Ultrafast Insulator-Metal Transition in VO_2 Driven by Intense Multi-THz Pulses

A. Grupp,[1] B. Mayer,[1] C. Schmidt,[1] J. Oelmann,[1] R. E. Marvel,[2] R. F. Haglund Jr.,[2] A. Leitenstorfer,[1] and A. Pashkin[1]

[1]*Department of Physics and Center for Applied Photonics, University of Konstanz, Universitätsstraße 10, 78464 Konstanz, Germany*
[2]*Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee 37235, USA*
Alexander.Grupp@uni-konstanz.de

Abstract: We demonstrate a non-thermal metallization of VO_2 induced by a non-resonant excitation at frequencies around 25 THz. An ultrafast switching time of 80 fs comprises only two cycles of the driving multi-THz field.

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Vanadium dioxide (VO_2) is a prime example of a transition metal oxide exhibiting a sharp insulator-metal transition at 340 K (67 °C). This high critical temperature renders several potential applications in optics and high-speed electronics. The delicate balance of competing interactions results in a high sensitivity to external perturbations and the metallic state can be induced by temperature, pressure, photoexcitation or electric fields.

Renewed interest towards VO_2 was triggered by the discovery of a photoinduced ultrafast insulator-metal transition [1]. By means of pump-probe spectroscopy switching times of less than 100 fs and the essential role of lattice dynamics in the photoinduced transition could be demonstrated [2-4]. These experimental observations have been well described by a quantitative model [5]. Nevertheless, control of the electronic state of VO_2 by applied electric fields is widely believed to be governed by resistive heating, dramatically limiting the switching speed [6].

Recent progress in the generation of intense terahertz (THz) pulses offers an attractive possibility to transiently apply extremely high electric fields without irreversible destruction of the structure. Thus, an excitation by THz transients offers an efficient way for the ultrafast non-thermal metallization of VO_2 by electric fields. A first demonstration of the field-induced switching in a VO_2 metamaterial using intense THz pulses has been reported lately [7]. However, the observed switching times of 8 ps are relatively long, indicating a thermal character of the transition driven by low-frequency THz transients.

Here, we demonstrate for the first time the insulator-metal transition in unstructured polycrystalline VO_2 films induced by high-field multi-THz waveforms on a sub-100 fs timescale. The thicknesses of films employed for the measurement are 120 nm and 200 nm, respectively. They have been grown by pulsed laser deposition on a CVD diamond substrate and a temperature-dependent measurement of the optical transmission verified the transition temperature to be 340 K. Difference frequency mixing was employed to generate broadband multi-THz transients with extremely high peak electric fields of up to 17 MV/cm [8]. Such a waveform and its corresponding amplitude spectrum centered at 27 THz is shown in Fig. 1(a). The electronic state of the samples used in our experiments is monitored by probing a transmission change utilizing 8-fs-short near-infrared pulses with a central wavelength of 1.2 µm.

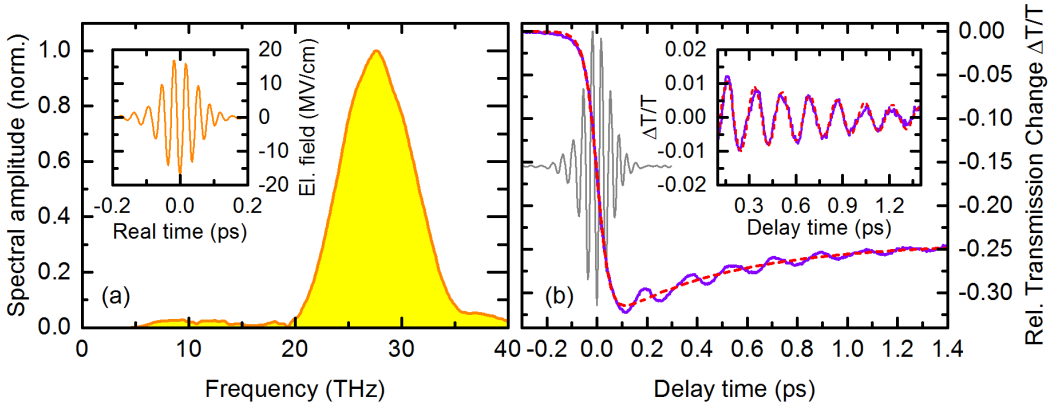

Fig. 1. (a) Amplitude spectrum and electric field profile (inset) of the multi-THz pump pulse; (b) Relative transmission change ΔT/T of the 200-nm-thick VO_2 film (solid purple line) induced by the multi-THz transient (gray line) with incident fluence of 40 mJ/cm^2 measured at room temperature. Red dashed lines show the fitting curves. Inset: Remaining coherent oscillation fitted by a damped harmonic oscillation with a frequency of 5.7 THz.

Ultrafast switching dynamics measured by a multi-THz pump / near-infrared probe experiment at room temperature are shown in Fig. 1(b). The relative transmission change ΔT/T is plotted as a function of the delay time between pump and probe pulses (purple line) and is compared to the driving multi-THz waveform (gray line). Ultrafast switching into the metallic state is succeeded by a relatively slow relaxation. A full recovery of the insulating state under a strong excitation requires timescales longer than 100 ps (not shown). A fit to the measured data [dashed line in Fig. 1(b)] reveals a remarkable short switching time of 80 fs, comprising only two cycles of the driving THz field. Interestingly, the metallization of the VO_2 film occurs within a time interval even shorter than the duration of the driving pulse. Clearly, such an ultrafast phase transition also requires a modification of the lattice structure which is evidenced by the coherent oscillation at a frequency of 5.7 THz observed in our experiment [see Fig. 1(b)]. The same type of oscillation assigned to coherent wave packet motion of the vanadium dimers has been reported in the metallic state of VO_2 induced by ultrashort near-infrared pulses [3,4].

In an additional experiment, the THz fluence dependence of the insulator-metal transition in a 120-nm-thick VO_2 film is studied. Fig. 2(a) demonstrates the time-resolved relative transmission change of the probe pulse for various incident excitation fluences. Remarkably, dynamics of the switching process is almost independent of the excitation fluence. However, the amplitude of the pump-probe signal shows a strongly nonlinear dependence on the THz fluence. For fluences below 10 mJ/cm^2 it is almost not observable and only starts to grow gradually at higher excitation fluences. This behavior is emphasized in Fig. 2(b) where the maximal transmission change is plotted as a function of the multi-THz fluence. A guide to the eye (red line) denotes the presence of a threshold fluence required for a creation of the meta-stable metallic phase. The value of the threshold fluence is of the same order as the values reported in the previous studies with near-infrared pump pulses [3,4].

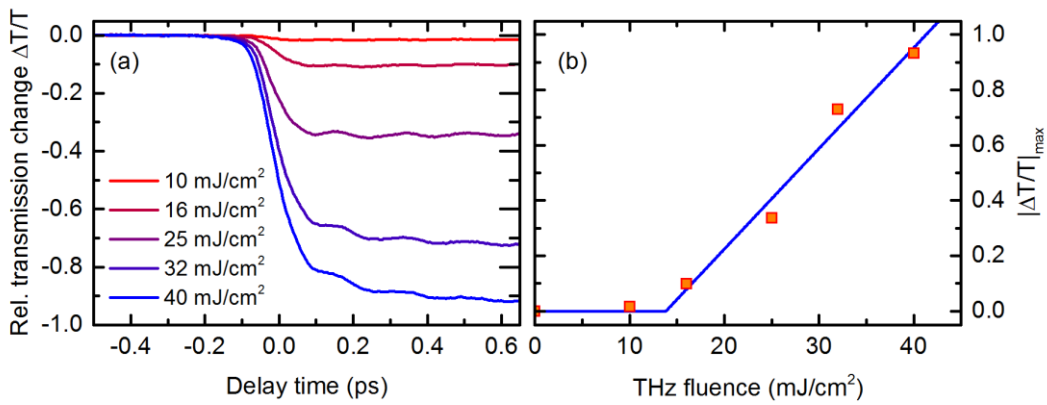

Fig. 2 (a) Relative transmission change ΔT/T measured at various excitation fluences; (b) Maximal |ΔT/T| as a function of the incident THz fluence. The blue line serves as a guide to the eye demonstrating the threshold behavior.

In conclusion, we observed sub-100 fs switching timescales and coherent lattice dynamics in VO_2, strongly supporting a non-thermal scenario of the field-induced insulator-metal transition. The physical mechanism of this fascinating phenomenon still has to be understood in detail requiring further experimental and theoretical efforts.

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