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Polarization modulation by vanadium dioxide on metallic substrates

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Abstract

Vanadium dioxide (VO_2) undergoing phase transition is known alters the polarization state of light in reflection owing to large changes in complex refractive indices. While this effect is promising for optical modulation applications, the usual VO_2 films on dielectric substrates tend to offer limited tunability for polarization modulation. In this paper, we show that metallic under-layers greatly enhance the performance by widening the spectral range and include visible wavelengths, by increasing the polarization modulation amplitude, and by widening the range of workable incidence angles. The imaginary part of the refractive index in the metallic layer is found to increase the relative phase shifts between s- and p-components of polarization as well as increasing the reflectance.

Keywords: Vanadium dioxide, Phase transition material, Polarization control, Light modulator, Phase control.

1. Introduction

Vanadium dioxide (VO₂) has found many applications in optics owing to its large refractive index variations when it transitions from an insulating to a metallic state. The phase transition can be activated thermally, by heating to a temperature of 68 $^{\circ}$ C [1], or on faster time scales by optical excitation [2–5]. Changes in optical properties have been useful mostly for applications of spectral filtering and field-amplitude modulation of infrared light, but recently, applications have also been found for phase and polarization modulation of visible and infrared light [6–10]. Phase and polarization modulations rely on the principle that in a material with complex-valued refractive indices like VO₂, optical phase shifts are generally unequal in s- and p-polarizations. A beam of light reflecting off a VO₂ layer therefore sees its polarization state change as the material undergoes a phase transition, making possible the rotation of linear polarization, among other possibilities [9,10].

One advantage of phase and polarization modulation over field amplitude modulation is a greater sensitivity over a wider spectrum: whereas applications of VO₂ were traditionally limited to wavelengths above 1000 nm [11,12], phase modulation has extended to range to include parts of the visible spectrum (> 500 nm). However, preliminary studies [7,9,10] showed that VO₂ layers on dielectric substrates only enable substantial polarization modulation for incident angles near Brewster's angle, i.e. $\sim 70^{\circ}$ in the case of VO₂. While we should not expect such applications to be possible at near normal incidence angles, where s- and p-polarizations become indistinguishable, it would be desirable to lower the angle for some applications.

In this paper, we study the effect of metallic substrates on polarization modulation of light by a VO_2 layer, a system which so far has only been studied in the context of perfect absorbers [13]. Here we show that not only a VO_2 /metal interface makes polarisation modulation possible at lower incidence angles and over a wider spectral range compared with VO_2 -on-dielectric layouts, it also increases reflectance significantly. Note that the term "metallic substrate" does not imply that the substrate should be made of bulk metal, only optically thick. In most cases, metal layer thicknesses greater than 100 nm would be sufficient.

2. Theory

The parameters relevant to the problem are defined in Figure 1. A beam of light with s- and ppolarization components arrives at an angle θ_i on a VO₂ layer of thickness *d* in contact with an optically thick layer of metal. Looking at the properties of the reflected light, we define $\delta = \phi_p - \phi_s$ as the phase difference between the two polarization components in reflection. This phase difference, along with the amplitudes of each polarization components, determine the polarization state of the reflected light. Since we are interested in how polarization is changed by the phase transition of VO₂, we need to consider $\Delta = \delta_1 - \delta_0$, the change in relative phase occurring during the phase transition, with δ_1 the relative phase when VO₂ is activated (metallic state) and δ_0 the relative phase when it is not activated (insulating state).



Fig 1. Sample geometry and definition of optical parameters.

Two cases are of particular interest: 1) if $\Delta = \pm \pi$, linearly polarized light may rotate by some angle while remaining linearly polarized, and 2) if $\Delta = \pm \pi/2$, linear polarization may become circular, or vice-versa. The first case is useful for producing large modulation amplitudes in transmission through polarizers.

From the values of the refractive indices of VO₂ and the metal, we can calculate the reflection amplitudes for either s- or p-polarizations and for the two states of VO₂, namely $r_{p,0}$, $r_{p,1}$, $r_{s,0}$ and $r_{s,1}$. These quantities are complex and contain the phase information of each polarization component relative to that of incident light. We then obtain the phase modulation term with

$$\Delta = \arg(z_r)$$

$$z_r = \frac{r_{p,1}r_{s,0}}{r_{p,0}r_{s,1}}$$

Since the film's Fresnel coefficients $r_{p,0} \dots r_{s,1}$ are functions of incidence angle θ_i and film thickness d, and since refractive indices depend on wavelength, the phase modulation term Δ also depends on all three parameters. As a result, at a given wavelength, there could be many sets of conditions where $\Delta = \pi$ is possible, for example.

3. Theoretical Results

where

To better visualize the role of each parameter and capture the full range of possibilities, we calculated Δ for many combinations of incidence angles and VO₂ film thicknesses. As a reference, we first calculate for the case of VO₂ deposited on amorphous silica. Figure 2 shows the result, with θ_i varied from 0 to 90° and *d* from 0 to 300 nm (the typical range of achievable VO₂ film thicknesses). Calculations use refractive index of VO₂ and amorphous silica experimentally measured by ellipsometry.





We note that VO_2 film thicknesses of less than 100 nm are sufficient. However, there is a narrow range of incidence angles, between 60 and 70°, where substantial phase modulation is possible, and modulation is much less pronounced in the visible spectrum as it is in the near infrared. Calculations for VO_2 on gold, on

(1)

(2)

the other hand, reveal an entirely different picture, as Figure 3 shows. Modulation is possible in the visible spectrum and the range of possible incidence angle reduced to 10° in some cases. While the required thicknesses to achieve $\Delta = \pi$ tend to increase, a side benefit is an enhanced reflectance due to the metallic layer. By calculating reflectance for all incidence angles, polarizations, and wavelengths from 400 nm to 2000 nm, an averaged reflectance of 0.2 is obtained for VO₂ layers (all thicknesses) on glass, whereas it increases to 0.4 for a 75 nm of VO₂ on gold, and to 0.6 for 50 nm of VO₂ on gold. If the VO₂ layer is thick (say, more than 200 nm), a metallic substrate has negligible effect because only a small fraction of incident light reaches it.



Fig 3. Relative phase modulation Δ during phase transition as a function of incidence angles and film

thickness for VO₂ on gold. The color code is normalized to $\Delta = \pi$.

To investigate the spectral range of phase modulation, Figure 4 shows the largest values of Δ , obtained by optimizing *d*, plotted as a function of wavelength and incidence angle. For VO₂ on dielectrics (amorphous silica and sapphire), sizeable phase modulation is limited to high incidence angles and for wavelengths above 600 nm. For VO₂ on gold and aluminum, the range of incidence angle is much widened, and phase modulation appears to be possible down to wavelengths as low as 400 nm.

VO2/SIO VO₂/Sapphire Incidence Angle ([°]) 00 00 00 0.9 0.8 0.7 0.6 0.5 VO₂/Au VO₂/AI 0.4 Incidence Angle (0.3 0.2 Wavelength (nm) Wavelength (nm)

Fig 4. The largest relative phase modulation Δ obtained as a function of incidence angle. Color code: blue $\Delta < \pi/2$, green $\pi > \Delta > \pi/2$, yellow $\Delta = \pi$.

But the enhancement is not observed with gold and aluminum substrates. Figure 5 reveals additional results for copper, silver and titanium. Here, the minimum incidence angles at which $\Delta = \pi$ is possible are plotted as a function of wavelength. The wavelength values where each curve begins and ends correspond to the shortest and longest wavelengths where $\Delta = \pi$ is possible. All metals studied here enable the lowering of incidence angle as compared to dielectric substrates. For applications in the visible spectrum, titanium appears to be particularly promising.

Calculations suggest that the imaginary part of the refractive index is largely responsible for the enhancements. For example, the results of Figure 4 are nearly the same if one tweaks only the real part of the refractive index; changing the absorption coefficient, on the other hand, alters the curves drastically. This is perhaps not surprising given the fact that Fresnel coefficients have the same phase for s- and p-polarizations when materials are dielectric, but different when they are absorbent.



Fig 5. Minimum incidence angle for achieving $\Delta = \pi$ with VO₂ films on various metallic substrates.

4. Experiments

In order to verify some of the properties described above, gold and aluminum films of 100 nm or more in thickness were deposited on glass substrates by thermal evaporation under vacuum. VO₂ films were then deposited on top of the metallic films by sputtering of metallic vanadium followed by post-treament oxidation at 500°C in one hour. The thickness of VO₂ layers was kept in the 85-100 nm range, values for which calculations show large enough phase shifts at wavelengths from 800 to 1000 nm. The complex refractive indices of gold and VO₂ were measured by ellipsometry and shown in Figure 6. The VO₂ film exhibits refractive index changes on the order of unity during the phase transition. While the refractive index of VO₂ varies from sample to sample according to the exact preparation conditions, the variations are typically small (\pm 5% or less).



Fig 6. (a) Real (n) and imaginary (k) parts of the refractive indices of gold, (b) real and (c) imaginary parts of VO₂ films at insulating state (25° C) and metallic state (95° C).

Figure 7 shows the experimentally measured relative phase modulations Δ at different incidence angles for a 87 nm thick film of VO₂ on glass, gold and aluminum. At most angles of incidence, VO₂ on glass produced values for Δ of less than 1 radian while in VO₂ on metals, it ranges from $-\pi$ to π . For VO₂ on glass samples, the incidence angle should be near Brewster's angle (greater than 65°) to attain $\Delta > \frac{\pi}{2}$. The performance of aluminum is especially remarkable given the fact that ellipsometry suggests that a 55 nm thick layer with the properties of Al₂O₃ formed between Al and VO₂ during the fabrication process. This oxide layer could be avoided by using a glass/VO₂/Al structure where light arrives on VO₂ through the glass substrate.

The thickness of VO₂ was chosen to achieve maximum Δ at 832 nm, the laser wavelength used in polarization rotation experiments. Thinner or thickner VO₂ layers would cause the peak of Δ to move to either shorter or longer wavelengths, as described in Figure 3.



Fig 7. Relative phase shift between s- and p-polarizations during phase transition of VO₂ on (a) glass, (b) gold and (c) aluminum.

Figure 8 shows a polarization rotation experimental setup with light at 832 nm (Mellet Griot diode laser) by a sample of VO₂ on metal. In this experiment, the laser beam is analyzed through a polarizer (P₂) after it reflects off sample (S) and compared for cases when VO₂ is insulating (room temperature) and metallic (high temperature). Since the refractive index is complex at room temperature, the linearly polarized beam becomes elliptical after reflection; to make it linear after reflection, we prepare the incident polarization accordingly with a KDP crystal acting as an adjustable phase compensator (C).

This way, we can rotate the linear polarization upon reflection by inducing only a phase transition in VO_2 , as Figures 9a-c show. At 50° incidence angle, rotation of linear polarization occurs, but at 40°, only conversion between linear and circular polarization is possible, in agreement with the results of Fig. 7b. Also shown are the corresponding modulation contrast ratios, defined as the ratio of the intensities transmitted through the polarizer at a fixed polarizer angle. This graph demonstrates the potential for applications in optical modulation. Even in cases where the polarization does not remain linear (e.g. Fig. 8c), contrast ratios of 25 can still be achieved.



Figure 8. Experimental setup for polarization rotation by a VO_2 sample (S) on metal. A phase compensator (C) renders the laser beam polarization linear upon reflection when VO_2 in its insulating state. The polarization state is analyzed with polarizer P_2 and detector D.







Fig 9b. Polarization state of reflected light at 50° incidence angle and the corresponding contrast ratio.



Fig 9c. Polarization state of reflected light at 40° incidence angle and the corresponding contrast ratio.

The results above confirm that the gold layer underneath the VO_2 film remarkably enhances polarization modulation and helps to lower the incidence angle.

5. Conclusions

Using calculations and experimental measurements, we investigated the relative phase shifts between s- and p-components of polarization occurring during the phase transition of a VO_2 layer on metallic substrates. Calculations used refractive indices obtained experimentally and phase shifts were also measured directly using ellipsometry and by polarized laser beam interaction with VO_2 samples.

Metallic substrates such as gold, aluminum, titanium, copper or silver were found to significantly

enhance the ability of VO_2 to modulate the polarization of light during the phase transition. The main advantages are to widen the spectral range of modulation to include parts of the visible spectrum, and lower the angle of incidence. With VO_2 on dielectrics, the phase shift is only significant near Brewster's angle and for a narrower range of wavelengths. The combination of VO_2 with metallic layers is therefore promising for light modulation applications.

References

- [1] F.J. Morin, Oxides Which Show a Metal-to-Insulator Transition at the Neel Temperature, Phys. Rev. Lett. 3 (1959) 34–36.
- [2] M.F. Becker, A.B. Buckman, R.M. Walser, T. Lépine, P. Georges, A. Brun, Femtosecond laser excitation of the semiconductor- metal phase transition in VO2, Appl. Phys. Lett. 65 (1994) 1507– 1509.
- [3] A. Cavalleri, C. Tóth, C.W. Siders, J.A. Squier, F. Ráksi, P. Forget, J.C. Kieffer, Femtosecond Structural Dynamics in VO2 during an Ultrafast Solid-Solid Phase Transition, Phys. Rev. Lett. 87 (2001) 237401.
- [4] S. Lysenko, A.J. Rua, V. Vikhnin, J. Jimenez, F. Fernandez, H. Liu, Light-induced ultrafast phase transitions in VO2 thin film, Appl. Surf. Sci. 252 (2006) 5512–5515.
- [5] M. Rini, Z. Hao, R.W. Schoenlein, C. Giannetti, F. Parmigiani, S. Fourmaux, J.C. Kieffer, A. Fujimori, M. Onoda, S. Wall, A. Cavalleri, Optical switching in VO2 films by below-gap excitation, Appl. Phys. Lett. 92 (2008) 181904.
- [6] T.V. Son, C.O.F. Ba, R. Vallée, A. Haché, Nanometer-thick flat lens with adjustable focus, Appl. Phys. Lett. 105 (2014) 231120.
- [7] T.V. Son, V.V. Truong, J.-F. Bisson, A. Haché, Nanosecond polarization modulation in vanadium dioxide thin films, Appl. Phys. Lett. 111 (2017) 041103.
- [8] T.V. Son, K. Zongo, C. Ba, G. Beydaghyan, A. Haché, Pure optical phase control with vanadium dioxide thin films, Opt. Commun. 320 (2014) 151–155.
- [9] T.V. Son, V.V. Truong, P.A. Do, A. Haché, Ultra-thin, single-layer polarization rotator, AIP Adv. 6 (2016) 085102.
- [10] P. Cormier, T.V. Son, J. Thibodeau, A. Doucet, V.-V. Truong, A. Haché, Vanadium dioxide as a material to control light polarization in the visible and near infrared, Opt. Commun. 382 (2017) 80–85.
- [11] M. Soltani, M. Chaker, E. Haddad, R. Kruzelesky, 1 × 2 optical switch devices based on semiconductor-to-metallic phase transition characteristics of VO 2 smart coatings, Meas. Sci. Technol. 17 (2006) 1052.
- [12] S. Zhang, M.A. Kats, Y. Cui, Y. Zhou, Y. Yao, S. Ramanathan, F. Capasso, Current-modulated optical properties of vanadium dioxide thin films in the phase transition region, Appl. Phys. Lett. 105 (2014) 211104.
- [13] J. Liang, L. Hou, J. Li, Frequency tunable perfect absorber in visible and near-infrared regimes based on VO2 phase transition using planar layered thin films, JOSA B. 33 (2016) 1075–1080.