Observing electron localization in a dissociating molecule in real time

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We observe real-time dynamics of electron localization and bond breaking in dissociating H_2^+ by performing a few-cycle pump-probe experiment. We determined that in that molecule in our experimental condition electron localization happens 15 fs after the initial ionization of neutral H_2.

Dissociation of diatomic molecules with odd number of electrons always causes the unpaired electron to localize on one of the two resulting atomic fragments. In the simplest diatomic molecule H_2^+ dissociation yields a hydrogen atom and a proton with the sole electron ending up on one of the two nuclei. That is equivalent to breaking of a chemical bond – the most fundamental chemical process. Here we observe electron localization in real time by performing a pump-probe experiment where both pump and probe are 5 fs pulses with controlled carrier-envelope phase (CEP). The more intense pump pulse ionizes a neutral H_2 molecule and initiates dissociation of H_2^+. The much weaker time-delayed probe pulse drives the electron motion in the dissociating molecule. By measuring the asymmetry of proton emission as a function of pump-probe delay we demonstrate that direction of electron localization can be controlled by the probe pulse only within the first 15 fs following ionization of H_2. For larger pump-probe delays direction of electron localization is completely determined by CEP of the pump pulse.

In our experiment, CEP-locked linear polarized ~5 fs few-cycle pulses with central wavelength of ~ 750 nm is diverted into a Mach-Zehnder interferometer to produce pump-probe pulse pair with a controllable delay. The pulses are tightly focused inside the Reaction Microscope (REMI) apparatus on a neutral H_2 supersonic jet, and full 3-dimensional momenta of ion fragments are measured by the REMI. The pump pulse ionizes the neutral H_2 and produces a dissociating nuclear wavepacket (NWP) via radiative coupling between $1s\sigma_g$ and $2p\sigma_u$ states. The delayed CEP-stabilized probe pulse is introduced during the dissociation process to drive the electron motion and affect the direction of electron localization [1].

The measured energy-resolved asymmetry of electron localization as a function of delay is shown in figure 1, and the data which are taken with the pump pulse only are shown on the far left side of the figure for comparison. The spectrum is characterized by a strong modulation of the asymmetry and change of its direction for delay times of less than 15 fs, and a much weaker modulation and constant negative direction of the asymmetry for larger delays. .

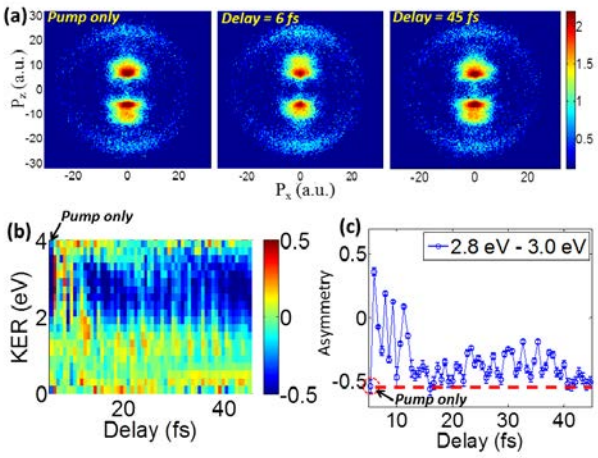

Figure 1 (a) Measured proton momentum distribution in laser polarization plane with pump pulse only (left), for pump-probe delay of 6 fs (middle) and for delay of 45 fs (right). (b) Measured delay dependent energy-resolved asymmetry of proton emission. The data for zero delay (indicated by the arrow) are measured with the pump pulse only. (c) Measured delay dependent asymmetry of proton emission for KER from 2.8 eV to 3.0 eV. The red dashed line indicates the value of the asymmetry measured with pump pulse only.

References

[1] M. F. Kling, et al., *Science* 312, 246 (2006).

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