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Capturing ultrafast molecular dynamics with time-resolved x-ray absorption, x-ray emission, and x-ray scattering[1]

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Ultrafast, time-resolved, laser-pump, x-ray-probe experiments are powerful tools for understanding and controlling the behavior of matter at the molecular level. Transient structural changes, both geometric and electronic, of single molecules after excitation by a laser pulse can be probed with high resolution and within complex or disordered environments, such as gases and liquids, taking advantage of the superior spatial resolution, elemental specificity and penetration power of x-rays. Third generation synchrotron sources, particularly the Advanced Photon Source (APS), provide x-rays with a unique combination of properties that are well suited for precision time-resolved measurements. These include a high flux (10^{13} photons/second/0.01% bandwidth) that is distributed in short pulses ($\sim$100 ps) with moderate intensity ($\sim 10^6$ photons/pulse) at a high repetition rate (MHz). Over the last decade laser-pump, x-ray-probe studies have been carried out at synchrotrons but a major challenge has been the low repetition rate (kHz) of standard amplified lasers resulting in underutilization of the synchrotron's high flux. This limitation has recently been removed with the installation of a high repetition rate laser system at 7ID-D at the APS. In this talk I will discuss measurements on the light-induced switching of Fe(II) complexes at 3.26 MHz pump-probe repetition rates which efficiently use the available x-ray flux. This efficiency enabled the complementary techniques x-ray absorption spectroscopy (XAS), x-ray emission spectroscopy (XES) and liquid phase x-ray scattering (XRS) to be used simultaneously to collect information on the structural and electronic dynamics on the picosecond time scale.

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