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Phonon polaritons enhance near-field thermal transfer across the phase transition of VO_2

P. J. van Zwol,[1,*] K. Joulain,[2] P. Ben-Abdallah,[3] and J. Chevrier[1]

[1]*Institut Néel, CNRS and Université Joseph Fourier Grenoble, BP 166, F-38042 Grenoble Cedex 9, France*

[2]*Institut P', CNRS-Université de Poitiers-CNRS UPR 3346, F-86022 Poitiers Cedex, France*

[3]*Laboratoire Charles Fabry, Institut d'Optique, CNRS, UMR 8501, Université Paris-Sud 11, 2, Avenue Augustin Fresnel, F-91127 Palaiseau Cedex, France*

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We show numerically that near-field heat flux can be modulated by orders of magnitude upon switching from the metallic to the insulating phase of vanadium dioxide. Furthermore, the resonant phonon polariton interaction for the insulating phase enhances near-field thermal transfer by three orders of magnitude. The effect should therefore be measurable with existing experimental setups and could find broad applications for systems where thermal control at the nanoscale is required.

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Discovered in 1959,[1] the phase transition of VO_2 at $T_c = 340$ K is nowadays a prototype of a metal-insulator transition (MIT) associated with a structural transition. The exact nature of this phase transition remains under scrutiny as it shows signs of both Peierls and Mott mechanisms, and has been subject of a vast literature.[1–10] The material can be switched reversibly in as little as 100 fs from an insulating state with a monoclinic structure to a metallic state with a tetragonal (rutile) structure.[4] Besides the change in structure, the optical and electrical responses are deeply affected. As a consequence, VO_2 has emerged as an important material for applications in optical and electrical systems. It can be used as an ultrafast optical shutter, for example, as the phase transition can be triggered by optical or electrical means.[6] Reversible electrical switching has been shown to work for over 10^7 cycles without failure in microscopic devices.[7]

An important, and thus far mainly neglected, feature of VO_2 is the existence of phonon modes in the infrared that result in surface phonon polaritons at energies close to the maximum of the Planck blackbody spectrum at room temperature. The existence of these phonon modes, which emerge as soon as the temperature is decreased below the transition temperature, is the key to the work presented here, which concerns near-field thermal transfer modulation in vacuum.

In general, it is difficult to tune the transfer of heat, as is analogously done with electrical current in modern transistors, due to the nonexistence of perfect thermal insulators. Large thermal conductivity contrasts of larger than an order of magnitude do not exist for switchable materials in either solids or in the far field. Yet temperature control is of great importance in many areas in physics and chemistry. Pioneering work has led to heat modulators with low cooling power,[11] or as concepts.[12] They employ either solid-fluid mechanisms or very low temperatures. A first effort for rectification of thermal transfer in the near field by 40% was reported in Ref. 13.

Near-field thermal transfer is enhanced as compared to far-field thermal transfer that is governed by the well-known Stefan-Boltzmann law.[14] In this regard one of the most intriguing effects discovered recently is that surface phonon polaritons can serve as unique channels of heat flow that dramatically increase the heat flux between two surfaces.[14–23] We show here that important unique thermal properties of VO_2 emerge. The MIT of VO_2 entails basically a change in surface phonon-polariton states, which has profound consequences for near-field thermal transfer.

Previous attempts have already shown that, whereas thermal conductivity and far-field heat transfer are difficult to tune, in the near field the radiative heat flux can be vastly altered.[17,24] In Ref. 24 the thermal transfer between two surfaces was varied by using the MIT of antimony-indium-silver-tellurium (AIST) alloys, a material that is well known for its use in compact disks and dense memories.[25] The thermal transfer change upon MIT (to which we will simply refer to as contrast) found in the near field was an order of magnitude larger than that for the thermal conductivity in the solid. While both AIST and VO_2 undergo a MIT, and have comparable conductivities for both their phases, we show that the surface phonon polaritons, which only exist for VO_2, lead to very large differences in thermal transfer as compared to AIST.

The response of a material to (thermal) photons in the IR is described by its dielectric function. While the dielectric behavior of monoclinic insulating VO_2 shows rich phonon absorption, rutile metallic VO_2 only exhibits Drude absorption without any observable phonon contributions [Fig. 1(c)]. This dielectric data is needed for numerical calculations which are not much different from those used in Ref. 24. However, monoclinic insulating VO_2 has an anisotropic dielectric response in the IR range, while the rutile metallic phase of VO_2 does not exhibit dielectric anisotropy. Anisotropic dielectric data for the two crystal orientations of ($\perp$ and $\parallel$ to the a_m unit vector) can be found in Refs. 3 and 8 [Figs. 1(a) and 1(b)]. There exists also ellipsometric data for a (200)-oriented thin film reported in Ref. 4. This data is not anisotropic because for thin films such as the one in Ref. 4, the a_m [Figs. 1(a) and 1(b)] vector points out of the surface, thus VO_2 thin films are a uniaxial medium with an optical axis orthogonal to the surface, and are thus isotropic in plane.

We employ standard stochastic electrodynamics[14] to calculate the heat flux exchanged between the two bodies. Accordingly, the flux per unit surface is given by the statistical average of the Poynting vector normal component S_z,

$$\langle S_z \rangle = \int_0^\infty \frac{d\omega}{2\pi}[\Theta(w,T_1) - \Theta(w,T_2)] \int \frac{dk_\|}{(2\pi)^2} T(\omega,k_\|), \tag{1}$$

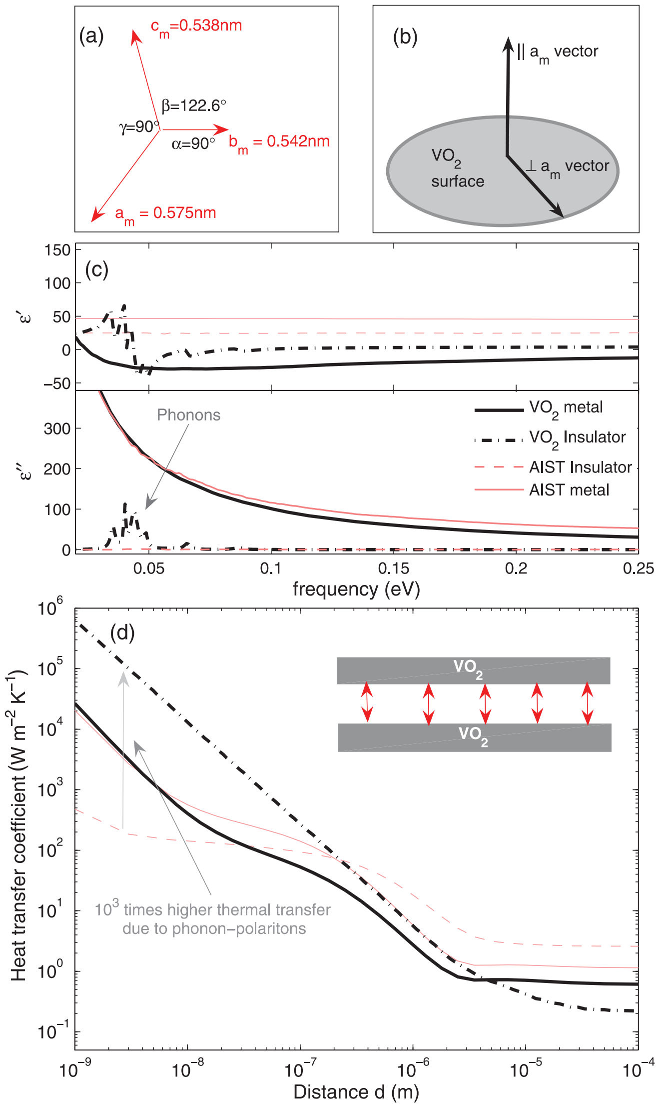


FIG. 1. (Color online) (a) The unit-cell dimensions of VO_2 as obtained from Ref. 8. While VO_2 has a monoclinic structure, only two distinct phonon spectra were found in Ref. 3. (b) It follows that thin films with (200) orientation (the circle) can be modeled as a uniaxial material. (c) Real and imaginary part of the dielectric functions for AIST and VO_2 as obtained by ellipsometry on deposited thin films from Refs. 3,4, and 24. For clarity, only the phonon state parallel to the a_m vector for insulating VO_2 is shown. (d) shows heat transfer between parallel plates of VO_2-VO_2, and AIST-AIST (Refs. 24 and 26). For (c) and (d) the legend of (c) applies.

where $\Theta(\omega,T) = \hbar\omega/[\exp(\hbar\omega/k_bT) - 1]$ is the mean energy of a Planck oscillator at frequency ω in thermal equilibrium where T_1 and T_2 are the temperatures of the two interacting surfaces, and $T(\omega,k_\parallel)$ is the energy transmission coefficient which can be written in terms of reflection operators of interacting surfaces.[18] Equation (1) is used in Ref. 18, and its application to anisotropic systems is well documented there. The methods employed here are not different from those employed in Ref. 18. We performed calculations using both the anisotropic theory with the data of Barker, as well as the theory for isotropic systems[24] using the data of Refs. 3 and 4. All calculations are done for parallel plates at approximately room temperature.

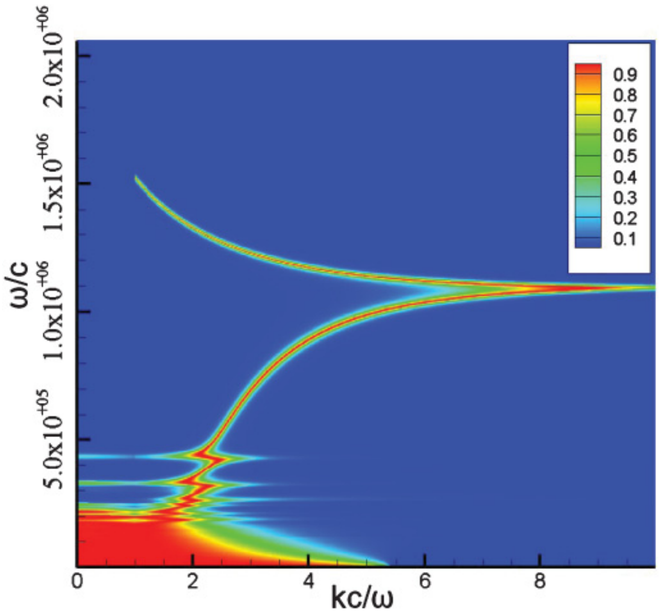
Figure 1(c) shows the dielectric functions for both AIST and VO_2 films and Fig. 1(d) shows near-field heat transfer results for both VO_2-VO_2 and AIST-AIST.[26] The dielectric responses for AIST and VO_2 in both states are rather similar over a large frequency range. Both are semimetals with very similar conductivity in the metallic case, and both are dielectrics in the insulating case. The main difference is that VO_2 supports surface phonon polaritons in its insulating state, whereas AIST does not, being a dielectric without apparent features in the IR range.

The thermal transfer for the metallic phase is very similar for both materials, as is expected from their highly similar semimetallic dielectric responses. Contrarily, the surface phonon polaritons enhance the heat transfer for the insulating phase of VO_2 by over three orders of magnitude as compared to insulating AIST [Fig. 1(d)]. The highest radiative heat transfer contrast for VO_2 is observable for distances below 10 nm, where thermal transfer approaches 100 W cm^{-2} K^{-1}. Both materials have similar contrasts in the extreme near field, being approximately a factor of 40, but the net thermal transfer for VO_2 is almost two orders of magnitude higher.

In order to understand the physics involved in these mechanisms, we have plotted in Fig. 2 the p-polarized part of the transmission coefficients $T(\omega,k_\parallel)$ [Eq. (1)] for both states of VO_2 at a given separation distance. For insulating VO_2 we see mainly a large magnitude of $T(\omega,k_\parallel)$ in the region of coupled surface phonon polaritons. The mode coupling is rather efficient in this region and responsible for a large heat transfer as $T(\omega,k_\parallel)$ is close to one over a large $k_\parallel$ range.[19] Note that there are discrete extraordinary surface modes at lower frequencies when $\varepsilon_\parallel/\varepsilon_\perp = 1$. Frustrated modes are also observed, and they are due to total internal reflection as propagating waves inside VO_2 become evanescent within the gap. Such modes give a non-negligible contribution to the heat transfer; more details on the physics thereof can be found in Ref. 18. On the other hand, for metallic VO_2 only a thin region at approximately $k = w/c$ over a broad spectral range contributes to the transfer. It corresponds to a spectral region where the real part of the permittivity is negative. Thus where in the near field AIST transfers heat in a broadband way for both insulating and metallic states, VO_2 can change from a metallic broadband emitter to one that emits strongly only at frequencies corresponding to interacting phonon polaritons.

The heat transfer contrast, being a factor of 40 for VO_2 in the near field, is much larger than the thermal conductivity contrast inside the material which increases from 3.5 to 5.5 W m^{-1} K^{-1} upon MIT.[5] The same is found when comparing to the change in far-field emissivity upon MIT, which is a factor of 2 [Fig. 1(d)]. Besides that, in the near field the heat transfer has increased by five orders of magnitude. This makes VO_2 a very interesting material for thermal management in nanosystems. To put things in perspective, 100 W cm^{-2} is the heat generated in modern computer chips, or the power emitted from a blackbody at 2000 K.

Unlike AIST in Ref. 24, VO_2 can also be switched when its dimensions are larger than a few hundreds of nanometers. This means that measuring the effect of phase transition should be much more straightforward. Glass spheres are currently the



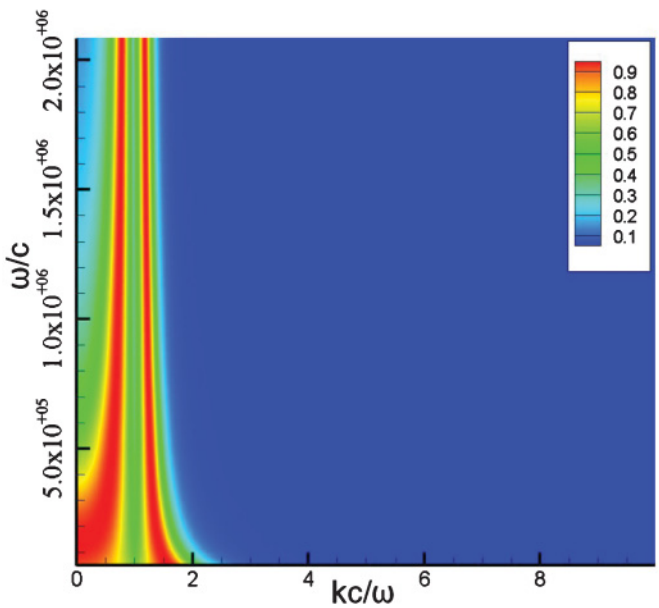


FIG. 2. (Color online) Top: Transmission coefficient (p polarized) between two insulating VO_2 surfaces. Bottom: The same for two metallic VO_2 surfaces. Both graphs were obtained at a separation distance between the surfaces of 500 nm. The spectral character of thermal transfer completely changes across the MIT.

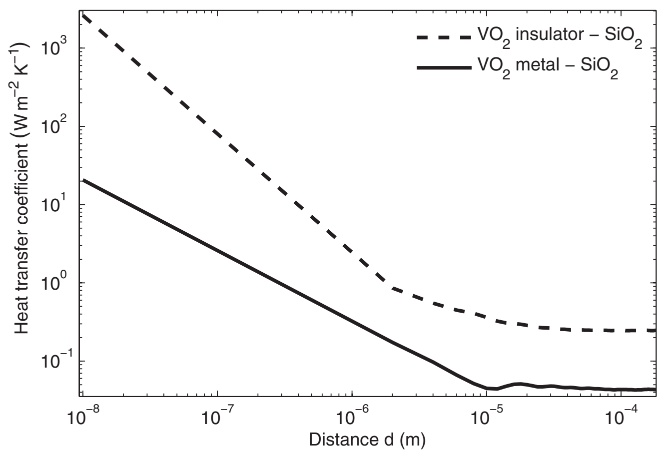


FIG. 3. Heat transfer between 100 nm VO_2 and a glass surface. The heat transfer change upon MIT is very large.

most used probes to accurately measure thermal transfer.[22] Both SiO_2 and VO_2 support surface phonon polaritons with similar frequencies, and thus one can expect the near-field interactions to be strong.

Near-field thermal transfer calculations for VO_2-SiO_2 are shown in Fig. 3, where it is seen that the magnitude of the net heat transfer is still large and well in the range of existing experimental setups. Moreover, we found a very large thermal contrast that is a factor of 5 in the far field and more than two orders of magnitude in the near field. Such a large effect should be well measurable. It may also find its way in applications, as heat transfer can effectively be switched from almost nothing to a large value, much as in electronic transistors.

Concerning measurements, for a plate-sphere setup the contrast is reduced. In this case lower-contrast far-field contributions are inevitably present. However, this should not be a problem since by definition experimental setups that probe the behavior of heat transfer with distance are only sensitive to near-field transfer, because the far-field transfer remains constant with distance. It therefore suffices to change the borosilicate sample used in Ref. 22 by a VO_2 sample and repeat the measurement. Finally (while this is not shown in Fig. 3), when the glass sphere is replaced by a gold sphere, the opposite behavior is observed, but the net thermal transfer is decreased by more than an order of magnitude, so this may be difficult to observe.

As far as possible applications are concerned, we note that the transition temperature of VO_2 may be lowered by doping VO_2 with tungsten.[9] Besides the well-known phase change materials[25] and VO_2, other vanadium oxides also undergo a metal-insulator transition, but at much higher (V_3O_5 at 428 K) or lower (V_2O_3 at ∼170 K) temperatures. These compounds also show strong phonon absorption in their insulating states[10] which may support surface modes. Manganite superlattices also exhibit phonon absorption in their insulating state combined with a metal-insulator transition.[27] Thus the phonon-polariton-enhanced near-field heat conductivity contrast may not be unique for VO_2, which implies also that the possibility to engineer the switching conditions and/or temperatures of the materials exists. We note that many transition-metal oxides can undergo a MIT, and some of them work as superconductors. Thus it might be possible to further increase the near-field heat conductivity contrast, with unique engineered switchable materials.

Concluding, by employing the metal-to-insulator transition, we have shown that tunable phonon polaritons, which exist on surfaces of insulating VO_2, can be used to modulate near-field thermal transfer from a high throughput narrowband state to a low throughput broadband state. The tuning of polaritons does not only enhance near-field transfer by orders of magnitude, but also results in a large near-field thermal conductivity contrast upon MIT. Thus the described effect should be well measurable with current experimental setups. Our finding may have implications for the control and transport of heat. The MIT can be used to drastically change heat exchanges in the near field. This may yield a powerful tool for thermal management at the nanoscale.

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*petervanzwol@gmail.com

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