Time-Resolved Dynamics of the Electrically-Triggered Phase Transition in VO₂ Probed Using MeV-UED

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Vanadium oxide (VO₂) is an exciting *functional material* that shows large changes in its optical and electronic properties across the insulator-metal transition (IMT). It is particularly significant for applications in the emerging area of brain-inspired (or neuromorphic) computing, where the IMT is triggered electrically ('E-IMT') in a nanoscale device using a fast voltage or current pulse. Understanding the dynamics of this transformation is crucial not only to uncover the fundamental mechanisms that govern the electrically-triggered phase transformation, but also to determine the ultimate limits of switching speed in futuristic computing devices. Here we report the development of a new time-resolved capability at the mega electron volt ultrafast electron diffraction (MeV UED) facility, that can resolve ultrafast structural dynamics in micro/nano devices under electrical excitation. This stroboscopic technique is based on in situ diffraction using ~3 MeV, ~100 fs electron pulses, with a temporal resolution limited only by the electrical transients of the device. Using this novel approach, we make the first simultaneous dynamical measurements of structure and transport during the electrically-triggered IMT in VO₂. Our results provide new insights into the mechanisms (e.g. the role of Joule heating) as well as pathways (e.g. the presence of new phases) underlying the transformation. Furthermore, by also probing the dynamics of the ultrafast photoinduced IMT with 800 nm femtosecond pulses, we make direct connections between the transient structures formed under electrical and optical excitation over a range of timescales from µs down to ps. Through comprehensive measurements, we have not only gained a deeper understanding of the nature of E-IMT in VO₂, but also established this pulsed "electrical pump" scheme as a new mode of operation at UED.