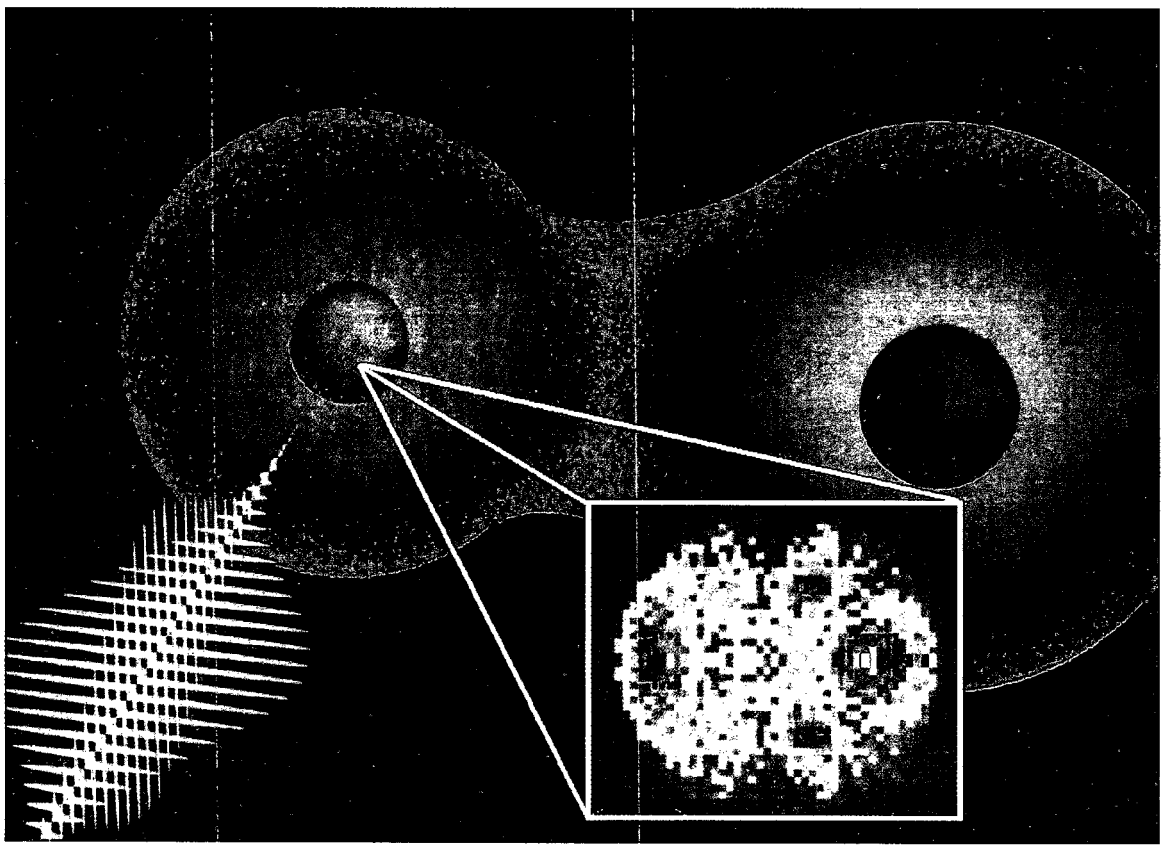
2001 Atomic, Molecular and Optical Physics

Research Meeting



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Chemical Sciences, Geosciences & Biosciences Division

Front Cover:

Schematic illustration of the x-ray excitation of the carbon atom in the CO molecule just above the *K* edge. The inset shows the photoelectron angular distribution for this molecular orientation, which features a pronounced *f*-wave resonance. Experiments performed using momentum imaging techniques at the Advanced Light Source.

Graphic courtesy of Lew Cocke, Kansas State University. For complete reference see: A. Landers, Th. Weber, I. Ali, A. Cassimi, M. Hattass, O. Jagutzki, A. Nauert, T. Osipov, A. Staudte, M. H. Prior, H. Schimdt-Böcking, C. L. Cocke and R. Dörner, *Phys. Rev. Lett.* **87, 013002 (2001).**

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U. S. Department of Energy
Office of Basic Energy Sciences

2001 Meeting of the Atomic, Molecular and Optical Physics Program
Molecular (Chemical) Physics

Sunday, Sept. 30

3:00-6:00 pm	**** Registration ****
6:00 pm	**** Reception (No Host) ****
7:00 pm	**** Dinner ****

Monday, Oct. 1

7:00 am	**** Breakfast ****
8:00 am	*Introductory Remarks* **Eric Rohlfing**, BES/DOE
Session I	Chair: **Eric Rohlfing**
8:15 am	*Intramolecular Dynamics from Frequency Domain Spectra: Patterns, Pattern-Recognition, and Resonance* **Robert W. Field**, Massachusetts Institute of Technology
9:00 am	*Molecules Illuminated from Within: Photoelectron Diffraction from Light Molecules Fixed in Space* **C. L. Cocke**, Kansas State University
9:30 am	*Femtosecond Time-Resolved Photoelectron/Photoion Coincidence Imaging* **Carl Hayden**, Sandia National Labs
10:15 am	**** Break ****
10:30 am	*Ultrafast X-Ray Beamline at the Advanced Light Source* **R. W. Schoenlein**, Lawrence Berkeley National Laboratory
11:00 am	*A Phonon Bragg Switch for X-Rays* **P. H. Bucksbaum**, University of Michigan
11:30 am	*Novel Radiation Sources from Ultra-high-intensity Laser-Plasma Interactions* **Donald Umstadter**, University of Michigan
12:00 pm	*Two-Electron Ionization of Lithium Minus by an Intense, Ultrashort Laser Pulse* **Anthony F. Starace**, University of Nebraska
12:30 pm	**** Lunch ****
5:00 pm	**** Reception (No Host) ****
6:00 pm	**** Dinner ****

Session II Chair: **David Schultz**

7:30 pm *New Electronic Structure Methods for Bond-Breaking*
Martin Head-Gordon, Lawrence Berkeley National Laboratory

8:15 pm *Rovibrational Modes and Classification of Triply Excited States of Atoms*
C. D. Lin, Kansas State University

8:45 pm *Correlation Effects in Two-Electron Transitions*
Jim McGuire, Tulane University

Tuesday, Oct. 2

7:00 am **** Breakfast ****

Session III Chair: **Chris Greene**

8:00 am *Reach Out and Control Something Quantum: Let the Wavepacket do the Thinking*
Herschel Rabitz, Princeton University

8:45 am *Nonlinear Optics in the EUV*
Henry C. Kapteyn, JILA/University of Colorado

9:15 am *Dissociative Recombination of Triatomic Di-Hydrides*
C. R. Vane, Oak Ridge National Laboratory

9:45 am *Ground State Dissociation (GSD) of HD^+*
Itzik Ben-Itzhak, Kansas State University

10:15 am **** Break ****

10:30 am *State-Resolved Probes of Photo-Induced and Thermal Reactions on Ag Surfaces*
Michael G. White, Brookhaven National Laboratory

11:15 am *Mode-Dependent Photoionization Dynamics in Small Molecules*
S. T. Pratt, Argonne National Laboratory

12:00 pm **** Lunch ****

Session IV Chair: **Tom Gallagher**

4:00 pm *State-Resolved Photoionization Dynamics of HCO and DCO*
Edward Grant, Purdue University

4:45 pm *Ionization Probes of Molecular Structure and Chemistry*
Philip M. Johnson, State University of New York at Stony Brook

5:30 pm *Nondipole Effects on Photoelectron Angular Distributions*
S. H. Southworth, Argonne National Laboratory

6:00 pm *Electron/Photon Interactions with Atoms/Ions*
Alfred Z. Msezane, Clark Atlanta University

6:30 pm **** Reception (No Host) ****

7:30 pm **** Dinner ****

Wednesday, Oct. 3

7:00 am	**** Breakfast ****
Session V	Chair: **Ali Belkacem**
8:30 am	*Near-Thermal Collisions of Multi-Charged Ions with Hydrogen and Multielectron Targets* **C. C. Havener**, Oak Ridge National Laboratory
9:00 am	*E1-M1 Transitions: Damping Interferences and Two-Photon Decay* **Robert W. Dunford**, Argonne National Laboratory
9:30 am	*X-Ray Spectroscopy of n=3→, 4→ and 5→1 Transitions in Heliumlike Ar, Ti and Cr* **A. J. Smith**, Morehouse College
10:00 am	**** Break ****
10:15 am	*Measurements and Isoelectronic Predictions of Transition Probabilities in Heavy Complex Atoms* **L. J. Curtis**, University of Toledo
10:45 am	*Measuring a Nuclear Magnetic Octupole Moment* **Carol E. Tanner**, Notre Dame University
11:15 am	*Closing Remarks* **Eric Rohlfing**, BES/DOE
11:30 am	**** Lunch ****
1:00 pm	Tour of the Research Facilities in the Physics Department at the University of Nevada, Reno (Nevada Terawatt Facility, Multicharged Ion Laboratory and Negative Ion Laboratory)

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"Generation and Characterization of Attosecond Pulses"

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I. PROGRAM SCOPE

This proposal exploits the use of nonlinear processes for both the formation and measurement of attosecond pulses. In one sense, the extreme interaction of an atom with an intense light field is used to generate a comb of harmonic radiation. In principle, the attosecond pulse is simply constructed by coherently summing the comb of harmonic bandwidth. However in practice, the coherent construction is strongly dependent on both microscopic and macroscopic amplitudes and phases of the harmonics. Consequently, detail knowledge of these quantities provides the essential roadmap towards attosecond pulse generation. The challenge is twofold, develop a novel source of high harmonic radiation well matched for complete optical characterization and extend the limits of the metrology towards shorter pulse duration (attosecond) and wavelength (x-ray). Meeting the latter challenge will also require the development of novel nonlinear optical mechanisms in atoms and molecules. The proposal outlines a series of investigations aimed at redefining the limits of ultra-short pulse generation and measurement.

II. RECENT PROGRESS

From a theoretical perspective the fundamental concepts of attosecond pulse generation are encouraging, but experimental realization faces many technical challenges. The core problem, which has hampered experimental progress, is simply related to the short wavelengths (<<100 nm) generated by the high harmonic process. Consequently, the primary impediment is that the metrology necessary to characterize short wavelength, ultra-fast pulses simply does not exist. The problem is a double-edged sword; harmonics have low peak power and nonlinearities are weak at short wavelengths. For example, the extreme sensitivity afforded these techniques in the visible region result from the ample availability of materials with large nonlinear susceptibilities, e.g. $\chi^{(2)}$, $\chi^{(3)}$. This is not the case below 200nm. One thrust of this proposal is the development of novel metrological approaches at short wavelength. However by exploiting the scalability of the intense laser-atom interaction, the relevant physics of attosecond pulse generation can be studied in great detail by shifting the high harmonic emission to a convenient spectral region. Thus, an intense, long wavelength mid-infrared fundamental field (3-4 μm) is used to produce high harmonic light in the visible and near-UV wavelength range. In this region, sensitive and sophisticated metrology does exist. Consequently, FROG and SPIDER methods can be immediately applied for the complete phase and amplitude characterization. A detailed understanding of the coherence properties of high harmonic

light is a crucial prerequisite for producing attosecond pulses and a roadmap towards x-rays.

In preliminary studies already performed as part of a previously funded BNL Laboratory Directed Research and Development proposal for FY1999 and FY2000, we have demonstrated the essential elements of a mid-infrared pumped harmonic source. In the experiment, high harmonic light from an alkali metal vapor target is produced by a tunable mid-infrared (3-4 μm) pulse with a peak intensity of 1 TW/cm^2. We have shown that high harmonic light is generated in sufficient quantity for applying complete spectral analysis throughout the near-ir/visible/near-uv region of the electromagnetic spectrum. Furthermore, the interaction of an alkali atom with the mid-infrared laser shows similar physics to harmonics generated from more tightly bound electrons (higher ionization potential atoms) at shorter wavelength. The findings of these studies are discussed in Refs. [1] and [2].

In FY2001, we have focused on developing and applying intensity autocorrelation for the temporal characterization of the harmonic light. An achromatic background-free second harmonic (SH) autocorrelator has been built and successfully applied to harmonics orders 5 through 11. Figure 1 shows typical SH autocorrelation traces for the 3.4 μm fundamental pulse (MIR), fifth (H5) and seventh (H7) harmonics. The result show a dramatic narrowing of the light with increasing harmonic order and is consistent with the predictions of perturbative scaling. However, we observe that the harmonic pulse widths are strongly dependent on the choice of experimental parameters, i.e. intensity, density. Understanding this behavior is still under active investigation but the findings suggest a realm of variation that can be utilized for obtaining optimal control of the output pulse.

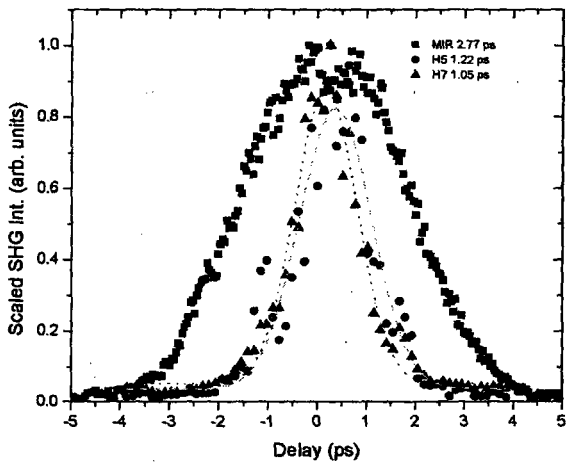

Figure 1: Intensity autocorrelation of the 3.4 μm fundamental, fifth and seventh harmonic light from cesium atoms. The pulse durations are indicated in the legend.

The influence of the atomic response on harmonic yield is also under examination. In this experiment, a cw-laser field is used to resonantly couple the ground state to the first

excited state (nS→nP transition) in the alkali atom. The harmonics generated by the interaction with the intense mid-infrared pulse with the incoherent target is then studied. Under certain conditions with the coupling laser present, enhancements by a factor of thirty are observed in the harmonic yield and the increase is dependant on the harmonic order. We are currently trying to understand how the change in harmonic yield is linked to the increased polarizability of the excited state. The obvious and intriguing extension of this study is to examine the intense laser-atom interaction for a "coherent" target.

III. Future Plans

The work on generation of high-harmonic radiation will continue at Brookhaven, supported by direct theoretical guidance and modeling at Livermore. In parallel with this effort, the Rochester team will extend current measurement techniques in ways that are easily adapted to the durations and spectral ranges expected in 4th Generation Light Sources. These developments will be supported by the BNL group's extensive experimental experience in photoionization. The techniques developed at Rochester will be implemented on the BNL source in the visible/near-UV, and together with the Dr. Agostini, on next generation tabletop sources in the VUV and x-ray regions. Furthermore, the Deep Ultra-Violet Free Electron Laser project under construction at BNL under the auspices of the National Synchrotron Light Source (NSLS) is scheduled for operation in the VUV regime in FY2002. This facility will provide a unique test-bed for exploring the dynamic interface between the metrology and 4th Generation Light Sources.

We have now established that our approach results in a scaled laser-atom interaction equivalent to more commonly studied inert gases interacting with visible light and that advance metrology is applicable to these harmonics. The next set of studies will focus on complete optical characterization of the low-order harmonics using second harmonic Frequency Resolved Optical Gating. The studies can begin to understand and control the entanglement of microscopic and macroscopic phases. This will be aided by our ability to perform experiments over a large dynamic range. Two main technical changes will be implemented over the next year to enhance our capabilities for performing these studies. First, the mid-infrared energy will be increased by a factor of five by the addition of a second optical amplifier. Second, the pulse duration of the mid-infrared light will be shortened into the femtosecond regime. In principle, a 1-2 cycle pulse should be obtainable with our apparatus. These changes should allow for higher harmonic energies, shorter pulse duration and investigation into more tightly bound systems.

The scaled alkali-mid-infrared interaction will provide a prototype for studying the influence of atomic "coherence" on the harmonic process. All harmonic experiments are performed on atoms initially in their ground state. However, the harmonic process is coherent and sensitive to the preparation of the initial state. Calculations [3,4] have shown that an initial "coherent" superposition state can dramatically alter the high harmonic distribution. Current harmonic experiments on inert gases are hampered by the large energy separation (VUV photons) between the ground and first excited state. However as we shown in FY2001, the alkali atoms have easily accessible transitions. In

fact, the D-lines (*ns*→*np* transition) of K, Rb and Cs are all within the titanium sapphire bandwidth and easily saturated while higher excited states are accessible to optical parametric amplifiers. We are currently examining a variety of approaches for preparing a superposition state between the *ns* and *np* state. We will first investigate the frequency spectrum and ultimately characterize the phase and amplitude of the harmonic field. Furthermore, these systems are amenable to our theoretical methods. Question such as; whether a resonance can develop as the atomic transition becomes commensurate with the mid-infrared field and how does the harmonic phase change with the initial coherence, can all be addressed. If, by virtue of our studies, the atomic coherence develops into an important tool for harmonic production, it is conceivable to apply these schemes in inert gases.

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3. F. I. Gauthey *et al.*, *"Role of Initial Coherence in the Generation of Harmonics and Sidebands from a Strongly Driven Two-Level Atom"*, Phys. Rev. A **52**, 525 (1995).
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Foreword

This volume summarizes the scientific content of the 2001 Research Meeting of the Atomic, Molecular and Optical Physics (AMOP) Program sponsored by the U. S. Department of Energy (DOE), Office of Basic Energy Sciences (BES). This meeting is held annually for the DOE laboratory and university principal investigators within the BES AMOP Program in order to facilitate scientific interchange among the PIs and to promote a sense of program identity. For the past three years, the meeting has included significant participation from scientists outside of the BES AMOP Program and has had a specific topical focus. This year's topic is rather loosely defined as "Molecular Physics" and our external invited speakers are all supported through the BES Chemical Physics Program. The fields of AMO and Chemical Physics are closely related in many ways and enhanced interaction between the two can only be beneficial to both.

The BES AMOP Program has grown modestly in recent years due to successful competition in BES research initiatives, including Complex and Collective Phenomena in FY1999, Novel X-Ray Light Sources in FY2000, and Nanoscale Science, Engineering and Technology in FY2001. This growth has helped to spur the evolution of the program from one of purely atomic physics to a more diverse portfolio that includes the interactions of intense laser fields with atoms and molecules, coherent control of quantum processes, development and application of novel x-ray light sources, ultracold collisional interactions, and quantum condensates. At the same time, the traditional role of the program continues through support of AMO science at the DOE synchrotron facilities and via ongoing projects involving highly charged atomic ions. The BES AMOP Program is characterized by the close coupling between experiment and theory; approximately 70% of the PIs are experimentalists and 30% are theorists. The quality of the science in the program has been and will continue to be high; roughly 60% of the PIs are fellows of the American Physical Society.

The AMOP Program is viewed within BES as one that provides fundamental physical insight and thus generally underpins more obviously energy-relevant programs in BES and the Office of Science (such as Fusion Energy Sciences), current and future BES facilities that probe matter with photons, electrons or heavy ions, and applied efforts in high- and low-temperature plasma modeling. The BES AMOP Program will continue to evolve in response to changes in the landscape of AMO science while maintaining solid, justifiable connections to the BES mission.

I gratefully acknowledge the speakers, particularly those not supported by the BES AMOP program, for their investment of time and effort and for their willingness to share their ideas with the meeting participants. Thanks also to the staff of the Oak Ridge Institute of Science and Education and the Granlibakken Conference Center for assisting with logistical aspects of the meeting.

Eric Rohlfing, Program Manager
Atomic, Molecular and Optical Physics
Chemical Sciences, Geosciences and Biosciences Division
Office of Basic Energy Sciences
August, 2001

Agenda

U. S. Department of Energy
Office of Basic Energy Sciences

2001 Meeting of the Atomic, Molecular and Optical Physics Program
Molecular (Chemical) Physics

Sunday, Sept. 30

3:00-6:00 pm	**** Registration ****
6:00 pm	**** Reception (No Host) ****
7:00 pm	**** Dinner ****

Monday, Oct. 1

7:00 am	**** Breakfast ****
8:00 am	*Introductory Remarks* **Eric Rohlfing**, BES/DOE
Session I	Chair: **Eric Rohlfing**
8:15 am	*Intramolecular Dynamics from Frequency Domain Spectra: Patterns, Pattern-Recognition, and Resonance* **Robert W. Field**, Massachusetts Institute of Technology
9:00 am	*Molecules Illuminated from Within: Photoelectron Diffraction from Light Molecules Fixed in Space* **C. L. Cocke**, Kansas State University
9:30 am	*Femtosecond Time-Resolved Photoelectron/Photoion Coincidence Imaging* **Carl Hayden**, Sandia National Labs
10:15 am	**** Break ****
10:30 am	*Ultrafast X-Ray Beamline at the Advanced Light Source* **R. W. Schoenlein**, Lawrence Berkeley National Laboratory
11:00 am	*A Phonon Bragg Switch for X-Rays* **P. H. Bucksbaum**, University of Michigan
11:30 am	*Novel Radiation Sources from Ultra-high-intensity Laser-Plasma Interactions* **Donald Umstadter**, University of Michigan
12:00 pm	*Two-Electron Ionization of Lithium Minus by an Intense, Ultrashort Laser Pulse* **Anthony F. Starace**, University of Nebraska
12:30 pm	**** Lunch ****
5:00 pm	**** Reception (No Host) ****
6:00 pm	**** Dinner ****

Session II Chair: **David Schultz**

7:30 pm *New Electronic Structure Methods for Bond-Breaking*
Martin Head-Gordon, Lawrence Berkeley National Laboratory

8:15 pm *Rovibrational Modes and Classification of Triply Excited States of Atoms*
C. D. Lin, Kansas State University

8:45 pm *Correlation Effects in Two-Electron Transitions*
Jim McGuire, Tulane University

Tuesday, Oct. 2

7:00 am **** Breakfast ****

Session III Chair: **Chris Greene**

8:00 am *Reach Out and Control Something Quantum: Let the Wavepacket do the Thinking*
Herschel Rabitz, Princeton University

8:45 am *Nonlinear Optics in the EUV*
Henry C. Kapteyn, JILA/University of Colorado

9:15 am *Investigation and Control of Strong-Field Processes in Atoms, Molecules and Clusters*
Robert R. Jones, University of Virginia

9:45 am *Ground State Dissociation (GSD) of HD^+*
Itzik Ben-Itzhak, Kansas State University

10:15 am **** Break ****

10:30 am *Single Molecule Vibrations and Spatially Resolved Chemistry*
Wilson Ho, University of California, Irvine

11:15 am *State-Resolved Probes of Photo-Induced and Thermal Reactions on Ag Surfaces*
Michael G. White, Brookhaven National Laboratory

12:00 pm **** Lunch ****

Session IV Chair: **Tom Gallagher**

4:00 pm *State-Resolved Photoionization Dynamics of HCO and DCO*
Edward Grant, Purdue University

4:45 pm *Ionization Probes of Molecular Structure and Chemistry*
Philip M. Johnson, State University of New York at Stony Brook

5:30 pm *Nondipole Effects on Photoelectron Angular Distributions*
S. H. Southworth, Argonne National Laboratory

6:00 pm *Electron/Photon Interactions with Atoms/Ions*
Alfred Z. Msezane, Clark Atlanta University

6:30 pm **** Reception (No Host) ****

7:30 pm **** Dinner ****

Wednesday, Oct. 3

7:00 am	**** Breakfast ****
Session V	Chair: **Ali Belkacem**
8:00 am	*Mode-Dependent Photoionization Dynamics in Small Molecules* **S. T. Pratt**, Argonne National Laboratory
8:45 am	*Dissociative Recombination of Triatomic Di-Hydrides* **C. R. Vane**, Oak Ridge National Laboratory
9:15 am	*Near-Thermal Collisions of Multi-Charged Ions with Hydrogen and Multielectron Targets* **C. C. Havener**, Oak Ridge National Laboratory
9:45 am	*E1-M1 Transitions: Damping Interferences and Two-Photon Decay* **Robert W. Dunford**, Argonne National Laboratory
10:15 am	**** Break ****
10:30 am	*X-Ray Spectroscopy of n=3→ , 4→ , and 5→ 1 Transitions in Heliumlike Ar, Ti and Cr* **A. J. Smith**, Morehouse College
11:00 am	*Measurements and Isoelectronic Predictions of Transition Probabilities in Heavy Complex Atoms* **L. J. Curtis**, University of Toledo
11:30 am	*Measuring a Nuclear Magnetic Octupole Moment* **Carol E. Tanner**, Notre Dame University
12:00 pm	*Closing Remarks* **Eric Rohlfing**, BES/DOE
12:15 pm	**** Lunch ****
2:00 pm	Tour of the Research Facilities in the Physics Department at the University of Nevada, Reno (Nevada Terawatt Facility, Multicharged Ion Laboratory and Negative Ion Laboratory)

Table of Contents

Invited Presentations (Ordered By Agenda)

Research Summaries (Multi-PI Programs, Alphabetical By Institution)

Research Summaries (Single-PI Grants, Alphabetical By PI)

Invited Presentations

(ordered by agenda)

Intramolecular Dynamics from Frequency Domain Spectra: Patterns, Pattern-Recognition, and Resonance

Robert W. Field, Department of Chemistry, Massachusetts Institute of Technology

DE-FG02-87ER13671

Each molecular eigenstate, taken by itself, is stationary. There is no intramolecular motion. Eigenstates, taken as a group, encode motion, but in an extremely opaque format. At high internal vibrational excitation of a small polyatomic molecule, the classical trajectory motions encoded by eigenstates are mostly chaotic. No simple, quasiperiodic ball-and-spring picture exists for such motions. Nothing is conserved that is not guaranteed to be conserved by some fundamental molecular symmetry. However, when we ask questions about "mechanisms" of intramolecular energy flow, we are interested in the early-time behavior of initially localized excitations. We are concerned with what is conserved *for a short time*. Any conserved dynamical quantity gives rise to a pattern that is multiply repeated in a spectrum or in a related set of spectra, provided that the frequency resolution ($\delta\nu$) of the spectrum is not so high that the reciprocal of the resolution ($1/2\pi\delta\nu$) is a time longer than the time that the dynamical quantity of interest is conserved. So we are interested in *early-time* dynamics and *low-resolution* spectra. But at low resolution, the dynamically interesting patterns overlap each other and it is difficult to disentangle them. EXtended spectral Cross-Correlation (XCC) is a pattern recognition technique, which is capable of recovering such patterns from spectra without prior knowledge of their nature or number. Once the patterns are revealed, it is possible to show that the early time energy flow mechanism is dominated by "resonance", which is the transfer of energy between two internal motions that have nearly identical classical frequencies. Energy resonance is at least as important as magnitudes of coupling constants. An understanding of intramolecular dynamics begins with the identification of the small number of dynamically important resonances. *This is a fundamental departure from the traditional approach to the spectra of small polyatomic molecules.*

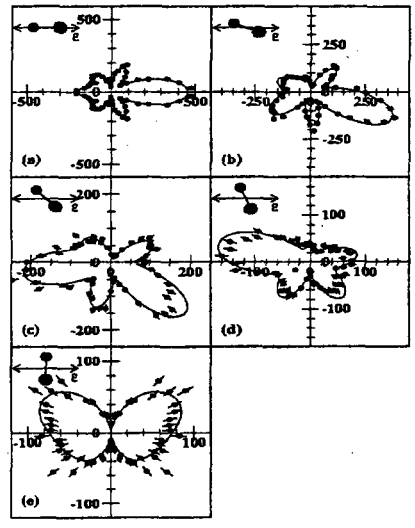
These ideas are illustrated by the 7 cm^{-1} resolution Dispersed Fluorescence (DF) spectra of acetylene. The approximately conserved quantities are polyad quantum numbers. Polyads are the spectrally overlapping repeated patterns that are disentangled by XCC. All polyads are represented by a quantum mechanical effective Hamiltonian, $\mathbf{H}^{\mathbf{eff}}$, and the matrices for different polyads are related by Harmonic Oscillator selection and scaling rules. The dimensionality of this quantum $\mathbf{H}^{\mathbf{eff}}$ is lower, by the number of polyad quantum numbers (3 in the case of acetylene) than the 3N–3 nuclear degrees of freedom. The quantum $\mathbf{H}^{\mathbf{eff}}$ is converted to a reduced dimensionality classical Hamiltonian, which permits the structure of phase space to be illustrated by surfaces of section. Almost as if by magic, extremely stable, large amplitude, local-bender motions appear directly along the minimum energy acetylene←>vinylidene isomerization path.

Molecules illuminated from within: photoelectron diffraction from light molecules fixed in space *

C.L.Cocke, Physics Department, J.R.Macdonald Laboratory, Kansas State University,Manhattan,KS 66506, cocke@phys.ksu.edu

The goals of this aspect of the JRM program are to explore the dynamics of photoelectron emission from and the instantaneous structure of small molecules. We measure the diffraction patterns of photoelectrons issuing from core photoionization of fixed-in-space light molecules. The electron emanates from a known site within the molecule and "illuminates" the structure from within. The diffraction patterns are shaped by the interaction of the electron with the instantaneous potential presented by the remaining nuclei and electrons of the molecule. The data provide an extremely comprehensive picture of the diffraction patterns, which in turn probe the overall charge density distribution within the molecule at the time of photoelectron emission. The experiments also provide information on the subsequent modes of breakup of the doubly-charged molecule. The experiments are carried out at the Advanced Light Source (ALS) on beamlines 4.0 and 9.3.2. The K shells of the target molecules (CO, N_2, C_2H_n) are photoionized slightly above threshold. The molecule subsequently undergoes Auger decay, and the doubly charged molecule dissociates. COLTRIMS techniques are used to measure the coincident photoelectron and two charged molecular fragments. The charged products are projected by an electric field onto the surfaces of time- and position-sensitive multichannelplate detectors, and their full coincident momentum vectors are reconstructed on an event-by-event basis. The molecule is "fixed in space" on an *a posteriori* basis, assuming a radial two-body dissociation. Further information on this project is provided in the research summaries.

A representative result for CO is shown in the figure to the right, where angular distributions of photoelectrons are plotted in the laboratory-fixed coordinate system with the linear polarization of the light parallel to the x-axis, and for various orientations of the molecule with respect to the polarization vector. The photon energy is 10.2 eV above the C-K edge, near the maximum of the well-known f-wave sigma shape resonance for this molecule. The f-wave character of the distribution is evident in the upper left-hand figure. The resonance is clearly weaker for the lowest figure (molecule perpendicular to polarization).



*Work carried out in collaboration with A.Landers (Western Mich. Univ.),R.Doerner, Th.Weber,M.Hattass,O.Jagutzki,A.Nauert,A.Staudte,H.Schmidt-Boecking(U.Frankfurt), M.H.Prior(LBL),A.Cassimi(CIRIL),T.Osipov, I.Ali(KSU).

Femtosecond time-resolved photoelectron/photoion coincidence imaging

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Sandia National Laboratories
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Femtosecond time-resolved photoionization imaging provides detailed probes of fast photodissociation dynamics. The time-resolved coincidence imaging approach generates three-dimensional energy- and angle-resolved images of photofragment ions and the corresponding photoelectrons. The resulting time-resolved, correlated distributions of photofragment and photoelectron energy can be used to determine the mechanisms of complex dissociation processes and to probe subsequent evolution of the products. Vector correlations between the photofragment and photoelectron recoil velocity vectors can be analyzed to yield photoelectron angular distributions referenced to the dissociation axis of the molecule. Changes in these molecular-frame photoelectron angular distributions directly reflect the time-dependent separation and reorientation of the dissociating photofragments. The capability for femtosecond time-resolved photoelectron imaging can also be used to probe fast intramolecular processes. Three-dimensional imaging provides complete energy and angular distributions of the photoelectrons. Photoelectron angular distributions (PADs) are sensitive to the nature of the molecular orbital that is ionized, the geometry and orientation of the molecule as the electron departs, and the dynamics of the photoionization process. Unfortunately, much of the information is lost when the PAD is measured from a randomly oriented sample of molecules and hence PADs have not been extensively studied. In femtosecond time-resolved experiments the time scale of the experiment is often short compared to molecular rotation times so it is possible to measure PADs from aligned or oriented molecules. Intramolecular processes, such as fast internal conversion, can be probed by the changes they produce in both the photoelectron energy and angular distributions.

Ultrafast X-ray Beamline at the Advanced Light Source

R.W. Schoenlein[a], A. Cavalleri[a], H.H.W. Chong[b], T.E. Glover[c]
P.A. Heimann[c], C.V. Shank[a], A.A. Zholents[d], and M.S. Zolotorev[d]

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Modern synchrotrons providing high-brightness, tunable x-ray beams, combined with techniques such as x-ray diffraction, extended x-ray absorption fine structure (EXAFS) and many others have driven rapid advances in our understanding of the 'static' structure of condensed-matter on the atomic scale. Of course the structure of matter is not static, and an important emerging area of research is the investigation of structural dynamics on the fundamental scale of a vibrational period, ~100 fs. This is the limiting time scale for structural changes that determine the course of phase transitions in solids, the kinetic pathways of chemical reactions, and even the function and efficiency of biological processes. A significant limitation of synchrotron sources is the pulse duration (typically >30 ps) as determined by the duration of the stored electron bunch. In contrast, femtosecond laser technology now enables the measurement of dynamic processes on time scales shorter than 10 fs. However, femtosecond lasers operating in the UV to IR region probe the extended electronic states of condensed matter which provide only indirect information about the underlying atomic structure.

This new research program is based on a unique bend-magnet beamline at the Advanced Light Source (ALS) designed to provide 100 fs x-ray pulses for time-resolved research. The initial focus of the program is on the development, characterization, and operation of the beamline, building on techniques from recent proof-of-principle experiments [1]. This program further includes the development of scientific applications and measurement techniques using femtosecond x-ray pulses, and provides scientific support in time-resolved x-ray science for beamline users.

Our approach for generating ultrashort pulses from a synchrotron relies on creating femtosecond time-structure on a long electron bunch by using a femtosecond laser pulse to modulate the energy of an ultrashort slice of the bunch [2]. In initial demonstration experiments, synchrotron pulses of ~300 fs duration were directly measured from ALS bend-magnet beamline 6.3.2 [1, 3].

Figure 1 illustrates the modulation and generation scheme. A femtosecond laser pulse co-propagates with the stored electron bunch through a wiggler (Fig. 1A). The field of the laser pulse modulates the energy of the underlying electrons as they traverse the wiggler. The optimal interaction occurs when the central wavelength of the spontaneous emission from an electron passing through the wiggler matches the laser wavelength (FEL resonance condition) [2]. In addition, the transverse mode of the laser beam must match the transverse mode of the spontaneous radiation from the electron passing through the wiggler, and the laser spectral

bandwidth must match the spectrum of the fundamental wiggler radiation averaged over the transverse mode.

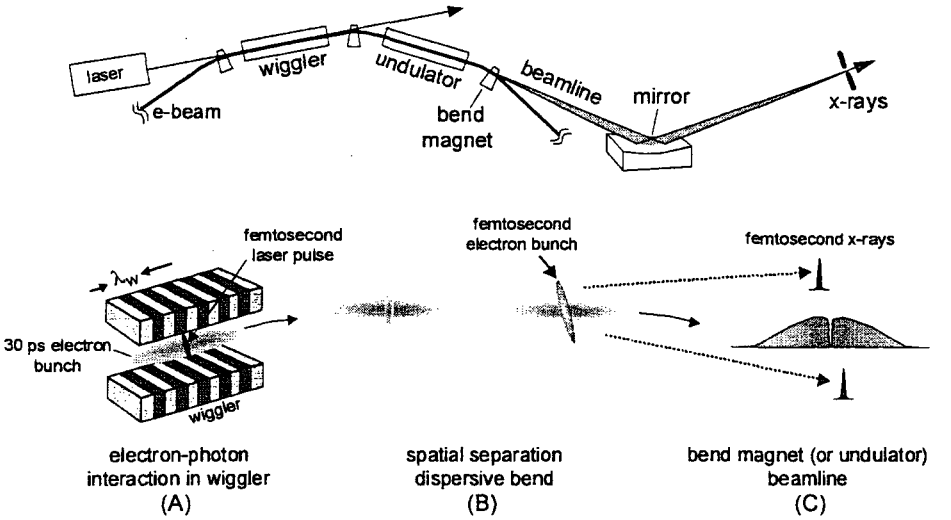

Figure 1. Schematic of method for generating femtosecond synchrotron pulses.

By creating an energy modulation that is significantly larger than the beam energy spread, the transverse dispersion of the storage ring causes a spatial displacement of the modulated electrons from the rest of the electron bunch (Fig. 1B). Finally, by imaging the synchrotron radiation from the displaced electrons to the experimental area, femtosecond x-rays can be separated from the long pulse using an aperture (Fig. 1C). The time structure of the temporally incoherent synchrotron radiation is directly determined by the time structure of the electron bunch and is invariant over the entire spectrum of the synchrotron emission, from infrared to x-ray wavelengths.

A new bend-magnet beamline dedicated to time-resolved x-ray research, and incorporating the above-described technique for generating 100 fs x-ray pulses has recently been constructed at the ALS (BL5.3.1). Figure 2 shows the beamline layout. It includes a grazing incidence toroidal optic which projects a 1:1 image of the bend-magnet source (250×50 μm^2) into the x-ray experimental hutch, where a pair of slits is used to select the femtosecond x-rays. A double-crystal monochromator (with interchangeable crystals) provides a spectral resolution of $\Delta\lambda/\lambda \sim 10^{-3}$ over the energy range from 1.8 to 12 keV. In addition, a dispersive (ruled grating) spectrometer provides measurement capability to ~300 eV. The beamline endstation includes an x-ray streak camera with a measurement resolution of ~2 ps, and a chopper (synchronized to the laser repetition rate). The laser system associated with the beamline has recently been upgraded to operate with TM_{00} mode at 2.5 kHz (5 kHz operation is anticipated in the near future) and includes two amplifiers operating in parallel providing separate pulses (~1 mJ, 75 fs) for electron beam interaction and sample excitation.

The beamline has been commissioned for long-pulse operation and the commissioning and characterization of femtosecond operation is presently in progress. Modifications to the wiggler have recently been completed which now enable this work to continue during normal user operation shifts at the ALS. The performance goal for this beamline is to provide a x-ray flux of $\sim 10^5$ ph/s/0.1% BW and a brightness of $\sim 10^8$ ph/s/mm^2/mrad^2/0.1 BW with a pulse duration of ~100 fs.

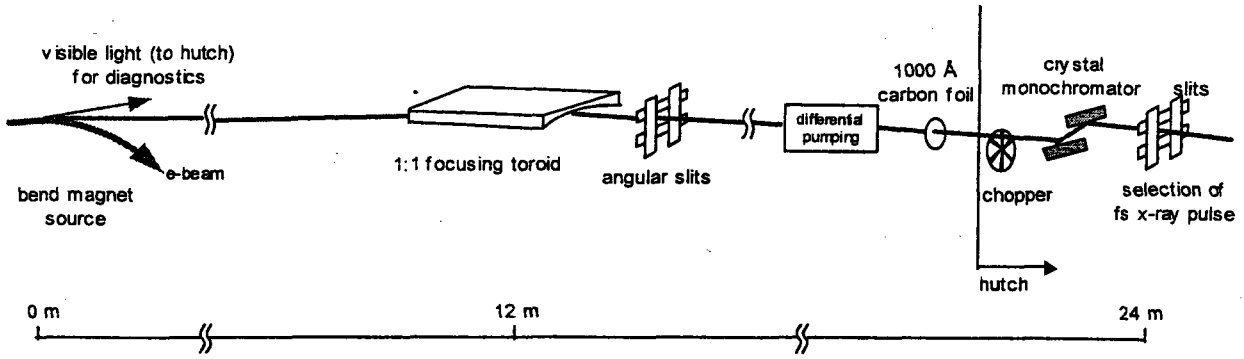

Figure 2. Schematic layout of ALS beamline 5.3.1

A diverse research program is being developed at beamline 5.3.1 consisting of experiments in several areas: solid-liquid and solid-solid phase transitions in laser-heated crystalline solids, structural dynamics of photo-induced spin-crossover transitions in molecular crystals, and modulation of x-ray absorption and ionization processes in laser-dressed atoms. The Falcone group (U.C. Berkeley, Physics Department) is using XANES techniques with an ultrafast x-ray streak camera (combined with a soft x-ray spectrometer) on BL5.3.1 to investigate changes in the electronic and ionic structure of amorphous Si foils excited to the warm dense matter regime (~1 eV) via laser heating. Initial results at 100 ps following excitation indicate distinct changes in the L_{II} and L_{III} absorption magnitudes as well as "smearing" of the edges over ~1 eV. The group of A. Belkacem and collaborators (LBNL Chemical Sciences Division) are applying x-ray photoelectron and ion spectroscopy techniques to investigate two-photon (visible plus x-ray) ionization of core levels in Ar gas. These experiments are expected to provide new knowledge about the physics underlying two-photon processes in atoms, and will provide a basis for future experiments of x-ray ionization of laser-dressed atoms. Our research group (Shank/Schoenlein LBNL Materials Sciences Division) is applying EXAFS techniques to investigate changes in coordination and bond distances (nearest-neighbor) during a laser-induced non-thermal solid-solid phase transition in crystalline VO_2. We are applying similar techniques to investigate the light induced spin-crossover transition in Fe^{II} molecular crystals. In both of these systems, the electronic transitions are known to occur on the ultrafast time scale. However, little is known about the dynamics of the structural changes that are strongly coupled to the ultrafast electronic processes. Finally, the user group of Prof. M. Chergui (University of Lausanne, Switzerland) has done some preliminary studies using XANES techniques to observe the photogeneration of iodine radicals in H_2O. The long-term goal of this work is to understand solvent structural dynamics. At present, many of these experiments are at a very early stage and are being initially developed with long x-ray pulses from the beamline. The development and optimization of pump-probe techniques with the high-flux, 30 ps x-ray pulses is an important first step toward ultrafast measurements using the lower-flux femtosecond x-rays.

References

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[3] R.W. Schoenlein, S. Chattopadhyay, H.H.W. Chong, T.E. Glover, P.A. Heimann, C.V. Shank, A. Zholents, and M. Zolotorev, *Appl. Phys. B*, **71**, (2000), 1.

A Phonon Bragg Switch for X-Rays"

Principal Investigator: P.H. Bucksbaum; Co-PI on renewal: David Reis
Physics Department, University of Michigan, Ann Arbor, MI 48109-1120
phb@umich.edu. Grant DEFG02-00ER15031

Program Scope

The Advanced Photon Source at Argonne National Laboratory provides tunable x-rays for research in physics, chemistry, biology, and engineering. Most spectroscopic or imaging problems at this third-generation synchrotron make use of its high average spectral brightness, which can exceed 10^{12} x-rays per second within a 10^{-4} fractional energy band, with beam divergence below 100μr. Still more impressive is the peak spectral brightness, which can approach 10^{4} monoenergetic hard x-*rays per picosecond.* This is high enough for diffraction or absorption measurements on the time scale of atomic motion in molecules and condensed media.

The x-ray pulse duration from the synchrotron is on the order of 100 *ps*, which is too long to resolve motion on the 0.1*ps*-1*ps* scale of molecular dynamics, so ultrafast x-ray gates or x-ray detectors are needed. Two years ago we began an experimental program to modulate the synchrotron x-ray beam by ultrafast laser excitation of x-ray diffractive optics, such as Bragg mirrors.

The work is done in conjunction with the MHATT-CAT collaboration, on Sector 7 of the APS. The hutch housing our experiment was completed late 1999. Since then we have had eight 7-10-day periods of access to x-rays from the ID (undulator insertion device) beam line.

In the two-year initial period of this project ("The phonon Bragg Switch:" see section below) we demonstrated coherent control of x-rays on sub-nanosecond time scales using laser-induced transient excitation of crystalline solids. Several new phenomena were observed that could lead to efficient x-ray sub-picosecond switches for physics and chemistry applications. We also made new measurements of strain propagation following laser excitation in semiconductors, which have helped to elucidate models of laser energy transfer to the lattice. These studies have helped to establish the field of ultrafast x-ray science at synchrotrons, and they form the foundation for ultrafast x-ray studies at future fourth-generation x-ray free electron lasers or energy recovery linac-based femtosecond x-ray sources.

Recent progress

Throughout the first grant period much of our effort was devoted to building the laser , feedback, timing, and gated detector systems that makes laser-x-ray experiments possible. Since no laser beams enter or leave the hutch, all aspects of laser and sample manipulation must be remotely controlled during experiments. The laser is a Ti:Sapphire multipass chirped-pulse amplified design, with the following output characteristics: 50-100 fsec pulse duration; 840-790 nm central wavelength; 1 kHz repetition rate; 0.5-1mJ pulse energy. The laser pulse is synchronized to the x-rays to better than the x-ray pulse

duration. This is accomplished by slaving the laser oscillator cavity round-trip period to the synchrotron accelerator RF signal.

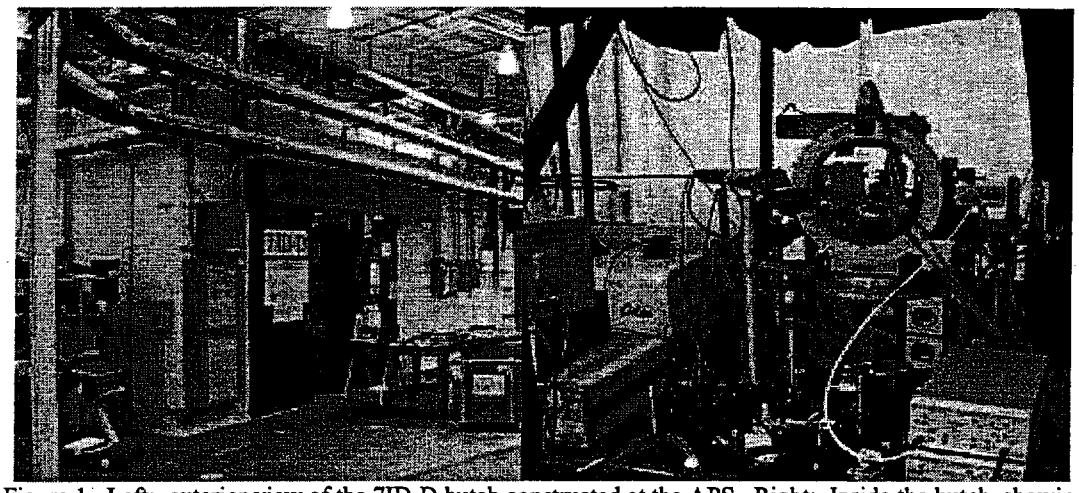

Figure 1. Left: exterior view of the 7ID-D hutch constructed at the APS. Right: Inside the hutch, showing part of thelaser system, the goniometer ans pump-probe apparatus.

The laser-x-ray system synchronization was initially tested with a series of measurements of impulsive acoustic excitation of crystalline InSb. We chose this crystal because previous work at the Berkeley Advanced Light Source has shown that laser-excitation produced large lattice distortions [Lindenberg, *et al. Phys. Rev. Lett*, **84**, 111-114 (2000)]. With the increased flux of the APS, we were able to make quantitative comparisons to the existing models. We could separate the transient strain into thermal-induced and coherent acoustic components, and found that models used to explain previous experiments significantly underestimate the contribution of coherent acoustic phonons to the overall strain. This finding was published in Physical Review Letters.

Our original idea for a Bragg switch was based on impulsive optical phonon generation by an ultrafast laser incident on a GaAs Bragg crystal. GaAs excitation of optical phonons is extremely inefficient, so we have searched for better materials. We showed that it was possible to obtain very large amplitude optical phonon fields in crystalline Bi, with atomic displacements on the order of 1%. This constitutes an order of magnitude increase from previously reported results. At high laser amplitudes, anharmonic distortions become visible in the phonon oscillation spectrum, and a redshift in the phonon frequency is observed. We demonstrated that it is still possible to exert simple control over the atomic displacements even in this anharmonic regime. This work has been published in Physical Revew B. We have also studied optical phonon generation in a number of other candidate materials for a Bragg switch. Good results were obtained in bismuth germinate ($Bi_4Ge_3O_{12}$, also called BGO), an optically transparent material used in gamma ray calorimeters, which can produce very large and high-Q optical phonon modes when sub-100 fsec optical pulses are transmitted through it.

Our most successful experiments on x-ray modulation have been in transmission geometries in crystalline Ge. We observed that an impulsively generated acoustic pulse

in a thick crystal modulates the coherent transmission of x-rays propagating many x-ray incoherent absorption depths through the material (a combination of the classical Borrmann effect and the Pendellösung effect). The excitation was observed to transfer energy between two x-ray beams in a time shorter than the synchrotron pulse width. Different arrangements were studied, including a switch made from two counter-propagating acoustic pulses that collide deep inside the crystal. This doubles the acoustic modulation frequency, and also provides one means to observe a localized transient strain inside an otherwise opaque material. The initial report of these studies was submitted to *Nature,* and has received favorable reviews.

The switching speed is ultimately limited only by the motion of atoms, and could be much less than a picosecond for optical phonon excitation. A high priority in our renewal project will be the study of sub-picosecond optical phonon-induced Pendellösung modulation of the x-ray beam.

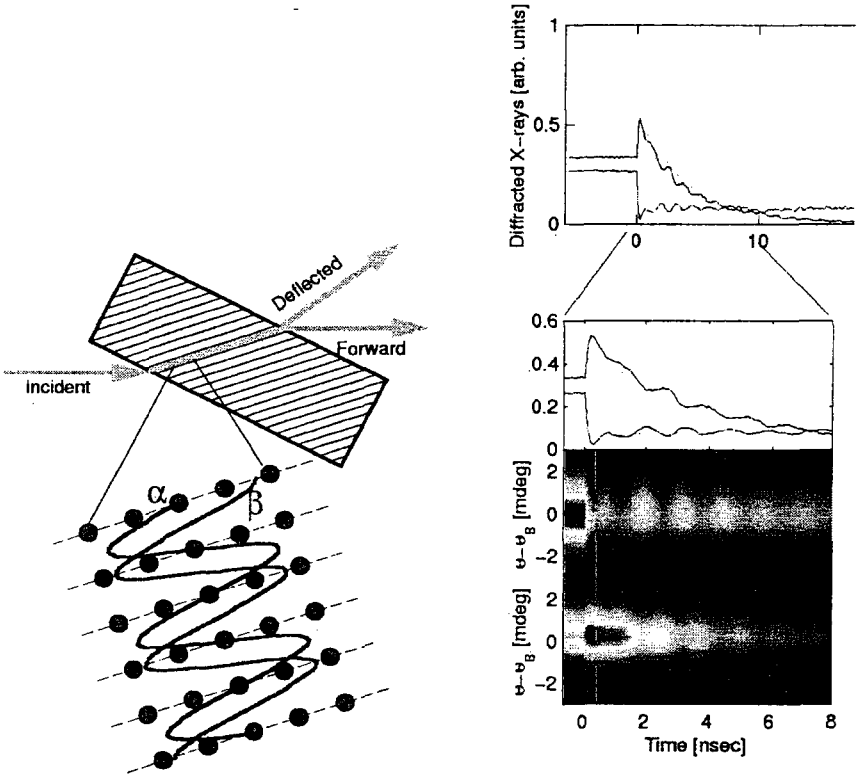

Figure 2. Time-resolved diffraction measurement of the $20\bar{2}$ reflection following laser impulsive excitation of an acoustic wave on the exit face of a thick Ge [001] crystal. Top: Forward (blue) and deflected (red) x-ray intensity as a function of time following excitation. Bottom: expanded scale showing the regions where the acoustic pulse is near the exit or entrance crystal surface. 2-d plots: the x-ray transmission vs. time and crystal angle near the forward (top) and deflected (bottom) diffraction peaks in normalized false color.

Plan of Research in the proposed grant period

We propose to continue to develop the new field of ultrafast x-ray coherent control in the next three years. The research plan has three main goals.

1. First we want to improve the quality of ultrafast x-ray modulators. This means developing methods to increase the speed, contrast, and efficiency, and also finding new methods that make these novel devices more accessible to other researchers at the APS and elsewhere.

2. Second, we want to develop the science of laser excitation and x-ray coherent scattering in solids that makes these switches possible in the first place. We are particularly interested in two research topics begun in the previous grant cycle: the partitioning of laser-deposited energy in semiconductors; and the rapid transport of energy into the crystal following initial surface excitation, by electron-hole plasma diffusion or other means. An x-ray streak camera would aid this effort by increasing our x-ray temporal resolution by two orders of magnitude with existing technology.

3. Third, we want to discover and develop new applications of ultrafast x-rays in atomic, molecular, chemical, or materials science. This will help to foster increased interest and activity in this new field, in advance of new x-ray sources developed in the coming years. One example is the development of Laue sideband detection of transient structure in complex crystals.

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Laser-Produced Coherent X-Ray Sources

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I. PROGRAM SCOPE AND DEFINITION

We experimentally explore the physics of ultra-high-intensity laser-plasma interactions and their applications to novel radiation sources. Our interests have shifted in the last few years from a more general study of high-field interactions towards the more directed goal of creating coherent keV-energy photons from a table-top source.

II. RECENT PROGRESS

A. Relativistic Nonlinear Thomson Scattering

The predicted figure-eight motion of relativistic electrons in field of high-intensity laser light was confirmed by us several years ago by the unique angular distribution of the harmonic light emission [1]. We later reported the observation of coherent emission in the forward direction (along the laser axis) from relativistic nonlinear Thomson scattering [2]. These harmonics are fundamentally different from those produced by the rescattering of electrons from the nucleus, but instead originate from intense laser light interactions with relativistic free electrons. Following is a summary of recent observations for low-order harmonics.

1. Imaging of coherent third harmonic in the forward direction

As shown in Fig. 1, a gaussian third-harmonic beam is generated under conditions of illumination by a laser beam with a gaussian profile.

FIG. 1. Image of the coherent third harmonic in the forward direction.

2. Dependence of harmonics on the light polarization

We have shown that there is significant harmonic emission even with circularly polarized light as expected on the basis of the fact that these harmonics are from free electrons. More interestingly, significant white light emission is seen from the plasma for linearly polarized light. When circular polarization is used only the harmonics are seen and there is no continuum emission. This is because of the fact that the atomic susceptibilities leading to the supercontinuum generation in the gas are of smaller magnitude for circular polarization as compared to linear.

3. High-order harmonics

Currently, we are conducting the first study of scattering from relativistic plasmas in the short-wavelength (XUV) spectral region. The experiments were carried out with an Nd:glass laser system with a peak intensity of 10^{19} W/cm^2, in underdense He plasma. First experiments on relativistic harmonics were carried out with the Seya-Namioka spectrometer with a toroidal grating. In the current experimental geometry we are studying incoherent harmonic emission. Preliminary estimates indicate that the harmonics extend to about 120 nm. The fact that these harmonics are from the free electrons has been verified by the fact the there is significant emission even when circularly polarized light is used. The harmonics are found to scale with the gas density as $n_e^{1.2}$ showing that these harmonics are incoherent and there are no many particle plasma effects involved. The dependence of the harmonic yield on the intensity is linear as shown in Fig. 2 and as expected on theoretical grounds.

Our results on the angular and spectral distribution are compared with theoretical calculations of relativistic laser-electron scattering. The relative importance of Thomson (incoherent [1] and coherent [2]) and Compton scattering is determined. The role of phase matching is elucidated in experiments used to study both the coherent and incoherent components of the scattered radiation field.

Preliminary experiments have been carried out to see if there is a contribution of Compton scattering to the observed spectra. Since our gas jets are known to produce a high energetic electron beam,

scattering of electrons off the incident laser light can produce broadband emission in the energy range being studied in our experiments. In fact preliminary data shows that this is indeed the case. When spectra are compared for cases where an electron beam is produced to those without a decrease in the in the harmonic yield is noticed along with a modification of the spectral profile of the emission.

The efficiency of this process opens up the possibility of using relativistic harmonics as a coherent short-wavelength radiation source.

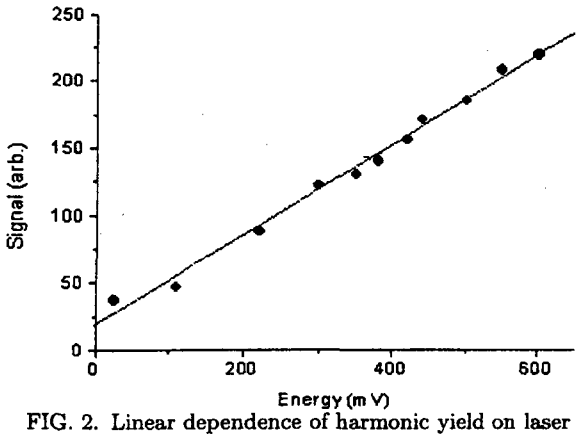

FIG. 2. Linear dependence of harmonic yield on laser intensity (arbitrary units).

B. X-ray Radiation from Dense Matter

We have continued our study the effects of extremely high fields on ion electronic structure, which result in strongly coupled effects such as continuum lowering, line broadening and line-merging. The experiments presented in this paper have been done with high-intensity ultrashort laser pulses. Using time-resolved spectroscopy in XUV spectral region, we demonstrate the effect of near solid density plasma on the emission spectra of multicharged ions in keV spectral region that results in continuum lowering, significant line broadening and line merging. This research provides a test-bed for atomic physics models of such plasmas. Shown in Fig. 3 are time-integrated emission spectra of a magnesium plasma produced by high-intensity laser pulse for two cases: low density (with a prepulse) and high density (high-contrast illumination). Low laser contrast heats the target prior to the arrival of the main pulse and thus produces a density that is lower than solid density. It can be clearly seen that narrow spectral lines only appear in the low-density case, when the fields of neighboring ions do not broaden the lines. In the high-density case, the lines are indistinguishable from the continuum.

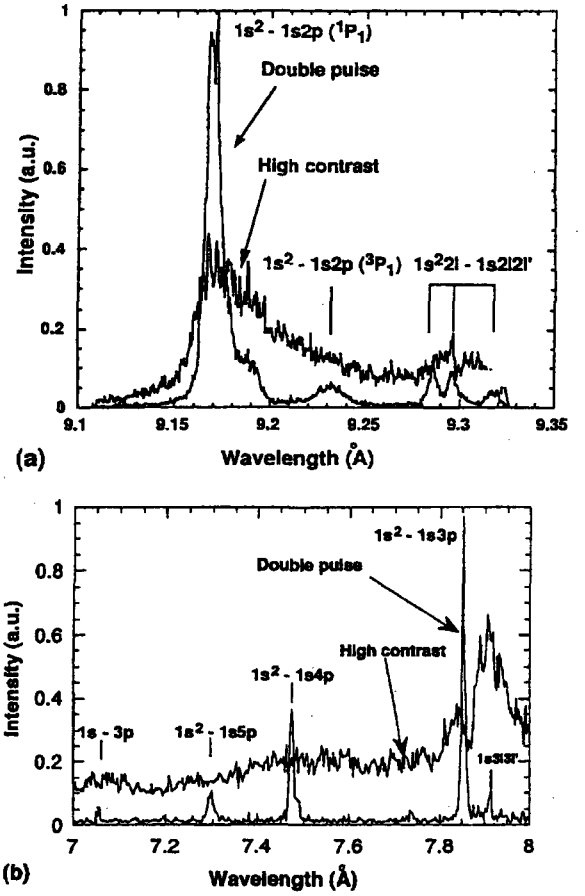

FIG. 3. Time integrated emission spectra of a magnesium plasma produced by high-intensity laser pulse with a prepulse and for high contrast laser illumination of the target: (a) in the vicinity of He-like resonance, intercombination and Li-like satellite lines (the intensity of the spectrum for high contrast illumination is multiplied by 2 for better viewing) and (b) in the vicinity of He-like 1s2–1snp lines. Spectrum for the high contrast illumination is shifted vertically for better viewing.

C. Laser-accelerated Ion Beams

Also reported are the results of experiments in which ultra-short duration (femtosecond) laser pulses from table-top lasers are focused to intensities above 10^{19} W/cm^2 onto solid-density matter. At such extreme electromagnetic field strengths (10^{11} V/cm), a plasma is formed in which the electrons oscillate relativistically, creating gigabar pressure. The displacement of electrons—but not the heavier ions—from the region of the laser focus drives large space-charge fields (exceeding 1 GeV/cm). The ensuing Coulomb explosion accelerates protons (or other ions) to energy in excess of 10 MeV in a well

collimated beam [3,4]. The spatial profile of a proton beam measured with a nuclear track detector, composed of CR-39 plastic, is shown in Fig. 4.

FIG. 4. Image of the spatial profile of laser-accelerated proton beam.

The scaling of the proton-beam energy with laser intensity is shown in Fig. 5, measured experimentally (squares), and simulated with a 2-D particle-in-cell (PIC) code (dots). The scaling appears to change as the intensity of the laser crosses a threshold given by 10^{19} W/cm^2, which corresponds to the demarcation between weakly and strongly relativistic electrons. Applications of these energetic and ultrashort duration ion beams include laboratory astrophysics, cancer radiotherapy, fast ignitor fusion, and high-current injectors for high-energy and nuclear physics [5,6].

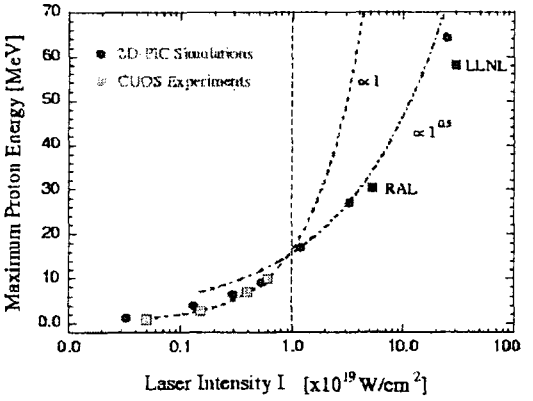

FIG. 5. Scaling of proton energy with laser intensity.

D. Laser-Driven Nuclear Transformations

One near-term application of laser-accelerated ions is the production of isotopes nuclear physics and medicine. As shown in Fig. 6, these well collimated beams of ionized hydrogen (deuterium) were can be "fused" with elements like boron to transform the latter into radioactive carbon. We observed the production of $\sim 10^5$ atoms of positron active isotope ^{11}C from the reaction ^{10}B(d,n)^{11}C. Such isotopes are commonly ingested by cancer patients and made to seek out tumors while they are undergoing radioactive decay. The resulting emission of gamma rays (energetic x-rays) is used to precisely locate the tumor in a commonplace technique called positron emission tomography, or PET. Up until now, a large device called a cyclotron was required to produce the radioactive isotopes for PET. But ions are accelerated by our high-power laser in a 10,000-times shorter distance than in a cyclotron, providing the possibility of a much more compact and inexpensive isotope source. The activation results also suggest that the deuterons were accelerated from the front surface of the target [7].

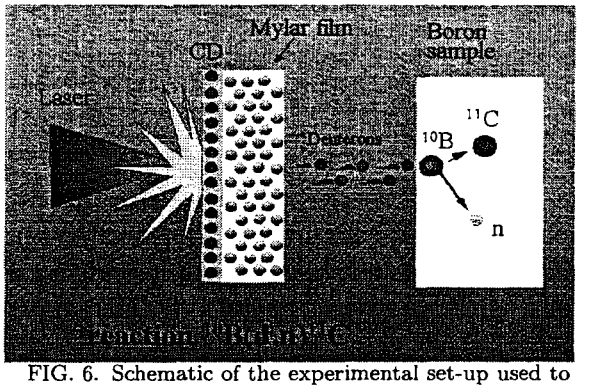

FIG. 6. Schematic of the experimental set-up used to create a radioisotope with laser-accelerated deuterons.

Because these laser-produced ions have extremely short pulse durations, they can be also be used to create short-lived isotopes for studies in nuclear physics. It is more difficult to study these transient nuclear states with longer duration ion pulses. The reason is that long ion pulses will produce isotopes for periods of time that are much longer than the radioactive decay time. It was also demonstrated that there are a sufficient number of electrons accelerated by laser-plasma accelerators to conduct pulsed radiolysis [8]. Koshichi Nemoto, visiting Michigan from the Central Research Institute of Electric Power Industry in Tokyo, also participated in this project.

III. FUTURE PLANS

Our future research will be focused mainly on the study of short-wavelength generation with much shorter laser pulses and higher laser intensities. This will be accomplished with a new laser, which will have 100-TW power and 30-fs duration and will be in operation by the end of the year. Several issues related high-order harmonics remain to be resolved, including the role of Compton scattering and some unusual spectral features (non-harmonic emission). Characteristics of nonlinear relativistic Thomson scattering predicted from exact analytical calculations and numerical codes, which are currently being developed in collaboration with Prof. Y.Y. Lau of the University of Michigan, will be compared to the experimental results. Harmonic emission from solid targets will be compared with that from gaseous density targets. Additionally, the scattering of short duration, high intensity laser pulses from our 30-MeV energy laser-driven electron beam we will be studied.

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DYNAMICS OF FEW-BODY ATOMIC PROCESSES

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PROGRAM SCOPE

This project aims to elucidate the role of many-body (specifically electron-correlation) effects on atomic processes. The main emphasis in the 1990's was on benchmark studies of negative ion photodetachment processes in the energy region of highly-excited two-electron resonances. The current emphasis is on the influence of electron correlation in higher-order photon processes, both in the perturbative (i.e., multiphoton) regime and especially in the non-perturbative (i.e., intense laser) regime. A nascent interest of this program concerns interacting spin 1/2 systems as quantum information devices.

RECENT PROGRESS

A. Angular Distributions for Double Ionization of Li^- by an Ultrashort, Intense Laser Pulse

[G. Lagmago-Kamta and A. F. Starace, *Phys. Rev. Lett.* **86**, 5687 (2001)]

Double photoionization remains a challenge in atomic physics due to the difficulty of describing correlated motion of two interacting continuum electrons in the field of an ion core. The simplest case of single photon double ionization is still the object of much research [1]. Most theories for this case evaluate triply differential cross sections (TDCS) in terms of transition matrix elements coupling initial and final states. While methods exist to obtain the initial state to arbitrary accuracy, the search for analytic representations for the asymptotic final state wave function continues [2]. Many *ab initio* approaches use the stationary Schrödinger equation to construct the final state numerically [1,3]. Direct numerical integration of the three-dimensional, time-dependent Schrödinger equation (TDSE) is the only approach adequate for studying the interaction of an atomic system with an ultrashort, intense laser pulse. For two-electron systems, this remains a challenging task. However, recent increases in both computer speeds and data storage capacities has allowed theorists to begin tackling this problem without gross approximations. Several approaches that account for the pulse shape have been developed [4,5]. For double ionization by intense fields, very little is known concerning the distribution of ejected electrons, despite recent electron coincidence [6] and COLTRIMS [7] measurements. On the theoretical side, although angle-averaged radial probability density plots (averaged over all angles except θ_{12}) indicate that the two electrons may be ejected with a small relative angle θ_{12} by an ultrashort intense laser pulse [8], an approach providing detailed and complete angular distributions is still needed. For this purpose, one requires a non-perturbative approach that includes correlation effects not only in the initial state but also in the time-dependent final state throughout the interaction time of the system with the laser field.

In this work, we have described such an approach that involves the solution of the TDSE for a real two-active electron system and have presented angular distributions following one- and multi-photon double ionization of Li^-. In our approach to solving the TDSE we treat the Li^- ion as a two-active electron system,

where each electron is assumed to move in the field of a potential $V(r)$ describing the Li^- core [9,10]. This potential accounts for the polarizability of the Li^+ core and contains semi-empirical parameters which are fitted to reproduce the experimentally measured energy levels of Li. It has been used successfully in time-independent photodetachment calculations for Li^- whose results agree well with experiment in the perturbative regime [10,11].

We solve the TDSE in a box of radius r_o chosen so that for the laser intensities and pulse durations employed the calculated two-electron probability is negligible for $r_{1,2} = r_o$. Our two-electron wave function is obtained as an expansion in coupled one-electron wave functions. The numerical approach used allows us to exclude spurious contributions to double ionization arising from bound and doubly-excited states lying below the double ionization threshold. (We have found that those spurious contributions are of considerable importance at higher laser intensities when electronic transitions exist at intermediate photon frequencies.)

We define the doubly differential double ionization probability (DDDIP) as the integration of the calculated two-electron probability at a time T (marking the end of the laser pulse) over the electron radial coordinates r_1, $r_2 > r_c$, where r_c is a cut-off radius chosen to encompass most of the ground state probability distribution. This definition is consistent with that used by others to calculate total double ionization probabilities [12]. As in [13] it obviates the need to specify asymptotic boundary conditions. A unique aspect of our calculations is that the spurious contributions arising from bound and doubly excited states are easily removed from our final state wave function owing to its representation in terms of two-electron configurations.

Our results show that the electron Coulomb repulsion and other selection rules strongly affect the emission angles of the two electrons in the one-photon double ionization process. In the multiphoton case electrons may possibly be ejected in all directions, but with sharp peaks corresponding to double electron ejection along the directions parallel and perpendicular to the polarization axis of the field. For the intensities studied, configurations where electrons are ejected in opposite directions are more likely than those for ejection in the same direction. These results were the subject of an invited hot topics talk by G. Lagmago-Kamta at the 2001 DAMOP meeting.

B. Role of Two-Electron Excited States in One- and Two-Electron Detachment of Li^- by an Ultrashort, Intense Laser Pulse
[G. Lagmago-Kamta and A. F. Starace]

Our approach to solving the time-dependent Schrödinger equation for two-electron systems in the presence of an ultrashort, intense laser pulse is capable of describing in detail excited two-electron states of the lithium negative ion complex. By choosing the laser pulse frequency on or off resonance with one of these two-electron excited states, we are able to examine the role of such states on ionization processes. Our as yet unpublished calculations show that such doubly-excited states contribute strongly to both single- and double-ionization processes. When the laser frequency is tuned to resonance with a doubly-excited state, the single-ionization probability is enhanced whereas the double ionization probability is reduced because of the competition with the decay of the doubly-excited state.

C. Role of Rescattering in One- and Two-Electron Detachment of Li^- by an Ultrashort, Intense Laser Pulse
[G. Lagmago-Kamta and A. F. Starace]

Our approach to solving the time-dependent Schrödinger equation for two-electron systems in the presence of an ultrashort, intense laser pulse is capable of treating arbitrarily-shaped laser pulses, including half-cycle and few-cycle pulses. Such pulses are ideal for determining the role of rescattering on two-electron ionization processes by an intense laser pulse. Classical models such as the "simple man model" [14,15] attribute a key role to rescattering processes in a number of intense laser-atom processes, such as high order harmonic generation. Our as yet unpublished quantum-mechanical calculations show that rescattering interactions contribute strongly to both single- and double-detachment processes. This work is being prepared for publication.

D. Dynamic Polarizability of the Helium Atom
[M. Masili and A. F. Starace]

In the past year we have published benchmark two-photon detachment cross sections for H^- [16]. These results agree with the accurate correlated B-spline results of van der Hart [17]. Our approach employs a generalization of the variationally stable method of Gao and Starace [18,19] for two-electron atoms and ions and incorporates a coupled-channel adiabatic hyperspherical approach [20]. The results [16,17] bring for the first time a consensus to theoretical predictions for the fundamental two-photon detachment cross section of H. We are presently extending these calculations to treat the He atom. Our first results for the dynamic polarizability of He were reported at the 2001 ICPEAC meeting.

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FUTURE PLANS

We are currently examining the role of anisotropy and external static fields on the entanglement of interacting spin 1/2 quantum dot systems. We are proceeding to obtain benchmark results for two-photon ionization of helium using our variationally stable approach. We are employing a new representation of the Coulomb Green's function to evaluate double ionization processes in helium perturbatively. We are investigating double ionization of helium by an intense ultrashort laser pulse, as well as two-electron effects in high-order harmonic generation.

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New Electronic Structure Methods for Bond-Breaking.

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To develop robust electronic structure methods that can describe very different parts of a potential energy surface at similar levels of accuracy remains a challenging problem. To do this with relatively simple wave functions virtually demands that one attempt to partition correlation effects into valence or static correlations, versus out-of-valence dynamical correlations. I shall discuss several recent developments from my group that we hope represent significant steps towards achieving feasible and accurate models along these lines based on coupled cluster theory. These developments deal first with the form of an appropriate coupled cluster approximation to the valence space Schrodinger equation. This can be done with good accuracy using only double excitations, as will be demonstrated with various numerical results. Indeed we shall show that cluster wavefunctions composed of a special set of generalized double excitations are in fact exactly equivalent to full configuration interaction! The second problem is the formulation of a true second order perturbative correction for the remaining non-valence correlations, which we are addressing via the general (2) correction we have recently proposed. Relative to traditional multi-reference approaches such as CASSCF+CASPT2, these new approaches offer the advantage of much reduced computational complexity, which permits the calculations to be performed in a full valence or 1:1 active space. In other words there is no need to select a small active space based on chemical intuition.

Rovibrational modes and classification of triply excited states of atoms

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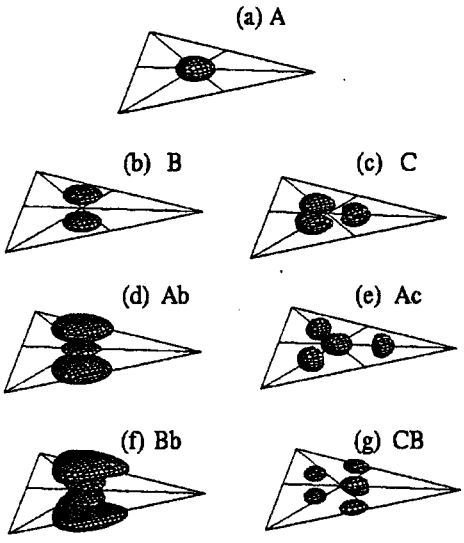
It is well understood that electron correlation plays a major role in determining the properties of multiply excited states of atoms. The shell model does not describe these states and alternative quantum numbers are needed to classify them. For doubly excited states, classification of these states in terms of new quantum numbers, $(K, T)^A$, have been available since the 1980's. The K, T and A are used to describe the collective bending and stretching motion of the two excited electrons, in analogy to the bending and breathing motion of a linear triatomic molecule. New spectral regularity emerges and propensity rules in processes involving doubly excited states have been found when they are ordered according to the new quantum numbers.

In the past few years we have been investigating the classification of triply excited states of atoms. We have developed a computational package for treating three-electron atoms in hyperspherical coordinates, but the emphasis in recent years has been on the classifications. *We have succeeded in classifying the intrashell triply excited states*. For example, there are eight intrashell triply excited states when all the three electrons are in the n=2 manifold, the so-called 2l2l'2l" triply excited states. The eight states have been separated into three groups. Within each group the three electrons perform motion akin to the vibrational motion of an XY_3 molecule with X being the nucleus and Y the electron, and the states within the group are identified as the rotational excited states of a symmetric top. The identification of the internal vibrational normal mode is supported by displaying the joint electronic density plots in appropriate coordinates and by analyzing the internal nodal surfaces of the wave functions as imposed by the quantum symmetries. Density plots for the three groups of the 2l2l'2l" triply excited states are shown as A, B and C in the plots here. As will be explained in the talk, in A the three electrons form a coplanar equilateral triangle with the nucleus at the center, in B the three electrons perform motion like that of an ammonia molecule and in C the three electrons can form an isosceles triangle while the equilateral triangle geometry is forbidden. Most recently we have extended the classification to the **sixty-four** 3l3l'3l" triply excited states. For this purpose we study the classification of a model atom with three electrons on the surface of a sphere. We have succeeded in classifying all these 64 states. Besides the fundamental A, B and C normal modes, excited vibrational modes such as Ab, Ac, Bb, etc., have been identified, see figure. We will show that the triply excited states of N^{2+} and N^{4+} can indeed be classified using the present scheme. Extension of the classification to intershell triply excited states is underway.



Correlation in dynamic multi-electron systems

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Cross sections for double excitation, ionization with electron capture and ionization with excitation in helium have been evaluated in full second Born calculations that include both electron correlation and time ordering. These full second Born calculations are compared to calculations in the independent electron approximation (iea), where spatial correlation between the electrons is removed. Comparison is also made to calculations in the independent time approximation (ita), where time correlation between the electrons is removed. Good agreement is found between our full calculations and experiment, except that the lowest velocities, where higher Born terms are expected to be significant. Both spatial electron correlation, arising from internal electron-electron interactions, and time correlation between electrons, arising from time ordering of the external interactions, can give rise to observable effects. Our method [1,2] can also be used for photon impact.

Most complex atomic and molecular systems cannot be adequately described in terms of independent transitions of electrons. In applications from molecular dynamics to quantum computing , correlation between electrons increases and modifies reaction pathways. These dynamic electronic connections can redistribute energy and facilitate transitions that would otherwise be forbidden. These correlations in time are caused by external interactions, such as strong electro-magnetic fields together with internal interactions, such as the Coulomb interactions between electrons or spin-spin interactions. These interactions can be used to shape and dynamically control nanostructures. The problem of time correlation in multi-electron systems has recently been addressed theoretically [1,2], with inclusion of corresponding energy non-conserving terms. Nevertheless, the effect of time correlation between electrons has not been previously confirmed by experiment, as reported here.

A new focus of our work has been time correlation in dynamic multi-electron systems. Interconnection of different electrons in time requires both internal electron correlation, which interconnects the electrons, as well as time ordering, which imposes a causal-like sequencing of interactions in quantum time propagation. Electron correlation is relatively well understood. Time ordering comes from the time-dependent Schrödinger wave equation, which determines the equation for the time evolution operator, $U(t_2,t_1)$, which describes how the system changes between time t_1 and time t_2 . The formal solution for the evolution operator may be expressed,

$$U(t_2,t_1)=\sum_{n=0}^{\infty}\frac{(-i\hbar)^n}{n!}\int_{t_1}^{t_2}dt^{n'}\ldots\int_{t_1}^{t_2}dt^{1'}\int_{t_1}^{t_2}dt^{1}\,T\,V(t^{n'})\ldots V(t^{1'})V(t^{1}). \qquad (1)$$

Here V(t) is the interaction (or sum of interactions) of a system of atoms with light or matter and T is the Dyson time ordering operator, which imposes the causality-like constraint that

$V(t^{l'})V(t^{k'}) = 0$ if any $t^{l'} < t^{k'}$. The time ordering operator T provides a time connection between pairs of interactions due to the constraint that the interactions occur in the order of increasing time.

The time ordering operator T may be decomposed into two terms, namely, $T = T_{cor} + T_{unc}$. The correlated term, T_{cor}, gives the time correlation between electrons and regulates sequencing of the interactions V(t). Time correlation arises from the enforcement of time ordering on the sequence of interactions, $V(t^{n'})$ .... V(t'') V(t'), which cause the system to change. The uncorrelated term, T_{unc}, is the time independent part of T, which does not connect the various interactions V(t) in time and thus eliminates both sequencing and time correlation in the time evolution of the system. For a two-electron transition occurring via $V_1(t)$ and $V_2(t)$ respectively, McGuire et al. [1,2] have shown that in second order,

$$T_{unc}\, V_2(t'')V_1(t') = 1/2\,(V_2(t'')V_1(t') + V_2(t')V_1(t'')), \qquad (2a)$$

so that,

$$T_{cor}\, V_2(t'')V_1(t') = 1/2\, sign(t''-t')[V_2(t''),V_1(t')], \qquad (2b)$$

Here *sign*(t'' – t') = (t'' – t')/|t'' – t'| is a unit vector in the direction of increasing time, and $[V_2(t''),V_1(t')] = V_2(t'')V_1(t') - V_1(t')V_2(t'')$ explicitly requires a time connection between interactions at different times, t' and t''. The Fourier transform of T_{cor} gives the principal value part of the energy propagator. This quantum term violates conservation of energy for short times. This effect is not present in the propagation of classical particles or waves.

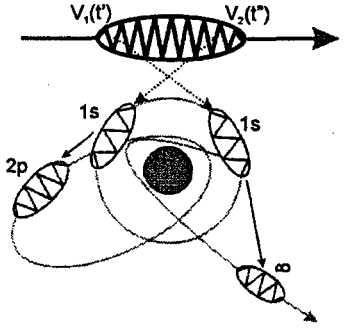

Fig. 1. Correlation for two electrons changing from a (1s,1s) initial state to a (2p,∞) final state. Time correlation occurs when $V_2(t'')$ is connected to$V_1(t')$. With quantum time ordering $V_2(t'')V_1(t')$ may differ from $V_1(t')V_2(t'')$. Then $V_2(t'')$ is connected to$V_1(t')$. Spatial electron correlation occurs when two electrons interact with one another (denoted by the arrows).

In the independent electron approximation, where spatial correlation between electrons is removed, the quantum commutator $[V_2(t'),V_1(t')]$ is zero. Then each electron evolves independently in time, i.e. the evolution operator, $U(t_2,t_1)$, reduces to a product of single electron evolution operators. When spatial correlation between electrons is included, e.g. via two-body Coulomb electron-electron interactions, $[V_2(t'),V_1(t')]$ is non-zero, and the time evolution of

different electrons may become entangled. Time correlation between electrons corresponds to cross correlation between the time propagation of amplitudes for different electrons, and describes how electrons communicate about time.

Time ordering is generally thought to be needed to understand the difference observed in double ionization of atoms by protons and anti-protons at high velocities. Nonetheless, there has been no previous direct experimental evidence for time correlation between different electrons. Our direct comparison of theory with data for ionization of helium with concurrent excitation of a second electron into various magnetic sublevels of the 2p excited state now provides evidence for correlation in the time evolution of different electrons. This means, for example, that one sequence of interactions may differ from another, as illustrated by the equation for T_{cor} given above.

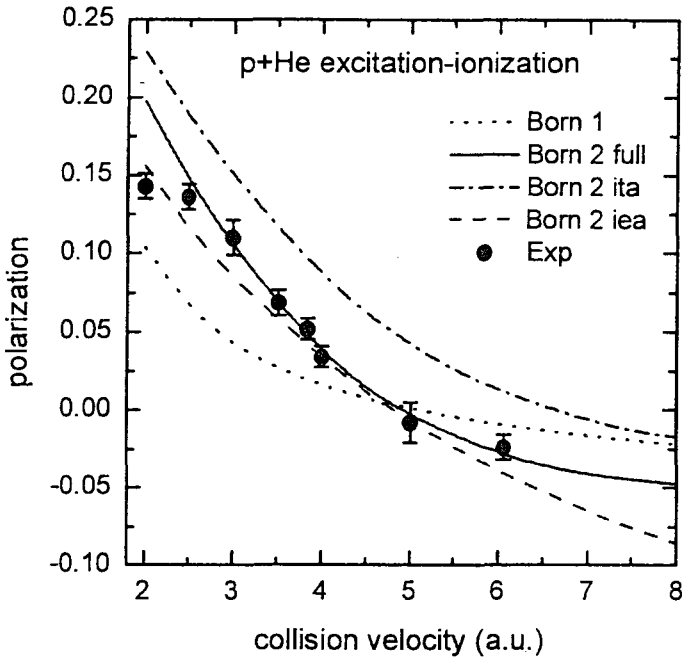

Fig. 2. Calculations with and without time correlation of electrons compared to experimental data. Here polarized light is emitted from helium following 1s – 2p excitation of one electron accompanied by ionization of the second electron. The polarization fraction is plotted as a function of the velocity of the incident proton. Born 1 is the first-order calculation that includes only spatial correlation. Born 2 (ita) is the second-order calculation without time correlation. Born 2 (iea) is the second-order calculation without spatial electron correlation. The full second-order calculation (Born 2 full) includes time correlation as well as spatial correlation. The data are from Merabet et al.

In figure 2 we present both data and theory for the polarization of light emitted from the 2p level of excited helium following excitation of one electron and ionization of another by an incident proton. It is evident that a first-order theory does not describe either the magnitude or the energy dependence of the polarized light observed. Second-order calculations in the independent time approximation (ita) and independent electron approximation (ita) provide some improvement, but still do not agree with the data. Only the second-order calculation including

time correlation between electrons is in good agreement with observation, except at the lowest energy shown, where it expected that higher order terms in V(t) are significant.

Other cases have also been considered [1,2], including examples where the independent time approximation works quite well. In many cases the effects of time correlation between electrons are small. This is fortunate in that the off-shell, time-correlated terms require more than one hundred times more computer time to evaluate than the simpler on-shell, energy-conserving terms. In calculations of dynamically correlated systems of particles, an independent time (or on-shell or wide band) approximation without time correlation is widely used to save computational time and effort. When this relatively easy independent time approximation is valid, calculations of relatively complex systems become feasible.

In summary correlations in the time evolution between electrons can be significant. This time correlation between electrons arises from energy-non-conserving quantum fluctuations occurring for short times in intermediate states of the system. Such time correlation between electrons may play a useful role in controlling quantum transmission of information via sequencing in complex multi-electron systems. We plan to extend this work to photon impact and to strong fields.

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REACH OUT AND CONTROL SOMETHING QUANTUM: LET THE WAVEPACKET DO THE THINKING

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Abstract:

Most quantum systems considered for control by external fields are plagued by a serious lack of complete information about the underlying Hamiltonian. In contrast, learning control techniques have a special role to play in the manipulation of quantum dynamics phenomena. The unique capabilities of quantum systems that make them amenable to learning control are (a) the ability to have very large numbers of identical systems for submission to control, (b) the high duty cycle of laboratory laser controls, and (c) the ability to observe the impact of trial controls at ultrafast time scales. Various learning algorithms have been proposed to guide this control process. This presentation will discuss these proposals, as well as some new perspectives.

Nonperturbative laser-atom interactions for nonlinear optics

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PROGRAM SCOPE

The goal of this work is to study of the interaction of atoms and molecules with intense and very short (<20 femtosecond) laser pulses, with the purpose of developing new short-wavelength light sources. We are also developing novel laser systems to enable this work.

PROGRESS TO DATE

We have made significant progress in a number of research areas supported under this grant. In work published in Nature in June 2000,[1,2] we demonstrated that we can use temporally shaped optical pulses to control and selectively enhance high-order harmonic generation. This work represents a milestone in the application of shaped optical pulses for coherent control of atomic wave functions, as well as representing a significant advance in the development of coherent radiation sources in the soft x-ray region of the spectrum. From the coherent control perspective, this work demonstrates for the first time that it is possible to use light pulses to both control and select very high-order nonlinear processes. Our work demonstrates that very subtle changes in the shape of a light pulse can dramatically alter the laser-atom interaction associated with high-harmonic generation. In more-recent work published in Chemical Physics and in Physical Review Letters,[3-5] we have developed a comprehensive theoretical description of this optimization process. This model elucidates the physics, and demonstrates that this effect is a control of the shape of the electronic wave function of an atom, on an unprecedented attosecond time-scale and angstrom scale-length. In the past few years, a "rescattering model" of high-intensity multiphoton ionization and harmonic generation has been established, where an electron liberated from an atom through high-field ionization can reencounter the parent ion, and scatter or recombine. Recent experiments have established that the quantum phase that the ionized electron accumulates during its trajectory influences the spectral characteristics of the emitted harmonic radiation. By applying an evolutionary feedback algorithm to a theoretical model of HHG, we allow the computer to recreate our experimental results. The excellent agreement between theory and experiment allows us to examine the microscopic physics of the process. In optimizing the shape of the laser pulse, we are "phasing" the x-ray emission that results from the periodic ionization and recollision process. On each half-cycle of the laser pulse,

a burst of laser field-induced ionization is followed by recollision x-ray burst. If we consider the emission of radiation at any particular harmonic "order," this emission is the result of a succession of bursts that can interfere constructively or destructively with each other, as shown in Fig. 1. Subtle reshaping of the pulse can optimize constructive interference between the emissions resulting from individual bursts—reshaping of the pulse on a time-scale that is short compared with the ~100 attosecond optical period of the high-harmonic radiation. In using shaped optical pulses, we are in essence demonstrating a new type of phase matching—a microscopic rather than macroscopic phase matching process. We are also spatially and temporally re-shaping the electronic wavefunction of an atom, so that it has an optimized dipole moment to enhance a particular harmonic.

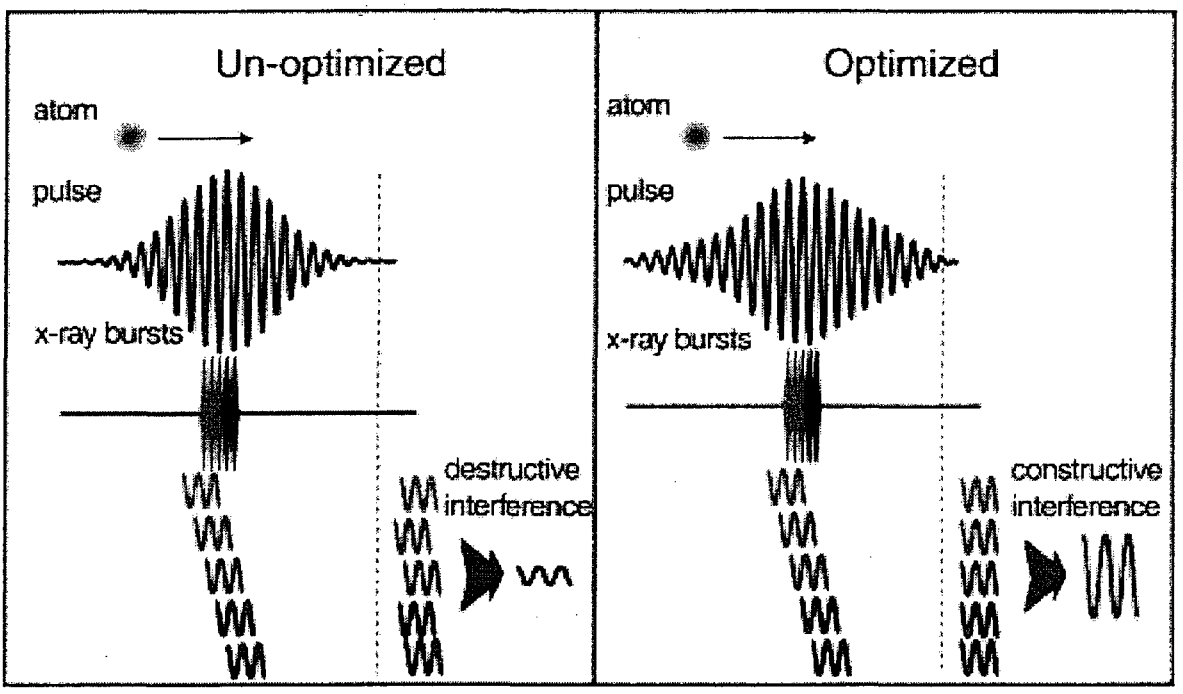

Figure 1: Schematic representation of intra-atomic phase matching.

In separate work recently published Physical Review Letters[6,7] we demonstrated for the first time that cascaded processes can be used to generate light efficiently in the VUV region of the spectrum. By generating 3rd harmonic light (3ω) very efficiently using a four-wave mixing technique we developed under prior DOE funding, we can cascade this process and use this 3ω light to in turn generate 4th and 5th harmonic light efficiently. This is possible only because we can generate 3ω light very efficiently, by using the long interaction lengths present in the hollow

core fibers, as well as the phase-matched geometry it allows. To demonstrate that we are using cascaded processes to generate this light, we used a novel multistage hollow-core fiber setup, where we can separately optimize the pressure in different sections of the fiber to phase-match different processes. This approach also allows us to optimize the efficiency of the cascaded process by more-sophisticated engineering of phase matching in this "gas-phase" nonlinear optics. A new type of quasi-phasematching was also observed in the VUV for the first time. Conversion using cascaded processes exhibits higher efficiencies, shorter pulse durations, and broader bandwidths than other schemes for generating light in the deep-UV, and will enable many applications in science and technology. This is a totally new type of "fiber-optics," applied to the EUV/ soft x-ray region of the spectrum.

In still other work,[8,9] we recently demonstrated an ultrafast laser system that generates an unprecedented-high average power for a small-laboratory-scale system, and that incorporates a pulse shaper for near-arbitrary optical waveform generation. This work recently appeared in Optics Letters.[9] This laser generates ~8 watts average power, in millijoule-energy pulses at 1-10 kHz repetition rates, and with ~20 fs pulse duration. This laser takes advantage of newly developed pump lasers that can generate large amounts (>80 Watts) of pulsed green light. The system we developed uses a cryogenically cooled single stage amplifier, and takes-up the space of about 1/4 optical table. We are currently building a high-harmonic generation test setup using this system.

FUTURE PLANS

During the coming year, we will work to demonstrate enhancement of x-ray generation at other wavelengths, and to understand the limits of this process. Better optimization algorithms will be developed, and modifications such as the use of cost functions (recently proposed by Hersch Rabitz) will be needed. We also believe that we may have demonstrated, for the first time, a nonlinear-optical frequency conversion process driven by EUV light; i.e. nonlinear optics in the extreme ultraviolet. We are currently doing experiments to try to verify our interpretation of this data. Finally, we hope to combine ultrafast laser amplifiers, phase stabilization techniques, and pulse shaping techniques to generate precisely controlled, phase stabilized, amplified pulses. This will allow us to enter a new regime of extreme nonlinear optics, enabling cycle-by-cycle control of light-matter interactions.

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Quantum Control of Time-Dependent Electron Correlation

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Program Scope

We use intense and/or short laser pulses to (1) manipulate and view time-dependent electron correlation within two-electron atoms; and (2) investigate and control strong-field ionization and fragmentation of atoms, molecules, and clusters. In the first line of experiments, coherent pulse sequences are used to generate doubly-excited Rydberg wavepackets in atoms with two valence electrons (i.e. alkaline-earth atoms). By controlling the excitation laser pulse (or pulse sequence), we can manipulate dielectronic dynamics and alter the branching ratio for electron ejection into different energy and angularly resolved continua.

In the second line of experiments, intense laser pulses are used to ionize and fragment atoms, molecules, and clusters. We are studying the role of electronic structure and electron correlation in strong field processes such as enhanced multiple ionization of atoms, population trapping via AC Stark shifted resonances in atoms, and the apparent suppression of ionization in diatomic molecules with triplet ground states. We are also examining the competition between laser induced ionization and fragmentation in larger molecules and small clusters. Our experiments compare and contrast process yields for different atomic and molecular species as a function of laser intensity, polarization, and wavelength. In addition, a liquid-crystal-based laser-pulse shaper is being used in conjunction with a genetic feedback algorithm to explore the enhancement and/or suppression of these different phenomena through the manipulation of the time-dependent laser field [1-3].

Progress During the Past Year

I. Manipulating Autoionization Lifetimes via Rydberg Wavepacket Control

We have demonstrated that the autoionization lifetime of doubly-excited Rydberg wavepackets can be significantly altered by controlling the relative phase of the constituent eigenstates in the wavepacket. In our experiments, a 1 psec laser pulse produces a coherent superposition of 4snd ($n \approx 40$) eigenstates in calcium. After a time delay, T, a 200 fsec laser pulse further excites the wavepacket to a 4pnd autoionizing configuration by driving a 4snd-4pnd isolated-core excitation (ICE). The time-dependent decay of the autoionizing 4pnd wavepacket is probed with a second, time-delayed, 200 fsec laser pulse which drives any atoms remaining in the 4pnd states to a 4dnd configuration. The higher energy electrons emitted during the decay of the 4dnd autoionizing states are readily distinguished from those ejected from the 4pnd levels.

We find that the 4pnd wavepacket decays in a "stair-step" rather than exponential fashion. Initially, the probability for finding the electron in the 4pnd doubly-excited state remains constant in time. However, after a timeτ, the 4pnd population drops suddenly to approximately 20% of its initial level. The population then remains constant for a time equal to one Kepler period, τ_K, of the Rydberg electron before suffering a second 80% reduction. The lifetime, τ, has a strong dependence on the delay T between the laser which initially produces the wavepacket and that which drives the 4snd-4pnd ICE. For the 4p40d wavepacket, we observe autoionization lifetimes with 1.5 psec $< \tau <$ 10 psec, and a functional form, $\tau = \tau_K - (T \bmod \tau_K)$.

Of course, the phase of each eigenstate in the coherent superposition evolves at a rate that is proportional to its binding energy. Therefore, our observation of the decay of the 4pnd wavepackets as a function of T is equivalent to a sequence of measurements of electron emission from a series of wavepackets, each having a different eigenstate phase distribution at the instant of its excitation [4]. Unfortunately, inspection of the phase distribution among the constituent eigenstates provides no obvious clues as to the source of the striking variation in the autoionization lifetimes. Instead, a semi-classical picture combined with the known time-dependence of the wavepacket's radial probability distribution provides an explanation. Autoionization occurs when the Rydberg and core electrons collide and exchange energy. Since the radial extent of the core electron's wavefunction is only a few atomic units, the Rydberg electron must be near the nucleus for autoionization to proceed. For small values of T, the Rydberg wavepacket has just been launched in a shell of probability density that travels *away* from the nucleus. As a result, no autoionization can occur and the 4pnd probability remains constant until the electron returns to the ionic core one Kepler period later. For $T = \frac{1}{2}\,\tau_K$, the Rydberg wavepacket is at its outer turning point and requires a time $\frac{1}{2}\,\tau_K$ to return to the nucleus. Therefore, the initial plateau in the decay curve is expected to have a duration of $\frac{1}{2}\,\tau_K$, as is observed. For T slightly less than or slightly greater than an integer multiple of τ_K, the electron is just approaching or just leaving the nucleus at the instant of the ICE, and the autoionization lifetime is approximately zero or τ_K, respectively. Time-dependent multi-channel quantum defect theory (MQDT) calculations based on ab initio K-matrices [5] confirm this intuitive picture and are in excellent quantitative agreement with our experimental results. A manuscript detailing the experimental and theoretical work is nearly complete.

II. Suppression of Ionization in Molecules with Triplet Ground States

Intense-laser ionization of molecules is an area of investigation that is motivated by basic science and practical concerns. While atomic ionization is relatively well described by single-active-electron tunneling or multiphoton models, in general, molecular ionization and fragmentation yields cannot be accurately predicted at this time. We have been investigating a relatively simple problem, strong-field ionization of diatomic molecules. Recent experiments have shown that while N_2 ionizes in accordance with the predictions of atomic tunneling models, the ionization of O_2 is relatively suppressed [6]. New theoretical work [7] suggests that an electron interference effect, similar to that observed for coherent light in a Young's double-slit configuration, is responsible for ionization suppression that is seen in O_2 but not in N_2. The model assumes that ground-state electron density is preferentially located at the site of the two nuclei. During intense-laser ionization, the electron amplitudes emitted from each atomic-core interfere, producing a modified angular distribution of electrons in the far-field. For low energy emission, the electron de Broglie wavelength is larger than the internuclear separation. As a result, the interference is isotropic, either completely constructive (for symmetric electronic ground-states) or completely destructive (for anti-symmetric ground states). Consequently, Muth-Bohm *et al.*[7] predict suppressed ionization of molecules with anti-symmetric electronic ground states in general, and F_2 in particular.

We have measured ionization probability vs. laser intensity for D_2, N_2, O_2, F_2, S_2, SO, NO, CO, and HD, and compared these to the ionization rates of companion atoms, Xe, Kr, or Ar, which have similar ionization potentials. We do not observe the predicted ionization suppression in F_2, a molecule with an antisymmetric singlet ground state. However, we do observe very strong suppression in S_2 and SO, molecules that, like O_2, have antisymmetric triplet ground states. In fact, the suppression of ionization in S_2 and SO is significantly greater than that observed in O_2. Yet, according to the interference model, ionization suppression in these molecules should be reduced relative to O_2. Due to the larger internuclear separations in SO and S_2, only the lowest energy electron channels will experience complete destructive interference. Our measurements indicate that molecules with singlet electronic ground states are not suppressed while those with doublet or triplet ground states are. We have no simple interpretation for this observation at this time. A manuscript describing the N_2, O_2, F_2, and S_2 work has been submitted to Physical Review Letters and a longer manuscript detailing the more comprehensive measurements is in preparation for submission to Physical Review A.

III. Control of Intense-Laser Fragmentation of Clusters

When molecules and clusters are subjected to intense laser fields, both ionization and fragmentation of the molecule can occur. In some applications, e.g. mass spectrometry, it may be desirable to produce only the parent molecular ion or specific charged fragments. However, due to the complexity of these systems, it is not generally possible to know *a priori* the specific laser characteristics that will maximize the desired yield for a particular target. Therefore, laser-pulse shapers and feed back algorithms[1] have recently been employed to control the relative fragmentation yields in large organo-metalic[2] and hydro-carbon species[8].

We have begun an investigation of intense laser fragmentation of S_8 cluster rings. Sulfur clusters provide interesting targets since they are atomically homogeneous and the ionization potential of the species, S_n ($1 \leq n \leq 8$), are approximately identical, 9.5 ± 1 eV. With low intensity (10^{11} W/cm^2) unshaped 100 fsec pulses, essentially all singly-charged fragments S_n^+ are seen, but the parent molecular ion, S_8^+ dominates the spectrum. However, with increasing intensity, smaller fragments are produced with higher efficiency. Above 10^{12} W/cm^2, S^+ and S_2^+ are essentially the only singly-charged species observed. Furthermore, if the pulse energy is held fixed while the pulse length is increased, either by chirping the pulse or restricting its bandwidth, lower mass species are produced with increased efficiency. These observations appear to be relatively insensitive to wavelength.

In an attempt to control the manner in which these clusters fragment, we enabled feedback control of the laser parameters using a genetic algorithm and liquid-crystal-based pulse-shaper. Although our observations are very recent and not fully analyzed at this time, we present some preliminary results. First, with phase-only shaping, transform-limited pulses appear to produce the maximum yield into any individual charged fragment. However, we have been able to enhance the *relative* yields of various charged fragments, e.g. S_2^+/S^+, by more than 100%. In maximizing the yield of heavier species relative to lighter species, the most effective pulses appear to be nearly transform limited with small but important modulations in the temporal wings of the pulse. In maximizing the ratio of lighter to heavier species, chirped pulses with small temporal modulations

appear to work best. Second, with phase and amplitude shaping 200% enhancements have been observed for various yield ratios. More importantly, these large enhancements are not accompanied by a significant decrease in yield for the primary species of interest. When attempting to maximize the ratio of heavier to lighter species, the algorithm typically selects pulses with lower energy but with temporal features similar to those produced with phase-only shaping. Not unexpectedly, maximizing lower to higher mass ratios leads to the selection of slightly chirped pulses with maximum pulse energy. More analysis is needed to determine the significance of the small temporal variations that appear to be important for the yield ratio enhancement of various species.

Future Plans

In the coming year, we intend to continue our investigations of strong-field phenomena in atoms, molecules, and clusters. First we will perform extensive analyses of our S_8 fragmentation results to determine if any physical insight into the fragmentation dynamics can be gleaned by comparing the characteristics of the different pulses that are found by the genetic algorithm to be the most "fit." We also intend to investigate the negative impact of spatial intensity variation on the effectiveness of pulse-shaping control in systems that possess many intensity-dependent "reaction" paths. Second, we plan to measure the role of electron correlation in trapping, via transient AC Stark-shifted resonances, electron population in highly-excited ionic states during intense-laser ionization of atoms [9]. We will explore population trapping in noble gas, alkali, and alkaline-earth systems where the first two ionization thresholds are both large, small and large, and both small, respectively. Third, we will begin construction of an amplified 20 fsec Ti:Sapphire laser that will enable us to investigate molecular fragmentation in a shorter pulse regime. The laser will be used in conjunction with a dual, multi-hit, spatially sensitive, charged particle spectrometer that is currently being built. The spectrometer is nearly identical to instruments used by other groups for COLTRIMS experiments, and will allow the determination of the energy and momenta of all charged particles produced via laser fragmentation of single molecules. These complete experiments will provide much needed, in-depth probes of molecular dynamics in intense fields.

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Ground state dissociation (GSD) of HD^+

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The fast removal of one electron from a hydrogen molecule is followed by very slow dissociation if the vibrational continuum of the electronic ground state is populated in this vertical transition. For the HD isotope this happens in about 1% of all transitions to the $HD^+(1s\sigma)$. For this isotope of hydrogen the dissociation favors $H^+ + D(1s)$ over $H(1s) + D^+$ production due to the small, 3.7 meV, energy gap between these two dissociation limits[1,2]. We have used this slow dissociation process of hydrogen to probe a couple of reaction mechanisms:

1. Charge transfer and elastic scattering in very slow $H^+ + D(1s)$ "half" collisions

We have studied charge transfer and elastic scattering in the $H^+ + D(1s)$ collision system. This system provides an interesting case study for theorists, since scattering calculations for this system depend in turn on HD^+ structure calculations, which must correctly account for the difference in nuclear mass. Our aim was to study the collision at energies from about 1 eV down to the charge transfer threshold at 3.7 meV, and below for the elastic channel. To accomplish this goal we used GSD of HD^+ to measure electron transfer from the $1s\sigma$ to the $2p\sigma$ state. Charge transfer occurs at an internuclear separation of about 12 a.u., around the avoided crossing resulting in the $H(1s)+ D^+$ final state. In contrast, elastic scattering leads to the $H^+ + D(1s)$ final state. The energy of the resulting H^+ or D^+ fragment (typically less than 300 meV) is determined for each fragment by imaging its momentum vector using a COLTRIMS-style apparatus.

The measured relative yields of H^+ and D^+ fragments as a function of kinetic energy in the HD^+ center of mass frame provide a direct measure of the electron transfer probability from the initial $1s\sigma$ to the final $2p\sigma$ state. This experimental approach provides a probe of very slow collisions. It is important to note, however, that during the GSD process the "avoided crossing" is traversed only once in contrast to twice in a "full" collision. As a result of the "half" collision nature of our experiment, we cannot compare directly to theoretical results available for full collisions[3]. Therefore, we performed coupled channels calculations of the transition probability for both half[2] and full collisions[3]. The experimental results are in good agreement with the "half" collision calculations except near the charge transfer threshold where better resolution and H_2 contamination subtraction are needed[2,4]. Improvements of the experimental setup are underway in order to probe the threshold behavior of charge transfer and the resonances in the elastic channel.

2. Momentum transfer to the hydrogen molecule CM or internal motion of its nuclei

In a collision, momentum can be transferred either to the CM motion of the molecule or to the internal motion of its nuclei in the CM frame. We have used the GSD process to probe and separate these two contributions from each other for slow $He^+ + H_2$ (or D_2) collisions (at a velocity of 0.25 and 0.5 a.u.). The GSD fragments are very sensitive probes of small amounts of momentum transfer because of their small dissociation speed. It was found that most of the momentum is transferred to the molecule as a whole for both electron capture and ionization processes. However, we observed some transverse-momentum transfer to the motion of the nuclei in the CM system for the ionization process and none for electron capture. It is suggested that direct scattering off one nucleus is the cause of this momentum transfer.

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Single Molecule Vibrations and Spatially Resolved Chemistry

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Inelastic electron tunneling spectroscopy with the STM (STM-IETS) allows us to reach the limit of sensitivity of vibrational spectroscopy, that of a single bond. While this represents an approximate nine orders of magnitude increase in sensitivity, a uniqueness of STM-IETS lies in the scanning capability of the microscope for providing the spatial distribution of the vibrational intensity with sub-Ångström resolution. An understanding of the intensities of vibrational features in STM-IETS depends critically on theoretical calculations and is emerging. The onset of inelastic tunneling associated with the excitation of a vibrational mode leads to a reduction of the elastic conductance. While this phenomenon is analogous to the Debye-Waller factor in the temperature reduction of the Bragg reflection intensities in x-ray, electron, and neutron diffractions where many phonons are involved, STM-IETS reduces this complexity to the emergence of a single vibrational excitation. Such theoretical analysis together with the experimental data provide a novel way to probe the coupling of electrons to the nuclear motions, which is the basis for many chemical and biological processes.

By combining single molecule vibrational spectroscopy with the unmatched imaging, manipulation, and chemical modification capabilities of the STM, chemistry can be probed at the spatial limit. Direct visualization of the nature of the chemical bonds and their transformations at the single molecule level not only provides convincing evidences but also fundamental understandings of chemical processes. The STM junction is effectively a nanoreactor in which the metallic tip and the substrate work together to induce chemical transformations of individual molecules adsorbed either on the substrate or the tip. Many aspects of chemistry can be probed by the STM, including the rotational, vibrational, and translational motions, the conformation changes, the energy transfer, the electrical conductivity, and bond breaking and formation of individual molecules. The STM images provide a direct view of the landscape in which experiments are carried out. Such information removes many ambiguities associated with either indirect or unknown knowledge of the working environment.

Since STM-IETS is spatially resolved, the energy, intensity, and lineshape of the vibrational features can be obtained at known points within a single molecule. While the study of isolated, single molecules is of fundamental interest, environmental effects are important as the molecules interact with each other to form systems with increasing complexity. By measuring the vibrational spectra to extract energy shifts, intensity changes, and lineshape variations for different separations between two adsorbed molecules, it is possible to address the question on how close the two molecules need to be in order for them to affect each other and for a reaction to occur. These intermolecular induced changes provide unique insights into the nature of chemical interactions and the elastic and inelastic couplings of electrons with the molecular vibrations. Since electron tunneling involves the overlap of electronic and vibrational wavefunctions of the tip, molecule, and the substrate, the STM is uniquely positioned to reveal the inner workings of molecules.

State-Resolved Probes of Photo-Induced and Thermal Reactions on Ag Surfaces

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State-resolved laser spectroscopy is being used to probe the internal-state, velocity and spatial distributions of molecules desorbed from metal surfaces resulting from photoexcitation of the substrate or from thermal reaction of adsorbed species. For simple photo-induced desorption, the final state distributions of the products reflect the energy transfer mechanism leading to desorption, whereas for thermal association reactions, such information can be used to obtain state- and energy resolved sticking coefficients via detailed balance. Generally, such measurements provide insight into the non-adiabatic coupling of the electronic and nuclear degrees of freedom of the adsorbate *and* substrate atoms which is unique to interfacial chemistry. Results will be presented for ongoing studies of photodesorption and photofragmentation of weakly-bound adsorbates on Ag(111) and thermal association reactions on oxygen-reconstructed Ag surfaces. Recent efforts towards the development of an ion imaging spectrometer for measuring the spatial distributions of desorbed neutrals based on velocity mapping will also be presented.

State-Resolved Photoionization Dynamics of HCO and DCO

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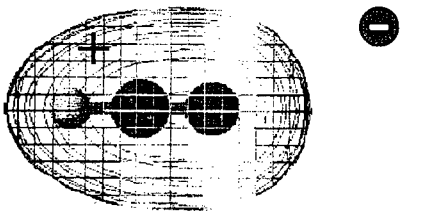

Neutral HCO appears as a central intermediate in mechanisms for the formation of photochemical smog. The cation, HCO^+, is among the most abundant charged polyatomics in the interstellar medium. Both the ion and neutral have important roles to play in the chemistry and thermodynamics of combustion, and, for many investigators, the radical signifies a collision complex in the reaction of H with CO. With their closed-shell cation core structure and convenient Rydberg-Rydberg accessibility, the higher excited states of HCO also present an ideal prototype for the study of electron-cation scattering dynamics in polyatomic molecules. Achieving an understanding of these dynamics is an ultimate aim of the present work. In the process of pursuing this goal, we have gathered a significant amount of spectroscopic information on the lower-lying excited states of the neutral and cation. This talk will present new results, using laser-assisted (1+1')-photon ionization spectroscopy, that characterize the combined effects of Renner-Teller coupling and Fermi resonance in this molecule's lowest Rydberg state ($3p\pi\ ^2\Pi$). Systematic double-resonance scans from selected levels of this intermediate state serve to decode the irregular structure of higher vibrational autoionizing states. Finally, from rovibrational-state detailed limits, we establish transition energies and anharmonic terms for higher vibrationally excited states of the ions, HCO^+ and DCO^+.

IONIZATION PROBES OF MOLECULAR STRUCTURE AND CHEMISTRY

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Photoionization processes provide very sensitive probes for the detection and understanding of molecules and chemical pathways relevant to combustion processes. Laser based ionization processes can be species-selective by using mass separation, resonances in the excitation of the neutral molecule under study, or by exploiting the fact that different molecules have different sets of ionization potentials. Therefore the structure and dynamics of individual molecules can be studied, or species monitored, even in a mixed sample.

The exploitation of Rydberg molecules has enabled orders-of-magnitude increases in the resolution available for recording the spectra of molecular ions. These spectra provide information equivalent to photoelectron spectra, but contain much more information by virtue of that resolution and the versatility of laser preparation of the states involved.

We have developed techniques called mass analyzed threshold ionization spectroscopy (MATI) and photoinduced Rydberg ionization spectroscopy (PIRI) to provide high resolution access to the spectroscopy of the electronic states of ions. To accomplish this we create high Rydbergs state just below an ionic threshold. A small field is used to separate the prompt ions from the Rydberg molecules and then after a delay of a few microseconds either a small electrical pulse field ionizes the Rydbergs (MATI) or a tunable laser beam is sent through the Rydberg molecules (PIRI). In the latter, if this laser is resonant with a transition of the ionic core (an isolated core excitation, or ICE), core-excited Rydberg molecules are created which promptly autoionize. These ions are again separated from the remaining Rydbergs and after a further few microseconds the various ion packets are sent into a TOF mass spectrometer, where they arrive as a distinct groups whose intensity can be recorded as the either the Rydberg preparation laser or the final laser is scanned. The resonant nature of MATI and PIRI are of great use in sorting out the vibrational structure of some ionic states.

The ground state and first excited state of benzene cation both exhibit substantial perturbations on the vibrational structure due to the Jahn-Teller effect. MATI and PIRI provide the means to study both of these states with the advantage that the multiple-resonant nature of the spectroscopy enables the absolute symmetry assignments of many of the vibrational transitions.

These two states, while both of necessity doubly degenerate, have different symmetries, E_{1g} and E_{2g}. Fundamental considerations from the model used to construct Jahn-Teller theory predict that first order coupling should be strong in the former and weak in the latter. These considerations have been somewhat ignored in the recent literature, and have never been tested experimentally.

In the $^2E_{2g}$ state, there is relatively strong quadratic or pseudo- coupling in every degenerate vibration that is identifiable. However, for mode 6, which should be the major vibration to exhibit a linear coupling, this drops from 0.4 in the ground state to 0.005 in the $^2E_{2g}$ state. For this mode, the quadratic effect is much larger than the linear one. This result establishes the general principle that only states that have a wavefunction with a single nodal plane through the principle axis (and therefore one quantum of pseudo-angular momentum) have a strong linear coupling. The ground state fulfills this condition and has a strong linear coupling, while the $^2E_{2g}$ state has two nodes and the coupling is weak. This principle should extended other molecules.

Nondipole Effects on Photoelectron Angular Distributions

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Atomic photoionization processes have been extensively explored by comparing theory with measurements of partial photoionization cross sections σ and photoelectron anisotropies β as functions of photon energy. Even in the independent-particle approximation, variations of σ and β with photon energy occur due to maxima and minima in dipole matrix elements and variations of phases of transition amplitudes. Many-electron interactions are also strongly manifested in σ and β in two general ways. The interaction of discrete states with continua produces rapid oscillations of σ and β over narrow energy ranges near ionization thresholds, and continuum-continuum coupling can modify σ and β over broad energy ranges. Relativistic interactions must also be treated in certain cases. New aspects of the photoionization of atoms, molecules, and materials have been investigated in recent years with studies of *nondipole photoelectron asymmetries*. Interference between electric-dipole (E1) transition amplitudes and amplitudes for higher-order interactions such as electric quadrupole (E2) produce asymmetries between the intensities of photoelectrons emitted forward and backward with respect to the photon propagation vector **k** (pure E1 angular distributions are forward-backward symmetric). Similar to σ and β, forward-backward asymmetries vary with energy due to one-electron and many-electron interactions and provide additional insight into photoionization processes.

We have developed a photoelectron spectrometer system using four parallel-plate analyzers to accurately and efficiently measure forward-backward asymmetries of atoms and molecules. Using hard x-rays at Argonne's Advanced Photon Source, we measured forward-backward asymmetries of Kr 1s photoelectrons from threshold to 8 keV kinetic energy and found excellent agreement with theory. We also searched for extra-atomic effects in the asymmetries of Br 1s photoelectrons from molecular Br_2 and $CBrF_3$. Recent theoretical studies predict surprisingly strong forward-backward asymmetries in low-energy ($\approx$10-200 eV) photoionization of valence electrons due to quadrupole autoionizing and shape resonances and variations of dipole amplitudes. These effects will be studied in experiments on a VUV beamline at the Synchrotron Radiation Center in Stoughton, Wisconsin.

ELECTRON/PHOTON INTERACTIONS WITH ATOMS/IONS

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Our presentation will be on sections A and C, focusing mainly on understanding measurements.

A. Generalized Oscillator Strengths for Atomic Transitions

A.1. Correlation Effects in the Generalized Oscillator Strengths of Noble Gas Atoms

The generalized oscillator strength (GOS) concept, introduced by Bethe [1], manifests directly the atomic wave functions and dynamics of atoms electrons. The interest in the GOS has been extensive and include the probing of the intricate nature of the valence- and inner-shell electron excitations [2]. We have investigated correlation effects in the GOS's of noble gas atoms, for the range of momentum transfer $0 \leq q < 2$ a.u. and energy transferred $0 \leq \omega < 120$ Ry [3]. The calculation used the one-particle Hartree-Fock approximation and with account of many-electron correlations using the random phase approximation with exchange (RPAE). Correlation effects have been found to be important in all dipole, monopole and quadrupole channels. Particularly important are the many-electron correlations at high q values, where new additional purely correlational maxima and minima appear.

A.2. Reinterpretation of the Recently Measured Absolute GOS for Nondipole Transition in Ar

The recent first experimental observation of the absolute GOS for Ar 3p-4p nondipole transition has been interpreted as a manifestation of quadrupole excitation [4]. Contrary to the experimentalists' assignment, we attribute the measured GOS to combined monopole, the dominant component, and quadrupole contributions [5]. Our RPAE results could have significant implications for other similar transitions, previously interpreted as quadrupole excitation and for interpreting other discrete transitions.

A.3. Minima and Maxima in GOS's of the Noble Gases and Na Resonance Transition

The GOS's for the transitions from ground state to the excited states $np^5(n+1)s$ of Ne (n=2), Ar (n=3), Kr (n=4) and Xe (n=5) have been calculated as a function of the momentum transfer squared using the RPAE and Hartree-Fock Approximation. The characteristic minima and maxima in the GOS's are in excellent agreement with measurements. For the noble gases, exchange and correlation effects are found to influence the positions of the minima insignificantly [6]. However, for Na 3s-3p transition the effects are important [7]. These minima could be important in studying nondipole effects as Cooper minima are in magnetic dichroism in atomic photoionization [8].

B. Nondipole Parameters in Angular Distributions of Electrons in Photoionization of Noble-Gas Atoms

The parameters that determine the nondipole (E1-E2) corrections to the photoelectron angular distribution have been investigated for the *s*- and *p*- subshells of the noble-gas atoms through comparing results calculated [9] in Hartree-Fock (HF) approximation and taking into account multielectron correlations, using the random-phase approximation with exchange. Our results cover the photoelectron energy range ε from the photoionization thresholds to 1.6 keV. We find the interesting result that near the photoionization thresholds these parameters are generally characterized by an oscillatory behavior as a

function of ε, exclusive of the parameter for the He 1s subshell. These oscillations are sensitive to multielectron correlations, except those of the Ar 3p subshell. This finding supports once more that the photoprocesses in these atomic subshells are of a collective character. We conclude that their correct description cannot be achieved within the framework of a one-electron approximation such as the HF approximation.

C. Regge Poles Description of Atomic Collision Processes

The recent successful Regge poles description of small angle electron scattering [10] and attendant normalization of the measured relative electron differential cross sections (DCS's) [11] as well as the novel Padé-Regge pole analysis of chemical reactions through analytical continuation of the S-matrix in the complex plane of the total angular momentum [12], have inspired the development of simple, accurate and efficient analytical methods to calculate Regge poles trajectories [13,14]. Two such theoretical approaches are presented, focussing particularly on a new simple semi-classical approach, based on the investigation of Stokes lines topology, to calculate Regge poles positions for singular potentials, viz. potentials more singular than r^{-2} at the origin. The method uses the solution of the Bohr-Sommerfeld quantization condition with the proviso that the positions of two turning points of the effective potential responsible for the Regge poles be relatively close together. Experimental small angle electron DCS's are analyzed and illustrative results are presented for both the polarization and Lennard-Jones potentials.

D. Electron/Photon Impact Studies of Ionized Atomic Systems

D.1 Correlation and Relativistic Effects in Fe X and Fe XV

We have investigated correlation and relativistic effects on fine-structure energy levels and oscillator strengths in Fe X using an extensive configuration interaction (CI) wave function with relativistic effects incorporated through the Breit-Pauli Hamiltonian. We found that the inclusion of the crucial correlation effects arising from the two-electron excitation configurations $3s^23p^33d^2$ and $3s3p^43d^2$ in the CI expansion resolved all but two of the energy discrepancies between a recent calculation and experiment [15].

Effective collision strengths for transitions among the energetically lowest 10 fine-structure levels belonging to the $(1s^22s^22p^6)$ $3s^2$, 3s3p and $3p^2$ configurations of Fe XV have been calculated in the electron temperature range of 10^5 to 10^7 K [16] using the recent Dirac atomic R-matrix code of Norrington and Grant. Large differences are observed for many transitions over almost the entire temperature range between our values and those of Eissner *et al* [17]. This is a continuation of our earlier work on Fe XV [18-20] in which we have investigated fine-structure transitions and contrasted our data with those of Griffin *et al* [21] and Eissner *et al* [17]. We have found [18] that the collision strengths of Ref. [21] are unreliable and the effective collision strengths of Eissner *et al* [17] generally underestimate our results, due mainly to deficiencies in their calculations near the lower threshold region. Resonances arising from the $3s4\ell$ and 3p4s configurations have not been included in the calculations of Ref. [17].

D.2. Strengths and Limitations of the R-Matrix Multichannel Quantum Defect Theory with Frame Transformation: Application to Ni^{14+}, Fe^{14+} and Neon-Like Fe

We continue our efforts to delineate the strengths and limitations of the R-matrix multichannel quantum defect theory with frame transformation. This method has proven to be adequate in describing relativistic effects in outer- and inner- shell photoionization of atoms and positively charged ions [22, 23],

thereby avoiding the computationally exacting Breit-Pauli and Dirac R-matrix methods. Recently, in contrast to previous work, we have focused on photoionization from an open-shell system $3s^2 3p^2(^3P_0)$, where there is significant term-coupling $3p^2(^3P_0)$ - $3p^2(^1S_0)$ in the initial state. A comparison of LS, Breit-Pauli and LS - JK frame transformation R-matrix calculations quantifies both initial and final state relativistic effects in the near threshold photoionization [24].

D.3. Electron-Impact Ionization of Mg^+ near the 2p Inner-Shell Edge

Ionization of an atom or positive ion by electron impact may occur directly by the emission of an outer electron, or indirectly by ejecting an inner-shell electron. In the case of Mg^+ the threshold for such ejection is about 50eV for the 2p subshell. The interference between the direct and indirect processes give rise to resonance structures. New R-matrix calculations have been performed on the L-shell processes in the electron-impact ionization cross section of Mg^+. Resonances arising from the interference of inner-shell and outer-shell channels have been quantified [25] and the $2p^5 3s^2 nl$ series discussed. Our background cross section agrees well with previous calculations and with the structure obtained in recent measurements. The main contribution is to unify the direct and indirect processes within a single calculation, leading to a proper treatment of resonances.

PLANNED / ONGOING RESEARCH ACTIVITIES

- Development of an optimal scattering angle algorithm for reliable measurement of small-angle DCS's.
- Our investigation of exchange forces in dispersion relations using circuit relations will be extended to the multichannel Static Exchange Approximation
- The application of external fields to the e-H scattering problem could conceivably move the triple pole closer to the positive real axis, thereby enhance its effect on measurable quantities.
- Many S-matrices exist for heavy atom- atom reactions. We will investigate possible identification of resonances in atom- atom collisions.
- We will use our present methods to investigate small-angle electron-atom scattering; even the most recent experiments on e- Kr still measure DCS's, down to only 10° (J. Phys. B**33**, 1895 (2000); - B**33**, 1921 (2000)).
- Our singular potential method will be used to investigate Bose-Einstein condensation and superfluidity of ^4He at extremely low temperatures as well as the possibility of forming dimer resonances in He -He collisions.
- Collision strengths for transitions among fine-structure levels in ions of importance to astrophysics and fusion plasmas will continue to be investigated, as well as correlation and relativistic effects in photoionization of positive ions.

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MODE-DEPENDENT PHOTOIONIZATION DYNAMICS IN SMALL MOLECULES

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The study of molecular photoionization and photodissociation dynamics can provide considerable information on the mechanisms responsible for the flow of energy and angular momentum among the electronic, vibrational, and rotational degrees of freedom in isolated molecules. In addition, the highly excited resonant states that decay by autoionization and/or predissociation can be viewed as reaction intermediates or activated complexes of a number of elementary chemical and physical processes. The study of these decay mechanisms can thus provide insight into these processes, which include dissociative recombination, ion-pair formation and ion neutralization, inelastic electron-ion scattering, and simple chemical reactions of neutral species. In small molecules, strong mode-specific effects are often observed in the decay dynamics, that is, the decay processes are found to depend strongly on the electronic, vibrational, and rotational quantum numbers of the initially prepared excited state. For example, in the highly excited states of nitric oxide, the branching between autoionization and predissociation depends strongly on the electronic symmetry and rotational quantum numbers of the excited state. Such mode-specific effects are sometimes found to persist in considerably larger molecules. I will discuss several of these mode-dependent effects using examples from recent work on ammonia, water, and nitric oxide.

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DISSOCIATIVE RECOMBINATION OF TRIATOMIC DI-HYDRIDES

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Our current research on electron-molecular ion interactions has centered on studies of dissociative recombination (DR) at the CRYRING heavy ion storage ring in Stockholm. We have continued measurements there of neutralizing fragmentation through DR of triatomic di-hydride molecular ions with low energy electrons, e.g., $H_2X^+ + e^- \rightarrow$ {X + H + H; XH + H; or X + H_2.} To develop a systematic base of experimental data, we have measured DR for H_3^+, H_2D^+, CH_2^+, NH_2^+, and H_2O^+. In all cases the three-body breakup channel dominates at 0 eV. These experiments have included measurements of fragmentation branching fractions, absolute cross sections, and three-body dynamics for vibrationally cold molecular ions interacting with electrons at relative temperatures (energies) down to ~ 10 °K (~ 1 meV).

In branching fraction measurements, molecular ions are merged with the electron cooler beam and ions undergoing neutralization pass undeflected through the next bending magnet on straight-line trajectories to a surface barrier silicon detector (SSD). Neutral atomic and molecular fragments that pass through a transmission grid placed in front of the SSD are detected and their individual total energies recorded, with the deposited energy identifying the mass of the fragment counted. The transmission grid provides a means of controllably isolating summed signals from separated fragment components which otherwise arise in the SSD from simultaneous multiple-particle hits. Branching fractions for systems we have studied to date are typified by results for OH_2^+ {O + H + H (73%); O + H_2 (8%); OH + H (19%)} - except for DR of NH_2^+ in which the H_2 two-body branch is significantly diminished. Previous measurements for DR of NH_2^+ at 0 eV had indicated no population in the N + H_2 decay branch. We have recently repeated these measurements with detector placement set to guarantee complete collection of all channels with the results: $NH_2^+ + e \rightarrow$ NH + H (41%); N + H_2 (4%); N + H + H (55%).

In studies of three-body DR kinematics, a multiple-particle, position- and time-sensitive detector system is used to image simultaneous hits of various fragments. Positions of the detected fragments are analyzed to yield component energies and angles of emission. For systems where energetically allowed electronic excitation is relatively simple, populations of the specific final-state branching have been determined from analysis of system dynamics. Recent results of these studies will be discussed.

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Near-Thermal Collisions of Multi-charged Ions with H and Multi-Electron Targets

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Electron capture by multi-charged ions from neutrals is important in many technical plasmas including those used in materials processing, lighting, ion source development, and for spectroscopic diagnostics and modeling of core, edge, and divertor regions of magnetically confined fusion plasmas. The ORNL ion-atom merged beams apparatus provides needed benchmark measurements for collisions between ions and ground state H or D. Relatively fast (q x keV) intense ion beams from the Multicharged Ion Research Facility (MIRF) are merged with fast (keV) neutral beams creating center-of-mass collision energies ranging from 20 meV/u to 5000 eV/u. Merged-beams measurements[1] have shown the importance of trajectory effects at near-thermal energies and are of sufficient precision to discriminate between theoretical approximations, e.g., in the molecular-orbital potentials used to describe the collision process. For several collision systems structure has been observed in the cross section but is not always reproduced by theory. The ion beams used are diagnosed at the MIRF facility for metastable contaminants by electron impact ionization measurements. Metastable contaminants have a dramatic effect on the measured cross section at near-threshold collision energies for endoergic reactions.

Recent measurements[2] of the electron capture cross section for Cl^{7+} + D shows a decrease in the cross section toward eV/amu collision energies, in contrast to the slightly increasing cross section observed for other 7+ ions. Fully quantal MOCC calculations performed at ORNL for N^{7+} + D also decreases toward lower energies and agrees with the Cl^{7+} measurements, suggesting that the Ne-like core of Cl^{7+} plays no significant role in the electron capture process. The fact that the cross section does not remain flat toward decreasing energies shows that the actual quasi-molecular structure and associated dynamics remain important. The proposed MIRF upgrade will allow measurements with heavier (e.g., Mo^{q+}, Fe^{q+}, ...) ions at eV/u energies to further explore mulitelectron core effects.

The duoplasmatron ion source on the merged-beams apparatus has been upgraded to a new Cs negative ion sputter source which allows measurements with a wide variety of neutral atom and molecular beams. Such neutral beams include Li, B, Na, Al, P, K, Ca, Cr, Fe... and molecular beams such as O_2, CH_2,.. The merged-beams technique is the only technique available to explore collisions at eV/u energies and below for these atomic targets. For vapor targets like Fe that can only be produced at high temperatures, the electron capture process is virtually unexplored. An interesting first measurement would be with He^{2+} + Li to try to observe in the total cross section the predicted (Shimakura, private communication) shape resonances formed due to the strong ion-induced dipole attraction between reactants. Measurements of such resonances would provide a new benchmark for theory which relies on accurate molecular potentials and a quantal description of the collision dynamics.

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E1-M1 Transitions: Damping Interference and Two-Photon Decay

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Under certain conditions atomic transitions can proceed by the combined action of electric dipole (E1) and magnetic dipole (M1) amplitudes. We will discuss two interesting cases of this which are being studied at the ATLAS accelerator at Argonne and at the SIS accelerator at GSI. One effect is the so-called damping interference of E1 and M1 amplitudes in the electric field quenching of the metastable 2s $^2S_{1/2}$ state in hydrogenlike ions, and the other is a rare two-photon decay mode of the heliumlike 2 $^3P_0$ level. This work has important implications for fundamental symmetries such as time reversal invariance and atomic parity nonconservation and it provides data needed to clarify atomic structure issues of current interest.

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X-ray Spectroscopy of n=3⟶ , 4⟶, and 5 ⟶ 1 Transitions in Heliumlike Ar, Ti, and Cr

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Scope of work: We have used the Livermore EBIT facility and the EBIT spectrometers to study x-ray spectra from 3 - 1, 4 -1, and 5 - 1 transitions in He-like Ar XVII, Ti XXI and Cr XXIII. EBIT uses a nearly monoenergetic electron beam compressed by a 3-T magnetic field to ionize atoms to high a degree, and to trap and excite these ions. The EBIT electron beam energy can be tuned to select for study various electron-ion interactions of interest to the exclusion of others. In this rather clean EBIT environment, we have made accurate determinations of line positions, cross sections, and resonance strengths. Our main interest is the atomic physics of highly ionized ion species, and our measurements provide data which are used in the development of spectral modeling codes, and in the development of density and temperature diagnostics for laboratory as well as for astrophysical plasmas. In this presentation we discuss some measurements we have carried out over the past three years.

1 Triplet to singlet intensity ratios for Kβ lines in He-like ions

The ratio of the intensity of the intercombination line, $1s3p\ ^3P_1$- $1s^2\ ^1S_0$ to the intensity of the resonance line , $1s3p\ ^1P_1$ - $1s^2\ ^1S_0$, has been measured for ions in the heliumlike charge state from Mg XI to Fe XXV. The ions were prepared and excited in the LLNL EBIT, using excitation energies just above threshold. At these experimental energies, direct excitation was expected to be by far the dominant mechanism for line formation. We have observed x-ray emission spectra with EBIT spectrometers which have a plane of dispersion that is perpendicular to the electron beam. We have compared our results with theoretical predictions and found that two different distorted wave codes significantly underestimate the measured values, and that the discrepancy between experiment and theory becomes wider at lower values of Z. Our results and the theoretical predictions are shown in Fig. 1.

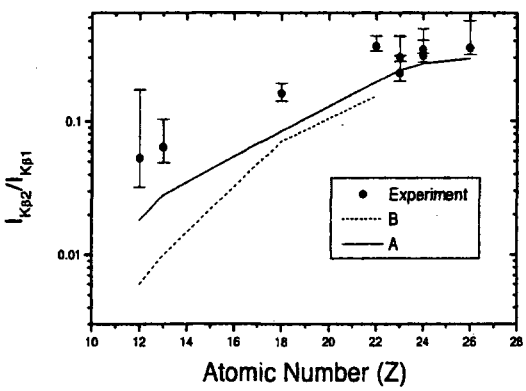

Figure 1. Ratio of the intensity of intercombination line to resonance line for He-like ions with Z from 12 - 26. The measured ratios are shown with error bars, and they lie mostly above the HULLAC code predictions line (A), and the predictions line from the Zang, Sampson and Clark code (B).

Part of the discrepancy can be accounted for by the uncertainties in the calculation of branching ratios. Since the upper states 1s3p $^3P_1$ and $^1P_1$ can decay to various states including 1s3s $^3S_1$, $^1S_0$, and 1s2s $^3S_1$, $^1S_0$ as well as to the ground state $1s^2\ ^1S_0$, it is important to calculate the fraction of the decays that are to the ground state. These branching ratios were calculated in MCDF theory as well as with the HULLAC code that uses distorted wave approximations (DWA). The two sets of branching ratios are significantly different from each other especially at low values of Z. Theoretical intercombination to resonance line intensities calculated with these branching ratios differ from each other and from experiment.

Uncertainties in branching fractions do not account for all of the discrepancy between experiment and theory. We note that branching ratios calculated to 1% using relativistic configuration interaction give results which lie between the HULLAC and MCDF values. Some of the discrepancy may also be due to difficulties in calculating cross sections in DWA, as well as to the transverse motion of electrons in the beam.

2 Dielectronic recombination in heliumlike Ar^{16+}

We have measured resonance strengths for $\Delta n \geq 1$ dielectronic recombination in heliumlike argon. DR is a resonant process in which a free electron is captured, and the kinetic energy plus the binding energy of the captured electron enable a bound electron to be simultaneously excited. The doubly excited state so formed stabilizes via the emission of a photon. In a KLM process, a free electron is captured into an M-shell while a K-electron is excited into an L-shell. Using a low resolution windowless Si(Li) detector we observe x-ray photons from the decay of either the M-shell or the L-shell electrons. We also observe x-ray photons from the process of radiative recombination into n=2. In addition to KLM, we have also observed the KLN, KLO, and KLP resonances in Ar XVII. For each resonance we obtain resonance strength for the n = 2 - 1 branch as well as for the n = j - 1, for j = 3 (KLM), 4 (KLN), 5 (KLO),and 6(KLP). We normalize the observed resonance strengths to radiative recombination cross sections, which like photoionization cross sections, can be calculated to an accuracy of 1%. Our measurements are in good agreement with MCDF calculations, and with similar measurements found in literature.

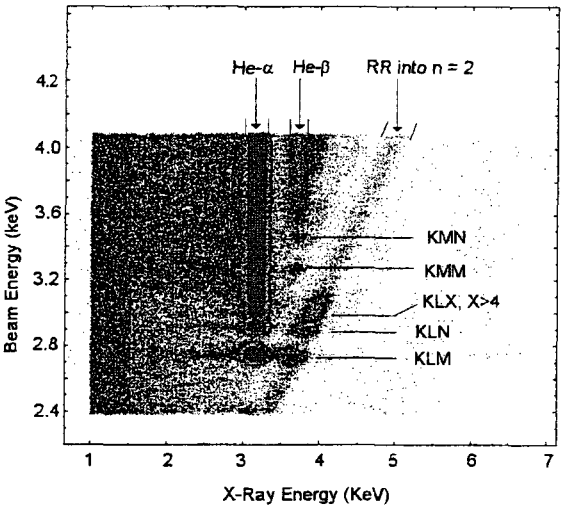

Figure 2. Scatter plot of x-rays emitted from Ar XVII ions shown as a function of x-ray energy and (EBIT) beam energy. Vertical lines represent lines excited directly by electron impact, angle lines represent radiative recombination, and intense dots represent dielectronic recombination resonances.

3 Dielectronic recombination in heliumlike Ti^{20+} and Cr^{22+}

We have extended our DR resonance strength measurements to higher-Z ions including Ti XXI and Cr XXIII, in separate measurements. Using the LLNL EBIT and a windowless Si(Li) detector we have observed the KLL, KLM, KLN and KLO resonances in these ions. As in the case of Ar, we sweep the electron beam energy and use a fast, multi-parameter event mode data acquisition system capable of archiving time, beam energy and photon energy for each photon event (see typical results for Ar XVII shown in Fig. 2). In the Ti XXI and Cr XXIII, we have also swept the electron beam current in addition to the beam energy in order to keep the electron-ion overlap constant.

4 Level specific DR resonance strengths in He-like Ti^{20+} and Cr^{22+}

We have measured the dielectronic satellite spectra for heliumlike Ti XXI and Cr XXIII, in separate experiments, using the LLNL EBIT and the EBIT high resolution Bragg crystal spectrometers. We sweep the electron beam energy across individual DR resonances, and thus we have measured level specific resonance strengths for the strongest DR resonances in doubly excited lithiumlike Ti XX and CrXXII. We have used the MCDF code to calculate the resonance strength, excitation energy, as well as the x-ray energy of these transition. We use various heliumlike or hydrogenlike lines excited directly by electron impact excitation and published data to wavelength-calibrate the spectrometers. Our measurements include not only the KLL resonances, which have been observed in tokamak plasmas for these and other ions, but also KLM and KLN resonances which to the best of our knowledge are being presented for the first time. We show a typical observed KLL spectrum and its comparison to theory in Fig. 3.

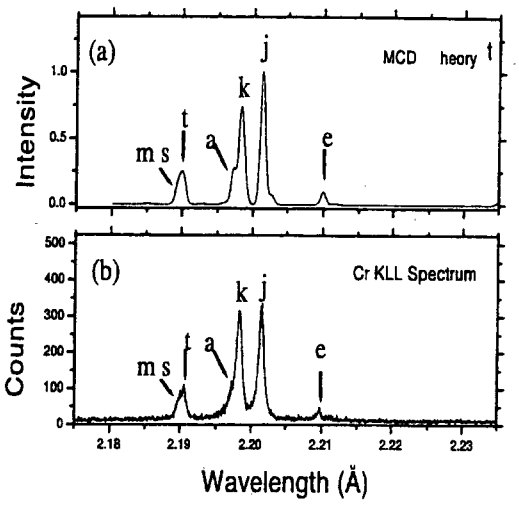

Figure 3. Dielectronic satellite spectra of heliumlike Cr^{22+} showing individual KLL resonances. The observed spectrum (b) was excited in EBIT and recorded with a high resolution Bragg crystal spectrometer using a LiF(200) crystal. The theoretical (a) spectrum was calculated with the MCDF code. Similar results have been obtained KLM, KLN and KLO resonances, and for Heliumlike Ti^{20+}.

5 Future Work

We plan to look more closely at the discrepancy between theory and experiment in connection with the ratio of the intercombination line to the resonance line in heliumlike systems. Since heliumlike systems are rather simple, it should be possible to make measurements that determine which models predict the best values for excitation cross sections and branching ratios.

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MEASUREMENTS AND SYSTEMATIC PREDICTIONS OF TRANSITION PROBABILITIES IN HEAVY COMPLEX IONS

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Program Scope and Definition

The exploitation of systematic regularities in precisely measured atomic data, both to make critical evaluations and to obtain interpolative and extrapolative predictions, is a primary means for achieving the high precision required for energy level and lifetime specification. These methods permit reliable specification of the properties of complex atoms by parametrizing a theoretical formula adapted from a simpler situation, wherein the increased complexity resides in the effective values for the parameters. The raw energy level data can be transformed by a one-to-one mapping onto the space of an effective quantum defect, a screening parameter, a core polarizability, an effective line strength, or some other parametric modulus. If this parameter can be expressed as a regular and slowly varying function of the charge state in an isoelectronic sequence, the principal quantum number in a Rydberg series, or some other attribute of a data subgroup, it can provide both high predictive power and insights into the dynamics of the complex many electron interactions.

The goal of this program is to enhance the available data base through carefully chosen precision measurements, and to systematize, categorize, and critically evaluate the data contained therein. Significant successes have been achieved in systematizing lifetime measurements for the resonance transitions in the alkalili-metallike, alkaline-earthlike, and inert-gaslike isoelectronic sequences of elements from low to medium Z. Most recently, the emphasis of these studies has been focussed in two directions. One area involves the investigation of extension of these methods from midrange isoelectronic sequences (40-50 electrons) to very complex systems (80 or more electrons). Here interesting dynamical effects have been observed to come into play in the vicinity of Z=60. Another area involves the development of new techniques for specifying branching fractions for multiply charged ions. There are necessary so that atomic lifetime data can be converted into transition probabilities, thus permitting systemization of line strengths of higher lying levels by these semiempirical methods.

Recent Progress

Lifetimes comparisons in the Pd and Pt sequences using selective excitation with a pulsed laser.

Earlier we made comparative homologous and isoelectronic studies of Ag II, Cd III, and In IV in the Pd I sequence, and of Au II and Hg III in the Pt I sequence using beam-foil excitation. Comparisons of these results were made with theoretical calculations using the Cowan code in the relativistic Hartree-Fock mode and a Superposition of Configurations code. There was general agreement between the experimental and theoretical results for the Pd sequence, but the experimental lifetimes were typically 30% longer than theoretical predictions for the heavier Pt sequence. While the beam-foil measurements BFS) made careful corrections for cascade repopulation from higher levels, it is well-known that incomplete accounting for these effects can lead to lifetime overestimates. Subsequently, cascade-free laser-excited measurements in Ag II were performed that confirmed the agreement between theory and experiment the result for the Pd sequence. To complete this investigation we performed a cascade-free laser induced fluorescence (LIF) excited lifetime study of Au II.

In this study Au ions were created in a plasma produced by irradiation of a gold target with a pulsed laser. Pulses from a Nd:YAG laser were compressed to 1 ns by Brillouin scattering in a water cell and used to pump a tunable dye laser, which was converted to wavelengths around 2100 Å by frequency tripling in nonlinear crystals. Whereas the LIF method is selective in its excitation, it does not have the broad applicability of the BFS method. However, we were able to access three of the eleven BFS measurements by LIF methods, and all three were confirmed (two agreed within two significant digits, and a third agreed to within error estimates, with the BFS result falling on the *short* lifetime side).

Having confirmed the measurements, new theoretical calculations were made using fully relativistic MCDHF methods. These also confirmed the measurements. Thus we conclude that, while computational methods that include perturbative relativistic corrections appear to be adequate for moderately heavy systems such as Ag, fully relativistic calculations are essential for describing systems as heavy as Au. This is consistent with the observation by Desclaux and Pyykkö that "the chemical difference between silver and gold may be a relativistic effect." We have also seen this in semiclassical self consistent field study, in which classical orbits undergo a collapse near Z=60 when a relativistic model was used , but not when the same problem was treated nonrelativistically.

Line strength factors for the Cd and Hg isoelectronic sequences

Beginning with the resonance transitions of the alkali-metallike isoelectronic sequences, we have developed methods whereby line strength factors can be presented in a linear representation that extrapolates to a high Z limit that corresponds to the hydrogenic limit. Thus, a critically evaluated base of accurate measurements can be used to make accurate interpolative and extrapolative predictions for any ion in the isoelectronic sequence to an accuracy that equals or exceeds that of any specific measurement in the data base. These methods have been extended to apply to alkaline-earthlike and inert-gaslike isoelectron sequences through a method that combines lifetime and spectroscopic energy level measurements to account for the effects of configuration interaction. In this approach, the line strength factors for the singlet-singlet resonance transitions and for the singlet-triplet intercombination transitions are placed in a common parametrization by to empirical determination of an effective singlet-triplet mixing angle deduced form of spectroscopic energy level data. While the singlet-triplet mixing angle is defined in the context of a single configuration model, it can also effectively include the effects of configuration interaction, higher magnetic effects such as spin-other-orbit interactions, and differences between the singlet and triplet wave functions.

These methods have been shown to work very effectively for the Be, Mg, and Zn alkaline-earthlike sequences, and for the Ne inert-gaslike sequence. In contrast to ab initio calculations, the semiempirical parametrizations seem to improve in reliability as the number of electrons in the core increased, perhaps as a result of an averaging process. However, as the complexity of the system increases, the question arises of whether the models used at low Z will continue to be applicable, or if a new domain will be entered (like the Z=60 demarcation describe above). For this reason we made extensive comparative studies, including experimental measurements, ab initio theoretical calculations, and semiempirical modeling for the Cd (46 electron) and Hg (80 electron) isoelectronic sequences.

For the Cd sequence the system is sufficiently complex to make it theoretically challenging, yet its midrange origin permits it to be followed through many stages of ionization before reaching the limits of nuclear stability. In contrast, the Hg sequence is substantially more complex, but only the first four members are radioactively stable. Since it is more difficult to measure the atomic structure properties of the radioactive members, (which are nonetheless significant in modeling calculations of radiation transfer in fusion plasmas), extrapolations are valuable.

For the Cd sequence, our calculations showed plunging levels from the unfilled 4f subshell perturb the 5s5p levels above Z=60 and eventually replace $5s^2$ as the ground state. In the Hg sequence, they indicated that the 6s6p and $6s^2$ levels remain below the plunging levels from the 5f and 5g subshells for all ions through uranium. Thus these resonance and intercombination transitions can be expected to dominate the radiative transfer among any of the ions in the Hg sequence whenever they exist in a hot plasma environment. The calculations also confirmed many of the trends that had been observed for the early members of the sequence. They showed that effective singlet-triplet mixing angle θ could be linearized by an exposition of $\cot(2\theta)$ vs $1/(Z-C)$, and that the reduced line strength factors for both the resonance and the intercombination lines could be linearized by an exposition of Z^2S_r vs $1/(Z-C)$. One significant difference was noted between the two sequences. In the case of the Hg sequence, a much better linearization of the intercombination line for of the Hg sequence was obtained if an additional correction to the mixing angle that includes the relativistic j dependence of the radial wave function is included.

Branching Fractions

The ns-np transitions in alkali-metal-like and ns^2-$nsnp$ in alkaline-earth-like systems are now well characterized for the entire isoelectronic sequence. However, these methods can presently be applied only to the lowest lying Δn=0 resonance and intercombination transitions since there the decays are unbranched, allowing line strengths to be deduced directly from lifetime measurements. For Δn>0 transitions, significant branching usually occurs, and the reduction of lifetime measurements to line strength factors requires values for the branching fractions, and such data are virtually non-existent for multiply charged ions. Intensity calibration of detection using continuum radiation standard lamps poses many problems for ion beam excitation, and line radiation intensity calibration standards in the UV are presently lacking. We have thus undertaken a task of developing intensity standards of line radiation in the UV to use in calibrating detection equipment.

We have shown that transitions of the form ns^2np^2-$n^2npn's$ in the Si, Ge, and Sn sequences are virtually free of configuration interaction, hence their branching fractions can be accurately predicted from intermediate coupling amplitudes deduced from measured spectroscopic energy level data. The high level of accuracy in the branching fraction predictions obtain in this manner has been demonstrated by comparision with precision measurements that are available for the visible light in the neutral atoms Si I, Ge I, and Sn I. We have extended these predictions into the VUV in studies of P II – Ar V in the Si sequence, of As II – Br IV in the Ge sequence, and of Sb II – Cs VI in the Sn sequence. This provides a set of calibration standards range from below 400 Å to above 1700 Å. These can be used to calibrate detection apparatus in a beam foil measurement.

We have begun a trial beam foil measurement using a Te III beam to calibrate the apparatus for a measurement in S II. The lines desired in S II are in three groups, in the vicinity of 910, 1055, and 1170 A. There are lines of known branching ratios at similar wavelengths in Te III. By choosing an energy for a Te beam that matches the velocity of the S II beam, corrections due to Doppler shifts and broadening are minimized, and should thus provide a reliable calibration. If this approach is successful, it should be widely applicable.

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Measuring a Nuclear Magnetic Octupole Moment

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We are investigating hyperfine energy level splittings with the hope of improving our knowledge of nuclear structure and the behavior of the electronic wave function near the nucleus. Our long-term goal is to study hyperfine interactions in both neutral and charged atomic systems. Presently, we are measuring the hyperfine splittings in neutral cesium. These measurements provide a means for testing nuclear theory, because hyperfine energy splittings arises through deviations of the nucleus from an ideal spherically symmetric point charge. The effects of the deviations can be described by the interaction between electric and magnetic multi-pole moments of the nucleus with those of the orbital electrons. These interaction energies are small and can be treated as perturbations to the fine structure of atoms. The ^{133}Cs 6p $^2P_{3/2}$ state maybe one of the few systems where one can measure the hyperfine energy splittings well enough to determine the nuclear magnetic octupole moment along with the magnetic dipole and electric quadrupole terms. In addition, the cesium atom plays an important role in the study of weak interactions, and our results could have on impact on the interpretation of atomic parity non-conservation measurements.

The hyperfine interaction is small enough to be treated as a perturbation to the fine structure of an atom and can be expressed in the usual form:

$$H_{hfs} = A(\mathbf{I}\bullet\mathbf{J}) + b\frac{3(\mathbf{I}\bullet\mathbf{J})^2 + \frac{3}{2}(\mathbf{I}\bullet\mathbf{J}) - (\mathbf{I}\bullet\mathbf{I})(\mathbf{J}\bullet\mathbf{J})}{2I(2I-1)J(2J-1)} + c\frac{10(\mathbf{I}\bullet\mathbf{J})^3 + 20(\mathbf{I}\bullet\mathbf{J})^2 + 2(\mathbf{I}\bullet\mathbf{J})[-3(\mathbf{I}\bullet\mathbf{I})(\mathbf{J}\bullet\mathbf{J}) + (\mathbf{I}\bullet\mathbf{I}) + (\mathbf{J}\bullet\mathbf{J}) + 3] - 5(\mathbf{I}\bullet\mathbf{I})(\mathbf{J}\bullet\mathbf{J})}{I(I-1)(2I-1)J(J-1)(2J-1)} \quad . (1)$$

The first term arises from the interaction of the nuclear magnetic dipole moment with the magnetic diopole moment of the electron wave function. The second term describes the electric quadrupole interaction, and the third describes the magnetic octupole interaction. The series can be continued to include higher order multi poles, but the strength of each successive term decreases rapidly. In fact, the third term in (1) is often ignored, but with the precision that can be achieved with contemporary laser spectroscopy, one can determine the magnetic octupole coefficient c through measurements of hyperfine energy splittings. The octupole coefficient can be shown to depend on electron radial matrix elements proportional to r^{-4} and r^{-5}. The r^{-5} terms are largest in systems where relativistic effects are important such as a heavy atom like Cs. The r^{-4} terms are

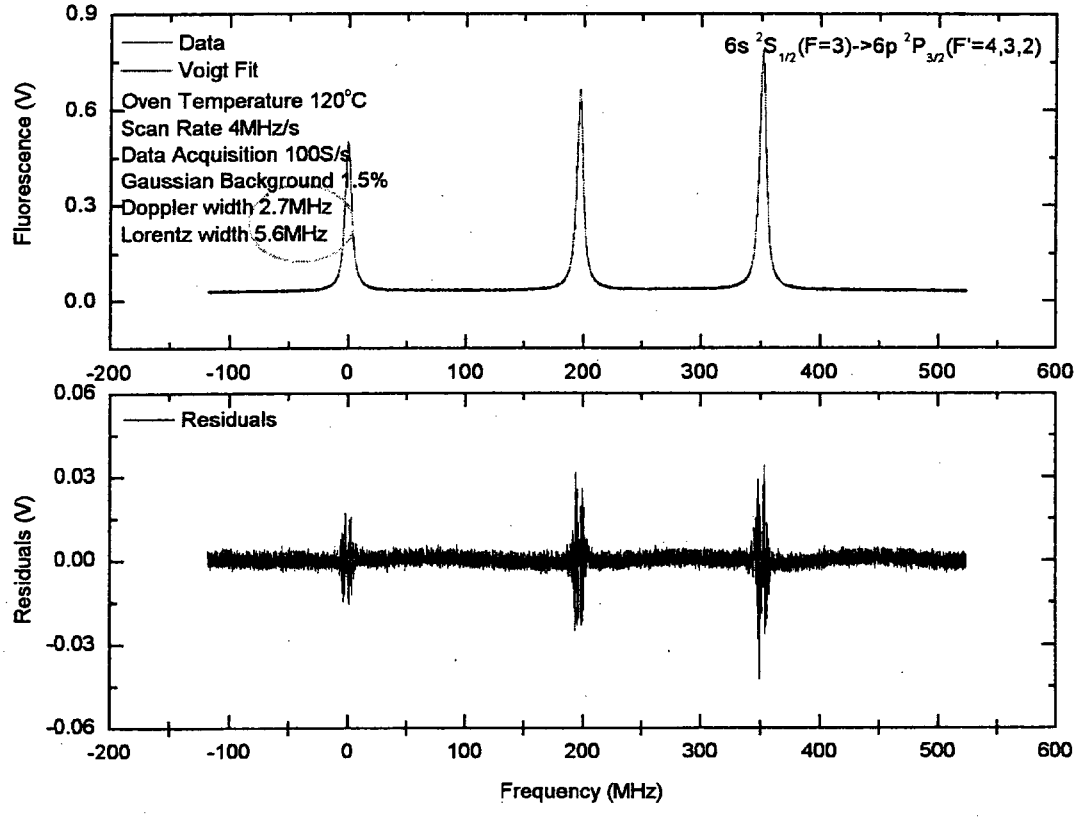
coincidentally largest for $p_{3/2}$ electrons in the non-relativistic limit. For the ^{133}Cs 6p $^2P_{3/2}$ state, we expect to reach the resolution required to determine the rarely observed magnetic octupole coefficient.

The previous best measurement of the Cs 6p $^2P_{3/2}$ state hyperfine splitting with a frequency resolution-to-linewidth ratio of 1 part in 10^3 did not have the resolution necessary to determine c. We are in the process of improving upon this measurement. The general scheme includes producing a dense well-collimated thermal atomic beam with perpendicular laser excitation. Two highly stabilized laser diodes (850 nm) are used. One is designed to scan the hyperfine structure of the 6s $^2S_{1/2}F=3,4 \rightarrow$ 6p $^2P_{3/2}F'=2,3,4,5$ transitions. The second is a reference laser that is absolutely locked near a single hyperfine transition. Portions of the output from each laser are combined on a fast photodiode where the frequency of the heterodyne beat note provides a high resolution absolute calibration. A technique similar to this was used to measure Stark shifts in Li and Cs with a frequency resolution-to-linewidth ratio of 6 parts in 10^5. Thus, the heterodyning technique should lead to an improvement by nearly a factor of 17 over the previous measurements in the 6p $^2P_{3/2}$ state making it possible to determine the magnetic octupole moment of the cesium nucleus.

This project includes a number of technological developments. A Thermal atomic beam is generated with a two-stage oven. The first stage contains Cs metal which his heated to approximately 120 C. This portion can be sealed with a metal seal valve from the rest of the oven and vacuum chamber. The second stage of the oven is a nozzle that is kept at approximately 150 C. The exit port of the nozzle is filled with closely packed small diameter stainless steal tubes. The array of thermal beams then passes through an additional collimator constructed of stacked microscope cover slips. The vertical and horizontal tilt of the collimator is adjustable to minimize asymmetries in the velocity distribution. The resulting atomic beam has a full angular divergence of less than 1 mrad and an experimentally determined Doppler width of 2.6 MHz which includes atomic velocity and laser beam divergence effects. Two highly stabilized diode lasers are required for this measurement. The first is a scanning laser stabilized to a linewidth of approximately 20 kHz using passive optical feed back from a resonant confocal optical cavity. The length of the cavity is approximately 10 cm and is. microscopically adjusted with a piezo-electric transducer (PZT) to tune the laser frequency. Both the voltage to the PZT and the current to the laser are computer controlled in order to scan the laser frequency over a tuning range of approximately 1 GHz. The second laser is used as an optical frequency reference. The reference laser is stabilized using optical feedback in conjunction with the Faraday effect. A portion of the laser output passes through a cell containing Cs vapor in a 1 Gauss magnetic field. On each side of the cell, the laser beam passes through a polarizer. The transmission axes of the polarizers are orthogonal. The

Faraday effect in the Cs vapor allows only a small amount of light to pass through the polarizer-cell-polarizer combination when the laser is tuned near an atomic resonance. The transmitted light is retro-reflected back through the optical system into the laser to provide optical feedback that results in passive locking of the laser frequency in the vicinity of the atomic resonance. The position of the retro-reflecting mirror is also controlled using with electronic feedback. By combining the outputs of the reference laser and the scanning laser on a fast photodiode, the beat note at the difference frequency provides an absolute frequency calibration for the scanning laser. This frequency is read with a precise RF frequency counter. As the first laser scans, a Labview based computer acquisition system records the fluorescence from the atomic beam and reads the frequency difference between the lasers from the counter.

Typical spectra starting from the F=3 ground state of Cs is shown in following figure. The frequency scan consists of ~50000 data points. The analysis is done using a customized Fortran



data-fitting program based on Levenberg-Marquardt method. The data is fitted with three Voigt profiles (a convolution between Gaussian and Lorentzian line shapes) and nonlinear backgrounds that take into account the velocity distribution of the residual Cs cloud in the chamber. For this data, the frequency differences between pairs of peaks can be determined to a precision of

approximately 2 kHz. We are presently evaluating potential sources of systematic error and implementing various improvements to minimize their effects.

Our future plans for this project include a number of improvements that should allow us to achieve a precision of approximately 2 kHz in the energy splittings. A critical element of this experiment is the heterodyne frequency-calibration technique, and several changes are intended to improve upon our preliminary observations. Improvements to the data acquisition system are needed to optimize the calibration. Also the reference laser stability will be improved in three ways. First, we will replace the retro-reflecting mirror to make the external resonant cavity more stable. Second, we will surround the laser and optical feedback system with a heavy airtight box to eliminate acoustic noise. Third, the retro-reflecting mirror will be controlled with electronic feedback derived from a separate Cs reference cell using RF sidebands to generate the error signal. In addition to eliminate the possibility of line shifts due to the Zehman effect, three pairs of magnetic field coils in the Helmholtz configuration will be placed around the vacuum system to zero the magnetic field in the interaction region.

In conclusion, the technical developments described have positioned us for realizing our goal of determining the nuclear magnetic octupole moment of cesium. We have taken preliminary data that demonstrate the capability or our apparatus to produce spectra with a high signal-to-noise ratio. Analysis of the data shows that we have the capability to measure spectral differences with a precision of approximately 2 kHz. We are presently installing improvements that will eliminate potential sources of systematic error. We expect to complete the cesium measurements in the coming months. In the future, we hope to apply the experience we have gained to other neutral and charged atomic systems.

Recent Publications:

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Research Summaries

(multi-PI programs alphabetical by institution)

X-ray Interactions with Atoms and Molecules

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We seek to establish a fundamental and quantitative understanding of x-ray interactions with free atoms and molecules. We have explored the broad energy range where the dominant interaction evolves from photoabsorption to scattering, with careful attention to regions near resonances and thresholds. The focus has been on understanding the limitations of theory, in particular the validity of the independent particle approximation and the role of multipole effects. Multipole effects are studied using a recently-developed angle-resolved photoelectron spectrometer system. The complex decay pathways following inner-shell excitation are being investigated using x-ray-ion coincidence techniques. Multielectron excitation is studied as a measure of electron-electron correlation in heavy atoms. We continue to address experimental and theoretical discrepancies (up to 10%) in atomic form factors by making high-precision absolute measurements.

I. Nondipole Effects on Photoelectron Angular Distributions

R. W. Dunford, E. P. Kanter, B. Krässig, S. H. Southworth, and L. Young

Atomic photoionization processes have been extensively explored by comparing theory with measurements of partial photoionization cross sections σ and photoelectron anisotropies β as functions of photon energy. Even in the independent-particle approximation, variations of σ and β with photon energy occur due to maxima and minima in dipole matrix elements and variations of phases of transition amplitudes. Many-electron interactions are also strongly manifested in σ and β in two general ways. The interaction of discrete states with continua produces rapid oscillations of σ and β over narrow energy ranges near ionization thresholds, and continuum-continuum coupling can modify σ and β over broad energy ranges. Relativistic interactions must also be treated in certain cases. New aspects of the photoionization of atoms, molecules, and materials have been investigated in recent years with studies of *nondipole photoelectron asymmetries*. Interference between electric-dipole (E1) transition amplitudes and amplitudes for higher-order interactions such as electric quadrupole (E2) produce asymmetries between the intensities of photoelectrons emitted forward and backward with respect to the photon propagation vector **k** (pure E1 angular distributions are forward-backward symmetric). Similar to σ and β, forward-backward asymmetries vary with energy due to one-electron and many-electron interactions and provide additional insight into photoionization processes.

We have developed a photoelectron spectrometer system using four parallel-plate analyzers to accurately and efficiently measure forward-backward asymmetries of atoms and molecules. Using hard x-rays at Argonne's Advanced Photon Source, we measured forward-backward asymmetries of Kr 1s photoelectrons from threshold to 8 keV kinetic energy and found excellent agreement with theory. We also searched for extra-atomic effects in the asymmetries of Br 1s photoelectrons from molecular Br_2 and $CBrF_3$. Recent theoretical studies predict surprisingly strong forward-backward asymmetries in low-energy ($\approx$10-200 eV) photoionization of valence electrons due to quadrupole autoionizing and shape resonances and variations of dipole amplitudes. These effects will be studied in experiments on a VUV beamline at the Synchrotron Radiation Center in Stoughton, Wisconsin.

II. X-Ray-Ion Coincidence Spectroscopy of Atoms and Molecules

G. B. Armen[a], and J. C. Levin[a],
E. P. Kanter, B. Krässig, S. H. Southworth, and L. Young

Atomic x-ray absorption typically excites or ejects a deep inner-shell electron, producing a vacancy state which relaxes by a series of radiative (x-ray fluorescence) and radiationless (Auger-electron emission) processes. A range of final ion charge states is observed due to alternative decay pathways. If the initial vacancy is produced in an atomic constituent of a molecule, a similar vacancy cascade occurs, but removal of delocalized valence electrons produces positive charge on two or more atomic centers and results in ion fragmentation (Coulomb explosion). Coincidence techniques, in which two or more ejected photons, ions, or electrons are detected simultaneously, allow specific decay pathways to be selected from a complex vacancy-cascade process. K-shell vacancies in high-Z atoms decay preferentially by K-L x-ray fluorescence in which the vacancy is transferred to the L-shell. By using this fluorescent x-ray as an event marker in a coincidence circuit, the ion charge-state distributions of Kr and Xe were measured as the absorbed x-ray energy was tuned across their respective K edges at 14.3 and 34.6 keV. The measured yields are being compared with a model which treats excitation of the 1s electron to bound and continuum states at the K edge. We also studied ion fragmentation of $CBrF_3$ following x-ray absorption at the Br K edge (13.5 keV). Information on fragment ion correlations was obtained by detecting two ions in coincidence with the same fluorescent x-ray. In our latest experiments, the decay pathways were further distinguished by using a Si(Li) detector to resolve diagram lines in the x-ray fluorescence spectrum.

III. Double K photoionization of heavy atoms

R. W. Dunford, D.S. Gemmell, E. P. Kanter, B. Krässig, S. H. Southworth, L. Young

We have previously demonstrated a relatively simple method to isolate the effects of electron-electron correlations in heavy atoms by measurement of simultaneous x-ray photoionization of both K-electrons in molybdenum. The presence of these double-hole states is signaled by the observation of $K_{\alpha,\beta}$ hypersatellite fluorescence detected in coincidence with the subsequent satellite transition filling the second vacancy. Recently, we extended these measurements to Ag (Z=47). Because of extensive previous studies of this system produced by the electron capture (EC) decay of ^{109}Cd, the shakeoff contribution is well-known experimentally for the single-electron final state produced in EC. Thus, our photoionization measurements will isolate the effects of the dynamic electron-electron scattering (TS1) term. Measurements were carried out at several energies from the double K-ionization threshold to the region of the expected maximum (from 50-90 keV) in the cross-section and analysis is currently underway. Preliminary analysis has confirmed the predicted rise in the double-ionization cross section throughout this region. These data are more complex to analyze however because of the use of Ge detectors, which were needed for these higher energies. Because of the higher efficiency of these detectors for high energy photons, and their larger angular acceptance, there was considerable background from the long tail of Compton scattering off of K-shell electrons which limits the accuracy of such measurements by the coincidence method. Analysis of these data is ongoing, and we anticipate the development of improved multidimensional fitting algorithms will reduce the contribution of the background to the experimental errors. In an effort to experimentally overcome this background problem, we have explored the use of a crystal spectrometer to isolate the hypersatellites in a singles measurement, as was recently demonstrated by Diamant *et al* in lighter atoms. Measurements were carried out on the BESSRC wiggler beamline with 125 keV photons and a thick Ag target. These measurements demonstrated the difficulty of separating the hypersatellites from the Lorentzian tail of the diagram line in heavy atoms. The natural linewidth grows with $\approx Z^4$ while the double K-ionization probability falls off with $\approx 1/Z^2$. Because the hypersatellite-diagram splitting only grows with $\approx Z^3$, the hypersatellite/tail intensity falls off as $1/Z^3$ and thus becomes the limiting background for such singles measurements, regardless of improvements in resolution or angular

acceptance. This strongly suggests that the coincidence method will be preferred in exploring heavier systems.

IV. High precision measurement of imaginary component of atomic form factors

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S.H. Southworth, L. Young, E.P. Kanter

Complex atomic form factors describe the primary interaction between x-rays and atoms; the real part of the form factor specifies the scattering processes and the imaginary component is directly related to the photoabsorption cross-section. These form factors are in widespread use for crystallography, medical diagnostics and radiological safety. However, there are discrepancies between theoretical tabulations and experiment of order of 10% in both the real and imaginary components. All general tabulations assume an isolated atom and invoke the independent particle approximation. Our earlier work focused on the real part of the form factor, elastic and inelastic scattering, in rare gases where comparisons with theory are "clean". There we were able to outline a method to correct existing IPA tabulations perturbatively for the inclusion of nonlocal exchange, electron correlation and **p•A** terms to an accuracy of 1%. The new experiment addresses the imaginary component of the form factor by precision measurement of attenuation on extremely well-characterized solid samples. The range of energies available at the APS allows extended coverage past the K-edges of many materials. In this experiment, performed on the bending magnet beamline at BESSRC, we measured attenuation in molybdenum and tin over the range of 30 keV to 75 keV. This range includes the K-edge of tin where data were taken in 1 eV intervals. A second monochromator was used to provide complete isolation from harmonic contamination in the incident beam. Systematic contributions from scattering were monitored using a CZT detector and a segmented Ge(i) detector. The preliminary analysis is consistent with a final precision of 0.1%.

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Structure and Collisions with Few-Electron Ions

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This program is focussed on topics of current interest in both atomic structure and ion-atom collisions involving few-electron ions. This work is connected to the study of astrophysical and laboratory plasmas. It also supports the field of many-body atomic theory which continues to be a challenging problem with connections to such fields as nuclear physics and atomic parity nonconservation. An important feature of the program is the emphasis on comparing the properties of ions at intermediate nuclear charge Z (studied at Argonne's heavy ion linear accelerator, ATLAS) with the properties of very high-Z ions (studied using the heavy ion synchrotron SIS at GSI in Germany).

I. Relativistic Quantum Mechanics and Fundamental Interactions

R. W. Dunford, E. P. Kanter, Th. Stöhlker[b], H. G. Berry[c], S. Cheng[a], L. J. Curtis[a], A. E. Livingston[c], and P. H. Mokler[b]

Our work on the physics of highly charged ions has a particular emphasis on studying atomic structure issues. The ATLAS facility can produce few-electron ions in the intermediate Z regime (we mean roughly $14 < Z < 40$). He-like ions in this regime are particularly suited for elucidating the interplay of relativistic effects and electron-electron interactions which is an area of current theoretical interest. At GSI, few-electron ions up to H-like Uranium are available. Here, the relativistic effects dominate over electron-electron interactions. Also at high Z, Quantum Electrodynamics (QED) makes large contributions to the energy levels. Although QED has been well tested, most of the data are at low fields and the theory has not been as well tested at high field strengths such as those acting on the electrons in the K-shell of uranium.

Lamb shift in U^{91+}. One of the frontiers in testing quantum electrodynamics (QED) is the study of electrons in intense electromagnetic fields. The important experiment is the measurement of the 1s Lamb shift in the most highly-charged, one-electron ion available in the laboratory U^{91+}. This is a relativistic system containing high momentum components and the electric field strength begins to approach the critical value for the spontaneous emission of electron-positron pairs. All of this makes it ideal for testing the limits of our understanding of bound state QED. We recently reported the first Lamb shift measurement in H-like Uranium to utilize the deceleration capability of the ESR storage ring [Stöhlker, et al, Phys. Rev. Lett. **85**, 3109 (2000)]. Deceleration leads to a reduction in the uncertainty due to the motion of the ions (i.e. the Doppler shift) and these data marked a significant advance in this field. The future goal of this work is to perform experiments which combine the use of the deceleration capability of the ESR storage ring with high-resolution devices such as crystal spectrometers, Doppler tuned spectrometers or bolometers. The needed experimental techniques are being developed in experiments done at the gas jet of the ESR storage ring.

E1-M1 Damping interference in Ar^{17+}. The metastable 2s $^2S_{1/2}$ state in one-electron ions decays to the ground state either by two-photon decay or by single-photon M1 decay. If an external electric field **E** is applied to the ion there is, in addition, a "Stark-induced" E1 amplitude. Interference between the E1 and M1 amplitudes leads to an asymmetry in the angular distribution of the photons (**k**) proportional to the invariant **k•E** which is of interest because it appears to violate time reversal (T) invariance. It has been shown however, that even in a T-conserving theory this term can be nonvanishing if damping is taken into account. We observed this "E1-M1 damping-interference" effect at ATLAS using metastable Ar^{17+} and have recently remeasured the asymmetry with an improved experimental technique. In the experiment, a large electric field is produced by motion of the ions through an applied magnetic field. We measure the angular distribution of the decay radiation with an array of Si(Li) x-ray detectors. In addition to the **k•E** asymmetry we also measure other angular correlations between the emitted photons and the applied electric and magnetic fields. These measurements can be used to determine a number of properties of the ions including the Lamb shift, the decay width of the 2p level and the amplitude for the M1 decay mode. In addition the angular distribution can be used to measure the polarization of the metastable ions.

Spectral shapes in two photon decay. A measurement of the exact shape of the continuum spectrum from two-photon decay of H-like or He-like ions tests our understanding of the entire structure of these ions since the theory requires a sum over a complete set of intermediate states and both energy levels and wavefunctions must be understood. The two-photon continuum is important in astrophysics because, for example, it contributes to the continuum radiation in the spectrum of planetary nebulae and it is important to understand this contribution to gain an understanding of these structures. Fully relativistic calculations for two-photon decay in He-like ions have been performed by Derevianko and Johnson. Their results show a marked dependence of the continuum shape on nuclear charge. We have studied this change by comparing the shapes of two-photon decay at intermediate nuclear charge Z in He-like Ni (Z=28) and at high-Z in He-like Au. The Ni experiments have been carried out at the ATLAS accelerator and the high-Z experiments have been done using the SIS facility at GSI in Germany. In the ATLAS experiment, two-photon decays were measured in both H-like and He-like nickel during the same run, switching between the two ions several times during the experiment. Since the continuum shape in the two-photon decay of the 2 $^2S_{1/2}$ level in H-like Ni is known precisely, this served as an on-line calibration of the spectral efficiency of the detection system used and this resulted in very precise and reliable data which agrees well with the theory. These also show good agreement with the theoretical calculation and a clear departure from the shapes at both high and low Z.

In future experiments we plan to measure two photon decay of the 2 $^3P_0$ level in He-like Uranium. This state decays 30% of the time via E1M1 two-photon decay. This decay mode has not been studied to date and has potential application to the measurement of atomic parity nonconservation. We also plan to study two photon decay of inner-shell vacancies in neutral atoms using Argonne's Advanced Photon Source.

II. K-shell Excitation of He-like Ions

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Coulomb excitation is an important mechanism leading to the production of characteristic projectile photons in collisions of fast charged particles with atomic targets. This has been a neglected area of the field of atomic collision physics. In particular, little information is available for state-specific excitation in the intermediate energy regime where the orbital velocity of the projectile electron is similar to the velocity of the ion. We are studying projectile excitation at intermediate energies at ATLAS using both foil and gas targets.

Initial measurements of the cross sections for excitation from the ground state of He-like Ni ions to the n=2 excited states have recently been completed. Identification of final states produced in the collisions was done by a combination of low-resolution x-ray spectroscopy using Si(Li) detectors and measurements of the lifetimes of the excited states. The results show that direct excitation to the triplet He-like states is important in excitation by both gas and foil targets. This is significant since excitation to this level from the $1s2s\ ^1S_0$ ground state requires a spin flip and is forbidden in the lowest order theories currently available.

In recent work we studied excitation in both ^{61}Ni and ^{58}Ni in order to resolve an ambiguity between the $^3P_0$ and the $^3S_1$ states which have indistinguishable lifetimes in ^{58}Ni. The isotope ^{61}Ni has a nuclear spin and the lifetime of the He-like $1s2p\ ^3P_0$ state in this system is shortened by the hyperfine interaction. By comparing data taken with the two isotopes, information about Coulomb excitation may be obtained for both the $1s2p\ ^3P_0$ and the $1s2s\ ^3S_1$ states.

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Ultracold Atoms: Applications

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The unique properties of cooled and trapped neutral atoms enable breakthrough applications in a variety of fields. Extreme isotopic selectivity combined with efficient capture probability has spurred the development of an ultrasensitive trace isotope analysis technique based upon counting single rare atoms, ATTA (atom trap trace analysis). The highly-localized, low-momentum spread sample of atoms available in a trap has also long been touted as a target for ionization experiments using COLTRIMS (cold target recoil ion momentum spectroscopy). With the development at Argonne of expertise in laser-manipulation of atoms, we are currently exploring the use of cooled and trapped atoms in these two areas.

Electron impact ionization of Li: absolute cross-sections and charge state distributions

M.T. Huang, L. Zhang, S. Hasegawa[1], S.H. Southworth, L. Young

Trapped neutral atoms are ideal targets to elucicate complex ionization mechanisms by using kinematically complete experiments involving recoil ions and electrons in coincidence. Li holds particular interest in that the three-electron Coulomb continuum is the next step (after the much-studied two-electron case) for understanding multielectron dynamical correlation in photoionization. In the area of strong-field photoionization Li holds special interest in that it is the atom with the greatest difference between the first and second ionization potentials, and thus, the largest intensity range over which the "non-sequential" ionization mechanism should be operative. Being a few-particle system, theoreticians can readily tackle issues involving the multi-electron continuum in the presence of a nuclear Coulomb field and strong-laser field.

Currently, we are using a Li magneto-optic trap (MOT) to measure low energy electron impact ionization cross sections where 80% differences between recent theories by Bray and Chang are not resolved by earlier measurements which differ by a factor of two. We employ the "trap loss" method, initially developed by Gould and coworkers for photoionization and extended by Schappe, Anderson, Walker and Lin for electron impact ionization cross-sections. In this method, the decay lifetime of the trap is measured with and without the presence of the ionizing beam. The additional decay rate induced by the beam is ascribed to ionization processes. The trap loss method provides an alternative to traditional measurements of absolute cross sections where beam intensities, sample densities and collection detection efficiencies need to be determined on an *absolute* scale. In contrast, the trap loss method requires only the electron current density to be absolutely detemined. However, we find several intricate problems associated with the electron-impact measurements. The most serious is that each electron pulse induces a pressure rise, which in turn increases the loss rate from the trap. The second is that additional electron scattering processes, such as excitation, can cause atoms to be ejected from the trap if the momentum transfer is sufficiently large. We are investigating various solutions to these issues and a final accuracy of 10% appears feasible. We have also tested a pulsed, ion-imaging time-of flight detector in conjunction with the MOT to measure the ratio of single, double and triple ionization of Li. These electron impact results will be used to compare with

analogous studies with photons and theoretical (semi-empirical) predictions. In the future, we plan to use the Li MOT target and recoil ion imaging technique to investigate photoionization in high-fields and at short wavelengths.

Optical production of metastable rare gases

L. Young, D. Yang[2], R. W. Dunford

Metastable rare gas atoms have numerous applications in fields such as atomic collisions studies, atom traps and rare isotope detection. They can also provide a means of obtaining hyperpolarized rare gases which are applicable in such fields as medical imaging, tests of fundamental symmetries, and materials research.

Metastable rare gas atoms are generally produced in beam form by bombarding a ground state beam with electrons or by direct extraction from a discharge. Such methods produce beams with a metastable fraction of less than 0.1%. For many of the applications it is desirable to have a significantly increased metastable fraction. For the experiments testing fundamental symmetries, achieving the required precision dictates that one work with the largest possible atomic samples. In the Atom Trap Trace Analysis (ATTA) technique rare isotopes of krypton in the metastable state are counted after capturing them in a trap. The small number of atoms available in this case requires good efficiency in excitation to the metastable state.

We have developed an optical method for excitation of metastable Kr atoms (5s J=2 level) which has potential for significantly increasing the metastable fraction available for the applications. In this method we employ a three-step scheme for excitation of the metastable state. In the first step, an ultraviolet lamp provides the 124-nm radiation needed to drive a transition from the atomic ground state to the 5s J=1 level in Kr. The excited atoms are then pumped to a 5p J=2 level using 819-nm light from a Ti-Sapphire laser. In the final step the 5p level decays to the metastable 5s (J=2) level with a branching ratio of 77%. This last step is accompanied by the emission of a 760-nm photon which can be used to monitor the production of metastables. We have demonstrated this method in a Kr gas cell, and plan to extend the basic technique to production of a beam of metastable atoms.

Atom Trap Trace Analysis

Z.-T. Lu[3], K. Bailey[3], C.-Y. Chen[3], X. Du[3], Y.-M. Li[3], T. P. O'Connor[3], L. Young

Ultrasensitive isotope trace analysis has been an important tool in modern science, with applications in radioisotope dating, medicine, and studies of transport processes in the ocean, atmosphere and groundwater. Two well-developed methods, low-level counting (LLC) and accelerator mass spectrometry have been widely used at the part-per-trillion level. Recent careful implementations of resonance ionization mass spectrometry approach this level.

We have developed a new method for ultrasensitive trace isotope analysis based upon laser manipulation of neutral atoms, atom trap trace analysis (ATTA). In this method, individual atoms are counted while residing in a magneto-optical trap (MOT), which affords extremely high isotopic selectivity. We have demonstrated a selectivity of 10^{-11} to 10^{-13} for (^{85}Kr/Kr) and (^{81}Kr/Kr) by counting these rare atoms in a natural atmospheric krypton sample. With no observed contamination from neighboring isotopes, the selectivity appears to be limited only by the number of atoms that can be sorted during a finite operation time. Key technical features are

an efficient capture rate and the ability to detect a single atom in the MOT. These two requirements are independently optimized with separate capture and detection phases. We initially obtained an overall efficiency of the system of 10^{-7}, with 10^{-4} arising from the need to produce metastable krypton atoms for the trapping process. A rf discharge source was found to increase the metastable excitation efficiency by a factor of ten over a dc discharge. Methods to improve the overall efficiency, by gas recirculation and optical excitation of the metastable state are presently under investigation.

While many applications are envisioned, two involving ^{85}Kr and ^{81}Kr should be of particular interest to the Department of Energy. ^{85}Kr (half-life 10.5 years) atoms are produced in nuclear fission processes. Trace detection of ^{85}Kr can be used to monitor environmental contamination around nuclear-waste storage areas, to monitor nuclear-fuel reprocessing activities around the world, and to provide advanced warning of reactor leakage problems. ^{81}Kr is a cosmogenic nuclide with a half-life of ≈229,000 years. Due to its gaseous and inert nature, it is homogeneously distributed over the earth. In addition, its concentration is unaltered by human activities with nuclear fission because stable ^{81}Br shields ^{81}Kr from the neutron-rich isotopes that are produced in nuclear fission. Thus, ^{81}Kr is an ideal tracer for dating polar ice and groundwater in the 100,000 year range.

In addition, an apparatus for detection of ^{41}Ca using ATTA has been constructed and tested. The applications for this isotope of 103,000 year half-life and natural abundance of 10^{-15} - 10^{-14} include medical use as a tracer for osteoporosis, nuclear activity monitoring and radioisotope dating of ancient bones. A slightly modified system could be of interest to DOE as an analyzer for ^{90}Sr, a fission product.

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Structure and Dynamics of Atoms, Ions, Molecules, and Surfaces:
Mechanisms for Transfer Ionization in High Z Ions on He

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The scope of the JRML research program is to study experimental and theoretical problems in ionic and photonic collisions with atoms, ions, molecules, laser cooled atoms, laser excited atoms, and surfaces. The experimental program is based on the study of collision physics using ion accelerators and laser systems in the J. R. Macdonald Laboratory, and the ALS synchrotron facility at Berkeley. The goal is to provide fundamental understandings of energy, momentum and charge transfer in interactions where one or more electrons and/or nuclear charge centers of the system are disrupted. This abstract presents recent progress and future plans in one of the experimental research projects. I. Ben-Itzhak, C. L. Cocke, B. D. DePaola and S. Hagmann present abstracts on other subjects in our experimental research program and can be found in these proceedings. Abstracts on subjects in our theoretical research program are presented by C. D. Lin, U. Thumm and B. D. Esry and also can be found in these proceedings.

Recent progress and future plans:

Mechanisms for Transfer Ionization in High Z Ions on He:
(R. Unal, P. Richard, I. Ben-Itzhak, C. L. Cocke, C. D. Lin, and H. C. Tseng) The process of removing two electrons from He by a charged ion has been studied for several years.[1-3] One of the interesting questions is the behavior of the transfer ionization process, TI, in which the projectile captures one of the electrons and the other is emitted to the continuum. Several features existing in the data are not understood to date[4] and to add intrigue to the problem, recent new data on H^+ + He showed a completely unexpected result[5] at collision velocities above 4.0 au. Below 4 au the ratio of TI-to-single electron capture, SC, is flat at a value of ~2.5%. The new results showed the ratio linearly increasing with velocity up to a value of ~4.4% in the range of 4.0 to 7.5 au. It was expected that the ratio would slowly fall with increasing velocity to the shake-off value of ~1.6%. The shake-off value is defined as the probability that a second electron will be lost by the He ion in its final state readjustment following SC. The electron emission is due to the incomplete overlap of the wave function of the intermediate He^+ state with that of the He^+ ground state. We have studied the TI/SC ratio for high velocity highly charged ions on He at velocities in the range of 6 to 10 au and observed, conversely, that the ratio is monotonically decreasing with velocity.[6] If we assume that the two-electron removal from He is occurring by independent interactions with the projectile, the ratio of TI-to-SC is expected to vary as Z^2, where Z is the projectile charge. Using this assumption, we can compare our results for F^{9+} projectiles to that of H^+ on He by using the appropriate scaling.

A comparison of the different data sets is shown in Fig. 1. The low velocity data (V ~ 2 to 4 au) from Shaw and Gilbody *et al.*[1] for H^+ + He are shown as solid squares, the new higher velocity data (V ~ 4.5 to 7 au) from Mergel *et al.*[5] are shown as solid squares and

the F^{9+} + He data from Unal *et al.*[6] are shown as solid circles. The plotted F^{9+} data are $R(scaled) = \frac{R(measured) - R(shake)}{Z^2} + R(shake)$, where R(shake) is taken as the theoretical value. This formula is used since the shake-off limit is independent of projectile Z.

This was the status of the TI/SC ratio until a few weeks ago, when Schmidt *et al.*[7] extended the H^+ + He measurements to higher velocity (V ~ 10 to 14 au) using the storage ring at Stockholm. Their results were reported at ICPEAC 2001 in Santa Fe this summer and are given in Fig. 1 as solid squares. The value of the ratio is observed to decrease from ~4 % to ~3% with velocity in the range of 10 au to 17 au. These latter results compare favorably with the behavior of our F^{9+} data. Earlier F^{9+} + He results from Shinpaugh *et al.*[2] show a turnover at lower energies similar to the H^+ + He data at about 7 au. These data were not taken with the COLTRIMS method used by Mergel *et al.*,[5] Unal *et al.*,[6] and Schmidt *et al.*[7] and suffer from fairly large errors. For this reason they are not used in the comparison, however those data are very suggestive of a favorable comparison between the velocity dependence of H^+ + He to a scaled F^{9+} + He assuming an independent electron model. This suggests that we attempt the difficult measurements of F^{9+} + He at velocities below 6 au using our COLTRIMS apparatus. C. D. Lin and H. C. Tseng have performed coupled channel calculations for the energy dependence of TI and SC for F^{9+} + He and find values slightly higher than our measured values, but with approximately the same energy dependence. We show this calculation scaled with the same formula as used for the experimental data. One of the difficulties of performing accurate calculations is the large basis set needed to properly describe the collision system.

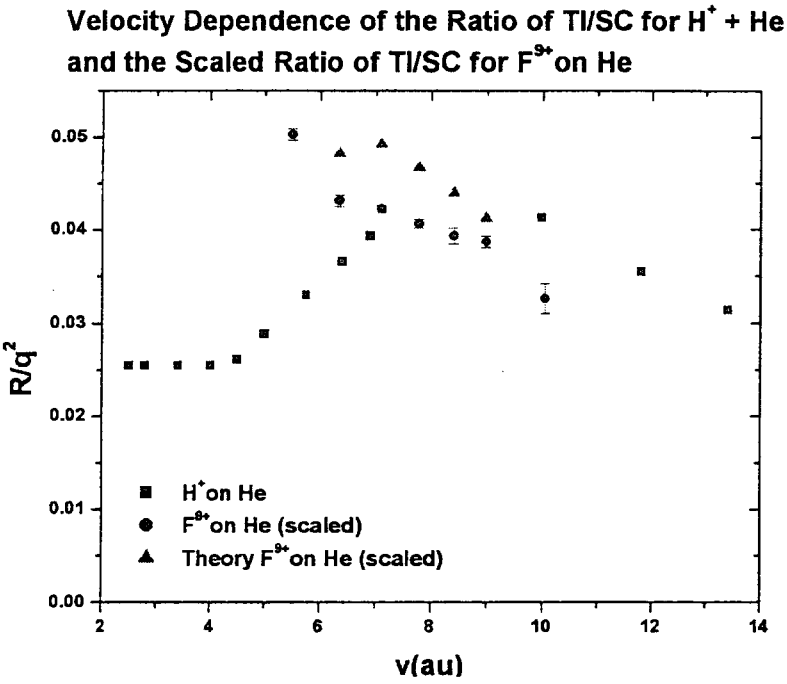

Fig. 1 Transfer Ionization to Single Capture Ratio

The TI process can proceed via two independent e-projectile ion interactions as discussed above [sometimes referred to as kinematical TI or TS2(two-step, two e-nuclear

interactions)] or it can proceed via the Thomas scattering mechanism [sometimes referred to as e-e Thomas TI of TS1(two-step, one e-nuclear interaction)]. The Thomas scattering mechanism transfers little momentum to the recoiling target nucleus, whereas kinematical SC transfers longitudinal momentum to the recoiling target nucleus. The same holds true for the corresponding TI processes. By using COLTRIMS one can separate the two TI processes based on the observed longitudinal momentum transfer to the recoiling target ion. Schmidt *et al.*[7] has done this for H^+ + He and reported that the e-e Thomas TI is 35% of the total TI. The e-e Thomas TI scattering becomes negligible for high Z projectiles due to the Z^2 scaling expected for the kinematical TI. The resulting kinematical TI/SC ratio for H^+ + He then compares very favorably with the scaled TI/SC ratio for F^{9+} + He in Fig. 1.

The other question that we have addressed with the TI/SC ratio is the projectile q dependence for projectiles greater than q=9. The big unanswered question here is the sudden fall in the TI/SC ratio for values of q above q = 20, as reported by Datz *et al.*[8] We have extended our studies[3,4] to new measurements for Si, Ti, and Cu as shown in Fig 2. We see a ratio that follows a q^2 dependence up to approximately q = 9. Above q = 9 the experiment exceeds the q^2 dependence prediction due to antiscreening. The new data go only up to q = 20 and show a smooth monotonically increasing TI/SC ratio. The inverted triangles are data for Ti^{15+} and Ti^{18+} from Montenegro *et al.*[3] The new data points for $Ti^{15,17,18+}$ have smaller errors, and when combined with the $Cu^{18,20+}$ reflect a more gently increasing behavior than might be inferred from the earlier Ti data. Our plan to complete this phase of the experiment is to push the charge state measurements up to q = 30 in order to overlap with the high q data of Datz *et al.*,[8] which showed TI/SC ratios of approximately 2, which is 5 times smaller than our TI/SC ratio for q = 20.

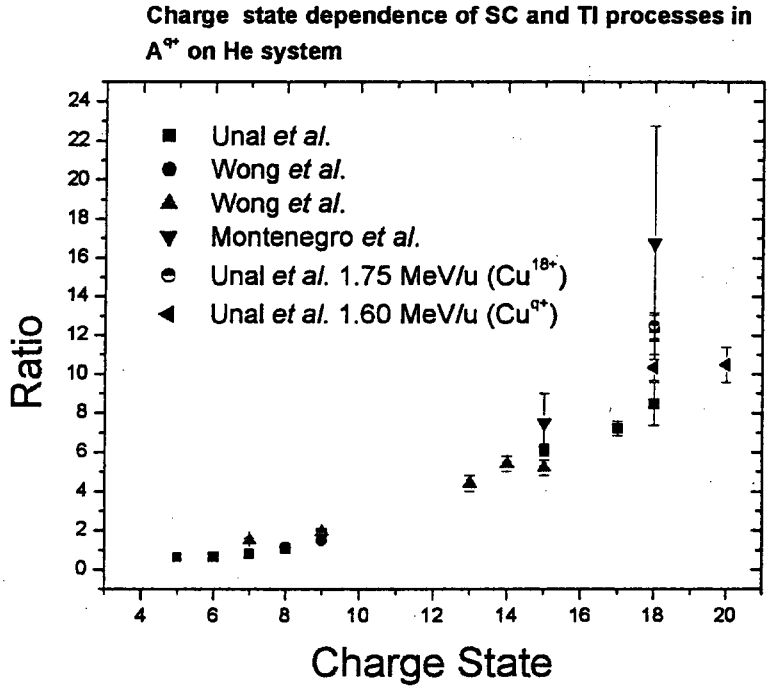

Fig. 2 Charge Dependence of TI to SC Ratio

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Structure and dynamics of atoms, ions, molecules and surfaces: Atomic physics with ion beams and synchrotron radiation

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The goals of this aspect of the JRML program are (1) to explore the dynamics of photoelectron emission from and the structure of small molecules, (2) to identify and explain basic mechanisms whereby electrons are removed from simple systems in capture and ionization processes.

Recent progress and future plans:

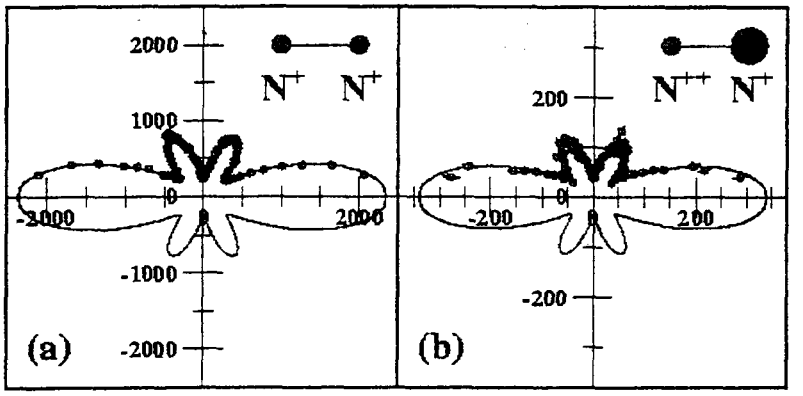
1) Photoelectron diffraction from light molecules fixed in space *A.Landers (Western Mich.Univ.), R.Doerner, Th.Weber, M.Hattass, O.Jagutzki, A.Nauert, A.Staudte, H.Schmidt-Boecking(U.Frankfurt), M.H.Prior(LBL), A.Cassimi(CIRIL), T.Osipov, I.Ali, C.L.Cocke (KSU).* We measure the diffraction patterns of photoelectrons issuing from core photoionization of fixed-in-space light molecules. The electron emanates from a known site within the molecule and "illuminates" the structure from within. The diffraction patterns are shaped by the interaction of the electron with the instantaneous potential presented by the remaining nuclei and electrons of the molecule. The data provide an extremely comprehensive picture of the diffraction patterns, which in turn probe the overall charge density distribution within the molecule at the time of photoelectron emission. The experiments also provide information on the subsequent modes of breakup of the doubly-charged molecule.

The experiments are carried out at the Advanced Light Source (ALS) on beamlines 4.0 and 9.3.2. The K shells of the target molecules (CO, N_2, C_2H_n) are ionized slightly above threshold. The molecule subsequently undergoes Auger decay, and the doubly charged molecule then dissociates. COLTRIMS techniques are used to measure the coincident photoelectron and the two charged molecular fragments. The charged products are projected onto the surfaces of time- and position-sensitive multichannelplate detectors by an electric field, and the full coincident momentum vectors of the three products are reconstructed on an event-by-event basis. The molecule is "fixed in space" on an *a posteriori* basis, assuming a radial two-body dissociation. The angular distributions of the photoelectrons can be plotted with respect to either the polarization vector (in the case of linear polarized light), the propagation vector or the light (in the case of circularly polarized light) or the molecular axis. The technique covers the entire reaction phase space, since all reaction products are measured simultaneously with an efficiency of order unity, and the data sets are very comprehensive and sufficiently large to admit (even invite) their presentation in the form of movies.

A summary of some results for CO and N_2 is:

1)From the photoelectron angular distributions the amplitudes and phases of the partial waves, including the relative phases between sigma and pi amplitudes, can be extracted. The results are in good agreement with theoretical calculations carried out by several groups for CO and N_2.

2)The data show at best very weak non-dipole contributions in the photoelectron spectra.
3)For low kinetic-energy-release channels, the photoelectron spectra wash-out, indicating that the intermediate fragment lifetime is long enough that the orientation of the molecule with respect to the initial axis is lost to rotational motion prior to fragmentation.



An example of the angular distribution of photoelectrons on the f-wave resonance for N_2, with the molecule aligned along the polarization vector, is shown in the above figure. The left-hand figure is for the N^+ - N^+ final channel, while the right-hand one is for N^+ - N^{2+} with the dication to the left. This figure shows that there is no correlation between the asymmetry of the final channel and the photoelectron emission.

A recent application of the technique to the C_2H_n sequence has been undertaken, partially because these molecules are widely used as adsorbates on surfaces and partially because a controversy exists concerning the possible non-existence of an f-wave resonance in these molecules. Full photoelectron spectra have been taken on and off the calculated position of the resonance for C_2H_2, C_2H_4 and C_2H_6. A clear indications of f-wave structure is seen on resonance, and a partial wave analysis is underway. For the case of acetylene (C_2H_2) an unexpected two-body breakup channel of the dication was observed, resulting in the fragments C^+ and CH_2^+. This is attributed to the rearrangement of the molecule from the acetylene to the vinylidene configuration after photoelectron emission but prior to fragmentation. The vinylidene and acetylene configurations are populated with similar probabilities. Preliminary analysis of the data indicates that, while a very marked f-wave structure is seen in the photoelectron spectra in the acetylene channel, this structure is partially lost in the vinylidene channel. This suggests that the lifetime of the vinylidene configuration may be longer than characteristic rotation times. This project is the Ph.D. project of Timur Osipov at KSU. Near-future work on this project will include obtaining complete data on and analysis of the C_2H_n data. More long range plans include attempting to extend these techniques to time-dependent probes of molecular structure.

2) COLTRIMS measurements of electron spectra from low energy ionization of atomic H and He targets *E. Edgu and C. L. Cocke* The goal of this project is to identify and characterize as cleanly as possible the process whereby a slow charged projectile

promotes into the continuum an electron from a neutral target. The projectile velocity is sufficiently low that direct kinematic ionization is forbidden, and saddle-point electron promotion, in some form, is expected to the major process. The electron momentum distributions show clear patterns that are much more definitive in determining the ionization mechanism than are total cross sections. Several theoretical treatments of this problem have been made, but the agreement between theory and experimental momentum space distributions of the continuum electrons is at best qualitative. The purpose of this experiment is to provide bench-mark data for the electron distributions for a *true one-electron system* (bare nuclei on atomic hydrogen), under conditions such that the collision plane and the transverse momentum transfer to the projectile are controlled. We use COLTRIMS techniques to image the momenta of the recoil ion and electron in coincidence. The major technical problem has been to develop a (semi-) cold target of atomic hydrogen. During the past year, we have attempted to do this using a Slevin atomic hydrogen source. The effort has failed, and we are now working to replace this with a 2.43 GHz microwave discharge source. In the meanwhile, a paper on ionization of He at proton energies between 25 and 100 keV is in preparation. Future work on this project will focus on making the experiment work. This project is the Ph.D. thesis work of Erge Edgu.

3) Non-sequential decay of doubly ionized D_2 formed by electron capture by slow Xe^{26+} projectiles *I. Ali, R. D. DuBois*, R..Olson*, S. Hagmann, C. L. Cocke.* When a diatomic molecule is fragmented by electron capture by a slow, highly charged ion, the ion can remain in the vicinity of the molecule during the fragmentation phase, and will thus distort the momentum distribution of the fragments from that of a freely disintegrating molecule. The time scale involved is near ten fs, comparable to that involved in the dissociation of D_2 by intense laser pulses. We have measured the kinetic energy release of the fragments emitted from the dissociation of D_2 in collision with slow Xe^{26+} ions from the KSU CRYEBIS. The energy of the fragments is calculated from the measured momentum vectors of both fragments emitted in the dissociation of the diatomic molecule using a three dimensional momentum imaging multi-hit detector system. The energy is measured separately for the ion pairs from the dissociation of the molecule into different dissociation channels. Clear distortion of the Coulomb sphere is observed, and correlated with the collision plane of the reaction. The results are in good qualitative agreement with CTMC calculations for this reaction. *Univ.of Missouri-Rolla

4) Electron capture from H_2^+ by doubly charged projectiles *I. Reiser, H.Braeuning(U.Giessen) ,C.D.Lin, E.Sidky and C.L.Cocke.* To what extent can a single electron capture from a two-center target be treated in terms of interfering amplitudes for capture from two separate atomic centers? This question has often been addressed within atom-ion collisions, but the experiments have often been rendered difficult to interpret by the fact that the target was not a true one-electron system. The purpose of this experiment is to address this question with a true one-electron molecule, namely H_2^+. The experiment is thus electron capture from H_2^+ by singly charged projectiles (Ar^{2+} , N^{2+} and He^{2+}, and is being carried out both on the JRML ion-ion collision facility and on the Univ. of Giessen Ion-Ion facility. The latter facility is being used to measure total cross sections for the capture, and the former, differential cross sections. The total cross section

measurements show that the cross sections for the two heavy projectiles are much larger, since these reactions are nearly resonant, while the cross section for the He^{2+} (endoergic) is substantially smaller. The results are in good agreement with a simple model based on interfering amplitudes from separated centers. The atomic amplitudes were calculated using a coupled-channel atomic expansion approach. If this model is correct, the differential cross sections are expected to reveal two-center interference patterns. The differential cross section experiment with N^{2+} projectiles is being carried out at KSU. Preliminary data show strong alignment effects, but further data analysis is required to extract complete cross sections as a function of the angle between beam and molecular axis. This project is the Ph.D. thesis work of Ingrid Reiser. Future plans are to complete the data set for N^{2+} , He^{2+} and Ar^{2+}.

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Structure and Dynamics of Atoms, Ions, Molecules, and Surfaces:
MOTRIMS: A New Tool for the Dynamical Study of Atoms interacting with Their Environment

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This abstract reports the progress and future plans of the MOTRIMS project, which is only one part of the J. R. Macdonald Laboratory's overall AMO program. The personnel involved in this project include Dr. C. Fehrenbach (Research Associate Professor), Dr. X. Flechard (Research Associate), and H. Nguyen (Graduate Student).

Recent progress and future plans:

MOTRIMS: A new tool for the dynamical study of atoms interacting with their environment:

Over the past few years, the COLd Target Recoil Ion Momentum Spectroscopy (COLTRIMS) technique has come into its own.[1,2] In this technique ionization processes of atoms and molecules are studied through the reverse kinematics approach of measuring the recoiling target's momentum vector. For this approach to give meaningful results, the momentum received by the target in its interaction must be large compared to its thermal momentum distribution. In COLTRIMS this condition is accomplished by cooling the target atoms or molecules, and then introducing the target into the interaction region *via* a supersonic expansion. The recoil ion momentum vector is measured by extracting the recoils using an electric field, and measuring their flight times and final positions. In an ion-atom collision, the component of the momentum lying perpendicular to the projectile axis is simply related to the projectile scattering angle, while the momentum component lying parallel to the projectile axis is simply related to the Q-value, or difference in the electron's initial and final binding energies. In general, because an electric field is used to extract all of the recoil ions, COLTRIMS is extremely efficient, having 4π steradian acceptance angle and nearly unity detection efficiency. Using the COLTRIMS technique, several different ionizing interactions of atoms and molecules have been studied, including electron- and ion-impact ionization of atoms and molecules, ion-induced dissociative ionization, and photo-ionization.

A recent variation on the COLTRIMS method is MOTRIMS, for Magneto Optically Trapped Recoil Ion Momentum Spectroscopy. In this approach, the target atoms are optically cooled and confined in the interaction region. The extraction and measurement methods are identical to COLTRIMS. MOTRIMS brings with it two main advantages over COLTRIMS. The first is that atoms can be cooled to a much lower temperature in a MOT than through supersonic expansion. Therefore, while in COLTRIMS the ultimate limitation in resolution is due to target temperature, in MOTRIMS the limit is due to position and timing resolution. Thus for example, the best COLTRIMS momentum resolution reported in the literature[1] is 0.06 atomic units for a helium target, whereas in our MOTRIMS setup we have obtained 0.03 atomic units for a rubidium target. (In principle, the more massive the target the *worse* the momentum resolution, all else being

equal. Thus one might expect even better momentum resolution with trapped lithium atoms.)

The second, and perhaps more significant, advantage of MOTRIMS is that the trapped atoms are of necessity accessible to optical excitation by lasers. In COLTRIMS, by contrast, the atoms which are most easily pre-cooled to very low temperatures and then expanded, are the noble gasses and simple molecules. Indeed, among this reduced set of possible targets, only helium comes close to allowing a momentum resolution even close to that possible with a typical MOTRIMS target. Nevertheless, there are many cases in which the conventional COLTRIMS technique is still superior to MOTRIMS. For example, it is not yet technically feasible to trap and cool a sufficient number of molecules to be useful as a target.

The remainder of this abstract addresses progress which has been made in the K-State MOTRIMS project, and includes a discussion of what studies we intend to pursue in the near future. The initial MOTRIMS experiment attempted was a charge transfer study in the Cs^+ + Rb(5s), Rb(5p) collision system.[3] Because the trapping laser was tuned to the 5s-5p transition, the target was a mixture of 5s and 5p states. Far from being a disadvantage, this circumstance allows for the simultaneous measurement of capture cross sections from *both* the 5s and 5p states. The key to this measurement is the MOTRIMS method's extraordinary Q-value resolution of 100 meV for this collision system. This allowed us to distinguish between capture from the two initial states, as well as the final states in the Cs into which the electron is captured. For each of these charge transfer channels we were able to measure the cross sections, differential in scattering angle to a resolution of 25 microradians. Thus these cross sections, differential in angle, and final and initial state provide theory with useful test values against which to test calculation. A typical Q-value spectrum and scattering angle plot are shown in Figure 1.

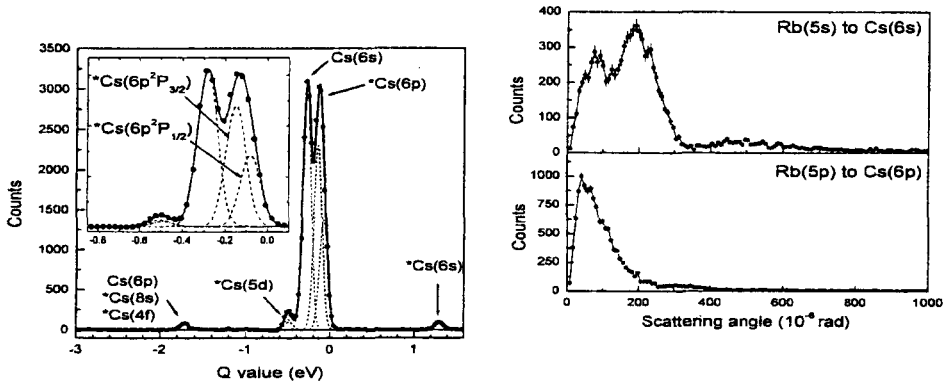

Figure 1 Q-Value and scattering angle spectra

What remains to be done along these lines is to continue the cross section measurements for a variety of alkali and alkali earth projectiles throughout a range of collision

velocities. This will be efficiently accomplished through the use of only two different thermionic ion sources, one which outputs beams of Li^+, Na^+, K^+, Rb^+, and Cs^+, and the other which outputs beams of Mg^+, Ca^+, Sr^+, and Ba^+. Rather than mass select these ions and carry out charge transfer experiments one element at a time, we intend to allow all of the ions species to participate in collision events at the same time. This is possible since the time-of-flight measurements easily distinguish between the different mass projectiles. Proof of principle for this mode of data acquisition has already been carried out using a Na^+ source which (inadvertently) contained a contamination of K^+. Besides having the obvious time advantage of massive multiplexing, this mode has the practical advantage of allowing us to compare on a *single relative scale* the charge transfer cross sections for all the different alkali (or alkali earth) ions.

The second line of experiments which on which we have begun using the MOTRIMS apparatus involves the use of a second laser to excite the rubidium from 5p-4d. Na^+ ions are then used to probe the resulting mixture of states *as a function of time* as the second laser is blinked on and off. Remarkably, the same resolution in Q-value that allows one to distinguish between various initial and final state channels, also allows one to separately determine the relative level populations and the relative charge exchange cross sections. That is, in the same measurement we are able to determine n_d/n_s,n_p/n_s, σ_d/σ_s, and σ_p/σ_s, where n_i and σ_i the population and transfer cross sections, respectively, of the i^{th} target state. This is possible since while the relative populations of the states vary on the tens of nsec time scale, the total target population remains constant on the ms time scale. Thus, the MOTRIMS apparatus used in this time-dependant mode can be seen as either a tool to measure relative cross sections from the various levels in a (laser-excited) MOT, or equally well as a technique which uses the ion beam as a diagnostic to study the dynamic development of the relative populations of the different levels in the MOT. The time dependant capture signals are represented in Figure 2.

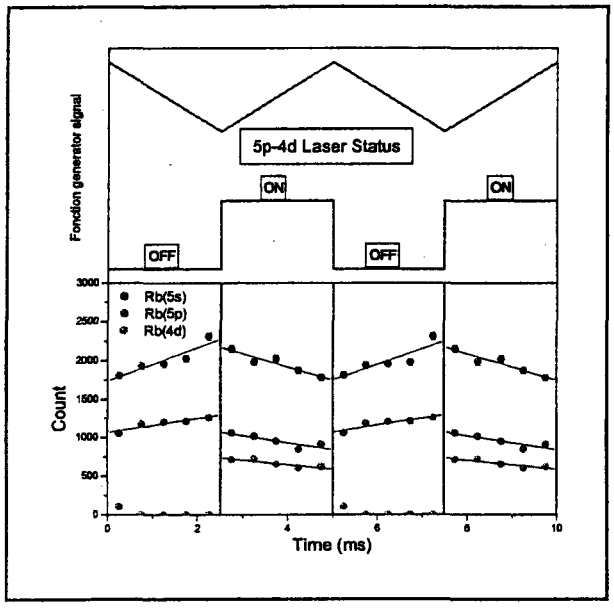

Figure 2 Time dependance of capture signals

The triangle wave is synchronous with the square wave, and both are sent to the data acquisition computer. The Q-value spectra are then sorted by the amplitude on the triangle wave and by whether or not the square wave has the value 0. Unlike a typical capture experiment, in which total charge transfer signals (*i.e.* capture from 5s, 5p, and 4d) would be collected and summed, the MOTRIMS technique allows one to distinguish between relative capture from each of these initial states. Thus, with this extra information, one can readily deduce both relative populations and relative cross sections. This technique is very general and could be used to analyze the effects of any number of processes taking place in a MOT. For example, in experiments in which one studies dimer formation[4] in cold collisions, the molecule production as well as the accompanying depletion of the 5s and 5p states could be measured. As another example, one could illuminate trapped ground state atoms with a femtosecond laser, and watch the time dependence of the decay of the various states excited in the process. The use of this technique as a probe of the dynamics of the trapped atoms under the influence of some external perturbation seems limited only by the ability of the technique to resolve the Q-values of the different capture channels.

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Structure and Dynamics of Atoms, Ions, Molecules, and Surfaces:
Multi-Particle Dynamics in Ion-Molecule Collisions

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Recent progress and future plans:

Ground state dissociation of HD^+

The fast removal of one electron from a hydrogen molecule is followed by very slow dissociation if the vibrational continuum of the electronic ground state is populated in such a vertical transition. For the HD isotope this happens in about 1% of all transitions to the $HD^+(1s\sigma)$. For this isotope of hydrogen the dissociation favors $H^+ + D(1s)$ over $H(1s) + D^+$ production due to the small, 3.7 meV, energy gap between these two dissociation limits[1,2]. We have used this slow dissociation process of hydrogen to probe a couple of reaction mechanisms:

1. Charge transfer and elastic scattering in very slow $H^+ + D(1s)$ "half" collisions – *E. Wells, I. Ben-Itzhak, K.D. Carnes, B.D. Esry in collaboration with H. Sadeghpour*

We have studied charge transfer and elastic scattering in the $H^+ + D(1s)$ collision system. This system provides an interesting case study for theorists, since scattering calculations for this system depend in turn on HD^+ structure calculations, which must correctly account for the difference in nuclear mass. Our aim was to study the collision at energies from about 1 eV down to the charge transfer threshold at 3.7 meV, and below for the elastic channel. To accomplish this goal we used ground state dissociation (GSD) of HD^+ to measure electron transfer from the $1s\sigma$ to the $2p\sigma$ state. Charge transfer occurs at an internuclear separation of about 12 a.u., around the avoided crossing resulting in the $D^+ + H(1s)$ final state. In contrast, $H(1s) + D^+$ is the final state of elastic scattering. The energy of the resulting H^+ or D^+ fragment is typically less than 300 meV and is determined for each fragment by imaging its momentum vector using a COLTRIMS-style apparatus.

The measured relative yields of H^+ and D^+ fragments as a function of kinetic energy in the HD^+ center of mass frame provide a direct measure of the electron transfer probability from the initial $1s\sigma$ to the final $2p\sigma$ state. This experimental approach provides a probe of very slow collisions. It is important to note, however, that during the GSD process the "avoided crossing" is traversed only once in contrast to twice in a "full" collision. As a result of the "half" collision nature of our experiment, we cannot compare directly to theoretical results available for full collisions[3]. Therefore, we performed coupled channels calculations of the transition probability for both half[2] and full collisions[3]. The experimental results[4], shown in figure 1, are in good agreement with the half collision calculations except near the charge transfer threshold where better resolution and subtraction of the H_2 contamination are needed[2,4]. Improvements of the experimental setup are underway in an attempt to probe the threshold behavior of charge transfer and the resonances in the elastic channel.

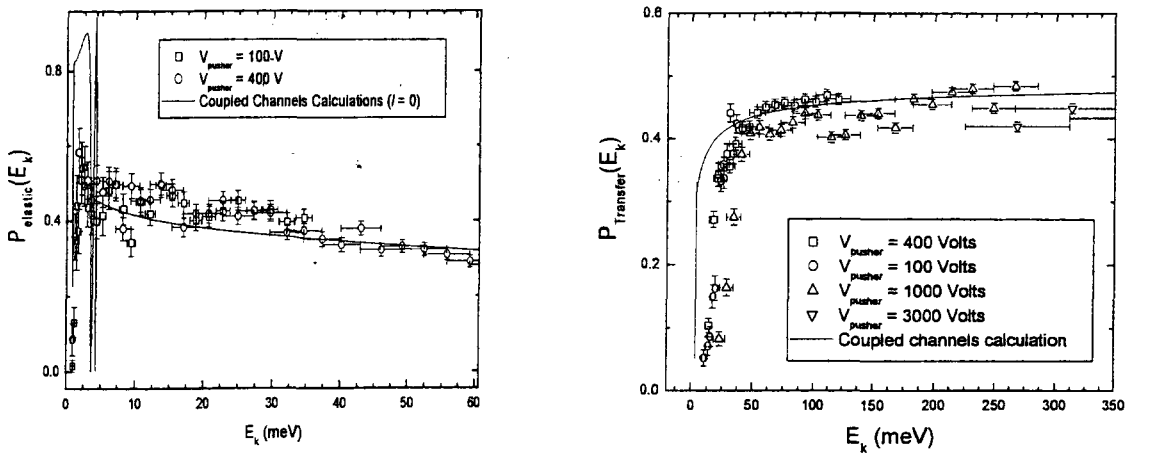

Figure 1: Right: Charge Transfer and Left: Elastic Scattering in $H^+ + D(1s)$ "half" collision.

2. Separating the momentum transfer to a hydrogen molecule as a whole from the momentum transfer to the internal motion of its nuclei in the CM frame -- *I. Ben-Itzhak, C.L. Cocke, M.A. Abdallah, M. Stöckli in collaboration with W. Wolff and H.E. Wolf*

In a collision momentum can be transferred either to the CM motion of the molecule or to the internal motion of its nuclei in the CM frame. We have used the GSD process to probe and separate these two contributions from each other for slow $He^+ + H_2$ (or D_2) collisions (at a velocity of 0.25 and 0.5 a.u.). The GSD fragments are very sensitive probes of small amounts of momentum transfer because of their small dissociation speed. It was found that for these collisions most of the momentum is transferred to the molecule as a whole for both electron capture and ionization processes. However, we observed some transverse momentum transfer to the motion of the nuclei in the CM system for the ionization process and none for electron capture. It is suggested that direct scattering off one nucleus is the cause of this momentum transfer.

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The high velocity limit of the double- to single-ionization ratio of hydrogen molecules -- *I. Ben-Itzhak, E. Wells, D. Studanski, Vidhya Krishnamurthi, K.D. Carnes, in collaboration with H. Knudsen*

The ratio of double to single ionization of helium by fast-ion impact has been a topic of intensive research. There has been particular interest in the high-velocity limit, which is a measure of the importance of electron-electron mechanisms for ionization of the second electron[1]. In contrast, the same ratio for the hydrogen molecule has not been determined as accurately. We have measured this ratio for fast proton impact as a function of the proton velocity. Using

electron-impact data[2] and our measurements the high-velocity limit was determined to be 0.0018±0.0002 %. This ratio is about a factor of 1.4 smaller than the same ratio for the atomic helium target. Most of this difference between the atomic and molecular two-electron targets stems from the higher probability to ionize one electron in hydrogen than in helium, which in turn is due to the lower binding energy in hydrogen. It was shown that, for 1-12 MeV proton impact, the single ionization of hydrogen is about a factor of 1.8 larger than that of helium. In contrast, the double ionization of these two-electron targets is approximately the same. It is suggested that the stronger electron-electron interaction in the helium target caused by the smaller average distance between the target electrons compensates for the higher probability for ionizing the first electron in hydrogen. Whether or not this is actually the case requires a quantitative theoretical treatment to determine.

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Formation and decay mechanisms of doubly charged molecular ions -- *I. Ben-Itzhak, Z. Chen, B.D. Esry, C.D. Lin, E.Y. Sidky. In collaboration with: A. Bar-David, J.P. Bouhnik, I. Gertner, B. Rosner, Z. Amitay, O. Heber, D. Zajfman, W. Koch, R. Röhse, W. Klopper*

The experimental mean lifetime of $^3He^4He^{2+}$ molecular ions (of about 300 ns) has been attributed to high angular momentum states. It has been suggested that the source of this angular momentum is the creation mechanism of the $^3He^4He^+$ parent molecular ions in the ion source. The suggestion that He_2^+ dimers are preferentially formed with high angular momentum is of great importance for those who are interested in ion chemistry. However, it is based on the assumption that the decay of the $^3He^4He^{2+}$ is by tunneling of the electronic ground state and that the potential energy curve is known precisely. To verify that the association of the measured lifetime with the tunneling of the ground state is correct we developed a new experimental method, which enables the simultaneous determination of the mean lifetime and kinetic energy release (KER) upon dissociation. This method is based on 3D imaging of the dissociating fragments using a CCD camera and a split anode photo multiplier tube coupled to a microchannel plate detector with a phosphor anode, as shown in figure 2. The first gives precise position information (δx~30 μm) while the second provides a precise measurement of the time difference between the two fragments (δt~0.31 ns). We tested this new method by measuring the decay rate of another interesting molecular ion[1], namely $^{12}C^{16}O^{2+}$.

Using the same method the mean lifetime and KER of long-lived $^3He^4He^{2+}$ molecular ions were determined to be τ=164±20 ns and $9.8\pm^{0.2}_{0.4}$ eV as shown in figure 3. A distribution of states peaked around v=1 and l=14,15 is in nice agreement with the data. To verify that no other decay process may contribute we have calculated many excited electronic states of the $^3He^4He^{2+}$ molecular ion and found one state, the $a^1\Sigma^+$ state, which can decay by dipole transitions. The computed mean lifetimes and KER of this excited state, however, are not in agreement with the data. Following the elaborate experimental and theoretical work, we can state that the original suggestion associating the measured mean lifetime with highly excited rotational states is verified. This high angular momentum has to originate from the formation mechanism of the parent $^3He^4He^+$ molecular ion, because the angular momentum does not change significantly in fast charge stripping collisions. Further studies of the collisions forming the $^3He^4He^+$ in the plasma are called for. In order to determine if these high angular momentum states were measured just because they matched the range of high experimental sensitivity or because He_2^+

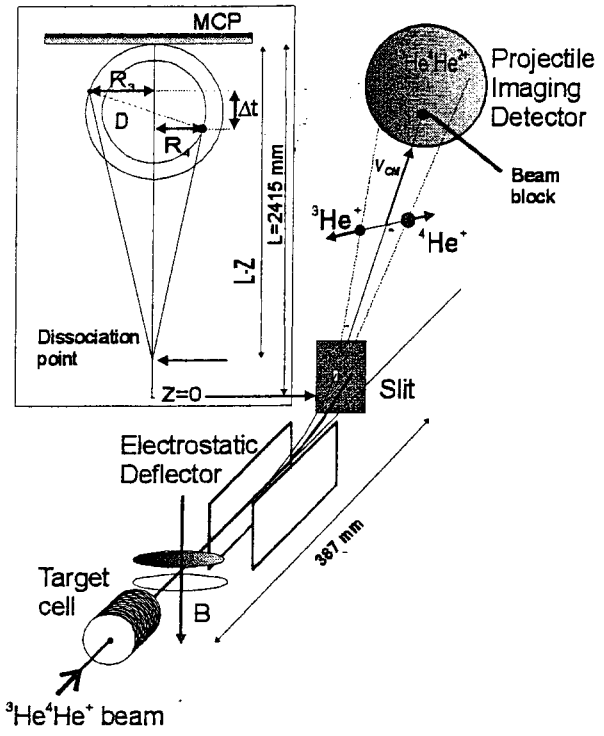

Figure 2. A schematic view of the 3D imaging experimental setup.

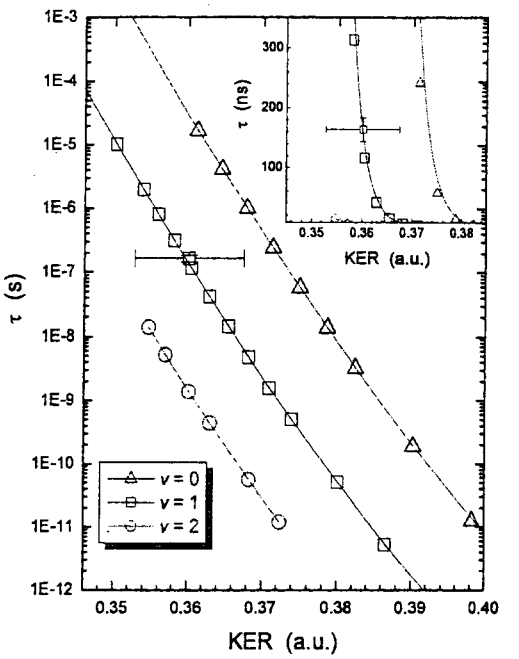

Figure 3. Calculated and measured mean lifetimes and KER values of many rovibrational states of $^3He^4He^{2+}$.

formation really peaks at high l values, we have recently conducted similar measurements of the homonuclear $^4He_2^{2+}$ dimers. For this isotope low l-states as well as high l-states are within the high sensitivity range of our experimental setup. Preliminary results suggest that high angular momentum states are preferred.

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1. "Measurements of the mean lifetime and kinetic energy release of metastable CO^{2+}," J.P. Bouhnik, I. Gertner, B. Rosner, Z. Amitay, O. Heber, D. Zajfman, E.Y. Sidky, and I. Ben-Itzhak, Phys. Rev. A **63**, 032509 (2001).
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Structure and Dynamics of Atoms, Ions, Molecules, and Surfaces:
4-Body Dynamics in Ion-Atom Collisions: towards (e,2e) Spectroscopy in Ions

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Recent progress and future plans:

Kinematically complete differential electron impact ionization cross sections for ions are, particularly in the relativistic regime, a formidable challenge to theory; however, such an experiment is not feasible using standard crossed beam technique due to low luminosity. We have now shown that in kinematically complete measurements of projectile ionization in ion-atom collisions the eN(ionizing interaction between the target nucleus and the projectile electron) and ee(target electron is considered quasifree and ionizes the projectile electron) channels can be separated. The kinematically complete measurement of the ee channel is equivalent to (e,2e) experiments measuring 5fold differential electron impact ionization cross sections of the projectile ion, but in inverse kinematics. For total and partial ionization already Wu et al/1/, Dörner et al/2/, based solely on the spectroscopy of recoil momenta and Zouros et al /3/ independently on resonance behavior of the ionization cross section, have demonstrated that the ionization of the projectile in an encounter with a target atom can proceed via the eN and the ee branch.

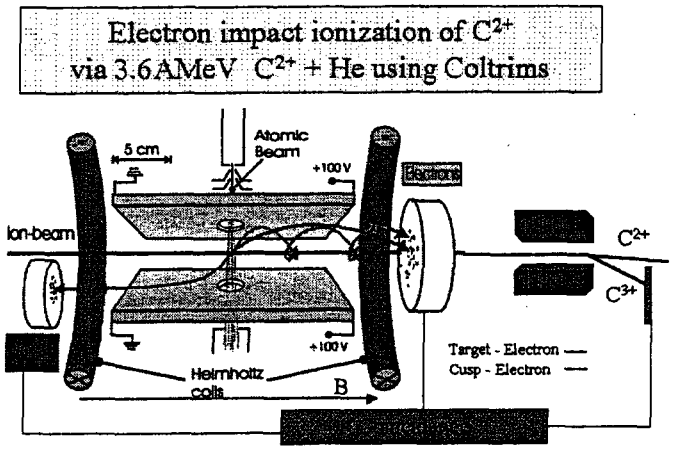

Fig.1. Experimental setup

The current experiment was carried out using the reaction microscope/4/ at the UNILAC at GSI. The setup is illustrated in fig.1. In the current setup the magnetic guiding field **B** was carefully configured to map not only slow electrons as usual but all fast electrons produced in the collision with velocities v_e close to the beam velocity v_{Proj} =12.a.u. and emitted into a narrow cone around the beam direction onto the 2D position sensitive multihit electron detector as well. Kinematic coincidences between a fast and a slow electron and a recoil ion are registered using standard fast electronics when a charge changed projectile $C^{2+} \rightarrow C^{3+}$ was detected in coincidence. A kinematically complete reconstruction of the momentum components of all collision partners is then possible.

It can be argued that ee and eN channels for projectile ionization shall exhibit distinguishing kinematical characteristics. In a collision where a projectile electron is ionized by the target nucleus an azimuthal angular correlation between the fast electron and the He ion recoiling with characteristic momentum is expected; the slow shake-off electron is not exhibiting an angular correlation with the He recoil.

When instead the interaction between projectile electron and He target electron effects the ionization the angular correlation is different: here the recoil remains a spectator with

a momentum distribution characterized by the Compton profile. The angular correlation between the two active electrons is then clearly expressed. This is indeed born out by an analysis of the coincident events as given in fig.2.

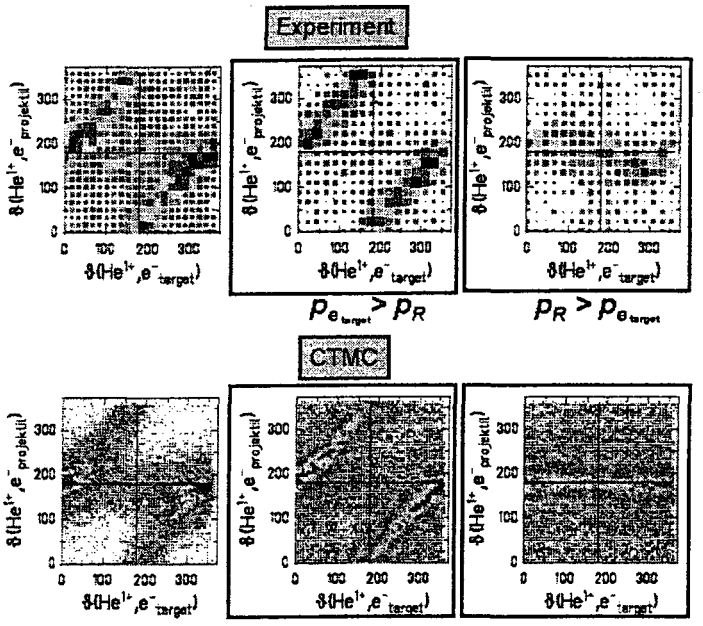

Fig.2. Azimuthal Correlation of Electrons withRecoil

In a 2D diagram azimuthal angles of the He^{1+} recoil ion with the slow target electron are mapped versus the azimuthal angle of the He^{1+} recoil with the fast electron. The top leftmost diagram in fig 3 contains all coincident events, in the top right most diagram, selecting $p_R > p_{e\text{-target}}$ it is apparent that He^{1+} and the fast electron are emitted 180^0 with respect to each other, a signature of the eN process. In the top center diagram events with very small recoil momenta $p_R < p_{e\text{-target}}$ are selected; the two diagonal lines indicate the strong azimuthal correlation $\Delta\varphi(e_{Proj}, e_{target})=180^0$ between the slow and fast electron. We point out that at the current collision energy the ee channel dominates the ionization cross section. In the second row CTMC calculations by Olson et al. for the same reaction is given, they show excellent agreement with the data.

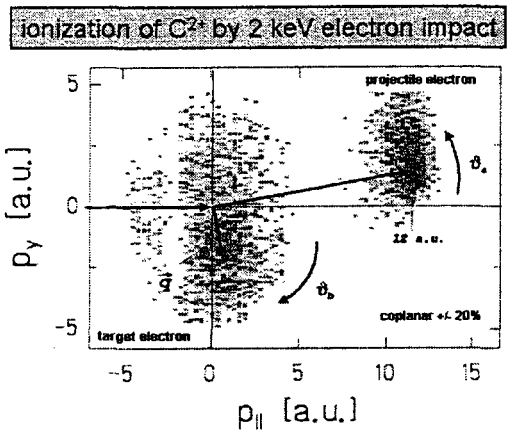

Fig.3. Scattering plane for electron impact ionization of C^{2+}

In fig. 3 the collision plane for equivalent ionization of C^{2+} by 2 keV electrons derived from the present experiment is presented, transformed into the C^{2+} restframe, no selection of a momentum transfer window is made due to lack of statistics. It is apparent that for the next experiment with enhanced statistics angular distributions for ionized electrons for a range of momentum transfers, i.e. full fivefold differential cross sections, can be derived.

This work was performed in collaboration with H.Kollmus, J.Ullrich, R.Moshammer.

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Correlated Three-Electron Continuum States in Triple Ionization by Fast Heavy-Ion Impact

M. Schulz, R. Moshammer, W. Schmitt, K. Kollmus, R. Mann, S. Hagmann, R. E. Olson, and J. Ullrich

Phys. Rev. A **61**, 022703-1 (2000)

Electron Correlation Observed through Intensity Interferometry

M. Schulz, R. Moshammer, W. Schmitt, H. Kollmus, B. Feuerstein, R. Mann, S. Hagmann, and J. Ullrich

Phys. Rev. Letters **84**, 863 (2000)

Momentum Profiles for Single- and Mini-Electron Continua in Strongly Perturbing Collisions of Heavy Ions with He, Ne, and Ar

S. Hagmann and I. Ali

Physica Scripta T**80**, 329 (1999)

Initial State Dependence of Low-Energy Electron Emission in Fast Ion Atom Collisions

R. Moshammer, P. D. Fainstein, M. Schulz, W. Schmitt, H. Kollmus, R. Mann, S. Hagmann, and J. Ullrich

Phys. Rev. Letters **83**, 4721 (1999)

Theoretical studies of interactions of atoms, molecules and surfaces:
Triply excited states of atoms and Ion Impact ionization of atoms

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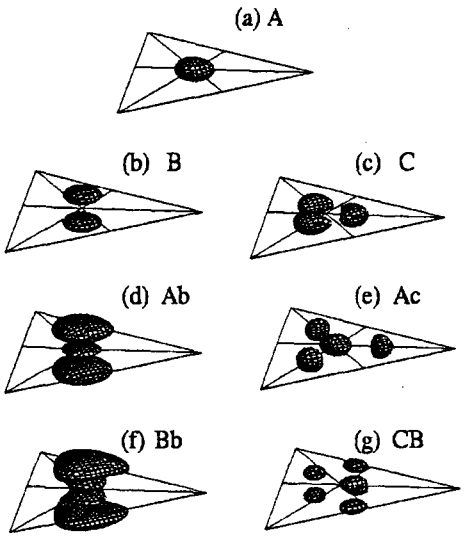
In this abstract we report progress and future plans in the theoretical developments in two areas: (1) the classification and analysis of rovibrational normal modes of triply excited states of atoms; (2) the electron momentum distributions and the role of potential saddle for ion impact ionization at low energies. Computations carried out in conjunction with new ion-atom, ion-ion and ion-molecule collision experiments and in radiative decay of helium doubly excited states are also summarized.

1. Classification and Analysis of Triply Excited States of Atoms

Background.

The goal of this program is to understand the nature of electron correlation in triply excited states of atoms and the eventual proposal of a new set of quantum numbers to classify these states. We have succeeded in the past in: (1) developing a general hyperspherical approach for calculating the adiabatic potential curves of a three-electron atom; (2) finding new ways of analyzing channel functions in the body-frame of the atom; (3) discovering the constraints of quantum symmetry imposed on the body-frame wavefunctions. These basic tools have allowed us to extract some information on the correlation properties of a selected subgroups of low-lying triply excited states in the last few years. In particular, when all three electrons are in the n=2 subshell, i.e., for the eight so-called $2l2l'2l''$ triply excited states, they have been found to fall into three groups---- each group with its own distinct correlation properties, or internal shapes. The next question we tried to address in the last couple of years is how this classification be extended to the higher intrashell triply excited states. There are sixty-four $3l3l'3l''$ triply excited states and the classification of these sixty-four states has been the main focus in the last year. Within the last few months, we have finally succeeded in classifying all these sixty-four states. For practical purpose we can claim that we have classified all the intrashell triply excited states.

Progress in the last year



To classify the $3l3l'3l''$ triply excited states we analyze the wavefunctions of a model atom with the three electrons confined to the surface of a sphere since intrashell states are distinguished by their relative angles with respect to each other. By analyzing the wavefunction in the body-frame where the overall rotational motion can be separated, each state is represented by a body-frame function which has three degrees of freedom. Using contour surfaces we have been able to actually visualize the wavefunctions directly. From the calculated electronic densities we have been able to identify the internal shape of each state. **Examples of such contour surfaces are shown on the left** without full explanations (see pub. A1 below). First, the groups A, B and C are the elementary normal modes that have been identified previously for the $2l2l'2l''$ states. However, new modes such as Ab, Ac, Bb and CB which are excited states of these normal modes have been found. The symbol Ab shows that it is a combined mode of A and B. With these excited modes identified we have been able to classify all the 64

$3l3l'3l''$ triply excited states of the model atom into groups of the rotational levels of a symmetric top, and the shape of each symmetric top is "visualized" through the contour surfaces above.

After all the sixty-four $3l3l'3l''$ states of the model atom have been classified, we found that the energy levels of the 64 intrashell $3l3l'3l''$ triply excited states of N^{2+} and N^{4+} calculated by Vaeck and Hansen [J. Phys. B25, 883 (1992)] can indeed be classified into manifolds of rotational states as in the model atom, proving the validity of the present classification scheme.

Future Plan

We have found the classification scheme for the $2l2l'2l''$ and $3l3l'3l''$ triply excited states. Thus the classification of intrashell triply excited states is basically complete. Our next goal in the immediate future is to classify ***intershell*** triply excited states. We are looking at the $2l2l'nl''$ states with n greater than 2. We need to identify the correlation between the inner pair electrons and the outer electron, and examine the possible normal modes. We need to find out what is the condition that intrashell states can be fit into the Rydberg series. This is the next step for us to reach an eventual full classification scheme for triply excited states of atoms.

2. Mechanism of ion impact ionization of atoms at low energies

Background

Ion impact ionization of a neutral atom is one of the prototype elementary reactions where the final products involve three charged particles. In analogy to the Wannier mechanism for electron impact ionization of a neutral atom near threshold, the potential saddle is expected to play an important role. Consider proton impact ionization of atomic hydrogen, the electrostatic potential experienced by the electron from the two nuclei has a potential saddle at the midpoint of the internuclear line. For slow collisions, ionization is unlikely except when the electron can straddle the potential ridge as the two nuclei separate, since away from the saddle the electron tends to fall into one of the potential wells, to form bound states in the final product. Over the last two decades there have been a lot of debates on the importance of these so-called saddle point electrons, and more importantly, what is the clear experimental signature of such a mechanism. In the last few years we have embarked a new theoretical approach aiming at calculating the electron momentum distributions of the ejected electrons. We developed a new method of solving the time-dependent Schrodinger equation in momentum space such that the momentum distribution of the ejected electrons can be obtained directly.

Progress in the last year

We have previously calculated the ejected electron momentum distributions for proton-hydrogen atom collisions and analyzed the role of saddle point mechanism. We concluded that saddle point mechanism is important only for slow collisions. However the □-electron type momentum distributions in the perpendicular direction, as well as the peak of longitudinal momentum distributions near the potential saddle were found **not** a signature of the saddle point mechanism since both occur at low energies as well as at higher impact energies. Our predictions are still waiting for experimental confirmation.

To pursue this issue further we examined the He^{2+}+ H system in the last year. At high energies, the calculated longitudinal momentum distribution was found to be near the target as expected. As the collision energy is lowered, the distribution is shifted toward the projectile. At further decrease, the distribution then shifts **back** toward the target. The latter shift is an indication of the importance of the saddle point mechanism for ionization at low energies. (see Pub. B1 below)

In the last year we have also performed careful calculations of the total impact ionization cross sections for proton-hydrogen system. In spite of the continuing theoretical and experimental efforts over many decades, the absolute total ionization cross sections for this system still differ by 25-40% in the 15-200 keV region. Our new calculations confirm the results from the largest close-coupling calculations carried out by Toshima. Both differ quite significantly from existing experimental data as well as from other less sophisticated calculations so far.

Future Plan

The direct solution of the time-dependent Schrodinger equation in momentum space we developed so far has some success. However we have recently realized that there is an intrinsic limitation in describing ionization event in a direct numerical approach. It is not practical to integrate the equation directly to large internuclear distances or for a long time since the numerical method would fail. Following the work of Sidky and Esry, it is found that direct numerical integration in a scaled space would eliminate such problems at large time. The latter method has been tested on one-dimensional problems. [see Report by Esry] In collaboration with Esry, we hope to generalize the scaled space approach to ion-atom collisions in the near future. It is hoped that this would allow us to propagate the time-dependent equation to the long time needed to extract the momentum distributions, so that they can be compared directly with experimental measurements for collisions at low energies.

3. Other topics in ion-atom collisions

We continued a number of studies in one- and two-electron processes in ion-atom collisions in conjunction with new experiments. This ranges from single electron capture cross sections to the alignment parameters of proton impact excitation of helium atoms, as well as transfer ionization cross sections at high impact energies. (See Pub. List C below)

Works in progress

A new effort at the J. R. Macdonald Laboratory in the last year is the so-called Motrim experiments in the group of *Brett DePaola*. With laser-cooled atoms in a MOT trap and a new low-energy ion source, they have been able to measure accurate state-selective charge transfer cross sections in the low energy regime with much higher precision. They also can perform experiments from the ground state as well as from the excited states at the same time. Together with the angular measurements, they are providing a new set of mutually normalized experimental data that are valuable for testing the theory critically. We are performing new calculations for systems that have been or will be measured. Already we have found that a more precise description of the internuclear potential is needed for reproducing the differential cross sections.

4. Radiative Decay of doubly excited states of He

In the last year, in collaboration with M. K. Chen (Chung-Hsin University, Taiwan) we have investigated the radiative vs Auger rates of the doubly excited states of He, in view of new experiments in the last couple of years. The radiative rates of He doubly excited states have been neglected by theorists so far since they are very weak, but these weak channels have been measured recently with the new intense synchrotron light sources. We have also calculated the lifetimes of low-lying doubly excited states of He and shown that the states in the A=0 series decay primarily via radiative processes. [see pub. D1]

There is no immediate plan for continuing in this area of work next year.

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A.Triply excited states of atoms

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C. Electron capture in ion-atom collisions

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C5.T. G. Lee, H. C. Tseng and C. D. Lin, " Evaluation of antiproton impact ionization of He atoms below 40 keV", Phys. Rev. **A61**, 062713(2000)

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D. Doubly excited states of atoms

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Theoretical studies of interactions of atoms, molecules, and surfaces: Laser-atom interactions and few-body systems

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1 Program Scope

This research program, begun in the last year, is intended to explore the few-body problem in a variety of circumstances including bound states, collisions, and in external fields. A major component of this program is to develop novel theoretical tools required for this effort. In the last year, special emphasis has been placed on developing a general method for treating time-dependent wave functions in scaled coordinates. Applications of this new approach and plans for other projects are also addressed.

2 Intense laser-atom interactions

2.1 Background

High harmonic generation and above threshold ionization are two of the most familiar phenomena that have emerged from studies of interactions of intense, ultrafast lasers with atoms. Both processes are inherently nonperturbative and involve one or more electrons in the continuum. As such, they are difficult to treat theoretically even after many years of study.

2.2 Recent progress

We (Sidky and Esry) have developed a representation for the time-dependent Schrödinger equation that simplifies the problem considerably. The new representation, a combination of a coordinate and a wave function transformation, removes nearly all of the effects of kinetic energy from the wave function. This "scaling" representation frees us from the necessity of absorbing boundaries, allowing essentially boundary-free propagation of the wave function. It is then only necessary to propagate the relatively smooth envelope of the continuum wavepacket. In collaboration with C.D. Lin, the scaling method has since been applied to the model problem of an intense laser interacting with a one-dimensional atom (Zhao, Esry, and Lin). It was found there that the scaling does indeed reduce the computational burden by roughly an order of magnitude on average. To simplify the wave function even further, the wave function was transformed to the acceleration gauge to account for the effects of the oscillating laser field. We were thus able to propagate the wave function to very large times, allowing us to see directly in the wave function localized continuum wavepackets corresponding to different numbers of photons absorbed, *i.e.* above threshold ionization.

2.3 Future plans

Based upon our experience with the one-dimensional problem, we have hopes that the scaling method can make two- or three-dimensional time-dependent calculations a more routine proposi-

tion than they are currently. The generalization of the method and code is reasonably straightforward, and we fully expect the same computational benefits realized in one dimension. The scaling method shows promise for essentially any time-dependent problem involving the continuum and many applications are possible. One application that we intend to explore in the near future is described in the next section.

3 Ion impact ionization of atoms

3.1 Background

After many years of investigation, the theory for ion-atom collisions is well developed and can handle bound-bound transitions quite reliably. Bound-free processes, on the other hand, are much less reliably treated, but progress is being made — largely with time-dependent methods. One problem that continues to plague current theoretical treatments is the inability to integrate the Schrödinger equation to large internuclear separations. Such long-time propagation is necessary to make the final state analysis less ambiguous, especially for low-energy collisions. The final momentum distribution of the ionized electron can then be extracted and compared with both experimental results and existing theory. It is hoped that with a cleaner asymptotic analysis we can help resolve the discrepancies remaining between them. The need to understand such a prototype process is clear, but is more fully motivated in the report by C.D. Lin.

3.2 Recent progress

This project is only in its beginning stages, but the time-dependent equations to be solved within the scaling method have been derived. We are considering the standard problem of one electron with straightline trajectories for the nuclei.

3.3 Future plans

This project is first on the list for my new postdoc. He will be charged with adapting existing three-dimensional time-dependent codes to solve the equations already obtained. The scaling method has the potential to remove the limitations on integration times, but remains to be explored for ion-atom collisions. There appear to be few obstacles to a successful implementation, but even within the scaling approach we are considering different schemes whose relative efficiency must be evaluated.

4 Helium trimer

4.1 Background

The helium trimer is an intriguing system to study because the helium dimer is so weakly bound. In fact, quantum chemistry texts often state that it is unbound based upon simple bond number analysis. It is bound, but barely so — and only for $^4He_2$; its single bound state has a binding energy of only about 1 mK. Such a weakly bound dimer implies very interesting physics for the trimer in the form of Efimov states. Efimov states are of keen interest to nuclear physicists as they are closely related to the halo states many neutron-rich nuclei possess. Our own previous calculations as well as those of several other groups confirmed that $^4He_3$ has two bound $J = 0$ states and that the excited state has Efimov characteristics. The question naturally arises whether there are any bound states with $J > 0$.

4.2 Recent progress

We have carried out calculations for the rotationally excited states of the helium trimer within the adiabatic hyperspherical approximation (Lee, Esry, Gou, and Lin). Simple estimates and calculations by two other groups had suggested that bound states might exist, but we found only completely repulsive adiabatic potential curves. Thus, no rotationally excited bound states are possible for the helium trimer. One especially interesting aspect of this work was the role of identical particle permutation symmetry. Since a helium atom is a boson, the spatial wave function of the trimer must be completely symmetric under permutations. This requirement forces the adiabatic potentials to be far more repulsive than they would be had the helium atoms been distinguishable.

4.3 Future plans

We currently have no plans to pursue similar calculations in the future, but will revisit the problem if more interesting physics presents itself.

5 Diabatization schemes

5.1 Background

The vast majority of problems of current interest in atomic physics are described by a Schrödinger equation that is not separable in any coordinate system. Adiabatic approximations, such as the Born-Oppenheimer approximation, are convenient and powerful tools for dealing with such equations. Unfortunately, at the very points where the physics gets interesting — near the avoided crossings — the numerical treatment gets difficult. Theoretically, then, a representation that does not have avoided crossings would be desirable. Such representations are generically called diabatic, but it is difficult in general to generate a diabatic representation that includes as much physics as compactly as the adiabatic representation.

5.2 Recent progress

We have recently finished developing one possible diabatization scheme (Esry and Sadeghpour). Standard, strict diabatic transformations are well known and require the nonadiabatic derivative coupling. We have dubbed our new representation the "split diabatic representation" as it is based upon the same equation as the strict diabatic transformation but with only part of the nonadiabatic coupling included. No approximations are made, however, since the remaining coupling is retained in its adiabatic form. The new representation can thus be considered a mixed representation since it is neither purely diabatic nor purely adiabatic, but the avoided crossings are eliminated. The split diabatic representation also avoids one of the problems of the strict diabatic representation, namely the unphysical behavior of the diabatic potentials at large distances. It turns out that because of long range nonadiabatic coupling — present for both the Born-Oppenheimer and the adiabatic hyperspherical representations — the strict diabatic potential curves oscillate sinusoidally at large distances. The long range portion of the nonadiabatic coupling need not be included in the definition of the split diabatic representation, though, so the asymptotic potential curves coincide precisely with the physically appealing adiabatic potentials.

5.3 Future plans

We intend to explore applications of the split diabatic representation. In particular, one advantage of a diabatic representation is the ability to selectively include or exclude individual channels. The advantage is computational since only physically important channels need be included. While the split diabatic representation produces physical potential curves and has the potential to reduce the number of channels, it is not suitable for automatic application to a large number of channels because someone must decide what part of the coupling to use in the definition. We will thus also seek diabatic representations that are suitable for large problems.

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Theoretical studies of interactions of atoms, molecules, and surfaces:

Particle–surface and laser–atom interactions

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1. Electron capture from thin metallic films

Recent progress: Based on the self-energy method [1,2] for charge transfer in ion–surface collision, we are investigating the broadening of atomic levels near thin metallic films. Size quantization in the growth direction of the film gives rise to characteristic structures in level widths, atomic occupation probabilities, and transition distances as a function of the film thickness [3]. Details of this structure depend on the orientation of the atomic Stark orbitals with respect to the film and can be related to the dependence of transition matrix elements on the active electron's wave vector component parallel to the surface. The large variation of the calculated transition distances with the film thickness may result in observable effects in atomic interactions with thin films.

Future plans: We plan to calculate the full electronic self-energy of hydrogenic atoms interacting with thin metallic films. Adiabatic resonance states generated from the self-energy for thin film targets will serve as basis states in time-dependent close-coupling calculations for the electronic dynamics in atom-film interactions, in close analogy to the semi-infinite case of a metal surface [4,5,6]. The extension of the present calculations to the case of layered nano–structures composed of arbitrary sequences of metals, semiconductors, and insulators, and involving arbitrary z-profiles, is straightforward. Possible applications of calculations of this kind may be found, e.g., in the analysis of electron transfer in ion desorption processes. These processes have considerable practical relevance in surface and thin film analytical methods.

2. Charge–transfer dynamics near metal surfaces

Recent progress: Within a new two-center close-coupling expansion, we solved the time-dependent Schrödinger equation for an active electron interacting with a slow projectile and a metal surface. The continuum of metal conduction band states is discretized in terms of wave packets. We obtained converged results for the time evolution of the atomic and metallic population amplitudes for an excited hydrogen atom near an aluminum surface [5,7,8]. In contrast to the self–energy approach [1], the continuum discretization resolves the active election's motion in the metal Hilbert space and allows for the study of projectile energy loss due to electron–hole pair excitations in the substrate.

Future plans: The efficient discretization of (ionization or conduction band) continua is an outstanding problem that requires the (often neglected) careful analysis of convergence, dephasing, and recurrence effects [6]. We intend to streamline our code and to increase the number of states that represent the discretized conduction band continuum in order to fully eliminate recurrence effects, even at very slow projectile velocities. We also intend to investigate the projectile energy loss due to electron–hole pair and plasmon excitations in the substrate.

3. Wave-packet propagation techniques applied to ion-surface interactions

Recent progress: Apart from contributing to the qualitative understanding of the interaction mechanisms, e.g., through computer animations, the direct propagation of the wave function on a numerical grid also enables the quantitative assessment of (one–electron) charge transfer. This method is flexible in that there are practically no restrictions on the effective one–electron potential used to describe the surface and projectile electronic structure (and the perturbations induced during the interaction).

Resonance positions and widths can be obtained by propagating the initial electronic state of the projectile Ψ_0 under the influence of the surface potential while keeping the ion at a *fixed* location $\vec{D}$ in front of the surface. The wave function at any time t is given by (unless stated otherwise, we use atomic units)

$$\Psi(\vec{r},\vec{D},t) = \exp\{-iH(\vec{r},\vec{D})\,t\}\;\Psi_0(\vec{r},\vec{D})\;, \quad H = T + V_{surface} + V_{atom} \tag{1}$$

The ion–survival probability is obtained by following the reflected projectile until the integrated probability density around the projectile has reached a stable value.

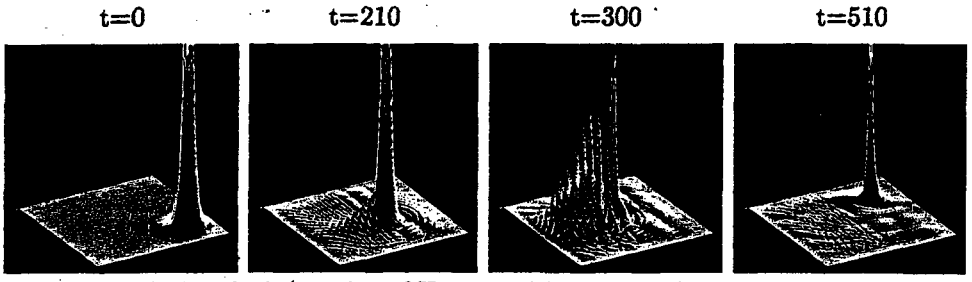

Fig. 1. Charge–transfer during the interaction of H^- ions with a model aluminum surface.

Fig. 1 shows the electronic probability density evolution for the scattering of H^- on a metal surface. The ion comes from the lower right corner of the x–z plane and is reflected to the upper left corner. The plot shows $ln|\psi(x,0,z,t)|^2$ obtained by three dimensional wave function propagation. The exploratory calculation is based on the corrugated surface potential

$$V_{surface}(\vec{r}) = 0.5[\Theta(z) - 1][1 + 0.5\cos(x)\cos(y)\cos(z)], \quad \Theta(z) = atan(z)/\pi + 0.5. \tag{2}$$

Θ is a smeared–out step function. The effective potential for H^- was modeled by regularizing

$$V_{atom}(r_p) = -(1 + \frac{1}{r_p})\exp(-2r_p) - \frac{\alpha}{r_p^4}\exp(-\frac{r_0^2}{r_p^2}) \tag{3}$$

at the origin. r_p measures the distance from the projectile center, $\alpha = 2.25$ is the atomic polarizability of H, and $r_0^2 = 2.547$. V_{atom} allows for the computation of the negative–ion ground state Ψ_0 of H^- and reproduces its affinity (0.75 eV). The propagator in (1) was constructed using the split–operator Crank–Nicholson technique on a 251 x 151 x 251 numerical grid with constant grid spacing, covering 100 x 60 x 100. a.u.^{3} in coordinate space. The projectile was led along a broken–straight–line trajectory, starting at $t = 0$ at a distance of 30 a.u. from the jellium edge of the surface with an incident velocity $(v_x, v_z) = (0.1, -0.1)$ and with $d_{min} = 2$. Boundary conditions at the edge of the grid were imposed by a suitable absorptive complex potential. Shortly after $t = 510$ (last frame in Fig. 1), the resonant flux of electron probability density has stopped. The ion–survival probability, obtained by integration over the atomic probability density at $t = 600$ amounts to 0.07.

Future plans: We intend a detailed investigation of the influence of crystal orientation, surface states, and image states of the substrate on the transfer of charge to and from a projectile.

4. Ionization in intense laser fields: distribution of emitted electron momenta

Recent progress: The basic features of single ionization by a strong, short laser pulse can be studied within the restricted dimensionality of a 1D model atom. Such a model target is given by the soft-core Coulomb potential, $V_{1D}(x) = -\frac{\gamma}{\sqrt{x^2+\alpha^2}}$, with parameters $\alpha = \gamma = 1$. The ground state $\psi_0(x)$ in this potential is bound with $\epsilon_i = 18.23\ eV$.

We expose this atom to a laser pulse of the form $V_L(x,t) = E_0 f(t) sin(\omega t)x$, with a Gaussian envelope function f(t) of width (FWHM) of 500. We further assume a laser wavelength of 780 nm, corresponding to the angular frequency $\omega = 0.0584$ and period $T = 107.59$. An assumed intensity of $I = 10^{15}\ W/cm^2 = 0.0285$ corresponds to an electric field amplitude $E_0 = 8.69 \times 10^8\ V/cm = 0.169$.

The given values of laser frequency, intensity and atom binding energy result in a ponderomotive energy of $U_p = I/(4\omega^2) = 57.6\ eV = 2.1$, a maximal classical excursion range of the electron of $x_{exc} = 4.46\ E_0/\omega^2 = 221$, and a Keldysh parameter $\gamma = \sqrt{\epsilon_i/(2U_p)} = 0.4$. The value of $\gamma < 1$ tends to favor tunneling as the most likely ionization mechanism.

Due to the interaction of the atom with the laser pulse, the electronic wave function evolves according to

$$\psi(x,t) = \exp\{-i\int^t dt' H(x,t')\}\,\psi_0(x) \tag{4}$$

where the Hamiltonian is given by $H(x,t) = T + V_{1D}(x) + V_L(x,t)$, with T for the electron's kinetic energy.

We obtain the time-dependent wave function $\psi(x,t)$ by numerically propagating ψ_0 using the split-operator, Cranck-Nicholson method. We use time increments $\delta t = 0.03$ and equally spaced spatial grid points that are $\delta x = 0.23$ apart. The spatial grid extents from $x_{min} = -280$ to $x_{max} = 280$ with the atom located at the center ($x = 0$). The stability parameter $\delta t/(2\delta x^2) = 0.31$ is smaller than one, as required for numerical stability. At the edges of the spatial grid, that is for $|x| > 250$, we introduce an absorptive potential in order to avoid non–physical reflections of probability density and in order to impose the correct boundary condition for ionization.

Recent photoionization experiments have determined the emitted electron momentum distribution for single ionization. In order to extract momentum-differential emission probabilities from our calculation, we introduce small 'detection intervals' to the right and to the left of the atom that lie outside the classical excursion range x_{exc} of the electron. We choose these intervals at $I_L = [-250, -225]$ and $I_R = [225, 250]$. Next, we define a momentum grid $\{p_j\}$ with step size $\delta p = 0.04$. δp is slightly smaller than a typical momentum resolution in recoil momentum experiments of $\delta p_{exp} = 0.1$. The largest momentum that we can represent on our spatial grid is given by Nyquist's formula as $1/(2\delta x) = 2.2$.

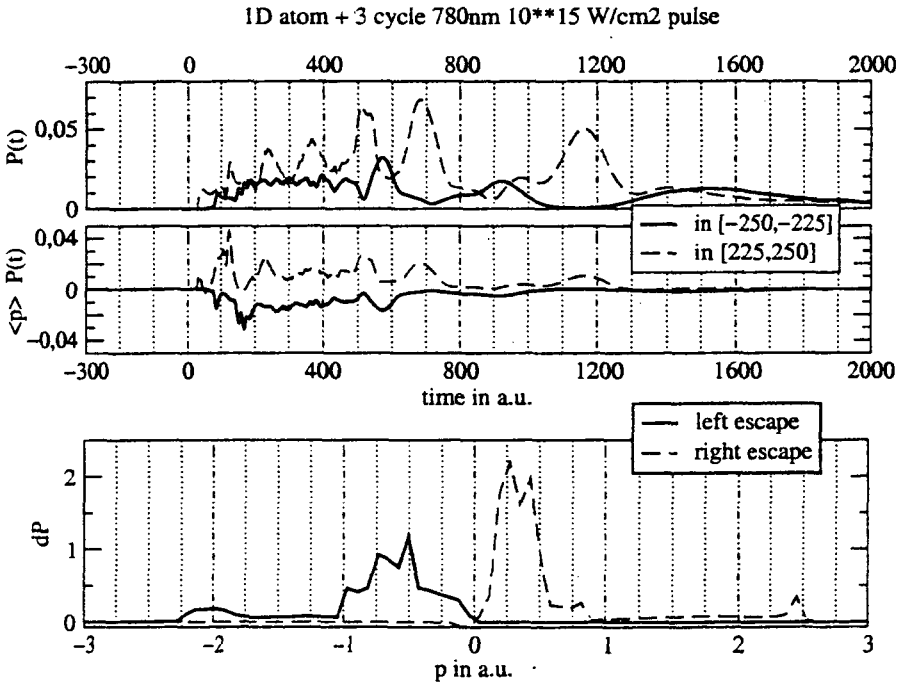

Fig. 2. Probability for finding the emitted electron at distances from the nucleus between [-250,-225] and [225,250] (top). Electronic current through these intervals (middle). Momentum distribution of emitted electrons (bottom).

The probability $dP_R(p_j, \delta p)$ for emission of an electron with momenta in the interval $I(p_j, \delta p) = [p_j - \delta p/2, p_j + \delta p/2]$ can now be related to the 1D released current

$$j_R = Re\{\int_{I_R} dx\ \psi^* p \psi\} \approx -\frac{1}{2} Re\{i \sum_{x_j \in I_R} \psi(x_j)^* (\psi(x_{j+1}) - \psi(x_{j-1}))\} \tag{5}$$

according to

$$dP_R(p_j, \delta p) = \int dt\ j_R \begin{cases} 1 & if < p >_R \in I(p_j, \delta p) \\ 0 & else \end{cases} \tag{6}$$

where $< p >_R$ is the momentum expectation value in the interval I_R. For emission to the left, $j_L, dP_L, < p >_L$ are obtained in the same way.

The top graph in Fig. 2 shows the probability for finding the electron in I_R and I_L for a three cycle pulse. Clearly visible are jets that arrive in the right interval I_R at multiples of the laser period. The middle graph shows j_R and j_L as functions of time. The fastest emitted electrons reach the detection interval I_R at $t \approx 50$; the slowest at $t \approx 1200$, long after the laser pulse. The momentum distributions dP_R and dP_L in the lowest graph indicate most likely emission of electrons with momenta between 0.15 and 0.5 to the right and between -0.9 and -0.2 to the left. The addition of dP_R and dP_L shows a dip around momenta $p = 0$, in qualitative agreement with the double-hump momentum distribution found in recent single ionization measurements.

Future plans: We intend to investigate the Fourier transform of $C(t) = \int dx\ \psi(x,t)^* \psi_0(x)$ in an attempt to identify and characterize field–induced resonances. We have started to investigate double ionization of 2D model atoms.

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Atomic, Molecular, and Optical Sciences at LBNL
Experimental program

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The goal of the program is to understand the structure and dynamics of atoms and molecules using photons, electrons and ions as probes. The current program focussed on studying inner-shell x-ray photo-ionization and photo-excitation of atoms, laser-dressed atoms, and cold molecules. The experimental and theoretical efforts are designed to break new ground and to provide basic knowledge that is central to the programmatic goals of the Department of Energy.

I. Photo-ionization at relativistic energies

This part of the program performs detail studies of atomic collision processes that involve large momentum transfer during the interaction. The goal of these studies is to expand the knowledge of atomic physics into new frontiers in the relativistic regime of atomic collisions and strong fields.

At relativistic energies, the cross section for the atomic photoelectric effect drops off as does the cross section for liberating any bound electron through Compton scattering. However, when the photon energy exceeds twice the rest mass of the electron, ionization may proceed via electron-positron pair creation. We made the first measurement of vacuum-assisted photo-ionization at relativistic energies using the GRAAL beam line at the European Synchrotron Radiation Facility (ESRF) in Grenoble-France. The 1 to 1.5 GeV photon beam is produced through back-scattering of a laser beam on the electrons of the ESRF storage ring. The first experimental results show that in the GeV-energy range inner-shell photo-ionization is dominated by mechanisms that involve pair production confirming theoretical prediction.

The process of vacuum-assisted photo-ionization can proceed either through the encounter of the positron (or electron) with the K-shell electron or through direct pair production on the K-electron. According to theory the first process will dominate for high-Z targets and the second for lower-Z targets. We used Gold and Silver targets to study the relative importance of both these mechanisms of vacuum-assisted photo-ionization. The most important preliminary result with respect to these two mechanisms is that the first one, encounter of the pair with the K-shell electron, is much smaller than the process of ionization triggered by pair production on the K-electron regardless of target-Z. This a significant result from the fundamental perspective because this result is a very strong evidence that the created electron and the positron pop-up (in the first 10^{-21} s) at distances with respect to each other much smaller that the Compton.

II. Laser-synchrotron two-color experiments

Multiphoton processes driven by combined synchrotron (x-ray) and laser (optical) radiation provide a basis for novel scientific directions. From a fundamental perspective, synchrotron x-rays can probe unique states of matter formed while a gaseous or solid target is exposed to intense electromagnetic (laser) radiation. The first experiments are focused on studies of picosecond core-level dynamics in laser-perturbed silicon and on studies of the modification of the x-ray absorption and excitation of argon atoms (gas phase) dressed with high intensity femtosecond laser field. Experiments are performed at the ALS beam line 7.3.1 for the silicon experiments and at beam line 5.3.1 for the gas phase experiments.

We combined a laser and synchrotron radiation to study core-level dynamics in Si samples undergoing rapid laser-induced phase-transitions. Measurements are performed with picosecond time resolution and indicate a rapid and recoverable shift of Si 2p core-state. We used a 400 eV synchrotron light and 800 nm laser pulses focused on Si to an excitation fluence of ~ 10 J/cm^2. A hemispherical analyzer records x-ray photoelectron spectra (XPS) in the vicinity of the Si 2p photoelectron peak. We observe two laser-induced modifications to the XPS: (1) the peak is shifted to lower binding energies and is broadened. These laser-induced modifications are transient and recover within the time-resolution of the measurement (~ 100 ps, set by the synchrotron pulse duration).

We set up the gas phase experiments in beam line 5.3.1 to study the charge state distribution of argon atoms when a K-shell electron is removed by a synchrotron x-ray. We measure the changes to this charge state distribution when the target is dressed with a laser. We built and installed a very efficient time-of-flight detection system that solves the very stringent problems associated with high gas densities that are needed in these experiments. Our measurements of the "laser-off" (static) charge state distribution show a strong post-collision interaction resulting in the recapture of the photoelectron by the argon ions for x-ray energies several eV above K-edge. The spectra measured with "laser off" agree well with similar measurements perfomed at the ALS by W. Stolte with very low gas pressures. We measured the charge state distribution for laser only and are performing the combined laser and x-ray measurements.

III. K-Shell photo-electron emission from oriented molecules

The ability to make complete diffraction images of simple molecules using one of the atomic centers as the photo electron source has been reported recently (PRL 87, 013002 , 2001). Here, the molecular orientation is determined from the measured momenta of the two ion fragments produced following the initial K-shell photoionization and the subsequent Auger decay of the highly excited system. The photoelectron momentum distribution plotted with respect to the fragment momenta then represents that from a "fixed in space" molecule if the system has not rotated significantly before it fragments (this is referred to as the "axial recoil approximation"). In more recent studies (to appear in J. Phys B.) we have continued and extended this approach to measure the angular distribution of electrons released from the K-shell of N_2 and the carbon K-shell of CO by absorption of single linear polarized photons at the LBNL Advanced Light Source, Beam Line 9.3.2 during two-bunch operation. For each ionization event yielding two charged fragments, we determined the angle of the photoelectron with respect to the fragment ion momenta. Measurement of both fragment momenta allows construction of a fragment kinetic energy release (KER) spectrum which reflects the energy levels of the transient doubly (or more) charged molecule excited by the photoionization and Auger processes.

For CO fragmenting to C^+, O^+, we observe resolved vibrational structure for levels associated with KER values below about 10.2 eV. For events in this region, the photoelectron angular distributions shows patterns consistent with emission from molecules which have rotated significantly before dissociation. However, for C^+, O^+ KER values above 10.2 eV, where the spectrum is broadly structured, the photoelectron patterns are characteristic of electron diffraction from fixed molecules. In contrast, similar measurements done in N_2 showed no change in the electron momentum distribution patterns for all KER values in both the N^+, N^+ and the N^+, N^{++} fragmentation channels, and these photoelectron patterns showed clear evidence of diffraction un-smeared by rotational averaging. We thus have found breakdown of the axial recoil approximation for CO for kinetic energies releases below 10.2 eV, whereas for N_2 that approximation is found valid for all fragment energies and doubly and triply charged, two-ion fragment channels. The assumption that the photo-electron emission and subsequent Auger decay are independent was not tested in this study and remains as a topic for future study. This work is collaborative with groups from Kansas State U., Western Michigan U. and U. Frankfurt.

IV. Intense Field Fragmentation of Simple Molecules

A simple molecule, such as H_2 or D_2, immersed in a short intense electromagnetic field, can disintegrate into electrons and heavy atomic or ionic fragments. The process bears some resemblance to multi-photon ionization of atomic targets, but the molecular nature, two heavy bound particles, leads to unique phenomena. We assembled an experimental setup to study this process using the techniques of momentum spectroscopy to ultimately map the distributions of ion and electron momenta following interaction of femto-second laser pulses (intensities up to several times 10^{14} watts/cm^2) with D_2 molecules, and using the femto-second laser facilities of Dr. Robert Schoenlein and colleagues (LBNL Materials Science Division).

We observed a new channel for the production of two D^+ ion fragments, yielding ions with substantially higher energy than previously observed, and with a much broader, nearly isotropic, angular distribution. This work points out how the efficiency and completeness of the coincident momentum spectroscopy approach allows observation of fragmentation channels not visible to earlier studies of the intense field breakup of hydrogen or deuterium molecules. This work has been prepared and submitted for publication; it formed the content of the Diplom Arbeit thesis of U. Frankfurt visiting student André Staudte. C.L. Cocke (Kansas State U.) contributed significantly to these measurements , while on sabbatical leave at LBNL.

V. Cold molecules

The goal is to determine the feasibility of constructing molecular synchrotrons to store, slow, and cool neutral polar molecules in a way that makes it easy to use in new experiments. Working with Juris Kalnins and Glen Lambertson, of LBNL, we developed an analytical approach for the design of focusing elements for strong-field-seeking polar molecules. This technique allows one to vary the relative multipole contributions of any order, producing a more linear element-focusing element with improved transmission and focusing properties. One example shows how focusing elements can transport a 560 m/s beam of methyl fluoride a distance of 30 m and then focus it.

IV. Future plans

Finish the measurements of the cross section of vacuum-assisted photo-ionization for Au and Ag targets. The relativistic electron is equivalent to a very intense photon pulse with a duration of 10^{-21} s. We plan to use the COLTRIMS at the 1.5 GeV electron beam line to study the orientation effects in the dissociation of simple molecules by this highly transient and strong transverse field.

In the two color experiment, perform the "laser-on" and "laser-off" x-ray absorption measurements in beam line 5.3.1 using Ar as a target and extend these measurements to K where the existing femtosecond laser matches the 4s-5p transition which can allowing a more laser controlled x-ray absorption.

Study of C_2H_2, C_2H_4, C_2H_6 will continue while drawing toward a conclusion in 2002. We plan to investigate the degree of correlation between photo-electron and subsequent Auger electron emission for selected molecular targets to compare with a recent report which claims that this can be large. During March 2002, measurements with circular polarized photons will be made at the ALS; these will augment the results taken with linearly polarized X-ray's by allowing unambiguous determination of the relative phases of partial wave amplitudes for the photo-electron emission.

Future momentum spectroscopic studies of the fragmentation of H_2 or D_2 molecules by intense short laser fields will include a more complete description of the newly observed double

ionization channel. In particular its evolution with the field intensity and, through use of photons with variable elliptical polarization, its sensitivity to electron rescattering collisions.

We plan to use accelerator lattice codes that have now been adapted for neutral molecules, by Hiroshi Nishimura, of LBNL, the characteristics and performance of table top synchrotron storage rings will be determined. Storage rings for strong-field seeking and weak-field seeking molecules will be studied. The minimum requirements are: a) molecules stored and detected after thousands of revolutions, b) high intensity through a large acceptance and storage of multiple pulses c) ability to perform experiments while continuing to store the molecules in the ring and adding new molecules.

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Electron-Atom and Electron-Molecule Collision Processes

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Program Scope: This project seeks to develop theoretical and computational methods for treating electron collision processes that are currently beyond the grasp of first principles methods. We are developing methods for treating low energy electron collisions with polyatomic molecules, complex molecular clusters and molecules bound to surfaces and interfaces, for studying electron-atom and electron-molecule collisions at energies above that required to ionize the target and for calculating detailed electron impact ionization probabilities for simple atoms and molecules.

Recent Progress: In the past year, we have made significant progress in two distinct areas covered under this project, namely electron-polyatomic molecule collisions and electron impact ionization.

A major goal of our research in the area of electron-polyatomic molecule scattering is to explore the mechanisms that control the flow of energy from electronic to nuclear degrees of freedom in such collisions. To this end, we have completed the second phase of our work on electron-CO_2 scattering, exploring resonant vibrational excitation in the 4 eV energy region. (The first phase of our attack on this problem was aimed at calculating, from first principles, accurate electronic cross sections for a number of fixed-nuclei geometries.) We have carried out time-dependent wavepacket studies in three dimensions using a novel and highly efficient suite of codes that employ the multi-configuration time-dependent Hartree (MCTDH) approach developed in Heidelberg by Prof H. Dieter-Meyer and coworkers. We have conclusively demonstrated the importance of considering both the bending and symmetric stretch degrees of freedom, which are strongly coupled by a Fermi resonance, and we have computed vibrational excitation cross sections that are in excellent agreement with the most recent experiments (see Fig. 1), reported this year in Physical Review Letters. This study represents the first time that all aspects of an electron-polyatomic collision, including not only the determination of the fixed-nuclear electronic cross sections, but a treatment of the nuclear dynamics in multiple dimensions, has been carried out entirely from first principles.

Our approach to electron impact ionization relies on a mathematical transformation of the Schrödinger equation called *exterior complex scaling* that allows us to solve for the wave function without detailed specification of the complicated boundary conditions demanded by the formal theory of ionization. This approach provided the first complete computational solution of the electron-impact ionization problem for atomic hydrogen which was described in *Science.* In that work, the breakup cross sections were obtained by a numerical extrapolation of the quantum mechanical flux. During the last year, we developed a new procedure for computing ionization amplitudes that provides a more direct and efficient route to computing the breakup cross sections. This provided an independent check on the fundamental correctness of the results we had initially

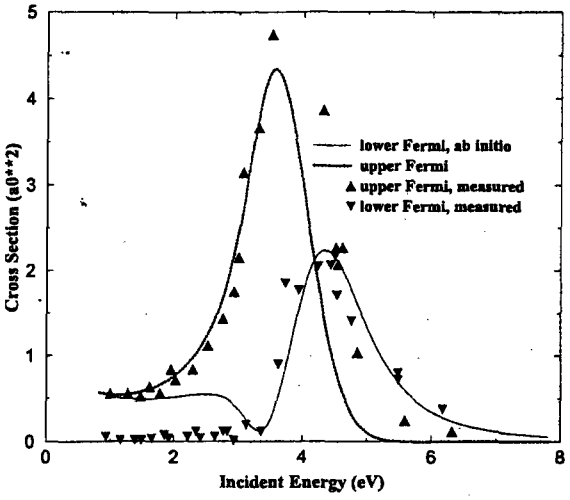

Fig. 1 Cross sections for resonant vibrational excitation of CO_2 Fermi dyad by electron impact. Comparison of *ab initio* results with experiment.

obtained. It also represents a dramatic improvement over the original technique, requiring smaller grids and removing any restriction of the range on secondary electron energies that can be handled. The computational approach we have developed has, to date, provided the only *complete* solution to the quantum mechanical three-body Coulomb at low collision energies, including not only energy sharing differential cross sections but doubly and triply differential cross sections (energy and solid angles for both ejected electrons).

Future Plans:

We will continue to explore the dynamics of vibrational excitation in e^- - CO_2 collisions. The primary mechanism at low energies is resonant excitation of the $^2\Pi_u$ temporary negative ion state. This state, which is doubly degenerate in linear geometry, splits into two distinct states ($^2A_1$ and $^2B_1$) upon bending. Our studies to date have only considered the $^2A_1$ state. Future work will incude studying vibrational excitation dynamics on the complex $^2B_1$ surface and the importance of including non-adiabatic coupling between the $^2A_1$ and $^2B_1$ resonance surfaces. There is new experimental data that suggests that non-Born-Oppenheimer effects may be playing an important role in the excitation mechanisms for certain vibrational transitions.

We will be undertaking similar investigations on the e^- - H_2O system. We will be carrying out coupled electronic channel calculations of direct electron-impact dissociation, as well as nuclear dynamics studies of low-energy vibrational excitation and dissociative electron attachment.

The theoretical approach we have developed for studying electron-molecule scattering has become known as the Complex Kohn Variational method. This approach allows for the inclusion of accurate target electronic states as well as the complete treatment of correlation between the incident electron and those of the target. To study

processes such as electron impact dissociation of a polyatomic molecule, we may need to construct a variational wave function that includes a large number of target excited states. Therefore, a major goal of this project will be to interface the Complex Kohn technology with a modern suite of electronic structure codes. We have chosen to carry out that development with NWCHEM, which was designed to run on distributed memory platforms. For electron collisions with polyatomic molecules our approach will be to build into the NWCHEM framework of codes the complete suite of capabilities that we have been developing for the past decade. We have hired a new postdoctoral fellow who has extensive experience with NWCHEM to carry out this development.

In the area of collisional breakup, a central objective is to extend the methods we have developed to the treatment of electron-impact ionization of complex atoms and small molecules. Key phenomena we want to attack include the quantum interference between direct ionization by electron impact and autoionization of metastable states excited by electron impact. We plan to investigate a new technique for computing the scattered wave function that does not require the solution of large systems of complex linear equations. This technique employs a time-dependent approach in constructing the collision wave function and will be implemented using the discrete variable representation in conjunction with a split-operator method. This time-dependent technique should scale better with particle number than does the linear equations approach. In particular, it will allow us to extend our ionization studies to systems with three active electrons.

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Atomic Physics at the Electron Beam Ion Trap (EBIT)

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Overview

The properties of "Highly Charged Ions (HCI)" and their interaction with electrons, atoms, molecules, clusters and surfaces are being studied at the LLNL Electron Beam Ion Traps (EBITs). *In-situ* studies of the ions produced in EBIT are aimed to determine atomic and nuclear parameters with highest accuracy for use in testing and guiding advanced atomic theory and to set experimental standards for collision physics calculations. In extracted-ion studies the interaction of HCIs with matter is studied. An initial ion-induced equilibrium distortion is caused by high energy density, ultrafast electronic excitation. The energy dissipation and equilibration dynamics of complex systems is investigated. The capability to form multi-component strongly coupled plasmas with HCIs in an ion trap (ion temperature < 1K), that we have developed, affords the study of the equilibrium conditions for these systems and will provide a basis for precision laser spectroscopy in the future as well.

Recent Extracted Ion-Surface Interaction Progress

The luminescence properties of silicon nanodots produced by slow, highly charged ion impact have been investigated. The dispersion of the excitonic features, observed in the luminescence from the silicon nanodots, with laser excitation wavelength (or energy) indicate that the laser-produced exciton does not thermalize before radiatively recombining. The square root dependence of the energy loss (laser excitation photon energy minus luminescence photon energy) on laser excitation energy is explained by a three phonon relaxation process. Two optical phonons are necessary for the vertical excitation and de-excitation transitions and production of one acoustic phonon provides the energy and momentum conservation.

Internal Dielectronic Excitation (IDE) is a correlated atomic physic process that takes place when the deexcitation of a Rydberg electron is accompanied by the excitation of a more tightly bound electron, resulting in a doubly-excited inner-shell configuration. Subsequent x-ray emission involving an electron transition to a shell that initially contained no vacancies identifies the IDE process. IDE is mediated by the electron-electron interaction in a manner similar to a time-reversed Auger transition, and can occur during the neutralization of a slow highly-charged ion interacting with a solid where there are many Rydberg levels that can give rise to correlated transitions to degenerate energy states. We have investigated IDE for a wide range of projectiles and solid targets by measuring the resulting x-ray emission. The characteristic features of the x-ray spectra suggest that IDE occurs above the surface of the solid.

Highly Charged Ion (HCI) Time of Flight (TOF) Secondary Ion Mass Spectrometry (SIMS) has been employed to analyze the changes in the surface composition of canary yellow triamino-trinitro-benzene (TATB) caused by low energy electron bombardment. Comparisons are made between this electron bombarded canary yellow TATB and TATB that has been "greened" by exposure to uv and gamma radiation. These changes, which have been observed through color alteration from yellow through green to black are found to be due to a loss of oxygen.

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In-situ Studies of Highly Charged Ions at the LLNL EBIT

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- **Program Scope**

The properties of highly charged ions and their interaction with electrons and atoms is being studied *in-situ* at the LLNL electron beam ion traps, EBIT-II and SuperEBIT. Spectroscopic measurements provide data on electron-ion and ion-atom interactions as well as accurate transition energies of lines relevant for understanding QED, nuclear magnetization, and the effects of relativity on complex, state-of-the-art atomic calculations.

- **Recent Progress**

The main effort during the past year of *in-situ* measurements of highly charged ions in the Livermore electron beam ion traps has been focusing on the identification and measurement the 1s hyperfine transition in hydrogenic thallium [1]. This measurement yields information that is crucial for understanding the atomic-nuclear interactions needed for atom-based parity violation experiments.

Two measurements of hydrogenic bismuth and lead were carried out in the past at the heavy-ion storage ring at the GSI facility in Darmstadt, Germany; three measurements of rhenium (two isotopes) and holmium were carried out at the Livermore SuperEBIT facility. These measurements have shown that the predicted wavelengths are off by as much as 40 Å, or 1% — a huge discrepancy given that these are "simple" one-electron ions.

We have set out to measure the 1s hyperfine transition in the two thallium isotopes because thallium has the best understood nuclear properties: both the nuclear charge distribution and its magnetic moment are well known from other measurements. Moreover, thallium is used in atom traps for parity violation experiments, and our measurements can therefore be directly used to improve the atomic calculations for the parity violation experiments. Moreover, we have constructed a very high-resolution optical spectrometer that in principle will allow us to measure the thallium lines with much higher accuracy than any previous measurement of the 1s hyperfine transition.

Before using the new optical spectrometer for the thallium measurements we performed careful measurements on a variety of optical transitions in order to understand the properties of the new instrument, understand the calibration procedures, and what problems may arise limiting the accuracy of our measurements. For example, we performed a measurement of the 3800-Å line in Ti-like tungsten that was published as a Rapid communication in Physical Review A [2]. This measurement achieved an accuracy of 0.1 Å, useful to test sophisticated predictions such as those carried out by Donald Beck in Michigan. Although the accuracy of our measurement was ten

times worse than we hoped for, it was five to twenty times better than comparable measurements at NIST and the Tokyo electron beam ion trap facility. The limitations in the accuracy were caused by thermal shifts in the instrumentation, and we learned how to avoid them in future measurements.

We also carried out a range of other measurements in the optical, ranging from nitrogen, neon, and argon spectra to krypton and xenon [3-5]. These measurements were designed to help us develop the best methods for calibration of our measurements. We developed a new technique dubbed "inverted-trap measurements". This technique allows us to calibrate optical spectra *in situ* with well known lines (tabulated in reference books on spectroscopy) from neutral and singly charged ions. We also carried out a series of radiative lifetime measurements that ascertained the role of metastable ion populations in our trap [6-8].

After these preparations, the measurement of the two thallium isotopes proceeded as planned. The only problem was that SuperEBIT had not run for 18 months. Upon startup the achievable charge balance was depressed, and the device could not produce as many hydrogenlike thallium ions as it otherwise would have. Our measurements were, therefore, limited by the count rate, as the signal was week, requiring about 3 months of continuous operation to identify and measure the lines.

The results were 3858.22 ± 0.30 Å for the 1s hyperfine transition in 203-Tl^{80+} and 3821.84 ± 0.34 Å in 205-Tl^{80+}. Despite the poor statistics, the accuracy of our measurement is twice as good as the best previous measurement (that of 209-Bi^{82+} by the GSI group). As SuperEBIT improves with time, we will achieve better statistics, increasing our measurement at least by a factor of five.

The improved accuracy is highly important for basic physics. Our present measurement of the two thallium isotopes for the first time showed that scaling the anomalous magnetic moment from measurements of neutral atoms does not provide the accuracy needed for parity violation studies. Our measurement of the anomalous magnetic moment of hydrogenic thallium differed by three sigmas from the scaled neutral value. This required a reassessment of theory that found that the difference is due to the assumption of a point magnetic dipole interacting with an extended charge distribution. The correct approach that provided agreement with our measurement is to assume an extended magnetization interacting with the extended charge distribution of the nucleus. Without doubling the accuracy of the measurements this crucial find would not have been possible.

In addition to the hyperfine measurements, we have pursued the physics of ion-electron interactions, including the measurement of dielectronic recombination with Prof. Smith from Morehouse College [9,10], electron-impact excitation measurements [11], and magnetic sublevel excitation cross section measurements [12,13], as well as the physics of ion-atom collisions [14-17]. A review of our BES-supported measurements was given in [18].

- **Future plans**

The SuperEBIT device has been moved to its new location in B194 (LLNL), and is again operating in the low energy mode (0.1 – 20 keV), as EBIT-I, and in the high-energy mode (≤ 250 keV), as SuperEBIT. It is now dedicated to *in-situ* ion research. As a result of its new focus, we expect to have an increased availability of run time during the upcoming year for investigating

important atomic physics issues with high accuracy. This also means we will have increased run time available for our current collaborators and future collaborators, who would like to utilize our facility.

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ATOMIC AND MOLECULAR PHYSICS AT OAK RIDGE NATIONAL LABORATORY

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Overview

The ORNL atomic physics program has as its overarching theme and goal the understanding and control of interactions and states of atomic-scale matter. The scientific objective is to enhance progress toward development of detailed understanding of the interactions of multicharged ions, charged and neutral molecules, and atoms with electrons, atoms, ions, surfaces, and solids. Towards this end, a robust experimental program is carried out by our group centered at the ORNL Multicharged Ion Research Facility (MIRF) and as needed at other world-class facilities such as the ORNL Holifield Radioactive Ion Beam Facility (HRIBF) and the CRYRING heavy ion storage ring in Stockholm. Closely coordinated theoretical activities support this work as well as lead investigations in complementary research. Specific focus areas for the program are broadly classified as particle-surface interactions, atomic processes in plasmas, and manipulation and control of atoms, molecules, and clusters, the latter focus area cross-cutting activities in the first two.

Low-energy Multicharged Ion-surface Interactions – *F.W. Meyer, V.A. Morozov, J. Mrogenda*, S. Datz, and C.R. Vane*

This section deals with investigations at the ORNL MIRF studying the interactions of slow, highly charged ions with metal, semiconductor, and insulator surfaces. Our goal is to improve fundamental understanding of neutralization and energy dissipation occurring in such interactions, and subsequently to apply the knowledge gained to probe and modify the surfaces of single crystals, thin films, and nanostructures.

This year we have extended our earlier studies [1,2] of the azimuthal dependences of low-energy projectile scattering in large angle quasi-binary collisions from Au(110). Measurements have been made for 20-keV Ar^{9+} at normal incidence, which are compared with our earlier measurements for this ion at 5 keV and 10^0 incidence angle. A deconvolution procedure based on MARLOWE [3] simulation results carried out at both energies provides information about the energy dependence of projectile neutralization during lattice-site-resolved interactions with target atoms on the ridges, sidewalls, and valleys of the Au(110) surface corrugation. Clear differences in neutralization probabilities and energy dependences between the various possible binary collisions occurring with surface atoms are seen. Such differences must be properly accounted for in order to permit meaningful comparison with theory. To test the sensitivity of the agreement between the MARLOWE results and the experimental measurements, we have performed simulations for both reconstructed and non-reconstructed Au(110) surfaces for 20-keV Ar projectiles, and for different scattering potentials that are intended to simulate the effects on scattering trajectory of a projectile inner-shell vacancy surviving the binary collision. In addition, we have carried out

simulations for a number of different total scattering angles, to illustrate their utility in finding optimum values for this parameter prior to the actual measurements.

As a follow up to similar studies [4] on Au(110), we have measured absolute scattered projectile charge fractions for Ar^{11+} ions with incident energies in the range 3–30 keV that have been 120^0 backscattered from CsI(100) in quasi-binary collisions. Use of a time-of-flight technique that incorporates a biased drift region permitted full separation of all scattered charged states, including neutrals. In contrast to our Ar^{11+} results [4] for Au(110), the scattered neutral fraction is smaller, and relatively independent of incident projectile energy over the entire investigated range. In addition, we have measured, at a fixed energy of 5 keV, scattered charged-state distributions as function of incident charge states in the range 1+ to 13+.

In a separate measurement [4] utilizing electrostatic instead of time-of-flight analysis of the scattered charge states, we attempted to evaluate the effect of surface charging (also known as the 'trampoline" [5] effect) on energy loss of low energy scattered projectiles by absolute measurements of the scattered 1+ energies of incident Ar^{11+} ions incident on CsI(100) at energies down to 10 eV/q. Apart from small deviations from the elastic binary collision energy loss expected for120^0 scattering that are ascribable to the image charge interaction, no measurable effect due to surface charging was found down to the lowest investigated energies.

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Ultra-fast Electron Dynamics in Nano-structures and Bulk Solids – *C. R. Vane and H. F. Krause*

We have mounted the ORNL von Hamos high-resolution X-ray spectrometer on a new target station installed at the HRIBF and initiated measurements using stable heavy ion beams from the tandem to produce characteristic target X rays containing information on the electronic properties of excited target atoms and the surrounding material environment. This research is based on previous measurements at the EN Tandem accelerator for chemical sensitivity of X rays excited by heavy ions [1-3], and is directly related to research on ion-surface neutralization by slow highly charged heavy ions at the MIRF. The spectral information obtained will be used to extract electron transfer rates and electron densities for the specific collision systems and target materials being examined. The precise energies and intensities of satellite

X rays emitted depend on the specific electronic nature of the target ion as it recoils slowly in the substrate. The multiply ionized recoil ion can be partially neutralized in flight by transfer of electrons from the surrounding material. The degree of neutralization prior to fast (femtosecond) radiative filling of an inner-shell vacancy depends on the availability of nearby weakly bound electrons, on their density of states, and on the relevant transfer mechanism rates. These aspects depend on the vacancy states populated in the recoiling ion and on the local and global electronic environments provided by the specific targets. From previous studies, it is anticipated that these transfer neutralization rates and the resultant variations in x-ray spectra will also be sensitive to the limited number and modified conduction electron states available in nano-sized structures, such as quantum dots and carbon nanotubes. Measuring the modifications in x-ray spectra as functions of size and electronic character (conducting versus insulating and/or semi-conducting) of the targets will give rather unique information on rapid electron response characteristics of these new structures. Research is in collaboration with members of the ORNL Solid State and Chemical Technology Divisions characterizing and supplying the carbon nanotubes and other nano-structured targets.

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Near-Thermal Collisions of Multicharged Ions with H and Multi-electron Targets – *C.C. Havener*

Electron capture by multicharged ions from neutrals is important in many technical plasmas including those used in materials processing, lighting, ion source development, and for spectroscopic diagnostics and modeling of core, edge, and divertor regions of magnetically confined fusion plasmas. The ORNL ion-atom merged beams apparatus provides needed benchmark measurements for collisions between ions and ground state H or D in the collision energy range from 20 meV/u to 5000 eV/u. Recent measurements[2] of the total electron capture cross section for Cl^{7+} + D shows a decrease in the cross section toward eV/amu collision energies, in contrast to the slightly increasing cross section observed for other 7+ ions. Fully quantal MOCC calculations performed at ORNL for N^{7+} + D also decrease toward lower energies and agree with the Cl^{7+} measurements, suggesting that the Ne-like core of Cl^{7+} plays no significant role in the electron capture process. The proposed MIRF upgrade will allow measurements with heavier (e.g., Mo^{q+}, Fe^{q+}, ...) ions at eV/u energies to further investigate multielectron core effects.

The duoplasmatron ion source on the merged-beams apparatus has been upgraded to a new Cs negative ion sputter source which allows measurements with a wide variety of neutral atom beams such as Li, B, Na, Al, P, K, Ca, Cr, Fe..., and molecular beams such as O_2, CH_2, ... Our first measurement will be with He^{2+} + Li to try to observe the predicted (Kimura, private comm.) shape resonances formed due to the strong ion-induced dipole attraction between reactants.

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Electron-Molecular Ion Fragmentation, Dissociative Recombination – *C.R. Vane*

A program of electron-molecular ion interaction research is being continued at the CRYRING heavy ion storage ring at Stockholm and, in the future, at the MIRF. One current study deals with developing a reliable base of accurate data on fragmentation of triatomic di-hydride ions in the process of Dissociative Recombination (DR). DR arises from recombination of a molecular ion with a free electron, resulting in disintegration of the molecular ion into several neutral fragments. Previous measurements [1] of DR of vibrationally cold NH_2^+ ions had indicated that the fragmentation of NH_2^+ deviates from that found for several other similar molecular ions. In those previous experiments the (N + H_2) product channel was observed to be completely missing. In all other systems studied (e.g., H_3^+, NH_2^+, and OH_2^+), the corresponding two-body channel has contributed approximately 10% of the DR total, and very nearly half (statistical weighting?) of the other two-body channel (e.g., NH + H). Calculations had shown that part of the signals might have been missed in the previous NH_2^+ branching fraction. An experiment was carried out this year designed specifically for complete neutral fragment collection for all possible DR channels by using a larger surface barrier detector placed closer to the electron cooler target. DR fraction measurements were performed for vibrationally cold NH_2^+ ions and 0 eV electrons. The resultant neutral fragment channel fractions were found to be N + H + H (55%), NH + H (41%), and N + H_2^+ (4%). Thus DR of NH_2^+ does indeed proceed somewhat differently than the other triatomic di-hydrides studied, with the N + H_2 channel reduced by roughly half and the NH + H channel more than correspondingly enhanced.

A second recent experiment involved study of the dynamics of three-body breakup by DR of CH_2^+. These measurements used coincidence timing of the three fragments on a two-dimensional detector, along with a foil-based energy degrading technique to identify the C atom. These methods were developed previously and used for measurements of H_3^+, NH_2^+, and OH_2^+. The data are still being analyzed, but preliminary results indicate that the dynamics of three-body DR of CH_2^+ at 0 eV behave similarly to NH_2^+, in which for the two possible three-body channels ($N(^4S)$ + H + H and $N(^2D)$ + H + H), decay to the excited $N(^2D)$ final state dominates. We will extend these studies of triatomic di-hydrides to develop a systematic base of data over a range of possible initial and final states. This will be implemented by complete studies of BH_2^+ and H_2S^+ ions using the same experimental apparatus and methods. Studies of more complex molecular ion systems, e.g., heavier hydrocarbons, will also be initiated using existing techniques, modified to accept heavier ions and more channels of decay. A program of electron-molecular

ion research is also being developed at MIRF, centering initially on dissociative excitation and ionization of hydrocarbon CH_2^+ and CH_3^+.

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Autodetachment of CS_2 Anions Measured in an Electrostatic Trap – *H. F. Krause*

We have studied the long-term lifetime of CS_2^- ions stored in a linear electrostatic trap. Previously this ion was not studied for times longer than about 10 μs in beam experiments. CS_2 is a candidate gas for use in high-power switching technology. In the experiment CS_2^- ions, formed in a Colutron discharge source containing CS_2, were accelerated to 2.8 keV and momentum analyzed before injection into a linear trap of the Zajfman design. The injected and stored ions oscillate back and forth until they lose their charge by autodetachment or undergo a charge-changing collision with residual gas (2.2×10^{-10} Torr). keV energy neutrals leaving the trap in the downstream direction are detected using a two-dimensional, position-sensitive detector, which indicates the angular distribution and the time dependence of the stored ion population. The metastable ion decay observed indicates three distinct exponential lifetimes of 2.4, 15, and 200 μs. There are no known electronically excited states of the CS_2^- ion. A fourth constant component observed corresponds to stable ground state CS_2^- ions ($0.5 < EA < 1.1$ eV). The unanticipated metastable components may be a consequence of ultra-slow autodetachment from high-rovibrational states of hot and bent CS_2^- ions. Theoretical calculations for the process, $CS_2^- \rightarrow CS_2 + e$, are warranted.

Theoretical Atomic Physics – *C.O. Reinhold, J.H. Macek, D.R. Schultz, S. Ovchinnikov, J. Burgdörfer*

The focuses of our theoretical program are the study of strongly perturbed atomic scale systems, such as atoms, ions, solids, surfaces, and nanostructures subject to electromagnetic fields or particle interactions, and strong synergistic support and interchange with the experimental program at the ORNL Multicharged Ion Research Facility. Recent work includes ion transport in solids, manipulation of Rydberg wavepackets, studies of cold collisions, treatments of ion-atom ejected electron spectra and correlated systems, and neutralization in ion-surface interactions.

For example, the internal state of a fast hydrogenic ion traversing a solid represents a simple open quantum system whose dynamics can be analyzed theoretically and experimentally in great detail. As the ion traverses the foil, the coherence of the internal state of the ion is destroyed due to the interaction with the environment, which is given by particles in the foil and the electromagnetic field. The former leads to multiple collisions whereas latter leads to the radiative decay of the ion. One of our ultimate goals is to be able to simulate the de-coherence of open excited states of atoms as exactly as possible. We study such systems using a Monte Carlo or test-particle discretization approach in which the density matrix is obtained as an average of a large but finite number of density matrices of quantum trajectories (pure states) whose dynamics is governed by a stochastic Schrödinger equation. We have recently shown that for multiple collisions this approach works quite well. This approach will be extended to treat spontaneous radiative decay on the same footing.

We are also exploring new avenues for steering and probing Rydberg wavepackets. Such wavepackets are usually prepared by subjecting stationary Rydberg states to designer pulses. We plan to analyze the possibility of steering wavepackets towards preferred target states using our knowledge of the phase space structure of the kicked Rydberg atom. Trains of pulses can be used to eliminate the part of the initial stationary state that resides in the chaotic sea by diffusion to the continuum. In turn, the fraction of the initial state that resides in one or more stable islands survives as a superposition of selected bound states of the system. In addition to investigating different pump schemes we would like to look into alternative probes. In the last few years we have shown that the time evolution of the Rydberg wavepackets can be probed using short half-cycle pulses, which are good probes of the momentum of the Rydberg electron. We would like to establish that field steps provide an alternative probe that can be used to extract information about the position of the Rydberg electron.

Study is also being aimed at exploring the application of atom optics to the manipulation and control of atomic processes. In particular, cold collisions involve atom-atom and ion-atom interactions where quantum properties are significant. Replacement of the normal adiabatic representation by the hyperspherical adiabatic representation is being developed as a way to treat the interactions rigorously. Our

work has concentrated on simple, universal expressions for dimmer breakup and recombination cross sections in the threshold region relevant to Bose condensation. Magnetic field effects have been included only via the two-body scattering length. Work is in progress to develop a more dynamical approach.

Extending in new directions previous work to develop and apply high-order lattice techniques of solution of the time-dependent Schrödinger equation, we have treated antiproton-impact ionization of helium in a four-dimensional grid model. Good agreement has been found with the most reliable theoretical approaches spanning the range of impact energies from 1 to 1000 keV, adding to the development of a theoretical consensus with contrasts with the lowest energy portion of the pioneering Aarhus-CERN experiments, and opening the possibility of applying the model to other fundamental systems such as H^+ + He or excitons in quantum confined structures. Work has also been undertaken to extend the method to treat the ejected electron spectrum, aiding the broad, fundamental interpretation of recent COLTRIMS experiments.

Finally, we are undertaking a many-body simulation to study the neutralization of highly charged ions near insulator surfaces. Such simulations should follow the projectile dynamics, electron transfer between the ion and the surface, the dynamics of the heavy particles in the crystal, and the hopping of the holes produced by the impinging ion. Since the HCI is repelled by the positive holes, the evolution and redistribution of the microscopic charge-up of the surface is important for understanding the neutralization dynamics. The dynamics of all the heavy particles and the electronic dynamics determining the transfer rates from/to the ion will be obtained using a classical trajectory approach. Using recent experimental information for molecular Auger rates we will attempt to evaluate inter-atomic Auger rates as well. The energy levels and the potentials entering the simulation will be first evaluated using quantum perturbation theory. The hole mobility will be treated as a random diffusive process with a hole velocity distribution obtained from the band structure of the solid.

Recent Publications

1. "Path-Dependent Neutralization of Multiply Charged Ar Ions Incident on Au(110)," V.A. Morozov and F.W. Meyer, Phys. Rev. Lett. **86**, 736 (2001).
2. "Target Orientation Dependence of the Backscattered Intensities and Charge Fractions Observed for Large-Angle Quasi-Binary Collisions of Ar^{q+} Ions with Au(110)," A. Morozov and F.W. Meyer, Phys. Scripta **T92**, 31 (2001).
3. "Charge Fraction Measurements for 2.4 – 35 keV Ar^{q+} (q = 2–13) Projectile Backscattered from Au(110)," F.W. Meyer, V.A. Morozov, S. Datz, and R. Vane, Phys. Scripta **T92**, 182 (2001).
4. "Multicharged Ion-surface Measurements at ORNL MIRF Using Decelerated Beams," F.W. Meyer and V.A. Morozov, *Nucl. Instrum. Methods Phys. Res.* **B157**, 297 (1999).
5. "Simultandous Energy Distribution and Ion Fraction Measurements Using a Linear Time-of-Flight Analyzer with Floatable Drift Tube," V.A. Morozov and F.W. Meyer, *Rev. Sci. Instum.* **70**, 4515 (1999).
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8. "Adiabatic Limit of Inelastic Transitions," P. Krstic, C.O. Reinhold, and J. Burgdörfer, Phys Rev A **63** 032103 (2001).
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13. "Electron Capture and Ionization of 33-TeV Pb Ions in Gas Targets," H.F. Krause, C.R. Vane, S. Datz, P. Grafström, H. Knudsen, U. Mikkelsen, C. Scheidenberger, R.H. Schuch, and Z. Vilakazi, Phys. Rev. A **63**, 032711 (2001).
14. "Investigating the Three-body Fragmentation Dynamics of H_3^+ and H_2D^+," R. Thomas, S. Rosen, F. Hellberg, M. Larsson, C.R. Vane, and S. Datz, *Proceedings, XXII Intl. Conf. on Photonic, Electronic, and Atomic Collisions*, p. 348, editors, S. Datz et al., July 18-24, 2001, Santa Fe, New Mexico.
15. "Lifetime of CS_2 Anions Measured in an Electrostatic Trap," H.F. Krause, C.R. Vane, B. Rosner, and S. Datz, *Proceedings, XXII Intl. Conf. on Photonic, Electronic, and Atomic Collisions*, p. 476, editors, S. Datz et al., July 18-24, 2001, Santa Fe, New Mexico.
16. "Radiative Electron Capture for 33-TeV Pb^{82+} Ions," C.R. Vane, H.F. Krause, S. Datz, P. Grafstrom, H. Knudsen, C. Scheidenberger, and R.H. Schuch, *Proceedings, XXII Intl. Conf. on Photonic, Electronic, and Atomic Collisions*, p. 522, editors, S. Datz et al., July 18-24, 2001, Santa Fe, New Mexico.

Research Summaries

(single-PI grants, alphabetical by PI)

Progress in Obtaining Properties of Transition Metal Atoms

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Introduction

Transition metal atoms are technologically important in plasma physics (e.g. as impurities), Atomic Trap Trace Analysis [1], astrophysical abundance and atmospheric studies, deep-level traps in semiconductors, hydrogen storage devices, etc. The more complicated rare earths, which we are beginning to study, are important in lasers, high-temperature superconductivity, advanced lighting sources, magnets, etc.

These species are computationally difficult to treat–particularly in an ab initio manner–due to the presence of open d and/or f subshell electrons, and the necessity to simultaneously include relativistic and correlation effects. We illustrate the difficulty in Table 1 for several $(d+s)^n\ p^m$ ($m = 0, 1$) levels for the important correlation effects associated with changing two d/s electrons to two f electrons. This table gives the number of eigenvectors (Full) of the correlation manifolds $(d+s)^{n-2}\ p^m\ f^2$, as well as the number of levels (# MCDF) of the $(d+s)^n\ p^m$ manifolds for the indicated J. Note how rapidly these numbers grow with n and m. In the past, cases with $n + m = 4$ were challenging [2]. Today, energy matrices of order 20 000 for transition metal atoms are becoming standard [3,4]. Even so, from Table 1, we see few correlation manifolds could be included within a Configuration Interaction framework [5], if the Full set of eigenvectors had to be kept, for $n + m > 4$.

Methodology and Hardware Improvements

As Table 1 shows, use of REDUCE is a means by which the matrix size may be greatly diminished. This method recognizes that energy matrix elements for fixed manifolds are a linear combination of a small number of radial integrals. This number can be further decreased by ignoring the Breit operator, and assuming that radial functions are independent of j. One can then "rotate" one manifold's eigenvectors to generate the largest number of zero elements. This is done when the second manifold is the reference function ("MCDF"). Consistent with a first-order wavefunction form, the zero vectors are discarded. The number of survivors is given in Table 1 ("Reduce").

In the past, generating the zero vectors involved much user intervention. Decisions had to be made as to which subset of vectors to use (the problem is an underdetermined one), which could vary somewhat from reference to reference. Previously ($n + m = 4$) this was tolerable because both the number of references and correlation vectors were reasonable. But, for $n + m = 7$ (e.g. Tc I, reported here), a user intervention approach is very inefficient. The Singular Value Decomposition (SVD) algorithm [6] was found to be both general and stable, once the proper choice of a reference was made (only ratios of coefficients are found). This change and completion of a data preparer for the reference functions input to REDUCE, dramatically improved user efficiency.

The downside of using REDUCE vectors (some accuracy, generally < 200 cm^{-1}, is also lost) is that each one uses the manifold's full set of determinants, so a lot of coefficients can be generated (several million in some cases). This increases the time needed to assemble the matrix element structure, which represents 70-80% of all RCI [7] CPU time. Some way of improving computational performance is needed. One idea is to use parallel processing, as construction of each matrix element group (fixed manifolds) can be done independently. We did such work in the mid-1990's [8].

Size is an easier problem to deal with. Our 500 MHz Alpha workstation is now configured with 1.5 GB of memory, and ~100 GB of disk space. This allows ~40% of the matrix elements of a energy matrix of order 20 000 to reside in memory, and about 50 million coefficients. Future increases can be obtained by rolling arrays in/out to disk.

It was possible to improve the efficiency of the f-value program dramatically for optical transitions. Previously, a calculation could take several hours per transition, to fully evaluate the non-orthonormality effects. This method [9] processes pairs of determinants one at a time. For optical electric dipole (E1) transitions, it is known that the final value scales as dE (Babuskin or length gauge) and as 1/dE (Coulomb or velocity gauge). By appropriately rescaling the results, a group (fixed J, J') of transitions may be run with a common energy difference (which appears in the spherical Bessel function), so that the determinantal processing occurs only once for the entire group.

We continued our development of algorithms which obtain properties of continuum states such as photoionization cross-sections, positions and widths of resonances, etc. These should be of utility to DOE-supported experimental studies such as those at the Advanced Photon Source. Tews and Perger have completed an algorithm to generate frozen-core relativistic continuum functions for complex atomic states, with proper normalization, core orthogonalization, and exchange, specific to the level of interest [10]. We are now developing an automated algorithm to evaluate N-electron matrix elements containing one continuum function, which includes the effects of between-state non-orthonormality [11].

Applications

Ta II Lifetimes

Last year we completed work on the Fe V d^4 J=0 $\rightarrow$ d^3 p J=1 transitions [12], and began work on $(d+s)^4$ J=1 $\rightarrow$ $(d+s)^3$ p J=1 transitions in Ta II, of interest to astrophysicists. In the case of Fe V, we were able to resolve some of the discrepancies between two prior calculations, as well as improve our understanding of what must be done to generate accurate f-values. Ta II is a more difficult case, due to interpenetration of d^3 p and d^2 s p levels, and only semi-empirical results were available [13]. Our ab initio calculations for lifetimes, hyperfine structure and Landé g-values of 24 (12) even (odd) parity levels are now complete [4]. We predict seven new lifetimes, all hyperfine structure, and 1 Landé g-value. Of the five measured lifetimes, we are in excellent agreement with two of them, and in good agreement with two others.

Tc I f-values

Investigation of Tc I f-values had been suggested to us by Linda Young [14] at last year's DOE workshop, as being of possible interest to ATTA studies [1]. As this involves transitions of the type $(d+s)^7 \rightarrow (d+s)^6$ p, it was also of considerable methodological interest, as it

would involve a dramatic increase in complexity, compared to the n + m < 5 cases studied earlier (e.g. see Table 1). For the higher n cases, we found a greater variation of the d radial between the $(d+s)^n$ p^m levels, as previously noted [15], which had to be properly accounted for.

Because of the complexity of the calculations, there was some sacrifice in accuracy for energy differences–average errors of 500 cm^{-1} were more typical, in place of 200 cm^{-1} errors, due to the omission of more core-valence effects. Sensitivity of atomic properties to errors was tested by shifting diagonal matrix elements to produce energy differences in agreement with experiment. In the aggregrate, structure files used ~35 GB of disk space, and the J=5/2 odd parity state took about 3 days of CPU time to produce. Reuse of structure and partial structure meant this cost was one time only.

We have identified the $4d^6$ 5s a $^6D_{9/2} \rightarrow 4d^6$ 5p z $^6F_{11/2}$ state as a potential ATTA transition. It has a high f-value (~0.3), the upper level decays strongly to a $^6D_{9/2}$, and a $^6D_{9/2}$ has a long lifetime, decaying by an E2 transition. The results are being written up, and will be submitted for publication shortly [16].

Future Plans

During the next year, we will complete the work on the continuum algorithms [11] and begin to apply them. We will complete a study of a rare earth ion of importance to the laser community, to see what correlation is essential to achieve good energy differences. Some earlier work we did on f^{13} and f^{14} [17] is proving helpful. We will also complete a study of the Yb II 6s → 6p transition, which requires careful correlation of the 4f electrons, due to the interpenetration of $4f^{14}6p$ and $4f^{13}5d6s$ levels.

Recent Publications Supported by DOE

Publications 4, 10, and 12; work on $3d^4$ W^{52+} and Bi^{61+} energy differences [18]; and lifetimes in Au^{66+} [19] were published in 1999–2001 and supported by DOE (Grant No. DE-FG02-92ER142828). Two Ph.D.'s (O'Malley and Norquist), and one M.S. (Tews) were also completed during this period.

TABLE I. Illustrating N-electron Basis Set Growth with Number of Electrons $(d+s)^2 \rightarrow f^2$ [a]

Label	# MCDF [b]	Full	Reduce [c]	RF [d]	Comments
$(d+s)^5$ J=5/2	29	333	111	3.0	Max J [e]
$(d+s)^6$ J=3	29	723	116	6.2	Near Max J
$(d+s)^7$ J=5/2	29	1068	121	8.8	Max J
$(d+s)^8$ J=3	15	1289	65	19.8	Near Max J
$(d+s)^3$ p J=1	40	83	no gain		Fe V, Ta II
$(d+s)^4$ p J=7/2	87	653	321	2.0	Near Max
$(d+s)^5$ p J=3	145	1963	558	3.5	Max
$(d+s)^6$ p J=7/2	155	4248	620	6.9	Tc I
$(d+s)^7$ p J=3	145	6382	602	10.6	Max J
$(d+s)^8$ p J=5/2	99	6891	424	16.3	Max

[a]Number of eigenvectors needed (fixed J_z=J) for the pair correlation where d/s electrons are replaced by two f electrons. Includes three manifolds.
[b]Number of eigenvectors (fixed J_z=J) in the d^m, $d^{m-1}s$ and $d^{m-2}s^2$ configurations. Most of

these would lie in the optical region.
[c]Number of surviving eigenvectors after REDUCE.
[d]RF = Full/Reduce.
[e]The J displayed is that for which (b) is maximum (or near max).

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Molecular Structure and Collisional Dissociation and Ionization

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Program Scope:

This program is aimed at investigating the molecular structure and the collisional dissociation and ionization of selected molecules and free radicals. The focus areas of the project are (1) ionization studies of selected molecules and free radicals and (2) the study of electron-impact induced neutral molecular dissociation processes. Targets of choice for ionization studies during the current funding cycle include SiF_4, $TiCl_4$, $SiCl_4$ and CCl_4 and its radicals, the molecular halogens Cl_2, Br_2, and F_2, $TiCl_x$ (x=1-3), and BCl_3 and its radicals. This choice is motivated on one hand by the relevance of these species in specific current and future etching applications (e.g. poly-Si and amorphous Si, Al and Al-Cu, and WSi_2 and $TiSi_2$ etching and the etching of thin magnetic films) and, on the other hand, by basic collision physics aspects ($SiCl_x$ and CCl_x are of similar molecular structure as the species studied previously and thus complement the existing data base for these targets; Br_2, F_2, and Cl_2 are simple diatomic molecules for which the collisional and spectroscopic data base is extremely scarce). The targets of choice for the neutral molecular dissociation studies include SiH_4, $Si(CH_3)_4$ (tetramethylsilane, TMS), SiF_4, and BCl_3. The fragments to be probed include $Si(^1S)$, $Si(^1D)$, $SiH(X\ ^2\Pi)$, $BCl(X\ ^1\Sigma)$, and $B(^2P^o)$.

The scientific objectives of the research program can be summarized as follows:

(1) provide the atomic and molecular data that are required in efforts to understand the properties of low-temperature processing plasmas on a microscopic scale

(2) identify the key species that determine the dominant plasma chemical reaction pathways

(3) measure cross sections and reaction rates for the formation of these key species and attempt to deduce scaling laws

(4) establish a broad collisional and spectroscopic data base which serves as input to modeling codes and CAD tools for the description and modeling of existing plasma processes and reactors and for the development and design of novel processes and reactors

(5) provide data that are necessary to develop novel plasma diagnostics tools and to analyze more quantitatively the data provided by existing diagnostics techniques

Recent Progress:

The main emphasis of the program up to now has been on ionizing electron collision processes and the target species have been chosen with two objectives in mind, (i) relevance to the physics and chemistry of low-temperature, non-equilibrium plasmas used in current technologies and (ii) relevance form a collision physics point of view (similar molecular and electronic structure, establishment of common trends and tendencies (or lack thereof), along iso-electronic or other sequences, families of species (i.e. hydrocarbons, fluorocarbons, etc.), and ability to develop scaling laws). The majority of targets studied so far fall into the matrix shown below (X = F, H or Cl), but other targets such as $TiCl_4$ and BCl_3 and their radicals have also been studied or will be considered as potential targets. 22 molecules and free radicals have so far been

investigated and a comprehensive data base of ionization cross sections and appearance energies has been compiled. Systematic trends, similarities as well as discrepancies in the data for targets of similar molecular and/or electronic structure have been established.

CX_4	CX_3	CX_2	CX
---	NX_3	NX_2	NX
SiX_4	SiX_3	SiX_2	SiX

The experimental efforts are complemented by calculations of ionization cross sections using semi-empirical and semi-classical approaches. Specific recent accomplishments:

Measurement of ionization cross sections for the $TiCl_x$ (x = 1 –3) free radicals

Ionization cross sections for the stable molecule $TiCl_4$ were measured earlier using a high-resolution double focusing mass spectrometer as well as a new time-of-flight mass spectrometer. We have now also measured the ionization properties of the $TiCl_x$ (x=1-3) free radicals. While we have good cross sections for the molecular fragment ions for each radical, the absolute calibration of the atomic Cl^+ and Ti^+ partial ionization cross sections is still affected by systematic uncertainties that must be investigated further before these cross sections can be published with the customary level of confidence. For $TiCl_3$, the $TiCl_2^+$ partial ionization cross section dominates with a peak value of about 4.3 x 10^{-16} cm^2 followed by the $TiCl^+$ cross section with a maximum of about 2.7 x 10^{-16} cm^2 and the parent $TiCl_3^+$ cross section with a peak value of 1.6 x 10^{-16} cm^2. The ionization of $TiCl_2$, on the other hand, is dominated by the $TiCl_2^+$ parent ionization cross section with a peak value of 3 x 10^{-16} cm^2 and a $TiCl^+$ fragment ionization cross section of 2 x 10^{-16} cm^2. Lastly, in the case of TiCl, we only have a reliable absolute cross section value for the parent $TiCl^+$ channel with a value of 1.8 x 10^{-16} cm^2.

Measurement of ionization cross sections for SiF_4

We completed the measurement of absolute total and partial dissociative ionization cross section measurements for silicon-tetrafluoride, SiF_4. We found that dissociative ionization channels are dominant. SiF_3^+ is the by far most dominant fragment ion with a maximum cross section of 4.5 x 10^{-16} cm^2 at 80 eV. The atomic fragment ions F^+ and Si^+ and the molecular SiF^+ fragment ion have comparable cross sections with maxima of 0.65 x 10^{-16} cm^2 (F^+), 0.5 x 10^{-16} cm^2 (Si^+), and 0.4 x 10^{-16} cm^2 in the energy range from 100 eV (SiF^+) to 160-200 eV (F^+ and Si^+). The SiF_2^+ fragment ions and several doubly-charged fragment ions have maximum cross sections below 0.1 x 10^{-16} cm^2. It is interesting to note that we found evidence of the presence of the metastable SiF_4^+ parent ion in our experiments. The determination of the appearance energies of all fragment ions indicates that all singly-charged molecular fragment ions are formed with little, if any excess kinetic energy, whereas the two singly-charged atomic fragment ions are formed with up to about 0.5 eV kinetic energy per ion.

Supporting semi-classical ionization cross section calculations

We have made significant progress in applying our semi-classical and semi-empirical ionization cross section calculation method to a variety of new singly-ionized molecular and radical targets and to highly ionized atomic targets.

To date, there have been few quantitative studies of the electron-impact neutral dissociation of molecules under controlled experimental conditions. This is mainly due to the difficulty in detecting neutral ground-state species with little or no internal energy. We have developed a triple-beam apparatus and use a combination of electron scattering techniques and laser-induced fluorescence (LIF) techniques to measure absolute cross sections for the electron-impact dissociation of molecules into neutral ground-state fragments. Prior to the use of LIF methods, a few non-optical approaches were applied which all have severe limitations in applicability and render the LIF approach due to its versatility the perhaps most promising techniques for these studies. The experimental arrangement consists of a crossed electron-beam - gas-beam set-up inside a stainless steel high-vacuum chamber (base pressure of 1×10^{-7} Torr). The two beams intersect at right angles. A tunable laser beam propagates either parallel or antiparallel to the electron beam in order to maximize the overlap of the three beams. Optical detection of the fluorescence from the interaction region is made perpendicular to both the electron beam and the gas beam. We now have absolute cross sections for the final-state-specific formation of $Si(^1S)$ atoms from SiH_4 for impact energies from 20 eV to 100 eV with a peak cross section of about 6×10^{-17} cm^2 at 60 eV.

Future Plans

As for the ionization studies, we are in the process of incorporating of a new detector system into the fast-beam apparatus. We have bought a new, commercially available dual-detector system from Roentdek GmbH in Germany. One fast position-sensitive MCP detector will replace the channeltron in the fast-beam apparatus and serve as the final product ion detector. The second detector will be installed as an alternative to the Faraday cup and will serve as a monitor of the fast neutral beam and the product ion beam prior to entering the electrostatic energy analyzer. This will allow us to obtain information about the excess kinetic energy of the fragment ions. In order to implement the detectors and to test their proper operation (which will be done in collaboration with Prof. Horst Schmidt-Böcking) we anticipate a 3-4 months down time of the fast-beam apparatus.

The main immediate emphasis is on the measurements of the electron-impact induced neutral molecular dissociation for the target molecules listed below together with the various atomic as well as molecular neutral fragments that will be probed.

Fragment	Parent Molecule	Transition	Pump Wavelength	Detection Wavelength
$BCl(X\ ^1\Sigma)$	BCl_3	$X\ ^1\Sigma \rightarrow A\ ^1\Pi^+$	272 nm	272 nm
$B(^2P^o)$	BCl	$(2p)\ ^2P^o \rightarrow (3s)\ ^2S$	250 nm	250 nm
$Si(^1S)$	SiH_4, SiF_4, $Si(CH_3)_4$	$(3p)^2\ ^1S \rightarrow (4s)\ ^1P$	391 nm	288 nm
$Si(^1D)$	SiH_4, SiF_4, $Si(CH_3)_4$	$(3p)^2\ ^1D \rightarrow (4s)\ ^1P$	288 nm	391 nm
$SiH(X\ ^2\Pi^+)$	SiH_4	$X\ ^2\Pi^+ \rightarrow A\ ^2\Delta$	415 nm	415 nm

Application of the proposed method to some of the above listed dissociation processes will require the implementation of a frequency doubler to pump the corresponding transitions (pump wavelengths below 350 nm).

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PROBING DYNAMICS AND STRUCTURE OF ATOMS, MOLECULES AND NEGATIVE IONS USING THE ADVANCED LIGHT SOURCE

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Program Scope

The objective of the research program is to probe the electronic structure and dynamics of atoms, molecules, and negative ions, and to advance fundamental understanding of their interaction with vuv/soft x-ray photons. The goal of the program is to understand, at the atomic and molecular level, detailed processes important to the understanding of the properties of complex materials. We use photons from the Advanced Light Source as our probes because they allow detailed knowledge and an impressive degree of precision. We have also developed and improved experimental techniques to achieve high detection efficiency and high precision measurements. We present here results completed and underway this past year and plans for the immediate future.

Recent Progress

1) Selective Resonant Inner-Shell Excitation and Dissociation in CO_2

Combining high-resolution spectroscopy with electron-ion coincidence measurements, we have been able to show the role of bending excitation in the dissociation of CO_2 after the C 1s $\rightarrow \pi_u^*$ resonant excitation (at 290.77 eV). The broad ($\approx$700 meV) C 1s$\rightarrow\pi_u^*$ resonance in the absorption spectrum of CO_2 has been decomposed into contributions from the Renner-Teller split core-excited states with bent and linear equilibrium geometries using resonant Auger spectroscopy. The C $1s^{-1}\pi_u^*$ excited state was found to decay primarily via participator Auger transitions to the A $^2\Pi_u$ state of CO_2^+. Analysis of the vibrational structure in the high-resolution Auger spectra, measured at several photon energies across the broad C 1s $\rightarrow \pi_u^*$ resonance, was accomplished using calculated Franck-Condon factors for the electronic excitation and de-excitation processes. Estimations of the geometries of the Renner-Teller split core-excited states were obtained from a comparison of the calculations with the resonant Auger spectra. Transitions to the bent core-excited state were found to contribute to the absorption profile exclusively at the photon energies below the maximum of the C 1s $\rightarrow \pi_u^*$ resonance, whereas the linear core-excited state become accessible at higher photon energies. The symmetric stretch vibrational progression of the linear core-excited state was identified and assigned. The minimum of the potential energy surface of the C 1s$\rightarrow\pi_u^*$ core-excited state at its linear configuration was estimated to be 290.4 eV above the ground vibrational level of the ground electronic state of CO_2. [1]. Using these high-resolution data with electron-ion coincidence measurements, we demonstrated that the predissociation of the dominating A state created through participator decay is not constant along the resonance profile. The interpretation is based on a preferential excitation of high levels of the bending mode at the low-photon energy side of the resonance, which favors the O^+ + CO decay channel.

[2]. We have also measured C 1s photoelectron spectra at higher photon energies (between 308-330- eV) with an instrumental resolution of about half the natural linewidth. The spectra have been analyzed to obtain vibrational spacings, vibrational intensities and the lifetime, τ, of the carbon 1s core-hole state, which we determine to be 99± 2meV, in disagreement with theoretical predictions of 66 meV . The calculations is based on a one-center model, which assumes that only the valence electrons localized on the atom with the core hole can participate in Auger de-excitation of the core hole. Our data however indicate that valence electrons on the oxygen atoms may play a role on the Auger decay of the carbon 1s hole in CO_2, and hence that a multicenter model may be necessary to describe the Auger process.[3]

2) Angle-resolved electron spectrometry studies in Li Atoms.

Precision measurements relating to the formation and autoionization processes of hollow Li atoms have been pursued due to the fact that this system allows the study of the simplest 4-body coulomb problem. The improvement in the detection system allowed us to push forward the spectral resolution in the measurement of singly, doubly and triply excited states as well as of Auger decay. These measurements allowed new understanding in both structure and dynamics of a light open-shell system, that nevertheless displays a complex behavior.

We have measured Li 1s photoelectron spectra, at several photon energies, in high electron and photon energy resolution, with resolved LS term structure of the Li^+ 1snl satellites transitions up to n=6. This study allowed us to resolve new final ionic states that could not be achieved in previous work. In particular, our measurement allowed for the first time, the 1s3s ^{3}S, 1s3s ^{1}S, 1s3p ^{3}P, 1s3d ^{1,3}D, and 1s3p ^{1}P photoelectron lines to be completely resolved, allowing a precise partition of the n=3 satellites into their individual components. The high-angular momentum satellite lines ($L \geq 2$) are found to contribute significantly to the 1snl satellite cross sections for n=3 and 4, and to become the dominant terms for $n \geq 5$. The high-angular momentum lines exhibit the same photon-energy dependence as the P lines, providing experimental evidence that continuum-continuum state coupling, equivalent to virtual electron collision processes, is responsible for the $L \geq 1$ terms in the satellite spectrum, in contrast to the electron relaxation (shake-up) mechanisms responsible for the S-terms.

Branching ratios and anisotropy parameters of individual lines, covering the 85-130 eV photon energy range, have also been compared with R-matrix calculations and with previous unresolved experimental works. The comparison with calculations shows that van der Hart [a] and our R-matrix calculations agree quite well with our measurements [4], unlike the configuration interaction based on overlap integrals of Armen et al.[b].

The total instrumental width of the 1s3s ^{3}S photoelectron line (FWHM=43 meV) results from the convolution of a 25 meV spectral resolution, a 25 meV width due to the electron spectrometer and a Doppler broadening of about 25 meV.

We have also, for the first time, measured the angle-resolved energy dependence of the electrons emitted over the energy range of the triply excited $2s^2 2p\ ^2$P Li resonance and have calculated the behavior of the angular distribution parameter β using the R matrix approximation. Experimental and theoretical results are in good agreement and show deep minima in the 1s2p ^{1,3}P ionic channels. We found that the energy at which the

minima occur does not coincide with the resonant energy, but is shifted towards higher energy. [5]
We also started experiments on doubly excited states and carried out measurements of electron spectra resulting from the Auger decay of the (2,3) doubly-excited states of Li^+ into the ground state of the Li^{2+} ion. The theoretical width of the 2s3s ^3S line is much lower than 1 meV, according to saddle-point calculations [c]. Thus, the observed width of 71 meV is entirely due to the spectrometer and Doppler broadening. This width is however significantly lower that the previous ultimate resolution obtained at the Super-Aco synchrotron radiation ring in Orsay, France. This data is presently being analyzed.

3) Double-Photoionization of Negative Ions.

We have finally started this year a new research project, conceptually planned eleven years ago, based on inner-shell photoionisation studies of negative ions. These systems provide a serious theoretical challenge because they are extremely sensitive to electron correlation and they provide a perfect testing ground to several different fundamental calculations. From an experimental point of view, inner-shell photodetachment is largely unexplored territory and most experiments have been on outer-shell studies using lasers. However, the availability of third generation light source has given us the opportunity to try the study of inner-shell photodetachment of negative ions. These experiments are very difficult since negative ions targets are very tenuous and one needs either huge photodetachment cross sections or a very intense photon source. We were motivated to undertake studies in this new area by recent theoretical work [d], untested experimentally, predicting large cross sections. We have installed this year a SNICS II negative ion source at the collinear photon-ion beam apparatus (located at the AMO center on the undulator photon beamline at the ALS) and have carried out successfully double-photodetachment experiments on Li^-. The experiment reveals dramatic stucture, differing substantially both qualitatively and quantitatively from the corresponding processes in neutral atoms and positive ions, as predicted by an enhanced R-matrix calculations [d]. The experimental/theoretical comparison shows good agreement over some of the photon energy range, and also reveals some puzzling discrepancies. Our work has been submitted to Phys. Rev. Lett.

4) Spin-and Angle Resolved Auger Spectroscopy.

In order to acquire new quantum mechanical information, such as dipole matrix elements and their relative phases, we added to our research program a new differential measurement dimension. In addition to measuring partial cross sections and angular distributions, we have now the ability to conduct spin resolved measurements on atoms and molecules allowing one to perform in some cases the "complete experiment". This type of study requires circularly polarized light available from the elliptical undulator of the ALS as well as an experimental system allowing electron spin-detection. We have built and tested successfully such a system based on three time-of-flight electron energy analyzers combined with retarding Mott polarimeters. The initial study has been applied to the photoionization case of the Xe 4d, which has been a show case subject of inner-

shell ionization for more than three decades and a benchmark experiment for theoretical models. The data is presently being analyzed.

Future Plans

The principal areas of investigation planned for the coming year are: (1) Continue our spin resolved studies in Xe and extend them to Ba and molecules, (2) continue our double photoionization/photodetachment studies in negative ions, (3) continue our high resolution measurements in atoms and molecules.

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Low-energy ion-surface and ion-molecule collisions

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Program Scope The focus of the present experimental research program is upon low-energy, ion-surface and ion- molecule collisions. In the case of the ion- surface studies, the goal has been to examine and understand the effects of adsorbates upon the secondary emission properties of both metallic and semiconductor substrates. Secondary emission is induced by positive and negative ion impact, the adsorbates studied thus far are oxygen and chlorine, and the substrates have included both crystalline and amorphous samples of Al, Mo, W, Ag and Si. The adsorbate coverage is indirectly determined and ranges from none up to a few monolayers. In the ion- molecule studies we have measured absolute cross sections for the collisional decomposition of SF_6^-. In both the ion- surface and ion- molecule collision studies, "low energy" implies collision energies ranging from a few up to 500 eV.

Recent Progress *Surface studies - Si(100) substrate.* The oxygen-chemisorbed silicon surface has been frequently explored owing to its connection to materials processing. The study of interactions of low-energy incident ions with these systems is related to understanding plasma interactions with the adsorbed silicon substrate, and sheds light on the nature of the chemisorption process as well. We have recently completed an investigation on the effects of an oxygen adsorbate on secondary electron and anion emission. This emission was effected with positive sodium ions, having an impact energy in the range of 50 to 500 eV. The use of low energy positive sodium ions as a surface probe precluded secondary electron emission via kinetic (momentum transfer) and potential routes, the former since the impact energies were below 500 eV and the latter since the ionization potential of sodium is less than twice the work function for most metals, a necessary condition for Auger neutralization, which leads to potential emission.

The absolute probabilities for secondary emission of electrons and anions are shown in Fig. 1 as functions of the impact energy; the relative

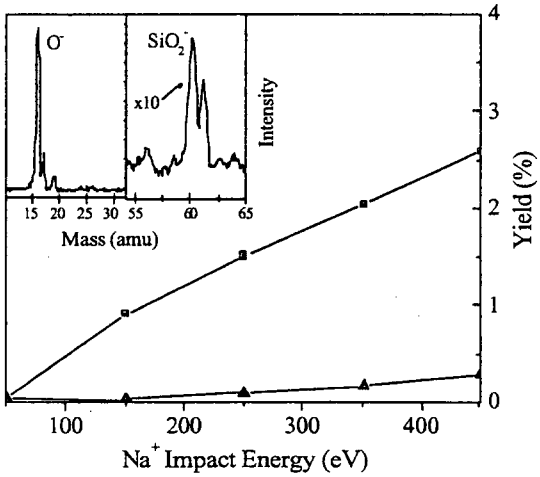

Figure 1 Secondary anion (squares) and electron (triangles) yields for Na^+ on Si(100) for an oxygen coverage of about 3/4 monolayer.

electron yield is seen to be quite small. In fact, the electron yield did not exceed 0.3% for any coverage or impact energy studied, in sharp contrast to what has been observed for metallic substrates. The anion spectrum arising from 250 eV Na^+ incident on the surface is shown in the inset of Fig. 1. The dominant peak is O^-, with a lesser contribution from OH^-; peaks at 60 and 61 amu, due to SiO_2^- and SiO_2H^-, were also present.

The kinetic energy spectra for both O^- and e^- are independent of the sodium impact energy and oxygen coverage suggesting that the dynamics descriptive of secondary emission involve electronic excitation and not collisional momentum transfer. Moreover electron kinetic energy spectra are very similar to those for the anions, suggesting that a common mechanism underlies emission of both. A model developed for ion-induced secondary emission from metallic substrates describes electronic excitation of a metal-adsorbate complex, e.g. MX^-, to a higher, repulsive potential, $(MX^-)^*$. This excitation can result in anion desorption into the vacuum, $(M + X^-)$, or decay of the system to yield a free electron (M + X + e or MX + e). Several factors complicate this picture when applied to an adsorbed semiconductor substrate, the most important of which is the presence of a band gap and fairly tightly bound electrons in the valence band. An electron transfer mechanism which can occur on insulator or semiconductor surfaces involves Auger de-excitation. Here, an electron from the valence band transfers to an unoccupied orbital of an excited species located near or on the surface, and an electron is simultaneously emitted to the vacuum from the excited state. This process is often referred to as "Penning ionization" in collisions of highly excited metastable atoms with atoms or surfaces, where secondary electron emission is routinely observed. In the present experiments, oxygen initially resides on the surface as O^-. We suggest that secondary anion and electron emission follows impact-induced excitation of SiO^-. A schematic diagram of this process is shown in Fig. 2, including the positions of Si(100) surface states and the energy levels of Si(100). The subsequent decay of $(SiO^-)^*$ may result in O^- being ejected into the vacuum intact, i.e., as O^-. The probability that O^- survives such a dissociation depends on how rapidly O^- leaves the surface and can be substantial for metallic substrates, as shown in previous experiments. Alternately, electron emission to the vacuum from $(SiO^-)^*$ could occur via a mechanism similar to the Penning ionization process if an electron from the silicon valence band transfers to fill the unoccupied orbital of (SiO^-). The rate for such a transfer process depends primarily on the overlap of the silicon valence band orbitals with those of the unoccupied states of (SiO^-). The relatively small probability for electron emission suggests that this orbital overlap is indeed small, perhaps due to a reduced spatial distribution of the surface wavefunctions. Furthermore, as $(O^-)^*$ recedes from the

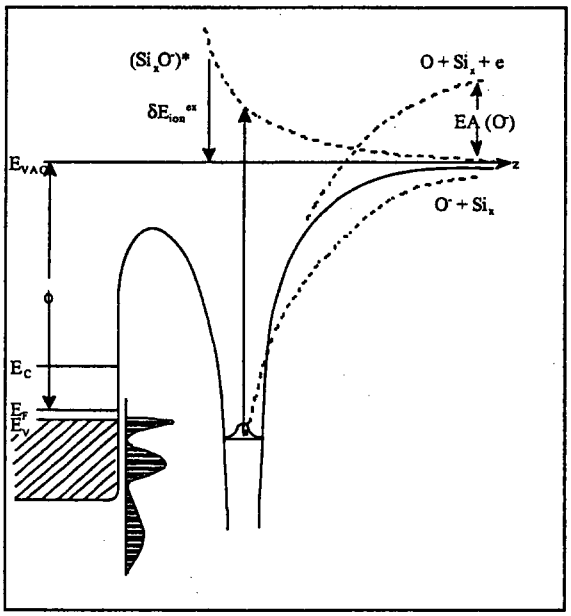

Figure 2 A schematic diagram illustrating the potentials descriptive of O^- and Si(100). E_C is the bottom of the conduction band, E_F the fermi level.

surface the energy level of the unoccupied orbital rises above the top of the valence band, and such autodetaching electron transfer will not occur. Secondary anions and electrons which originate from the decay of $(SiO^-)^*$ will have asymptotic kinetic energies (with respect to the vacuum) which depend on the initial position of $(SiO^-)^*$ on the antibonding state curve, as illustrated in Fig. 2. The energy distributions for e^- and O^- would then be similar; this is, in fact, observed. Finally, the way in which the total emission probability increases with impact energy is compatible with what is observed for binary collision electronic excitation mechanisms.

Surface studies - Rare gas ion projectiles. We have initiated a series of experiments which have incorporated a new ion source to study secondary emission processes from metallic substrates and the effects of adsorbates upon those processes. The first substrate/adsorbate investigated was Al/O and the results were unexpected. Fig. 3 illustrates the case for Ne^+ impacting a Al/O surface. The squares are the results for a clean surface and the circles (triangles) represent a coverage of about one-half (one) monolayer of oxygen on the Al substrate. The electron yield goes down as the coverage goes up, despite the fact that the macroscopic work function of the Al/O system *decreases* slightly with oxygen coverage in this range. Based upon previous experimental results where potential electron emission was precluded, one might expect that electron emission initiated by Ne^+ would simply expressed as a sum of the auger-type potential emission and the adsorbate-related process described above. Clearly the situation is much more complicated. On the other hand the secondary yield for O^- is very similar in all respects to that initiated by ions with very low ionization potentials.

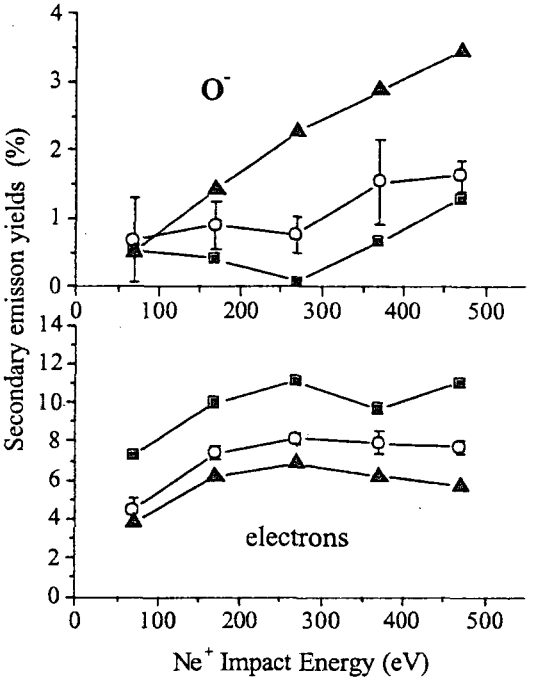

Figure 3 Secondary yields for Ne^+ on Al/O are shown for clean(■) Al; ~½ ml (○), and ~one ml oxygen (▲) on the substrate.

Collisional decomposition of SF_6^- .We have measured total cross sections for electron detachment and collision induced dissociation for collisions of SF_6^- with N_2 for collision energies ranging up to a few hundred eV. It has been suggested that N_2 / SF_6 mixtures might, in some applications, serve as replacements for pure SF_6. In order to improve the understanding of dielectric properties of N_2 / SF_6 mixtures, total cross sections for electron detachment and collision induced dissociation (CID) have been measured for $SF_6^- + N_2$ for relative collision energies $2 \lesssim E \lesssim 80$ eV. Within this energy range, cross sections for electron detachment are small, and CID is the dominant destruction mechanism for SF_6^-. These experimental results are remarkably similar to those obtained earlier for inert gas targets, suggesting that the processes are largely independent of target's structure. Hence, collisional decomposition has been modeled with a two-step mechanism in which collisional excitation of SF_6^- to SF_6^{-*} is followed by the latter's unimolecular decomposition. Unimolecular decomposition rates are based upon recent thermochemical data and a statistical phase space theory

The model results are in qualitative agreement with the experimental observations.

Future directions: During the coming contract period we will focus principally upon ion-induced secondary emission processes on surfaces. We will continue to examine the effect of adsorbates on these processes and will employ a host of impacting ions to probe the surface. These ions will include the rare gas ions; H^-, O^- and Cl^- anions; and such complicated molecular ions as CF_3^+, a popular etching species. Substrates will include both poly- and single- faceted metallic surfaces and Si(100).

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100 femtosecond X-ray detector

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I. PROGRAM SCOPE

Ultrafast X-ray source development has been undergoing rapid progress during the last decade. Whereas a lot of research has been devoted to the sources, very little effort has been spent towards the improvement of ultrafast X-ray detectors. Detectors with ~100 fs resolution will be very desirable because many chemical reactions and phase transitions are driven by the motion of atoms on the time scale of one vibration period (~100 fs). The streak camera is the fastest linear detector in the hard X-ray range. The goal of this project is to develop an X-ray streak camera with ~100 fs temporal resolution and timing accuracy.

Our development of the 100 fs X-ray streak camera has focused on two main issues:

a. Find new methods to improve the time resolution of the streak tube in order to improve the intrinsic resolution of the streak camera.

b. Implement novel techniques to decrease the synchronization jitter of the ramp voltage generator in order to improve the time resolution when the camera integrates signals over many shots.

This is a two-year project. Zenghu Chang, Bing Shan (CUOS fellow) and Adrian Cavalieri (graduate student), all from the Center for Ultrafast Optical Science (CUOS) at the University of Michigan, are the main contributors of the first year's achievements. In the second year, the grant will continue at Kansas State University. We have established collaborations with Roger Falcone (University of California Berkeley) and Phil Bucksbaum (University of Michigan) on time-resolved studies with 3rd generation sources (ALS and APS). We also have been working with Jin Wang (Argonne National Lab) on preparing a streak camera for the 4th generation source.

II. RECENT PROGRESS

a. Femtosecond X-ray Source Generated with OPA

We proposed to use ultrashort x-ray pulses from high harmonic generation as the light source for streak camera calibration. However, so far only soft x-rays (sub keV) can be produced with high harmonic generation. We experimentally demonstrated a powerful method that dramatically extended the harmonic cutoff. By using 1.5 μm pulses from an

optical parametric amplifier (OPA), we doubled the cutoff photon energies of xenon and argon atoms pumped with commonly used 0.8 μm lasers [1,2]. The results for Xe gas are shown in Fig.1. The experiments were done at an intensity of 10^{14} W/cm^2. We expect to produce 2 keV x-rays by exciting helium atoms with 1.5 μm pulses at the 10^{15} W/cm^2 intensity level.

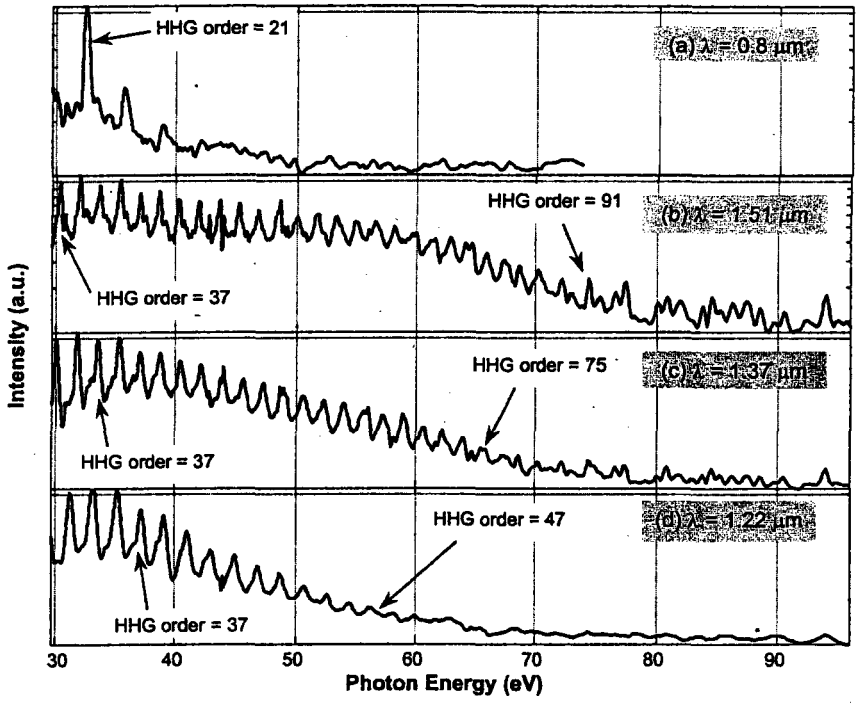

Fig. 1. High harmonic spectrum using different pump wavelengths but same pulse energy and focal point size

The new method of high harmonic generation will not only allows us to test the streak camera with keV x-rays, but also will provide an unique light source for time-resolved x-ray studies. By tuning the OPA between 1.2 to 1.5 μm, we can tune the harmonic wavelength to fill the gaps between two adjacent odd harmonics, which can be seen in Fig.1 [3]. This tuning method is much simpler than the methods demonstrated so far. This will have a big impact on experiments that require precise x-ray wavelengths such as resonate excitation of atoms and molecules.

b. Construction of a Newly Designed Streak Camera

To reach our 100 fs resolution goal, we proposed operating the streak tube in a UHV housing to increase the electric field in the cathode to anode region. To test our idea, a streak camera housing with $2x10^{-9}$ torr vacuum was built with which we can run the streak tube at 20 kV/mm. Under such a high field, the temporal resolution of the streak tube using KBr photocathode should be 140 fs. We are working on improving the vacuum and the cathode/anode assembly design to further increase the field [4].

To obtain 100 fs with our camera, we proposed driving the streak tube sweep plates with a 100 ps or faster photoconductive switch. We developed a new photoconductive switch that has rise time shorter than 100 ps, which meets our requirements. New deflection plates with a 150 ps response time were developed recently, which need further improvements to have a 100 ps response time.

c. Time Resolved X-ray studies with Streak Camera

We have continued our collaboration with the team lead of Prof. Roger Falcone at University of California, Berkeley. A streak camera was set up on a dedicated femtosecond x-ray beam line at the Advanced Light Source. Recently works done with the streak camera include the coherent control of acoustic phonons in an SbIn crystal and a time resolved absorption spectroscopy study of warm-state matter produced by femtosecond laser pulses [5,6,7].

We started a new collaboration with Phil Bucksbaum's group at the University of Michigan who has built an ultrafast setup at the Advanced Photon Source, Argonne National Laboratory. Recently, X-rays that transmit through several types of thin wafers were measured with our streak camera.

III. FUTURE PLANS

a. Femtosecond, Tunable, keV X-ray Source

We plan to extend our high harmonic generation source to keV photon energies. The PI of this project, Zenghu Chang, derived a formula to calculate the cutoff photon energy of high harmonic generation:

$$h\nu_{cutoff} = I_p + \frac{0.5 I_p^{3+a} \lambda^2}{\left[\ln \frac{0.86 \Delta t 3^{2n^*-1} G_{lm} C_{n^*l^*}^2 I_p}{-\ln(1-p)}\right]^2} \qquad (1)$$

where I_p is the ionization potential of the atom/ion, λ is the wavelength of the laser, and Δt is the pulse duration of the laser. G_{lm} and $C_{n^*l^*}$ are determined by the quantum number of the electron. P is the ionization probability.

According to Eq. (1), a ~2 keV HHG can be obtained by using a 1.6 μm, 25 fs driving field with helium gas, which is much higher than the highest harmonics (0.5 keV) generated so far by the 0.8 μm laser. Work is in progress to increase the OPA intensity further so that the expected keV HHG emission can be generated by driving helium gas with OPA at 10^{15} W/cm^2. The high intensity OPA is under development at the J.R. Macdonald Laboratory at Kansas State University. The X-ray will be measured with a spectrometer on loan from Lawrence Berkeley Laboratory.

b. Streak Camera Testing

The new camera will be assembled at Kansas State University and will be tested using the ultrafast laser and x-ray source under construction there. We expect to get sub-picosecond resolution in the accumulation mode by the end of 2001, so that it can be used on the ALS beamline for experiments that desperately need high time resolution. In 2002, we will continue our efforts to improve the camera and debug problems that may surface during the applications.

c. Applications

We plan to implement the new camera on the ALS femosecond X-ray Beamline in 2002. Another camera will be built for the 4th generation source development at the APS. That camera may be shared with X-ray experiments using the current 3rd APS source.

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Multiphoton Quantum Dynamics and Optimal Generation of Coherent X-Ray Harmonic Emission

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Program Scope

In this research program, we address the fundamental physics of the interaction of atoms and molecules with intense ultrashort laser fields. The main objectives are to develop new theoretical formalisms and accurate computational methods for *ab initio* comprehensive investigations of multiphoton quantum dynamics and very high-order nonlinear optical phenomena of one- and multi-electron quantum systems in intense and superintense laser fields, taking into account detailed atomic and molecular structures. Particular attention will be paid to the exploration of novel physical mechanisms, time-frequency spectrum, and coherent control of multiple high-harmonic generation (HHG) processes for the development of table-top x-ray laser light sources. Also to be investigated is the fundamental AMO theory on the interactions of ultrafast, intense x-ray pulses with atomic and molecular systems, including multiphoton and high-field effects.

Recent Progress

DOE BES support of this program began at Sept. 15, 2000. Our major accomplishments in the past year are summarized below.

1. Spectral and Temporal Structures of High-Order Harmonic Generation of Na Atoms in Intense Mid-IR Laser Fields

The recent advancement of mid-IR laser technology [1] opens the possibility of studying multiphoton processes in systems with lower binding energies (such as alkali atoms) and allows the exploration of fundamentally different strong-field phenomena at longer wavelengths. Recently it has been also suggested that an intense mid-IR laser light source may be used to generate HHG in the visible to UV regime, allowing the application of frequency resolved optical grating [2] for the full characterization of the harmonic's amplitude and phase. Motivated by such recent advances, we have performed a 3D quantum study of HHG of Na atoms in mid-IR laser fields [3].

To describe the electronic structure of Na atom, we have constructed an accurate angular momentum-dependent model potential. The valence-electron -core interaction is chosen to have the correct long-range polarization terms, and the parameters that determine the short-range part of the potential are constructed so that both the energy levels and experimental spectra of low-energy electron scattering phase shifts are reproduced to very high accuracy for all angular-momentum symmetries. The time-dependent Schrodinger equation of Na in laser fields is solved by means of the *time-dependent generalized pseudospectral* (TDGPS) method recently developed [4]. The procedure allows *nonuniform* and optimal spatial discretization and accuracy and efficient time propagation of the wavefunction in space and time. Excellent agreement of the HHG power spectrum in the *length* and *acceleration* forms is obtained from the lowest harmonic to the cutoff. The HHG power spectrum shows fine structure and significant enhancement of intensities of the lower harmonics due to the strong coupling of the 3s-np states and the 3s-3p multiphoton resonance. We use a wavelet transform to

perform a detailed time-frequency analysis for the whole range of HHG power spectrum. The results reveal unexpected details of the spectral and temporal fine structures of individual harmonic, providing insights regarding different HHG mechanisms in different frequency regime of Na atoms at longer wavelengths [3].

2. Development of Self-Interaction-Free Time-Dependent Density Functional Theory for Nonperturbative Treatment of Many-Electron Quantum Systems in Intense Laser Fields

To study strong-field processes of many-electron systems using the *ab initio* wavefunction approach, it is necessary to solve the time-dependent Schrödinger equation of 3N spatial dimensions in space and time (N = the number of electrons), which is beyond the capability of current supercomputer technology. The *single-active-electron* (SAE) model with *frozen core* is commonly used and has been successful for some strong-field processes where only one valence electron plays the dominant role. However, within the SAE model, important physical phenomena such as excited state resonances, different dynamical response from different valence spin-orbitals, inner core excitation, and the dynamical electron correlations cannot be treated. Clearly, a more complete formalism beyond the SAE model is very desirable for further progress in the study of the atomic and molecular physics in strong fields. Recently we have initiated a series of new developments in *self-interaction-free time-dependent density functional theory* (TDDFT) for probing strong-field processes of many-electron atomic systems, taking into account electron correlations and detailed electronic structure. More recently, the theory has been also extended to the two-center diatomic molecular systems as well.

The traditional DFT, widely used in quantum chemistry and condensed matter electronic structure calculations [5], is largely limited to the study of the *ground-state* properties of atoms, molecules, and solids. Due to the approximations used in the exchange-correlation (xc) energy functionals, conventional DFT calculations contain a spurious *self-interaction energy*, and the xc potentials so generated do not have the proper *long-range* Coulomb (-1/r) potential behavior. As a result, while the total electronic energies of the ground states are rather accurate, the excited and resonance states, as well as the ionization potentials (obtained from the highest occupied orbitals) are not satisfactory. Typically the ionization potentials are 30-50% too low. The goal of the recent development of DFT with *optimized effective potential* (OEP) and *self-interaction-correction* (SIC) is to overcome some of these fundamental difficulties encountered in conventional DFT electronic structure calculations [6,7]. The DFT/OEP-SIC procedure uses only *orbital-independent* single-particle *local* potentials that are self-interaction *free* and have the proper short- and long-range- potential behaviors. It is shown that the method is capable of providing reasonably high accuracy for the excited states and autoionizing resonances [6], as well as for the ionization potentials (well within a few percents of the experimental values) across the periodic table [7]. The DFT/OEP-SIC formalism has been recently extended to the *time* domain for atomic [8,9] and molecular [10] systems. Given below is a brief summary of the progress in the last year.

2a) Multiphoton Ionization and High-Order Harmonic Generation of Rare Gas Atoms in Intense Laser Pulsed Fields

We perform a detailed *all-electron* study of multiphoton ionization (MPI) and high-order harmonic generation (HHG) processes of rare gas atoms (He, Ne, and Ar) in intense pulsed laser fields [9] by means of the *self-interaction-free* time-dependent density functional theory (TDDFT) recently developed. The time-dependent exchange-correlation (xc) potential with proper short- and long- range potential is constructed by means of the *time-dependent optimized effective potential* (TDOEP) method and the incorporation of an explicit *self-interaction-correction* (SIC) term. The TDOEP-SIC equations are solved accurately and efficiently by the use of the *time-dependent generalized pseudospectral* technique [4]. In this study, all the valence

electrons are treated explicitly and nonperturbatively and their partial contributions to the ionization and HHG are analyzed. The results reveal instructive and qualitatively different behavior from each subshell orbital. Moreover, we found that the HHG yields from Ne and Ar atoms are considerably larger than that of the He atom in strong fields. Two main factors are identified for accounting the observed phenomena: (a) the binding energy of the subshell valence electron, and (b) the orientation of the valence electron orbital (with respect to the electric field polarization). In particular, we found that the np_0 valence electrons (in Ne and Ar), with lowest binding energies and electron orbital orientation parallel to the electric field direction, make the dominant contributions to both ionization and HHG processes in sufficiently strong fields [9].

2b) High-Order Harmonic Generation of H_2 in Intense Laser Fields

We develop a new and general *self-interaction-free* TDDFT for nonperturbative and comprehensive treatment of multiphoton processes of *multi*-electron *molecular* systems in intense laser fields [10]. The resulting TDDFT equations are structurally similar to the time-dependent Hartree-Fock equations, but include the many-body (electron-correlated) effects through an orbital-independent single-particle *local* time-dependent exchange-correlation (xc) potential. The latter is constructed by means of the time-dependent OEP/SIC method. A numerical time-propagation technique is introduced for accurate and efficient solution of the TDDFT/OEP-SIC equations for two-center diatomic molecular systems. The procedure involves the use of a *generalized pseudospectral method* for *nonuniform* optimal grid discretization of the Hamiltonian in prolate spheroidal coordinates [11] and a split-operator scheme in the *energy* representation for the time development of individual electron orbital wavefunctions. High-precision time-dependent wavefunctions can be obtained by this procedure with the use of only a modest number of grid points. The theory is applied to the first *all-electron* study of HHG processes of H_2 molecules in intense pulsed laser fields. Particular attention is paid to the exploration of the spectral and temporal structures of HHG by means of the wavelet time-frequency analysis. The results reveal striking details of the fine structures (sub-peaks) of the time profile of individual harmonic, providing new insights regarding the underlying HHG mechanisms in different energy regimes, including low-lying multiphoton dominant regime, near ionization-threshold regime, plateau regime, and near cut-off regime, for a molecular system for the first time [10].

2c) Exact Relations of the Quasienergy Functional and the Time-Dependent Exchange-Correlation Potential

In the steady-state DFT study, one major unsolved problem is the lack of the exact and universal form of the exchange-correlation energy functionals and potentials. Thus approximate xc energy functionals must be used. While there is considerable progress in the refinement of the approximate form of the steady-state xc energy functionals in the past decade [5], the study of the corresponding *time-dependent* xc energy functional forms is still at its infancy and remains a largely unexplored area of fundamental research. To advance this field, we have recently initiated a study of the *exact* relations of quasienergy functionals [12] based on the extension of the *generalized Floquet formulation* of TDDFT recently developed [13]. Several exact relations involving different parts of the quasienergy functionals are obtained. These relations hold when the exact densities and xc energy functionals are employed. They can serve as the constraints when searching for the approximate forms of the time-dependent xc functionals and potentials in the future. The general results are illustrated on an exactly soluble model, two-electron Hook's atoms in a linearly polarized monochromatic laser field [12].

Future Research Plans

In addition to continuing the ongoing researches discussed above, we plan to also initiate the following several new projects: (a) Extension of the self-interaction-free TDDFT to the study of MPI and HHG of more complex molecular systems (such as N_2). The goal is to explore the nonlinear response of individual molecular orbital to intense fields, a subject of largely unexplored area of strong-field molecular physics. (b) *Ab initio* quantum mechanical study of the coherent control of HHG in intense laser fields by means of the *genetic algorithm* optimization of the *"intra-atomic"* dynamical phase matching on the sub-optical cycle and the wavelet time-frequency analysis of quantum dipole emission. This research is prompted by the recent JILA experiment on the attosecond time-scale feedback control of coherent x-ray generation by optimizing the shape of an ultrafast laser pulse [14]. (c) Development of *quantum fluid dynamics* approach to the treatment of multiphoton dynamics and HHG processes of many-electron quantum systems in intense laser fields.

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Theoretical Investigations of Atomic Collision Physics

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A diverse array of atomic and molecular collision phenomena has been studied theoretically. Of particular interest has been the behavior of atomic and molecular collisions at ultracold temperatures.

Calculations of the quenching of vibrationally excited molecular hydrogen in collisions with H and ^{4}He have been extended to collisions of ^{4}He with O_2 and CO and of ^{3}He with CO. Differences in the quenching rate coefficients of CO colliding with ^{4}He and ^{3}He become much more apparent at very low temperatures where the quenching is affected by resonances trapped in the long range van der Waals well.

A substantial effort has been put into developing numerical codes that can solve the close-coupling equations for reactive collisions at low energies. The method we used is an extension of the so-called ABC quantum reactive scattering program of Skouters, Castillo and Manopoulos (Comp. Phys. Comm. **133**, 128, 2000) which employes Delves hyperspherical coordinates for all the rearrangement channels. We obtained results for the reaction

$$F + H_2 \rightarrow FH + H$$

and determined the rate coefficients at near zero temperature for the open rovibrational states of the product molecule FH. For the total reactive rate coefficient at zero temperature, we found a value of 5×10^{-13} cm^3 s^{-1}. The preferred product is the v = 2 vibrational level. It appears that the long duration of the collision allows tunneling through the repulsive barrier even in the limit of zero velocity.

The accelerated cooling of hydrogen by mixing it with an alkali metal atom was explored by determining the scattering lengths and it was found that mixing with Na might be particularly effective.

Investigations were carried out of the collisions of hydrogen and antihydrogen atoms at low energies using a version of the distorted wave approximation. Rearrangement collisions resulted in the formation of protonium in a highly excited state and positronium. The possibility of cooling antihydrogen by colliding with cold hydrogen was explored. The quasi-molecule formed by the approach of H and $\overline{\text{H}}$ has a dipole moment which may lead to specific radiation and provide a unique diagnostic of the presence of $\overline{\text{H}}$. Collaborators in the study of antihydrogen are given in the publication list.

The quenching of metastable hydrogen atoms in collisions at thermal energies was investigated and it was concluded that the dominant quenching mechanism is not Penning ionization, though it does occur, but double excitation transfer to a pair of excited $2p$ atoms which then radiate Lyman alpha photons. The situation may differ at ultralow temperatures where the Lamb shift and fine-structure splitting must be included in the energetics.

In connection with the various scattering problems we have applied what are by now standard methods to compute the van der Waals coefficients for various systems of experimental interest.

In addition, Dr. A. Derevianko carried out many body perturbations calculations of the electric dipole amplitude of transitions between low-lying levels of Mg, Ca, Sr and Rb and presented an analysis of parity-nonconservation in Cs that resolved a discrepancy between the measurements and calculations using the standard model.

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Femtosecond photon echo techniques for manipulation of quantum states and computation

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1. Introduction

Our DOE supported project is aimed at exploring the coherent manipulation of multiple quantum mechanical states using three-pulse photon echo techniques. The originally proposed experiments were aimed at writing information in the form of phase and amplitude in multiple vibrational states of molecular iodine and reading that information using spectrally dispersed and time-resolved detection. These experiments depend on the interaction of the sample with three laser pulses and the detection of the coherent emission resulting from the four-wave mixing process. We now have more ambitious goals. We hope to use three-pulse four-wave mixing with cooled molecules for quantum computation. Femtosecond amplitude and phase shaped pulses encode information into a coherent superposition of vibrational states. The coherent coupling between the quantum states and the consecutive interactions with shaped pulses is used as quantum logic gates to create the final superposition of states. The resulting coherent virtual or stimulated photon echo signal corresponds to a vector or a matrix output, respectively. The concept, experimental setup, and preliminary results are presented.

The main requirement for a quantum computer is a quantum set of states that can be addressed coherently and controlled without the loss of coherence [1,2]. We propose to use three-pulse four-wave mixing (FWM), in particular the stimulated photon echo (SPE) and virtual echo (VE) pulse sequences, to prepare and control coherent ensembles of vibrational states [3]. The transformations are achieved with femtosecond amplitude and phase shaped laser pulses. The proposed experiments involve the coherent manipulation of vibrational states in molecular iodine. Recently many groups have shown various modes of FWM signal from this sample [3-7]. Zadoyan et al. recently considered the manipulation of a ro-vibronic superposition of states using time-frequency-resolved coherent anti-Stokes Raman scattering for quantum computing [8]. Our approach does not depend on the internal dynamics (as proposed in [8]) and instead uses amplitude and phase shaped pulses to control the transition probabilities between ground and excited states.

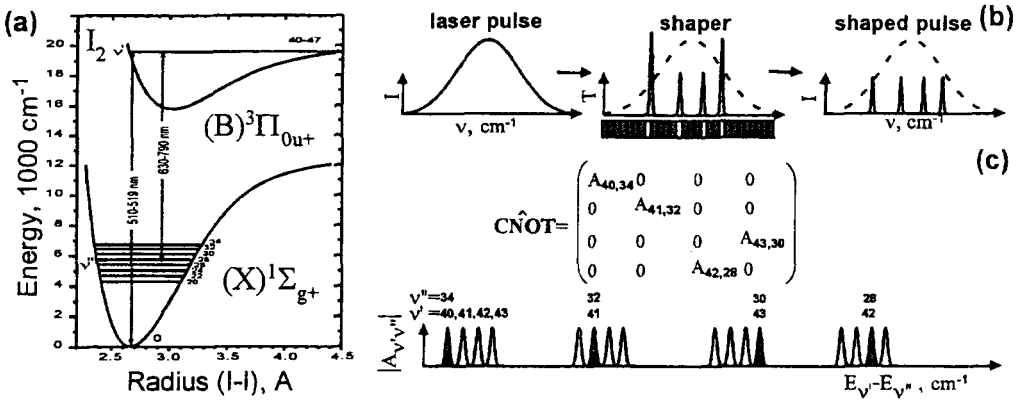


Fig. 1. (a) Energy curves, vibrational levels of I_2 and optical transitions for a 3-qubit processor. (b) The initial laser pulse passes through a shaper and is transformed into coherent light consisting of several narrow spectral components. (c) Electric field components responsible for the CNOT quantum gate for a 2-qubit operator.

The main idea is to save information as quantum amplitudes on selected eigenstates (Fig.1a) and to manipulate the information with shaped coherent light pulses (Fig.1b). The shaped pulses contain specific spectral components that address individual v'-v" transitions. To construct a controlled NOT quantum gate, for example, we use spectral components for the transitions $40 \rightarrow 34$, $41 \rightarrow 32$, $42 \rightarrow 28$, $43 \rightarrow 30$. Each specific vibrational eigenstate is a component of a vector in Hilbert space and the manipulation with complex amplitudes of these vector components allows us to perform quantum computations.

Here we present two different arrangements that provide the foundation of a more general setup for vectorial computation. Signal due to the R_2 and R_3 Liouville pathways is known as SPE, whereas signal from R_1 and R_4 is known as VE [9]. The first pulse spans the wavelength range 510-520 nm, where the absorption cross-section from the ground state is maximal. The second (Stokes) pulse is tuned to a wavelength (640-780 nm) where no further B

state excitation can be achieved from the ground state (v'' = 0). These two responses are separated by their different phase matching conditions [3-5].

2. Theory

The first shaped *pump* pulse creates a coherent superposition of vibrational states in the excited state. The complex amplitude of each excited vibrational state is defined by the spectrum of the electric field. The *Stokes* pulse promotes transitions to the coherent superposition of vibrational states in the ground electronic state. If the wavelengths in the *probe* pulse are equal to those in the *Stokes* pulse, then the *probe* pulse promotes transitions from the ground state to the exited state (R_4). If the wavelengths in the *probe* pulse are equal to those in the *pump* pulse, then the probe pulse promotes transitions from the initial ground state to the excited state (R_3).

The VE signal is formed by the coherence between the initial ground state and the third order wave packet in the excited state. The SPE signal is formed by the coherence between the second order wave packet in the ground state and the first order wave packet in the excited state. VE and SPE emissions have phase matching directions $\mathbf{k}_3$-$\mathbf{k}_2$+$\mathbf{k}_1$ and $\mathbf{k}_3$+$\mathbf{k}_2$-$\mathbf{k}_1$, respectively. The third order polarization and resulting emitted light are given by

$$P^{(VE)} \propto \langle \Psi^{(0)} | \mu | \Psi^{(3)}(\tau_{12},\tau_{13}) \rangle \text{ and } P^{(SPE)} \propto \langle \Psi^{(2)}(\tau_{12}) | \mu | \Psi^{(1)}(\tau_{13}) \rangle, \quad (1)$$

where

$$|\Psi^{(1)}\rangle \propto \sum_{i'} a_{i'} |i'\rangle; \quad |\Psi^{(2)}(\tau_{12})\rangle \propto \sum_{jj'} M_{jj'} |j\rangle; \quad |\Psi^{(3)}(\tau_{12},\tau_{23})\rangle \propto \sum_{ijj'} Q_{ij} M_{jj'} |i\rangle; \quad |\Psi^{(1)}(\tau_{13})\rangle \propto \sum_{i} b_i |i\rangle. \quad (2)$$

Based on Equation 1 and 2 the signal for VE is a *vector* $b_i \propto Q_{ij} M_{jj'} a_{i'}$ and the signal for SPE is a *matrix* $O_{ij} \propto (M_{jj'} a_{i'})^{\dagger} b_i$. This approach is applicable for weak fields; the strong-response has been analyzed in [10].

3. Experiment

Ultrafast pulses are generated using non-collinear optical parametric amplifiers. The femtosecond pulses are shaped using spatial light modulators. The information carried by the emitted light has to be spectrally dispersed and heterodyne (phase sensitive) detected. The experimental implementation is shown as a schematic in Fig. 2a,b.

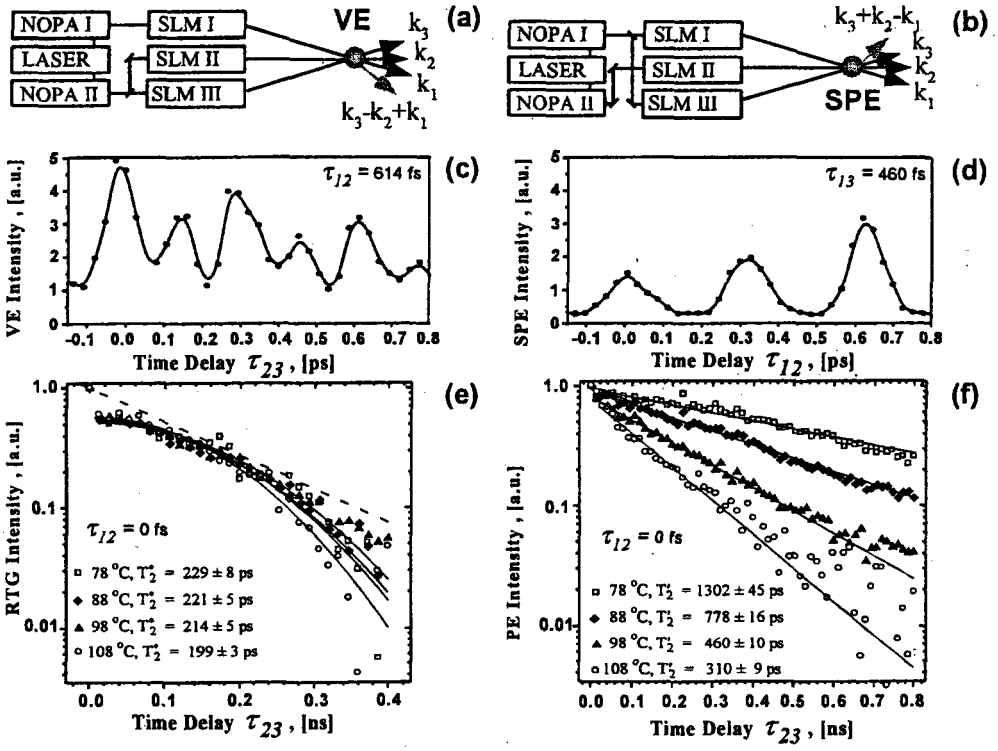


Fig. 2. Experimental setup for VE (a) and for SPE (b) types of computing. The NOPAs are pumped by a femtosecond *Ti:sapphire amplified laser. Shaped pulses are obtained using spatial light* modulators (SLM). The homodyne-detected intensities of VE (c) and SPE (d) as function of time delay time between the pulses are presented. Inhomogeneous (e) and homogeneous (f) coherence relaxation measurements.

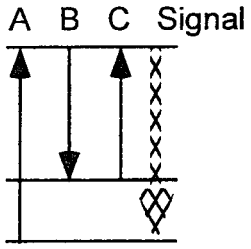
The data on figure 2 was obtained with a simplified setup: room temperature I_2, degenerate transform limited pulses, and homodyne detection to demonstrate the feasibility of the experiments [4]. The experimental data

presented in Fig.2c corresponds to the VE setup. Because the three beams are the same wavelength (620 nm), the timing of the first two pulses is used to determine which pathway leads to signal formation [3]. The data obtained for $\tau_{12} = 614$ fs is the data that is relevant for the R_4 response. The signal shows the time evolution of the ground state coherence prepared after the first two pulses. The experimental data presented in Fig. 3d corresponds to the SPE setup. Here $\tau_{13} = 460$ fs, the time delay between the first and last pulses, is set to cancel the contributions from the R_2 response [3]. It is clear that every 307 fs, which is the period of oscillation of the wave packet in the excited state, the signal achieves a maximum value. All laser pulses have their phases set to zero, so no specific quantum information as been introduced yet. Here we show the coherent manipulations of ground and excited state wave packets and the control over the desired responses. The data in Figs. 2e and 2f show the electronic coherence decay. In Fig. 2e inhomogeneous broadening reduces the coherence time to about a hundred picoseconds; however, the data in Fig. 2f, shows the homogeneous decoherence of the sample to be longer than a nanosecond. The quantum computation scheme we envision requires that all laser pulse interactions occur with minimal loss of coherence. With supersonic jet cooling, the homogeneous dephasing time can be extended to hundreds of nanoseconds. The ratio between the time for quantum computation (10^{-11} s) and coherence lifetime (10^{-7} s) is approximately 10^4, this implies minimal loss of coherence.

Recently we have implemented the VE setup to demonstrate the feasibility of the proposed method. The experimental data are shown in Fig. 3. The data were obtained with a *pump* beam at 585 nm; a shaped *stokes* beam centered at 800 nm and a *probe* beam at 800 nm. The phase-matched signal emerged centered near 590 nm and was clearly modulated by the rotationally broadened vibronic transitions. Although the sample was held near 470 K, the individual transitions can already be discerned. Jet cooling, however, will sharpen considerably the spectral resolution.

Fig. 3

Experimental Data Obtained with the VIrtual Echo Setup



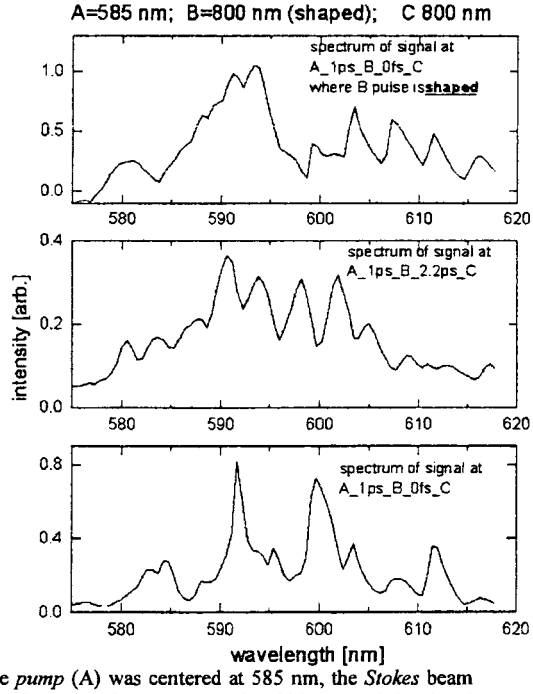


Fig. 3. Experimental data for a VE setup. The *pump* (A) was centered at 585 nm, the *Stokes* beam (B) was shaped and centered at 800 nm, the *probe* beam (C) was centered at 800 nm, and the signal was centered near 590 nm. The signal beam was frequency dispersed. Three different spectra resulting from different conditions are shown. The variables for each spectrum are indicated in the upper right corner. Clearly the time between the pulses and shaping of the *Stokes* beams changes the outcome in the observed signal.

4. Conclusions and Future Work

From an experimental point of view the proposed scheme is analogous to NMR based quantum computation, which is the most successful demonstration to date [11]. As pointed out by Warren, the NMR quantum computer has two problems [12]. The first is low clock frequency, approximately 10-100 Hz. In our case, using optical frequencies, the clock rates are on the order of 10^{11}-10^{12}Hz. The second is that NMR operates in the high temperature limit. The NMR ratio between the photon energy and temperature is $h\nu/k_BT<10^{-6}$ for a 10 spin system. Echo sequences,

analogous to the multiple pulse NMR, operate in the low temperature limit $h\nu/k_BT>10^3$. This difference makes the optical experiments much less susceptible to quantum noise.

From a mathematical point of view, the proposed VE and SPE processes can perform the following functions: write, store in memory, read, sum, scalar product, direct products, matrix product between matrices and vectors, and can be used to construct circuits and networks. The large Hilbert space and the freedom to construct complex quantum gates give us the possibility to operate with massive coherent quantum information even within a single three-pulse sequence.

We are very encouraged by our first data for a number of reasons. Most importantly, the signal level is relatively strong and can be separated easily from background scattered light. We expect the arrival of a second NOPA this month that will allow us to design background free experiments. We are looking forward to the purchase of two additional pulse shapers and the adaptation of our supersonic jet machine to finalize the full implementation. We hope to secure the funds for the additional equipment in the near future.

Our funding period started April 2001. We do not have publications of research supported by DOE in print yet but we have already submitted four publications. A post-deadline paper on this research was presented at the first International Conference on Quantum Computation, ICQI, Rochester NY, June 2001.

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High Intensity Laser Interactions with Atomic Clusters

Progress report (Fall 2001)

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Program Scope:

The development of ultrashort pulse table top lasers with peak pulse powers in excess of 1 TW has permitted an access to studies of matter subject to unprecedented light intensities. Such interactions have accessed exotic regimes of multiphoton atomic and high energy-density plasma physics. Very recently, the nature of the interactions between these very high intensity laser pulses and atomic clusters of a few hundred to a few thousand atoms has come under study. Such studies have found some rather unexpected results, including the striking finding that these interactions appear to be more energetic than interactions with either single atoms or solid density plasmas. Recent experiments have shown that the explosion of such clusters upon intense irradiation can expel ions from the cluster with energies from a few keV to nearly 1 MeV. This phenomenon has recently been exploited to produce DD fusion neutrons in a gas of exploding deuterium clusters. Under this project, we have undertaken a general study of the intense femtosecond laser cluster interaction. Our goal is to understand the macroscopic and microscopic coupling between the laser and the clusters with the aim of optimizing high flux fusion neutron production from the exploding deuterium clusters or the x-ray yield in the hot plasmas that are produced in this interaction. In particular, we are studying the physics governing the cluster explosions. The interplay between a traditional Coulomb explosion description of the cluster disassembly and a plasma-like hydrodynamic explosion is not entirely understood, particularly for small to medium sized clusters (<1000 atoms) and clusters composed of low-Z atoms. We are focusing on experimental studies of the ion and electron energies resulting from such explosions through various experimental techniques. We are also examining how an intense laser pulse propagates through a dense medium containing these clusters.

Recent Progress

During much of this previous year, I was involved in moving my research effort from LLNL to the University of Texas. At UT we are now constructing a 20 TW, 30 fs Ti:sapphire laser which will be used to drive the cluster explosions. We intend to reinitiate our experimental program this fall.

Much of the research undertaken previously under this project has been focused on the interaction of a intense 30 fs pulses with deuterium clusters. These initial experiments utilized the 5 TW Falcon laser at LLNL. This work is motivated by the recent observation of DD fusion in a gas of laser

irradiated deuterium clusters. One of the principal goals of this project is to understand the explosion mechanisms of these clusters so that they can be manipulated to enhance the fusion yield. We have also undertaken studies of third harmonic generation from these intense cluster interactions to gain understanding of the electron oscillation dynamics during the interactions. Finally, we have examined hot electron generation from quite large clusters to gain information about how the nature of the interaction evolves from the sub wavelength scale clusters to clusters of size approaching the laser wavelength (a size scale which may also, ultimately, be interesting for fusion research).

1) Interactions with deuterium clusters

During late 1999 and through 2000, we conducted a variety of studies on the interaction of 30 fs pulses focused to intensity $>10^{16}$ W/cm^2 into gases of deuterium clusters. For example, we have conducted deuterium ion time of flight energy spectroscopy. These measurements were conducted from ions escaping the plasma in a high density gas jet (and were consequently not ideal because of space charge problems and ion slowing down in the surrounding gas. Future experiments solve these problems.) This measurement indicated that ions with energy out to 30 keV are produced in the exploding clusters, however, detailed studies of ion energy spectra require an apparatus enabling interactions with a low density cluster beam. One of the principal diagnostics in these experiments has been the measurement of the 2.45 MeV fusion neutron production. To gain information on the explosion mechanisms, we examined the fusion neutron yield as a function of cluster size (see ref [1]). We find that the fusion yield increases rapidly as the average cluster size increases from 5 to 8 nm. We vary the average cluster size by changing the temperature of the gas jet backing reservoir with liquid helium cooling. The rate of the yield increase with cluster size increase appears to be consistent with a Coulomb explosion model of the ion ejection (In this model, the laser field strips the cluster of its electrons on a time scale much faster than the cluster expansion. The Coulomb forces between ions then drive an explosion. In this simple picture, the ion energies should scale as the square of the cluster size.) In addition, we find that as we increase the average cluster size further to 10 nm, the yield rolls over.

We have also conducted extensive interferometric probing of the plasmas to gain information on the laser propagation dynamics and now believe that this fusion yield roll over is the result of increased laser absorption and energy depletion in the front edges of the gas jet. We have used analysis of the blast waves produced in these deuterium jets to derive direct information on the spatial distribution of energy deposited by the incident laser. These results confirm that laser energy is strongly depleted as it enters the deuterium cluster jet [].

2) Two color pump probe experiments

We also begun pump probe experiments at LLNL to explore the expansion dynamics of larger, laser heated clusters. In particular, we have constructed an experiment to examine the third harmonic generation of 800 nm pulses in a gas of xenon clusters as the clusters expand from the photoionization and heating of a an initial pump pulse (which has a wavelength of 400 nm). This pump-probe experiment is designed to yield information on the nonlinear oscillation dynamics of the electron cloud in an expanding cluster. This experiment follows on experiments conducted at LLNL two years ago on the linear absorption of a probe laser pulse in xenon clusters as a function of delay after an ionizing pump.

We conducted initial experiments using the Falcon laser at LLNL and have seen some variation of third harmonic signal as delay between the 800 nm and 400 nm pulses. We have not yet reproduced the

original absorption measurement (which used 800 nm pulses for both pump and probe) and attribute this to inadequate intensity in the pump pulse to drive the xenon explosion. We have procured appropriate optics to enhance the pump intensity and will begin another round of experiments in the fall. These experiments will be conducted on the 5 TW Ti:sapphire laser in Mike Downer's labs at UT.

3) Interactions with large, wavelength scale clusters

Finally, we conducted a series of experiments on the production of fast electron during the interaction of our intense femtosecond pulses with clusters of diameter approaching that of the laser wavelength [4]. In these experiments, we produced water clusters with diameter around 1 μm using a special jet developed in a collaboration with Tom Donnelly at Harvey Mudd College. The 30 fs laser pulse was focused to an intensity of nearly 10^{18} W/cm^2 into a spray of these small water droplets (whose sizes were characterized by Mie scattering).

The laser produced hot electrons which then produced hard x-rays via bremsstrahlung. We measured the x-ray spectra to gain information about the electron spectra. These measurements indicated that the laser irradiation produced electrons with an effective temperature of around 1 MeV [4]. This result is remarkable because hot electrons produced from a planar target under nearly identical irradiation conditions exhibited a hot electron temperature only half that from the water droplets. We have compared these experimental results with particle-in-cell calculations and have found that this enhancement in hot electron temperature is a result of the laser field distribution around the wavelength scale particle. We are conducting further studies to ascertain the scaling of hot electrons with cluster size.

Future Research Plans

Our future research plans are aimed at an understanding of the interactions between the laser and single clusters to understand in more detail the energy deposition mechanisms. Studies during 1999 and 2000 have concentrated on interactions of the laser pulse with high density gas jets containing clusters. By using a low density molecular beam of clusters, we intend to undertake a series of studies on single cluster interactions. All of these future experiments will be conducted on the new 20 TW laser that will come on line at UT later this fall.

To do these future experiments we have constructed a time-of-flight spectrometer coupled to a molecular beam. This target chamber and spectrometer have been moved from LLNL to UT. We have fitted the chamber with a cryogenically cooled gas jet, capable of producing large hydrogen and deuterium clusters with sizes ranging from a 1 to 10 nm. Laser pulses are focused with an aspheric lens into this beam of clusters and fast ejected particles are detected along an axis perpendicular to both the laser and the cluster beam. This spectrometer is designed to yield information on both ion and electron energy spectra. Ion spectra will be characterized both through direct, field free ion time of flight as well as through the use of charged retarding grids. This spectrometer will allow us to examine ions with energy up to 1MeV and will allow charge state differentiation on ions with energy to charge state ratios up to 20 keV/Z. We will also analyze electron spectra, which will be measured by scanning the voltage on the retarding grids.

1) Our fist set of experiments at UT will be to examine the ion energy distributions from hydrogen clusters. This is important information in calculating the expected fusion yield from exploding deuterium clusters. We intend to examine these distributions as a function of average cluster size

(varied by changing the gas jet temperature) as well as laser intensity, wavelength and pulse width. We will also examine electron energy distributions. This information will be compared to simulations of the laser-hydrogen cluster interactions currently underdevelopment in my group at UT. We are developing a particle dynamics code to model the classical (relativistic) motion of electrons and ion in the cluster during strong field excitation. This round of experiments and simulations will essentially examine explosions in the pure Coulomb explosion regime from low Z species. Information derived here will then be used to optimize fusion yield in the high density deuterium gas jet experiments.

2) We then intend to examine spectra from higher Z species (namely N_2, Ar, Kr and Xe) This will allow us to explore the nature of the cluster explosions as it evolves from a pure Coulomb explosion to a hydrodynamic explosion. Here, electron and ion spectra will be compared with particle dynamics simulations (i.e. the Kulander model) as well as with hydrodynamic simulations (i.e. the Hyades hydro-code).

3) Additional experiments will follow up on the two color pump probe experiments. While we intend to continue the third harmonic experiments in Xe clusters, we will also begin to look at electron and ion spectra from these two pulse interactions. Once again, we will likely concentrate on deuterium clusters with an eye toward optimizing ion energies for fusion studies.

4) Finally, we will follow up on the micron scale droplet experiments. In particular, we will likely examine the electron spectra directly. We will begin with the electrostatic TOF spectrometer, however, it is likely that we will require a magnetic spectrometer to fully characterize the fast electrons produced in these interactions. We are now developing a Paul trap to trap micron sized polystyrene spheres to act as a monodisperse target for these studies.

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Few-Body Reaction Imaging

Department of Energy 2001

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The research described here remains part of a general effort in the AMO community to advance our fundamental understanding of collective few-body excitations. We thus continue to seek to characterize excitations for which independent particle motions are superseded by subtle balances of charge and inertia across the system as a whole, and therefore excitations for which collective few-body motion is the requisite zeroth-order description. At the same time, we have shifted aspects of our research to impact nanoscience, work we have already begun with recent DOE support. We are thus investigating the basic physics of nanoscale science, engineering, and technology (NSET) based on our long time experience in the highly-evolved AMO industry of collision and few-body phenomena. We find there exists opportunity to contribute to at least two goals of the NSET initiative of the DOE Office of Basic Energy Sciences (BES): (i) Attain a fundamental understanding of nanoscale phenomena, and (ii) develop experimental characterization tools and theory to understand, predict, and control nanoscale phenomena.

We thus consider few-body reaction detection—a long-standing theme of the BES program—from the more general perspective of reaction imaging. We accordingly distinguish two parallel efforts with (i) new emphasis on *detection with interferometers* while (ii) pursuing ongoing work on *collective Coulomb excitations*. As in the past, we continue to place strong priority on research relevant to experiment and to maintaining collaborations with experimental groups in this country and in Europe.

Detection with Interferometers

Advances in coincidence detection technology for collisions involving charged particle or photon impact have evolved into a new field of few-body fragmentation spectroscopy.[1] Full kinematic information can now be extracted systematically on all collision fragments of few-body states from measurements of the momentum vectors of each fragment. With supersonically cooled targets, momentum detection to 0.1 au is now routinely achieved. With confined and cooled targets in magneto-optical traps, this precision is likely to improve in the near future by an order of magnitude.[2] Parallel to this remarkable progress have been the developments in the field of *atom optics* concerned with manipulating with micro-structures and light fields the motion of atoms and molecules[3] and Bose-Einstein condensates[4] to observe their interference and diffraction just like waves of light. Here attention has centered on imaging the quantum state (Wigner function) of just the diffracted particle's internal and center of mass motion,[5] although beautiful progress has been made towards extracting information on path interactions.[6]

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[5] Ch. Kurtsiefer et al., Nature **386**, 150 (1997) and related references.

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In pioneering work on quantum correlation, Wootters and Zurek[7] analyzed how two-slit interference with photons changes when the recoil of a distant collimator slit is monitored. Considering complementary momentum and position measurements, they made clear that the shape of the interference pattern depends critically on the collapse of the entanglement between the wavefunctions of the diffracted photon and the recoiling collimator. In recent work, we have applied basic scattering theory to extend the description of Wootters and Zurek to projectile interferometry and target-fragment recoil detection.[8] In some respects, our motivation parallels a recent analysis of scattering with pulsed beams in which coherence is established instead between wavepackets in the incident beam also requiring off-diagonal elements of the system density matrix for a complete description.[9] Our intention is to combine few-body fragmentation spectroscopy with tools from atom optics to extract addtional amplitude and phase information from the reaction dynamics. This effort is thus much in the spirit of recent work by Forrey and coworkers to determine electron scattering amplitude information with electron interferometry.[10] At the same time, we show that the technology can be applied to manipulating microscopic devices and provide for example simple schemes for establishing basic elements of quantum computing.

Our Born-approximation analysis is readily generalized within the *impulse approximation* to non-Coulombic collisions.[11] In effect, one replaces the Born amplitude with the exact one for scattering between the projectile and any target component. For example, one might consider scattering of slow neutrons off a molecule, or (p,n) reactions near threshold, and bring to bear the considerable technology which has been developed for neutron interferometry. Along with information transfer, we are working to extend the scheme to include n-slit diffraction and other *quantum tomographic* techniques[5,6] to image for example the extremely fragile few-body Coulomb states generated near fragmentation thresholds[12] and fundamental to the few-body Coulomb problem.

In a related effort, we have considered schemes for realizing a Grover database search with wavepacket propagation and special spatial filtering. We intend to explore the method by considering electron wavepackets diffracted through mesoscopic junctions, and in particular how the efficiency of such a device might be improved with correlated electron pairs. Our approach relies on superposition without requiring quantum entanglement. However, because this special quantum correlation is widely regarded to be a key advantage of quantum over classical computing, we are working to implement it in our simulations. We note in this regard that Deutsch has shown some time ago[21] that one can extract the phase of an unknown target qubit, if one entangles it appropriately with a known 'ancillary' qubit. Grover's original proof[22] relies on a similar entanglement to track the phase of the unknown target state, although no actual device or experiment has been reported utilizing it.[13]

Single-Electron Circular Dichroism

Parallel to advances in double ionization with circularly polarized photons, there has been a renewed interest over the past ten years in photo *single* ionization and the resulting photoelectron angular distributions for departures from the dipole approximation. Following the theoretical

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[8] J. M. Feagin and Si-ping Han, Phys. Rev. Lett. **86**, 5039 (2001).

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[13] See also S. Lloyd, Phys. Rev. A**61**, 010301(R) (1999).

work of Cooper,[14] Bechler and Pratt,[15] and others, Krässig and coworkers have reported measurements of nondipolar asymmetries in photoelectrons from Ar and Kr ionized with x rays in the 3–5 keV energy range from the National Synchrotron Light Source.[16] They have shown that retardation effects will result in a forward or backward asymmetry in the ejected-electron angular distribution along the photon beam, which increases as the photon energy is increased. They have thus extracted nondipolar angular asymmetry parameters over a continuous and remarkably wide range of ejected electron energies, 30–2000 eV, not possible ten years ago.

Two-slit detection would provide new probes of these photoionization angular distributions. Even for photo *single* ionization, it appears possible[17] to generate a dichroism analogous to the well established effect seen in photo *double* ionization with circularly polarized photons.[18] Electron-pair circular dichroism is observed in the electron angular distributions and can be used to extract phase information unavailable with linearly polarized photons. (We have also considered the generalization of this effect to molecular photo double ionization.[19] See below.) The momentum vector $\mathbf{k}_\gamma$ of the incident photon along with the momentum components $\mathbf{k}_1$ and $\mathbf{k}_2$ of the two outgoing electrons can be used to mediate a handedness, which transfers to the electron pair from the circularly polarized photon. This dichroism is a measure of the difference Δ in the photo double ionization cross sections between left and right circularly polarized photons. For dipole excitation in helium, $\Delta_{12} \sim (\mathbf{k}_1 \times \mathbf{k}_2) \cdot \mathbf{k}_\gamma$.

In the case of photo single ionization, the idea is this: Introduce a two-slit detector and let the pair of momentum vectors $\mathbf{k}_\pm$ of the *single* ionized electron entering both slits take on the chiral role of the electron-pair momentum vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ in photo double ionization. In effect, we use the incident circularly polarized photons to distinguish right and left slits. One thus obtains a one-electron circular dichroism of the form $\Delta \sim (\mathbf{k}_+ \times \mathbf{k}_-) \cdot \mathbf{k}_\gamma$ as well as phase information unavailable with linearly polarized photons. Moreover, the dependence of the dichroism on the wavevectors changes if nondipolar contributions are considered. Thus, two-slit detection should prove useful in analyzing the breakdown of the dipole approximation. A new generation of experiments to study nondipolar effects at the advanced light sources are underway by Young, Krässig and coworkers.[20]

Collective Coulomb Excitations

The conventional detection of two electrons following photo-double ionization or electron-impact single ionization of simple atoms and molecules is already a highly-advanced technology, and one which has contributed enormously to our understanding of the correlated motion of electrons in the field of a positive ion. In this respect, the study of the photo double ionization of the helium atom near threshold has played a significant role. Not only is the final state one involving three unbounded particles interacting via pure Coulomb forces, but the dipole symmetry change is simple and well defined. This system has thus served as a benchmark for both experimental[21] and theoretical[22] work.

The coincident measurement of two continuum electrons has been extended to the photo double ionization of molecular hydrogen in the isotopic form D_2,[23] and recently including

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[21] R. Dörner et al, Phys. Rev. A**57**, 1074 (1998). (*Feagin is a coauthor on this paper.*)

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coincident detection of the deuterons.[24] This is of fundamental interest since the final state is one of four unbound Coulomb-interacting particles and introduces several new aspects to the few-body problem:

- The two-center geometry of D_2 introduces a line of symmetry and therefore a new vector quantity into the description of the ejected electron pair.
- The 'Coulomb explosion' of the nuclei following the removal of the electrons in D_2 gives final ionic energies of several eV, which can be of the same order of magnitude as the ejected electron-pair energy.
- The four-body Wannier threshold configuration is inaccessible by (single) photon absorption, since a vertical Frank-Condon transition puts the molecule at an energy (corresponding to the Coulomb explosion energy) well above the true four-body break-up threshold.

We have thus developed a basic description of the photo double ionization cross section for diatomic molecules[25] based closely on this cross section for helium. We derive a dependence of molecular excitation amplitudes on electron energy sharing and dynamical quantum numbers labeling internal modes of excitation of the escaping electron pair. We consider both linear and circular polarizations. We thus make predictions regarding the continuum molecular symmetries as well as the circular dichroisms in the angular distributions of the electron and ion pairs for advanced light source experiments in planning.

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Theoretical Studies of Atomic Transitions

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Program Scope and Definition

The atomic structure project is concerned with the accurate determination of wave functions from which atomic properties can be predicted. Of particular importance are properties associated with energy transfer mechanisms such as transition probabilities, where relativistic effects are essential. Light elements have been investigated primarily in the Breit-Pauli approximation, but for heavier elements our methodology is relies on variational Dirach-Hartree-Fock methods that include correclation and the Breit interaction.

Recent Progress

1. Breit-Pauli Studies

We have continued to refine and improve our "spectrum" calculations where all the energy levels up to a certain excitation level are computed along with all transitions between these levels, thus making it possible to determine lifetimes for excited states as well as brancing ratios. Codes have been ported to the most advance parallel computer at NERSC, namely the IBM SP "seaborg" computer. The transition data results are being made available for ready access on the internet :

(See `http://www.vuse.vanderbilt.edu/~cff/mchf_collection`).

We have published the evaluation of Be- [21], B- [24], and C-like [34] sequences. Calculations for N-, O-, F-like are completed but not yet evaluated, Ne-like is in progress. Our computational method is systematic allowing for an estimation of the uncertainty in the transition rates, many of which are accurate to 1% or better, though exceptional cases, where the line strengths are small due to cancellation, will always be less accurate.

2. Relativistic Multiconfiguration Dirac-Hartree-Fock Calculations

Many questions remain about how multiconfiguration Dirac-Hartree-Fock calculations can be performed accurately and efficiently. Particulary troublesome are intercombination lines. Paper [29] considered an intercombination line in the Mg-like sequence, namely $3s^2\ ^1S_0 - 3s3p\ ^3P_1$ and showed how, with a non-relativistic off-set correction, and certain energy corrections, results indendent of the details of optimization could be obtained. This work was extended to higher members of the sequence and to forbidden (M1, M2, E2) transitions between levels [32].

In some of the heavy atoms, like Lr (Z=103) and Rf (Z=104), ground states have been determined only by coupled cluster theory and resonance transitions are unknown. We have

started work on validating the coupled cluster energies, but to do so, we have first investigated Lu (Z=71) where experimental energy level data is available for comparison. So far, our findings have been that correlation in the $5p^6$ core is important, not only core-polarization. This work relies on the parallel computers available at NERSC. Similar calculations for Lr are also in progress. The graspVU codes have been improved to perform transition calculations and angular integrations for transition calculations modified for parallel execution.

Future Plans

Our spectrum codes perform well for lower members of a spectrum. Calculations for Rydberg series, in many respects, are more like continuum calculations. Since the spline approach was shown to be effective in photodetachment [11, 13] and earlier in Rydberg series calculations, we plan to extend these for more general studies. Of crucial importance will be the inclusion of some non-orthogonal orbitals as well as Breit-Pauli effects. Our first system will be transitions among Rydberg series in carbon where astrophysical observations have predicted transition rates that differ substantially, in some cases, with present theory.

We plan to investigate the $3d^4 - 3d^3 4p$ transitions in Fe V where there was a considerable discrepancy between the Breit-Pauli extension of the R-matrix method (BPRM) and the relativistic CI results obtained by Beck. However, the BPRM method neglects the two-body Breit-Pauli operators which we consider essential in many cases. Our initial approach will therefore be a variational MCHF calculation wiht Breit-Pauli corrections since, for Fe (Z=26) we believe this approximation should be adequate.

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Studies of Auotionizing States Relevant to Dielectronic Recombination

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In this program we are investigating multiple excitations of two valence electron alkaline earth atoms by laser spectroscopy. The first goal of the program is to reach an understanding of the autoionization of doubly excited Rydberg states and how it relates to the inverse process, dielectronic recombination (DR). In these experiments we take advantage of the fact that one electron absorbs a single photon, but the energy is always shared between the two electrons. The other goal is to explore the effect of intense laser pulses on two electron atoms. For intense laser pulses short relative to the Kepler time of the Rydberg electron it is possible that one electron absorbs several photons and keeps all the energy while none or very little is given to the other electron.

In the past year we have worked on two topics; dielectronic recombination (DR) from a continuum of finite bandwidth in the presence of time varying fields, and multiphoton inner electron ionization (IEI).

The recombination of an ion and an electron through an intermediate doubly excited autoionizing state, DR, is the dominant electron-ion recombination mechanism in high temperature plasmas. Such plasmas are found in fusion plasmas and in stars. The Rydberg autoionizing states play a central role in the process, and as a result, it is not surprising that small electric fields can have a significant effect.[1] Electric fields mix low angular momentum character into high angular momentum states so that more states contribute to DR, which raises the total DR rate. On the other hand, electric fields also depress the ionization limit, reducing the number of available states, which lowers the DR rate. In controlled beam experiments on DR there are always macroscopic motional electric fields, which have a profound effect,[2] and in a plasma there are microscopic electric fields, which come from the ions and electrons of the plasma. The ions produce a quasi static field, and the electrons produce a rapidly varying field.[1] The effects of quasi static fields are well understood, but the effects of time varying fields are not.

We have investigated DR from a continuum of finite bandwidth in the presence of linearly and circularly polarized microwave fields of frequencies from 4 to 12 GHz. The continuum of finite bandwidth is the broad Ba $6p_{3/2}11d$ state which straddles the Ba^+ $6p_{1/2}$ limit. We excite Ba atoms to the continuum of finite bandwidth, the $6p_{3/2}11d$ state, at a well defined energy. The quasi continuous 11d electron can be captured into $6p_{1/2}\,n\ell$ state, which decays radiatively to the bound $6s_{1/2}\,n\ell$ state, which we detect by field ionization. Here n, ℓ, and m, denote the principal, orbital angular momentum, and azimuthal angular momentum quantum numbers. We have observed a striking resonant enhancement of DR when the microwave frequency matches the $\Delta n = 1$, 2, or 3 spacing of the autoionizing Rydberg states.[3] We have verified that the effect is resonant by changing the microwave frequency and observing that the

enhancement in DR occurs at the energy for which the Δn spacing matches the microwave frequency. The origin of the resonant enhancement is easily understood. When the microwave frequency matches the Δn separation the microwave field couples high ℓ states differing in both ℓ and n by one. In other words, it is resonant ℓ mixing analogous to that produced by a static electric field.

In the past year we have worked on two aspects of this problem, extending the frequency range to 4 GHz from 8-12 GHz, and altering the microwave pulse. We wished to change the frequency to verify that the resonant enhancement occurs over a larger frequency range. In our initial experiments we used a resonant cavity to produce the microwave field, and, due to the Q of the cavity, the field cannot be turned off faster than 100 ns. During this period the recombined atoms can be ionized by the microwave field. We measured this ionization and corrected for it, but a slightly cleaner approach is to turn off the microwave field more quickly, in 10 ns, which requires a broad band microwave system, not a cavity, and more microwave power. We have done measurements with the microwave field turned off 10 ns after the laser pulse using frequencies of 4, 8, and 12 GHz. We found that the corrections we were making for microwave ionization of recombined atoms were reasonable. Perhaps more interesting, we also observed DR above the $6p_{1/2}$ limit. In other words, we observed the microwave analog of the pulsed field recombination reported by Bensky et al[4] and Wesdorp et al.[5]

In the coming year we plan to do several things. First we plan to do calculations of the couplings produced by linear and circularly polarized microwave fields. We hope to understand why the resonances in the enhancement are as broad as they are or why there is no difference between linear and circular polarization. We plan to extend the measurements to high frequencies, at first to 18 GHz, and then to 60 GHz. We have in mind two objectives, the first is simply exploring DR in higher frequency resonant microwave fields, and the second is to assess the possibility of carrying out resonance measurements between autoionizing states. Finally, we plan to explore further the low frequency dependence of recombination observed above the limit in a microwave field.

The second area of research is IEI. When an alkaline earth Rydberg atom is exposed to a laser pulse of duration shorter than the Kepler orbit time of the Rydberg electron it is possible to eject the inner electron while leaving the outer electron bound to the atom.[6,7] The proposed explanation is simple enough. If the pulse is so short, in some of the atoms the outer Rydberg electron does not come near the ion core and can not absorb the photon, while the inner electron easily absorbs the photon and leaves the atom. The outer electron is then projected onto the ionic Rydberg states.

The explanation given above is a time domain description which suggests that if we started from a Rydberg wavepacket we should see the probability of IEI oscillate as the wavepacket moves radially in and out. We have made wavepackets of Sr and have observed the expected oscillation in the probability of IEI. While our experiment shows that the time domain picture is essentially correct, it also shows that it has deficiencies. To be precise, the final Sr^+ Rydberg states produced should depend on the position of the outer electron in a well defined way, which is not observed in the experiment. We see lower lying Sr^+ Rydberg states than expected, supporting the suggestion of Rosen et al that a long range multipole interaction is involved.[8]

Somewhat to our surprise, IEI turns out to be a far better way to detect atomic wave packets than other methods, presumably because it requires that the outer electron be very close to the ion core to suppress IEI. We have demonstrated that we can see up to five revivals of the

wave packet, and, more important, that we can reconstruct the wavefunction of the wavepacket. We have also worked out the analytic theory for wavepacket oscillations for non hydrogenic wavepackets with arbitrary, i.e., not hydrogenic, energies.

In the coming year we plan to explore the ℓ dependence of IEI in Sr to see if we can develop a clear picture of the range of the interaction between the two electrons which is responsible for inhibiting IEI.

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EXPERIMENTS IN MOLECULAR OPTICS

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1. Program Scope

The objectives of this program are to develop optical methods of deflecting, focusing, and aligning beams of neutral molecules. Optical manipulation of molecules is a potentially useful tool for controlling chemical reactions, inducing specific optical transitions, and creating nanostructures by focusing particles onto a surface in a controlled way. The central idea is to use the dipole force produced by a focused non-resonant laser beam to induce a dipole moment in a polarizable atom or molecule. The potential energy produced by a focused laser beam is given by $-¼\ \alpha I$, where α is the polarizability at frequency ω, and I is the laser intensity. Depending on the sign of α, the particles will either be drawn into or repelled out of the laser focus. Moreover, if the polarizability tensor is anisotropic, the molecules may be aligned along the direction of the electric vector of the field.

Because the above discussion is general, virtually any atom or molecule may be manipulated by the dipole force, so long as the intensity is not high enough to ionize the particle. The tools that are available for manipulation depend on the properties of α and I. In one ongoing experiment we are attempting alter the value of the polarizability by changing the value of the principle quantum number, n, of the target molecule. The static polarizability is known to vary as n^7. This effect is not very useful, however, because the ionization probability of a Rydberg state varies as n^{10}. For an ac field the dynamic polarizbility has a complicated energy dependence, depending on the proximity of resonant states. At very high values of n, α becomes negative and approaches $-4a_0^2(R_y/\hbar\omega)$, where R_y is the Rydberg constant and a_0 is the Bohr radius. Clearly, for infrared radiation the magnitude of α can be very large. Because atoms and molecules in high Rydberg states are not easily photoionized, it should be possible to utilize their large polarizabilities to create a molecular mirror.

In a second experiment we control the alignment of a molecule by varying the pulse duration of the laser field. In most previous experiments, the pulse duration was much longer than the rotational period of the molecule. In this limit, the molecule aligns adiabatically with the laser field, returning to its original free-rotor state at the end of the pulse. Because one is typically interested in studying the behavior of particles under field-free conditions, adiabatic alignment produced by long pulses is of limited use. This problem may be overcome by using short pulses to create a rotational wave packet, which displays rotational recurrences long after the pulse is over. Such wave packets are produced by multiple Raman transitions in which many rotational levels are populated.

In the strong field limit the maximum alignment is achieved after the pulse is over, and recurrences occur with a period $\tau_{rec} = (2B_e c)^{-1}$, where B_e is the rotational constant of the molecule and c is the speed of light.

2. Recent Progress

The goal of the first experiment is to produce molecules in high Rydberg states that are long-lived in a strong optical field. Such states may be detected by pulsed-field ionization, producing zero-kinetic energy (ZEKE) electrons. A key part of this experiment is to measure the intensity threshold for ionization of these states by the non-resonant aligning pulse. Figure 1 is the ZEKE spectrum of OCS produced by a single UV photon, showing vibrational structure of both spin-orbit states of the ion core. This spectrum was measured in a time-of-flight apparatus that is dedicated to other experiments. We are currently constructing a new photoelectron apparatus designed specifically for the molecular optics project.

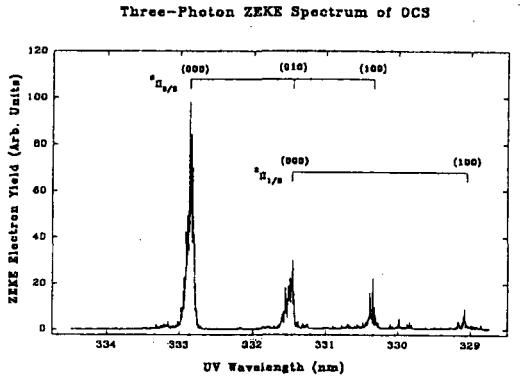

Figure 1. ZEKE spectrum of OCS.

In the second experiment we have observed the alignment of I_2 and ICl by 800 nm pulses having a duration of several ps. The molecule of interest was entrained in a He carrier gas and expanded into a vacuum chamber through a pulsed molecular beam valve. A pair of linearly polarized, 800 nm laser pulses intersected the molecular beam. The duration of the first, aligning pulse was adjusted by an expansion/compression grating pair. The relative energy of the pulses was determined by a variable beam splitter, and the delay between them was set by a roof prism mounted on a translation stage. The fragment ions were accelerated electrostatically towards a microchannel plate detector backed by a phosphor screen. Images produced on the screen by ejected electrons were captured by a CCD camera mounted outside of the vacuum chamber.

Preliminary results for the alignment of I_2 by a 2.5 ps are shown in Figure 2, where a revival at the half recurrence time of 222.5 ps is evident. The polarization direction of the aligning pulse is along the vertical axis of the image, whereas the probe polarization is along the flight direction (perpendicular to the image). The aligning pulse by itself does not produce any signal. The images show the difference between the signal produced by the probe laser alone and the signal from both pulses. The effect of the first

pulse is to align molecules perpendicular to the probe electric vector and therefore to *reduce* the total ion signal. This configuration of polarization vectors eliminates the possibility of enhanced ionization by the probe pulse of those molecules that happen to be aligned along the probe electric vector and also accounts for the observed intensity along the horizontal direction in the image. The difference signal at 60 ps is essentially zero, with a residual that displays no anisotropy. The alignment observed at $\tau_{rec}/2$ and its absence at an intermediate time are indicative of wave packet evolution.

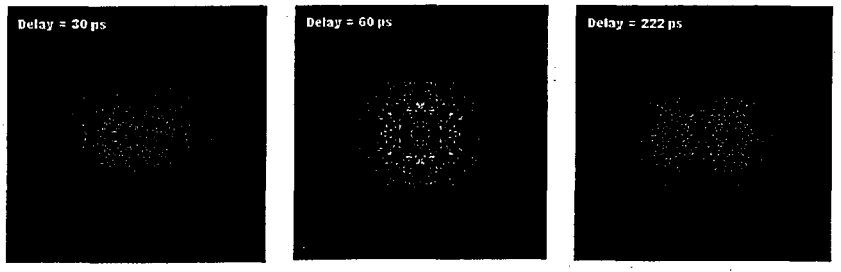

Figure 2. 2D symmetrized ion images showing the alignment of I_2 rotational wave packets.

3. Future Plans

During the coming year we plan to continue the two experiments described above. Once the new photoelectron spectrometer is operational, we plan to measure the ionization thresholds of ZEKE states of nitric oxide and pyrizine by 1064 nm radiation. When that experiment is completed, we will move the experiment over to our imaging apparatus, where we will measure the defocusing of a molecular beam by a focused laser beam.

Concurrent with the high-Rydberg experiment, we will continue our short-pulse alignment studies of rotational wave packets. We are currently building a new laser amplifier that is designed to produce 1 mJ/pulse @ 800 nm @ 1 kHz, which is an order of magnitude more powerful than our current pulse energy. We are also designing a piezo-electric driven pulsed nozzle source for our molecular beam that will be capable of operating with corrosive gases at 1 kHz. With these improvements we plan to study the effects of laser intensity, pulse duration, and chirp on the alignment. In a later experiment we plan to study the effect of multiple pulses designed to "kick" the rotor at its natural recurrence frequency.

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Experiments in Ultracold Collisions

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Program Scope:

The ability to cool and confine atoms with laser light has enabled many new and exciting research directions in atomic, molecular, and optical (AMO) physics. These techniques allow the generation of atomic vapors at high densities (e.g., $n>10^{11}$ cm^{-3}) and ultracold temperatures (e.g., $T<100$ μK). Such samples are the starting point for many applications, including Bose-Einstein condensation (BEC) and atom lasers, degenerate Fermi gases, improved atomic clocks, photoassociative spectroscopy, ultracold molecule production, optical lattices, ultracold Rydberg atoms and plasmas, and fundamental atomic and nuclear physics experiments with radioactive isotopes. In many of these research areas, the effects of ultracold collisions are important. For example, at high atomic densities, inelastic collisions can eject atoms from traps and/or increase their temperature. Improved knowledge and control of these collisional interactions will benefit applications of ultracold atoms. The main goal of our experimental program is to better understand collisions between ultracold atoms, in particular, those which occur in a typical laser trap environment.

In addition to the importance of our experiments to applications of laser-cooled atoms, there is a great deal of novel physics associated with these extremely low energy (e.g., $\sim 10^{-8}$ eV) collisions. Although various collisional channels exist, no kinetic energy is available, so only exoergic processes are allowed. Since the collisions generally involve low angular momentum, quantum effects are often important. In contrast to collisions at higher energy, the dynamics can be dominated by the long-range ($R\sim 100$ nm) excited-state potentials, i.e., the colliding atoms begin to "feel" each other at extremely long range. This allows control of the collision dynamics with laser light. Finally, the combination of the long range of the potential and the low collisional velocity yields a collisional time scale which can exceed the excited-state lifetime. The atomic excitation can therefore spontaneously decay during the course of a slow collision, dramatically affecting the outcome.

All of our experiments utilize near-infrared (~780 nm) diode lasers to capture, cool, and confine rubidium atoms in a magneto-optical trap (MOT). The choice of rubidium is based on three factors: 1) the convenient match of its resonance lines with commercially available diode lasers; 2) the existence of two abundant isotopes (^{85}Rb and ^{87}Rb) to check isotopic effects in collisions; and

3) the popularity of ^{87}Rb in BEC experiments. Atoms are loaded into the MOT from either a laser-slowed beam or a room-temperature vapor. Measurements of inelastic collisions are carried out by monitoring the loss rate of atoms from the trap. Specific processes which can eject atoms from the trap include: radiative escape (RE), where the excited-atom pair emits a less energetic photon than it absorbed; fine-structure-changing (ΔJ) collisions; and hyperfine-changing (ΔF) collisions.

Recent Progress:

During the past year, we have: 1) completed our measurements of photoionization of the highly-excited Rb 5D level; 2) made significant progress towards ultracold collisions induced by chirped laser light; and 3) demonstrated and characterized the frequency modulation of injection-locked diode lasers. We briefly describe each of these advances below.

Recent experiments on the highly-excited 5D level in Rb have focused on photoionization. This is a loss process process which competes with ultracold 5D collisions, so we needed to know its cross section. To efficiently prepare cold atoms in the 5D level, we use two-photon diode-laser excitation from the ground state: 5S$\rightarrow$5P$\rightarrow$5D, with pulses applied in the "counterintuitive" order, i.e., 5P$\rightarrow$5D followed by 5S$\rightarrow$5P. The photoionization cross section is determined by measuring the increased loss rate of atoms from the trap when photoionizing light is applied. In addition, by using intense pulsed laser light, we completely ionize all 5D atoms, which gives a direct measure of the 5D excitation efficiency. We have measured the absolute photoionization cross section over a range of wavelengths from 532 nm to 1064 nm, obtaining very good agreement with theoretical calculations performed by H.R. Sadeghpour.

In a previous experiment, we observed ultracold collisions in real time by using two pulses, delayed with respect to each other in a pump-probe configuration. The first pulse, tuned close to the atomic resonance, excites the colliding atom pair at very long range. The atoms accelerate towards each other on the attractive excited-state potential, decaying back to the ground-state as they approach short range. A second pulse, detuned significantly below the atomic resonance, re-excites this enhanced collisional flux, causing an observable inelastic process at short range. If instead of applying temporally separated pulses at different frequencies, we use a frequency chirp, we can exert a higher degree of control over the collisions. With a blue-to-red chirp, we can maintain resonance during the course of the collision. With a sufficiently intense red-to-blue chirp, the population can be adiabatically inverted, significantly enhancing the collision rate. This type of active control may prove useful in enhancing the rate of cold molecule formation via photoassociation, a process which occurs at relatively short range. We have achieved chirp rates exceeding 1 GHz in 100 ns which is well matched to the temporal dynamics of the collisions. Preliminary results of these experiments are promising.

An important technical advance in our lab involves the modulation of diode lasers at microwave frequencies. Cooling and trapping of rubidium atoms requires laser light at two frequencies separated by 2.9 GHz for ^{85}Rb (6.6 GHz for ^{87}Rb). We have previously used two

separate external-cavity diode lasers to generate these trapping and repumping beams. A simpler scheme is to stabilize one external-cavity laser at the trapping frequency and inject the light from this "master" laser into a free-running "slave" laser. The frequency of the slave laser is thereby injection-locked to that of the master. We then modulate the current in the slave laser at 2.9 GHz (or 6.6 GHz), which generates the desired sideband for repumping. There are two advantages to this scheme. The slave laser provides a higher power than the master, and only one laser needs to be locked to an atomic resonance.

Future Plans:

We have two main experiments in ultracold collisions planned for the coming year. First, we will continue our work with frequency-chirped collisions, building on the promising preliminary results we have obtained. This will involve characterizing the collisions as a function of the rate, range, and direction of the frequency chirp, as well as the intensity of the laser. The intensity is particularly important since it determines whether the excitation is adiabatic. We will attempt to increase our intensity by injecting a "slave" laser with frequency chirped light. In addition to our collisional investigations, we need to understand the effects of the chirped light on atomic excitation, since this can result in perturbations of the atomic cloud.

In our second planned experiment, we will expand the temperature range of our recent measurements of ^{87}Rb ground-state hyperfine-changing collisions, both in the presence and absence of laser light. We also plan to make analogous measurements on the other isotope, ^{85}Rb. This will hopefully allow us to further constrain the ground-state potentials and determine the origin of the light-induced enhancement which we previously observed.

Recent Publications:

"Ultracold Collisions Observed in Real Time", S.D. Gensemer and P.L. Gould, Phys. Rev. Lett. **80**, 936 (1998).

"Ultracold ^{87}Rb Ground-State Hyperfine-Changing Collisions in the Presence and Absence of Laser Light", S.D. Gensemer, P.L. Gould, P.J. Leo, E. Tiesinga, and C.J. Williams, Phys. Rev. A **62**, 030702(R) (2000).

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Annual Technical Progress Report for the period July 1, 2000 – June 30, 2001
DOE Grant: *Physics of Correlated Systems*
Principal Investigator: Chris H. Greene

1. Connecting closed-orbit theory with quantum defect theory

The PhD dissertation of Brian Granger was completed in August, 2001. His research work developed a rederivation of closed-orbit theory within the context of a semiclassical approximation to quantum defect theory [1]. He has extended those techniques to treat atoms more complex than hydrogen, in the presence of a uniform electric and/or magnetic field, with the applications to date limited to diagmagnetic Rydberg states. A major theme of the research has been to unscramble the physics associated with the quantum mechanical pathways associated with core-scattered classical orbits, which cause additional interferences and peaks in the time domain that are absent from hydrogenic spectra. A major development in this research in recent months has been an understanding of how "core-scattered" recurrence peaks in the time domain are intimately related with the so-called "non-classical ghost orbits" that have been discussed by others such as J. Main. A manuscript on this subject is expected to be ready for submission soon.

One unexpected development that arose during the past six months was Granger's realization that the semiclassical methods he has developed should permit a simple interpretation of the electron nodal patterns of long range Rydberg molecules. This led to a long paper that will appear in print soon.[2] This collaboration is expected to continue at a reduced scale during the coming year, after Granger assumes a postdoctoral position at ITAMP during the fall of 2001. One application intended for study is an application of the reformulated theory to a broader class of systems that have not been accessible to closed-orbit theory in its existing forms. A prime example is photoabsorption in the presence of an external electric or magnetic field, by an atom possessing a complex, multichannel ionic core.

2. Correlations between the electronic and nuclear motions in an electron-molecule collision

The process $e + HD^+ \rightarrow H^+ + D^-$ has recently been studied in a storage-ring experiment at Stockholm. Such resonant ion-pair formation events proceed through an intermediate doubly-excited resonant state of HD, which then dissociates following the initial electronic excitation. The experiment showed around a dozen sharp peaks in the cross section for D^- formation. The P.I. was involved in developing a Landau-Zener-Stueckelberg description that semiquantitatively explains the observed peaks as resulting from a quantum mechanical interference between competing dissociation pathways.[3]

3. Bose-Einstein condensation studies

One BEC-related portion of this project was a study of the behavior of a condensate with a negative or attractive scattering length. What happens in real time is that the number of Bose-Einstein-condensed atoms grows up to some maximum number, after which it then collapses. A condensate of ^{7}Li atoms, for instance, is the most prominent example since it has been studied extensively by the group of R. Hulet. Its maximum size has been measured to consist of approximately 1400 atoms, in agreement with theoretical expectations. After the condensate grows to contain this many atoms, calculations by H. van der Hart show how it undergoes a rapid collapse to a very small but nonzero number of atoms remaining in the condensate. A paper describing the model and the numerical experiment was published during the past year.[4]

4. Scattering amplitudes for electron-impact ionization

This grant has supported the continuation of work on electron-impact ionization initiated by Mark Baertschy while he was a graduate student at U.C. Davis. A complete solution to this problem at any given energy usually means calculating differential cross sections that describe the scattering angles of both the scattered and ejected electron as well as the distribution of kinetic energy between the two outgoing electrons. We are working towards a more thorough understanding of the fundamental dynamics of a state

with two outgoing electrons, so we focus our attention on the relatively simple yet physically relevant problem of electron-hydrogen scattering.

The most significant accomplishment in this project is the development and implementation of an entirely new formalism for extracting ionization information from a wave function, described in a recent paper. [5] This formalism gives an integral expression for the ionization amplitude that provides a more accurate and widely applicable means of generating differential cross sections than does our original method. Also, the computational efficiency of the codes used for solving the time-independent Schroedinger equation has been improved and the codes have been ported to a faster supercomputer. As a result, a complete wave function for a particular collision energy can be produced in about a half to a third of the time.

Our studies have focused primarily on energies within a few eV above the ionization threshold. Correlation between the two outgoing electrons is most significant in the near-threshold region. Consequently, formalisms based on separable expansions, such as convergent close-coupling (CCC) and various R-matrix hybrids, have difficulty calculating the energy-distribution for collision energies within about 30 eV of threshold. There has been much controversy surrounding the discrepancy between our results and those of CCC. It is hoped that continued work on the near-threshold behavior of ionization will resolve these problems.

5. Atoms in an intense laser field

In this project we are studying the double ionization of a helium atom by a short, intense laser pulse in order to understand more about the specific mechanisms leading to double photoionization. We have developed algorithms for propagating the time-dependent Schroedinger equation for a two dimensional model atom that has physical properties similar to those of helium. Our preliminary results appear to favor the recollision model where the second ionization event is caused by electron-impact from the first electron to be ionized rather than by a second photoionization. Definitive results will require a more efficient numerical representation of the two-dimensional wave function. To that end we are developing a hyperspherical approach where the two-dimensional wave function is represented in terms of coupled one-dimensional functions of the hyperradius. We expect to see sufficiently rapid convergence with respect to the number of hyperspherical potential curves that this method will be more efficient for propagating the time-depedent Schroedinger equation on larger grids than methods based on Cartesian coordinate systems. This project and the next one will increasingly become the focus of this research effort during the next year of funding.

6. Coherent control of rotational wave packets

A recent collaboration has emerged between our group and the experimental group of Kapteyn and Murnane at JILA. Using femtosecond laser pulses they are able to form coherent rotational wave packets of CO_2 molecules. At regular intervals, signatures in the spectral data from subsequent "probe" pulses have been identified as indications of periodic revivals of the coherent wave packets. Using a simple rigid rotor model we have been able to reproduce this spectrum in detail. Following completion of our current preliminary study we plan to extend the model to provide an even more detailed description of the system.

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Quantum Theory of Collective Effects in the Atom Laser

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I. Overview of research activity

During the past year, we have continued to develop a fully quantum mechanical description of both condensed bosonic gases and quantum degenerate fermionic gases. The motivation for this project has been to formulate quantum kinetics as required to provide an adequate description of nonequilibrium devices such as the atom laser. Our core results to date have been applied to a variety of interesting topics that are currently being studied throughout the world in investigations of the collective behavior of quantum degenerate gases.

II. Resonance superfluidity in a Fermi gas

A major highlight of the research activities this year was a prediction that it should be possible to achieve superfluidity in a quantum degenerate Fermi gas at temperatures which are currently accessible experimentally [1].

In general terms, the phenomenon of superfluidity is closely related to Bose-Einstein condensation, as was shown in the foundation of the microscopic theory of superfluid ^{4}He. In bosonic fluids the phase transition is marked by the appearance of a macroscopic number of bosons in the lowest quantum state. In fermionic systems the occurrence of superconductivity and superfluidity in superconductors and liquid ^{3}He is due to the emergence of a pairing field and thereby, in a generalized sense, to a condensation of Cooper pairs. We showed that a dilute fermionic alkali gas could undergo a transition to a superfluid state at an extraordinarily high transition temperature T_c, that can be up to half the Fermi temperature T_F. This remarkable value of T_c can be achieved through a Feshbach resonance pairing mechanism.

The application of regular BCS theory to a dilute Fermi system involves a treatment of the interatomic interactions by means of a mean-field energy proportional to the scattering length a. A superfluid phase transition will generally only occur when a is negative. It has been previously pointed out (e.g. by Stoof and coworkers) that the presence of a scattering resonance can be used to obtain a very large negative value for a and thus a high superfluid transition temperature. However, direct application of the regular BCS theory close to resonance cannot be done due to the breakdown of a number of underlying assumptions. These include violation of the diluteness criterion for the gas, and the inapplicability of the assumption that the scattering phase shift is independent of the scattering energy. In order to address this problem, we derived a renormalizable low energy effective field theory that also holds in close proximity to a resonance. The Hamiltonian was formulated by separating out the resonance state and treating it more or less exactly which is similar in spirit of the original Feshbach description of a scattering resonance.

The possibility to vary the magnetic field gives the unique opportunity to explore the intriguing crossover regime between the known BCS and BEC systems (see Fig. 1). Since the value of T_c at resonance is larger than the temperatures already achieved in a degenerate Fermi gas of potassium or lithium atoms, it may be possible to create this new type of quantum matter with current technology.

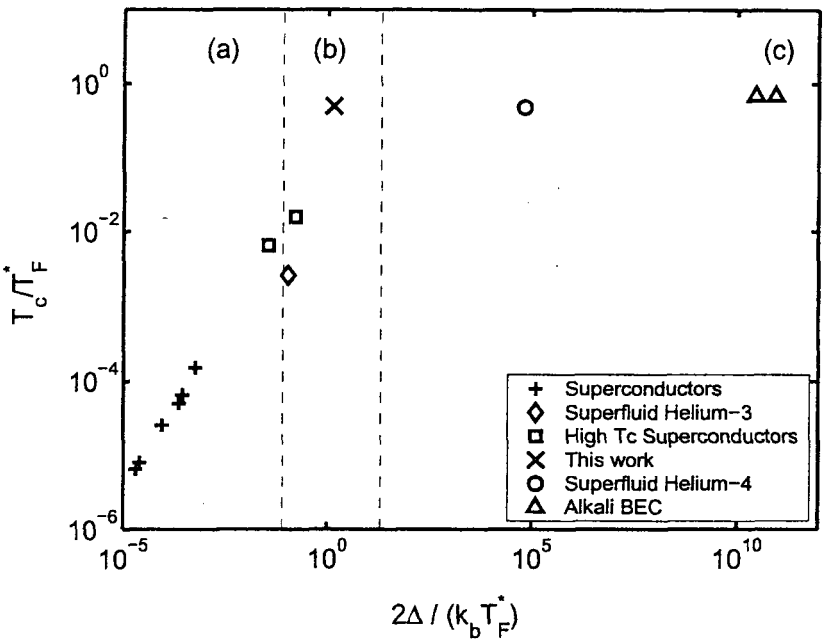

Figure 1: The transition temperature T_c is shown as a function of the relevant gap energy 2Δ for six distinct regimes for quantum fluids. Both quantities are normalized by an effective Fermi temperature T_F^*. For the BCS systems in region (a) and (b), 2Δ is the energy needed to break up a fermion pair. The systems in region (c) are all strongly bound composite bosons and exhibit BEC phenomenology. Here 2Δ is the energy needed to break the composite boson up into an electron and an ion, and T_F^* is the ionic Fermi temperature.

III. Kinetic theory of Bose-Einstein condensates

During the past year we have also continued research into the development of quantum kinetic theory to describe phenomena such as the initiation of condensate growth and phase diffusion in dilute trapped gases. A few years ago, we provided a general solution for the coupled master equations describing the time-evolution of condensate mean fields, thermal normal densities, and anomalous fluctuations, which interact through binary collisions, and reduce to the Gross-Pitaevskii equation and the quantum Boltzmann equation in the respective limits. We have now solved these equations numerically under the assumptions of vanishing anomalous fluctuations and ergodicity for the normal fluctuations, which implies neglecting quantum coherences. However, the inclusion of these coherences was the novel and salient

feature of the approach, and thus a new way to obtain solutions of the full equations would be highly beneficial.

To approach this problem, we reformulated the above theory in terms of Bogoliubov quasiparticles. The equation of motion for the quasiparticles took the shape of a much simpler quantum Boltzmann equation, which was capable of describing the system in equilibrium at temperatures above and below the phase-transition as well as out of equilibrium. We found that the second-order collisional terms take a very symmetric and simple form. This shows that the complicated collision operators that appear in the JILA kinetic equations of [4] and complicate the direct solution of these equations do not reflect physical structure. We thus believe that our new equations are more accessible to numerical simulations.

A very significant point has been the demonstration of the equivalence of this JILA kinetic theory and the non-equilibrium Green's function approach originally proposed by Kadanoff and Baym and more recently applied to inhomogeneous trapped systems by M. Imamović-Tomasović and A. Griffin. We established this equivalence by deriving the JILA kinetic equations from the Kadanoff-Baym equations of motion, using the gapless, second-order Beliaev approximation for the collisional self energies. We renormalized our theory by upgrading the two-body potential to T-matrices, which removed the divergences associated with the anomalous averages. M. Imamović-Tomasović resolved this issue by dropping the anomalous averages, which we find to be not negligible in numerical simulations, such as [4].

IV. Phase diffusion in a continuous wave atom laser

We have considered the description of phase diffusion in a continuous wave atom laser by a simplified model consisting of a dilute gas of atoms trapped in an isotropic spherical box. The reason for using such a box is that the problem is then reduced to an effective one dimensional problem with eigenmodes generated by spherical Bessel functions. In this system, we consider s-wave scattering and a contact interaction as applicable to low energy scattering. We have developed such a simple model to allow us to easily solve the quantum kinetic equations that we have formulated for general systems.

The quantum kinetic theory involves separating the physical variables into two components; a mean field (representing the condensate), and a fluctuation piece (representing the thermal cloud). We treat the continuous loading of the atom laser by coupling the spherical box to a reservoir of thermal atoms resulting in a constant input flux. The thermal atoms interact via binary collisions. Above a threshold energy level, hot atoms are removed via evaporative cooling. In addition, cold atoms are also removed from the lowest energy level to describe output coupling. Providing certain threshold conditions are satisfied, this situation may lead to the formation of a condensate in steady state. Thus we have a system which outputs a continuous beam of coherent atoms in analogy with the optical laser.

Although the complete predictions of this model represents work in progress, to date we have examined the solution in the following situations:

1. In the absence of fluctuations, we have solved the Gross-Pitaevskii equation to obtain the steady state solution. We typically start with an initial guess for the spatial dependence of the mean field and use a steepest descent method to obtain the steady state solution. The solution we obtain is consistent with the expected Thomas Fermi

solution providing a sufficiently large number of atoms is considered.

2. In the absence of a condensate, we have solved the kinetic equations for the fluctuations including all collision terms. The first order terms give the reversible evolution which was confirmed by our simulation. Particle number and energy was perfectly conserved in the first order calculation. The second order terms represented the collisional effects. They involved off the energy shell scattering events. We have shown that our equations contain the expected behavior that any initial distribution relaxes to the steady state Bose Einstein distribution with particle number conservation satisfied. We have also confirmed the expected energy conservation.

We have therefore completed development of the necessary computer code to describe the nonequilibrium kinetics, and future work will involve the incorporation of the mean field with the normal component, and the inclusion of the input and output feeding and loss processes. We anticipate that these should be straightforward steps and will then give a sophisticated and complete treatment of the collective atom laser physics.

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Toward Cooper Pairing of Fermionic Atoms

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Our previous work [1], as well as more recent work by two other groups [2], introduced a new quantum system by cooling a trapped gas of Fermionic atoms to ultralow temperatures. An exciting possibility in this system is a phase transition to a Cooper-paired state, analogous to superconductivity in metals. If found experimentally, this predicted new phase in the atomic gas system would have enormous potential in elucidating the underlying physics of superconductivity. In particular, recent theoretical efforts have pointed out that Cooper pairing in the atom gas could very well occur in a regime of temperature compared to the Fermi temperature that is more similar to exotic high temperature superconductivity than to standard BCS superconductivity [3]. One of the primary challenges in realizing this predicted new phase is in cooling the gas to sufficiently low temperatures relative to the Fermi temperature. In this project we will be cooling a Fermi gas indirectly through thermal contact with a directly cooled Bose gas. With the appropriate choice of bosonic and fermionic atoms, this system has the potential to reach further into the quantum regime than any previous result. Additionally, the Bose/Fermi mixture will open up new possibilities for investigating a mixed quantum system.

We are building an apparatus to produce Bose/Fermi mixtures using ^{87}Rb and ^{40}K. Many experiments worldwide have shown that a gas of ^{87}Rb atoms can be cooled efficiently to produce stable Bose-Einstein condensates. In addition, our group has expertise in studying a Fermi gas of atoms using ^{40}K. Using the more efficient cooling of ^{87}Rb in the combined system we should be able to cool the fermionic atoms further into the quantum regime and begin exploring the possibility of Cooper pairing in the Fermi gas. Recent progress has been in setting up the new apparatus and achieving the first ^{87}Rb/^{40}K two-species magneto-optical trap (MOT).

The vacuum chamber has been assembled, baked out, and pumped down to low pressure. Both a homemade enriched K atom source and a commercial Rb atom source were included in the chamber. A novel laser system that can provide up to 500 mW of light, divided among the four frequencies needed to trap both ^{40}K and ^{87}Rb, has been developed and tested. We have now independently characterized the ^{87}Rb and ^{40}K MOT's as well as the simultaneous two-species ^{87}Rb/^{40}K MOT in this system. A manuscript describing these results has been submitted [4].

For the next stage of cooling (forced evaporative cooling of Rb and sympathetic cooling of K) the pre-cooled atoms will be loaded from the MOT into a purely magnetic trap. Once in the magnetic trap the gas will be transferred to a higher vacuum portion of the chamber simply by mechanically moving the magnetic trap coils. In the higher vacuum part of the chamber we expect lifetimes on the order of 100's of seconds for the trapped gas. Here the atoms will be transferred into a Ioffe-Pritchard type magnetic trap and then evaporatively cooled. (Forced evaporation will be used to cool the ^{87}Rb gas, while the ^{40}K gas will be cooled sympathetically simply through its thermal contact with the ^{87}Rb gas). The first magnetic trap coils have been constructed and tested, and are mounted on a computer controlled track for the mechanical transfer to the high vacuum part of the chamber.

In the immediate future we will be transferring the atoms from the MOT into this magnetic trap. We are also developing a Ioffe-Pritchard-type magnetic trap for the evaporative cooling. Our first experiment will be to measure the collision rates in the mixed gas in preparation for sympathetic cooling of the fermionic atom gas. After demonstrating sympathetic cooling of ^{40}K we plan to explore the ultimate limits of this cooling strategy in our two-species system. Ultimately we will investigate interactions in the mixed Bose/Fermi gas, including the possibility of a boson-mediated attraction that could drive Cooper pairing of the Fermionic atoms at ultralow temperature.

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High Resolution Spectroscopy of Cluster Ions in Discharges, Clusters in Jets, and Nanoparticles

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1.0 Introduction

Our objectives are: (1) measure the spectra of ro-vibrational states of cluster ions formed in high-pressure discharges and to study their dynamics, (2) measure the surface and bulk electronic quantum states of nanocrystals produced by a unique method of synthesis--laser ablation of microspheres (LAM), and (3) study the dynamics of excitons in structures of nanoparticles. Surface defect states produced by LAM are expected to be different from those grown chemically; and the knowledge of their defect states are essential to their use as optical devices. In contrast to chemical synthesis of nanocrystals, LAM is compatible with standard microelectronics fabrication techniques.

Using near-field scanning microscopy (NSOM) and microluminescence techniques, we are working to obtain the spectra of single nanocrystals correlating their spectra with size. This size selectivity is limited by the Δz resolution of near-field techniques. These spectra will be compared with spectra obtained in a supersonic jet apparatus using resonance excitation followed by photoionization (REMPI) with time-of-flight mass analysis. In the latter experiments, semiconductor nanoparticles produced by LAM will be precooled to ~150 K and then expanded through a pulsed nozzle to achieve phonon temperatures less than 20K. Such experiments have the advantage of obtaining spectra in vacuum--free of substrate and environmental effects; and REMPI can obtain spectra of nonradiative, surface defect states. The disadvantage of REMPI spectra is that the accumulation of the spectrum requires the observation of a large ensemble of particles. Though mass selected, and therefore of a uniform size, there may be a distribution of defects among the particles. We hope that a comparison of the single-particle fluorescence spectra of nanoparticles with REMPI will develop a clear understanding of the quantum states for semiconductor nanocrystals formed by LAM.

2.0 Single-particle spectra of semiconductor clusters

First experiments attempting to measure microluminescence from silicon nanoparticles produced by LAM were unsuccessful. Fluorescence was quenched either by nonradiative trap states on the surface of the particle, by tunneling to the substrate, or the bulk bandgap has filled by strain produced by surface reconstruction.[1] Experiments by Krauss, et. al[2] suggest that native oxide on silicon should prevent tunneling to the substrate.

Following initial failures, we prepared samples of silicon nanoparticles produced by LAM in He/H_2 and pure He carrier gases. In the case of the He/H_2 carrier gas, hot nanoparticles (T~5000K) are ejected into the cooling He/H_2 laser plasma after nucleation where we hoped they would grow a hydride capping layer before surface reconstruction. A second set of samples produced in pure He carrier gases were exposed to air after collection so they formed native oxide coatings; a third set of samples were first exposed to H_2 at the collection wafers. All three types of particles were collected dry onto either Si wafers with stable native oxides, or onto flat and polished sapphire wafers. In regions with a sparse deposition of nanoparticles, we have observed no fluorescence.

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[1]Bare silicon nanoparticles with surface reconstructrion are expected to be metallic, see Lei Liu, C. S. Jayanthi, and Shi-Yu Wu, "Factor Responsible for the Stability and the Existence of a Clean Energy Gap of a Silicon Nanocluster," to be published J. Appl. Phys.

[2]Krauss TD and Brus LE, "Charge, Polarizability, and Photoionization of Single Semiconductor Nanocrystals," Phys. Rev. Lett., 83 (23), 1999, pp. 4840-4843

Since surface defects have proved to be less of a problem for CdSe nanoparticles, we measured spectra of isolated, individual particles collected on sapphire substrates. Charging and bleaching was not a problem, and we found that 50% of the nanoparticles observed were fluorescent. We did not observe the blinking of the particles as observed by Bawendi, and coworkers,[3] perhaps because of dry collection. An example spectrum is shown in Fig. 1 where we observed a broadened spectrum as expected for excitons at room temperature. Subsequent experiments attempting to observe similar particles at low temperatures were not successful. Analysis of these particles by XRD using high-resolution TEM demonstrated that the particles had become Cd rich over several weeks of storage (in argon) while waiting access to the microluminescence apparatus. Oxidation, even in an argon filled glove box, resulted in loss of Se in the form of SeO_2.

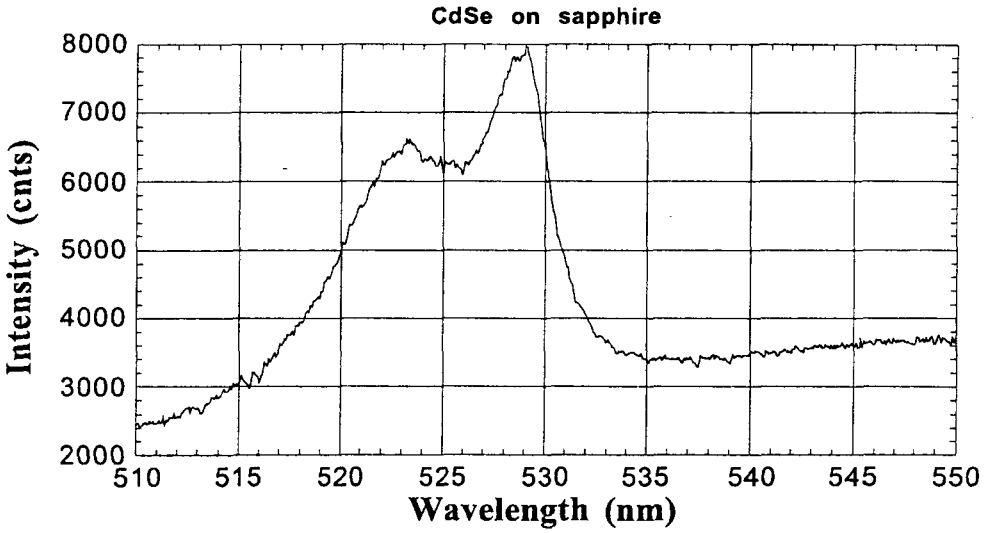

Fig. 2. Microluminescence of CdSe nanocrystals collected dry on sapphire substrates. The particles were produced in 1 atm of N_2 and collected dry on sapphire substrates. The shoulder appearing at wavelengths >535nm results from sapphire fluorescence.

Though our ultimate goal is to prepare dry samples for single particle spectroscopy, it has proven more productive to test different techniques of synthesis on particles collected at large density in solution. These are then tested using a fluorimeter to determine the quantum efficiency for fluorescence and to measure the room temperature absorption and fluorescence spectra. We first tried supersonic impaction into either Nonanoic acid or a solution of 10% by mass Tri-octylphosphine oxide (TOPO) in octanol. We can recirculate the solution through the collection region many times and hence produce very high densities of nanocrystals in the liquid. We have demonstrated that the particles are spherical in shape and remain non-agglomerated at densities of 1 x 10^{17} cm-3 which corresponds to approximately a 25% concentration by mass.

Nonanoic acid as a collection liquid has several interesting properties. It has a strong affinity for the surface of the CdSe nanocrystal, however, the surface passivation is not complete, resulting in loose flocculation of the particles. This property is valuable because it allows nanocrystal solutions to be further concentrated from the collected solution and even "slurries" of nanoparticles can be produced approaching 50% mass density. Unfortunately, the incomplete surface capping leads to an inability to carry out size selective precipitation so as to produce size distribution with ~5% dispersion.

[3]Shimizu, K.; Empedocles, S. A.; Neuhauser, R.; Bawendi, M. G. Stark spectroscopy investigation of spectral diffusion in single CdSe quantum dots Proc. - Electrochem. Soc., 98-19 (Quantum Confinement: Nanostructures), 1999, pp. 280-285

We have experienced similar behavior with silver nanoparticles captured in Nonanoic acid. As produced, the sample solutions are opaque and a dark gray to black in color. If allowed to settle a dense precipitate forms at the bottom of the test tube. We find, however, that we can stabilize the nanoparticles in solution by annealing the sample at temperatures of 140 C for several hours. After dispersing the particles ultrasonically, a sequence of beautiful color changes occur with temperature, progressing from a muddy brown color, to dark orange, and translucent yellow. Finally the samples become as transparent as water, though the density of nanoparticles is ~1 g/50ml. Once transparent, it is difficult to remove the particles by even centrifugation. We believe this successful dissolution of the particles results from a change in the surface coverage, perhaps due to a change in orientation of the surface binding of the molecule.

Spectra of similar samples of annealed, CdSe particles in Nonanoic acid were obtained and we obtained only weak fluorescence. Perhaps the surface interaction of CdSe with Nonanoic acid results in a non-fluorescent surface trap state. A system which has been demonstrated to provide excellent passivation of the CdSe nanocrystal surface is TOPO in Anhydrous-butanol or Octanol. The TOPO strongly binds to the surface and can provide enough steric hinderance to attractive van der Waals forces that the particles remain unflocculated. Our best results have used a 10% solution by weight of TOPO in octanol.

The second issue we addressed is that oxidation of the CdSe nanocrystals destroys their optical properties. Alivisatos and co-workers showed that TOPO capped particles suffer preferential oxidation of Se from the surface of the crystal. SeO_2 was found to be volatile. It is believed that the TOPO molecule binds primarily to the Cd on the surface, leaving the Se exposed to oxygen which diffuses through the TOPO capping layer. We hypothesized that a hydrogen capping layer on the bare CdSe nanocrystals would lead to a surface more resistant to oxidation.

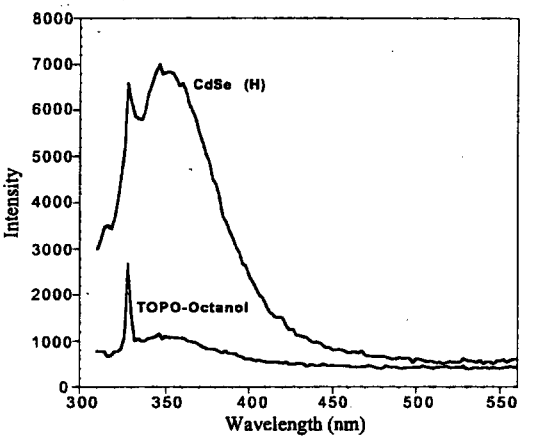

Fig. 2. Spectra of hydrogen capped CdSe nanoparticles collected in TOPO/octanol. The lower spectrum of the solvent without particles results from Raman scattering of the 300 nm excitation lamp.

To test this hypothesis we injected 4% by volume of Hydrogen into the microparticle carrier gas so that it reacts with the nanocrystals as they are ejected from the exploding microsphere into the aerosol carrier gas. The particles readily react when injected hot (T~5000 C) into the cooling laser plasma. These particles are then supersonically deposited directly onto a carbon coated transmission electron microscope (TEM) grid and imaged in a JEOL 2010 high resolution TEM equipped with an energy dispersive x-ray fluorescence spectrometer (EDS) to determine elemental composition from small areas of the sample. Preliminary results have been encouraging and indicate that the hydrogen enhances oxygen resistance of the dry particles significantly. The hydrogen capping begins to breakdown after about four days when stored in ambient air while the non-hydrogen capped samples loose significant amounts of Se by the second day.

Finally, we have obtained fluorescence from TOPO/octanol solvated CdSe nanocrystals produced by LAM and capped with hydrogen. A spectra of nanoparticles produced in a helium/hydrogen aerosol is shown in Fig. 2. The fluorescent wavelength shown in Fig. 2 is significantly shorter than that observed in Fig. 1. This blue shift is consistent with a mean particle diameter near one nm, which is also consistent with our previous experiments studying the size dependence of Ag nanoparticles with aerosol pressure and gas type.[4] Both spectra demonstrate a

[4]William T. Nichols , Gokul Malyavanatham , Dale E. Henneke ,James R. Brock , Michael F. Becker, John W. Keto, and Howard D. Glicksman, *"Gas and Pressure Dependence for the Mean Size of Nanoparticles Produced by Laser Ablation of Microparticles in Aerosols"*, J. of Nanoparticle Res. **2**, 141-145(2000).

significant quantum confinement of the excited electron. We will now attempt to obtain spectra of hydrogen capped CdSe particles which are dry deposited.

3.0 Future Research: Supersonic jet experiments

We have also built and tested a new design for a powder feeder so that semiconductor clusters can be produced by LAM in a supersonic jet. A schematic of the apparatus when modified is shown in Fig. 3. The new aerosol generator employs a small drum head driven by an oscillating solenoid. Powder resting on the drum head is driven vertically into the atmospheric gas and then descends slowly. As the gas column traverses the cloud particles are carried down stream toward the nozzle. We have measured particle densities of 10^7 cm^{-3} in the output tube of this device. The design is compact, operates better at the high pressures preferred by the nozzle, and operates independently of the pressure and flow rate of the aerosol. To avoid chaining, the laser ablation cylinder is positioned within 5 cm upstream of the pulsed nozzle. The filament observed in Fig. 3 is planned so as to neutralize the nanocrystals which are expected to be photoionized by laser ablation. The first grid of the TOF unit can be biased so as to reject charged clusters.

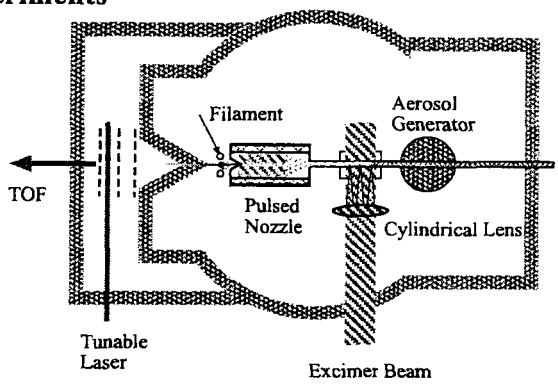

Fig. 3. Schematic of operation of supersonic jet apparatus for generation of semiconductor nanoparticles.

We have finished the construction of an apparatus to deposit nanoparticles on single, crystal, clean substrates held in ultra-high vacuum. The samples can then be moved into a second chamber where high band-gap semiconductors or glasses can be grown epitaxially by laser or ion beam sputtering. The apparatus has achieve pressures of 10^{-10} Torr, and neutralized ion-beam sputtering has been demonstrated. Samples of hydrogenated Si nanoparticles will be passivated by a high bandgap semiconductor (ZnO) or SiO_2 coating with ~10 nm thickness. In addition to the spectrocopy experiments above, single particle spectrocopy will then be completed on these new samples.

4.0 DOE Supported Publications

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2. Parminder S. Bhatia and John W. Keto, "Pressure and power dependence of the optically heterodyne Raman-induced Kerr effect line shape," *Phys. Rev.* **A59**, 4045(1999).
3. H.Htoon, Hongbin Yu, D. Kulik, J. W. Keto, O. Baklenov, A.L.Holmes Jr., B.G. Streetman, and C.K.Shih, *"Quantum dots at the nanometer scale: Interdot carrier shuffling and multiparticle states,"* Phys. Rev. B. **60**, 11026-11029(1999).
4. H. Htoon, J. W. Keto, O. Baklenov, and A. L. Holmes, Jr., C. K. Shih, "Cross-sectional Nano-Photoluminescence Studies of Stark Effects in Self-Assembled Quantum Dots," Appl. Phys. Lett.**76**, 700-702(2000).
5. H. Htoon, Hongbin Yu, D. Kulik, J. Keto, C.K. Shih, O.l Baklenov, and A. L. Holmes, "Nano-photoluminescence studies of self-assembled quantum dots," *Self-organized Processes in Semiconductor Alloys-Spontaneous Ordering, Composition Modulation, and 3-D Islanding*, (Material Research Soc., Pittsburg), 2000.
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7. W.T. Nichols, D.E. Henneke, G. Malyavanatham, M.F. Becker†, J.R. Brock, and J.W. Keto, and H. D. Glicksman, "Large scale production of nanocrystals by laser ablation of aerosols of microparticles," Appl. Phys. Lett. **78**, 1128-1130(2001).

Program Title:

"Ion/Excited-Atom Collision Studies with a Rydberg Target and a CO_2 Laser"

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Program Scope:

The program involves three related projects, all involving the interaction of multiply-charged ion beams with a Rydberg target.

1) Continued studies of charge transfer, aiming to reveal details about the L-distributions formed in the collisions.

2) Studies of X-rays emitted from the highly-excited Rydberg ions formed in these collisions.

3) Studies of the fine structure of high-L Rydberg ions, in order to extract measurements of dipole polarizabilities and quadrupole moments of the positive ion cores.

Recent Progress:

Project 1) During this year, we completed this study and prepared a paper reporting the result which will appear soon in Phys. Rev. A. Our study provided the first test of an unusual aspect of CTMC predictions of L-distributions in ion-Rydberg charge transfer collisions, which is illustrated in Fig. 1. It is predicted that the qualitative features of the L-distributions obtained in n-levels above and below the most probable n-level will be different, with the lower n levels being high-L rich, while the upper n-levels are high-L poor. Our study confirmed this prediction, showing remarkable agreement with CTMC predictions.

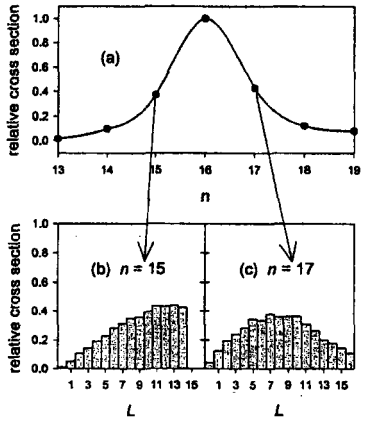

Fig. 1. Typical variation of L-distributions predicted by CTMC for two different values of principal quantum number n, for a q=3 ion on a 7F target at v=0.031.

Project 2) We made substantial progress on this project during the year. A major step was demonstrating that an external electric field noticeably modifies the X-ray spectrum emitted by both H-like and He-like Silicon ions, when they are formed by charge capture from the Rydberg target. After these initial observations, as often happens, a considerable effort was required to obtain reliable and reproducible measurements documenting this effect. We have now concluded that the major problem we encountered was associated with uncontrolled variations in the thickness of the Rydberg target, caused largely by variations in the conditions of the Rb oven that pumps it. These, in turn, caused secondary Rydberg collisions, which apparently changed the characteristics of the X-ray emitting populations. Having identified this problem, we are optimistic that we will be able to complete this initial study before the end of the calendar year. Our goal is to provide a measurement of the low-m fraction (m=-1, 0,1) in the population distribution formed in the charge transfer collision with the Rydberg target. Our initial data suggest a fraction of about 25%.

Project 3) No progress has been made towards obtaining a Th^{3+} beam at Kansas State University. In view of this, we have modified the immediate objective of this study. Our present plan is to carry out microwave spectroscopy of Rydberg states built on several different ionization states of Silicon, beginning with Si^{3+}, but including ions with charge ranging from 1 to 6. These studies will be feasible with the present ECR source at KSU. We hope they will provide a "proof of principal" demonstration of the capabilities of this technique that will help to justify the additional effort and expense that will be required to finally obtain Thorium and Uranium beams in the future.

In addition to these three projects, we have recently added a fourth. We are collaborating with Brett DePaola to extend his MOTRIMS studies, where he uses the COLTRIMs technique to monitor collisions between an ion beam and the atoms trapped and cooled in a Magneto-Optical Trap (MOT). Our contribution is to provide the additional lasers which can excite the Rb atoms in the MOT to higher excited states. As a first step in that direction, we added our 1529 nm laser which excited upwards out of the $5P_{3/2}$ state into the $4D_{5/2}$ state. The results of this test were very encouraging. A paper describing our observations is now in preparation.

Immediate Future Plans:

Project 2) We expect to complete the initial study of Stark-induced X-ray emission, which uses a field configuration applying an electric field along the beam axis. Our next step in this study will be to design and construct a device to apply an electric field at an arbitrary angle to the beam axis. This will be similar to the "Stark-barrel" design used by K.B. MacAdam recently. This device should enable us to extend the existing studies by providing a more complete data set. We plan to concentrate both phases of this experiment on H and He-like Silicon, but if acceptable beams of heavier ions are obtained from the CryEBIS source, this could provide attractive extensions.

Project 3) We are ready to begin microwave studies of Si^{q+} Rydberg states, and expect to make substantial progress during the next calendar year.

Project 4) We expect to refine the 2-laser experiments, exciting the MOT to the $4D_{5/2}$ state, by building a new frequency locking system for the 1529 laser which will enable us to carefully control the frequency of this laser. One objective of this improved study will be to elucidate the mechanisms responsible for increased MOT loss-rate when the 1529 laser is on. Later, we will add a third laser, the Ti-Saphire laser, which will excite the MOT up to nF states. One question which we need to answer immediately is whether this third laser results in an unacceptable background of ion production.

Recent Publications:

1) "Energy transfer in charge exchange collisions between slow ions and Rydberg atoms", D.S. Fisher, C.W. Fehrenbach, S.R. Lundeen, E.A. Hessels, and B.D. DePaola, Phys. Rev. Lett., 81, 1817 (1998)

2) "Energy transfer in Ion-Rydberg-atom charge exchange", D.S. Fisher, S.R. Lundeen, C.W. Fehrenbach, and B.D. DePaola, Phys. Rev. A 63, 052712 (2001)

3) " Experimental Studies of Resonant Charge Transfer from Rydberg states by Highly-Charged Ions", Physica Scripta, T92, 71-75 (2001)

4) "Experimental Studies of L-distributions from charge capture by Si^{3+} on Rydberg atoms", S.R. Lundeen, R.A. Komara, C.W. Fehrenbach, and B.D. DePaola, to be published, Phys. Rev. A (2001)

PROGRESS REPORT
PROJECT: ELECTRON COLLISIONS IN PROCESSING PLASMAS

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PROJECT DESCRIPTION

This project aims to develop and apply accurate, scalable methods for first-principles computational study of low-energy electron–molecule collisions. In particular, the goals are (i) to extend the Schwinger multichannel (SMC) method [1] to allow multiconfigurational descriptions of the target molecular electronic states and (ii) to improve the computational efficiency of high-level studies of elastic scattering by introducing compact representations of polarization effects. Because our focus in applications is on polyatomic molecules, for which calculations are highly numerically intensive, the code developed is designed to run efficiently on large-scale parallel computers.

PROGRESS

During the past year, we have focused primarily on completing the implementation of multiconfiguration target states within our scattering code. We are employing a formulation in which all states of interest are described within a compact set of configurations obtained from a calculation of the state-averaged, multiconfiguration self-consistent field (SA-MCSCF) type. Such a description allows us to capture not only the important qualitative properties of excited electronic states (and of the occasional ground states that are not well described at the single-configuration level) but also, in quantitative terms, a large percentage of the correlation effects omitted at the single-configuration level. However, because a comparatively small set of electronic configurations is employed, the approach is amenable to a distributed-memory parallel implementation, in that data describing the wavefunctions can be replicated across processors without placing heavy demands on memory or, worse, requiring the use of disk storage.

We have tested limited multiconfiguration cases in single-channel (i.e., elastic) calculations and are currently working to implement multichannel cases, which involves completion of computer code to handle the matrix elements arising from channel coupling. Code writing and debugging are proceeding normally, and we expect to proceed to running test cases soon.

During the year, we also received an equipment gift from Intel Corp. to upgrade the workstation cluster that we use as a virtual parallel computer for electron-molecule collision calculations. The upgrade increases the cluster size from 16 to 32 processors, with an increase from 300 MHz to 933 MHz in processor speed and roughly commensurate increases in aggregate memory and disk storage. In addition, Intel donated a quad-processor server with over 200 Gbyte of disk for use in large single-processor jobs as well as small multiprocessor jobs. We have spent some time in configuring hardware and software for the cluster, including the setup of a gigabit ethernet interconnect using three 8-port switches.

PLANS

Over the next few months, we will complete the implementation of the multiconfiguration description of excited states in our scattering code, which will also allow us to study elastic scattering from multiconfiguration ground states with polarization effects included. Some period of code assessment, debugging, and refinement will follow.

Looking further ahead, we anticipate taking two principal directions in our work over the next few years. First, we will apply the enhanced capabilities that we have developed to carry out studies of electron collisions with moderate-sized polyatomic molecules having both scientific and technological interest. The DNA base pairs form one such group of molecules; recent experiments [2] demonstrate that dissociative attachment of electrons causes single- and double-strand breaks in DNA and is therefore a mechanism for genetic damage by low-energy secondary electrons produced by ionizing radiation. Because dissociative attachment is mediated by shape resonances in the electronically elastic cross section, and because the resonance character (including its lifetime) will change as the molecule moves along the dissociation coordinate, studying the process in DNA base pairs will draw on just the capability we have been developing—that is, the capability to treat polarization effects efficiently within a scalable implementation, so that large-scale parallel hardware can be employed to make accurate calculations possible for molecules that would otherwise be out of reach. We will also continue our studies of polyatomic fluorocarbon and hydrofluorocarbon etchants of high interest in plasma-processing applications, as well as of fluorocarbon radicals that may be formed within plasmas, whose cross sections can be very difficult to obtain from experiment.

Second, we will undertake highly detailed studies of small molecules, taking advantage of the small electron count to employ more accurate wavefunctions and more extensive channel-coupling schemes. Target systems in this work will include H_2O, H_2, and N_2, all of which are of interest both in natural (atmospheric and/or biological) contexts and as often-studied prototypes. For molecules of this size, we will be able to carry out extended multichannel studies employing accurate representations of the target wavefunctions involved, and we will be able to repeat those calculations at numerous nuclear geometries in order to account for vibrational effects. We are collaborating with a group of experimentalists at California State University, Fullerton (M. Khakoo and coworkers) and at the Jet Propulsion Laboratory (S. Trajmar and coworkers) in this work, and we expect that the calculations and measurements will be mutually supportive. Preliminary calculations and the analysis of experimental data are currently under way for N_2.

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[2] B. Boudaïffa, P. Cloutier, D. Hunting, M. A. Huels, and L. Sanche, Science **287**, 5458 (2000).

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1998

"Electron Collisions with Nitrous Oxide (N_2O)," C. Winstead and V. McKoy, Phys. Rev. A **57**, 3589 (1998).

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"Low-Energy Electron Scattering by CH_3F, CH_2F_2, CHF_3, and CF_4," M. T. do N. Varella, C. Winstead, V. McKoy, M. Kitajima, and H. Tanaka, Phys. Rev. A (submitted).

"Electron Collisions with Tetrafluoroethene, C_2F_4," C. Winstead and V. McKoy, J. Chem. Phys. (submitted).

"Electron Transport Properties and Collision Cross Sections in C_2F_4," K. Yoshida, S. Goto, H. Tagashira, C. Winstead, V. McKoy, and W. L. Morgan, J. Appl. Phys. (submitted).

Ultrafast Coherent Soft X-rays: A Novel Tool for Spectroscopy of Collective Behavior in Complex Materials

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Program Scope

In this new project, nonlinear time-resolved spectroscopy of condensed matter with coherent soft x-rays is being attempted. The primary experimental objective is to obtain direct access to both femtosecond time resolution and mesoscopic length scale resolution. The length scale is defined through transient grating, or time-resolved four-wave mixing, measurements in which the grating spacing is on the order of the soft x-ray wavelength. The primary scientific objective is to measure both the correlation length scale and the correlation time scale, and to determine whether there is any direct association between them, for collective responses including structural relaxation in polymers, supercooled liquids, and other complex materials. and to determine whether there is any direct association between the length and time scales.

Femtosecond pulses at soft x-ray wavelengths are produced through high harmonic generation. [1] It is now possible to reach nanojoule energies in such pulses, sufficient for nonlinear time-resolved spectroscopy of condensed matter. Given the high spatial coherence and focusability of the output, intensity levels comparable to those typically used in condensed-matter femtosecond spectroscopy with visible pulses can be reached. Thus there is strong reason to hope for successful extension of spectroscopic methods now widely used in visible and nearby wavelengths to the soft x-ray regime.

In current time-resolved measurements of condensed-matter collective dynamics, the relevant time scales may be measured but the relevant length scales are rarely determined and often are not well understood. For example, structural relaxation in simple molecular liquids occurs in femtosecond/picosecond time scales and is only weakly temperature-dependent. In supercooled liquids, the responses are often highly nonexponential, extending over a wide range (several decades) of time scales even at a simple temperature and changing enormously (i.e. by many decades) as the temperature is cooled and the viscosity increases. [2] A great many

materials including synthetic polymers and biopolymers, molecular and ionic liquids, and some acqueous solutions show this behavior. In the case of polymers, it is tempting to associate time scales with length scales, i.e. to imagine that the faster components of structural relaxation correspond to the motions of end groups, somewhat slower components to small side chain motions, still slower components to polymer backbone rearrangements, and the slowest components to relative motions of whole polymer molecules. However, supercooled polymer and small-molecule or ionic liquids show very similar temperature-dependent dynamics, and in the latter cases there is no obvious hierarchy of length scales based on structural elements like those of polymers. How are we to understand the observed hierarchy of time scales? Current time-resolved measurements cannot resolve this issue since essentially all the important correlation lengths are far smaller than any length scale defined experimentally. In this respect the situation is similar to ultrafast measurements of molecules, but in the case of molecules we have extensive independent information about bond lengths and molecular potentials to guide our understanding of the dynamical responses observed. In the case of collective dynamics, we usually do not have such guidance concerning correlation length scales. Thus we do not know whether small or large groups of constituents are involved in different components of structural relaxation, or whether there is any consistent or important connection between length and time scales.

In transient grating measurements, the grating fringe spacing defines the length scale over which correlated dynamics are measured. [3] In familiar cases, the fringe spacing is used to define an acoustic wavelength or the length scale over which thermal or mass diffusion is measured. In such cases the observed dynamics can be associated directly with the experimentally defined length scale. This is the basis for transient grating study of a wide range of phenomena including thermal, compositional, collective vibrational and electronic dynamics. However, structural relaxation dynamics are associated with density or molecular orientational correlation lengths or polymer persistence lengths that generally extend over nanometers, not microns. Direct measurement of the length and time scales is still possible through transient grating experiments, but the light wavelength must be sufficiently short that fringe spacings on the order of the correlation lengths can be produced. This requires soft x-ray wavelengths, i.e. wavelengths on the order of tens of nanometers or shorter.

In the experiments planned, two coherent soft x-ray pulses produced through high harmonic generation will be crossed to excite grating patterns with nanometer wavelengths. Heating at the transient grating maxima will lead to thermal expansion and acoustic wave generation. A third coherent soft x-ray pulse will be used as a variably delayed probe, incident at the phase-matching angle for diffraction off the grating pattern. The time-dependent diffraction intensity is expected to show acoustic oscillations and thermal diffusion, in both cases associated with the grating spacing. From the acoustic responses, measured as functions of temperature and grating spacing (i.e. acoustic wavelength or wavevector), complex structural relaxation dynamics can be determined. The measurements should extend those made with visible wavelengths, yielding acoustic responses throughout much of the Brillouin zone. In particular, acoustic responses at wavelengths ranging from about 50 nm to 500 microns, corresponding to acoustic frequencies in roughly the 10 MHz – 100 GHz frequency range, will be characterized. The results, along with complementary optical measurements of slower structural relaxation dynamics, will yield relaxation dynamics in the 10 ps – 1 ms range. This is sufficient for description of complex structural relaxation dynamics at most temperatures significantly above the liquid-glass transition temperature. Crucially, the correlation length scales can also be

determined in the soft x-ray measurements. Association of these with relaxation time scales will be examined in detail.

Recent Progress

In the initial stages of the project, mask patterns for soft x-ray photolithography have been designed and are being fabricated by the group of Prof. H.I. Smith of the MIT Dept. of EECS. These are to be used for diffraction-based generation of transient gratings with 50-nm fringe spacings. A single mask pattern will be used for shadow imaging of the grating pattern in preliminary measurements. An interferometer consisting of two mounted and precisely aligned grating patterns will be used for diffraction of an incident pulse into two orders which are then recombined through diffraction to meet at the sample. The probe pulse may be incident on the same interferometric setup to generate variably delayed probe and reference pulses, the latter used for heterodyne detection of diffracted signals.

Independent efforts are under way to generate the two excitation pulses through two separate fiber-optic high-harmonic generation systems. In this case the two outputs will be directed to meet at the sample, at which grating formation will take place. This arrangement offers higher total intensity, but may prove less robust since the interference pattern may be more susceptible to fluctuations.

Measurements with optical wavelengths, using femtosecond pulses and pulse sequences to generate GHz-frequency acoustic responses, are under way to provide results that will be compared to those generated with soft x-ray pulses.

Future Plans

Transient grating measurements with soft x-ray wavelengths will be attempted during the coming year. Demonstation and optimization of the experimental method will be the first objectives. Key issues are successful generation of transient grating signals, determination of the responses of interest, and elucidation and understanding of any competing signals. Simple samples will be examined at first for the purpose of methodology development. Following this stage, systematic study of supercooled liquid structural relaxation dynamics will follow.

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Energetic Photon and Electron Interactions with Positive Ions

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Program Scope

The objective of this experimental program is to probe multielectron interactions in atomic ions using energetic beams of photons and electrons. In addition to precision spectroscopic data for ionic structure, quantitative measurements of cross sections for photoionization and electron-impact ionization provide critical benchmarks for the theoretical calculations that generate opacity databases. The latter are used to model hot, dense astrophysical and laboratory plasmas. Examples of special relevance to DOE interests include the Z pulsed-power facility at SNL, which is the world's brightest and most efficient x-ray source, and the National Ignition Facility under development at LLNL. Three significant milestones were achieved by this program during the 2000-2001 period. Absolute photoionization measurements were achieved for the first time at the Advanced Light Source, and an electron-cyclotron-resonance ion source was developed and implemented at ALS, facilitating the first photoionization measurements on multiply charged ions. In addition, a new electron spectrometer was calibrated and implemented the first coincidence measurements at UNR of the energies of Auger electrons from metastable states of ions.

Recent Progress

Photoionization of Ions

The major milestones during the 2000-2001 period were the absolute calibration of the ion-photon-beam (IPB) endstation, the development and successful implementation of an ECR multicharged ion source, and the first photoionization measurements by an independent investigator using the IPB endstation. Absolute photoionization cross-section measurements were completed for outer-shell photoionization of O^+, Ne^+, Ar^+, C^{2+} and Ar^{2+}. Photoionization of Ca^+ was studied in collaboration with A. Müller and S. Schippers of the University of Giessen, Germany, who were the first independent investigators to be awarded ALS beamtime to use the IPB endstation. This work constitutes part of a NATO-sponsored collaboration. The photoionization results are being analyzed in detail and compared with available theoretical calculations. One manuscript comparing experiment and theory for photoionization of metastable O^+ has been submitted to Physical Review Letters for publication and two other manuscripts are currently in preparation.

Figure 1 presents an example of the comparison between experiment and theory for photoionization of Ne^+. While there is general agreement between the absolute measurement and the 10-state Breit-Pauli R-matrix calculation [B. McLaughlin, unpublished, 2001], some significant differences are evident in the positions and magnitudes of the resonance features, and in the magnitude of the non-resonant cross section. In addition, K-shell photoionization measurements have been made recently for C^+, C^{2+} and C^{3+}, and L-shell photoionization cross sections have also been measured for Ar^+. A fourth paper is in preparation on the K-shell results for C^+. During 2000-2001, this contract supported wholly or in part the participation in this project of two UNR postdoctoral fellows (A.M.

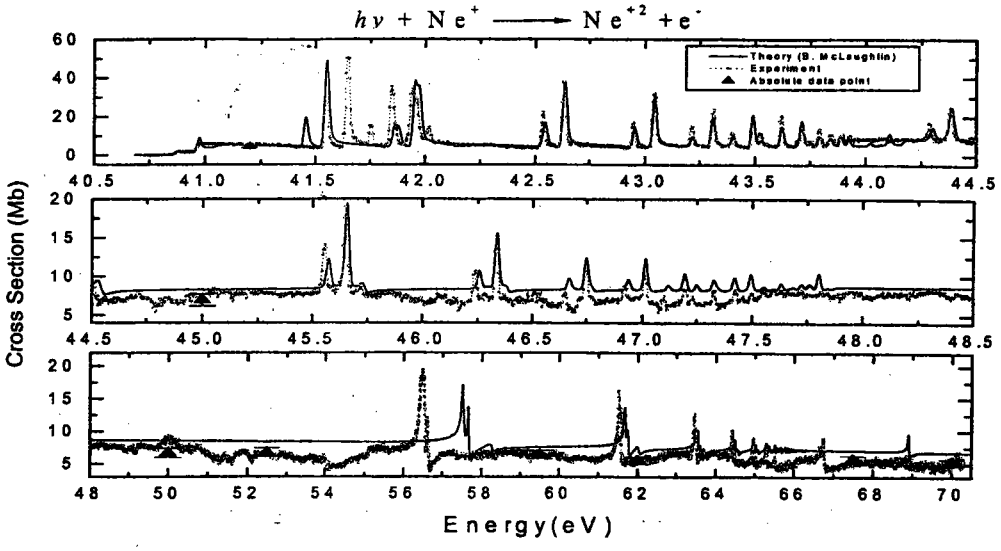

Figure 1. Comparison of absolute measurements and R-matrix theory of McLaughlin (unpublished, 2001) for photoionization of Ne^+ at a resolution of 22 meV.

Covington and G. Hinojosa) and two UNR graduate students (A. Aguilar and M. Gharaibeh).

Electron Collisions with Multiply Charged Ions

This project focused in 2000-2001 on the calibration and implementation of a high-efficiency electron spectrometer, and on development of an electron-ion coincidence technique to realize the full potential of the spectrometer for electron-ion collision experiments. Measurements were completed of the Auger electron spectra of the $2p^5 3s 3p\,^4D_{7/2}$ autoionizing metastable states of Na-like Ar^{7+}, Cl^{6+} and S^{5+}. The time-of-flight and electron energy spectra for Ar^{7+} are presented in Figure 2. The present measurements are believed to be the first for the autoionizing metastable states of Cl^{6+} and S^{5+}, and constituted the M.S. thesis of M. Lu, who was supported by this contract. A manuscript on this work is being prepared for publication.

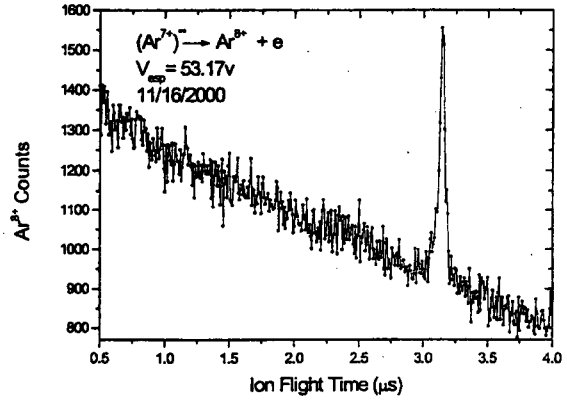

Figure 2. Time-of-flight spectrum for coincidence between electrons and Ar^{8+}.

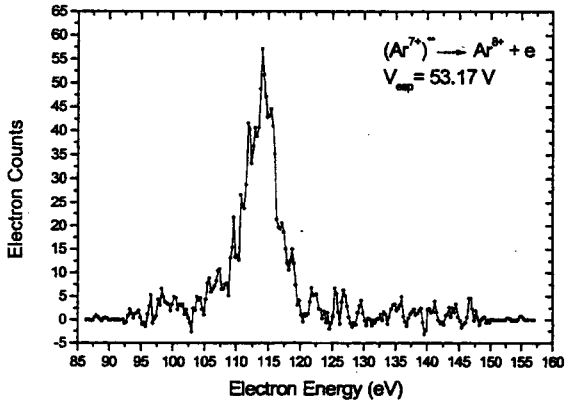

Figure 3. Electron energy spectrum for decay of the $2p^5 3s 3p\,^4D_{7/2}$ autoionizing metastable state of Na-like Ar^{7+}.

Measurements at the TSR storage ring in Heidelberg of the effects of combined external electric and magnetic fields on dielectronic recombination of Li-like Ni^{25+} were analyzed and published during the reporting period., as well as the results of a precision experimental and theoretical study of autoionizing resonances inelectron-impact ionization of Li-like O^{5+}.

Future Plans

Photoionization of Ions

Future research on photoionization will focus on systematic studies of photoionization of multiply charged ions along isoelectronic and isonuclear sequences. Further measurements of ions in the N-isoelectronic sequence (F^{2+}, Ar^{3+}) will complement initial measurements on O^{+} and constitute the Ph.D. dissertation research of A. Aguilar. Systematic measurements on ions of the Fe-isonuclear sequence (Fe^{2+}, Fe^{3+}, Fe^{4+} and Fe^{5+}) will constitute the Ph.D. dissertation research of M. Gharaibeh. Initial measurements on the broad $3p^5 3d^2\ ^2F$ autoionizing states of K-like Ca^{+} performed in collaboration with the Giessen group will be extended to Sc^{2+} and Ti^{3+}. Measurements on Na-like Mg^{+} and Al^{2+} are planned in collaboration with a group of collaborators from Aarhus University. Both experiments will require the implementation of an oven for producing metallic ions in the ECR ion source.

Electron Collisions with Multiply Charged Ions

Following the successful electron spectroscopy measurements of autoionizing metastable states of Na-like ions, the electron-ion coincidence technique will next be applied to 4d-nl electron-impact excitation-autoionization of ions in the Xe isonuclear sequence (Xe^{3+}, Xe^{4+}, Xe^{5+}), where multiple excitations with large cross sections are predicted. This will present a significant technical challenge, and will constitute the Ph.D. dissertation research of M. Lu.

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High-Angular-Momentum Rydberg Atoms in Magnetic Plasma Environment

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1 Physical system under investigation and research goals

We study the properties, the excitation and the decay mechanisms of high-angular-momentum (high-m_l) Rydberg states in clouds of cold atoms in strong magnetic fields, and the role these states play in magnetized cold plasmas. Since the diamagnetic interaction of free electrons and (quasi-free) Rydberg electrons causes strong anisotropy and alters the very nature of Rydberg states, such systems are expected to behave very different from Rydberg gases and cold plasmas in small magnetic fields.

In strong magnetic fields, high-m_l Rydberg states are highly unusual, as becomes evident already by a consideration of classical electron orbits (see Fig. 1). The nature of these orbits allows for an adiabatic separation of the motion into a cyclotron, a bounce and magnetron motion, similar to the separation procedure used for the motion of ions and electrons in Penning traps [1]. High-m_l Rydberg states in strong magnetic fields essentially correspond to an $\mathbf{E} \times \mathbf{B}$ drift motion of the Rydberg electron and are therefore unrelated to the well-known low-B circular Rydberg states, which correspond to Kepler orbits. At B-fields as large as ours ($\sim 5T$), in the anticipated relevant energy range -30meV< W <100meV most of the classically allowed phase space of the Rydberg electron is filled by high-m_l drift trajectories, such as the ones shown in Fig. 1. Due to this fact and the anticipated long radiative lifetimes of quantum-mechanical high-m_l Rydberg states, we expect Rydberg atoms in such states to be ubiquitous in near-equilibrium cold, magnetized plasmas. This is in contrast to beam experiments with Rydberg atoms, where optical selection rules and the presence of chaos in the low-m_l manifolds largely spoil efforts to excite high-m_l Rydberg states.

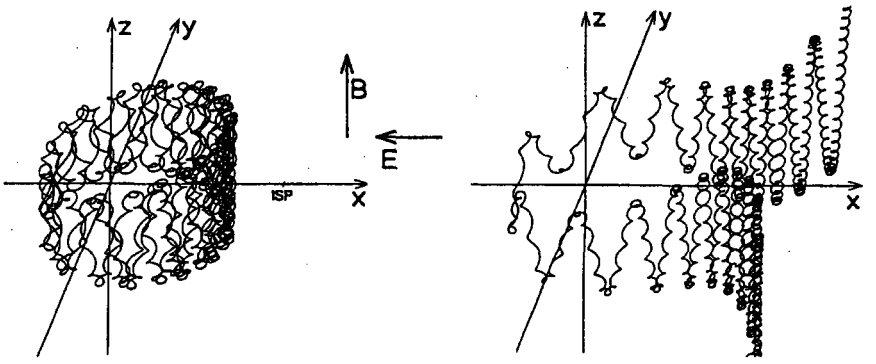


Figure 1: Bound (left) and free (right) high-m_l orbits (= drift orbits) of Rydberg electrons for $B = 5.63$T, $E = 58$V/cm, and an excitation energy of 41.2cm^{-1} below the field-free photo-ionization threshold of the atom.

To study cold, magnetized plasmas and the high-m_l Rydberg atoms in them, a superconducting high-B atom trap is being built. The atom trap has a B-field minimum of order 5T at its center. After loading the atom trap with a large density of rubidium atoms in the state $5S_{1/2}, m_s = 1/2$, the atoms will be excited by a pulsed dye laser into optically accessible low-m_l bound Rydberg states. Collision-induced state mixing will then promote the (chaotic) low-m_l states into long-lived high-m_l states. If the initial excitation is slightly above the ionization threshold, a cold plasma is produced. In this case, recombination should populate bound high-m_l Rydberg states. Recombination is known to play a role in non-magnetic cold plasmas [2] and has recently been observed in such plasmas [3].

Using a time-dependent electric field parallel to the B-field, the experimental system will be suitable to first extract the plasma components and to then field-ionize the remaining Rydberg atoms in a controlled manner. It will thus be possible to measure the formation of Rydberg states due to collisions, three-body

and radiative recombination. The lifetimes and state distributions of the produced Rydberg states will be determined by measuring the Rydberg atom population versus time and field ionization voltage. Later, it is planned to investigate the RF and microwave spectra of atoms in high-m_l Rydberg states. The spectra should reflect the three characteristic oscillation frequencies of the high-m_l-states (cyclotron, bouncing and magnetron oscillation). We also anticipate studies of plasma waves in photo-excited magnetized plasmas.

2 Relation of the work to other research

Currently a number of groups produce and study cold Rydberg gases and cold plasmas by photo-exciting clouds of laser-cooled and trapped atoms into Rydberg or continuum states. The scale size of all these systems is of order one mm, and typical numbers of Rydberg atoms and/or ion-electron pairs range from a few thousand to about 10^7. Depending on their electron density, electron temperature and applied magnetic field B, the cold plasmas that form in these systems fall into one of the following categories:

Low-B regime. The cyclotron radius of the electrons at their typical energy is larger than the plasma scale size. Typical magnetic fields are less than a few Gauss, and the magnetic pressure plays only a minor role. Cold, strongly coupled plasmas of the low-B case are currently studied at NIST, the University of Connecticut, the University of Virginia, and in my group.

Moderate-B regime. The average electron cyclotron radius is smaller than the plasma scale size but larger than the size of the Rydberg atoms present in the plasma ($B \sim$ 100Gauss). In this regime, it should be possible to achieve significant enhancements of the plasma lifetime and anisotropic plasma properties. The structure of the Rydberg atoms that are formed in the plasma will not be significantly different from the low-B case.

High-B regime (this work). In this regime, the electron cyclotron radius is smaller than the size of the Rydberg atoms present in the plasma. Both the plasmas and the Rydberg atoms are highly anisotropic. Due to the large magnetic pressure, the large depths of superconducting magnetic traps, and the low vacuum pressure in cryogenic UHV systems we expect long plasma trapping times. Most Rydberg atoms in the plasma should be in drift states, which resemble electron trajectories in Penning traps ([1] and Fig. 1). Cold, strongly magnetized plasmas should therefore be an ideal environment for the study of these unusual atomic states. We expect our research to be relevant to efforts elsewhere that aim at the formation of anti-hydrogen in high-B Penning traps. We also believe that long-lived states of positronium found in those experiments are closely related to the unusual Rydberg states studied in our project.

3 High-B atom trap

A high-B magnetic trap with a B-field minimum of order 5T forms the central part of our experiment. The system has been ordered in December 2000 and is since then under construction at AMI. At present, the magnet fabrication - the most critical phase of the construction - is finished and the magnet system is being tested. The magnet and dewar system may arrive in a month. Once delivered, the system will provide unique experimental opportunities for the study and the trapping of cold, magnetized plasmas and Rydberg atom gases.

The 4-coil magnet system, which emerged from combined designing efforts at AMI and in my group, contains a main split-coil dipole magnet that provides a $\sim 5T$ bias field at the trap center. A pair of racetrack-shaped coils inserted into the dipole magnet add a strong quadrupole field to the dipole field, thereby creating a Ioffe-Pritchard-type local minimum of the B-field, which acts as a magnetic trap for neutral atoms [4] and as a magnetic bottle for ions and electrons. An auxiliary center tap on the split-coil dipole magnet can be used to move the trap minimum in axial direction and to thereby overlap it with the geometrical symmetry point of the coil arrangement. The system provides six-axis optical access with two horizontal channels having a cross section of 1.9cm x 6.0cm and four channels having a cross section of 2.3cm x 3.6cm.

The cryostat is being built by Janis. The large access channels of the magnet end in 8-inch CF flanges that

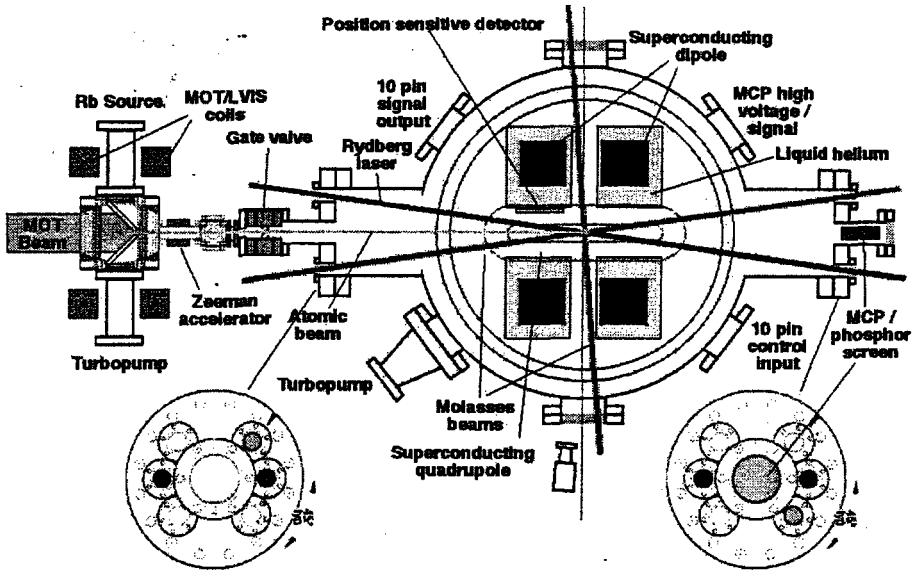
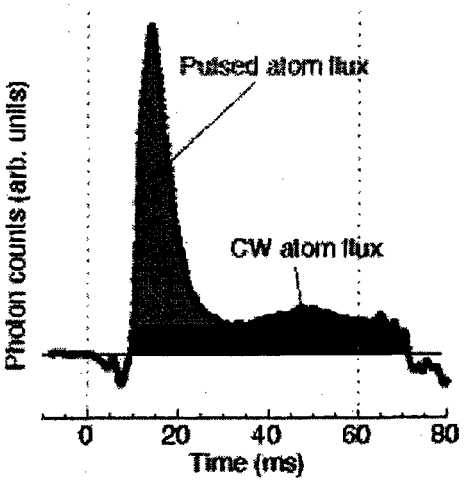


Figure 2: High-B atom trap with loading beam apparatus.

provide mounting space for electron and ion detection equipment, guiding and ionization electrodes, and an optional electron source. Planar 4K and 77K radiation baffles located in the access cones provide removable and modifiable mounting platforms for instrumentation and tubular radiation baffles for the laser beams. It will be possible to directly observe the trap from outside the cryostat; this will greatly simplify the alignment of the molasses lasers and the B-field minimum.



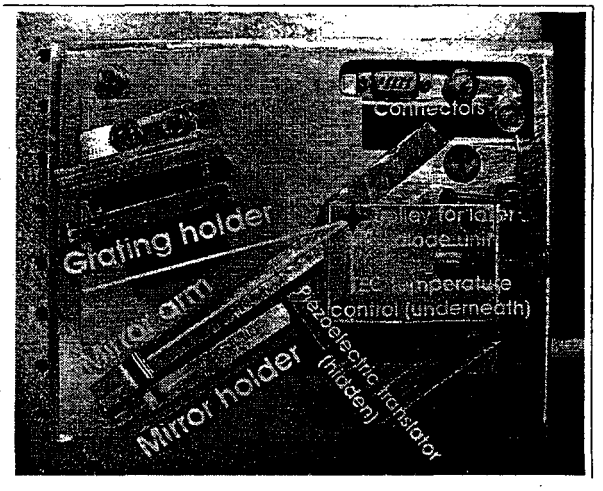


Figure 3: **Left:** Measured fluorescence of an LVIS atomic beam intersecting a probe laser. The LVIS is activated at time $t = 0$. The delay time and the shape of the resultant pulse (red) and the geometry of the setup allow us to determine the velocity distribution of the atomic beam. The level of the constant signal observed after about 30ms (blue) allows us to estimate the cw flux of atoms. **Right:** Photograph of the Littman laser that we use for the high-B optical molasses. Key parts of the setup are identified. Optical elements have been mounted after this picture was taken.

The high-B trap will be loaded with an atomic beam having about 30m/s velocity. The atomic beam is produced by an LVIS magneto-optic trap [5] and a short Zeeman accelerator, as shown in Fig. 2. The LVIS, which utilizes a pyramidal magneto-optical trap (MOT), generates a beam of atoms with ~ 10 m/s forward velocity and a flux of 10^9 to 10^{10} s^{-1}. The LVIS-MOT is located at a distance of 70cm from the center of the high-B trap. Since the fringe field of the high-B trap at the location of the LVIS-MOT still amounts

to $\sim$ 200Gauss, it was necessary to build a pair of water-cooled coils for the MOT operation. A typical fluorescence measurement of the atomic beam emitted by the LVIS-MOT is shown in Fig. 3. To obtain the required loading velocity of 30 m/s, a Zeeman acceleration region with a length of a few cm is placed between the LVIS and the high-B trap (see Fig. 2).

The slow atomic beam produced by the loading manifold will be trapped and cooled in the high-B-trap using a 6-beam optical molasses [6]. While in high-B there is no need for a molasses re-pumping laser, the molasses laser itself has to be stabilized to an uncomfortable frequency approx. 100 GHz away from the field-free rubidium D2 transition. The stabilization and the locking of the molasses laser is greatly simplified by the use of an external cavity setup that allows one to scan mode-hop-free over 100 GHz. Since Fall 2000, a graduate student has designed and built a Littman laser for this purpose (see Fig. 3). The main characteristics of the Littman design is that the planes of the mirror surface, the grating surface and the diode laser emitting surface, indicated by green lines in Fig. 3, intersect in a common pivot line (red star), which has to coincide with the effective rotation axis of the mirror arm. The type and the location of the piezo-electric translator have been chosen such that we should achieve a $\sim$ 200 GHz continuous scan range. We have ordered an anti-reflection-coated laser diode from Sacher for our Littman laser. The laser will be stabilized to an actively length-stabilized Fabry-Perot interferometer, which has also been constructed during the recent months.

4 Other aspects of the work

Team. A post-doctorial research associate (Jeffrey Guest) works full-time on the project since June 2000. A graduate student has been supported from this grant during summer 2000. A second graduate student, who is funded from a fellowship, has joined in Fall 2000.

Miscellaneous. For the MOT operation and other purposes, a few stabilized laser diodes in Littrow configuration have been set up. A complete pulsed YAG and PDL laser system has been modified such that it can be used to excite rubidium Rydberg atoms. A new lab section of approx. 500sf has been fitted with utilities and infrastructure for this project. We have moved from a temporary lab space into the new lab in December 2000. Ongoing efforts also include theoretical studies of transitions between high-m_l Rydberg states in strong magnetic fields, their radiative decay rates and their recombination cross sections.

The following publications acknowledge partial support by this grant:

"Ponderomotive Optical Lattice for Rydberg Atoms", S. K. Dutta, J. R. Guest, D.Feldbaum, A. Waltz-Flannigan, G. Raithel, Phys. Rev. Lett. 85, 5551 (2000).
"High-angular-momentum states in cold Rydberg Gases", S. K. Dutta, D. Feldbaum, A. Walz-Flannigan, J. R. Guest, G. Raithel, Phys. Rev Lett. 86, 3993 (2001).
"Tunneling resonances and coherence in an optical lattice", B. K. Teo, J. R. Guest, G. Raithel, submitted (2001).
"L-changing collisions in cold Rydberg gases", G. Raithel et al., to be submitted to the proceedings of ICPEAC 2001.
"Spectroscopy of Rydberg atoms in non-neutral cold plasmas", G. Raithel et al., to be submitted to the proceedings of the Workshop on Non-Neutral Plasmas 2001.

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"Quantum/Classical Atomic Interactions"

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Project Scope

There are two separate projects that are being investigated in this theory proposal. They are related in the sense that both explored issues of nonstandard scattering. In the first project, I study the stimulated attachment of an electron to an ion using a pulsed electric field; besides the inherent interest in understanding the physical process, it is possible that the method could be used to create high Rydberg states of complex molecules or to make anti-Hydrogen at CERN. In the second project, I study the interaction of quantum targets with matter beams containing some longitudinal coherence; this project addresses basic issues of scattering theory and how to use the beam coherence to extract new quantum parameters.

Recent Progress

In the first project, we recently published[1] the large version of the paper that describes how a ramped electric field can be used to stimulate the recombination of a free electron with an ion. Previous results were described in two short papers[6] before this project began. Reference [1] presents both new calculations and new experimental results. The experiments showed that the method worked in medium strength magnetic fields (60 mT) and when using trapped electrons. On the theory side, I show the regions of space that contribute to the recombination and give other parameters that cannot be directly obtained from experiment (for example, the energy distribution of the recombined electron).

In November 2000, C. Wesdorp attempted to use pulsed field recombination to create anti-Hydrogen at CERN in collaboration with Gabrielse's group. Calculations showed that anti-Hydrogen signal should have been observed for the parameters that were available in the experiment; this was not a simple extrapolation of previous calculations because the magnetic field was substantially stronger and the ramp rates were substantially slower due to the presence of filters. Despite the favorable prediction, it was judged that the data did not clearly demonstrate the presence of anti-Hydrogen. The suspicion is that the vXB motional electric field was stripping the Rydberg positron from the anti-proton. I am currently performing calculations which show there really isn't a vXB motional electric field for high Rydberg states: neutral particles cross magnetic field lines in a simple way only when the internal time scales are short compared to cyclotron periods. The results of the calculations predict that highly excited anti-Hydrogen atoms were not being stripped but were not leaving the magnetic field at the rate that was originally expected.

In the second project, there were four papers published on different aspects of quantum scattering using a beam that has longitudinal coherence. This project grew out of the short paper[7] that showed it might be possible to measure the

novel processes that occur when a quantum target interacts with a longitudinally coherent matter beam. The first paper[2] showed that the basic idea was correct by directly solving the time dependent Schrodinger equation for a model 3 electron problem; this showed that the predictions from the original formulation correctly predicted novel scattering processes. In particular, we showed that the predictions for multiple electron pulses interacting with a quantum target were quantitatively correct. The second paper[3] gave a full description of the theoretical formulation and presented a number of examples that seem to be within reach of current experimental technology. In particular, I showed that it is possible to cause transitions even when the beam misses the target; also, I showed that the longitudinal coherence could be used to control the excitation probabilities.

The two most recent papers elaborate the basic phenomena of scattering with a longitudinally coherent beam. Reference [4] shows how classical large scale phenomena emerge from the theory by applying perturbation theory. As an example, we show how the idea of the interaction of the target with the overall electric field of the beam emerges from the formulation. The calculations for Reference [5] were mainly performed by M. Ferrero, an *undergraduate* who worked with me during summer 2000. This paper describes the application of the pulsed electrons to electron molecule scattering; in particular, we showed that it is possible to obtain non-trivial molecular information by comparing a direct solution of the time dependent Schrodinger equation with approximations developed from the theory. We applied the formalism to an electron pulse interacting with a molecule in a vibrational wave packet; it is possible to obtain electronic excitation probabilities as a function of internuclear distance.

Future Plans

The prospects for the coming year are good for the theory side of the project. For example, I have plans to examine the behavior of Rydberg atoms in strong magnetic fields; in particular, the motion of the center of mass and whether a Rydberg atom will be ionized by a strong magnetic field will be investigated. This is a basic question in classical mechanics and could have implications for the measurement of anti-Hydrogen. If there are further attempts to create anti-Hydrogen using pulsed field recombination, then it will be important to understand how Rydberg atoms move across magnetic field lines. I also plan to simulate the motion of the Rydberg atoms through the exact electric field configurations used in one of the working experiments at CERN.

Experiments are being planned to study the interaction of a pulsed electron beam with Rydberg atoms. This will test the ideas we have proposed for the interaction of a longitudinally coherent beam with a quantum target. There is no need for further theoretical investigations until the experiments are at a more advanced stage.

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Development and utilization of bright tabletop sources of coherent soft x-ray radiation

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Program Description

Compact capillary discharge lasers and high order harmonic up conversion of ultrashort pulse tabletop lasers have both produced high brightness soft x-ray pulses [1,2]. These tabletop sources can provide intense beams of soft x-ray radiation for numerous applications. Their use time-resolved surface spectroscopy [3], material characterization [4], material ablation [5], characterization of soft x-ray optics [6] and plasma interferometry [7] has been demonstrated. Moreover, these compact sources can contribute to the development of techniques that are useful to the next generation of accelerator-based sources of coherent ultrashort wavelength radiation, in two respects. First, these sources may be used to seed future soft x-ray free-electron lasers. Second, they can provide very intense soft x-ray radiation pulses at a high repetition rate for the development of imaging and focusing techniques, and their use in applications.

In the proposed research, we are studying the amplification of picosecond HHG pulses in a 46.9nm collisionally-excited amplifier, and are exploring the techniques necessary for the application of these table-top sources of coherent short-wavelength radiation. A compact capillary discharge pumped amplifier based on the generation of a population inversion in Ne-like Ar, developed for this purpose at Colorado State University, will be combined with a HHG source developed at the University of Colorado. The amplification of XUV pulses in a capillary discharge amplifier presents its own unique challenges for the HHG source, which are of interest both for the application at hand as well as for understanding the HHG process itself. The efficient production of HHG "seed" pulses with optimum bandwidth at this wavelength will be attempted using a new technique of two-color high-order difference-frequency mixing. This HHG amplification scheme has potential for the generation of highly coherent XUV picosecond pulses of significant energy at multi-hertz repetition rates with a compact tabletop set up. Extraction of a fraction of the energy stored in a discharge pumped amplifier could produce pulses with energy several orders of magnitude larger than the HHG seed pulse, and with pulse width more than two orders of magnitude shorter than those of the unseeded discharge-pumped amplifier. Moreover, the experiments will provide a test-bed for the development of the techniques and understanding necessary to seed amplified spontaneous emission amplifiers with ultrashort high order harmonic pulses. The results obtained could impact the use of high order harmonic pulses as a seed for proposed x-ray free-electron laser amplifiers.

Figure 1 is a schematic representation of the harmonic amplification experimental set up. The high order harmonic pulse is generated using phase-matched frequency conversion in a capillary waveguide. The pulse is subsequently injected into a compact discharge pumped amplifier that produces gain at 46.9nm by electron impact excitation of the $3p\ ^1S_0$- $3s\ ^1P_1$ transition of Ne-like Ar. The realization of the

experiment requires: 1) The development of a new compact discharge-pumped amplifier tailored for the injection and amplification of "seed" harmonic pulses, using an open-end structure to allow the injection and extraction of the high order harmonic pulses, and using a femtosecond laser- triggered spark-gap. 2) The optimization of harmonic pulses at 46.9nm utilizing phase-matching and pulse-shaping techniques. 3) The temporal and spatial overlap of the high order harmonic pulses with the gain pulse in the discharge amplifier.

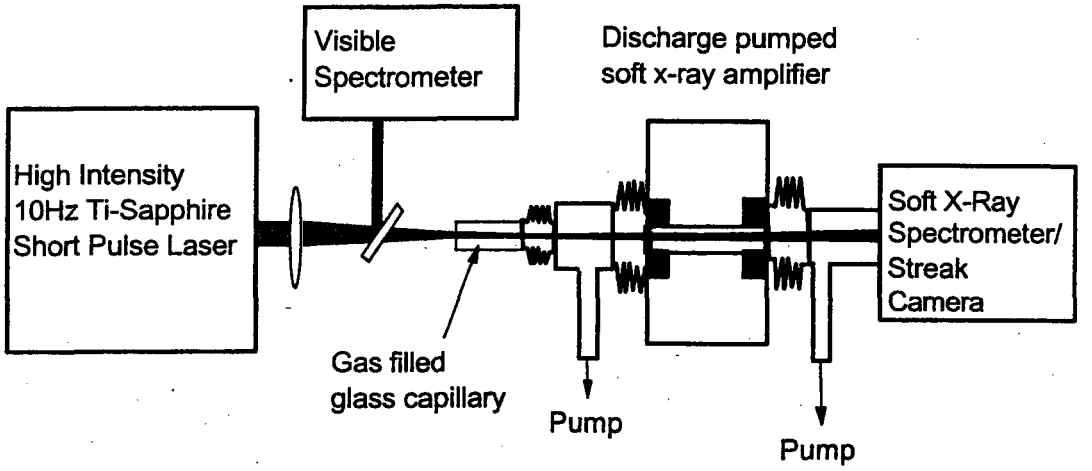

Figure 1: Schematic representation of set up for the amplification of high order harmonic pulses.

In a second set of experiments we plan to utilize these tabletop sources to demonstrate imaging and focusing techniques for the application of intense beams of coherent XUV radiation. For this purpose we are utilizing new high reflectivity Si/Sc multilayer-coating optics optimized for these wavelengths, to conduct high resolution imaging of materials surfaces.

Recent progress towards the amplification of high order harmonic pulses in a discharge pumped soft x-ray amplifier, and related work.

Progress achieved during the first year of research includes:

a. **Development of 46.9nm compact discharge amplifier for high order harmonic amplification:** A new capillary discharge Ne-like amplifier was designed and constructed for this purpose at Colorado State University. The amplifier is designed to provide a gain of approximately e^{10} at 46.875 nm, and to allow the free propagation of harmonic pulses along the axis. Gain has been observed, and tests of the amplifier performance and its optimization are currently underway.

b. **Demonstration of low jitter laser-triggered high voltage spark-gap:** The proposed experiment requires a precise synchronization between the gain pulse and the arrival of the harmonic seed pulse. To achieve this low jitter we have developed a high voltage spark-gap that can triggered by a femtosecond laser pulse of a few hundred microjoules of energy. Such trigger pulse can be extracted from the same Ti:sapphire laser amplifier that will be used to create the high order harmonic seed pulses. A first series of test of a prototype of this spark-gap showed a shot-to-shot time delay statistical fluctuations with a standard deviation as low as $\sigma = 0.1$ns. Our most recent tests with optimized laser trigger pulses have achieved a jitter of ± 0.04 ns for voltages of 10 KV [8]. It is expected that the spark-gap modifications necessary to operate it at the high power levels required to pump the capillary discharge amplifier will somewhat increase the jitter. Nevertheless, the concept of

femtosecond laser-triggering of high voltage spark-gap we have demonstrated is expected to meet the synchronization requirements for the amplification of high order harmonic pulses in the discharge amplifier.

c. **Optimization high order harmonic generation at 46.9nm**

The 46.9 nm wavelength corresponds to the 17th harmonic of a ti:sapphire laser; our preliminary investigations for this project have centered-around determining the optimum configuration for generating this harmonic. In recent work, we have determined that two-color high-harmonic generation unexpectedly gives a higher efficiency of conversion into the 17th harmonic than simple HHG when nonlinear frequency conversion is done in argon gas. We are in process doing a direct comparison of direct HHG using krypton gas, compared with two-color HHG in argon. We have also implemented temporal pulse-shaping on the laser system with which we are doing these investigations, and soon will be using this to investigate in detail methods for efficiently obtaining narrow-bandwidth pulses.

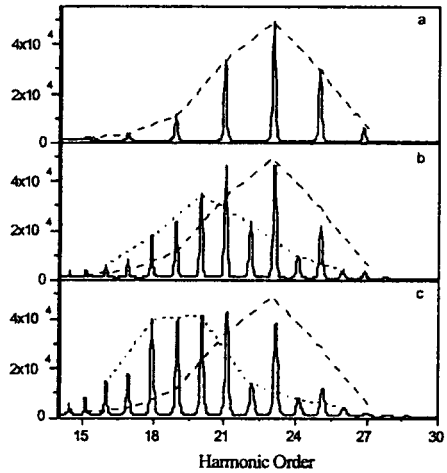

Figure 2: (a) Harmonic output when excited by 90μJ of fundamental (ω_1); (b), (c) when excited by a combination of 90μJ of fundamental and a weak 5μJ and 15μJ second harmonic field (ω_2), respectively. This data shows an unexpected shift of EUV energy to longer wavelengths that is attributed to four-wave difference frequency mixing driven by both the EUV high harmonics and by the 400 nm and 800 nm fields. This data demonstrates a new method for tuning and optimization of HHG radiation.

d) **Demonstration of essentially full spatial coherence in a high-average power table-top soft x-ray laser**

We have conducted an experiment that demonstrated for the first time an extraordinarily high degree of coherence in a soft x-ray laser beam, using a table-top discharge-pumped 46.9nm laser. [9]. A fast capillary discharge excitation was utilized to produce plasma columns with both very high axial uniformity and length to diameter ratio exceeding 1000:1. In such elongated plasma columns, rapid coherence buildup occurred as a result of gain guiding and strong refractive anti-guiding. This intrinsic mode selection mechanism makes it possible to achieve essentially full spatial coherence with the available plasma column length. A series of Young's interference experiments verified the rapid coherence buildup and show the evidence of approaching full spatial coherence with a plasma column length of 36 cm. This resulted in the generation of spatially coherent, milliwatt-level average power soft x-ray radiation from a tabletop device. Moreover the laser's peak coherent power is estimated to reach 6×10^4 W, within a spectral bandwidth $\Delta\lambda/\lambda \leq 1 \times 10^{-4}$. The simultaneously generating such high average coherent power and peak spectral brightness in soft x-ray region would open new opportunities in science and technology.

Future plans

Objectives for the second year of the project include the completing the development of the capillary discharge amplifier by integrating a low jitter laser-triggered spark-gap, and generation of on optimized seed pulse. In the second semester we plan to interface the high harmonic generation set up with the capillary discharge amplifier and detection system, and to conduct the initial high order harmonic amplification experiments. To realize this, the capillary discharge amplifier developed at Colorado State University will be installed in a laboratory at the University of Colorado. In addition, we plan to further advance the modeling of the amplification of the harmonic pulsed in the discharge pumped plasma column and develop an imaging system for use in combination with the coherent table-top soft x-ray sources.

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CORRELATED CHARGE-CHANGING ION-ATOM COLLISIONS

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I. PROGRAM SCOPE

Experimental investigations of atomic processes occurring in collisions of systems with few electrons are conducted. Additionally, photon interactions with molecules are studied. Major emphases are the study of the *electron-electron* interaction (i.e., *correlation*) and *multi-step* processes related to various collision phenomena, and the investigation of connections to photoinduced processes. Results are of interest from both fundamental and applied points of view. In addition to work at WMU, experimental work is carried out at the J.R. Macdonald Laboratory (Kansas State), the Advanced Light Source, and at GANIL (Caen, France).

II. RECENT PROGRESS

1. Formation of hollow three-electron atomic systems

Studies of double-K-shell-vacancy production in atomic Li and Li-like systems are investigated to determine the role and manifestations of the *electron-electron* (*e-e*) interaction in the two-electron transitions required to produce so-called "hollow" systems. The unique and simple structure of three-electron systems facilitates interpretation of the data and permits comparison with theory. Interactions leading to hollow states are generally expected to depend on the perturbation strength Z/v, and perhaps on v as well at relativistic velocities (v/c ~ 0.5). Measurements are being conducted over wide ranges of Z/v and v to elucidate the nature of the mechanisms that give rise to two K vacancies. Experimental work is carried out at WMU for intermediate-velocity (v ~ 10 a.u.) collisions, and at GANIL for high velocities (v ~ 0.5c) (in collaboration with Dr. N. Stolterfoht from the Hahn-Meitner-Institute, Berlin).

High-resolution Auger-electron emission spectra resulting from states with two K-shell vacancies in Be^+ and Li are shown in Figs. 1 and 2. Significant similarities between these

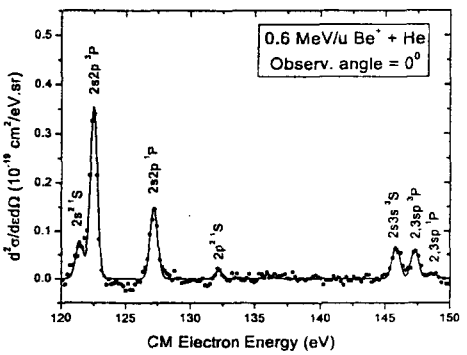

Fig. 1. Auger electron emission from Be double-K-shell-vacancy states formed in 0.6 MeV/u Be^+ + He (v/c = 0.04; Z/v = 0.4) collisions.

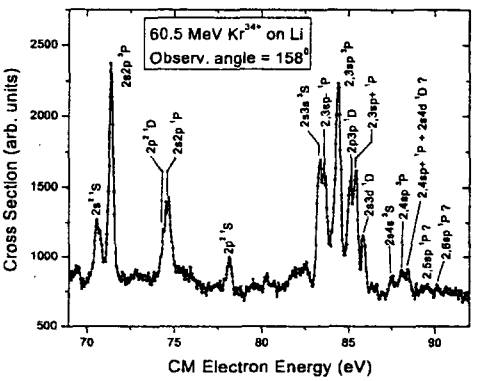

Fig. 2. Auger electron emission from Li double-K-shell-vacancy states formed in 60.5 MeV/u Kr^{34+} + Li (v/c = 0.3; Z/v = 0.7) collisions.

spectra are seen, and both spectra consist mainly of two-electron states, indicating that K-shell ionization followed by K-shell excitation is the dominant two-step process leading to the observed excited states. Analysis of these spectra indicates that the *e-e* interaction plays a significant role in the formation of the $2s^2\ ^1S$ and 2s3s ^3S states. On the other hand, the 2s2p ^3P and 2s2p ^1P states are attributed to a combination of *e-e* interactions and nucleus-electron (*n-e*) interactions. To control the correlation strength, additional measurements have been conducted at WMU for the Li-like ions B^{2+}, C^{3+}, and O^{5+}. These results can be compared with corresponding photoinduced spectra [1], where double-core excitation can only result from the *e-e* interaction. New measurements for N^{4+} ions and for Li atoms are planned at WMU.

2. Multicenter effects in the ionization of molecules

An important aspect of molecular ionization by ions and photons is the possibility for *multi-center* effects. These effects can lead to interference, orientation, or multiple scattering effects in the ejection of electrons.

An interesting aspect of molecular ionization is the possibility for interference in electron emission. Since the two atomic centers are indistinguishable, their contributions to ionization add coherently and interference effects might arise. Such interference, predicted by Cohen and Fano [2], is analogous to Young's double-slit experiment. Recently, we obtained evidence for interference in electron emission from H_2 caused by the impact of 60.5 MeV/u Kr^{34+} ions [3] as shown in Fig. 3. The measurements were conducted at the GANIL facility (Caen, France). The data and the calculations show a full sinusoidal-like oscillation in the emitted electron energy range up to ~250 eV. Further measurements for incident protons are in progress at WMU.

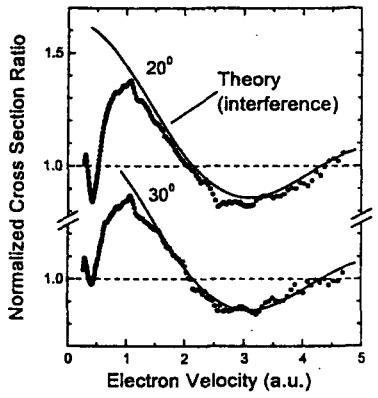

Fig. 3. Normalized (experiment divided by theory) cross sections for electron emission from H_2 produced by 60.5 MeV/u Kr^{34+} impact. The electron observation angles are 20^0 and 30^0, respectively. The solid lines represent model calculations of the present work (see Ref. [3]).

In addition to these interference studies, other work has shown that the initial orientation of a molecule relative to the incident ion can be a significant factor in determining the outcome of molecular ionization and fragmentation processes. Specifically, double-ionization and ionization-excitation of D_2 leading to dissociation are being studied by determining the complete momentum vectors of the resulting ion fragments in addition to those for one continuum electron. Results to date indicate that double ionization of D_2 by incident 1 MeV/u F^{8+} is more likely to occur when the molecular axis is oriented perpendicular to the beam axis. The reason for this effect is not yet understood. The experimental portion of this work is being conducted at Kansas State in collaboration with Profs. P. Richard and C.L. Cocke. A spectrometer is presently being constructed at WMU to expand these studies to new areas.

3. Superelastic electron scattering from metastable 1s2s ^{3}S states of low Z ions

A particularly interesting phenomenon occurs in electron scattering when electrons are scattered by an initially excited atomic system in such a way as to gain energy while simultaneously deexciting the parent system to its ground state. An electron that gains energy as a result of such an event is said to be *superelastically* scattered. Superelastic electron scattering can be considered as the time-reversed equivalent of inelastic electron scattering, i.e., electron impact excitation of ions, where the incident electron loses energy in the interaction.

Superelastic scattering of quasi-free H_2 electrons colliding with metastable 1s2s ^{3}S C^{4+} or O^{6+} has been investigated in measurements conducted at WMU. Here, the superelastically scattered electron gains several hundreds eV (the actual amount depending upon whether it is scattered from C^{4+} or O^{6+}), while the metastable ion is deexcited to $1s^2$ ^{1}S. The superelastic cross section is found to be about four orders of magnitude smaller than that for elastically scattered (i.e., binary encounter) electrons. Using the quasi-free electron approximation, *R*-matrix calculations for the time-reversed inelastic excitation process show good agreement with experiment.

4. Multi-particle continuum states from the photoionization of CO

A collaborative research effort, initiated by Dr. Allen Landers who is supported entirely by funds from this grant, to explore the dynamics of photoionization in atoms and molecules is also in progress. This work is being done at the Advanced Light Source (ALS) in collaboration with the Frankfurt (Germany) group of Prof. H. Schmidt-Böcking, the Kansas State group of Prof. C. L. Cocke, and M. Prior at Berkeley. The experiments utilize momentum-imaging techniques that measure the momentum vectors of all outgoing particles following photoionization. Recent measurements of the K-shell photoionization of CO reveal a richly structured electron diffraction pattern that can be correlated with a fixed initial orientation of the CO molecule [4].

III. FUTURE PLANS

As mentioned above, the primary emphasis of our research is the study of the *electron-electron* interaction and *multi-step* processes as related to the dynamics of various collision phenomena, and the connections to photoinduced processes. New work will be conducted at WMU, and in collaborative efforts with researchers at other universities and laboratories, thereby enabling us to carry out comprehensive and complementary investigations of fundamental and applied processes occurring in atomic collisions. Specific planned projects include: (1) production of doubly-K-shell vacant states in three-electron systems, (2) interference effects in the coherent emission of electrons from H_2, and (3) the dynamics of molecular ionization and fragmentation.

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