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Atomic and Molecular Dynamics by Attosecond Four Wave Mixing¹

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Isolated attosecond pulses in the extreme ultraviolet combined with near infrared pulses from a Ti:sapphire laser are used to initiate and measure electronic and vibrational superpositions and decaying states in atoms and small molecules. Especially, the background-free method of attosecond four-wave mixing allows a new level of time-dynamic analysis, and multidimensional methods with near infrared pulse shaping can be used to isolate individual states. Attosecond pulses create massive superpositions of electronic states in atoms as well as vibrationally selectable states in molecules, and few-femtosecond decay dynamics are obtained due to autoionization or competing predissociation. Alignment-dependent core-hole decays in molecules reveal the role of nonlocal interactions on decay processes. Detailed outcomes are compared to theory.

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