Table-top Ultrafast X-ray Spectroscopies using a Laser Plasma X-ray Source and Microcalorimeter Sensors

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We present recent successful demonstrations of table-top Time-Resolved X-ray Absorption Spectroscopy (TR-XAS) and Emission Spectroscopy (TR-XES) using a laser plasma x-ray source and a 240 element microcalorimeter array. Using TR-XAS, we studied the photoreduction of ferrioxalate, a reaction that has been the subject of a long-running debate in the literature, including contradictory x-ray measurements. Our results \cite{1} strongly support a picture in which reduction of the central iron is complete by 100 ps and contradict a theory in which the photoreduction occurs on much longer timescales. Using TR-XES, we studied spin cross-over in photoexcited iron tris-bipyridine and accurately measured the lifetime of the quintet state from simultaneous observations of the iron K\textalpha and K\beta features \cite{2}. We also determined the time resolution of our apparatus to be better than 6 ps. Better time resolution in TR-XES has only been demonstrated at two x-ray free electron lasers. These results are the first laboratory-scale demonstration of ultrafast TR-XES. They were enabled by a multi-institution collaboration and by the unique combination of spectral resolution and collecting efficiency provided by microcalorimeter sensors \cite{3,4}. In particular, the collecting efficiency of these devices allowed the quintet lifetime to be measured using 100-1,000\times fewer x-rays delivered to the sample than comparable work performed at a synchrotron.

Finally, we describe the planned use of microcalorimeter sensors for time-resolved x-ray science at large light sources. Building on previous deployments of microcalorimeter spectrometers to three synchrotrons for static x-ray science, our team, together with collaborators at SLAC and Stanford University, has recently been selected to build a first-light instrument for the Linac Coherent Light Source upgrade (LCLSII). The instrument will employ recently developed GHz readout technology and is intended to be scalable to several thousand sensing elements. Delivery of the detector package from NIST to SLAC is tentatively scheduled for 2019 with science operations planned to begin in 2020.

References: