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# Ultrafast x-ray probes of inner- and outer-shell electron dynamics

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X-ray free electron lasers potentially have the pulse duration and intensity to probe electron motion on the intrinsic timescales found in atoms and molecules, as characterized by the Bohr orbital period of ~150 attoseconds. While sub-orbital period dynamics for deep inner-shell electrons may still out of reach using direct time-domain probes, readily available few femtosecond x-ray pulses are well suited to characterize the relaxation pathways of the highly energetic hole state created by x-ray photoionization, i.e. the competition between the atomically localized Inner-shell decay (Auger emission, fluorescence), intramolecular charge dynamics and Coulomb explosion.

For studies of molecular inner-shell dynamics, we have taken advantage of accelerator-based developments at LCLS [1,2] that engineer two femtosecond x-ray pulses with adjustable duration, wavelength and time delay to probe the first steps following an inner-shell photoelectron ejection event [3,4]. Inner-shell dynamics in two prototypical molecules have been studied using the x-ray pump/x-ray probe methodology: XeF2 via recoil-ion spectroscopy, and, CO via photoelectron spectroscopy, demonstrating both the potential power and limitations of these methods.

For studies of outer-shell electron dynamics, we use the more standard optical pump/x-ray probe configuration. Here the strong-field optical pump generates a distinctive ion signature by impulsively removing an electron from a high-lying orbital to open an isolated x-ray resonance characteristic of the valence hole that can then be used to track its subsequent dynamics [5,6]. We will report on an XFEL-based experiment that addresses the origins of the long-lived electronic coherence in strong-field ionized water previously observed in all-optical experiments that track the hydrated electron [7].

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