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Towards momentum-space imaging of electronic and nuclear motions in molecules

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Abstract

The momentum distribution of electrons and atoms (nuclei) in a molecule provides a sensitive probe of molecular orbital shape [1, 2] and nuclear quantum effects [3]. It can be measured by utilizing Compton scattering of high energy electrons (> keV). Electron momentum spectroscopy (EMS [1, 2]) utilizes electron Compton scattering that most nearly corresponds to the collision of the incident electron with the target electron to be ionized. Accordingly, EMS can measure electron momentum distributions for individual electron orbitals. On the other hand, atomic momentum spectroscopy (AMS [4, 5]) utilizes electron-atom Compton scattering where the instantaneous motion of the scattering atom causes a Doppler broadening in the energy of the scattered electrons. One can obtain with AMS the information on the momentum distribution of atoms with different masses. Advances in highly-sensitive multichannel spectrometer opened up new possibilities of EMS and AMS, such as EMS study on photo-excited molecules [6] and precise mapping of the intramolecular H-atom motion [7]. Recently, we have made theoretical [8] and experimental progresses to broaden the applicability of EMS/AMS to molecular science. Experimentally, we have developed a new highly-sensitive electron spectrometer that can be employed for both EMS and AMS studies. Results of EMS study on relativistic effects on electron orbital shape, as well as those of AMS study on elements heavier than hydrogen (i.e., carbon and nitrogen) will be presented to demonstrate and discuss the ability of the developed apparatus.

The momentum distribution of electrons ejected from molecules can be mapped in the molecular frame by means of photoelectron-photoion coincidence velocity map imaging spectroscopy [9]. We have measured with a linearly-polarized femtosecond laser (~10^{13} W/cm^2) recoil-frame photoelectron angular distributions (RF-PADs) for above threshold ionization of CH_4 and CF_4. It was found that the RF-PADs can be partly interpreted by anisotropy of tunnel ionization followed by recollision with the ion. Therefore, RF-PADs offers a direct imaging of photoionization dynamics of molecules in intense laser fields.

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