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TITLE: "STRUCTURE AND DYNAMICS OF ATOMS, IONS, MOLECULES, AND SURFACES"

INSTITUTION: Kansas State University
Department of Physics
J.R. Macdonald Laboratory
Manhattan, KS 66506-2604

Principal Investigator: Charles L. Cocke
Director, J.R. Macdonald Laboratory

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## A. STRUCTURE AND DYNAMICS OF ATOMS, IONS, MOLECULES, AND SURFACES

## A. Continuation Progress Report Overview

The joint experimental/theoretical program at the J.R.Macdonald Laboratory focuses on the dynamics of atomic and molecular systems in intense electromagnetic fields. The major emphasis of the laboratory has shifted in recent years from accelerator-based collisions work to ultrafast laser work and the interface between the two areas. There are many projects involving close experimental/theoretical collaboration, and involving group efforts between different experimental PIs. The individual projects are described in more detail by the individual PIs on the following pages. This summary is intended to give a brief overview of the laboratory activities of the last year and projection of the near future.

Current laser AMO projects include a joint theoretical/experimental (COLTRIMS) investigation of new mechanisms for multiple ionization of simple molecules and the use of the emerging understanding to probe the structure of the outer orbitals. Associated understanding of the dynamic alignment of the molecules in the pulse is also under study. Pump-probe experiments and analyses, including the use of Coulomb imaging, are being carried out to follow, in real time, heavy particle rearrangements which occur in the laser pulse. Harmonic generation from the interaction of the focused laser pulse with matter is under study. Polarization gating has been shown successful in generating for the first time a “super-continuum” of hard photons. Characterization of these “attosecond” pulses in real time is underway. As an entrance to “attosecond science”, theoretical analysis of possible experiments which could be done with these pulses is being carried out. For example, theoretical analysis of the time-evolution of auto-ionizing states in intense laser pulses and Stark-induced resonances is underway. The use of the harmonics as a “beam” of soft x-rays in time-resolved COLTRIMS experiments has been initiated.

The rapidly developing frontier of ultrafast science requires that the Kansas Light Source (KLS: the central Ti:sapphire laser facility in the JRM laboratory) be constantly upgraded and improved. Recent improvements include the development of routinely available 8 fs pulses, the installation of a new compressor and the installation of an OPA. Upgrade projects under way include the stabilization of the carrier-envelope phase and the addition of a second amplifier for this heavily subscribed facility. The laser runs reliably up to 24 hours a day, 7 days a week, typically feeding up to three parallel experiments.

In the ion-atom collisions area, fast highly-charged ions and protons are being used to dissociate water molecules and to study isotopic dependences of bond rearrangements, as well as ground state dissociation of HD. The long-standing expertise of the JRM laboratory is being used to study doubly and triply excited states of highly-charged fast projectile systems from our Tandem/LINAC, and theoretical analysis of up to quadruply excited systems is being carried out. In the area of low-energy collisions, investigations of mechanisms for dissociative capture from isotopically selected molecular hydrogen targets are being carried out and a new theoretical treatment of low-energy collisions using a hyperspherical-coordinate close-coupling method has been developed. MOTRIMS is being used to study momentum spectroscopy of electron capture from ground state and excited Rb with alkali beams, and extension to the use of highly charged beams from the EBIS is planned. This approach is also being used to study the dynamics of strongly coupled few-level systems in the clean environment of the MOT, a process with close connections to the fs-laser dynamics program, but on a longer time scale.

The historical expertise of the JRM laboratory in electron spectroscopy has been used in conjunction with the KSL facility to identify predicted but previously unobserved image-potential states formed above nanotubes. This work is a joint experimental/ theoretical study. Additional theoretical work in the surface/cluster area is being carried out on negative ions near conducting surfaces.

We are making heavy use of the unique availability of the KLS laser and ion beams in the same laboratory. Single electron removal from a true one-electron system, $H_2^+$, is being investigated experimentally using the ion beam from the ECR source interacting with the KLS laser pulse. Supporting theoretical work using lattice solutions to the time dependent Schroedinger equation, including a comprehensive treatment of all relevant processes, is addressing this most fundamental of all molecular systems. Extension of this approach to other molecules is also in progress.

A project to use the KLS beam to "picopulse" the Tandem is underway. The goal is to produce few-picosecond pulses of energetic heavy ions as a new tool in the ultrafast arsenal for studying real-time dynamics in a pump-probe arrangement. Proposed experiments include time-resolved heavy-particle diffraction and collision experiments with laser-modified/prepared targets. Related theoretical work on laser-assisted electron capture is being carried out. The transfer line which brings the KLS beam to the Tandem will also enable other direct laser experiments on the negative ion beam from our ion sources and on the fast ion beam from the accelerator.

While the JRM Laboratory is no longer an official BES User Facility, we continue to host many users. Steve Lundeen of Colorado State University maintains a very active program in the laboratory on x-ray production following capture by highly-charged ions from Rydberg targets and spectroscopy of highly ionized systems. Theo Zouros from Univ. of Crete maintains a program on electron-ion collisions and electron spectroscopy of highly charged systems. Our accelerator continues to serve both DOE and non-DOE users who use our beams for radiation damage studies. We continue to have a steady flux of collaborators from other institutions in many of the programs discussed here. In addition, members of the JRM laboratory continue to carry out research with collaborations at the ALS at LBNL and the Weizmann Institute.

In the coming year we expect to see a continuing shift of emphasis from pure ion-atom work to ultrafast dynamics using the KLS and to experiments which make special use of the availability of both ion beams and laser in the same laboratory. This work is almost always collaborative in nature, involving typically several faculty members and constantly involving interplay between experiment and theory.

## 1. Molecular Dynamics with Ion and Laser Beams

*I. Ben-Itzhak [ibi@phys.ksu.edu]:*

*The goals of this part of the JRML program are to study the different mechanisms leading to molecular dissociation and charge exchange following fast collisions, slow collisions, or interactions with an intense short laser pulse.*

**1. Dissociation and ionization of molecular ions by ultra-short intense laser pulses**, *I. Ben-Itzhak, P. Wang, J. Xia, A.M. Sayler, M.A. Smith, K.D. Carnes*, and *B.D. Esry*, – partly in collaboration with *Z. Chang*'s group, *C. Fehrenbach*, and *C.L. Cocke*. We have experimentally explored laser-induced dissociation and ionization of diatomic molecular ions, such as $H_2^+$, $HD^+$, $N_2^+$ and $O_2^+$, using coincidence 3D momentum imaging. Only a handful of experimental studies of intense ultrashort laser interaction with molecular ions have been conducted so far [J. Phys. Rev B 33, 2743 (2000); Phys. Rev. Lett. 85, 4876 (2000); Phys. Rev. A 62, 023401 (2000); Phys. Rev. Lett. 86, 5695 (2001); Eur. Phys. J. D 26, 39 (2003)]. The vibrationally excited molecular ion beam (4-8 keV), in our case, is crossed by an ultrashort intense laser beam (28-150 fs, $10^{13}$-$10^{14}$ W/cm$^2$). The resulting fragments are recorded in coincidence by a position-sensitive detector. Complete angular distribution and kinetic energy release maps are reconstructed from the measured dissociation-momentum vectors. Dissociation of the vibrational states around $v$=9 is notable in the low intensity measurements. The dissociating $H_2^+$ exhibits stronger alignment with increasing energy difference from KER 0.77 eV. However, lower KER values align along the laser polarization while the higher values are associated with alignment away from the polarization direction. Dissociative ionization was found to be smaller than dissociation in our measurements and increased with laser intensity and the alignment of the molecular axis with the laser polarization vector. The data is compared with recent calculations performed by *Esry*. These results will be presented as an invited talk in the upcoming International CAARI 2004 meeting. Previous intense laser work conducted at the University of Virginia (UVA) in collaboration with *R.R. Jones* and *E. Wells* was recently published [43], and a couple of other manuscripts were recently submitted.

Future plans: We plan to measure the dependence of ionization and dissociation of $H_2^+$, and other simple molecular ions, on the duration and intensity of the laser pulse.

**2. Isotopic effects in bond-rearrangement of water ionized by fast proton impact.** *M. Leonard, A.M. Sayler, P. Wang, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak*. Studies of ionization and fragmentation of water molecules by fast protons and highly-charged ions have revealed an interesting isotopic preference for H-H bond rearrangement. Specifically, the dissociation of $H_2O^+ \rightarrow H_2^+ + O$ is about twice as likely as $D_2O^+ \rightarrow D_2^+ + O$, with $HDO^+ \rightarrow HD^+ + O$ in between. Further investigations of this isotopic effect lead us to discover that bond rearrangement also occurs when the water molecule is multiply ionized, i.e. $H_2O^{2+} \rightarrow H_2^+ + O^+$, $H_2O^{3+} \rightarrow H_2^+ + O^{2+}$, etc. [14]. Calculations are underway to determine the relative production rates for the different isotopes from the overlap of the initial and final vibrational wave functions and the time evolution of the final wave function. These results were presented as an invited talk in the FIAC04 meeting in Hungary 2004.

Future plans: The isotopic enhancement in the $H_2O^{2+} \rightarrow H_2^+ + O^+$ dissociative double ionization channel requires further investigation to determine if it is similar in magnitude to that found in single ionization.

**3. Electron impact neutralization of small negative clusters.** *(I. Ben-Itzhak in collaboration with the group of D. Zajfman, at the Weizmann Institute of Science where I spent my Sabbatical year).* We studied electron detachment from small negative carbon and aluminum clusters by electron impact and photo-detachment. The clusters were cooled to their electronic ground state by storing them in an electrostatic ion trap and later interrogated either by an electron beam crossing their path inside the trap (*Heber et al*. accepted for publication in Rev. Sci. Intrum.) or by a laser beam overlapping their path. The measured electron impact cross sections exhibit unexpected dependence on the number of atoms in the linear carbon clusters, which we attributed to the increasing polarizability of these clusters. In contrast, the aluminum clusters followed the expected dependence on the binding energy of the loosely bound electron [44]. These findings were presented in a few international conferences this summer. Another main effort I was involved with was the construction of an experimental setup for studies of the interaction of molecular ions with ultrashort intense laser pulses (similar to our *JRML* setup) in collaboration with the laser group of *Yaron Silberberg*. First tests of this system are underway.

Future plans: We plan to continue our collaborative studies of (i) electron impact and photo-detachment of small negative clusters, and (ii) investigate the effect of the laser pulse shape on the ionization and dissociation of small molecular ions.

**4. Molecular dissociation imaging of collision induced dissociation and dissociative capture in slow $H_2^+$ + Ar (He) collisions.** *M. Leonard, A.M. Sayler, P. Wang, I. Ben-Itzhak, K.D. Carnes.*

Future plans: After a year break we plan to renew our studies of dissociative capture (DC, e.g. $H_2^+ + Ar \rightarrow H + H + Ar^+$) and collision induced dissociation (CID, e.g. $H_2^+ + Ar \rightarrow H^+ + H + Ar$) collisions. Our previous work indicated that CID is caused either by electronic or vibrational excitation. As in the past, the 3D molecular dissociation imaging technique will be used in our measurements. However, improvements to the experimental setup are underway in order to allow the detection of the recoil ion in addition to the molecular fragments, thus providing kinematically complete information about processes resulting in target ionization. In addition, the energy resolution should improve in the new setup. These measurements will be conducted focusing on the effect of the target species and the collision energy. Our preliminary results indicate significant differences in vibrational CID between Ar and He targets. Furthermore, we plan to investigate both these processes for a few additional simple molecular ions, such as $HeH^+$, $He_2^+$ and $H_3^+$.

**5. Ground state dissociation of $HD^+$.** *E. Wells et al. (Augustana College)* in collaboration with *I. Ben-Itzhak* and *K.D. Carnes*. An improved apparatus for 3D imaging of the slow $H^+$ and $D^+$ fragments from single ionization of HD was tested recently, and preliminary measurements of the angular distribution of single ionization by fast proton impact were conducted.

Future plans: We plan to continue these measurements and determine the dependence of pure single ionization on the angle between the projectile velocity and the molecular axis. In addition we plan to study very slow (a few meV) $H^+$ + D (1*s*) "half" collisions [Phys. Rev. A 67, 032708 (2003)]. In particular, we will investigate (1) the threshold behavior of electron transfer and (2) the Feshbach resonances in elastic scattering below the electron transfer threshold.

## 2. Attosecond Pulse Generation and Femtosecond Streak Camera Development

*Zenghu Chang [chang@phys.ksu.edu]:*

**1. Attosoecond supercontinuum generation and characterization,** *Bing Shan, Ghimire Shambhu, Jiangfan Xia and Zenghu Chang.* In the past year, we demonstrated experimentally that a supercontinuum covering the plateau and cutoff region is generated when a driving laser pulse with a time-dependent elliptically is composed by a few cycle pulses [10a]. The broad spectral should support much shorter pulses than that produced with the linearly polarized laser. In the experiments, the output beam from *Kansas Light Source* laser system was focused into a hollow-core fiber. The pulse passed through two pairs of chirp mirrors to be compressed down to 8 fs. The pulse with a time-dependent ellipticity was then produced with a delay of 15 fs induced by a 0.5 mm quartz plate. Finally the pulse was focused by a parabolic mirror (f = 250 mm) into an argon gas jet. The pulse energy was ~ 260 μJ which yielded an intensity of $\sim 1.5\times 10^{14}$ W/cm$^2$ on the target for the linear portion of the pulse. The high-harmonic spectrum showed a supercontinuum from 25 nm to 40 nm, corresponding to the harmonic order of 19 to 31. The supercontinuum yielded a single attosecond pulse with the transform limited pulse duration of 190 attoseconds. To the best of our knowledge, this is the first time that a supercontinuum was obtained by using polarization gating.

We plan to study the effect of the carrier-envelope phase on the supercontinuum generation. The phase of the *Kansas Light Source* laser system will be stabilized, which is a major upgrade. Our theoretical studies indicate that the supercontinuum is generated when the phase is 90 degree, which corresponds to the generation of two attosecond pulses. When the phase is zero degrees, discrete harmonic peaks appear, consequently two pulses are generated [10a]. We are working on the measurement of the duration of the attosecond pulse generated with the polarization gating. An apparatus based on measuring the angular distribution of the photoelectron in a circularly polarized laser field is under construction. We should be able to retrieve the attoseccond pulse duration in the similar way as the Frequency–Resolved Optical Gating technique widely used for femtosecond lasers.

**2. Approaching 100 fs resolution with an accumulative x-ray streak camera,** *Mahendra Shakya and Zenghu Chang.* In the past year, we improved the camera resolution from 580 fs to 280 fs by confining the electron beam size with a variable slit in the streak tube [11a]. The 5 μm slit is located in front of the electrostatic lens. To the best of our knowledge, the 280 fs is the best resolution ever achieved with a camera operating at the accumulation mode. We believe that the deflection aberration is not the limiting factor anymore for the camera with the slit. Rather it is the transit-time dispersion that limits the time resolution. We plan to use curved-optical-axis design to compensate the transit-time dispersion in order to improve the camera resolution to 100 fs. The photoelectrons with high initial velocity traverse longer distance than the ones with slower velocity. The camera will be tested with a femtosecond x-rays from a high-harmonic generation setup under developing.

## 3. Atomic Physics with Ion Beams and Synchrotron Radiation

*C.L. Cocke [cocke@phys.ksu.edu]:*

**1. Double ionization of small molecules by short intense laser pulses,** *A S. Alnaser, S. Voss, X.-M. Tong, C. Maharjan, P. Ranitovic, B. Ulrich, B. Shan, Z. Chang, C.D. Lin and C.L. Cocke.* During the past two years we have used COLTRIMS techniques to study two recently discovered processes for double ionization of molecular hydrogen by intense laser pulses: rescattering ionization (RS) and sequential ionization (SI) with ultrashort pulses. We used laser pulses from the Kansas Light Source (KLS) with pulse durations between 8 and 35 fs, to reach peak intensities between 1 and 12 x $10^{14}$ w/cm$^2$. Our major results are:

(a) Rescattering ionization dominates at the lowest intensities, is relatively weakly dependent on the angle ($\theta$) between laser and molecular axis. Peaks in the kinetic energy release spectra caused by several sequential returns of the first electron to the molecular core are seen in the data, showing that the electron bursts can be used as an fs clock for timing the evolution of the nuclear wave packet. Suppression of repeated returns is achieved with the 8 fs pulse. (b) At high intensity and short pulses, SI dominates. Again, the energy release is used to determine the time between emission of the first and second electron, showing the operation of a different but similar fs clock. (c) By adjustment of the pulse length and laser intensity, both of these processes, as well as the well established enhanced ionization, can be revealed as clearly distinguishable and separate processes in a single momentum spectrum. (d) In collaboration with C.D. Lin and X.-M. Tong, a full model for the RS and SI processes has been constructed [3, 8, 39, 41, 6a] which is in very good agreement with the experimental data and shows that understanding of these mechanisms has been brought under very good control. The results of the experimental work are described in publications 7, 35 and 7a.

Armed with this understanding, the COLTRIMS measurements have been extended to nitrogen and oxygen molecules. Unlike nitrogen, oxygen has been observed to display suppressed ionization and an extended harmonic spectrum, a widely discussed effect which has been associated with the different symmetries of the outer orbitals of these molecules. By studying the momentum distributions of coincident singly-ionized fragments from these molecules we have found that: (a) The fragmentation proceeds through well-defined and identified states of the dication, a previously undocumented result in laser fragmentation of molecules; (b) For low intensities the dependence of the ionization rate on the angle between polarization vector and molecule is very different for oxygen and nitrogen and maps out the shape of the outer orbital of the most loosely bound electron in each molecule. This result is in good agreement with the molecular ADK theory of Tong and Lin; (c) For higher intensities and long pulse lengths, the effect of the (post-collision) alignment on the ions is large, tending to send the fragments along the laser beam. This result is consistent with a model of Tong and Lin, and shows the importance of alignment not only prior to but during the fragmentation process. These results are discussed in publications 7a and 8a.

**2. Picopulsing the Tandem,** *Z. Chang, K. Carnes, V. Needham, C.L. Cocke.* We are implementing the production of (sub) ps pulses of fast heavy ions from our Tandem Van de Graaff accelerator. Intense laser pulses from the KLS will be focused into the source gas in the high-energy column of our 7MV EN Tandem to produce ions pulses with very small longitudinal and transverse emittance. Preliminary calculations indicate that, by choosing the appropriate ratio between acceleration and drift length and by keeping the number of ions in each pulse sufficiently low, pulses of MeV ions with pulse lengths in the several ps region can be delivered on target. We explore the possibility that these pulses can be used in a pump-probe arrangement to study physical processes on a ps time scale. Experiments planned include time resolved heavy-particle diffraction for the probing of phase transitions in solids, laser assisted collisions and collisions with transiently modified targets.

**3. Photoelectron diffraction from small molecules** This project now encompasses two projects, one at the ALS (a) and one at KSU (b).

**a. Synchrotron radiation:** *T. Osipov, A. Alnaser, P. Ranitovic, C. Marhajan, B. Ulrich, C.L. Cocke (KSU), A. Landers (Auburn Univ.), R. Dörner, Th. Weber, L. Schmidt, A. Staudte, H. Schmidt-Böcking, et al. (U. Frankfurt), M.H. Prior (LBL).* At the Advanced Light Source, we have measured the correlated momentum-space distributions of photoelectrons and charged photofragments from energetic photons on small molecules using COLTRIMS. This work is carried out by a large multi-laboratory collaboration involving the University of Frankfurt, LLBL, Auburn Univ. and KSU. During the past year, emphasis has been on molecular hydrogen where both dissociative ionization and double ionization processes have been studied [6, 26, 27 and 36].

**b. Harmonic generation source:** *P. Ranitovic, A. Alnaser, C.L. Cocke, B. Shan, Z. Chang*. Using harmonics generated from a gas jet illuminated by pulses from the KLS at KSU, we have initiated a project to measure photoelectron angular distributions from "fixed in space" molecules. We used soft x-rays in the 10-80 eV range to ionize "inner" valence electrons from the target in a modified COLTRIMS geometry. The goal is to perform time-resolved photoelectron spectroscopy from small molecules, in a pump-probe arrangement, with the infrared KLS beam as the pump and the x rays as the probe.

Future plans: We will continue to use COLTRIMS to explore to what extent the mapping of the outer orbitals seen in oxygen and nitrogen is generally successful for other molecules. We will explore the use of the fs clock by extending our measurements to include electron momentum measurements in coincidence with the ions. We will develop the picopulse facility: the transfer line from the KSL to the Tandem, the mechanism for mounting the mirror inside the terminal and the target chamber are under construction. First tests are expected in fall 2004. We will develop the use of soft x-rays from harmonic generation as a means to doing time-resolved photoelectron spectroscopy.

## 4. Collision and Coherent Excitation Dynamics in Atomic Systems

*Brett D. DePaola [depaola@phys.ksu.edu]:*

**1. Charge transfer cross section measurements, differential in angle and initial and final states**, *H. Nguyen, R. Brédy, H. A. Camp, T. Awata, and B. D. DePaola.* We have used the MOTRIMS methodology to measure charge transfer cross sections between ground- and excited state Rb (5s, 5p, and 4d) with a variety of singly charged ions of interest. In all cases the cross sections are differential in initial and final state. Each of the charge transfer channels was measured differential in scattering angle as well. Rather than attempting to catalogue a complete database, we concentrated on systems of special interest, such as the symmetric system: $Rb^+ + Rb(5s) \rightarrow Rb(5p) + Rb^+$ and $Rb^+ + Rb(5p) \rightarrow Rb(5s) + Rb^+$. Systems such as these provide valuable information on, for example, the validity of semi-classical calculation approaches such as the Demkov model. The results of these measurements have either been published [32, 33, 34] or are in preparation for publication.

**2. Dynamics of coherent excitation in a 3-level ladder system**, *H. A. Camp, M. A. Gearba, M. Shah, M. Trachy, B. D. DePaola.* We have used the MOTRIMS methodology to measure the time evolution of the population of states in a 3-level ladder system undergoing resonant, coherent excitation. We have focused our attention on the STIRAP (stimulated Raman adiabatic passage) prescription for coherent excitation of $^{87}Rb$ from the ground state to the 4d state, by-passing the intermediate 5p state. The essential idea behind the methodology is that the relative areas of the peaks in the Q-value spectra for charge transfer collisions between $Na^+$ and Rb(*nl*) are proportional to the products of the relative populations of the initial states and the cross sections for capture from those states. Using previously measured cross sections for capture from the 5s, 5p, and 4d states by 7 keV $Na^+$, as in section 1 above, we have been able to measure the relative populations of these states as they evolve on the nanosecond timescale. Because the populations of all three levels are measured simultaneously and as functions of time, we can readily distinguish between adiabatic and non-adiabatic excitation, the two cases having different temporal dependencies. Details of this work are being written up for submission to *Nature*.

**3. Dependence of excited-state fraction on the detuning of the trapping laser in a MOT**, *H. A. Camp, M. A. Gearba, M. Shah, B. D. DePaola.* We have used MOTRIMS to directly measure the fraction of atoms in the 5p state in a $^{87}Rb$ MOT as a function of the detuning of the trapping laser. Unlike previous measurements which have relied on fluorescence, the MOTRIMS methodology allows us to make model-independent measurements of excited-state fraction, in the same manner as described in section **2)** above. Comparison of these measurements with simple (previously *assumed*) excitation models will very helpful to the cold atom community which uses fluorescence as the principal diagnostic in determining the number of atoms in their MOTs. A manuscript containing the details of the results of these experiments is under review for publication.

**4. Three-dimensional Spatial imaging in multi-photon ionization rate measurements**, *R. Brédy, H. A. Camp, H. Nguyen, T. Awata, B. Shan, Z. Chang, and B. D. DePaola.* A consistent problem in measurements of above-threshold ionization is that the measurements are, to some extent, integrated over a range of laser intensities, due to the focal properties of the beam. The problem is exacerbated because above-threshold ionization is a non-linear function of intensity. Previous attempts to reduce this source of measurement uncertainty have included using time-of-flight or aperturing to reduce the integration to 1 or two dimensions. In our approach, our momentum recoil spectrometer was operated in *spatial imaging* mode. The spectrometer was thus converted into an ion lens (in 2-dimensions) and having a magnification of about 5. This, together with the flight time of the ions yields true 3-d spatial imaging with magnification. Details of the technique, including direct measurements of the lens parameters and limits on the system imposed by target temperature are described in publication [2a].

**5. Associative ionization and Penning ionization in a MOT**, *H. A. Camp, M. A. Gearba, M. Shah, M. Trachy, B. D. DePaola.* Several research groups have noted loss of Rb atoms from MOTs upon excitation to the 5d state. In the past it was assumed that the dominant process by which ionization took place was photo-ionization of the 5d. In the KSU MOT we have excited the Rb atoms to the 4d state and noticed similar trap loss. In the case of Rb(4d), however, 1-step photo-ionization is not energetically possible. We have begun a series of experiments in which it was discovered that the dominant processes for trap loss were associative and Penning ionization. By using a combination of time-of-flight spectroscopy and MOTRIMS, we have begun to measure the relative cross sections for several separate associative ionization and Penning ionization channels.

Future plans: Besides finishing off projects 2 and 5 above, we will expand the use of MOTRIMS to include studies of photo-ionization. In particular, we will generate short pulses of 3$^{rd}$ harmonic light from the 800 nm fundamental coming from the KLS. We will then measure relative single photon ionization rates of cold trapped Rb in the 5s, 5p, and 4d states. We will start with unaligned Rb and then proceed with photo-ionization measurements of laser-aligned Rb. For these latter experiments, we will use narrow line width, cw lasers to optically pump the Rb into aligned states. Using MOTRIMS techniques, we will measure the photo-ionization rate as a function of the angle between the target quantization axis and the polarization axis of the 3$^{rd}$ harmonic.

## 5. Time-Dependent Treatment of Three Body Systems in Intense Laser Fields

*Brett Esry [esry@phys.ksu.edu]:*

The primary goal of this program is to understand the behavior of $H_2^+$ in an intense laser field. Because the system is so complex, past theoretical descriptions have artificially reduced the dimensionality of the problem or excluded one or more important physical processes such as electronic excitation, ionization, vibration, or rotation. One component of this work is thus to systematically include these processes in three dimensions and gauge their importance based on actual calculations. Further, as laser pulses get shorter and more intense, approaches that have proven useful in the past may become less so. A second component of this program is thus to develop novel analytical and numerical tools to describe $H_2^+$. The ultimate goal is to understand the dynamics of these strongly coupled systems from first principles.

Future Plans: Our calculations to date have included a simple grid method to solve the time-dependent Schrödinger equation that was relatively inefficient. These calculations included three degrees of freedom -- two for the electron and one for the nuclei. Nuclear vibration and electronic excitation and ionization were thus included. We are in the process of adapting coordinate scaling methods to these calculations, making them more numerically stable and accurate. In fact, we have just submitted a paper describing the results to Physical Review A. In the next year, we will look for better spatial representations to further reduce the computational burden. With a more efficient code, we can follow up on our initial studies of carrier-envelope phase difference effects more easily [3a]. We can, for instance, explore the initial vibrational state dependence, eventually combining the results weighted by the Franck-Condon factors from the H2 ground state as is appropriate for molecular ion beam experiments. To complete the comparison with experiment, several intensities can also be calculated, allowing an intensity average to be performed. More differential information, such as the nuclear kinetic energy distribution, will also be calculated.

Since it includes ionization, the above code is most appropriate for intensities above about $7\times10^{13}$ W/cm$^2$. For lower intensities where dissociation is the dominant channel, solving the time-dependent Schrödinger in the Born-Oppenheimer representation is a more efficient approach. We have developed such a code and used it to carry out extensive calculations to describe the experiments from I. Ben-Itzhak's group. The dependence of the dissociation probability on peak intensity, pulse length, molecular orientation, and initial vibrational state has been studied. The data will be analyzed, compared with experiment, and published in the coming year.

The combination of Born-Oppenheimer and grid methods allows us to treat $H_2^+$ for all laser intensities accessible in our lab with some limitations. We will work to reduce these limitations in the next year and will work closely with I. Ben-Itzhak's group to help keep the theoretical work relevant to experiment.

## 6. Theoretical Studies of Laser-Atom, Laser-Molecule Interactions and Ion-Atom Collisions

*Chii-Dong Lin [cdlin@phys.ksu.edu]:*

**1. Ionization of molecules by short laser pulses: D**uring the past year, we have examined the ionization of molecules by intense laser pulses where the kinetic energy spectra of the fragmented ions have been determined. In particular, theoretical modeling for the double ionization of $H_2$ and $D_2$ have been carried out to investigate the dependence of the kinetic energy spectra on the laser's peak intensity, pulse length and mean wavelength. For the lower peak intensity, in the so-called sequential ionization regime, double ionization proceeds through three steps: the tunneling ionization of the first electron, followed by electron impact excitation to excited states, and then further ionization by the laser field. The simulation shows that the time interval for each process occurs at sub-femtosecond durations, and such information on time can be obtained to sub-fs accuracy from the measured kinetic energy spectra of the fragmented ions. The full account of the rescattering theory was published in [3, 8, 6a].

At higher peak laser intensity, double ionization occurs through two successive single ionizations and the time interval between the two ionizations can be calculated from the kinetic energy spectra of the fragmented ions as well. This work was published in [39]. We have also performed additional calculations for experiments carried out at KSU and they appeared in [7, 7a].

In the coming year, we will be investigating the angular distributions of the fragmented ions. Such experiments have been carried out at KSU and we have identified a new mechanism—post-ionization alignment effect, which is very important for determining the fragment's angular distributions.

**2. Attosecond physics:** We have developed a strong field approximation which allows us to calculate the electron spectra for laser-assisted autoionization by an attosecond light pulse. The theory can be applied to the Auger decay and to any autoionization process. From the time-delay electron spectra we showed how to extract the lifetime of the resonances. We have also explored the emergence of additional resonances in a two-color IR+XUV experiments where new resonances appear due to the Stark effect of the IR laser. In the process we have also studied the DC Stark effect on the doubly excited states of He [37]. Further extension of these works will be carried out in the coming year.

**3. Hyperspherical close-coupling method for low-energy collisions:** In the last year we have demonstrated clearly the power of diabatic hyperspherical close-coupling method that we have developed recently. The method allows the elmination of channels that couple weakly with the processes of interest. We have published two papers [23, 38] showing the advantage of channel truncations. Other calculations using the HSCC method can be found in [1, 2, 18, 22]. In the coming year, this method will be further extended to other systems, for example, the positronium formation in positron-Na collisions where outstanding discrepancy exists in the literature. We also will work on the collisions such as $\mu^- p^+ + (O,N)$ for the capture of muon by atomic nuclei. Such collisions are being investigated by some experimentalists and isotope effect has been shown to be significant.

**4. Quadruply excited states:** We have classified all the singly, doubly, triply and quadruply states of an atom under the $s^4$ model and the paper has been submitted for publication. No additional effort is planned for this work in the coming year.

## 7. Interaction of Molecules with Intense Femtosecond Laser Pulses

*Igor V. Litvinyuk [ivl@phys.ksu.edu]:*

After having joined the laboratory in January of 2004, the following upgrades to the existing equipment were implemented: 1) Piezo-electric slits were installed in the molecular beam source of the Laser COLTRIMS chamber. That allows us to produce a narrow (<50 μm) molecular beam, which is needed to keep a low count rate and high correlation at high laser intensities, while maintaining low beam temperature. This upgrade was critical for successful experiments on Coulomb explosion imaging and also on electron-ion coincidences spectroscopy. These experiments rely heavily on a high relative coincidence rate, which requires a very small interaction volume. In addition, having the target narrower than the Raleigh range of the laser focus (~ 150 μm) helps control the focal volume effect, which always affects intense laser experiments. 2) A larger diameter laser input window and a shorter focal length focusing mirror were installed on the chamber. That allows the expansion of the laser beam diameter and decreases the diameter of the focal volume. As a result, we further decrease the interaction volume and can achieve higher peak intensities without increasing the total pulse energy.

We also installed the following additional equipment: 1) Two computer-controlled optical pump-probe delay set-ups. One is based on Mach-Zehnder interferometer for long (> 30 fs) pulses used in molecular alignment experiments. Another one is for ultra-short (sub-10 fs) pulses used in Coulomb explosion experiments. We now have the capability for conducting pump-probe studies of molecular dynamics. 2) TOPAS optical parametric amplifier (OPA) with two additional mixing stages was purchased and installed in the KLS room. We now can generate short (~ 50 fs) laser pulses in broad spectral range (230 nm – 2600 nm). The OPA significantly extends KLS's range and capabilities. Two main implications are: 1) single photon excitation of molecules with UV pulses is now possible, giving access to excited state dynamics; 2) much larger electron re-collision energies can be achieved and controlled by using longer wavelengths (800 – 2600 nm).

**1. Effect of orbital structure on strong-field dissociative ionization of molecules**, *A.S. Alnaser, X.-M. Tong, C.L. Cocke and I.V. Litvinyuk.* We measured angular distributions of ion fragments produced in dissociative double ionization of $CO_2$, CO and $C_2H_2$ by intense ultra-short (8 fs) laser pulses. These experiments extend similar recent studies of $O_2$ and $N_2$ [8a] to a wider set of molecules. We found that for sub-10 fs pulses of sufficiently low intensity the fragment angular distributions for all studied molecules are determined by angular dependence of the first ionization step. Those experimental angular distributions were in good agreement with angular dependent ionization probabilities calculated with the Molecular ADK theory. The measured angular distributions directly reflect the symmetry of corresponding molecular orbitals.

**2. Laser-Induced Coulomb Explosion Imaging for Studying Ultrafast Molecular Dynamics**, *A. S. Alnaser, X.-M. Tong, B. Ulrich, C.L. Cocke and I.V. Litvinyuk.* We conducted a series of pump-probe studies of ulrafast dynamics for $H_2$, $N_2$, $O_2$, CO and $C_2H_2$ molecules during the first 100 fs following ionization. We used single or double ionization by an 8 fs pulse as a pump and dissociation by more intense delayed 8 fs pulse as a probe. We observed various wavepacket dynamics, including fast dissociation and dephasing internuclear vibrations with 5 fs time resolution. The results of these experiments are still being analyzed.

**3. Strong Laser Field Interactions with Dynamically Aligned Molecules** *A.S. Alnaser, C. Marhajan, C.L. Cocke and I.V. Litvinyuk.* We measured full momentum distributions of electrons produced by strong field ionization of $N_2$ and $O_2$ molecules, after the molecules were dynamically aligned by a preceding laser pulse. Analysis of results obtained in these experiments is also ongoing.

**Continuing collaboration with Ottawa groups of Paul Corkum and Thomas Brabec:**

**4. Laser-Induced Interference, Focusing, and Diffraction of Rescattering Molecular Photoelectrons**, *S. N. Yurchenko, S. Patchkovskii, I. V. Litvinyuk, P. B. Corkum, and G. L.Yudin.* We solve the time-dependent Schrödinger equation in three dimensions for $H_2^+$ in a one-cycle laser pulse of moderate intensity. We consider fixed nuclear positions and Coulomb electron-nuclear interaction potentials. We analyze the field-induced electron interference and diffraction patterns. To extract the ionization dynamics we subtract the excitations to all significant low-lying bound states explicitly. In the simulation we follow the time evolution of well-defined wavepacket, which is formed near the first peak of the laser field. We observe the subsequent fragmentation of the wavepacket due to molecular focusing. We show how to retrieve diffraction molecular image by taking the ratio of the momentum distributions in the two perpendicular directions. Even at moderate laser intensities, the position of the diffraction peaks is well described by the classical slit diffraction rule.

**5. Shake-up excitation during intense field tunnel ionization**, *I. Litvinyuk, F. Légaré, P. W. Dooley, D. M. Villeneuve, P. B.Corkum, J. Zanghellini, A. Pegarkov, C. Fabian, and T. Brabec.* "Shake-off" and "shake-up" play an important role in the X-ray double ionization of atoms. The first electron is ionized by absorbing an X-ray photon and shakes up a second electron on its way out. We investigate shake-up in the opposite, low frequency, strong field limit, both theoretically and experimentally. Theoretically, a complete analytical theory of shake-up in intense laser fields is developed. We predict that shake-up produces one excited $\sigma_u$ $D_2^+$ state in $10^5$ ionization events. Shake-up is measured experimentally by using the molecular clock provided by the internuclear motion. The number of measured events is found to be in excellent agreement with theory.

**6. Imaging ultra-fast molecular dynamics using laser Coulomb explosion,** *F. Légaré, K.F. Lee, I.V. Litvinyuk, P.W. Dooley, A.D. Bandrauk, D.M. Villeneuve, P.B. Corkum.* We demonstrate a novel technique for direct imaging of ultrafast molecular dynamics. The technique is based on pump-probe approach employing few-cycle (8 fs) laser pulses. The technique is applied to directly map nuclear motion in vibrating $D_2^+$ and dissociating $SO_2^{2+}/SO_2^{3+}$ molecules. In each case, the dynamics is launched by ionizing a neutral molecule with the pump pulse. The time-dependent molecular structure is then interrogated with a delayed probe pulse by analyzing the momenta of the Coulomb explosion fragments. The method traces atomic motion with 5 fs time resolution.

## 8. Electronic Excitations in Carbon Nanotubes via Short-Pulse Pump-Probe Interactions and in Highly Charged Ions via Collisions

*Patrick Richard [richard@phys.ksu.edu]:*

**1. Time–resolved photoexcitation of image–potential states in carbon nanotubes:** *M. Zamkov, N. Woody, S. Bing, H.S. Chakraborty, Z. Chang, U. Thumm, and P. Richard.* During the past year we have initiated a study of multi walled nanotubes. This project utilizes the new capabilities of the JRML ultra-high intensity, fs laser facility. In this effort we have obtained the first experimental evidence for the existence of image-potential states in carbon nanotubes. The observed features constitute a new class of surface image states due to their quantized centrifugal motion. These types of states were recently predicted in a Phys. Rev. Letters article by B.E. Granger, P. Kral, H.R. Sadeghpour, and M. Shapiro. This has been extremely satisfying result in view of the somewhat lukewarm comments from the review panel of our three year renewal proposal. Some reviewers expressed skepticism at our ability to perform these types of experiments.

Quantized states are known to form in front of flat surfaces due to the polarizing image–interaction of an external electron. For years the investigation of these states above metal surfaces has served as a powerful tool for probing a variety of physical and chemical phenomena on the nanometer scale. The unique properties of image states are determined by the extreme sensitivity of "image" electrons to any changes in the dielectric susceptibility at the surface. Therefore, through measurements of their binding energies and lifetimes it is possible to elucidate many complex processes that ultimately promote our knowledge of surface structure.

The experimental setup consisted of a Ti:sapphire laser system generating 35 femtosecond pulses at 2 kHz. Frequency–tripled UV photons were produced through non–linear effects during the photoionization of $N_2$ molecules. The fluence of the resulting 100 fs UV pump–pulse used for promoting an electron population into unoccupied image states was estimated to be $40\mu J/cm^2$. Binding energies and temporal dynamics of photoexcited electrons were subsequently probed with a delayed IR probe–pulse having a fluence of $30\mu J/cm^2$. In order to increase the count rate to pulse fluence ratio, the laser spot size on the sample for both UV and IR beams was maintained relatively large (~ $400\mu m$). Following the photoionization by IR pulses, electrons drifted into the magnetically and electrically shielded 30-cm long spectrometer tube and were detected with a strip–and– wedge position sensitive detector. The overall energy resolution of the system in the case of 1 eV electrons was 20 meV. In the present study, the energy of the UV photon exceeded the sample work function of 4.24 ± 0.10 eV by 0.47 eV, resulting in a pump–only photoemission of low–energy electrons. In order to isolate the photoemission originating from image–potential states, we recorded the change in the photoelectron signal induced by the IR probe–pulse (UV pump IR probe electron signal minus the UV pump electron signal). The corresponding "excitation" difference resulting in a series of excitation peaks which can be correlated with the expected positions of image potential states of the multi walled carbon nanotubes. The lifetimes of these states are also obtained in a series of measurements of the electron signal versus the pump-probe delay time. A Physical Review Letters paper has been written on this work and has recently been accepted [1a]. We have also written a theoretical paper that presents results of image-potential states of single and multi walled carbon nanotubes. This paper has recently been accepted in Phys. Rev. B [4a]. During the next contract period we plan to investigate the lifetimes of charge carriers in multi walled carbon nanotubes and also to extend our experiment to the investigation of double walled carbon nanotubes contingent on obtaining suitable DWCN targets.

**2. Doubly and Triply Excited States formed in ion-atom collisions:** *M. Zamkov, E. P. Benis, T.J.M. Zouros, T.W.Gorczyca, A. Habib, and P. Richard.* We have continued our investigation of doubly and triply excited states of highly-charged ions using the JRML accelerators. The recent results of this work are published this year in several papers [10, 11, 12, 13, 24]. We are installing a new LabView data acquisition system for operation of the Crete electron spectrometer system. We plan to complete the work on triply excited states during the coming year.

## 9. Interactions of Ions and Photons with Surfaces, Molecules and Atoms

Uwe Thumm [thumm@phys.ksu.edu]:

### 1. Laser-assisted collisions:

*We are developing numerical and analytical tools to efficiently predict the effects of a strong laser field on the dynamics of electron capture and emission in ion-atom collisions. These investigations assist in the planing of future experiments with crossed laser and particle beams at the J.R. Macdonald Laboratory.*

We investigated the effects of a strong 1064nm laser field on electron capture and emission in slow (keV) proton-hydrogen collisions within a reduced dimensionality model of the scattering system in which both, the motion of the active electron and the laser electric field vector are confined to the scattering plane [30]. By solving the time-dependent Schrödinger equation, we examined the probabilities for electron capture and ionization as a function of the laser intensity, the projectile impact parameter b and the laser phase Φ that determines the orientation of the laser electric field with respect to the internuclear axis at the time of closest approach between target and projectile. Our results for the b-dependent ionization and capture probabilities show a strong dependence on both Φ and the helicity of the circularly polarized laser light. For intensities above $2 \times 10^{12}$ W/cm$^2$ our model predicts a noticeable circular dichroism in the capture probability for slow proton-hydrogen collisions. Interestingly, this dichroism persists after averaging over Φ. Capture and electron emission probabilities defer significantly from results for laser-unassisted collisions. The difference in the capture cross sections for co- and counter rotating collisions, $\sigma_{cap}^{+}$ and $\sigma_{cap}^{-}$, amounts to up to 40 % at a laser intensity of $5 \times 10^{13}$ W/cm$^2$. We consider these differences as upper limits for the dichroism effect and expect them to decrease slightly in full three-dimensional calculations. Furthermore, we find evidence for a charge resonance enhanced ionization mechanism that may enable the measurement of the absolute laser phase Φ.

### 2. Surface-morphology effects in the neutralization of negative hydrogen ions near silver and copper surfaces

*In this project we model and compute resonant transfer of a single electron, initially bound to the projectile, during the reflection of a slow ion or atom on a metal surface. In the long run, this work may improve our understanding of surface chemical reactions, catalysis, and diagnostics.*

We compared the neutralization of hydrogen anions in front of plane Ag and Cu surfaces of symmetries (100) and (111) using the Crank-Nicholson wave-packet propagation method [29,9a]. The Ag (100) surface binds the surface state that is degenerate with the valence band and rapidly decays while being populated by the ion. For Ag (111), in contrast, the population of a quasi-local Shockley surface state inside the projected L-band gap impedes the electron decay into the bulk along the direction normal to the surface. This difference in the decay pattern strongly affects the survival of 1 keV ions scattered from these surfaces and scattering off the Ag (111) surface results in about an order of magnitude higher ion-survival probabilities as compared to that off Ag (100). Our results for Ag (111) show good agreement with the measurements [29]. The anion-survival near the (111) surface is much higher since, on the time scale of the ion-surface interaction, the surface state retains electrons and enables their recapture. Taking advantage of the flexibility of the wave-packet-propagation scheme with regard to the choice of an effective surface potential, we have begun to investigate resonance formation and charge exchange near near vicinal and nano-structured surfaces.

### 3. Image-Potential States of Single- and Multi-Walled Carbon Nanotubes

*The approximate cylindrical symmetry of carbon nanotubes enables the formation of long-lived image states, that provide new setting for the investigation of nanotube surface phenomena. such as surface reactivity, structural and optical properties at interfaces, and the dynamics in nanotube heterojunctions.*

In a collaborative effort with Prof. Richard's group, we investiagted the formation of image--potential states near the surfaces of carbon nanotubes. We helped in calculating binding energies and wave wave function by modelling the interactions inside the nanotube with a cylindrical, jelliumlike short-range potential that is parameterized to ensure the correct vacuum to surface transition [4a]. Recent experiments in the J.R. Macdonald laboratory provide first evidence for the existence of these states [1a].

## B. Financial Report

It is anticipated that there will be no unexpended funds for the current funding period.

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