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Attosecond probing of state-resolved ionization and superpositions of atoms and molecules.¹

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Isolated attosecond pulses in the extreme ultraviolet are used to probe strong field ionization and to initiate electronic and vibrational superpositions in atoms and small molecules. Few-cycle 800 nm pulses produce strong-field ionization of Xe atoms, and the attosecond probe is used to measure the risetimes of the two spin orbit states of the ion on the 4d inner shell transitions to the 5p vacancies in the valence shell. Step-like features in the risetimes due to the subcycles of the 800 nm pulse are observed and compared with theory to elucidate the instantaneous and effective hole dynamics. Isolated attosecond pulses create massive superpositions of electronic states in Ar and nitrogen as well as vibrational superpositions among electronic states in nitrogen. An 800 nm pulse manipulates the superpositions, and specific subcycle interferences, level shifting, and quantum beats are imprinted onto the attosecond pulse as a function of time delay. Detailed outcomes are compared to theory for measurements of time-dynamic superpositions by attosecond transient absorption.

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