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Ultrafast Transient Absorption and Photoelectron Spectroscopy with High Order Harmonics[1]

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Laser-produced high order harmonics are used to probe chemical dynamics of atoms and molecules on femtosecond timescales. Two basic methods are developed, ultrafast transient absorption and photoelectron spectroscopy. The high order harmonics are produced with an 800 nm Ti:sapphire laser focused into a capillary or rare gas jet. Both inner shell core levels and outer shell valence states are investigated. The transient absorption of xenon ions produced by high field ionization of neutral xenon atoms is probed by core level spectroscopy. The alignment of the vacancy created in forming the ion is measured as a function of pump-probe delay by promotion of an inner d electron to the vacancy in the outer shell. Small molecules are excited to repulsive dissociative states and individual harmonics are used to obtain time-resolved photoelectron spectra. A wave packet on the dissociative state of bromine molecules is detected, as well as the production of atoms at longer time delays. By the use of velocity map imaging, the angular distributions of outgoing photoelectrons are analyzed. In a new experimental system, carrier-envelope phase-stabilized few-cycle pulses will be used to create attosecond pulses of the high order harmonics, to study electronic-time-scale processes in atoms and molecules.

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