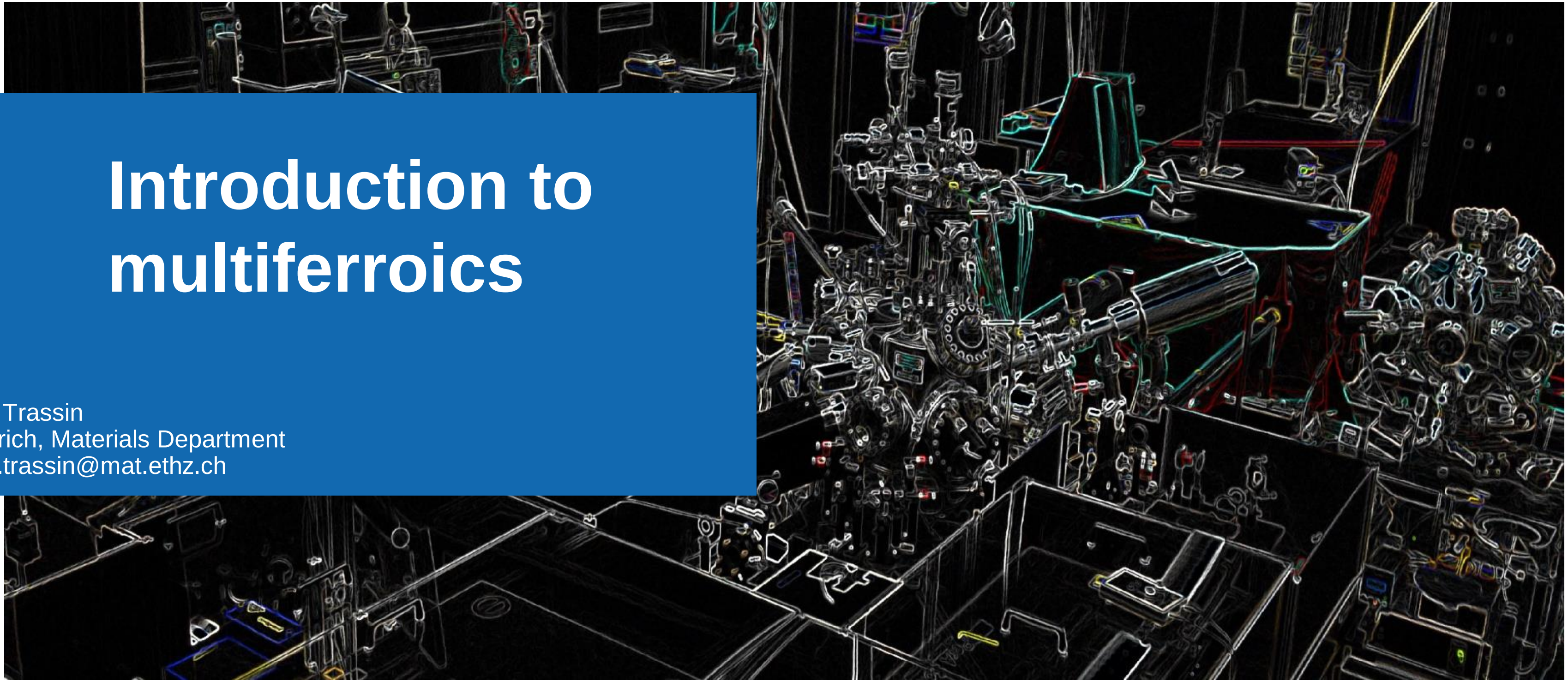


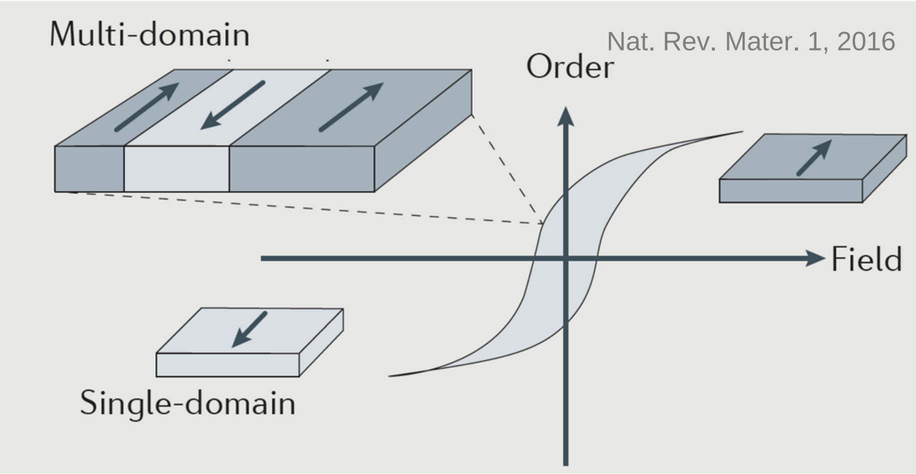
Introduction to multiferroics

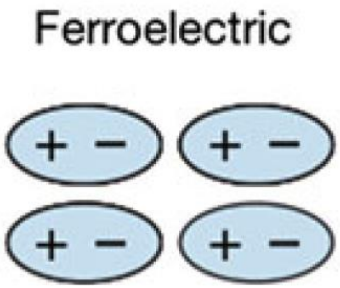
Morgan Trassin
ETH Zurich, Materials Department
morgan.trassin@mat.ethz.ch

Ferroic Materials (brief definition)

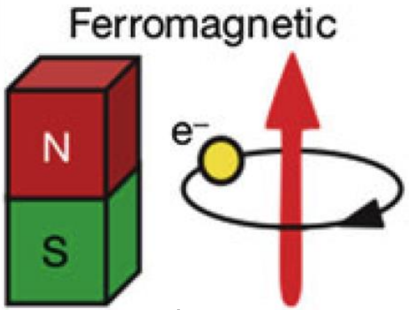
Materials displaying a long-range order with respect to at least one macroscopic property. They develop domains which orientation can be changed by a conjugate field. The ferroic state results in the observation of a nonvolatile switching and a hysteresis

Nat. Rev. Mater. 1, 2016
J. Phys.: Condens. Matter 28 (2016) 033001
Nature 449, 702 (2007)

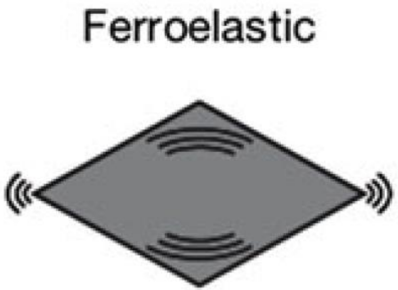




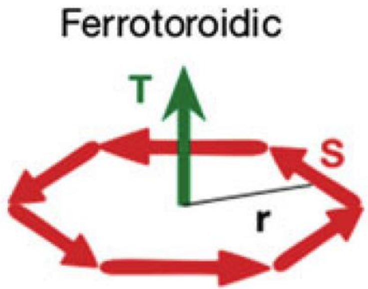
spontaneous polarization



spontaneous magnetization



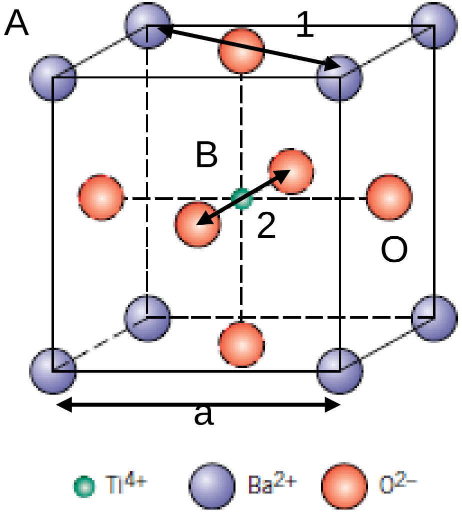
spontaneous strain



spontaneous magnetic vortex

Ferroelectric Materials

(brief reminder –
the perovskite case)



t = 1: Structure is frozen in the cubic phase

t > 1: B cation is small, can “move” within the unit cell

t < 1: B is too large, deviation from perovskite structure (oxygen octahedral distortion)

In the perfect cubic structure

$$a\sqrt{2} = 2R_A + 2R_O \qquad (1)$$

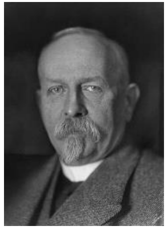
And

$$a = 2R_O + 2R_B \qquad (2)$$

Goldschmidt factor

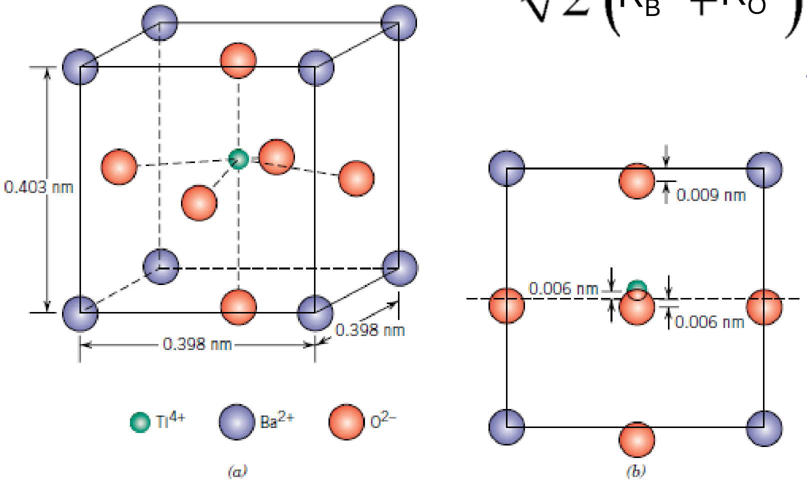
Defines the stability of crystal structures.
Predicts deviations from the cubic unit cell

$$t = \frac{R_A + R_O}{\sqrt{2}\,(R_B + R_O)}$$



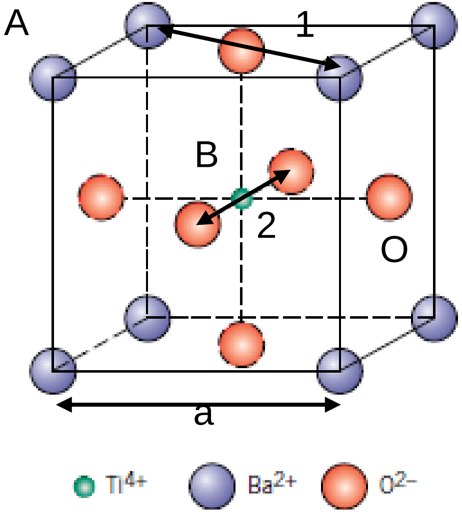
Victor Goldschmidt
(1888-1947)

Figure 18.35 A barium titanate ($BaTiO_3$) unit cell (*a*) in an isometric projection, and (*b*) looking at one face, which shows he displacements of Ti^{4+} and O^{2-} ions from the center of the face.



Ferroelectric Materials

(brief reminder –
the perovskite case)



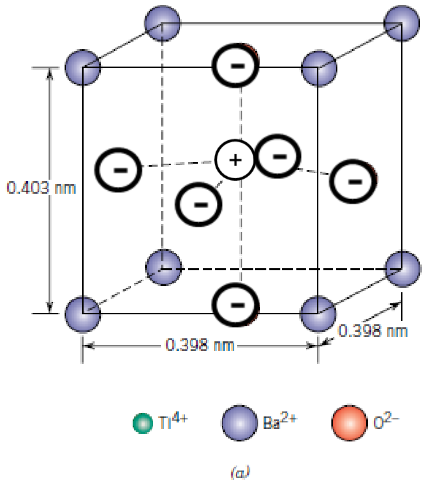
t = 1: Structure is frozen in the cubic phase

t > 1: B cation is small, can “move” within the unit cell

t < 1: B is too large, deviation from perovskite structure (oxygen octahedral distortion)

The B cation off-centering leads to the emergence of an electric dipole in the unit cell and a macroscopic polarization (**P**) within the material

Insulating (can’t be metallic)



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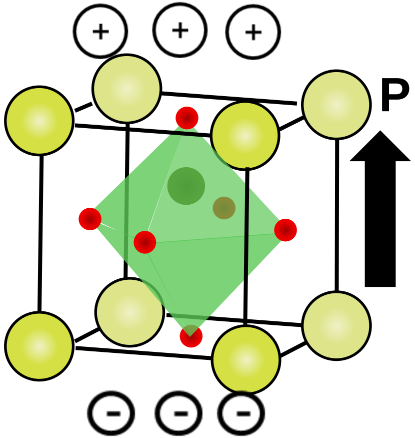
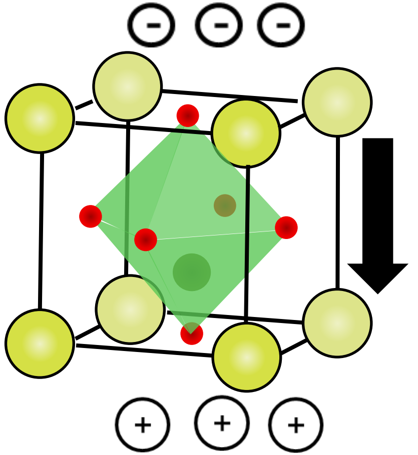




Ferroelectric Materials

(brief reminder –
the perovskite case)

How to measure ferroelectric polarization
In the ideal case (perfect insulator), we measure: $Q = 2P_r A$

Q charge measured, P_r polarization (at remanence), A electrode area

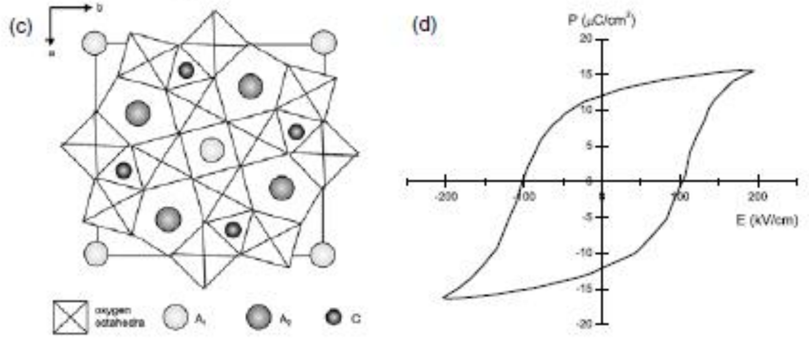
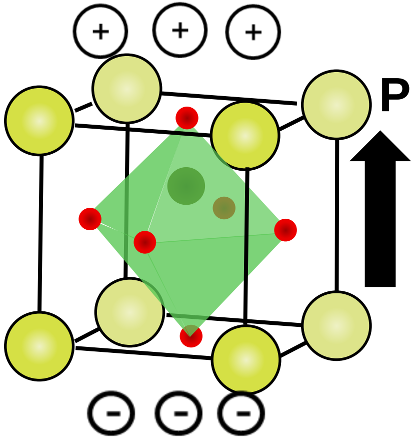
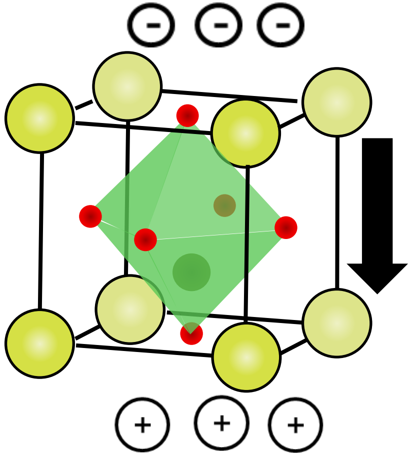


Figure 1. (a) Charge versus voltage loop typical for a lossy dielectric, in this case the skin of a banana (b) electroded using silver paste. The hysteresis loop for a truly ferroelectric material such as $Ba_2NaNb_5O_{15}$ (c) is shown in (d) ferroelectric hysteresis curve for ceramic barium sodium niobate (data from [24]).

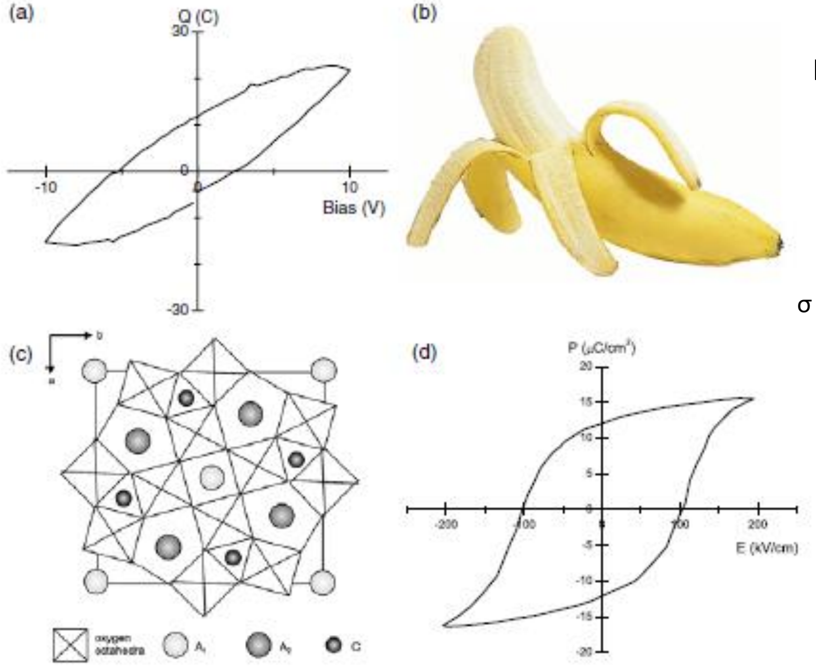
J. Phys.: Condens. Matter **20** 021001





Ferroelectric Materials

(brief reminder –
the perovskite case)



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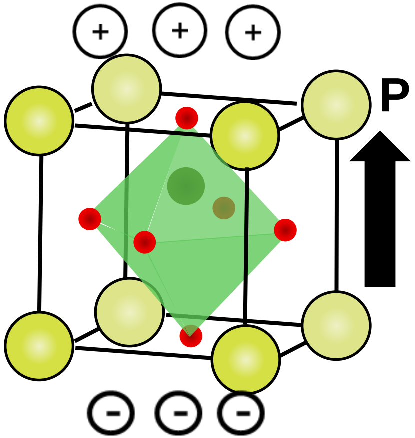
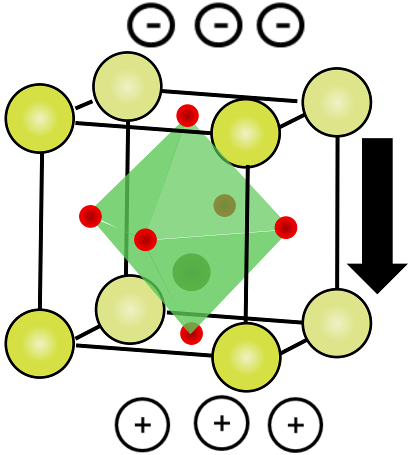
In reality, we measure:

$$Q = 2P_r A + \sigma E A t.$$

σ electrical conductivity, E applied field, t measuring time

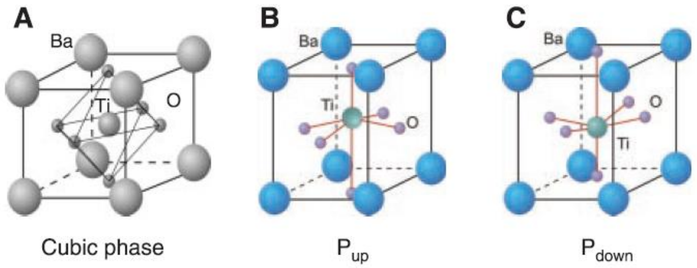
J. Phys.: Condens. Matter **20** 021001





Ferroelectric Materials
(brief reminder)

Ceramics

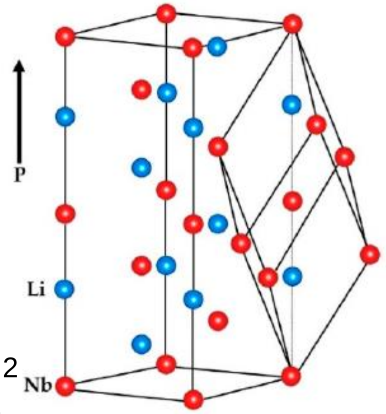


Science (2004), 303, pp. 488-491

$BaTiO_3$, P ≈ 20 μC/cm^2

Pb(Zr,Ti)O_3, P ≈ 40 μC/cm^2

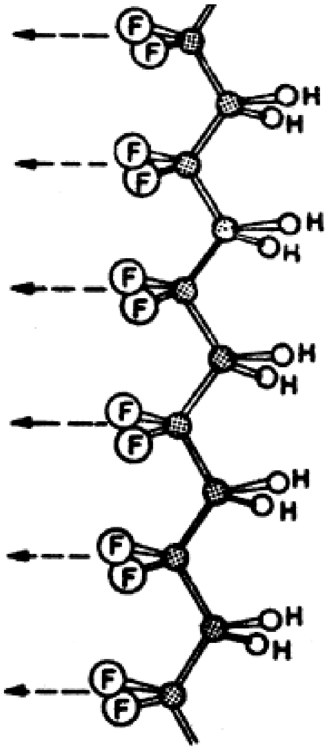
$BiFeO_3$, P ≈ 60 μC/cm^2



$LiNbO_3$, P ≈ 50-90 μC/cm^2

Crystals 2020, 10, 973

Polymers



Polyvinylidene fluoride (PVDF)
P ≈ 5 μC/cm^2

Polymers, Ferroelectric (2003)

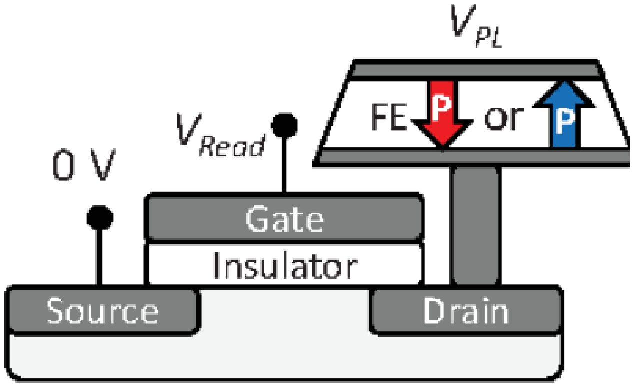
Ferroelectric Materials

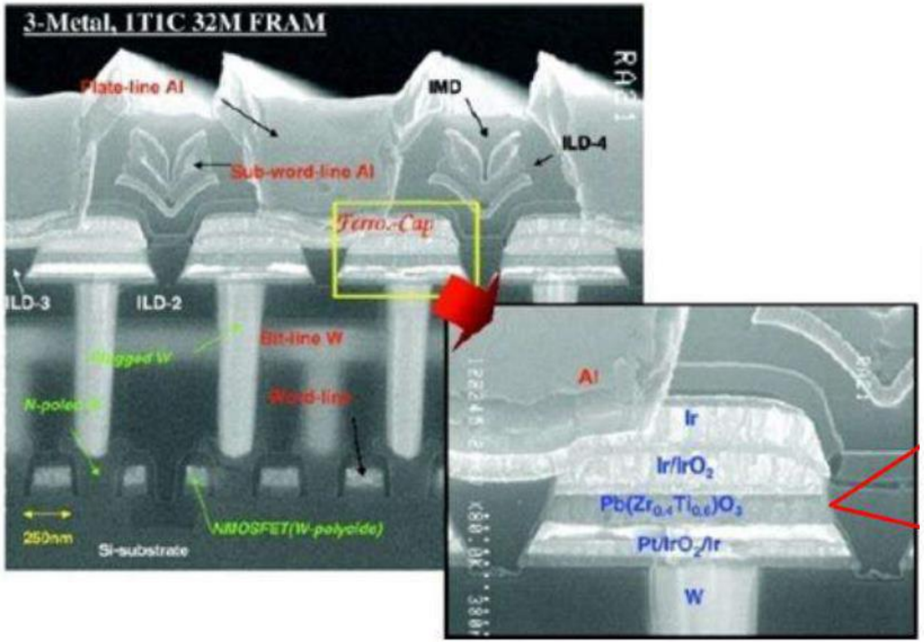
(brief reminder)

- ## Ferroelectric memory and DRAM

DRAM is a volatile type of memory, the charged accumulated in the capacitor slowly leaks and the memory needs to be rewritten constantly (refreshed ~ 16 times/sec).

The insertion of ferroelectric materials in FeRAM renders the memory storage non-volatile.



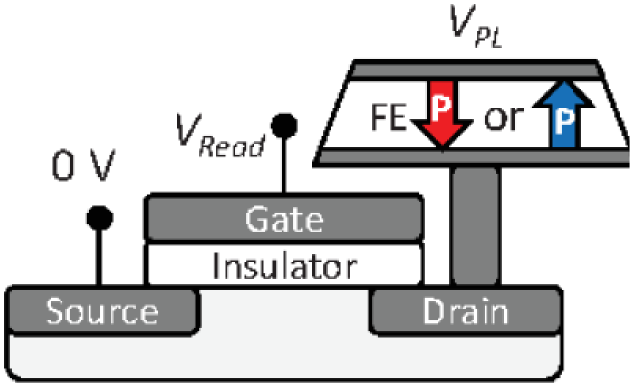


Ferroelectric Materials
(brief reminder)

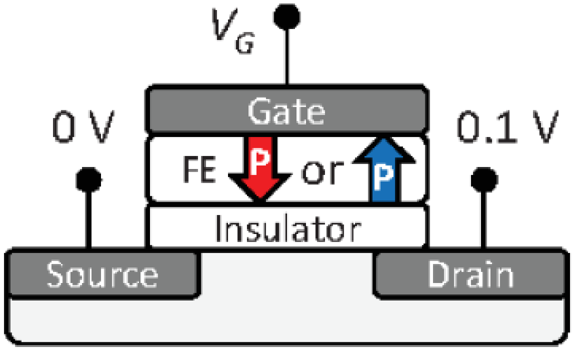
- Ferroelectric memory, the Ferroelectric Field Effect Transistor (FeFET)

Writing the state of the of the Ferroelectric is performed by applying an electric field. The reading process is based on the switching current detection. The reading is destructive, the state needs to be restored after each read operation.
The ferroelectric field effect transistor architecture is now developed for non-destructive readout.

FeRAM



FeFET



Ferroelectric Materials

(brief reminder)

PRL **94,** 246802 (2005) PHYSICAL REVIEW LETTERS week ending 24 JUNE 2005

Giant Electroresistance in Ferroelectric Tunnel Junctions

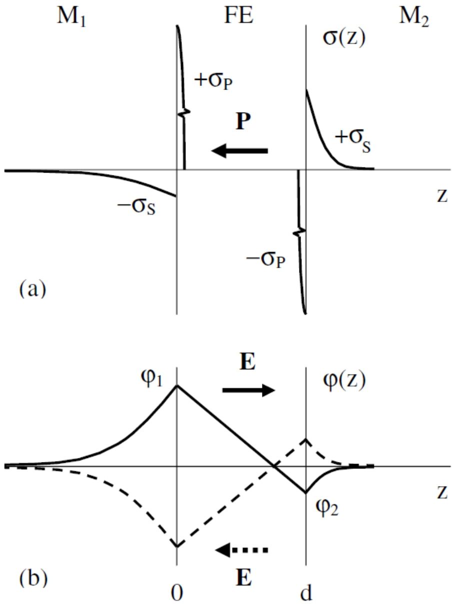
M. Ye. Zhuravlev,[1] R. F. Sabirianov,[2,3] S. S. Jaswal,[1,3] and E. Y. Tsymbal[1,3,*]

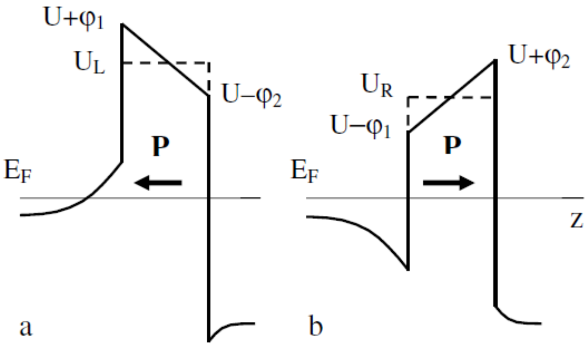
[1]*Department of Physics and Astronomy, University of Nebraska, Lincoln, Nebraska 68588-0111, USA*

[2]*Department of Physics, University of Nebraska, Omaha, Nebraska 68182-0266, USA*

[3]*Center for Materials Research and Analysis, University of Nebraska, Lincoln, Nebraska 68588-0111, USA*

(Received 2 February 2005; published 20 June 2005)





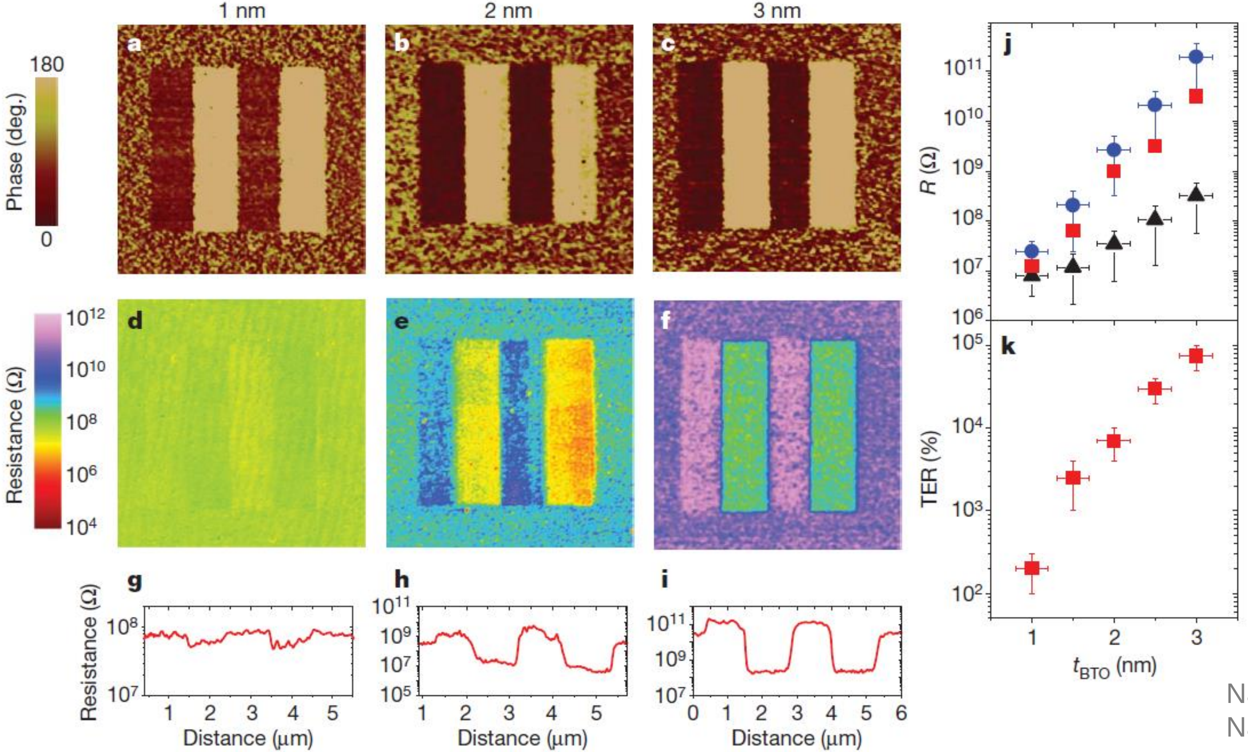
Ferroelectric tunnel junction (FTJ)

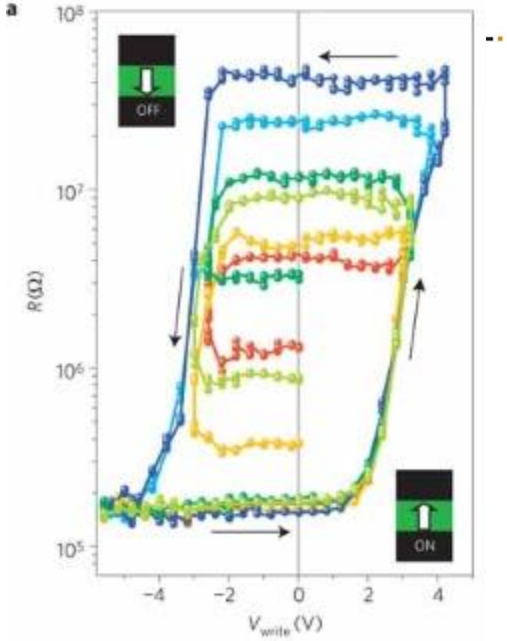
- Electrostatic effect – Imperfect screening at the ferroelectric/metal interface leading to asymmetric potential profile seen by the transport electrons.
- Interface atomic displacement, polarization dependent orbital hybridization at the interface.
- Strain effect, tunnel barrier width vary under electric field application.

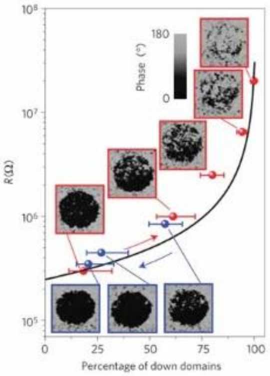
Ferroelectric Materials

(brief reminder)

Non volatile readout of polarization state







Nature 460, 81 (2009)

Nat. Mater.11, 860 (2012)

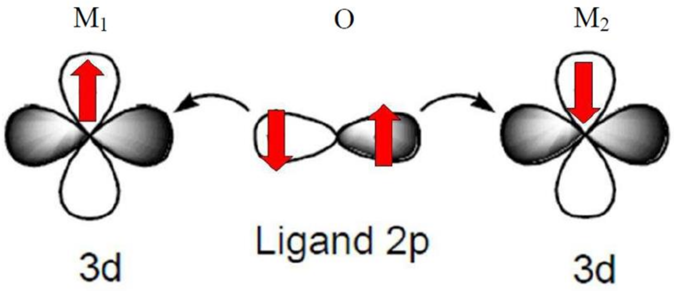
DMATL.

Magnetically ordered Materials
(brief reminder –
the perovskite case)

In oxides two types of interactions dominate

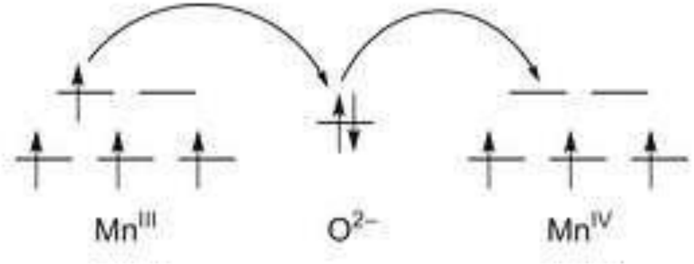
- Super exchange

Magnetic ions separated by O^{2-}
The distance between d orbital does not allow a direct overlap



- Double exchange

Magnetic species exhibiting mixed-valence states (Mn^{3+}, Mn^{4+} in La,SrMnO_3 or Fe^{2+},Fe^{3+} in Fe_3O_4) The electron mobility of the less oxidized state stabilizes a parallel arrangement of the spins



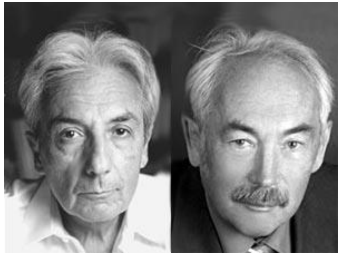
Magnetically ordered Materials

(brief reminder –
the perovskite case)

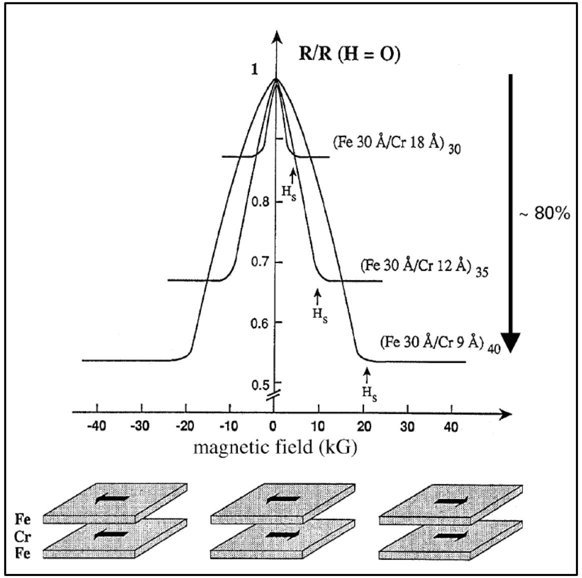
The discovery of “Giant Magnetoresistance” (GMR), leading to the radical miniaturizing of hard drives

Magnetic based data storage

Alber Fert, Peter Grünberg
Physics Nobel 2007



Phys. Rev. Lett. 61 (1988) 2472.



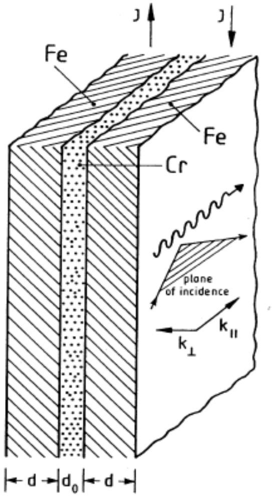
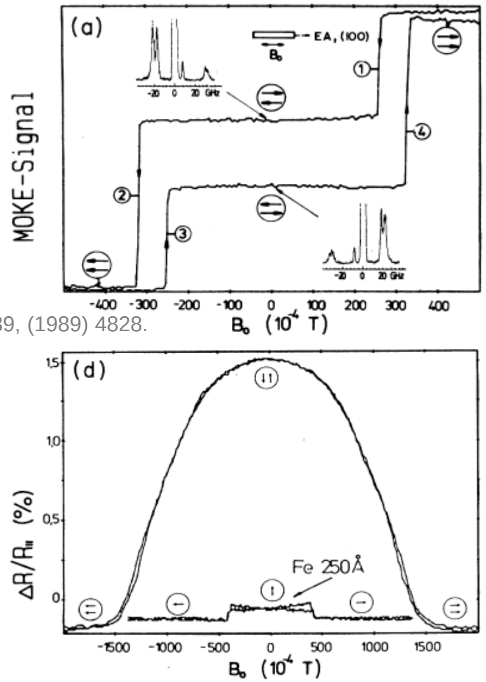


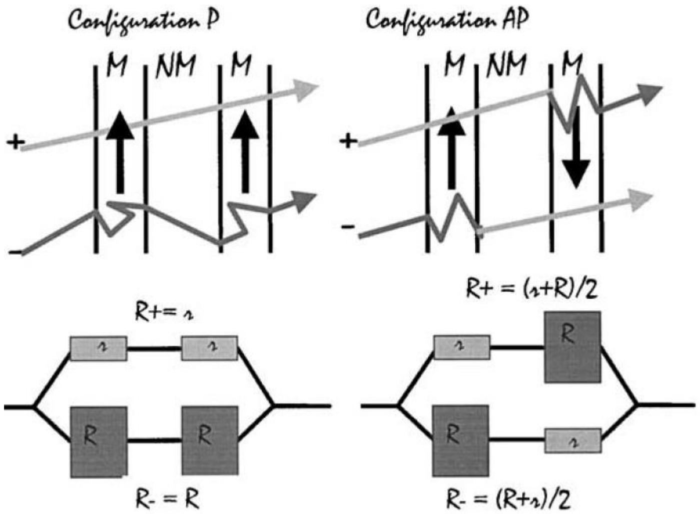
FIG. 1. Ferromagnetic double layer with antiparallel alignment of the magnetizations. Also indicated is the plane of incidence of the laser light for the observation of light scattering from spin waves and hysteresis curves via MOKE.



Phys. Rev. B 39, (1989) 4828.

Magnetically ordered Materials (brief reminder – the perovskite case)

J. of Mag. and Mag. Mater. 242–245 (2002) 68–76



1956 IBM R&D Laboratory
1000 kg
5 Mo
500 bits/cm2

https://www.computerhistory.org

https://www.cpu-galaxy.at/

Nowadays
few grams
> 8 TB

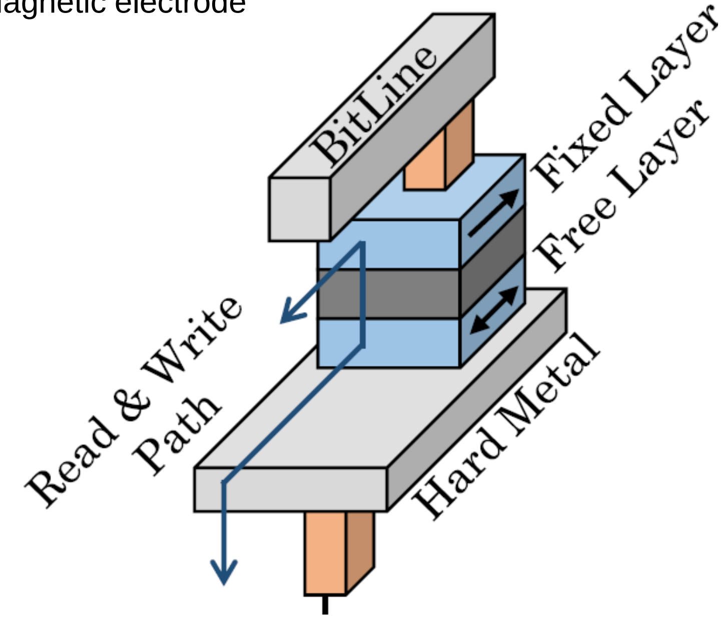
From 1997, GMR read head were out. At this point it becomes cheaper to store data on magnetic hard disk than to print it out on paper

-> great impact on our society (social media, google, big data, etc…)

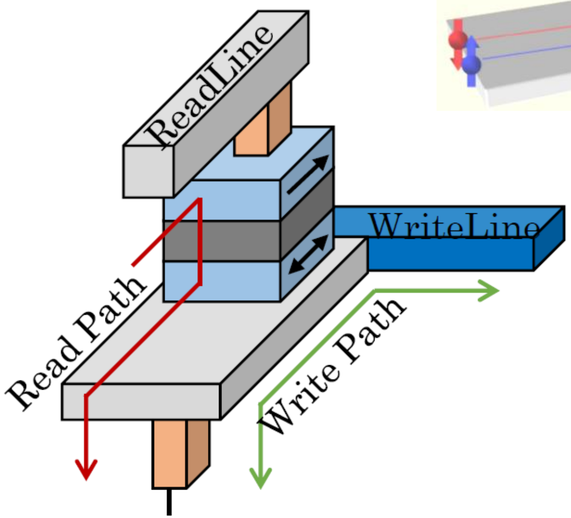
Magnetically ordered Materials

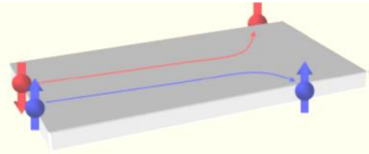
(brief reminder –
the perovskite case)

Spin Transfer Torque
switching using spin-polarized currents generated through the magnetic electrode



Spin Orbit Torque
The spin Hall effect in high spin-orbit material (Pt, W) results in a spin accumulation that induces a magnetic torque





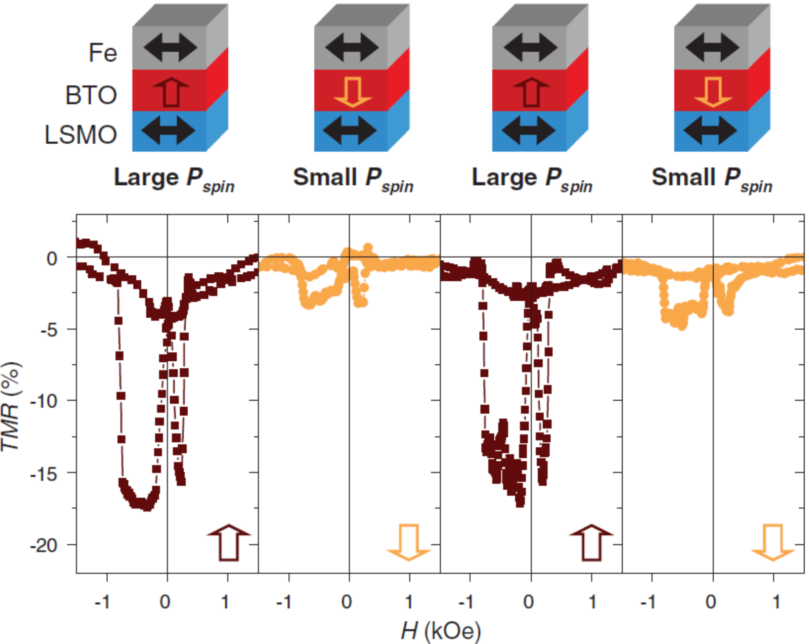
J. Low Power Electron. Appl. 2017, 7(3), 23
2008 J. Magn. Magn. Mater. 320 1190
2010 Nat. Mater. 9 230
J. Phys.: Condens. Matter 28 (2016) 033001

Combining Ferroelectric and Magnetic order

Multiferroic memory
Data storage indepently in polarization and magnetic states
Combining magnetoresistance and electroresistance

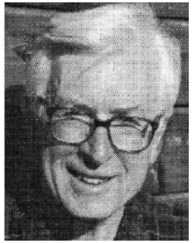
Using magnetic electrodes and ferroelectric barrier
Increased storage density, 4 resistance states

Science 327, 1106 (2010)



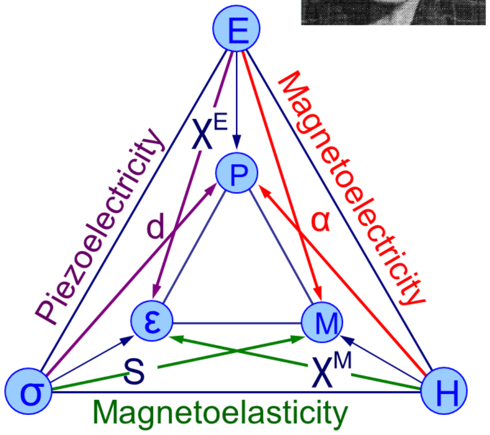
Using a **multiferroic material**

Crystals can be defined as multiferroic when two or more of the primary ferroic properties are united in the same phase.



Hans Schmid
Ferroelectrics 162, 665 (1994)

J. Phys. D 38, R123 (2005)
Nature 442, 759 (2006)
Nat. Mater. 6, 21 (2006)
Nat. Mater. 6, 13 (2006)

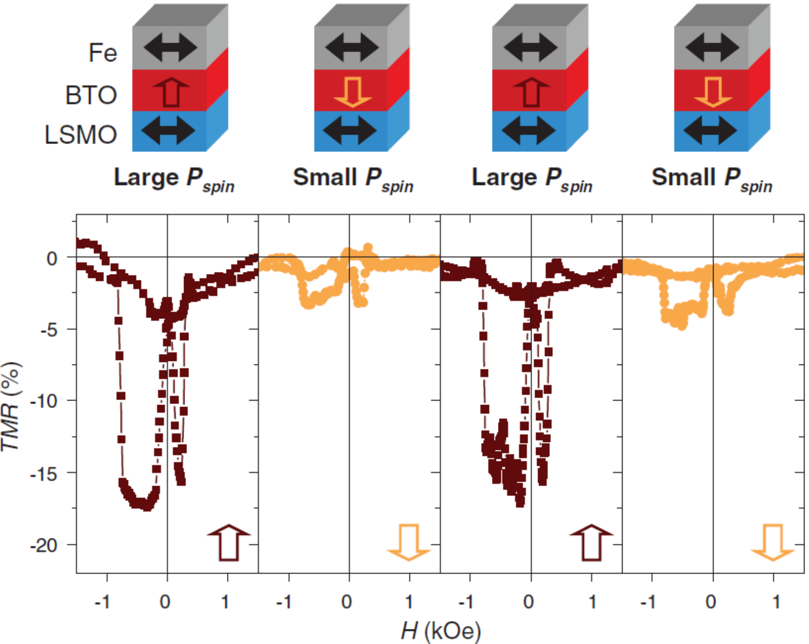


Combining Ferroelectric and Magnetic order

Multiferroic memory
Data storage indepently in polarization and magnetic states
Combining magnetoresistance and electroresistance

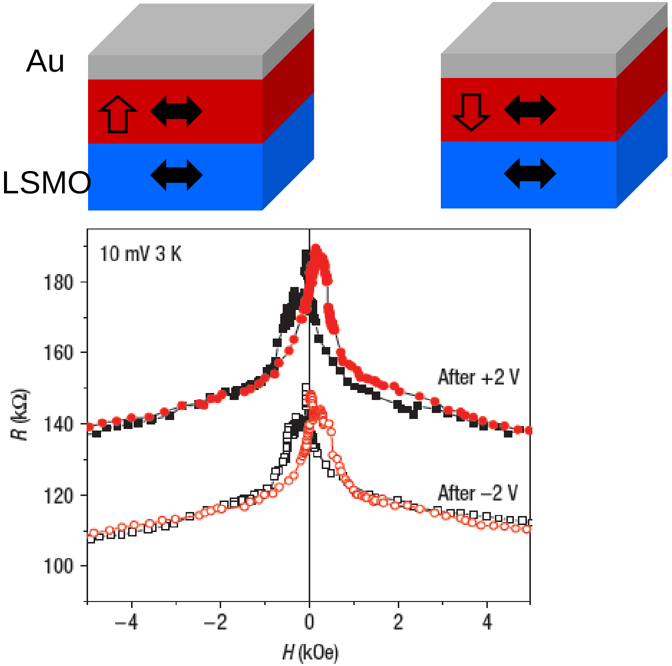
Using magnetic electrodes and ferroelectric barrier
Increased storage density, 4 resistance states

Science 327, 1106 (2010)



Using a **multiferroic material**

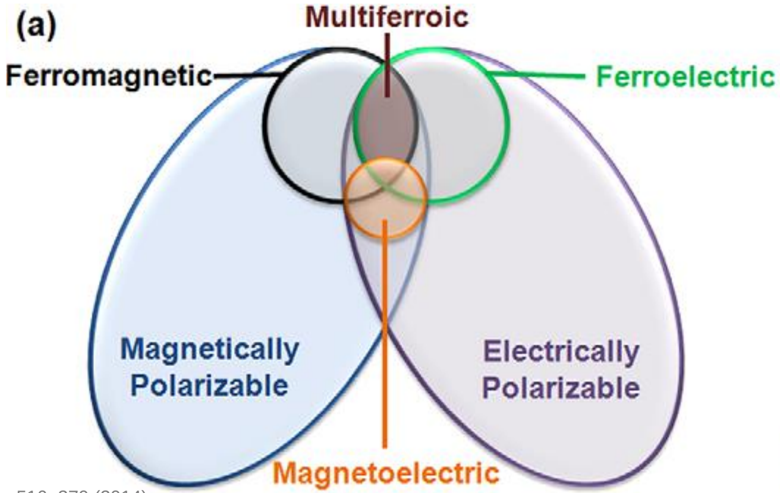
Using a multiferroic tunnel barrier



Nat. Mater. 6, 296 (2007)

Combining Ferroelectric and Magnetic order

Magnetoelectric memory
«Easy» magnetic readout
Low energy consuming ferroelectric «write»



Nature 516, 370 (2014)

Using multiferroic magnetoelectric materials with coexisting and coupled magnetic and ferroelectric order parameters

Electrical Manipulation of Magnetization Reversal in a Ferromagnetic Semiconductor

D. Chiba,[1] M. Yamanouchi,[1] F. Matsukura,[1] H. Ohno[1,2*]

SCIENCE VOL 301 15 AUGUST 2003

PRL **101,** 137201 (2008) PHYSICAL REVIEW LETTERS week ending 26 SEPTEMBER 2008

Surface Magnetoelectric Effect in Ferromagnetic Metal Films

Chun-Gang Duan,[1] Julian P. Velev,[2,3] R. F. Sabirianov,[3,4] Ziqiang Zhu,[1] Junhao Chu,[1] S. S. Jaswal,[2,3] and E. Y. Tsymbal[2,3]
[1]*Key Laboratory of Polarized Materials and Devices, Ministry of Education, East China Normal University, Shanghai 200062, China*
[2]*Department of Physics and Astronomy, University of Nebraska, Lincoln, Nebraska 68588, USA*
[3]*Nebraska Center for Materials and Nanoscience, University of Nebraska, Lincoln, Nebraska 68588, USA*
[4]*Department of Physics, University of Nebraska, Omaha, Nebraska 68182, USA*
(Received 25 June 2008; published 22 September 2008)

nature materials LETTERS
PUBLISHED ONLINE: 13 NOVEMBER 2011 | DOI:10.1038/NMAT3172

Induction of coherent magnetization switching in a few atomic layers of FeCo using voltage pulses

Yoichi Shiota[1], Takayuki Nozaki[1,2†], Frédéric Bonell[1], Shinichi Murakami[1,2], Teruya Shinjo[1] and Yoshishige Suzuki[1,2*]

Ferroelastic Materials

Ferroelasticity is defined by its hysteresis, as are its sister ferroic properties, ferroelectricity and ferromagnetism (**Figure 1**). An elastic hysteresis represents the effect of the mechanical switching between at least two orientation states of a crystal by external stress. One of the first full hysteresis loops was seen in 1976 for the prototypic material $Pb_3(PO_4)_2$ (3, 9). Macroscopic hysteresis relates to the switching of atomic positions, usually across twin boundaries. Such switching occurs between ferroelastic states. These states relate in $Pb_3(PO_4)_2$ to the displacement of Pb inside its oxygen coordination (**Figure 1**) (10–16). The Pb atom establishes chemical bonds with two oxygen positions with shorter bond distances than the distances between Pb and the remaining four oxygen atoms. This anisotropic bonding shears the structural network and lowers the symmetry of the crystal structure from trigonal to monoclinic. The different orientations of short bond distances correspond to the different orientational strain states, and mechanical switching occurs between these states. The size of the ferroelastic hysteresis depends on thermodynamic

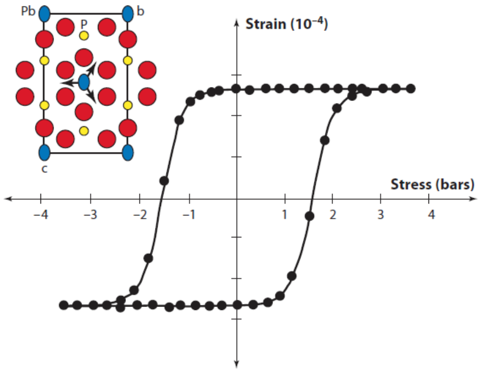
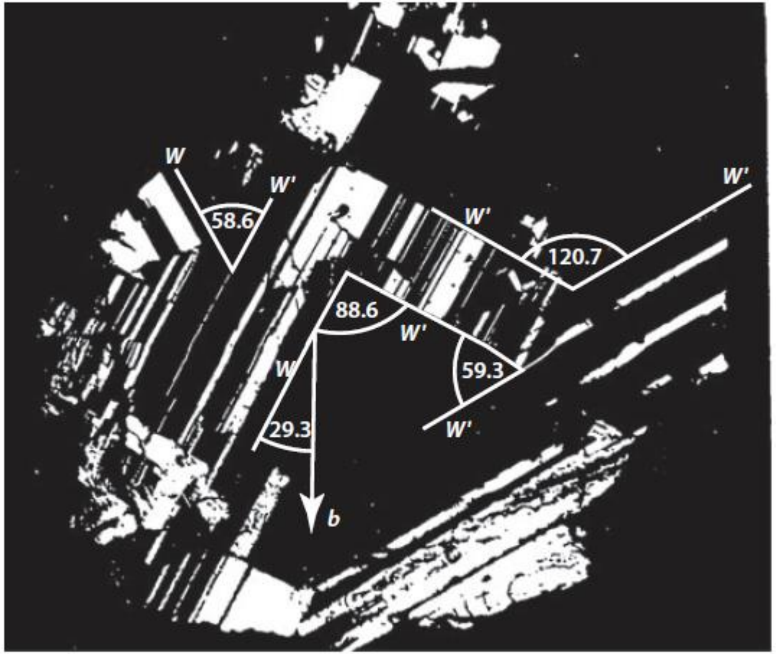


Figure 1

Ferroelastic hysteresis and atomic switching in $Pb_3(PO_4)_2$. The structural mechanism for ferroelasticity originates from the shift of the Pb atom away from the center of its oxygen cage towards one pair of oxygen. This leads to a monoclinic distortion of the crystal structure that can be inverted or rotated (in the direction of the arrows) under external stress.

Some ferroelectric switchings correspond to ferroelastic events, 90˚ domains in $BaTiO_3$



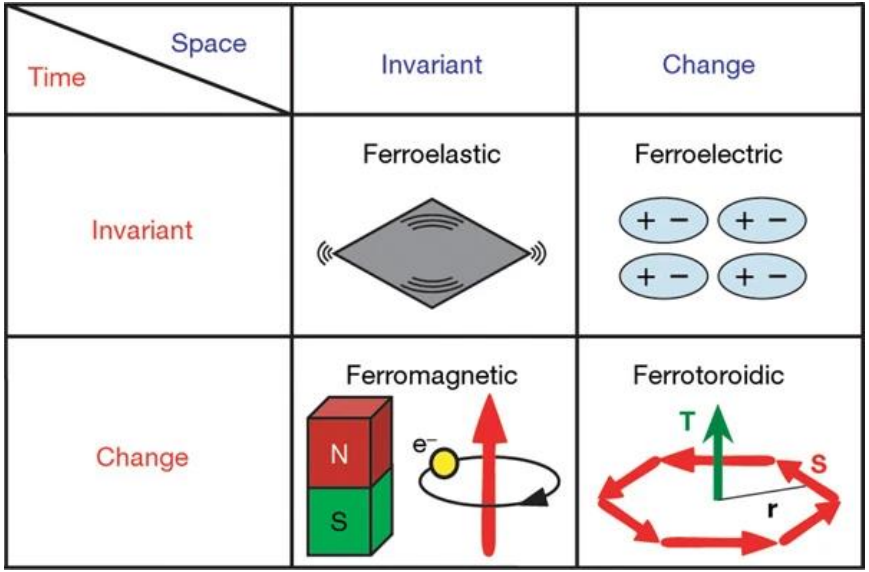
Annu. Rev. Mater. Res. 42, 265-283 (2012)

Ferrotorroidic Materials

D MATL.

Classification of the ferroic states

Nature 449, 702 (2007)



The magnetic order breaks inversion symmetry, inherent magnetoelectric materials

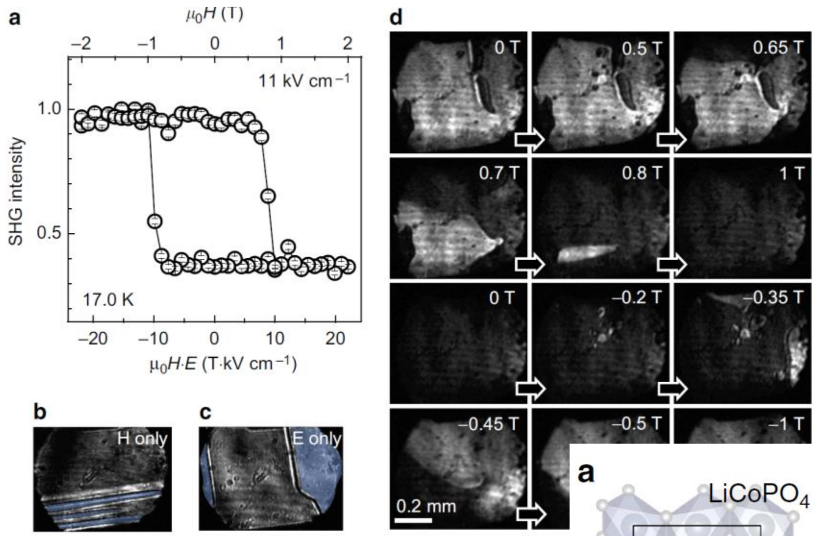
ARTICLE

Received 28 Nov 2013 | Accepted 24 Jul 2014 | Published 5 Sep 2014

DOI: 10.1038/ncomms5796

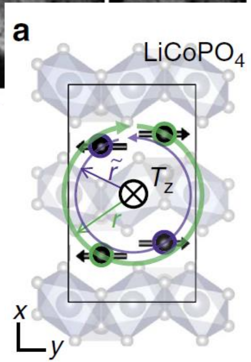
Ferroic nature of magnetic toroidal order

Anne S. Zimmermann[1], Dennis Meier[2] & Manfred Fiebig[2]



The conjugate field is a combination of electric and magnetic field

The magnetic sublattices in $LiCoPO_4$ play the role of the torus

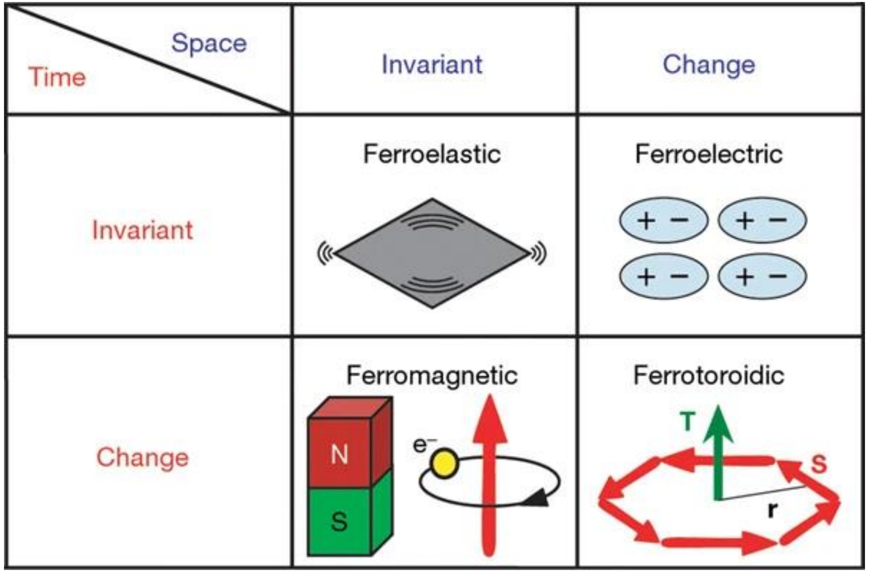


ETH zürich

Ferrotorroidic Materials

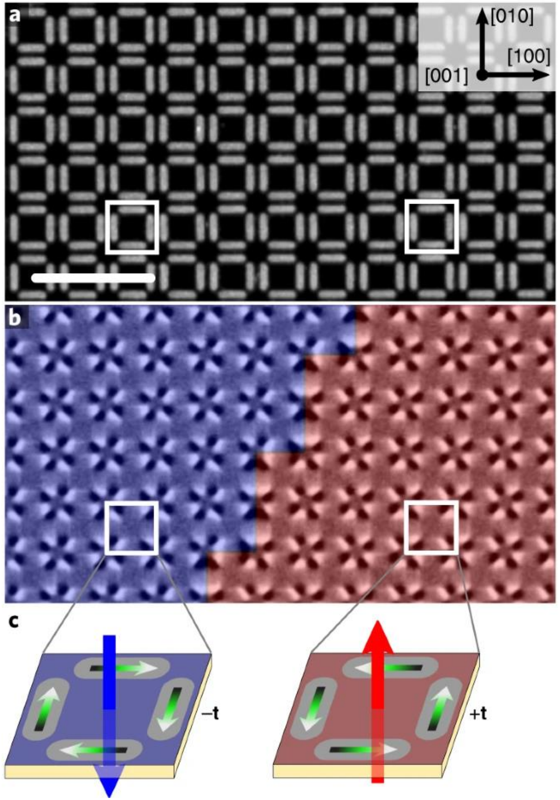
Classification of the ferroic states

Nature 449, 702 (2007)



The magnetic order breaks inversion symmetry, inherent magnetoelectric materials

Alternative to antiferromagnetic poling



Nat. Nano. 14, 141–144 (2019)

Multiferroics

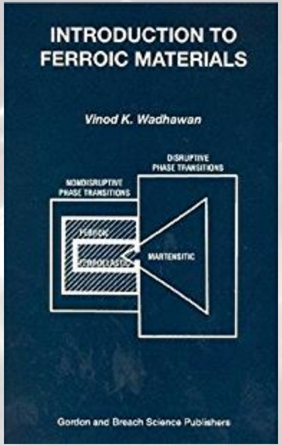
- Why are there so few ferromagnetic-ferroelectric materials?
- Mechanisms promoting the coexistence of magnetic and electric long-range orders (type I and type II multiferroics)

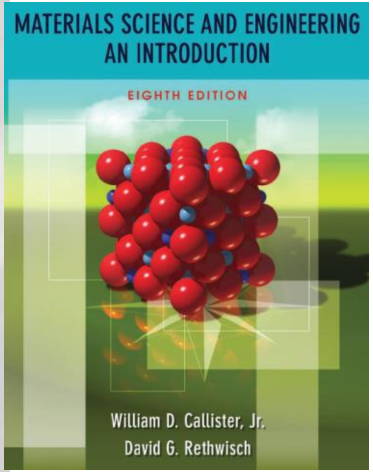
Electric-field control of ferromagnetism using multiferroics

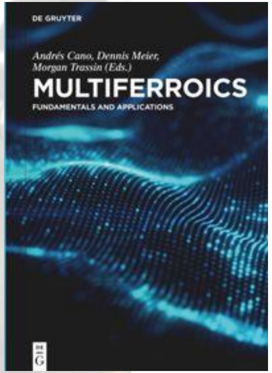
- Artificial / synthetic multiferroics
- Electric-field-induced magnetization reversal

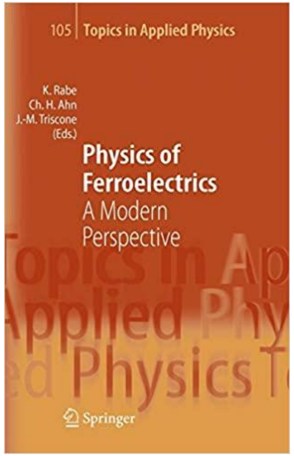
On the way to the ultimate goal

- Characterization of multiple order parameters
- Beyond the low energy control of magnetization









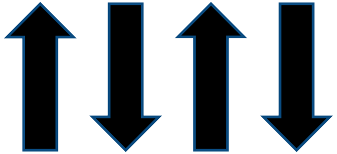
Magnetically ordered Materials

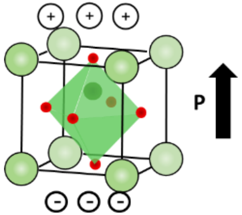
(brief reminder –
the perovskite case)

In oxides two types of interactions dominate

- Super exchange

Magnetic ions separated by O^{2-}, The distance between d orbital does not allow a direct overlap

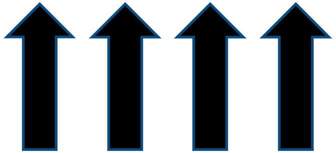


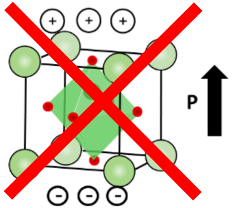


No net M

- Double exchange

Magnetic species exhibiting mixed valence states (Mn^{3+}, Mn^{4+} in La,SrMnO_3 or Fe^{2+},Fe^{3+} in Fe_3O_4) The electron mobility of the less oxidized state stabilizes a parallel arrangement of the spins





M

6694 J. Phys. Chem. B 2000, 104, 6694–6709

D MATL.

FEATURE ARTICLE

Why Are There so Few Magnetic Ferroelectrics?

Nicola A. Hill
Materials Department, University of California, Santa Barbara, California 93106-5050

Received: January 7, 2000; In Final Form: April 25, 2000

The B cation displacement is stabilized an orbital hybridization
In $BaTiO_3$, the Ti^{4+} is formally in a d^0 state so that the lowest unoccupied energy levels are d states that tend to hybridize with O 2p ions.

→ Size of B cation and "d^0-ness"

By definition, ferroelectric materials break inversion symmetry

Among the 31 point group allowing a spontaneous polarization (Pyroelectrics), only 13 allow coexisting spontaneous magnetization

→ Symmetry

The polarization in ferroelectric materials can be reversed by the application of an external electric field

Hence a ferroelectric material must be an insulator otherwise an applied electric field would induce an electrical current flow rather than causing an electric al polarization reversal.

→ Electrical property

Ferroelectricity mechanisms compatible with magnetic order

(Type I multiferroics)

Nat. Rev. Mater. 1, 2016

Lone pair stereochemical activity at the A-site, spontaneous electric polarization due to the displacement $6s^2$ lone pairs in BiFeO3 T_c ~ 1100 K and BiMnO3 T_c ~ 760 K

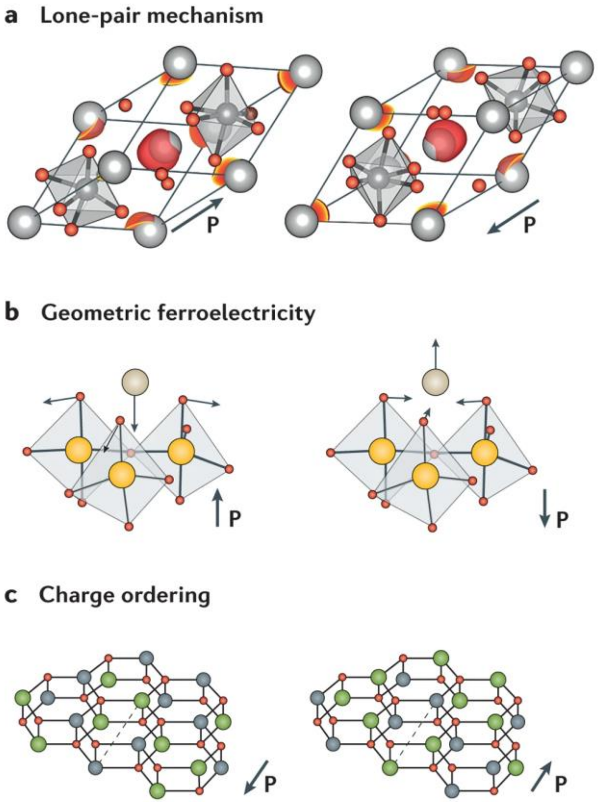
Magnetic order emerges from B cation

Geometric ferroelectrics, the electric polarization is driven by a rotational instability of the polyhedra and displacement of the A-site cation, $BaNiF_4$ or hexagonal $YMnO_3$ T_c ~ 930 K

Magnetic order emerges from the manganese lattice

Spatial ordering of charges in a non-centrosymmetric arrangement, Fe2+ and Fe3+ in $LuFe_2O_4$

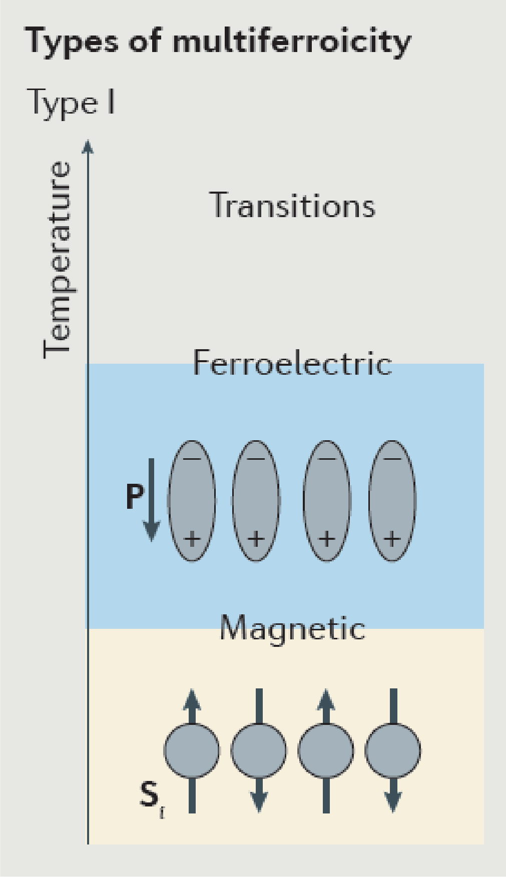
Magnetic order emerges from the iron lattice



Ferroelectricity mechanisms compatible with magnetic order

(Type I multiferroics)

Nat. Rev. Mater. 1, 2016



Lone pair stereochemical activity at the A-site, spontaneous electric polarization due to the displacement $6s^2$ lone pairs in BiFeO3 T_c ~ 1100 K and BiMnO3 T_c ~ 760 K

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Magnetic order emerges from the manganese lattice

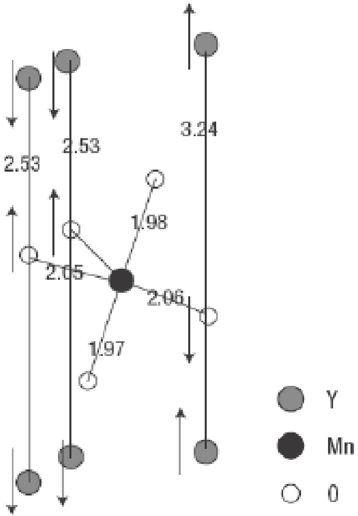
Spatial ordering of charges in a non-centrosymmetric arrangement, Fe2+ and Fe3+ in $LuFe_2O_4$

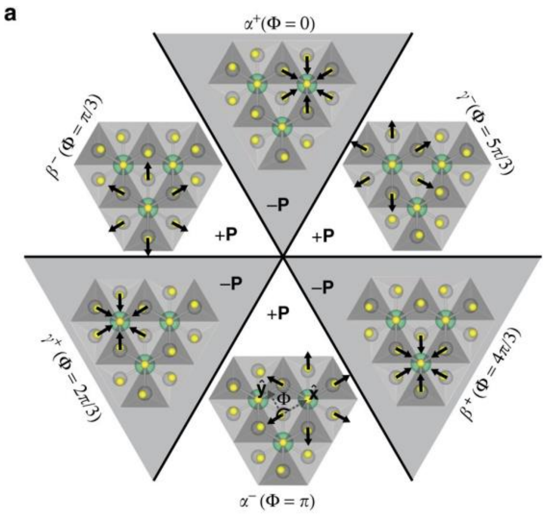
Magnetic order emerges from the iron lattice

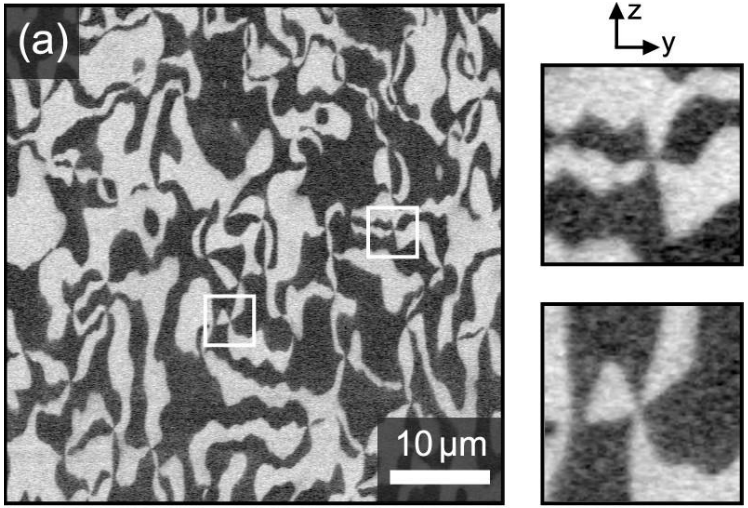
The classics
(Type I multiferroics)

Hexagonal $YMnO_3$

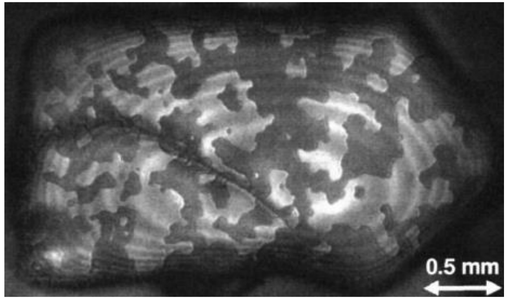
Geometric ferroelectrics, the electric polarization is driven by a rotational instability of the polyhedra and displacement of the A-site cation, $BaNiF_4$ or hexagonal $YMnO_3$,







T_N~ 77 K



Nat. Mater. 3, 164 (2004)
Appl. Phys. Lett. 97, 012904 (2010)
J. Appl. Phys. 83, 6560 (1998)
Nat. Commun. 5, 2998 (2014)

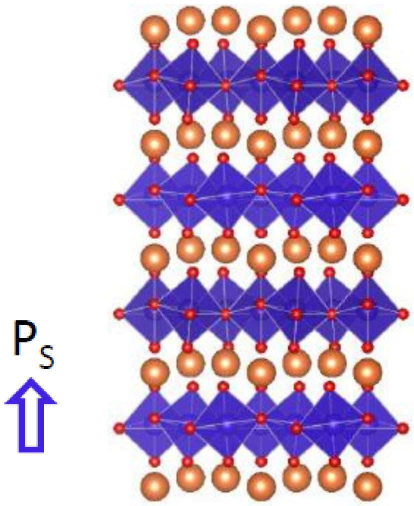
The classics
(Type I multiferroics)

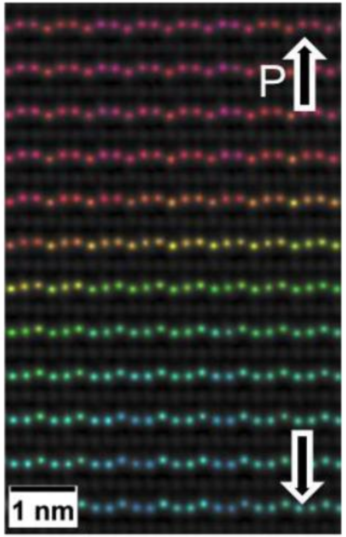
Hexagonal $YMnO_3$

Geometric ferroelectrics, the electric polarization is driven by a rotational instability of the polyhedra and displacement of the A-site cation, $BaNiF_4$ or hexagonal $YMnO_3$,

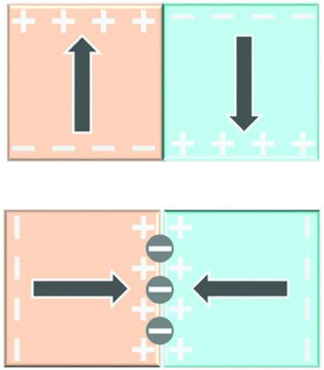
Geometric ferroelectric domain formation is not affected by electrostatics in contrast to “classical” or “proper” ferroelectrics like $BaTiO_3$

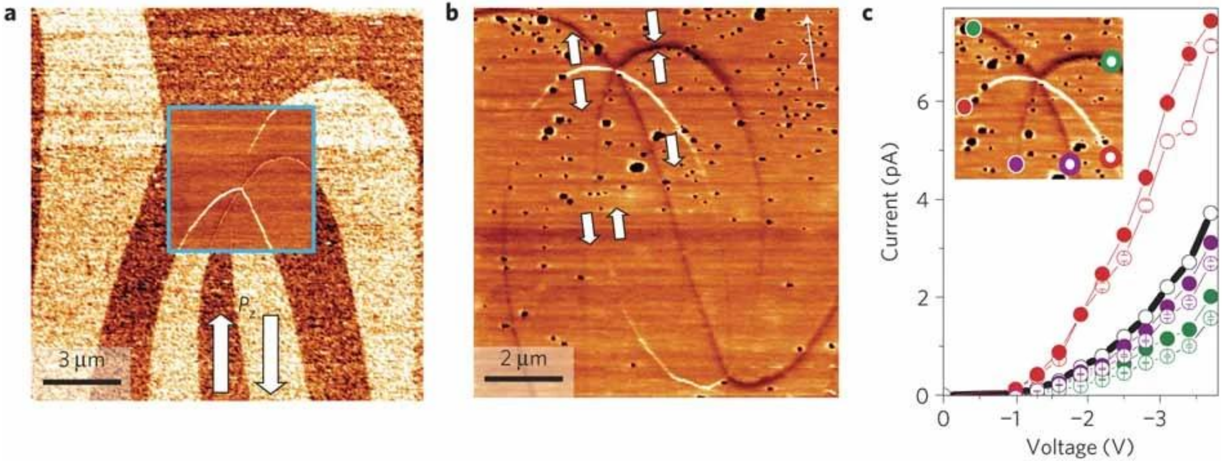
Head-to-head and tail-to-tail charged domain walls may form.





Phys. Sciences. Rev. 0067 (2019)
Nat. Mater. 3, 164 (2004)
Microscopy and Microanalysis, 23, 1636 (2017)
Nat. Mater.11, 284–288 (2012)

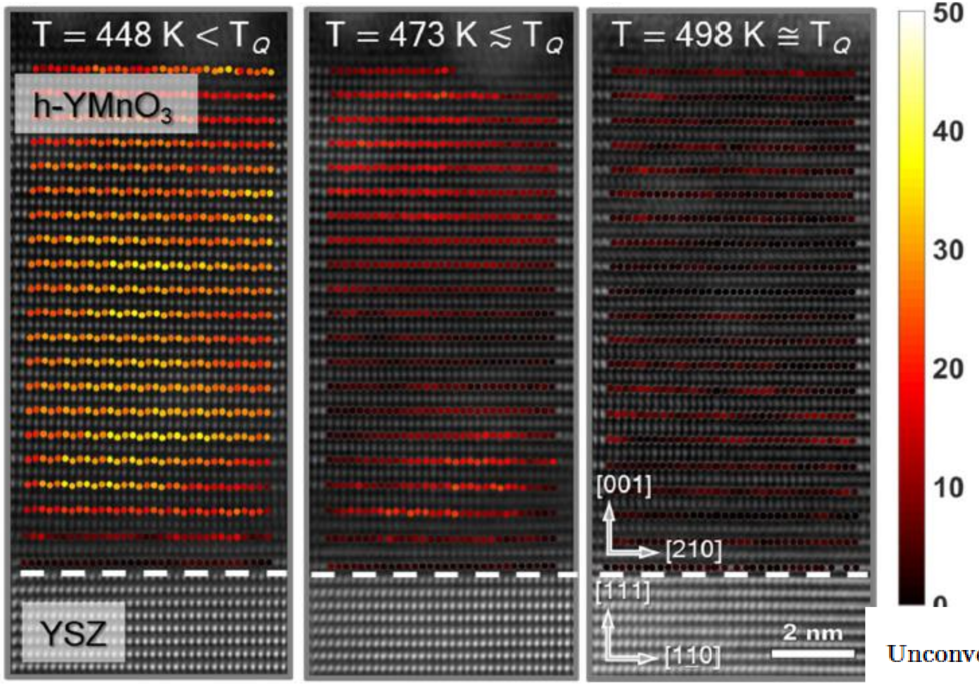




The classics

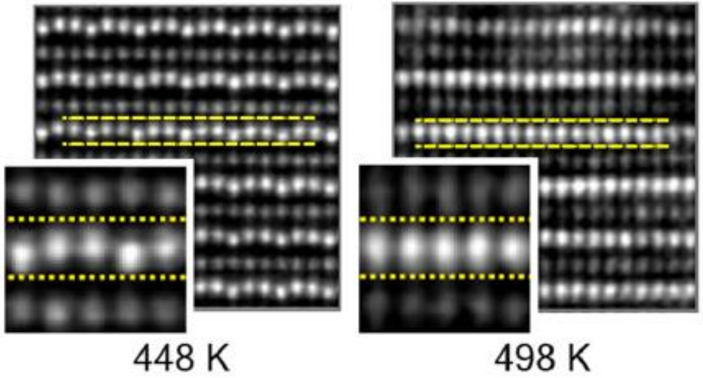
(Type I multiferroics)

Hexagonal $YMnO_3$



Nat. Commun. 10, 5591 (2019)

Polarization suppression and order-disorder phase transition in real space

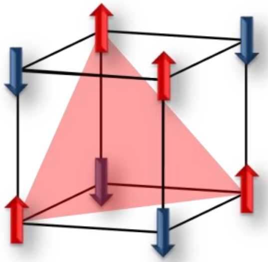


Unconventional order-disorder phase transition in improper ferroelectric hexagonal manganites

Sandra H. Skjærvø,[1,*] Quintin N. Meier,[2] Mikhail Feygenson,[3,4] Nicola A. Spaldin,[2] Simon J. L. Billinge,[5,6] Emil S. Bozin,[5] and Sverre M. Selbach[1,†]

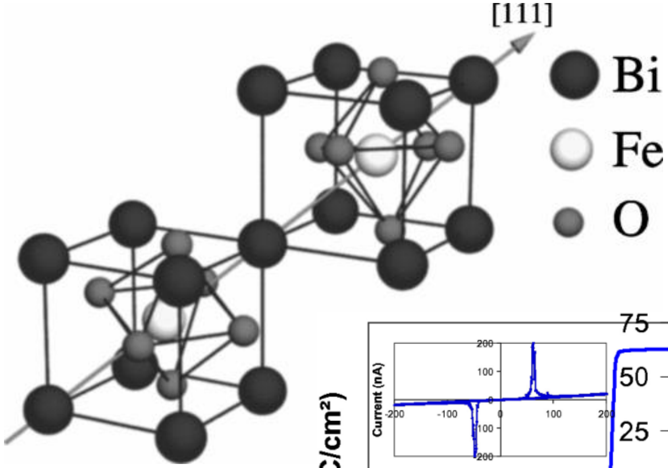
The classics
(Type I multiferroics)

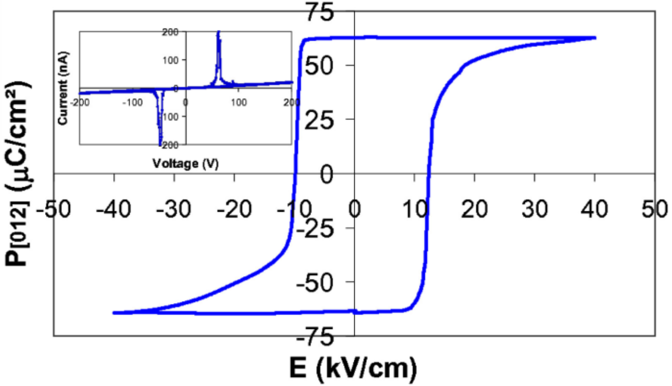
G-type antiferromagnetic order



$BiFeO_3$

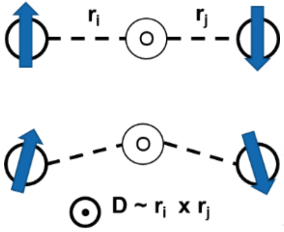
Lone pair stereochemical activity at the A-site, spontaneous electric polarization due to the displacement $6s^2$ lone pairs in $BiFeO_3$ T_c ~ 1100 K

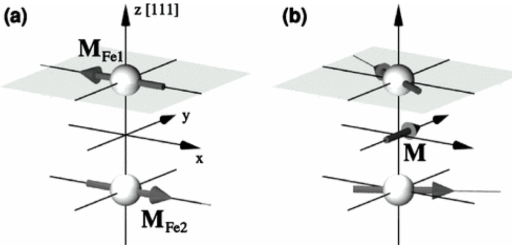




$$H_{DM} = \boldsymbol{D} \cdot \boldsymbol{S}_i \times \boldsymbol{S}_j,$$

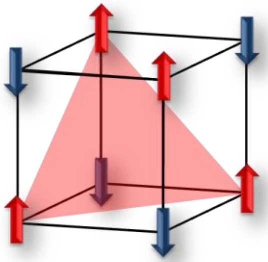
Dzyaloshinskii-Moriya interaction
A non centrosymmetric environment promotes antisymmetric exchange, non collinear $S_{i,j}$





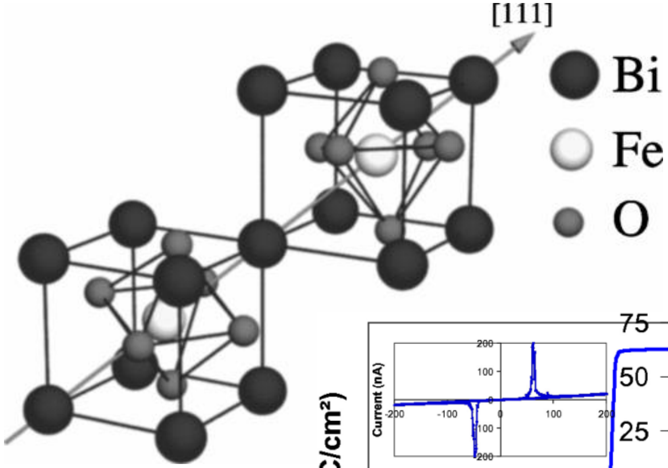
The classics
(Type I multiferroics)

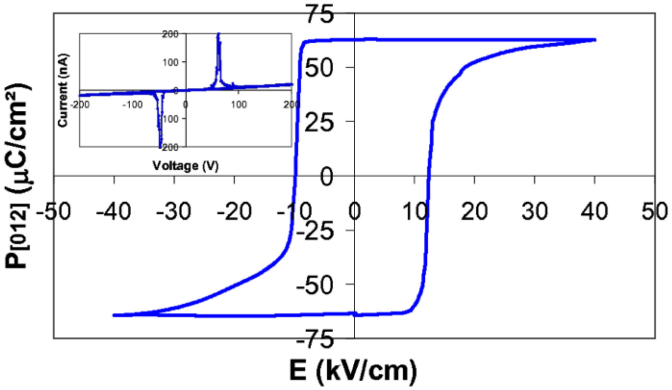
G-type antiferromagnetic order



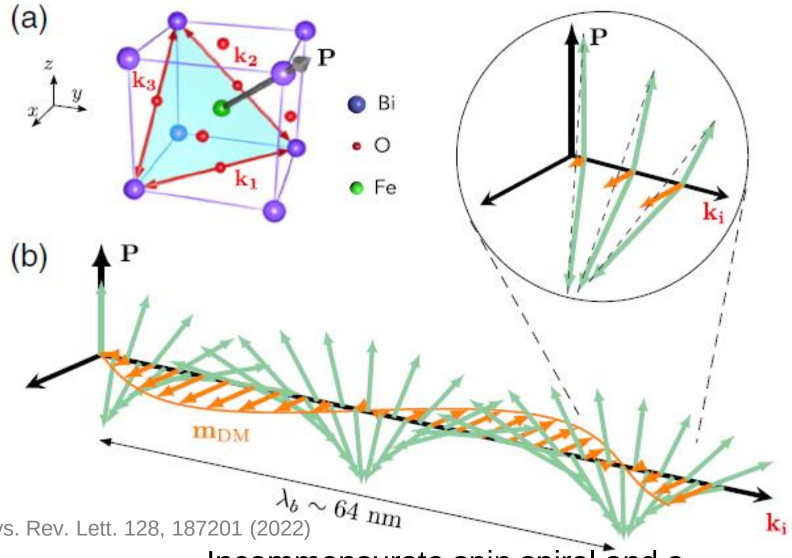
$BiFeO_3$

Lone pair stereochemical activity at the A-site, spontaneous electric polarization due to the displacement $6s^2$ lone pairs in $BiFeO_3$ T_c ~ 1100 K





Second DMI



Phys. Rev. Lett. 128, 187201 (2022)

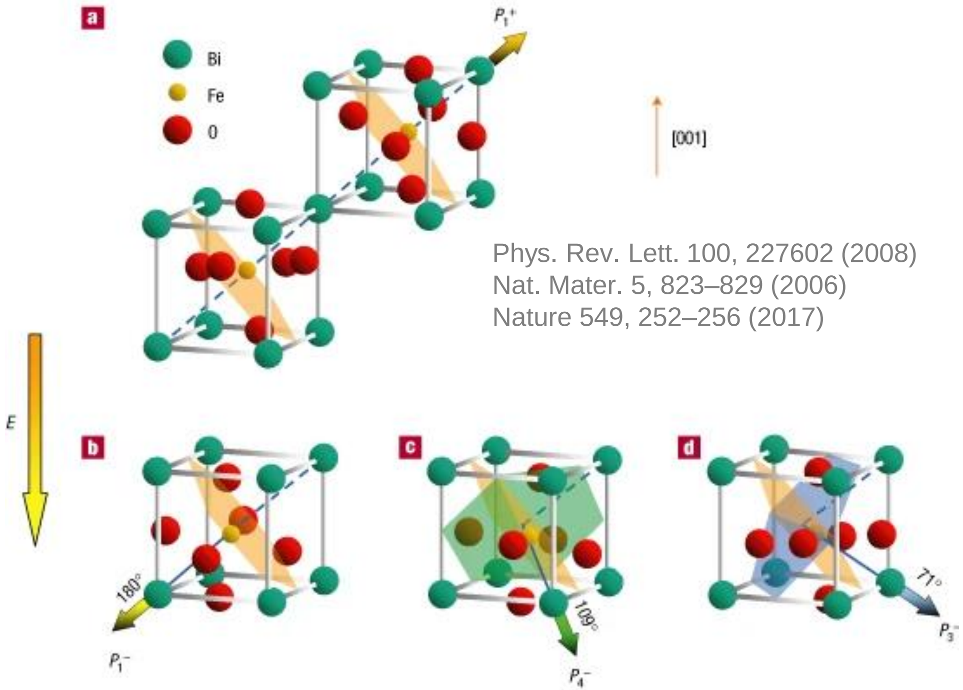
Incommensurate spin spiral and a modulation of uncompensated moment

The classics

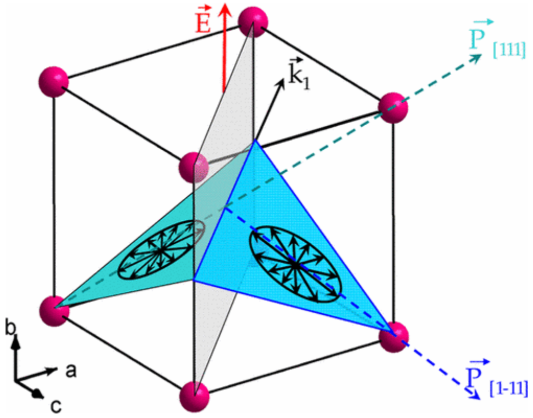
(Type I multiferroics)

$BiFeO_3$

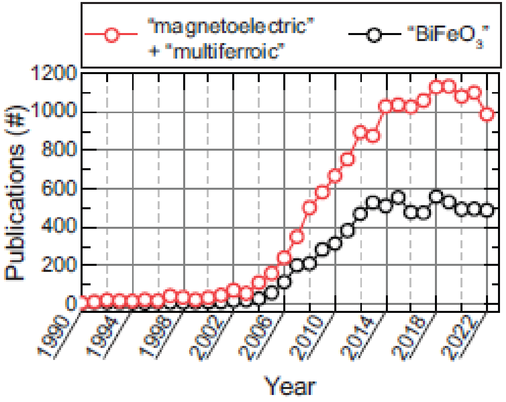
Lone pair stereochemical activity at the A-site, spontaneous electric polarization due to the displacement $6s^2$ lone pairs in $BiFeO_3$ $T_c \sim 1100$ K



Phys. Rev. Lett. 100, 227602 (2008)
Nat. Mater. 5, 823–829 (2006)
Nature 549, 252–256 (2017)

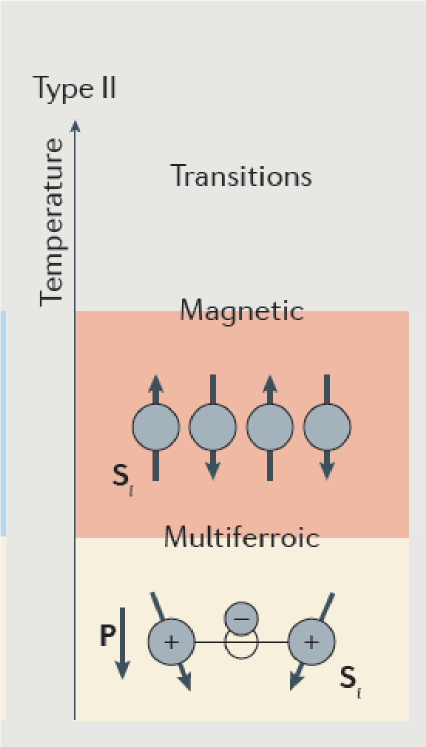


At room temperature

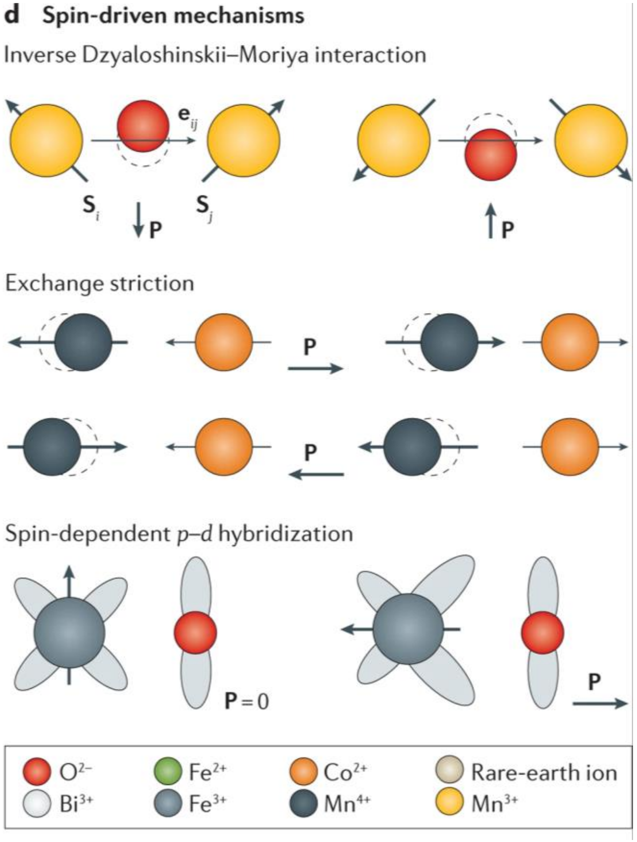


M. Mueller PhD thesis, ETH Zurich (2023)

Ferroelectricity induced by the magnetic order (Type II multiferroics)

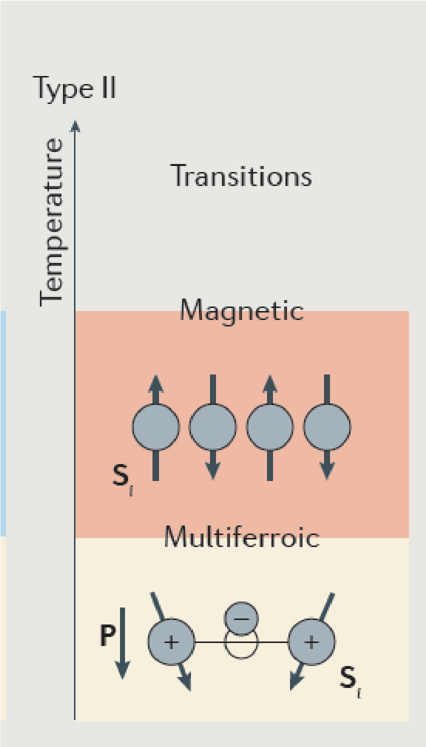


Nat. Rev. Mater. 1, 2016

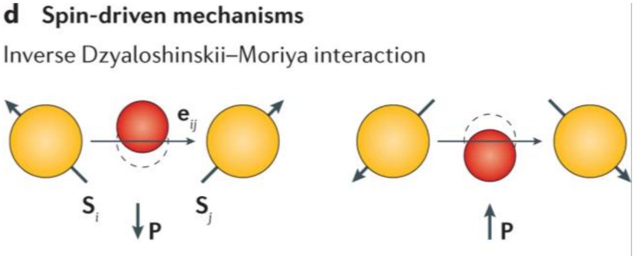


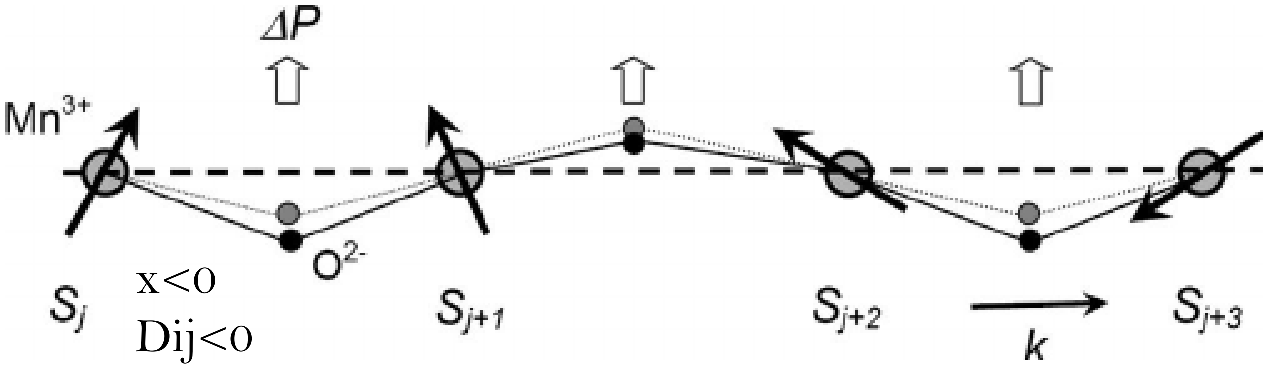
Ferroelectricity induced by the magnetic order (Type II multiferroics)

In the case of a frustrated magnetically ordered system, a canted spin ordering leads to an atomic displacement via the inverse Dzyaloshinskii-Moriya interaction, orthorhombic $TbMnO_3$ $T_c \sim 27$ K



Nat. Rev. Mater. 1, 2016



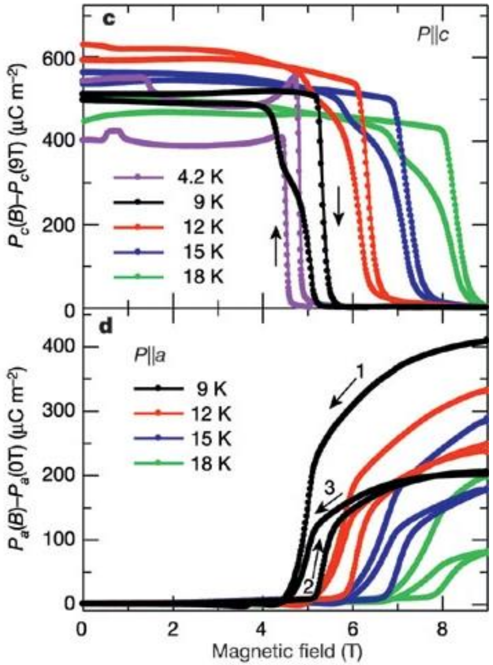


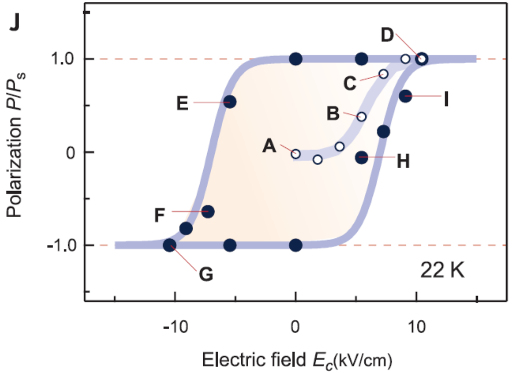
$S_i \times S_j$, is fixed and D_{ij} is proportional to x x r_{ij}

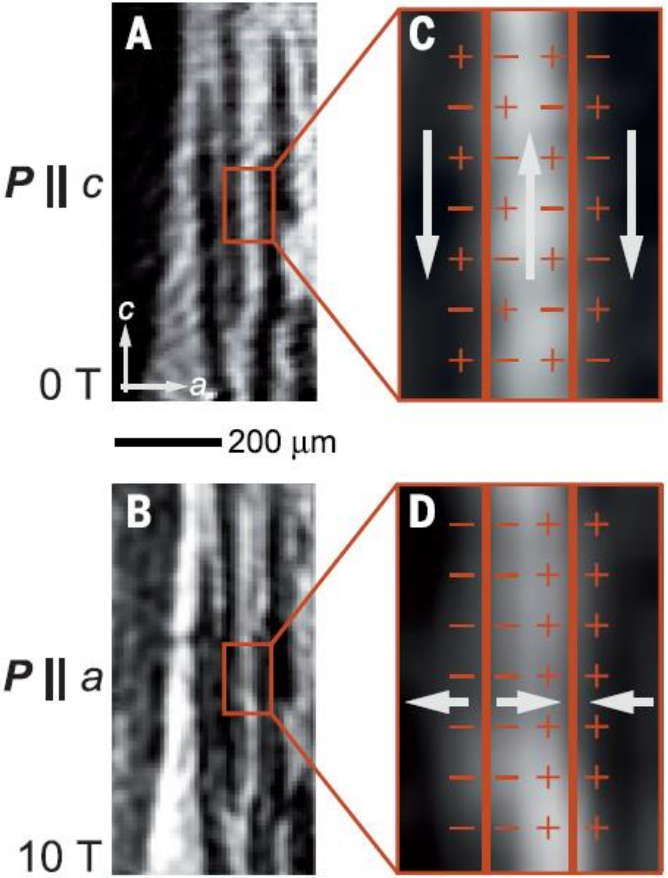
Ferroelectricity induced by the magnetic order (Type II multiferroics)

$TbMnO_3$

Field-induced domain dynamics of $TbMnO_3$
first-order spin-flop transition

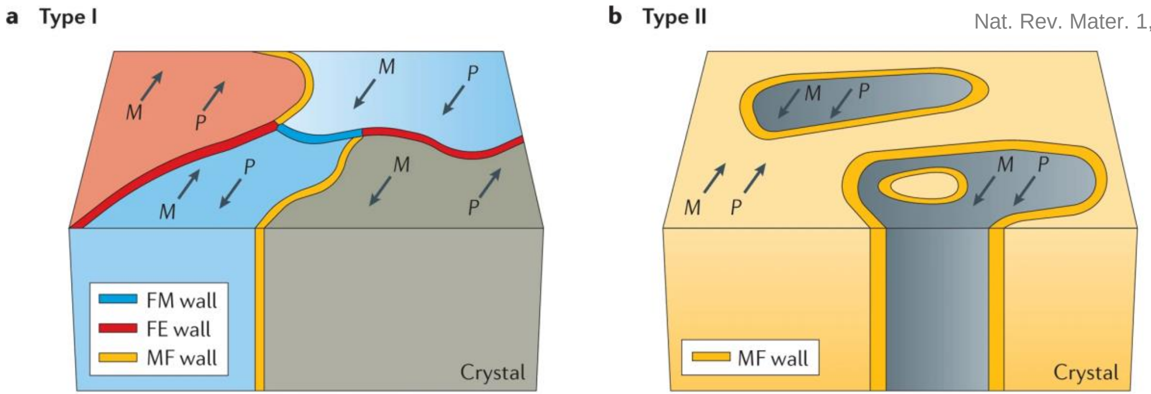






Type I and Type II multiferroics

A domain correlation naturally appears in Type II (magnetoelectric coupling expected due to the common origin of magnetic order and ferroelectricity)
However Low P_s ~ 0.1 µC/cm^2, Low T_c 20 K

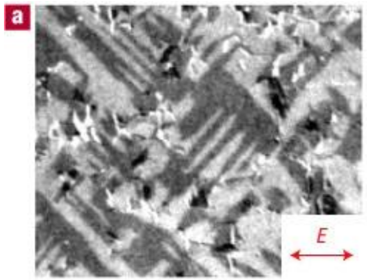


Nat. Rev. Mater. 1, 2016

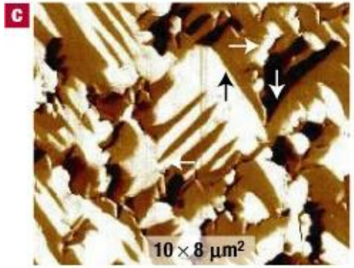
A ferroelectric and antiferromagnetic domain coupling can appear in type I multiferroics

BiFeO$_3$ bulk

Antiferromagnetic domains

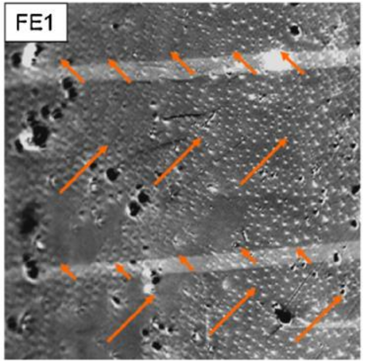


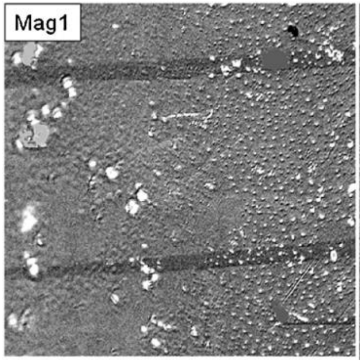
Ferroelectric domains



BiFeO$_3$ films

Nat. Mater. 5, 823–829 (2006)
Phys. Rev. Lett. 103, 257601 (2009)



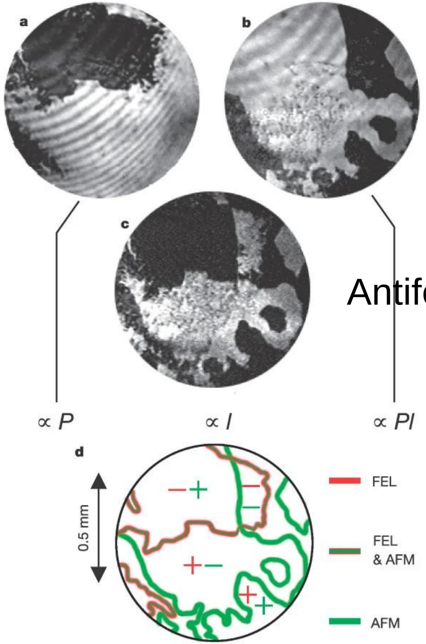


Type I and Type II multiferroics

A ferroelectric and antiferromagnetic domain coupling can appear in type I multiferroics

Ferroelectric domains

bulk $YMnO_3$



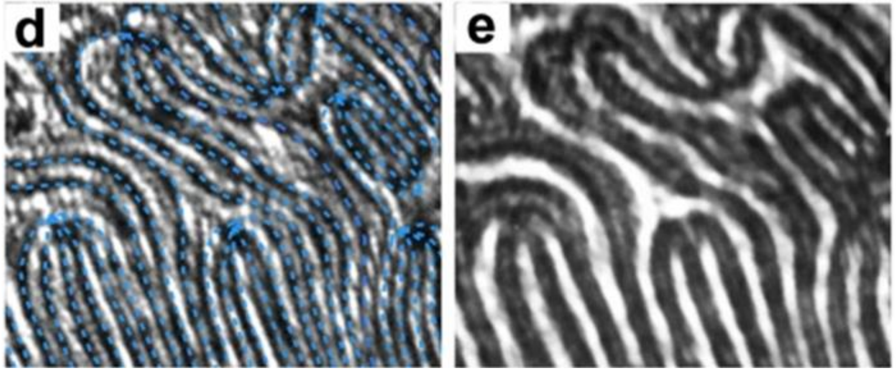
Antiferromagnetic domains

Nature 419, 818 (2002)

The lattice trimerization impacts the spin ordering and the ferroelectric domain architecture

Ferroelectric domains

Antiferromagnetic domains



Nat Commun 12, 3093 (2021)

bulk $ErMnO_3$

D MATL.

Multiferroics

- Why are there so few ferromagnetic-ferroelectric materials?
- Mechanisms promoting the coexistence of magnetic and electric long-range orders (type I and type II multiferroics)

Electric-field control of ferromagnetism using multiferroics

- Artificial / synthetic multiferroics
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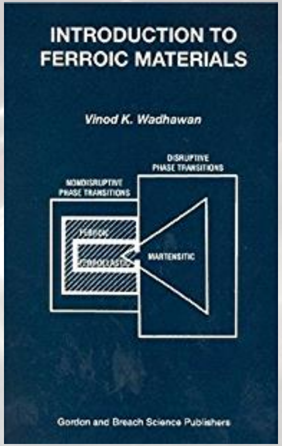
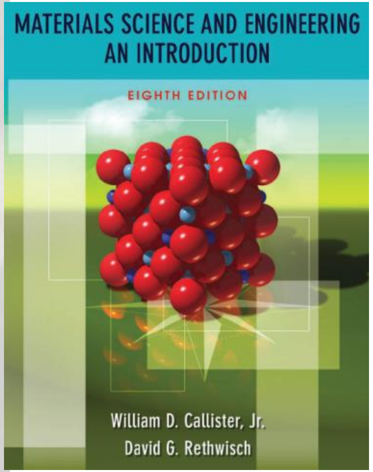
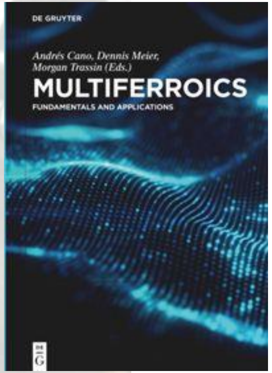
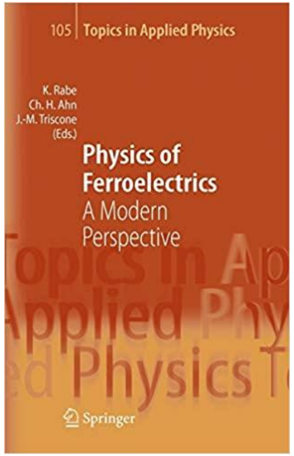
On the way to the ultimate goal

- Characterization of multiple order parameters
- Beyond the low energy control of magnetization





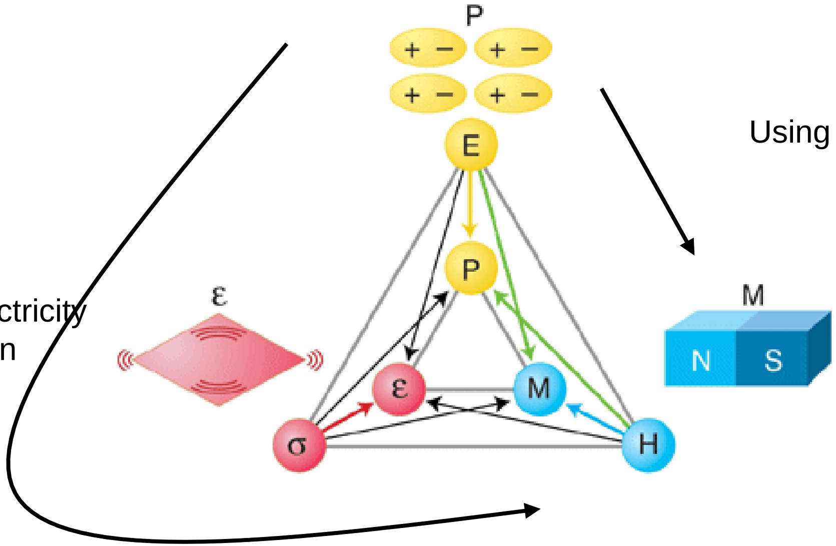




Electric field control of ferromagnetism

Two ways to act on magnetic order using multiferroics magnetoelectrics

Combining piezoelectricity with magnetostriction

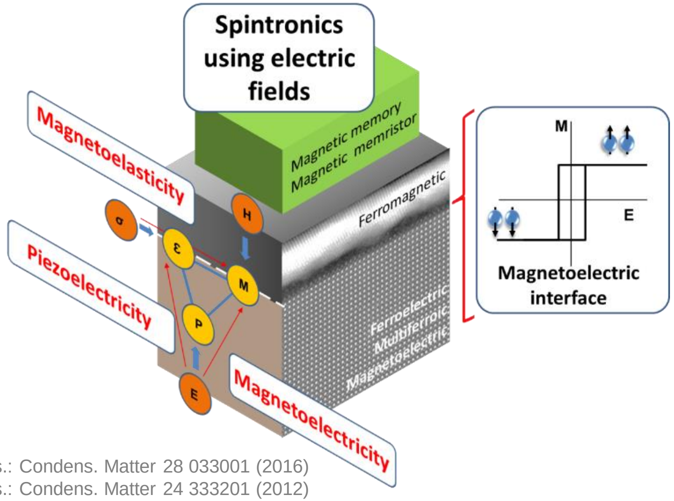


Science 309, 391 (2005)

Using the magnetoelectric coupling

Symmetry restriction
No net M (Type I)
Low temperature (Type II)

In the so-called artificial multiferroic heterostructures, each layer brings its own ferroic contribution

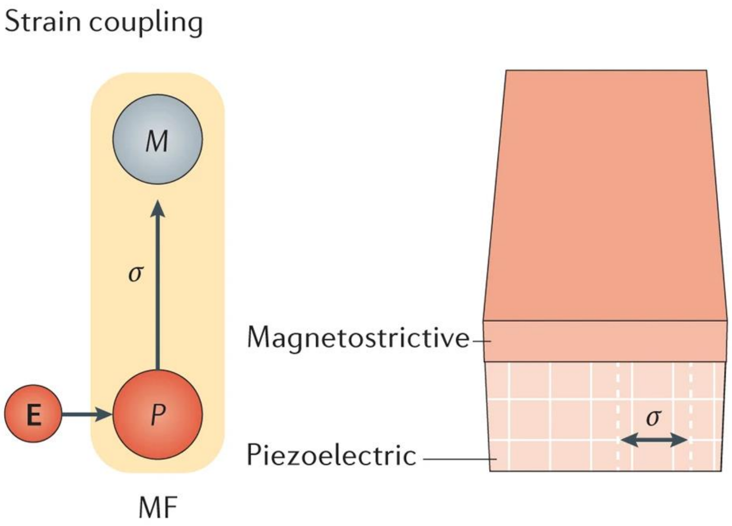


J. Phys.: Condens. Matter 28 033001 (2016)
J. Phys.: Condens. Matter 24 333201 (2012)

Electric field control of ferromagnetism

Acting on magnetic order using artificial multiferroics heterostructures

Nat. Rev. Mater. 1, 2016



Applying a mechanical strain to a ferromagnetic material induces a magnetoelastic anisotropy, also know as inverse magnetostriction effect.

$$E_{me} = -K_{me} \sin^2 \phi$$

For a sufficient strain value ε_c, the magnetoelastic energy dominates over the magnetocrystalline anisotropy

$$\varepsilon_c = \frac{|K_1|}{|B_1|}$$

Ferromagnet

magnetoelastic anistropy, high magnetostrictive coefficient
Ni, Co50Fe50, Terfenol-D, etc..

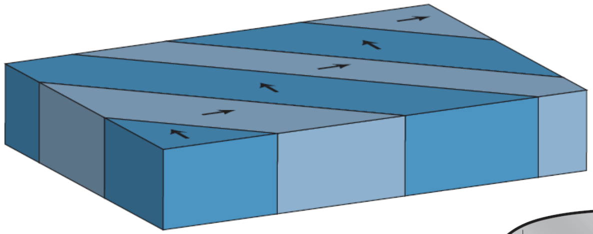
Ferroelectric/Piezoelectric

Switching events need to involve a ferroelestic event, no equivalent initial and final strain state

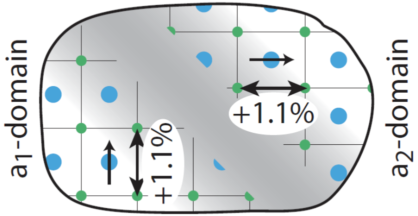
BaTiO3, PZT, PMNPT

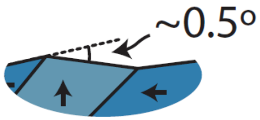
Electric field control of ferromagnetism

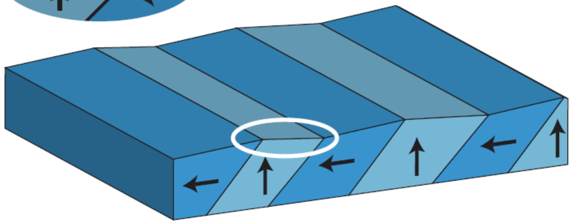
Acting on magnetic order using artificial multiferroics heterostructures



As-grown 90° domains in-plane







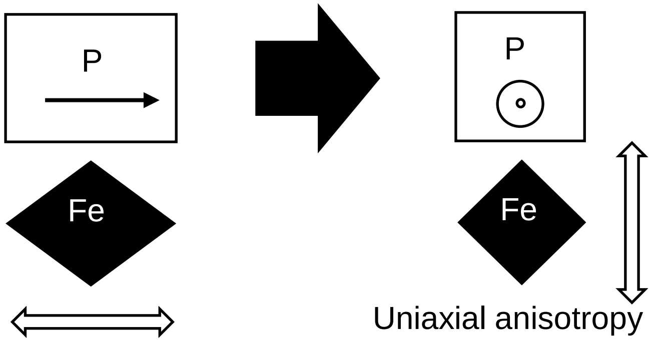
Voltage out-of-plane:
a- to c-domains

Lahtinen T H E, et al., 2012 Sci. Rep. 2 258
Lahtinen T H E, et al., 2011 Adv. Mater. 23 3187
Franke K J A, et al., S 2014 Phys. Rev. Lett. 112 017201
Franke K J A, et al., 2015 Phys. Rev. X 5 011010
Lathinen Thesis

Fe on $BaTiO_3$ bulk

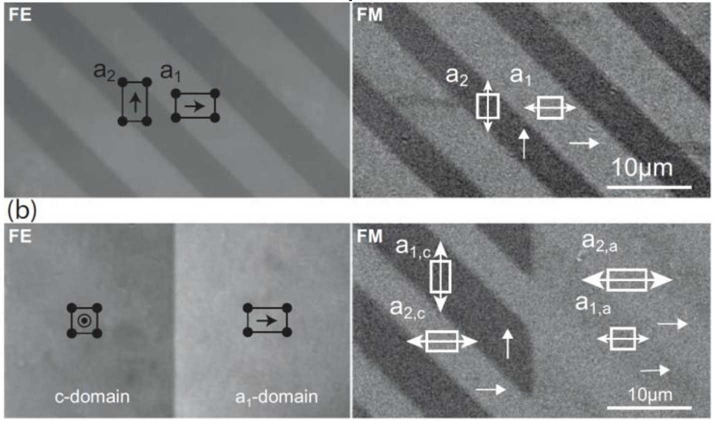
a-domain

c-domain



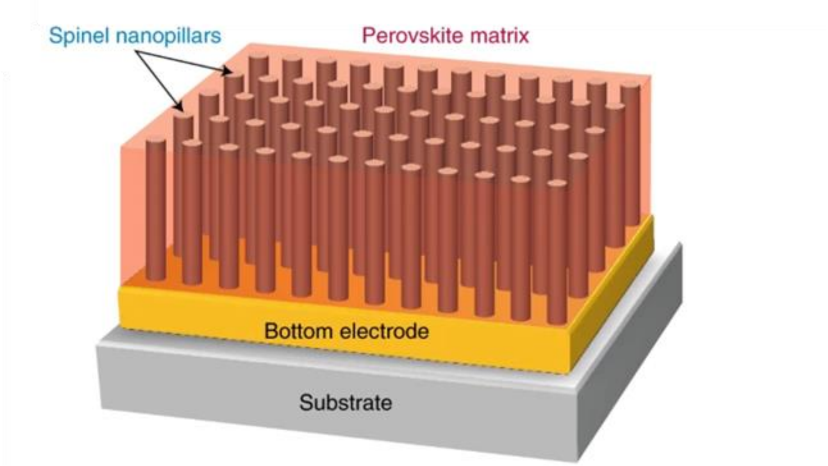
Uniaxial anisotropy

1.6% compressive strain

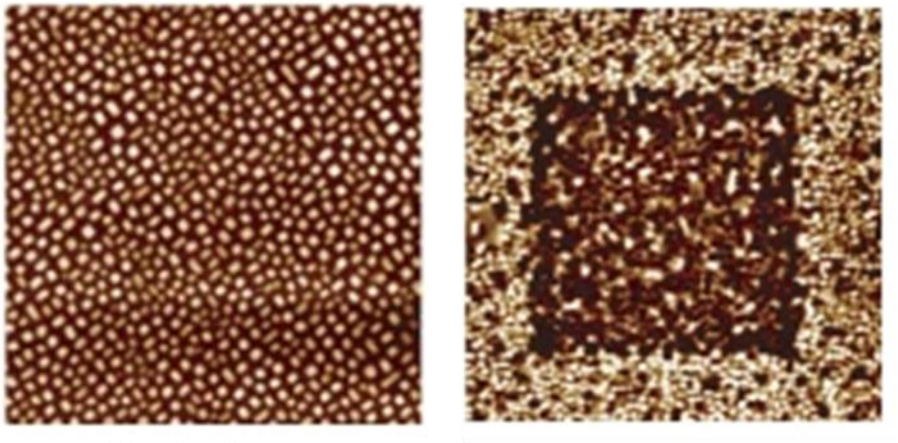


Electric field control of ferromagnetism

Acting on magnetic order using artificial multiferroics heterostructures



$BiFeO_3$ and $CoFe_2O_4$



Nano Lett. 2005, 5, 9, 1793
Nat. Mater. 6, 21–29 (2007)

JOURNAL OF APPLIED PHYSICS VOLUME 87, NUMBER 9

Novel magnetostrictive memory device

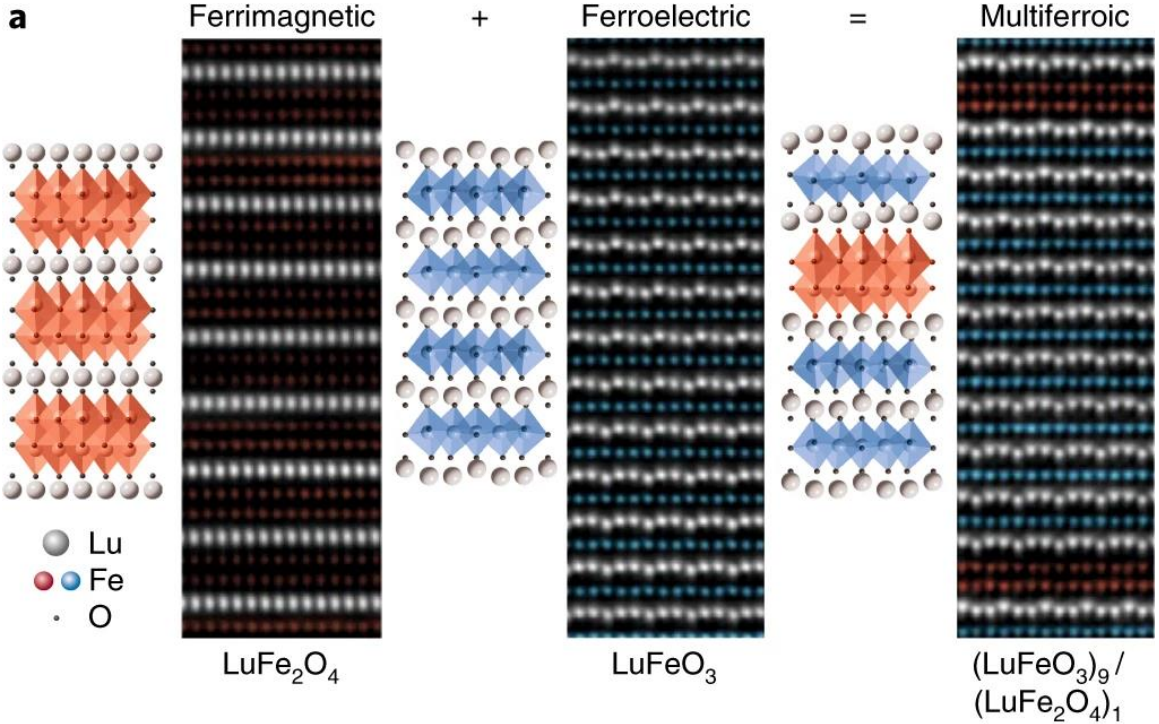
V. Novosad, Y. Otani, A. Ohsawa, S. G. Kim, K. Fukamichi, J. Koike, and K. Maruyama
Department of Materials Science, Graduate School of Engineering, Tohoku University, Aoba-yama 02, Sendai 980-8579, Japan

O. Kitakami and Y. Shimada
Institute for Scientific Measurements, Tohoku University, Sendai 980-8577, Japan

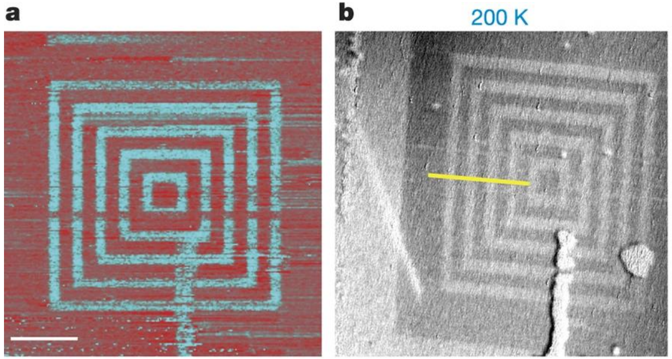
Electric field control of ferromagnetism

Synthetic multiferroics

$LuFeO_3$ and $LuFe_2O_4$



Growth of oxides thin films with atomic precision. Interface driven magnetoelectric properties in superlattices

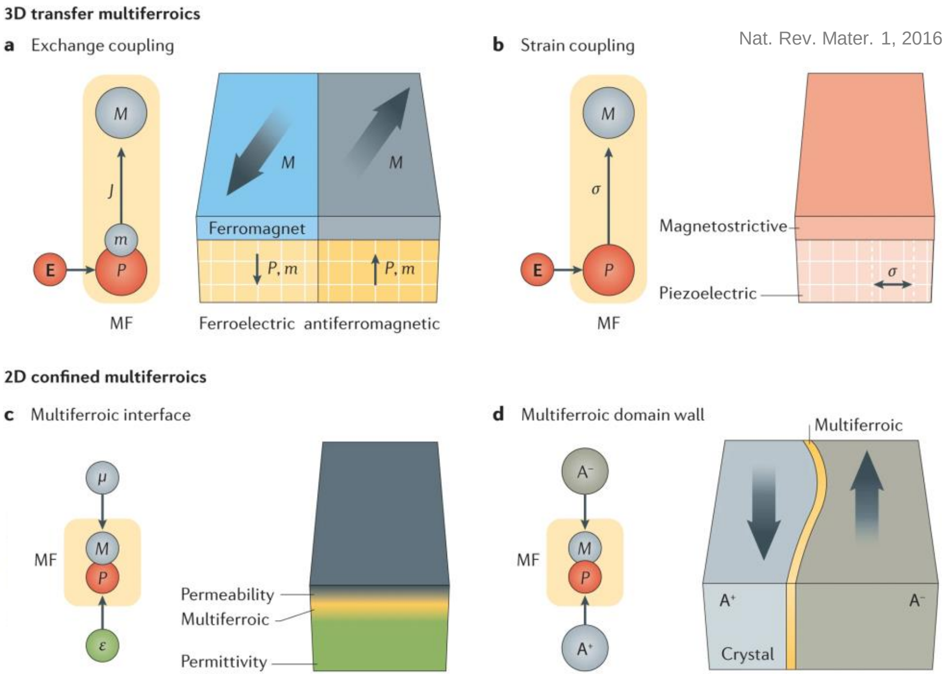


Nature 537, 523 (2016)

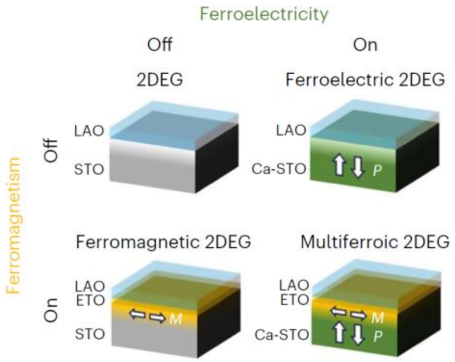
Electric field control of ferromagnetism

Acting on magnetic order using artificial multiferroics heterostructures

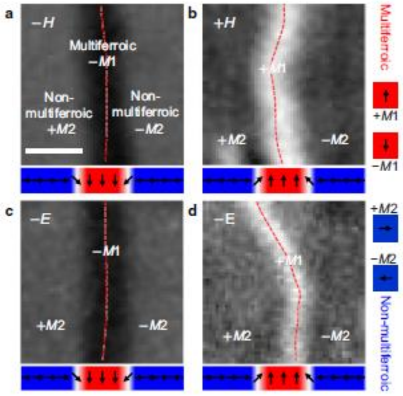
Nat. Rev. Mater. 1, 2016



Nat. Mater. 10, 753 (2011)
Nat. Phys. 19, 823 (2023)



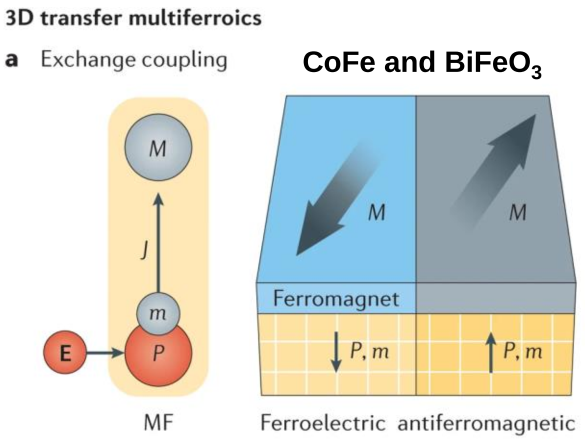
Nat. Commun. 12, 2755 (2021)



Electric field control of ferromagnetism

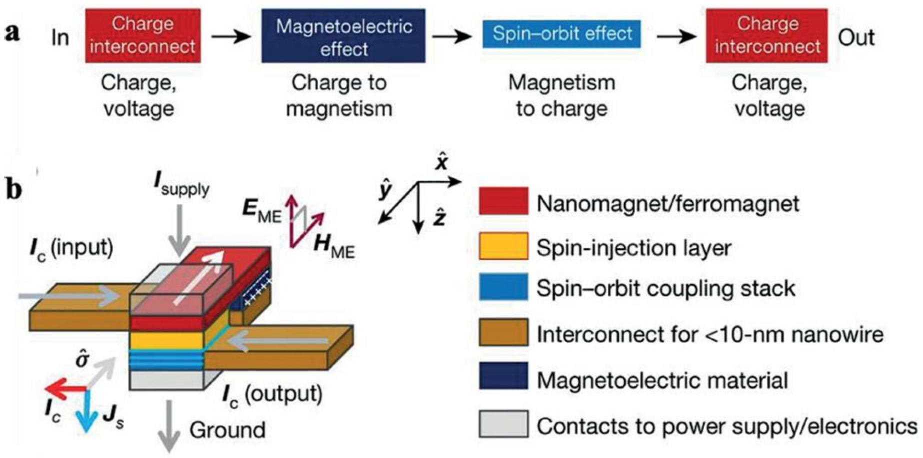
Acting on magnetic order using artificial multiferroics heterostructures

CoFe and $BiFeO_3$



Nat. Rev. Mater. 1, 2016

Integration into magnetoelectric spin orbit devices



Nat. Phys. 14, 338 (2018)
Nature 565, 35 (2019)
arXiv:2302.12162

Selected for a Viewpoint in *Physics*
PRL **107**, 217202 (2011) PHYSICAL REVIEW LETTERS week ending 18 NOVEMBER 2011

Electric-Field-Induced Magnetization Reversal in a Ferromagnet-Multiferroic Heterostructure

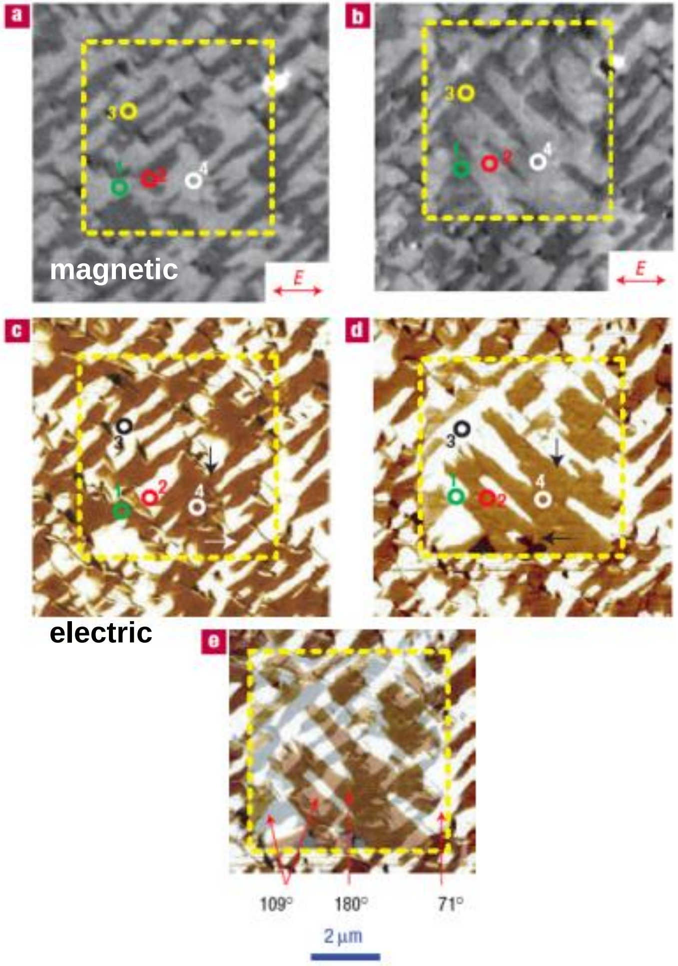
Electric field control of ferromagnetism

Nature Materials 5, 823 (2006)

Electrical control of antiferromagnetic domains in multiferroic $BiFeO_3$ films at room temperature

T. ZHAO[1*†], A. SCHOLL[2‡], F. ZAVALICHE[1‡], K. LEE[1], M. BARRY[1], A. DORAN[2], M. P. CRUZ[1,3], Y. H. CHU[1], C. EDERER[4], N. A. SPALDIN[4], R. R. DAS[5], D. M. KIM[5], S. H. BAEK[5], C. B. EOM[5] AND R. RAMESH[1†]

No net magnetization

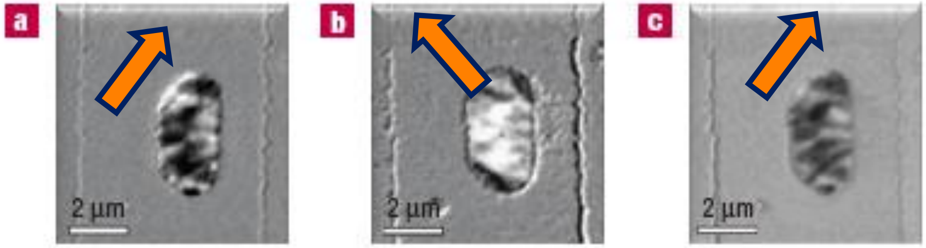


A Co dot is exchange coupled to the antiferromagnetic order in $BiFeO_3$

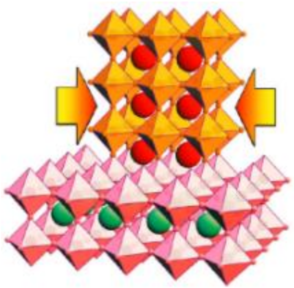
Electric-field control of local ferromagnetism using a magnetoelectric multiferroic

YING-HAO CHU[1,2,3*], LANE W. MARTIN[1,3*†], MIKEL B. HOLCOMB[2,3], MARTIN GAJEK[2], SHU-JEN HAN[4], QING HE[2], NINA BALKE[2], CHAN-HO YANG[2], DONKOUN LEE[4], WEI HU[4], QIAN ZHAN[1,2], PEI-LING YANG[1,2], ARANTXA FRAILE-RODRÍGUEZ[5], ANDREAS SCHOLL[6], SHAN X. WANG[4] AND R. RAMESH[1,2,3]

Nature Materials 7, 478 (2008)



Electric field control of ferromagnetism

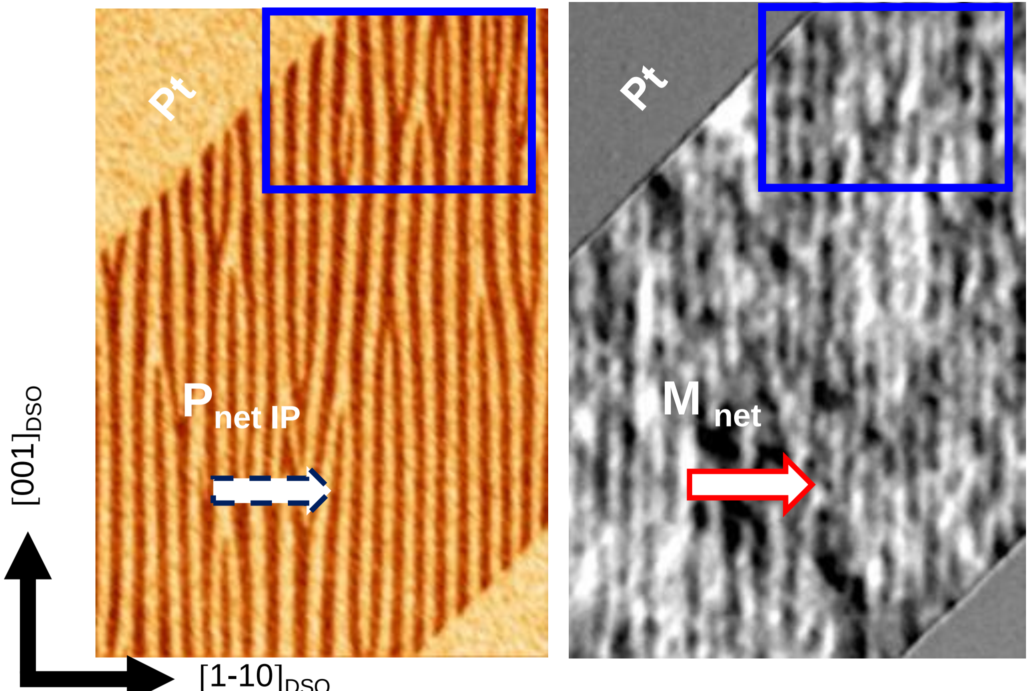


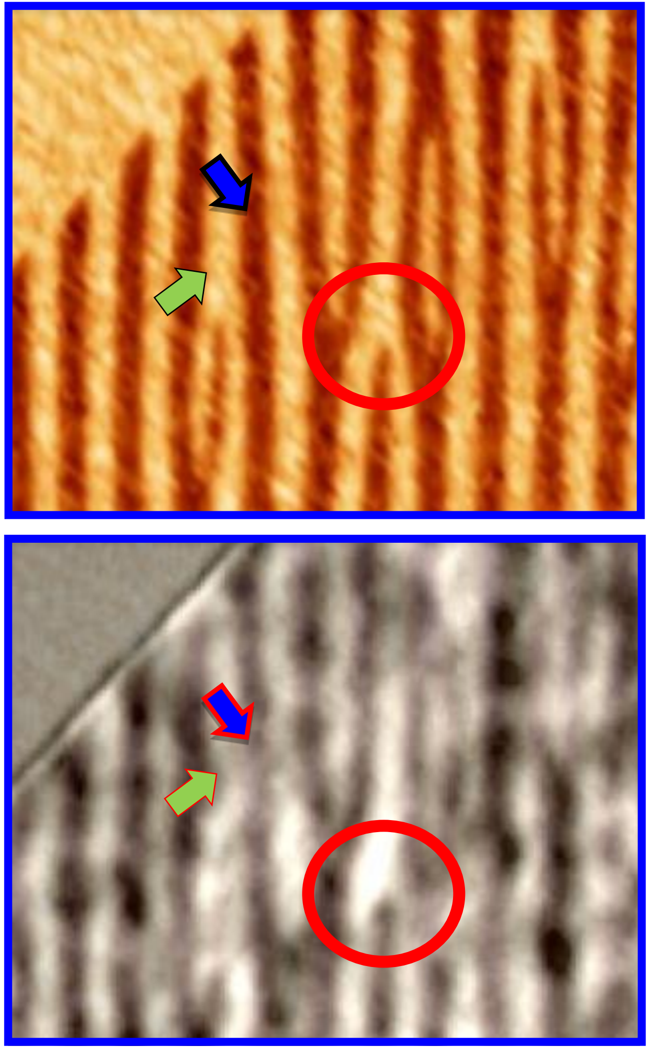
Epitaxial growth of $BiFeO_3$ thin films with well defined ferroelectric domain architecture.

Net in-plane polarization, Net in-plane magnetization

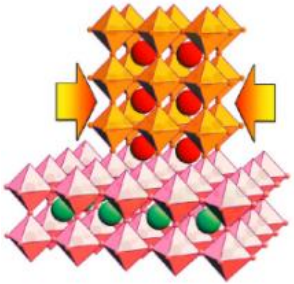
CoFe and $BiFeO_3$

PRL. 107, 217202 (2011)
PRB 87 134426 (2013)





Electric field control of ferromagnetism



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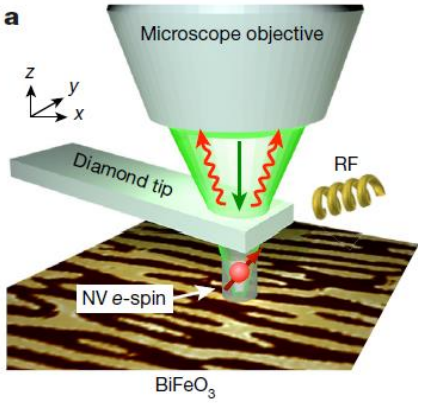
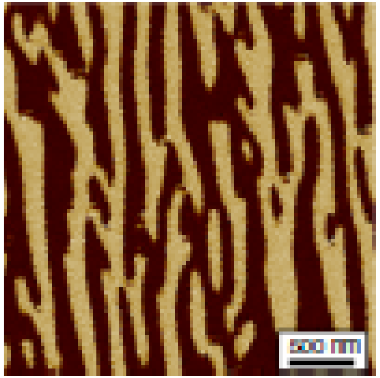
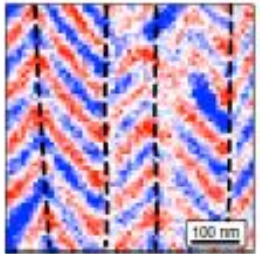
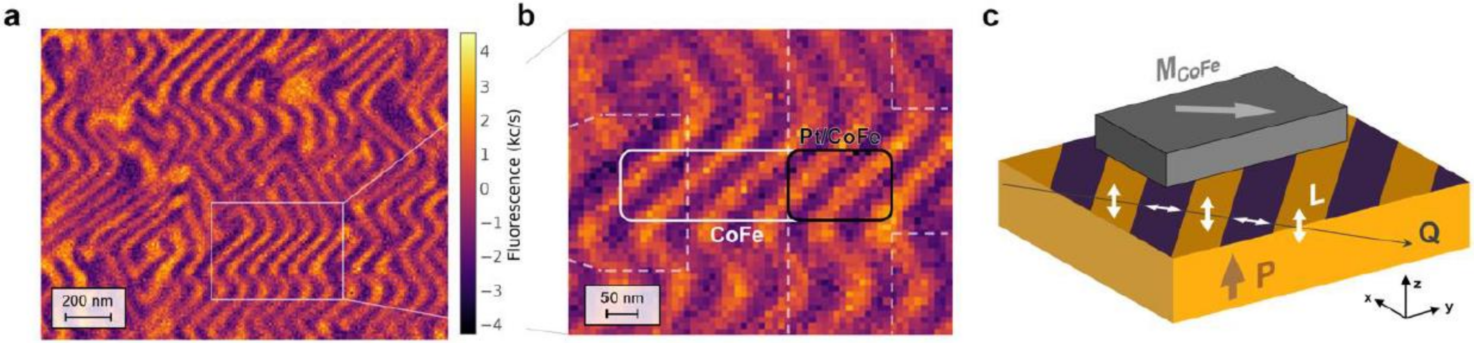




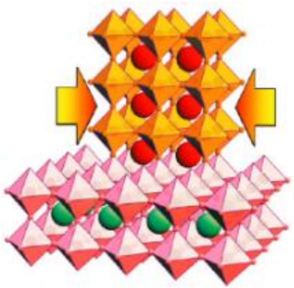


Direct mapping of uncompensated moment using single spin magnetometry

Nature 549, 252 (2017)
Nat. Commun 11, 1704 (2020)
arXiv:2302.12162



Electric field control of ferromagnetism



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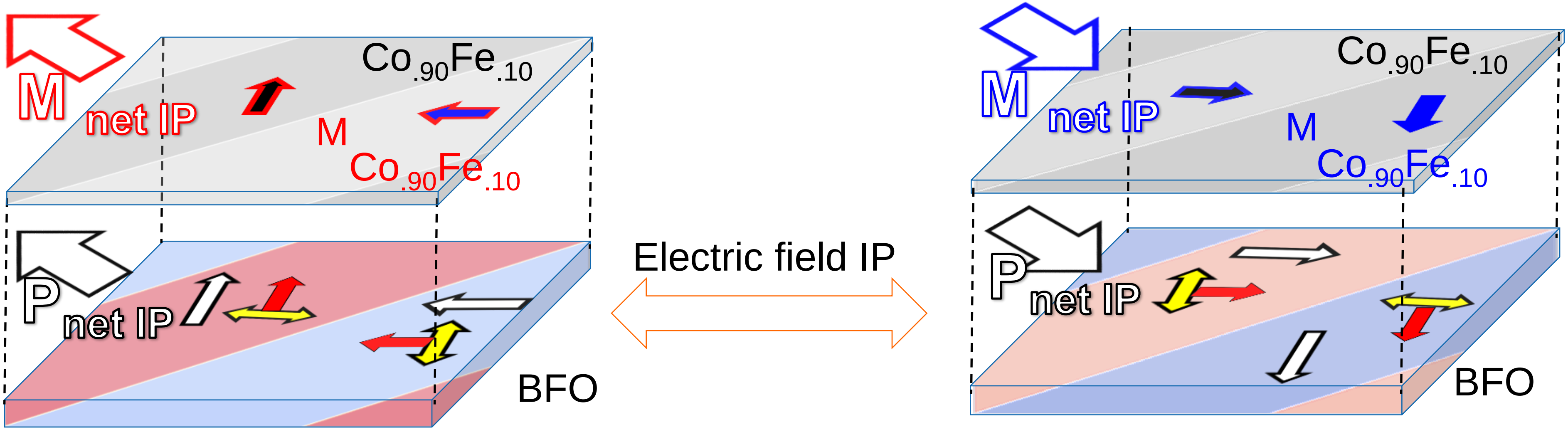
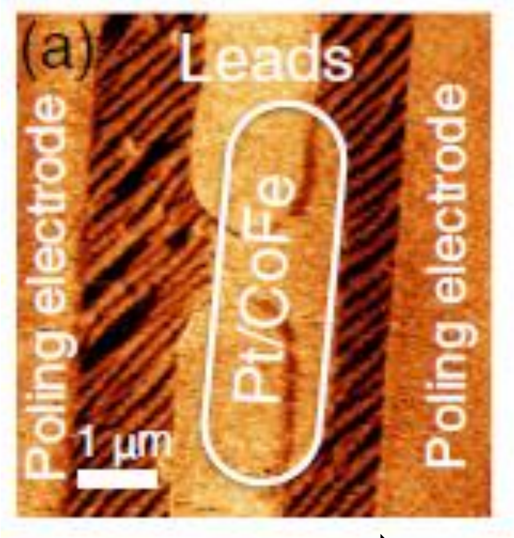


PRL. 107, 217202 (2011)
PRB 87 134426 (2013)

Electric field control of ferromagnetism

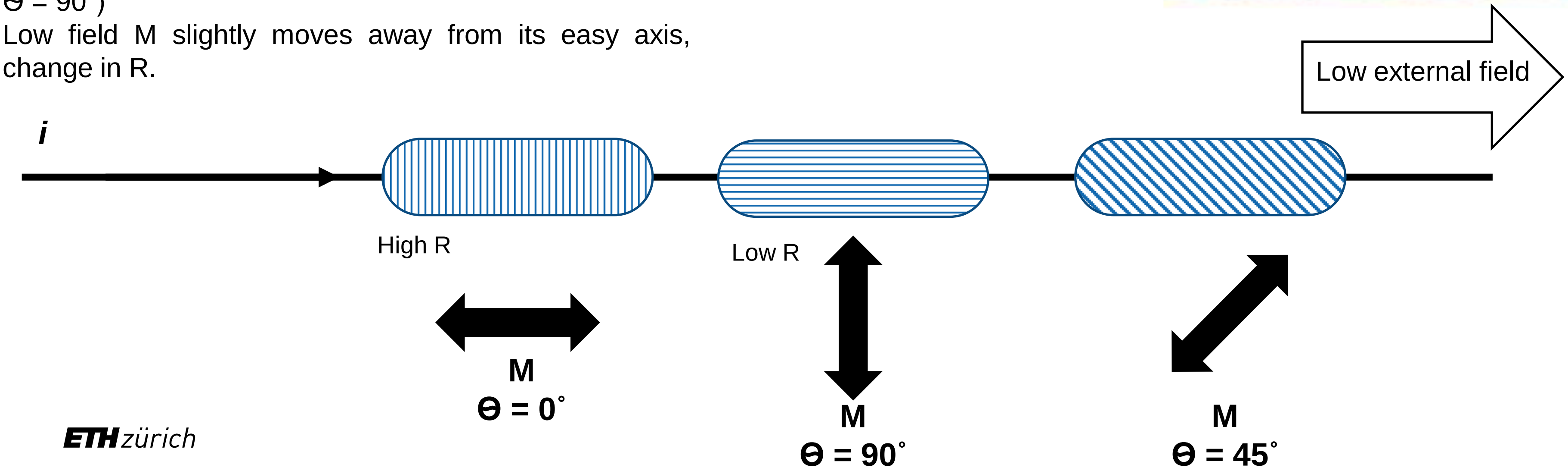


Probing electric field induced magnetization reversal

Using anisotropic magnetoresistance

$$R(\theta) = R_0 + (R_{\parallel} - R_0)\cos^2(\theta)$$

The magnetic layer is spin filtering the current mainly when the current is along its magnetization (R min when ϴ = 90˚)
Low field M slightly moves away from its easy axis, change in R.



ETH zürich

Electric field control of ferromagnetism

Probing electric field induced magnetization reversal

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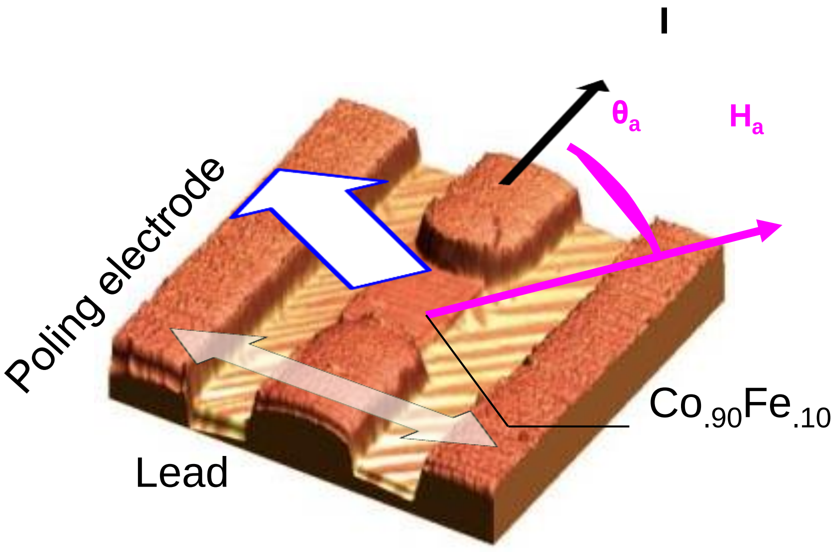
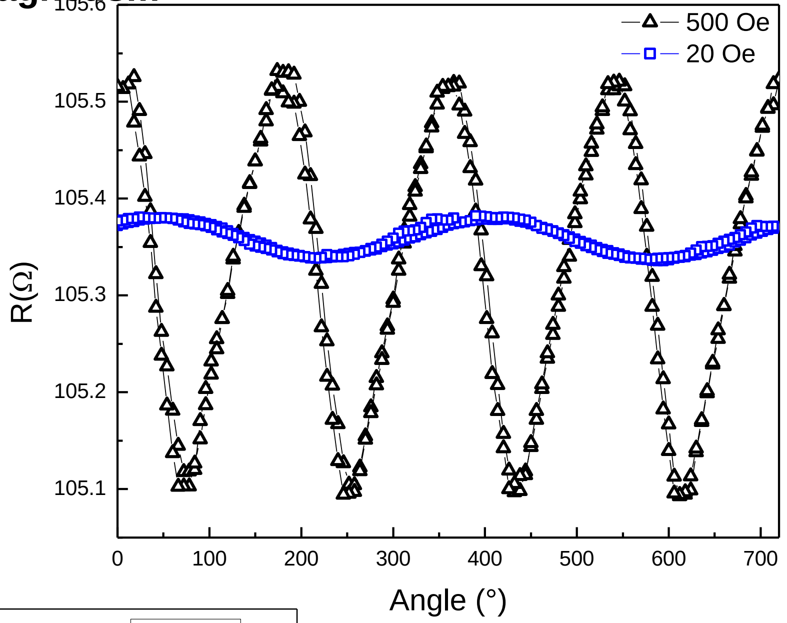
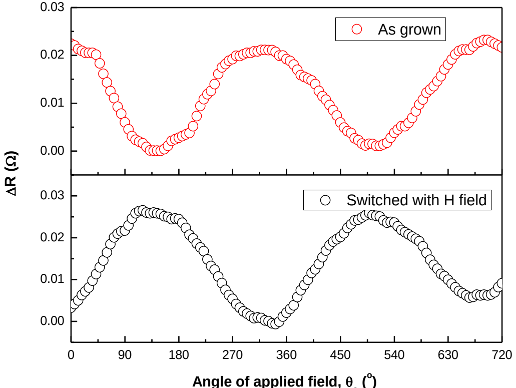
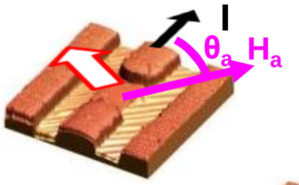
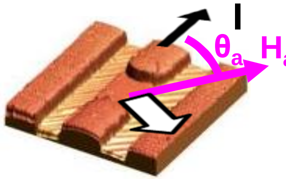


Magnetically induced reversal



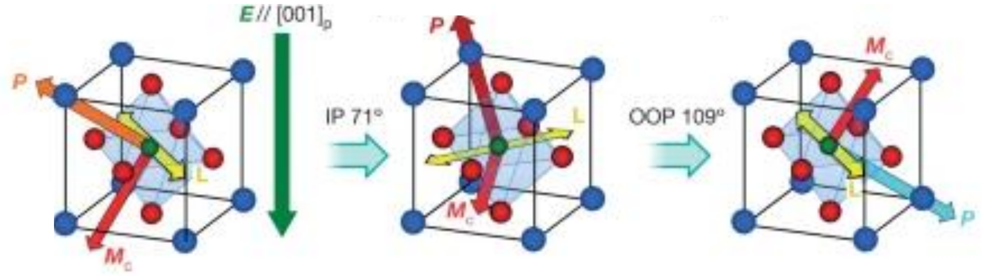


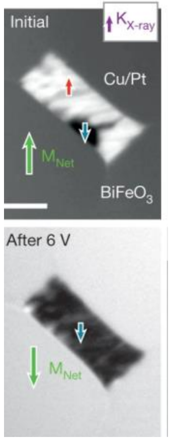


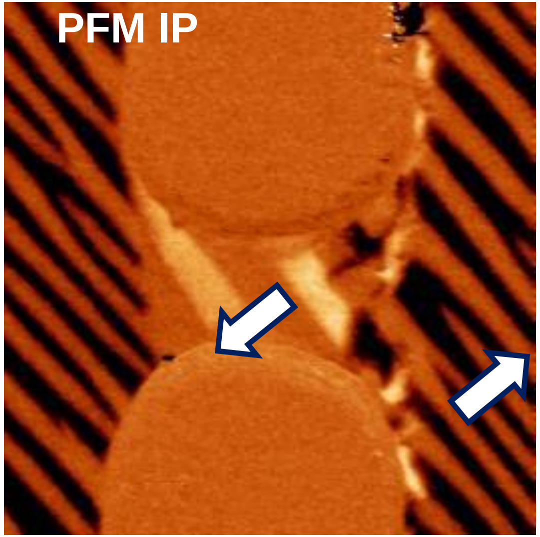


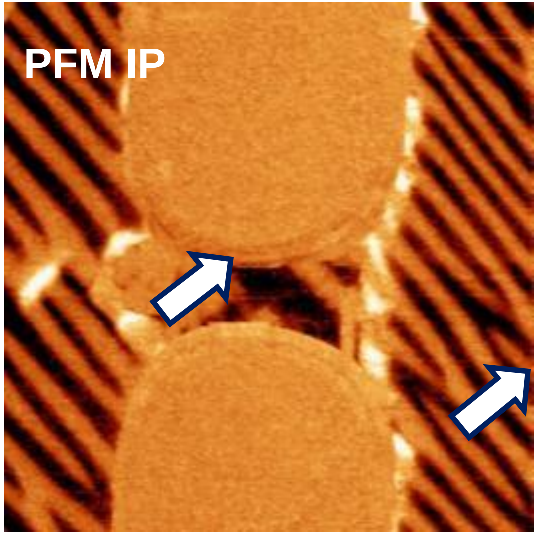
PRL. 107, 217202 (2011)
PRB 87 134426 (2013)

Electric field control of ferromagnetism

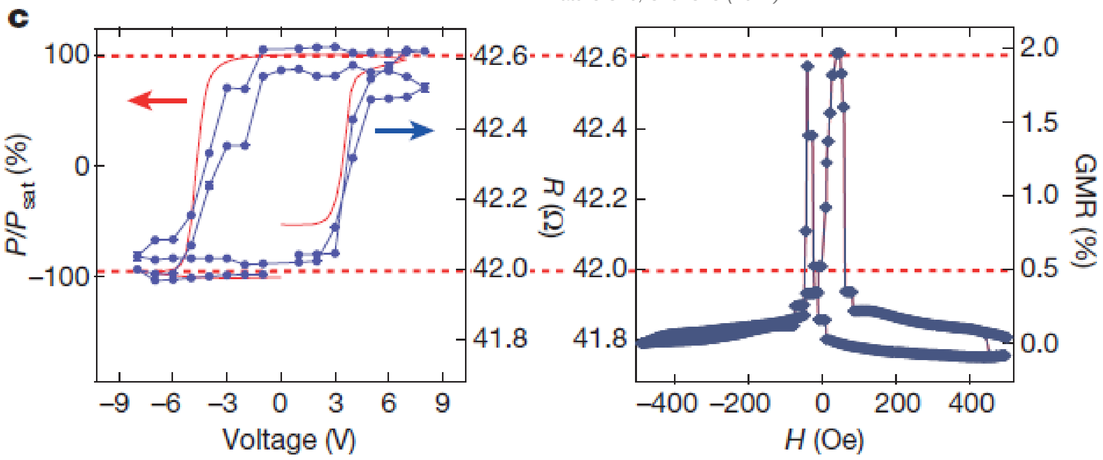






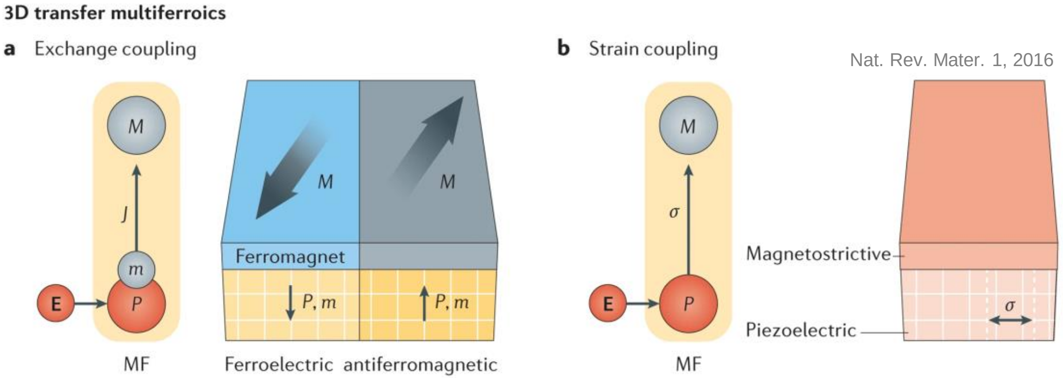


Nature 516, 370–373 (2014)



Electric field control of ferromagnetism

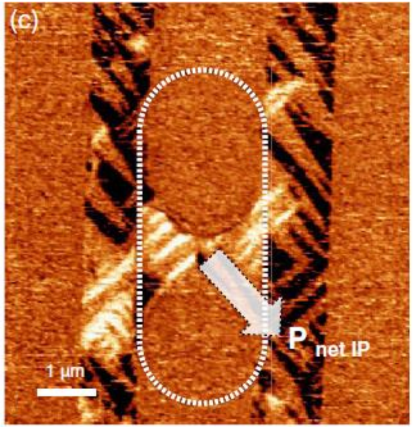
The domain correlation drives multiferroics heterostructures functionality



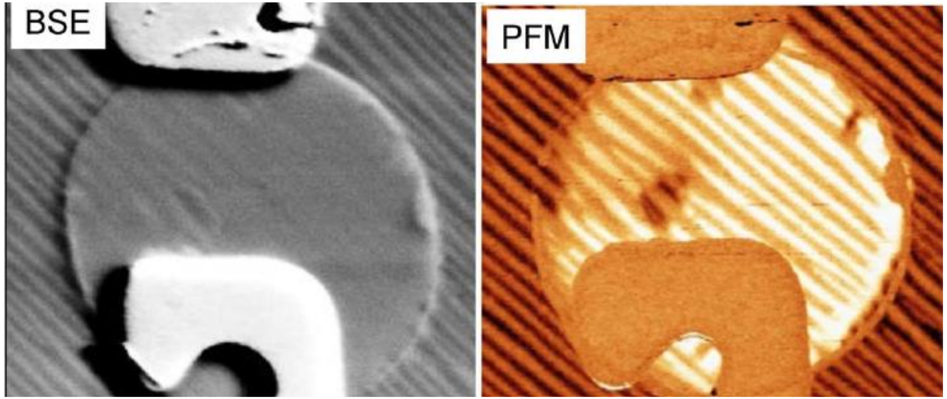
Nat. Rev. Mater. 1, 2016

Difficulty to probe buried domains, antiferromagnetic domains

APL Mater. 2, 076109 (2014)



PRL. 107, 217202 (2011)
PRB 87 134426 (2013)



Multiferroics

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- Mechanisms promoting the coexistence of magnetic and electric long-range orders (type I and type II multiferroics)

Electric-field control of ferromagnetism using multiferroics

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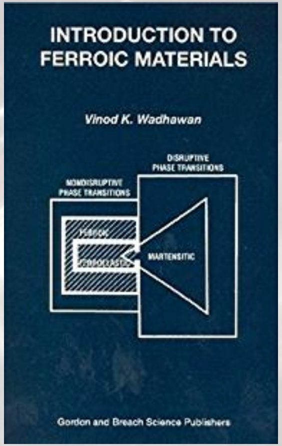
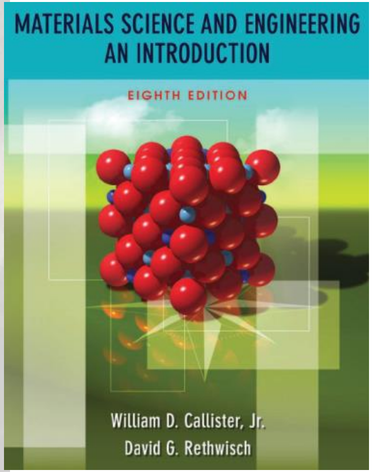
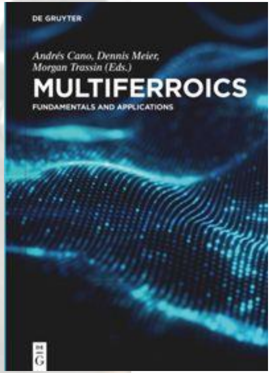
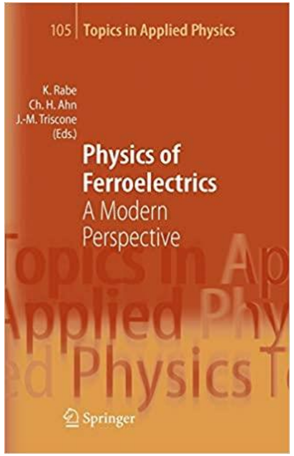
- Beyond the low energy control of magnetization







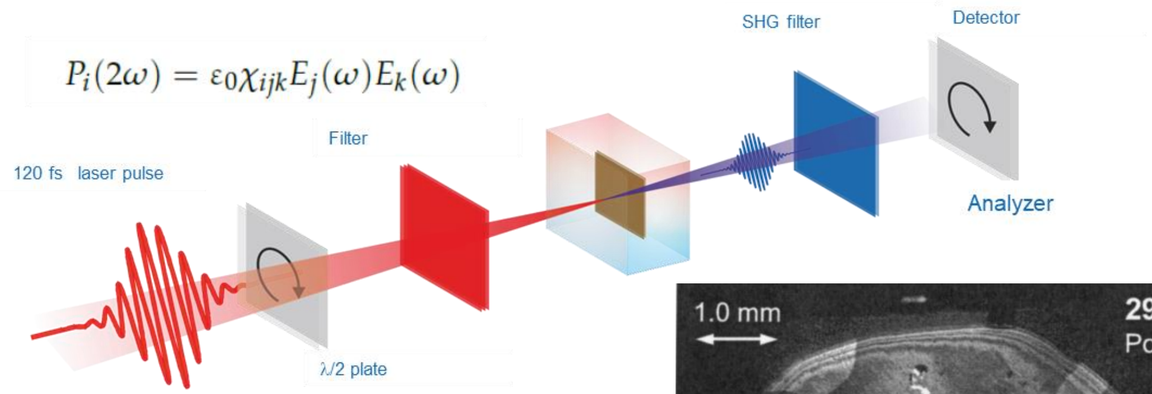


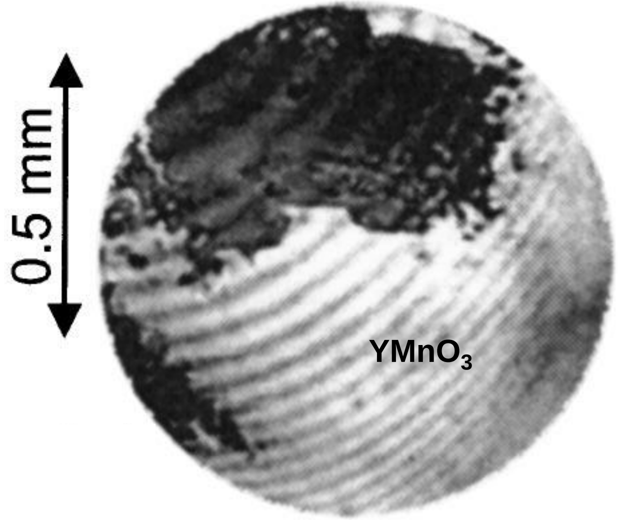
Probing multiferroic domains

Non invasive probes

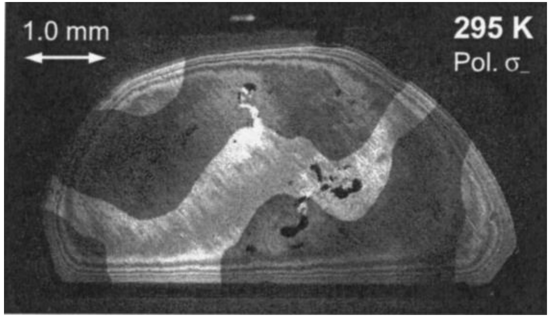
Optical second harmonic generation

$$P_i(2\omega) = \varepsilon_0 \chi_{ijk} E_j(\omega) E_k(\omega)$$

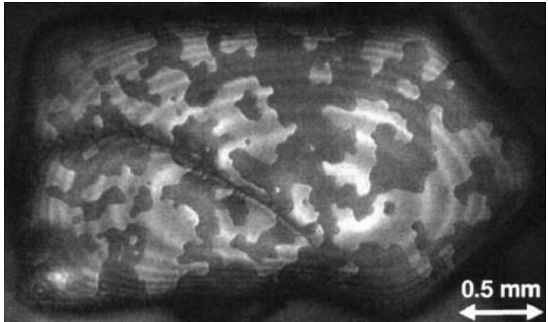




Cr_2O_3



$YMnO_3$

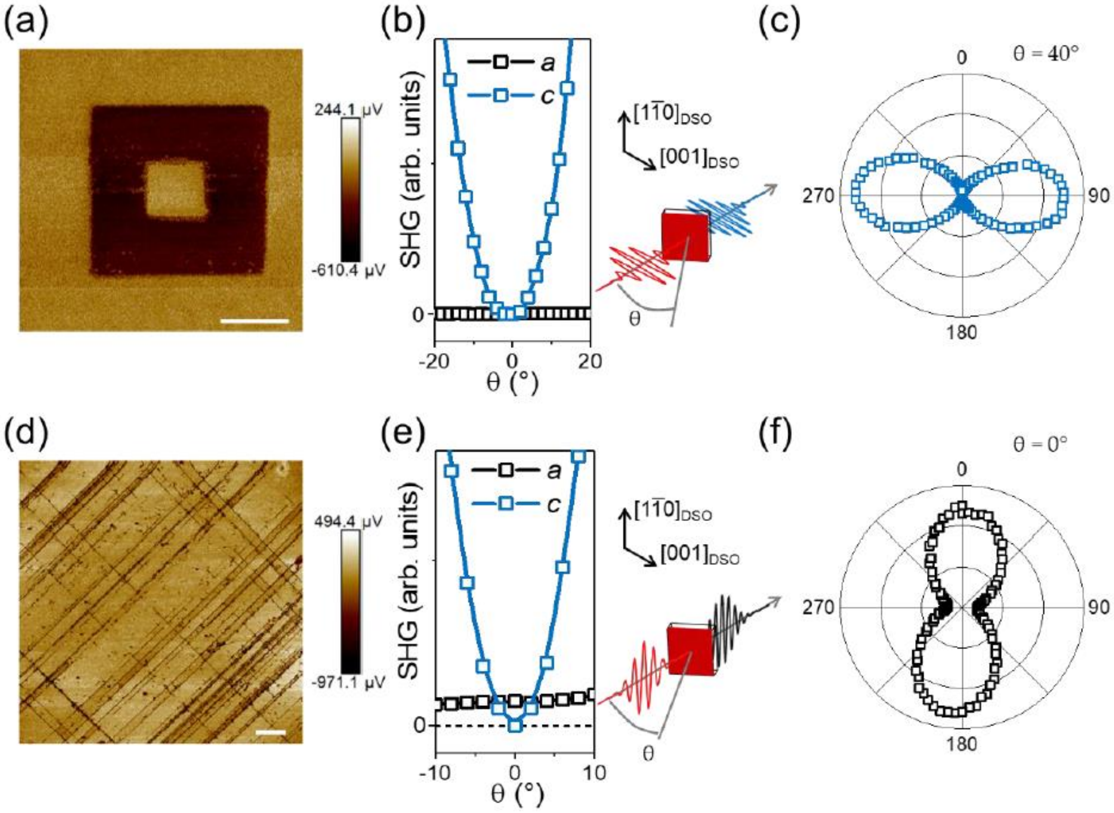


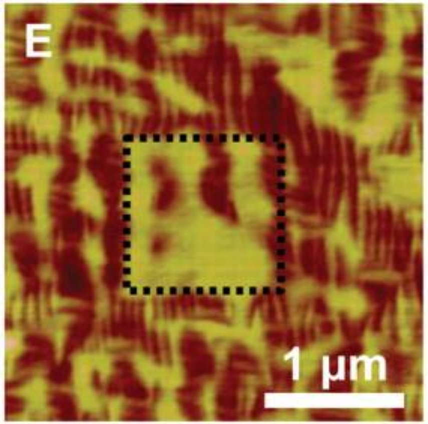
J. Am. Ceram. Soc.94, 2699 (2011)
Opt. Soc. Am. B 22, 96 (2005)
Appl. Sci. 2018, 8, 570
J. Appl. Phys. 83, 6560 (1998)

J. Am. Ceram. Soc.94, 2699 (2011)
Opt. Soc. Am. B 22, 96 (2005)
Appl. Sci., 8, 570 (2018)

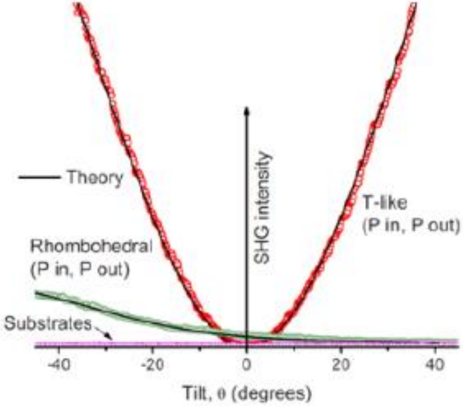
Probing multiferroic domains

Probing polarization states and ferroelectric phases in thin films.





Science 326, 977 (2009)
Appl. Phys. Lett. 97, 112903 (2010)



J. Am. Ceram. Soc.94, 2699 (2011)
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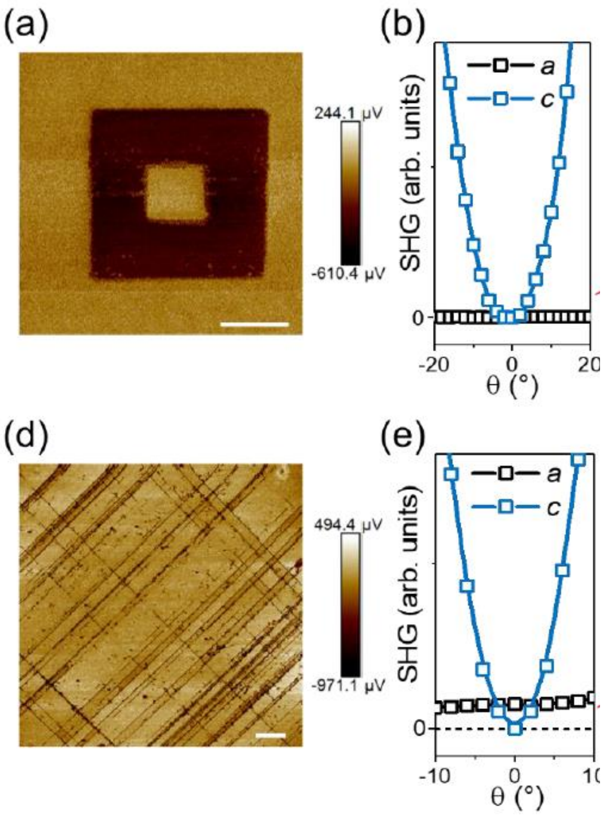
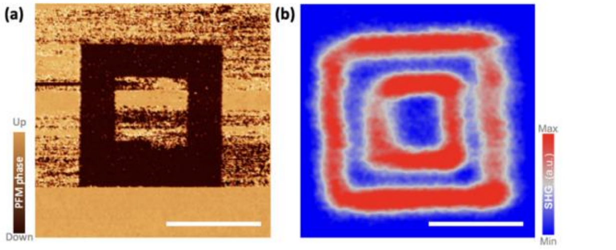
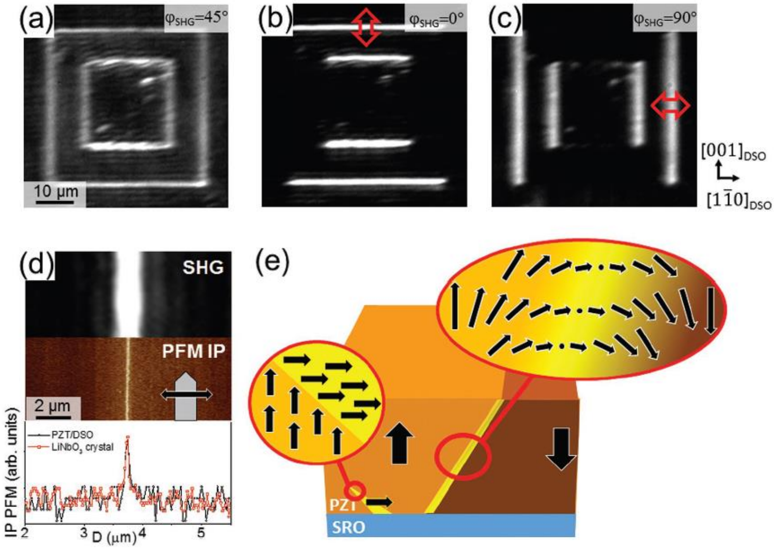


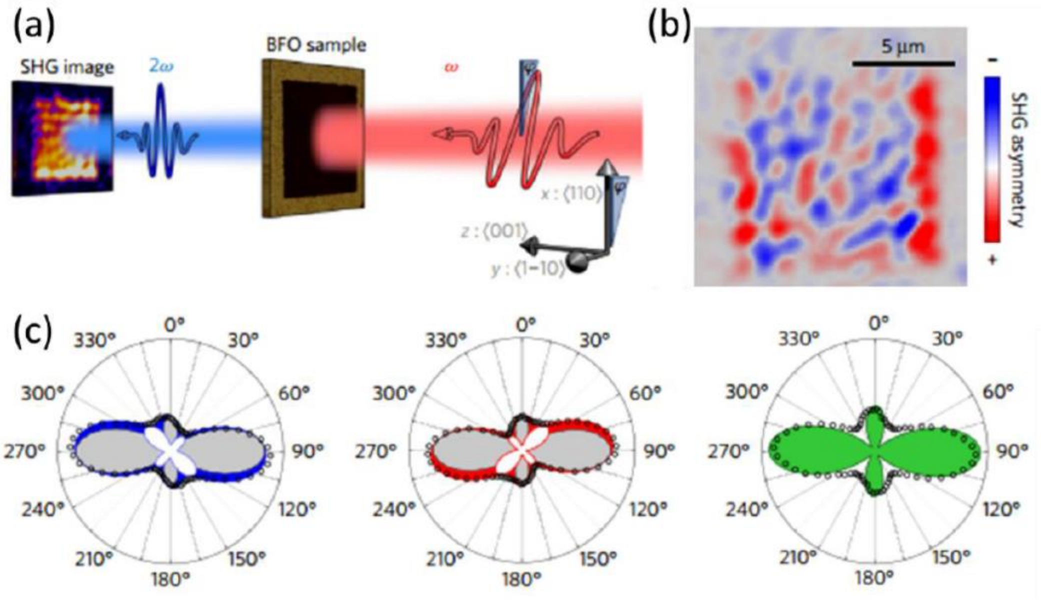


Phys. Rev. B 106, L241404 (2022)
Adv. Mater. 2017, 29, 1605145

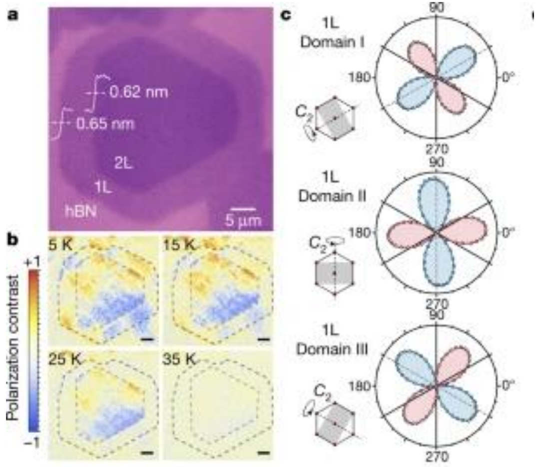


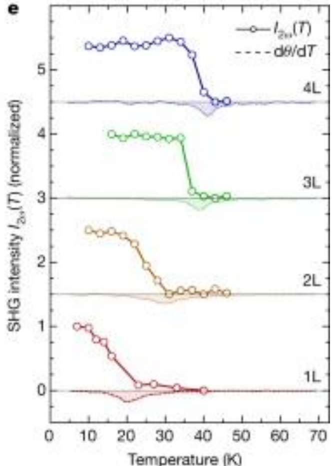
Probing multiferroic domains

Probing multiferroic states in thin films.



Nat. Mater. 16, 803 (2017)



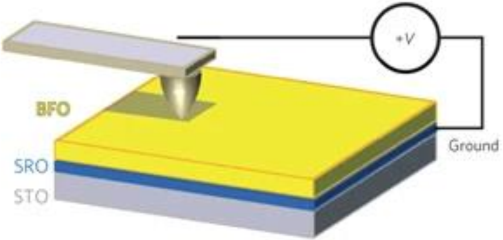


type-II multiferroic in van der Waals NiI_2

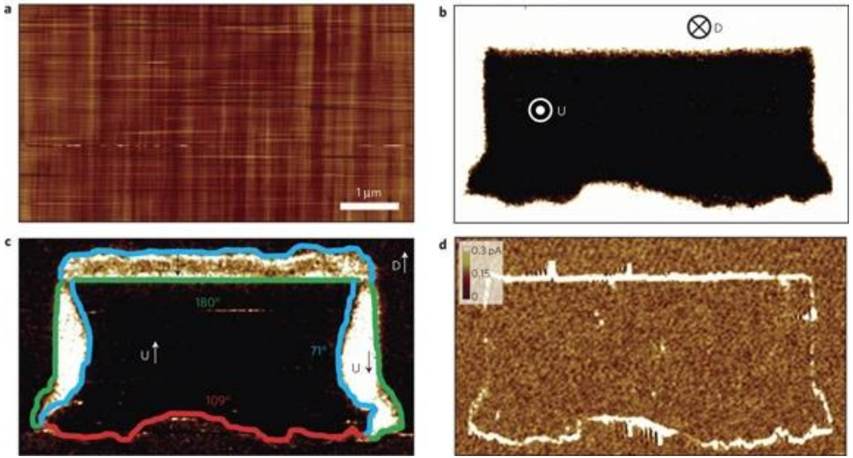
Song, Q. et al., Nature 602, 601 (2022)

Beyond the electrical control of magnetism : ferroelectric domain wall conduction

Probes sensitive to ferromagnetic and ferroelectric states
Piezoresponse force microscopy



Nat. Mater. 8 229–234 (2009)

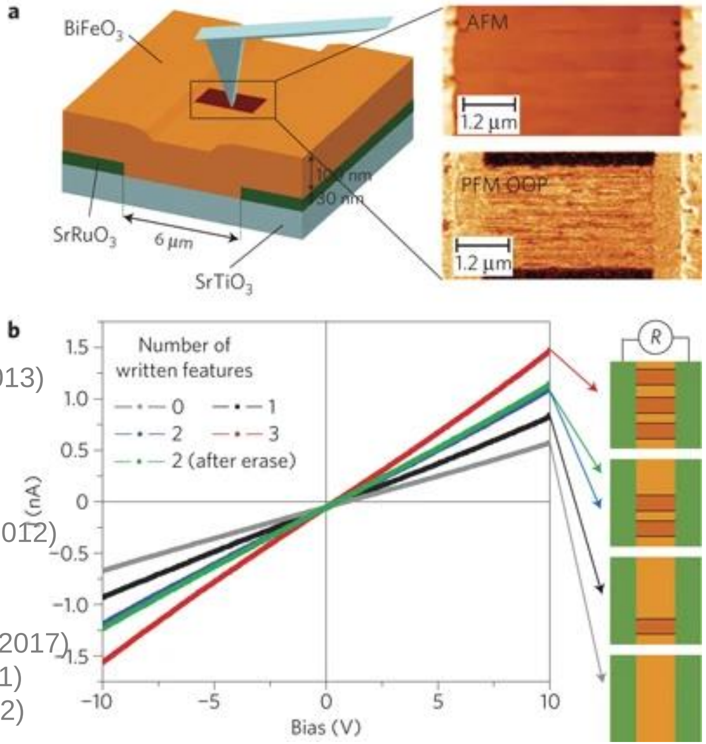


BTO
Nat. Commun. 4, 1808 (2013)

YMO
Nat. Mater.11, 284–288 (2012)

PZT
Adv. Mater. 29, 1605145 (2017)
Adv. Mater. 23, 5377 (2011)
Nano Letters 12, 209 (2012)

Discovery of conduction at charged ferroelectric domain walls in $BiFeO_3$

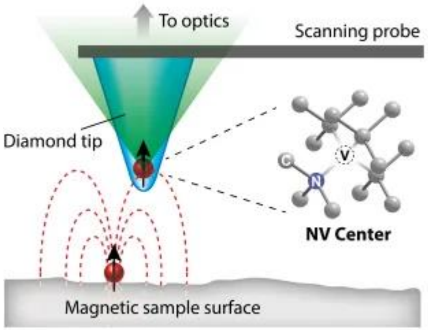


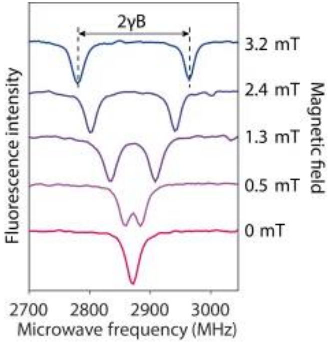
Probing multiferroic domains

Probes sensitive to antiferromagnetic states

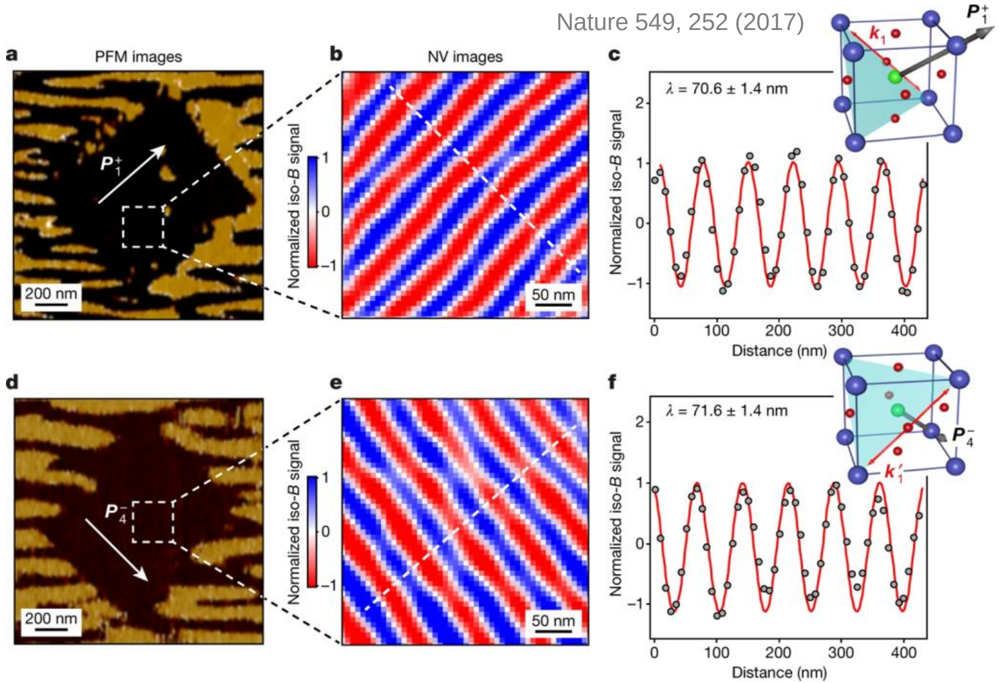
Single spin magnetometry : Nitrogen-vacancy (NV) defect in diamond

The electronic spin of a single NV defect is placed at the apex of a diamond scanning probe tip for atom-sized magnetic field sensor (nT)





Nature 549, 252 (2017)

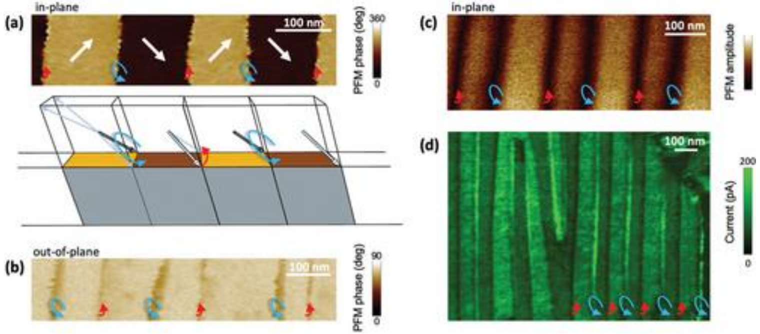


The magnetic interaction is probed by microwave excitations. When the energy of the microwaves equals the magnetic interaction: rapid reorientation -> drop in fluorescence

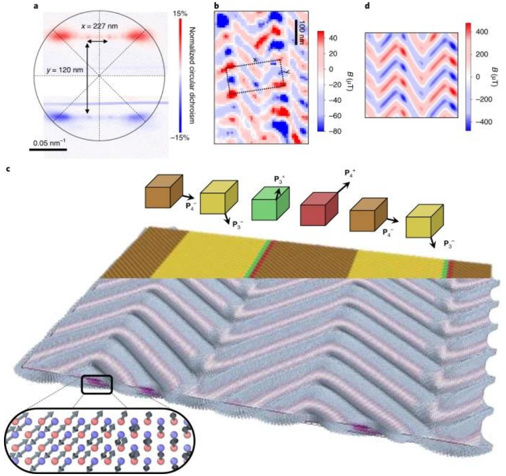
D MATL.

Beyond the electrical control of magnetism : ferroelectric domain wall chirality

Chirality at polar domain walls and electric analogue of the DMI

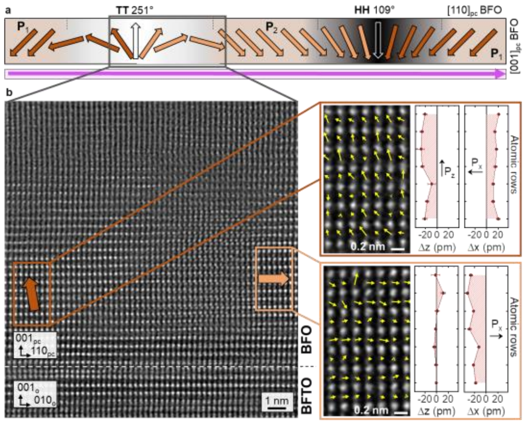


Adv. Electron. Mater. 2022, 8, 2101155



Homochirality in magnetic
and ferroelectric domains walls

Nature Materials,19, 386 (2020)

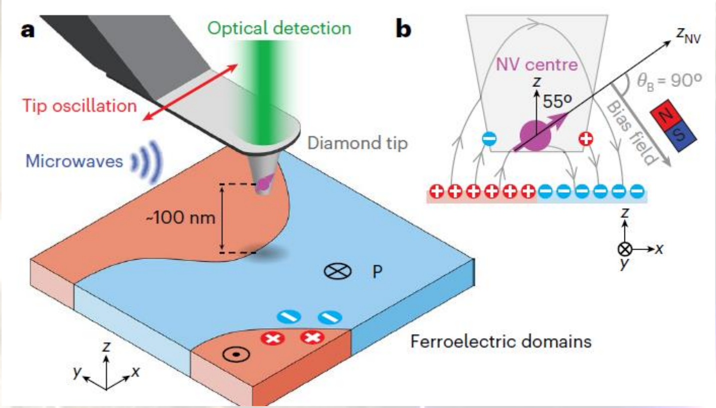


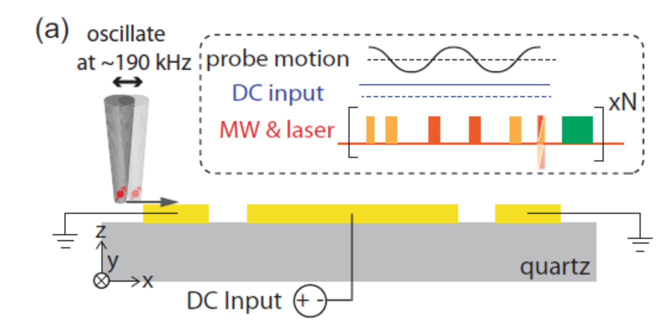
Origin of the homochirality
Symmetry allowed non collinear electric dipoles (mediated by oxygen octahedral tilting)
Bulk or interface?

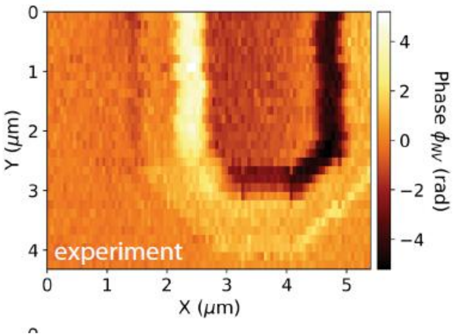
Nature Materials 20, 341 (2021)
Physical Review B 102, 024110 (2020).

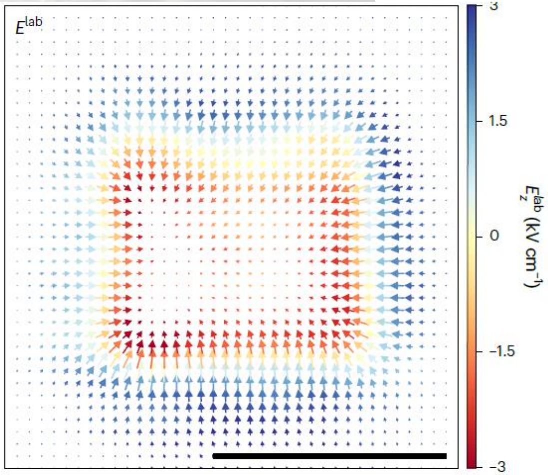
What's next?

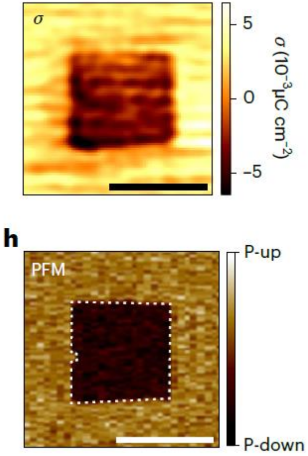
Single spin **electrometry**











Synchronizing the NV response to the tip oscillation frequency allows for electric field sensing.

Electric field variation faster than screening charge displacement at the surface of the tip:
Stark effect detection enables

Nat. Phys. 19, 644 (2023)
npj Quantum Inf 8, 107 (2022)