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High Fidelity Ultrafast Time Resolved X-ray Absorption Spectroscopy of the Insulator to Metal Transition in VO2

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We introduce a novel method for soft x-ray absorption spectroscopy (XAS) featuring high sensitivity and compatibility with time resolved ultrafast short-pulse FEL experiments. As a demonstration, we have measured the optically induced insulator to metal transition in VO2 with time resolved XAS. Probing the oxygen electronic system, we observe a fast sub-100 fs response in VO2 followed by a slower, few picosecond, final transition into the metallic state. The hundred-fold increase in sensitivity realized by our new XAS method enabled careful measurements of VO2, not simply as a function of photon energy and response time, but also excitation fluence. Our results provide a glimpse into the spectroscopic data quality expected from future high repetition rate XFEL sources.
Our new method employs nanofabricated diffractive optics to divide a monochromatic x-ray beam into a reference beam to provide an Io and an identical beam used to illuminate the sample. Both beams are independently detected on a low noise area detector (CCD). The sensitivity of the CCD enables single photon sensitivity and superb linearity. Consequently, the transmission of the sample is measured with a precision approaching the counting statistics. In our measurements we are able to achieve 0.2% sensitivity with a single x-ray pulse. This enables the rapid high quality data collection that is required for systematic measurements.

We expect this method will be used in the future to study dilute liquid systems by transmission through sheet jets. Our demonstration will also open up new possibilities to study thin film solids including transition metal oxides. This method will benefit from increased repetition rate at next generation XFEL sources.

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