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August 27, 2002

Dr. Eric A. Rohlfing, SC-14
Chemical Sciences, Geosciences and
Biosciences Division
Office of Basic Energy Sciences
U. S. Department of Energy
19901 Germantown Road
Germantown, MD 20874-1290

Re: Continuation Request
DE-FG02-86ER13491
New Grant Period: 2/15/03 - 2/14/04
Grant Amount: $2,350,000

Dear Dr. Rohlfing,

Attached is the continuation progress report for the above referenced DOE grant to the J. R. Macdonald Laboratory at Kansas State University. This report is for the period 2/15/02 – 2/14/03. Also included are a statement of the anticipated unexpended funds at the end of the budget period, and a list of publications resulting from the grant period.

The continuation progress report contains a brief summary of accomplishments by each of the nine investigators on the grant.

Sincerely yours,

Patrick Richard

Patrick Richard
Principal Investigator
Director, J. R. Macdonald Laboratory
Cortelyou-Rust Distinguished Professor

C O N T I N U A T I O N P R O G R E S S R E P O R T

FOR GRANT NO: DE-FG02-86ER13491

GRANT PERIOD: 15 February 2002 - 14 February 2003

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

INSTITUTION: Kansas State University
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August 2002

[Letter-Numbers in parenthesis refer to the original Renewal Proposal when applicable]

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1. Atomic Physics with Ion Beams and Synchrotron Radiation

C. L. Cocke [cocke @phys.ksu.edu; (785) 532-1609]

1.1 (A6) Photoelectron diffraction from C_2H_2, C_2H_4 and other small molecules, *T. Osipov, C. L.Cocke (KSU), A.Landers (Western Mich. Univ.), R.Doerner, Th.Weber, L.Schmidt, A..Staudte, H.Schmidt-Boecking (U.Frankfurt), M.H.Prior (LBL)* Using beamlines 4.0 and 9.3.2 at the Advanced Light Source, we have measured the correlated momentum-space distributions of photoelectrons and charged photofragments ejected when the K-shells of acetylene and ethylene are photoionized. During the past year our work has centered on the analysis of diffraction patterns from C_2H_2 and C_2H_4. Our attempt to identify in acetylene an f-wave shape resonance, well known in N_2 and CO but controversial for C_2H_n, was initially frustrated by the interesting observation that this molecule often undergoes isomerization to the vinylidene configuration before it dissociates. The separation of decays from the acetylene and vinylidene channels proved challenging, but has finally been resolved, with the following conclusions:
(1) Using only molecules decaying approximately along the polarization vector, the acetylene channel shows the expected f-wave structure near the predicted resonant energy (2) The vinylidene channel shows that this structure is considerably washed out. This can only occur because the molecule rotates the bond angle after the photoelectron departs (a fast process) but before the dissociation is complete. This rotation angle provides information on the isomerization time: if the proton re-alignment occurs slowly, the correlation between axial dissociation and photoelectron emission will be entirely lost. Analysis of the data reveals that the rotation of the bond angle is very close to the minimum value consistent with the mass redistribution if the proton travels peripherally around the molecule to its new home. We deduce that the time scale for isomerization is no longer than approximately 100 fs.

For the case of C_2H_4, no such complication occurs, and full photoelectron angular distributions have been taken as a function of photoelectron energy spanning the range of the expected shape resonance; a partial wave analysis is in progress.

1.2 (A1.1) COLTRIMS measurements of electron capture from and low energy ionization of atomic H *E. Edgu-Fry and C. L. Cocke* The goal of this project is to establish an atomic hydrogen source compatible with use in a COLTRIMS experiment. It will be used 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. In addition, electron capture from atomic hydrogen by multiply charged ions will be studied. The use of the true one-electron target removes uncertainties associated with the comparison of one-electron dynamics calculations with data from multielectron targets. The major difficulty of this project has been to develop a COLTRIMS-compatible atomic hydrogen target. We finally have a working microwave discharge source, which provides a jet of atomic hydrogen with an acceptable momentum spread. Using this target, electron capture from atomic hydrogen by Ar^{8+} projectiles has been studied, differential in both the Q-value of the reaction and the transverse momentum transfer. These data are under analysis. Preliminary data have been taken for the ionization of atomic H by protons. It is expected that the performance of this ion source will be adequate to enable the completion of this project.

1.3 (A5.1) Electron capture from D_2^+ by doubly charged projectiles *I. Reiser, H.Braeuning (U.Giessen), C.D.Lin, E.Sidky and C.L.Cocke*. The dependence of the cross section for electron capture from H_2^+ by the doubly charged projectiles He^{2+}, N^{2+} and Ar^{2+} on the angle between the beam axis and the molecular axis has been measured. The purpose of this experiment was to seek

evidence for a "two-slit" interference of amplitudes for capture from the two centers. The price one pays for the clean nature of this experiment, which involves only one active electron, is that it is an ion-ion collision with the attendant low count rates and ion-optics difficulties. The experiment has now been completed. The results show a clear tendency for the capture to optimize for molecules aligned perpendicular to the beam axis. This result is qualitatively consistent with the expectations of the "two-slit" interference picture and is also confirmed by a model calculation based on a coherent combination of calculated atomic scattering amplitudes. This work was the thesis work of Ingrid Reiser and is being prepared for publication.

1.4 (A6) Identification of a rescattering mechanism in the double ionization of D_2 by intense laser pulses, *T .Osipov,E. P. Benis, A. Wech, C. Wyant, B. Shan, Z. Chang and C. L. Cocke.* It is now well established that so-called "non-sequential" ionization of neutral atoms by fs laser pulses with intensities in the range 10^{14} -10^{15} watts/cm^2 can occur through a rescattering mechanism whereby the electron liberated in the single ionization process returns to the ion with sufficient energy to further ionize the singly charged ion through an (e,2e) process. The equivalent process in the ionization of molecules has not been seen until very recently [Niikura et al.,Nature, to be published 2002; and ref.1 in publication list], partially due to the fact that charge-resonance-enhanced ionization is usually the dominant process for multiple ionization. Using the newly installed fs light source in the JRM laboratory, we have established, using linearly and circularly polarized light, that the high energy group reported by Staudte et al. is caused by a rescattering process. The energy distribution of the resulting deuterons is consistent with the picture that the vibrational wave packet created in the single ionization of the D_2 molecule, trapped in the gerade potential well of the D_2^+ ion, is further ionized by the returning electron. This second step must itself proceed through excited states of the D_2^+ ion, however, since the returning electron lacks sufficient energy to ionize the molecular ion in a single step. The observed deuteron energy distribution extends between limits that are consistent with the reflected energies from the turning points of the vibrational wave packet in the D_2^+ gerarde potential well.

II. Future plans:

1.1 The ALS collaboration: In addition to the completion of the analysis of the C_2H_n data, future plans include the experimental studies at the ALS of ionization-excitation of D_2 below the double ionization threshold, photo double ionization of D_2 far enough above threshold (>150 eV) that interference between photoelectron waves from the two centers might be observable, and coincident high-resolution-Auger/photoelectron/KER experiments for which the limits of the validity of the two-step picture of Auger and dissociation process may be probed. We continue to have approximately two two-bunch runs a year at the ALS, and expect to continue this. We are attempting to arrange the attachment of Dr. Thorsten Weber of U. Frankfurt as a Feodor Lynen Fellow, partially supported also by KSU and LBL, as a longer-term resident member of this group.

1.2 The COLTRIMS project at the EBIS with the atomic hydrogen target: This project is the Ph.D. thesis work of Erge Edgu. The major future plan for this project is to complete the investigation of the mechanism for low energy ionization from atomic hydrogen over as extended a range of proton energies and as highly differential as possible. We are presently engaged in a discussion of collaborative COLTRIMS work at the EBIS on capture by highly charged ions with C. Havener of ORNL and R. Schuch at U. Stockholm.

1.4 Laser interactions with neutral gases and beams: We plan to

1) Continue investigation of the rescattering process for molecules, including theoretical modeling of the process in collaboration with KSU and other theorists, with an eye to exploiting the generation of short electron bursts in the first step to explore the dynamics of the outgoing vibrational wave packet on a fs time scale.
2) Construct a special ultra-high-vacuum COLTRIMS chamber suitable for investigating correlated momentum spectra of both electrons and heavy fragments from laser ionization of atoms and molecules; use of this chamber to carry out kinematically complete ionzation studies of neutral targets.
3) Investigate the ionization of ion beams from the KSU ECR source by intense laser pulses. Target beams will include H_2^+ (in collaboration with I. Ben Itzhak) and singly charged noble gas beams. The ion-ion collision facility is being converted to an ion-laser collision facility.

2. Atomic Physics with High Velocity, Highly Charged Ion Beams

P. Richard [richard@phys.ksu.edu; (785)532-6783]

The goals of this part of the JRML program are 1) to study doubly and triply excited states formed in high velocity, highly charged ion–neutral target collisions by state selective electron-ion resonances and by triple electron capture and 2) to study the dynamics of one and two electron processes in which electrons from bound neutral target states are brought to continuum and bound states of high velocity, highly-charged ions.

2.1 (A2.2) Doubly and Triply Excited States in High Z Few Electron Ions

H. Aliabadi, E.P. Benis, P. Richard, M. Zamkov, H. Tawara, T. G. Lee, C. D. Lin, T.J.M. Zouros (U of Crete, Greece), and Tom Gorczyca (Western Michigan U, Kalamazoo, MI)

Resonance excitation of doubly excited states: We continue our investigation of doubly excited states of ions formed by electron resonant transfer and excitation in ion-atom collisions. The process is detected by observing the Auger electron emission from the projectile resonant state at zero degrees in the laboratory frame. The projectile electron emission spectrum can be analyzed as an electron-ion collision system by including the effects of the Compton profile of the target electrons and the kinematics of the collision. It can then be directly compared with electron-ion differential elastic and inelastic scattering calculations. We have successfully demonstrated that the $2p^2\ ^1D$ Auger emission from C^{5+}, N^{6+}, O^{7+}, and F^{8+} + H_2 collisions, when analyzed in this way, gives very good agreement with R-Matrix calculations for $C^{5+}(1s) + e^- \rightarrow C^{4+}(2p^2, {}^1D) \rightarrow C^{5+}(1s) + e^-$ (and similarly for N^{6+}, O^{7+}, and F^{8+}) resonance elastic scattering. We have recently extended these measurements to the complete *2lnl' and 1s2lnl'* manifolds of resonance states for B^{4+} and B^{3+} projectiles, respectively. R-Matrix calculations are in very good agreement for the two electron 2*l*n*l*' states as indicated above and in overall good agreement for the case of three electron resonance states (e.g. $1s2p^2\ ^2D$). We have also measured the inelastic scattering channel of the electron-ion scattering process by observing the Auger decays to the 2s/2p final states. The first manifold above the inelastic scattering threshold is the 3*l*3*l*' configuration. The states in the 3*l*3*l*' configuration decay primarily via the inelastic scattering channel. The experiment was done for $F^{8+}(1s) + e^- \rightarrow F^{7+}(3l3l') \rightarrow F^{8+}(2s/2p) + e^{-`}$. We have recently extended the measurements to B^{4+} projectiles. The resonances in this case are poorly resolved, however the gross structural features are reproduced by R-matrix calculations.

Resonance excitation of three-electron hollow ions: In the process of studying resonance states we noticed that for two-electron ion beams we could enhance the production of certain states by controlling the manner in which the beam was prepared. The effect is due to the 1s2s ^3S metastable component in the beam. We undertook a systematic study of the production of metastable ion beams, since what we discovered was that we could preferentially, resonantly populate three-electron hollow ions, e.g. $1s2s + e^- \rightarrow 2sn$*l*n*l*' resonances. Below is a brief summary of these studies. <u>FIRST STUDY</u>: We observed that the metastable fraction in $B^3(1s^2\ ^1S;1s2s\ ^3S)$ beams after traversing a foil is constant over the investigated energy range, whereas the fraction produced in collisions with gas targets strongly depends on the incident beam energy. K-vacancy production followed by electron capture to the 2s state in the ion-beam stripping process is identified as a dominant mechanism. <u>Z DEPENDENCE</u>: The metastable fraction in fast He-like B, C, N, O, and F beams produced in collisions with thin carbon foils was studied. It was found that the C^{4+} beam contained a significantly lower metastable fraction than

that of the other beams. The observed deviation is explained on the basis of K-vacancy sharing, which is known to have the highest probability for symmetric collisions. NEW TECHNIQUE: A new experimental technique was developed, which utilizes only the electron yields of the $1s2p^2$ ^2D and 1s2s2p ^4P doubly excited states produced in collisions of two-electron ion beams with H_2 or He targets. The metastable beam fraction is *in situ* determined in two successive measurements at the same beam energy but having different metastable fractions. The results are in good agreement with those from our first study discussed above, but they do not rely on theoretical cross sections. HOLLOW IONS: We have used the metastable ion production information to study the resonance excitation of three-electron hollow ions. Accurate measurements of the double differential cross sections are made on the basis of the known metastable ion beam fractions and known cross sections for resonance transfer and excitation from the ground-state component of the beam. The first system investigated was $B^{3+}(1s2s\ ^3S)$ + H_2 leading to the $B^{2+}(2s2p^2\ ^2D)$ resonance that decays to $B^{3+}(1s2s\ ^3S)$, the elastic scattering channel and to $B^{3+}(1s2s\ ^1S)$ and $B^{3+}(1s2p\ ^3P)$, two inelastic scattering channels.

Doubly and triply excited three-electron ions formed by triple capture: At this point in our research we observed that we could readily populate and observe doubly and triply excited states in triple capture measurements. The first experiment involved producing states of the 1s2*l*2*l*' manifold. We later figured a way to populate more exotic and interesting triply excited manifolds, in particular the $2p^3$ states. This work is in the early stages.

2.2 (A2.1) Study of One- and Two-Electron Processes in Ion-Atom Collisions: Single Capture and Transfer Ionization, *R. Unal, P. Richard, N. Woody, E. P. Benis, I. Ben-Itzhak, C. L. Cocke, M. J. Singh, H. Tawara, C. D. Lin, and T. G. Lee.*

We have investigated the charge state and energy dependences of Transfer Ionization (TI) and Single Capture (SC). The main emphasis of this research is to provide reliable, measured cross sections that can be compared with new state of the art calculations. The collision systems studied are $F^{(4-9)+}$ ions interacting with helium produced in a collimated supersonic jet. The measurements were made for beam energies between 0.5 and 2.5 MeV/u, which corresponds to projectile velocities between 6 and 10 au. A recoil ion momentum spectrometer was used to separate TI and SC based on the longitudinal momentum transfer as well as the time-of-flight of the recoil ions. The ratio, $\sigma_{TI}/\sigma_{SC,}$ was accurately obtained by this method, and the individual cross-sections for the cases of bare and hydrogen-like fluorine ions were determined by using the measured ratios and the previously measured total charge exchange cross-sections. New calculations were undertaken to compare with this accurate data set. Previous data obtained from time of flight of the recoil ions suffer from large errors and are not in agreement with the present data or previous calculations. A more consistent independent electron model, using the two-center semiclassical close coupling method was used to calculate the SC and TI cross sections. Good agreement was found between the new measurements and the new calculations.

Future Plans: 2.1 Doubly and Triply excited States: We will continue the study of triply excited three-electron ions. We will compare our results with existing perturbative calculations by Safronova *et al*. and with predictions from newer multielectron theories. We will modify the hemispherical analyzer to rid electron spectra of rescattered cusp electrons.

2.2 TI/SC ratios: We plan to make the final measurements of the TI/SC ratios for F^{q+} beams.

3. Molecular Dynamics with Ion and Laser Beams

Itzik Ben-Itzhak [ibi@phys.ksu.edu, (785)-539-2155]

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.

3.1 (A.3.5 & A.3.6) Molecular dissociation imaging of collision induced dissociation and dissociative capture in slow H_2^+ + Ar (He) collisions, *D. Hathiramani, I. Ben-Itzhak, J.W. Maseberg, A.M. Sayler, M.A. Smith K.D. Carnes.* The dissociation of hydrogen molecular ions following a slow collision (keV) is studied by 3D momentum imaging of the fragments. The two main processes at this collision energy, collision-induced dissociation (CID, e.g. $H_2^+ + Ar \rightarrow H^+ + H + Ar$) and dissociative capture (DC, e.g. $H_2^+ + Ar \rightarrow H + H + Ar^+$), are experimentally separated in the method we recently developed. Using an electric field in the target region followed by a field free region, we managed to separate the CID from the DC in time. Thus, it is possible to evaluate the relative importance of these two processes, because both CID and DC are measured simultaneously. The same is true for the ratio of the two possible CID channels for heteronuclear molecules, i.e. A^++B or A+B^+. Moreover, this method allows one to distinguish experimentally between two different mechanisms of CID and thus study each one of them in detail. The two mechanisms differ in the momentum transfer to the projectile: (i) very small momentum transfer for CID caused by an electronic excitation to a repulsive state, and (ii) very large momentum transfer for CID caused by a vibrational/rotational excitation. This distinguishes the present work from previous studies [e.g. Los and Govers, in *Collision Spectroscopy* (ed. R.G. Cooks, Plenum Press, NY 1978) p-289], in which no such separation was possible. Results for CID of 3 keV H_2^+ by an electronic excitation indicate that the dissociating fragments align along the beam direction for "short" molecular ions and perpendicular to the beam for "long" ones, as predicted by Green and Peek. [Green and Peek, *Phys. Rev*. **183,** 166 (1969)]. The vibrational/rotational dissociation mechanism shows very strong alignment effects. First, molecular ions aligned perpendicular to the beam velocity (i.e. θ=90°) are much more likely to dissociate by this mechanism. Second, the dissociation velocity is preferentially aligned along the momentum transfer. Further analysis of the DC and CID processes as well as model calculations of the vibrational CID mechanism are underway. Recently we have conducted studies of CID of HD^+ in similar collisions in search of the isotopic effects observed previously for this molecular ion [see, for example, Lehman *et al*. *Int. J. Mass Spectrom. Ion Proc.* **69**, 85 (1986)], namely, that the dissociation into H + D^+ was favored over H^+ + D. Preliminary results indicate that such effects, if they exist, are much smaller than those reported in previous work. These results will be presented as an invited talk in the CAARI 2002 meeting.

Future plans: Systematic studies of DC and CID caused by either electronic or vibrational excitation 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^+. While conducting these studies on the existing system, an improved setup, which includes a cold jet target, will be assembled and tested. This will enable the simultaneous measurement of the recoil ion momentum for DC reactions, thus providing kinematically complete information about the process.

3.2 Isotopic effects and asymmetries in bond-rearrangement and bond-breaking processes in water ionized by fast proton impact. *A.M. Sayler, Z.S. Casey, J.W. Maseberg, D. Hathiramani, 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 a similar isotopic effect following double ionization of water, i.e. in $H_2O^{2+} \rightarrow H_2^+ + O^+$, although these results are preliminary. In addition, we have observed large asymmetries in bond breaking in the HDO isotope. For example, $HDO^+ \rightarrow H^+ + OD$ is more likely than $HDO^+ \rightarrow D^+ + OH$ by about a factor of 1.5, suggesting that it is easier to break the O-H bond than the O-D bond. This preference is even larger in the dissociation of HDO^{2+}. 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. In addition, we determined the relative cross sections of all dissociation channels including $H^+ + H^+ + O$ which typically are not measured by TOF techniques. These results were presented by Max Sayler as an invited talk in the undergraduate research session of DAMOP 2002.

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. We also intend to study ionization and fragmentation of water by fast highly charged ions and compare them to fast proton impact.

3.3 (A.6) High intensity lasers interactions

3.3.1 Evidence for pondermotive-gradient field-ionization in an intense focused laser beam, *E. Wells (UVA), I. Ben-Itzhak, and R.R. Jones (UVA)*. We have measured the Rydberg ion population produced during intense laser ionization in Xe, Kr, and Ar. The branching ratio for production of Rydberg ions, $A^{(q-1)+*}$, to ions A^{q+} has been measured as a function of laser intensity and polarization. Using 100 fs, 790 nm laser pulses, singly and double charged Rydberg ions are observed in Xe, but in Kr and Ar only singly charged Rydberg ions are seen. Model calculations of ionization by the gradient of the pondermotive potential (∇U_p) of the focused laser beam suggest that the Rydberg population is inversely proportional to the ionization potential of the ion species. These calculations are in agreement with our data except for the Xe^{2+*}/Xe^{3+} ratio, which is anomalously large, approximately by a factor of two higher than the Xe^{+*}/Xe^{2+} ratio.

3.3.2 Photo ionization and photo dissociation of H_2^+ by an intense short pulse laser, *I. Ben-Itzhak, J.W. Maseberg, A.M. Sayler, M.A. Smith K.D. Carnes, Z. Chang, C. Fehrenbach, and C.L. Cocke*. We have recently begun measurements of ionization and dissociation of a few keV H_2^+ beam crossed by an intense short-pulse laser beam using 3D molecular dissociation imaging on a newly installed apparatus employing techniques similar to those described in 3.1.

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

4. Structure and Dynamics of Atoms, Ions, Molecules, and Surfaces: Ion-Atom Collisions with Laser Prepared Targets

B. D. DePaola [depaola@phys.ksu.edu;(785)532-1623]

4.1 (A1.2) Charge Transfer in Extremely Slow Collisions Between Highly Charged Ions and Helium (B. D. DePaola, C. L. Cocke, C. W. Fehrenbach, K. Okuno, and C. Verzani)

In this project we measure single and double (net) charge exchange in collisions of highly charged ions with He and H_2 targets. The main pieces of apparatus used are the CRYEBIS and an OPIG (octupole ion guide). The latter was developed at Tokyo Metropolitan University and is used to decelerate the projectile ions, while at the same time keeping them confined in a tight beam. Collisions with the target gas occur in a cell within the OPIG; the product projectile ions are charge-state analyzed using a magnet and two-dimensional position-sensitive detector (2D-PSD).

In the last progress report we reported that we had improved both the product detection system and the ion optics located downstream from the OPIG. Since then, the last of the apparatus irregularities have been addressed and charge transfer cross sections for several collision systems have been investigated. Spread in CRYEBIS beam energy of a few eVq sets the practical lower limit on the collision energies studied. Systems studied to date include: (He/ 2 /He), (N/ 3-7 /H_2), (O/ 2 /H_2, He, Ne, Ar), (Ne/ 3-6 /He), (Ar / 5-9,11 /H_2, He), and (Xe/ 24 /He). Notation is (Ion Specie/ q /target) .

From this list one can see that, with a reliable lower energy limit of 3eVq, typical minimum collision "velocities" of approximately 400 eV/u were studied. The results obtained thus far will be submitted for publication in the near future. In the coming year we will attempt to add to these data, in the process of looking for and trying to understand systematics in the cross section trends, and comparing measurements with various theoretical models and semi-empirical scaling laws.

4.2 (A4.3) COLTRIMS with a Laser-Cooled Target: MOTRIMS (B. D. DePaola, Z. Chang, C. W. Fehrenbach, X. Flechard, R. Bredy, H. Camp, and H. Nguyen)

a) Cross sections, differential in scattering angle.

At the time of the last progress report the MOTRIMS apparatus was working and already being used to measure, with unprecedented resolution, relative charge exchange cross sections, differential in initial state of the target, final state of the projectile, and projectile scattering angle. As the test case in those studies, Cs^+ at collision energies between 2 and 7 keV was used as the projectile. As a natural consequence of being in a MOT, the target was in a mixture of 5s and 5p states. As noted in the last report, because of the extremely high Q-value resolution, it is possible by chopping the trapping and repump lasers, to determine the ratio of cross sections for capture from Rb(5p) and Rb(5s), without relying on model-dependent fluorescence measurements. Since the last reporting period we have taken advantage of all the advantages of the MOTRIMS technique to carry out charge exchange studies for a variety of projectile ions over a range of collision energies. Because the ion source currently in place on the MOTRIMS apparatus is of the thermionic type (chosen so as to have a very small spread in projectile velocity) the experiments are limited to singly charged alkali and alkali earth projectiles. In the coming year, the alkali and alkali earth cross section measurements will be completed, and the results will be written up and submitted for publication.

In addition to starting the measurements of these cross sections, quite a bit of effort has been spent on enabling the fast chopping of the MOT's quadrupole B-field. Based on numerical simulations of ion trajectories, it is believed that at least a threefold improvement in scattering angle resolution will be achieved once this upgrade is implemented. During the coming year, we will try to implement the B-field chopping electronics and use the expected enhancement in scattering angle resolution to look for structure in $d\sigma/d\Omega$, as predicted by theory for certain selected collision systems.

b) Use of charge exchange as a diagnostic of population dynamics.

We have explored the capabilities of the MOTRIMS apparatus for measuring excited state fractions in the MOT. An important test case was to measure cross section ratios (as described in the preceding section) while deliberately varying the excited state fraction by adjusting the detuning of the trapping laser. While the excited state fraction was measured to vary by a factor of 2 (from 15% to 30%) the cross section ratios remained constant – as they should for a fixed collision energy. A second important test was the measurement of the *time dependence* of the excited state fraction in the MOT as the repump laser was chopped on and off. The experiment confirmed what is already known: the excited state fraction falls to zero when the repump laser is blocked as the trapping laser optically pumps the target atoms to the "dark", optically inaccessible lower hyperfine level. The success of these experiments opens the door to studies of population dynamics in the MOT caused by a variety of processes, including dimer formation due to 3-body collisions, photo-association, and stimulated Raman adiabatic passage (STIRAP). With the MOTRIMS apparatus, excited state dynamics can be studied with a temporal resolution of a few nanoseconds, over a time span of a few milliseconds. Unlike the use of an optical probe, the ion beam interacts so weakly with the target that the measurements may be viewed as non-destructive. The results of these experiments have been submitted to *Physical Review Letters*.

c) **Ion spatial imaging to measure above threshold ionization rates.**

A long-time problem in the measurement of ionization rates in interaction of intense laser pulses with atomic and molecular gases and vapors is in the accurate determination of the laser intensity which caused the ionization. The difficulty is that intensity variation in a focused laser beam can vary over more than an order of magnitude. Thus, ions extracted from the interaction region, were produced by a field whose value has large uncertainty. A possible solution to this problem has been explored by using ion optics to image the laser interaction region onto a 2D-PSD. The apparatus consisted of the MOTRIMS momentum spectrometer, to which potentials were applied that were appropriate to spatial, rather than momentum, imaging. With two dimensions provided by the PSD, and the third dimension provided by the ion the time-of-flight, a 3-D image of the ionization region was built up, event by event. The spatial variation of the laser beam intensity was separately measured with a scanning CCD camera. Thus, with knowledge of the target thickness, one could deduce, through a single image of thousands of events, the absolute ionization rate as a function of laser intensity, for a range of intensities. In principle, one could introduce any number of atomic or molecular gases or vapors into the chamber for ionization studies. In this test case, Rb vapor supplied by the "getter" source used in MOT experiments was studied. The target thickness was directly measured from absorption studies, which made use of one of the diode lasers normally tuned for trapping. The methodology was found to work beautifully. Results were presented at the 2002 ICAP (*International Conference on Atomic Processes)*, and will soon be submitted for publication.

5. Study of High Harmonic Generation

Z. Chang [chang@phys.ksu.edu; (785) 532-1621)]

The goals of this part of the JRML program are 1) to develop a high repetition rate, high intensity laser system for laser-atom interaction studies, 2) to study high order harmonic generation from molecules using the laser, and 3) to study high order harmonic generation using an optical parametric amplifier.

5.1 Development of the *Kansas Light Source*, a high repetition rate and high intensity laser system, *Bing Shan, Chun Wang, Shambhu Ghimire and Zenghu Chang*

The new inhouse-built femtosecond laser laid the foundation for the initiative to study high intensity laser-atom interactions at KSU. It is a high repetition rate laser to reduce data acquisition time and to avoid space change effects. It can operate at a 1 to 2 kHz repetition rate to accommodate the data acquisition electronics of the COLTRIMS and other time-of–flight spectrometers. Strong optical fields (about 1 atomic unit) can be achieved with a moderate focusing lens. The laser oscillator and amplifier are pumped with diode pumped solid-state lasers to assure shot-to-shot and long time stabilities. The laser has been used by six users from the JRML and from outside, which include:

1. Zenghu Chang: high harmonic generation (one paper submitted).
2. C. Lew Cocke: laser-molecule interactions in a COLTRIMS.
3. Brett DePaola: laser-atom interactions in a MOTRIMS.
4. Patrick Richard and Zenghu Chang: laser-atom interactions using an imaging detector.
5. Jin Wang (Argonne National Laboratory): streak camera testing (one paper submitted).
6. Itzik Ben-Itzhak: Laser-molecular ion beam interactions.

5.2 High harmonic cutoff extension with molecules, *Bing Shan, Mahendra Shakya and Zenghu Chang*

Since the early discovery of high harmonic generation, HHG, at the end of 1980s, most of the studies have been concentrated on rare gas atoms. There have been a number of experimental studies of HHG from molecules. In an earlier experiment, it was found that "the harmonic spectra from molecular gases are very similar to those obtained in atomic gases, with a plateau and a cutoff whose location is strongly correlated to the value of the ionization potential". As an example, the HHG spectra of O_2 are close to that of Xe. Their ionization potentials are comparable (~12 eV). Compared to the preliminary study of HHG from molecules, the ionization of molecules has been studied more extensively. Recently, it was found that the ionization of O_2 is suppressed by about one order of magnitude when comparing with Xe. However, there is no ionization suppression for N_2 when comparing with Ar, which has nearly the same ionization potential as N_2. As is known, HHG is closely related to the ionization in the intense laser. The suppressed ionization of O_2 should lead to a significant extension of harmonic spectra, which was not observed in the previous HHG measurements. Stimulated by such a controversy and the desire to further explore the relationship between HHG and ionization, we studied the HHG cutoff behavior for molecules and their companion atoms, for cases with a strong ionization suppression (O_2, Xe), and for cases with no ionization suppression (N_2, Ar).

The experiment was carried out with the newly established high intensity laser facility at the J. R. Macdonald laboratory. We compared Ar and N_2, which have nearly the same ionization potentials, at 15.76 eV and 15.58 eV, respectively, and we found they have nearly the identical harmonic cutoff (The cutoff positions for Ar and N_2 are q_c = 63 and q_c = 57, respectively). On the other hand, while Xe and O_2 have nearly the same ionization potentials, at 12.13 eV and 12.06 eV, respectively, the harmonic cutoff for O_2 (q_c=53) is much higher than for Xe (q_c=29). We attributed this to the O_2 molecule being much harder to ionize than the Xe although they have the same ionization potential. Therefore, the harmonic cutoff extension and the ionization suppression are strongly correlated. The cutoff extension caused by the ionization suppression provides another avenue for obtaining higher energy x-ray photons.

5.3 High Harmonic study with the OPA, *Bing Shan and Zenghu Chang*
Much effort has been devoted to extend HHG into the keV x-ray regime, which will enable x-ray spectroscopy studies of the inner shell electrons and diffraction/absorption studies of solids with sub-femtosecond resolution. It is well known that the cutoff photon energy scales as the square of the laser wavelength. The difficulty is to generate high intensity long wavelength pulses. We demonstrated that by using a long wavelength (~1.5 μm) infrared pump pulse with 10^{14} W/cm^2 from an optical parametric amplifier; the cut-off photon energies of Ar and Xe atoms were increased by a factor of two or more compared to that by a conventional 800nm pump laser. For Xe gas, the cutoff is ~40 eV with an 800nm pump and is ~80 eV with a 1.5 μm pump. It also shows that a harmonic wavelength can be easily tuned to cover any wavelength from the 5th order to the cutoff by changing the OPA wavelength. This tuning method is much easier to implement as compared to other approaches such as using two color laser pulses that involves a sensitive temporal and spatial overlap of the two pulses.

Future Plans
5.1 Laser system upgrade: We are building a high intensity optical parametric amplifier that will produce 10^{15} W/cm^2 intensity in the 1-1.6 μm wavelength range. We are also working on a hollow-core fiber compressor that will produce high intensity pulses with less than 10 fs duration.

5.2 High harmonic generation from molecules. We will generate high harmonics with two laser pulses. The first pulse is to align the molecules and the second pulse generates high harmonic radiation.

5.3 keV High harmonic generation with intense long wavelength field. With the new, more intense OPA, we will perform experiments on He and Ne gases and expect to extend the cutoff to the keV x-ray range. One of the major microscopic factors that affects that efficiency is the quantum diffusion of the electron wave packet in the laser field. Since quantum diffusion is sensitive to the change of the excitation wavelength, we will study this effect qualitatively by tuning the wavelength of the OPA.

6. Atomic physics with ion- and electron beams

S. Hagmann, [J. R. Macdonald Lab shagmann@phys.ksu.edu and Inst. für Kernphysik, Universität Frankfurt, Frankfurt, Germany s.hagmann@gsi.de]

The projects presented in this section cover advances in spectroscopy of few electron very heavy ions, a new access to (e,3e) spectroscopy of atoms, and delta electron spectroscopy for heavy ions traversing condensed matter.

Recent progress and future plans :

6.1 (A2.3) Electron spectroscopy of relativistic ions in the ESR storage ring

S. Hagmann (KSU and IKF, Frankfurt), Th. Stöhlker (IKF,Frankfurt), J. Ullrich, R. Moshammer (MPI, Heidelberg , Ch. Kozhuharov (GSI, Darmstadt)

We have designed a new electron spectrometer to be used in the ESR storage ring; its purpose is twofold: a) investigate the role of strong ($E \geq 10^{16}$ V/cm) and rapidly varying ($\tau \leq 10^{-18}$ sec) electromagnetic fields active in fragmentation of ions and atoms during relativistic collisions using high resolution projectile electron spectroscopy; b) investigate (e,2e) spectroscopy of ions in inverse kinematics using a combination of the electron spectrometer with a new reaction microscope.

The new forward electron spectrometer D60-QT-D60 analyzes electrons emitted near 0^0 around the beam direction with $v_e \cong v_{Proj}$. Taking advantage of relativistic kinematics allows for $\beta \cong c$ very high resolution spectroscopy of high lying autoionizing Rydberg states (e.g. with $n \geq 100$ in few electron U ions) in such collisions. We can thus study coherent states via anisotropies of Rydberg states and the corresponding asymmetry[1] of ELC and ECC Cusp peaks. For kinematically complete experiments, i.e. when jointly operated with the reaction microscope, we can reconstruct the scattering plane for (e,2e) experiments on ions performed in inverse kinematics. In the commissioning phase of the instrument we have shown that the spectrometer is effectively suppressing the intense beam-induced background and is transmitting only electrons of selected momenta onto the 2D position sensitive detector.

A first application after completion of this phase of commissioning will be a combined study of the shape of the ECC Cusp in coincidence with the Radiative Electron Capture into Continuum RECC photons- a process which is closely related to the elementary process of bremsstrahlung.

At least one new Ph.D student will be joining our group in fall 2002 to work on this topic.

1. J. Burgdörfer , Phys. Rev. **A33** (1986) 1578

6.2 Two-electron Lambshift in high-Z He-like Ions

A. Gumberidze, Th. Stöhlker, F. Bosch, Ch. Kozhuharov(GSI, Darmstadt) X. Ma(IMP Lanzhou), S. Hagmann(KSU and IKF, Frankfurt), Y. Zou(Fudan Univ, Shanghai)

We are using radiative recombination (RR) into the 1s state of bare and H-like U to measure with high precision 2-electron contributions to the ground state of He-like U and thus gain access to 2-electron QED effects. The experiment will be conducted in the electron cooler region of the ESR storage Ring and is taking advantage of the deceleration technique where the beam energy of the bare and H-like U ion is taken from 400 AMeV - where bare and H-like ions can be produced very efficiently - to 43.5 AMeV. With bremsstrahlung intensity much attenuated and Doppler corrections strongly reduced, this allows for an accuracy of 5eV for the total 2e contribution of 2.2 keV in U^{90+}.

6.3 He-double ionization via electron impact: (e,3e) spectroscopy

A. Knapp, L. Schmidt, R. Dörner, O. Jagutzki, H. Schmidt-Böcking(IKF,Frankfurt), S. Hagmann(KSUand IKF, Frankfurt)

We constructed a COLTRIMS type spectrometer that will allow us to measure kinematically complete 5fold differential ionization cross sections for double ionization of He under electron impact. At present the ion- and electro-optical properties of the entire spectrometer are mapped, first experiments will focus on single ionization of He, followed by studies of double ionization for low collision energies, where the projectile electron´s energy loss is significant. Here discrepancies with theory are large. The coincidence rates are estimated to be a factor 100 higher than in traditional configurations.

This is the Ph.D. project of A. Knapp

6.4 An electrostatic toroidal electron energy analyzer for delta electrons excited in collisions of heavy ions up to relativistic collision velocities

S. Dreuil , G. Kraft(GSI, Darmstadt) S. Hagmann(KSU and IKF, Frankfurt), H. Rothard(CIRIL, Caen)

In radiotherapy with relativistic heavy ions the success in cell deactivation and thus an effective treatment is largely based on understanding the details of the mechanism of energy deposition(via creation of free electrons) in the intracellular medium leading to double strand breaks of DNA.

The biological effective dose optimization at the core of inverse treatment planning[1] codes thus depends – as a key part of it – on the electron emission pattern observed for swift heavy ions passing through dense matter. Our goal is a systematic measurement of doubly differential cross sections for electron emission and of electron transport for a variety of condensed matter targets, emphasizing variations in solid state density and electron density to assess their respective influence on creation and transport of continuum electrons.

Two additional Ph.D students are expected to join this project later this fall.

1. W. K. Weyrather et al Int. J. of Radiation Biology 75(1999) 1357

6.5 Laser Spectroscopy of Hyperfine Transitions in very heavy H- and Li-like ions in the ESR

Th. Kühl, W. Nörtershäuser, A. Dax, M. Tomaselli, R. Sanchez , (GSI-Darmstadt), S. Hagmann(KSU and IKF, Univ. Frankfurt), Th. Stöhlker(IKF, Univ. Frankfurt)

The simplest and most basic magnetic interaction in atomic physics is the hyperfine splitting of the 1s ground state of a one-electron ion or atom. For very heavy ions around Pb and Bi QED effects - which amount to just 10^{-6} for protons - rise to several percent. Recently discrepancies have been found[1] between high resolution optical spectroscopy of hyperfine transitions and calculated wavelength both, for $^{207}Pb^{81+}$ and $^{209}Bi^{82+}$ in the ESR storage ring. In order to address questions with respect to a possible nuclear origin of the observed discrepancy, we are improving the experimental setup and measure Li-like 2s and H-like 1s hyperfine transitions for $^{209}Bi^{82+}$.

1.P. Seelig et al. Phys. Rev. Lett. **81** (1998) 4824.

7. Theoretical Studies of Interactions of Atoms, Molecules and Surfaces

C. D. Lin [cdlin@phys.ksu.edu; (785) 532 1617]

7.1 (B1) Tunneling ionization of molecules by intense laser fields

(X. M. Tong, Z. X. Zhao and C. D. Lin)

In conjunction with the new experimental initiative in the J. R. Macdonald Laboratory, we have also started a new theoretical program in intense laser interactions with atoms and molecules.

The ionization of atoms, especially rare gas atoms, by femtosecond Ti:Sapphire lasers has been well studied experimentally, and the ionization yield has been found to be well-described by the tunneling ionization model, or the so-called ADK model. In this model ionization by an intense laser field is calculated from the ionization rate of a static electric field, which can be approximately calculated in analytic form. The ADK model has been widely used by experimentalists to calculate the ionization rate of atoms.

When comparing the ionization rates of molecules by an intense laser, experimentalists have found that the ADK model works for some molecules, but not for others. For example, N_2 and Ar have similar ionization potentials and their ionization rates are about the same. On the other hand, O_2 and Xe also have nearly identical ionization potentials, but the ionization rate for O_2 is significantly smaller than that of Xe. Thus O_2 ionization is suppressed. The origin of ionization suppression of O_2 has been controversial. It has been "explained" in terms of many-electron effects [Guo, PRL85, 2276 (2000)], or in terms of the interference from the two atomic centers [Muth-Bohm et al, PRL85, 2280 (2000)].

We have succeeded in extending the ADK model to describe tunneling ionization of molecules. The basic idea is rather simple. Since the ADK model was derived for the one-center atoms, to apply it to molecules, one needs to extract the one-center parameters from the two-center (for diatomic molecules) molecular wavefunctions. By fitting the asymptotic electronic wavefunction of a molecule in proper form, we derived the parameters needed for the ADK model for molecules. With these parameters the ionization rates for each molecule can be obtained for any pulse shape, laser intensity and pulse duration, in analytic form, so that experimentalists can readily calculate the rates for molecules.

We have completed two papers on molecular tunneling ionization so far. The theoretical paper, which detailed the theory, has been submitted for publication. In this paper we also calculated the ratio of the ionization yield of a molecule with its companion atom, as a function of the peak laser intensity. The molecular tunneling model results are compared to the experimental data from Bob Jones' group in Virginia. We found general good agreement with their data.

The molecular tunneling ionization model predicts that most molecules would show ionization suppression if its valence electron is a π orbital. This is the case for O_2. Since ionization suppression means a higher saturation intensity to ionize the molecules, then the cutoff for the high order harmonic generation would be greater for O_2 than for its companion atom Xe. This has been shown to be the case experimentally. The first intense laser experiment carried out at the J. R. Macdonald Laboratory is the measurement of harmonic cutoff done by Z. H. Chang's group. Indeed the cutoff for O_2 was found to be twice that for Xe, but N_2 and Ar have essentially identical cutoffs.

Our molecular tunneling ionization model also predicts the dependence of the ionization rates on the alignment of molecules. In the coming year we will calculate the ionization yields of molecules that are aligned. It is well known that a short pulse can align molecules at each

"revival" time. We will predict the ionization rates of molecules in the short interval near revival, which can be tested by experiments. We expect the dependence for O_2 and N_2 to be quite different. We also intend to extend the molecular tunneling ionization model to include all internuclear distances. This would lead us eventually to study dissociative ionization of molecules.

7.2 (B2) Hyperspherical approach to ion-atom collisions at low energies

(C. N. Liu, A. T. Le and C. D. Lin)

Ion-atom collisions at low energies are usually carried out using the close-coupling expansion with molecular orbitals. This "standard" approach has serious difficulties since the theory is not Galilean invariant. In the past few decades electron translational factors were introduced in an *ad hoc* manner to account for such deficiency. The validity of such an approach is difficult to assess. We have developed a new approach for ion-atom collisions at low energies based on hyperspherical coordinates, which bypasses the need of electron translational factors. In the last month, we finally have all the programs developed and a first calculation on ion-atom collisions using hyperspherical coordinates has been carried out. We calculated the charge transfer cross sections for He^{++} on H from 500 eV down to about 10 eV in the center-of-mass frame, where the cross section drops by a factor of about 10^{10}. We have found that the results at low energies differ significantly from calculations based on the molecular orbitals where translational factors have been used. A manuscript is being prepared for this work.

7.3 (B3) Shakeoff theory revisited

(T. Y. Shi and C. D. Lin)

The shakeoff theory that has been used in the literature is all based on the calculations carried out by Aberg in 1970, which is valid only at the limit where the first electron leaves at an infinite velocity. We have correctly calculated the shakeoff theory prediction for He when the first electron is left with a high but finite velocity. With this new calculation, we showed that the ratio of double to single photoionization cross sections of He and the ratio of transfer ionization to single electron capture of He by high-velocity protons could be interpreted with the shakeoff model. A manuscript based on this work has been accepted for PRL.

7.4 (B4) Intershell triply excited states of atoms

(T. Morishita* and C. D. Lin)

Thus far we have been able to classify most of the intershell states in the $2\ell 2\ell' 3\ell''$ manifold of Li. We have found the additional quantum numbers that are needed as compared to the intrashell triply excited states. This work is still in progress since we are still unable to classify a number of the higher excited states within the manifold.

* Also at University of Electrocommunications, Tokyo, Japan.

7.5 (B5) Total and differential charge transfer cross sections from MOTRIM experiments

(Teck Lee and C. D. Lin)

We have used the close-coupling code to obtain total and angular differential charge transfer cross sections for ions on Rb targets that have been measured at the J.R. Macdonald Laboratory in DePaola's group. We have found general good agreement with experiments. But discrepancies were found at the lower energies. The angular resolution from the experiment is still not good enough to test the oscillatory structure predicted by the theory. In the low energy end we also see some discrepancy, which may be due to the limitation of calculations based on the atomic orbital model. We will use the newly developed hyperspherical code to repeat the calculations. Improved resolution from the experiment is also expected.

8. Interactions of Ions and Photons with Atoms, Surfaces, and Molecules

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8.1 (B9.2) Neutralization of negative hydrogen ions near metal surfaces

(T. Niederhausen, H. Chakraborty, U. Thumm)

We have applied wave-function propagation techniques to charge-transfer processes in ion-surface interactions, using self-consistent potentials to represent the electron-surface interaction. Apart from contributing to the qualitative understanding of the interaction mechanism through computer animations, this project has led to the quantitative assessment of (one-electron) charge transfer and to the characterization of surface resonances in terms of level shifts and decay widths as functions of the ion-surface distance. Using the split-operator Crank-Nicholson propagation in conjunction with carefully adjusted absorbing potentials near the numerical grid edges, we have performed 3D propagation calculations for the decay of negative hydrogen ions near metal surfaces.

In particular, we have completed calculations for the neutralization of negative hydrogen ions near two copper surfaces of different symmetries, Cu(100) and Cu(111). In these (adiabatic) calculations, we kept the ion-surface distance D fixed and propagated the electronic wave function. To start with, we used a 1D propagation scheme where the active electron is restricted to move along the surface normal. To model the electron-surface interaction, we used suitable parameterizations of self-consistent LDA potentials. For numerical convenience, we performed these calculations for a 100 monolayer thick Cu films and extended the numerical grid 200 atomic units on the vacuum side. In order to test the accuracy of our Cu potentials, we diagonalized the effective one-electron Hamiltonian and were able to reproduce the experimentally known energies of the upper and lower limits of the surface projected L-band gap, the surface-state energy, and the energies of the few lowest image states.

We obtained resonance positions and widths by first propagating the initial negative hydrogen state (one-electron approximation) for fixed D. Next, we calculated the projected density of states (PDOS) by Fast Fourier Transformation of the autocorrelation function, i.e., the overlap of the electronic wave function at time t with the initial electronic state. We repeated this calculation for various D and identified peaks in the PDOS as resonant states that asymptotically (for large D) correspond to the affinity level of the negative hydrogen ion, to image states, surface states, and bulk states of the 100 layer thick film. The lifetimes of those resonance states were extracted from the line widths of the corresponding peaks in the PDOS. We compared our results for the Cu(100) and Cu(111) surfaces with results for a structureless Cu-jellium potential.

By plotting the level shifts and widths for Cu(100), Cu(111), and Cu(jellium) as a function of D, we noticed several avoided crossings between image-state, surface-state and affinity-level resonances. Interestingly, the detailed behavior of both level widths and shifts differs strongly for decay in Cu(100) and Cu(111). Qualitatively, we understand this difference in terms of the different energetic location of image states, surface state, and surface band gap.

Future plans: We intend to extend our one-dimensional adiabatic propagation calculations to two and three dimension. We will try to quantify to what degree the inclusion of the active electron's motion in the surface plane affects resonance widths, and shifts. Next, we plan to include the motion of the projectile in order to provide neutralization probabilities for the scattering of negative ions near surfaces and to understand the interplay of elementary processes at different

time scales (decay into surface states, decay of surface states, effects of the electron's motion parallel to the surface, the role of lifetimes and interaction times).

8.2 (B10) Laser-Matter Interactions

(M. Alcantara Ortigoza, B. Feuerstein, U. Thumm).

We have investigated the interaction of 25 fs, 0.2 PW/cm^2, 780 nm pulses with H_2^+ molecular ions within a reduced-dimensionality model that represents both the nuclear and electronic motion by one degree of freedom. We carefully adjusted the adiabatic molecular electronic potential by introducing a "soft-core function" a(R) in the electron-nucleus interaction potentials $1/(x \pm R/2 + a(R))$ that depends on the internuclear distance R instead of the commonly used fixed soft-core constant a. We obtained molecular model potentials that reproduce accurate three-dimensional results for the known number of 19 vibrational states in the electronic ground state and for the dipole oscillator strength.

We solved the time-dependent Schrödinger equation on a two-dimensional numerical grid and designed a simple, but as far as we know new, method to calculate the flux of emitted electrons and protons by means of "virtual detectors" for electrons and protons. These detectors are placed outside the excursion range of the electron and at a distance R where the amplitudes of bound vibrational states have become irrelevant.

Our results reproduce the main features of measured kinetic-energy release spectra, support the "charge-resonance enhanced" ionization mechanism, and allow us to clearly distinguish between molecular dissociation (MD) into field-dressed final channels and fast, ionization-induced Coulomb explosion (CE). Both MD and CE appear as distinct peaks in the kinetic energy release spectra. We find that MD dominates for molecular ions that are prepared in the two lowest vibrational states only, while CE becomes increasingly dominating for higher vibrational states.

Future plans: For two short laser pulses of variable delay, we started to resolve in time the interplay between MD and CE. We intend to further investigate the pump-probe dynamics for two short pulses. We will explore the possibility of adding a further dimension to the electronic motion. Motivated by new experiments in the Macdonald Laboratory, we have stated to investigate the ionization of model atoms under the combined influence of a few-cycle intense Laser pulse (10^{14} to 10^{15} W/cm^2) and a significantly longer and less intense pulse (about 10^{12} W/cm^2). The two pulses may or may not be phase coherent.

9. Theoretical Studies of the Dynamics of Few-Body Systems

B.D. Esry[esry@phys.ksu.edu,(785) 532-1620]

9.1 (B.6) Time-Dependent Treatment of Continuum Phenomena

a. Charge exchange in $\alpha + H_2^+$ collisions. *S.C. Cheng and B.D. Esry*

My graduate student, Shu-chun (Amy) Cheng, has recently been working on solving the time-dependent Schrödinger equation for electron capture from H_2^+ by an ion. The calculations were motivated by an experiment carried out by C.L. Cocke's group in the JRM lab. The essential physics is that when the electron is captured from, H_2^+, the molecule will dissociate. Amy is propagating the Schrödinger equation in three-dimensions for the electronic wave function. The projectile is assumed to move along a straight-line trajectory, and the molecular nuclei are assumed stationary. At the collision energies relevant to the experiments, both assumptions are quite good.

To help her understand the propagation methods, Amy started working on a one-dimensional model of the system. In the model, the molecule is in line with the trajectory of the incoming ion. The approaching ion comes within some minimum distance of the molecular center of mass, then recedes along its entrance direction. We used this model to test both the propagation code and a masking function that damps all of the electron wave function save that on the molecular ion. Since we are only interested in the total electron capture, this approach is both sufficient and efficient. Amy has obtained the capture probability in this model as a function of the distance of closest approach and presented the results at a poster at DAMOP.

Future plans Amy is in the process of developing the three-dimensional code, and no problems are expected. Once the total electron capture probability is obtained for this case, the dissociation probability is known since they are the same. The angular and energy distributions of the molecular nuclei are then known as well. We can thus obtain a quantity that is directly comparable to experiment. We plan to begin generating the data soon. It will take some time, though, since the capture probability depends on five parameters: the impact parameter b, the orientation of the molecule θ and ϕ, the distance between the nuclei R, and the velocity of the incident ion v. In the end, we will integrate over b, ϕ, and R, weighting the integrals with the appropriate nuclear wave function. As long as only one electron is active, there is, in principle, no difficulty to include a polyatomic target, or even a molecular projectile. These cases will, of course, require careful modelling to reduce them to one active electron, but they could be quite interesting to study.

b. Ionization in ion-atom collisions *W. Guo and B.D. Esry*

We have worked on applying the scaled coordinate method to the time-dependent Schrödinger equation for ionization in ion-atom collisions. The advantage of the scaling method is that it analytically removes much of the known behavior of the ionized wave function, leaving only a smooth, stationary envelope to propagate. In particular, it alleviates the need for the absorbing boundaries normally used in such calculations.

The primary effort on this project came from my postdoc, Wei Guo. He arrived in August 2001 and worked to apply the scaling method to a one-dimensional model of the collision. Using this model gave us the opportunity to explore the scaling method for ion-atom collisions in an efficient manner since the one-dimensional model lready contains all of the elements peculiar to the scaling method. Wei eventually got the code working and began to generate data for the

one-dimensional problem. The project was not completed the prior to Wei's departure. A replacement, Vladimir Roudnev, has been hired and should arrive around October 2002.

Future plans. While the numerical aspects of this method require careful consideration, there appear to be no fundamental difficulties in extending the scaling to the three-dimensional ion-atom collision problem. My plans for the near future, however, do not include this project since other efforts appear more timely.

9.2 (B.7) Rearrangement Processes in Asymmetric Three-Body Systems

a. Protonium formation in $\bar{p}$+H(1s) collisions. *H. Sadeghpour (ITAMP) and B.D. Esry*

The low-energy collision of $\bar{p}$ with hydrogen is one of the most asymmetric systems one can consider. The difficulty lies in the fact that $\bar{p}$ and p can form bound states with a ground state energy of approximately –459 a.u. By comparison, the energy in the incident channel is --0.5 a.u. The formation of protonium, Pn or $\bar{p}p$, occurs when the proton is captured by the antiproton, ionizing the electron in the process. Based on simple energy arguments, the most likely Pn states produced are in the n=30 manifold. The extreme change in length and energy scales --- going from truly quantum mechanical in the initial state to nearly classical in the final state --- makes the problem extremely difficult to treat theoretically.

It is tempting to apply the Born-Oppenheimer approximation to this problem, and many people have. The Born-Oppenheimer curves, however, do not allow for the formation of Pn; the united atom limit is a free electron. We have instead used the adiabatic hyperspherical approach to generate potentials that do indeed correlate to the $\bar{p}$+H(1s) limit as well as all of the Pn($n\ell$)+e^- channels. Unfortunately, about 450 potential curves are required to cover the energy range of interest. This multitude of channels and the strong coupling between them is at the heart of the problems with asymmetric systems. To gain some insight into the physics, we have artificially reduced the masses of p and $\bar{p}$ and carried out the scattering calculations with these simpler systems. By examining several values of the mass, we could identify the important channels. This work has been submitted for publication and has so far resulted in two invited talks: one at an ITAMP workshop and the other at an ECT* workshop in Trento, Italy.

Future plans

I want to combine the physical insight gained in our scaled-mass calculations with a diabatic representation in order to reduce the number of channels in the full-mass problem. While we have recently completed the manuscript describing our split diabatic approach, another diabatic approach will likely be needed. The split diabatic representation requires one to identify all of the curve crossings by hand --- an unreasonable task with 450 channels. We will thus seek alternative diabatization schemes that can be applied more automatically. I anticipate that any method successful for this system can be profitably applied to other collision systems, allowing quantum mechanical calculations where none are now possible.

9.3 (B.8) Hyperspherical Approach to Four-Body Systems

Low-energy Ps+Ps collisions. *R. Krems (ITAMP), H. Sadeghpour (ITAMP), and B.D. Esry*

Roman Krems, a Predoctoral Fellow at ITAMP who is soon to be a postdoc there, recently spent two weeks at Kansas State to begin implementing my formulation of an adiabatic hyperspherical approach for the low-energy collision of two Ps (e^+e^-) "atoms". The only four-

body adiabatic hyperspherical treatments that have been done to date are for three electron atoms (see C.D. Lin's report). With an infinitely heavy nucleus, this system has a natural center about which to define approximately good angular momentum quantum numbers. The equal mass problem that we are considering, however, does not, making the necessary expansion over partial waves more slowly converging.

Future plans

The code seems to be nearly complete, but we are still struggling to achieve convergence in the partial wave expansion. Roman will continue to work on this problem as a postdoc.

FINANCIAL REPORT

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

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[2001 – 2002]

1) "Observation of a Nearly Isotropic, High-Energy Coulomb Explosion Group in the Fragmentation of D_2 by Short Laser Pulses,"
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M. Zamkov, H. Aliabadi, E. P. Benis, P. Richard, H. Tawara, and T.J.M. Zouros
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