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Imaging electronic motions in atoms by energy-resolved ultrafast electron diffraction¹ HUA-CHIEH SHAO, ANTHONY F. STARACE, University of Nebraska-Lincoln — We present a general analysis of energy-resolved ultrafast electron diffraction for imaging target electronic motion and numerical simulations of time-resolved spectra of ultrafast electrons scattered from the breathing, wiggling, and hybrid modes of electronic motion in the H atom². We consider pump-probe processes in which a laser pulse creates a coherent superposition of target states that are probed by the electron pulses. Varying the pump-probe delay time, the delay-dependent scattering intensities record the ensuing electronic motions. The kinematics of the scattered electrons is fully resolved; both the scattering angles and the kinetic energies are measured. Therefore, besides the spatial and temporal information, the energy content of the electronic motions can be retrieved from the energy-resolved diffraction patterns, which provide unequivocal interpretations of the electronic motions. Because of this, we are able to explain the counterintuitive temporal behavior of the diffraction images, which show a quite different temporal behavior and little connection to the electron densities.

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