Magneto-ionic control of perpendicular anisotropy in epitaxial Mn_4N films

Silvia Damerio, Teodor Apetrei and Can Onur Avci

Institut de Ciencia de Materials de Barcelona (ICMAB-CSIC), Campus de la UAB, Bellaterra, 08193 Spain

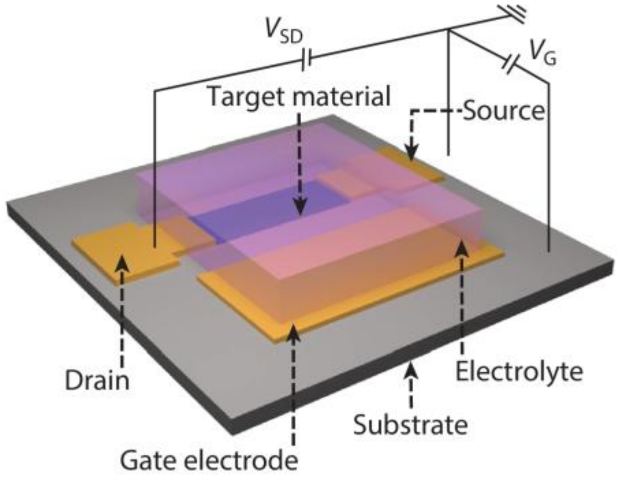
40 ANYS ICMAB
INSTITUT DE CIÈNCIA DE MATERIALS DE BARCELONA
EXCELENCIA SEVERO OCHOA

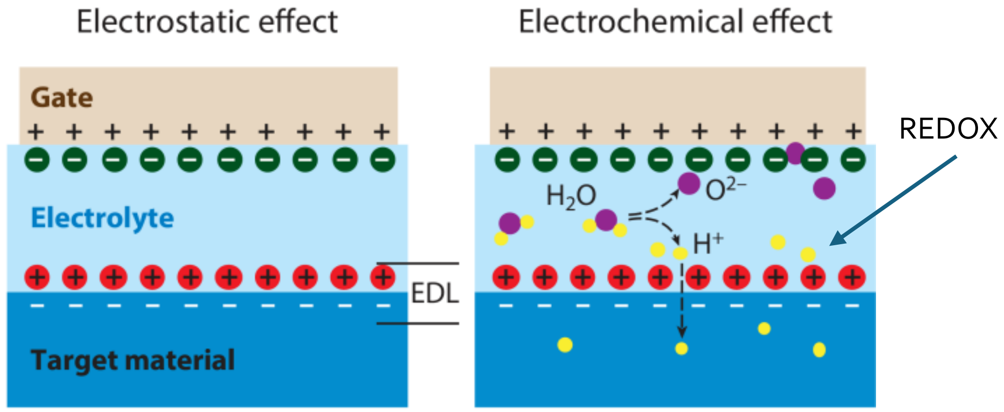
Outline

- ❑ Introduction:
  - Ionic gating
  - Motivation
- ❑ Synthesis and characterization of Mn_4N thin film
- ❑ Tunable magnetic properties in Mn_4N
- ❑ Summary

Ionic gating

Electric-field control of physical properties via gate-induced ion migration

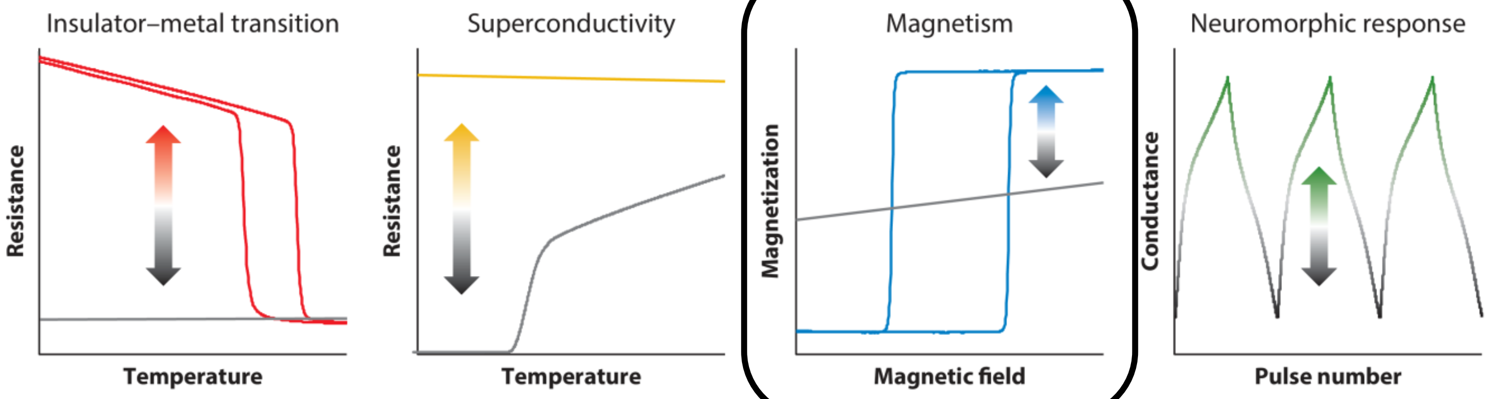




Electrolytes:
- polymers
- ionic liquids
- ion gels
- inorganic solids: SiO_2, Al_2O_3, HfO_2, and GdO_x

Ions:
- anions: O^{2-}, N^{3-}
- cations: Li^+
- protons: H^+

Tunable properties:



Orbital currents

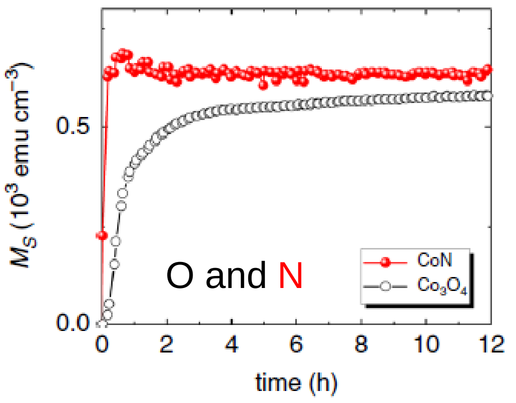
S.D. *et al.* Nano Letters **25**(6), 2181–2187(2025)

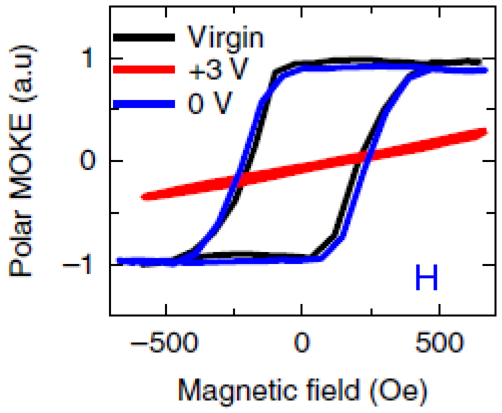
Y. Guan Annu. Rev. Mater. Res. **53**, 25–51 (2023)

Magneto-ionics

Magnetic properties

Co thin films



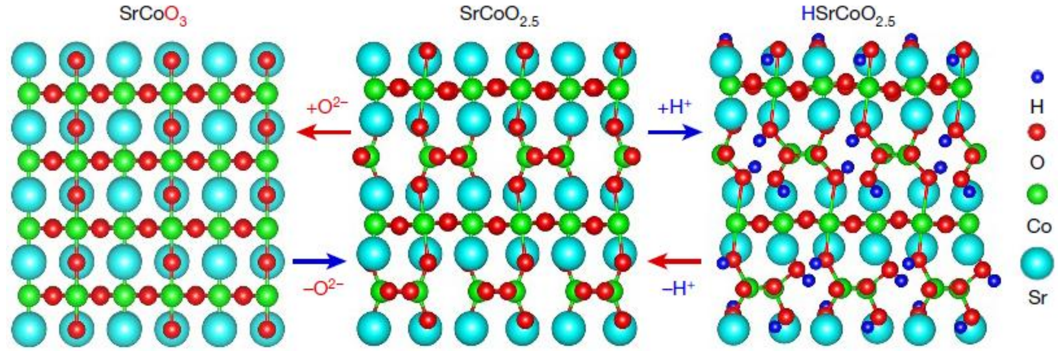


J. de Rojas, *et al.* Nat. Commun. **11**, 5871 (2020)

A.J. Tan, *et al.* Nat. Mater. **18**, 35–41 (2019)

Magnetic order

N. Lu, *et al*. Nature **546**, 124–128 (2017)



Perovskite FM metal

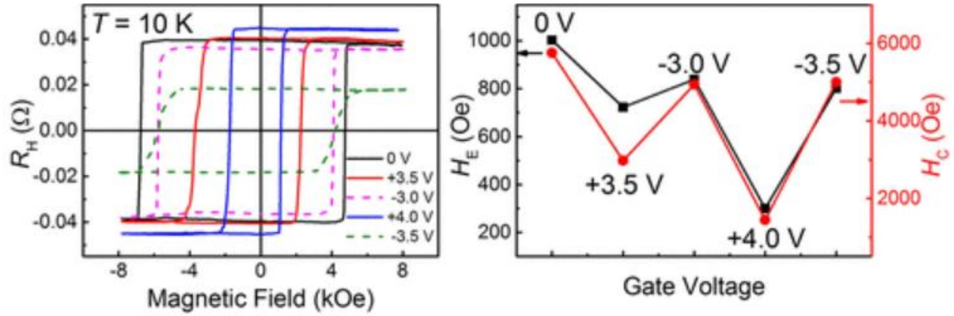
Brownmillerite AF insulator

Perovskite FM insulator

Exchange bias

X. Zhuo, *et al.* J. Phys. Chem. C **120**, 1633–1639 (2016)

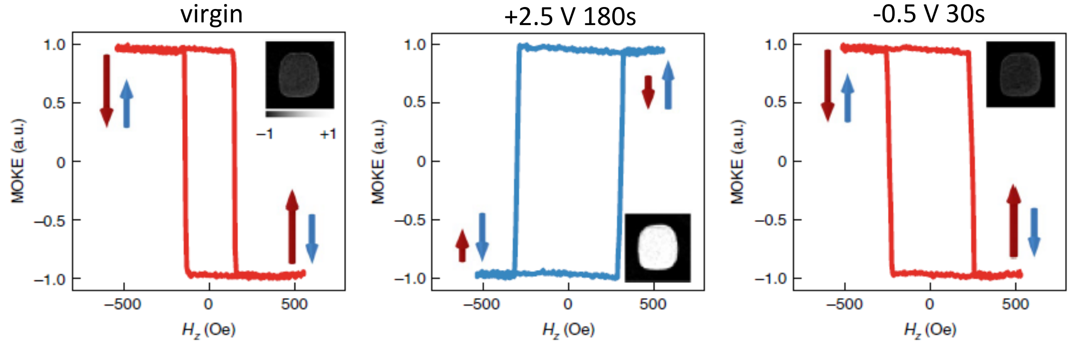
NiOx/Co



Ferrimagnetism

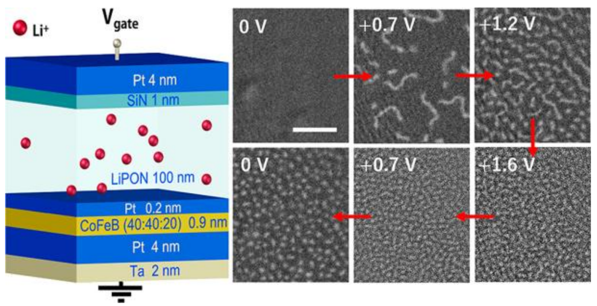
M. Huang, *et al.* Nat. Nanotech. **16**, 981–988 (2021)

GdCo alloys



Skyrmions

$Pt/Co_{40}Fe_{40}B_{20}/Pt$

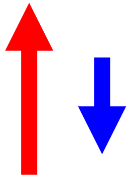


M. Ameziane *et al.*, Nano Lett. **23**, 8, 3167–3173 (2023)

Ferrimagnets for spintronics

Two coupled antiparallel sublattices

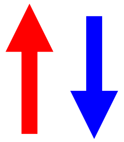
$|M_1| \neq |M_2|$



$M_{\mathrm{net}} \neq 0$

Parameter tuning →

$|M_{\mathrm{TM}}| = |M_{\mathrm{RE}}|$



$M_{\mathrm{net}} = 0$

or $A_{\mathrm{net}} = 0$

compensation

- Efficient SOT switching
- Ultrafast optical magnetization switching
- AFM-like fast domain wall motion
- Strong magnon-mode coupling

SK. Kim *et al.*, Nat. Mater. **21**, 24–34 (2022)

Rare-earth 3d metal alloys

- Amorphous, atomically disordered
- Tunable via composition
- Electrically conductive

✗ Easily deteriorated

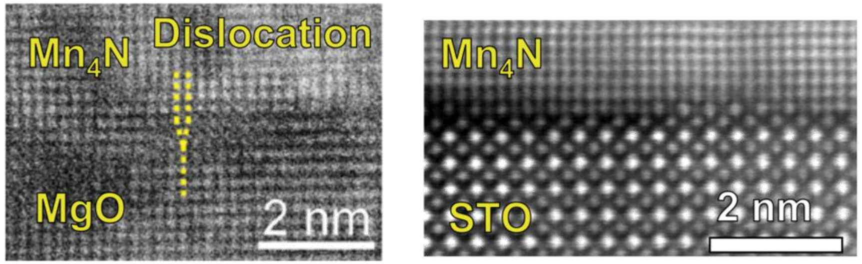
Oxide compounds

- Crystal, atomically ordered
- Low damping
- Tunable via strain and composition

✗ Electrically insulating

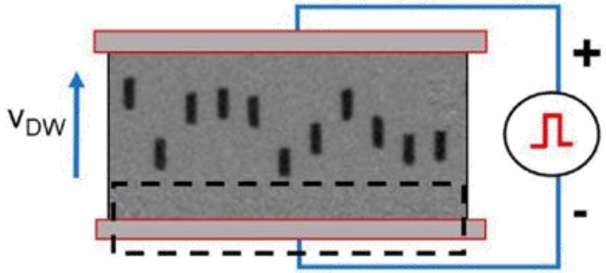
Mn_4N

✔ Crystal, atomically ordered
✔ Electrically conductive
✔ Tunable via strain and composition
✔ Rare-earth free



Perpendicular magnetic anisotropy

X. Shen *et al.*, Appl. Phys. Lett.**105**, 072410 (2014)



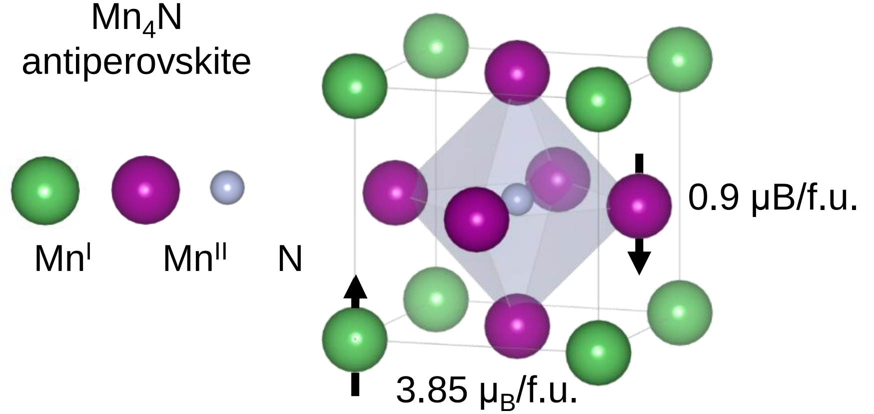
STT: DWs speed up to 3 km/s

S. Gosh et al., Nano Lett. **21**, 2580-2587 (2021)
T. Gushi et al., Nano Lett. **19**, 8716–8723 (2019)

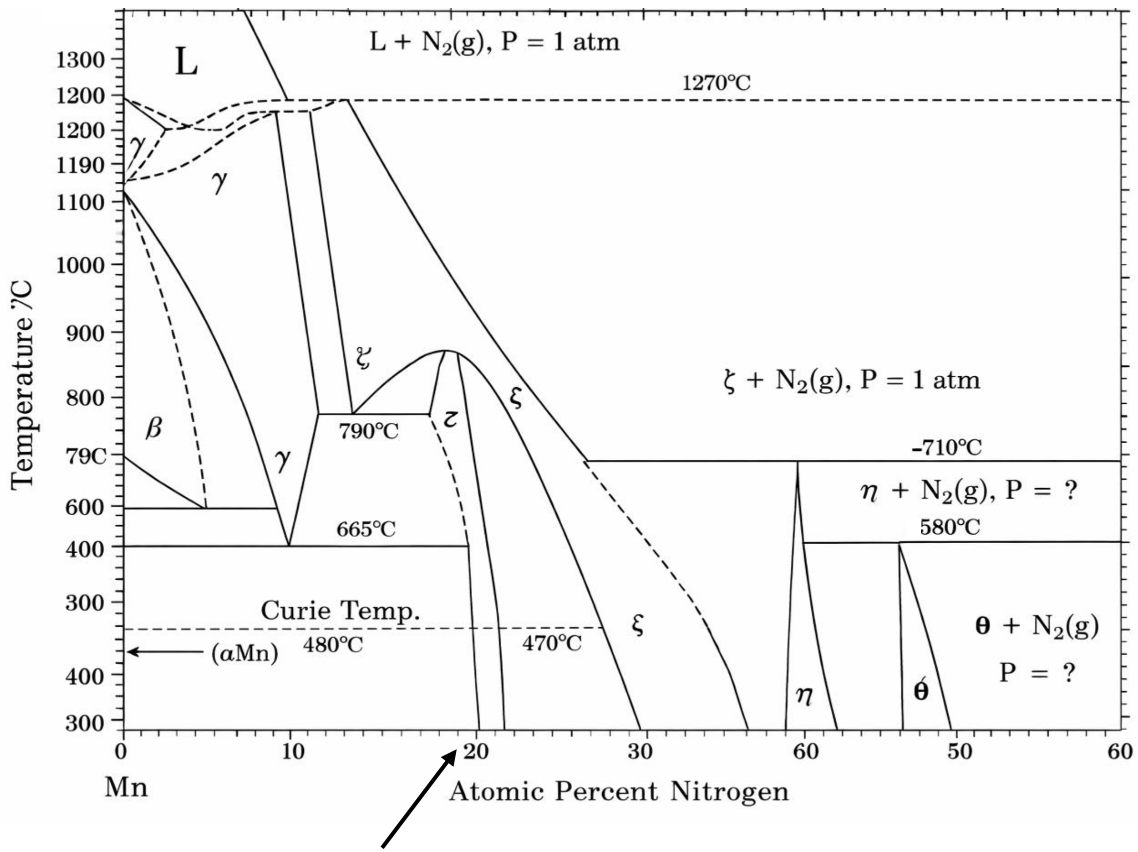
Outline

- Introduction:
  - Ionic gating
  - Motivation
- Synthesis and characterization of Mn_4N thin film
- Tunable magnetic properties in Mn_4N
- Summary

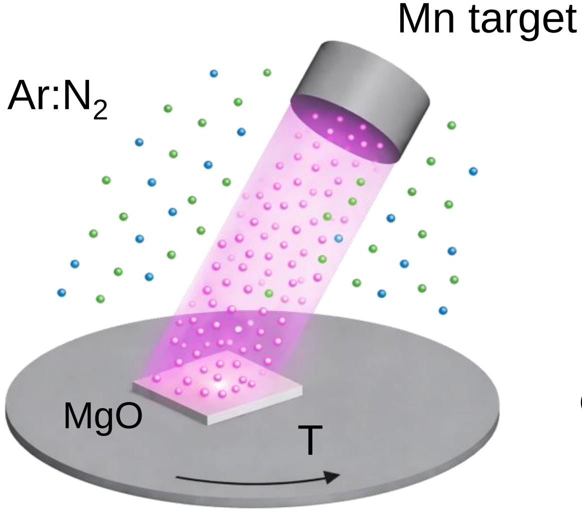
Mn_4N thin films



Z. Zhang, W. Mi, J. Phys. D: Appl. Phys. **55**, 013001 (2022)

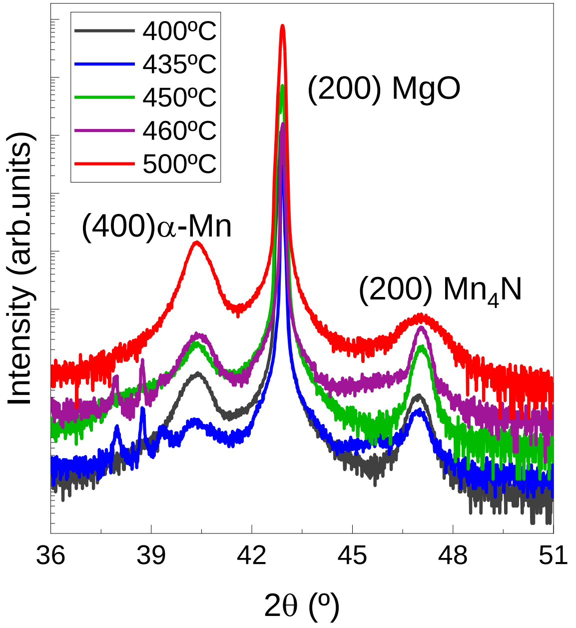


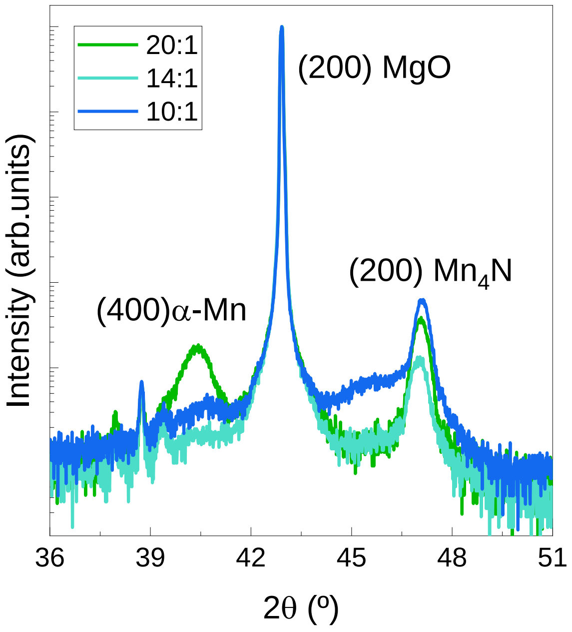
Mn_4N only ferrimagnetic phase



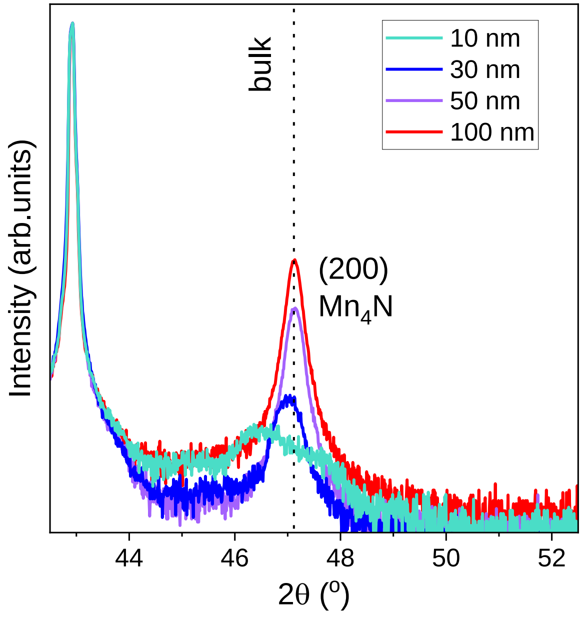
Growth methods:
- MBE on STO/MgO
- (Facing target) sputtering on STO/MgO
- Ionically driven synthesis on Si

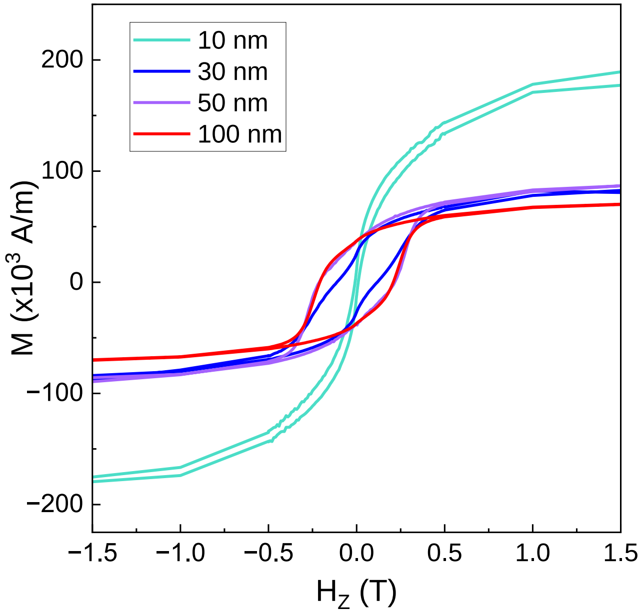
Growth optimization

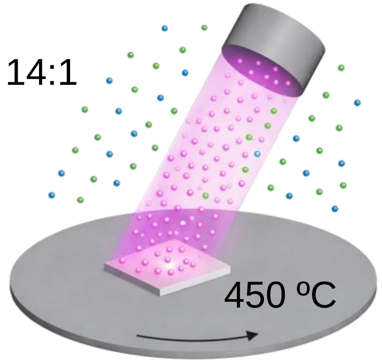




- Control of teperature
- Control of Ar:N_2 ratio

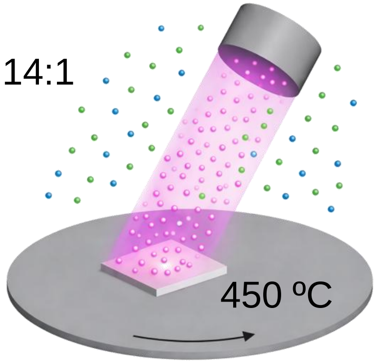


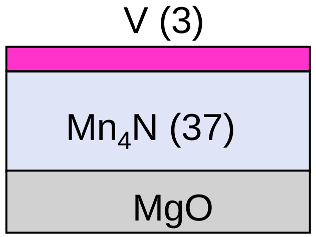


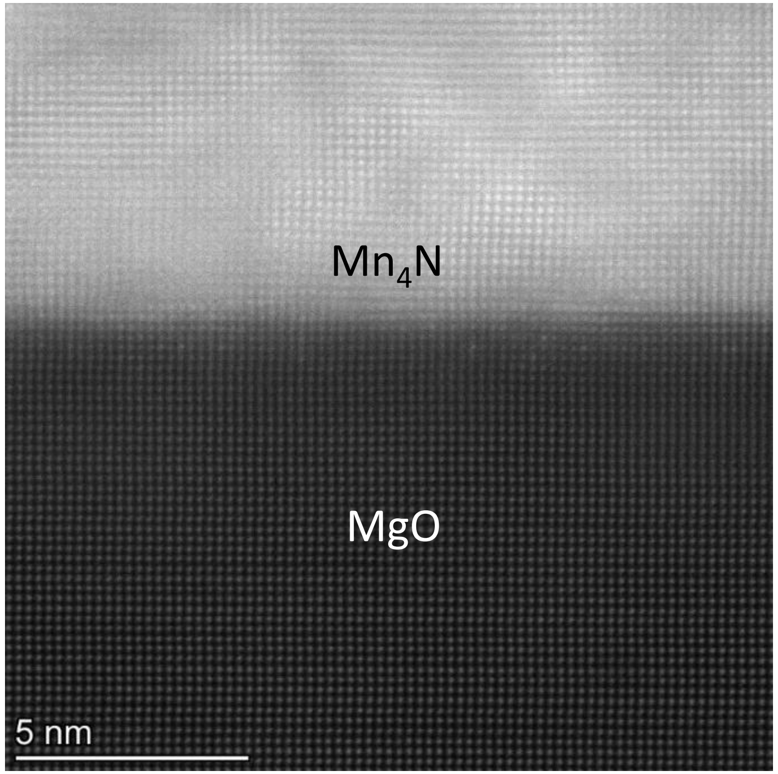


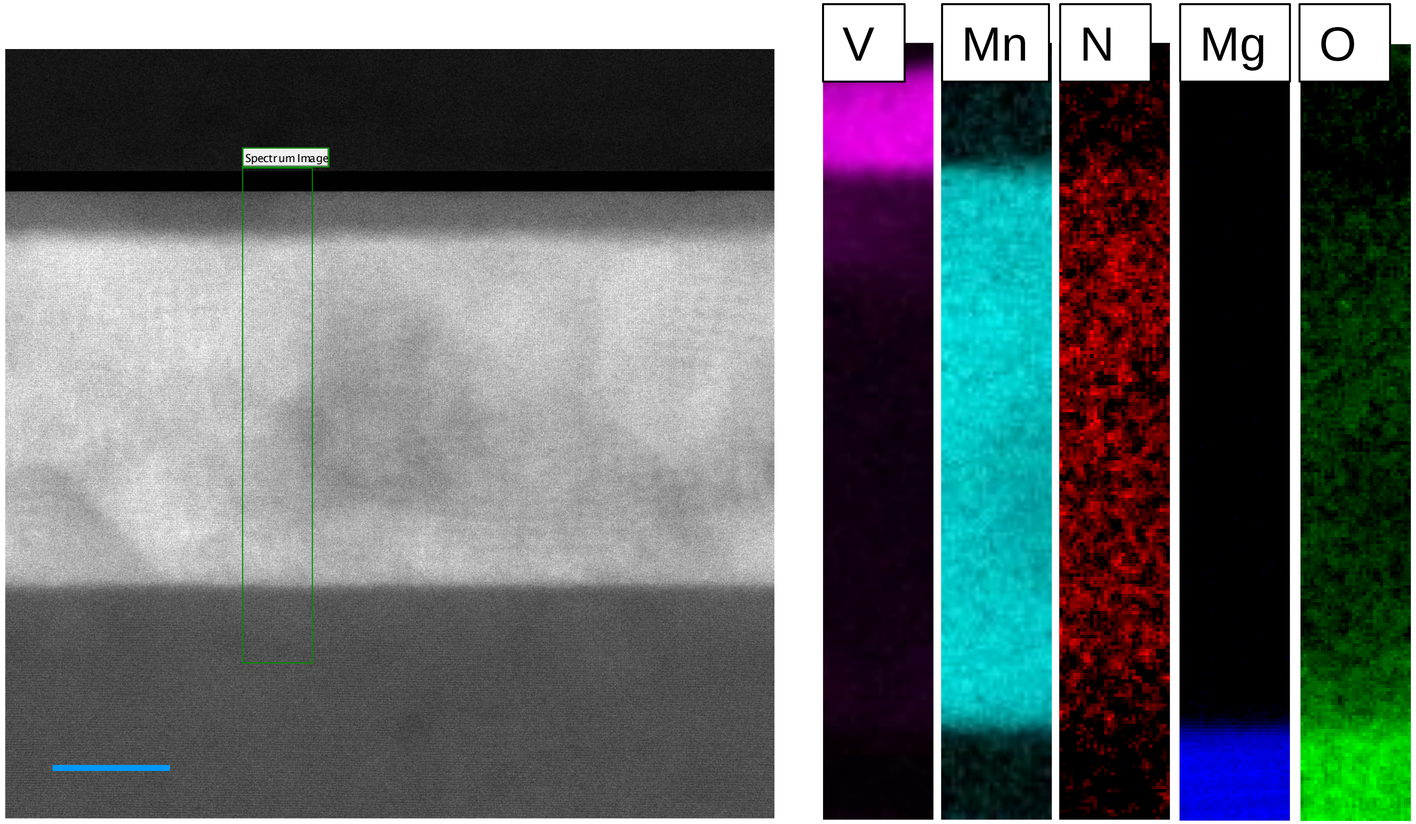
- Strain relaxes with increasing thickess
- PMA doesn’t scale with strain

Mn_4N thin films

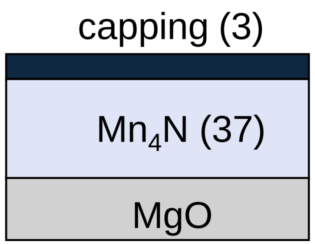


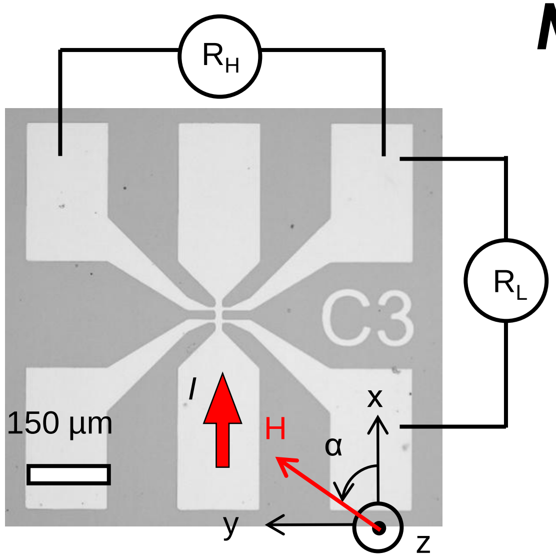






Magneto-transport

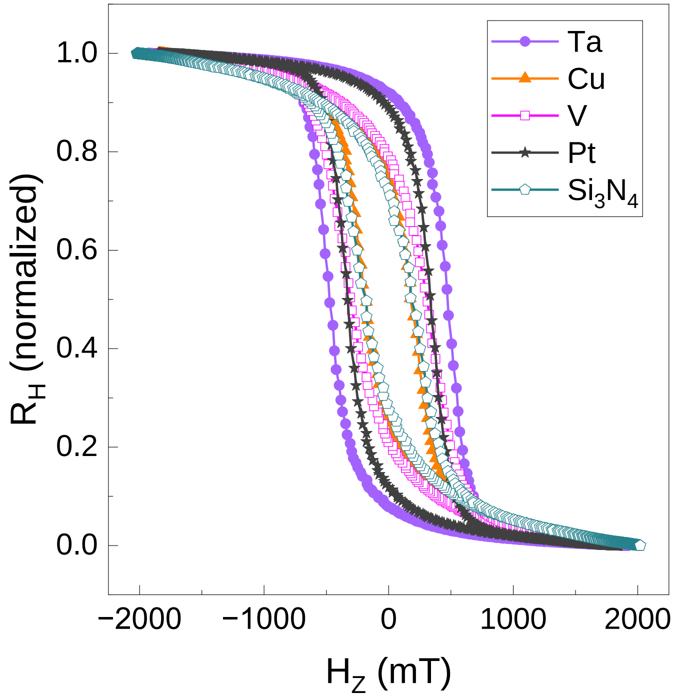


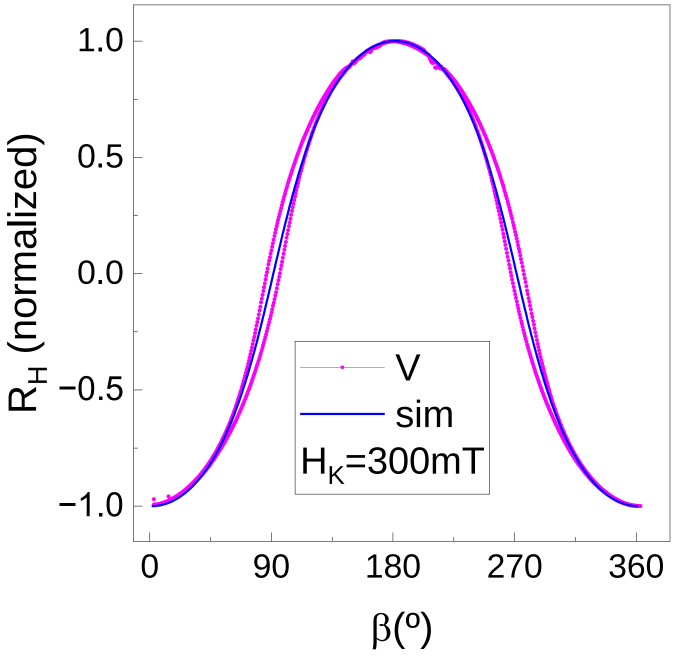


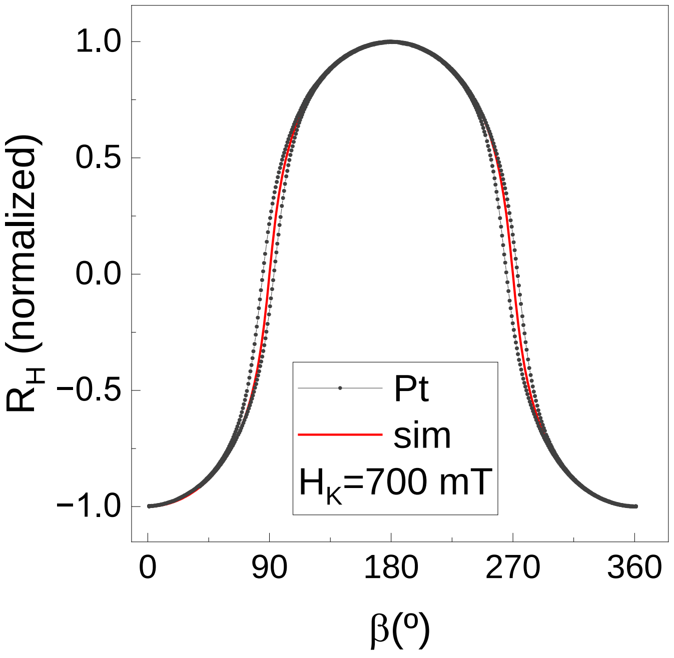
$$R_H = R_\omega + R_{2\omega} + \ldots$$

$$R_H^\omega = R_{AHE} \cos\theta + R_{PHE} sin^2\theta * sin2\alpha$$

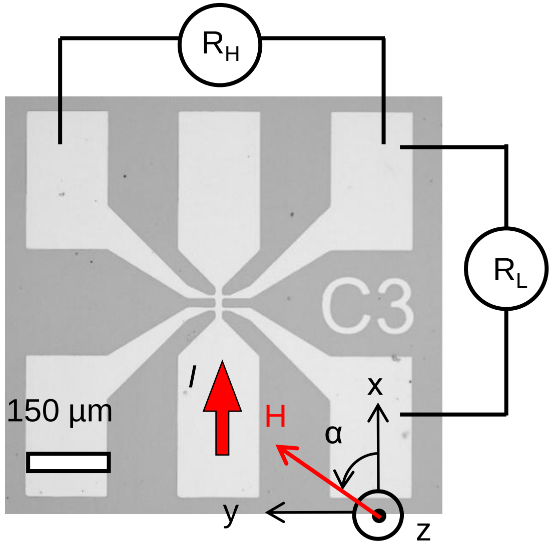
- The magnetic anisotropy depends on capping layer material

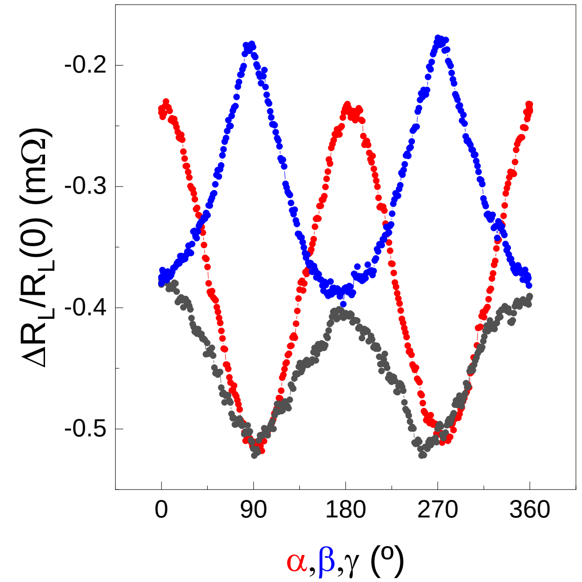






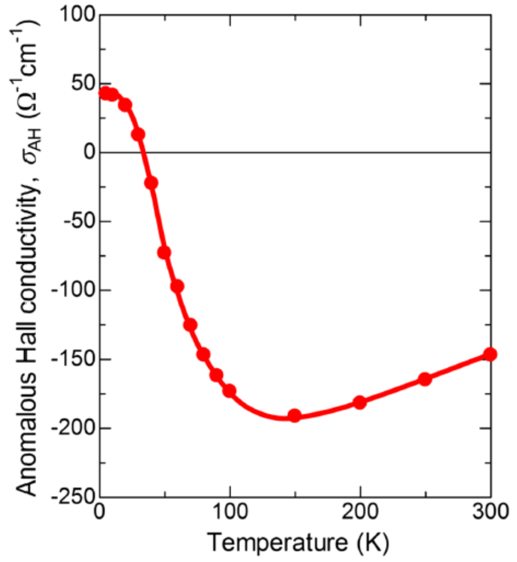
Magneto-transport



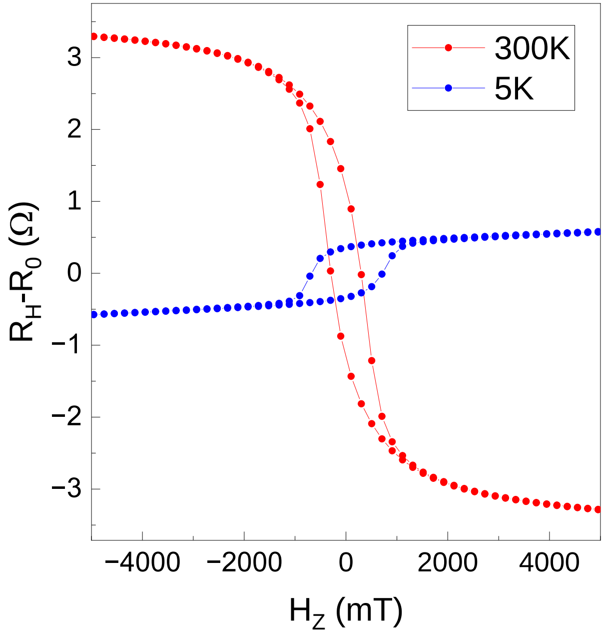


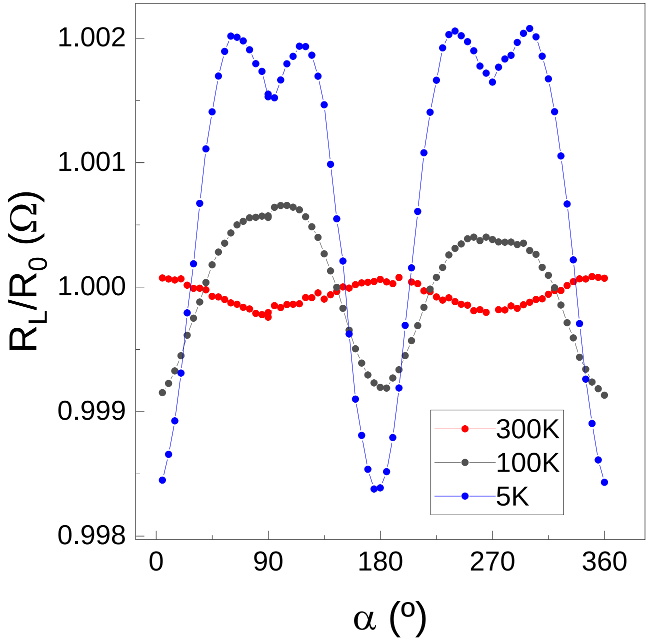
$$R_L^{\omega} = R_0 + R_{AMR} sin^2\theta * cos2\alpha$$

- Magnetic hard axis along z
- Low temperature anomalies of Hall conductivity caused by the splitting of the 3*d* orbitals in tetragonal crystal field



K. Kabara *et al.*, AIP Advances **7**, 056416 (2017)



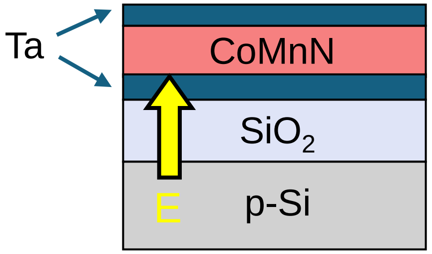


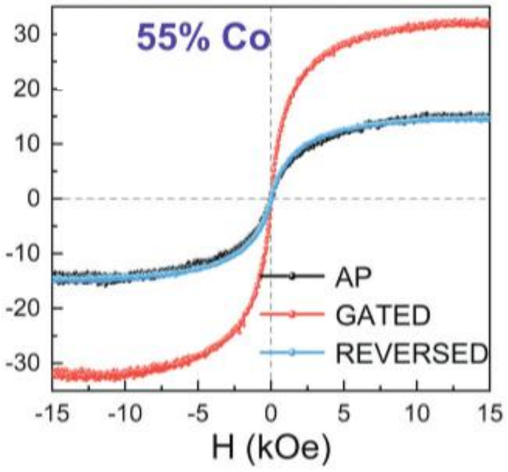
Outline

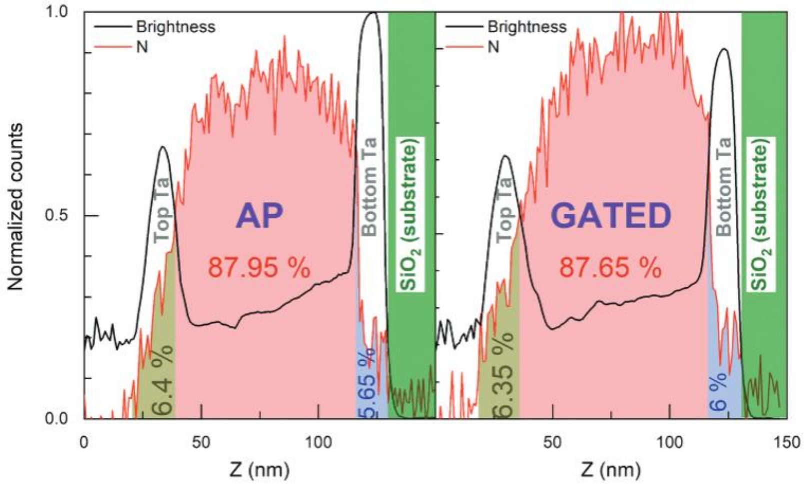
- ❑ Introduction:
  - Ionic gating
  - Motivation
- ❑ Synthesis and characterization of Mn_4N thin film
- ❑ Tunable magnetic properties in Mn_4N
- ❑ Summary

Magnetoionics

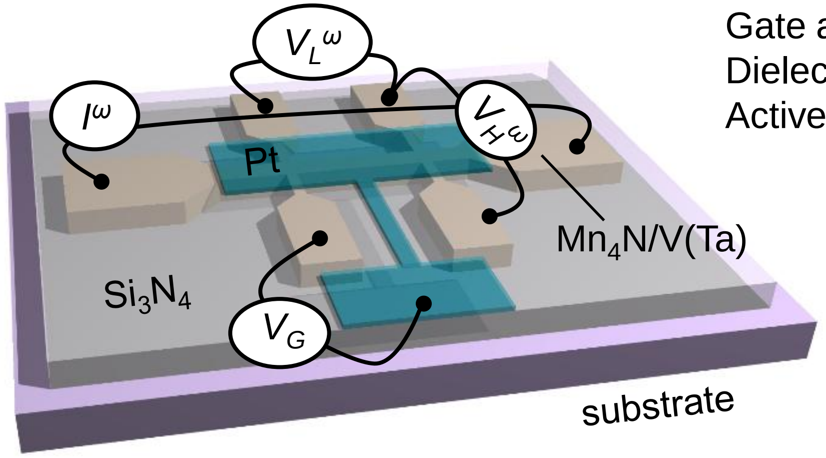
- continuous film
- amorphous FM
- in-plane M
- Linear N^{3-} front



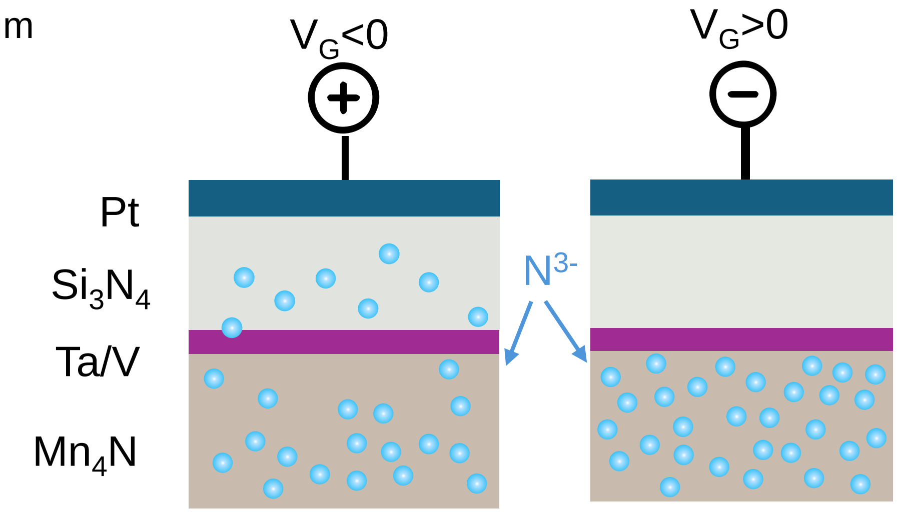




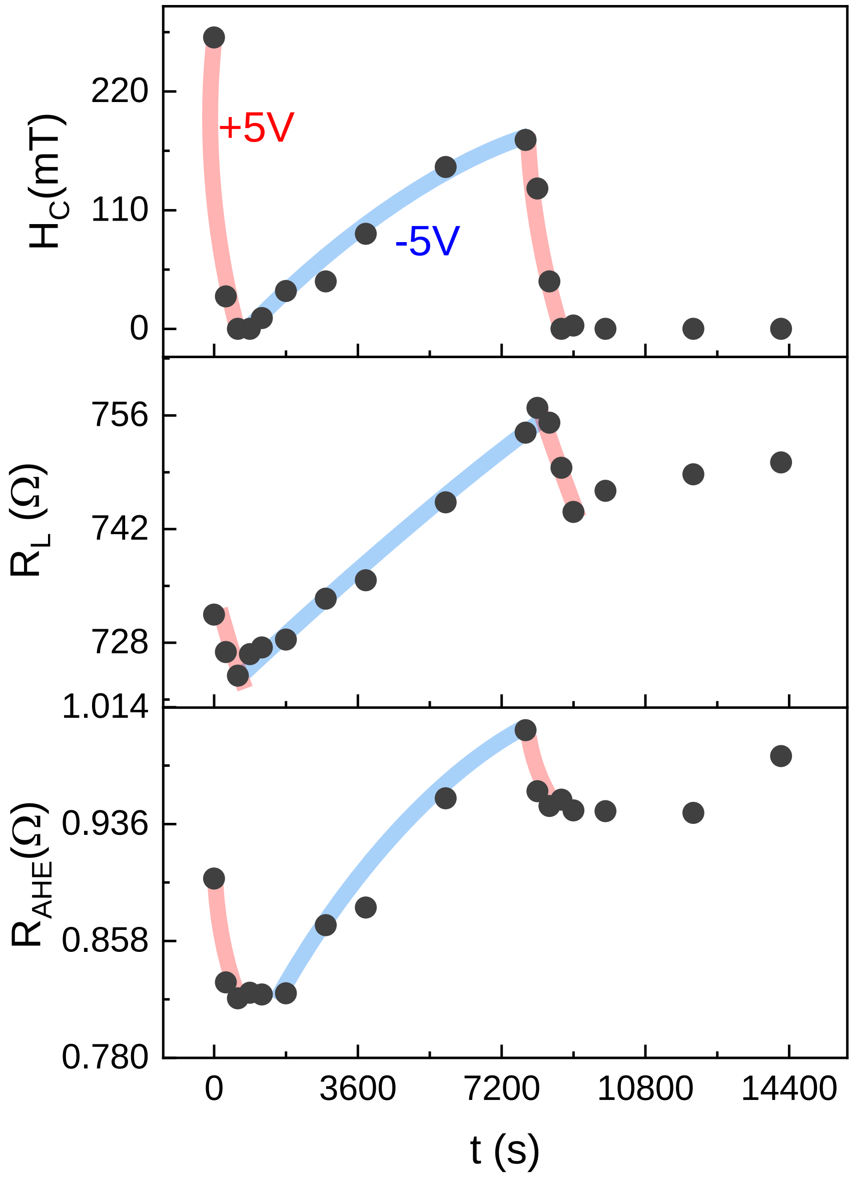
N. López-Pintó, *et al.* Adv. Funct. Mater. **34**, 2404487 (2024)



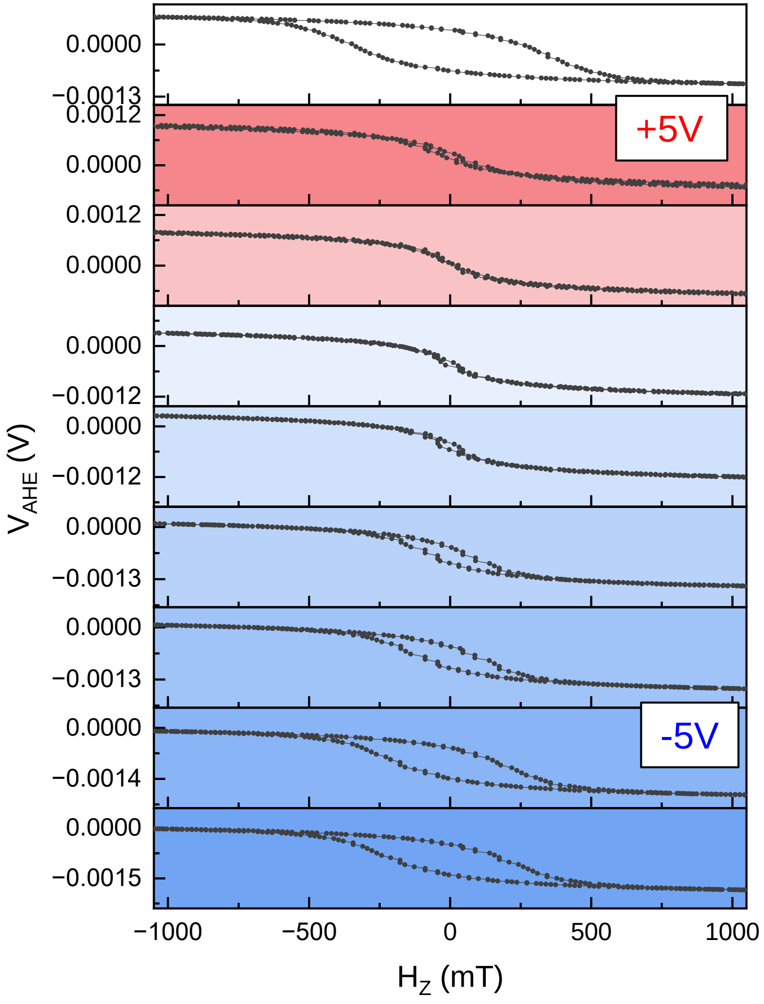
Hall bar dimensions: 30 µm x 7.5 µm
Gate area: 30 µm x 20 µm
Dielectric thickness: 25-50 nm
Active layer: 15-40 nm

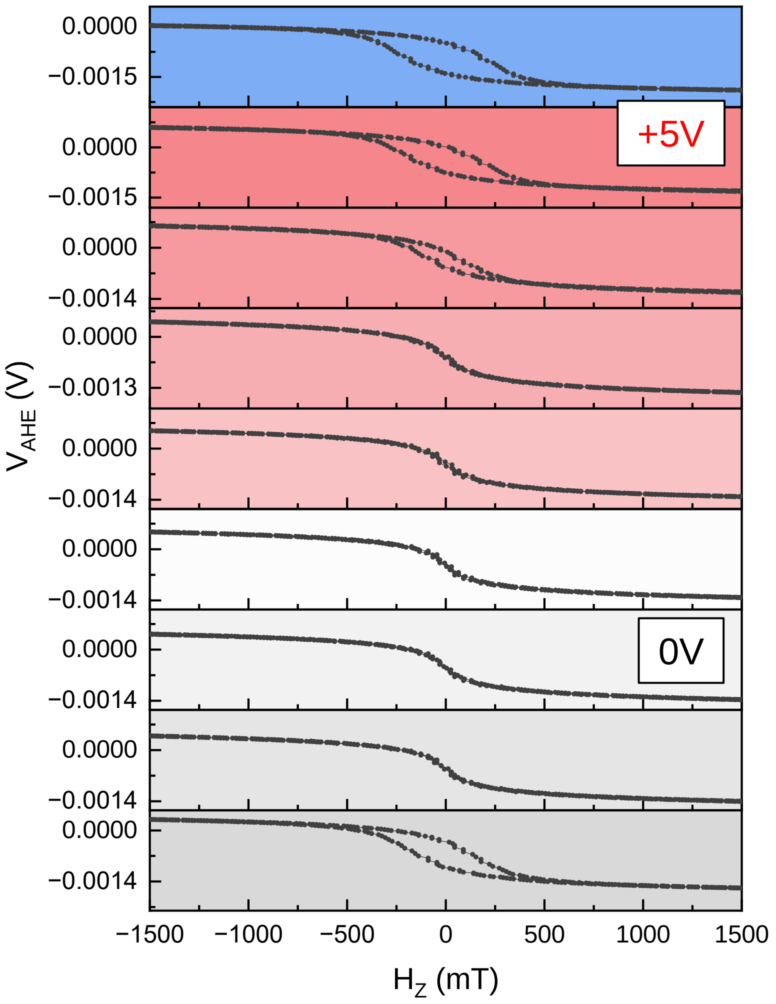


Magnetoionics



AHE evolution during gating

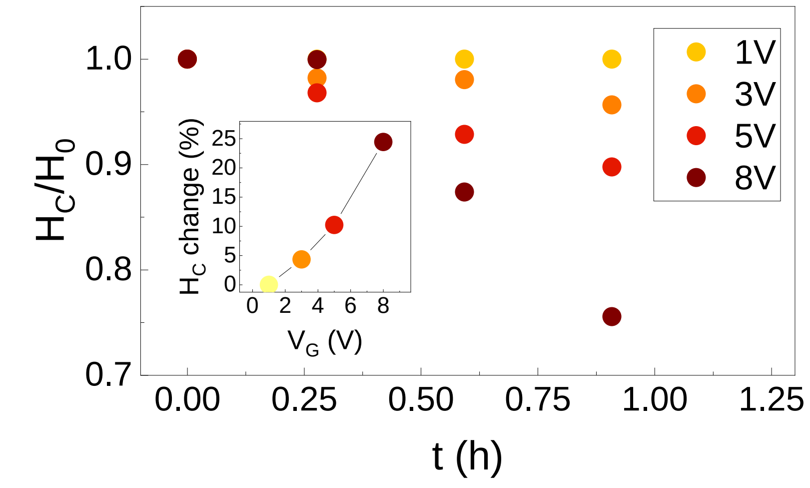


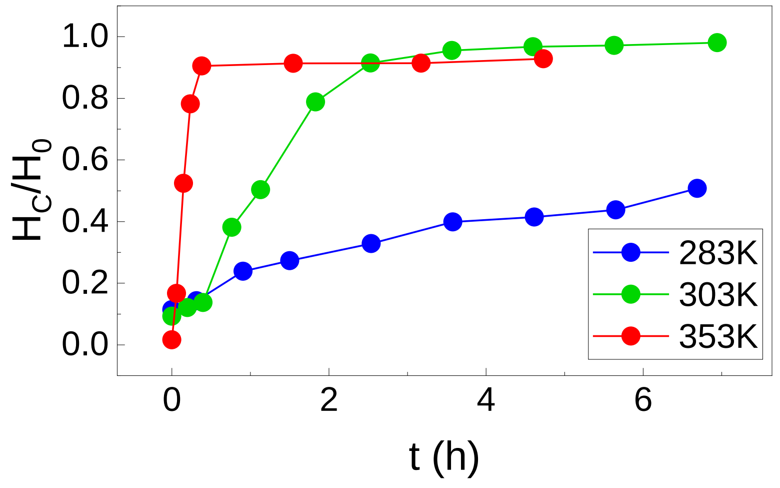


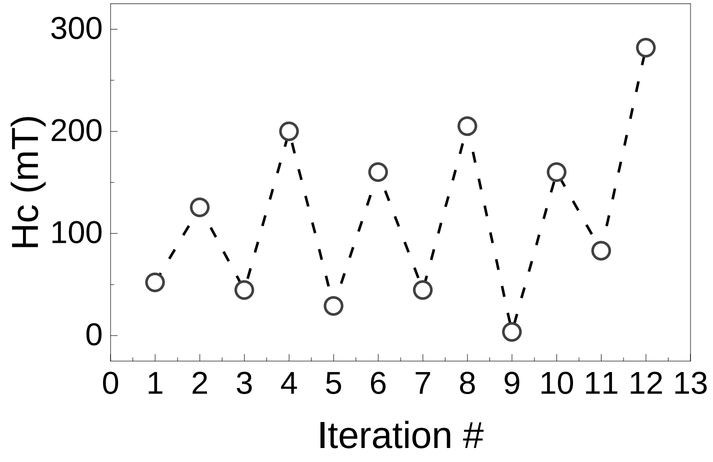
Magnetoionics

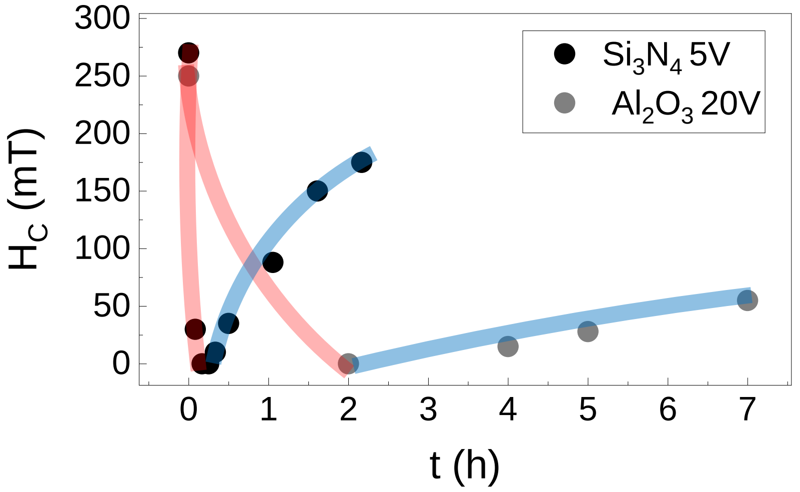
Mott and Gurney hopping mechanism for Ion motion in solids: $i = zecav\, e^{\frac{-E_a}{kT}} e^{\frac{azeE}{2kT}}$

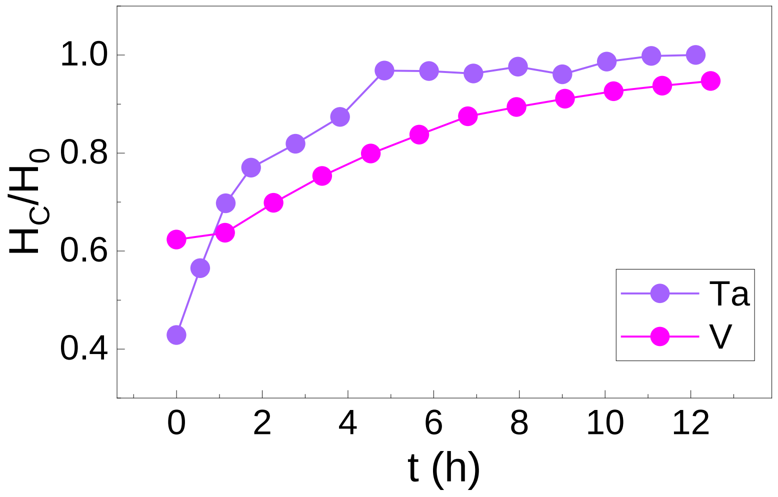
R. Waser *et al.* Adv. Mater. **21**, 2632–2663 (2009)



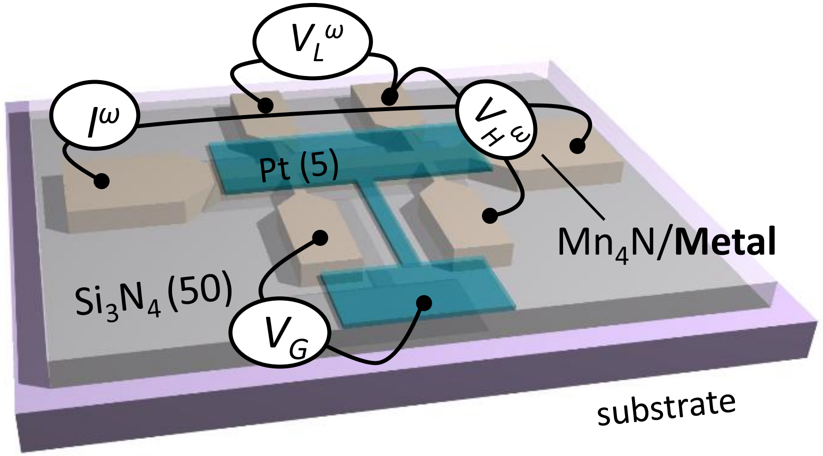




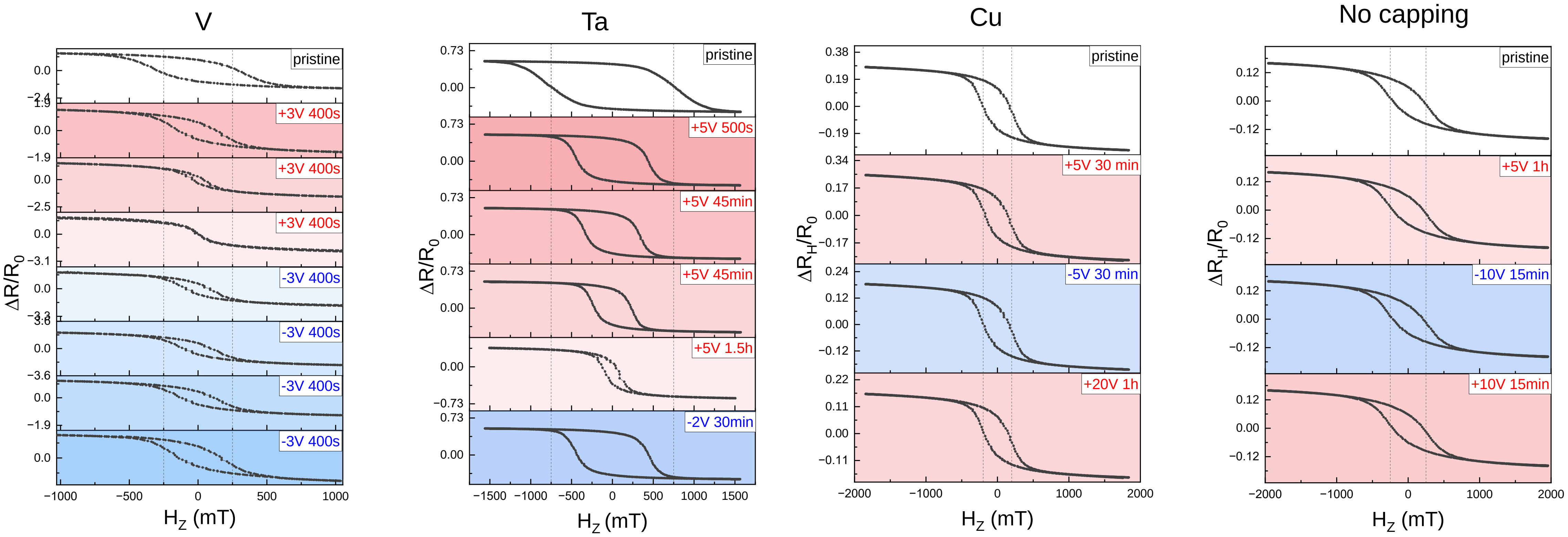




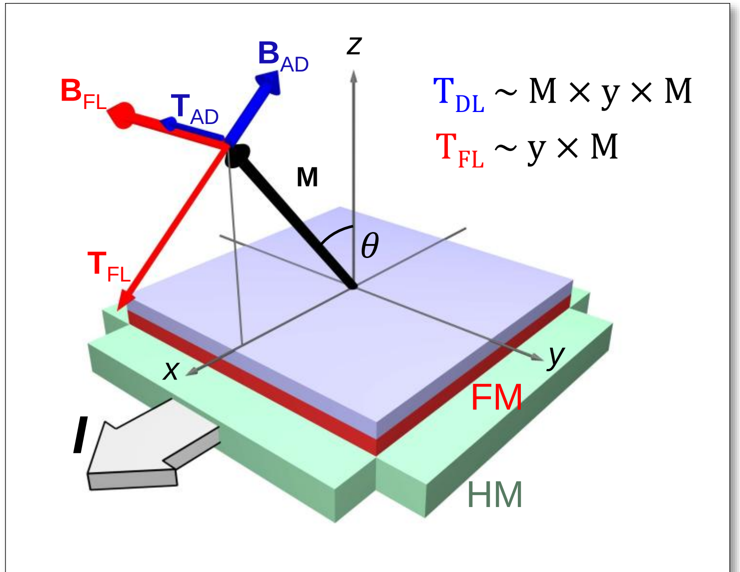
Magnetoionics



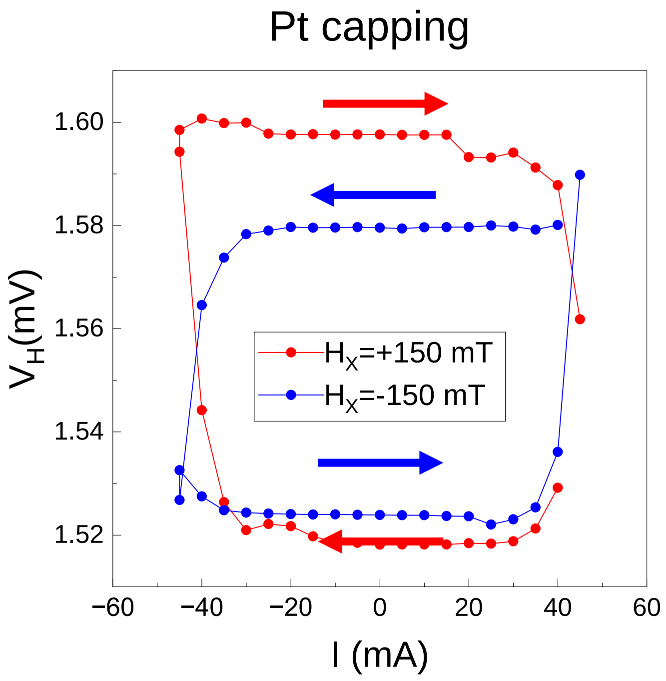
Metal	Reactivity with N_2	M-N Bond Strength	Works
Cu	Very Low	~150–250 kJ/mol	✗
Pt	Low	~200–500 kJ/mol	✗
Ti	High	~400–600 kJ/mol	✗
V	Very High	~400–700 kJ/mol	✓
Ta	Very High	~500–800 kJ/mol	✓

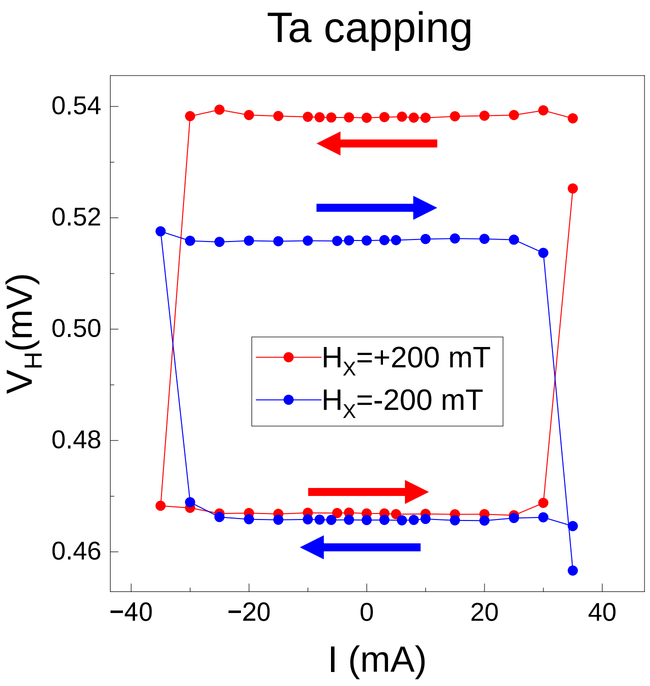


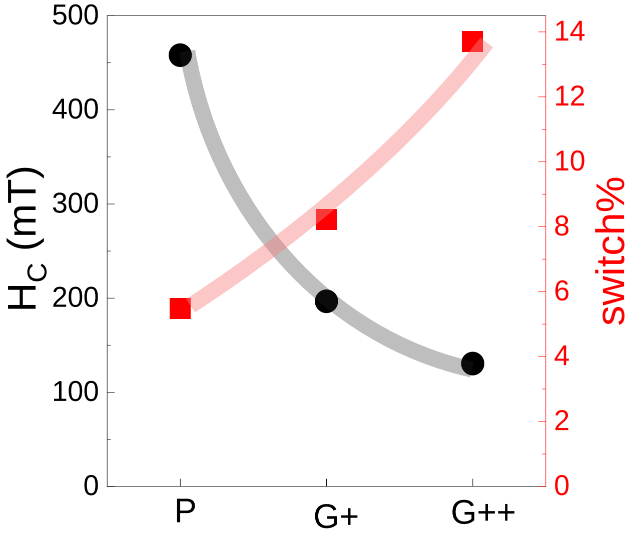
Tunable spintronic devices



C.O. Avci, *et.al.,* Phys. Rev. B **90**, 224427 (2014)



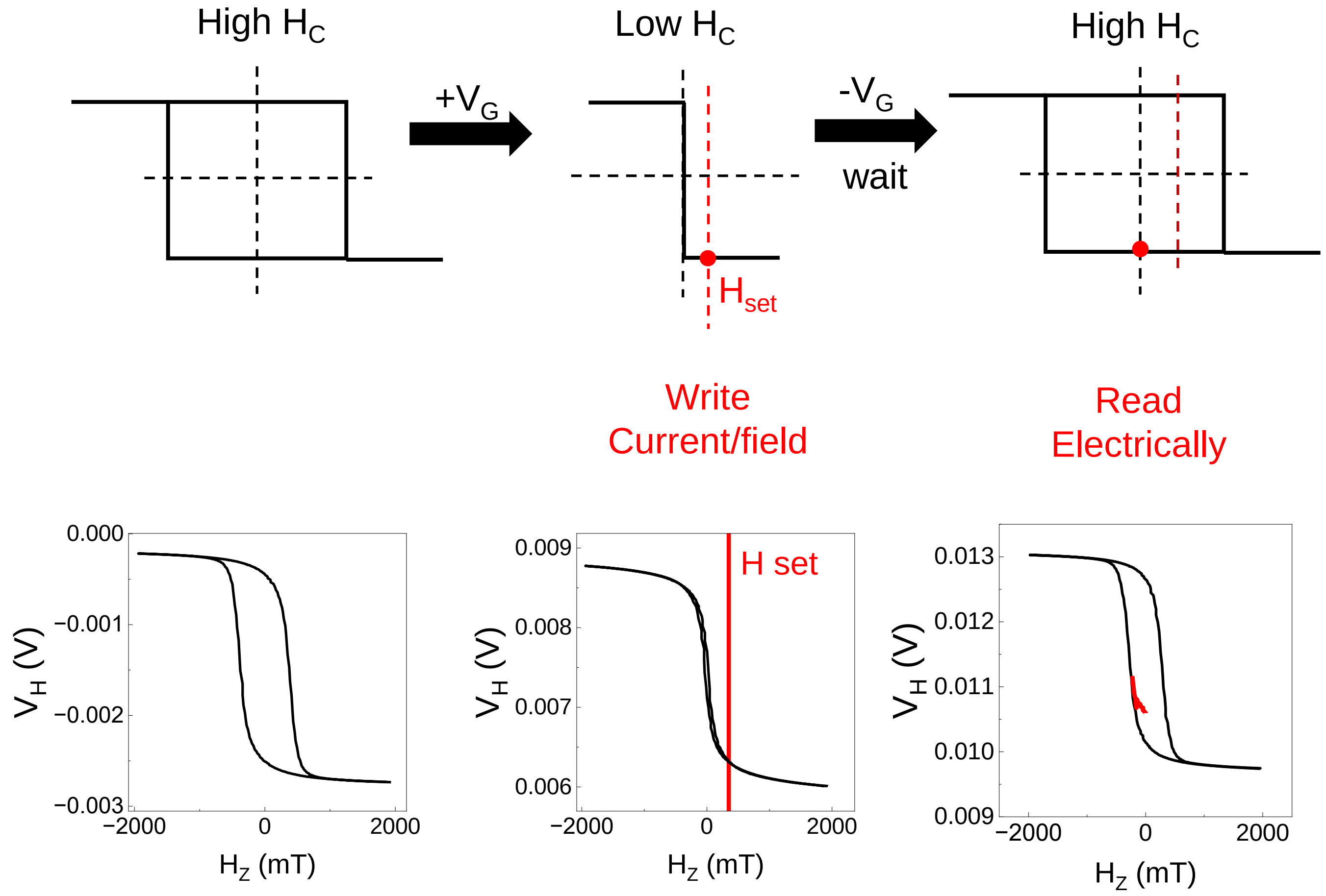


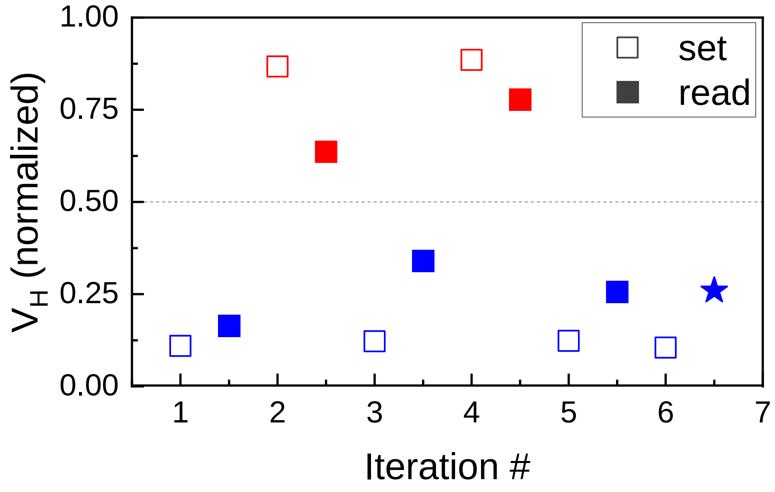


- Opposite switching polarity for Pt and Ta indicates SOT mechanism
- Switching efficiency increases after gating

Tunable spintronic devices

Example of application: magnetoionic memory





The magnetization state set in the low coercivity state is retained after gating

Summary

- High quality epitaxial Mn_4N films can be grown by sputtering in N_2 rich atmosphere on a heated MgO substrate. The films display PMA originating from a combination of strain and capping layer material, large AHE and good magnetotransport properties.

T. Apetrei, E. Demiroglu, C. Deger, C.O. Avci and S. Damerio Phys. Rev. Mater. (submitted)

- Voltage-driven nitrogen ion migration in solid-state devices allows to reversibly tune the magnetic anisotropy of Mn_4N films, increasing the SOT switching efficiency in Mn_4N/Ta bilayers and allowing magnetoionic memory functionality

S. Damerio, T. Apetrei and C. O. Avci Appl. Phys. Lett. **127**, 262405 (2025)

- **Overall voltage driven ionic motion is a powerful tool to tune materials' properties**

Can Onur Avci

Teodor Apetrei

THANK YOU!