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Abstract for an Invited Paper for the DAMOP19 Meeting of the American Physical Society

Imaging electronic and molecular motions by ultrafast electron diffraction and impact ionization¹ HUA-CHIEH SHAO, University of Nebraska - Lincoln

Owing to advances in ultrafast electron technologies, it is now possible to generate and manipulate fs electron pulses with sub-ångström de Broglie wavelengths. Therefore, it is now feasible to directly image and visualize electronic and molecular transient motions during reactions. We have theoretically studied and explored various schemes for utilizing ultrafast electron pulses to image electronic and molecular motions [1-6]. In particular, time-resolved (e, 2e) electron momentum spectroscopy has been considered as a means of probing the momentum profile of electronic motion in lithium atoms [6]. Specifically, we have studied the adiabatic population transfer of the electronic state of the Li atom from the ground state (2s) to the first excited state (2p) driven by a chirped ps laser pulse. During the population transfer, the time-varying valence electronic motion is imaged by time-delayed 100- and 1-fs electron pulses through the mechanism of high-energy impact ionization. The simulations show that the momentum distribution of the valence electron at the moment of collision can be retrieved from the time-resolved spectrum. However, the level of detail of the information about the motion depends on the pulse duration and the time scale of the electronic motion.

Recently, we have studied ultrafast electron diffraction as a means of imaging the oriented ro-vibrational motions of deuterated lithium hydride (LiD) and hydrogen (HD) molecules. The molecular motion is assumed to be initiated by a pump pulse that impulsively excites an electron from the ground state to some excited electronic state. Then the ensuing molecular motion in the excited state is imaged by 1-fs electron pulses. The simulated diffraction images show a delay-dependent ring pattern owing to the interfering scattering amplitudes from the constituent atoms as the molecule vibrates. Moreover, the centrosymmetry of the diffraction images is violated in time-resolved measurements, which exhibit asymmetric angular distributions that relate to the direction of motion of the atoms.

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