

Controlling spins with Electric field in Multiferroic architectures

Agnès Barthélémy

Unité Mixte de Physique CNRS/Thales, Palaiseau, France

Agnes.barthelemy@thalesgroup.com



<http://www.trt.thalesgroup.com/ump-cnrs-thales>

Why?



The electron has



a charge ($-e$)

a spin (\uparrow, \downarrow)

Electronics

Magnetism

Spintronics

Information is carried by

Charge

Magnetization

Electron spin

Control

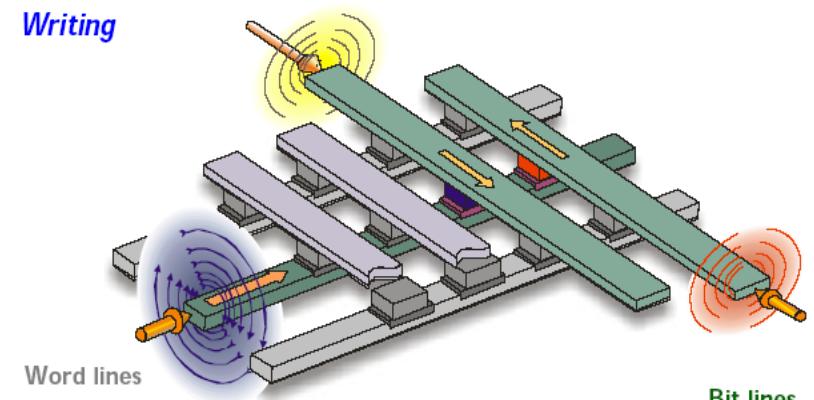
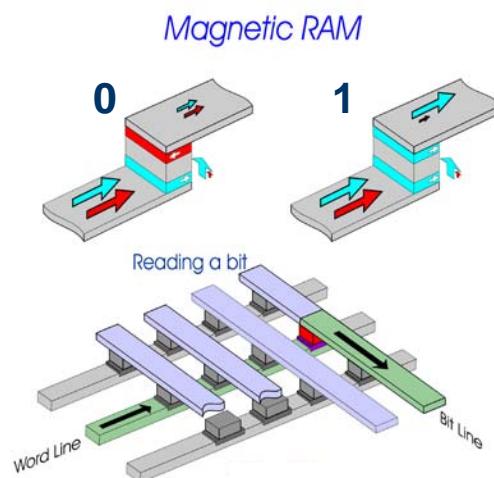
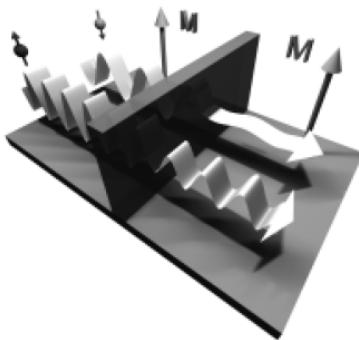
Electric field

Magnetic field

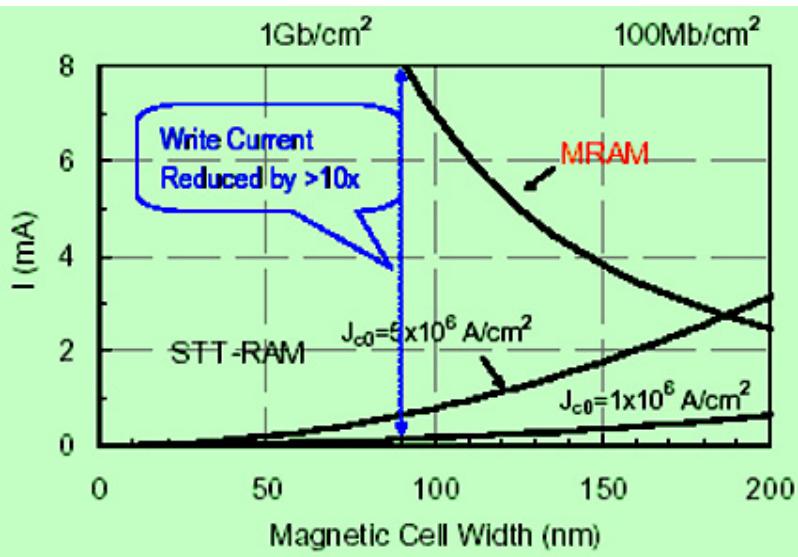
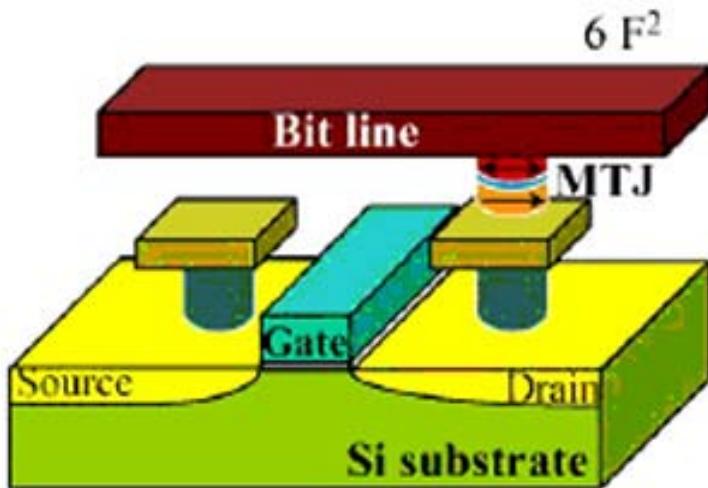
Magnetic field,
spin-polarized current

Key improvement in spintronics: Electric Control of magnetization or spin polarization

Non volatile Magnetic (Magnetoresistive) Random Access Memories (MRAMs)



Limitation for integration



From [http://www.embedded.com/design/real-time-and-performance/4026000/The-future-of-scalable-STT-RAM-as-a-universal-embedded-memory-\(Grandis\)](http://www.embedded.com/design/real-time-and-performance/4026000/The-future-of-scalable-STT-RAM-as-a-universal-embedded-memory-(Grandis))



STT-RAM architecture simpler, Size smaller
Reduction of the power



Nevertheless inevitably Joule heat losses

Other solution: E field control in heterostructures with ferroelectric or piezoelectric and magnetic materials: multiferroic architectures

Why?



The electron has

a charge ($-e$)

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Electronics

Information is carried by

Charge

Magnetism

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Spintronics

Electron spin

Control

Electric field

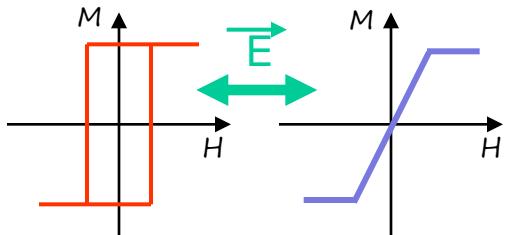
Magnetic field

Electric field

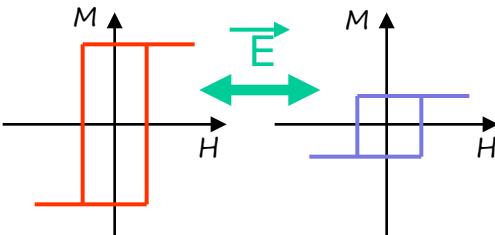
Controlling spins with electric field

Intrinsic Multiferroics or Artificial **multiferroic** heterostructures combining **ferroelectric** and **magnetic** materials:

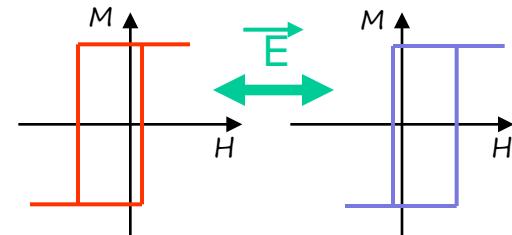
Magnetic anisotropy



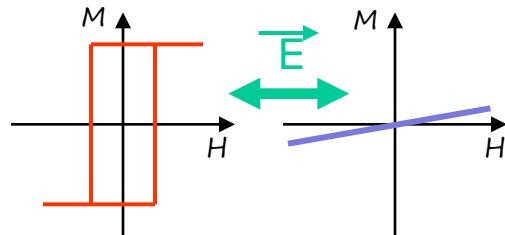
Magnetic moment



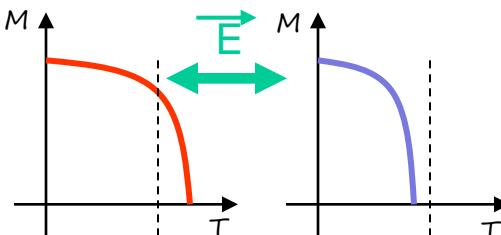
Exchange bias



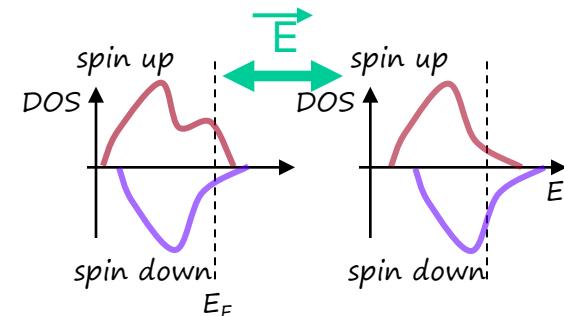
Magnetic order



Curie temperature



Spin polarization



$$P_{\text{spin}} > 0$$

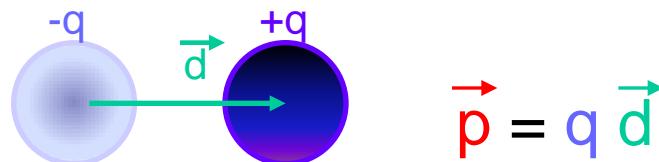
$$P_{\text{spin}} < 0$$

- Various magnetic properties can be controlled by electric field

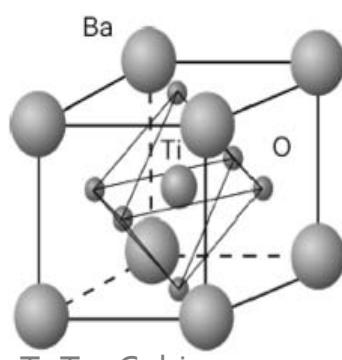
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Basics of Ferroelectricity / Piezoelectricity

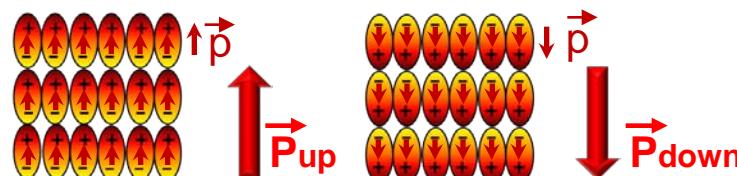
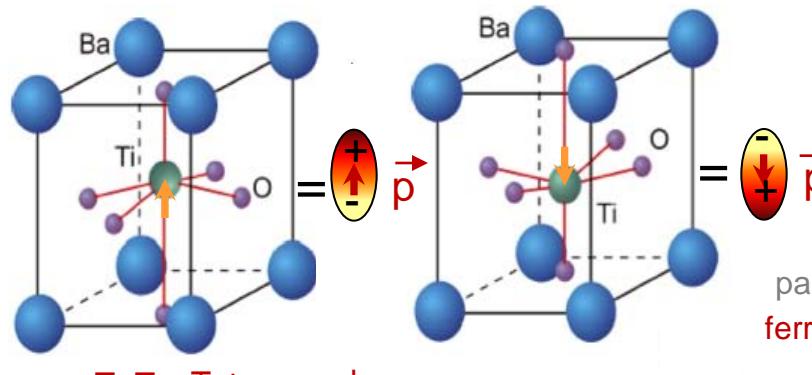
FM materials: magnetic moment $\vec{\mu}$ / FE (FerroElectric) materials: dipolar moment \vec{p}



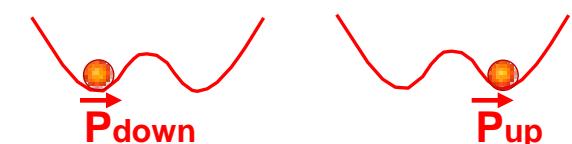
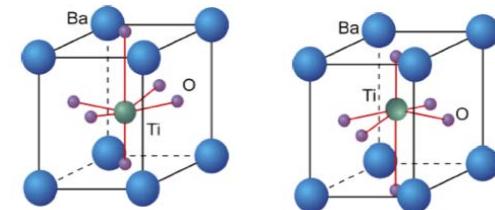
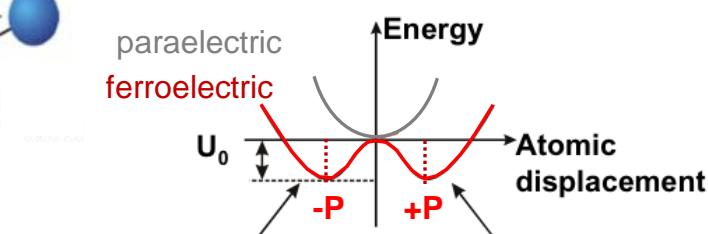
Prototypical FE: BaTiO_3



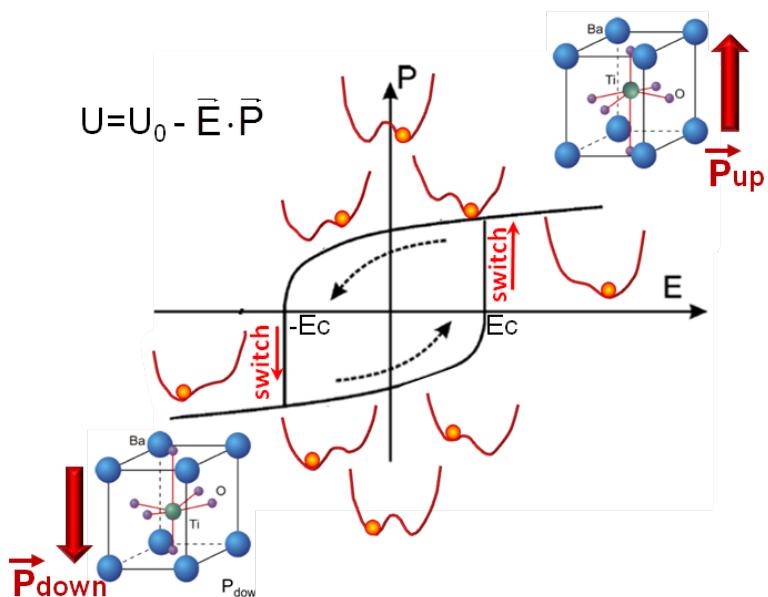
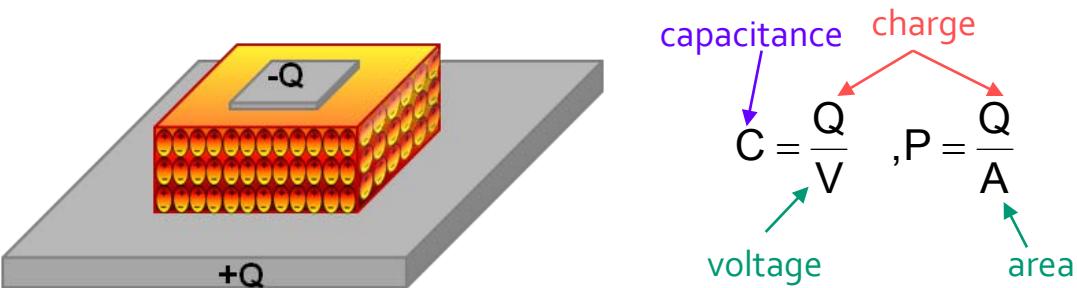
$T > T_c$: Cubic.
Paraelectric
 $P=0$



$$\Rightarrow \text{Polarization } \vec{P}: \quad \vec{P} = \frac{d\vec{p}}{dV} = \frac{\overrightarrow{p_{\text{unit cell}}}}{V_{\text{unit cell}}} \neq 0$$



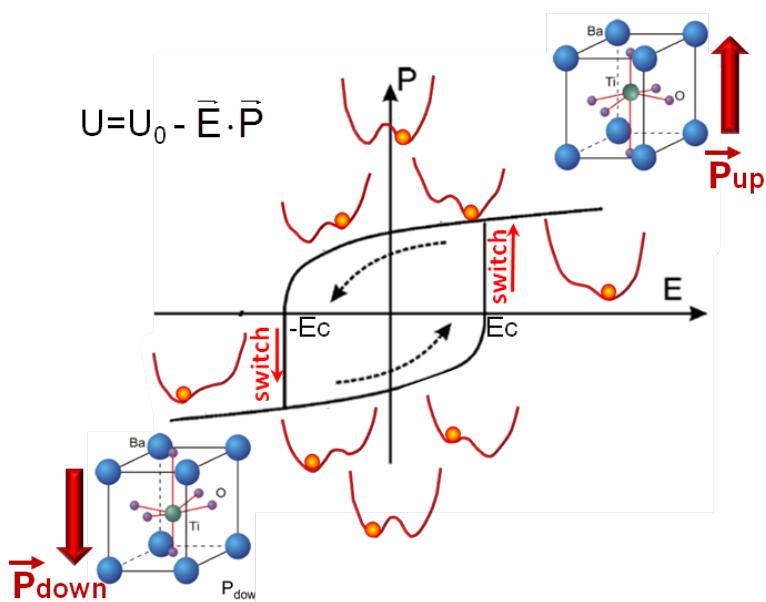
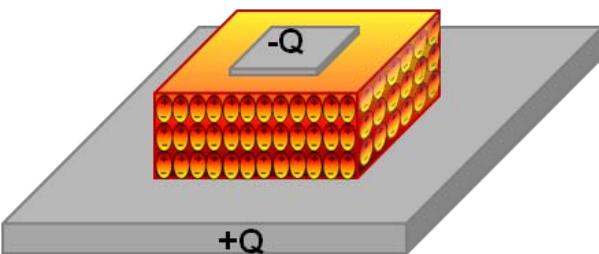
Polarization vs electric field loops



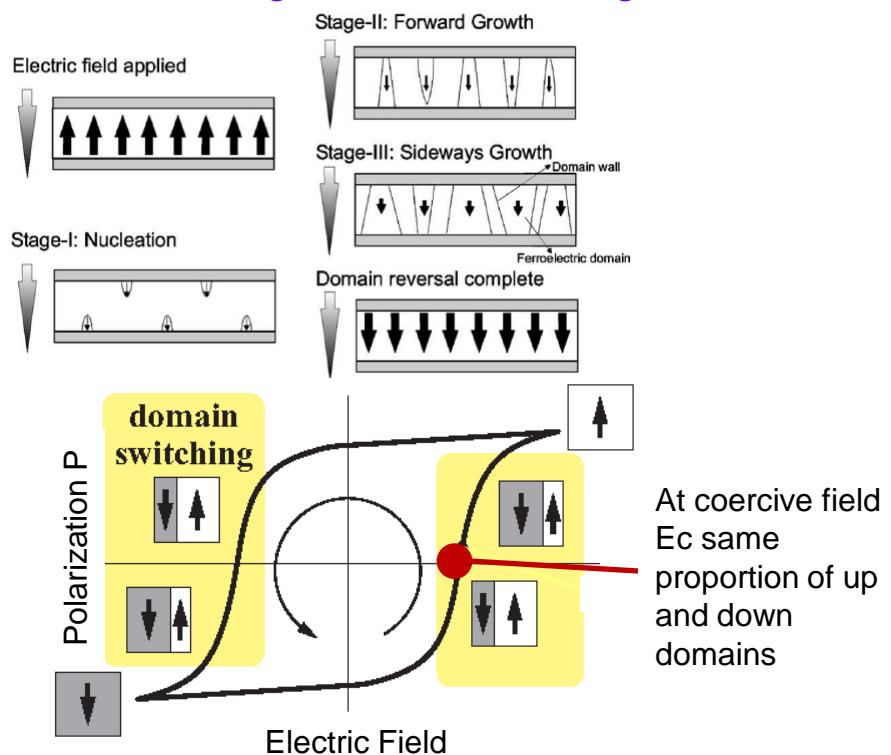
Very similar to the shape of magnetic loop
BUT not possible for the polarization to
rotate (always along a high symmetry axis).

Polarization vs electric field loops

Usually reversal through nucleation and growth of domains

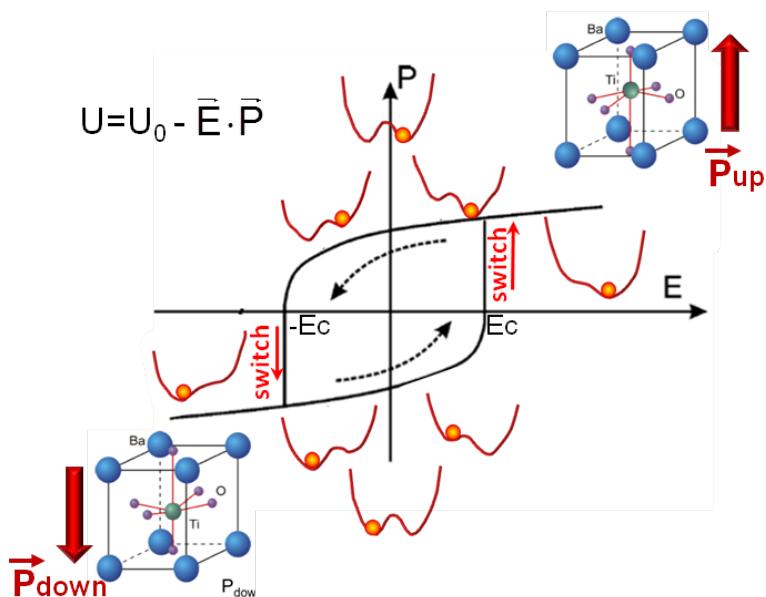
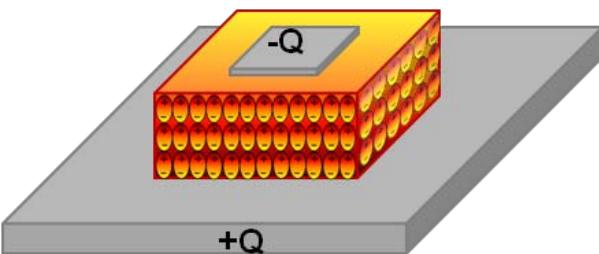


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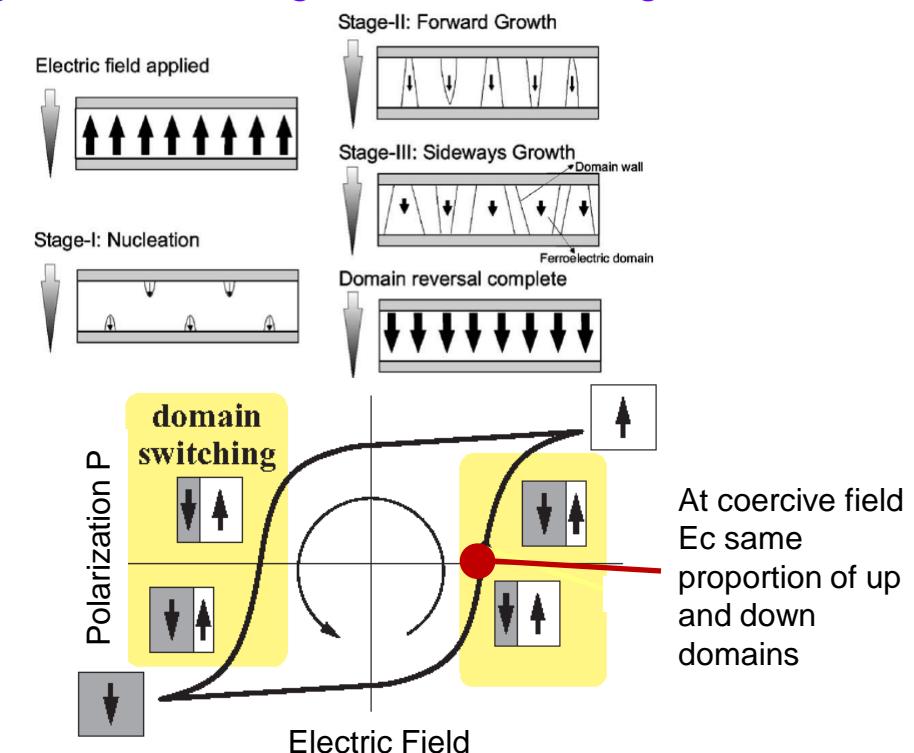


Polarization vs electric field loops

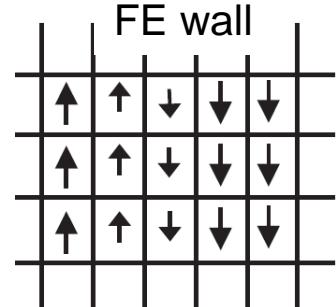
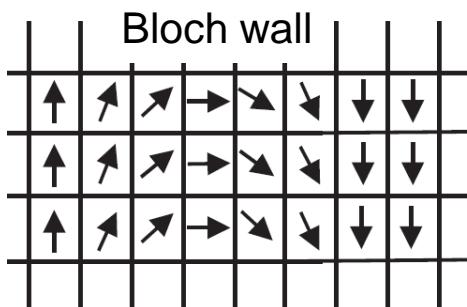
Usually reversal through nucleation and growth of domains



Very similar to Stoner Wohlfarth BUT **not possible for the polarization to rotate** (always along a high symmetry axis).

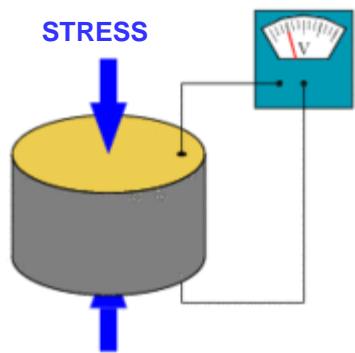


Another difference: FM DWs are large (hundreds of unit cells), **FE DWs are very thin** (few unit cells)

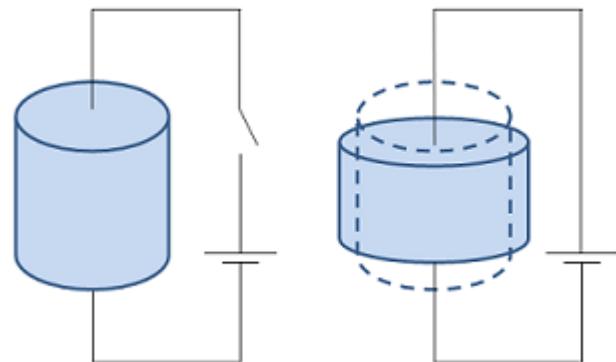


Every Ferroelectric material is a Piezoelectric material

Piezoelectric effect

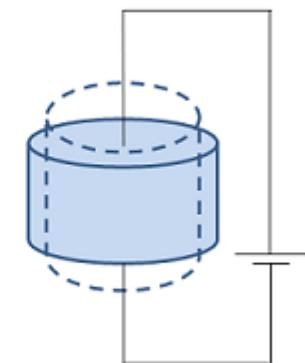
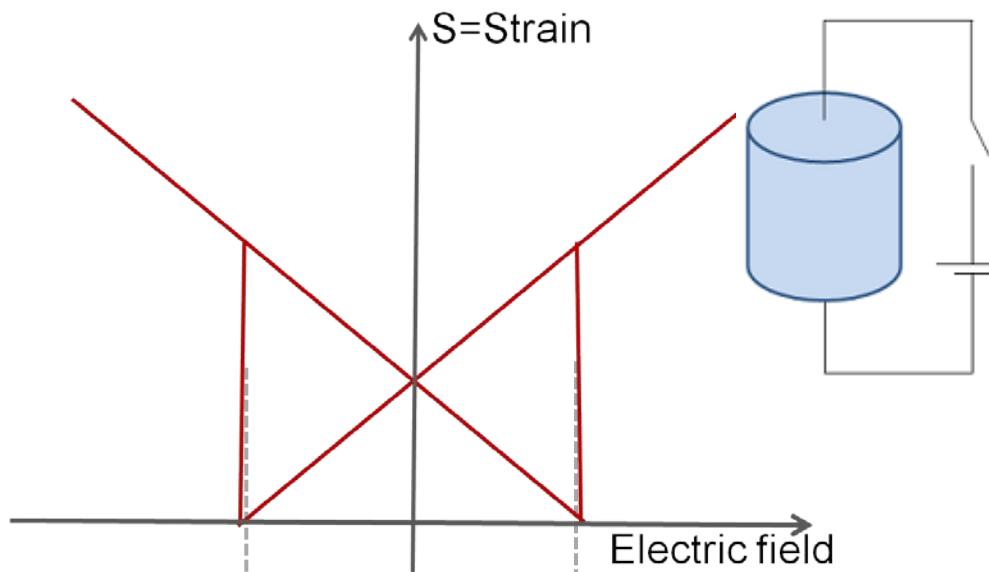


Converse Piezoelectric effect

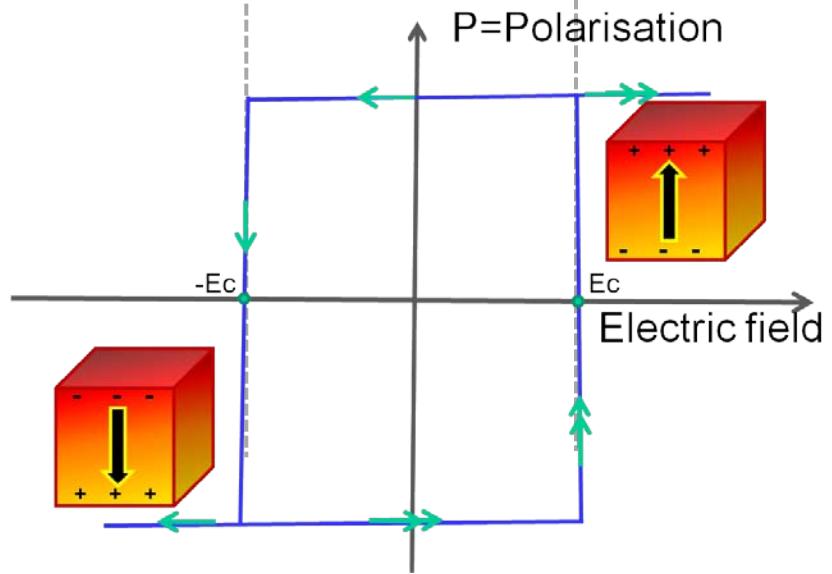


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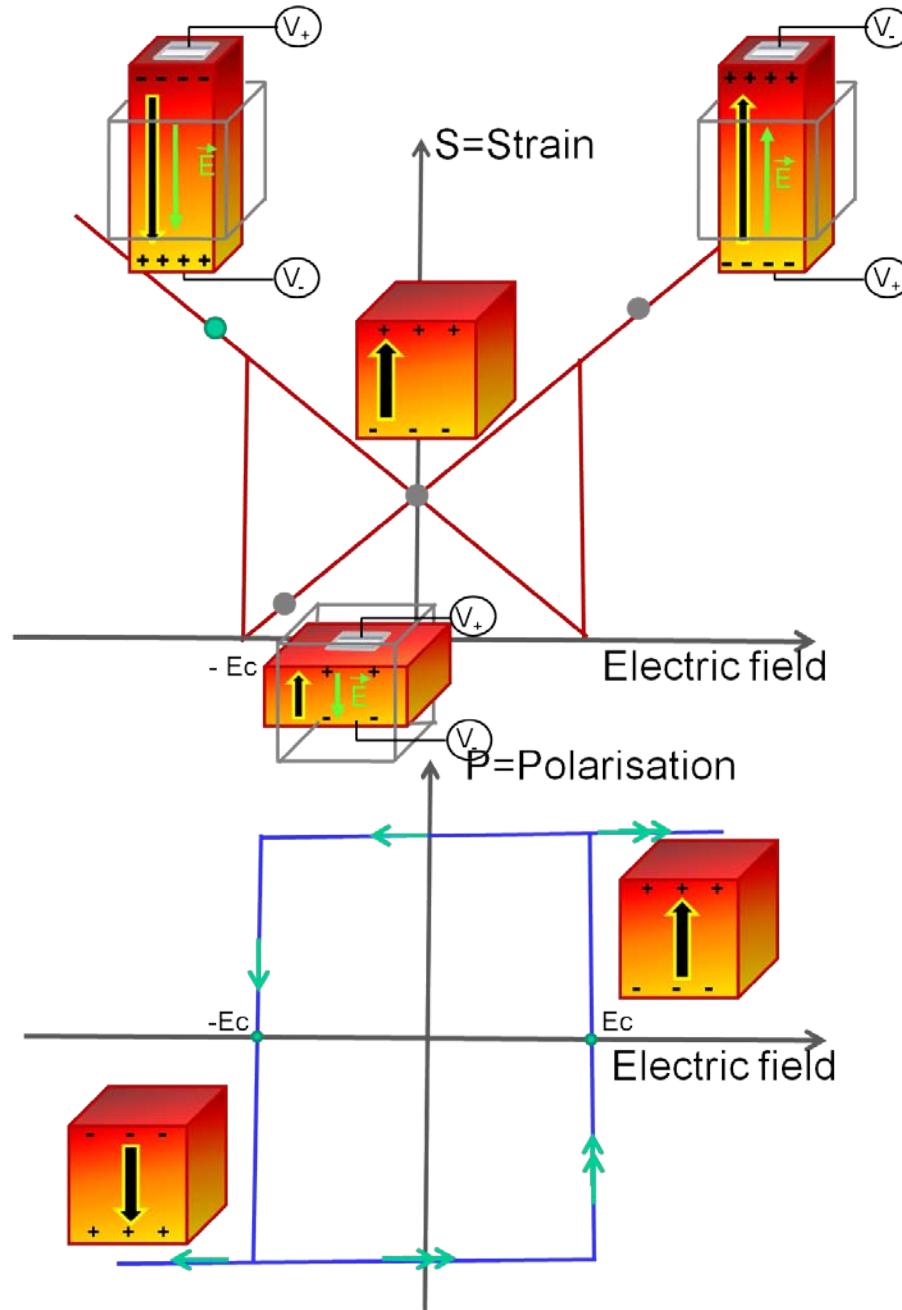
Converse Piezoelectric effect



$P = \text{Polarisation}$

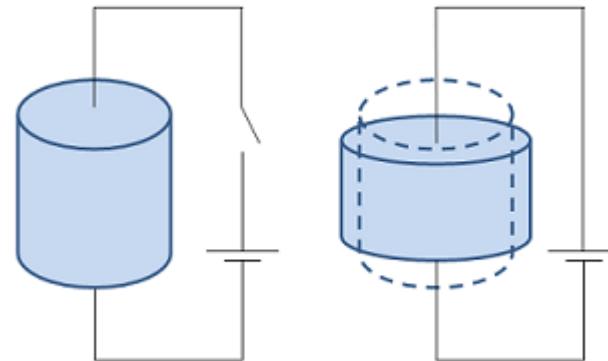
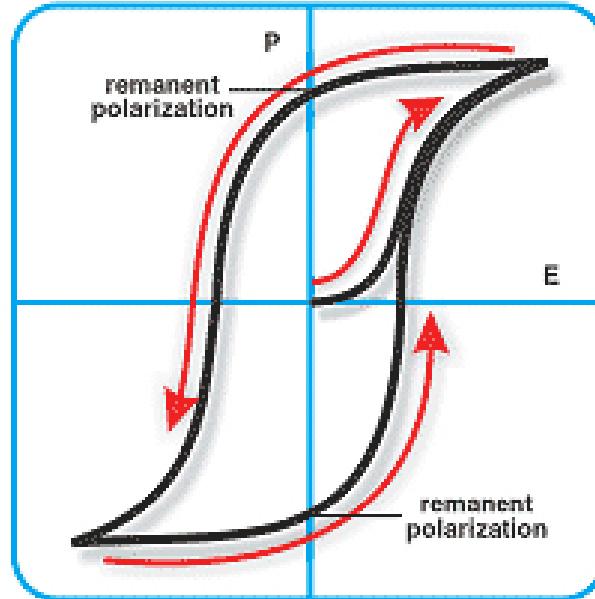
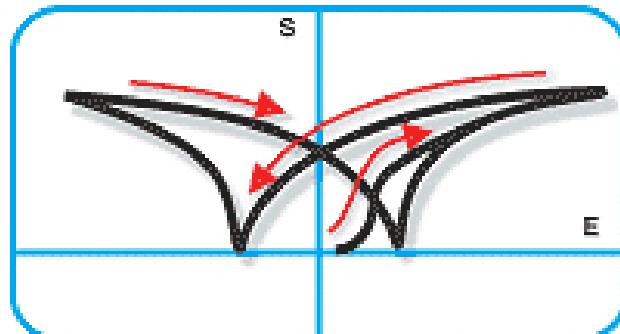


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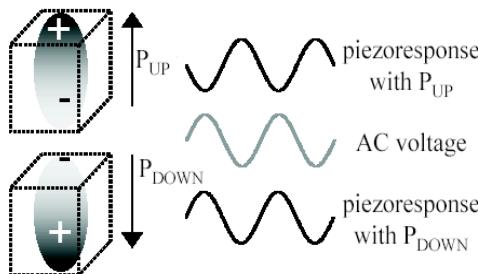
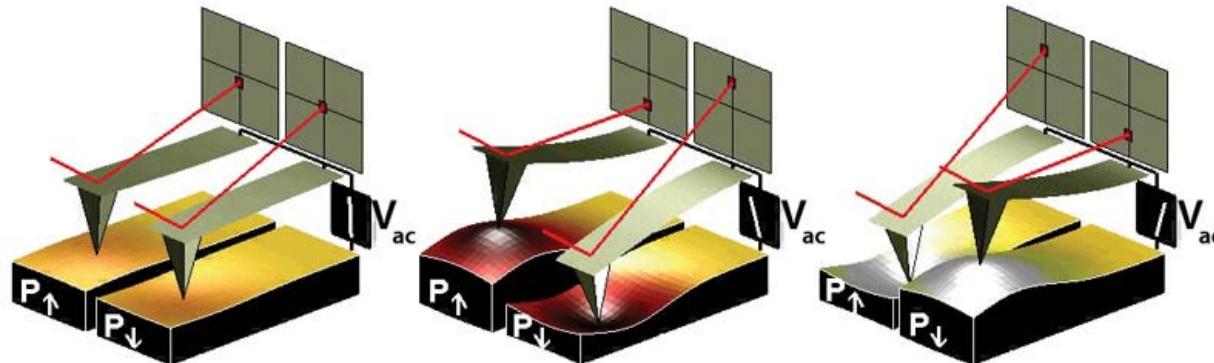
Every Ferroelectric material is a Piezoelectric material

Converse Piezoelectric effect



Effect used in actuator,
transducers, microsensors...

This piezoelectric character can be used to image ferroelectric domain:
Piezo-response force microscopy (PFM)



P_{up} domains : 180° out of phase with AC voltage

P_{down} domains in phase with AC voltage

$V = V_0 \cos(\omega t)$
 $\Delta Z = d_{33} V_0 \cos(\omega t + \varphi)$
 with $\varphi = 180^\circ$ for P_{up} domains
 and $\varphi = 0$ for P_{down} ones

Allows to image FE domains

Image of FE domains in BiFeO_3 $t_{\text{BFO}}/\text{(La,Sr)MnO}_3/\text{SrTiO}_3$ heterostructure

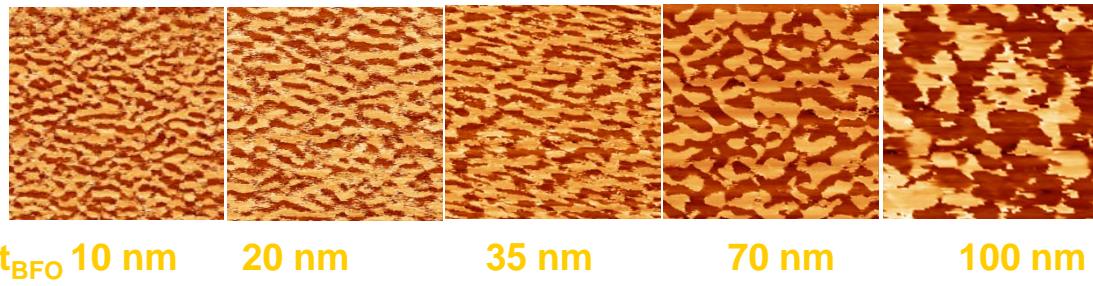
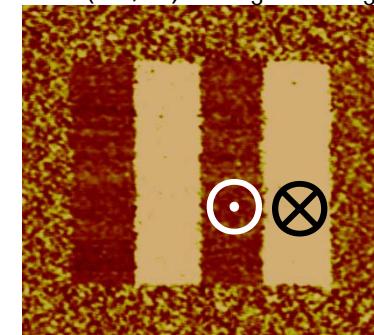


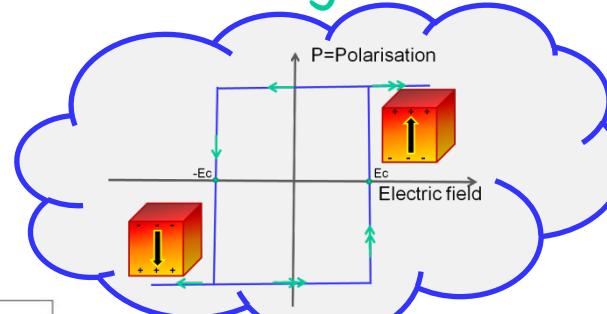
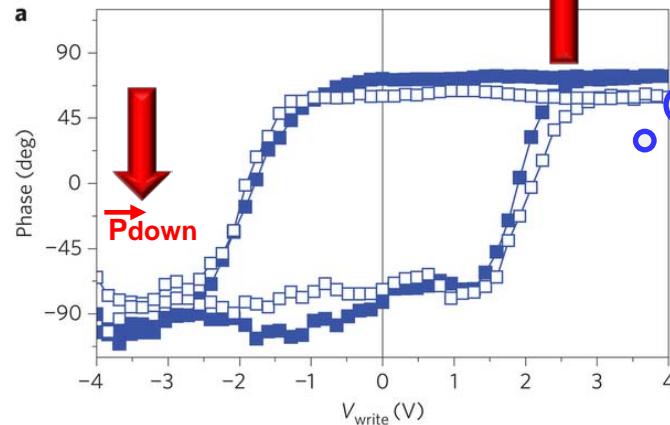
Image of written FE domains in BaTiO_3 $1\text{nm}/\text{(La,Sr)MnO}_3/\text{SrTiO}_3$ heterostructure



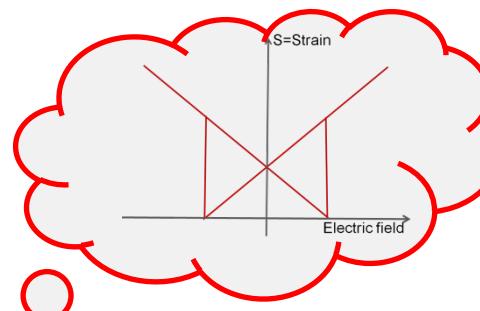
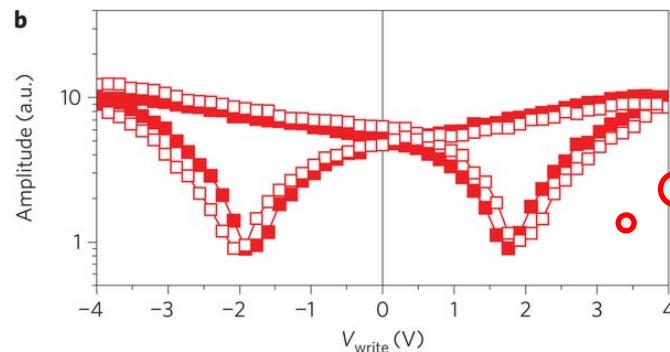
This piezoelectric character can be used to image ferroelectric loops: Piezo-response force microscopy (PFM)

Allows to determine E_c

$$V = V_{DC} + V_0 \cos(\omega t)$$



Phase cycle similar to polarization vs electric field loop: allows to deduce coercive field ($2V$ in that case)



Amplitude cycle similar to strain vs electric field loop: allows to deduce coercive field ($2V$ in that case)

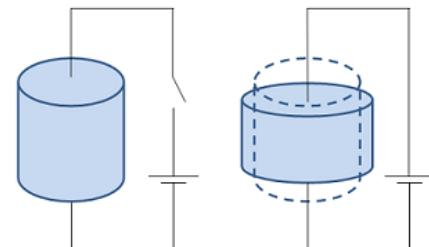
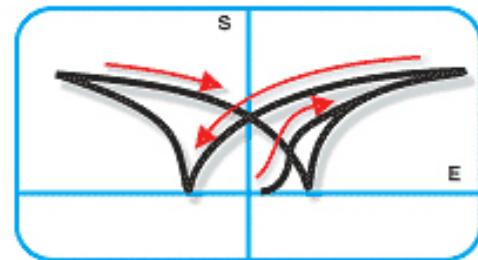
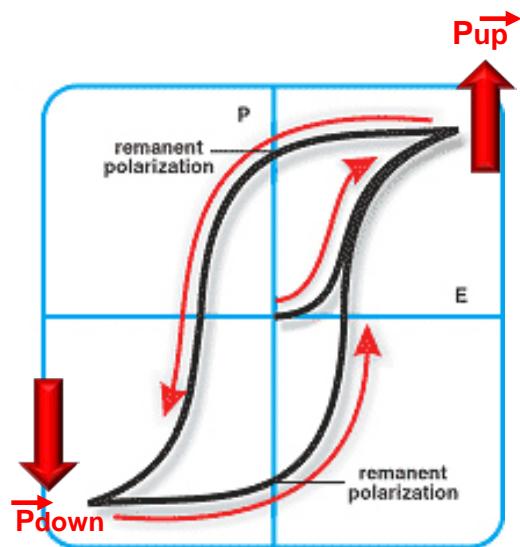
Evolution of Phase and amplitude of PFM signal for $\text{BiFeO}_3/\text{BaTiO}_3(2\text{ nm})/(\text{La},\text{Sr})\text{MnO}_3/\text{NdGaO}_3$ heterostructure

Chanthbouala et al.; Nature Nanotechnology 7, 101 (2012)

Material	Polarization ($\mu\text{C}/\text{cm}^2$)	Tc (K)
BaTiO ₃	26	393
PbTiO ₃	75	763
PbZr _{0.52} Ti _{0.48} O ₃ (PZT)	25	670
BiFeO ₃	100	1100

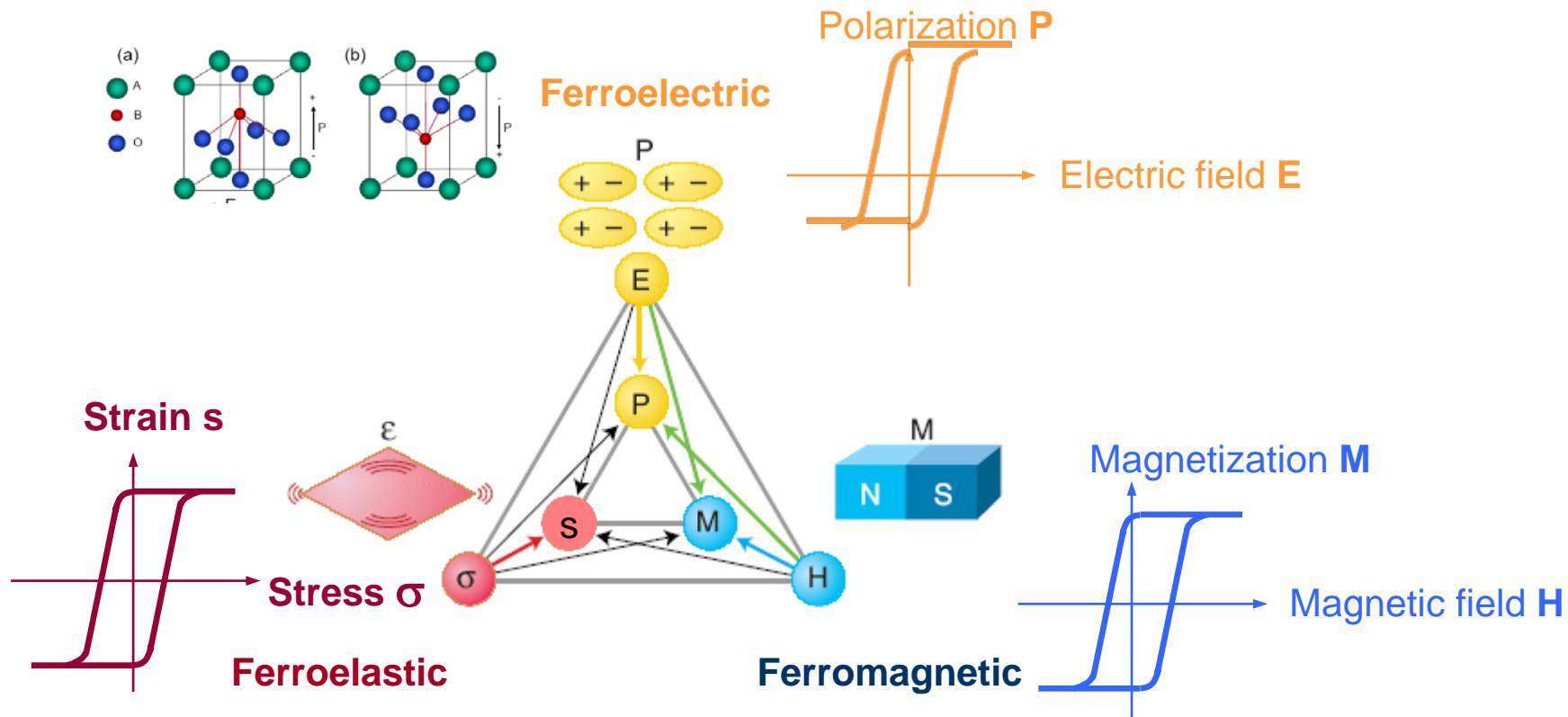
Sum up

- FE materials are characterized by their hysteresis loop $P(E)$:
- Two states at remanence: can be used to store information FERAM (equivalent to MRAM): FERAM= capacitor with Pup or down: disadvantage: necessary to reverse the polarization to read whereas in MRAM: information simply read by measuring the resistance
- As in FM materials reversal through domain nucleation and expansion
- Polarization $\Rightarrow \exists$ of charges on surface $Q = P \times A \rightarrow$ can be used to control magnetism
- Also Piezoelectric: their size changes when an electric field is applied:
 - Used to design actuators, transducers, sensors...
 - Used to image FE domains in PFM experiments
 \rightarrow can also be used to control magnetism: straintronics



Multiferroics : definition

H. Schmid, *Ferroelectrics* 162, 317 (1994): "Crystals can be defined as multiferroic when two or more of the primary properties are united in the same phase"



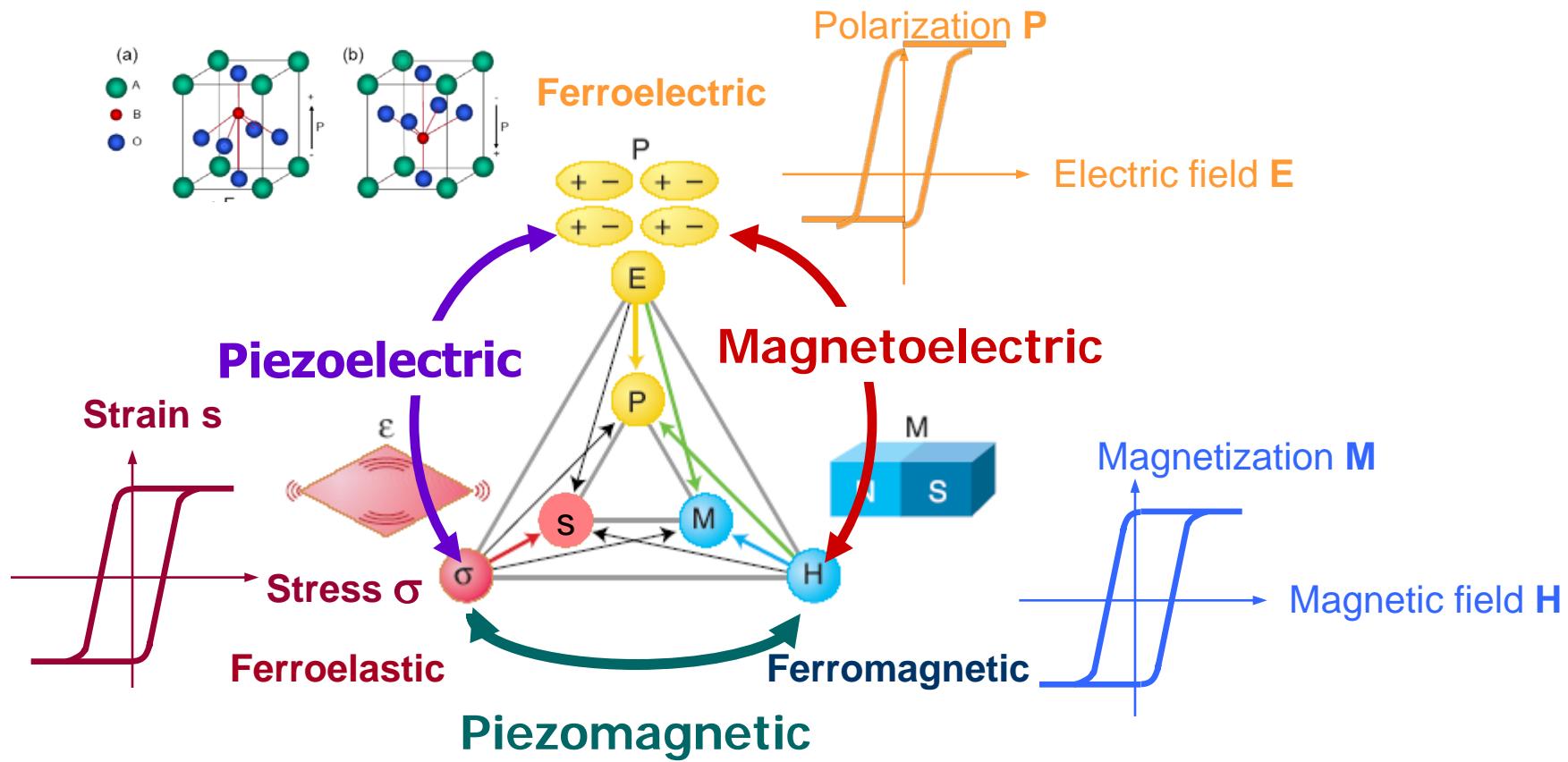
Definition generally enlarge to antiferroic orders

Intrinsic multiferroic (BiFeO_3 , BiMnO_3 , YMnO_3 ...) or **artificial**: combination of FE and magnetic

Reviews: M. Fiebig; *J. Phys. D Appl. Phys.* 38, R123 (2005); N. Spaldin and M. Fiebig; *Science* 309, 391 (2005); W. Eerenstein et al.; *Nature* 442, 759 (2006); K. F. Wang et al.; *Adv. in Phys.* 58, 321 (2009)

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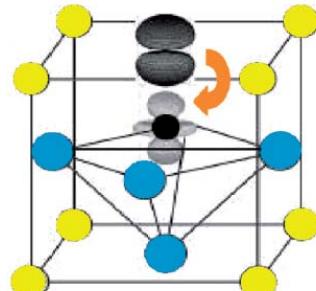
1. Few materials are Multiferroics

In perovskite ABO_3 : Ferroelectricity related to the displacement of the TM atom from the center of the O_6 octaedra to form a strong covalent bond: only possible for d^0 TM

On the contrary magnetism necessitates d^N atom

N. Hill; J. Phys. Chem. B, 104 6694 (2000)

Solution: A cation responsible of FE character & B cation at origin of magnetism



2. Most of them are AFM or WFM (Noticeable exceptions of $\text{La}_x\text{Bi}_{1-x}\text{MnO}_3$, CoCr_2O_4)

	magnetism		ferroelectricity	
	Type of order	Critical temperature (K)	Type of order	Critical temperature (K)
YMnO_3	AF	70	FE $P=5.5 \mu\text{C}/\text{cm}^2$	920
TbMnO_3	AF	42	FE $P=80 \text{nC}/\text{cm}^2$	27
$(\text{La},\text{Bi})\text{MnO}_3$	FM $M=3.6\mu_B/\text{f.u}$	105	FE $P=0.1-1 \mu\text{C}/\text{cm}^2$	770
BiFeO_3	AF, WF	640	FE $P=100 \mu\text{C}/\text{cm}^2$	1100



3. Low (magnetic) critical temperature

4. All of them do not present magnetoelectric coupling

$$\mathbf{F} = \mathbf{F}_0 - \vec{\mathbf{P}}_s \cdot \vec{\mathbf{E}} - \mu_0 \vec{\mathbf{M}}_s \cdot \vec{\mathbf{H}} - \frac{1}{2} \epsilon_0 \chi_E \mathbf{E}^2 - \frac{1}{2} \mu_0 \chi_M \mathbf{H}^2 - \alpha \vec{\mathbf{E}} \cdot \vec{\mathbf{H}}$$

$$\mu_0 \mathbf{M} = - \frac{\partial \mathbf{F}}{\partial \mathbf{H}} = \mu_0 \mathbf{M}_s + \mu_0 \chi_M \mathbf{H} + \alpha \mathbf{E} \quad \text{with } \alpha = \mu_0 \frac{\partial \mathbf{M}}{\partial \mathbf{E}} = \frac{\partial \mathbf{P}}{\partial \mathbf{H}} \text{ in T.m.V}^{-1} = \text{s.m}^{-1}$$

and coupling limited by “Cannot be larger than the geometric mean of electric and magnetic permeability” *Brown et al.; Phys. Rev. 168, 574 (1968)*

Review: Fiebig; J. Phys. D 38, R123 (2005)

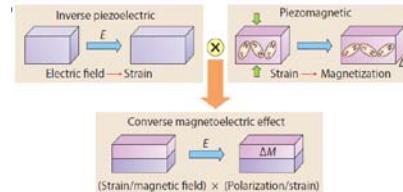
$$\alpha^2 < \epsilon_0 \mu_0 \chi_E \chi_M$$

Solution: artificial multiferroic architecture: combination of FE and magnetic materials

Mechanisms of control of magnetism by ferroelectricity:

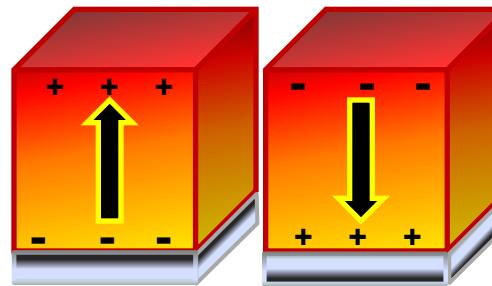
In artificial multiferroics FE/FM architectures through:

- ✓ strain-mediated coupling

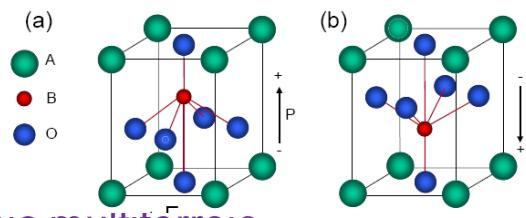
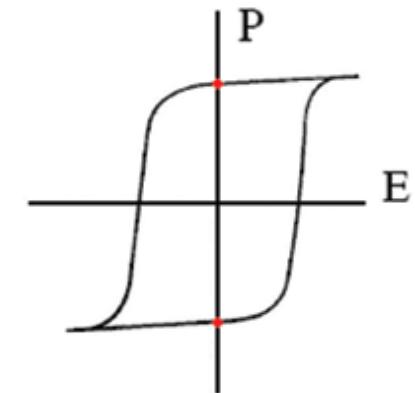


Wang et al.; NPG Asia Mater. 2, 61 (2010)

- ✓ effect of polarization direction on electronic structure of FM:
→ Field effect: accumulation/depletion



→ Different hybridization

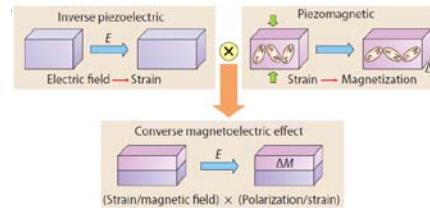


- ✓ direct coupling using an intrinsic multiferroic

Mechanisms of control of magnetism by ferroelectricity:

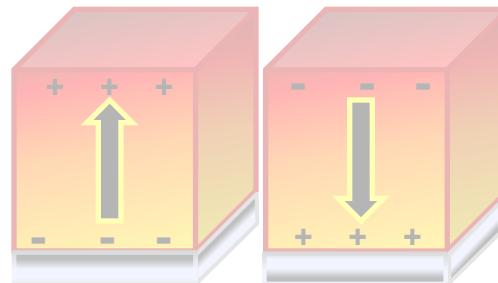
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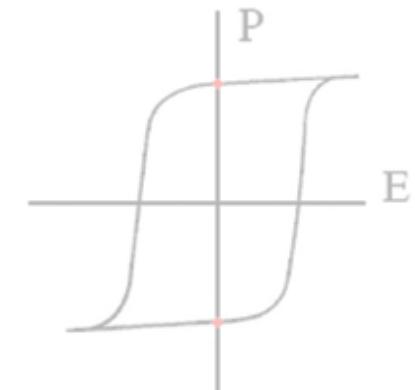
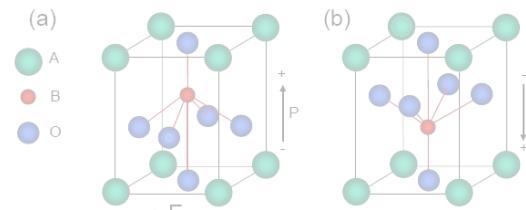


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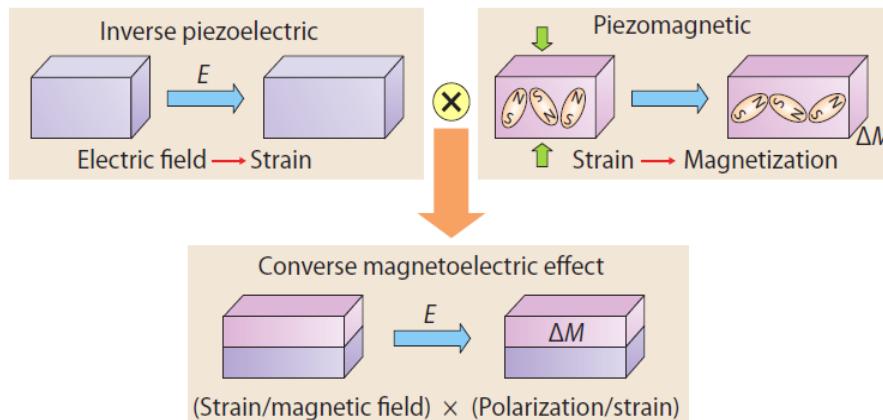


- ✓ direct coupling using an intrinsic multiferroic

Mechanisms:

In artificial multiferroics **FE/FM** architectures through:

- ✓ strain-mediated coupling

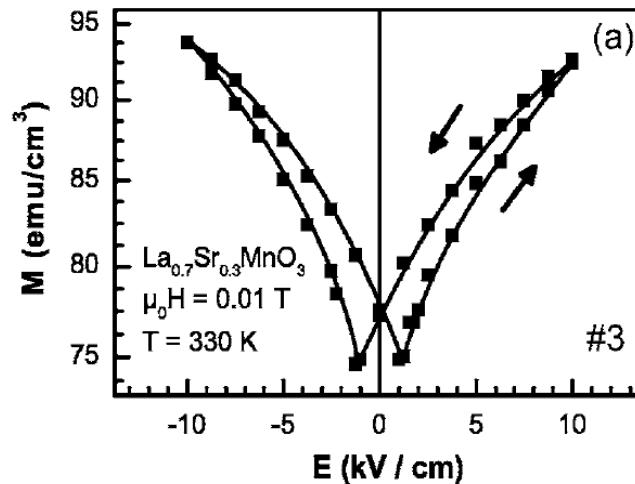
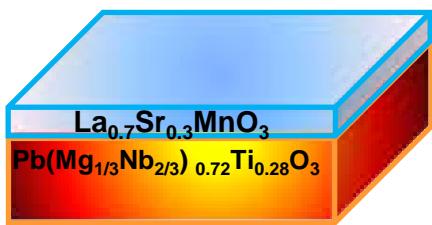
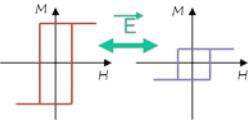


Wang et al.; NPG Asia Mater. 2, 61 (2010)

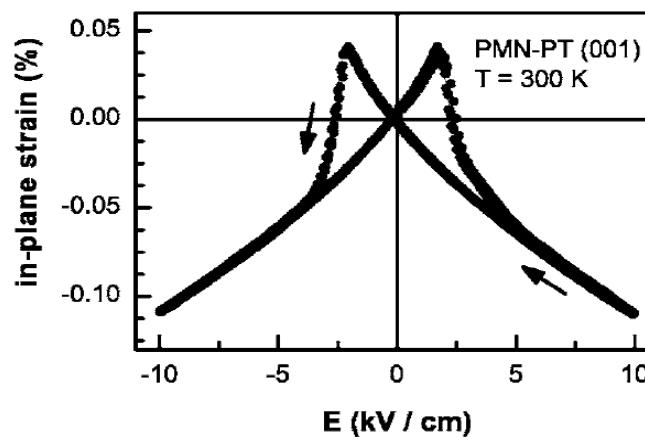
$$\text{Converse ME effect} = \frac{\text{Mechanical}}{\text{Electric}} \times \frac{\text{Magnetic}}{\text{Mechanical}} = \text{piezoelectric} \times \text{magnetostriuctive}$$

$$\mu_0 \Delta M = \alpha \Delta E$$

Magnetic moment

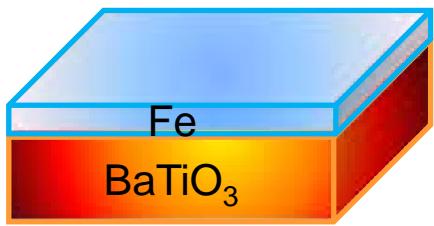
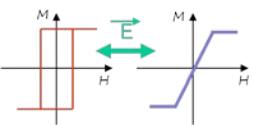


Reflects the piezoelectric loop

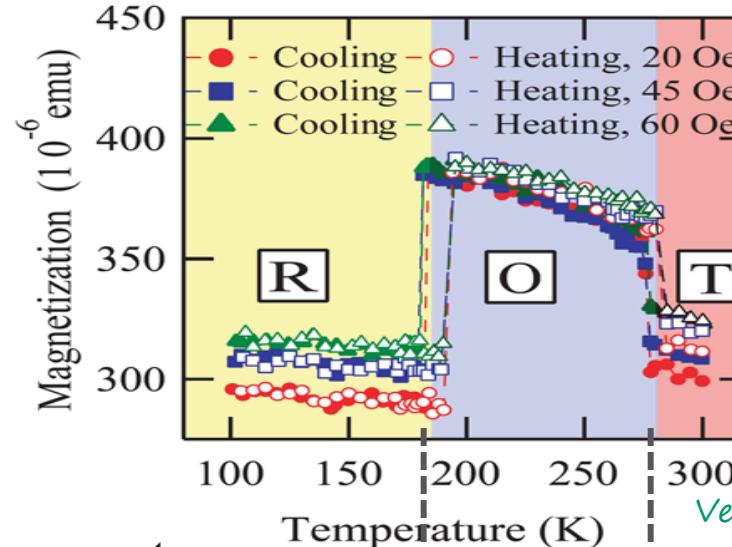


Example: Fe//BaTiO₃(001): control of magnetic anisotropy

Magnetic anisotropy

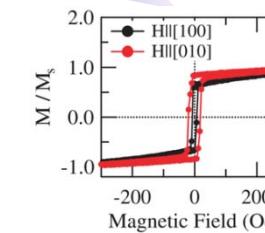
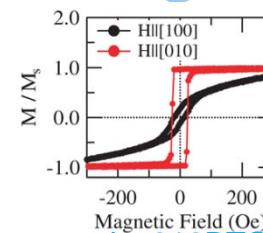
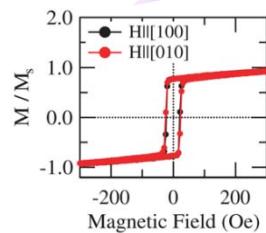
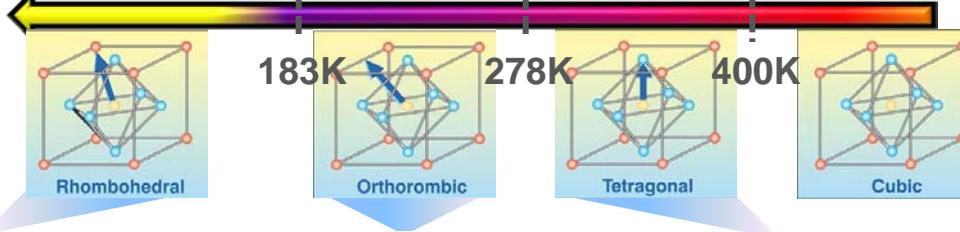


Epitaxial Fe is rotated by 45° on BaTiO₃(001):
(100) BaTiO₃ ≡ (110) Fe

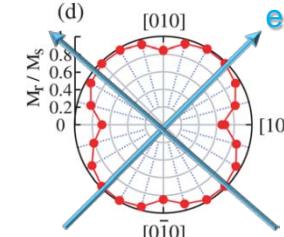
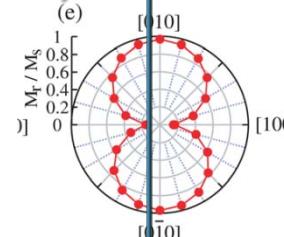
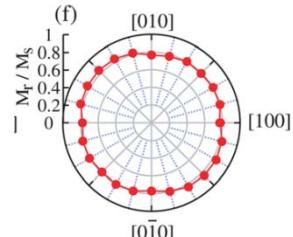


Reflects the ≠ strains
states imposed by the ≠
phases of BaTiO₃

Venkataiah et al.; APL 99, 102506 (2011)



easy axis: 010BTO=110Fe

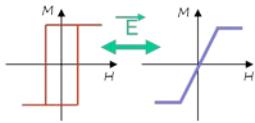


easy axis: 110BTO=001Fe

Shirahata et al.; APL 99, 022501 (2011)

BaTiO_3/Fe ; BaTiO_3 in T phase

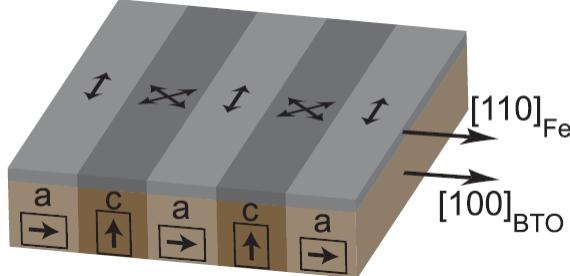
Magnetic anisotropy



c domains

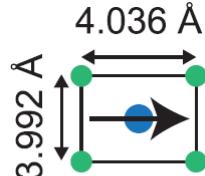
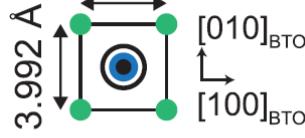


a domains

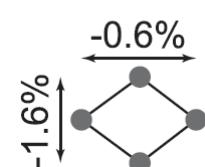
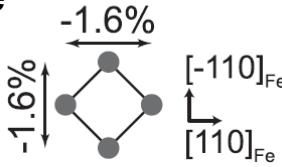


BTO

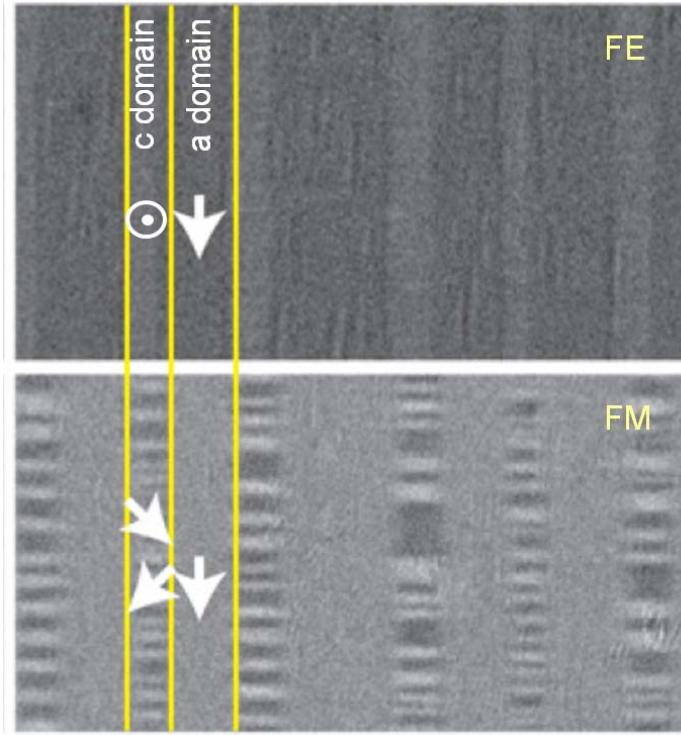
3.992 Å



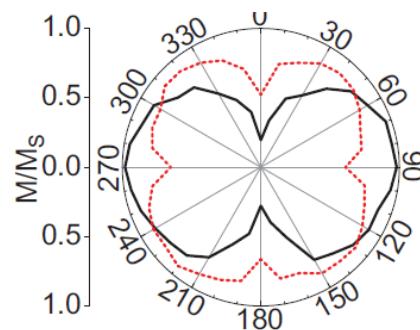
Fe



Optical microscopy experiments: image of Fe and FM domains



On a domains: uniaxial anisotropy
c domains: fourfold anisotropy
(magnetocrystalline anisotropy)



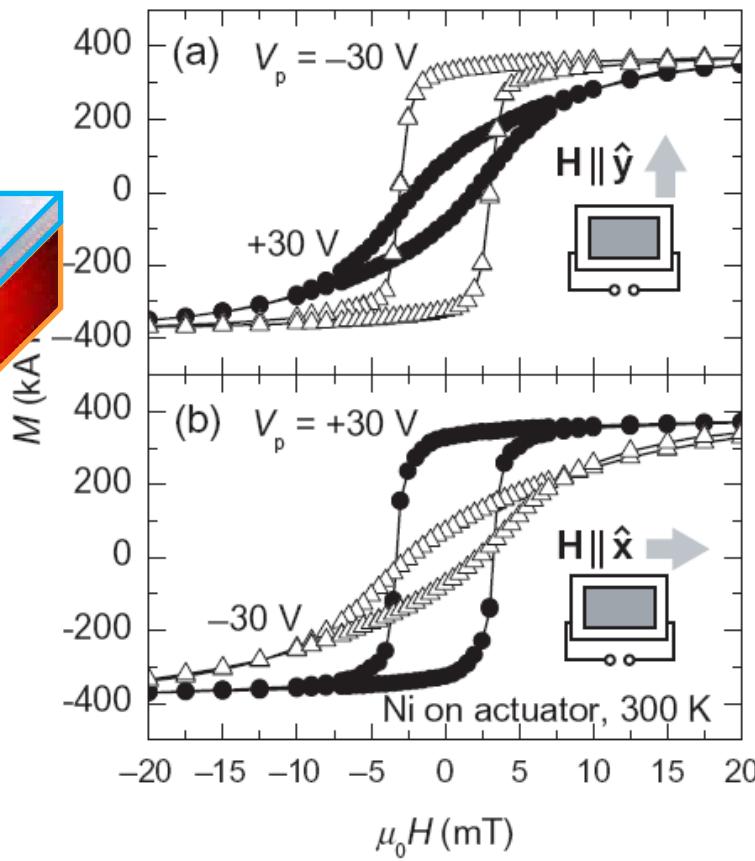
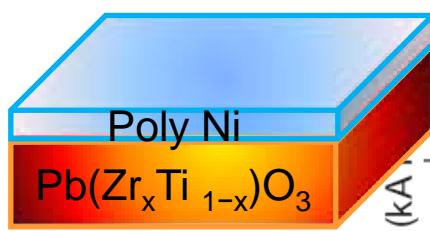
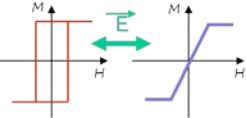
Transfer of FE domain pattern onto the FM

Lahtinen et al.; APL 101, 262405 (2012)

Electric field control of magnetic anisotropy via strain

Example : $Pb(Zr_xTi_{1-x})O_3$ actuator with Ni polycrystalline film

Magnetic anisotropy



$V_p < 0$: y = easy axis; x = hard axis
 $V_p > 0$: y = hard axis; x = easy axis
⇒ Clear Rotation by $\approx 90^\circ$ while changing the voltage polarity

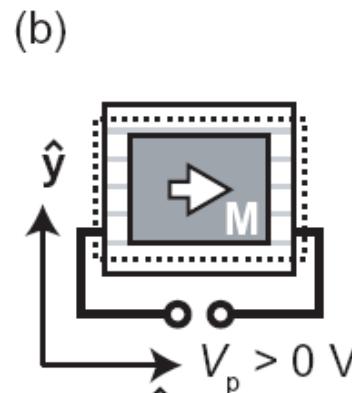
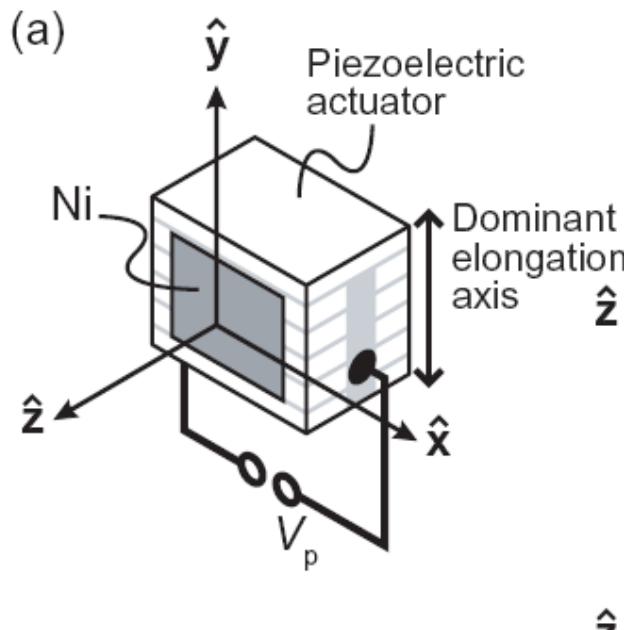
Electric field control of magnetic anisotropy via strain

Example : $Pb(Zr_xTi_{1-x})O_3$ actuator with Ni polycrystalline film

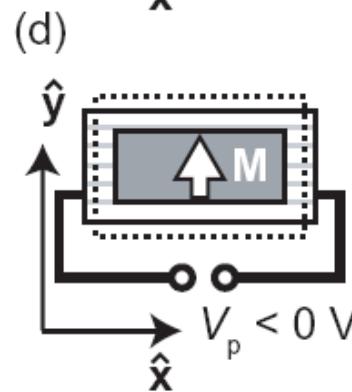
How it works?

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 **magnetostriiction** in Ni, strain modifies the magnetic properties



$V_p > 0$ the y expands (x, and z contract), the Ni film is then strained tensilely along y and compressed along x.
 $M(H)$ loops \Rightarrow y=hard axis
x=easy axis



$V_p > 0$: the y axis contracts (x and z expand), the Ni film is then strained compressively along y and tensilely along x.
 $M(H)$ loops \Rightarrow y=easy axis
x=hard axis

Electric field control of magnetic anisotropy via strain

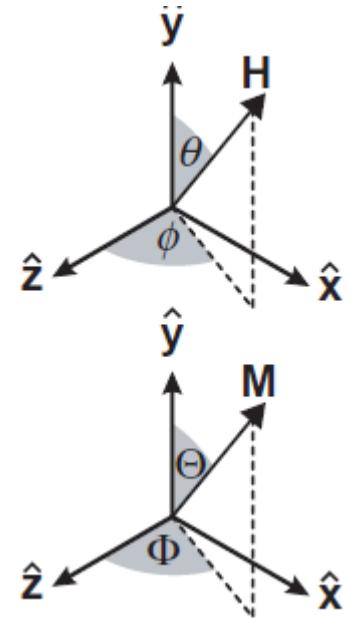
$$F = F_{\text{Zeeman}} + F_{\text{magstat}} + F_{\text{magnetocryst}} + F_{\text{magel}}$$

$$F_{\text{Zeeman}} = -\mu_0 \vec{M} \cdot \vec{H} = -\mu_0 M H \cos(\Theta - \theta)$$

$$F_{\text{magel}} = \frac{3}{2} \lambda \left(c_{12}^{\text{Ni}} - c_{11}^{\text{Ni}} \right) (1 + \nu) \varepsilon_2 \cos^2(\Theta)$$

(λ : magnetostriction) c_{ij} : elastic coefficients of Ni
 ε_2 : Strain along y

$\left(c_{12}^{\text{Ni}} - c_{11}^{\text{Ni}} \right)$ and λ are both negative in Ni



Considering a linear dependence of the length L of the actuator with voltage:

$$\varepsilon_2 = \frac{\Delta L_{\max}}{L_0} \frac{V_p}{V_{\max}}$$

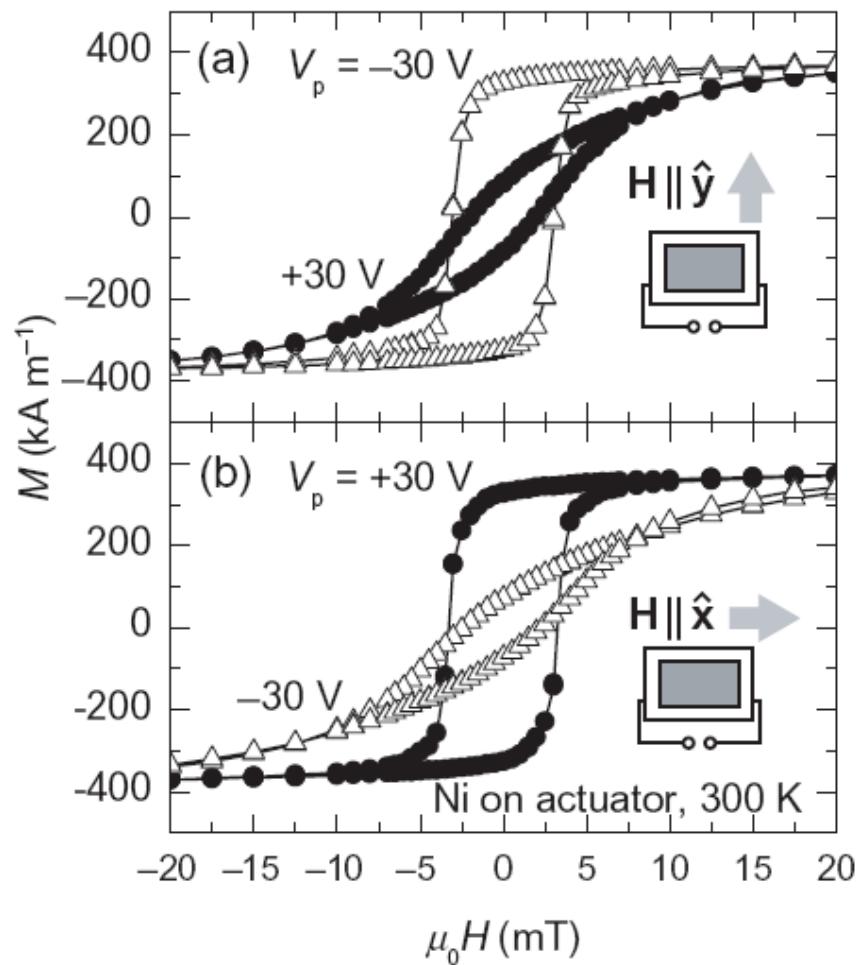
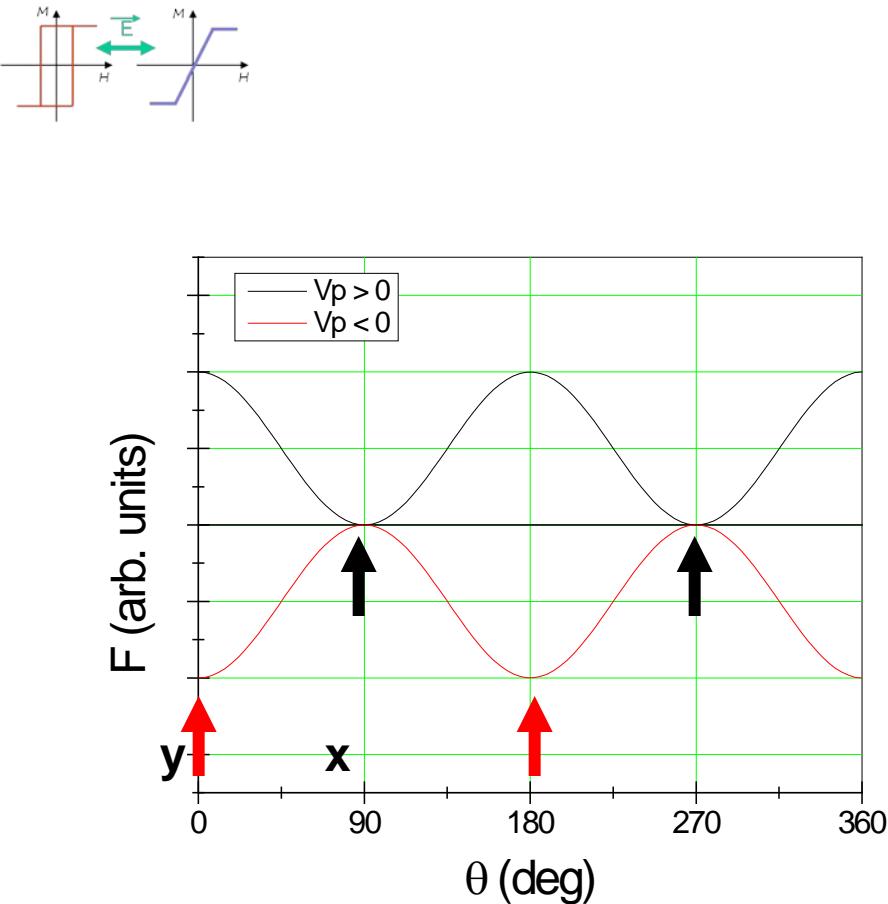
Piezoelectric properties of the PZT actuator ;
 $\Delta L_{\max}/L_0 = 1.3 \times 10^{-3}$; $V_{\max} = 180$ V

$$F_{\text{magel}} \propto \frac{V_p}{V_{\max}} \cos^2(\theta)$$

Easy axes of magnetization determined by the energy minima of F vs θ

Electric field control of magnetic anisotropy via strain

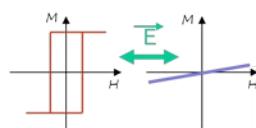
Magnetic anisotropy



Electric-field induced control of magnetization easy axis

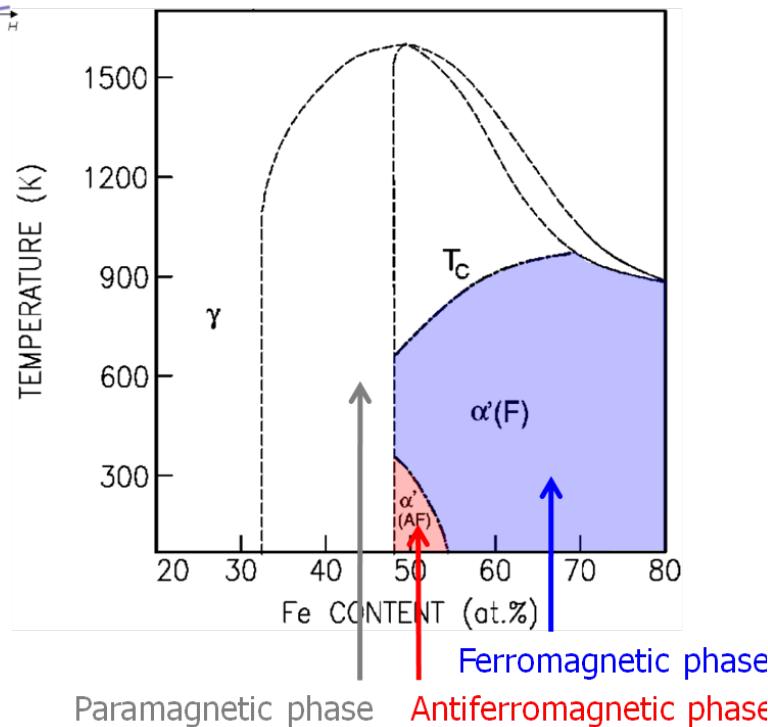
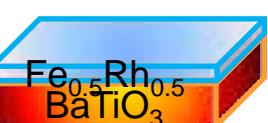
Electric field control of magnetic transition via strain

Magnetic order



Example : $\text{BaTiO}_3//\text{FeRh}$

FeRh

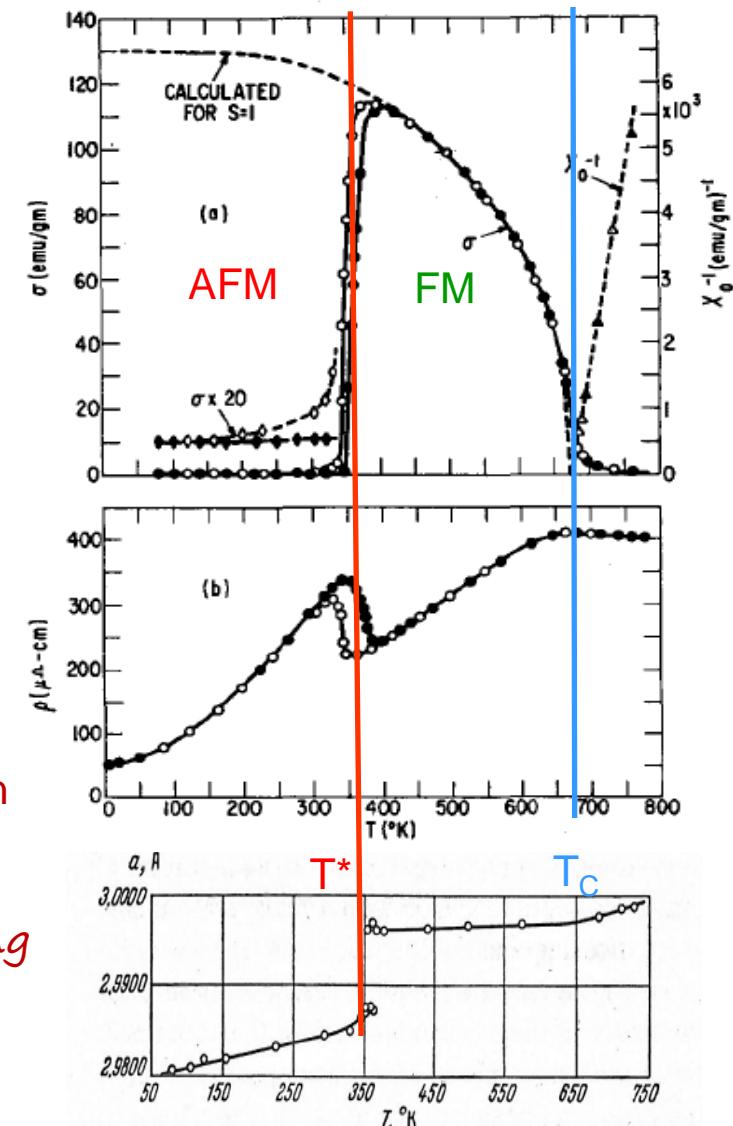


◎ γ phase : fcc

◎ α' phase : Fe/Rh ordered bcc: 1st order transition from G-AFM to FM @ 370°K

◎ Associated with large resistivity drop

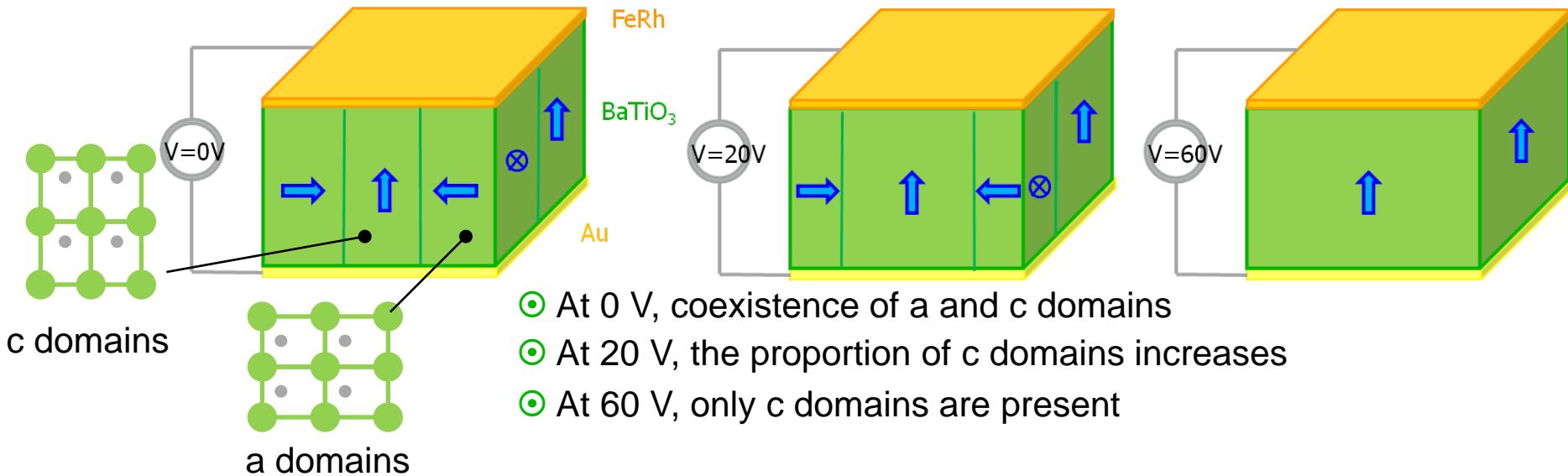
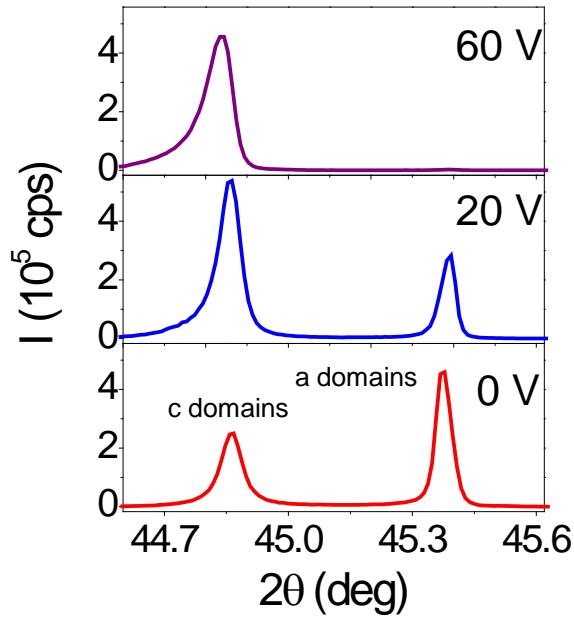
◎ Jump of cell volume by ~1% at T^* : coupling between structural and magnetic orders



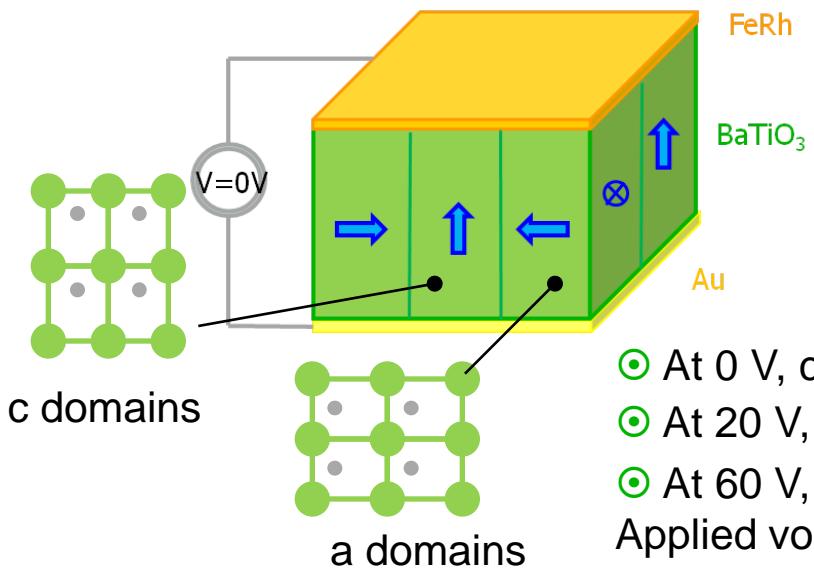
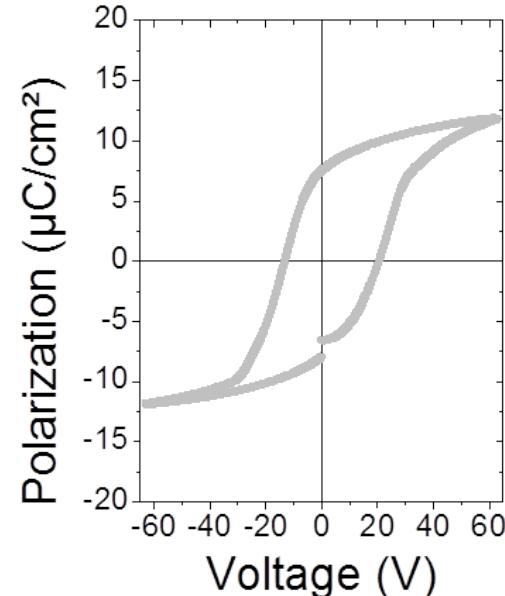
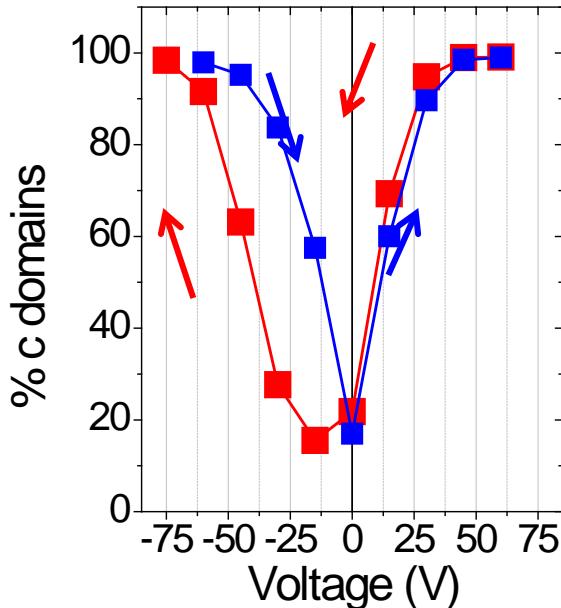
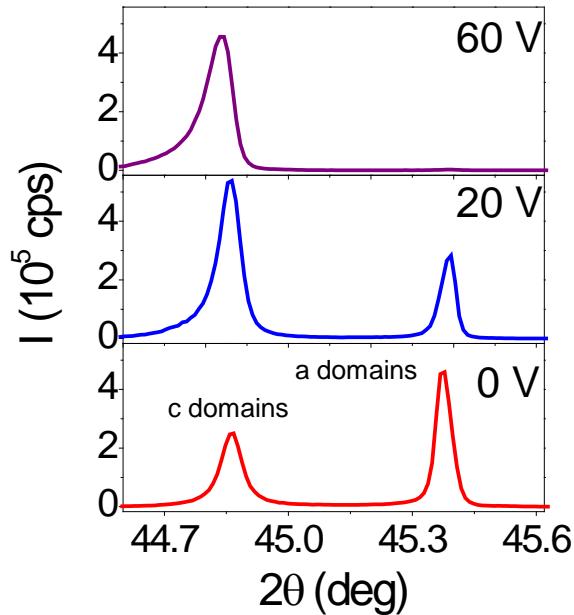
van Driel et al, JAP 85, 1026 (1999)

Kouvel et al, JAP 33, 1343 (1962)
Maat et al, PRB 72, 214432 (2005)

BaTiO_3 under electric field (X-ray diffraction study)

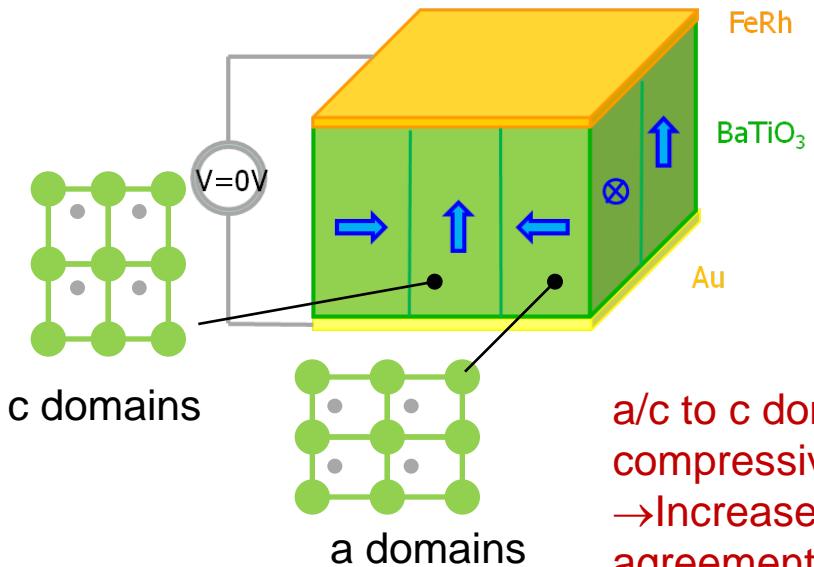
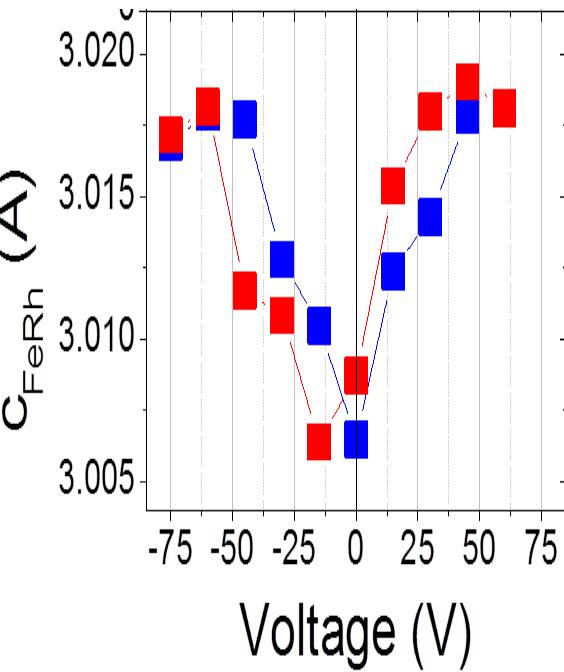
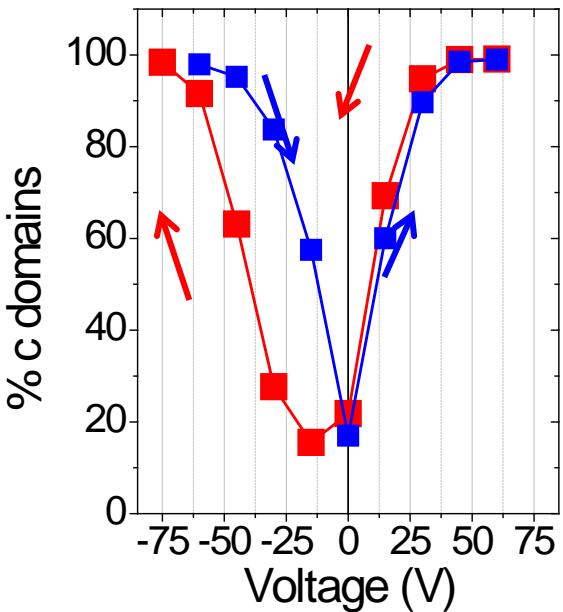
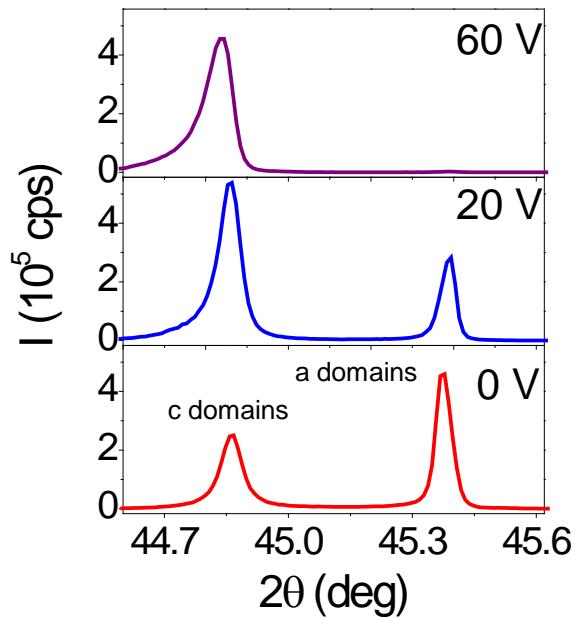


BaTiO_3 under electric field (X-ray diffraction study)



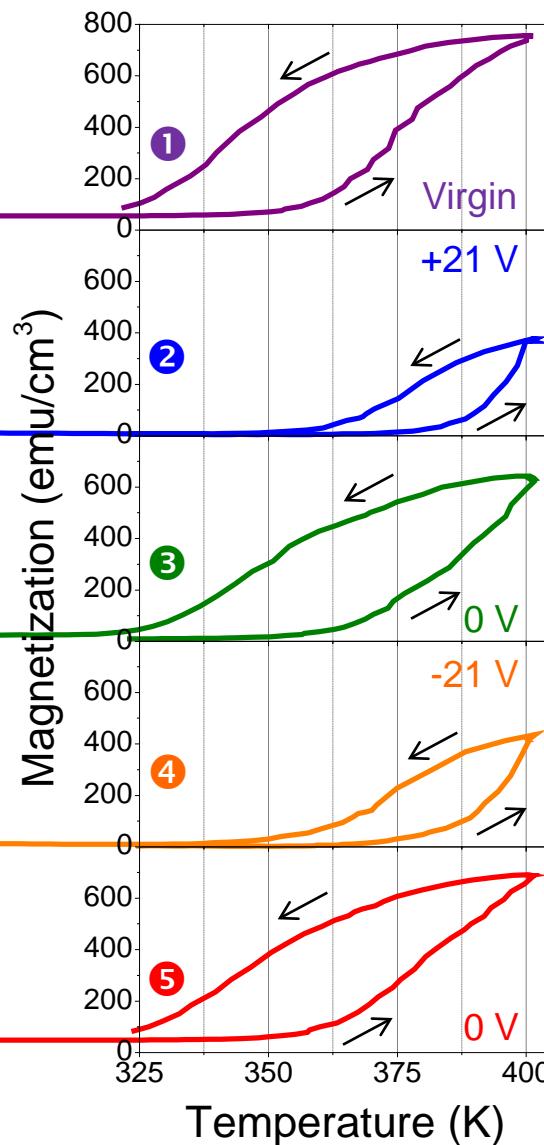
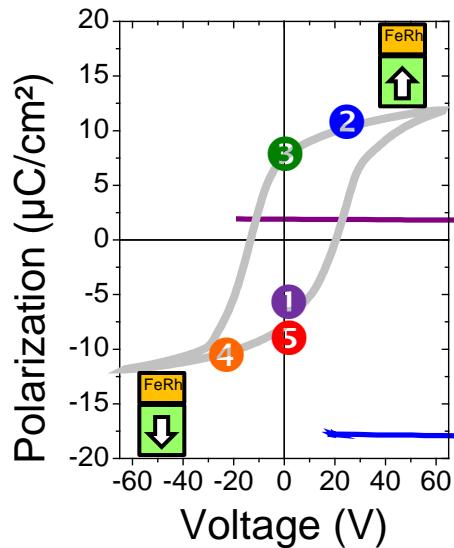
- At 0 V, coexistence of a and c domains
 - At 20 V, the proportion of c domains increases
 - At 60 V, only c domains are present
- Applied voltage increases the proportion of c domains

BaTiO_3 under electric field (X-ray diffraction study)



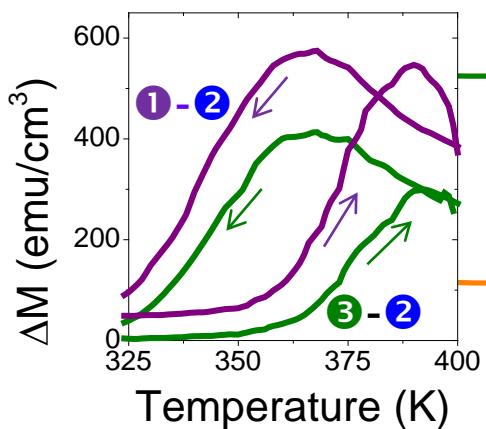
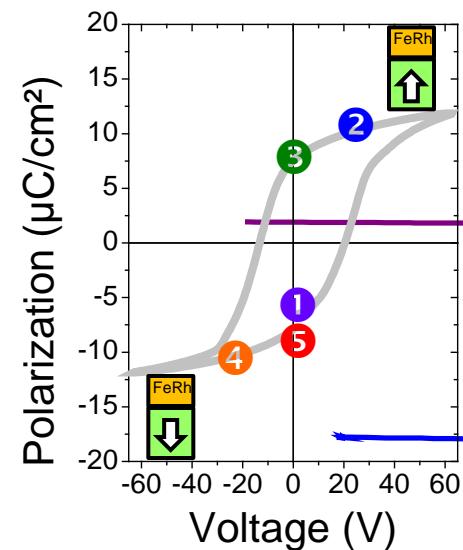
a/c to c domain configuration: increase of the in plane compressive strain by 0.47%
 → Increase in the FeRh out of plane parameter by 0.52%:in good agreement with strain (Poisson ratio:=0.31)

Influence of voltage on magnetic properties

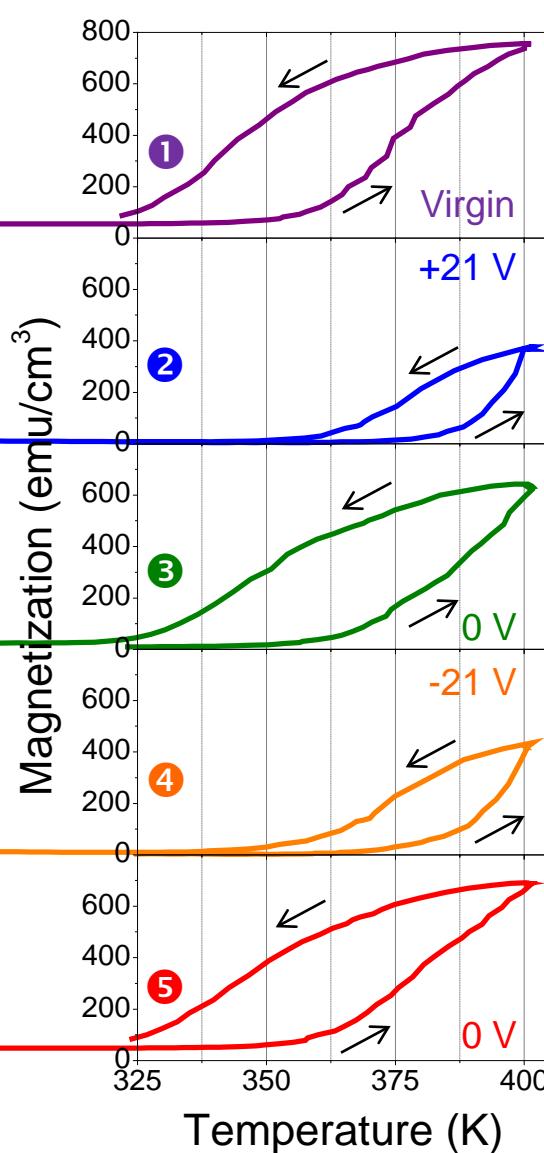


- At 0V at 20 kOe, $T^* \approx 360$ K
- Voltage shifts T^* by ~ 20 K
- Effect is reversible
- Positive or negative voltages give roughly similar effect

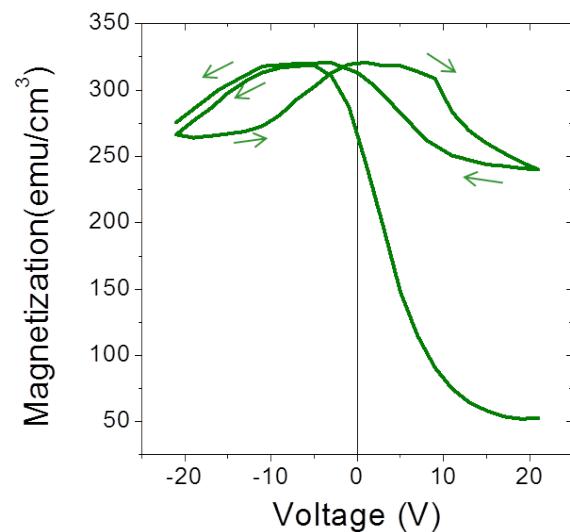
Influence of voltage on magnetic properties



- Max magnetization change $\sim 550 \text{ emu}/\text{cm}^3$
- Very large magnetoelectric coupling: $\alpha = 1.6 \cdot 10^{-5} \text{ s/m}$



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



Symmetrical effect:
reflects the strain

Electric field control of magnetism via strain:

Advantages :



- ✓ effect is bulk-related, i.e. it applies to the whole ferromagnetic film
- ✓ it can be applied to all ferromagnetic materials with magnetostriction and not too large intrinsic magnetocrystalline anisotropy

Inconvenients :

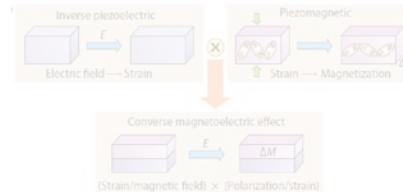


- ✓ No modification at remanence
- ✓ limited to piezoelectric with large coefficients
- ✓ needs to be demonstrated with low voltages
- ✓ fatigue ?

Mechanisms of control of magnetism by ferroelectricity:

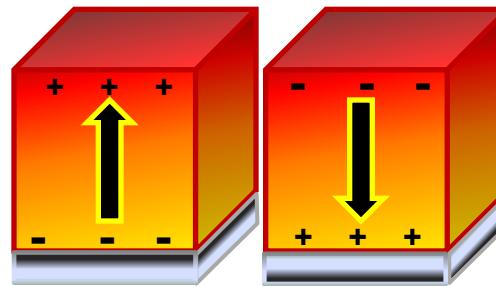
In artificial multiferroics FE/FM architectures through:

- ✓ strain-mediated coupling

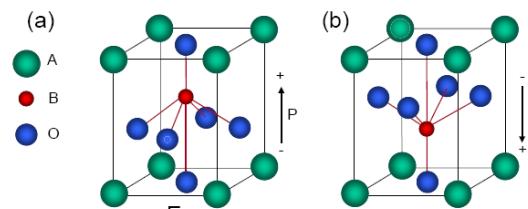
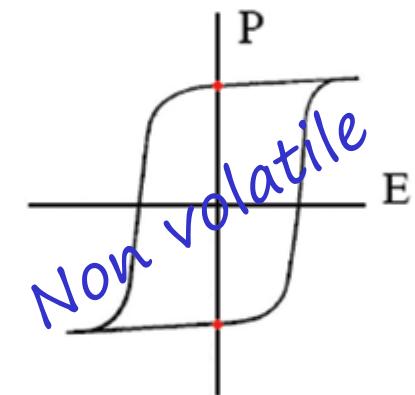


Wang et al.; NPG Asia Mater. 2, 61 (2010)

- ✓ effect of polarization direction on electronic structure of FM:
→ Field effect: accumulation/depletion



- Different hybridization



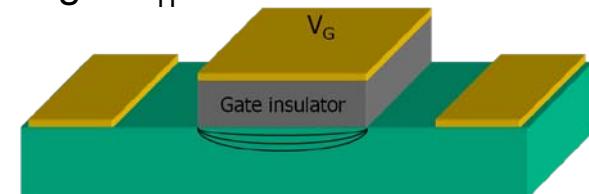
- ✓ direct coupling using an intrinsic multiferroic

effect of polarization direction on electronic structure and magnetism of FM:

Principle : like in standard FET the gate voltage locally decreases / increases the carrier density. effect efficient over the Thomas-Fermi screening length λ_{TF}

λ_{TF} proportional to $\sqrt{DOS E_F}$

$$n = n_0 e^{-\frac{t}{\lambda_{TF}}}$$

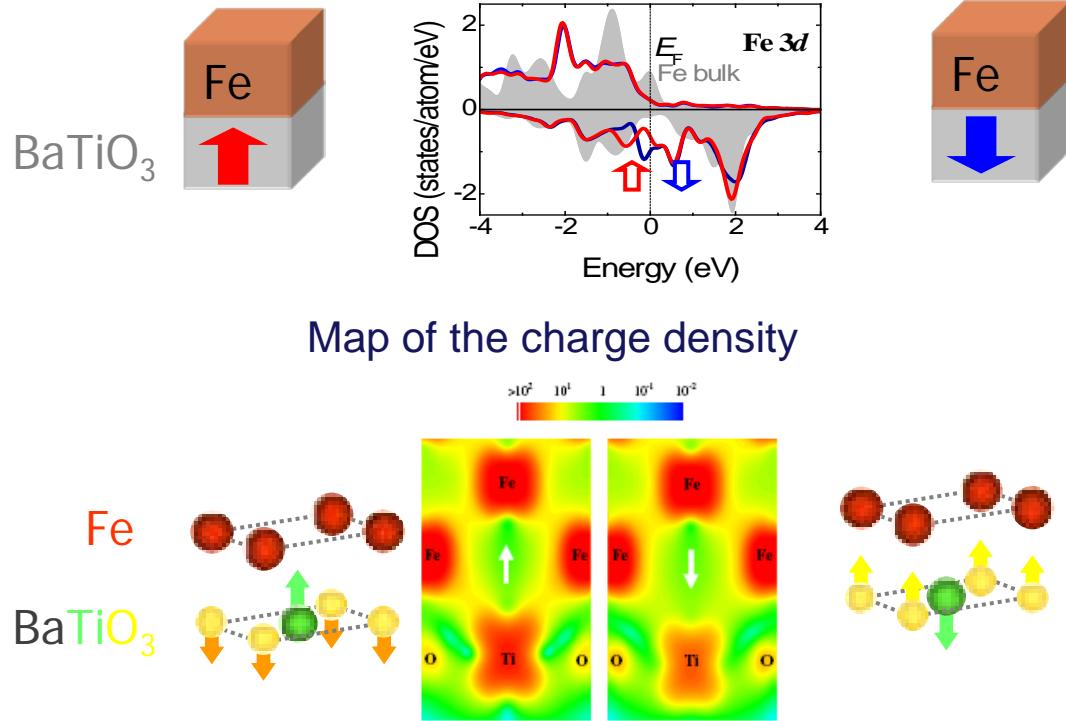


- Very thin channel ($\lambda_{TF}=0.1$ nm in metals, 1 nm in SC).
- particularly efficient in ferromagnets with a carrier-mediated magnetic interaction like **mixed-valence manganites** like $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$, **diluted magnetic semiconductors** like Mn-doped GaAs

Additional effects in FM:

- Anisotropy is determined by electron occupation of orbitals : by affecting orbital occupation at the interface, should change the **interface anisotropy**
- $n\uparrow \neq n\downarrow$: the screening is different for the two spin direction and will affect differently the DOS for spin up and spin down: results in modification of the **magnetization** (*Zhang; Phys. Rev. Lett. 83, 640 (1999)*)
- **change in the orbital overlap** between the FM and FE materials: change in DOS of spin \uparrow and spin \downarrow (*Duan et al., PRL 97, 047201 (2006)*)

Change in hybridization at the interface



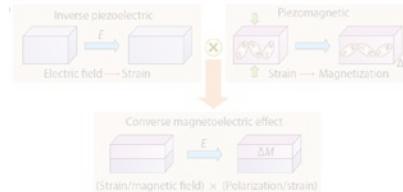
Duan et al., PRL 97,047201 (2006)

Change direction of P: change in orbital overlap between Fe and Ti:
change in the charge transfer between Fe and Ti:
change in the DOS of Fe at interface.
DOS for spin $\uparrow \neq$ spin \downarrow : affect differently the two DOS:
change in spin polarization

Mechanisms of control of magnetism by ferroelectricity:

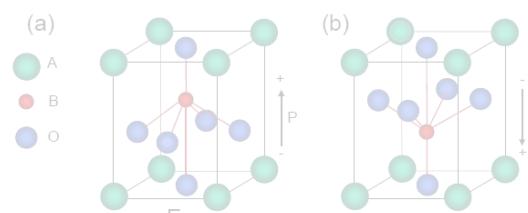
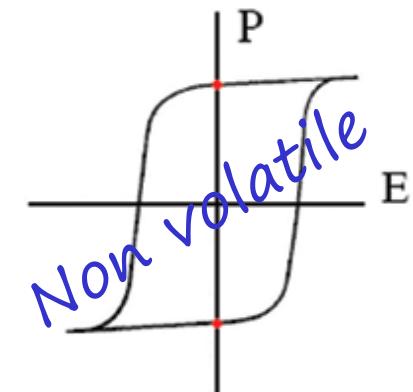
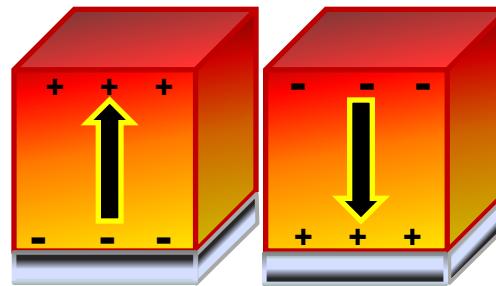
In artificial multiferroics FE/FM architectures through:

- ✓ strain-mediated coupling



Wang et al.; NPG Asia Mater. 2, 61 (2010)

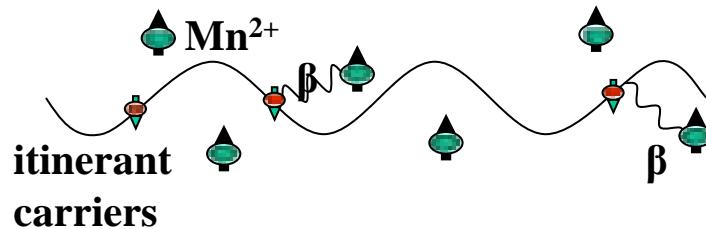
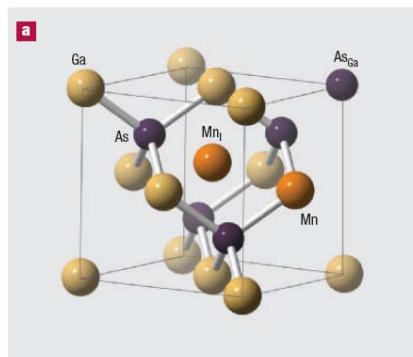
- ✓ effect of polarization direction on electronic structure of FM:
→ Field effect: accumulation/depletion



- ✓ direct coupling using an intrinsic multiferroic

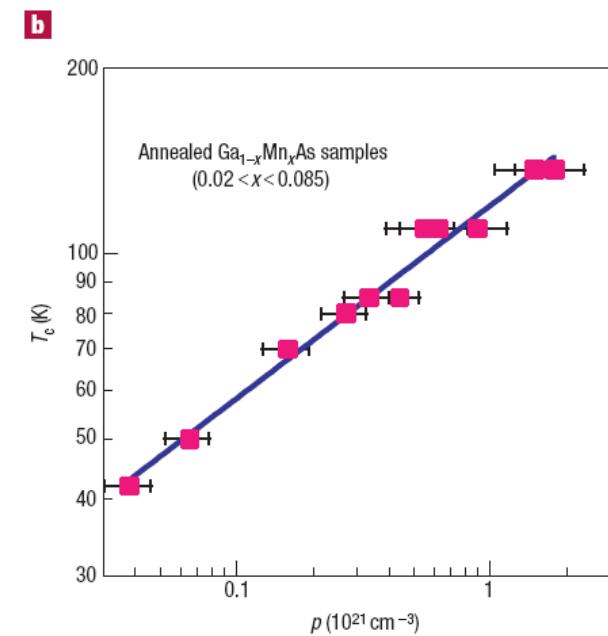
Gating experiments on carrier mediated ferromagnets:

Diluted magnetic semiconductors: Mn-doped GaAs

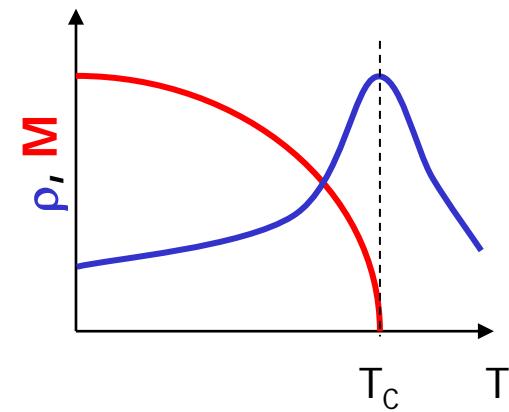


The Curie temperature is strongly dependent on the carrier density

→ Changing the carrier density by an electric field should modify T_c

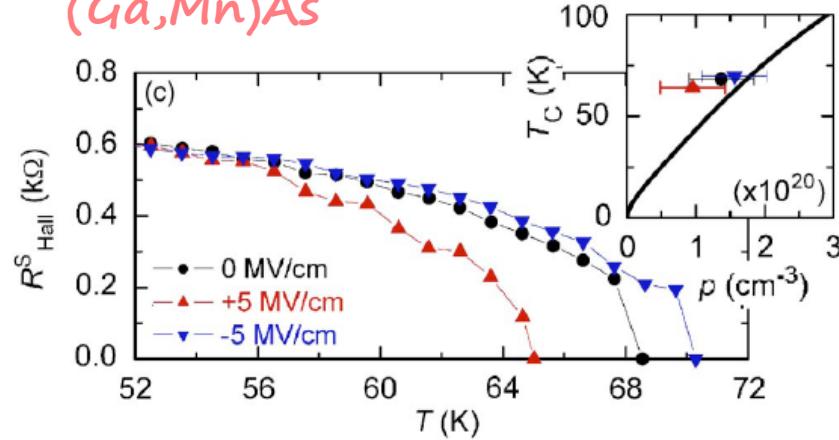


McDonald et al, Nature Mater. 2005

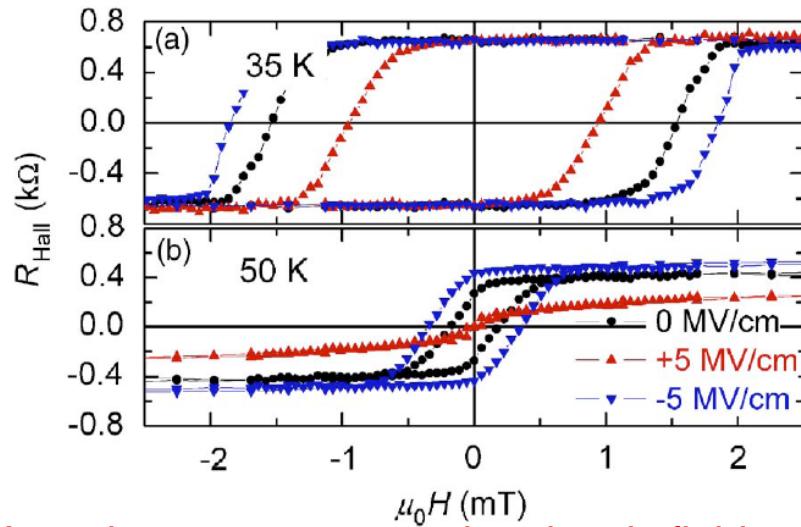


effect of gate voltage on magnetism

(Ga,Mn)As



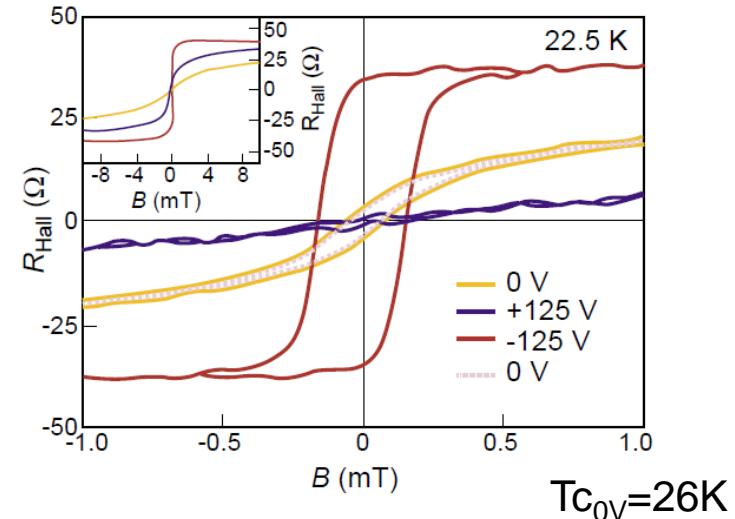
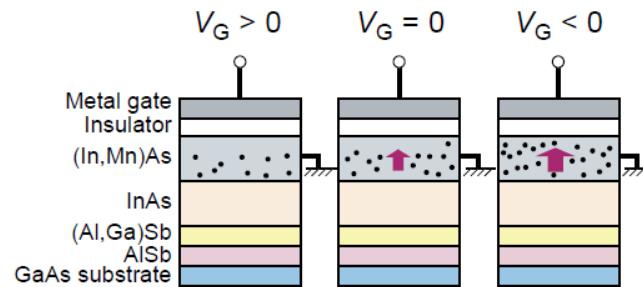
✗ The application of a positive or negative gate voltage (electric field) changes the T_c



✗ At a given temperature, the electric field can be used to change the magnetic properties (anisotropy) or even suppress ferromagnetism

Chiba et al, APL 2006 & Nature 455, 515 (2008)

(In,Mn)As



$T_c|_{0V} = 26\text{K}$

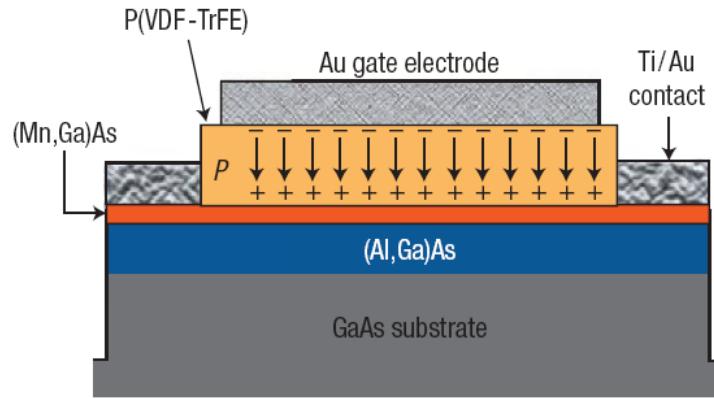
Ohno et al.; Nature 408, 944 (2000)

However, the effect is volatile

FE Gating on magnetism

(Ga,Mn)As

How to make the field effect **non-volatile**? Use a **ferroelectric gate insulator**



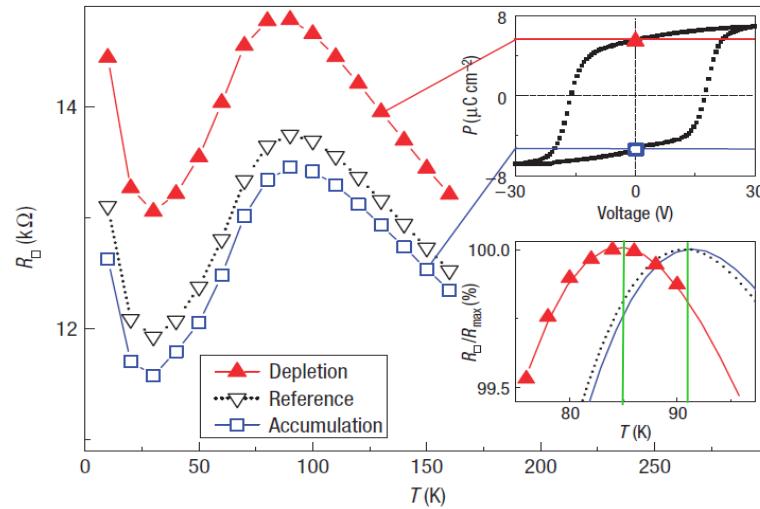
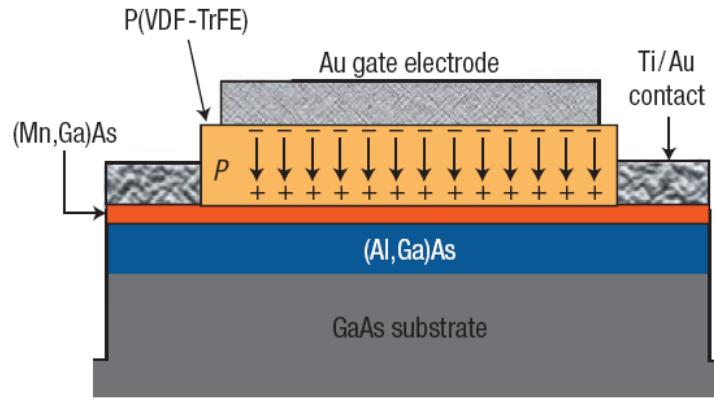
$$\Delta n_{sq} = \frac{2P}{e} = 6 \cdot 10^{14} / \text{cm}^2 \quad \text{for } P = 50 \mu\text{C}/\text{cm}^2$$

$$d = 6 \text{ nm} \Rightarrow \Delta n = \frac{P}{ed} = 10^{21} / \text{cm}^3$$

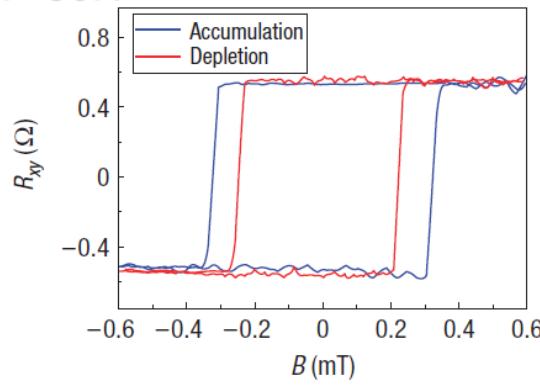
FE Gating on magnetism

(Ga,Mn)As

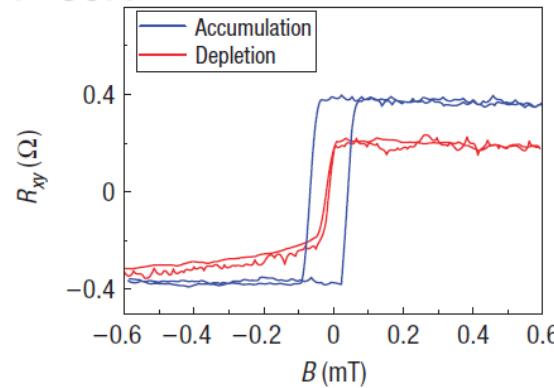
How to make the field effect **non-volatile**? Use a **ferroelectric gate insulator**



$T=50\text{K}$



$T=60\text{K}$

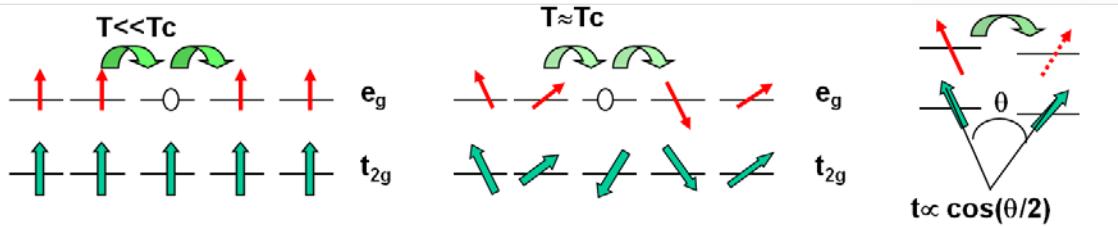
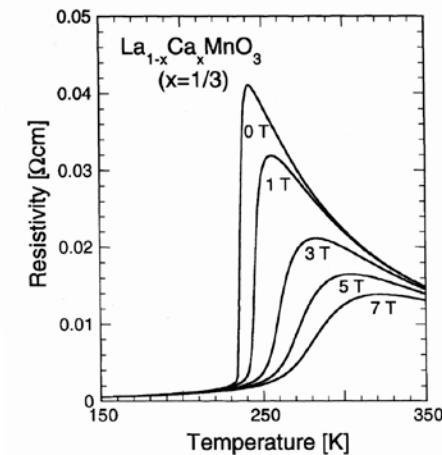
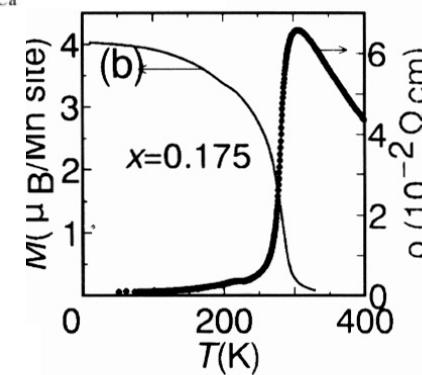
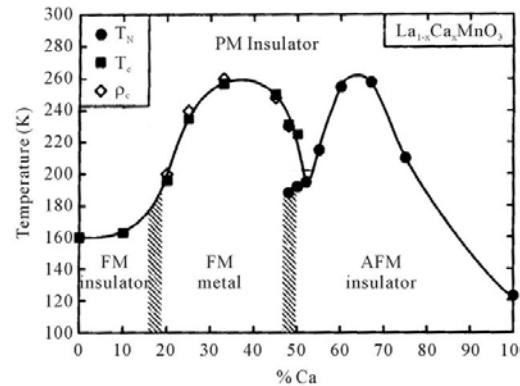
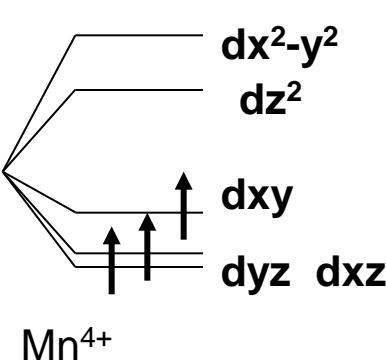
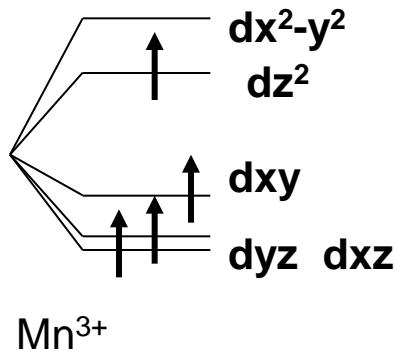
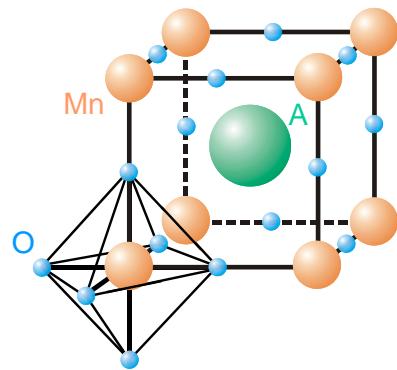


- ✗ non volatile Change in T_c : $\Delta T_c/T_c = 5\text{K} / 85\text{K}$
- ✗ Change in magnetic anisotropy

Stolichnov et al, Nature Mater. 7, 464 (2008)

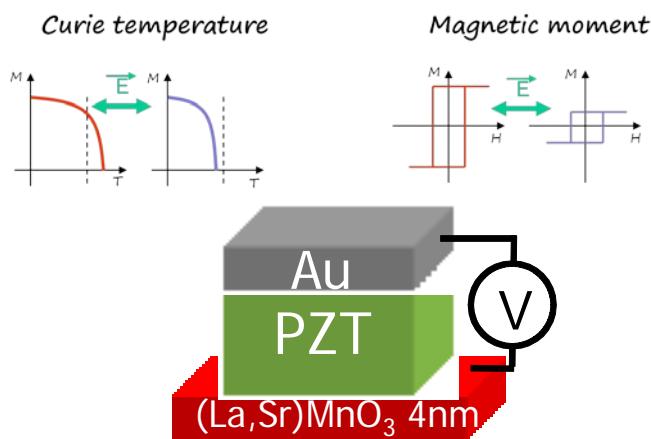
FE Gating on magnetism

Manganites

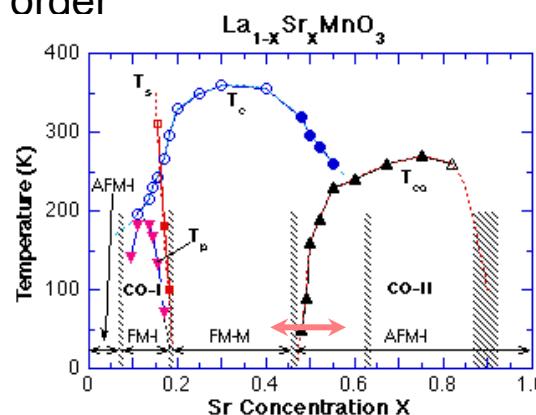
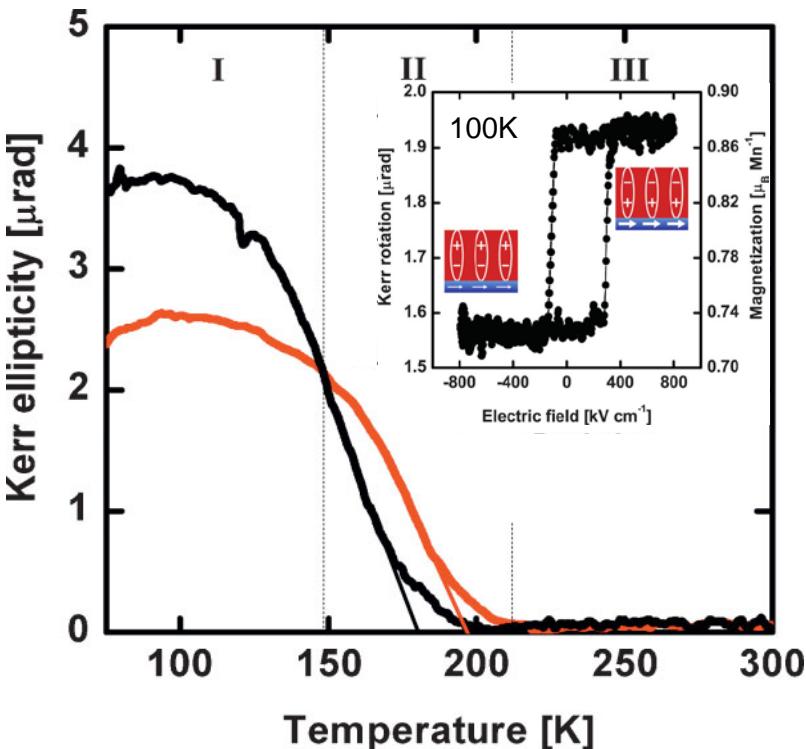


FE Gating on magnetism

Manganites



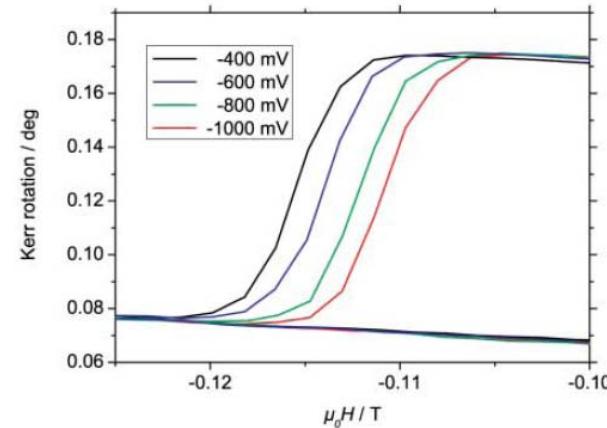
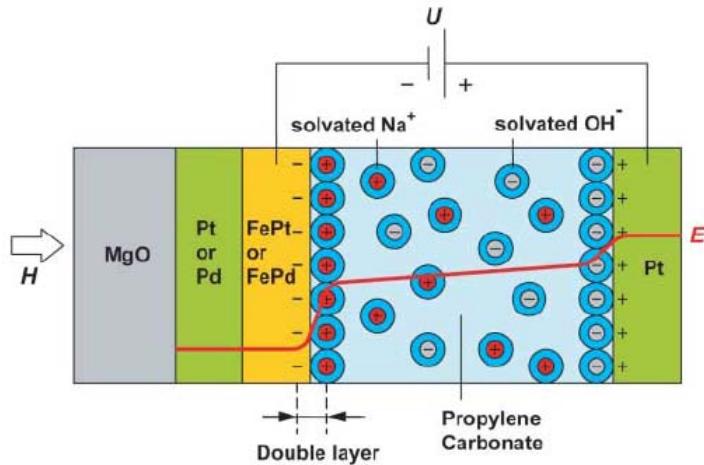
- ◎ Switching P in PZT produces charge accumulation/depletion in manganite
 - Change T_C of manganite
 - Change in magnetization amplitude
- ◎ Attribute to change in carrier concentration that induced a transition from FM (to AFM order at interface



Field effect control of magnetism at RT?

Need to use TM-FM

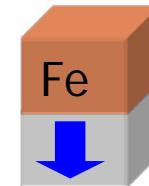
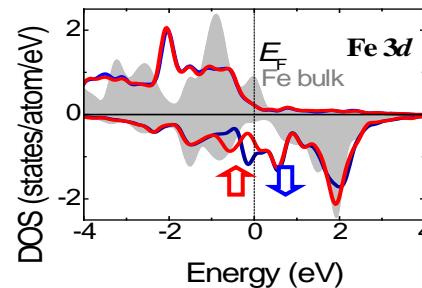
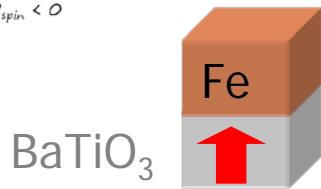
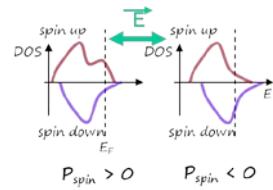
Pb: have a large carrier density $10^{23}/\text{cm}^3$ compared to DMS or manganites ($10^{21}/\text{cm}^3$)
BUT feasible:



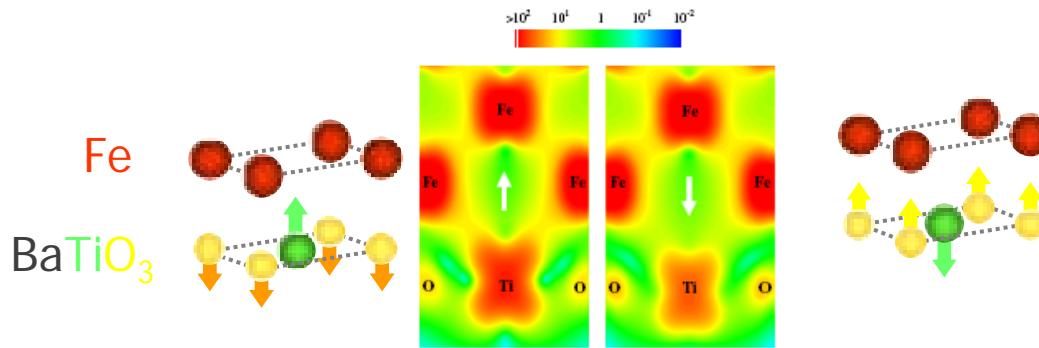
Change in magnetic coercive field at 300K induced by an electric field in an ultrathin FePt film:
Accumulation/depletion change the orbital occupancy at interface: changes the interface anisotropy
thus the coercive field

Change in hybridization at the interface

Spin polarization



Map of the charge density

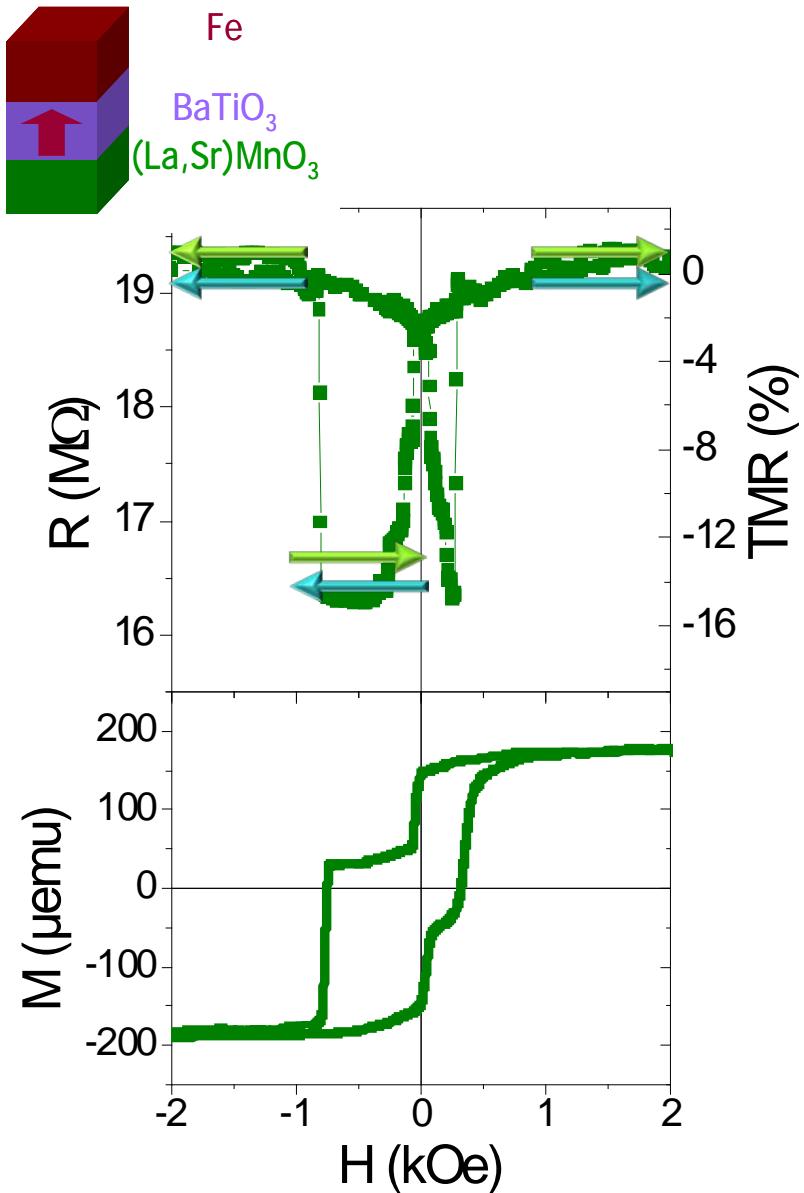


Duan et al., PRL 97,047201 (2006)

Change direction of P: change in orbital overlap between Fe and Ti: change in the charge transfer between Fe and Ti: change in the DOS of Fe at interface.
 DOS for spin $\uparrow \neq$ spin \downarrow : affect differently the two DOS: change in spin polarization

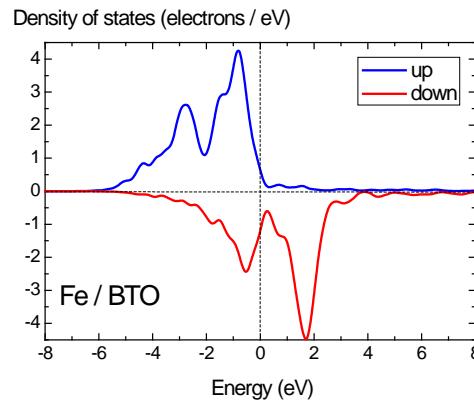
Change in hybridization at the interface

FM tunnel Junctions Fe/ BaTiO₃ 1nm/La_{0.7}Sr_{0.3}MnO₃ : TMR



$$TMR = \frac{R_{AP} - R_P}{R_P} = \frac{2SP_1SP_2}{1-SP_1SP_2}$$

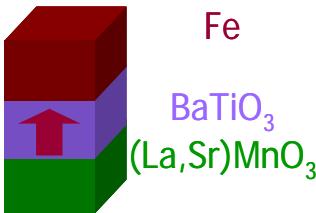
- ✓ Clear negative tunnel magnetoresistance (TMR)
- ✓ Negative spin-polarization at Fe/BTO interface



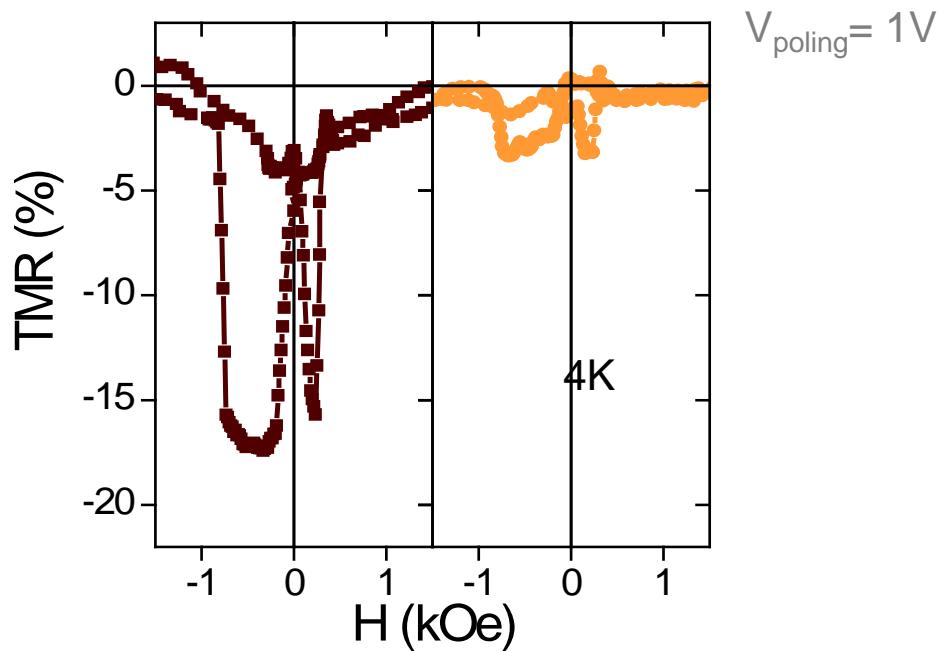
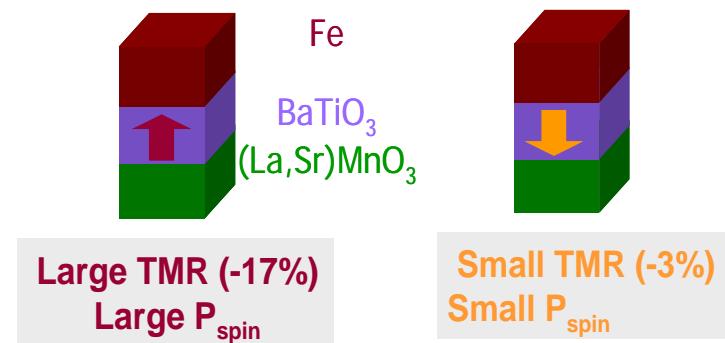
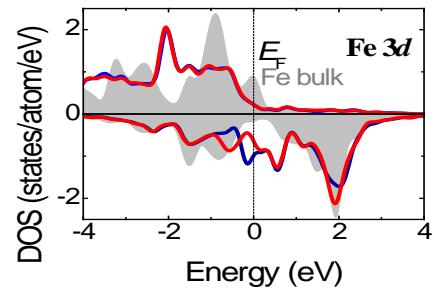
$$SP = \frac{N_{\uparrow}(E_F) - N_{\downarrow}(E_F)}{N_{\uparrow}(E_F) + N_{\downarrow}(E_F)} < 0$$

Change in hybridization: Electric control of the spin polarization

FM & FE tunnel Junctions Fe/ BaTiO₃ 1nm/La_{0.7}Sr_{0.3}MnO₃ :



→ Change in the TMR amplitude reflects change in the DOS of Fe at the interface and the consequent change in spin polarization when FE polarization direction is changed
Measured at 4K (due to LSMO) but in principle feasible at RT.

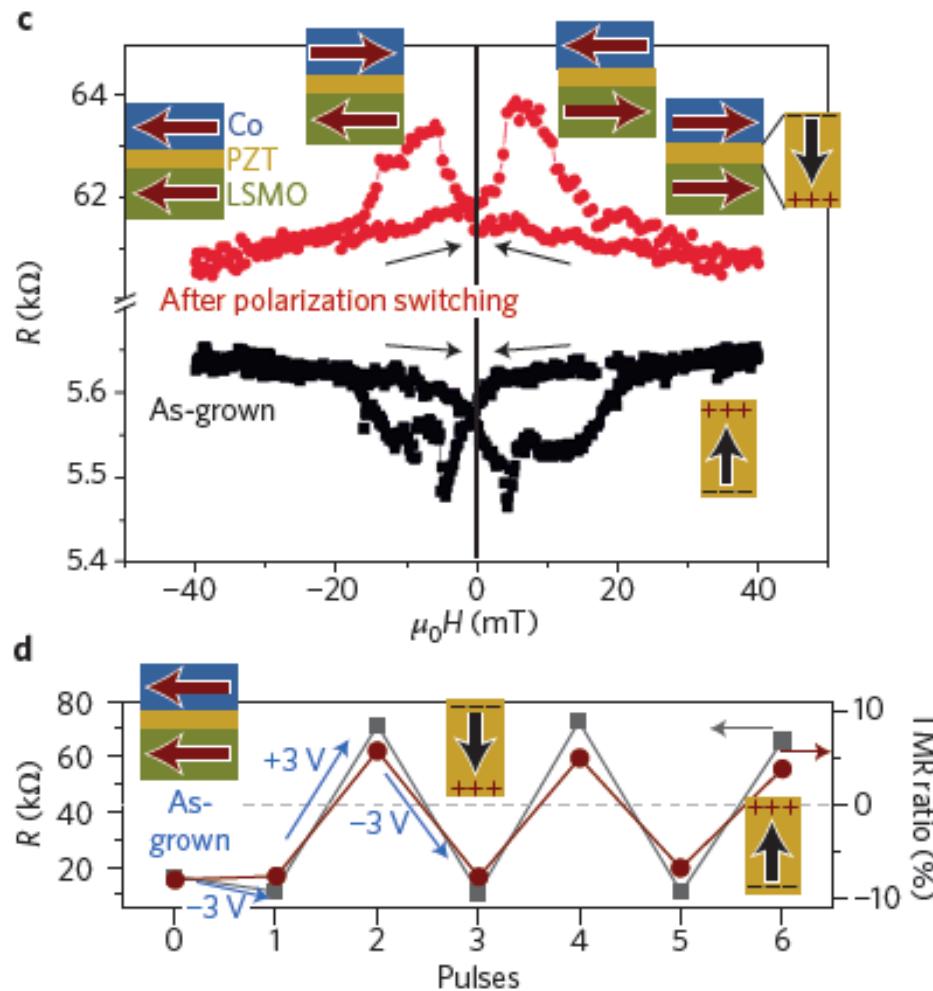


Garcia et al.; Science 327, 1106 (2010)

Change in hybridization: Electric control of the spin polarization

FM & FE tunnel Junctions Co/ PbZrTiO₃ 1nm/La_{0.7}Sr_{0.3}MnO₃:

- Change in the TMR sign
- reflects change in the DOS of Fe at the interface and the consequent change in spin polarization when FE polarization direction is changed



FE control of electronic structure

Very rich physics with large number of mechanisms

Advantages :



Substantial change in T_c may be achieved
close to T_c the magnetic properties can be tuned
Substantial change in anisotropy
Change in spin polarization: particularly attractive in MTJs

Inconvenients :

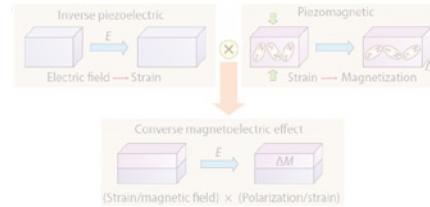


very local modification (over a thickness of a few nm at most)
effect is small
effect is mostly restricted to carrier-mediated ferromagnets

Mechanisms of control of magnetism by ferroelectricity:

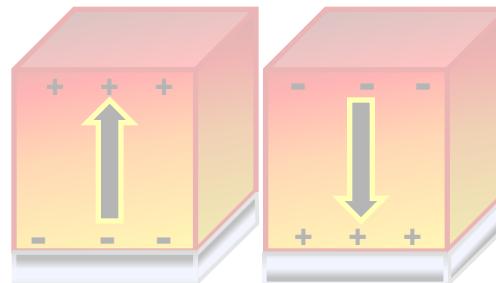
In artificial multiferroics FE/FM architectures through:

- ✓ strain-mediated coupling

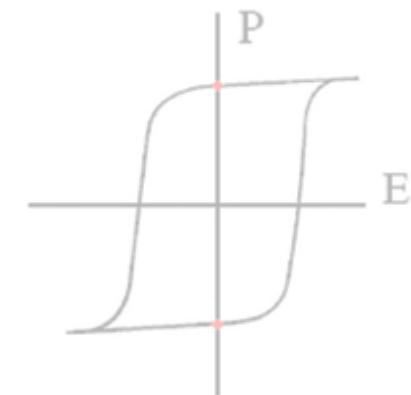
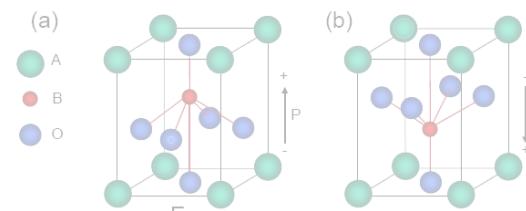


Wang et al.; NPG Asia Mater. 2, 61 (2010)

- ✓ effect of polarization direction on electronic structure of FM:
→ Field effect: accumulation/depletion



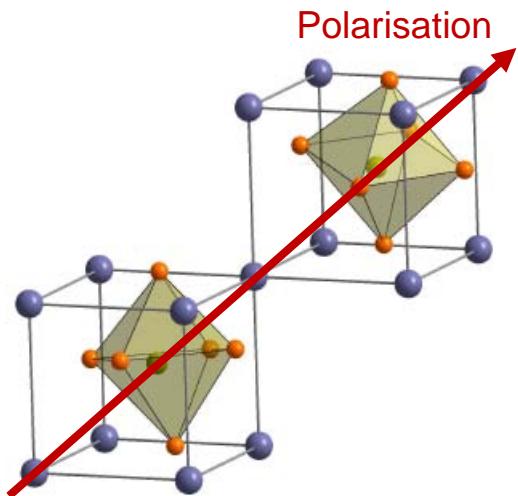
→ Different hybridization



- ✓ direct coupling using an intrinsic multiferroic

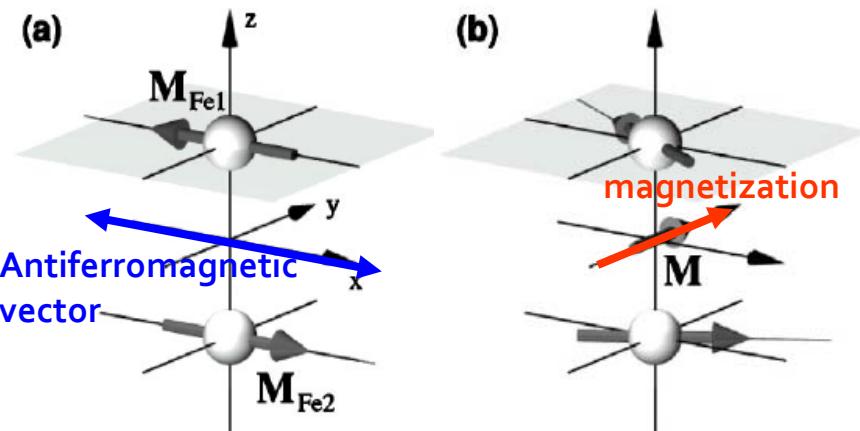
BiFeO_3 (BFO): an AFM-FE Multiferroic @ RT

Rhombohedrally distorted perovskite
(R3c) $a=3.96\text{\AA}$ $\alpha=89.5^\circ$



Polarization along [111] direction
 $T_c=1100\text{K}$
 $P_s=100\mu\text{C}/\text{cm}^2$

Lebeugle et al.; APL 91, 022907 (2007)



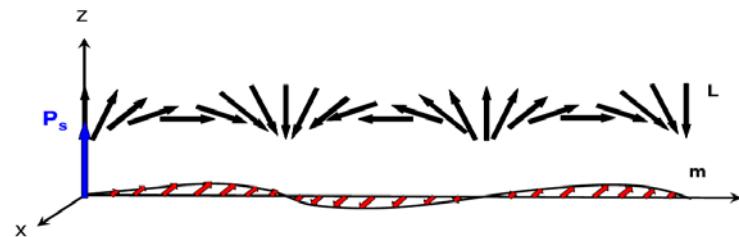
C. Ederer & N. Spaldin, PR B, 71, 060401 (R) (2005)

Antiferromagnetic of G type :

Superexchange: AF $T_N=640\text{K}$

Canted spins \rightarrow weak ferromagnet $M_S=0.01\mu_B/\text{f.u.}$

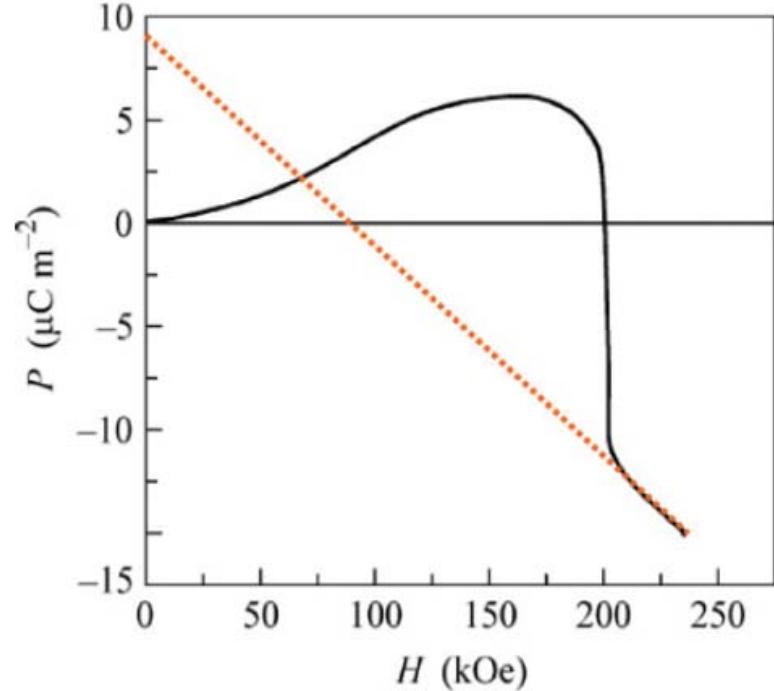
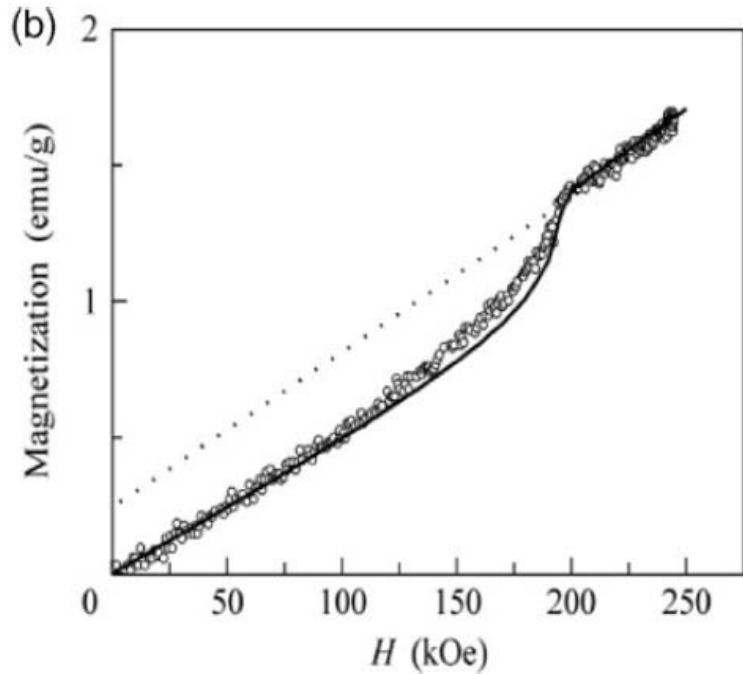
P. Fisher et al., J. Phys. C, 13, 1931 (1980)



cycloidal modulation \Rightarrow Averaging to zero of the linear ME effect

Review by G. Catalan & J. Scott; Adv. Mat. 21, 2463 (2009)

BiFeO_3 (BFO): an AFM-FE Multiferroic @ RT



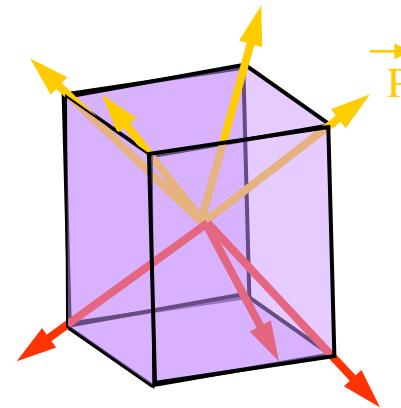
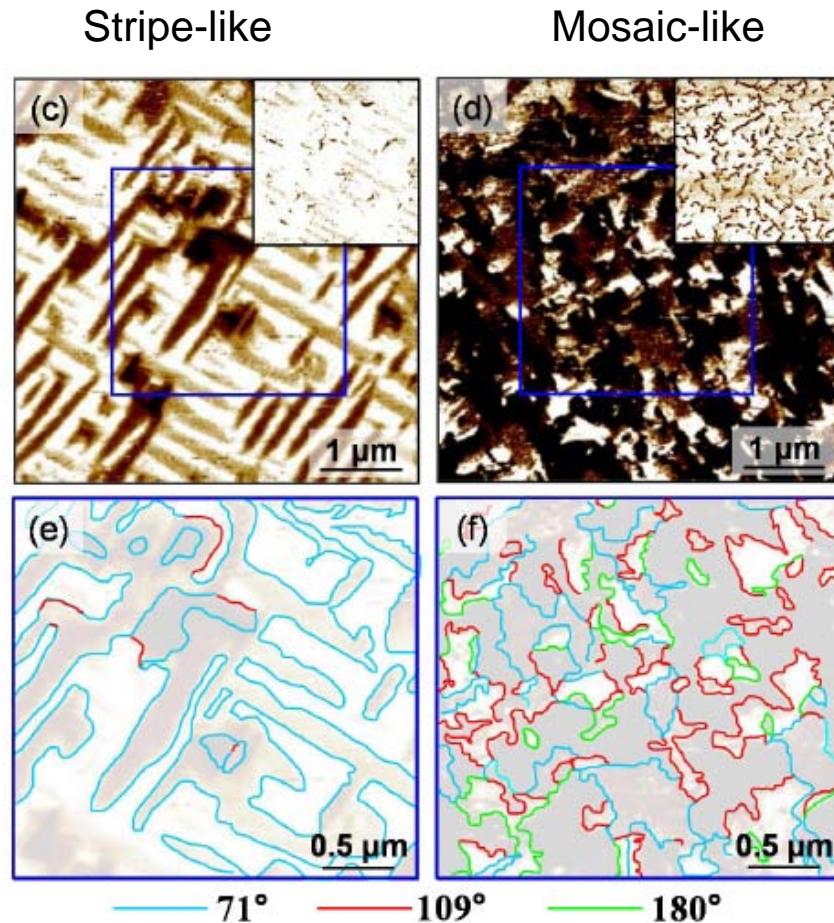
In bulk: Above 20T: the cycloidal modulation is destroyed:

- recovery of a **small magnetic moment**
- **linear magnetoelectric coupling** reflected by the linear dependence of the magnetically induced polarisation

Y. F. Popov et al; JETP Lett. 57, 69 (1993); A. K. Zvezdin et al.; Jmmm 300, 224 (2006)

In thin film the cycloidal modulation is destroyed (*Béa et al., Phil. Mag. Lett. 87, 165 (2007)*): the linear magnetoelectric coupling is allowed

FE domain structure (PFM) in BiFeO_3 thin films

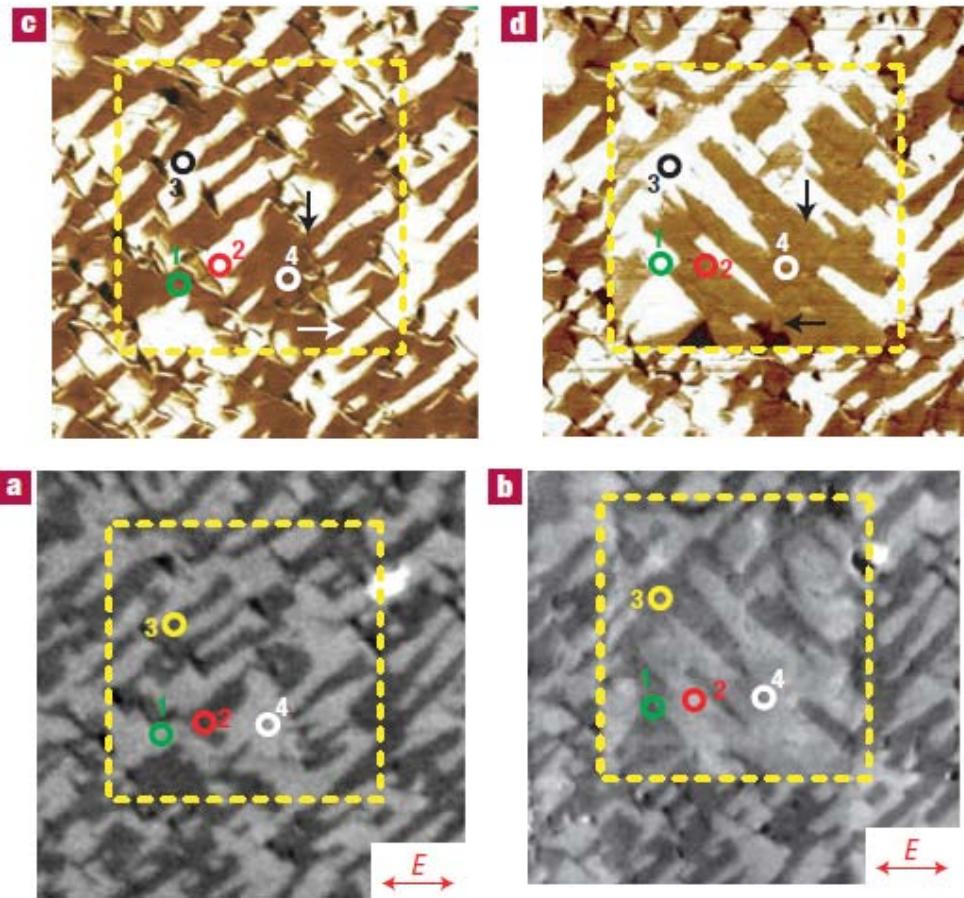


Stripe-like domains: mainly 71° DWs

Mosaic-like domains: 109° + 71° + 180° DWs

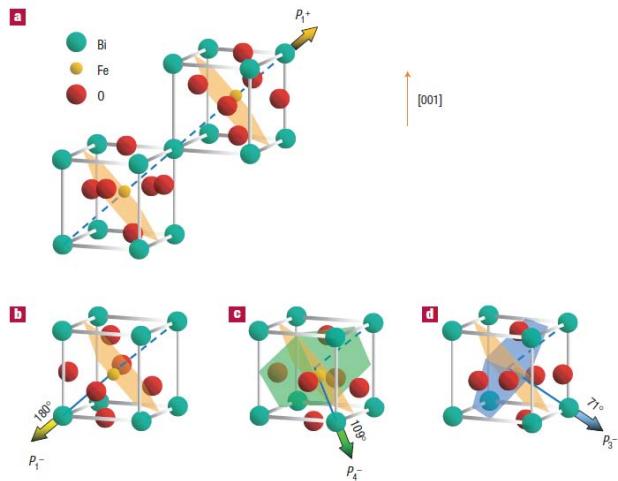
BFO: evidence for the magnetoelectric coupling

Combination of PFM and XLD-PEEM experiments



1 & 2 : 109° ferroelectric switching
3: 71° switching
4: 180° switching
1 & 2 the PEEM contrast reverses after electrical poling.

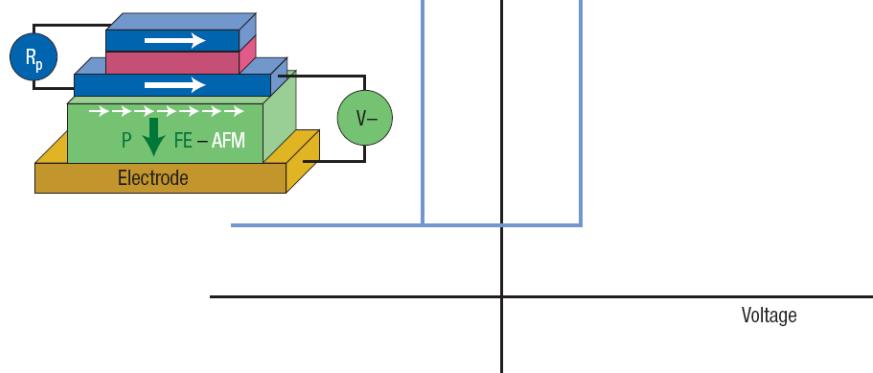
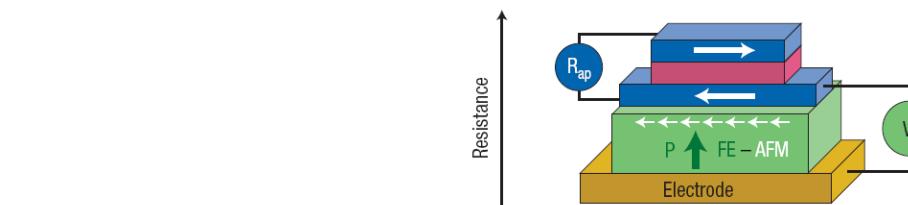
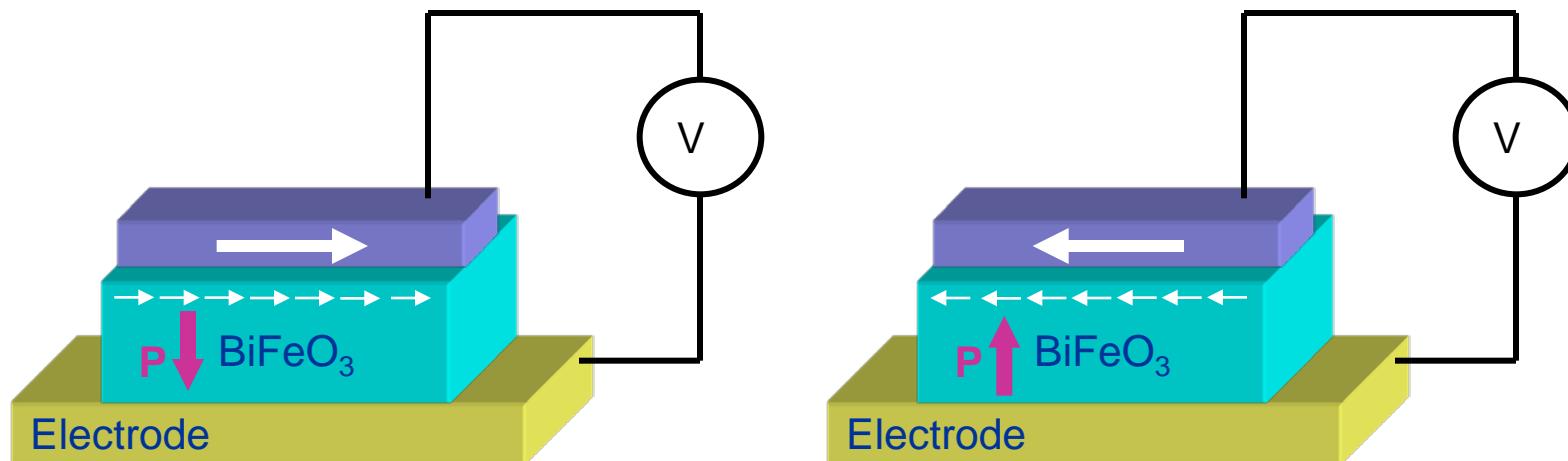
71 & 109° : change in the AFM plane
 180° : same AFM plane



T. Zhao et al.; Nature Materials 5, 823. (2006)

To exploit this magnetoelectric coupling it is necessary to couple BFO with a ferromagnetic materials through an exchange bias interaction: i.e. to design an **artificial multiferroic**

How to exploit such AFM-FE material to obtain an electric control of magnetic properties? Couple it by exchange bias with a FM.



exchange bias

Discovery in 1956

New Magnetic Anisotropy

W. H. MEIKLEJOHN AND C. P. BEAN

General Electric Research Laboratory, Schenectady, New York

(Received March 7, 1956)

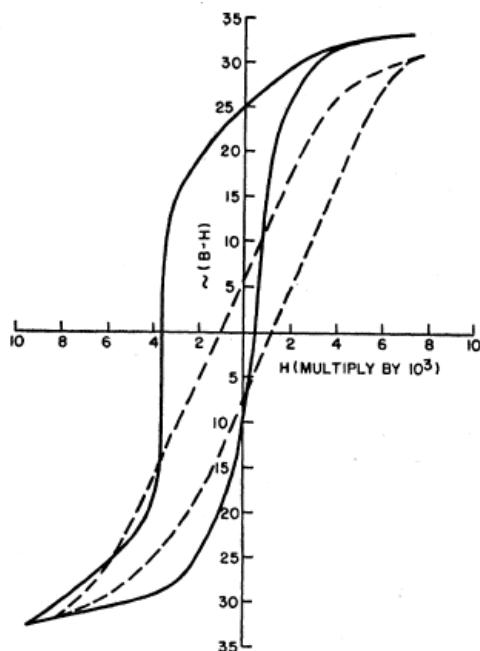
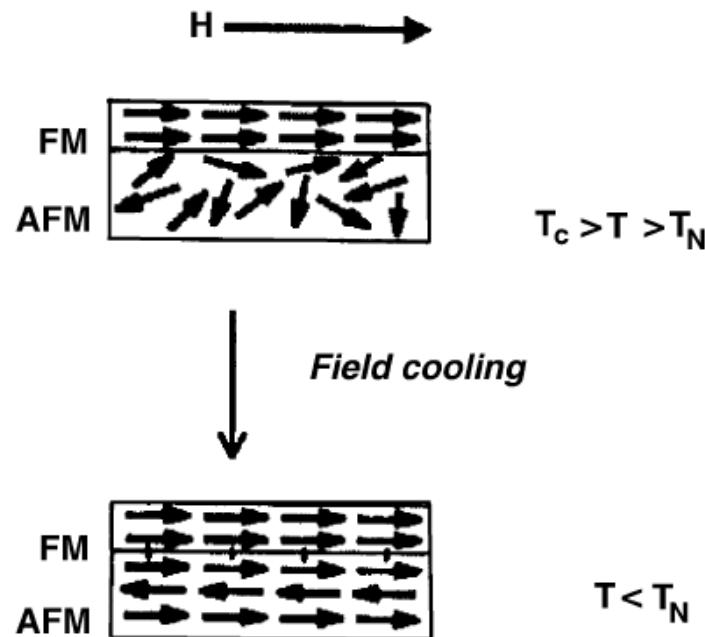


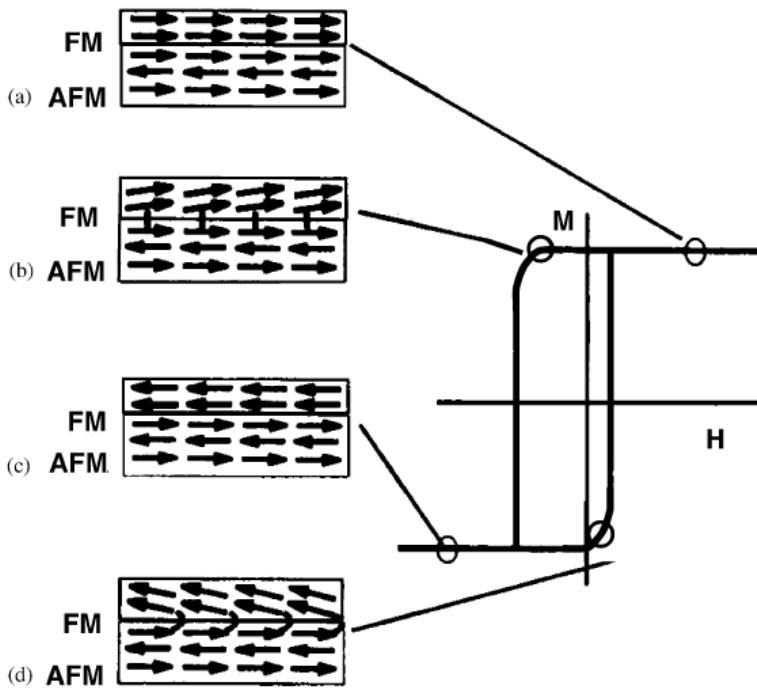
FIG. 3. Hysteresis loops of fine oxide-coated particles of cobalt taken at 77°K. The dashed lines show the hysteresis loop when the material is cooled in the absence of a magnetic field. The solid lines show the hysteresis loop when the material is cooled in a saturating magnetic field.

Meiklejohn and Beam : Co/CoO particles

Appears when a FM/AF system is cooled in a magnetic field through the Néel temperature of the AFM



Reviews : Noguès et al, JMMM 192, 203 (1999)
Noguès et al, Phys. Rep. 22, 65 (2005)

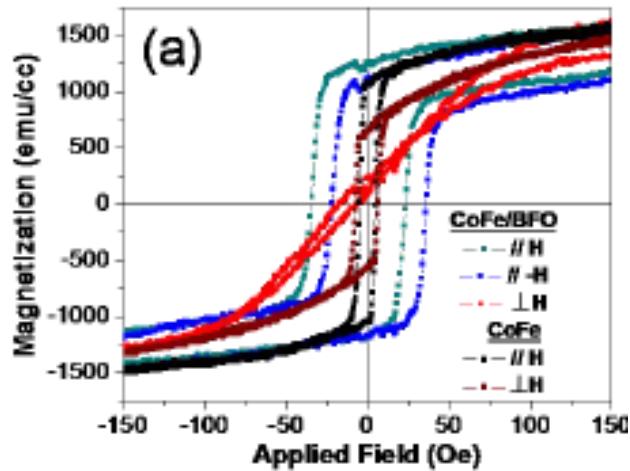


- Simple picture assumes the presence of **uncompensated spins** at the interface (due to the presence of defects)
- Some are **pinned** (by the magnetic anisotropy in the antiferromagnet) : **these are the ones responsible for the exchange bias: shift of the loop**
 - Some of the uncompensated spins are **unpinned** (free to rotate with the magnetic field, like the magnetization of the ferromagnet):**the loop is enlarged**

exchange bias coupling with BiFeO_3

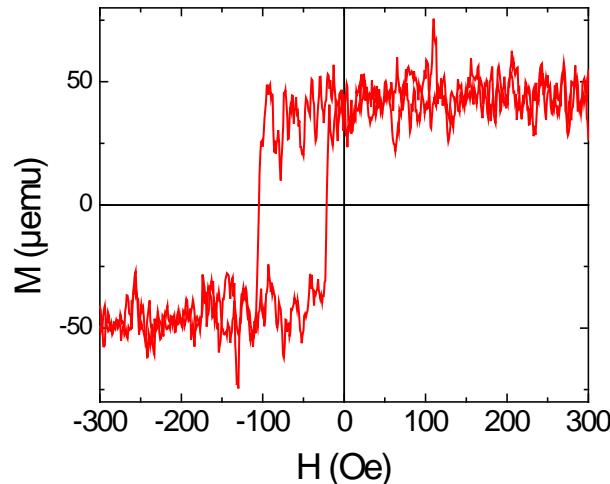
Stripe-like domains

$\text{CoFe}(2.5\text{nm})/\text{BFO}(70\text{nm})/\text//\text{STO}(001)$



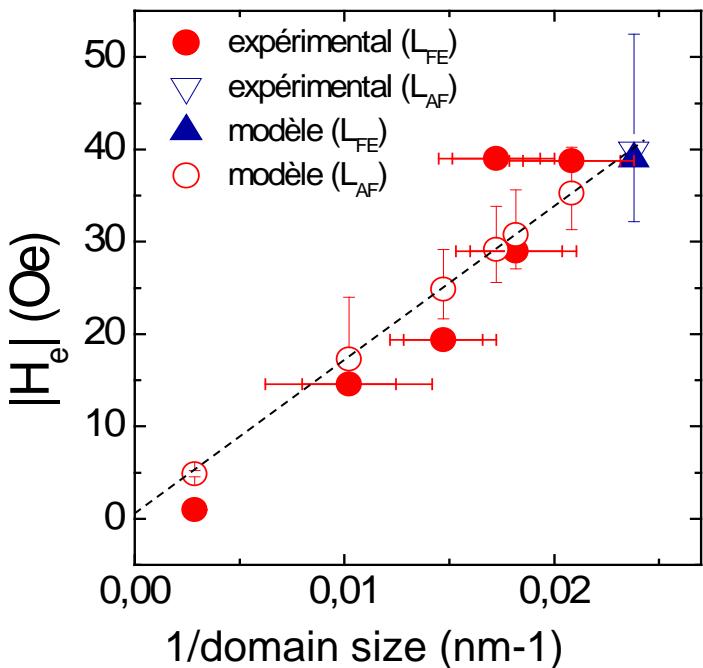
FM grown by sputtering in a field $H_{\text{dep}} = 200 \text{ Oe}$

$\text{CoFeB}(7.5\text{nm})/\text{BFO}(70\text{nm})/\text//\text{STO}(001)$



- ◎ On BFO, $M(H)$ cycle is enlarged (**coercivity enhancement**) and shifted (**exchange bias**)
- ◎ Stripe like domains: only 109° DWs: only enlarged hysteresis loop
- ◎ Mosaic like domains: $71^\circ + 109^\circ + 180^\circ$ DWs: $M(H)$ cycle is enlarged (**coercivity enhancement**) and shifted (**exchange bias**)

exchange bias with BiFeO_3

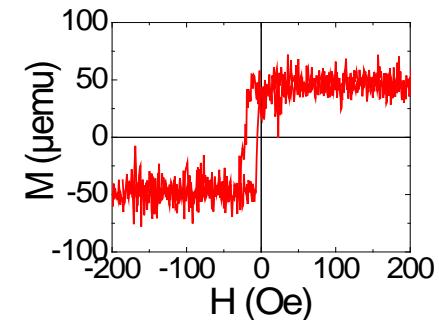
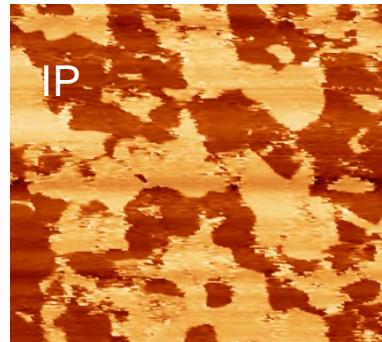


Malozemoff 's model extended to multiferroics:
 Malozemoff, Phys. Rev. B, 35, 3679 (1987)

$$H_e = -\frac{2zS_{AF}S_{FM}J_{ex}}{\mu_0 M_{FM} t_{FM} aL}$$

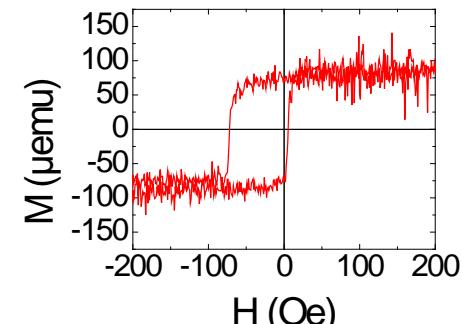
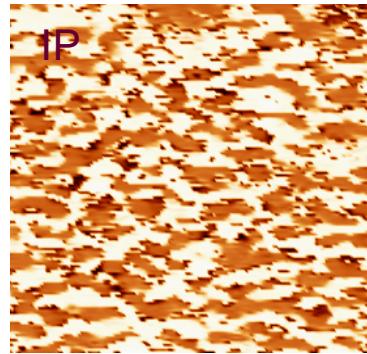
- H_e varies as the inverse of the **FE domain size**
- **Strong suggestion of Magnetoelectric coupling**

BFO(70nm)/SRO//STO (001)



L_{FE} large → H_e small

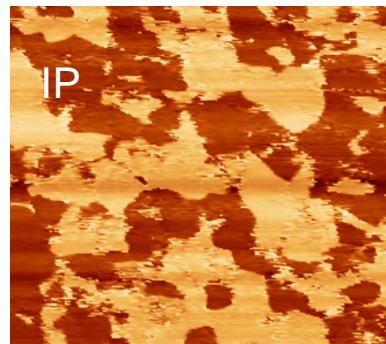
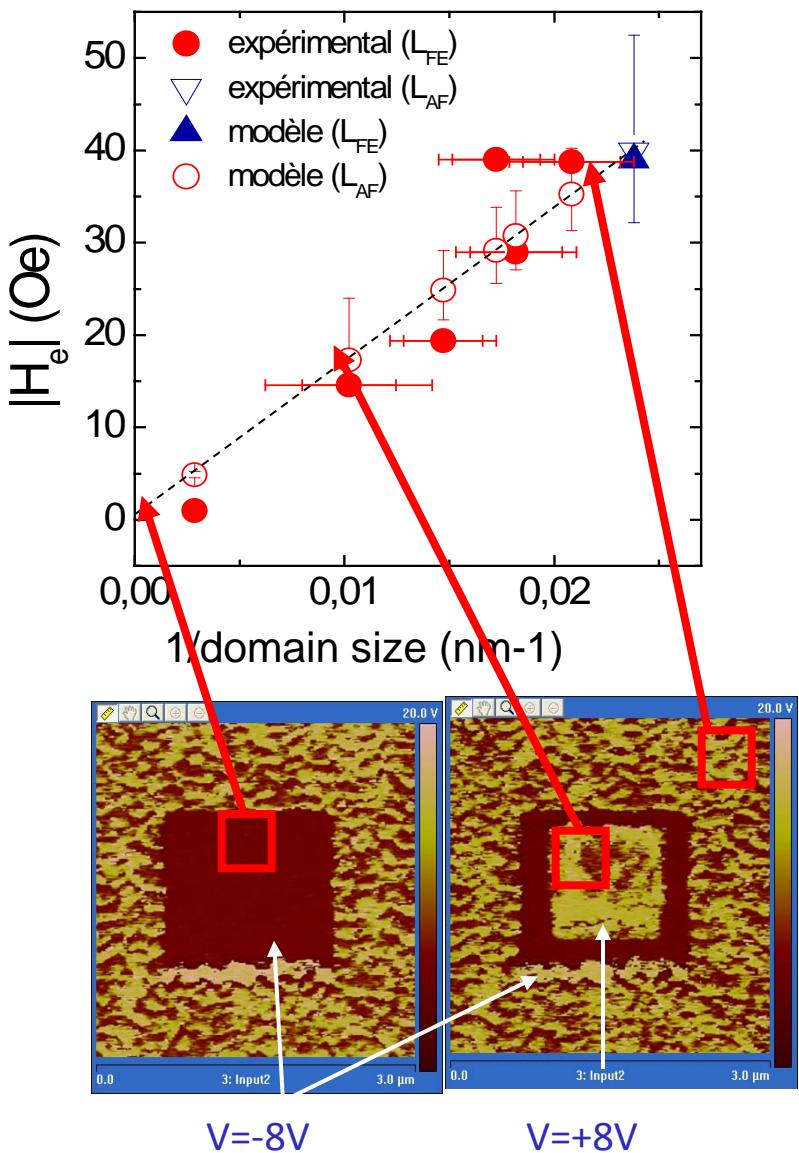
BFO(70nm)/SRO//STO (111)



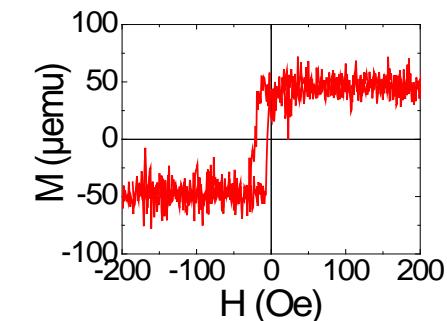
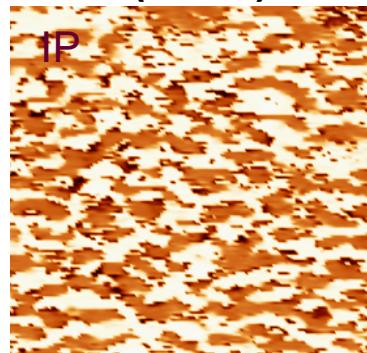
L_{FE} small → H_e large

exchange bias with BiFeO_3

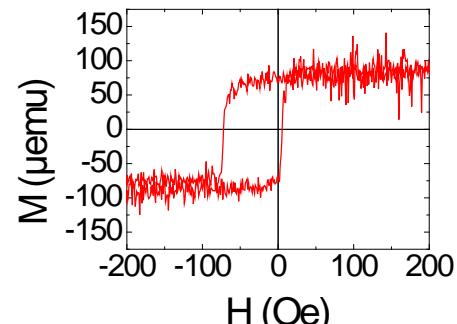
BFO(70nm)/SRO//STO (001)



BFO(70nm)/SRO//STO (111)



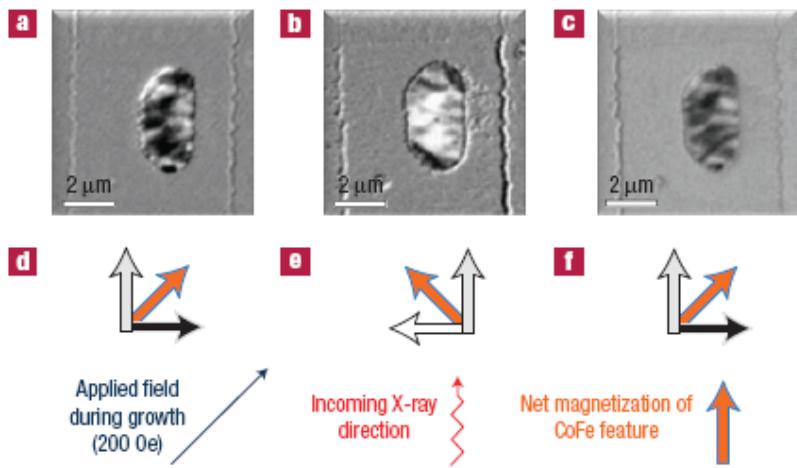
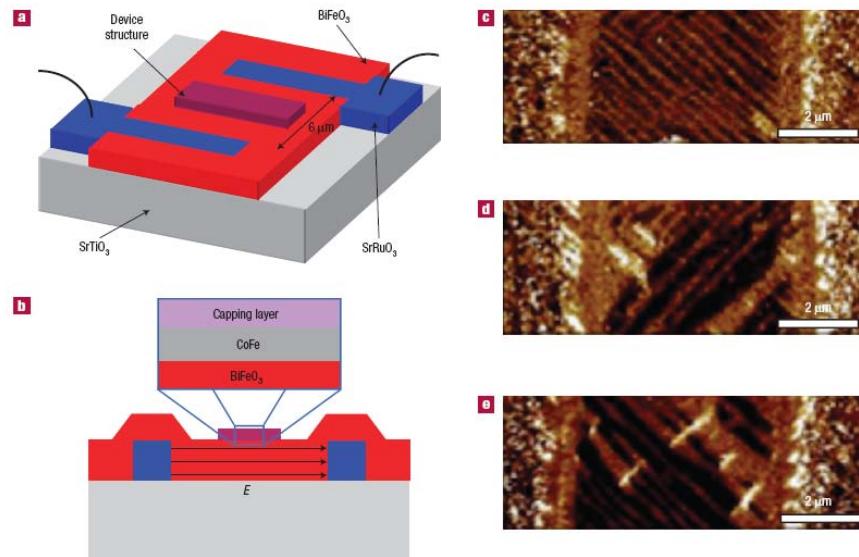
L_{FE} large \rightarrow H_e small



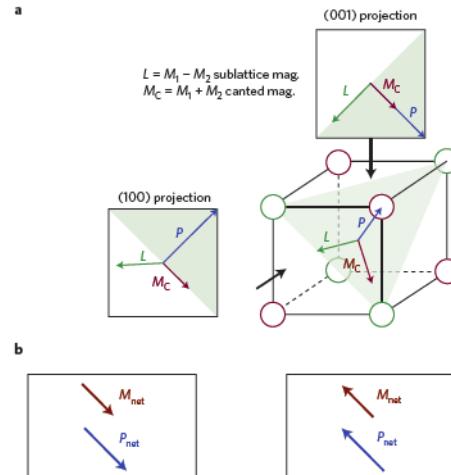
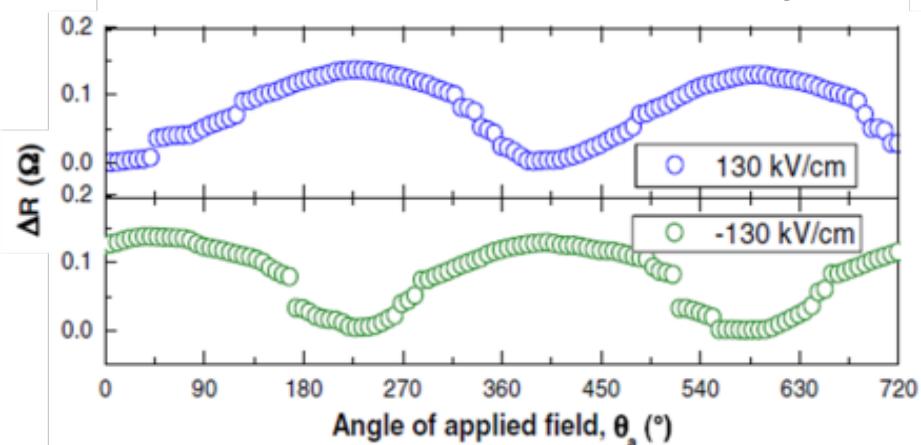
L_{FE} small \rightarrow H_e large

Towards the electric control of magnetic layer

Combination of PFM and XMCD-PEEM

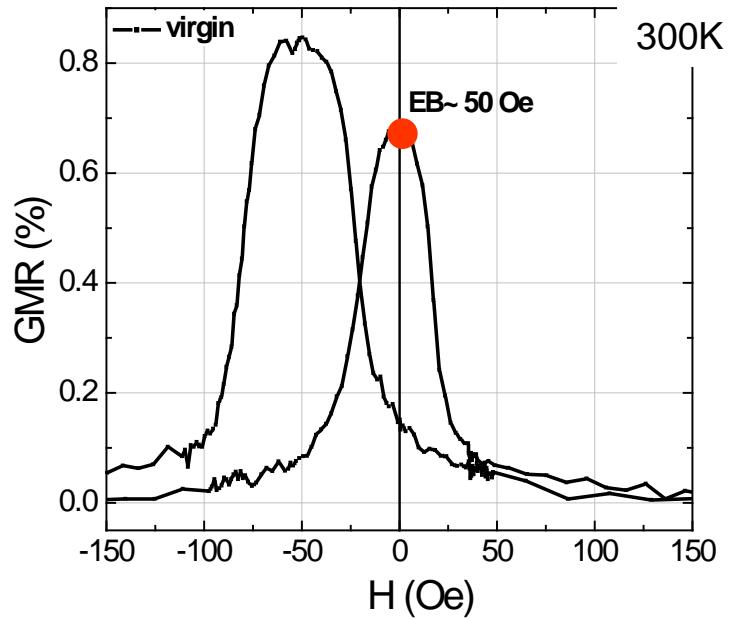
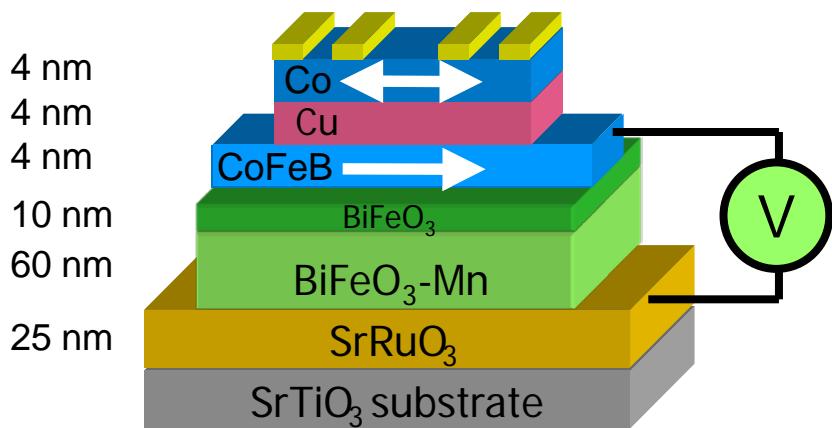


180° shift in the AMR: switching of magnetization



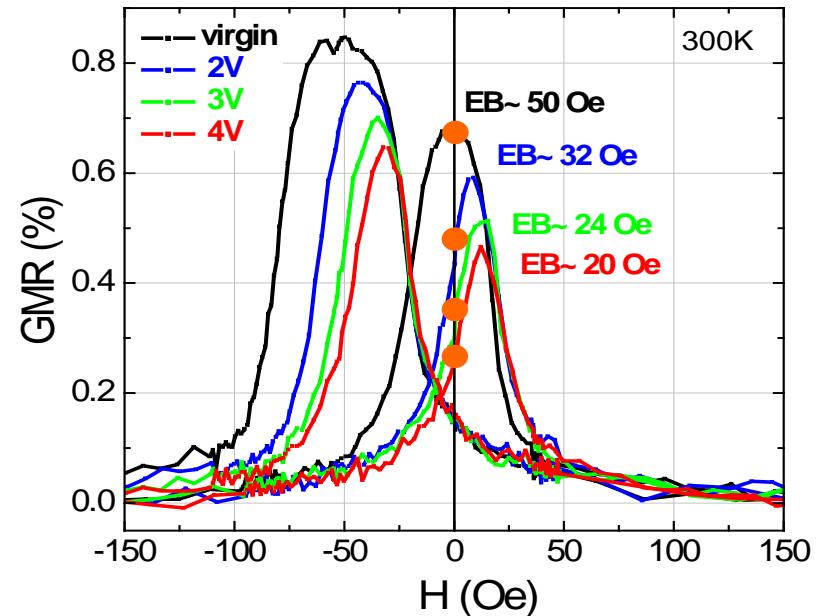
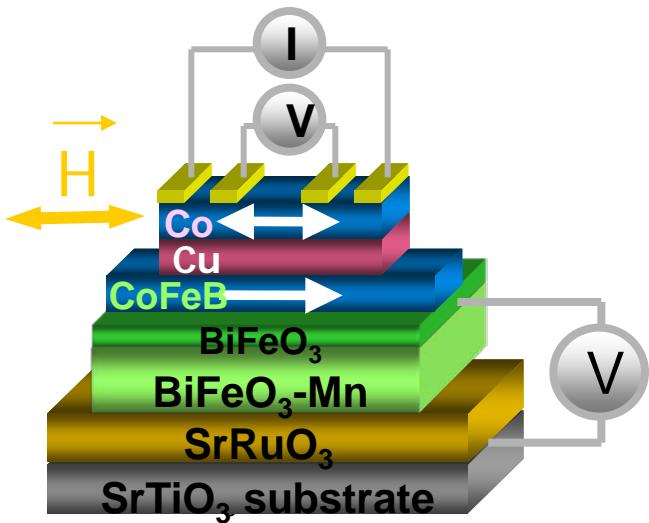
E. Chu et al.; Nat. Mat. 7, 478 (2008)
Heron et al.; PRL 107; 217202 (2011)

Towards the control of the spin valve by E-field

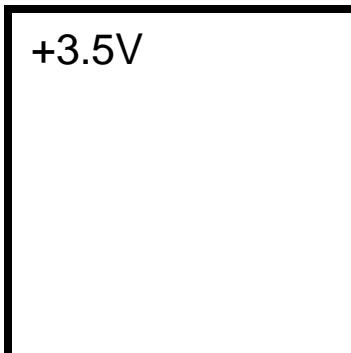
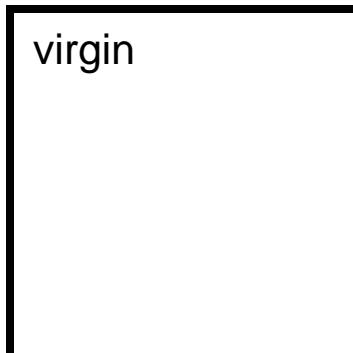


- ✓ Sizeable GMR effect
- ✓ Shifted by the exchange bias

Towards the control of the spin valve by E-field



- ✓ Change with E-field reflecting the change in exchange bias (related to FE domain)
- ✓ But non reversible
- ✓ Reversible effect obtained at LT using LuMnO_3 a FE/AFM non ferroelastic material (Skumryev et al.; PRL 106, 057206 (2011))



Electric control of Exchange-bias using a multiferroic

Advantages :

180° reversible rotation demonstrated at RT in planar device: so feasible applicable to all TM-FM



Inconvenients :

As to be demonstrated in vertical device: high density and small voltage
Mechanisms not yet very clear



That's it!

Thank you for your attention

Reviews:



- C.W. Nan et al.; J. Appl. Phys. 103, 031101 (2008)
- C. A. F. Vaz et al.; Adv. Mater. 22 (2010) 2900-2918; J. Phys. Condens. Matter. 24, 333201 (2012)
- J. Ma et al.; Adv. Mater. 23, 1062 (2011)
- A. Barthélémy, M. Bibes, Annual Review of Mat. Res. 44, 91 (2014)
- Garcia et al.; C. R. Physiques 16, 168 (2005)
- Matsukura et al.; Nat.Nano. 10, 2009 (2015)