# DOE-BES Ultrafast Roundtable

## Opportunities at the Frontiers of XFEL Ultrafast Science

Tony F. Heinz

*SLAC National Accelerator Laboratory*

*Stanford University*

# Outline of Today's Presentation

- Scientific setting and background
- Roundtable process
- Priority Research Opportunities
- Cross-cutting Research

# Ultrafast Science

<u>Ultrafast time scales</u>

| | |
|---|---|
| Light: | 300 nm/fs |
| Electron: | 0.3 nm/fs |
| Nuclei: | 0.003 nm/fs |

***We can now freeze motion of relevant fundamental processes in chemical and material systems***

- **Examine motion of electrons, nuclei, and their interactions to address fundamental problems in material and chemical science**
- **Not only observe motion, but drive it into new nonequilbrium regimes**

# Ultrafast Science: X-rays

**Importance of ultrafast x-rays**

- **Structural analysis**
- **X-ray spectroscopy: chemical specificity**
- **Beyond Fourier-transform constraint of optical radiation: attosecond time resolution**
- **Light-matter interactions in regime of high x-ray intensities and nonlinear x-ray processes**

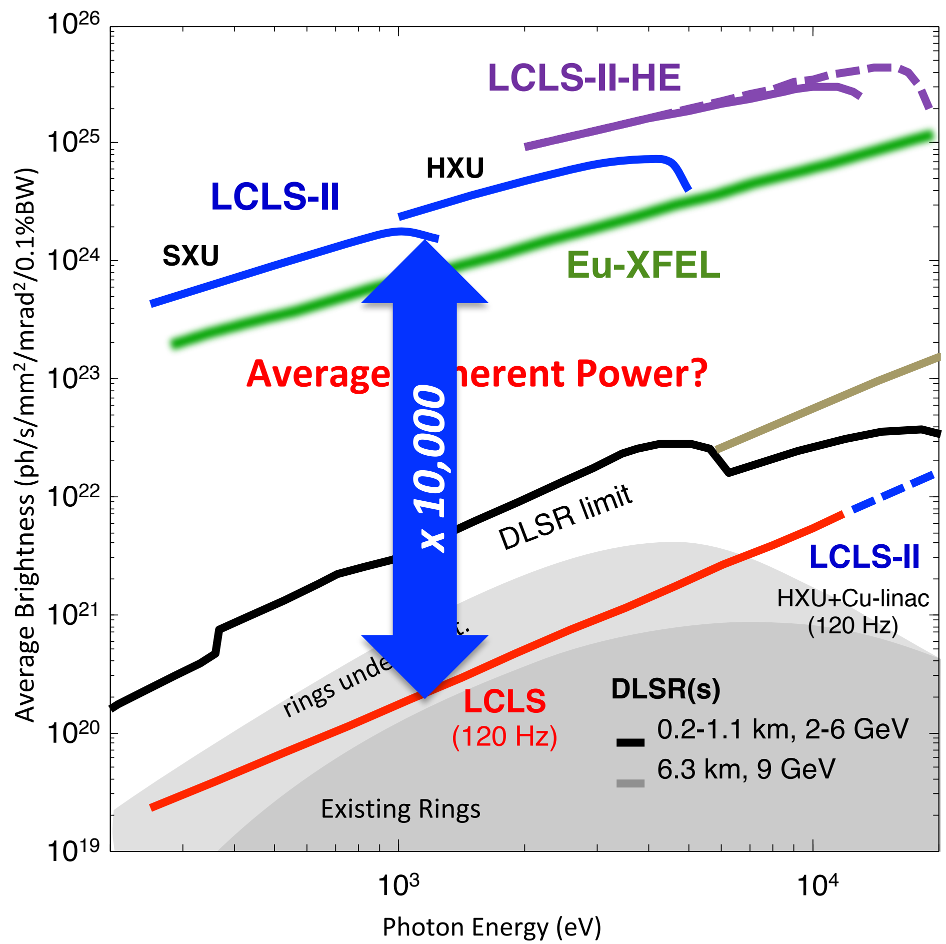
# X-rays Free Electron Lasers: LCLS

Coherent x-rays across the spectrum
with pulse durations down to 10’s of femtoseconds

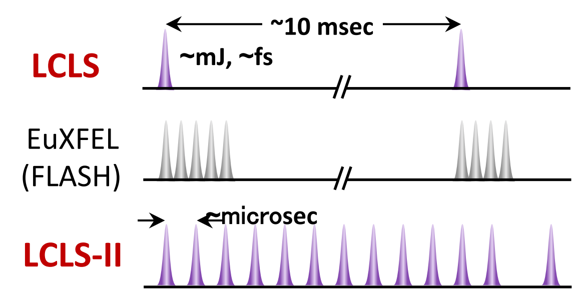
# LCSL and LCSL II

## LCLS-II Project – 1st Light 2020


Photon Energy (eV)




**LCLS-II:**
- CW-SCRF linac (4 GeV) in 1st km of linac
- Two new tunable undulators
- Repetition rate up to 1 MHz
- Photon energy reach – 25 keV (120 Hz)
- Stability, coherence (seeding)



# DOE-BES Ultrafast Roundtable Charge

The focus of this roundtable is to **identify the research priorities, key science drivers and research strategies for the BES research portfolio that uses LCLS, including its prospective upgrades.** The roundtable will provide **input to optimize BES' research investment in ultrafast x-ray science research and ensure the scientific impact of this research to the broader BES mission.**

Also to be explored are opportunities for enhanced ultrafast community synergy across BES scientific disciplines. The roundtable and subsequent report will provide an assessment of the current status of BES ultrafast x-ray science, and define a path for optimal utilization of future LCLS-II capabilities by BES-supported **research programs in the 2-10 year timeframe, extending well beyond the first LCLS-II experiments**. The roundtable will serve to illuminate areas where gaps exists between the current BES research portfolio and the LCLS-II experimental capabilities including the associated ultrafast scientific challenges and opportunities. The roundtable output will be used by BES to lay the foundations for new directions and support for future ultrafast science research.

DOE-BES Ultrafast Roundtable

# Opportunities at the Frontiers of XFEL Ultrafast Science

Oct. 25-26, 2017


Co-Chairs: Tony Heinz, SLAC
Oleg Shpyrko, UCSD

DOE team:

| | |
|---|---|
| Helen Kerch (MSE) | Linda Horton (MSE) |
| Tom Settersten (CGSB) | Jeff Krause (CSGB) |
| Lane Wilson (MSE) | Bruce Garrett (CSGB) |

# Roundtable Plan

- **Imaging Nuclear Dynamics**
- **Imaging Charge Dynamics**
- **Inducing and Probing Collective**
- **High Field, Attosecond Frontier**

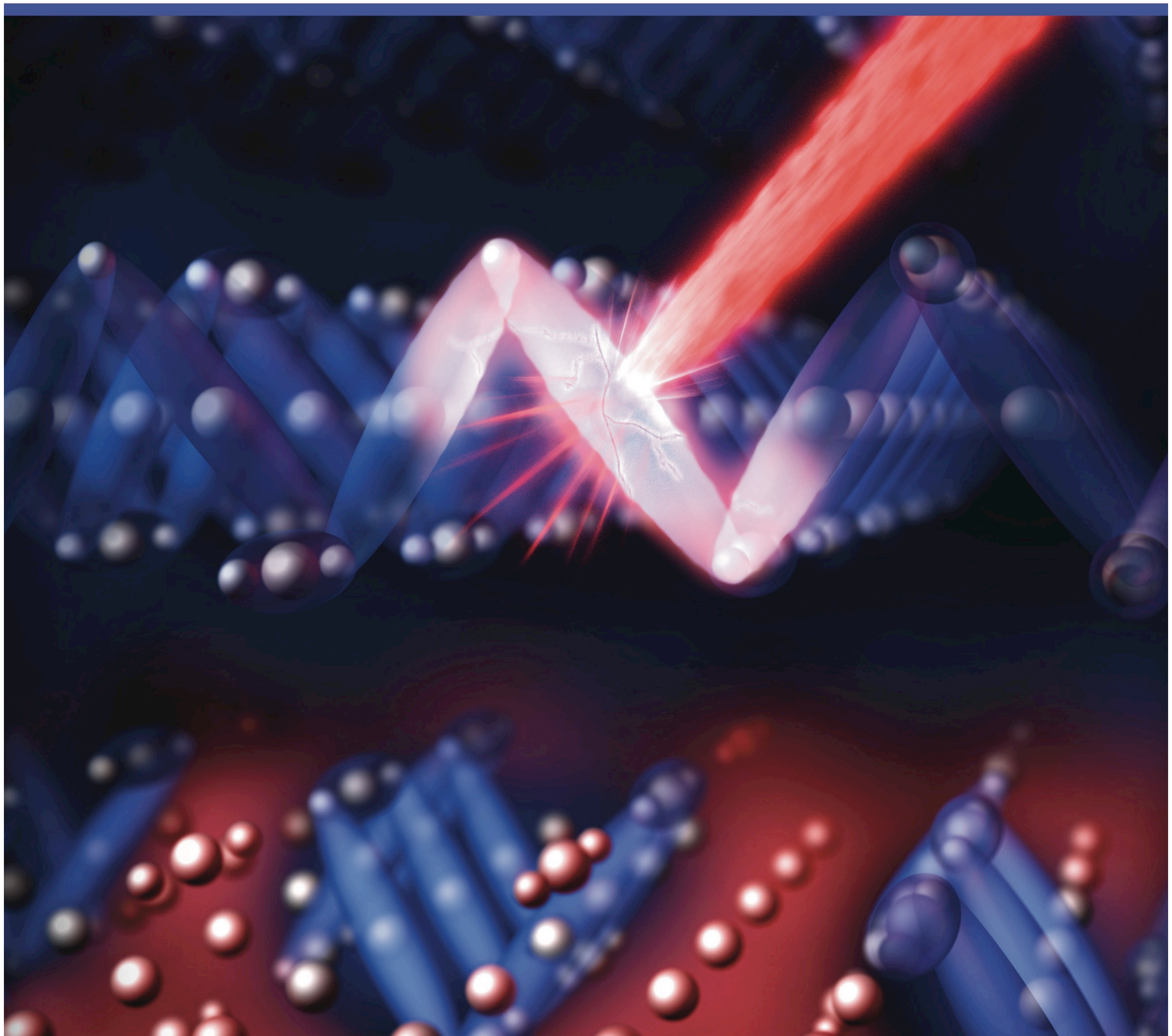
Panel leads: Dmitri Basov (Columbia)
Nora Berrah (U Conn)
Oliver Gessner (LBNL)
Rohit Prasankumar (LANL)

# Priority Research Opportunities

- **Attosecond electron dynamics within a molecule**
- **Light-induced states of matter**
- **Dynamics of spontaneous transformation of matter**
- **Cross-cutting research**

Basic Energy Sciences Roundtable

# Opportunities for Basic Research at the Frontiers of XFEL Ultrafast Science



October 25–26, 2017

Will be posted shortly

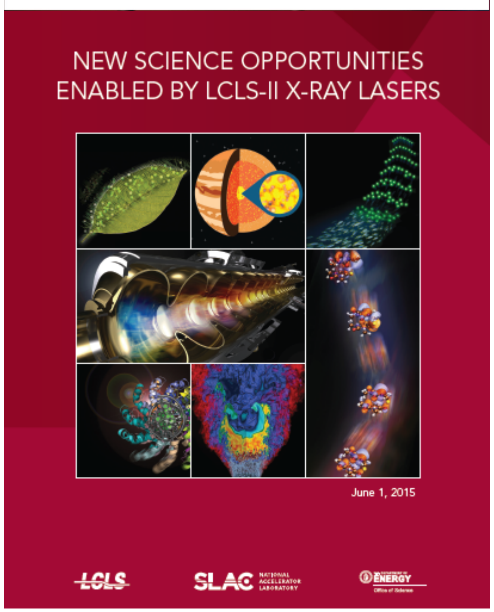
# Opportunities at the Frontiers of XFEL Ultrafast Science

- Focuses on new possibilities enabled by LCLS-II
- Complements earlier planning documents for LCLS-II, such as 2015 report
- Complementary techniques are also very important, but not focus of the roundtable

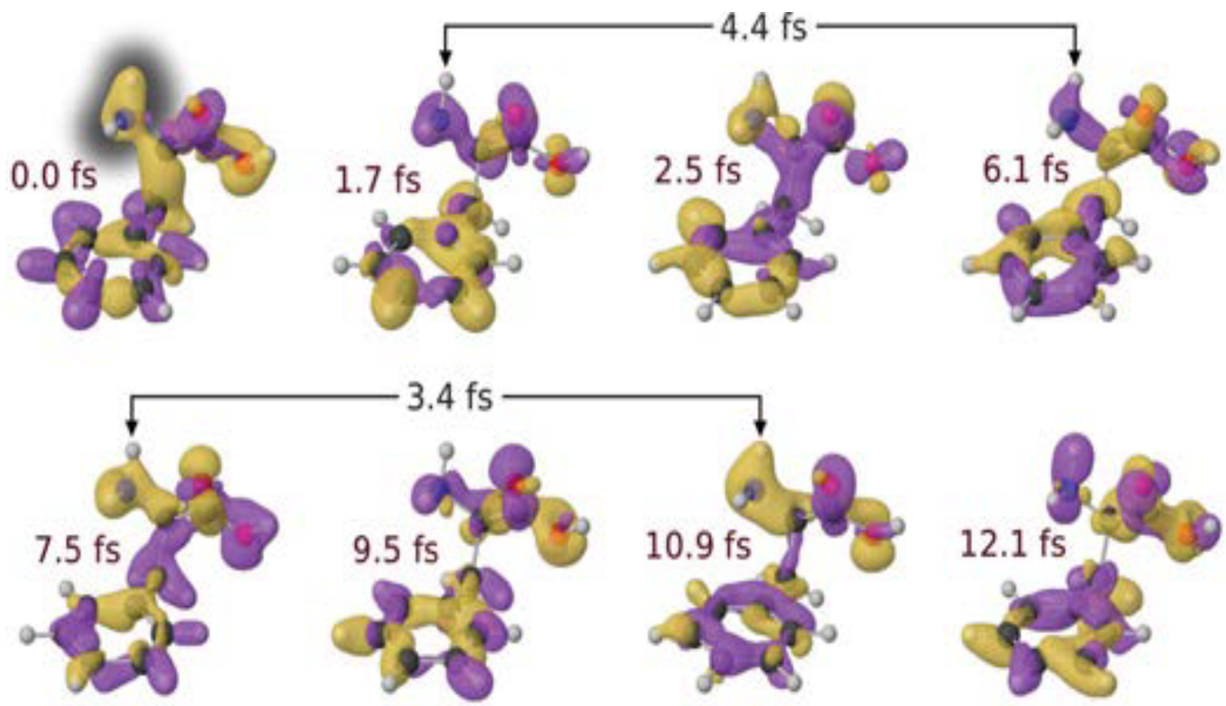
    Conventional modelocked lasers

    High-harmonic generation

    Ultrafast electron diffraction

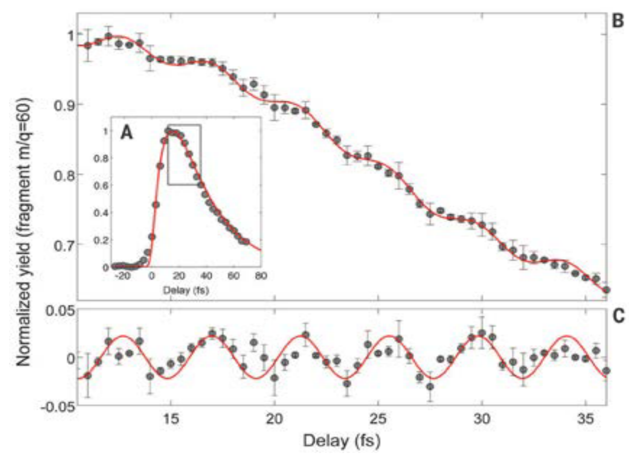



# PRO 1: Electron Motion within a Molecule



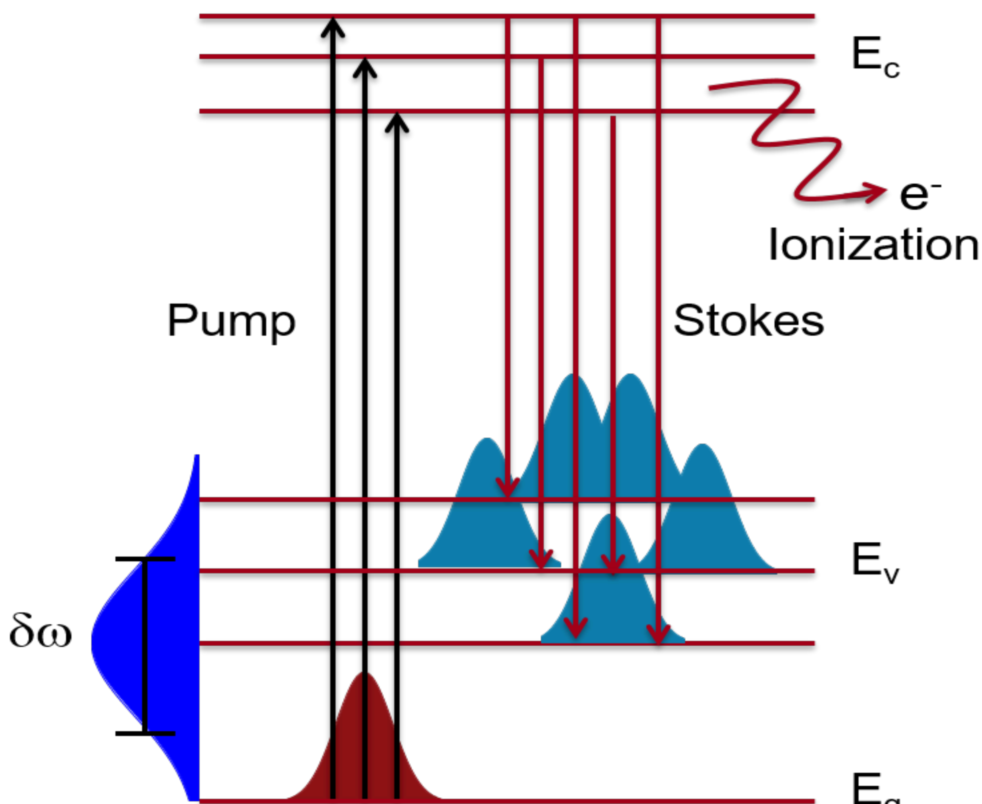

Electron dynamics and coherence strongly influenced by

- electron-electron correlations
- Slight relaxation of positions of atoms




Calagari et al. Science (2014): theory and HHG

# PRO 1: Probing and Controlling Electron Motion within a Molecule




Creating localized electronic excitations through nonlinear x-ray interactions with matter

*Stimulated x-ray Raman scattering*

# PRO 1:  Need for XFELs

**Broad coherent bandwidth** permits simultaneous coherent excitation of the entire valence spectrum of a system, enabling control of the initial state of the system.

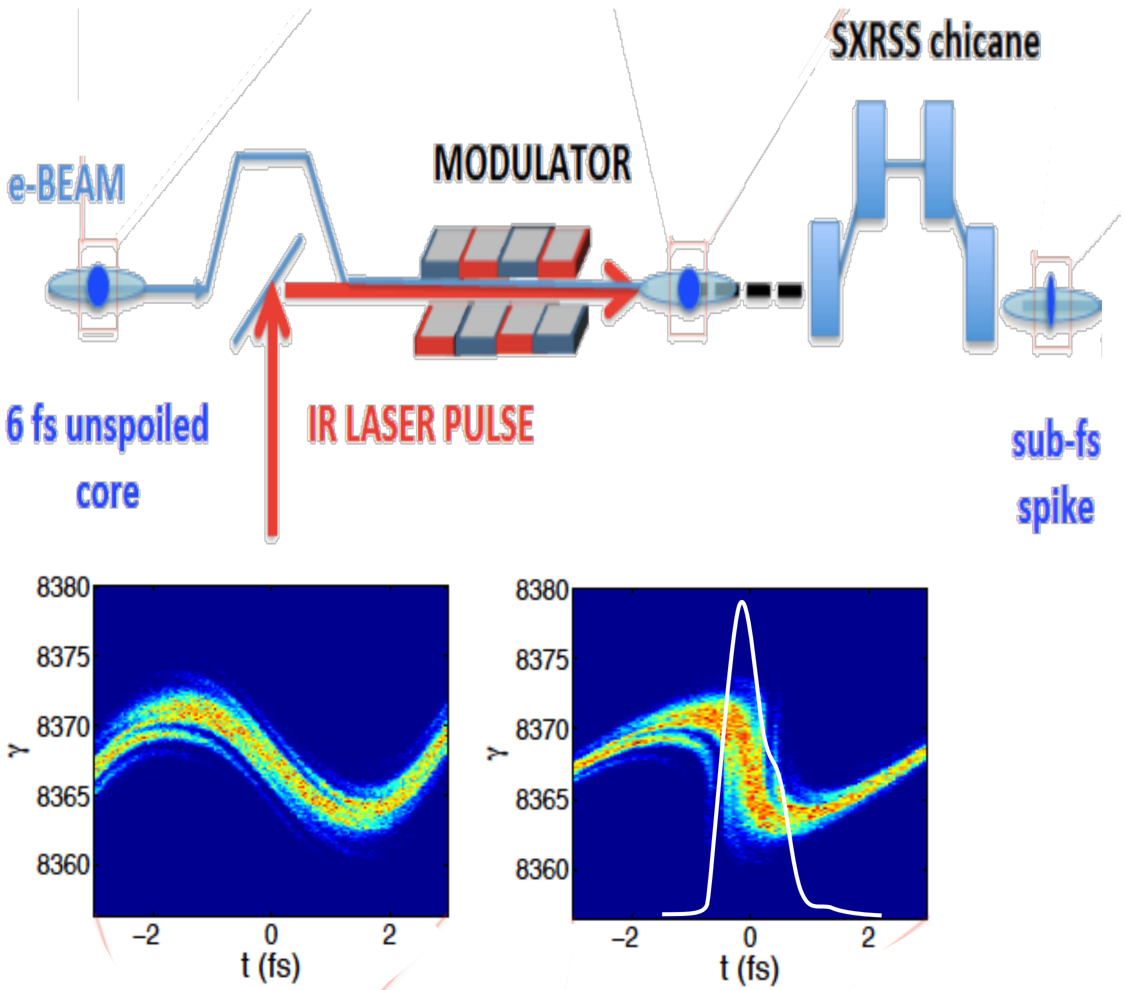
**Attosecond pulse duration** is the critical characteristic for probing below the electronic timescale as defined by the energy spread between electronic states.

**Short wavelength** permits access to atom-specific core-to-valence transitions in molecules.

**High repetition** rate permits low density and subtle effects to be recorded with high fidelity.

**High pulse energy** makes it possible to employ nonlinear spectroscopies.

# Generation of Sub-Femtosecond XFEL Pulses



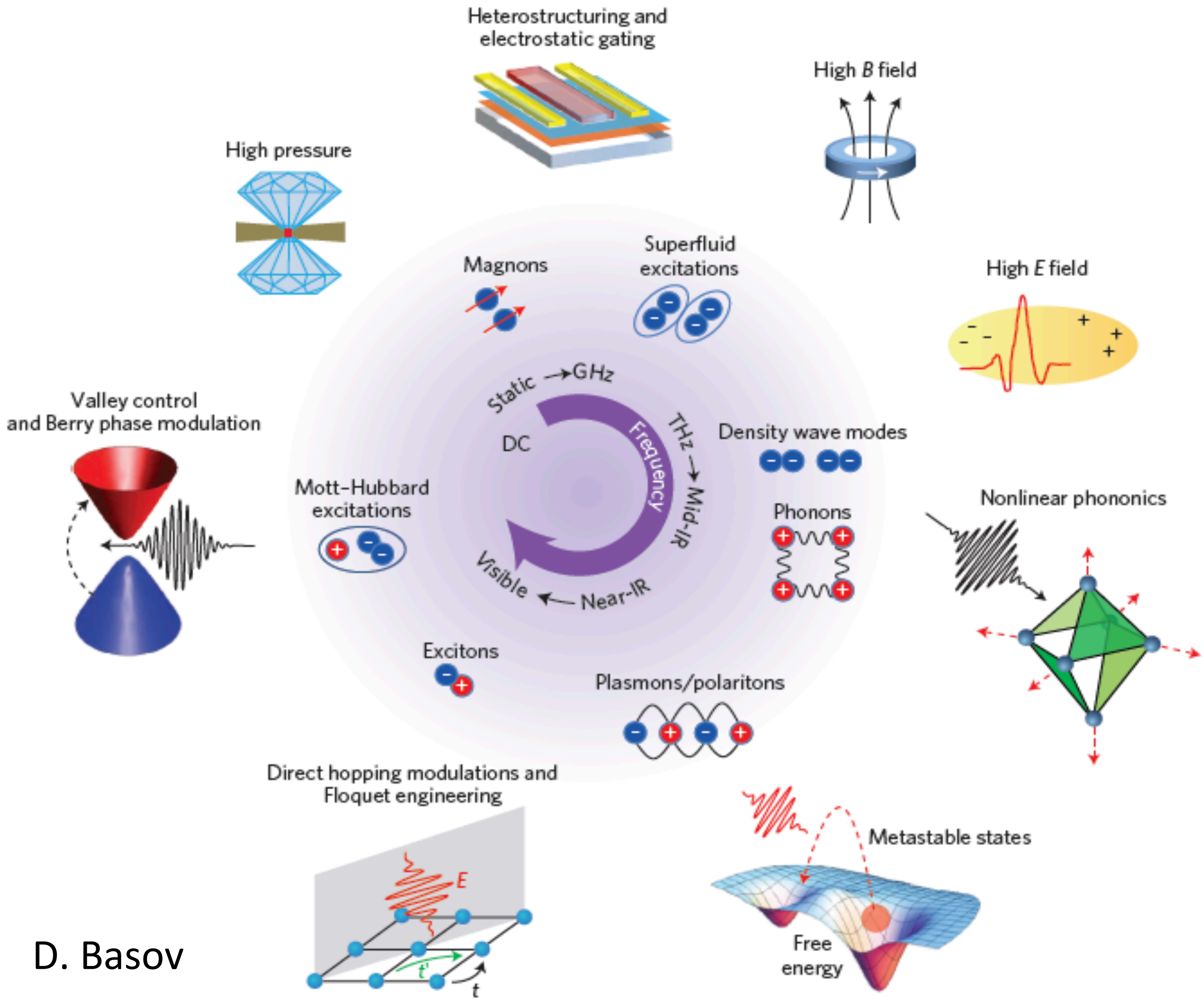

**Production of sub-femtosecond X-ray pulses**

**A. Marinelli - SLAC**

# PRO 1: Probing and Controlling Electron Motion within a Molecule

- How does electronic charge move from atom to atom in a molecular system?
- How do electron-electron interactions and correlations alter this motion?
- How do the atoms rearrange following this electronic motion and, conversely, how does this atomic motion affect the coherent electronic motion?
- Can this coupled and correlated electronic motion be exploited to affect longer-timescale dynamics?

# PRO 2: Discovering Novel Quantum Phases through Coherent Light-Matter Coupling



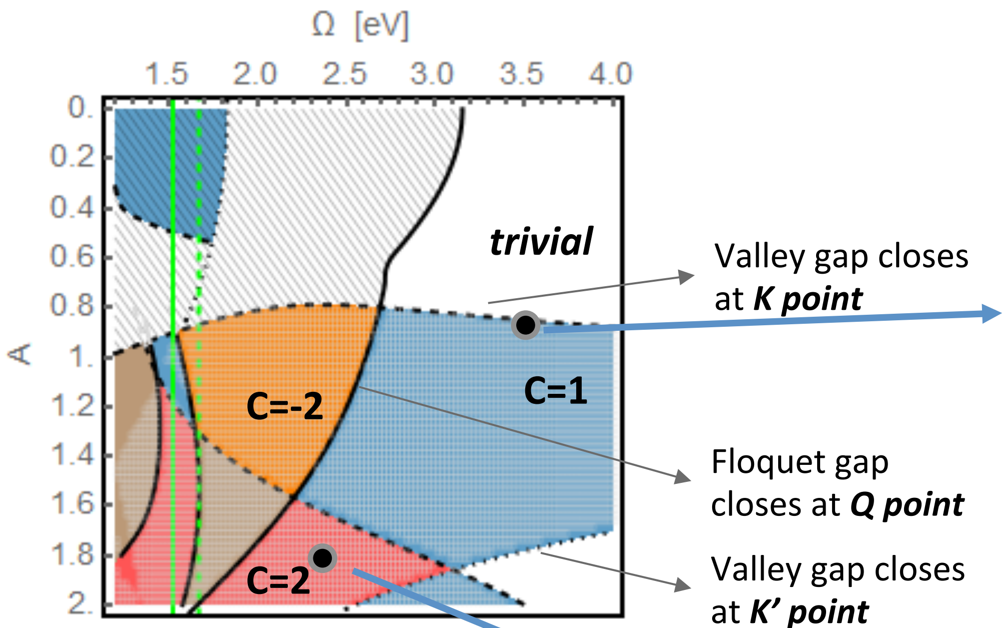

**Hidden photoinduced nonequilibirum states**

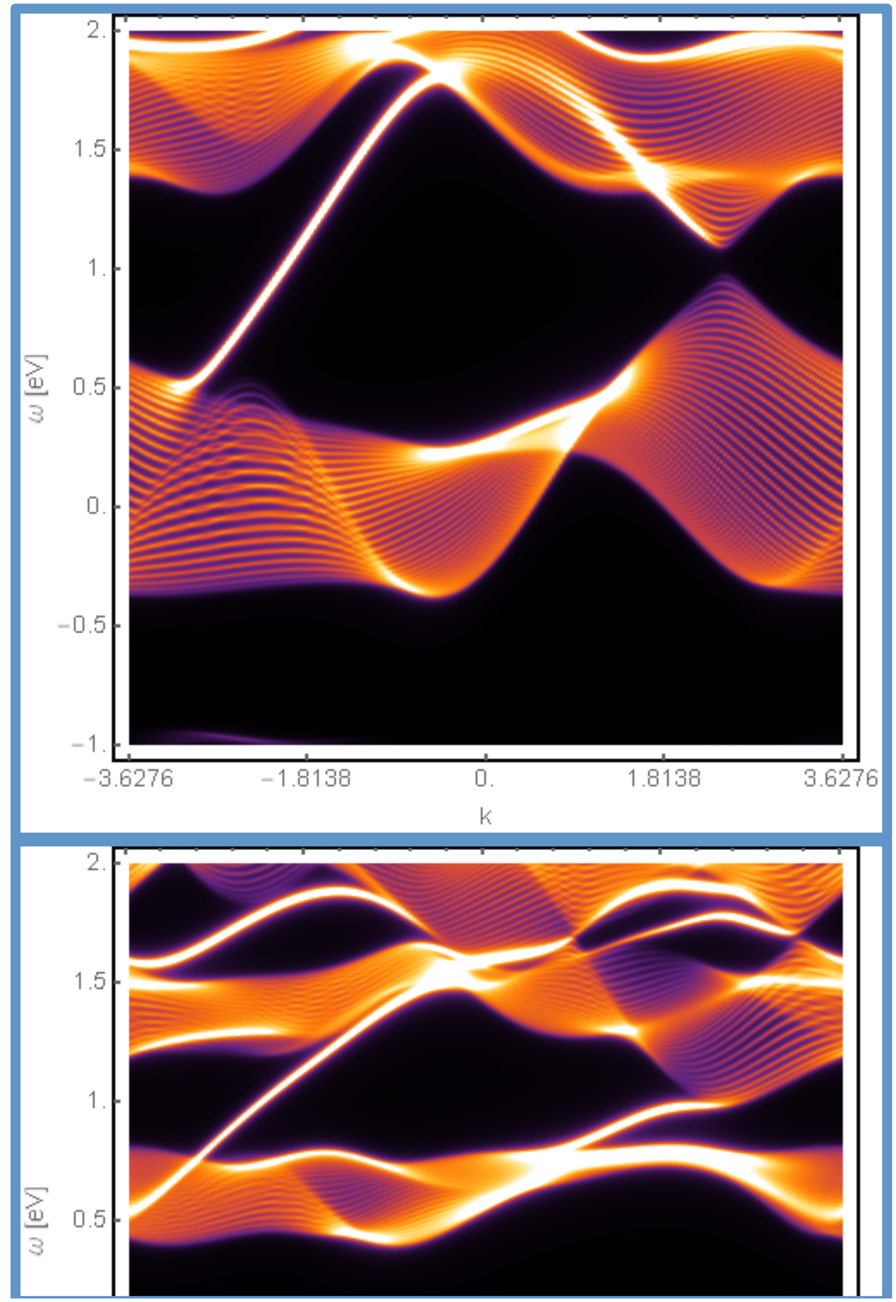
**Transient room-temperature superconductivity**

**Photoinduced Floquet-Bloch topological states of matter**

D. Basov

# PRO 2: Tuning Band Structure with Light

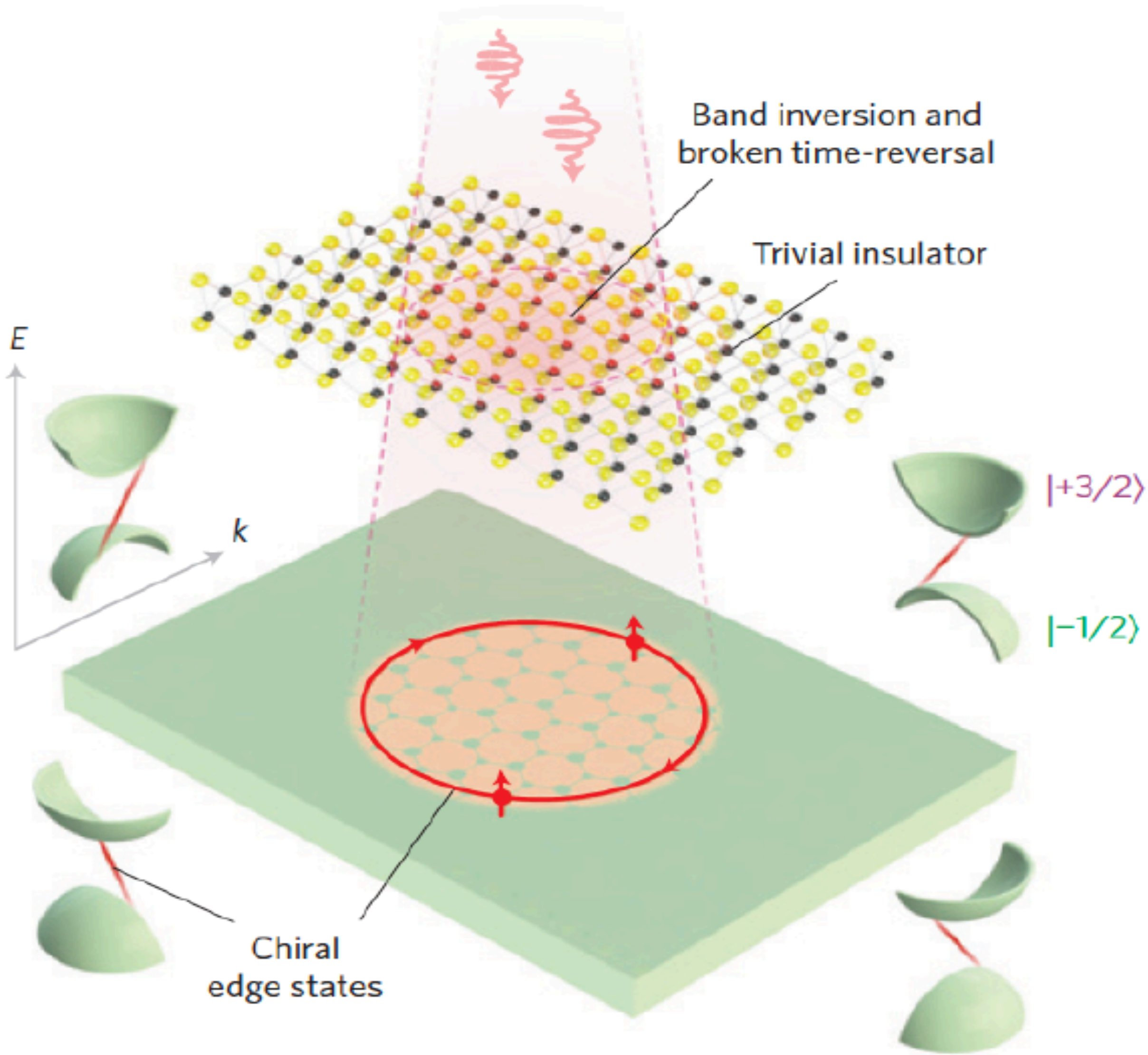



Tuning the band structure of a 2D material with light to create new topological phases and edge states.

(Devereaux group)

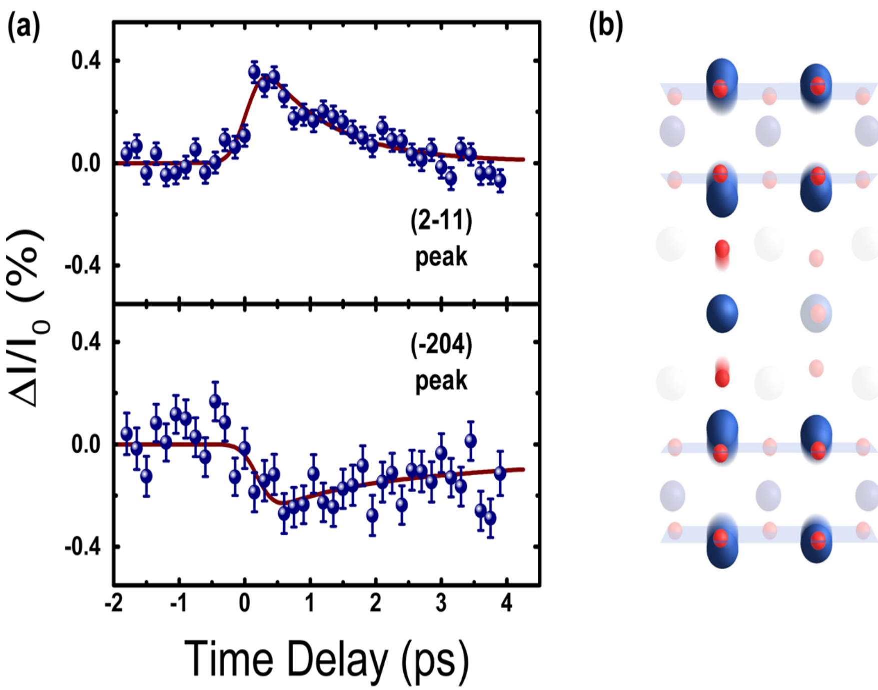
# PRO 2: Tuning Band Structure with Light



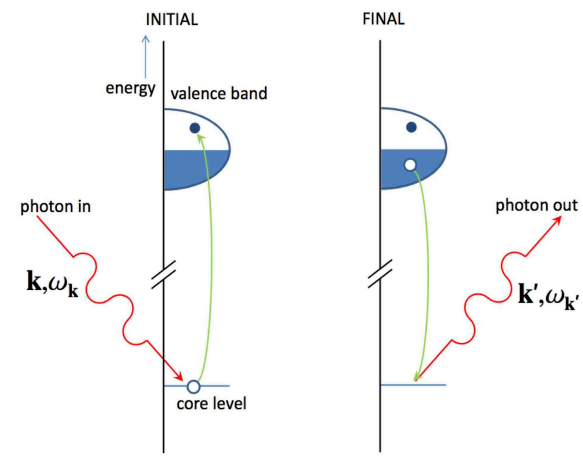

Spatial control of topological phases by light

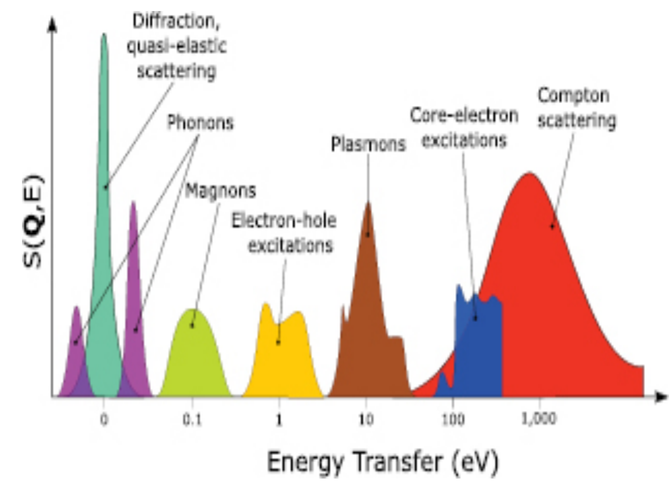
X. Zhang et al.
2-016

# PRO 2:  Probing New Transient Phases




**Time resolved XRD for photoinduced superconductivity (Cavalleri et al)**




**RIXS (x-ray Raman) to probe excitations across the Brillouin zone**

# PRO 2: Discovering Novel Quantum Phases through Coherent Light-Matter Coupling

- How can light be used to create novel quantum phases of matter exhibiting properties that do not exist in equilibrium?
- How can ultrafast optical excitations be utilized to impose quantum coherence or create new topological states?
- How can the lifetime of novel transient quantum phases be extended and manipulated for practical use?

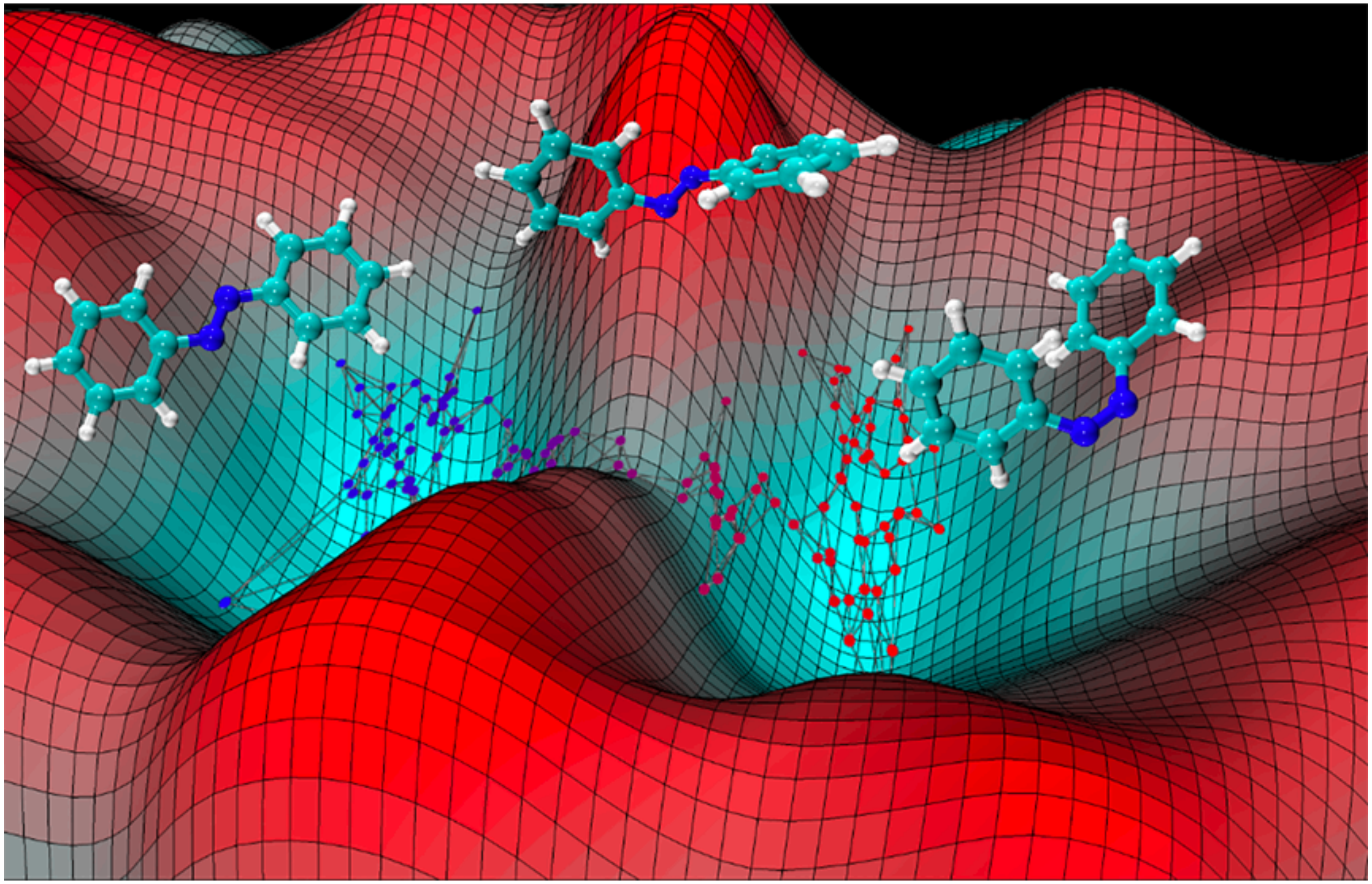
# PRO 3: Capturing Rare Events & Intermediate States in the Transformation of Matter

- Time-resolved measurements almost always use pump-probe techniques. This typically requires
  - Strong perturbation from equilibrium
  - System in which process can be repeated many times to collect stroboscopic data

- What about changes near equilibrium
  - Chemical transformations
  - Phase transformations

# PRO 3: Capturing Rare Events & Intermediate States in the Transformation of Matter

- **Critical events** may are often **fast** and **rare**, as in thermally activated barrier crossing.



# PRO 3: Capturing Rare Events & Intermediate States in the Transformation of Matter

- **New approach: Take advantage of high repetition rate of LCSL-II to accumulate many snapshots of processes**

- **At 1 MHz, we can, in principle, collect $10^{10}$ snapshots in a three hour run.**

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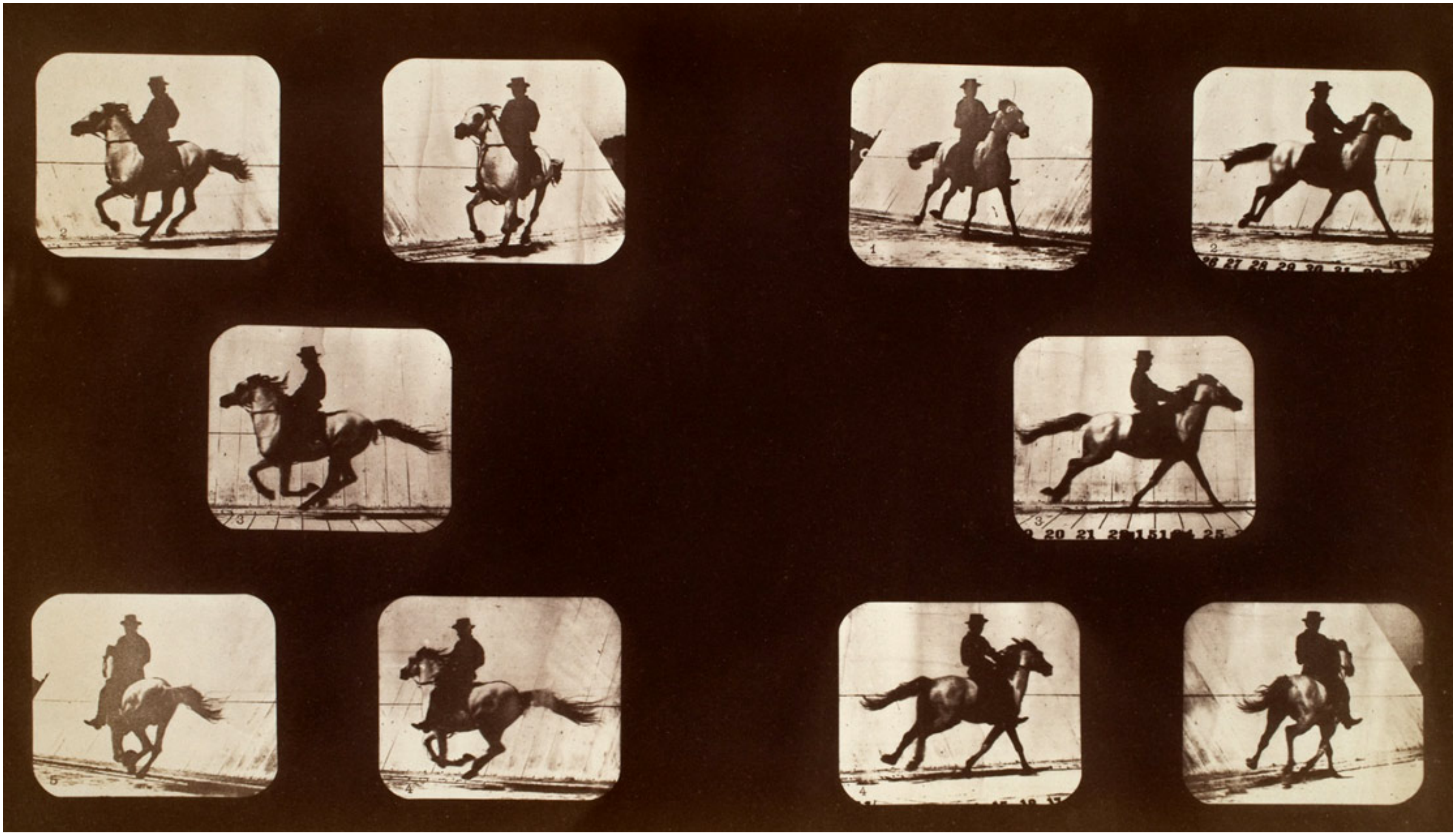


# Looking for Temporal Evolution in Spontaneous Processes

- X-ray photon correlation spectroscopy

  - Down to microseconds using successive frames directly from LCLS

  - Down to femtoseconds using split-pulse techniques

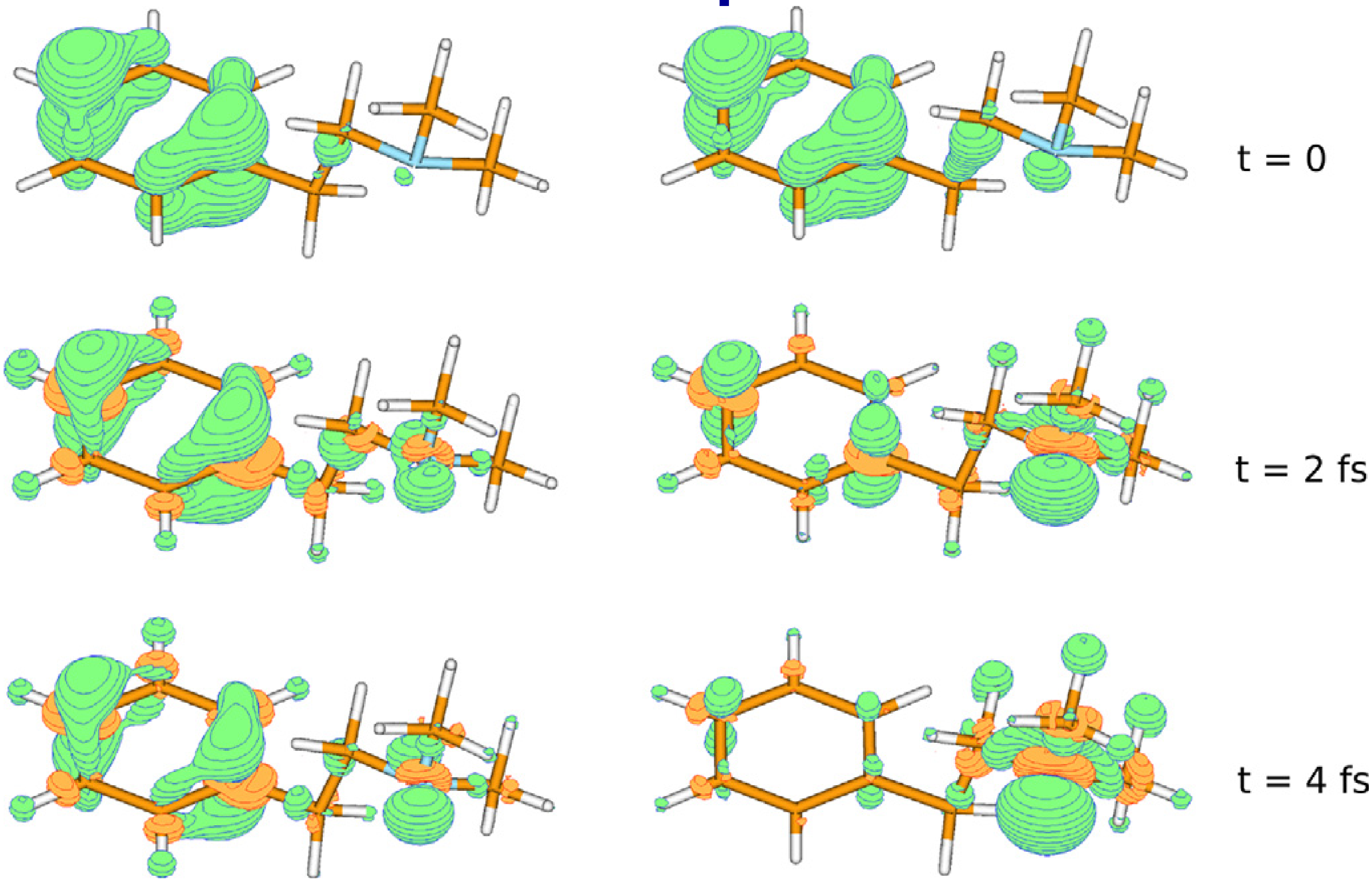
# PRO 3: Capturing Rare Events & Intermediate States in the Transformation of Matter

- How can rare events be captured without the use of external stimulation, as in conventional pump-probe measurements?

- What new theoretical advances and computational methodologies can rapidly translate large experimental datasets into detailed information about transient states and rare events?

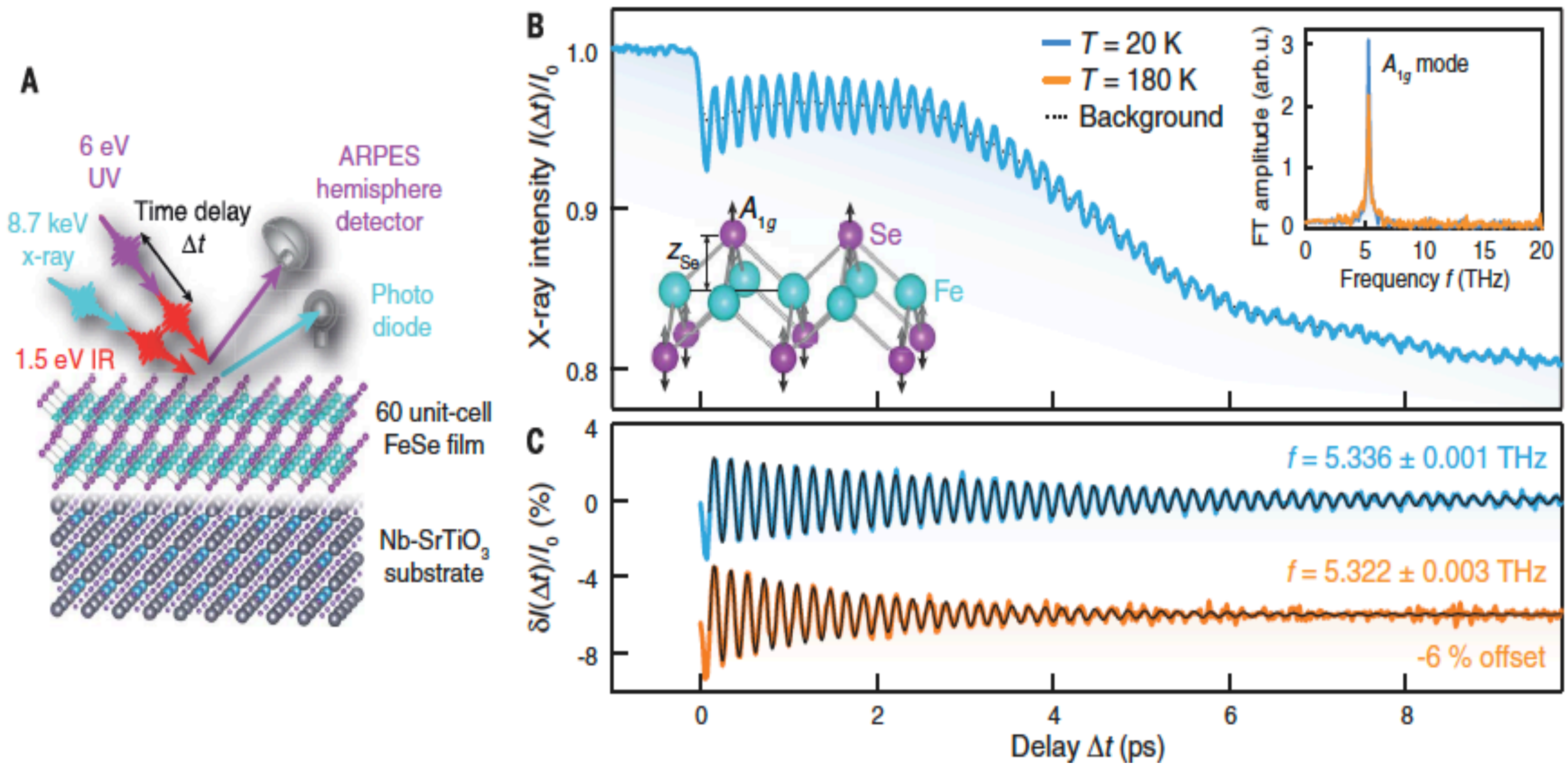
- How can models and theories translate the newly available insights on the role of fluctuating local environments into a predictive understanding and, ultimately, control of macroscopic rates and reaction products?

# Cross-cutting Research:
# Theory of dynamical processes far from equilibrium



# PRO 3: Cross-cutting research
# Multimodal ultrafast measurements

- Dynamic structure function relation accessible by combined measurements of physical and electronic properties/states



# Summary: Roundtable on Opportunities at the Frontiers of XFEL Ultrafast Science

**Fascinating new insight into the fundamental properties of molecules and materials to be revealed with ultrafast measurement capabilities of XFELs**

PRO 1 Probing and controlling electron motion within a molecule

PRO 2 Discovering novel quantum phases through coherent light-matter coupling

PRO 3 Capturing rare events and intermediate states in the transformation of matter

Cross-cutting opportunities in ultrafast x-ray science