Controlling the resistive switching hysteresis in VO_2 thin films via application of pulsed voltage

O. Murtagh,[1, a)] B. Walls,[1] and I. V. Shvets[1]
School of Physics and Centre for Research on Adaptive Nanostructures and Nanodevices (CRANN), Trinity College Dublin, Dublin 2, Ireland

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We investigate the origin of the variation in resistive switching hysteresis of VO_2 thin films. Using pulsed electrical measurements in textured VO_2 thin film devices, we show that the hysteresis observed in I-V curves results from Joule heating effects, particularly in the low-resistance state. The hysteresis is reduced by increasing the cooling time between pulses. Based on a mechanism of Joule heating-induced metal-insulator transition, numerical simulations are performed which agree with the experimental variation in the hysteresis. Finally, a framework for engineering the I-V curves of VO_2 devices is proposed.

The metal-insulator transition (MIT) is a topic of long standing interest in condensed matter physics and materials sciences.[1,2] This transition deals with the increase in conductivity of a material with external stimulus such as heat,[1] light,[3] strain[4] or electric field.[5] Systems exhibiting MIT behaviour include organic compounds[6] and transition metal oxides.[7,8] Along with the resistivity; optical[9] and magnetic properties[10] can also undergo a sharp change through these transitions. Generally, the control parameters for the MIT can be classified into three categories: temperature control; bandwidth control[4] (e.g. strain); and band-filling control[11] (e.g. doping or applied fields), although more specific classifications can be found in this article by Imada *et al*.[12]

The use of an electric field to induce a change in resistivity is referred to as resistive switching (RS), and has attracted much attention due to the potential applications in electronics such as oscillators, neuromorphic devices and memory.[13,14] For many RS systems, their I-V curve shows some level of hysteresis, i.e. the current does not retrace itself in voltage sweeps up and down.[6,15]

The V-O system is a family of compounds with a large number of phases, many of which undergo a MIT.[16] Of particular focus in this study is vanadium dioxide (VO_2), with a MIT transition temperature (T_{MIT}) of 340K. Over a range of 0.1K, single crystal VO_2 undergoes a structural transition, accompanied by a 5 order of magnitude change in conductivity.[17] There is also a intrinsic thermal hysteresis of $\sim$1K.[18] VO_2 has also been shown to exhibit RS when external electric fields of 6.5×10^7 V m^{-1} are applied across it.[19] For thin films of VO_2, the amplitude of the transition decreases, while the hysteresis and range over which the transition occurs increases, as described by structural effects such as phase propagation and grain size.[20] VO_2 RS takes place at either voltage polarity with well-pronounced hysteresis.[21] In many examples, the power required to trigger the MIT in the insulating state is equal to the power required to maintain the metallic state before it returns to the insulating phase.[22]

The mechanism for the RS is debated, with arguments made for temperature control via Joule heating, and band-filling control via carrier injection.[23] Experiments using different methodologies have isolated these mechanisms,[24,25] but in most cases both simultaneously contribute to the RS. Understanding the relative contributions of the mechanism in any given system is important. Furthermore, understanding the dynamics of each mechanism - for example temperature change induced by Joule heating - can provide insight into morphology, structure and phase changes, which is vital for device optimisation. Ideally, in electronic applications, heat fluctuations and power dissipation should be kept to a minimum to reduce energy consumption and mitigate adverse heating effects, while the ideal profile of the hysteresis varies between applications.[14] The ability to tune this I-V profile is therefore an important consideration in RS devices.

In this study, we determine the origin of hysteresis in RS of our VO_2 thin films. The influence of pulsing on hysteretic behaviour and the temperature profile is discussed. Through both continuous and pulsed voltage measurements we show that the hysteretic behavior originates from local Joule heat-

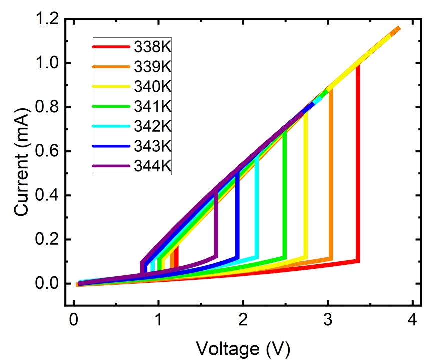

FIG. 1. The IV curves of the VO_2 device at different temperatures under continuous voltage sweeping conditions. Voltage here is the drop across the entire circuit (V_s). It it clear that the switching voltage (V_{sw}^{on}) reduces with increasing temperature as expected for the Joule heating picture.

a)Electronic mail: murtagho@tcd.ie

ing of the channel once the system is switched to the low-resistance state. We further demonstrate how pulsing - in particular the time between pulses - can change the hysteresis. A thermal model, predicting channel temperature using only Joule heating and environmental cooling, agrees with the data and predicts substantial temperature fluctuations between the metallic and insulating states. The model demonstrates that one needs to consider heating and thermal conductivity beyond the VO_2 material itself.

Textured VO_2 thin films were grown on c-plane sapphire substrates via pulsed laser deposition to 30nm thickness. A KrF laser (248nm wavelength) and a pure vanadium target were used. Distance from target to substrate was 7cm. During growth the substrate was held at 600°C, base pressure was approximately 1×10^{-5}mbar, O_2 pressure was held at 15×10^{-3}mbar and the laser energy used was 170mJcm^{-2}. The films were characterised by x-ray diffraction and resistance measurements, showing the VO_2<010> (Figure S1) family of reflections[26,27] and an approximately 3 order of magnitude resistance change across the MIT. Films grown by this method have been shown to be granular.[28] Sheet resistance measurements were performed using a 4 point probe and a ceramic heating stage. The sheet resistance was measured to be 120kΩ at 330K, 16kΩ at 342K and 150Ω at 360K. Due to hysteresis, upward and downward switching temperatures were 350K (T_{MIT}^{on}) and 345K (T_{MIT}^{off}), characteristic of thin film VO_2.[29] The wider hysteresis is evidence of a granular film of slightly mixed phase causing a percolative effect, while the high switching temperature suggests a slight oxygen excess as compared to single crystal VO_2 (Figure S2).[20,30]

Devices for two-terminal measurements were prepared using photolithography. VO_2 channels of length 4μm and width 8μm were fabricated using a CF_4 RIE etch followed by deposition of layered Ti/Au contacts of thickness 5nm/30nm via electron beam evaporation. An image of the device can be seen in Figure S3. Different devices showed variations in threshold switching voltages and resistance due to variations in local film structure and lithography accuracy.

Contacting the samples was performed using a JANIS probe station with a heated stage. All measurements were made in the local Dublin atmosphere. For electrical characterisation, input waveforms were created using the LeCroy ArbStudio 1102 arbitrary signal generator. A resistor (560 Ω) was placed in series with the selected VO_2 device and the Digilent Analog Discovery 2 oscilloscope was used to collect voltage data from the signal generator and across the resistor. The circuit is described in Figure S3. This arrangement allowed a statistically relevant number of measurement to be taken quickly. Two different measurements were performed: first, a continuous sweep of the voltage up and down at; and second, a pulsed sweep implemented with controlled pulse parameters. These measurements were performed on different devices.

At stage temperatures below the transition temperature, the upward continuous sweeping I-V curve of the VO_2 device shows a discontinuity at a critical voltage (V_{sw}^{on}) as the material enters the metallic state and the current jumps. The closer the stage temperature is to the transition temperature, the less voltage is required (Figure 1). As the voltage is lowered, the device will return to an insulating state at some V_{sw}^{off}. For a continuous voltage sweep, V_{sw}^{off} is always lower than V_{sw}^{on} resulting in a well-defined hysteresis.

In the pulse experiments voltage pulses 10μs in length were applied with varying gaps of 0V between pulses, from 100μs to 32ns. The input waveform was generated by multiplying this pulsed wave with a triangular wave. The beginning of each pulse occurs at voltage intervals of 0.01V (Figure S4). For this device, the high and low resistance states of the VO_2 device were found to be between 10-15 kΩ and 60-260 Ω respectively measured via the I-V curves. This variance is believed to be due to conduction filaments[31] within the VO_2 channels changing or rerouting as the experiment progressed.

Two measured quantities from the experiment are total voltage dropped over the circuit (the source voltage, V_s) and the voltage drop over the resistor (V_R). The current through the circuit is calculated via $I = \frac{V_R}{R}$ where R is the resistance of the resistor, while the voltage drop across the device $V_D = V_s - V_R$.

Figure 2 shows I-V curves of the VO_2 device using V_D. This data is used to calculate the potential and power dissipated across the isolated VO_2 device. Figure 3 shows the relationship between gap length, V_{sw}^{off} and V_{sw}^{on} for the VO_2 device (using V_D). 100 measurements were taken for each gap length. Each data point represents the average and standard deviation of these measurements. Measurements were take with a stage temperature of 342K. Two trends are clear from Figure 3. First, V_{sw}^{on} slightly increases with increasing gap time. Second, V_{sw}^{off} increases, with increasing gap time. More importantly, $\Delta V \; (= V_{sw}^{on} - V_{sw}^{off})$ decreases as the gap time increases.

In this system, it's demonstrated that both V_{sw}^{on} and V_{sw}^{off} are dictated by the self heating and cooling of the VO_2 channel device. This will be demonstrated qualitatively before a quantitative description using a simple model of channel temperature.

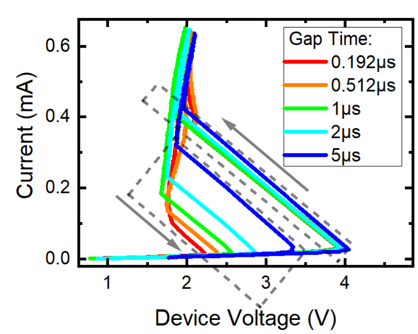

FIG. 2. The IV curves of pulsed measurements using device voltage (V_D). Dashed boxes represent regions of V_{sw}^{On} and V_{sw}^{Off} for varying gap time. As gap time increases, V_{sw}^{On} slightly increases while V_{sw}^{Off} increases faster, reducing the hysteresis.

The sample begins at the set temperature (T_{set}: the stage temperature). In the continuously applied voltage regime, power I × V is applied to the channel, heating it. Any heat added will be conducted away by the channel, contacts and the highly thermally conductive sapphire substrate.[32] At V_{sw}^{on}, input power will be enough to bring the channel temperature (T_{ch}) to T_{MIT}^{on} (power = P_{on}). The resistance of the channel will drop and the current will increase. In this state the power applied is much higher, heating the channel. The channel will now stay in a metallic state until the applied power is low enough (P_{off}) to allow the channel to cool to T_{MIT}^{off}. Because the device is in a low resistance state, this will be at a much lower voltage (V_{sw}^{off}). Note, in this case $P_{on} \approx P_{off}$.

However, if a pulsed voltage is used, the channel is able to cool between consecutive pulses. Therefore, more power will be required to heat the channel to T_{MIT}^{on}, and for longer gaps the V_{sw}^{on} will increase as observed. Similarly, the device will switch back to an insulating state once T_{ch} reaches T_{MIT}^{off}. For a very large gap between pulses, this would occur at a voltage very close to V_{sw}^{on} as no heat would be retained in the channel between pulses. However as the gap decreases, ΔV will grow as heat is retained in the device, lowering the voltage required for T_{ch} to reach T_{MIT}^{off}. This will be the trend until the gap disappears and the system is in the continuous voltage regime again. Thus, the experimentally observed changes in switching voltage in relation to the gap between pulses (Figure 3) is consistent with the heating picture.

As qualitatively described above, V_{sw}^{on} and V_{sw}^{off} are related to the current local temperature of the channel. To describe the movement of V_{sw}^{off} position in sweeps with different pulse parameters, a model is used to estimate the effective channel temperature as a function of applied electrical power. This model is used in a similar experiment by Fursina *et al*.[33] Detailed thermal modeling is very challenging due to the highly local character of the heating and difficulty of capturing all relevant heat transfer processes in these nano-structures. Instead a simple model where return to equilibrium is described by a

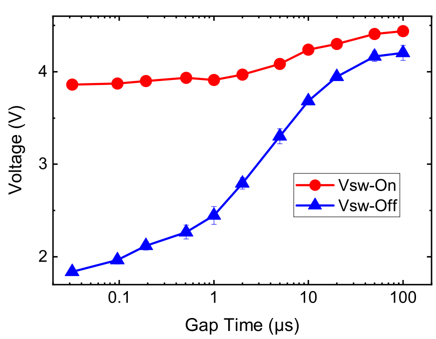

FIG. 3. This graph illustrates the dependence of V_{sw}^{on} and V_{sw}^{off} on the gap time. The hysteresis of the device switching increases when the gap time is reduced.

relaxation time, τ.[34] τ is defined by the temperature change over time with no applied heating:

$$\frac{\partial T}{\partial t} = -\frac{T - T_{set}}{\tau} \tag{1}$$

Where T_{set} is the stage temperature. τ is dependent on the thermal conductivity of the device and substrate. The heat flux into the sample is given by $dQ_{in}/dt = V \times I$. We further assume that the heat dissipation can be described as:

$$\frac{dQ_{out}}{dt} = -C_v \frac{T - T_{set}}{\tau} \tag{2}$$

Note this model assumes heat capacity C_v and τ are independent of temperature.[35] The temperature variation with respect to time t considering total heat flux (heating and cooling) of the system is given by:

$$C_v \frac{\partial T}{\partial t} = \frac{\partial}{\partial t}(Q_{in} + Q_{out}) = V \times I - C_v \frac{T - T_{set}}{\tau} \tag{3}$$

Solving Eq. (3) an expression for channel temperature is given by:

$$T(t+dt) = T_{set} + \frac{V \times I \times \tau}{C_v}\left[1 - exp\left(-\frac{dt}{\tau}\right)\right] + [T(t) - T_{set}]\, exp\left(-\frac{dt}{\tau}\right) \tag{4}$$

There are two fitting parameters in this model that predict the temperature; heat capacity C_v and relaxation time τ. Equation (4) was used in a python script to model the channel temperature and calculate the fitting parameter values. V_D and I values for each time step were taken from the measured data. The fitting parameters were adjusted so that the temperatures reached at V_{sw}^{on} and V_{sw}^{off} corresponded to T_{MIT}^{on} and T_{MIT}^{off} respectively.

An example of the temperature calculated by Eq. (4) is shown in Fig. 4. The orange line traces the maximum temperatures reached during pulses (voltage on), while the red line traces the minimum temperatures reached between pulses (voltage off). At V_{sw}^{on} the power jumps as the device becomes metallic and this leads to an increase in the temperature. The opposite occurs at V_{sw}^{off}. It is worth noting from Figure 4 how erratically the channel temperature changes between pulses, rapidly heating and cooling while reaching a maximum temperature of 430K. This is similar but lower in magnitude to temperatures calculated in similar experiments,[36] with difference attributed to the lower powers used here. The adjusted optimum values of C_v and τ are 3.6×10^{-11} J/K and 1.8×10^{-6} s respectively. All pulse measurement simulations match V_{sw}^{on} and V_{sw}^{off} with respective temperatures T_{MIT}^{on} and T_{MIT}^{off} within 1K.

Using the Eq. (4) the voltage hysteresis was simulated as a function of gap length, and is depicted in Figure 5. The dependence of the hysteresis magnitude on the gap time is

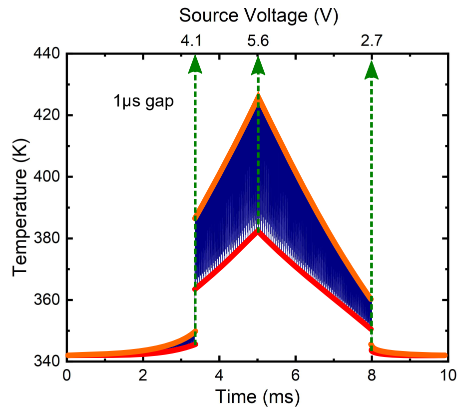

FIG. 4. The heat simulation of the device with an input waveform from 0-5.6V, a pulse width of 10μs and a 1μs gap time. The dark blue area is the fluctuating temperature data. The orange and red line represent the temperature envelope, orange being the upper temperature profile, and red being the lower. Switching on occurs when the orange line hits T_{MIT}^{On} and off when the red line reaches T_{MIT}^{Off}

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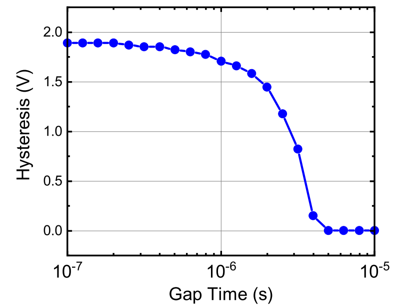

FIG. 5. A simulation of hysteresis using the model above and a simplified input dataset. The curve shape and magnitude of the hysteresis match experimental results.

reproduced, further illustrating the Joule heating mechanism and the role of the gap time in facilitating cooling and controlling the hysteresis magnitude. The change in the hysteresis is mostly due to the change in V_{sw}^{off}, in agreement with experiment.

This value of C_v can be used to approximate the size of the channel. The specific heat capacity C_v^s of VO_2 is 530-600 $JK^{-1}kg^{-1}$.[37] Using the equation;

$$C_v^s = \frac{C_v}{m} = \frac{C_v}{\rho V} \tag{5}$$

and using the estimated heat capacity, this gives a switching volume of 1.5 $\times 10^{-17}$ m^3. Given that the volume of the VO_2 channel is 9.6×10^{-19} m^3, about 16 times smaller, it can be assumed that the substrate plays a substantial role in absorbing heat in this experiment, an expected result due to the high thermal conductivity of the Al_2O_3 substrate.

In this experiment the applied field over the device never exceeds $1 \times 10^7 Vm^{-1}$, lower than the field of $6.5 \times 10^7 Vm^{-1}$ required to cause a purely electronically driven switch.[19] Because of this, it can be assumed that the dominant mechanism for the MIT is self-heating. Further evidence is seen by the shift of V_{sw}^{on} in Fig. 3, as a higher power is required to heat the channel to T_{MIT}^{on}. A purely field driven switch would see a constant V_{sw}^{on} at the critical field. Despite this, it is maintained that even in channels which solely switch via electric field, this self-heating effect is still what dominates their substantial hysteresis. For this reason, the temperature of VO_2 in the on state needs to be considered in the use of VO_2 electronics, and cooling methods to prevent the material from reaching unstable temperatures or use of voltage pulsing should be implemented to reduce achieved temperatures.

We have shown that the hysteretic behaviour seen in resistive switching of VO_2 thin films can be altered via pulsing. Increasing the gap time, or allowed cooling between pulses, raises the V_{sw}^{off} value, reducing hysteresis. This effect is shown to be thermal, agreeing with the work of Lee *et al*.[36] We have also shown that V_{sw}^{on} increases with increasing gap time, with the cooling adding to the power requirement. Increasing the period of the applied waveform, or the pulse width, would allow more heating at low voltages, decreasing V_{sw}^{on}. This agrees with the results of Lee *et al*. That work has also shown that by decreasing the resistive load in series with the device, V_{sw}^{off} and V_{sw}^{on} can be lowered without changing the hysteresis. From our model, this can be attributed to increasing the power applied to the channel, reducing the required voltage.

With this information a picture of tuning the RS curve of VO_2 can be built. V_{sw}^{on} may be raised or lowered by the period of applied voltage waveforms. The hysteresis of the device may be altered via pulsing of this waveform, and the magnitude of the switching voltages can be altered by changing the power applied to the channel.

Of course, this doesn't take into account the cooling effects of substrates or the intrinsic material properties of the VO_2 film itself. Our model shows the effect of substrate heat capacity plays a substantial role, while the effect of structural properties will change how the film reacts to changing temperatures and electric fields. Strict environmental temperature control will also be a requirement. However, this is proposed as a useful framework for engineering electronic devices using VO_2 films.

The data that support the findings of this study are available from the corresponding author upon reasonable request.

See supplementary material for the resistivity vs temperature graph of the VO_2 film, the XRD spectrum of the film and diagrams of the circuit and input waveform used.

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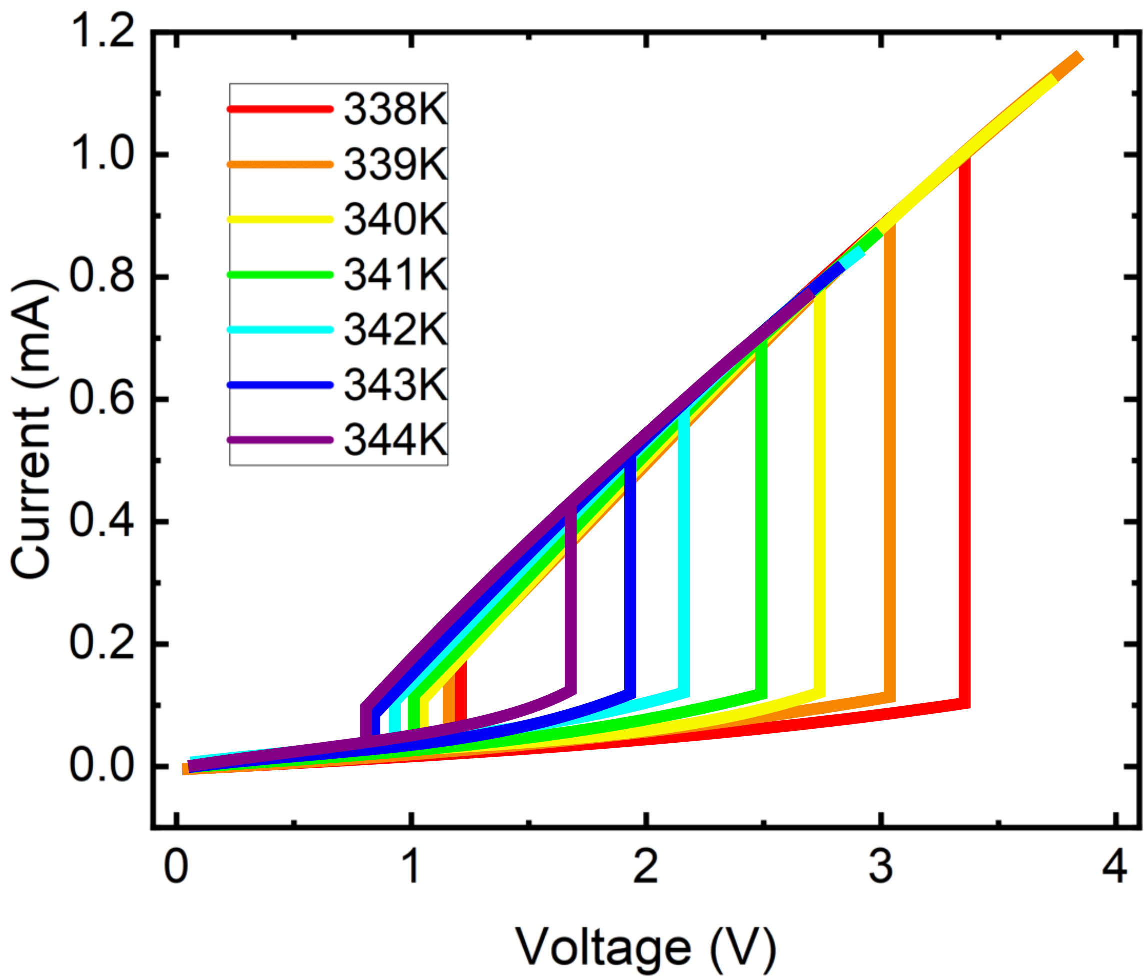
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340K
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342K
343K
344K
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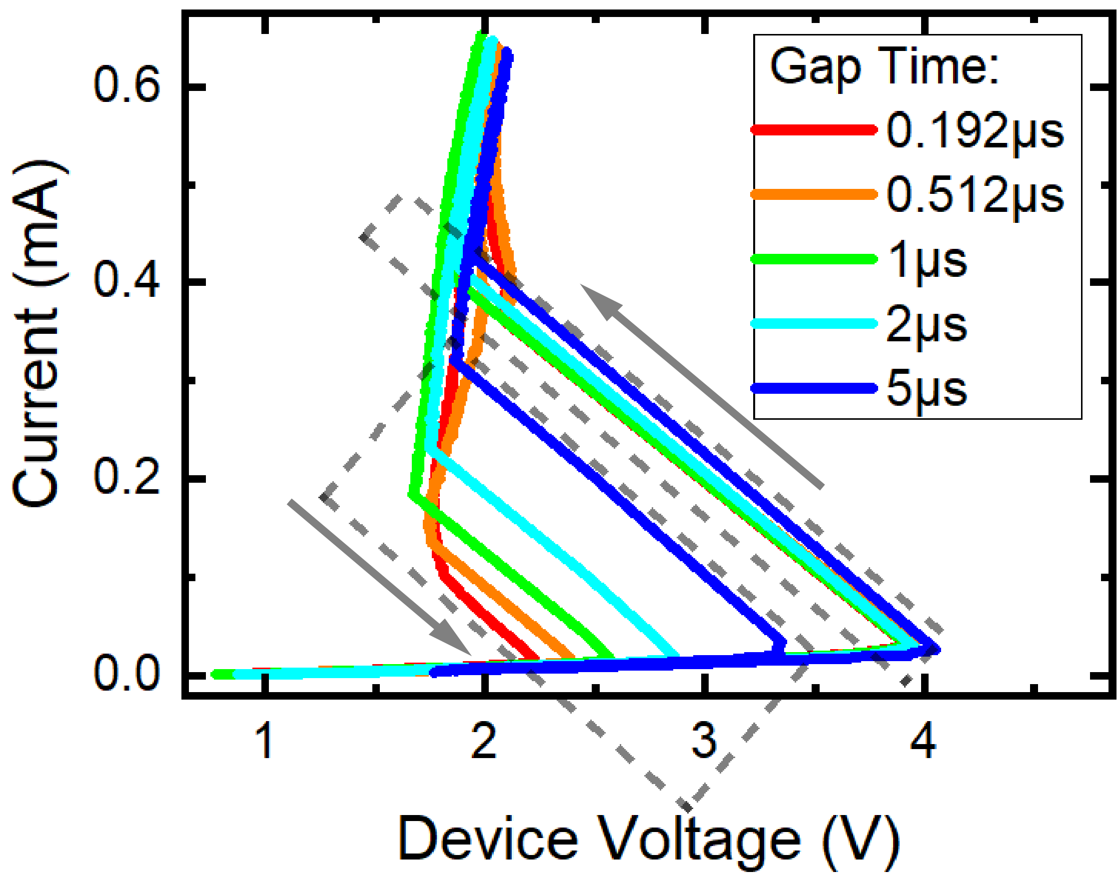
Voltage (V)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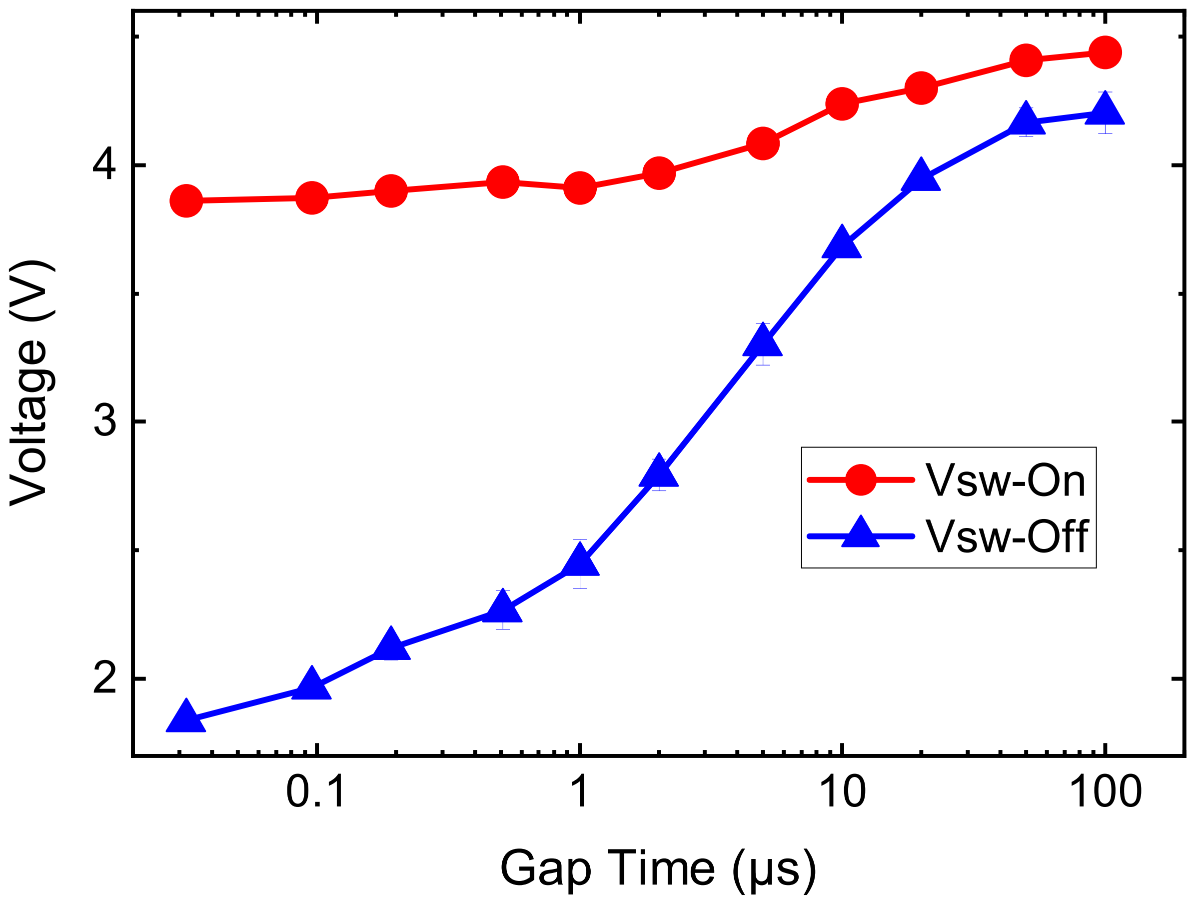
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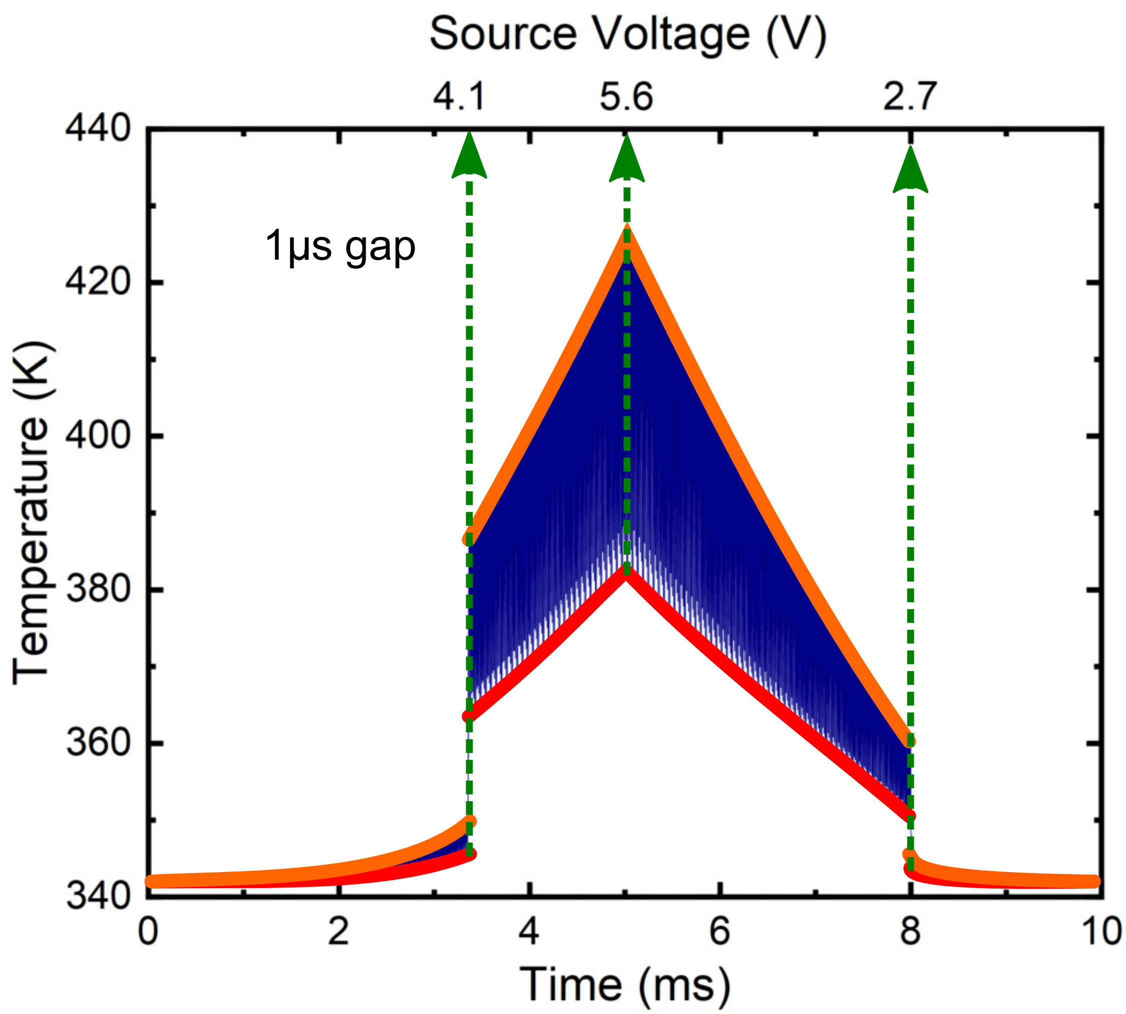
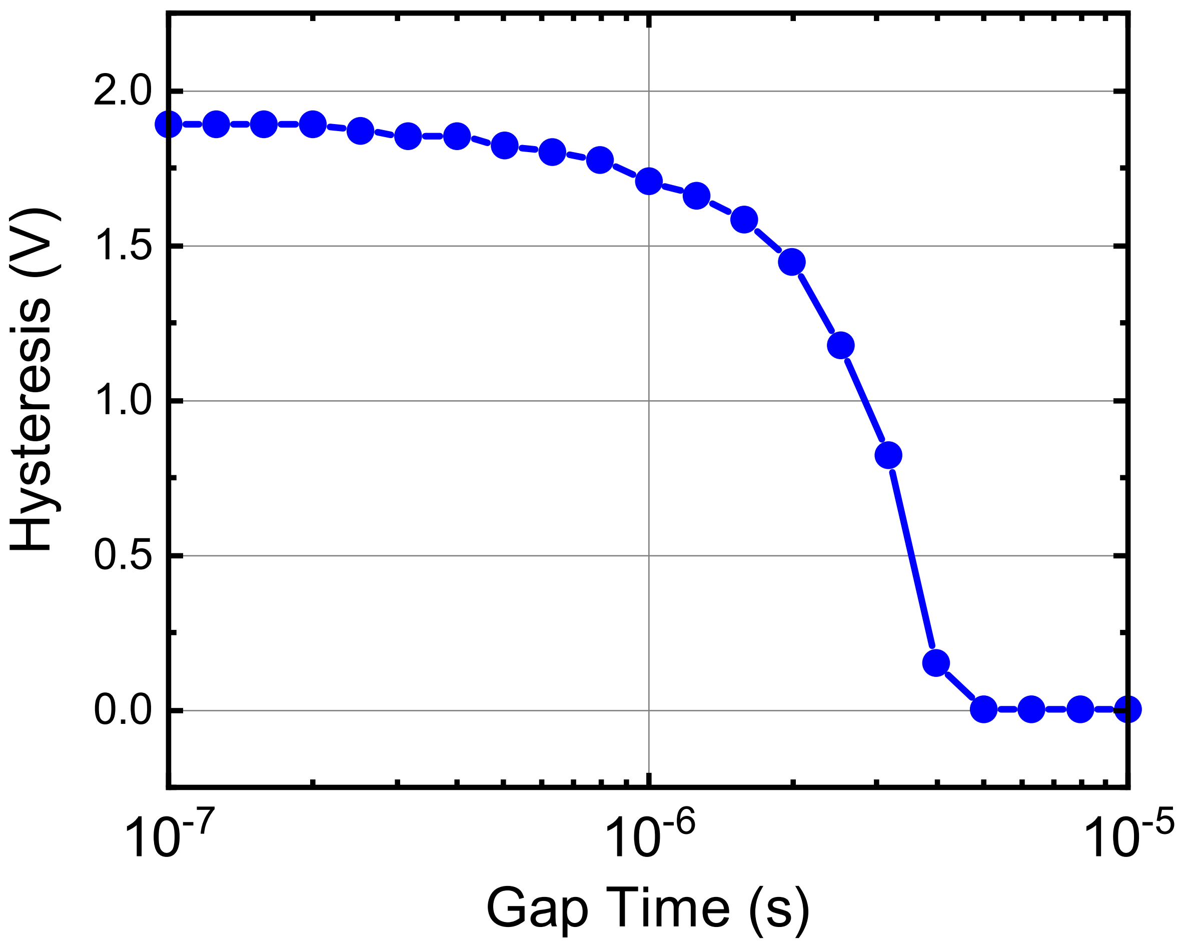
Vsw-On
Vsw-Off


Source Voltage (V)
4.1
5.6
2.7
1µs gap
Temperature (K)
440
420
400
380
360
340
0
2
4
6
8
10
Time (ms)


Hysteresis (V)
2.0
1.5
1.0
0.5
0.0
10^{-7}
10^{-6}
10^{-5}
Gap Time (s)