

Surface plasmon polaritons in VO₂ thin films for tunable low-loss plasmonic applications

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We report on the first observation of optically excited surface plasmon polaritons (SPPs) in the conducting phase of vanadium dioxide (VO₂) thin films. VO₂ is low-loss optical material that undergoes an insulator-metal transition (IMT) under suitable thermal, optical, or electrical stimulation, thus enabling tunable SPP excitation of the conducting phase. Here we applied IR light (1520 nm) to excite SPPs while thermally inducing the IMT by changing the VO₂ temperature, and observed a clear trend from nonabsorption in the insulator phase to high absorption in the conducting phase due to SPP excitation in the latter phase. Tunable SPPs in VO₂ enable a range of opportunities for low-loss optoplasmonic applications since the rate of the IMT excitation can also be tailored. © 2012 Optical Society of America

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Conventional noble metals, such as Au and Ag, have traditionally been used in the field of metamaterials and plasmonics, opening up a number of exciting possibilities [1–3]. However, due to their large optical losses and large negative real permittivities, metals are not suitable for novel metamaterial devices [4–7]. Reducing losses in metals or searching for alternative low-loss materials is of paramount importance to expand the range of applications. Conducting metal oxides such as indium tin oxide (ITO) [5], aluminum doped zinc oxide [8], and ruthenium oxide [9] are alternatives to noble metals for use in metamaterial and plasmonic applications due to their low optical losses in the visible and near-infrared ranges. Of special interest is vanadium dioxide (VO₂), which has been known for a long time as a material exhibiting an insulator-metal phase transition (IMT) around 340 K. During the IMT, it is known that VO₂ transforms from a monoclinic structure to a tetragonal structure. The electrical and optical properties of VO₂ are also dramatically changed, the conductivity jumps by a factor as large as 10⁵ [10], and the infrared (IR) transmission decreases around 60% [11]. Interestingly it has also been reported that the IMT can be optically [12] and electrically [13] induced. The phase-dependent optical properties of VO₂ make it an outstanding candidate to achieve surface plasmon polariton (SPP) tunability, since SPPs can be excited in the conducting or metallic phase and then tuned by applying adequate light, heat, or electric field excitation. Besides, VO₂ has a small real part of the permittivity and relative low losses in the IR region, which makes it suitable as a potential low-loss alternative plasmonic material. This tunable plasmon excitation in VO₂ enables interesting opportunities for plasmonic components such as switching devices.

In this manuscript, we present an experimental and theoretical investigation of the excitation of SPPs in VO₂ thin films for their possible applications to tunable plasmonic elements. The structural, optical, electrical, and plasmonic properties of VO₂ thin films grown on

c-Al₂O₃ substrates were investigated, and the results provided a path on how to tailor VO₂ thin films for optimized excitation and tunability of SPPs.

VO₂ thin films were prepared on *c*-Al₂O₃ substrates using the reactive biased target ion beam deposition technique, which is described at length elsewhere [13]. Samples with film thicknesses ranging from 50 to 100 nm were investigated in order to study the thickness dependence of the SPP excitation. The structure of the VO₂ thin films was characterized by x ray diffraction (XRD) using a Rigaku diffractometer with Cu K α radiation. Figure 1(a) shows XRD symmetric scans ($\theta - 2\theta$ scan) of three VO₂ thin films with different thicknesses on *c*-Al₂O₃ substrates. Only M-VO₂ (020) (M means monoclinic structure) peaks are observed for all the thicknesses, indicating single phase VO₂ thin film. The average grain sizes extracted from the width of the XRD reflections increase from 52 to 88 nm for films with thicknesses ranging from 50 to 100 nm.

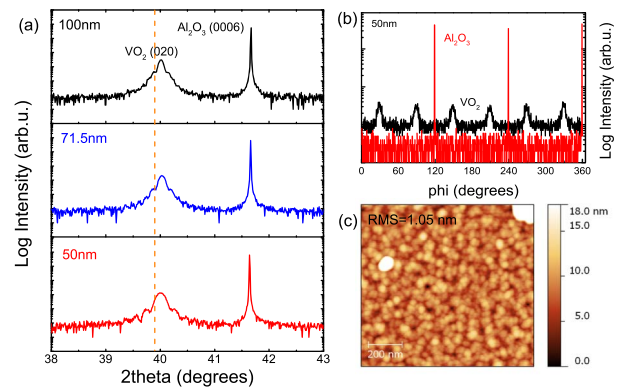


Fig. 1. (Color online) (a) XRD $\theta - 2\theta$ scans for VO₂ thin films on *c*-Al₂O₃ substrates with different film thicknesses. A single phase VO₂ (020) was observed. The dashed vertical line shows the position of the bulk VO₂ (020) peak. (b) XRD ϕ scans of the (110) planes of the 50 nm-thick VO₂ thin film and the (2204) planes of the Al₂O₃ substrate. (c) AFM image of the 50 nm-thick VO₂ film surface.

Asymmetric XRD phi scans for the M-VO_2 (110) and Al_2O_3 (2204) planes were performed to investigate the in-plane epitaxial relation. As shown in Fig. 1(b), phi scans of the 50 nm VO_2 thin film at $2\theta = 27.75^\circ$ and $\psi = 45^\circ$ show six reflections corresponding to the (110) orientation of VO_2 . The presence of six peaks in the phi scans is due to the presence of three preferred orientations of M-VO_2 on sapphire, which means the VO_2 grains are rotated in-plane while showing a single out-of-plane orientation in $\theta - 2\theta$ scans. The three-fold symmetry of the (2204) Al_2O_3 reflection measured at $2\theta = 26.26^\circ$ and $\psi = 57.60^\circ$ is also shown in the same figure. For each sample, the (110) VO_2 reflections are rotated by 30° with respect to the (2204) Al_2O_3 reflections, demonstrating an epitaxial relation $[100](010)\text{VO}_2 \parallel [10\bar{1}0](0001)\text{Al}_2\text{O}_3$. We also carried out atomic force microscopy (AFM) to characterize the surface morphology of the films. Figure 1(c) shows an AFM image of the 50 nm-thick VO_2 film surface, indicating very smooth surface with root mean square (RMS) roughness of 1.05 nm. The XRD phi scans and AFM images of the 71.5 and 100 nm VO_2 thin films are not presented here, since they exhibit similar properties to those of the 50 nm film.

The IR transmission ($\lambda = 1520$ nm) of the VO_2 thin films was measured at normal incidence as a function of temperature in order to determine the temperature dependent optical properties of the thin films. The VO_2 sample was attached to a thermoelectric cooler (TEC) with a hole in the center [inset of Fig. 2(a)]. As shown in Fig. 2(a), an optical transmittance change up to 45% takes place during the IMT for all the VO_2 films investigated. Transmission is thickness-dependent only, with the thicker films transmitting less. The narrow hysteresis width of 1.5 K attests to the high structural quality of the VO_2 thin films. Four-point probe measurements at different temperatures were performed to characterize the resistivity changes of the films, as shown in Fig. 2(b). All the VO_2 thin films show quite similar behavior, exhibiting a sharp resistivity change of four orders of magnitude during the IMT. Characterization of the structural, optical, and electrical properties of these VO_2 thin films further attest to their good quality.

VO_2 is a low loss optical material and an outstanding candidate for tunable SPPs due to its phase dependent optical properties. For the present studies we have concentrated on the thermally induced IMT, but it is important to point out that other IMT excitation methods can also be used to trigger this transition [12,13], opening up the possibility of ultrafast or integrated devices depending on the choice of light or electric field excitation. In

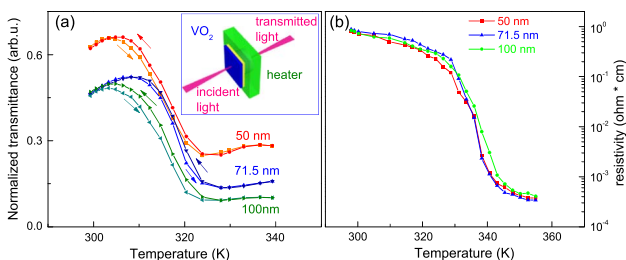


Fig. 2. (Color online) Temperature dependence of the IR transmittance at $\lambda = 1520$ nm (a) and resistivity (b) of three VO_2 thin films with different film thicknesses.

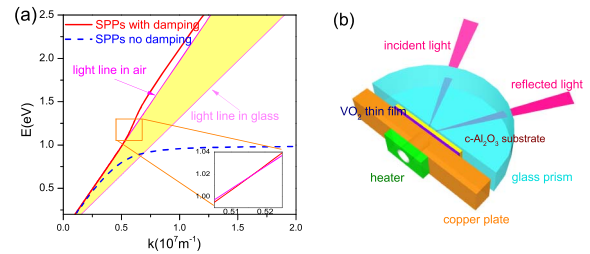


Fig. 3. (Color online) (a) Dispersion relation curves of SPPs at the VO_2 /air interface with (solid) and without (dash) damping. The shadowed region is bounded by the light lines in air and in glass. Dispersion relations are plotted versus the real part of the in-plane wave vector k . (b) Schematic of SPPs measurement setup using Kretschmann configuration combined with a heater.

the present case, when the film is in the metallic phase at higher temperatures, SPPs can be excited due to the presence of free electrons, which vanish below the transition temperature when the VO_2 is in its dielectric (insulating) state. In Fig. 3(a) we plot the calculated dispersion relation curves, a function of the incident energy E and the plasmon wave vector k for propagating SPP modes at the VO_2 /air interface in the metallic state. The dielectric constants used in the calculations were obtained from reference [14]. Leaky SPPs can be excited within the shadowed region between the light lines in air and glass, which in the Kretschmann configuration [as shown in Fig. 3(b)] corresponds to the angular region $[\theta_c, 90^\circ]$ (θ_c is the total internal reflection angle or critical angle). When considering the optical absorption of VO_2 , SPP modes are allowed between the light lines of air and glass for incident energy $E < 1$ eV, which means that the SPPs can be excited in the IR region ($\lambda > 1240$ nm).

The optical response of VO_2 thin films under SPP excitation was investigated in the Kretschmann configuration [15], in which the VO_2 sample was attached to a semicylindrical glass prism ($n = 1.5018$) using refractive index-matching oil ($n = 1.5018$), as shown in Fig. 3(b). The VO_2 sample was attached to a TEC to carry out the temperature-dependent measurements. An automated high-resolution goniometer allowed measurement using variable incident angles. The VO_2 thin films were illuminated using p -polarized He-Ne IR laser (1520 nm), and the reflected intensity was detected using a Ge photodetector preceded by a p -oriented polarizer. The optical response under SPP excitation at a different temperature was investigated in depth on a 50 nm VO_2 thin film. As discussed above, a strong excitation of SPPs is expected

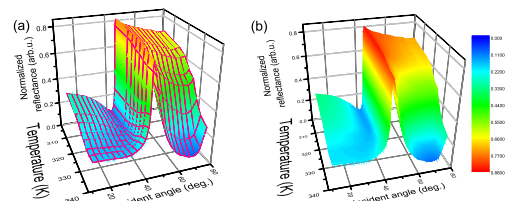


Fig. 4. (Color online) Experimental (a) and simulated (b) reflectance of a 50 nm VO_2 thin film illuminated in the IR region as a function of temperature and incident angle. A high absorption is observed at high temperature due to SPP excitation in the metallic state.

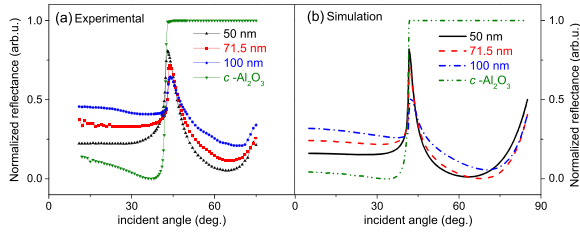


Fig. 5. (Color online) Experimental (a) and simulated (b) angular dependence of the reflectance for VO_2 thin films with thicknesses ranging from 50 to 100 nm. Experimental reflectance and simulation results of $c\text{-Al}_2\text{O}_3$ substrate are also included as comparisons. The absorption due to SPPs of thinner films (50 and 71.5 nm) is stronger than that of thicker film (100 nm).

in the IR region. Figure 4(a) shows the normalized reflectance as a function of temperature and incident angle in the IR region. It shows a clear trend from nonabsorption in the insulator phase to a high absorption in the metallic phase due to SPP excitation in the latter state. The absorption changed from around 10% to almost 100% across the IMT after the critical angle. This absorption trend correlates with the trend of the VO_2 metallic phase growth. We carried out simulations using matrix transfer formalism [16] and the previously obtained temperature dependent optical constants [14], as shown in Fig. 4(b). Simulations agree very well with the experimental results, showing low absorption in the insulator state (low temperature), and an extremely high absorption in the metallic state (high temperature) due to the SPP excitation. These results demonstrate the high potential of VO_2 as a tunable plasmon material where it is worthwhile considering that it is also possible to tailor the rate of this transition by adequate choice of the excitation process, i.e., optical, electrical, or thermal depending on the ultimate application goal.

We also investigated the effect of the thin film thickness on the SPP excitation. Figure 5 shows the angular dependence of the reflectance measurements of three VO_2 thin films with thicknesses of 50, 71.5, and 100 nm. The experimental and simulated reflectance for the $c\text{-Al}_2\text{O}_3$ substrate alone are also shown for comparison. The measurements were performed in the VO_2 metallic state, around 350 K. As shown in Fig. 5, the experimental results [Fig. 5(a)] agree very well with the simulations [Fig. 5(b)]. The SPP excitation strongly depends on the film thicknesses, where the thinner films (50 and 71.5 nm) show higher absorption due to stronger SPP excitation, while for thicker film (100 nm) less light reach the VO_2 /air surface to excite SPPs, so the light is reflected after undergoing a weak absorption. Besides, there is an optimum thin film thickness that allows optimum coupling, i.e., a zero in the reflectance after the critical angle. Apparently it is close to 50 nm for

VO_2 . The width of SPP excitation is not as narrow as noble metals, but comparable to other conducting metal oxide, like ITO [17].

In conclusion, we have studied the SPP excitation in VO_2 as a potential tunable low-loss plasmonic material. We found that the SPPs could be excited in the conducting or metallic state in the IR region. For the present case, the absorption intensity due to SPPs increases as temperature goes up. We find that the SPP excitation highly depends on film thickness. Our findings have broader impact, since the results of these experiments demonstrate the possibility of active control as well as the ability to tailor the properties of optoplasmonic devices for different applications and technologies.

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