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Attosecond pre-determination of reaction dynamics in polyatomic molecules[1]

MARKUS KITZLER, Vienna University of Technology

An important aim of Ultrafast Laser Science and Attosecond Physics is the measurement of valence electron dynamics in molecules during complex restructuring and fragmentation reactions. Such reactions may be triggered by the removal of electrons, e.g., by ionization with intense, ultrashort laser pulses. Depending on the valence-shell from which the electrons are removed, the molecular ion might be put into a binding state or a certain dissociative state. With control over the ionization process it might thus be possible to gain control over the subsequent restructuring and fragmentation processes on a purely electronic level. Detailed insight into the process of electron removal can be obtained from interferometric measurements using two bound state wavepackets released at different times within a sub-cycle of a laser field. I will present results of experiments that exploit this kind of electron wavepacket interferometry with sub-10 attosecond resolution for tracing the evolution of the phase of the bound state of an atom or molecule during the removal of an electron. I will furthermore present two conceptually similar schemes that allow pre-determining the outcome of molecular restructuring and fragmentation processes in polyatomic molecules on sub-femtosecond time-scales by gaining control over the process of electron removal. The first method involves recollision double ionization in intense few-cycle laser fields with a known carrier-envelope phase (CEP). Tuning of the CEP allows controlling the removal of inner-valence electrons and the controlled population of dissociative excited states. Using this method I will show experimental CEP-control over various fragmentation reactions of a series of polyatomic molecules. The second control scheme uses the strong preponderance of ionization from specific molecular orbitals to the alignment of the molecular axis with respect to the laser polarization direction for determining which valence level the electrons are removed from. I will demonstrate experimental control over different fragmentation pathways of the acetylene molecule using the field-free alignment angle as a control knob.

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