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# Phonon-Polaritons enhance near field thermal transfer across the phase transition of $VO_2$

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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 3 orders of magnitude. The effect should therefore be measureable 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 Tc= 340K 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 femtoseconds from an insulating state with monoclinic structure to a metallic state with 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, 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, that 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 non-existence of perfect thermal insulators. Nor in solids, nor in far field, do large thermal conductivity contrasts of larger than an order of magnitude exist for switchable materials. Yet the control of temperature 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-fluids mechanisms or very low temperatures. A first effort of rectification of thermal transfer in near field by 40% was reported in ref [13].

Near field thermal transfer is enhanced as compared to farfield thermal transfer that is governed by the well known Stefan-Boltzman law [14]. In this regard one of the most intriguing effects discovered recently is that surface phonon-polaritons can serve as new channels of heat flow that dramatically increase the heat flux between two surfaces [14-23]. We show that precisely here, important new thermal properties of $VO_2$ emerge. The MIT of $VO_2$ entails basically a change of 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 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 discs and dense memories [25]. The thermal transfer change upon MIT (to which we will simply refer to as contrast) found in 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 AIST.

The response of a material to (thermal) photons in the IR is described by its dielectric function. While the dielectric behaviour of monoclinic insulating $VO_2$ shows rich phonon absorption, rutile metallic $VO_2$ only exhibits Drude absorption without any observable phonon contributions (Fig 1c,d). 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 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 2 crystal orientations of ($\perp$ and $\parallel$ to the $a_m$ unit vector) can be found in ref [3,8] (fig 1a,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 like the one in ref [4] the $a_m$ (fig. 1a,b) vector points out of the surface, thus VO$_2$ thin films are a uniaxial medium with 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 $Sz$

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

where $\Theta(\omega, T) = \hbar\omega/[\exp(\hbar\omega/k_bT)-1]$ is the mean energy of a Planck oscillator at frequency $\omega$ in thermal equilibrium 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 [18]. We performed calculations using both the anisotropic theory using the data of Barker, as well as theory for isotropic systems [24] to the data of [3] and [4]. All calculations are done for parallel plates at around room temperature.

Figure 1c shows the dielectric functions for both AIST and VO$_2$ films and figure 1e shows near field heat transfer results for both VO$_2$-VO$_2$ and AIST-AIST [27]. The dielectric response 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 semi-metallic dielectric responses. Contrarily the surface

phonon polaritons enhance the heat transfer for the insulating phase of $VO_2$ by over 3 orders of magnitude as compared to insulating AIST (fig 1d). The highest radiative heat transfer contrast for $VO_2$ is observable for distances below 10nm, where thermal transfer approaches 100W $cm^{-2}$ $K^{-1}$. Both materials have similar contrasts in the extreme near field, being about a factor of 40, but the net thermal transfer for $VO_2$ is almost 2 orders of magnitude higher.

In order to understand the physics involved in these mechanisms we have plotted in Fig. 2 the transmission coefficients T (eq. 1) in the $(\omega,k_\parallel)$ plane of p-modes for both states of $VO_2$ at a given separation distance. For insulating $VO_2$ we see mainly a large magnitude of T in the region of coupled surface phonon polaritons. The mode coupling is rather efficient in this region and responsible for a large heat transfer (T is close to one over a large $k_\parallel$ range) [19]. There are also discrete extraordinary surface modes at lower frequencies when $\varepsilon_{//}\varepsilon_\perp=1$ as well as frustrated modes due to total internal reflection (propagating waves inside $VO_2$ become evanescent within the gap). Such modes give a non-negligible contribution to the heat transfer [18]. On the other hand, for metallic $VO_2$ only a thin region around the 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 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 near field contrast being a factor of 40 for $VO_2$ in 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 [18]. The same is found when comparing to the change in farfield emissivity upon MIT, which is a factor of 2 (fig. 1d). Besides that in near field the heat transfer has increased by 5 orders of magnitude. This makes $VO_2$ a very interesting material for thermal

management in nanosystems. To put things in perspective: $100 Wcm^{-2}$ is the heat generated in modern computer chips, or the power emitted from a blackbody at 2000K.

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

Near field thermal transfer calculations for $VO_2$-$SiO_2$ are shown in figure 3, where it is seen that the magnitude of the net heat transfer is still large and well in range of existing experimental setups. Moreover we found a very large thermal contrast that is a factor of 5 in farfield and more than two orders of magnitude in 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 like in electronic transistors.

Concerning measurements, for a plate-sphere setup the contrast is reduced. In this case lower contrast farfield contributions are inevitably present. This should however not be a problem since by definition experimental setups that probe the behaviour of heat transfer with distance are only sensitive to near field transfer, because the farfield 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 figure 3 when the glass sphere is replaced by a gold sphere the opposite behaviour 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 428K) or lower ($V_2O_3$ at ~170K) 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 [26]. Thus the phonon-polariton enhanced near field heat conductivity contrast may not be unique for $VO_2$, which implies also that there exists the possibility to engineer the switching conditions/temperatures of the materials. 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 near field heat conductivity contrast, with novel engineered switchable materials.

Concluding, by employing the metal to insulator transition we have shown that tuneable 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 result 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 control and transport of heat. The MIT can be used to drastically change heat exchanges in near field. This may yield a powerful new tool for thermal management at the nanoscale.

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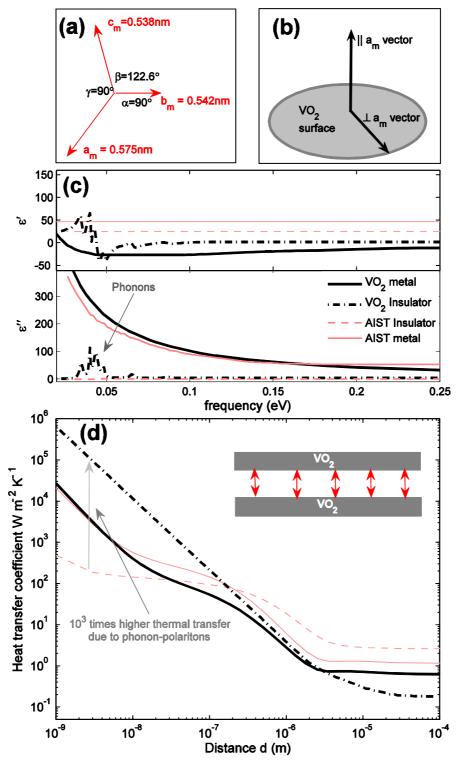
Figure 1 (Color online): (a) The unit cell dimensions of VO$_2$ as obtained from ref [8]. While VO$_2$ has monoclinic structure, only 2 distinct phonon spectra where found in ref [3]. (b) It follows that thin films with (200) orientation (the circle) can be modelled as a uniaxial material. (c) Real and Imaginary part of the dielectric functions for AIST and VO$_2$ as obtained by ellipsometry on deposited thin films from refs [3, 4, 24]. For clarity only the phonon state parallel to the $a_m$ vector for insulating VO$_2$ is shown. Plot (d) shows heat transfer between parallel plates of VO$_2$-VO$_2$, and AIST-AIST [24,27]). For all figures 1c,d the legend of fig. 1c applies.
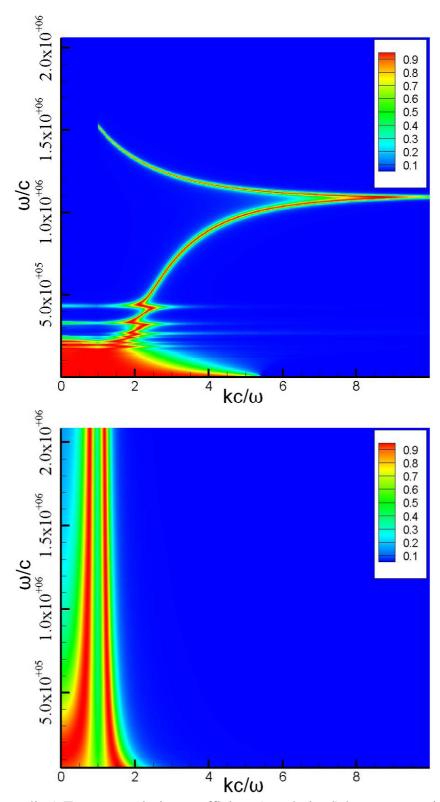
Fig 2: (Color online) Top, transmission coefficient (p polarized) between two insulating $VO_2$ surfaces. Bottom, same for two metallic $VO_2$ surfaces. Both graphs were obtained at a separation distance between the surfaces of 500nm. The spectral character of thermal transfer completely changes across the MIT.
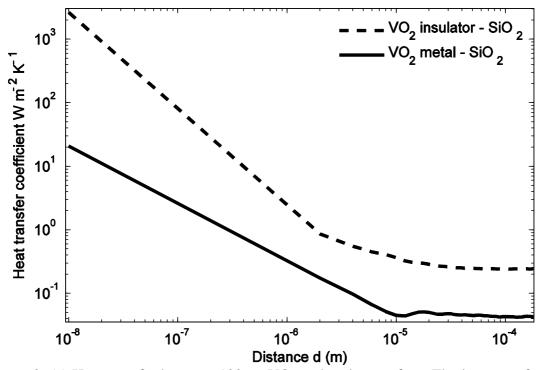
Figure 3: (a) Heat transfer between 100nm $VO_2$ and a glass surface. The heat transfer change upon MIT is very large.