Optical pump – THz probe response of VO_2 under high pressure

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Abstract. We present the ultrafast THz response of VO_2 under high pressures. A clear anomaly is observed around 8 GPa indicating a pressure-induced phase transition. Our observations can be interpreted in terms of a bandwidth-controlled Mott-Hubbard transition.

1 Introduction

Vanadium dioxide (VO_2) is a prime example of a strongly correlated system undergoing insulator-to-metal transition (IMT) at a critical temperature T_c = 340 K. The sharp drop in resistivity occurs simultaneously with a structural transformation from a dimerized monoclinic to a tetragonal rutile structure.

Ultrafast pump-probe studies have demonstrated that VO_2 can be switched non-thermally into the metallic state on sub-100 fs timescale [1,2]. In particular, probing electronic and lattice degrees of freedom using ultrashort THz pulses has provided an important insight into the mechanism of the photo-induced IMT in VO_2 [2]. The cooperative character of the ultrafast IMT is evidenced by the fact that only photoexcitation above a certain threshold fluence creates a long-lived metallic phase [2,3].

Application of external pressure provides another non-thermal route for the IMT in VO_2. However, the crystal structure in the high-pressure metallic state remains dimerized in stark contrast to the temperature-driven IMT [4,5]. Thus, the mechanism behind the pressure-induced IMT must be qualitatively different and its understanding is vital for identifying the roles played by electronic and lattice degrees of freedom in the properties of VO_2.

In this study we use time-resolved optical pump – THz probe spectroscopy in order to understand the phenomena occurring in VO_2 under high pressure.

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2 Experiment

The pressure experiment was performed using a diamond anvil cell. CsI powder was used as a pressure-transmitting medium in order to ensure good contact between the studied VO_2 crystal sample and the surface of a diamond anvil. Photoexcitation by near-infrared pulses with photon energy of 1.55 eV excited carriers above the band gap of VO_2. The response of photoexcited charge carriers was probed by a delayed probe pulses with a central frequency of 30 THz reflected from the surface of the sample. The spot sizes (FWHM) of the pump and probe beams were around 55 and 35 μm, respectively.

3 Results and discussion

The pump-probe measurements at different excitation fluences have been performed for pressure between 0 and 20 GPa. Fig. 1(a) shows time-resolved reflectivity changes at the pressure of 2.9 GPa for pump fluences up to 20.6 mJ/cm^2. The initial steep reflectivity increase with the duration limited by the temporal resolution of our experiment is followed by a characteristic relaxation on sub-ps timescale in agreement with previous studies [2,3]. When the excitation fluence exceeds a certain threshold, the reflectivity change $\Delta R/R$ remains finite on a multi-ps timescale indicating the onset of a metastable metallic phase. The threshold fluence Φ_{th} can be determined by plotting the fluence dependence of the pump-probe signal level after the initial relaxation process as depicted in Fig. 1(b). We define Φ_{th} as the crossing point of asymptotes to linear parts of the fluence dependence. Fig. 1(c) shows the pressure dependence of Φ_{th} determined for two VO_2 crystals together with the linear transmissivity at the THz frequency.

Similar to previous high-pressure studies we assign the vanishing THz transmissivity to the filling of the band gap caused by the pressure-driven phase transition in VO_2 [4]. The critical pressure p_c = 6–8 GPa observed in our experiments is somewhat lower compared to the previous report [4] that might be related to pressure non-hydrostaticity effects. Interestingly, the threshold fluence experiences an anomalous drop exactly at the transition pressure p_c. Moreover, in contrast to the thermally driven IMT, Φ_{th} increases for $p < p_c$ and does not vanish for $p > p_c$ showing only a gradual decrease as depicted in Fig. 1(c). Thus, the threshold behavior is observed in the whole studied pressure range of our experiment in spite of the appearance of intragap states evidenced by the strong drop in the linear THz transmissivity.

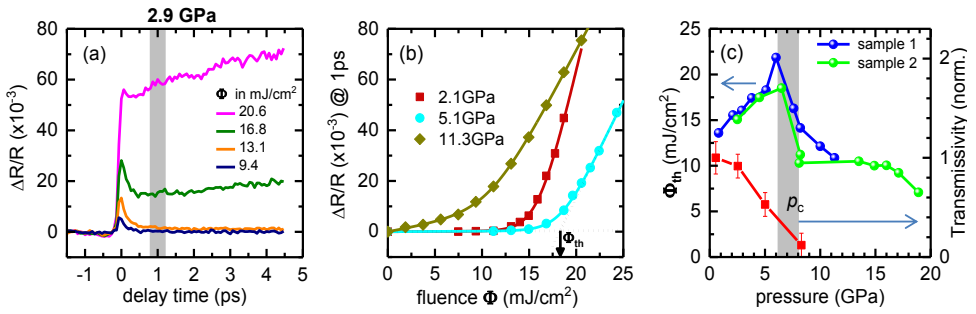

Fig. 1. (a) Normalized pump-probe signals measured at a pressure of 2.9 GPa for different excitation fluences Φ; (b) Fluence dependence of pump-probe signals averaged around the delay time of 1 ps; (c) Pressure dependence of the threshold fluence Φ_{th} (blue and green) and the normalized THz transmissivity (red).

We suggest that the signatures of the electronic localization and the band gap filling observed simultaneously for $p > p_c$ can be understood within the scenario of the bandwidth-driven Mott-Hubbard transition [6]. In this picture the band structure of the metallic phase near the transition retains the lower and upper Hubbard bands split by the onsite Coulomb interaction and the metallic conductivity originates from a limited amount of states in a quasiparticle peak which appears at the Fermi level. Thus, it is still possible to increase the number of free charge carriers beyond the critical limit by optical pumping and to drive the complete closure of the band gap. This can explain why the pump-probe response of VO_2 remains partially similar to its insulating phase even for $p > p_c$. On the other hand, the linear transmissivity/reflectivity is mostly affected by the free or weakly localized charge carriers in the quasiparticle peak and shows a drop/increase above the transition.

The observed initial increase of the threshold fluence can be interpreted as a stabilization of the monoclinic structure under high pressure. As a result, the photo-induced switching requires higher excitation fluence in order to overcome the energy barrier to the non-dimerized rutile structure. The fact that the observed pressure-induced transition in VO_2 is not related to a Peierls instability [4,5] provides a further support for the suggested picture of a purely electronic Mott-Hubbard transition driven by the pressure-induced broadening of the electronic bands.

4 Conclusion

Our results provide important hints for understanding the nature of the pressure-induced correlated state of VO_2. The complete picture of the linear and nonlinear THz response of pressurized VO_2 can be explained using the scenario of a purely electronic bandwidth-driven Mott-Hubbard transition completely decoupled from a Peierls instability.

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