

STUDYING ULTRAFAST PHOTO-INDUCED MESOSCOPIC PHASE TRANSITIONS

Phenomena such as insulator-to-metal phase transitions (IMTs), colossal magnetoresistance, and high-temperature superconductivity often involve the spontaneous appearance of spatially separated regions having distinct structural, magnetic, and electronic properties. The IMT of vanadium dioxide (VO_2) is of particular interest, as the correlated-electron material exhibits an orders-of-magnitude change in electrical conductivity across the transition, in addition to a change in lattice symmetry. This close-to-room-temperature transition can be triggered by optical excitation on ultrafast time scales, and so promises important applications in low-power ultrafast electronics. The ultrafast nature of the transition is also of fundamental interest because this seemingly simple binary oxide compound may serve as a model system that provides new insights into high-temperature superconductivity, charge stripes, quantized spin and charge fluctuations, and ferroelectricity. Dynamic solid-solid phase transitions involving spatially separated regions that span length scales from nanometers to micrometers are very difficult to characterize because critical information underlying the phase transitions, such as localized phase competition, cannot be resolved by measurements that are spatially averaged over many phase-separated regions. An international team of researchers working at the APS developed a state-of-the-art laser-pumped x-ray diffraction imaging technique for just such applications.

The time-resolved x-ray diffraction microscopy technique, which has 350-nm spatial resolution and 100-ps temporal resolution (Fig. 1a), was applied to quantitatively studying structural phase propagation during a photo-induced phase transition in a VO_2 thin film. The experiment, which was conducted at the XSD 7-ID-B,C,D beam-line of the APS, represents the first hard x-ray measurement with sub-nanosecond and sub-micrometer resolutions that directly captures a mesoscopic structural phase transformation in a correlated material. The sub-micrometer length and sub-nanosecond time resolutions allowed the scientists from Argonne, the University of Wisconsin-Madison, the IBM Almaden Research Center, and the Max Planck Institute for Microstructure Physics (Germany) to follow the structural phase progression

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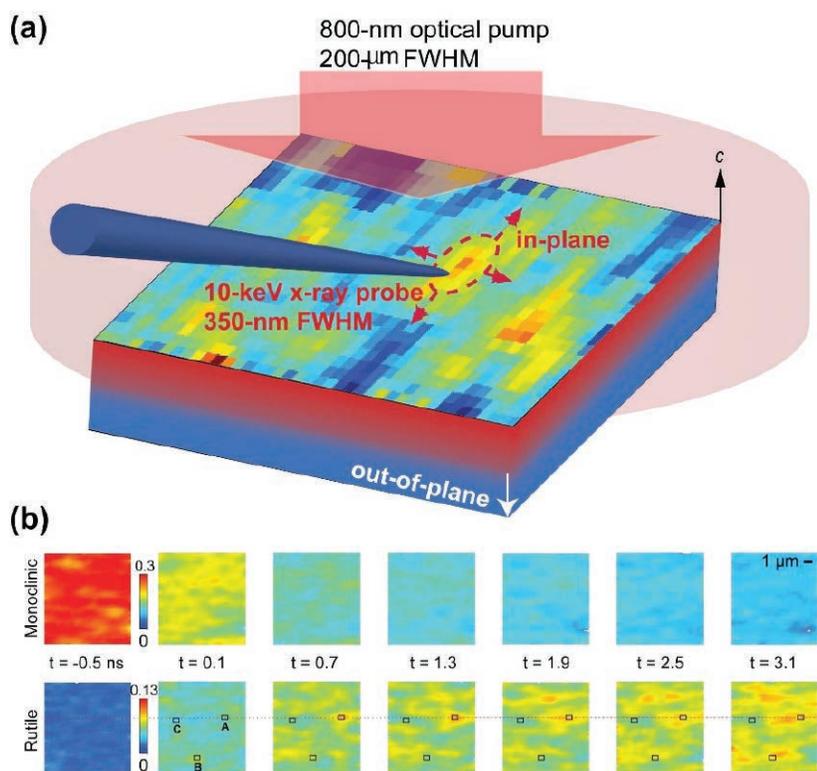


Fig. 1. (a) Experimental setup of the laser pump/hard x-ray diffraction microscopy probe technique. The structural phase progression of a VO_2 film along in-plane and out-of-plane directions, indicated by red and white arrows, respectively, is probed by synchrotron-based focused x-ray pulses upon homogenous optical excitation along the in-plane direction. The blue and red regions represent monoclinic (M) and rutile (R) phases respectively. (b) The intensity maps of the M and R phases measured at a sequence of time delays excited by an optical pulse with a fluence of $1.4 \text{ mJ}/\text{cm}^2$. The x-ray diffraction intensity from the M phase regions decreases after laser excitation, while the R phase regions emerge at discrete sites (e.g., site A) and spread out in the thin film. The color bars show the normalized diffraction intensity.

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demonstrates that vwBCDI preserves the strain-sensitive 3-D imaging capability of current rocking-curve-based BCDI methods without requiring any sample motion. This capability will greatly simplify certain *in situ* strain measurements in environments in which it is difficult to accurately rotate the sample about a precise center of rotation or is otherwise cumbersome. Currently the new method does not incorporate the energy dependence of the scattering factor. Thus, vwBCDI scans should be performed far away from absorption edges of the elements in the sample. However, enabling element-sensitive vwBCDI may be feasible with near-edge energy scanning, if additional resonant scattering effects are incorporated into the phase-retrieval algorithm. — *Vic Comello*

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during the photo-induced phase transition.

Time-resolved mapping of the real-space structural transformation was achieved by tracking the Bragg diffraction signatures of the structural phases. The first stage in the phase transition is a symmetry change from the monoclinic (M) state to the rutile (R) state at isolated nucleation sites on the unit-cell level, which occurs on a femtosecond time scale following optical excitation (Fig. 1b). These nucleation sites possibly relate to both local strain states that favor the stabilization of the high-temperature R phase and variations in grain size, since smaller single crystalline particles require less activation energy for the phase transition.

At this point, the optical excitation has created an unstable high-energy M state in which the energy to supply the latent heat for the transition to the R state has already been stored at the electronic level. The second stage occurs when the lattice is superheated above the transition temperature a few picoseconds later, as a result of electron-phonon coupling during which the absorbed radiation energy is transferred to the lattice. At this stage, the M state film has sufficiently high energy for the structural phase transition, but a lattice symmetry change has not yet occurred. The third stage is a domino-like lattice transformation propagating from the nucleation sites into the excited but not yet transformed M state film. This process is a displacive lattice transformation, in which the lattice symmetry change results from straightening V–V atom pairs without diffusive heat transport. The displacive lattice transformation occurs at a speed that is faster than in-plane thermal diffusion but slower than the sound speed in VO₂.

The visualization of phase transformations by means of the time-resolved x-ray imaging technique opens new opportunities for studying ultrafast nanoscale phase transitions and separations in correlated materials. Further studies using the same technique on different VO₂ thin films will reveal, for example, the dependence of phase progression speed on grain size and crystalline orientations. These insights are important not only for understand-

ing mesoscale phase transformations but also for designing ultrafast optoelectronic devices using VO₂. For example, faster structural phase transformations as a whole may occur in VO₂ with more nucleation sites, while a faster phase front progression that approaches the sound speed may occur in ultrathin VO₂ films that contain fewer grain boundaries. — *Vic Comello*

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