Correlated metallic state of vanadium dioxide

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The metal-insulator transition and unconventional metallic transport in vanadium dioxide (VO_2) are investigated with a combination of spectroscopic ellipsometry and reflectance measurements. The data indicate that electronic correlations, not electron-phonon interactions, govern charge dynamics in the metallic state of VO_2. This study focuses on the frequency and temperature dependence of the conductivity in the regime of extremely short mean free path violating the Ioffe-Regel-Mott limit of metallic transport. The standard quasiparticle picture of charge conduction is found to be untenable in metallic VO_2.

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I. INTRODUCTION

Transition-metal oxides constitute outstanding systems to explore strong correlations in solids. A rich variety of phase transitions and exotic electronic and magnetic ground states in this class of materials are at the focus of condensed-matter physics.[1] One system that has received considerable attention is vanadium dioxide (VO_2) that undergoes a first-order metal-insulator transition (MIT) at $T_c \approx 340$ K between a high-temperature rutile metallic state and a low-temperature insulating state. The renewed interest in this particular oxide is, in part, due to unresolved roles of the structural transformation and of Coulomb interactions in the MIT.[2–7] A better understanding of the metallic state is required to elucidate the nature of the MIT.

Yet another remarkable property of VO_2 pertains to the unconventional behavior of the resistivity for $T > T_c$.[8] The electronic mean free path inferred from the resistivity data was nearly the same as the lattice constant, yet resistivity showed linear T dependence without signs of saturation. In typical metals, the resistivity increases with increasing temperature but begins to saturate when the mean free path (l) becomes comparable to the lattice constant (d).[9] Ioffe and Regel, and subsequently Mott, have expressed the view that the electron mean free path cannot be less than the lattice constant, and $l \simeq d$ is commonly referred to as the Ioffe-Regel-Mott (IRM) limit.[10–12] However, in certain exotic metals, including the metallic phase of VO_2 and the high-T_c superconductors, the resistivity continues to increase beyond the IRM limit.[13,14] A complete understanding of this behavior in these so-called "bad metals"[15] is lacking. More importantly, when $l \lesssim d$, the conventional picture of a quasiparticle with a well-defined momentum undergoing occasional scattering events breaks down. Such a regime can no longer be described by the Boltzmann equation, signalling a breakdown of Fermi-liquid theory.[9] Fermi-liquid theory has been remarkably successful in describing the electronic properties of typical metallic systems. Understanding the deviations from this theory in exotic metals is a central theme of modern research.[1]

Significant progress has been made recently in the understanding of resistivity in the regime of short electronic mean free path.[13,14,16] Gunnarsson *et al.*, in particular, have supplemented early qualitative discussion with numerical work, including the role of correlations.[13] They devised a new criterion for resistivity saturation in correlated metals taking advantage of optical sum rules. The new criterion predicts saturation resistivities at values exceeding the IRM limit and this is attributed to the reduction of electronic kinetic energy due to correlation effects. Thus motivated, we have investigated the metallic phase of VO_2 with infrared and optical spectroscopy over a wide frequency range and up to high temperatures. Surprisingly, the optical conductivity shows typical metallic behavior beyond the IRM limit with the Drude peak centered at zero frequency persisting at high temperatures. In this work, we discover that beyond the IRM limit, the linear increase of resistivity of VO_2 with temperature is due to the linear increase of scattering rate of charge carriers. We show that although VO_2 violates the IRM criterion, the data are consistent with a higher value of saturation resistivity based on Gunnarsson *et al.*'s. model.[13] We conclude that electronic correlations, not electron-phonon interactions, dominate charge dynamics in the high temperature metallic phase of VO_2 and that the correlated charge carriers are not conventional Landau quasiparticles.

II. EXPERIMENT, RESULTS, AND DISCUSSION

VO_2 films of thickness ≈ 1000 Å were grown on Al_2O_3 (sapphire) substrates with the sol-gel method. X-ray diffraction shows that the films are single phase VO_2. The resistivity decreases by four orders of magnitude upon entering the metallic state above $T_c \approx 340$ K. The details of film growth and characterization are given in Ref. 17. The optical constants were obtained directly from a combination of ellipsometric measurements in the spectral range 50 meV–5.5 eV and near-normal incidence reflectance data in the spectral range 6–90 meV.[18]

In Fig. 1, we display the real part of the optical conductivity $\sigma_1(\omega)$ below and above T_c. VO_2 shows a band gap of ≈ 0.6 eV with a clear threshold in $\sigma_1(\omega)$ in the insulating state ($T = 295$ K). The band-gap value is in accord with previous data,[7,20,21] and the presence of a clear threshold confirms the good quality of sol-gel grown VO_2 films. Three distinct peaks at 1.4, 2.5, and 3.4 eV that arise from interband transitions can be discerned in the insulating state.

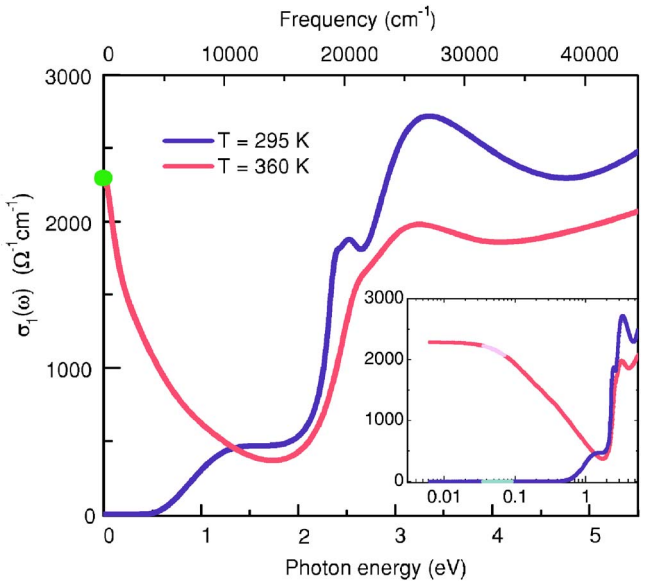

FIG. 1. (Color online) Real part of the optical conductivity $\sigma_1(\omega)$ of VO_2 in the insulating state (T=295 K) and metallic state (T=360 K). The solid circle refers to $\sigma_1(\omega\rightarrow 0)$=2300 Ω^{-1} cm^{-1}. Inset: the same data are shown on a logarithmic scale (in eV). Part of the data with phonons removed are depicted by lighter shades (Ref. 19).

There is significant rearrangement of the spectrum as VO_2 enters the metallic regime (T=360 K). A rather broad Drude-like peak emerges at low energies and signifies metallic behavior. There is a shift of spectral weight from the interband transitions at 2.5 and 3.4 eV to the Drude peak. The difference in the conductivity of the two phases extends beyond the upper cutoff of our measurements at 5.5 eV. Similar broad energy scales are involved in the redistribution of spectral weight between the insulating and metallic states in other correlated electron materials[1] including a close counterpart V_2O_3.[22] Therefore data in Fig. 1 indicate the presence of electronic correlations in VO_2.

A more quantitative approach to assess the importance of correlations is through their effect on the kinetic energy of the conduction electrons. If electron-electron interactions are important, then the measured kinetic energy is expected to be substantially reduced from the value given by band theory.[23] This has indeed been observed in both the hole-doped and electron-doped cuprates.[24] The optical conductivity provides us the means to obtain the expectation value of the kinetic energy of the conduction electrons in a solid through the partial sum rule:[13]

$$\frac{\omega_p^{\ 2}}{8} = \int_0^{\omega_c} \sigma_1(\omega) d\omega = -\frac{\pi d^2 e^2}{6N\Omega\hbar^2}\langle T_k\rangle. \qquad (1)$$

Here, ω_p is the plasma frequency, d is the lattice constant, Ω is the volume of a unit cell, N is the number of unit cells, and $\langle T_K\rangle$ is the expectation value of the kinetic-energy operator. Thus the expectation value of the kinetic energy is simply proportional to the square of the plasma frequency ω_p. We obtain the plasma frequency by integrating the area

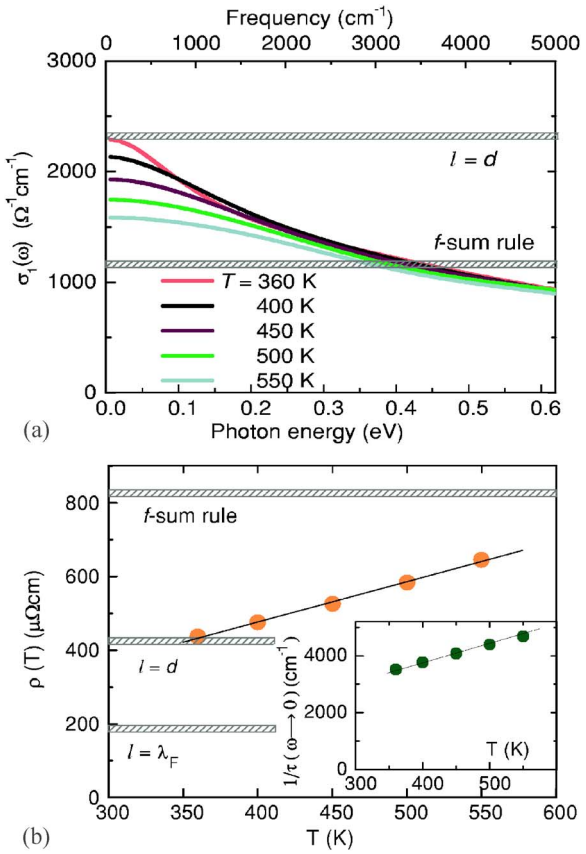

FIG. 2. (Color online) (a) Frequency dependence of $\sigma_1(\omega)$ of metallic VO_2 at different temperatures. Phonons have been removed in the frequency range 0.04–0.08 eV (Ref. 19). (b) Resistivity $\rho(T)$ of metallic VO_2, defined as $1/\sigma_1(\omega\rightarrow 0,T)$, is plotted as a function of temperature (circles). Inset: The scattering rate $1/\tau(\omega\rightarrow 0)$ is plotted as a function of temperature (circles). The solid lines in panel (b) and the inset are guides to the eye. The annotated horizontal lines in panels (a) and (b) depict dc conductivity and resistivity values at which saturation is expected according to the criteria mentioned in the text.

under the intraband Drude part of the conductivity up to a cutoff (ω_c) determined by the minimum in the conductivity at 1.7 eV and find ω_p=2.75 eV. The square of this value is only 50% of that obtained from band theory calculations.[8,25] This result is not particularly sensitive to the choice of the cutoff and confirms the importance of correlations in the conducting state of VO_2.

We investigate further the metallic phase of VO_2 at elevated temperatures. In Fig. 2(a), we display the frequency dependence of $\sigma_1(\omega)$ in the metallic phase of VO_2 for successively increasing temperatures. The conductivity decreases with increasing temperature as coherent spectral weight is transferred above 0.6 eV. However, the conductivity retains the Drude-like shape even at the highest temperature attained. In Fig. 2(b) we plot the "optical" resistivity $\rho(T)=1/\sigma_1(\omega\rightarrow 0,T)$. The resistivity continues to increase almost linearly with temperature up to the upper limit of our measurements at T=550 K without any sign of saturation. This is consistent with lack of saturation seen in dc resistivity measurements.[8]

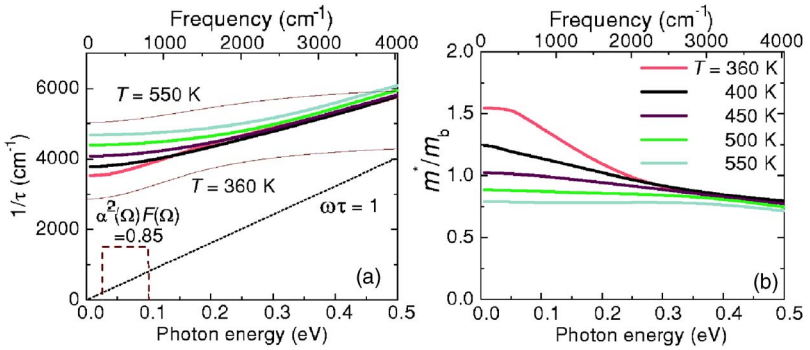

FIG. 3. (Color online) (a) The scattering rate $1/\tau$ and (b) the mass enhancement factor m^*/m_b in the metallic state of VO_2 are plotted as a function of frequency for different temperatures [labeled in panel (b)]. Extended Drude analysis was performed after phonons were removed from the conductivity in the frequency range 0.04–0.08 eV. In panel (a), the dotted line satisfies the equation $\omega\tau=1$, the dashed line is a model phonon spectral density $\alpha^2(\Omega)F(\Omega)$ (see text for details), and thin solid lines are the electron-phonon scattering rates at $T=360$ K and $T=550$ K calculated from $\alpha^2(\Omega)F(\Omega)$.

Next, we perform an extended Drude analysis to obtain the frequency and temperature dependence of the effective scattering rate $(1/\tau)$, and mass enhancement factor (m^*/m_b) of the charge carriers.[26,27] These are plotted in Figs. 3(a) and 3(b).

$$\frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi}\frac{\sigma_1(\omega)}{\sigma_1^2(\omega)+\sigma_2^2(\omega)}, \tag{2}$$

$$\frac{m^*(\omega)}{m_b} = \frac{\omega_p^2}{4\pi\omega}\frac{\sigma_2(\omega)}{\sigma_1^2(\omega)+\sigma_2^2(\omega)}. \tag{3}$$

In the above equations, $\sigma_2(\omega)$ is the imaginary part of the conductivity and m_b is the band mass of the charge carriers. We find that the scattering rate (in the zero-frequency limit) increases linearly with temperature and thereby leads to the corresponding increase of resistivity [see Fig. 2(b) and inset]. The absolute value of the scattering rate at all measured temperatures and infrared frequencies is higher than the excitation energy $(\hbar\omega)$. The canonical criterion for well-defined quasiparticles is that $1/\tau \ll \omega$.[9,26] The scattering rate in VO_2 certainly violates this criterion in common with the high-T_c cuprates.[26] Allen has defined the criterion for the validity of the Boltzmann equation and Fermi-liquid theory as $\hbar/\tau \ll E_F$ [or $N(0)\hbar/\tau \ll 1$] where E_F is the Fermi energy and $N(0)$ is the density of states at the Fermi level.[9] In Fig. 3(a), we see that $\hbar/\tau \approx 0.5$ eV which is comparable to $E_F \approx 0.5$ eV.[3] Alternatively, taking $N(0)=4.16/$ eV molecule,[2] we find that $N(0)\hbar/\tau$ is of order unity. Hence charge carriers in metallic VO_2 are not well-defined quasiparticles.

In Fig. 3(b) we see that the mass enhancement at $T=360$ K is a strong function of frequency. The frequency dependence of m^*/m_b becomes weaker at higher temperatures. Also, m^*/m_b in the low-frequency limit increases significantly with decreasing temperature, i.e., as one approaches the insulating state. Mass enhancement has been predicted in a strongly correlated electron gas in the vicinity of a transition to the Mott insulator.[28] In VO_2, the absolute value of m^*/m_b is modest at $T=360$ K consistent with recent photoemission results.[6] The increase in m^*/m_b with temperature on approaching the insulating state is, nevertheless, significant and provides evidence for correlations in VO_2. However, the mass enhancement is not as spectacular as seen with doping in $Sr_{1-x}La_xTiO_3$, a classic system that exhibits a mass-diverging-type Mott transition.[29] Several correlated systems are known to avoid mass divergence near the MIT.[1,30]

We now investigate the metallic behavior of VO_2 in light of the IRM criterion. At high temperatures, the resistivity of typical metals and alloys saturates at values which give mean free paths (l) of the quasiparticles that are comparable to the lattice spacing (d).[9] A complementary viewpoint is that resistivity saturates when l becomes comparable to the de Broglie wavelength λ_F ($\lambda_F=2\pi/k_F$, where k_F is the Fermi momentum).[15] The real part of the optical conductivity in the dc limit $\omega\to 0$ (Fig. 1) can be used to estimate the mean free path of the electrons in the metallic phase of VO_2 from the equation $\rho=\frac{3\pi^2\hbar}{e^2k_F^2l}$.[13] Here, ρ is the dc resistivity $[\rho=1/\sigma_1(\omega\to 0)]$. Since a single d electron per vanadium atom is delocalized,[2,3,5] one obtains $k_F \approx 10^8$ cm^{-1} assuming a spherical Fermi surface for simplicity. Thus the mean free path is ≈ 2.8 Å at $T=360$ K, i.e., l is nearly equal to the lattice spacing (2.85 Å) and less than λ_F (≈ 6 Å).[31] Therefore the Bloch-Boltzmann transport model no longer applies and the quasiparticle concept breaks down. It is observed that departure from the Boltzmann transport theory typically occurs for $l<10$ Å in metals.[8] If VO_2 were to avoid breakdown of Boltzmann transport through resistivity saturation similar to common metals, then saturation is expected to occur already at $T=360$ K [Fig. 2(b)]. Instead, resistivity increases continuously beyond the IRM limit.

It has been argued that a quantum theory of electrons which also includes correlation effects is required to give a correct account of resistivity behavior at short values of the electronic mean free path.[13] Within this theoretical framework, the optical conductivity of a correlated metal provides important insights into saturation phenomena. The low-energy response of correlated metals at reduced temperatures often reveals a Drude-like mode at far-IR frequencies attributable to well-defined charge carriers followed by a broad, incoherent part at higher energies. With increasing T, the

coherent Drude weight is transferred to the incoherent part.[1] Resistivity saturation is expected to occur when there is no further coherent spectral weight to be shed and only the incoherent part remains. In this model, the limiting dc conductivity $\sigma_s(0)$ of the incoherent part (corresponding to resistivity saturation) can be estimated from the *f*-sum rule:

$$\frac{\sigma_s(0)W}{\gamma} = \hbar \int_0^{\omega_c} \sigma_1(\omega) d\omega. \tag{4}$$

Here W is the one-particle bandwidth and γ is a weighting factor (≈ 2) depending upon the shape of the incoherent part of the conductivity.[13] We integrate the optical conductivity in the metallic phase of VO_2 up to the frequency of the minimum in the conductivity, beyond which interband transitions are clearly present. We take $W=2.59$ eV from recent LDA calculations.[3] Then using Eq. (4), we obtain $\sigma_s(0) \approx 1200\ \Omega^{-1}\ \mathrm{cm}^{-1}$. We will refer to this lower limit of the conductivity (upper limit of resistivity) as the "*f*-sum rule limit." Note that the *f*-sum rule resistivity is higher than the IRM resistivity [see Figs. 2(a) and 2(b)]. The conductivity decreases with increasing temperature but no saturation is observed because $\sigma_1(\omega \to 0)$ at $T=550$ K is still above the *f*-sum rule limit. The coherent part of the conductivity (the Drude peak at $\omega=0$) persists beyond the IRM limit in a regime where the quasiparticle picture is no longer valid. This is remarkable because some other bad metals with exceptionally short mean free paths usually exhibit a peak at *finite* frequency in $\sigma_1(\omega)$.[14,32]

At this stage, we briefly discuss the effect of phonons on the electronic properties of metallic VO_2. We calculate $1/\tau$ for electron-phonon scattering using a square-shaped Eliashberg function $\alpha^2(\Omega)F(\Omega)$ that mimics the phonon spectral density of VO_2,[26]

$$\frac{1}{\tau}(\omega,T) = \frac{\pi}{\omega}\int_0^{\infty} d\Omega \alpha^2(\Omega)F(\Omega)\left[2\omega \coth\left(\frac{\Omega}{2T}\right) - (\omega+\Omega)\coth\left(\frac{\omega+\Omega}{2T}\right) + (\omega-\Omega)\coth\left(\frac{\omega-\Omega}{2T}\right)\right]. \tag{5}$$

We note that the precise form of $\alpha^2(\Omega)F(\Omega)$ is not known for VO_2 but the shape of the calculated $1/\tau$ plots is not very sensitive to it. The upper cutoff for $\alpha^2(\Omega)F(\Omega)$ is taken as 0.1 eV which is the maximum phonon energy of VO_2.[33] As shown in Fig. 3(a), electron-phonon coupling theory predicts $1/\tau$ to be independent of frequency and strongly temperature dependent for frequencies greater than the maximum phonon energy.[26] However, in VO_2, $1/\tau$ is frequency dependent and nearly temperature independent at frequencies well above the maximum phonon energy [Fig. 3(a)]. Spectroscopic evidence and the lack of resistivity saturation indicate that electron-phonon scattering plays a less significant role in charge dynamics in the metallic phase of VO_2 compared to electronic correlations. Therefore the linear increase of resistivity and $1/\tau$ ($\omega \to 0$) with temperature cannot be due to electron-phonon scattering.

We note that there is broad consensus on the breakdown of the conventional quasiparticle picture for $l \lesssim d$ (or $l \lesssim \lambda_F$).[9,13,15] Despite attempts to understand transport in bad metals, including proposals beyond conventional Fermi-liquid theory, there is no convergence of views.[13,15,16,34,35] On the other hand, one cannot rule out the possibility of electronic phase segregation on a length scale smaller than infrared wavelengths. Also, silver films grown under certain conditions are known to exhibit bad metal behavior due to their peculiar microstructure and not due to any novel physics.[36] Nevertheless, empirical evidence indicates that a common feature of bad metals is the predominance of electronic correlations in their transport properties due to proximity to a Mott insulating phase.[14] We find that metallic VO_2 fits into this picture and suggest that the on-site electronic Coulomb repulsion is crucial to bad metal behavior and the MIT even though the insulating state may not be a conventional Mott insulator.[3,5]

III. CONCLUSION

We have determined the optical constants of VO_2 films in the insulating and metallic states over a wide temperature and frequency range. We find that VO_2 is a correlated metal above T_c and the charge dynamics are dominated by electronic correlations rather than electron-phonon scattering. In the metallic state, the length and energy scales associated with the scattering processes make it problematic to describe the charge carriers within Fermi-liquid theory. However, in contrast to some other bad metals,[14,32] the Drude peak persists in VO_2 for $l \lesssim d$. We establish that in metallic VO_2, the increase in resistivity with temperature beyond the IRM limit is due to the increase of scattering rate of charge carriers rather than a decrease of carrier density. The data are consistent with the model based on the fundamental *f*-sum rule in optics in which resistivity saturation is not expected at the IRM limit.[13]

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