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

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We present 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 [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 Kα and Kβ features [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 three 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 [3,4]. In particular, the collecting efficiency of these devices allowed the quintet lifetime to be measured using 100-1,000× fewer x-rays delivered to the sample than comparable work performed at a synchrotron.

Finally, we present a systematic study of the valence-to-core x-ray emission lines for different titanium compounds using cryogenic sensors. With an energy resolution of approximately 4 eV at the Ti Kβ lines, we measured the $K\beta_{2,5}$ features of different titanium compounds and compared our results to previously published synchrotron data. The use of microcalorimeter sensors allowed us to simultaneously measure how compound chemistry affects the titanium Kα and Kβ emission lines, something that is not possible using wavelength dispersive detection techniques.

[1] G. O'Neil, L. Miaja-Avila, Young Joe, et al., J. Phys. Chem. Lett. **8**, 1099 (2017).

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[3] J. Uhlig, W. Doriese, J. Fowler, et al., J. Synchrotron Radiat. **22**, 766 (2015).

[4] Young Joe, G. O'Neil, L. Miaja-Avila, et al., J. Phys. B **49**, 024003 (2016).