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The metal-insulator transition in VO_2 studied using terahertz apertureless near-field microscopy

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We have studied the metal-insulator transition in a vanadium dioxide (VO_2) thin film using terahertz apertureless near-field optical microscopy. We observe a variation of the terahertz amplitude due to the phase transition induced by an applied voltage across the sample. The change of the terahertz signal is related to the abrupt change of the conductivity of the VO_2 film at the metal-insulator transition. The subwavelength spatial resolution of this near-field microscopy makes it possible to detect signatures of micron-scale metallic domains in inhomogeneous VO_2 thin films. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2801359]

Vanadium dioxide (VO_2) is prototype metal oxide with a complex phase behavior dictated by strong electron-lattice correlations.[1] It undergoes a first-order metal-insulator phase transition as well as a structural phase transition, from a low-temperature semiconducting phase to a high-temperature metallic phase, with associated significant changes to the conductivity and optical properties.[2–4] Recent work has shown that an electric field can also trigger the phase transition and that the transition temperature depends on the external bias.[5] In order to describe the infrared properties of VO_2 films near the metal-insulator transition (MIT), Choi *et al.* introduced a composite medium model, in which the VO_2 film is described as an inhomogeneous medium composed of metallic and insulating domains and the first-order phase transition is treated as a process of domain growth and eventual coalescence.[6] Recent experiments suggest that a destabilization of the V–V dimer leads to both the structural and the electronic phase transitions and that this process can be photoinduced.[7] Other recent work suggests that the structural and electronic phase transitions are distinct, separated by a monoclinic metallic phase. In this picture, the electronic transition is hole mediated and is not related to domain growth dynamics.[4] These two proposals predict very distinct microscopic characteristics in the vicinity of the phase transition. However, prior work on this material has always measured macroscopic (i.e., not intrinsic) conductivity, and no microscopic studies have been reported so far.

Terahertz apertureless near-field scanning optical microscopy (ANSOM) has recently been shown to provide the ability for broadband terahertz measurements with subwavelength resolution.[8,9] Recently, Jepsen *et al.* reported studies of the MIT of VO_2 thin films by terahertz transmission spectroscopy, showing a large change in the spatially averaged terahertz dielectric response associated with the change in conductivity of the sample.[10] Since the optical properties of the insulating and metallic phases are significantly different at terahertz frequencies, it is promising to use terahertz ANSOM to study VO_2 thin films for characterization of their phase behavior.

In this letter, we present an observation of a phase transition using terahertz ANSOM. The near-field terahertz signal scattered from a VO_2 thin film is studied as a function of temperature and external voltage, which is applied across the sample to trigger the MIT. We observe a significant enhancement of the amplitude of the terahertz signal around the transition from semiconducting to metallic phase, consistent with a dramatic change in the dielectric properties of the sample. With the approximately micron-scale spatial resolution of terahertz ANSOM, this enhancement can be observed either earlier than or later than the macroscopic MIT voltage, depending on the microscopic details of the metallic domain growth.

As in our earlier work, a fiber-coupled broadband terahertz system is used for terahertz generation and detection in the ANSOM setup.[8] The terahertz transmitter and the receiver are both photoconductive antennas based on low-temperature GaAs. As shown in Fig. 1, terahertz pulses are

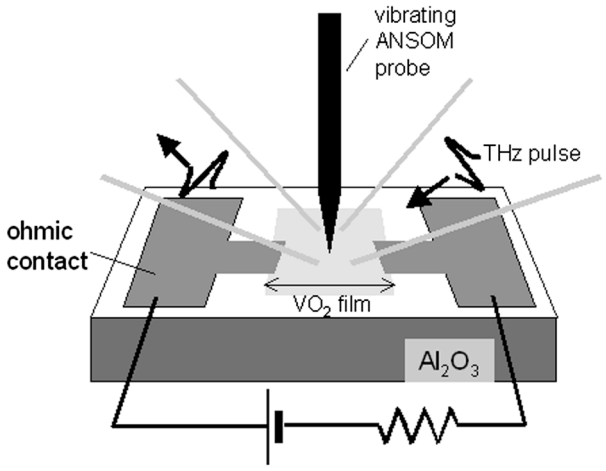

FIG. 1. (Color online) Schematic of terahertz ANSOM experiment. The terahertz beam is focused onto the surface of the VO_2 thin film. The scattered terahertz radiation is detected by a photoconductive antenna in the far field.

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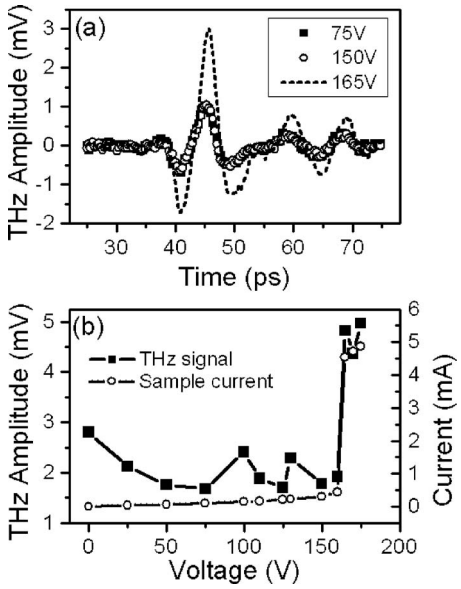

FIG. 2. (a) Time-domain waveforms of the terahertz signals scattered from the VO_2 thin film at 25 °C at three different voltages. The terahertz signal jumps abruptly when the voltage reaches 165 V. (b) Terahertz peak-to-peak amplitude and sample current as a function of the applied voltage across the sample at 25 °C.

focused on a subwavelength copper-beryllium tip of 1 μm point radius, which is held close (less than 500 nm) to the sample surface. The scattered terahertz radiation is modulated at 800 Hz by vibrating the tip normal to the sample surface with amplitude of about 750 nm. The terahertz signal is collected by the receiver in the far field with a lock-in amplifier referenced to the tip frequency, giving subwavelength resolution in the immediate neighborhood of the tip apex.[11–13] While the antenna effects of the long ($L \gg \lambda$) probe shaft can lead to substantial temporal reshaping of the terahertz waveform,[8] the measured scattered field is nevertheless an accurate indicator of the amplitude of the near field at the surface induced by the far-field illumination.

The VO_2 thin films were grown on Al_2O_3 substrates by laser ablation.[5,14] The thickness of the VO_2 thin films is about 100 nm. Two-terminal devices were fabricated with Au/Cr electrodes for the Ohmic contacts. In order to protect the VO_2 thin film, we connect an external resistor in series to limit the sample current. The VO_2 samples are mounted on a temperature-controlled stage. The channel lengths of the VO_2 samples vary from 3 to 100 μm. The transition voltage depends both on the sample length and the temperature. At room temperature (21 °C), the MIT occurs at a voltage above 210 V for a channel length of 50 microns, but at higher temperatures, the voltage required to induce the MIT decreases, as reported earlier.[5]

Figure 2(a) shows the time-domain plots of terahertz signals measured on the VO_2 thin film at 25 °C for different values of the applied dc bias. We observe a significant enhancement of the measured terahertz amplitude at the transition voltage (165 V), coincident with an abrupt jump of the dc current flowing through the sample, as shown in Fig. 2(b). When crossing through the MIT, the terahertz signal increases by a factor of 2, while the change of the dc conductivity of the VO_2 thin film is more than two orders of magnitude. These results are consistent with the fact that the metallic surface produces a stronger interaction with the near-field tip than the semiconducting surface due to the formation of an image dipole.[15] We may predict the enhancement of the terahertz ANSOM signal by estimating the change of the dielectric response of the VO_2 thin film at the MIT. The scattered terahertz signal is proportional to the scattering cross sections of the probe tip, $E_{\mathrm{THz}} \propto C_{\mathrm{sca}} = (k^4/6\pi)|\alpha_{\mathrm{eff}}|^2$, where

$$\alpha_{\mathrm{eff}} = \alpha(1+\beta)\left(1 - \frac{\alpha\beta}{16\pi r^3}\right)^{-1}.$$

Here, $\beta = (\varepsilon - 1)/(\varepsilon + 1)$ relates the image dipole p' with the dipole moment of the probe tip as $p' = \beta p$, ε is the complex dielectric constant of the region of the sample situated directly beneath the probe, α is the polarizability, and r is the tip-sample distance.[15] As in several previous studies,[6,10] we can use the Drude model, evaluated at the central frequency of our measured pulse, to obtain ε_m of the metallic domains. We further assume, following Jepsen *et al.*,[10] that ε_i of the insulating domains is equal to the high-frequency dielectric constant, $\varepsilon_i \approx \varepsilon_\infty = 9$. We then calculate that $E^m_{\mathrm{THz}}/E^i_{\mathrm{THz}} \propto |\alpha^m_{\mathrm{eff}}/\alpha^i_{\mathrm{eff}}|^2 \approx 1.3$, which is somewhat smaller than the measured enhancement of the terahertz signal. This discrepancy may result from the aforementioned approximations or may be the result of other effects that could enhance the terahertz signal. For example, approach curve measurements (not shown) indicate that the terahertz signal is very sensitive to the tip-sample separation when the VO_2 is metallic, but much less sensitive when it is in an insulating state. As a result, small changes in this separation, induced, for example, by changes in the dipolar attractive force on the tip, could lead to changes in the scattered terahertz amplitude.

The composite medium model mentioned above predicts that small metallic domains begin to grow in the thin film as the voltage is increased toward the MIT. Eventually, these domains coalesce and a continuous conducting pathway is formed. This results in an abrupt change in the macroscopic conductivity of the sample at a particular voltage V_{MIT}, which depends on the sample temperature. However, on a microscopic scale, one might expect that the transition from insulating to metallic behavior could be observed at slightly lower voltages $V < V_{\mathrm{MIT}}$. If a microscopic metallic domain happens to be located immediately below the position of the ANSOM tip, that region will become metallic at a lower voltage. On the other hand, if the region of the sample beneath the tip remains insulating even after the coalescence of the metallic domains, then the rise of the terahertz signal may even lag slightly behind the macroscopic transition voltage V_{MIT}. We observe both of these effects in our experiments—multiple measurements on a given location on a particular sample can give rise to slightly different transition voltages for the terahertz signal (see Fig. 3).

We have also studied the temperature dependence of the scattered terahertz signal from the VO_2 thin films. We observe that at higher temperatures, the jump in the scattered terahertz signal occurs at a lower voltage and is smaller [Fig. 3(b)], consistent with dc *I-V* measurements. This result is less easy to reconcile with the composite medium model, since the conductivity of the metallic rutile phase should not depend strongly on temperature, even in a single micron-sized domain. On the other hand, it is consistent with the existence of an intermediate monoclinic phase, which has

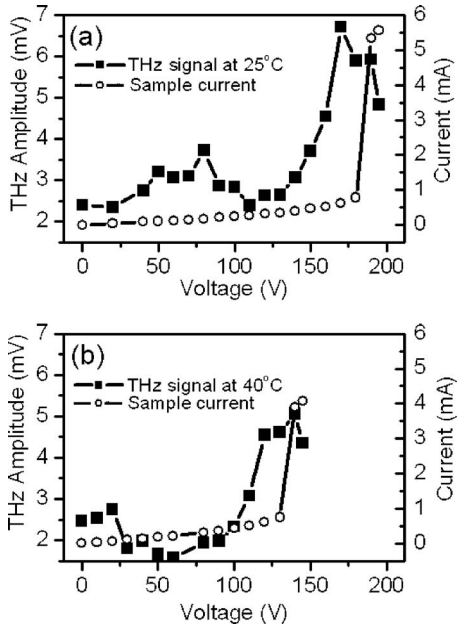

FIG. 3. Terahertz peak-to-peak amplitude and sample current as a function of the applied voltage across the sample at (a) 25 °C and (b) 40 °C. In (a), the terahertz signal rises at a lower voltage than the bulk phase transition voltage, which is an indication of spatially inhomogeneous conductivity. In (b), the smaller jump in the terahertz signal indicates a smaller conductivity of the metallic phase, which could be an indication of the intermediate monoclinic correlated metallic phase described in Ref. 4.

been shown to have a strongly temperature-dependent conductivity.[16]

In conclusion, we have demonstrated that terahertz ANSOM can be used to study the metal-insulator transition in a prototype metal oxide VO_2. These results provide evidence for the existence of an intermediate correlated metallic phase, as proposed recently.[4] They also suggest that spatial inhomogeneities in the film conductivity exist on a micron length scale even when the external voltage is below the transition voltage. Thus, a complete description of the phase transition in VO_2 may require *both* an understanding of the monoclinic phase and some version of a composite medium model in which percolation conduction plays a role. Future measurements in which the terahertz field is spectrally and spatially resolved should permit us to distinguish between Ohmic and non-Ohmic behavior, as well as to image the formation of inhomogeneities in the conductivity of the thin film.

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