

## All-optical short pulse translation through cross-phase modulation in a VO<sub>2</sub> thin film

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Received 13 August 2015; revised 27 November 2015; accepted 30 November 2015; posted 3 December 2015 (Doc. ID 247826); published 6 January 2016

**VO<sub>2</sub> is a promising material for reconfigurable photonic devices due to the ultrafast changes in electronic and optical properties associated with its dielectric-to-metal phase transition. Based on a fiber-optic, pump-probe setup at 1550 nm wavelength window, and by varying the pump-pulse duration, we show that the material phase transition is primarily caused by the pump-pulse energy. For the first time, we demonstrate that the instantaneous optical phase modulation of probe during pump leading edge can be utilized to create short optical pulses at probe wavelength, through optical frequency discrimination. This circumvents the impact of long recovery time well known for the phase transition of VO<sub>2</sub>.** © 2016 Optical Society of America

**OCIS codes:** (060.5060) Phase modulation; (320.7130) Ultrafast processes in condensed matter, including semiconductors; (160.4670) Optical materials; (310.6860) Thin films, optical properties.

<http://dx.doi.org/10.1364/OL.41.000238>

The search for advanced photonic materials with high modulation efficiency and fast response time has been a continuous challenge in the field of nanophotonics. Realization of functional photonic devices, such as optical modulators and switches that provide rapid and energy-efficient optical response at compact size, relies heavily on the fundamental properties of materials [1–3]. Vanadium dioxide (VO<sub>2</sub>) is a promising candidate for a variety of photonic and optoelectronic applications that encompass large, broad-spectrum and ultrafast optical responses while suitable for integration with silicon photonics when needed [4]. A distinctive property of VO<sub>2</sub> is the capability of reversible dielectric-metal phase transition (DMT), which occurs near a critical temperature  $T_c$  in the vicinity of 67°C. This transition is associated with drastic changes in the refractive index and resistivity of the material [5,6]. DMT can be induced thermally [7], optically [8], and electrically [9] on an ultrashort time scale of less than 100 fs [10]. Although the material properties of VO<sub>2</sub> have been studied extensively, argu-

ment on the exact mechanism behind phase transition still exists. Various applications utilizing its optically induced refractive index modulation ( $\Delta n$ ) have been reported, such as tunable resonators [4], filters and modulators [11,12], optical phase controllers [13], switches, and photodetectors [14]. However, a major drawback of VO<sub>2</sub> is the relatively long recovery time of the transition back from the metal state to the dielectric state, which is usually longer than 10 ns. This largely asymmetric response limits many potential high-speed applications.

In this work, we present a systematic measurement, using a fiber-optic, pump-probe setup in a 1550 nm optical communications wavelength window. We show that the optically induced material phase transition in VO<sub>2</sub> is primarily related to the pulse energy. Although the refractive index change of VO<sub>2</sub> triggered by the pump pulse has a long recovery time, the induced optical frequency change on the probe, which is proportional to the time derivative of the index change ( $dn/dt$ ), has a time scale only determined by the leading edge of the pump pulse, which can be shorter than 100 fs. This allows for ultrafast, optically induced modulations. In particular, we demonstrate the feasibility of converting a continuous wave (CW) probe source into a pulsed waveform due to the effect of short pump pulses on the probe through cross-phase modulation in VO<sub>2</sub> followed by a frequency discriminator. In addition to clarifying the fundamental mechanism behind optically induced phase transition in VO<sub>2</sub>, the frequency discrimination technique demonstrated in this Letter opens the possibility of converting ultrashort optical pulses from the pump wavelength to the probe wavelength. Because DMT in VO<sub>2</sub> is a broadband phenomenon covering both visible and near infrared (NIR) wavelengths [5], the proposed wavelength translation can be performed anywhere across this region. The duration of the translated pulses is limited only by half the leading edge of the DMT, which is shorter than 100 fs [10].

VO<sub>2</sub> thin films (200 nm) were deposited on *c* axis, single crystal sapphire substrates by pulsed laser deposition (PLD) [15–17]. Details of the deposition conditions of VO<sub>2</sub> thin film have been described elsewhere [5,17]. Briefly, a KrF

excimer laser (Lambda Physik LPX 305,  $\lambda = 248$  nm, pulse duration = 30 ns) was operated at a pulse rate of 10 Hz and was focused onto a rotating VO<sub>2</sub> target with a laser beam energy density of 1.5 J/cm<sup>2</sup> and a target-to-substrate distance of 5.8 cm. The target was a 2" diameter by 0.25" thick sintered oxide ceramic disk supplied by ACI Alloys, USA. The deposition chamber was initially evacuated to  $<10^{-5}$  Torr and then oxygen gas was introduced into the chamber to maintain the desired pressure of 20 mTorr. The substrate temperature was kept at 550°C during deposition. The as-deposited samples were in situ annealed for 1 h, maintaining the deposition temperature and oxygen pressure to obtain pure VO<sub>2</sub> phase. The samples were cooled down to room temperature at the same oxygen pressure. In order to allow optical transmissivity measurements, both sides polished sapphire substrates were used in this work. The deposited films exhibited a significant resistivity drop by four orders of magnitude and ~60% optical transmittance drop at near IR (2500 nm) as the temperature increased up to 100°C above the transition temperature [17].

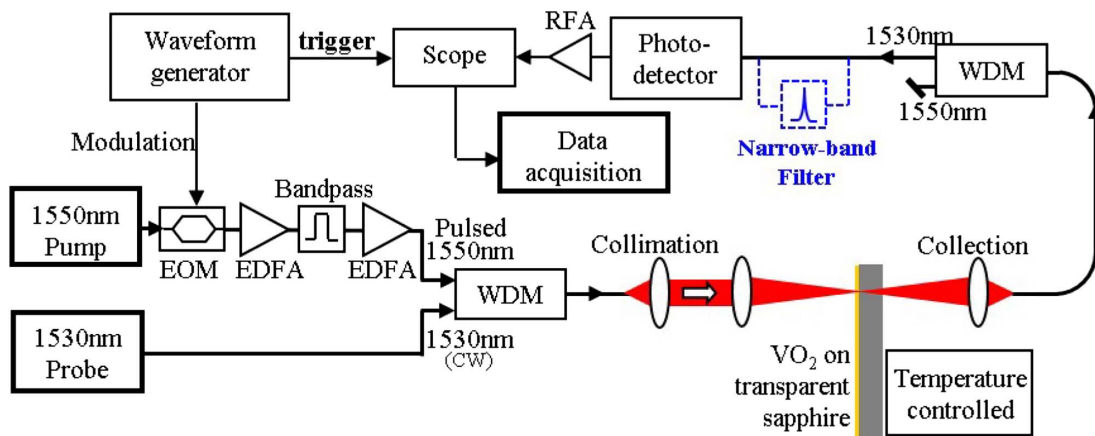
Figure 1 illustrates the fiber-optic experimental setup used to measure the transmission of the VO<sub>2</sub> sample in the 1550 nm optical communications wavelength window. Two tunable semiconductor lasers were used as the pump and the probe sources, and both of them were operating in the CW. The pump laser emitting at 1550 nm was intensity-modulated through a fiber pig-tailed LiNbO<sub>3</sub> electro-optic modulator to create width-tunable optical pulses ranging from 200 ps to 10 ns. The wavelength of the probe laser was 1530 nm. The electrical waveform applied to the electro-optic modulator was generated by an arbitrary waveform generator (AWG) with a sampling rate of 25 GS/s, and the analog bandwidth of the modulator is 10 GHz. After intensity modulation, the pump-optical pulse train was amplified by two erbium-doped fiber amplifiers (EDFA), with a bandpass optical filter (1 nm bandwidth) sitting in between them to minimize the saturation effect due to amplified spontaneous emission (ASE) on the second EDFA. The pump and probe were combined through a fiber-optic wavelength division multiplexing (WDM) coupler and focused onto the preheated VO<sub>2</sub> sample through free space. Transmitted light through the sample was collected using a 10× objective and coupled into the single-mode fiber pigtail of a second WDM coupler, which operated as a demultiplexer to reject the pump wavelength, allowing only the probe wavelength

to reach the photodetector with a 12 GHz electrical bandwidth. The detected electrical waveform of the probe was amplified and recorded by a real-time digital scope with a 50 GS/s sampling rate, which was triggered by the AWG. The VO<sub>2</sub> sample temperature was set at approximately 58°C by a temperature controller, which is in the vicinity of the phase transition temperature of VO<sub>2</sub> [5] and was chosen to maximize the impact of pump pulse on the transmission reduction of the probe. The measurement was performed with 500 kHz pump-pulse repetition rate, with the width of the pump pulse being varied from 200 ps to 10 ns. The peak optical power of the pump was fixed at approximately 5 W, which was primarily limited by the EDFA saturation and stimulated Brillouin scattering (SBS) of the single-mode fiber.

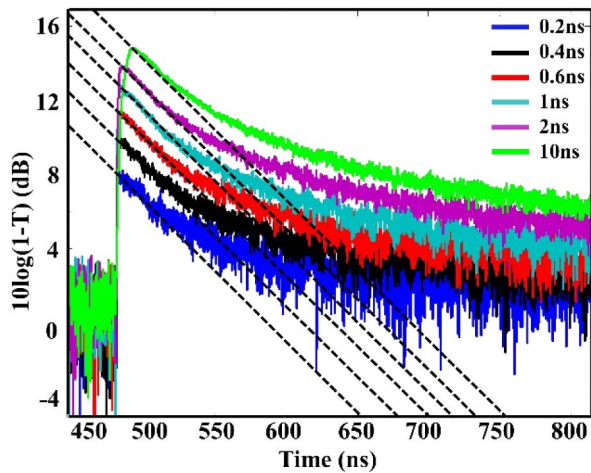
As the VO<sub>2</sub> undergoes the phase transition from dielectric to metallic state, a dramatic change in its optical and electrical properties is anticipated, leading to an increase in the optical loss and a reduction of probe transmission. Figure 2 shows the measured probe absorption waveforms ( $1 - T$ ) at different widths of the pump pulse plotted in a logarithm scale (in dB), where  $T$  is the probe power transmissivity.

These waveforms show that  $T$  is reduced instantaneously at the leading edge of the pump pulse due to an abrupt increase of the sample temperature. Previous studies have reported response times of less than 100 fs for VO<sub>2</sub> phase transition upon fs pump excitation [10]. On the other hand, the recovery time is much longer due to the slow heat dissipation after the pump pulse with the temperature reduction following a typical exponential decay process. Considering a constant pump peak power was used in this experiment, the pulse energy increases linearly with the pulse width, and the reduction in the probe transmission also increases accordingly. Figure 2 also shows that the time constant of temperature decay gradually increases after the pump pulse because the reduction of temperature gradient near the focal point over time, which slows down the heat dissipation. This is shown as the slope reduction of the decay curves further away from the pump excitation event. Nevertheless, the decay rate of probe absorption immediately after the pump pulse is almost constant for all pump pulse widths from 0.2 ns to 10 ns shown as dashed straight lines, which is 37.5 ns.

At a more fundamental level, it is important to understand the basic mechanism behind the optically induced VO<sub>2</sub> phase transition and clarify whether this transition is caused by the

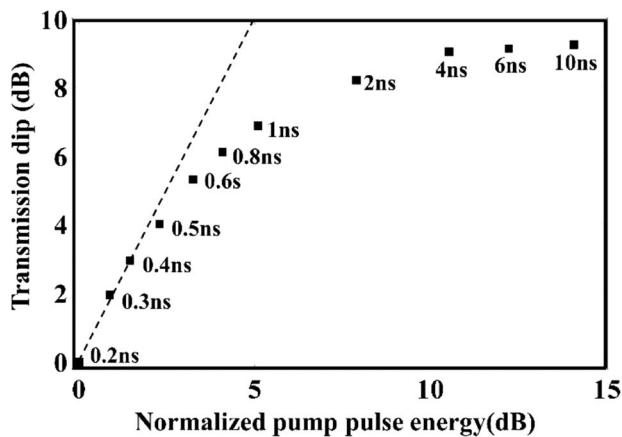


**Fig. 1.** Experimental setup. WDM, wavelength division multiplexer/demultiplexer between 1530 and 1550 nm; EOM, electro-optic modulator; EDFA, erbium-doped optical amplifier; RFA, radio frequency amplifier.



**Fig. 2.** Measured waveforms of probe transmission  $T$  upon optical excitation of a 200 nm  $\text{VO}_2$  film via pump pulses of different time durations. Here  $(1 - T)$  are plotted in logarithm scale to better demonstrate recovery time constants: The slopes of the dashed lines indicate a recovery time of approximately 37.5 ns for all six curves near their pulse peak.

peak power or by the energy of the optical pump pulse. To answer this question, a more systematic experiment was performed which measured the depth of probe absorption at pump pulses with different durations. In this case, the pump peak power was kept constant so that the width change is equivalent to varying the pulse energy. The results are shown in Fig. 3, where solid squares are the normalized depth of probe absorption measured with pump pulses of different widths on a log-log plot. This figure indicates that when the pump pulse duration is much shorter than the phase recovery time of the  $\text{VO}_2$  film, the depth of probe absorption is linearly proportional to the energy of the pump pulse. When the pump pulse is sufficiently long and no longer negligible in comparison with the phase recovery time constant ( $\sim 37.5$  ns in this case), the efficiency of probe absorption starts to reduce. This is caused by the non-negligible thermal dissipation within the duration of



**Fig. 3.** Normalized depth of probe absorption as a function of pump-pulse energy on a log-log scale. The value marked near each solid square indicates the width of the pump pulse. The dashed straight line is a linear fit at shorter pulse durations.

the long pump pulses. Consequently, the sharpness of the phase change (from dielectric to metal) decreases accordingly. Therefore, the probe absorption starts deviating from its initial rate and tends to saturate with longer pump pulse durations. Note that in Fig. 3 the linear fit at short pump-pulse widths reveals an approximately 2 dB increase of probe absorption for a dB increase of the pump-pulse energy. This super-linear characteristic is possible because the sample was preheated to the middle of its sharp phase transition region.

The optically induced phase transition in  $\text{VO}_2$  is associated with the change of the complex refractive index. It has been shown that the refractive index of  $\text{VO}_2$  can be switched from  $n_s = 3.243 + j0.346$  for the dielectric state to  $n_m = 1.977 + j2.53$  for the metallic state [18]. The measurements of pump-induced probe absorption increase discussed so far in the literature were only related to the change of the imaginary part of the refractive index, while the optical phase modulation on the probe relies on the pump-induced change related to the real part of the  $\text{VO}_2$  refractive index. Considering the  $\text{VO}_2$  film used in the experiment with a thickness  $d = 200$  nm, for a complete material phase transition from dielectric to the metallic state, the maximum optical phase change ( $\delta\varphi$ ) on the probe wave at  $\lambda = 1530$  nm wavelength would be approximately:

$$\delta\varphi = \frac{2\pi d}{\lambda} \text{Re}(n_m - n_s) \approx 1 \text{ rad.}$$

The time-domain response of this probe optical phase modulation  $\delta\varphi(t)$  is expected to have the same waveform as the probe absorption when the material phase transition is optically induced by a pump pulse. The largely asymmetric response (shown in Fig. 2) with an ultrafast leading edge and a significantly slower falling edge prohibits the generation of short optical pulses of the probe when passing through the  $\text{VO}_2$  during its phase transition. However, the dynamic frequency modulation induced on the probe beam is proportional to the derivative of the optical phase modulation ( $d\varphi(t)/dt$ ). Thus a large optical frequency modulation may be obtained, corresponding to the fast leading edge of the optical phase modulation, while the much slower falling edge would no longer contribute to this optical frequency modulation.

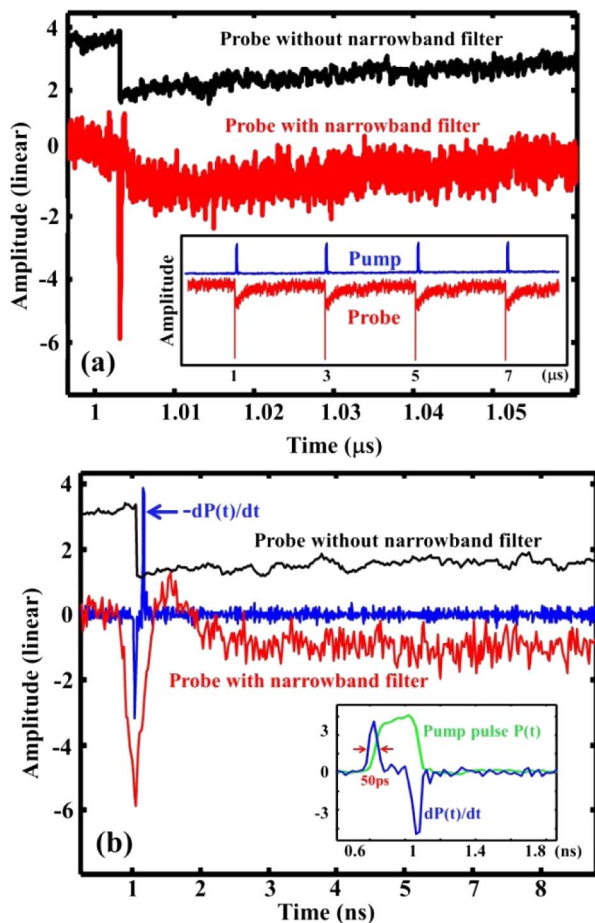
In our experiment with 10 GHz electrical bandwidth of the electro-optic modulator, the rising time of the pump pulse was approximately 50 ps. The leading edge of the optical phase modulation on the probe has the same time scale. The dynamic optical frequency shift of the probe can be estimated as

$$\delta f = \frac{1}{2\pi} \frac{d\varphi(t)}{dt} = \frac{1}{2\pi} \frac{1}{5 \times 10^{-11}} = 3.2 \text{ GHz.}$$

However, due to the limited pulse peak power used in our experiments, the material phase change was limited to only a small fraction of the maximum phase transition between the dielectric and the metallic states of  $\text{VO}_2$ . Therefore, the  $\delta f$  value calculated here only provides an upper limit, while the actual instantaneous frequency deviation of the probe obtained should be less than a gigahertz.

The pump-induced instantaneous optical frequency shift on the probe can be translated into an intensity modulation through a frequency discriminator such as an optical filter. As shown in Fig. 1, this frequency modulation (FM) to intensity modulation (IM) conversion can be accomplished by adding a tunable-fiber Fabry-Perot (FP) filter (shown in blue in Fig. 1) before the photodetector. This FP filter has 100 GHz





**Fig. 4.** (a) Comparison of probe waveforms measured without (top) and with (bottom) the narrowband FP filter. Inset: pump and probe waveforms in a longer time span. (b) Detailed view of probe waveform change in the transition region along with the inverse differential of the pump waveform ( $-dP(t)/dt$ ). Inset: 200 ps pump-pulse waveform  $P(t)$  in green and its time derivative  $dP(t)/dt$  in blue.

free-spectral range and a finesse of 100 so that the width of the passband is approximately 1 GHz, which is narrow enough to provide high FM/IM conversion efficiency for the probe signal with small FM modulation index. By setting the center of the FP passband to the static wavelength of the probe, an instantaneous frequency deviation of the probe would result in a reduced transmission through the FP filter, creating an IM that can be detected by the photodetector. Figure 4(a) compares the measured probe waveforms without (top) and with (bottom) the FP filter before the photodetector. A sharp notch clearly appears in the waveform when the FP filter was applied for the FM-IM conversion.

Figure 4(b) reveals further details of the probe waveforms near the transitional region and the comparison with the inverse differential of the pump waveform ( $-dP_{\text{pump}}(t)/dt$ ). The sharp notch in the FP-filtered probe waveform corresponds with the leading edge of the pump pulse, which instantaneously induces the phase transition in the VO<sub>2</sub> film, whereas the material phase transition after the falling edge of the pump

pulse is too slow to induce any meaningful frequency deviation on the probe. The inset of Fig. 4(b) shows the pump waveform  $P(t)$  with 200 ps pulse width together with its time derivative  $dP(t)/dt$ . This plot indicates that the differential of the pump-pulse leading edge has a temporal width of approximately 50 ps.

In conclusion, we have measured the phase transition of a 200 nm VO<sub>2</sub> film on a sapphire substrate at different pump pulse durations. The results show that the phase transition from dielectric to metallic state of VO<sub>2</sub> specifically depends on the energy of the excitation pump pulse, as long as the width of the pulse is much smaller than the phase recovery time from metallic back to the dielectric state. Moreover, during the leading edge of the pump pulse, a fast phase transition in the sample occurs and consequently an optical phase ( $\delta\phi$ ) transient is introduced in the probe. We have shown that the derivative of this optical phase transient causes an instantaneous frequency shift of the probe. Therefore, by using a narrowband optical filter centralized at the probe wavelength, we were able to transform the optical FM into an IM and consequently generate short pulses at the probe wavelength with the same repetition rate as the pump. The width of the optical pulse generated in the probe is independent of the phase recovery time. However, the periodicity of the probe pulse waveform has to be longer than the phase recovery time of the VO<sub>2</sub>.

**Funding.** Army Research Office (ARO) (0910295, W911NF-16-1-0029); National Science Foundation (NSF) (1105986, 1337737, 1508494).

## REFERENCES

- Q. F. Xu and M. Lipson, *Opt. Express* **15**, 924 (2007).
- A. Liu, R. Jones, L. Liao, D. Samara-Rubio, D. Rubin, O. Cohen, R. Nicolaescu, and M. Paniccia, *Nature* **427**, 615 (2004).
- Q. Xu, B. Schmidt, S. Pradhan, and M. Lipson, *Nature* **435**, 325 (2005).
- J. D. Ryckman, K. A. Hallman, R. E. Marvel, R. F. Haglund, and S. M. Weiss, *Opt. Express* **21**, 10753 (2013).
- H. Kim, N. Charipar, M. Osofsky, S. B. Qadri, and A. Piqué, *Appl. Phys. Lett.* **104**, 081913 (2014).
- J. Nag and R. Haglund, Jr., *J. Phys.* **20**, 264016 (2008).
- G. I. Petrov, V. V. Yakovlev, and J. A. Squier, *Opt. Lett.* **27**, 655 (2002).
- A. Cavalleri, C. Tóth, C. W. Siders, J. A. Squier, F. Ráksi, P. Forget, and J. C. Kieffer, *Phys. Rev. Lett.* **87**, 237401 (2001).
- H.-T. Kim, B.-G. Chae, D.-H. Youn, G. Kim, K.-Y. Kang, S.-J. Lee, K. Kim, and Y.-S. Lim, *Appl. Phys. Lett.* **86**, 242101 (2005).
- A. Cavalleri, T. Dekorsy, H. H. Chong, J. C. Kieffer, and R. W. Schoenlein, *Phys. Rev. B* **70**, 161102 (2004).
- R. M. Briggs, I. M. Pryce, and H. A. Atwater, *Opt. Express* **18**, 11192 (2010).
- J. M. Choi, R. K. Lee, and A. Yariv, *Opt. Lett.* **26**, 1236 (2001).
- T. V. Son, K. Zongo, C. Ba, G. Beydaghyan, and A. Haché, *Opt. Commun.* **320**, 151 (2014).
- A. Joushaghani, J. Jeong, S. Paradis, D. Alain, J. S. Aitchison, and J. K. S. Poon, *Opt. Express* **23**, 3657 (2015).
- H. Kim, M. Osofsky, R. C. Y. Auyeung, and A. Piqué, *Appl. Phys. Lett.* **100**, 142403 (2012).
- H. Kim, M. Osofsky, M. M. Miller, S. B. Qadri, R. C. Y. Auyeung, and A. Piqué, *Appl. Phys. Lett.* **100**, 032404 (2012).
- H. Kim, N. Charipar, E. Breckenfeld, A. Rosenberg, and A. Piqué, *Thin Solid Films* **596**, 45 (2015).
- J. T. Kim, *Opt. Lett.* **39**, 3997 (2014).