

A Momentum Microscope Views Many-Body Dynamics

How has nature created all the wonderful complexity surrounding us from such simple ingredients as positively charged nuclei and negatively charged electrons? What coordinates all the subtle motions and changes that animate our world on a microscopic level? The general answer to these questions comes from the fact that electrons do not move individually and independently of each other but mostly in a highly correlated fashion; that is, the motion of one electron affects the motions of the others through the Coulomb force that causes like charges to repel and opposite charges to attract. An international team working at the ALS has constructed a unique apparatus to study this correlated motion and applied it to the emission of two electrons from a deuterium molecule after absorption of an x ray. This is the first time that the correlated electron emission from a molecule has been mapped in complete detail.

Dynamical electron correlation—the fact that changes in electron motion, or more precisely their quantum-mechanical wavefunctions, occur in a highly correlated fashion—is determined by the complex, many-particle, momentum exchange via the Coulomb force between the charged particles (electrons and ions) in an atom or molecule. This momentum exchange is constrained by quantum-mechanical symmetry conditions imposed on all composite systems by their total angular momentum and parity.

A long-term project at the ALS is yielding new insight into this fundamental problem of electron correlation by investigating in as much detail as possible simple few-body processes that can serve as paradigms for more complicated cases. To do this, an endstation equipped with a novel momentum-space microscope (termed COLTRIMS) is used. It has been developed by a collaboration among

Universität-Frankfurt, Kansas State University, and Berkeley Lab. This device provides multidimensional images of the correlated momenta of all charged particles after photofragmentation of an atom or small molecule. Subatomic resolution and high detection efficiency are achieved for each electron and ion, no matter in which direction they may be moving.

Using this unique tool, we have investigated the ejection of two electrons from helium and neon. More recently, we have applied this technique to study the complete fragmentation of a hydrogen (deuterium) molecule by absorption of a single photon. The final state for this process consists of two protons and two electrons. The momentum vectors of each of the four particles have been measured in coincidence. The fact that a single photon can couple to two electrons at all is a remarkable consequence of electron correlation. In a world of isolated, uncorrelated electrons, the probability for coupling one photon to two electrons would be zero.

After absorption of the photon, the two electrons are emitted very quickly. On a much longer time scale, the two heavy nuclei then fly apart back-to-back along the molecular axis because of the repulsive Coulomb force between them. Thus, detecting the two nuclei yields the orientation of the molecular axis at the instant of ionization. With this trick one can map the correlated emission pattern of the two electrons from a “fixed-in-space” molecule.

In a first deuterium experiment at the old Beamline 9.0.1 (which later moved to become Beamline 10.0.1), we measured the energy and angular distribution of one of the two electrons with respect to the two heavy nuclei. Then, in two successive experiments at Beamline 7.0.1, both nuclei and both electrons were finally measured (Figures 1 and 2). This is the first time that the correlated electron emission from a molecule has been mapped in complete detail. Astonishingly, the gross features of the two electron wave functions do not depend strongly on the molecular orientation and are somewhat

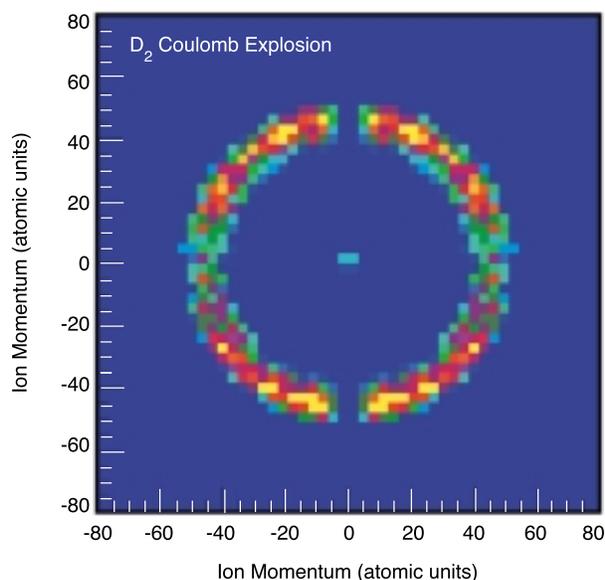


Figure 1

Momentum distribution of the D^+ fragments from photo-double-ionization of deuterium by 58.8-eV linearly polarized light. The electric field vector is horizontal, and the light propagates in the vertical direction. The gap in the “ring” is caused by inability to separate two ions arriving at the detector at nearly the same time.

similar to the emission pattern from a helium atom. The two new molecular effects to be seen are interference due to the two indistinguishable centers of the molecule and the consequences of a transfer of angular momentum from the electronic to the nuclear wave function.

The plot represented in Figure 2 is only one of many ways to visualize the results of these measurements using the COLTRIMS momentum microscope. The power of the technique allows investigators to view the correlations among the particle momenta without enforcing *a priori* restrictions on any of them.

Investigators

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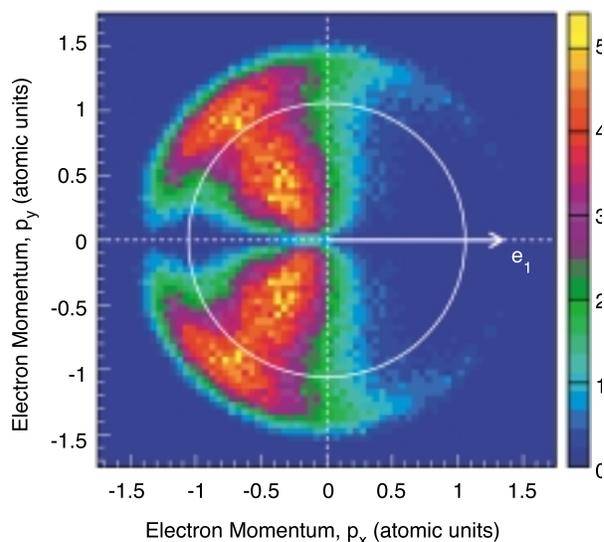


Figure 2

Correlated two-electron momentum distribution after double ionization of deuterium by photons 25 eV above the double-ionization threshold. In this figure, the momentum vector of one electron points along the horizontal axis to the right (arrow), so the momentum distribution shown is that of the second electron. Electron repulsion is responsible for the dominant emission of the two electrons into opposite half spheres. The node for back-to-back emission is a result of the odd-parity symmetry of the final-state electron wave function. The data are integrated over all orientations of the molecule and all directions of the polarization axis.

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Funding

German Federal Ministry for Basic Research (BMBF); German Science Foundation (DFG); Alexander von Humboldt Stiftung; Habilitanden Programm der DFG; and U.S. Department of Energy, Office of Basic Energy Sciences.

Publications

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Symmetry Resolution Yields Evidence for Shape Resonance

Many people are familiar with acetylene as a welding fuel, but it also plays an important role in industrial chemistry. Acetylene molecules that are fixed (adsorbed) onto a specially prepared surface serve as the foundation for the synthesis of certain types of plastics, such as polypropylene. To achieve the desired end result, the initial conditions of the acetylene molecules on the surface must be well understood. It would therefore be extremely useful to have a simple way to determine the properties of these molecular building blocks. Shape resonances are spectroscopic features that have shown promise for probing the geometry of molecules that are not in the gas phase. Now, the first convincing evidence for the existence of a shape resonance in acetylene has been obtained at the ALS through high-resolution photoelectron spectroscopy, where small differences in the energy of molecular orbitals can be resolved.

Shape resonances in photoelectron spectroscopy were first proposed to exist 15 years ago. Shape resonances occur when photoelectrons are temporarily trapped by a potential-energy barrier in the molecule. Because the shape of the potential depends upon the configuration of the molecule, it was initially thought that shape resonances might be a simple way to obtain information about molecular geometry. In particular, it was thought that the

energy at which a shape resonance occurred could be correlated with the length of molecular bonds.

An attempt was made 15 years ago to quantify the relationship between shape-resonance energy and bond length for a number of molecules, including acetylene. However, shape resonances were manifested as broad, somewhat ambiguous peaks in the photoelectron cross section, and the criteria for identifying shape resonances in general were unclear. Subsequent studies found only weak evidence for a shape resonance in acetylene. But now symmetry-resolved photoelectron spectroscopy measurements have finally provided the first convincing evidence for the existence of a shape resonance in acetylene.

The high brightness of undulator radiation and the high resolution afforded by Beamline 10.0.1 (old Beamline 9.0.1 was moved to sector 10 in mid-1998; data were obtained before and after the move) made it possible to see features of the photoelectron spectrum that were not readily apparent in previous work. As a result, it was possible to resolve the carbon 1s peak of acetylene into contributions from electrons of g (symmetric) and u (antisymmetric) character (Figure 3).

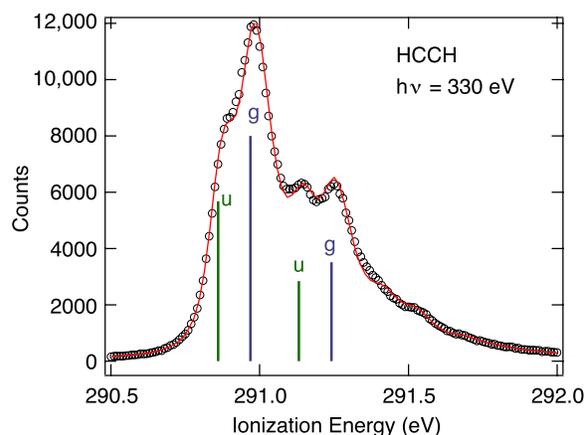


Figure 3

Symmetry-resolved photoelectron measurements show relative intensities of $1\sigma_u$ and $1\sigma_g$ ionization. The circles are experimental data, and the solid line is a fit to theory with the assumption that carbon-carbon stretching is the only vibrational mode contributing. The vertical bars indicate the position and intensity for ionization with vibrational quantum number $v = 0$ (left pair of bars) and $v = 1$ (right pair of bars).