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# Ultrafast photo-induced dynamics across the metal-insulator transition of VO$_2$

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**Abstract** – The transient reflectivity of VO$_2$ films across the metal-insulator transition clearly shows that with low-fluence excitation, when insulating domains are dominant, energy transfer from the optically excited electrons to the lattice is not instantaneous, but precedes the superheating-driven expansion of the metallic domains. This implies that the phase transition in the coexistence regime is lattice-, not electronically-driven, at weak laser excitation. The superheated phonons provide the latent heat required for the propagation of the optically-induced phase transition. For VO$_2$ this transition path is significantly different from what has been reported in the strong-excitation regime. We also observe a slow-down of the superheating-driven expansion of the metallic domains around the metal-insulator transition, which is possibly due to the competition among several co-existing phases, or an emergent critical-like behavior.

**Introduction.** – For decades, transition metal oxides have been subject of basic [1–3] and applied [4–8] research due to their unique electrical, magnetic, optical, thermal, mechanical, and chemical properties. An important example, vanadium dioxide (VO$_2$) [1,9], exhibits a first-order metal-insulator transition (MIT) with a 4–5 orders of magnitude change in resistivity, accompanied by a structural phase transition (SPT) from a low-temperature monoclinic structure to a high-temperature rutile structure. In VO$_2$ thin films the MIT exhibits phase coexistence [10] and occurs through multiple resistance jumps which are ascribed to avalanches [11]. The transition can be induced by heating [9,12], current [13,14], electric field [15–17], and laser excitation [18,19], and its origin remains the subject of much debate [14,20–25].

Ultrafast optical pump-probe spectroscopy was extensively used to study the dynamics of the MIT and SPT of VO$_2$, from sub-picosecond to nanosecond time scales [18,19,21,26–28]. Most ultrafast experiments were carried around room temperature well below the disorder-broadened metal-insulator transition temperature ($T_C$). These experiments show a clear fluence threshold [18,27], needed to overcome the latent heat associated with the first-order transition. At the high pump fluences which are necessary to induce the transition, clear evidence of the SPT appears in sub-picosecond time-resolved x-ray diffraction, electron diffraction, and coherent phonon studies [18,21]. There is still controversy involving the actual nature of this non-thermal pathway to the SPT, which may involve either a predominantly electronic mechanism (*i.e.*, carrier-induced perturbation of the lattice potential), or direct laser excitation of coherent lattice displacements. It is quite clear from other experiments that the initial seeding of metallic domains by the non-thermal mechanisms is followed by much slower, superheating-driven growth of metallic domains, at the expense of the

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insulating phase, on typical hundreds of picoseconds time scales [27]. This process is observed for a wide range above-threshold fluences and base temperatures.

Few experiments probed the effects of base temperature on the dynamics of the MIT. A decrease of the threshold fluence [27] together with a shortening of the initial, sub-picosecond non-thermal response [29] was observed. Surprisingly, a detailed study of the dynamics as the base temperature approaches $T_C$, over a broad time scale range (sub-picosecond to nanosecond) has not been carried out. Moreover, no measurements have been reported at low-fluence excitation at base temperatures across the MIT. This regime presents a totally different physical scenario, since the existence of precursor metallic domains in the coexistence regime negates the necessity of introducing metallic seeds by strong excitation, while also creating a different optical landscape with spatially-varying absorption. Significantly, the ability to excite the material in this regime with a lower-fluence pump has the potential to reveal effects, which are otherwise masked by the strong drive which is always necessary when exciting from a homogeneous, single-phase initial state at low base temperatures. This is truly an experiment in uncharted waters (at least in terms of ultrafast laser spectroscopy), since describing the sample as being in a coexistence regime of insulating and metallic phases is an oversimplification. As recently concluded from experiments in disorder-free $VO_2$ microcrystals, micro-beams and nano-beams, the MIT and SPT in thin, strained $VO_2$ films involve the coexistence of two insulating monoclinic phases, an insulating triclinic phase, the rutile metallic phase, and a broad range of transition temperatures [29–31]. A recent ultrafast electron diffraction experiment has revealed yet another possible monoclinic metallic phase during the optically induced MIT [32]. It is therefore not obvious how the inhomogeneity of the system, which apparently occurs over a wide range of length scales, might change the dynamics of the phase transition.

Here we report time-resolved optical pump-probe measurements over a *broad range of base temperatures and pump fluences*, with emphasis on low-fluence excitation in the *phase coexistence regime*. Our broad time range (sub-picosecond to sub-nanosecond) measurements reveal details of the optically-induced MIT and SPT which have not been reported before. We clearly show that at low-fluence when the optical response of the insulating domains is dominant, energy transfer from the optically-excited electronic degrees of freedom to the lattice (*i.e.*, the phonon bath) is not instantaneous. Yet it appears to be a condition for the superheating-driven expansion of the metallic domains. This implies that the SPT in this regime is lattice not electronically-driven (at least for sufficiently weak excitation), with the superheated phonon bath providing the latent heat required for the propagation of the SPT. This path to the SPT is significantly different from earlier reports in the strong-drive regime, where the ions and the lattice potential are driven hard enough to produce an instantaneous collapse to the rutile phase. Furthermore, we observe a slow-down of the superheating-driven expansion of the SPT of the statically-measured phase transition curve. This is possibly due to competition between the different insulating and metallic phases at the transition [33], or perhaps due to an emergent critical-like behavior [11] observed in second-order phase transitions [34].

**Experimental.** – Thin films $VO_2$ samples were prepared by reactive RF magnetron sputtering of a $1.5''$ $V_2O_3$ target ($>99.7\%$, ACI Alloys, Inc.) on an r-cut sapphire substrate. A mixture of ultrahigh-purity argon and oxygen was used for the sputtering. The total pressure during deposition was $4\,\mathrm{mTorr}$, and the oxygen partial pressure was optimized at $0.32\,\mathrm{mTorr}$ ($8\%$ of the total pressure). The substrate temperature during deposition was $600\,^\circ\mathrm{C}$, and the RF power was $100\,\mathrm{watts}$. The data presented here was obtained with a $130\,\mathrm{nm}$ thick sample, but similar results were obtained with other samples. Considering that the optical penetration depth of $VO_2$ is $\sim 7\,\mu\mathrm{m}$, absorption throughout the film thickness is expected to be homogeneous[1].

The pump-probe measurements used an amplified femtosecond Ti:sapphire laser system. Laser pulses were generated by a Ti:sapphire–based oscillator (Coherent Mira seed) and amplified by a multi-pass Ti:sapphire amplifier (Odin Quantronix). The $\sim 100\,\mathrm{fs}$ pulses were centered near $800\,\mathrm{nm}$, and had a pulse energy of up to $\sim 600\,\mu\mathrm{J}$, at a $1\,\mathrm{kHz}$ repetition rate, long enough to allow for complete sample relaxation between consecutive pump pulses. The weak probe signal was measured by a combination of mechanical chopping, lock-in detection, and computer averaging.

**Results.** – Figure 1(a) shows the thermal hysteresis of the static reflectivity ($R$-$T$) of the sample, normalized to 0 at $T = 320\,\mathrm{K}$ and to 1 at $T = 360\,\mathrm{K}$. The midpoint of the heating branch of the thermal hysteresis (red curve in fig. 1(a)) is at $T_C = 343\,\mathrm{K}$, and the width of the transition is $\sim 5\,\mathrm{K}$. Figure 1(b) shows, on logarithmic scale, time-dependent differential reflectivity curves, $\Delta R = R(t) - R(0)$, measured at a base temperature $T = 319\,\mathrm{K}$, with three pump fluences: $F = 9, 18$, and $36\,\mathrm{mJ/cm^2}$. The lowest fluence of $9\,\mathrm{mJ/cm^2}$ is slightly above the threshold, consistently with the base temperature and the high thermal coupling to the sapphire substrate [32]. The three curves show different behaviors in the first 10 ps, but are similar on longer time scales (20–200 ps). In all three cases the initial response is a fast increase of $\Delta R$, with a characteristic time $\tau_1 = 450\,\mathrm{fs}$. The evolution of $\Delta R$ is very different after that: well above the fluence threshold (at $F = 36\,\mathrm{mJ/cm^2}$) $\Delta R$ increases monotonically, while at lower fluences (18 and $9\,\mathrm{mJ/cm^2}$) $\Delta R$ decreases after reaching a maximum at $\tau_1$. This behavior

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[1]The relative permittivity is measured for a similar $VO_2$ film ($\varepsilon = \varepsilon_1 + \varepsilon_2 = 8.822 + 0.055i$), giving a penetration depth $\delta = \frac{\lambda}{2\sqrt{2}\pi(-\varepsilon_1 + (\varepsilon_1^2 + \varepsilon_2^2)^{\frac{1}{2}})^{\frac{1}{2}}} \approx 7\,\mu\mathrm{m}$, where $\lambda = 800\,\mathrm{nm}$.
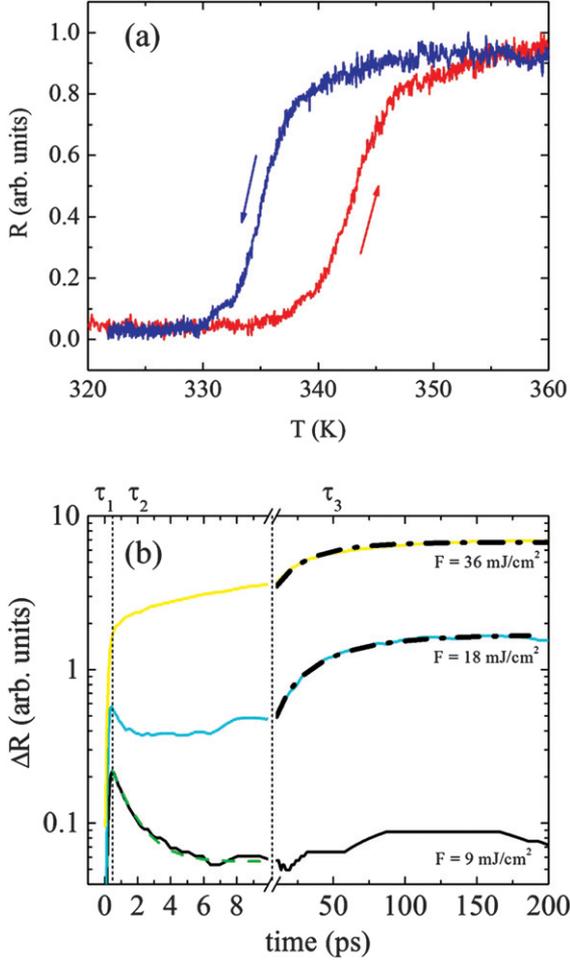
Fig. 2: (Colour online) The time dependence of $\Delta R$ for $F = 9\,\text{mJ/cm}^2$ at different base temperatures from $319\,\text{K}$ to $361\,\text{K}$. The data for $344\,\text{K}$ is not shown for clarity. The black dash-dotted lines are single exponential fits to the experimental data.

Fig. 1: (Colour online) (a) Static reflectivity *vs.* temperature ($R$-$T$) of a VO$_2$ film. The reflectivity at $T = 320\,\text{K}$ and $T = 360\,\text{K}$ is set to 0 and 1 (in arbitrary units), respectively. Red and blue curves correspond to the heating and cooling branches of the hysteresis, respectively. (b) Time dependence of $\Delta R$ (in logarithmic scale), for three different laser fluences ($F$) at a base temperature $T = 319\,\text{K}$: $F = 9\,\text{mJ/cm}^2$ (black curve), $F = 18\,\text{mJ/cm}^2$ (cyan curve) and $F = 36\,\text{mJ/cm}^2$ (yellow curve). The green dashed line is a fit that combines a single exponential decay, with a time constant of $\tau_2 = 1.3\,\text{ps}$, and a flat background. The black dash-dotted lines are single exponential fits with time constants of $\tau_3 = 43\,\text{ps}$ for $F = 18\,\text{mJ/cm}^2$ and $\tau_3 = 29\,\text{ps}$ for $F = 36\,\text{mJ/cm}^2$.

is consistent with previous measurements on disorder-free VO$_2$ samples [32], where it was assumed that a decrease of $\Delta R$ reflects sub-threshold excitation, with insufficient energy to induce the phase transition. Yet our measurements clearly show that even at low fluence (but above the threshold) $\Delta R$ recovers and starts increasing again (note that these details could not be discerned in previous measurements, *e.g.*, [27]). At $F = 18\,\text{mJ/cm}^2$ $\Delta R$ reaches a minimum at $t \sim 3\,\text{ps}$, and then shows a monotonous increase that on longer time scales (20–200 ps) appears similar to that observed for $F = 36\,\text{mJ/cm}^2$. For excitation slightly above threshold, $F = 9\,\text{mJ/cm}^2$, the decrease
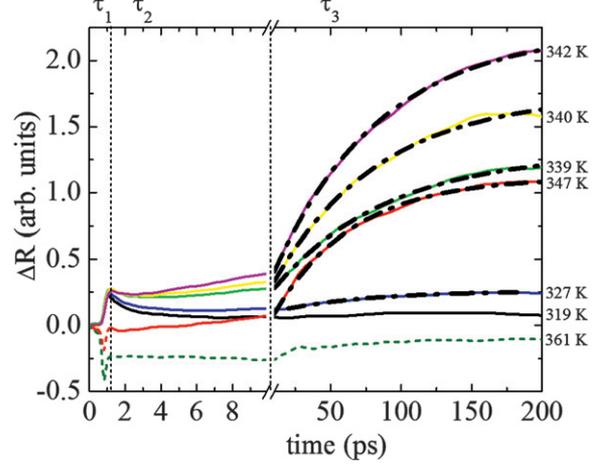
of $\Delta R$ after $\tau_1$ is much more pronounced. In this case $\Delta R$ reaches a minimum at $t \sim 10\,\text{ps}$, followed again by a gradual increase on longer (20–200 ps) time scales (note the logarithmic scale). The initial decrease of $\Delta R$ after $\tau_1$ in this measurement fits a single exponential decay with a time constant $\tau_2 = 1.3\,\text{ps}$ (indicated by a green dashed line in fig. 1(b))[2].

The gradual increase on longer time scales (20–200 ps) in all the measurements depicted in fig. 1(b) implies that for all three fluences a MIT is induced. Even at the lowest fluence ($F = 9\,\text{mJ/cm}^2$) $\Delta R$ is clearly positive at $t = 200\,\text{ps}$. Thus, even this low fluence is above the threshold required to induce the MIT at $T = 319\,\text{K}$ [27]. The long-time (20–200 ps) dependences of $\Delta R$ for $F = 18\,\text{mJ/cm}^2$ and $F = 36\,\text{mJ/cm}^2$ fit well a single-exponential growth (indicated by black dash-dotted lines in fig. 1(b)), with time constants (which we denote $\tau_3$) of 43 ps and 29 ps, respectively. The low signal-to-noise ratio prevents a reliable fit to the data for $F = 9\,\text{mJ/cm}^2$.

The effect of base temperature on the dynamics of the phase transition is quite striking. Figure 2 shows the time-dependent differential reflectivity curves measured, with the lowest laser fluence of fig. 1(b) ($F = 9\,\text{mJ/cm}^2$), at several base temperatures across the transition, from $T = 319\,\text{K}$ to $T = 360\,\text{K}$. The curves show two distinct responses as a function of the base temperature. In the temperature range between $319\,\text{K}$ and $342\,\text{K}$ (*i.e.*, below $T_C$) the response is similar to that shown (for the same fluence) in fig. 1(b), namely $\Delta R$ first increases rapidly, reaching a maximum at $\tau_1 = 450\,\text{fs}$, then slightly decreases, and finally settles on a steady increase that saturates at $t \sim 200\,\text{ps}$. However, as the base temperature approaches $T_C$, the reversal from decrease to increase

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[2]Note: We perform all the fittings with a general exponential function of the form $\Delta R = \Delta R_0 + \Delta R_1 e^{-t/\tau}$ within each temporal regime and obtained the corresponding characteristic time ($\tau_2$ and $\tau_3$).
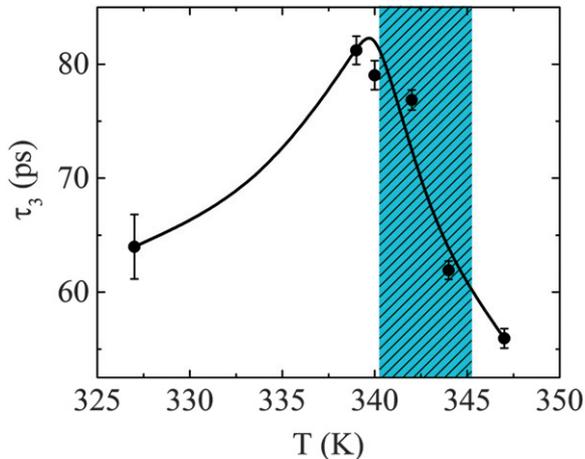
Fig. 3: (Colour online) The temperature dependence of the rise time $\tau_3$ obtained from the data in fig. 2. The error bars are the standard deviations from the fit to an exponential rise. The shaded area indicates the MIT temperature range.

occurs much earlier. The magnitude of the rise of $\Delta R$ at longer times (20–200 ps), associated with the growth of metallic domains, increases dramatically from $T = 319\,\mathrm{K}$ to $T = 342\,\mathrm{K}$, until, at $T = 342\,\mathrm{K}$, becomes similar to the response shown in fig. 1(b) for $T = 319\,\mathrm{K}$ and $F = 18\,\mathrm{mJ/cm^2}$. This is consistent with the idea that less energy is necessary to induce the MIT when the base temperature is higher.

A significant change is observed in the transient reflectivity measurements at $T = 347\,\mathrm{K}$ and $T = 361\,\mathrm{K}$ (phase coexistence with predominantly metallic phase). The initial response is now a *decrease* to a (negative) minimum, followed by partial recovery, all within $\tau_1$, in agreement with ref. [35]. The long-time response at $T = 347\,\mathrm{K}$ is qualitatively similar to that at $T = 342\,\mathrm{K}$, but weaker. The negative initial response and the decrease of the magnitude of the long-time response are clear signatures of the prevalence of the metallic phase [36,37].

The time constants $\tau_3$ obtained from single exponential fits to the long-time rise of $\Delta R$ (indicated by black dash-dotted lines in fig. 2) are plotted in fig. 3 as a function of the base temperature. To make sure that $\tau_3$ only reflects the dynamics of the MIT, we subtracted the contribution of the metallic phase from the experimental data. For this we used $\Delta R(t)$ at 361 K, and factored in the fraction of the metallic phase for each temperature, which we deduced from the normalized static reflectivity shown in fig. 1(a). Note that while the total absorption of the sample decreases with increasing base temperature, the deposited energy *density* in the insulating domains is expected to remain unchanged, and, therefore, the observed behavior of $\tau_3$ must reflect an intrinsic property of the material. The measurements in ref. [27], showed that $\tau_3$ is a constant below 320 K at low base temperatures. On the other hand, closer to the transition our fig. 3 shows that $\tau_3$ varies across the MIT temperature range, with the longest $\tau_3$ (*i.e.*, slowest response) observed around $T_C$.

**Discussion.** – The fast initial $(t < \tau_1)$ $\Delta R$ increase that we observe with low excitation fluence at base temperatures below $T = 347\,\mathrm{K}$ must be attributed to the electronic response of the insulating $VO_2$ ("photo-doping" [21]), *i.e.*, the excitation of hot electrons from the valence band to the conduction band of insulating $VO_2$ (the initial *negative* response of the metallic phase only appears at higher base temperatures). The initial, non-equilibrium distribution of photo-excited electrons reaches an internal equilibrium within $\tau_1$, slightly longer than the laser pulse-width, with an effective temperature which is significantly higher than the phonon temperature. The slow $\Delta R$ decay that follows $\tau_1$, at low fluences, implies that the effective electron temperature decreases as the electron system transfer energy to the lattice via inelastic electron-phonon scattering, with a characteristic time $\tau_2 \sim 1\,\mathrm{ps}$ (electron-phonon thermalization). While this type of response has been observed many times in pump-probe experiments on semiconductors [38,39] and metals [40,41], this is the first time that it is observed in $VO_2$ (see footnote [3]).

Only the much slower rise of $\Delta R$ on a time scale $\tau_3 \gg \tau_2$ reflects the MIT in $VO_2$, namely the appearance of a highly-reflecting metallic phase. With the phonon bath now at an effective (non-equilibrium) temperature which is higher than $T_C$, the MIT and SPT are driven by super-heating, in agreement with the interpretation in ref. [27]. Our data shows that, in contrast with what is implied by earlier experiments (including ref. [27]), this process starts instantaneously only in the limit of high-fluence excitation (see footnote [3]). It is in fact remarkable that the electron-phonon thermalization process is observed in our experiment even deep into the coexistence regime, when the material is already seeded with metallic domain. This is a strong indication that the MIT and SPT are driven by the superheated phonons and not by electronic excitation, at least in the weak excitation limit (when the hot-electron density is insufficient to modify the lattice potential). This path to the SPT is significantly different from what has been reported in the strong-drive regime, where the ions and the lattice potential are driven so hard that a large part of the crystal instantaneously collapses to the rutile phase [18,21,29,42,43], although the retarded response in the latter case must also be due to the superheated phonons.

The time scale $\tau_3$ is determined by the propagation speed of the MIT and SPT, which reflects the motion of the interface between the insulating and metallic phases [33,44,45]. The temperature dependence of $\tau_3$ across the MIT, which is determined here for the first time, shows a slow-down of the superheating-driven expansion of the MIT and SPT on the low-temperature side of the statically-measured phase transition curve. This finding is very surprising, since one would expect the

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[3]Note that the $\tau_2$ decay was not reported in ref. [26], possibly due to the low measurement resolution when the THz conductivity is low.

pre-existing, thermally-excited metallic domain to accelerate the propagation speed of the MIT and SPT. At this point we can only speculate that the observed slowdown is possibly due to competition between the different insulating and metallic phases in the reported "triple point" of the transition [30], or perhaps due to an emergent critical-like behavior [11,34]. It is clear, though, that as the abundance of metallic domains grows, the expansion rate of the metallic phase increases, as evidenced by the decrease of $\tau_3$ on the high-temperature side of the transition.

**Conclusions.** – Our temperature-dependent transient reflectivity measurements reveal details of the optically-induced MIT and SPT in VO$_2$ which have not been reported before. They clearly show that with low-fluence excitation, when the optical response of the insulating domains is dominant, energy transfer from the optically-excited electronic degrees of freedom to the lattice (*i.e.*, the phonon bath) is not instantaneous. Yet the transfer of heat to the lattice appears to be a condition for the superheating-driven expansion of the metallic domains. This implies that the SPT in this regime is lattice-driven and not electronically-driven (at least for sufficiently weak excitation), with the superheated-phonon bath providing the latent heat required for the propagation of the SPT. This path to the SPT is significantly different from what has been reported in the strong-drive regime, where the ions and the lattice potential are driven hard enough that a large part of the crystal instantaneously collapses to the rutile phase. Furthermore, we observe a slowdown of the superheating-driven expansion of the SPT on the low-temperature side of the statically-measured phase transition curve. This is possibly due to a competition among the different intermediate phases close to the transition [30], or perhaps due to an emergent critical-like behavior [11,34].

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