Extreme Ultraviolet Transient Grating Measurement of Insulator-Metal Transition Dynamics of VO_2

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Abstract: We demonstrate spectrally resolved transient grating spectroscopy in the extreme ultraviolet near the M-edge of vanadium dioxide. Time-dependent measurements of the ultrafast photo-induced insulator-to-metal transition disentangle pure electronic response from electron-phonon coupling and thermal effects.
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1. Introduction

Multidimensional spectroscopy (MDS) has proven a powerful technique in chemistry and materials science, particularly in the infrared, visible and near-ultraviolet [1]. A form of MDS, transient grating (TG) spectroscopy has been highly successful in examining the electronic response of complex materials, electron-phonon coupling, and acoustic waves on surfaces [2]. In addition to being intrinsically background-free, TG experiments have proven highly sensitive to both surface and bulk acoustic waves and are widely used to characterize thin films. Few MDS experiments have been performed in the extreme ultraviolet (EUV) to study the acoustic response of a sample [3]. We further develop the technique to study the electronic sample response close to characteristic resonances in the EUV region. We demonstrate this for the example of vanadium dioxide (VO_2), which is a strongly-correlated material of particular interest because of a photo-induced, ultrafast insulator-to-metal transition (IMT) (see for instance [4]). The extensive literature documents a debate over the mechanism of the IMT in VO_2, which can also be induced thermally, by heating the sample above about 70 °C. The phase transition is thought to involve both structural and electronic dynamics [5-6]. The use of EUV TG spectroscopy for studying the photo-induced transition allows for probing of the electronic structure with sensitivity to the vanadium M-edge during the IMT.

2. Experiment

High harmonics of an 800nm, 25 fs, Ti:Sapphire laser are generated with a loose (~1.5m) focusing geometry in an argon cell. The transient grating pump consists of two synchronized pulses of 800nm light, crossed on the sample and impinging close to the surface normal. The grating period of ~15 μm is achieved with a total fluence of approximately 15 mJ/cm^2. The high harmonic probe beam is refocused with a 2m focal length toroidal mirror, and probes the VO_2 sample surface at an incident angle of 22 degrees before reaching the focus at the plane of the detector. Harmonics of order 15-29 (22.5-43.5 eV) are resolved in the first order diffracted signal and integrated using a back-illuminated CCD camera. The experimental geometry for the TG is shown in Figure 1. The average harmonic intensity is monitored through the current drawn by photoemission from the toroidal mirror and recorded to check source stability during long acquisitions [7].

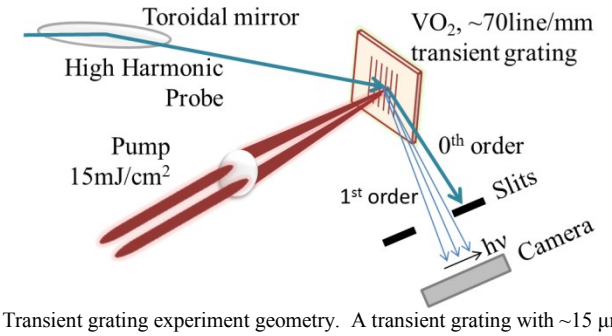

Figure 1: Transient grating experiment geometry. A transient grating with ~15 μm period created on the VO_2 surface by two 800nm beams. High harmonics generated in a high pressure gas cell are refocused using a toroidal mirror. First order diffraction from the sample surface is separated from the 0-order beam with slits and detected with a CCD camera.

3. Results

The time dependence of the transient response of VO_2 was studied with the sample below and above the thermally induced IMT (typically between 60-70 C depending on growth condition). In the (heated) metallic phase, the pump is expected to drive acoustic phonon modes in the sample, but no IMT. The measured thermal grating has a rise-time of approx. 20 ps. The response of harmonic order 25 (HH25) is included in Figure 2 (green x). All other recorded harmonic orders yield similar behavior.

The colder room temperature sample exhibits two different temporal behaviors as a function of probe wavelength. Harmonic orders below HH25 (38.8 eV) display only the slow acoustic signal known from the hot measurements. Harmonics 25 and above also exhibit a much faster transient feature before the slow acoustic signal. Figure 2 shows the time-dependence of HH21 (red ○), which exhibits only the slow acoustic response, and HH25 (blue □), which increases rapidly for a few picoseconds, before continuing to rise with the acoustic behavior.

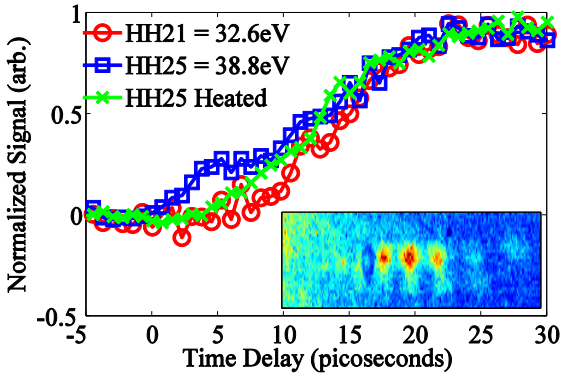

Figure 2: Integrated transient grating signal. For sample temperatures above the IMT transition the 1st order diffracted harmonic signal increases according to the acoustic response of the material (green x). With a room temperature sample (below the thermal IMT) low order harmonics show a slow acoustic response. Harmonics close to the V M-edge resonance (order 25 (blue □) and above) show a faster electronic response not seen in the lower-order harmonics (red ○). The inset shows a typical image of different diffracted harmonics on the detector.

The fast rise-time exhibited by the highest harmonic orders is likely to be the electronic response of the VO_2 as it undergoes the IMT. The measured few picosecond rise-time is our current resolution limit due to the grazing incidence of the beamline. The highest-order harmonics have an increased electronic sensitivity since they are at the onset of the vanadium M-edge [8].

We measure a maximal diffraction efficiency of order 10^{-2}, consistent with a heat induced sample expansion. The large efficiency increase over 10^{-8} efficiency reported for IR and visible TG measurements [2] demonstrates the advantage of EUV TG spectroscopy. Furthermore, we note that accumulated damage to the surface of the sample creates a permanent grating, producing a diffracted signal under the transient signal. This opens the interesting opportunity for future heterodyne measurements allowing for signal amplification and decomposition in real and imaginary material constants.

By generating the pump's grating pattern with diffractive optics and utilizing a tilted wavefront scheme, we plan to compensate for the time-resolution limitations caused by the grazing incidence probing.

4. References

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