Transition metal oxides under strong electric fields, from resistive switching to artificial synapses and neurons

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CNRS – Université Paris-Sud
2017 @ UCSD-CNRS LIA

What is Resistive Switching (in TMOs) **?**

It is the sudden change in *resistance* due to a strong electric stress (*V* or *I*)

1) The change may be permanent, ie ***non-volatile***, and ***reversible***

(Obvious) Application as electronic memory device: **RRAM** (aka: ReRAM, OxRAM, memristors)

2) The change may be non-permanent ie ***volatile***

Less obvious applications are practical realizations of:

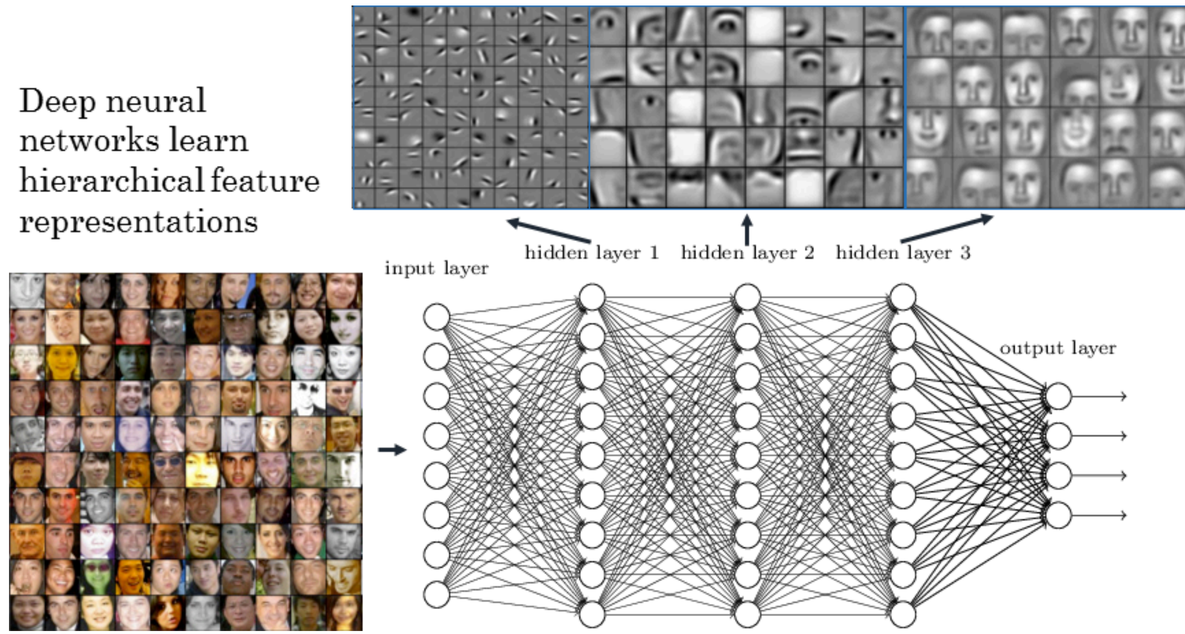
artificial synapses (1) and ***artificial neurons*** (2)

New functionalities of TMO materials

Neuromorphic circuits and computation is a very hot topic

Bio-chips (CMOS hardware)

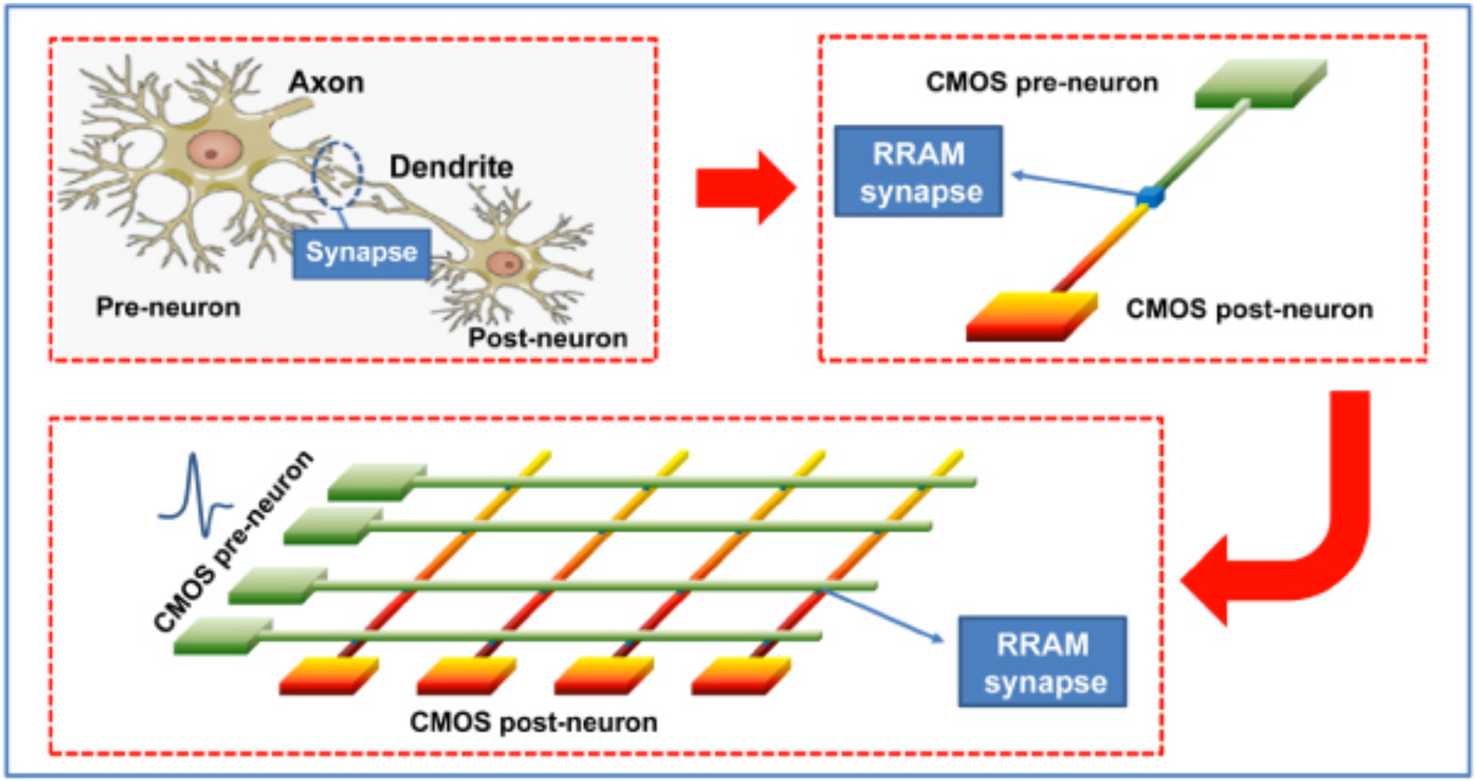
Deep Neural Networks (software)



- DARPA's Synapse Program
- EU Human Brain Project
- Facebook
- Google (DeepMind, AlphaGo)

human brain:
10^{11} neurons
10^{15} synapses

Novel electronic devices for neuromorphic systems



Park et al Nanotechnology ‘13

Neurons and **Synapses**:

Great oportunity for **oxyde electronics** !

1 - **Non-volatile** Resistive Switching

Basic concepts
Physical mechanism
Simple model

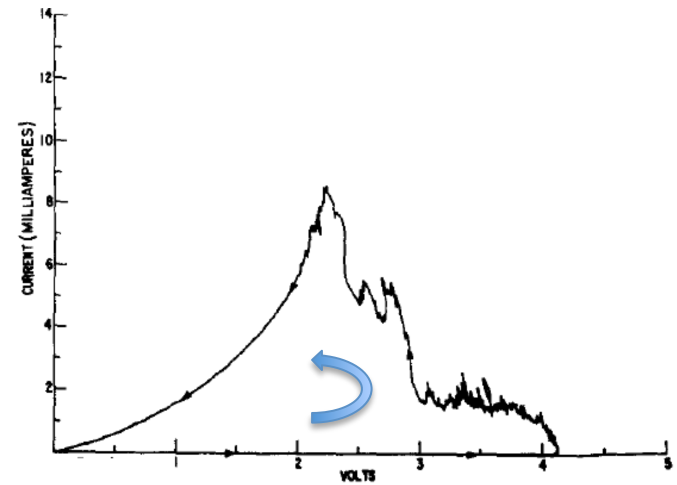
RS research is not new

it begun more than 50 years ago...

JOURNAL OF APPLIED PHYSICS VOLUME 33, NUMBER 9 SEPTEM

Low-Frequency Negative Resistance in Thin Anodic Oxide Films

T. W. Hickmott
General Electric Research Laboratory, Schenectady, New York
(Received February 5, 1962)



New Conduction and Reversible Memory Phenomena in Thin Insulating Films

J. G. Simmons; R. R. Verderber

Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences, Vol. 301, No. 1464 (Oct. 3, 1967), 77-102.

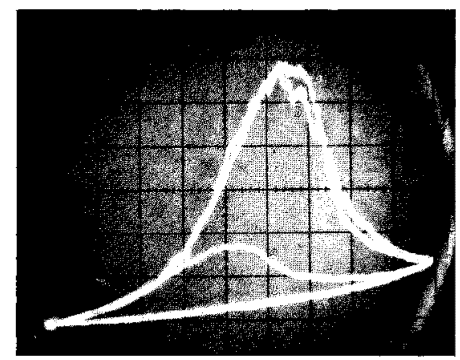
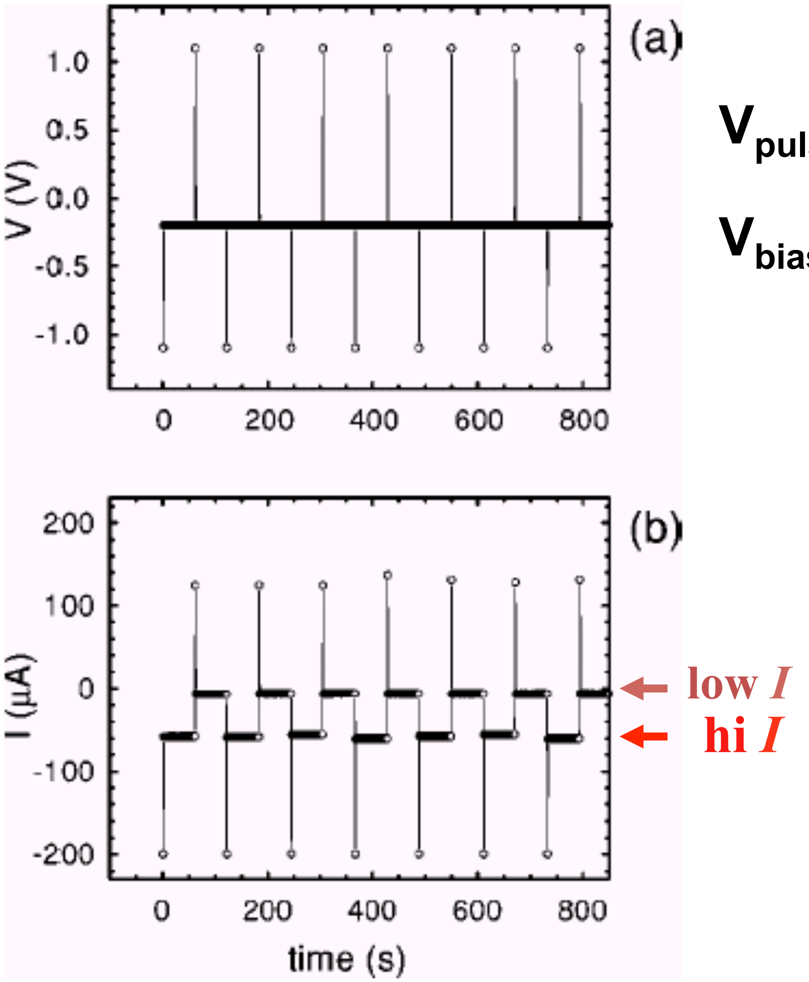
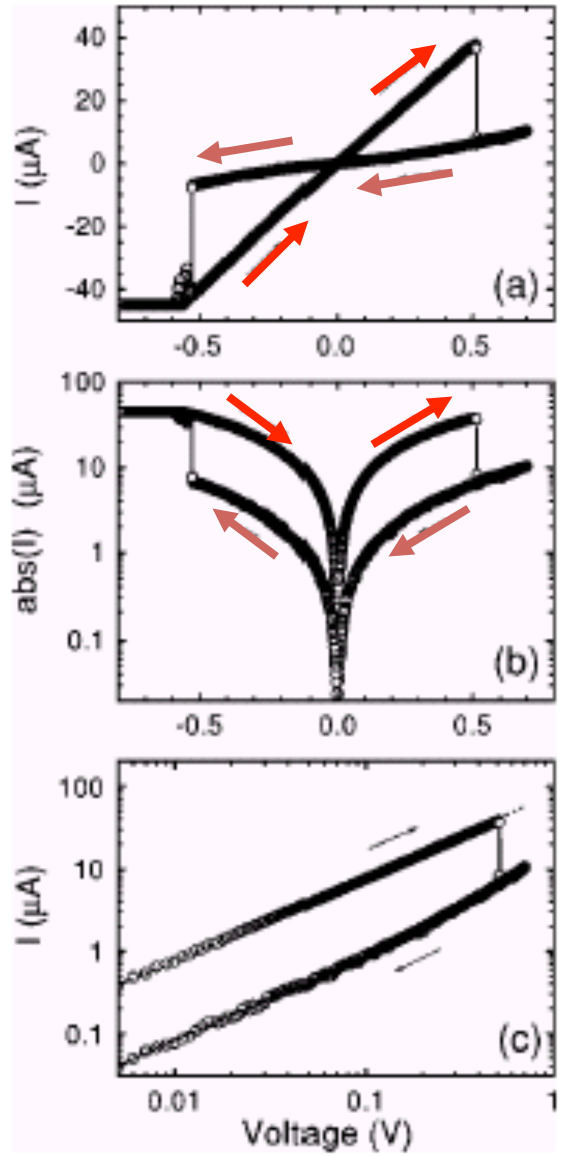


Figure 10. Photograph of X–Y oscilloscope V–I trace for a complete voltage cycle between en 0 and 9 V at (a) 300 °K and (b) 77 °K. Scales are x = 1 V/div, y = 10 mA/div.

switching

hysteresis (*I-V*)



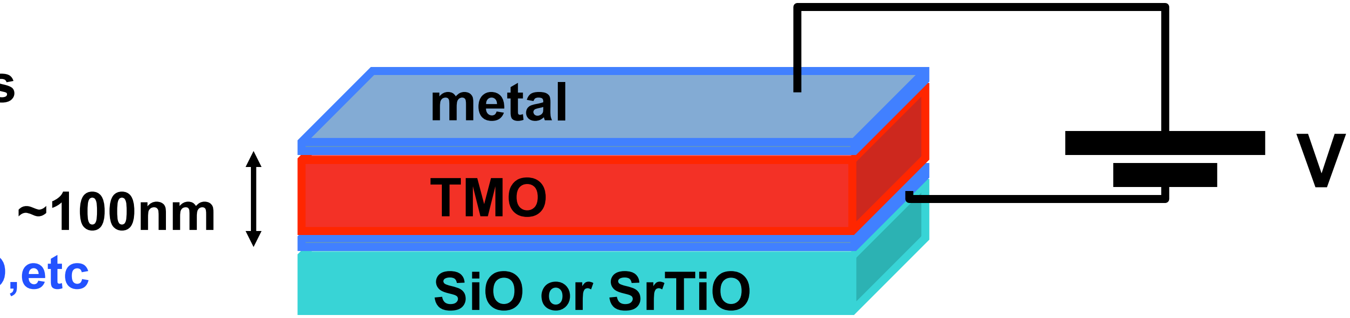


Cr-doped SrZrO

IBM group APL'00

Typical RRAM systems (aka ReRAM, OxRAM, memristor)

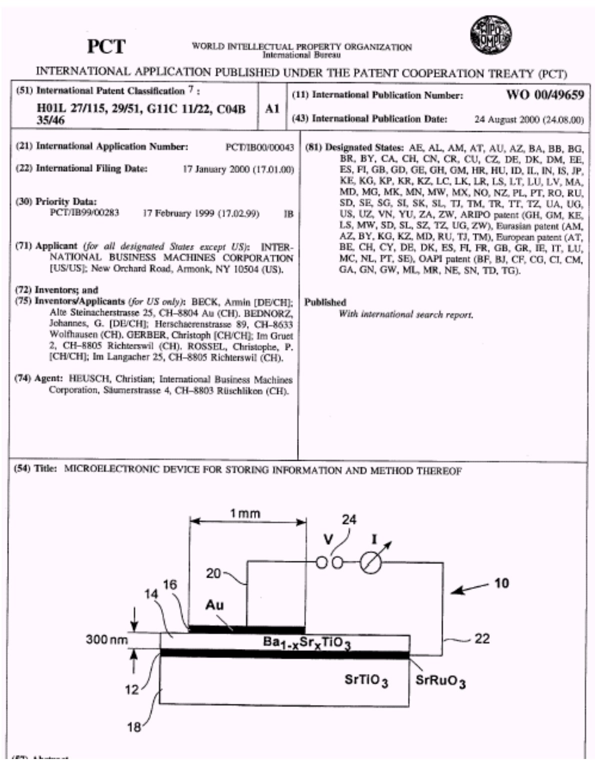
PLD made films



Au, Pt, Ag, SrRuO,etc

PrLaCaMnO, YBaCuO,LaCuO, SrTiO, SrZrO, TiO, CuO, NiO, etc…

IBM-Zurich patent



PCT

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Published
With international search report.

(54) Title: MICROELECTRONIC DEVICE FOR STORING INFORMATION AND METHOD THEREOF

UT Houston & Sharp group

Novell Colossal Magnetoresistive Thin Film Nonvolatile Resistance Random Access Memory (RRAM)

W. W. Zhuang[1], W. Pan[1], B. D. Ulrich[1], J. J. Lee[1], L. Stecker[1], A. Burmaster[1], D. R. Evans[1], S. T. Hsu[1], M. Tajiri[2], A. Shimaoka[2], K. Inoue[2], T. Naka[2], N. Awaya[2] K. Sakiyama[2], Y. Wang[3], S. Q. Liu[3], N. J. Wu[3], and A. Ignatiev[3]

1: Sharp Laboratories of America, 5700 NW Pacific Rim Blvd, Camas, WA 98607, USA
2: Sharp Corporation, IC Group, 2613-1 Ichinomoto-cho, Tenri, Nara 632, Japan
3: Texas Center for Superconductivity and Advanced Material , University of Houston, Houston, Texas 77204-5002 USA

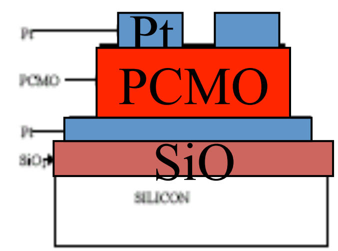


Fig.2 Spin-coating deposited (MOD) memory resistor structure. Both top and bottom electrode is Pt. The thickness of PCMO is 100nm to 200nm

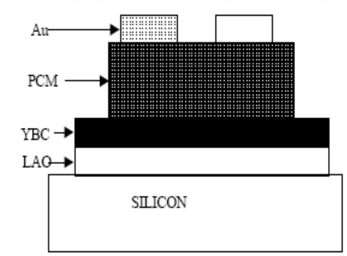


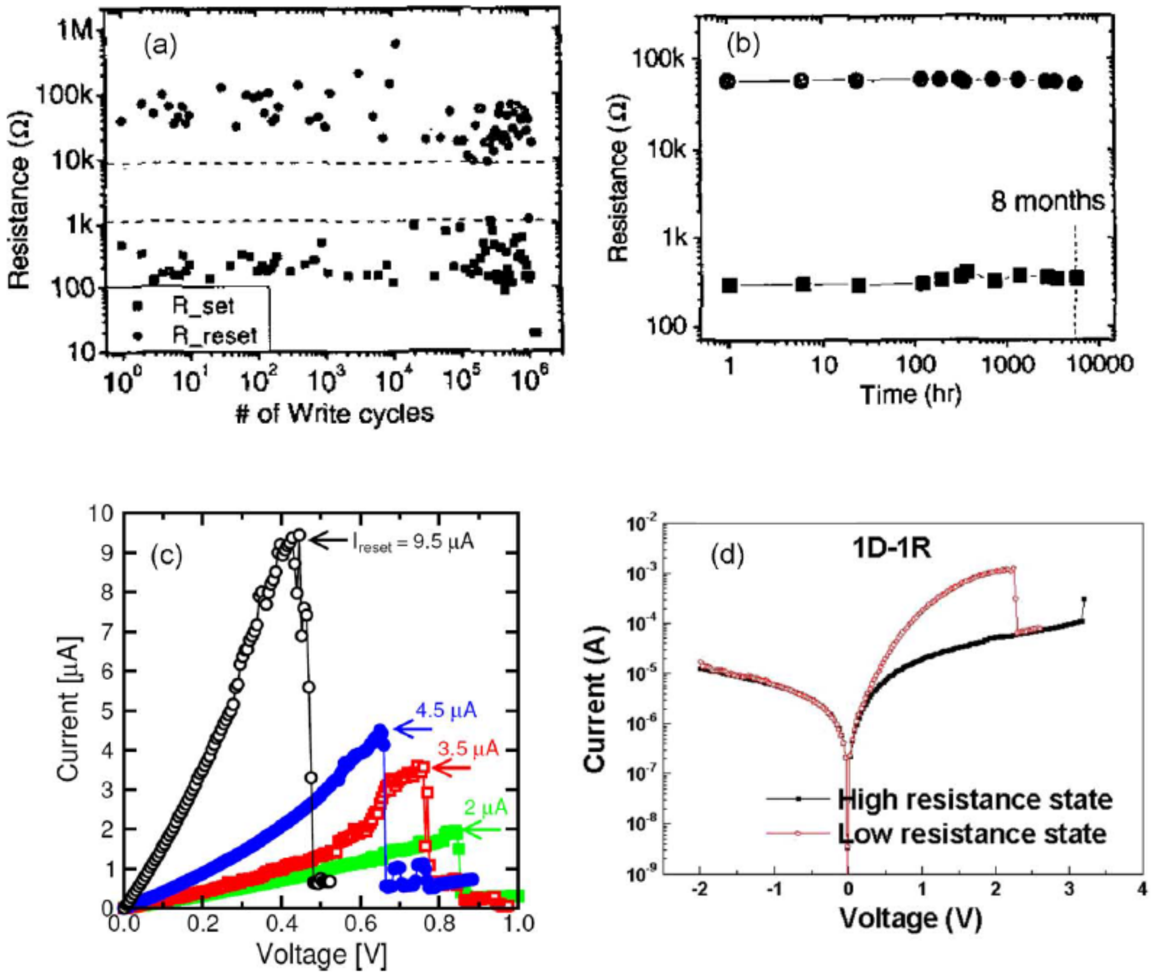
Fig.1 Pulsed Laser Deposited (PLD) test memory resistor structure. The memory material is PCMO ($Pr_{0.7}Ca_{0.3}MnO_3$). The double bottom electrode is formed with YBCO ($YBa_2Cu_3O_{7-x}$) on LAO ($LaAlO_3$)

Key issues

- Endurance
- Retentivity
- Resistance on-off ratio
- Power dissipation
- Commutation speed

NiO
Baek el al 2004

High Endurance **High Retentivity**

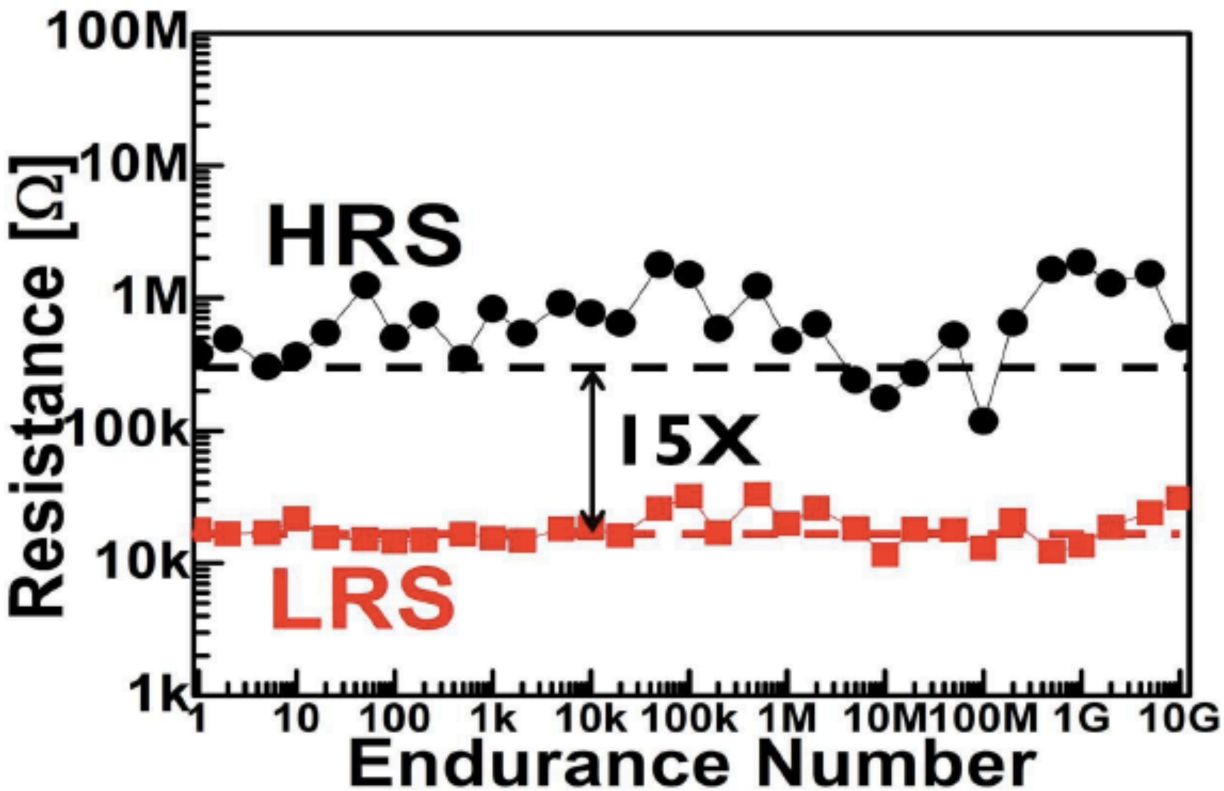


High Resistance on-off ratio

Low Power dissipation

Wong et al IEEE 2012

Fast commutation speed   nsec



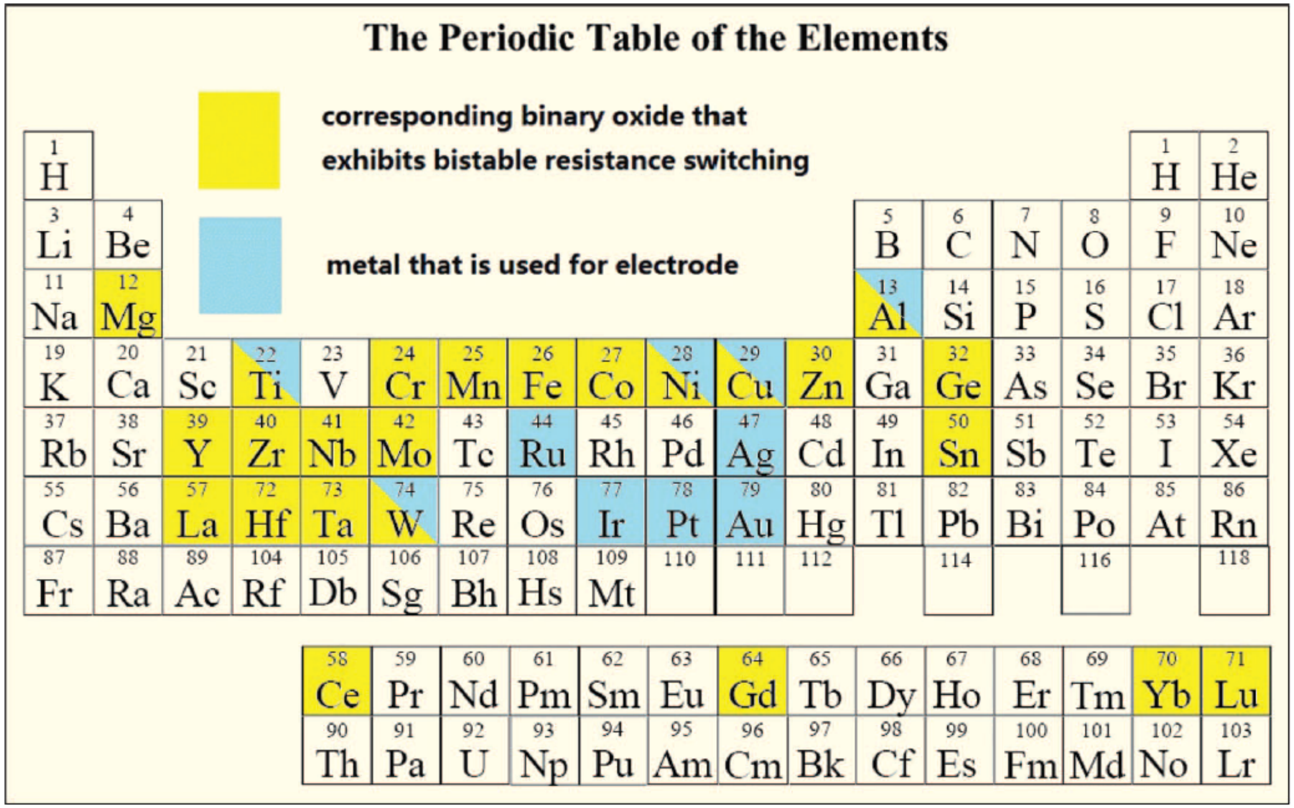
By balancing the SET pulse WL=1V, BL=1.8V, 5ns and RESET pulse WL =3V, SL=1.8V, 10ns, 10^{10} pulse endurance could be achieved on 40nm Hf/HfO_2 1T1R devices.

YY Chen et al 2012

Key issues

- Endurance
- Retentivity
- Resistance on-off ratio
- Power dissipation
- Commutation speed

- **Physical mechanism ?!?**
  Voltage – time dilema



Astonishingly universal!

Wong et al IEEE 2012

Oxygen vacancies
are involved in RS!

Not surprising…
Universal functionality
of TMOs
and
Universal presence
of O in TMOs

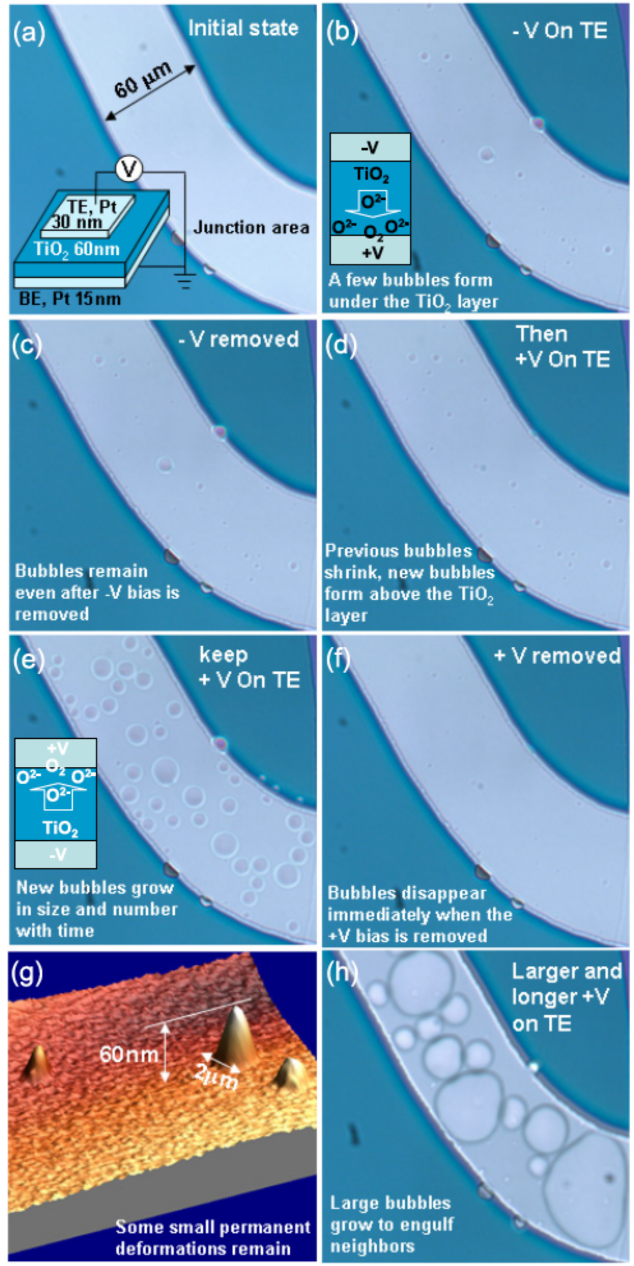
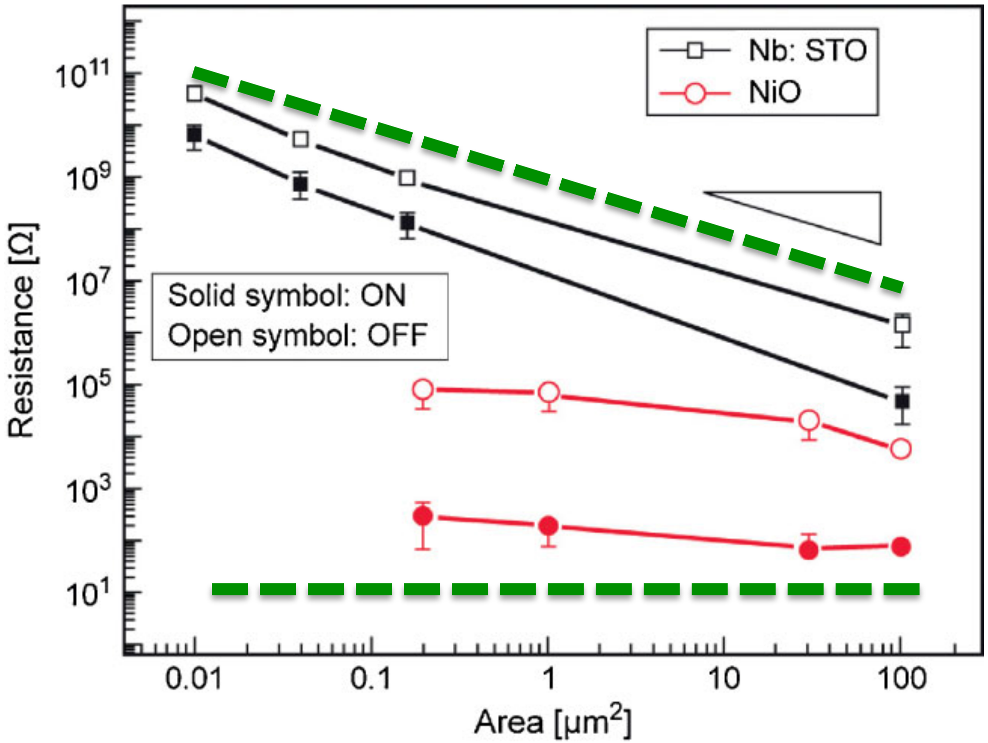


Figure 2. Gas bubble behavior under (b), (c) negative bias, then under (d)–(h) positive bias. (g) Atomic force micrograph of eruption features remaining after the bias voltage was removed. Videos of bubble evolution are available in the supplemental information (available at stacks.iop.org/Nano/20/215201).

Oxygen bubbles!

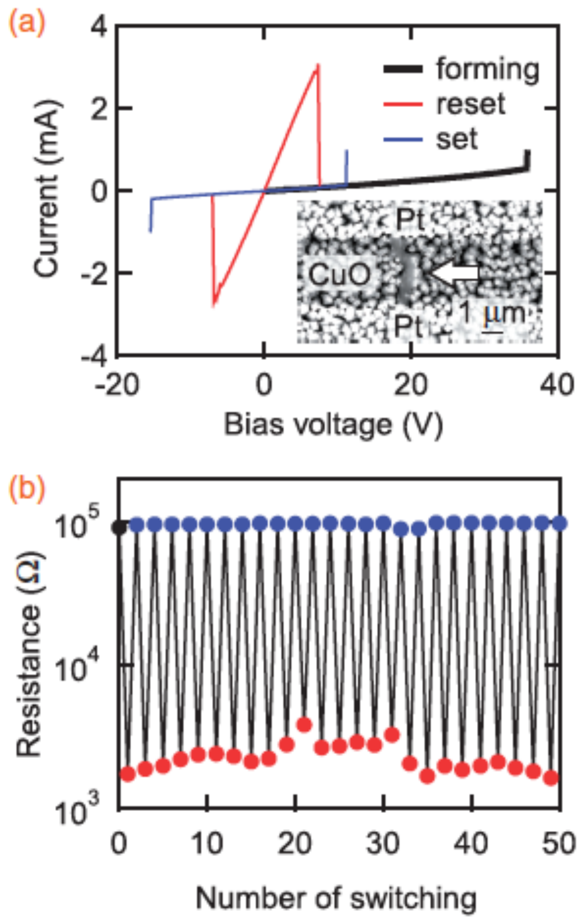
JJ Yang et al 2009

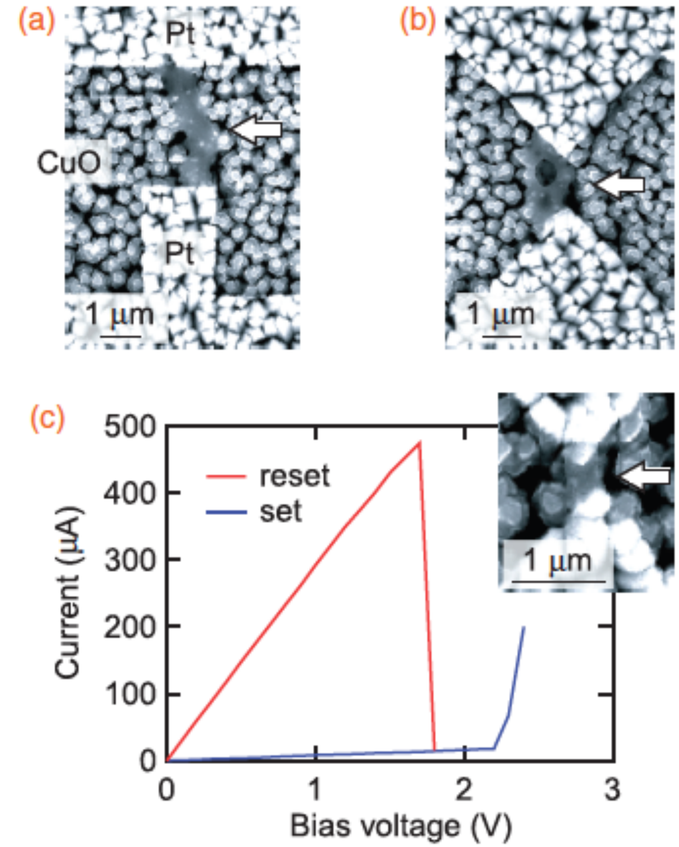
Evidence of different type of switching
Filamentary and non filamentary conductive structures



Sim et al 2005

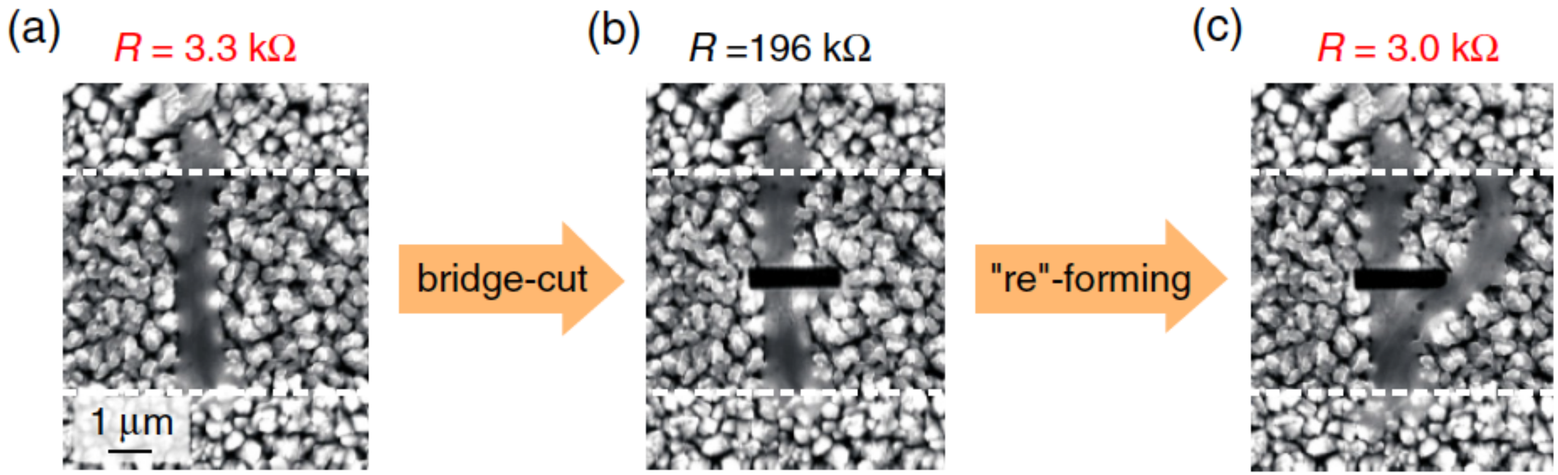
Observing the filaments (CuO)

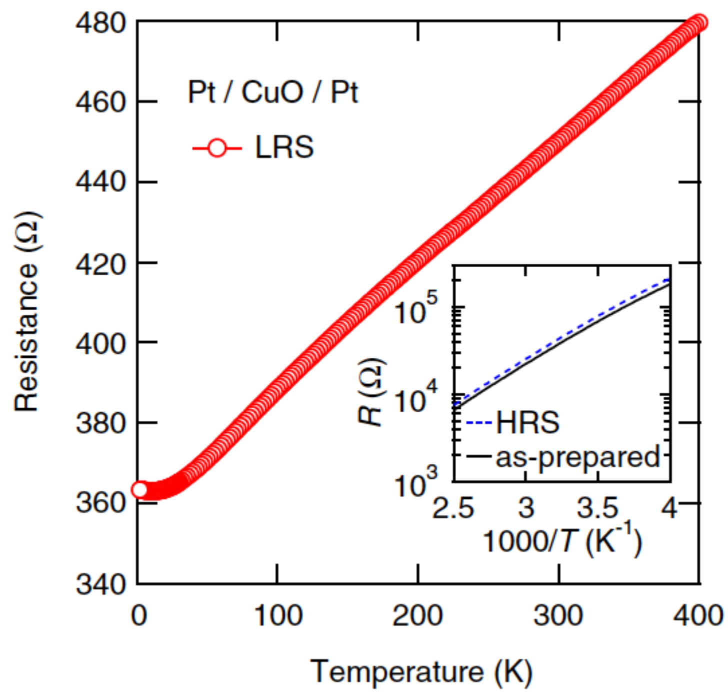




Fujiwara et al 2008

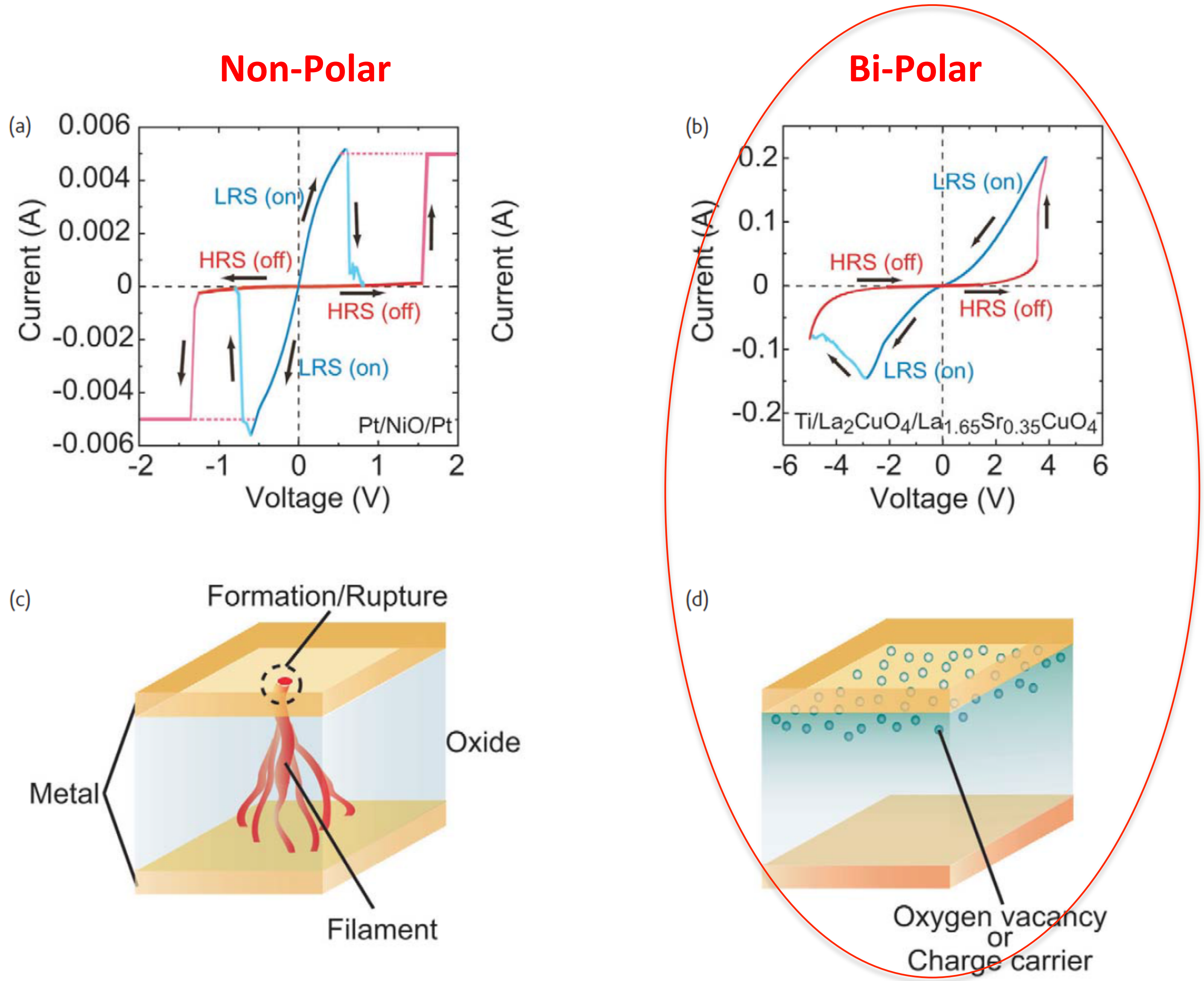
LRS **HRS** **LRS**





Fujiwara et al 2008

There are two main types of Non-volatile Resistive Switching:



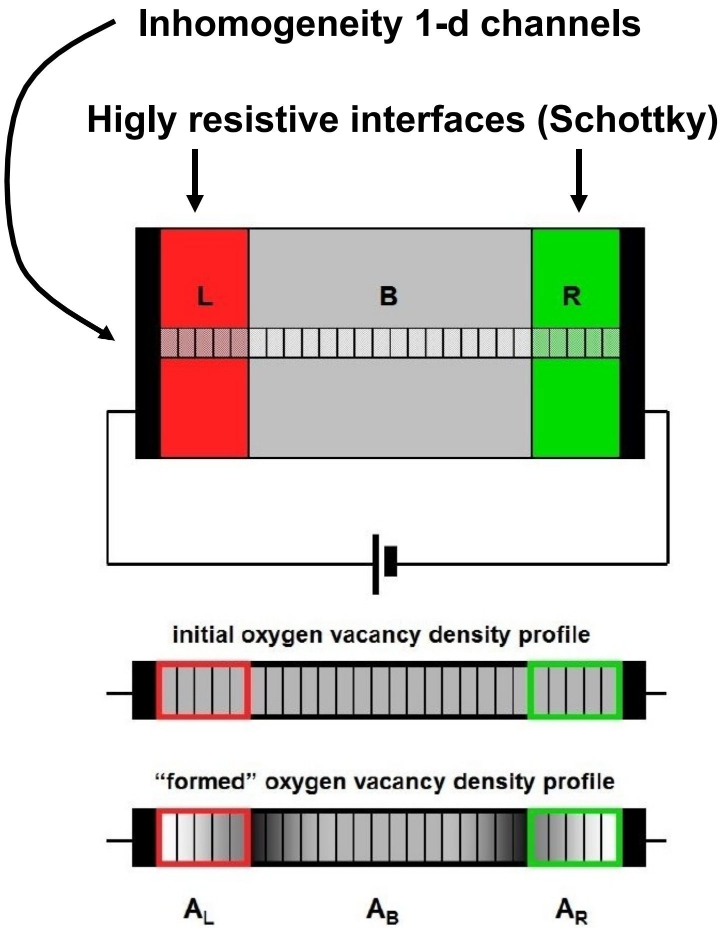
A Sawa, Mat Today (2008)

A simple model for bi-polar RS

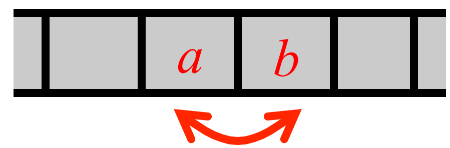
Voltage-enhanced Oxygen drift (VEOD) model

MR, Sanchez, Weht, Levy, Acha PRB '10

(see also Jeong,Schroeder and Waser et al PRB'09 and R. Meyer et al NVMTS2008)



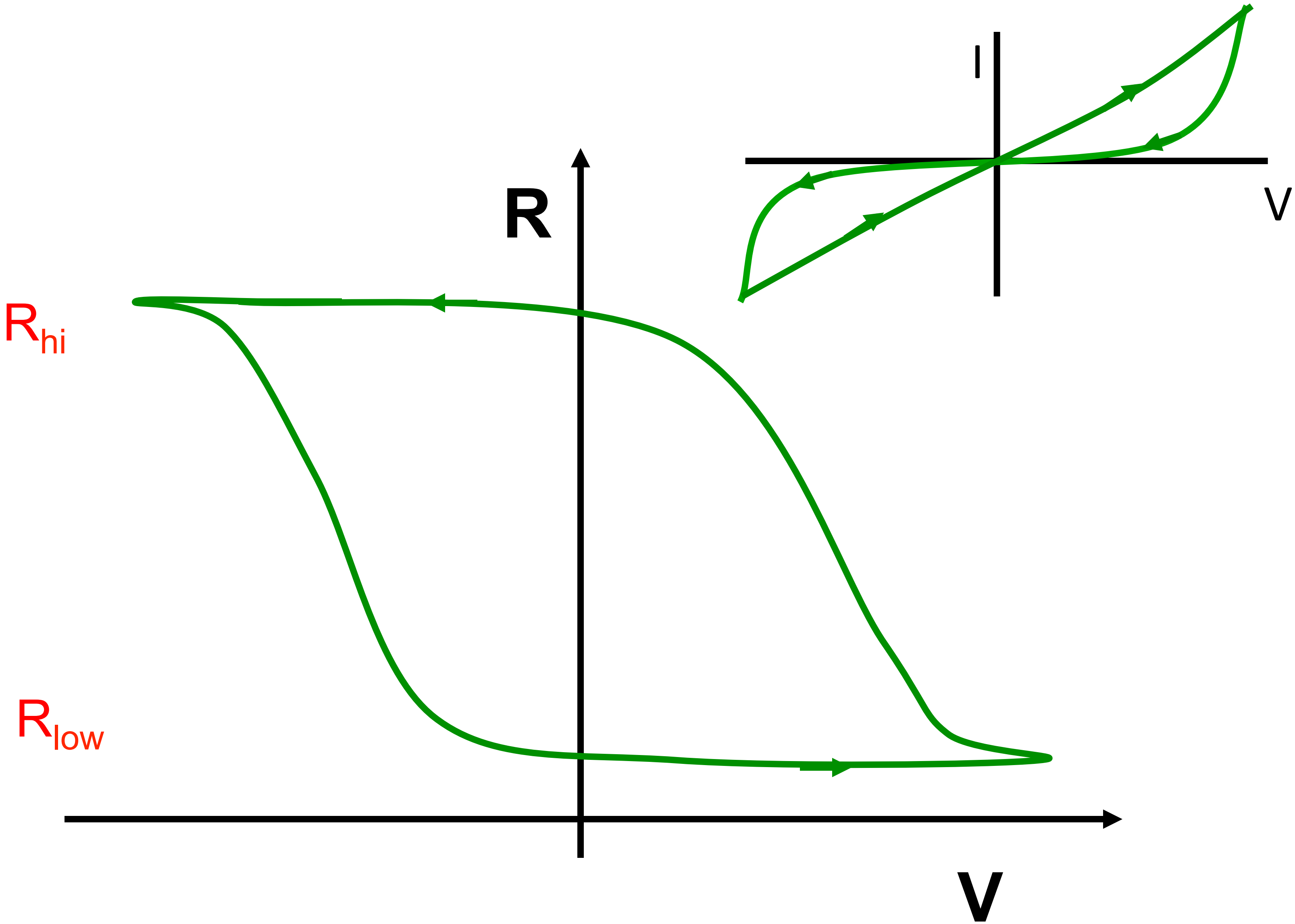
Oxygen drift (enhanced by V)



$$p_{ab} = \delta_a (1 - \delta_b) \exp(-V_0 + \Delta V_a)$$

$$\rho_i = A_\alpha \delta_i$$

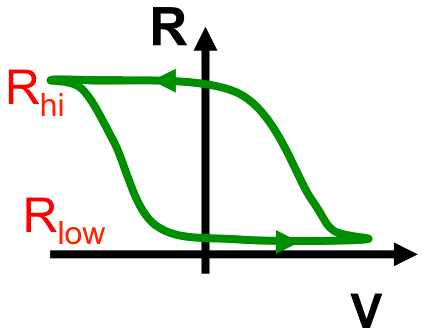
$$R_T = c \sum_{i=1}^{N} \rho_i = \sum_{\alpha} \sum_{i \in \alpha} A_\alpha \delta_i$$



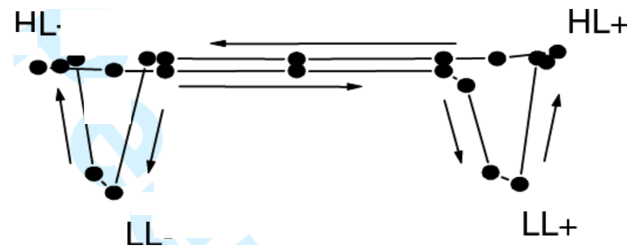
R
V
R_{hi}
R_{low}
I
V

Non-trivial test: “Table with legs mystery”

MR, Sanchez, Weht, Levy, Acha PRB ’10

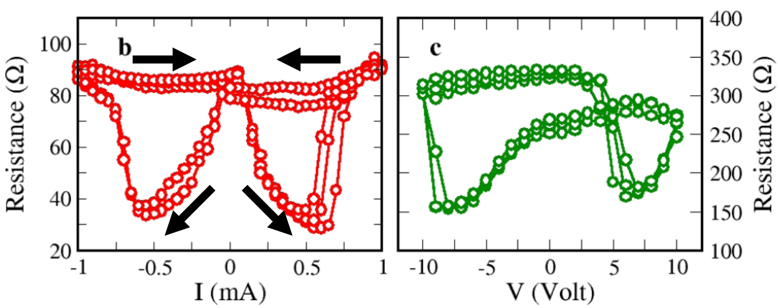


R vs V data on LCMO



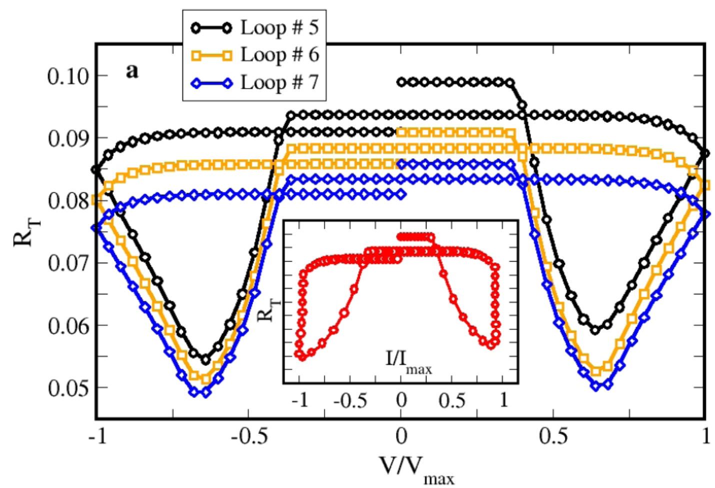
Chen Ignatiev, APL’05

experiments

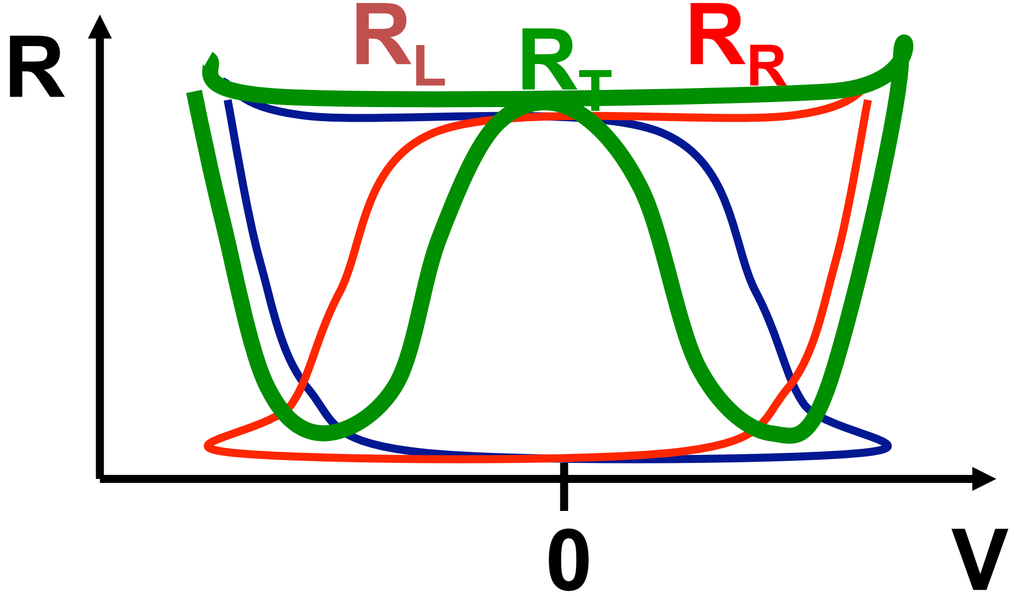


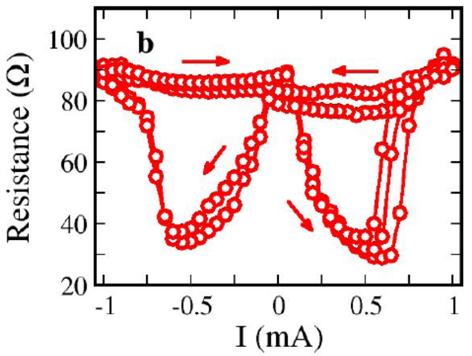
PLCMO **YBCO**

model simulations



Sum of two symmetric interface contributions



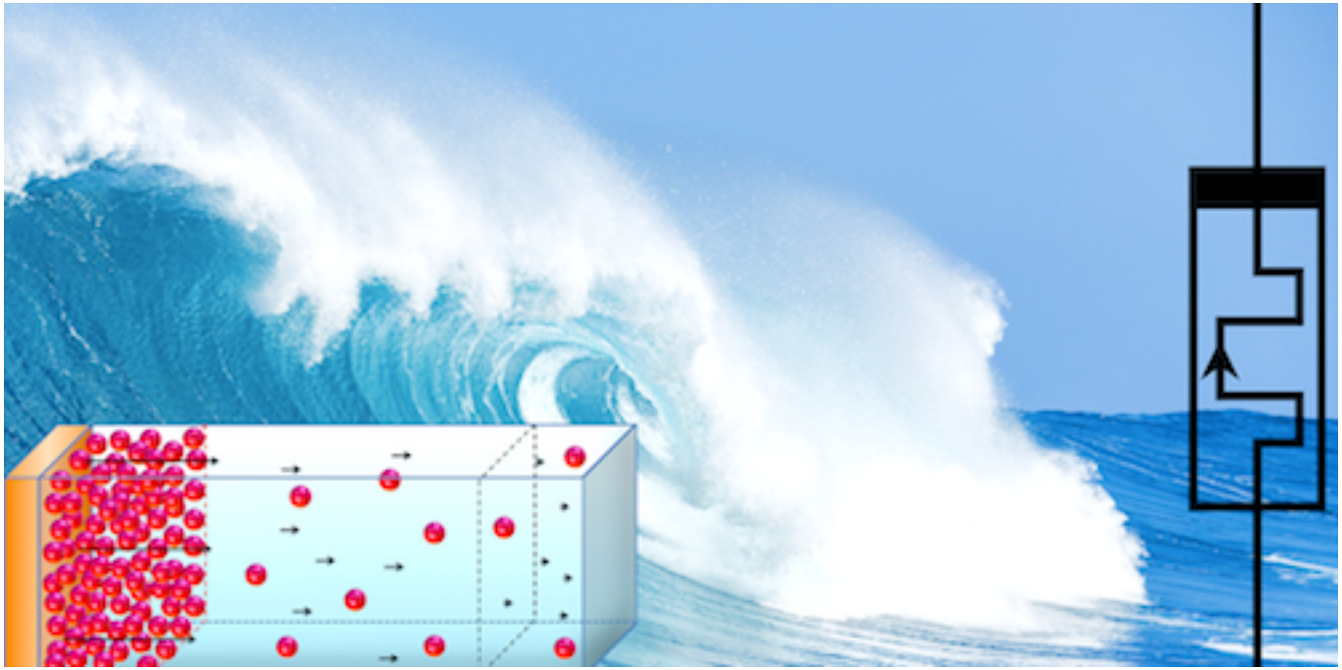


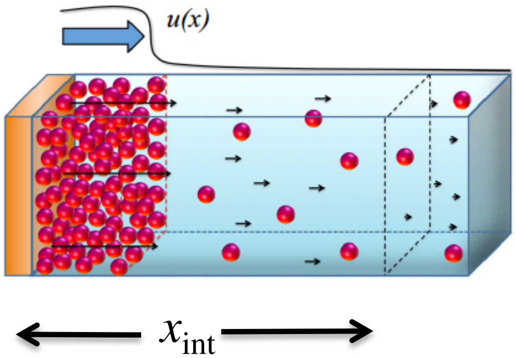
Some new theoretical insight

Shock Waves and Commutation Speed of Memristors

Phys. Rev. X 6, 011028 (2016)

Physics Synopsis: Waves That Shock Resistance





$$\delta R(t^*) = 1 - \ln\left(1 + t^*/\tau_2\right)/\ln(2)$$

$$\tau_2(I) = \left(x_{\mathrm{int}}^2/DIR_{\mathrm{HI}}\right)\exp\left(-IR_{\mathrm{HI}}/x_{\mathrm{int}}\right)$$

x_{int} width of Schottky barrier

R_{HI} resistance of HI-R state

Strong correlation effects?

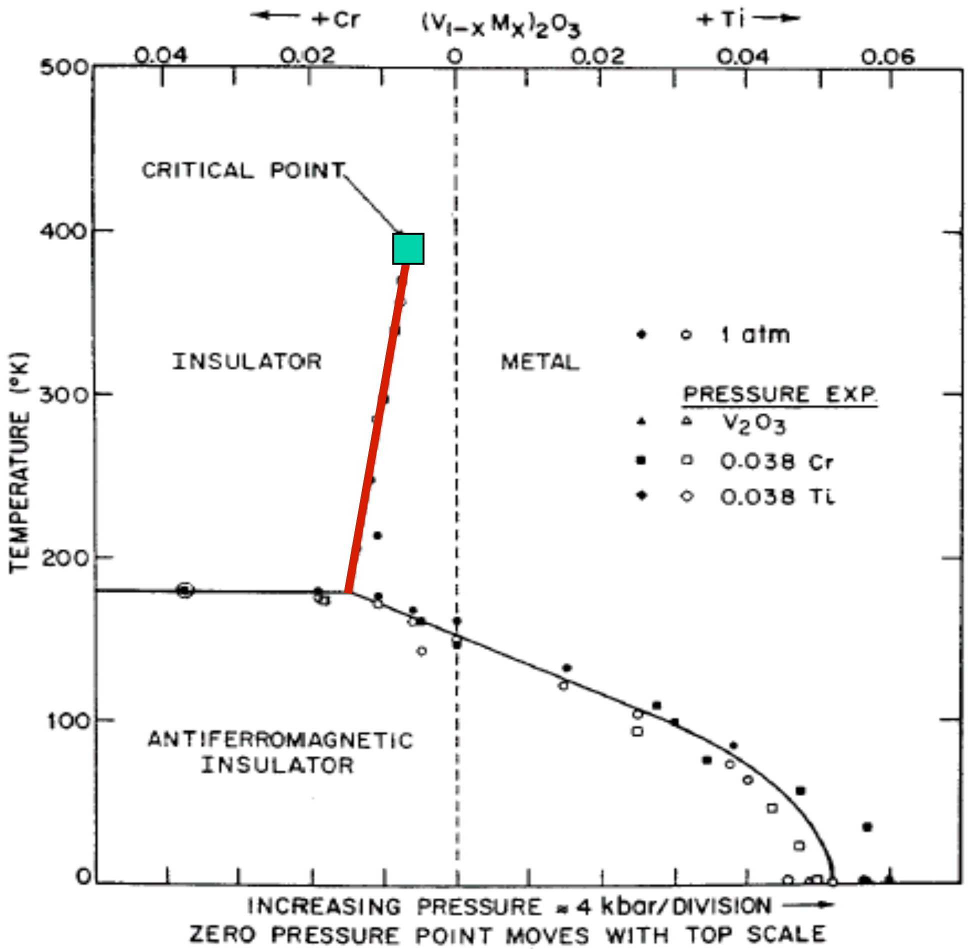
2 – Volatile Resistive Switching

in 3-dimensional **Mott** insulators

« Mottronics »

The classic example: Mott transition in V_2O_3

T



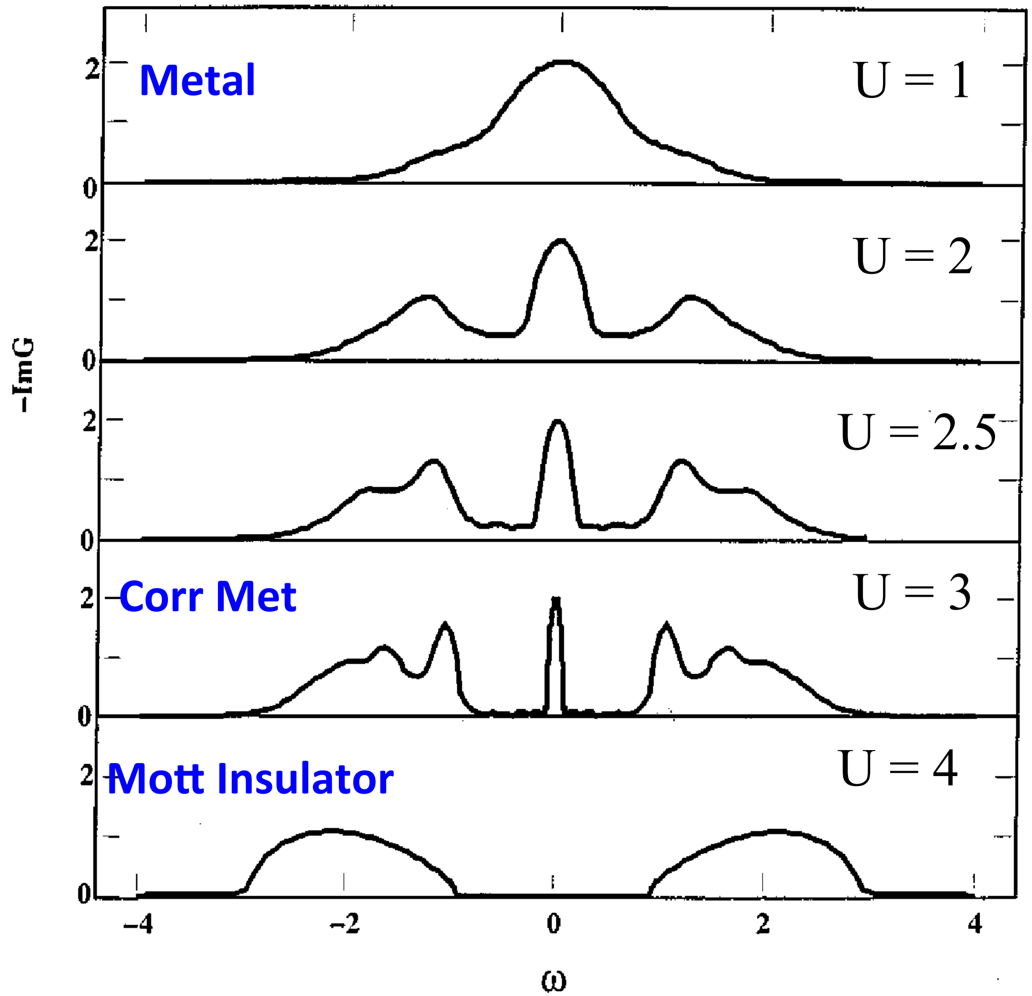
pressure or chemical substitution

McWhan et al PRB '71 '73

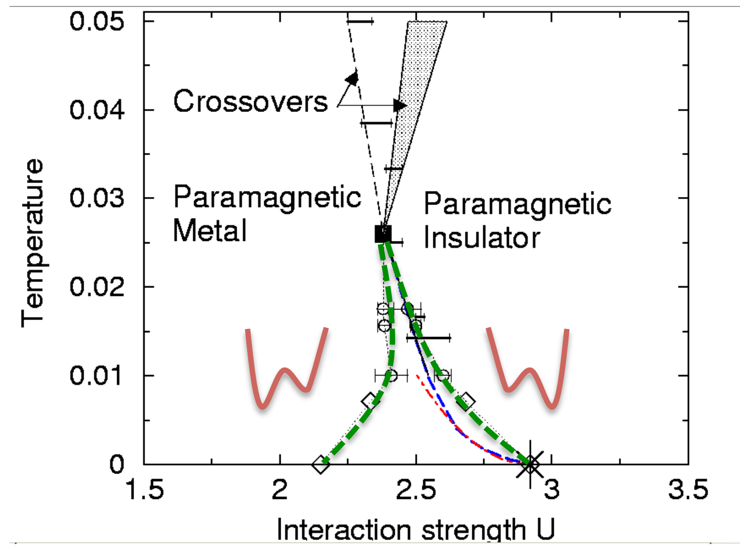
DMFT of the Mott – Hubbard transition

Georges, Kotliar, Krauth & MR, RMP '96

Georges, Kotliar PRB '92
Zhang, MR, Kotliar PRL '92



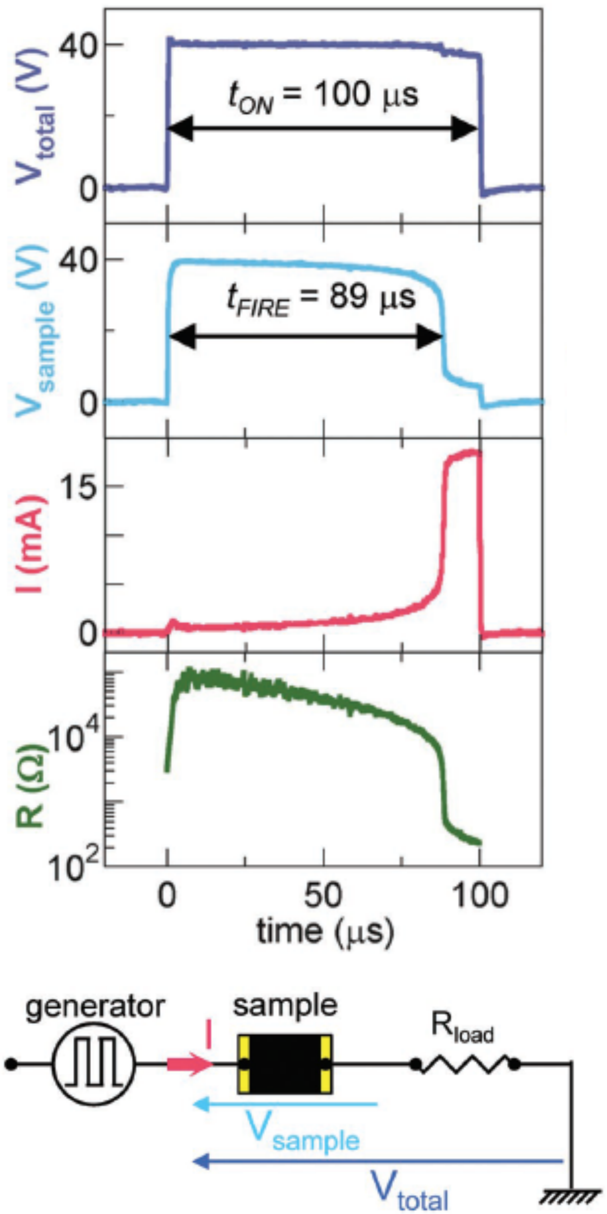
Coexistance region: 2 solutions



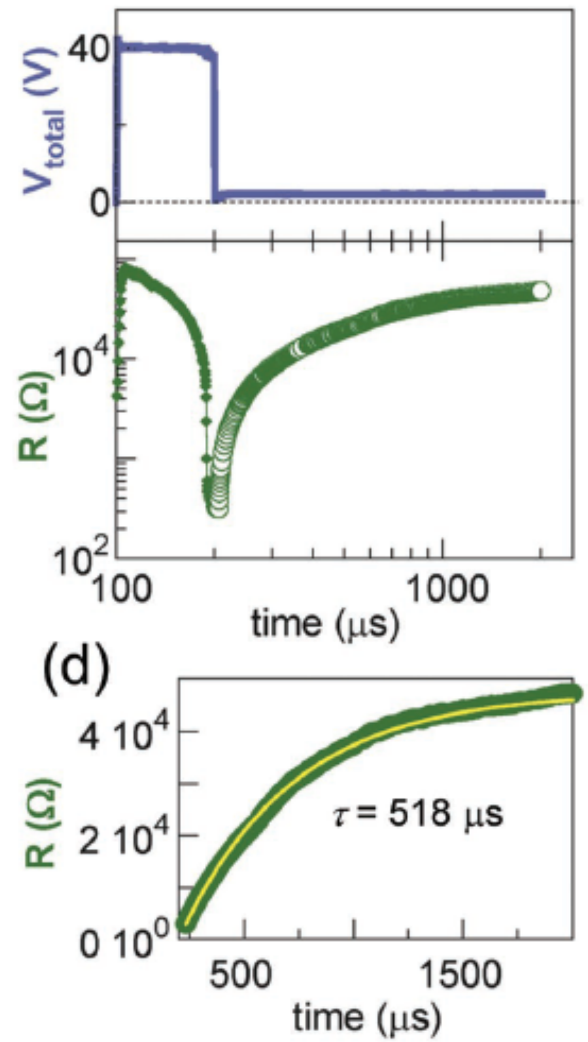
Mott physics + electronics
« Mottronics »

Applying strong E-fields to
Mott systems

Volatile RS in 3D Mott insulators



Stoliar et al Adv Func Mat 2016

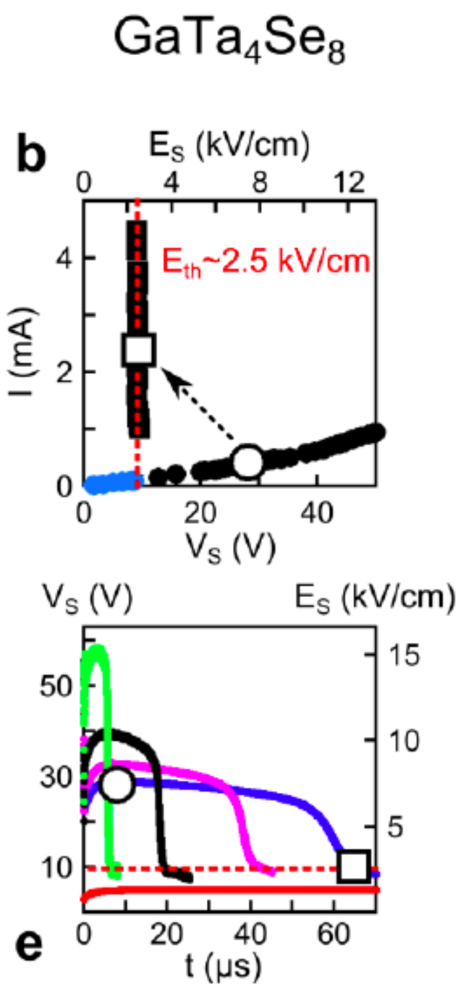


Perfectly reproducible!!

$GaTa_4Se_8$ single x-tal @ 74K

A. Camjayi, et al PRL 2014

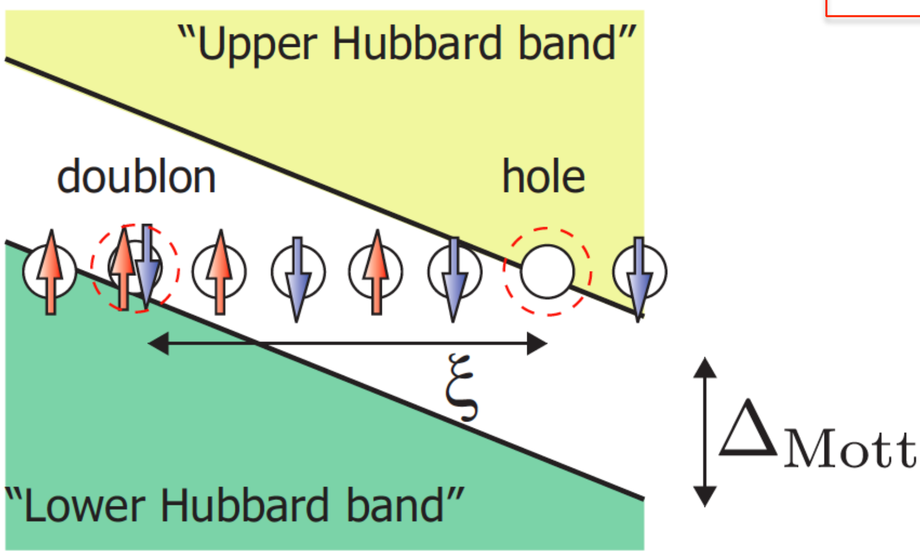
Volatile RS in 3D Mott insulators



P. Stoliar et al Adv. Mater. (2013)

What is the origin of the Mott electric-breakdown?

Hubbard model 1D

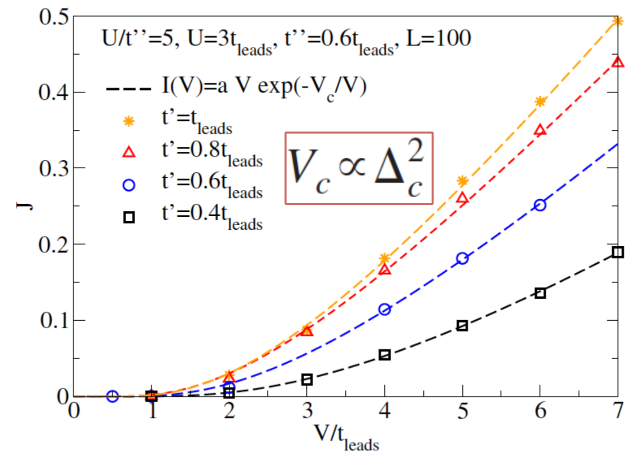


$F_{\rm th} \propto \Delta^2_{\rm Mott}$

$1/\xi \sim \Delta_{\rm Mott}$ (1D)

$$F_{\rm th} \simeq \frac{\Delta_{\rm Mott}}{2\xi}$$

$$J \sim \Gamma_p = \frac{F_0}{2\pi} \exp\left(-\pi \frac{F_{\rm th}}{F_0}\right)$$



T. Oka et al. '03 '05 '10 '12
F. Heidrich-Meisner et al '10

M. Eckstein et al. '09 '10 '11 (DMFT)
M. Schiro and M. Fabrizio '10 (TGW)
A. Amaricci et al. '12 (DMFT)

??? $1/\xi \sim \Delta_{\rm Mott}$ (3D)

??? $\xi \sim \mu$m (3D)

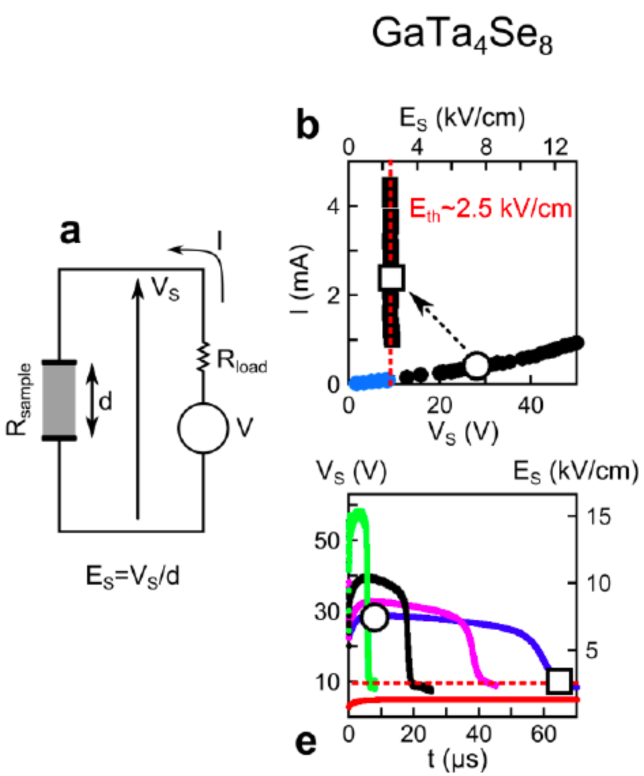
$\Delta \sim 10^{-1}$ eV
$\xi \sim 1$ nm = 10^{-7} cm
$F_{Th} \sim 1$ MV/cm

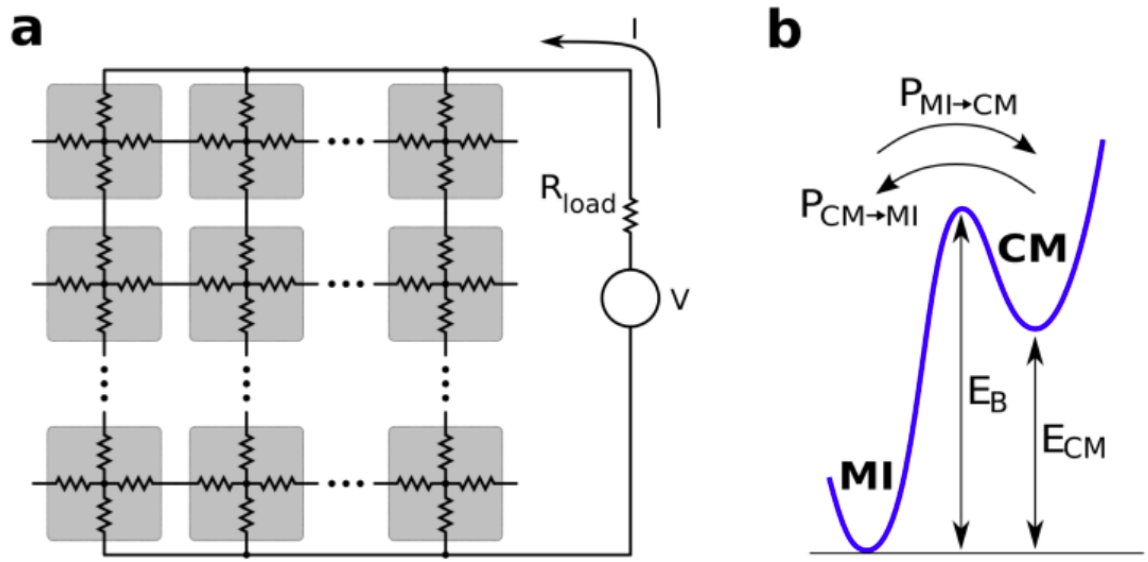
$E_{Th} \sim 1$ KV/cm !!!!

Model of the Mott resistive transition

(with inspiration from DMFT)

P. Stoliar et al Adv. Mater. (2013)





Two states: MI – Mott insulator

CM – Correlated metal

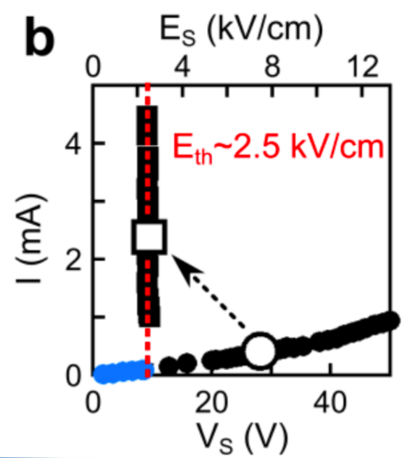
$R_{MI} >> R_{CM}$

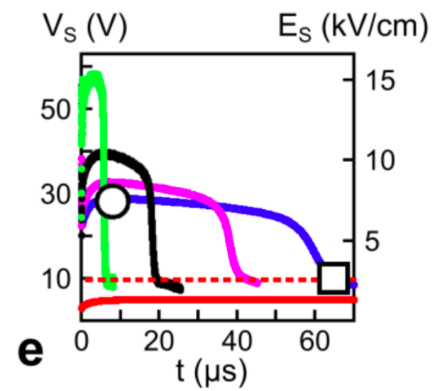
$P_{\text{MI->CM}}$ and $P_{\text{CM->MI}}$ are transition probabilities

$$P_{\text{MI}\rightarrow\text{CM}} = \nu e^{-(E_B - q\Delta V)/kT} \qquad P_{\text{CM}\rightarrow\text{MI}} = \nu e^{-(E_B - E_{CM})/kT}$$

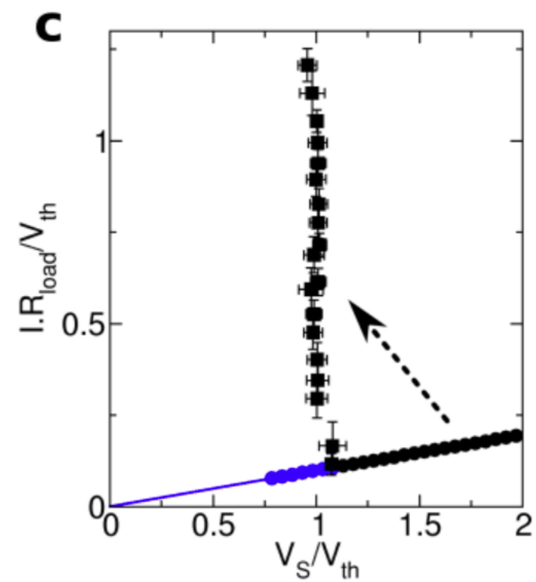
Model results: Threshold Mott resistive transition

Experiment

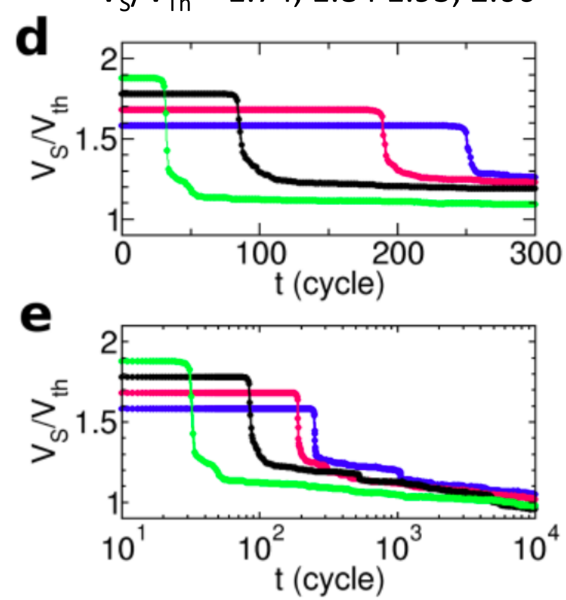




Theory

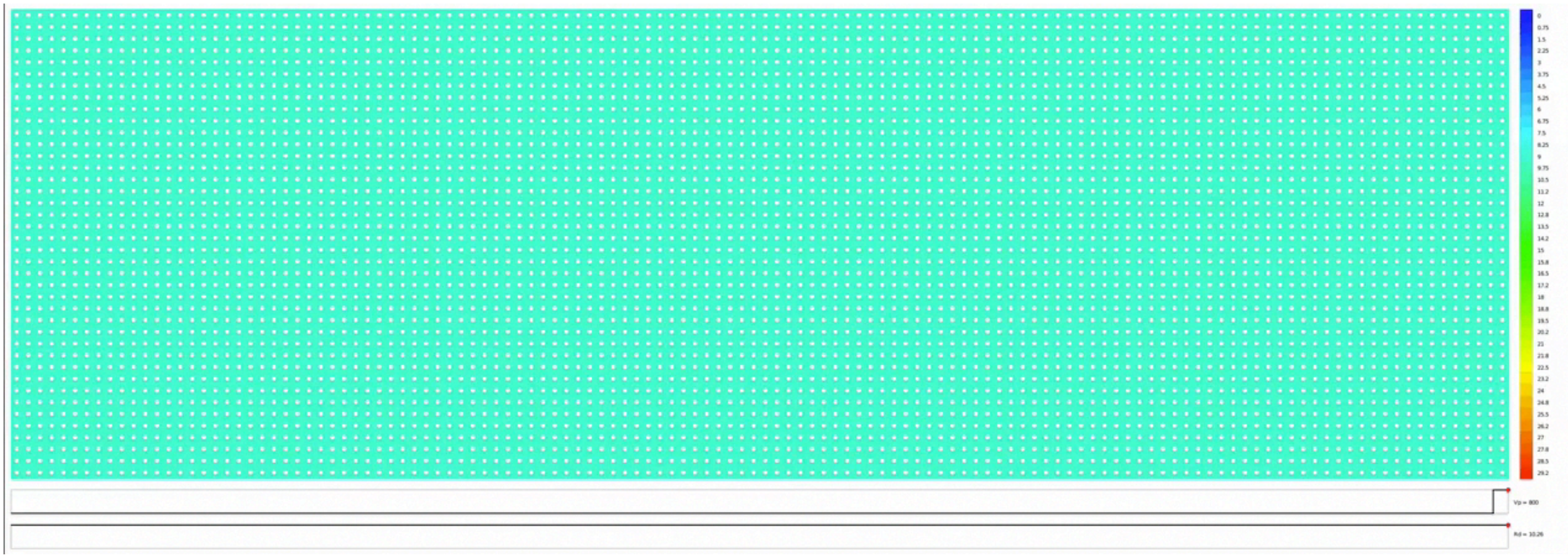


V_S/V_{Th} = 1.74, 1.84 1.95, 2.06



How the transition evolves in time?

Top electrode

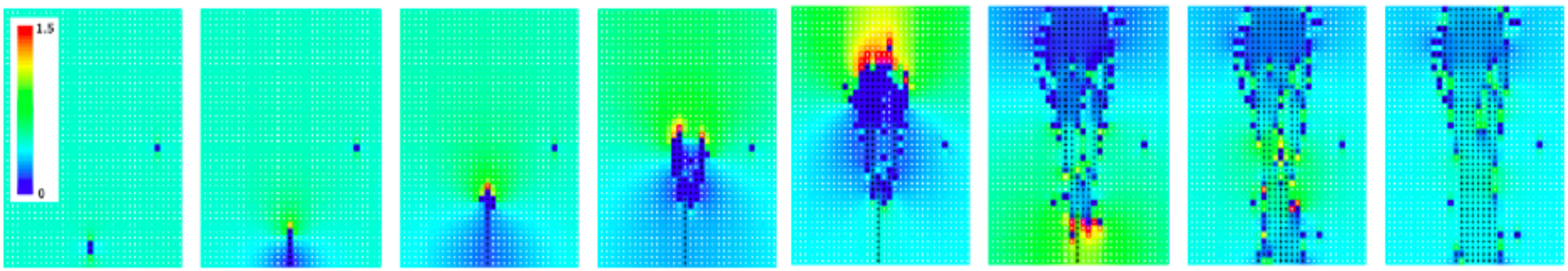


Bottom electrode

Each pixel is a cell of the resistor network model

Color intensity indicates the local ΔV drops (ie local E)

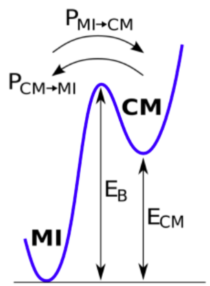
How the transition evolves in time? (snapshots)



time

Model validation

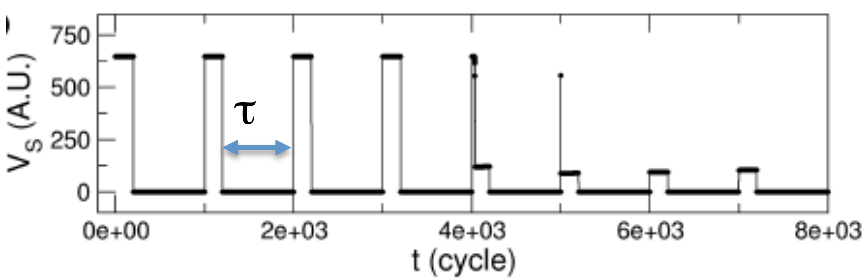
P. Stoliar,MR, et al., Adv. Mat. (2013)

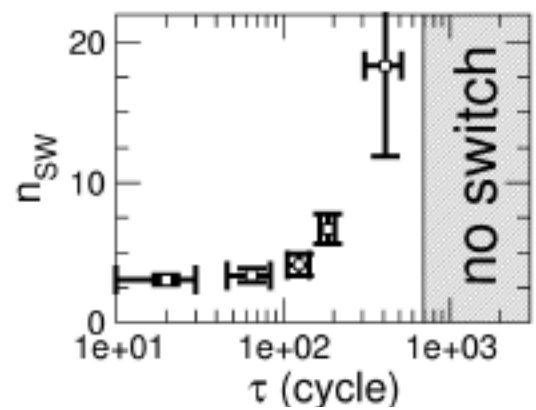


Transition rates imply the existence of a relaxation time scale t_{relax}

Short pulses (< t_{delay}) are sent at intervals $\tau < t_{relax}$

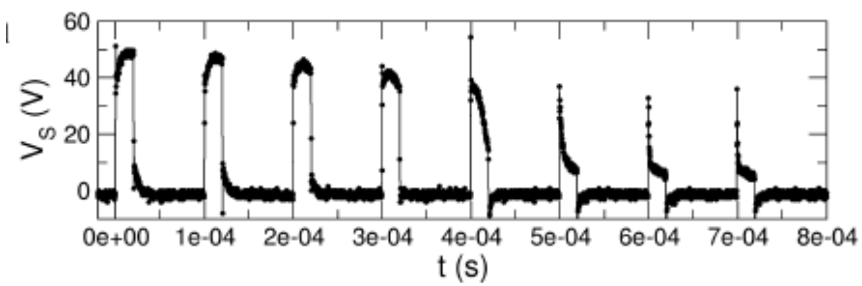
Model prediction

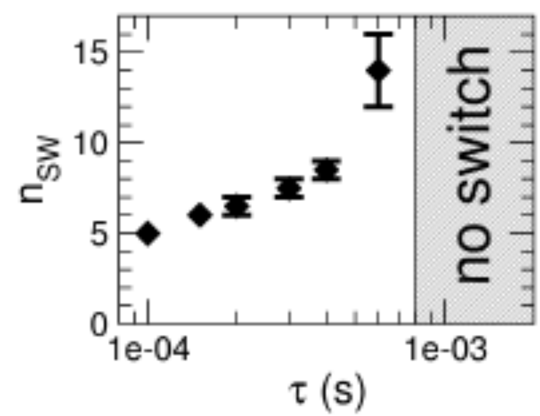




Transition after 5 pulses

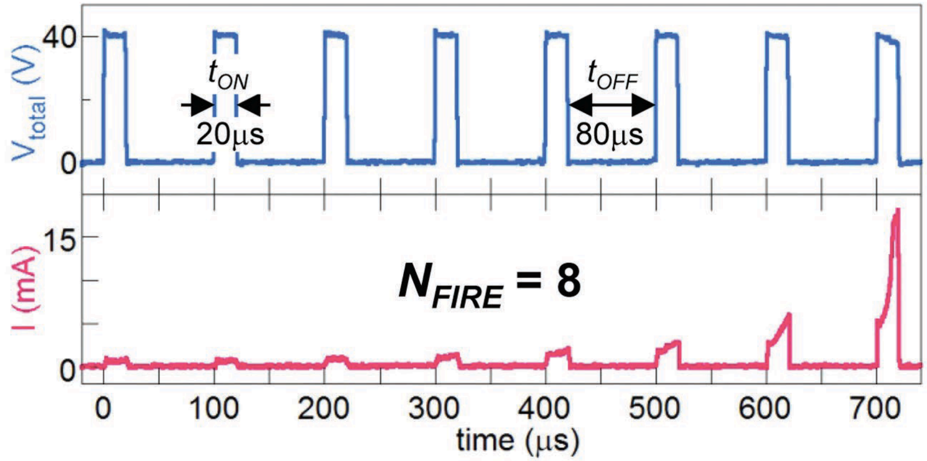
Experiment

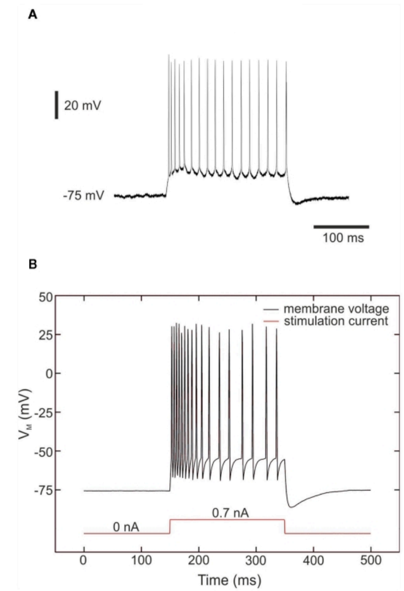




A Leaky-Integrate-and-Fire Neuron Analogue realized with a Mott insulator

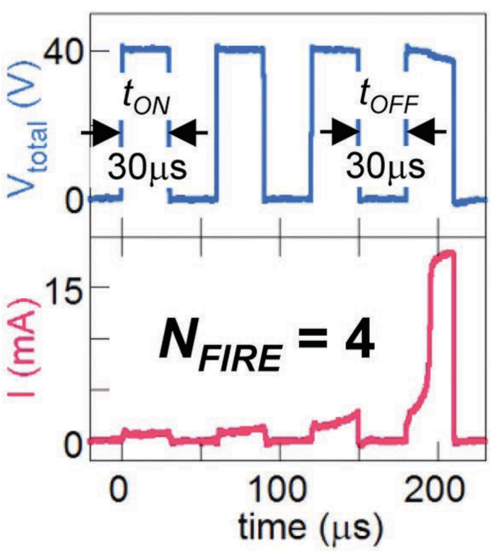
P. Stoliar, MR, et al Adv Funct Mat (2017)



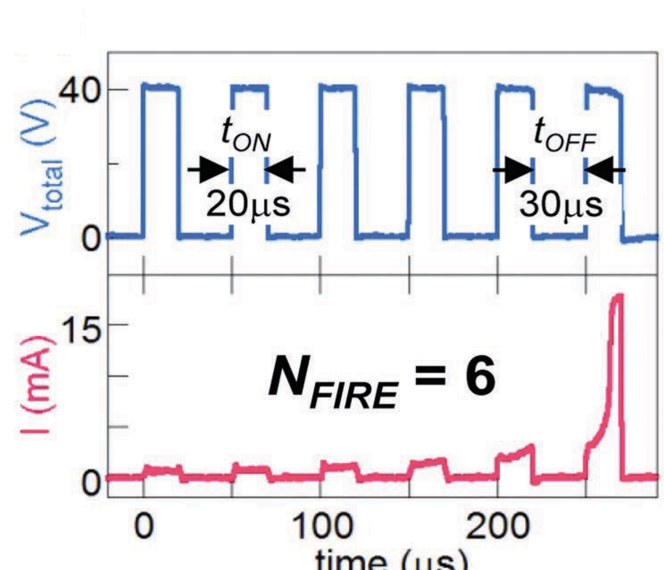


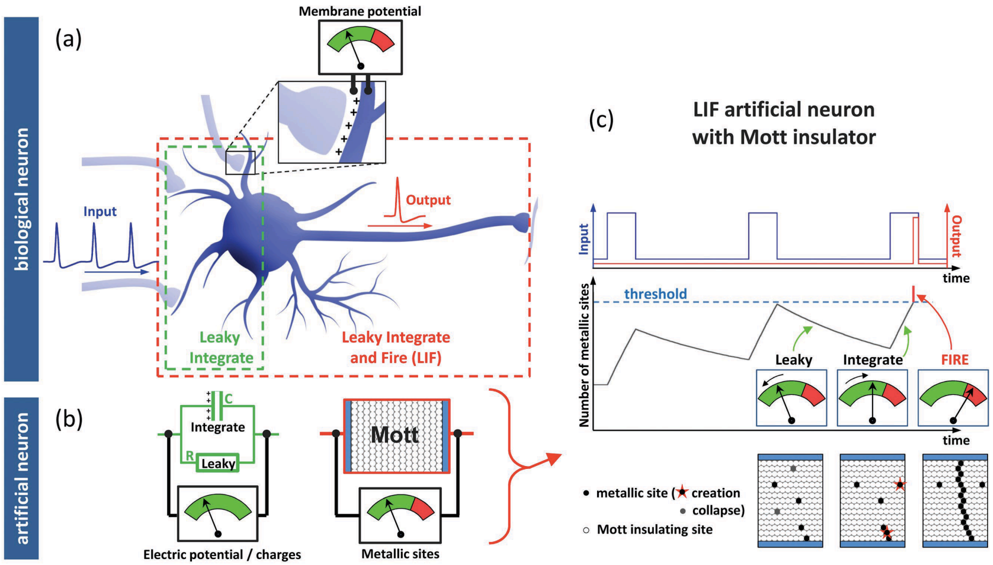
French patent n1453834.
"Neurone artificiel mono-composant à base d'isolants de Mott,
réseau de neurones artificiels et procédé de fabrication correspondants".

higher frequency

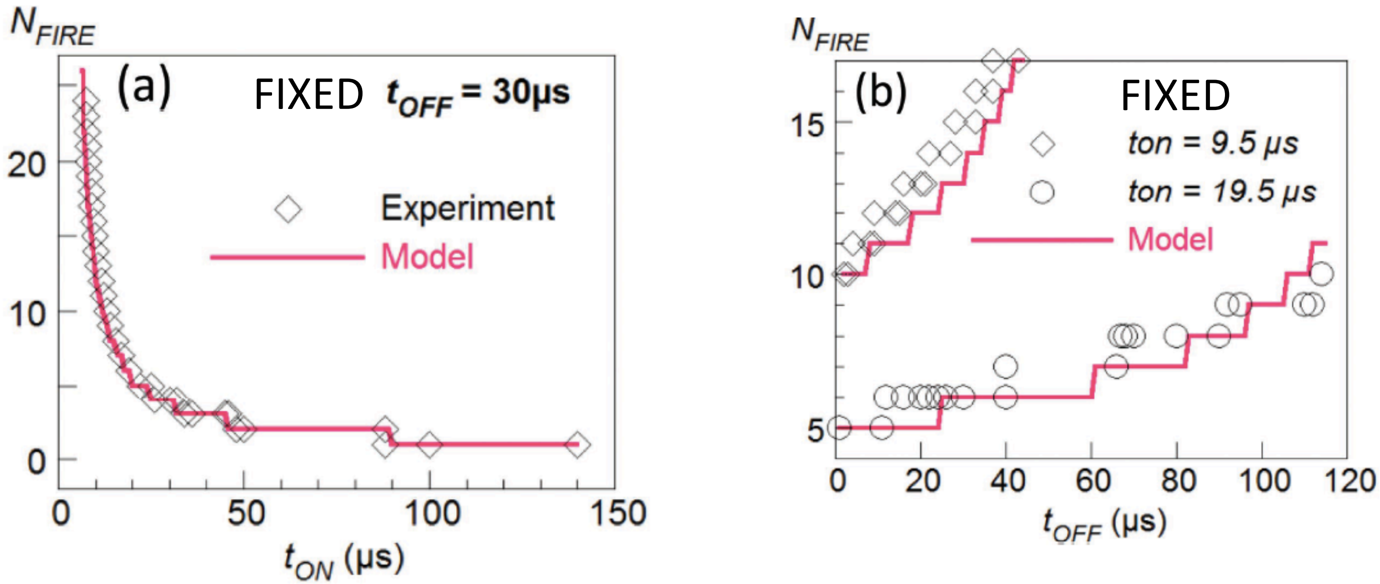


higher strength





	LIF model	Mott LIF neuron
Integrated variable	Membrane potential v	Fraction metallic regions n_{CM}
Model	$\frac{\partial}{\partial t}v = -v\frac{1}{RC} + \frac{w}{C}s(t)$	$\frac{\partial}{\partial t}n_{CM} = -n_{CM}P_{CM\rightarrow MI} + Ap(t)$
Input variable	Dirac delta function	Voltage pulse
Output variable	Not defined	Current pulse
Leaking time constant	RC	$1/P_{CM\rightarrow MI}$
Synaptic input	$s = \sum_i \delta(t - t_i)$	$p = \sum_i [H(t - t_i) - H(t - t_i - t_{ON})]$
Spike contribution	w/C	At_{ON}
Number of pulses for FIRE	$N_{FIRE} = \text{ceiling}\left(1 - \frac{\ln\left[e^{t_{OFF}/\tau} - \frac{t_{FIRE}}{t_{ON}}\left(e^{t_{OFF}/\tau} - 1\right)\right]}{t_{OFF}/\tau}\right)$	



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Summary

- We now have artificial synapses and neurons made of simple 2 terminal oxide devices whose physics is based on the physical phenomenon of resistive switching

- Theoretical modeling may provide useful guidance for experiments

- The way is open for neuromorphic aplications

Reviews:

<u>Non-volatile Resistive Switching:</u>

M. Rozenberg, Scholarpedia 6(4):11414 (2011) (short introductory)
H-S Philip Wong et al., Proceedings of IEEE v100 p1951 (2013)
D. Ielmini et al. Phase transitions v84 p570 (2011)
J.J. Yang et al, Nature Nanotechnology, v8 p13 (2013)

<u>Volatile Resistive Switching in Mott insulators:</u>

E. Janod et al Adv Func Mat Adv. Func. Mat. (2016)

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L. Cario
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