

Ultrafast X-ray Science at the ALS: Resolving Atomic and Electronic Structural Dynamics

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X-rays are ideal probes of atomic structure since they interact with core electronic levels that are closely bound to the atomic nucleus. Moreover, x-rays offer important advantages for probing electronic structure (valence states, bonding geometry etc.) via transitions from core levels with well-defined symmetry and element specificity. An important frontier in x-ray research is the investigation of atomic and electronic structural dynamics in condensed matter using techniques such as XANES and EXAFS on the fundamental time scales of electronic scattering processes, ~ 1 fs, and atomic vibrational periods, ~ 100 fs. These are the relevant time scales for understanding the interplay between atomic structure and electronic properties in condensed matter. In correlated electron systems, time-resolved x-ray studies are instrumental to understanding the coupling between charge, spin, and lattice degrees of freedom, which give rise to remarkable material properties. In molecular systems, time-resolved measurements on the time scale on which chemical bonds are formed and broken, and on which conformational changes occur, promise to provide new understanding of chemical reaction dynamics.

We have developed a novel technique for generating femtosecond x-ray pulses from the Advanced Light Source (ALS) using ultrashort laser pulses to manipulate the temporal and spatial structure of stored electron bunches[1, 2]. Based on this technique, we have constructed a simple bend-magnet beamline for time-resolved x-ray spectroscopy. Current research on this beamline is focused on time-resolved NEXAFS measurements in the correlated electron system VO_2 and on molecular crystals of the spin-crossover complex $[\text{Fe}(\text{tpen})]^{2+}$. VO_2 undergoes an insulator-metal transition, accompanied by a structural transformation from monoclinic to rutile, which can be optically induced on the sub-picosecond time scale. The Fe(II) complex undergoes a $\Delta S=2$ spin transition within 500 fs of optical excitation, and this is accompanied by a dilation of the ligand cage of the Fe ion. The goal of our present research is to apply NEXAFS and EXAFS spectroscopy on the femtosecond time scale to elucidate the relationship between electronic and atomic structural dynamics in these systems.

In order to meet the stringent flux requirements for future x-ray spectroscopy experiments on the femtosecond time scale, we are developing an undulator beamline at the ALS which will provide ~ 200 fs duration x-ray pulses in the 0.3-10 keV range. The facility will incorporate a high repetition rate femtosecond laser system with an average power approaching 100 W. This contribution will provide a status report of current time-resolved x-ray experiments, and describe the development of the new femtosecond undulator beamline at the ALS.

References

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- [2] R. W. Schoenlein, S. Chattopadhyay, H. H. W. Chong, T. E. Glover, P. A. Heimann, C. V. Shank, A. Zholents, and M. Zolotarev, Science, **287**, 2237, 2000.