Magneto-ionics: using ionic motion to control magnetism

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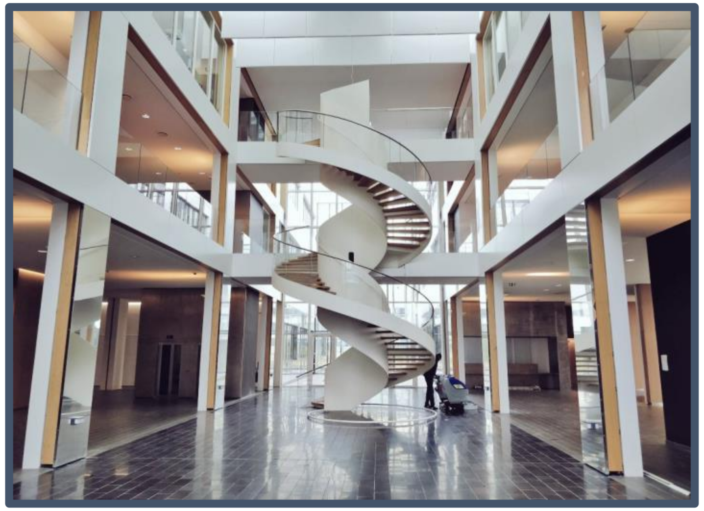
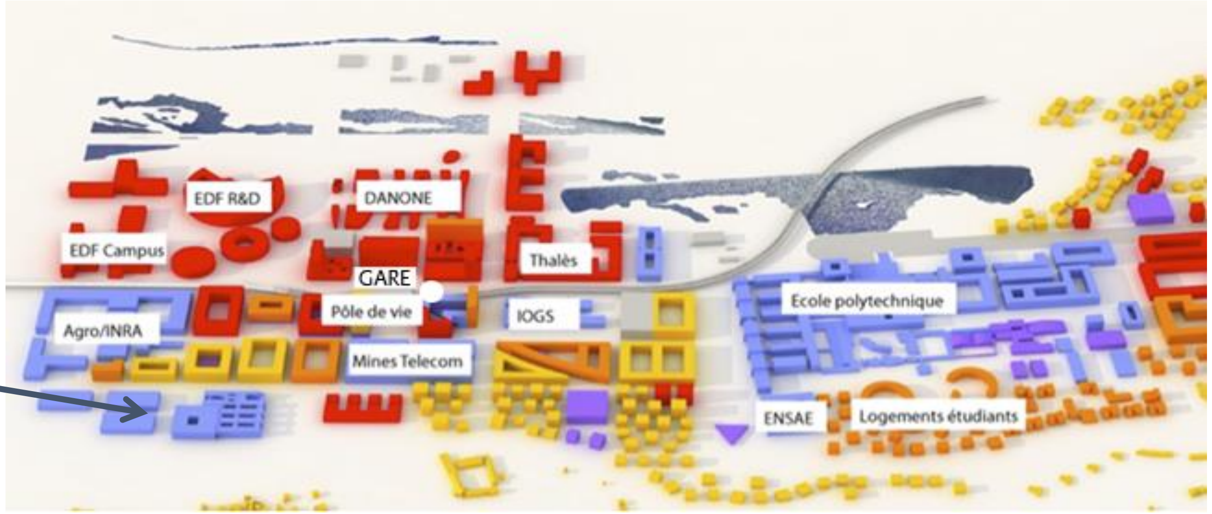
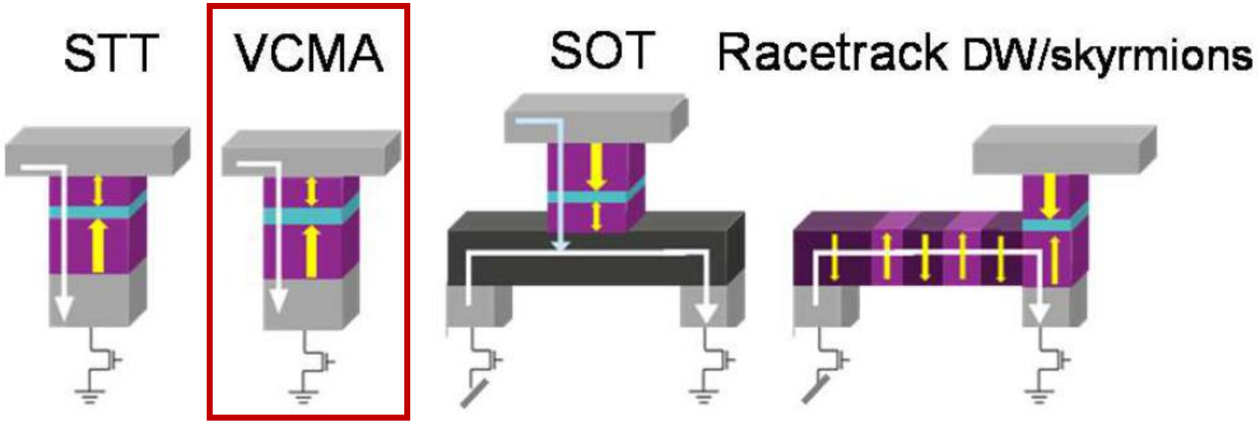


- 400 members
- 4 research departments:
  - Nanoelectronics
  - Photonics
  - Materials
  - NanoBio Fluidics
- 3000 m² of cleanroom



Why do we want to control magnetism with a gate voltage?

Magnetic memory architectures



B. Dieny et al. Nature Electronics, 3, 446–459 (2020).

Challenges:

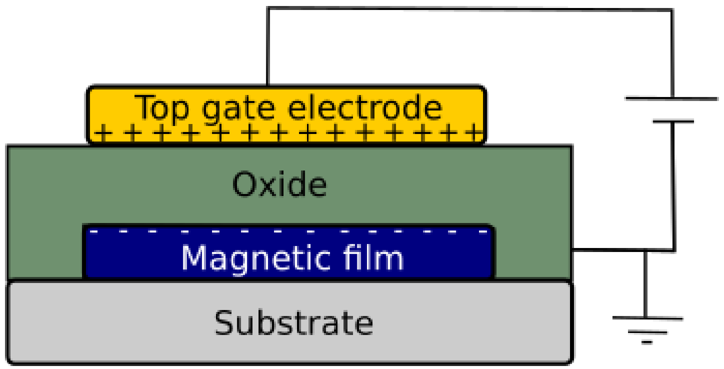
- **Lower writing current while keeping long retention times**
- Field free switching, improve charge-spin conversion and lower resistivity in SOT RAM
- High DW/skyrmion pinning in nano-tracks, skyrmion Hall effect

We need a switch between:

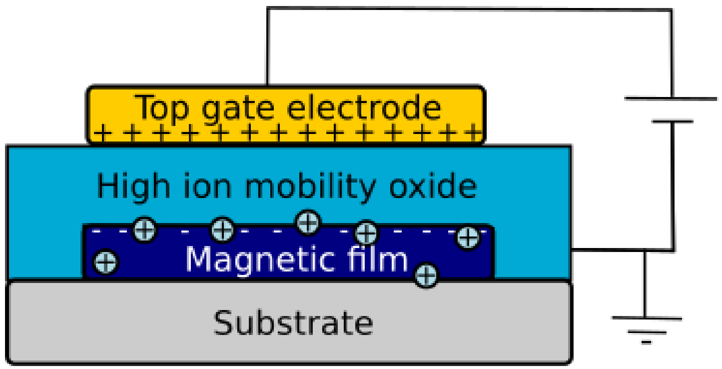
soft magnetic states for easy information writing	**E-field controlled**	**hard magnetic states for reliable information retention**
DW/skyrmion pinning and nucleation	⟷	**DW/skyrmion depinning and anhililation**

How do we control magnetism with a gate voltage?

Charge effects



Ionic effects



✓ High charging speed
✓ Compatible with conventional electronics

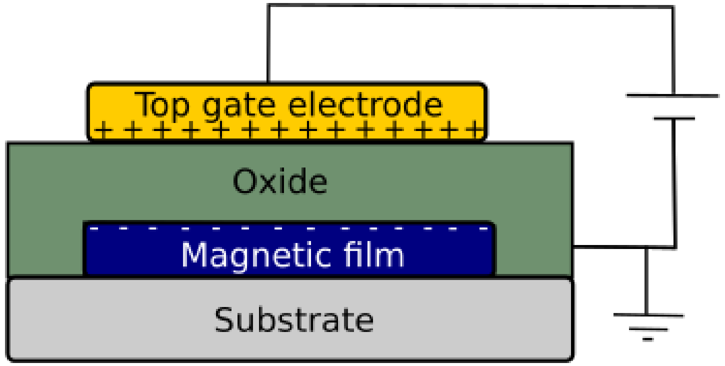
X Volatile
X Limited to the top interface

✓ Non volatile
✓ Effects on both top and bottom interfaces

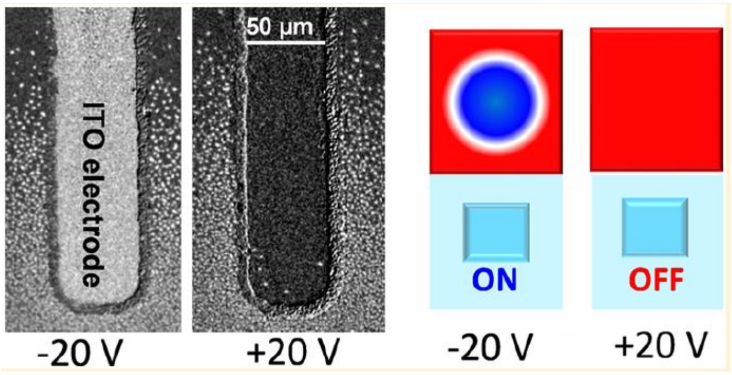
X Speed limited by ionic diffusion dynamics
X Reversibility and cyclability affected by chemical reactions

How do we control magnetism with a gate voltage?

Charge effects

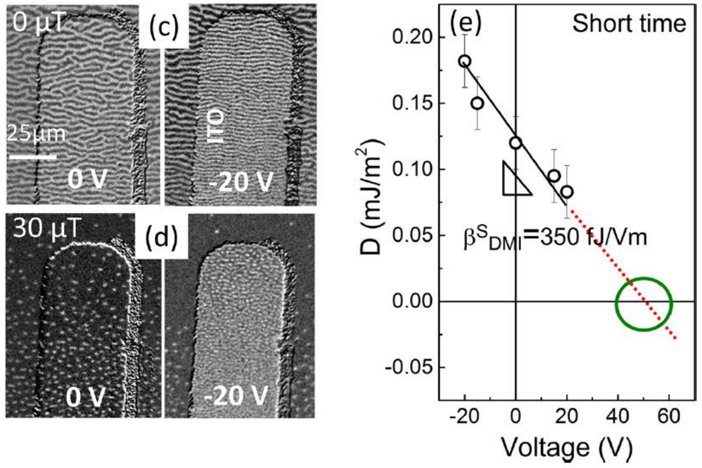


$Pt/Co/Al_2O_3/HfO_2$



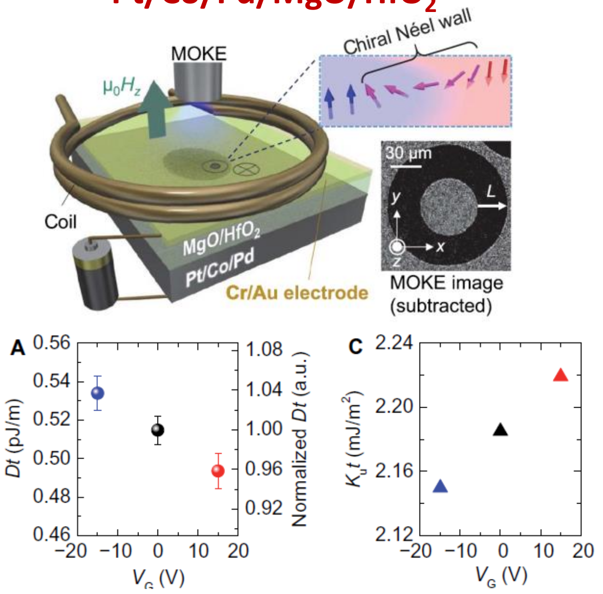
M. Shott et al., Nano Lett. 17, 3006 (2017)

$Ta/CoFeB/TaOx/HfO_2$



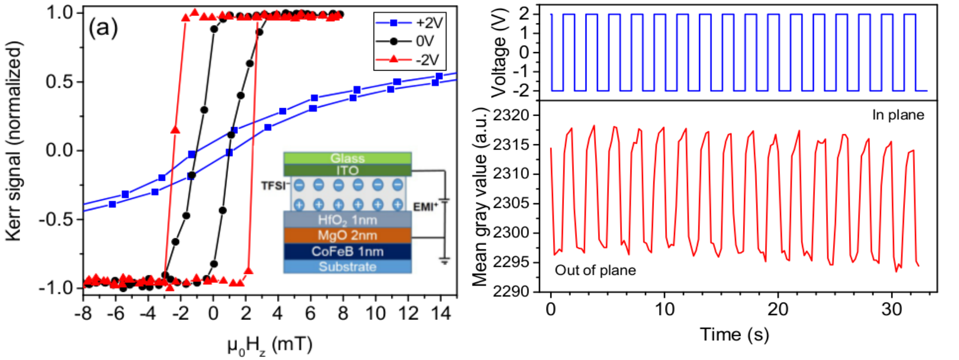
T. Srivastava et al., Nano Lett. 18, 4871 (2018)

$Pt/Co/Pd/MgO/HfO_2$



T. Koyama, et al. Science Advances 4, 0265 (2018).

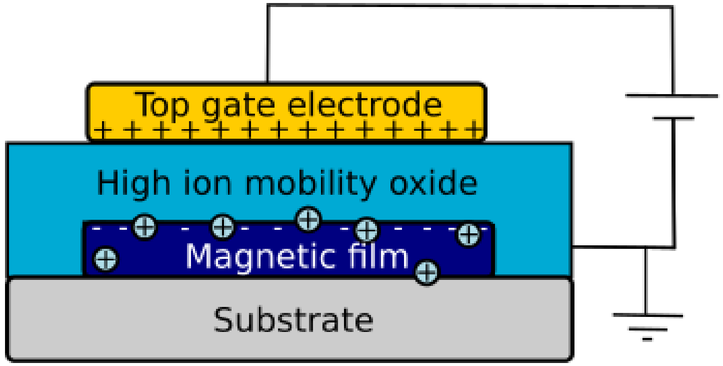
$Ta/CoFeB/HfO_2$



Y. Liu, *et al.*, J. Appl. Phys. 122, 133907 (2017).

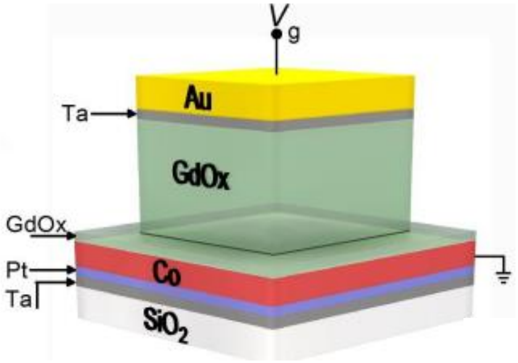
Magneto-ionics: devices

Ionic effects = Magneto-Ionics



Solid state devices

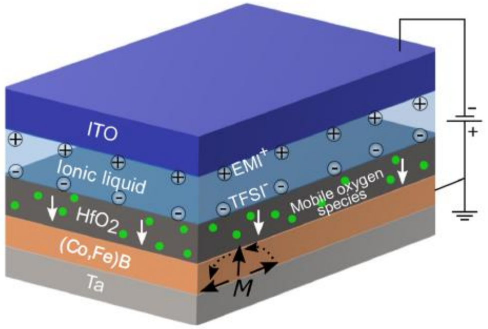
- ✓ More compatible with applications
- ✓ Perspectives for miniaturisation



U. Bauer, *et al. Nature Materials* 14, 174–81 (2015).

Ionic liquid gating

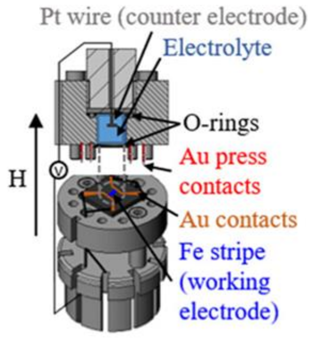
- ✓ Robust and simple device design
- ✓ High E-fields over large areas



R. Pachat, *et al.*, Phys. Rev. Appl. 15, 064055 (2021).

Electrochemistry

- ✓ In-depth characterisation of electrochemical reactions



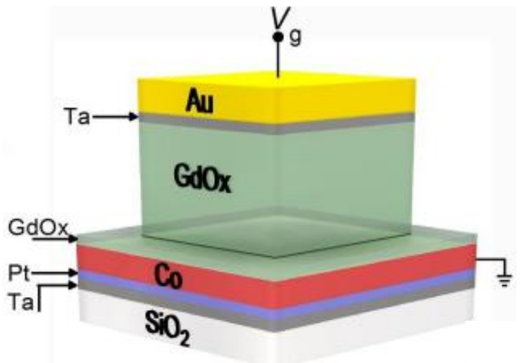
In situ transport cell

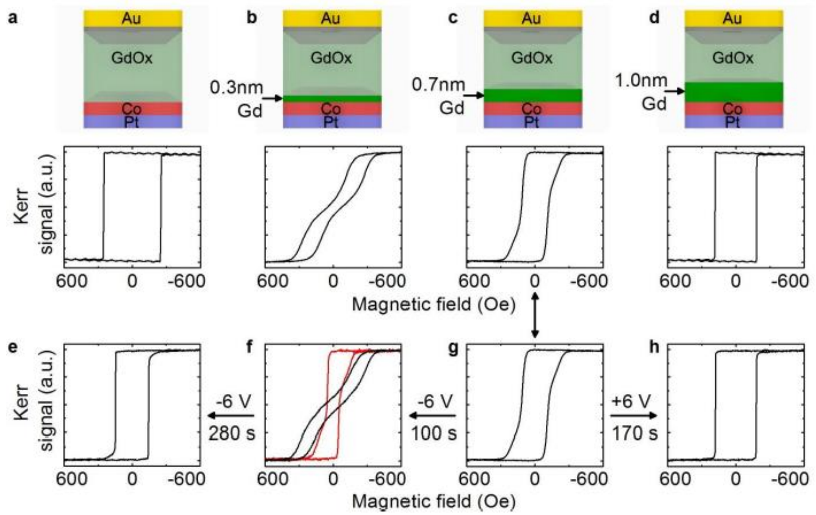
M. Nichterwitz, *et al*. ACS Appl. Electron. Mater, 2, 8, 2543–2549 (2020).

What are the mobile ions?

Oxygen ions

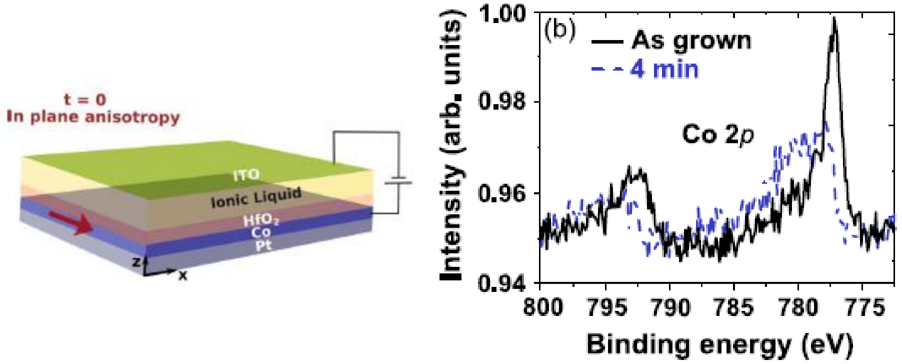
The oxidation front can be controlled by gate voltage and temperature



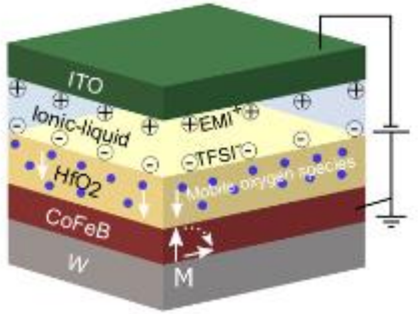


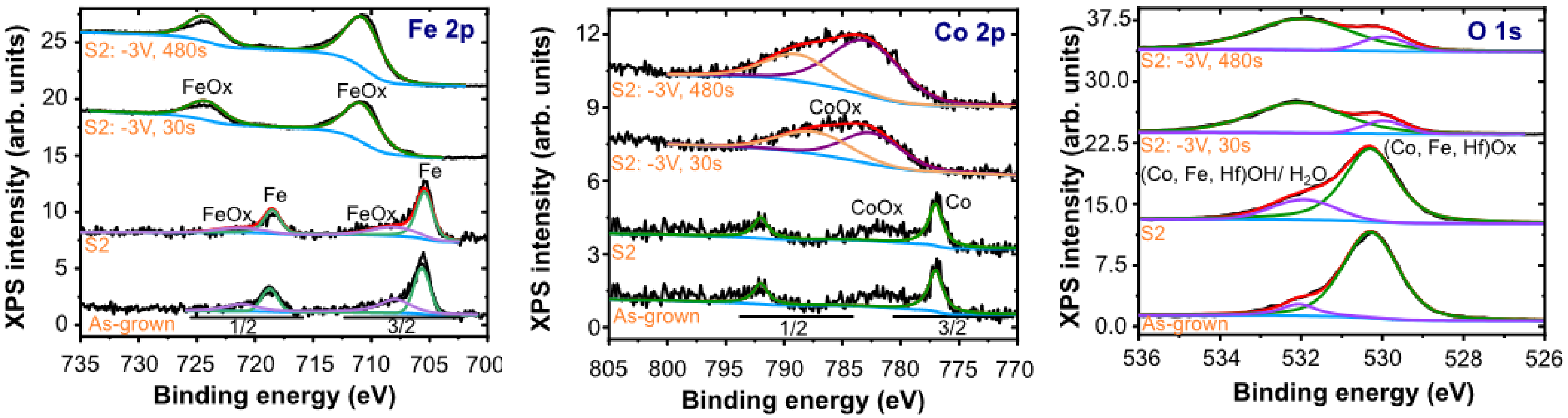
U. Bauer, *et al. Nature Materials* 14, 174–81 (2015).

Co oxidation induced by voltage at room temperature



L. Herrera Diez *et al.,* Phys. Rev. Appl. 12, 034005 (2019)





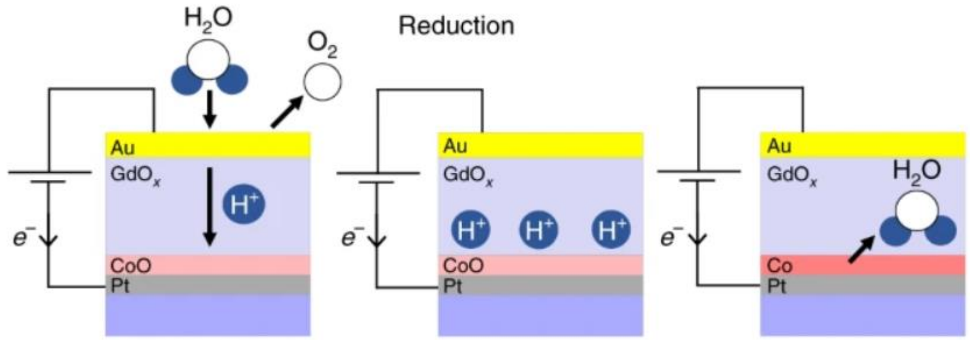
R. Pachat, *et al.*, Phys. Rev. Appl. 15, 064055 (2021).

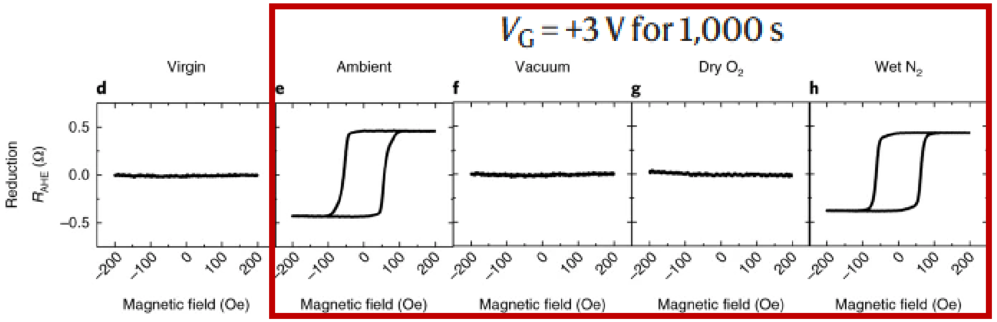
R. Pachat, *et al.*, Submitted (2022).

High mobility of oxygen species and contribution of atmospheric water

What are the mobile ions?

Hydrogen ions





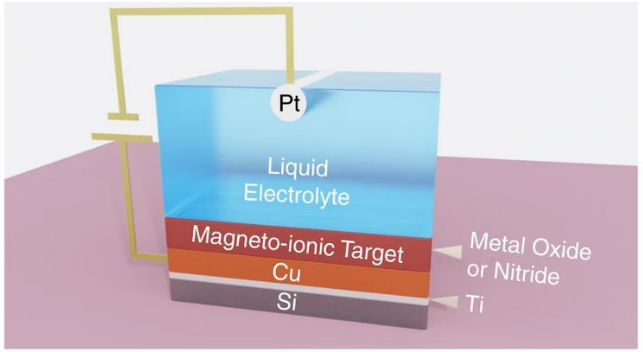
A. J. Tan *et al.* *Nature Materials* 18, 35-41 (2019).

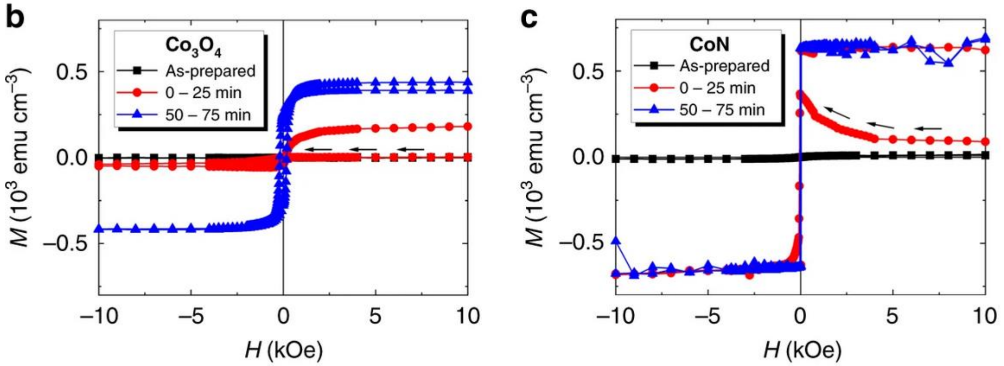
Strong indication of water electrolysis

$$\text{Anode (Au)}: \quad 2H_2O \rightarrow 4H^+ + O_2 + 4e^-$$

$$\text{Cathode (Pt)}: \quad 4H^+ + 2Co^{2+} + 2O^{2-} + 4e^- \rightarrow 2H_2O + 2Co$$

Nitrogen ions



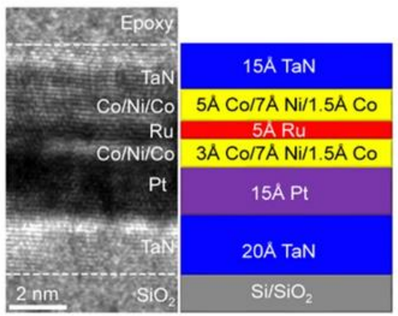


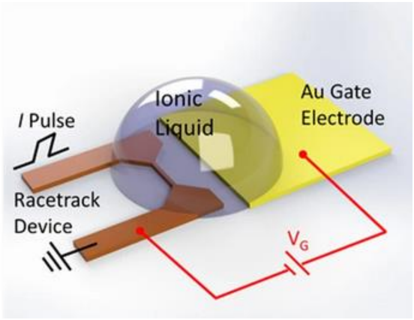
J. De Rojas *et al.* *Nature Communications* 11, 5871 (2020).

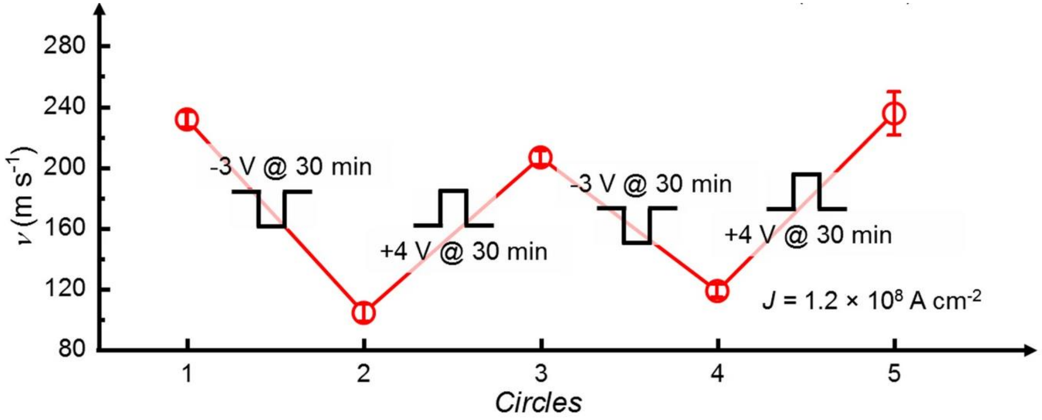
N^{2-} transport is more homogeneous and requires less activation energy than for O^{2-} ions

Not only in ferromagnets

Synthetic antiferromagnets



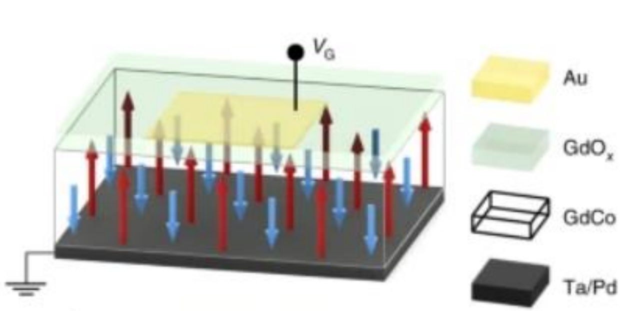


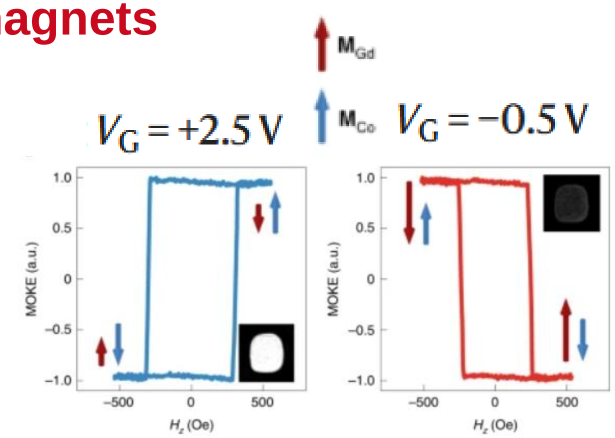


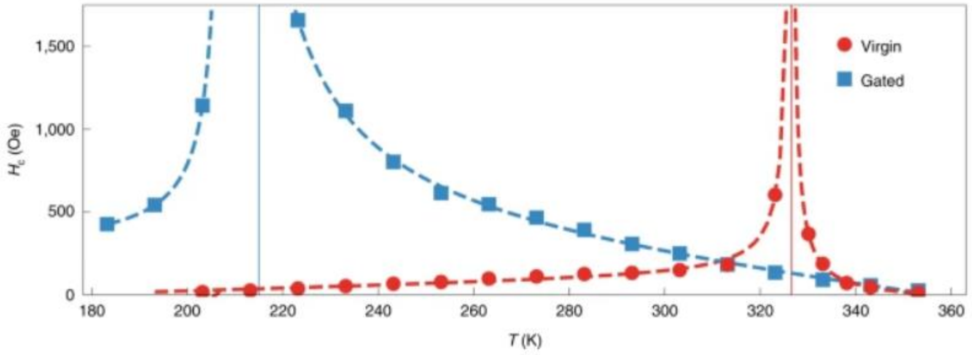
Y. Guan *et al*. *Nature Communications* 12, 5002 (2021).

Ionics used to tune the moment compensation between the CoNi layers to increase domain wall velocity

Ferrimagnets





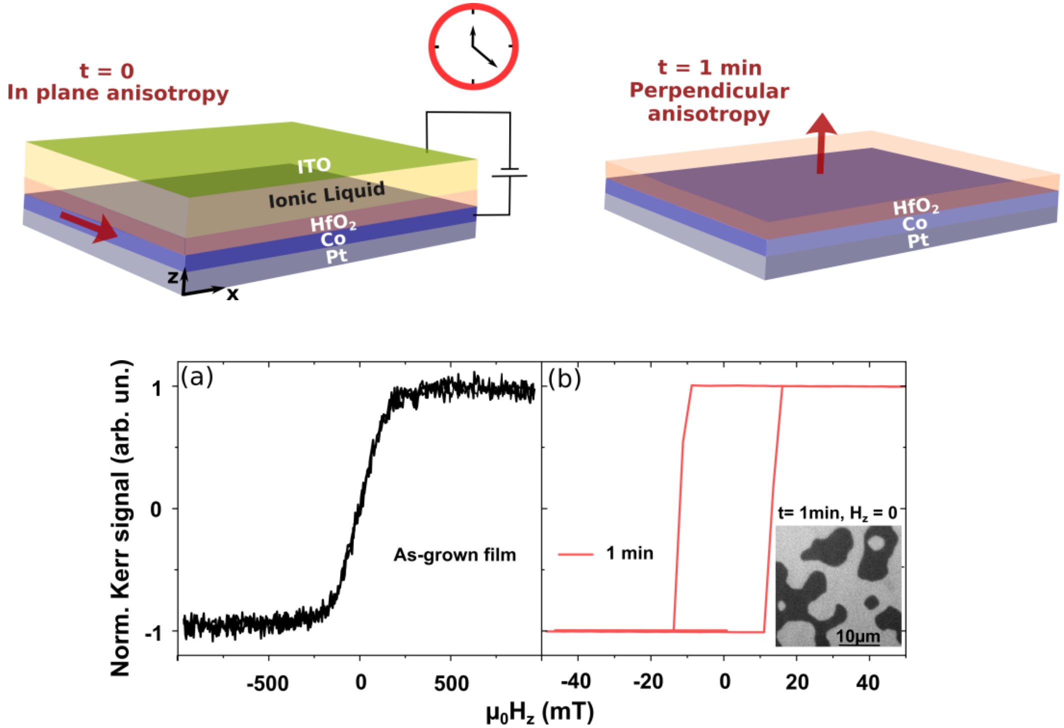


M. Huang *et al*., *Nature Nanotechnology* 16, 981–88 (2021).

H^+ ionic motion allows for defining the dominant sublattice in GdCo devices and gating of the compensation temperature

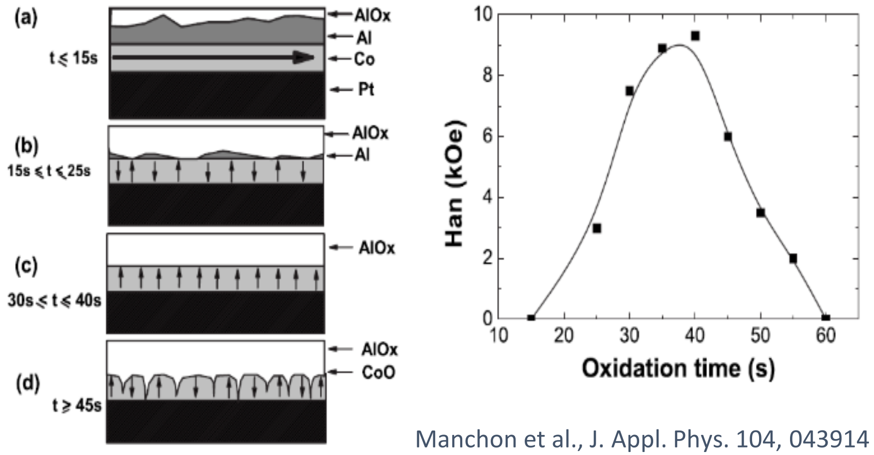
Non-volatile ionic control of DMI in Pt/Co

$Pt/Co/HfO_2$



L. Herrera Diez *et al.*, Phys. Rev. Appl. 12, 034005 (2019)

Pt/Co: surface perpendicular anisotropy



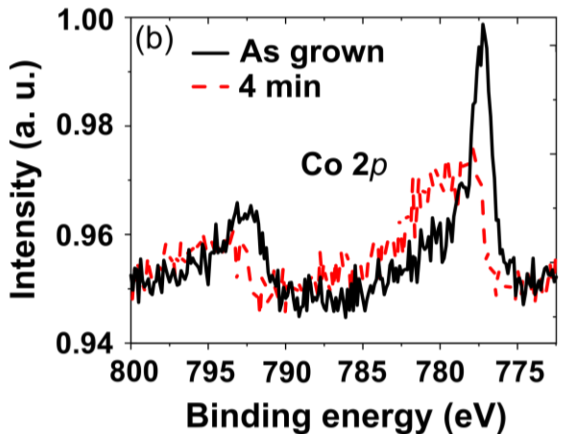
Manchon et al., J. Appl. Phys. 104, 043914 (2008)

Gate-driven oxidation promotes the transition between in-plane and perpendicular anisotropy

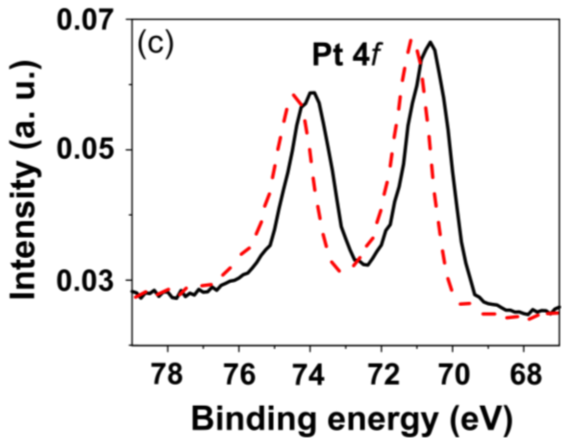
Non-volatile ionic control of DMI in Pt/Co

X-ray Photoelectron spectroscopy
A. Lamperti, CNR Agrate Brianza, Italy

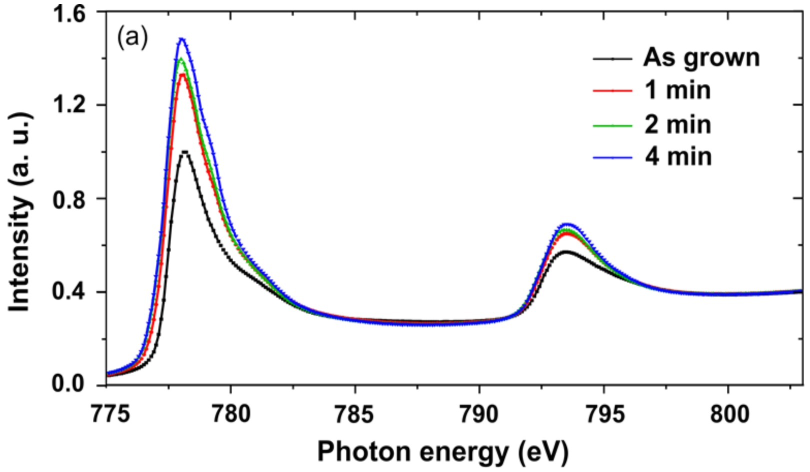
X-ray Absorption spectroscopy
D. Gilbert, NIST/Univ. Tennessee



New oxide peak



Shift to higher binding energies



Gentle oxidation without satellite peaks

Ion migration does not only reach the Co/HfO2 interface but also the Pt/Co interface

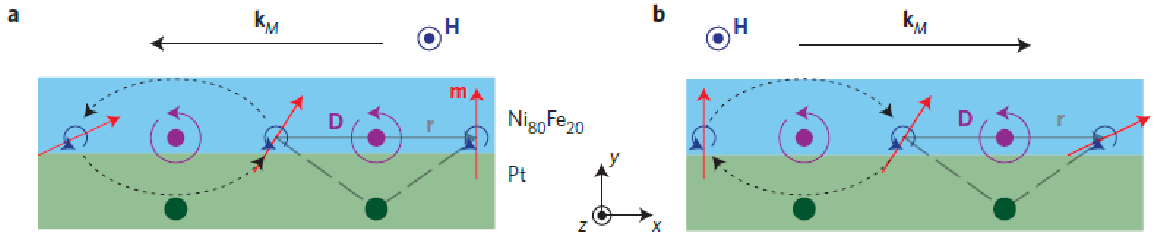
⬇

What happens to DMI?

L. Herrera Diez *et al.*, Phys. Rev. Appl. 12, 034005 (2019)

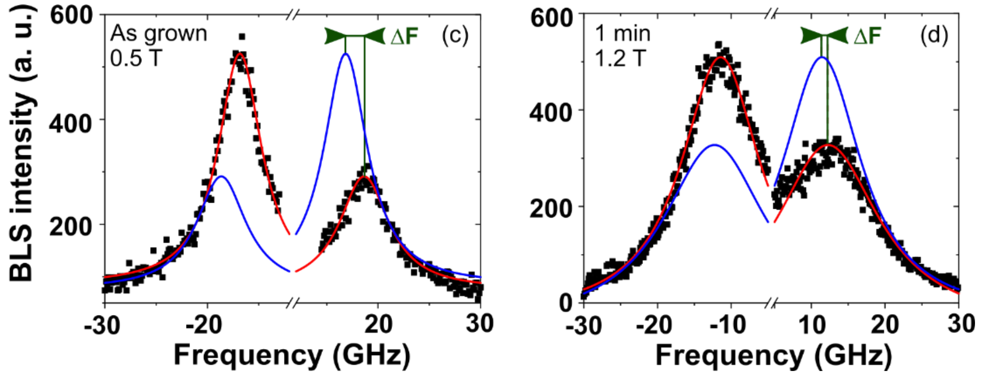
Brillouin light scattering: measure the non-reciprocity of spin wave propagation

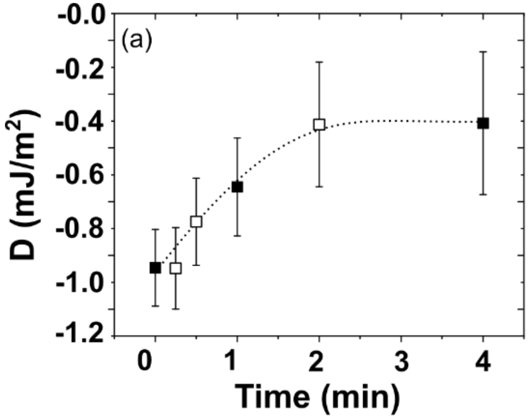
$$\Delta f = \frac{2\gamma}{\pi M_s} D k_{sw}$$



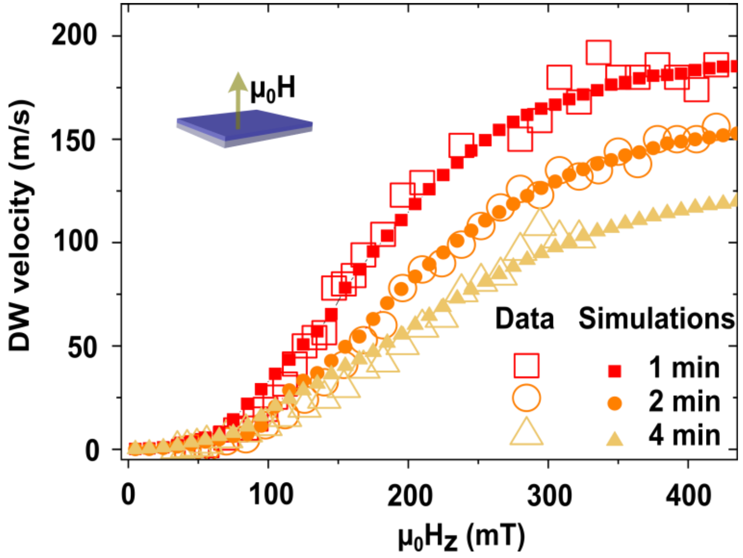
H. Nembach et al., Nat. Physics 11, 825 (2015)

BLS, M. Belmeguenai/Y. Roussigné, Univ. Paris Nord





Mumax E. Martinez University of Salamanca

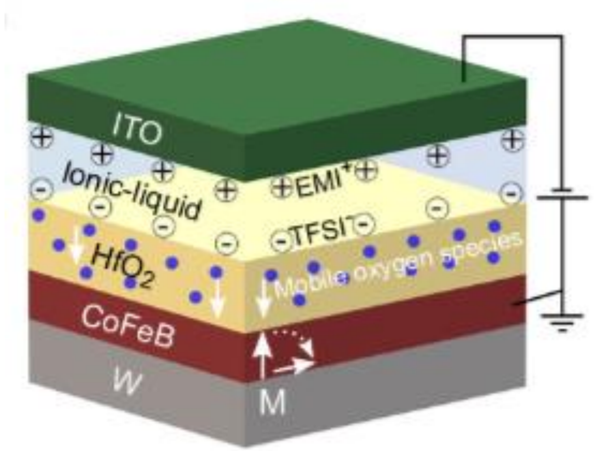


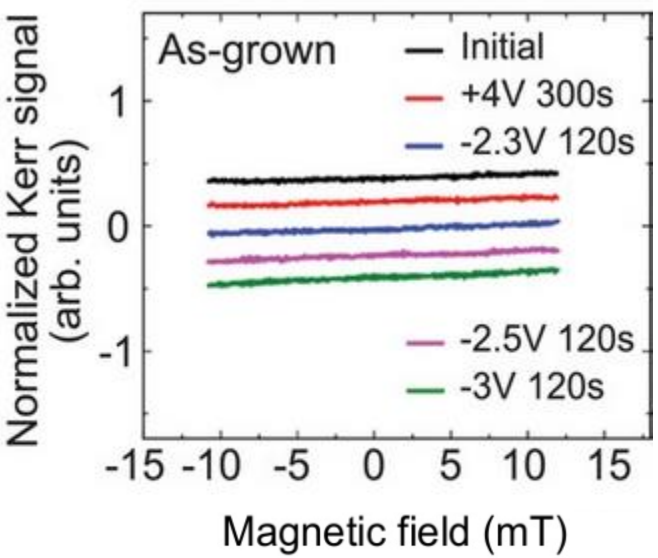
- Decoupling between Pt and Co due to ionic intercalation
- Co interfaces asymmetry affected by changes in chemical composition

L. Herrera Diez *et al.*, Phys. Rev. Appl. 12, 034005 (2019)

Non-volatile control of DMI

W/$Co_{20}Fe_{60}B_{20}$/HfO_2





Controlled oxidation does not induce PMA

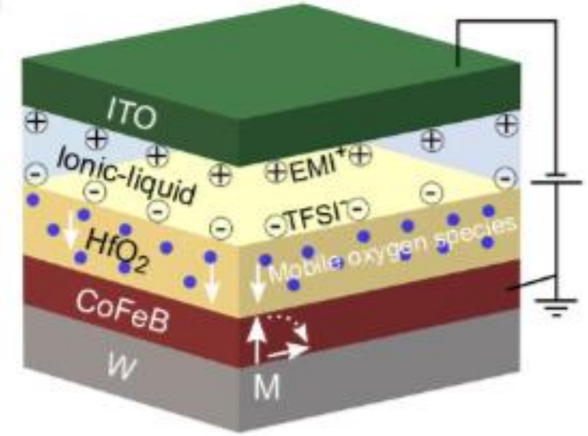
Annealing

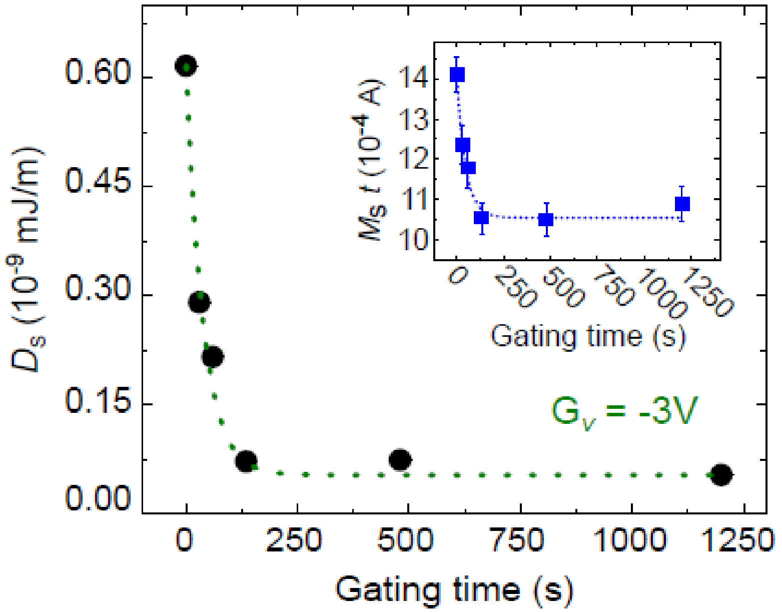
Rohit Pachat
PhD student

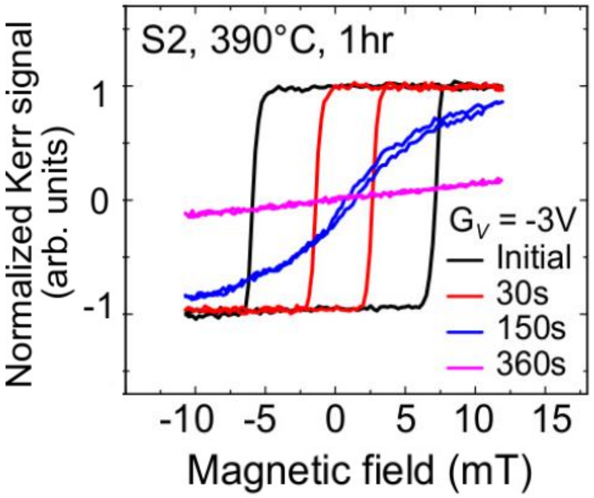
R.Pachat *et al.*, Adv. Mater. Interfaces 2022, 2200690.

Non-volatile control of DMI

$W/Co_{20}Fe_{60}B_{20}/HfO_2$







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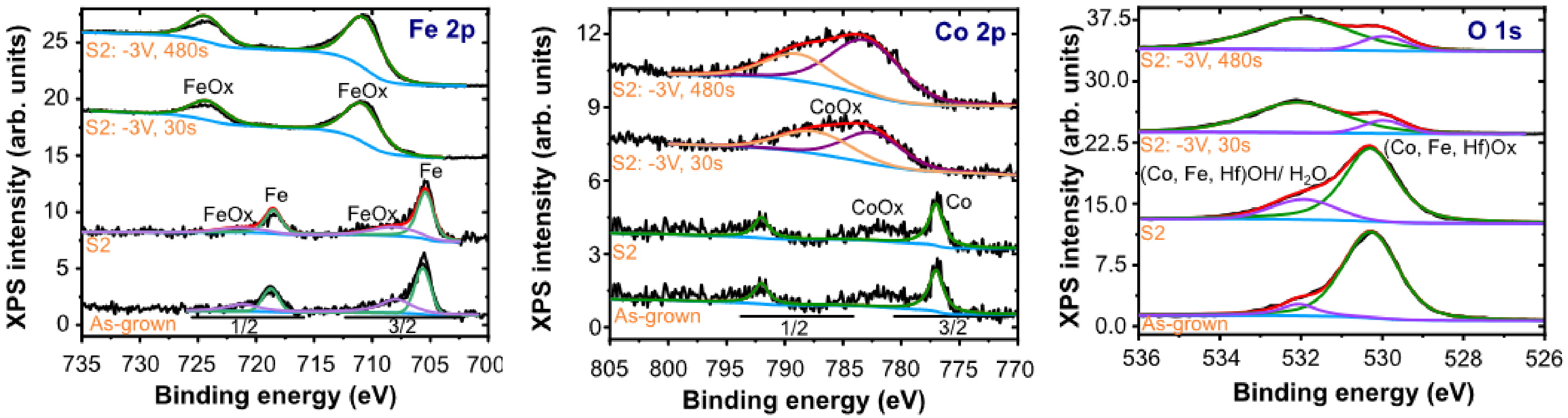
PhD student

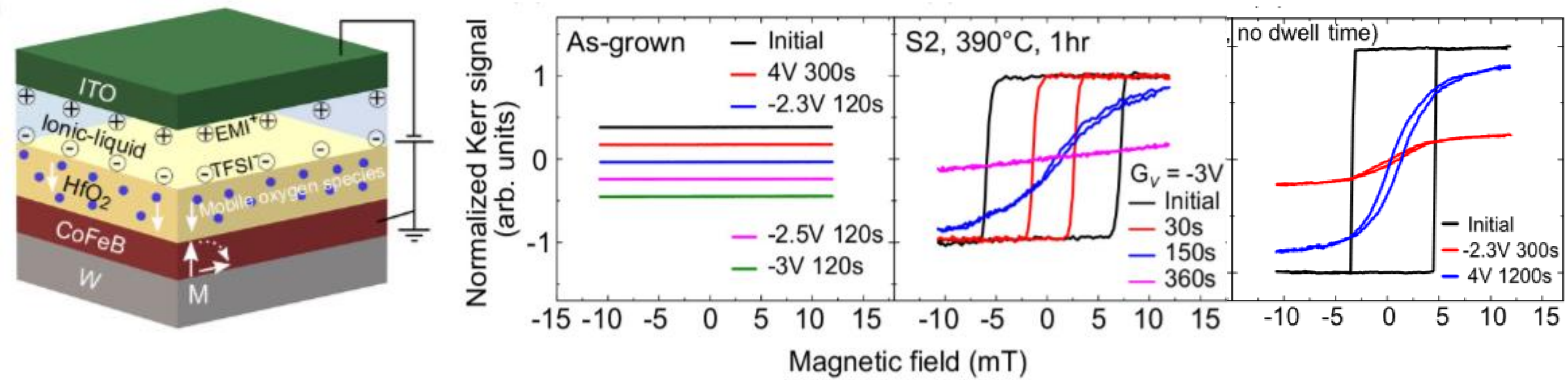
Oxygen introduced by gating shows as a new O peak in XPS



R.Pachat *et al.*, Adv. Mater. Interfaces 2022, 2200690.

Non-volatile control of DMI

As-grown	390°C, 1h	350°C, no dwell time
X No PMA/DMI	✓ DMI	✓ Partially reversible
X No gating effects	X Non reversible	X Very small DMI

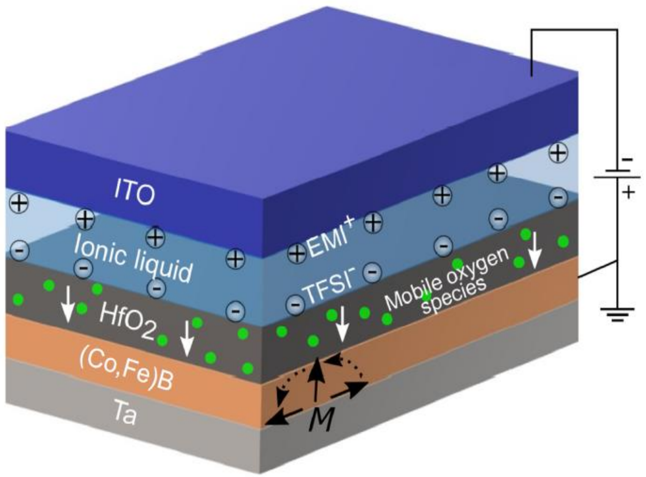


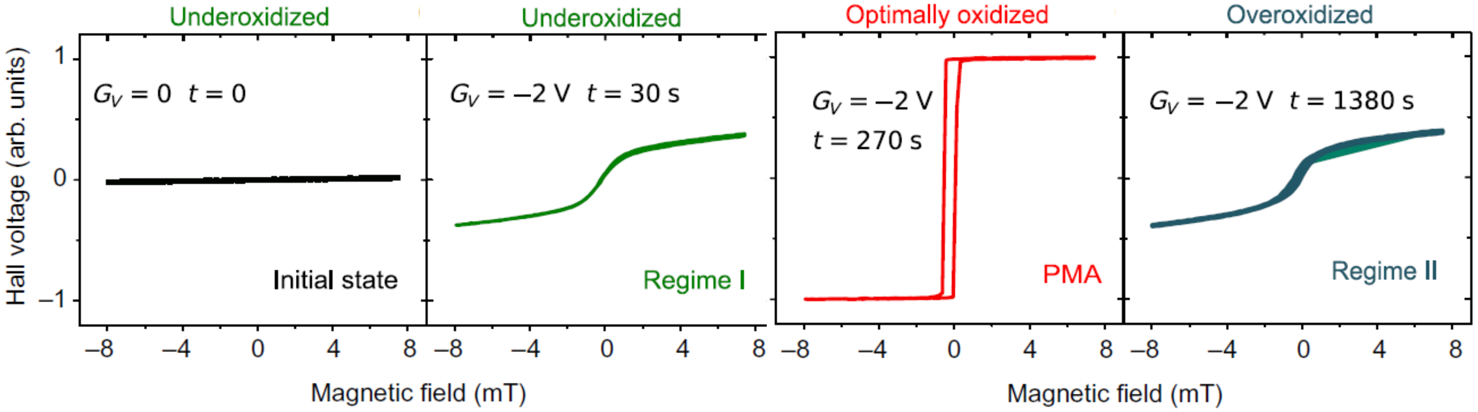
Amorphous system (IPA) → **Annealing** → Crystalized system (PMA) ⇄ Over-oxidation (IPA)

Annealing T dependent

R.Pachat *et al.*, Adv. Mater. Interfaces 2022, 2200690.

Reversibility and multiple magneto-ionic regimes in CoFeB





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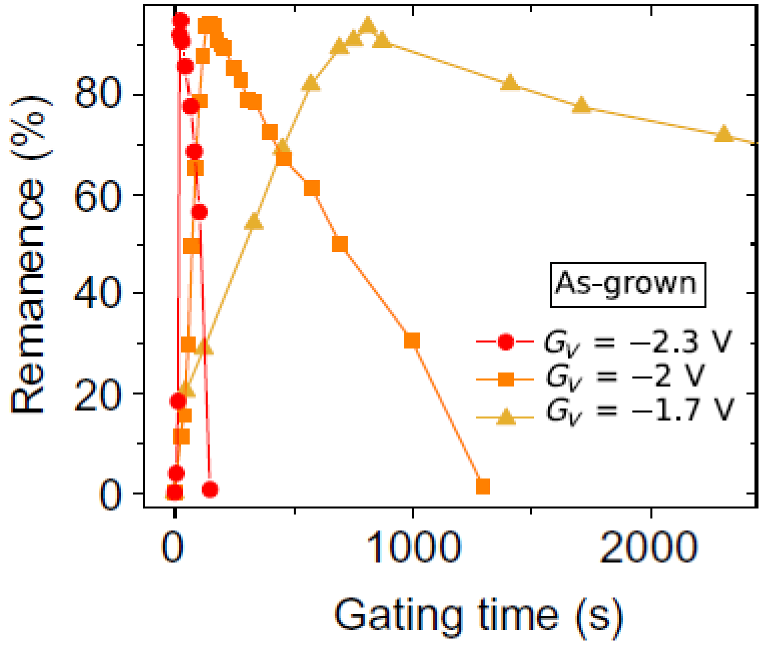
PhD student

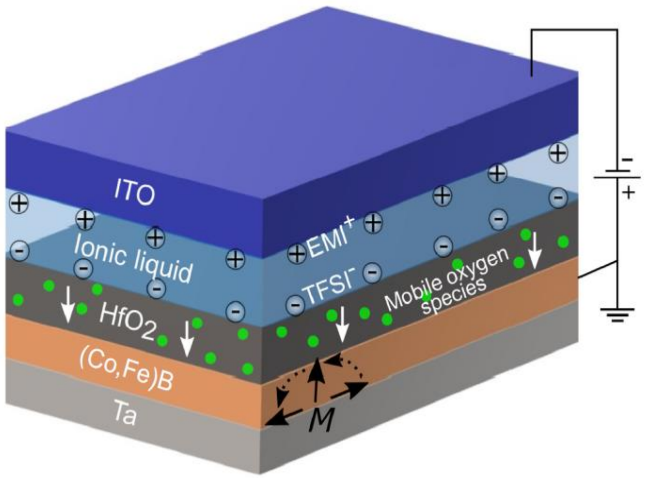


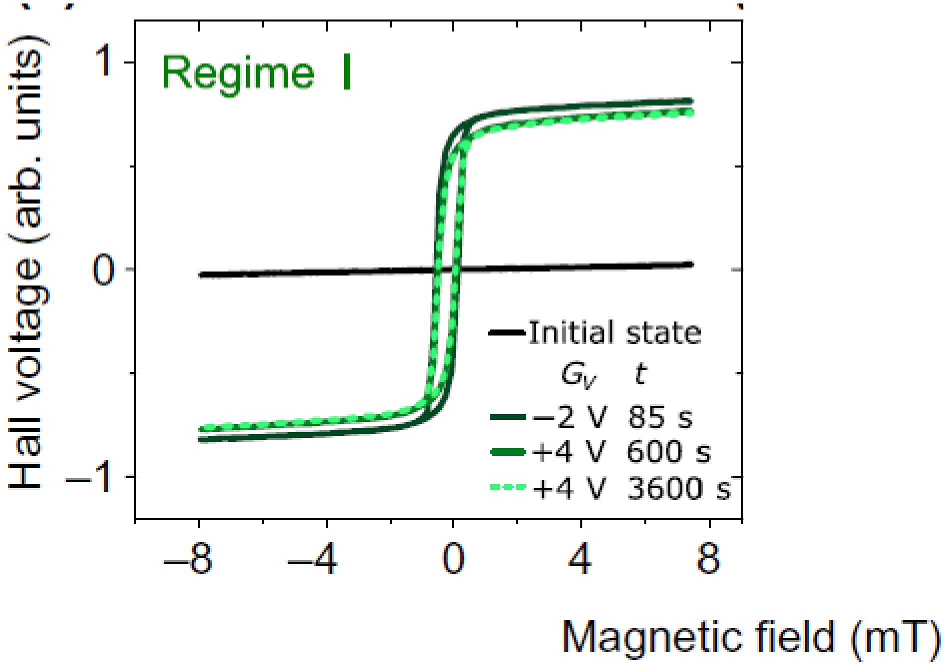
Under-oxidation (IPA) → **Optimum-oxidation (PMA)** → **Over-oxidation (IPA)**

Regime I **Regime II**

R. Pachat, *et al.*. Phys. Rev. Appl. 15, 064055 (2021).

Reversibility and multiple magneto-ionic regimes in CoFeB





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Under-oxidation (IPA) → **Optimum-oxidation (PMA)**

Regime I

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Reversibility and multiple magneto-ionic regimes in CoFeB

Under-oxidation (IPA) → Optimum-oxidation (PMA) ⇄ Over-oxidation (IPA)

Regime I

Regime II

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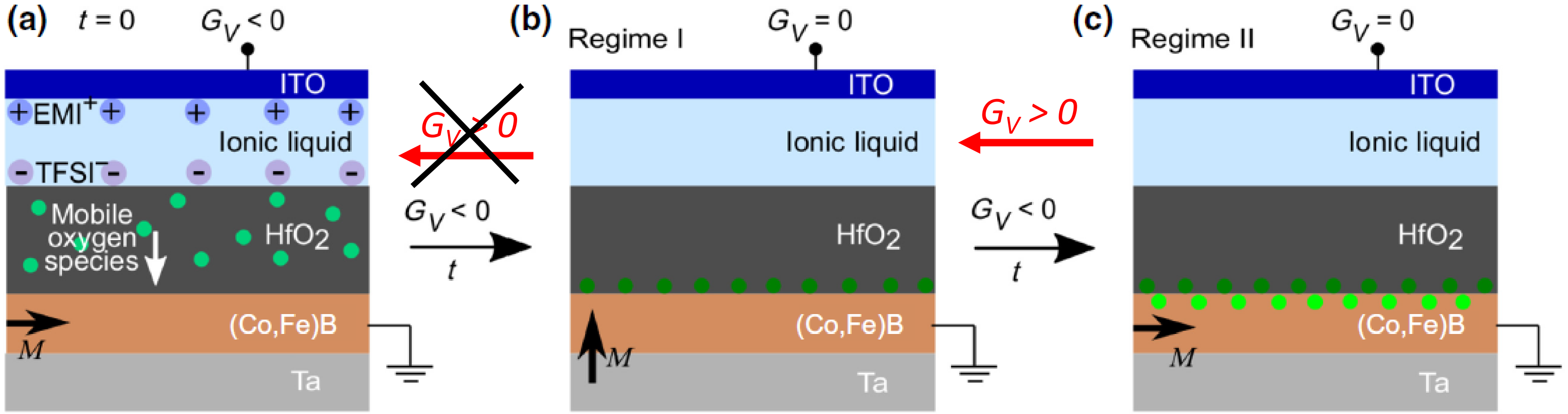


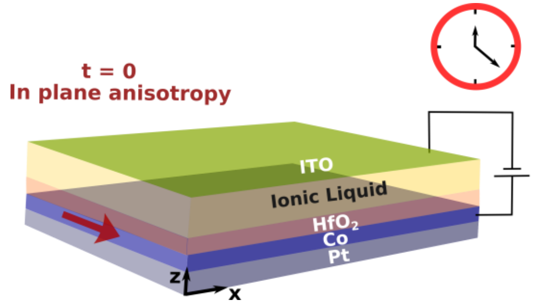
Metallic Fe → Fe^{2+} ⇄ Ox Ox Fe^{2+} Ox Ox → Fe^{3+}

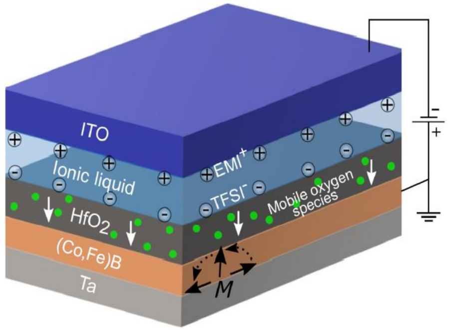
High energy barrier

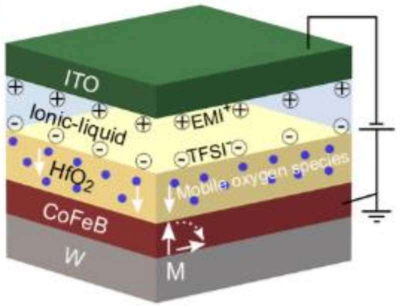
Easily reversible

R. Pachat, *et al.*. Phys. Rev. Appl. 15, 064055 (2021).

Conclusions







- Large changes in PMA and DMI
- Better to use amorphous materials
- Reversibility and cyclability affected by chemical reactions between the ferromagnetic metal and the mobile ions

- **Magneto-ionics has great potential for spintronics applications mostly for energy efficiency and new functionalities**
- **Magneto-ionics means chemistry! Interdisciplinarity is a must**

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