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Probing ultrafast x-ray induced inner-shell processes.[1]

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Short and intense pulses produced by x-ray free electron lasers (XFELs) initiate complex processes when they interact with matter through inner shell ionization, resonant excitation or even non-linear interaction. The ability to follow these processes in time is a continuing challenge, but it is of high importance to be able to understand quantitatively the basic mechanisms at play during any measurement using intense x-rays. In this talk, I will illustrate progress towards that goal using two separate approaches involving detection of direct photoelectrons and/or Auger electrons to track the inner-shell decay mechanism. The first approach takes advantage of the ability to produce pairs of x-ray pulses with a well-defined delay and different colors. By measuring the time-dependent chemical shifts, one can access the changing chemical environment due to electronic and nuclear rearrangement following inner shell interaction with a few-femtosecond resolution. The second approach exploits the electron energy modulation in the presence of a strong linearly polarized laser field. Using a self-referenced analysis, decay mechanisms in atoms and small molecules have been studied with sub-femtosecond resolution. Both approaches are poised to take advantage of the upcoming LCLS-2, with full wavelength tuning ability in two-pulse mode, high repetition rate and reliable attosecond pulse generation.

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