Clustering & Global Challenges
April 07-09, 2021
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Health
Smart Energy
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Advanced Materials

Magneto-ionics for energy efficiency: challenges and opportunities

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CNM Centre Nacional de Microelectrònica IMB

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OUTLINE

1. Introduction

- The problem of heat dissipation in magnetic actuation
- Converse magneto-electric actuation: mechanisms
- Magneto-ionics

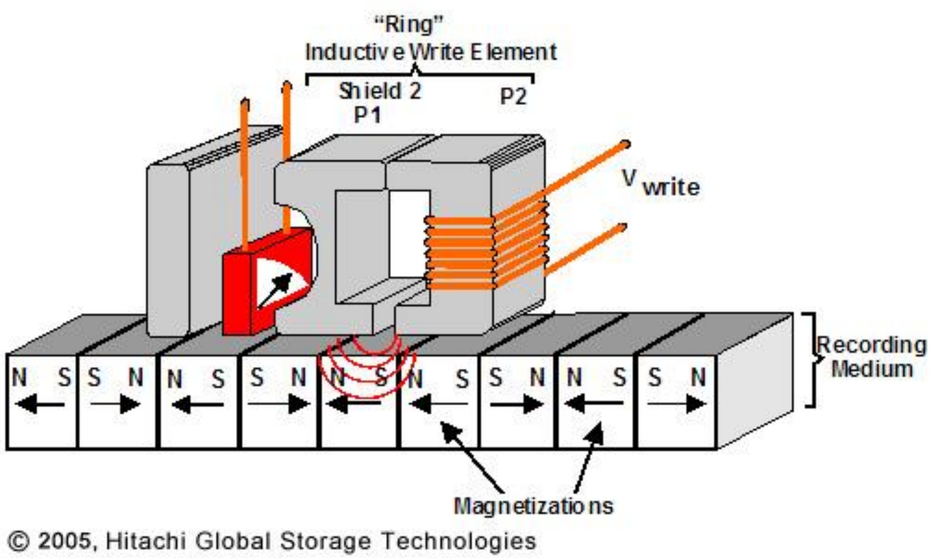
2. Results and discussion

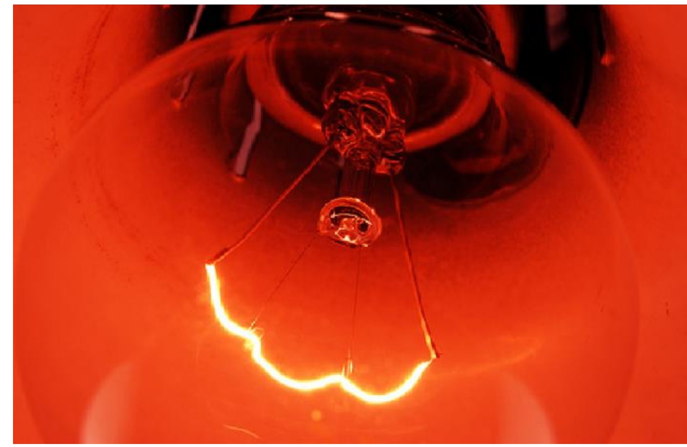
- Magneto-ionic effects in Co_3O_4 films
  - ✓ Morphology and structural characterization
  - ✓ Magnetic properties *vs.* applied voltage: ON-OFF magnetism!
  - ✓ Effect of the electric field configuration
- Extrapolation to other materials: N-magneto-ionics

3. Conclusions

1. INTRODUCTION

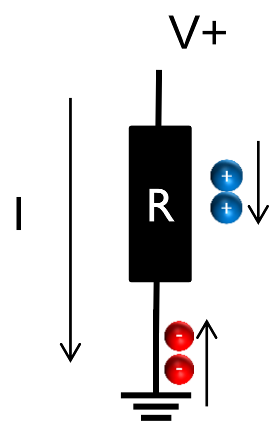
1. INTRODUCTION: POWER DISSIPATION IN MAGNETIC RECORDING





Joule Effect

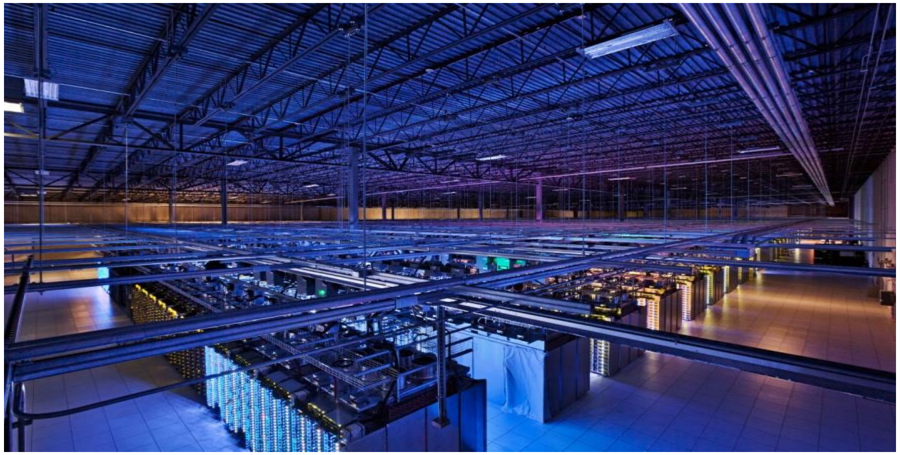
- Almost **40%** of electric power can be **wasted due to heat dissipation!**



$$V=IR$$

$$P=IV=I^2R$$

1. INTRODUCTION: POWER DISSIPATION IN MAGNETIC RECORDING



Technology | Thu Oct 27, 2011 6:30am EDT

Related: TECH, FACEBOOK

Facebook likes Sweden for first Europe server site

STOCKHOLM

Social networking site Facebook is to build its first data center outside the United States in the northern Swedish town of Lulea, awarding an initial construction contract of $121 million, the companies said on Thursday.

The data center, set to be the largest of its kind in Europe, will take advantage of the climate in Lulea, among the coldest in Sweden, to cool tens of thousands of servers.

"Those servers basically are what allow us to support all of the Facebook products for our users. Friend requests, tags, user updates will be accessed through this facility," Tom Furlong, Facebook's director of site operations, told Reuters.

"It will mostly serve European users and ideally improve performance for them," he added in a telephone interview.

PHOTOS OF THE DAY

Our top photos from the last 24 hours. Slideshow »

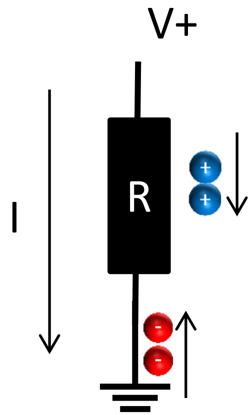
Trump rally mayhem

A refugee childhood

Microsofts is throwing data centers underwater

What can be done to avoid power dissipation?

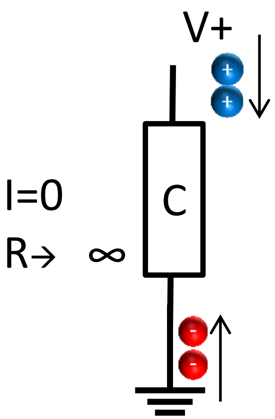
- Try to replace current by DC voltage.
- Move from resististive to capacitative systems.



$$V=IR$$

$$P=IV=I^2R$$

Resistive System



$$P=IV=V^2/R$$

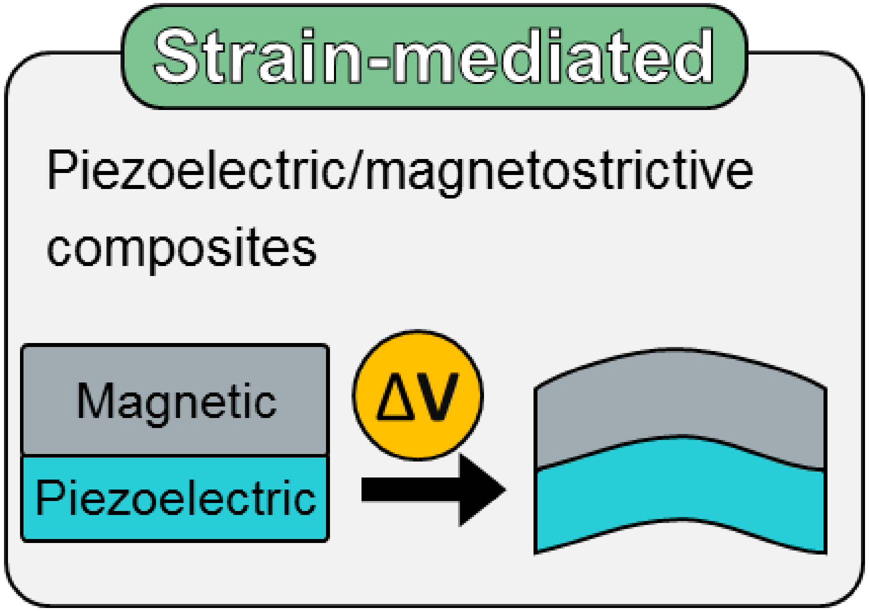
$$P=0$$

Capacitive system

- But… is it posible to **control magnetism with voltage**?

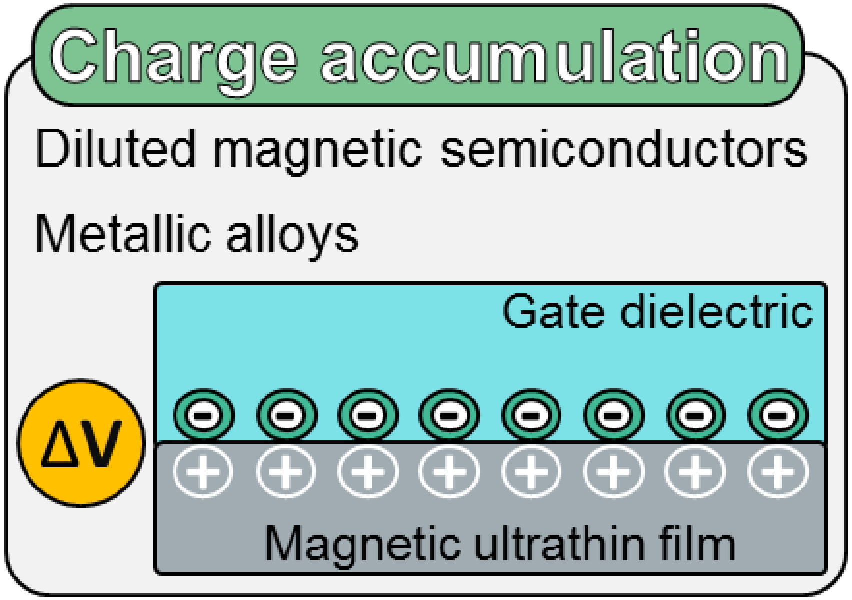
➢ Conventional voltage-driven magnetic actuation approaches

- Strain mediated piezoelectric + magnetostrictive composites (clamping effects, need of epitaxy, fatigue-induced failure)
- Single phase multiferroics (basically at low T)



- **Magnetism mediated by electrostatic charges (voltage-driven)**

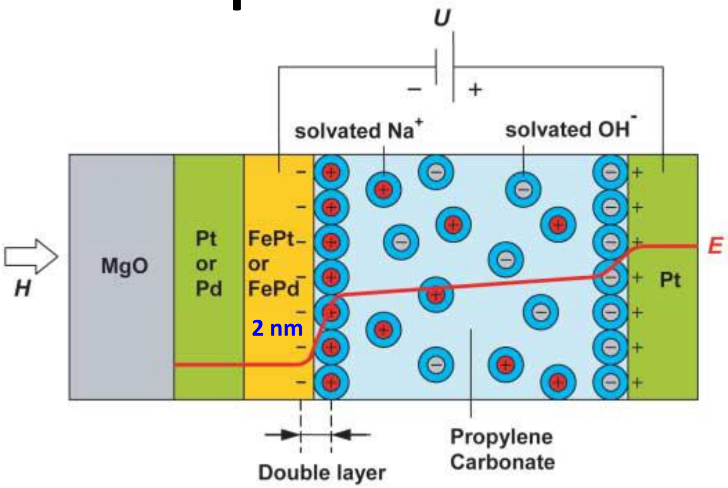
- Typically proven in:
  - i) Low-T magnetic semiconductors
  - ii) **Metallic** **ultra-thin films** and nanoparticles (electric field only at the surface!)

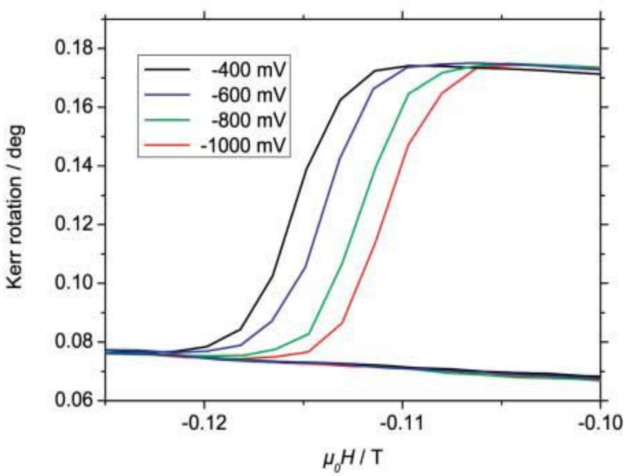


In metals λ_{TF} = 0.5 nm! Effects within the exchange length (l_{ex}=20 nm)

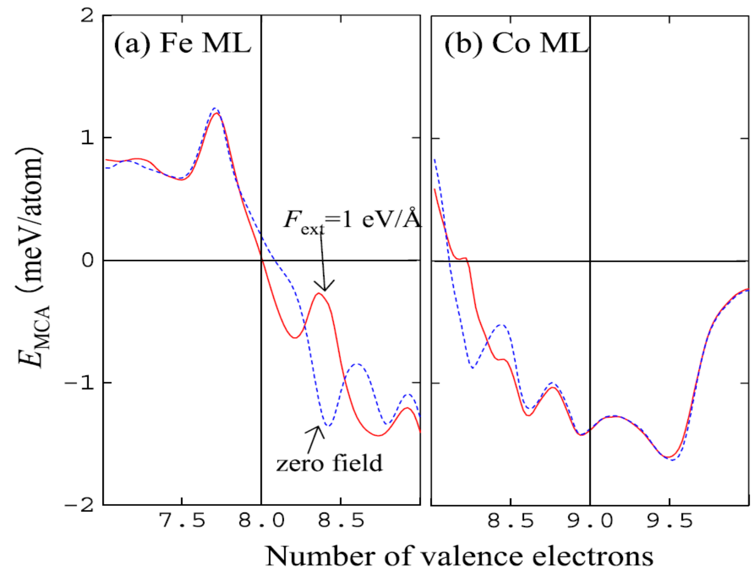
1. INTRODUCTION: MAGNETO-ELECTRIC ACTUATION

➢ Some previous works in ultra-thin films (electrostatic charging)





M. Weisheit, Science 315 (2007) 349.



K. Nakamuira, *Phys. Rev. Lett.* 102 (2009) 187201.

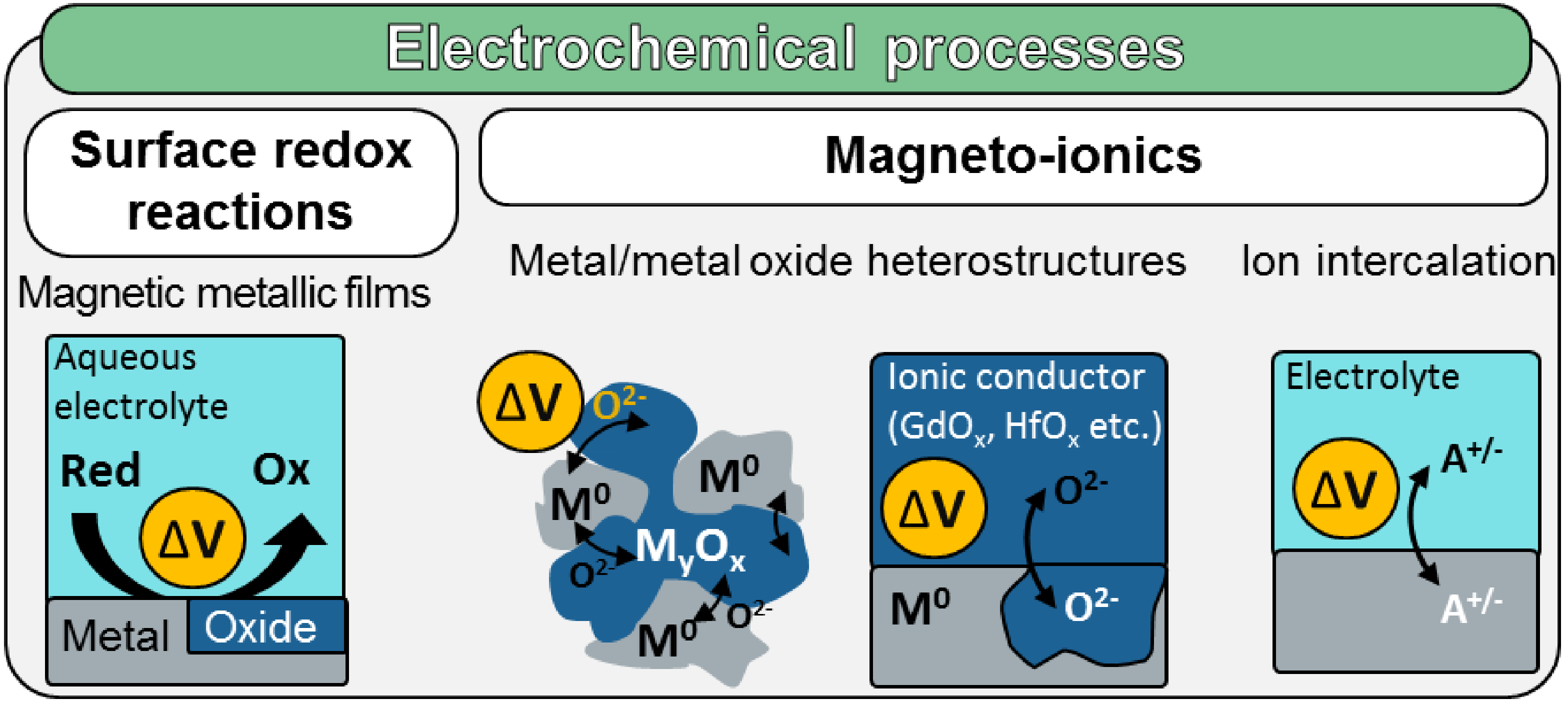
➢ Voltage induces changes in the **electronic band structure**

➢ Coercitivity **lowers by 4,5%**!

➢ Magnetism mediated by voltage-driven ion migration (magneto-ionics)

Nat. Mater. 14 (2015) 174
ACS Nano 12 (2018) 10291
Adv. Funct. Mater. 30 (2020) 2003704
Nat. Commun. 11 (2020) 5871
APL Mater. 7 (2019) 030701

- ❖ Strongest non-volatile effects (permanent changes)
- ❖ Usually slow at room temperature (ionic conductivity is thermally activated)



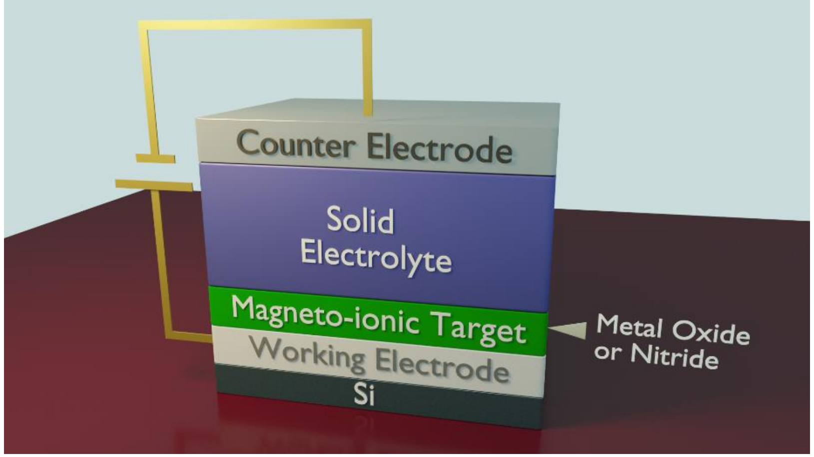
1. INTRODUCTION: MAGNETO-ELECTRIC ACTUATION

Magneto-ionics = modulation of oxidation state by voltage-induced ion motion (**O**, Li, H, F and N ions)

System = Layered heterostructure containing a magneto-ionic target

Ways to apply voltage (DV):

Solid electrolytes
all-solid systems: solid heterostructure/solid



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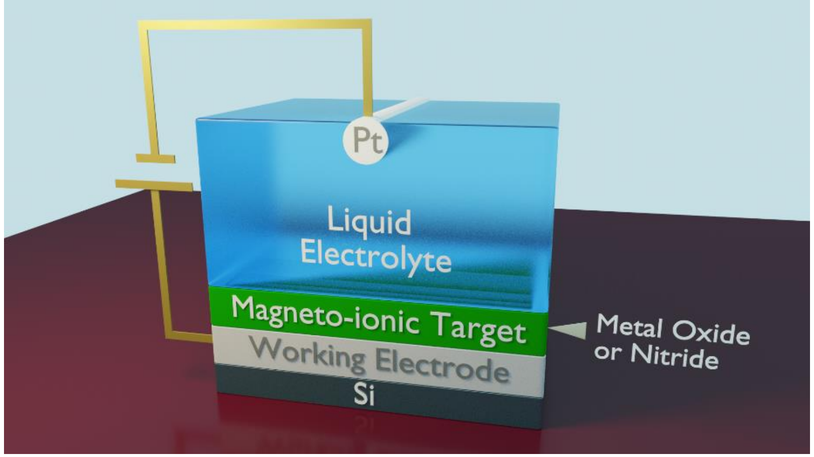
Liquid electrolytes
solid/liquid systems: solid heterostructure/liquid



*Adv. Funct. Mater.***27** (2017) 1701904
ACS Nano **12** (2018) 10291
APL Mater. **7** (2019) 030701

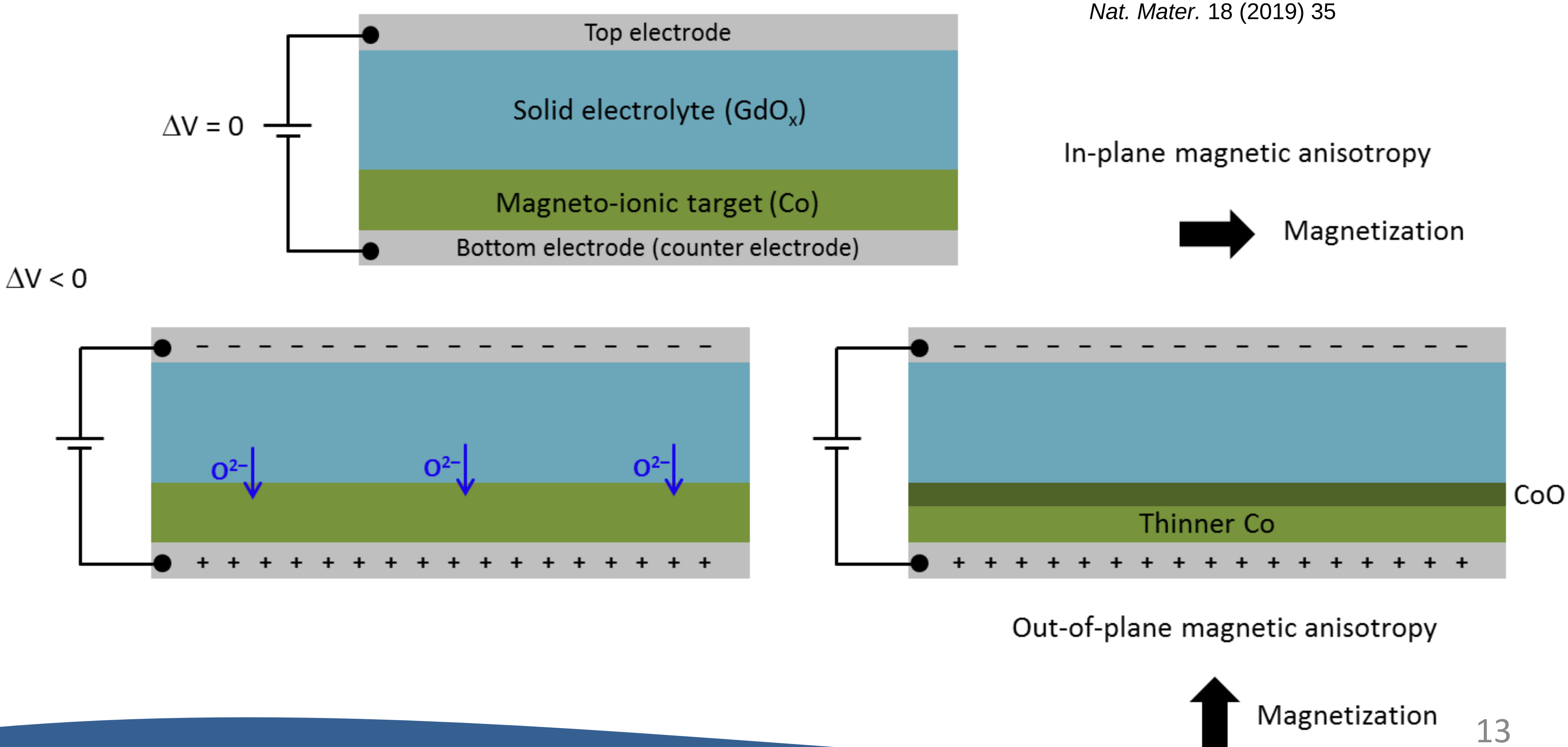
Electrolyte = ion reservoir to donate/accept (O, Li, H, F and N) ions depending on voltage polarity

1. INTRODUCTION: MAGNETO-ELECTRIC ACTUATION

Oxygen magneto-ionics using all-solid layer heterostructures: thin ferromagnetic layers (Co, Fe…) grown adjacent to solid electrolytes (GdO_x, HfO_2…)

Nat. Mater. 14 (2015) 174
Phys. Rev. B 98 (2018) 214441
Nat. Mater. 18 (2019) 35

Condenser-like configuration

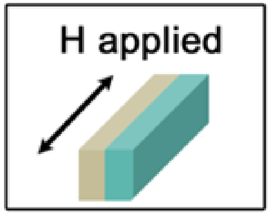


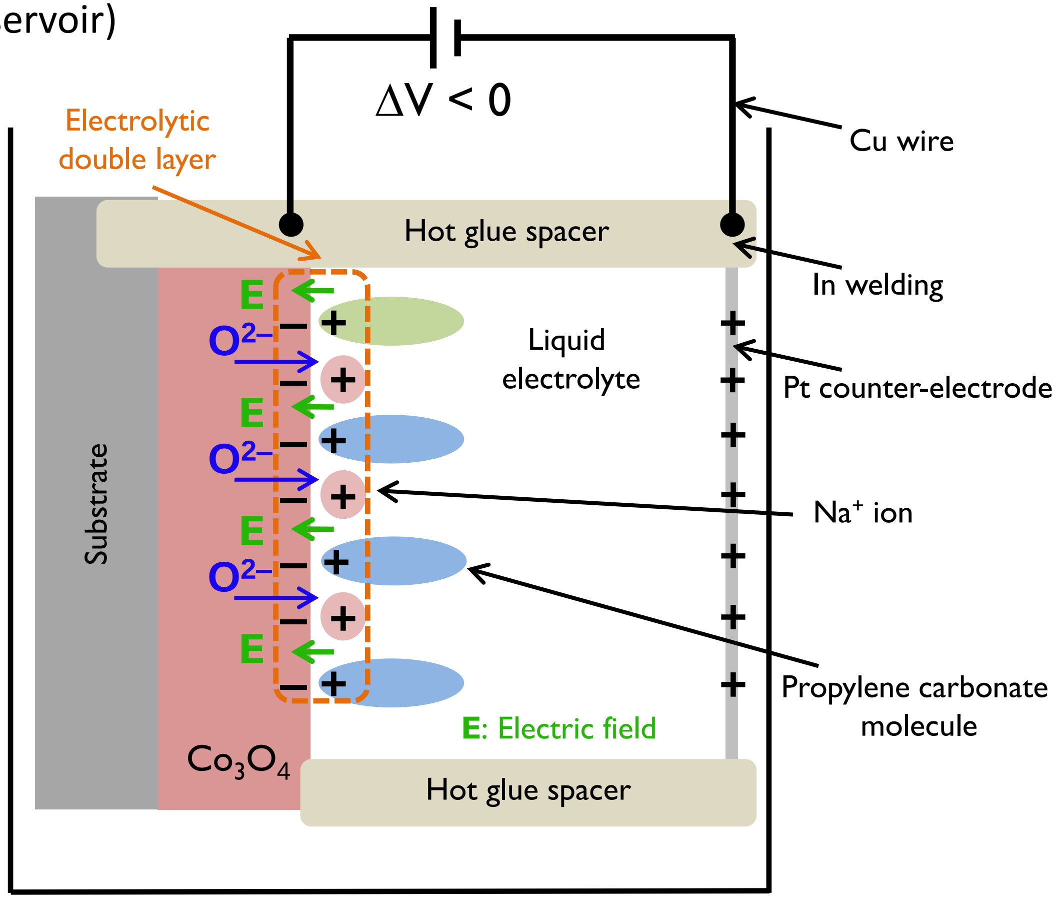
1. INTRODUCTION: MAGNETO-ELECTRIC ACTUATION

Voltage actuation by **electrolyte-gating** (liquid electrolytes: e.g., non-aqueous propylene carbonate with Na^+)

Propylene carbonate can dissolve oxygen (reservoir)

- Electrolytic cell adaptable to VSM holder





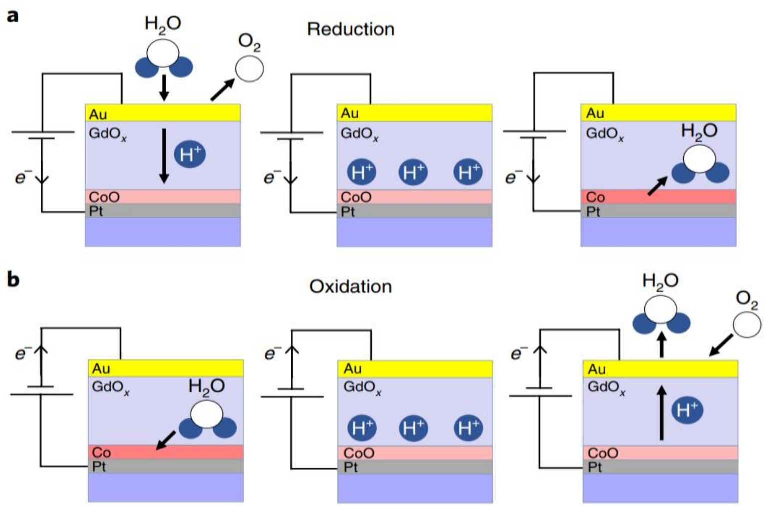
Why in liquid state?

- ➢ To ensure proper V actuation through the formation of an **electric double layer (EDL)**, with no pinholes
- ➢ To obtain high electric field with low or moderate voltages (thickness of EDL < 1 nm)

1. INTRODUCTION: MAGNETO-ELECTRIC ACTUATION

Hydrogen (H^+) magneto-ionics gives **faster** and **very strong effects!**

SOLID configuration



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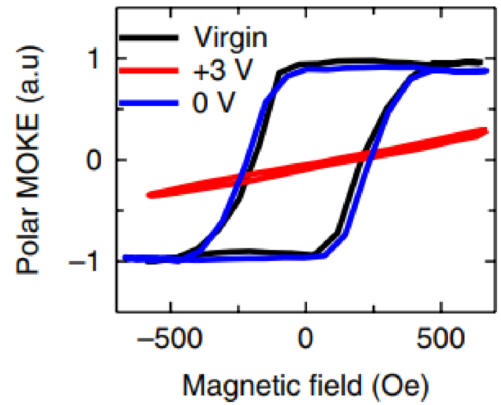
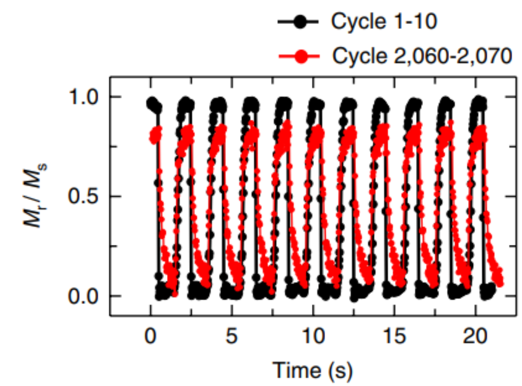
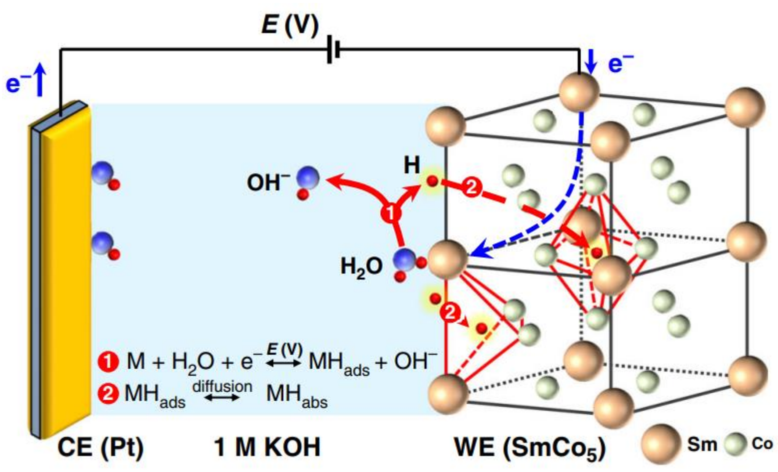
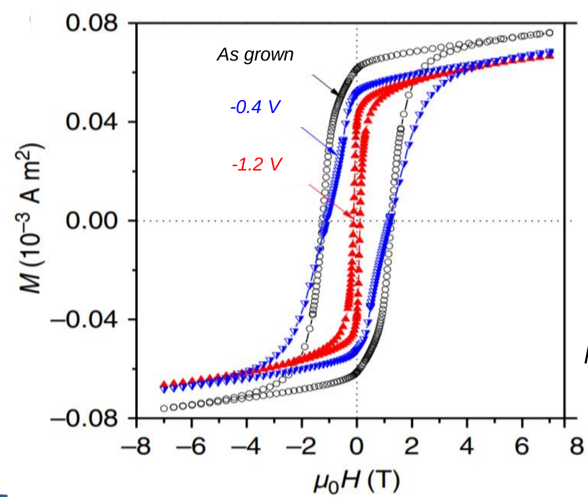








LIQUID configuration

Nat. Commun. 11 (2020) 4849

2. RESULTS & DISCUSSION

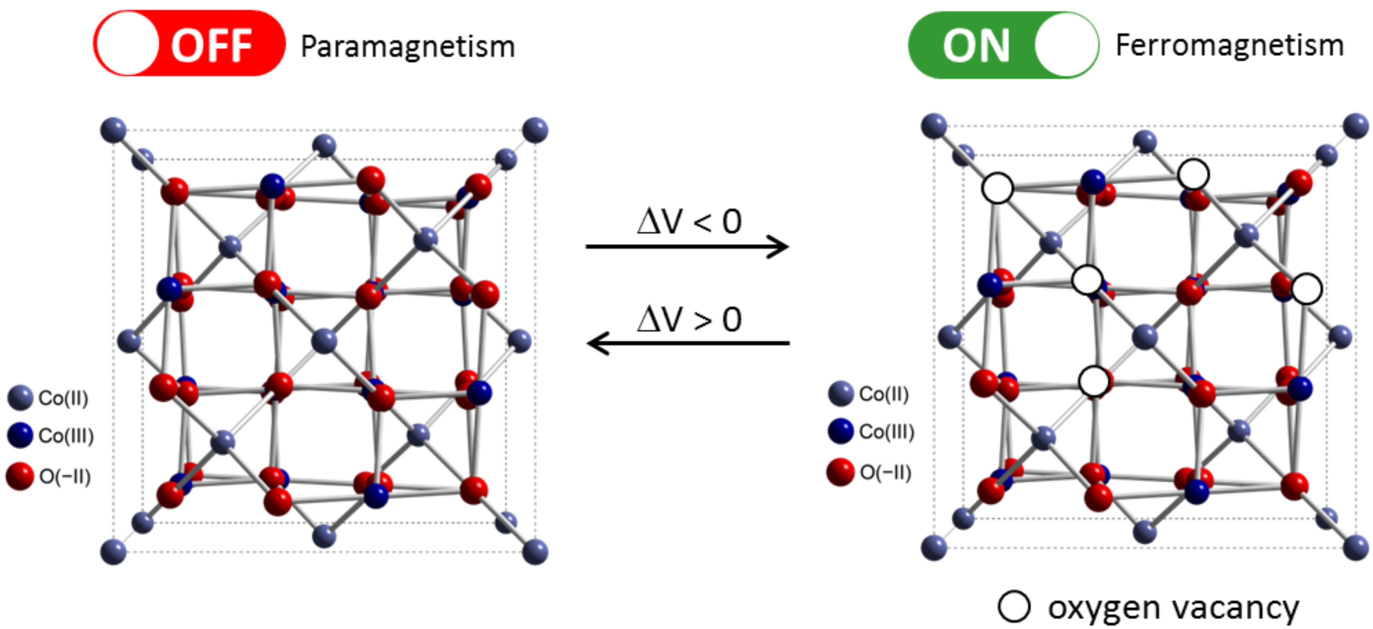
3. RESULTS AND DISCUSSION: **ON-OFF magnetism** in Co_3O_4 dense films ?

Voltage-driven motion of structural oxygen ions: the case of Co_3O_4

- **OFF-ON-OFF… (OFF: paramagnetism & ON: ferromagnetism) ??**

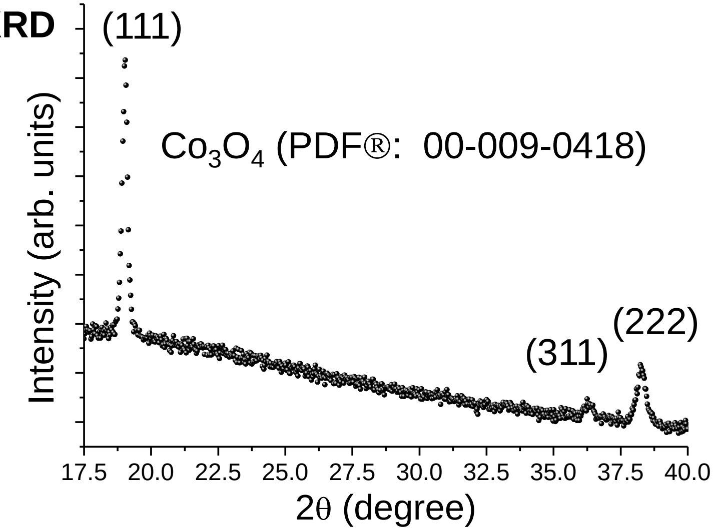
- Spinel structure

  Co_3O_4 (spinel structure)
  a = 8.0840 Å
  (PDF®: 00-009-0418)



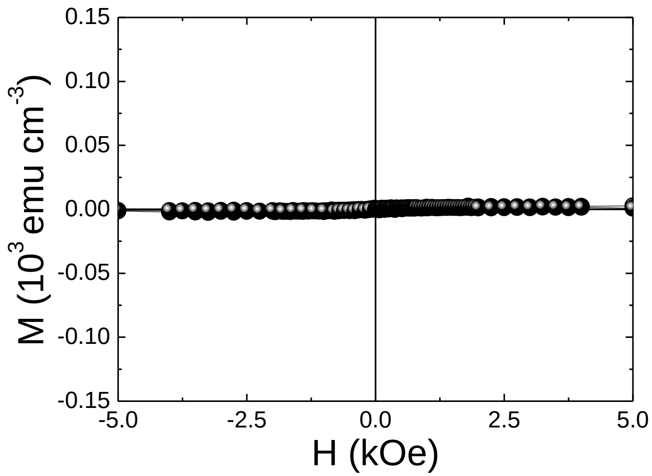
3. RESULTS AND DISCUSSION: ON-OFF magnetism in Co_3O_4 dense films ?

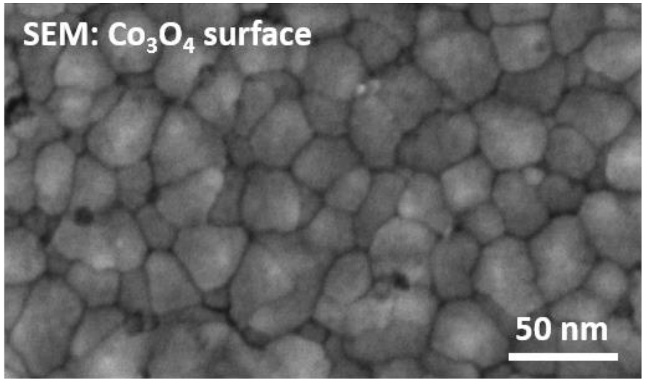
θ/2θ XRD

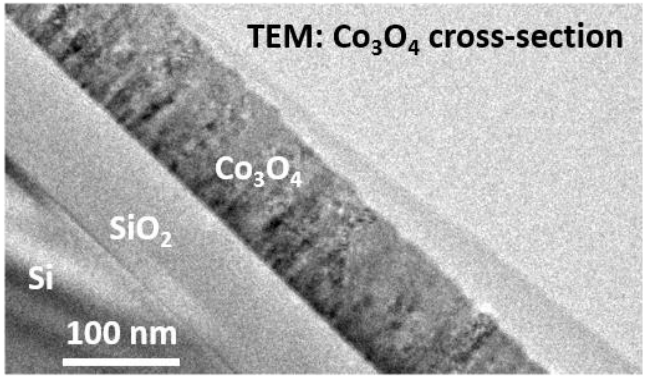


M vs. H dependence

Non-ferromagnetic (OFF)

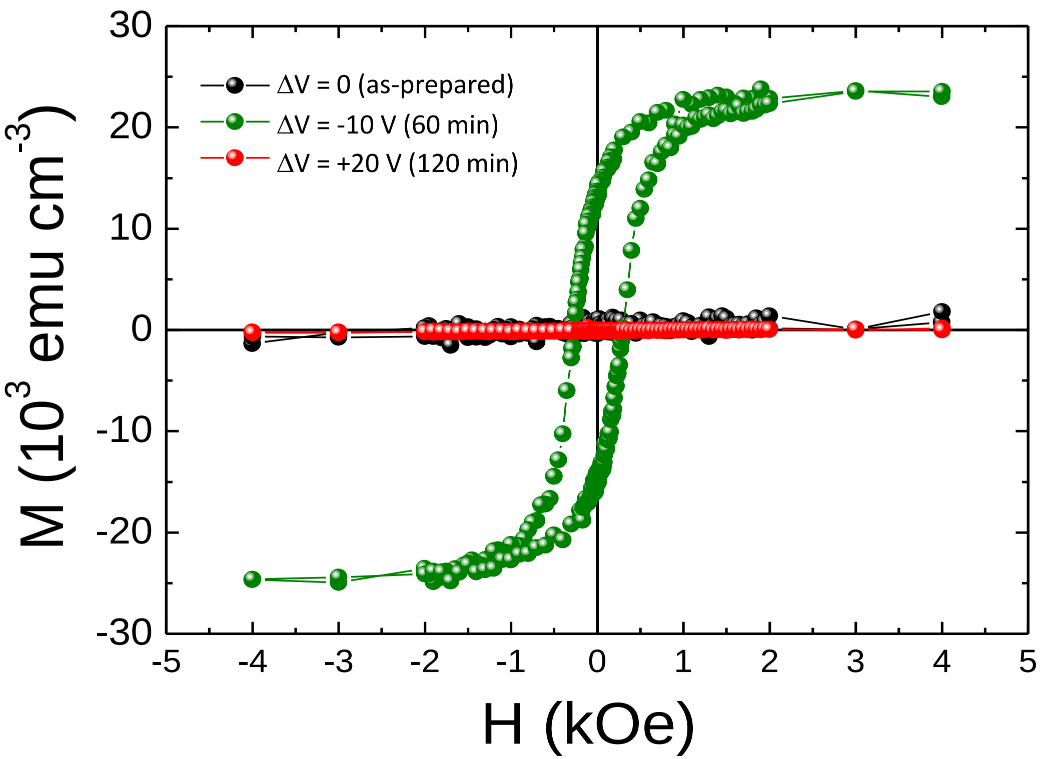






≈ 100 nm-thick **Co_3O_4** thin films by atomic layer deposition on thermally-oxidized [100]-oriented Si (100 nm-thick SiO_2/Si)

3. RESULTS AND DISCUSSION: Magneto-ionic effects in Co_3O_4 dense films



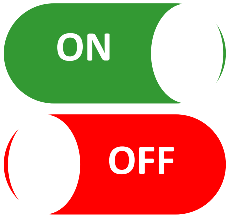
ONSET ΔV = -10 V

Full reversibility for mild voltages and moderate times

ACS Nano 12 (2018) 10291
Adv. Funct. Mater. 30 (2020) 2003704

Magnetic switch

ΔV < 0

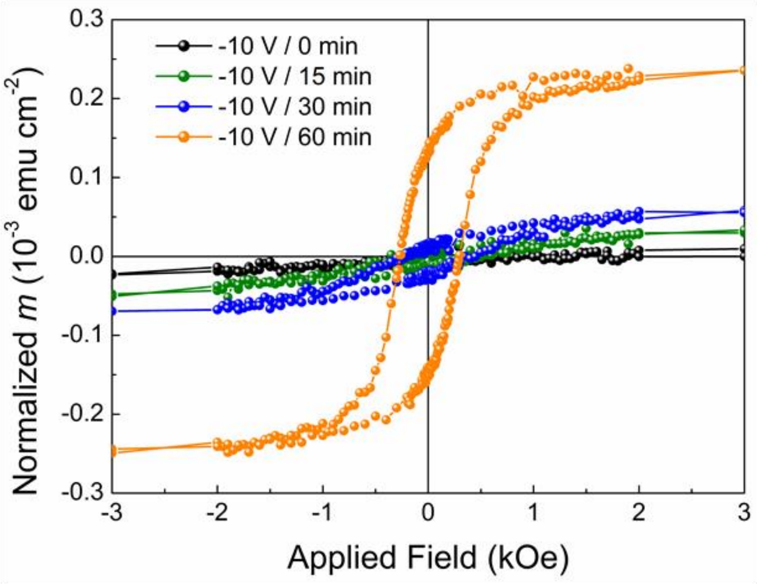


Ferromagnetism

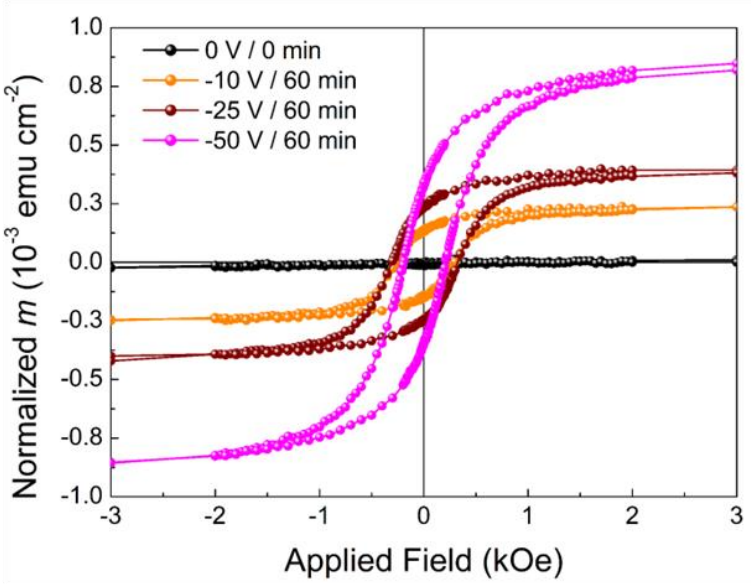
ΔV > 0 Paramagnetism

3. RESULTS AND DISCUSSION: Magneto-ionic effects in Co_3O_4 dense films

Magnetoelectric measurements by vibrating sample magnetometry



m scales with time

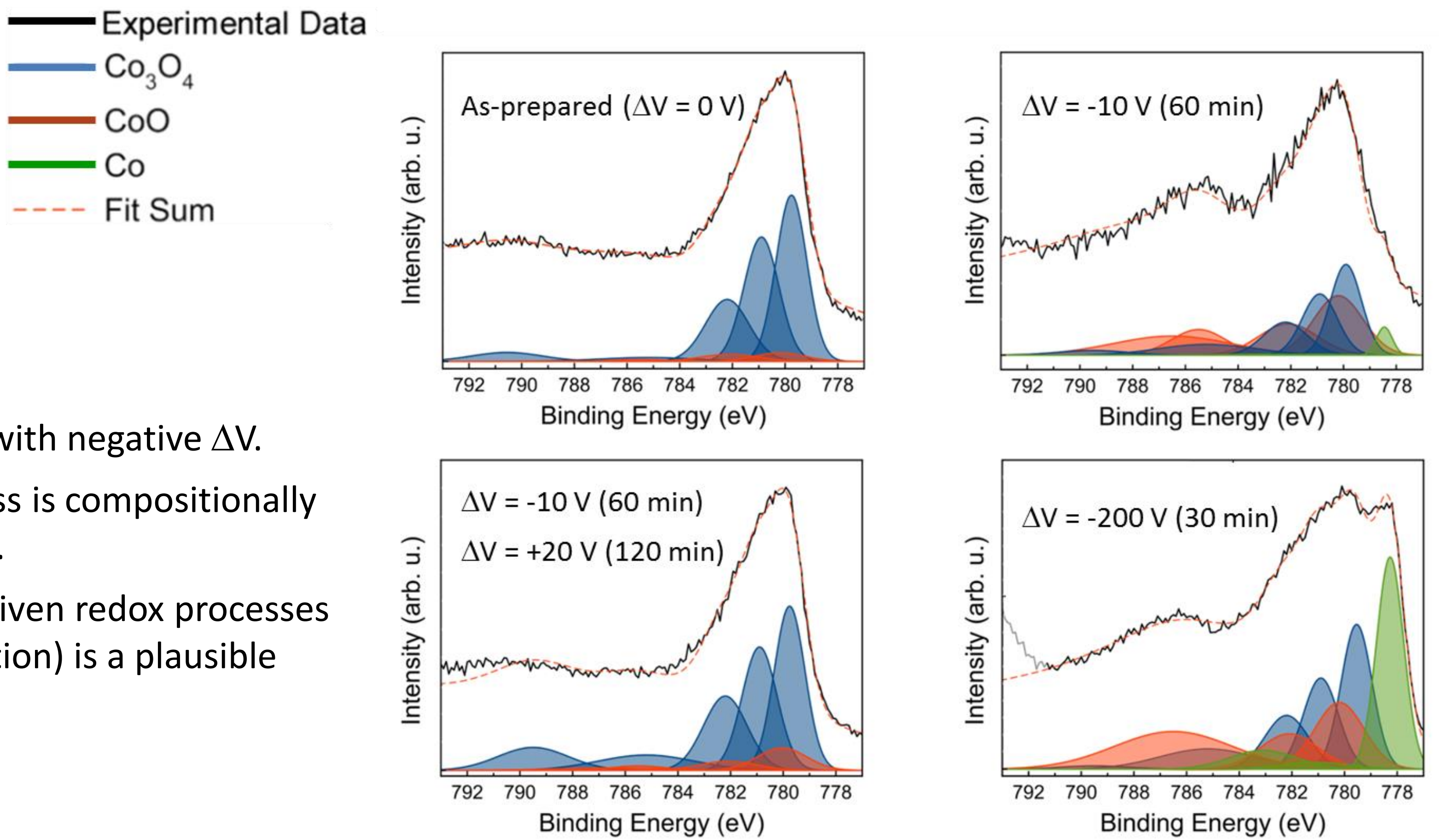


m scales with negative ΔV
(voltage-activated effect)

m (–50 V / 60 min) is equivalent to a metallic 6 nm-thick Co film (assuming FCC-Co, 164.8 emu/g, 8.9 g/cm³)

3. RESULTS AND DISCUSSION: Structural characterization

➤ What is happening from a compositional viewpoint? (XPS analysis)

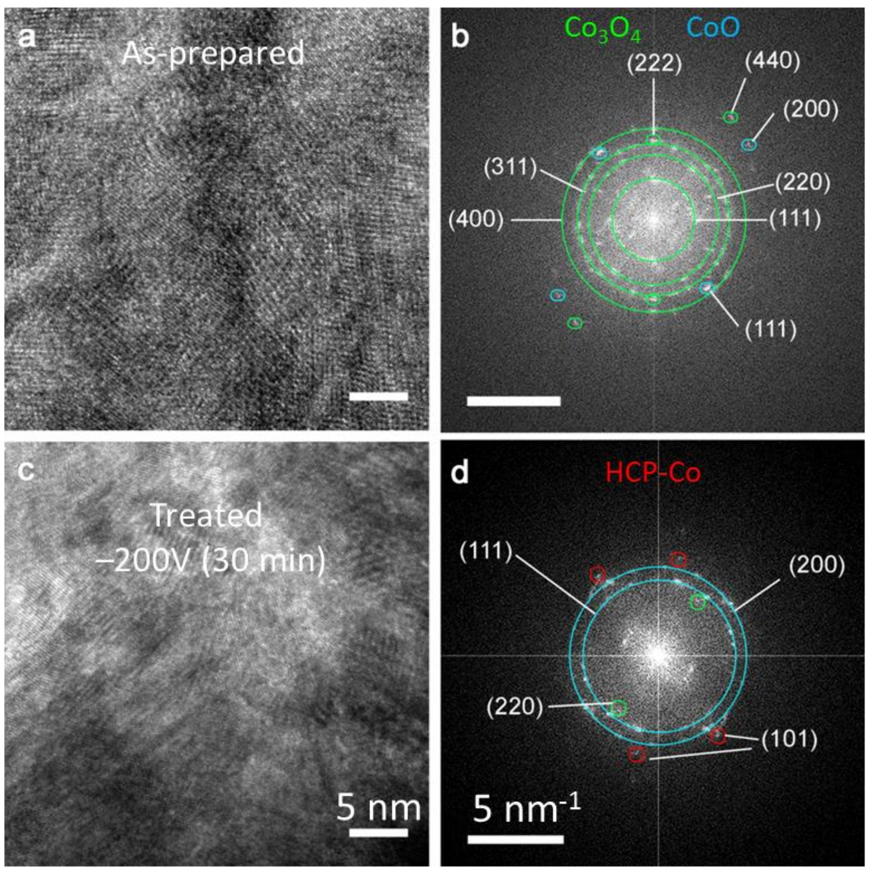


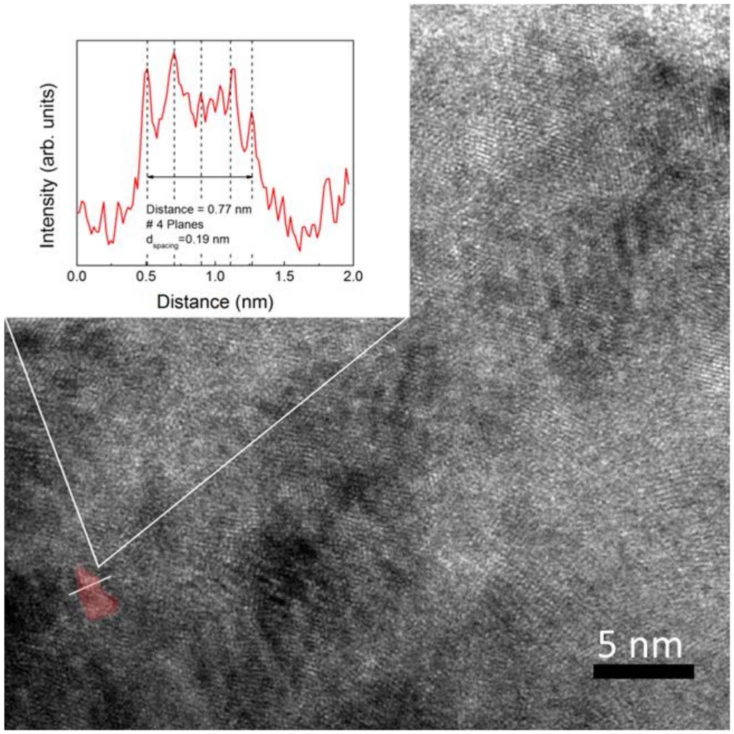
- Co scales with negative ΔV.
- The process is compositionally reversible.
- Voltage-driven redox processes (O^{2-} migration) is a plausible scenario.

3. RESULTS AND DISCUSSION: Structural characterization

What is going on from a structural point of view?

HRTEM FFT



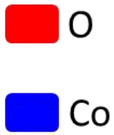


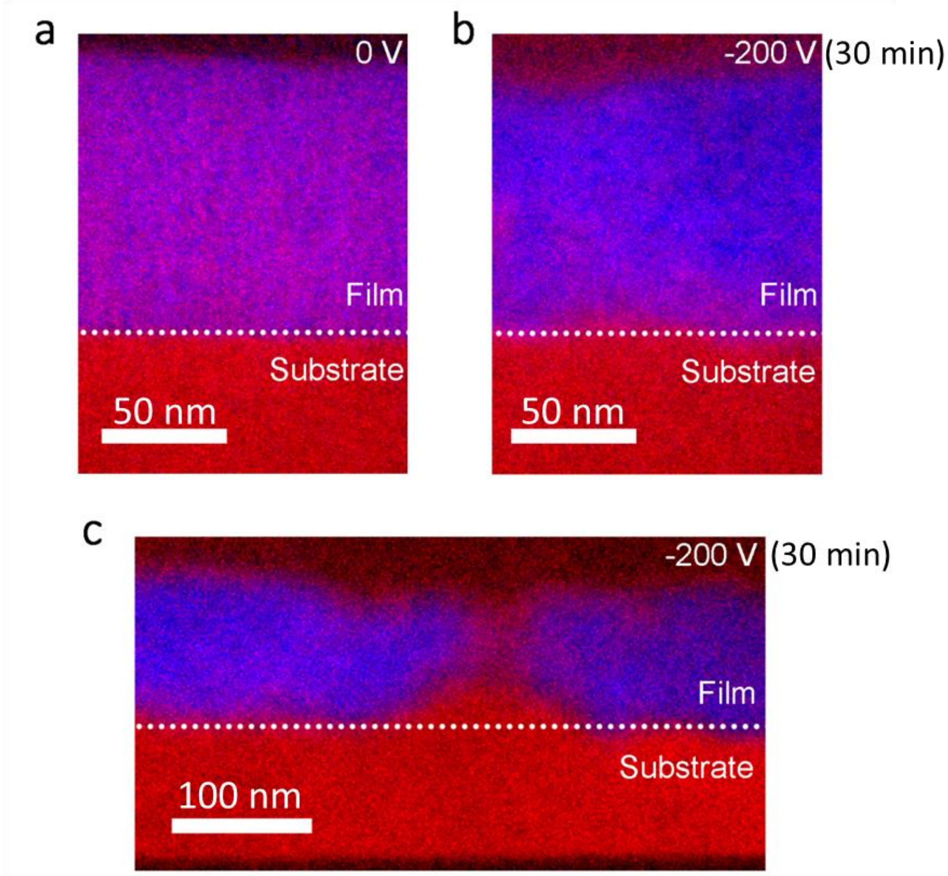
HRTEM → HCP-Co nanocrystals

3. RESULTS AND DISCUSSION: compositional characterization

Further compositional and structural insights:

EFTEM

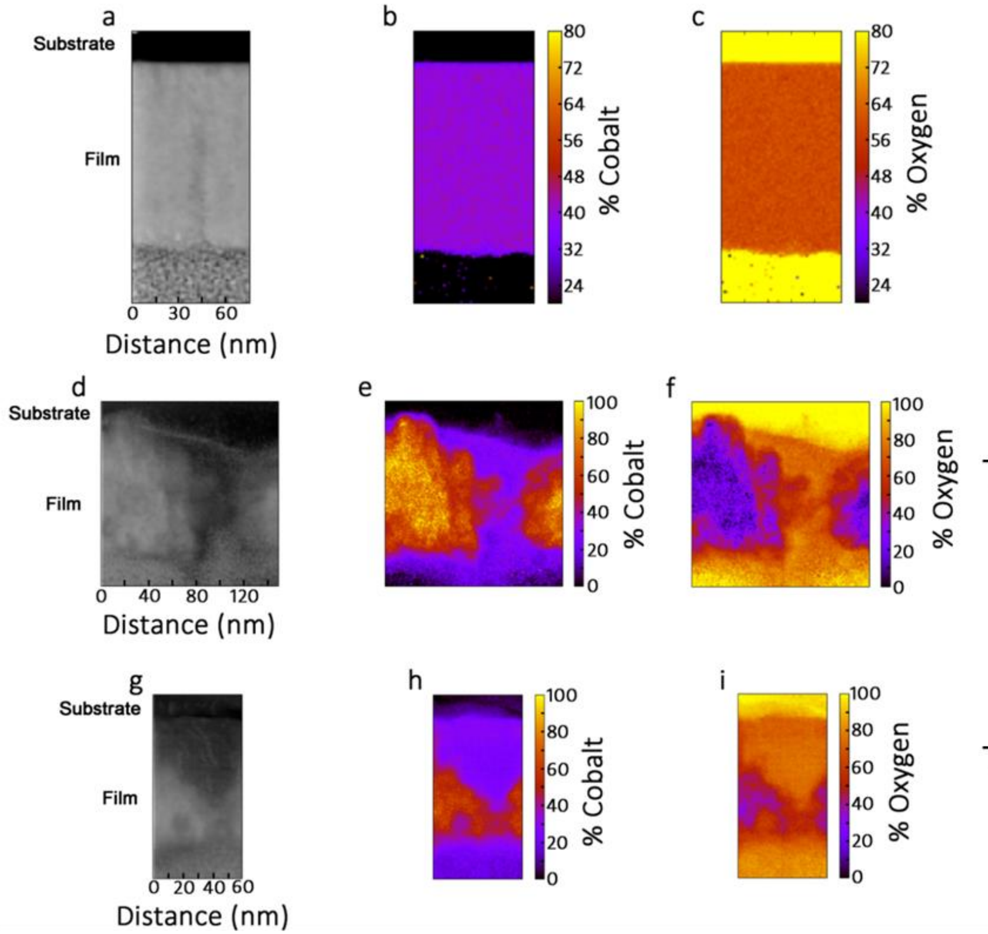




O-rich channels (O diffusion paths)

3. RESULTS AND DISCUSSION: compositional characterization

EELS



As-prepared

Treated –200V (30 min)

Treated –200V (30 min)

- O-rich channels (diffusion paths which allow for a large incorporation of O due to lack of crystallinity)
- Not only O migration! Co and O ion migration (segregation)

3. RESULTS AND DISCUSSION: in-depth structural characterization

Ion migration mechanism:

PALS (Positron Annihilation Lifetime Spectroscopy)

↓

To disentangle size and density

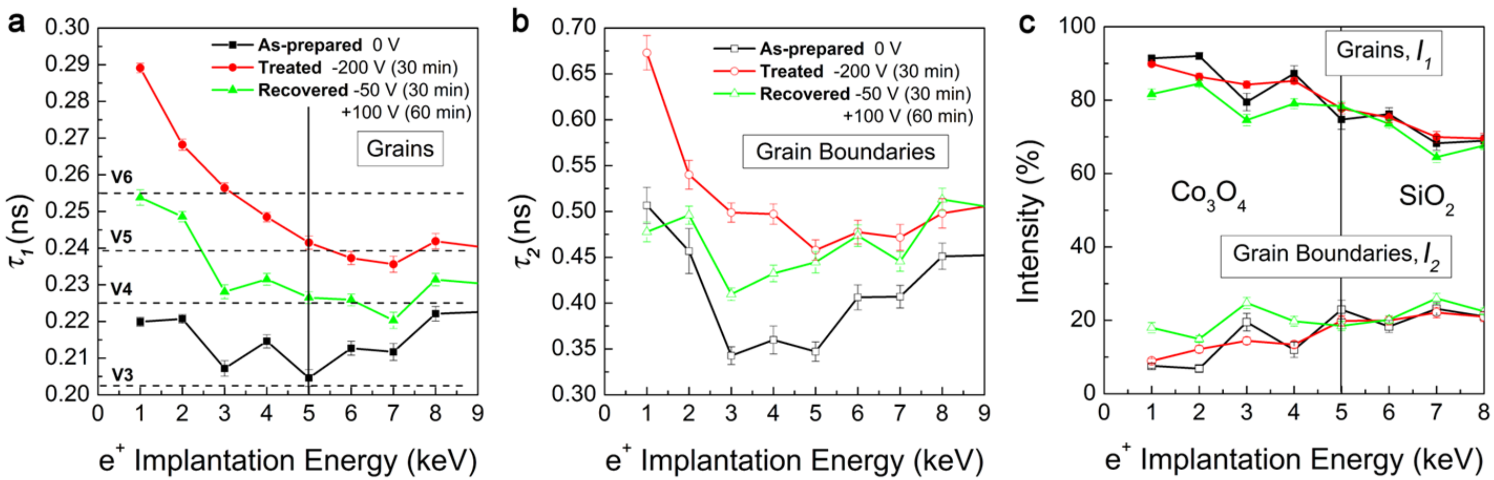
("τ" accounts for defect size and "Intensity" for defect density)

τ_1 (< 0.30 ns): vacancy clusters inside the grains ✓

τ_2 ($0.35 < \tau_2 < 0.70$ ns): grain boundaries ✓

τ_3 ($1.50 < \tau_3 < 4.00$ ns): large voids (1-3 nm in diameter) ✗

A. Quintana et al., *ACS Nano* 12 (2018) 10291.



3. RESULTS AND DISCUSSION: in-depth structural characterization

Ion migration mechanism:

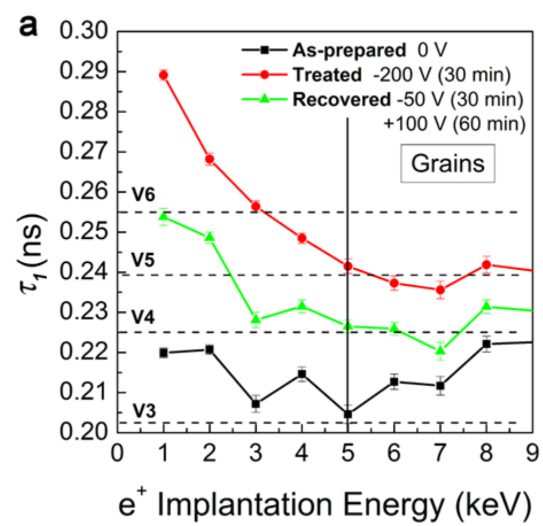
Calculated positron lifetimes τ using the atomic superposition (ATSUP) method

Co_3O_4	No. of vacancies within a complex	Vacancy type	Positron lifetime τ (ns)
	0	-	0.1188
Co vacancy clusters	1	V_{Co} (monovacancy)	0.1646
	2	V_{2xCo} (dimer)	0.1757
	3	V_{3xCo} (trimer)	0.1785
	4	V_{4xCo}	0.1795
O vacancy clusters	1	V_O (monovacancy)	0.1201
	2	V_{2xO} (dimer)	0.1255
	3	V_{3xO} (trimer)	0.1347
Mixed vacancy clusters	2	$V_{Co} + V_O$	0.1816
	3	$V_{Co} + V_{2xO}$	0.1952
	3 (V3)	**$V_{2xCo} + V_O$**	**0.2030**
	4 (V4)	**$V_{3xCo} + V_O$**	**0.2251**
	5 (V5)	**$V_{3xCo} + V_{2xO}$**	**0.2394**
	6 (V6)	**$V_{3xCo} + V_{3xO}$**	**0.2526**

J. Olsen *et al. Phys. Status Solidi C* **4** (2007) 4004

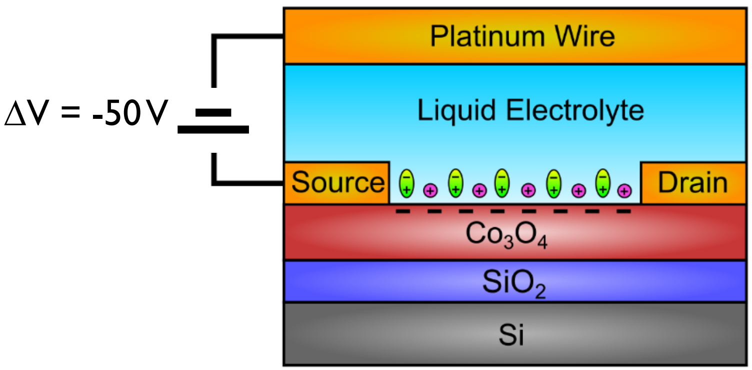
A. Quintana et al., *ACS Nano* **12** (2018) 10291.

Experimental τ

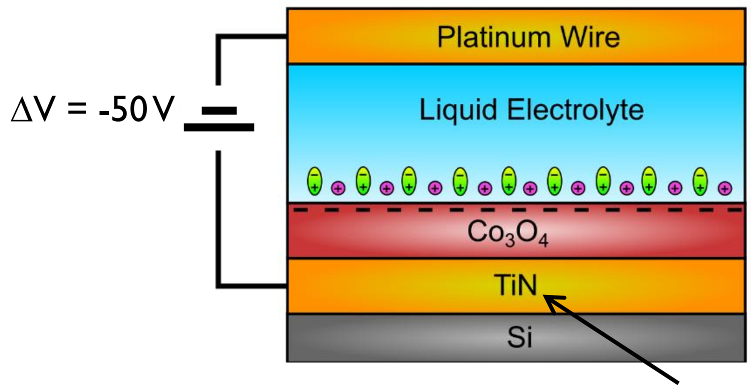


- Only mixed vacancy clusters are compatible with the experimental results!
- Not only O migration! Co and O ion migration

3. RESULTS AND DISCUSSION: Influence of the electric field configuration

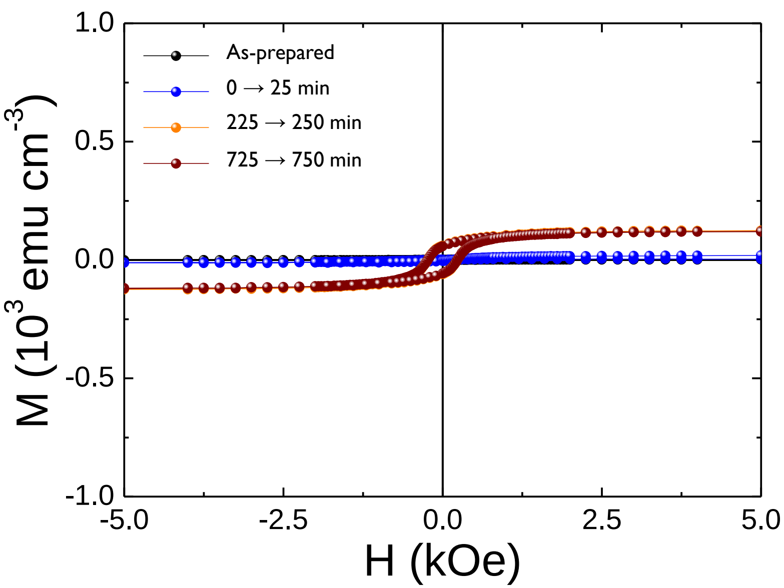


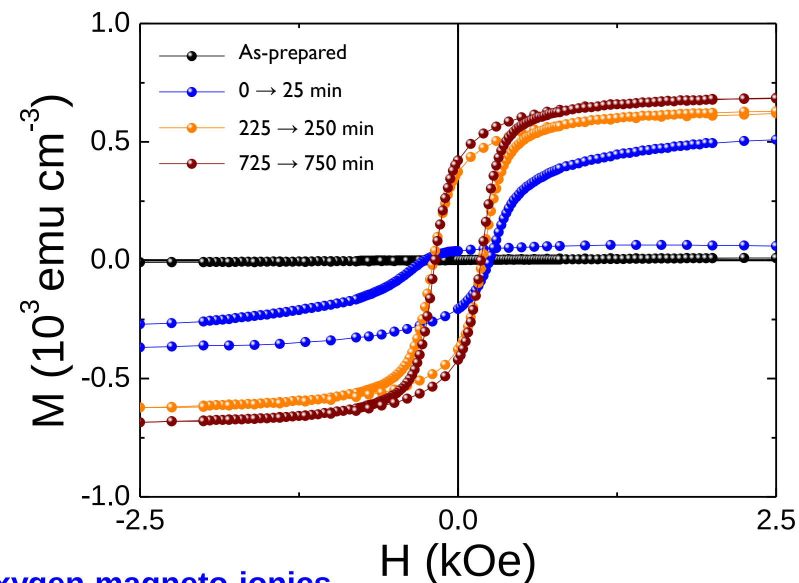
Transistor-like configuration



Capacitor configuration

Conducting buffer layer





ON-OFF oxygen magneto-ionics

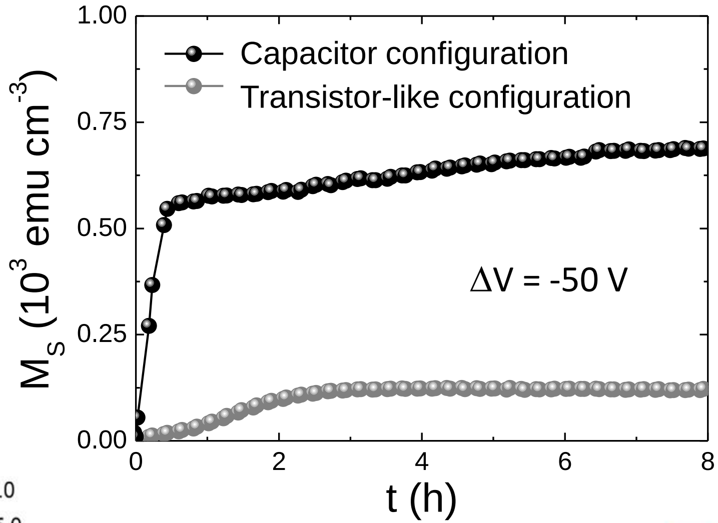
Adv. Funct. Mater. 30 (2020) 2003704

3. RESULTS AND DISCUSSION: Influence of the electric field configuration

Transistor-like configuration

Ion motion rate ≈ 30 emu cm^{-3} h^{-1}

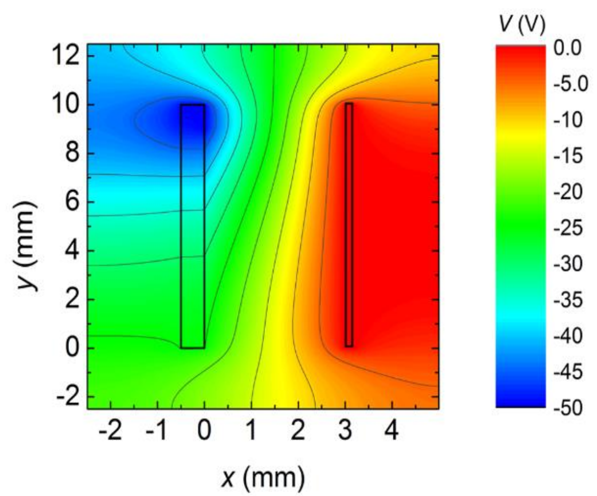
M_S (t → ∞) ≈ 120 emu cm^{-3}



Capacitor configuration

Ion motion rate ≈ 1200 emu cm^{-3} h^{-1}

M_S (t → ∞) ≈ 700 emu cm^{-3}



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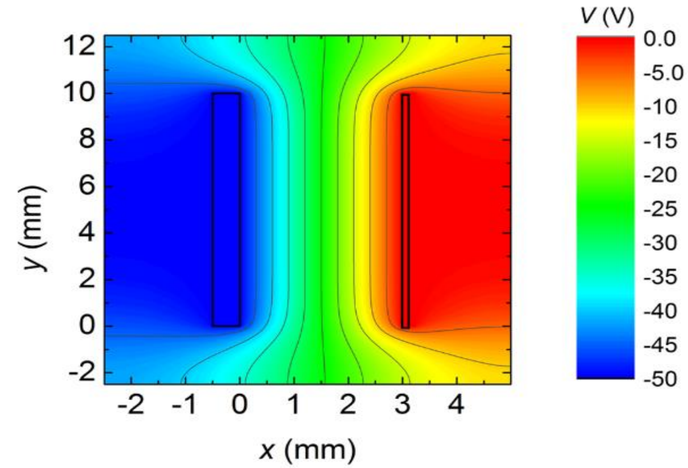
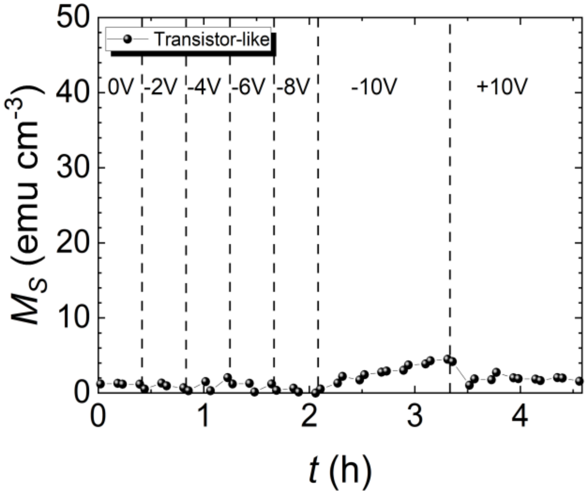
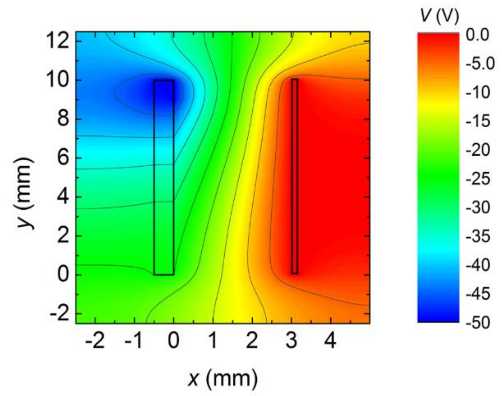


3. RESULTS AND DISCUSSION: Influence of the electric field configuration

Transistor-like configuration

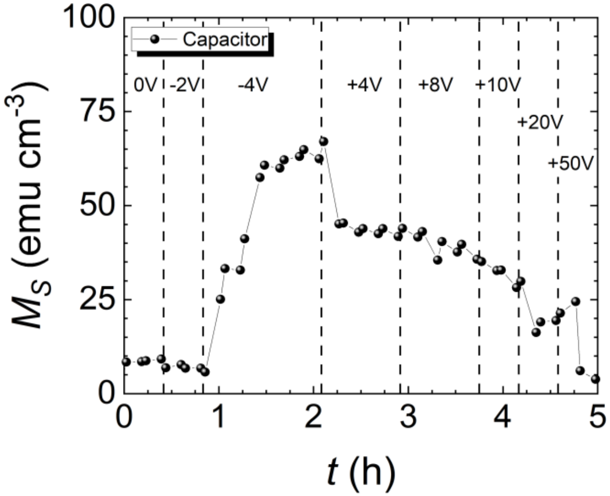


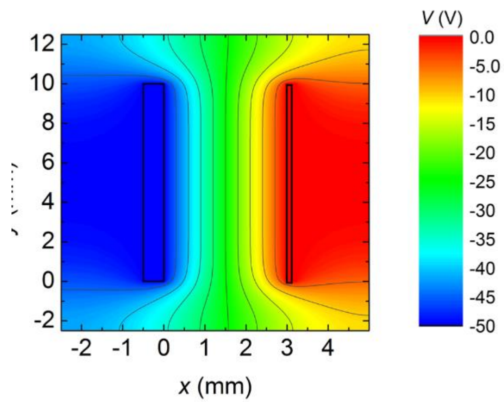


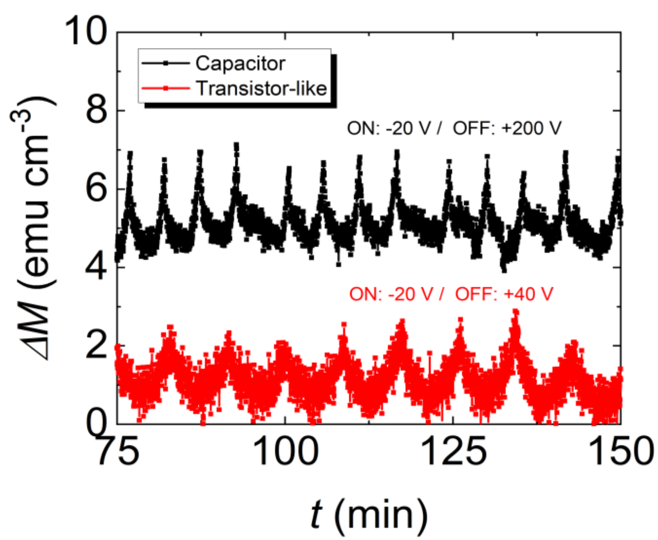
ONSET ΔV = -10 V

Cyclable (~ 10^2 cycles)

Capacitor configuration







ONSET ΔV = -4 V

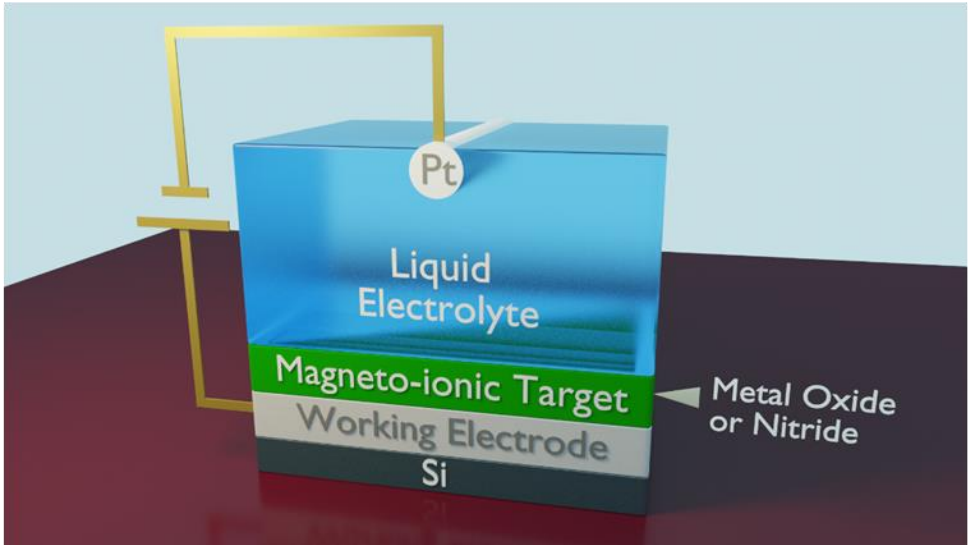
Cyclable (~ 10^2 cycles)

Adv. Funct. Mater. 30 (2020) 2003704

3. RESULTS AND DISCUSSION: What about **nitrogen magneto-ionics**?

- Paramagnetic CoN by reactive sputtering
- Interstitial N (expanded austenite)
- Ionic radius N (-III) = 132 pm > Ionic radius O (-II) = 124 pm
- CoN: a = 4.2840 Å (PDF®: 00-016-0116) *vs.* Co_3O_4: a = 8.0840 Å (PDF®: 00-009-0418)
- N is less electronegative than O & $E_{Cohesion}$ (Co nitrides) < $E_{Cohesion}$ (Co oxides) → Co-N bonds weaker than Co-O bonds

Voltage actuation by **electrolyte-gating** (non-aqueous propylene carbonate with Na^+) using a capacitor configuration

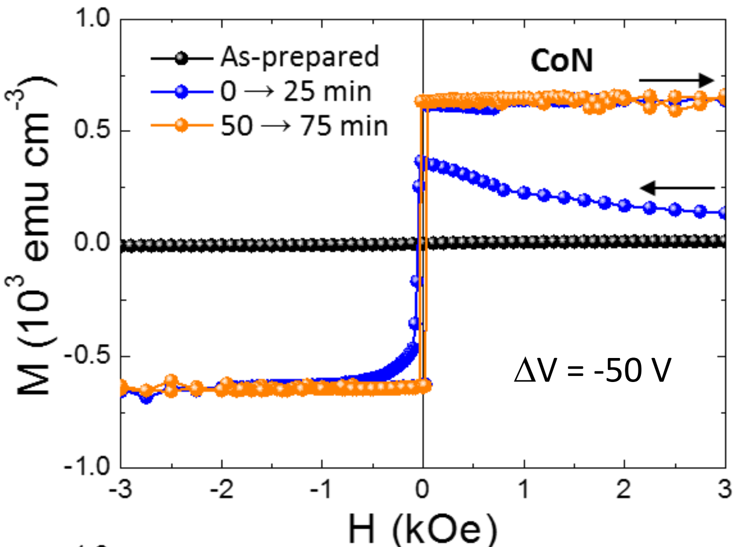


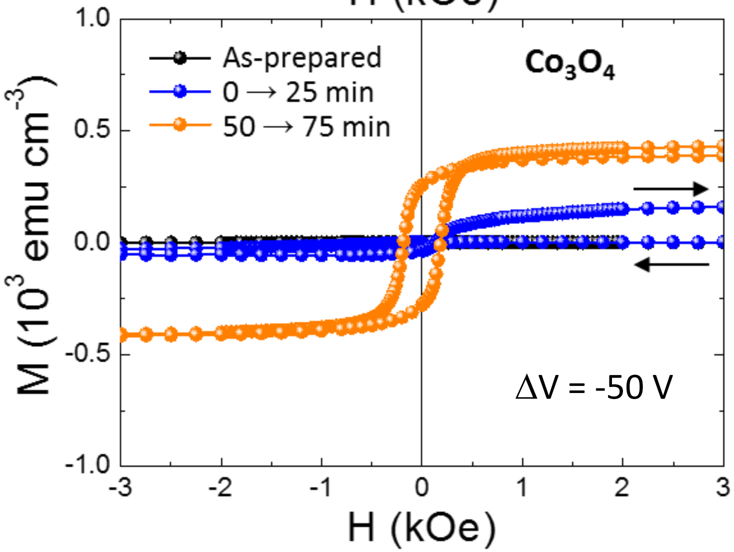
N *vs.* O magneto-ionics
(CoN *vs.* Co_3O_4)

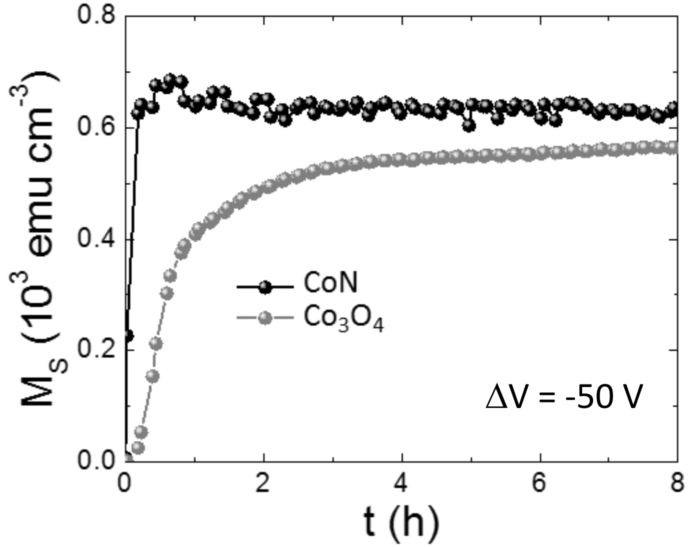
(working electrolyte: Ti/Cu seed layer)

Nat. Commun. 11 (2020) 5871

3. RESULTS AND DISCUSSION: What about **nitrogen magneto-ionics**?







Co_3O_4

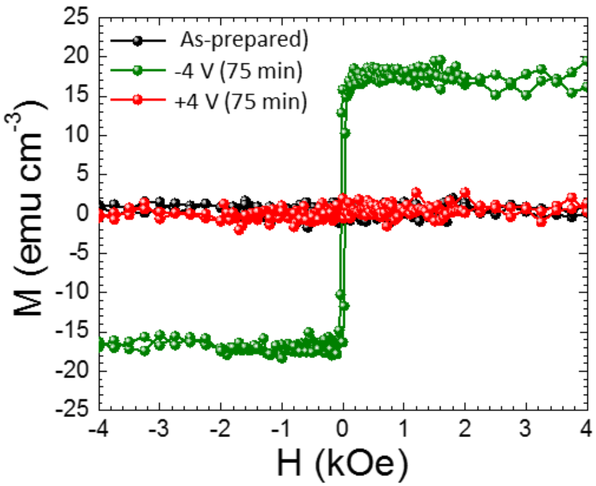
- Ion motion rate ≈ 470 emu cm^{-3} h^{-1}
- M_S ($t \rightarrow \infty$) ≈ 590 emu cm^{-3}
- ONSET ΔV = -6 V
- Cyclable (~ 10^2 cycles)

CoN

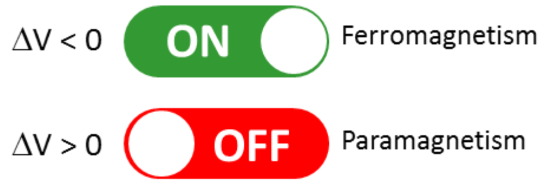
- Ion motion rate ≈ 2600 emu cm^{-3} h^{-1}
- M_S ($t \rightarrow \infty$) ≈ 640 emu cm^{-3}
- ONSET ΔV = -4 V
- Cyclable (~ 10^2 cycles)

N magneto-ionics as fast as Li-ion intercalation

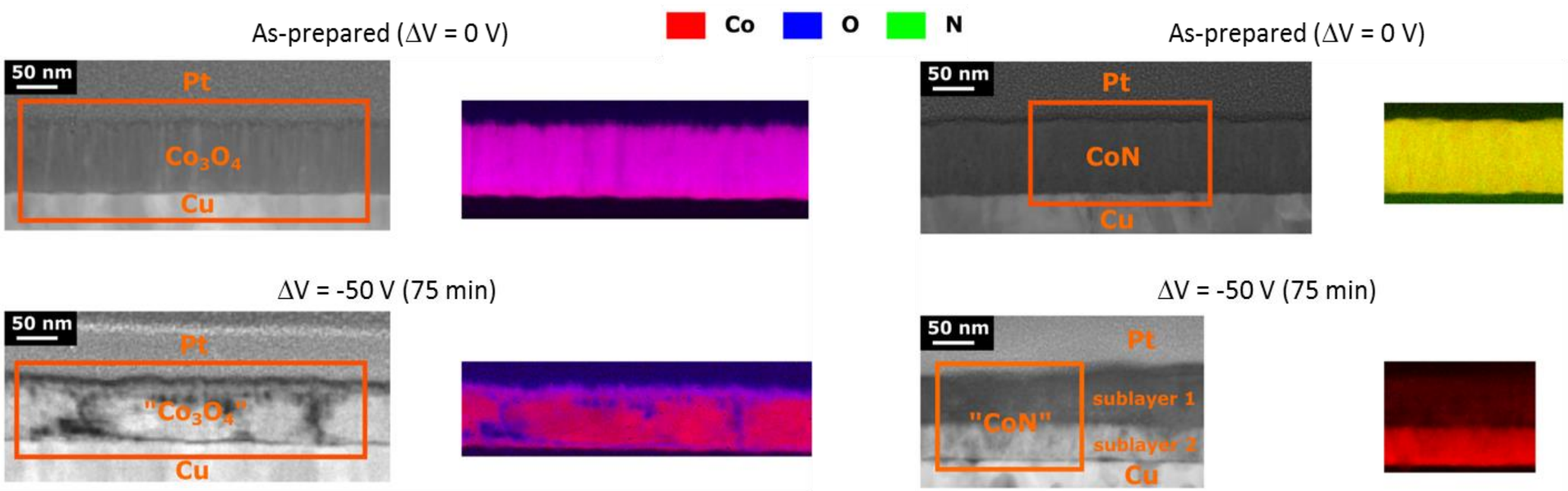
3. RESULTS AND DISCUSSION: What about **nitrogen magneto-ionics**?



Magnetic switch



Ion transport mechanisms (by electron energy loss spectroscopy)



O transport assisted by channels

N transport via a plane-wave-like migration front

3. CONCLUSIONS

4. CONCLUSIONS

- ✓ **ON-OFF ferromagnetism by O and N magneto-ionics (stable, tunable & reversible).**
- ✓ **The way to apply electric field is crucial to determine ion speed and generated magnetization (capacitor configuration better).**
- ✓ **N transport via plane-wave-like migration fronts while O transport is assisted by diffusion channels.**
- ✓ **N magneto-ionics shows lower ONSET voltages and enhanced ion motion.**
- ✓ **N magneto-ionics interesting to integrate this effect with other nitrides used in microelectronics.**

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THANK YOU FOR YOUR KIND ATTENTION !