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## Pure optical phase control with vanadium dioxide thin films

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## ABSTRACT

Under certain conditions, films made of vanadium dioxide exhibit wavelengths at which transmittance or reflectance do not change as the material undergoes insulator to metal phase transition, in spite of refractive index changes on the order of unity. Exploiting this effect, we demonstrate control of optical phase at 800 nm in transmission and at 1310 nm in reflection. With a 68 nm film, the optical phase is adjusted while leaving all other properties of light unchanged, including amplitude, polarization and frequency. The phase change per unit of propagated distance is  $\Delta k = 107$  rad/m, orders of magnitude higher than typically obtained with electro-optic effects. We discuss potential application to nano-sized phase devices or thin film lenses.

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Vanadium dioxide ( $\text{VO}_2$ ) is a correlated material that undergoes a reversible insulator-to-metal transition (IMT) above 68 °C, owing to a rearrangement of its crystalline structure [1–2]. This phase transition is accompanied by an increase in conductivity and a sharp drop in optical transmittance in the infrared. This IMT has been exploited in a wide range of photonic applications, including tunable metamaterials [3–5], spectrally selective filters [6–7], optical switches and limiters [8–11] and infrared imaging systems [12–13].

So far, much research has focussed on the amplitude modulation capabilities of this material, which are indeed remarkable in both range of amplitude (orders of magnitudes) and spectral width, spanning from the near infrared to the terahertz. While phase shifting of THz beams by  $\text{VO}_2$  films has been demonstrated [14], the capabilities of the material for phase and polarization modulation have been relatively unexplored. Yet, refractive index changes of  $\text{VO}_2$  during phase transition are on the order of unity, with the potential of phase control in thin films. While conventional techniques for phase shifting and modulation use piezoelectric actuators, the electro-optic effect and liquid crystals, the prospect of phase modulation with  $\text{VO}_2$  thin films is interesting for at least three reasons. First, conventional techniques are implemented in devices that are micrometers to centimeters in size, while  $\text{VO}_2$  thin films offer the possibility of nanometer-sized phase devices. Second, it is sometime difficult to disentangle phase control from polarization and amplitude effects. For example, in some configurations of Pockels cells or liquid crystals, the material

anisotropy causes the polarization state of light to change, which in turn modifies the field amplitude when combined with polarization-dependant optical components. With  $\text{VO}_2$  thin films, as this paper demonstrates, it is possible to shift the optical phase of light beams without affecting its amplitude, frequency and polarization. Such “pure phase” control is made possible in both transmission and reflection at specific wavelengths where  $\text{VO}_2$  exhibits the same reflectivity or transmittance in its metallic and insulating states. Third, non-uniform phase shift profiles in the film could lead to beam focusing and nanometer-sized flat lenses.

As above mentioned, when a  $\text{VO}_2$  film undergoes a phase transition from insulator to metal, a drop in transmittance is generally observed across all wavelengths in the infrared, while the reflectance increases accordingly. However, in some samples, typically with thicknesses of about 100 nm or less, there are specific wavelengths where transmittance or reflectance does not change. Fig. 1(a) shows an example of such effect. The transmittance, here measured at normal incidence, is plotted for the insulating and the metallic states of the  $\text{VO}_2$  film, with an arrow indicating where the two curves cross over (825 nm). A similar effect is observed in reflection at 1300 nm, as shown in Fig. 1(b). Both graphs were obtained with a 68 nm-thick layer of  $\text{VO}_2$  deposited on glass using a technique described in Refs. 15–16. In Fig. 2 are images taken by a scanning electron microscope and an atomic force microscope revealing the characteristic microstructure of the  $\text{VO}_2$  films deposited on glass. During the film deposition, only half of the substrate was exposed, and during post treatment, the whole film was treated, creating a uniform film to the edge (a necessary condition for experiments described later).

A priori, there are two possible explanations for why transmittance or reflectance may remain the same during phase transition at some wavelengths: 1) the optical constants of the material

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happen to be the same in both material phases at these wavelengths, or 2) the optical constants do change, but interference effects lead to unchanged optical properties. The first hypothesis can be verified by measuring the film's complex refractive index (given as  $n+ik$ ) by ellipsometry.

We used an ellipsometer adapted to measure the film reflectance and transmittance simultaneously at temperatures ranging from 20 to 90 °C. The home-built instrument uses silicon and InGaAs detectors covering the 400 to 2000 nm spectral range and measures for incidence angles from 10 to 80°. Once the field amplitudes of the  $s$  and  $p$  polarizations are measured, the optical constants are extracted by fitting to a fourth-order polynomial, appropriate for non-dielectric media. The model assumes a single,

uniform layer of VO<sub>2</sub> on glass. The refractive indices shown in Fig. 3 were obtained. While keeping in mind that optical constants of VO<sub>2</sub> are sensitive to stoichiometry and microstructure, we compared our ellipsometry results with that of Refs. 17 and 18 and found many similarities. During phase transition, the real part of the refractive index drops considerably, namely  $\Delta n = -0.82$  at 825 nm and  $\Delta n = -1.11$  at 1300 nm. Meanwhile, the imaginary part varies by  $\Delta k = 0.28$  and  $\Delta k = 1.63$  at the same respective wavelengths. We can therefore conclude that interference effects are responsible for the constant transmission and reflection.

From ellipsometry data, we verify theoretically a possible optical phase shift associated to these refractive index changes by calculating the phase of the complex field amplitudes in transmission  $t$  and in reflection  $r$  in a thin film

$$r = r_{12} - \frac{t_{12}t_{21}}{r_{21}} \left( \frac{1}{1 - r_{12}r_{23}e^{2i\phi}} - 1 \right) \quad (1)$$

$$t = \frac{t_{12}t_{23}e^{i\phi}}{1 - r_{23}r_{21}e^{2i\phi}} \quad (2)$$

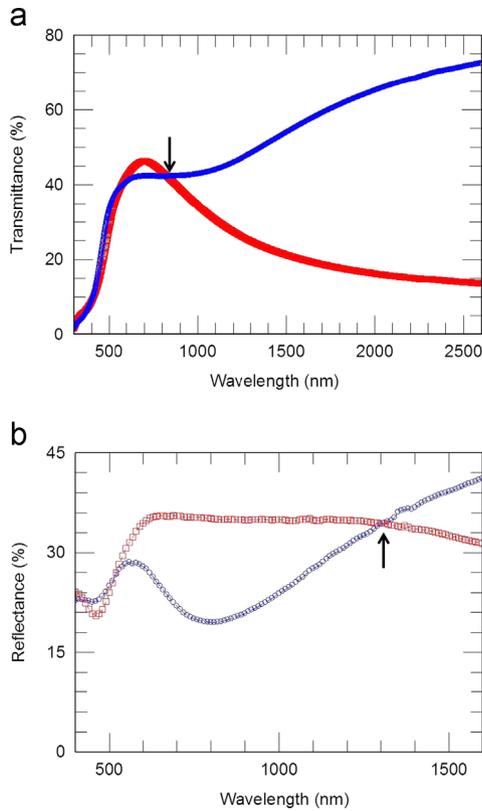
where  $\phi$  is the phase through a film of thickness  $d$

$$\phi = \frac{2\pi d}{\lambda}(n+ik) \quad (3)$$

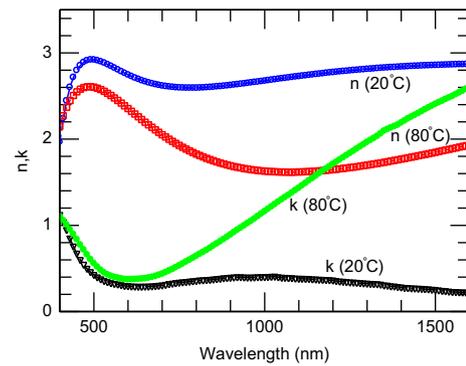
and the following Fresnel coefficients are used:

$$r_{12} = -r_{21} = \frac{1-n}{1+n} \quad (4)$$

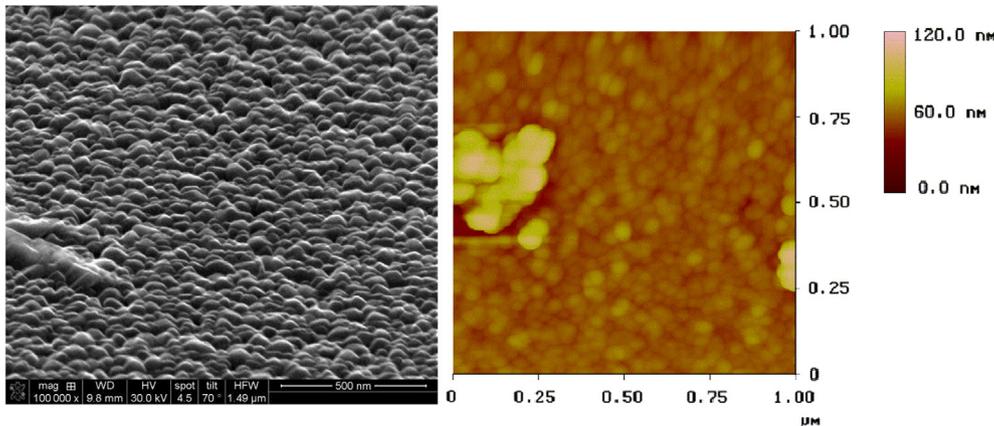
$$r_{23} = \frac{n-n_t}{n+n_t} \quad (5)$$



**Fig. 1.** (a). Transmittance of a VO<sub>2</sub> film at temperatures of 23 °C (blue) and 80 °C (red). (b). Reflectance of film of VO<sub>2</sub> at temperatures of 23 °C (blue) and 80 °C (red). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)



**Fig. 3.** Optical constants of a 68 nm VO<sub>2</sub> film measured by ellipsometry.



**Fig. 2.** SEM (left) and AFM (right) images showing the microstructure of the VO<sub>2</sub> films deposited on glass.

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