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Capturing Ultrafast Molecular Motions and Lattice Dynamic Response in a Spin Crossover Thin Film Using Femtosecond Electron Diffraction and X-ray Diffraction

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Structure determines function, and one of the great dream experiments in science is to directly observe atomic motions during the defining moment when matter transforms from one state to another and witness the key reaction modes directing chemistry. X-ray diffraction (XRD) on solid-state samples provides direct information on how atoms specifically respond and rearrange to stabilize the excited state. Structural dynamics from XRD and electronic dynamics from optical and X-ray spectroscopy are complementary to fully understand photochemical mechanisms. The European XFEL provides capabilities to carry out these studies, in particular high photon flux with ultra-short X-ray pulses, high repetition rates, and extreme focusing. In the Hamburg research landscape, the ultrafast electron diffraction (UED) at REGAE and the XRD at EuXFEL have proven to be invaluable resources for investigating how atoms specifically respond and rearrange to stabilize the excited state in solid-state materials.

In this contribution, we show experimental data obtained by two complementary methods, including UED and XRD at the EuXFEL beamline FXE, as a showcase of the potential application of these two new ultrafast probes. The developments of MeV-electron and X-ray free electron laser (FEL) facilitates the application of UED and ultrafast XRD with sub-100 fs time resolution, high signal-to-noise and momentum transfer (q) resolution. Therefore, it is now possible to simultaneously monitor the molecular motions and the lattice volume changes in quasi-crystalline SCO nanoparticle of well-controlled size and morphology with such high temporal resolution.

The results on a novel SCO materials Fe(HB(tz)3)2 (HBTZ) (tz= 1,2,4-triazol-1-yl) nanocrystalline thin film [1-2] unveiled a detailed picture of the transient coupling between spin-state switching and vibrational degrees of freedom, along with the development of nanometric strain wave on thin films upon photoexcitation. Due to the epitaxial orientation of the thin film with the c-axis perpendicular to the surface and the high electron energy, only Bragg peaks of the (h k 0) family have been detected. This renders it impossible to refine the atomic motions during the HBTZ phase transition and study the lattice volume expansion along the c-axis (Fig. 1b). However, from our electron diffraction results, one noteworthy feature is a plateau behavior for time delay 2 ps to 12 ps (Fig. 1c), which is after the initial photoinduced SCO and before the latter lattice response. This is indicative of the formation of the photoexcited high-spin state in unrelaxed lattice. We further carried out the time-resolved XRD experiments at the FXE end-station of the European XFEL. Thanks to the high q-resolution in low q-range of X-rays, we have been able to study changes in Bragg peak positions from more crystal lattice planes. Therefore, we could elucidate the development of nanometric strain on the thin film in ultrafast timescale (Fig. 1d and 1e).

1. Ridier, K. et al. Phys. Chem. Chem. Phys. 20, 9139–9145 (2018).
2. Ridier, K. et al.. Adv. Mater. 31, 1901361–1901361 (2019).

I plan to submit also conference proceedings

No

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