



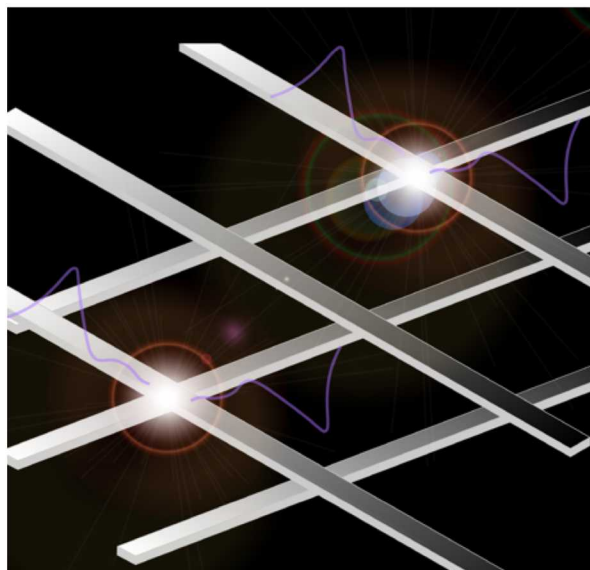
Sandia
National
Laboratories

SAND2019-9702PE

Electrochemical materials for analog memory and neuromorphic computing

PRESENTED BY

Yiyang Li



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Computers vs the Human Brain



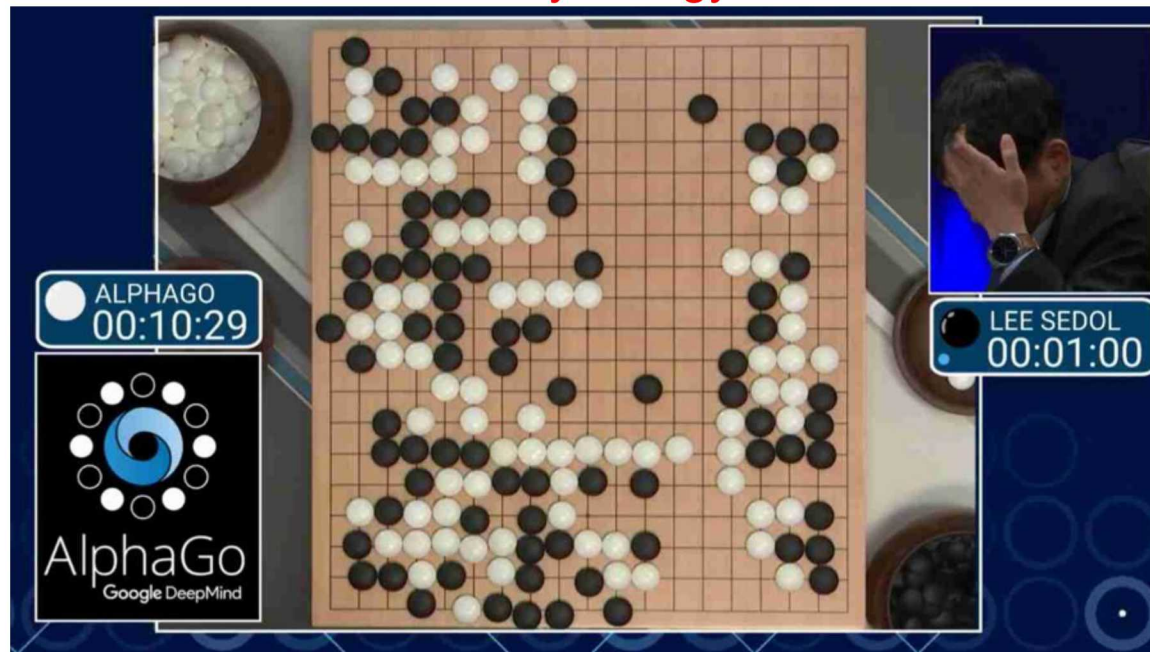
10^{12} FLOPS



~1 FLOPS

Machine learning can beat humans in certain tasks,
but are extremely energy intensive

10^6 Watt



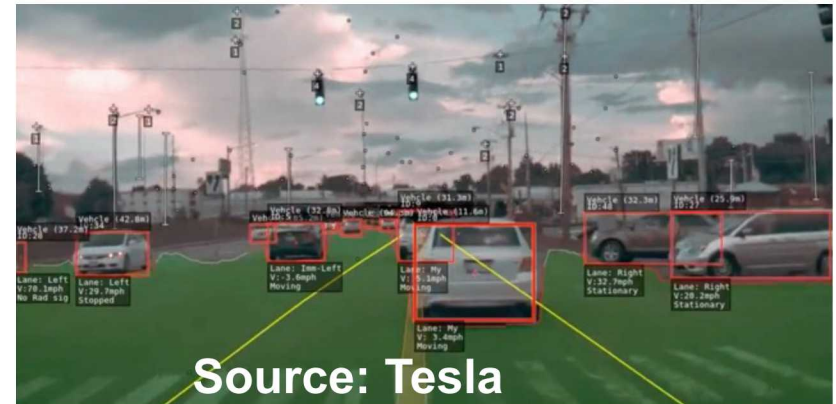
20 Watt



Example: autonomous driving



Chevy Bolt

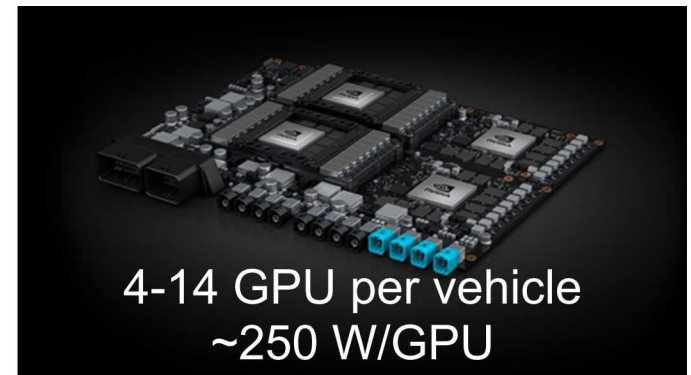


~100 GB/sec

Human brain: 10 Watt

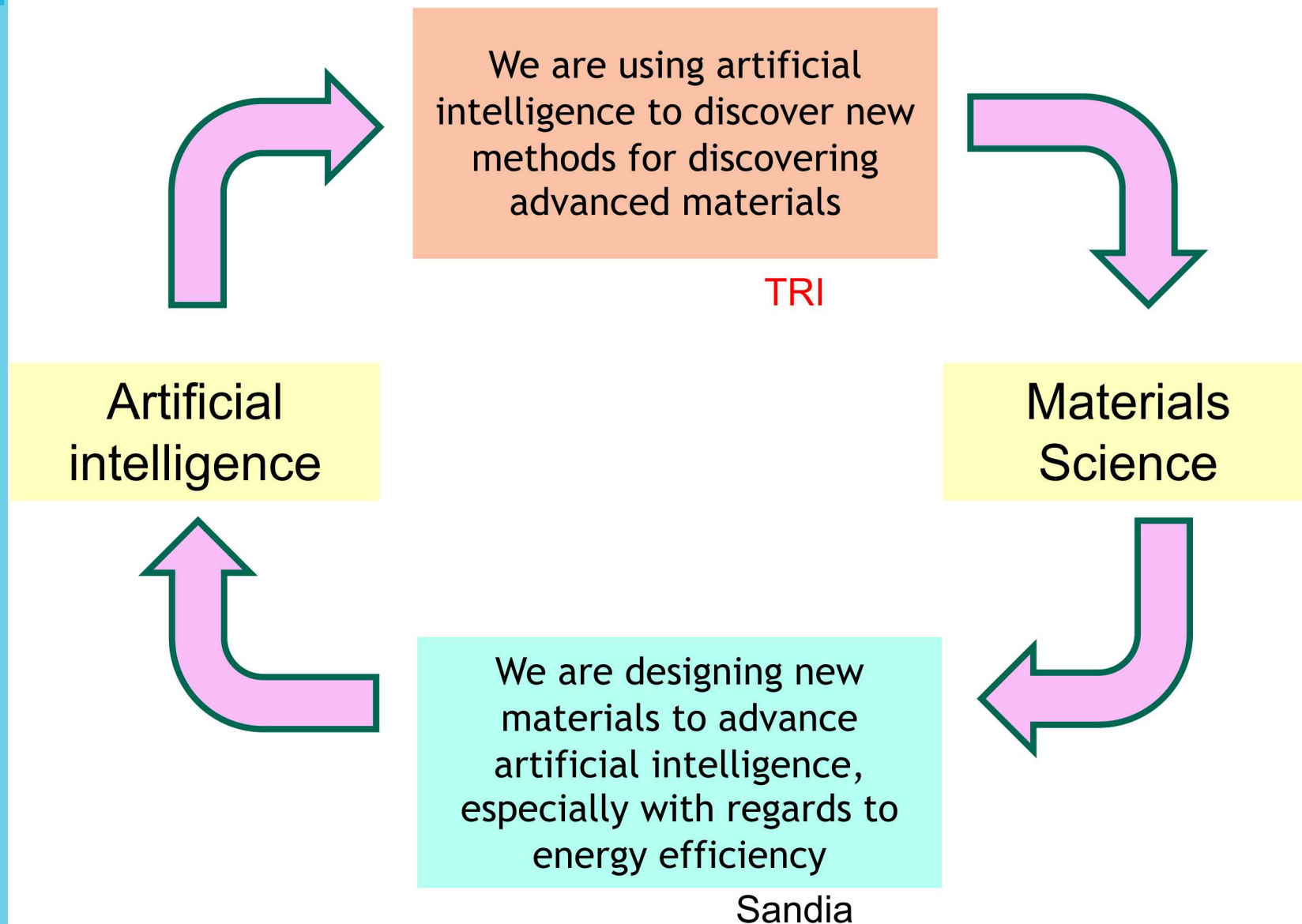


Computing: 1-4 kW



4-14 GPU per vehicle
~250 W/GPU

Nvidia





Artificial neural networks

Which one of these images is a cat?



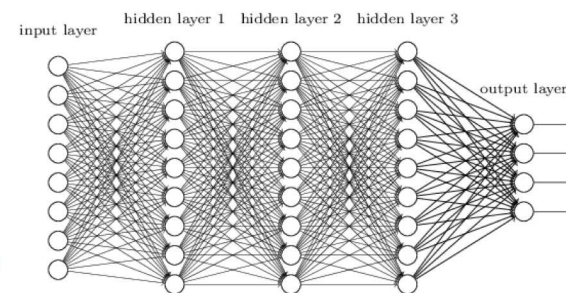
Image recognition
Autonomous driving
Natural language processing

Artificial neural networks: use training examples and error backpropagation to find the matrix weights that correctly maps the input $\mathbf{x}$ onto the desired output $\mathbf{y}$

$$\begin{bmatrix} y_1 \\ \vdots \\ y_m \end{bmatrix} = \begin{bmatrix} w_{1,1} & \cdots & w_{1,n} \\ \vdots & \ddots & \vdots \\ w_{m,1} & \cdots & w_{m,n} \end{bmatrix} \begin{bmatrix} x_1 \\ \vdots \\ x_n \end{bmatrix}$$

Inference and training are very energy expensive

$n, m > 1000$

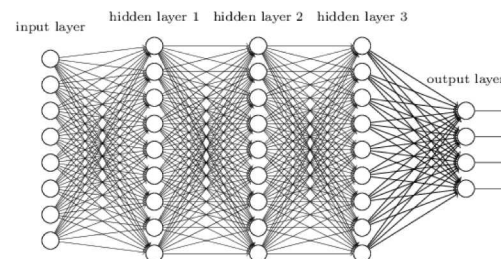




Digital and analog implementations of neural networks

$$\begin{bmatrix} y_1 \\ \vdots \\ y_m \end{bmatrix} = \begin{bmatrix} w_{1,1} & \cdots & w_{1,n} \\ \vdots & \ddots & \vdots \\ w_{m,1} & \cdots & w_{m,n} \end{bmatrix} \begin{bmatrix} x_1 \\ \vdots \\ x_n \end{bmatrix}$$

$n, m > 1000$

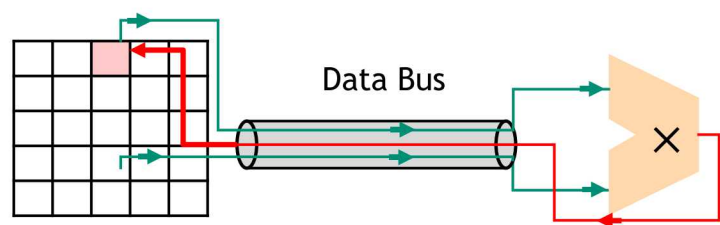


Von Neumann Digital

Separate logic and memory structures

SRAM to store the weights

Arithmetic logic unit for multiplication



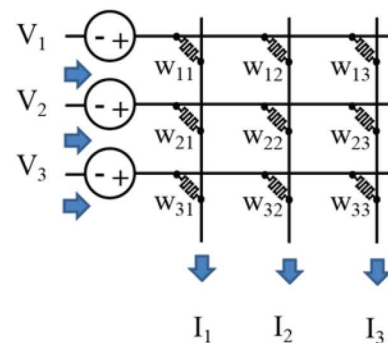
Uses established CMOS technology
Data bus results in latency and power

In-memory Parallel Analog

Use non-volatile memory

Crossbar for matrix multiplication

Conductance of each element can be changed in a predictable manner



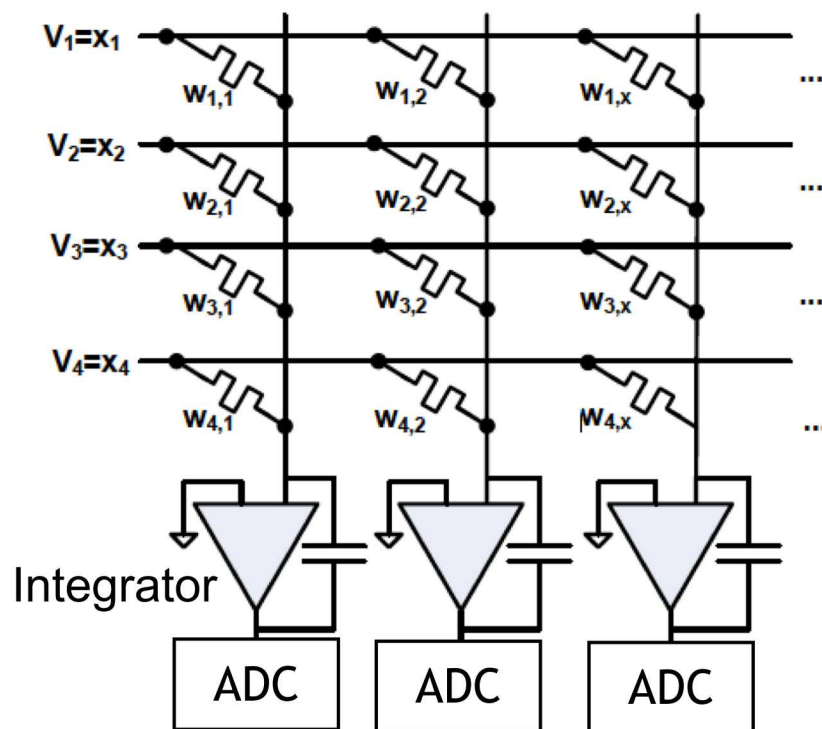
$$I_1 = V_1 W_{11} + V_2 W_{21} + V_3 W_{31}$$

Simultaneous logic and memory
3 orders of magnitude less power



Energy consumption: analog vs digital

Analog core: doing $O(n^2)$ operations using $O(n)$ inputs and outputs



Energy per operation per array,
1024X1024 synapses
1 Million multiply-and-accumulate

	Analog	Digital
Vector matrix multiply	13 nJ	2600 nJ
Outer product update	2 nJ	3600 nJ

Analog energy breakdown:

Read: 0.4 nJ
Write: 1.66 nJ
Integrator: 2.8 nJ
ADC: 9.4 nJ

M. Marinella, *IEEE Circuits and Systems*, 8, 86-101, 2018

Analog: ~200 Tera-OPS/watt

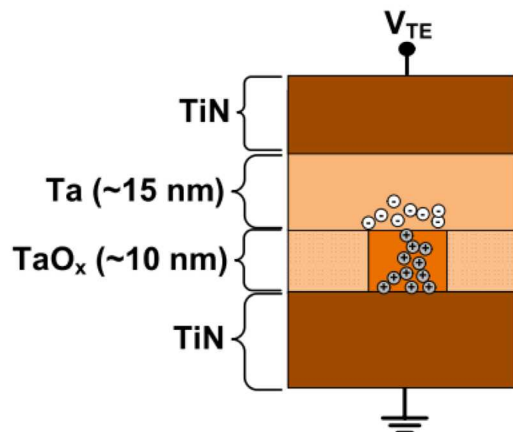
Digital: 1-5 Tera-OPS/watt

Challenge: analog non-volatile memory that meets these requirements do not exist

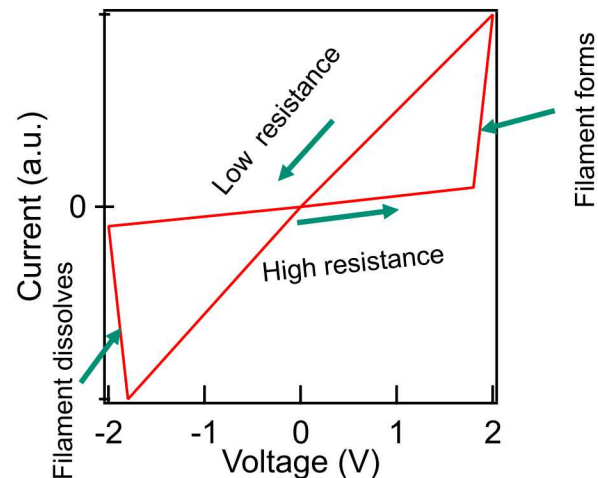


Two-terminal resistive memory

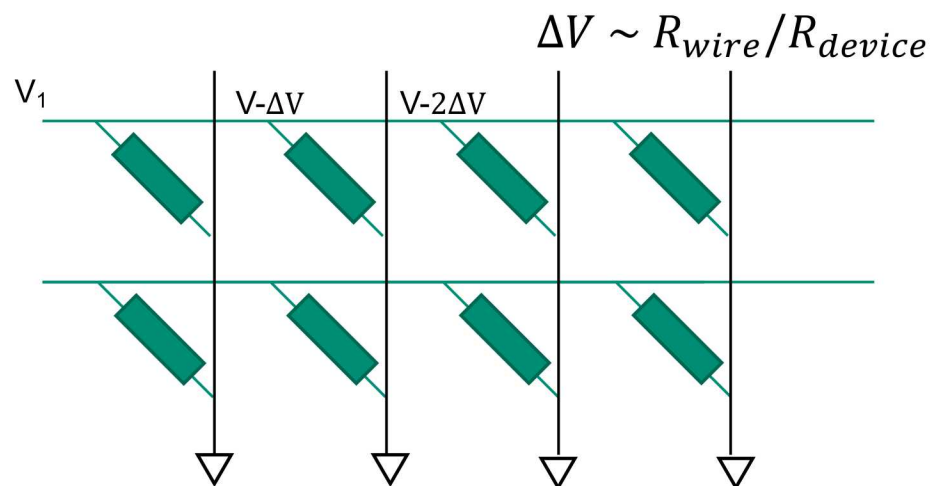
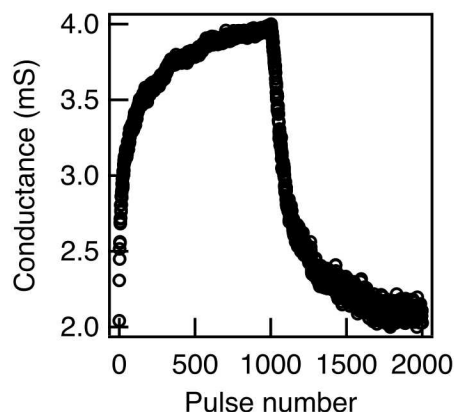
TaO_x resistive memory cell



Resistive memory switch between high resistance and low resistance states



Analog resistive memory is nonlinear and unpredictable

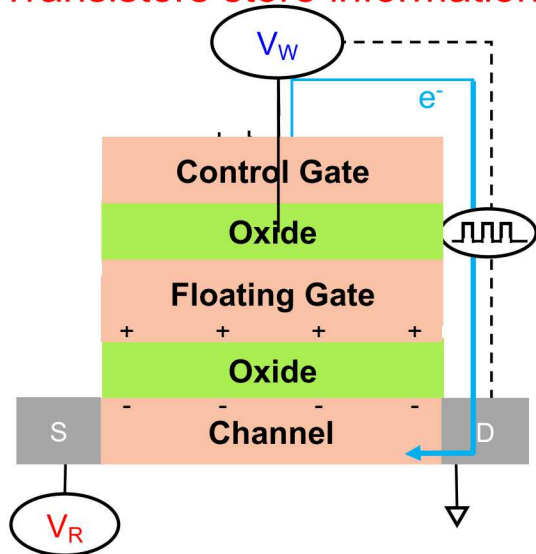


Target: $R_{\text{device}} = 10^7\text{-}10^9 \Omega$



Three-terminal memory

Transistors store information by moving charge between the gate and the channel



$$\tau_{\text{retention}} = C_{\text{gate}} R_{\text{OFF}}$$

$$C_{\text{gate}} = \epsilon_0 \epsilon_r \frac{A}{t_{\text{ox}}} \approx 10^{-6} \text{ F cm}^{-2} = 10^{-16} \text{ F (A = 100 nm}^2\text{)}$$

Option 1: transistor switch: $R_{\text{OFF}} \sim 10^{15} \Omega$

$\tau_{\text{retention}} < 1 \text{ s}$ (Resistive Processing Unit, dynamic RAM)

S. Ambrogio, G. Burr, et al. *Nature*, **558**, 60 (2018)

Option 2: floating gate oxide switch, $R_{\text{OFF}} \sim \infty$

$\tau_{\text{retention}} > 10 \text{ yr}$ (Flash memory)

10V, high power, limited endurance (10^5)

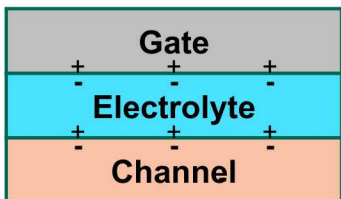
Improving retention by increasing C_{gate} ? $C = \frac{\Delta Q}{\Delta V}$

Electrochemical double-layer transistors

Electrons stored at the **interface**

$$C_{\text{gate}} \sim 10^{-4} \text{ F cm}^{-2}$$

$$t_{\text{ox}} < 1 \text{ nm}$$



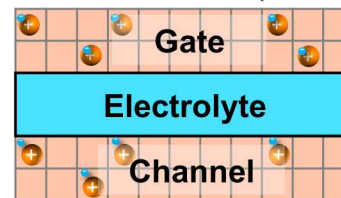
P. Gkoupidenis, G. Malliaras, *Adv Mater*, **27**, 7176 (2015)

Redox transistors

Electrons (and ions) are stored in the **bulk** of the material (pseudo-capacitor)

Electron/ion neutral ambipolar pair enables ΔQ without electrostatic charging

$$C_{\text{gate}} \sim 10^{-1} \text{ F cm}^{-2} \text{ (100-nm film)}$$

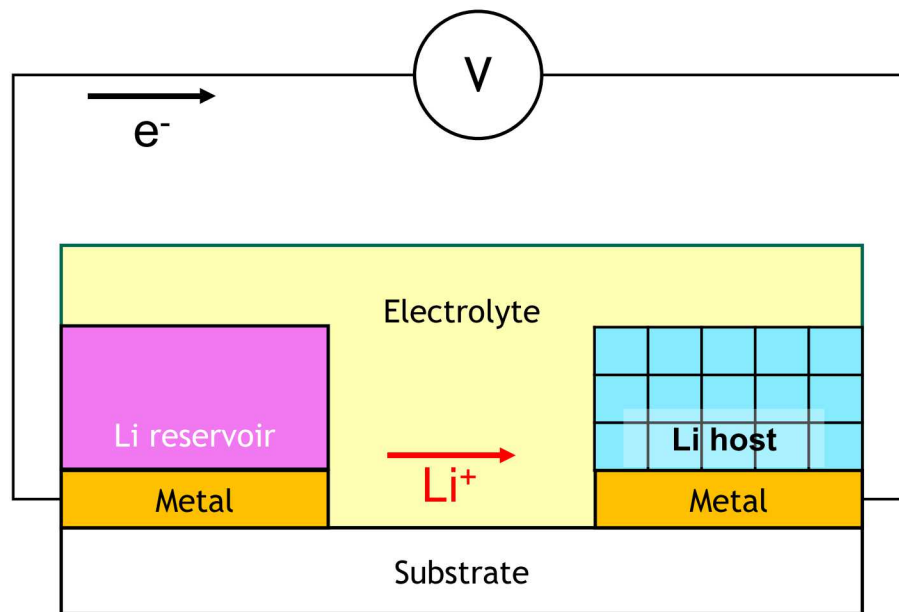


E. Fuller, *Adv Mater*, **29**, 1604310 (2017)

Y. van de Burgt, *Nat. Mater.*, **16**, 414 (2017)



Redox transistors based on electrochemical ion insertion



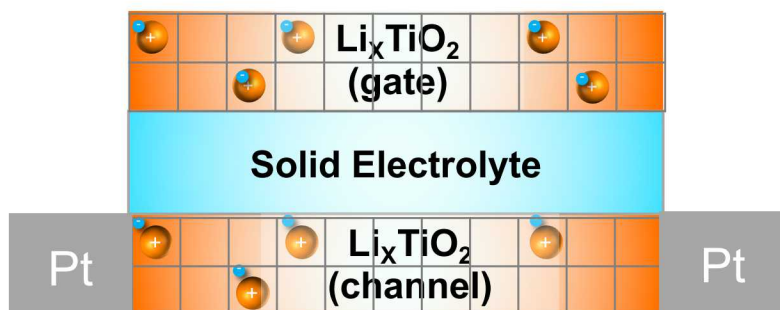
Dynamic control of the lithium doping level using current and voltage

In batteries, intercalation provides a high density of energy storage;
In redox transistors, intercalation provides a high density of information storage

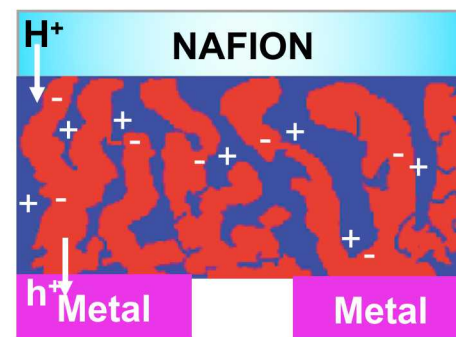


Outline

Part 1: Inorganic redox transistor Lithium insertion into TiO_2

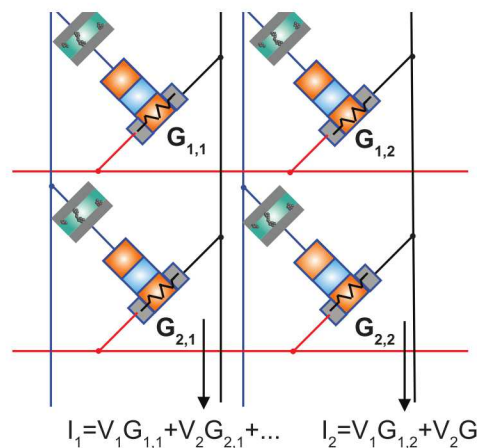


Part 2: Organic redox transistor Proton insertion into PEDOT:PSS



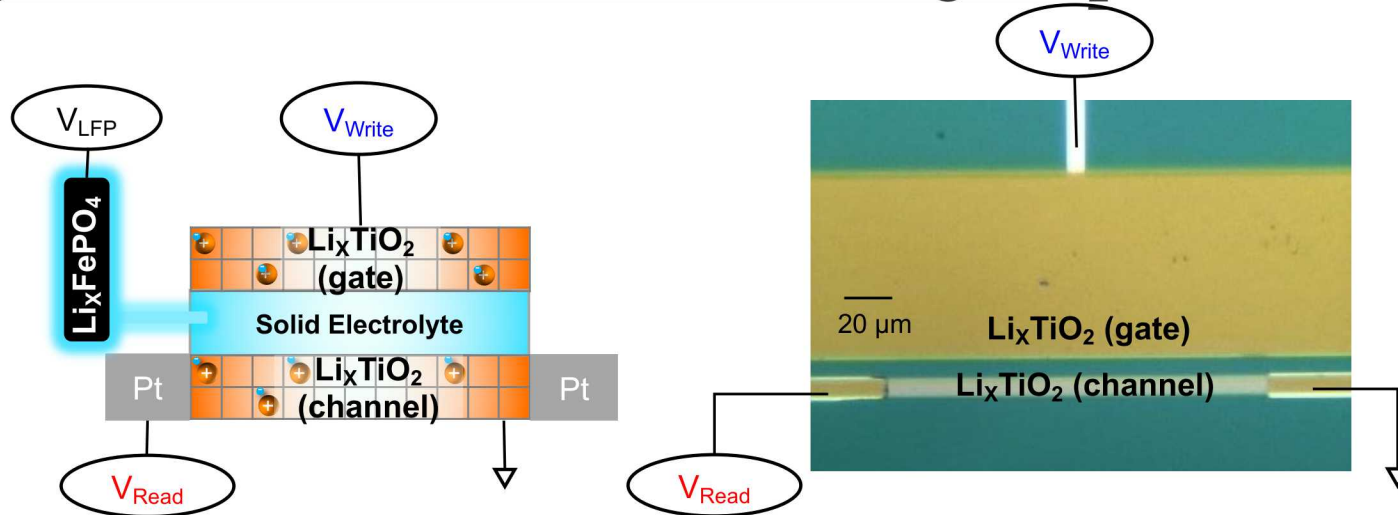
H^+ : proton; h^+ : hole
PEDOT:PSS

Part 3: Crossbar Array integration





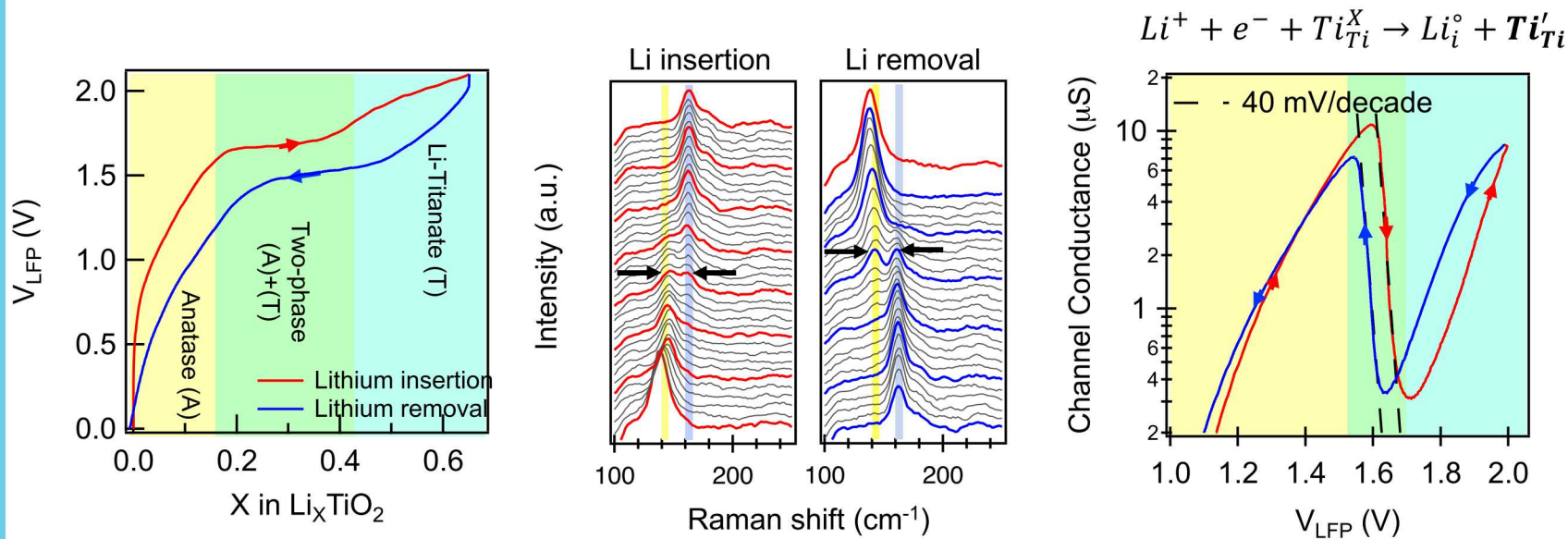
Symmetric redox transistor using TiO_2



A. Talin

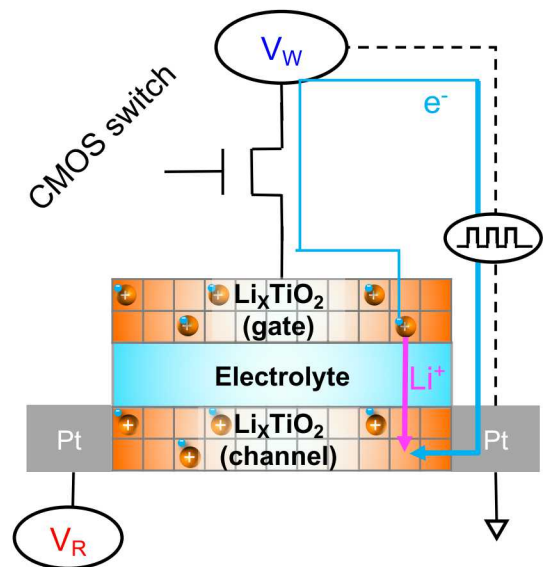


E. Fuller

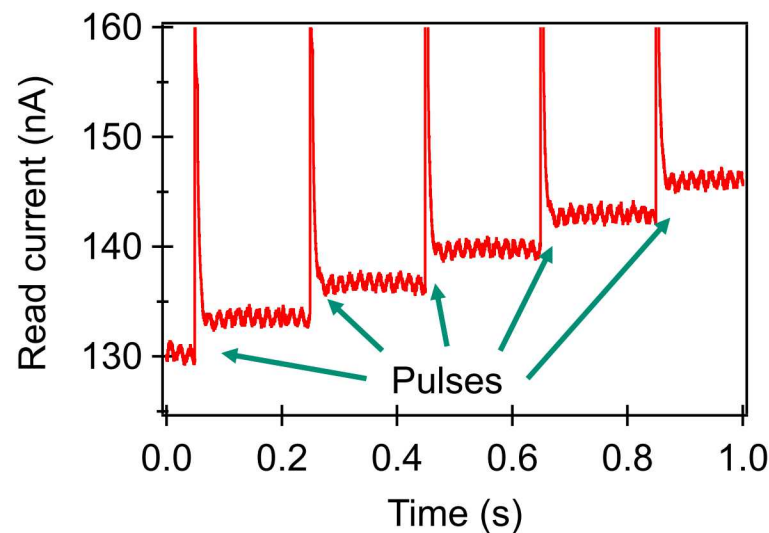
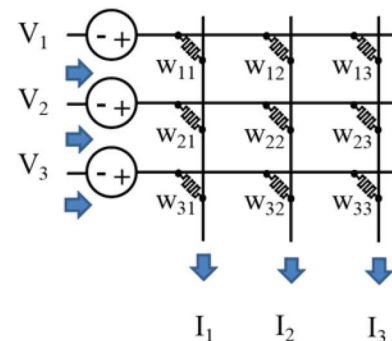
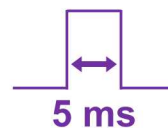




Redox transistor using Li_xTiO_2 (anatase)



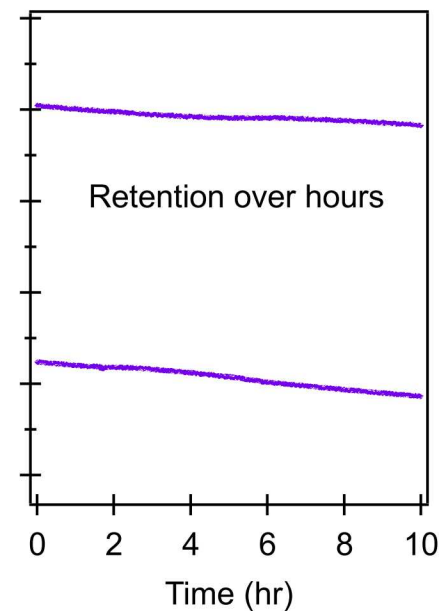
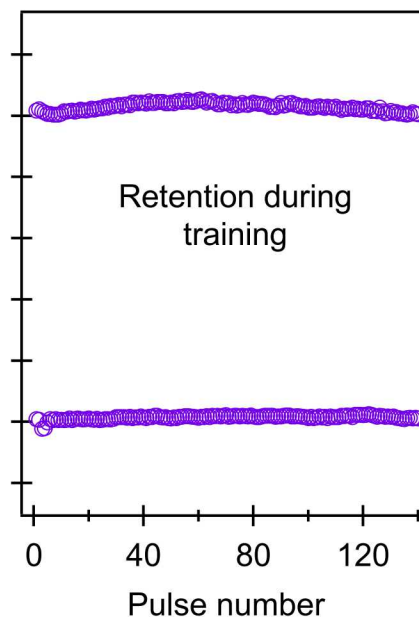
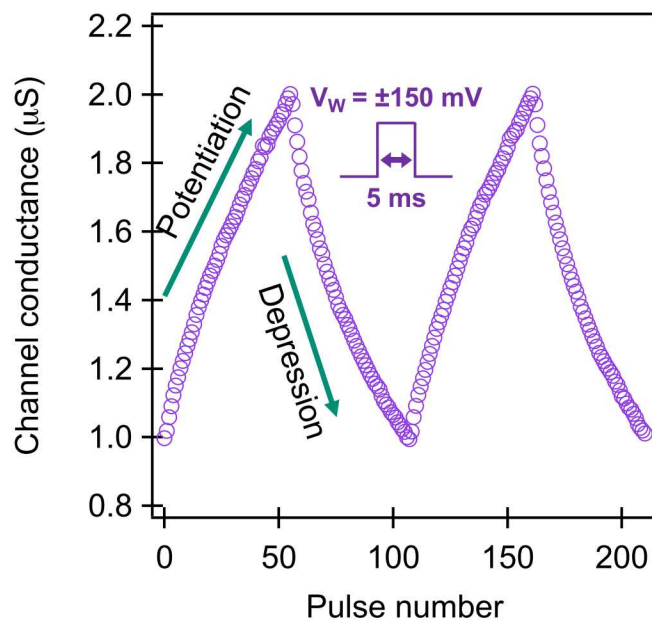
$$V_W = \pm 150 \text{ mV}$$



- Linear and symmetric programming
 - 150-mV write pulses
 - Analog, non-volatile states



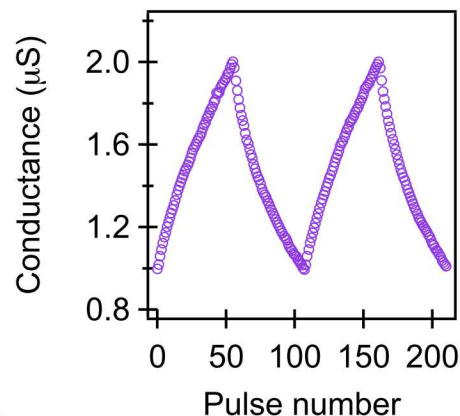
Redox transistor using Li_xTiO_2 (anatase)



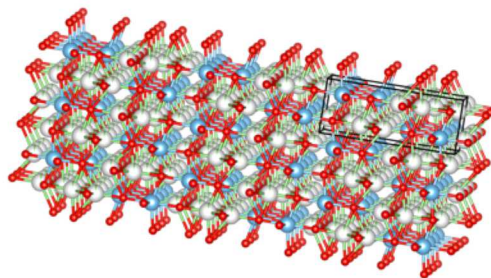


Linear programming and high accuracy

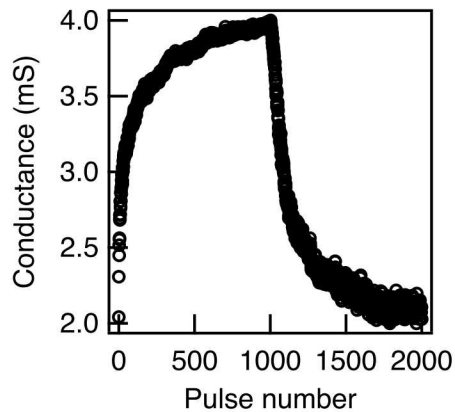
Li_xTiO_2 transistor
Voltage: ± 0.15 V



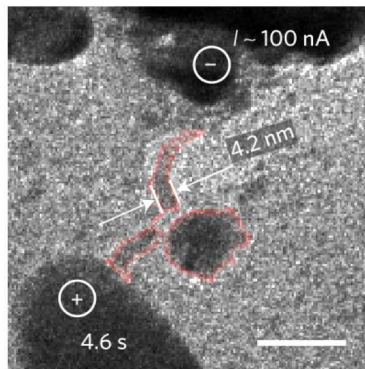
Redox transistors store information continuously as dopants in a crystal



TaO_x Memristor
Voltage: ± 1 V



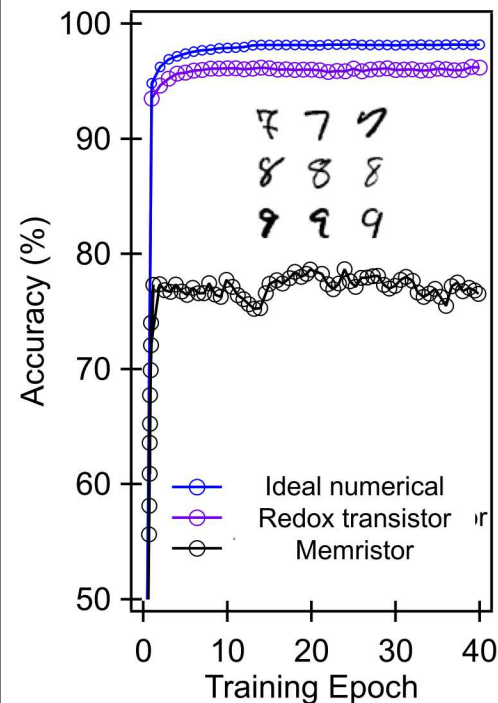
Memristors store information at filaments



R. Jacobs-Gedrim et al. *ICRC*, 2017

Wang et al. *Nat Mater.* 2017

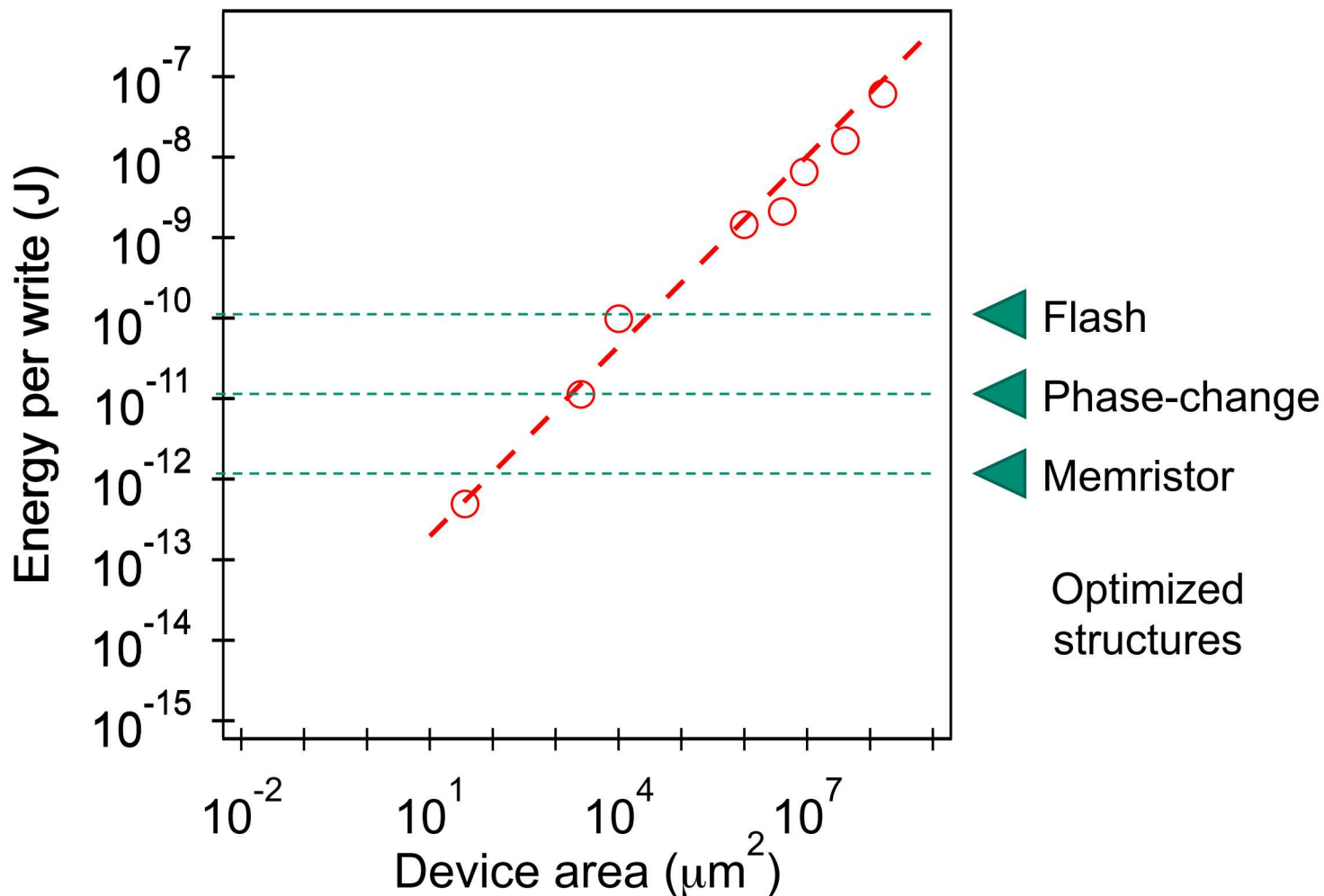
Linearity is essential to train an accurate neural network



Crossbar Simulator



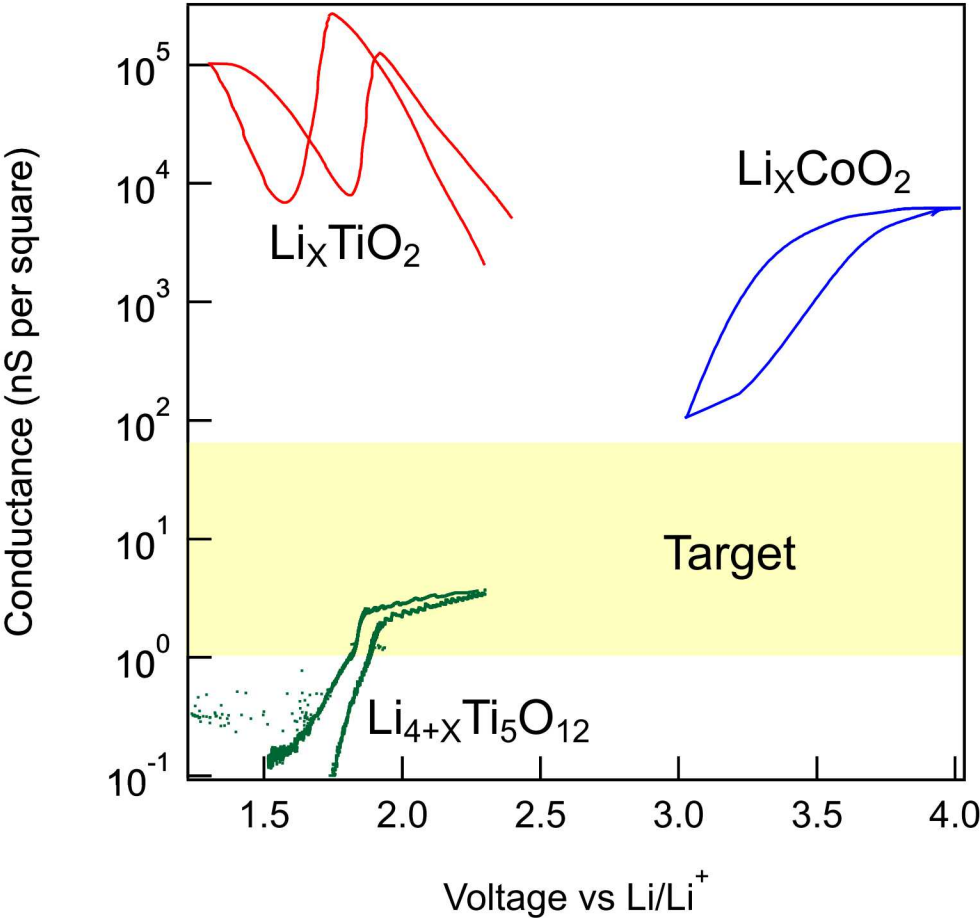
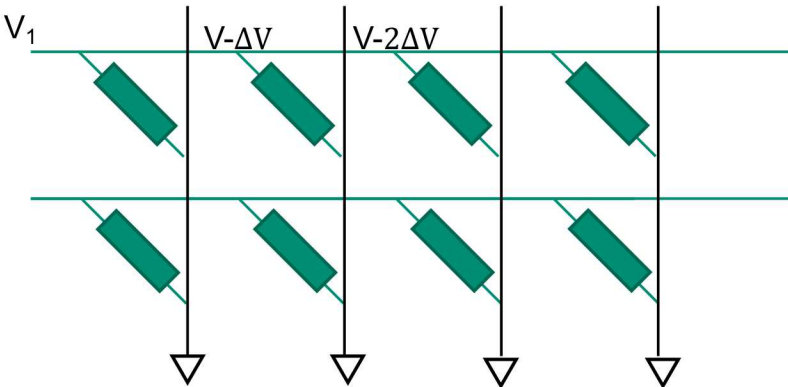
Low write energies



Y. van de Burgt, *Nat Mater*, 16, 414(2017)
M. Sharbati *Adv. Mater.* 30, 1802353 (2018)
J. Yang et al. *Nat. Nano.* 8, 13(2012)



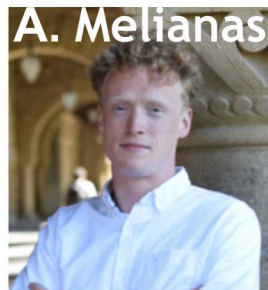
High resistance devices



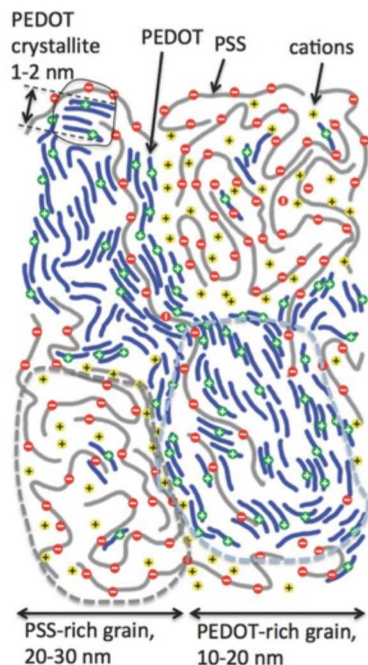
} Memristors



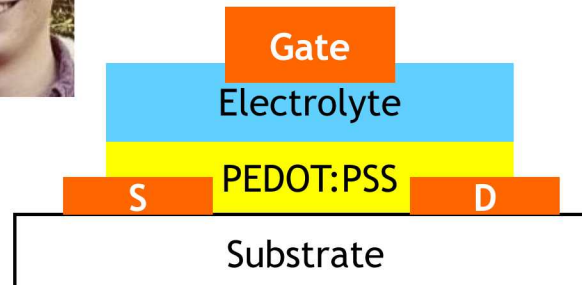
Part 2: Organic redox transistor based on PEDOT:PSS



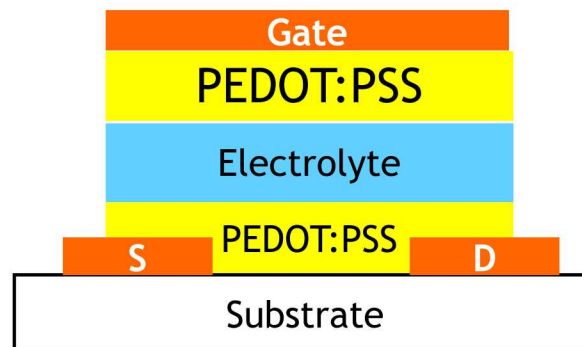
PEDOT (hole conductor)
PSS (proton conductor)



A. Volkov, et al. *Adv. Funct. Mater.* 27, 1700329 (2017)



Metal/electrolyte double-layer capacitance $\sim 20 \mu\text{F}/\text{cm}^2$

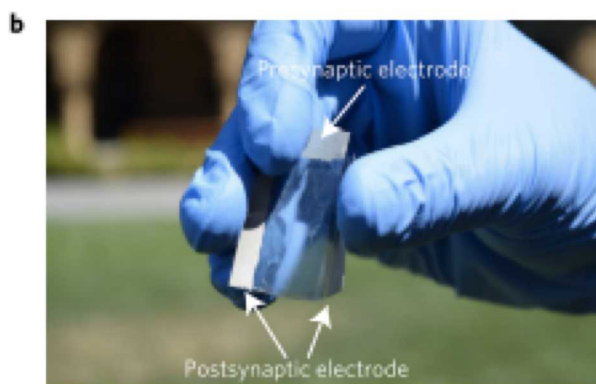
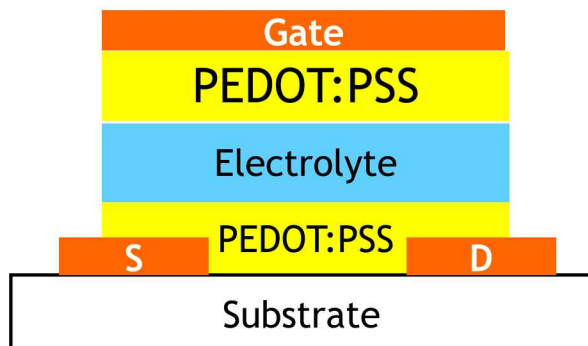


Volumetric charge density: $40 \text{ F}/\text{cm}^3$
 $\sim 400 \mu\text{F}/\text{cm}^2$ (100-nm film)

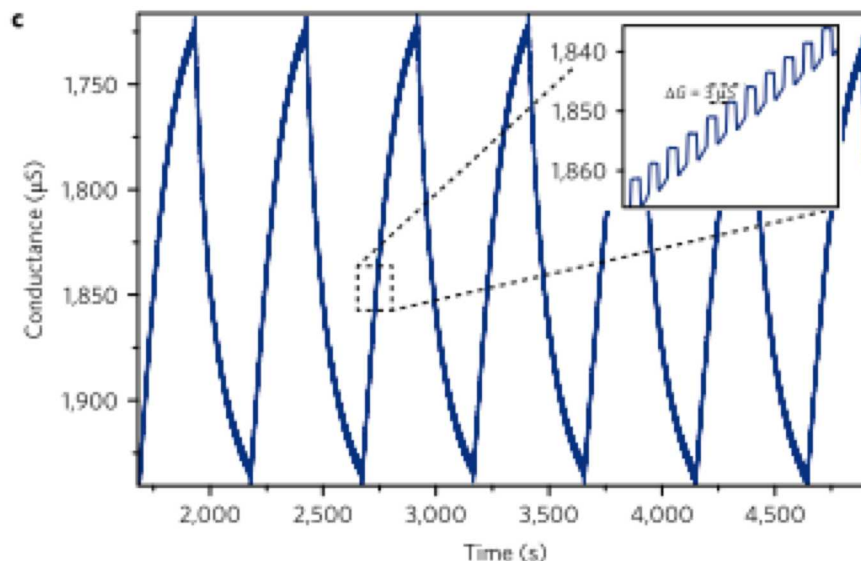


PEDOT:PSS Redox Transistor

Flexible device

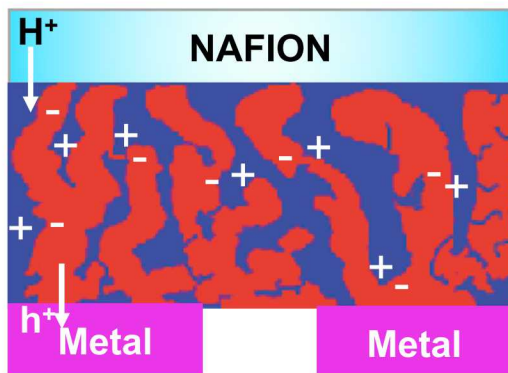


Linear weight updates





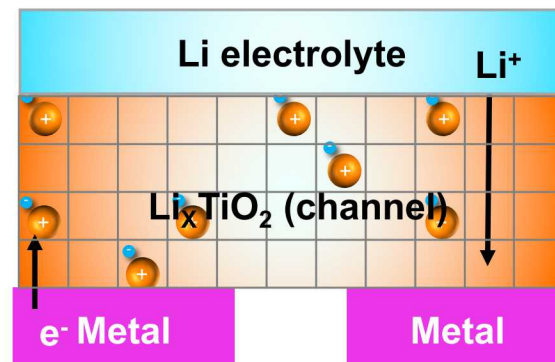
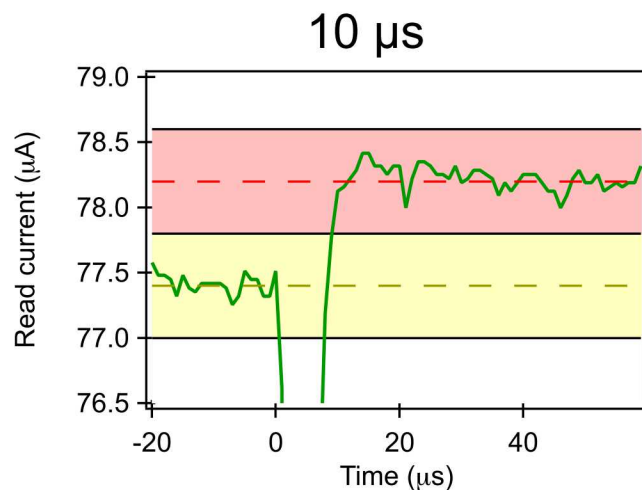
Super-capacitative vs intercalation redox transistor



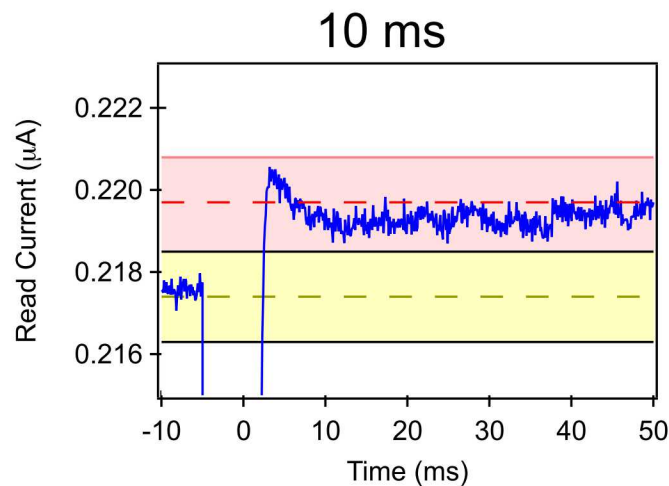
H^+ : proton; h^+ : hole

PEDOT:PSS

No charge transfer
Fast single-specie diffusion
Low capacitance: 40 F cm^{-3}



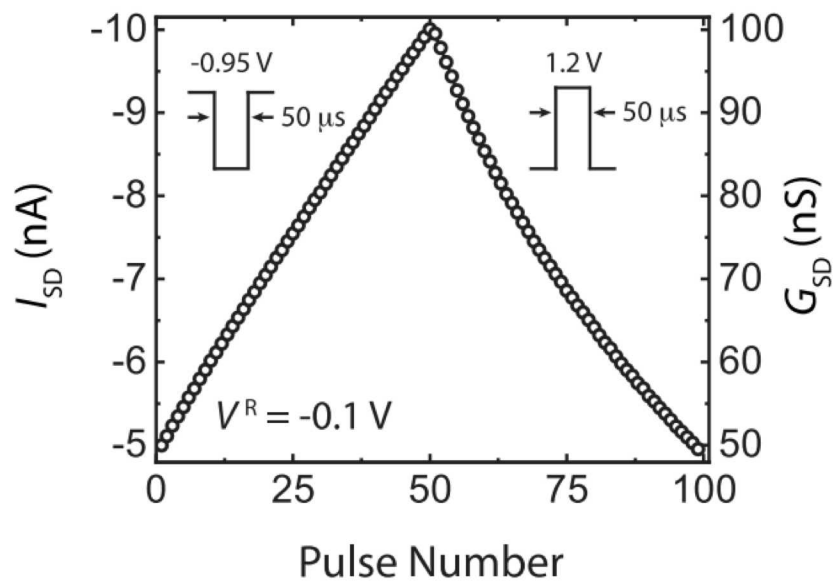
Interfacial charge transfer
Slow chemical diffusion
High capacitance: 5000 F cm^{-3}





High resistance devices

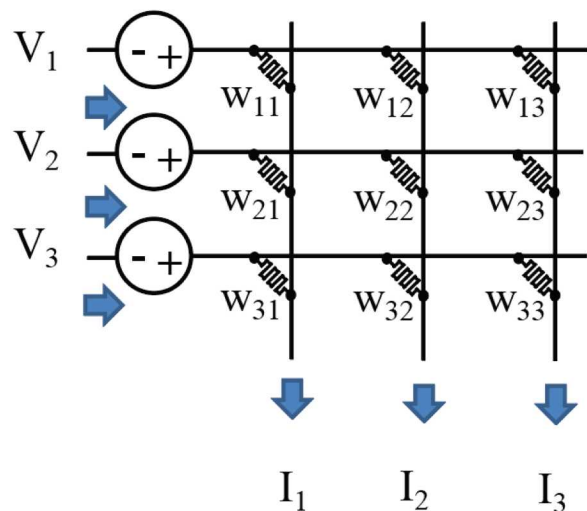
10^7 Ohm achieved by doping with filler PEI



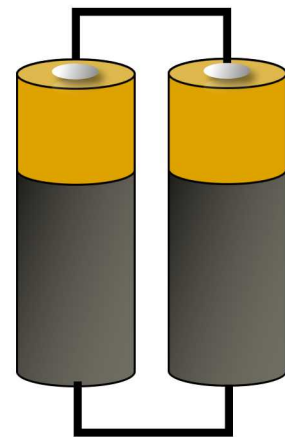
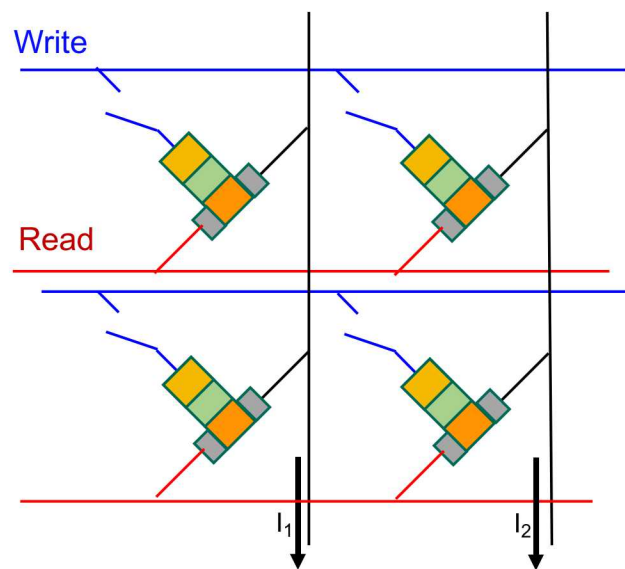


Part 3: Crossbar array integration

Matrix multiplication crossbar



Gates and channels are shorted to each other



Challenge: find a highly nonlinear switch with a high ON/OFF ratio at low voltages

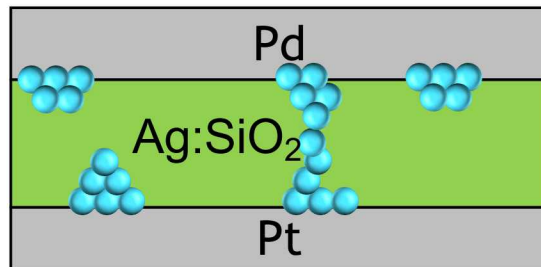
$$\tau_{retention} \sim R_{switch} * C_{device}$$



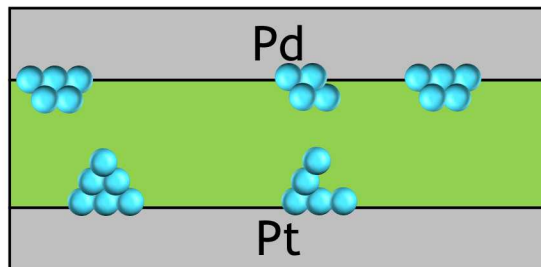
Diffusive memristor

Two-terminal switch

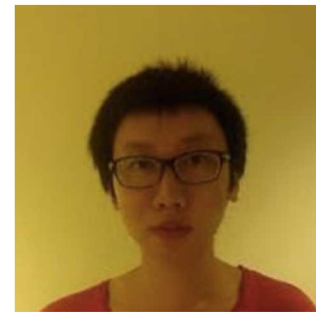
ON



OFF



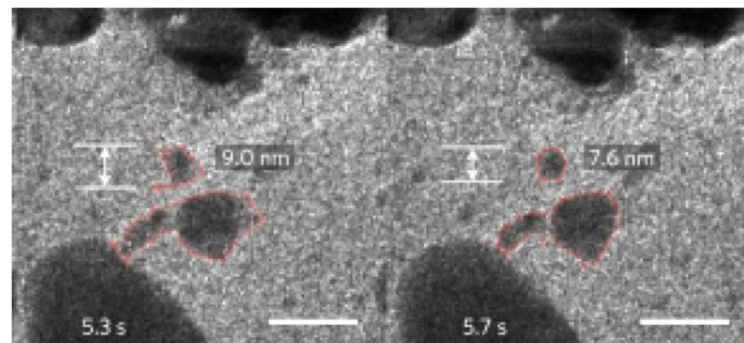
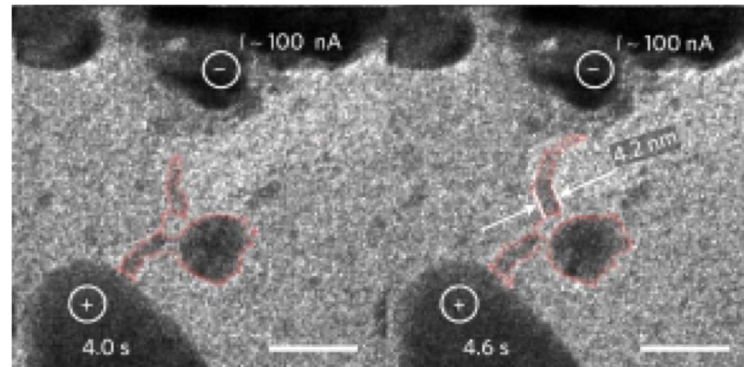
J. Yang



Z. Wang



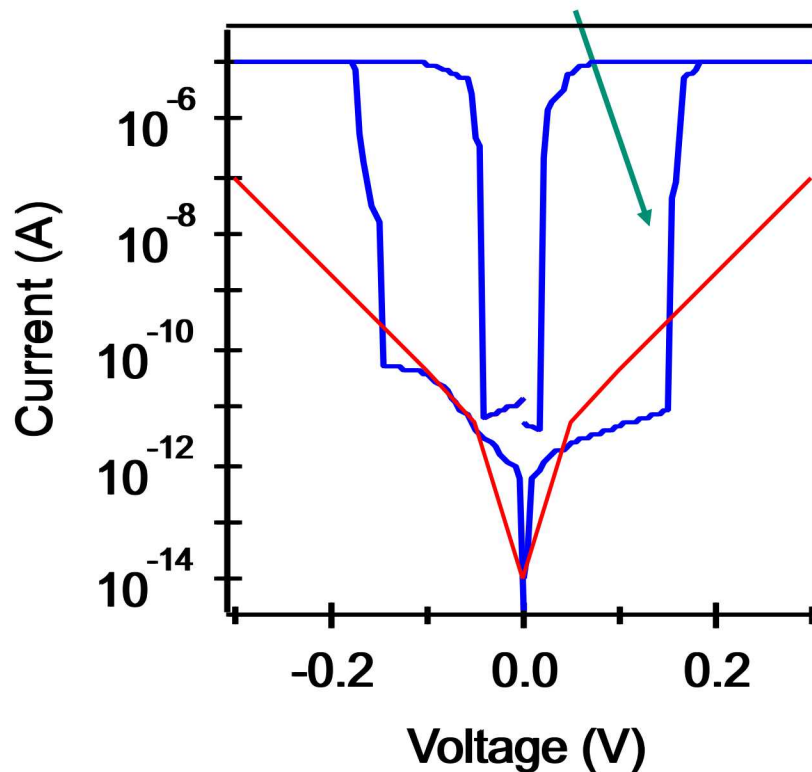
S. Asapu





High nonlinearity at low voltages

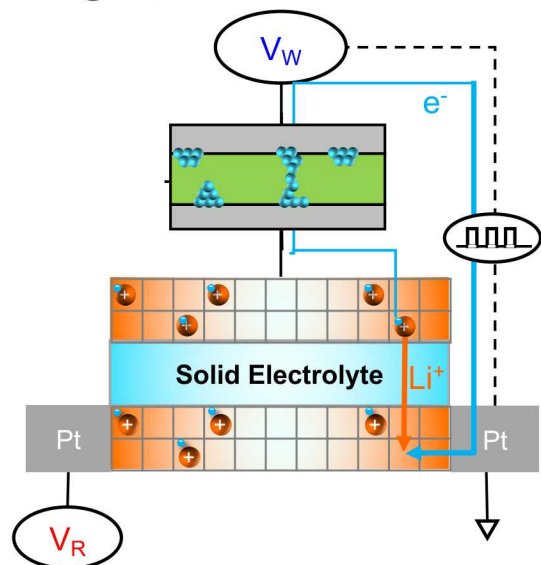
Diffusive Memristor: ~ 1 mV/decade,
 10^7 ON/OFF



Ideal diode: 60 mV/decade
Need much higher voltages
for same ON/OFF ratio



Low-voltage, Si-free electrochemical memory



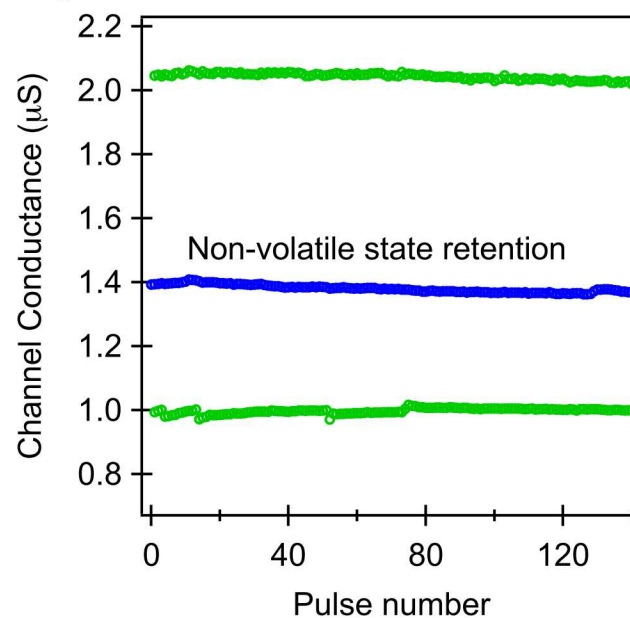
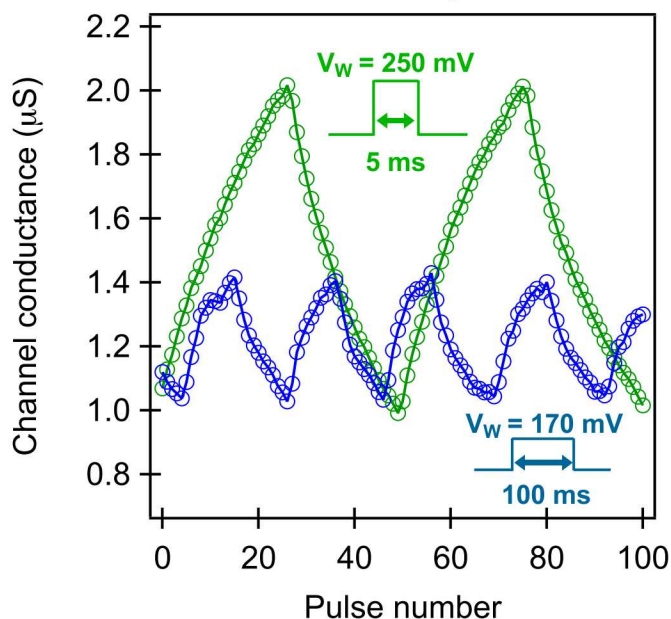
Diffusive memristor (Ag in SiO_x): high ON/OFF ratio

Redox transistor: high charge density via bulk storage

Both: Low switching voltages

Wang et al. *Nat. Mater.* 2017

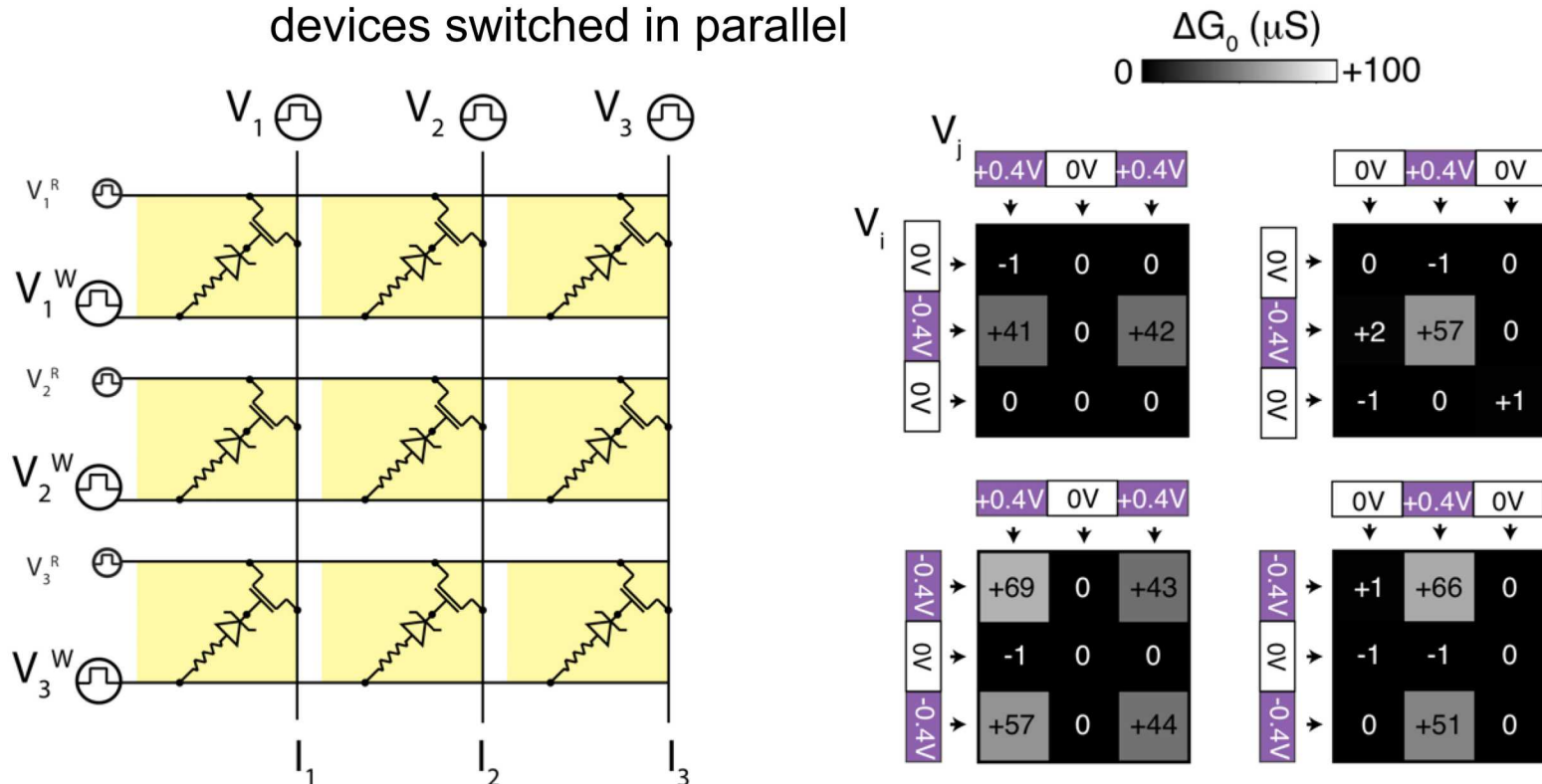
Non-volatile memory that switches at just 6 times the thermal voltage





Parallel weight updates in crossbar arrays

Speed of a network $\sim$ speed of a device * # of devices switched in parallel



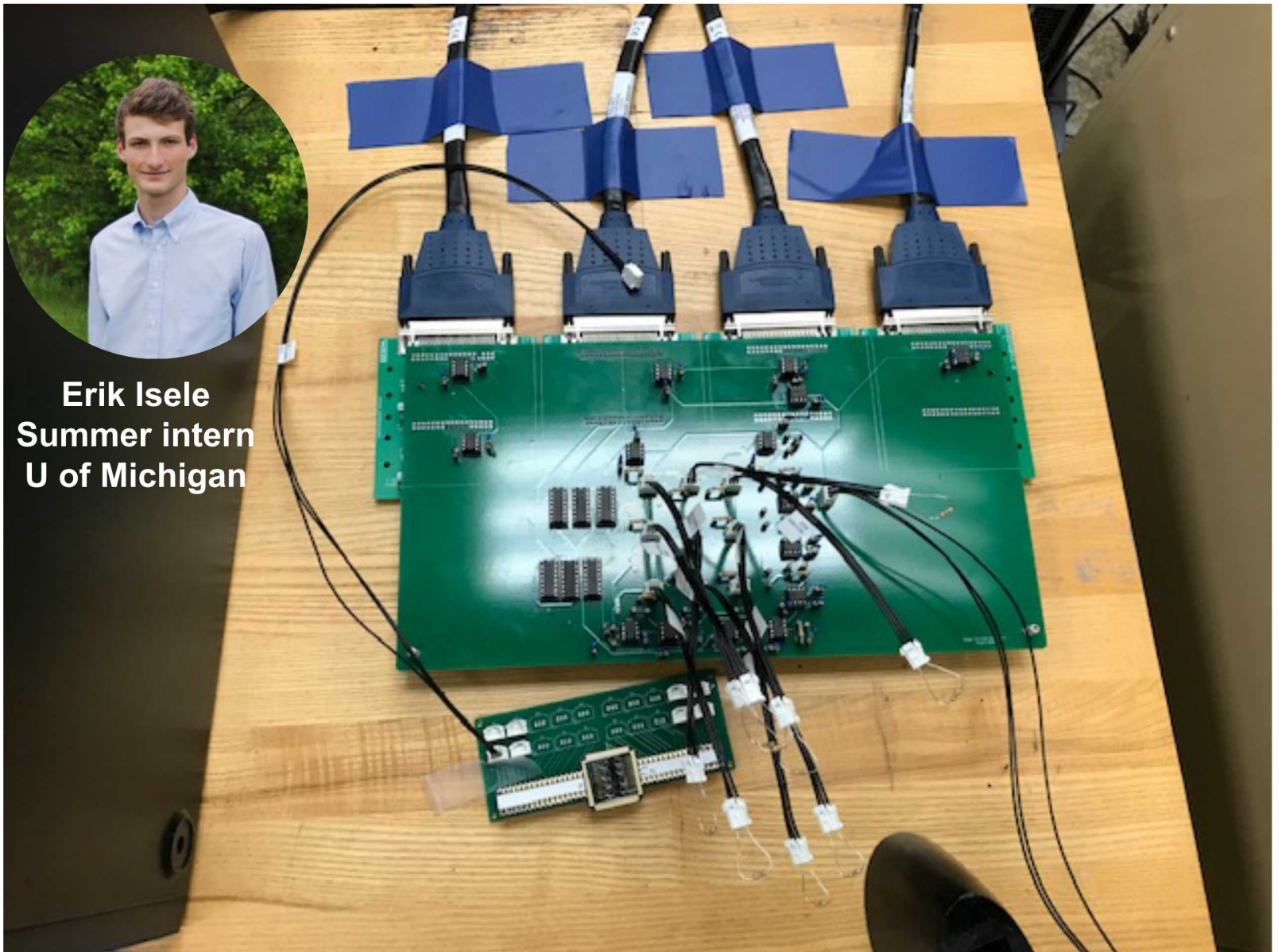
We can select the rows and columns in this crossbar to update the weights of many synapses in a highly parallel fashion in accordance with an outer product update



Next step: learning demonstration



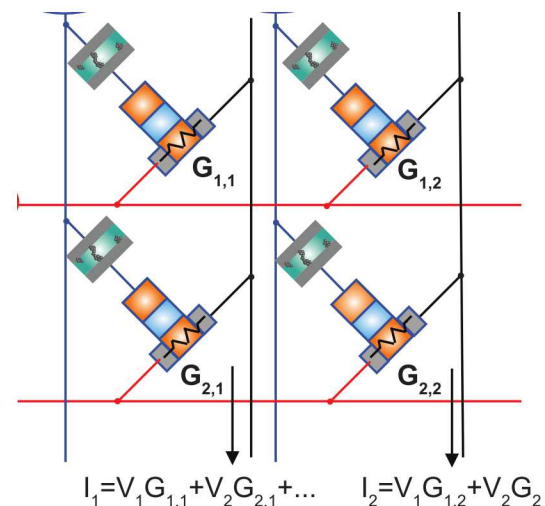
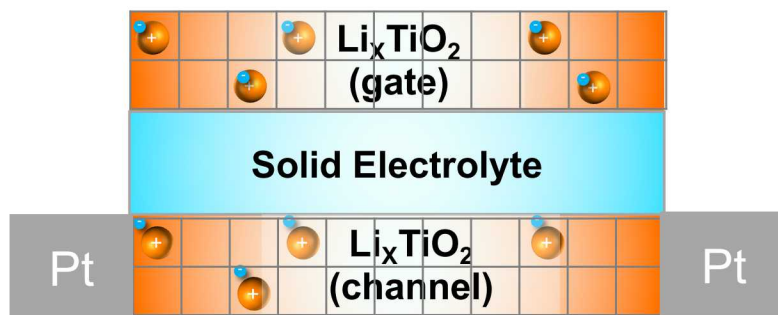
Erik Isele
Summer intern
U of Michigan





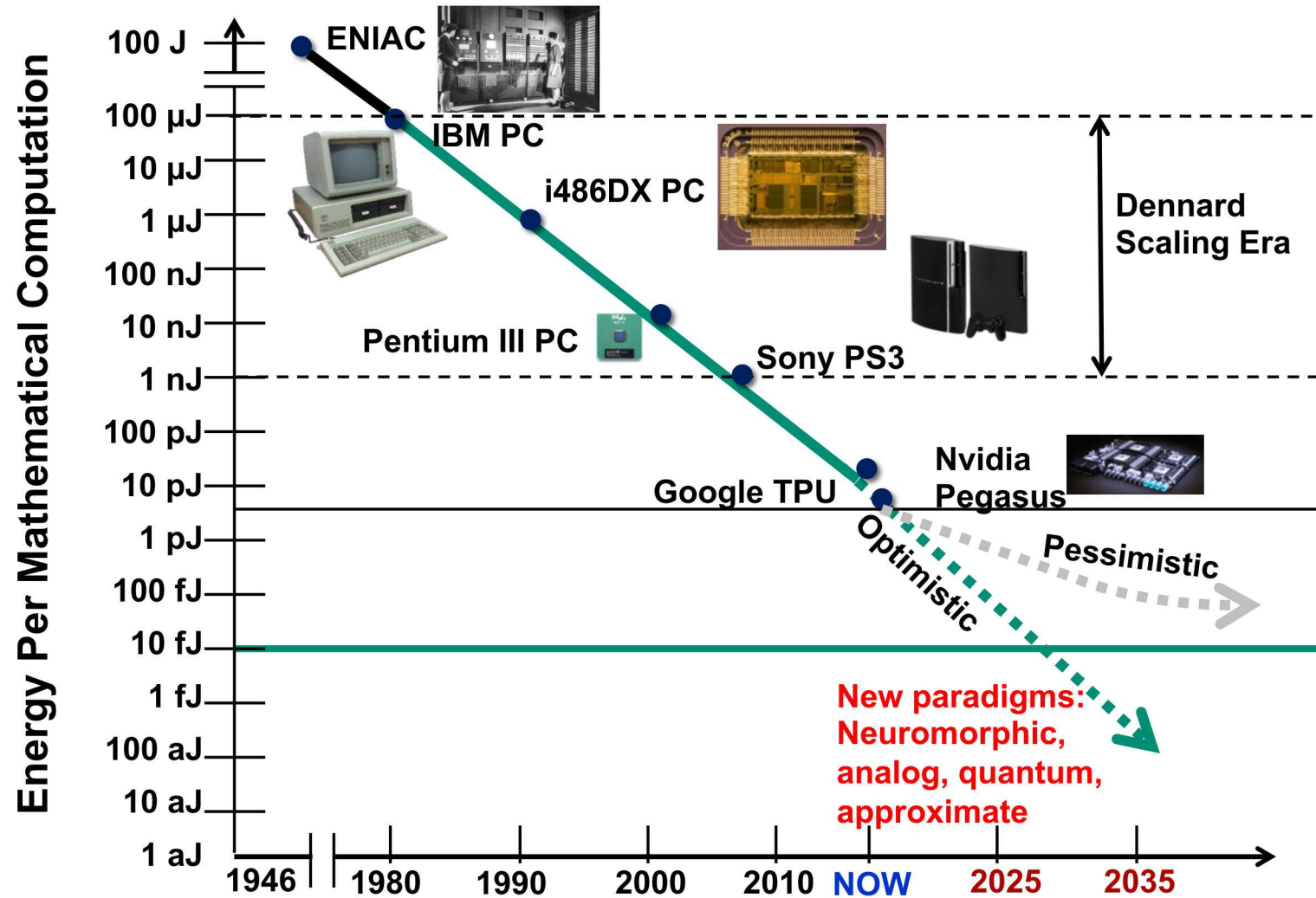
Summary

- In-memory analog computing can be much more energy efficient than digital computing, but there exists significant materials challenges.
- Redox transistors based on electrochemical ion insertion fulfill the energy and accuracy needs for in-memory computing.
- Super-capacitive redox transistors attain sufficient speeds for most neural applications
- Crossbar arrays enable parallel weight updates at low energy.





Evolution of Computing Machinery



Just as artificial intelligence can be used to advance materials science,
materials science can be used to advance artificial intelligence



Acknowledgements

Sandia

Alec Talin

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