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Origin of variation in switching voltages in threshold-switching phenomena of VO$_2$ thin films

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We investigated the origin of the variation in switching voltages in threshold-switching of VO$_2$ thin films. When a triangular-waveform voltage signal was applied, the current changed abruptly at two switching voltages, i.e., $V_{\text{ON}}$ (insulator-to-metal) and $V_{\text{OFF}}$ (metal-to-insulator). $V_{\text{ON}}$ and $V_{\text{OFF}}$ were measured by changing the period of the voltage signal, the temperature of the environment, and the load resistance. We observed that either $V_{\text{ON}}$ or $V_{\text{OFF}}$ varied significantly and had different dependences with respect to the external parameters. Based on the mechanism of the metal–insulator transition induced by Joule heating, numerical simulations were performed, which quantitatively reproduced all of the experimental results. From the simulation analysis, the variation in the switching voltages for threshold-switching was determined to be thermal in origin.

Threshold-switching is a volatile and reversible transition that occurs between the high- and low-resistance states. The phenomenon is of great interest in condensed matter research because it causes significant changes in a material’s electrical, optical, and mechanical properties and can be triggered by different external (thermal, electrical, or optical) stimuli. VO$_2$ is an example of a typical threshold-switching material. VO$_2$ threshold-switching has been widely studied for various technological applications, such as electrical switches, smart windows, terahertz nanoantennas, and memory metamaterials.

To utilize the VO$_2$ threshold-switching for practical purposes, it is important to understand and control detailed properties of the phenomenon. In particular, control over the switching voltages, where abrupt current changes occur by application of electric fields, is crucially important for stable performance of VO$_2$ devices. Despite such importance, intentional control over the switching voltages has not been quantitatively investigated to date. We address a key question in this work: can the switching voltages be varied by changing the external parameters, and if they can, what is the origin of the variation?

Here, we measured the switching voltages of threshold-switching in VO$_2$ thin films by applying a triangular-waveform voltage signal. We found significant variation in switching voltages with changes in the period of the voltage signal, environmental temperature, and load resistance. We reproduced all of these experimental findings using numerical simulations based on the mechanism of the metal–insulator transition induced by Joule heating. Our simulation results indicated that the variation in the switching voltages was determined by the generation and dissipation of Joule heating.

We deposited insulating VO$_2$ thin films onto Al$_2$O$_3$ (0001) substrates by pulsed laser deposition. The films were grown at a substrate temperature of 560 $^\circ$C, an oxygen pressure of 6.5 mTorr, and a laser fluence of 1.5 J/cm$^2$. Photolithography and DC-magnetron sputtering were used to deposit 200 x 200-$\mu$m$^2$ area Pt electrodes onto the VO$_2$ thin films. The inset of Fig. 1(a) shows a schematic diagram of the Pt/VO$_2$/Al$_2$O$_3$ sample geometry and the electrical measurement setup. (b) The resulting current $I(t)$, restricted by the current flow of the load resistor. We defined the switching voltages $V_{\text{ON}}$ and $V_{\text{OFF}}$ as $V(t)$ at the time when $I(t)$ abruptly increased and decreased, respectively. (c) Procedure for formation of a channel on the surface of the VO$_2$ thin film; the channel has different optical properties than its surroundings.

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measurement setup. For all of the electrical measurements, we selected two adjacent Pt electrodes, which were separated by 25 μm.

Threshold-switching was induced in the VO2 thin films by applying a triangular-waveform voltage signal, as shown in Figs. 1(a) and 1(b). The triangular-waveform voltage signal \( V(t) \) was swept linearly over time \( t \), with a fixed amplitude of \( V_{\text{max}} = 60 \, \text{V} \), using a Yokogawa FG300 synthesized function generator (Fig. 1(a)). The resulting current \( I(t) \) was measured simultaneously, using a Yokogawa DL7100 digital oscilloscope (Fig. 1(b)). To avoid pulse shape deformation, the impedance was matched using an nF HSA4101 high-speed bipolar amplifier. An abrupt increase and decrease in \( I(t) \) occurred at \( V(t) = V_{\text{ON}} \) and \( V_{\text{OFF}} \), indicating insulator-to-metal and metal-to-insulator transitions, respectively. The current flow was limited by connecting a load resistor of resistance value \( R_{\text{load}} \) in series; this prevented permanent sample damage by an abrupt current jump at \( V_{\text{ON}} \).

We observed the formation of a channel with different optical property between the two electrodes when the VO2 thin film became metallic. Figure 1(c) shows one example of sequential snapshots of channel formation during the voltage application (Fig. 1(a)). Color changes were monitored in the VO2 thin film by an optical charge-coupled device camera. The first snapshot shows that there was no channel on the VO2 film surface at \( V(0) = 0 \, \text{V} \). When the VO2 thin film became macroscopically metallic at \( V(t) > V_{\text{ON}} \), a darker localized channel appeared in the film, as shown in the second snapshot. Further increases in \( V(t) \) made the channel wider (third snapshot); however, the channel width became narrower when \( V(t) \) decreased (fourth snapshot). When the VO2 thin film became macroscopically insulating at \( V(t) < V_{\text{OFF}} \), the channel disappeared (fifth snapshot). This analysis indicated that the channel was highly conducting, compared with other regions; i.e., the threshold-switching of the VO2 thin film was accompanied by conducting channel formation and rupture.

To collect information on the external parameters affecting the switching voltages, we first measured \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) by changing \( t_p \), the period of the triangular-waveform voltage signal (Fig. 1(a)). \( V_{\text{ON}} \) decreased significantly as \( t_p \) increased, but \( V_{\text{OFF}} \) did not change within the standard deviation of the experimental values. Figure 2(a) shows an example of the \( t_p \)-dependence of \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) for room temperature and a fixed \( R_{\text{load}} = 1 \, \text{kΩ} \). \( V_{\text{ON}} \) decreased from \( \sim 50 \) to \( \sim 25 \, \text{V} \) (red closed circles), whereas \( V_{\text{OFF}} \) remained almost constant at \( \sim 20 \, \text{V} \) (blue open circles), when \( t_p \) increased from \( 10^{-3} \, \text{s} \) to \( 10^{2} \, \text{s} \), as shown in Fig. 2(a).

We then investigated the dependence of \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) on the environmental temperature, \( T_e \), using a probe station equipped with liquid nitrogen. We found that both \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) decreased as \( T_e \) increased. Figure 2(b) shows an example of the \( T_e \)-dependence of \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) for a fixed \( t_p = 0.1 \, \text{s} \) and \( R_{\text{load}} = 1 \, \text{kΩ} \). \( V_{\text{ON}} \) decreased from \( \sim 58 \) to \( \sim 38 \, \text{V} \) (red closed circles), and \( V_{\text{OFF}} \) also decreased from \( \sim 25 \) to \( \sim 20 \, \text{V} \) (blue open circles), when \( T_e \) increased from 240 to 290 K.

We investigated the \( R_{\text{load}} \)-dependence of \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) in the range of \( 500 \, \Omega \leq R_{\text{load}} \leq 2000 \, \Omega \) and observed that both \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) increased as \( R_{\text{load}} \) increased. In this case, \( R_{\text{load}} \) was smaller than the in-plane resistance of the insulating VO2 thin film and larger than that of the metallic VO2 thin film. Figure 2(c) shows an example of the \( R_{\text{load}} \)-dependence of \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) for room temperature and a fixed \( t_p = 0.1 \, \text{s} \). \( V_{\text{ON}} \) increased from \( \sim 31 \) to \( \sim 46 \, \text{V} \) (red closed circles), and \( V_{\text{OFF}} \) also increased from \( \sim 15 \) to \( \sim 29 \, \text{V} \) (blue open circle), when \( R_{\text{load}} \) increased from 500 to 2000 Ω (Fig. 2(c)).

The origin of the conductance transition of VO2 thin films triggered by an electric stimulus is still debatable. Some researchers suggest that the transition occurs by purely electric-field-driven phase transitions. Other research groups have attributed the thermally induced phase transition to Joule heating, due to a referential experiment that revealed a sudden metal–insulator transition near 340 K in VO2 films. If we consider only the electric-field-driven phase transition as the origin for our experimental results, then it is hard to understand the \( t_p \)- and \( T_e \)-dependences for the switching voltages because the electric field strength does not depend on them. Note that the RC time for the electric circuit in our experiments was on the order of 10 ns. Thus, the electric-field distribution over the entire circuit immediately relaxed within the experimental time range (≥1 ms). This indicated that the RC time is not the origin of the \( t_p \)-dependence. As a result, the metal–insulator transition observed in our experiments could be attributed to thermally induced phase transition by Joule heating.

![Figure 2](https://example.com/figure2.png)

**FIG. 2.** Dependence of \( V_{\text{ON}} \) and \( V_{\text{OFF}} \) on (a) the period of the triangular-waveform voltage signal, \( t_p \), (b) the environmental temperature, \( T_e \), and (c) the resistance of the load resistor, \( R_{\text{load}} \).
We performed numerical simulations based on the mechanism of the thermally induced metal–insulator transition to quantitatively understand the observed variation in the switching voltages. In the simulations, a two-dimensional lattice for the VO$_2$ thin film was assumed, whose lengths in the $x$- and $y$-directions were $L_x = 200$ $\mu$m (40 divisions) and $L_y = 25$ $\mu$m (1 division), respectively, as shown in the inset of Fig. 3(a). Note that the film thickness was set to 100 nm. We confirmed that the number of divisions in the $L_y$ direction had no significant effect on the simulation results. The three-dimensional lattice for Al$_2$O$_3$ was attached under the VO$_2$ lattice, whose thickness was 1 mm (30 divisions for feasible calculation). We assumed that all of the VO$_2$ cells in the lattice were in the insulating state at the beginning of the simulation and were held at a temperature of $T_o$. With this initial configuration, the external voltage $V_{ext}$ increased over time, starting from zero. The current density, $j_n$, flowing through a cell indexed by $n$, is given by the Ohm’s law. Here, the current was assumed to flow along the direction perpendicular to the electrode, only in the VO$_2$ layer. To investigate the time evolution of the temperature profile, we solved the heat equation:

\[ c_p^n \rho_n \frac{\partial T_n}{\partial t} = \nabla \cdot (\kappa_n \nabla T_n) + j_n^2 / \sigma_n, \]  

where $c_p^n$, $\rho_n$, $\kappa_n$, and $\sigma_n$ are the specific heat, density, thermal conductivity, and electric conductivity of a cell indexed by $n$, respectively. Here, $T_n$ is the temperature at the center of the cell $n$. The time-step increment was fixed at $10^{-7}$ s for all simulations. We assumed that the heat-diffusion effect was negligible at the electrodes and that the cells in contact with the ambient surroundings were cooled by air convection. To account for the thermally induced phase transition in our simulation, the following switching rules were used for the state of the VO$_2$ cell:

- Insulator $\rightarrow$ metal when $T_n > T_c$,
- Metal $\rightarrow$ insulator when $T_n < T_c$.

For the VO$_2$ thin film, $T_c = 340$ K. The $\kappa$ and $\sigma$ values of the VO$_2$ cell depended on its state; we used $\kappa_i = 3.5$, $\kappa_m = 6$ W (mK)$^{-1}$, and $\sigma_i = 300$, $\sigma_m = 2 \times 10^6$ $\Omega^{-1}$ m$^{-1}$ (Refs. 11–13), where index $i$ and $m$ denote insulator and metal, respectively. We used $c_p = 690$ J (kg-K)$^{-1}$ and $\rho = 4340$ kg m$^{-3}$ for VO$_2$ and $c_p = 880$ J (kg-K)$^{-1}$, $\rho = 4000$ kg m$^{-3}$, and $\kappa = 45$ W (mK)$^{-1}$ for Al$_2$O$_3$.

When a triangular-waveform voltage signal with $V_{max} = 60$ V was applied, our simulation resulted in the threshold-switching $I(t)$–$V(t)$ curve, with abrupt current jumps at $V_{on}$ and $V_{off}$, as shown in Figs. 3(a) and 3(b). Figure 3(c) shows the sequential channel formation and $T_n$-distribution when the threshold-switching (Fig. 3(b)) occurred. With zero voltage bias, $T_n = T_o$ ($< T_c$) and there was no metallic channel, as shown in the first snapshot. When $V(t) > V_{on}$, $T_n$ of some cells reached a temperature higher than $T_c$, due to Joule heating; a local channel formed, as shown in the second snapshot. Further increases in $V(t)$ led to an increase in the channel width (the third snapshot), while the channel width decreased as $V(t)$ decreased (the fourth snapshot). Finally, when $V(t) < V_{off}$, $T_n$ for all of the cells became lower than $T_c$, and the channel disappeared, as shown in the fifth snapshot. The simulated metallic channel formation agreed well with experimental observation, as shown in Fig. 1(c).

Figure 4(a) shows the simulated $t_p$-dependence of $V_{on}$ and $V_{off}$. Similar to the experimental results in Fig. 2(a), $V_{on}$ decreased as $t_p$ increased; however, $V_{off}$ showed no apparent change. The importance of Al$_2$O$_3$ should be noted; without the Al$_2$O$_3$ lattice, the temperature of the VO$_2$ film would rise too fast, and the simulation would be unable to reproduce the experimental results quantitatively. To understand the simulated $t_p$-dependence analytically, consider a “lump” of VO$_2$ directly connected to the heat bath held at temperature $T_e$. Equation (1) can then be written as follows:

\[ C_d \frac{dT}{dt} = -\frac{1}{R_{th}} (T - T_e) + \frac{V_{VO2}^2}{R}, \]

where $C_d$, $T$, $R_{th}$, and $R$ are the heat capacitance, temperature, effective thermal resistance, and resistance of the VO$_2$ film, respectively. $V_{VO2}$ is the applied voltage to the VO$_2$ thin film, which is given by $RV_{ext}/(R + R_{load})$. Note that $R = R_{th}$ and $R = R_{on}$ when VO$_2$ is in an insulating and metallic state, respectively. $V_{ext} = 2V_{max}/t_p$ for $t < t_p/2$ and $V_{ext} = 2V_{max}(1 - t/t_p)$ for $t > t_p/2$. Because Eq. (2) is a first differential equation, it can be easily solved. In particular, for $t < t_p/2$, one can show that

![Image](image_url)
after-saturation behavior of connected substrate and VO2 film in the actual experiment. For $t_p < t_{th}$, we observed that both $V_{ON}$ and $V_{OFF}$ decreases further, rather than saturate. We think that this originates from the effectively larger $C$ due to the larger environment, which includes the connected substrate and VO2 film in the actual experiment. For $t > t_p/2$, after some calculation, one can show that

$$V_{OFF} \approx B', \quad \text{for} \quad t_p \gg CR_{th} \left( B' = \left( T_e - T_c \right) / \left( R_m + R_{load} \right) \right).$$

Plugging $R_{th} \sim 300 \text{sK} / \text{J}$ into Eq. (4) gives $B' \sim 20 \text{V}$, which corresponds to the saturation value for $V_{OFF}$ in the simulation. These analytic results clearly demonstrate that $V_{ON}$ and $V_{OFF}$ are determined by thermal processes resulting from Joule heating.

Figure 4(b) shows the $T_e$-dependence of $V_{ON}$ and $V_{OFF}$. Both $V_{ON}$ and $V_{OFF}$ decreased with increasing $T_e$, as we observed experimentally in Fig. 2(b). These behaviors can be easily understood from Eqs. (3) and (4). Figure 4(c) shows the $R_{load}$-dependence of $V_{ON}$ and $V_{OFF}$. Both $V_{ON}$ and $V_{OFF}$ increased with an increase of $R_{load}$, as experimentally shown in Fig. 2(c). This can also be understood from Eqs. (3) and (4). The agreement between the numerical simulations and the experimental results indicates that the variation in switching voltages of threshold-switching in VO2 thin film has a thermal origin.

In summary, we explored the effect of various external parameters on the switching voltages for threshold-switching in VO2 thin films. We applied a triangular-waveform voltage signal, we observed that $V_{ON}$ decreased with an increase of $t_p$. For a fixed $t_p$, we observed that both $V_{ON}$ and $V_{OFF}$ decreased (or increased) as $T_e$ or $R_{load}$ increased. A simulation based on the mechanism of thermally induced phase transition reproduced all of the experimental results quantitatively. We also gave a simple analytic explanation for the simulation results. Thus, our simulation and analytic results demonstrate that variation in switching voltages originates from thermal processes induced by Joule heating and its dissipation, i.e., the switching voltages for VO2 threshold-switching can be intentionally controlled or accidentally changed by variation of the thermal environment. This indicates the importance of thermal architecture for stable performance of VO2 applications.

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