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Time-resolved Pump–Probe Spectroscopy to Follow Valence Electron Motion

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Synopsis After an initial few-cycle UV pump pulse, the time-dependent spatial locations of holes and particles produced in the valence level can be probed by exciting core electrons.

One of the major endeavors of chemical physics over the last decades has been the study of microscopic dynamics on ever shorter time scales. Indeed, the time-resolved study of nuclear motion is now a staple field of science, made possible by femtosecond laser pulses that excite transitions among electrons, which move on time scales faster than the nuclei.

Recent advances in light sources, such as the free-electron laser (FEL) and high-harmonic generation (HHG), make it timely to consider the next natural step, which is to probe the fastest time scales relevant to chemistry, the motions of valence electrons. Anticipating the experimental realization of attosecond pulses [1] with photon energies of a few hundred eV to 1 keV [2, 3, 4], we have developed a simple theory which connects the evolution of a non-stationary electronic state (presumed to be created by a few-cycle UV pump pulse) to an XUV/X-ray probe signal.

The electronic states we wish to follow evolve on time scales of a few to several femtoseconds. The essential principle is that the dynamic valence occupancy structure of these states can be probed, resolved in both space and time, by taking advantage of the inherent locality of core–valence transitions and the comparatively short time scale on which they can be produced [5].

The formalism is built in terms of the time-dependent single-particle reduced density matrix, and it can therefore be supported by any *ab initio* model for the electronic structure, as well as provide a compact theoretical framework for understanding experimental results. We have applied it to dynamic density matrices obtained from time-domain electronic structure calculations at the extended, second-order algebraic diagrammatic construction [ADC(2)-x] level [6].

The transition from the complexities of many-body theory to an intuitive picture of dynamic local occupancy structure will be presented along with some key numerical results, which we hope to compare with future experiments.

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