Temperature dependence and control of the Mott transition in VO$_2$-based devices

Hyun-Tak Kim (htkim@etri.re.kr), B. G. Chae, D. H. Youn, S. L. Maeng, and K. Y. Kang

Telecom. Basic Research Lab, ETRI, Daejeon 305-350, Korea

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The transition voltage of an abrupt metal-insulator transition (MIT), observed by applying an electric field to two-terminal devices fabricated on a Mott insulator VO$_2$ film, decreases with increasing temperature up to 334 K. The abrupt current jump disappears above 334 K near the MIT temperature. These results suggest that the mechanism of the abrupt MIT induced by temperature is the same as that by an electric field. The magnitude of the current jump (a large current) decreases with increasing external resistance; this is an important observation in terms of applying the abrupt MIT to device applications. Furthermore, the temperature and resistance dependence of the MIT cannot be explained by the dielectric breakdown although a current jump known as breakdown is similar to that observed in an abrupt MIT.

Thin films of vanadium dioxide, VO$_2$, have been extensively studied for electronic and electro-optic device applications [1–3] of an abrupt first-order metal-insulator transition (MIT) at a critical temperature $T_c \approx 340$ K [4–6]. Recently, a new type of abrupt MIT, having an abrupt jump of driving current in a two-terminal device fabricated on an epitaxial VO$_2$ film, has been demonstrated by electric field excitation [7]. The abrupt MIT does not undergo a structural phase transition [8], as predicted by Mott for an abrupt first-order MIT driven by strongly correlated electronic Coulomb energy [9].

When an abrupt MIT occurs and excess current flows in a device, the device can be damaged or its characteristics can be degraded. Furthermore, the electric I-V characteristics of the abrupt MIT are similar to those of dielectric breakdown observed at high electric fields in thin AlO$_x$ and HfO$_2$ gate insulators [10,11]. Clearer evidence of the Mott transition, a control method of the excess current for device applications utilizing abrupt MITs, and evidence of the difference between a breakdown and an abrupt MIT are important unresolved issues in this field.

In this paper, we measure the temperature dependence of abrupt MITs in VO$_2$ driven by a DC electric field to suggest evidence of the Mott transition, and control the magnitude of the abrupt current jump (or excess current) using an external resistance. The effect of measurement on the magnitude of the observed current is briefly discussed. An important difference between a breakdown and an abrupt MIT is also given.

Thin films of VO$_2$ have been deposited on (1102) Al$_2$O$_3$ and Si substrates by laser ablation [12]. The thickness of the VO$_2$ films is about 900 Å. For two-terminal devices, Ohmic-contacted Au/Cr electrodes on VO$_2$ films with a channel width of 25 µm and a channel length of 5 µm were patterned by photo-lithography and lift-off. I-V characteristics of the devices were measured by a precision semiconductor parameter analyzer (HP4156B).

Figure 1 (a) shows the temperature dependence of the resistance of an epitaxial VO$_2$ film I. The resistance decreases with increasing temperature and shows an abrupt MIT at a critical temperature $T_c \approx 340$ K (68°C). This is consistent with previous measurements [4,5]. It was proposed that this abrupt MIT is due to the structural phase transition from monoclinic below $T_c$ to tetragonal above $T_c$ [13,14]. The decrease of the resis-

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tance up to 340 K indicates an increase of hole carriers, and two kinds of electron and hole carriers coexist near \( T_c \approx 340 \) K, as shown in the inset of Fig. 1 (a). Owing to a mixing of electrons and holes, the number of carriers at temperatures from 332 to 340 K is not exactly determined. The number of hole carriers at \( T_c \approx 340 \) K is expected to be \( n_c \approx 3 \times 10^{18} \text{ cm}^{-3} \) from the Mott criterion [1,9], based on an exponential decrease of resistance (increase of carrier) with increasing temperature. \( n_c \) corresponds to 0.018% of d-band charges. In the metal regime above 340 K, the major carriers are electrons, as shown in the inset of Fig. 1 (a). Fig. 1 (b) shows the drain-source-voltage dependence of conducting current, \( I_{DS} \), density, \( J_{DS} \), of the flow between two terminals (drain-source) for a VO\(_2\) film II. An abrupt current jump near the transition voltage \( V_t = 20 \) V is shown and Ohmic behavior as a characteristic of metal is also exhibited over the transition voltage. This is a typical characteristic of a first-order transition and is reproducible more than 1,500 times.

This arises from excitation of hole charges by temperature. At 338 K, near the transition temperature of the abrupt MIT, and beyond, \( I_{DS} \) follow Ohmic behavior without any current jump, in contrast to the MITs with a current jump below 338 K; this is observed for the first time. Note that the device was protected by a compliance current of 3 mA and the measurement was carried out without external resistance.

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\Delta n = n_c - n_{free}(T,E) \approx 0 \quad \text{for} \quad n_c = 3 \times 10^{18} \text{ cm}^{-3},
\]

as predicted by Mott [1,9]. At \( T_c \approx 340 \) K, it is suggested, as decisive evidence of the Mott transition, that the abrupt current jump will disappear, because \( n_{free}(T = 340K,E = 0) \approx n_c \) (i.e. \( \Delta n = 0 \)) is excited by only temperature, as shown in Fig. 2(a).

Below \( T_c \approx 340 \) K, the abrupt MIT voltage decreases with increasing temperature, because, from \( n_c \equiv n_{free}(T,E) = n_{free}(T) + n_{free}(E) \), the increase of \( n_{free}(T) \) with increasing temp-

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**FIG. 2.** Temperature dependence of abrupt MIT observed at device II fabricated on a VO\(_2\) film II/Al\(_2\)O\(_3\) substrate above (Fig. a) and below (Fig. b) room temperature. Above \( T = 66^\circ \text{C} \), apparent metallic behavior appears.

Figures 2 (a) and (b) show the temperature dependence of the abrupt MIT measured at device II fabricated on a VO\(_2\)/Al\(_2\)O\(_3\) film III. The transition voltage of the abrupt MIT decreases with increasing temperature.

**FIG. 3.** External resistance dependence of abrupt MIT measured, (Fig. a) at device I fabricated on a VO\(_2\) film II/Al\(_2\)O\(_3\) substrate, (Fig. b) at device III fabricated on a VO\(_2\) film IV/SiO\(_2\)/Si substrate.

The temperature dependence also provides decisive information for revealing the mechanism of the abrupt jump. When the number of total holes, \( n_{tot} \), in the hole levels is given by \( n_{tot} = n_b + n_{free}(T,E) \), where \( n_b \) is the number of bound holes in the levels and \( n_{free}(T,E) \) is the number of holes freed by temperature, \( T \), and electric field, \( E \), from the levels, \( n_b \) decreases with increasing \( n_{free}(T,E) \). For the abrupt jump, \( \Delta n \equiv n_c - n_{free} = 0 \) should be satisfied, where \( n_c \approx 3 \times 10^{18} \text{ cm}^{-3} \), as predicted by Mott [1,9]. At \( T_c \approx 340 \) K, it is suggested, as decisive evidence of the Mott transition, that the abrupt current jump will disappear, because \( n_{free}(T = 340K,E = 0) \approx n_c \) (i.e. \( \Delta n = 0 \)) is excited by only temperature, as shown in Fig. 2(a). Below \( T_c \approx 340 \) K, the abrupt MIT voltage decreases with increasing temperature, because, from \( n_c \equiv n_{free}(T,E) = n_{free}(T) + n_{free}(E) \), the increase of \( n_{free}(T) \) with increasing tem-
temperature decreases $n_{free}(E)$. Thus, it is revealed that the mechanism of the abrupt MIT excited by temperature (Fig. 1) is the same as that by an electric field. In addition, if the abrupt current jump occurs by breakdown due to a high field, the temperature dependence and the change near 338 K of MIT cannot be explained.

Figures 3 (a) and (b) show the external resistance dependence of the magnitude of abrupt current jumps observed at device II fabricated on a VO$_2$/Al$_2$O$_3$ film II and device III fabricated on a VO$_2$/SiO$_2$/Si film III, respectively. With increasing external resistance, the magnitude of the abrupt current jump decreases and the MIT voltage increases. This can be explained by our model, as shown in Fig. 4 (a), where the metal region decreases with increasing external resistance in the measurement region. If only the metal region in Fig. 4 (a) is measured, the magnitude of the current jump might be of an order of $\sim 10^7$ A/cm$^2$, the current density of a good metal. That is, $I_{DS}$ observed at 5 KΩ do not have the characteristics of a current jump, while $I_{DSS}$ measured at 1 KΩ and less resistance, display jumps, as shown in Figs. 3(a) and (b). This indicates that the observed $I_{DS}$ changes with an external resistance change, even though the intrinsic metal characteristic remain unchanged. Thus, the observed current density, $J_{DS}$, of an order of $\sim 10^5$ A/cm$^2$ in Fig. 3 (a) is an average of the metal region over the measurement region, as shown in Fig. 4 (b). The average is the effect of measurement. Furthermore, since the VO$_2$ film has electrons and holes, as observed by Hall measurement in Fig. 1, it is regarded that the VO$_2$ film is intrinsically inhomogeneous, although an external resistance effect is excluded. The inhomogeneity is an intrinsic characteristic of a material with an abrupt current jump and was confirmed by high resolution cross-sectional transmission-electron microscopy [12]. Thus, a true current jump cannot be measured in an inhomogeneous system, as shown in Fig. 4, as has been explained by the extended Brinkman-Rice picture [15].

In conclusion, the temperature dependence of the abrupt MIT explains the mechanism of the abrupt Mott MIT. In particular, the external resistance dependence of the magnitude of the current jump is an important key to application of the abrupt MIT to optic and electronic devices. Furthermore, the breakdown phenomenon observed in thin gate insulators [10,11] may be the abrupt Mott MIT.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig4.png}
\caption{When a device is measured, the resistance is included in the measurement region. This indicates that the measurement region is inhomogeneous.}
\end{figure}

We have observed a true dielectric breakdown which forms a current path with a very low source-drain voltage (or field) and a high current such as a 'short' phenomenon, after a device is applied to a very high current or electric field. After the dielectric breakdown occurs, the current jump is not observed. The dielectric breakdown is regarded as a device breakdown. The abrupt current jump is reproducible without degradation even in thousands of measurements, when an external resistance is attached in the circuit. The external resistance is a decisive key to controlling the abrupt MIT for application devices.

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