Observation of the Mott Transition in VO₂ Based Transistors

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An abrupt Mott metal-insulator transition (MIT) rather than the continuous Hubbard MIT near a critical on-site Coulomb energy \( U/U_c = 1 \) is observed for the first time in VO₂, a strongly correlated material, by inducing holes of about 0.018% into the conduction band. As a result, a discontinuous jump of the density of states on the Fermi surface is observed and inhomogeneity inevitably occurs. The gate effect in fabricated transistors is clear evidence that the abrupt MIT is induced by the excitation of holes.

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In a strongly correlated system, a metal-insulator transition (MIT) near a critical on-site Coulomb energy, \( U/U_c = 1 \), has long been controversial in terms of whether the transition is abrupt or continuous in experiments [1-4], although a first-order MIT with temperature was observed by Morin [5]. An abrupt MIT indicates the Mott transition (first order) and a continuous MIT is the Hubbard transition (second order). The MIT breaks down an energy gap, formed by the strongly correlated Coulomb energy, between sub-bands in a main band. Mott first predicted that the abrupt MIT occurs when a lattice constant is larger than a critical value [1]. Brinkman and Rice theoretically demonstrated an abrupt MIT near \( U/U_c = 1 \) for a strongly correlated metal with an electronic structure of one electron per atom [6]. Hubbard first derived that, when sub-bands overlap just below \( U_c \), there is a finite minimum density of states (DOS) at the Fermi level, the DOS increases with decreasing \( U \), and the system is metallic [7]; this is Hubbard’s continuous MIT. Later, the continuous MIT was confirmed in the infinite-dimensional Hubbard model [2].

Applying an electric field to a two-terminal structure, Kumai et al. [8] measured Ohmic behavior in an organic Mott insulator in a regime, where conduction from non-conduction (insulating behavior) occurs. Through a theoretical consideration based on the Hubbard model, Oka et al. [9] described the Ohmic behavior in terms of a universal Landau-Zener quantum tunneling. Boriskov et al. [10] also observed a similar metallic behavior for VO₂ to that measured by Kumai et al. [8]. Thus, on the basis of the metallic behaviors and Oka’s analysis, the MIT just below \( U_c \) seems to follow Hubbard’s continuous model. However, considering the abrupt MIT observed in resistance measurement (Fig. 1), an abrupt MIT in the electric field should also be found.

In this letter, we observe an abrupt jump of current (or DOS) at an electric field in a two-terminal structure and measure the gate effect of the jump in a three-terminal device (switching transistor). The abrupt jump is analyzed in terms of an abrupt MIT (Mott transition). Inducing internal hole charges of about 0.018% in hole levels into the conduction band with a source-drain field or a gate field of a fabricated transistor [11] is an effective method of revealing the MIT mechanism. Note that Mott criterion, \( n_c \approx 3 \times 10^{18} \text{ cm}^{-3} \), is obtained from \( n_c^{1/3} a_H \approx 0.25 \), where \( a_H \) is the Bohr radius for VO₂ [12]. \( n_c \) corresponds to about 0.018% of the number of carriers in the half-filled band, when one electron in the cell volume, \( 59.22 \times 10^{-24} \text{ cm}^3 \), of VO₂ is assumed; the number of electrons is about \( 1.7 \times 10^{22} \text{ cm}^{-2} \).

**FIG. 1.** a, Temperature dependence of the resistance of a VO₂ film. Hysteresis is shown. b, The number of carriers measured by Hall effect. A change of carriers from holes to electrons is shown at 332 K. The minus sign indicates that the carriers are holes.

Thin films of a Mott insulator, VO₂, with a sub-energy gap of about 1 eV in the d band [13,14], have been deposited on Al₂O₃ and Si substrates by laser ablation. The thickness of the VO₂ films is about 900 Å. The resistance of the film decreases with increasing temperature and shows an abrupt MIT at a transition temperature, \( T_{n_c} = 340 \text{ K (68°C) (Fig. 1a). This corresponds with that measured by De Natale et al. [15] and Borek et al. [16].} \)
It was proposed that the abrupt MIT is the structural phase transition from monoclinic below $T_{tr}$ to tetragonal above $T_{tr}$ [5]. The decrease of the resistance up to 340 K indicates an increase of hole carriers, and two kinds of electron and hole carriers coexist near $T_{tr}=340$ K (Fig. 1b). From 332 to 340 K, the number of carriers is not discernable because of mixing of electrons and holes. We speculate that the number of hole carriers may be the Mott criterion, $n_e \approx 3 \times 10^{18} \text{ cm}^{-3}$, at $T_{tr}=340$ K on the general basis that an exponential decrease of the resistance with temperature in semiconductor physics indicates an exponential increase of carriers. Generally, in oxide materials, there are holes of about $5.5 \times 10^{18} \text{ cm}^{-3}$ which corresponds to 0.034% to $d$-band charges [17 - 19]. The holes are coupled to optical phonons [17]. In the metal regime above 340 K, the carriers are electrons (Fig. 1).

We fabricated transistors to observe the Mott transition. A schematic diagram of the transistors is shown in Fig. 2. Transistor 1 of a channel length, $L_{ch}=3 \mu m$, and a gate width, $L_w=50 \mu m$, was fabricated on Al$_2$O$_3$ substrates by lithography processes (Fig. 2a). A gate insulator of transistor 1 was an amorphous Ba$_{0.5}$Sr$_{0.5}$TiO$_3$ (BSTO) which was deposited on the VO$_2$ film. The interface between the VO$_2$ film and the amorphous gate insulator was sharp. A gate-source current, $I_{GS}$, between the gate and the source for transistor 1 is an order of $10^{-13}$ A, which indicates that there is sufficient insulation between the gate and the source. Transistors 2 and 3 of a gate length, $L_{ch}=5 \mu m$, and a gate width, $L_w=25 \mu m$, were manufactured on Si substrates. Their structure is shown in Fig. 2b. SiO$_2$ as the gate insulator was thermally treated. It is revealed that an interface between the polycrystal VO$_2$ film and the amorphous SiO$_2$ film is not sharp and complicated, and that the VO$_2$ films are inhomogeneous [20]. However, the SiO$_2$ insulator is strong with respect to a high field and is superior to the BSTO insulator for electronic application. Au/Cr electrodes were prepared for Ohmic contact. WSi is used as gate electrode. Characteristics of the transistors were measured by a precision semiconductor parameter analyzer (HP4156B). To protect transistors from excess current, the maximum current was limited to 20mA.

![FIG. 2. Schematic diagram of transistors. The dotted line between VO$_2$ and insulator is a channel. The gate insulators are an amorphous Ba$_{0.5}$Sr$_{0.5}$TiO$_3$ for transistor 1 (Fig. a), and thermally treated amorphous SiO$_2$ for transistors 2 and 3 (Fig. b).](image)

FIG. 3. Left-axis side: $J_{DS}$ vs. $E_{DS}$ for transistor 1. Right-axis side: Density of states (DOS), $dI/dV$, on the Fermi surface obtained from derivation with respect to $V_{DS}$. Inset: Ohmic behavior from $E_{DS}=0.8$ up to $4 \text{ MV/m}$ is shown. The maximum current was limited to 20mA to protect the transistor.

Figure 3 shows the drain-source current density, $J_{DS}$, vs. the drain-source electric field, $E_{DS}$, for transistor 1, measured at $V_{gate}=0$ V (two-terminal structure). $J_{DS}$ and $E_{DS}$ are obtained from the drain-source current and the drain-source voltage, respectively; $I_{DS}=J_{DS}S_{DS}(S_{DS}:\text{cross section})$ and $V_{DS}=E_{DS}L_{ch}$. $J_{DS}$ behavior below point A (Inset of Fig. 3), as observed by Boriskov et al. [10] and Kumai et al. [8] who used an organic Mott-insulator, is linear from $E_{DS} \approx 0.8$ to 4MV/m, but is nonlinear in the total regime. It was suggested that the linear Ohmic behavior occurs due to an applied field [10] and an induced current [8], not an increase of sample temperature due to leakage current. The Ohmic behavior was well described through a theoretical consideration in terms of a universal Landau-Zener quantum tunneling based on the Hubbard model [9]. However, since holes were observed below 334 K (Fig. 1b), it is asserted that the carriers for the Ohmic behavior are holes and the number of holes is very small. Thus, the Ohmic behavior is not an intrinsic property of metal and may be due to scattering of only a few carriers existing at room
Abrupt jumps of $J_{DS}$ at point A and the DOS at point $dA$ are shown in Fig. 3. The measured maximum-current density at point B is $J_{measured} \approx 2 \times 10^5$ A/cm², which is an order of the current density observable in a dirty metal. Note that the true maximum current density is much higher than $J_{measured}$, because the measurement was limited to 20mA. The jump of $J_{DS}$ between point A and point B corresponds to a jump of conductivity, $\sigma$, because $J_{DS} = \sigma E = n_{free}ev$ and $E$ is constant between points A and B; $\sigma \propto n_{free}$, where $n_{free}$ is the number of electron carriers (above point A) on the Fermi surface, and $v$ is the carrier velocity on the Fermi surface and is constant between points A and B due to constant $E$. This indicates that the number of carriers discontinuously increases, and that the DOS on the Fermi surface jumps from points $dA$ to $dB$ (Fig. 3). The jump of the DOS is a typical behavior of a first-order MIT which was first theoretically derived by Brinkman and Rice [6]. Note that carriers for current measured in semiconductors and metals are on the Fermi surface. Derivatives, $dI/dV$, at respective fields correspond to the DOSs on the Fermi surface (Fig. 3). The DOS in Fig. 3 can be expressed in terms of the Fermi level (Fig. 4a). Moreover, hysteresis loops of 5 times for transistor 2 were continuously measured (Fig. 4b), which may be due to the Joule heating and is also evidence of a first-order MIT. The abrupt jump was also measured more than 1,500 times without breakdown in a transistor. Fig. 4d shows Ohmic behavior after jump, which indicates that this phase is metal. Thus, we suggest that the jump at point A is the abrupt MIT (or Mott transition). If Hubbard’s continuous MIT exists, the jump should be not observed and the nonlinear behavior with an increasing field in the inset of Fig. 3 should be continuously exhibited from point A to point B in the electron system. However, continuous behavior is not found.

Figure 4c shows the off-current density, $J_{off-current}$, vs. the MIT drain-source electric field, MIT-$E_{DS}$, at $V_{gate}=0$ V (two-terminal structure). Although $V_{gate}=0$ V, the gate-source currents were very small to be ignored. The off-current is defined as $I_{DS}$ near $V_{DS}=0V$ and $V_g=0V$ (or gate open); other researchers refer to off-current as leakage-current. Data were selected from 5 transistors. MIT-$E_{DS}$ increases with decreasing $J_{off-current}$, which also provides information for revealing the mechanism of the abrupt jump. The off-current is caused by the excitation of holes in impurity levels such as oxygen deficiency. When the number of total holes in the hole levels is given by $n_{tot} = n_b + n_{free}$, where $n_b$ is the number of bound holes in the levels and $n_{free}$ is the number of holes freed from the levels. $n_b$ decreases with increasing $n_{free}$, because $n_{tot}$ is constant. The larger off-current is attributed to the increase of $n_{free}$. For the abrupt jump, $\Delta n \equiv n_c - n_{free}=0$ should be satisfied, where $n_c \approx 3 \times 10^{18}$ cm⁻³, as predicted by Mott. Hence, the decrease of $\Delta n$ (increase of $n_{free}$) contributes to the
reduction of the MIT-\(E_{DS}\).

Figure 4d shows \(J_{DS}\) vs. \(E_{DS}\) near the abrupt jump of transistor 3. Its characteristics are as follows. First, the gate effect at \(E_{DS}=2.8\text{MV/m}\) and \(V_{\text{gate}}=-10\text{V}\) is due to induced holes and occurs suddenly; this indicates attainment of \(n_c\). Second, the MIT-\(E_{DS}\) increases with increasing negative gate voltage (or field), which is due to a decrease of the conductivity; \(J_{DS}\)s at MIT points can be nearly regarded as constant and \(\sigma_{DS}=J_{DS}/E_{DS}\). This is due to an increase of hole carriers generated by the negative gate fields and indicates an increase of inhomogeneity (injection of holes to electron system). It was also observed that the MIT-\(E_{DS}\) decreases with increasing positive gate voltage when the off-current is large. Third, in the metal (electron system) regime over \(J_{DS}=0.9\times10^4\text{mA/cm}^2\), Ohmic behavior differs from the Ohmic behavior in Fig. 3 and arises from a dirty interface between the polycrystal VO\(_2\) film and the amorphous SiO\(_2\). The dirty interface causes resistance \cite{20} and is a channel where current flows (Fig. 2b). We suggest that the Ohmic behavior in Fig. 4d is a true metallic characteristic. Finally, the gate induced abrupt MIT reveals that the MIT depends upon the hole carriers in the semiconductor regime, and that inhomogeneity also inevitably arises from hole doping \cite{21}.

In conclusion, in the Mott insulator of VO\(_2\), MIT near \(U/U_c=1\) abruptly occurs in company with inhomogeneity through semiconduction as a doping process of internal holes of \(n_c\approx0.018\%\). This will be observed in all Mott insulators.

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