

Visualizing Ultrafast Chemical Reactions by Coulomb Explosion Imaging and Photoelectron Holography 1a-TH-3

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Understanding molecular dynamics in chemical reaction is one of the central subjects of chemistry. Here, we discuss two novel approaches to visualize directly how molecules change their structure in the course of reactions by using few cycle intense laser pulses and the high-order harmonics in the soft X-ray region with an unprecedented time resolution. First, we present time-resolved Coulomb explosion imaging of ultrafast acetylene (DCCD) – vinylidene (D₂CC) isomerization of C₂D₂²⁺ by intense few-cycle laser pulses (<10 fs, ~1 PW/cm², 800 nm) [1]. A pair of ultrashort intense laser pulses (9 fs, ~1 PW/cm², 800 nm), generated by a pulse compressor consisting of a hollow fiber and dispersive mirrors, is employed as pump and probe pulses. The pump laser pulse is used to ionize the target molecule as well as to trigger the nuclear dynamics, whereas the probe laser pulse, introduced with a time delay Δt, ionizes the molecule further to induce the three-body Coulomb explosion, C₂D₂³⁺ → D⁺ + C⁺ + CD⁺, from the triply charged parent ion. The momenta of the resultant fragment ions are determined in the molecular frame for each explosion event by using the coincidence momentum imaging method [2]. The correlation among the fragment momenta and its dependence on the time delay Δt shows that the migration of a deuterium atom proceeds in a recurrent manner: The deuterium atom first shifts from one carbon site to the other in a short time scale (~90 fs), and then migrates back to the original carbon site by 280 fs, in competition with the molecular dissociation.

In the second part, we present the development of a new reaction imaging system based on laser high-order harmonics (HH). The HH source generates the harmonics of the fundamental laser pulses (800 nm) in the soft X-ray region (~91 eV@59th) with high photon flux 3×10⁹ photons/s and small divergence (1.1mrad). The 59th HH pulses are then separated from other harmonics by a pair of Mo/Si dielectric mirrors for the innercore ionization of molecules. The pulse duration is estimated to be 1.8 fs at the Fourier transform limit from the 4d photoelectron spectra of Xe. Inner-core photoelectrons are emitted from specific atomic sites and then scattered by surrounding atoms within the molecule. The interference between the scattered electron wave and the original (unscattered) wave forms a holographic pattern that precisely records the position of the surrounding atoms. The time-resolved photoelectron holography being developed here, which directly captures the ultrafast changes in the molecular structure in the course of the reaction, will promote a deeper understanding of the interaction between molecules and intense laser fields, and provide a key to realize reaction control with high efficiency.

References

[1] A. Hishikawa, A. Matsuda, M. Fushitani, E. J. Takahashi, *Phys. Rev. Lett.* **99**, 258302 (2007)

[2] A. Hishikawa, E. J. Takahashi, A. Matsuda, *Phys. Rev. Lett.* **97**, 243002 (2006)

[3] M. Fushitani, A. Matsuda, E. J. Takahashi, A. Hishikawa, *J. Phys. Conf. Ser.* submitted

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