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Tracking the Phase Transition in VO₂ using High Harmonic Spectroscopy

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We present a study that uses a technique extending upon high harmonic spectroscopy, which is generated as a function of photoexcitation pump fluence to probe the resulting ultrafast dynamics of the insulator-to-metal phase transition in VO₂ [1]. When heated to above ~343 K, VO₂ experience a complete atomic rearrangement of the crystal lattice structure from a monoclinic insulator (*M1* phase) to a metallic rutile crystalline structure (*R* phase). When initiating this IMT via photoexcitation, the pathways are even more complex. After photoexcitation, the ultrashort pulse immediately excited electrons creating the *M1* state. With sufficient pump energy, the periodic lattice of the *M1* phase can transition completely into the *R* phase. If there is insufficient energy, after a few hundred femtoseconds, the excited photodoped electrons in the *M1* state relax into a pseudothermal state in which the thermalized photodoped populations have the same chemical potential (*M1,b* state). After about a picosecond, the *M1,b* state then transitions into a long-lived metastable monoclinic metallic *M* state. If the pumping fluence is between these two thresholds, a final metallic mixed state of rutile and monoclinic is produced (*R + M*). So far, only Morrison et al. have reported the existence of the monoclinic metallic *M* state [2]. Here, we show that the IMT dynamics in VO₂ can be tracked by measuring the yield of the 5th intraband harmonic. The temporal evolution of the harmonic yield reveals both time scales, i.e. ~300fs to reach the pseudothermal state and ~1.5ps for the monoclinic metallic state.

[1] M.R. Bionta et al., "Tracking ultrafast solid-state dynamics using high harmonic spectroscopy," under review (2018).

[2] V.R. Morrison et al., "A photoinduced metal-like phase of monoclinic VO₂ revealed by ultrafast electron diffraction," *Science* 346, 455 (2014).

[3] M.R. Bionta et al., "Probing the phase transition in VO₂ using few-cycle 1.8 μm pulses," *Phys. Rev. B* 97, 125126 (2018).

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