

Nonreciprocal switching of VO₂ thin films on microstructured surfaces

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We demonstrate that the combination of near-field focusing and a switchable oxide layer permits construction of a modulator with different optical switching thresholds in two opposing directions. For that purpose VO₂ layers are deposited onto spherical micrometer-sized particles. The phase transition in VO₂ is induced by a nanosecond-pulsed Nd:YAG laser from two different directions. The measured thresholds differ by a factor of 2.4, consistent with calculated differences in the intensities in the two directions. © 2010 Optical Society of America

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Vanadium dioxide is one of the most studied transition-metal oxides because of its reversible thermochromism. It undergoes a semiconductor-to-metal phase transition (SMT) at a critical temperature T_c of 68 °C [1]. The optical and electrical properties of the oxide change significantly above T_c , resulting in dramatically decreased transmission through metallic VO₂ in the near-infrared. Current application interest in VO₂ is driven by the fact that the phase transition can also be induced optically by a pulsed laser [2–7]. While optical modulation using VO₂ has been demonstrated on nanoparticle arrays [8] and sub-wavelength hole arrays [9], light-induced switching has not to our knowledge been demonstrated on thin film structures with well-defined curvature.

In this Letter, we demonstrate the use of near-field focusing to implement nonreciprocal optical switching in a thin layer of VO₂ deposited on silica (SiO₂) microspheres. Optical transmission measurements show a difference in the intensity threshold for switching depending on the direction of incidence of the switching beam with respect to the planar microstructure. This experiment thus represents the first practical use of the third-order nonlinearity in VO₂ [10] to implement a planar microstructured optical switch.

The samples were prepared in two steps. A colloidal solution of SiO₂ particles with a diameter of 1.54 μm was deposited by micropipette onto clean glass substrates, after which the solvent was allowed to evaporate in ambient atmosphere. Solvent evaporation leads to the formation of compact structures including monolayers of silica particles on portions of the substrates [11,12]. Then 140-nm-thick films of VO₂ were deposited on the microstructures by pulsed laser deposition followed by thermal oxidation [13]. The film thickness near the top center of the spheres is expected to be identical to that on the bare substrate; however, the film thickness probably decreases with increasing angles from the deposition axis [14].

The SMT in the microstructured oxide layers was induced by a frequency-doubled pulsed Nd:YAG laser (532 nm); the beam spot on the sample was approximately circular with a diameter of order 5 mm; the pulse duration was approximately 5 ns, and the typical fluence was around 10 mJ/cm². Optical transmission was measured by using a cw laser diode (980 nm) with a Gaussian profile, a diameter of less than 1 mm, and an output power less than 10 mW. At this power level, heating by the probe can be neglected. Two different measurement configurations were employed, as shown in Fig. 1. In configuration 1 (C1), the heating and probing beams were incident on the sample from the substrate side [Fig. 1(a)]. In configuration 2 (C2), the heating beam was incident from the film side, while the probe beam was incident from the substrate side [Fig. 1(b)]. The beams were adjusted to be collinear and parallel to the surface normal and with the probe beam centered in the heating-beam spot. Filters and polarizers in front of the photodiode blocked the Nd:YAG beam to prevent any contribution of the Nd:YAG light to the transmission signal. The data were saved in a fast digital oscilloscope and normalized to the transmission in the semiconducting state.

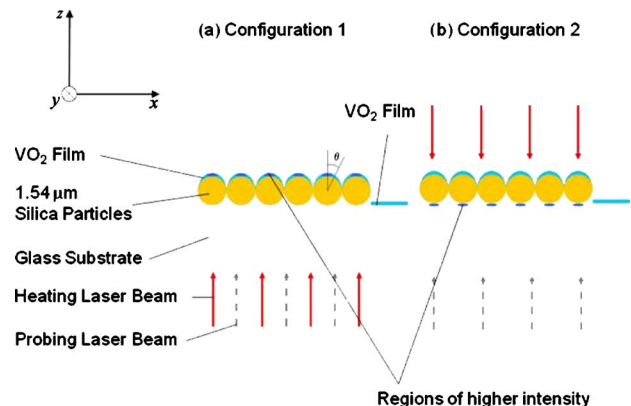


Fig. 1. (Color online) Schematic diagram of the samples and the two measurement configurations.

A typical plot of normalized transmission versus time is shown in Fig. 2. The transmission exhibits a fast transient decrease (A_1) when the Nd:YAG pulse initiates the SMT, followed by a slower decrease (A_2). We attribute the initial fast drop (A_1) to the light-induced switching. The slower drop (A_2), also observed in femtosecond-laser-induced switching experiments [15], may be ascribed to the subsequent lateral diffusion of heat out of the probe-beam focal spot. In Fig. 3, we compare the transmission ratio A_1/I_0 for the two measurement configurations as a function of the Nd:YAG laser pulse energy, where I_0 is the transmitted intensity in the semiconducting VO_2 state. Filled circles refer to the values obtained in C1, while filled squares are for C2. The amplitude drops are maximal at 1.2 and 2.9 mJ/pulse for C1 and C2, respectively, as indicated by the arrows. We define the energy values at which the amplitudes saturate as the switching thresholds for the SMT.

For comparison, the value of A_1 for bulk films is also shown in Fig. 3. The amplitude decrease for bulk films is a factor of ~ 4 greater than that for $\text{VO}_2:\text{SiO}_2$ in C2, and the two curves reach their plateau values at different intensities. The $\text{VO}_2:\text{SiO}_2$ shows a plateau, signifying attainment of the metallic state, at a lower threshold, consistent with a much smaller volume of material reaching the transition temperature. The measured ratios A_1/I_0 as a function of heating-laser pulse energy can be fitted well by a single exponential function $A_1/I_0 = R_0 \exp[-E_{\text{laser}}/\Delta]$, where Δ is the energy required to switch a fraction $(1-1/e)$ of the VO_2 from insulating to metallic. The values of Δ calculated from the fits are 1.099 ± 0.08 mJ for the bulk film, 1.096 ± 0.11 mJ for VO_2 in C2, and 0.285 ± 0.018 mJ for C1.

The key observation from Fig. 3 is that the switching threshold and switching energy are strongly affected by the direction of incidence of the heating beam. The threshold in C2 is a factor of ~ 2.4 larger than that measured in C1. At a laser intensity of ~ 1.2 mJ/pulse, switching is nearly complete in C1, while there is still no change in the amplitude in C2.

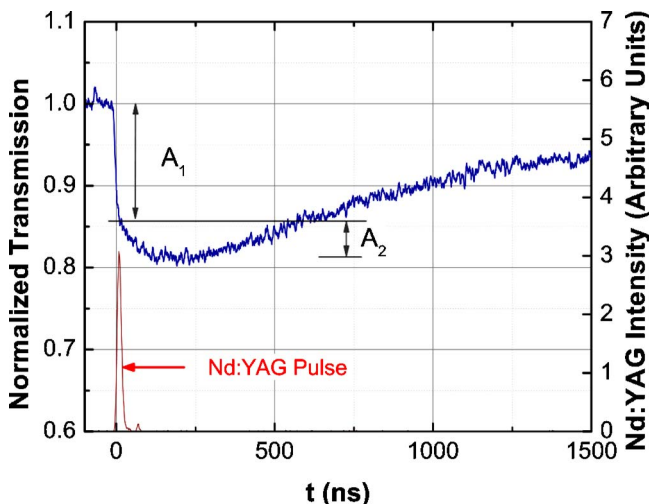


Fig. 2. (Color online) Normalized transmission through 140 nm VO_2 bulk film as a function of time at 980 nm (in C2).

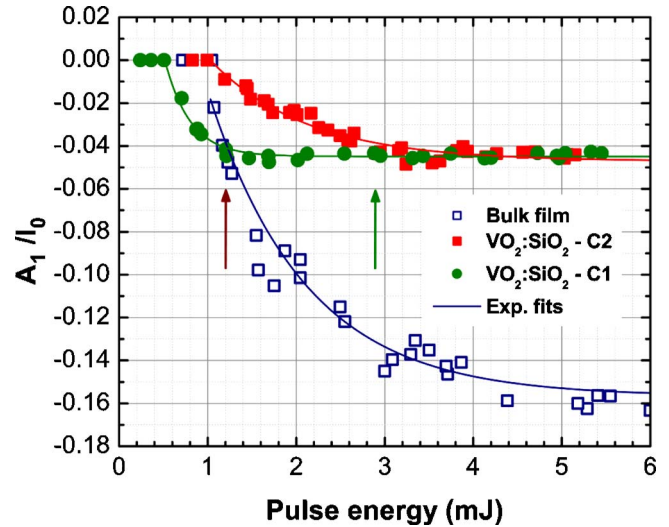


Fig. 3. (Color online) Amplitude A_1 normalized by the transmitted intensity I_0 in the semiconducting state, for $\text{VO}_2:\text{SiO}_2$ in both measurement configurations and for bulk film, as a function of Nd:YAG pulse energy at a wavelength of 980 nm. The arrows indicate the beginning of the plateaus.

The very large difference in switching thresholds and in Δ for C1 and C2, and the similarity between switching in the bulk film and in C2, can be explained by the near-field focusing effect of the silica microspheres in C1. A micrometer-size particle under laser illumination acts as a lens in the near-field region when its size is comparable with the wavelength of the light. The result is a local intensity enhancement in the optical near field that has been exploited successfully in laser nanostructuring [16,17] and in superresolution optical data storage [18,19]. This inference is consistent with simulations carried out by using the three-dimensional finite-difference time-domain code Lumerical. As shown in Fig. 4, the sample was modeled as a hexagonally close-packed array of $1.54 \mu\text{m}$ diameter silica spheres capped with a 140 nm thick layer of VO_2 . Figures 4(a) and 4(b) show a side view (X - Z plane, in the coordinate system of Fig. 1) in C1 and C2, respectively, with the direction of the heating-laser beam shown by the white arrows. The near-field focusing effect is evident in C1, where there is a strong maximum in intensity just at the top surface of the sphere. In C2, on the other hand, the much lower intensity maximum is near the bottom of the sphere. Figures 4(c) and 4(d) show a top view of the (X - Y) plane $1.54 \mu\text{m}$ above the substrate. The intensity at the center of the VO_2 capping layer is markedly less when the heating laser is incident from above (C2) than from below the sphere (C1).

Figure 5 shows a plot of calculated heating-laser intensity as a function of z (in micrometers) as measured from the bottom of the sphere. The intensity in C1 at $z = 1.54 \mu\text{m}$, where the heating laser enters the layer of VO_2 , is approximately 5.6 times the intensity at $z = 1.68 \mu\text{m}$ (C2). The $1/e$ absorption length of VO_2 at 532 nm is approximately $0.140 \mu\text{m}$; however, the absorption length calculated from the attenuation of

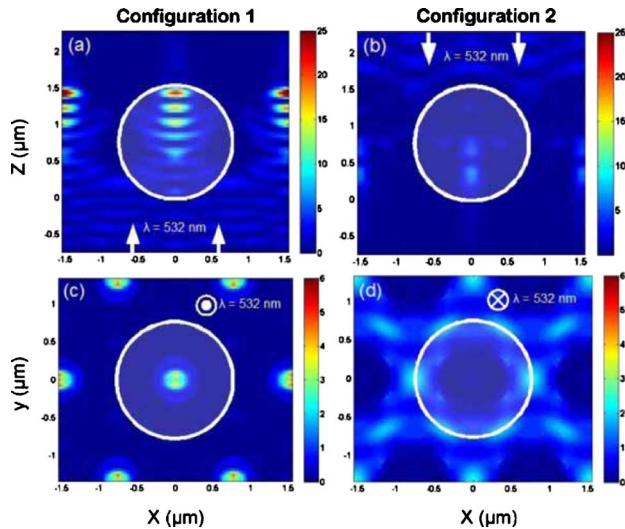


Fig. 4. (Color online) False-color, two dimensional intensity profiles of calculated heating-beam intensities for different measurement planes. The heating-laser beam is always incident along the z axis; all intensities are normalized to unit input. Cut through the center of the microsphere array in the X - Z plane in (a) C1 with beam incident from below and (b) in C2 with beam incident from above. Calculated intensity in the X - Y plane at the surface of the microsphere array with (c) beam incident from below and (d) with beam incident from above. Note the difference in the intensity scales in (a), (b) and (c), (d).

the forward-going wave in C1 is near $0.025 \mu\text{m}$. This calculated result is consistent with the hypothesis that nonlinear, near-field absorption effects are the proximate cause of the lowered switching threshold

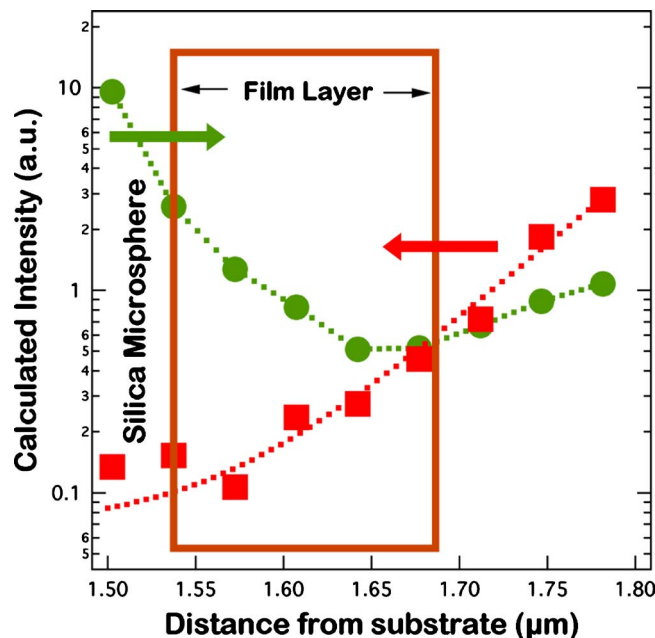


Fig. 5. (Color online) Logarithmic plot of the calculated pump-beam intensity (markers) as a function of distance from the substrate for the forward (green circles, C1) and backward (red squares, C2) illumination of the SiO_2 microspheres covered with VO_2 . The dotted curves connecting the markers are only guides to the eye.

in C1 and with the measured difference in the switching energies for C1 and C2 (Fig. 3).

In conclusion, the threshold for transmission switching through VO_2 films deposited on the curved surfaces of SiO_2 particles shows a difference depending on whether the heating beam is incident on the sample from the substrate or the film side. The observed nonreciprocal behavior can be explained by the near-field focusing effect of the spherical particles on which the thin VO_2 film is deposited.

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