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Ultrafast Dynamics in the Insulator-to-Metal Phase Transition of VO₂ via Attosecond Transient Absorption

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Abstract: The extreme ultraviolet absorption of vanadium dioxide is probed with attosecond transient absorption during a photoinduced phase transition. Measurements reveal an ultrafast rearrangement in the density of states suggestive of electron correlation and Mott physics.

OCIS codes: (160.0160) Materials; (320.0320) Spectroscopy; (300.0300) Ultrafast optics

The mechanism and driving force of the insulator-to-metal phase transition (IMT) in vanadium dioxide (VO₂), and the accompanying crystallographic rearrangement, has been the subject of controversy since its discovery in 1959. Electron correlation effects and a structural instability have both been hypothesized to play a role in the creation of a 0.7 eV band gap in the material below 340 K. The electronic picture supports a Mott-type mechanism, where upon exciting sufficient free carriers the nuclear potential is screened, and the bound and localized d states in the valence band are no longer supported. The structural picture alternatively suggests a Peierls-type mechanism, where a Jahn-Teller like structural instability is the origin of the band gap, and changes to the band structure follow from the motion of the ions [1].

To gain access to the dynamics and changes in electronic structure that occur both before and after the lattice has time to respond, and start to separate these effects, attosecond transient absorption measurements are performed on solid state VO₂ thin films. The IMT is initiated by a few-femtosecond carrier-envelope-phase stabilized near infrared (NIR) pulse (6.5 fs FWHM, 760 nm central wavelength) focused to 30 mJ/cm². The absorption of a time-delayed, collinearly propagating attosecond pulsed light source (produced by high harmonic generation in krypton) is measured around the vanadium M-edge with an extreme ultraviolet (XUV) spectrometer. Static M-edge spectra are additionally measured for both insulating and thermally induced metallic phases, and compared with the photoinduced changes.

In the transient measurements, as shown in Figure 1, a fast change in absorbance is observed across a broad spectral range surrounding the vanadium M-edge at 39 eV [3], where this energy corresponds to the spacing between the vanadium 3p core level and the Fermi level. While changes occur over a >30 eV bandwidth, the strongest two features are a rise from 38-45 eV and a bleach from 46-50 eV. These prominent features grow in within the first 15 fs, and remain roughly constant over the remaining 250 fs time window measured.

Figure 1. Transient changes in the XUV absorption spectrum of VO₂

The change in absorption upon photoexcitation can be compared to the static absorption spectrum, as well as the changes upon heating as shown in Figure 2, in order to see these effects in context. Both the heating and photoinduced changes show an increased absorption at and above the Fermi level with respect to the vanadium 3p core level, at 39 eV. This is consistent with an increased density of states at that energy, as would follow from a collapse of the band gap, and would be consistent with recent photoemission experiments which saw electron density appear in the bandgap within their pulse duration of 60 fs [3]. Additionally, the broad bandwidth of the changes (compared to the NIR pulse energy of 1.6 eV) is indicative of strong electron correlation and screening effects, as opposed to simple carrier excitation and the subsequent state opening / blocking.

Figure 2. A comparison of photoinduced changes with the static absorption spectrum and thermally induced changes. The static spectrum is plotted on the right hand y-axis, as indicated by the arrow.

A more full analysis will be done to disentangle changes to the density of states from screening effects that lead to small energy shifts of the core levels. However, it is apparent that changes in certain spectral regions converge to match the thermal phase completely within the first 30 fs, pointing to an electronic origin of those spectral signatures, where other regions must wait for lattice motion before converging to the thermal phase. This highlights the utility of attosecond transient absorption as a powerful tool for separating electronic and structural effects on the ultrafast timescale in complex systems.

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