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

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

Recent progress and future plans:

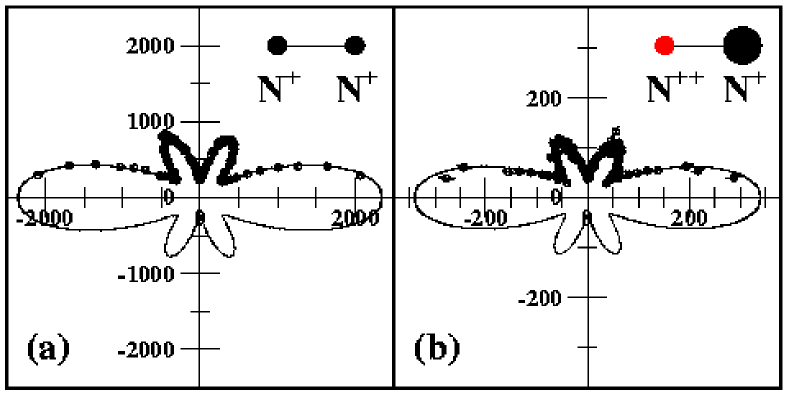
1) Photoelectron diffraction from light molecules fixed in space *A.Landers (Western Mich.Univ.), R.Doerner, Th.Weber, M.Hattass, O.Jagutzki, A.Nauert, A.Staudte, H.Schmidt-Boecking(U.Frankfurt), M.H.Prior(LBL), A.Cassimi(CIRIL), T.Osipov, I.Ali, C.L.Cocke (KSU)*. We measure the diffraction patterns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

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

A summary of some results for CO and N_2 is:

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

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



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

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

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

3) Non-sequential decay of doubly ionized D_2 formed by electron capture by slow Xe $^{26+}$ projectiles *I. Ali, R. D. DuBois*, R..Olson*, S. Hagmann, C. L. Cocke.* When a diatomic molecule is fragmented by electron capture by a slow, highly charged ion, the ion can remain in the vicinity of the molecule during the fragmentation phase, and will thus distort the momentum distribution of the fragments from that of a freely disintegrating molecule. The time scale involved is near ten fs, comparable to that involved in the dissociation of D_2 by intense laser pulses. We have measured the kinetic energy release of the fragments emitted from the dissociation of D2 in collision with slow Xe $^{26+}$ ions from the KSU CRYEBIS. The energy of the fragments is calculated from the measured momentum vectors of both fragments emitted in the dissociation of the diatomic molecule using a three dimensional momentum imaging multi-hit detector system. The energy is measured separately for the ion pairs from the dissociation of the molecule into different dissociation channels. Clear distortion of the Coulomb sphere is observed, and correlated with the collision plane of the reaction. The results are in good qualitative agreement with CTMC calculations for this reaction. *Univ.of Missouri-Rolla

4) Electron capture from H_2^+ by doubly charged projectiles *I. Reiser, H.Braeuning(U.Giessen) ,C.D.Lin, E.Sidky and C.L.Cocke.* To what extent can a single electron capture from a two-center target be treated in terms of interfering amplitudes for capture from two separate atomic centers? This question has often been addressed within atom-ion collisions, but the experiments have often been rendered difficult to interpret by the fact that the target was not a true one-electron system. The purpose of this experiment is to address this question with a true one-electron molecule, namely H_2^+. The experiment is thus electron capture from H2+ by singly charged projectiles (Ar $^{2+}$, N $^{2+}$ and He $^{2+}$, and is being carried out both on the JRM Ion-Ion collision facility and on the Univ. of Giessen Ion-Ion facility. The latter facility is being used to measure total cross sections for the capture, and the former, differential cross sections. The total cross section measurements show that the cross sections for the two heavy projectiles are much larger, since these reactions are nearly resonant, while the cross section for the He^{2+} (endoergic) is substantially smaller. The results are in good agreement with a simple model based on interfering amplitudes from separated centers. The atomic amplitudes were calculated using a coupled-channel atomic expansion approach. If this model is correct, the differential cross sections are expected to reveal two-center interference patterns. The differential cross section experiment with N^{2+} projectiles is being carried out at KSU. Preliminary data show strong alignment effects, but further data analysis is required to extract complete cross sections as a function of the angle between beam and molecular axis. This project is the Ph.D. thesis work of Ingrid Reiser. Future plans are to complete the data set for N^{2+} , He^{2+} and Ar^{2+}.

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