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# Phase Transition in strongly-correlated VO<sub>2</sub>: Time-domain Assignment of Cause and Effect.

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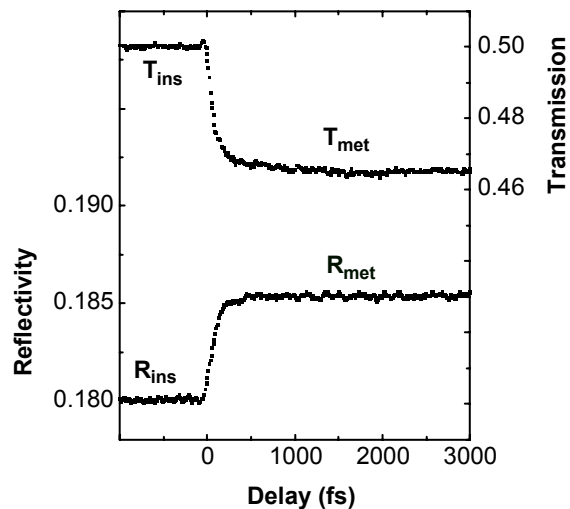
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**Abstract.** We establish time-domain hierarchy between structural and electronic effects in the strongly correlated system VO<sub>2</sub>. The insulator-to-metal transition is driven directly by structural change rather than by electron-electron correlations.

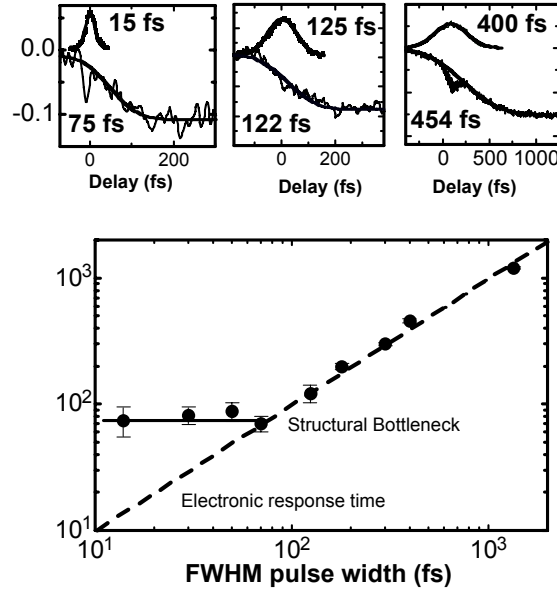
We study VO<sub>2</sub>, a strongly-correlated compound that exhibits cell doubling in “concomitance” with electron localization and a metal-insulator transition below 340 K [1]. Previous time-resolved optical [2] and x-ray diffraction [3] experiments demonstrated that impulsive photo-excitation of the low-T insulator causes an ultrafast transition in both electronic properties *and* atomic structural arrangement toward those of the high-T metal. Cause-effect relationship is still unassigned. We now report evidence of a limiting structural timescale for the formation of the metallic phase. Such evidence provides strong evidence in favor of the structurally driven mechanism.



**Figure 2:** Time-resolved reflectivity/transmission evolution of VO<sub>2</sub> / Si<sub>3</sub>N<sub>4</sub> structures after 100-fs excitation pulses at 790-nm wavelength and 50 mJ/cm<sup>2</sup>.

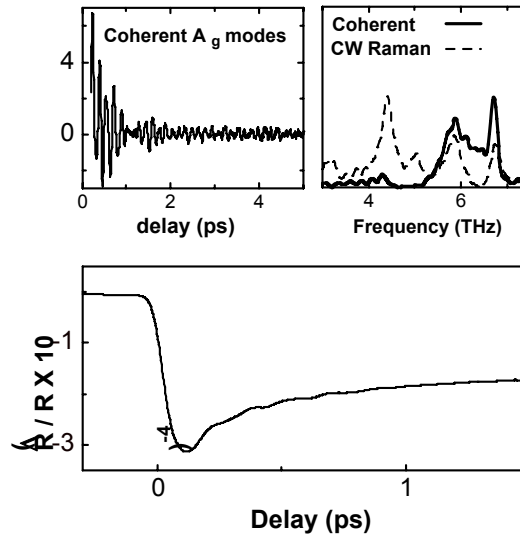
Time-resolved optical spectroscopy was performed at several wavelengths in the visible using a 1-KHz amplified Ti-Sapphire laser system, white-light continuum generation and an optical parametric amplifier. An insulator-to-metal transition is evidenced by increase of the reflectivity and decrease in the transmission (figure 1).

The fundamental transition time was then measured as a function of pulse duration in the range between 1.5 ps and 15 fs. A bottleneck timescale of 75 fs appeared, even in the cases where excitation was complete on the 10-fs timescale (figure 2).



**Figure 2:** Pump-probe reflectivity experiments of the photo-induced phase transition. The experiments are performed in the non-etched structure with pulse durations between 1.5 ps and 15 fs.

The nature of the bottleneck timescale was further investigated using Raman and coherent phonon spectroscopy, used to address the nature of the elementary structural distortions that give rise to the bottleneck of figure 2. These experiments revealed that photo-excitation results in coherent structural motion, where only modes of Ag symmetry are excited. Remarkably, the bottleneck timescale for the phase transition observed at higher fluence corresponds approximately to half period of the two coherent modes. Analysis of the real-space motion of atoms reveals that the coherently excited modes are the same modes that project the atomic arrangements of the two crystallographic phases onto one another. These modes have been identified as the order parameter for the transition and project the space group of the low-T phase onto that of the high-T phase. Importantly, the change in symmetry of the unit cell renormalizes the A1g, zone-center and raman active phonons of the low-T phase to the zone edge in the high-T phase. Any vibrational coherence in the product phase would thus be invisible to optical probing.



**Figure 3:** Time-resolved evolution of the reflectivity in the low-T phase. The coherently excited modes are symmetric  $A_g$  modes with displacements that project onto the high-T crystallographic phase, as revealed by comparison with the CW Raman spectrum.

In summary, we have shown that ultrafast spectroscopy on the sub-vibrational timescale can be applied to resolve ambiguous cause-effect assignments across phase transitions in strongly correlated electron systems. Based on the ultrafast response of to photo-excitation, we conclude that that the atomic arrangement of the high-T unit cell is necessary for the formation of the metallic phase of  $\text{VO}_2$ , even if the correlated d band is highly depleted (hole-doped). This result is suggestive of significant Band-like character for the low-T insulator. A highly controversial issue on the nature of the insulating phase in this prototypical, correlated system is settled by means of time-resolved spectroscopy.

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