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Attosecond Precision Pump-Probe Experiments: Visualizing Molecular and Electronic Dynamics

JOACHIM ULLRICH, Max-Planck-Institut für Kernphysik, D-69117 Heidelberg

A series of time-resolved experiments is reported with the goal (i) to map, completely characterize and finally to control femtosecond nuclear motion in simple molecules, (ii) to generate and observe ultra-fast electronic wave-packets and, (iii) to study correlated sub-fs few-electron dynamics in strong-field multiple ionization. For that purpose, we have developed a unique combination of a 'reaction microscope' spectrometer (with an integrated Li-MOT target) imaging the complete many-particle final-state momentum space and a pump-probe setup providing two 7 fs, ca. 0.1 PW/cm**2 pulses at variable delays between 0 and 3300 fs, reaching absolute and relative precisions as good as 70 as and 1 as, respectively. (i) Via Coulomb explosion imaging we reconstruct the time-dependent probability density of the dissociating, rotating and vibrating nuclear wave-packets in the most fundamental molecular systems, the hydrogen and deuterium molecular ions. We observe the 'collapse' and 'revival' of their vibrational wave packets, investigate their composition via Fourier analysis, show novel routes to directly visualize field modified potential curves yielding a complete characterization of the field-induced ultra-fast molecular dynamics and, most recently, study the formation of hydrogen molecular ions in laser-induced fragmentation of methane. A one attosecond relative accuracy is demonstrated mapping the vibrational motion in the neutral deuterium molecule and the corresponding excitation mechanism is identified by determination of the absolute quantum phase of the motion. (ii) Using a Li-MOT target as an effective one-electron target we have coherently populated and observed fast electronic wave packets in low-lying states. (iii) For multiple ionization of atoms recoil-ion momentum distributions allow us to distinguish different ionization pathways and to reveal first time information on few-electron emission. For Ne we observe signatures of highly correlated recollision-induced three- and four-electron processes measured to occur on a 500 as time scale.