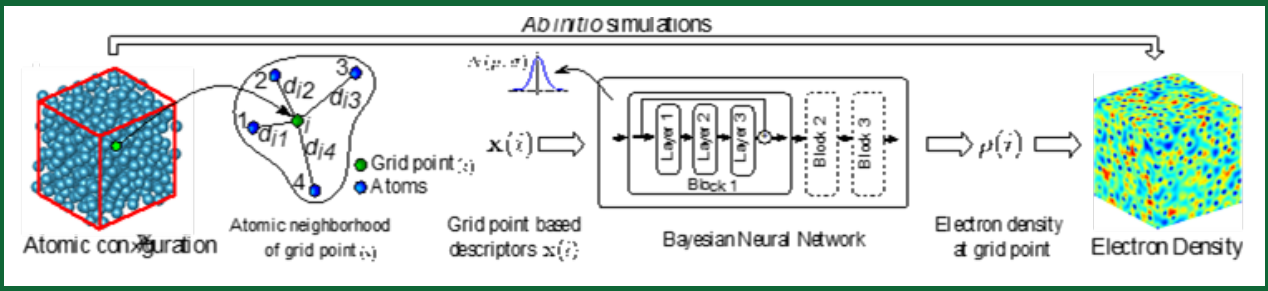
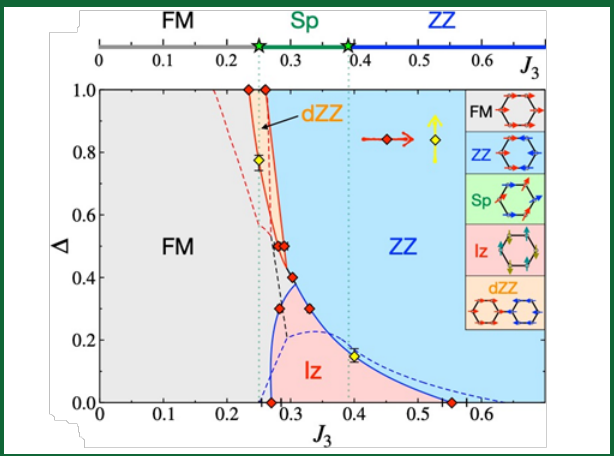
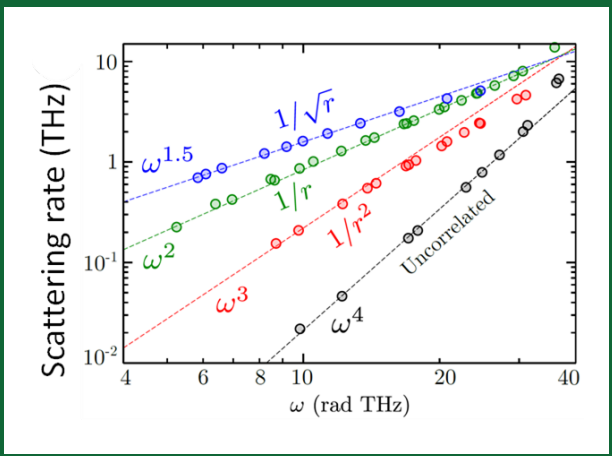
Theoretical Condensed Matter Physics Principal Investigators' Meeting

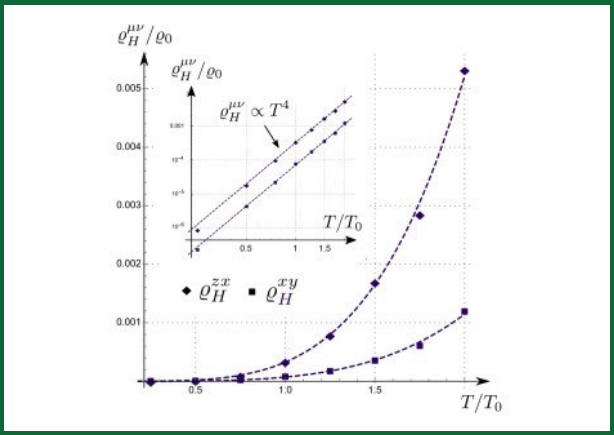
April 10-12, 2024

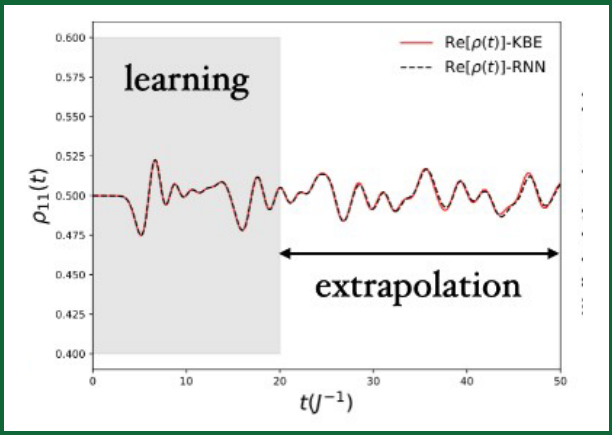
Program and Abstracts







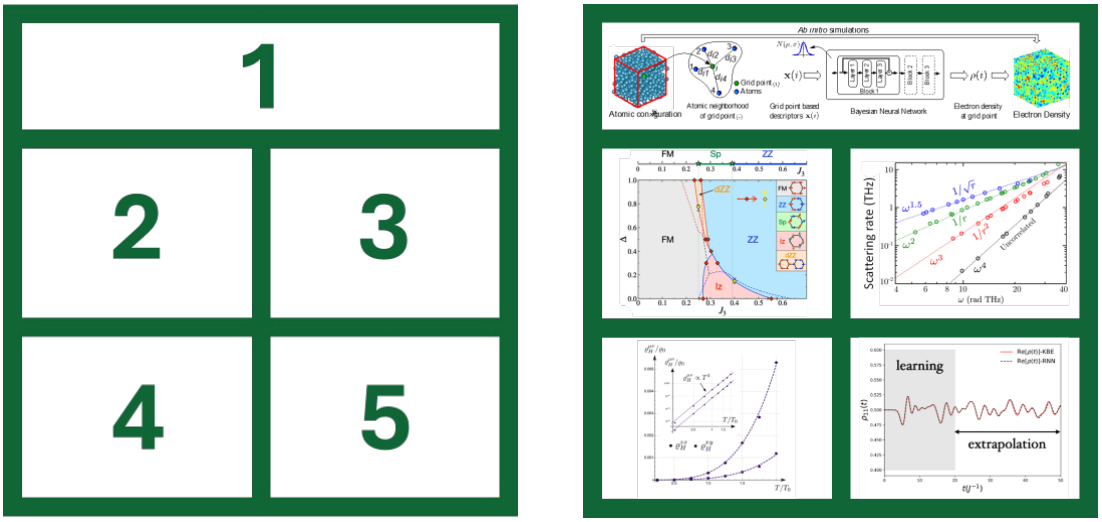




U.S. DEPARTMENT OF ENERGY | Office of Science

Office of Basic Energy Sciences
Materials Sciences and Engineering Division

On the Cover



1. PI Amartya Banerjee:Prediction and Tuning of Spin Selectivity Properties of Chiral Nanomaterials via an Integrated Machine Learning -- First Principles Approach, shown is a Bayesian Neural Network based ML model for electron density prediction.
2. PI Alexander Chernyshev: Quantum Dynamics in Anisotropic Magnets, shown are (a) classical and (b) quantum phase diagrams of the J_1–J_3 honeycomb-lattice ferro-antiferromagnetic model. Novel double-zigzag and Ising-z phases supersede classical spiral phase.
3. PI Valentino Cooper: Simulation, Design, and Discovery of Complex Materials, shown is correlated disorder causing breakdown of Rayleigh's ω^4 law. Results are extracted from large-scale simulations including those with no correlations and 1/r correlations (optimal).
4. PI Leon Balents: Theory of fluctuating and critical quantum matter, shown is the thermal Hall resistivity from many body phonon skew scattering off of antiferromagnetic magnons of a layered 2d magnet. The results show a T^4 power law and effects both for heat currents within and perpendicular to the 2d layers, a hallmark of phonon heat transport.
5. PI Vojtech Vlcek: Real-time dynamics of driven correlated electrons in quantum materials, shown is the Recurrent Neural Network approach extrapolating the Green's function in time, which leads to orders of magnitude computational savings without loss of accuracy.

Forward

This book contains extended abstracts of presentations made at the 2024 Theoretical Condensed Matter Physics (TCMP) Principal Investigators' Meeting sponsored by the Materials Sciences and Engineering Division of the U.S. Department of Energy (DOE), Office of Basic Energy Sciences (BES), which was held April 10-12, 2024. This was the first in-person meeting since the COVID-19 pandemic and convened scientists whose awards are managed within the TCMP program by the BES to present the most exciting, new research accomplishments and proposed future scientific directions in their supported research awards.

The purpose of the Principal Investigators (PIs) meeting is to bring together the PIs in the TCMP program to share the latest exciting scientific knowledge and discoveries, facilitate exchange of ideas and promote collaborations. For the BES and the participating PIs, the meeting serves the purpose of providing an overview and assessment of the whole program, which helps BES to identify new research areas and chart future directions for the program. This meeting brings together leading experts in topical areas of research supported by the TCMP program and is designed to stimulate and inspire new ideas.

The mission of the BES program is to support fundamental research to understand, predict, and ultimately control matter and energy at the electronic, atomic, and molecular levels. More specifically, the TCMP program supports fundamental research in quantum physics with an emphasis on quantum materials, materials discovery and design, out-of-equilibrium quantum dynamics, and materials theory related to DOE missions. Major scientific themes include electron correlations, quantum phases of matter, topological states, quantum magnetism, superconductivity, multiferroic and ferroelectric materials, and excited states phenomena. Research spans from analytical to computational approaches with a strong emphasis on theory, methods, and technique development, as well as prediction and interpretation of novel quantum phenomena.

The TCMP program supports primarily single-PI university projects, small and large team projects at national laboratories, as well as interdisciplinary, multi-investigator research in response to special topic funding opportunity announcements. For example, small and large team Computational Materials Science (CMS) projects support the Materials Genome Initiative (MGI) and the exascale computing initiative by developing widely applicable open-source software, utilizing the range of midscale computing all the way to DOE's exascale computing facilities. They provide software platforms and data necessary for the design of new functional materials with a broad range of potential applications, including alternative and renewable energy, microelectronics, low-power and opto-electronics, data storage, materials processing, and functional materials for quantum information science.

The computational ecosystem is complemented by next-generation, high-performance computing projects supported through the Scientific Discovery Through Advanced Computing (SciDAC) partnership with the Office of Advanced Scientific Computing Research (ASCR). SciDAC supports interdisciplinary teams, which bring together experts in key areas of science and energy research, applied mathematics, and computer science to address computing challenges and take maximum advantage of DOE's supercomputers, allowing them to accelerate the pace of

scientific discovery.

The meeting was held in a hybrid format, with in-person oral presentations and webcasting. The format afforded eight oral and four poster sessions covering the range of activities. Short lightning talks were presented by new awards recently added to the TCMP program. Two panel discussions focused on “Exciting Directions in TCMP” and “Artificial Intelligence & Machine Learning for TCMP”. The meeting was attended by over 100 scientists, organized in 44 regular talks, 7 lightning talks, and 89 poster presentations.

We appreciate the contributions of the co-chairs for the scientific program, Dr. Aditi Mitra (New York Univ.), Dr. Brian Moritz (SLAC), and Dr. Qimiao Si (Rice Univ.), and the PIs for sharing their exciting ideas and latest findings. We are also grateful for the excellent support provided by Ms. Tia Moua and Mr. Bruce Warford of the Oak Ridge Institute for Science and Education (ORISE) and by Ms. Teresa Crockett of BES, for their efforts in organizing the meeting.

Drs. Matthias Graf and Claudia Mewes

Program Managers
Theoretical Condensed Matter Physics
Division of Materials Sciences and Engineering
Office of Basic Energy Sciences
Office of Science
U.S. Department of Energy

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2024 DOE-BES Theoretical Condensed Matter Physics Principal Investigators' Meeting

Meeting Chairs: Matthias Graf and Claudia Mewes (DOE-BES) Meeting Co-Chairs: Aditi Mitra (New York University), Brian Moritz (SLAC), and Qimiao Si (Rice University)

Wednesday, April 10, 2024

7:30-8:15 AM	**Breakfast**
8:15-8:30 AM	**Matthias Graf and Claudia Mewes**, TCMP Program Managers, DOE-BES *Welcome and Introductory Remarks*
Oral Session 1	Session Chair: **Brian Moritz**, SLAC
8:30-8:45 AM	**Garnet Chan**, California Institute of Technology *Quantitative descriptions of cuprate superconductors from ab initio quantum many-body theory*
8:45-9:00 AM	**Daniel Agterberg**, University of Wisconsin - Milwaukee *Crystal-structure driven odd-parity superconductivity*
9:00-9:15 AM	**Elbio Dagotto**, Oak Ridge National Laboratory *High-Tc resurrection: $La_3Ni_2O_7$ with Tc≈80Ks*
9:15-9:30 AM	**Steven Johnston**, University of Tennessee - Knoxville *Origin of charge order in the bismuate superconductors*
9:30-9:45 AM	**Dong-Ning (Donna) Sheng**, California State University - Northridge *Unconventional Superconductivity in the Doped Triangular Lattice Moiré System*
9:45-10:00 AM	**Richard Scalettar**, University of California - Davis *Photoinduced enhancement of superconductivity*
10:00-10:30 AM	**Break**
Oral Session 2	Session Chair: **Fernando Reboredo,** Oak Ridge National Laboratory
10:30-10:45 AM	**Andrey Chubukov**, University of Minnesota *Pairing by nematic fluctuations in doped FeSe*
10:45-11:00 AM	**Mark van Schilfgaarde**, National Renewable Energy Laboratory *Vertex controls superconductivity in FeSe*
11:00-11:15 AM	**Igor Zutic**, University at Buffalo *THz Spin-Light Coupling in Proximitized Dirac Materials*
11:15-11:30 AM	**Michael Hermele**, University of Colorado - Boulder *Fracton self-statistics*
11:30-11:45 AM	**Paul Kent**, Oak Ridge National Laboratory *Gapless topological surface states are explained by stacking-fault disorder*
11:45 AM-12:00 PM	**Rahul Nandkishore**, University of Colorado – Boulder *Multipole groups and new fracton phenomena on arbitrary crystalline lattices*
12:00–12:15 PM	**Irena Knezevic**, University of Wisconsin - Madison *Plasmon-enhanced optical nonlinearity in graphene nanomeshes*
12:15-12:30 PM	**Feliciano Giustino**, The University of Texas at Austin *How polarons form in two-dimensional crystals*
12:30-1:30 PM	**Lunch and Concurrent Presentation**

1:00-1:30 PM	**TCMP Info & Updates**, TCMP Program Managers
Oral Session 3	Session Chair: Sahar **Sharifzadeh**, Boston University
1:30-1:45 PM	**Giulia Galli**, Argonne National Laboratory *Large scale calculations of excited state energy surfaces*
1:45-2:00 PM	**Michael Zaletel**, University of California - Berkeley *Benchmarking quantum computations with 100+ qubits*
2:00-2:06 PM	**Ezio Iacocca**, University of Colorado - Colorado Springs *Lightning Talk: Multiscale model for magnetization dynamics*
2:06-2:12 PM	**Arunima Singh**, Arizona State University *Lightning Talk: 2D Materials Heterostructures for Solar Energy Conversion*
2:12-2:18 PM	**Chinedu Ekuma**, Lehigh University *Lightning Talk: Advanced computing unveils exotic physics in quantum materials*
2:18-2:24 PM	**Herbert Fotso**, University at Buffalo *Lightning Talk: Nonequilibrium Dynamics of Disordered Correlated Systems*
2:24-2:30 PM	**Renat Sabirianov**, University of Nebraska - Omaha *Lightning Talk: Novel narrow-gap magnetic semiconductors: $YCrB_4$ and $YMnB_4$*
2:30-3:00 PM	**Break**
3:00-5:00 PM	**Poster Session 1**
5:00-6:00 PM	**Dinner and Concurrent Panel Discussion**
5:30-6:30 PM	**Exciting Directions in TCMP** Moderator: Mike Zaletel (University of California – Berkeley / Lawrence Berkeley National Laboratory) Panelists: **Leon Balents** (University of California – Santa Barbara), **Giulia Galli** (Argonne National Laboratory / University of Chicago), **Sinéad Griffin** (Lawrence Berkeley National Laboratory), **Felipe Jornada** (Stanford University), **Mark van Schilfgaarde** (National Renewable Energy Laboratory)
6:30-8:00 PM	**Poster Session 2**

Thursday, April 11, 2024

7:30-8:15 AM	**Breakfast**
Oral Session 4	Session Chair: **Ivar Martin**, Argonne National Laboratory
8:15-8:30 AM	**Long-Qing Chen**, The Pennsylvania State University *Thermodynamics and Phase-field Simulations of Light-induced Transformations of Mesoscale Polar Structures*
8:30-8:45 AM	**Wang-Kong Tse**, University of Alabama - Tuscaloosa *Photo-induced Magnetic Interaction Predicted*
8:45-9:00 AM	**Benedetta Flebus**, Boston College *New many-body quantum platform revealed*
9:00-9:15 AM	**Alexander Chernyshev**, University of California - Irvine *Quantum phases in a honeycomb-lattice model*
9:15-9:30 AM	**Carsten Ullrich**, University of Missouri *New first-principles tools for noncollinear magnetism*
9:30-9:45 AM	**Rebecca Flint**, Ames National Laboratory *Enhancing anisotropy with valence fluctuations*
9:45-9:52 AM	**Marisol Alcántara Ortigoza,** Tuskegee University *RENEW Lightning Talk: New optically controllable spin qubit revealed*
9:52-10:00 AM	**Armin Rahmani**, Western Washington University *FAIR Lightning Talk: Supersymmetry on a quantum computer*
10:00-10:30 AM	**Break**
Oral Session 5	Session Chair: **David Broido**, Boston College
10:30-10:45 AM	**Inna Ponomareva**, University of South Florida *Persistent and Quasipersistent Spin Textures in Halide Perovskites Induced by Uniaxial Stress*
10:45-11:00 AM	**Valentino Cooper**, Oak Ridge National Laboratory *Breaking Rayleigh's Law Suppresses Thermal Conductivity*
11:00-11:15 AM	**Tadashi Ogitsu**, Lawrence Livermore National Laboratory *Phonon Driven Ballistic Photocurrents*
11:15-11:30 AM	**Lucas Lindsay**, Oak Ridge National Laboratory *Translational symmetries constrain how phonons interact in layered magnets*
11:30-11:45 AM	**Chris Marianetti**, Columbia University *Precisely computing phonons via irreducible derivatives*
11:45 AM-12:00 PM	**Victor Galitski**, University of Maryland *Surprises in Non-Equilibrium Materials: Slow Thermalization*
12:00-1:00 PM	**Lunch and Concurrent Presentation**
12:30-1:00 PM	**BES Updates & more TCMP Info,** TCMP Program Managers

Oral Session 6	Session Chair: **Paul Kent**, Oak Ridge National Laboratory
1:00-1:15 PM	**Massimiliano Di Ventra,** University of California - San Diego *Transformer quantum state: A multipurpose model for quantum many-body problems*
1:15-1:30 PM	**Massimiliano Di Ventra**, University of California - San Diego *Transformer quantum state: A multipurpose model for quantum many-body problems*
1:30-1:45 PM	**Gia-Wei Chern**, University of Virginia *Machine Learning for spin dynamics of metallic magnets*
1:45-2:00 PM	**Amartya Banerjee**, University of California - Los Angeles *Making machine learning based electronic structure predictions systematic, reliable and efficient*
2:00-2:15 PM	**Sugata Chowdhury**, Howard University *Capturing dynamical spin correlations with machine learning*
2:15-2:30 PM	**Qimin Yan**, Northeastern University *Contrastive learning for density functional design*
2:30-3:00 PM	**Break**
3:00-5:00 PM	**Poster Session 3**
5:00-6:00 PM	**Dinner and Concurrent Panel Discussion**
5:30-6:30 PM	**Artificial Intelligence & Machine Learning for TCMP** Moderator: **Hal Finkel** (Acting Division Director, Computational Science Research and Partnership, DOE ASCR) Panelists: **Chinedu Ekuma** (Lehigh University), **Susanta Gosh** (Michigan Technological University), **Jason Munro** (Lawrence Berkeley National Laboratory), **Vojtech Vleck** (University of California – Santa Barbara)
6:30-8:00 PM	**Poster Session 4**

Friday, April 12, 2024

7:30-8:15 AM	**Breakfast**
Oral Session 7	Session Chair: **Mitra Aditi**, New York University
8:15-8:30 AM	**Sahar Sharifzadeh**, Boston University *Revealing the Nature of Optical Transitions within a Doped Carbon Nanotube*
8:30-8:45 AM	**James Freericks**, Georgetown University *Avoiding the heat death of driven electrons*
8:45-9:00 AM	**Steven Louie**, Lawrence Berkeley National Lab *Self-driven exciton-Floquet phenomena*
9:00-9:15 AM	**Ziqiang Wang**, Boston College *Entanglement Probe and Control in Material*
9:15-9:30 AM	**Diana Qiu**, Yale University *Mixing Bulk and Topological Surface States with Chiral Excitons in Bi_2Se_3*
9:30-10:00 AM	**Break**
Oral Session 8	Session Chair: **Qimiao Si,** Rice University
10:00-10:15 AM	**Natalia Perkins**, University of Minnesota *Fractionalized Excitations Probed by Ultrasound*
10:15-10:30 AM	**Patrick Lee**, Massachusetts Institute of Technology *Emergent particle and gauge field in a spin liquid*
10:30-10:45 AM	**Romain Vasseur**, University of Massachusetts - Amherst *Fluctuations in many-body quantum systems*
10:45-11:00 AM	**Senthil Todadri**, Massachusetts Institute of Technology *Non-fermi liquids from kinetic constraints*
11:00-11:15 AM	**Aldo Romero**, West Virginia University *Site and orbital selective Mott transition from DMFT*
11:15-11:30 AM	**Matthias Graf and Claudia Mewes**, TCMP Program Managers, DOE-BES Concluding Remarks

Poster Session 1

Wednesday, April 10, 2024, 3:00-5:00 PM

Last Name	First Name	Institution	**Project Title** (Poster Title not available at time of printing)
Agterberg	Daniel	UW-Milwaukee	Superconductivity in quantum materials
Balents	Leon	UCSB	Theory of fluctuating and critical quantum matter
Chan	Garnet	Caltech	Ab initio complete cell quantum embedding and diagrammatic coupled cluster for correlated materials phase diagrams
Chubukov	Andrey	U of M	Superconductivity and Competing Orders in Correlated Electron Systems
Dagotto	Elbio	ORNL	Theoretical Studies of Complex Collective Phenomena
Devereaux	Thomas	SLAC	Theory Institute for Materials and Energy Spectroscopies (TIMES)
Feiguin	Adrian	Northeastern	Time-Dependent Phenomena in Correlated Materials
Giustino	Feliciano	UT at Austin	Toward exascale computing of electron-phonon couplings for finite-temperature materials design
Hermele	Michael	CU-Boulder	Symmetry in Correlated Quantum Matter
Johnston	Steven	UT-Knoxville	AI and data science enabled predictive modeling of collective phenomena in strongly correlated quantum materials
Kent	Paul	ORNL	Center for Predictive Simulation of Functional Materials
Knezevic	Irena	UW-Madison	Wigner Equation with Electromagnetic Fields for Electron Transport in Quantum Materials
Liu	Zi-Kui	Penn State	Zentropy Theory for Transformative Functionalities of Magnetic and Superconducting Materials
Louie	Steven	LBNL	Theory of Materials
Martin	Ivar	ANL	Correlations and Dynamics in Quantum Materials
Nandkishore	Rahul	CU-Boulder	Fractons and Beyond

Narayanan	Badri	U of L	Controlling reversible phase transitions in rare-earth nickelates for novel memory devices
Prokofiev	Nicolay	U Mass-Amherst	Diagrammatic Monte Carlo Approach to Real-frequency response Functions and the Spin-Fermion Model of Hot Spots
Rahman	Talat	UCSF	Theoretical and Computations studies of Excitations in Functional Nanomaterials
Sheng	Donna	CSUN	Global Quantum Phase Diagram and Topological Superconductivity in Strongly Interacting Systems
Si	Qimiao	Rice	Electron Correlations, Bad-Metal Behavior and Unconventional Superconductivity
Tsvelik	Alexei	BNL	Condensed Matter Theory
van Schilfgaarde	Marc	NREL	Ab initio theory of unconventional superconductivity
Wan	Liwen	LLNL	Atomic to Mesoscale Models of Phase Transitions for Energy Materials (lead PI: Adelstein)
Zhu	Qiang	UNC Charlotte	Data-driven Discovery of Inorganic Electrides for Energy Applications
Zutic	Igor	UB	Quantum Materials: Magnetism, Spin-Orbit Coupling, and Superconductivity

Poster Session 2

Wednesday, April 10, 2024, 6:30-8:00 PM

Last Name	First Name	Institution	**Project Title** (Poster Title not available at time of printing)
Bernevig	Andrei	Princeton	Moire and Heavy Fermions: A New Strongly Interacting Paradigm
Broido	David	Boston College	Coupled electron-phonon transport from first principles
Ekuma	Chinedu	Lehigh University	Custom Design of van der Waals Materials
Fernandes	Rafael	U of M	Intertwined and Vestigial Electronic Orders in Correlated Systems
Fotso	Herbert F	UB	Nonequilibrium Dynamics of Non-Ideal Quantum Materials
Freericks	James	Georgetown	Theory for pump/probe experiments in charge-densitywave materials
Galli	Giulia	ANL	Midwest Integrated Center for Computational Materials (MICCoM)
Iacocca	Ezio	UC-Colorado Springs	Fourier and fractional neural operators to unveil topological textures in 3D magnetism
Jain	Jainendra	Penn State	Quantitative Studies of the Fractional Quantum Hall Effect
Lee	Patrick	MIT	Strongly correlated electronic systems: Local Moments and Conduction Electrons
Mulligan	Michael	UC Riverside	Duality and Strongly Interacting Systems
Perkins	Natalia	U of M	Identification of Fractionalized Excitations in Quantum Spin Liquids and Related Materials
Sabirianov	Renat	UN-Omaha	A High-Throughput Computational and Experimental Approach to the Design of Unconventional Magnets
Singh	Arunima	ASU	Functionalization of 2D Materials Heterostructures for Solar Energy Conversion
Todadri	Senthil	MIT	Unconventional Metals in Strongly Correlated Systems
Vasseur	Romain	U Mass-Amherst	Universality and Complexity of Non-equilibrium Quantum Dynamics
Wang	Ziqian	Boston College	Disorder and Interaction in Correlated Electron Systems
Zaletel	Michael	UC Berkeley	Quantum Simulation and State Preparation for TwoDimensional Materials

Poster Session 3

Thursday, April 11, 2024, 3:00-5:00 PM

Last Name	First Name	Institution	**Project Title** (Poster Title not available at time of printing)
Alcántara Ortigoza	Marisol	Tuskegee U	HBCU Undergraduate Program toward ab initio Prediction of SinglePhoton-Emitters and Spin Qubits in Defected 2D Semiconductors
Chen	Long-Qing	Penn State	COMMS: Center for Computational Mesoscale Materials Science
Chernyshev	Alexander	UC-Irvine	Quantum Dynamics in Anisotropic Magnets
Claassen	Martin	U Penn	Harnessing Quantum Geometry of Correlated Electrons for Next-Generation Photovoltaics
Coleman	Piers	Rutgers	Spin and Orbital Physics in Novel Correlated Materials
Cooper	Valentino	ORNL	Simulation, Design, and Discovery of Complex Materials
Flebus	Benedetta	Boston College	Exploring the quantum potential of magnetic systems
Flint	Rebecca	Ames	Exploiting the interplay of mixed valence and magnetic anisotropy in rare earths
Galitski	Victor	UMD	Theory of Fluctuations in Strongly-Correlated Materials
Ke	Liqin	Ames	Quantum Control and Tuning of Magnetic 2D van der Waals Heterostructures

Kovalev	Alexey	UN-Lincoln	Spin Currents in Magnetic Systems and Heterostructures
Lambrecht	Walter	Case Western	Excited state electronic structure and phase stability in 2D materials
Lindsay	Lucas	ORNL	Elucidating the Nature of Chiral and Topological Phonons in Materials for Energy Technologies
Marianetti	Chris	Columbia	A new approach to the interacting phonon problem
Mele	Eugene	U Penn	Structure and Electronic Properties of Dirac Material
Mitra	Aditi	NYU	Nonequilibrium Phenomena in Topological Insulators
Munro	Jason	LBNL	Materials Project (lead PI Persson)
Ogitsu	Tadashi	LLNL	Center for Non-Perturbative studies of functional materials under Non-EQuilibrium conditions (NPNEQ)
Ponomareva	Inna	USF	Co-Functional (Anti)ferroic Perovskites with Topological Phases: Statics and Dynamics at Finite Temperatures
Rahmani	Armin	WWU	Emergent phases, transport, and nonequilibrium dynamics of interacting Majorana fermions
Reboredo	Fernando	ORNL	Extending the Reach of Computational-Theoretical Methods to Materials at the Energy Frontier

Tse	Wang-Kong	UA-Tuscaloosa	Interaction and Transport Effects in Driven Magnetic and Topological Materials
Ullrich	Carsten	MU	Time-dependent density-functional approaches for noncollinear spin dynamics
Wu	Ruqian	UC-Irvine	First Principle Investigation for Magnetic Properties of Innovative Materials
Zang	Jiadong	UNH	Topological Spin Textures in Chiral Magnets: From 2D to 3D
Zunger	Alex	CU-Boulder	Understanding and Control of Charge Carriers in Quantum Materials

Poster Session 4

Thursday, April 11, 2024 6:30-8:00 PM

Last Name	First Name	Institution	**Project Title** (Poster Title not available at time of printing)
Banerjee	Amartya	UCLA	Prediction and Tuning of Spin Selectivity Properties of Chiral Nanomaterials via an Integrated ML & First Principles Approach
Barraza-Lopez	Salvador	UA	Toward optical quantum entanglement on a 2D ferroelectric/ferroelastic platform
Chern	Gia-Wei	UVA	Machine learning aided multi-scale dynamical modeling of functional electronic materials
Chowdhury	Sugata	Howard	Machine Assisted Quantum Magnetism
Di Ventra	Massimiliano	UCSD	Memcomputing the Spectrum of Correlated Quantum Systems
Hosur	Pavan	UH	Nontrivial consequences of non-centrosymmetry in topological and trivial metals
Khemani	Vedika	Stanford	The Non-Equilibrium Quantum Frontier
Louie	Steven	LBNL	Center for Computational Study of Excited-State Phenomena in Energy Materials (C2SEPEM)
Raghu	Srinivas	Stanford	Atomic Engineering Oxide Heterostructures: Materials by Design

Romero	Aldo	WVU	Applications of Nickelate perovskites for neuromorphic computing from electronic structure and ML
Santos	Luiz	Emory	Catalysing Chiral Quantum Matter at Hofstadter Van Hove Singularities
Sharifzadeh	Sahar	Boston U	Theoretical understanding of the excitonic properties of low-dimensional materials
Tserkovnyak	Yaroslav	UCLA	Nonequilibrium thermodynamics in magnetic nanostructures
Vlcek	Vojtech	UCSB	Real-time dynamics of driven correlated electrons in quantum materials
Wagner	Lucas	UIUC	QMC-HAMM: High accuracy multiscale models using quantum Monte Carlo
Wong	Bryan	UC-Riverside	DECODE: Data-driven Exascale Control of Optically Driven Excitations in Chemical and Material Systems
Woods	Lilia	USF	Dispersive Interactions in Quantum Materials: Interplay between Anisotropy, Doping, and Non-linearity
Yan	Qimin	Northeastern	Synthesis of motif and symmetry for accelerated learning, discovery, and design of electronic structures for energy conversion applications

Laboratory Abstracts

Simulation, Design, and Discovery of Complex Materials

V. R. Cooper, T. Berlijn, R. S. Fishman, L. R. Lindsay, D. S. Parker (Oak Ridge National Laboratory)

Keywords: high entropy, phonons, weak interactions, stability

Research Scope

The overarching goal of this FWP is to co-design structural stability and function to engender and control emergent properties in complex materials. We address this goal through three specific aims: (1) Designing defect- and disorder-derived thermal transport, (2) Unraveling the influence of doping and disorder on materials stability and properties, and (3) Examining the robustness of van der Waals density functionals. During the past two years, key achievements included: (i) the development of approaches to predict and understand lattice thermal properties due to mass disorder and (ii) the development of tools to explore stability and emergent properties in multicomponent oxides. Fundamentally, this work continues to build towards a framework for codesigning stability and function (, magnetism, polarization, and phonon transport) in layered heterostructures and materials with disorder and defects.

Recent Progress

Ultralow thermal conductivities in lead-free halide perovskites. To develop insights into the phonon transport behaviors of two prototypical inorganic lead-free halide double perovskites (Cs_2SnI_6 and γ-$CsSnI_3$) of potential interest for optoelectronic and thermoelectric applications, we employed state-of-the-art density functional theory (DFT) calculations [P10]. Their thermal conductivity values are ultralow (0.26 W/m/K and 0.72 W/m/K, respectively, at room temperature) and cannot be understood in terms of standard DFT Boltzmann transport theory [1] as temperature-induced anharmonic dynamical effects are critically important. This behavior is dictated by SnI_6 octahedral rotations and Cs rattling. Figure 1 depicts the temperature-dependent thermal conductivities of Cs_2SnI_6 at T=300K. Here, we demonstrated that phonon renormalization (accounting for temperature) [2] and wave-like coherent contributions [3], in addition to particle-like phonon contributions, are critical to understanding the full nature of transport in these materials.

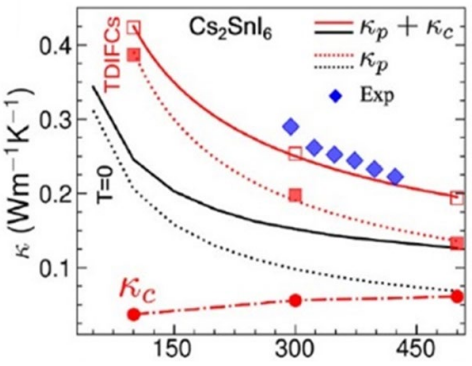

Figure 1 Calculated thermal conductivities (k) of Cs_2SnI_6 comparing phonon (k_p) and coherent (k_c) contributions to k with measured data and differing levels of DFT [P10].

Breaking Rayleigh's law for phonons. Finding ways to suppress the thermal conductivity of insulators and semiconductors is a grand challenge of condensed matter physics, with crucial relevance to energy applications such as thermal insulation, energy storage, and thermoelectrics. Since heat-carrying low-frequency acoustic phonons are insensitive to randomly distributed point defects due to Rayleigh's law, researchers have for decades tried to scatter them using nanostructures and extended defects. In our recent work, we used large-scale nonperturbative Green's function techniques involving tens of millions of atoms to show that spatially correlated point defects can break Rayleigh's law (see Fig. 2) [P1]. Furthermore, we demonstrated that this leads to strong scattering of low-frequency phonons, resulting in an order of magnitude reduction in room temperature thermal conductivity. In addition, we analytically determined that to minimize thermal conductivity, correlations should decay as 1/r. Our work provides a unifying framework guiding theoreticians and experimentalists for fine tuning phonon transport in nanostructured materials. This effort lays the groundwork for future studies of experimentally realizable systems with correlated disorder such as high entropy oxides.

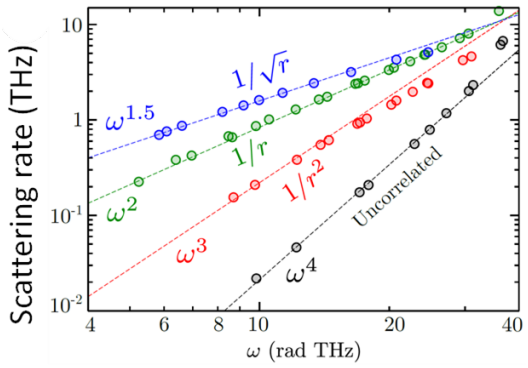

Figure 2 Correlated disorder causing breakdown of Rayleigh's ω^4 law. Results are extracted from large-scale simulations including those with no correlations and 1/r correlations (optimal) [P1].

Computationally accelerated discovery of high entropy ceramics. Building on our prior work demonstrating the ability to predict stable, single-component high entropy oxides (HEOs) [4], we employed a combination of first-principles and Metropolis Monte Carlo simulations to design a new class of high entropy pyrochlore oxides [P9]. Due to the range of stoichiometries of competing compounds, we now incorporate oxygen vacancy formation energies that enable access to synthesis-like conditions as a function of oxygen partial pressure (Fig. 3). Most significantly, these calculations guided the experimental synthesis of four new phase pure and one 97.4% pure multicomponent entropy stabilized pyrochlore oxides. A key advantage of this approach is the simultaneous evaluation of potential impurity phases, ionic disorder, and oxygen vacancy concentrations for realistic predictions of material synthesizability. Extending our success in predicting phase stability as a function of oxygen pressures and temperatures, we revisited our previous predictions of Fe-based rocksalt high entropy oxides to demonstrate that stable Fe- and Mn-based HEOs can exist due to the stabilization of Fe and Mn in the *A*O structure under oxygen poor conditions. These predictions explained the recent realization of an Fe- and Mn-containing HEO, previously thought unattainable

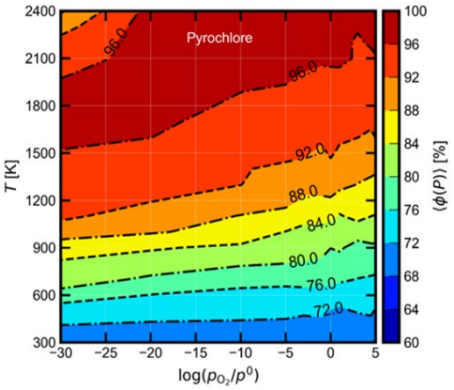

Figure 3 Heat map of pyrochlore phase fraction, ϕ, for a high entropy oxide as a function of temperature and oxygen partial pressure p_{O_2}

[P3]. Our results provide fresh insights into the design and property tailoring of emerging classes of disordered ceramics.

Future Plans

Future research plans seek to extend our efforts using large-scale nonperturbative numerical simulations of disordered alloys to the realization of breaking Rayleigh's Law in realistic systems. Furthermore, we will combine this work with our research on designing and discovering high entropy oxides with unique functional properties. Here, we will employ first-principles calculations to find experimentally realizable high entropy oxides with correlated disorder. The goal will be to understand the behavior of phonons in these systems as they are quenched just above their structural transition temperatures. Simultaneously, we will develop approaches to predict the vibrational entropy contributions to the phase diagrams of multicomponent oxides. Here, we will emphasize HEOs with unequal dopant contributions such as Li-doped pyrochlore oxides (which are being explored in the experimental BES programs at ORNL as potential cathode materials). While the vibrational entropy component can be simply derived from an integration over the phonon density of states, the key challenge remains in computing the phonon density of states in multicomponent disordered materials, where differences between average views and specific atomic configurations can result in large variations in computed vibrational entropies. Here, we hypothesize that since each cation in an oxide is surrounded by an oxygen cage, the long-range interactions will be screened and may lead to a more efficient approach to studying phonons in these materials. This work complements the aforementioned materials specific evaluation of the effects of force and mass disorder on phonons. Together, these efforts will allow us to build a clearer picture of phonon behaviors and vibrational entropies in strongly disordered materials.

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Theoretical Studies of Collective Phenomena

Lead PI: Elbio Dagotto [Oak Ridge National Laboratory (ORNL) and University of Tennessee at Knoxville (UTK)]. Co-PIs: Thomas Maier (ORNL), Adriana Moreo (ORNL/UTK), and Satoshi Okamoto (ORNL)

Keywords: Quantum materials, computational, multi-orbital Hamiltonians

Research Scope

Quantum materials display a remarkable variety of exotic fermionic states. By studying appropriate Hamiltonians, theorists can provide conceptual frameworks to understand experimental observations and predict new properties. The *overarching goal* of our project is to gain a quantitative understanding of the many-body states generated in models for quantum materials with simultaneously active spin, charge, and orbital degrees of freedom, and electronic Hubbard repulsion, Hund, and spin-orbit couplings. This research will be performed via state-of-the-art many-body computational techniques in combination with analytical methodologies. This project aligns with the mission of the Department of Energy to guide the development of future practical technologies through fundamental science.

Recent Progress

Our FWP at ORNL produced 64 publications during FY21-23, including several in high impact factor journals. During FY24, we already published 8 additional manuscripts. Below we focus only on a subset of five publications representative of the many topics covered by our team.

(1) *Ni-based High-Tc Superconductivity*. In the summer of 2023, surprising results were announced. The compound $La_3Ni_2O_7$ (LNO) was reported to be superconducting at temperatures as high as 80 K, albeit with the need of a high pressure of 14 GPa. This triggered a considerable effort in the international scientific community. We have contributed to this effort with several publications, but here for simplicity we will only focus on Refs. [1,2]. Figure 1 contains the primary ingredients of our results, which arise from a combination of density functional theory (DFT), with or without Hubbard U, and the random phase approximation (RPA) applied to a two-orbital Hubbard-Hund model that we constructed, with hoppings and crystal field from DFT.

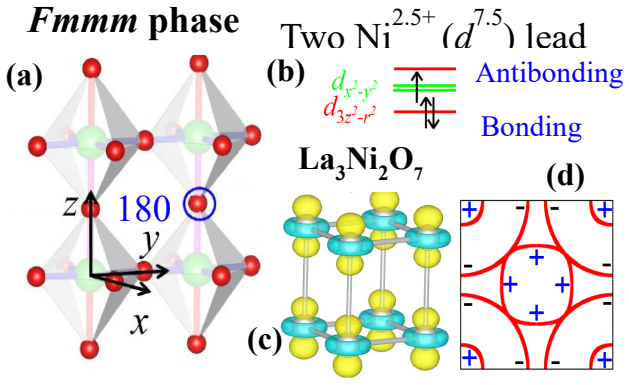

Figure 1: (a) *Fmmm* atomic phase that becomes superconducting at high pressure. **(b)** "Dimers" that form between adjacent Ni ions in bilayers, with one Ni in each plane. They form bonding-antibonding states, as shown. **(c)** Dominant orbitals $d_{3z2\text{-}r2}$ in LNO. **(d)** In red Fermi Surface of LNO in the *Fmmm* state. Shown are also the sign distribution of the order parameter in the superconducting $s\pm$ state

(2) *Altermagnets.* There is a novel state of magnetism mixture of the canonical ferromagnetic and antiferromagnetic orders, dubbed altermagnetism. Each species of spin, up or down, is surrounded by a different atomic environment that differs by a π/2 rotation, as in Fig. 2(a), and thus in an antiferromagnetic *metallic* state their Fermi Surfaces (for up and down spin) are rotated by π/2. Our efforts thus far have focused on a minimal two-dimensional single-orbital model, studied by Hartree Fock (HF) and the RPA [3]. Figure 2 illustrates work in progress addressing a multiorbital system for the t_{2g} sector of RuO_2, one of the material candidates to display this physics. As in the previous case [3], this model will be studied with HF, RPA, and also the Dynamical Cluster Approximation (DCA). We aim to predict quantities of value for inelastic neutron scattering experiments at ORNL, such as S(**q**,ω).

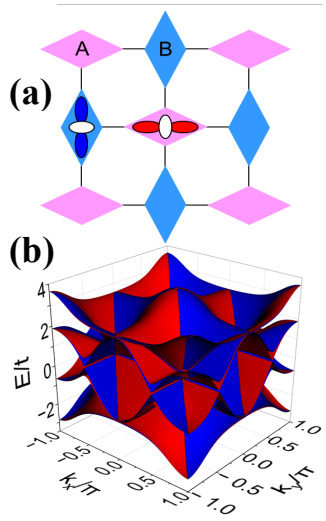

Figure 2: (a) Schematic of the e_g-orbital model for altermagnets, under study. Sublattices A and B have different Jahn-Teller (JT) type crystal fields, which lowers the $3x^2$-r^2 level (red) relative to the y^2-z^2 (not shown) on sublattice A, and lowers the $3y^2$-r^2 level (blue) relative to z^2-x^2 (not shown) on sublattice B. With AFM staggered ordering, spin-up and -down on different JT sublattices, altermagnetism is achieved. **(b)** Electronic band structure. Spin splitting is denoted by red (blue) for spin-up (down) bands.

(3) *Mechanism for insulating ferromagnetism.* We proposed a novel mechanism to stabilize FM insulating states in multiorbital systems [4]. FM insulators are rare, compared with AFM insulators which are common. A large *inter-orbital* nearest-neighbor hopping and a robust Hund coupling are needed in our mechanism. However, the stability of the phase depends on the interplay between the Hubbard repulsion U and the crystal-field splitting Δ_2 [see Fig. 3 with the phase diagram obtained using the density matrix renormalization group (DMRG)]. The FM insulating phase occurs when the Hubbard repulsion U is larger than the bandwidth W. For details see Ref. [4]. This mechanism explains the FM insulating state of quasi-1D $Ce_2O_2FeSe_2$. Thus, to synthesize materials with strong FM insulating

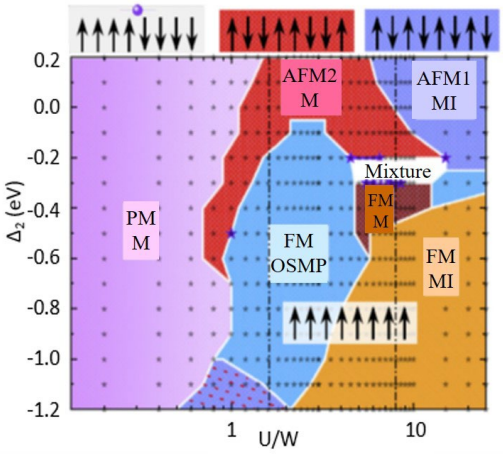

Figure 3: Phase diagram of a three-orbital Hubbard model [4] varying U/W and crystal-field splitting Δ_2 of the $d_{x^2-y^2}$ orbital, using DMRG. The Hund coupling is $J_H/U = 1/4$. Electronic and magnetic phases are indicated by solid regions and labels: paramagnetic metal (PM M, pink), ferromagnetic orbital-selective Mott phase (FM OSMP, light blue), ferromagnetic Mott insulator (FM MI, orange), antiferromagnetic Mott insulator (AFM1 MI, purple), ferromagnetic metal (FM M, maroon) and antiferromagnetic metal (AFM2 M, red). The bottom purple region is a mixture of tendencies. Data points are marked by small gray stars.

states, experimentalists should study compounds with large orbital entanglement and strong crystal-field splitting between half-filled and fully occupied orbitals.

(4) *Topological superconductivity (TSC) in the Rashba-Hubbard model.* We reported spin-singlet TSC in a 2D attractive Rashba-Hubbard model with Zeeman magnetic field, using DCA and DMRG [5]. When spin-orbit coupling is present, the spin-degenerate bands split into two pseudospin bands, and with an additional Zeeman field, the Dirac-like portion of the band structure acquires a gap. With the chemical potential tuned inside that gap, only a single Fermi surface for a given pseudospin remains, and the resulting metallic state is spinless. If superconductivity develops, this superconductivity is topological. In this case, the spin-singlet *s*-wave state in the original spin-basis has odd-parity *p*-wave triplet structure in the chiral basis.

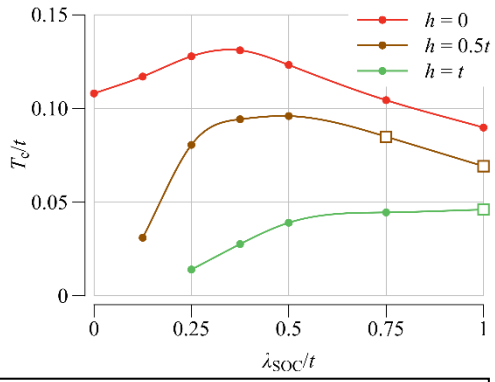

Figure 4: The superconducting transition temperature Tc/t in units of the hopping t, in the attractive Rashba-Hubbard model vs. Rashba SOC λ_{SOC}/t for different Zeeman magnetic fields h, calculated with DCA [73]. Data points with square symbols indicate parameters for which the superconducting state is found to be topologically non-trivial, based on results for the Fermi surface [5]. Results are shown for an electron filling $n = 0.25$ and the DCA calculations were performed for an 8-site cluster.

Future Plans

1. Within Ni-based superconductors, we will focus on "What causes pairing in LNO? Beyond RPA, in what channel is the pairing?" We will employ DCA in 2D, and DMRG in two-leg ladders to clarify these questions.

2. As described above, for altermagnets the next objective is to generalize our previous efforts from one- to multi-orbital models.

3. For TSC with a Rashba term, our immediate plan is to extend our previous results for the attractive Hubbard model to a repulsive case, both using DCA and DMRG on two-leg ladders.

4. We continue efforts in the field of Majoranas, now moving them in real time and focusing on fusion, non-adiabatic effects, and eventual braiding. The spinless Kitaev model will be used in chains, as well as in X and Y geometries, with various superconducting phases at the arms.

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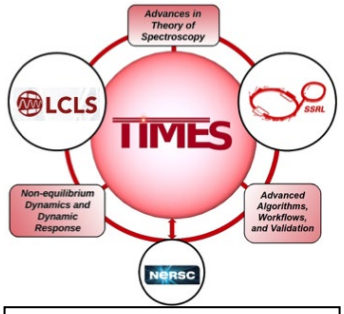
Theory Institute for Materials and Energy Spectroscopies (TIMES)

Thomas Devereaux, Brian Moritz, Hong-Chen Jiang (SLAC); John Rehr, Joshua Kas (Univ. of Washington)

Keywords: advanced theories, computational methods, x-ray spectroscopies, non-equilibrium

Research Scope

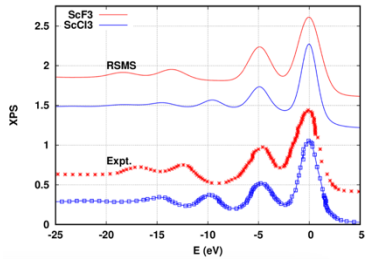
The Theory Institute for Materials and Energy Spectroscopies (TIMES) provides leadership in the development of advanced theories and computational methods for investigations using x-ray spectroscopies. These synergistic activities play a vital role in addressing cutting-edge problems in materials and energy sciences now being investigated at current and next-generation photon facilities, with a goal of moving from qualitative to predictive simulation. Our developments cover equilibrium, non-equilibrium and time-dependent phenomena, as well as advanced materials. While this focus is primarily on the capabilities of LCLS and SSRL at SLAC National Accelerator Laboratory, TIMES has a broader impact in the photon sciences community, providing a strong link between theory, computation and experiment that enhances overall scientific productivity.



Problems of scale enabled by linking efforts in spectroscopy and simulation in a national lab environment.

Recent Progress

Finite-temperature x-ray spectroscopy theory: We have continued our efforts to extend our real-space Green's function theory of x-ray spectroscopy to include non-equlibrium and finite temperature properties. These developments are particularly relevant for interpreting investigations of materials e.g., at the LCLS. To facilitate these calculations with our FEFF10 x-ray spectroscopy codes, we have recently developed an efficient approach for including self-energy effects at finite temperatures, up to the warm dense matter (WDM) regime [1].



Real-space multiple scattering calculation of the *2p* XPS of ScF_3 and $ScCl_3$ showing near quantitative agreement with experimental satellites.

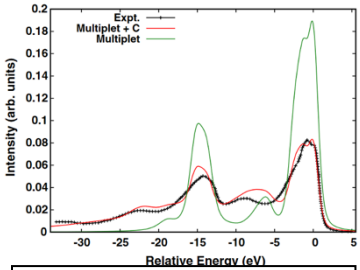
Real-space Green's function approach for Intrinsic Losses: Intrinsic inelastic losses in x-ray spectra characterize the features observed in x-ray photoemission spectra (XPS), as well as many-body effects such as satellites and edge-singularities in x-ray absorption spectra (XAS). We have developed an *ab initio* real-space Green's function (RSGF) approach to calculate these losses in terms of the dynamically screened core-hole interaction $W_c(\omega)$ and the independent particle response function [3]. Illustrative results show near quantitative agreement with other theoretical and experimental results in a variety of systems,

including the electron gas, sodium, and some early transition metal compounds. This generalized RSGF approach can also be applied to other problems, such as calculations of van der Waals interactions in condensed matter.

Theoretical and experimental XAS investigation of potassium-organic matter interactions: Potassium (K) is an essential nutrient for plant growth. However, the mechanisms underpinning microbial K uptake, transformation, storage, and sharing are poorly resolved. To better understand the controls on microbial K transformations, our colleagues at SSRL performed K K-edge x-ray absorption near-edge structure (XANES) spectroscopy on K-organic salts, which are frequently secreted by soil microbes. Theoretical XANES simulations based on TDDFT and BSE elucidate why the spectral shapes differ and indicate that the K K-edge spectra are associated with the entire ligand despite similar first-shell bonding environments around the K center. This study provides an important toolkit to understand how K is transformed by microbial processes.

Superconductivity models: Recent experiment has unveiled an anomalously strong electron-electron attraction in one-dimensional copper-oxide chain $Ba_{2-x}Sr_xCuO_{3+\delta}$. While the effect of the near-neighbor electron attraction V in the extended Hubbard chain has been examined recently, its effect in the Hubbard model beyond the 1D chain remains unclear. We report a DMRG study of the extended Hubbard model on long four-leg square cylinders [2]. We find that the attractive V can notably enhance the superconducting (SC) correlations while suppressing the charge-density-wave correlations. For a modestly strong attractive V, the SC correlations become dominant with an exponent $K_{sc}\sim1$. Our results provide a promising way to realize superconductivity in the doped Hubbard model.

DMRG phase diagram calculations: We studied the ground state phase diagram of spin-*1/2* J_1-J_2 XY model on the square lattice using both iDMRG and DMRG approaches [4]. We show that a plaquette valence bond phase is realized in an intermediate region $0.50 \leq J_2/J_1 < 0.54$ between a Neel magnetic ordered phase at $J_2/J_1 < 0.50$ and a stripy magnetic ordered phase at $J_2/J_1 \geq 0.54$. The plaquette valence bond phase is characterized by finite dimer orders in both the horizontal and vertical directions. Contrary to the spin-1/2 J_1-J_2 Heisenberg model, we do not find evidence for a quantum spin liquid phase in the J_1-J_2 *XY* model.



Experimental 2p XPS of Hematite compared to multiplet - only (green) and multiplet + cumulant calculations.

Combined multiplet + cumulant approach for treating effects of long- and short-range correlations in x-ray spectra [5]. This novel approach is based on a convolution of the atomic multiplet spectrum and the cumulant spectral function. The atomic multiplet spectrum describes the spitting of the local atomic states due to strong coulomb repulsion, while the cumulant describes longer range excitations such as charge transfer and plasmons that are missing in conventional ligand-field multiplet treatments. The real-space, real-time cumulant can be interpreted in terms of the density fluctuations induced by the sudden appearance of the core-hole. This

provides a real-time analysis of charge transfer and excitation densities to elucidate orbital contributions to the excitations. Results compare well with experimental XPS, which provide a direct probe of electronic correlations in condensed matter and molecular systems.

Future Plans

Cumulant approximation for the exciton Green's function: This approach extends the cumulant theory to XANES, while treating intrinsic, extrinsic, and interference effects in a consistent manner. Briefly, based on a simplification of work by Cudazzo and Reining, the valence electrons respond to an exciton-density rather than to the core-hole alone. This exciton density can be calculated via a solution of the Bethe-Salpeter equation using the OCEAN code or with FEFF10 and the final-state rule. The cumulant can be calculated via the real-time TDDFT or real-space Green's function approach.

Nearly ab initio multiplet-ligand field theory (MLFT). As an extension of our work on MLFT, we plan to develop a nearly *ab initio* approach for core-to-core x-ray emission spectra. We have recently applied this approach to the XES of transition metal oxides.

X-ray spectroscopy investigations of anionic redox in battery materials: In a continuation of our current efforts, we plan to perform charge-transfer, full atomic multiplet simulations for a variety of battery cathode materials, with a focus on charge valence states during voltage cycling and battery operation.

Theory and simulation of transient XAS in Warm Dense Matter: We plan to continue the development of a novel theoretical approach for simulations of transient XAS in non-equilibrium systems probed by intense pulsed x-ray sources. A preliminary account was presented at the March 2024 APS meeting.

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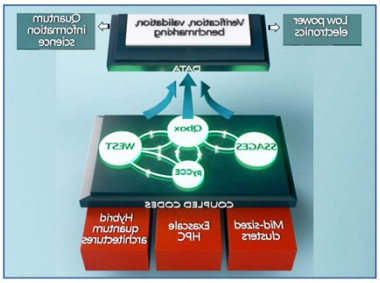
Midwest Integrated Center for Computational Materials

Giulia Galli (Argonne & Univ. Chicago), Maria Chan (Argonne), Juan de Pablo (Argonne & Univ. Chicago), Andrew Ferguson (Univ. Chicago), Marco Govoni (Univ. of Modena & Reggio-Emilia, Italy), Francois Gygi (UC, Davis), Joseph Heremans (Argonne), Jie Xu (Argonne), Jonathan Whitmer (Notre Dame Univ.)

Keywords: Quantum Simulations, Electronic structure calculations, Molecular Dynamics, Quantum Information, Low Power Electronics.



Research Scope

The Midwest Integrated Center for Computational Materials (MICCoM, http://miccom-center.org/) develops and disseminates interoperable computational tools – open-source software, data, simulation methods, and validation procedures – that enable the community to simulate and predict the properties of functional materials. We focus on (i) materials for quantum information science (QIS) and on (ii) materials for low power electronics. The former class of materials represents an area with potential impact in designing qubits, quantum sensors, and materials for quantum communications. The latter class of systems is another important area related to the design of materials for microelectronics, with potential, long term impact on semiconductor manufacturing. The Center's scientific strategy is built under the premise that the functionality of materials depends critically on the integration of dissimilar, often defective components and on the interfaces that arise between them. Hence our emphasis is placed on understanding and characterizing defects and interfaces, and on predicting finite-temperature spectroscopic and coherence properties. The description of dissimilar and defective components requires the development of first-principles electronic-structure methods, coupled to appropriate dynamical descriptions of matter and advanced sampling techniques, in order to predict multiple properties of complex systems and to capture the relevant length and time scales of importance to materials design. A key success of MICCoM has been the coupling of classical and quantum codes, together with their use on several high-performance computing (HPC) architectures. At present, we are developing methods to compute electronic excited states, including highly correlated states and excited state potential energy surfaces. To enhance the accuracy and efficiency of these methods, as well as those of the techniques used to predict ground state properties, we are developing automatic collective variable searches for advanced sampling. We are expanding the scope of codes performing advanced sampling, electronic structure calculations beyond density functional theory and coherence properties calculations, and we are porting several of them to additional GPU based HPC architectures. In particular, some of the codes developed by the center are being prepared for exascale and hybrid classical/quantum architectures. Hence, important activities of MICCoM are code maintenance, verification, and testing, including testing codes at scale. A summary of the

materials' properties of interest to the center together with methods and codes that are being developed is given in Figure 1.

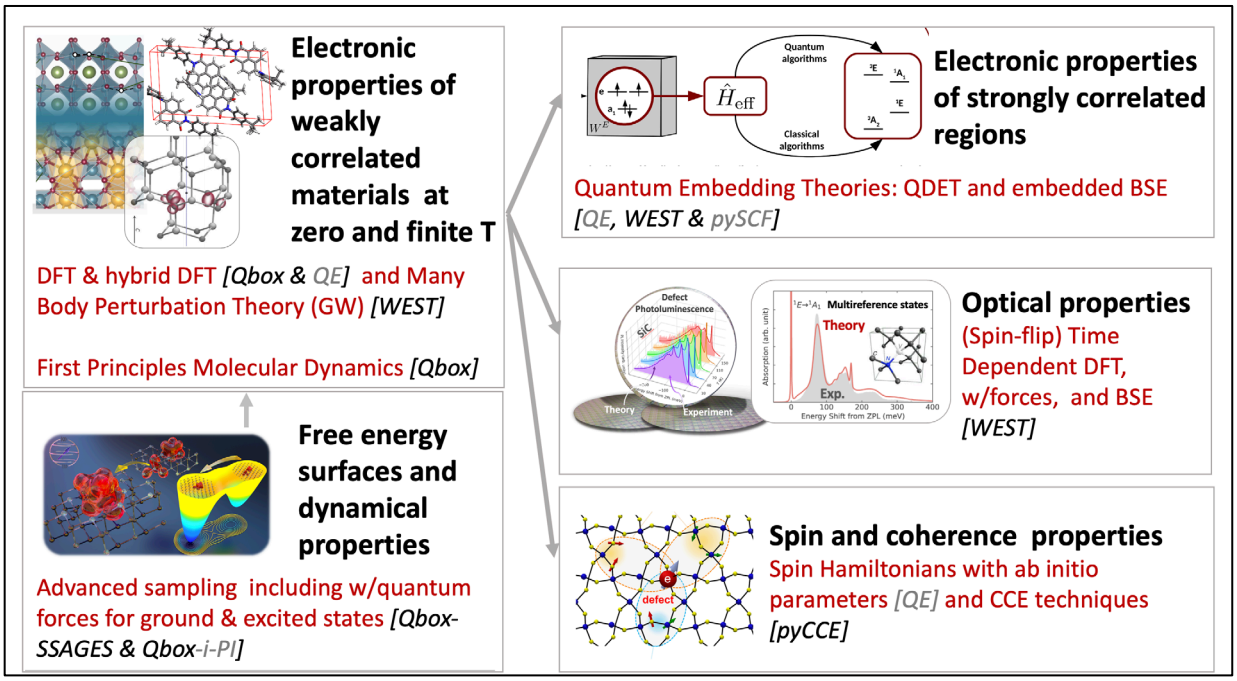


Figure 1. Summary of materials' properties (in black) of interest to the Center, together with methods used and developed (in red) and codes (within square brackets: codes developed within MICCoM are in black while codes used by the MICCoM team and developed elsewhere are in gray). QE (Quantum Espresso: https://www.quantum-espresso.org/; pySCF: https://pyscf.org/; i-PI: http://ipi-code.org/).

The Center's V&V and benchmarking activities are focused on the definition of first principles molecular dynamics and free energy benchmarks, quantum defect embedding theory benchmarks and validation of pseudopotentials used in electronic structure calculations. Our community outreach, in addition to documentation of codes and use cases made available to the public on a regular basis, includes the organization of tutorials, schools and workshops.

Recent Progress

The activities of the center have seen progress on multiple fronts: the development of methods and release of codes for advanced sampling [1] and electronic structure calculations of excited states [2]; the application of first principles molecular dynamics (FPMD) coupled with advanced sampling techniques to the study of defect formation in solids [3] and of FPMD to study vibrational properties of interfaces [4]; the study of free energy surfaces [5,6] and quantum vibronic effects in molecular solids [7] and defective materials [8]; the development and application of all-electron electronic structure methods to validate pseudopotentials [9], and the development of methods to study spin-coherence in solids [10].

Future Plans

Methods: Our ongoing method developments include (i) the implementation of new, improved features of the quantum defect embedding theory (QDET), in particular the implementation of self-consistent procedures; (ii) the calculation of excited potential energy surfaces using analytical forces derived within GW-BSE, building on our recent TDDFT implementation; (iii) the development of automatic Collective Variable searches for advanced sampling; (iv) the development and implementation of calculations of ab-initio spin-phonon dynamics in solids, which is critical to the study of materials for QIS.

Codes: We are expanding the scope of PySAGES with the addition of advanced sampling methods and unifying the methodologies and code bases for SSAGES and PySAGES. In addition, we are porting the WEST code, with all of its recent features, to AMD and, in the future, we will port some of the WEST features to Intel GPUs. WEST is already running on NVIDIA GPUs. We are also expanding the functionalities of the pyCCE code to include interacting spin defects and augmenting the use cases released to the community. Additionally, we are optimizing a brand-new version of Qbox implemented on NVIDIA GPUs. Through collaborations with other centers and research groups and TOPS (seed) projects, we are exploring the use of Qbox-generated datasets as training for deepMD potentials for large-scale simulations (with focus on oxide materials).

Applications: We are carrying out calculations on specific categories of materials and properties: (i) semiconductors and oxides for qubits and quantum sensing applications, with focus on the optical, coherence and formation properties of spin-defects in WBG semiconductors and oxides; and (ii) transition metal oxides and organic materials for low power electronics, including neuromorphic organic electronics, with focus on electrochemical-transistor (OECT).

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Understanding magnetic anisotropy and mixed valence in rare earth intermetallics

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Keywords: Mixed valence, magnetic anisotropy, Kondo physics

Research Scope

High performance magnets require large magnetizations, high operating temperatures, and strong magnetic anisotropy. The 4f electrons of rare earths are the only sufficiently strong source of this anisotropy currently known, and commercial magnets require large quantities of these critical materials. To use our current resources to their fullest potential, and find sustainable alternatives, we explore two complementary approaches: using cerium, the most abundant rare earth, and reducing the proportion of rare earths required by more effectively transferring anisotropy to the transition metals that provide the bulk of the magnetization. Both approaches require understanding the interplay between 4f valence, magnetic anisotropy, and electronic correlations. Our goal is to develop a comprehensive and quantitative picture of how mixed valence and magnetic anisotropy evolve in different rare earth materials by a collaborative combination of analytical theory based on model Hamiltonians, novel ab initio computational approaches; and experimental synthesis and characterization of targeted compounds to validate the theoretical approaches.

Our recent work has been along three distinct thrusts: understanding how valence fluctuations and magnetic anisotropy affect one another in Kondo and Anderson impurity models; improving density functional theory crystal field parameter calculations throughout a rare earth series, including benchmarking these against experiment in several series of compounds; and discovering novel rare earth transition metal intermetallics via machine learning combined with more traditional approaches.

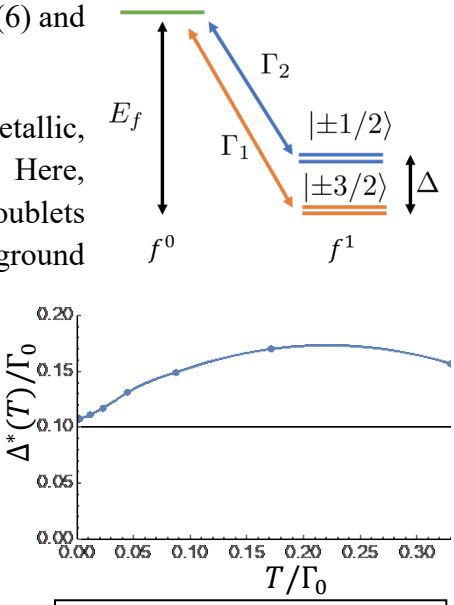
Recent Progress

Interplay of valence fluctuations and magnetic anisotropy in impurity models

We have shown that valence fluctuations can actually enhance the magnetic anisotropy energy at intermediate temperatures, which is potentially relevant for Ce-based permanent magnets. We have approached this problem in two complementary ways: using poor man's scaling to examine the renormalization group flows of a generic anisotropic SU(4) Kondo model with tetragonal crystal field, and using the numerical renormalization group (NRG) to study the related infinite-U Anderson model with two crystal field doublets and extract the renormalization of the crystal field.

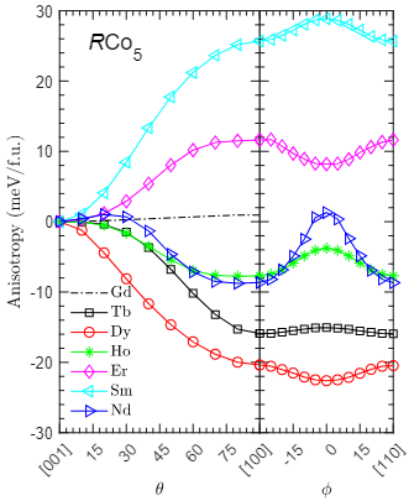
In the poor man's scaling, we first derived a generic anisotropic SU(4) Kondo model froma J=3/2 Anderson model with all symmetry-allowed hybridization terms using a Schrieffer-Wolff transformation, which led to four distinct Kondo couplings. We analyzed the RG equations and found the same fixed points as previous, less sophisticated approaches that kept a single SU(4) Kondo coupling: a marginally stable SU(4) Kondo fixed point and a stable SU(2) Kondo fixed point where one doublet is fully screened in the Kondo singlet and the other is totally decoupled. We calculated the corrections to the magnetic susceptibility and showed that instead of the single, overall screening [1] $\chi_{\perp,z}(T) = \chi^{SI}_{\perp,z}(T)[1 - 2\rho J_K(T)]$, there are three distinct components of the susceptibility screened by three distinct Kondo couplings, leading to an anisotropic onset of Kondo screening. In contrast to recent work on SU(2) S≥ 1 models [2], we do not find that the anisotropy changes sign for the anisotropic SU(4) models, either in poor man's scaling or NRG. Our approach here should generalize straightforwardly to the anisotropic SU(6) and SU(8) models relevant for Ce- and Yb-based materials.

We have also studied a simplified model of Ce in a metallic, anisotropic environment with an easy axis using NRG [3,4]. Here, there are two crystal field doublets (compared to the three doublets present in Ce), with the moment lying along the c-axis in the ground state doublet and in the plane for the excited doublet. There are also valence fluctuations where an electron hops out of the Ce orbitals into the sea of conduction electons, with energy cost |Ef| and hybridization $\boldsymbol{\Gamma}_2 = \boldsymbol{\Gamma}_1$ for both doublets. These valence fluctuations screen the moment at low temperatures, where we can see the anisotropic onset of Kondo screening for the c-axis and in-plane magnetic susceptibilities, where the c-axis susceptibility does not start to be suppressed until lower temperatures, in agreement with the poor man's scaling. Highly anisotropic susceptibilities persist for very large degrees of mixed valency. The magnetic anisotropy energy can be probed by calculating the spectral function for both the ground state and excited f-electrons, and extracting the effective crystal field splitting as a function of temperature. We find a small enhancement (~5-10%) of the effective crystal field splitting at low temperatures, which then increases starting above the Kondo temperature and has a maximum near the "valence" temperature, Tv, before decreasing again. The valence temperature is the temperature where roughly half of the change in valence (f0 occupation) between the high temperature minimum and the lowest temperatures has taken place, and it is orders of magnitudes larger than the Kondo temperature. For one case, with 8% mixed valence, the crystal field splitting was enhanced by 70%. Ce-based permanent magnets could potentially be tuned such that the desired operating temperatures are in this region of enhanced anisotropy.



(Top) Infinite-U impurity model used for NRG calculations. (Bottom) Enhancement of the crystal field splitting as a function of temperature for Δ=.1Γ_0, E_f = -8 Γ_0, where T_K is 10^{-4} Γ_0

Benchmarking rare-earth anisotropy in DFT+U



Magnetocrystalline anisotropy calculated in DFT+U for RCo_5, both in and out of plane. The minimum agree with experimentally determined ordering directions.

In order to quantitatively predict the magnetic anisotropy in rare earth intermetallics, the zeroth order requirement is to be able to calculate the magnetic anisotropy in density functional theory with Hubbard U (DFT+U), or at least to know when DFT+U results can be applied. Calculating this magnetic anisotropy has been a challenge for DFT, which fails to reproduce the correct Hund's rule ground state of 4f elements, due to significant orbital dependence of the self-interaction error for the 4f shell, and the lack of explicit proper orbital polarization treatment. Calculations may find erroneously find the true ground state as a metastable state above lower energy states that violate Hund's rules. However, as the self-interaction error and orbital polarization are, in principle, rotationally invariant, the magnetocrystalline anisotropy of the true ground state might be expected to remain correct if Hund's rules are enforced by hand. We have benchmarked this approach on materials with heavy rare earth atoms with saturated moments where Hund's rules are expected to be satisfied and the single Slater determinant description is suitable, notably RCo_5, RFe_{12}, and $R_2Fe_{14}B$, where the calculated easy directions (including easy axes, planes and conical angles) have all agreed well with low-temperature experimental measures. Here we show the magnetocrystalline anisotropy energy calculated for RCo_5, which predicts easy axis anisotropy for R = Sm, Er and easy plane anisotropy for R=Nd, Ho, Dy, Tb, and finds unexpectedly large in-plane anisotropies.

Work in progress is to continue this benchmarking for rare earth series compounds without magnetic transition metals, where the rare earth moments are not necessarily saturated, as well as for the light rare earths. Preliminary results suggest good agreement, with significantly larger changes in the crystal field parameters, A_n^m along the series than naively expected (and found in, say, the RMn_6Sn_6 and RV_6Sn_6 materials[5]).

Using machine learning to predict novel Ce-T intermetallics

We have integrated machine learning approaches with first-principles calculations to efficiently explore low-energy ternary Ce-Co-Cu compounds. Our study reveals several structures that are energetically as well as dynamically stable, along with a number of metastable ones close to the convex hull. Notably, two Co-rich metastable compounds exhibit high magnetization, suggesting their potential as doped Ce_2Co_{17}-based permanent magnets. We have also explored low-energy ternary Ce-Fe-X compounds, where X = Ag, Bi is immiscible with Fe and have found a number of low energy metastable states. We are currently attempting to synthesize these

predicted compounds, where we have potentially synthesized a material whose x-ray diffraction pattern is consistent with one of the predicted novel compounds.

Future Plans

In the future, we will continue to explore the interplay of anisotropy and mixed valence in the impurity models, where we can look at lower symmetries and models appropriate for Ce, Yb, as well as understand how the anisotropic Kondo interactions can be extracted from experimental data. We will also extend these ideas to lattice models, where slave particle mean-field theories will be combined with data from DFT+U to make realistic Kondo or Anderson lattice models that can capture a wide variety of experimental effects, which will be used to benchmark the approaches, as well as results from Correlation Matrix Renormalization Theory, a parameter free ab initio approach.

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Publications

All publications are in preparation.

Center for Predictive Simulation of Functional Materials

Paul Kent (PI), Panchapakesan Ganesh, Jaron Krogel, Ho Nyung Lee (Oak Ridge National Laboratory); Anouar Benali, Anand Bhattacharya, Ye Luo, Hyeondeok Shin (Argonne National Laboratory); Brenda Rubenstein (Brown University); Lubos Mitas (North Carolina State University); Raymond Clay, Luke Shulenburger, Joshua Townsend (Sandia National Laboratories

Keywords: Quantum Materials, Nanomaterials, Electronic Structure, Electron Correlation, High-performance Computing/Exascale

Research Scope

The Center focuses on providing accurate and reliable materials specific theory novel quantum materials and two-dimensional nanomaterials where strong electronic correlations, van der Waals and spin-orbit interactions delicately interplay with the underlying atomic structures, resulting in novel quantum phases and potential value for energy applications. In these materials, established electronic structure methods are challenged, requiring the development of new theories, methods, codes, and data, to provide simulations and analysis with sufficient reliability and accuracy.

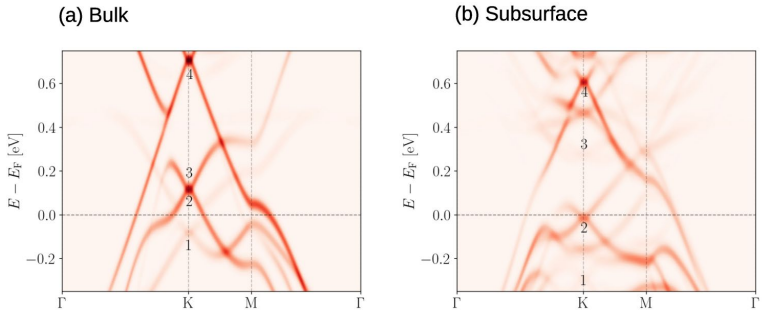
Project Members, CPSFM Annual Meeting, Nov. 2023.

The overarching goal of the Center for Predictive Simulation of Functional Materials is therefore the development, validation, and distribution of external-parameter-free methods and open-source codes to predict and understand the properties of functional materials, emphasizing those with strong electronic correlations, van der Waals and spin-orbit interactions. The Center develops advanced Quantum Monte Carlo (QMC) techniques and the open source QMCPACK code[1, 2] to reach this goal. By solving the Schrödinger equation stochastically, QMC techniques avoid uncontrolled approximations, and the few controlled approximations are increasingly testable and systematically reducible in real materials. As a Computational materials Sciences Center, the project applies these methods to specific materials and phenomena where the accuracy is required and also develops code, data, and tutorials, and holds workshops to expand the user base and for application to general materials.

Recent Progress

During the last years we have invested in fundamental improvements in the methodology to increase the range of materials and physical phenomena that can be studied with QMC methods, both in general and with the QMCPACK code. In particular, we have implemented the spin-orbit

interaction and the necessary steps to use the spinor wavefunctions. Additionally, we have continued to expand our range of highly-accurate correlation-consistent effective core potentials (ccECPs), see https://pseudopotentialibrary.org . These are developed and validated completely within a many-body framework and now cover approximately half of the periodic table. As a result of these enhancements, we have applied QMC to challenging scientific questions in a range of materials and phenomena including complex transition metal oxides[3-5], to 2D nanomaterials[6,7] and their heterostructures[8], and to layered Kagome-lattice quantum materials[9].



The Dirac crossing, where the Chern gap manifests, in bulk $TbMn_6Sn_6$ is only ~120 meV above the Fermi level, in close agreement with scanning tunneling microscopy experiments. The proximity of the Chern gap is corroborated by slab calculations, where the Fermi level lies within the same gap.[9]

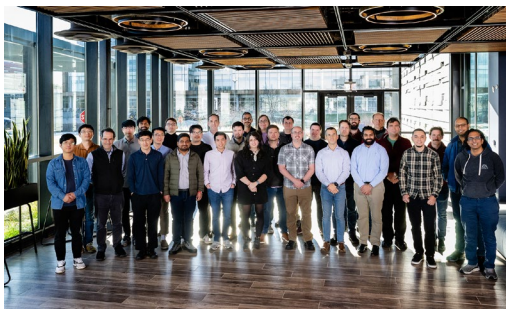
Combining many of these advances, in [9] we were able to perform predictive calculations of the energy level of the Chern gap in the Kagome metal $TbMn_6Sn_6$. This is the first application of QMC to Kagome systems with f-electrons. The presence of a Chern gap in this material can lead to chiral edge-states, enabling dissipationless transport, but uncertainties remained regarding the energy level of the gap. We resolved these uncertainties and provided a predictive approach for other Kagome compounds. Key steps included the development of ccECPS and an extensive series of calculations to enumerate the systematic biases of single-reference QMC calculations.

As mentioned above, we extended the range of ccECPs to include some lanthanides and other heavy elements of interest to the Center and requested by users, e.g. Zr, Nb, Pt, Gd, and Tb[10]. The transferability is adjusted using binding curves of hydride and oxide molecules. We address the difficulties encountered with f elements, such as the presence of large cores and multiple near-degeneracies of excited levels. For these elements, we construct ccECPs with core–valence partitioning that includes 4f subshell in the valence space. The developed ccECPs achieve an excellent balance between accuracy, size of the valence space, and transferability and are also suitable to be used in plane wave codes with reasonable energy cutoffs.

We developed a new twist-averaging approach for treating metals within QMC[11]. This twist-averaging technique is tailored to sample the Brillouin zone of magnetic metals, though it naturally extends to nonmagnetic conducting systems. The proposed scheme aims to reproduce the reference magnetization and achieves charge neutrality by construction, thus avoiding the large energy fluctuations and the post-processing needed to correct the energies. It shows the most robust

convergence of total energy and magnetism to the thermodynamic limit when compared against existing schemes, e.g., for the magnetic moments of α-Fe.

Significant effort has been spent preparing QMCPACK for the exascale era. This has required a large redesign to support the accelerated architectures from NVIDIA, AMD, and Intel and new algorithms so that the code can be both performant and maintained in a practical, sustainable manner. We recently presented (APS March 2024) full Frontier runs. We continue to work with the vendors, including HPE, on improvements to their software to reduce the workarounds currently needed and increase scientific productivity. This code is in full production use on CPU and NVIDIA GPU systems, and the workflows are becoming robust enough – for small systems -- for use in data generation for machine-learning applications.[12]



In person attendees of the hybrid QMCPACK workshop held at ANL 12-14 December 2023. Recordings are available on the QMCPACK YouTube channel and materials are archived on the QMCPACK GitHub organization.

In December 2023 we held a hybrid virtual/in-person users' workshop at Argonne National Laboratory, with 50 total participants. Besides tutorials and science presentations, significant time was dedicated to obtaining user feedback and requests. Addressing these requests will be part of Center activities in the remainder of 2024.

Future Plans

A key goal of our future work is to be able to apply the methodology where the input trial wavefunctions are most in question, such as when both the magnetic ground state and geometry of a material is incorrectly predicted by an approximate density functional. This is necessary to meet the challenges observed in numerous quantum and topological material systems. Accurate predictions for these systems require the optimization/prediction of both the trial wavefunction and geometry within the QMC, so that the results become independent of starting point. Demonstrations of these capabilities have been performed individually, finding the geometry of CrI_3 or wavefunctions in FeO. However, they have yet to be combined efficiently and in a general manner for materials, where the thermodynamic limit must be reached for comparison to experiment. Improvements in the treatment of many-body finite-size errors and new uses of symmetry are expected to be required. e.g. Because forces display a finite-size error, in most cases forces obtained in a small simulation cell cannot be used directly. Expanding the range of experimental properties that can be computed directly as well as increasing the ease of use and robustness of the methodology will continue to be a focus. This is necessary for wider use and also for use in machine learning and artificial intelligence-based approaches where the workflows must be highly robust and hands-off, as has been shown by other condensed matter theory program projects.

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10 Most Relevant Publications

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Elucidating the Nature of Chiral and Topological Phonons

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Keywords: phonons, thermal conductivity, chirality

Research Scope

The overarching goal of this project is to understand how vibrational chirality and other symmetries govern the fundamental quasiparticle couplings that determine thermal properties of bulk and low dimensional complex unit cell materials. To this end, we are closely examining symmetry-derived behaviors of phonons, electrons, magnons, and polaritons derived from density functional theory (DFT) and model Hamiltonian methods in non-symmorphic chiral and achiral materials. We are building a dynamical description of these excitations with fractional symmetries built in, thus elucidating connections between symmetry-enforced band crossings, quasiparticle interactions, topological behaviors, thermal transport, and unexpected observables from neutron, x-ray, and Raman scattering experiments. This general dynamical description has been applied to phonon and electron band structures in a variety of one-dimensional, van der Waals (vdW) layered, and bulk crystalline materials. This project is developing deeper insights into chiral and topological quasiparticles, while advancing understanding of thermal behaviors for smarter multifunctional materials.

Recent Progress

Non-symmorphic structures with twist symmetry: Building on previous efforts, we expanded and generalized our description of twist phonon phases in chiral and achiral crystals of different rotational symmetries, *i.e.*, non-symmorphic materials with 2-, 3-, 4-, and 6-fold screw operations. We further extended our previous insights regarding symmetry-protected band crossings, topological invariants, and observables of neutron scattering measurements to this broader class of materials. We specifically examined chiral and achiral phonons in elemental Te, GaN, FeSi, TiO_2 (comparing with inelastic neutron scattering (INS) data), $MgAs_4$, and In_2Se_6, all with varying degrees of symmetry [1]. This research enables more intuitive understanding of band degeneracies at Brillouin zone centers and boundaries on screw axes and varying degrees of topological charges at band crossings at intermediate wavevectors. Furthermore, we provided insights into scattering intensity variations among different zones in reciprocal spaces of chiral materials that were confounding expectations, *e.g.*, varying acoustic velocities among zones and finite slope zone boundary modes.

Vibrational spectra and lattice thermal transport in large conventional unit cells: We stretched the twist phase concept to describe translational phases relating the dynamics of primitive unit cells with their larger conventional unit cell counterparts. This occurs in complex vdW layered and covalently bonded systems for which a conventional unit cell is more convenient to use than the lower symmetry primitive cell. We applied this translational dynamics to understand phonons

and transport in N_2, GeTe, and $CrCl_3$ [2, 3]. Experimental geometries and theoretical analysis are often based on conventional cells despite having larger numbers of atoms. This work also led to a novel understanding of INS observables (see Fig. 1 for INS data for $CrCl_3$), interaction of magnetic structure with vibrational dynamics, and has made transport calculations significantly more efficient: it combines the relative simplicity of conventional structural descriptions with the efficiency of primitive cell calculations. Thus, we can make thermal transport calculations in large conventional cells at a significantly reduced cost. This will allow us to tackle more complex materials in future calculations, *e.g.*, spin-dependent phonon thermal transport in α-$RuCl_3$.

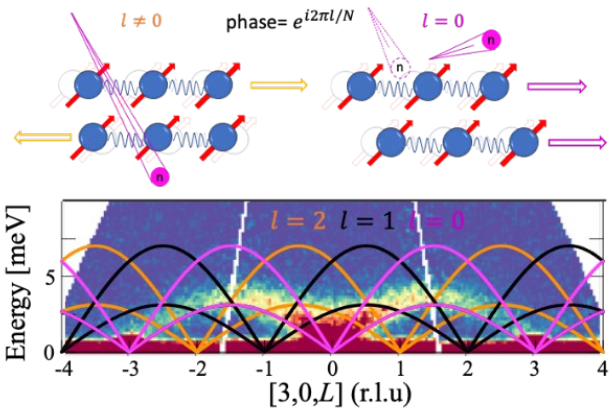

Figure 1: (top) Neutrons are strongly scattered when layer vibrations are in-phase ($l = 0$). (bottom) Measured and calculated dispersion of $CrCl_3$. Destructive interference conditions for $l \neq 0$ (black and orange curves) limit scattering intensity.

Scattering driven thermal transport in short-period superlattices: As alluded to above, structural twist symmetries of AlN and GaN lead to phase related folded bands and degeneracies along their 6_3 screw axes. This symmetry is broken in superlattices (SLs) composed of these, which leads to gapping of the bands and increased band folding. These dispersion features lead to reduced phonon velocities and increased intrinsic phonon scattering, both of which lower cross-plane thermal conductivities. Our DFT-derived calculations of thermal transport in coherent (*i.e.*, no boundary resistance) short-period SLs demonstrate that lowered phonon velocities play only a minor role in this reduction. Increased phase space for scattering underlies most of the conductivity reductions with increasing SL period [4].

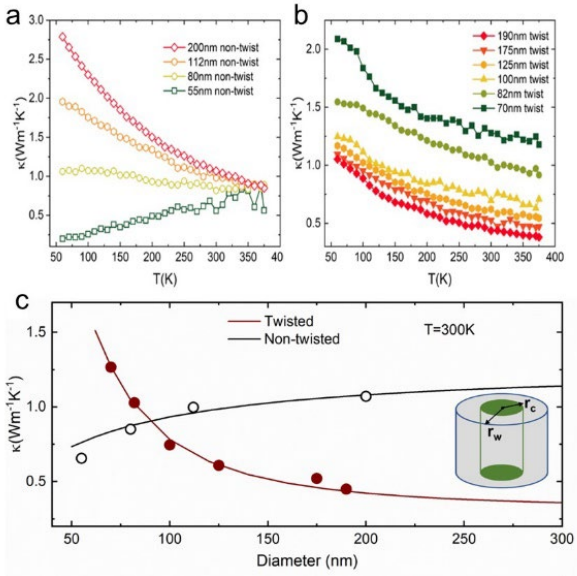

Figure 2: Temperature-dependent measured thermal conductivities of (a) non-twisted and (b) twisted GeS nanowires with varying diameters. (c) DFT-informed core-shell thermal conductivity model (curves) of diameter dependence of twisted and non-twisted nanowires at room temperature.

Phonons and polaritons in nanowires: Contrary to typical boundary scattering expectations and transport in non-twisted GeS nanowires, measurements have demonstrated that thermal conductivities of twisted GeS nanowires increase with decreasing diameter (see Fig. 2). Structural characterizations and DFT vibrational and transport calculations point to thermal

conductivity enhancements in the twisted nanowire cores deriving from dislocation-induced strain and compression. This is further reinforced by agreement with diameter-dependent thermal conductivities for twisted and non-twisted wires from a DFT-informed core-shell thermal conductivity model. In separate work, combined nanoscale measurements and calculations demonstrated significant surface phonon-polariton thermal transport in SiC nanowires with gold coated endcaps [5]. Calculations based on an equilibrium Landauer ballistic transport formalism severely underestimates this transport pathway and hints at the highly non-equilibrium nature of the polariton transport mechanism. Significant polariton derived thermal conductivity enhancements offer novel design pathways for improving thermal management in microelectronics.

Future Plans

Over the course of the next year, we will continue to build on the above-described efforts to construct theoretical and numerical algorithms to improve our understanding of quasiparticle interactions and resulting thermal properties of chiral and achiral materials with varying degrees of freedom. We plan to build better transport theories for bulk and surface polariton modes to understand how behaviors vary over different geometric configurations and understand their non-equilibrium nature. We will also combine large-scale supercell calculations with our recently developed dynamical phase symmetry concepts to derive insights into charge density wave and phason behaviors of twisted and incommensurate systems. Another investigative track involves applying phase symmetry concepts to understand chiral magnons, phonons, and their interactions in magnetic materials, particularly as these govern transport behaviors.

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Center for Computational Study of Excited-State Phenomena in Energy Materials (C2SEPEM)

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Keywords: Interacting-particle Green's functions; multiple-particle excitations/spectroscopies; driven time-dependent phenomena; interoperable and portable algorithms/codes; experimental validation.

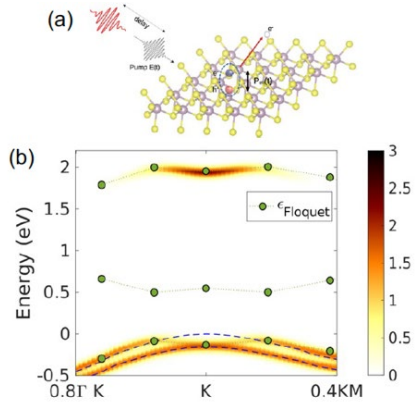
Research Scope

Excited-state phenomena in materials typically give rise to their defining attributes and determine their usefulness. Computational discovery and design of functional materials utilizing excited-states properties require accurate and predictive descriptions of many-body interactions, such as electron-electron (e-e), electron-hole (e-h), electron-phonon (e-ph) couplings, and the correlation and coherence of excitations, both in- and out-of-equilibrium**.** The mission of the Center for Computational Study of Excited-State Phenomena in Energy Materials (C2SEPEM) is to develop new *ab initio* theories, methods, algorithms, and open-source software to elucidate and predict excited-state phenomena. It aims to advance the frontiers of fundamental theories and methods, their efficient and scalable implementation utilizing exascale and post-exascale high performance computing (HPC) resources, their experimental validations, and open-access software with interoperability, through efforts from a diverse, multidisciplinary, and multigenerational team. In the past two years, C2SEPEM has developed and applied advanced *ab initio* methods for ultrafast dynamics and correlated multiparticle excitations and their interactions, discovered novel properties and deepened understanding of emergent quantum material systems, and scaled the software on DOE leadership class HPC resources.

Recent Progress

In the recent two years, C2SEPEM advanced the frontiers of *ab initio* computation of excited-state properties via a multi-pronged strategy, resulting in 50+ publications in *Nature, Science, Proceed. of Natl. Acad. Sci. USA, Nature family journals, Phys. Rev. Lett., Nano Lett., ACS Nano, J. of Comp. Phys.,* etc. journals. Below are some selected highlights.

With our time-dependent adiabatic GW (TD-aGW) method, we discovered an exciton-Floquet phenomenon (see figure), predicted shift current vortex crystals in moiré superlattices, and discovered an excitonic channel for transitions of intralayer to interlayer excitations in the ultrafast optical responses in two-dimensional (2D) heterolayers. With our GW perturbation theory (GWPT), we revealed strong self-energy effects in the e-ph coupling of transition-metal oxides. We developed new approaches to study exciton transport in molecular crystals, phonon screening on excitons, and exciton effects in second-order nonlinear responses as well as in the X-ray absorption spectra of matter.



(a) Time-resolved ARPES spectroscopy (tr-ARPES) setup. (b) tr-ARPES intensity (color) for monolayer MoS2 from *ab initio* TD-aGW theory. Green dots are exciton-induced Floquet quasienergies.

A strategy to compute e-h interactions in moiré superlattices led to a 10^6 speedup in computation and the discovery of novel intralayer moiré excitons. A new theory on exciton-phonon coupling was developed, as well as an *ab initio* method on exciton lifetime and optical line width profile via exciton–phonon interactions. New techniques, such as a mixed stochastic-deterministic approach, a Lanczos method for frequency-dependent interactions, and a non-uniform sampling technique, speed up GW and GW plus Bethe-Salpeter-equation (GW-BSE) calculations by orders of magnitude. A novel dynamic mode decomposition approach was developed for the long-time propagation of the Kadanoff-Baym equations (KBE) of model systems.

We expanded the interoperability, exascale-readiness, and portability for our BerkeleyGW package, which fully scales to leadership class HPC centers. We optimized GPU-acceleration across different GPU architectures including NVIDIA, AMD, and Intel. BerkeleyGW serves as the main engine on many projects at DOE HPC centers, including 3 INCITE projects, an ALCC project, and was selected in the NESAP and Early Science Program. Our theory and software advances were validated and led to cutting-edge applications. These include the experimental validation of our predictions on moiré excitons and phonon-induced exciton localization/anisotropic transport in molecular crystals, which required the development of a novel laser-STM experimental technique and spatio-temporally resolved ultrafast spectroscopies.

Future Plans

Our plans are to develop new *ab initio* quantum many-body methods and advance the Center's main highly versatile, interoperable, and portable software package BerkeleyGW to further expand the frontiers of excited-state phenomena in the following areas.

Theories for Correlation and Nonequilibrium Phenomena. This thrust area focuses on new ideas and theoretical methods for predictive calculations and discoveries of phenomena from different

interactions (light-matter, e-e, e-h, e-ph, exciton-phonon, exciton-spin, etc.). We will study third- and higher-order nonlinear optical responses with excitonic effects using TD-aGW, opening a door for understanding phenomena such as four-wave mixing, optical Kerr effect, and quantum geometry and topology. We will investigate excitonic effects in light-induced Floquet states and their nonequilibrium dynamics, a novel avenue for dynamical manipulation. We will study the exciton-spin dynamics in 2D magnets to understand transient magneto-optical phenomena. We will develop *ab initio* theories for the spectroscopic properties of excitonic insulators, a novel quantum phase of matter. We propose to study e-ph coupling phenomena with GWPT, including phonon-mediated superconductivity, zero-point renormalization, and their dimensionality effects. We will explore the hybridization between exciton states induced by phonon screening as well as the frequency dependence of the phonon screening kernel with the aim of better understanding their impact on the optical absorption spectra. We will incorporate e-ph interactions into the TD-aGW framework to capture from *ab initio* nonequilibrium exciton-phonon dynamics and dephasing and decoherence processes.

Advanced Methods and Algorithms. This thrust area creates advanced numerical methods and algorithms for the concepts and theories developed above. We plan to build on the successful methods from the previous period and develop new, impactful computational techniques. We will expand on the Lanczos method to efficiently perform large-scale GW and GW-BSE calculations. We will extend the hybrid stochastic-deterministic approach to speed up calculations of optical and excitonic properties. We will generalize the full KBE model solver for real-materials simulations of non-equilibrium dynamics, meanwhile incorporating machine learning methods for computational acceleration, such as the long short-term memory and the physics-informed neural operator. We will utilize exciton Wannier functions to compute nonlinear optical responses and Berry phase effects. We will design neural networks that predicts GW quasiparticle excitations with atomic configurations as input, facilitating large-scale calculations and the construction of GW materials database.

Software development in exascale and post-exascale era. This thrust area focuses on the development of our open-source community code, BerkeleyGW, and its efficient employment in HPC, especially for exascale and post-exascale computing applications, and interoperability to different platforms. We will advance the code with the continuous DOE HPC developments, and achieve portability across different hardware architectures utilizing directive-based programming models. We plan to adapt and scale our Center-developed new methods, such as TD-aGW, GWPT, and full KBE solver, for leadership class HPC systems. To increase interoperability, we will collaborate with other community software (e.g., EPW) to enable novel physics previously inaccessible. We will leverage hardware solutions offered by third party cloud infrastructures and provide BerkeleyGW as a community computational tool and library that can be accessed as needed and incorporated into complex workflows, i.e., “BerkeleyGW as a service.”

Applications of advanced methods and their validations. This thrust area performs verifications and validations of our methods, as well as applications to functional materials to demonstrate their impact and versatility. Our experimental effort will focus on advanced optical spectroscopies (including ultrafast pump-probe and micro spectroscopy) of novel excitonic phenomena to validate predictions, e.g., of exciton insulator states as well as the influence of excitons on the Franz-Keldysh effect in wide bandgap insulators such as GaN. We plan to validate calculations of exciton transport and diffusion in molecular crystals, halide perovskites, and 2D materials against femtosecond-scale stroboscopic transient microscopy technique. We plan to study the influence of carrier density on multiparticle excitations in 2D materials and in bulk polar and nonpolar systems. We will explore the origin and nature of emergent excited states at interfaces, in collaboration with experimental efforts. We will verify our TD-aGW approach with developers of the Yambo code which exploits a different formulation.

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None.

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Theory of Materials

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Keywords: photo excitations and charge dynamical phenomena; quantum phases with novel charge, spin & topological states; emergent properties and behaviors at multiple scales; *ab initio* and correlated electron computational and data science methods

Research Scope

The goal of this program is to explore, understand and compute material properties through theory and modeling, as well as to develop concepts and methods for such studies, and thus to produce enabling knowledge relevant to the basic missions of DOE-BES. A variety of techniques within quantum many-body framework are employed, including first-principles electronic structure calculations, field-theoretical methods and topological invariants, new conceptual and computational methods suitable for complex and strongly interacting systems, and advanced algorithms. Emphases are both to investigate realistic material systems as well as physically motivated model systems. Research is carried out in four areas: 1) photoexcitations and charge dynamical phenomena; 2) quantum phases with novel charge, spin & topological states; 3) emergent properties and behaviors at multiple scales; and 4) *ab initio* and correlated electron computational and data science methods. Photoexcitations and transport properties are crucial to phenomena such as optical response, photocatalysis, optoelectronic behaviors, etc. that are central to energy research and applications. Activities in areas 2) and 3) are aimed at understanding and discovery of new phases and new classes of materials with novel properties. Activities in area 4 are aimed at developing cutting-edge computational methods that allow accurate, predictive studies. Close interactions with experimentalists are maintained. The program uses and interacts strongly with unique resources at LBNL, including the National Energy Research Scientific Computing Center (NERSC), the Molecular Foundry, and the Advanced Light Source (ALS).

Recent Progress

Our activities in the last two years covered advances on novel phenomena in atomically thin reduced-dimensional materials, properties of complex bulk materials, exotic phases and properties of quantum materials, and new theoretical and computational methods. Over 30 papers were published including publications in *Nature*, *Science*, *Proc. Nat. Acad. Sci. USA*, *Phys. Rev. Lett., Nature family journals, Nano Letters, J. Am. Chem. Soc.*, etc. The work has led to formulation of new concepts, prediction of novel phenomena, and explanation of experiments for a range of materials. Some selected results are:

For reduced-dimensional systems, we have: unmasked the band renormalization effects driven by excitonic correlations in monolayer semiconductor MoS2; performed analysis and calculations of novel moiré excitons in 2D materials and predicted the existence of intralayer charge-transfer moiré excitons in van der Waals superlattices; performed first-principles calculations on the magnetism and interlayer bonding in pores of Bernal-stacked hexagonal boron nitride; elucidated the spin-dependent vibronic response of a carbon radical ion in two-dimensional WS2; predicted from *ab initio* the spatial confinement of intralayer exciton in moiré superlattices and with experimentalists verified the results using hyperspectral imaging; studied a 2D topological insulator (substrate-supported monolayer bismuthine) leading to discovery of optically controlled single-valley exciton doublet states with tunable internal spin structures and spin magnetization generation; elucidated excitons and trions in MoTe2 monolayers as a function of carrier doping; studied the structure and properties of new forms of 1D chains in nanotubes -- GeX_2 (X = S or Se) and MX3 (M is Cr or V, and X is Cl, Br, or I); studied how charging can drive the phase transition and stability of the different atomic configurations of nanotube encapturated HfTe2 nanoribbons.

For properties of complex bulk materials, we have: studied phonon-induced localization of excitons in pentacene, showing that anharmonic phonons localize excitons and strongly renormalize exciton band structure, explaining experiments; studied Au-based perovskites, reporting the first realization of spin-polarized Au in a solid; revealed the central role of electron-phonon coupling in the phase transitions of excitonic insulator candidate Ta2NiSe5; studied the origin of isolated flat bands in copper-substituted lead phosphate apatite, a purported room temperature superconductor, suggesting that site-selectivity was necessary for correlated physics in this system; elucidated the effects of anisotropy and isotope in the superconducting properties of solid hydrogen and deuterium under high pressure.

For exotic phases and properties of quantum materials, we have: developed an approach for bosonization of topological insulator and superconductors (extending our previous work of bozonizing gapless free Dirac and Majorana field theories); studied the O(6) non-linear sigma model, with the level-1 Wess-Zumino-Witten term in 3+1 space time dimensions - showed that this theory exhibits electro-magnetic duality, where the charge and monopole are the solitons of the O(6); demonstrated that in frustrated magnets when several conventional orders compete and are “intertwined" by a Wess-Zumino-Witten (WZW) term, the possibility of spin liquid arises which can carry fractional spins and obey non-trivial self/mutual statistics; showed that chiral gauge transformation induces a Berry’s phase which is equal to Fujikawa’s Jacobian in high energy physics; demonstrated emergent superconductivity and non-Fermi liquid transport in a doped valence bond solid insulator, investigated using a sign-problem-free quantum Monte-Carlo method.

For theoretical and computational method developments, we have developed: a method and the computational code for the optical and magneto-optical properties of magnetic two-dimensional materials and applied to the understanding of ferromagnetic monolayer $CrBr_3$; a full

spinor GW-BSE formalism and computer code for the computation of the optical properties of materials with strong spin-orbit coupling; a framework for understanding vibrational side bands of deep in-gap states associated with point defects in 2D materials; a many-body second-order perturbation theory and analysis for the optical response of the exciton lifetime and optical linewidth via exciton-phonon interaction; a gauge consistent method for Wannier interpolation of electronic states and electron-phonon coupling; a theory of spin liquid from the Ginzburg-Landau action with topological terms; a method for using linear magneto-conductivity as a DC probe of time-reversal symmetry breaking.

Future Plans

Planned activities will be focused in the 4 thematic areas stated above: 1) photoexcitations and charge dynamical phenomena; 2) quantum phases with novel charge, spin & topological states; 3) emergent properties and behaviors at multiple scales; and 4) *ab initio* and correlated electron computational and data science methods. Some selected projects include:

- Understanding structural relaxation/reconstruction on correlated symmetry breaking ground states/mechanisms in moiré heterostructures.
- Understanding how non-Hermitian topological phases can arise from the imaginary self energy of interacting electrons.
- Developing an accurate DFT-based interatomic potential for moiré stacks of transition metal dichalcogenide monolayers, allowing the exploration of electronic structure of arbitrary twist angles.
- Theory and computation of exciton-phonon coupling induced new pathway for ultrafast intralayer-to-interlayer exciton transitions and interlayer charge transfer in TMD heterostructures.
- *Ab initio* and model studies of properties of excitons in topological insulators.
- Exploring the temperature-dependence of exciton binding energies of novel semiconductors and halide perovskites, using our newly developed phonon screening methodology.
- Theory and computation of excitonic properties of 2D molecular crystals on monolayer transition metal dichalcogenides.
- *Ab initio* study of time evolution of excitons in molecular solids.
- Study of the optical response of 1D phosphorus nanoring/helix structures, in particular excitons of mixed dimensionality in these systems that have been recently synthesized.
- Exploration of properties of axions and possible experimental signatures in magnetic topological insulators.
- Understanding and classifying the topological natures of systems protected by “dynamical space-symmetries”, including the search for a general recipe for model constructions in engineered Floquet topological phases.
- *Ab initio* study of electronic correlation effects on novel topological materials.

- Investigation of an identified candidate material for correlation-induced topological phase transition ($CaMn_2O_4$) using DFT calculations and model studies. Current work is building up a Wannier-based tight-binding model to incorporate in a DMFT solver.
- Study of doping site selectivity and the role of electron-phonon coupling in the experimental report of superconductivity in Cu-substituted lead apatite.
- Study of mechanism for the experimental observation of strong electromigration of molecular adsorbates on graphene.
- Understanding the stability and properties of new successfully synthesized novel 1D semiconductor alloys based on $Si_{1-x}Ge_x(Se,S)_2$.

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None.

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Correlations and Dynamics in Quantum Materials

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Keywords: Hamiltonian Engineering, Artificial Quantum Matter, Quantum Dynamics, Open Quantum Systems

Research Scope:

Engineering desired Hamiltonians in artificial quantum many-body simulators is an important challenge. Usually, the Hamiltonians realized natively in these systems from qubits whose states differ from one another by some internal degrees freedom of atoms. When these atoms are thus made to interact with one another, rarely do many relevant symmetries in conventional condensed matter systems, such as in plane or SU(2) spin rotational invariance manifest natively. On the other hand, various many-body symmetries are necessary for protecting the topological nature of the quantum ground state, and essential to realize edge modes. Thus, it is an important task to be able to engineer these symmetries for the purposes of controlled quantum simulation of those materials using these artificial matter setups. A natural way of implementing such symmetries is using dynamical decoupling techniques [1,2]. For instance, to implement full spin SU(2) symmetry, one can imagine pulsing the atomic system rapidly and periodically to implement discrete rotations of the qubit. If these rotations effectively span the surface of the Bloch sphere in a uniform way, the effective Floquet Hamiltonian can develop an SU(2) spin-rotational invariance. While this idea is appealing at the outset, driving in general causes heating, and is especially pronounced in a many-body system [3]. Moreover, imperfections in the driving, or noise from the environment, can lead to a loss of coherence and the symmetry that is intended to be generated dynamically. The aim of our work is to examine these issues.

Recent Progress:

In previous work [4], we developed a class of "polyfractal" dynamical decoupling protocols for realizing multiple Z_2 symmetries in a many-body quantum spin system, and provided bounds for the timescales up to which the effective symmetric Floquet Hamiltonian could be used to describe the dynamics of the system at stroboscopic time intervals. In a subsequent work [5], we studied how such symmetries could be used to realize and protect bosonic symmetry protected topological phases in quantum spin chains, and Majorana edge zero modes in a system of imperfect (overlapping) Majoranas.

Besides intrinsic heating caused by driving, these protocols are also susceptible to noise and imperfections of driving, which also tend to destroy the effective symmetries. In [6], we developed a novel protocol to study the effect of noise on these protocols. Specifically, we consider randomized and quasi-random imperfections in the times at which stroboscopic dynamical decoupling pulses are applied. We justify such an approach in two ways. First, we argue that spectrally, the most important noise contribution comes from noise at frequencies proximate to the driving frequency. This is because noise at very high frequencies is largely ineffective at heating the system because it requires multiple spin flips for the system to absorb the radiation and heat up, and is thus exponentially suppressed in the order n of flips required. Noise at much lower frequencies is also ineffective since it represents slow fluctuations which

can be suppressed by driving (similar to spin-echo protocols). It is thus expected that the noise at frequencies $f \sim 1/T$, where T is the drive period that is most important, which is born out in our simulations. Second, while noise can couple to arbitrary operators in the system, the most pernicious couplings are those that involve operators associated with the many-body symmetry being dynamically engineered. Fortunately, these are also the same operators that we use to drive the system with. With the above in mind, irregularities in the application of drive at the timescale of the drive period T capture the most important features of noise the system is subject to.

Although completely random irregularities in the applications of the driving fields represent a faithful model of noise in the system, numerically it is difficult to evolve the system for the (quasi-) exponentially long times we need to see the breakdown of the dynamically engineered symmetries. To address this issue, we introduce quasi-random noise, which appears random on short time scales, but has correlations on longer time scales. Specifically, we consider the case when the irregularity in the application of the noise is binary---either the driving pulse is applied early by some time εT, or late by a related time---and whether the pulse is applied later or earlier than period T is determined by the Fibonacci sequence. We note here we have multiple driving pulses and this creates additional technical complications in implementing such a sequence although these challenges can be surmounted. In particular, we are able to simulate dynamics generated in the presence of such quasirandom noise to exponentially late times using the recursion relations built into the Fibonacci sequence.

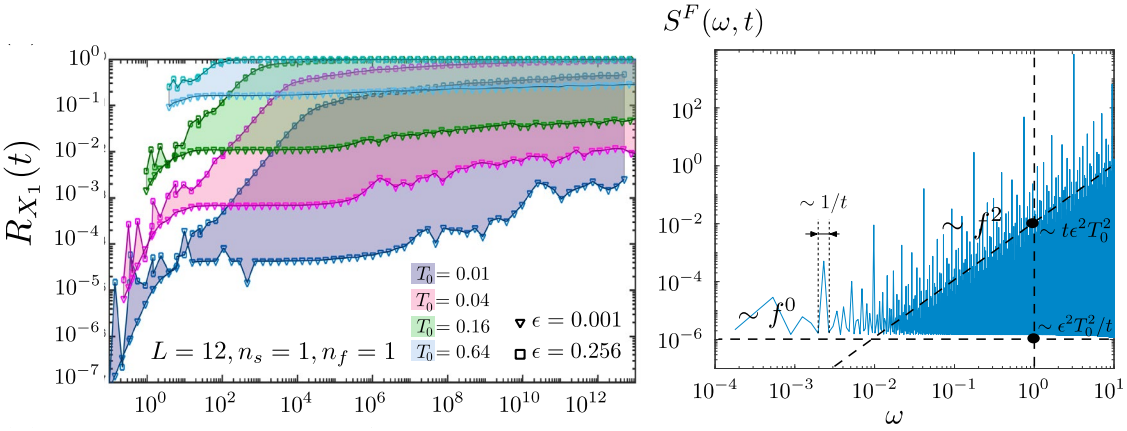

Fig 1a (left): decay of autocorrelator of many-body Z_2 symmetry generator X_1 in the presence of quasiperiodic noise. Fig 1b (right): Spectrum of quasiperiodic noise simulated has a noise floor that decreases in time as 1/t---this floor effectively describes the `random' part of the noise at various timescales, and is crucial in computing relaxation dynamics.

We find (see Fig. 1a) that the dynamically engineered symmetries in the Floquet Hamiltonian relax in three stages. Provided driving frequencies are faster than microscopic timescales, initial relaxation on short timescales gives way to a prethermal plateau. This prethermal plateau then gives way to linear heating ramp that can be explained by Fermi's Golden Rule like arguments. Up to this point, relaxation dynamics in true noise and quasiperiodic noise is identical and justifies our study of quasiperiodic noise which allows us to push the study of dynamics to long time scales. Unique to quasiperiodic noise, a final

slow logarithmic decay is observed. The emergence of this logarithmic plateau can be explained by the appearance of isolated spectral peaks in the quasiperiodic noise spectrum above the noise floor which decays in time as *1/t* (see Fig. 1b). We provide convincing numerical evidence and analytical arguments to justify the scaling of all relevant time scales and relaxation amplitudes with system size and noise amplitude. A summary of the results are provided in Fig. 2.

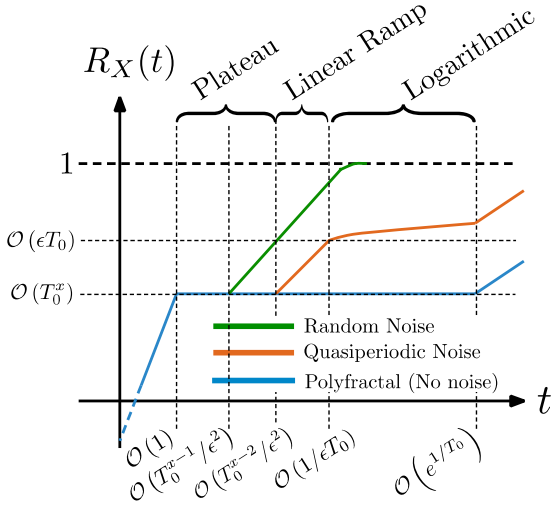

Fig 2: Summary of relaxation dynamics in the presence of random and quasirandom noise.

Future Plans:

In our work to date we demonstrated the utility and stability of novel dynamical decoupling protocols for implementing nontrivial symmetries in many-body interacting systems, focusing on one dimensional systems. In the future we will apply these ideas to higher dimensions, and also to stabilization of more complex symmetries.

A holy grail in this regard is the implementation of gauge symmetries, as well as higher form symmetries. This will likely require additional resources in the form of extra degrees of freedom (ancillas). We expect that using these techniques, generic low-symmetry Hamiltonians can be sculpted into any desired Hamiltonian given enough dynamical control and sufficiently large Hilbert space (including ancillas).

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Nonperturbative Studies of Functional Materials Under Nonequilibrium Conditions (NPNEQ)

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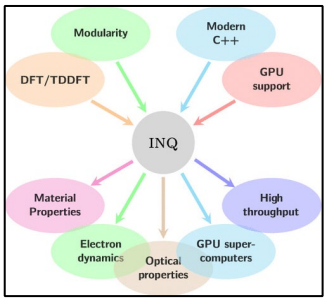
Keywords: quantum dynamics, electron-ion-spin-photon coupling, open source software, TDDFT, GPU computing

Research Scope

The research scope of the CMS Software Center NPENQ is to develop, validate and disseminate open source software tools for simulation of ultrafast quantum dynamics. Such software includes a state-of-the-art GPU optimized (TD)DFT code named INQ and time-dependent tight-biding code TBINQ, whose parameters could be derived from INQ simulations and will be used to extend the time and length scale of quantum dynamics simulations. Our software dissemination effort will leverage hackathon, internship and collaborative scientific research including our own experimental effort on code validation in topics such as high harmonic generation and non-linear optical effects.

Recent Progress

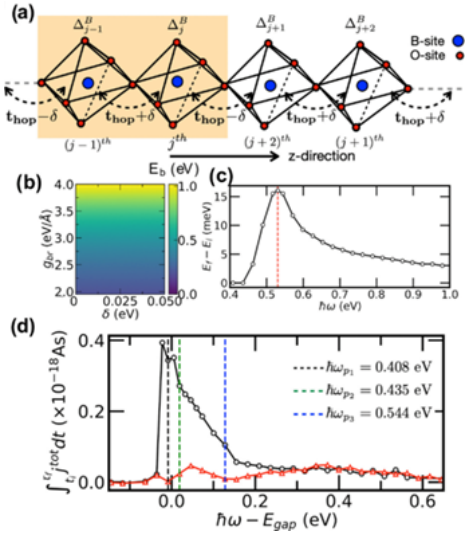


Software development/dissemination: In the previous period, we developed INQ code with basic functionalities such as ground state DFT calculations, TDDFT and Ehrenfest dynamics simulations with PBEGGA on multiple GPUs (Ref. [1]).

In the past two years, we introduced a few new features, ported on multiple DOE/ASCR HPC systems, and improved user interface. New features: spin-polarized collinear calculations for magnetic systems and simulations of ultrafast demagnetization. Time-dependent hybrid functionals including optimizations such as the low-rank Hartree-Fock operator (ACE) (Ref. [2]). Porting on DOE/ASCR HPC systems: INQ code was ported to NERSC Perlmutter (Nvidia GPU), and we are currently working on porting on Argonne Aurora (Intel), Oak Ridge Frontier (AMD MI250 GPU), LLNL El Capitan (AMD MI300 APU) wherein technical information are gathered by Andrade and Correa via participating the Hackathons including a series held by LLNL. Improving user interface: INQ can now read various input formats, such as CIF and Vasp's POSCAR, to maintain compatibility with Materials Project's databases and a programmable building of structures. Note: Materials Project switched data format from CIF/POSCAR to PyMatgen (future plan). Community engagement: has been fostered through the Rutgers Hackathon (Pavanello Group), the TDDFT School, and Berkeley GW school, as well as the ongoing LLNL's Computational Chemistry Materials Science Summer Institute (Director: Alfredo Correa).

Scientific research: We have pioneered the use of real-time tight-binding simulations as a tractable method for understanding photocurrents in complex materials (Pub. [4-5]). This approach has the advantage of capturing non-perturbative effects which lie beyond conventional response theory, including saturation, strong-field effects, and photo-induced phase transitions. The use of a tight-binding basis allows us to access longer time and length scales beyond the reach of density functional theory, addressing issues such as relaxation and long-range order.

Our work on the materials system of $Pr_xCa_{1-x}MnO_3$ has resulted in the discovery of a new way to enhance the bulk photovoltaic effect by optically induced phase transitions (Pub. [4]). Using real-time simulations, we show that this strongly correlated oxide perovskite displays strong photocurrent enhancement when it is driven into a hidden magnetic phase transition. This highly non-perturbative phenomenon shows that novel routes to photocurrent enhancement may be found through transient magnetic and orbital orders.



Simulations of photocurrent (a) in a linear perovskite-type chain, considering electron-phonon interactions (b) and absorption (c) processes. Turning on phonon dynamics strongly enhances the photocurrent (d).

In Pub. [5], we study ballistic photocurrent driven by optical phonon modes in a polaronic ferroelectric. We show that optical phonon modes can be harnessed to drive strong bulk photocurrents, instead of acting as a purely dissipative mechanism. We find that electronic ferroelectric materials with polaronic character are especially suitable for this. Time domain methods are used to simulate phonon-assisted ballistic current. Being non-perturbative, this method allows us to observe a crossover between linear and nonlinear photocurrent regimes. We link our results to materials design considerations, finding that bonding asymmetry directly correlates to stronger ballistic photocurrents, while electron-phonon coupling strength does not. In parallel, Lindenberg is developing experimental efforts capable of tracking these photocurrents directly through time domain terahertz emission spectroscopy, capturing the radiated electromagnetic fields from time-dependent currents (Ref. [4]).

Furthermore, with tight-binding nonadiabatic dynamics we have studied carrier transport properties and local polarization in halide perovskites. By including forces from time-dependent electronic states and machining-learning ionic potentials, we observed band transport behavior at high temperature (with carrier mobility in good agreement with experimental results) while the local polarization as a pancake structure at low temperature. Such behavior may well explain the physical observation of long electron-hole recombination lifetime in halide perovskites and the

ferroelectric relaxor behavior at low temperature observed experimentally (Ref. [5]). This work is currently under preparation.

Future Plans

Software development: In order to lower the barrier of adoption, a Python frontend and a simple command line interface are being developed. The Python interface is based on the ASE (Atomic Structure Environment) and provides an ASE-compatible "calculator" to integrate into Python workflows. Programmable request of data in the Python Materials Genomics format from the LBNL lead Materials Project (Ref. [3]) is being investigated.

Software validation: Full-time-dependent Spin Dynamics, including non-collinear and spin-orbit coupling, and nonlinear field driven phenomena including High harmonic generation and Floquet band engineering are two applications that are fundamental for experimental validation in these regimes. To this end, Lindenberg is carrying out high harmonic generation experiments in the topological semimetals WTe_2 and $MoTe_2$ and in a class of ferroelectric oxide superlattices driven by mid-infrared pulses in both a static and pump-probe mode.

Scientific research: We will continue the line of research in bulk photocurrents by performing real-time time-dependent density functional theory simulations of bulk photocurrents in real materials using the INQ code. These first-principles calculations will verify the above predictions made by tight-binding models and provide quantitative predictions which can be verified by experiments. The importance of optical phonon modes and the polaronic effect will be investigated with Ehrenfest TDDFT. Multiferroic oxides and charge-transfer organic salts are potential candidates for the realization of strong ballistic photocurrents. In multiferroic oxides, ferroelectricity originates from the underlying charge and orbital order that are strongly coupled to local atomic distortions, where the band gap is sensitive to these distortions.

With implementation of non-colinear spin and spin-orbit coupling to INQ, one will be able to simulate spin relaxation and dephasing, time-resolved Kerr rotation of semiconductors and magnetic systems. In semiconductors, spin polarization can be generated with circularly-polarized light in semiconductors, then relaxes and dephases due to spin-orbit, electron-electron, and electron-phonon couplings, which can be measured experimentally by time-dependent Kerr rotation. The calculated results by TDDFT can be compared to real-time density-matrix dynamics developed recently (Ref. [6]). In parallel, magnetic excitation (such as magnon) in AFM/FM systems will be simulated for magnon dispersion in 2D magnets (in collaboration with experimentalists in UW-Madison). By turning on electron-ion interaction, magnon-phonon coupling can be simulated which gives rise to the magnon line-width.

In addition, we will use INQ to model how low-dimensional materials, like graphene and transition metal dichalcogenides respond when driven with a strong time periodic partially coherent field. Preliminary real-time tight-binding calculations on graphene ribbons indicate that it is possible to

drive a topological phase transition from a semimetal to a chern-1 insulator even as the coherence time of the driving field becomes comparable to its period. We will reproduce these results using TDDFT as implemented in INQ to further bolster our confidence. Next using a combination of TDDFT and Ehrenfest dynamics, we will explore the possibility of realizing the same effect but with the external driving field replaced by internal phonon oscillations. Lindenberg is carrying out sub-gap photoexcitation experiments coupled to nonlinear optical probes as a means of testing these calculations.

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The Materials Project: A Decade of Materials Discovery

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Keywords: materials science, computational materials, high-throughput workflows, machine learning

Research Scope

Fueled by our abilities to compute materials properties and characteristics orders of magnitude faster than they can be measured, and recent advancements in machine learning, we are entering the era of the fourth paradigm of science: data-driven materials design. For over ten years The Materials Project [1] (www.materialsproject.org) has used supercomputing together with state-of-the-art quantum mechanical theory to compute the properties of all known inorganic materials and beyond, design novel materials and offer the data for free to the community together with open-source analysis and design algorithms. The current release contains data derived from quantum mechanical calculations for over 154,000 materials and millions of properties. The resource feeds a growing community of data-rich materials research, currently supporting over 450,000 registered users making millions of data records requests, and consuming tens of terabytes of data per month. From this data have come a number of material prediction success stories with novel compounds being synthesized and verified experimentally across a number of different application areas.

Recent Progress

In the last year, progress has been made in areas involving high-throughput workflows and screening, core data quality and expansion, machine learning, and automated synthesis. More specifically, high-throughput calculations and screening have been applied to generate new promising material and property predictions in areas including ferroelectrics and polar metals, battery material, transparent conductors, and charged defects, to name a few. Studies involving workflow creation have had their code integrated into the open-source packages like atomate or atomate2, with related analysis code integrated into pymatgen. With regards to core data, work has been done to both enhance its quality and quantity via the creation of a high-throughput framework for obtaining Hubbard U and Hund J values [2], as well calculation expansion using the r2SCAN meta-GGA functional. In total, hundreds of thousands of high-quality molecule and material calculations have been added to the core database, which provide new properties related to thermodynamics, bonding, charges, electronic structure, redox activity, spins, and vibrational modes [3]. This new data has also contributed to the success of a number of machine learning

related projects on material property prediction and interatomic potentials. More specifically, this includes the training of equivariant networks for elasticity tensor and chemical activation barrier prediction, as well as graph networks for the creation of more accurate interatomic potentials for use in molecular dynamics simulations. Lastly, synthesis related efforts have come in the form of data and software contributions to the A-lab project and calculation data integration in collaboration with the GnOME effort from Google Deepmind [4]. This has helped enable the automated synthesis of a number of new theoretical oxide materials which have been predicted thermodynamically stable.

Future Plans

Future plans involve both data, platform, and research efforts. For data, expansion of the core database is planned via increased property coverage using higher quality meta-GGA functionals, as well as machine learned interatomic potentials such as M3GNet and CHGNet. The latter also involves an effort to improve the overall quality of the models via generation of a standardized and comprehensive open-source reference training dataset. On the platform side, improvements to community contributions are planned via expansion of the MPContribs platform and portal. This will broaden the type and size of computational and experimental data that can be ingested by the Materials Project, in addition to making it more visible to the community. For research efforts, there are a few areas of focus including increased high-throughput DFT accuracy and capabilities, calculations beyond bulk stoichiometric compound space, and data-driven realization and validation of target solid-state inorganic materials. The first involves moving towards the development of high-throughput calculation capabilities in non-collinear magnetic ground states and finite-temperature phonons. This will be enabled through the application of novel agent-based optimization schemes and the compressive sensing sampling approach. For the second, workflows are planned that are able to capture tensorial properties of disordered multi-component solid solutions. For the last synthesis related focus, further integration of the Materials Project software and data into the A-lab's autonomous synthesis capabilities is planned. This will further accelerate the rate of experimental feedback on theoretical materials predictions made by other core team members and collaborators.

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Emergent properties of highly correlated electron materials

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Keywords: Quantum critical phenomena, quantum transport, unconventional superconductivity, quantum materials with disorder and interactions

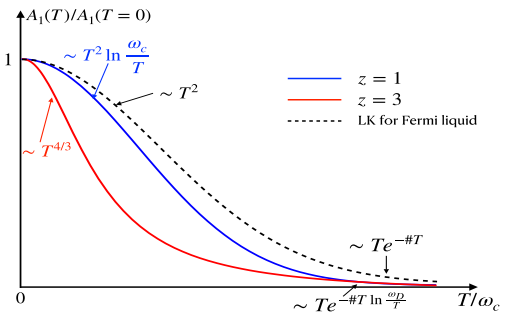
Research Scope

The main focus of the PI's current program has been on universal behavior of quantum matter stemming from the effects of topology, electron-electron interactions and quenched randomness. The PI has taken a multifarious approach, attempting to "triangulate" complex phenomena via 1) numerical studies of paradigmatic model Hamiltonians, 2) analytic studies of asymptotic behavior of quantum systems, and 3) phenomenology to tackle experimental observations head-on. Recent highlights include, 1) New predictions for quantum oscillations in three-dimensional quantum critical metals[2], 2) New pairing mechanisms for pair density wave superconductivity[1,5,6] and resulting predictions for model Hamiltonians as well as material systems, and 3) Phenomenology of reentrant superconductivity in heavy fermion systems such as UTe_2 [3,8].

Recent Progress

1) Magnetic oscillations in quantum critical metals

Magneto-oscillations are spectacular macroscopic quantum effects in which thermodynamic and transport quantities in metals oscillate in the presence of a magnetic field B. The period of oscillations is proportional to 1/B, and such quantum oscillations have been used for decades to understand shapes of Fermi surfaces in metals. The conventional theory of quantum oscillations is based on the seminal work of Lifshitz and Kosevich (LK), who analyzed quantum oscillations of a free Fermi gas. Extensions of LK theory by Luttinger included effects of Fermi liquid corrections but were otherwise still based on the idea of long-lived quasiparticles. What happens, then, when metals are tuned to quantum critical points (QCP) where the quasiparticle notion breaks down?



Temperature dependence of magnetization oscillations of a 3d marginal Fermi liquid with dynamical exponent z=1 and z=3, in comparison with the standard LK theory.

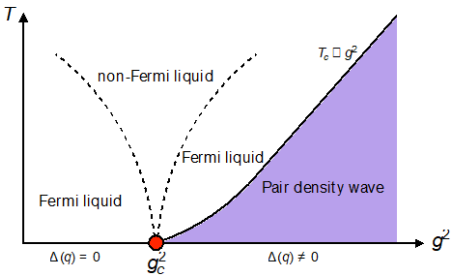
In recent work[2], we investigated this question. We showed that naïve extensions of LK theory to a three-dimensional quantum critical metal results in a *violation* of the third law of thermodynamics: the entropy density becomes finite in the limit of zero temperature. We showed that the violation occurs when the order parameter correlation length associated with the QCP exceeds the cyclotron radius. The breakdown of LK theory occurs because (i) fermion self-energies have non-analytic temperature dependence, (ii) fermion self-

energies themselves oscillate in a magnetic field and such oscillations, which are usually ignored in LK theory, and have dramatic consequences, and must be treated with care. Lastly, (iii) quantum oscillations are profoundly affected by non-trivial dynamical scaling laws that occur at the QCP[2].

We carefully considered these effects within the framework of Migdal-Eliashberg theory. Upon doing so, the laws of thermodynamics remain satisfied, and we obtained exact expressions for the entropy density at a quantum critical point. Finally, as a result of our work, we obtained *new predictions* for the amplitude of magnetization oscillations in a 3d quantum critical metal and showed dramatic difference in temperature dependence in comparison to the LK theory. Our theory has testable predictions that can be studied in 3d quantum critical metals such as $ZrZn_2$.

2) Robust mechanisms for pair density wave superconductivity

In certain exotic superconductors, Cooper pairs have finite center of mass momentum. Such superconductivity has been actively studied recently (where it is referred to as "Pair density wave (PDW) superconductivity") and is thought to play an important role in the phase diagram of the cuprate superconductors. In recent years, there has been growing evidence for PDW order occuring in Kagome metals such as CsV_3Sb_5, and in UTe_2. Despite such tantalizing experimental developments, there has been relatively little in the way of theoretical studies of pairing mechanisms for PDW order, because such superconductivity requires intermediate coupling, where relatively little in the way of controlled theory can be done.



Phase diagram consisting of Fermi liquid and PDW phases, as well as non-Fermi liquid behaivor near a PDW QCP.

Recently, we uncovered precisely such a robust mechanism for PDW formation[6]. We showed that if metals had sufficiently *repulsive* BCS interactions (i.e. pair tunneling), that are also *non-monotonic* in space, pair density wave order can occur. In our first study, we confronted the resulting intermediate coupling problem using large N saddle point techniques of statistical and quantum field theory. Subsequently, we showed how such resulting interactions occur naturally in Kagome metals when the Fermi level lies close to a van Hove singularity[5]. We have shown how such interactions naturally can occur in multi-band materials, where BCS interactions can be non-monotonic and repulsive as a consequence of form factors that arise when going from a local orbital basis to a momentum space band basis[1].

3) Phenomenology of reentrant superconductivity in UTe_2.

The heavy fermion superconductor UTe_2 hosts superconductivity that survives in excess of 60 Tesla, while the transition temperature itself is about 1.5 Kelvin. Why is superconductivity so robust in this system and how can we understand the complex phase diagram of this material?

One exotic aspect of the phase diagram of UTe_2 is the existence of reentrant behavior in a field: a magnetic field applied along certain directions enhances rather than suppresses superconductivity in this system. A tempting explanation for such behavior is that the system may possess a proximate quantum critical point, from which soft order parameter fluctuations enhance the tendency towards pairing. However, the proximate phase transitions in this system are strongly first order at low temperatures, which suggests that other mechanisms may be at play. In recent work, we suggested that there may be *multiple superconducting phases*, which give rise to reentrant behavior[8]. At the same time, we made our predictions, ongoing experiments revealed multiple superconducting phases in NMR and magnetocaloric studies. In more recent work, we used first-principles based microscopic models to study superconducting pairing symmetries in this material and have made concrete predictions that are roughly consistent with ongoing experimental studies[3].

Future Plans

1) Shot noise in strongly correlated metals

Many phases of condensed matter are defined by their electrical resistance. Recent experimental studies of correlated metals have involved non-equilibrium extensions wherein current noise at low temperatures in the presence of an applied voltage difference reveal the nature of charge carriers, as well as their extent in space: i.e. whether they flow through a wire in discrete classical "lumps" of charge or are more non-local, extended objects that are entangled with their surroundings. The PI is studying such shot noise, inspired by recent experiments on YbR_2hSi_2, and is studying a variety of systems which have strong interactions and quantum fluctuations, including metals near Kondo breakdown transitions, Luttinger liquids with extended interactions, and phenomenological descriptions of marginal Fermi liquids.

2) Fractionalized phases and emergent gauge fields

In a groundbreaking recent experiment, the fractionalized version of the celebrated quantum spin Hall effect has been discovered. The PI is actively working on understanding the behavior in the neighborhood of the observed phase, in particular on the role of screened interactions, disorder and exchange (Hubbard-like) interactions between the fractionalized degrees of freedom. Of particular interest is the nature of electromagnetic response near the observed fractional spin Hall effect phase and implications therein for novel forms of topological order producing this phase. The PI is investigating the role of emergent gauge fluctuations in this system and they way in which they affect the somewhat fragile topological order observed in experiments.

3) Phenomenology of superconductivity in infinite layer Nickelates and in UTe_2.

The PI has ongoing collaborations with his colleague Harold Hwang. Current projects include phenomenological studies of superfluid stiffness (penetration depth) in infinite layer

Nickelate superconductors to uncover possible order parameter symmetries, and the effects of strain on thin films of nickelate superconductors. The PI also has ongoing research projects on the nature of the high field "Lazarus superconductivity" of UTe_2 based on anomalous Hall effects coming from flat bands with berry curvature giving rise to robust superconductivity at extremely high fields.

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Advanced theoretical and computational methods for quantum materials

PI: F.A. Reboredo, co-PI K. Saritas, M. Eisenbach, M. Yoon, and J. Krogel.

Program Scope

In this project we focus on theoretical-computational studies using *ab initio* based approaches and high-performance supercomputers. The development of the theoretical/computational approach is key to achieve the accuracy required to predict new materials with competitive degrees of freedom. The overarching goal of this project is to understand and predict the electronic and magnetic properties of correlated and topological 2D layered materials and interfaces, and the effects of defects and dopants, using high-performance computing (HPC)-enabled many-body ab initio approaches to achieve the accuracy required. This goal will be pursued via the following three **Specific Aims.** (1) Understand electron correlation and magnetic effects in layered materials and interfaces. (2) Through understanding the underlying topological states, discover and design novel topological materials that have enhanced functionalities. (3) Develop and advance novel computational approaches that exploit HPC with targeted applications in quantum materials. We will develop and apply complementary many-body approaches based on quantum Monte Carlo techniques and density functional theory in combination with high-throughput, machine learning (ML), and artificial intelligence approaches. Computational methods based on ML will be developed to generate inexpensive surrogate models to predict the thermodynamic stability and finite temperature properties of novel quantum materials at energies and scales that are accessible through experimental synthesis and characterization approaches. This hierarchy of techniques will allow us to extend the accuracy of many-body approaches into increasingly less detailed methods that can reach larger sizes taking maximum advantage of current and future HPC assets to guide and assist the interpretation of experimental studies of quantum materials. Computationally assisted discovery methods based on machine learning will be developed to generate effective models and to predict novel quantum materials that are accessible through experimental synthesis and characterization approaches. This set of complementary techniques will allow us to have a comprehensive picture of materials systems across different energy and length scales involved in the experimental program supported by MSED BES DOE at ORNL.

Keywords: Magnetism, Topology and Correlation

Recent Progress

Existence of unobserved defects uncovered with DMC methods

The properties of $LaMO_3$ (M: 3d transition metal) perovskite crystals strongly depend on their point defects, whether introduced accidentally or intentionally. The most studied defects in La-based perovskites are the oxygen vacancies and doping impurities on the La and M sites. We identified [1] that intrinsic antisite defects, replacing La by the transition metal M, can be formed under M-rich and O-poor growth conditions, based on results of an accurate many-body ab initio approach. Our fixed-node diffusion Monte Carlo (FNDMC) calculations of $LaMO_3$ (M = Mn, Fe, and Co) **(Fig. 1**) find that such La-M antisite defects can have low formation energies and are magnetized. Complementary DFT-based calculations show that Mn antisite defects in $LaMnO_3$ may cause the p-type electronic conductivity. These features could affect spintronics, redox catalysis, and other broad applications. Our bulk validation studies establish that FNDMC reproduces the antiferromagnetic state of $LaMnO_3$, whereas DFT with Perdew–Burke–Ernzerhof

(PBE) strongly constrained and appropriately normed (SCAN) [128], and the local density approximation with Hubbard U (LDA+U) functionals all favor ferromagnetic states, are at variance with the experiments.

Antisite defects are difficult to observe in transmission electron microscopy because the presence of antisite defects would be masked by the La atoms on the same column of atoms on the surface of the samples. This is because the Mn, Fe, and Co sites have significantly fewer electrons (25, 26, and 27) than La (57). However, we found that antisite defects affect the electronic and magnetic properties of the perovskite host. Our partial density of states analyses showed that the antisite defects render the $LaMnO_3$ metallic, introducing holes and energy levels inside the band gap of $LaFeO_3$ and $LaCoO_3$. Mid-gap levels could be a signal that antisite defects have formed in experiments. Our FNDMC calculations also showed that the antisite defects have local magnetization, which, if measured, could confirm theoretical prediction.

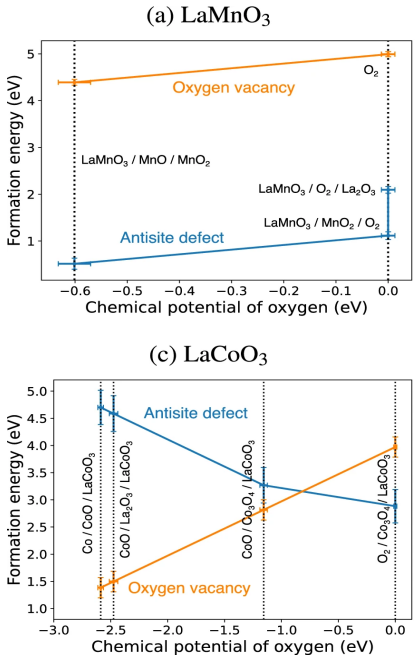

Figure 1 FNDMC prediction of the antisite defect and the oxygen vacancy formation energies as a function of O chemical potential for (a) (b) , and (c) . The vertical lines indicate the chemical potential where three compounds coexist.

Dynamical control of a topological material with an applied field

The core of our scientific contribution is the prediction of a strong coupling between topological and magnetic properties mediated by electric fields in $MnBi_2Te_4$ (MBT) when interfaced with ferroelectric materials. This finding is significant because it provides a new way to control the properties of quantum materials via heterointerface engineering, specifically in magnetic topological materials interfaced with ferroelectric materials. Its significance lies in potentially revolutionizing the way we understand and exploit interactions at material interfaces to dynamically control material properties.

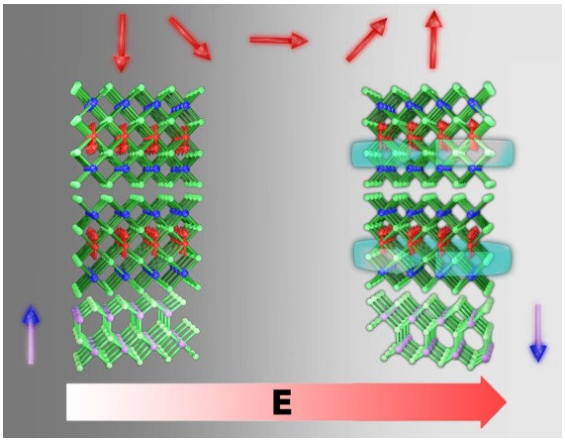

Figure 2 Nonvolatile control of magnetism and topology is predicted in an MBT/InSe heterostructure, in which the ferroelectric polarization switching induced by an applied electric field (E) can transition the system from an antiferromagnetic ground state (left) ferromagnetic (right) carrying topologically protected currents.

Building on our recent studies on MBT [2], we have performed a study of MBT heterostructures—materials that blend magnetism and topological properties [3]. The examples showing the importance of these interfaces are mentioned in Section 4.1. One of the hurdles with these materials is their antiferromagnetic ground state, which typically requires the use of high magnetic fields to adjust the spin orientation into a ferromagnetic

state, posing a challenge for practical applications. To solve this challenge, we designed a heterostructure that combines MBT with a 2D ferroelectric material. The innovation of our approach lies in exploiting the polarization of the ferroelectric substrate, which can be changed by an applied electric field, to dynamically control and induce a charge transfer at the interface (**Figure 2**). This technique can potentially stabilize the ferromagnetic state of the MBT and lead to topological phase transitions.

Our research builds on theoretical models and shows that reversing the polarization of the ferroelectric material could induce a topological phase transition in MBT. This approach could lead to a nonvolatile method for modulating these states in an MBT/InSe heterostructure, potentially switching between antiferromagnetic and ferromagnetic states. It is a promising step toward the development of new topological heterostructures with multifunctional capabilities. Our results contribute to a better understanding of quantum materials and open new avenues for future research and technological innovation.

Methodological advances in DMC

We are replacing the pseudopotential approach with more accurate pseudo-Hamiltonians, as proposed in our previous review document. In the pseudo-Hamiltonian approach, the kinetic energy is modified in addition to the potential. This additional degree of freedom allows the use of a local potential while still retaining the scattering properties of the all-electron atom. This strategy removes the source of locality error, which is one of the major sources of error in pseudopotential evaluation (**Fig. 3**). *We have successfully produced pseudo-Hamiltonians for transition metals from Cr to Zn [4,5].* We will use our new pseudo-Hamiltonian in our proposed work to improve accuracy.

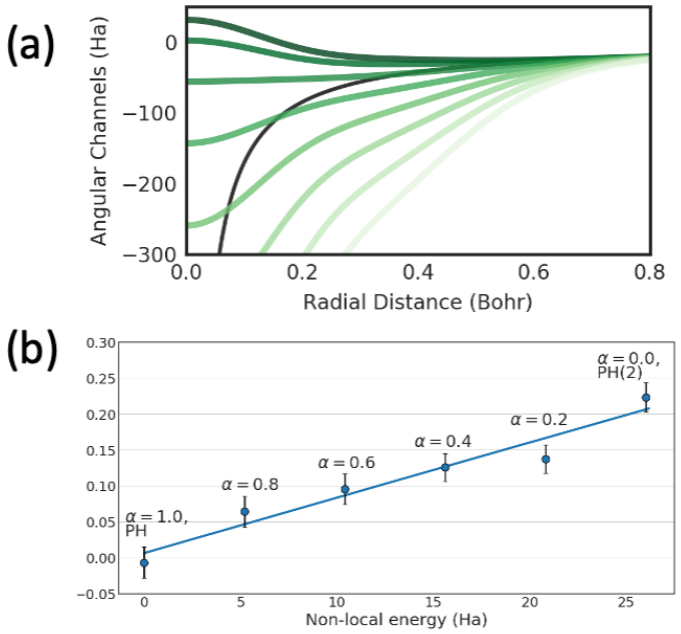

Figure 3 (a) Scattering channels of the quantum chemistry optimized pseudo-Hamiltonian for Co. (b) Elimination of DMC localization errors. The error vanishes, simultaneously with the non-local energy, as the Co core varies smoothly between a semi-local potential and the pseudo-Hamiltonian.

We uncovered an approach to obtain the excitation spectra and include missing correlations via an auxiliary bosons approach in DMC. Within the field of stochastic methods, the electronic excitation spectra have been evaluated primarily using VMC-related approaches.[6–10] VMC is known to miss correlations compared to DMC. We have theorized an approach that could introduce missing correlations in the excitation spectra via DMC [11]. Our approach has been tested in very simple models. We will continue this effort by implementing it for small ab initio systems in this funding cycle before a potential implementation in QMCPACK.

Future Plans: A few examples of current and immediate research are listed below. Please contact for discussions/collaborations.

- Fermi surface of La3Ni2O7 at high pressures (materials application of DMC and SHDMC)
- Oxygen deficiency in La3Ni2O7 (materials application of DMC)
- Spin/charge transitions in Co-containing materials ($LaCoO_3$ methods/theory for challenging magnets)
- Tunable Mott Insulators Li_xCoO2 (mott transition under (ordered) doping DMC and DFT)
- Energetics and properties of delafossites: (materials design: novel topo/etc ordered phases)
- QM materials: proximity effects/heterointerfaces/Moiré superlattices (experimental design or interpretation: topo/edge states in Moiré nanoflakes)
- Electronic instabilities in $HfFe_6Ge_6$-type 166 compounds (materials design or property search: CDW/topo states vs composition)
- Shandites and FeSn (explaining interfacial and surface reconstruction)
- Advanced wavefunctions for strongly correlated oxides and itinerant magnetic metals
    - SHDMC acceleration by noise filtering approaches
    - SHDMC Reference state optimization via singles expansions
- Calculations of excited states with DMC many-body corrections (expanded capabilities: spectra)
- Toward high-throughput quantum Monte Carlo methods

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Condensed Matter Theory

Alexei M. Tsvelik (lead PI), co-PIs: Robert Konik, Andreas Weichselbaum, Julia Wildeboer, Weiguo Yin, Brookhaven National Laboratory

Keywords: novel phases, anyons, machine learning

Research Scope

Our research in correlated and topological systems is aimed at establishing how simple and elegant fundamental principles of physics lead to the emergence of intricate complexity in the real world. In the Condensed Matter Theory FWP (CMT FWP) we address these questions with three principles in mind: i) To advance the boundaries of theoretical understanding of solid state quantum materials and statistical mechanical systems; ii) To provide insight and understanding of the experiments conducted by our colleagues in the Condensed Matter and Materials Science Division (CMPMSD); and iii) To connect to the research priorities articulated by the Department of Energy-Basic Energy Sciences (DOE-BES).

In regard to the first of these principles, advancing the state of the art of theoretical physics, we aim both to bring new insights into the physics of quantum materials and to advance methodologies than enable such insights. Notable examples of the former from the past three years include our work on Kondo-Kitaev models [R1] and order fractionalization therein, understanding mechanisms for topological phase transitions in $ZrTe_5$ [P1], and our insight that Majorana fermions exist in the $FeTe_{1x}Se_x$ family of iron-based superconducting materials [R3]. For the second principle governing our research, we strive to connect to the activities of our CMPMSD experimental colleagues. This is a strength of this FWP: out of 20 FWP publications in the past two years, 6 have involved collaborations with the experimental DOE-BES FWPs within the Division. Here, for example, we have lent theoretical understanding to non-equilibrium studies of Dirac semi-metals, bilayer iridates, the prototypical Mott insulator VO_2, the topological thermoelectric SnSe, and the 2D transition metal dichalchogenide 1T-TaSeTe.

For the third and final principle, we align our research interests with DOE BES priorities. Our overall work on quantum materials is informed by two BESAC reports, “Directing Matter and Energy: Five Challenges for Science and the Imagination” and the “Basic Research Needs Workshop on Quantum Materials”.

Bearing these three principles in mind, our research is organized around the following Focus Questions (FQ):

FQ1: What kinds of novel and exotic states of matter including topologically ordered ones can strong correlations generate?
FQ2: What new insights into quantum energy materials can be gained with inclusion of orbital degrees of freedom?
FQ3: How can placing a correlated system out-of-equilibrium lead to interesting non-thermal behavior?

FQ4: How do we find long-lived excitations in correlated quantum systems that may serve as the building blocks for quantum computation and information?
FQ5: How do we use machine learning to explore the properties of correlated and topological materials?

We discuss our recent progress and future planned work in terms of these focal questions.

Recent Progress on the Focus Questions

FQ1: Twisted bilayer graphene has a phase diagram similar to the one of the cuprates which has given rise to suggestions about similar physics. We use the topological heavy fermion (THF) model [R1] and its Kondo Lattice (KL) formulation to study the symmetric Kondo state in twisted bilayer graphene [P2,P3] . The conclusions are that near the integer filling where the Fermi surface is reduced to a point the physics is similar to the one of a ferromagnet.

FQ1: Work to extend Hamiltonian truncation methods to systems in two and three spatial dimensions is an active area of research [R4]. Here we have made progress by showing how Krylov methods can be combined with Hamiltonian truncation methods [P4] that naturally extend to higher dimensional systems.

FQ2: Ref. [P5] has demonstrated that a spin frustration can be induced in ferromagnets with nonuniform opposite Landé *g* factors such as the copper-iridium oxide Sr_3CuIrO_6. The frustrated state is characterized by a mutual interplay of typical ferromagnetic (FM) and antiferromagnetic (AFM) features, such as the zero-field susceptibility being FM-like at low temperatures but AFM-like at high temperatures. It is also found to contain an exotic zero-temperature "half-fire, half-ice" critical point at which the spins on one sublattice are fully disordered and on the other sublattice are fully ordered. We anticipate realization of the frustration and "partial fire, partial ice" states in certain antiferromagnets, lattice gas, and neuron systems. Such frustration could be used to drive novel phase transitions.

FQ3: Light-induced metal-insulator transition in the parent compounds of ruthenium pnictide superconductors [P6]. The phase diagram of ruthenium-pnictide superconductors $Ru_{1-x}Rh_xP$, $Ru_{1-x}Rh_xAs$, and RuSb is similar to that of the cuprates. We have recently completed a study on the interplay of the structural phase transition, flat electronic band dispersion, and metal-to-insulator transition (MIT) in the parent compounds of the Ru-pnictide superconductors using Density Functional Theory-based electron and phonon calculations. In this work, we examined the nature of pseudo-gapped monoclinic phases as well as predicted a light-induced two-step insulator-to-metal and orthorhombic to monoclinic structural transitions.

FQ4:[P7,P8] contain a description of symplectic chiral Kondo lattice which we suggest to use for quantum information processing. This suggestion is based on our previous result maintaining that symplectic Kondo impurities is quantum critical and contains non-Abelian anyon in the ground state.

FQ5: [P9] In [P9] machine learning techniques were applied to the so-called t−t′−t"−J model, where the determination of the parameter t′ in the context of cuprate superconductors is challenging. The self-consistent Born approximation was employed to generate a dataset of

about 1.3×10^5 spectral functions. The results suggest that it may be possible to use deep-learning methods to predict materials parameters from experimentally measured spectral functions.

Future Plans

FQ1: Study of the 3D CDW phase transition in kagome metals (J. Wildeboer and A. Tsvelik): This project is part of an effort to better understand the behavior of kagome metals. We plan to perform Monte Carlo simulations to determine the character of the CDW transition in this system. That such a transition is possible was suggested in analytical work performed by A. Tsvelik in conjunction with a postdoc, S. Sarkar [P10]. J. Wildeboer has implemented a Metropolis Monte Carlo simulation for a model Hamiltonian that describes the system of interest. Preliminary measurements suggest the existence of a (possibly first order) phase transition. Additional measurements are currently underway.

FQ2: Frustration-driven decoupling of the zigzag chains and emergence of spinons in layered trimer iridate $Ba_4Ir_3O_{10}$ (W. Yin, A. Weichselbaum, A. Tsvelik): This study looks for the origin of the exotic effective decoupling of the zigzag quantum spin chains in the honeycomb lattice.

FQ2: Orbital-assisted CDW and metal-insulator transition in the parent compounds of ruthenium pnictide superconductors (W. Yin, N. Aryal, A. Tsvelik): We will pin down the mechanism of the exotic hexamer formation in the insulating CDW state of Ru(P,As) and quantify the relative importance of electron-phonon and electron-electron interactions.

FQ3: Universality classes for transport in low dimensional spin chains and itinerant systems (R. Konik and A. Tsvelik): We will investigate how the nature of low energy interactions in low dimensional materials is connected to the presence and nature of ballistic transport in a material. We posit that these materials have two universality classes dependent on whether excitations can exchange their quantum numbers when they scatter off one another.

FQ4: NRG study of the symplectic Kondo effect (A. Weichselbaum and A. Tsvelik): This work is based on earlier work by A. Tsvelik *et.al.* [P7,P8] in which it was suggested that an anyon-based quantum computational scheme could be based on Kondo impurities (qubits) with symplectic symmetry. Here we are checking whether the scheme is robust with respect to devia-ons from symplectic symmetry breaking.

FQ4: Hunt for quantum many-body scars in real materials (J. Wildeboer, A. Weichselbaum, and R. Konik): In this work we are investigating the presence of quantum many-body scars in systems that harbor a special set of eigenstates that will result in a material not thermalizing in the standard fashion. We are particularly interested in the presence of scars in realistic models of low dimensional magnetic systems. J. Wildeboer has produced a working exact diagonalization (ED) code for multiple spin systems focusing on XXZ spin chains and ladders and two-dimensional lattices such as the square lattice.

FQ5: Search for potential material realizations of spin-unpolarized topological insulator systems able to realize the symplectic Kondo effect (N. Aryal and A. Tsvelik): In this project, we will use data mining and large language model-based artificial intelligence to scan the literature and material databases to look for possible material candidates that can realize the symplectic

Kondo effect. To realize this effect, we need ferromagnetic topological insulator systems with spin-unpolarized edge modes of Chern number 2 [P8].

FQ5: Deep neural network descriptions of the ground-state wavefunctions of frustrated Systems (W. Yin, A. Tsvelik, R. Konik, A. Weichselbaum, and J. Wildeboer): In this project we will develop the ability to represent wavefunctions of frustrated systems using deep neural networks. The training of such networks will be performed using variational Monte Carlo.

FQ5: Machine learning for efficient electronic structure calculations (N. Aryal and W. Yin): We are currently investigating a potential use of neural network for configurational averaging methods such as calculating temperature dependent band structure due to electron-phonon interaction. Our preliminary work has shown that a properly designed neural network can indeed reduce the computational cost involved by reducing the number of expensive first-principles calculations necessary to perform such configurational averaging. We plan to extend this methodology to study systems with defects and disorders.

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Ab initio Theory of Superconductivity

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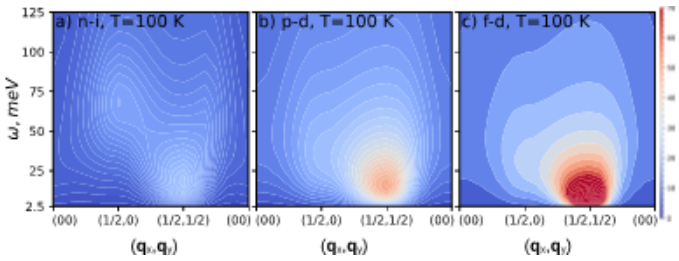
Keywords: Unconventional superconductivity, strong correlations, Green's functions, many body perturbation theory

Research Scope

We have formulated an *initio* Green's-function-framework, by combining self-consistent many-body perturbation theory with dynamical mean field theory that is effective in dealing with strong correlations. One- and two-particle Green's functions give both one-particle and two-particle properties, which can predict instabilities to broken symmetry states, such as antiferromagnetism or unconventional superconductivity (UCS). UCS are distinguished from conventional superconductors in that the electrons forming the superconducting state bind through many-electron interactions instead of through lattice vibrations. Yet the origins of UCS remain one of the outstanding unsolved problems in theoretical condensed matter physics today, perhaps the most important one. Our current research has a "methods" track, and an "applications "track to investigate some important UCS. On the methods side, we are implementing a field-theoretic description of the electron-phonon interaction. In this formulation, the theory adds a phonon contribution to the screened coulomb interaction *W*, instead of generating the usual diagrams to construct the self-energy directly. This puts all the bosons in one term, which makes it possible to formulate a theory of superconductivity that incorporates electron-phonon, charge and spin fluctuations in one description. There is an abundance of experimental evidence today that phonon play some role in the cuprates, the Fe superconductors, and many other ones.



FeSe: enhancement in low energy magnetic glue for superconductivity on intercalation. The vertex corrected dynamic and momentum resolved magnetic susceptibility $\mathrm{Im}\chi^{m}(\omega, \mathbf{q})$ is shown for (a) pristine FeSe (b) structure partially distorted by intercalation (c) structure fully distorted where the axial ratio is relaxed, $\mathrm{Im}\chi^{m}(\omega, \mathbf{q})$ becomes more intense at low energies, particularly at q = (1/2, 1/2) which corresponds to the anti-ferromagnetic instability vector in 2-Fe atom unit cell of FeSe.

On the applications side, we are investigating a variety of superconductors, described in **Recent Progress**. In all of them we shed new light on longstanding questions in a number of systems, made possible by the unique power of *ab initio* Green's function methods we developed.

Recent Progress

FeSe We have applied to Quasiparticle Self-Consistent GW theory combined with Dynamical Mean Field theory to pristine and intercalated FeSe. We elucidate the mechanism responsible for superconductivity in pristine FeSe (T_c = 9 K), and identify the vertex (the magnetic two-particle Green's function) as playing a central role. It is known T_c is enhanced almost universally

by a factor of ~4–5 when it is intercalated with alkali elements, but why T_c changes so dramatically is not known. Canonical approaches to date apply BCS or BEC theory (sometimes highlighting a "BCS/BES crossover"), in particular the density of states at E_F, $D(E_F)$. However, a theory based on one-particle properties utterly fails to explain what happens here: intercalation scarcely modifies $D(E_F)$. We show that the vertex is the primary driving force that enhances T_c. (In BCS theory a vertex enters implicitly through an undetermined coefficient, but it is not known and never calculated). Moreover, the widely use random phase approximation for χ^m (as an improvement to BCS theory) fails to capture the effect.

We show that there is one-to-one correspondence between the enhancement in magnetic instabilities at certain q vectors and superconducting pairing vertex, even while the nuclear spin relaxation rate $1/(T_1T)$ may not reflect this enhancement. Intercalation modifies electronic screening both in the plane and also between layers. We disentangle quantitatively how superconducting pairing vertex gains from each such changes in electronic screening. Intercalated FeSe provides an archetypal example of superconductivity where information derived from the single- particle electronic structure appears to be insufficient to account for the origins of superconductivity, even when they are computed including correlation effects. Another key finding of the theory is the usual neutron measurements used to assess the importance of spin fluctuations, do not apply as expected. superconductivity. We also find that the dxy orbital is by far the dominant one responsible for superconductivity. Nevertheless, the five-fold enhancement in T_c on intercalation is not sensitive to the exact position of this orbital at Γ point, provided it stays close to E_F. Finally, we show that intercalation significantly softens the collective charge excitations, suggesting the electron-phonon interaction could play some role in intercalated FeSe.

In a separate work, we benchmarked the spin susceptibility and the effect nematicity has on it. In the low-temperature rhombohedral phase, FeSe has a pronounced nematicity (asymmetry in the x- and y- directions), whose effect is seen in many experiments, including spin susceptibility. There has been much speculation that nematicity modifies the critical temperature; we show that even while it has a pronounced effect on many properties, its modification of T_c is small.

YFe_2Ge_2 has received much recent attention because susceptibility measurements suggest it has a near ferromagnetic instability, which makes it a good candidate to be a spin triplet superconductor. Within our Questaal framework we show that the system has a generic ferromagnetic instability which, however, gets strongly suppressed as the low spin state of the Fe starts to fluctuate strongly at low temperatures leading to singlet Kondo physics. Such strong competition between an instability towards a ferromagnetic order and Kondo physics was not explored in its full rigor in the literature. With QSGW+DMFT we can now describe many of its high and low temperature properties in excellent agreement with experiments performed by our collaborators (Dessau group at CU, Boulder). In addition to explaining spin susceptibility and the Kondo physics, we explain why YFe_2Ge_2 is a heavy fermion system with a d_{xz}+d_{yz} state lying at the Fermi level, having a very high mass ($m_{\text{eff}} \sim 25m_e$). The origin of the large mass---which drives many-body effects---is unique: it originates less from the usual "Hundness" picture and more from one-body hopping matrix elements as in twisted graphene.

UCoGe is a rare system, for which of ferromagnetism ($T_{\text{Curie}} = 2.7$ K) and superconductivity ($T_{sc} = 0.5$ K) coexist. This promises a unique situation for triplet superconductivity. Both Co and U have partially filled orbitals and are present at the Fermi energy. However, the bandwidths and the degree of coherence of the *d* and *f* states are rather different and they can lead to different

kinds of superconductivity. By employing our Questaal approach, in collaboration with the experimental group of Alix McColuum that performs high field quantum oscillation measurements, and computing vertex functions in magnetic and superconducting channels that include both the Co-d and U-f states, we have gained insights into the superconductivity and the fine balance between triplet and singlet nature of the superconductivity driven by the U and Co atoms respectively. A paper is nearly complete.

Rare-earth nickelates $RNiO_3$ (R=rare-earth element) exhibit a rich phase diagram that hosts, metal-insulator transition (MIT), magnetism, structural distortions and also superconductivity under certain perturbations. The ability to disentangle the coupled nature of the electronic, magnetic and structural transition is key to understanding the crucial parameters that control the interactions between the degrees of freedom that control the low temperature properties of these systems, including superconductivity. The body of literature has been muddled with opinions in absence of parameter free approaches that can compute the many-body interactions involved. With QSGW and DMFT we show that the d^7 valence of Ni comes with spin fluctuations and large orbital moments that can only get quenched if the system lowers its crystalline symmetry by coupling to breathing modes of the lattice. Such reliable determination of the electronic structure, gives us reasonable confidence that we are poised to address the superconductivity in Sr doped (Nd,Pr)NiO_3 and NdNiO3/$SrIrO_3$ heterostructures. A paper is nearly complete.

$TiSe_2$ has received much attention in part because it becomes superconducting when doped with Cu, and also because there has been longstanding wide speculation that it is an excitonic insulator. Excitonic insulators are much sought after materials with the promises for strong coupling between electrons and holes that can gap out the Fermi surface and drive a metal-insulator phase transition. By combining our self-consistent many-body perturbative approach with *ab-initio* molecular dynamics calculations, we are able to show that the strong lattice fluctuations drive the metal-insulator transition are not driven by excitonic correlations, but results from generalized Umklapp processes. It is the first instance we are aware of where a metal-insulator transition occurs as a consequence of dynamical nuclear motion. An initial paper has been put on arxiv and a final paper is essentially complete, including a new Brillouin zone unfolding scheme that elucidates the nature of the scattering processes.

JMRAM can form the basis of memory (JRMAM) possibly with orders of magnitude faster switching and lower power than conventional MRAM --- if materials issues can be solved [A]. Imperfections vitiate the effect in part, but also the predictions are based on free electron models and simplified models of scattering, which shaped how JRAM was understood. We have performed the first (an only so far) *ab initio* calculation of an MJJ [B], combining the Landauer-Buttiker formalism in density functional theory and the Bogoliubov-de Gennes model. We studied the Nb/Ni/Nb junction for J-MRAM. By studying the role of ferromagnet thickness, magnetization, and crystal orientation we show how the supercurrent decays exponentially with thickness and identify two mechanisms responsible for the effect that lay outside the scope of prior models.

Future Plans

Methodologically, our wider aim to implement a general-purpose, high-fidelity *ab initio* theory of superconductivity that does not rely on models, builds on the considerable successes we have had while also encountering limitations as noted above. The most important methods advance for us is the unification of all important bosons in one theory. This is necessary to understand

unconventional superconductors where it appears the electron-phonon interaction plays some role, of which there appear to be many.

We will continue studies of cuprates, where charge-density waves are adjacent to superconducting phases, which makes it likely the phonons play a role in superconductivity. What drives Tc in cuprates is poorly understood [2]; for example why *n*-layer cuprate superconductors made of sandwich structures have higher Tc than found in the tri-layer compound. Particularly interesting are **one-dimensional cuprates.** where the present paradigm predicated on separation of spin and charge into distinct collective excitations, for which we believe there is a more conventional explanation, which emerges with a high enough fidelity theory.

We will also study Fe(Se,Te), a materials system with a remarkable confluence of a Dirac point, surface ferromagnetism, and superconductivity [3].

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Exploratory Development of Theoretical Methods

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Keywords: Correlated Materials; First-principles methods; Machine learning; Materials discovery, Magnetism

Research Scope

The scope of this FWP is to develop new theories and computational methods to solve challenging basic science problems to significantly advance the mission of Department of Energy's Basic Energy Science and Ames National Laboratory. We focus on developing computationally efficient *ab initio* theories and computational methods for accurate calculation of correlated-electron materials. These methods aim to treat the energetic stability, electronic structure, and magnetism in an equal footing for correlated materials containing both localized and itinerant electrons so that fundamental physics and chemistry of rare earth (RE) materials can be correctly described and predicted. We also integrate machine learning (ML) and data science with the state-of-the-art materials modeling and simulation algorithms to develop robust ML-guided feedback frameworks to dramatically accelerate the design and discovery of novel materials with desirable structures and functionalities for energy applications.

Recent Progress

Ab initio CMRT method for correlated electron materials. Based on Gutzwiller many-body wavefunction, we developed an *ab initio* correlation matrix renormalization theory (CMRT) and computational code for accurate and efficient calculations of realistic strongly correlated materials. We showed that while the CMRT has the accuracy like that of LDA+DMFT, it is much faster with the speed of minimal-basis Hartree-Fock calculation. No choice of U and J parameters is needed and no double counting issues in CMRT. In addition to correctly predicting the volume collapse and related valence collapse physics in fcc Ce, CMRT also describes the behaviors of fcc Pr in good agreement with LDA+DMFT calculations and experiment [1], including the energy and pressure verses volume curves as well as the electron occupations in different 4*f* states as shown in **Fig. 1**. Moreover, spin-polarized CMRT formulism and computational code are also developed and tested for the ferromagnetic state of bulk bcc iron [2]. We showed that the CMRT describes the equilibrium physical properties and the pressure-volume curve in a very good agreement with experiments. The variation of magnetization with pressures in bcc Fe predicted by CMRT is also much more accurate than those from DFT-type calculations. The spin polarized CMRT is also being applied to

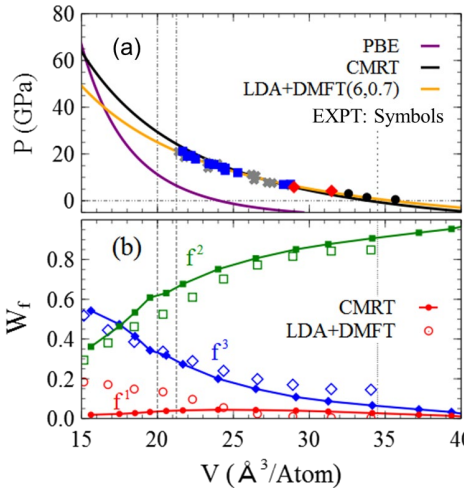

Fig. 1 CMRT calculation on fcc Pr is compared with GGA, DFT+DMFT, and experiments (EXPT). (a) Volume-Pressure (V-P) curves. (b) Occupation weights of different *f* states.

magnetic phases of rare earth materials. Preliminary results indicate that CMRT gives a better description of spin-splitting in Gd metal in comparison with other available *ab initio* methods. Our achievement opens the door for accurate and efficient prediction of strongly correlated materials.

Machine learning (ML) assisted materials discovery. We developed a ML framework for novel materials discovery as illustrated in **Fig. 2**. This ML assisted framework effectively integrates a crystal graph convolutional neural network (CGCNN) ML model and materials databases with adaptive genetic algorithm (AGA) and *ab initio* calculations, as well as experimental validation/feedback for efficient materials discovery. We demonstrated that the ML assisted framework enables 100-1000 times acceleration in ternary and quaternary materials discovery in comparison with conventional approaches [3,4]. As an essential part of our ML-guided framework, we also developed neural-network ML interatomic potentials for complex binary and ternary systems which enable reliable molecular dynamics simulation and analysis to address the important issues in materials discovery and synthesis, such as temperature and dynamics effects in the phase stability and phase selection/transformation. Complex phases with 3 or more chemical elements are a vast untapped potential for new materials. Our integrated ML approach is applicable to a wide range of complex compounds of interest, offering a new paradigm to enable dramatic acceleration in the discovery of novel materials.

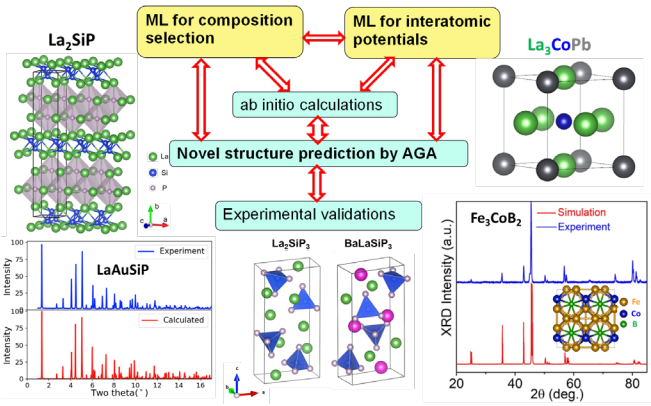

Fig. 2 Our ML-guided framework has been demonstrated to accelerate the novel materials discovery by 100-1000 times in comparison with conventional computational approaches.

Prediction of coexistence of low/high spin states in a complex La-Co-Pb compound. We performed *ab initio* calculations to study the electronic structure and magnetic properties of a newly predicted ternary compound $La_{18}Co_{28}Pb_3$ from our ML approach. This compound is complex with 98 atoms per unit cell and contains an immiscible pair of Co and Pb elements. As shown in **Fig. 3**, a peculiar localized-itinerant transformation from a high-spin ferromagnetic (FM) phase to a low-spin FM phase and the appearance of other competing metastable antiferromagnetic (AFM) magnetic phases are observed. We showed that this complex ternary compound would exhibit strong spin fluctuations and a quantum criticality because of the competing magnetic states, leading to magnetic instability and magnetic phase transitions under marginal pressure (or volume) changes.

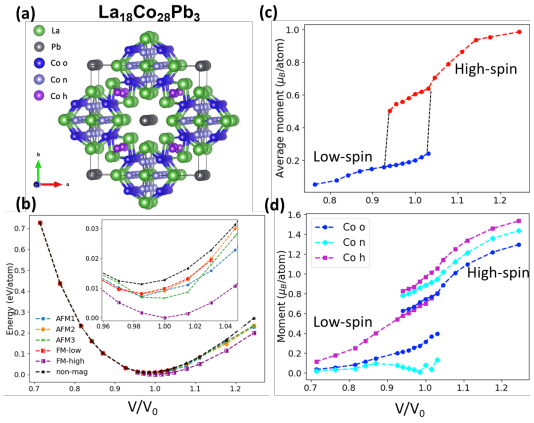

Fig. 3 (a) The structure of $La_{18}Co_{28}Pb_3$. (b) The total energy vs volume for various magnetic states. (c) The averaged magnetic moment per Co atom and (d) The magnetic moments for the non-equivalent Co atoms at different volume.

Future Plans

We will further develop *ab initio* theories and computational methods to describe correlated-electron materials, especially those containing 4f electrons, with desired accuracy and fast computational speed. In addition to spin polarization, spin-orbit coupling will be developed and implemented in CMRT so that complex materials with interplay between itinerant and localized electrons can be accurately described to greatly enhance the predictive power of *ab initio* calculations for correlated materials. We will also develop other methods for calculating the electronic structure in the localized (*4f* electron), itinerant, and mixed magnets. These methods will include a reliable description of crystal field splitting in RE systems with *3d* atoms. One fundamental challenge in treating RE containing systems is that an accurate quantum-chemical description of the RE atom is highly demanding, requiring a large active space. We plan to develop an effective atomic Hamiltonian for RE atoms within minimal basis by fitting it to the well-established experimental atomic spectra. The effective atomic Hamiltonian will then be incorporated through embedded theory to study the interaction of the localized *4f* states with the environment in RE-containing systems.

We will further develop AI/ML-guided methods for efficient discovery and synthesis of novel functional materials. Our current ML-guided framework will be extended to less explored and more complex quaternary compounds. AI/ML models will also be developed to provide accurate and efficient screening and prediction of materials properties. Currently, many computationally predicted compounds cannot be synthesized, largely due to lack of knowledge of the necessary synthetic pathways. We will address this challenge by developing accurate and efficient neural network ML interatomic potentials for reliable molecular dynamics simulations to timely acquire thermodynamics data and phase selection/nucleation kinetics to understand the local energy landscape between transient or intermediate products and suggest synthesis pathways to guide precision synthesis. The ML interatomic potential development will also be integrated with adaptive genetic algorithm (AGA) for efficient new crystal structure prediction in our ML-guided framework. Our ML-guided framework with feedback from experimental validation will greatly accelerate materials design, discovery, and synthesis to meet the mission of DOE and the strategic goals of Ames National Laboratory.

We will improve the state-of-the-art *ab initio* methods by effectively incorporating the many-body perturbations (e.g., using developed dynamic RPA to add spin fluctuations) to correctly describe the interaction of localized and itinerant electrons in magnetic systems containing *3d* and/or *4f* electrons. We will perform such *ab initio* calculations to reveal complex material physics governed by delicate interplays among electron correlation, spin fluctuation, and magnetism, especially in the correlated materials containing *4f* electrons. We will study the different magnetic interaction parameters in the localized (*4f* electron), itinerant, and mixed magnets. A fundamental understanding of microscopic interactions is vital for understanding these materials' properties,

formulating simulation models at finite temperatures, and accelerating the design and discovery of novel and/or optimization of existing materials for energy-relevant applications.

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University Abstracts

Atomic to mesoscale models of phase transitions for energy materials

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Sabrina Liwen Wan, Tae Wook Heo & Marissa Wood – Lawrence Livermore National Laboratory

Keywords: multiscale simulations, polymorphic phase transition, machine learning

Research Scope

Numerous energy production and storage technologies rely on materials with many metastable phases (polymorphic); often one phase is more effective than others for a given application. These polymorphic phases can coexist and undergo phase transformations during synthesis or device operation since their energies are relatively similar, unlike for example graphite and diamond. Transition metal (TM) oxides with nickel, manganese, and cobalt are promising polymorphic materials for catalysts, battery electrodes, and supercapacitors. However, their long-term performance and efficiency are impacted by phase transitions between their competing polymorphic phases. Extensive work has been done to address the thermodynamic differences between the polymorphic phases. In application, kinetics is also a primary driving force for any undesired phase transformations, which is far less understood. Our simulations are designed to determine the kinetics of phase transitions and phase nucleation and growth, focusing on MnO_2, since significant simulations of its thermodynamics have already been performed.

Multiscale simulations that employ three cutting-edge computational techniques will bridge the quantum, atomic, and mesoscales to provide an innovative and comprehensive model of polymorphic MnO_2. Quantum and atomic scale simulations will determine phase transition pathways, transition states, and energy barriers. A phase-field method will provide microstructure-level mesoscale simulations to determine phase evolution. Machine learning force fields (MLFF) will be developed to perform meta-dynamics simulations that will determine nucleation and growth parameters. The MLFF will also be used to simulate interfacial energies and elastic properties for the mesoscale model. MnO_2 synthesis and characterization using X-ray diffraction and SEM by Lawrence Livermore National Laboratory (LLNL) will inform the microstructural models and can provide experimental validation of the simulations.

Current work focuses on training the MLFF using quantum simulations of the interfaces between various MnO_2 phases and molecular dynamics of phases at temperature. We test if including small interfaces in the MLFF training set improves optimization of larger interfaces. The MLFF development will use the most rigorous methods for training and testing the potentials so that they can be trusted to accurately simulate the complex phase transitions between polymorphic TM oxides. MLFF meta-dynamics simulations of nucleation and growth will provide insights into the phase transformations of the technologically important MnO_2. In addition, these simulations will help identify phase transition pathways, which will inform the set-up quantum calculations of

energy barriers and transitions states. Our simulations will be one of the few studies that tackles solid-solid nucleation and growth of complex TM oxides or the mesoscale phase evolution of polymorphic materials.

Recent Progress

We have generated MLFF training data by running molecular dynamics simulations of each individual phase (α, β, γ, δ, ε, λ, R) at various temperatures. Since we need the MLFF to simulate nucleation and growth, we generated interfaces between the phases for input into the MLFF (Figure 1). A Python code is being developed to generate and optimize these interfaces. Interfaces with high and low strain/energy are required to train the MLFF.

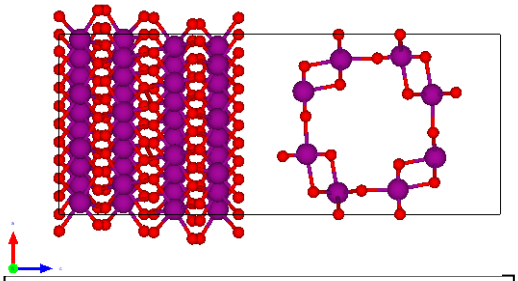

Figure 1. This interface of R-α is low strain and was constructed by-hand.

The procedure to train the MLFF is being tested with phase of MnO_2 and Li-MnO_2 and uses the Multi Atomic Cluster Expansion (MACES) code[1,2]. Our tests have shown that Li- MnO_2 potentials training by this code is fast and accurate. We have tested the potential against the speed of two other codes that employ neutral networks: n2p2[3] and SIMPLE-NN[4].

Whether or not to include spin-polarized calculations is determined by looking at the spread in energy of different spin configurations (both ferromagnetic and anti-ferromagnetic) for the individual phases and the interfaces. The fidelity of the force field is tested on larger interface (around 1000 atoms) optimization. The size of the interface is set by reasonable simulation times using Density Functional Theory codes.

At San Francisco State, the PI has started student networking efforts to improve mentoring and support our diverse student researchers. She established a LinkedIn page and advertised student research positions on the page.

Future Plans

Once the MLFF is trained to handle phase transitions, the PI will run nucleation and growth simulations, where a seed of one phase is inserted into another phase and the structure evolves at various temperatures. If a phase transition occurs, the atomic-scale pathway will be used to set up calculations of energy barriers and transition states.

Over the summer, the PI will invite LLNL scientists to visit students at SFSU and give talks about their research. If a suitable student is identified, this student will start synthesis of MnO_2 phases over the summer at LLNL. The collaboration with LLNL will advance the PI's computational expertise and capacity to train undergraduate and master's students at San Francisco State University, a Hispanic Serving Institution. LLNL will actively mentor three students in both

simulations and experiment and support efforts to increase diversity and inclusion in the workforce training of under-represented groups in the sciences.

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Publications

N/A

Superconductivity in quantum materials

Daniel Agterberg, University of Wisconsin-Milwaukee

Keywords: Correlated superconductors, Bogoliubov Fermi surfaces, nodal degeneracies, spin-orbit coupling

Research Scope

This project addresses unconventional superconductivity of quantum materials. It does so by solving effective theories of correlated fermions guided by experimental results and by collaboration with density functional theory (DFT) experts. A key emphasis is to move beyond the commonly used single-band picture to include the spin, orbital, valley, sublattice, and non-symmorphic degenerate electronic degrees of freedom needed to properly describe quantum materials. This project focuses on the role of these electronic degrees of freedom on the physics of unconventional superconducting states. Current research emphasizes three topics: i) the interplay between non-symmorphic symmetries and superconductivity with an application to $CeRh_2As_2$; ii) magnetic response of superconductors with strong spin-orbit coupling with an application to the odd-parity superconductor UTe_2; and iii) magnetically driven topological and non-topological Bogoliubov Fermi surfaces with an application to $FeSe_{1-x}S_x$ superconductors.

Recent Progress

Non-symmorphic symmetries and superconductivity: Our earlier collaborative experimental and theoretical research on $CeRh_2As_2$ [1] revealed a c-axis field-induced superconducting transition between an even and odd-parity (possibly topological) superconducting state. Key to the theory of this transition are the two Ce atoms in each unit cell. These two non-symmophic-symmetry required Ce atoms form two sublattice degrees of freedom that are related by inversion symmetry. Even and odd-parity superconducting states correspond to in-phase and out of phase spin-singlet pairing on these two sets of Ce atoms. We have shown that the non-symmorphic space group ensures that the electronic spin-orbit coupling (SOC) is the dominant single particle interaction for electronic Fermi surfaces near the Brillouin zone boundary. This is the case in $CeRh_2As_2$ and this enhanced SOC in turn stabilizes the odd-parity state [2]. In addition, as shown in Fig. 1, we have shown that our theory can quantitatively explain the field induced even to odd-parity transitions for fields tilted from the c-axis, strengthening the interpretation of a field induced even to odd-parity transition [3].

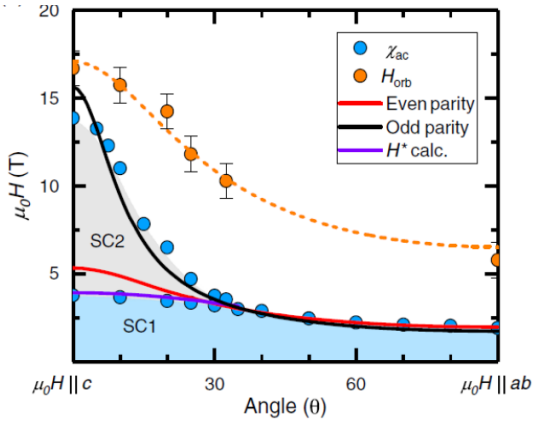

Fig. 1 Field-angle dependence of the even (SC1)- and odd (SC2) -parity superconducting phases at 45mK. H_{orb} is the orbital limiting field. From [3].

Underlying the theory of the odd-parity superconducting state for $CeRh_2As_2$ is the non-symmorphic space group (P4/nmm) of the crystal. This motivated us to ask more generally if non-symmorphic space groups can provide a generic means to stabilize odd-parity superconducting states. They can, provided the Fermi surfaces that dominate the superconducting state lie near the Brillouin zone boundary [4]. We found that when a non-zero SOC splits non-symmorphic band degeneracies at the zone boundary, the resultant pseudospin has different symmetry properties than usual spin ½, implying that the magnetic response and Cooper-pair structure of superconductors formed from this pseudospin will be unusual. We have classified the non-symmorphic space groups that exhibit this anomalous pseudospin, analyzed the resultant unusual superconducting states, and identified a series of candidate materials [4]. A brief synopsis is given in Fig. 2.

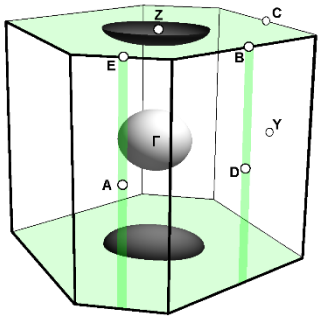

Fig. 2 Non-symmorphic band degeneracies, nodal planes and nodal lines (green), that exist without SOC. Fermi surfaces near nodal planes exhibit unusual superconducting properties including: field-induced odd-parity states, large critical fields, and pair density wave transitions [4].

More recently we have examined general consequences of electronic interactions on electronic ground states derived from non-symmorphic sublattice degrees of freedom [5]. We have examined 2D space groups that enable co-incident Van-Hove singularities, two Van-Hove points that occur at the same energy and momentum. We have classified the symmetry allowed interactions near these coincident Van-Hove points for eight 2D space groups. We have examined how these interactions are enhanced by both the Van-Hove divergent density of states and by the divergent intra-band superconducting susceptibility. Surprisingly, we find both superconducting and non-superconducting states [5] are stable ground states. The appearance of non-superconducting states stabilized by the intra-band superconducting susceptibility is not found in related so-called patch models and provides a non-nesting based weak-coupling mechanism to stabilize such states.

Magnetic response of superconductors with strong spin-orbit coupling: The role of SOC on the magnetic response of even and odd-parity superconductors has been poorly examined and yet plays an important role in interpreting experiments. In Ref. [6], we have carried out a general analysis of this problem that shows that usual expectations for magnetic response can be qualitatively incorrect for odd-parity superconductors and further reveals two quantities are sufficient to determine this response. The first quantity is the well-known effective *g*-factor and the second is the field-fitness function. For even parity superconductors, the field-fitness function is always one, so only the effective *g*-factor can play a role in magnetic response. For odd-parity superconductors, the field-fitness function can lead to counterintuitive results, as we have shown for j=3/2 materials [6]. In collaboration with the research group of Prof. Raghu at Stanford University, we have applied our results to odd-parity superconductivity in UTe_2 [7].

Magnetically driven topological and non-topological Bogoliubov Fermi surfaces. Our original work on Bogoliubov Fermi surfaces revealed that when time-reversal symmetry is broken in even-parity superconductors, traditionally anticipated point or lines nodes do not exist [8]. The quasi-particle excitation spectrum is either fully gapped or has topologically protected Bogoliubov Fermi surfaces. Underlying this result was the realization that symmetry requires the appearance of momentum varying magnetic fields when time-reversal is broken and that these fields can be created intrinsically by superconductivity. Recently we have examined this question more generally [9,10] by classifying the different ways time-reversal symmetry can be broken. We have found that in addition to even-parity broken time-reversal symmetry, odd-parity broken time-reversal symmetry can lead to Bogoliubov Fermi surfaces. In this case the Bogoliubov Fermi surfaces are not topologically protected. Furthermore, we have shown that for some odd-parity broken time-reversal orders, the superconductor is required to become a pair density wave superconductor. We have applied this to $FeSe_{1-x}S_x$ and have shown that checkerboard antiferromagnetic order corresponds to an odd-parity broken time-reversal magnetic order that gives rise to PDW superconductivity and naturally accounts for the observation of nematic Bogoliubov Fermi surfaces [11].

Future Plans

In the final year of this grant, in addition to continuing our theoretical work on odd-parity superconductivity in $CeRh_2As_2$, we plan to develop a DFT approach to calculate the Pauli limiting field in even parity superconductors with strong SOC. This approach will be based on our general results for the magnetic response of superconductors [6]. We will apply this approach to BiS_2-based superconductors and to Ti_4Ir_2O. In addition, we plan to extend our work on superconducting symmetries in 2D [12] and the relationship of these to the appearance of nodal excitations including Bogoliubov Fermi surfaces. Finally, based on our symmetry-based explanation of nematic Bogoliubov Fermi surfaces in $FeSe_{1-x}S_x$, we will develop a more microscopic understanding of the interplay between superconductivity, checkerboard magnetic order, and Bogoliubov surfaces in FeSe-based superconductors.

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HBCU Undergraduate Program toward ab initio Prediction of Single-Photon-Emitters and Spin Qubits in Defected 2D Semiconductors

Marisol Alcàntara Ortigoza and Sergey Stolbov

Keywords: Spin-Qubits, Single-Photon Emitters, electronic structure, optical excitation, triplet

Research Scope

Spin Qubits: Defect-based optically controllable spin qubits may serve as the building blocks of quantum information technology. We introduce an avenue to rationally design local defects in semiconductors with spin-qubit characteristics and to reveal their optical spin-polarization cycle. By applying this methodology, we predict a new defect, a sulfur-vacancy defect in AlN, whose optical properties are more favorable for qubit functionality than those of the NV- center in diamond.

Single Photon Emitters: Single photon emitters (SPE) in the near-infrared (NIR) range are important elements of the quantum communication technology. In order to reveal efficient NIR SPEs, we rationally preselected substitutional-doping defects in SnS_2. We found that the Si_S, P_S, and N_S defects have sharp excitation peaks in the NIR region. Our calculations indicate that the optical emission associated with these excitations will have narrow zero-phonon lines with negligible phonon side bands, which makes them excellent NIR SPE candidates.

Recent Progress

SPIN QUBITS

The optical readout and manipulation of a spin qubit is a complex process requiring a combination of several specific properties of the defect. It is thus not surprising that for the first and most studied spin qubit defect – the negatively charged nitrogen-vacancy center in diamond (NV^- center) – it took about two decades to detect its charged nature, to demonstrate a feasible preparation-readout qubit cycle, to describe its electronic structure, and to formulate a consistent theoretical description of the entire cycle process. [1, 2]

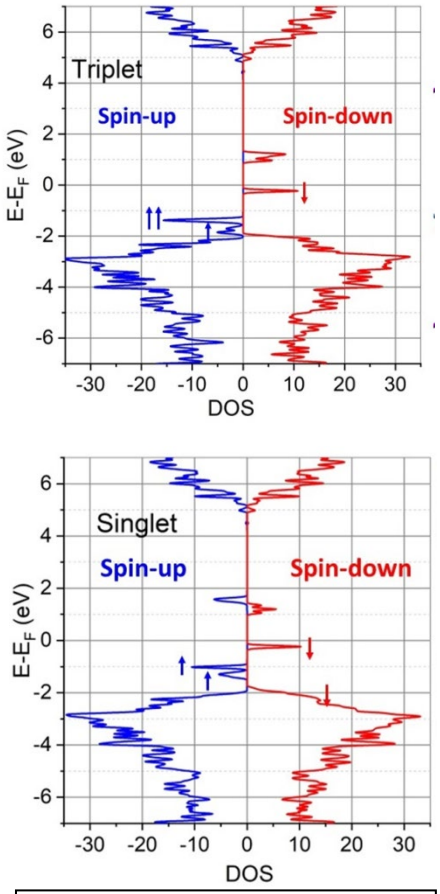
Based on our rational methodology, we selected the neutral defect in wAlN and evaluated its spin qubit functionality from first principles. This defect combines an Al vacancy and a S atom substituting the N next to the vacancy ($V_{Al}S_N$). Our design is based on the following reasoning: Removal of an Al atom decreases the even number of *p*-electrons by one, while the replacement of N with S brings back one electron making the number of *p*-electrons even again, which is necessary for triplet formation. Also, the relatively low electronegativity of S hints that it will not bind as strongly as O, possibly shifting the defect states to the band gap. Moreover, the vacancy creates three N dangling bonds, which is beneficial for spin polarization. We also expected that this defect will have C_{3v} symmetry as the NV^- center does, which is favorable for the spin-

polarization cycle. Our calculations revealed that the structure does have C_{3v} symmetry, is thermodynamically stable, and has a triplet spin state.

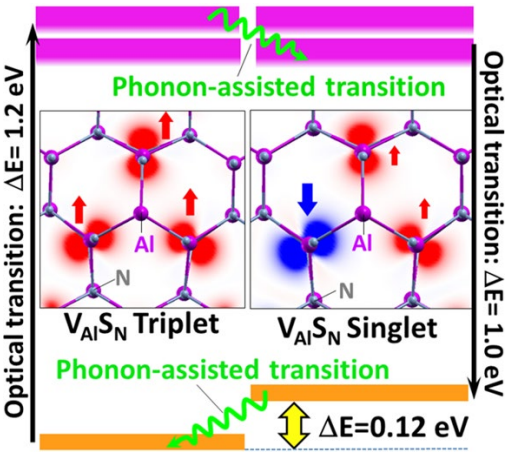
The GW electronic structure of the $V_{Al}S_N$ defect in the triplet state is found to be promising for spin qubit functionality. Indeed, the four electrons determining the triplet ground state (TGS) in $V_{Al}S_N$ occupy narrow GW independent-quasiparticle (IQP) peaks located in the band gap. The spin density is evenly distributed among three dangling bonds associated with the three N atoms next to the vacancy, yielding a total magnetization per supercell of 2.0 μ_B. Using the Bether-Salpeter equation method we calculate the optical excitations of the triplet states of the $V_{Al}S_N$ defect in wAlN. The optical excitations correspond to numerous BSE eigenstates grouped between 1.0 and 1.2 eV.

The next step is to find a singlet state which results from the structural transformation the triplet excited state (TES) undergoes. To locate the singlet state, we designed the following method: First, we evaluated the changes in electron charge density caused by the optical excitation in the triplet. To mimic the possible lattice reaction to the charge redistribution, we assumed that a decrease in the the local valence charges within the Wigner-Seitz spheres (QWS) of the N atoms causes a decrease in bond length between the corresponding N atom and its neighboring Al atoms, while an increase in QWS induces an increase in the corresponding N-Al bond lengths. Our approach resulted in a thermodynamically stable structure that has a singlet spin state.

Since the energy difference between TES and SGS is large (~0.9 – ~1.1 eV) compared to the phonon energies of the system, a direct nonradiative phonon-induced transition from the former to the latter is unlikely. Thus, some intermediate singlet excited state (SES) may be needed. To find such a state, we calculated the optical excitation spectrum for the singlet using the BSE method. The spectrum is divided in two distinct energy groups. The lower energy excitation group is between 0.9 – 1.4 eV and all corresponding excitations originate from the spin-down states. It may serve as an intermediate state in the path of the spin-selective decay of the spin-polarization



Densities of the IQP states calculated for the $V_{Al}S_N$ defect in the triplet (upper) and singlet (lower) ground states using the self-consistent GW method.



Spin density (red and blue lobes) and spin-polarization cycle energy diagram obtained for $V_{Al}S_N$.

cycle. Remarkably, the oscillator strength for the singlet excitation is practically the same as that for the triplet, which may be critical for an efficient spin-polarization cycle.

We proved that the energy barrier for the transition from the SGS to TGS is not higher than 0.15 eV, which makes the transition feasible because such barrier can be easily overcome via phonon excitations. Finally, we built the energy diagram for the entire optical spin-polarization cycle of the $V_{Al}S_N$ defect in wAlN. The cycle is very similar to that of the NV^- center: an optical excitation from the TGS to TES changes the charge distribution and reduces the symmetry of the system, which leads to a presumably spin-selective phonon-assisted transition from the TES to SES. Next, an optical emission brings the system to SGS, followed by a nonradiative phonon-assisted transition back to TGS. Importantly, our BSE calculation of the oscillator strength of the optical transition between the singlet ground and excited states suggests that this step for the $V_{Al}S_N$ defect is optically more favorable for the spin-polarization cycle than that for the NV^- diamond center.

SINGLE PHOTON EMITTERS

An efficient single-photon emitter (SPE) suitable for quantum communication applications is supposed to emit photons with high intensity and well-defined energy and long lifetime in the near-infrared (NIR) range. This former condition is satisfied if a zero-phonon line (ZPL) dominates in the spectrum, while the phonon sideband (PSB) has a small contribution to the intensity of the SPEs.

Our rational search for efficient NIR SPEs is based on two premises: (1) if a substitutional defect changes the number of valence electrons in the system, it is likely that it will create local peaks (occupied and unoccupied) of the density of electronic states (DOS) inside of the band gap. Thus, if an excitation occurs between these local peaks, there is little capacity for the phonon-assisted energy reorganization, and, as a result, the defect emits a sharp ZPL peak, which is a favorable condition for SPEs. (2) A rough tuning of the emitted photon energy can be based on varying of the energy separation between the local peaks involved in the optical transition. This separation depends on hybridization between the dopant and host electronic states and on the bandgap of the host material: the larger the gap in the host semiconductor the stronger the hybridization between the host's electronic states. Thus, it is fair to assume that these states can also strongly hybridize with the defect's electronic states, and, therefore, lead to a large energy separation between the peaks associated to the defect.

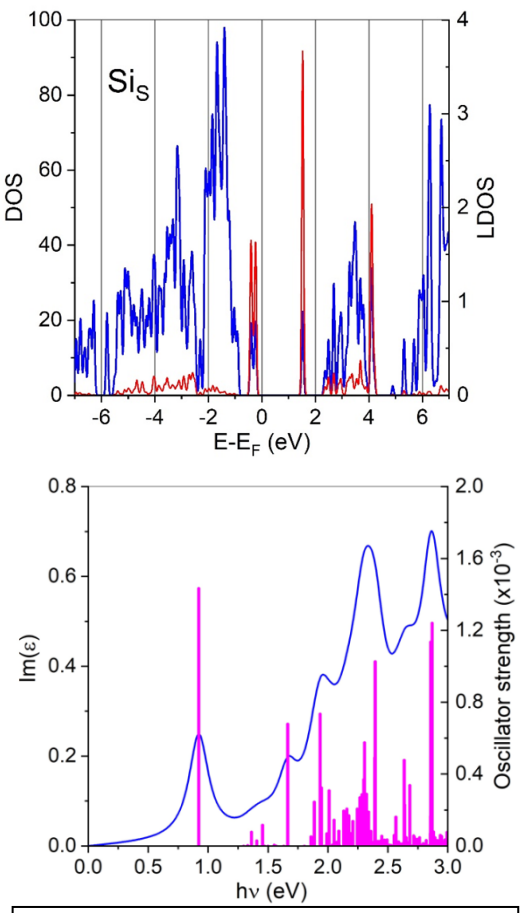
Based on the above consideration, we select two-dimensional (one monolayer) SnS2 as a host for promising defects. This semiconductor has a band gap of ~ 2.5 eV. The substitutional defects are chosen so that they change the number of valence electrons in the system by one or two. It is known that if the change is by one electron, most probably, the defect will be spin-polarized, which adds a degree of freedom to the expected properties. To fulfill these requirements, and considering that the components of the host, the selected defects were YX, where Y represents the dopant and X the substituted element, X = Sn or S, and Y = C, Si, Ge, N, P, and As. We evaluate from first principles their stability, electronic structure, and optical properties. The electronic structure of the considered systems has been calculated using the GW method with a single iteration (G0W0). To

obtain the optical excitation spectra, we apply the BSE formalism. Our calculations singled out three promising defects: the Si_S, P_S-, and the N_S- defects

Analysis of the IQP eigenstates that contribute the most to the corresponding BSE eigenstates of these defects indicate that these optical excitations may induce the emission of sharp ZPL peaks, without the presence of a significant phonon-side band, and consequently with the photon energy close to that of the excitation peaks. The latter in turn shows that the Si_S, P_S-, and N_S- defects are excellent SPEs candidates operating in near-infrared emission region.

Future Plans

We have preliminary evaluations of the electronic structure of cubic, wurtzite, and hexagonal boron-nitride with defects including vacancies and oxygen and sulfur substitutional defects. We found them to be promising spin qubits. Thus, we will perform detailed calculations and evaluate their properties related to the spin qubit functionality. In regard to SPEs, we have selected hosts $GeSe_2$, GeS_2 –which have a band gap of 2.7 eV and 3.7 eV, respectively– for future consideration. The defects in these hosts will be selected according to the methodology described in the Recent Progress section.



(Upper panel) Total density of IQP states (blue) and *p*-states of Si_S defect in SnS_2 (red). (Lower panel) Im(ε) (blue lines) and the oscillator strengths (pink bars) calculated for Si_S defect in SnS_2.

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Theory of fluctuating and critical quantum matter

Leon Balents, University of California, Santa Barbara

Keywords: thermal Hall effect, Berry phase, spin liquid, moiré

Research Scope

This program aims to understand phenomena in materials involving strong quantum effects and/or fluctuations. This includes the theory of spin liquid states of quantum magnets, as well as other quantum many body phases of two dimensional moiré systems and fluctuating condensates. One aspect of the research is into specific quantum magnetic materials, investigating the mechanisms by which they may exhibit spin liquid and related fluctuating quantum states. Another thrust is into experimental signatures of quantum coherent dynamics. In the latter, we have been working to understand the origin of thermal Hall effects through a systematic development of theory of transverse transport.

Recent Progress

Spin liquid physics of an inorganic triangular system: Recently our experimental collaborators discovered a novel magnetic material, $NaRuO_2$, which has a highly layered structure involving layers of triangular lattices of Ru^{3+} ions, well separated from one another. The material shows broad magnetic response, with a majority of magnetic weight measured by neutron scattering centered around 25meV. Combining a variety of thermodynamic measurements with the neutron scattering, we were led to propose a model of the magnetism similar to that proposed for two dimensional triangular lattice organic materials, with substantial ring exchange and proximity to an itinerant state playing an important role [pub 3]. In a follow-up theoretical work, we verified this picture from *ab initio* calculations jointly with the group of Roser Valentí [pub 4]. The results place $NaRuO_2$ in a category mixing Kitaev physics with ring exchange and itineracy, and make a spinon Fermi surface state a plausible candidate.

Thermal Hall effect and unconventional quasiparticle dynamics: The thermal Hall effect has become an increasingly studied probe of correlated quantum systems. Our group's interest has been primarily driven by proposals of thermal Hall effects in spin liquids, and by experiments on diverse magnetic materials. One of the outcomes of these experimental studies has been a primary role of phonons in the heat transport, despite the naïve expectation that phonons should be insensitive to magnetic fields because they are neutral. From this, we set out to study different mechanisms by which phonons might acquire knowledge of applied magnetic fields and time-reversal symmetry breaking. This can occur either via scattering of phonons from degrees of freedom which do see the magnetic field, or via intrinsic Berry phase effects that descend from non-dissipative coupling to such degrees of freedom. On examining these effects, we have been led to work more comprehensively to develop theory of dynamics and disequilibrium of bosonic quasiparticles such as phonons.

To put this in context, the description of near-equilibrium dynamics in terms of quasiparticles underlies much of the theory of transport and response. Modern research highlights how quantum effects enter this description through Berry curvature and quantum metric, as well as through exotic scattering channels. Our research aims to combine and systematize these mechanisms via an exact derivation of quantum kinetics order by order in a quantum parameter.

In one set of papers [pubs 1,2], we study the scattering of phonons by collective modes, showing how higher order terms in the Born series are needed to capture the "skew scattering" processes that break detailed balance and time-reversal symmetry, which are required to induce a Hall effect. To our knowledge this is the first systematic treatment of many-body skew scattering, and can be applied generally to phonons coupled to fluctuating modes such as magnons or paramagnons, or even to quantum critical modes. In we derived a result which allows completely general correlations between the collective modes to be included. We applied this result to the problem of phonon thermal Hall effect induced by scattering from magnons in an antiferromagnet (see Fig.1).

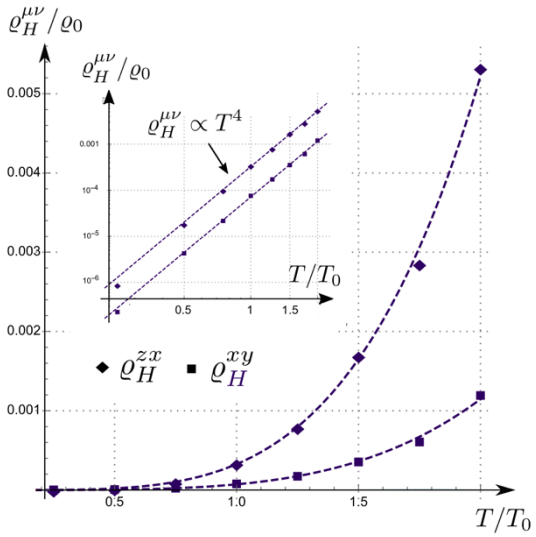

Fig. 1: Thermal Hall resistivity from many body phonon skew scattering off of antiferromagnetic magnons of a layered 2d magnet. The results show a T^4 power law and effects both for heat currents within and perpendicular to the 2d layers, a hallmark of phonon heat transport.

An alternative mechanism for the phonon thermal Hall effect is intrinsic, due to a Berry phase effect on the phonons due to their coupling to electrons or spins. This is known as phonon Hall viscosity. In the literature, we found that the Hall effect was obtained in a very opaque manner by a formal Kubo calculation, which limits understanding and is difficult to combine with other effects like the above scattering one. Hence we were led to try to understand the Hall viscosity effect through a kinetic equation. We found that an exact derivation of such a kinetic equation including Berry curvature for bosons was lacking in the literature, and therefore worked to obtain it. Our recent result is a general procedure to fully diagonalize the density matrix of an arbitrary free boson system in a quasi-classical expansion (in for example gradients of a potential, of temperature, etc.). In this way we thereby exactly obtain the Berry phase terms for both the kinetic equation and associated currents.

Through this formulation, we obtain the intrinsic thermal Hall effect from phonon Hall viscosity, as well as another result for Hall effect of magnons, much more directly and transparently. In addition, the kinetic method also gives the spatial profile of thermal currents

(see Fig.2). Thus the new methodology allows a full calculation of spatially resolved thermal properties in near-equilibrium Bose systems. These results are currently in preprint form [pub. 7]

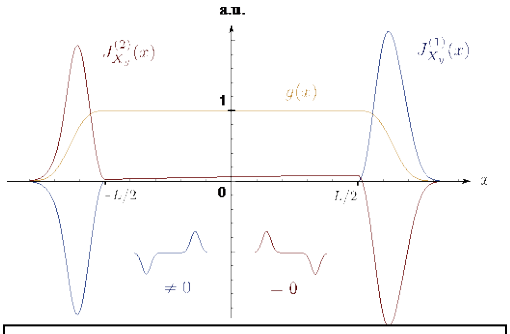

Fig. 2: Thermal currents in a Hall bar of phonons with Hall viscosity, calculated from the quantum kinetic approach.

Moiré Mott insulators at fractional filling: Experiments on two dimensional transition metal dichalcogenide materials with moiré patterns imposed due to incommensurate layers have proven that they can act as excellent simulators of triangular lattice Hubbard physics, with the additional ingredient of substantial further-neighbor interactions. The very strong coupling physics has been observed to stabilize "Wigner crystal" states which realize Mott insulators at fractional fillings, with spontaneously formation of various spatial patterns, including superlattices with triangular, honeycomb, and kagomé forms. The magnetic structure of these superlattices remains an intriguing open question. To address it, we developed an approach based on parton methods which allows simultaneously to study (see Fig.3) the formation of the spatial ordering itself, the onset of the Mott transition, *and* the magnetism [pub.5]. We find that both spin liquid and singlet valence bond solid states can be formed for certain particular fractional fillings.

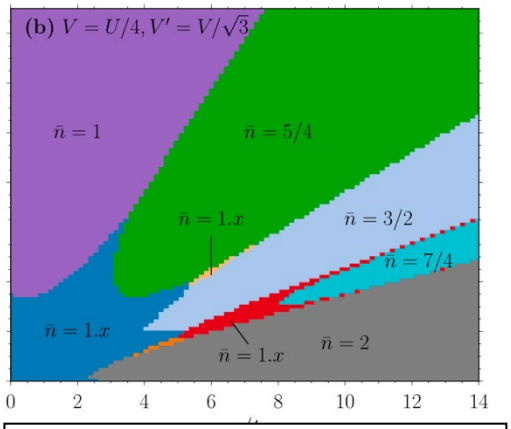

Fig. 3: Phase diagram in the chemical potential-interaction plane for moiré TMDs via the parton method.

Spin orbit coupled bosons: We collaborated with the group of Glenn Fredrickson, an expert on simulations of the statistical mechanics of polymer systems, to develop an approach to simulating quantum bosons with substantial thermal and quantum fluctuations. In our work [pub 6], we discovered that bosons with a Rashba-type spin-orbit coupling evolve from a striped state at low temperature to a fluctuating "microemulsion" phase, analogous to structure of some liquid crystals. Unlike the classical analogs, however, there are well-defined phase gradients and anisotropic superfluid correlations appearing in this intriguing state.

Future Plans

We are continuing to work on some of the above topics. In particular, we are working to apply the quantum kinetic approach to systems with natural spatial textures, to take advantage of the ability to spatially resolve physical quantities that is a feature of this method. We intend to apply this to skyrmion textures, and extend the method to fermionic quasiparticles as well. In addition, we would like to explore higher order semiclassical effects, which include the quantum metric and its influence of non-linear transport.

We are also working to extend the treatment of fractionally filled moiré systems beyond the parton mean field approach, using density matrix renormalization group methods and analytic approximations together.

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Prediction and Tuning of Spin Selectivity Properties of Chiral Nanomaterials via an Integrated Machine Learning -- First Principles Approach

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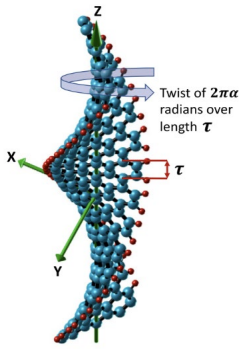
Keywords: Chiral Matter, First Principle Calculations, Machine Learning, Complex Alloys, Uncertainty Quantification

Research Scope

Chiral matter, i.e., structures with non–superimposable mirror images, offers unparalleled opportunities for impacting the design of novel quantum, electromagnetic, and spintronic devices [1]. Some chiral nanomaterials — particularly, quasi-one-dimensional helical nanostructures — display an unusual enhancement of electron spin transport properties (i.e, the Chiral Induced Spin Selectivity or CISS effect) [2], that can be harnessed for the design of new types of sensing technologies and room temperature quantum computing hardware. The overarching goal of the proposed research is to generate insights into the fascinating electronic and transport properties of chiral 1D nanomaterials, and thus enable their widespread adoption in the next generation of technologies. The PIs aim to accomplish this by developing novel computational and machine learning (ML) models, which will allow for the discovery and characterization of such materials. Specific goals are:



Chiral matter, like the twisted nanoribbon shown here, is often associated with fascinating electronic and transport properties.

1. First principles framework for chiral matter: To develop an efficient, systematic, helical symmetry adapted ab initio calculations framework for chiral 1D nanomaterials that explicitly considers critical factors influencing spin transport (including exchange interactions and unconventional spin-orbit coupling; to use this framework to study novel forms of chiral matter.

2. Spin transport calculations: To develop a computational framework for the calculation of spin transport and the spin selectivity effect in prototypical systems, that uses the above framework.

3. Machine Learning Models: To develop Machine Learning (ML) models using Bayesian approaches to predict electronic fields and spin transport properties from system parameters.

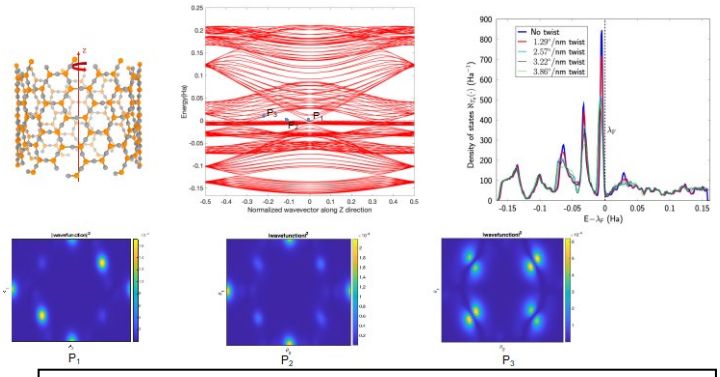
4. Machine-Learning Guided Materials Discovery: To discover novel material candidates with enhanced spin transport and spin selectivity properties — specifically by using the above ML models to search through a large composition–structure–deformation design space.

Recent Progress

We have made significant progress on all four fronts listed above. Specifically:

Novel computational scheme for electronic structure calculation of 1D chiral nanomaterials: We have formulated and implemented a novel spectral method for solving the single-electron problem, as it applies to 1D materials and structures (**Publication 1**). This allows for computation of the electronic structure of important technological materials such as nanotubes (of arbitrary chirality), nanowires, nanoribbons, chiral nanoassemblies, nanosprings and nanocoils, in an accurate, efficient and systematic manner. The computational savings offered by this scheme can be several orders of magnitude, when compared to conventional planewave codes. Recent and ongoing work includes the development of a mixed spectral–finite-difference technique for solving the Poisson problem, for fast evaluation of exchange interactions.

Discovery of novel forms of chiral matter: Exploring exotic phenomena in nanostructures, that emanate from flat band physics and strong electronic correlations is promising, especially in wire-like geometries. Such materials may allow the simultaneous study of dispersionless electronic states and chirality-induced transport effects. We used computational tools (described above) to study carbon Kagome nanotubes (CKNTs), a new carbon allotrope (**Publication 2**), and P2C3 "double Kagome" nanoribbons and nanotubes (manuscript under preparation). Both materials host electronic flat bands near the Fermi level and a singular peak in their electronic density of states. P2C3's electronic properties remain stable under deformations, whereas CKNTs show electronic phase transitions when deformed. Ongoing work covers structural phase transitions in these materials and experimental synthesis strategies.

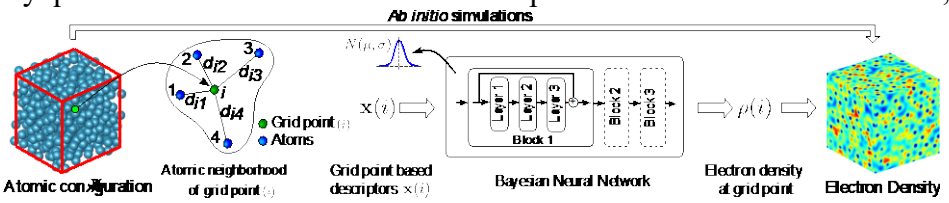


"Double Kagome" P2C3 nanotubes feature flat bands at the Fermi level, along with Dirac cones at the gamma point, The electronic states are resilient to mechanical

Charge and spin transport in chiral nanomaterials: We have been developing a framework for computing coherent transport in 1D nanomaterials. Our recent investigations have focused on charge transport, specifically Bloch and super-Bloch oscillations, and the role of Landau-Zener transitions in chiral nanomaterials with quadratic band crossing (like the CKNT and P2C3 nanotubes described above). Ongoing investigations include extension of this framework to spin transport in these materials.

Machine Learning Models: The ground state electron density, as can be evaluated from Density Functional Theory (DFT) calculations, is an attractive quantity for prediction via machine learning (ML) techniques. A strategy adopted by the PIs for this project is to predict the electron density of chiral materials of interest as a verifiable intermediate step, instead of direct prediction of the ultimately desired spin transport or spin selectivity properties. However, the computational expense of DFT scales cubically with system size which tends to stymie training data generation, making it difficult to develop quantifiably accurate, transferable ML models. To address this and other related challenges, we have:

a) Developed uncertainty quantification enabled ML models for prediction of electronic structure, that employ transfer learning to leverage the multi-scale nature of the training data, while comprehensively sampling system configurations using thermalization (**Publication 3**). The ML models (based on Bayesian neural networks) developed here are less reliant on heuristics and can reduce training data production costs by over 50%. We have shown that our ML models allow confident — and when verifiable, accurate — predictions for a wide variety of systems well beyond training, including ones with defects, different alloy compositions, and at unprecedented, multi-million-atom scales (while using only modest computational resources).



Bayesian Neural Network based ML model for electron density prediction.

b) Developed novel loss functions for Bayesian Neural Networks, to overcome key limitations of the state-of-the art Kullback-Leibler (KL) divergence-based variational inference. Theoretical analysis and computational experiments suggest that the proposed loss functions perform better, especially when the data sets are noisy or biased (**Publication 4**).

c) Proposed novel descriptors to represent the atomic neighborhoods that offer increased efficiency and accuracy when compared to existing alternatives. We have tested these new descriptors on the ternary Si-Ge-Sn (medium entropy) alloy system. When combined with Bayesian Optimization, such that the most informative compositions are used for training, our ML model can predict electronic structure across all composition space, while having been trained on very few compositions (manuscript under preparation).

Other Investigations: A few other research directions pursued over the past year include:

a) Development of a multiscale model that allows the investigation of strain and atomic relaxation effects in heterostructures (**Publication 5**). The model takes advantage of a bicrystallography framework, incorporates data from first principles simulations, and is applicable to important materials systems such as twisted bilayer graphene.

b) First principles investigations of a novel 2D allotrope of silicon featuring electronic flat bands (manuscript in preparation). Structural and electronic properties of this material, amorphization instabilities and stabilization through substrate strains have been investigated.
c) Cataloging strain induced electronic phase transitions in a wide variety of 2D materials — specifically, split and line graphs of bipartite lattices (manuscript in preparation).

Future Plans

Over the coming year, we will:

1. Extend the first principles computational framework for chiral matter, to enable hybrid DFT calculations for such systems, thus leading to better prediction of their electronic, optical and transport properties. We will also implement spin transport calculation methods for chiral structures and interface this framework with the first principles techniques described above.
2. Extend the ML models described above, to predict spin densities (instead of simply charge densities), to ultimately enable ML prediction of spin transport and selectivity.
3. Extend the ML models to enable materials optimization across a large structure-composition space. We will also develop a framework to ensure statistical validity of the ML models, so as to select material structure/stoichiometry optimally for training. Furthermore, we will reduce heuristics in the ML model using Optimal Transport Theory techniques.

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Physical behavior of two-dimensional materials with reduced symmetries

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Keywords: Reduced symmetries, phase transitions, topological properties, optical properties, 2D materials

Research Scope

This research encompasses multiple aspects of the theory of two-dimensional materials such as: structural phase transitions and the thermally-induced sudden modification of their elastic parameters, model 2D Hamiltonians displaying a topological crystalline insulating phase, topological antipolar-to-ferroelectric phase transitions in ferroelectric moiré homobilayers, magnetoelastic couplings in 2D magnets and 2D antiferromagnets, and non-linear optical behavior in 2D materials without a center of inversion. **Publication 1** provides an updated review of the field.

Recent Progress

a) Structural phase transitions in low-buckled silicene, germanene, and stanene. The clearest realization of two-dimensional topological insulators is the recent experiment by Bampoulis and collaborators [1] in which germanene is grown on a metal substrate. We hypothesize a situation in which silicene, germanene, and stanene exist without a supporting substrate and note that such structure has a degeneracy upon a mirror reflection with respect to the z-direction in Fig. 1 [2]. While graphene has a planar single energy minima in an energy landscape parameterized by the lattice parameter and the buckling height (Figs. 1a and 1d), silicene, germanene, and stanene all have two degenerate global minima and two additional local minima. For silicene, the global minima sits at the low-bickled configuration (Figs. 1b and 1e), while the high-buckled structure (Fig. 1c) is the global minima for germanene and stanene (Figs. 1f and 1g). As a result, freestanding silicene undergoes a structural transformation from a low-buckled phase onto an average planar phase at around 600 K. Details can be found in **Publication 2**.

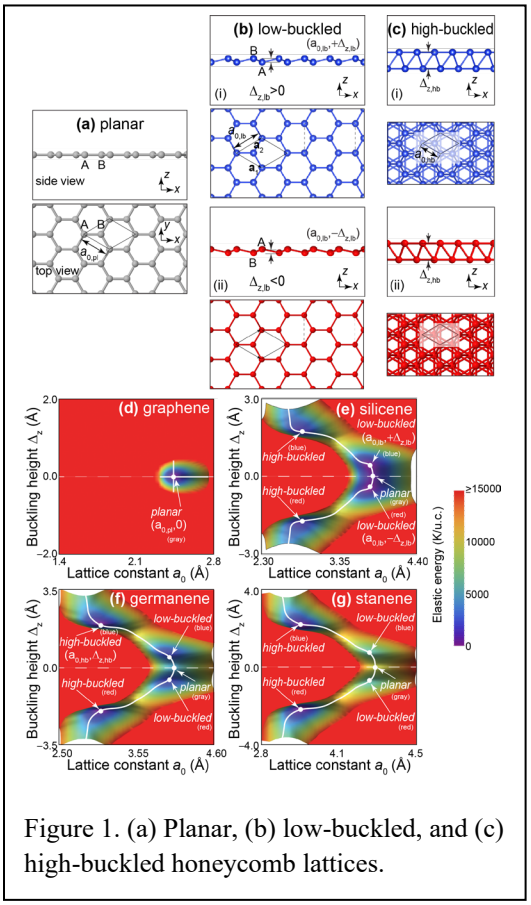

Figure 1. (a) Planar, (b) low-buckled, and (c) high-buckled honeycomb lattices.

b) Understanding the evolution of elastic properties on ferroelastic 2D materials. Commonly, elastic properties estimated by DFT are obtained at zero temperature. Nevertheless, some phase-

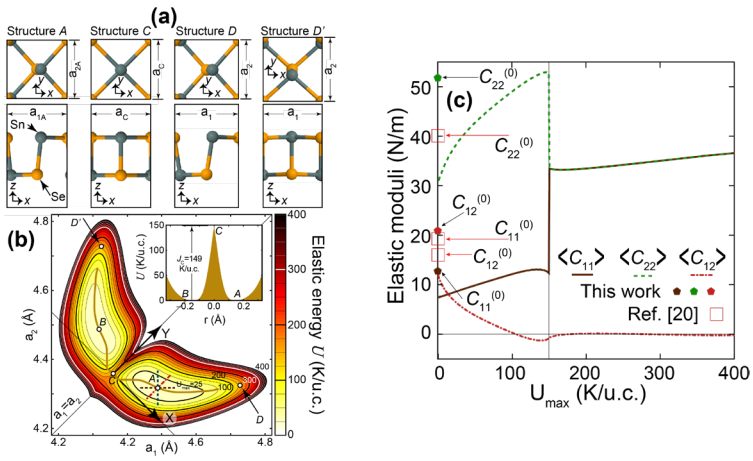

Figure 2. (a) Global minima (structure A), paraelectric (C), and two additional crystalline configurations (D and D') in the energy landscape (subplot b) of a SnSe monolayer. (c) The presence of the two degenerate minima results in a drastic modification of elastic parameters as the monolayer turns from ferroelectric to paraelectric.

changing materials with anisotropic lattice parameters and anisotropic elastic properties turn isotropic at a certain *Tc*, and one should expect that elastic properties turn isotropic in-plane as well. **Publication 3** deploys a methodology to calculate elastic coefficients at finite temperature from the energy landscape (Fig. 2).

c) 2D topological crystalline insulators without out-of-plane inversion symmetry. The original paper on topological crystalline insulators [3] focused on bulk and semi-infinite slab Hamiltonians. Along with G. Naumis, we demonstrated that the bulk Hamiltonian could be thinned down to slabs with a single unit-cell thickness, and that two topological measures (the vector field of $H^2(\mathbf{k})$—which becomes a 2×2 matrix—and the Pfaffians) remarkably remain invariant in the bulk and 2D limits. This naturally means that Fu's original bulk model ends up furnishing a 2D topological crystalline insulator as well. **Publication 4** contains the details.

d) Topological antipolar-to-ferroelectric phase transitions in ferroelectric moiré homobilayers. On a manuscript just resubmitted, thermally-driven phase transitions from antipolar moiré homobilayers were analyzed. Moiré bilayers possess a non-trivial structural topology in which AB and BA triangular domains are separated by dislocation lines meeting at AA topological nodes [4], and the density of AA nodes remains unchanged at any finite temperature.

e) Understanding magnetoelastic couplings in 2D antiferromagnets. On a manuscript currently under preparation, student John Davis has been able to diagonalize force constants from phonon dispersion calculations, to provide a unique perspective on magnetoelastic couplings of a $CrSiTe_3$ antiferromagnetic 2D material. Concurrently, Heisenberg spin models are being extracted directly from DFT data. A manuscript will be submitted in April, 2024.

Future Plans

A tool for optical second harmonic generation based on localized atomic orbitals. As a necessary step toward the creation of sources for entangled photons based on 2D materials, calculating the second-harmonic generation tensor $X^{(2)}$ is necessary. This type of calculation is not

available in most DFT tools, and it must be developed. Over the past year, student Luis Enrique Rosas has overcome the main technical challenge of those calculations: calculating electric dipole matrix elements on the localized atomic basis created by the SIESTA DFT code. Together with Postdoctoral Associate Angiolo Huaman and the PI, we are currently writing code to calculate $X^{(1)}$ and $X^{(2)}$ for a ferroelectric AB hexagonal boron nitride bilayer and expect to have a manuscript ready for publication in Fall, 2024. At that point, a method to estimate the creation of probabilistic entangled photon pairs will be devised, and a second publication is expected out of those results. Time allowing, we will calculate $X^{(2)}$ at atomistic defects as well, with the long-end goal of providing estimates for the probability to create deterministic sources of entangled photon pairs out from engineered atomistic defects in 2D materials.

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Topological Heavy Fermions In Twisted Bilayer Graphene

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Keywords: Moire samples, twisted bilayer graphene, heavy fermions, strongly interacting systems

Research Scope

The current research tries to obtain a model of the myriad of phases that appear in twisted bilayer and trilayer graphene. I will review the topological heavy fermion model of twisted bilayer graphene and review recent progress in analytical DMFT approximations on this model that can be used to compute transport coefficients. We show calculations that we believe uniquely can match recent Seeback coefficient experiments which show the presence of different carriers with asymmetric mass and asymmetric lifetime. We then show how strain and particle hole breaking relaxation can be easily incorporated into the heavy fermion model which leads to the following results: 1.

the explanation of the observation of stronger correlated insulator states on the electron doped side than the hole doped side of tbg, despite the bare dispersion being stronger on that side and 2. An analytical understanding of the stability of the IKS state recently found in STM experiments to be the ground state of the correlated CI including its wavevector. We will also discuss preliminary results on the superconductivity of tbg.

Recent Progress

Very recently, we studied the interacting transport properties of twisted bilayer graphene (TBG) using the topological heavy-fermion (THF) model. In the THF model, TBG comprises localized, correlated *f*-electrons and itinerant, dispersive *c*-electrons. We focus on the Seebeck coefficient, which quantifies the voltage difference arising from a temperature gradient. We find that the TBG's Seebeck coefficient shows unconventional (strongly-interacting) traits: negative values with sawtooth oscillations at positive fillings, contrasting typical band-theory expectations. This behavior is naturally attributed to the presence of heavy (correlated, short-lived *f*-electrons) and light (dispersive, long-lived *c*-electrons) electronic bands. Their longer lifetime and stronger dispersion lead to a dominant transport contribution from the *c*-electrons. At positive integer fillings, the correlated TBG insulators feature *c*- (*f*-)electron bands on the electron (hole) doping side, leading to an overall negative Seebeck coefficient. Additionally, sawtooth oscillations occur around each integer filling due to gap openings. Our results highlight the essential importance of electron correlations in understanding the transport properties of TBG and, in particular, of the lifetime asymmetry between the two fermionic species (naturally captured by the THF model). Our findings are corroborated by new experiments in both twisted bilayer and trilayer graphene, and show the natural presence of strongly-correlated heavy and light carriers in the system.

We also developed heavy fermion DMFT calculations to test the physics of TBG in relation to the Heavy Fermion predictions. We use the topological heavy fermion (THF) model and its Kondo Lattice (KL) formulation to study the symmetric Kondo state in twisted bilayer graphene. Via a large-N approximation, we find a symmetric Kondo (SK) state in KL mode at fillings ν=0,±1,±2. In the SK state, all symmetries are preserved and the local moments are Kondo screened by the conduction electrons. At the mean-field level of the THF model at ν=0,±1,±2,±3, we also find a similar symmetric state. We study the stability of the symmetric state by comparing its energy with the ordered states and find the ordered states to have lower energy. However, moving away from integer fillings by doping holes to the light bands, we find the energy difference is reduced, which suggests the loss of ordering and a tendency towards Kondo screening. In order to include many-body effects beyond the mean-field approximation, we perform dynamical mean-field theory (DMFT) calculations on the THF model. We find the spin susceptibility follows a Curie behavior at ν=0,±1,±2 down to ∼2K where the onset of screening of the local moment becomes visible. This hints to very low Kondo temperatures at these fillings, in agreement with the outcome of our mean-field calculations. At non-integer filling ν=±0.5,±0.8,±1.2 DMFT shows deviations from a 1/T-susceptibility at much higher temperatures, suggesting a more effective screening of local moments with doping. Finally, we study the effect of a $C3z$-rotational-symmetry-breaking strain via mean-field approaches and find that a symmetric phase (that only breaks $C3z$ symmetry) can be stabilized at sufficiently large strain at ν=0,±1,±2. Our results suggest that a symmetric Kondo phase is strongly suppressed at integer fillings, but could be stabilized either at non-integer fillings or by applying strain.

Future Plans

We will develop further transport calculations in TBG through the prism of heavy fermions, and work with our experimental colleagues to test their validity

Publications

1. Kondo Lattice Model of Magic-Angle Twisted-Bilayer Graphene: Hund's Rule, Local-Moment Fluctuations, and Low-Energy Effective Theory; Haoyu Hu, B. Andrei Bernevig, and Alexei M. Tsvelik; Phys. Rev. Lett. 131, 026502, July 2023

2. Symmetric Kondo Lattice States in Doped Strained Twisted Bilayer Graphene; Haoyu Hu, Gautam Rai, Lorenzo Crippa, Jonah Herzog-Arbeitman, Dumitru Călugăru, Tim Wehling, Giorgio Sangiovanni, Roser Valentí, Alexei M. Tsvelik, and B. Andrei Bernevig, Phys. Rev. Lett. 131, 166501, October 2023

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Coupled Electron-Phonon Transport from First Principles

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Keywords: Electron-phonon coupling; electron and phonon drag; *ab initio* thermoelectric transport;

Research Scope

The focus of this project is to implement a predictive *ab initio* approach, free of adjustable parameters, to explore the transport regime in which the interaction between electrons and phonons results in strong electron and phonon drag effects. The approach is based on an efficient, home-grown computational scheme to fully solve the coupled electron and phonon Boltzmann transport equations without approximation. It accurately describes the momentum-conserving (normal) and momentum-relaxing (Umklapp) intrinsic electron-phonon and phonon-phonon scattering processes as well as extrinsic scattering processes such as from charged impurities and defects. The computational approach satisfies by construction the Kelvin-Onsager relation, a fundamental requirement of thermodynamics. The primary materials focus is on doped semiconductors and semimetals. *Ab initio* coupled electron-phonon transport in the latter has been little studied. New materials will be identified in which strong electron-phonon drag behavior occurs.

A side-project has been the development of an *ab initio* approach to calculate thermal properties in magnetic materials. The approach incorporates the renormalization of phonon modes by (1) chemical disorder (i.e. alloying); (2) temperature- and pressure-induced vibrational and magnetic disorders; (3) spin–lattice coupling. Fully non-collinear magnetism with spin–orbit coupling is included.

Recent Progress

Our recent work to study coupled electron-phonon transport from first principles implements the elphbolt code [1], a home-grown code that numerically solves the coupled Boltzmann transport equations for electrons and phonons in applied electric field and temperature gradient. Outputs are the non-equilibrium electron and phonon distribution functions from which all thermoelectric transport coefficients: electrical conductivity, Seebeck thermopower, electronic and phonon contributions to the Peltier thermopower and the thermal conductivity are calculated. The code also obtains decoupled solutions where phonons (electrons) are taken to remain in

equilibrium when considering electron (phonon) transport (Bloch approximation). The differences between the calculated transport coefficients for these two cases gives the drag enhancement for each transport coefficient. Excellent agreement was obtained with the measured drag enhanced Seebeck thermopower in both n-doped and p-doped silicon [R1, 1, 2], providing preliminary validation of the predictive power of the approach.

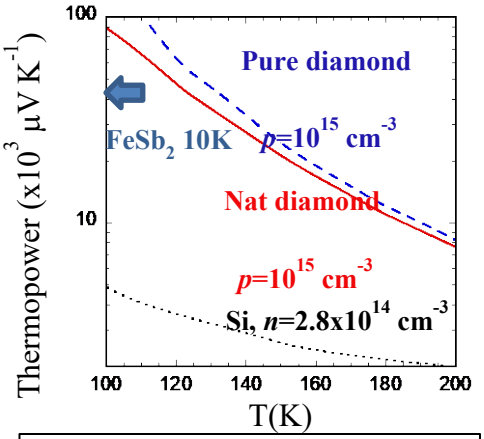

Figure 1: Calculated thermopower of diamond and Si .vs. temperature. Solid red (dashed blue) curve shows the thermopower for naturally occurring (isotopically pure) hole-doped diamond. The calculated thermopower of Si (black dotted) is provided for comparison. Blue arrow shows the measured thermopower of $FeSb_2$.

(i) Colossal drag enhanced thermopower in diamond [1]: Strong drag behavior requires weak dissipative scattering channels for electrons and phonons in order to retain the momentum exchanged between electron and phonon subsystems that occurs through electron-phonon interactions. With this in mind, we examined the thermopower of lightly doped diamond. Diamond has exceedingly weak anharmonic decay of phonons, which promotes retention of quasi-momentum in the electron-phonon system even though electron-phonon interactions in diamond are relatively weak. We identified enormous drag enhancements to the diamond thermopower, with values of around 100,000 μV K^{-1} being found at 100 K (Fig. 1). These values are significantly larger than the highest measured values in the correlated metal $FeSb_2$. Moreover, they occur at an order of magnitude larger temperatures.

(ii) Large drag thermopower in semiconductors at high carrier density [2]: In lightly-doped semiconductors, large drag enhanced thermopower values can occur at low temperatures through momentum exchange between carriers and low frequency acoustic phonons, when the anharmonic decay rates for these phonons are small. In contrast, the thermopower values in many metals are orders of magnitude smaller even though acoustic phonons across the full frequency range are coupled to electrons. The regime of carrier densities connecting these two extremes has been little explored. To begin to fill this knowledge gap, we calculated the thermopowers of two semiconductors, silicon and boron arsenide (BAs), up to high hole densities of 10^{21} cm^{-3}. Surprisingly, we found that large drag enhancements to the Si and BAs thermopowers occur, extending even up to room temperature (Fig. 2a). Moreover, the drag enhancements arise from momentum exchange between carriers and high-frequency acoustic phonons (Fig. 2b), in contrast to the known behavior at low carrier densities, in which only low frequency phonons participate. Weak anharmonic decay of these high frequency phonons in BAs leads to particularly large drag behavior. These unexpected results motivate exploration of electron-phonon drag in semimetallic systems, which have been relatively unstudied using *ab initio* schemes.

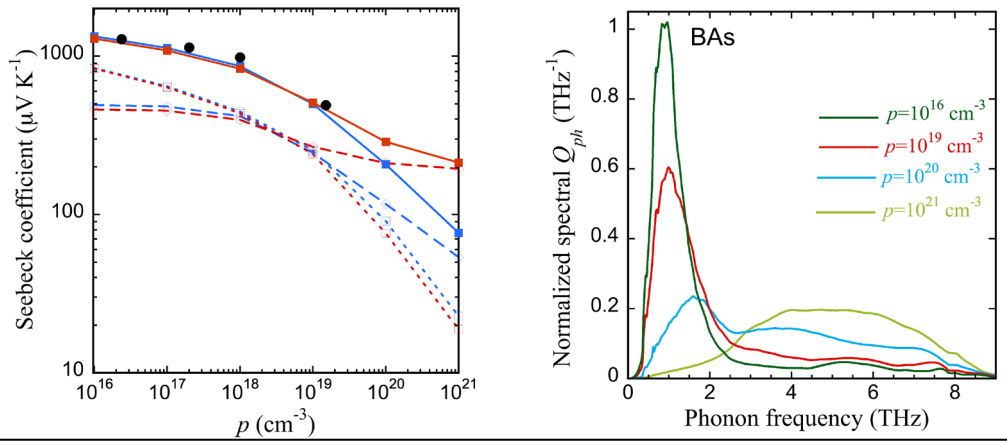

Figure 2: a) Seebeck coefficients, S, for BAs and Si as a function of hole density, p, at 300 K. BAs: solid red curve; Si: solid blue curve. Black circles are measured data for Si [R2]. Short-dashed and long- dashed curves give diffusive (S_{diff}) and drag (S_{drag}) contributions, with $S_{diff}+S_{drag}=S$. b) Normalized spectral phonon contributions to the room temperature Peltier thermopower of BAs for four hole densities: p= 10^{16} cm^{-3}, 10^{19} cm^{-3}, 10^{20} cm^{-3}, and 10^{21} cm^{-3}. A shift in spectral contributions to higher frequency is evident at high p.

(iii) Thermal properties of magnetic materials with unknown entropy—Application to Invar [3, 4]: We developed a theoretical approach to study the thermal properties in magnetic materials where an accepted model for the magnetic interactions does not exist [3]. In place of a specific model for magnetic interactions, the approach integrates measurements of temperature or pressure dependent magnetization of the studied material into a first principles computational scheme that incorporates the renormalization of phonon modes by alloying, temperature- and pressure-induced atomic displacements and magnetic moment orientations, and spin–lattice coupling. Fully non-collinear magnetism with spin–orbit coupling is also included.

We applied the approach to investigate the anomalously small thermal expansion in the Invar alloy, $Fe_{0.65}Ni_{0.35}$, which extends over a wide range of temperatures and pressures. An accepted explanation for the underlying cause of this behavior has eluded researchers since its first observation by Guillaume in 1897. Through our independent theoretical work and in collaboration with our experimental colleagues who measured the pressure dependences of phonon and magnetic contributions to the entropy, we found that the small Invar thermal expansion coefficient occurs because the normal positive contribution to thermal expansion from phonons is nearly exactly canceled by a large negative magnetic contribution [3, 4].

Future Plans

Coupled electron-phonon transport in semimetals: The large drag effects found in the thermopower of highly-doped semiconductors motivate examination of coupled electron-phonon transport in semimetals, in particular those for which anharmonic phonon decay is weak. Toward that end, we have initiated first principles calculations for theta-phase tantalum nitride (θ-TaN). θ-TaN has been predicted to be a topological semimetal [R3] and a high thermal conductivity

material similar to BAs [R4]. Calculations are focused on understanding the complex interplay between electron-phonon and phonon-phonon scattering as the Fermi surface evolves with changing chemical potential. Guided by these results, calculations will also be performed for other semimetals (including 2D systems) for which large electron-phonon drag behavior may occur.

Electron-phonon drag in semimetals in magnetic field: We will examine electron-phonon drag behavior in semimetals in a magnetic field, by extending the elphbolt code to include this capability. Longitudinal and transverse thermoelectric transport coefficients will be calculated up to high field strengths where the transverse transport coefficients should be large enough for accurate numerical resolution. Extrapolation to low field will allow direct comparison to available measured data.

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Ab initio complete cell quantum embedding and diagrammatic coupled cluster for correlated materials phase diagrams

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Keywords: Correlated quantum materials, many-body theory, electronic structure

Research Scope

Work in my group funded under this program focuses on the quantitative description of the electronic properties of correlated quantum materials. While in prior years we have devoted much effort towards the definitive solutions of models (such as our work on resolving the ground-state stripe order in the Hubbard model [1]) in recent years, we have focused on moving beyond low-energy models. In particular, we are developing techniques to describe correlated quantum material electronic properties with structural and compositional specificity. In practice, this means that our starting point is the full ab initio electronic Hamiltonian, and we choose to retain this complexity throughout our treatment without any downfolding. The computational tools we are developing in this program are based on quantum embedding theories, such as density matrix embedding theory [2] and dynamical mean-field theory, in the latter case within the full cell, all-electron picture we previously introduced [3]. We employ many-body solvers ranging from ab initio tensor network methods to methods taken from quantum chemistry, such as coupled cluster theories. These tools are then incorporated into large-scale electronic structure calculations employing the full interacting electronic Hamiltonian.

Recent Progress

Recent highlights include the successful demonstration of the above strategy in simulating material-specific trends in magnetism and superconductivity in the cuprates, and material specific-trends in Kondo temperatures for magnetic impurities in hosts. More specifically, (1) Across a series of cuprate materials and structures, we have deployed large scale quantum embedding calculations (e.g. embedding an impurity with over 900 orbitals). In our first work, this allowed us to reproduce the layer effect in magnetism, and identify its origin in fluctuations between the copper-oxygen plane and the apical oxygen and buffer orbitals (Science, 377, 1192 (2022) [4]). In a second work, we showed that we obtain the pressure dependence and layer dependence of the pairing order across a series of four cuprate structures and three cuprate materials as a function of pressure and composition (arxiv:2306.16561 [5]), (2) We have also targeted the quantitative description of Kondo spectra and temperatures for a series of transition metal impurities embedded in bulk copper, aiming to converge to exactness. (By exact, we mean all aspects of the electronic problem are converged with quantifiable error, and any remaining errors stem from non-electronic effects such as phonons). Across the transition metal series, we reproduce the trends in Kondo temperatures, and quantitatively capture physics beyond the “effective spin” model.

Future Plans

Our future plans center on extending the above framework to more materials, with more complex structures, with greater fidelity and reliability in the predictions. While we do not (and cannot) claim to describe all the physics in these complex quantum materials, it is clear that our framework contains relevant ingredients to describe some interesting physics with material specificity. Our immediate target therefore is to improve the robustness and efficacy with a view to future efforts in correlated materials screening and discovery.

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Traversing the "death valley" separating short and long times in non-equilibrium quantum dynamical simulations of real materials

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Co-PIs: Marco Bernardi, Caltech, Emanuel Gull, Michigan, Eran Rabani, Berkeley, David Reichman, Columbia, Andrew Millis, Columbia, Chao Yang, LBL, Susan Woodward, LLNL

Keywords: Correlated quantum materials, non-equilibrium, quantum dynamics, many-body theory, tensor networks

Research Scope

Our collaboration aims to tackle the fundamental bottlenecks that inhibit the faithful simulation of non-equilibrium quantum materials: the need to incorporate realistic interactions and the need to cross the "death valley" separating the very short times accessible to brute force simulation and the long times where hydrodynamics takes over. Our work is organized around three intellectual thrusts: (1) improving first principles simulations to reach longer time-scales, and improve the treatment of electron/phonon/exciton interactions; (2) diagrammatic methods for impurity models out of equilibrium; (3) long time-scale tensor network simulations to reach longer times and higher dimension. Common to all thrusts are a set of cross-cutting mathematical and computational science challenges: propagating/extrapolating stiff equations of motion for long times, handling large amount of data in time-propagation (especially for multi-time quantities), and the need for scalable and GPU-performant implementations.

Recent Progress

In the last year we have made significant progress both with respect to the common crosscutting mathematical challenges as well as in the science drivers of quantum dynamics.

For example, in the first category, we have developed and implemented several kinds of reduced order models and extrapolations to reduce the cost of time-propagation and to reach longer times. In [1], we show that an application of dynamic mode decomposition (DMD), which decomposes the Koopmans operator to define reduced space-time coordinates to extrapolate the long-time dynamics, allows us to propagate stochastic Green's functions to obtain converged optical spectra. Similarly in [2], we show that DMD allows us to push simulations of the Boltzmann equation in the strong-field driving of semiconductors all the way to reach the steady-state regime, thus crossing the death valley of simulation. We have also developed denoising strategies based on positive definiteness of spectral functions [3] which enable reliable time extrapolations from noisy short-time data.

We have also developed better time-integrators, including multi-rate methods for non-equilibrium electron-phonon dynamics. These exploit the fact that phonon-phonon interactions are very expensive to compute due to the fine momentum grid, while electron-phonon interactions are less expensive and on a much finer time-scale. For a given step size, we find that multi-rate methods are orders of magnitude faster. We have further modified adaptive time-integrators for the Kadanoff-Baym 2-time equations, reorganizing the computation to take advantage of the causal structure of the equations. Finally, we have demonstrated the ability to use hierarchical off-diagonal compression (in particular, the HODLR decomposition) to reduce the storage and efficiently manipulate the two-time Green's function, which greatly reduces the cost of the history integrals in non-equilibrium Green's function simulations [4].

In the context of science drivers and the physics of quantum dynamics, we mention in particular three results. In the first, we have uncovered the detailed structure of tensor network influence functionals, obtaining the correct continuum limit, the connection to quantum embedding theories, and carrying out some of the first simulations of fermionic impurity dynamics within this formalism [5, 6]. In the second, we show that we can directly compute the stationary state of non-equilibrium impurity dynamics within the diagrammatic picture (we apply this to show the destruction of the Kondo resonance under applied bias [7]), thus avoiding the need for difficult time propagation altogether. Finally, in the third driver, we show that we can carry out long-time scale Trotterized tensor network dynamics far beyond what has previously been simulated, using a mixed Heisenberg-Schrödinger picture and ideas from belief propagation to evaluate tensor network expectation values [8].

Future Plans

In addition to further progress along the computational and algorithmic problems above, we aim in the near future to use our new methods to reproduce, analyze, and explain some recent physical non-equilibrium experiments at a more microscopic level. An example of this is the unusual long time-scales of metal insulator transition observed in a non-equilibrium textured Mott insulator and recently studied phenomenologically (in terms of a free energy landscape) in our collaboration [9].

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Keywords: mesoscale, phase-field, phase transitions, quantum materials, domain patterns

Research Scope

The central goal of COMMS is to develop mesoscale computational models, efficient numerical algorithms for exascale computation, and software validated for quantum and functional materials. Built upon the accomplishments of the prior report period, the specific efforts of current research have been focused on correlated electronic systems and complex topological structures. We developed a dynamical phase-field model integrating polarization dynamics, elastodynamics, and electronic carriers and a phase-field model of coupled structural and electronic phase transitions in the presence of both electronic carriers and oxygen vacancies as well as a two-temperature dynamical phase-field model of strongly correlated electron systems. We have achieved a fundamental understanding of coupled lattice and electronic degrees of freedom, including the discovery of intrinsic phase and voltage oscillations during metal-insulator transitions and of the critical roles of oxygen vacancies and sublattice-dependent antiferromagnetic transitions in rare earth nickelates. We elucidated the role of electronic carriers in mesoscale topological polar structural transformations and their pathways validated by experiments. We have made significant progress in the development of numerical solvers based on multigrid pre-conditioners and have developed an alpha version of our software module for modeling metal-insulator transitions as well as several effective software tools for postprocessing and characterizing the mesoscale structures and topologies produced by phase-field simulations in both real and Fourier spaces.

Recent Progress

Insulator-to-metal (IMT) Transitions in the Strongly Correlated VO_2: Our team not only pioneered the phase-field model development of metal-insulator transitions but also using the example of VO_2 to reveal a fascinating electro-chemo-mechanical mechanism at play that includes coupled electronic-structural phase transitions in the presence of oxygen vacancies, the simultaneous evolution dynamics of electronic carriers, structural orders, as well as new non-equilibrium phases [1]. This teamwork has also led to the computational prediction of intrinsic self-oscillations [2] and the discovery that the vacancy redox reactions and the insulator-metal transitions mutually promote each other, leading to faster voltage self-oscillations in VO_2 [3].

Discovery of Sublattice-Dependent Antiferromagnetic Transition in Perovskite Nickelates [4]*:* We used the Landau theory to investigate the coupled insulator-metal, structural, and magnetic phase transitions in another fascinating correlated material, perovskite rare earth nickelates. We reproduced the experimentally measured temperature-tolerance factor phase diagram and found that the antiferromagnetic transition is sublattice-dependent, which illustrates the existing but

confusing experimental susceptibilities. Our results also proved the noncollinear nature of the magnetic structure of bulk nickelates, shedding new light on the relevant long-standing debate.

Coupled Electron and Lattice Systems in Correlated Material Systems: We have established a novel dynamical phase-field model for coupled electron and lattice systems in strongly correlated materials under ultrafast excitation on the ps-ns timescales and model the electronic and lattice dynamic responses of the non-equilibrium states of the prototypical correlated material $Ca_3Ru_2O_7$ on the ultrafast timescales [5]. We also formulated a comprehensive mesoscale thermodynamic model of coupled electronic phase transitions including metal-insulator transitions, magnetic phase transitions, charge density waves (CDW) formation, superconducting phase transitions with the initial focus on the couple of CDW formation [6].

Mesoscale Modeling of Complex Topological Systems: We have developed a *dynamical* phase-field model (DPFM) for modeling the co-evolution of electronic carriers and ferroelectric domains [7]. Our phase-field simulations reveal the existence of rich mesoscale structures that can transform from one to another under external biases and the key role of electronic charge carriers in the thermodynamic stability and phase transitions among different mesoscale polar phases induced by an above-bandgap excitation [8]. In particular, we study the thermodynamics of nanoscale polar structures in $PbTiO_3/SrTiO_3$ ferroelectric superlattices induced by above-bandgap optical excitation using a phase-field model explicitly considering both structural and electronic processes (Figure 1). We demonstrate that the light-excited carriers provide the charge compensation of polarization bound charges and the lattice thermal energy, both of which are key to the thermodynamic stabilization of a previously observed supercrystal, a three dimensionally periodic nanostructure, within a window of substrate strains, while different mechanical and electrical boundary conditions can stabilize a number of other nanoscale polar structures by balancing the competing short-range exchange interactions responsible for the domain wall energy and long-range electrostatic and elastic interactions. The insights into the light-induced formation and richness of nanoscale structures from this work offer theoretical guidance for manipulating the stability of nanoscale polar structures employing a combination of thermal, mechanical, and electrical stimuli as well as light.

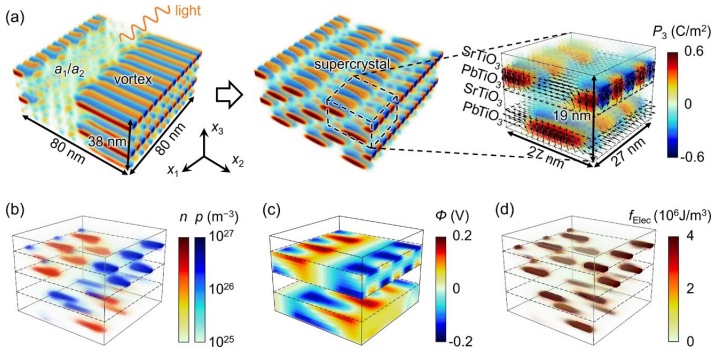

Fig. 1: Phase-field simulation of light-induced transformation of a mesoscale vortex/twin ferroelectric polar structure to another mesoscale ferroelectric supercrystal. (a) The $PbTiO_3/SrTiO_3$ superlattice transforms from a pristine state consisting of vortex tube and a_1/a_2 twin domain regions to a 3-dimensional ferroelectric supercrystal upon application of an above-bandgap light pulse. The right panel shows the polarization vectors (black arrows) of a selected region corresponding to a supercell of the supercrystal, with the $PbTiO_3$ and $SrTiO_3$ layers labeled and the $PbTiO_3/SrTiO_3$ interfaces marked by black dashed lines. (b)-(d) The spatial distributions of (b) the charge carrier concentration n and p, (c) the electric potential Φ, and (d) the electrostatic energy density f_{Elec} within the same selected region, upon formation of the supercrystal [8].

Numerical Algorithm and Open-Source Software Package Developments: Our team has made substantial progress in enhancing solvers for phase-field equations, achieving significant improvements in speed. We integrated the HX preconditioner into our solver for the Maxwell equation's linear system resulting from discretization with markedly enhanced performance [9]. In addressing the complexities of superconductivity models coupled with elasticity, we have developed numerical schemes and solvers utilizing the finite element method. We achieved enhanced solver efficiency, as evidenced by our numerical experiments. We have made significant progress on the Q-POP framework, an open-source suite designed for flexible phase-field simulations. The initial release offers a variety of capabilities, including modules for simulating insulator-metal transitions (Q-POP-IMT) with documentation. Furthermore, we have developed several pre- and post-simulation utility tools including a routine (Q-POP-Diffraction) to compute the X-ray diffraction patterns of phase-field predicted structures to allow a direct comparison between phase-field simulations and diffraction experiments [11]. We have developed a software tool to simulate the second harmonic generation (SHG) termed ♯SHAARP of crystals [12].

Future Plans

Develop a phase-field model of electronic antidoping and apply it to simulate the synaptic resistance trees found in hydrogenated perovskite nickelates and illustrate its mechanism.

Develop the macroscopic superconducting phase-field model in entire-temperature regime by extracting dynamics from microscopic superconducting theory; incorporate the coupling of the superconducting order parameter with other quantum orders and fluctuations and calculate the temperature-doping/density phase diagram of the fluctuating and high-T_c superconductivity; and develop the phase-field model of the superconducting phase coherence in Josephson junctions, and design/propose the scheme for experimental detection and application of quantum devices.

Develop a new characterization tool (Q-POP-NLO) for the direct evaluation of second-harmonic generation (SHG) and other non-linear optical responses in ferroelectric crystals with arbitrary polarization distributions and a GPU-accelerated finite-difference method based 3D dynamical phase-field model that incorporates coupled dynamics of strain, polarization, charge, and electromagnetic waves in ferroelectrics-based multiphase system (Q-Pop-FerroDyn) to simulate the nonlinear interaction between strong transient THz/optical pulse and ferroelectric systems with inhomogeneous polarization patterns; and demonstrate the possibility of performing basic quantum operation (Rabi oscillation and Ramsey interference) for quantum computing based on strongly coupled THz photons and optical phonons (polarization).

Develop robust and efficient solvers for Maxwell's equations in mixed formulation, ensuring the parameter robustness and design advanced time discretization methods and corresponding fast solvers for the time-dependent Ginzburg-Landau model to significantly enhance simulation efficiency; Develop fast and robust solvers specifically for phase-field models that are coupled with Maxwell's equations in mixed formulation.

Advance the implementation of highly-performant, portable, and scalable multigrid, finite-element solvers for phase-field equations governing insulator-metal transitions, electron carrier dynamics, polarization dynamics, spin dynamics, and superconducting phase transitions. To achieve this, exascale-capable libraries such as FEniCS-DOLFINx (CPUs-only) and MFEM (supporting both CPU and GPU architectures) will be leveraged.

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Machine Learning Assisted Multi-Scale Dynamical Modeling of Functional Electronic Materials

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Keywords: Machine Learning, Artificial Neural Network, Strongly Correlated Electron Systems, Nonequilibrium Dynamics, Magnetism and Magnetization Dynamics.

Research Scope

The objective of this project is to develop a machine learning framework that enables multi-scale dynamical modeling of functional electronic materials, which are at the forefront of research in modern condensed-matter physics and materials science. The complexity of these functional materials often leads to intricate electronic textures that emerge at the mesoscopic scale, such as stripes and checkerboards in high-temperature superconductors, metallic filaments in resistive-switching materials, and skyrmions in itinerant magnets. An accurate and comprehensive dynamical modeling of these electronic textures, however, is a challenging multi-scale task. On the one hand, large-scale dynamical simulations are necessary to properly describe mesoscopic pattern formation processes. On the other hand, sophisticated electronic structure methods and many-body techniques have to be employed to compute the electronic contribution to the driving forces of the mesoscale textures. Repeated quantum calculations that are computationally expensive thus significantly limit the accessible scale of dynamical simulations.

The machine learning (ML) force-field methods, originally pioneered in quantum chemistry to enable large-scale *ab initio* molecular dynamics, are generalized to broad condensed-matter systems, including correlated electron models and itinerant magnets. The central idea of this approach is to utilize the universal expressive capability of deep-learning neural networks to develop an accurate and efficient model for computing the generalized force fields. By effectively emulating the time-consuming many-body calculations, the ML framework combines the best of two worlds: the efficiency of classical force-field models and the desired accuracy of quantum calculations. Specific implementations of descriptors, which are a crucial component for ensuring symmetry and enhancing performance of ML models, will be benchmarked and optimized. Also importantly, in order to describe dynamical behaviors of driven systems, the ML framework will be further developed to model force fields that originate from highly nonequilibrium electrons.

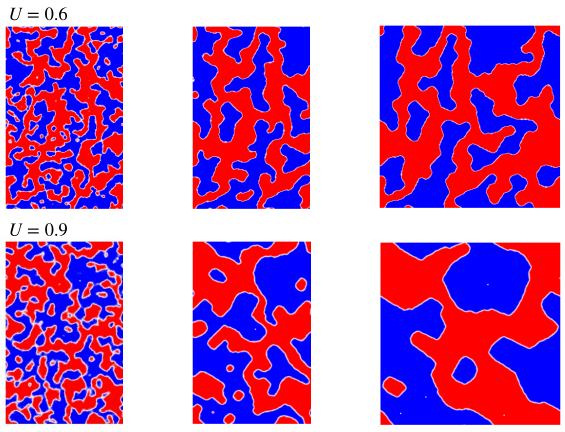
The dynamical behaviors of complex mesoscopic structures are not only of fundamental interest, but also crucial to the understanding of novel material functionalities. For example, the properties of the electronic liquid crystalline states are believed to play a crucial role in the emergence of high temperature superconductivity. Another case in point is the dynamical control and manipulation of complex spin textures by electrical means, which are central to the success of the spintronics technology. Large-scale dynamical simulations enabled by ML force-field models will shed new light on the dynamics of complex electronic textures, which in turn will lead to valuable insights on the design and engineering of these functional electronic systems. Finally, the ML

framework developed in this project is expected to open new frontiers in the research of correlated electron materials and other complex electronic systems.

Recent Progress

ML force-field models for itinerant electron magnets and spintronics: Over the past few years, our group has been developing ML frameworks to enable large-scale dynamical simulations of itinerant electron magnets. While the fundamental ideas are similar to the ML force-field models for *ab initio* molecular dynamics (MD) [1], one crucial difference is the proper symmetry-invariant representation of a local magnetic environment, which we called a magnetic descriptor. The magnetic descriptors must account for the symmetry in both the spin-space and the real-space. For lattice systems, the real-space symmetry is described by point groups associated with the lattice site. We have formulated a general theory centered on the group-theoretical methods for the magnetic descriptor. Explicit implementation of magnetic descriptors is demonstrated for three types of itinerant magnets: the metallic spin glass, Kondo-lattice model, and spin-orbit-coupled chiral magnet. In particular, we have applied the ML force-field models to investigate for the first time the crystallization dynamics of skyrmions in itinerant magnets.

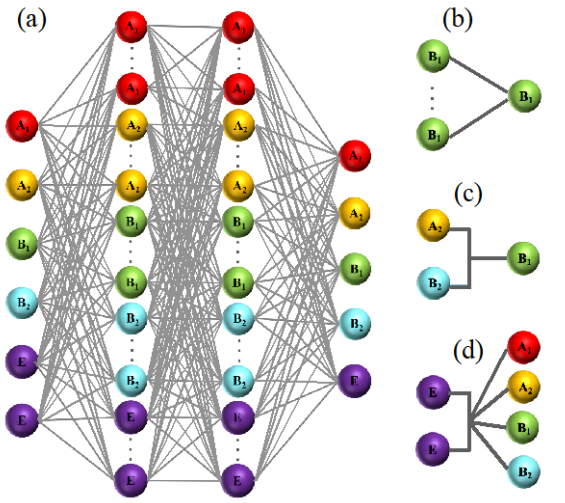
ML force-field models for strongly interacting electron lattice systems. In several of our previous works, we have shown that ML force-field approaches can be successfully applied to several well-known lattice models (double-exchange, Falicov-Kimball, Holstein, etc). These systems are characterized by a nontrivial interaction between classical dynamical degrees of freedom and free electron system on a lattice. To go beyond lattice models with free electrons, we have also developed ML models for correlated electron systems with Hubbard interactions. In particular, building on our work on ML application to the Holstein model, we have recently generalized the approach to enable large-scale adiabatic dynamical simulations of the Holstein-Hubbard model on a square lattice. The Gutzwiller/slave-boson method coupled with real-space exact diagonalization were used to generate the training dataset. Our large-scale thermal quench simulations uncover an anomalous growth of the CDW domains that deviates significantly from the expected Allen-Cahn law for phase ordering of Ising-type order parameter field. Our work demonstrates, for the first time, how kinetics of phase ordering in correlated electrons can be affected by Hubbard U. We observe an intriguing non-monotonic dependence of CDW coarsening on the Hubbard repulsion U, indicating nontrivial interplay between electron correlation and CDW dynamics.



Coarsening of CDW domains at two different U in the Holstein-Hubbard model.

ML force-field models for non-conservative and nonequilibrium forces: Partly due to symmetry considerations, conventional ML models for quantum MD simulations are designed to predict the system energy [1]. Atomic forces are then obtained from derivatives of the energy. As a result, the conventional ML methods cannot be used to describe non-conservative forces which arise in out-of-equilibrium electron systems. Although the ML modeling of general nonequilibrium forces is a challenging open problem, we have formulated a general ML approach to both conservative and non-conservative effective fields for spin dynamics. Applying this formulation to a driven double-exchange system, we showed that the generalized ML force-field model not only can accurately predict electron forces computed from the non-equilibrium Green's function (NEGF) method, LLG simulations based on our new ML-potential also successfully reproduce the voltage-induced insulator-to-metal transition obtained from NEGF-LLG simulations.

ML force-field models based on Equivariant Neural Networks. As discussed above, a crucial requirement for a faithful ML model is to preserve symmetries of the original physical systems. In conventional Behler-Parinello type ML schemes, this is accomplished through a proper symmetry-invariant representation of local environment called the *descriptor*. In the past few years, we have formulated a general theory of descriptor for condensed-matter systems. A properly designed descriptor can then be combined with a conventional fully connected neural network to form a ML model. A novel alternative approach is to incorporate symmetry properties directly within the neural net itself. Such so-called equivariant neural networks (ENNs) are characterized by the properties that neurons at each layer follow specific transformation rules of the governing symmetry groups [2]. One way to achieve this is to make sure that each node in ENN belongs to a specific irreducible representation (IR) of the symmetry group; see Figure on the right for an example. We have recently implemented such a scalable ENN force-field model that incorporates the D_4 point group of the square-lattice Holstein model and applied it to study large-scale coarsening dynamics of CDW.



(a) Schematic diagram of fully connected equivariant neural network. (b)-(d) Inter-layer connections are governed by IR multiplication rules.

Future Plans

Building on our ML models for itinerant electron magnets, we plan to investigate large-scale dynamical behaviors of skyrmion systems and their emergent collective behaviors. These include, for example, the crystallization and melting dynamics of skyrmion lattices (SkL) either driven by temperature or current. Preliminary results from thermal quench simulations indicate

unusual freezing behaviors that obstruct the crystallization of skyrmions. To study dynamical phenomena driven by an applied current, we are currently applying the ML framework for non-conservative forces discussed above to provide an accurate and efficient approximation of spin-transfer torques (STT). The NEGF-LLG method has been developed for a Rashba s-d model to generate datasets for training a generalized ML force-field model, which already shows promising preliminary results. We are currently working on scale up the ML model to large system sizes.

In addition to providing an elegant solution to the symmetry requirements, ENN also offers an approach to model non-conservative forces for out-of-equilibrium electron systems. This is because the output nodes of ENN can be directly designed to produce driving forces, thus bypassing the local energy required in the conventional BP-type ML scheme. Encouraged by our works of equivariant neural network (ENN) for the Holstein model, we plan to develop ENN-based ML force-field models for other condensed-matter systems with more complex dynamical variables, in particular for itinerant magnetic systems and spintronics applications.

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Quantum Dynamics in Anisotropic Magnets

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Keywords: bond-dependent interactions, quantum magnets, dynamics, spin-nematic, transport

Research Scope

The primary goal of this program is to advance theoretical understanding of the emergent phases in the novel quantum magnets, which form a rapidly growing and diverse group of materials that demonstrate a remarkably broad spectrum of complex quantum phenomena. Our research is focused on demystifying enigmatic quantum selection of the non-classical ground states in quantum magnets with strongly-anisotropic bond-dependent interactions, on their unconventional excitations, and on their dynamical and transport properties. Our effort can be expected to yield predictions of significant new effects and result in a deeper understanding of a large class of natural and synthetic materials, offering crucial insights into their potentially transformative properties, while also providing critical theoretical modeling of experiments and guiding future developments in this field of study.

Recent Progress

Novel quantum phases in the honeycomb-lattice ferro-antiferromagnets.–Novel quantum phases are found to emerge in a model that is directly relevant to a large group of new materials.

The significant role of quantum effects in magnets with competing interactions remains at the forefront of condensed matter physics for over 50 years, inspiring a multitude of quests for exotic states, models that can realize them, and real materials that can host them. The elusive spin-liquid states with strong entanglement are but one example; others include valence-bond phases with spatial symmetry breaking, quantum spin nematics that are quantum analogues of liquid crystals, and an especially extensive class of unconventional magnetically ordered phases that do not appear in the classical solutions of the underlying spin models. It is the latter group of phenomena that creates a broader context for the continuing research effort of our group concerning the search for and an identification of the novel ground states of quantum magnets.

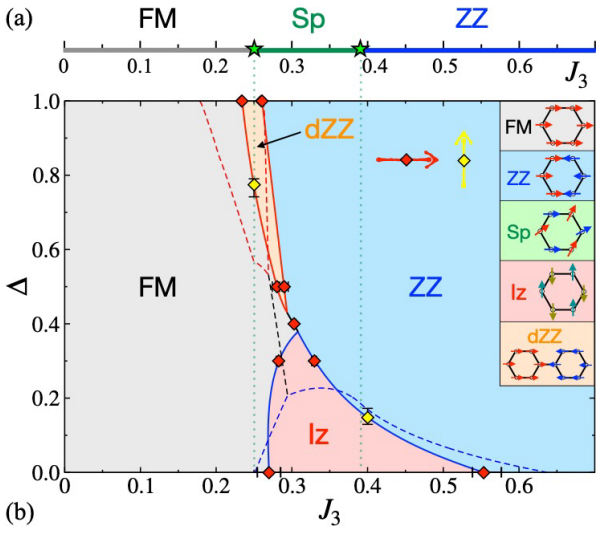

Figure 1. From [1]. (a) Classical and (b) quantum phase diagrams of the J_1–J_3 honeycomb-lattice ferro-antiferromagnetic model. Novel double-zigzag and Ising-z phases supersede classical spiral phase.

In the highlighted work [1], we have studied one of the paradigmatic models in quantum magnetism that is also attracting a significant

recent interest because it appears to be providing a tantalizingly close description for many of the newly synthesized materials of the cobalt and ruthenium families. We have demonstrated that the phase diagram of the quantum ferro-antiferromagnetic model on the honeycomb lattice differs dramatically from the classical one. It hosts the double-zigzag and Ising-z phases as unexpected intermediaries between ferromagnetic and zigzag states that are also extended beyond their classical regions of stability, see Fig. 1. In broad agreement with quantum order-by-disorder arguments, these collinear phases completely supersede the classical spiral state. It is established beyond any reasonable doubt that the two unconventional quantum phases occupy a significant portion of the phase diagram of this important model. The proposed minimally-augmented spin-wave theory is advertised as an effective tool to explore quantum phases beyond their classically stable ranges at a fraction of the numerical effort [1].

Our study [1] provides a much-needed framework to this area of research, creates a foundation for future studies of a large group of materials with anisotropic exchanges, connects and extends different approaches used in this broad field of study, clears the path to a consistent interpretation of the current and future experiments, and gives important new insights into fundamental properties of quantum magnets with spin-orbit-generated low-energy spin systems.

Magnon interactions in the quantum paramagnetic phase of $CoNb_2O_6$.–The strongly-anisotropic quasi-one-dimensional ferromagnet $CoNb_2O_6$ in transverse field has provided a spectacular realization of the magnon decay effect in its paramagnetic phase.

In our recent work [2], a self-consistent theory has been proposed to regularize unphysical divergences while preserving the integrity of the singular thresholds of magnon decay, demonstrating quantitative agreement with the neutron-scattering results throughout the field regime inaccessible by the standard spin-wave theory. Moreover, our results for the spectrum gap are in a close accord with the DMRG calculations for the same model parameters, see Fig. 2.

In this study, we have also reanalyzed the symmetry-based approach to formulating anisotropic-exchange model of $CoNb_2O_6$ and proposed its connection to the broader class of such models, studied for a wide variety of materials with complex bond-dependent exchanges. We have also clarified the role of a phenomenological constraint that has been used to restrict parameter space of $CoNb_2O_6$, and investigated the effects of the residual terms, neglected in the previous studies, using real-space perturbation theory and unbiased DMRG approach. The effects of longitudinal fields were explored as well.

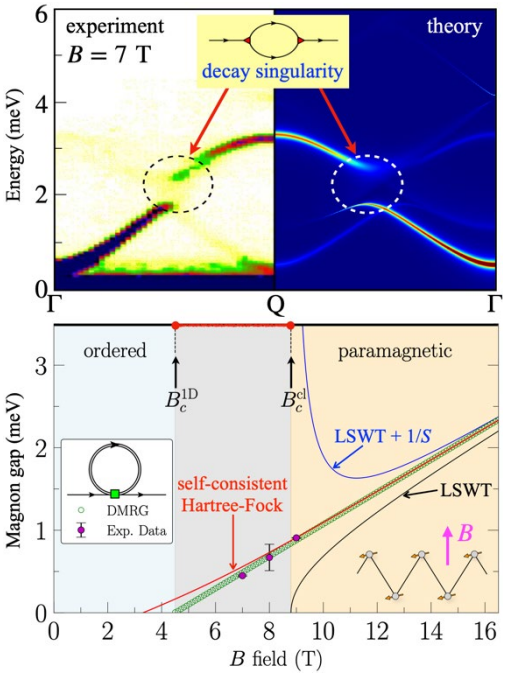

Figure 2. From [2]. INS experiment and theory results for the dynamical structure factor of $CoNb_2O_6$. Spectrum gap vs field by our theory, DMRG, and experiment.

Bond-dependent anisotropy and magnon decay in cobalt-based Kitaev triangular-lattice antiferromagnet.–Following tremendous success of the graphene-derived research, the magnetic van der Waals materials that can be cleaved into monolayer two-dimensional atomic crystals have emerged as a new platform in the studies of low-dimensional physics and in the design of artificial heterostructures with novel properties. At the same time, the search for the Kitaev-like bond-dependent anisotropic interactions, induced by the strong spin-orbit coupling, has evolved from the compounds of the *5d* and *4d* transition metals, to that of the *3d* group, with Co^{2+}-based materials producing a number of promising candidates for the realization of an exotic physics.

While many Kitaev candidate materials order magnetically due to additional interactions, their bond-dependent anisotropy manifests in the unusual spin dynamics. In our study [3], we reported on the bond-dependent interactions in the triangular-lattice cobalt-based van der Waals magnet CoI_2. An extremely rich spin dynamics was uncovered [3]: the momentum and energy-resolved inelastic neutron scattering measurements in CoI_2 show substantial magnon decay and level repulsion. A thorough examination of excitations in the paramagnetic and magnetically ordered states has demonstrated that the Kitaev-like bond-dependent anisotropy is the origin of both the noncollinear magnetic spiral order and the magnon decay effect found in CoI_2.

Moreover, the decays and avoided decays are observed over the broad momentum space. These phenomena originate from the strong magnon-magnon interactions due to bond-dependent anisotropy and nontrivial magnetic order. Our findings are essential for understanding complex bond-dependent anisotropy and noncollinear magnetic order in quantum magnets and provide the basis for future studies of the interplay between Kitaev magnetism and geometric frustration.

The elusive spin nematic.–Liquid crystals–which combine properties of a liquid and a solid that seem mutually exclusive–were considered an exotic state of matter for nearly a century before becoming ubiquitous in technology. Their quantum analogues have been hypothesized and pursued in several contexts, such as electronic nematic states in strongly correlated materials, spin nematics in frustrated magnets, and supersolids in He^4 and cold atomic gases. Quantum spin nematics are particularly elusive, as they should interpolate between a magnetically ordered spin solid and a spin liquid, another exotic and elusive state. Like spin liquids, spin nematics lack conventional dipolar magnetic order, but instead break spin-rotational symmetry with a higher-rank multipolar ordering, making their experimental detection challenging.

In our study [4], the existence of the *d*-wave nematic phase in the phase diagram of the paradigmatic ferro-antiferromagnetic J_1–J_2 model was firmly established using analytical and DMRG insights. A nematic state occurs if a dipolar magnetic order due to a BEC of single spin flips is preempted by a BEC of their pairs. In the 2D J_1–J_2 model, the nodes of the $d_{x^2-y^2}$ harmonics avoid magnon band minima at the non-trivial **Q**-points, rendering other channels unfavorable and providing a stable solution for an arbitrarily weak attraction, as for the *s*-wave Cooper pairing, elucidating broader criteria for the existence of spin nematics.

The two most dramatic results of our work are the switch of the *pair*-repulsion, which is necessary for the formation of the stable nematic condensate, to the *pair*-attraction, leading to a collapse of the nematic state in zero field, and a suppression of the single-spin-flip gap by an

attraction to the pair-condensate, which leads to a drastic order-of-magnitude contraction of the nematic phase compared to the naive expectations. The hallmark of the nematic phase is the sharp drop in the magnetization in a narrow field range near saturation without any dipolar order.

Our work [4] gives vital guidance to the theoretical and experimental searches of the elusive quantum spin nematics, arming them with realistic expectations. Our phase diagram applies to a variety of models and materials, suggesting important directions for future research.

Future Plans

For the following years, our studies of the quantum phenomena in anisotropic-exchange magnets will focus on their novel phases and dynamical properties. Specifically, we plan:

- To expand our recent successful insights onto significant novel properties of the cobaltite family, providing a generic approach to identifying their microscopic spin models.
- To expand our recent understanding of the quantum spin nematics, with the goal of a comprehensive exploration of the nature of the bound states in them.
- To work out universal rules for the non-reciprocal spin excitations in quantum magnets.
- The easy-axis triangular-lattice systems show unexpected ground states and dynamical properties. We are currently working on their phase diagram and dynamical responses.

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Machine-Assisted Quantum Magnetism

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Keywords: Machine learning, quantum magnetism, quantum spin liquid, skyrmions, X-ray scattering

Research Scope:
This project combines the scientific domains of quantum magnetism using a theoretical model, high-performance computing, and multimodal experimental (X-ray photon fluctuation spectroscopy, ultrafast X-ray and electron diffraction, and THz spectroscopy) workflows to achieve real-time, machine-assisted control and analysis of x-ray scattering studies at LCLS to significantly reduce the time to discovery of new exotic quantum states. Specific materials include (1) skyrmions in systems, such as Fe/Gd multilayers and CrI_3; (2) the pyroxene family of 1D Mott insulators and 1D-cuprate $CuGeO_3$; and (3) quantum spin liquid states in $RuCl_3$/graphene heterostructures. The project will give insight into areas of spontaneous fluctuations, quantum criticality, etc.

Our research goal stands on five interlinked pillars: **(1)** First-principles modeling to identify suitable candidate materials and the associated effective model Hamiltonians; **(2)** Higher-level theory for accurate treatment of electron correlation effects; **(3)** Development of ML models to identify the correct model Hamiltonian; **(4)** Experimental work on the most promising candidate materials and to verify the theory; **(5)** Development of software tools to build a platform for real-time control of experiments guided by theory, modeling, and ML.

Recent Progress

Highlights are as follows. (1) An ML model based on simulated spectra was built to predict Hamiltonian parameters by adapting neural implicit representations from computational photography to model inelastic scattering data. The model was tested on new data on La_2NiO_4.[1,4] (2) As key steps toward achieving real-time steering of experiments: (a) Feasibility of capturing and analyzing single-shot X-ray data at LCLS using ML algorithm was demonstrated;[2] (b) An ML-enhanced Bayesian approach to guide measurements via theory simulations was developed; and (c) a convolutional neural network for faster analysis of sparse soft X-ray speckle data was studied.[4] (3) Progress made toward deploying ML tools in our materials studies includes: (a)

Impurity bands in heavily doped semiconductors was elucidated via a supervised deep learning method and a convolutional neural network; (b) Datasets from three different light sources were combined and analyzed via a variety of simulations to understand magnetism of $NiPS_3$;[3] (c) A high-performance algorithm was implemented for atomistic spin dynamics simulations for modeling skyrmion lattices;[5] (d) Materials studied with interesting magnetic states include, helical spin ordering in room-temperature metallic antiferromagnet Fe_3Ga_4,[6] topological phases of MnA_2X_4 (A=Bi, Sb; X = Se, Te),[7,8] spiral magnetic order in SmAlSi,[9] stripe helical magnetism in NdAlGe,[10] and chiral fluctuations in Kagome Lattice.[11]

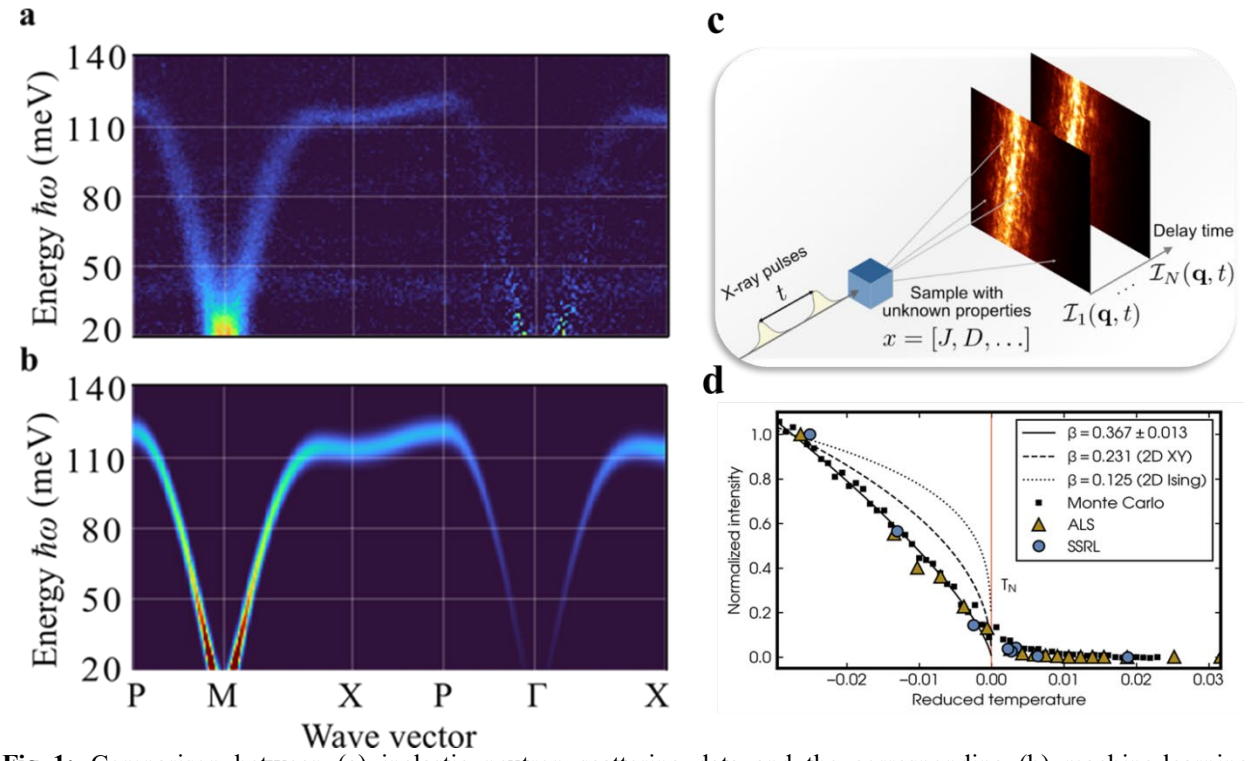

Fig 1: Comparison between (a) inelastic neutron scattering data and the corresponding (b) machine-learning prediction.[1] (c) A schematic illustration of machine-learning enabled Bayesian experimental design framework. Speckle pattern shown in the left panel is the magnetic scattering from a van der Waals magnetic system at the Ni K-edge.[2] (d) Integrated intensity obtained at SSRL (blue dots) and ALS (brown triangles), normalized to the values at T=0.97×TN, shows their functional form as they approach the magnetic phase transition. Corresponding theoretical predictions are shown.[3]

Future Plans

The progress outlined above provides a firm foundation for implementing our vision of harnessing data-science tools for real-time steering of beamlines and for advancing the understanding of quantum magnetism in complex materials. Major activities proposed are: (1) Continuing to adapt and deploy data-driven tools that are especially tailored for handling experimental spectroscopic data; the time required to return results will be benchmarked to assess the viability of our tools for handling large volumes of live data at high repetition rates; (2) Persisting to build, test, and deploy

ML-driven tools for rapid determination of coupling constants for spin Hamiltonians describing quantum magnets, including Kitaev and DMI interactions, as well as other non-trivial couplings (e.g. electron-phonon, magnon-phonon, etc.), and for handling complex ground and excited states and their dynamics. Automated generation of Wannier functions and fine-grained band structures needed for accurate calculation of response functions will also be pursued. (3) Integrating theory/computation focused on model Hamiltonians with experimental data in a tight feedback loop using ML algorithms. Utilizing ML models trained with simulated data during ongoing experiments, we will actively refine the model details and enhance prediction accuracy as more experimental data is collected. This will increase the accuracy of the model and its predictive capabilities over time to enable real-time experimental control. Although we have explored a variety of materials with interesting magnetic states (e.g. helical spin ordering in Fe3Ga4, spiral magnetic order in SmAlSi, and helical magnetic stripes in NdAlGe), our ongoing work suggests that the van der Waals magnets will provide attractive materials family for investigating exotic magnetic states with our platform for real-time steering of experiments.

Broader Impacts and Workforce Development: The project trains undergraduate, graduate, and postdoctoral students to pursue theoretical and experimental research in quantum materials, quantum information sciences, and data-driven analysis. A special feature of this project, due to its location at Howard University, which is a leading HBCU, is that it directly impacts the training of underserved communities for careers in the quantum workforce for the industries of tomorrow. The project is also helping to improve the quantum infrastructure at Howard University, which will continue to impact the university's ability to provide quantum training to minorities long after the project has ended.

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Superconductivity and Competing Orders in Correlated Electron Systems

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Keywords: Superconductivity, nematic fluctuations, spin/valley orders, van Hove singularities, phase fluctuations

Research Scope

My current research (2023-2024) is along three directions.

The first is the analysis of the role of orbital physics in pure and doped FeSe. My main goal here is to understand the origin of nematic order in pure and doped FeSe and the interplay between nematic order and superconductivity, particularly near a nematic quantum-critical point (QCP). Recent experiments clearly indicated that superconducting state near a nematic QCP is quite different from that in pure FeSe and speculated that the pairing glue can be different. Besides, there is strong evidence from specific heat measurements that superconducting state near a QCP is highly unconventional.

The second is the analysis of valley physics in twisted bilayer graphene and in biased Bernal bilayer graphene (BBG) and rhombohedral tri-layer graphene (RTG). My current goals here are (i) to understand the sequence of spin and valley (isospin) ordered states, observed in experiments, (ii) explain experimental data showing two seemingly incompatible features: presence of soft collective modes near an isospin order and strong first order transition into a state with maximal possible order, and (iii) explain superconductivity observed near the onset of valley polarization in the presence of either a finite magnetic field or Ising spin-orbit interaction induced by a monolayer WSe2.

The third is the analysis of response functions in a superconductor with pairing by nematic fluctuations. My current research in this direction is on (i) development of a novel critical Fermi liquid theory near a nematic QCP, in which the divergence of the effective mass is compensated by the divergence of non-nematic components of the quasiparticle interaction function, (ii) on the comparative analysis of response functions in superconductors mediated by spin fluctuations and nematic fluctuations, and (iii) on the applicability of Eliashberg theory for nematic-mediated superconductivity and for normal state with strong nematic fluctuations.

Recent Progress

I will focus on the work on superconductivity due to nematic fluctuations, which I will present at the meeting. With my student K. Islam, we performed a comprehensive analysis of superconductivity in a multi-orbital fermionic system near the onset of a nematic order. For doped FeSe we associated the nematic order with spontaneous polarization between d_{xz} and d_{yz} orbitals. The first question we addressed is how one can get an attractive pairing interaction mediated by nematic fluctuations given that nematic fluctuations are charged fluctuations peaked at zero momentum transfer q. A bare pairing interaction mediated by small q charge fluctuations is repulsive. We found, however, that beyond bare theory, there exists an attractive pairing component induced by density-density interaction between a hole and an electron pocket. We derived this pairing interaction and explicitly demonstrated that it is proportional to a nematic susceptibility and hence becomes the strongest pairing interaction near a nematic QCP. The second question we addressed is the value of superconducting Tc and the structure of superconducting gap. The source of non-triviality here is the structure of nematic-mediated pairing interaction: it not only scales with the nematic susceptibility, but its strength also depends on the position of a fermion on the Fermi surface, specified by angle θ as $\cos^2(2\theta)$. As the consequence, right at the nematic QCP, superconducting gap opens up at Tc only at special points and extends into finite arcs at T < Tc. In between, the

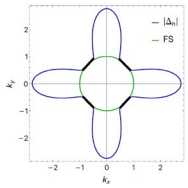

Fig. 1 Theoretical gap structure at a nematic QCP.

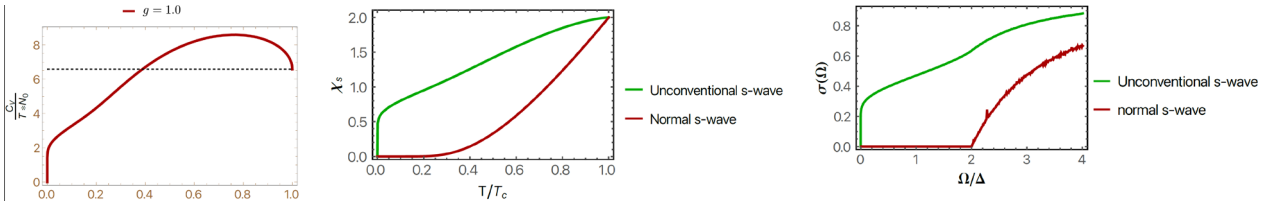

Fig. 2 Theoretical behavior of observables in a superconducting state near a nematic QCP. Left – specific heat, middle- uniform susceptibility, right – optical conductivity. The behavior of all observables is highly unconventional.

arcs the Fermi surface remains intact (Fig. 1). This gives rise to highly unconventional behavior of the specific heat, with no jump at Tc and to a finite offset at T=0, when extrapolated from a finite T (Fig. 2a). We also found a highly nontrivial behavior of other characteristics, like spin susceptibility and optical conductivity (Figs 2b,c). In addition, Tc does go down under irradiation, even when the gap symmetry is an s-wave. Away

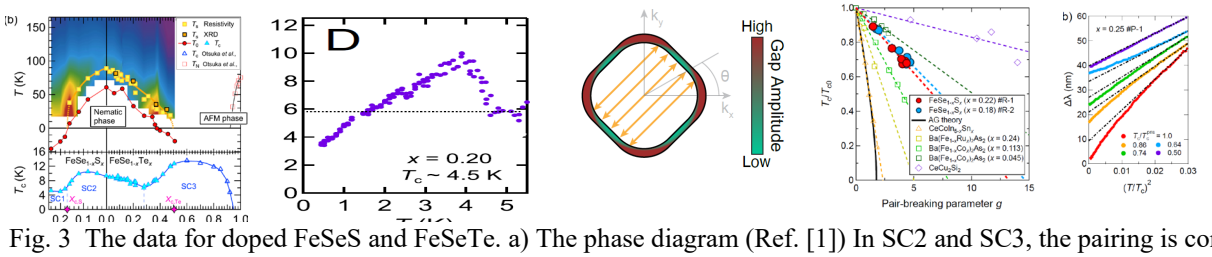

Fig. 3 The data for doped FeSeS and FeSeTe. a) The phase diagram (Ref. [1]) In SC2 and SC3, the pairing is consistent with nematic scenario; b) Specific heat data near a nematic QCP [2], consistent with Fig. 2a; c) Gap function extracted from STM data [3], consistent with Fig. 1; d-e) Data for Tc and penetration depth in irradiated samples [4]. We reproduce these results in our theory.

from a QCP, the gap emerges at Tc everywhere on the Fermi surface, except, possibly, special points, yet gap anisotropy remains.

Pairing by nematic fluctuations is an alternative to pairing by spin fluctuations, which was extensively discussed for Fe-pnictides and pure FeSe. We compared in detail the gap structure in the two pairing scenarios and argued that there is at least one qualitative difference: in nematic scenario the maximum of the gap on the hole pocket is along X and Y directions in 1Fe zone, while in magnetic scenario it is at 45° compared to X or Y. For pure FeSe, the direction of gap maximum is consistent with the magnetic scenario. However, in $FeSe_{0.81}S_{0.19}$, which is close to a QCP, recent STM study found gap maxima along X and Y, consistent with the nematic scenario (Fig. 3c). Other experimental data in Fig. 3 are also consistent with the nematic scenario.

Our microscopic theory of highly anisotropic gap function due to pairing by soft nematic fluctuations is an alternative to phenomenological theories which assume the existence of a Bogolubov Fermi surface in a superconducting state.

In another recent work on non-trivial aspects of the pairing by soft nematic-like fluctuations, my former postdoc Shang-shun Zhang (now at U. of Tennessee) and I analyzed the spectral function in a superconducting state above a nematic QCP in a one-orbital model with emphasis on the search of the effects of strong fluctuations and non-Fermi liquid behavior. A normal state at a nematic QCP in 2D is a non-Fermi liquid with no coherent quasiparticles. A superconducting order gaps out low-energy excitations, and at a first glance, restores coherent quasiparticle behavior. We argued that this is not the full story as the fermionic self-energy remains singular above the gap edge. This singularity gives rise to markedly non-BCS behavior of the density of states and to the appearance of a non-dispersing edge mode (Fig. 6). We applied the results to doped FeSe and also showed that similar behavior holds for cuprates Bi2201 and Bi2212, where ARPES experiments detected an edge mode [5]. We made a detailed comparison between theory and experiment (Fig. 4)

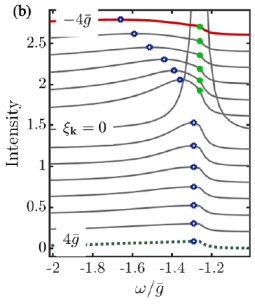

Fig. 4 Spectral function of a SC out of a NFL, with a non-dispersing mode (blue/green).

Future Plans

On nematic superconductivity, my plan is to extend the analysis to regions away from a nematic QCP, compute the observables, and compare with the data. We are also searching for smoking gun experiment to distinguish our scenario from the ones about Bogolubov Fermi surface.

On isospin order in BBG and related , my plans are to (i) analyze the origin of a nematic order, which accompany spin/valley ordered states, (ii) to develop the full Ginzburg-Landau description of the ordered state, not assuming that the order is weak and analyze whether a strong order breaking time reversal symmetry can explain anomalous Hall effect observed in BBG, (iii) to obtain the full spectrum of collective modes, (iv) to analyze superconductivity emerging out of the ordered states.

On response functions, my goal is to analyze qualitatively new features of a superconductor emerging out of a non-Fermi liquid and propose specific experiments to probe them.

My other plan is to obtain full understanding of superconductivity out of a pseudogap phase, which includes the interplay between pair formation and phase coherence, and apply the results to electron-doped cuprates, for which detailed ARPES data in a SC state have been reported recently. Besides, I plan to analyze in great detail how bi-polaron SC emerges out of a state pseudogap polaron state and the interplay between this theory and Eliashberg theory of superconductivity.

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Harnessing Quantum Geometry of Correlated Electrons for Next-Generation Photovoltaics

Martin Claassen, University of Pennsylvania

Keywords: Strongly-correlated electrons, quantum geometry, photogalvanic effect, nonlinear optics

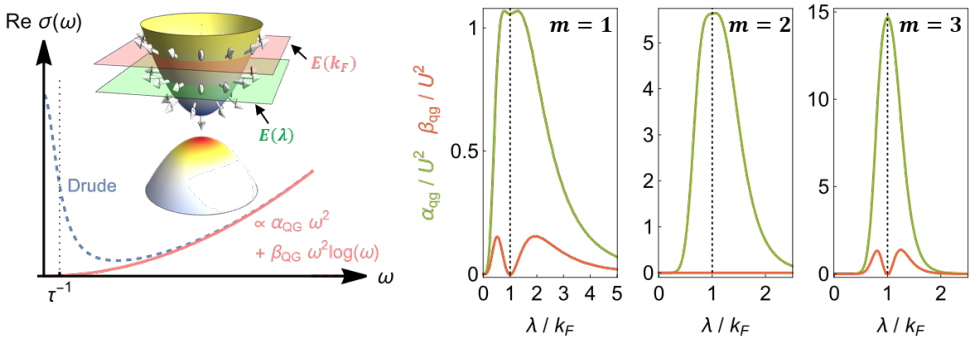
Research Scope

This project focuses on devising a theoretical and computational foundation for emergent non-linear optical and photogalvanic responses in strongly-correlated quantum materials, to probe and harness the interplay between strong electronic interactions and the quantum-geometric structure of electronic wave functions that couple to light. In recent years, connections between optical responses of band insulators and quantum-geometric properties of the material's free-electron Bloch wave functions have become increasingly well understood [1]. However, thus far, no analogous insight exists for Mott insulators and strongly-correlated electrons. Such systems are most naturally addressed from a real space perspective of charge localization in Wannier orbitals subjected to strong screened interactions, with their shape, extent and coupling to light playing a dual role to the usual momentum-space quantum geometry [2]. However, non-linear and photogalvanic responses from the quantum many-body behavior of electrons in correlated quantum materials, as well as a microscopic understanding of the emergent coupling of photons to low-energy excitations in correlated materials, remain a key open theoretical and methodological challenge. A central goal of this project is the development of a theoretical framework and computational foundation for studying quantum-geometric optical responses of correlated electrons and Mott bulk photovoltaics, materials that are insulators due to strong Coulomb repulsion between electrons but can rectify light into electrical current. Key objectives include predicting many-body photogalvanic effects in moiré materials and low-dimensional Mott insulators, devising quantum-geometrical guiding principles for photogalvanic responses in correlated and topological van der Waals heterostructures, as well as the development of new computational tools which combine first principles modelling with many-electron simulations to drive the discovery of strongly-correlated materials with useful non-linear optical and photogalvanic responses.

Recent Progress

Since the start of this project in the fall, we pursued a two-pronged approach: First, a key objective is the development of a framework to describe *emergent* quantum-geometric contributions for non-linear optical and photogalvanic responses of interacting electrons. Conventional low-energy theories of correlated electrons rest on effective Wannier orbital tight-binding descriptions of the active strongly-interacting electronic bands in solids. However, a microscopic description of their electromagnetic response at frequencies ω well below interband transitions Δ necessarily requires carefully integrating out virtual interband transitions (out of the manifold of active bands) in the presence of strong Coulomb interactions. In a first work in this project, we have shown that this

leads to a new, emergent, and purely quantum-geometric contribution to the linear optical conductivity of correlated metals near a topological band inversion, which can play a crucial role in the finite-frequency incoherent response for small Fermi surfaces at low temperatures. We find that this effect originates from an emergent photon-induced scattering of electron *pairs*; it enters as an adiabatic contribution from self-energy diagrams and vertex corrections involving interband processes, and appears in addition to real interband excitations that are suppressed by ω/Δ. Usefully, it can be equivalently understood as an interaction-mediated response to light due to the imperfect localization of the real-space Wannier functions of the band that hosts the Fermi surface. Near a time-reversal-invariant topological band inversion or its higher-angular-momentum generalizations in 2D, we find that dilutely-doped correlated metals can reach an almost-Galilean-invariant "chiral limit" with parabolic bands, for which the optical conductivity becomes a purely quantum-geometric quantity. The finite-frequency optical conductivity scales $\sim \omega^2$ and is drastically enhanced if the orbital character of Bloch states changes substantially across the Fermi surface; in addition, a logarithmic correction $\sim \omega^2 \log(\omega/\varepsilon_F)$ probes the Berry phase of the Fermi surface. Our results predict a new THz signature of the interplay of quantum geometry and electronic correlations in moiré materials and topological metals, while providing a foundation for diagrammatic studies of interaction-mediated quantum-geometric non-linear and photogalvanic responses, to be pursued next.



Quantum-geometric optical response of two-dimensional correlated metals near a topological band inversion with angular momentum m.

In parallel, we are investigating the role of quantum-geometric bounds of Wannier function extents in governing optical and non-linear responses in Mott insulators or strongly-frustrated moiré materials [3]. In systems with spin rotation symmetry, the electric field can formally couple to magnetic excitations via the scalar spin chirality, however, such couplings are theoretically expected to be strongly suppressed in linear THz responses or Raman scattering in the A_{2g} channel [4]. In current work, we show that this ceases to apply to Mott insulators that form from imperfectly-localized Wannier orbitals, leading to a quantum-geometric scalar spin chiral coupling that grants access to chiral magnetic fluctuations at leading order in t/U mediated via emergent photon two-electron scattering.

Complementarily, we have implemented a matrix-product-state-based approach to compute the photogalvanic response for strongly-correlated electrons using Chebyshev expansions and DMRG. We find that the photocurrent for interacting electrons can be compactly expressed in terms of a

many-body shift vector, that, for the linear photogalvanic response, can be recast in dipole gauge into a two-sided expansion in terms of matrix product state Chebyshev vectors that can be conveniently evaluated via recurrence relations. The computational effort for the nonlinear photocurrent matches the usual computation of linear responses. Benchmarking this computational tool for the Rice-Mele-Hubbard model, we are studying its photovoltaic response in Mott and charge-transfer insulator regimes, and are investigating a generalization of our non-linear kernel polynomial approach to circular photogalvanic responses.

Future Plans

Continuing the directions above, we will pursue three near-team goals: (1) We will study the photogalvanic response in interacting flat bands of moiré heterostructures, as a probe of non-trivial quantum geometry and a design principle for enhanced photovoltaic effects. Formally, a diagrammatic extension to non-linear responses yields new quantum-geometric triangle-diagram contributions from emergent photon two-electron scattering, which enter as a generic correction for correlated electrons but are expected to dominate the non-linear responses in almost-flat moiré bands. We are especially interested in photogalvanic responses of correlated insulating phases at optical frequencies across the monolayer gap in twisted transition metal dichalcogenide bilayers, as a potentially sensitive probe of Wannier obstructions. (2) We plan to study the many-body circular photogalvanic effect (CPGE) in correlated insulators with non-trivial quantum geometry, to shed light on a qualitatively distinct contribution arising from the non-commutativity of band-projected many-body dipole operators in analogy to Landau level projections, which can drastically enhance CPGE in topological Mott phases. (3) In collaboration with Angel Rubio's group, we will develop a td-DFT-derived time-dependent Wannier basis to determine emergent light-matter interactions for correlated electrons in the material's active bands *ab initio*, providing a microscopic foundation for computing non-linear optical and photogalvanic responses using many-body methods. Combining first principles modelling with many-electron simulations, a central goal is to drive the discovery of strongly-correlated materials with enhanced non-linear optical and photogalvanic responses.

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Spin and Orbital Physics in Novel Correlated Materials

Piers Coleman, Rutgers, The State University of New Jersey.

Keywords: Twisted bilayer Graphene, Moire materials, Fractionalization and Topology, Kondo insulators

Research Scope

The current research involves three thrusts, a new local moment approach to moiré materials inspired heavy fermion physics; an ongoing development of the triplet resonating valence bond approach to iron-based superconductors and a study of fractionalization and topology in Kondo insulators.

Recent Progress

We have carried out[1] an extensive study of the effects of valence fluctuations within a topological mixed valence model of twisted bilayer graphene. Our approach builds on the work of Song and Bernevig[2], seeking to understand the evolution of the localized f-electron states with interaction. Our study of this problem emphasizes the role of the back-gate in the physics, demonstrating that the existence of the tight localization of charge inside the moiré unit cells creates a modulated binding potential, created by the image charges in the back gate. This effect stabilizes local moment behavior. Utilizing a auxilliary-rotor mean-field theory[3], we formulate a single model of interacting MATBG that captures physics across all filling factors. In our treatment, signatures of two electron fluidity emerge with widely separated Kondo ordering temperature when hybridization turns on, and coherence temperature when heavy quasiparticles become quantum.

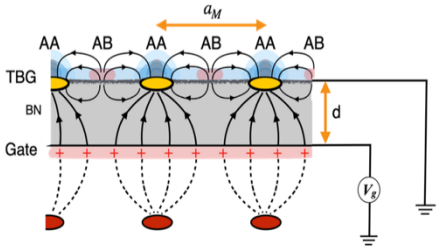

Figure 1. Schematic of the electric field lines for gated TBG in the Image Charge

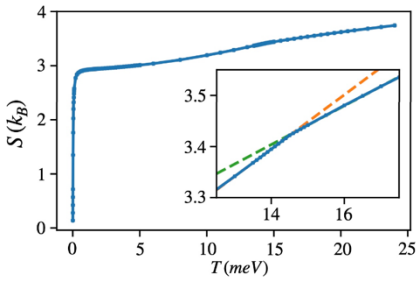

Figure 2. Two fluid behavior in our theory of TBG. Though the topology develops at high temperatures, the f-states quench at a much lowr temperature, giving rise to a thermalized flat band over a wide range of temperature.

Finally, As part of our work on possible fractionalization in Kondo lattice systems, we have developed an analytic method of calculating the vison gap which stabilizes a Z_2 spin liquid[4].

Future Plans

We have been developing a model for the spin-polarized tunneling of topological edge states in STM studies with a Kondo insulator nanowires of SmB_6[5]. Since topological edge states are

not Wannier localizable, naively we would not expect them to transmit through a point STM tip. Tentative results suggest that the tunneling results from odd-frequency magnetism that develops in the STM tip.

We plan to extend our heavy fermion approach to moire graphene to multi-layer systems, and we hope to examine whether the observed two-fluid physics in our mean-field theorycan be used as a basis to understand whether scattering off these flat bands could be an origin of the strange metal behavior seen in these compounds. We will also extend the auxiliary rotor we have employed to model the full doping range of twisted bilayer graphene, to incorporate superconductivity by allowing the rotor to move in the full 3 dimensions of charge space.

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MemComputing phenomena in correlated systems

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Keywords: MemComputing, long-range order, correlated systems, Machine Learning

Research Scope

The current research effort is to understand and take advantage of the long-range order (LRO) of memcomputing machines [1] to compute the properties of correlated systems efficiently. This requires an RG-type analysis of time non-locality which induces spatial and temporal LRO in the system. With this work we are then able to better tailor this new computing paradigm for the study of many-body systems.

Recent Progress

We have worked in two parallel directions. In one, we have suggested the transformer quantum state as a versatile machine learning model for quantum many-body problems. This opens up the possibility to generate the entire phase diagram within a single model. In the other direction, we have analyzed the long-range order in some experimentally realized neuromorphic systems to understand the role of memory in generating it. We have found that time non-locality induces phases of long-range order even in systems coupled locally and in the absence of criticality. This phenomenon is at the core of the efficiency of memcomputing machines and it has implications in our understanding of the brain dynamics.

Future Plans

We are planning to combine the transformer quantum state with memcomputing to accelerate even further the calculation of correlated many-body systems.

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Transformer quantum states. Inspired by the advancements in large language models based on transformers, we have introduced the transformer quantum state (TQS) (Fig. 1): a versatile machine learning model for quantum many-body problems. We have found that, in sharp contrast to Hamiltonian/task specific models, TQS can generate the entire phase diagram, predict field strengths with experimental measurements, and transfer such a knowledge to new systems it has never been trained on before, all within a single model. With specific tasks, fine-tuning the TQS produces accurate results with small computational cost. TQS is versatile by design, and it can be easily adapted to new tasks, thereby pointing towards a general-purpose model for various challenging quantum problems.

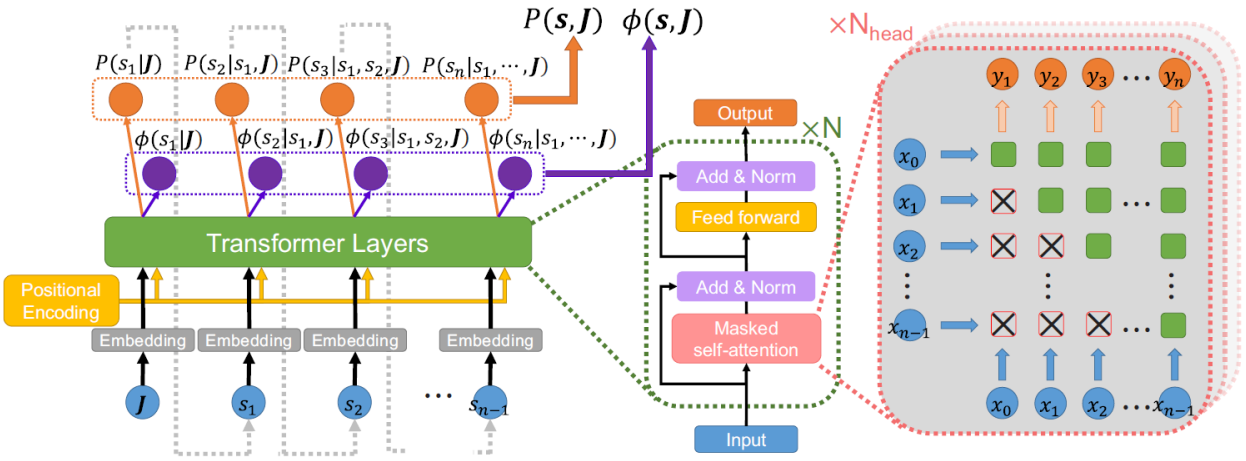

Fig. 1 Structure of a TQS. Left: the overall architecture of our model. We use the standard encoder-only transformer architecture, utilizing an embedding layer to map different inputs into a single unified feature space, and pass them through N identical transformer encoder blocks followed by two different output heads, parametrizing the amplitude P and phase φ of a quantum state, respectively. Middle: the structure of a transformer encoder block. Right: the mask structure in a masked self-attention operator. Squares with a cross represent the masks, blocking the flow of information, so that each site only has access to its predecessors. This ensures that the autoregressive property is satisfied.

Memory-induced long-range order in coupled neuristors. In the pursuit of scalable and energy-efficient neuromorphic devices, recent research has unveiled a novel category of spiking

oscillators, termed ``thermal neuristors." These devices function via thermal interactions among neighboring vanadium dioxide resistive memories, closely mimicking the behavior of biological neurons. In this research direction, we have shown that the collective dynamical behavior of networks of these neurons showcases a rich phase structure, tunable by adjusting the thermal coupling and input voltage (Fig. 2). Notably, we have identified phases exhibiting long-range order (LRO) that, however, does not arise from criticality, but rather from the time non-local response of the system. In addition, we have shown that these thermal neuristor arrays achieve high accuracy in image recognition tasks through reservoir computing, *without* taking advantage of this LRO. Our findings highlight a crucial aspect of neuromorphic computing with possible implications on the functioning of the brain: criticality may not be necessary for the efficient performance of neuromorphic systems in certain computational tasks.

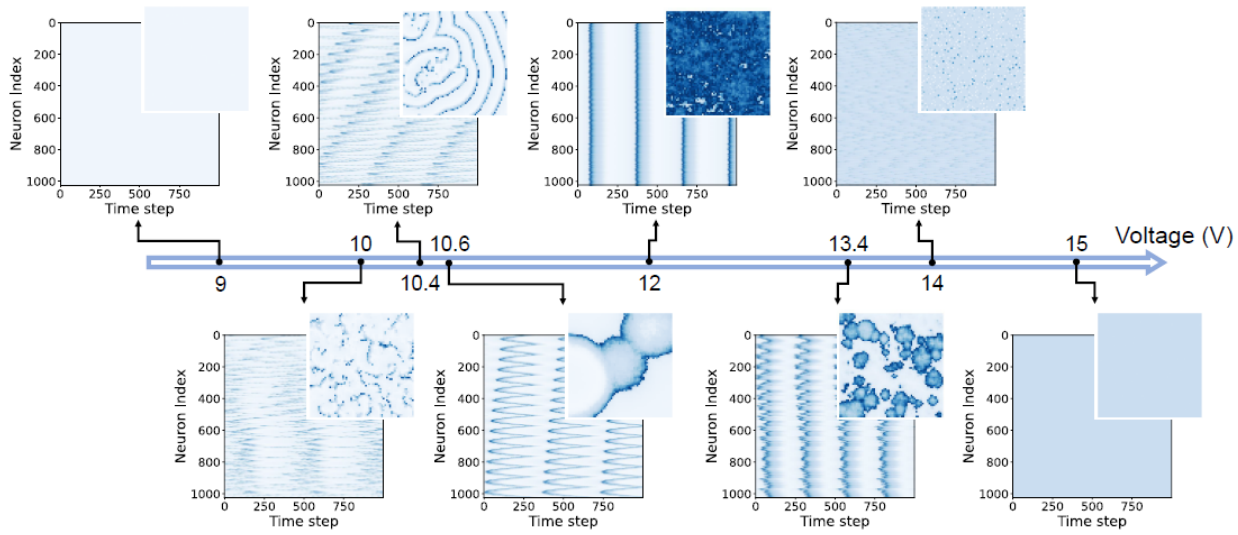

Fig. 2 Snapshots of different oscillation patterns in a 64×64 array of thermal neuristors. In each panel, color indicates current level: white signifies no current, while shades of blue denote current spikes. The main panels show collective current-time plots for the first 1024 neuristors (concatenated from the first 16 rows), and each inset captures a specific moment in the 64 × 64 array. The system exhibits no activity at very low input voltages. As the voltage increases, a sequence of dynamic phases unfolds, including correlated clusters (10 V and 13.4 V), system-wide waves (10.4 V and 10.6 V), synchronized rigid states (12 V), and uncorrelated spikes (14 V), culminating again in inactivity at excessively high voltages. The thermal capacitance is fixed at the experimentally estimated value.

Custom Design of van der Waals Materials

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Keywords: Quantum materials, intercalation, first-principles methods, machine learning, materials design

Research Scope

This project aims to pioneer the development and characterization of new and hybrid materials, focusing on 2D van der Waals[1–4] intercalated with a diverse array of intercalants, ranging from elemental intercalants like zerovalent atoms to complex conjugated and organometallic molecules (Figure 1). We envision these dynamically tunable optoelectronic materials playing a crucial role in advanced optoelectronics, where modulation of the electronic and optical properties in response to external stimuli is critical. By functionalizing these 2D-based structures through the introduction of intercalants into their intrinsic van der Waals (vdW) gap, we aim to engineer new quantum materials with unprecedented properties. Intercalation provides a pathway to modify the electronic structure, carrier mobility, and light-matter interaction of these materials, potentially leading to breakthroughs in optoelectronic device efficiency, sensitivity, and functionality. Tailoring absorption spectra and electrical conductivity through controlled intercalation opens up development avenues for novel sensors, photodetectors, and flexible electronic components.

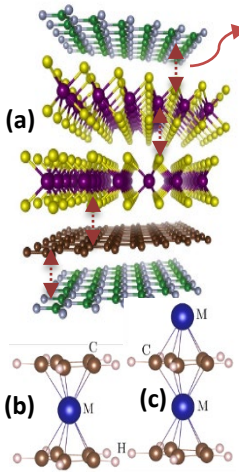

Figure 4 | Representative crystal structure of the (a) diversity of layered 2D-based materials and (b-c) metallocene, an organometallic molecule with (b) conventional and (c) double-decker configurations proposed in this program, where M=Co, Fe, Ni, Na, Li, Cr, etc.

Given the vast compositional landscape introduced by both host materials and the myriad of potential intercalants, we recognize the critical need to develop advanced machine learning (ML) techniques tailored to materials science. These data-driven solutions are essential for navigating the complex interplay between composition, structure, and properties in intercalated quantum materials. Figure 2 illustrates an active learning multiobject ML strategy using a deep neural network (DNN) to enable the simultaneous optimization of mutually exclusive properties. By leveraging materials informatics in a computational framework, we aim to accelerate the discovery and optimization of these hybrid systems, moving beyond Edisonian materials discovery. We employ a synergistic approach that combines *ab initio* materials modeling with cutting-edge ML algorithms to predict and identify new materials and optimal intercalation strategies. This methodology not only facilitates understanding the fundamental mechanisms governing the properties of intercalated van der Waals materials, but also enables efficient exploration of the material design space.

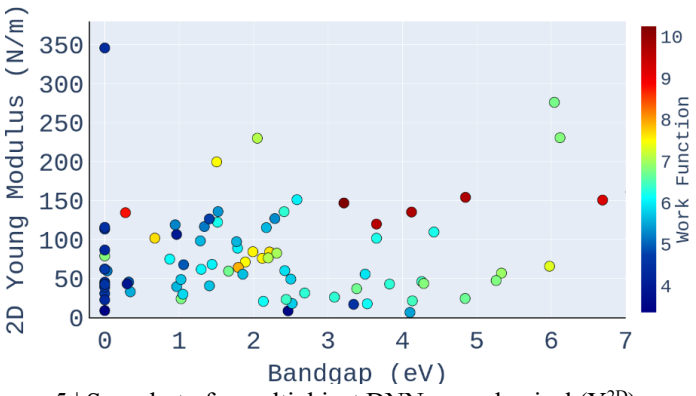

Figure 5 | Snapshot of a multiobject DNN – mechanical (Y^{2D}), energy bandgap (E_g), and the work function for 2D-based *TMCs.*

While our project will establish a general framework for the high-throughput intercalation of quantum materials focusing on 2D transition metal dichalcogenides such as MoS_2 and group-IV monochalcogenides like GeSe (TMCs) as host materials, we will undertake extensive advanced computations to characterize the properties of promising candidate materials. These advanced simulations will involve density functional theory (DFT) plus effective Coulomb interaction to account for strong correlation effects; Green's function and screened Coulomb (GW) to capture the intrinsic many-body effects in 2D-based structures; using the Bethe-Salpeter equation (BSE) to describe absorption spectra that incorporate electron-hole interaction effects; density functional perturbation theory to characterize vibrational properties and Raman and Infrared spectroscopy; dynamical mean-field theory to explore the role of intrinsic defects and many-body interactions; and other advanced computational methods essential to elucidating emerging properties.

Recent Progress

Although the project started a few months ago, we are diligently working towards our project goals.

Data-driven intercalation of 2D vdW materials. Motivated by the potential of discovering novel quantum materials for low-cost and flexible devices, as supported by the improved performance of organic semiconductor devices when integrated with 2D materials, we recently developed a "*high-throughput and data-driven computational framework for novel quantum materials"* (*under review*). The high-throughput data-driven pipeline (Figure 3) has the capability of screening large compositional spaces of intercalated hybrid materials using the intercalation energy – the energy required to insert atoms, ions, or molecules into the host matrix's van der Waals gap – as the screening parameter. Our pipeline combines DFT and active learning to efficiently identify the most stable hybrid materials, accurately identifying them by evaluating just 5% of the entire compositional space. Although the intercalation energy was the focus here, the algorithm's flexibility supports various screening parameters, enabling the exploration of large material spaces for properties such as the bandgap, work function, and magnetic moment.

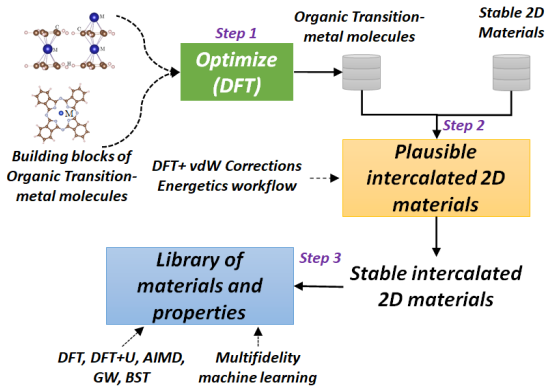

Figure 6 | Workflow for high-throughput discovery

Engineering intermediate states in quantum materials. We have computationally engineered a new class of quantum materials featuring many-body-driven quasi-localized mid-gap electronic states (Figure 4). These states are created by intercalating zerovalent Cu atoms into the vdW gap of the GeSe/SnS heterostructure. Advanced *ab initio* many-body computations reveal that these intermediate band states are driven by strong electron-electron interactions hosted by the coupling between the *s* and *p*-states of Cu and the adjacent chalcogen atoms. Our findings, detailed in the manuscript "*Chemically tuned intermediate band states in atomically thin Cu_xGeSe/SnS quantum material for photovoltaic applications*," (*in press, Science Advances*), demonstrate the potential of these materials for photovoltaic applications. Specifically, the GeSe/SnS heterostructure with Cu intercalation exhibits intermediate bands with sub-bandgap energies (~0.78 and 1.26 eV), closely aligned with the optimal values (~0.71 and 1.24 eV) for efficient intermediate band solar cells (IBSC) as per the seminal predictions of Luque and Marti.[5] Cu intercalation modified the optical spectra, introducing new low-energy states in the infrared and visible regions while eliminating energy-dissipating plasmon modes found in the pristine heterostructure. Leveraging the unique absorption

spectra from intermediate band states, we designed a prototype thin-film IBSCs, where this material acts as the primary light-absorbing layer (Figure 4c). By calculating device parameters like the external quantum efficiency (EQE) through the transfer matrix method and considering the complex refractive index derived from exciton absorption spectra, we found that the designed solar cell exhibits significant solar absorption (>90%) and an EQE (~190%) across a broad spectrum, including infrared and visible light (Figure 4d). Although these findings are based on computational analysis, they are corroborated by recent experimental work. Time-resolved ultrafast THz spectroscopy measurements by our collaborators, reported in the manuscript "*Tailoring ultrafast near band gap photoconductive response in GeS by zerovalent Cu intercalation*" (*in press, ACS Applied Materials & Interfaces*), show that Cu-intercalated GeS exhibits reduced photoconductivity, a shorter carrier lifetime, and increased mobility.

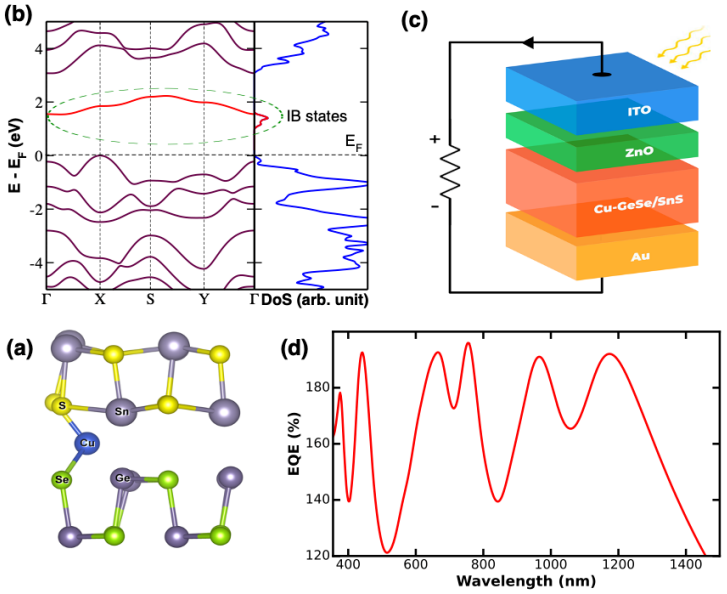

Figure 7 | Figure 1. Features of Cu_xGeSe/SnS: (a) Crystal structure. (b) Electronic structure with intermediate band states, obtained using GW method. (c) Prototype IBSC utilizing Cu_xGeSe/SnS as the active layer. (d) External quantum efficiency (EQE) of the IBSC device.

Future Plans

Work being done to identify and perform advanced calculations on promising candidate materials for advanced optoelectronic applications. We are exploring the functionalization of 2D-based group IV monochalcogenides with other elemental intercalants and the diversity of organometallic transition molecules such as the metallocene family. Simultaneously, we are refining many-body methodologies that employ effective Hamiltonians to elucidate the topological states present in these quantum materials. In conjunction with these efforts, we are integrating materials informatics with multifidelity modeling, enhanced by our application of active learning and statistical design of experiments (DoE) to maximize the extraction of information, thereby improving both the scalability and performance of our high-throughput materials screening and design process. This advanced pipeline is being generalized to accommodate a broad spectrum of 2D host materials and organic intercalants.

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Time-dependent phenomena in correlated materials

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Keywords: strongly correlated electrons, non-equilibrium phenomena, time-resolved spectroscopies, ARPES, RIXS, Auger

Research Scope: Strongly correlated electronic materials, also referred-to as "quantum materials", are systems where several phases compete producing extremely complex many-body states. Although a single phase may dominate the ground state, competing instabilities are often hidden at higher energies, which can be accessible with intense ultrafast pulses of light. Experimentalists can probe the system's dynamics by shaking it with a pulse of light, which allows them to access metastable states. These types of non-thermal states often contain coexisting orders that are not usually present in the standard ground or thermal states, making a remarkable difference in the way the system evolves after photoexcitation.

Non-equilibrium states can be probed using time-resolved spectroscopies, such as time-resolved ARPES. Recent advances in radiation sources and instrumentation have also enabled core-hole spectroscopies such as RIXS, XAS and Auger. However, computing response functions away from equilibrium has remained the main challenge to establish a connection between theory and experiment.

The present project aims at studying models and time-dependent processes to understand light-matter interaction in strongly correlated materials, and the interplay between electronic, orbital, vibrational, and spin degrees of freedom. The time scales involved in the creation and recombination of excitons, or the relaxation of the system after a perturbation, will be dictated by the way light couples to the different excitations, and how these excitations exchange energy and momentum. Our research will advance our understanding of these processes (related to photochemistry, opto-electronics, and light-harvesting applications), and the interpretation of different equilibrium and time-resolved spectroscopies.

Our project encompasses two main themes: (i) developing and refining computational techniques to study non-equilibrium spectroscopies including non-perturbative effects and (ii) applications to non-equilibrium phenomena.

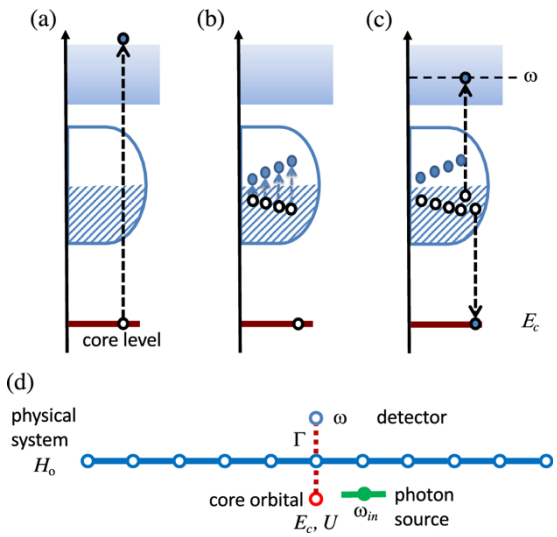
The method of choice to study time-dependent phenomena is the time-dependent density matrix renormalization group (tDMRG), which was co-developed by the PI, and has had a remarkable success expanding our knowledge of correlation effects to the time-domain.

Recent progress:

During the previous funding cycles, the PI has introduced three innovative computational approaches to study non-equilibrium spectroscopies --such as time-resolved ARPES (trARPES)-- and core-hole spectroscopies --such as Resonant Inelastic X-Ray Scattering (RIXS). As described in this proposal, these ideas are nothing short of ground-breaking. The premise is to simulate the full scattering event including all the "actors" (*i.e.* the scattering particles --photons, electrons, neutrons--, core orbitals, etc.) and

numerically solve the time-dependent Schrödinger equation. The response function is measured in a "detector", akin an actual scattering experiment. This approach not only overcomes the limitations of perturbation theory but provides a solution to all orders allowing one to study higher order contributions to the spectra and other many-body phenomena discussed in this project.

Auger electron spectroscopy: We extended our time-dependent scattering approach to study Auger electron spectroscopy (AES) beyond the ultra-short core-hole relaxation time approximation. The Auger spectrum is usually calculated by assuming that, after the creation of a core-hole, the system thermalizes almost instantaneously. This leads to a relatively simple analytical expression relating the scattering cross-section to the pair spectral function. On the other hand, our numerical calculations allow one to study the transient dynamics of the system in terms of the pulse duration and in the non-perturbative regime. Time-dependent density matrix renormalization group calculations reveal that the relaxation process involves the creation of a polarization cloud of doublon excitations that have an effect similar to photo-doping. By changing the duration of the light-pulse, the entire screening process can be resolved in time.



Schematic representation of the regimes involved in the core-hole creation and non-radiative Auger recombination: (a) core-hole creation; (b) transient regime in which the core hole is screened creating a polarizarion cloud; (b) an electron recombines with the core-hole while a second one is ejected into the continuum. (d) Geometry used in the calculations

Non-radiative recombination: We further extended the previous approach to provide a more realistic description of Auger processes responsible of non-radiative core-hole recombination in X-ray experiments. The mechanisms responsible for the lifetime of the core-hole are not completely understand, but usually attributed to Auger recombination. However, in experiments and theory, the lifetime is introduced as an adjustable phenomenological parameter to fit the data. We make progress in understanding the interplay between radiative and non-radiative recombination taking place concurrently at the same time, and shed light onto the effects of the Auger mechanism on the lineshape of RIXS spectra.

Effective one band model for the 1d cuprate $Ba_{2-x}Sr_xCuO_{3+d}$: We derived an effective low energy Hamiltonian for CuO chains starting from a four-band model with one relevant orbital per Cu or O atom. We find that, in order to account for the effect of excited triplets, the one-band Hubbard model should be supplemented by other terms not usually considered. In addition, the hopping term depends on the occupancy of the sites involved. For energies below the value of the effective Coulomb repulsion U, it is more convenient to use a generalized *t-J* model. Our DMRG results for the ARPES spectra are in semiquantitative agreement with experiment.

Quasi-Fermi liquids, a new paradigm: We present numerical evidence for a new paradigm in one-dimensional interacting fermion systems, whose phenomenology has traits of both, Luttinger liquids and

Fermi liquids. This new state, dubbed a “quasi-Fermi liquid” possesses a discontinuity in its fermion occupation number at the Fermi energy while lacking the associated Landau quasiparticles in the hole sector. Such a state is realized in a one-dimensional spinless fermions Hamiltonian by fine-tuning the interactions to a regime where they become irrelevant in the renormalization group sense. We show, using uniform infinite matrix products states and finite-entanglement scaling analysis, that the system's ground state is characterized by a Luttinger parameter $K = 1$ and a discontinuous jump in the fermion occupation number. We support the characterization with calculations of the spectral function, that show a particle-hole asymmetry reflected in the existence of well-defined “delta-like” Landau quasiparticles above the Fermi level, and edge singularities without the associated quasi-particles below. These results indicate that the quasi-Fermi liquid paradigm can be realized beyond the low-energy perturbative realm.

Probing entanglement with spin-transfer torque and X-rays: Using the nascent concept of quantum spin-transfer torque, we predict that a charge current pulse can be harnessed to entangle localized quantum spins of two spatially separated ferromagnets (FMs) within a FMp/NM/FMa spin-valve spintronic device, where a normal metal (NM) spacer separates a spin-polarizer (FMp) from a spin-analyzer (FMa) FM layer. The dynamical build-up of mixed-state entanglement between the FM layers is quantified by calculating the mutual logarithmic negativity, entanglement entropy and mutual information over time via fully quantum many-body approaches. The effect of decoherence on our scheme, the use of multi-electron pulses and the scaling with system size are also analyzed in an effort to ascertain the robustness of our predictions under realistic experimental conditions. Finally, we propose a “current-pump/X-ray-probe” scheme, utilizing ultrafast X-ray spectroscopy, which can witness nonequilibrium entanglement of the FM layers by extracting their time-dependent quantum Fisher information.

Future Plans:

In this funding period we aim at cementing our collaboration with experimental groups and extending our methods to multi orbital problems to study realistic Hamiltonians. It is subsequently divided into four (4) thrusts: (i) Applications of time-resolved ARPES; (ii) a DMRG-RIXS code for multi-orbital problems; (iii) Many-body Floquet phononics; (iv) Pump-driven phase transitions. Specifically:

The fate of quasi-particles after a pump: It is reasonable to assume that in ladders, a finite temperature or a pump will overcome the binding that holds the polaron-like quasiparticle together and only deconfined holons, spinons, or triplons would survive. We will numerically study the fractionalization of the polaron-like quasiparticle into deconfined excitations after a pump near half-filling by analyzing the “melting” of the bound states into the continuum with time-resolved ARPES and neutron scattering in *t-J* ladders. This would indicate a path toward understanding and interpreting the physics on non-equilibrium quantum matter beyond the conventional quasi-particle description.

Pair breaking after a pump: When pairs are broken by a pump, the resulting state would have the properties of the parent `normal' state without superconducting correlations. Presumably, the charge carriers in this state will be polarons (bound states of a spinon and a holon across the rung). However, the binding energy that keeps the polaron together is of the same order as the pair binding energy and, as a consequence, might not be stable either. As a result, we might get a “soup” of deconfined holons and spinons with a completely incoherent spectrum and only a continuum remaining. We will study the non-equilibrium photo-emission spectrum of a doped *t-J* ladder after a pump to unveil the nature of the excitations in the underlying normal state.

Two-electron ARPES: Collaborator Andrea Damascelli at UBC will study quantum materials with a novel technique to directly probe electron correlations: coincidence two-electron angle-resolved photoemission spectroscopy (2e-ARPES). His group has been working since 2021 on the development of instrumentation for Momentum-Resolved Electron-Pair Spectroscopy, a one-of-a-kind such infrastructure. This approach uniquely combines coincidence electron-pair detection with high momentum-, energy-, and dynamic time-resolution. In this experiment, two electron will be ejected from the sample kinetic energies ω_1 and ω_2 and momenta k_1, k_2 ,which are separately and simultaneously measured in two detectors. We will use our recently developed tunnelling approach to simulate different scenarios using the repulsive and attractive extended Hubbard chains that realize both spin singlet and spin triplet paired phases, FFLO and η-pairing, and identify the spectral signatures of each order.

Floquet phononics: The field of ``nonlinear phononics" concerns the selective excitation of phonon modes of the crystal lattice. In this case, while the driving is induced by light, the pump does not interact directly with the electronic degrees of freedom. The targeted excitation of a particular phonon mode opens additional pathways for mode-selective control that are not accessible in traditional driving schemes. We will consider a problem of atoms vibrating classically such that their atomic positions vary periodically in time oscillating about their equilibrium positions. In particular, we will focus our attention on the effect of the time-varying distance on the atomic overlaps. The problem consists of understanding and developing a modified Floquet theory in a time-evolving Hilbert space when atomic overlaps are taken into consideration.

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Intertwined and Vestigial Electronic Orders in Correlated Systems

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Keywords: Correlated-electron systems; intertwined orders; unconventional superconductivity; twisted moiré systems; exotic electronic phases.

Research Scope

The overarching goal of this project is to investigate and elucidate the intertwining between various electronic orders realized in correlated materials, going beyond the competing-order paradigm and focusing on the framework of vestigial electronic orders. The innovative aspect of this research is its focus on the behavior of intertwined and vestigial orders in both crystalline and non-crystalline elastic media to elucidate how the lattice degrees of freedom modify, constrain, and even enable unique collective electronic behavior. During this period, the PI developed a complete classification of electronic nematicity in all crystalline and quasi-crystalline point groups and demonstrated the emergence of an exotic critical nematic phase displaying pseudogap behavior in twisted moiré systems. The PI also developed a general framework to explore electron-phason coupling in twisted moiré systems, demonstrating the existence of an unusually extended temperature regime in which this scattering process promotes linear-in-T resistivity. Finally, the PI discovered the ubiquitous presence of sub-leading vestigial charge-4e order instabilities with s-wave and d-wave characters in multi-component superconductors, showing that they can be locally stabilized in nematic and magnetic domain walls.

Recent Progress

Nemato-elastic interactions in diverse crystalline structures. The electronic nematic and the lattice degrees of freedom are deeply intertwined, since uniform strain acts as a conjugate field to the nematic order parameter. This not only enables external control over this quantum state of matter, but it also enhances the impact of random strain ubiquitously present in real materials. During this period, the PI made significant advances in the elucidation of nemato-elastic interactions in correlated systems. The fact that signatures of electronic nematicity are observed in an ever-growing list of materials with diverse electronic properties and crystalline structures, such as tetragonal iron-based superconductors, trigonal doped topological insulators, and correlated twisted materials with triangular moiré

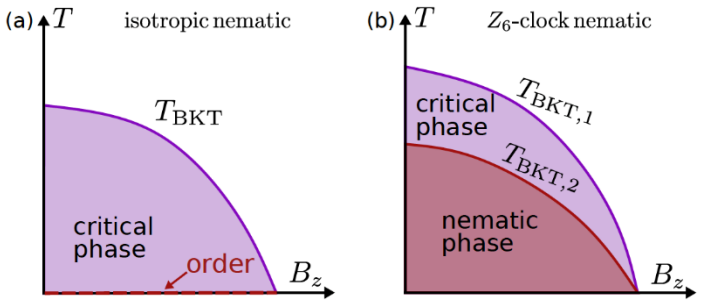

Figure 1: Temperature-magnetic-field nematic phase diagram of (a) an ideal isotropic system and (b) a 30°-twisted hexagonal bilayer. In the latter, before the onset of long-range nematic order, a critical phase with quasi-long-range nematic order is stabilized via a BKT transition. Inside this phase, nematic fluctuations promote a pseudogap-like behavior. From Ref. [2].

superlattices, motivated the PI to carry out a thorough classification of nematic phenomena in all crystalline and quasicrystalline point groups [1]. This revealed various promising routes to realize exotic nematic phenomena, such as emergent continuous nematic fluctuations and critical nematic phases. In [2], the PI explored one of these routes, which consists of twisting two hexagonal monolayers nematic materials by 30°. The resulting phase diagram, shown in Fig. 1, reveals many interesting effects that are enabled by the fact that the twisted bilayer behaves as an artificial quasicrystal, as it displays an incommensurate "lattice" and a crystallographic-forbidden twelve-fold rotational symmetry. This enhanced symmetry enables this system to host a nematic phase diagram that shares many properties with that of the ideal isotropic nematics. In particular, a critical phase with quasi-long-range nematic order emerges below a BKT (Berezinskii-Kosterlitz-Thouless) transition, which can be tuned by an external out-of-plane magnetic field acting as a transverse nematic field via an effect akin to the Hall viscosity in viscoelastic media. Remarkably, the low-energy nematic fluctuations present throughout the critical phase mediate electronic interactions that give rise to a pseudogap-like behavior in the electronic spectrum, enabled by the anomalous critical exponent that characterizes nematic quasi-long-range order.

Electron-phason coupling in twisted moiré systems. In contrast to bulk crystals, the elastic excitations of the emergent moiré superlattice that characterizes twisted bilayer graphene are not acoustic phonons, but phasons. In contrast to the former, the latter can display a small gap and, most importantly, displays overdamped dynamics at low energies originating from the friction between the layers. These soft collective elastic excitations mediate important interactions between the low-energy moiré electronic degrees of freedom, which are qualitatively different from the electron-phonon interactions of regular crystals. In Ref. [3], the PI and collaborator, after having previously developed a formalism to describe moiré phason excitations, investigated how electron-phason scattering impacts the transport properties of twisted bilayer graphene. In monolayer graphene, the electron-phonon coupling causes a linear-in-temperature (T) resistivity down to the Bloch-Grüneisen temperature, below which the resistivity shows a T^4 behavior. The situation changes considerably in twisted bilayer graphene due to the overdamped dynamics of the phasons, which renders phason scattering an efficient channel for entropy production. At low enough temperatures, the resistivity always shows a "metallic-like" T^2 behavior, rather than T^4. Moreover, when the temperature scale

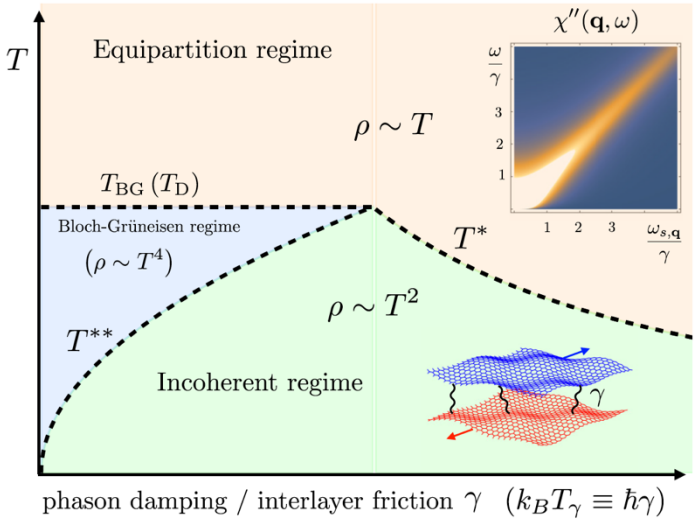

Figure 2: Different regimes for the temperature-dependence of the resistivity caused by scattering between electrons and phason excitations of the moiré superlattice. The inset shows the imaginary part of the phason susceptibility, which displays a broad incoherent peak reflecting the phason damping enabled by the friction between the layers. From Ref. [3].

associated with the damping experienced by the phasons is large enough, the equipartition regime of linear-in-T resistivity is extended to temperatures much smaller than the Bloch-Grüneisen one. Specifically, this new temperature scale could be as small as 0.1K, which might explain the puzzling linear-in-T resistivity experimentally observed in twisted bilayer graphene over a wide range of doping and down to very low temperatures.

Cascades of intertwined vestigial orders. Vestigial orders are characterized by the partial melting of an underlying electronic order, such as charge and magnetic stripes. Previous works by the PI have shown that multi-component superconductors provide a fertile ground for the realization of vestigial order. Indeed, the ground state of multi-component superconductors often break either rotational or time-reversal symmetry. As a result, it is not surprising that vestigial nematic and ferromagnetic order can emerge. In Ref. [4], the PI demonstrated that, besides these types of vestigial order, multi-component superconductors generally support vestigial exotic charge-4e order, which can have s-wave or d-wave symmetry. However, in most cases these vestigial charge-4e instabilities, which are described by complex-valued composites, are sub-leading compared to the nematic and magnetic ones, which are described by real-valued composites. Motivated by these results, in Ref. [5], the PI showed that when the leading vestigial order is suppressed, such as in nematic and magnetic domain walls, charge-4e order is stabilized before the onset of superconductivity. This provides a promising setting to search for local charge-4e order.

Future Plans

The PI will continue the investigations about the impact of lattice disorder on electronic nematicity, in order to provide a realistic description of this phenomenon in materials. In particular, instead of assuming a random distribution of internal strains, the PI will consider a distribution of correlated and long-range strain generated by random defects. Still in what concerns nemato-elastic interactions, the PI will elucidate how the long-range nematic interactions mediated by acoustic phonons impact electronic nematicity in different crystalline structures, such as cubic materials or incommensurate lattices with phason excitations. Finally, the PI will also continue Quantum Monte Carlo investigations of vestigial phases by simulating a multi-band Hubbard model with inter-band-only repulsive interactions. This model, which does not suffer from the fermionic sign problem, has been previously shown by the PI to display superconductivity and magnetism. Stabilization of a stripe-type magnetic state opens the door to look for vestigial nematic order.

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Exploring the quantum potential of magnetic systems

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Keywords: Magnetic systems, cooperative quantum phenomena, quantum entanglement, magnetization dynamics, dissipation.

Research Scope

The possibility of engineering entanglement and quantum superpositions in many-body systems using tailored environments and harnessing these resources to realize new dynamical states of matter and novel applications in quantum technologies has gained vast traction in quantum optics [R1]. At light-matter interfaces, atomic emitters interact with each other dissipatively, i.e., through the emission or absorption of *real photons* in a shared photonic reservoir, i.e., see Fig.1(a). When multiple quantum emitters radiate in the shared bath, the collective spontaneous emission rate of an ensemble of atomic emitters is initially enhanced (*super-radiance*) compared to that of an isolated emitter, as shown in Fig.1(b). While relaxing towards the ground state, super-radiant states evolve into *sub-radiant* states, i.e., long-lived many-body states exhibiting nontrivial quantum correlations that can be harnessed for quantum-enhanced metrology and quantum-information

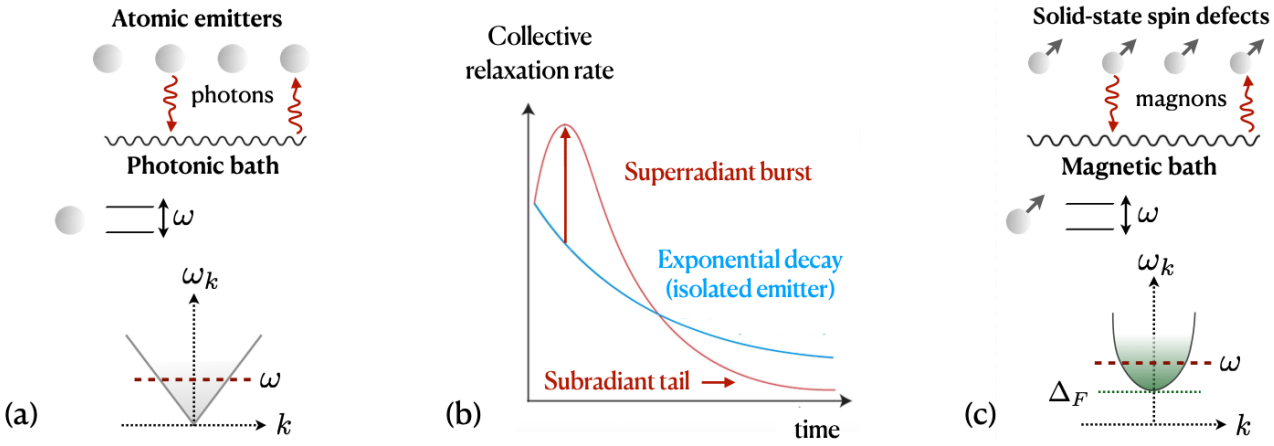

Figure 1. (a,b) At light-matter interfaces, atomic emitters interact *dissipatively* with a shared photonic bath — with gapless dispersion ω_k — by emitting (absorbing) *real photons* into (from) the bath. This interaction can establish *nontrivial quantum correlations* among the quantum emitters. As a result, the relaxation dynamics of a qubit ensemble initialized in a trivial excited state relaxes towards the ground state via a subradiant burst followed by a subradiant tail — rather than displaying the exponential decay characteristic of an ensemble of uncorrelated emitters. **(c)** When their resonance frequency ω is larger than the gap Δ_F of the spin-wave dispersion ω_k of a shared magnetic bath, the solid-state spin defects interact *dissipatively* with the bath, in close analogy to the regime illustrated in **(a)**.

processing and storage applications.

The recent progress in quantum optics suggests that the *minimal* ingredient for harbouring dissipation-driven nontrivial correlations in a quantum many-body system is an ensemble of long-lived quantum systems interacting *dissipatively* with a shared reservoir. Inspired by this recent

progress and by the lack of efficient pathways for the generation of *long-range* couplings among solid-state spin qubits, our current research is set to explore the uncharted regime in which *an ensemble* of solid-state spin defects interact *dissipatively* with a *shared* magnetic bath, sketched in Fig.1(c).
A current key research focus is the identification of experimentally feasible setups in which dissipative qubit-bath interactions can lead to observable signatures of many-body quantum dynamics and the exploration of their robustness against the impact of finite-temperature and disorder effects.

Another avenue of ongoing research is motivated by recent proposals for generating qubit-qubit entanglement through the quantum dephasing noise of a common reservoir [R2]. Following recent experimental results reporting that proximity between a NV-center and a domain wall texture is responsible for a significant enhancement of the NV-center dephasing time [R3], we are currently exploring the generation of many-body entanglement in an ensemble of solid-state spin defects interacting via the Goldstone modes of a domain wall.

Recent Progress

We have developed a comprehensive theoretical framework for exploring the uncharted regime in which dissipative interactions between an ensemble of spin qubits and a shared solid-state bath can generate nontrivial quantum correlations within the qubit ensemble. Our formalism applies to a wide range of solid-state spin defects and any solid-state reservoir whose fluctuating spin, pseudospin, or charge degrees of freedom can generate magnetic fields.

To understand whether dissipative correlations can play a relevant role in a realistic experimental setup, we have applied our general framework to an NV-center array interacting with a simple ferromagnetic bath, i.e., a Yttrium Iron Garnet (YIG) film. Our results [P1], together with recent experimental progress [R4], suggest that experimental signatures of super- and subradiance should be already detectable in the relaxation dynamics of the disordered and inhomogeneously broadened NV-center ensembles previously used to probe room-temperature properties of YIG spin waves [R5], provided that the YIG film is cooled down to low temperatures, i.e., $T \sim 100$ mK.

Furthermore, our (yet unpublished) investigation of an ensemble of solid-state spin defects interacting via the Goldstone modes of a domain wall has confirmed that the low-frequency quantum noise generated by a *nonreciprocal* ferromagnetic domain wall can establish many-body entanglement in the qubit ensemble.

Future Plans

A near-future goal is to explore the many-body quantum dynamics of an ensemble of spin qubits interacting with a squeezed magnetic reservoir. Specifically, we aim to identify a magnetic reservoir that: i) is compatible with the GHz energy scales of solid-state spin defects, and ii) supports squeezing that can be toggled on and off using experimental controls compatible with solid-state spin-defect setups.

Inspired by the progress in topological quantum photonics, we will also investigate the interplay between topology and quantum many-body dynamics in spin-qubit arrangements that yield a multi-band structure, such as 2D arrays or 1D lattices with a double unit cell.

Finally, we intend to leverage the insights gained from the current research to develop a theoretical framework describing nonlocal dissipation and quantum corrections to the magnetization dynamics of bulk magnetic systems and heterostructures.

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Nonequilibrium dynamics of correlated quantum systems with disorder under an interaction quench

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Hanna Terletska, Middle Tennessee State University

Keywords: Nonequilibrium, Correlated Systems, Disorder, Quench

Research Scope

The interplay of interaction and disorder gives rise in equilibrium to a vast array of intriguing properties and has thus rightfully received a great deal of attention. Away from equilibrium however, the transient dynamics of many-particle correlated systems that feature both interaction and disorder is rather challenging despite this significant interest. To enable the investigation of the nonequilibrium dynamics of interacting systems in the presence of disorder, we recently introduced the nonequilibrium DMFT+CPA method [1]. An embedding scheme that combines the nonequilibrium extensions of both the dynamical mean field theory (DMFT) [2,3] and the coherent potential approximation (CPA) [4,5]. This framework opens up the possibility for detailed exploration of the effects of disorder on the dynamics away from equilibrium of correlated quantum systems with connections to various fundamental questions and practical applications..

The approach was previously benchmarked on the equilibrium solution of the Anderson-Hubbard model describing itinerant electrons with on-site interaction as well as a site dependent random on-site energy. The analysis reveals among other features the disorder-induced insulator-to-metallic phase transition that occurs for Mott gap of strong interactions is filled when the disorder strength is increased. In recent efforts, we have analyzed the effect of disorder on the nonequilibrium dynamics of this correlated system, described by the Anderson-Hubbard model, under an interaction quench. The system, initially in equilibrium at a given temperature, $T_{initial} = 1/\beta_{initial}$, has the interaction abruptly switched from zero to a finite value at a given time.

Recent Progress

To investigate the role of disorder on the nonequilibrium dynamics of a correlated many-particle system, we use our effective medium approach to calculate, through the nonequilibrium Green's functions, for different values of the final interaction and varying disorder strengths, the distribution functions as the system evolves in time. This allows us to determine the effective steady state temperature after the quench and to analyze the effects of disorder on the thermalization for various interaction strengths. The analysis shows that disorder can tune the final temperature of the system across a broad range of values [6]. Very importantly this analysis also reveals that for a quench from a non-interacting system to a weakly interacting one, the steady

state temperature is increased with increasing disorder strength whereas this trend is reversed for moderate final interaction strengths.

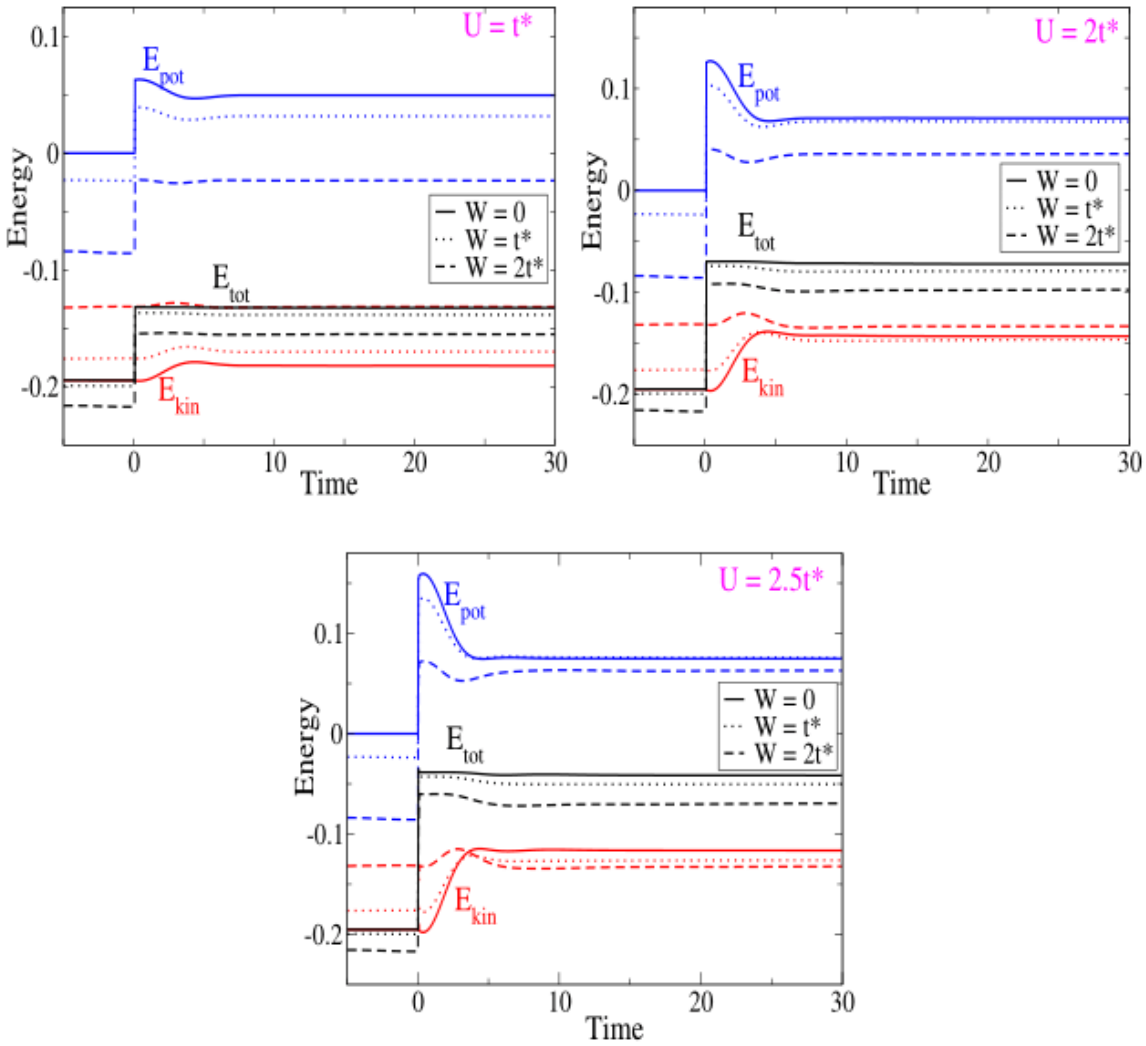

Figure 8: Relaxation of the kinetic, potential and total energy of the system across the interaction quench for different interactions as a function of disorder strength [7].

Within our nonequilibrium DMFT+CPA framework, we consider various types of disorder and analyze the thermalization of the system under an interaction quench. Figure 1 shows the relaxation of the kinetic, potential and total energy across an interaction quench for and interaction quench occurring at time $t = 0$ from $U_1 = 0$ to $U_2 = t^*$, $2t^*$, and $2.5t^*$ for different values of disorder. It shows that for for weak final interaction, the final kinetic energy following the nontrivial transient immediately after the quench is increased with increased disorder strength and that this trend is reversed when the final interaction is increased [7].

Figure 2 shows the steady state inverse temperature as a function of final interaction U_2, and disorder strength W, for a system that is quenched at an earlier time from $U_1 = 0$ to U_2. Energy is measured in units of the effective hopping amplitude *t*[6]*.

Future Plans

The location in phase space of the thermalization crossover highlights a subtle yet important feature of the free energy landscape of the disordered correlated systems. We intend to examine the thermodynamic properties of the Anderson-Hubbard model in equilibrium to shed some light on this unexplored aspect of our system. The present studies have been conducted for "box" and binary disorder. We will carry out a comparative study of how different other types of disorder affects the nonequilibrium dynamics that follows an interaction quench.

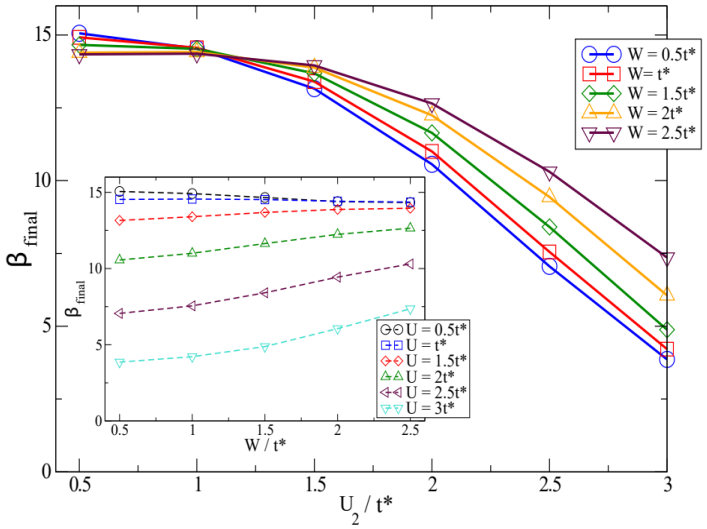

Figure 9: Effective steady state temperature of the system after thermalization for different interaction strengths as a function of disorder strength.

Next, we plan on implementing a strong coupling solver in order to be able to effectively address the dynamics of the system beyond the weak coupling regime. Considering the effect of disorder on the Mott insulator phase, we anticipate that the nonequilibrium dynamics might show important dependence on disorder.

In parallel with this development of the strong coupling solver, we will derive and implement our formalism for the field-driven Anderson-Hubbard model. This will allow the solution of a new nonequilibrium problem and the characterization of various current experiments. We are also involved in ongoing discussion with collaborators at Louisiana State University (LSU) and at the University at Buffalo, to apply our framework to other nonequilibrium problems.

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Theory for pump/probe experiments in charge-density-wave materials

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Keywords: Nonequilibrium many-body problem, pump/probe spectroscopy, electron-phonon coupled systems, charge-density-wave systems

Program Scope

Objectives: This project is focused on theoretically describing pump-probe experiments. One of the most studied classes of materials are charge-density-wave (CDW) ordered quantum materials that illustrate a wide range of complex and interesting behavior. We want to determine the fundamental behavior of electron-driven and phonon driven charge-density-wave materials. The most significant technical developments we have undertaken are to develop a new computational theory to describe pump-probe experiments in electron-phonon mediated materials that can compute effects out to tens of picoseconds where the phonon dynamics occur. The approach initially worked in the adiabatic limit, where the phonon mass is taken to the limit of infinity. Next, we generalized the approach to incorporate Ehrenfest forces on the phonons in a semiclassical fashion. We are planning to generalize to working with coherent states, which will incorporate additional quantum effects for the phonons. Using this numerical approach, we have discovered a robust mechanism to avoid the heat-death in Floquet-driven electronic systems and we have been able to provide a microscopic description of a number of recent pump-probe experiments in CDW systems.

Recent Progress

The first group of projects we have completed involve nonequilibrium dynamics in electron-phonon coupled systems. We have first studied the response of an electron-phonon coupled system to a large DC electric field. The current folklore says this should heat up to infinite temperature. But, instead, we find that the gauge-invariant electronic momentum distribution quickly becomes an even function of momentum. Once this occurs, the current vanishes and the system can no longer heat up, even though an electric field is present (because the Joule heating is given by $E \cdot J$, which vanishes if J=0). The steady state that emerges is quite nonequilibrium. The density of states develops broadened Wannier-Stark bands that are occupied by electrons in a Fermi-like distribution for each miniband (rather than an overall Fermi-Dirac distribution for the set of bands). The distribution in each miniband has a different effective temperature. Depending on the initial conditions (initial temperature of the system and strength of the DC electric field that is turned on), we can heat to infinite temperature, to a negative temperature, or to a finite temperature. The heating ceases on a short time scale on the order of 100 femtoseconds or less. These results are summarized in Fig. 1. This work has been published in Phys. Rev. Lett.

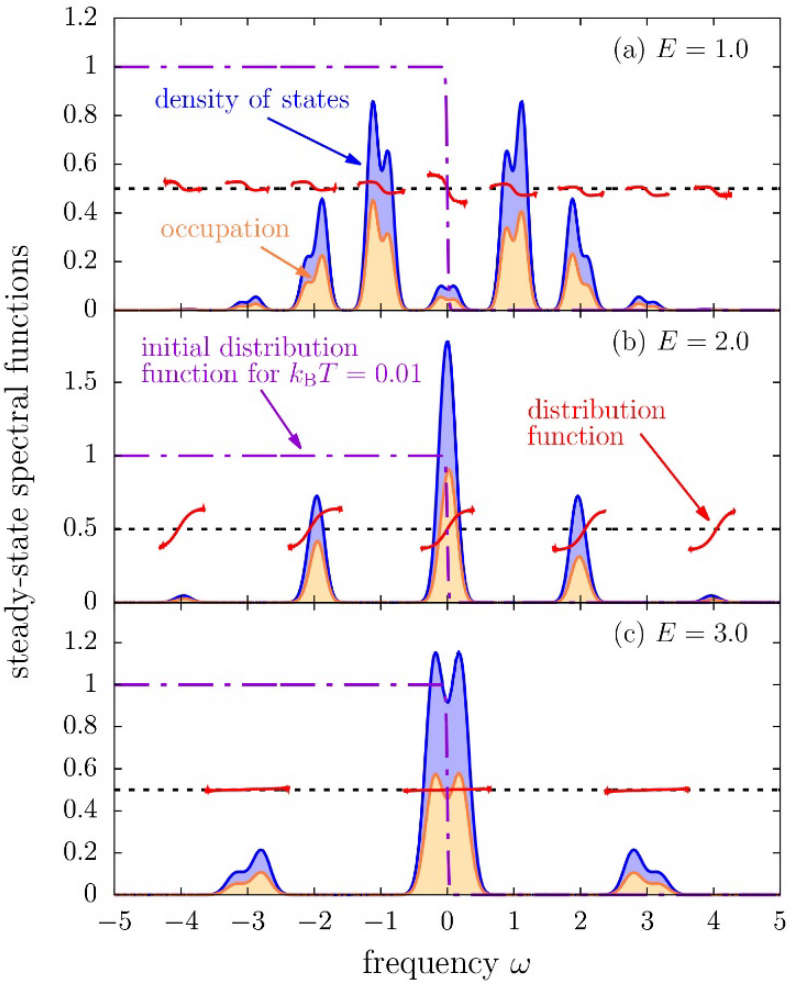

Fig. 2. Nonequilibrium density of states (blue) and occupancy (beige). One can see the individual Wannier-Stark minibands, partly split by the interactions, with a distribution (red lines) that is given by high-temperature Fermi-Dirac distributions. This behavior is highly nonequilibrium, but is stable, because the runaway heating of typical Floquet systems is shut off due to the dynamical symmetrization of the electron momentum distribution. In the top panel, we have an evolution to a positive finite temperature. In the middle panel to a negative temperature and in the bottom panel to infinite temperature. The steady state is determined here simply by changing the driving field magnitude.

We have also have investigated time-resolved photoemission spectroscopy and other probes in CDW systems. Our numerical calculations span many 10s of picoseconds, so we can faithfully simulate the behavior of these systems on the phonon timescales. We see similar behavior for the oscillation of the band edge of the photoemission spectroscopy with that seen in experiment on a typical CDW system. The results are shown in Fig. 2. We also have investigated two-dimensional spectroscopy, where we apply two pumps, with a time delay, and we have examined x-ray diffraction. In the latter case, we see fluence dependence to the driving frequency and the damping rate, precisely as seen in experiments. This work is on the third round of reviewing with Phys. Rev. X.

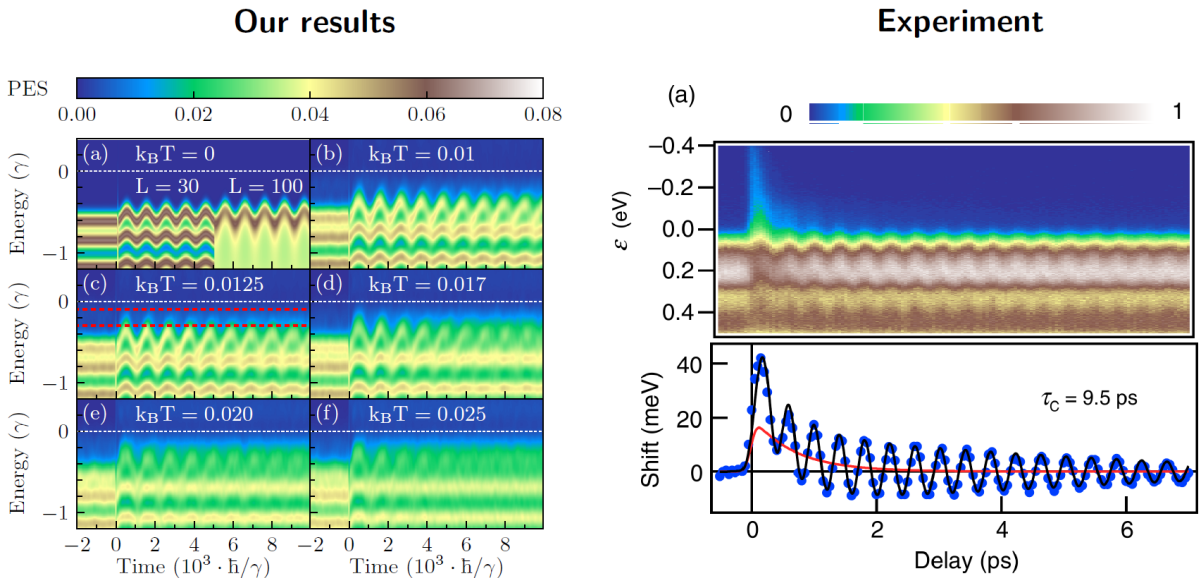

Fig. 2. (Left) Time-resolved photoemission spectroscopy of a CDW system at different initial temperatures and (right) experimental photoemission spectroscopy for a CDW system (from the Perfetti group). As T rises, the excited phonon is more and more damped. The oscillation of the phonon can be seen by the oscillation of the band edge of the photoemission. We see similar damping to experiment at intermediate Ts.

We also have investigated the dynamical transition seen as a function of fluence in CDW systems. When we are in the CDW phase, the phonon potential has a double-well nature to it. As we pump energy in the system, it changes to a single well. Interestingly, if one can tune the fluence to drive the phonon right to the bottom of the single well, then it will just sit there. This leads to a dynamical decoupling of Holstein phonons, since their interaction is proportional to the phonon coordinate. In this case, they would have no interaction on average, and very small interaction in general. The transition is very sharp in the fluence and has been seen by Nuh Gedik's group. We summarize our calculations in Fig. 3. A paper on these results is currently being written up.

Future Plans

Our main effort is to continue the work with our new code in the following way. We will improve the algorithm by adding in more quantum effects. We will examine direct driving of the phonon (as opposed to electron driving, which we have initially investigated). We also plan to work with the Murnane group on experiments they have been conducting on topological insulators and charge-density-wave systems. In addition, we hope to investigate electron-driven CDWs that might exhibit emergent quantum critical behavior, which we have examined previously. Finally, we plan to examine a number of formal aspects related to using photon wavepackets in the theory as opposed to single-mode photons (which brings in time-dependence

of the signals for arrival times at detectors) and we will also investigate how to connect reflectivity data to optical conductivity data in nonequilibrium.

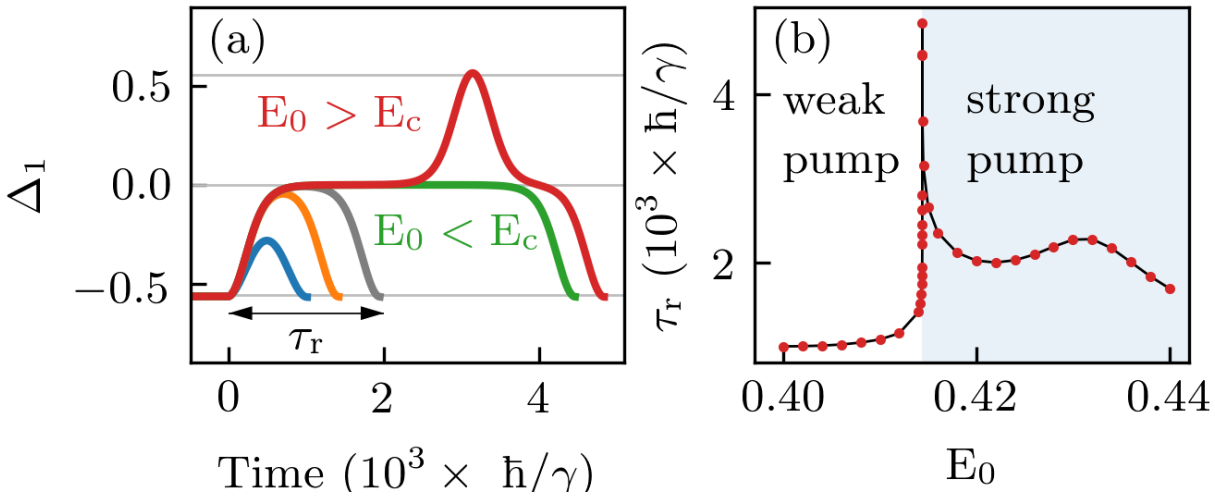

Fig. 3. (Left panel) the phonon coordinate as a function of time for different fluences. For weak fluence, the phonon coordinate oscillates about the CDW-ordered value. At high fluence, it oscillates from one ordered value to the other, as if the double well has become a single well. At the critical fluence, the phonon coordinate is driven to the origin (center of the single well) and stays there. (Right panel) The rise time as a function of fluence measures the time it takes the phonon to reach its maximal variation due to the pump. In the critical region, this rise time sharply increases. Similar results have been seen in Nuh Gedik's experiments on CDW systems.

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Quantum Dynamics of Strongly Correlated Materials

PI: Victor Galitski (Joint Quantum Institute, University of Maryland)

Keywords: chaos, ergodicity, thermalization, non-equilibrium, superconductivity

Research Scope

This DOE project is focused on studies of quantum chaos in interacting many-body systems. When it comes to electron systems, there are two types of chaotic dynamics: Single-particle quantum chaos is driven by electron scattering off disorder potential. It dominates momentum relaxation and transport in most materials [1], but has no relation to thermalization, which requires inelastic scattering. Many-body quantum chaos on the other hand implies the emergence of universal energy level statistics due to interactions [2], which in turn leads to thermalization. A related topic is hydrodynamics, which describes collective dynamics of translation-invariant media. Indeed, the Navier-Stokes equation is effectively a momentum conservation law. Electronic hydrodynamics [3,4] and many-body chaos are connected because in both cases dynamics are driven by momentum-conserving scattering. There is a growing class of materials, where interaction-driven quantum dynamical phenomena dominate.

The research focus of the current DOE project has been on the frontier topic of time-dependent evolution and driven quantum dynamics of many-body systems. In particular, the PI and his group has focused on the following questions:

1. Why many seemingly unrelated correlated quantum systems exhibit the same universal energy level statistics? The goal here has been to understand the fundamental mathematical underpinnings of many-body quantum chaos in a variety of physical systems: from abstract quantum dynamical systems to quantum circuits to realistic electron systems.
2. While to calculate non-equilibrium quantum dynamics of strongly interacting systems from first principles is often an unsurmountable task, PI's group has been exploring random matrix theory (RMT) methods to derive and constrain quantum dynamics assuming the underlying many-body RMT level statistics from the outset. The questions of importance are: what determines the thermalization scales? How does the universality of Wigner-Dyson RMT level statistics manifest itself in the dynamic response?
3. Another question specific to pure electron systems has been to identify the experimental signatures of hydrodynamic transport (as opposed to Drude transport). Mapping known phenomena from classical hydrodynamics to quantum electronic materials is another research activity within this project.
4. An important application of PI's general results has been to describe non-equilibrium dynamics of realistic electron materials, including driven superconductors. This has been motivated experimental reports of long-lived transient superconducting-like state in driven superconductors [4]. PI's work has focused on developing computational tools to accurately describe dynamics of non-equilibrium superconductors.

Recent Progress

PI's group has made significant progress in understanding the foundations of many-body quantum chaos both from the mathematical perspective and for more specific interacting systems, including developing practical tools to quantitatively describe thermalization dynamics in superconductors. Below is a brief description of the main highlights of PI's research within the current project:

1. *Mechanism of many-body quantum chaos.*

In a series of papers [1, 7–10], the PI and his group have developed a general theory elucidating the mechanism for emergence of universal random matrix level statistics in a variety of unrelated interacting systems. This progress is rooted in Michael Berry's influential early work [6] on single-particle quantum chaos, which related the appearance of the linear-in-time ramp in the spectral form factor (the hallmark of quantum chaos) to interference of periodic orbits. Specifically, it was understood that the linear ramp is the consequence of time-translation symmetry. The PI generalized it to many-body physics [9]. The key insight is that in non-interacting metals, periodic orbits preserve coherence for each electron. Interactions lead to dephasing and only global time-translation symmetry remains (see, Fig. 1). This symmetry breaking restores universal RMT picture at the many-particle level.

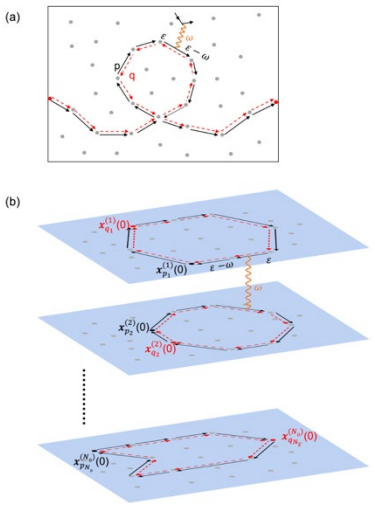

FIG. 1. Qualitative picture of the dephasing mechanism for (a) weak localization and (b) periodic-orbit theory of chaos.

2. *Quantum ergodicity and dynamics from random matrix theory.*

Ergodic theory provides a rigorous mathematical description of chaos in classical dynamical systems, including a formal definition of the ergodic hierarchy. How ergodic dynamics is reflected in the energy levels and eigenstates of a quantum system is the central question of quantum chaos, but a rigorous quantum notion of ergodicity remained elusive. In a recent comprehensive work [11], the PI and his student have resolved this questioned and connected random matrix theoretic description to time-dependent behavior of quantum systems. This includes deriving rigorous bounds on thermalization dynamics as reported in recent Letter [7].

3. *Slow Thermalization in Light-Induced Superconductors*

Light-induced transient superconductivity is arguably one of the most interesting experimental puzzles in condensed matter physics [5]. The PI and his group have developed novel analytical and (open-source) computational tools to accurately describe quantum dynamics of out-of-equilibrium superconductors [12]. Specifically, the PI addressed the slow thermalization puzzle of transient superconductivity experiments by the Cavalleri group, who have observed proxies of light-induced superconductivity far above equilibrium transition temperature and anomalously slow relaxation to equilibrium. This theory work explains these phenomena as a crossover from integrable coherent dynamics to ergodic dynamics/thermalization. The resolution is due to the bottleneck effect due to the lack of quasiparticles at early times, needed to break integrability.

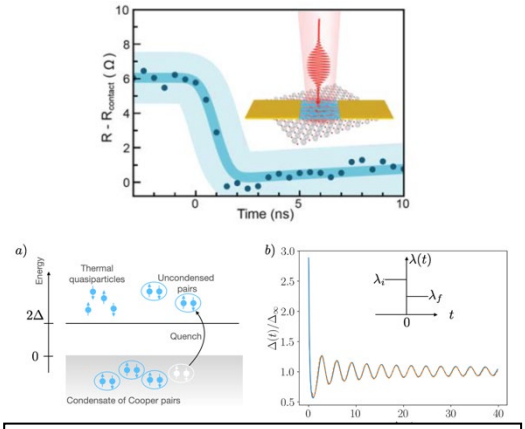

FIG. 2. Schematic of an experiment and time dependent optical conductivity data (see, Ref. [5] for a review). a) Schematic representation of the physical processes inside the quenched superconductor at finite temperature. b) Dynamics of the order parameter as derived in Ref. [12].

Future Plans

PI's future research plans in this DOE program are the following:

1. To develop a general theory of time-dependent dynamics of interacting systems out of equilibrium. It is generally a daunting task, but using PI's recent results [7], one can gain practical insights by relying on universality of random matrix theory that can be connected to time dependence of observables.

2. The PI plans to continue studies of non-equilibrium superconductivity. Recent work by the PI [12] showed that dynamics of the order parameter involves a coherent bosonic dynamic and chaotic relaxation (see, Fig. 2). PI's open-source code base will be further utilized and expanded to provide quantitative insights into driven superconductivity for other regimes including Floquet superconductivity under periodic driving with an eye on identifying optimal driving protocols.

3. The PI also proposes to further focus on manifestations of many-body quantum chaos and quantum hydrodynamics in electronic materials. In particular, the PI proposes to identify smoking gun signatures of hydrodynamic transport and experimentally relevant setups to explore Navier-Stokes electron systems. Of particular interest is to connect the microscopic foundations of many-body quantum chaos with the foundational questions in quantum hydrodynamics.

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Toward exascale computing of electron-phonon couplings: Polarons and Auger-Meitner recombination in materials for energy and microelectronics

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Keywords: Electron-phonon interactions, polarons, Auger-Meitner effect, solar cells, 2D materials

Research Scope

In this project we are developing methods and software to enable accurate, fast, scalable, and reproducible *ab initio* calculations of electron-phonon couplings for the design of advanced materials in the areas of renewable energy and microelectronics. Electron-phonon couplings play a significant role in the electronic and optical properties of semiconductors. For example, the electrical conductivity of semiconductors decreases with temperature because of electron scattering by phonons, and the photovoltaic energy conversion efficiency decreases with temperature because of phonon-assisted Auger-Meitner non-radiative recombination. Describing both processes requires predictive and efficient first-principles calculations of electron-phonon couplings. With this goal in mind, we are developing, testing, and deploying new *ab initio* methods, algorithms, and software for electron-phonon physics that go beyond the state-of-the-art in the field; we are consolidating and expanding the open-source software project EPW; and we are optimizing its performance portability on leadership-class DOE supercomputers.

This project builds on the foundations laid out during our prior DOE/BES CMS award "Toward exascale computing of electron-phonon couplings for finite-temperature materials design" and aims to address more complex challenges and enable *ab initio* calculations of new properties. Within our prior award, we established the *ab initio* many-body theory of polarons; we advanced *ab initio* calculations of carrier transport in semiconductors under electric and magnetic fields; we brought the EPW code into the pre-exascale regime; and we engaged with the scientific community via online user support and online as well as in-person events.

In the current project, we are pushing the frontiers of the theoretical and computational description of polarons, high-field transport in wide-gap semiconductors, transport in topological materials, and non-radiative Auger-Meitner processes. We are investigating time-dependent Boltzmann transport, non-Boltzmann polaron hopping transport, polaron-induced band structure and band gap renormalization, the role of large polarons in 2D materials and in halide perovskites, and strain engineering of phonon-assisted Auger-Meitner recombination rates.

Recent Progress

During the past year we have made sustained progress on three fronts: (i) Theory and computation of polarons in 2D materials; (ii) Theory and computation of phonon-assisted Auger-Meitner recombination; (iii) Refactoring and release of EPW v5.8 and accompanying technical manuscript. We describe these advances below.

Theory and computation of polarons in 2D materials. We developed the first *ab initio* theory of polarons in two-dimensional atomic crystals, and we established the universal laws that govern polaron formation in real 2D materials.

Polarons are emergent quasiparticles that consist of electrons dressed by a phonon cloud. Polarons are responsible for many technologically important phenomena, such as thermally activated transport and luminescence enhancement. Recently, new experimental capabilities such as ultrafast X-ray and electron diffraction have enabled the direct observation of polarons in bulk 3D semiconductors. Unlike polarons in bulk 3D materials, little is known about polarons in 2D atomic crystals. It is unclear whether polarons can form in strictly 2D materials, whether they are localized and to what extent, do they respond to external electric and magnetic fields, and whether collective phenomena such as bosonic condensates of bipolarons are possible in reduced dimensions. During the past year, we have made the first key steps toward answering these questions by developing an atomic-scale *ab initio* theory of polarons in 2D crystals [1]. We have found that the physics of polarons in 2D is fundamentally different from their 3D counterparts owing to a surprising effect whereby the effective Coulomb interaction between electrons and phonons becomes short-ranged. This effect alters the energetics and localization of polarons and gives rise to an unexpected and hitherto-unknown critical condition for the existence of polarons in two dimensions, which has no counterpart in bulk materials. To illustrate these conceptual developments, we have performed the first *ab initio* calculation of polarons on bulk and monolayer hexagonal boron nitride (h-BN). Monolayer h-BN has recently emerged as a versatile platform to realize hyperbolic phonon polaritons, single-photon emitters for quantum information processing, and light-emitting devices for deep ultraviolet optoelectronics, but the nature of electron-phonon coupling and polarons in this material remains poorly understood. We found that monolayer h-BN hosts large Frohlich-type polarons that arise from the coupling of carriers to longitudinal-optical polar phonons, supporting a recent experimental proposal. Furthermore, we expanded these observations to other 2D materials by developing a simple yet general model that links widely available materials properties such as dielectric constants and carrier effective masses to the polaron formation energy and size. Using this framework, we screened 2D materials for strong polaronic effects, and discovered that polarons in 2D can only form when the ionic dielectric screening and the carrier effective masses fulfil a critical condition. This new paradigm will stimulate and guide experimental investigations of polaronic effects in 2D through a variety of experimental probes, from strain-field mapping to ultrafast pump-probe spectroscopy.

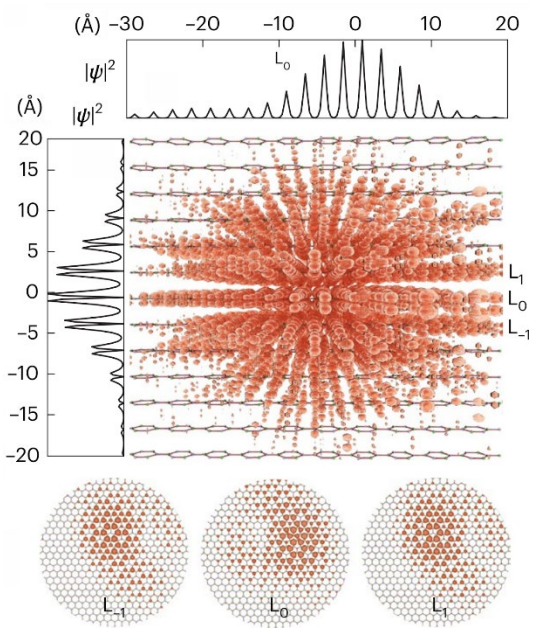

Fig. 1: Wavefunction of large hole polaron in h-BN: isosurface and cross-sections. From Ref. [1].

Theory and computation of phonon-assisted Auger-Meitner recombination. Auger-Meitner recombination (AMR) is an intrinsic non-radiative recombination process that is known to limit the performance of a variety of different electronic and optoelectronic devices. Given the broad

technological importance of silicon and the detrimental effect of AMR in silicon devices, there is a pressing need to understand AMR at a fundamental microscopic level in silicon and related photovoltaic materials. However, experimental efforts are constrained by the difficulty of measuring a non-radiative mechanism, and theoretical and computational efforts have been limited by the computational complexity of the calculations involved. These challenges are exacerbated when considering phonon-assisted processes, which are important for indirect-gap materials like silicon.

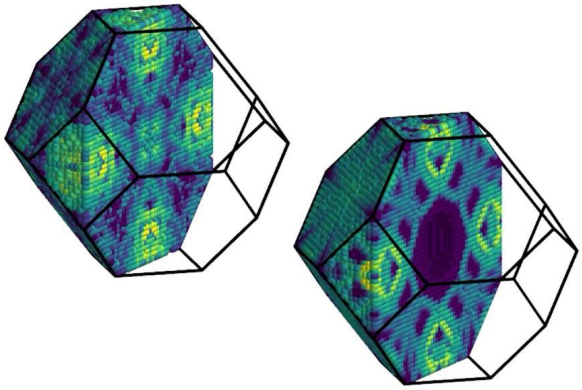

Fig. 2: Intensity map of AMR processes in the silicon Brillouin zone. From Ref. [2].

During the past year, we conducted a detailed theoretical characterization of the direct and phonon-assisted AMR processes in silicon from first principles. We determined the AMR coefficients as a function of temperature and carrier concentration, and we leveraged our predictive methodology to decompose the AMR process in terms of contributions from different phonons and valleys. Our analysis revealed the importance of zone-edge acoustic phonons to the phonon-assisted mechanism and indicated a route for modulating the AMR rate via strain engineering [2]. Following up on this development, we applied our approach to analyze the effects of compressive and tensile biaxial strain on AMR in silicon. We found that the application of tensile strain can reduce the hole-hole-electron AMR coefficient by approximately 40%. We then used EPW to calculate the AMR coefficients under the various strain conditions and analyze the contributions from different phonon modes. Our calculations show that tensile strain can simultaneously increase the lifetime and the in-plane mobility of minority electron carriers in *p*-type silicon, indicating a potential new pathway to improve the performance of silicon solar cells.

Refactoring and release of EPW v5.8. Work during the past year has led to the release of EPW v5.8. This new release contains several significant advances and is described in an extended technical manuscript that was published last year [3]. We developed a magneto-transport module for predicting charge carrier mobilities in semiconductors and conductivities in metals under external electric and magnetic fields. In this module, we incorporated corrections for the electron-phonon scattering matrix elements resulting from polar phonon scattering and from quadrupole scattering, both in 3D and 2D materials. These corrections are critical to perform reliable calculations of the transport properties of 2D materials. Furthermore, we implemented carrier scattering by ionized impurities, and we optimized and released a module to compute both small polarons and large polarons.

Future Plans

During the next reporting period, we will focus on (i) the investigation of large polarons in halide perovskites; (ii) the development of methods and software for computing polaron hopping transport based on the *ab initio* theory of polarons introduced by our group; (iii) the consolidation and release of the AMR code within EPW; (iv) the improvement of Brillouin-zone sampling techniques for accurate calculations of transport coefficients in topological semimetals; (v) the implementation and testing of time-dependent and stochastic approaches for performing

calculations of transport coefficients under high fields; and (vi) the demonstration of GPU portability for the EPW code.

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Fracton self-statistics and modularity

Michael Hermele (University of Colorado Boulder)

Keywords: fractons, braiding statistics, quantum error correcting codes, quantum spin liquids

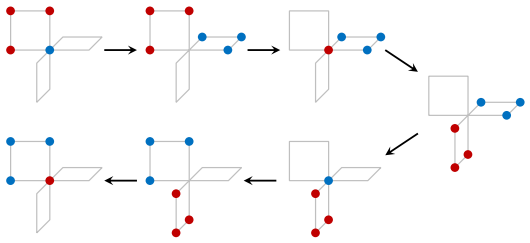
Research Scope

The research to be presented at this meeting focuses on the properties of excitations of fracton phases of quantum matter. By definition, fracton phases have excitations of restricted mobility, with the term "fracton" referring to an excitation that is completely immobile in isolation. Fracton systems are of interest as resources in quantum information processing, for their connections to non-equilibrium dynamics, and as a new and poorly understood class of quantum phases of matter. While description by effective quantum field theories underpins much of the understanding of more conventional quantum phases of matter, the role and existence of useful such continuum theories for fracton systems remains unclear. Fracton phases are interesting in part because they seem to lie beyond the usual paradigms for understanding condensed matter in terms of quantum field theory.

In the well-developed theory of anyons in two spatial dimensions, one gives a mathematical description of fusion and braiding statistics of excitations. Our past work [1] took an important step in this direction by giving, for fracton systems, a mathematical description of fusion that also incorporates mobility of excitations. Moreover, we showed that restricted-mobility excitations can undergo statistical processes analogous to braiding of anyons – these processes are, however, more complicated because they do not conserve particle number. For anyons, any statistical process can be decomposed into full and half braids, which allows for a precise algebraic description of anyon statistics. In fracton systems, by contrast, we do not have a similar understanding of possible statistical processes. More generally, there are few nicely-behaved invariants for fracton phases, which has made it difficult to make sense of a proliferation of examples in exactly solvable models.

Recent Progress

Fracton excitations are immobile in isolation, so it is natural to expect that they do not have any kind of analog of braiding exchange statistics, which after all requires particles to move around in space in order to be exchanged. In recent work [2], we showed that fractons surprisingly can be exchanged via processes that do not conserve particle number, and that these processes are characterized by a statistical exchange phase just as in the exchange of conventional mobile quasiparticles. While mobile particles in three dimensions can only be bosons or fermions, we



Depiction of a "windmill" fracton exchange process. Initially, a blue excitation and a composite of three red excitations are fractons of the same type, which can only move via certain processes that change the particle number. Six steps are shown after which the blue and red excitations are exchanged.

showed that fractons can have more general statistics, and gave an explicit example of a model where the fractons are semions. Self-statistics of fractons is a new and previously unappreciated universal property of fracton orders, and an important step toward a full understanding of all statistical processes for fracton systems.

Fracton self-statistics gives an example of a phase invariant, a simple property that can be used to distinguish different quantum phases of matter. In general, the invariants of fracton phases are not well understood. This goes even for simpler invariants that are only sensitive to mobility of excitations and not to statistics. In work in preparation [3], we have identified a large class of fracton systems for which many new invariants can be constructed. The defining property of these systems is dubbed *p*-modularity, which means that any non-trivial point-like excitation can be remotely detected via braiding with planons (these are quasiparticles constrained to move only within a fixed spatial plane). *p*-modular fracton models include important examples such as the X-cube model, but not Haah's cubic code and other models with fractal geometrical structure. We discovered a family of invariants capturing the mobility properties of *p*-modular fracton models, and used these to identify, for the first time, a Z_2 fracton model that neither has fractal structure nor is a stack of models with X-cube-like mobility (which we define precisely).

Future Plans

We plan to further explore the properties of *p*-modular fracton systems. There are reasons to hope that a full classification of *p*-modular fracton phases might be possible. Important ingredients will be to understand the relationship between *p*-modular fracton systems and gauged subsystem symmetries, and whether there is a simple description of the excitation statistics in *p*-modular systems.

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Nontrivial consequences of non-centrosymmetry in topological and trivial metals

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Keywords: Chiral, non-centrosymmetric, kinematic, superconducting diode effect, localization

Program Scope

Noncentrosymmetric metals have historically been rare in nature. However, fervent research on Weyl semimetals (WSMs) over the past decade has paved a new and prolific route to such metals [1]. Inspired by this development, this project theoretically explores novel phenomena emerging from the interplay between noncentrosymmetry and metallicity and unlock their potential for the next generation of electronic devices.

One half of the program ("chiral kinematics") entails studying the effects of rotation and acceleration on electrons in 3D *chiral* metals – non-centrosymmetric metals that break all improper symmetries besides. One such phenomenon, well-studied in high-energy physics, is the chiral vortical effect (CVE), defined as an axial current that develops in a relativistic chiral fluid upon rotation [2]. This project aims to explore, generalize, and propose ways to harness this effect in settings exclusive to solid-state physics.

The second half ("band asymmetric metals") entails studying non-centrosymmetric metals that are also magnetic but lack Berry phases. These properties skew the energy-momentum relationship for the constituent electrons, and we dubbed them *band asymmetric metals* (BAMs). Conventional metals rely on the absence of skewness for most physical properties such as their response to electromagnetic fields, behavior under disorder and Cooper pairing tendencies. In BAMs, the skewness immediately brings these properties under question. This project aims to explore disorder-induced localization and superconducting phenomena in BAMs.

Recent Progress

Progress on chiral kinematics: So far, we have established one facet of the linear response theory of chiral kinematics, where both the driving and responding fields are velocities. We first derived an expression for the chiral vortical effect and introduced a closely related gyrotropic vortical effect using the Kubo formula. In general, vortical effects are defined as rotational motion driving linear motion along the rotation axis. Our work generalized previous Boltzmann

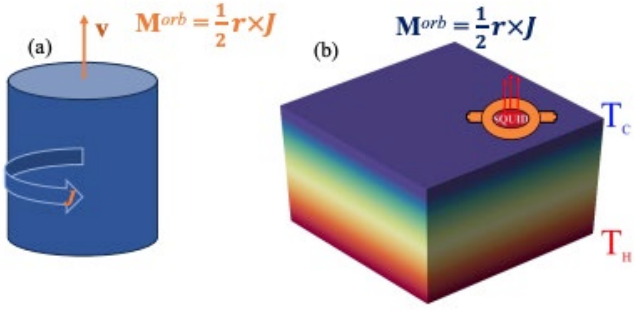

Figure 10: (a) Schematic of the converse vortical effect. (b) Device geometry for observing the effect disguised as a magneto-thermal effect.

and hydrodynamic results for Weyl fermions rotating at constant angular velocity to arbitrary band structures, rotation varying in space and time, and finite relaxation time. In the process, we introduced a momentum-like quantity $\mathbf{Q}^{\mathbf{k}}$, that is the kinematic counterpart of the current operator in electromagnetic response. It resembles the continuum momentum but preserves momentum space periodicity in a lattice. We then proposed converse vortical effects (Fig. 1), defined as linear motion driving rotational motion, and derived relevant Kubo response functions and a semiclassical free energy functional. Interestingly, the free energy includes a modification of the phase space measure from a coupling between Berry curvature and fluid vorticity which is expected be crucial for the full response theory.

Progress on band asymmetric metals: We studied the consequences of proximity-induced conventional superconductivity in BAMs and discovered remarkable behaviors such as an equilibrium supercurrent that evades a no-go theorem (Fig. 2) and a perfect superconducting diode effect (SDE). In addition, we investigated the SDE in Dirac and WSMs and found an enhancement of the effect upon tilting the Dirac/Weyl node, which effectively converts the node into a BAM.

Figure 2: Depositing a BAM wire on a conventional superconductor will generate an equilibrium current, a SDE in general, and a perfect SDE if the band asymmetry exceeds a threshold determined by the critical Cooper pair momentum of the parent superconductor.

In disordered BAMs, we studied weak localization physics in 1D. For perturbative disorder, we analytically found a suppression of weak localization due to mismatched left and right Fermi velocities. Specifically, we found the weak localization correction to conductivity:

$$\sigma^{WL} = -\frac{e^2}{h}\frac{2\pi v\tau}{\sqrt{\frac{l}{l_\phi}+\frac{\delta v^2}{v^2}}}$$

where v is the mean speed of the left and right Fermi modes, τ is the Born lifetime, δv is the difference in their speeds, l is the mean free path and l_ϕ if the phase coherence length. In complementary quench numerics, we saw sharp delocalization tendencies in the Anderson localization length (Fig. 3) and the long-time saturation of the participation ratio after starting from a localized state due to band asymmetry.

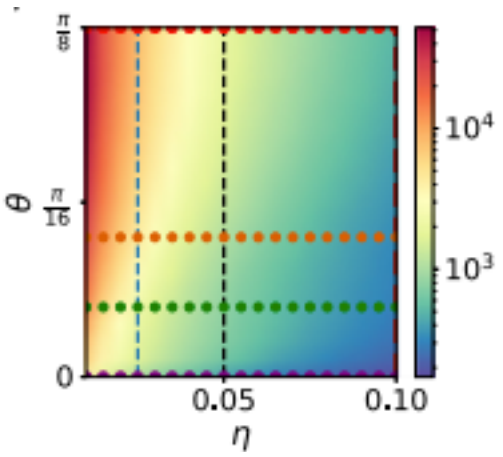

Figure 3: Localization length (color, ξ) as a function of band asymmetry (θ) and disorder strength (η). ξ increases sharply with θ.

Future Plans

We plan to continue and expand our research program on non-centrosymmetric metals through 4 interrelated Thrusts.

In Thrust 1, we will establish a standalone theory of chiral kinematics in the perturbative regime. In other words, we will derive a framework to describe the response of electrons in a chiral band structure to spacetime dependent velocity field $\mathbf{v}(\mathbf{r},t)$. We require $|\mathbf{v}|$ to be much smaller than typical band velocities for perturbation theory to be valid; however, unlike usual approaches, we will *not* rely on analogies between $\mathbf{v}(\mathbf{r},t)$ and the electromagnetic vector potential $\mathbf{A}(\mathbf{r},t)$. We will first study lattice contributions to linear kinematic responses captured by the "Bloch gauge field," a gauge field arising from the freedom to shift lattice momentum by an arbitrary reciprocal lattice momentum. Then, we will focus on linear responses where the driving field, responding field or both fields are momentum fields, i.e., linear or angular momentum. These investigations together will complete the linear response theory of chiral kinematics and set the stage for studying non-linear responses and deriving an effective action.

Thrust 2 will promote chiral kinematics to a quantum regime and unearth various intriguing phenomena. Here, we will begin by solving the "problem zero" of 3D chiral quantum kinematics, namely, calculating the quantized spectrum of a rotating Weyl fermion and describing how the CVE, well-known in the semiclassical regime, emerges in the quantum limit. Then, we will explore a different quantum effect, namely, a quantum anomaly that resembles the famous Adler-Bell-Jackiw anomaly in electromagnetism but is purely kinematic in nature. For both these questions, a naïve calculation paralleling the one in electromagnetism gives a vanishing effect. Our key insight is that the effects emerge from a non-equilibrium state that is stabilized by a delicate interplay between adiabaticity and locality. Having gained a solid understanding of quantum mechanics of chiral kinematics, we hope to predict quantized responses in two different quantum limits, ultrafast rotation and quantum confinement.

Thrust 3 strives to facilitate the development of digital logic based on superconductors instead of semiconductors. For this purpose, we will exploit a unique superconducting property of non-reciprocal metals, namely, a large or even perfect SDE under suitable conditions. Thus, we will examine the SDE in a candidate platform, Rashba nanowire under a magnetic field proximity-coupled to a conventional superconductor. This system has been extensively studied in the search for Majorana fermions in condensed matter and many engineering and materials science challenges have already been solved. This will be followed by the construction of circuits that function as transistors and basic logic gates, thus providing the building blocks for superconducting digital electronics.

In Thrust 4, we will investigate systems that harbor both chirality and band asymmetry, namely, WSMs with tilted bands. In recent years, interest has mushroomed in so-called type-II WSMs, defined by a Weyl node tilted so strongly that a portion of the conduction (valence) band moves

below (above) the Fermi level. Here, we will focus on moderately tilted Weyl nodes that they are still type-I, and study quantum effects in localization phenomena. As a precursor, we will with a tilted type-I 2D Dirac node where weak anti-localization is expected under disorder. The results will pave the way for studying quantum effects in localization in WSMs with multiple tilted nodes. We will be especially interested in thin films, where disorder is marginal in the renormalization group sense. As a result, the competition between weak localization and anti-localization determines whether the films are localized or delocalized in the thermodynamic limit.

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Fourier and fractional neural operators to unveil topological textures in 3D magnetism

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Keywords: Magnetization dynamics, solitons, modulational instability, spin superfluidity

Research Scope

Recent developments in magnetism and magnetic materials enable us to access both far-from-equilibrium and nonlinear physics, including the stabilization of solitons [1,2] and surprisingly fast soliton dynamics [3]. Understanding these phenomena requires a theoretical approach that captures short- and long-range interactions on equal footing. Our research aims to provide such an approach in a manner that is both computationally advantageous and analytically tractable. These qualities are needed to integrate physical models into machine-learning approaches and predict new magnetic materials with tailored properties.

Recent Progress

Pseudo-spectral Landau-Lifshitz model

We introduced a pseudo-spectral Landau-Lifshitz equation (PS-LLE) to describe the magnetization dynamics in the transition between atomistic and continuum regimes. This model does not introduce new physics in the system; instead, it enforces the conservation of both energy and momentum to guarantee a smooth transition between spatial and temporal scales. In other words, the model ensures that the physical dispersion relation of magnons is correctly described across scales. This is similar to the idea of "dispersion engineering" more commonly used in fluid dynamics. The PS-LLE model is given by:

$$\frac{\partial}{\partial t}\mathbf{m} = -\mathbf{m}\times(\gamma\mu_0 M_{\text{eff}}\mathbf{h}_l - \mathcal{F}^{-1}\{\omega(k)\widehat{\mathbf{m}}\}) + \alpha\mathbf{m}\times[\mathbf{m}\times(\gamma\mu_0 M_{\text{eff}}\mathbf{h}_l - \mathcal{F}^{-1}\{\omega(k)\widehat{\mathbf{m}}\})]$$

where $\mathbf{m}$ is the normalized magnetization vector, γ is the gyromagnetic ratio, μ_0 is the vacuum permeability, M_{eff} is the effective magnetization, $\mathbf{h}_l$ contains local field contributions (uniform external field, and anisotropies), and $\omega(k)$ is the convolution kernel that captures the magnon dispersion relation. In Figure 1(a), we have verified the implementation of the model for a 1-dimensional ferromagnetic chain and recovered the known dispersion relation for magnons, shown by a solid red curve. In contrast, the micromagnetic dispersion proportional to k^2 is shown by a dashed red curve, clearly leading to an unphysical energy for short-wave magnons. This artificially high energy typically leads to a mismatch between atomistic and micromagnetic models [1].

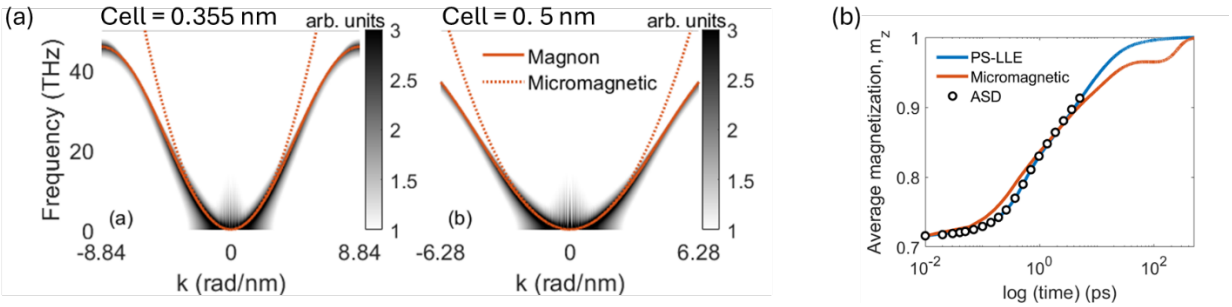

Figure 1. (a) Numerical dispersion from the PS-LLE generated by the spatiotemporal evolution of a delta function. The color scale represents the numerical dispersion obtained from the PS-LLE model, the solid red curve is the magnon dispersion relation, and the dashed red curve is the micromagnetic dispersion relation. (b) Magnon dispersion relation obtained from a grid with a cell-size of 0.5 nm, incommensurate to the lattice constant, demonstrating the grid-independence of the PS-LLE model. (c) Evolution of the spatially-averaged easy-axis magnetization from transient grating. The PS-LLE model (blue curve) follows the atomistic spin dynamic simulation (black circles) while the micromagnetic approach (red curve) fails.

Because the PS-LLE is a spectral approach, grid independence is also expected. This is shown in Figure 1(b) where we use a cell size that is incommensurate to the lattice constant. Notable, the PS-LLE recovers the correct dispersion relation, truncated at the maximum resolved wavenumber.

After recovering the expected dispersion relations, we tested the PS-LLE against atomistic spin dynamics. We modeled an ultrafast transient grating experiment where both short- and long-wave components are present. In these experiments, two ultrafast lasers are interfered creating a tunable spatial pattern that leads to a grating in the sample's magnetization, periodically varying from randomized to ordered distributions. We followed the evolution with the PS-LLE model and atomistic spin dynamics in a quasi-1D configuration. In Figure 1(c) we show the time evolution of the spatially-averaged easy-axis magnetization component as a metric for comparison. The PS-LLE model (blue curve) follows the atomistic spin dynamic simulation (black circles) almost exactly from the same initial conditions. The error was found to be on the order of 0.1%. The micromagnetic implementation is quantitatively different. Note that the time is in logarithmic scale so that the PS-LLE model returns a better description at the sub-nanosecond timescales.

Generalized spin hydrodynamics

Spin hydrodynamics provides a framework to analytically investigate nonlinear states and their stability. Under the PS-LLE model, it is possible to express the equations of motion as an effective fluid. While there is an exact transformation for the micromagnetic equations into Euler equations, the PS-LLE model can be expressed in Fourier space or as convolutions. The dispersion relation of magnons on top of a spin hydrodynamic state determines the stability of perturbations. So far, it is generally accepted that in-plane magnets are stable while uniaxial magnets are unstable. However, under the PS-LLE model, we find that this situation is more complex. In Figure (2), we

show the stability of perturbations by computing the magnetic Mach number when the easy-plane magnetization is 0.5. In-plane magnets (a) are stable for more than half the first-Brillouin zone, but shorter wavelengths become modulationally unstable (gray area, MI), indicating that solitons may be favored. In contrast, uniaxial magnets (b) have the opposite behavior and indicate stability for short wavelengths. In addition, the Mach number is greater than one for textures shorter than the exchange length. This indicates that defects, either magnetic or nonmagnetic, can give rise to phase slips. These conditions are expected to be relevant to understanding the nucleation and stabilization of topological textures with atomic-scale defects and at ultrafast timescales using the PS-LLE model.

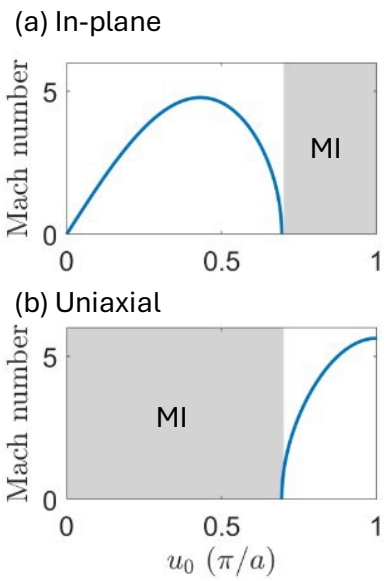

Figure 2. Mach number obtained from generalized spin hydrodynamics for (a) in-plane and (b) uniaxial magnets. The gray regions indicate modulational instability (MI), where the amplitude of waves grow exponentially, i.e., due to the complex dispersion relation. The Mach number is mostly larger than 1, indicating that phase-slips are prone to occur due to defects.

Future Plans

1) The PS-LLE will be extended to three dimensions plus the inclusion of the long-range nonlocal dipole field. The nonlocal dipole field is typically described in Fourier space, thus being naturally integrated with the PS-LLE. This numerical tool will be used to verify analytical predictions and to investigate the nucleation of topological objects.
2) Far-from-equilibrium conditions will be investigated to understand the evolution of topological defects in both in-plane and out-of-plane magnets. It must be noted that in-plane materials remain poorly investigated in far-from-equilibrium conditions. Short-wavelength instabilities would open the possibility of understanding the nucleation and stabilization of topological defects in in-plane materials, such as topological magnetic monopoles [2].
3) The PS-LLE model will be integrated with machine learning algorithms and contrasted with "Fourier neural operators" (FNOs) [5]. One scope of this research is to retrieve the dispersion relation based on a desired outcome, guiding the search for novel magnetic materials. We will investigate how to transform activation functions used in machine learning algorithms to recover the dispersion relation or propose alternative machine learning algorithms that intrinsically consider the dispersion relation in such activation functions and matrix weights.
4) Other forms of magnetic order will be included in the PS-LLE. We are interested in antiferromagnets and ferrimagnets due to their use in far-from-equilibrium experiments and potential applications. Such multi-sublattice materials will also provide an ideal testbed to further extend our approach to consider, e.g., altermagnetism.

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Topological superconductivity in the quantum Hall regime

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Keywords: topological superconductivity, Majorana zero modes, spin-orbit coupling, quantum Hall effect

Research Scope

The search for topological superconductors is motivated by the prediction that they support Majorana modes, which are quasiparticles that are their own antiparticles and which obey non-Abelian statistics in which successive exchanges of particles do not always commute. We have investigated whether an ordinary superconductor can be turned topological by application of a magnetic field. For this purpose, we obtain self-consistent solutions of the mean-field Bogoliubov-de Gennes equations (a large set of nonlinearly coupled equations) for electrons with an attractive interaction subjected to a magnetic field. The nature of the superconductivity and of the Abrikosov lattice is an outcome of the solution.

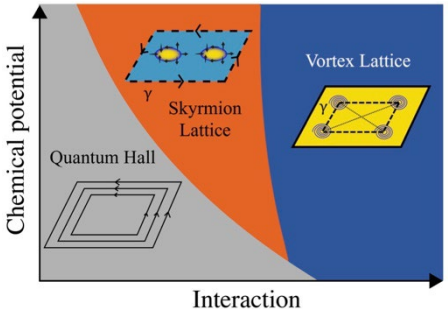

Fig. 1: The schematic phase diagram of spinless electrons in a magnetic field with nearest neighbor attractive interaction.

In [1], we consider a system of spinless electrons on a lattice with nearest neighbor attractive interaction (which models a p-wave superconductor) subjected to a magnetic field and find a rich phase diagram as a function of the chemical potential, magnetic field and the interaction (Fig. 1). As the strength of the attractive interaction is increased, the system first makes a transition from a quantum Hall phase to a superconductor that is fully gapped and has topological chiral edge current, characterizing a topologically non-trivial state. The magnetic field is accommodated through a lattice of skyrmions, where a skyrmion has h/4e flux (half of a superconducting flux quantum h/2e) associated with it. This is followed by a vortex lattice phase in which the h/2e vortices carrying Majorana modes form a lattice. The spectrum of this phase contains a low-energy Majorana band arising from the coupling between neighboring vortex-core Majorana modes, but it does not have chiral edge currents. For certain parameters, a dimer vortex lattice occurs with no low energy Majorana band.

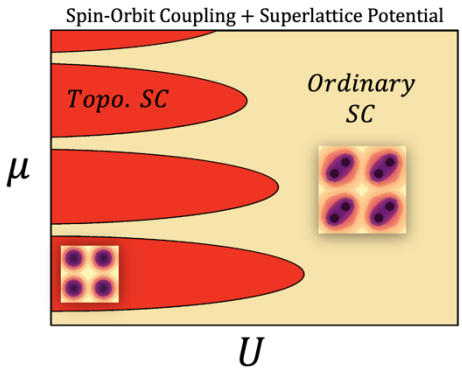

Fig. 2: Schematic phase diagram of an s-wave superconductor with spin-orbit coupling, subjected to a periodic superlattice potential, and exposed to a magnetic field.

In [2] we ask whether a two-dimensional superconductor with ordinary s-wave pairing can be rendered topological by the application of a strong magnetic field. We find that the topological

superconductivity is facilitated by a combination of spin-orbit coupling and a coupling to an external periodic potential which gives dispersion to Landau levels and which also distorts the Abrikosov lattice. As shown in Fig. 2, the broadened Landau levels support, for a range of parameters, topological superconductivity with Majorana zero modes. This superconductivity is typically accompanied by a lattice of h/e vortices, as opposed to the usual h/2e Abrikosov vortices. We believe that our work suggests a realistic proposal for achieving topological superconductivity, as well as a helical order parameter and unusual Abrikosov lattices.

Recent Progress

The objective of the grant is to explore pairing in the quantum Hall regime in two contexts: pairing that arises naturally at certain filling factors and pairing that can be engineered by either proximity-coupling a QHE state to a superconductor or by exposing a superconductor to a magnetic field. These states have been predicted to support excitations, such as Majoranas, that obey non-Abelian braid statistics.

The abstract (above) describes our work in which either a p-wave or an s-wave superconductor is exposed to a magnetic field. We have also made progress on how pairing between composite fermions can be induced naturally by varying certain parameters. Our previous calculations have suggested that at even-denominator filling factors, a Fermi sea of composite fermions is stabilized when the interaction between the electrons is strongly repulsive at short distances, but a transition into a paired-composite fermion state may occur as the strength of the short-range repulsion is reduced. In [3] we predicted that pairing at half and quarter filled Landau levels can be achieved by increasing Landau-level mixing. This prediction was confirmed experimentally [Chengu Wang et al., Phys. Rev. Lett. 131, 266502 (2023)] at quarter filling in hole type systems where the Landau level mixing is significant.

In [4] we predicted that the localization length exponent associated with the plateau transitions in the fractional quantum Hall effect (FQHE) is the same as that in the integer quantum Hall effect, and thus universal. This paper has already motivated three new experiments. In Phys. Rev. Lett. 130, 226503 (2023), Madathil et al. have found the exponents to be non-universal. However, further experiments from the same group (unpublished) and by S. Kaur et al. (arXiv:2312.06194) have shown that the exponents are universal provided that the disorder is such that it causes short range scattering. A similar dependence on the nature of disorder had been obtained about two decades ago for plateau transitions in the integer quantum Hall effect. These studies represent real progress on this longstanding issue, which is important example of localization-delocalization transition in a strongly correlated, topological system.

The graduate student Jonathan Schirmer (supported by this grant) was encouraged to work independently toward the end of his PhD, and I was pleased to see that he has had two publications in which I am not a coauthor.

Future Plans

A new student has joined the group and is studying the model of a bilayer superconductor with a magnetic field parallel to the layer. Landau-Ginzburg theory predicts, with increasing magnetic field, a transition into an FFLO phase, which has been observed experimentally. The objective is to go beyond the Landau-Ginzburg theory to determine if the pair momentum remains spatially uniform, and if addditional phases may occur. Preliminary results indicate an "FFLO stripe phase." We will also study proximity induced superconductivity in the quantum Hall regime within this model. This work is in collaboration with C.-X. Liu.

Certain theoretical papers have considered superconductivity arising out of flat bands in time-reversal invariant systems and argued that there exists a lower bound on the superfluid weight that depends on the Chern number. This sets the energy scale for phase fluctuations and is responsible for the Meissner effect and persistent supercurrents. We plan study the superfluid weight for Cooper pairs formed in flat bands in a magnetic field, namely Landau levels, using self-consistent mean-field theory. We will study how the superfluid weight evolves under the application of a periodic potential that pins the Abrikosov vortex lattice and also induces dispersion in the Landau levels. Jonathan Schirmer made progress on this before he finished his PhD and is continuing to work on it.

In [1] and [2] we have studied the interplay between the integer quantum Hall effect and superconducvitity. The next step would be to explore what structures arise when we couple *FQHE* and superconductivity. A straightforward extension of [1] and [2] is not possible, because the FQHE requires repulsive interactions whereas superconductivity attractive. There are two possible ways to circumvent this hurdle: use an interactioin that is repulsive at short distances but attractive at large distances, and to consider spatially separated domains of the two as a function of the coupling between them. We will pursue both.

With the recent discovery of FQHE in zero magnetic field (dubbed fractional quantum anomalous Hall effect or fractional Chern insulator), coupling FQHE and superconductivity is now well within experimental reach, and much new physics is expected in the coming years. We are hoping to make predictions and also to be in a position to address any surprises that experiments present.

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Artificial intelligence and data science enabled predictive modeling of collective phenomena in strongly correlated materials

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Keywords: machine learning, effective models, strongly correlated electrons, inelastic scattering.

Research Scope.

This project is leveraging advanced computational methods, including machine learning (ML), artificial intelligence (AI), and data science, to advance predictive modeling of correlated quantum materials and accelerate discovery in quantum materials research. Our work is organized around three primary aims: 1) generating and validating novel low-energy effective models for correlated quantum materials, 2) accelerating the time to solution for these models using advanced computational methods, and 3) creating novel workflows integrating theory and experiment, with a particular focus on resonant inelastic x-ray scattering (RIXS) and inelastic neutron scattering (INS) experiments. These new methodologies are being prototyped in studies of correlated quantum materials, with an emphasis on materials hosting a mix of localized and delocalized excitations. Examples include systems containing magnetic 4d, 5d, and 4f ions, unconventional superconductors, and materials with complex collective phenomena and nontrivial topological properties.

Recent Progress.

Effective Hamiltonians for unconventional superconductors – We have conducted several studies aimed at determining the electronic models of unconventional superconductors. In paper 1, we performed O K-edge RIXS measurements of $La_4Ni_3O_8$ and $La_{2-x}Sr_xCuO_4$ with equivalent effective doping (x = 1/3) and extracted effective multi-orbital Hamiltonians for these materials by matching the spectra to small cluster calculations (see Fig. 1). The results demonstrated that the nickelate has a mixed charge-transfer/Mott-Hubbard character with $U \approx \Delta$, which explains the moderate magnetic superexchange observed previously. In paper 2, we then studied $La_4Ni_3O_8$'s charge ordered phase using Ni L_2-edge RIXS measurements and analyzed the data by introducing charge order via the application of a site-dependent potential difference to our prior model. The calculations identified the involvement of

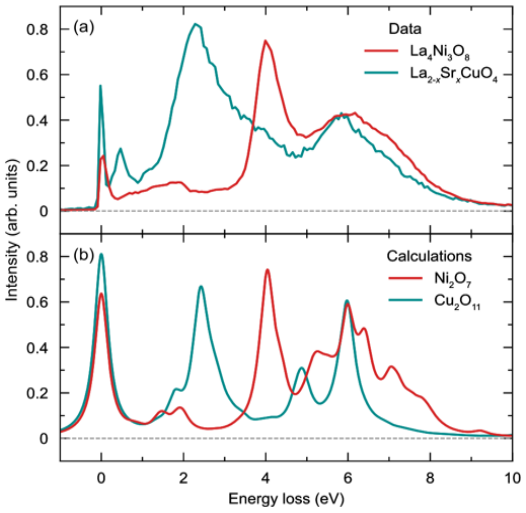

Fig. 1: (a) O-K edge RIXS measurements and (b) calculations for $La_4Ni_3O_8$ and $La_{2-x}Sr_xCuO_4$ with equivalent effective doping (x = 1/3), adapted from Paper 1.

the Ni $3d_{x^2-y^2}$, $3d_{3z^2-r^2}$, and O $2p_\sigma$ orbitals; however, the hybridized planar orbitals primarily shape the charge distribution, leading to a Ni site-centered charge order. We have also conducted two studies extracting an effective extended Hubbard model description of the cuprate ladders $Sr_{14-x}Ca_xCu_{24}O_{41}$ from RIXS and INS data, finding evidence for an *attractive* nearest-neighbor interaction like that identified recently in doped spin-chain cuprates [1].

Reliably downfolding the Kondo Lattice Model (KLM) – In paper 3, we have introduced an ML protocol for extracting effective models from high-energy Hamiltonians and deployed it in applications to the KLM. The KLM describes localized spins of itinerant (conduction) and localized electrons interacting via the Kondo exchange J; its low-energy effective model retains only the localized spin degrees of freedom with effective spin-spin interactions obtained by integrating out the conduction electrons. Deriving the form of these interactions is challenging because they are non-analytic functions of J, which precludes perturbation theory beyond the second order. Our approach solves this problem and allows us to extract effective two- and four-spin interactions, obtain an effective spin model that reproduces the KLM's phase diagram, and reveals effective four-spin interactions that stabilize a field-induced skyrmion crystal phase. Crucially, we can perform efficient calculations of the static and dynamic properties of the ML-derived spin model at a significantly reduced cost, thus accelerating discovery (see also paper 4).

A flexible and efficient open-source implementation of DQMC – We have put considerable effort into developing and releasing SmoQyDQMC.jl, a fast and efficient Julia implementation of the DQMC algorithm (see Ref. 2 and paper 5). This package supports a broad class of Hamiltonians on arbitrary lattice geometries with both Hubbard and *e*-ph interactions. It uses hybrid Monte Carlo (HMC) sampling to enable simulations of realistic low-energy dispersive optical or acoustic phonons and general momentum-dependent *e*-ph couplings. The package has a novel scripting interface, allowing users to tailor it to their needs and directly integrate it into new AI-powered workflows. We have subsequently deployed this tool in a range of studies including examining stripe order in the three-band Hubbard and Hubbard-Su-Schrieffer-Heeger (SSH) models and superconducting correlations in both the two-dimensional SSH and bilayer Hubbard models. We are also currently using these packages to implement a quantum parallel tempering algorithm to improve the autocorrelation time of *e*-ph simulations in the strong coupling limit.

Efficient simulations of magnetic Hamiltonians – We have extended the theory of generalized SU(N) spin dynamics and developed highly efficient algorithms to simulate it (see paper 6). While SU(N) spin dynamics was originally formulated as a generalization of the Landau-Lifshitz equation, we discovered that significant gains can be made using a Schrödinger picture, where the dynamics of the problem are governed by a nonlinear Schrödinger equation with a canonical Hamiltonian structure. This makes it possible to design integration methods that exactly respect the symplectic geometry of phase space. Such integrators enable long-time dynamical simulations with zero energy drift and are a powerful basis for the measurement of long-time dynamical spin-spin correlations (small energy transfer). Data from these simulations often produce remarkable

agreement with INS data [3]. All algorithms designed in this project have been implemented in the open-source code, Sunny, where they can now serve as fast solvers for the dynamical spin correlation functions of magnetic insulators, a crucial component for extracting model Hamiltonians from scattering data. We have also leveraged these methods to discover new mechanisms for stabilizing CP^2 skyrmions in paramagnetic systems (see paper 4).

Learning off-diagonal correlations through structural complexity measures – We have demonstrated that a multi-scale complexity measure can be used to identify phase transitions involving subtle off-diagonal order using only projective measurements in a diagonal basis. This structural complexity measure uses ideas from the renormalization group flow and is based on dissimilarity of patterns at different scales. It can pinpoint the transition to and from the bond ordered wave phase of the 1D extended Hubbard model with an off-diagonal order parameter, sandwiched between diagonal charge and spin density wave phases, using only diagonal descriptors (see Fig. 2 and papers 7 & 8). We also studied the structural complexity of snapshots of the 2D Fermi-Hubbard model as a function of doping and temperature using projective measurements generated by DQMC simulations and taken from optical lattice experiments for comparison (see paper 8). These complexities are linked to physical observables like the entropy and double occupancy, and their behavior captures the development of correlations and relevant length scales in the system. These results demonstrate that data-driven techniques can circumvent limitations in measuring off-diagonal correlation functions, which can enable new experimental studies of exotic phases.

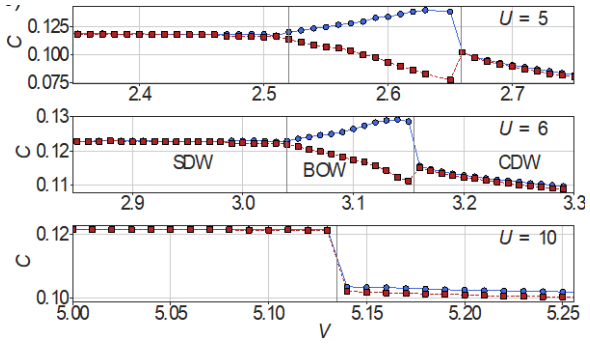

Fig. 2: Multi-scale structural complexity as a function of nearest-neighbor interaction V in the half-filled 1D extended Hubbard model.

Future Plans

We now aim to automate our model extraction methods as applied to inelastic scattering data, focusing on new workflows for RIXS experiments. We have begun work on automating the extraction of crystal field Hamiltonians from RIXS measurements, using the EDRIXS software as the direct solver. We also intend to develop new solvers for the RIXS cross-section based on QMC and DMRG methods, which will allow us to treat more extensive systems and determine low-energy effective models from the measured collective excitations. We will prototype these methods in applications to topical research problems, including unraveling the electronic structure of the nickelate bilayer superconductors and identifying the role of *e*-ph coupling in the cuprates.

We are also pursuing several promising applications of ML and AI methods to improve many-body simulations. Recurrent neural networks. (RNNs) have recently emerged as efficient and versatile approaches for encoding many-body wave functions [4], which we would like to use for improved variational Monte Carlo (VMC) ground state calculations of canonical Hubbard and Hubbard-Holstein models. We have also begun examining two methods for accelerating many-

body simulations. The first is a self-learning DQMC frameworks, where an ML model is trained to predict the forces driving the fictitious dynamics of the lattice in HMC sampling of e-ph Hamiltonians. The second focuses on creating new ML model surrogates for improving rates of convergence in quantum cluster methods like DCA, which build directly on proof-of-concept studies we conducted earlier in this project.

Finally, we are also exploring new methods for analytically continuing QMC data to the real frequency axis. Previously, we explored data-driven ML frameworks and applications of genetic algorithms. Next, we will research physics-informed ML algorithms that will enforce the known analytic structure of causal correlation functions and simplify the computer's learning objective. We are particularly interested in applying these methods in calculating the superconducting gap function, which provides important insight into a system's pairing mechanism.

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Quantum Measurements and the Non-Equilibrium Frontier

Vedika Khemani

Keywords: many-body quantum dynamics, measurement induced phase transitions, quantum state learning, quantum transport

Research Scope

Synthetic quantum devices, with their unprecedented controllability, have opened a vast new domain for studying and harnessing many-body quantum physics. The operation of such devices naturally implements dynamics that is highly out-of-equilibrium, combining unitary time evolution with *controlled* quantum-coherent *measurements*, giving us experimental access to "active" quantum matter for the very first time. In condensed matter physics, measurements have typically played a passive role, functioning as *probes* of static correlations. However, measuring a quantum system can also *create* novel correlations, so that measurements can be promoted to playing an active role i.e. forming an intrinsic *part* of the dynamics. My current research includes two complementary thrusts, motivated by this dual role of measurement as probe and protagonist.

(i) We explore how novel capabilities for controlled and site-resolved measurements in quantum simulators can be used to learn properties of complex quantum states efficiently, and to reveal new order parameters for many-body phenomena. We find that these new probes can reveal novel and distinct dynamical universality classes.

(ii) We study novel measurement-induced dynamical phenomena in which the observer plays a central role. We find new phases of quantum dynamics sculped by measurements and characterized by new information theoretic order parameters.

Recent Progress

I. Measurements as Probes: Advances in Quantum State Learning and New Order Parameters from Measurement Snapshots

A. Optimizing quantum state learning via insights from quantum dynamics

The development of controllable quantum simulators has enabled the creation of complex and highly entangled quantum states in laboratory settings These advances raise the issue of how to efficiently characterize such quantum states. Full quantum state tomography requires exponentially many measurements in the size of the system, motivating the need for more scalable and efficient state-learning protocols. Recent progress in this direction has come from the development of *classical shadows*, a method to extract many physical properties of states with a dramatically smaller number of measurements [1]. Classical shadows is a protocol developed in quantum information which uses “randomized measurements” to form a compact representation

of a many-body quantum state, where the measurement basis is randomized by a random unitary operation U chosen from a suitable ensemble. Results existed on the best choice of the unitary ensemble for learning strictly local (one-body) observables and fully global N-body observables. However, there are many condensed matter settings in which one may want to learn observables intermediate in size between these limits (e.g. string order parameters).

In [2], we shed light on the inner workings of classical shadows protocol by making connections to foundational ideas in quantum dynamics on the spreading and equilibration of operators. In particular, we related the sample complexity (the number of measurements needed to learn a certain observable) to the dynamics of the observable under the randomizing unitary. Using these insights, we formulated an optimal protocol (choice of random unitary ensemble) which gives an exponential gain in sampling complexity for learning k-body observables that are intermediate in size between one-body and fully global. Such observables are essential for characterizing complex correlations in many-body phases. More generally, by connecting a practical task in quantum information to fundamental ideas in quantum many-body dynamics, our work paves the way for the development of highly-optimized applications of classical shadow protocols for near-term quantum devices.

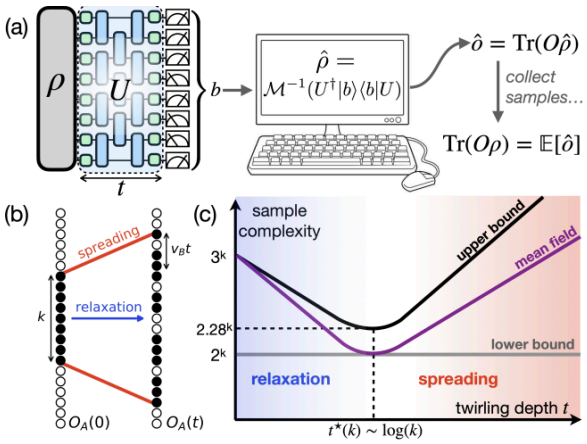

Fig from [2]. (a) Illustrates the classical shadows protocol. A state is measured after randomizing with a twirling unitary U and the measurement data is classically processed to obtain Pauli expectation values. (b, c) The state learning problem is related to the dynamics of operator spreading and relaxation to obtain an exponential gain in the sampling complexity.

B. New order parameters from measurement snapshots

Traditionally, the dynamics of a system is characterized by thermalization properties (i.e. is the system thermalizing or many-body-localized etc.) or by transport properties (diffusive vs. subdiffusive etc). More recently, new observables accessible to quantum simulators have revealed new layers of depth to the question of quantum thermalization. For example, while hydrodynamics is traditionally a property of the expectation values of local conserved charges, experiments in platforms such as ultracold atoms are not limited to measuring expectation values, but can instead take simultaneous 'snapshots' of *all* the particles in a system. These give access to the *'full counting statistics'* or the full quantum distribution of charges, whose first moment is related to transport, but the higher moments go beyond and contain much more information. In recent work, we have shown that various spin chains with diffusive spin transport on average can nevertheless belong to distinct dynamical universality classes revealed by the statistics of snapshots [3].

This work furnishes a fundamental new insight: systems with the same hydrodynamics can nevertheless belong to distinct universality classes, and these distinctions can be probed using new observables that are natural to near-term simulators. This insight can be applied to various settings in future work, for example, in near-integrable or localized models.

II. Active Measurements: New Phases of Monitored and Interactive Dynamics

A. Learnability Phase Transitions in Monitored Dynamics

Monitored quantum dynamics combining unitary evolution and controlled measurements have been a subject of intense recent interest. In this type of evolution, the role of the *observer* is central: the observer's measurements shape the dynamics of the system and drive it to sharply different possible ensembles of late-time states, with 'measurement-induced phase transitions' (MIPTs) defining a new paradigm for phase structure in open systems away from equilibrium. Despite the fact that the transition is driven by the observer's measurements, the traditional formulation of MIPTs is in the form of the entanglement properties of quantum trajectories of the *system*.

Instead, in [4], we study monitored dynamics *from the point of view of the observer* who has access to the classical measurement outcomes, but not to the quantum many-body system. Once again, we marry works in quantum dynamics with results in quantum information. We find novel classes of MIPTs formulated as *learnability* transitions within the framework of classical shadows (discussed in IA above). We show that a measure of information flow from the quantum system to the classical measurement record—the informational power—undergoes a sharp phase transition. This transition determines the observer's (in)ability to learn properties of an unknown initial quantum state of the system from measurements. This introduces a novel class of information-theoretic transitions in monitored quantum dynamics. Several existing formulations of the MIPT (including with symmetries) can also be understood within our framework, which also has the advantage of connecting the large bodies of work on classical shadows and monitored dynamics. This also furnishes a promising new approach for constructing new information-theoretic order parameters of monitored dynamics

B. Entanglement and Absorbing State Transitions in Interactive Quantum Dynamics

Quantum simulators can not only make controlled measurements, but can also perform controlled feedback operations based on the results of measurement outcomes. In [5], we explore dynamics which combine measurement and feedback, wherein one attempts to (locally) *use* measure outcomes to steer a quantum system towards a target many-body state. We study the resulting phase diagram as a function of measurement and feedback rates. We find an *absorbing phase* when the steering succeeds, and the absorbing state transition can be well separated from the measurement-induced entanglement phase transition. We clarify that the former is a transition in a trajectory averaged density matrix (and hence experimentally accessible), while the latter is a transition in *individual* quantum trajectories conditional on measurement outcomes, and hence much harder to observe experimentally because of the exponential cost of postselecting a given trajectory. As an aside, we note that in the prior 2021 Abstract we reported on theoretical approaches to mitigate this postselection problem of observing the measurement induced entanglement phase transition using a novel spacetime duality approach. We successfully implemented this approach in collaboration with the Google Quantum AI group and reported a large experimental demonstration of this transition ([3] in Publication List).

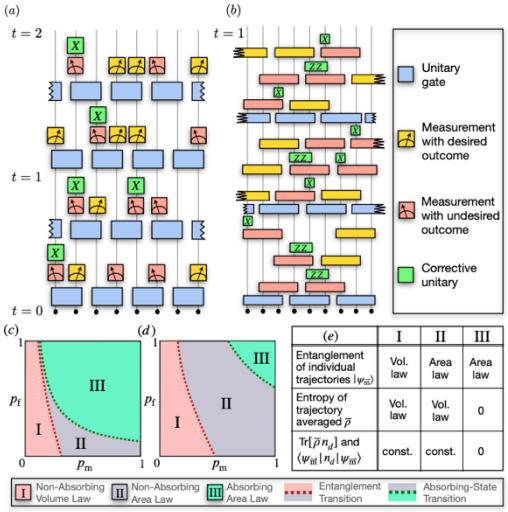

Fig from [5]. Monitored quantum circuits with feedback, and their phase diagrams showing entanglement and absorbing state transitions.

Future Plans: There are many directions we are exploring that build on these approaches of using the ingredients of controlled measurements and feedback. In particular, we are thinking about phases of open quantum systems in the language of error correction (which involves noisy dynamics, controlled measurements of syndromes and feedback, and displays a sharp threshold transition as a function of error rate). We are also thinking about various directions on dynamics in integrable, scarred and many-body localized systems (some results on these have not been discussed in this report, but are listed in the publications).

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Modeling Light–Matter Interaction at the Nanoscale

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Keywords: Quantum materials, light–matter interaction, computational electrodynamics, multiphysics simulation

Research Scope

Modeling light–matter interactions at the nanoscale requires accurate handling of both quantum and electromagnetic systems. Such models must self-consistently couple electrodynamics with quantum electronic transport with full account of the phenomena arising from new materials, low dimensionality, small size, edge topography, quantum geometries, substrate choice, and impurity placement. We present recent progress on the development of a multiphysics modeling framework that responds to this challenge.

Recent Progress

The control of light–matter interaction at the nanoscale is a grand challenge that cuts across modern photonics, plasmonics, and optoelectronics. Nonlinear optics offers a promising platform for controling and manipulating light at the nanoscale for nanophotonic applications. It relies on matter-mediated photon-photon interactions, which are intrinsically very weak. Therefore, realizing nonlinear nanophotonics requires new materials and structures that will enhance nonlinear optical effects.

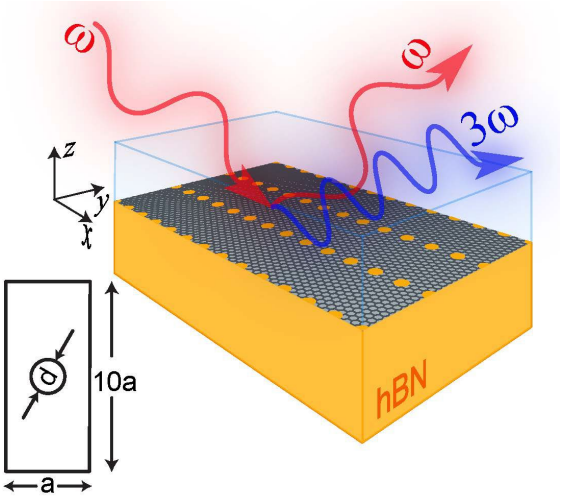

Fig. 1: Schematic of a graphene nanomesh, placed on an hBN substrate. Incident light with in-plane TM polarization (meaning nonzero electric-field component along the plasmon-propagation direction) causes linear as well as third-order nonlinear optical responses. Inset: The unit cell of the rectangular superlattice: The lattice constants along the x and y axes are a and $10a$, respectively. Antidot diameter is d.

In [1], we used the density-matrix formalism and theoretically showed that graphene nanomeshes (GNMs)—graphene sheets patterned with antidots (Fig. 1)—have large plasmon-enhanced nonlinear optical response. GNMs can be designed to behave as quasi-one-dimensional plasmonic crystals in which plasmons with large propagation lengths are efficiently excited. The associated third-order Kerr and third-harmonic-generation susceptibility can be as high as 10^{-7} and 10^{-9} m^2/V^2, respectively, over the mid-to-near-infrared frequency range. Furthermore, carrier-density tuning in GNMs can flip the propagation direction of plasmonic waves and enables bidirectional switching of optical signals, which makes GNMs a promising core material for nonlinear optics.

Figure 2(a) shows the loss function as a function of the Fermi level and frequency. The bright narrow strips correspond to plasmon resonances. For each diffraction order, there is a plasmon branch comprising a pair of bright narrow strips: forward-propagating (FWD) and backward-propagating (BKWD) modes along the x-direction. The splitting of forward and backward propagation comes from the asymmetry with respect to the polarization of the input light and can be analytically calculated. For the Fermi energies and frequencies of interest, this splitting is about 5 meV. Figures 2(b)–(d) show the loss function, third-order Kerr susceptibility and thirt-harmonic-generation (THG), respectively, for the FWD modes as a function of frequency and for a fixed Fermi level. For the Fermi level equal to 1 eV, the Kerr and THG susceptibilities are as high as 10^{-7} and 10^{-9} m^2/V^2, respectively. The loss function in Fig. 2(b) quantifies plasmonic field enhancement, and can be as high as 300–400 at resonance. While plasmon enhancement increases the GNM optical nonlinearity to unprecedented large values, its effect is narrowband. Owing to the long propagation length of plasmons, the loss-function peaks at plasmon resonances are narrow and, therefore, plasmons significantly enhance the nonlinear optical response, but over a narrow frequency range. The nonlinear optical response can be broadened by tuning the plasmon resonances via a back-gate voltage.

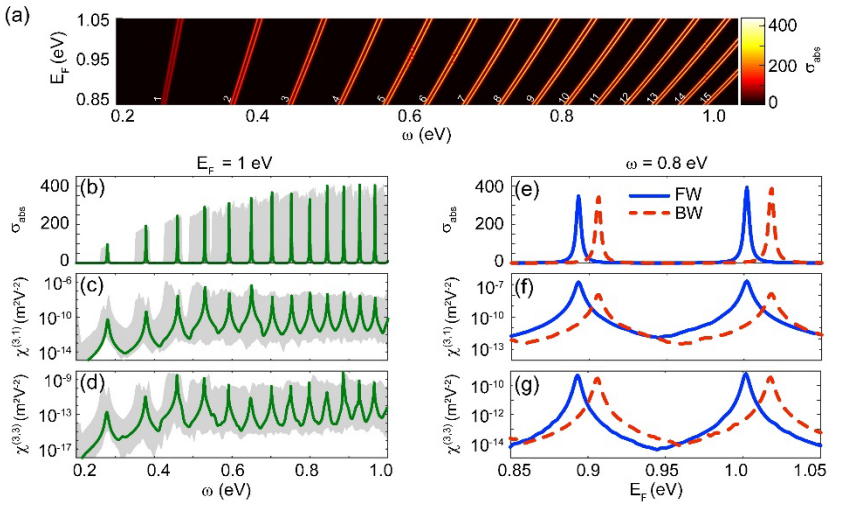

Fig. 2: (a) The GNM loss function as a function of the Fermi energy and frequency. Each branch comprises two modes: forward- and backward-propagating waves. (b) Loss function, (c) magnitude of the third-order Kerr susceptibility, and (d) magnitude of the third-harmonic-generation (THG) susceptibility for a fixed Fermi energy (EF = 1 eV) as a function of frequency for forward-propagating modes. The gray-shaded areas show the corresponding quantities calculated for the Fermi energy in the range of 0.85 to 1.05 eV. (e) Loss function, (f) magnitude of third-order Kerr susceptibility, and (g) magnitude of the THG susceptibility for a fixed frequency (ω = 0.8 eV), as a function of the Fermi energy for forward- and backward-propagating modes.

Multiphysics Simulation of Light–Matter Interaction Using FiPo FDTD and DuPo FDTD

In [3], we introduced a novel self-consistent field–potential (FiPo) FDTD algorithm using a first-order time derivative **A**–ϕ formulation in conjunction with the traditional **E**–**H** Maxwell's equations. The first-order formulation retains the key benefits of standard FDTD while enabling the calculation of potentials needed for integration with various quantum-transport frameworks. By coupling the FiPo FDTD solver with a quantum transport solver, FiPo FDTD enables the calculation of potentials for the quantum transport solver, and the current and charge densities can

be fed back into FiPo FDTD as sources. Additionally, we have formulated a complex-frequency shifted form of the convolutional perfectly matched layer, which we will simply refer to CPML here. In terms of speed, accuracy, and the quality of absorbing boundary conditions, FiPo FDTD is on par with the state-of-the-art of traditional FDTD, which is the mainstay of computational electromagnetics. FiPo FDTD computational advance is particularly useful for calculating the potentials from known sources that are used to drive quantum systems some distance apart (e.g., radiation problems) and where retardation effects must be carefully accounted for. Figure 3 presents wave scattering from a conducting cylinder as calculated using FiPo.

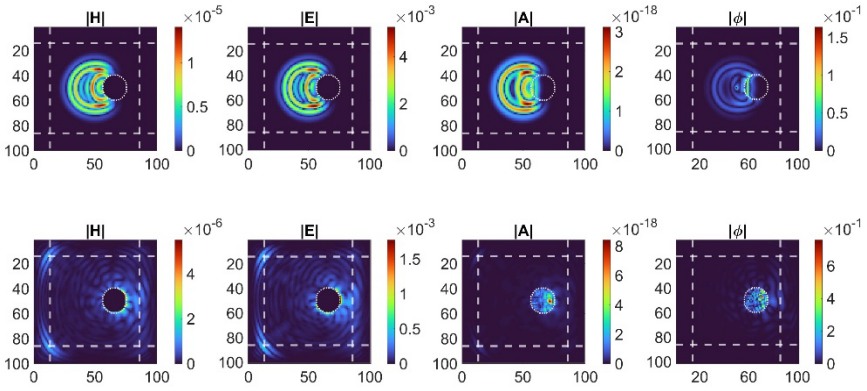

Fig. 3: Snapshots of the magnitudes of electromagnetic fields sourced by a differentiated Gaussian Az pulse at the center of the domain, as calculated by FiPo Hybrid. The wave scatters from an off-center conducting PEC disk identified by a white dash line. Ten-cell-thick CPML boundary surrounds the domain. The snapshots are taken (a) after 200 time steps, which is before the wave reaches the boundary, and (b) after 300 time steps, which is after the wave has reached the boundary and has been absorbed by the CMPL.

In [4], we introduced dual-potential (DuPo) FDTD for accurate treatment of near-field phenomena, basically trying to self-consistently and simultaneously couple quantum transport and electrodynamics *where the quantum transport is happening and when it is actually sourcing the electromagnetic fields*. The essence of the electromagnetic arm of the simulation is the dual-potential outlook on Maxwell's equations. In addition to the standard magnetic vector potential **A** and electric scalar potential ϕ, we introduce the electric vector potential **C** and the magnetic scalar potential ψ. This formulation is not unknown, but is far from standard, and has not been previously numerically implemented in any systematic, efficient, or scalable way. We also show the first fully coupled simulation of device-scale time-resolved fully coupled quantum transport and electrodynamics on the case of electrically fed bowtie nanoantenna in Figure 4. Note that the tunneling current across the feed gap, which is a parasitic phenomenon happening in very small antennas, gets naturally captured in this self-consistent simulation. This is something no widely used electromagnetic simulator, conventional or otherwise, can do, and the simulation is fully time-dependent and can thus be used for a variety of broadband excitations to characterize the frequency response of quantum materials and systems.

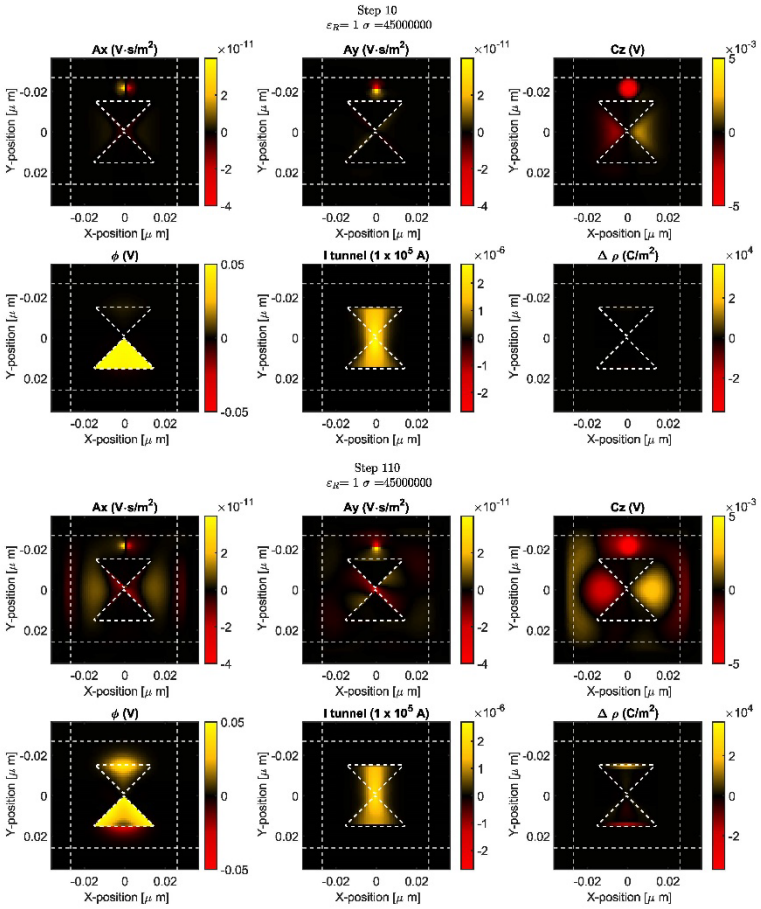

Fig. 4: Example of a fully-coupled self-consistent DuPo FDTD and quantum transport loop. The white dashed outline indicates the boundary of the PML region and the region of high conductivity (metallic patches). In panel a) we see the situation briefly after starting the simulation (10 timesteps). ϕ is set to be positive on the bottom patch of the bowtie antenna (bottom left), resulting in electron tunneling from top to bottom (bottom middle). Additionally, a radiating dipole source is placed above the metallic patch. The dipole source is scaled down to be very small in magnitude, so we can see the interference effects with the small solenoidal contribution of the tunneling current density. Panel b) shows the simulation after 110 timesteps, and we can now see that electrons (negative charge) have moved from top to bottom (bottom right), resulting in a different potential distribution (bottom left). Dur to potential difference between patches decreasing, tunneling current is smaller (bottom middle).

Plans for the upcoming funding period:

Now that DuPo FDTD is ready, coupling of Wigner solver with DuPo FDTD electrodynamics will be underway for full integration of quantum transport with dissipation and electrodynamics. We also plan to release the code base for FiPo and DuPo.

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Spin transport in superconducting systems and magnetic heterostructures

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Keywords: Spin superfluids, magnons, magnetic interfaces.

Research Scope

The project is primarily aimed at gaining a deep understanding of spin currents and how they behave in the settings of superconductor-ferromagnet hybrids, spin-triplet superconductors, and magnetic heterostructures. While grounded in similar spintronics principles, the project encompasses a broad spectrum of scenarios, ranging from the analysis spintronic effects in conventional magnetic heterostructures that utilize heavy metals to the investigation of spin currents in the context of superconductor-ferromagnet hybrids and spin-triplet superconductors. The spintronics foundational principles continue to serve as the catalysis for further developments, and the integration of innovative materials, such as quantum materials, has broadened our comprehension of the underlying physics. In this project, we expect that applying these core principles to noncollinear antiferromagnets, altermagnets, superconductor-ferromagnet hybrids, and spin-triplet superconductors will reveal novel and fascinating aspects of physics allowing control of magnetic states and spin transport with almost no dissipation. This exploration will encompass topics related to fundamental concepts, such as topological phase transitions and the realization of Majorana bound states.

Recent Progress

Spin superfluidity and the Berezinskii-Kosterlitz-Thouless (BKT) transition – the PI, in collaboration with Edward Schwartz (graduate student) have studied interplay between spin superfluidity and the BKT transition. In this work, we propose to use the Hall response of topological defects, such as merons and antimerons, to spin currents in two-dimensional magnetic insulator with in-plane anisotropy for identification of the magnetic Berezinskii-Kosterlitz-Thouless (BKT) transition in a transistor-like transport geometry (see Figure 1). Our numerical results relying on a combination of Monte Carlo and spin dynamics simulations show transition from spin superfluidity to conventional spin transport, accompanied by the universal jump of the spin stiffness and exponential growth of the transverse vorticity current. Based on our results, we propose a superfluid spin transistor in which the spin

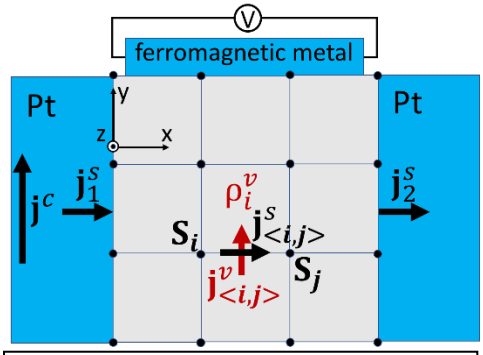

Figure 1: Spin current is injected into an easy-plane magnet using a heavy metal such as Pt. Spin and vorticity currents, and vorticity density are shown for a single plaquette. The vorticity current can be detected by a top ferromagnetic metal contact magnetized along the z axis. The spin current can be modulated by tuning the in-plane magnet below or above the BKT transition via modifications of exchange interactions and anisotropy.

and vorticity currents are modulated by changes in density of free topological defects, e.g., by injection of vorticity or by tuning the in-plane magnet across the BKT transition by changing the exchange interaction, magnetic anisotropy, or temperature. The papers have been published in Physical Review Research and PRB Letters.

Superconductor hybrids, magnetic textures, and realizations of Majorana bound states – the PI, in collaboration with Utkan Gungordu and Hamed Vakili (postdoc) have studied topological superconductivity and realizations of Majorana bound states in superconductor hybrid systems. In our previous works, we have demonstrated how magnetic textures can be used to induce synthetic or fictitious spin–orbit interactions, and, thus, stabilize Majorana bound states. We have described a general approach that works for arbitrary textures and applied it to skyrmions. We have shown how Majorana bound states can be stabilized by elongated skyrmions, certain higher order skyrmions, and chains of skyrmions, and how braiding operations can be performed with Majorana bound states stabilized on magnetic skyrmions. In our latest work, we have extended our approach to d-wave superconductors. We have studied phase-controlled planar Josephson junctions comprising a two-dimensional electron gas with strong spin-orbit coupling and d-wave superconductors, which have an advantage of a high critical temperature (see Figure 2). We have shown that a region between the two superconductors can be tuned into a topological state by the in-plane Zeeman field and can host Majorana bound states. The phase diagram as a function of the Zeeman field, chemical potential, and the phase difference between superconductors exhibits the appearance of Majorana bound states for a wide range of parameters. We have further investigated the behavior of the topological gap and its dependence on the type of d-wave pairing, i.e., d, d+is, or d+id′, and noted the difficulties that can arise due to the presence of gapless excitations in pure d-wave superconductors. On the other hand, the planar Josephson junctions based on superconductors with d+is and d+id′ pairings can potentially lead to realizations of Majorana bound states. Our proposals could be realized in cuprate superconductors, e.g., in a twisted bilayer, combined with the layered semiconductor Bi2O2Se. The papers have been published in Journal of Applied Physics and PRB Letters.

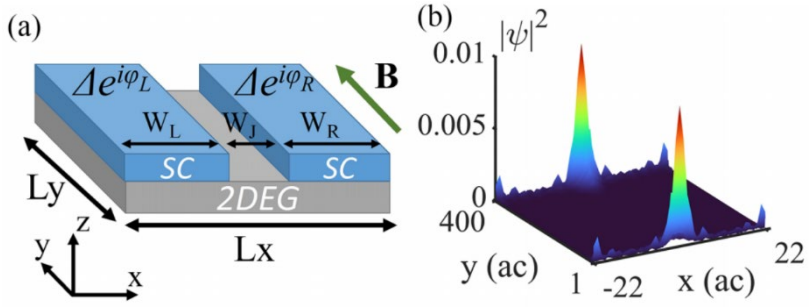

Figure 2: (a) Schematic diagram of the planar Josephson junction heterostructure. The 2DEG is covered by d-wave superconductors, except for a narrow junction between the two superconductors.

(b) The probability function ($|\psi|^2$) of Majorana bound states is plotted.

Drift-diffusion of transverse spin transport in disordered Pt films – the PI, in collaboration with Kirill Belashchenko, Mark van Schilfgaarde, Paul Haney, and Mark Stiles

have studied spin transport in Pt films. In particular, we have studied the interplay between the intrinsic and extrinsic spin transports. To this end, spin-accumulation and spin-current profiles have been calculated for a disordered Pt film subjected to an in-plane electric current within the nonequilibrium Green's function approach. In the bulklike region of the sample, this approach has captured the intrinsic spin Hall effect found in other calculations. Near the surfaces, the results have revealed qualitative differences with the results of the widely used spin-diffusion model, even when the boundary conditions are modified to try to account for them. One difference is that the effective spin-diffusion length for transverse spin transport is significantly different from its longitudinal counterpart and is instead similar to the mean-free path. This feature may be generic for spin currents generated via the intrinsic spin Hall mechanism because of the differences in transport mechanisms compared to longitudinal spin transport. We have found that the orbital accumulation in the Pt film is only significant in the immediate vicinity of the surfaces and has a small component penetrating into the bulk only in the presence of spin-orbit coupling, as a secondary effect induced by the spin accumulation. Our results warrant reinterpretation of experiments involving spin-orbit torques. The paper has been published in Physical Review B.

Future Plans

Conversion of spin currents at interfaces within magnetic heterostructures and mechanisms of spin-orbit torques – the PI will study the conversion of spin currents at magnetic interfaces in the context of spin-orbit torques. The PI aims to investigate model systems with spin-orbit interactions to enhance the understanding of spin-charge conversion and identify new mechanisms of spin-charge conversion [1,2]. Such studies should further resolve some of inconsistencies associated with the orbital mechanism of spin-orbit torque generation. In our previous study, we have also identified inconsistencies in interpretations of spin-orbit torques in systems with dominant intrinsic mechanism of spin current generation, e.g., Pt. To improve our understanding of such systems, some simple models and analytical calculations are warranted. The PI plans to perform analysis of spin-orbit torques using the nonequilibrium Green's function approach obtaining analytical and numerical results. Developing an understanding of this physics through straightforward analytical models can significantly contribute to the design of more efficient memory and computing devices reliant on spin-orbit torques.

Longitudinal and transverse spin Seebeck effect involving both collinear and noncollinear insulating magnets and altermagnets – the PI will study spin currents in collinear and noncollinear insulating magnets and altermagnets and the conversion of spin currents at magnetic interfaces in the context of the longitudinal and transverse spin Seebeck effects. Recent experimental realization of the transverse spin Seebeck effect in LuFeO3/Pt warrants development of new theoretical descriptions of the longitudinal and transverse spin Seebeck effects [3]. To this end, the PI plans to develop transport methods for studying numerically and analytically spin transport in heterostructures containing insulating magnets such as noncollinear antiferromagnets and

altermagnets. Given the prevalence of insulating magnetic materials with above properties, conducting an extensive material analysis in the context of the longitudinal and transverse spin Seebeck effects could potentially unveil a significant pool of other promising candidates.

Spintronics effects in superconductor-magnet hybrid systems – the PI aims to investigate the manipulation of the triplet order parameter in spin-triplet superconductors and the control of the magnetic order parameter in superconductor-magnet hybrid systems [4]. The overarching aim is to expand spintronics principles, e.g., related to spin-orbit torques, into the realm of superconducting systems, potentially yielding innovative solutions for low-temperature, low-dissipation electronics. In this task, the PI plans to employ mean-field methods, analytical and numerical techniques utilizing the scattering matrix approach and accounting for the Andreev reflection, and tight binding simulations using the Kwant package. We will compute the Matsubara Green's function self consistently to subsequently determine observables.

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Excited state electronic structure and phase stability in 2D materials

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Keywords: excitons, Bethe Salpeter Equation, quasiparticle

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Research Scope

The proposal is focused on excited state properties of 2D systems. A recently developed extension[i] of the quasiparticle self-consistent (QS) *GW* method which includes ladder diagrams in the screened Coulomb interaction W is applied to the electronic band structure and optical properties of various 2D and ionic materials. Emphasis is placed on the exciton spectrum, including dark excitons. In the Bethe Salpeter equation (BSE), the imaginary part of the dielectric function $\varepsilon_2(\omega)$ is obtained as a mixture of vertical inter-band transitions, but symmetry can lead to destructive interference effects, resulting in dark excitons. In some cases, these excited exciton states can be related to the Wannier-Mott hydrogenic Rydberg series but modified by the 2D aspects of screening and lower symmetry of the system. Converging the excitonic spectrum is challenging in terms of the dense **k**-mesh required. Visualization of the excitons in real and k-space provides insights in the origin of dark and brightness. In other cases with flat band edges, very large exciton binding energies are obtained.

Recent Progress

The first system considered here is an ultra-wide band gap semiconductor $LiGaO_2$.[ii] The QS$G\widehat{W}$ where the ^ indicates that W is calculated including ladder diagrams predicts a quasiparticle gap of 7.02 eV, somewhat reduced to 6.66 eV by zero-point motion electron phonon coupling effects, but exciton formation leads to a lowest optical gap of about 6.0 eV, in excellent agreement with recent photoluminescence excitation (PLE) spectroscopy. The exciton is split in three sublevels because of the crystal field splitting of the valence band maximum (VBM), also in agreement with experiment. The excitons are shown to be derived from the lowest conduction band (CBM) and the top three different symmetry valence bands but require a fine k-mesh to adequately converge and extrapolate their exciton binding energy. The large exciton binding energy allows us to identify a series of excited state excitons, which can be viewed as a modified Rydberg

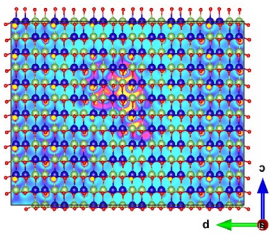

Figure 1: real space exciton function for 2s-type exciton in $LiGaO_2$.

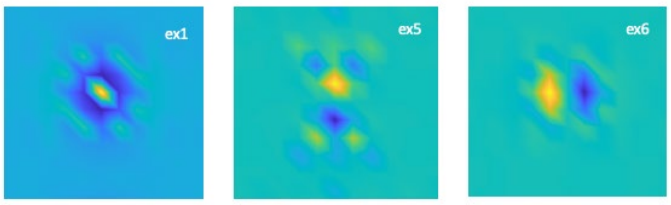

Figure 2: k-space envelope function of some excitons in $LiGaO_2$, showing symmetry related nodal lines, explaining darkness of the exciton.

series. The Fourier transform to real space of the expansion coefficients $A_{vc\mathbf{k}}^{\lambda}$ of a given exciton provides the slowly varying envelope function of the exciton in the Wannier-Mott theory. We can identify either radial nodes in this function of angular nodes consistent with the site symmetry, which explain the darkness or brightness of the excitons. Examples of our real space and k-space analysis are given in Figs. 1-2.

A related study[iii] of the optical properties in $ZnGeN_2$ revealed only the lowest excitons and in this case, still strongly overestimates the binding energy because dynamic screening by the lattice polarization needs to be included. We showed that by carefully extrapolating the exciton energies as function of **k**-mesh and then rescaling the binding energies by the ratio of the static to high-frequency dielectric constants, we could obtain binding energies of the right order of magnitude of a few 10 meV but a precise calculation would require further developments of the dynamic screening effects. On the other hand, we here analyzed the relation of the optical dielectric function in the continuum region in terms of specific interband transitions near critical points and its relation to the parent compound GaN by band folding effects. (Fig. 3)

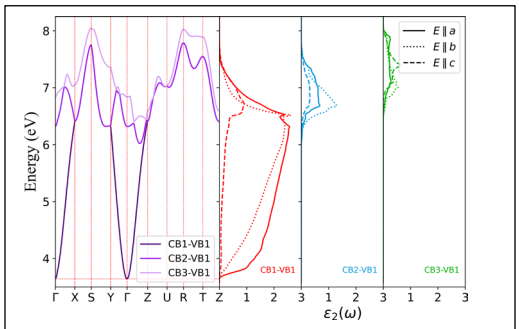

Figure 3: Relating band-to band transitions to features in $\varepsilon_2(\omega)$ in $ZnGeN_2$.

The third system investigated is WSe_2 monolayers.[iv] Here, the first task is to obtain the isolated monolayer limit by studying the behavior as function of interlayer distance and extrapolating the *1/d* slow convergence of the quasiparticle self-energy, which is however compensated by the equally slowly converging exciton binding energy in the optical gap. (Fig. 4) The exciton angular functions in this case are 2D cosine and sine functions if the long-range potential is azimuthally symmetric in the plane. We find the lowest bright exciton (A-exciton) to be in good agreement with experiment and other recently calculations, when a fine enough k-mesh is used and spin-orbit splitting coupling are added a-posteriori. Again, we find some dark and some semi-bright excitons. The semi-bright one can be identified with the A' exciton usually assigned as a 2s exciton. The first dark excitons show a 3-fold nodal pattern indicating a f-like envelope function.

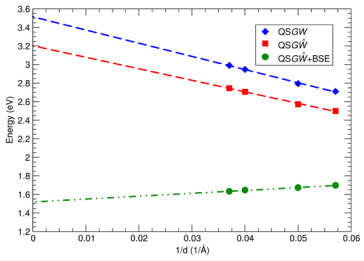

Figure 4: Quasiparticle and optical gaps as function of the inverse of the interlayer spacing.

Finally, we study V_2O_5, which is a layered oxide.[v] In this case, we find extremely high exciton binding energies related to the relatively flat conduction and valence band edges. We also study the behavior of the band structure and excitons and overall optical dielectric function as function of interlayer distance. Upon increasing the distance between the layers, initially, the remaining

weak interlayer hopping is broken and leads to a change in location of the lowest direct and indirect band gaps. Subsequently the quasiparticle self-energy keeps slowly increasing in inverse relation to the distance between the layers but the excitonic binding energy also increases leading again to a much faster convergence of the optical gap. The spatial extent of the exciton wave functions are analyzed and manifest the charge transfer nature of these excitons. In this system we also identified very strong local field effects for the direction perpendicular to the layers. These lead to a strong suppression of the independent particle imaginary part of the dielectric function but were also seen to lead to sharp peaks at high energy. The latter however were found to be very sensitive to the number of conduction bands included in the BSE and become strongly suppressed when transitions to higher, strongly dispersing bands are included.

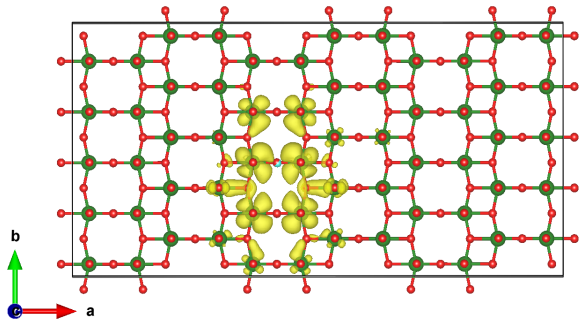

Figure 5 Lowest bright exciton in V_2O_5, probability to find electron when hole at bridge O in the center.

Future Plans

We have initiated a study of band structures in α-As monolayers, which have a puckered form similar to black phosphorus. We already studied its band structure evolution as function of gradually undoing the puckering and transforming to a flat hexagonal honeycomb lattice. We plan to identify the crystalline symmetry related topological band inversions in this system and calculate corresponding optical spectra, including excitons. We have also started extending our study of V_2O_5 to intercalated LiV_2O_5. We find a new band of optical transitions from the now partially filled split-off conduction band to higher bands which is strongly polarized in the a-direction and could lead to re-absorption of luminescence from polaronic states at this energy. This would explain recent cathodoluminescence observations of suppression of the luminescence around 1.8 eV. A paper on this is in preparation. Further studies of this system's antiferromagnetic properties and ordering and in the related γ-structure are planned.

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Magnetization oscillations is quantum spin liquid: emergent spinon and gauge magnetic field.

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Keywords: spin liquid, strong correlation, quantum oscillations.

Research Scope

In metals, electrons in a magnetic field undergo cyclotron motion, leading to oscillations in physical properties called quantum oscillations. This phenomenon has never been seen in a robust insulator because there are no mobile electrons. Working with the experimental group of Lu Li at the University of Michigan, we discovered the first exception to this rule. [1] We study a Mott insulator on a kagome lattice which does not order magnetically down to milli-Kelvin temperatures despite antiferromagnetic interactions. The experimentalists observe a plateau at magnetization equal to 1/9 Bohr magneton per magnetic ion, accompanied by oscillations in the magnetic torque, reminiscent of quantum oscillations in metals. The temperature dependence obeys Fermi distribution. We provide a theoretical interpretation of the data which allows us to fit the rather complex data set with a single equation. [1] The theory is based on a quantum spin liquid state whose excitations are fermionic spinons with a Dirac-like spectrum coupled to an emergent gauge field. We also find a new mechanism for producing the gauge magnetic field which is unique to this kagome system, taking advantage of the Dyaloshinskii-Moriya (DM) interaction. [2] We find a gauge field strength which is about 4 times that of the applied magnetic field which is what is needed to explain the experiment.

Recent Progress

In order to explain the 1/9 plateau with a fermionic particles, there must be a gap or a Dirac node in the fermion band so that the down (up) spin bands holds 5 (4) fermions. This requires 9 bands which can come only from a tripling of the unit cell. This is accomplished either by breaking translation symmetry, or (without doing that) by imposing $2\pi/3$ flux per unit cell. In either case, we sketch the schematic band structure of the 4th to 6th bands in Fig.**1**. Furthermore, a V shaped $\chi=dM/dH$ suggests a linear density of states. In order to explain the V shaped χ, we assume a Dirac node with velocity v_D between band 5 and 6. A small gap at the Dirac node will not change our analysis. We show that this band structure for the spinon can explain the 1/9 plateau and its temperature dependence simply by assuming smearing by a fermi distribution function.

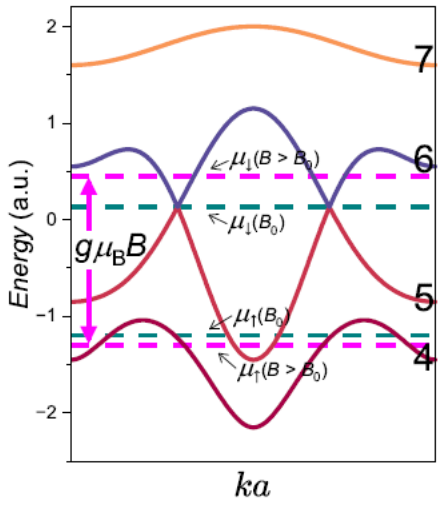

Figure 1, proposed spinon band showing Dirac spectrum between band 5 and 6.

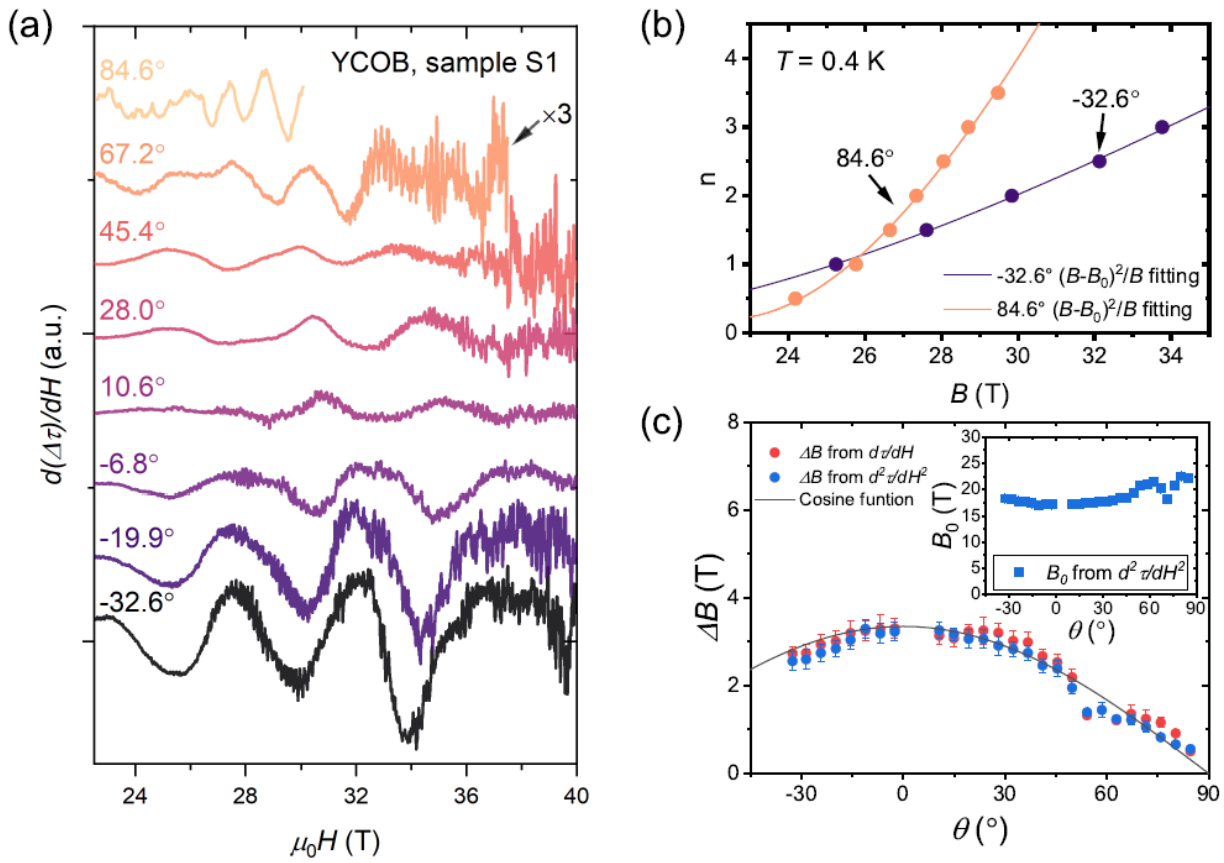

Figure 2. (a) Magnetization oscillations for different angles of the applied B field relative to the c-axis. (b) Fit of the maximum and minimum positions of the oscillations vs B for two angles using equation (1). (c) DB extracted from the bit follows the predicted cos(f) behavior, indicating the orbital origin of the oscillatios. The extracted values of B_0 is shown in the inset.

The above discussion shows that the susceptibility data alone already place considerable constraints on the nature of the fermion bands. We show that the same band structure explains the quantum oscillations. Owing to the large charge gap, the fermions must be charge neutral, leading us to interpret the fermions as fractionalized spinons which are necessarily coupled to an emergent gauge field. Recall that conventional two dimensional (2D) metals exhibit magnetization oscillation due to Landau quantization:

$$M(B) \propto -\sin\left(\frac{A_{FS}\varphi_0}{2\pi B\cos(\theta)} - \phi\right). \qquad (2)$$

where A_{FS} is the Fermi surface area, θ is the angle between the magnetic field B and the c-axis, and $\varphi_0 = h/e$. The application of this equation to our problem requires several modifications. First, for $B > B_0$ a Fermi surface is formed in the down spin band with an area $A_{FS} = \pi k_F^2$ where $k_F=\mu_B(B-B_0)/v_D$. Because it is 2D the gauge magnetic field b is always perpendicular to the plane and produces Landau levels in the spinon spectrum. It is useful to introduce the parameter $\alpha = b/(B\cos(\theta))$ to characterize the relative strength of the gauge field and B. Then we can simply replace B in the denominator of Eq.2 by $|\alpha|B$.

Putting everything together, for $kT \ll \mu_\downarrow$ the oscillatory part of the torque per area is given by

$$M(B) \propto -\sin(2\pi(B-B_0)^2/(B\Delta B) - \phi) \qquad (3)$$

where $\Delta B=C_0\cos(\theta)$.

Figure 2(a) shows the observed magnetization oscillations after background subtraction. For a give angle θ, the oscillations is well fitted by equation 3, as shown in fig. 2(b). Furthermore, the extracted ΔB follows the cos(θ) form predicted, as shown in fig 3c. This show that the oscillations are orbital in origin.

We find that in order to fit the data, a strong gauge magnetic field that is comparable to the applied B field is needed, i.e. $|\alpha|$ is of order unity. This is not expected for a large gap insulator. We propose a new mechanism to generate the gauge magnetic field using the DM term which is known to be present in these kagome structures. [2] Our calculations show that the gauge magnetic field is about 4 times that of the applied field and is of the order needed to explain the data.

Future Plans

We plan to pursue a microscopic version of the phenomenological theory using the method of variational Monte Carlo studies of projected wavefunction. We will try a number of trial wavefunctions for the kagome structure in a strong magnetic field to capture the 1/9 plateau as well as the quantum oscillations.

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Zentropy Theory for Transformative Functionalities of Magnetic and Superconducting Materials

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Project participants: Shun-Li Shang, Nigel Lee En Hew, and Luke Myers

Keywords: density functional theory, statistical mechanics, magnetic materials, ferroelectrics, superconductors

Research Scope

The current research aims to apply the zentropy theory developed by the PI's group to examine the ground-state configurations of complex magnetic, ferroelectric (FE), and superconducting materials and construct their non-ground-state configurations by considering the internal degrees of freedom of their respective ground-state configurations. Their Helmholtz energies are subsequently predicted by DFT-based first-principles calculations, and their statistical probabilities are calculated from statistical mechanics. In the zentropy theory, the partition function of each configuration is evaluated by its Helmholtz energy rather than its total energy, as commonly done in the literature. It was demonstrated in the PI's previous publications that this is the key to enabling accurate predictions of experimental observables in a number of magnetic materials [1], such as singularity at critical points in Ce and Fe_3Pt, negative thermal expansion in Invar Fe_3Pt [2], and strongly correlated physics in $YNiO_3$ [3].

Recent Progress

In the last one and a half years of the project, progresses have been made in ferroelectric $PbTiO_3$, magnetic FeSe, and superconductors of Al, Pb, and $YBa_2Cu_3O_{7-x}$ (YBCO) with both published and unpublished results shown below.

1. $PbTiO_3$

The ground-state configuration of $PbTiO_3$ is of the FE tetragonal structure. The main internal degrees of freedom are the change of polarization directions due to thermal fluctuations, resulting in the formation of local domain walls (DW). The high temperature paraelectric (PE) macroscopical cubic phase detected by X-ray diffractions is due to the fast switching among those fluctuations which could not be detected by X-ray diffractions [4]. Using the two types of DWs for $PbTiO_3$, i.e., 90° and 180° DWs, denoted by 90DW and 180DW, respectively, and their DW energies at 0 K in the literature, the predicted FE-PE transition is in reasonable agreement with the experimentally observed value [5]. We have been calculating the Helmholtz energies of the FE, 90DW, and 180DW configurations using our mixed-space approach [6] to account for the long-range dipole-dipole interactions in ferroelectric $PbTiO_3$ in DFT-based calculations. The predicted

properties of PbTiO3, including derivatives of Helmholtz energy and the FE-PE transition temperature, will be presented at the TCMP PI meeting.

2. FeSe

Fe Se has a complex magnetic ground-state configuration. In the literature, it is generally agreed that DFT-based calculations predict a "staggered dimer" or "pair checkerboard" configuration to be the ground state as shown in Fig. 1 [7]. However, through extensive investigation on supercell shapes and magnetic spin configurations, we find the single stripe configuration with each FeSe layer aligned antiferromagnetically to be the ground state. This configuration lowers the total energy by 40meV/atom. We are performing energy-volume and phonon calculations to validate the calculation results and will report them at the TCMP PI meeting.

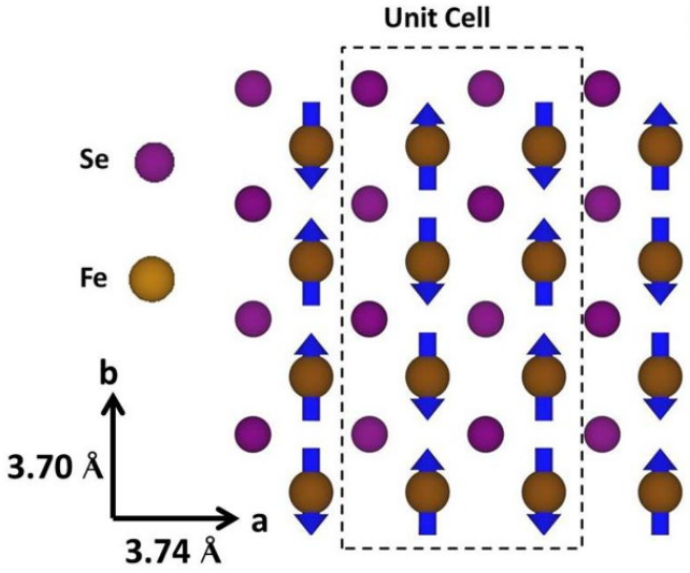

Fig. 1: Schematic top view of the pair-checkerboard AFM in an FeSe layer. Each spin of the Fe atom has only one neighbor spin aligned ferromagnetically while the other neighbor spins are all aligned antiferromagnetically. The rectangle enclosed by the dashed lines denotes the magnetic unit cell. From Cao et. al [7].

3. Superconductors

Due to the weak pairing interaction ($\sim 10^{-3} eV$) of Cooper pairs, thermal energy can easily break them, resulting in conventional superconductors only at low temperatures in terms of the Bardeen–Cooper–Schrieffer (BCS) theory. Based on the zentropy theory, we [1] postulated that in high T_C superconductors, the layered structures in complex phases reduce the entropy due to phonons, thus preserving the Cooper pairs within the layers to higher temperature, and T_C is thus determined by the competition between the superconducting and non-superconducting configurations. The key challenge is to define the superconducting ground-state configurations in the framework of DFT.

Even though DFT considers valence electrons to be independent, it is based on rigorous multi-body interactions, and the interactions are represented by an average effective potential collectively determined by the nuclei and all electrons. Consequently, the electron density should accurately reflect the multi-body interactions even for superconducting-state configurations at 0 K. It is important to emphasize that the results from DFT-based calculations at 0 K for ground-

state configuration only are not comparable with experimental observations due to the statistical mixture of many configurations above zero temperature.

Through detailed DFT calculations and analyses of electronic structures of many configurations, we have discovered the superconducting-state configurations in Al, Pb, and YBCO and are in the process of assembling them into a manuscript for publication. We anticipate that the paper will be sent out for publication before the TCMP PI Meeting.

Future Plans

We plan to finish three manuscripts based on the results mentioned above and submit them for publication before the TCMP PI Meeting.

We plan to perform the following research tasks in the next year.

- Study the magnetic transitions in Fe-Ni Invar alloys;
- Study the magnetic and superconducting transitions in FeSe;
- Expand the calculations to other conventional and high T_C superconductors.

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A new approach to the interacting phonon problem

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Keywords: phonons; phonon interactions; lattice dynamics

Research Scope

The major goals of this project are two-fold. The first goal is to be able to compute all group theoretically allowed phonon interactions in arbitrary crystals within some range and up to some order in a well defined, efficient, and reproducible manner; and to communicate our irreducible phonon interactions to the broader community for further use. The foundation for accomplishing this first goal is provided by our lone and bundled irreducible derivative approaches (Ref. 1). The resulting irreducible phonons and phonon interactions can then be used to compute a wide variety of classical or quantum observables, including thermal expansion, temperature dependent elastic constants, thermal conductivity, structural phase transitions, and any other vibrational observable. Particular emphasis will be placed on extracting minimal models of anharmonicity which can be used to truly build understanding of complex phenomena.

The second goal is to develop and implement a hierarchy of approaches to solve the interacting phonon Hamiltonians which we compute using our irreducible derivative methods. Standard approaches include molecular dynamics, which provides the numerically exact classical solution, and the Hartree-Fock approximation for phonons, which provides a baseline quantum solution. More advanced techniques include self-consistent perturbation theory, which sums additional families of diagrams beyond Hartree-Fock, in addition the centroid molecular dynamics, which allows the computation of accurate real frequency in the quantum regime. This hierarchy of approaches allows one to rapidly obtain a baseline solution, which is critical for big data approaches, or spend more computational resources to get a very robust solution.

Recent Progress

During the past reporting cycle, numerous tasks were accomplished. First, we published our self-consistent diagrammatic perturbation theory approach (publication 5), where the Hartree-Fock or quasiparticle Green's function is used to evaluate various families of diagrams, including the bubble and sunset diagrams. These self-consistent perturbation theory results were rigorously tested in the classical limit by comparing to irreducible derivative molecular dynamics, which delivers the numerically exact classical result (see figure 1a and 1b). Second, we succeeded in implementing unfolding of irreducible phonon interactions (publication 3). In a previous funding cycle, we implemented phonon unfolding within our irreducible derivative approaches (publication 7), though phonon unfolding has been achieved in the literature long ago; but it is still not ubiquitous. However, we are not aware of any previous work on unfolding phonon interactions.

This technical achievement allowed us to use DFT+U to compute and unfold the cubic and quartic phonon interactions in the antiferromagnetic Mott insulating phase of UO_2, and directly compare the linewidths with inelastic neutron scattering experiments by accounting for the q-voxel (publication 6). We used our self-consistent perturbation theory to provide highest level of diagrammatic perturbation theory currently available in the literature. We demonstrated good agreement with the experimental linewidth, in addition to the thermal conductivity. UO_2 is a classic material, and our technical developments have finally allowed for a definitive understanding of the vibrational behavior in this system at intermediate and high temperatures.

The third accomplishment was an application of our irreducible derivative methodology to assess the quality of neural network (NN) based empirical potentials (publication 2). Our irreducible derivative methods provide the "ground truth" for derivatives in crystals, and high order derivatives provide a critical test of NN based potentials which continue to explode in popularity. Our analysis demonstrated that NN potentials performed surprisingly well, yielding very reasonable results for quartic derivatives, and only seriously deteriorating for sixth derivatives.

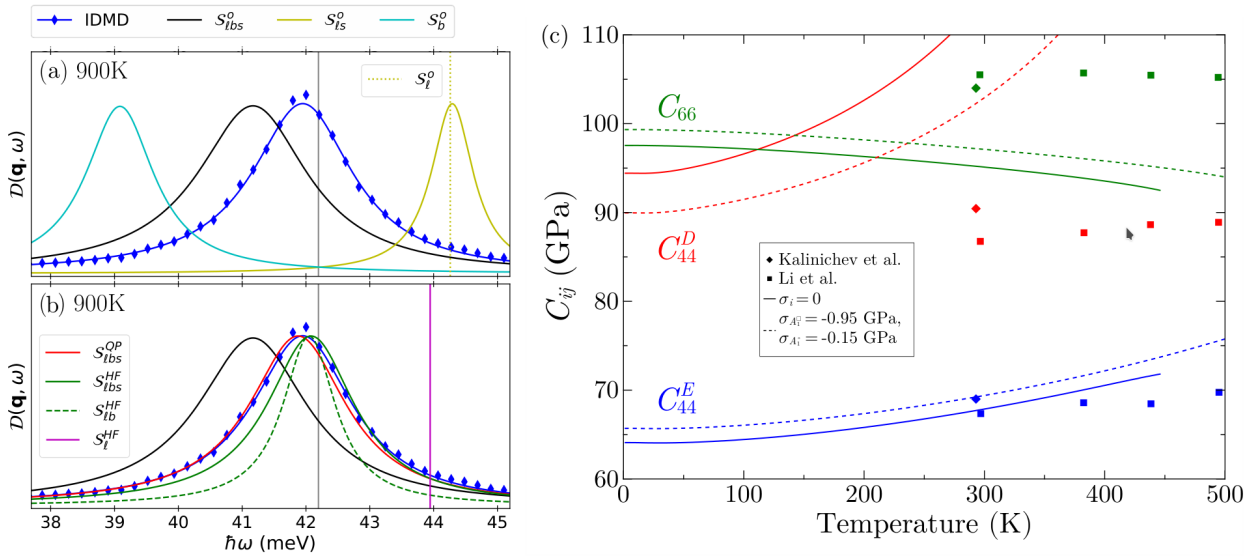

Figure 1: (panels a and b) The classical spectral energy density of CaF_2 at T=900 K at the Γ point in an energy window around the T_{2g} phonon frequency (gray vertical line). The blue points are the numerically exact results from irreducible derivative molecular dynamics, while the lines are obtained from various levels of self-consistent perturbation theory in the classical limit (see publication 5 for further details). (panel c) Various elastic constants as a function of temperature and stress in ferroelectric $PbTiO_3$ computed using the generalized quasiharmonic approximation (lines) and compared to experiment (points) (see publication 1 for further details).

The fourth accomplishment was particularly important, providing the elastic properties at finite temperature and stress in ferroelectric materials (publication 2). Properties such as the piezoelectric coefficient are of critical fundamental and technological importance, yet there are no practical methods to compute such quantities at finite temperature and stress including quantum

effects. We achieved a number of technical developments which allowed us to apply our generalized quasiharmonic approximation to ferroelectric $PbTiO_3$, producing the elastic constants and piezoelectric coefficient as a function of temperature and stress for the first time (see figure 1c for elastic constants). While these results are an important technical achievement, comparison with experiment suggests that the quasiharmonic approximation is insufficient to capture the physics, and therefore we will need to go one step further and implement a strain dependent version of our Hartree-Fock approximation for phonons. The strain dependent formalism that we developed is completely general, allowing for Hartree-Fock to be straightforwardly applied, though at a much greater computational expense.

The final accomplishment of the previous reporting cycle was in the context of software development. We have continued to push forward our software suite, Principia Materia. We implemented the ability to handle non-symmorphic space groups, which we previously had been handling using the corresponding lower symmetry symmorphic subgroup. Another important thrust was beginning yet another major rewrite of the code from the ground up, using an object relation mapper to greatly streamline how we handle complex data structures that inherently must be dealt with in our code suite. We officially set our first public release date as September 1st 2024.

Future Plans

Several major tasks are planned for the next funding period. First, we will execute a detailed study of the phonon anharmonicity of the canonical quantum paraelectric $SrTiO_3$. While there are anharmonic Hamiltonians in the literature from decades ago, our preliminary studies indicate that they have massive discrepancies. Additionally, it is unclear if neural network based potentials, which are now being used on this system, are sufficient to capture the delicate anharmonic terms associated with the soft modes in this system. We will provide the ground truth for the anharmonic Hamiltonian of the Gamma and R points, and provide a minimal description of the overall vibrational Hamiltonian. Once the vibrational Hamiltonian is established, we will use our hierarchy of approaches to solve the Hamiltonian, including Hartree-Fock and self-consistent perturbation theory. We also plan to use this system as a launching point for our centroid molecular dynamics and quantum Monte-Carlo, in order to deliver highly robust quantum solutions which can benchmark our diagrammatic approaches.

An important technical development that we promised is an enhanced Fourier interpolation routine, and we will address this in the coming funding period. Specifically, we will develop and approach for merging phonon interactions using different finite translation groups, potentially exploiting the long wavelength behavior which can be probed with anharmonic strain derivatives. Another important technical point we will study is a detailed comparison between our irreducible derivative approaches, which are based on finite difference, with density functional perturbation theory. Our preliminary results suggest that our approaches may converge much more rapidly with respect to k-points and plane wave cutoff in charge density wave systems. Proving this will be an important milestone for our methods.

Finally, we plan finish our most recent rewrite of our code and issue a beta release of this branch. We continue to add more external users to our code, and we believe that we will be sufficiently close to a finished product in the next funding cycle for our first public beta release on September 1st 2024. As part of this code release, we are computing the phonons in several complex systems, including the representative cuprates and hybrid perovskites.

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Bloch Oscillations in Two Dimensional Artificial Crystals

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Keywords: Transport phenomena, Collective dynamics, Superlattices, Topological Materials, Bloch wave theory

Research Scope

Moiré superlattices and other two-dimensional artificial materials that are patterned into large-period structures are promising platforms for observing new nonlinear electric current responses in crystals in the high-field regime. Anticipated since early work on semiconductor heterostructures in the 1970's, a one dimensional superlattice fractures a Bloch band into minibands in which a propagating wave packet can be Bragg reflected, generating an oscillatory current in response to a static electric field: a Bloch oscillation [1]. Moiré materials lift this phenomenology into two dimensions with an important twist. In two dimensions the oscillatory current from the zone-periodic variation of the group velocity generally coexists with a nonclassical current response perpendicular to the driving field due to the anomalous velocity in a Bloch band [2]. Remarkably, in large period two dimensional superlattices one often encounters situations where the bands are very narrow and spectrally isolated, but a substantial Berry curvature persists. This combination acts to suppress the conventional group velocity mechanism for longitudinal Bloch oscillations but it admits the fully quantum-geometric transverse response [3]. As a result one can directly see nonclassical contributions to the current transport in the high field regime through the dependence of the oscillating current on field strength and direction [4]. This turns out to provide a sensitive measure of the distribution of Berry curvature in momentum space, even in situations where symmetry requires the band-integrated Berry curvature to vanish.

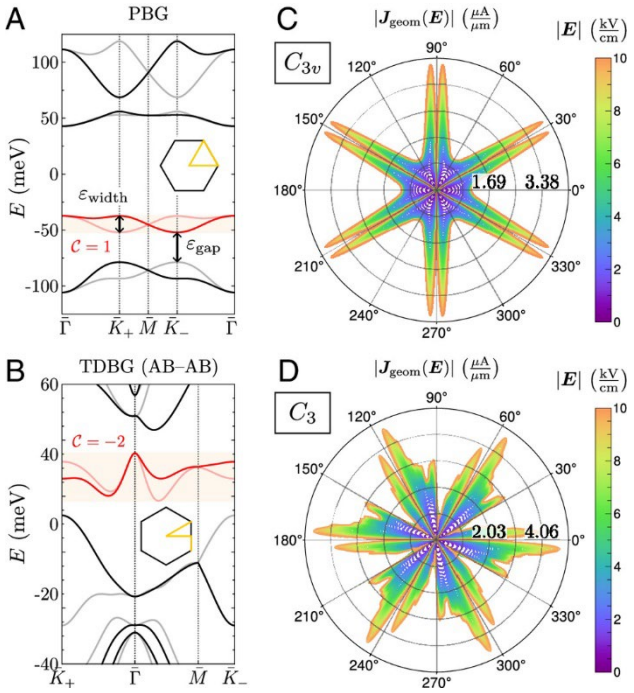
Recent Progress

Field Scaling of Bloch Oscillations [3]

We find that the Berry curvature can induce an oscillating trajectory of a band-projected electron wave packet transverse to an applied static electric field. Though analogous to conventional Bloch oscillations, this novel oscillatory behavior is driven entirely by the quantum geometry in momentum space instead of by the group velocity in a dispersive band. In conventional Bloch oscillations the oscillating current can be suppressed by increasing the field strength, giving a signature negative differential conductance in the high field regime. By contrast, the current from the geometric contribution saturates to a *nonzero plateau* in the strong-field limit. Furthermore we find that in nonmagnetic materials, the geometric oscillations are even under inversion of the applied field, whereas the Bloch oscillations are odd, a property that can be used to further distinguish these effects when they coexist. We estimate that for a typical relaxation time of 1 ps [56], the onset of geometric oscillations requires $E \approx 0.5$ kV/cm, which is both experimentally feasible and below the threshold for Zener tunneling.

Rose patterns in the High Field Response [4]

In the high-field regime, the nonlinear current response of a two-dimensional crystal features a strong angular dependence, which encodes information about both the energy band dispersion and the Berry curvature of its isolated electronic Bloch minibands. In this regime, when an electron is accelerated by the field, it explores nearly the entirety of the folded Brillouin zone before a scattering event can relax its momentum. Hence, in real space the electron becomes sensitive to features of the crystal potential on the lattice scale which produces a strongly anisotropic response as a function of the direction of the electric field. In particular, one finds that the dependence of the current on the field direction is given by "flowers" (rose patterns) whose petal structure is symmetry constrained and whose explicit analytical form we obtain from an expansion of the band energy and Berry curvature in real-space translation vectors. To illustrate the transport theory, we performed calculations for artificial crystals with time-reversal and trigonal symmetry, which are relevant for many moiré and other two-dimensional superlattices. Specifically, we considered periodically-buckled graphene and twisted double bilayer graphene, and conclude that these effects should be accessible at experimentally-relevant field strengths. (Figure 1)



The right panel gives the angular anisotropy of the current from Bloch oscillation as a function of field direction in a two dimensional superlattice. The color scale shows the effect of increasing the field strength. The calculations are carried out for a single layer of periodically buckled graphene (top) and for an AB-BA double graphene bilayer (adapted from Ref. 4)

Berry Curvature Spectroscopy [5]

Artificial crystals such as moiré superlattices can have a real-space periodicity greatly exceeding the underlying atomic scale. This facilitates Bloch oscillations in the presence of a static electric field. We investigated the optical response of such a system when the bands states are dressed with a static electric field. One finds that the optical response of the dressed system becomes resonant at the Bloch oscillation frequency, typically in the terahertz regime when the lattice constant is of the order of 10 nm. In particular, we studied the dressed optical conductivity within the semiclassical band-projected theory. The most important new result is that the magnitude of resonances in the dressed optical Hall conductivity are proportional to the lattice Fourier components of the Berry curvature and independent of the momentum-relaxation time. This is a direct consequence of the presence of geometric Bloch oscillations [3]. Consequently, we find that

coupling light to geometric oscillations allows for a spectroscopic probe of the momentum distribution of the Berry curvature.

Future Plans

Zener tunneling between Bloch minibands

We propose to generalize these interesting results to take into account the possibility of interband coupling in the form of Zener tunneling between different Bloch bands. To address this, we are developing a full quantum theory of the static drive. This is achieved by first treating the static electric field in temporal gauge such that a uniform electric field enters the Bloch Hamiltonian with a vector potential through the usual minimal substitution. Interestingly, the resulting time-dependent description becomes a Floquet problem for fields that lie along a reciprocal lattice vector where time periodicity of the Bloch Hamiltonian is inherited from the periodicity in momentum space. In this treatment, the physically time-independent problem in D spatial dimensions turns into a time-dependent problem in D-1 spatial dimension, since the longitudinal momentum becomes a gauge degree of freedom that is absorbed in the time origin. The corresponding Floquet quasienergies then yield the familiar Wannier-Stark ladder with interband coupling included to all orders. By further coupling the clean system to a bath using the Floquet Green's function formalism, we can calculate band-resolved currents and investigate the effect of interband mixing on the current response and in particular on the transverse nonlinear Hall response in the regime of geometric Bloch oscillations. Our initial results show that the band-projected theory remains valid within a wide range of parameters as long as either time-reversal or spatial inversion symmetry is present which suppresses the lowest-order interband corrections. When both symmetries are broken, the latter contribute to the transverse current response, an effect known as the quantum metric Hall response. Importantly, while the predicted plateau in the transverse response due to the geometric oscillations does not persist indefinitely with increasing field strength, a characteristic peak in the corresponding transverse differential conductance can still be observed.

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A universal model of Floquet operator Krylov space

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Project Participant: Hsiu-Chung Yeh

Keywords: Non-equilibrium systems, Floquet systems, topological phases, Krylov techniques.

Research Scope

The study of driven quantum systems with a focus on understanding their topological properties.

Recent Progress

It is shown [1] that the stroboscopic time-evolution under a Floquet unitary, in any spatial dimension, and of any Hermitian operator, can be mapped to an operator Krylov space which is identical to that generated by the edge operator of the non-interacting Floquet transverse-field Ising model (TFIM) in one-spatial dimension, and with inhomogeneous Ising and transverse field couplings. The latter has four topological phases reflected by the absence (topologically trivial) or presence (topologically non-trivial) of edge modes at 0 and/or pi quasi-energies. It is shown that the Floquet dynamics share certain universal features characterized by how the Krylov parameters vary in the topological phase diagram of the Floquet TFIM with homogeneous couplings. These results are highlighted through examples, all chosen for numerical convenience to be in one spatial dimension: non-integrable Floquet spin 1/2 chains and Floquet Z3 clock model where the latter hosts period-tripled edge modes.

Future Plans

We plan to exploit the mapping found by us between any Floquet system in any dimension to the one-dimensional inhomogenous transverse field Ising model. We also plan to study two dimensional Floquet systems, exploring flat bands and the possibility of anomalous Landau levels, the latter arising due to the periodicity of the Floquet spectrum.

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Coulomb-enhanced quantum critical electrical conductivity

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Keywords: quantum phase transitions, effective field theory, duality, transport, disorder

Research Scope

The primary objective of the research is to advance our understanding of anomalous metallic states of strongly interacting electron systems [1] using effective field theory.

Recent Progress

There were five published papers over the past two years (and there are one or two preprints likely soon to appear). In the highlighted paper (listed last in the publication list), we find that an unscreened Coulomb interaction enhances the zero-temperature dc electrical conductivity at superconductor-insulator transitions in the dirty (2+1)d XY universality class. This result advances our understanding of disordered quantum criticality using a paradigmatic quantum critical point, which may be relevant to various quantum materials and that has only recently been understood theoretically. In our study, we used a (dual) model consisting of a single Dirac fermion at zero density, coupled to a Chern-Simons gauge field and in the presence of a quenched random mass, with or without an unscreened Coulomb interaction. To control our calculation, we used the $1/N_f$ expansion, where N_f is the number of Dirac fermions. We found that $\mathcal{O}(1/N_f)$ gauge field and disorder fluctuation corrections do not preserve "self-duality.

Future Plans

One thrust may consider the interplay of quenched disorder and interactions in low-temperature anomalous metals. A future project may use the strong disorder fixed point of composite fermion mean-field theory to study the effects of an unscreened Coulomb interaction, with the goal of finding a strong disorder fixed point with finite Coulomb interaction. A related project might consider interaction-driven insulators, such as the Hall insulator, within the composite fermion framework.

A second thrust may consider more general theoretical approaches to systems with a Fermi surface. One project might study the effects of (irrelevant) electron-electron interactions and phonons on the (in)famous linear in temperature scaling of the electrical resistivity of "strange metals" [2], with the goal of understanding the robustness of this linear in T phenomenon. Another project might consider the implications of the loop group symmetry of the Fermi liquid [3] for non-Fermi liquid metals, building on our prior DOE-supported work.

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Fractons and beyond

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Keywords: Fractons, lattice, contextuality, entanglement, quantum advantage

Research Scope

The project is concerned with exploring fracton phases (and related topologically ordered phases). Questions of interest include exploring the dynamics and thermodynamics of fracton and related phases, identifying means of characterizing such phases, identifying routes to laboratory realization, and better understanding the role of the lattice.

Recent Progress

I will present two key recent results. (1) Much of the physics of fractons can be understood as arising from multipolar symmetries. Multipolar symmetries have previously only been studied in one dimension, or else on square/cubic lattices. We have shown how multipole symmetries may be treated on arbitrary crystalline lattices. Given as inputs a space group and a set of occupied Wyckoff positions, we can enumerate all possible consistent multipole groups, and explore the consequences thereof. Some novel phenomena are identified on the triangular and breathing Kagome lattices.

(2) We introduce a new way to characterize and exploit such exotic phases on quantum hardware, based on nonlocal quantum games. Previously known quantum games require fine tuned resources. We are exploring how topological (and fracton) ordered states can be used to provide robust quantum advantage at a range of device oriented tasks.

We have demonstrated how a quantum state in the same topologically ordered phase as the `toric code' may be used to gain robust quantum advantage at a non-local coordination task. We have explicitly demonstrated this quantum advantage on the *Quantinuum* quantum computer, and have demonstrated how topological order endows this advantage with greater robustness than available in nontopological states. We have also demonstrated how one may tune through a topological transition – witnessed by the loss of quantum advantage – using a finite depth circuit (with measurements and feedback) on currently available quantum hardware.

Future Plans

The work (1) provides a powerful framework for exploring fractons on arbitrary lattices. In future work we propose to explore gauging multipolar symmetries on arbitrary lattices, and thereby to discover new fracton phases. Meanwhile, the work (2) described above constitutes `proof of principle' that topological order can afford robust quantum advantage at a device oriented task. In future work we plan to elucidate (a) how different topological (and fracton) orders can be characterized using this `quantum games' framework (b) how topological and fracton orders may

be harnessed to gain advantage at the task of computing arbitrary nonlinear Boolean functions and (c) how the protocols we introduced above may be improved to enhance the amount of quantum advantage throughout the respective phases.

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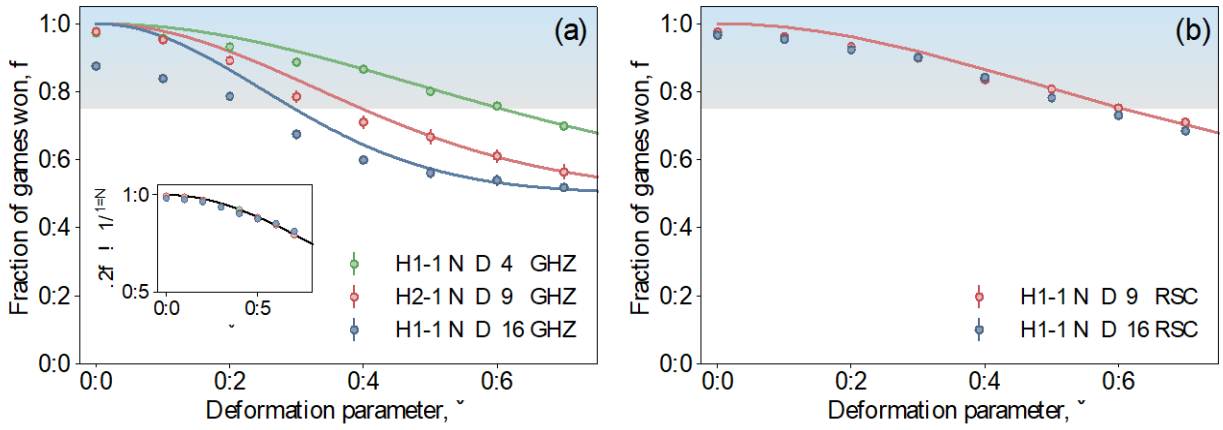

Figure 1: The `parity game' provides a crisp example of a device oriented task at which quantum resources can afford practical advantage. The shaded area represents a region of performance that cannot be accessed via any purely classical strategy. The plots above show our performance at this task on the *Quantinuum* quantum device using two kinds of quantum resource states (a) a (deformed) GHZ state (not topologically ordered), and (b) a (deformed) toric code state (topologically ordered). The points represent data (obtained by averaging 256 shots on the device). The solid lines represent the theory prediction. Both states provide quantum advantage when available in fine tuned form (no deformation). However, with a GHZ resource state the advantage is rapidly lost as the state is deformed, and is lost more rapidly as the `size' of the state increases – the finite size collapse in the inset indicates that there is no quantum advantage in the thermodynamic limit at any non-zero deformation strength. In contrast, for the toric code there is no loss of performance as the state is increased in size, and robust quantum advantage persists to the thermodynamic limit, as long as we remain in the topologically ordered phase.

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Controlling reversible phase transitions in rare-earth nickelates for novel memory devices

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Collaborators: Dillon Fong, Subramanian Sankaranarayanan (Argonne National Laboratory)

Keywords: Metal-insulator transitions, Complex oxides, Resistive Switching, Atomistic Simulations, Machine Learning

Research Scope

Resistive switching in strongly correlated oxides is lucrative for emerging applications in neuromorphic computing, and densely scaled non-volatile memory. Electrical conductance of such complex oxides can be controllable switched across multiple orders of magnitude by either (a) electroforming a conduction channel (e.g., in tungsten oxide), or (b) inducing Mott-Hubbard transition (e.g., in rare-earth nickelates)– both via controlled migration of defects (such as oxygen vacancies) under applied bias. Nevertheless, the promise of such defect-driven electronic transitions are far from realized due to a lack of fundamental understanding of the atomic-scale processes that underlie migration and spatiotemporal evolution of oxygen vacancies over nano-to-mesoscopic length/timescales under applied electric field. In this project, we employ a synergistic integration of density functional theory (DFT) calculations, *ab initio*/classical molecular dynamics (AIMD/CMD) simulations, machine learning (ML), precision synthesis, and multi-modal X-ray imaging experiments to address this knowledge gap. Such an integrated approach offers to elucidate the correlations between subtle structural distortion and oxidation states; treat localized charge carriers; describe defect/ion transport in the presence of electric field; and, in turn, greatly advance the current understanding of microstructural evolution in complex oxides under applied bias. The fundamental knowledge gained from this work will enable precise control over hierarchical defect structures and unravel new routes to manipulate resistance states in complex oxides. This, in turn, will accelerate design of novel devices with desired set of neural functionalities, and high-speed densely-scaled resistive random access memory technologies.

Recent Progress

1. Controlled formation of conduction channels in memristive devices to improve reliability: Accuracy of the state-of-the-art memristors is not sufficient to realize practical neuromorphic computers. This lack of accuracy stems from the intrinsic randomness in the electroforming process required to form conduction channel (region containing high concentration of oxygen vacancies) in the oxide memristor. Specifically, the electroforming process depends on the local concentration of defect concentration, which results in large variation in performance across devices. Even for a given device, the location, size, and shape of the conduction channel can change dramatically from one switching cycle to another. Using planar device as model system, we demonstrate that conduction channel can be formed at predictable and controllable locations by introducing sharp protrusion in electrode gap, as observed by multi-modal X-ray imaging (Figure 1a-d). CMD simulations show that such controlled formation of conduction channels occurs due to localized electric field near the notches in the electrode (Figure 1e-j). For a flat electrode (i.e., no notches) (Figure 1(a)), the vacancy distributions obtained under forward bias

after the first and second switching cycles are markedly different from each other, indicating stochastic formation of conduction channel (Figure 1e-g). In contrast, the confinement of the electric field provided by triangular protrusions in the electrode yields similar vacancy distributions after forward-reverse-forward bias cycles (Figure 1 h-j). This allows the controlled formation of a conduction channel. Importantly, the reproducibility of the vacancy distribution during switching is not affected by the initial distribution of vacancies. These simulations demonstrate that electric field confinement reduces variability in the performance of devices, as well as the cycle-to-cycle variance for a given device.

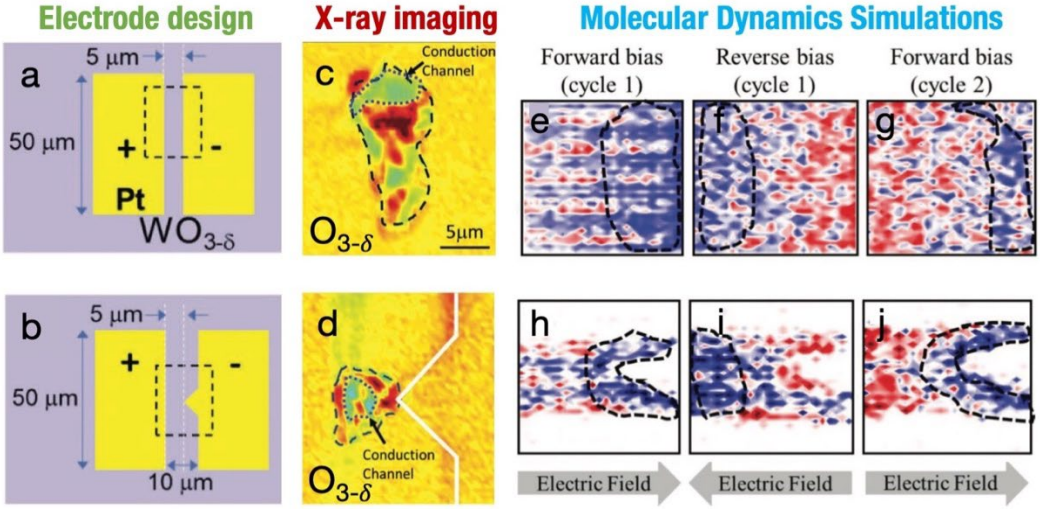

Figure 1. Controlled formation of conduction channel in oxide memristor by careful engineering of sharp protrusions in electrode gap. Schematic illustration of electrode design with (a) flat surface and (b) triangular protrusion. Multi-modal X-ray imaging of conduction channel for electrode configuration (a) without, and (c) with triangular protrusion. Spatial distribution of oxygen vacancies obtained from representative CMD trajectories (averaged over the last 50 ps) of atomic models for devices with flat electrode (e-g), and that with triangular protrusion (h-j). In panels (e-h), regions of high oxygen vacancy concentration are shown in blue, while oxygen vacancy deficient regions are shown in red. The black dotted lines in panels (c-j) indicate the conduction channel.

2. Machine learning interatomic potentials for understanding dynamics of electron-lattice coupling in rare-earth nickelates: We introduce a deep learning neural network (DNN)[1] interatomic potential to accurately describe atomic interactions in $SmNiO_3$, a representative rare earth nickelate using a large dataset obtained from AIMD simulations within the framework of DFT + *U*. This newly developed DNN predicts atomic forces and cohesive energies within 0.50 meV/Å/atom and 0.1 meV/atom of DFT+*U* values, respectively. Additionally, to capture the chemical bonding and local electronic configuration of Ni, we develop two additional DNNs to predict (a) atomic Bader charges and (b) number and location of Maximally Localized Wannier Functions (MLWFs) around Ni and O atoms in any given structure. These DNN models predict atomic charges within ~0.01*e* of DFT+*U*, and can reproduce the correct DFT+*U* *d*-electron configuration with ~90% accuracy. Using these DNN models, we performed long-time CMD simulations to understand the atomic-scale dynamical phenomena that control thermally induced metal-insulator transition (MIT) in $SmNiO_3$ (Figure 2) We found that MIT is driven by a series of cooperative lattice breathing modes (including rotation, tilting and distortion of NiO_6 octahedra) that causes concomitant Ni-O bond and Ni-charge disproportionation with decrease in temperature (Figure 2a,b). This, in turn, facilitates a transition from a metallic state with a predominant d^7 configuration to an insulating state featuring two sets of Ni atoms with inequivalent electronic

configuration (i.e., d^6 and d^8) at low temperature (Figure 2c-e). Our DNN-CMD simulations also showed that these dynamic processes can be suppressed (and consequently decrease critical temperature for MIT) by increasing the inter-octahedral tilt angle (Ni-O-Ni), such as via applied pressure, substrate-induced epitaxial strains, or chemical pressure induced by rare-earth cations.

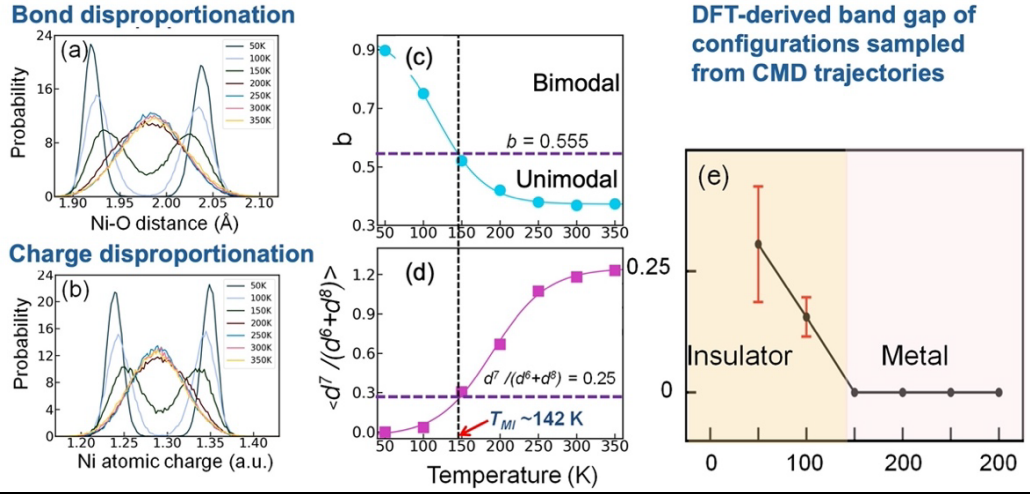

Figure 2. Dynamical processes underlying temperature induced MIT in $SmNiO_3$ from DNN based molecular dynamics simulations. Distribution of (a) average Ni-O bond lengths within a NiO_6 octahedron, and (b) charge on Ni atoms as a function of temperature. (c) Bimodality of the distribution of Ni-O bond lengths or Ni-charge, and (b) relative fraction of the population of Ni with various *d*-configurations (derived using our DNN-MLWF model), as a function of temperature averaged over the last 1 ns of classical MD trajectories based on our DNN-FF interatomic potential. (e) Electronic band gap of 10 representative structures at each temperature sampled from classical MD trajectory calculated at the DFT + *U* level of theory

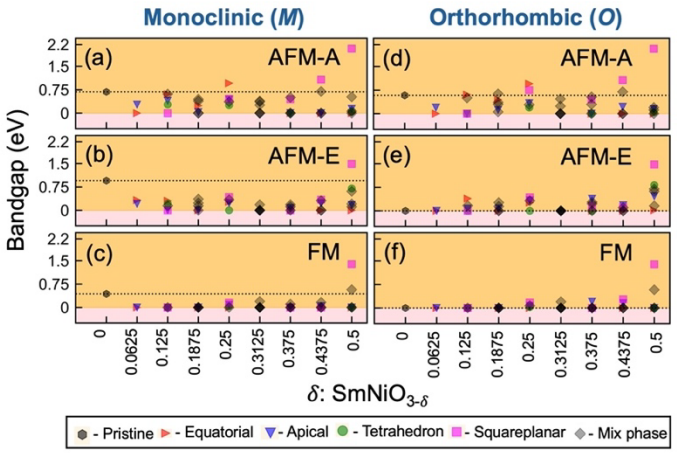

Figure 3. Oxygen-vacancy induced band-gap opening in SNO. The band gaps for all possible configurations of NiOx motifs at different levels of oxygen deficiency are shown for (a-c) *M*- and (d-f) *O*-phases with different magnetic ordering.

3. Stability, electronic behavior, and migration of oxygen vacancies in rare-earth nickelates: SNO exhibits a perovskite structure, and can exist in orthorhombic (*O*) and monoclinic (*M*) forms. Controlled introduction of OVs can result in a rich variety of vacancy-ordered structures (depending on OV amount) composed of various NiO_x motifs, including (a) octahedral NiO_6, (b) pyramidal (NiO_5) featuring either a missing O in the square-base (equatorial pyramid) or apex (apical pyramid) of the NiO_6 octahedron, (c) square planar (NiO_4), and (d) distorted tetrahedron (NiO_4). Our DFT +*U* calculations on all possible OV configurations (i.e., all possible NiO_x combinations for oxygen deficiency δ = 0-0.5) show that irrespective of δ, $SmNiO_{3-\delta}$ shows an energetic preference for *M* phase, although the *M* and *O* phases are energetically close (~10–50 meV/u.f depending on magnetic ordering). Similarly, for both *M* and *O* phases, ferromagnetic ordering (FM) is the most stable. Notably, the square planar motifs play a significant role in stabilizing the structure (at least ~0.2 eV/u.f.). Nevertheless,

at any given concentration, several possible distributions of NiO_x motifs lie within ~100 meV/u.f. of each other; which makes them all synthesizable using non-equilibrium methods (e.g., pulsed-laser deposition). Interestingly, the *O* phase exhibits MIT upon introduction of OVs, with a rich diversity of gaps afforded by different combinations of NiO_x motifs and magnetic ordering (Figure 3). In all these cases, band gap opening occurs due to a combination of (a) on-site Coulombic repulsion due to electron filling of e_g state of Ni in NiO_6 octahedra (yielding a correlated state), and (b) crystal field splitting introduced by the symmetry of NiO_x motifs. For instance, pristine *M*-SNO with FM ordering is a narrow band-gap insulator (~0.3 eV) featuring two sets of Ni atoms with inequivalent electronic structure (i.e., high-spin $3d^8$ Ni^{2+} and low-spin $3d^6$ Ni^{4+}). In vacancy-ordered $SmNiO_{2.5}$ with alternating NiO_6 octahedra and NiO_4 square planar motifs, electron-filling occurs in e_g state of NiO_6 (yielding high-spin Ni^{2+} ($t_{2g}^6 e_g^2$); ~1.3μ_B) , while the t_{2g} and e_g states in NiO_4 split (resulting in a low spin Ni^{2+} ($t_{2g}^4 d_{z^2}^2 d_{xy}^2 d_{x^2-y^2}^0$); ~0.15$\mu_B$). The valence band maximum is controlled by hybridization of $3d_{z^2}$ of octahedral Ni and O-$2p_z$, while conduction band minimum is dictated by crystal field splitting in NiO_4, and Ni-$3d_{x^2-y^2}$-O-$2p_x$,$2p_y$. The extent of the Ni-O hybridization, on-site repulsion, and crystal field splitting can be controlled by rare-earth ion (with small rare earth yielding highest band gap opening). We also found that the kinetic barrier associated with OV migration is strongly controlled by the distribution of NiO_x motifs (e.g., barriers for OV migration in $SmNiO_{2.6875}$ range from 0.9 – 1.7 eV for different OV distributions), owing to various tilt patterns induced by the NiO_x motifs.

Future Plans

We are currently working on (1) extending the DNN interatomic potential to capture structure, energetics, and dynamics of oxygen vacancies in rare-earth nickelates, (2) understand evolution of defects under applied electric field, and formation of hierarchical extended defects, (3) identify earth-abundant oxides susceptible to metal-insulator transition via electron doping, and (4) understand variation in oxidation states of Ni/Co in CoO_2 and NiO_2 with Li-intercalation, in collaboration with RXES experiments by collaborators at Argonne and ORNL.

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Publications

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1. H. Liu, Y. Dong, M. Galib, Z. Cai, L. Stan, L. Zhang, A. Suwardi, J. Wu, J. Cao, C. Tan, S. Sankaranarayanan, B. Narayanan†, H. Zhou†, D. Fong†. *Controlled Formation of Conduction Channels in Memristive Devices Observed by X-ray Multimodal Imaging.* Advanced Materials **34**, 2203209 (2022) DOI**:** https://doi.org/10.1002/adma.202203209 (Highlighted by Nature Review Materials)

Manuscripts either submitted or close to submission

2. M. Galib, R. Pidathala, B. Narayanan. *Machine learning interatomic potential to understand dynamics of metal-insulator transition in strongly correlated perovskite nickelates.* Manuscript submitted
3. R. Pidathala, B. Narayanan. *Thermodynamic stability, electronic properties, and migration of oxygen vacancies in oxygen-deficient samarium nickelate*. Manuscript close to submission
4. D. Bhagat, R. Pidathala, B. Narayanan. *Effect of rare-earth cation on electronic properties of ordered-vacancies in rare-earth nickelates*. Manuscript close to submission

Phonon dynamics in Kitaev materials
Natalia B. Perkins, University of Minnesota

Keywords: Quantum spin liquid, sound attenuation

Research Scope

The scope of our research encompasses the investigation of the coupling between collective spin and lattice degrees of freedom, which plays a crucial role in determining the fundamental properties of correlated materials. Our work has specifically focused on elucidating the effects of spin-phonon coupling in Kitaev magnets and revealing observable signatures of fractionalized excitations in the phonon dynamics of these materials [1-5]. In a recent study, we examined the potential of sound attenuation as a probe to study fractionalized excitations in alpha-$RuCl_3$, a promising Kitaev spin liquid candidate [5].

Recent Progress

Recent progress in our research comprises two key components. Firstly, we performed a purely theoretical exploration of the phonon dynamics in generalized Kitaev models describing quantum spin liquid in the proximity to the exactly solvable model but in the presence of additional interactions characteristic for the real materials [1-5]. Secondly, in collaboration with experimentalists, we performed the ultrasound investigations to probe fractionalized excitations in alpha-$RuCl_3$ [6]. Our main results are summarized in Figure 1.

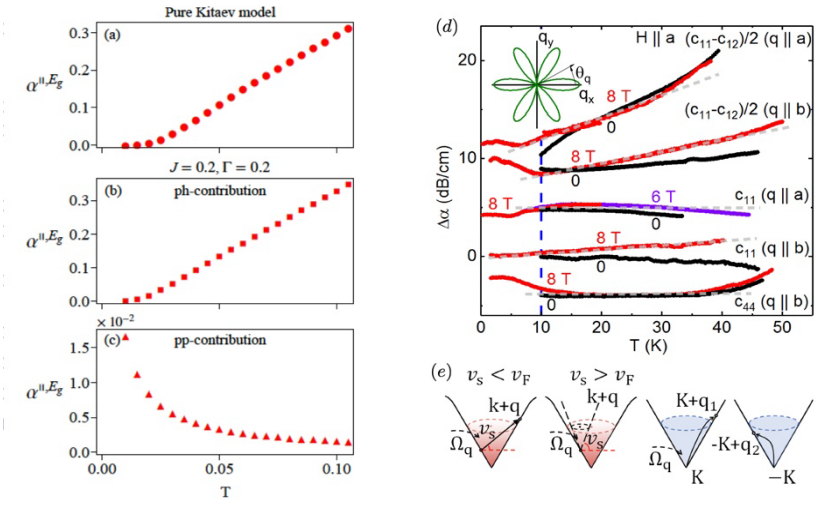

Figure 1: The temperature evolution of the sound attenuation coefficient is shown for (a) the pure Kitaev model, (b) the extended Kitaev model from particle-hole processes, and (c) the extended Kitaev model from particle-particle processes. Panel (d) displays the change in sound attenuation versus temperature for selected acoustic modes under zero and finite magnetic field applied along the **a**-direction. Panel (e) illustrates various scattering channels near the bottom of the Dirac cone, distinguishing between acoustic phonon velocities less than and greater than the Fermi velocity.

The theoretical study comprised several steps. First, we developed a theoretical framework to study the effects of phonon dynamics in the pure Kitaev model and showed that the fractionalized spin excitations of the Kitaev spin liquid—specifically, the itinerant Majorana fermions and static $Z2$ fluxes— have distinct effects on the phonon dynamics. This suggests the potential use of phonon dynamics as a valuable tool for exploring materials that could host the Kitaev spin liquid phase. Then we

computed the sound attenuation due to a phonon scattering off a pair of Majorana fermions, revealing a linear temperature dependence ($\sim T$) [1] (see Fig.1 (a)). We argued that the attenuation coefficient has a particular angular dependence, and its magnitude is modified by the thermal excitation of the $Z2$ fluxes, reflecting the effects of the emergent disorder on the Majorana fermions introduced by the $Z2$ fluxes [2].

Second, considering that candidate materials for the Kitaev spin liquid typically exhibit residual interactions beyond the Kitaev coupling, we investigated how fractionalization signatures are influenced by these additional interactions. To do so, we conducted a mean-field analysis of sound attenuation in the generalized J-K-Γ model [4]. Our results indicate that if the system remains in the spin liquid phase, distinctive characteristics of sound attenuation persist, even in the presence of residual interactions (see Fig.1 (b) and (c)).

Lastly, we tackled the challenge of characterizing the Kitaev spin liquid in the presence of defects, such as lattice distortions, stacking faults, dislocations, and impurities. We recognized that disorder can dramatically alter the low-energy excitations in the Kitaev model, impacting both dynamical and thermodynamic properties, including phonon dynamics. To gain deeper insight into this impact, we calculated the sound attenuation coefficient in the diluted Kitaev honeycomb model [5].

The experimental effort in studying phonon dynamics was focused on studying sound attenuation in alpha-$RuCl_3$ [6]. Our experimental collaborators obtained the temperature and in-plane magnetic-field dependence of the sound velocity and sound attenuation for several longitudinal and transverse phonon modes propagating along high-symmetry crystallographic directions (see Fig.1 (d)). Our comprehensive data analysis above the ordered state provided strong evidence of phonon scattering by Majorana fermions. As predicted by our prior theoretical investigations [1,2], this scattering depends sensitively on the value of the phonon velocities relative to the characteristic velocity of the low-energy fermionic excitations (see Fig.1 (e)) describing the spin dynamics of the underlying Kitaev magnet. Remarkably, in alpha-$RuCl_3$, the phonon dynamics exhibit two distinct attenuation regimes due to the significant difference in velocities between the longitudinal and transverse phonon modes relative to dispersing Majorana fermions. In one regime, a phonon scatters an occupied fermion state to a higher-energy state (particle-hole channel), leading to linear temperature-dependent sound attenuation. In the other, a phonon decays into two fermions, both with positive energy (particle-particle channel), resulting in almost temperature-independent attenuation, with only a slight decrease observed for temperatures above 30-40 K. Additionally, we conducted a detailed investigation of the field-temperature phase diagram of alpha-RuCl3 for an in-plane magnetic field applied perpendicular to the Ru-Ru bonds, leveraging ultrasound's high sensitivity to various phase transitions.

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Future Plans

We plan to expand our study of phonon dynamics to moire systems, such as twisted bilayer graphene (TBG) and transition metal dichalcogenide (TMD) bilayers. These systems offer novel pathway to explore correlated-electron and topological effects in highly tunable narrow bands, as well as provide a groundbreaking approach to quantum two-dimensional magnetism. For TBG, several studies suggest that the electron-phonon interaction significantly influences the phase diagram, either in conjunction with strong electron-electron correlations or by potentially driving instabilities independently, such as superconductivity. Similar effects from magnetoelastic coupling are anticipated in TMD systems.

Motivated by the success of the use of sound attenuation as an effective probe in Kitaev materials, we plan to study sound attenuation in these systems. For example, we anticipate that TBG may exhibit distinctive features in sound attenuation related to its electronic structure and phonon dynamics. In TMD bilayers, the interplay between spin, valley, and lattice degrees of freedom can lead to rich physics and potentially unique sound attenuation characteristics.

Our plan also involves studying the elastic collective modes of a moire superlattice using Raman scattering. These phonon modes arise not from vibrations of a rigid crystal but from the relative displacement between the constituent layers. Despite their similarity to acoustic phonons, these modes, called phasons, are not protected by any conservation law. Their spectrum can be derived from the original linear dispersions of graphene's acoustic phonons, which are reconstructed into superlattice minibands. The structure of these minibands and their dependence on the twisting angle can be explored through phonon Raman scattering.

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Co-functional (anti)ferroic perovskites with topological phases: statics and dynamics at finite temperatures

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Keywords: Rashba effects, ferroics, spin textures, finite-temperature simulations, Rashba ferroelectricity co-functionality

Research Scope

The project focus is on the development, implementation, and application of computational methodologies to study Rashba effects, Rashba ferroelectricity co-functionality in ferroics at finite temperatures. We report (i) discovery of giant spin splitting in a family of hybrid organic-inorganic perovskites and its fundamental origin; (ii) development of finite-temperature computational methodology to study newly emergent ferroelectric semiconductors among halide perovskites, and their spin-related properties; (iii) and the possibility of enhancement of spin splitting through nanostructuring of halide perovskites.

Recent Progress

Prediction of giant spin splitting in $MHyPbX_3$ (X=Br, Cl). Most recently we computationally discovered a compound with giant spin-splitting. Spin splitting is defined as removal of double spin degeneracy in the energy level of an isolated system or an energy band of a solid. It often originates from the relativistic effect of spin-orbit interaction. The spin-orbit coupling contributes a weak perturbation to the Hamiltonian of the system, which is proportional to $(\nabla V \times p) \cdot \sigma$. Consequently, the effect originates from the presence of (local) electric field, which could be due to the lack of inversion symmetry in bulk material, interfacial asymmetry in heterostructures, and even local asymmetry in centrosymmetric compounds. The effect leads to spin-momentum locking and intriguing physical manifestations for many emerging spin-orbitronic applications, including intrinsic spin-Hall effect, gate-controlled spin precession, (inverse) spin galvanic effects, photogalvanic effects, and chiroptic effects, that rely on manipulation of the spin degrees of freedom by electrical, optical, or magnetic means. Naturally, these intriguing effects and their emergent and potential applications rely on strong spin-momentum coupling and large spin splitting, which in turn originate from large spin-orbit coupling term. For example, for room temperature applications it is desirable to have a material that exhibits spin splitting in excess of 30 meV. Unfortunately, being weak relativistic effect spin-orbit coupling typically is not strong enough, and so far only a handful of materials exhibited experimentally measurable or technologically significant values. One example, is a polar semiconductor BiTeI which possesses a high Rashba spin-splitting of 100 meV [1]. Recently, hybrid organic-inorganic perovskites with chemical formula ABX_3 came into the focus of attention. Here A is the organic, while X could be organic ligand or inorganic element. We carried out first-principles DFT calculations on some representative of $MHyPbX_3$ (X=Br, Cl) hybrid organic-inorganic perovskites family in order (i) to predict the giant spin splitting of up to 400 meV; (ii) reveal its fundamental origin; (iii) report the

unique spin textures in these materials, which are not present in other materials with giant spin splitting.

Figure 1 shows our computational data. Panel (a) visualizes the structure of the material, panel (b) gives the band structure computed with spin-orbit coupling turned on. It reveals spin splitting in the vicinity of both CBM and VBM. Figure 1(c) shows the actual spin splitting values obtained along different directions of the Brillouin zone. Our data predict that the giant value of up to 400 meV can occur in these materials. This is the largest value reported in the literature, to the best of our knowledge. To investigate the temperature evolution of this giant value and in to eliminate the possibility that the giant spin splitting is an artifact of simulations, we computed Rashba spin splitting for the structures obtained from different structural relaxation techniques, including ab initio Molecular Dynamics and present data in Figure 1(d). The data reveals that the values remain large at finite temperatures as well.

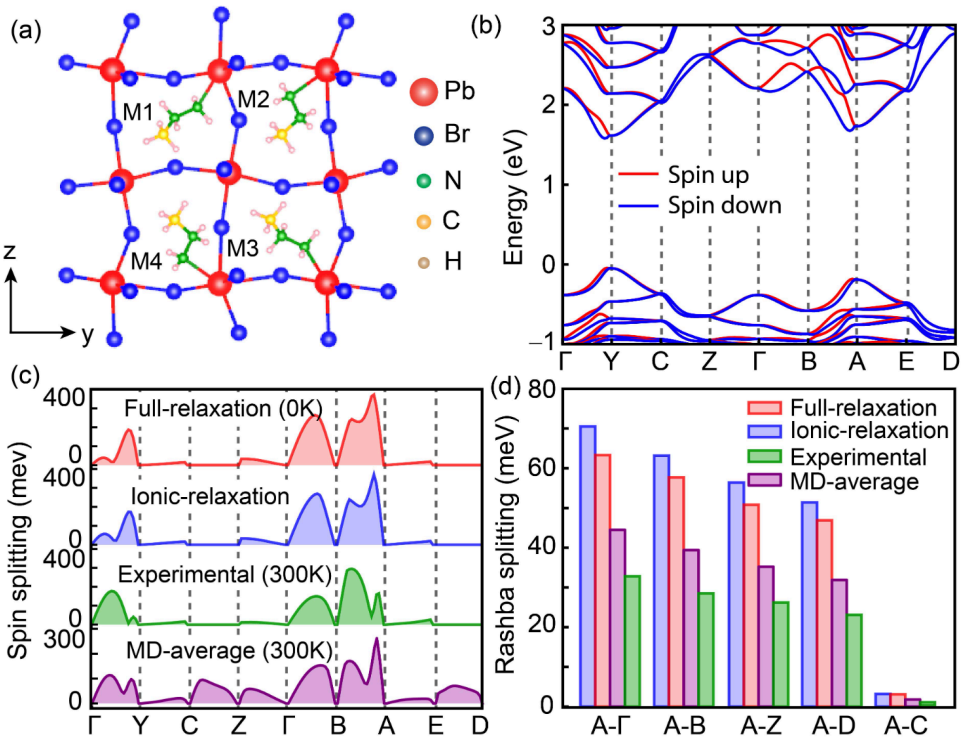

Figure 1: (a) Structural visualization of $MHyPbX_3$; (b) band structure of $MHyPbX_3$ computed with spin-orbit coupling turned on; (c) spin splitting along different directions in the Brillouin zone; (d) Rashba spin splitting obtained from different structural relaxation techniques as indicated in the legend.

Finite-temperature methodological development. Recently, ferroelectricity has been demonstrated in the family of halide perovskites: $CsGeX_3$ (X=Cl, Br, I). We developed a first-principles-based computational approach to probe finite-temperature properties of this family. Figure 2 shows the temperature evolution of polarization and lattice parameter and demonstrates that the material undergoes a single phase transition. Panels (c) and (d) shows the hysteresis loops and the dependence of the coercive field on the temperature, respectively, demonstrating excellent agreement with experimental measurements from the literature. We have used these finite-temperature data to compute dependence of structure and spin splitting on the temperature and across phase transition.

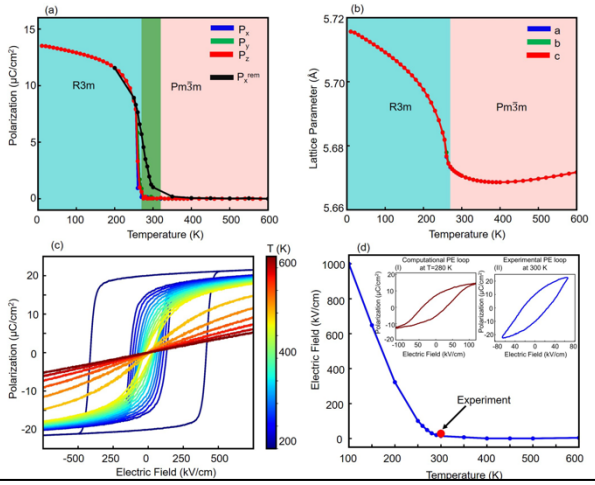

Figure 2: Temperature evolution of polarization (a) and lattice parameter (b) in $CsGeBr_3$; (c) simulated hysteresis loops at different temperatures; (d) comparison of our computational data with experimental ones from the literature [2].

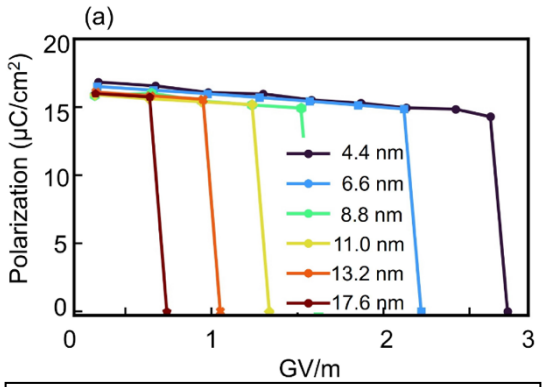

Figure 3: Dependence of polarization on the residual depolarizing field in $CsGeBr_3$ thin films with thicknesses indicated in the legend.

Enhancement of spin splitting in $CsGeBr_3$ through nanostructuring. We have applied our finite-temperature computational methodology to study $CsGeBr_3$ ultra thin films. Surprisingly, we found that such films are capable of sustaining residual depolarizing field of up to 3 GV/m (see Fig.3). Such a field could greatly enhance Rashba spin splitting in $CsGeBr_3$ nanostructures. Our next step is to compute electronic structure of these films to demonstrate the effect.

Future Plans

We have already prescreened numerous hybrid organic-inorganic compounds for their electronic structure properties, Rashba ferroelectricity co-functionality, and spin-splitting. Our next step is to develop efficient computational methodology for the most promising ones in order to study temperature evolution of these properties as well as their behavior across phase transitions. Same electronic structurer properties are being investigated in ferroics with topological polar structures.

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Real-frequency Bethe-Salpeter equation and Landau damping in Fermi liquids.

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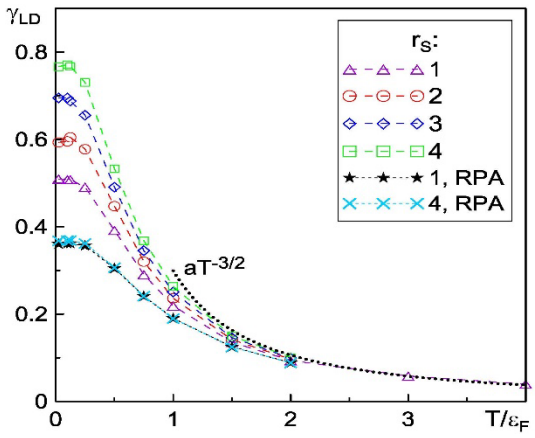
Keywords: Polarization, Landau damping, Bethe-Salpeter equation

Research Scope

Self-consistent Hartree-Fock approximation combined with solutions of Bethe-Salpeter equation is a powerful scheme for studies of strong correlation effects in materials. The standard finite-T approach would be to first solve the problem in the Matsubara representation and apply numerical analytic continuation to the real-frequency axis to link theoretical results with experimental probes. We demonstrate that the ladder-type finite-temperature Bethe-Salpeter equation in the Hartree-Fock basis for the 3-point vertex function and, ultimately, systems's polarization can be accurately solved directly on the real frequency axis using the diagrammatic Monte Carlo technique and series resummation. We illustrate the method by applying it to the homogeneous electron gas model [1] and demonstrate how multiple scattering events renormalize Landau damping [2]. We establish how this damping mechanism depends on the Coulomb parameter r_s and changes with temperature between the correlated liquid and thermal gas regimes. In a broader context of dielectric response in metals, we also present the typical dependence of the exchange-correlation kernel on frequency at finite momentum and temperature within the same computational framework.

Recent Progress

We found an efficient way of implementing algorithmic Matsubara integration approach for ladder diagrams and used it to study dynamic response at arbitrary temperature with extremely high accuracy.



Landau damping coefficient for different values of r_s as a function of temperature

Future Plans

Current scheme is based on expansion in terms of the static screened Coulomb potential. Next step would be to perform an expansion in terms of the dynamic screened interaction.

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First Principles Approach to Exciton Transport in Energy Materials

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Keywords: exciton dynamics, many-body perturbation theory, 2d materials, theoretical spectroscopy, topological materials

Research Scope

This project aims to develop and apply methods in *ab initio* many-body perturbation theory (MBPT) to understand exciton dynamics in materials of interest for optoelectronics and energy harvesting and transport. The proposed work is broadly organized under three objectives that address exciton dynamics in the ballistic, diffusive, and intermediate regimes. The first objective is to understand how features of crystal symmetry and dimensionality manifest in exciton dynamics through band-like exciton transport. The second objective is to understand the role of exciton-phonon interactions in exciton dynamics by developing and performing predictive calculations of exciton-phonon relaxation and decoherence times. The third objective is to model exciton diffusion processes by parametrizing a Boltzmann-like equation for exciton transport from *ab initio* calculations. The materials we will focus on include 1) monolayer transition metal dichalcogenides (TMDs), which exhibit exceptionally strong light-matter coupling and spin-momentum locking, 2) layered metal organic hybrid halide perovskites, which are promising materials for photovoltaics and broadband light emitting diodes (LEDs), and 3) topological insulators (TIs), where chiral excitons forming from transitions between high energy surface states hold the potential for dissipationless energy transport. The methods we employ build on the *ab initio* GW plus Bethe Salpeter equation (GW-BSE) approach.

Recent Progress

One of the major focus areas of our research in the past two years has been on the study of high-energy resonant excitons and their dynamics. Excitons arising from surface states in topological insulators have attracted recent interest for both their potential to couple topologically protected spin currents with the helicity of light and to couple bulk and surface states. These excitons are typically resonant states with electrons or holes in valleys or bands at higher energies

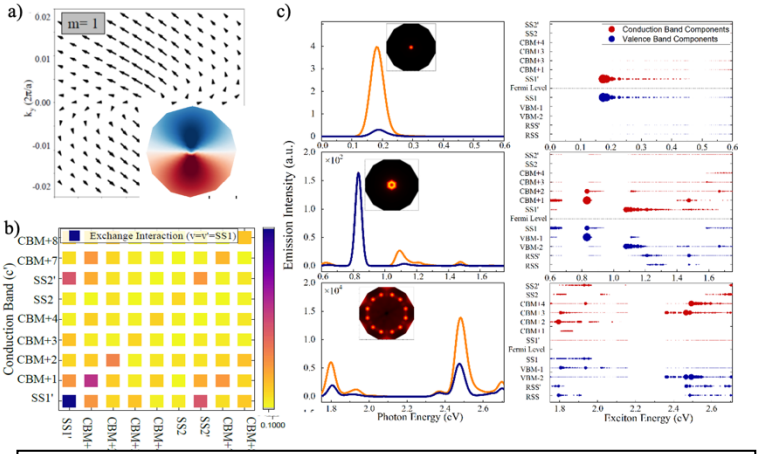

Fig. 1: a) Phase winding of the envelope of the lowest energy bright exciton in Bi_2Se_3 shows p-like symmetry. b) BSE exchange matrix elements show coupling of bulk electron states and topological surface states (SS1, SS2). c) Instantaneous emission of left (blue) or right (orange) circularly polarized light after excitation with right circularly polarized light. Exciton composition is shown on the right. Figure adapted from Ref. [1].

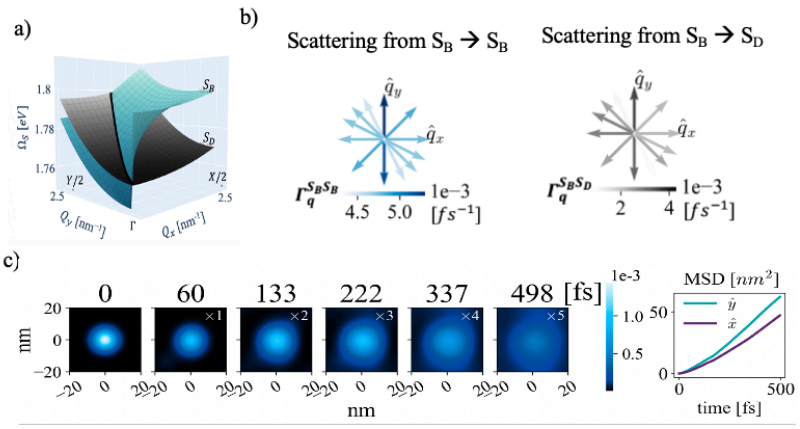

Fig. 2: a) Dispersion of lowest energy bright (S_B) and dark (S_D) singlet exciton in crystal pentacene. b) Asymmetry of phonon scattering from the bright state at zero momentum to finite momentum bright states (left) and finite momentum dark states (right). c) Time evolution of an exciton wavepacket in real-space with exciton-phonon interactions included. Figure adapted from Ref. [4].

than the linearly dispersing topological surface states (TSS) near the Fermi level. Moreover, TSS in the prototypical topological insulator Bi_2Se_3 are frequently characterized using optical probes around 1.5-2 eV, but electron-hole interactions and their effect on surface localization and optical response of the TSS remain largely unexplored. We used *ab initio* calculations to understand excitonic effects in the bulk and surface of Bi_2Se_3 [1]. We identified multiple series of chiral excitons that exhibit both bulk and TSS character, due to exchange-driven mixing of electron-hole pairs residing on the surface and bulk (Fig. 1). Our results address fundamental questions about the degree to which electron-hole interactions can relax the topological protection of surface states and dipole selection rules for circularly polarized light in TIs by elucidating the complex intermixture of bulk and surface states excited in optical measurements and their coupling to light.

Another type of resonant exciton we have been exploring is a high-energy exciton (HX) in monolayer TMDs, coming from transitions between the valence band maximum at the K point and the third highest conduction band, which has a negative effective mass [2]. Previous experimental measurements have shown that the HX has a nonradiative lifetime comparable to that of the lowest energy bright exciton state [2]. We calculate the exciton-phonon relaxation time of the HX state in monolayer MoS_2 and WS_2, which requires including exciton-phonon matrix elements for up to ~3000 exciton bands. We find that the resonant state coming from the negative mass conduction band has a lifetime that is ~3 times longer than other excitons in the same energy range, though the calculated lifetime of ~10 fs is still considerably shorter than the experimentally measured lifetime.

The other major thrust of our work focuses on the relation between the exciton bandstructure, exciton-phonon interactions and exciton transport in real space. Non-radiative exciton relaxation processes are critical for energy transduction efficiencies in optoelectronic materials, but how these processes are connected to the underlying crystal structure and its associated electron, exciton, and phonon band structures is poorly understood. We developed a first-principles approach to explore exciton relaxation pathways in pentacene, a paradigmatic

molecular crystal and optoelectronic semiconductor [3]. We compute the momentum- and band-resolved exciton-phonon interactions and use them to analyze key scattering channels. We find that exciton intraband transitions on femtosecond timescales leading to dark-state occupation is a dominant nonradiative relaxation channel in pentacene. We further show how the nature of real-time propagation of the exciton wavepacket is connected with the longitudinal-transverse exciton splitting [4], stemming from crystal anisotropy, and concomitant anisotropic exciton and phonon dispersions (Fig. 2). Our results provide a framework for understanding time-resolved exciton propagation and energy transfer in molecular crystals and beyond.

Future Plans

Over the course of the next year, we will expand on our model of exciton-phonon propagation in real space by studying exciton transport in various 2D materials. In particular, we will characterize and compare carrier, exciton, and heat transport in bulk, monolayer, and few-layer black phosphorus (BP), where highly anisotropic exciton transport has been observed. In addition to exciton-phonon scattering, we will build terms into our kinetic equation that account for exciton-exciton interactions that depend on the exciton density. This work will lay the foundation for the first-principles study of exciton diffusion.

Recent time-resolved angle-resolved photoemission spectroscopy (TR-ARPES) experiments revealed long-lived indirect excitons in the three-dimensional topogological insulator Bi_2Te_3. These indirect excitons are believed to be composed of holes in the bulk and electrons at the bulk-surface mixing point, where the topological surface state merges with bulk conduction states. As a follow-up to our work on Bi2Se3, we will characterize the indirect exciton in Bi2Te3. We will try to understand exciton binding energy, the degree of spatial localization at the surface, and the reason for its long experimentally-observed lifetime.

Finally, our planned future work and the first principles techniques we have developed are very computationally intensive requiring dense sampling of the Brillouin zone and a large number of bands to accurately capture energy and momentum-conserving scattering processes for states far from the band edge. In the next stage of our project, we will develop machine learning (ML) models that will allow us to accurately extrapolate real-time dynamics and perform down-stream prediction of many-body calculations from lower-cost density functional theory calculations (DFT). In these efforts, we will focus on generative unsupervised learning approaches, moving away from supervised feature selection that was the focus of previous work.

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Excitons, phonons, and excited state charge dynamics in novel 2D materials

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Keywords: Novel 2D materials, excitons, Coulomb interaction, phonons, ultrafast dynamics

Research Scope

The overarching goal of this project (DE-FG02-07ER46354) is to understand, explain, and predict striking equilibrium and non-equilibrium properties of low dimensional materials, particularly their response to ultrafast probes. While ground state electronic structure and the excitation spectra reveal information on novel steady-state properties of these materials, deeper insights into the impact of electron correlations, phonon scattering and multi-excitation processes on the excited state dynamics are obtained in the non-equilibrium regime, in response to ultrafast probes. For the purpose, we have developed a density matrix-based time dependent density functional theory (DM-TDDFT) code with which, in addition to absorption and emission spectra, we calculate the characteristics of quasiparticles (excitons, trions, biexcitons) and collective excitations. To unveil the role of electron correlation, we have developed techniques combining dynamical mean field theory (DMFT) and TDDFT through which has allowed rationalization of experimental data such as the femtosecond demagnetization of Ni and ultrafast charge dynamics in the Mott insulator VO_2. One focus in two dimensional (2D) materials stems from the pronounced manifestations of their bound excitations (excitons, trions, biexcitons) whose reduced Coulomb screening and interesting ultrafast dynamics can be tracked by the above computationally feasible codes. Spatially resolved picosecond (ps) dynamics of the excited states and emission spectra of single layer (1L) and bilayer (2L) transition metal dichalcogenides (TMDs), studied also experimentally through pump-probe measurements, are our main targets. Our momentum-resolved calculations track the dynamics of the bright and dark excitons in these materials, as well as the relative contributions of the intra and interlayer excitons (electron and hole in the same or different layers, respectively,) to the emissive properties. Through coupling of excitons, electrons, and holes to phonons we follow the charge dynamics at experimental timescales (fs to ps) both for rationalization of data and prediction of material properties. Investigation of other interacting degrees of freedom – electrons, holes, spins, and excitons – for selected hetero-bilayer TMDs, twisted and untwisted, is also planned. These effects play an important role in nonlinear dynamics and emission. A related question is how the properties above depend on the twist angle, and how dramatic are correlation effects at small angle.

Recent Progress

1. DM-TDDFT approach for *intervalley* excitons and dark excitons in 1L WSe_2

We have formulated an approach based on TDDFT to describe characteristics of both intra- and inter-valley excitons in semiconductors (Fig. 1)[1]. The inter-valley case was a challenge since it required the derivation of the Wannier equation for non-vertical transitions. This TDDFT Wannier equation for exciton polarization is quite general since it describes both intra- ($\mathbf{k} = \mathbf{q}$) and inter-valley ($\mathbf{k} \neq \mathbf{q}$) transitions. On testing the above formulation for several types of exchange-correlation kernels to calculate binding energies and energetics of intra-valley bright and intra and intervalley dark excitons in 1L and 2L WSe_2, we find the nanoquanta exchange-correlation (XC) kernel to be the most suitable. For 1L WSe_2, these ab initio calculations (a first to our knowledge) find the binding energy of the intravalley bright exciton (91 meV) and the energy difference of 22 meV between the intravalley (132 meV) and intervalley (110 meV) dark excitons to be in reasonable agreement with experimental results.[2] Furthermore, we trace the energy difference between the intra- and inter-valley dark excitons to the exchange contribution to the XC energies of these states (the exchange interaction energy for the intra-valley dark exciton is zero because of the orientation of the electron and hole spins). From the calculated transition contribution maps of the excitonic states (absolute value square of the corresponding momentum component), we find that the states consist of hybridized excitations around the corresponding valleys which leads to brightening of the dark excitons, i.e., significantly decreasing their lifetime which is reflected in the photoluminescence (PL) spectrum. In addition, through application of many-body perturbation theory[3] to calculate the phonon contribution to the bandgap and exciton energies and their temperature dependencies, we find that as the temperature increases the bandgap decreases (by tens of meV at room temperature), while the binding energy of the excitons increases, while the lifetime of excitons decreases with increasing temperature. These results define the ultrafast charge dynamics in 1L WSe_2.[1]

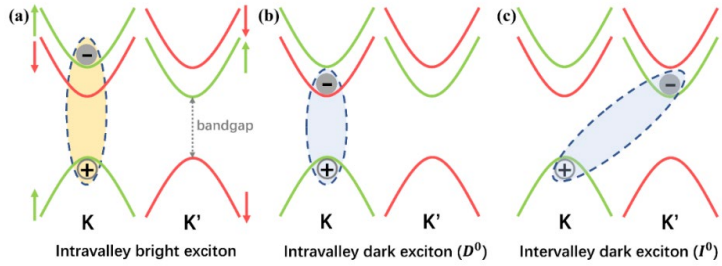

Figure 1 Bright and dark Excitons in 1L TMDs formed at time-reversal K and K' points in the Brillouin zone.

2. Intra- and inter-layer excitons and ultrafast charge dynamics in 2L WSe_2

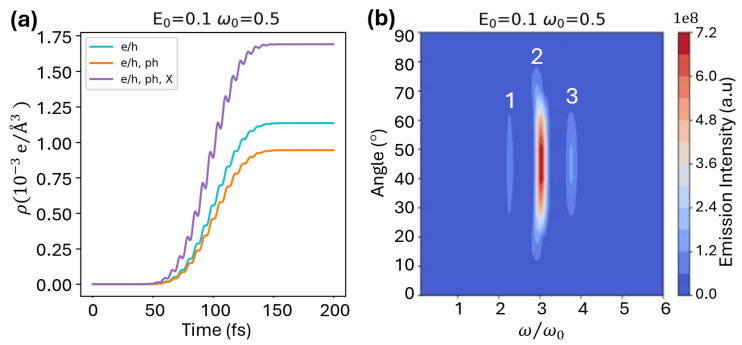

Figure 2 (a) Temporal evolution of electron charge density in 2L WSe_2 excited by 100 fs duration laser pulse. Non-interacting charge density (blue curve), charge density with phonon scattering (orange), and charge density with both phonons and excitons (purple). **(b)** The harmonic spectrum (arb. units) at different angles of the incident pulse with included free charges, excitons, and phonons.

To understand the interplay between intra- and inter-*layer* excitons as reflected in the ultrafast charge dynamics and emissive properties of bilayer systems, we have developed a real-space DM TDDFT approach. Application of the technique to 2L WSe_2, using the nanoquanta XC kernel with the electron and hole in the same and in two different layers, we find that the lowest energy intra-and inter-layer excitons are formed by electrons and holes localized mostly at the K valley. The binding energy of the *intralayer* and *interlayer* excitons turn out to be 141 meV and110 meV, respectively, in reasonable agreement with experiments (~230 meV).

Combined DM TDDFT and many-body theory analysis[3] shows the intra- and inter-layer coupling between excitons and charges (electrons and holes) leads to interlayer migration of excitons that is reflected in the emission spectrum. While momentum direct (bright) intra- and inter-layer excitons have the largest binding energy the indirect (dark) excitons also contribute to the emission spectrum. Furthermore, phonons significantly enhance the number of excited excitons (Fig. 2a) by transforming them from intra- to intervalley states with longer decay times. The role of electrons, phonons and excitons in the charge dynamics of 2L WSe_2 are further disentangled in the calculated harmonic spectrum at different angles of the incident field (with respect to the surface normal). With excitons (Fig. 2b) we get third-harmonic emission, rather than the harmonic (linear emission). Moreover, there is directional dependence of emission (maximum for pulse at 45^0). We also find that *interband* charge dynamics is always strong, independent of the pulse

strength, while the *intraband* response becomes important at strong pulses, i.e., when the system is in a non-linear regime. In the latter regime, the role of exciton is especially pronounced, as also suggested by experiments.

The calculated emission spectrum which is in a good agreement with experimental data[4] shows that both *intra-* and *interlayer* excitons account for direct ("exciton A") transition (red curve in Fig. 3a). With *intra-layer* excitons alone, the emission is predominantly indirect (Fig. 3a, black low-energy peak). That is the electron in the *inter-layer* exciton, being weakly bound to the hole, moves more easily to the K point than the *intra-layer* one because of phonon scattering and then recombines. The time evolution of the excited charge density (Fig. 3b) shows the dominant role of

the interaction of intralayer excitons with charges and phonons, which is distinct from that of interlayer excitons. Moreover, *interlayer* excitons contribute predominantly to the strong third-order harmonic generation in the system (Fig. 2b). These results establish the important role of phonon scattering in the ultrafast dynamics and optical response in bilayers TMDs.

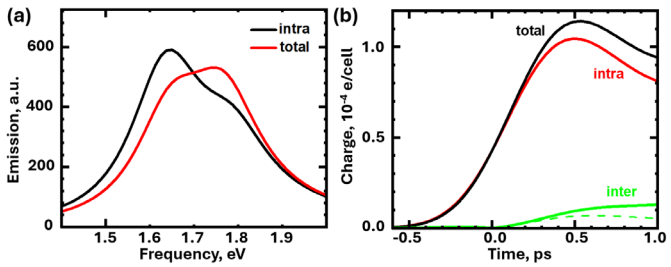

Figure 3 (a) Calculated emission spectrum of 2L WSe_2 including effects of intra- and interlayer excitons (total) and only intralayer excitons (intra). The pulse excitation parameters were chosen to compare with experimental data.[4] **(b)** Excited charge density as function of time for a pulse perturbation in different approximations: exact solution (total), with only interlayer (intra) and interlayer (inter) excitons. Dashed line – the interlayer exciton with no interaction between excitons and charges.

3. Exciton-phonon interaction and anomalous isotope effect in 1L MoS_2

With the techniques mentioned above we have explained an unusual isotope effect in the experimentally observed optical bandgap in 1L MoS_2 which found a red shift of *13 (±7) meV* of the PL peak with increasing Mo isotope mass, corresponding surprisingly to an increase in the exciton binding energy.[5] In full agreement with experimental results, we show that exciton-phonon interaction shifts the binding energy of the exciton from 0.562 eV for Mo^{92} to 0.578 eV for Mo^{100} while the PL peak shifts from 1.809 eV to 1.790 eV. We attribute this anomaly to the large binding energy of the exciton thereby a domination of exciton-phonon interaction over electron-phonon interaction. Furthermore, we find the dependence of the exciton binding energy on temperature to be stronger for the lighter isotopes, in agreement with experiments.[5]

Future Plans

Future work can be divided into two categories: 1) Examinations of excitations, spatially resolved charge dynamics, and photoluminescence in 2L TMDs (WSe_2, $MoSe_2/WSe_2$, WS_2/WSe_2), twisted and untwisted, and correlated antiferromagnetic bulk material (V_2O_3); 2) Further developments in DFT (TDDFT): enhanced exchange correlation potentials to include nonlinear and spatial and temporal non-local effects. Novel aspect of the work is the focus on spatially resolved nonhomogeneous response to external probes with inclusion of coupling of the bound and unbound charges to phonons that define the picosecond system dynamics. In the case of TMDs, a small twist-angle in the systems shows a variety of interlayer Moiré excitons, and exciton diffusion. These and other static, dynamics and PL properties will be studied in detail. In the case of V_2O_3, the effects of phonons will be analized for both charge and spin subsystems as well. For this system, our focus is ps spatially-resolved metallization and magnetization. In addition to phonons, the main addressed questions will be How do the AFM and FM domains grow? Which V and O orbitals are responsible for the metallization that is expected to predominate in the AFM vanadium planes? What is the role of spin orbit coupling in spin-flip processes? What is the contribution of electron correlations to the system response?

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Realizing and demonstrating emergent supersymmetry on quantum computers

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Keywords: Majorana fermions, emergent supersymmetry, quantum computing, variational quantum algorithm, strong correlations

Research Scope

It is known that an interacting chain of Majorana fermions with Majorana hopping and four-fermion interaction in a cluster of four consecutive Majorana sites gives rise to the tricritical Ising model at a quantum phase transition point between a gapless Ising phase and a gapped symmetry-broken phase [1,2]. The tricritical Ising model is the simplest superconformal field theory. Thus, this one-dimensional model provides an example of emergent spacetime supersymmetry in an interacting condensed matter system. However, this system has not been realized in materials-based experiments. One goal of this research project is to use the existing quantum computing devices to demonstrate the signatures of emergent supersymmetry.

Another aspect of the project concerns theoretical investigations of the nature of excited states as we perturb away from the well-understood tricritical Ising point. Of particular interest is the gapped phase in the vicinity of the tricritical point, which inherits the emergent supersymmetry but is not described by a conformal field theory. How does supersymmetry manifest itself in the excitations of this gapped phase? This project aims to answer the question above by exploring the nature of the low-energy excited states in the gapped phase adjacent to the tricritical point.

Finally, we are interested in the role of twist defects in interacting Majorana chains. When a Majorana chain is mapped to spins, the noninteracting part maps to the transverse field Ising model, with interactions mapping to multi-spin couplings. In addition to the spin-flip defects, an Ising chain supports duality defects corresponding to a Kramer-Wannier duality twist. Interestingly, the duality defects do not appear in pairs. This defect binds a single Majorana mode that is not hybridized with a partner and has an infinite lifetime, even in a finite system. This physics has been studied in the noninteracting case (transverse filed Ising). However, the behavior of the duality twist and the Majorana bound state in an interacting Majorana chain is unexplored. This project investigates an interacting Majorana chain with duality twisted boundary conditions.

Recent Progress

Of the three questions outlined above, the most substantial progress has been made on the first question, namely the realization of

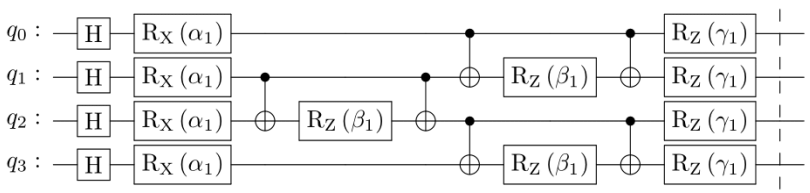

Fig. 1: An example of the structure of the variational circuit for 4 qubits, which contains 3 variational parameters.

supersymmetry on quantum computers. As a step toward experimental demonstration of supersymmetry, we created this tricritical Ising state in the IBM superconducting quantum computer and measured the correlation function of the supercharge operators.

We utilized the O'Brein-Fendley version of the model [3], which exhibits smaller finite-size effects. A mapping of the interacting Majoranas to spins, i.e., qubits, gives a spin chain with three-spin interaction. We constructed a variational quantum circuit with a quadratic number of gates (in the number of qubits) that can accurately produce the tricritical state using the variational eigenvalue solver (VQE). An example of this circuit for four qubits is shown in Fig. 1. To verify supersymmetry in the quantum device, we mapped the model's supercharge operators to spins and measured the correlation functions of these operators on the IBM device, verifying that they produce the correct critical exponents, as shown in Fig. 2.

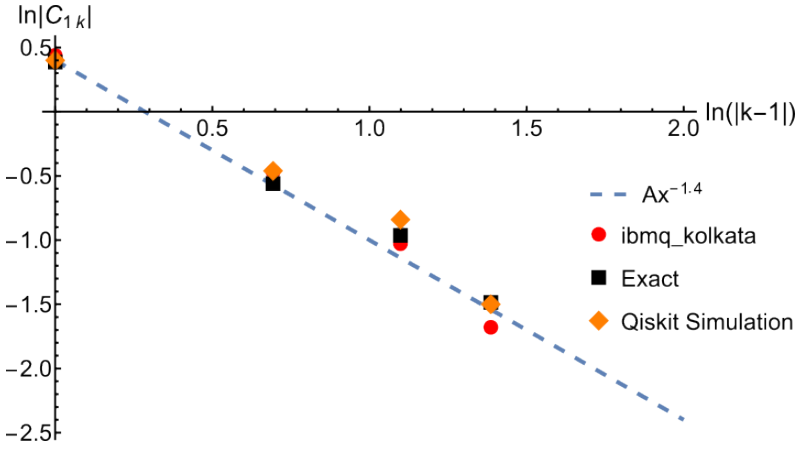

Fig. 2: Logarithm of the correlation functions of supersymmetry generators between sites 1 and k in a 7-qubit system. Exact results, Qiskit simulations of the variational circuit, and quantum hardware

Other signatures of the supersymmetric state come from various ratios of energy gaps with different boundary conditions and fermion parity (which maps to a Z2 qubit symmetry) and the central charge of the conformal field theory. We have done theoretical and numerical investigations of ways of probing these signatures in quantum computers. For gap ratios, we applied excited-state VQE [4] to create excited states of the model using a variational circuit with two copies of the ground-state circuits shown in Fig. 1. In this approach, one copy of the circuit targets the ground state, and another copy targets the excited state by adding a term proportional to the overlap of these two states to the cost function. The overlap can be measured and combined with the energy expectation value. Regarding the central charge, we have found that the central charge of the tricritical Ising model can be extracted from bitstring probabilities in the computational, i.e., Z-spin, basis, with very small finite-size effects.

Another related direction in which we have made substantial progress is the nature of the excited states in the gapped phase next to the tricritical point. Supersymmetry extends into this gapped phase even though it is spontaneously broken in the Ising phase. The ground states in the gapped phase are characterized by ordering in the occupation of certain Dirac fermions formed by combining neighboring Majoranas. We have found, by using density-matrix-renormalization-group calculations with the addition of infinitesimal perturbations to pin solitons and antisolitons,

that the low-energy excitations can be viewed as a superposition of soliton-antisoliton pairs between regions with the same ordering as the ground state. Interestingly, each soliton and antisoliton binds an emergent Majorana mode. The system with periodic boundary conditions exhibits degeneracy between even and odd fermion-parity sectors. The fermion parity for the first excited states is switched by the occupation of a Dirac mode made out of the emergent Majoranas bound to the soliton and the antisoliton.

Future Plans

The immediate future plans involve finishing and writing up two papers on the quantum computer implementation and soliton-antisolition pairs and then pushing forward the investigation of duality twisted boundary conditions. We have made preliminary progress by deriving the interacting Hamiltonian in terms of both Majoranas and spins in the presence of the duality twist and writing nonuniform density-matrix-renormalization-group codes. We have found that the ground-state degeneracy changes with duality-twisted boundary conditions. Further investigations include the nature of ordering on the two sides of the defect, associated correlation functions, and investigation of the fate of defect-bound Majorana as interactions ramp up.

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Publications

All papers supported by BES are still in preparation.

Navigating the Complex Landscape of Strongly Correlated Materials: Insights from DMFT and Machine Learning

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Keywords: Strongly correlated materials, Metal-insulator transition, Rare-earth nickelates, Dynamical mean-field theory, Machine-learning potentials

Research Scope

The electronic correlations in materials drive a variety of fascinating phenomena, from magnetism to superconductivity, due to the coupling between electrons' spin, charge, ionic displacements, and orbital ordering, also called strongly correlated materials (SCMs). These materials exhibit diverse electronic phases resulting from the interactions between electronic spins, charges, and orbitals. Within this realm, the dynamic interplay—whether cooperative or competitive—among these correlated electron phases can unveil unexpected electronic behaviors and functionalities such as metal-to-insulator transitions (MIT). Within SCMs, rare-earth nickelates (RNiO3) stand out as a fascinating class of perovskite nickelates that showcase thermally- and doping-induced MIT stemming from subtle manipulation of the electronic orbital occupancy due to non-degeneracy in the energy levels in the Ni-O framework. Recent experimental breakthroughs revealing proton-induced MIT in nickelates present a novel avenue for controlling and manipulating resistance states within correlated semiconductors using charged dopants that can migrate under electric fields. However, the dynamics of these processes, especially at nanosecond timescales pertinent to microelectronics applications, remain largely unexplored.

This research investigates methods to control and comprehend metal-insulator transitions (MIT) in rare earth nickelates under diverse stimuli, including oxygen vacancies, hydrogen doping, and alloying. Overcoming limitations of density functional theory (DFT), advanced techniques like dynamical mean-field theory (DMFT) are utilized to calculate various nickelates' energies, forces, and charges. A classical machine learning model is constructed, validated, and tested against sophisticated methodologies. Molecular dynamics simulations explore structural changes induced by proton doping, vacancy migration, and external electric fields. By comparing different nickelates, the study elucidates microstructural modifications' structural origins, advancing the understanding of correlated quantum materials and expediting the discovery of novel metastable phases.

Recent Progress

We have generated over 1000 uncorrelated configurations of SmNiO3 by simulating a Boltzmann distribution at finite temperatures. These configurations were then subjected to DMFT calculations to derive energies, which serve as the basis for training our Machine Learning (ML) potential. Our DMFT calculations were conducted using our internally developed DMFT package

(DMFTwDFT), an open-source tool designed for electronic structure calculations on SCMs. Using the data generated by DMFT, we constructed ML potentials by training neural networks (NN). The structural and energy information was fed to NequIP (Neural Equivariant Interatomic Potentials) software [1], which is an E(3)-equivariant neural network approach for learning interatomic potentials from ab-initio calculations for MD simulations. Unlike traditional symmetry-aware models employing invariant convolutions acting solely on scalars, NequIP employs E(3)-equivariant convolutions to handle interactions of geometric tensors, resulting in a more information-rich and faithful representation of atomic environments.

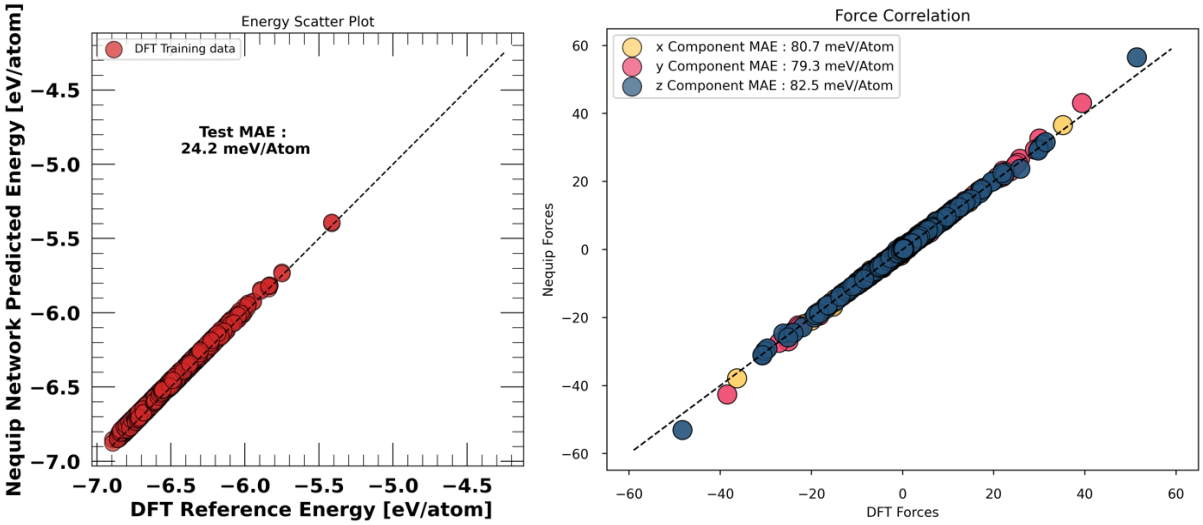

Fig 1: Performance of the NN model trained using NequIp. The left panel shows the correlation for the energy prediction of the model against the DFT reference and the right panel shows correlation for the force prediction of the model against the DFT reference for the three components in the x,y,z directions.

The outstanding correlation, with average errors of approximately 24 meV/atom for energy and around 80 meV/Angstrom for forces, positions this model as an excellent candidate for reliably probing the dynamics of H-doped $SmNiO_3$ using MD techniques. Experiments have shown chemical doping in nickelate perovskites as a way to drive MIT under isothermal conditions. While earlier DFT studies with static correlations fell short of explaining the recent MIT observations at lower hydrogen concentrations, we conducted a comprehensive computational analysis using an advanced approach. We combine DFT with DMFT (DFT+DMFT) to efficiently analyze the insulating behavior of hydrogen-doped $SmNiO_3$. In contrast to previous theoretical works, our calculations predict an insulator transition occurring at a reduced doping level of H:Ni=0.5:1. Specifically, while the DFT+U method reveals a gap opening between p-to-d orbitals, the DMFT approach highlights a gap opening between d-to-d orbitals. Our findings uncover a selective Mott transition in site and orbital characteristics, with the Ni ions proximate to the doped hydrogen exhibiting Mott-like traits. Notably, DMFT calculations highlight a pronounced dependence on Hund's parameter J, implying the presence of Hundness in the Mott insulator. Our study underscores the necessity of accounting for dynamical correlations to accurately describe the electronic structure of strongly correlated electron-doped rare-earth nickelates. Further to explore

the activation energy for H-intercalation, we computed the energy barrier for hydrogen migration in $SmNiO_3$ by employing climbing-image Nudged Elastic Band (CI-NEB) calculations. Initially, different possible pathways for migration were calculated at the level of DFT+U. The optimized structures corresponding to the peak and end-points of the minimum energy pathway were then used to obtain the energy barrier height within DMFT. A comparison of DMFT and DFT+U results indicates correlation effects on migration pathways. Further calculations are being performed to establish a better understanding of these correlation effects on hydrogen doping.

Due to the strong coupling between multiple degrees of freedom, oxygen vacancies in correlated complex oxides lead to a variety of intriguing emergent phenomena that govern magnetism, superconductivity, colossal magnetoresistance and MITs in many different oxides. Manipulating oxygen vacancies in strongly correlated RNO_3 materials enables the tuning of their elusive MIT, providing a better handle for control over their electronic properties. However, as the number of vacancies in a system increases, the possible configurational space of vacancy structures rises drastically, rendering a computational study of them quite expensive. To address this, we have studied the effects of oxygen vacancies on the electronic properties of the correlated perovskite $LaNiO_3$ using DFT+DMFT through a symmetry-adapted configurational ensemble method to obtain a reduced configurational space, thus lowering the computational cost. Using DFT+DMFT, we showed that certain vacancy configurations undergo a MIT based on the positioning of their vacancies. We also computed the transition path energy of a single oxygen vacancy through means of the NEB method. We found that the diffusion energy profile calculated through DFT+U differs from that of DMFT, possibly arising from the correlation effects that are not quite well captured with the former method. We also investigated the effect of using different double counting correction types on the height of the energy profile.

We have generalized our methods to consider the Virtual Crystal Approximation (VCA) along with DMFT to study strongly correlated alloys. For this purpose, we used two different methods, the electronic structure obtained from VCA and interfaced with our DMFT package and the Site Occupation Disorder (SOD) to study the correlated alloy $Nd_{1-x}La_xNiO_3$ as our test case. The main goal of the study was to verify if the VCA method could be used in place of the computationally expensive supercell method to study alloys. As such, we have shown that the electronic structure resulting from both methods are indeed equivalent. Following this verification, we continue to use the method to study additional systems of correlated alloys including Gd doped $SmNiO_3$.

Future Plans

The ML potential currently in testing stage, will be improved by incorporating the DMFT calculated energies during the training phase of the ML model. This generated potential will enable us to conduct large-scale (micron sized) and long run (~tens of nanoseconds) classical MD with the DMFT quality albeit at the expense of classical simulation. Utilizing the ML potentials for $SmNiO_3$, we will execute standard molecular dynamics simulations using the LAMMPS

package. As this is including the nickelate and the protonation, this can help us to understand the dynamics of the hydrogens within the nickelate. Additionally, with the classical potential, we will explore heterostructures and configurations not considered in the training data. Moreover, the generated potential will elucidate the influence of strain, oxygen vacancies, proton doping, crystal phase variation, substrate effects, vibrational effects (such as octahedral rotation), grain boundaries, and defects in the response of a MIT material under the presence of temperature and voltage. Furthermore, extensive simulations will be conducted to examine the impact of different stimuli on MIT hysteresis, as experimentally suggested. Selected MD snapshots will be analyzed using more precise theories like DMFT to provide an accurate electronic description and investigate orbital and charge rearrangements under diverse conditions.

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A High-Throughput Computational and Experimental Approach to the Design of unconventional magnets

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Keywords:

High-Throughput, Materials design, Electronic structure, synthesis, unconventional magnets

Research Scope

Prediction of a new family of dimerized quantum magnets in ternary metal borides

Dimerized quantum magnets are fascinating crystalline systems in which Bose-Einstein condensation of bosonic magnetic excitations can occur. However, the discovered dimerized quantum magnets are limited to only a few oxides and halides. Quantum magnets have attracted great interest due to their numerous exotic phenomena. In quantum magnets, tuning the quantum phase transition across the quantum critical point (QCP) can lead to different novel states of matter. One of the simplest and most intensely studied quantum magnets is dimerized quantum magnets, which are systems constituted by strongly coupled spin dimers. Antiferromagnetic intradimer coupling results in a disordered quantum paramagnetic singlet ground state due to quantum zero-point spin fluctuations. Its first excited state is an ordered triplet state. Interdimer interactions make the triplet bands dispersive. External parameters such as magnetic field, pressure, and doping have been shown to close the spin gap and generate magnetic order. Various descriptions of the ground state of the triplets, such as Bose-Einstein condensate, a triplet crystal, or a supersolid, have been proposed. Bose-Einstein condensation (BEC) of the bosonic triplons is a particularly interesting phenomenon since BEC is well-known for its role in the superconductivity of Cooper pairs and the superfluidity of 4He. In dimerized quantum magnets, at the QCP, the singlet ground state crosses the bottom of the lowest triplet band dispersion, where BEC occurs. Exploring novel

quantum magnets, states of matter in the vicinity of BEC, and parameters near the QCP is of great significance.

Recent Progress

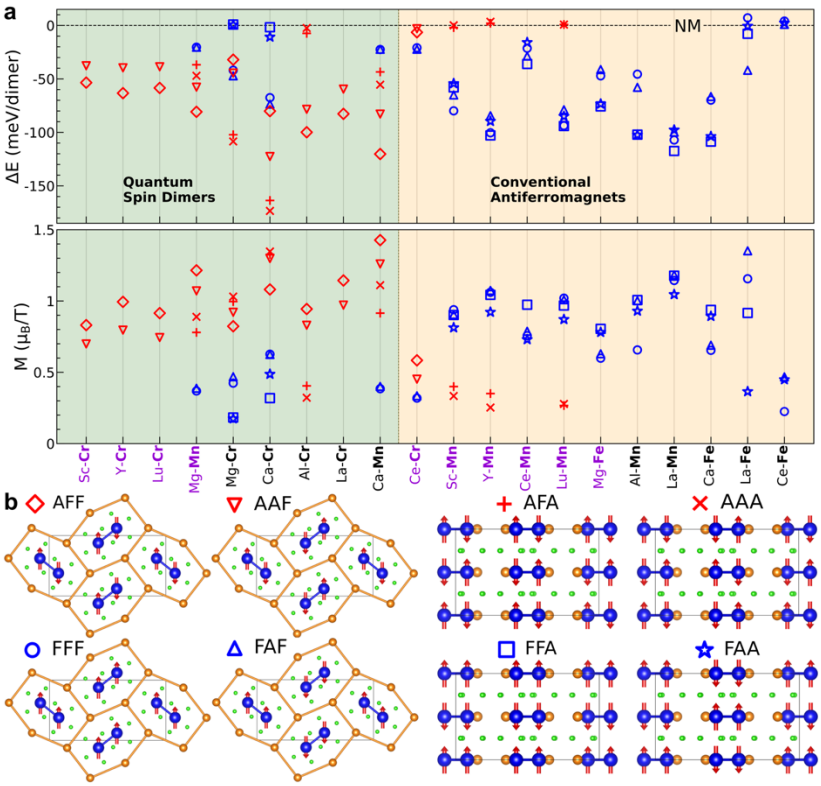

Figure 1. **$YCrB_4$-type crystal structure. a** [001] and **b** [100] projections. Orange, blue, and green spheres show M, T, and B atoms, respectively. Red arrows show magnetic moments. AFF magnetic ordering is displayed in the figure as an example. The three-letter notation for the magnetic configuration indicates the relative alignment of the local moments inside a dimer (D), between the nearby dimers in the same plane (L2), and between the neighboring layers (Z).

Using first-principles calculations, we identified 21 structurally stable and 25 metastable ternary metal borides of MTB_4-type (M = Sc, Y, La, Ce, Lu, Mg, Ca, Al; T = V, Cr, Mn, Fe, Co, and Ni). Among them, we uncovered 20 magnetic systems, the rest being non-magnetic. Electronic and magnetic calculations for this family of materials reveal similarities in their electronic and magnetic structures for those compounds with the same valence shell electron count. Magnetism in these compounds is dominated by strong antiferromagnetic Cr (and Mn for M = Mg and Ca) or ferromagnetic Mn (and Fe for M = Mg and Ca) interactions within the structural dimers, with much weaker interactions between the dimers. The magnetic ground states in DFT are semiconducting for Cr systems or weakly metallic for Mn systems. Mn compounds are predicted to be conventional Néel antiferromagnets with layered (A-type) ordering and T_N below room temperature. In contrast, Cr compounds are proposed to be close to a quantum critical point between a singlet spin-dimer phase, with a spin gap above room temperature, and the conventional Néel antiferromagnetic phase. We identified nine dimerized quantum spin-gap systems and 11 conventional antiferromagnets. Notably, we proposed 4 stable ($ScCrB_4$, $YCrB_4$, $LuCrB_4$, and $MgMnB_4$) and 5 metastable ($MgCrB_4$, $CaCrB_4$, $AlCrB_4$, $LaCrB_4$, and $CaMnB_4$) quantum spin-dimer magnets. They provide a unique possibility for investigating Bose-Einstein condensation of magnons [1]. The prediction of this new family of dimerized quantum magnets greatly expands the materials

inventory, allowing such an investigation. Experimental methods to produce single-phase $YFeB_4$ samples suitable for characterization are also reported. All the stable and metastable MTB_4 systems identified in this study, no matter quantum-magnetic, conventionally magnetic, or non-magnetic, provide a platform with numerous possibilities for future doping on the M site or the T site. Tuning the exchange coupling by doping may facilitate the study of this rare type of quantum phase transition across the spin-gap quantum critical point.

Future Plans

We will perform a high-throughput study of the following systems

Quantum Critical Point (QCP) Monoboride Alloys Analysis:

- We identified new quantum critical point monoboride alloys (for instance, Mn, Fe in (Ni, Co)B). We are now working on a complete theoretical and experimental analysis of the structure, excitations, and magnetism around this QCP.
- We identified a new family of potentially altermagnetic systems by screening materials in materials databases.

Borides:

- 1-1-2 borides family (>100 systems) to determine stable magnetic systems and grow some for magnetic measurements.
- Ternary borides structures (1-1-4, 1-2-2, 2-3-5). These systems provide an opportunity to tune magnetic properties by increasing a concentration of TM element. Preliminary computational data indicated systems with altermagnetic effects, as well as magnetocaloric effects in these systems (see preliminary results). For the novel borides, stability and synthesizability for known structures with 3d-metal substitutions will be computationally accessed and experimentally verified.
- The intrinsically *chiral* borides will be studied due chiral crystal structures (CCS): $A_2Os_3B_5$(A = Ca, Eu), $CePt_2B_{1+x}$, REM_2B_2 (M = Ru, Os, RE=La-Gd), ANi_3B_2 (A = Li, Mg). The alloying by 3d-elements could result in magnetic ground state, while CCS will ensure chirality of the magnetic states.

Multinary high-T_c chalcogenides:

- For the quaternary and higher multinary chalcogenides $(RE)_6(TM)_{0.5\text{-}2}X_2Q_{14}$ the stability of a number of members has been demonstrated as well as variability of the transition metal content and TM-TM distances in the linear 1D chains of transition metals.
- Chiral sulfides and selenides, space group $P6_3$, will be studied on the premise that compounds with 3d transition metals, V-Ni, will generate chiral magnetism in $REa_6M_xSi_2S_{14}$ and $RE_6M_xAl_2S_{14}$ (M is transition metal and x is governed by M oxidation state).

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Publications

1. Zhen Zhang, Andrew P. Porter, Yang Sun, Kirill Belashchenko, Gayatri Viswanathan, Arka Sarkar, Kirill Kovnir, Kai-Ming Ho, and Vladimir Antropov, “A High-Throughput Computational and Experimental Approach to the Design of unconventional magnets”, submitted

Catalysing Chiral Quantum Matter at Hofstadter Van Hove Singularities

Luiz Santos, Emory University

Keywords: Chiral Quantum Matter, Topological Phases, Fractal Lattices, Electronic Singularities.

Research Scope

This project seeks to investigate and comprehend novel electronic phases within chiral quantum materials. These materials possess distinctive properties due to their chirality, originating from sources such as atomically thin layered materials or from the surfaces of three-dimensional topological materials. The focus lies on delving into the complex nature of interacting electronic orders within topological and fractal Hofstadter electronic bands while exploring their rich structure of Van Hove singularities. These explorations lay the groundwork for the emergence of new electronic states of matter, potentially leading to the development of innovative materials with tailored electronic properties, thus advancing areas such as quantum computing, electronics, and energy storage. The project is structured around three primary goals: (1) to identify novel electronic states characterized by fractal dimension and energy bands featuring higher-order Van Hove singularities as platforms for emergent correlated electronic states; (2) to characterize critical phenomena in topological and fractal lattices, exploring connections with symmetry, topology, and quantum geometry; (3) to characterize novel competing electronic orders in two-dimensional lattices supporting topological and Hofstadter electronic states, aiming to uncover new mechanisms for unconventional superconductivity, chiral charge and spin density wave orders, gapless fermionic matter beyond the Fermi liquid paradigm, and fractionalized states of matter.

Recent Progress

(i) In **publication P1**, we have introduced a renormalization group (RG) framework to explore competing electronic orders in Hofstadter lattices supporting near flux quantum per unit cell, motivated by their experimental realization in moiré heterostructures (Figure 1). Exploring symmetry and topology aspects that are characteristic to Hofstadter systems, our research has demonstrated that repulsive interactions among fractal electrons can drive instabilities towards correlated charge and spin density wave states, as emerging chiral topological superconductivity. Furthermore, in **publication P2**, we have demonstrated that Hofstadter systems are fertile ground to achieve pair-density wave (PDW) superconductivity, where we uncovered new microscopic mechanisms for these exotic phenomena in the pi-flux Hofstadter square lattice when the Fermi energy lies near logarithmic Van Hove singularities. These findings not only advance our understanding of

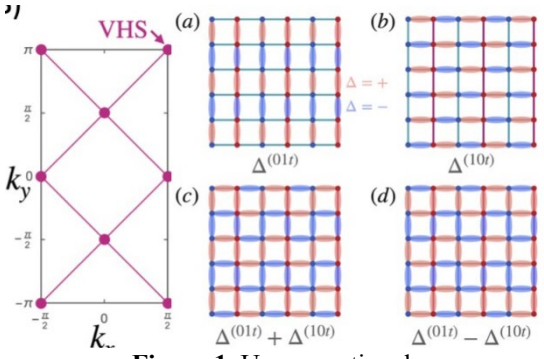

Figure 1. Unconventional superconductivity in Hofstadter lattices. Publications: Nat. Comm. (2022) and Phys. Rev.

unconventional superconductivity beyond the BCS paradigm but also open new avenues for designing quantum platforms for topological superconductivity.

(ii) We have identified new topological band structures supporting higher-order Van Hove singularities (HOVHS), and proposed mechanisms for their realization in quantum materials (Figure 2). In **publication P3**, we achieved a new understanding of HOVHS in Chern bands. In particular, our analytical classification of Fermi surface topological transitions unveiled a rich landscape of HOVHS in the Haldane honeycomb model, opening a new platform to investigate correlated electronic phenomena in chiral topological systems. Motivated by these findings, in **publication P4** we have demonstrated a promising path to realizing these higher-order singularities through heterostructures of topological insulators and ferromagnetic materials. Our findings deepen the knowledge of electronic phenomena and suggest intriguing avenues for future research, especially regarding the interplay between topology and magnetism.

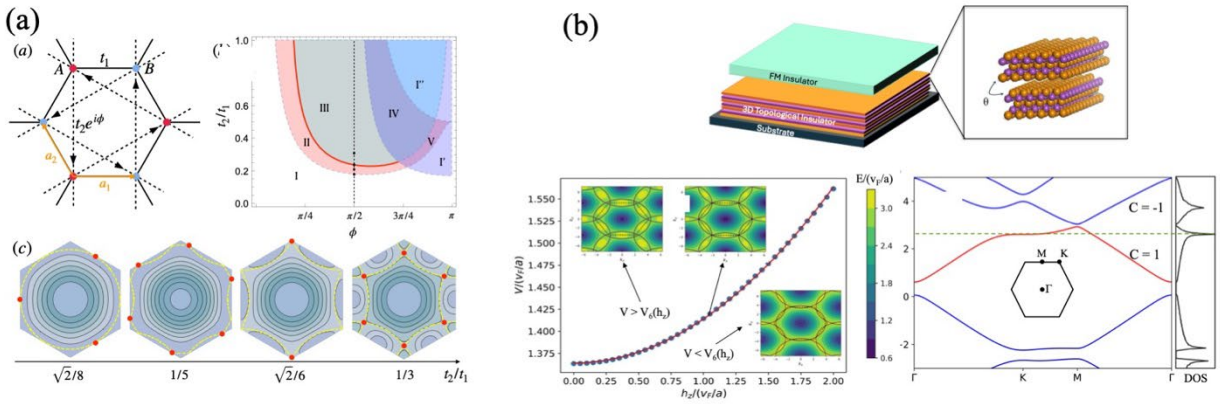

Figure 2. (a) Classification of HOVHS on the Haldane model (Phys. Rev. Lett. 2023). (b) Mechanism to realize HOVHS in Chern bands on the surface of topological insulator – ferromagnetic heterostructure.

(iii) We have uncovered new platforms and mechanisms for realizing interacting gapless chiral fermions and fractionalized topological matter. In **publication P3,** we have shown that the interplay of band topology and repulsive interactions can stabilize a new interacting non-Fermi liquid state manifesting anomalous Hall conductivity in the absence of an external magnetic field, dubbed Chern supermetal. We have found that this exotic gapless state forms due to a sublattice polarization in topological bands that strongly constraints the effect of local interactions. In **publication P5** we have extended this sublattice polarization framework to time-reversal invariant topological bands in moiré bilayers. Our work uncovers a new platform for realizing the fractional quantum spin Hall (FQSH) effect, laying out a framework to characterize this long sought-after effect that has recently been reported on twisted MoTe2 bilayers.

(iv) In **publication P6** we have proposed and investigated a new class of time-reversal symmetric quantum fractal states and demonstrated a path to their realization on the surface of three-dimensional topological insulators (Figure 3). We dubbed these emerging electronic fractal states *Dirac Fractals,* and characterized their novel fractal properties through measures of inverse-participation ratio and fractal dimensionality, achieved via large-scale exact diagonalization. This accomplishment expands the realm of fractal electronic states beyond Hofstadter systems and opens a research direction to achieve unconventional transport phenomena through self-similar fermionic states in topological materials.

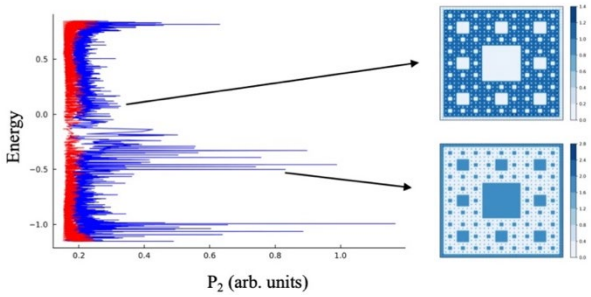

Figure 3. Dirac Fractals on the surface of a topological insulator. Phys. Rev. B 108, 155430 (2023).

Future Plans

Our framework to investigate electronic instabilities near Van Hove singularities present in topological bands paves new routes to explore novel electronic orders in topological quantum materials. We will investigate whether unconventional wave superconductivity and quantum critical phenomena can be stabilized by local interactions in 2D quantum materials supporting quasi-flat bands. We will explore these prospects in kagome lattices and moiré heterostructures (graphene and transition metal dichalcogenide) having electronic states highly tunable via external electric and magnetic fields, while harboring valley-polarized chiral electrons near Van Hove singularities.

Drawing from our understanding of Hofstadter systems and unconventional superconductivity, we will investigate emergent fractionalization phenomena within Hofstadter lattices. This endeavor is spurred by recent groundbreaking experimental discoveries of Fractional Chern Insulators (FCI) and Fractional Quantum Spin Hall (FQSH) states in moiré heterostructures. In light of these developments, we will (i) characterize novel Abelian FCIs using Hofstadter composite fermion theory, (ii) explore pairing of Hofstadter composite fermions to achieve novel Non-Abelian FCI and FQSH states beyond Landau level paradigm, and (iii) probe the stability and nature of fractionalized lattice fermions through exact diagonalization and large-scale Density Matrix Renormalization Group methods.

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Quantum Simulations of the Interplay of Charge Density Wave, Magnetic, and Pairing Correlations

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Keywords: Quantum Simulations, Magnetism, Exotic Superconductivity

Research Scope

Understanding the competition of different low temperature phases in strongly correlated electron systems is a core endeavor of condensed matter physics. Here we describe our progress using Quantum Monte Carlo (QMC) methods to simulate models which incorporate multiple types of electron-phonon interactions, studies of the Su-Schrieffer-Heeger-Hubbard Hamiltonian, and the use of the 'sign problem' (SP) as a tool to infer critical behavior. We then discuss future plans- AI methods for electron-phonon models, quantum simulations of N-component fermions, and continued exploration of the SP.

Recent Progress

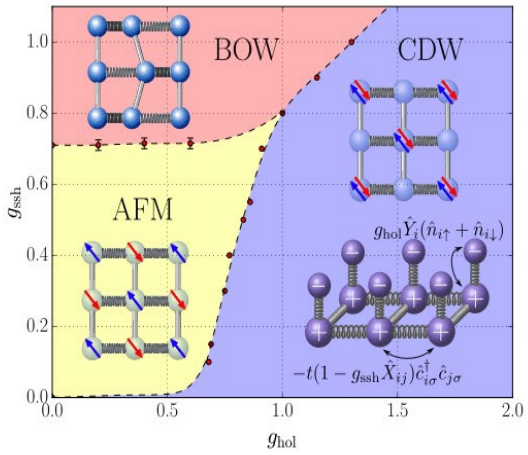

Figure 1: The low-temperature phase diagram of the SSH-Holstein model. CDW order is present at any $g_{\rm hol}$ for $g_{\rm ssh} = 0$. BOW dominates at large $g_{\rm ssh}$. For $g_{\rm ssh} \leq 0.7$, antiferromagnetic order is present for low Holstein couplings in place of BOW. Bottom right (in the blue zone): A sketch of the SSH-Holstein model.

a) Su-Schrieffer-Heeger-Holstein Hamiltonian: The interaction of electrons with vibrational modes of a solid is a key area of study of material sciences, with the potential dramatically to alter the motion of 'dressed' quasiparticles (polaron formation) and also to result in various charge and bond ordered phases and superconductivity. Two fundamental types of interaction are commonly studied- vibrations which couple to the density of electrons on a site, and those coupling to intersite hopping. We have computed the equilibrium properties of the Su-Schrieffer-Heeger-Holstein (SSHH) Hamiltonian which combines both these modes,

$$\begin{aligned}\widehat{\mathcal{H}} = \; & -t\sum_{\langle i,j\rangle,\sigma}\left(1-\alpha_{\rm ssh}\hat{X}_{ij}\right)\left(\hat{c}^\dagger_{i\sigma}\hat{c}^{\pm}_{j\sigma}+\text{h.c.}\right)-\mu\sum_{i,\sigma}\hat{n}_{i\sigma}+\sum_{\langle i,j\rangle}\left(\frac{\hat{P}^2_{ij}}{2M_{\rm ssh}}+\frac{M_{\rm ssh}}{2}\omega^2_{\rm ss}\right. \\ & \left.+\alpha_{\rm hol}\sum_{i,\sigma}\hat{Y}_i\,\hat{n}_{i\sigma}+\sum_i\left(\frac{\hat{\Pi}^2_i}{2M_{\rm hol}}+\frac{M_{\rm hol}}{2}\omega^2_{\rm hol}\hat{Y}^2_i\right)\right.\end{aligned} \quad (1)$$

We considered a square lattice geometry and obtained the phase diagram shown in Fig. 1.

b) Su-Schrieffer-Heeger-Hubbard Hamiltonian: We similarly studied the SSH-Hubbard model where an attractive electron-electron interaction replaces the Holstein coupling in Eq. 1. Figure 2 shows the abrupt changes in various observables as phase boundaries are crossed.

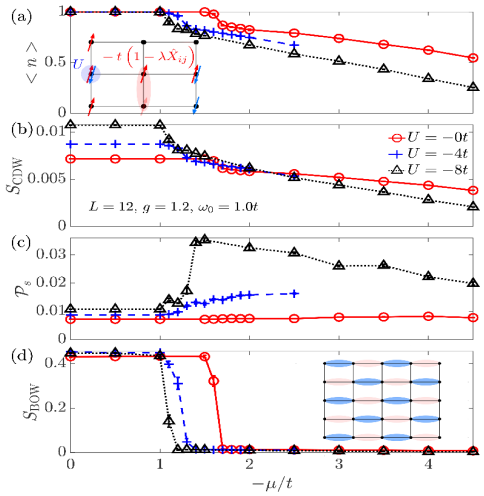

Figure 2: QMC simulations of the SSH Hamiltonian with an on-site attractive electron-electron interaction U. (a) Average density $\langle n \rangle$ as a function of chemical potential μ. The plateau measures the BOW gap, and is reduced by U. b) The CDW structure factor S_{CDW} remains small for these parameter choices. (c) s-wave pairing correlation $\mathcal{P}_s$; (d) The BOW structure factor S_{BOW} decreases rapidly when $\langle n \rangle$ shifts from commensurate filling (panel (a)); Inset of panel (a): representation of the el-el and el-ph interactions. Inset of panel (d): representation of the $q = (\pi, \pi)$ BOW. Different bond colors emphasize the alternation in bond lengths. The spatial lattice is 12×12, electron-phonon coupling $\lambda = 1.2\,t$ and phonon frequency $\omega_0 = 1.0\,t$.

c) Criticality and the Sign Problem: The "sign problem" (SP)[1] is the fundamental limitation to simulations of strongly correlated materials in nuclear matter, quantum chemistry, condensed matter physics, and quantum chromodynamics. In this project we undertook a study[2] of possible connections between the SP in determinant quantum Monte Carlo (DQMC) to quantum critical behavior in several fundamental models of condensed matter physics, including the spinful and spinless Hubbard Hamiltonians on a honeycomb lattice and the ionic Hubbard Hamiltonian, both of whose critical properties are relatively well understood. We then proposed a reinterpretation of the low average sign for the Hubbard model on the square lattice when away from half-filling, an important open problem in condensed matter physics, in terms of the onset of pseudogap behavior and exotic superconductivity.

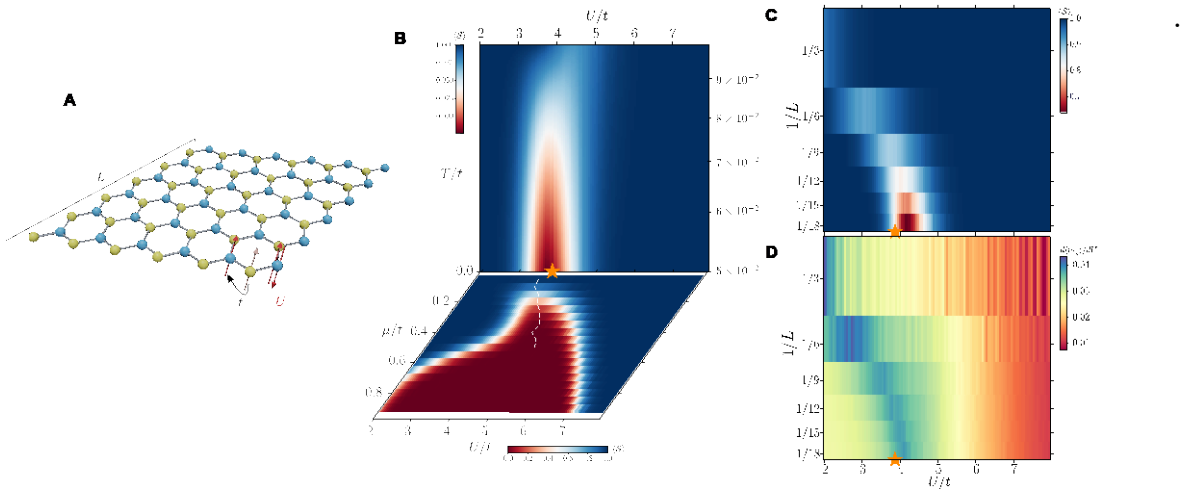

Figure 3: (A) Cartoon depicting a honeycomb lattice with $N = 2L^2$ sites ($L = 6$ here), accompanied by the relevant terms in $\widehat{H}$. (B) Contour plot of the average $\langle \mathcal{S} \rangle$ in the T/t (μ/t) vs U/t in the upper (lower) panel. Here $L = 9$ and $\mu/t = 0.1$ ($T/t = 1/20$) in the upper (lower) panel. (C) The average sign extrapolated with the linear system size L, using $T/t = 1/20$ and $\mu/t = 0.1$. (D) Similar extrapolation as in (C), but displaying a local quantity (the derivative of the double occupancy), which is an indicator of the QCP. In all panels with

data, the prediction for the ground-state phase transition occurring at $U_c/t = 3.869$ [3] is depicted by a star marker.

Figure 3 shows one example of our results the Hubbard Hamiltonian on a honeycomb lattice. In such a geometry (Fig. **A**), the $U = 0$ Hubbard Hamiltonian has a semi-metallic density of states which vanishes linearly at $E = 0$. Unlike the square lattice that displays AF order for all $U \neq 0$, the honeycomb Hubbard model at $T \to 0$ remains a semimetal for small nonzero U, turning to an AF insulator only for U exceeding a critical U_c[3]. Despite the fact that it is a QMC construct rather than a physical observable, the behavior of the sign seems to show sharp features at the semimetal to antiferromagnet boundary. Finite size scaling reinforces the connection.

Future Plans

a) AI methods for electron-phonon models: Figure 4 exhibits preliminary work applying a machine learning method known as 'learning by confusion' (LbC) [4] to the CDW phase transition of the Holstein Hamiltonian. The idea behind LbC is to present a collection of configurations to an artificial neural network across a parameter region $\lambda_1 < \lambda < \lambda_2$ spanning a phase transition. The configurations are then labeled as being above or below a critical coupling (or temperature) λ_c and the ability of the ANN to reproduce the label is measured. A plot of the accuracy $\mathcal{A}(\lambda_c)$ after training takes on a 'W' shape, with the central maximum determining the true value of the critical coupling. Precisely this structure is observed in Fig. 4, which represents the first attempt to use LbC for an interacting fermion Hamiltonian. We plan to use LbC, and a broader set of AI approaches, to study the CDW, BOW, and SC phase transitions in the Holstein and SSH Hamiltonians.

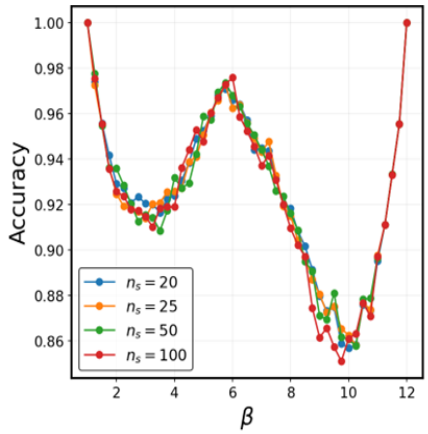

Figure 4: Using 'learning by confusion' to locate the critical temperature of the 2D Holstein Hamiltonian: Phonon frequency $\omega_0 = t$ and electron-phonon coupling $\lambda = t$. The parameter n_s controls the number of Monte Carlo sweeps between snapshots, so that $n_s = 100$ corresponds to $1/5$ as much data as $n_s = 20$. Here we train on density profiles $n_{\mathbf{i}\uparrow} = 1 - G_{\mathbf{i}\uparrow}$, but we have also explored using the phonon coordinate $x_{\mathbf{i}}$. LbC yields a value for $\beta_c \sim 5.9\,t$, in good agreement with estimates based on finite size scaling of the CDW structure factor [5].

b) QMC for multi-orbital fermions: We plan to develop further QMC codes for N-component fermion systems. These are of interest to study novel types of magnetic ordering and also multi-orbital and twisted multilayer systems.

c) Using the sign problem to detect superfluid phase transitions: An interesting generalization of the idea of using the sign to probe non-analyticities associated with phase transitions in fermionic models exploits the idea that a phase transition in a bosonic system can also be revealed by the sign problem. We plan to explore the possibility of detecting a superfluid transition in 2D hardcore bosons via the SP which arises in world-line QMC methods for non-interacting fermions.

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First-Principles Understanding of Optical Excitations within Low-Dimensional Materials

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Keywords: two-dimensional materials; excitons; electron-phonon interactions

Research Scope

Understanding how to control the nature, energy, and movement of bound electron and holes (excitons) formed upon optical excitation is a challenge in designing better optoelectronic and photonic materials. The objective of this project is to utilize first-principles computational approaches to understand optical excitations within low-dimensional materials, with the ultimate goal of designing new materials by modifying the chemical and physical structure on the nanoscale. Controlling the nature and migration properties of excitons is a challenge because of the complex relationship between electrons, phonons, spins, defects, disorder, and dynamics. By analysis of highly accurate density functional theory (DFT) and many-body perturbation theory (MBPT) calculations, we aim to decompose these complexities into simple, physically intuitive models.

The specific goals of this project are to 1) develop a theoretical understanding and simple physical models of electron- and exciton-phonon interactions in low-dimensional and layered materials; 2) develop a model of electron excitation dynamics at interfaces; 3) understand excitons in layered magnetic van der Waals materials; and 4) quantify and tune the localization of excitons, exciton-phonon interactions, and dephasing in defective low-dimensional materials.

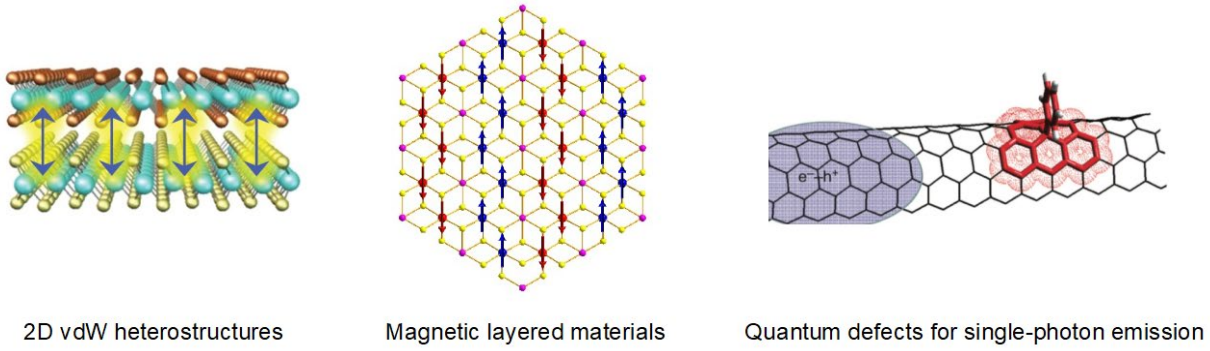

Figure 1: A schematic of the Project Scope. Left: interlayer interactions in 2D materials. Center: Excitonic effects in anti-ferromagnetic layered materials; Right: excitonic effects in the presence of quantum defects in carbon nanotubes.

Recent Progress

I. Electron- and Exciton-Phonon Interactions

Electron-phonon interactions can dominate the electronic behavior of materials. As such, they can be exploited for technological applications or may need to be minimized for improved materials. Understanding the microscopic origins of electron-phonon interactions is crucial for comprehending the functional behavior of materials, particularly low-dimensional materials, where reduced dimensionality leads to increased interactions [1]. However, electron-phonon interactions are complex with many degrees of movement associated with atoms. Here, we aim to develop a physical intuition for understanding electron- and exciton-phonon interactions.

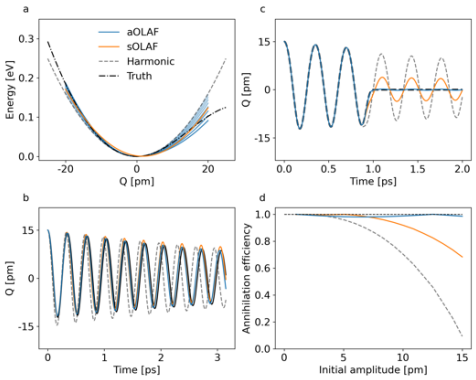

Figure 2: a) The calculated potential energy surface along the A1g coordinate of bulk bismuth compared with the harmonic approximation of the PES, the symmetric (s), and asymmetric (a) effective harmonic approximation (OLAF). b) Phonon trajectories initialized with an amplitude of 10 pm, corresponding to the ground truth PES, harmonic, and two OLAF approximations. c) Coherent phonon annihilation via 2-pulse excitation within the three approximations for an initial amplitude of 15 pm. d) The proportion of the oscillation amplitude that is removed after the second pulse. From Ref. 9.

We performed first-principles density functional theory (DFT) and time-dependent DFT (TDDFT) calculations of coherent phonons in bismuth and antimony and formulated a classical, parametrized model for phonon dynamics, introducing an effective linearization of anharmonic effects associated with coherent phonons. Using this model, we created a protocol for coherent control of these phonons, which we termed OLAF, that is predictive and able to describe weakly anharmonic behavior of coherent phonons using linear ordinary differential equations (Figure 2). This work has been published [9] and we are working with experimentalists to test our model.

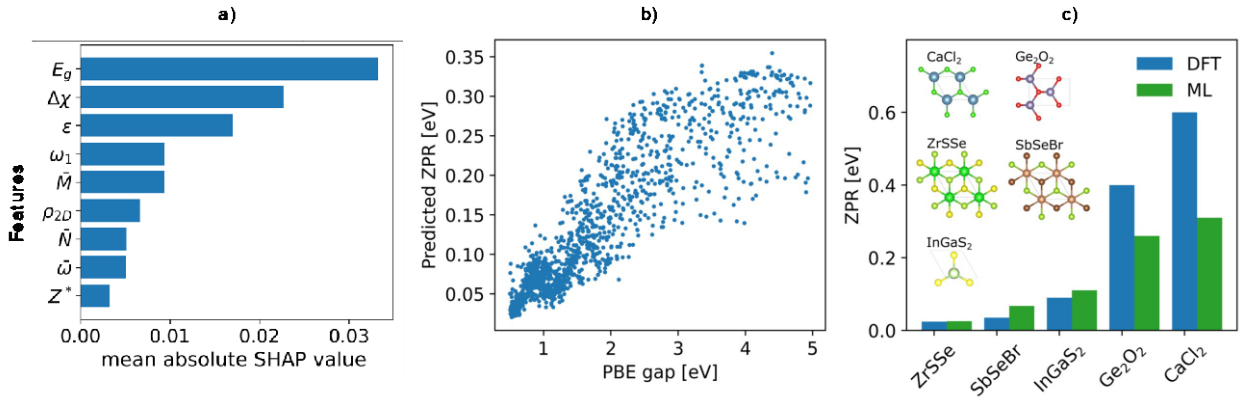

Figure 3: a) Important features that correlate with ZPR as predicted by feature selection analysis. b) ML model prediction of the ZPR of 1302 materials as a function of the DFT-predicted bandgap. c) Comparison of ML and DFT-predicted ZPR five randomly selected materials studied. From Ref. 6.

We performed a data-driven analysis of electron-phonon interactions in 2D materials. We applied the special displacement method [2] to describe the thermally-averaged impact of phonons on the electronic structure of over 100 materials included within the MC2D database and determined underlying relationships between material properties and phonon-induced band gap renormalization using an ensemble-model-based approach (Figure 3). We determined that the band gap, electronic dielectric constant, ionicity, and average mass are the most important features determining electron-phonon interactions. We applied the machine learning (ML) model to the C2DB database, which includes over 1300 2D materials. This model, based on extra random trees [3], allows us to predict a complex material property that has so far remained expensive and intractable in a high-throughput manner [6].

II. Quantum Defects in (6,5) Single-Walled Carbon Nanotube (SWCNT)

Quantum defects in low-dimensional materials can lead to trapped excitons and single photon emission (SPE) [4]. The magnitude of the exciton trapping potential determines the operating temperature of these devices while the dephasing time determines their functionality [5]. Within this class of materials, exciton-phonon coupling can lead to a larger trapping energy, improving the device, while simultaneously leading to exciton decoherence, reducing its dephasing time. Here, we aim to explore the relationship between exciton localization, the trapping energy, exciton-phonon interactions, and SPE.

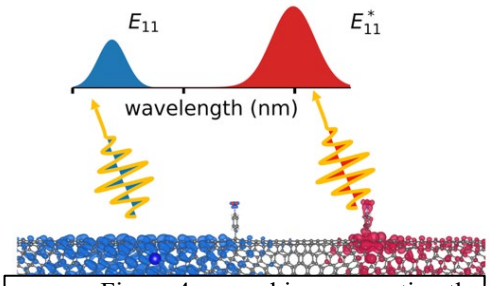

Figure 4: a graphic representing the optical and emission wavelength of doped SWCNT along with the distribution of the exciton. From Ref. 10.

To understand the excitonic properties of this system, we applied both TDDFT and MBPT, and utilized constrained DFT to quantify the Stokes shift. By comparing the predicted and experimental optical absorption and photoluminescence for nitrophenyl-doped (6,5) SWCNT, we determined that the exciton associated with the observed single-photon emission is a valence to conduction type excitation red-shifted due to defect-induced symmetry breaking and the presence of the defect-induced in-gap state. Furthermore, we found an asymmetry between the contribution of the two spin channels, suggesting this system has potential for spin-selective optical transitions [10]. We are now studying (6,5) SWCNT doped with sulfur dioxide (SO2), which was synthesized and displayed similar low-energy peaks as sp3 defects. By calculating the adsorption energy of SO2 and its derivatives, and comparing with measurements, we determined that SO2 breaks down to either S or SO on the surface and are studying the excited-states associated with these likely defects.

Future Plans

We aim to continue to build a more comprehensive understanding of the functional characteristics of excitons: exciton wavefunction localization, spin-selectivity of excitations, and electron and exciton coherence.

We will continue to study exciton-phonon interactions, focusing on polar layered materials, particularly hybrid perovskites where we have previously found polaronic effects were important [8]. While our initial calculations provided qualitative insight into the experimental finding of the role of ligand modification on exciton-phonon coupling, we were missing important physical

features such as polaronic distortions and the dynamic screening by the lattice of the electron and hole polaron. The overall goal of this work is to elucidate the complex interplay between electronic and lattice dynamics in this important class of materials.

We will continue to study exciton localization within defective (6,5) SWCNT, comparing to experimental studies of single photon emission in these systems. The zero-spin system based on SO2 provide a contrast to our previous studies of sp3 defects where an unpaired spin was introduced at the bonding site. Additionally, for the defects with a net magnetic moment, we will study spin selectivity of optical excitations. For this system, we will also explore new methods for determining exciton coherence times. Furthermore, we have initiated and will continue to study NH3 adsorption on defective MoS2, a precursor to the formation of Mo5N6. We will study excitonic and electrocatalytic properties of Mo5N6, particularly with defects.

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Novel topological quantum phases and topological superconductivity for Moiré superlattices

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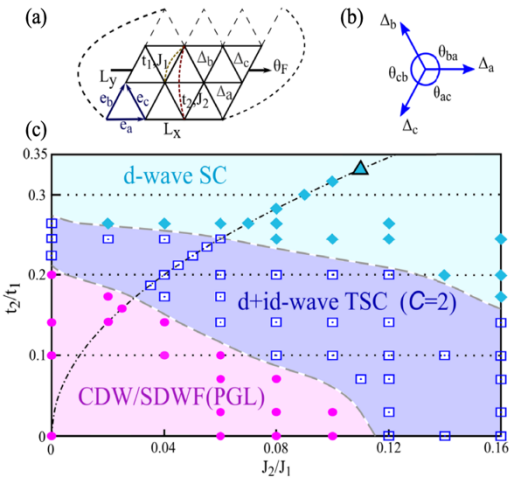
Keywords: superconductivity, topological order, pair density wave, quantum anomalous Hall effect

Research Scope

This project aims to answer fundamental questions regarding the emergence of topological phases including topological superconductivity, fractional Chern insulator, and novel quantum phases with intertwined topological and symmetry breaking orders in the strongly interacting systems modeling Moir`e superlattices. Through advancing the numerical algorithms based on density matrix renormalization group (DMRG) and exact diagonalization, the PI and her team extensively explore quantum phase diagrams for different classes of strongly correlated systems. We aim to identify and reveal the interplay of topological superconducting states with other correlated states in the quantum phase diagrams and uncover the mechanism of the emergence of the unconventional superconductivity in these systems. We have made significant progress in the following areas: (i) Identifying the quantum phase diagram for doped Mott insulators on triangular lattice with including further neighboring couplings, chiral interaction and displacement field suitable for Moir`e systems. (ii) Revealing the competition and interplay between charge density wave, spin order, and unconventional superconductivity for cuprate systems, which lead to an improved understanding for the mechanism of high-Tc superconductivity. (iii) Identifying and predicting a novel quantum anomalous Hall insulator phase with co-existing topological order, spontaneous ferromagnetism and charge density wave for Moir`e superlattice systems. The improved understanding and quantitative prediction on the complex properties of these interacting systems will advance our knowledge of the correlated physics, provide valuable information and guidance for the development of analytic theories and experimental discovery of new topological states of matter.



Quantum phase diagram for extended t-J model describing Moir`e system on triangular lattice. We identify charge density wave, topological chiral superconductivity (SC) and d-wave SC.

Recent Progress

Quantum phase diagram, spontaneously emergent topological chiral superconductivity and pair density wave for triangular lattice systems—The topological superconducting state is a highly sought-after quantum state hosting topological order and Majorana excitations. In this direction, we first explore the mechanism to realize the topological superconductivity (TSC) in the doped

Mott insulators with time-reversal symmetry modeling the twisted bilayer transition metal dichalcogenide (TMD) systems with short-range interactions. Through large-scale DMRG study of an extended triangular-lattice t−J model on the six- and eight-leg cylinders, we identify a d+id-wave chiral TSC with spontaneous TRS breaking, which is characterized by a Chern number C=2 and quasi-long-range superconducting order. We map out the quantum phase diagram with by tuning the next-nearest-neighbor (NNN) electron hopping and spin interaction. In the weaker NNN-coupling regime, we identify a pseudogap phase with a charge stripe order coexisting with fluctuating superconductivity, which can be tuned into d-wave superconductivity by increasing the doping level and system width. The TSC emerges in the intermediate-coupling regime, which has a transition to a d-wave superconducting phase with larger NNN couplings. The emergence of the TSC is driven by geometrical frustrations and hole dynamics which suppress spin correlation and charge order, leading to a topological quantum phase transition.

Furthermore, we model the homobilayer TMDs under displacement field which can be described by a generalized triangular-lattice Hubbard model with a spin-dependent hopping phase θ. To explore the effects of spin-phase on the system, we perform DMRG calculations for the relevant triangular lattice *t-J* model. By changing θ at small hole doping, we obtain a region of quasi-long-range superconducting order coexisting with charge and spin density wave within $0<\theta<\pi/3$. The superconductivity is composed of a dominant spin singlet d-wave and a subdominant triplet p-wave pairing. Intriguingly, the $S_z=\pm1$ triplet pairing components feature pair-density waves. In addition, we find a region of triplet superconductivity coexisting with charge-density wave and ferromagnetism within $\pi/3<\theta<2\pi/3$, which is related to the former phase at smaller θ by a combined operation of spin-flip and gauge transformation. Our findings provide insights and directions for experimental search for exotic superconductivity in twisted TMD systems.

Mechanism to cuprate superconductivity--The square-lattice Hubbard and closely related t−J models are considered as basic paradigms for understanding strong correlation effects and unconventional superconductivity (SC). Recent large-scale DMRG simulations on the extended t−J model have identified d-wave SC on the electron-doped side (with the next-nearest-neighbor hopping $t_2>0$) but a dominant charge density wave (CDW) order on the hole-doped side ($t_2<0$), which is inconsistent with the SC of hole-doped cuprate compounds. We re-examine the ground-state phase diagram of the extended t−J model by employing the state-of-the-art DMRG calculations with much enhanced bond dimensions, allowing more accurate determination of the ground state. On six-leg cylinders, while different CDW phases are identified on the hole-doped side for the doping range $\delta=1/16-1/8$, a SC phase emerges at a lower doping regime, with algebraically decaying pairing correlations and d-wave symmetry. On the wider eight-leg systems, the d-wave SC also emerges on the hole-doped side at the optimal 1/8 doping, demonstrating the winning of SC over CDW by increasing the system width. Our results not only suggest a new path to SC in general t−J model through weakening the competing charge orders, but also provide a unified understanding on the SC of both hole- and electron-doped cuprate superconductors.

Identifying quantum anomalous Hall-crystal—TMD moir`e systems have been experimentally demonstrated to host various fractional Chern insulating phases with quantized Hall conductance at zero magnetic field. Based on exact diagonalization simulations, we predict the emergence of a novel state of matter with intertwined ferromagnetism, charge order and topology in fractionally filled moir'e superlattice bands. Remarkably, these quantum anomalous Hall (QAH) crystals exhibit a quantized integer Hall conductance that is different than expected from the filling and Chern number of the band. Microscopic calculations show that this phase is robustly favored at half-filling at larger twist angles of the twisted semiconductor bilayer $tMoTe_2$. Our numerical results demonstrate that the ground state of $tMoTe_2$ at half-filling is a QAH-crystal that exhibits spontaneous ferromagnetism, 2x2 CDW order, and quantized Hall conductance σ_{xy}=1 (in units of e^2/h). This state exemplifies a topological quantum phase possessing multiple symmetry-breaking order parameters. Furthermore, we also present numerical evidence for a QAH-crystal at filling number ν=3/4.

Future Plans

We will systematically search and discover novel correlated states such as non-Abelian QAH states, competing or intertwined charge-spin orders for complex systems on triangular or honeycomb lattices relevant for twisted TMD systems. Furthermore, we will study the quantum phase diagram for unconventional superconductivity for emerging material platforms based on moir`e superlattice systems with including more complex interactions such as spin-orbital coupling, topological band and spontaneous ferromagnetism.

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Electron Correlations, Bad-Metal Behavior and Unconventional Superconductivity

Principal Investigator: Qimiao Si, Rice University

Keywords: Correlated electrons, unconventional superconductivity, topology, flat bands, quantum phase transitions

Research Scope

This project aims to deepen our understanding of electron correlations, bad-metal behavior, and their connections with electronic orders and superconductivity in the Fe- and Ni-based superconductors (FeSCs and NiSCs) and correlated flat band systems. One direction is to address new frontiers on the orbital-selective correlations in the FeSCs, in particular the role of inter-orbital dehybridization, and the new NiSCs. Another direction is to investigate the role of multi-orbital correlation on bulk electronic topology, especially in the FeSCs. The third direction is to explore the effects of electron correlations in flat band systems. The projects will shed light on the multi-orbital correlations, superconductivity and topology in both bulk and low-dimensional systems.

Recent Progress (primarily 2023-2024)

Topic #1 -- Electron correlations in topological flat-band systems

1.A Kondo description of coupled flat and wide bands in frustrated lattices. Flat bands amplify correlation effects and are of extensive current interest. They provide a platform to explore both topology in correlated settings and correlation physics enriched by topology. In recent work [P1], we identify an orbital-selective Mott transition in any system of coupled topological flat and wide bands. This was made possible by the construction of exponentially localized and symmetry-preserving Wannier functions, which, in turn, leads to an effective Kondo-lattice description. Our findings show how quasiparticles are formed in such coupled topological flat-wide band systems and, equally important, how they are destroyed. Our work provides a conceptual framework for the understanding of the existing and emerging strange-metal properties in kagome metals and beyond.

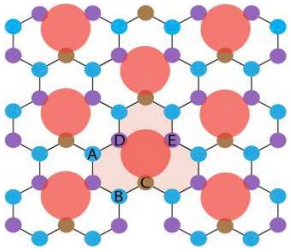

Figure: Compact molecular orbitals and Kondo lattice.

1.B Emergent flat bands and non-liquid behavior in frustrated lattices. Using the Kondo description, in a recent work [P2] we show, within a Hubbard model on a clover lattice, that electron correlations produce emergent flat bands that are pinned to the Fermi energy. This effect is important because noninteracting flat bands are generically located far away from the Fermi energy, which limits their capacity to partake in the low-energy

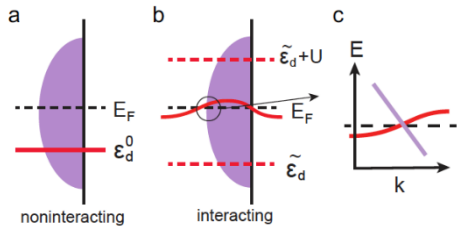

Figure: Emergent flat band.

physics. In our work, we show that the correlation effect cooperates with symmetry constraints to produce a topological Kondo semimetal. We also collaborate an experimental group to demonstrate such emergent flat band in CuV2S4 [P3], and demonstrate non-Fermi liquid behavior.

1.C Flat bands and Kondo effect in moire transition metal dichalcogenides. Using moiré structures represent a setting to realize flat bands. In recent work [P4[, we address the recently observed Kondo-driven heavy fermion state and its destruction in AB-stacked hetero-bilayer transition metal dichalcogenides, which can be controlled by the gate voltages. By studying an effective interacting Hamiltonian using the slave spin approach, we obtained a phase diagram with the total filling factor and the displacement field strength as the tunable parameters. In an extended range of the tunable displacement field, our numerical results show that the relative filling of the d orbital, which is associated with the highest moiré band from the MoTe2 layer, is enforced to be vd ≈ 1 by the interaction. This agrees with the experimental observation. We also argue that the observed high coherence temperature scale could be explained by the non-negligible bandwidth of the d orbital. Our results set the stage to address the amplified quantum fluctuations that the Kondo effect may produce in these structures and new regimes that the systems open up for Kondo-destruction quantum criticality.

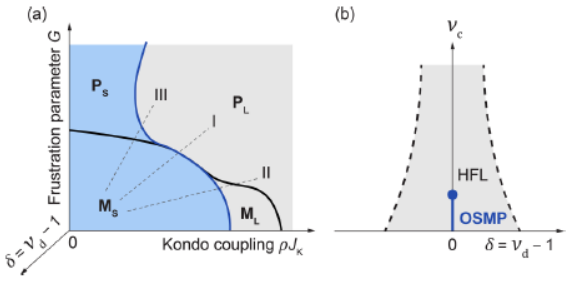

Figure: Kondo effect in AB-stacked hetero-bilayer TMDs.

Finally, we outlined a perspective on the generality of Kondo description for flat band systems in frustrated lattices and beyond [P5], and collaborated with experimentalists to topological signatures [P6].

Topic #2 -- Orbital-selective correlations and topology

Strong correlations lead to emergent excitations at low energies. When combined with symmetry constraints, they may produce topological electronic states near the Fermi energy. Within this general framework, we address [P7] the topological features in iron-based superconductors. We examine the effects of orbital-selective correlations on the band inversion in the iron chalcogenide FeSexTe1−x near its doping of optimal superconductivity, within a multiorbital model and using a U(1) slave spin theory. The orbital selectivity of the quasiparticle spectral weight, along with its counterpart of the energy level renormalization, leads to a band inversion and Dirac node formation

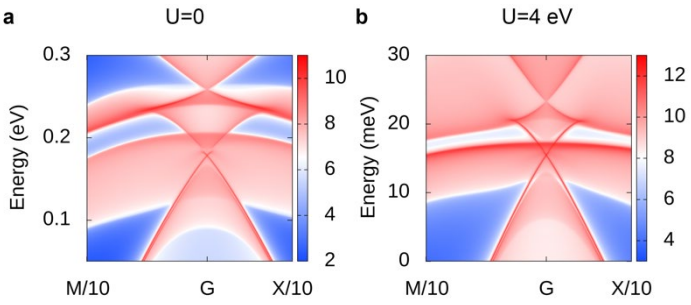

Figure: Topological surface states, showing the effect of orbital-selective correlations.

pinned to the immediate vicinity of the Fermi energy. Our work demonstrates both the naturalness and robustness of the topological properties in FeSexTe1−x, and uncovers a new setting in which strong correlations and space-group symmetry cooperate in generating strongly correlated electronic topology.

Topic #3 – Orbital-selective correlations and superconductivity

3.A Orbital-selective correlations and superconductivity in double-layer nickelates. The superconductivity community has been excited by the recent discovery of superconductivity in La3Ni2O7 under pressure. We discuss [P8] the basic ingredients of a model that captures its microscopic physics under pressure tuning. We anchor our description in terms of the spectroscopic evidence of strong correlations in this system. In a bilayer Hubbard model, we show the ground state of the model crosses over from a low-spin S=1/2 state to a high-spin S = 3/2 state. In the high-spin state, the two x2-y2 and the bonding z2 orbitals are all close to half-filling, which promotes a strong orbital selectivity in a broad crossover regime of the phase diagram pertinent to the system. Based on these results, we construct an effective multiorbital t-J model to describe the superconductivity of the system, and find the leading pairing channel to be an intraorbital spin singlet with a competition between the extended s-wave and dx2−y2 symmetries. Our results highlight the role of strong multiorbital correlation effects in driving the superconductivity of La3Ni2O7.

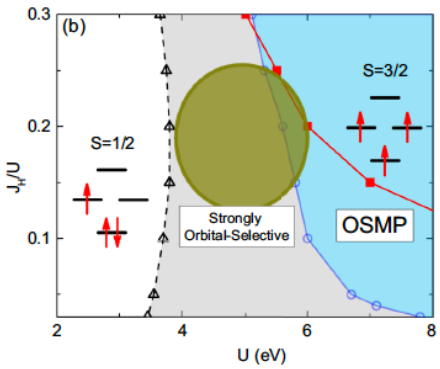

Figure: Calculated phase diagram for a multi-orbital model for the double-layer nickelates.

3.B New directions of orbital-selective correlations in FeSCs. provided a status report of the field of FeSCs in Physics Today [P9]. We make the case that fifteen years after the surprising discovery of superconductivity in iron-based materials, researchers are beginning to impart some of their newfound wisdom on a slew of emerging superconductors that display similar traits. Motivated by our recent work on the multi-orbital superconductivity in FeSCs, we have outlined a perspective about similar behavior in the heavy fermion superconductors [P10]. Finally, we studied [P11] a multiorbital Hubbard model and demonstrated a mechanism for a substantial change to the Fermi surface, namely, orbital selectivity of the energy-level renormalization cooperating with its counterpart in quasiparticle spectral weight. Our results set the stage to understand the origins and nature of both the unconventional superconductivity and likely electronic topology in this prototype iron pnictide and, more generally, reveal a remarkable

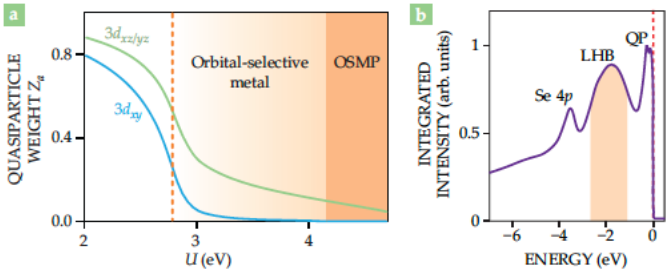

Figure: Status summary of orbital-selective correlations in FeSCs.

degree of universality out of the seemingly complex multiorbital building blocks across a broad range of strongly correlated superconductors.

Topic #4 – Bad metallicity and electronic order

We [P12] study spin excitation anisotropy in a model for FeSe, and collaborated with a RIXS group to show that the high-energy spin excitations are dispersive and underdamped. We understand the results from a local-moment perspective.Taking together the large energy scale far beyond the dxz/dyz orbital splitting, we suggest that the nematicity in FeSe is likely spin-driven. We also explore [P13] how the spin-excitation anisotropy, which directly reflects the existence of nematic spin correlations, provides clues about nematicity in the FeSCs. By analyzing the high-energy nematic spin correlations, we demonstrate the essential role of spin fluctuations in driving electronic nematicity.

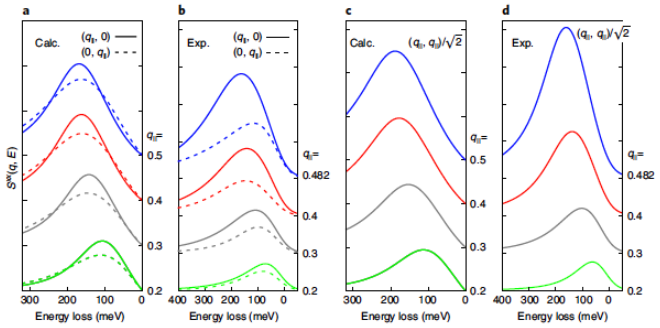

Figure: Calculated spin excitation spectra of the antiferroquadrupolar (AFQ) phase and their comparison with the fitting curve of the RIXS experiment.

Future Plans

Our plan for the coming year includes:

Quantum criticality in moire transition metal dichalcogenides. We plan to study how the effective Kondo physics can generate quantum criticality in AB-stacked transition metal dichalcogenides.

Flat bands induced quantum criticality in pyrochlore systems. Motivated by the aforementioned experimental result on non-Fermi liquid behavior in pyrochlore lattices, we plan to study the effect of flat bands on quantum criticality. A parallel study will be carried out in CsCr3Sb5, motivated by the exciting recent observation of quantum criticality and unconventional superconductivity in this system.

Electron correlations and superconductivity in the nickelates. This topic is receiving explosive theoretical interest. We are one of the earliest theory groups to address the topic. Our immediate focus is to characterize the orbital-selective correlation effects through a comparison with optical conductivity and ARPES measurements, and consider the implications for superconductivity.

Nematic correlations in FeSe. The unusual quantum-disordered magnetic ground state intertwined with superconductivity and electronic nematicity in FeSe has been a research focus in

iron-based superconductors. Recently, our experimental collaborators have mapped out the spin excitations of FeSe dewtinned with a uniaxial-strain device. The exciting new results provide a motivation to study a spin-interaction phase diagram, with the goal of elucidating the physics of FeSe in the overall phase diagram that contains the antiferroquadrupolar, Neel, and collinear AF ordering regimes. The work is expected to shed considerable new insights into the origin and entwining of the emergent orders in iron-based superconductors.

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Functionalization of 2D Materials Heterostructures for Solar Energy Conversion

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Keywords: 2D materials, water-splitting, heterostructures, materials discovery

Research Scope

The goal of this project is to use computational modeling, big data, and machine learning to discover robust and efficient 2D material-bulk heterostructures as tandem catalysts for solar-driven water-splitting to produce hydrogen. Solar-driven water-splitting processes provide clean, renewable, and sustainable form of hydrogen for generating electricity via fuel cells thus enabling solar energy to electricity conversion. An approach to push the limits of solar-to-hydrogen efficiency is to utilize record-breaking photoabsorbers to maximize the capture of sunlight then use an outstanding catalyst to drive the catalytic reaction at a superior reaction rate. However, the interfaces of bulk photoabsorbers and catalysts are poorly understood. In this project, we are expanding our understanding of heterostructures of record-breaking photoabsorbers, like silicon, gallium arsenide, and cadmium telluride, and two-dimensional (2D) materials which excel as catalysts due to high specific surface area and a wide-range of tunable properties. Through high-throughput *ab initio* simulations and physics-based machine learning (ML) modeling, the atomic-scale structure, electronic-structure, and interface physics of 2D-bulk material heterostructures are being studied and curated in an open-source database. The physics-based ML models and the computed data will be used for a data-driven discovery of water-splitting photocatalytic heterostructures that push the limits of catalytic reactivity and at the same time yield corrosion-resistant durable heterostructures with massive solar energy capturing capacity. In particular, we plan to study a) the role of an electric field at the 2D-bulk materials interface in enhancing the reactivity of the 2D catalysts, b) the corrosion-resistance of 2D materials that can form protective barriers to the inherently corrosive photoabsorbers and c) and the fundamentals of heterostructures' interface-physics that yields massive number of photogenerated electrons to facilitate the water-splitting reaction.

Recent Progress

The funding period for this grant started in Fall. Since then we have simulated the energetic stability and the interface-charge transfer for ~50 2D materials (Janus MXY-type) on ~20 elemental, cubic phase, and metallic bulk materials. We found that out of the >1000 heterostructures that are obtained with these 2D-bulk pairs, more than 500 are thermodynamically stable with adsorption energies less than zero. We launched an open-source 2D-bulk materials data site, the *ab initio* heterostructure database (aiHD),[1] which consists of these van der Waals (vdW) corrected density functional theory (DFT) simulations. This dataset was sufficient to obtain a ML model with high R2 score (0.93) and low MAE (30 meV/atom), Figure 1b. Furthermore, the ML models developed on the aiHD data showed that stability of 2D Janus-bulk heterostructure are dependent mostly on bulk material properties.[2] Figure 1c shows the top five features for predicting

the 2D-bulk binding energy from the ML model and the percent feature importance. The dominance of bulk material properties in the ML feature importance is surprising, however, it can

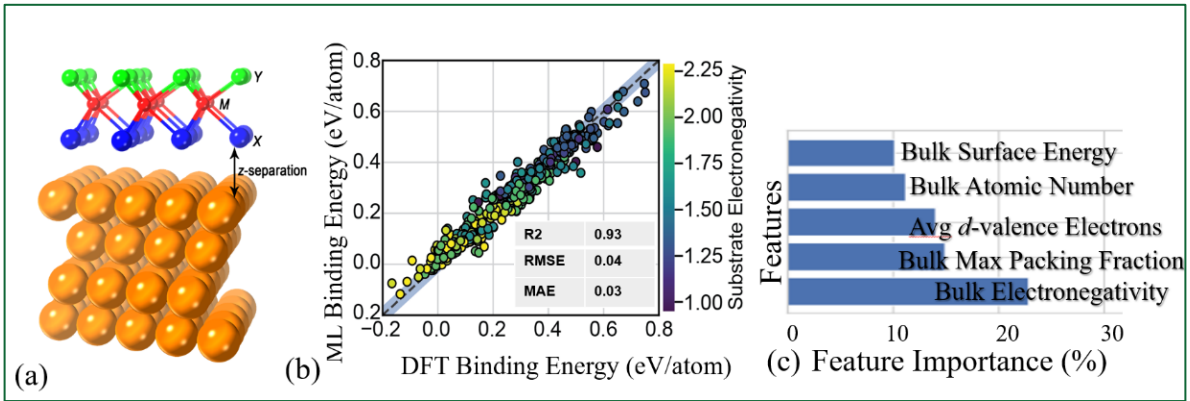

Figure 1 a) A 2D Janus material and bulk material heterostructure model. b) Comparison of DFT computed and machine learned binding energy for the Janus 2D-bulk heterostructure configurations. ML model has a root mean squared error of only 0.04 eV/atom. c) Top five most predictive features for the ML model. The most significant features are dependent on the bulk material properties.

be explained by comparing the energetics of the surfaces of bulk materials, which have high-energy broken bonds at their surface, with that of the 2D materials that have much weaker vdW bonds broken at their surface. Using the ML models trained on excited state theory, GW_0, and GW_0-BSE simulations we also discovered bulk photo absorbers that have low exciton binding energies, high integrated absorption in visible- or ultraviolet -light, and low anisotropy in absorption.[3] With ~6000 starting candidates we identified 159 visible-light and 203 ultraviolet-light photoabsorber materials utilizing the Materials Project database and our ML models. In summary, our work thus far has advanced the understanding of fundamental physics of the 2D-bulk heterostructures and led to the discovery of new photoabsorbers using first-principles simulations and physics-based ML models.

Future Plans

In the next year of the project, we will continue to study more 2D-bulk heterostructure pairs and the studies will include high-efficiency photoabsorbers as bulk materials. The large throughput of the simulations will be managed by our high-throughput workflow codes *hetero*2D[4] and *py*GWBSE[5]. In addition, we will be studying the charge transfer at the heterostructure interfaces, developing generalized descriptors for assessing charge-doping, dipole moment, and the impact of interface strain on these properties. Furthermore, we will employ continual ML modeling to train our existing ML models with the increasing first-principles computed data in the aiHD.

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Unconventional metals in strongly correlated systems

Senthil Todadri, Massachusetts Institute of Technology

Keywords: Non-fermi liquids, strange metals, superconductivity

Research Scope

The goals of the research are to develop a theoretical framework capable of describing the observed unconventional metallic behavior in quantum materials with strong inter-electronic interactions, and their relationship with more conventional phases.

Recent Progress

A paradigmatic and well understood, model of strong correlations is the Bose Hubbard model. As an example that illustrates some novel phenomena, I (with my student E. Lake, and M. Hermele (Boulder)) studied[1] a simple model of interacting bosons on a d-dimensional cubic lattice whose dynamics conserves both total boson number and `center-of-mass' of the bosons i.e their dipole moment). This model provides a simple framework in which several remarkable consequences of the constrained dynamics due to center-of-mass conservation can be explored. As a function of chemical potential and hopping strength, the model can be tuned between gapped Mott insulating phases and various types of gapless condensates. The condensed phase realized at large hopping strengths, which we dub a Bose-Einstein insulator, is particularly interesting: despite having a Bose condensate, it is insulating, and despite being an insulator, it is compressible. In a subsequent paper[2] (with Lake, MIT visitor Jung-Hoon Han, and Hyun Yong Lee), I studied in great detail the phase diagram of 1d boson models with dipole conservation with analytic arguments and numerical calculations.

Later (with student Ethan Lake), I studied[3] the fermionic Hubbard model with both particle number and center-of mass (dipole moment) conservation. This has the remarkable consequence that, in $d > 1$, the ground state at generic densities is naturally a non-fermi liquid. This understanding lead us to propose a route for a quantum simulation of a non-fermi liquid state in a system of cold atomic fermions in a tilted optical lattice. This system is described (in a `pre-thermal' regime) by a dipolar fermi Hubbard model, and hence realizes the non-fermi liquid state.

In other directions, I also studied a number of quantum critical strange metals[4], developed a phenomenological understanding of superconductivity in twisted bilayer graphene[5] , and described Mott and Wigner-Mott transitions in 2d semiconducting moire materials.

Future Plans

For the near future, I will continue my recently initiated study of quantum criticality associated with various kinds of exciton condensation in 2d materials, and explore experimental

signatures in transport. I will also continue exploring kinetically constrained non-fermi liquids and the possibility of quantum simulation of such states.

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Interaction and Transport Effects in Driven Magnetic and Topological Materials

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Department of Physics and Astronomy

University of Alabama

Program Scope

The scope of this program encompasses investigations of fundamental non-equilibrium effects arising from magnetic, transport and interaction-induced phenomena in periodically-driven low-dimensional materials and structures. Electronic band structures are known to become renormalized both on or off resonance under irradiation, with the renormalized bands occupied by a non-equilibrium distribution of photon-dressed electrons. One can tailor the parameters of driving field to design the system Hamiltonian to achieve the desired responses in the illuminated material. We have made considerable progress along the following directions: indirect exchange interaction between impurity spins, RKKY and Dzyaloshinsky-Moriya interactions between spin chains and ferromagnetic layers, and strong-field nonlinear magneto-optical effects.

Keywords: Non-Equilibrium, Light-Matter Interaction, Floquet, RKKY, Dzyaloshinsky-Moriya

Recent Progress

(1). Impurity Levels in Non-Equilibrium RKKY Interaction. We have investigated [1] the effects of off-resonant circularly polarized irradiation on the indirect exchange interaction between impurity spins coupled to graphene via a strong onsite potential. An important motivation was to extend the theory we had previously developed [2] to cases where the effects of the onsite potential have to be treated non-perturbatively. To this end, we have developed a theory for the RKKY interaction using a combination of Floquet Keldysh Green's functions and T-matrix formalism. Due to an optically induced band gap, undamped impurity levels can exist inside the gap which give rise to new behaviors of the exchange interaction. For impurities located at either the same (AA) or different (AB) sublattice sites, we found that irradiation extends the spatial range of antiferromagnetic behavior of the RKKY interaction in the presence of a strong onsite potential. The exchange energy is found to exhibit a strong resonance feature (Fig. 1) that originates from the crossing of the Fermi energy with one of the impurity levels. Remarkably, unlike in equilibrium where such a resonance feature only occurs in the AB case, we found that irradiation can induce a resonance feature in both AA and AB cases. When the Fermi level is located at the middle of the light-induced gap, the case without any potential scattering offers a limited tunability on the exchange energy only through its magnitude. However, for impurities with strong potential scattering, the exchange energy becomes more extensively tunable not only through its magnitude but also its sign. Thus, the presence of strong potential impurities enhances even more the

tunability of the exchange interaction by the driving field and may present an interesting prospect for light-controlled switching of antiferromagnetic/ferromagnetic exchange coupling.

(2). Optical Control of Dzyaloshinsky-Moriya Interaction. Generalizing our previous investigations on irradiated graphene, we have investigated the effects of light irradiation on the indirect exchange interaction in quantum materials with spin-orbit coupling. We studied two systems: (1) surface states of the strong topological insulator Bi_2Se_3 [3], and (2) bulk states of the Rashba semiconductor BiTeI [4]. In our first study, we have obtained the irradiation dependence of the Ising, Heisenberg, and Dzyaloshinsky-Moriya exchange couplings between isolated impurity spins and impurity spin chains (Fig. 2). Our results showed that not only can a stronger driving strength extend the RKKY oscillation periods, but it can also enhance the oscillation envelope of the Heisenberg exchange coupling while suppressing that of the Ising and Dzyaloshinsky-Moriya couplings.

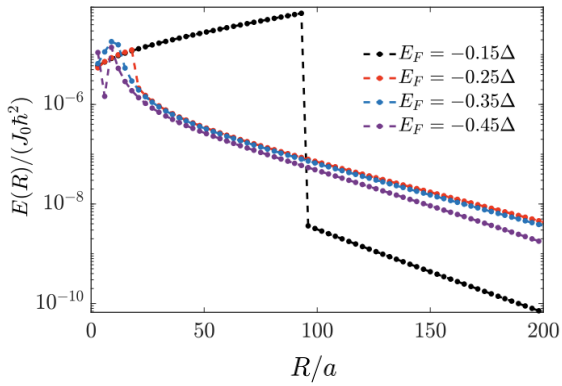

Fig.1: Time-averaged RKKY coupling versus distance between magnetic impurities in driven graphene ($A = 0.06$). The impurities are situated at sublattices

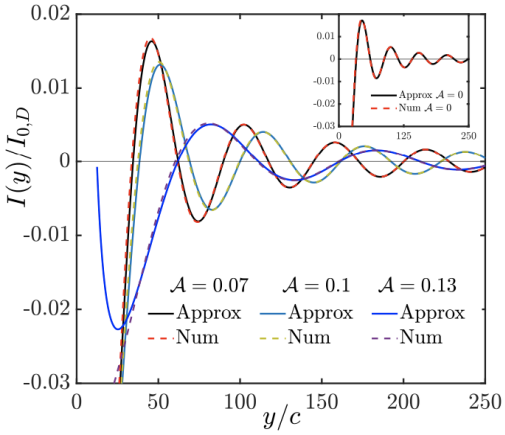

Fig.2: Dzyaloshinsky-Moriya interaction for different values of light-matter coupling as a function of the

In our second study, we have analyzed the light-driving effects on the interlayer indirect exchange coupling mediated by the bulk Rashba semiconductor BiTeI in a magnetic multilayer setup. The collinear magnetic exchange coupling mediated by the photon-dressed spin-orbit coupled electrons of BiTeI develops light-induced oscillation periods and displays new decay powers laws, both of which are enhanced with an increasing light-matter coupling. For magnetic layers with non-collinear magnetization, we find a non-collinear magnetic exchange coupling uniquely generated by light-driving of the multilayer (Fig. 3). These two works have provided useful insights and tools for understanding and realizing Floquet engineering of both collinear and non-collinear indirect exchange interactions in systems with spin-orbit coupling.

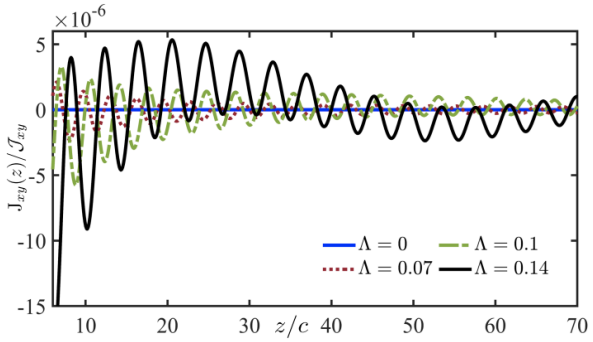

Fig.3: Non-collinear exchange interaction Jxy as a function of the BiTeI thickness z for different values of light-matter coupling. Note that this

(3). Floquet Engineering of Tilted and Gapped Dirac Band Structure. We have investigated [5] the photon-dressed band structure of the 2D van der Waals material 1T'-MoS_2. In contrast to the more well-known 2H form, the equilibrium dispersion of 1T'-MoS_2 is anisotropic exhibiting a tilted Dirac cone at each valley with a band gap. It displays many unique features, such as tunable anisotropy, band gap, spin- and valley-polarized states, and coexistence of different topological phases similarly to silicene. One important piece of physics we have elucidated in this work is the dependence of the photon-dressed bands on the polarization of the driving field. Because the equilibrium dispersion of 1T'-MoS_2 is already anisotropic, we found that illumination with linearly polarized light can substantially tune the degree of anisotropy of the dressed dispersion. On the other hand, illumination with circularly polarized light preserves the original degree of anisotropy and renormalizes the band gap. This work provides a detailed understanding on the effects of off-resonant light illumination on the band structure of van der Waals materials with an anisotropic tilted Dirac dispersion.

(4). Magnetic-Field Tuning of Second-Harmonic Generation. We have developed a theory for the second harmonic generation from the surface states of a strong topological insulator Bi_2Se_3 under an external perpendicular magnetic field. The approach we used in this investigation is perturbative and complementary to the approach used in our investigations (1) and (2) outlined above. Topological insulator surface states can generate second harmonics due to the reduced symmetry caused by trigonal warping. The second harmonic signal is found to be directly proportional to the trigonal warping parameter, and therefore measurement of the second-order conductivities could in principle offer a means to quantify the trigonal warping effect. We have also investigated the effects of Landau level quantization on the second-order conductivities for second harmonic generation. Our results have elucidated the dependences of the second-order conductivities on the light frequency and polarization, Fermi level, and magnetic field. In particular, our work has elucidated the single-photon and two-photon resonance processes between Landau levels that underlie the enhancement features in the second-order conductivities. This work is currently being prepared for publication.

Future Plans

Based on the progress we have obtained in understanding the optical driving effects on the indirect exchange interaction in metallic host materials, we plan to study the case of indirect exchange interaction in insulating host materials, known as the Bloembergen-Rowland (BR) interaction. Compared to the RKKY interaction in metallic systems, the BR interaction is short-ranged and does not undergo any oscillation. We will obtain the dependence of the time-averaged BR interaction on the driving field parameters such as the driving amplitude and frequency and elucidate the mechanism by which such an interaction can be coherently controlled. Our investigation will cover first the case of isolated impurities and then the case of a ferromagnetic layers sandwiching an insulating spacer. Another direction we will continue to investigate is the electron-electron interaction effects under periodic driving. We are currently making progress in understanding the interaction-induced transport properties in van der Waals heterostructures driven by a time-periodic laser field. Finally, we plan to continue investigating the nonlinear optical conductivities in various quantum materials under an external magnetic field to elucidate the interplay between Landau quantization and multiphoton excitation processes.

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Nonequilibrium thermodynamics in magnetic nanostructures

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Keywords: Spin superfluidity, nonequilibrium topological textures, quantum entanglement generation and dynamics

Program Scope

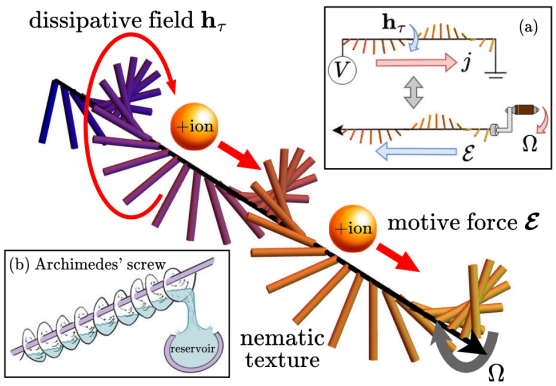
Over the last few years, this DOE award supported the work in my group that can be subdivided into four related areas: (i) *Topological transport phenomena,* (ii) *Spin impurities for qbits and sensing,* (iii) *Thermoelectric spin transport of electrons and nuclei,* and (iv) *Coherent pumping and torque-induced dynamics.* In recent years, the interplay of real- and momentum-space topologies of quasiparticles and collective order-parameter textures led to a resurgence in materials science, quantum-matter classifications, and novel transport and dynamic phenomena. We contributed to these developments within the first cluster of projects. The second thrust develops new solid-state strategies for designing and probing scalable quantum systems. To this end, we study Andreev bound states in magnetic/superconducting heterostructures, spin liquids, and continue exploring integration strategies of quantum color centers with spintronic systems. The latter effort is supplemented with our continued collaborations with experimentalists on nitrogen-vacancy sensing. Our traditional strength in various thermoelectric modalities of spin transport is leveraged in the third topic. Here, we further expand into topological materials, such as Weyl semimetals, and address also thermally-driven nuclear spin dynamics, which was observed experimentally. Finally, within the last topic, we are pursuing the subject that has remained mainstay since the inception of this project: Collective order-parameter dynamics and pumping, in the presence of various quasiparticle torques, dynamic condensation, and topological order-parameter textures. In the most recent developments, we are building bridges with quantum optics and liquid-crystal communities. The latter relying on superradiance and lasing perspectives and the former exploring nematic texture dynamics and ionic pumping in analogy to magnetic textures in solid state.

Recent Progress

Here, I will touch upon three highlights from the most recent work. The full publication list for the work supported by this Award over the last year is appended below.

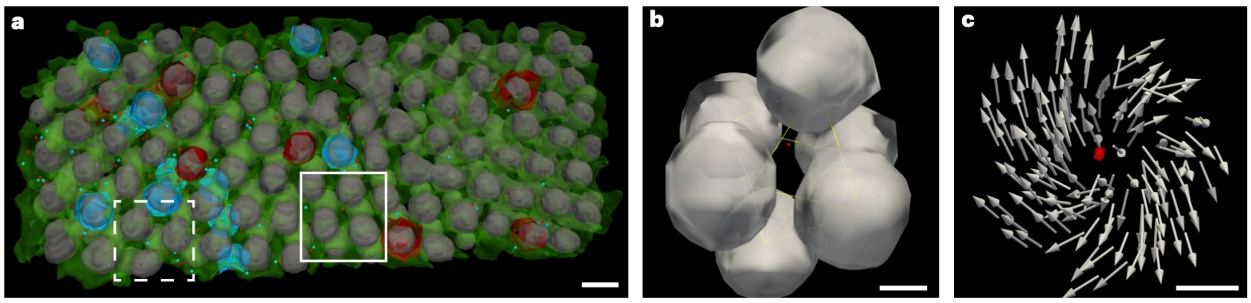
Ref. [1] constitutes an interdisciplinary effort. Here, adopting a spintronics-inspired approach, we studied the reciprocal coupling between ionic charge currents and nematic texture dynamics in a uniaxial nematic electrolyte. Assuming quenched fluid dynamics, we developed equations of motion analogously to spin torque and spin pumping. Based on the principle of least dissipation of energy, we derived the adiabatic ``nematic torque" exerted by ionic currents on the nematic

director field as well as the reciprocal motive force on ions due to the orientational dynamics of the director. We discussed several simple examples that illustrate the potential functionality of this coupling. Furthermore, using our phenomenological framework, we proposed a practical means to extract the coupling strength through impedance measurements on a nematic cell. Exploring further applications based on this physics could foster the development of nematronics - nematic iontronics, by drawing on the extensive knowledge and ideas developed within magnetism and spintronics over the past three decades. We hope that such venturing into other adjacent fields may benefit interdisciplinary cross-pollination and always keep our eyes open for new opportunities, on both quantum and classical fronts.



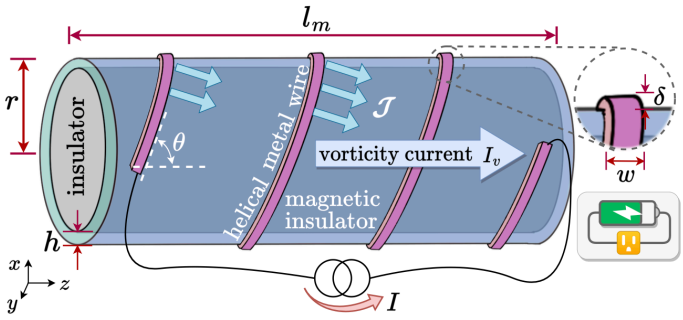
The nematic analog of the Archimedes screw. An ionic current (driven by a voltage bias) induces an effective field $\mathbf{h}_\tau$ on the nematic director field, causing the director field to rotate. Inversely, ions are pumped by the motive force $\mathcal{E}$, which is induced when the director is rotated at frequency Ω through external means. In panel (a), red arrows denote the external drives and blue arrows denote induced forces. Panel (b) depicts the actual Archimedean screw transporting water, in analogy with the nematic screw transporting charge.

Topological magnetic hedgehogs (or Bloch points) are three-dimensional (3D) non-local spin textures that are robust to thermal and quantum fluctuations. Until recently, it has been difficult to directly measure their 3D magnetization vector field and characterize their interactions at the nanoscale. Teaming up with our colleagues at an X-ray imaging lab at UCLA, we studied magnetic textures in a frustrated magnetic metamaterial in Ref. [5]. The magnetic medium consists of nickel infiltrating the interstitial space of a metalattice made of close-packed silica nanospheres. The resultant magnetic textures, rich in (anti)hedgehog features, were imaged with the state-of-the-art soft X-ray vector ptycho-tomography, and the distribution of the associated topological charges analyzed based on the ideas from our earlier theoretical work.\cite{zouPRL20} The spatial resolution of 10~nm is comparable to the magnetic exchange length of transition metals, enabling us to probe interactions of the topological charges. In addition to the Bloch points within the interstitial magnetic material, we also observed virtual (anti)hedgehogs hosted by magnetic voids in the metalattice. This work demonstrated that ferromagnetic metalattices could be used as a platform to create and investigate the interactions and dynamics of 3D topological charges. Furthermore, we expect that soft X-ray vector ptycho-tomography can be broadly applied to quantitatively image 3D vector fields in magnetic and anisotropic materials at the nanoscale, which will stimulate further theoretical investigations.



3D spatial distribution of 68 hedgehogs (red dots) and 70 anti-hedgehogs (blue dots) in the silica FCC metalattice, where the surfaces of the magnetic voids in red and blue blobs represent virtual topological charges. The solid square marks a region of interest shown in b. The location (b) and 3D spin textures (c) of a hedgehog within a tetrahedral site of the metalattice. Scale bars: a, 60 nm; b, 25 nm; c, 10 nm. The voxel size of the magnetization vector field is $5 \times 5 \times 5$ nm^3, which is set by the experimental resolution.

One of our key projects within the topological rubric concerns an energy-storage idea we have reported on in the previous award cycle. In the present work (Ref. [9]), we studied the transport of vorticity, which is manipulated to store magnetic winding energy in ring-like structures, on curved 2D magnetic membranes. We found that topological transport of vorticity can be controlled by geometrically reducing symmetries, which can enable processes that are not present in flat magnetic systems. To this end, we constructed a vorticity 3-current that obeys a continuity equation in curved space. This continuity equation is immune to local fluctuations of the magnetic texture as well as spatiotemporal fluctuations of the membrane. We then showed how electric current can manipulate vortex transport in geometrically nontrivial magnetic systems. As an illustrative example (shown in the figure below), we proposed a minimal setup that can realize an experimentally feasible energy-storage device. The thermodynamic efficiency of the device is formulated in terms of a figure of merit that reflects the strength of the electrical and topological flux cross coupling, analogously to the thermo-electric figure of merit known as *ZT*.



Schematic of a minimal setup for geometrically controlled vortex transport. A metallic wire is wrapped around a cylindrical magnetic insulator membrane as a helix. An applied electric current I induces vorticity flux $\mathcal{J}$ transverse to the wire, resulting in vorticity current I_v along z. The side panel indicates that this system realizes a battery (based on the free energy stored in the azimuthal magnetic winding).

Future Plans

In the future work, we plan to advance our efforts on nonequilibrium thermodynamics of complex spin systems, with diverse microscopic constituents spanning a broad range of energy and lengthscales. Of particular interest to us will be the interplay of topological texture flows and quantum correlations, arising in response to thermodynamic biases.

Specifically, bolstered by the recent advances in understanding and controlling the coupled spin and energy transfer in complex magnetic materials and structures, I plan to focus on the following three thrusts over the coming years:

• Non-Hermitian (classical and quantum) dynamics in magnetic heterostructures

• Scalable quantum dynamics via dissipative coupling out of equilibrium

• Spin-superfluidity inspired SQUID for electric-field sensing

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Time-dependent density-functional approaches for noncollinear spin dynamics

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Project Scope

The objective of this research is to develop new approaches, based on time-dependent density-functional theory (TDDFT), for spin-dependent nonequilibrium phenomena in quantum materials, and to implement and apply them in the linear and nonlinear regime. Magnetic systems out of equilibrium are of great scientific interest and offer many potential applications, such as spin waves in (anti)ferromagnetic and topological systems, light-driven demagnetization, control of single-spin qubits, or skyrmion dynamics. First-principles descriptions must account for noncollinear spins, many-body interactions, and spin-orbit coupling. Standard DFT approaches use exchange-correlation (xc) functionals obtained by "upgrading'" the usual LSDA and GGA for noncollinear spins. While this often works reasonably well, subtle xc magnetic torque effects are missed in this way, which affects the description of magnetization dynamics. To include these xc torques, new (TD)DFT approaches are needed. This project will develop new types of multipurpose, orbital-based xc functionals for the noncollinear case which can produce xc torques. These functionals will be implemented in INQ, which is a new code platform for large-scale DFT and TDDFT calculations. In addition, new types of correlation functionals for Hubbard-type lattice Hamiltonians will be developed. These TDDFT methods will be applied to describe noncollinear spin dynamics in situations where xc torque effects may be expected to be significant. *Magnonics.* Magnon dispersions in magnetic materials will be calculated using real-time TDDFT in bulk and heterostructures. The goal will be to obtain atomistically resolved magnonic boundary conditions at interfaces and to describe nonlinear phenomena such as magnon gating and switching, instabilities of parametrically driven magnons, and generation of higher harmonics. *Ultrafast magnetization dynamics.* Simulations of ultrafast demagnetization in Fe, Co, and Ni and in perovskite materials such as $NdNiO_3$ will be carried out, to better understand the experimentally observed rapid collapse of magnetic order following photoexcitation. Ultrafast magnetization dynamics will be analyzed in systems where ordered and spin-frustrated disordered magnetic phases are in close competition. The noncollinear TDDFT approaches proposed here will have fundamental and practical impact, leading to reliable and robust first-principles predictions of spin dynamics in a broad spectrum of materials. This could lead to novel applications in data storage and processing, encoding and transmitting information via spin waves, and the design of ultrafast spintronics devices.

Keywords:

Density-functional theory, noncollinear magnetism, magnonics, ultrafast magnetization dynamics, Hubbard model

Recent Progress

1. DFT for noncollinear magnetism. The standard approach for first-principles electronic structure calculations of noncollinear magnetic materials is based on a simple adaptation of DFT methods for collinear spins: exchange-correlation (xc) functionals such as the local spin density approximation (LSDA) or generalized gradient approximations (GGA) are evaluated using a locally fixed spin quantization axis. The resulting xc magnetic fields are always parallel to the local magnetization. However, this is not true in general: there should be local xc torques that influence the magnetic structure and dynamics [1]. The PI has developed a semilocal xc functional for noncollinear spin DFT based on short-range expansions of the spin-resolved exchange hole and the two-body density matrix [Publication #2]. The functional is U(1) and SU(2) gauge invariant and produces nonvanishing xc torques. These xc functionals have been implemented in the Octopus code, and applied to small chromium clusters, see Fig. 1. The exchange part performs favorably compared to the more expensive Slater or exact-exchange potentials; a delicate interplay between exchange and correlation torques is uncovered.

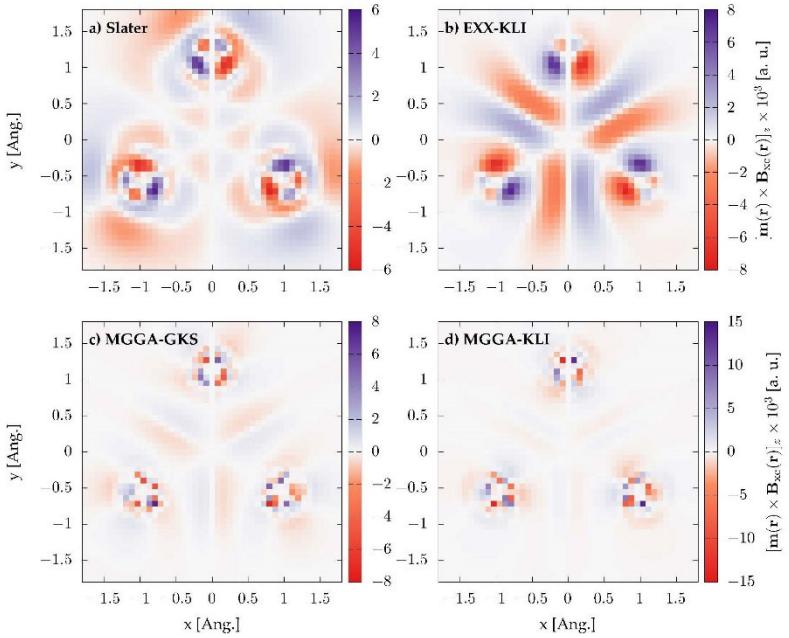

Fig.1. Local xc torques around the Cr atoms in a Cr3 cluster, computed with the Slater potential (top left), exact-exchange-DFT (top right), and with our new noncollinear meta-GGA (MGGA) xc functionals (bottom) [Publication #2].

2. Topological magnetic quasiparticles in nanomagnets. The ubiquitous domain wall kinetics under magnetic field or current application describes the dynamic properties in nanostructured magnets. However, when the geometrical size of a nanomagnetic system is constricted to the limiting domain wall length scale, the competing energetics between anisotropy, exchange and dipolar interactions can cause emergent kinetics due to quasiparticle relaxation, similar to bulk magnets of atomic origin. We have carried out a joint experimental and theoretical study to support this argument: constricted nanomagnets made of antiferromagnetic and paramagnetic Nd thin films with honeycomb motif reveal fast kinetic events at ps time scales due to the relaxation of chiral vortex loop shaped topological quasiparticles that persist to low temperature in the absence of any external stimuli. We found the kinetics in the Nd nanostructures to be quantitatively similar to that found in ferromagnetic (permalloy) structures of the same geometry. This suggests that a universal,

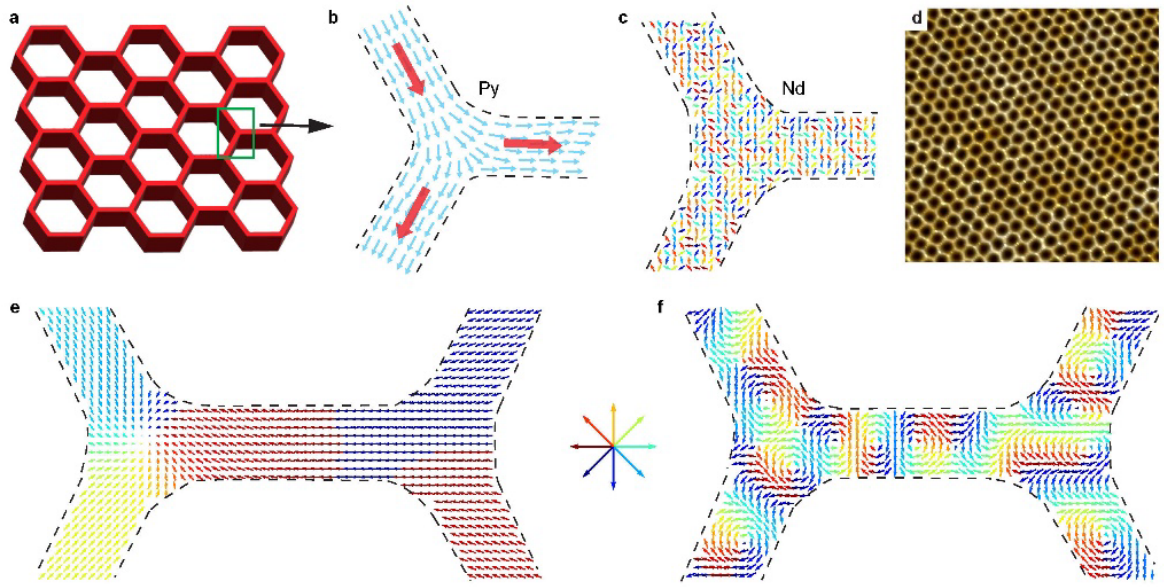

Fig.2. Quasiparticle-mediated dynamics in antiferromagnetic and ferromagnetic honeycomb lattices of constricted nanomagnetic elements, illustrating the kinetics of vortex-type topological defects [Publication #5].

topological quasiparticle mediated dynamical behavior can be prevalent in nanoscopic magnets, irrespective of the nature of the underlying magnetic material. The geometries and magnetization patterns are illustrated in Fig. 2. This research has been carried out in collaboration with the experimental group of Prof. Deepak Singh at the University of Missouri, who fabricated the samples and conducted neutron scattering measurements. The PI's postdoc, Dr. Daniel Hill, developed a theoretical model based on a Hamiltonian reflecting the sample geometry, exchange interactions, and dipolar interactions, which lead to skyrmion-like quasiparticles without the need to include spin-orbit effects (DMI). The thermal fluctuations and relaxations were then modeled using a stochastic approach, leading to an excellent agreement with experimental data. This work has been published in Nature Communications [Publication #3], and has been submitted to Nature Physics [Publication #5].

Future Plans

Ab-initio (TD)DFT for noncollinear spin dynamics. In collaboration with Dr. Nicolas Tancogne-Dejean from the Max-Planck-Institute of Structure and Dynamics of Matter, Hamburg, Germany, we have developed MGGA and self-interaction corrected xc functionals for noncollinear magnetism. We will implement and test these functionals in the INQ code [2], in collaboration with the group of developers at LLNL (Takashi Ogitsu, Xavier Andrade) and with the group of Prof. Andre Schleife at the University of Illinois. The work will be mainly done by the PI's postdoc, Dr. Daniel Hill. INQ is a GPU-based code for TDDFT, with the capability of simulating noncollinear magnetism dynamics. At the early test stages, we will simulate ultrafast

demagnetization of bulk ferromagnets, and test the influence of our noncollinear MGGA functionals on magnon dispersions.

Source-free xc functionals for spin waves. This is a project with the PI's graduate student Jenna Bologa. We are testing a recently proposed xc functional for magnetic materials [3] which subtracts the longitudinal component of the xc magnetic fields. While this seems to be mildly advantageous for ground-state magnetic properties, we find that there are some problems in the dynamical case: most notably, Larmor's theorem for spin waves is violated. We expect this project to be completed by mid-2024.

Correlation energy functional for the Hubbard model. The objective of this project is to construct correlation energy functionals for lattice models. In previous work under DOE support, the PI and his group have worked extensively on noncollinear spin dynamics for Hubbard clusters [4]. We found that dynamical correlations are significant, which motivates the search for designated correlation energy functionals [Publication #1]. To do this, we will work with a correlated wave function ansatz for the 2-body density matrix, similar to the Colle-Salvetti functional from quantum chemistry. An undergraduate student, Justin Shotton, has been involved in this project.

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Full counting statistics of charge in chaotic many-body quantum systems

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Keywords: Quantum transport, Fluctuations, Hydrodynamics, Quantum Circuits

Research Scope

The probabilistic nature of measurement in quantum mechanics endows transport in quantum systems with fluctuations. Uncovering the nature of these fluctuations has become a recent focus in quantum gas microscopy and fluorescence imaging experiments, where the experimental readout – 'snapshots' – give access to the full distribution of measurement outcomes. At the same time, the 'full counting statistics' of transport has been computed analytically in a number of models. However, so far these have been limited to non-interacting and integrable systems. This leaves important questions in generic, chaotic systems unanswered, one of which we highlight in this work: are fluctuations effectively classical or are additional ingredients required to include the quantum coherent effects on transport?

Just as in equilibrium statistical mechanics, universal behavior also emerges in the relaxation of a system out-of-equilibrium. This dynamical universality corresponds to the emergence of a hydrodynamic description, with the most famous examples being the diffusion equation and Kardar-Parisi-Zhang (KPZ) equations for which their respective universality classes are named. Our understanding of dynamical universality has so far almost exclusively focused on mean transport, encoded in the classical equations of hydrodynamics. The extent to which this universality can be extended to fluctuations is far less understood, but is a problem that can now be explored experimentally using the single-site resolution measurements of noisy intermediate scale quantum devices, and through the nonlinear response measured in pump-probe spectroscopy.

Recent Progress

In a recent work [2], the PI and collaborators addressed this question of the "quantumness" of fluctuations in many-body quantum systems by investigating charge transport in a paradigmatic example of chaotic quantum dynamics – random unitary circuits. In order to study the statistics of transport analytically, we restricted our attention to circuits with a conserved charge. Crucially, our approach relies on recent analytical tools developed in part by the PI and collaborators to study the properties of ensembles of random quantum circuits by mapping them to effective replicated statistical mechanics models. While those tools were originally developed with quantum information theoretic questions in mind [1,4], our work also shows that they can be applied to study fundamental quantum transport questions as well.

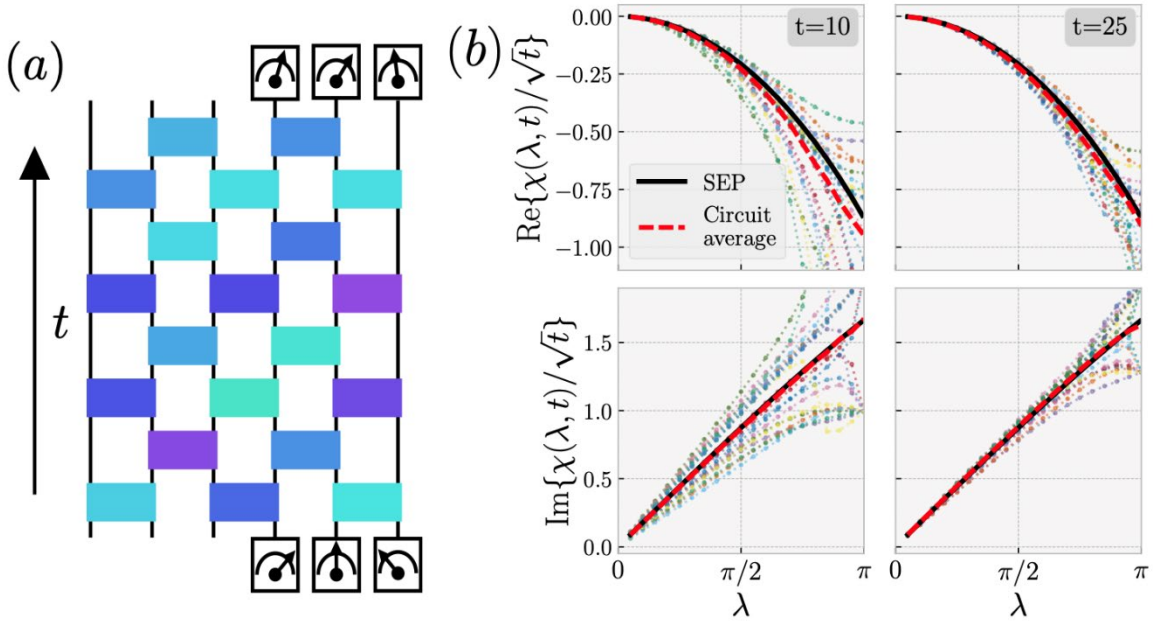

Figure 11: (a) The statistics of charge transfer across the central bond of a random unitary circuit with a U(1) conserved charge is measured using a two-time measurement protocol. The charge in the right half of the system is measured at times 0 and t. (b) The cumulant generating function χ(λ, t) of the transferred charge with a step initial step at times t = 10 and t = 25 for different circuit realizations (multi-colored) from matrix-product state simulations, the circuit averaged cumulant generating function with 35 samples (red dashed) and the late time analytical prediction from the effective statistical mechanics model (black solid). The cumulant generating functions is self-averaging at long times.

Leveraging a controllable limit (large on-site Hilbert space), we proposed an effective classical statistical model that captures the charge fluctuations. This effective theory allows, for the first time, the computation of full counting statistics in chaotic quantum models. We showed that transport and its fluctuations are identical to a simple stochastic classical in the long-time limit, and that quantum fluctuations enter as sub-leading finite time corrections. Our effective theory also goes beyond equilibrium, enabling us to see the emergence of classical fluctuations with far- from-equilibrium initial states. We verified these results with matrix product state numerics of the underlying quantum dynamics.

Future Plans

Our results thus establish that the current fluctuations of individual realizations of random quantum circuits are described by a simple fluctuating hydrodynamic equation: a diffusion equation with a non-linear multiplicative noise term. To fully establish the emergence of fluctuating hydrodynamics in many-body quantum systems, it would be interesting to consider ensembles of circuits with more general diffusion constants that would depend on density: there

as well we expect a similar mapping onto effective statistical mechanics models with a replica structure. The PI and collaborators are also investigating generalizations to systems with kinetic constraints (such as dipole moment conservation leading to fracton-like behavior).

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SciDAC: Real-time dynamics of driven correlated electrons in quantum materials

Vojtech Vlcek (UCSB - Lead PI), Khaled Ibrahim (LBNL), Gabriel Kotliar (Rutgers), Lin Lin (UC Berkeley), Daniel Neuhauser (UCLA), K. Birgitta Whaley (UC Berkeley), Chao Yang (LBNL), Dominika Zgid (U Michigan)

Keywords: Many-body theory, Non-equilibrium dynamics, First principles calculations, time-resolved spectroscopy, time evolution algorithms

Research Scope

The partnership aims to develop a theoretical framework and establish and implement an efficient methodology for practical simulations of ultra-fast non-equilibrium electron dynamics in quantum materials. The suite of tools aims to address the state-of-the-art experiments that capture the evolution of charge carriers at fs timescales. We combine new computational first-principles wavefunction and Green's function based tools, with novel numerical and AI algorithms leveraging the DOE HPC resources. The goal is to predict time-resolved photoemission and optical spectroscopy accurately.

Recent Progress

The team has focused on developing complementary new theoretical and algorithmic tools and multi-layered methodology. The major achievement is reducing the computational cost of methods used in each layer (while maintaining or increasing accuracy) and deciphering their fundamental limitations for correlated systems under strong non-equilibrium conditions.

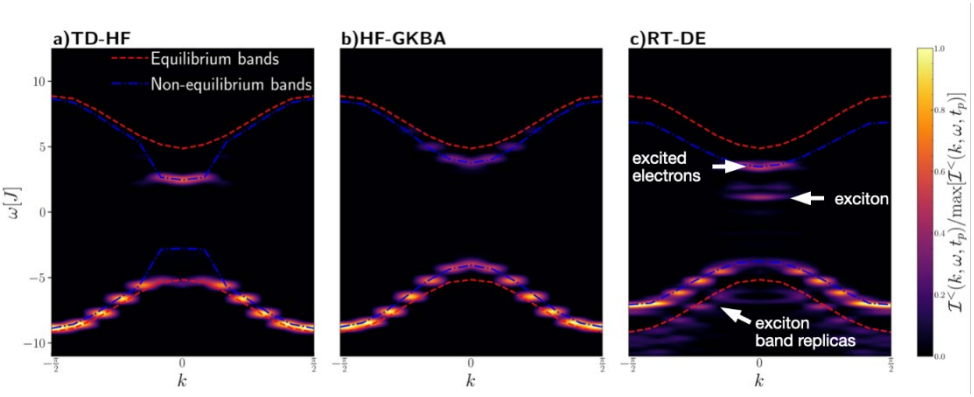
We rely on a combination of mean-field and perturbative many-body (MB) techniques to describe weakly correlated electronic states. The implementation is based on energy scale separations, and a mean-field treatment sufficiently approximates most electronic states. To this end, we have advanced real-time real-space methods by sparse stochastic fragmented resolution of the identity and stochastic fitting. These approaches were validated by simulations on realistic nanoscale systems and represent a critical jumping-off point for the MB techniques necessary for the quantitative treatment of quantum materials. [1]

The major effort resulted in developing new Green's function (GF) based methods capturing strong dynamical correlation induced by external driving. We have overcome conceptual and computational bottlenecks related to the time-non-local memory, which is subject to a rigorous bound and can be drastically truncated in many practical applications. This truncation does not have a negative impact on the accuracy and leads to orders of magnitude decrease in the computational cost. [2]

A major success is the development of the Real-Time Dyson Expansion (RT-DE) scheme, which addresses the lack of accurate predictions for time-dependent photoemission spectra (Fig.1). Typical computational techniques are subject to significant errors and practically do not improve

upon the mean-field results. The RT-DE approach corresponds to the standard many-body perturbation theory (MBPT) at equilibrium but enables reconstructing the non-equilibrium spectral properties with a cost similar to the mean-field approaches. The method captures both band renormalization and the emergence of new quantum phenomena: excitonic features and excitonic replicas, which agree with exact results. Most importantly, the RT-DE scheme can be readily combined with the existing real-time stochastic MBPT tools.

Fig 1: Theoretical time-resolved photoemission spectrum. RT-DE method captures strong dynamical correlations and new emergent phenomena (excitons and band replicas)



The team has further developed numerical methods, achieving large computational savings by employing efficient and accurate tools for extrapolating the time evolution of GFs based on dynamical mode decomposition extrapolation. [3] Simultaneously, we also developed a data-driven methodology employing a recurrent neural network (RNN – Fig 2), which decreases the cost of long-time evolution even when the memory effects are fully included. Unlike common data-driven approaches, RNN is employed to represent the integral operator, which is constructed as a GF functional. This eliminates the need to evaluate the costly integral in each time step. In essence, RNN allows reformulating the integrodifferential equation as an ODE that can be treated efficiently by existing solvers. We have demonstrated the effectiveness of the approach on model problems, including a nonlinear Dyson equation. [4]. Our results also demonstrate the method's wide applicability to generic systems. The extension of the RNN techniques to long-time simulations of realistic problems is underway.

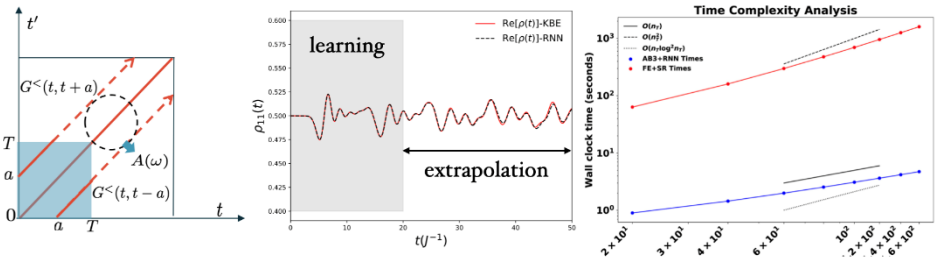


Fig 2: RNN approach extrapolates the Green's function, and leads to orders of magnitude computational savings without loss of accuracy.

The out-of-equilibrium dynamics of correlated electrons in materials require that the excitations associated with strong interactions are treated beyond MBPT. The team developed alternatives to existing electronic structure methods, which are as accurate as DMFT but significantly less

expensive. Our formalism is based on the Gutzwiller – Rotationally Invariant Slave Boson method, and we demonstrated its accuracy for models and realistic systems in [5]. The second critical aspect of the separation of weakly and strongly correlated regimes relies on identifying the conditions under which strong correlations are induced by external driving fields. Fig 3 clearly indicates such regimes (using the natural orbitals entropy) under which a system that is weakly correlated at equilibrium enters a strongly interacting regime under driving. For such conditions, standard perturbative wavefunction methods (such as coupled cluster – CC) fail.

The entire team has been involved in an extensive and unique comparison of distinct real-time non-equilibrium methods (Fig 3), determining the optimal setup of the multilayered methodology and the limits of applicability of individual approximations. Green's function techniques are numerically stable (albeit missing out on high-frequency response due to overdamping). The strongly correlated systems are, however, practically exactly described by selected/truncated configuration interaction techniques. Hence, the multilayered method employs a combination of RT-DE and TD-CI.

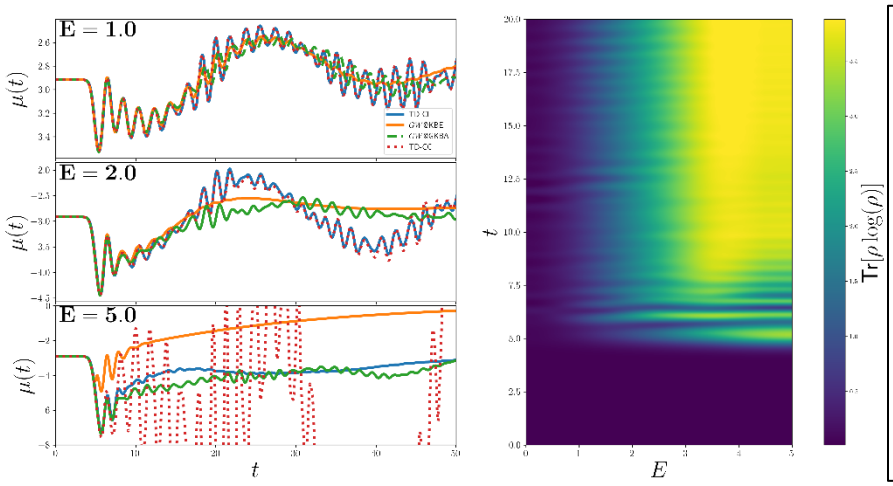


Fig 3: Non-equilibrium dynamics described by various methods for distinct pulse strength (E). TD-CC fails for high correlations but Green's function approaches are stable. Driven strong correlation shows high entropy of density matrix occupation numbers (yellow region in the right plot).

Future Plans

In the current and upcoming research efforts, the team combines the new developments (particularly RNN with RT-DE) and performs first-principles simulations of realistic semiconductors and quantum materials out of equilibrium. In particular, we investigate the scaling laws governing the renormalization of gaps and the emergence of new states. We also investigate the appearance of time-non-local quantum signatures for systems with embedded strong correlations (i.e., correlated quantum materials). To this end, we will employ the embedding of time-dependent selected CI and DMRG methods. The embedding will further employ the rotationally invariant slave boson formalism and include the development of hybridization fitting, which will enable applying GF techniques to open quantum systems modeled by the Lindblad equation. Finally, the newly produced computational tools are optimized for DOE HPC resources.

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QMC-HAMM: High accuracy multiscale models using quantum Monte Carlo

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Keywords: first principles calculations, quantum Monte Carlo, multiscale modeling, machine learning, effective Hamiltonian

Research Scope

The QMC-HAMM project seeks to improve the quality of multiscale models by coupling with high accuracy first principles calculations. QMC-HAMM leverages the quantum Monte Carlo class of methods, which directly treat electron correlation. These methods represent a significant improvement over the state of the art density functional theory calculations because they treat electron correlations explicitly. Critical to this goal is not just to improve the input data for models learned from ab initio simulation, but also to evaluate uncertainties in the models.

Recent Progress

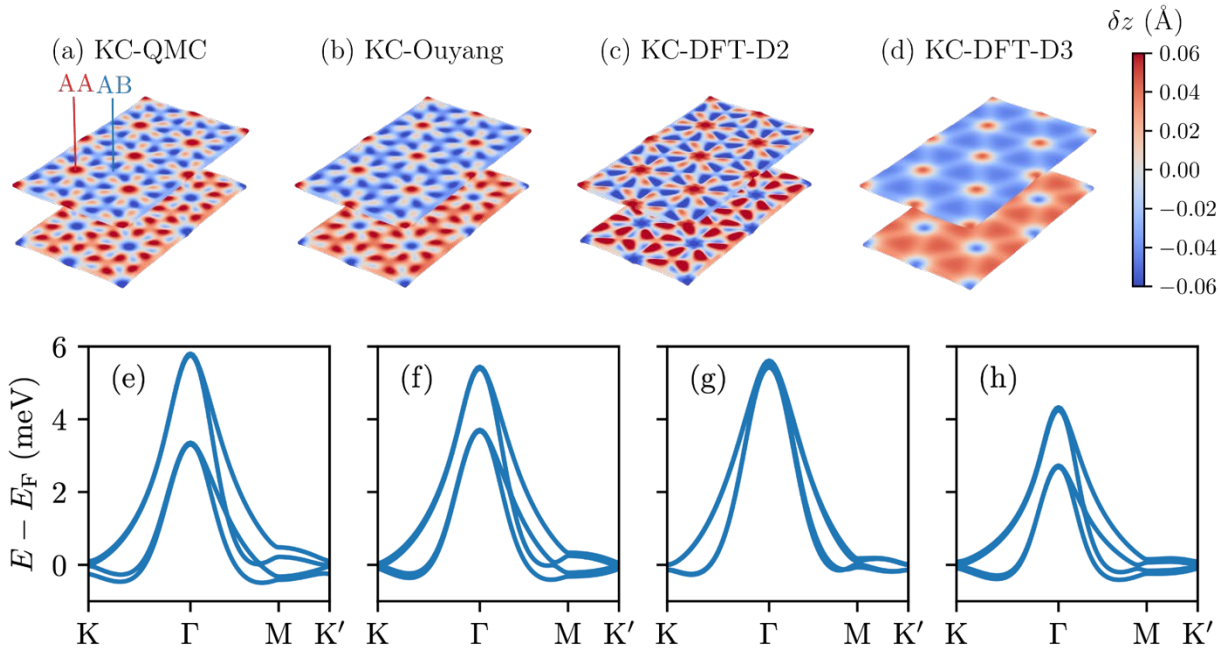

Figure 1: (top) Dependence of corrugation on the potential used to simulate the atomic motion. (bottom) The one-body band structures for the corresponding corrugation models. Note that the bandwidth can change by 50% or more depending on the accuracy of the van der Waals interaction.

Our collaboration focuses on four goals: 1) Improving models of materials by linking them to high accuracy first principles calculation, with error assessments 2) Methods to make quantum Monte Carlo simulations more useful for deriving larger length scale models 3) Application of the above

two ideas to understanding the structure and electronic behavior of layered graphene and other 2D materials, and 4) Application of the above two ideas to understanding hydrogen at high pressure.

Graphene Twisted bilayer graphene, and other stackings of graphene, have been observed to exhibit several unusual electronic phases, some of which appear to be indicative of strong electronic interactions. The minimum number of atoms that can be used to model this system number in the tens of thousands, out of reach for standard first principles methods, so models are used. Over the past few years, it has become clear that corrugations in the layers due to the Moire' patterns are critical in determining the electronic properties of the system. To model these corrugations accurately, an accurate treatment of van der Waals interactions is required, which is an electron correlation effect.

In Ref 1, we used quantum Monte Carlo methods, implemented in the TCMP-supported QMCPACK package, to evaluate the van der Waals interactions in graphene layers as a function of the relative coordinates of the layers. We then used this data to fit an atomic potential that included the very high accuracy interactions. Both the data and atomic potential are available from our collaboration.

In Ref 2, we used feature selection techniques from machine learning to learn an electronic tight-binding model from first principles calculations. This model treats the changes in electronic behavior on stretching and bending the graphene much more accurately than previous models, as we show in the paper. These modes are critical in understanding the effects of corrugation on the electronic behavior of layered graphene.

In Fig 1, we show the equilibrium structure of graphene at 0.99 degrees, evaluated with the new potential and the previous state of the art. Using our electronic model from above, we evaluated the sensitivity of the electronic structure to the corrugation induced by our more accurate model and found that the accuracy of the corrugation is critical to obtaining accurate band structures in this system. These differences are critical to understanding this system; for example, many pictures of this system rely on particle-hole symmetry, in which there is a symmetry between adding and removing an electron. Our results indicate that there is very little particle-hole symmetry in this system. We have also found (Ref 3) several unusual new corrugation patterns at low energy in bilayer graphene. As mentioned above, all the models and data supporting this work are available through our website and are being made available on the Materials Data Facility.

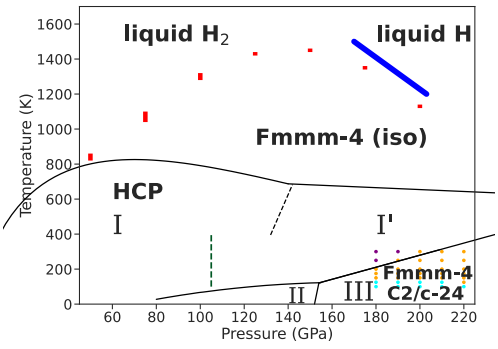

Figure 2: New phase diagram of hydrogen.

New phases of hydrogen. The primary interest of this application area is the determination of the phase diagram at high pressure over a large range of temperatures. In Ref 4, we used quantum

Monte Carlo simulations to derive an atomistic potential accurate across multiple phases of hydrogen. These simulations resulted in the discovery of a new potential phase in hydrogen at high pressure and temperature, as shown in Fig 2.

As part of the hydrogen project, we have published the QMC data on our website and in the Materials Data Facility. This data will be helpful in developing models of both electrons in hydrogen and in developing atomic-level models.

Methods development and improvement. While computation of coarse-grained atomistic potentials from high accuracy first principles quantum data is well-known, the same is not true for the computation of interacting electronic models. In Ref 5, we take a step towards achieving this goal for a test system, vanadocene, which emulates a correlated defect in a material. Such defects are responsible for the color of many gemstones, as well as a potential platform for quantum information processing. However, understanding the many-electron states of these defects remains challenging. We performed a cross-method benchmark of effective models from this system, using quantum Monte Carlo and quantum chemistry methods as reference, and learned guidelines for electronic model creation.

Future Plans

Graphene: We are working to incorporate a learned model for electronic interactions into a model. This new model will thus include a complete description of coarse-grained electronic interactions, one-body terms, and lattice motion. We plan to use this model to investigate electron-phonon effects and correlated electron phases in this system, using a very accurate reference.

Hydrogen: We are using hydrogen as a testbed for optimal learning algorithms and uncertainty quantification of models derived from Monte Carlo data. We are also investigating the capability of machine-learning models to describe solid-liquid phase transition accurately.

Methods development: We plan to extend our systematic approach to electronic models to a number of defects relevant to quantum information in collaboration with the MiCCom collaboration.

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Disorder and Interaction in Correlated Electron Materials

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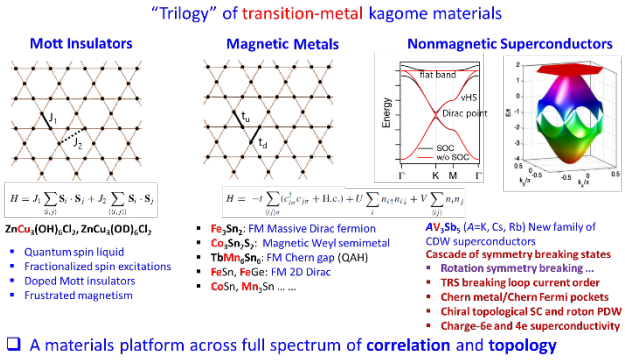
Project Scope

The program investigates correlated and topological electronic states and their low energy excitations in transition metal oxides, pnictides, chalcogenides, shandites, and antimonides. These include Cu-based, Fe-based, and Ni-based high-T_c superconductors, kagome magnets and the newly emerged V-based kagome metals and superconductors. The program maintains close collaborations with many experimental groups. These collaborative efforts have led to impactful discoveries and advanced the theoretical understanding of the collective properties of quantum materials including the role of electron correlation and its interplay with lattice geometry and electronic topology. They have in turn inspired more experimental discoveries. Our recent research activities have focused on studying the topological states and excitations in Fe-based superconductors and the rapidly developing field of kagome metals and superconductors.

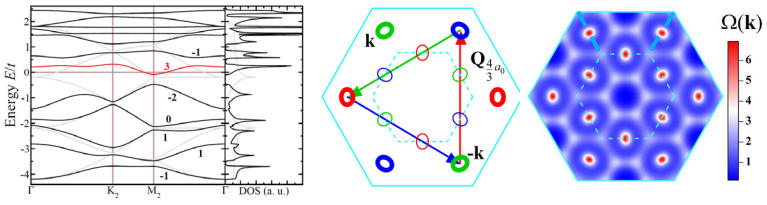
Recent Progress

Trilogy of kagome materials. We describe here our recent work on vanadium-based kagome metals AV_3Sb_5 (A=K, Rb, Cs). These compounds are *nonmagnetic* with the Fermi level close to the van Hove (vH) singularities and undergo a charge density wave (CDW) transition. Together with kagome insulators and magnetic metals, they form a trilogy of transition-metal kagome materials. The field has leapt forward after superconductivity was discovered in 2020. Since then, the field has attracted enormous attention with more than 600 papers published and posted on the arXiv to date. A plethora of thought-provoking correlated and topological quantum phenomena amid a cascade of symmetry breaking transitions have been discovered, straddling several frontiers of quantum materials. At the center of the revelation is the evidence for spontaneous time-reversal symmetry (TRS) breaking in the CDW state, which led to our original proposal of persistent orbital loop current (LC) order in these nonmagnetic kagome metals [R1]. The superconducting (SC) state is also highly intriguing and extraordinary. It coexists with the CDW order and breaks TRS, exhibiting novel pair density wave order and pseudogap phases. Evidence for charge-6e flux quantization has been observed in thin films ring devices, suggesting unprecedented higher-charge superconductivity. We have contributed to some of these original discoveries [R1-R5]. The physical origin and the broad implications of these remarkable observations have attracted great interests as the importance of the underlying physics goes beyond the field of kagome materials.



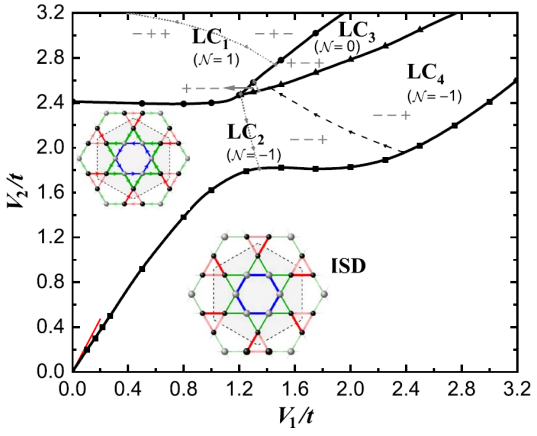
Loop-current Chern metal and Chern Fermi pockets. Recently, we proposed a plausible theoretical mechanism that the most essential part of the phenomenology can be captured by a bond CDW with LC order [P1]. The TRS breaking topological CDW gaps out the vH singularity and creates quasiparticle bands

with nonzero topological Chern numbers. Close to the vH filling, the Fermi level crosses a partially filled Chern band and gives rise to a LC Chern metal with small Fermi surface pockets we termed as Chern Fermi pockets (CFPs). The CFPs carry concentrated Berry curvature of the Chern band and orbital magnetic moment, and give rise to intrinsic anomalous Hall effect observed in the CDW state. They are consistent with the low-frequency orbits carrying large Berry phases detected by quantum oscillations. More recently, Fermi pockets at the predicted locations have been observed by combined ARPES and STM. Polarized neutron scattering and circular dichroism ARPES are underway to determine whether they are consistent with the predicted CFPs.



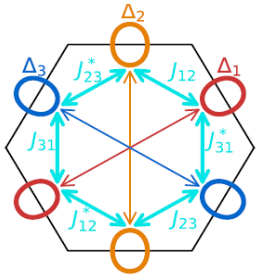
Due to the Fermi surface reconstruction by the $2a_0 \times 2a_0$ CDW, the CFPs are connected by ¼ and ¾ of the reciprocal lattice vectors, and raise the possibility for additional density wave formation with $4a_0$ or $\frac{4}{3}a_0$ spatial modulations. Nonzero center of mass momentum pairing over the CFPs can indeed describe the novel 3Q PDW with $\frac{4}{3}a_0 \times \frac{4}{3}a_0$ spatial modulations observed in the SC and pseudogap phases of CsV_3Sb_5 [R2]. We find that a plethora of intertwined and vestigial correlated topological states emerges, including a chiral LC pseudogap phase and charge-4e and charge-6e superconductivity in staged melting of the roton PDW.

Model realization of LC order. An outstanding question is whether and how the LC order can be produced in concrete theoretical models with electronic correlation on the kagome lattice. To this end, we studied the minimal single-orbital model under extended Coulomb interactions. An especially important and unique property of the kagome lattice is the sublattice polarization of the p-type vH singularity at the three M points, i.e. the electronic states at one M point reside exclusively on one of the sublattices. This is responsible for the bond ordered CDW states at wave vectors connecting different M points, and makes the intersite (inter-sublattice) Coulomb interactions V more relevant than the onsite-U. However, previous model studies could not produce LC order. Recently, we made concrete theoretical progress [P2]. We calculated the susceptibility of the real and imaginary bond order at vH filling. We found that for the nearest neighbor bond, the susceptibility of the real part of the bond diverges faster than that of the imaginary part, leading to an instability toward a real bond CDW. Remarkably, for the next nearest neighbor bond, it is the imaginary bond susceptibility that diverges faster than the real bond, which gives rise to an instability toward a complex bond CDW with LC order. A self-consistent mean field theory of the t-V_1-V_2 model is studied, where V_1 and V_2 are the nearest and next nearest neighbor Coulomb repulsion. The phase diagram reveals that the region dominated by V_1 is occupied by the real bond CDW order with the inverse of Star-of-David (ISD) configuration. In contrast, a large part of the phase diagram with significant V_2 is indeed occupied by various LC orders both at and



close to the vH filling. These findings provide a concrete model realization of the LC Chern metal in the normal state of the kagome superconductors.

Extraordinary superconductivity from LC Chern metal. We then made progress on the extraordinary SC state that emerges from the LC Chern metal normal state. Understanding the SC state of a partially filled Chern band is a central issue of correlation and topology. Physically, the LC in the Chern metal must turn into loop supercurrent circulating around vortices and anitvortices. We develop a concrete theoretical model description of the novel TRS breaking SC state when the quasiparticles residing on the CFPs undergo a pairing instability. We find that pairing over the CFPs is described by a three-component SC order. The components are coupled by complex Josephson couplings due to the circulating LC. Remarkably, the phase of the Josephson coupling contains dominant contributions from the Berry curvature of the CFPs, in an intriguing realization of the quantum geometry for the Cooper pairs. The leading SC instability is thus driven toward an *intrinsic* chiral topological superconductor where the relative phases of the three SC components are locked at 120-degrees. This exotic superconductor carries persistent loop-supercurrents circulating around an emergent vortex-antivortex (V-AV) lattice. In contrast to a uniform pairing state coexisting with charge density modulations, the Cooper pair wave functions exhibit V-AV roton PDW modulations with the reciprocal lattice momenta. Fully self-consistent meanfield calculations reveal remarkable spatial sublattice configurations of the orientation ordered superconductor and an anisotropic gap around the CFPs. We termed this a hexatic superconductor in analogy to hexatic liquid crystals. We argue this hexatic chiral SC state is frustrated on the kagome lattice. Strong relative phase and V-AV fluctuations suppress the true transition temperature to be well below the meanfield transition, resulting in an extended region of SC fluctuations [P3] observed experimentally. Intriguingly, a charge-6e bound state formed by the product of three 120-degree oriented Cooper pairs is free of the chiral phases, and thus decouples and is unaffected by the internal phase fluctuations. As a result, a phase coherent state of charge-6e states emerges over the extended fluctuation region upon melting of the charge-2e hexatic chiral superconductor, leading to fractionalized hc/6e flux quantization and charge-6e superconductivity observed in ring structures of kagome superconductor CsV_3Sb_5 thin films [R5]. The significance of these findings goes well beyond kagome superconductors.



Future Plans

Our research will continue to focus on the correlated and topological quantum states in kagome metals and superconductors, especially the interplay between rotation and TRS breaking. We plan to develop theoretical descriptions of our recent experimental discovery of the nonzero piezo-magnetic coupling that unambiguously revealed the TRS breaking and the important role of the lattice strains [P4]. Studying the multiorbital and three-dimensionality of the electronic structure provided insights and motivates us to explore the design and construction of multiorbital flat bands in lattices made of corner-sharing motifs such as the kagome and pyrochlore lattices, a direction to extend the horizon of electron correlation, topology, and geometry for extraordinary phases of matter in quantum materials.

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P9. Meng Li, Geng Li, Lu Cao, Xingtai Zhou, Xiancheng Wang, Changqing Jin, Ching-Kai Chiu, Stephen J. Pennycook, Ziqiang Wang & Hong-Jun Gao, "Ordered and tunable Majorana-zero-mode lattice in naturally strained LiFeAs", **Nature** 606, 890 (2022). https://doi.org/10.1038/s41586-022-04744-8

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Entanglement Witnessed by Spectroscopy and Multi-Point Correlations

Yao Wang, Emory University

Keywords: quantum simulations, spectroscopy, entanglement witness, multi-point correlations

Research Scope

Quantum materials are at the forefront of both scientific research and technological development. One of the defining features of these materials is that their electrons exhibit collective behavior underpinned by quantum entanglement, which sets them apart from classical band theory. The quest to understand, control, and design quantum materials for specific functionalities, such as superconductors and batteries, stands as a crucial research domain in the field of energy science. Achieving these groundbreaking applications requires precise and efficient characterization of the collective properties of electrons using advanced spectroscopy techniques. Nevertheless, due to the presence of quantum entanglement in these materials, conventional theoretical predictions using band theory and density functional theory face limitations. To accurately simulate quantum many-body systems, one must resort to computational approaches that scale exponentially, underscoring the imperative need for quantum computing or simulation techniques.

Our research employs quantum simulation techniques to reconcile microscopic quantum theories with macroscopic solid-state experiments. Leveraging quantum few-body systems as a platform, we elucidate the high-order correlations indicative of quantum entanglement that transcend classical interpretations. These correlations are then associated with specific spectral fingerprints. This approach enables the reconstruction of solid-state spectroscopies from analog quantum computers based on ultracold atoms or quantum dots, which will be accessible in the near future. Concurrently, this methodology facilitates the derivation of quantum information metrics from solid-state spectroscopies, which are accessible with existing materials and beamline facilities. Through collaborations with experimentalists, our methodology also guides the characterization and design of quantum materials.

The research scope is not restricted to the equilibrium properties of quantum systems and quantum materials. By extending the methodology to nonequilibrium many-body states, our research also aims to elucidate the control of quantum properties using ultrafast laser pulses, leveraging pump-probe spectroscopy techniques. Given the widespread lab accessibility in both cold-atom quantum simulators and solid-state materials, the ultrafast laser stands out as a promising control approach due to its rich degrees of freedom. Our studies based on quantum many-body simulations enable deciphering quantum information properties from the instantaneous status of materials.

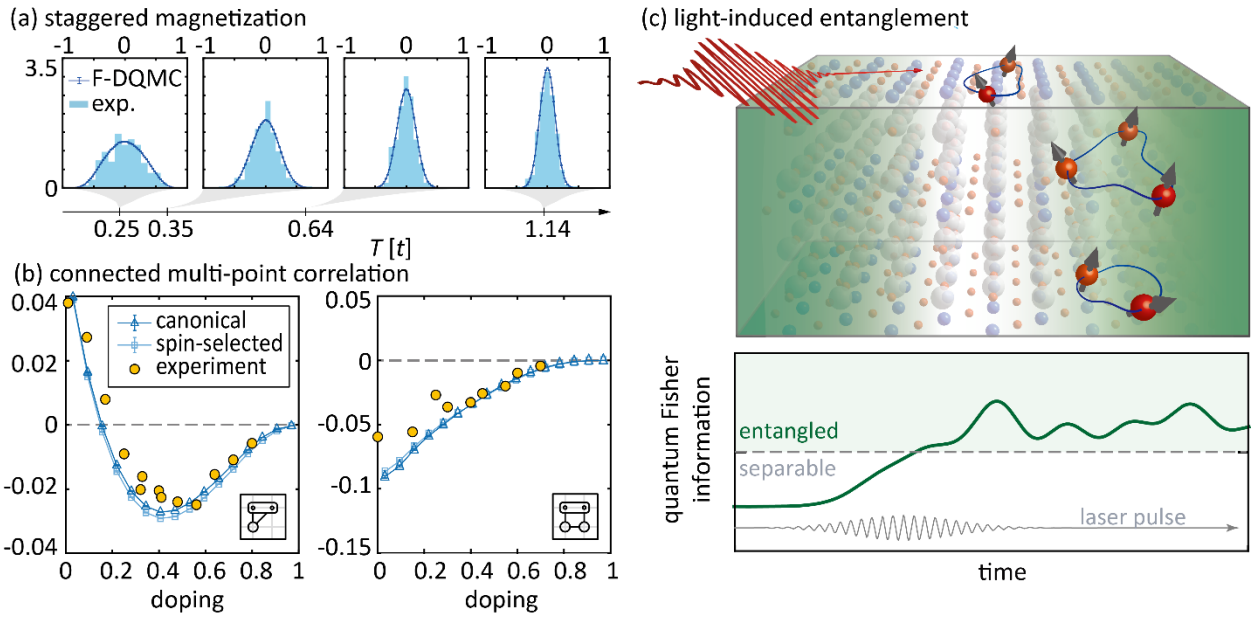

Figure. (a) Staggered magnetization in a finite-temperature Hubbard model, simulated using the Fock-state samples obtained by our quantum Monte Carlo method (solid line) and measured by ultracold atom experiments. (b) The simulated three-point (left) and four-point (right) correlations for canonical ensemble and spin-selected ensembles across varying doping levels, compared against the experimental data (yellow circles). (c) Light-induced spin quantum Fisher information in a strongly correlated system, described by the single-band extended Hubbard model. The gray dashed line indicates the boundary between witnessed separable and entangled state.

Recent Progress

The recent progress of this project can be bifurcated into two significant two aspects. Firstly, we developed a Fock-state quantum Monte Carlo algorithm that efficiently samples the Fock-state configurations of a many-body wavefunction. This algorithm allows for a faithful emulation of the quantum gas microscope in ultracold-atom quantum simulators.[1] Leveraging the sampled snapshots from this emulator, we unveiled multi-point correlations that align quantitatively with experimental results from quantum simulators [see panel (a) in the Figure]. Notably, the emergence of intensitive spin-spin-hole correlations in a doped Hubbard model signals the spin polaron's formation, whereas the four-point spin-spin-hole-hole correlation highlights intricate entangled electronic wavefunction across a wide range of doping [see panel (b) in the Figure].

The second aspect of progress lies in utilizing solid-state spectroscopies as a proxy for witnessing entanglement. Through a self-consistent approach, we demonstrated that instantaneous quantum Fisher information of time-dependent many-body wavefunctions can be quantified by a sequence of time-resolved x-ray spectroscopies.[2] This method facilitates the elucidation of microscopic quantum information in a laser-engineered nonequilibrium state of a macroscopic material via pump-probe x-ray scattering spectroscopy. Using a one-dimensional correlated material as an example, we showcased the potential for entanglement enhancement via ultrafast laser pulses [see

panel (c) of the Figure]. Then, by combining this approach with the multi-point correlations in quantum simulators, we further extended entanglement witnessing to basis-independent fermionic correlations. Our work reveals that the cumulant reduced density matrix, acting as an entanglement witness for indistinguishable fermions, can be inferred from the nonlinearity observed in x-ray scattering experiments.

Future Plans

We aim to establish comprehensive quantitative links between quantum simulators and solid-state spectroscopic analyses. The future plan extends from quantifying quantum properties across both domains to reconstructing spectral features using quantum simulations. Our immediate objective involves elucidating the dynamical spin spectrum, akin to inelastic neutron scattering outcomes, by harnessing multi-point spin correlations derived from sampled Fock-state configurations. Further, the X-ray spectrum and nonlinear spectral features necessitate a deeper investigation into time-domain dynamics. Moreover, we will leverage the high-throughput sampling of electronic Fock states, enabled by our innovative Fock-state quantum Monte Carlo method. Our methodologies will be tested and verified using classical computers and will apply to analog quantum computers in the near future based on ultracold atoms.

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DECODE: Data-driven Exascale Control of Optically Driven Excitations in Chemical and Material Systems

PI: Bryan M. Wong (University of California, Riverside), Co-PIs: Christian R. Shelton (University of California, Riverside), Zizhong Chen (University of California, Riverside), Khaled Z. Ibrahim (Lawrence Berkeley National Laboratory), Mauro Del Ben (Lawrence Berkeley National Laboratory)

Program Scope

This collaborative project will harness massively parallelized computation and advanced machine-learning approaches to design tailored optical excitations for controlling electron-driven dynamics in chemical/material systems. **Figure 1** summarizes the overall scope and inter-connected approaches used in this project for designing tailored, optically-driven excitations to control and manipulate the long-term dynamics of chemical/material systems. Within this coordinated approach, **(1)** RT-TDDFT calculations are first utilized to create a simulation set (i.e., pulse width, excitation frequency, and amplitude) for **(2)** calculating target "control" descriptors for enhanced electron/energy transfer efficiency. These computationally intensive calculations will be **(3)** optimized to efficiently run on leadership-class pre-exascale and exascale HPC systems to finally enable **(4)** Bayesian-based hyperparameter optimization algorithms to construct tailored optimal pulse shapes for controlling specific, driven excitations in chemical/material systems (such as polarization switching in ferroelectric materials or directed electron-transfer excitations on surfaces). Collectively, these four synergistic approaches address far-from-equilibrium

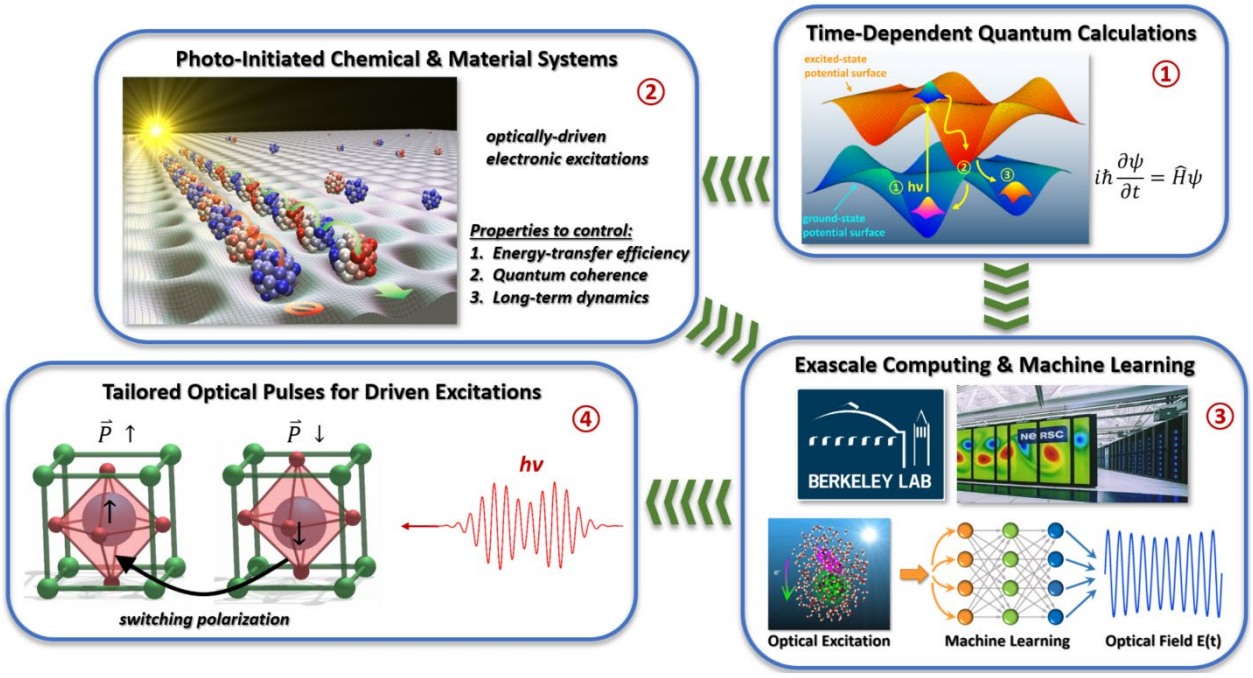

Figure 1: Overall scope and interconnected approaches for optimizing optically driven excitations to control the dynamics of chemical/material systems. **(1)** RT-TDDFT calculations with periodic boundary conditions are first utilized to generate data for **(2)** calculating target control parameters for enhanced electron/energy transfer efficiency in material systems. The ability to generate this high-quality data is enabled by **(3)** exascale computing and machine-learning approaches with the end goal of constructing tailored optimal pulse shapes for **(4)** controlling specific, driven excitations in chemical/material systems.

phenomena with advanced scientific computing, behavior of properties in electric/magnetic fields, and ultimately controlling chemical/material processes under the influence of driven excitations to predict their long-term dynamics. Due to the sheer volume of data and number of CPUs/GPUs used in these calculations, this project also addresses fault-tolerance computational issues, which has not received appropriate attention in most large-scale parallelization efforts. Finally, the LBNL team members in this project are also directly involved in the architecture exploration path for NERSC10 and beyond, which ensures that this project has direct relevance to upcoming DOE ASCR and exascale initiatives.

Keywords: time-dependent density functional theory, quantum dynamics, quantum control, massive parallelization, machine learning

Recent Progress

We have made progress on a new massively parallelized velocity-gauge real-time, time-dependent density functional tight-binding (VG-rtTDDFTB) implementation in the open-source DFTB+ software package for probing electronic excitations in large, condensed matter systems. Our VG-rtTDDFTB approach enables real-time electron dynamics simulations of large, periodic, condensed matter systems containing hundreds to thousands of atoms with a favorable computational scaling as a function of system size, described further below.

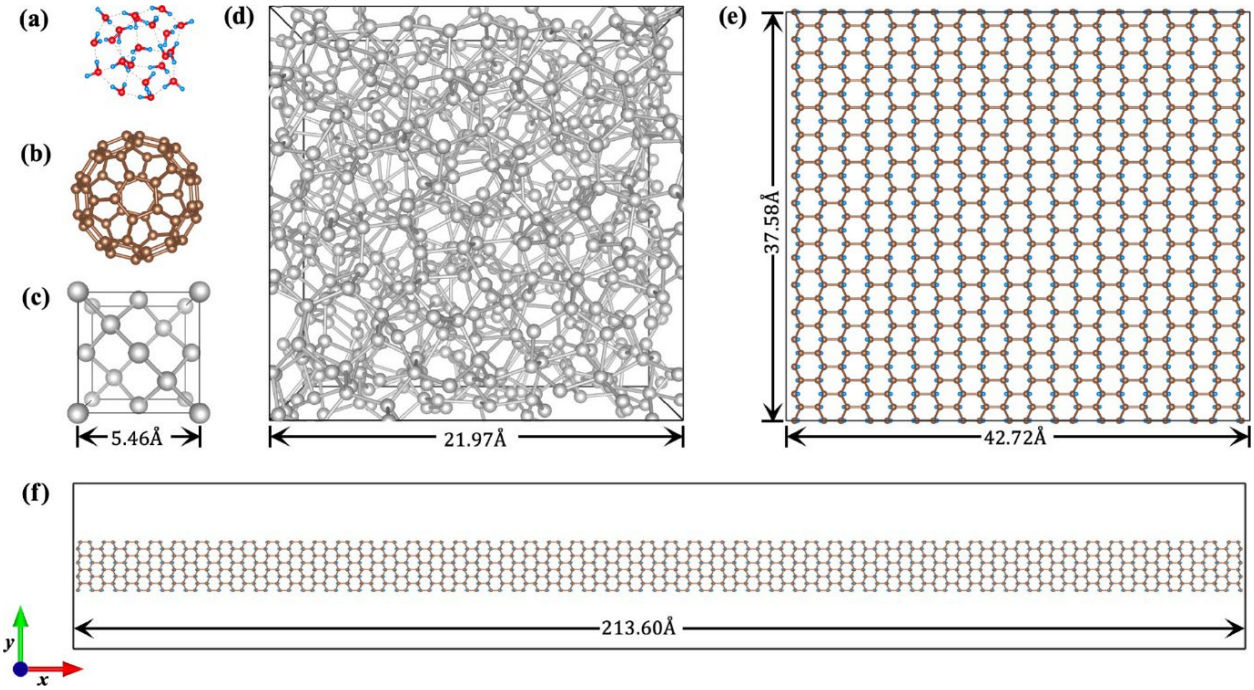

Figure 2: Molecular and periodic structures examined with our VG-rtTDDFTB approach: (a) $(H_2O)_{21}$ cluster, (b) C_{60}, (c) c-Si, (d) a-Si, (e) 2D-graphane ($C_{600}H_{600}$), and (f) 1D-graphane ($C_{800}H_{800}$).

To calculate optically-induced effects in material systems, the VT-rtTDDFTB approach solves the time-dependent Kohn-Sham equation of the form:

$$i\hbar\frac{\partial}{\partial t}\psi_{n,k}(\boldsymbol{r},t) = \widehat{H}_{\mathrm{DFTB}}(\boldsymbol{r},t)\psi_{n,k}(\boldsymbol{r},t), \quad (1)$$

where $\psi_{n,k}(\boldsymbol{r},t)$ is the wavefunction of the system in the Bloch representation, and $\hat{H}_{\mathrm{DFTB}}$ is the time-dependent DFTB Hamiltonian given by

$$\hat{H}_{\mathrm{DFTB}}(\boldsymbol{r},t) = \frac{1}{2}\left[\hat{p} - \int_0^t \mathbf{E}(\tau)\, d\tau\right]^2 + V_{\mathrm{DFTB}}[\rho(t)], \tag{2}$$

where $\hat{p}$ is the electron momentum operator, V_{DFTB} is the DFTB potential energy operator, and $\rho(t)$ is the time-dependent electron density. The time-dependent electric field, $\mathbf{E}(\tau)$, in **Eq. (2)** *can adopt any temporal dependence* and allows us to simulate incoming electromagnetic of various strengths and durations. In collaboration with Mauro Del Ben and Khaled Z. Ibrahim at Lawrence Berkeley National Laboratory (LBNL), we carried out quantum dynamics simulations of several large chemical/material systems described briefly below.

Figure 2 shows (a) a $(H_2O)_{21}$ cluster, (b) C_{60}, (c) a periodic crystal silicon (c-Si) structure, (d) an amorphous silicon (a-Si) structure containing 512 atoms, (e) 2D-graphane, and (f) 1D-graphane. We implemented our VG-rtTDDFTB approach with a hybrid MPI/OpenMP parallelization to enable large-scale electron dynamics simulations. Our parallelization is accelerated by distributing the *k*-point index over MPI ranks because the electron dynamics simulation is largely independent of each k-point and requires only minimal inter-core communications. At the node level, for each *k*-point, the computational workloads are distributed among cores using the multithreaded OpenMP parallelization. To evaluate the computational efficiency for our parallelized VG-rtTDDFTB implementation, we simulated various systems for 18,000 steps (36 fs) on the NERSC Perlmutter supercomputer with a δ-function "kick" field. **Table 1** shows that our parallelized VG-rtTDDFTB implementation enables extremely efficient electron dynamics simulations of large complex condensed matter systems that are too computationally expensive with standard rtTDDFT approaches.

System	Periodicity	Slater-Koster set	*k* mesh	# of basis function	# of CPU cores	Walltime (h)
$(H_2O)_{21}$	0	*mio*	1x1x1	126	1	0.01
C_{60}	0	*mio*	1x1x1	240	1	0.06
Crystal Si	3	*siband*	16x16x16	72	64	0.35
Amorphous Si	3	*siband*	1x1x1	4608	128	11.51
$C_{600}H_{600}$	2	*pbc*	1x1x1	3000	64	2.43
$C_{800}H_{800}$	1	*pbc*	1x1x1	4000	128	5.37

Table 1. Walltime for Various VG-rtTDDFTB Simulations (18000 steps, 36 fs) on the NERSC Perlmutter Supercomputer.

In collaboration with Mauro Del Ben and Khaled Z. Ibrahim at LBNL, we applied our new RT-TDDFTB approach to radiation-induced dynamics in massive, amorphous nanostructures. This work was featured prominently on the November 2023 cover of *J. Chem. Theory Comput.* (cf. **Fig. 3**). In particular, our results showed that radiation-induced effects in amorphous silicon are markedly different than in its pristine state, and the former can only be captured by our large-scale RT-TDDFTB calculations. Currently, this RT-TDDFTB code is limited to electromagnetic-induced interactions with the nuclei fixed, which is a good approximation for solid-state materials. However, to capture non-adiabatic bond-breaking processes, which are necessary for describing decoherence or light-induced reactions, we are currently augmenting our code to account for these

effects. We anticipate that some of the recent improvements in parallelization implemented by our LBNL collaborators in RT-TDDFTB will further reduce computational cost, enabling real-time dynamical effects of nuclear movement in these large systems. Consequently, this new computational capability would provide the necessary means to address optically-induced mechanisms in a large range of material systems and opens up the possibility for transformative collaborations between domain scientists and computer scientists in this SciDAC project.

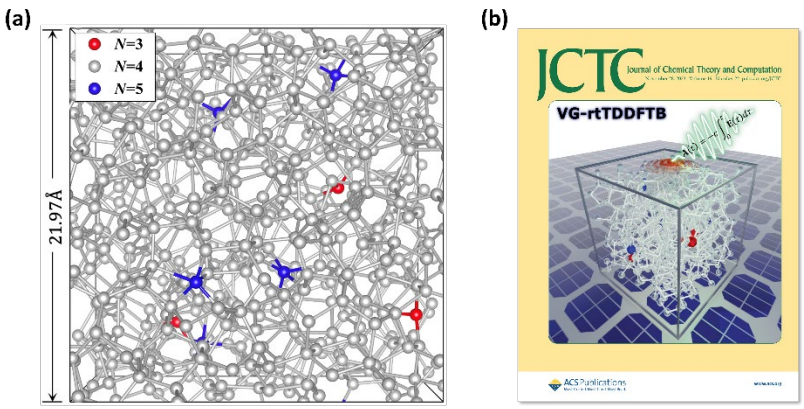

Figure 3. (a) Amorphous silicon box with various *N*-coordinated sites calculated with excited-state TDDDFTB (b) Nov. 2023 cover of *J. Chem. Theory Comput.* depicting a picture of amorphous silicon irradiated with an electromagnetic pulse calculated with excited-state RT-

Future Plans

For our future plans, we plan to fine tune the performance optimization of RT-TDDFTB to exploit GPU acceleration. We will leverage the portable OpenMP-target implementation to ensure performance portability across vendor architectures such as NVIDIA, AMD, and Intel. Within this implementation, we aim to target systems of 10k atoms with orders of seconds per time step to enable long dynamics with excellent time to solution. In addition, in collaboration between Wong (UCR), Shelton (UCR), and Ibrahim (LBNL), we aim to use Bayesian optimization to construct desired excitations in these quantum systems and explore how to utilize parallel simulations within our Bayesian optimization method. Our initial explorations suggest that the Gaussian process used as the typical surrogate model for Bayesian optimization, while performing well, may not be ideally suited for high-dimensional quantum systems. We will also explore the development of Bayesian radial basis function networks, which we believe can more easily model the objective landscape for this quantum control problem.

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Dispersive Interactions in Quantum Materials: Interplay between Anisotropy, Doping, and Non-linearity

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Keywords: van der Waals interactions, Casimir phenomena, optical response, topological materials, nonlinearity

Research Scope

Electromagnetic fluctuations exchange between objects gives rise to the ubiquitous van der Waals (vdW)/Casimir interactions [1]. Even though such interactions are universal, they are influenced by many factors making their investigation challenging. In addition to deep understanding of the materials properties and their dependence upon various factors, advanced theoretical and computational models capturing more diverse circumstances are needed for the successful modeling of vdW/Casimir phenomena. The fundamental understanding of vdW/Casimir phenomena requires an in-depth knowledge of the electronic and optical response properties of the underlying materials within effective medium and atomistic models. The unusual properties of Dirac materials, Weyl semimetals, and Chern insulators lead to unexpected outcomes in their fluctuation-induced interactions, giving rise to repulsion, quantization, lack of retardation, strong thermal fluctuations, etc…. Layered materials and heterostructures formed by stacking of different 2D monolayers have given an impetus to the development of new computational methods for vdW interactions that are compatible with density functional theory (DFT) codes. This research is inspired by novel materials, and it is driven by the connection between advanced models of their electronic and optical response properties and computational frameworks beyond currently available schemes. The ultimate goal is to examine various aspects of vdW/Casimir phenomena for better control and modulation, and also to advance basic science of materials for the benefit of broader communities working in the areas of condensed matter physics and optics.

Recent Progress

Casimir Interactions and twisted bilayered graphenes at magic angles: The short atomistic periodicity of graphene together with the long moiré superlattice period of *AA-AB* domains in a graphene bilayer creates an environment with unique properties. At special (magic) twist angles θ, electronic correlations can drive an array of phases by tiny changes in doping [2]. The electronic structure description is quite challenging, and here we have employed a ten-orbital tight binding model by introducing *phenomenologically* at the mean field level different order parameters that mimic the appearance of electronic phases due to breaking of C_2, C_3, and T symmetries. This is followed by computations of the optical conductivity tensor within the Kubo formalism by taking into account the coupling between electrons and electromagnetic fields via a Peierls substitution. By varying these order parameters, we are able to obtain anomalous quantum Hall effect (QAHE), quantum spin Hall insulator effect (QSHE), Dirac state, and nematicity in magic angle TBG. These have nontrivial consequences in the optical response, which further affect the Casimir interaction phenomena of two magic angle TBGs (Fig. 1). By employing a generalized Lifshitz formalism [1], we find that the Casimir energy for an uncorrelated Dirac TBG state is simply twice that of two graphene sheets. For TBGs with a broken T symmetry, the energy is $E = -\frac{\hbar c \alpha^2}{8\pi^2 d^3} \mathcal{C}_1 \mathcal{C}_2$ ($\mathcal{C}_{1,2}$ –

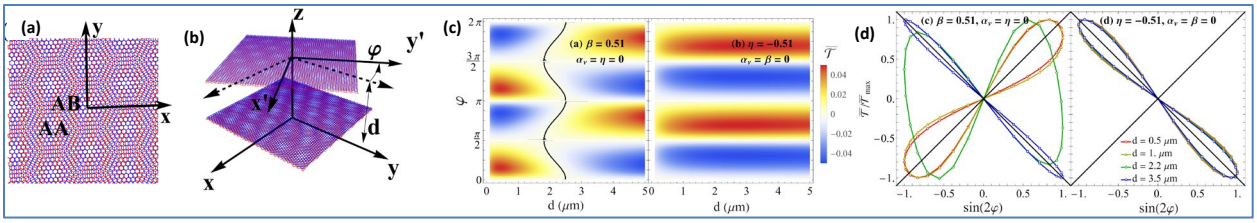

Figure 1 (a) A twisted bilayer graphene (TBG) with AA and AB stacking patterns; (b) two identical TBGs separated by a distance d with a relative angle φ of their optical axis; (c) density maps of the normalized Casimir torque $\bar{\mathcal{T}} = \frac{1}{E_M}\frac{\partial E}{\partial \varphi}$ for different parameters ($E_M = -\frac{\pi^2\hbar c}{720d^3}$); (d) the ratio of $\bar{\mathcal{T}}/\bar{\mathcal{T}}_{max}$ as a function of $\sin 2\varphi$, where $\bar{\mathcal{T}}_{max}$ denotes the maximum torque.

Chern numbers for both TBGs; α – fine structure constant), while for the QSHE state with broken C_2 symmetry $E \sim -\frac{\alpha^2}{d^5}$. On the other hand, for nematic TBGs E behaves similarly as the interaction between two metals with $E_M = -\frac{\pi^2\hbar c}{720d^3}$. Furthermore, the anisotropy in nematic TBGs results in a Casimir torque $\mathcal{T}$: the misaligned inequivalent optical axis of the TBGs causes a relative rotation induced by the Casimir force (Fig. 1).

The effect of nonlinearity in van der Waals interactions: At nanoscale separations, which is the vdW regime, effective medium models (as in the case of TBGs) are not adequate anymore, and one must consider discrete object representations. It is noted that atomistic schemes currently available are primarily focused on calculations within linear response theory [3]. However, many materials, such as non-centrosymmetric transition metal dichalcogenides and Weyl semimetals, are found to exhibit much enhanced second-order and third-order nonlinear hyperpolarizabilities. To broaden vdW computations, we developed a *Discrete Coupled Nonlinear Dipole* method as a generalized framework for dispersive interactions between nanoparticles. The method relies on a quantum mechanical Hamiltonian H for interacting non-linear dipoles fluctuating around equilibrium positions by taking into account $\boldsymbol{\alpha}$ (linear polarizability, tensor of rank 2), $\boldsymbol{\beta}$ (second order nonlinear hyperpolarizability present in noncentrosymmetric materials only, tensor of rank 3), and $\boldsymbol{\gamma}$ (second order nonlinear hyperpolarizability, tensor of rank 4). The scheme utilizes a diagonalization procedure supplemented by perturbation theory to treat explicitly second and third order nonlinear hyperpolarizabilities of each nanoparticle in the system. Setting $\boldsymbol{\beta} = \boldsymbol{\gamma} = 0$, one recovers our previous many-body dipolar approach based on linear response theory [4].

The method is illustrated in two types of molecules: azulene ($C_{10}H_8$) and 4-dimethylamino-4'-nitrostilbene ($C_{16}H_{16}N_2O_2$) found to be optically anisotropic with nontrivial $\boldsymbol{\beta}$ and $\boldsymbol{\gamma}$ tensors. In Fig. 2, calculations are shown for the vdW energy of two $C_{10}H_8$, two $C_{16}H_{16}N_2O_2$, and two chains of ten $C_{10}H_8$ molecules. It is found that the interaction between two molecules is strongly affected by the anisotropy (parameter ε) in the polarizability and the hyperpolarizabilities making it very different than the usual London-type energy between two molecules. In the case of molecular chains, repulsion is also possible. Fig. 2 shows that while two azulenes in the displayed orientations attract each other, two chains of the same molecules experience repulsion. We find that this is due to the strong second order nonlinear response and its specific tensorial structure. These results give strong evidence that indeed optical nonlinearities can affect significantly the magnitude and stacking patterns of vdW interactions, and the *Discrete Coupled Nonlinear Dipole* approach gives a new computational platform for further examinations of vdW interactions in nonlinear materials.

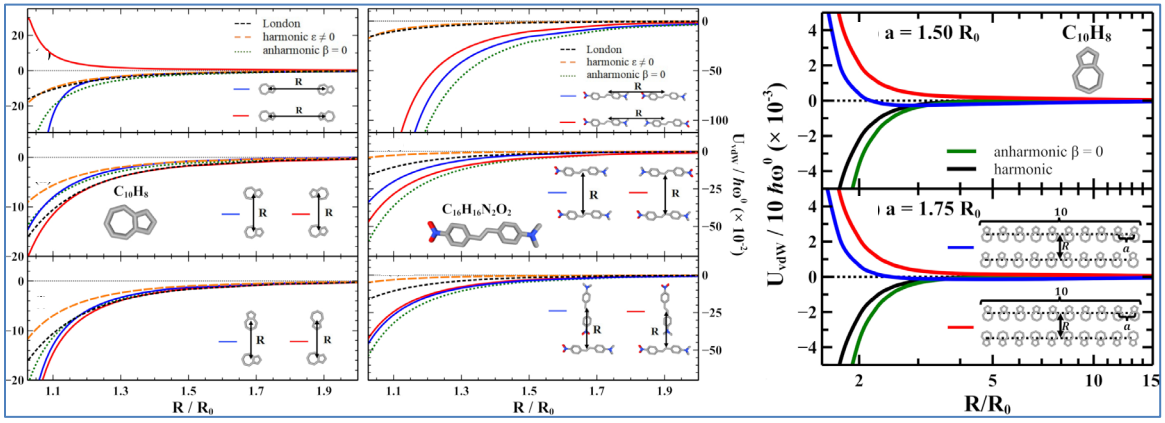

Figure 2 VdW energy between two $C_{10}H_8$ molecules (left column), two $C_{16}H_{16}N_2O_2$ molecules (middle column) and two chains of ten $C_{10}H_8$ molecules (right column) as a function of separation scaled by the size of the dipole $R_0 = \sqrt[3]{\alpha/2\pi\epsilon_0}$ for various orientations. Here ϵ is an anisotropy parameter and a denotes molecule separation in each chain. Also, harmonic calculations correspond to ignored perturbation contributions.

Data-driven models for vdW interactions with a machine learning approach: Available repositories containing various information for different systems are stimulating *data-driven* emerging directions in materials science. In addition to developing models and computational tools to calculate Casimir/vdW interactions, some of our efforts in this cycle are devoted towards exploring how data can be used for statistical modeling of dispersive interactions and related frictional properties [5]. When two objects are displaced against each other, the resulting energy dissipation gives rise to friction. Here we explore a data-driven approach to extract and understand materials trends and patterns in adhesion, corrugation, and van der Waals energies, $E_{adh}, E_{corr}, E_{vdW}$, of nanoscaled materials: properties that are closely related to friction at the nanoscale. We perform statistical modeling based on machine learning (ML) algorithms of bilayered materials composed of identical monolayers. Using DFT simulations we create a *bilayered dataset (BMDS)* containing over 760 materials in different stacking configurations and their basic electronic and transport properties. Using tree-based and gradient boosting ML algorithms statistical models are constructed with carefully optimized feature selections including electron affinity, packing fraction, mass density, atomic radii, unit cell, interlayer distance, etc…The ML algorithms are trained on 90% of the dataset and validated on the remaining 10% materials using the Scikit-learn package. A deeper insight into the physics behind the ML models is gained by further analyzing how different atomistic and monolayer features affect the frictional properties using the SHAP analysis. The numerical relations uncovered via SHAP give *quantitative* means to relate the strong correlation between the vdW and adhesion energy and the polarizability of the atoms. The relations between the commensurability of the bilayers, the magnitude of the unit cell, and electron affinity and the corrugation energies further help us identify types of materials and sliding paths for much reduced (or enhanced) friction [5].

Future Plans

The future research aims at developing a deeper understanding of fluctuation induced interactions by moving forward with new computational frameworks and analytical models. I continue building on the past and ongoing work currently supported by this grant and draw inspiration from new materials discoveries. One broad objective is to present a new Wannier Function (WF)

computational method for vdW interactions. The method is based entirely on electronic properties (captured in WF spreads and centers) and it enables the inclusion of optical nonlinearities within a many-body approach. This is an electron-based approach and it is different than the atom-based existing schemes; it broadens our computational capabilities beyond linear response descriptions. Another objective is to pursue a quantum many-body modeling of radiative heat transfer between objects with realistic optical nonlinearities. A data-driven approach based on computed response properties of layered materials within first principles is also planned in order to uncover materials signatures (electronic and atomic properties, hyperbolicity, specific surface modes) for much enhanced energy transfer. Efforts to bring forward theoretical understanding of vdW/Casimir interactions modulated by phonons and currents are also planned. Phonons, typically overlooked in such problems, couple to electromagnetic excitations in piezoelectrics, for example, and can affect the interaction energy in unexpected ways. Rarely considered dc currents introduce nonequilibrium and nonreciprocity conditions, which also hold much promise for new ways of vdW/Casimir interaction control.

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First Principles Investigations for Magnetic Properties of Innovative Materials

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Keywords: **topological properties, spin filtering, chiral materials, interface superconductivity**

Research scope

This project aims to explore the innovative realm of low-dimensional heterostructures through the development and application of theoretical models grounded in density functional theory. Our objective is to discover novel materials and phenomena that could pave the way for technological advancements. We delve into the intricate interactions between electron, spin, phonon, and magnon excitations to define new principles for manipulating and utilizing exotic properties of 2D materials and chiral crystals.

Recent progress

1. Control of two-dimensional magnetic and topological properties

We recently investigated the possibility of tuning topological phases of diverse materials with electric field and lattice strain. As shown in Fig 1a, we used a model with two interfaces between TI and magnetic insulator (MI). With a basis set of $|t\uparrow>$, $|t\downarrow>$, $|b\uparrow>$, and $|b\downarrow>$ (t and b represent the top and bottom surface states and $\uparrow$, $\downarrow$ represent the spin up and spin down states) the low-energy effective Hamiltonian including the surface states, proximity exchange effect and potential difference between the two surfaces can be expressed as

$$H(k)=\begin{bmatrix} 0 & iv_f k_- & m(k) & 0 \\ -iv_f k_+ & 0 & 0 & m(k) \\ m(k) & 0 & 0 & -iv_f k_- \\ 0 & m(k) & iv_f k_+ & 0 \end{bmatrix}+\begin{bmatrix} \Delta & 0 & 0 & 0 \\ 0 & -\Delta & 0 & 0 \\ 0 & 0 & \Delta & 0 \\ 0 & 0 & 0 & -\Delta \end{bmatrix}+\begin{bmatrix} V & 0 & 0 & 0 \\ 0 & V & 0 & 0 \\ 0 & 0 & -V & 0 \\ 0 & 0 & 0 & -V \end{bmatrix} \quad (1)$$

Here, $k_\pm = k_x \pm ik_y$, v_f is the Fermi velocity, and $m(k) = m_0 + m_1\left(k_x^2 + k_y^2\right)$ describes the interaction between the top and bottom surface states. By solving this Hamiltonian, the boundary between quantum anomalous Hall (QAH) and normal insulator (NI) is given at $\sqrt{V^2 + {m_0}^2} = \Delta$. Typically, the exchange field (Δ) of a given vdW

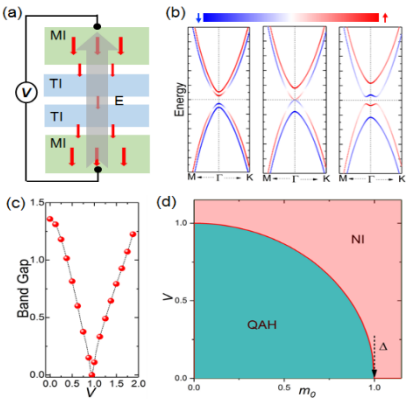

Figure 1. (a) Schematic diagram of a thin vdW heterostructure. (b) and (c) Evolutions of band structure and band gap with increasing electric field. (d) The phase diagram with a fixed Δ. Here the band gap, V and

heterostructure is fixed, and m_0 can be adjusted by controlling the film thickness, i.e., $m_0$increases as the film thickness decreases. For thin heterostructures with $|\Delta| > |m_0|$, the initial nontrivial band gap shrinks by applying a small electric field. As the potential difference V varies in a sufficiently large range, a reversible topological phase transition occurs between the quantum anomalous Hall effect and the normal insulator states as shown by the evolution of band gap and bands structures with increasing electric field in Fig. 1b and c. From the phase diagram in Fig. 1d, there is a large region for the realization of the QAH state. Recent experiment done by collavorators indeed reveal such electic-field driven topological phase transition in several model systems.

2. High dimensional spin filtering properties of chiral materials

We investigated the role antisymmetric spin-orbit coupling (ASOC) on the chiral-induced spin selectivity (CISS), exploring terms such as Rashba [$H_R = \lambda_R(k_y\sigma_x - k_x\sigma_y)$], Weyl [$H_W = \lambda_W(k_x\sigma_x + k_y\sigma_y)$], Dresselhaus [$H_D = \lambda_D(k_x\sigma_x - k_y\sigma_y)$ and $H_{D'} = \lambda_{D'}(k_y\sigma_x + k_x\sigma_y)$] terms. In systems with heavy elements, these ASOC terms become significantly influential, heralding a range of emergent properties. Integrating these terms with the standard CISS Hamiltonian, $H_{CISS} = \lambda_C k_z\sigma_z$, our analysis reveals that they can generate varied spin textures in reciprocal space. This demonstrates intricate spin-momentum locking, paving the way for realizing advanced spin Hall effects. Remarkably, in chiral crystals where either Weyl or Dresselhaus SOC prevails, the system may shift into a phase of heightened spin-current coherency, exhibiting spin selectivity in all three spatial directions. Importantly, we found that indium selenoiodide (InSeI), characterized by its helical chains rich in heavy elements and a substantial twist angle of 135° (refer to Fig. 2 a and b) may serves as a template to realize our theoretical predictions. Collaborative experimental efforts have led to the synthesis of InSeI nanowires and crystals. Our first-principles calculations indicate that its first valence (conduction) band is notably influenced by Weyl (Rashba) ASOC, as shown by the large SOC dependence of the band structure and density of states in Figures 2c, as well as by direct calculations of the spin accumulation. Furthermore, we observe that ASOC's sensitivity to structural symmetry alterations can trigger a phase transition, such as spin-flipping, by symmetry reduction (e.g., from C_{4V} to C_{2V}). Current experimental efforts are focused on refining synthesis techniques to enhance control over the dimensions and chirality uniformity of InSeI samples, aiming to corroborate our theoretical findings. These findings offer unprecedented opportunities for manipulating spin transport features of chiral materials by slightly altering their crystalline symmetry—a method of tunability fundamentally distinct from changing chemical morphology.

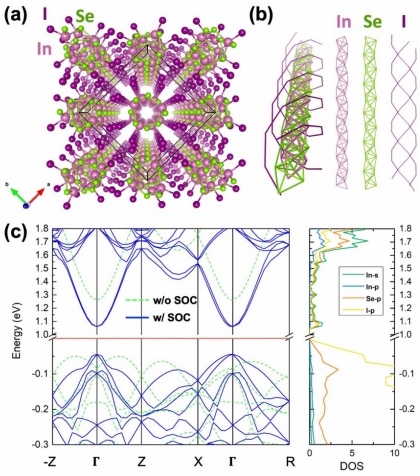

Fig. 2. (a) The bulk InSeI crystal with right-handed chirality. (b) The chains decomposed for In, Se, and I. (c) The band structures of InSeI with and without SOC and the orbital projected DOS.

Future Plans

- Investigate the effects of electric fields, mechanical strain, and interfacing on the topological properties of diverse two-dimensional materials.
- Examine how chiral structures in materials respond to various external stimuli, including electric and magnetic fields, as well as mechanical strain.
- Create computational codes to facilitate the study of chiral phonons and magnons within one-dimensional InSeI wires.
- Conduct research on the interactions between phonons and magnons in both two-dimensional and three-dimensional materials.
- Assess how structural changes at the interface affect the superconductivity of FeSe on substrates like SrTiO3 (STO) and BaTiO3 (BTO).

10 Publications through 2022

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Synthesis of motif and symmetry for accelerated learning, discovery, and design of electronic structures for energy conversion applications

Qimin Yan, Northeastern University

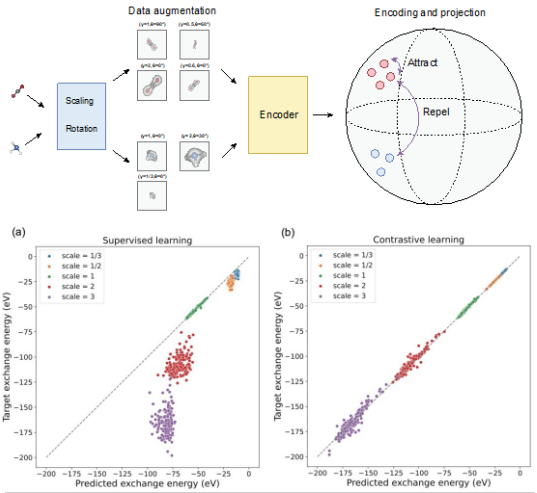
Keywords: motif, symmetry, physical constraint, neural network, graph, density functional theory

Research Scope

The scope of the proposed research is to develop symmetry-incorporated and motif-centric machine learning frameworks with a focus on the prediction of materials properties related to electronic structures at a dramatically reduced cost compared to ab-initio atomistic simulations. The developed machine learning (ML) models will be combined with symmetry tools and high-throughput computations to discovery and design functional oxides and 2D materials for energy conversion applications.

Recent Progress

In a data-driven paradigm, machine learning (ML) is the central component for developing accurate and universal exchange-correlation functionals in density functional theory. Exchange-correlation functionals must satisfy several physical constraints, such as density scaling, spin scaling, and derivative discontinuity. In a recent work,[1] we demonstrated that contrastive learning is a computationally efficient and flexible method to incorporate equality-based physical constraints in ML-based density functional design. We proposed a schematic approach to incorporate the uniform density scaling property of electron density for exchange energies by adopting contrastive representation learning during the pretraining task. The pretrained hidden representation is transferred to the downstream task to predict the exchange energies of molecule systems. The results demonstrate that incorporating exact constraints through contrastive learning can enhance the understanding of density-energy mapping using neural network models, which represents a viable pathway toward the machine learning design of a universal density functional via representation learning.



Top: a contrastive learning framework for incorporating uniform density scaling in ML-based density functional design. Bottom: Predicted exchange energies for ~10,000 molecules based on supervised learning and contrastive leraning.

Future Plans

Following the proposed research objectives in the proposal, we are working on the development of crystal hypergraph neural networks to create a universal learning framework to incorporate structure motifs and other higher-order material information. Also, we are developing a structure-motif-based material network to connect materials with motifs and learn the effective representations of both materials and motifs using both bipartite graphs and heterogeneous graphs. Another ongoing effort is the development of a Wannier-function-based and physics-informed multi-function learning model for both structural and electronic structure predictions of large-scale complex material systems (such as twisted multi-layer materials).

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Quantum simulation and state preparation for two-dimensional materials

Michael Zaletel, UC Berkeley

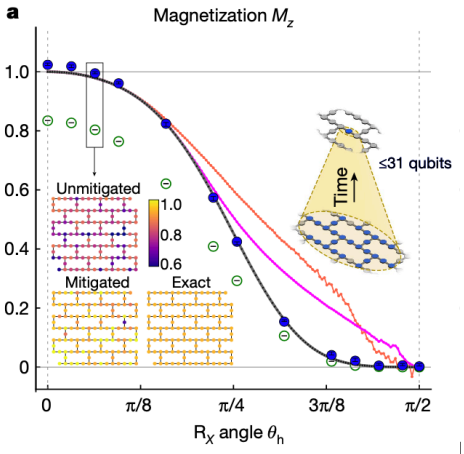
Keywords: Quantum simulation, tensor networks, quantum state preparation, frustrated magnetism, quantum dynamics

Research Scope

This research seeks to develop methods for quantum simulation and state preparation on near term quantum devices. The first thrust is the development of "isometric tensor network" (isoTNS) representations. First proposed as a tool for classical simulation of quantum materials, the ansatz allows for "holographic" quantum state preparation with a qubit count scaling only as the surface area of the system to be modeled. However, the variational power of the ansatz is not fully understood, nor whether there are useful instances for which there is a quantum advantage when manipulating them. The second thrust focuses on analog simulation of frustrated magnetism using Rydberg atom arrays. We first aim to understand what spin liquid phases can be stabilized within this space of Hamiltonians, and second, how can low entropy states be prepared and probed.

Recent Progress

In a collaboration with IBM research, we used a combination of exact and tensor network calculations to validate an error mitigation procedure for superconducting qubit platforms, "zero noise extrapolation" (ZNE). Comparing theoretical calculations with results on their 128-qubit Eagle device, we showed ZNE could give accurate results for simulation of quantum dynamics out to surprisingly large circuit depths.[1] In a followup, [2] we showed how improved classical tensor network algorithms could be used to match the results from Eagle in all circuits tested.

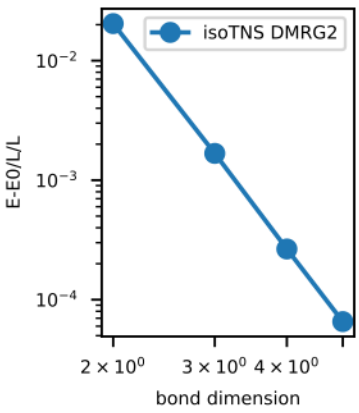


Comparing quantum dynamics on IBM's Eagle and classical approaches. Pure-state based tensor networks perform poorly in some regimes, which is not surprising given the linear growth in entanglement.

In works [3, 5], we extended the isoTNS ansatz to fermion systems, and systems which are infinite in one direction. These methodological developments are a step towards being able to apply isoTNS to interacting fermions in the thermodynamic limit, to study e.g. the Hubbard model.

Finally, in [4], we collaborated with the experimental group of A Browaeys to show that Rydberg atom arrays could be used to study both ferromagnetic and antiferromagnetic XY models. We found evidence for finite-temperature symmetry breaking stabilized by the long range (dipolar) nature of the interactions. While this study benchmarked the platform on unfrustrated square

lattice, ongoing experiments concern lattices where frustration is predicted to lead to spin liquid states.



Convergence of the isoTNS DMRG2 algorithm applied to the 2+1D quantum **critical** Ising model. For a quantum, critical state , as the bond dimension is increased from 2 to 5, the energy accuracy appears to improve as a power law.

Future Plans

First, we are nearing completion of a fully variational algorithm for optimizing 2D tensor networks, which we dub "DMRG^2". With this tool it should finally be possible to assess whether isoTNS provides a practical and reliable method for 2D ground state search. We will extensively benchmark the method, comparing with Monte Carlo, DMRG, and PEPs methods, in order to characterize the convergence rate.

We are also nearing completion of a finite-temperature isoTNS algorithm using a 2D generalization of the "METT's" algorithm. We plan to gather evidence on how the time complexity of this algorithm depends on the temperature and phase of matter being explored; the problem has interesting relations to measurement based quantum computation.

Finally, we are working to predict optical tweezer geometries which will stabilize gapped spin liquid phases in Rydberg atom aways. Most promising is a "breathing" versions of the Kagome lattice, which we finds drives a phase transition out of the usual Kagome spin liquid phase into a gapped "chiral spin liquid." Future research will determine optimal adiabatic state preparation protocols and measurement sequences for detecting the chiral spin liquid phase in Rydberg systems.

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Topological spin textures in chiral magnets: from 2D to 3D

Jiadong Zang, University of New Hampshire

Keywords: skyrmion, topological Hall effect, topological spin texture

Research Scope

This grant award focuses on the study of both fundamental physical property and material realization of magnetic spin textures with nontrivial topology. It consists of three major parts:

1. Investigating transport signatures of spin textures. Topological Hall effect (THE) has been abused for the purpose of identifying skyrmions. We will systematically examine THE in various systems and explore other mechanisms for THE. Current driven dynamics of skyrmions will be investigated as well.
2. Unlocking 3D topological spin textures. We will model the energy landscape and novel dynamics of 3D topological spin textures, hopfions for example, and develop new experimental methods to image 3D spin textures.
3. Developing a machine learning-based algorithm for 3D magnetic reconstruction. Its application in electron holography and X-ray will be investigated.

Recent Progress

1. Transport of spin textures
   We have studied the dynamics and Hall effects of skyrmions in several systems. Main breakthroughs include:
   a. Topological Hall effect in target skyrmions
      It has been believed by the community that topological Hall effect, featuring a hump in the Hall resistivity-magnetic field plot, is nonzero only when the overall topological charge of the spin texture is nonzero. The target skyrmion is a novel texture recently observed, in which the conventional skyrmion is wrapped by a circular helical stripe (Fig.1b). Although it is skyrmion related, the target skyrmion has zero topological charge, and the topological Hall effect is expected to be zero. However, we showed in this work that the topological Hall effect is still nonzero[1]. Using semiclassical study of electron trajectory and full quantum mechanical treatment using the Landauer-Büttiker formulism, we showed the sign reversal of the topological Hall effect depending on the Fermi level. Quantum coral states are also observed due to the confinement from the helical stripe.
   b. Thermal fluctuation induced topological Hall effect

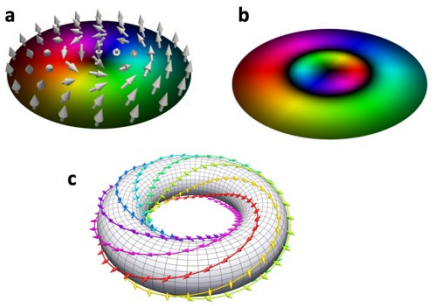

Fig1: Sketch of a skyrmion (**a**), a target skyrmion (**b**), and a hopfion (**c**)

In an earlier work, we predicted the topological Hall effect at high temperatures where the skyrmion long longer exists. We have confirmed this prediction recently by studying a realistic system, the bilayer Pt/LiAlFeO4. Using field theoretic approach and Monte Carlo simulation, we determined the scaling relation of thermally generated topological charge. The work will shortly be submitted to Nature Physics.

c. Topological Hall effect in intercalated $Cr_{1+x}Te_2$ compounds

$CrTe_2$ is a van der Waals material. Band structure calculation shows that it is also a Weyl semimetal. Interstitial Cr atoms can be intercalated into the Te-Te van der Waals gaps. As a result, a plethora of compounds have been discovered. We have investigated these compounds systematically in a recent work in collaboration with Soumyanarayanan group at National University of Singapore and Okada from Okinawa Institute of Science and Technology[2]. Intrinsic anomalous Hall effect was studied, which undergoes a sign flip at δ=0.33. It is explained in terms of the emergence of Weyl points therein.

There is also interesting topological Hall effect observed in $Cr_{1+x}Te_2$ compounds, but the origin is elusive. In a recent work submitted to Nature Communications, we did first principles calculations and suggested that the topological Hall effect is originated from the addition of anomalous Hall effects from ordered pristine Cr and intercalated Cr layers.

d. Manipulation of skyrmion dynamics by electric currents

Theory of current driven skyrmion motion has been out there for a while, but the realization of skyrmion motion in bulk chiral magnets was lacking. We made the first observation of the skyrmion dynamics in bulk chiral materials[3]. Via the theoretical design in terms of micromagnetic simulation, a notch was introduced into the FeGe nanoribbon. Electric current sending through the notch thus creates skyrmion clusters. Skyrmion Hall effect was also clearly identified, whose sign depends on the topological charge. Our micromagnetic simulations well reproduced all experimental observations.

A bottleneck of skyrmion-based device is the low skyrmion speed under current. We theoretically designed a new architecture in which skyrmions are confined in between helical lanes. Skyrmions therein receive a boost of speed by a factor determined by the ratio of non-adiabatic spin transfer torque and the Gilbert damping. This factor is surprisingly large in FeGe. Our first-principles calculation explains that in terms of small spin gap therein.

2. 3D topological spin textures

Skyrmion is mathematically a 2D entity. It is interesting to explore 3D topological spin textures. In a recent work[4], we found a skyrmionic vortex in FeGe tetrahedra with sub-100nm sizes. Due to the geometric confinement, topological charge therein is no longer integer-valued, but essential feature s of skyrmions are still kept.

This is a rich platform showing the existence of various topological textures such as skyrmion, vortices, and fractional edge vortex. We revealed a new mechanism called geometric chiral frustration for for formation of these textures.

3. Machine-learning based 3D magnetic reconstruction
   We have made preliminary success in reconstructing 3D magnetic structure using a U-shaped network architecture. Our algorithm outperforms existing non-machine learning methods[5]. It has been demonstrated to successfully resolve complicated skyrmion/helical structures.

Future Plans

1. Transport of spin textures
   We are currently working on an interesting resistor network model. The idea is to see the possibility of deriving topological Hall effect-like signals solely by the anomalous Hall effect in a simple multi-domain system. We have developed all necessary modeling and coding. Preliminary results show very positive signature. We still need to figure out what determines the size of topological Hall resistivity. It is very likely that the emergence of topological Hall-like signature is intimately related to the percolation phase transition.
   Some efforts will be devoted to understanding the non-adiabatic spin transfer torque. Our work on skyrmions confined in between helical lanes is the only work so far to measure coefficients of this torque directly. There is also a sign reversal of this torque but cannot captured by any theory so far. We plan to use a microscopic model to compute this torque by linear response theory.
2. 3D topological spin textures
   We will focus on the transport signature of the hopfion. A hopfion is a three-dimensional topological spin texture with nonzero Hopf index (Fig. 1c). Our preliminary result shows that the nonlinear conductivity in a hopfion lattice is associated to the moment of topological charge.
   Another exciting revenue we are developing is the optical response of hopfion lattice. The net magnetization is regarded as angular momentum $l = 0$ channel of the magnetization distribution, Recent development of orbital angular momentum light source can be used to detect nonzero l moments. A hopfion naturally consists of both $l = 1$ and $l = 2$ channels. It is thus an ideal platform to discuss interplay between nonlinear spin texture and nonlinear lights.
3. Machine-learning based 3D magnetic reconstruction
   Progress of this direction in the past year was slow due to the personal issue of the student involved. Fortunately, we have another student recently studied necessary techniques of machine learning. He will continue working on this project.

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Data-driven Discovery of Inorganic Electrides for Energy Applications

Qiang Zhu, University of North Carolina at Charlotte

Keywords: Materials Screening, Machine learning, Crystal Structure Prediction, Global Optimization

Research Scope

Electrides represent a unique class of materials where excess electrons trapped inside crystal cavities behave as anions. The trapped electrons are loosely bound near the Fermi energy level and can be used to design new materials with low work functions or minimum thermodynamic work to remove electrons from the solid, high electron mobility, and nontrivial band topology. This research aims to accelerate the discovery of electrides through developing an advanced materials screening method that combines group theory, crystal structure prediction, machine learning, and high-throughput screening. Specific objectives of this research include: (1) incorporating symmetry relations into materials structure screening, (2) developing machine learning models that can perform efficient crystal structure prediction and evaluations of materials' structural and electronic properties; and 3) constructing an electride database by screening promising material structures within a large chemical space. The simulation results and database will provide the materials science community with a large number of potential electrides, allowing the experimental community to test these predictions and probe potential technological applications. These computational approaches will be transferable across all classes of inorganic materials and may be utilized for a wide range of energy research activities.

Recent Progress

To achieve the goal aforementioned objectives, we have launched the following projects at UNLV (2021-2023) and UNC Charlotte (2023-present).

1. **Symmetry relation analysis**. The transition between two adjacent low-energy basins is often related by symmetry. To investigate the displacive phase transition at the atomic scale, we have implemented a numerical algorithm to automate the detection of the symmetry relations between any two candidate crystal structures [1]. Using this algorithm, we systematically screened all possible polar–nonpolar structure pairs from the Materials Project database and establish a library of 4500 pairs that can be connected through a continuous phase transition with small atomic displacements. In future, we also plan to include it as a structure variation operator to boost the performance the existing crystal structure prediction algorithms [2].
2. **Electride materials screening.** The Zhu group has a long tracking record in the computational screening of electrides [3]. Recently, we have switched to structurally more complicated zeolite materials that has the potential to overcome the thermal stability issue of most know electride materials. To achieve this goal, we employed our newly developed

symmetry analysis function to systematically search for zeolite structures with the high symmetry Wyckoff sites that are occupied by anions. Using this as guide, we have identified a set of sodalites that can become thermally stable electrides due to the complex structural framework and thus promising for practical applications such as catalysts and spintronics [4].

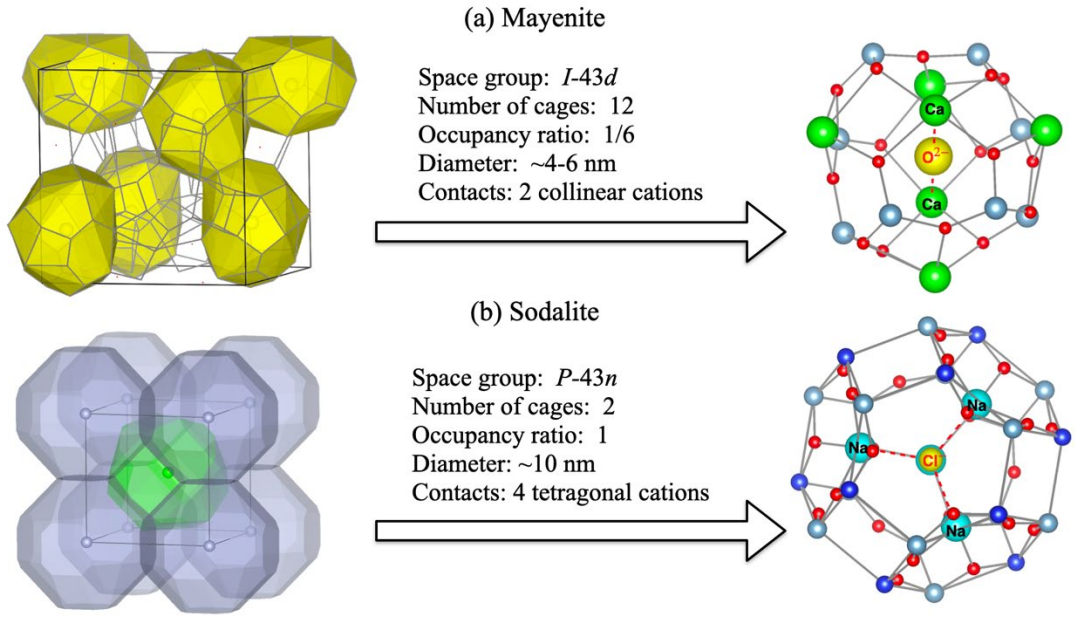

Fig. 1. Designing thermally stable electrides from the known zeolites (Kang & Zhu, JPCC, 2023)

3. **Machine learning Approaches for enhancing the existing atomistic simulations.** In this subtask, we focus on two aspects (1) development of machine learning interatomic potentials and (2) combining machine learning structure descriptor for innovative structure prediction method. First, we have been developing the PyXtal_FF package that can train the ML potential from different set of descriptors and regression method. Using PyXtal_FF, we developed a tailor MLP to simulate the phase transition of GaN. For the first time, we observe sequential change of phase transition mechanism from collective modes to nucleation and growths. The observed change of atomistic mechanism manifests the importance of statistical sampling with large system size in phase transition modelling [5]. Second, we are developing an alternative approach to predict complex crystal packing without energy minimization from expensive electronic structure calculations. Assuming the local atomic environments are well defined, we seek to directly perform global optimization using the metric of

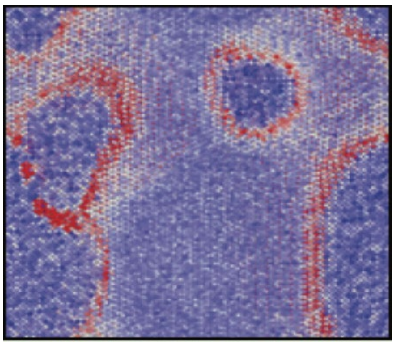

Fig. 2. Simulation GaN phase transition by MLP (Santos-Florez, PRL, 2022).

similarity with respect to the reference local environments that are represented by machine learning structure descriptors. Furthermore, we plan to use recent generative models to accelerate the exploration of latent chemical space. The proposed new methodology is expected to significantly boost the performance of existing crystal structure prediction methods and accelerate computational materials discovery.

Future Plans

In the future, we plan to focus on two following aspects,

1. **Extended computational screening of more electrides.** In particular, we will focus on the screening of 2D electrides and zeolite electrides. If possible, we will also seek for collaboration opportunities with the experimental groups to validate our theoretical predictions. Finally, we expect to collect all simulation results and publish a database for all identified electride materials.
2. **Code development for new generation of crystal structure prediction methods.** In the recent years, first-principles crystal structure prediction has become a popular tool for the design of new materials. However, most practical structure prediction approaches heavily rely on expensive energy minimization of many trial structures and the predictive capability is largely limited to a few tens of atoms in the unit cell. In this work, we will continue to develop the new structure prediction approach as described in task 3. Moreover, we plan to introduce the generative deep leaning model and reinforcement learning approach to boost the optimization process. We expect this new approach will be generally applied to the screening and design of complex materials such as battery materials, zeolites, molecular organic frameworks and molecular crystals.

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Symmetry breaking forms split-off flat bands in quantum oxides controlling metal versus insulator phases

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Keywords: Quantum Oxides; Symmetry breaking; Insulator to metal transition; Another Magnetism.

Research Scope: The overarching theme of this DE-SC0010467 program is "Understanding and Control of Charge Carriers in Quantum Materials". The list of publication after the previous TCMP meeting resulting from this BES supported program is provided at the end under "Publications". The current report concentrates on a recent direction, (indicated by the title) within this general subject. Pertinent questions addressed include:

- What controls the selectivity of undoped Quantum Oxides in being either persistent metals (as in $BaVO_3$, $SrVO_3$, $LaNiO_3$), or persistent insulators (as in $SrBiO_3$, $LaFeO_3$, $LaVO_3$), or being able to readily transform from metal to insulator (VO_2)
- How is this selectivity between "being metallic" and "being insulating" (Ref.1), predictable from evaluation of different forms of symmetry breaking (SB), often through the formation of *split-off flat bands.*
- The way in which such split-off bands occupied either by electrons ($LaTiO_3$, $LaVO_3$) or holes ($SrBiO_3$, $LaFeO_3$) offer natural predictors to the familiar phenomena of *intrinsic off-stoichiometry,* whereby certain oxides like to be oxygen poor, others oxygen rich.
- Implications for creating free carriers via impurity doping in such Mott insulators, to the benefit of potentially realizing the missing class of dopeable "Mott *Semiconductors*".

The results obtained in this study are intimately related to removal of band degeneracy. We will therefore briefly discuss in passing the connection with the ideas of *Non-Relativistic Spin Splitting (NRSS)* in Antiferromagnets and its enabling symmetry conditions (Ref.2,3). It turns out that the recently discussed Alter Magnetism (Ref. 4) represents subgroups of such known general NRSS.

Recent Progress: Senior colleagues who recall conferences on Cuprates and transition metal oxides in the early days of High T_c superconductivity tell stories on how theorists presenting mean field band structure calculations of such undoped compounds and predicting metallic characteristics instead of the known insulators, were almost driven out of the lecture rooms. Electronic structure calculations generally require as "input" a specification of (i) the level of description of inter electronic interaction, as well as (ii) the atomic-scale structure. Predictions of "false metal" phases alluded to above (some compiled in Ref.1) were often attributed to shortcomings in (i), such as lacking strong electronic correlations. Recently, increased attention was placed on group (ii) factors, namely the effect on predicted band structures of the microscopic degrees of freedom (m-DOF) nested within the crystallographic structure. These include local motifs such as atom-atom Peierls dimers, distorted octahedra, Jahn-Teller features, and local magnetic configurations. Such motifs have often been observed experimentally via local probes

(EXAFS, PDF, etc.) that avoid looking at the simplified "average crystallographic structures" that overlooks such local motifs.

We therefore use for input (i) deliberately just mean field like DFT, whereas for input (ii) we explore the correct macroscopic cell symmetry (say, cubic), but allow local sites to develop different (energy lowering) symmetries. We observe that when such local symmetry breaking total energy-lowering motifs are considered for their effect on electronic properties, one finds in a rather broad range of quantum oxides the formation of split-off flat bands that can also control the metallic versus insulating characteristics.

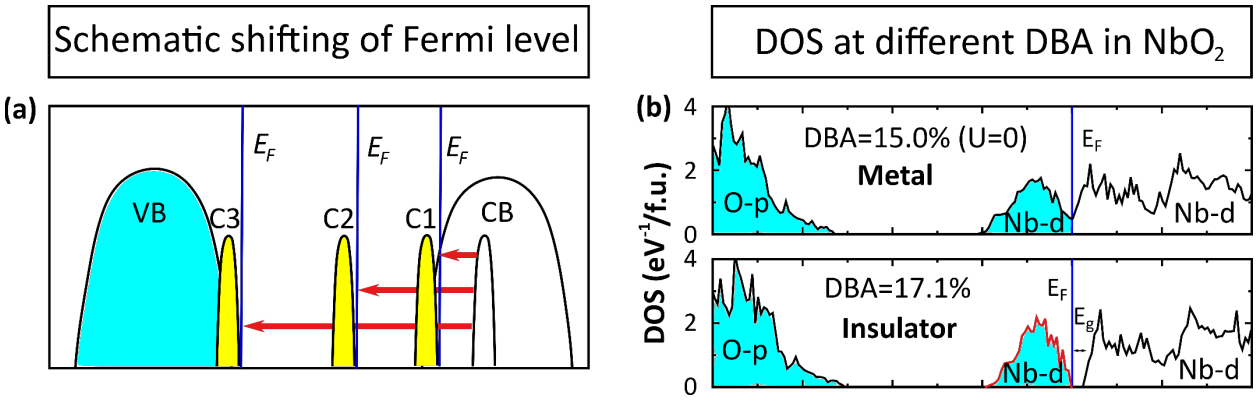

Figure 1. (a) Schematic showing how split-off bands (yellow peaks C1, C2 or C3) can split off due to symmetry breaking from the broad conduction band (CB), creating occupied, generally narrow bands. The shifts, denoted by the red arrows can be small (C1), intermediate (C2), or large (C3) depending on SB type and strength. (b) DFT density of states (DOS) at different degree (%) of Nb-Nb bond alternation (DBA) in NbO2 transitioning from a metal (C1case) to an insulator (C2 case).

Figure 1(a) illustrates three prototypes (C1, C2, C3) split off cases, whereas Fig 1(b) provides quantitative DFT results of NbO_2 at different degree of Nb-Nb bond alternation. If the perturbation is weak (as in $BaVO_3$ or $SrVO_3$ that hardly lower their energy) the system remains a metal (the C1 process). If the perturbation is stronger the split off band can pop up into the gap area creating an insulator (the C2 process, as in $LaVO_3$, $LaTiO_3$). In all cases the local motifs deciding the perturbation are obtained by examining total energy minimization in the appropriate supercell. Analogous effects are obtained for splitting off states from the valence band (VB) up as in $LaNiO_3$ (case V1), $SrBiO_3$ (case V2) or NiO (case V3). The split-off flat bands here are different types of flat bands in twisted bilayer graphene or structurally topological crystals.

Generality: Examples of our calculated range of quantum materials for the six types are given in Fig. 2. Interestingly, such split-off band effect is found to exist in magnetic and non-magnetic

oxides; in d-orbital (Mott-like) band edges (d, d*), or in more conventional charge-transfer oxides with (p, d*) band edges, or in oxides with "ligand hole" (p, p*) band edges, including even non-transition metal oxides. In other words, the effect transcends Mott physics. Neglecting computationally such m-DOF effects explains inconsistencies in the designation of quantum oxides as metals versus insulators, as noted routinely in DFT databases (listed in Ref. 1).

Association with tendencies to non-stoichiometry: Given that split-off bands tend to have specific charge polarity (trapped electrons as in $LaTiO_3$ or trapped holes as in $LaFeO_3$) their presence implies a correlation with spontaneous formation of specific intrinsic defects. For example, cation vacancies (oxygen vacancies) are generally hole-producing acceptors (electron producing donors) that are stabilized in crystals having electron-trapped (hole-trapped) isolated bands, respectively.

Type	Material	Symmetry breaking mode	ΔE_T (meV/f.u.)	Band edges	Metal or insulator	
					Before symmetry breaking	After symmetry breaking
C1	$CaVO_3$ $BaVO_3$	Magnetic Magnetic	-24 -34	(p, d*) (p, d*)	Electron Metal	Electron Metal
C2	NbO_2 $LaTiO_3$ $LaVO_3$	Dimerization Magnetic Mag., JT	-64 -302 -477	(d, d*) (d, d*) (d, d*)	Electron Metal	Insulator (Occupied split-off flat bands)
C3	To be found	-	-	-	Electron Metal	Insulator
V1	$LaNiO_3$ $SrFeO_3$	Magnetic Magnetic	-39 -1690	(d, p*) (d, p*)	Hole Metal	Hole Metal
V2	$YNiO_3$ $SrBiO_3$ $LaFeO_3$ $LaCrO_3$	Mag. disp. Bond disp. Magnetic Magnetic	-133 -763 -1778 -2967	(d, d*) (p, p*) (p, d*) (p, d*)	Hole Metal	Insulator (Unoccupied split-off flat bands)
V3	NiO MnO	Magnetic Magnetic	-1469 -4290	(p, d*) (p, d*)	Hole Metal	Insulator

Figure 2. Example materials of six types [Fig. 1(a)] of flat bands emerging from different symmetry breaking modes, the total energy lowering ΔE_T, and band edge orbital characters.

Future plans: Doping of Broken symmetry bands in Quantum Oxides: We found that split-off symmetry broken narrow bands can trap and localize native electrons (for states that split down in energy from the CB) or native holes (for states that split up from the VB). What happens if such undoped compounds would be intentionally doped by impurity atoms that are expected to be "electron producers" (donor dopants being n-type) or "hole producers" (acceptors dopants, being p-type)? Using "the modern theory of doping" (based on dependence of the solutions on Fermi energy and chemical potentials and structural relaxation) a number of groups noted: p-type doping normally expected to shift E_F down towards the VB, shifts instead the split-off bands containing trapped electrons up towards the CB. Understanding this "anti-doping" might hold the key to

realize the hope of converting Mott insulators into hitherto missing class of "Mott Semiconductors" where E_F can be shifted controllably by doping as in semiconductors.

Relation to other degeneracy removal effects and to "Alter Magnetism": The formation of split-off bands discussed here represents a degeneracy removal mechanism. The Rashba degeneracy removal represents a consequence of an enabling symmetry (broken inversion) in a non-magnet with spin-orbit coupling (SOC). The relativistic Rashba spin splitting (SS) raised the hope for a *non*-relativistic analogue. Pekar and Rashba raised this possibility in 1964 (Ref. 2) to potentially arise from the inhomogeneous magnetic field h(r) in antiferromagnets (AFM). The scenario was dormant for half a century; In 2020, in collaboration with E. Rashba, Yuan et al (Ref. 3) identified two enabling symmetry conditions (i.e. without which the effect does not exist) for the non-relativistic spin splitting (NRSS) in AFM. These are the two symmetries $\mathbf{S}_I$ and $\mathbf{S}_T$ each representing a product: $\mathbf{S}_I = \theta I$ (where I = inversion and θ = time-reversal) and $\mathbf{S}_T = UT$ [where U is spin-reversal SU (2) symmetry and T is fractional translation]. In collinear ordered magnetic materials with zero net magnetization the dual scale ($\mathbf{S}_I$ and $\mathbf{S}_T$) provides the identification of all the symmetries that can force the spin-up and spin-down energy bands to be degenerate throughout the Brillouin zone: AFM with θI symmetry obeyed but UT symmetry violated [Yes,No] are classified as Spin Splitting Type SST-1 (no spin splitting) such as CuMnAs. The simultaneous violation conditions [No, No] of [$\mathbf{S}_I$; $\mathbf{S}_T$] gives SST-4 as non-relativistic spin splitting. Using these enabling symmetries it has been possible to identify from magnetic data bases specific AFM compounds that have NRSS (CoV_2O_6, $Y_2Cu_2O_5$, $Cu_2V_2O_7$) and predict the spin splitting in DFT (MnF_2, $LaMnO_3$, $MnTiO_3$).

The term Alter Magnetism has been coined more recently (Ref. 4) for cases where in addition to the fundamental enabling symmetries [$\mathbf{S}_I$; $\mathbf{S}_T$] = [No, No] for NRSS (Ref. 3) there are rotation symmetries connecting spin-up and spin-down sublattices. These "Alter Symmetries" represent subgroups of SST-4. One such subgroup is SST-4 without additional Alter symmetries where the AFM phase still has proper zero magnetization (the numbers of valence spin-up and valence spin-down electrons are equal leading to zero total spin). Unlike other SST-4 subgroups, this type of SST-4 can have strong Γ-point spin splitting (L.D. Yuan, private communication); Such SST-4 AFM have the advantage of ferromagnetic systems for efficient spin operation while maintaining the advantage of zero net magnetization of AFM for fast spin operation.

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Quantum Materials: Magnetism, Spin-Orbit Coupling, and Superconductivity

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Keywords: proximity effects, spin-orbit coupling, magnetism, superconductivity

Research Scope

Advances in quantum materials and their heterostructures provide fascinating opportunities to reveal novel quantum phenomena and transfer them to other classes of materials. Our proposed research reflects the renewed interest in emergent and many-body phenomena in heterostructures, which can support magnetism, spin-orbit coupling, and superconductivity, as well as novel ways to implement such heterostructures. Rather than with the MBE growth, atomically sharp interfaces can be also realized by mechanical exfoliation of van der Waals materials. The resulting monolayers are a striking example of ultrathin nanostructures which can be transformed through proximity effects. However, even seemingly well-understood spin-orbit coupling provides many unexplored manifestations, such as the conversion between spin, charge, and light, in monolayers and heterostructures, or noise in superconducting tunnel junctions, which we elucidate in our research and guide their experimental realization.

Recent Progress

Terahertz Spin-Light Coupling in Proximitized Dirac Materials

Two-dimensional (2D) Dirac materials, such as graphene, are known to inherit both magnetic and spin-orbit coupling (SOC) proximity effects from its neighbors [R1]. We find that such proximitized Dirac material reveal a remarkable terahertz (THz) spin-light interaction, with a giant increase in the optical absorption that can be probed by the electron dipole spin resonance (EDSR) [1], which shares many similarities with the widely studied electron spin resonance (ESR). However, in EDSR the spin-flip transitions are induced by ac electric field, allowed in the presence of SOC and, therefore, EDSR is one of the manifestations of the spin-charge conversion. In a proximitized graphene, depicted in Fig. 1, the role of magnetic substrate provides the proximity-induced SOC and spin splitting, Δ, which removes the need to apply any static magnetic field to realize intraband spin-flip transitions. From the calculated optical absorption, shown in Fig. 2, we find that in the THz range it even significantly exceeds the universal absorption in graphene of 2.3% and that it shows a strong dependence with the strength of the Bychkov-Rashba SOC. This predicted EDSR, has striking differences from its previously studied mechanisms and leads to a remarkable increase of both the spin susceptibility and THz absorption, while also showing an anomalous polarization structure. We complement our Kubo

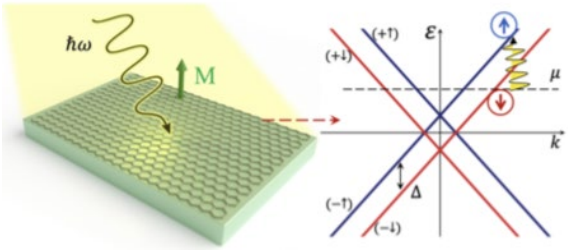

Fig. 1 Electric field of THz radiation causes intersubband spin-flip transitions in a graphene on a substrate with a magnetization, **M**. The Dirac spectrum with a proximity-induced spin splitting, Δ, wave vector, **k**, and the Fermi energy, μ [1].

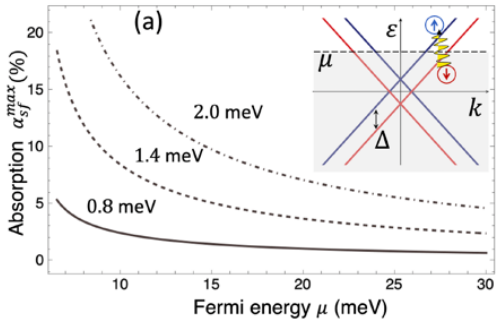

Fig. 2 THz EDSR absorption with the Fermi energy and SOC, λ_{SO} = 0.8, 1.4, and 2.0 meV.

formula calculations by using the classical picture of the spin resonance and introducing the underlying generalized Bloch equations, which describe the corresponding dynamics of the spin and lattice pseudospin, inherent to graphene. We find that the origin of the giant increase in the THz EDSR and its anomalous polarization structure comes from the resonant coupled spin-pseudospin dynamics [1]. This underlying mechanism is the previously overlooked dynamics of coupled spin and pseudospin torque.

Controlling the Helicity of Light by Electrical Magnetization Switching

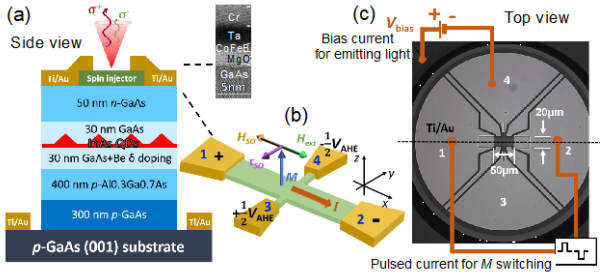

Fig. 3 SOT spin-LED. (a) Schematic structure with InAs QD active layer emitting light of positive/negative helicity. The spin injector and its STEM image. (b) **M** switching in the injector by the spin Hall effect, monitored by the anomalous Hall effect. (c) Top view of the optical microscopy image of the device. A pulsed current is injected to switch **M.** Vertical bias is applied to pump the LED [2].

Room-temperature spintronics is typically limited to magnetoresistive effects and unipolar devices where their transport is dominated by the carriers of a single charge polarity [R2]. In contrast, spin-light emitting diodes (spin-LEDs) are example of bipolar devices (both electrons and holes play a crucial role) and the conversion between the spin, charge, and light. Unlike all the prior work, studied for over 25 years [R2,R3], which is limited to low temperatures, an applied magnetic field (to realize out-of-plane magnetization, **M**, required that the dipole optical selection rules yield circularly polarized light), or quasi-static operation, our realization in Fig. 3 overcomes all of these constraints in a novel spin-orbit torque (SOT) spin-LED [2]. The experimental work led by our collaborator, Yuan Lu (University of Lorraine, France), used well-established magnetic tunnel junctions with out-of-plane **M**, common to magnetic hard drive and magnetic memories, which we have previously studied together for the SOC-induced spin-triplet superconductivity [R4]. We demonstrate that by passing a pulsed current through a neighboring heavy metal (Ta) with a strong SOC generates spin Hall effect with transverse spin current sufficient for **M** switching, even in the absence of any applied magnetic field. This resulting spin-orbit torque (SOT) mechanism, used in commercial memory applications, allows modulating spin polarization of the injected carriers in the InAs quantum dot where the carrier recombination and the conservation of the angular momentum is responsible for the helicity reversal of the emitted light. Through the conservation of angular momentum between photons, electrons, and ferromagnets, we demonstrate the missing link between the magnetization dynamics and modulation of the helicity of the emitted light [2].

Topological Flat Bands in Epitaxial Kagome Thin Films

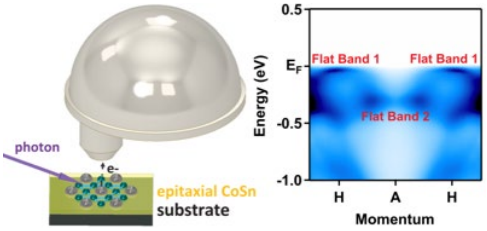

Fig. 4 Left: Schematic of the photoemission measurement of the epitaxial thin-film CoSn. Right: The corresponding electronic structure reveals characteristic flat bands [3].

Strongly-correlated electronic systems are one of the focus of condensed matter physics due to the emergence of interesting many-body ground states. Materials with dispersionless bands, i.e., flat bands, are ideal systems for studying the physics of strongly correlated electronic states due to the smaller bandwidth as compared to the Coulomb repulsion. One important class of materials exhibiting such flat bands is those based on quasi-2D kagome lattices, composed of equilateral triangles and regular hexagons. For realizing flat-band-induced phenomena in kagome metals, it is important to fine-tune the flat band position relative to the Fermi level, since many physical properties are dominated by states at the Fermi level. Using synchrotron-based angle-resolved photoemission spectroscopy (ARPES), shown in Fig. 4, the group of our collaborator, Roland Kawakami (Ohio State) has directly measured the band structure of the CoSn thin films and revealed multiple flat bands [3]. At the Γ point, SOC gaps were observed between one of the flat bands and the quadratic bands, suggesting the nontrivial topology of this flat band. Using density functional theory calculations, we found that the tunability of the flat bands through carrier doping is consistent with the ARPES experiments. Using the calculated band structure and semiclassical transport theory, we have quantitatively explained the measured transport properties of CoSn [3].

Tuning Corner States in Topological Insulators with Bulk-Boundary Obstruction

Second-order topological insulators (SOTIs) support topological states beyond the usual bulk-boundary correspondence and provide important connections between quantum chemistry and topology. A hallmark of the 2D SOTIs is the emergence of corner states, which usually arise from the topologically nontrivial obstructed states in the bulk. In contrast, we reveal a very different scenario where even trivial obstructed bulk states can induce corner states due to their open boundaries. We show that these two types of corner states can coexist in a single system and predict, that the monolayer C_2N is a promising candidate for their observation [4]. To overcome the limitation in manipulating corner states, we demonstrate it can be accomplished using a magnetic exchange field, where the corner states can be fully spin polarized and moved into the bulk states as we show from first principles in the C_2N/CrI_3 van der Waals (vdW) heterostructure.

Future Plans

The role of SOC in ferromagnetic/superconductor junction is known to strongly modify the conductance and its anisotropy. In contrast, the influence of SOC on the shot noise in such junctions is not understood. Our theoretical studies will be supported by the experiments led by the group of Farkhad Aliev (Autonomous University of Madrid, Spain). We will study effects of

Rashba SOC on excitons in proximitized vdW Heterostructures using Bethe-Salpeter equation.

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