CFEL SCIENCE

SEMINAR

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CFEL – Building 99, seminar room I+II (ground floor)

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Probing the insulator to metal phase transition in VO_2 using high harmonic spectroscopy

We extend the high harmonic spectroscopy (HHS) technique in solids with a systematic study of the generated spectra as a function of photoexcitation pump fluence to time-resolve the electronic dynamics associated to the insulator-to-metal (IMT) phase transition in VO_2 [1]. When heated above ~343 K, VO_2 experiences a complete atomic rearrangement of the crystal lattice structure from a monoclinic insulator (M_1 phase) to a metallic rutile crystalline structure (R phase). When initiating this IMT via photoexcitation, the pathways are even more complex. During photoexcitation, the ultrashort pulse instantaneously excites electrons creating the M_1^* state. With sufficient pump energy, the periodic lattice of the M_1 phase can completely transit into the R phase. With insufficient pump energy, the excited photodoped electrons in the M_1^* state relax after a few hundred femtoseconds into a pseudothermal state in which the thermalized photodoped populations have the same chemical potential ($M_1^{*,b}$ state). After about a picosecond, the $M_1^{*,b}$ state reaches a long-lived metastable monoclinic metallic $\mathcal{M}$ state. When the pumping fluence is between these two thresholds, a final mixed state between rutile and monoclinic is produced (R + $\mathcal{M}$). So far, only Morrison et *al.* have reported the existence of the monoclinic metallic $\mathcal{M}$ state using ultrafast electron diffraction (UED) measurements [2]. The question we asked: Is high harmonic spectroscopy sensitive to the evolution of the IMT?

We track the IMT dynamics in VO_2 by measuring the yield of an intraband harmonic, requiring a conduction band with an anharmonic band structure. A mid-infrared laser pulse (drive) at 10 μm is used to drive high harmonic generation (HHG) from a 100 nm thick, epitaxial VO_2 sample [3]. The sample is photoexcited with a 50 fs, 1.5 μm laser pulse (pump) to initiate the IMT. The fifth harmonic signal is recorded as a function of the time delay between the pump and the high harmonic driver. Within the range of pump fluence where Morrison et *al.* have reported the observation of the monoclinic metallic $\mathcal{M}$ state, we measured a drop of the harmonic signal at zero delay when electrons are promoted to the conduction band, followed by a recovery of the harmonic yield within a picosecond as measured using UED [2]. This demonstrates the potential of high harmonic spectroscopy as a complementary technique for tracking ultrafast dynamics in solids. Furthermore, recent results will be presented showing additional information extracted using high harmonic spectroscopy.

[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).

Host: Francesca Calegari / CFEL Molecular and Ultrafast Science Seminar