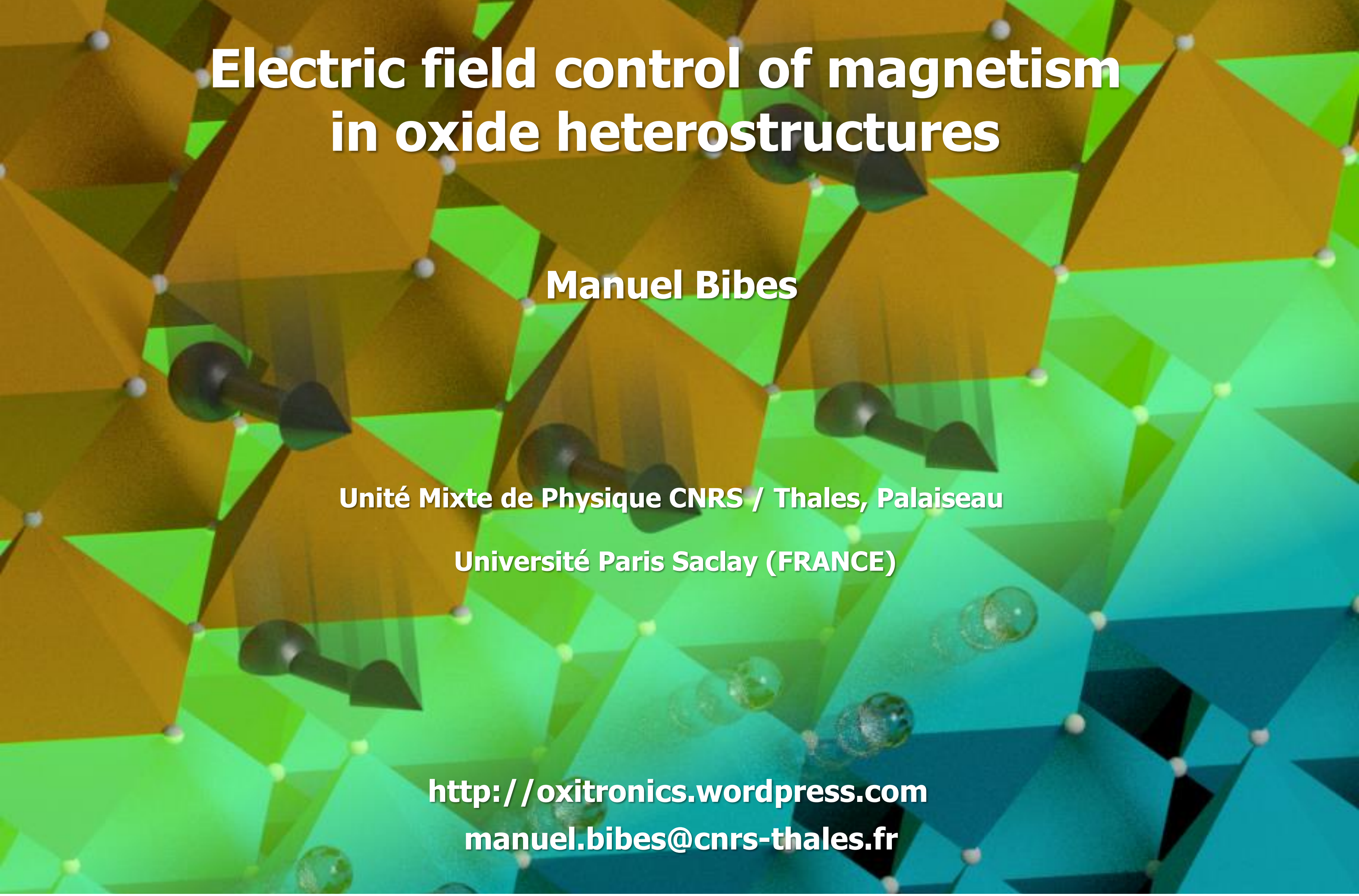


Electric field control of magnetism in oxide heterostructures

Manuel Bibes

Unité Mixte de Physique CNRS / Thales, Palaiseau

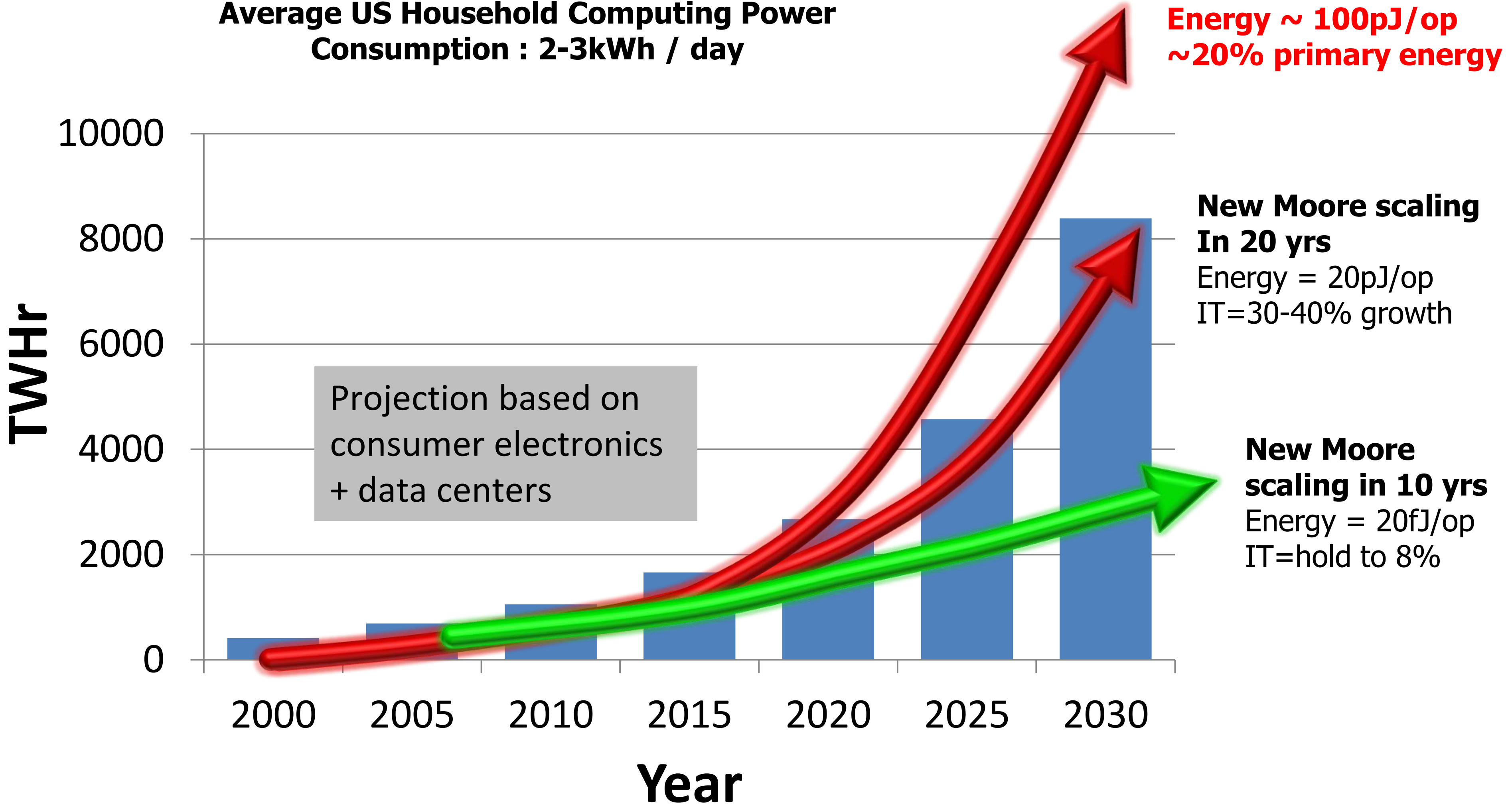
Université Paris Saclay (FRANCE)

http://oxitronics.wordpress.com
manuel.bibes@cnrs-thales.fr

Power consumption of ICT systems

Global Semiconductor market size ~ $5 trillion by 2030

Average US Household Computing Power Consumption : 2-3kWh / day

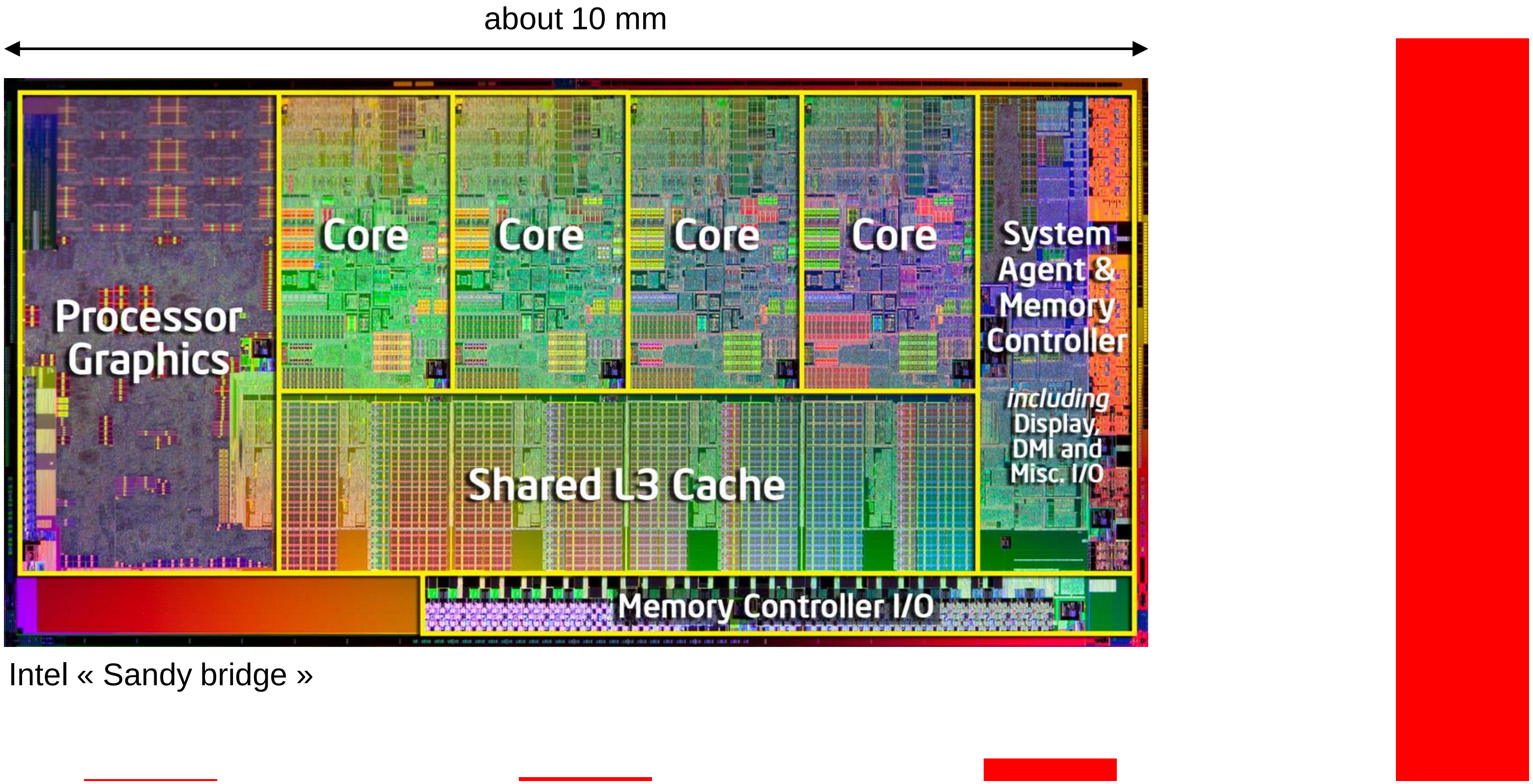


Projection based on consumer electronics + data centers

www.alliancetrustinvestments.com/sri-hub/posts/Energy-efficient-data-centres
www.iea.org/publications/freepublications/publication/gigawatts2009.pdf

Courtesy R. Ramesh

Power needs in microprocessors



Intel « Sandy bridge »

Read one 64-bit number in SRAM

14 pJ

Multiply two 64-bit numbers

50 pJ

Move one 64-bit number 10 mm away

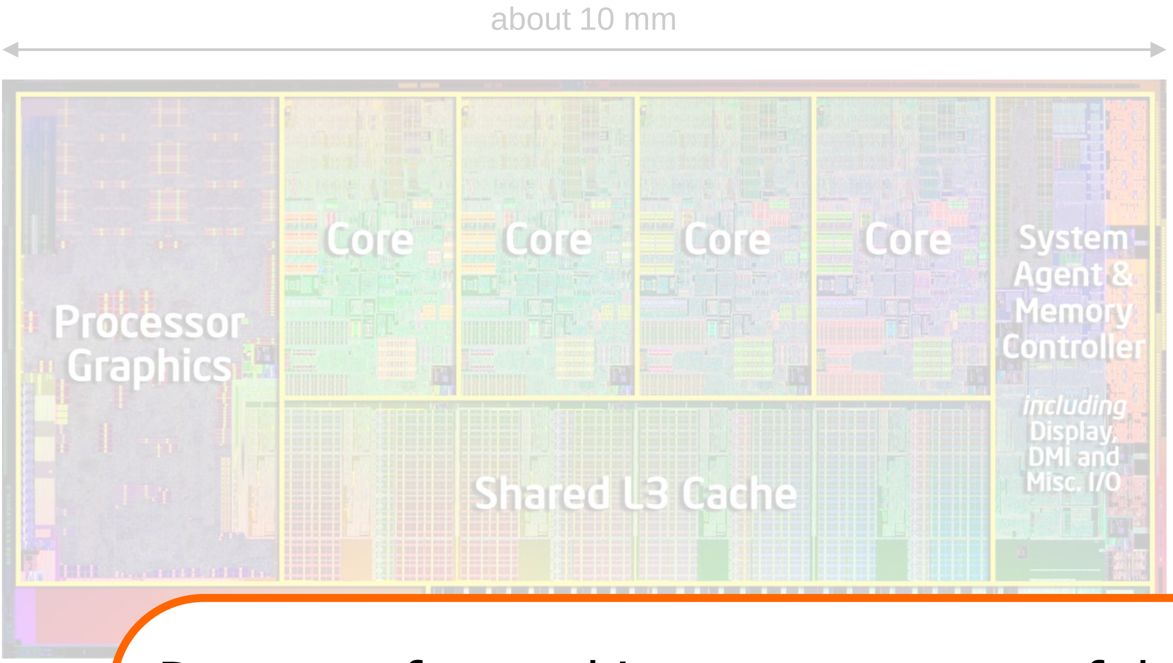
300 pJ

Move one 64-bit number from external RAM

10000 pJ

GPUs and the future of parallel computing, W.J. Dally et al, IEEE Micro (2011)

Power needs in microprocessors



Intel «

Read
numb

1

mber
AM

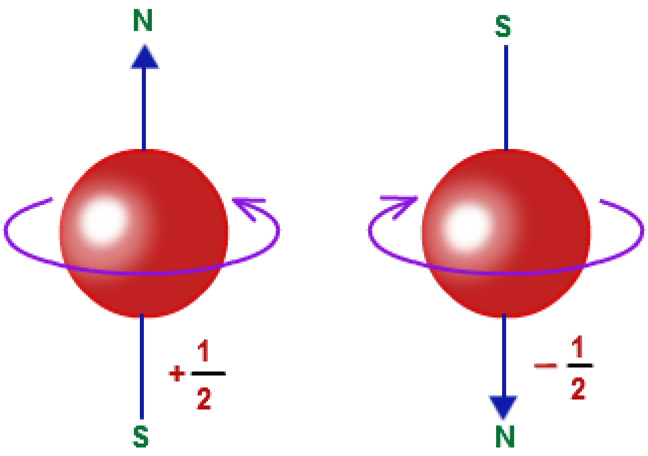
Data transfer on-chip consumes most of the power !

➔ « **Bring memory into logic** »
➔ Embed memory elements in the logic units

➔ **Ferroic materials** can bring solutions for **beyond CMOS electronics**

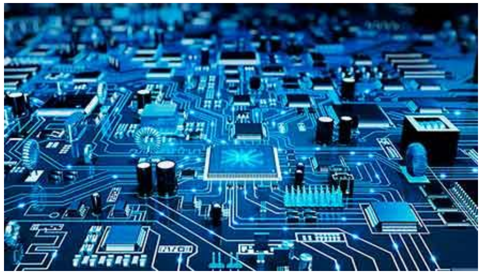
GPUs and the future of parallel computing, W.J. Dally et al, IEEE Micro (2011)

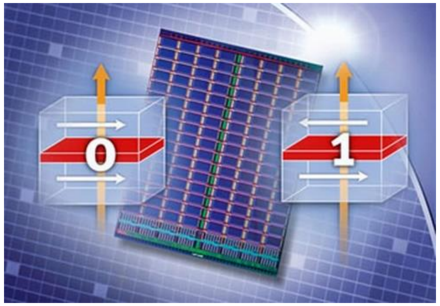
Enter spintronics…



The electron has

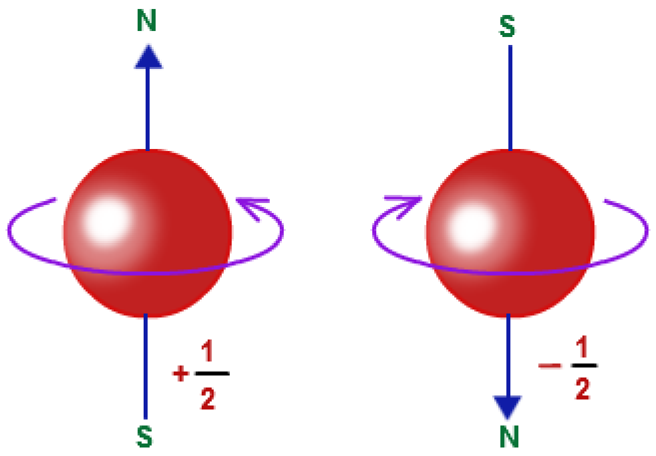
- a charge (-e)
- a spin (↑,↓)





	Electronics	**Magnetism**	**Spintronics**
Information vector	Charge	Magnetization	Electron spin
Control	Electric field	Magnetic field	Magnetic field, spin-polarized current
Detection	Current or voltage measurement	External element (magnetometer)	Current or voltage measurement

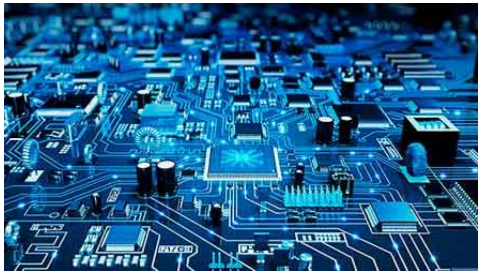
Enter spintronics…

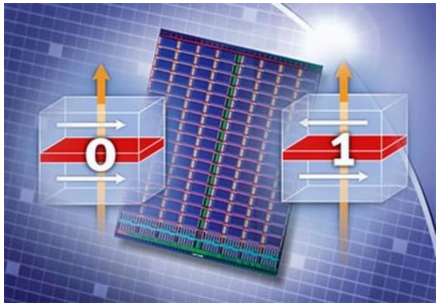


The electron has

a charge (-e)

a spin (↑,↓)

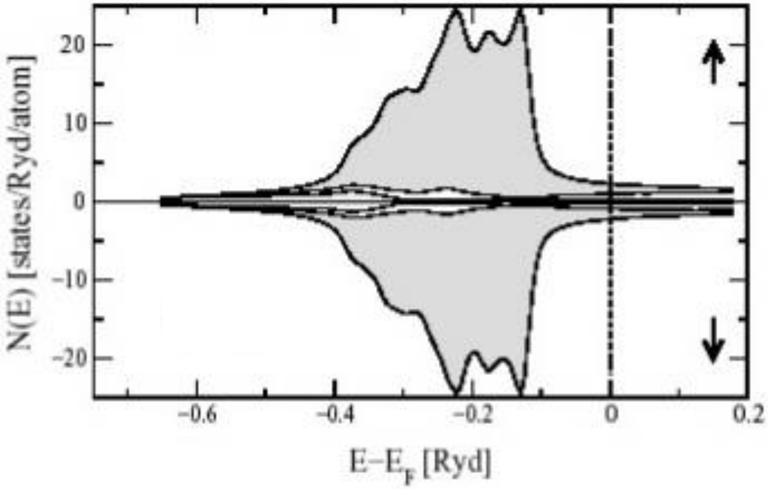




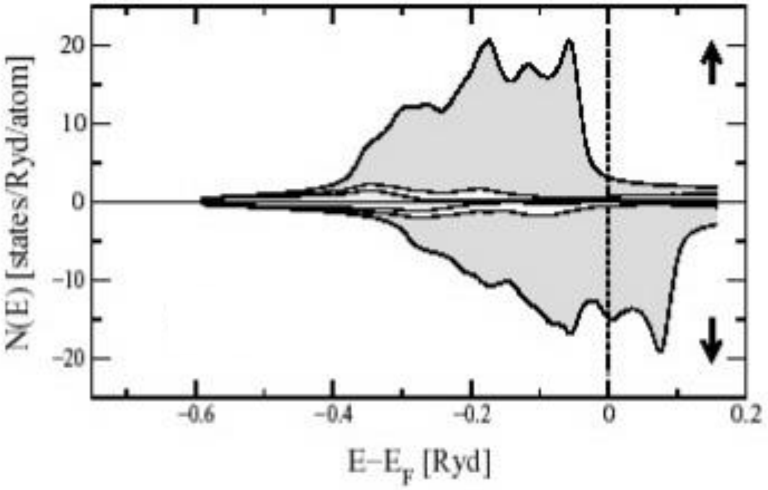
	Electronics	**Magnetism**	**Spintronics**
Information vector	Charge	Magnetization	Electron spin
Control	Electric field	Magnetic field	**Electric field ?**
Detection	Current or voltage measurement	External element (magnetometer)	Current or voltage measurement

Spintronics and ferromagnetic materials

Normal metal (Cu)



Ferromagnetic metal (Co)

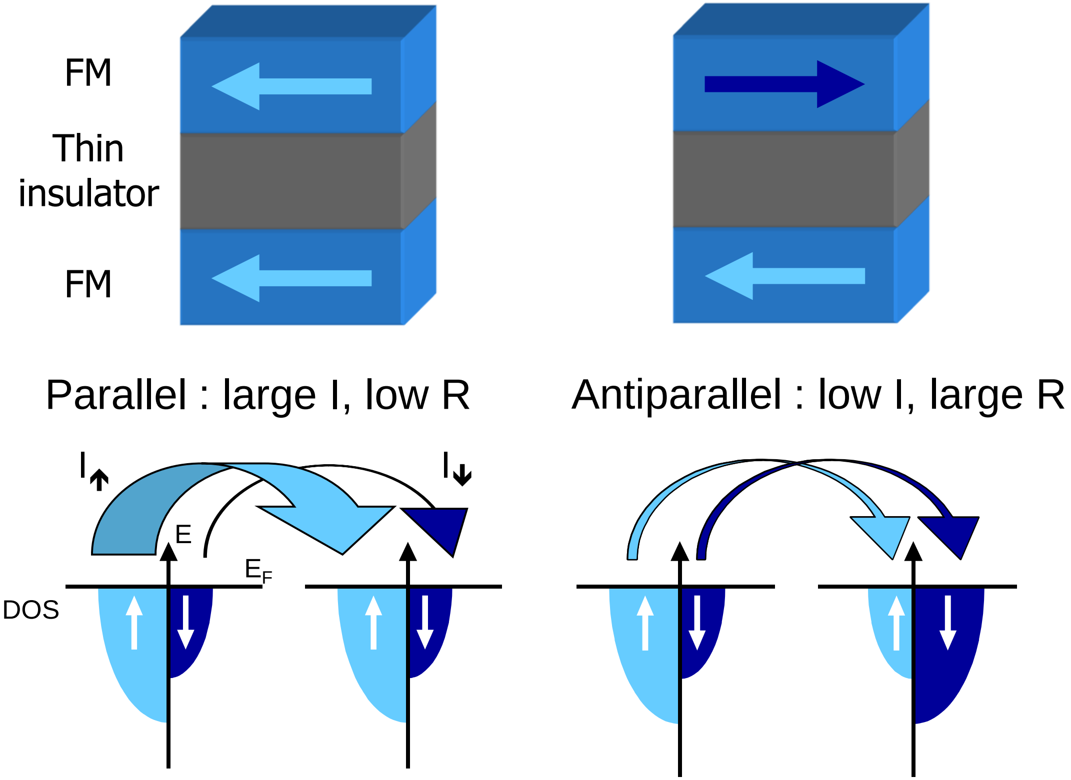


- Non-magnetic metal : DOS is the same for spin up and spin down
- Ferromagnetic metal : different DOS for spin and and spin down
  - spin-polarization of electrons near E_F
  - different transport properties for **spin up and spin down electrons**

Spintronics exploits spin-sensitivity of electron transport

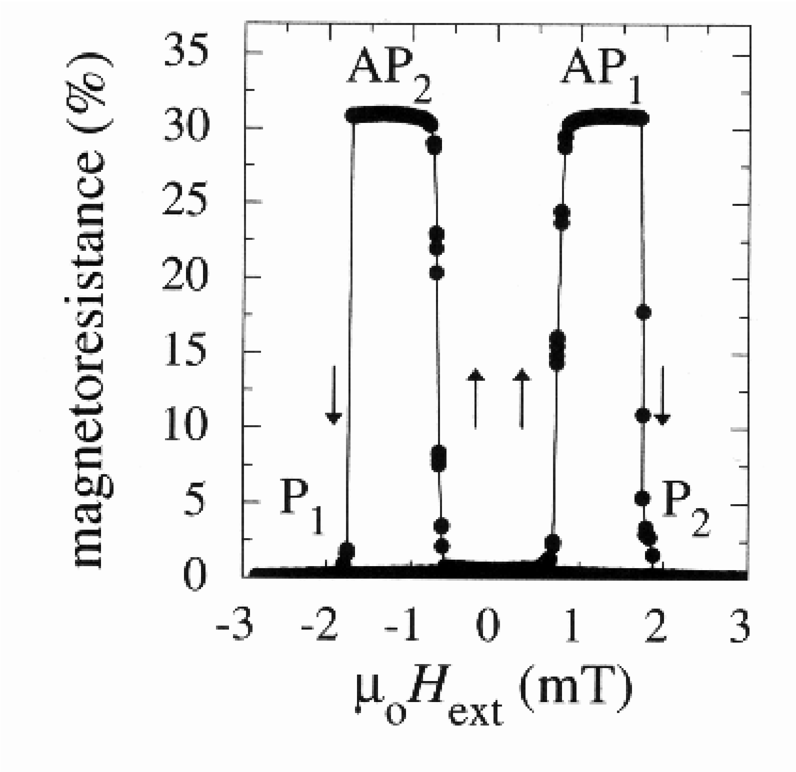
Typical spintronic device : magnetic tunnel junction

MTJ : Trilayer device : two ferromagnetic electrodes separated by thin insulator



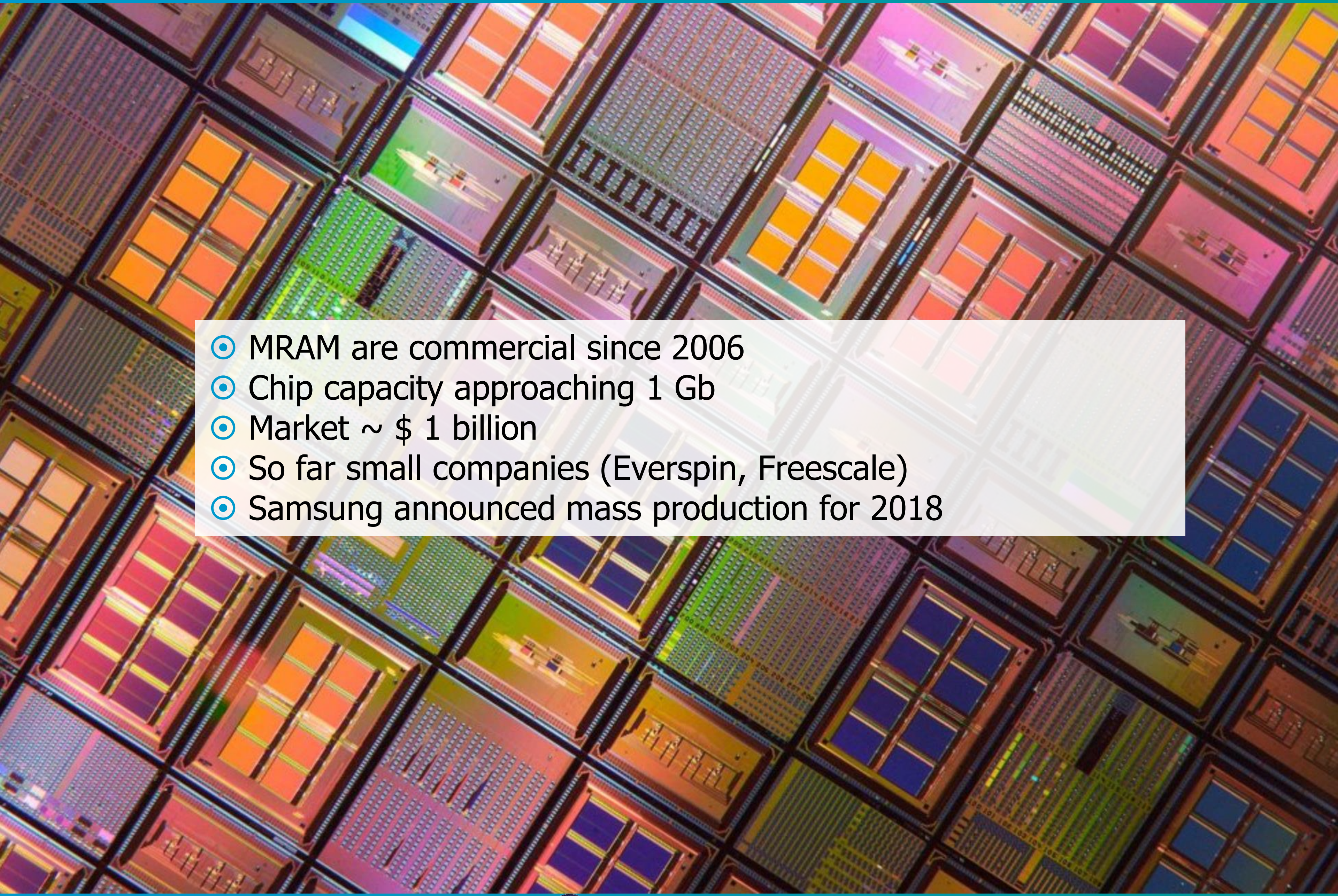
Parallel : large I, low R

Antiparallel : low I, large R



- MTJs show tunnel magnetoresistance (TMR)
- The TMR amplitude increases with the spin-polarization of the electrodes
- Typically switching between P and AP configuration is done by a magnetic field
- Useable for **non-volatile data storage**

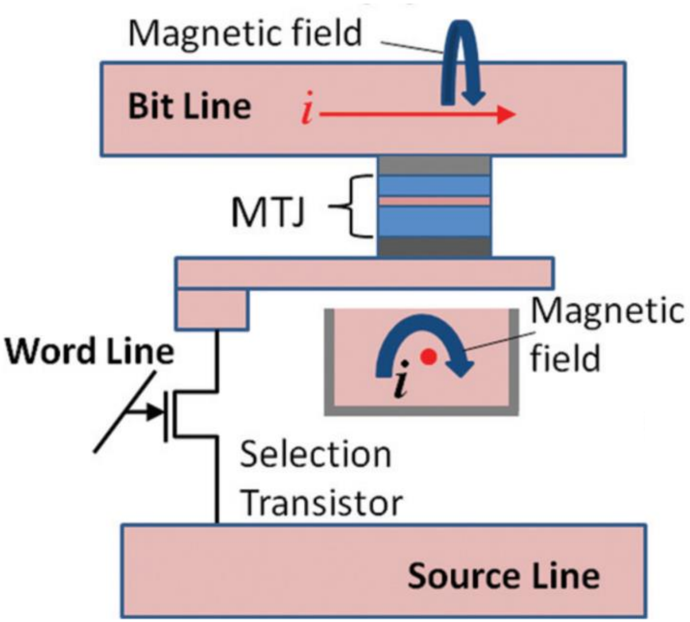
Magnetic random access memory (MRAM)



- MRAM are commercial since 2006
- Chip capacity approaching 1 Gb
- Market ~ $ 1 billion
- So far small companies (Everspin, Freescale)
- Samsung announced mass production for 2018

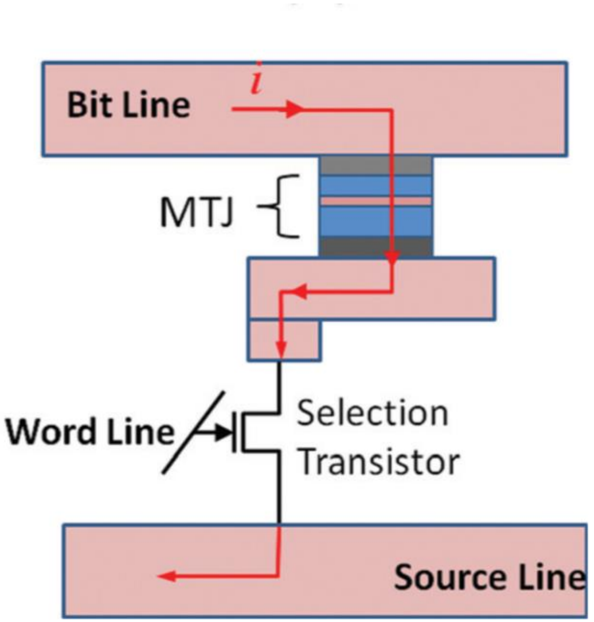
Magnetic random access memory (MRAM)

Magnetization switching with current-induced magnetic field



E > 1000 fJ/bit

Magnetization switching with spin-transfer torque



E ~ 10-100 fJ/bit

GOAL

E < 0.1 fJ/bit

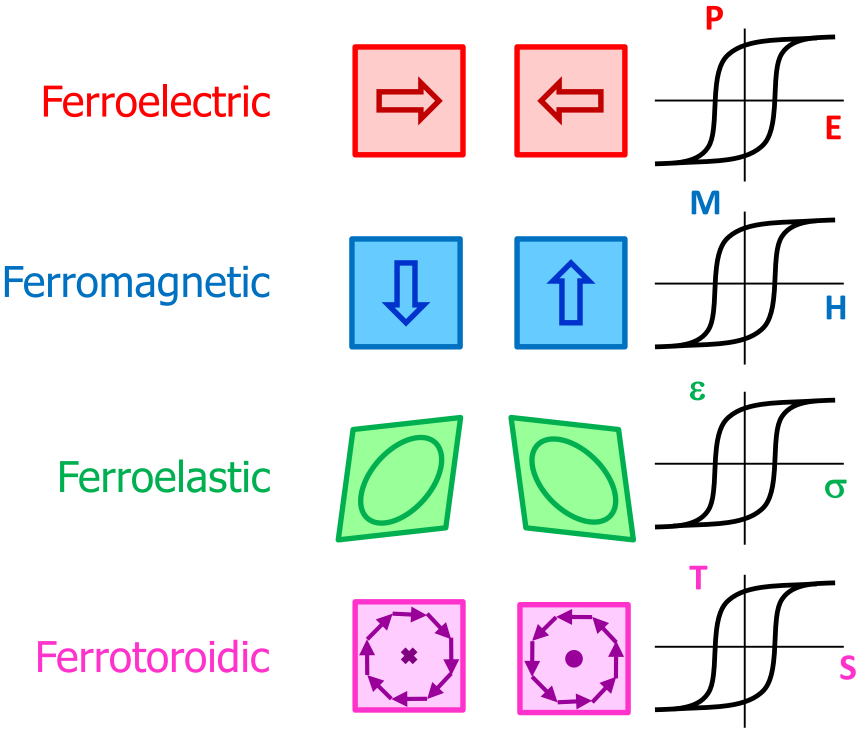
- Move from current-based approaches to electric-field-based approaches
- Electric-field applied across an insulator : power consumption ~0

How to achieve electric-field control of magnetism ?

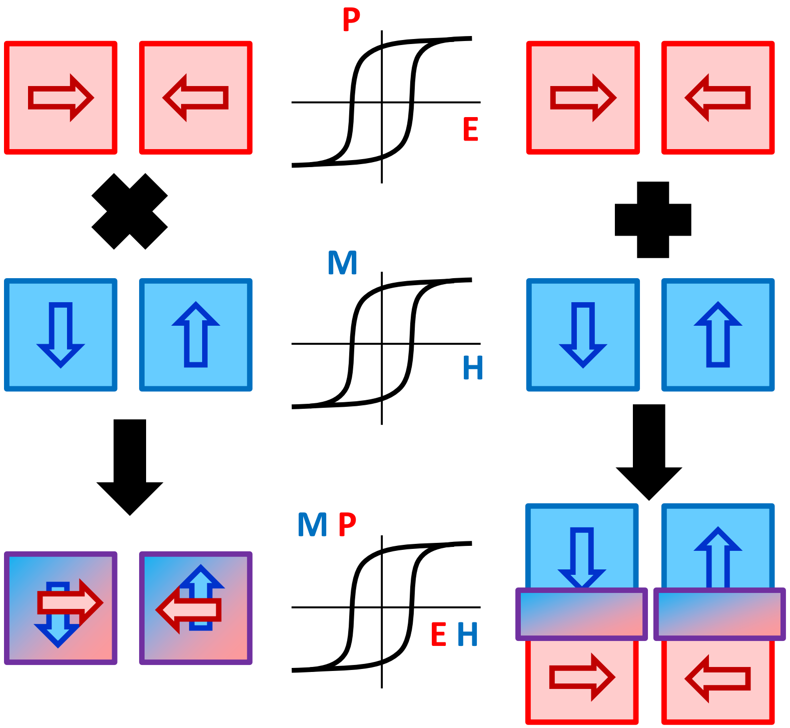
Use multiferroic materials or multiferroic architectures

Introduction to ferroic orders

Ferroic orders



Multiferroic / Magnetoelectric



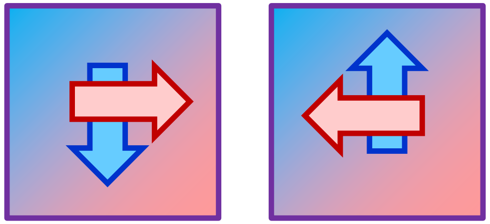
« Intrinsic » « Artificial »

- Hysteretic dependence of order parameter : good for data storage
- Multiple order parameters : increased storage density
- Coupled orders : enhanced flexibility for data writing

MB, Nature Mater. 11, 354 (2012)

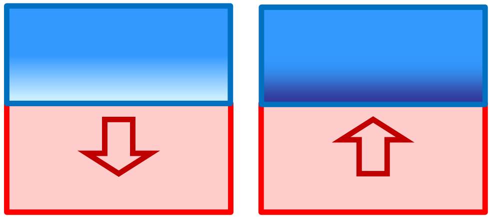
Different approaches for E-field control of magnetism

Intrinsic magnetoelectric



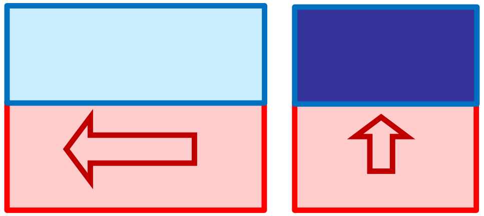
Use single-phase multiferroic material

Field-effect



Combine strong ferroelectric with carrier-mediated ferromagnet

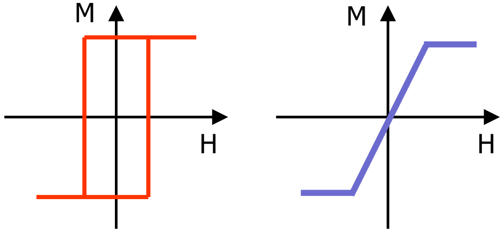
Strain-driven



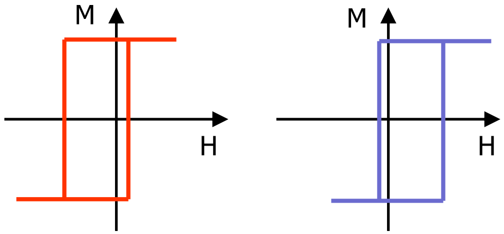
Combine piezoelectric or ferroelectric/ferroelastic with magnetostrictive ferromagnet

Controlling magnetism with electric fields

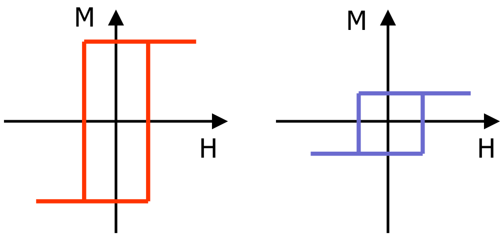
Magnetic anisotropy



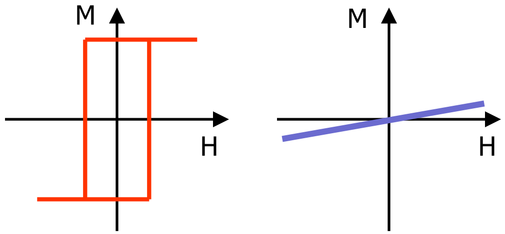
Exchange bias



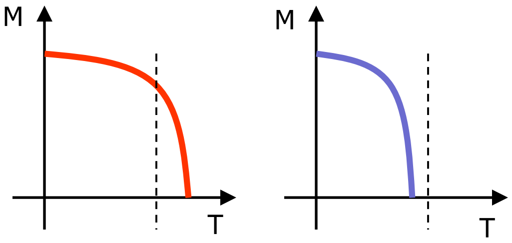
Magnetic moment



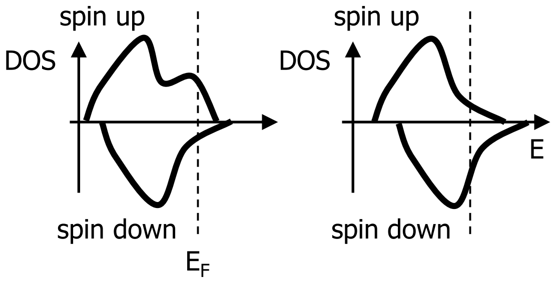
Magnetic order



Curie temperature



Spin polarization

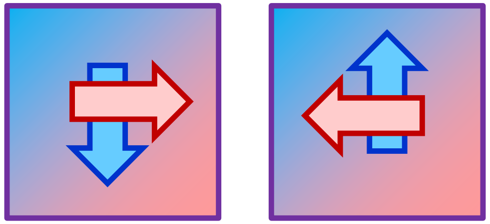


$P_{spin} > 0$ $P_{spin} < 0$

MB, Nature Mater 11, 354 (2012) & MB et al, Annu. Rev. Mater. Res. 44, 91 (2014)

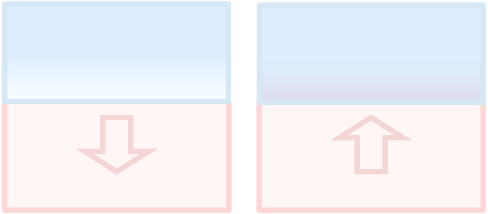
Different approaches for E-field control of magnetism

Intrinsic magnetoelectric



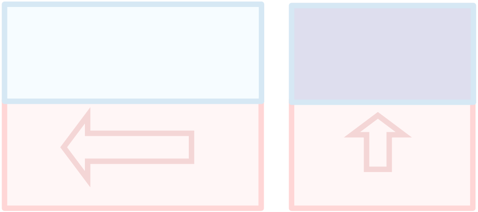
Use single-phase multiferroic material

Field-effect



Combine strong ferroelectric with carrier-mediated ferromagnet

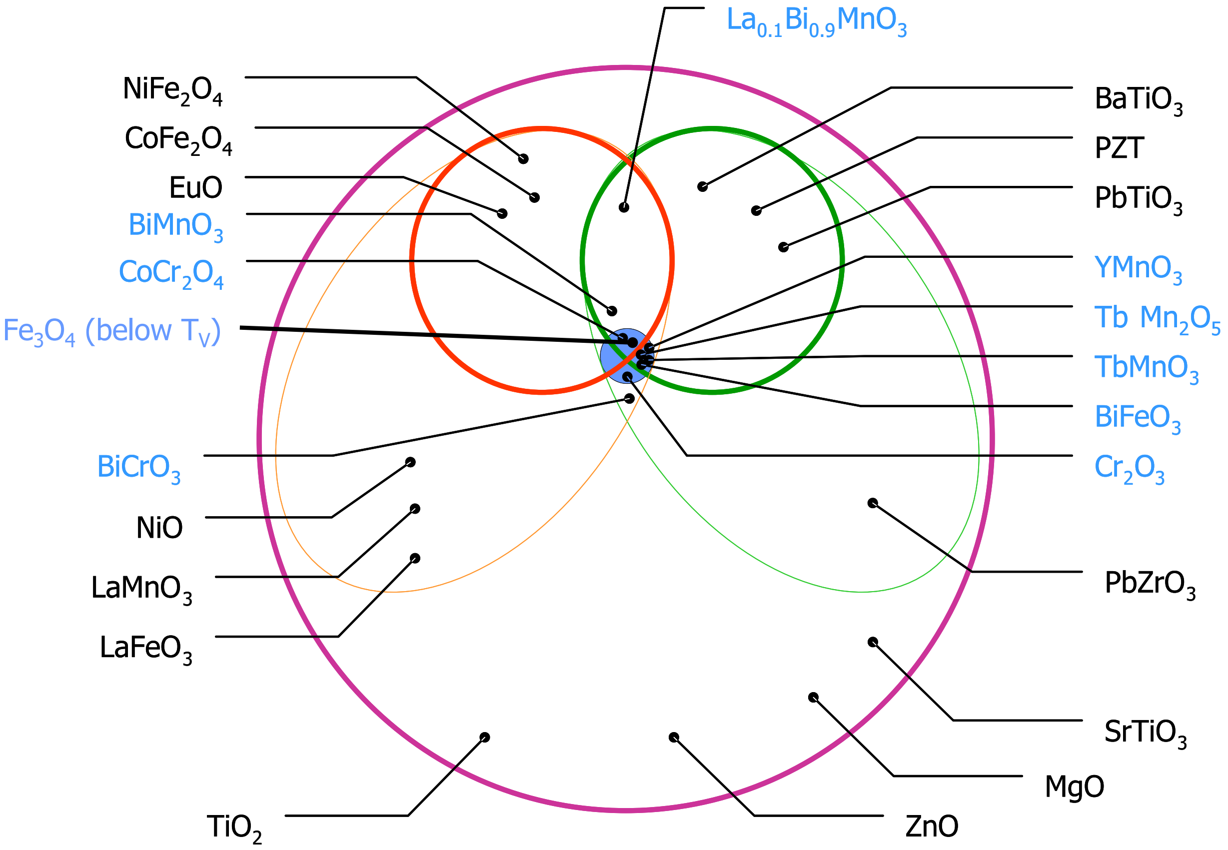
Strain-driven



Combine piezoelectric or ferroelectric/ferroelastic with magnetostrictive ferromagnet

E-field control of magnetism with intrinsic multiferroics

There are very few (room-temperature) multiferroics



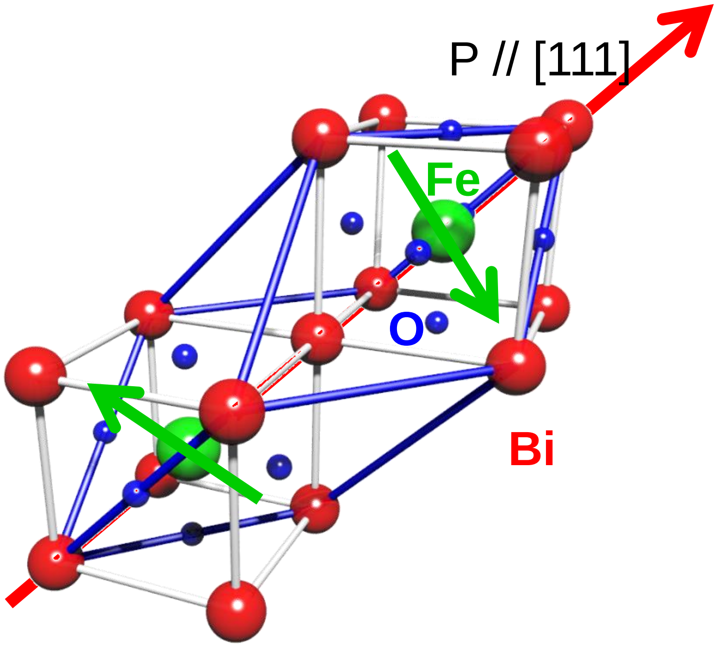
Magnetically polarizable
Ferro(i)magnetic

Electrically polarizable
Ferro(i)electric

Magnetoelectric

H. Béa, MB et al, J. Phys.: Condens. Matter 20, 434221 (2008)
Derived from Eerenstein, Mathur and Scott, Nature 442, 759 (2006)

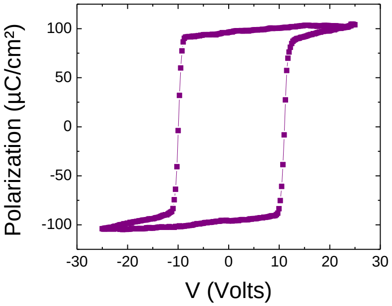
$BiFeO_3$: a room-temperature multiferroic



Ferroelectric properties

- Very high $T_C \approx 1100$ K
- Very large P=100 μC/cm²

Fisher et al., J. Phys. C, 13, 1931 (1980)
Béa, MB et al, APL 93, 072091 (2008)



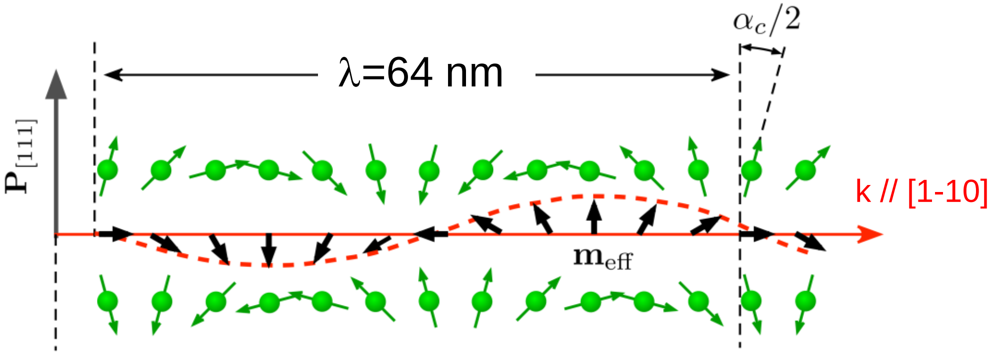
Magnetic properties

- G-type antiferromagnetic

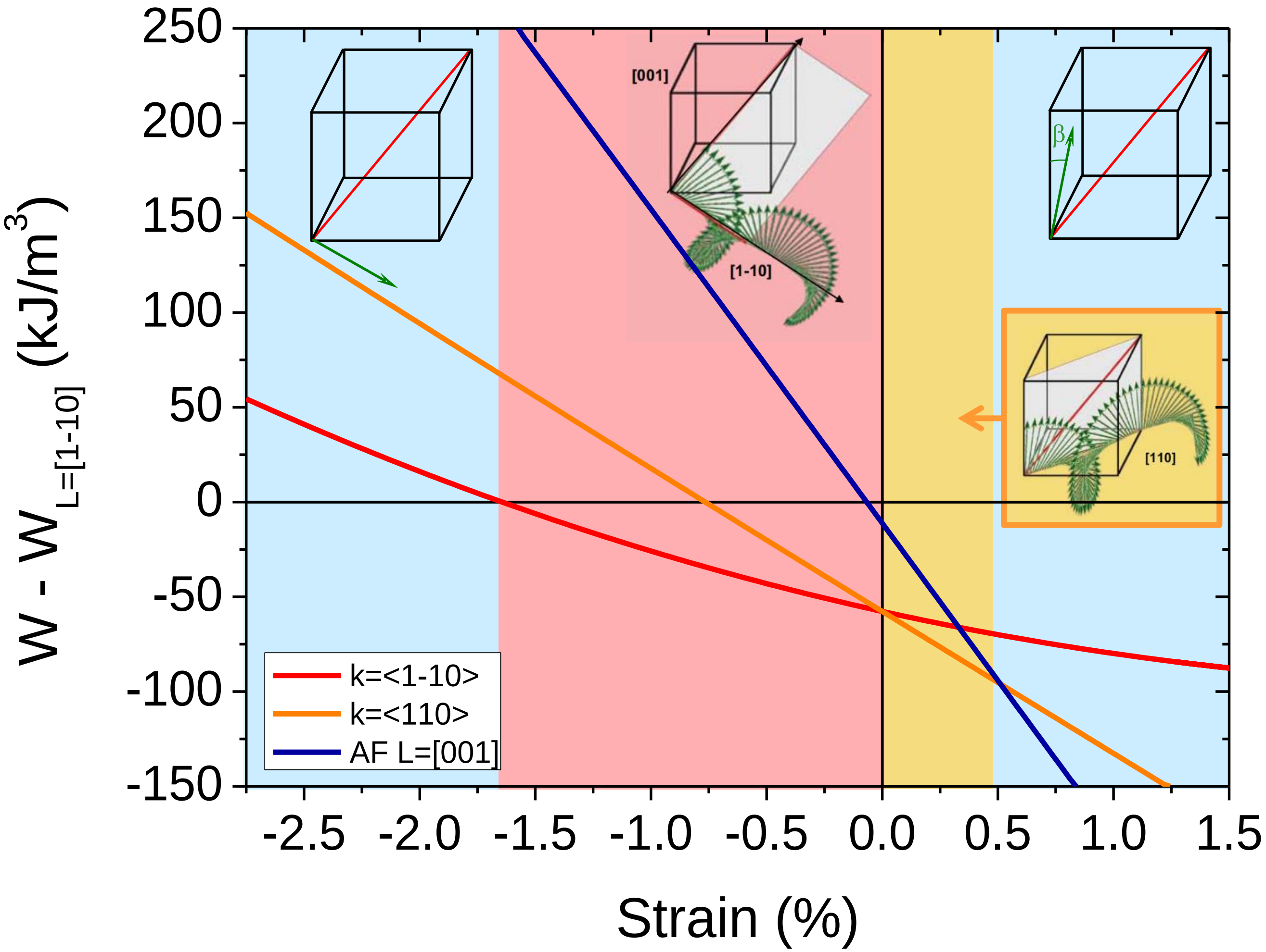
+ cycloidal modulation (λ=62 nm)

- Weak moment with periodic modulation
- $T_N \approx 640$ K

Sosnowska et al., J. Phys. C, 15, 4835 (1982)



Influence of epitaxial strain on the magnetic properties of $BiFeO_3$



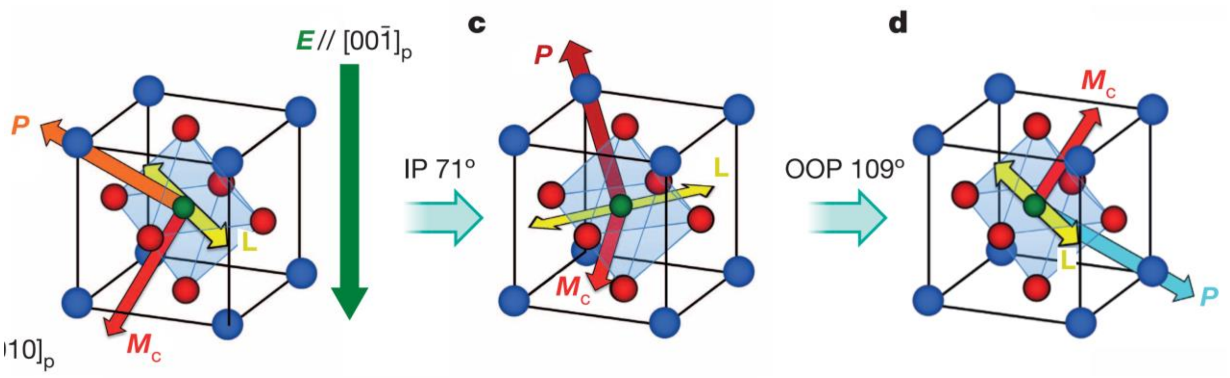
- Cycloidal state is destabilized by strain-induced (magnetoelastic) anisotropy
- Weak-FM state at high tensile or compressive state
- New cycloid stabilized at low tensile strain

} Mössbauer spectroscopy + theory

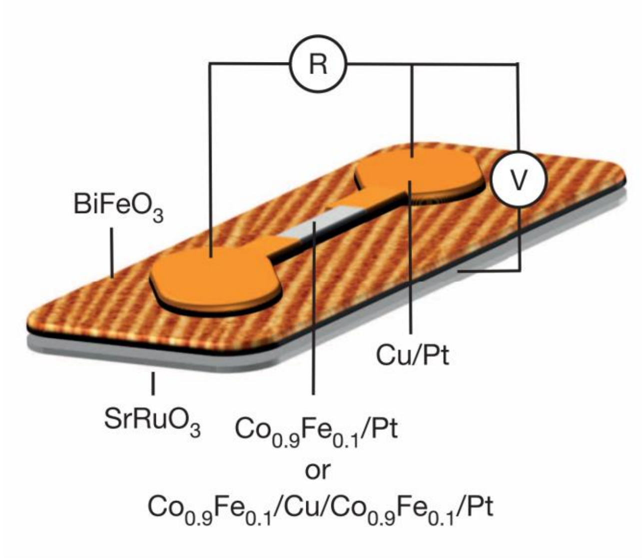
D. Sando, MB et al, Nature Mater. 12, 641 (2013)

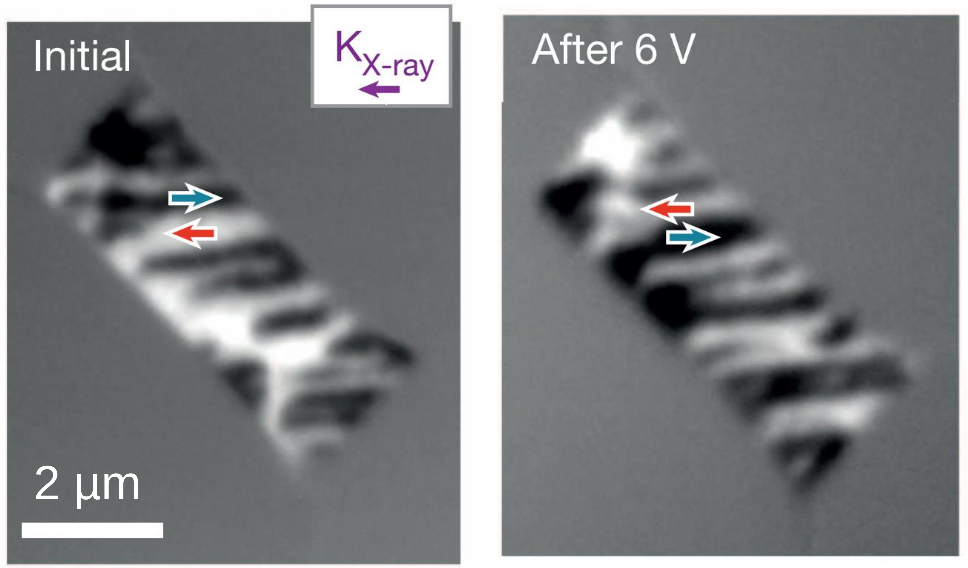
E-field induced magnetization switching with $BiFeO_3$ thin films

Sequential switching of P promotes switching of weak M



Heron et al, Nature 370, 516 (2014)

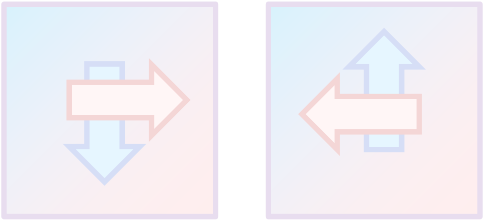




- Application of out-of-plane voltage to BFO film promotes local switching of magnetization in Co film grown on top of BFO

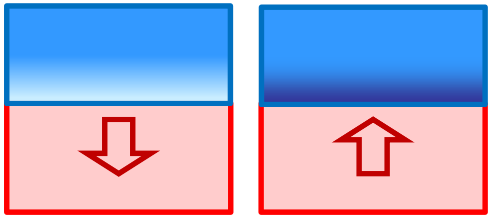
Different approaches for E-field control of magnetism

Intrinsic magnetoelectric



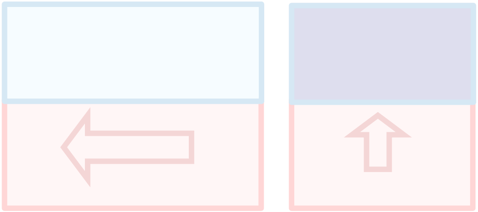
Use single-phase multiferroic material

Field-effect



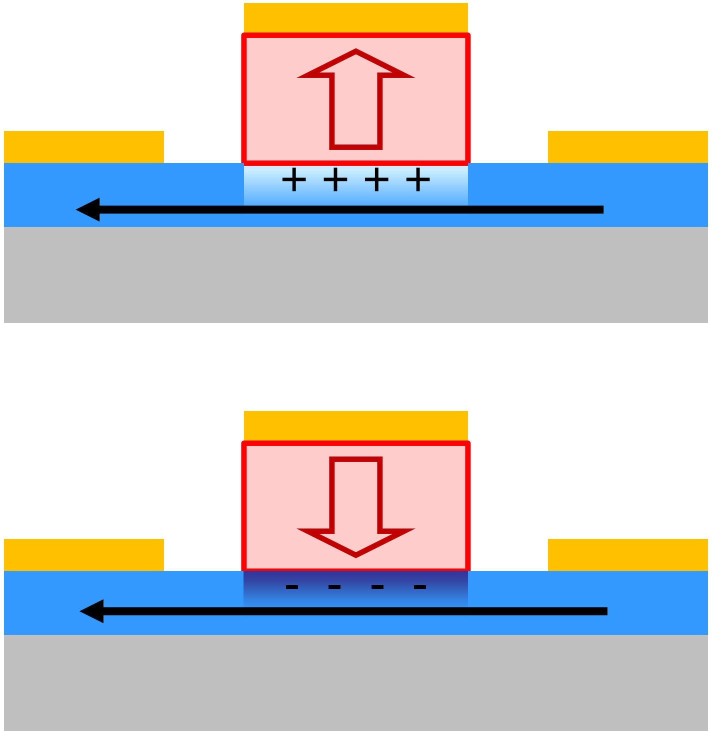
Combine strong ferroelectric with carrier-mediated ferromagnet

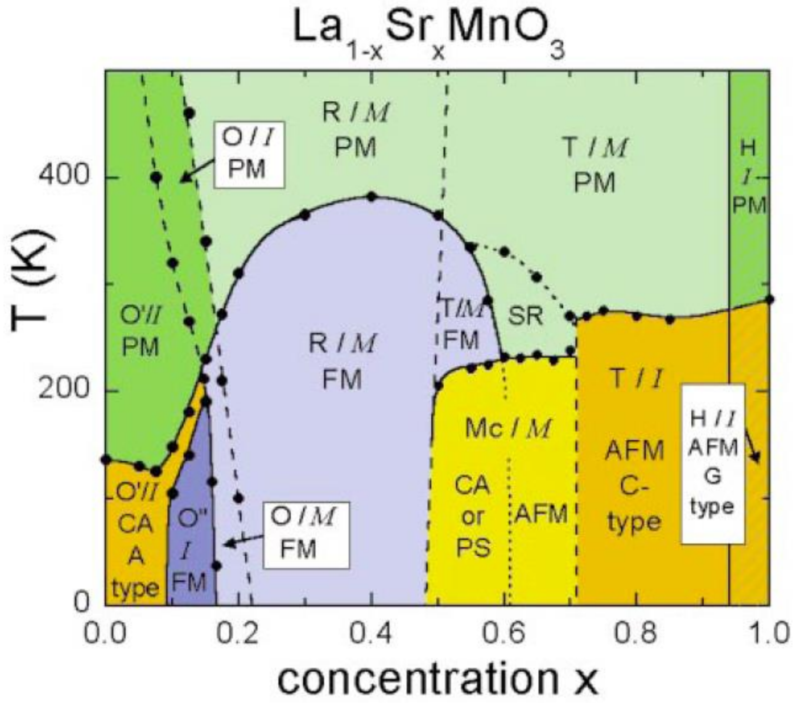
Strain-driven



Combine piezoelectric or ferroelectric/ferroelastic with magnetostrictive ferromagnet

Field-effect control of magnetism





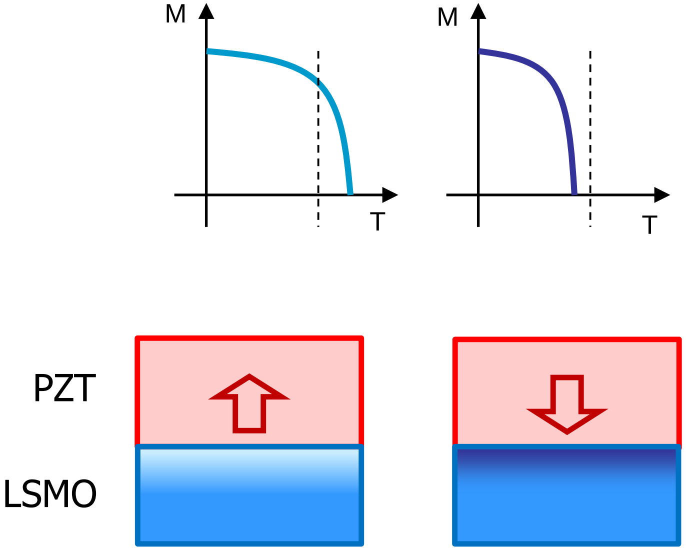
Hemberger et al., PRB, 66, 094410 (2002)

- ⦿ Charge accumulation / depletion thanks to a dielectric or ferroelectric (non-volatile)
- ⦿ If magnetism in channel material is (highly) sensitive to carrier density

➔ **Change of magnetic properties by electric field**

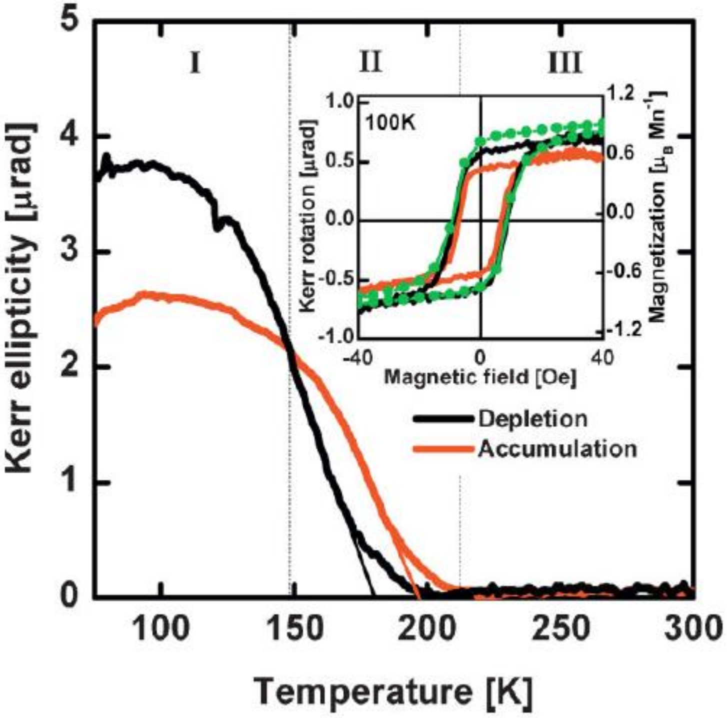
- ⦿ Effect occurs over small distance, typically **Thomas Fermi screening length** (Å for metals, nm for oxides)

Field-effect control of Curie temperature



deplete electrons
accumulate holes
T_C increases

accumulate electrons
deplete holes
T_C decreases

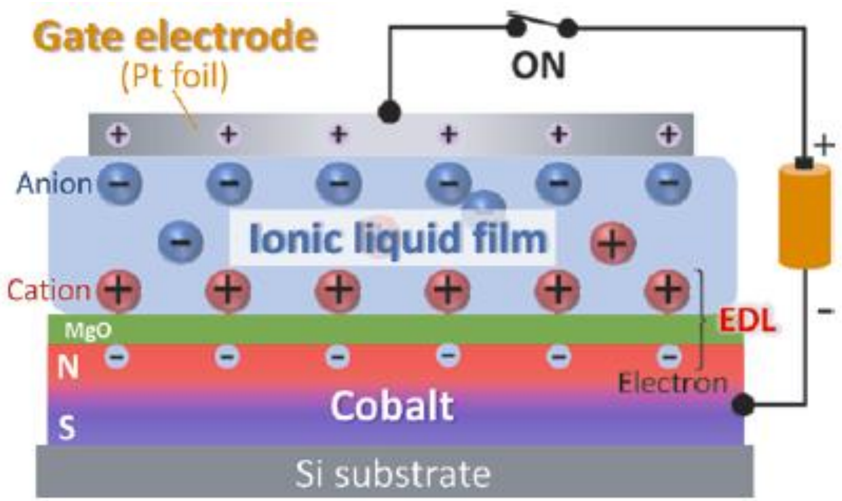


Vaz et al, PRL 104, 127202 (2010) & Molegraaf et al, Adv. Mater. 21, 3470 (2009)

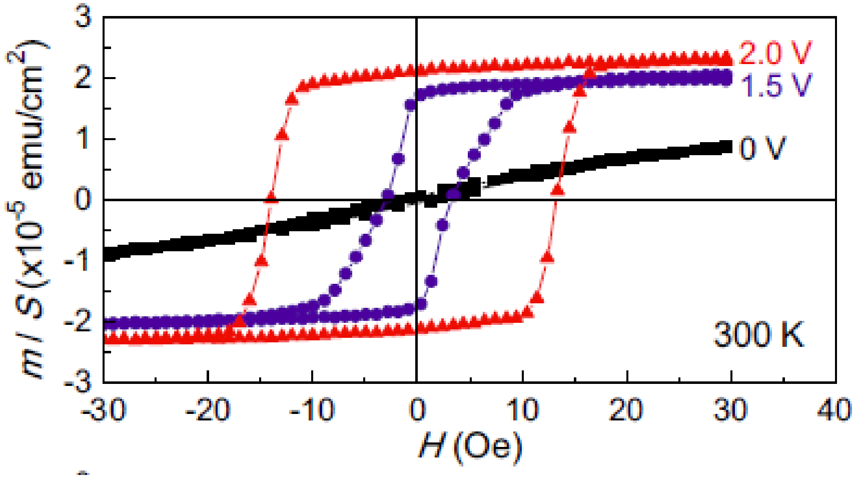
- Combination of a **ferroelectric** and a **carrier-mediated ferromagnet**
- Switching P in ferroelectric PZT produces charge accumulation/depletion in manganite
- ➔ Change T_C of manganite
- Limited to low-temperature (also with GaMnAs)

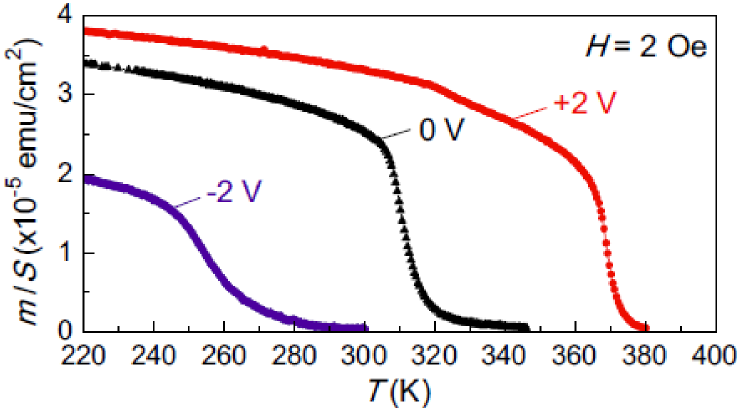
Field-effect control of Curie temperature

Increase accumulated charge density : ionic liquids



Chiba and Ono, J. Phys. D 46, 213001 (2013)



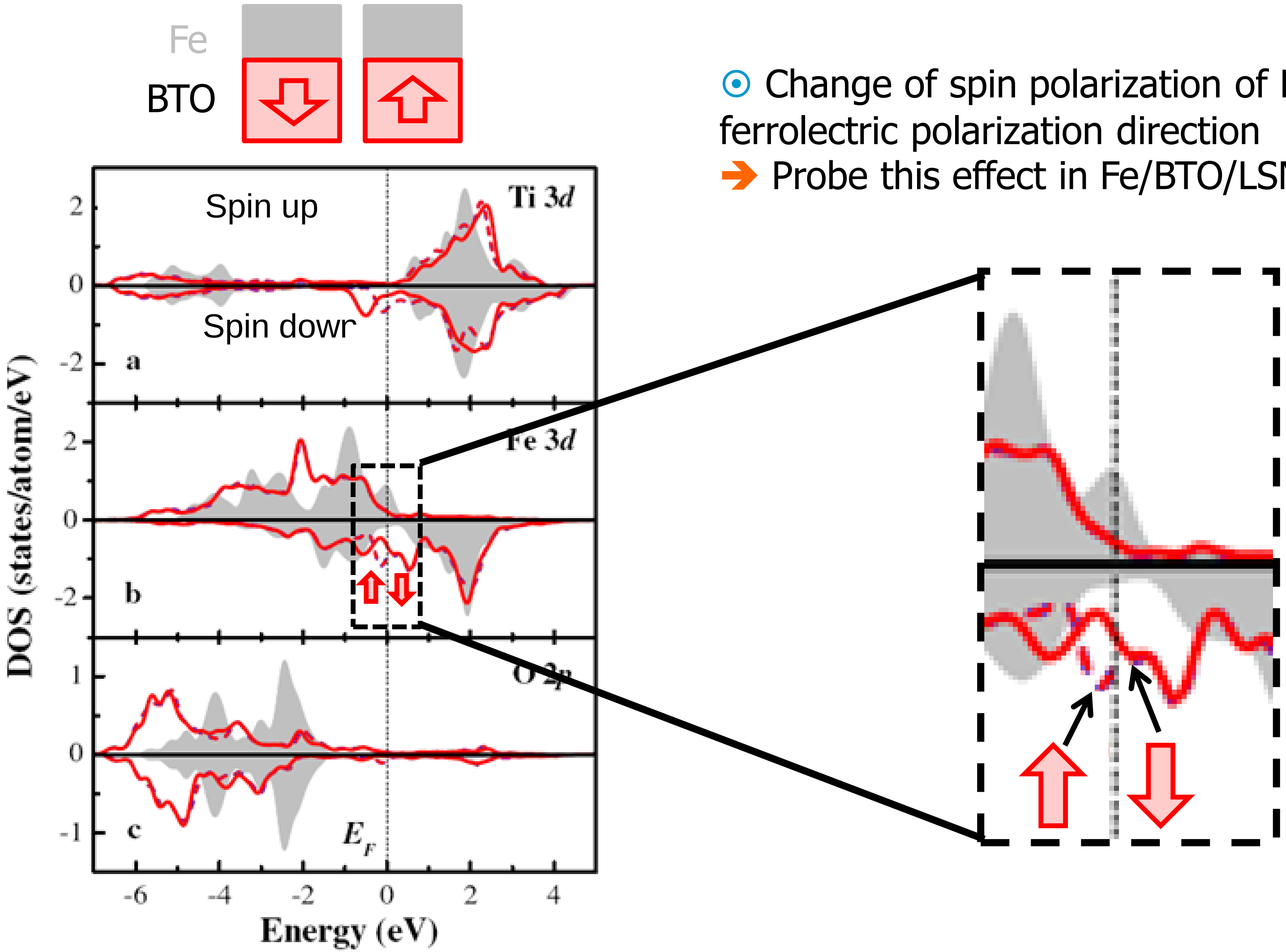


- Large field effect in 0.6 nm Co film using ionic liquid gating
- Possible with ferroelectrics (i.e. PZT/ultrathin Co) ?

Electric-field control of spin polarization

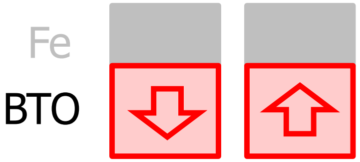
- Change of spin polarization of Fe depending on ferrolectric polarization direction

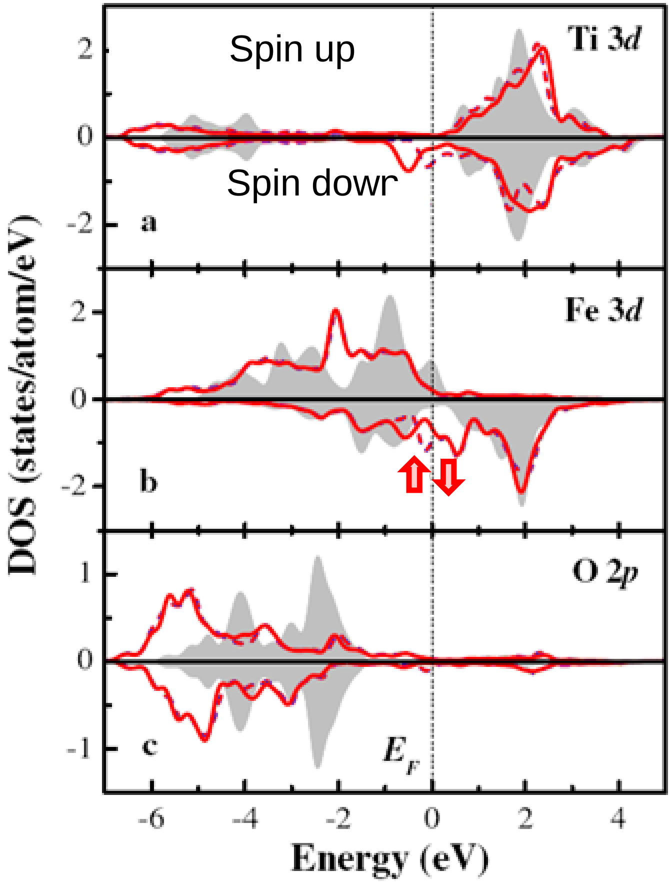
➔ Probe this effect in Fe/BTO/LSMO tunnel junctions



Duan et al., PRL 97, 047201 (2006)
Fechner et al, PRB 78, 212406 (2008)

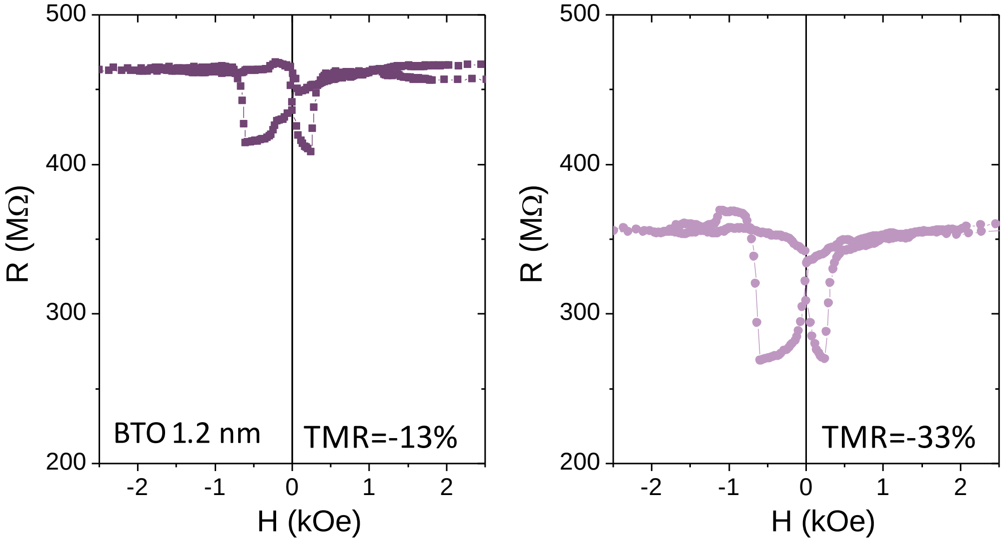
Electric-field control of spin polarization





Duan et al., PRL 97, 047201 (2006)
Fechner et al, PRB 78, 212406 (2008)

- ⊙ Change of spin polarization of Fe depending on ferrolectric polarization direction
- ➔ Probe this effect in Fe/BTO/LSMO tunnel junctions

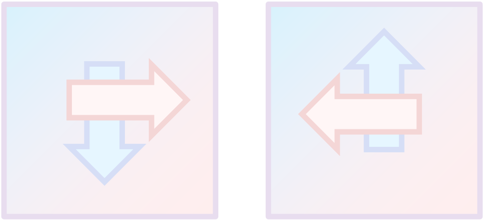


- ⊙ TMR amplitude depends on direction of P
- ➔ **Ferroelectric control of spin polarization**
- ⊙ Combination of field-effect and hybridization changes

V. Garcia, MB et al, Science 327, 1106 (2010)
S. Valencia, MB et al, Nature Mater. 10, 753 (2011)

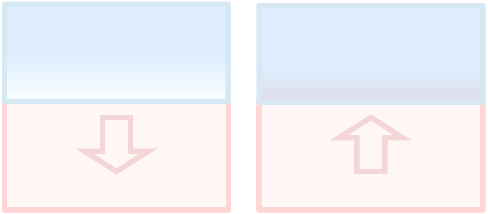
Different approaches for E-field control of magnetism

Intrinsic magnetoelectric



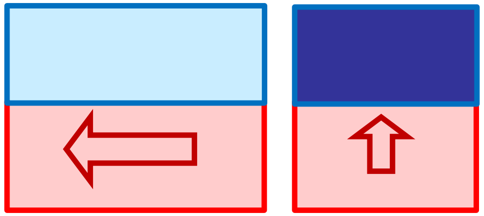
Use single-phase multiferroic material

Field-effect



Combine strong ferroelectric with carrier-mediated ferromagnet

Strain-driven



Combine piezoelectric or ferroelectric/ferroelastic with magnetostrictive ferromagnet

Strain-induced control of magnetic anisotropy

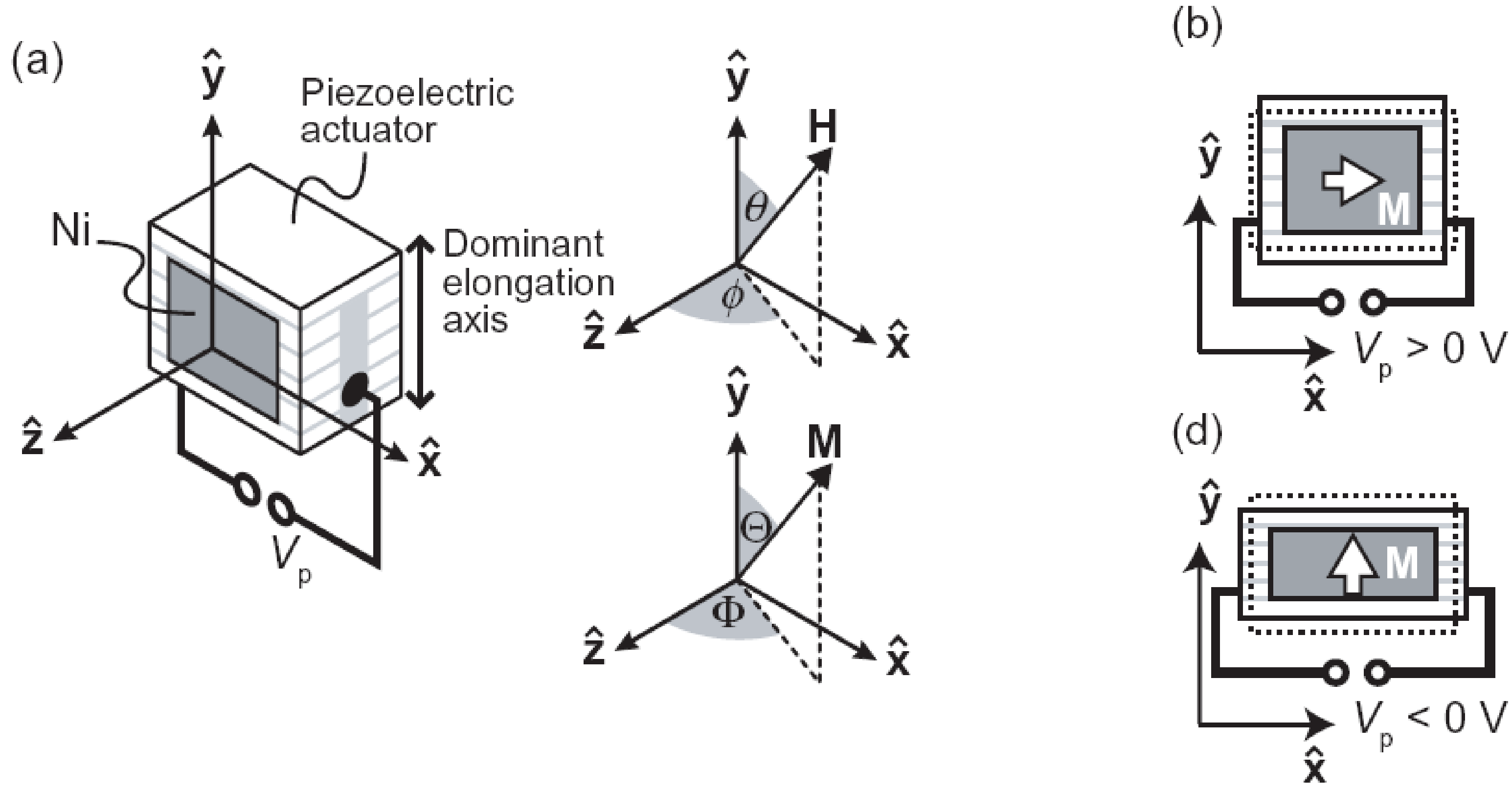
Example : experiments on PZT/Ni Weiler et al, New J. Phys 2009

Principle :

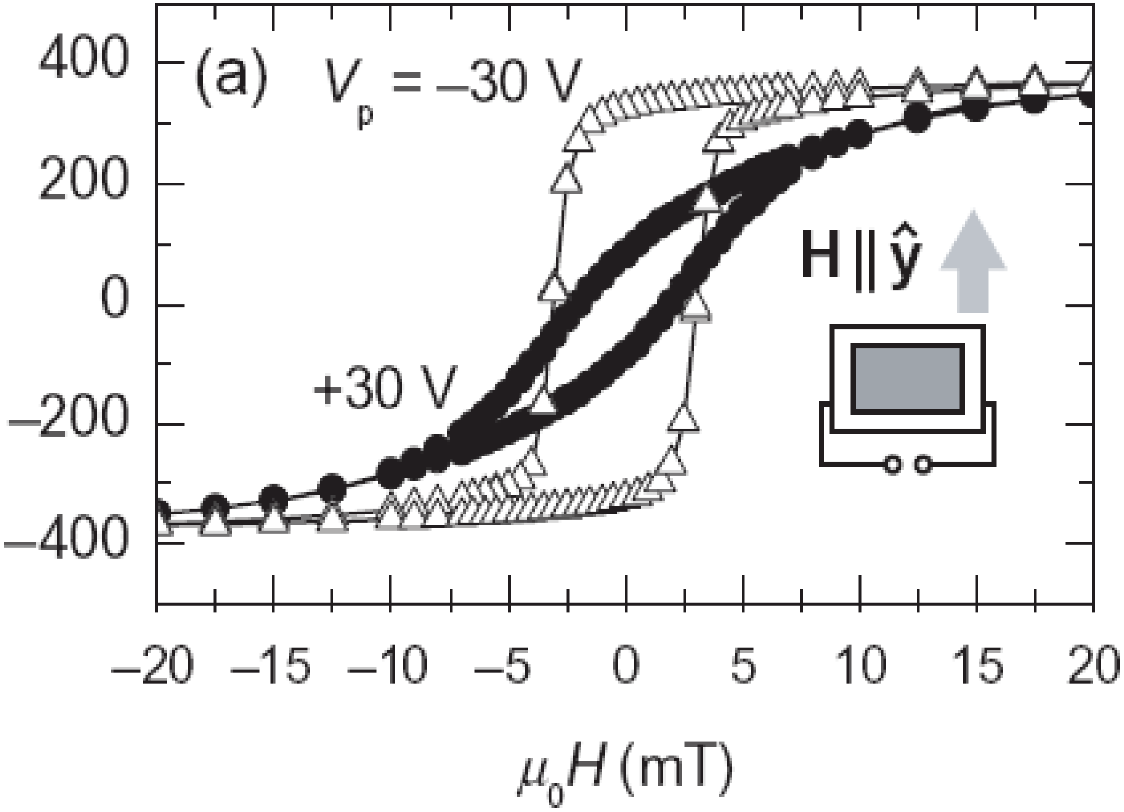
E-field applied to PZT : change in PZT dimensions due to **converse piezoelectric effect**

➔ Change in dimensions induced in Ni : strain effect

➔ Due to **magnetostriction** in Ni, strain modifies the magnetic properties



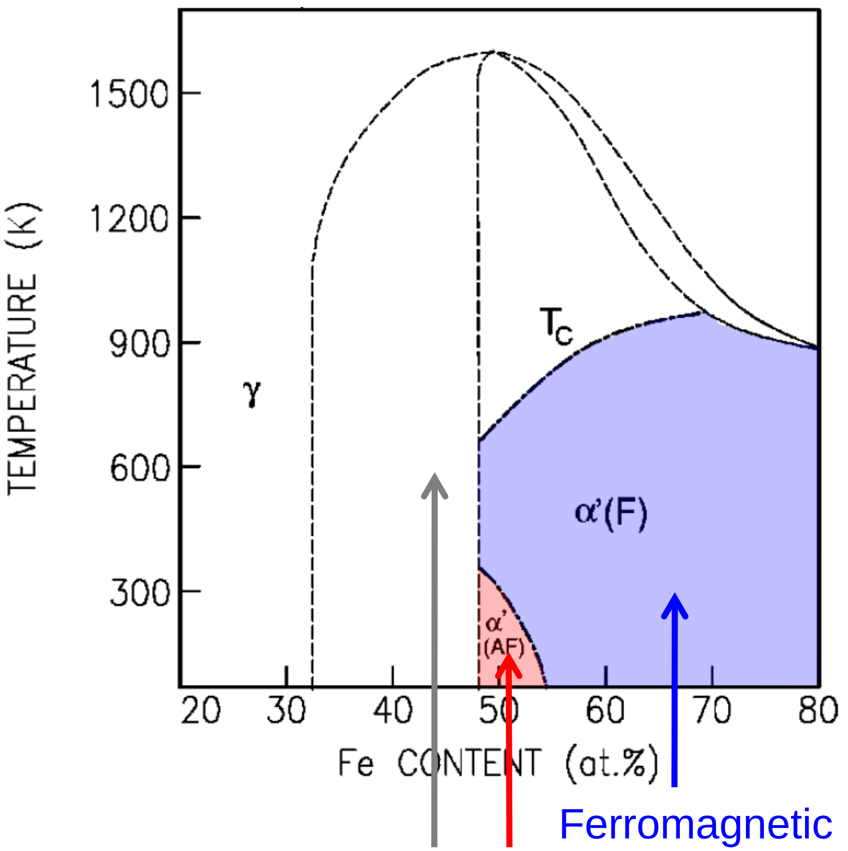
Strain-induced control of magnetic anisotropy



Weiler et al, New J. Phys 2009

Electric-field induced control of magnetization easy axis

Strain-induced control of magnetic order



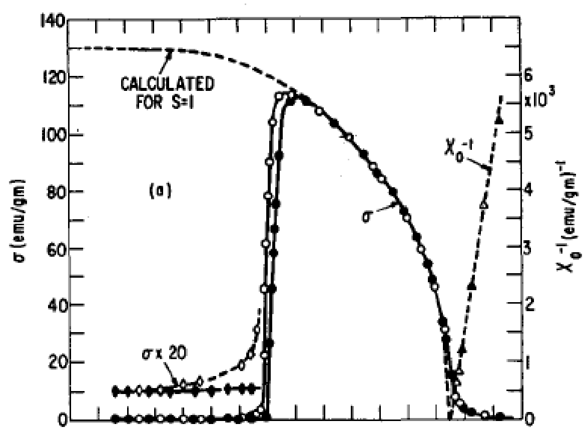
Ferromagnetic phase

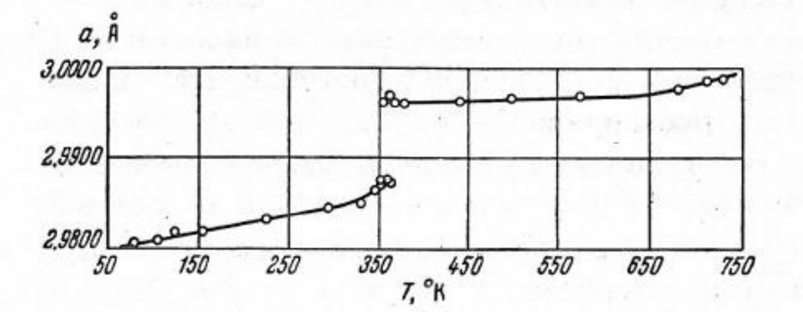
Paramagnetic phase Antiferromagnetic phase

γ phase : fcc ; α phase : disordered bcc

α' phase : Fe/rh ordered bcc

van Driel et al, JAP 85, 1026 (1999)





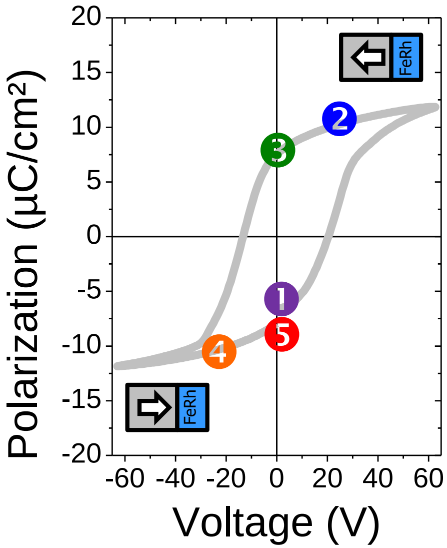
- Near $Fe_{50}Rh_{50}$, transition from AFM to FM at about 370K
- Transition is first order
- Associated large resistivity drop
- Jump of cell volume by ~1% at T*

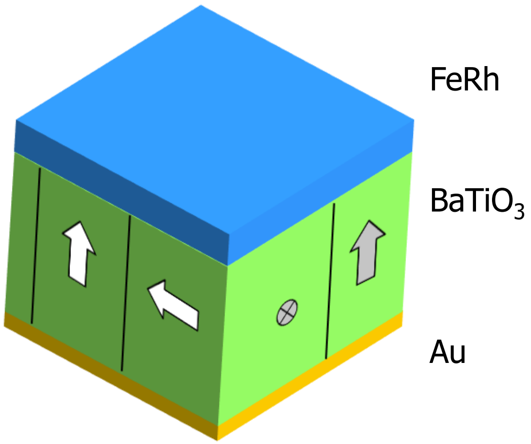
Zakharov et al, Sov. Phys. JETP 19, 1348 (1964)

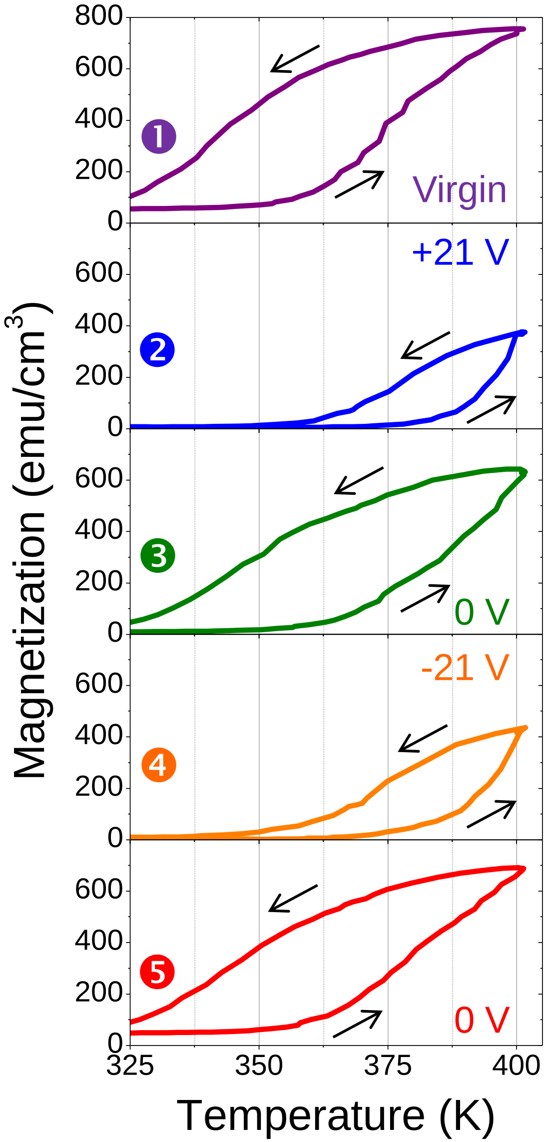
- Magnetic state of FeRh is sensitive to pressure

➔ Grow on **ferroelectric/ferroelastic $BaTiO_3$** substrate to achieve E-field control

Strain-induced control of magnetic order







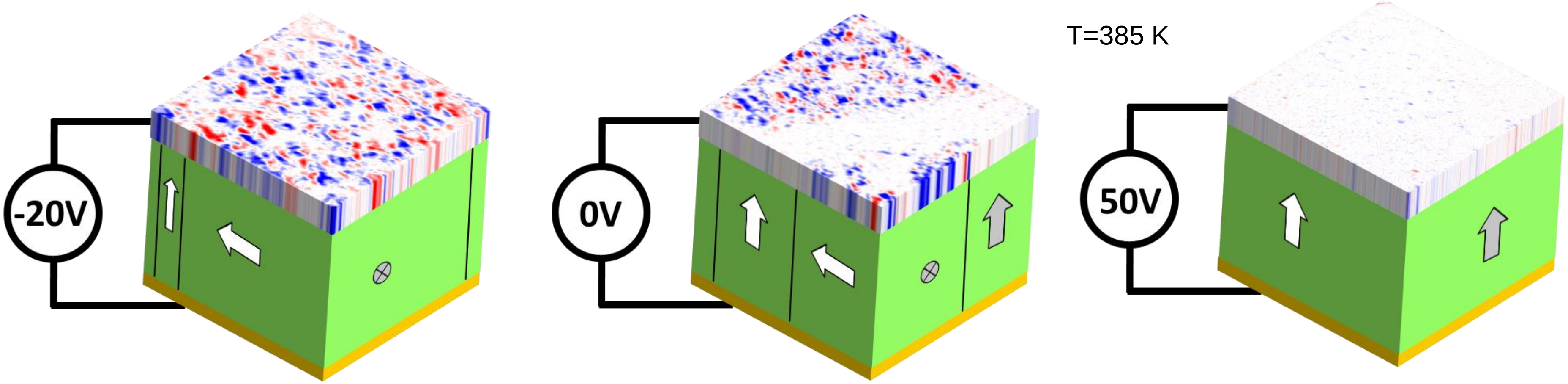
- At 0V at 20 kOe, $T^* \approx 360$ K
- Voltage shifts T^* by ~20K
- Effect is reversible
- Positive or negative voltages give roughly similar effect

- Max magnetization change ~600 emu/cm^3
- ME coupling $\alpha = 1.6.10^{-5}$ s/m
- Larger than in any single phase material by 5 orders
- Larger than in any artificial multiferroic by factor >10

Cherifi, MB et al, Nature Mater. 13, 345 (2014)

Strain-induced control of magnetic order

Direct imaging of magnetic state using XCMD-PEEM



Ferromagnetic state

Mixed ferro/ antiferromagnetic state

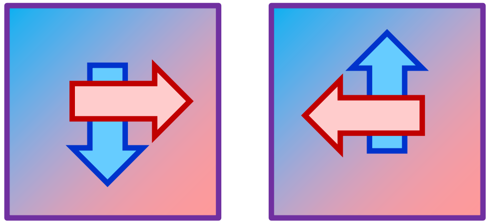
Antiferromagnetic state

- Switch ferromagnetism **OFF and ON** by electric field, just above room temperature

Phillips, MB et al, Sci. Rep. 5, 10026 (2014)

Different approaches for E-field control of magnetism

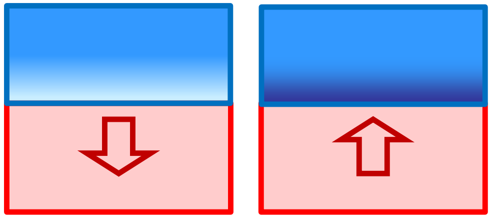
Intrinsic magnetoelectric



Use single-phase multiferroic material

✓ Simple approach, just one material
✓ Beautiful physics, potential for new science
✘ BFO only RT multiferroic
✘ Can be leaky, hard to switch

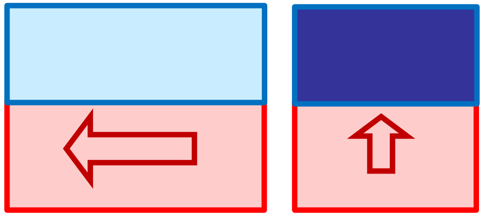
Field-effect



Combine strong ferroelectric with carrier-mediated ferromagnet

✓ Broader choice of materials
✓ Well-suited for perpendicular transport
✘ Few ferromagnetic oxides with high T_C ; need simple metals
✘ Effect occurs over very small thickness (few nm max)
✘ Needs very large fields

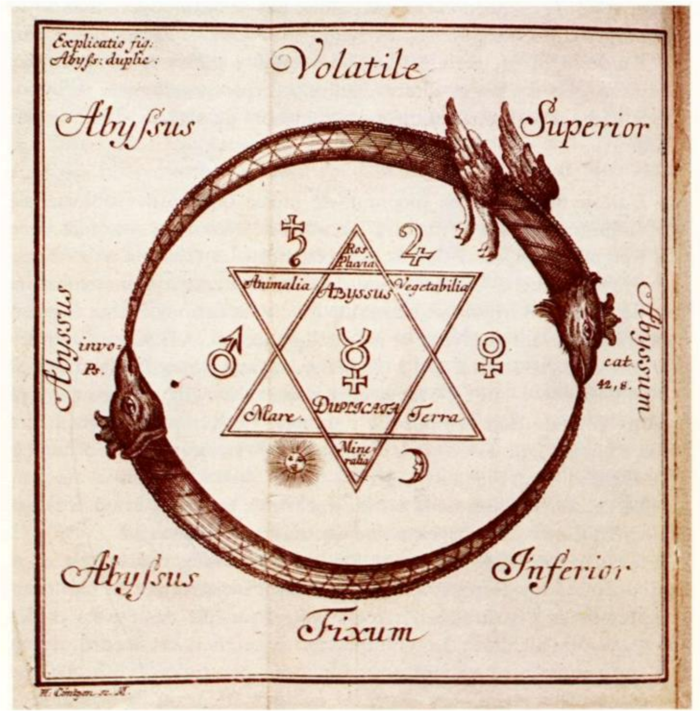
Strain-driven



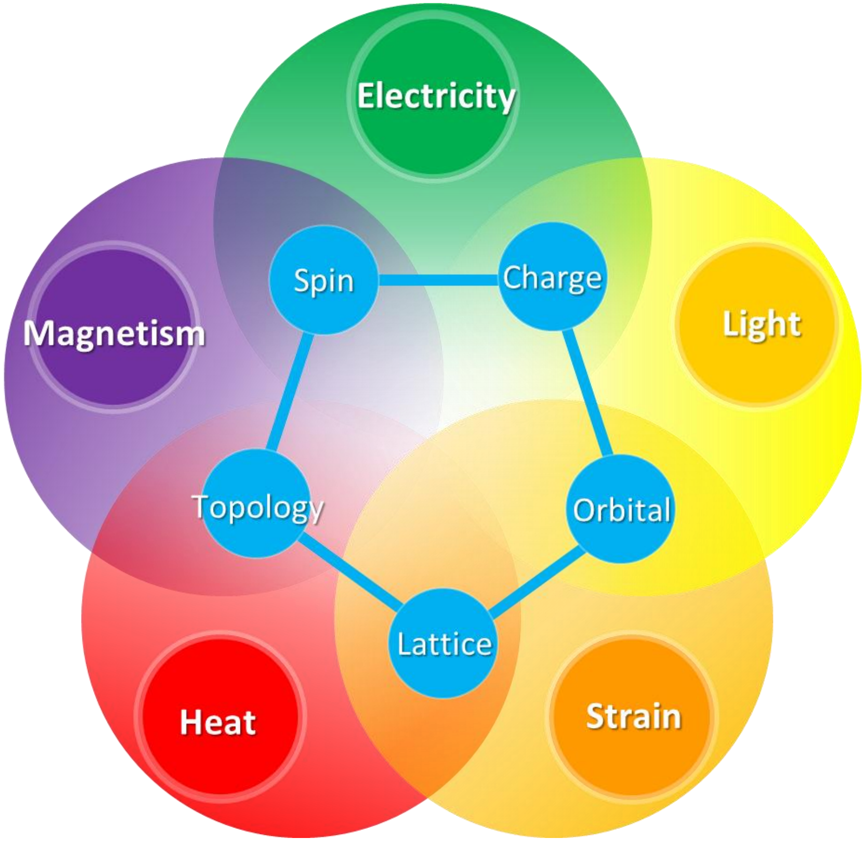
Combine piezoelectric or ferroelectric/ferroelastic with magnetostrictive ferromagnet

✓ Broader choice of materials
✓ Effect occurs over whole FM film
✘ Fatigue + low endurance
✘ Hard to miniaturize

Different approaches for E-field control of magnetism



Hermes Trismegistus « Emerald tablet »

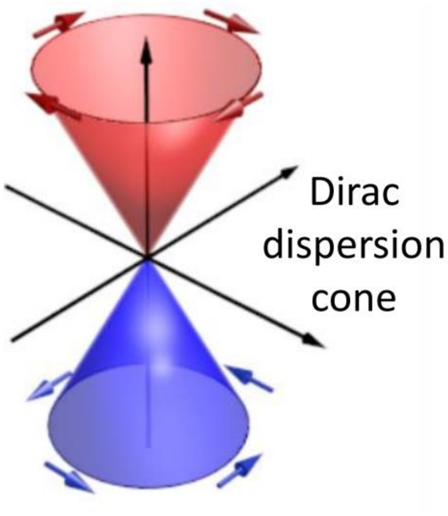


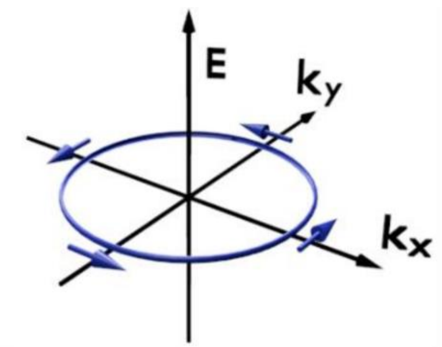
Yoshinori Tokura « Quantum Science on Strong Correlation Report 2014 »

- ⊙ New approaches / new ingredients
- ➔ Interconvert charge and spin currents using **spin-orbit coupling**
- ➔ Engineer **topological spin-textures** controlable by electric fields

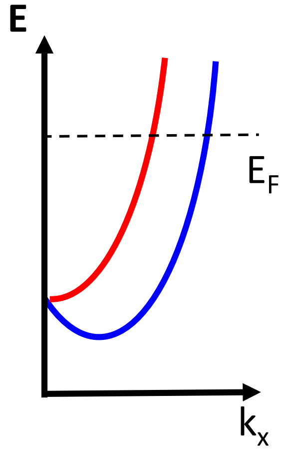
Fermi contours in 2DEGs

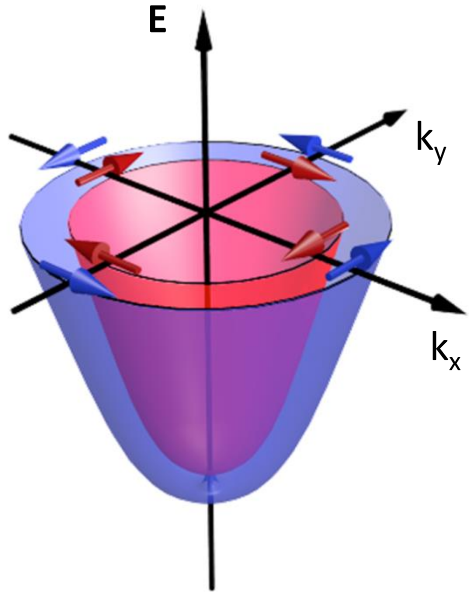
Topological insulator

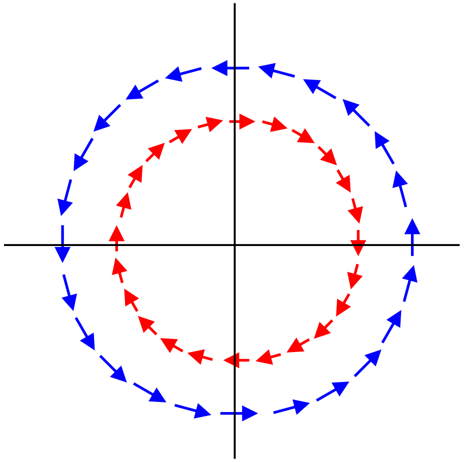


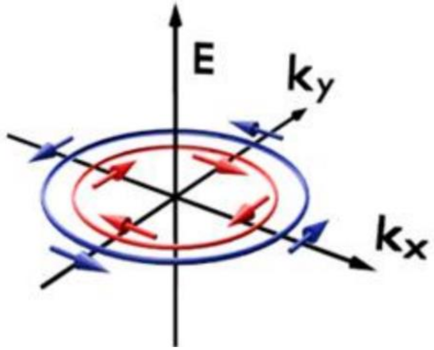


Rashba interface







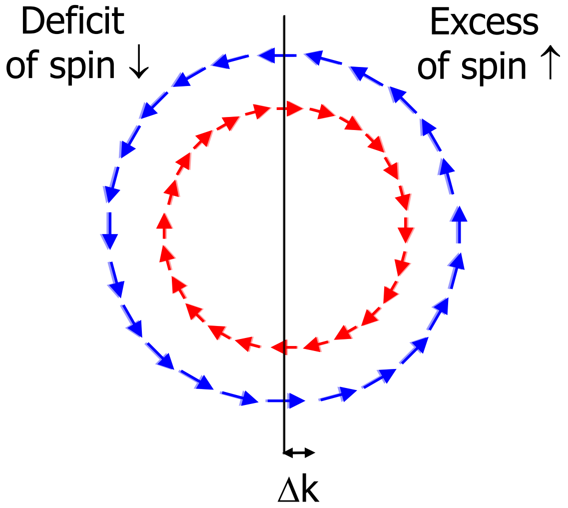


Zhang et al, Nature Phys. 5, 438 (2009)
Xia et al, Nature Phys. 5, 398 (2009)

E. I. Rashba, Sov. Phys. Solid State 2, 1109 (1960)
Yu. A. Bychkov & E. I. Rashba, Sov. Phys. JETP Lett. 39, 78 (1984)

Direct and inverse Rashba-Edelstein effects

Direct Rashba-Edelstein effect

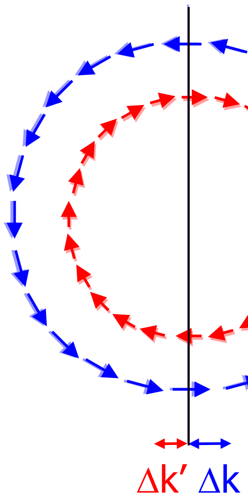


- Inject a charge current J_C

➔ Generation of a spin accumulation ➔ Spin current J_S

V. M. Edelstein, Solid State Commun. 73, 233 (1990)
A. G. Aronov & Y. B. Lyanda-Geller, JETP Lett. 50, 431 (1989)

Inverse Rashba-Edelstein effect



- Inject a spin current J_S
- Inequivalent shift of both Fermi contours

➔ Generation of a charge current J_C

J.-C. Rojas-Sánchez et al., Nat. Commun. 4, 2944 (2013)
K. Shen et al., Phys. Rev. Lett. 112, 096601 (2014)

- 3D spin current produces 2D charge current

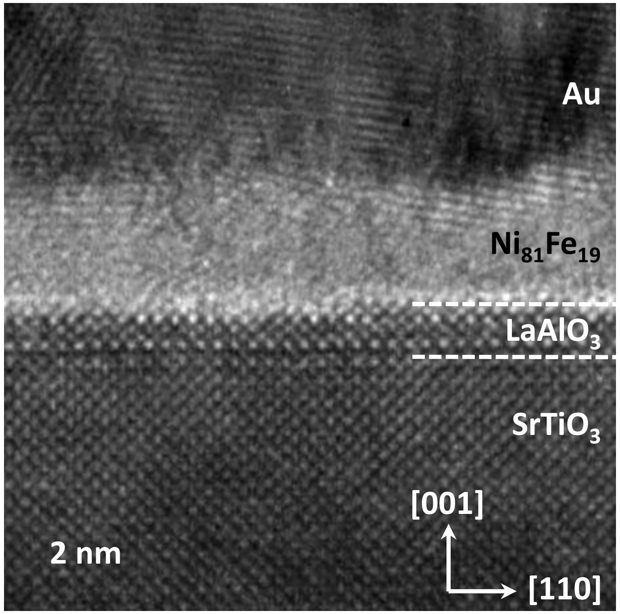
➔ **figure of merit is a length**

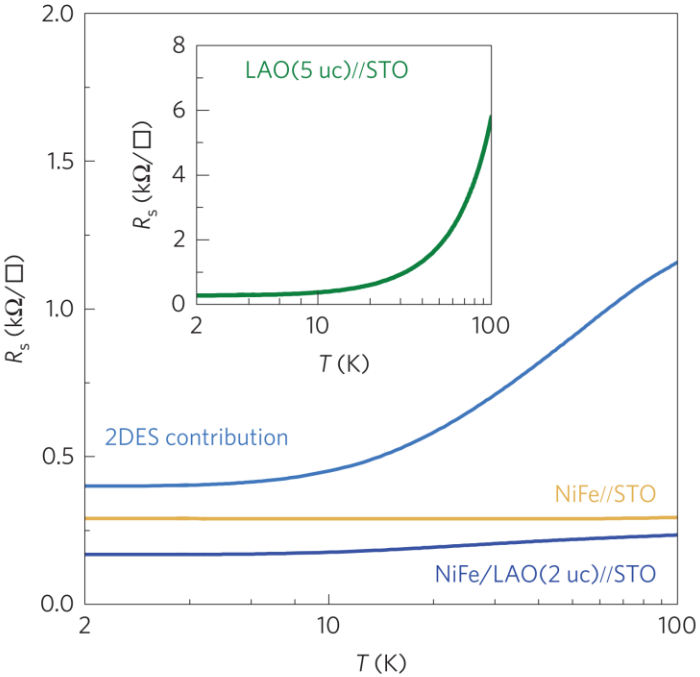
$$\boxed{\lambda_{\mathrm{IEE}} = \frac{j_c}{j_s} = \frac{\alpha_R \tau}{\hbar}}$$

α_R : Rashba coeff.
τ : scattering time

$LaAlO_3/SrTiO_3$ interface system

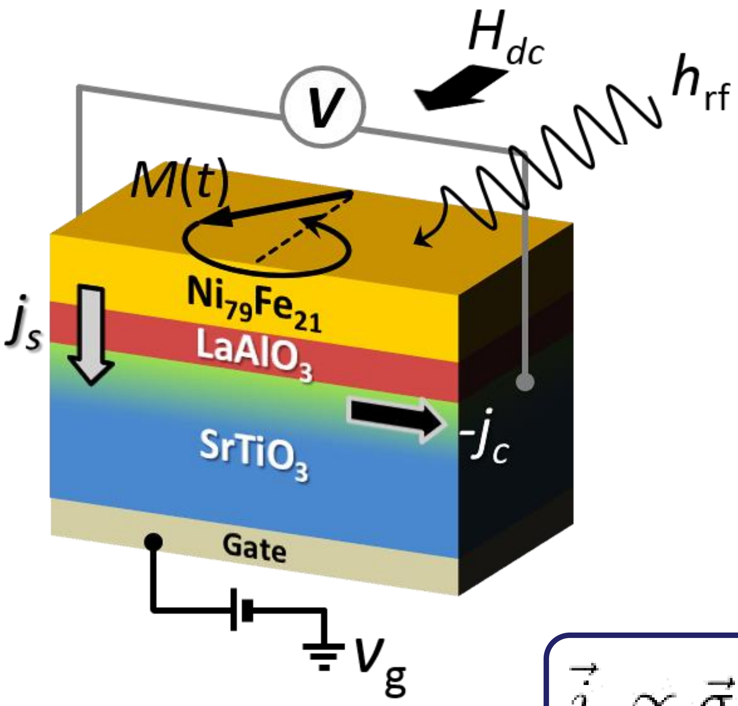
- Well-known oxide interface system Ohtomo & Hwang, Nature 427, 423 (2004)
- 2-dimensional electron gas forms at interface despite both LAO and STO being two insulators
- High mobility (>1000 cm²/Vs) and low carrier density (~10^{13} cm^{-2})
- Gate-tuneable **Rashba spin-orbit coupling** A.D. Caviglia et al., PRL. 104, 126803 (2010)



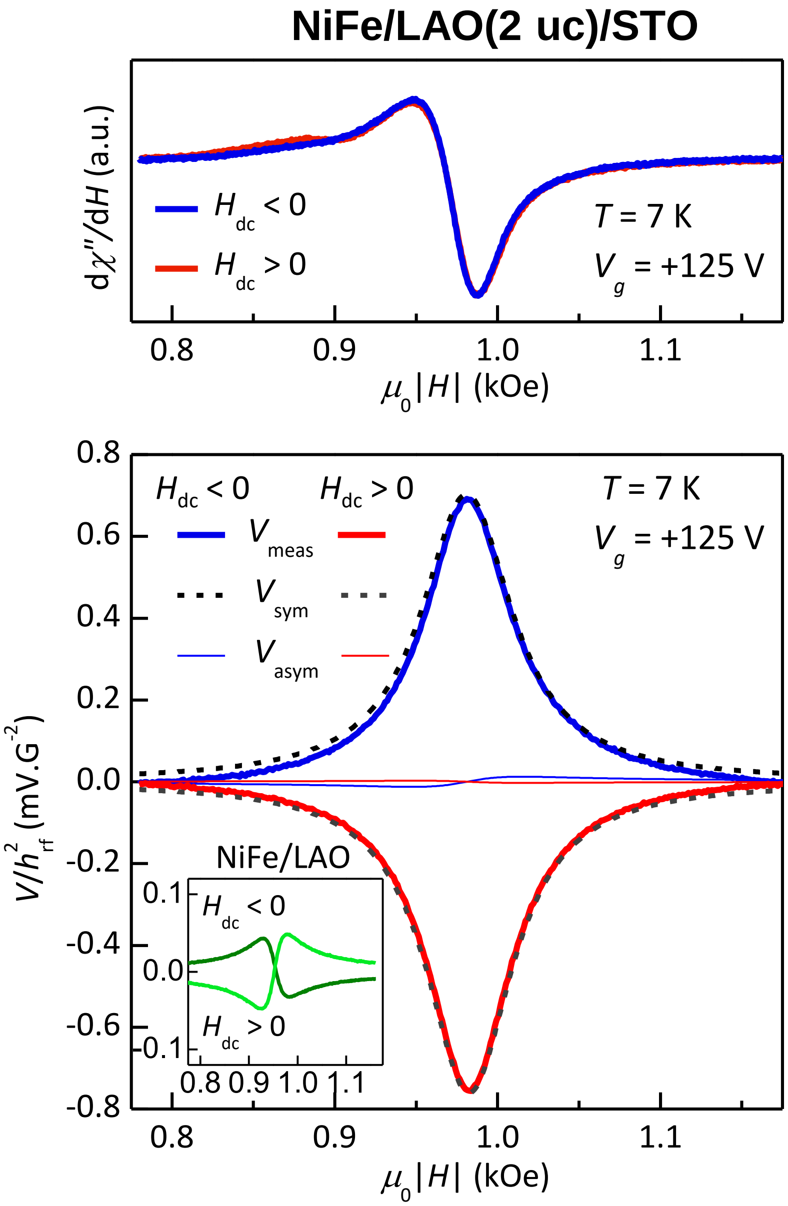


- Combine 2 unit-cells of LAO and 2 nm of NiFe (permalloy)
- TEM and AFM analysis indicates smooth surface and interfaces

FMR-spin pumping



$$\boxed{\vec{j}_c \propto \vec{\sigma} \times \vec{j}_s}$$

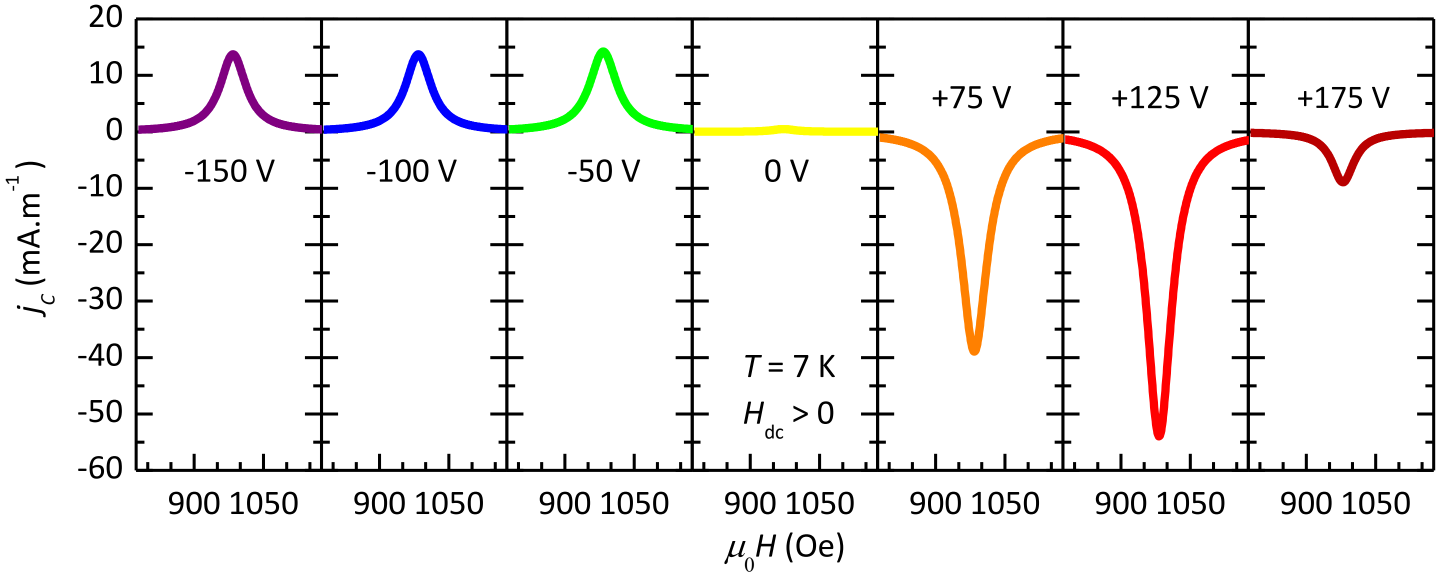


- Detected voltage:
- Symmetric Lorentzian shape
- Sign reversal upon $+H \rightarrow -H$ inversion

$+\sigma \rightarrow -\sigma$

$+V \rightarrow -V$

Spin to charge current conversion



- Efficient **spin-to-charge current conversion** via the Inverse Edelstein Effect (IEE) :

$$\lambda_{\mathrm{IEE}} = \frac{j_c}{j_s} = \frac{\alpha_{\mathrm{R}}\tau}{\hbar}$$

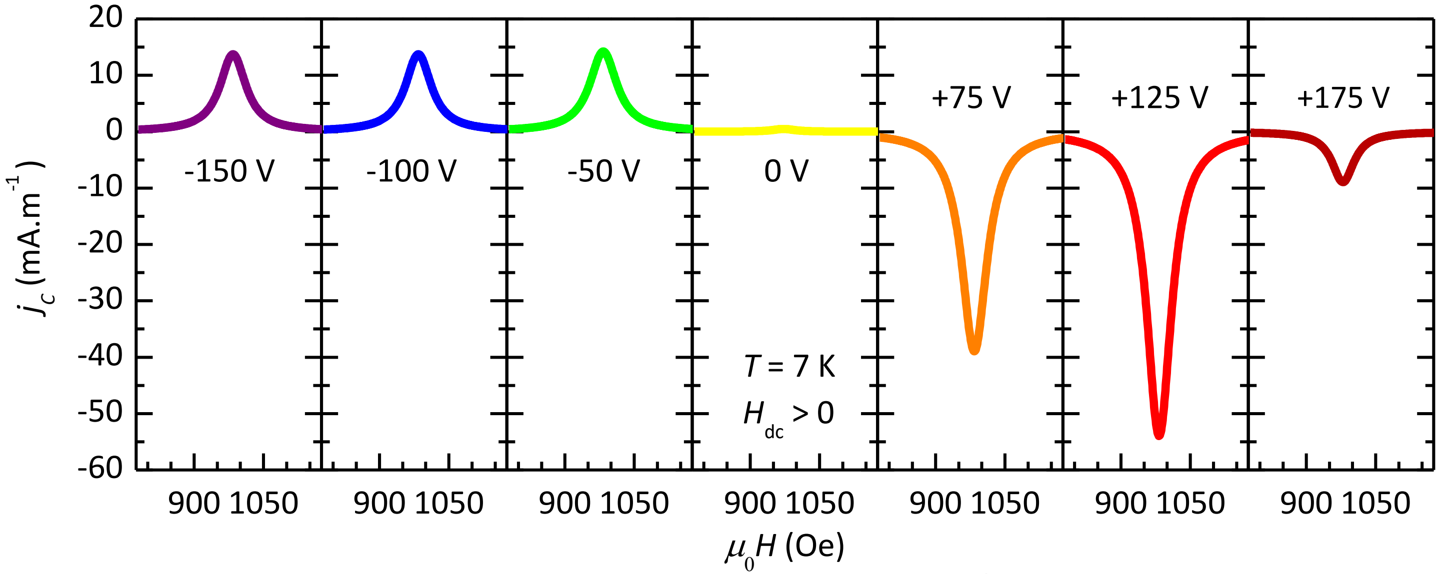
- Strong gate dependence, reminiscent of gate dependent Rashba coefficient in WAL data

A. D. Caviglia et al., Phys. Rev. Lett. 104, 126803 (2010)
A. Fête et al., Phys. Rev. B 86, 201105(R) (2012)

- Can one quantify λ_{IEE} ?

E. Lesne, MB et al., Nature Mater. 15, 1261 (2016)

Spin to charge current conversion



- Efficient **spin-to-charge current conversion** via the Inverse Edelstein Effect (IEE) :

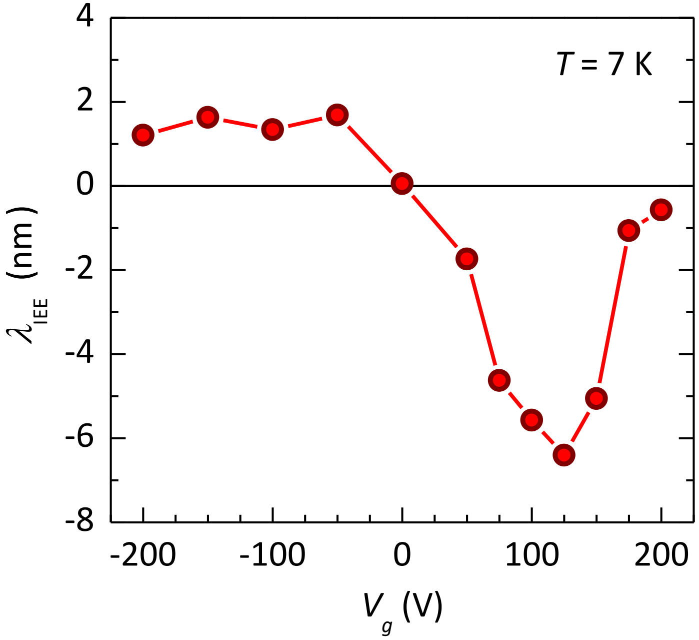
$$\lambda_{\text{IEE}} = \frac{j_c}{j_s} = \frac{\alpha_R \tau}{\hbar}$$

- **Largest efficiency reported for any material**

- Efficient larger than in Bi/Ag interface (λ_{IEE} =0.3 nm) and in topological insulator α-Sn (λ_{IEE} =4 nm)

  J.-C. Rojas-Sánchez et al., Nature Comm. 4, 2944 (2013)
  J.-C. Rojas-Sánchez et al., Phys. Rev. Lett. 116, 096602 (2016)

- Values of λ_{IEE} compatible with reported values of α_R~ 0.01-0.05 eV.Å and measured τ ~ 10-100 ps



E. Lesne, MB et al., Nature Mater. 15, 1261 (2016)

Conclusions and perspectives

- Oxide materials and heterostructures offer **many possibilities for E-field control of magnetism**
  - With multiferroics (works but at room temperature limited to $BiFeO_3$)
  - Through field effect (limited efficiency, requires ultrathin films and large fields)
  - Through field-controlled strain (efficient but hard to miniaturize)
  - New possibilities for **spin/charge interconversion** at oxide interfaces with **Rashba SO coupling**
    - Could work at room temperature ? Be larger in other systems than LAO/STO ?

Topological states at the (001) surface of $SrTiO_3$ M. Vivek et al, ArXiv 1702.05974

- Opportunities for devices in spintronics
  - Memory : future generation of MRAMs ?
  - **Memory into logic :** new spin-based transistors combining ME coupling + spin-charge conversion

Spin-Orbit Logic with Magnetoelectric Nodes S. Manipatruni et al (INTEL), ArXiv 1512.05428

- Opportunities for new physical effects
  - **Topology meets correlations**
  - In real space :
    - Topological spin textures ➔ new physical effects (topological Hall effect, top. orbital moment)
    - Effects amplified by correlations ?
    - Controllable through field-effect ?

Role of Berry-phase for describing orbital magnetism J. Hanke et al, PRB 94, 12114 (2016)
Interface-driven topological Hall effect in $SrRuO_3$-$SrIrO_3$ bilayer J. Matsuno, Sci. Adv. 7, e1600304 (2016)
Giant topological Hall effect from skyrmion bubbles in correlated manganite thin films, L. Vistoli, MB et al, submitted

  - In reciprocal space :
    - Novel types of topological materials (iridates, osmates, etc)
    - New state variables for information processing (beyond spin and charge)

Correlated quantum phenomena in the strong spin-orbit coupling regime W. Witczak-Krempa et al, Ann. Rev. 5, 57 (2014)