Structure and Dynamics of Atoms, Ions, Molecules and Surfaces:
Atomic Physics with Ion Beams, Lasers and Synchrotron Radiation

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The goals of this aspect of the JRML program are to explore mechanisms of ionization of atoms, ions and small molecules by intense laser pulses and ions and to investigate the dynamics of photoelectron emission from small molecules interacting with x-rays from harmonic generation and synchrotron radiation.

Recent progress:

1) Momentum imaging of doubly charged ions of Ne and Ar in the sequential ionization region, *C. M. Maharjan, A. S. Alnaser, X. M. Tong, B. Ulrich, P. Ranitovic, S. Ghimire, Z. Chang, I. V. Litvinyuk, and C. L. Cocke*. One theme of "AMO ultrafast" science is to develop "clocks" for following the dynamics of atomic and molecular processes in real time. The project described here developed from our attempt to use the rotation of the electric field vector in circular polarization to measure the time interval between the emission of two electrons when an atom is doubly ionized by a short intense laser pulse. The plan was to measure the azimuthal angle between the momentum vectors of the two emitted electrons, using COLTRIMS and measuring the doubly charged ion and the two electrons in coincidence. This experiment has not yet been successful, due largely to the limitation of the repetition rate of our laser (1 kHz). However, we were able to gain important information concerning the sequential ionization process from the momentum spectra of the singly and doubly charged recoil ions of Ar and Ne when neutral gas targets of these elements were ionized by 8 fs pulses of circularly polarized 800 nm radiation at intensities approaching 10^{16} w/cm^2. The recoil momentum spectra of the singly charged ions, taken in the plane of the electric field vector, show the expected "donut" pattern, reflecting the momentum corresponding to the vector potential of the laser at the time of emission. The projections of these donuts onto the axis along which we have out best resolution are shown in fig. 1 for argon. A similar projection of the momenta of doubly charged ions, also shown in fig. 1, shows considerably more interesting and more informative structure. This structure can be interpreted as the folding together of two "donuts" , each with a radius corresponding to the magnitude of the vector potential at the time of emission of the electrons. Roughly speaking, the outer peak in the projection corresponds to the sum of the magnitudes of the vector potentials at the times of first and second ionization, and the inner peak corresponds to the magnitude of the difference of these vector potentials. This information, when combined with knowledge of the time-profile of the pulse, allows the deduction of the approximate times of ionization of the two electrons, although not with the resolution which the angular measurement would provide. The experimental momentum spectra were interpreted quantitatively by a model calculation of X.-M. Tong. This work is described in publication 3.

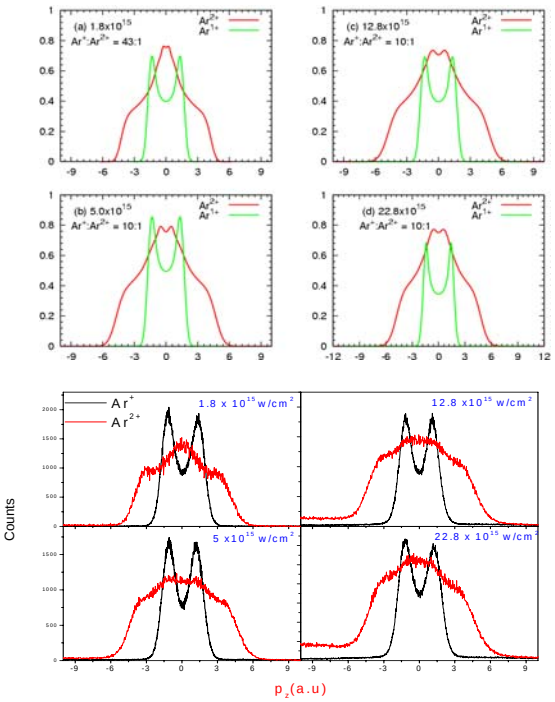

Fig. 1. (Lower half) Spectra of the momenta of singly and doubly ionized Ar ions projected onto the z-axis (along the collection field of the spectrometer) for circularly polarized radiation at four different peak laser intensities. (Upper half) Model calculations by X.-M.Tong of the p_z distributions, corresponding to the ionization data.

2) Wavelength dependence of momentum-space images of low-energy electrons generated by short intense laser pulses at high intensities, *C. M. Maharjan, A. S. Alnaser, I. Litvinyuk, P. Ranitovic, and C. L. Cocke*. Recent measurements of the momentum spectra of low energy electrons released from atomic targets by intense laser pulses, in an intensity region where tunneling ionization was expected to dominate, have shown marked structure in both electron energy and angle. The exact origin of this structure has not yet been unambiguously identified. Both "Freeman resonances" and diffraction are thought to play a role. In an attempt to provide experimental data on this subject, we used COLTRIMS to measure electron momentum spectra over wavelength range between 400 and 800 nm at high enough intensities that one might naively expect tunneling to dominate over multi-photon ionization. Complex structure was observed, as shown in fig. 2. By following individual features over a range of wavelengths, some of the energy structure could be associated with particular Rydberg resonances in the atom. Some could not. The angular structure was observed to maintain a preference for an angular momentum of 6 over a range of energy structures. The observed structures are too complex to be easily interpreted without a complete solution to the Schrödinger equation. Integration of theoretical calculations over the interaction volume is crucial in the interpretation. At least two groups, including that of C.D.Lin, are presently making progress in quantitatively interpreting this structure. See publication 3.

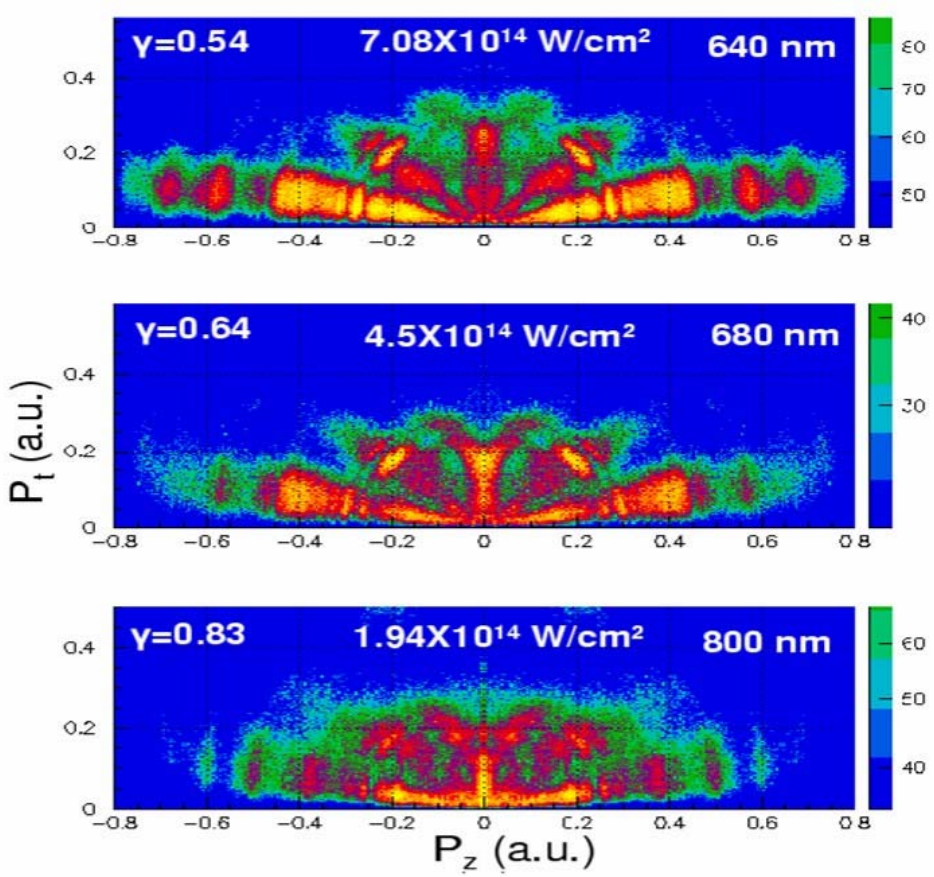

Fig. 2. 3D electron momentum distributions measured for single ionization of Ar. Horizontal axis corresponds to momentum component along the polarization direction, vertical axis represents value of electron momentum perpendicular to laser polarization.

Future plans:

3) Infrared pump/XUV probe work discussed in last year's progress report has continued. We have successfully produced photoelectrons from harmonics up to approximately 45 eV in a COLTRIMS geometry and are building a two-component mirror to allow time-resolved photoelectron spectroscopy. One of the initial experiments will be to try to launch a wave packet into the dissociative $2p\sigma_u$ potential curve of D_2^+ with an infrared pulse and to photoionize this state at a known later time with the XUV probe. This work is an extension of our infrared pump-probe work on H_2^+ and D_2^+discussed in last year's progress report and in publication 5. We will also use photoelectron spectra generated by the harmonics in the presence of the infrared field to characterize the time structure of the attosecond pulse trains represented in the harmonics.

4) Other future projects: We will continue our infrared pump-probe work using CEP stabilized pulses and investigate ways to control the dissociation/ionization routes in H_2, D_2 and other light molecules.

Publications appearing in 2005-2006 not previously listed:

1. "Photon-ion collisions and molecular clocks", T. Osipov, A. L. Alnaser, S. Voss, M. H. Prior, T. Weber, O. Jagutzki, L. Schmidt, H. Schmidt-Böcking, R. Dörner, A. Landers, E. Wells, B. Shan, C. Maharjan, B. Ulrich, P. Ranitovic, X. M. Tong, C. D. Lin, and C. L. Cocke, J. of Modern Optics 52, 439 (2005).

2. "Simultaneous real-time tracking of wave packets evolving on two different potential curves in H_2^+ and D_2^+", A. S. Alnaser, B. Ulrich, X. M. Tong, I. V. Litvinyuk, C. M. Maharjan, P. Ranitovic, T. Osipov, R. Ali, S. Ghimire, Z. Chang, C. D. Lin, and C. L. Cocke, Phys. Rev. A 72, 30702 (2005)

3. "Momentum imaging of doubly charged ions of Ne and Ar in the sequential ionization region," C. M. Maharjan, A. S. Alnaser, X. M. Tong, B. Ulrich, P. Ranitovic,S. Ghimire, Z. Chang, I. V. Litvinyuk, and C. L. Cocke, Phys. Rev. A 72 041403 (R) (2005).

4. "Resonant excitation during strong-field dissociative ionization," A. S. Alnaser, M. Zamkov, X. M. Tong, C. M. Maharjan, P.Ranitovic, C. L. Cocke, and I. V. Litvinyuk, Phys. Rev. A 72 041402 (R) (2005)

5. "Wavelength dependence of momentum-space images of low-energy electrons generated by short intense laser pulses at high intensities," C. M. Maharjan, A. S. Alnaser, I. Litvinyuk, P. Ranitovic, and C. L. Cocke, J. Phys. B: At. Mol. Opt. Phys. 39 1955 (2006).

6. "Momentum-imaging investigations of the dissociation of D_2^+ and the isomerization of acetylene to vinylidene by intense short laser pulses," A.S.Alnaser, I.Litvinyuk, T.Osipov, B.Ulrich,A. Landers, E.Wells, C.M.Maharjan, P.Ranitovic, I.Bocharova, D.Ray and C.L.Cocke, J. Phys. B: At. Mol. Opt. Phys. 39, S485(2006).