Electronic Supplementary Information

Metastable State-Induced Consecutive Step-like Negative Differential Resistance Behaviors in Single Crystalline VO_2 Nanobeams

Jung Inn Sohn,[*ab] Seung Nam Cha,*[a] *Seung Bae Son,*[c] *Jong Min Kim,*[a] *Mark E.Welland,*[b] *and Woong-Ki Hong*[*c]

[a]Department of Engineering Science, University of Oxford, Oxford OX1 3PJ, United Kingdom

[b]Nanoscience Centre, University of Cambridge, Cambridge CB3 0FF, United Kingdom

[c]Jeonju Center, Korea Basic Science Institute, Jeonju, Jeollabuk-do 54907, Republic of Korea

*Corresponding author: E-mail: (J.I.S) junginn.sohn@eng.ox.ac.uk; (W.-K.H) wkh27@kbsi.re.kr

Table of Contents

1. The parameters and related values used for theoretical simulations

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

3. Threshold voltage and current (V_{th}, I_{th}) versus the calculated temperature ($T_{cal.}$) and temperature change versus V_{th}

4. Calculated temperature rise at 60 °C for the VO_2 nanobeam with a change in thermal conductivity

5. The effect of the interface thermal conductance on temperature rises

6. Current-voltage (I-V) characteristics for an epitaxial 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

8. Current-voltage characteristics of the VO_2 nanobeam device measured for the current-controlled mode under different temperatures

9. Raman spectra at 60 °C of the epitaxial VO_2 nanobeam

1. The parameters and related values used for theoretical simulations

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

Parameters	Values	References
Heat capacity of VO_2	690 J/kgK	*J. Phys:Condens. Matter.* 12, 8837 (2000), *J. Mater. Sci.* 44, 5345 (2009)
Mass density of VO_2	4340 kg/m^3	*J. Phys:Condens. Matter.* 12, 8837 (2000), *J. Mater. Sci.* 44, 5345 (2009)
Thermal conductivity of VO_2	6.5 W/mK	*Phys. Rev.* 185, 1022 (1969)
Thermal conductivity of Al_2O_3	30 W/mK	COMSOL Multiphysics, *Nature Commun.* 2012, 3, 827
Heat transfer coefficient	10 W/m^{2}K	*J. Mater. Sci.* 2009, 44, 5345
Voltage	Threshold voltages	Experimental values extracted from Figure 2(b)
Current	Threshold currents	Experimental values extracted from Figure 2(b)

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

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

Set temperature	Maximum temperature rise	Temperature deviation
40 ºC	2.6 ºC	± 0.4 ºC
50 ºC	3.0 ºC	± 0.55 ºC
55 ºC	2.8 ºC	± 0.45 ºC
60 ºC	3.7 ºC	± 0.5 ºC
65 ºC	4.4 ºC	± 0.8 ºC
70 ºC	4.6 ºC	± 0.85 ºC

3. Threshold voltage and current (V_{th}, I_{th}) versus the calculated temperature ($T_{cal.}$) and temperature change versus V_{th}

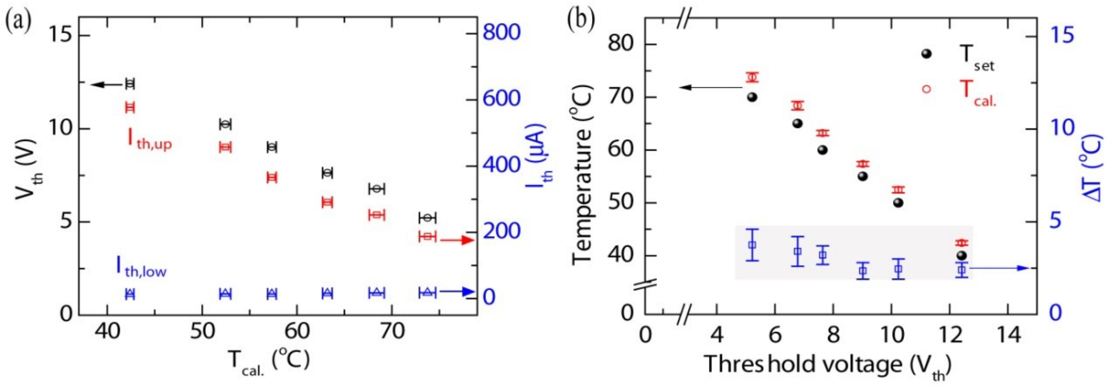

Figure S1. (a) Threshold voltage (V_{th}) and threshold current (I_{th}) as a function of the calculated temperature ($T_{cal.}$). (b) Temperature change as a function of threshold voltage. $\Delta T = T_{set} - T_{cal.}$. The gray-colored region shows temperature rise is less than 5 °C.

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 ºC 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.

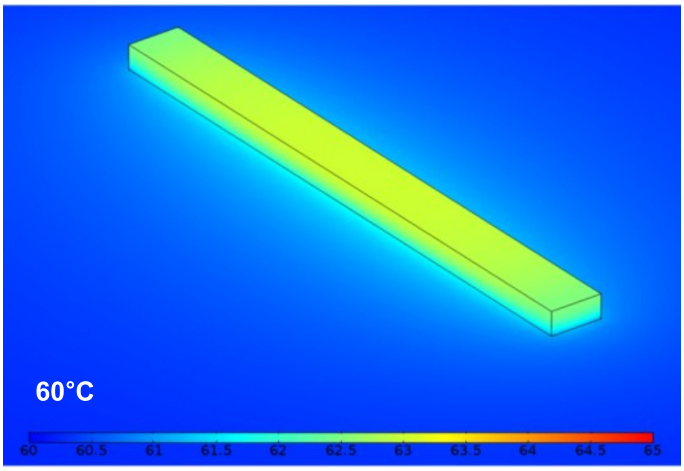

Figure S2. Calculated temperature rise at 60 °C for the VO_2 nanobeam with a change in thermal conductivity

5. The effect of the interface thermal conductance on temperature rises

We considered heat dissipation through the solid interface of the VO_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_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.*,[S4] the thermal conductance of solid interfaces between metal and dielectric is a range of 10 - 100 MW/m^2K. 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^2K, we performed additional calculations of the temperature rise at 60 ºC. As shown in Figure S3 below, the simulated temperature rise was found to be approximately 4.4 and 3.7 ºC for 10 and 100 MW/m^2K, 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.

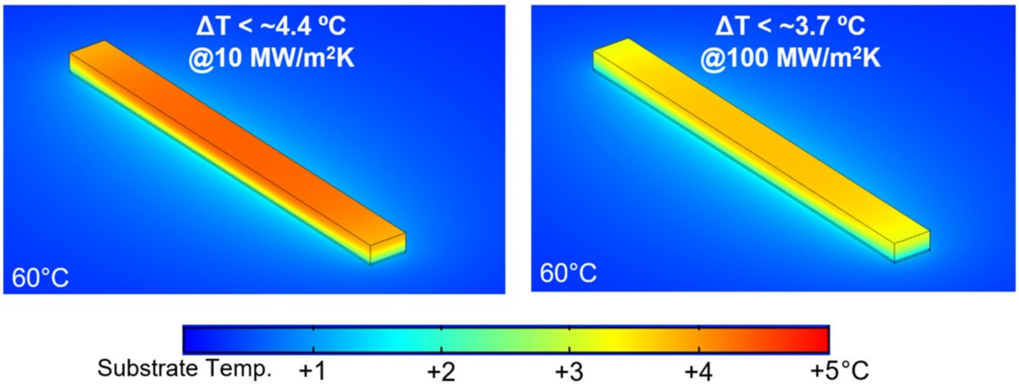

Figure S3. Simulated temperature rise for the interface thermal conductance of 10 and 100 MW/m^2K

6. Current-voltage (I-V) characteristics for an epitaxial VO_2 nanobeam device with an external resistor of 100 kΩ

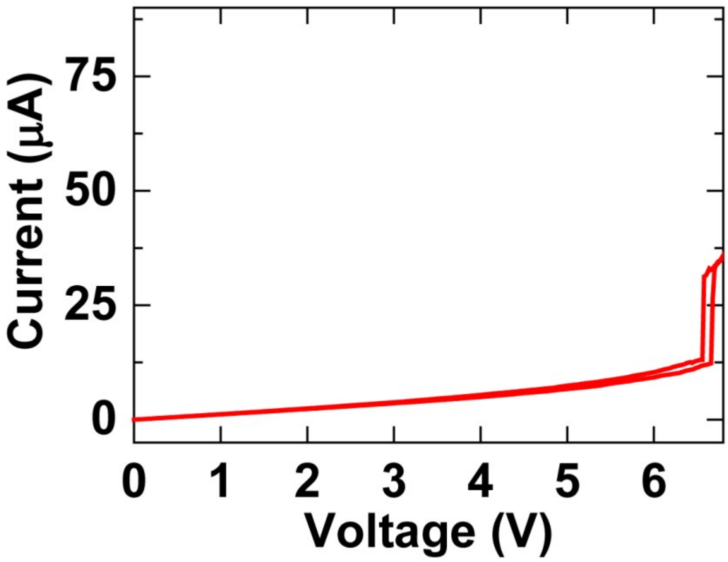

Figure S4. Representative current-voltage characteristics measured consecutively at 65 °C by varying the applied voltage both in the forward- and reverse-sweep from 0 to 7 V for the VO_2 nanobeam device with an external resistor of 100 kΩ.

7. Current-voltage (I-V) characteristics measured at (a) 70 and (b) 55 °C

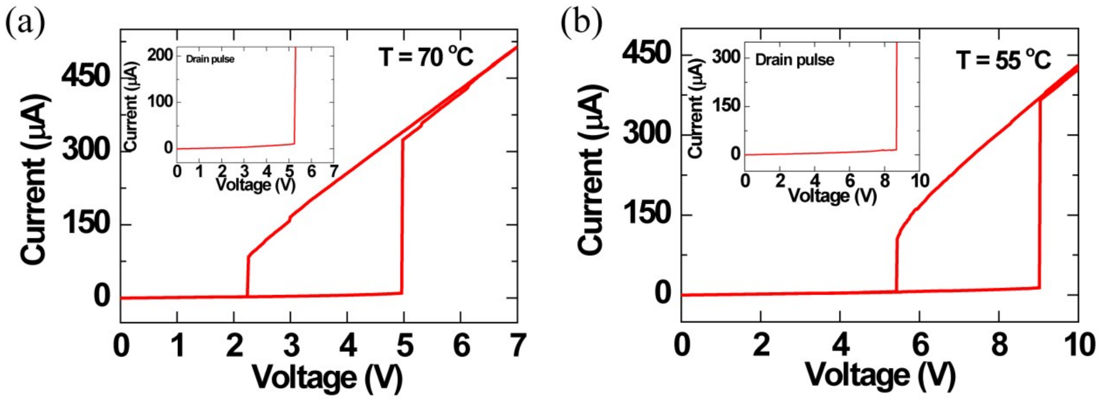

Figure S5. Current-voltage (I-V) characteristics measured at (a) 70 and (b) 55 °C by varying the applied voltage both in the forward- and reverse-sweep directions for a epitaxial VO_2 nanobeam device. The insets show I-V characteristics of the epitaxial VO_2 nanobeam device with R_{ext} = 10 kΩ measured 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

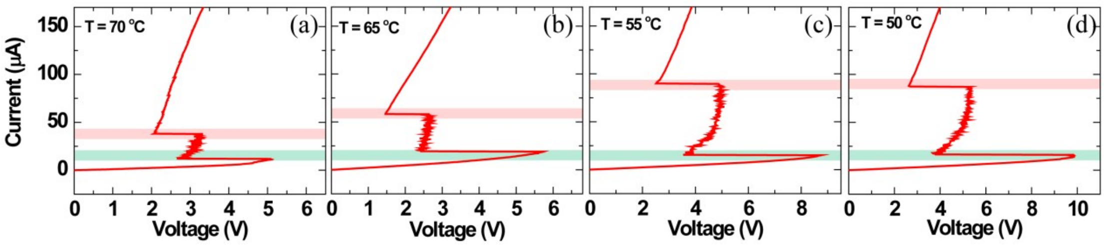

Figure S6. Current-voltage characteristics of the VO_2 nanobeam device with R_{ext} = 10 kΩ measured for the current-controlled mode under different temperatures ((a) 70, (b) 65, (c) 55, and (d) 50 °C).

9. Raman spectra at 60 °C of the epitaxial VO_2 nanobeam

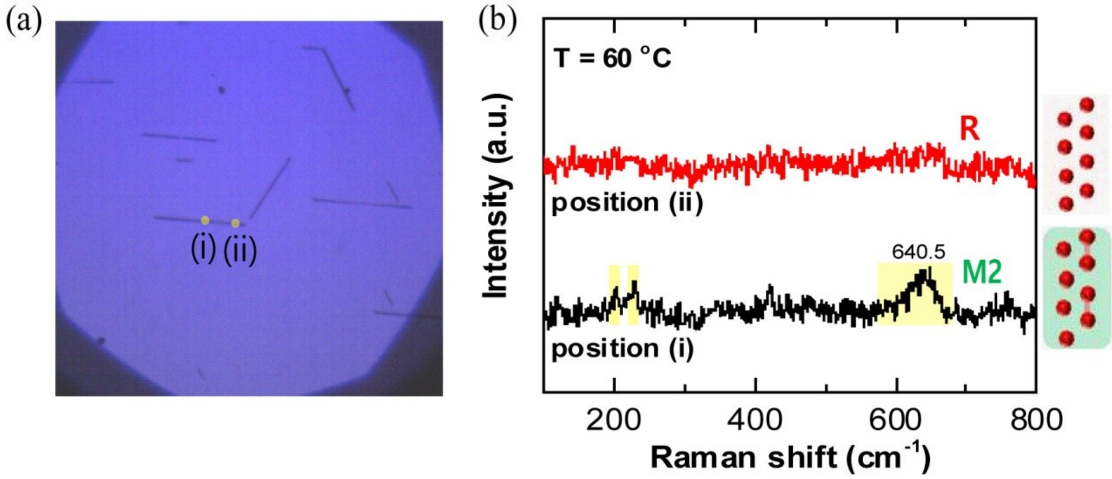

Figure S7. (a) Optical image of the epitaxial VO_2 nanaobeams grown on a c-cut sapphire substrate. (b) Raman spectra at 60 °C of the VO_2 nanobeam in (a). The schematic drawings indicate the crystal structure of R and M2 phases.

References

[S1] C. N. Berglund and H. J. Guggenheim, *Phys. Rev.* 1969, **185**, 1022.

[S2] S. Lee, K. Hippalgaonkar, F. Yang, J. Hong, C. Ko, J. Suh, K. Liu, K. Wang, J. J. Urban, X. Zhang, C. Dames, S. A. Hartnoll, O. Delaire, J. Wu, *Science* 2017, **335**, 371.

[S3] D. -W. Oh, C. Ko, S. Ramanathan, D. G. Cahill, *Appl. Phys. Lett.* 2010, **96**, 151906.

[S4] H. -K. Lyeo and D. G. Cahill, *Phys. Rev. B* 2006, **73**, 144301.