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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 mononiclinic 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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