Preisach-Percolation Dynamics of Multilevel Resistive Switching in Phase-Separated LPCMO

A Statistical Model of Thermal Hysteresis and Joule-Heating-Driven Memory

Prof. Juan Gabriel Ramírez

jgramirez@uniandes.edu.co

with Ali C. Basaran, Diego Carranza-Celis, Christian T. Wolowiec, P. Salev, and Ivan K. Schuller

[1] D. Carranza-Celis et al., *Phys. Rev. Mater.* **5**, 124413 (2021). Thin films **[2]** D. Carranza-Celis et al., *Phys. Rev. Mater.* **8**, 054401 (2024). Bulk

[3] G. Gomide, D. Carranza et al., *APL Mater.* **13**, 041122 (2025). Bulk **[4]** D. Carranza-Celis et al., *In preparation.* Bulk

Universidad de los Andes
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Neuromorphic computing: why analogue memory?

The von Neumann bottleneck

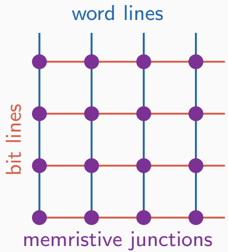
- Modern AI: data shuttles between CPU and RAM on every operation
- Energy cost $\propto$ data movement — unsustainable at scale
- GPT-scale inference: $\sim$MW per data center

Neuromorphic approach

- Memory and computation co-located (like a synapse)
- Analogue weight stored as *resistance* state
- Vector–matrix multiply in hardware: $I = G \cdot V$
- Orders-of-magnitude energy gain over digital

Key requirement: a device with

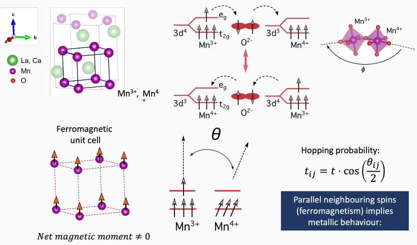
- Non-volatile multilevel resistance states
- Deterministic, reproducible switching



Crossbar array: each junction stores an analogue synaptic weight

Two parent compounds at $x = 3/8$: opposite ground states

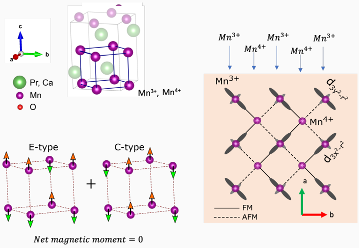
$La_{1-x}Ca_xMnO_3$



- Large La^{3+} → undistorted lattice
- e_g electrons hop freely (double exchange)

⇒ **Ferromagnetic metal**

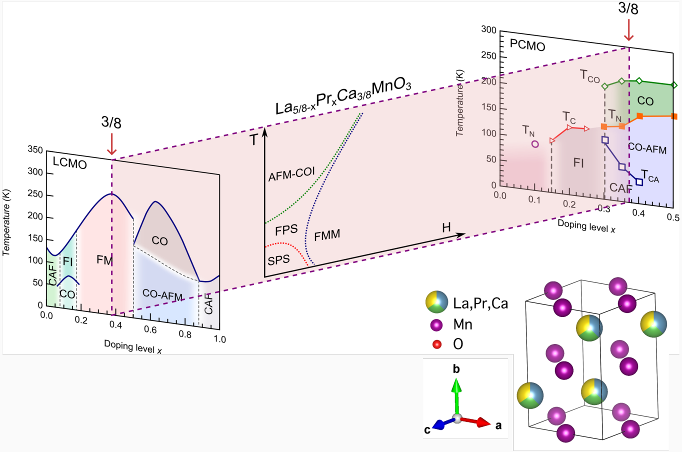
$Pr_{1-x}Ca_xMnO_3$



- Smaller Pr^{3+} → lattice distortion (Jahn-Teller)
- Mn^{3+}/Mn^{4+} lock in checkerboard: charge ordering

⇒ **Charge-ordered insulator**

LPCMO: tuning between the two extremes



$La_{5/8-x}Pr_xCa_{3/8}MnO_3$

Tuning x interpolates:

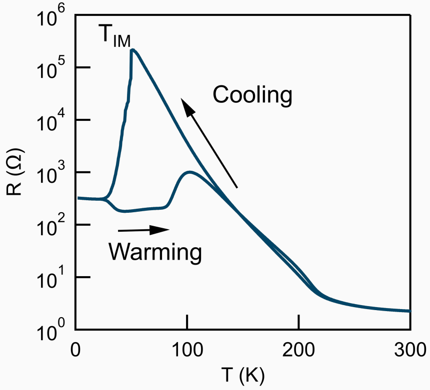
- $x \to 0$: LCMO (FM metal)
- $x \to 5/8$: PCMO (insulator)
- $x \approx 0.4$: both compete

Nanoscale phase separation:

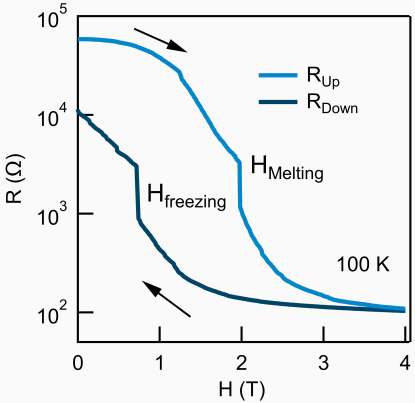
- FM clusters in COI matrix
- Neither phase wins globally
- R set by percolating FM network

Fingerprint of phase competition: $R(T)$ and $R(H)$

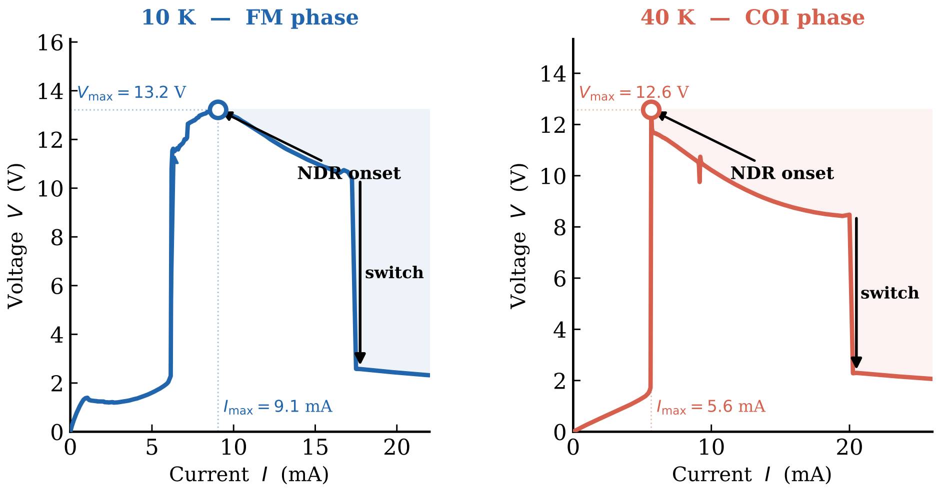
Resistance vs temperature
($x = 0.40$, zero field)



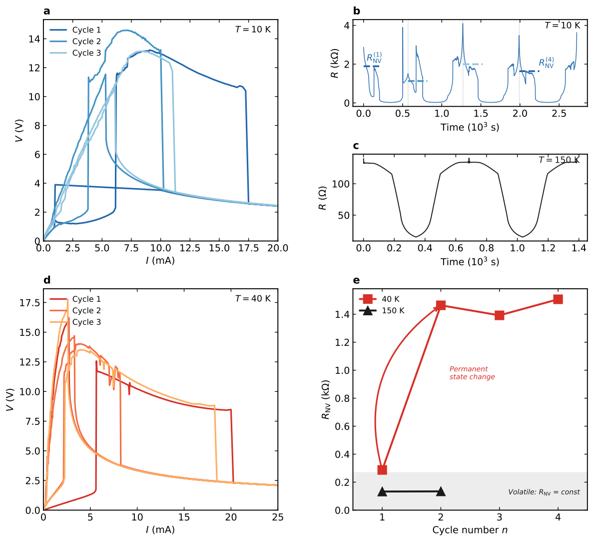
Resistance vs magnetic field
($T = 100$ K, inside the hysteresis region)



The device: current-controlled resistive switching



Non-volatile vs volatile: two distinct regimes



At 10 K (FM phase)

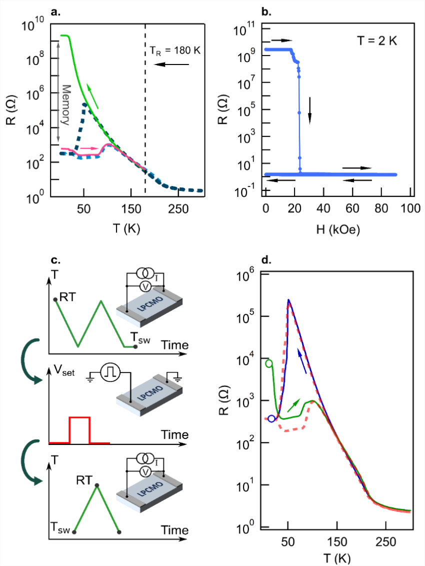
- R_{NV} drifts upward cycle-to-cycle
- Each pulse leaves the device in a higher-resistance state
- Memory is **non-volatile**: R does not relax
- Mechanism: pulse drives FM $\to$ COI fraction permanently

At 150 K (COI phase)

- $R(t)$ is perfectly periodic — volatile
- Device returns to same baseline after each pulse
- Local heating is insufficient to cross T_R

Question: What determines whether a pulse leaves a permanent trace or not?
$\to$ **The thermal pathway**

The thermal pathway: minor loops in R–T



Full hysteresis loop:

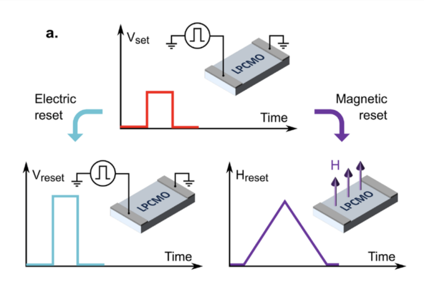
- Cool from 300 K → 2 K: FM metallic state, $R \approx 300\,\Omega$
- Warm 2 K → 300 K: crosses $T_{\mathrm{MIT}}^{\mathrm{warm}} = 102$ K

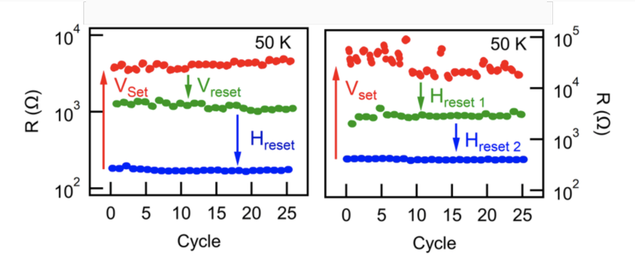
Minor loops (Joule pulse simulation):

- Warm to T_r then re-cool to 2 K
- For $T_r < T_R = 180$ K: re-cooling follows a minor cooling curve — device ends in higher R state
- For $T_r > T_{\mathrm{COI}} = 210$ K: full FM recovery on re-cooling

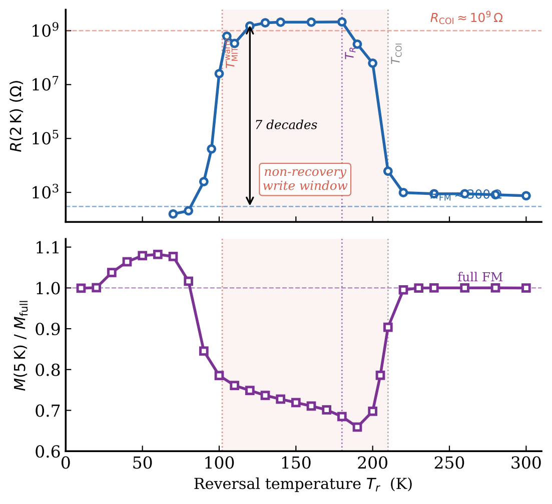
Bifurcation at $T_R \approx 180$ K
Above: FM metal restored
Below: COI fraction frozen in

Non-volatile resistive memory





What a current pulse writes: $R(2\,\text{K})$ and $M(5\,\text{K})$ vs T_r



Experiment: cool to 2 K → warm to T_r → re-cool to 2 K. Measure R and M after re-cooling.

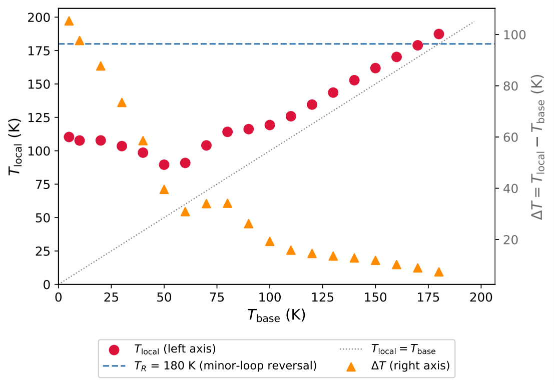
Three regimes:

- $T_r \lesssim 90$ K: no change — FM phase intact, $R \approx 300\,\Omega$
- $90 \lesssim T_r \lesssim 210$ K:
  non-recovery write window
  R jumps **7 decades** to $\sim 10^9\,\Omega$
  M drops to 57% of full value
  ⇒ FM fraction permanently suppressed
- $T_r > T_{\text{COI}} \approx 210$ K:
  full FM recovery, R and M return

The open question

Why does re-cooling from $T_r < 210$ K **fail to restore** the FM phase?
→ **The Preisach + non-recovery model**

Key result: T_{local} converges to $T_R \approx 180\,\mathrm{K}$



Method: at the NDR onset, $R_{\mathrm{switch}} = V_{\max}/I_{\max}$. Map onto the R–T cooling branch $\Rightarrow T_{\mathrm{local}} = T(R_{\mathrm{switch}})$.

Two regimes:

FM phase ($T_{\mathrm{base}} \lesssim 50\,\mathrm{K}$)

- $T_{\mathrm{local}} \approx 100$–$110\,\mathrm{K}$ — nearly constant
- Self-regulation: heating through T_{MIT} chokes current
- ΔT up to 105 K

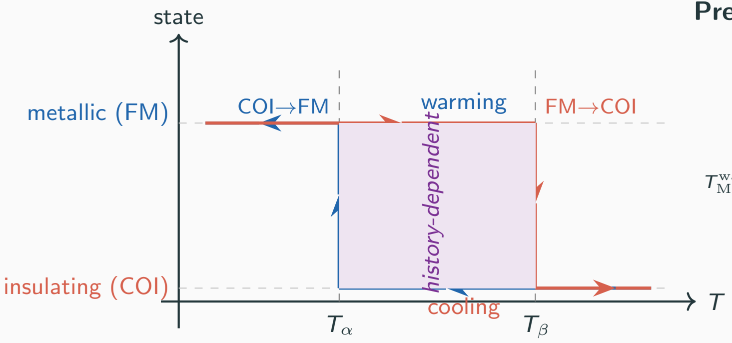
COI phase ($T_{\mathrm{base}} \gtrsim 60\,\mathrm{K}$)

- T_{local} increases linearly with T_{base}
- Converges to $T_R = 180\,\mathrm{K}$
- ΔT shrinks from $\sim 34\,\mathrm{K}$ to $\sim 7\,\mathrm{K}$
- At $T_{\mathrm{base}} = 170\,\mathrm{K}$: $T_{\mathrm{local}} \approx 179\,\mathrm{K} \approx T_R$

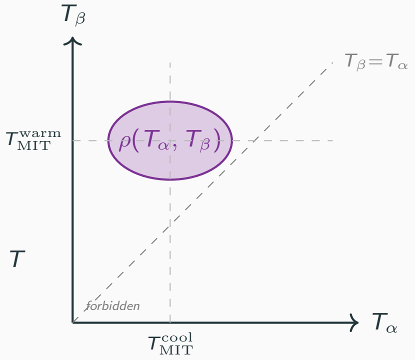
Central result

Throughout the insulating phase, the Joule pulse heats the sample to exactly the minor-loop bifurcation temperature $T_R \approx 180\,\mathrm{K}$

The Preisach model: the hysteron



Preisach plane



Each domain = one point (T_α, T_β) in the plane

ρ = density of domains (encodes quenched disorder)

The Preisach model: ensemble and effective medium

One hysteron = one microscopic domain

A two-state unit with quenched disorder:

- **Cooling:** insulator → metal at T_α
- **Warming:** metal → insulator at T_β
- $T_\beta > T_\alpha$ always — the hysteresis gap
- Between T_α and T_β: state depends on **history**

Full material = ensemble of $N \gg 1$ hysterons, $(T_\alpha^{(i)}, T_\beta^{(i)})$ drawn from $\rho(T_\alpha, T_\beta)$

FM volume fraction:

$$\eta_{\mathrm{FM}} = \iint_{\mathrm{FM\,region}} \rho(T_\alpha, T_\beta)\, dT_\alpha\, dT_\beta$$

Effective resistance:

$$\log R = \eta_{\mathrm{FM}} \log R_{\mathrm{FM}} + (1-\eta_{\mathrm{FM}}) \log R_{\mathrm{COI}}$$

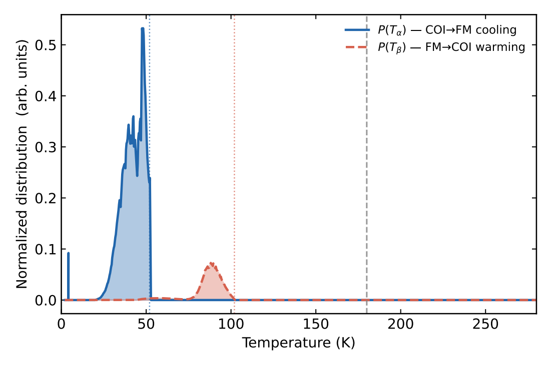
Statistical physics analogy

Spins	↔ FM/COI domains
Ext. field	↔ temperature T
Disorder	↔ spread of $(T_\alpha^{(i)}, T_\beta^{(i)})$
Order param.	↔ η_{FM}

Key difference from Ising:
each "spin" has *two* switching thresholds $(T_\alpha \neq T_\beta)$ ⇒ rectangular loop per domain

Macroscopic R–T hysteresis emerges from the distribution of individual rectangular loops

The statistical model: distributions $P(T_\alpha)$ and $P(T_\beta)$



How the distributions are extracted

- $P(T_\alpha) \propto \left|\frac{d\log R}{dT}\right|_{\mathrm{cool}}$ (cooling branch)
- $P(T_\beta) \propto \left|\frac{d\log R}{dT}\right|_{\mathrm{warm}}$ (warming branch)
- Both normalized so $\int P\,dT = 1$

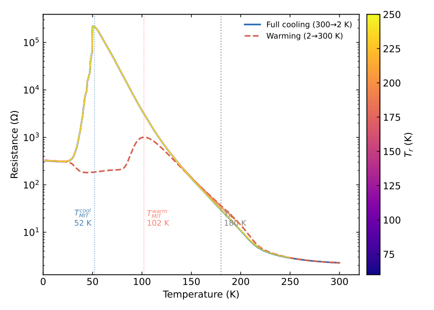
Physical meaning

- Peak of $P(T_\alpha)$ at $T_{\mathrm{MIT}}^{\mathrm{cool}} \approx 52\,\mathrm{K}$: most domains convert COI→FM around here
- Peak of $P(T_\beta)$ at $T_{\mathrm{MIT}}^{\mathrm{warm}} \approx 102\,\mathrm{K}$: most domains revert FM→COI around here
- Width of distributions = degree of phase separation / quenched disorder

These distributions are the core of the statistical model: they encode the full thermal hysteresis in a single Preisach density $\rho(T_\alpha, T_\beta) \approx P(T_\alpha) \cdot P(T_\beta)$

Minor-loop formula: the Mayergoyz vertical-shift rule

$$R_{\text{minor}}(T) = R_{\text{cool}}(T) + \underbrace{\left[R_{\text{warm}}(T_r) - R_{\text{cool}}(T_r)\right]}_{\delta(T_r)\text{: inter-branch gap at } T_r}$$



Vertical translation only — minor cooling curve is the full cooling curve shifted up by $\delta(T_r)$

At $T_r = T_R = 180\,\text{K}$: $\delta \approx 6.7\,\Omega$

No free parameters — fixed by the full R–T loop alone

Non-recovery: why minor loops freeze a new resistance state

Full loop: warming past $T_{\mathrm{COI}} \approx 210\,\mathrm{K}$ resets the COI microstructure $\Rightarrow$ optimal FM re-nucleation on re-cooling

Minor loop ($T_r < T_{\mathrm{COI}}$): COI domains kinetically frozen — fraction $\alpha(T_r)$ of FM domains fail to reform:

$$\eta_{\mathrm{FM}}^{\mathrm{minor}}(2\,\mathrm{K}) = \eta_{\mathrm{FM,max}}\left[1 - \alpha(T_r)\,F_\beta(T_r)\right]$$

$$\alpha(T_r) = \alpha_{\max}\cdot\sigma\!\left(\frac{T_r - T_{\mathrm{MIT}}^{\mathrm{warm}}}{\sigma_{\mathrm{warm}}}\right)\cdot\left[1 - \sigma\!\left(\frac{T_r - T_{\mathrm{COI}}}{\sigma_{\mathrm{COI}}}\right)\right]$$

$\sigma(x) = (1+e^{-x})^{-1}$: 1st sigmoid = onset above $T_{\mathrm{MIT}}^{\mathrm{warm}}$; 2nd = off above T_{COI}

Percolation resistance :

$$R(2\,\mathrm{K}) = \frac{1}{\left[\max\!\left(0,\ \frac{\eta_{\mathrm{FM}}-\eta_c}{1-\eta_c}\right)\right]^{1.5}/R_{\mathrm{metal}} + 1/R_{\mathrm{COI}}}$$

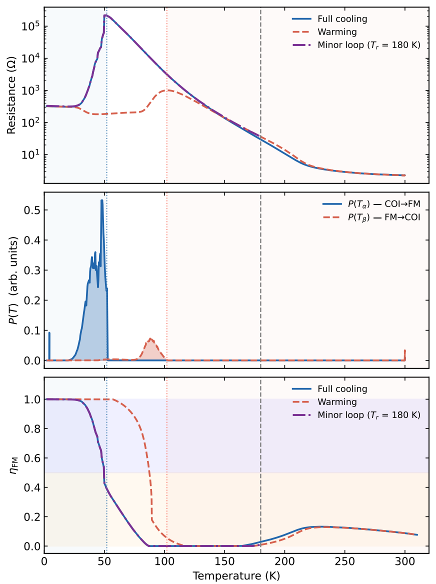
Three regimes of T_r:

- $T_r \lesssim 90\,\mathrm{K}$: $\alpha \approx 0$, $\eta_{\mathrm{FM}} \approx 1 > \eta_c$ $\Rightarrow$ FM intact, $R \approx 300\,\Omega$
- $90 \lesssim T_r \lesssim 210\,\mathrm{K}$: non-recovery window $\eta_{\mathrm{FM}} \approx 0.57 < \eta_c = 0.65$ $\Rightarrow$ sub-percolation $\Rightarrow$ $R \sim 10^9\,\Omega$
- $T_r > T_{\mathrm{COI}}$: microstructure reset $\Rightarrow$ full FM recovery, $R \approx 300\,\Omega$

Key result

Standard Preisach alone gives $\Delta R(2\,\mathrm{K}) \approx 6.7\,\Omega$.
Non-recovery + percolation: $\Delta R \sim 10^9\,\Omega$ — **7 decades**

Model prediction vs experiment



What the model predicts (no free parameters)

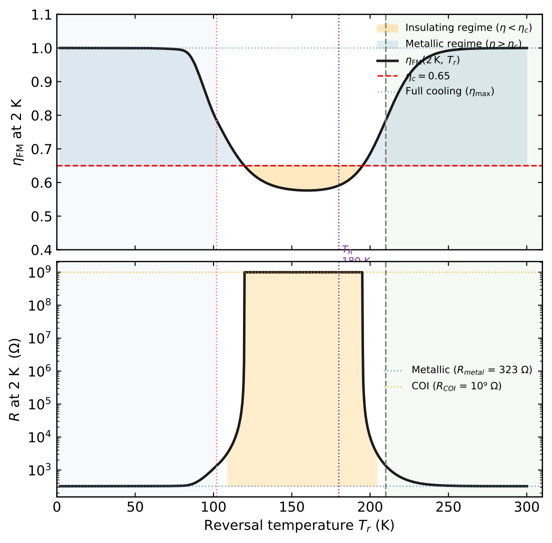
- **Minor-loop R–T:** predicted curve matches measured minor-loop trajectory to within experimental noise
- **$R(2\,\mathbf{K})$ vs T_r:** monotonically increasing — higher T_r leaves more COI frozen in on re-cooling
- **FM fraction $\eta_{\mathrm{FM}}(T)$:** crosses percolation threshold $\eta_c = 0.65$ only for $T_r > T_R$

Quantitative agreement

At $T_r = T_R = 180\,\mathrm{K}$: $\Delta R(2\,\mathrm{K}) = 6.7\,\Omega$
Matches the measured resistance jump cycle-to-cycle in Fig. 2b

The Preisach model makes **parameter-free predictions** of the non-volatile resistance state given only the R–T loop as input

Percolation and the bifurcation at $T_R = 180\,\mathrm{K}$



Why is $T_R = 180\,\mathrm{K}$ special?

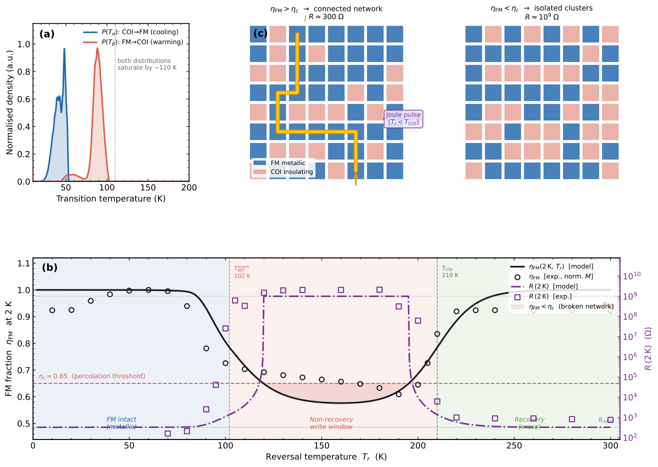
- FM phase forms a percolating network through the COI matrix
- Percolation threshold: $\eta_c \approx 0.65$
- **Key:** for $T_r < T_R$, re-cooling leaves $\eta_{\mathrm{FM}} < \eta_c$ — FM network is fragmented — insulating
- For $T_r > T_{\mathrm{COI}} = 210\,\mathrm{K}$, full FM percolation restored — **metallic**

Percolation resistance model:

$$R(2\,\mathrm{K}) \sim |\eta_{\mathrm{FM}} - \eta_c|^{-t}, \quad t = 1.5$$

Seven-decade resistance jump explained by crossing η_c

Experiment and model: the full picture



What the figure shows:

- **(a)** R–T minor loops: model (solid) vs experiment (symbols) — parameter-free agreement
- **(b)** $R(2\,\mathrm{K})$ vs T_r: 7-decade jump across the non-recovery window, reproduced quantitatively
- Dashed line at $T_R = 180\,\mathrm{K}$: the bifurcation point linking Joule heating to the memory mechanism

One model, two observables

Same Preisach + non-recovery + percolation prediction accounts for both the R–T trajectory *and* the final resistance state

Summary

Three-step mechanism for non-volatile switching in LPCMO:

1. **Joule heating**
   Current pulse heats locally to $T_{\mathrm{local}} \approx T_R = 180\,\mathrm{K}$, regardless of bath temperature
2. **Preisach minor loop**
   Re-cooling freezes a sub-percolation FM fraction in the COI matrix
3. **Percolation threshold**
   FM fraction $< \eta_c = 0.65 \Rightarrow$ 7-decade resistance jump — non-volatile

Key quantitative result

$T_{\mathrm{local}} \approx T_R = 180\,\mathrm{K}$ across all T_{base} in the COI phase, directly linking Joule heating to the thermal-pathway memory mechanism

Outlook

- Optimal pulse design: target T_r precisely
- Multilevel resistance states via T_r tuning
- Connection to STDP / synaptic plasticity at cryo temperatures
- Filament-free, scalable switching mechanism

Backup: numerical results i

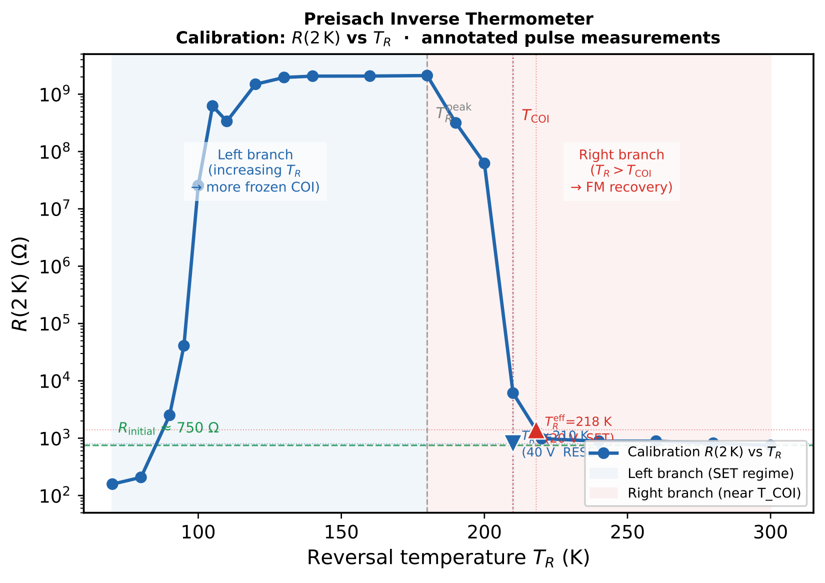
T_{base} (K)	I_{max} (mA)	V_{max} (V)	R_{switch} (Ω)	T_{local} (K)	ΔT (K)
10	—	—	—	≈ 100	≈ 90
40	—	—	—	≈ 108	≈ 68
60	—	—	—	≈ 94	≈ 34
77	—	—	—	≈ 130	≈ 53
100	—	—	—	≈ 148	≈ 48
125	—	—	—	≈ 162	≈ 37
150	—	—	—	≈ 171	≈ 21
170	—	—	—	≈ 179	≈ 9

Values from `joule_heating_results.csv`; fill in when Diego's raw I-V files are processed.

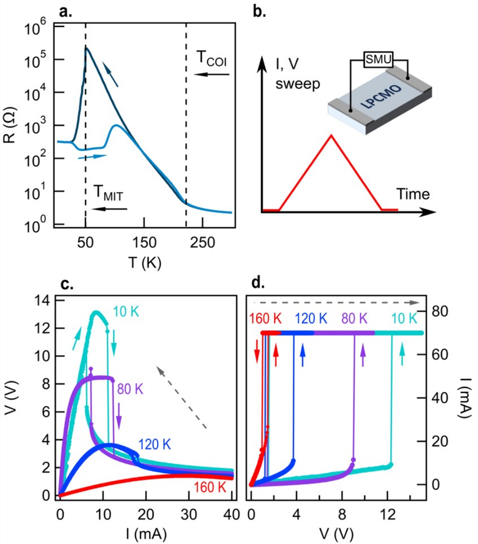
Backup: Preisach model parameters

Parameter	Symbol	Value	Source
Cooling MIT peak	$T_{\mathrm{MIT}}^{\mathrm{cool}}$	52 K	R–T data
Warming MIT peak	$T_{\mathrm{MIT}}^{\mathrm{warm}}$	102 K	R–T data
Minor-loop reversal	T_R	180 K	Fig. 3a of ms.
Charge-order boundary	T_{COI}	210 K	Fig. 1a of ms.
Percolation threshold	η_c	0.65	M(5 K) data
Conductivity exponent	t	1.5	percolation theory
FM resistance (2 K)	R_{FM}	$\sim 300\,\Omega$	R–T data
COI resistance (2 K)	R_{COI}	$\sim 10^9\,\Omega$	R–T data

Backup: calibration figure



The material: LPCMO and its giant hysteresis loop



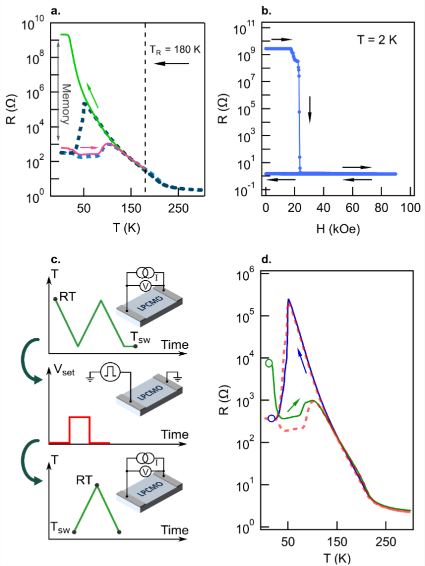
Resistance vs temperature: cooling (blue) and warming (red)

Key temperatures

Symbol	Value	Role
$T_{\mathrm{MIT}}^{\mathrm{cool}}$	52 K	phase transition (cooling)
$T_{\mathrm{MIT}}^{\mathrm{warm}}$	102 K	phase transition (warming)
T_R	180 K	bifurcation point
T_{COI}	210 K	full metallic recovery

The bifurcation at T_R: warming to 180 K then re-cooling leaves the device in a **different state** than warming to 210 K

The thermal pathway: minor loops in R–T

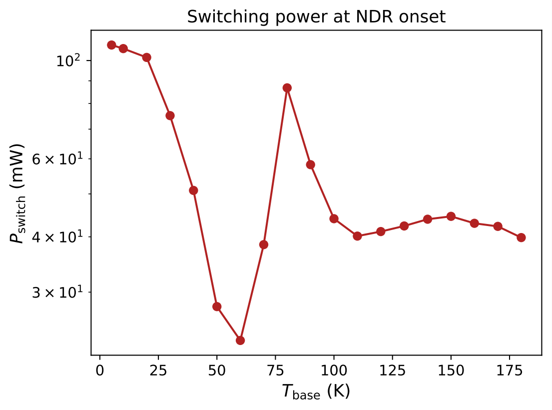


Minor loops (Joule pulse simulation):

- Warm to T_r then re-cool to 2 K
- $T_r < T_R$: device ends in higher R state
- $T_r > T_{\mathrm{COI}}$: full FM recovery

Bifurcation at $T_R \approx 180\,\mathrm{K}$

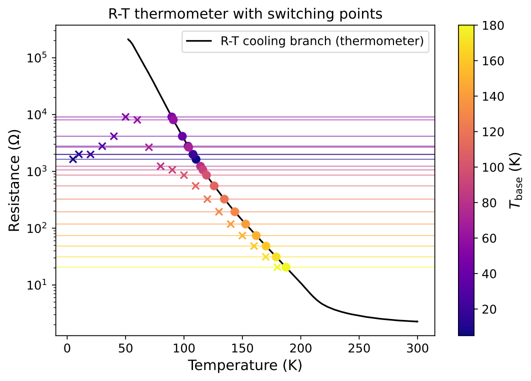
Switching power P_{switch} vs bath temperature



FM phase ($T_{\mathrm{base}} \lesssim 50$ K): $P_{\mathrm{switch}} \approx 100$ mW, minimum at T_{MIT}

COI phase ($T_{\mathrm{base}} \gtrsim 60$ K): P_{switch} rises again, local peak near 80 K

R–T as a local thermometer: Joule heating estimate

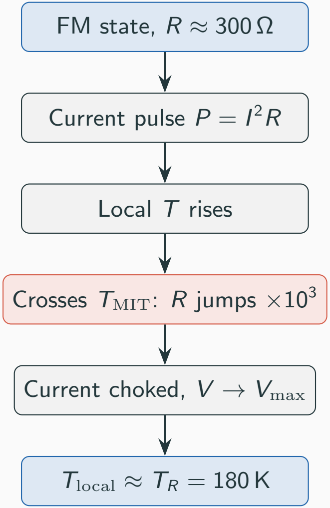


Method:

1. At NDR onset: $R_{\mathrm{switch}} = V_{\mathrm{max}}/I_{\mathrm{max}}$
2. Map onto high-T cooling branch of R–T
3. $T_{\mathrm{local}} = T(R_{\mathrm{switch}})\big|_{\mathrm{cooling}}$;
   $\Delta T = T_{\mathrm{local}} - T_{\mathrm{base}}$

T_{local} is a **lower bound**: filamentary flow means actual hotspot $>$ bulk estimate

Self-regulation: why T_{local} saturates at T_R



FM regime: T_{MIT} jump chokes current; T_{local} self-limits at 100–110 K

COI regime: no abrupt jump; power dissipates until COI→FM domains deplete at $T_R \Rightarrow$ natural convergence

The system acts as its own **thermal thermostat**