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Isolated attosecond pulse probing of atomic and molecular dynamics¹

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Extreme ultraviolet pulses are produced by high order harmonic generation and used to probe ultrafast and attosecond atomic and molecular dynamics. Methods to produce and characterize isolated attosecond pulses are discussed. Time-dynamic processes are probed by core level transient absorption spectroscopy in atoms that absorb the extreme ultraviolet light. These so-called “reporter” atoms, even when incorporated into molecules, exhibit characteristic transitions that are altered by charge state and electronic state, and the transitions also exhibit dramatic shifts upon vibrational excitation. With removal of electrons from multiple specific orbitals in molecules, correlated dissociative dynamics is characterized. With isolated attosecond pulses, processes of high field ionization resulting in alignment, light induced states, Autler Townes splitting, and coherent superpositions of electronic and vibrational states are investigated on femtosecond and subfemtosecond timescales. Results are compared to theory, and fundamental challenges regarding the use of transient absorption methods for time-dynamic attosecond processes are discussed.

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