Neuromorphic Computing with Polymers

PRESENTED BY

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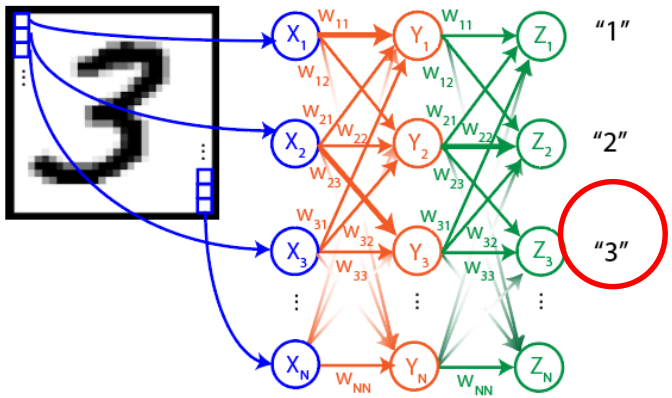
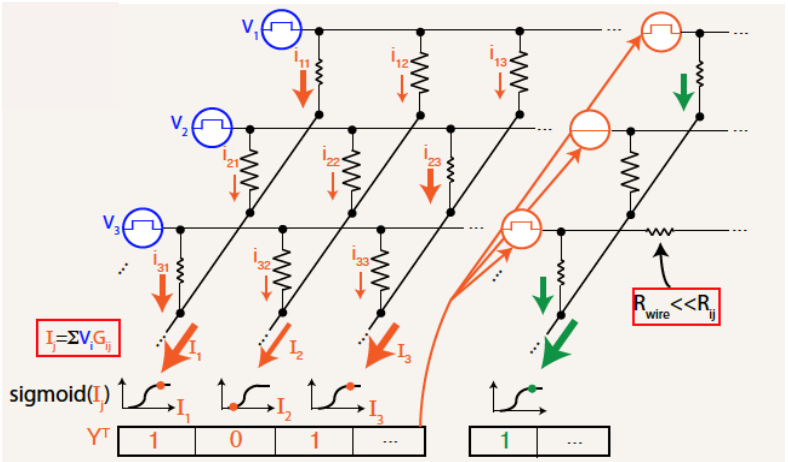
One Laboratory – Two primary sites

Albuquerque

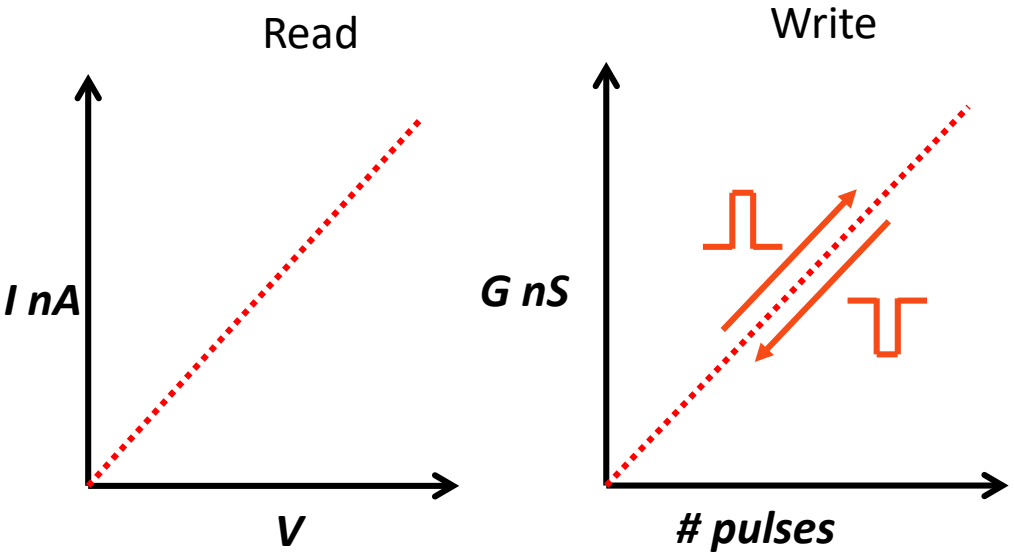
Livermore

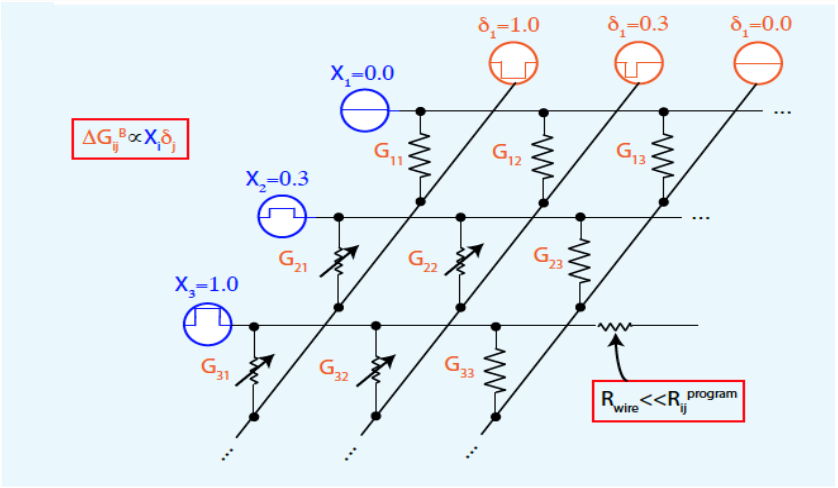
Need: Ohmic I-V, low conductance, fast, high endurance





vector-matrix multiply (inference)





outer product (learning)

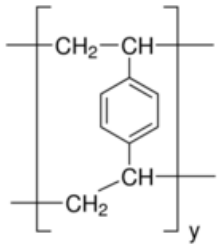
Organic non-volatile memory R&D >5 decades

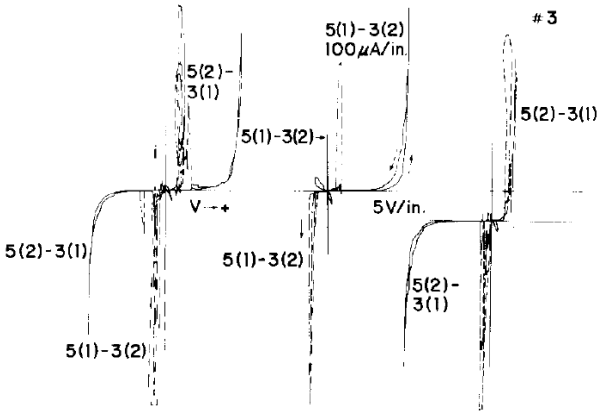
ELECTRICAL CONDUCTIVITY OF POLYDIVINYLBENZENE FILMS*

L. V. GREGOR

IBM Components Division, East Fishkill Facility, Hopewell Junction, N.Y. 12533 (U.S.A.)

(Received December 30, 1967; in revised form April 1, 1968)





Electrical switching and memory phenomena in Cu-TCNQ thin films

R. S. Potember and T. O. Poehler

Applied Physics Laboratory, Johns Hopkins University, Laurel, Maryland 20810

D. O. Cowan

Department of Chemistry, Johns Hopkins University, Baltimore, Maryland 21218

(Received 20 November 1978; accepted for publication 10 January 1979)

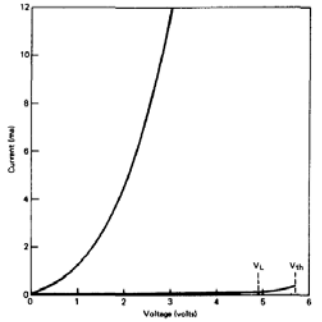


FIG. 1. Typical dc current-voltage characteristic showing bistable switching in a 5-μm-thick Cu-TCNQ sandwich structure.

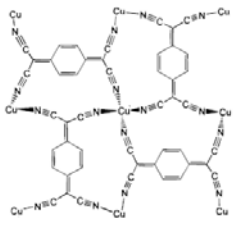
Inorg. Chem. 1999, *38*, 144−156

New Insight into the Nature of Cu(TCNQ): Solution Routes to Two Distinct Polymorphs and Their Relationship to Crystalline Films That Display Bistable Switching Behavior

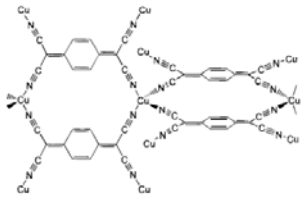
Robert A. Heintz,† Hanhua Zhao,† Xiang Ouyang,† Giulio Grandinetti,† Jerry Cowen,‡ and Kim R. Dunbar*,†

Departments of Chemistry and Physics & Astronomy and The Center For Fundamental Materials Research, Michigan State University, East Lansing, Michigan 48824

Received October 13, 1998



Cu(TCNQ) Phase I

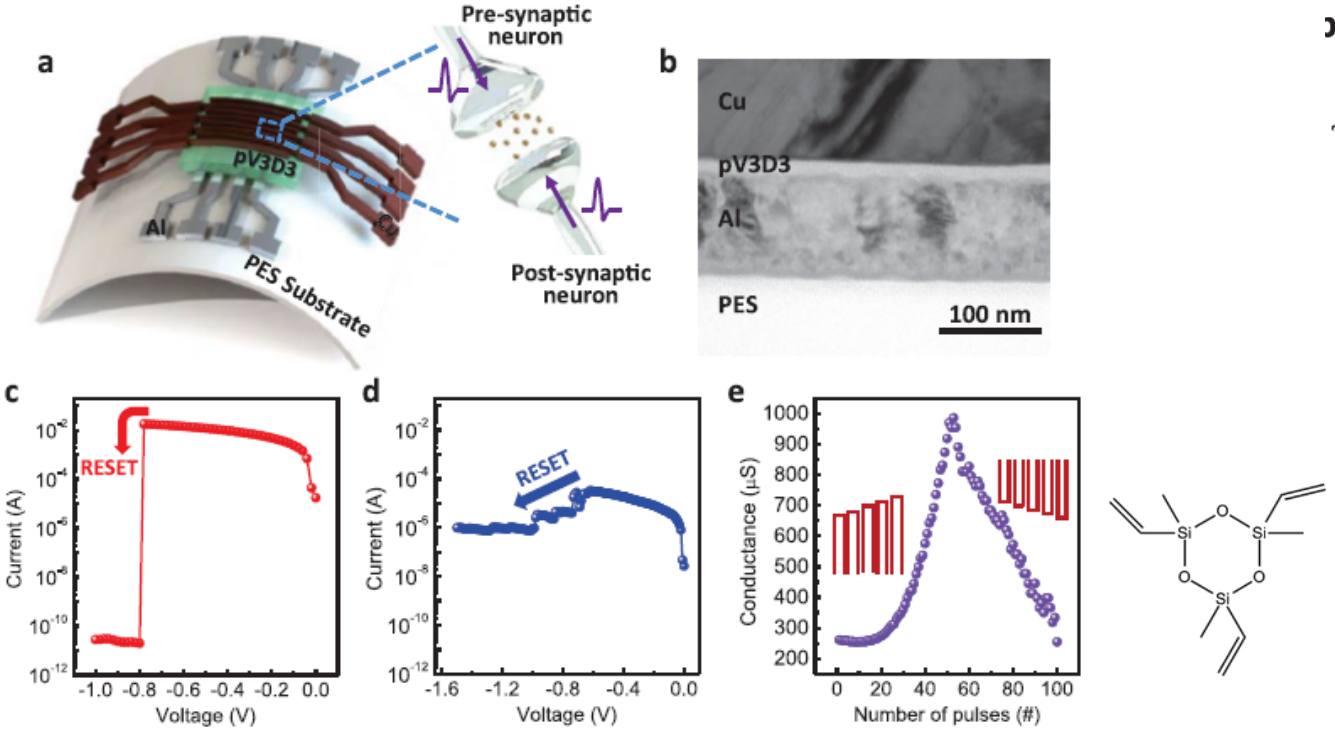


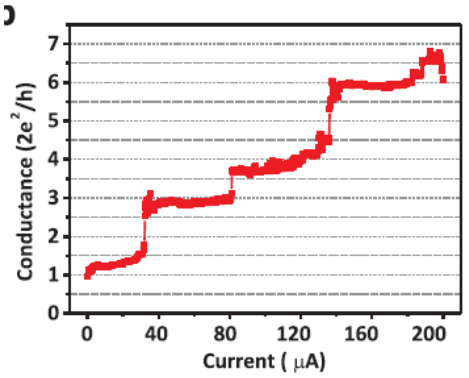
Cu(TCNQ) Phase II

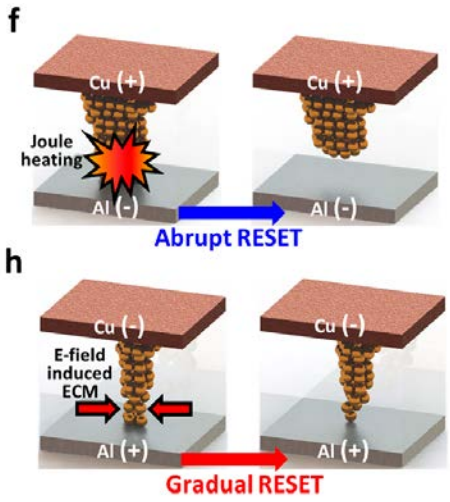
Polymers memristors

Recent review: Gao et al., Chem. Soc. Rev., 48, 1531, (2019)

- ***Many proposed mechanisms….***
- ***Non linearity, asymmetry, noise, need for reset, high conductance…***

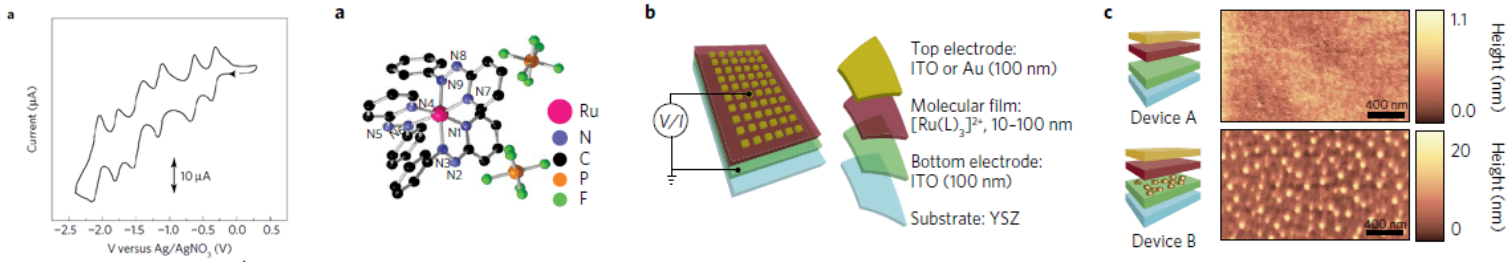


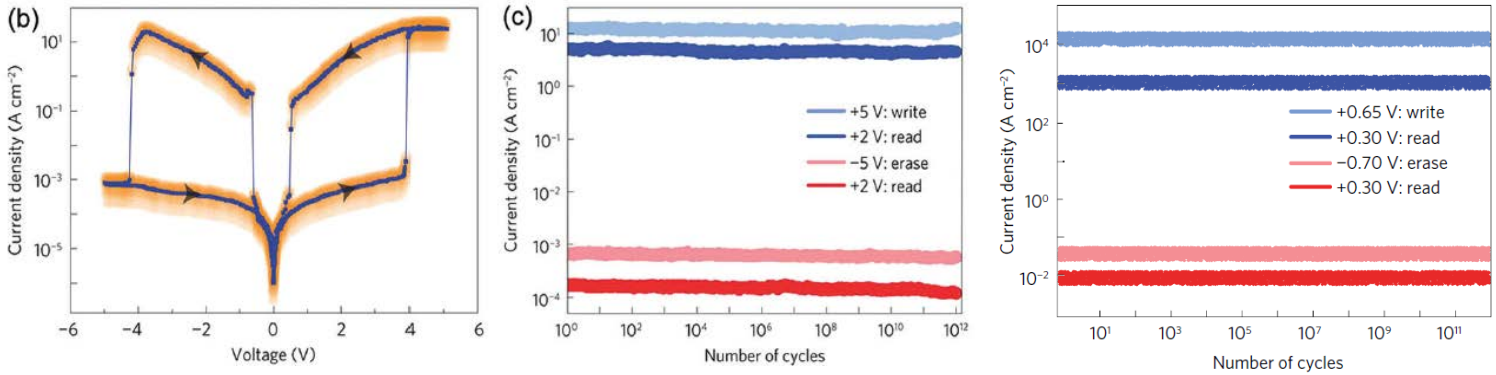


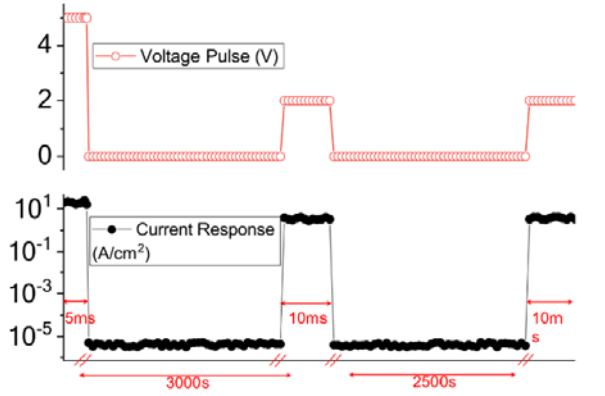


B. C. Jang et al., Nano Lett. 19, 839, (2019); pV3D3: trivinyl-trimethyl cyclotrisiloxane

Solution processable azo-aromatic metal complexes



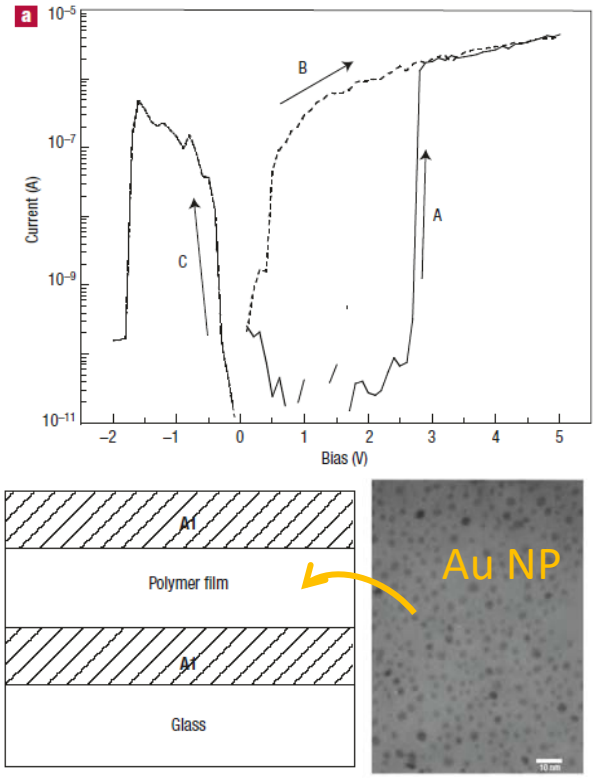




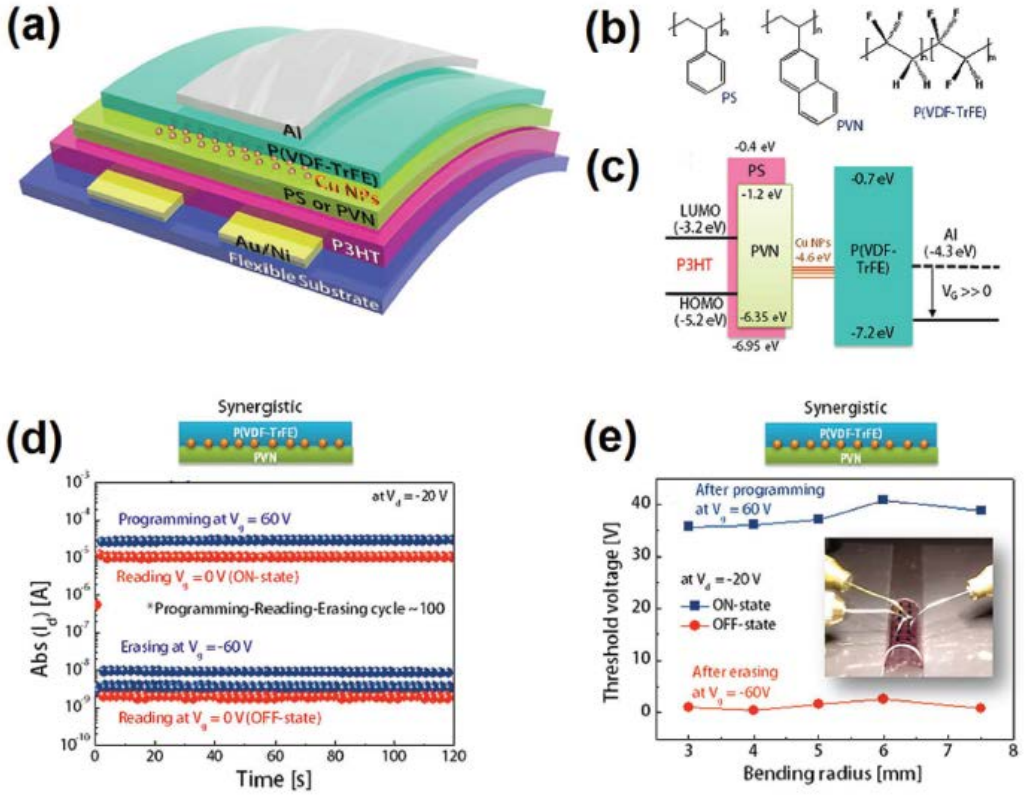
S. Goswami et al., Nat. Materials 16, 1216 (2017)

Charge trapping polymer memory (organic Flash)

High write voltage; retention time 10^3 – 10^8 sec

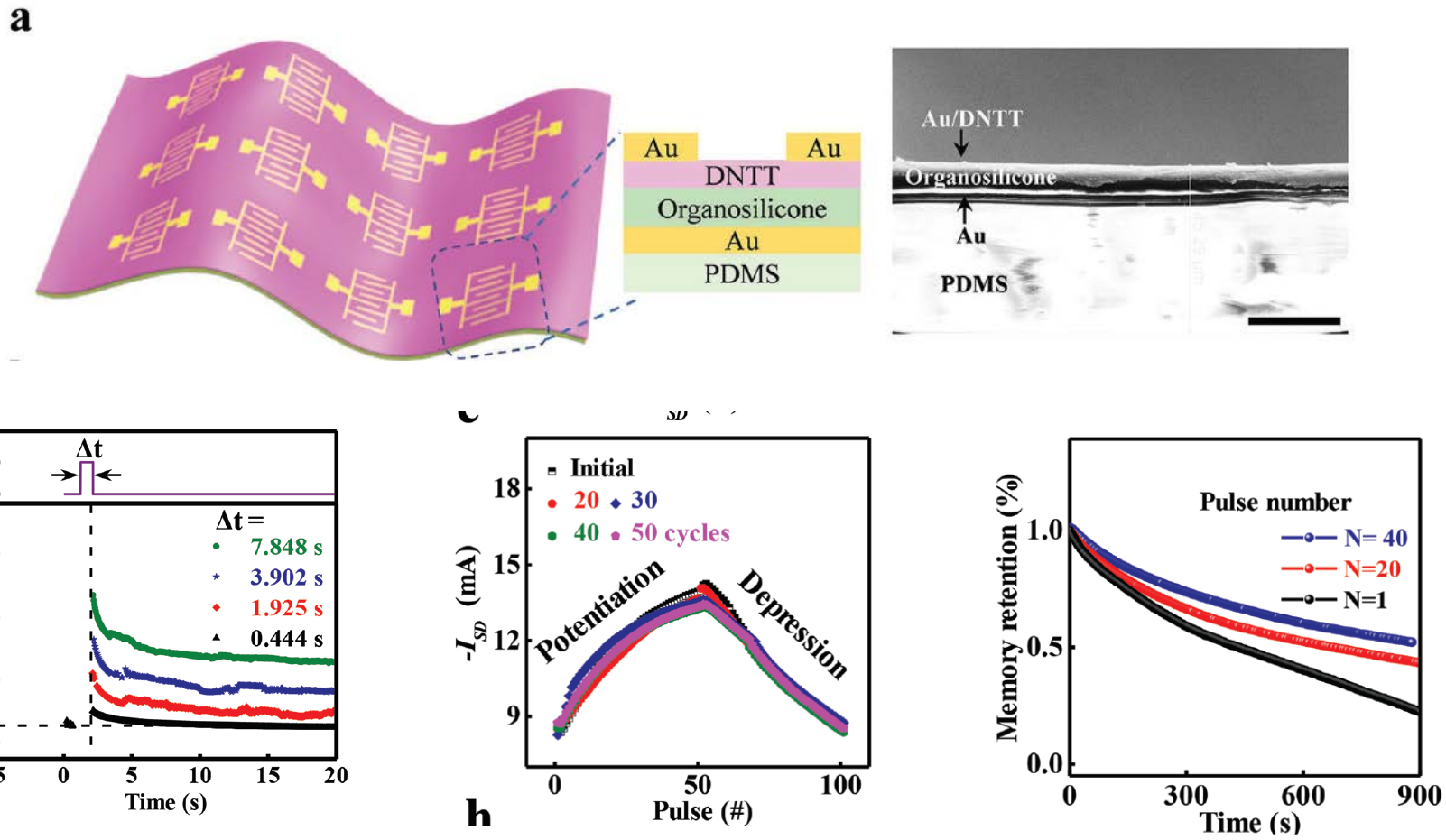


Ouyang et al., Nature Materials 3, (2004)



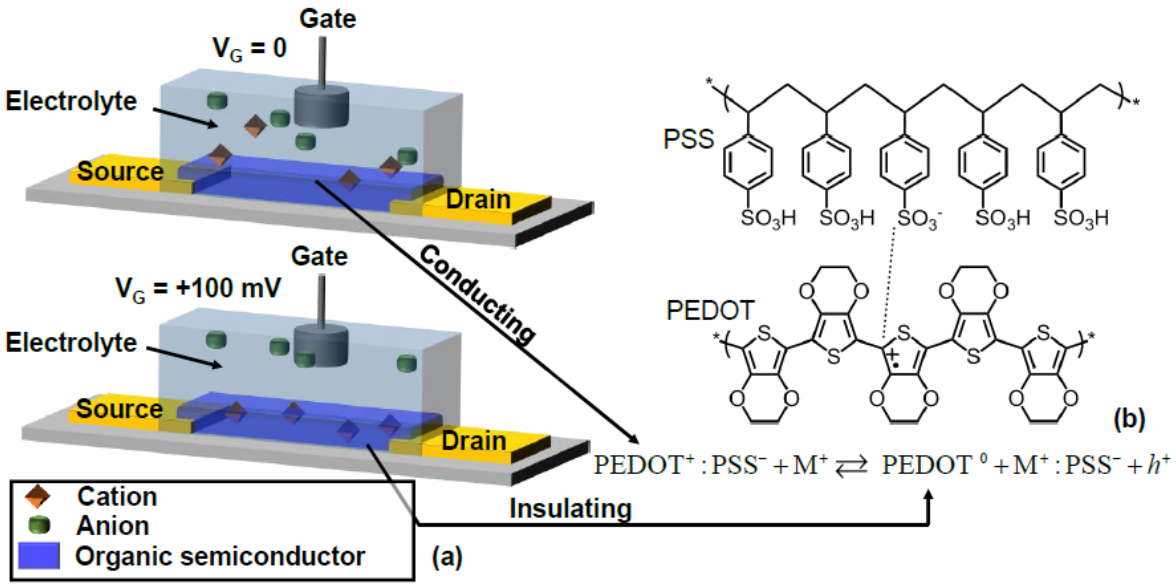
Scientific Reports 5, 12299 (2015)

Organic polarization transistor

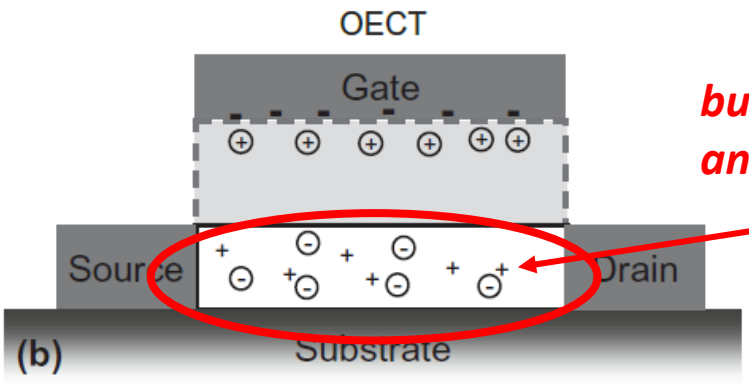


Wang et al., Adv. Funct. Mater. 29, 1901107 (2019)

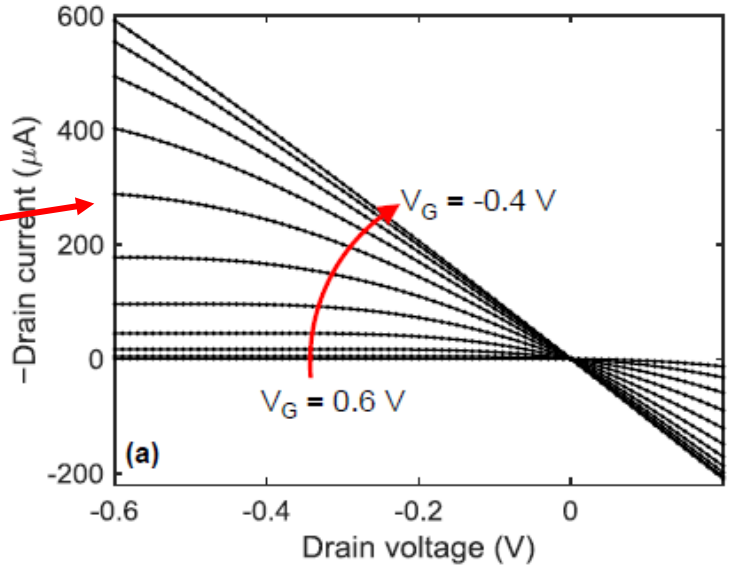
Organic electrochemical transistors (OECTs)



Friedlein et al., Organic Electronics 63 398, (2018)



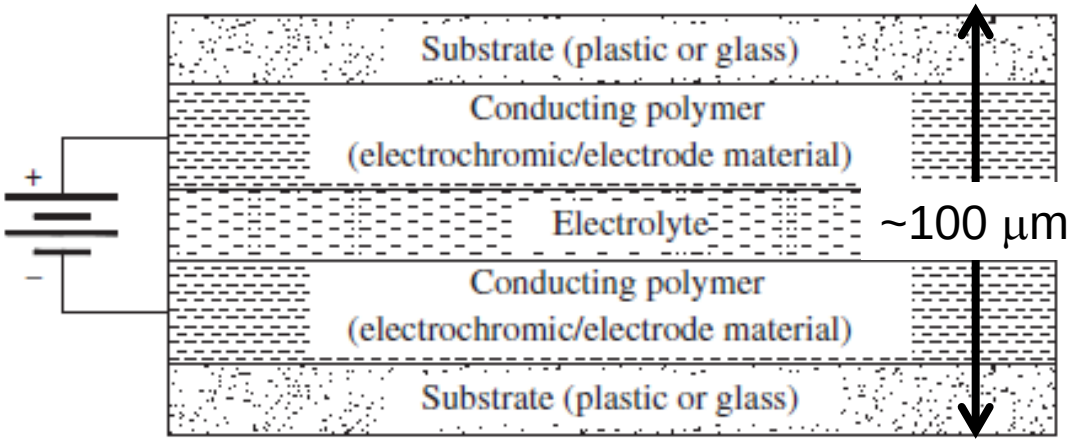
bulk redox analog states



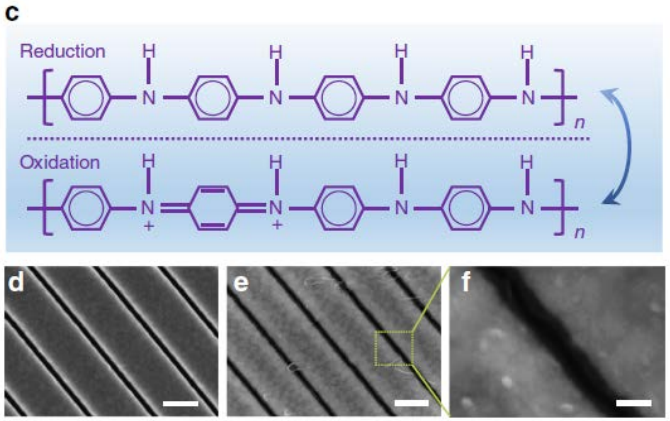
OECT is a volatile device (like a FET)

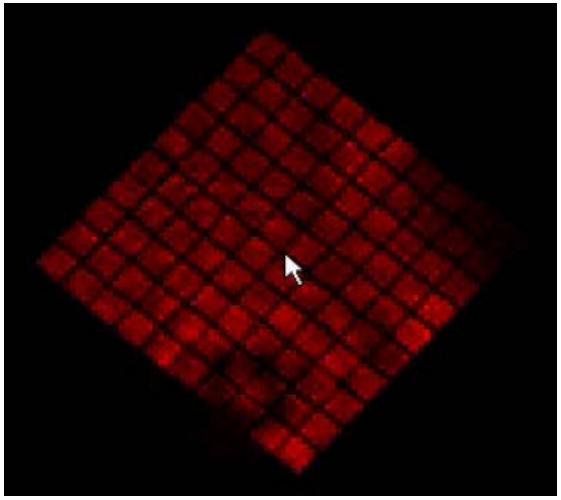
Electrochromics store optical absorption (state of charge)

https://www.gentex.com/products-technology/automotive/dimmable-glass/



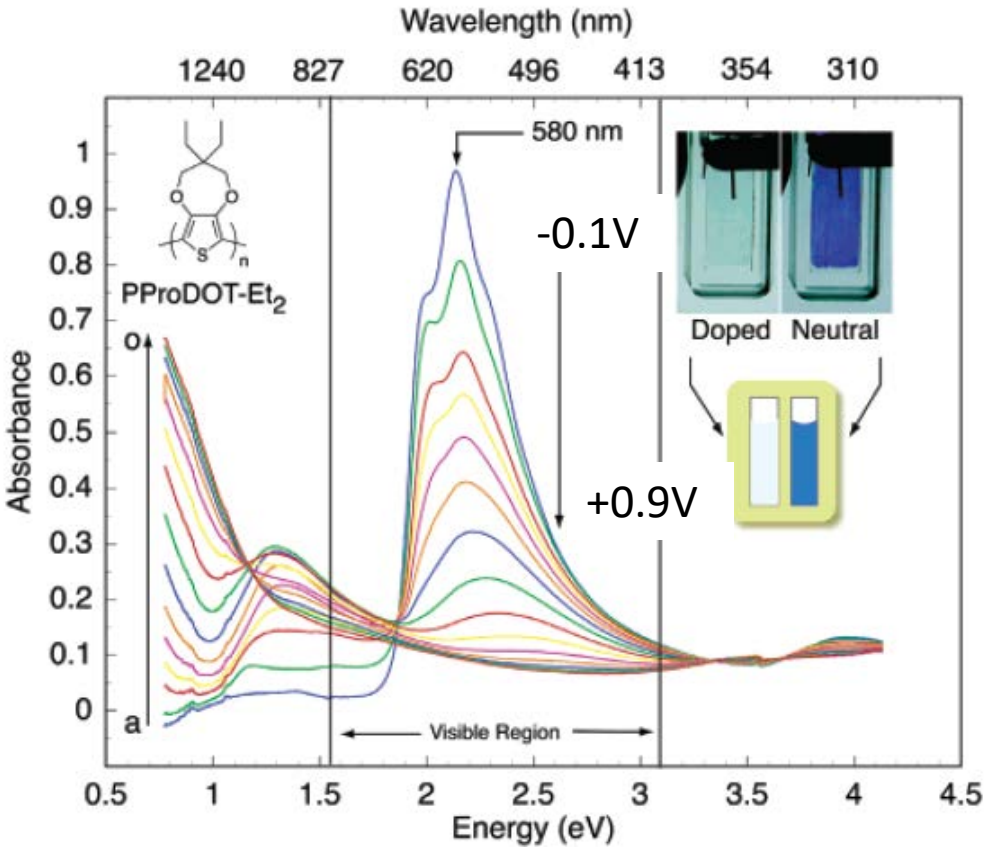
F. Carpi, D. De Rossi Opt. Laser Tech. 38, 292 (2006)



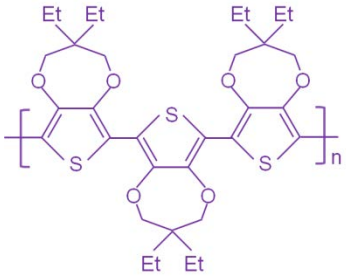


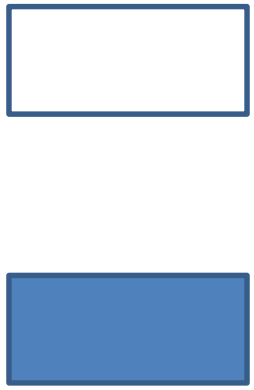
T. Xu, E. C. Walter, A. Agrawal, C. Bohn, J. Velmurugan, W. Zhu, H. J. Lezec, A. A. Talin, *Nature Comm*. 7, 10479, (2016).

Analog optical state tuning of PEDOT:PSS

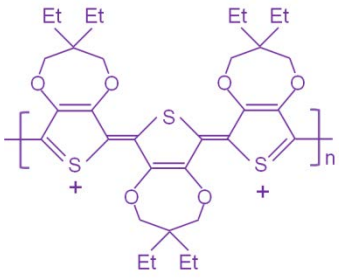


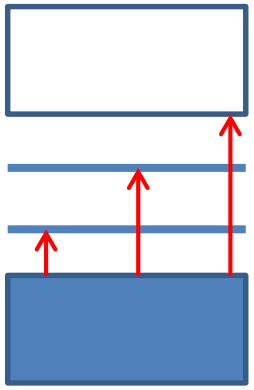
Neutral/
opaque





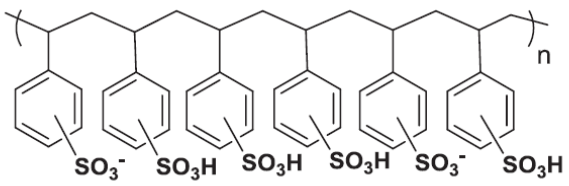
Doped/
transparent (Vis)



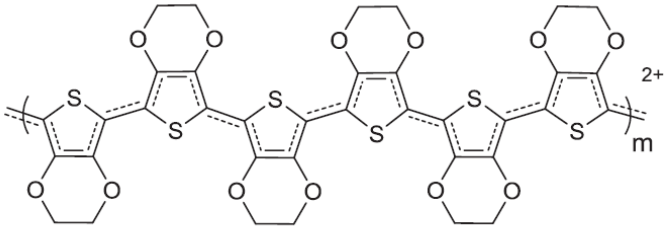


Argun et al., Chem. Mater., *16, 4401, (2004)*

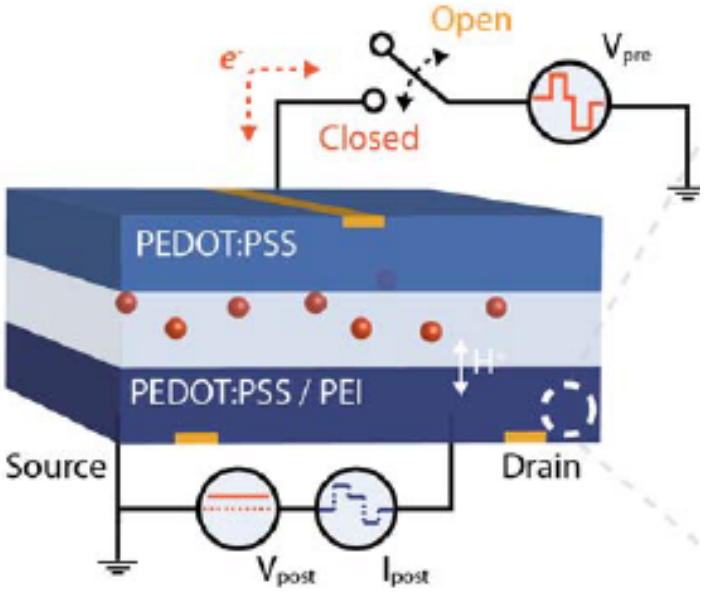
Polymer redox transistor inspired by ECs, battery

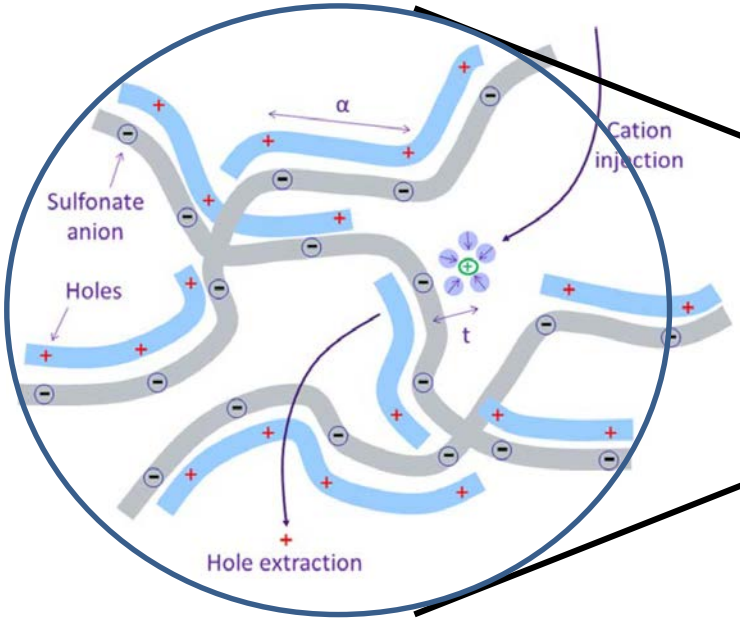


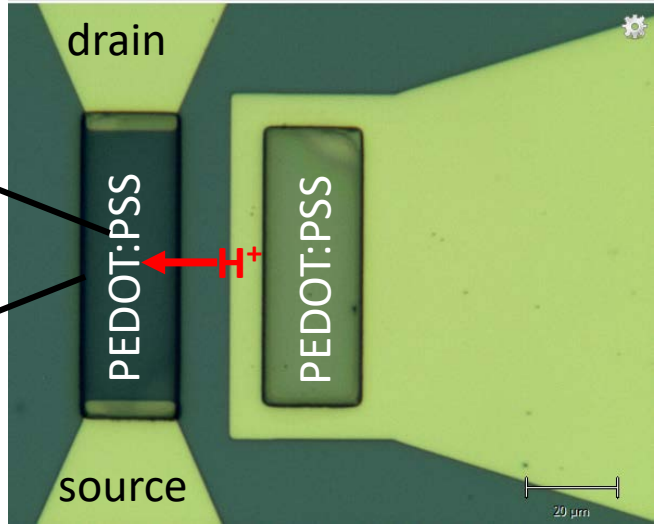
PSS



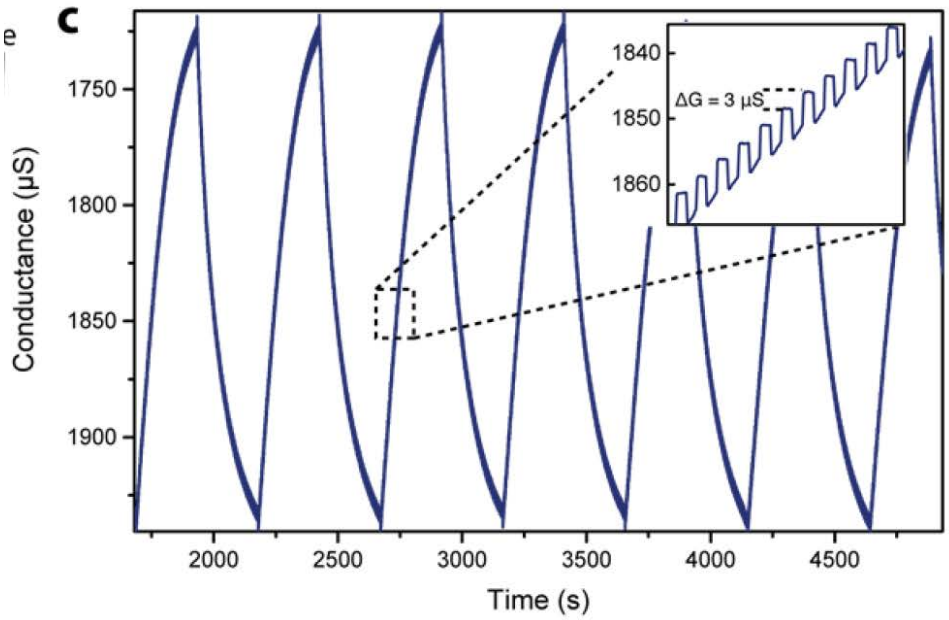
PEDOT

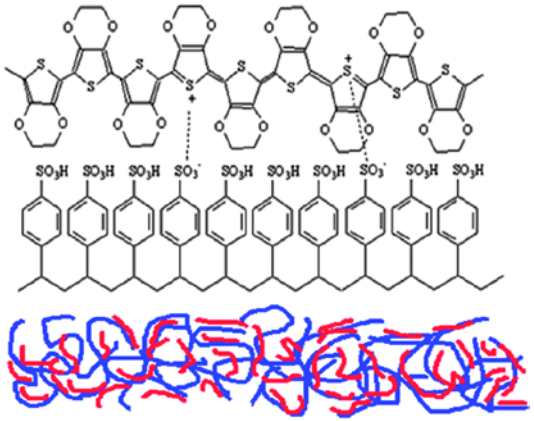


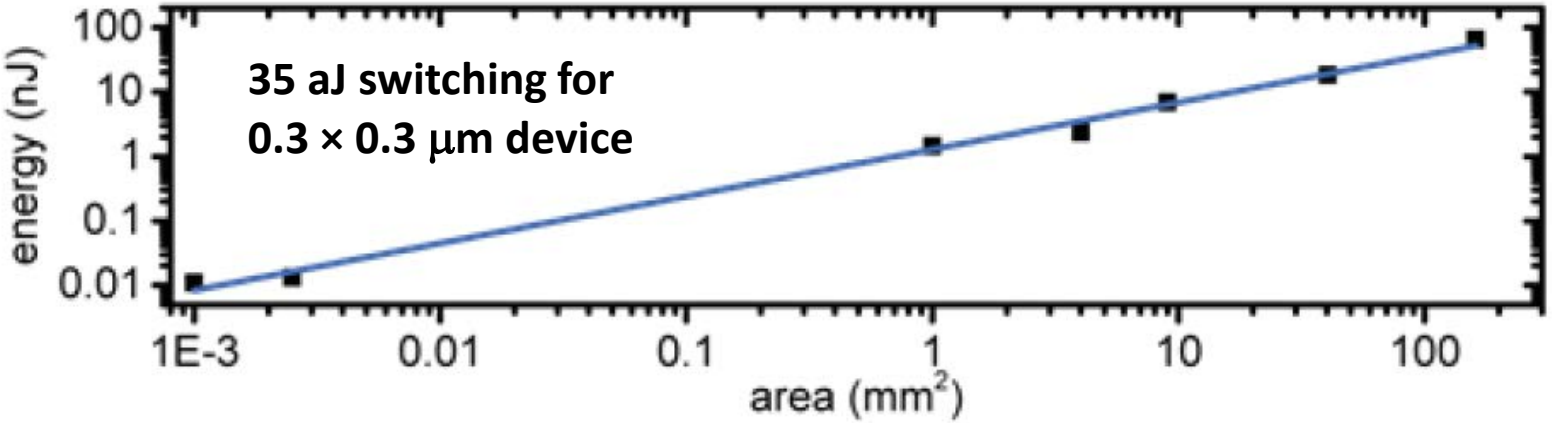




Projected switching energy of 35 aJ for 0.1 μm² device

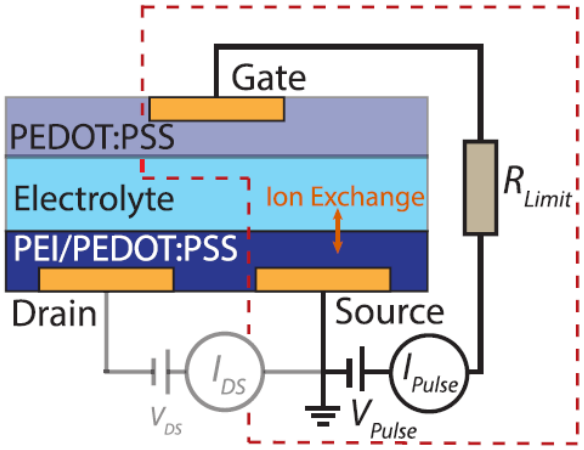


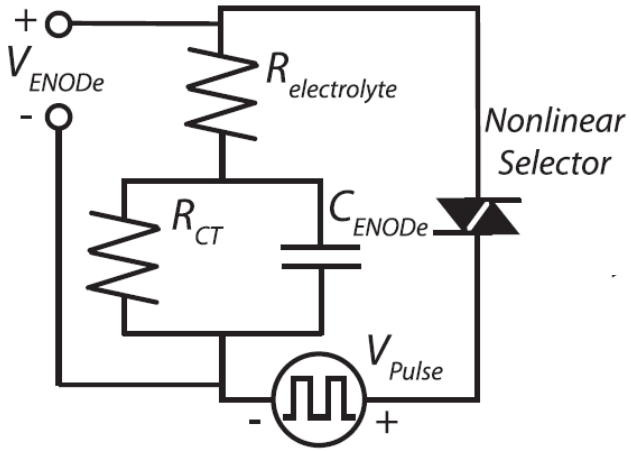


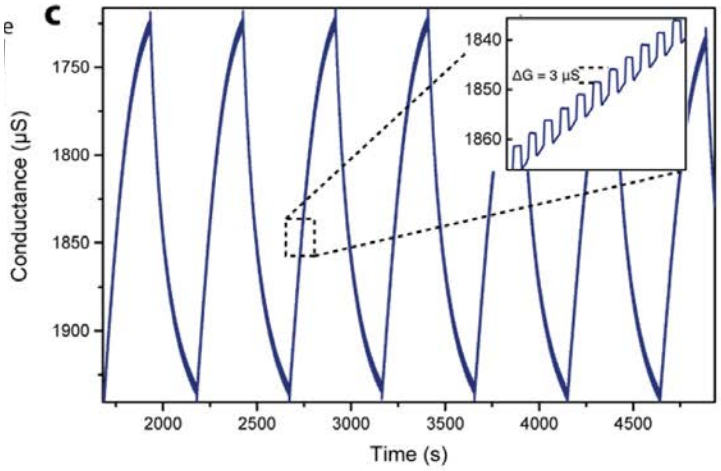


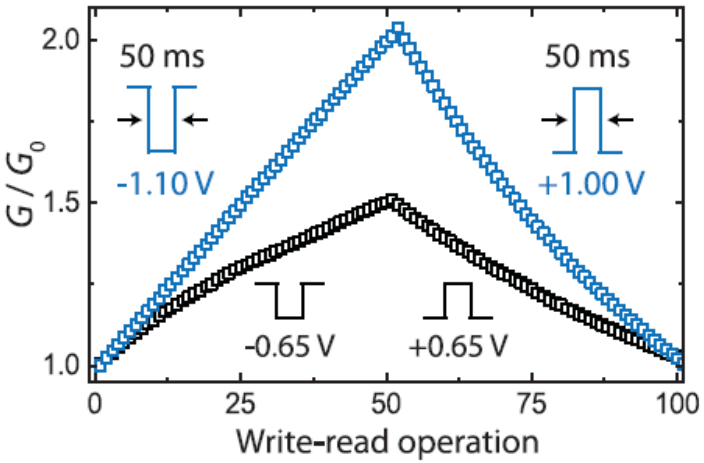
Y.B. van de Burgt, E. Lubberman, E.J. Fuller, S.T. Keene, G.C. Faria, S. Agarwal, M.J. Marinella, A.A. Talin*, A. Salleo*, Nature Materials 16, 414, 2017

Selector enables fast, low-V switching + retention



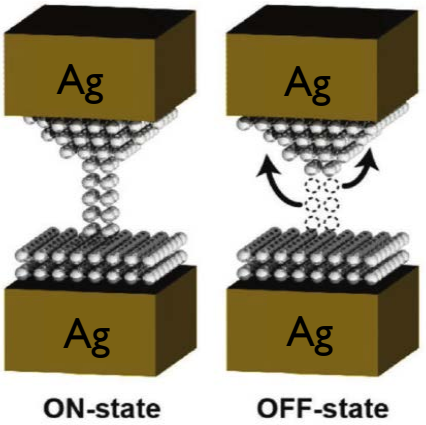






Use binary, volatile selector

selector: diffusive memristor
normally OFF, $V_T = 150 - 400$ mV

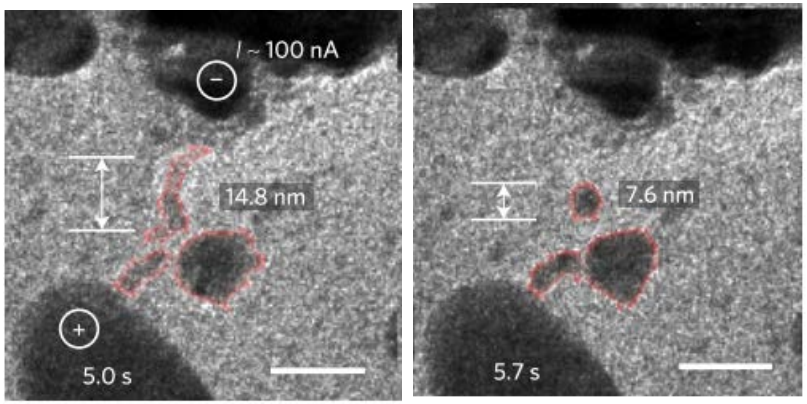


SiO_x

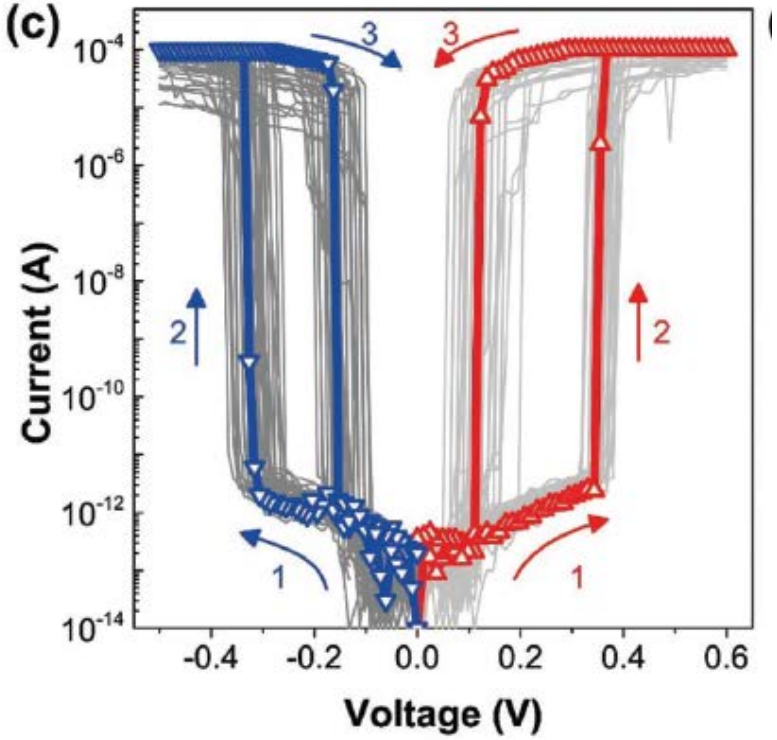
E_a ~10 kT

Pt/Ag/SiO_x:Ag/Ag/Pt

Midya et. al., *Advanced Materials*, 29(12), 2017



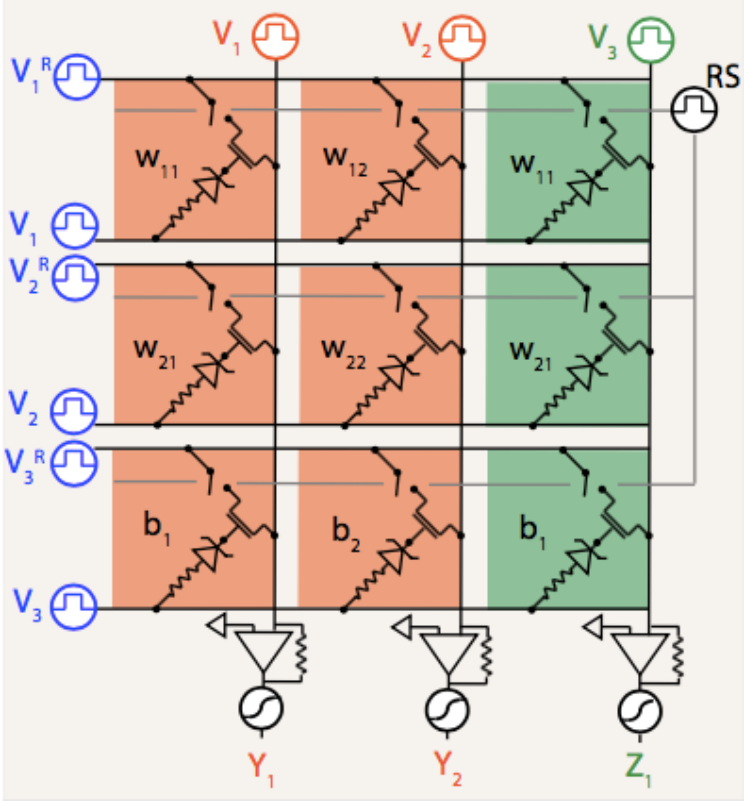
Wang et. al. *Nature Materials* 16, 101-108 (2016)

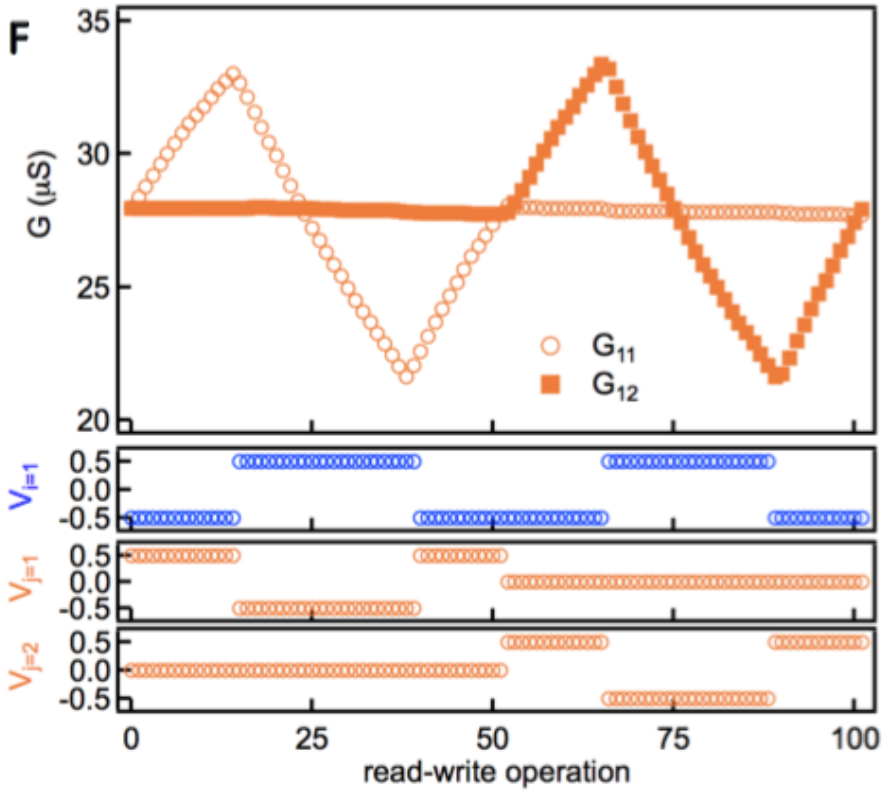


symmetric thresholding mechanism for program selectivity within array

1 mV/decade slope

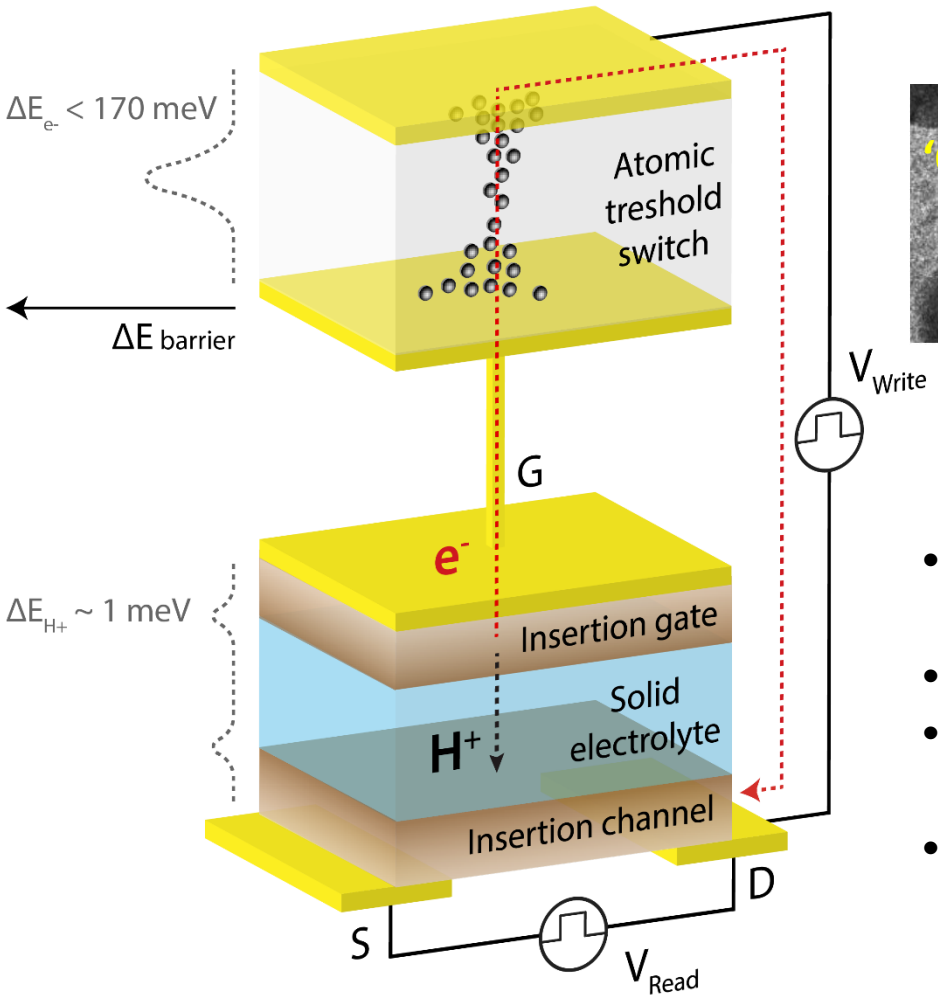
First test of addressability



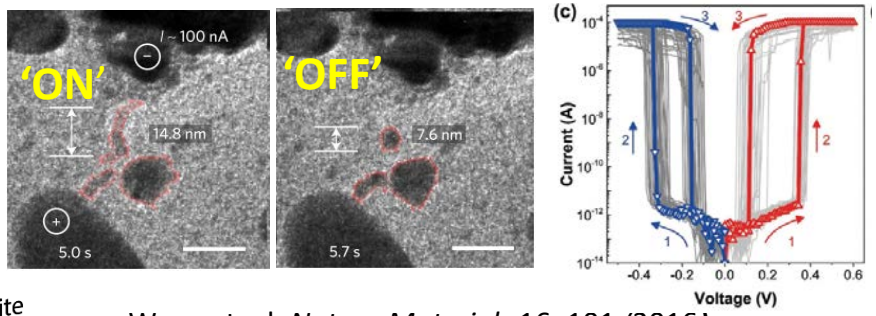


E. J. Fuller, S. T. Keene, A. Melianas, Z. Wang, S. Agarwal, Y. Li, Y. Tuchman, C. D. James, M. J. Marinella, J. J. Yang, A. Salleo, A. A. Talin, *Science*, online

Non-volatile redox memory – a floating gate memory



Threshold switch: only on for V>V$_{th}$

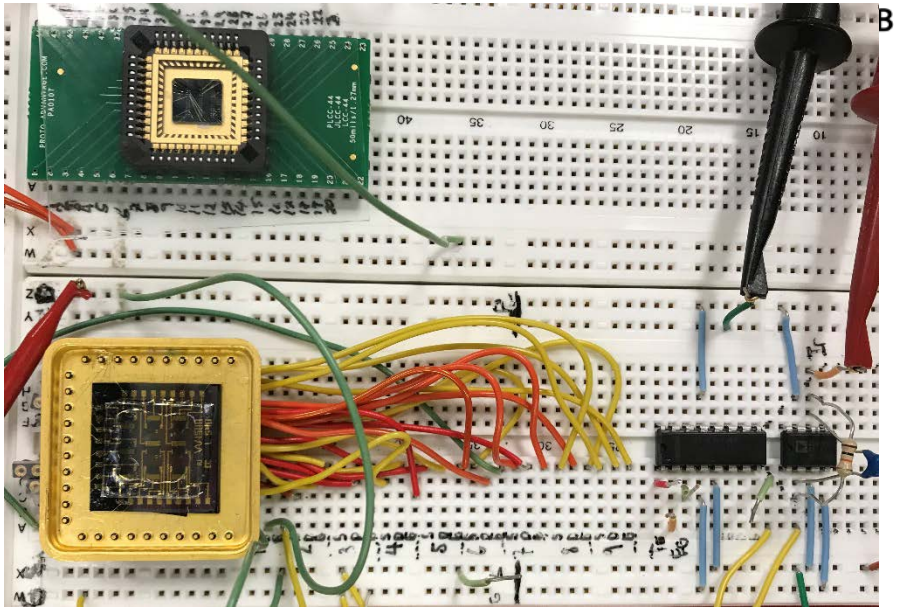


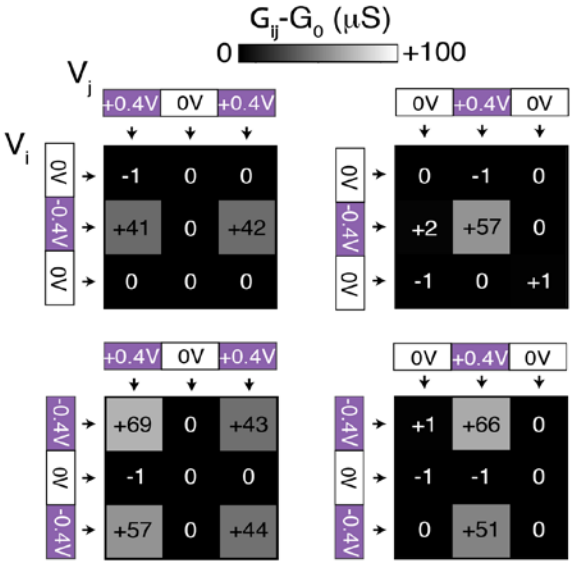
Wang et. al. *Nature Materials* 16, 101 (2016)

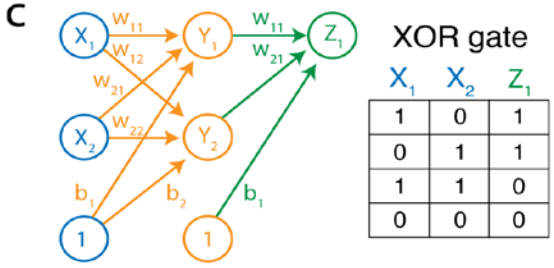
- Redox transistor is an electrochemical concentration cell
- Redox state → channel conductance
- Threshold switch enables electron flow ONLY during 'write' step
- State retained after 'write' because electron flow is cut off.

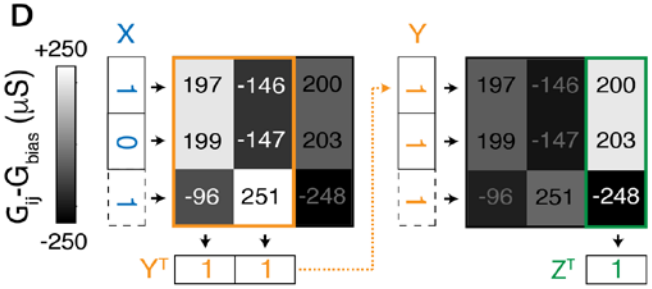
E. J. Fuller, S. T. Keene, A. Melianas, Z. Wang, S. Agarwal, Y. Li, Y. Tuchman, C. D. James, M. J. Marinella, J. J. Yang, A. Salleo, A. A. Talin, *Science*, 364, 570, (2019).

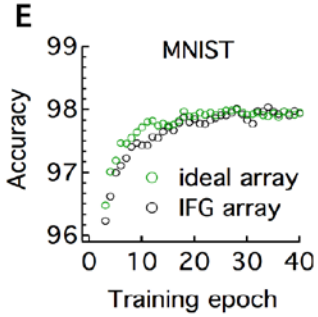
Parallel updates in a 3x3 array, 'XOR' operation

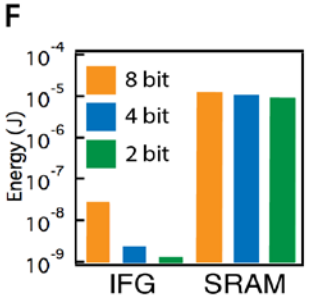


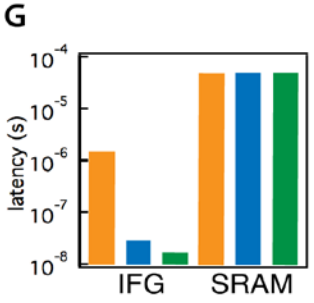


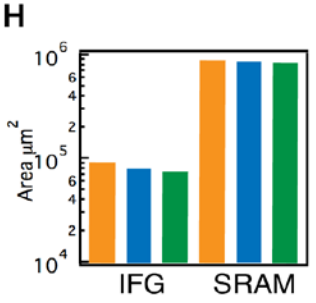






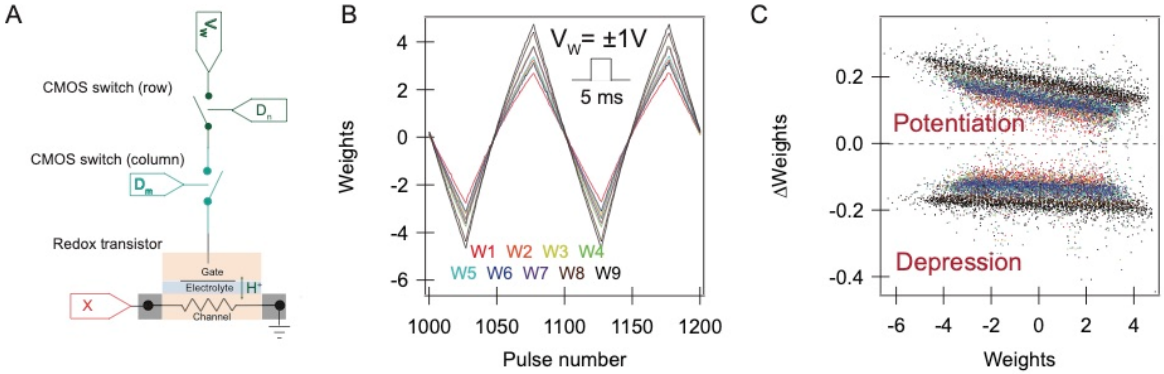


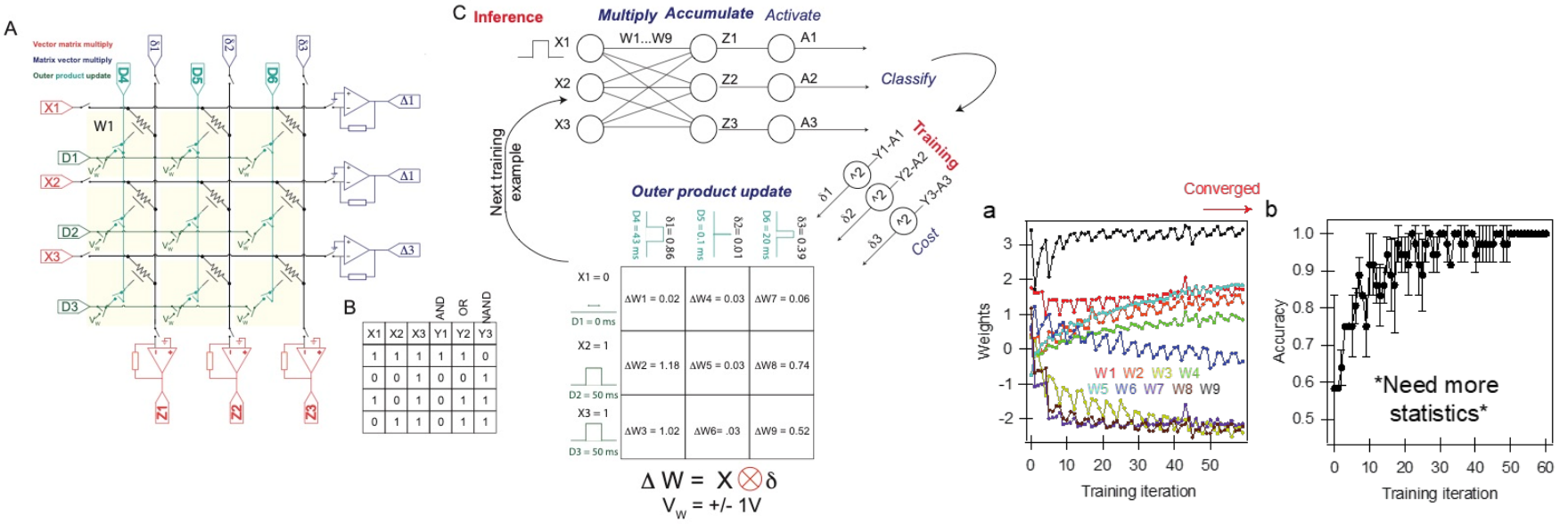




Parallel training with a 3x3 array

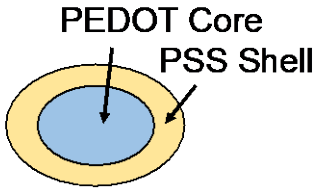
Y. Li et al., in prep.



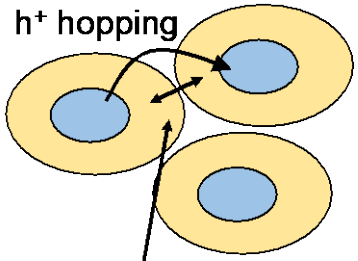


Polymers are tunable

Hypothesis



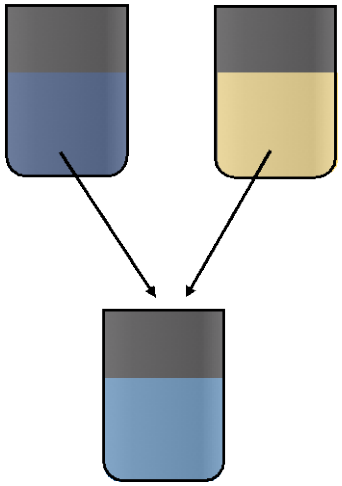
Lang et. al., *Adv. Func. Mater.* (2009), Zhou et. al., *J. Mater. Chem. C* (2014)



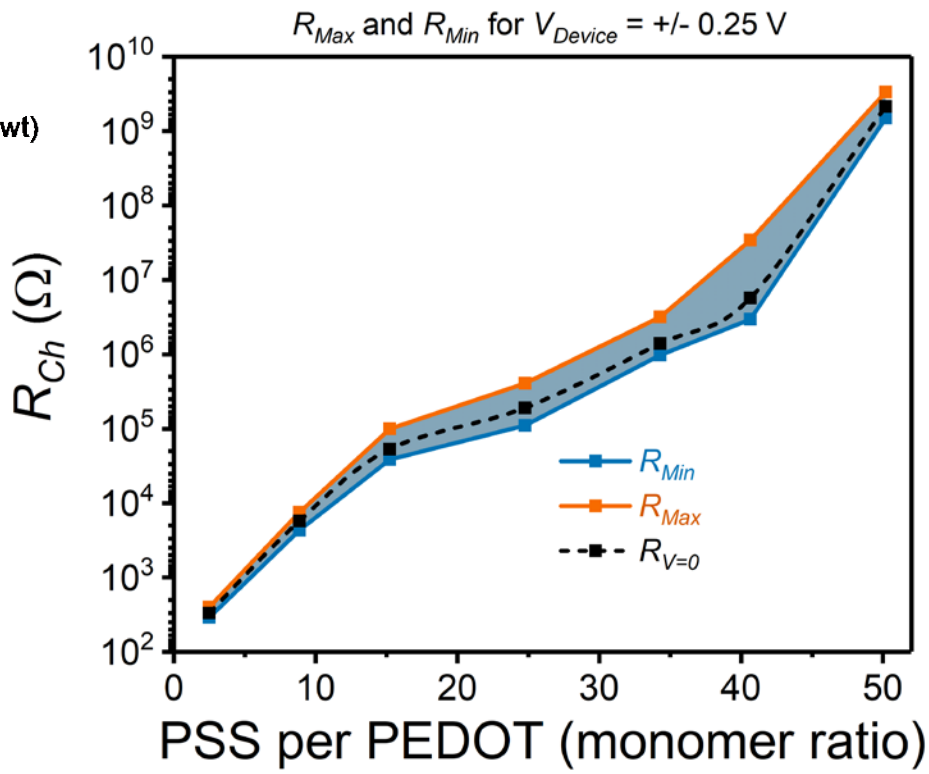
Sample Prep

PEDOT:PSS (PH1000) with EG + DBSA (1.3 %wt)

PSS:Na (1.3 %wt)

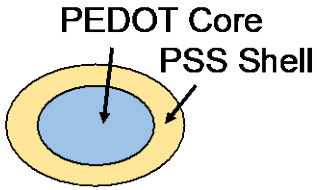


Diluted PEDOT:PSS

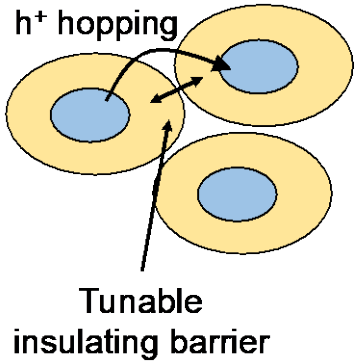


Polymers are tunable

Hypothesis



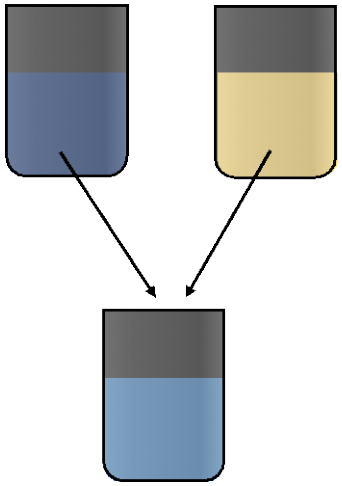
Lang et. al., *Adv. Func. Mater.* (2009), Zhou et. al., *J. Mater. Chem. C* (2014)



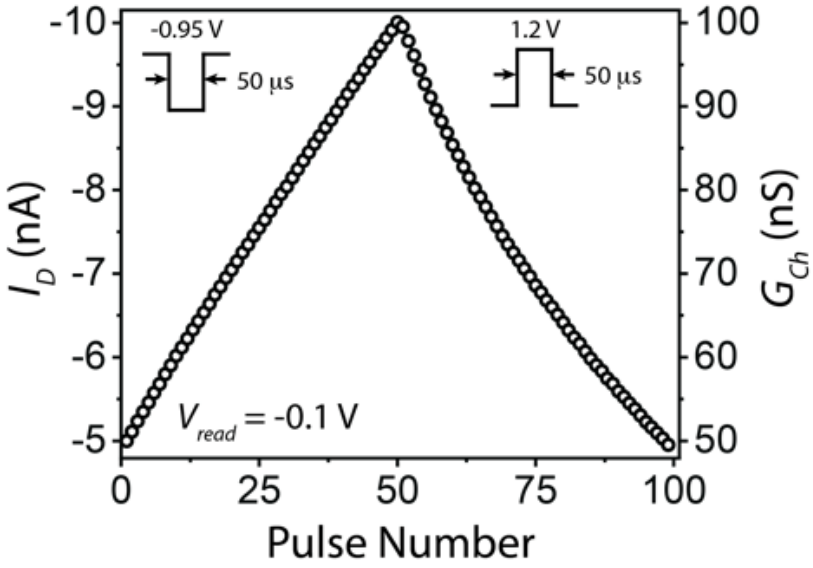
Sample Prep

PEDOT:PSS (PH1000) with EG + DBSA (1.3 %wt)

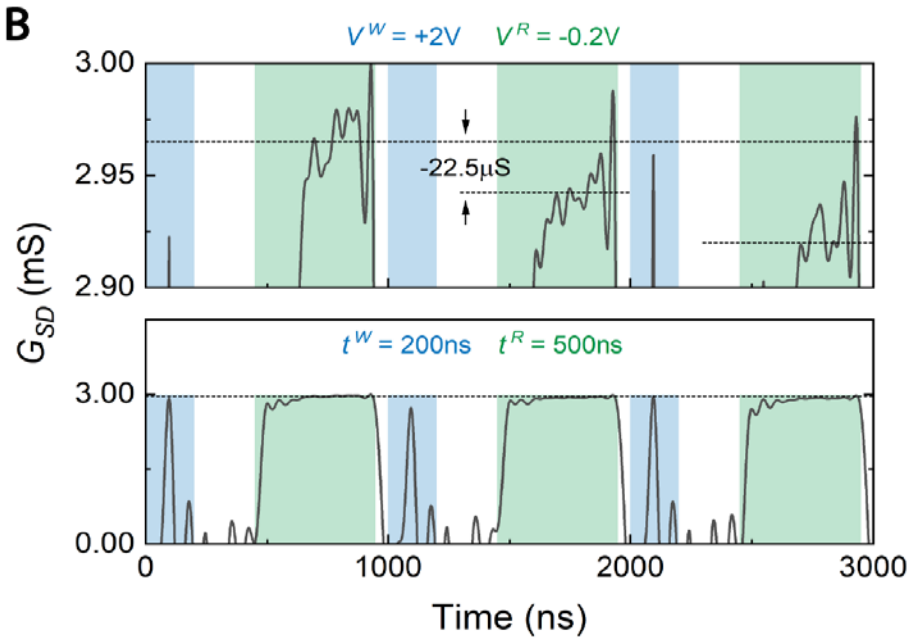
PSS:Na (1.3 %wt)

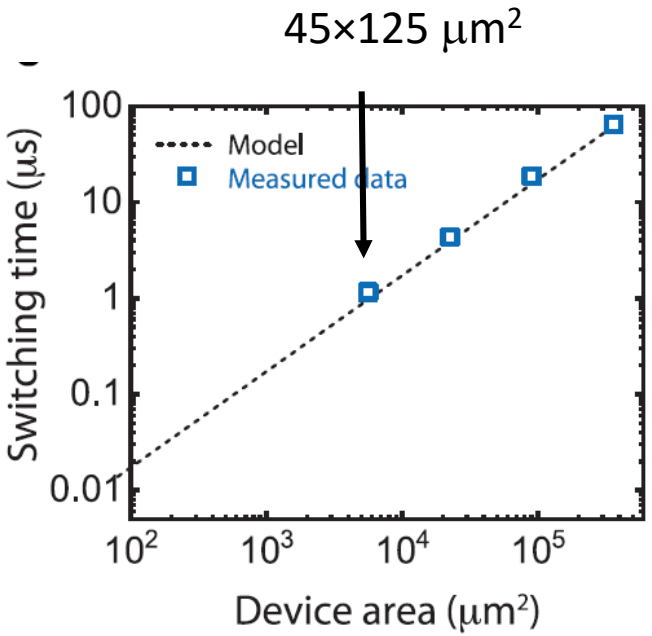


Diluted PEDOT:PSS



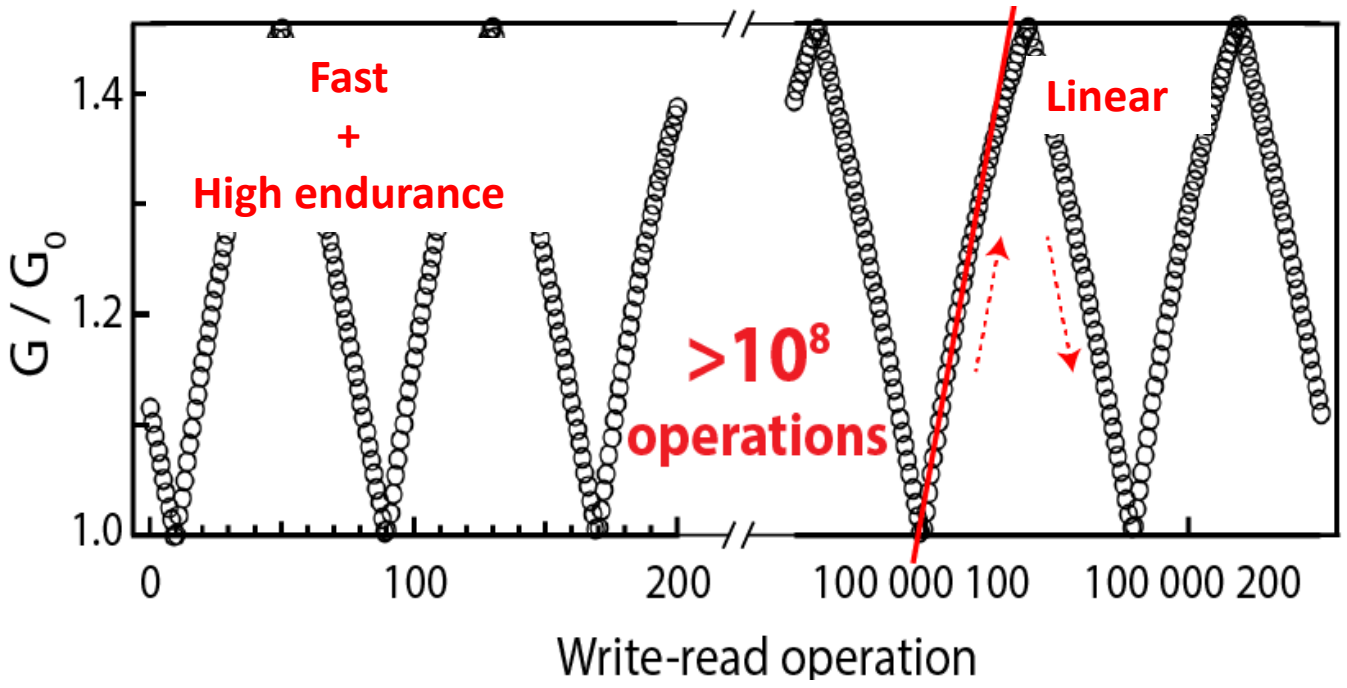
Rapid H^+ transport enables fast switching





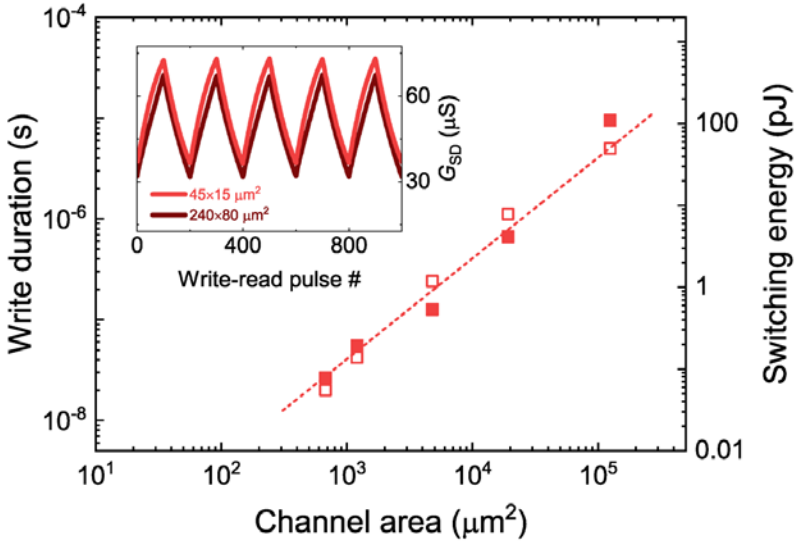
E. J. Fuller, S. T. Keene, A. Melianas, Z. Wang, S. Agarwal, Y. Li, Y. Tuchman, C. D. James, M. J. Marinella, J. J. Yang, A. Salleo, A. A. Talin, *Science* 364, 570, (2019).

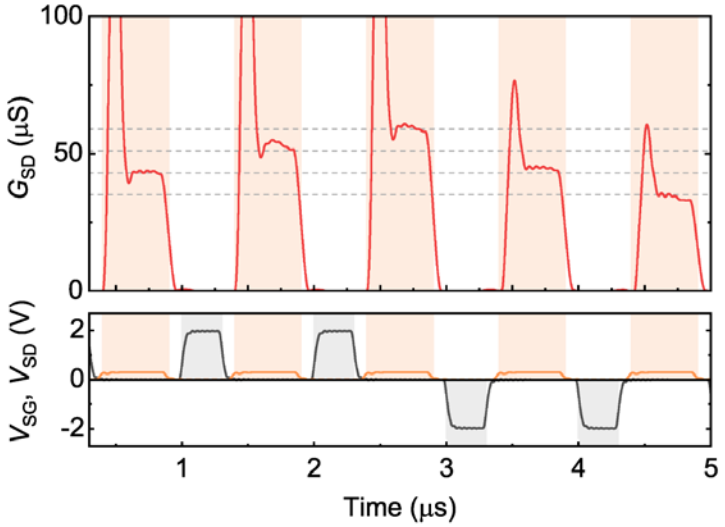
Fast switching enable endurance testing



Further reduction in switching time with scaling

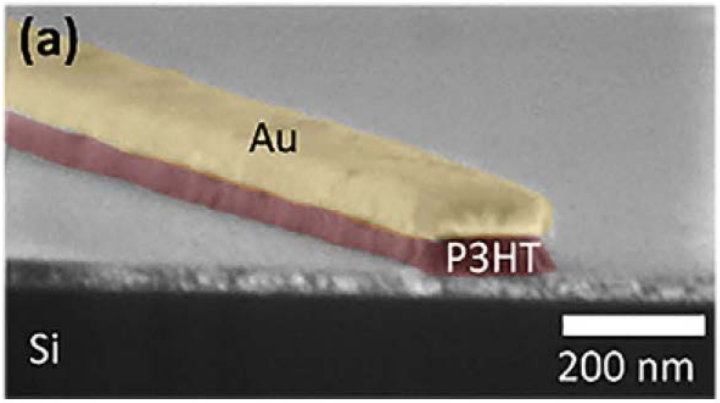
(Salleo group)



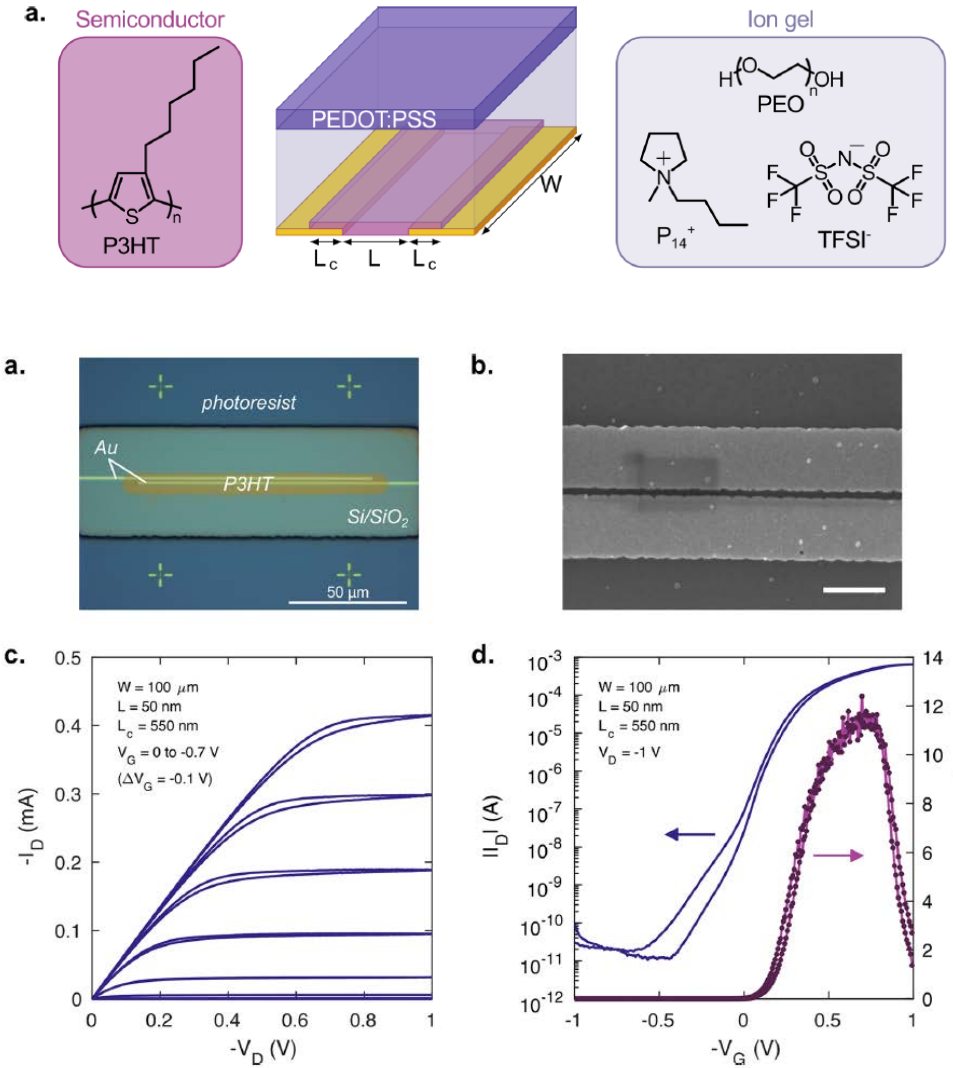


- < 1μs write-read cycle
- 20 ns switching achieved
- < 100 fJ write energy measured

50 nm polymer transistor demonstrated



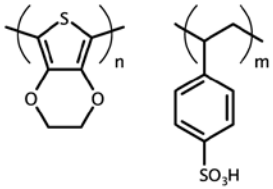
Wilbers et al., Scientific Reports DOI:10.1038/srep41171



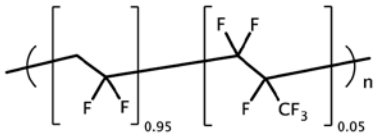
Thiburce et al., Nano Lett. 19, 1712, (2019)

Materials selection for increased resistance, range

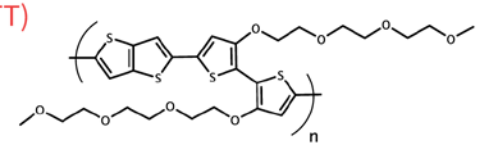
PEDOT:PSS



PVDF-HFP

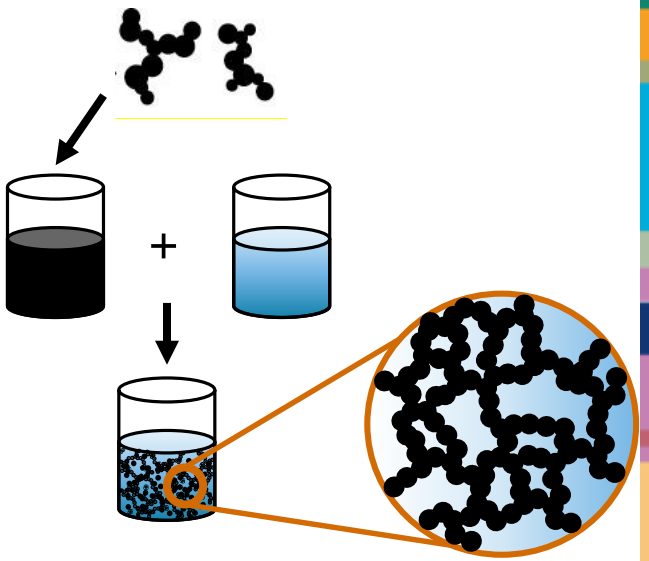


p(gT2-TT)

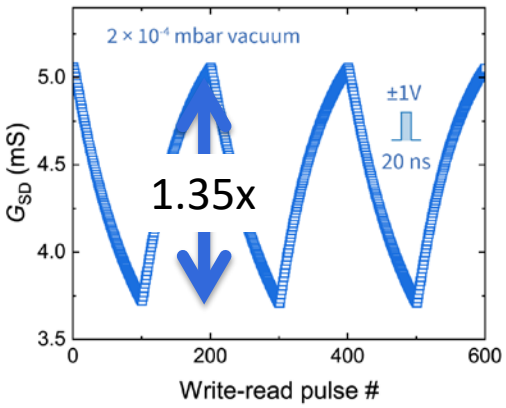


EIM:TFSI

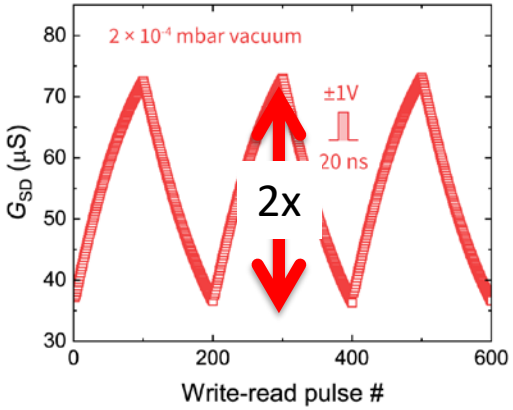
EMIM:TFSI

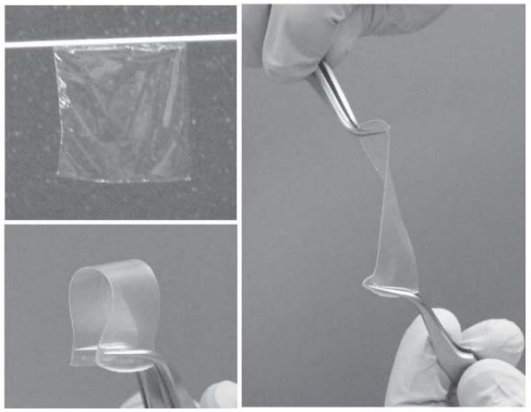


PEDOT:PSS w/ EIM:TFSI PVDF-HFP



p(gT2-TT) w/ EMIM:TFSI PVDF-HFP

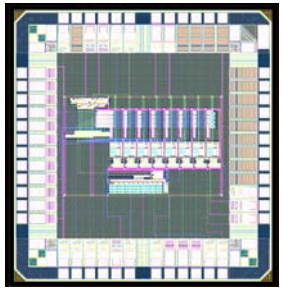




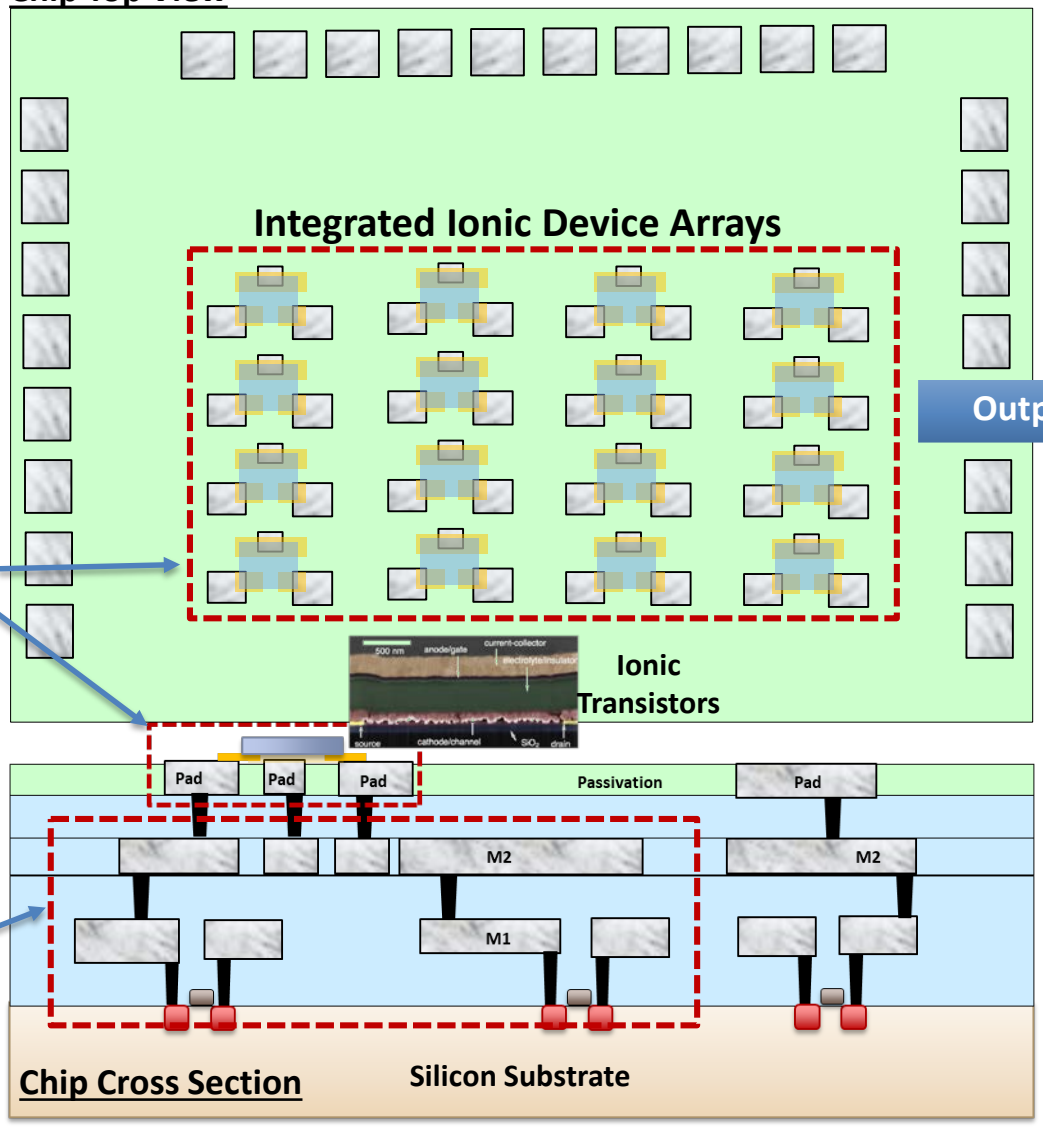
D. Frisbie et al.
Advanced Materials 24, 4457-4462 (2012)

CMOS-Integrated Ionic Transistors

Ionic Array Neural Characterization Chip

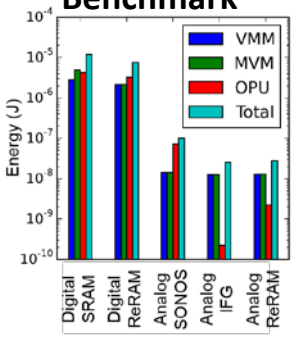


Chip Top View

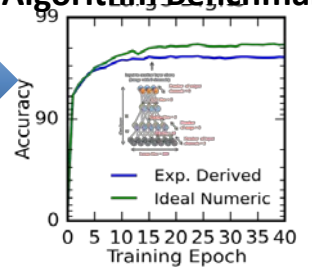


Outputs

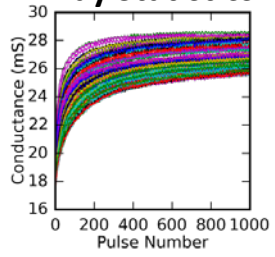
Energy & Performance Benchmark



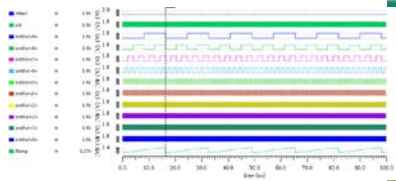
Algorithm Benchmark

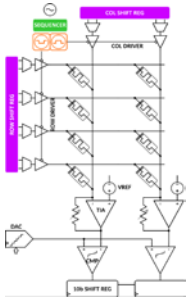


Array Statistics



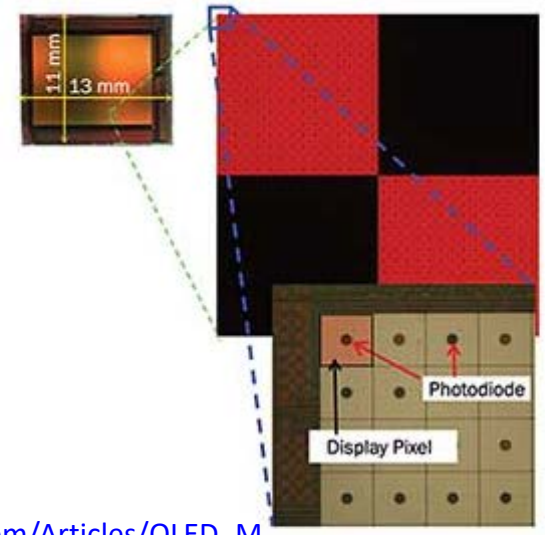
Ensemble Dynamics

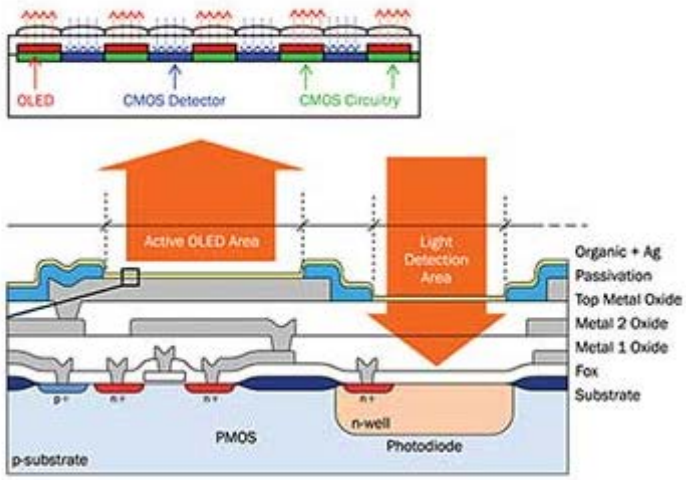




180nm CMOS Analog Neuro Circuits

Organic electronics technology rapidly advancing

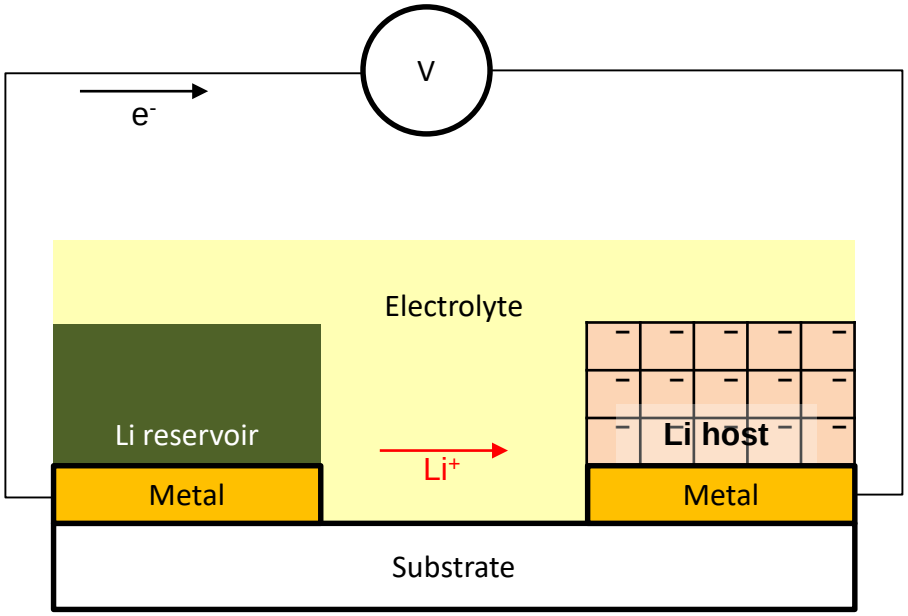


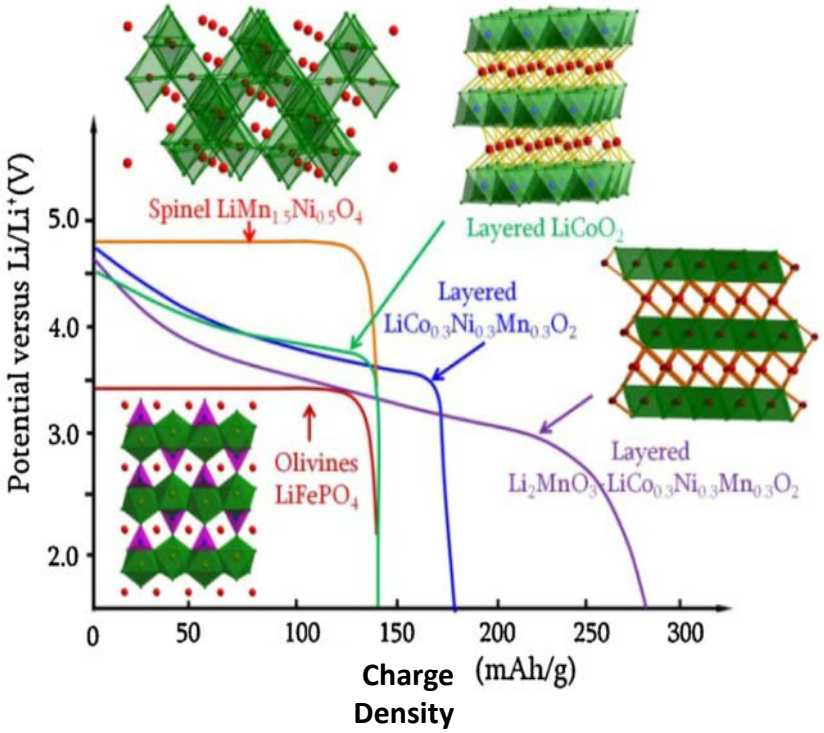


https://www.photonics.com/Articles/OLED_Microdisplays_Advancing_Virtual_and/a58438

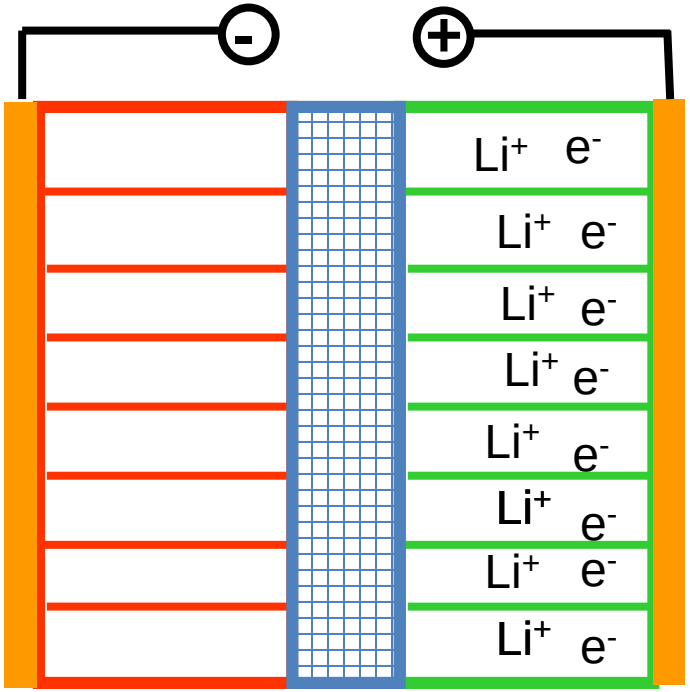
*General setup of a bidirectional OLED microdisplay (**left**). Courtesy of Fraunhofer FEP.*

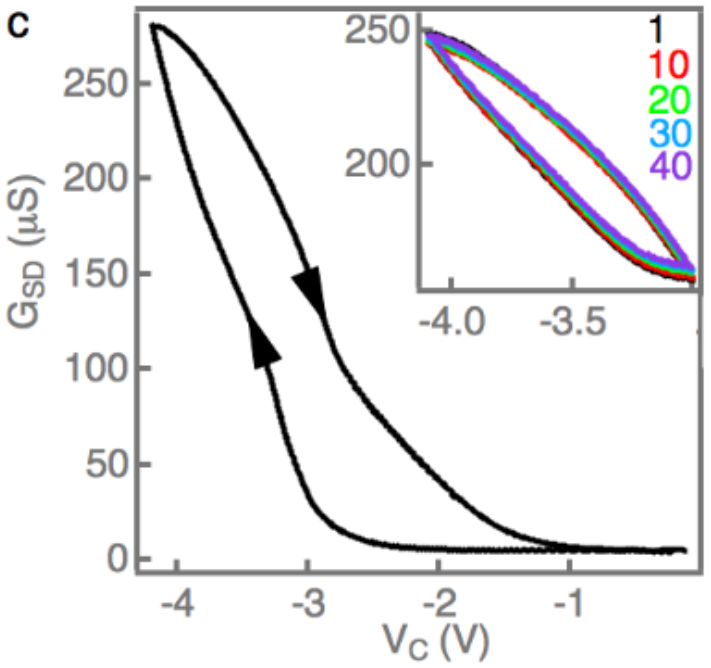
Ion-insertion electrodes for neuromorphic computing

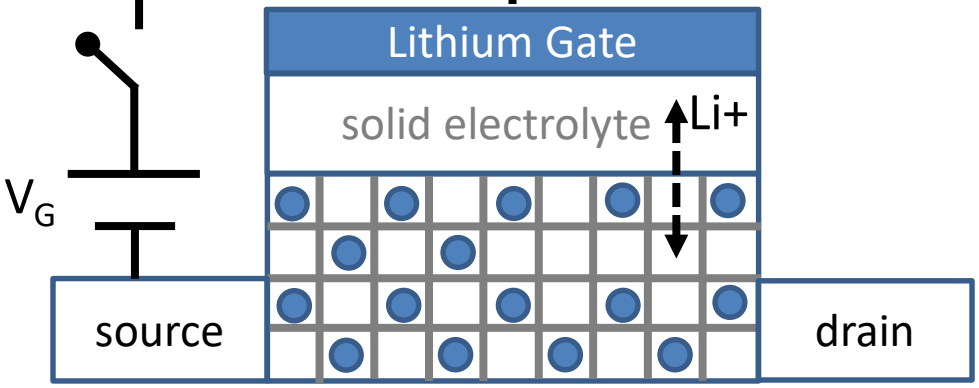


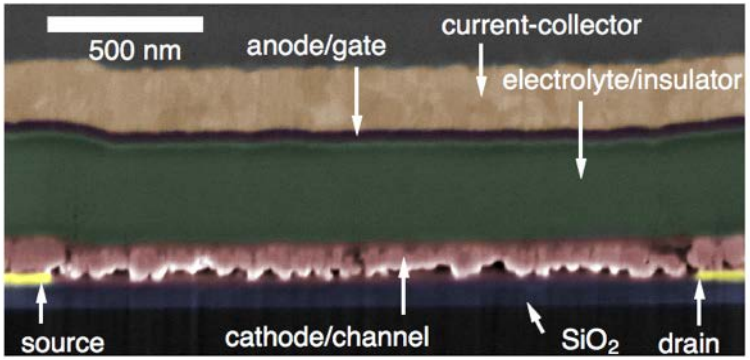


Store energy ↔ store information



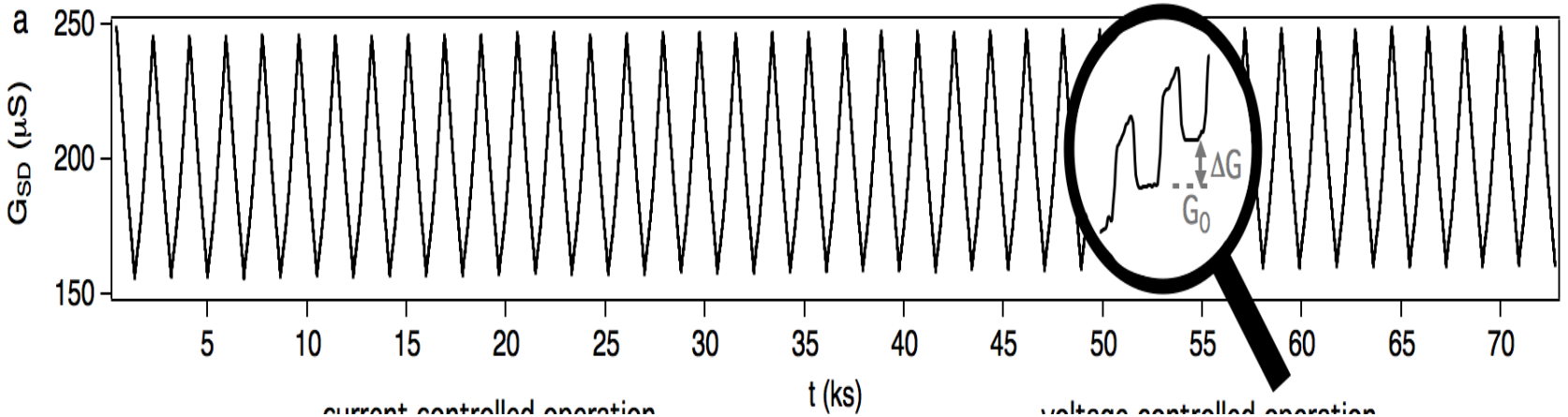


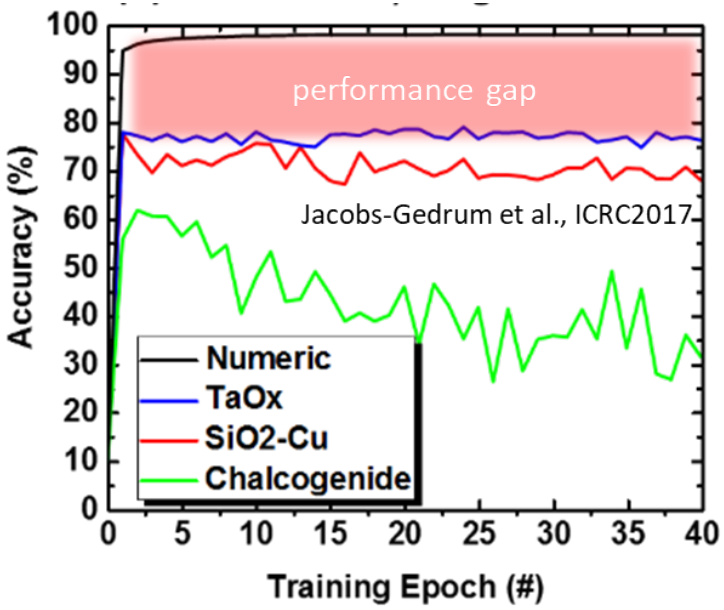


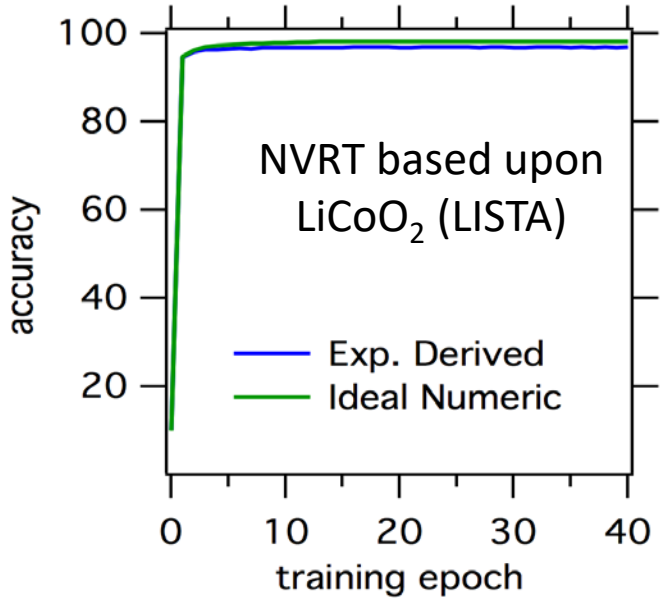


E. J. Fuller, F. El Gabaly, F. Léonard, S. Agarwal, S. J. Plimpton, R. B. Jacobs-Gedrim, C. D. James, M. J. Marinella, A. A. Talin, Adv. Mat., 29, 1604310, 2017

Highly predictable, stable operation, ~200 states

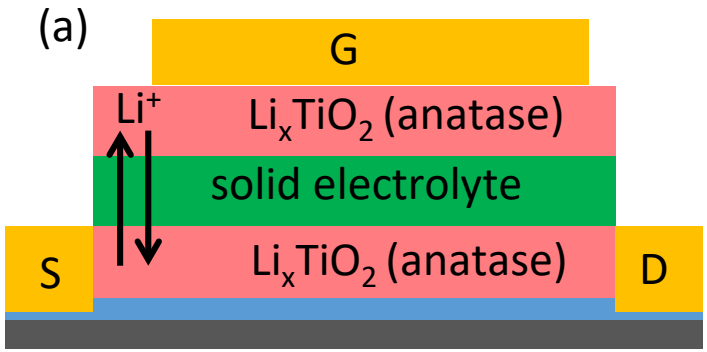


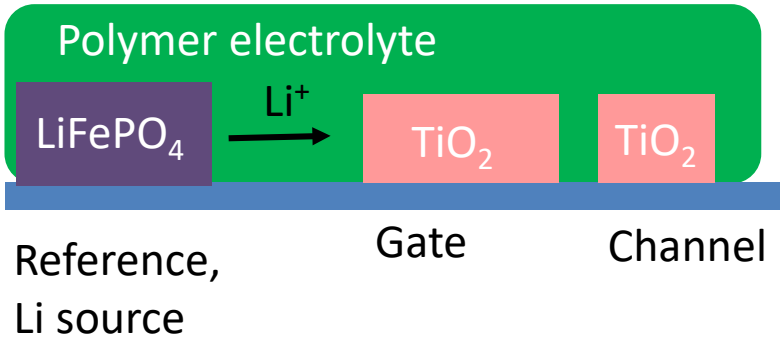


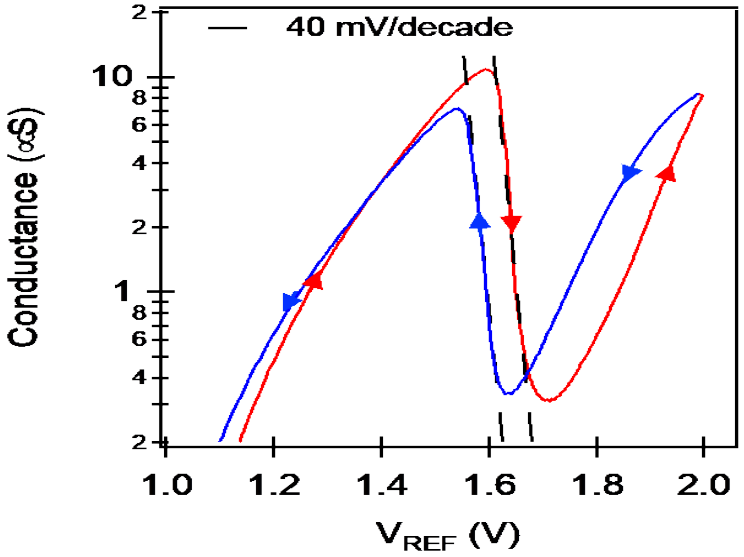


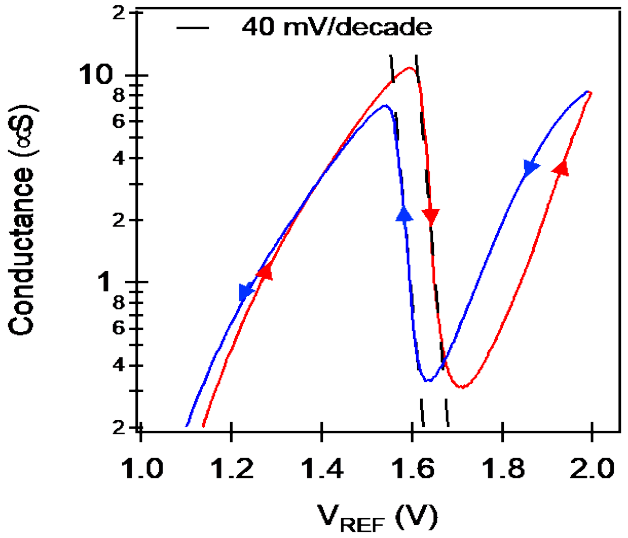
E. J. Fuller, F. El Gabaly, F. Léonard, S. Agarwal, S. J. Plimpton, R. B. Jacobs-Gedrim, C. D. James, M. J. Marinella, A. A. Talin, Adv. Mat., 29, 1604310, 2017

Li_xTiO_2-Li_xTiO_2 symmetric redox transistor





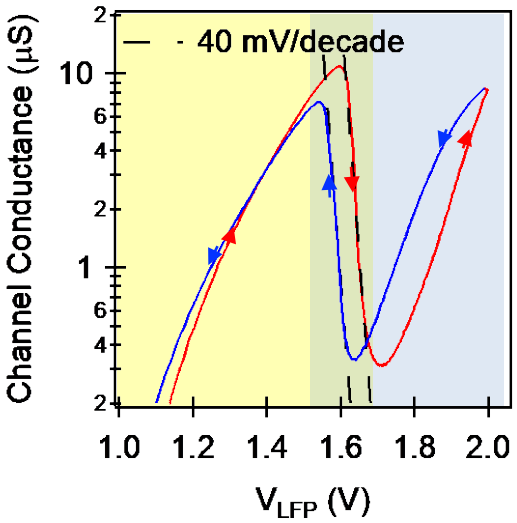


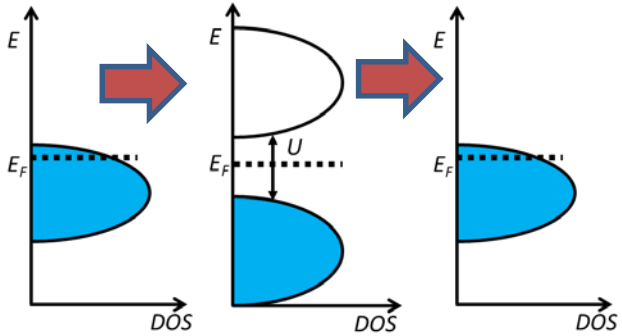


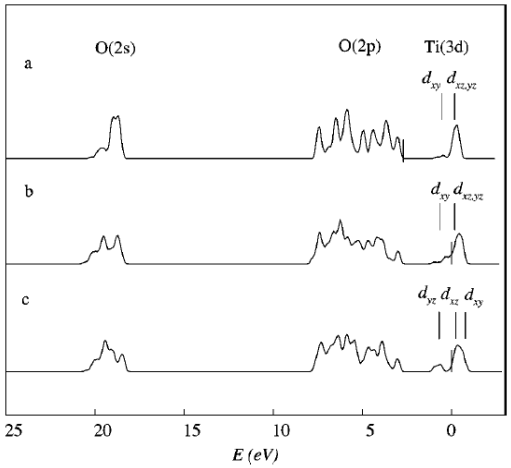
Y. Li, E. J. Fuller, S. Asapu, S. Agarwal, T. Kurita, J. J. Yang, A. A. Talin, ACS App Mat Int., 11, 38982, (2019)

Electronic structure <-> ion insertion relationship

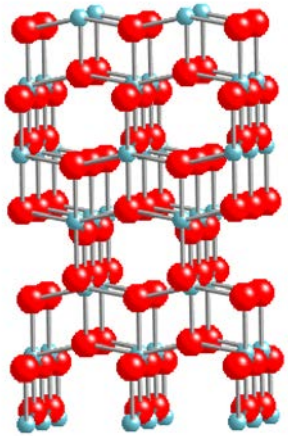
$$Li^{+} + e^{-} + Ti_{Ti}^{X} \rightarrow Li_{i}^{\cdot} + \boldsymbol{Ti'_{Ti}}$$



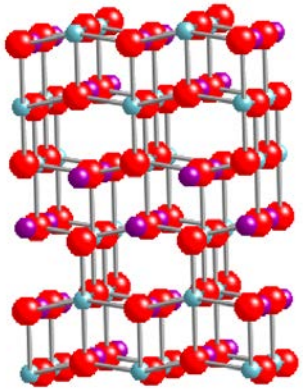




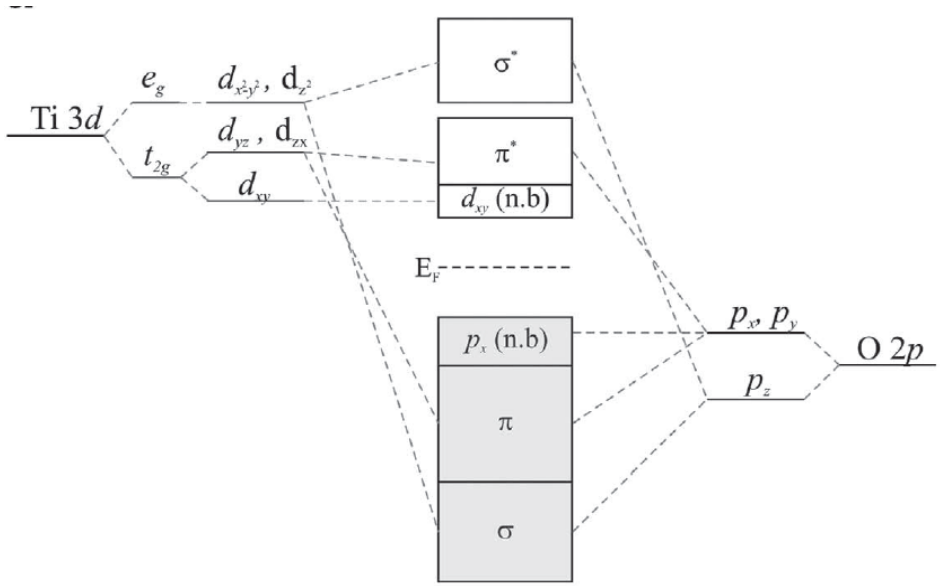
M. V. Koudriachova, S. W. de Leeuw, N. M. Harrison, Phys. Rev. B 69, 054106, 2004



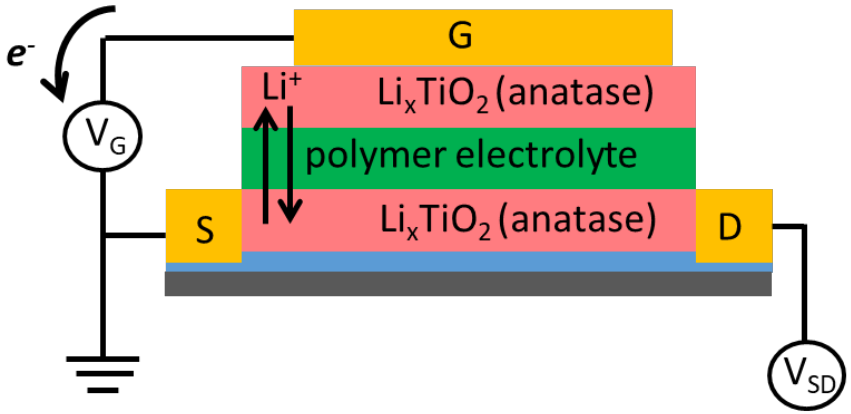
tetragonal, *a, b*=3.787Å and *c*=9.515



orthorombic, *a=3.819* *b*=4.084 Å and *c*=9.066

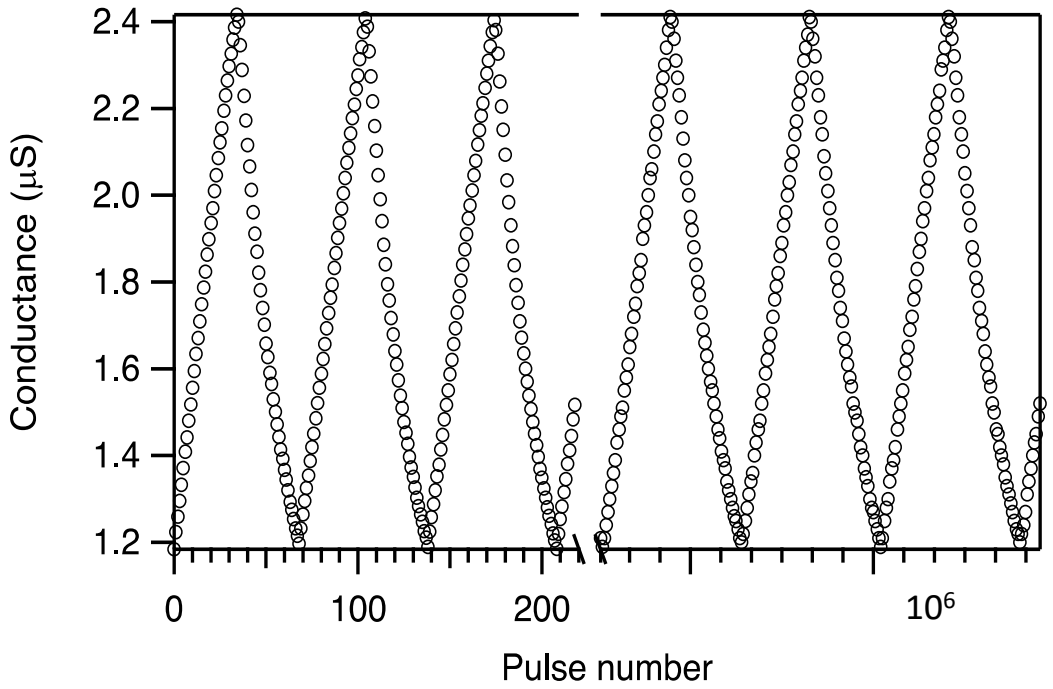


Li_xTiO_2-Li_xTiO_2 symmetric redox transistor



Gate: $Li_xTiO_2 \rightarrow Li_{x-\Delta x}TiO_2 + \Delta xLi$

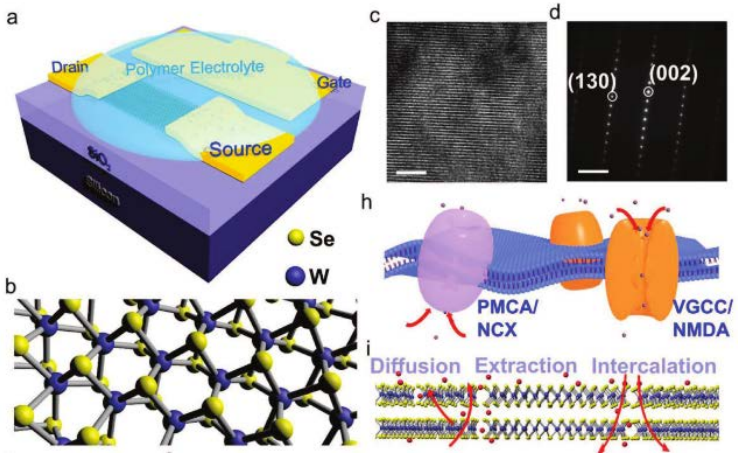
Channel: $Li_xTiO_2 + \Delta xLi \rightarrow Li_{x+\Delta x}TiO_2$



Y. Li, E. J. Fuller, S. Asapu, S. Agarwal, T. Kurita, J. J. Yang, A. A. Talin, ACS App Mat Int., DOI: 10.1021/acsami.9b14338

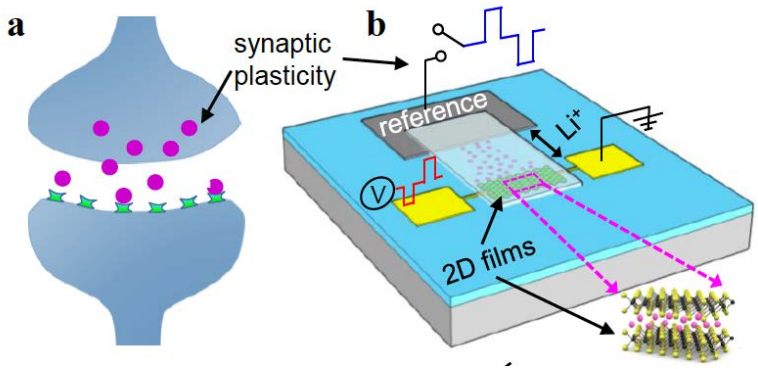
LISTA work is inspiring new 2D systems

LISTA - WSe_2



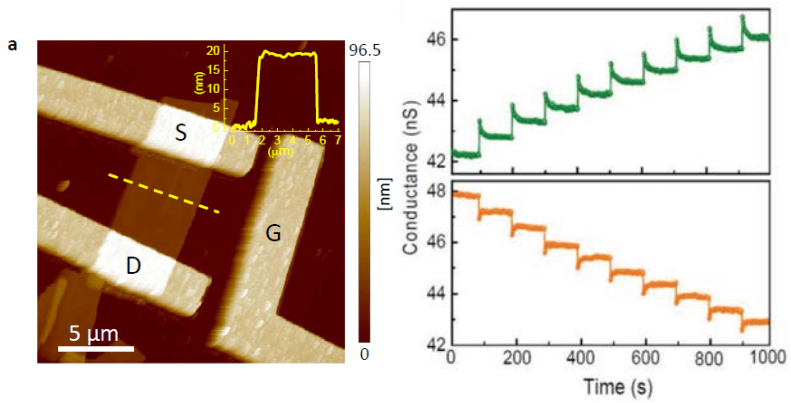
Zhu et al *Advanced Materials* 2018

LISTA - Graphene



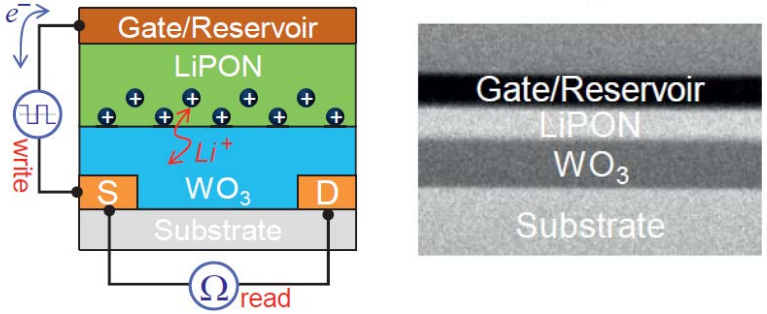
Sharbati et al *Advanced Materials* 30, 1802353, 2018

LISTA - MoO_x



C-S. Yang, D-S. Shang, N. Liu, E. J. Fuller, S. Agrawal, A. A. Talin, Y-Q. Li, B-G. Shen, Y. Sun, Adv. Funct. Mat. 1804170 2018

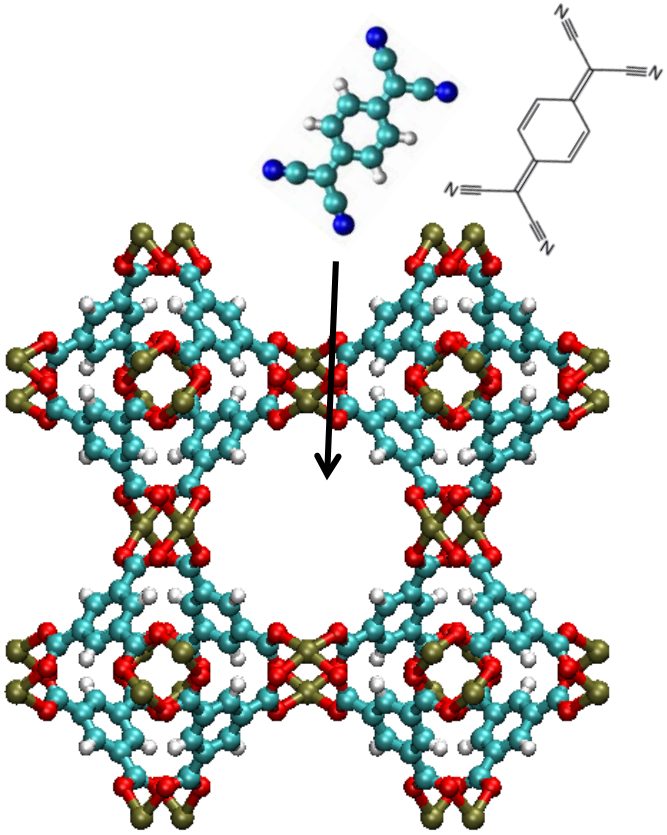
LISTA – Wo_x (IBM)

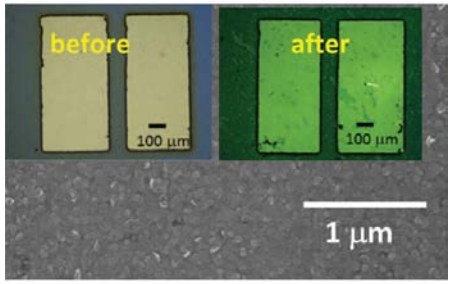


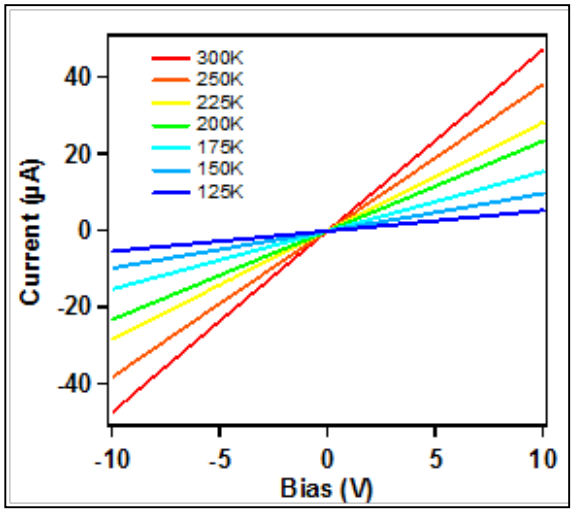
J. Tang, et al., IEDM 2018

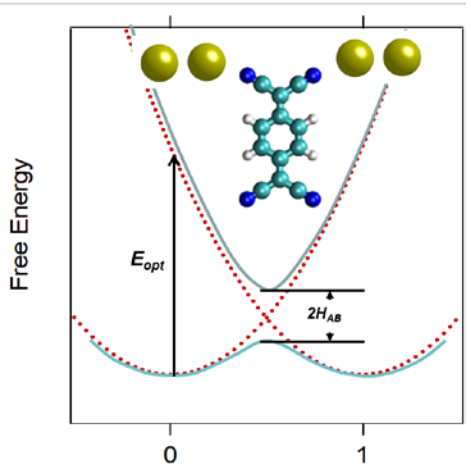
Metal organic frameworks (MOFs)

- *high density of redox states*
- *structural porosity for high ionic transport*
- *tailor electronic transport through metal-ligand coupling*



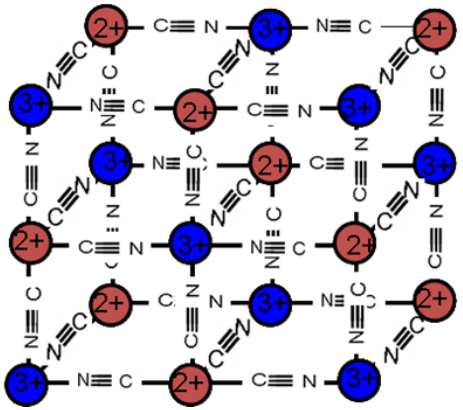


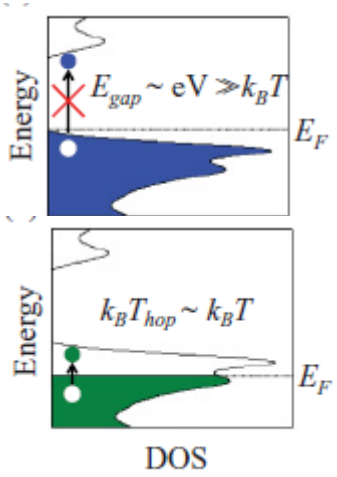


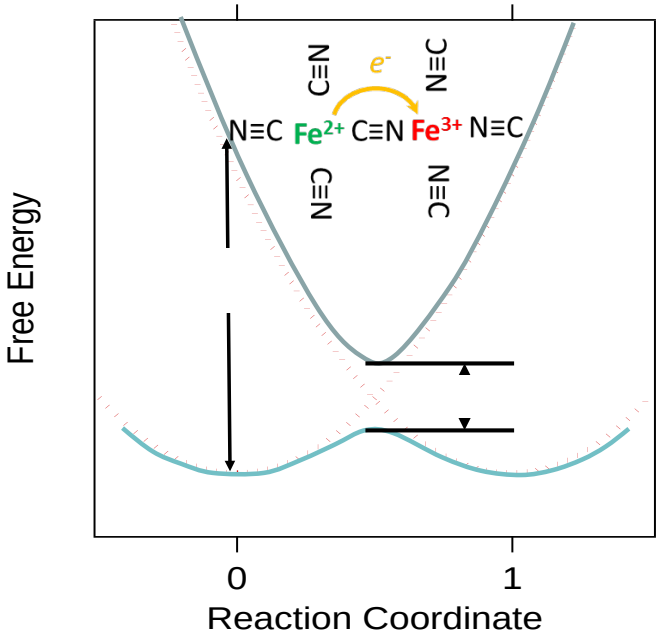


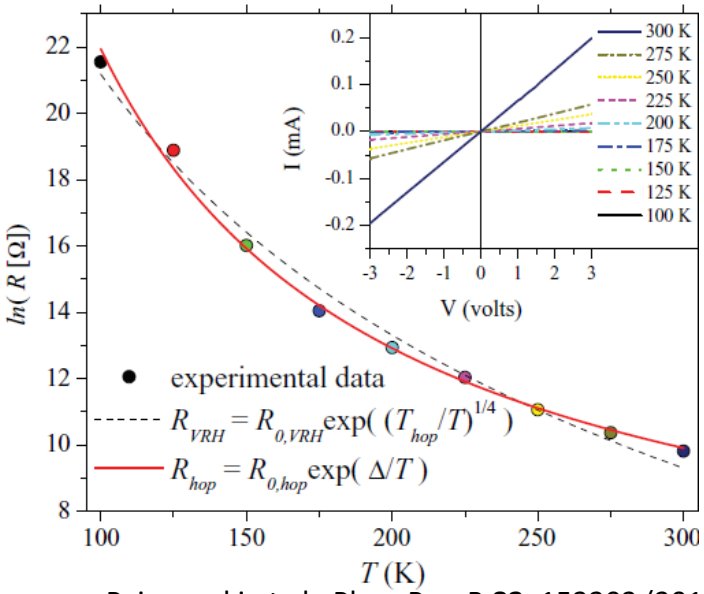
A. A. Talin, A. Centrone, A. Ford, M. E. Foster, V. Stavila, P. Haney, R. A. Kinney, V. Szalai, F. El Gabaly, H. P. Yoon, F. Léonard, M. D. Allendorf, Science 343, 66 (2014).

PB analogs display ultrafast H^+, tunable e^- transport









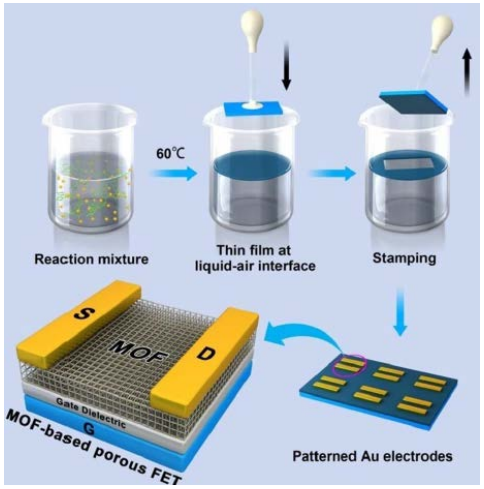
Pajerowski et al., Phys. Rev. B **83**, 153202 (2011)

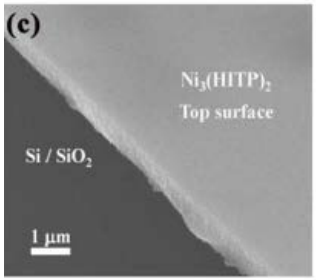
$$\Delta G^* = \frac{(\lambda - 2H_{AB})^2}{4\lambda} \qquad H_{AB} = \langle \Psi_A | H | \Psi_B \rangle$$

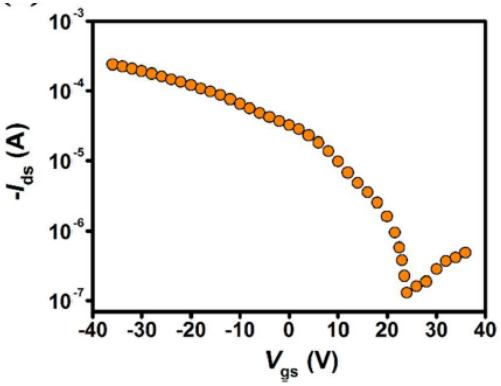
Class I: ~ weak/no coupling
Class II ~ moderate coupling
Class III ~ strong coupling

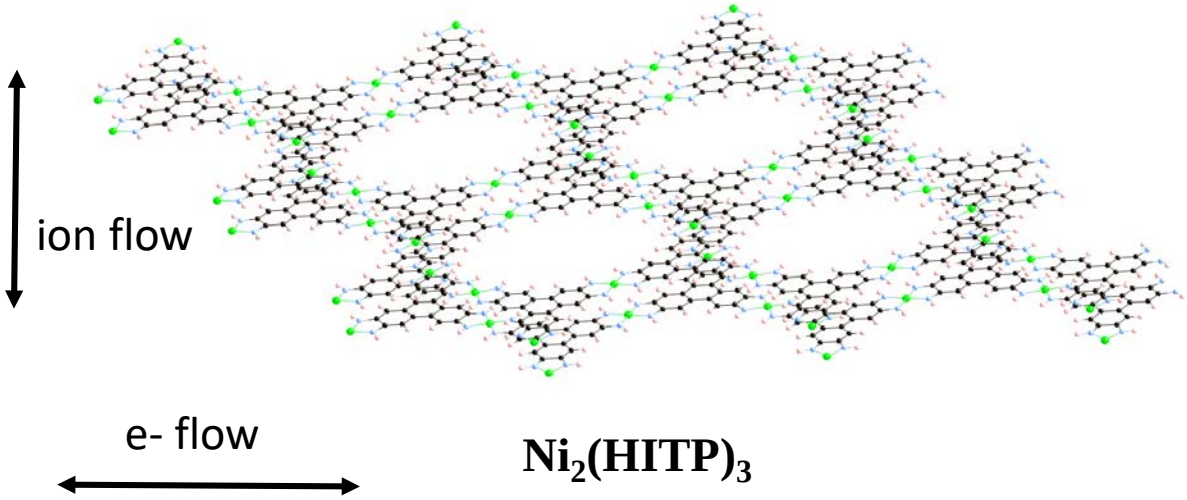
2D semiconducting MOFs

G. Wu, J. Huang, Y. Zang, J. He, G. Xu, JACS





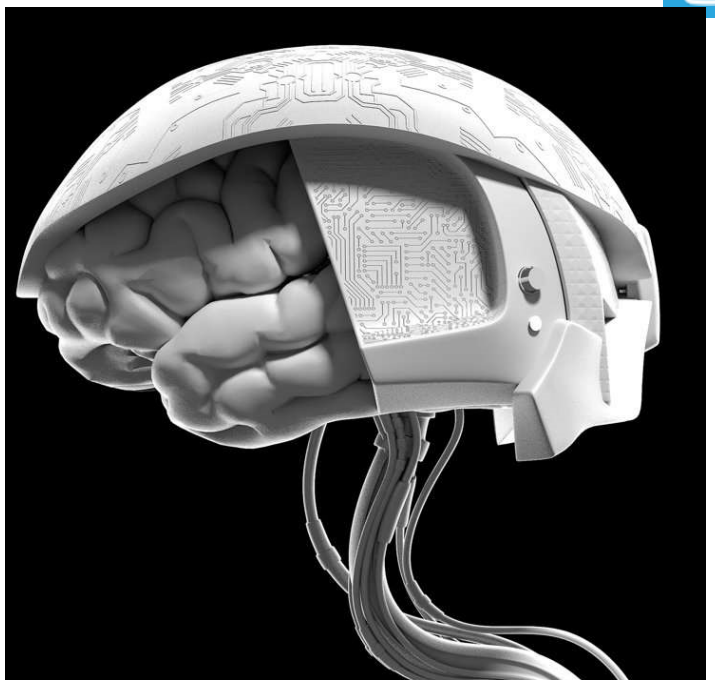


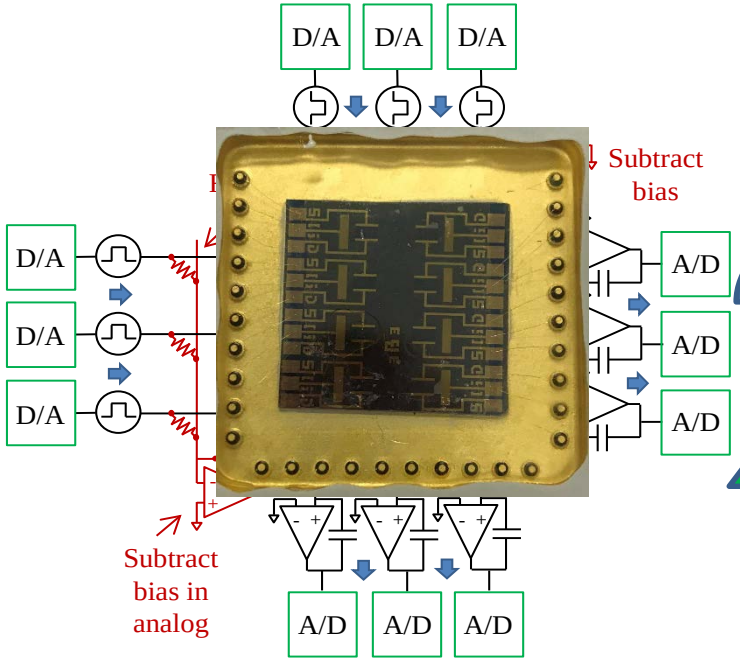


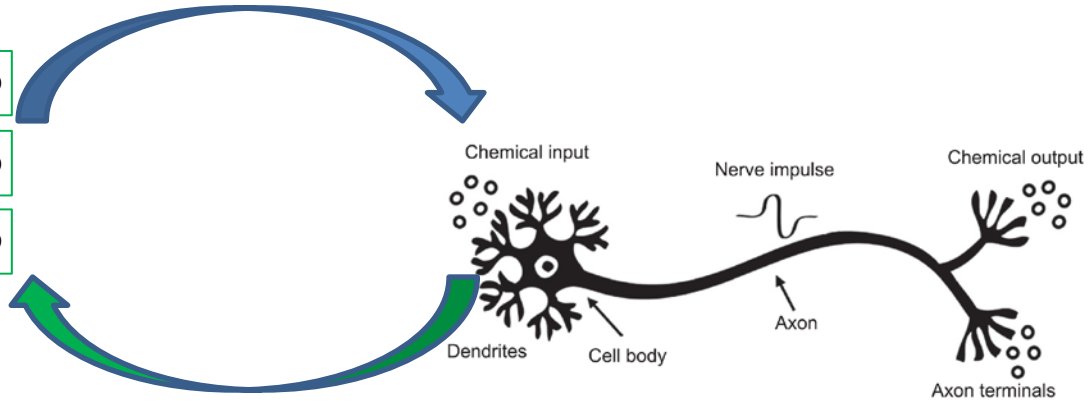
The Pentagon's Push to Program Soldiers' Brains

The mission is to make human beings something other than what we are, with powers beyond the ones we're born with.

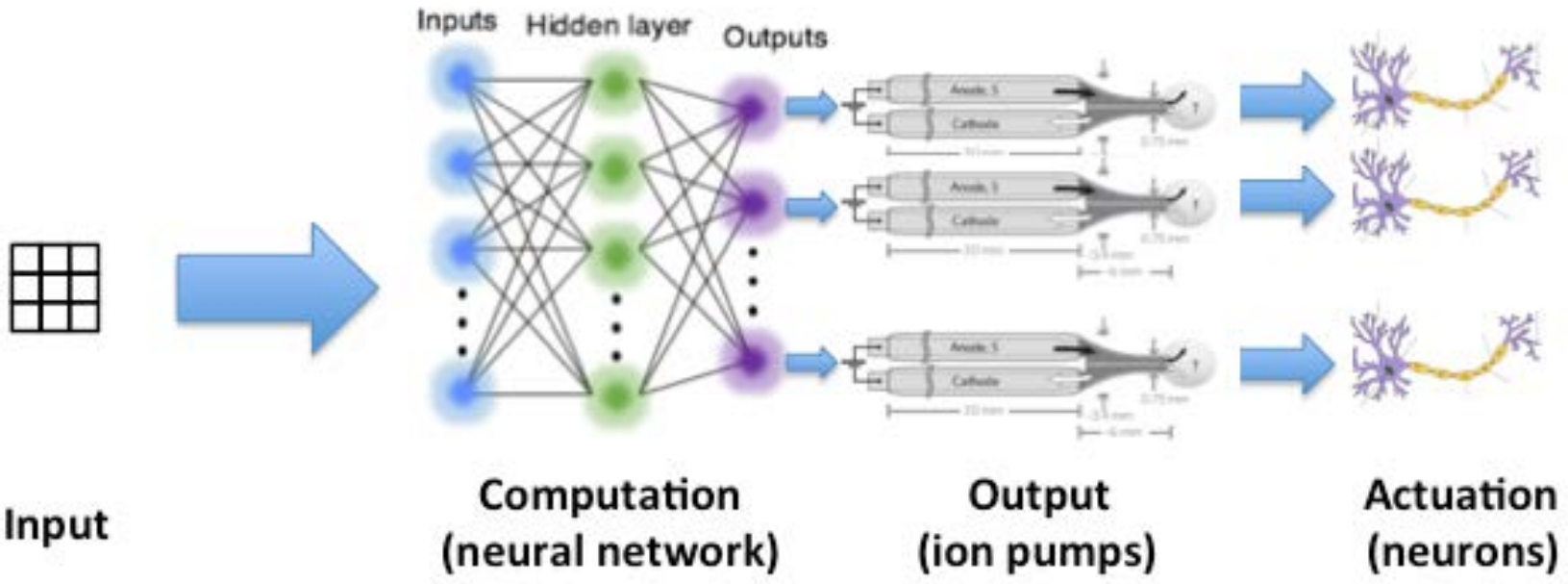
https://www.theatlantic.com/magazine/archive/2018/11/the-pentagon-wants-to-weaponize-the-brain-what-could-go-wrong/570841/



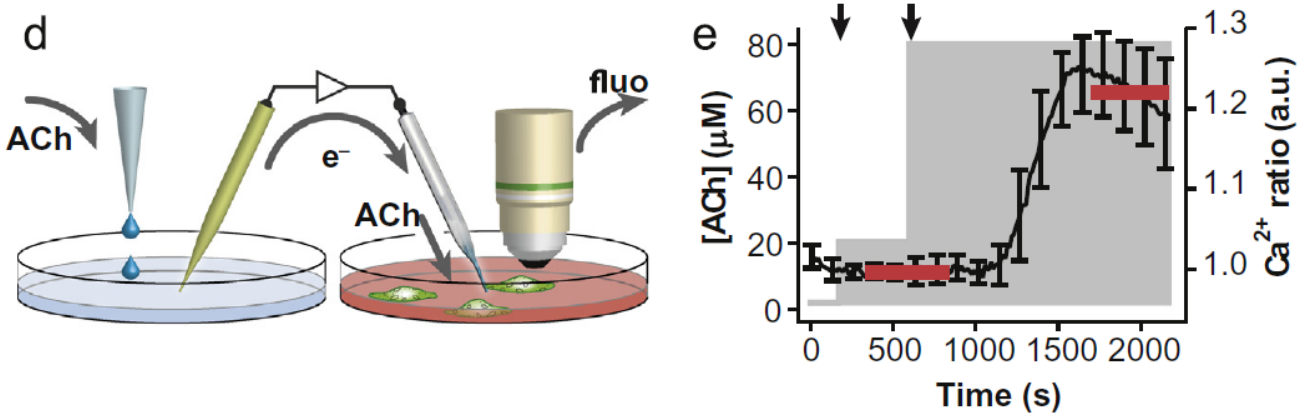




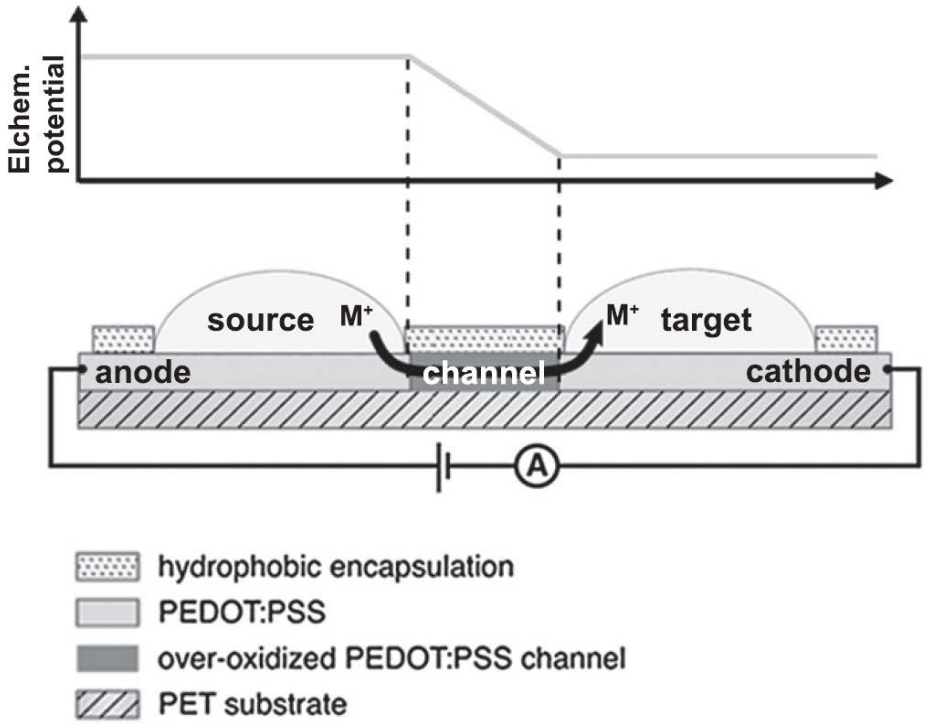
Polymer NVRM for brain-machine interface



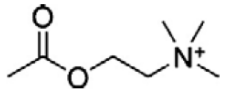
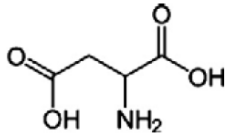
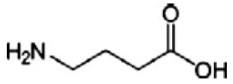
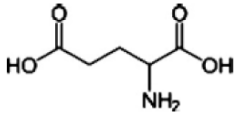
Simon et al., Biosensors andBioelectronics 71 359 (2015)



PEDOT:PSS ionic-electronic neurotransmitter pumps



K. C. Larsson, P. Kjäll, A. Richter-Dahlfors, Biochimica et Biophysica Acta 1830 (2013) 4334

Ions			
Na^+	Involved in the initiation of action potentials.		80%
K^+	Maintain the resting potential in excitable cells.		100%
Ca^{2+}	Important second messenger in cell signaling, dictates release of neurotransmitters.		100%
Neurotransmitters			
Acetylcholine	Neurotransmitter in the CNS and PNS.		100%
Aspartate	Excitatory neurotransmitter in the CNS.		16%
GABA	Major inhibitory neurotransmitter in CNS.		77%
Glutamate	Major excitatory neurotransmitter in the CNS.		37%

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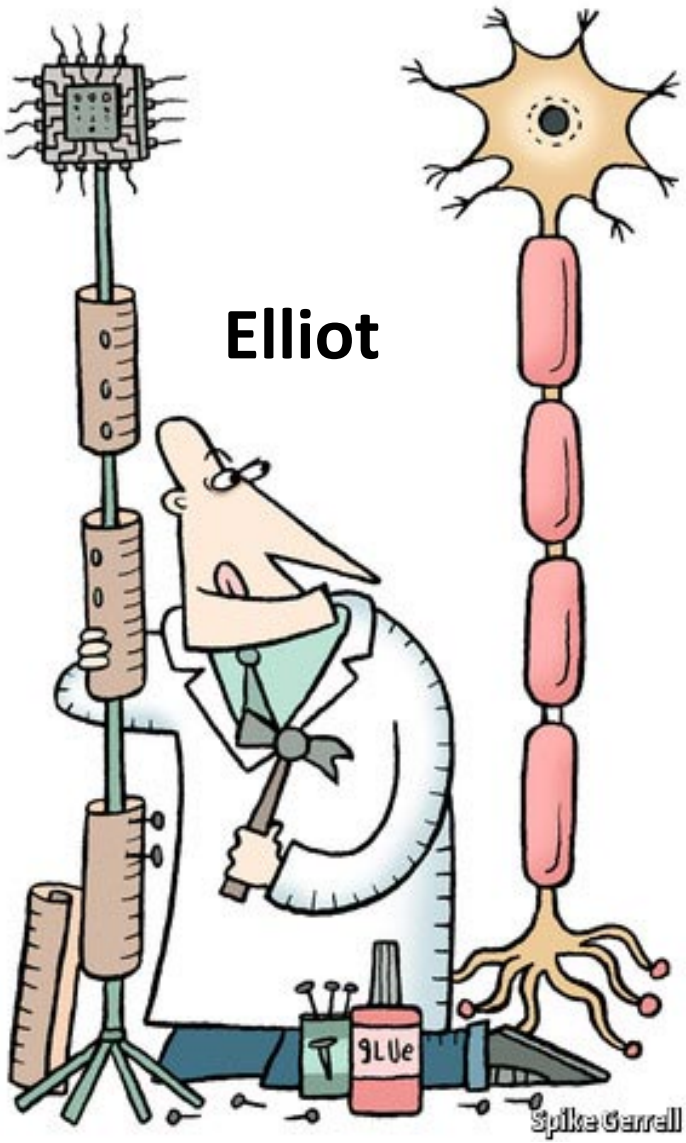
U.S. DEPARTMENT OF ENERGY | Office of Science

NEES NANOSTRUCTURES for ELECTRICAL ENERGY STORAGE


Elliot
GLUE
Spike Gerrell

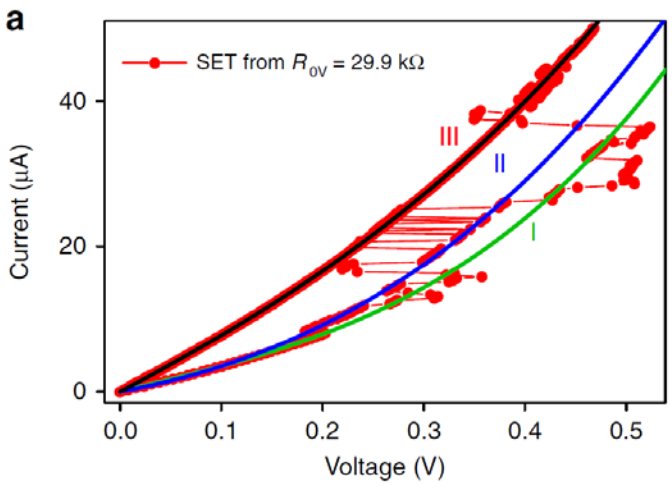
Operating in conductance limit increases noise

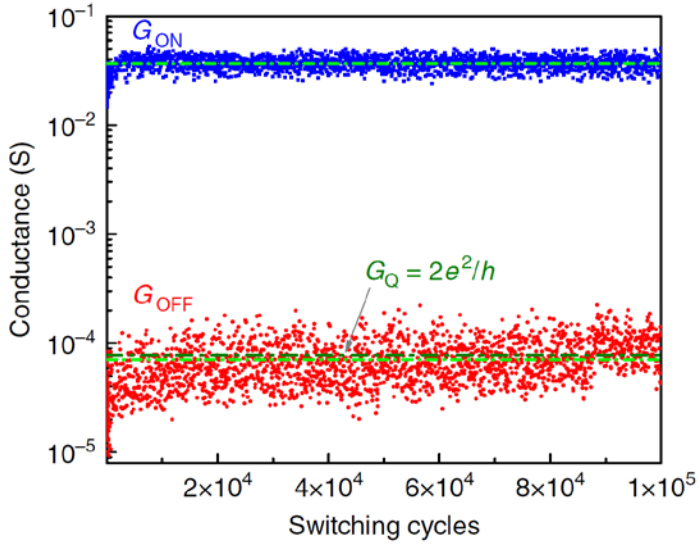
Received 25 Aug 2015 | Accepted 25 Feb 2016 | Published 4 Apr 2016

DOI: 10.1038/ncomms11142 OPEN

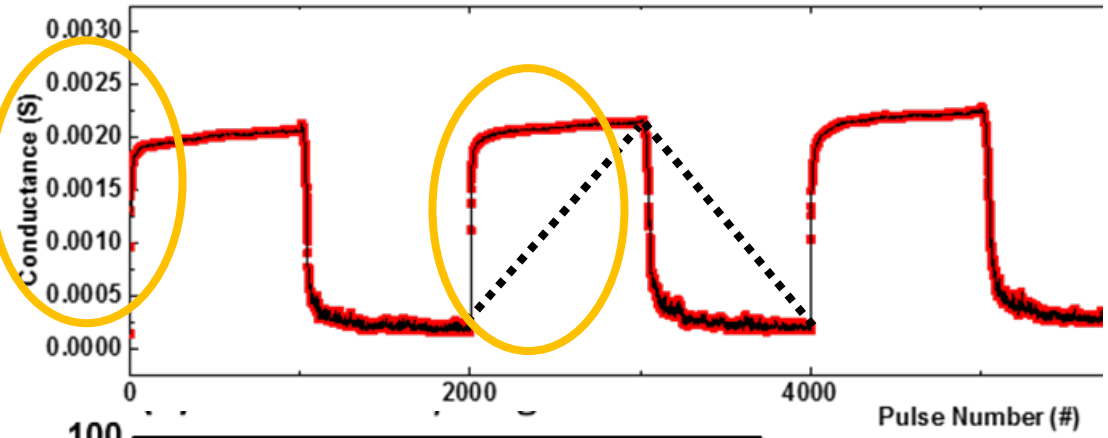
Quantized conductance coincides with state instability and excess noise in tantalum oxide memristors

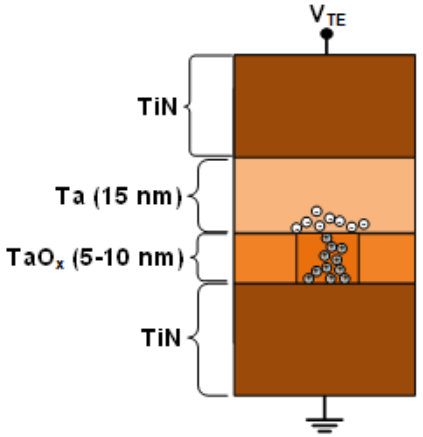
Wei Yi[1,2], Sergey E. Savel'ev[3], Gilberto Medeiros-Ribeiro[1,4], Feng Miao[1,5], M.-X. Zhang[1], J. Joshua Yang[1,6], Alexander M. Bratkovsky[1,7,8] & R. Stanley Williams[1]

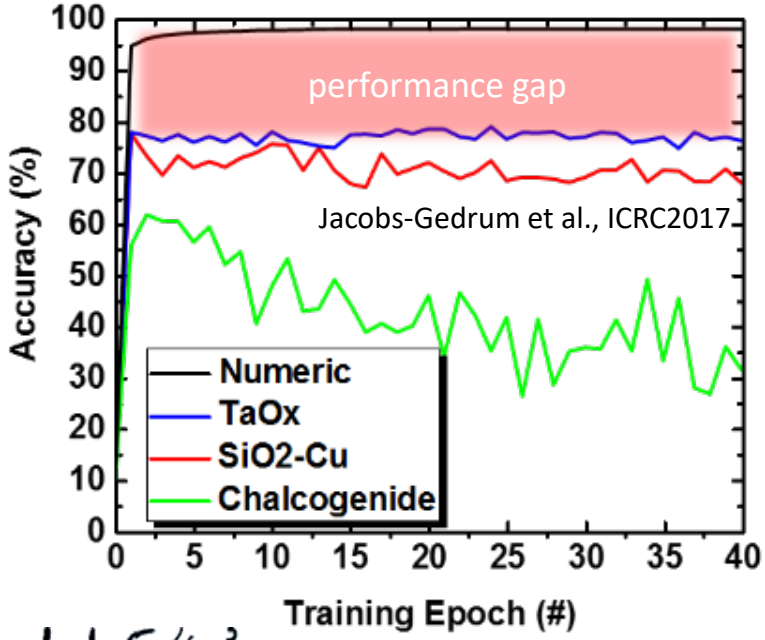


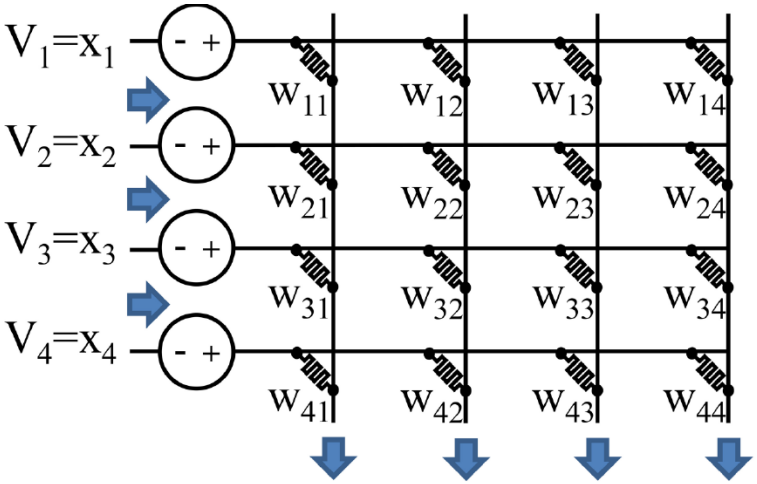


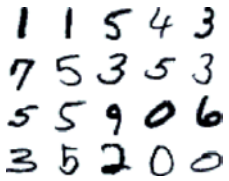
Non-linearity, high conductance limit accuracy, scaling







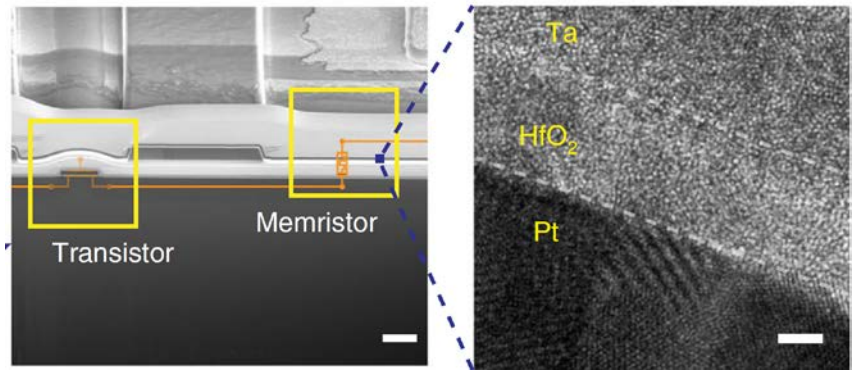


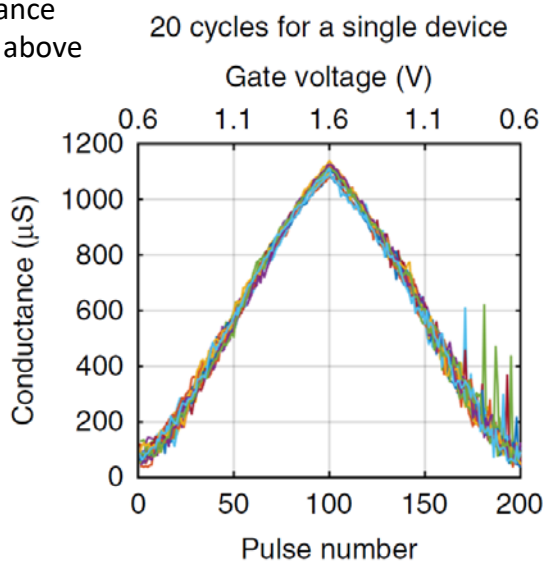


Data set	# Training Examples	# Test Examples	Network Size
MNIST Large Digits	60,000	10,000	784×300×10

Linearity BUT conductance high, needs reset

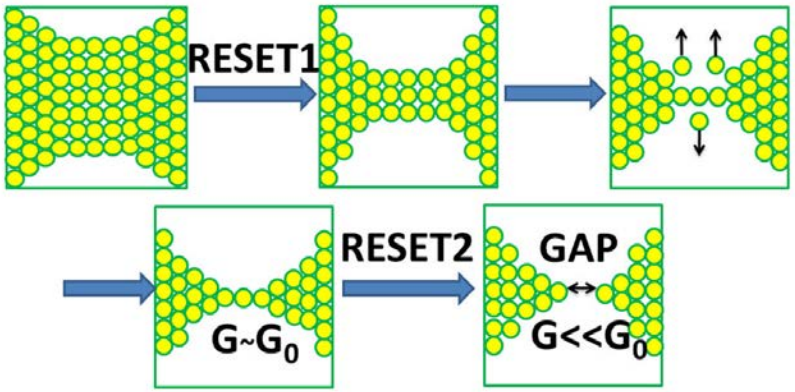
- Increase conductance → synchronized V+ to memristor, FET gate; set I compliance
- Decrease conductance→ reinitialize memristor, then increase conductance as above





100 μS→
0.1mA @1V!

Li et al., Nature Comm. 9, 2385, (2018)



$$G_o = {}^{2e^2}/_{h} = 13k\Omega$$

S. Long, X. Lian, C. Cagli, X. Cartoixa, R. Rurali, E. Miranda, D. Jimenez, L.Perniola, M. Liu, J. Sune, APL 102, 183505 (2013)