

Spatiotemporal characterisation of the field-induced insulator-to-metal transition

- J. Del Valle, N. M. Vargas, R. Rocco, P. Salev, Y. Kalcheim, P. N. Lapa, C. Adda, M.-H. Lee, P. Y. Wang, **L. Fratino**, M. Rozenberg, I. K. Schuller “Spatiotemporal characterization of the field-induced insulator-to-metal transition,” *Science*, vol. 373, no. 6557, pp. 907–911, 2021
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- P. Salev, **L. Fratino**, D. Sasaki, R. Berkoun, J. Del Valle, Y. Kalcheim, Y. Takamura, M. Rozenberg, I. K. Schuller, “Transverse barrier formation by electrical triggering of a metal-to-insulator transition,” *Nature communications*, vol. 12, no. 1, pp. 1–7, 2021



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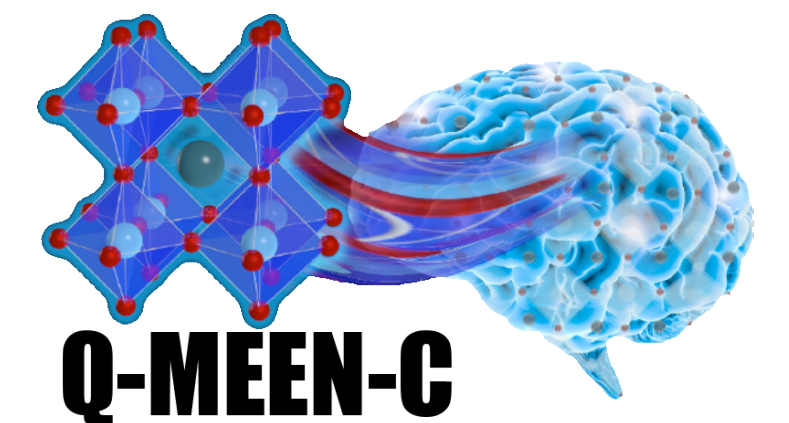
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GDR Meeticc 2023, Bordeaux



DOE-US EFRC project Q-MEEN-C (UCSD-CNRS)

Experiment:



Pavel Salev



Javier del Valle



Ivan K. Schuller

Theory:



Rodolfo Rocco

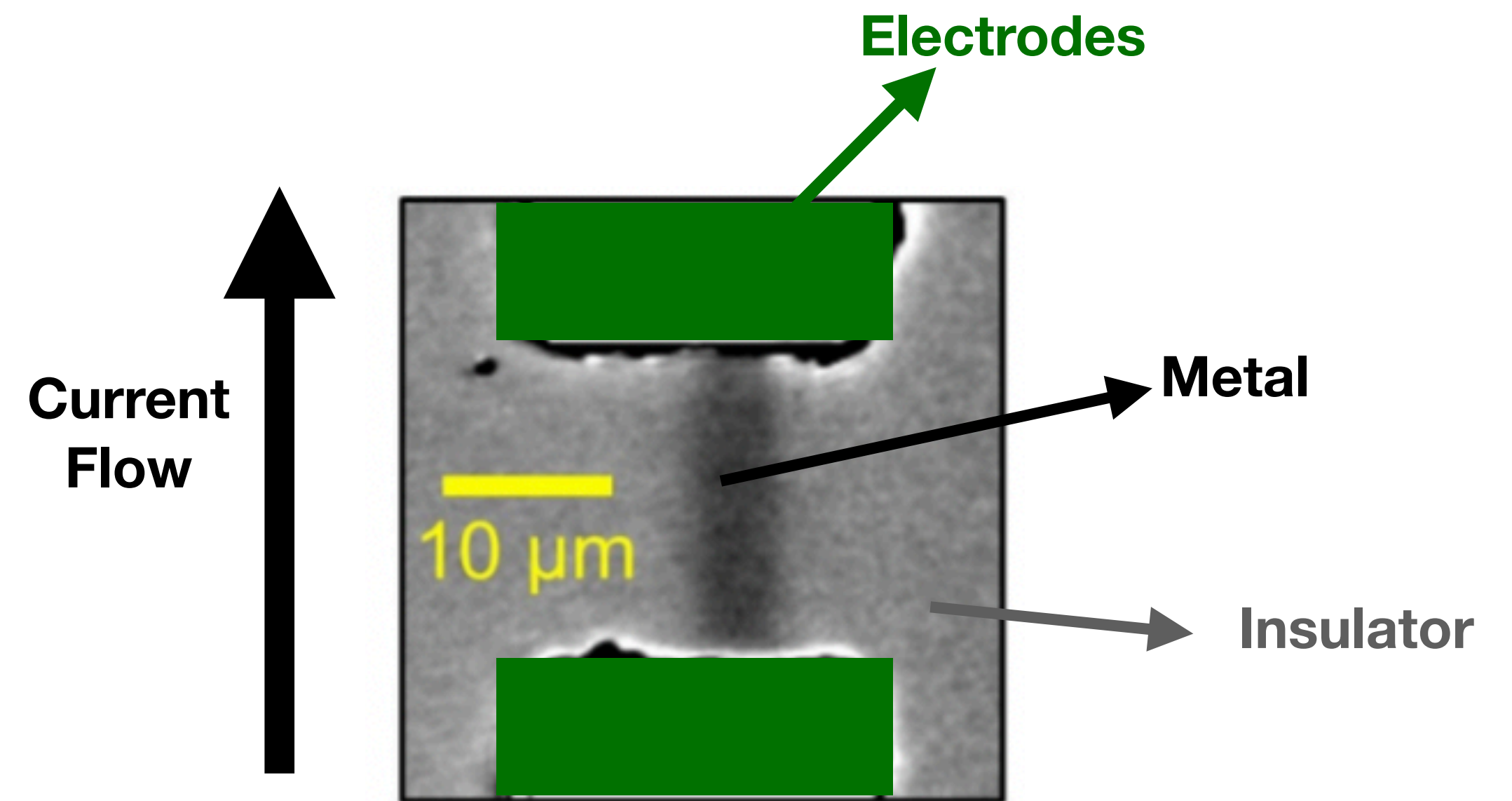
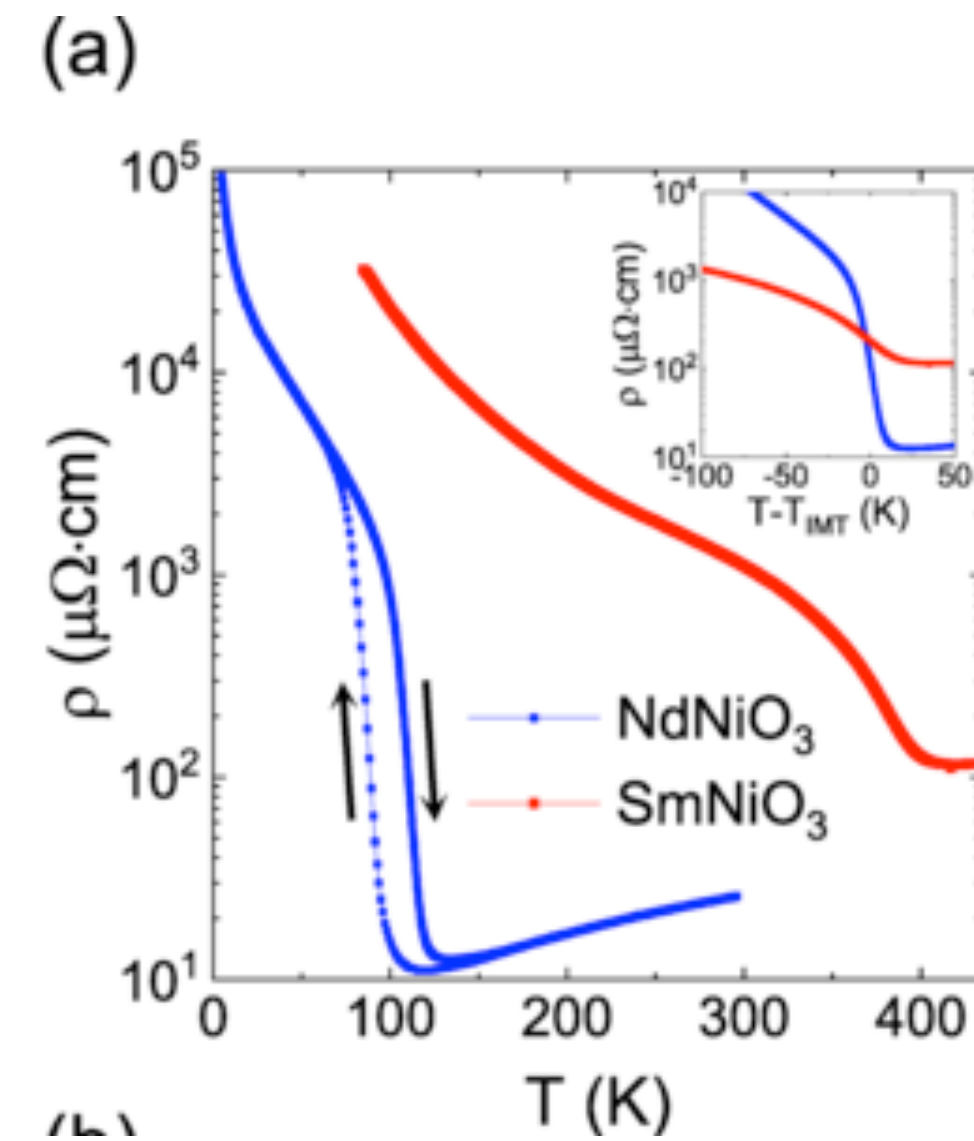
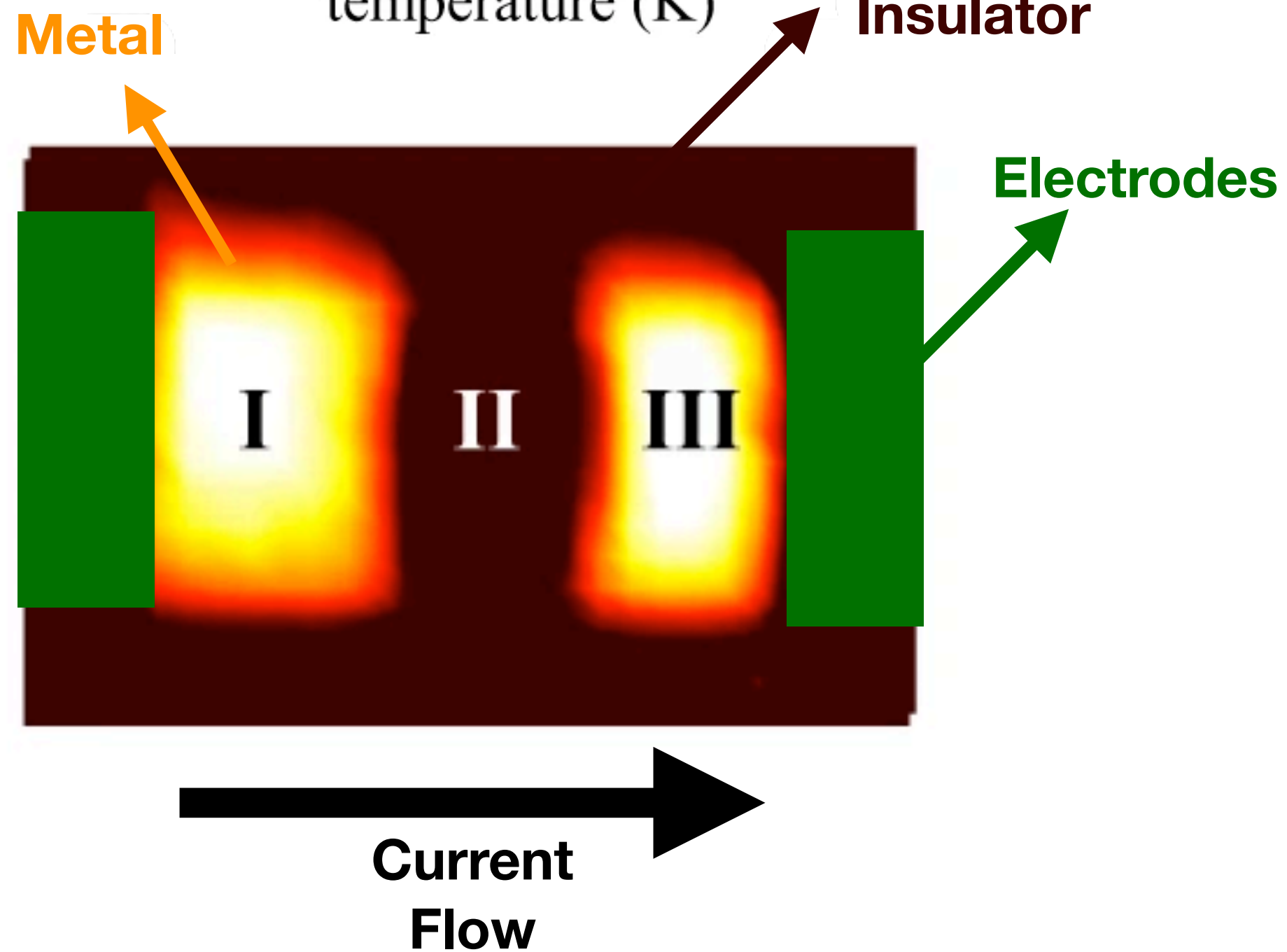
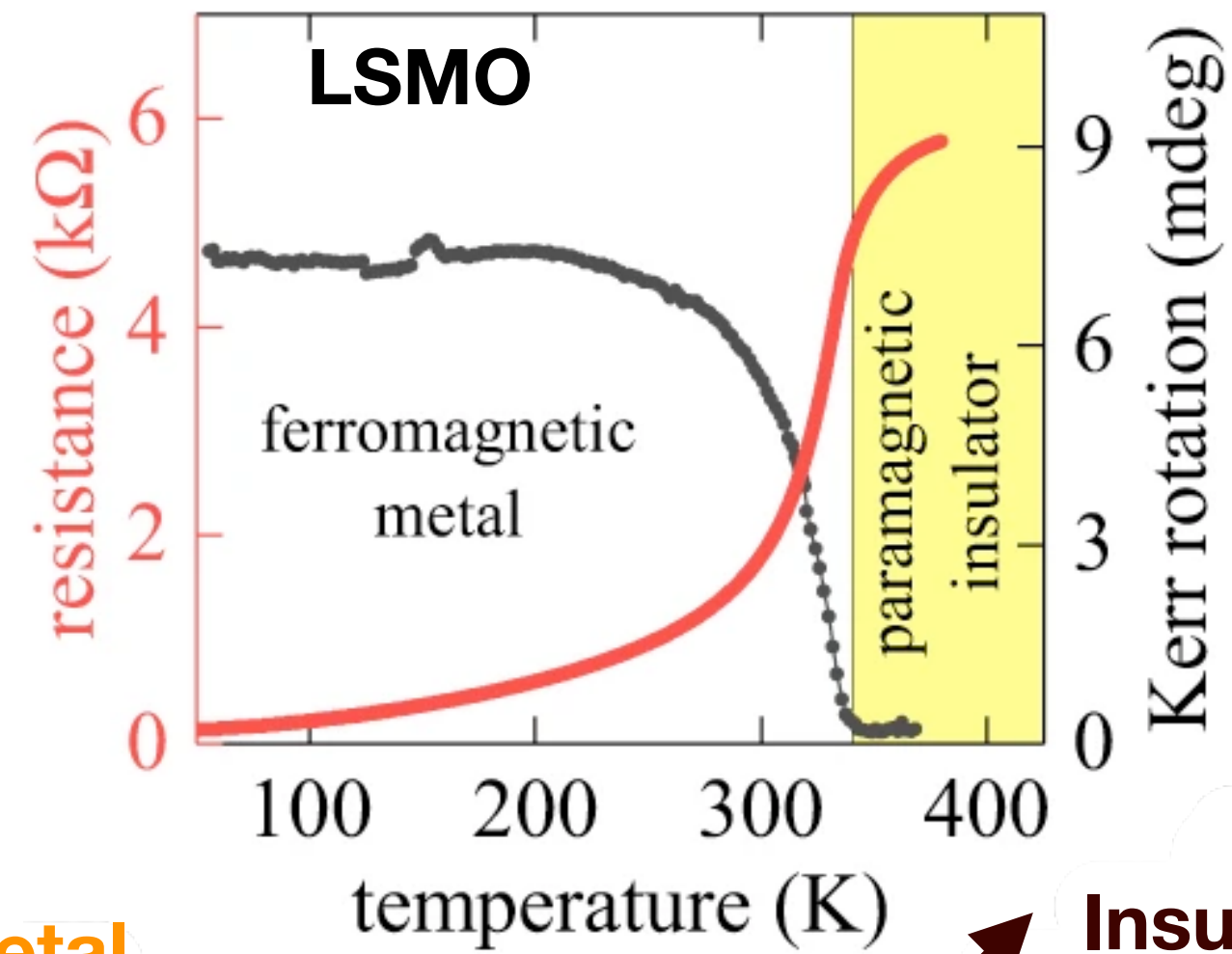


Soumen Bag

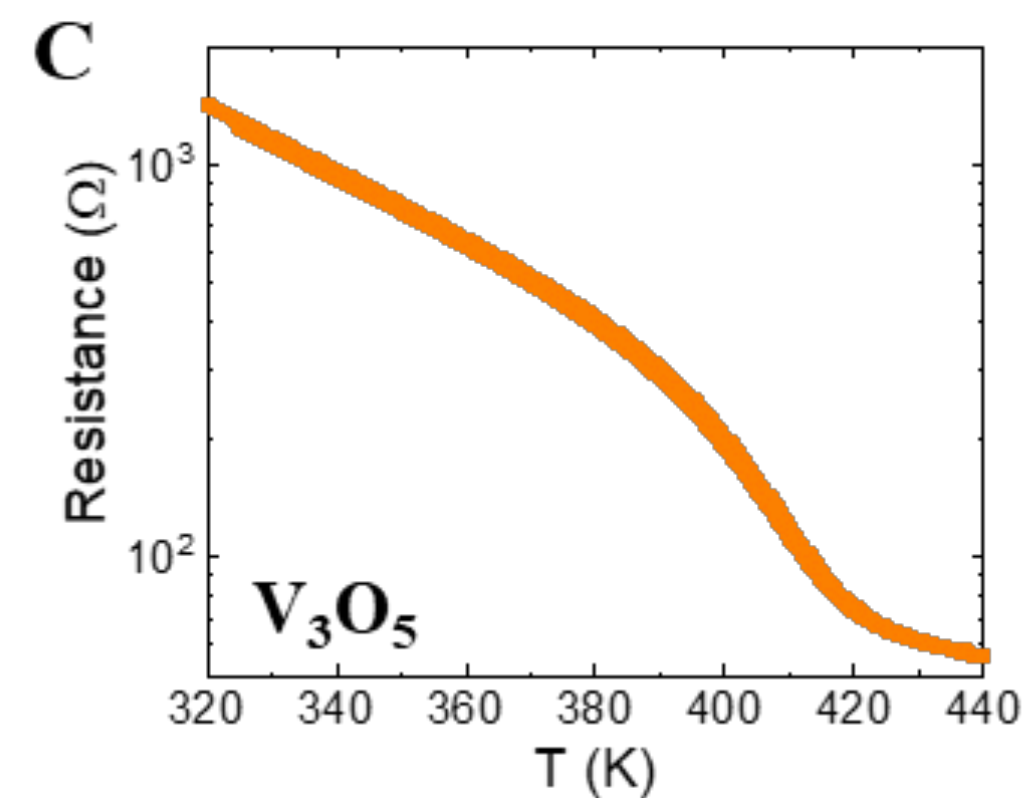
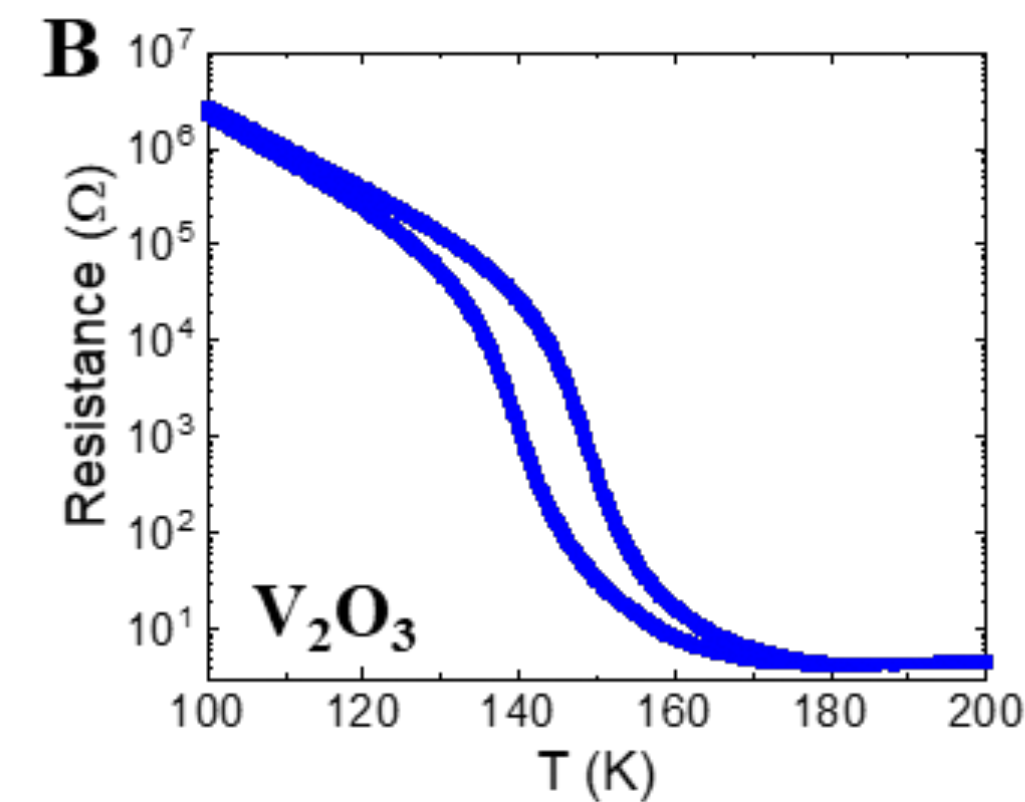
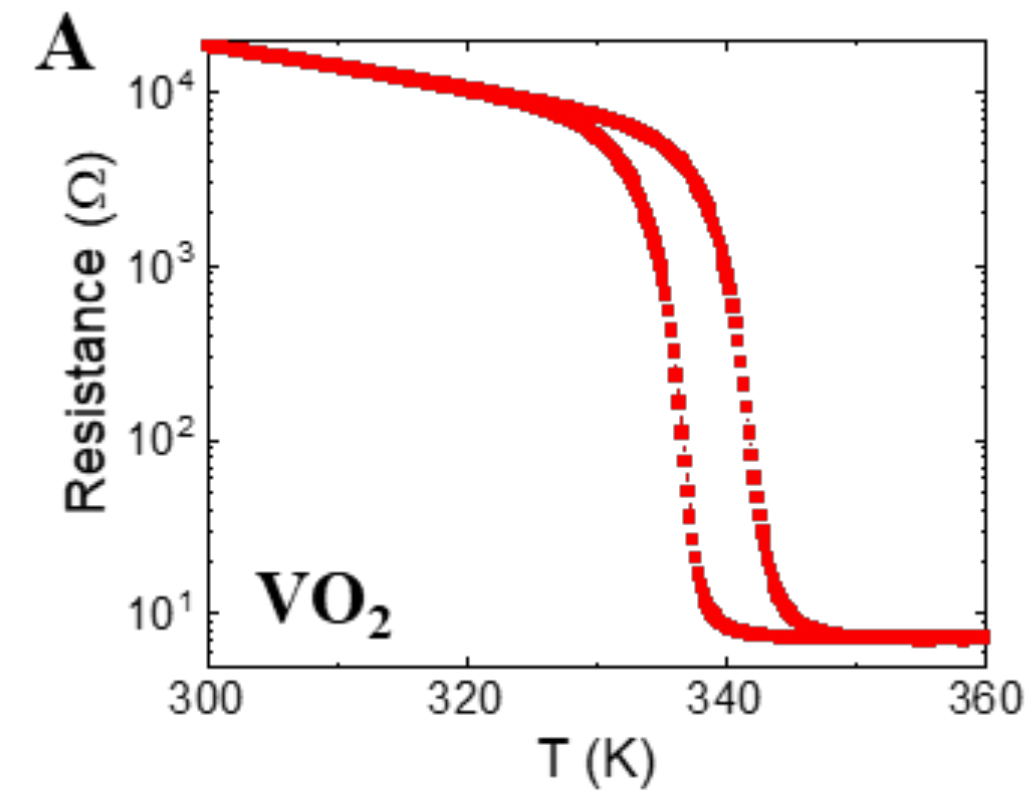


Marcelo J. Rozenberg

Resistive switching: Barrier Vs filament formation



Insulator to Metal transition



We study the transition dynamics in three well-known oxides featuring an IMT:

VO_2 (TIMT ≈ 340 K)

V_2O_3 (TIMT ≈ 160 K)

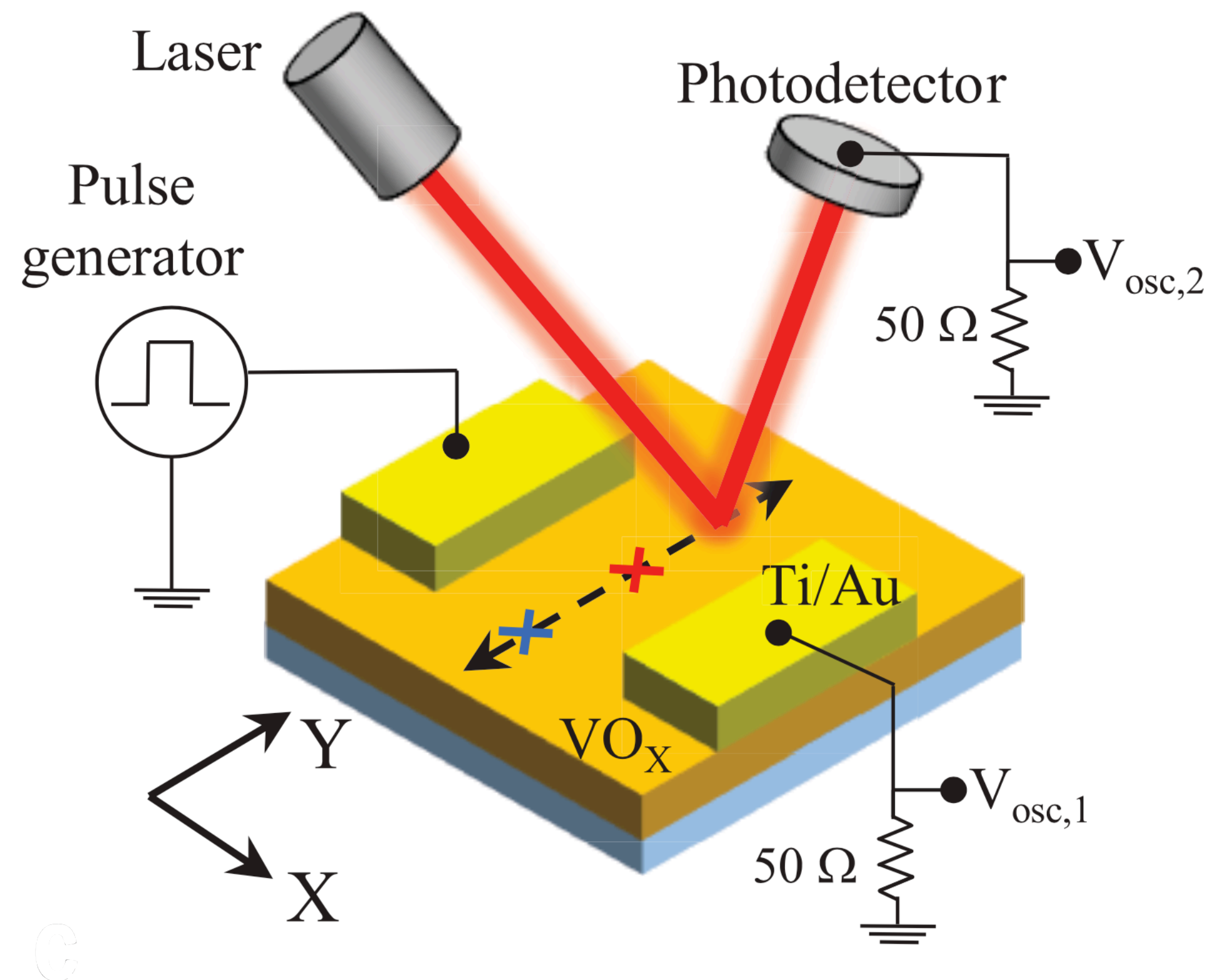
V_3O_5 (TIMT ≈ 420 K)

In V_2O_3 and VO_2 , the IMT is a **first-order transition** accompanied by an abrupt change of the crystal lattice symmetry and dimensions

In V_3O_5 , the IMT is a **second-order transition** and happens **without a substantial structural change**.

This distinction allows us to examine the importance of various factors and single out the most relevant properties governing the field-driven IMT.

Schematic representation of the measurement setup



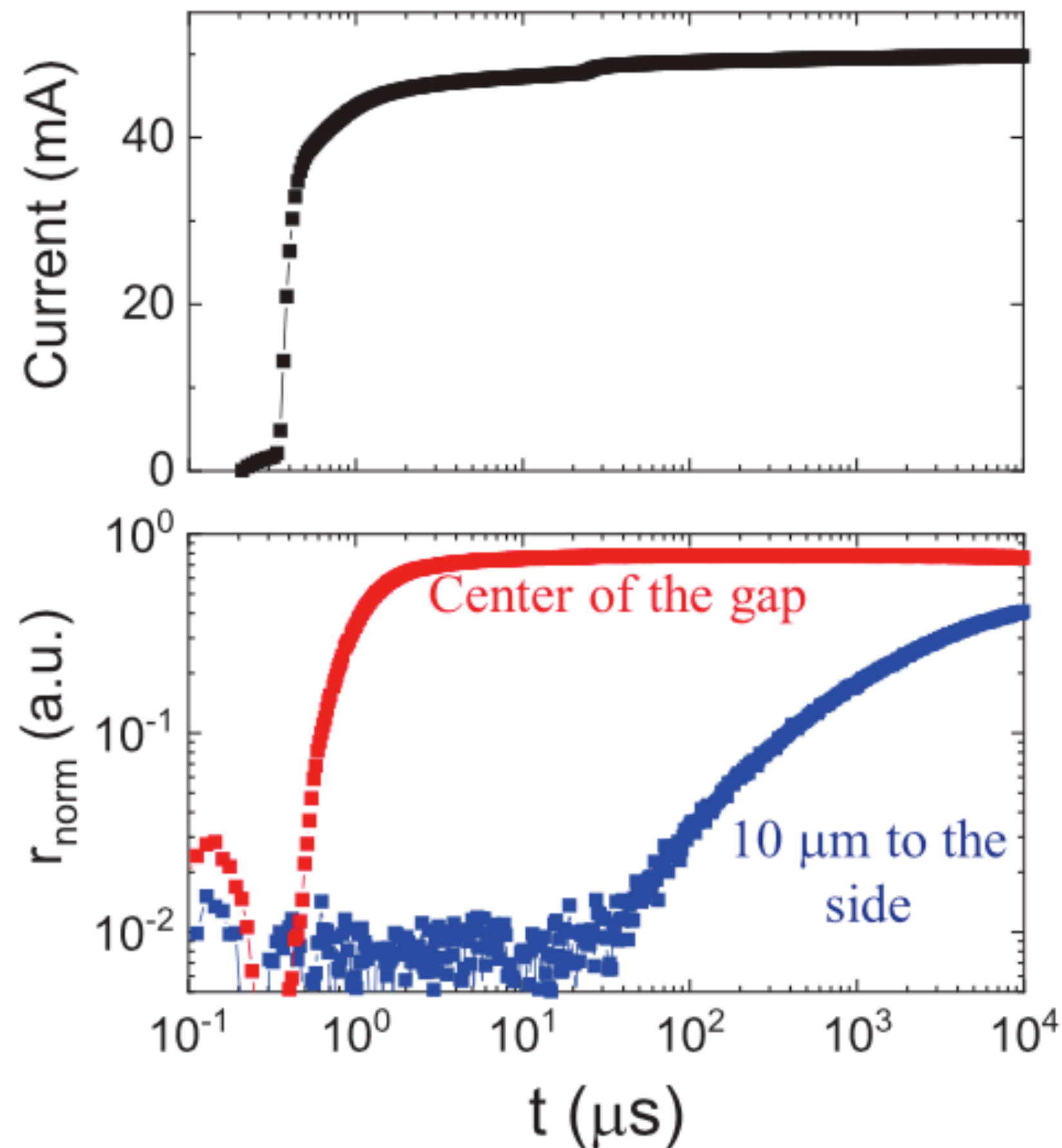
The **IMT** is induced by applying a voltage step using a function generator.

A 660-nm laser is focused to a 3-mm spot between the electrodes, and the **reflectivity** is measured with a photodetector.

Current and reflectivity are monitored with an oscilloscope.

Spatial resolution is enabled by moving the laser spot in the direction perpendicular to the current.

Measurement of the metallic phase fraction



Current versus time when a 24-V step is applied on VO₂.

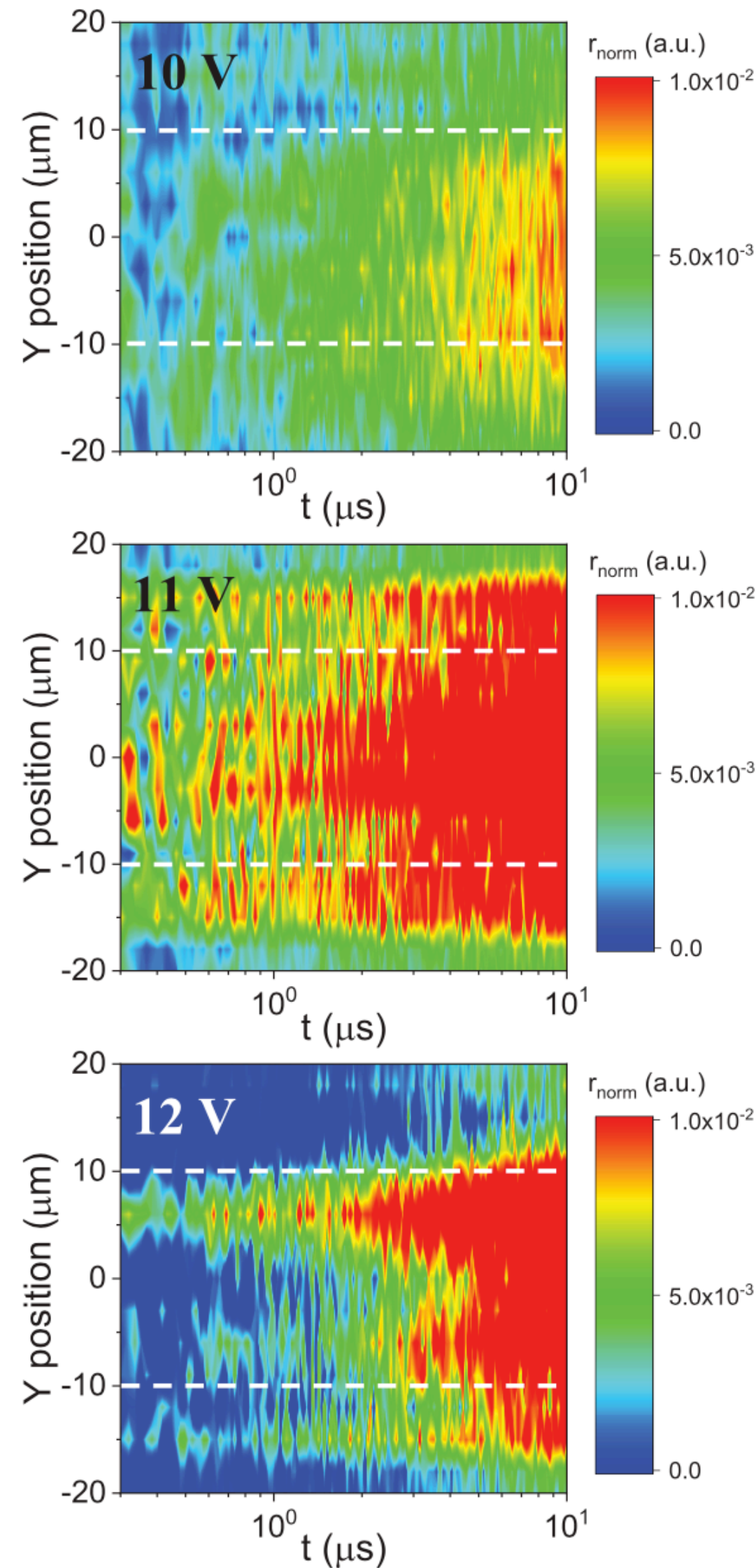
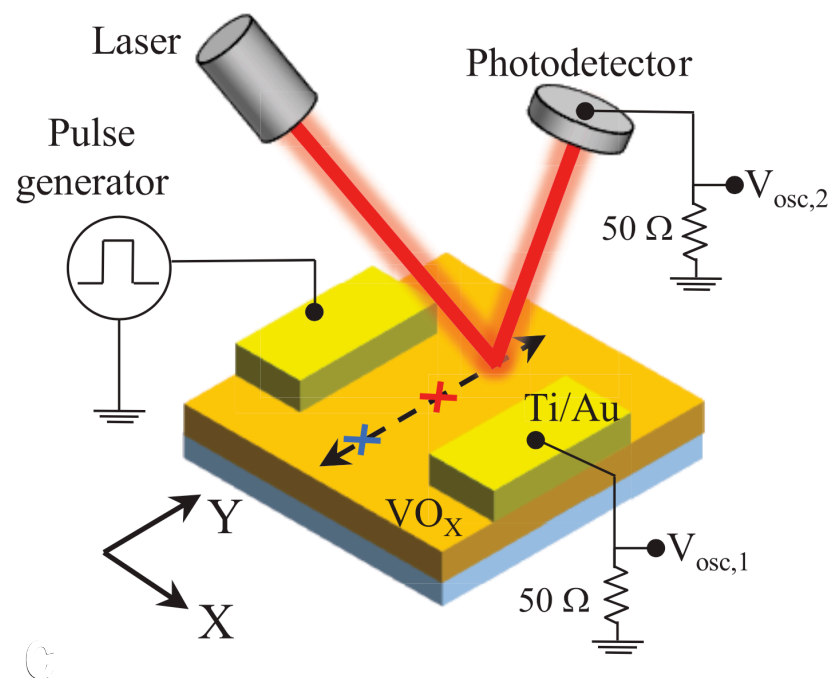
After an incubation time of $\sim 300 \text{ ns}$, a filament is formed

Normalized reflectivity in the center of the gap (red) and 10 μm away (blue).

$$r_{\text{norm}} = \frac{r - r_{\text{ins}}}{r_{\text{met}} - r_{\text{ins}}}$$

Metallization happens fast in the center on a time scale of $\sim 10^{-7} \text{ s}$ and **then expands at a much slower rate.**

Nucleation dynamics of the field-driven IMT



As the electric field is applied, the current flows inhomogeneously, Defects tend to partially suppress the IMT and lower the film resistivity which helps in focusing the current. As Joule heating concentrates in these hotspots, the temperature increases locally, further metallizing them and concentrating the current even more.

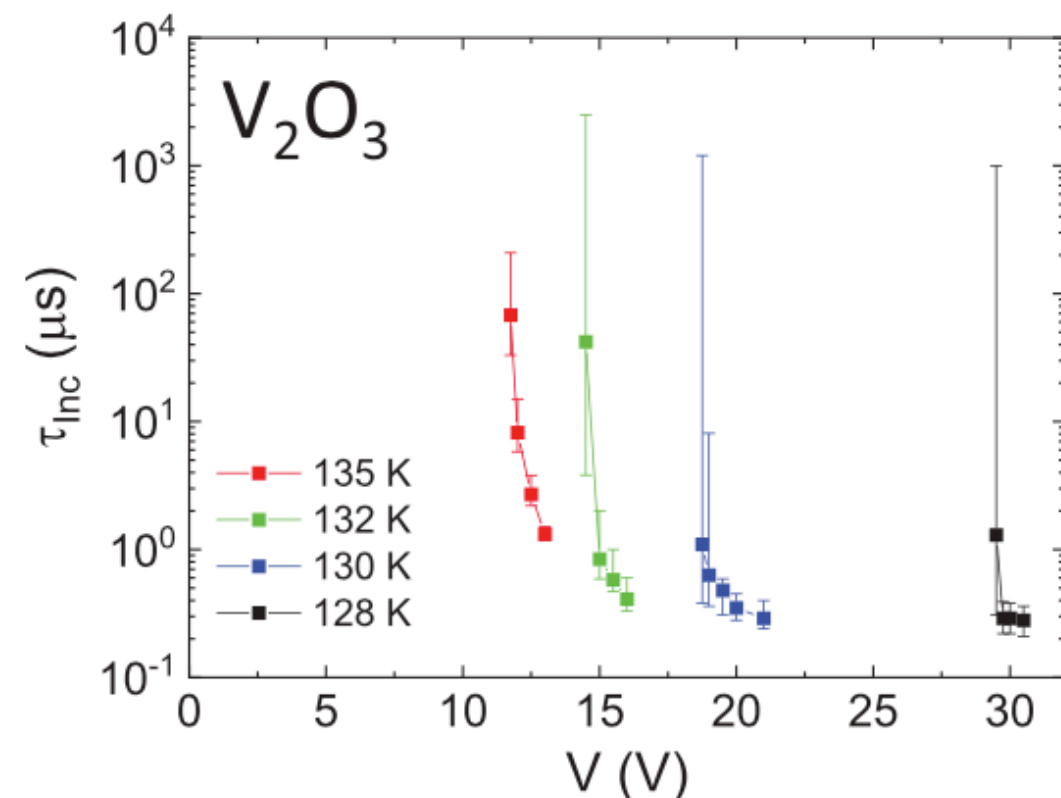
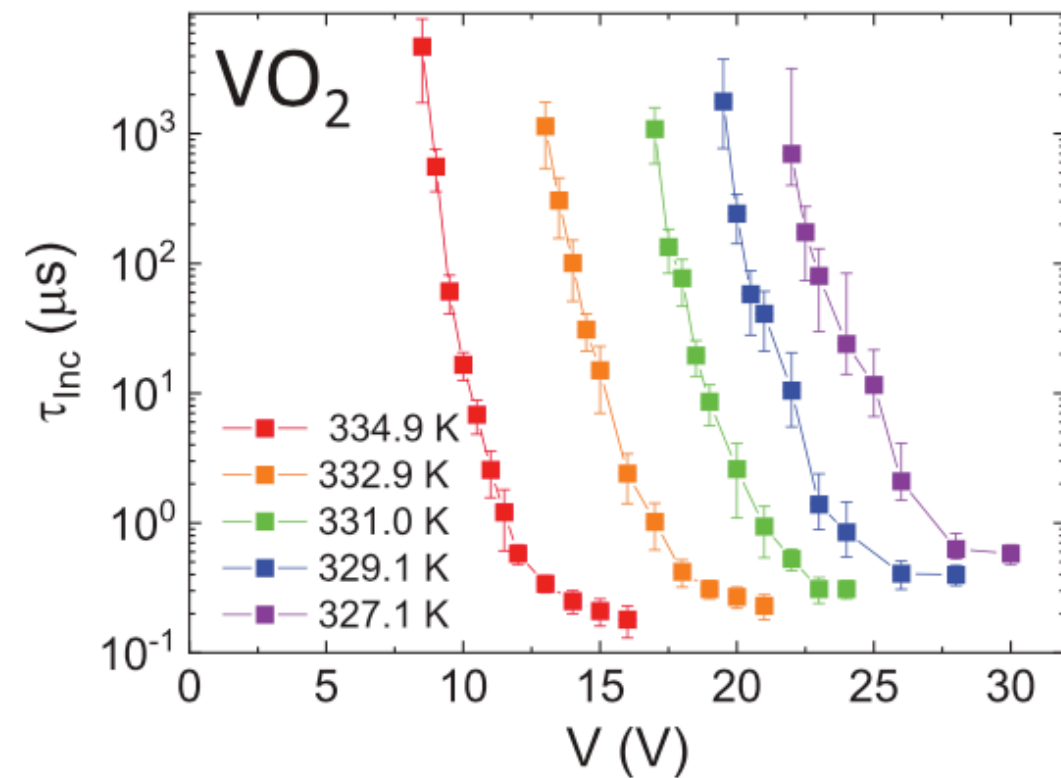
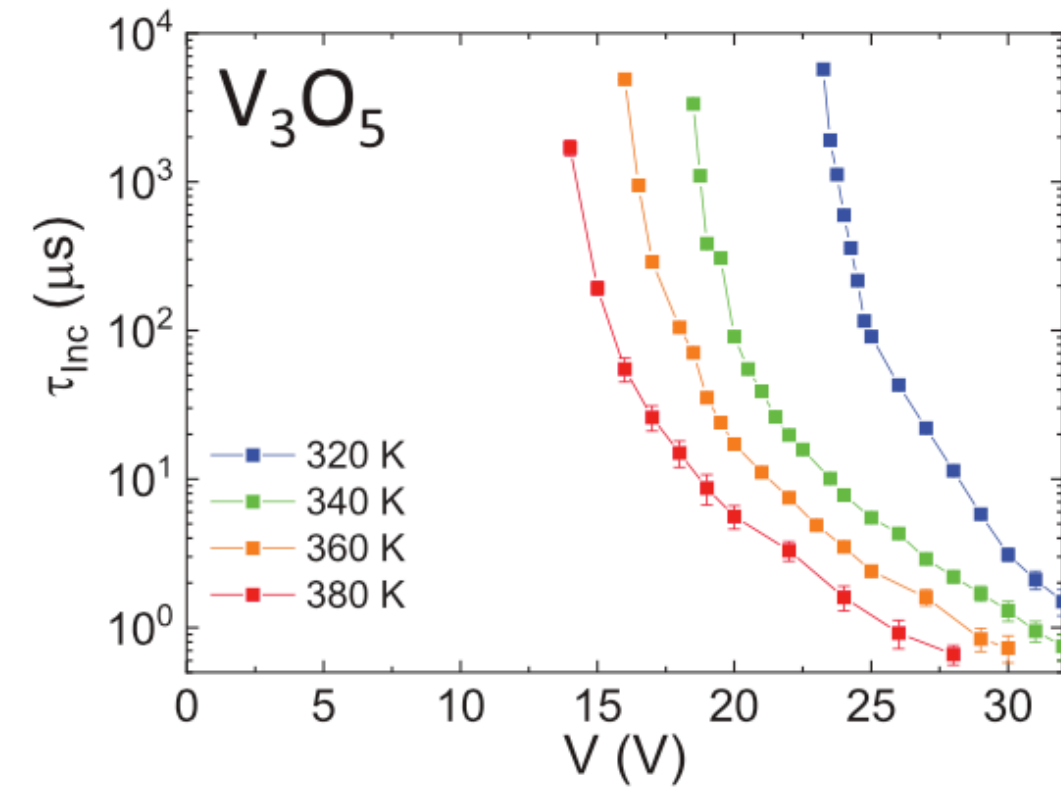
A positive feedback loop is established, leading to an instability and ultimately filament formation.

$$\frac{\partial T}{\partial t} \propto \frac{E^2}{\rho(T)} - k(T - T_0)$$

Small voltage variations can result in big differences in the nucleation process.

$$r_{norm} = \frac{r - r_{ins}}{r_{met} - r_{ins}}$$

Nucleation dynamics of the field-driven IMT



Subtle voltage variations can change t_{inc} by several orders of magnitude.

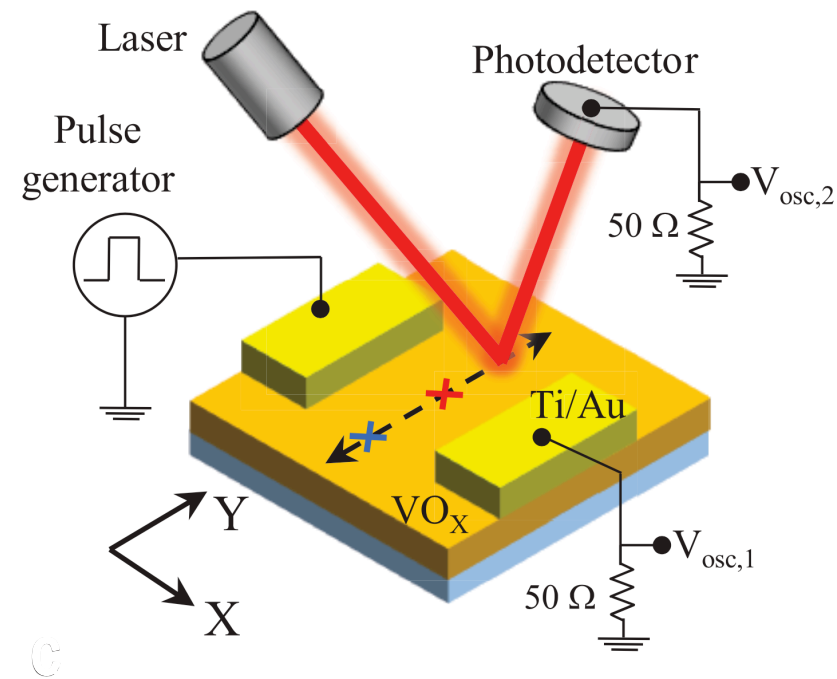
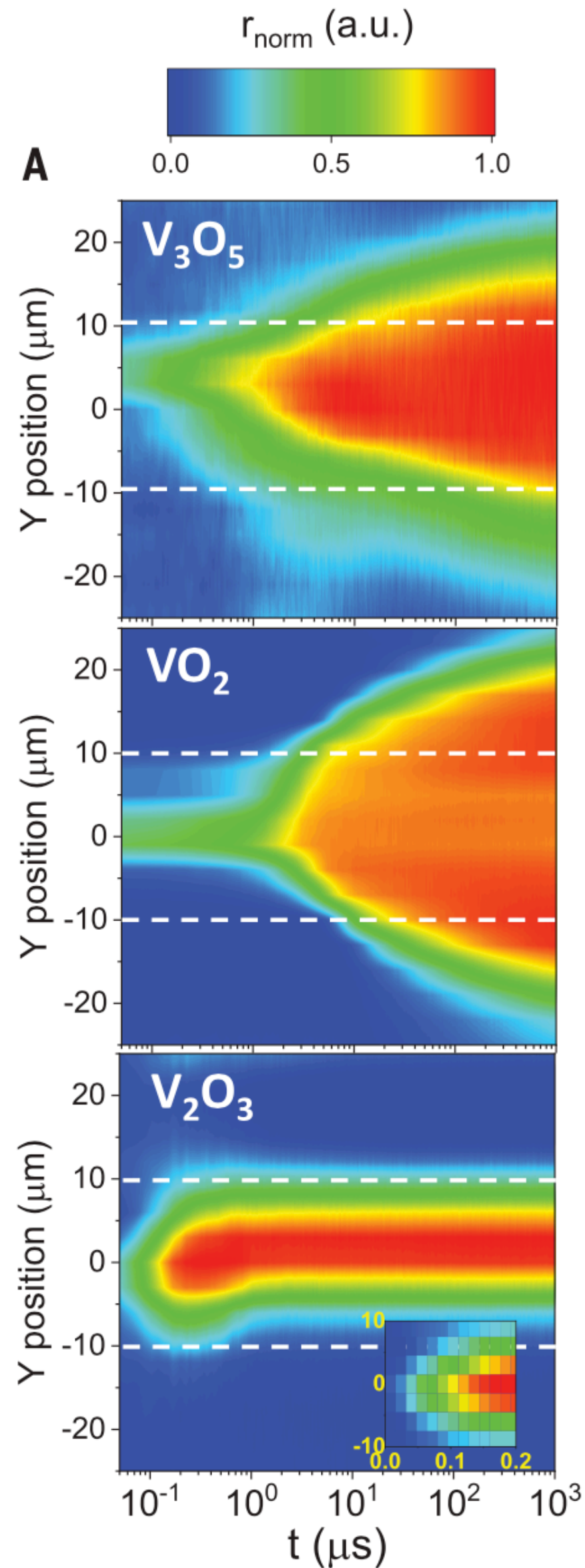
The t_{inc} sensitivity to voltage changes is relatively low for V₃O₅, higher for VO₂, and very high for V₂O₃.

In the V₂O₃ case, it approaches an all-or-nothing behavior, with t_{inc} decreasing from infinity to a few microseconds. V₂O₃ has a more pronounced thermo-electronic transition characterized by Joule heating and electric current concentration in thin filamentary domains.

For more details: **Thursday 17:20 - 17:40** → Stochastic filament formation in Mott materials under an applied voltage implements neuronal firing of exponential escape rate models - **Rodolfo Rocco**

Rocco, R., del Valle, J., Navarro, H., Salev, P., Schuller, I. K., & Rozenberg, M. Exponential escape rate of filamentary incubation in Mott spiking neurons. *Physical Review Applied*, 17(2), 024028, (2022)

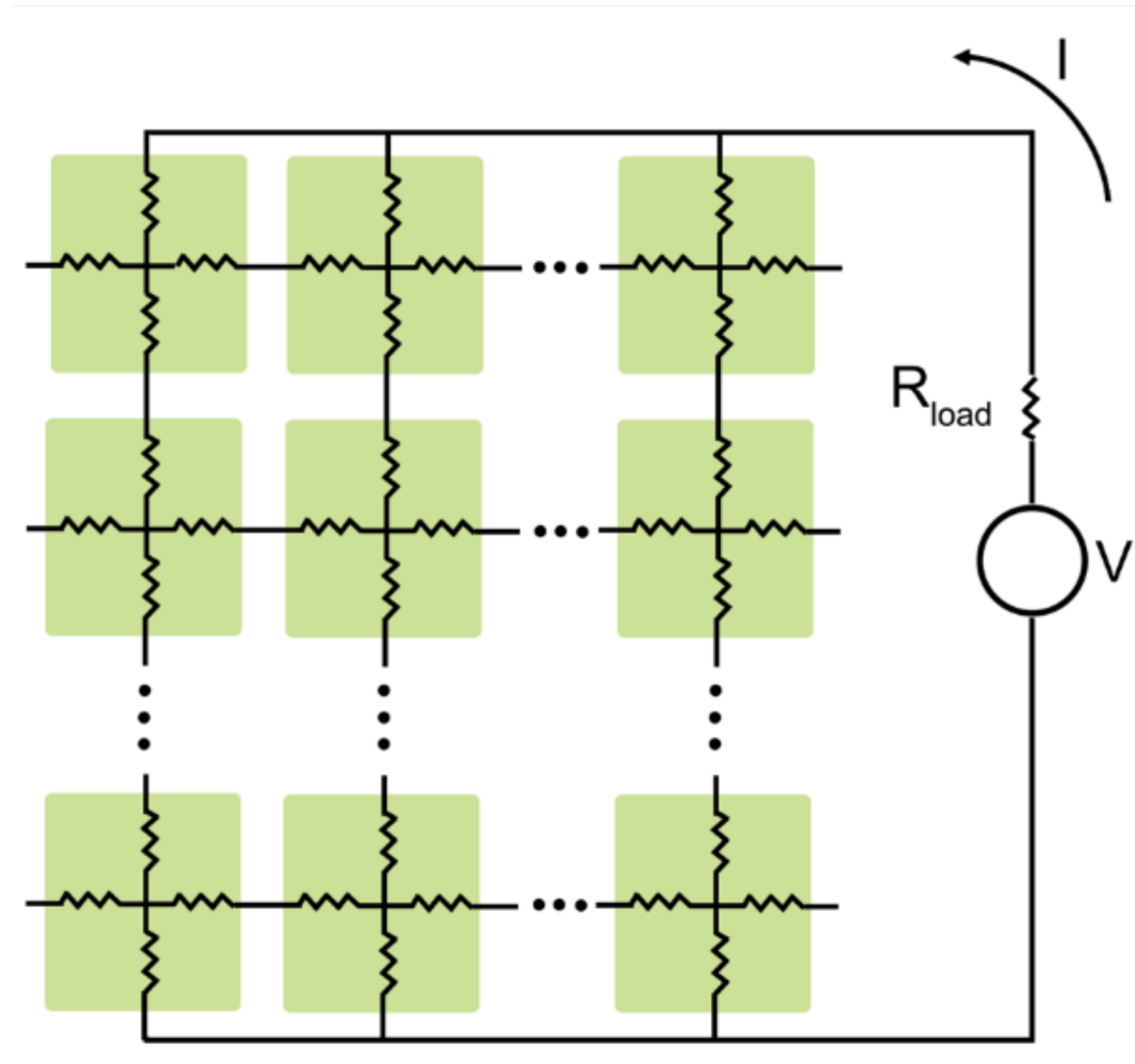
Dynamics of filament expansion



The growth dynamics of V_2O_3 look completely different when compared with VO_2 and V_3O_5

- For V_3O_5 and VO_2 , the filament is initially confined to a narrow path, but it immediately starts widening and eventually reaches its final shape. As the filament grows thicker, the current density goes down, causing a decrease in Joule heating and local temperature.
- For V_2O_3 The filament reaches a stationary configuration in 1 to 2 ms, remaining unaltered after that.

Mott resistor network model



We model the system as the Mott resistor network model in the figure.

A load resistance R_{load} is making a voltage divider circuit, with voltage of the sample given by:

$$V_S = \frac{R_S}{R_S + R_{load}}$$

Each site of our 2D lattice is represented by 4 resistors of

2 possible values:

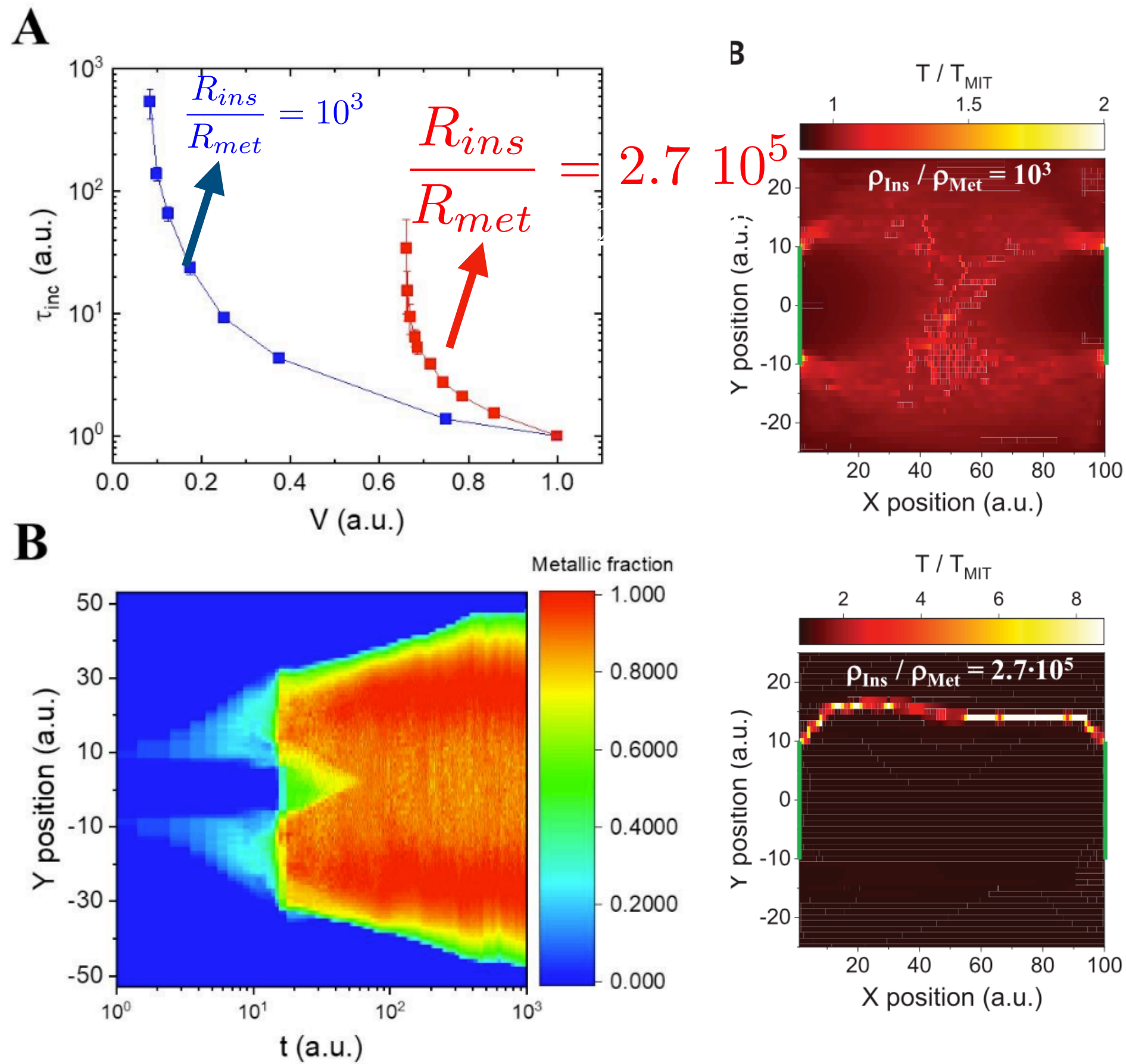
$$R_{cm}^{VO_2}, \text{ and } R_{MI}^{VO_2}$$

P. Stoliar, L. Cario, E. Janod, B. Corraze, C. Guillot-Deudon, S. Salmon-Bourmand, V. Guiot, J. Tranchant, and M. Rozenberg, *Advanced Materials* 25, 3222 (2013)

V. Guiot, L. Cario, E. Janod, B. Corraze, V. Ta Phuoc, M. Rozenberg, P. Stoliar, T. Cren, and D. Roditchev, *Nature communications* 4, 1722 (2013)

P. Stoliar, J. Tranchant, E. Janod, B. Corraze, M.-P. Besland, F. Tesler, M. Rozenberg, L. Cario
Adv. Funct. Mat. 27, 1604740 (2017)

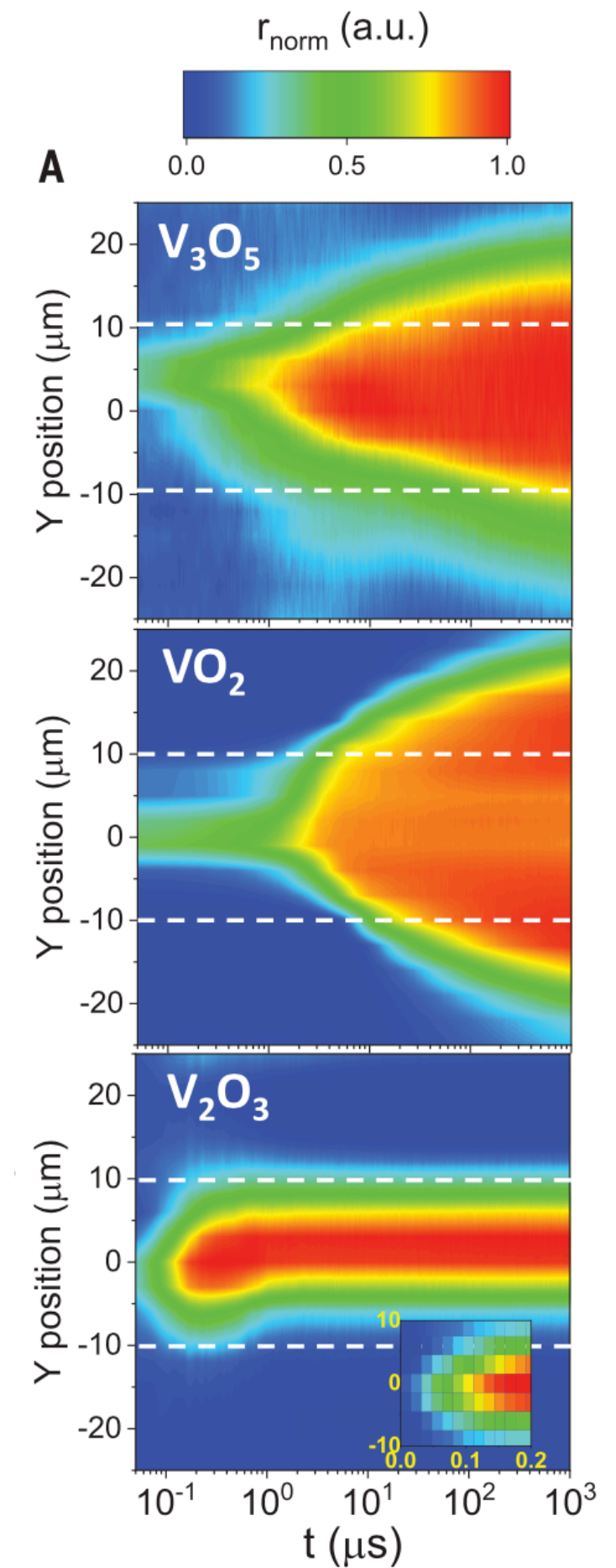
Simulations of filament nucleation and growth dynamics



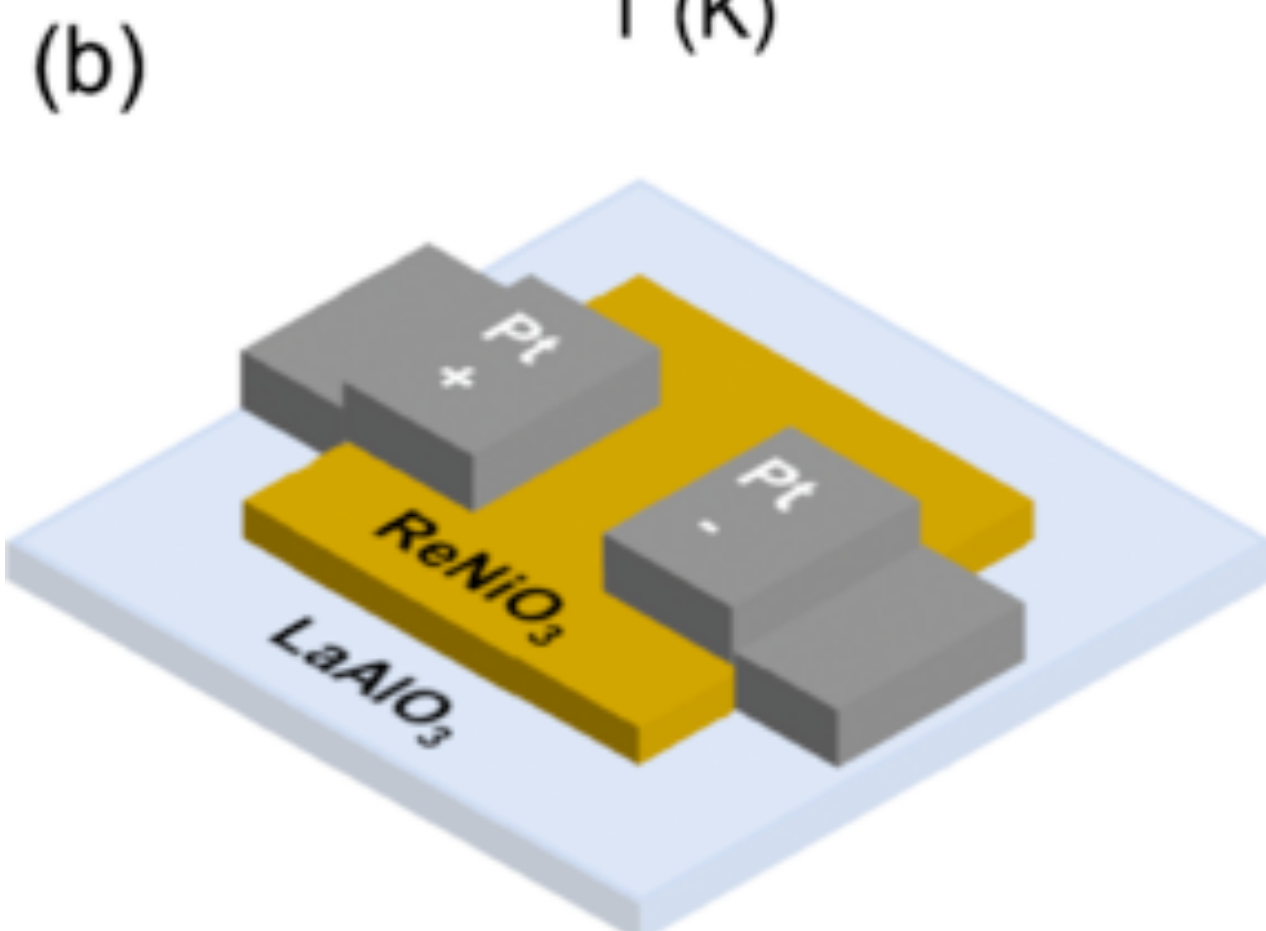
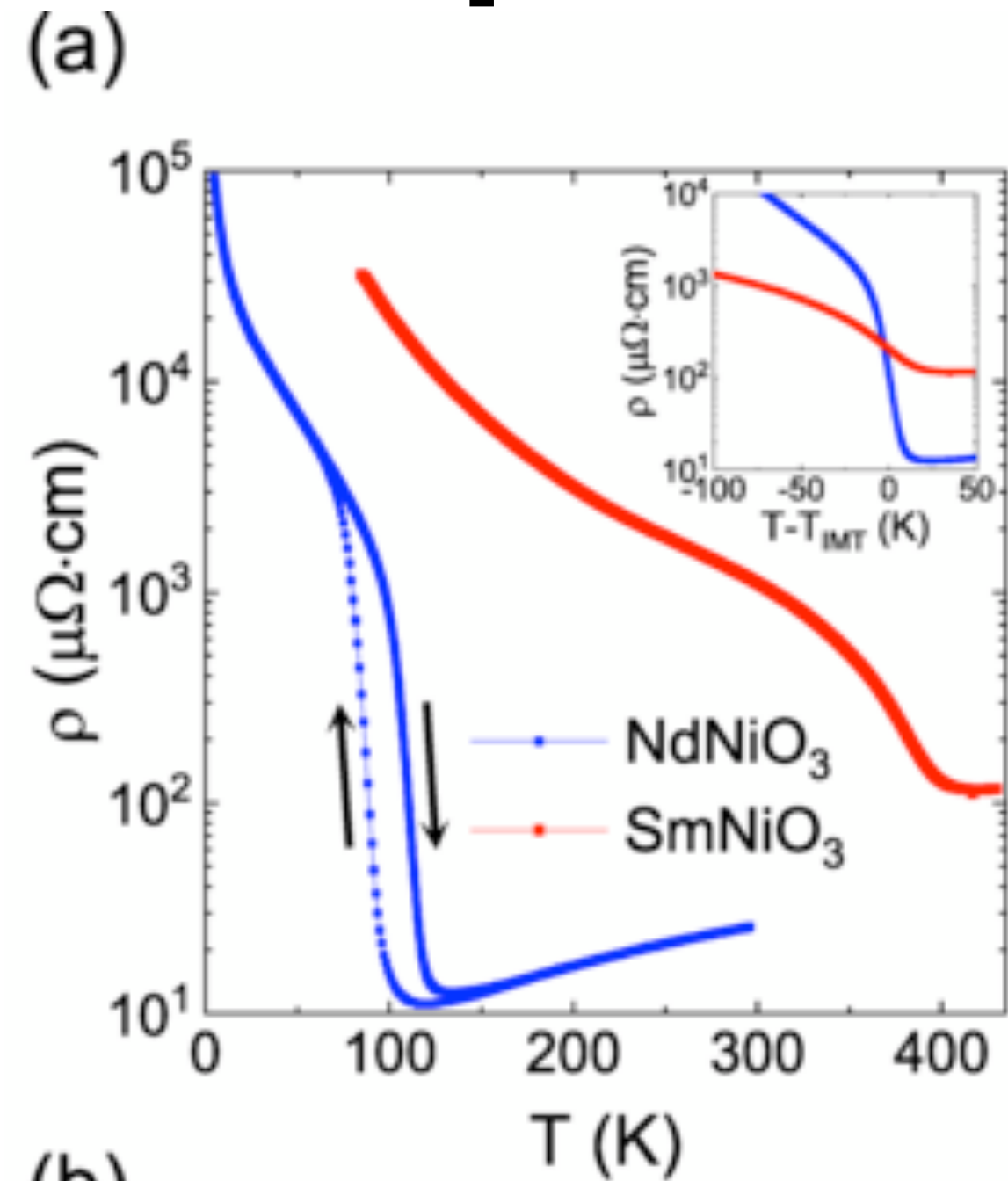
τ_{inc} is more sensitive to voltage changes for larger R_{ins}/R_{met} ratios.

Stochastic behavior in filament formation increases with an increasing r_{ins}/r_{met} ratio.

For large R_{ins}/R_{met} , a very small volume of the sample controls the initial nucleation.



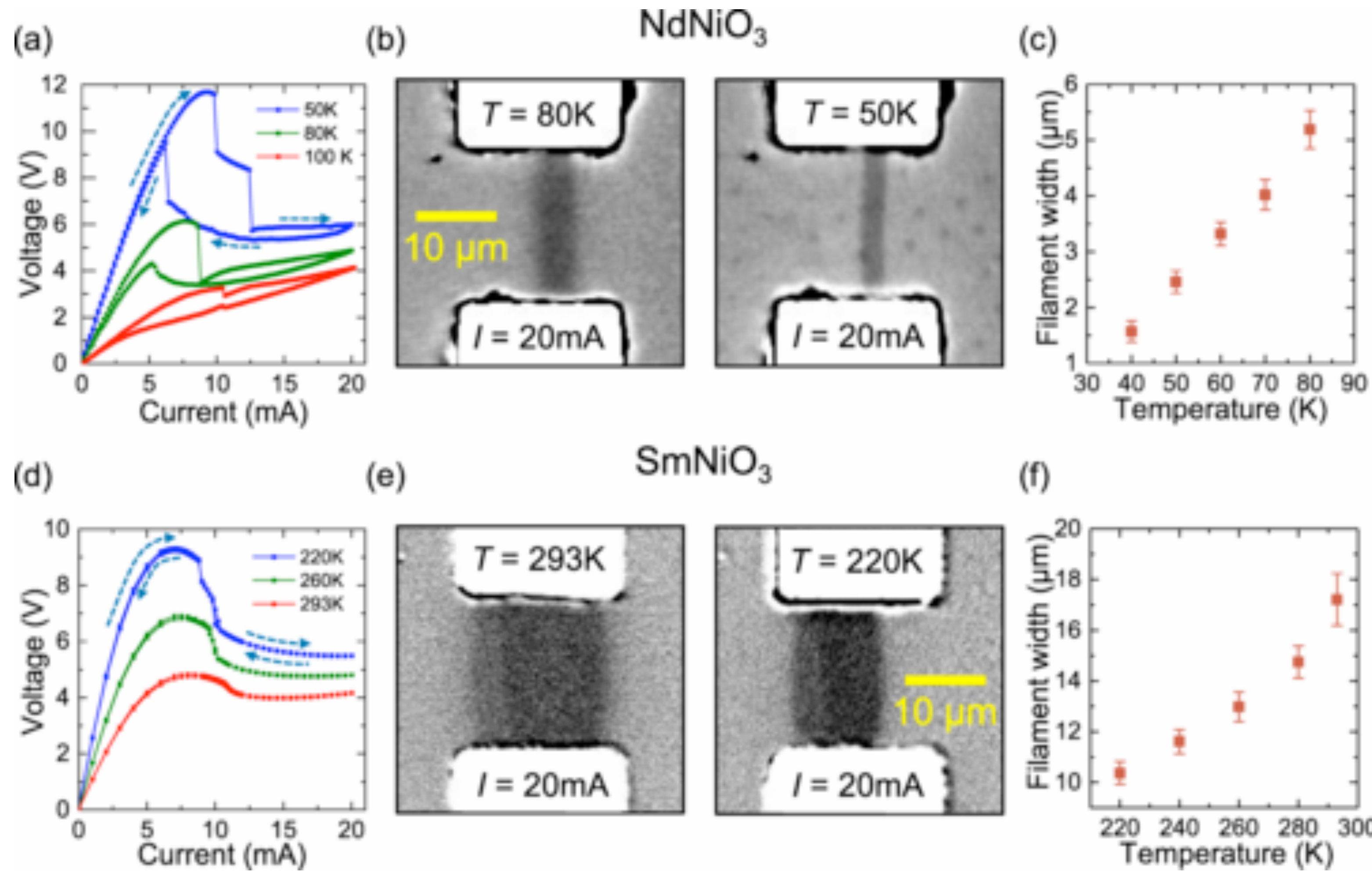
What sets the size of these filaments, and how does this impact resistive switching properties?



- **NdNiO_3 has a sharp IMT ~ 120 K with a resistivity drop of more than two orders of magnitude**
- **SmNiO_3 on the other hand, displays a smooth IMT ~ 400 K, with an order of magnitude resistivity change**

Such different IMTs allow us to contrast the results from both materials and to determine which parameters govern filament length scales.

Connection between resistive switching properties and filament size

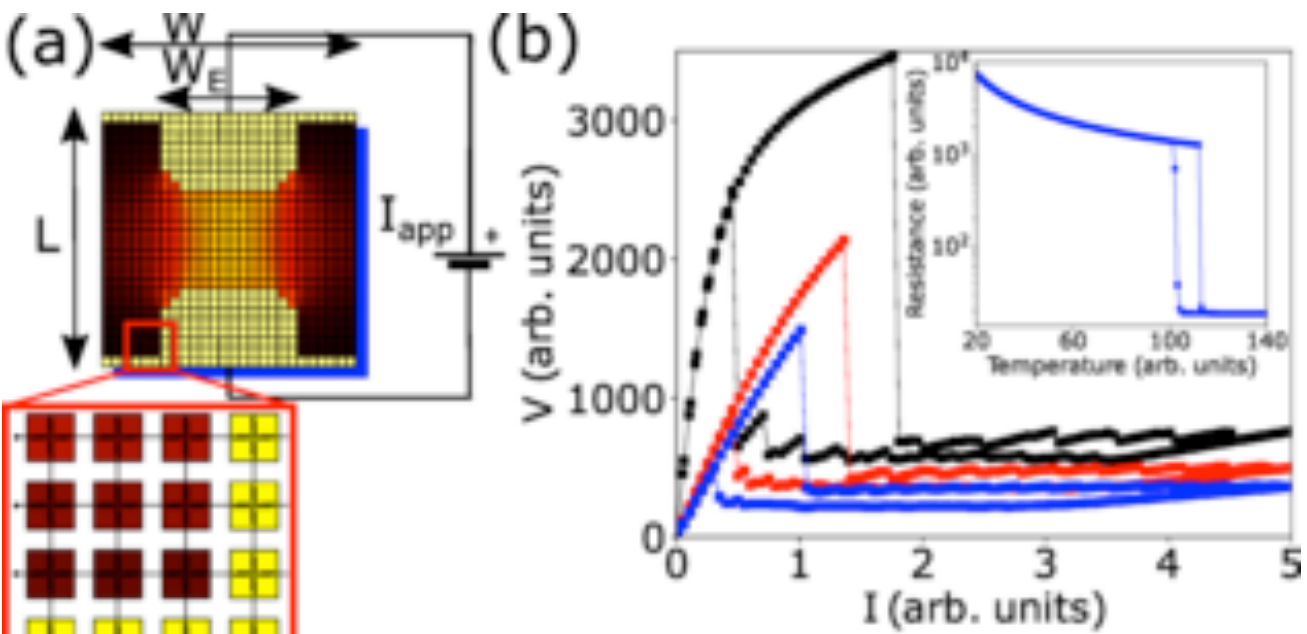


For a fixed bias current, the filament size is strongly dependent on the material and the base temperature.

We establish a strong connection between filament size and V-I characteristics:

Thinner filaments (higher current densities) lead to sharper and larger resistive switching.

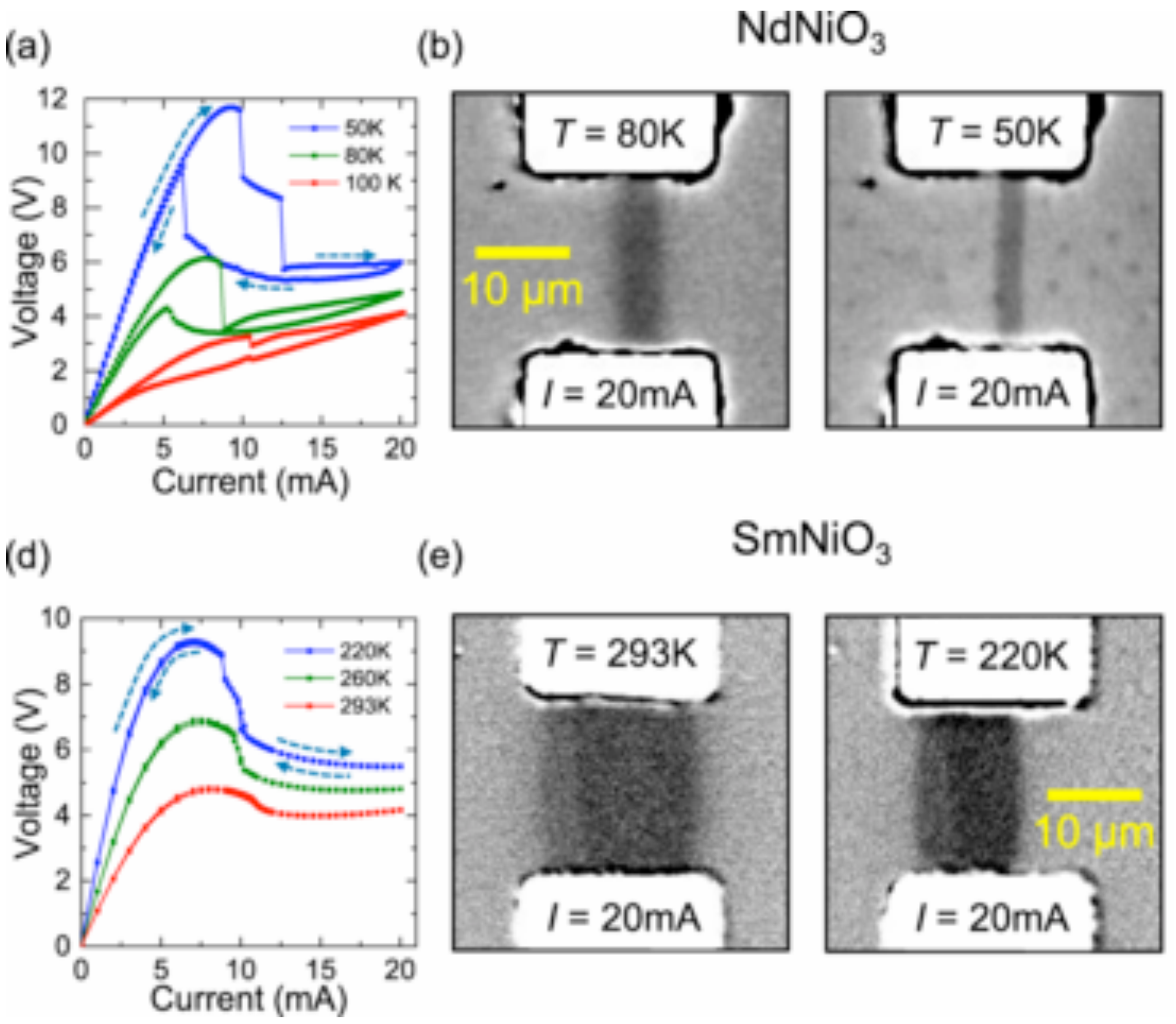
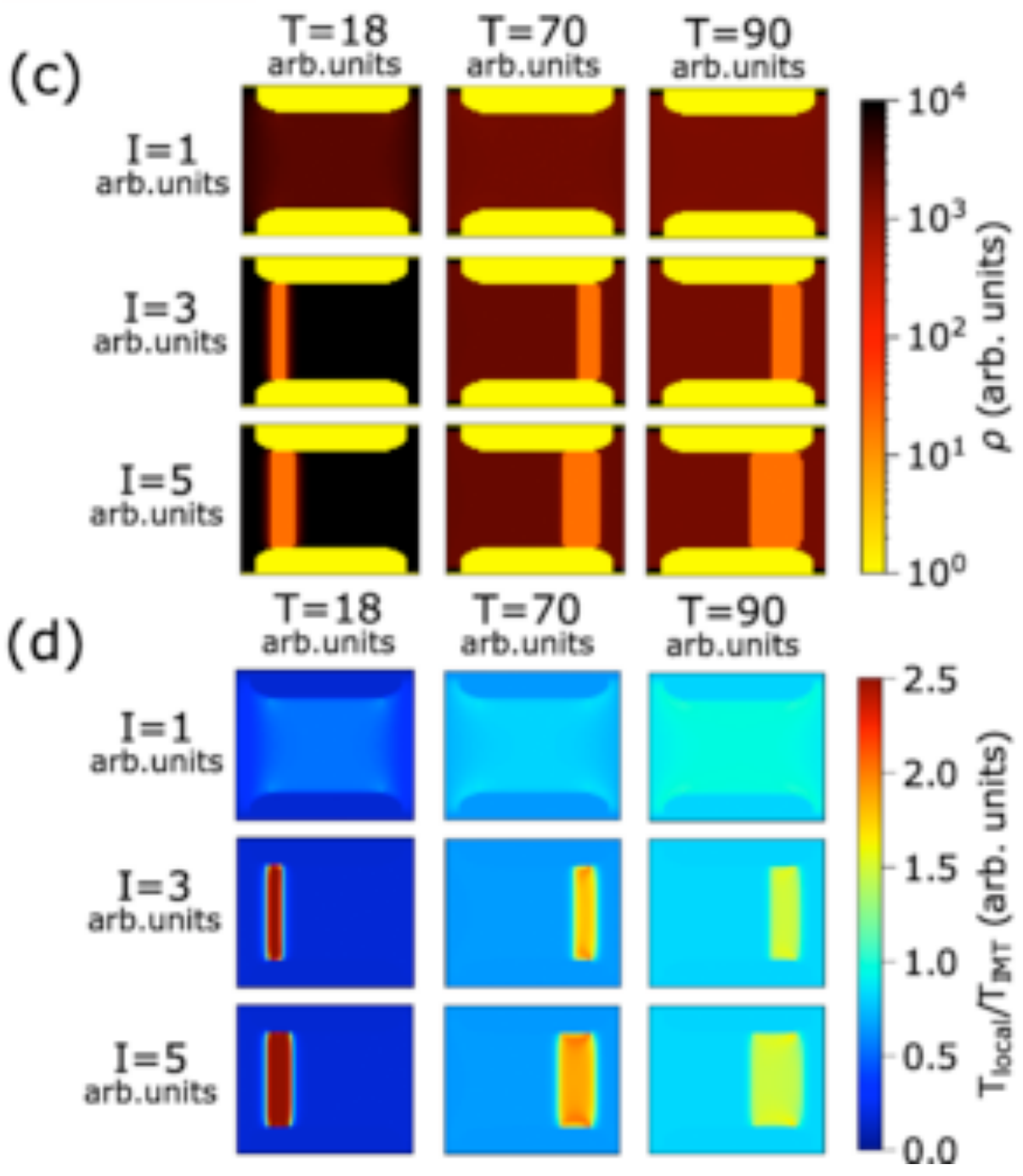
Resistor network simulations and current focusing effect



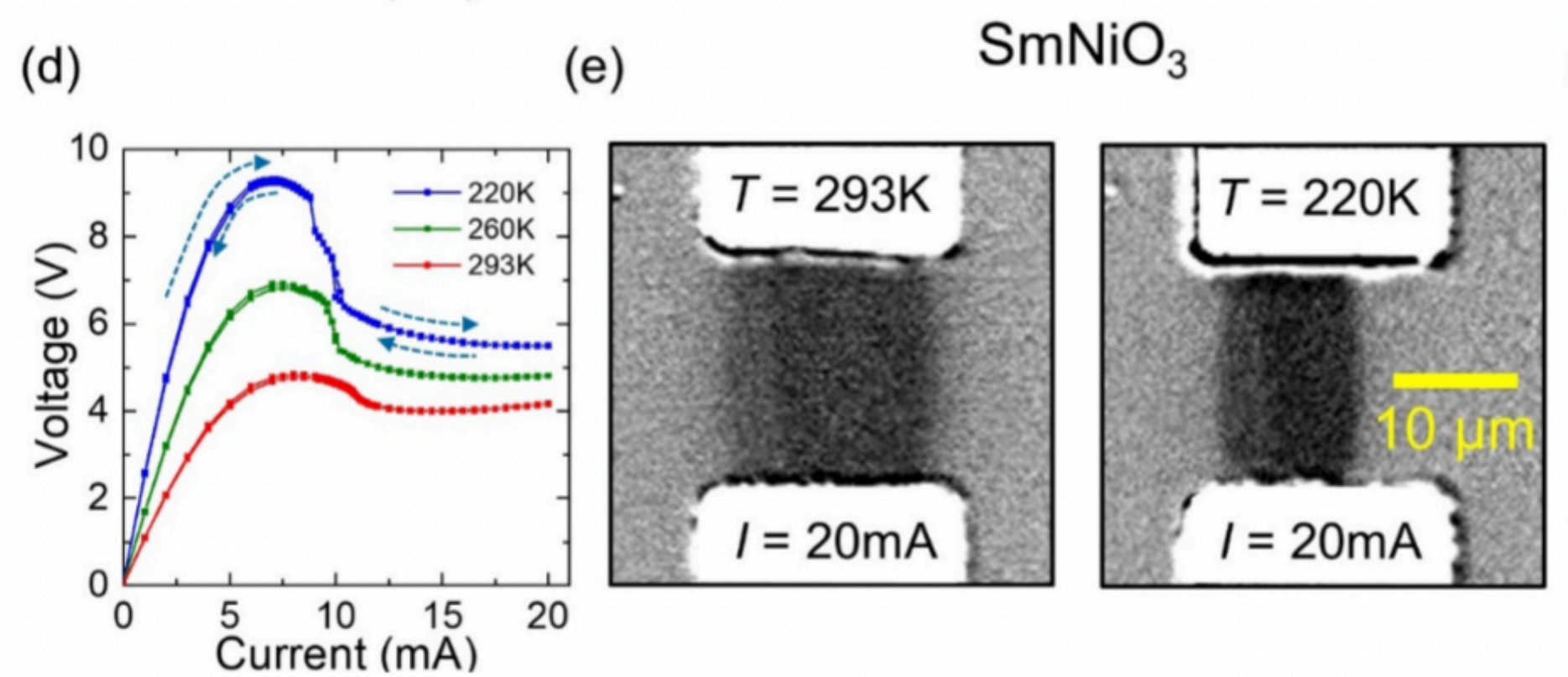
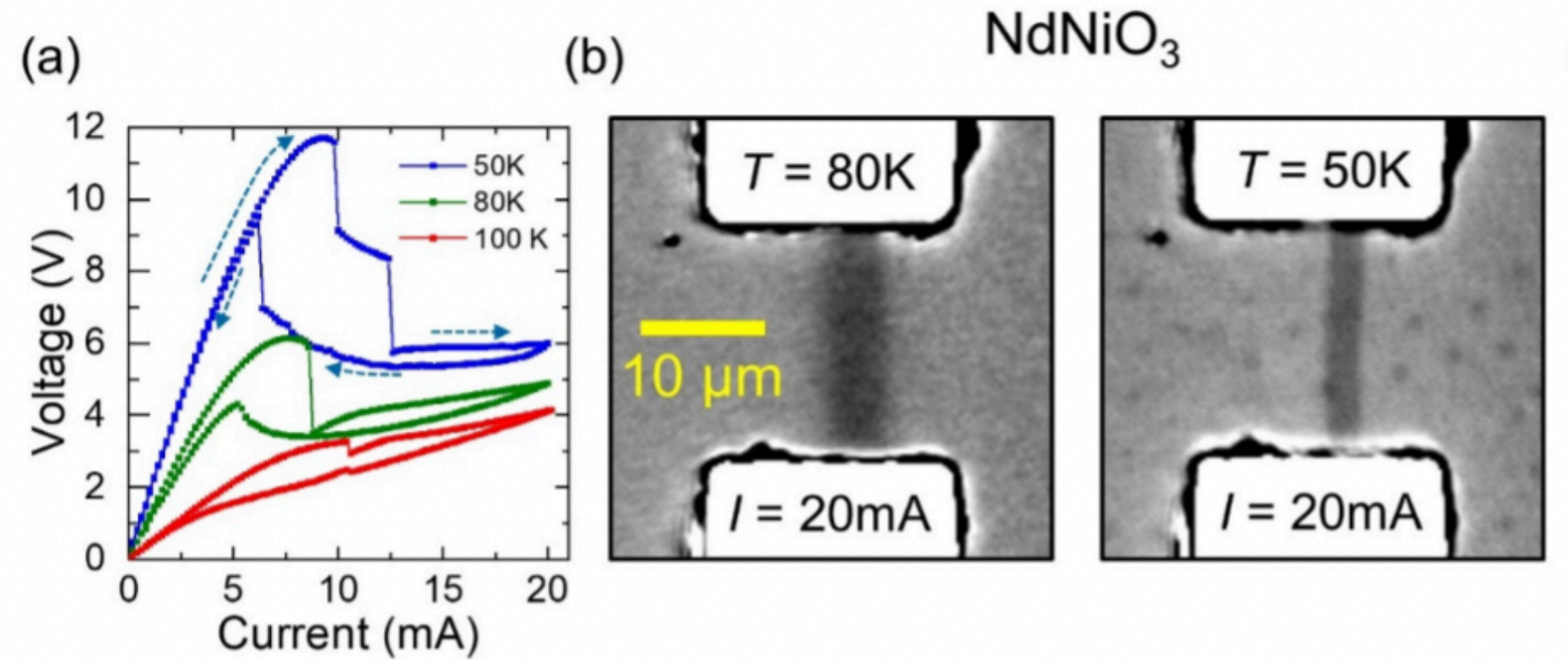
Filaments are strongly dependent on the bias current.

Filaments become narrower as the base temperature is lowered. As the filament narrows, its inner temperature increases.

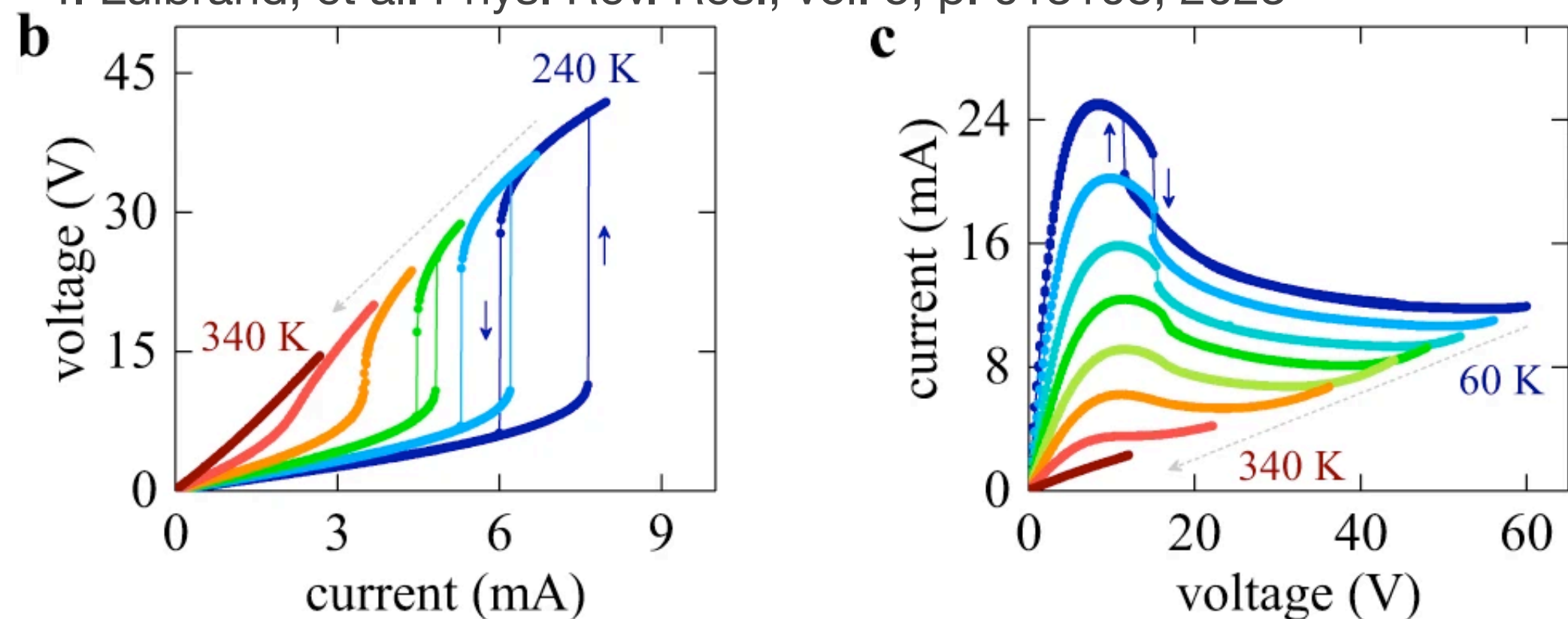
As a result, the thinner the filament and the higher its current-temperature feedback, the larger the discontinuities in the V - I curves.



Volatile resistive switching in MIT materials



T. Luibrand, et al. Phys. Rev. Res., vol. 5, p. 013108, 2023



P. Salev, et al. Nature communications, vol. 12, no. 1, pp. 1–7, 2021

- Two switching types are possible:**
- insulator-to-metal (I→M), e.g. in VO₂, V₃O₅, V₂O₃, NdNiO₃ and SmNiO₃
 - metal-to-insulator (M→I), e.g. in LSMO

These two switching types have contrasting behaviors:

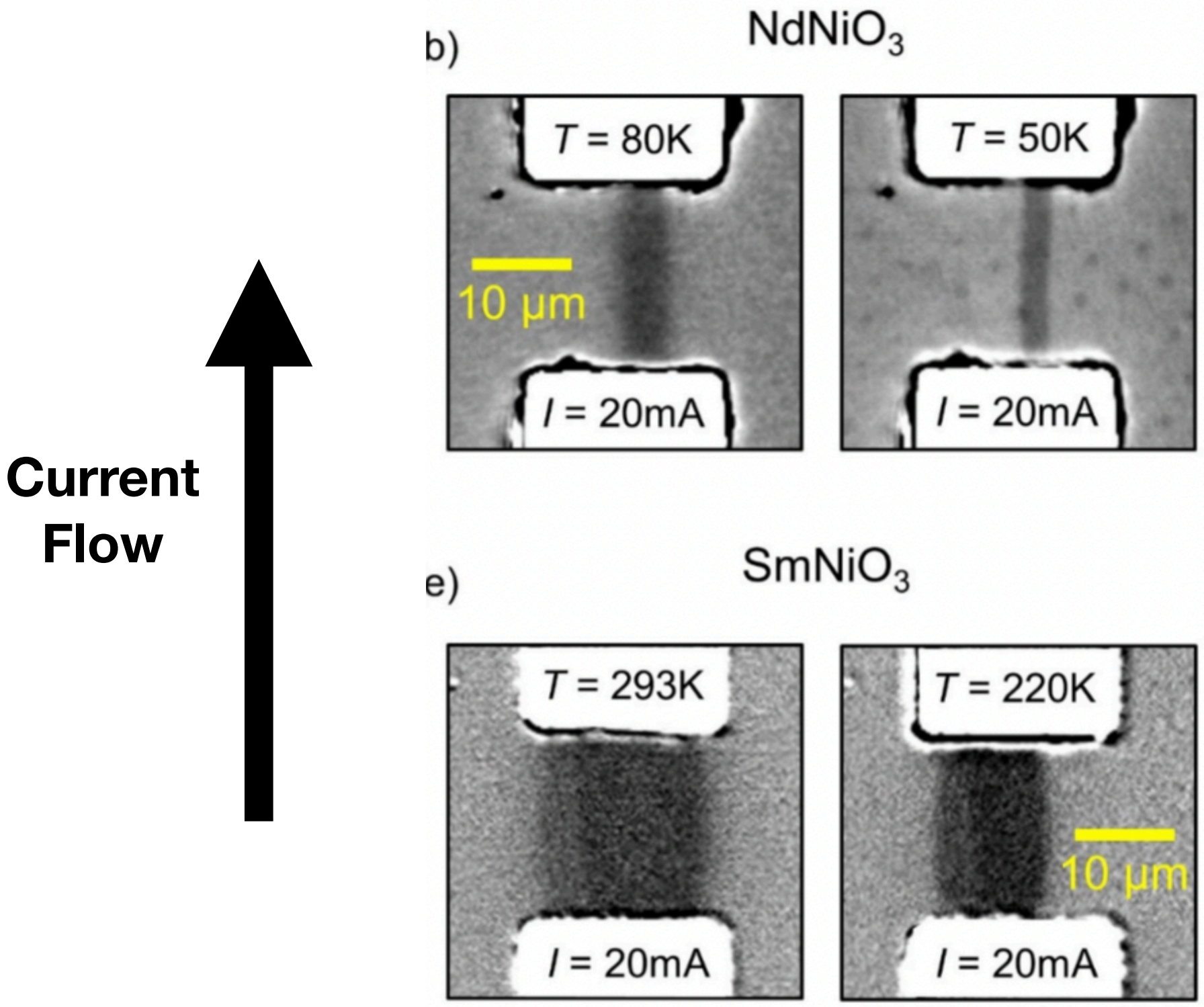
Under voltage biasing:

- I→M is abrupt and hysteretic
- M→I is gradual

Under current biasing:

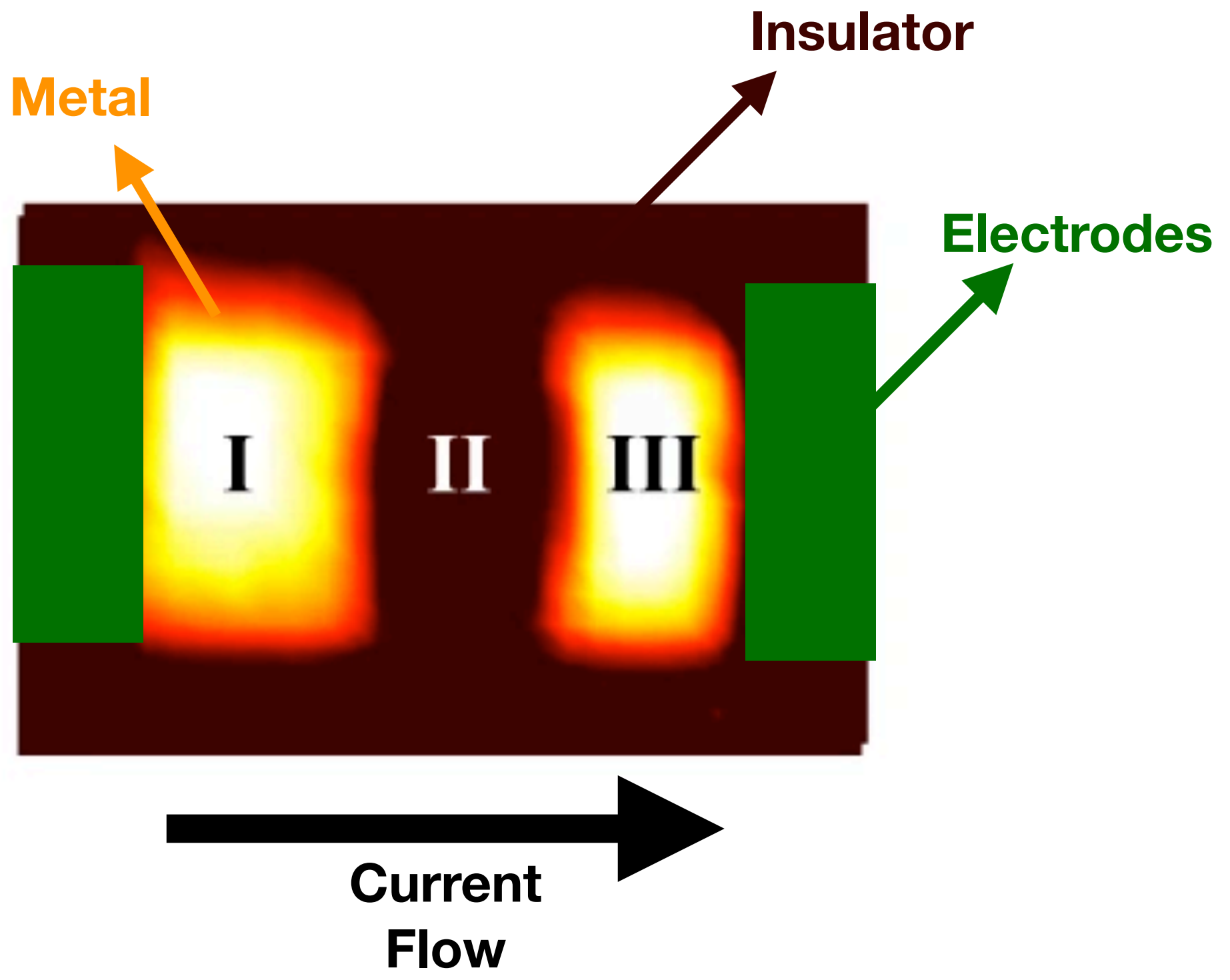
- I→M occurs gradually
- M→I is abrupt and hysteretic

Volatile resistive switching in MIT materials



The gradual switching is accompanied by a spatially inhomogeneous transition:

- In I→M, this characteristic pattern is a conducting filament parallel to the current flow
- In M→I an insulating barrier perpendicular to the current flow forms during the switching



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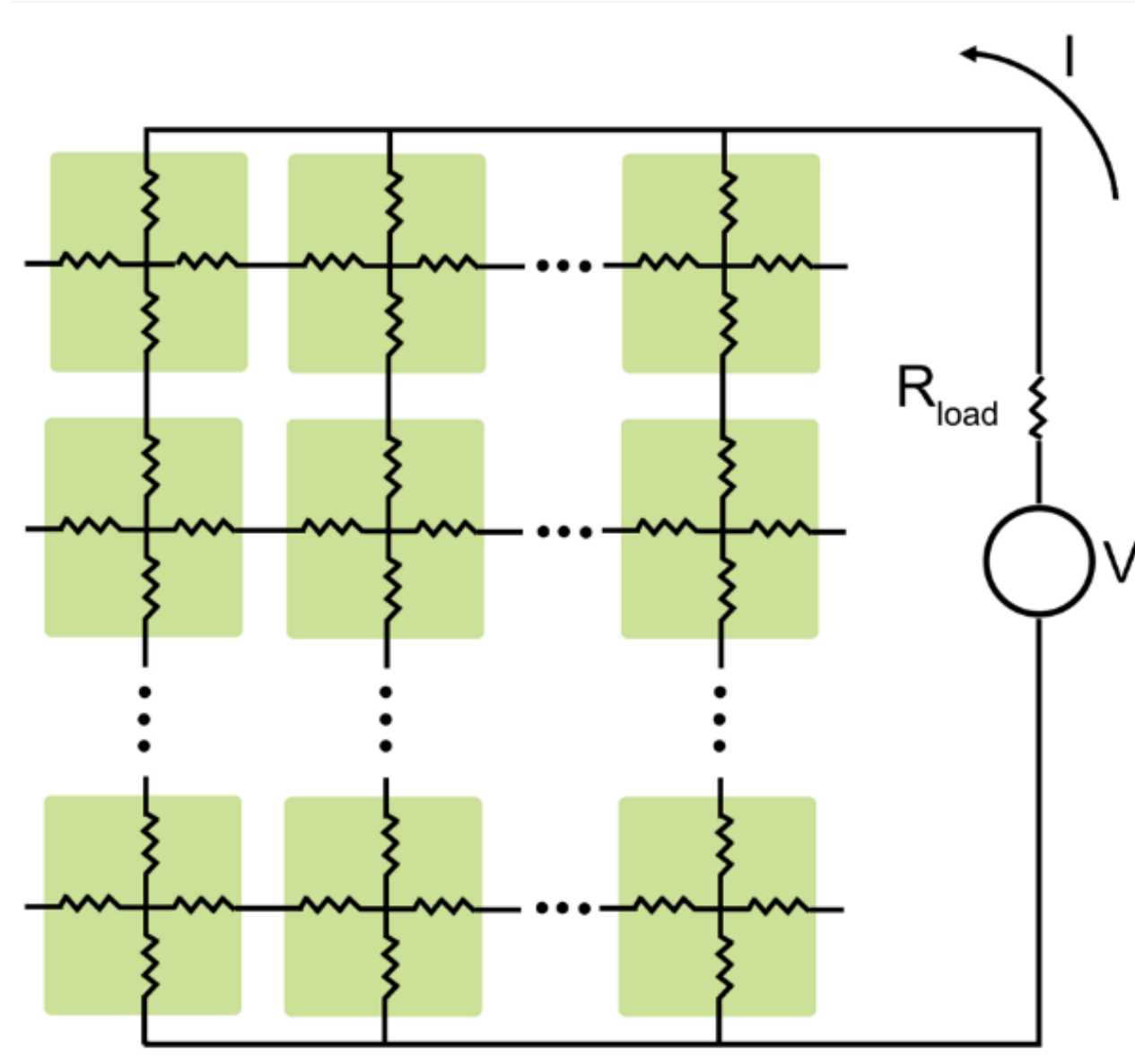
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Thank you for the attention!

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Mott resistor network model algorithm

All resistors of the network are given by substrate temperature



At each time-step is applied a certain voltage V_{app} and the full **resistor network circuit is solved using Kirchhoff law**, obtaining all the local voltage drops. The **local temperature** is updated using the heat equation through the discrete Laplacian approximation:

$$c_h \frac{\partial T_{ij}}{\partial t} = P_{ij} - k \left(5T_{ij} - \sum_{\text{neighbors}} T_i \right)$$

Then for each resistor of the lattice we assign a new value of the resistivity given by the new site temperature