Metastable State-Induced Consecutive Step-like Negative Differential Resistance Behaviors in Single Crystalline VO$_2$ Nanobeams

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1. The parameters and related values used for theoretical simulations

Table S1. The parameters and related values used for theoretical simulations.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity of VO₂</td>
<td>6.5 W/mK</td>
<td><em>Phys. Rev.</em> 185, 1022 (1969)</td>
</tr>
<tr>
<td>Heat transfer coefficient</td>
<td>10 W/m²K</td>
<td><em>J. Mater. Sci.</em> 2009, 44, 5345</td>
</tr>
<tr>
<td>Voltage</td>
<td>Threshold voltages</td>
<td>Experimental values extracted from Figure 2(b)</td>
</tr>
<tr>
<td>Current</td>
<td>Threshold currents</td>
<td>Experimental values extracted from Figure 2(b)</td>
</tr>
</tbody>
</table>

2. The simulated temperatures at different device (set) temperatures

Table S2. Information about the simulated temperatures at different device (set) temperatures.

<table>
<thead>
<tr>
<th>Set temperature</th>
<th>Maximum temperature rise</th>
<th>Temperature deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>40 °C</td>
<td>2.6 °C</td>
<td>± 0.4 °C</td>
</tr>
<tr>
<td>50 °C</td>
<td>3.0 °C</td>
<td>± 0.55 °C</td>
</tr>
<tr>
<td>55 °C</td>
<td>2.8 °C</td>
<td>± 0.45 °C</td>
</tr>
<tr>
<td>60 °C</td>
<td>3.7 °C</td>
<td>± 0.5 °C</td>
</tr>
<tr>
<td>65 °C</td>
<td>4.4 °C</td>
<td>± 0.8 °C</td>
</tr>
<tr>
<td>70 °C</td>
<td>4.6 °C</td>
<td>± 0.85 °C</td>
</tr>
</tbody>
</table>
3. Threshold voltage and current ($V_{th}$, $I_{th}$) versus the calculated temperature ($T_{cal.}$) and temperature change versus $V_{th}$

**Figure S1.** (a) Threshold voltage ($V_{th}$) and threshold current ($I_{th}$) as a function of the calculated temperature ($T_{cal.}$). (b) Temperature change as a function of threshold voltage. $\Delta T = T_{set} - T_{cal.}$ The gray-colored region shows temperature rise is less than 5 °C.
4. Calculated temperature rise at 60 °C for the VO$_2$ nanobeam with a change in thermal conductivity

According to Berglund et al.$^{S1}$ thermal conductivity of bulk VO$_2$ across the MIT showed no significant temperature dependence. Most recently, Lee et al.$^{S2}$ demonstrated that single-crystal VO$_2$ nanobeams exhibited very little change in thermal conductivity across the MIT, which was consistent with a previous study on bulk VO$_2$$^{S1}$. Although a change in thermal conductivity of VO$_2$ across the IMT is very little, we further performed additional calculations, assuming that the thermal conductivity increases by as much as 60 % in the metallic phase$^{S3}$. As shown in Figure S2 below, the simulated temperature was found to decrease from approximately 3.7 to 3 °C while increasing the thermal conductivity by 60 % in the metallic phase. This implies that the change in thermal conductivity of VO$_2$ through the IMT doesn’t affect much the temperature rise below the transition temperature of the VO$_2$ nanobeam.

![Figure S2. Calculated temperature rise at 60 °C for the VO$_2$ nanobeam with a change in thermal conductivity](image-url)
5. The effect of the interface thermal conductance on temperature rises

We considered heat dissipation through the solid interface of the VO\textsubscript{2} and the insulating dielectric substrate (sapphire substrate) as one of primary heat dissipation processes when we carried out the simulation. The thermal conductance across the solid dielectric substrate (the contribution of conduction to heat transfer) is significantly much larger than that through air (the contribution of convection heat transport to air from the remaining surfaces of VO\textsubscript{2}) because of large differences in thermal conductivity between solid, liquid, and gas states of matter. This means that the temperature rise is mainly affected by the convective heat transfer to the ambient air (rather than a heat dissipation through the solid interface) as shown in the inset of Figure 3, showing the distribution of calculated temperature rise in the cross-sectional image.

According to the Lyeo et al.\textsuperscript{54} the thermal conductance of solid interfaces between metal and dielectric is a range of 10 - 100 MW/m\textsuperscript{2}K. As we described in Table S1, we chose the thermal conductivity of sapphire as 30 W/mK as our simulation value for the sapphire dielectric substrate. It should be noted that it is very hard to directly compare the thermal resistance due to the complex heat conduction mechanism across metal-insulator interface, involving energy conversion and coupling among different energy carriers in metal and phonons in dielectric insulators. However, to approximately estimate how much the temperature increases for the interface thermal conductance of 10 - 100 MW/m\textsuperscript{2}K, we performed additional calculations of the temperature rise at 60 °C. As shown in Figure S3 below, the simulated temperature rise was found to be approximately 4.4 and 3.7 °C for 10 and 100 MW/m\textsuperscript{2}K, respectively. This is because the temperature rise is mainly induced
around the VO$_2$ surface near air by the convective heat transfer to the ambient air, as shown in the cross-sectional image of temperature distribution in the inset of Figure 3 in the original manuscript.

Figure S3. Simulated temperature rise for the interface thermal conductance of 10 and 100 MW/m$^2$K
6. Current-voltage (I-V) characteristics for an epitaxial VO$_2$ nanobeam device with an external resistor of 100 kΩ.

**Figure S4.** Representative current-voltage characteristics measured consecutively at 65 °C by varying the applied voltage both in the forward- and reverse-sweep from 0 to 7 V for the VO$_2$ nanobeam device with an external resistor of 100 kΩ.
7. Current-voltage (I-V) characteristics measured at (a) 70 and (b) 55 °C

**Figure S5.** Current-voltage (I-V) characteristics measured at (a) 70 and (b) 55 °C by varying the applied voltage both in the forward- and reverse-sweep directions for a epitaxial VO\textsubscript{2} nanobeam device. The insets show I-V characteristics of the epitaxial VO\textsubscript{2} nanobeam device with R\textsubscript{ext} = 10 k\textohm measured after the application of pulse voltage.
8. Current-voltage characteristics of the VO$_2$ nanobeam device measured for the current-controlled mode under different temperatures

![Figure S6](image-url)

**Figure S6.** Current-voltage characteristics of the VO$_2$ nanobeam device with R$_{ext}$ = 10 kΩ measured for the current-controlled mode under different temperatures ((a) 70, (b) 65, (c) 55, and (d) 50 °C).
9. Raman spectra at 60 °C of the epitaxial VO₂ nanobeam

**Figure S7.** (a) Optical image of the epitaxial VO₂ nanobeams grown on a c-cut sapphire substrate. (b) Raman spectra at 60 °C of the VO₂ nanobeam in (a). The schematic drawings indicate the crystal structure of R and M2 phases.
References


