Spintronics with Ferroelectrics

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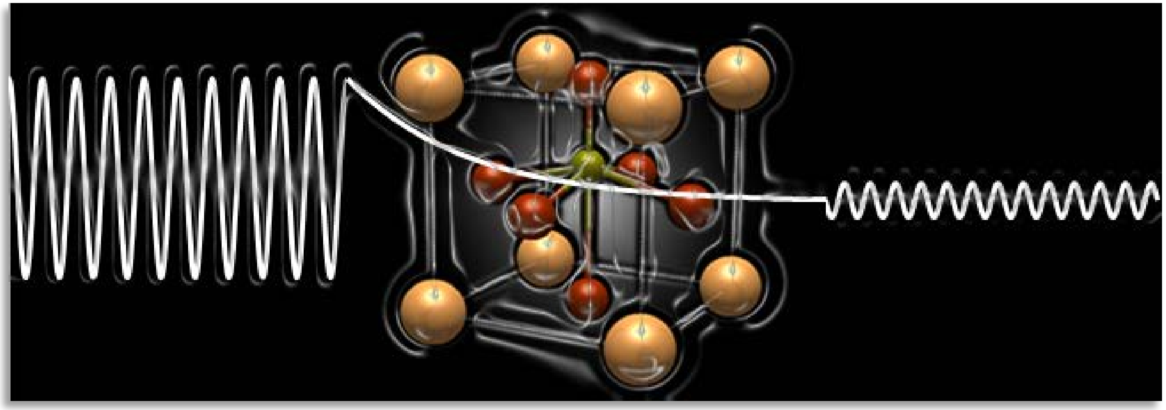
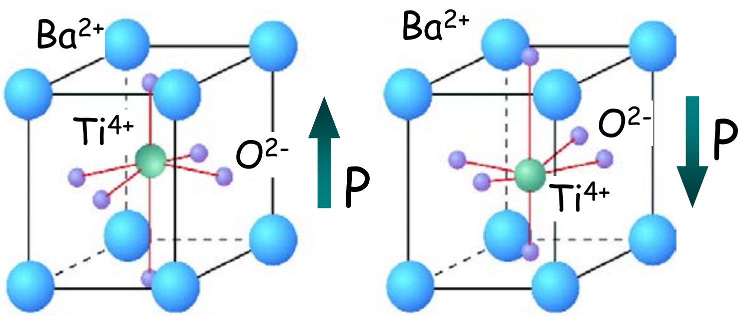
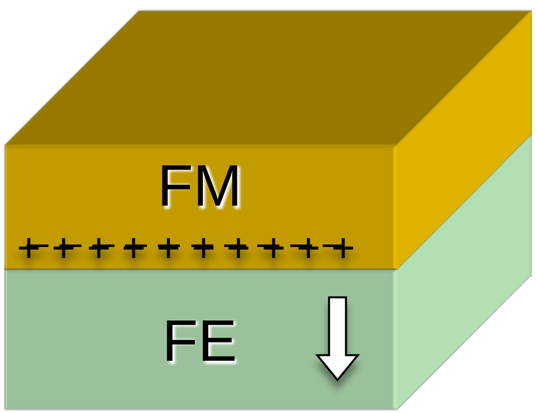


Ferroelectrics: Field Effect

Ferroelectric $BaTiO_3$





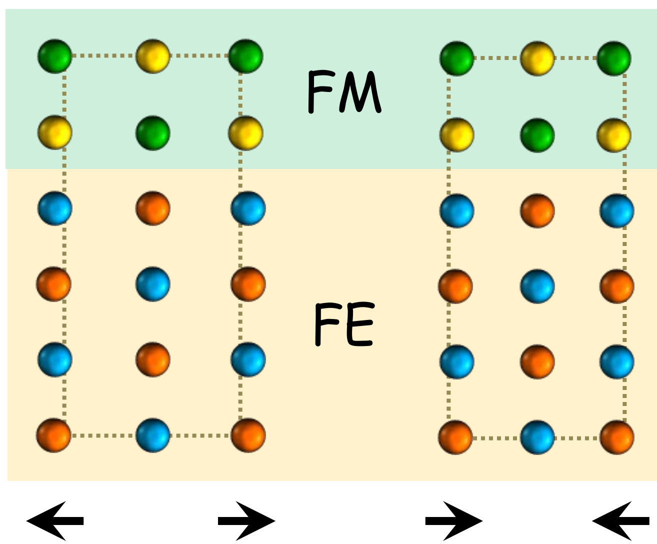
- Spontaneous electric polarization switchable by electric field
- Polar displacements ~ 0.1-0.2Å
- Perovskite structure
- Large polarization charge: 10 – 100 μC/cm² (~ 0.1 – 1 e/a²)
- Polarization charge screening: "field effect". Effective electric fields up to 10 V/Å (1 GV/cm)
- Controlled by fields of ~ 1 MV/cm Three order in magnitude smaller!
- Non-volatile capability

- Control of spin-dependent properties relevant to spintronics

Ferroelectrics: Strain and Bonding

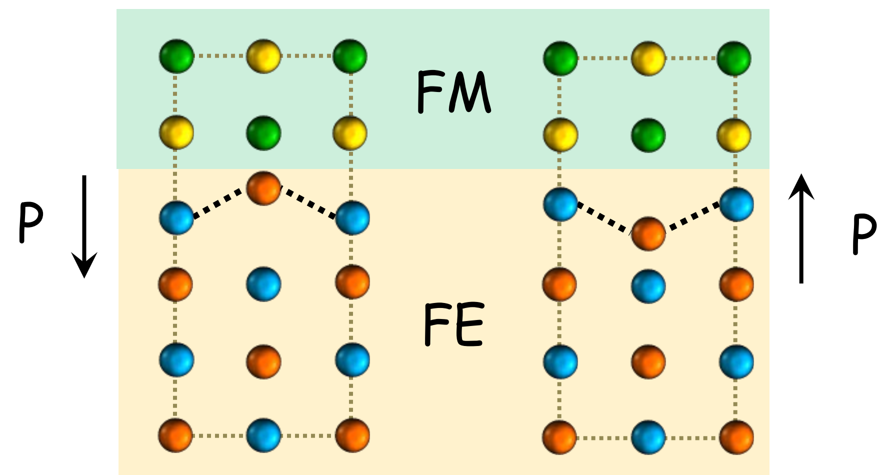
Ferroelectrically induced:

- Interface strain effects



Tensile Compressive

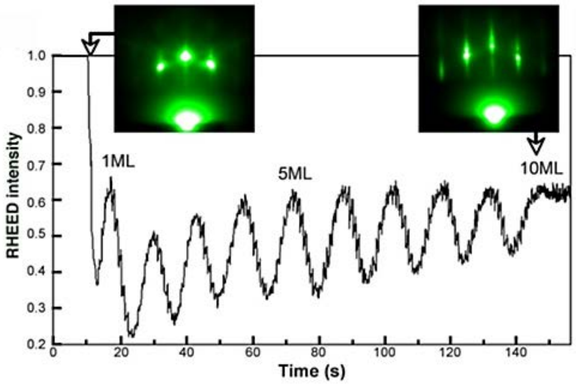
- Interface bonding effects



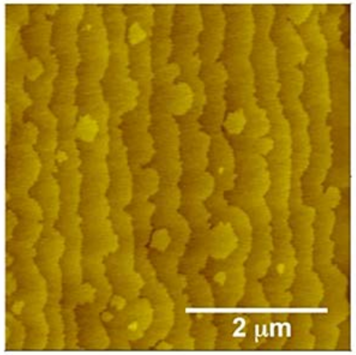
Ferroelectrics: Epitaxial Thin Films

- High quality epitaxial structures with nearly atomic precision interface control: PLD, MBE
- Epitaxial strain to stabilize polarization
- Stable and switchable polarization in sub nm thick films

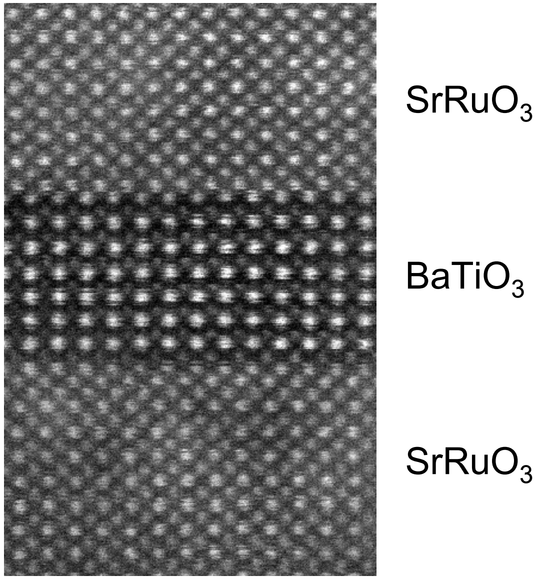
RHEED



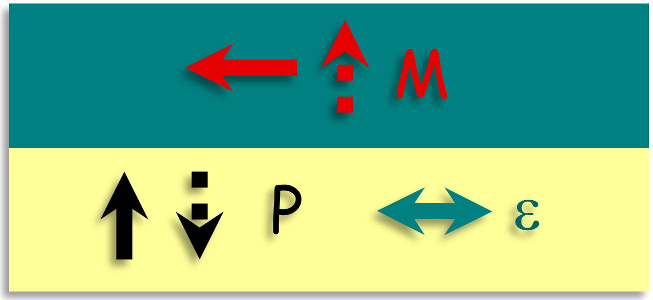
AFM



HRTEM



Magnetoelectric Interfaces



Magnetic (Ferromagnetic, Ferrimagnetic, Antiferromagnetic)

Ferroelectric

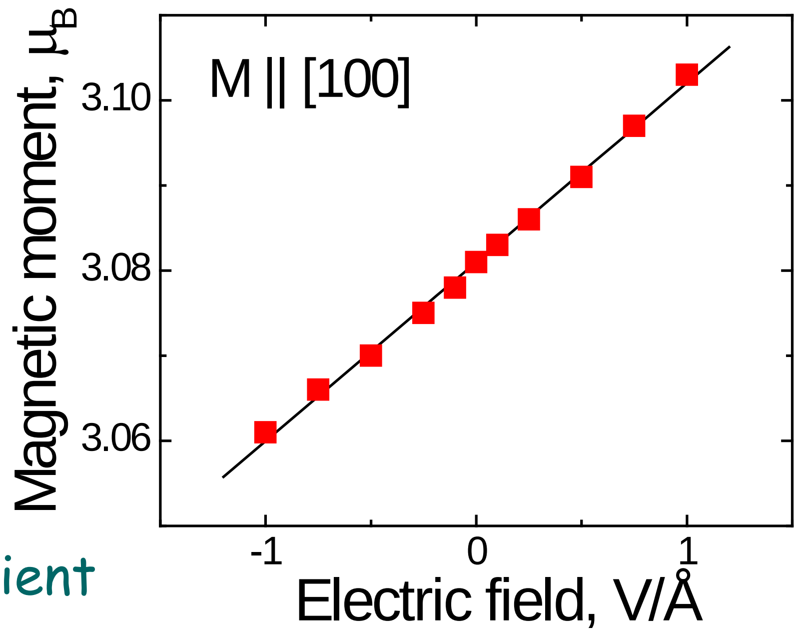
- Strain mediated:
  Piezoelectricity & Magnetostriction or Piezomagnetism
- Electronically driven:
  Spin-polarized screening or interface bonding

  Effects of ferroelectric polarization on:

- Interface magnetization
- Magnetocrystalline surface anisotropy
- Interface magnetic order
- Curie temperature
- Electron and spin transmission across interfaces
- Exchange bias
- Spin waves

Surface Magnetoelectric Effect on Fe (001)

Duan et al, PRL **101**, 137201 (2008)



- broken space inversion & time reversal symmetries

$$\mu_0 \Delta M = \alpha_s E$$

α_s – surface magnetoelectric coefficient

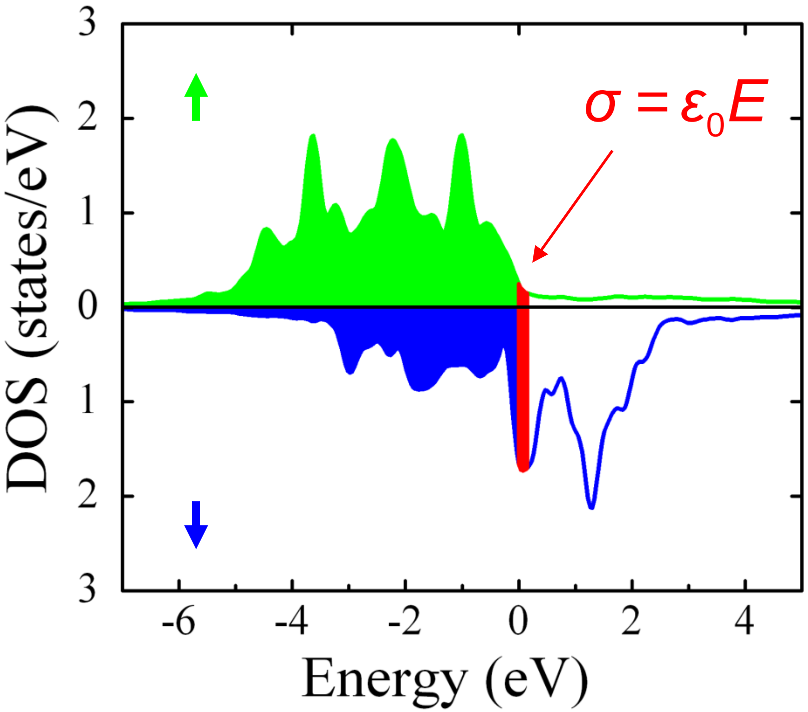
$\alpha_s \approx 2.4\times10^{-14}$ G cm^2/V

E = 1 V/nm -> $\Delta m \approx 2\times10^{-3}\mu_B$ per Fe

- Electric field induces a surface magnetic moment
- Effect is small

Origin of Magnetoelectric Effect

Fe(001) Surface DOS



$$\sigma = \sigma^{\uparrow} + \sigma^{\downarrow}$$

$$\sigma^{\uparrow} = \varepsilon_0 E \frac{n^{\uparrow}}{n}; \ \sigma^{\downarrow} = \varepsilon_0 E \frac{n^{\downarrow}}{n}$$

$$\Delta M = \mu_B \frac{\sigma^{\uparrow} - \sigma^{\downarrow}}{e} = \frac{\varepsilon_0 E \mu_B}{e} \frac{n^{\uparrow} - n^{\downarrow}}{n^{\uparrow} + n^{\downarrow}}$$

Magnetoelectric coefficient:

$$\alpha_s = \frac{\mu_B}{ec^2} \frac{n^{\uparrow} - n^{\downarrow}}{n^{\uparrow} + n^{\downarrow}} = \frac{\mu_B}{ec^2} P$$

$$\mu_0 \Delta M = \alpha_s E$$

- Fundamental limit for the linear ME effect
- Universal constant for half-metals: $\alpha_s = \frac{\mu_B}{ec^2} \approx 6.44 \times 10^{-14} \text{ Gcm}^2 / \text{V}$

Duan et al., PRB **79**, R140403 (2009)

Ferroelectric Effect on Interface Magnetization

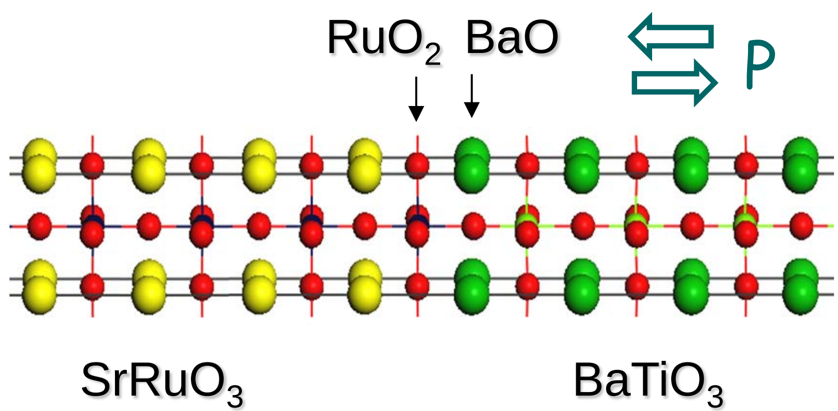
Niranjan et al, APL 95, 052501 (2009)

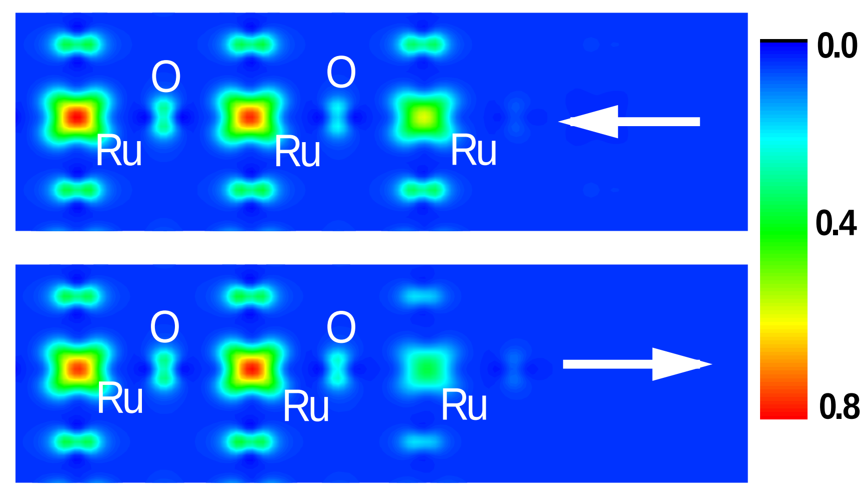
$SrRuO_3$

- Itinerant ferromagnet (~1 μ_B/f.u.)
- Curie temperature 160 K
- Perovskite structure
- Good lattice match to $BaTiO_3$
- Epitaxial strain to stabilize polarization ($SrTiO_3$ substrate)

$\Delta m_{Ru} = 0.31\ \mu_B$

$E_c \sim 0.1$ V/nm



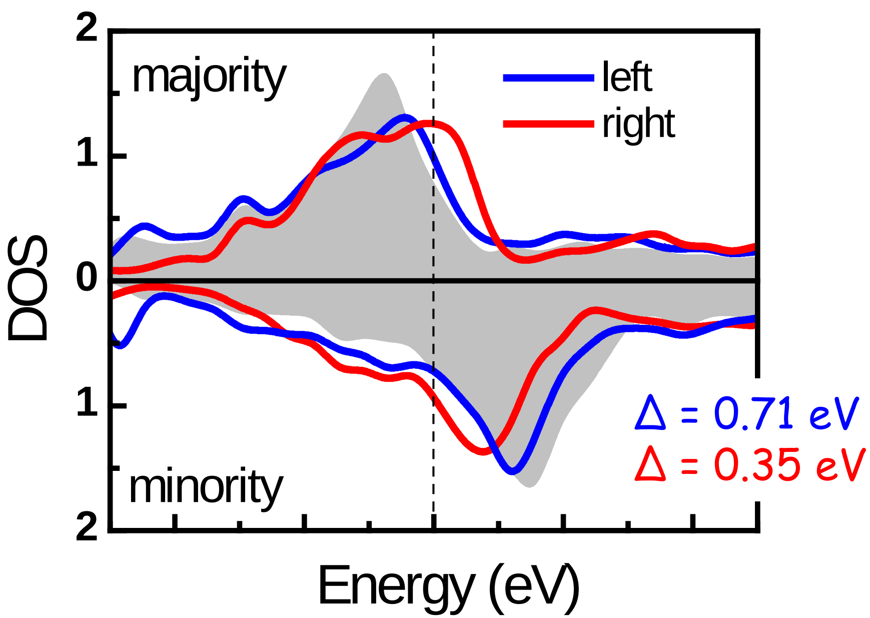


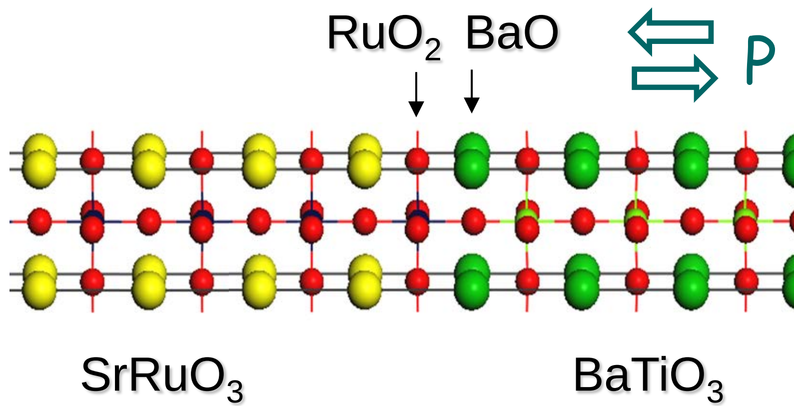
Spin density

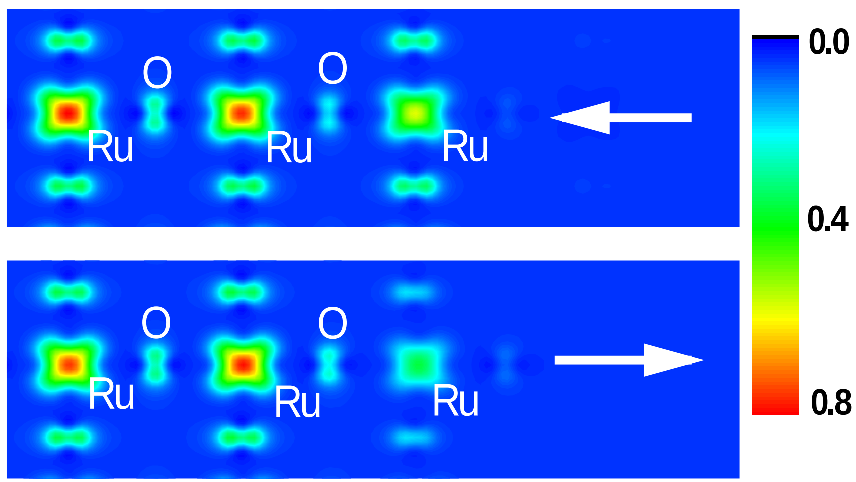
Ferroelectric Effect on Interface Magnetization

Niranjan et al, *APL* 95, 052501 (2009)

Spin resolved density of states







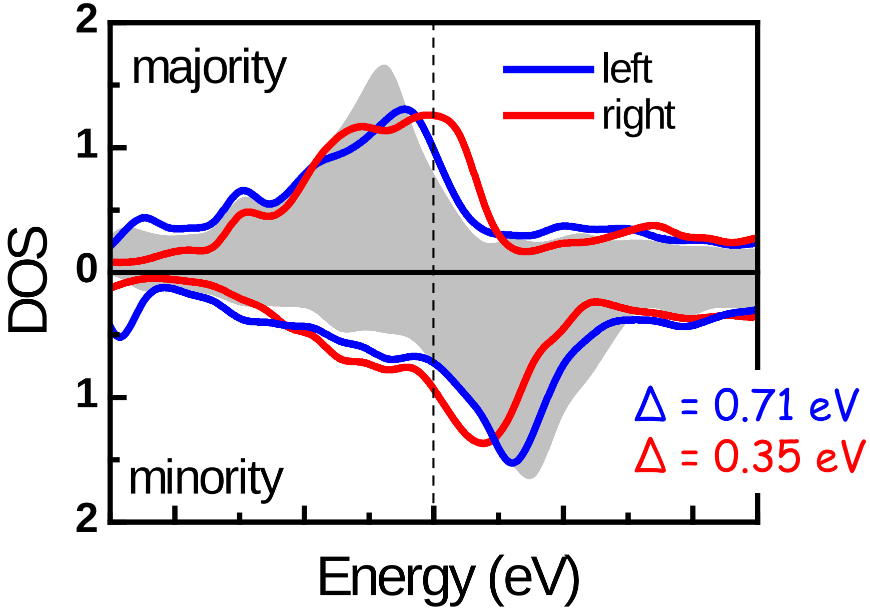
Spin density

- Change in the exchange splitting at the interface

Ferroelectric Effect on Interface Magnetization

Niranjan et al, APL 95, 052501 (2009)

Spin resolved density of states



- Change in the exchange splitting at the interface

Stoner model for itinerant magnetism:

$$\Delta = Im$$

I – Stoner exchange constant

$$\Delta = \frac{2\sqrt{\rho_F^2 I^2 - 1}}{|\rho_F'| I}$$

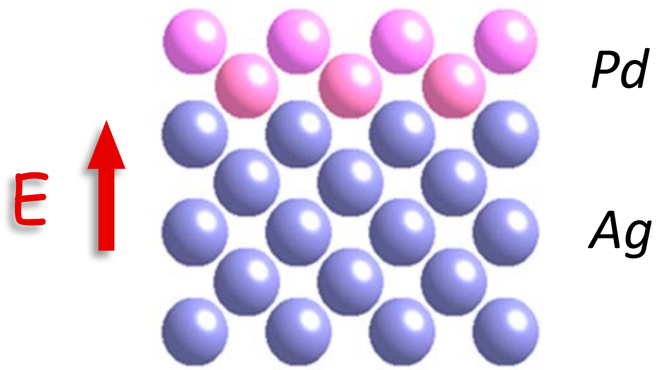
- Explains calculated change in exchange splitting with FE polarization reversal

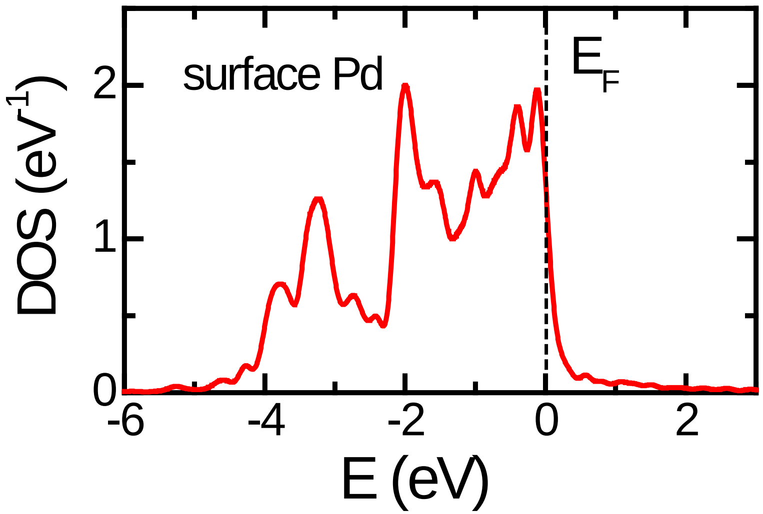
Inducing Magnetism on a Non-Magnetic Surface

Sun et al., PRB 81, 064413 (2010)

Stoner criterion for itinerant magnetism

$$I\rho_F > 1$$

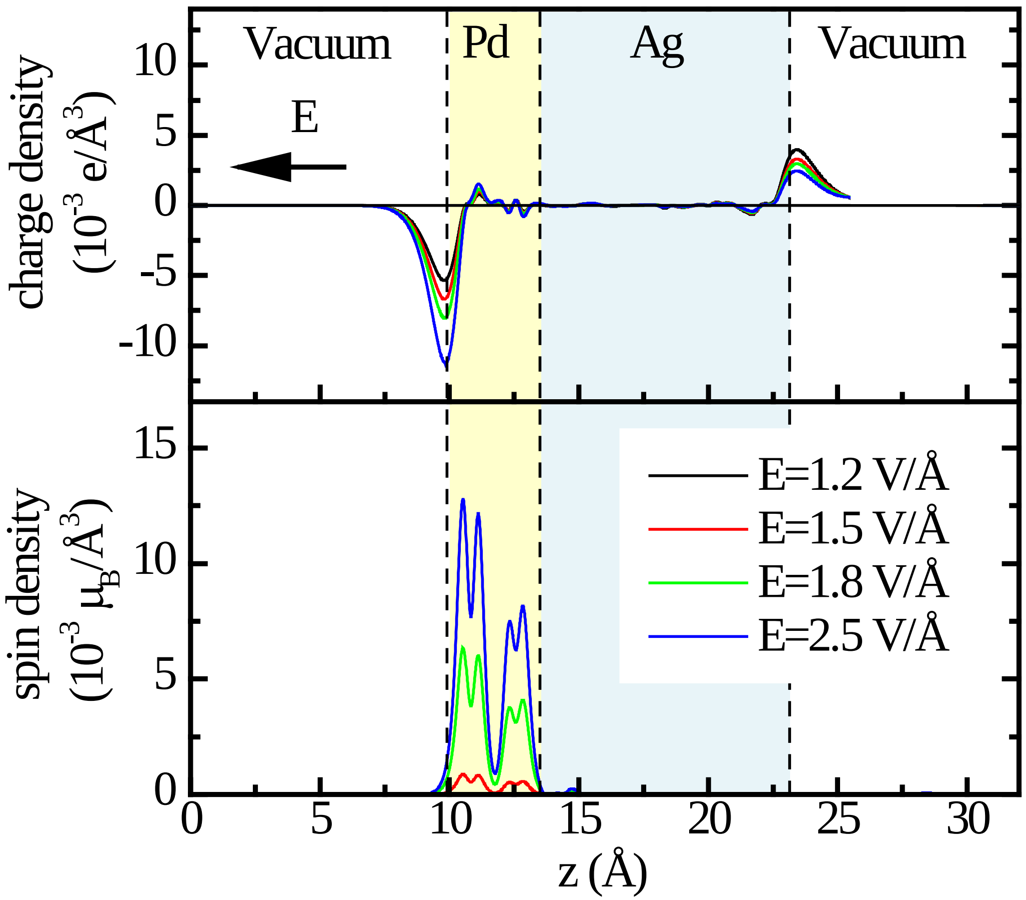




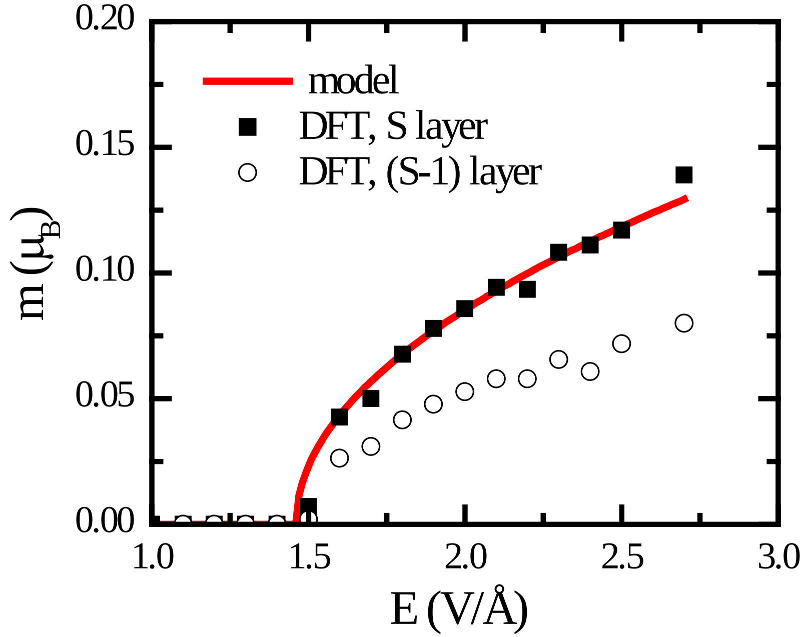
$$I \approx 0.65\text{eV} \; ; \quad I\rho_F \approx 0.96$$

- Can we induce a surface magnetism by electric field ?

Electric-Field Induced Magnetism on a Pd (001) Surface



Sun et al., PRB 81, 064413 (2010)



- Second-order phase transition
- Require very large electric fields which may be produced by an adjacent ferroelectric

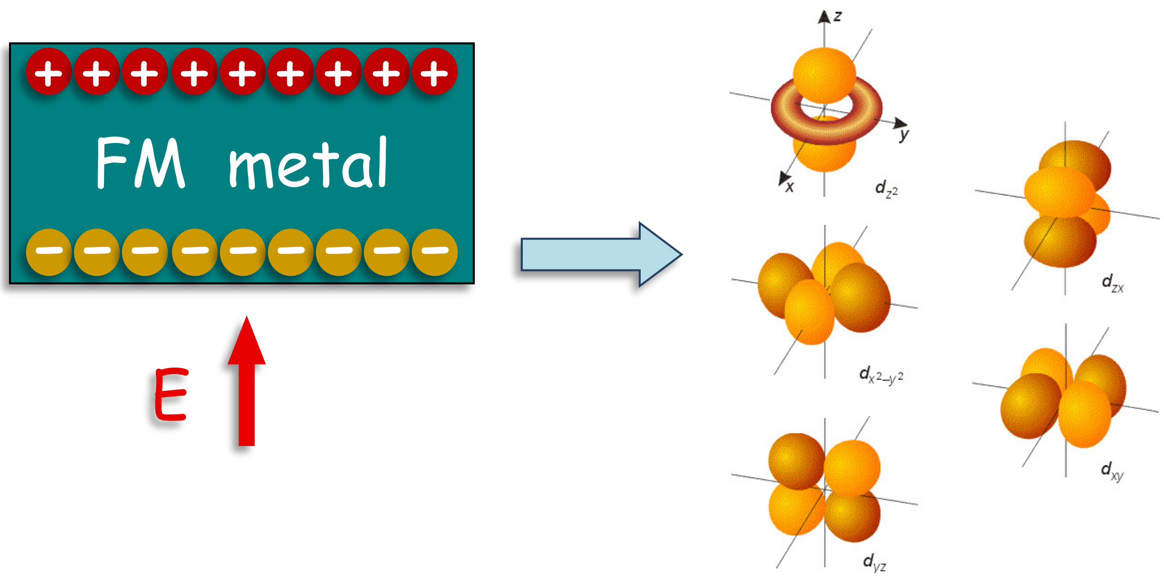
Linearized Stoner model:

$$m(E) = \frac{2\sqrt{2\alpha\rho_F}}{I\left|\rho_F'\right|}\sqrt{E - E_c}$$

$$\rho_F(E) = \rho_F + \alpha E$$

Electric Field Effect on Magnetocrystalline Surface Anisotropy

➢ Electric field control of magnetization orientation



- Screening charge induced by electric field

→ Change in relative population of d-orbitals at the surface

→ Change in surface magnetocrystalline anisotropy (MCA)

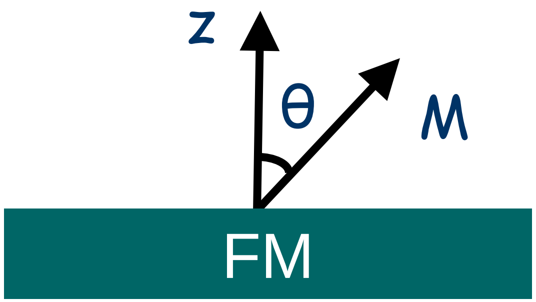
$$\Delta K = \beta_s E$$

β_s – VCMA coefficient

Magnetic Anisotropy

- Bulk magnetocrystalline anisotropy (MCA)
- Surface (interface) MCA
- Shape anisotropy

MCA energy determined by spin-orbit interaction: $H = \xi \mathrm{L} \cdot \mathrm{S}$



In thin films:

$E_{shape} \approx \mu_0 M^2 \cos^2\theta$ - volume

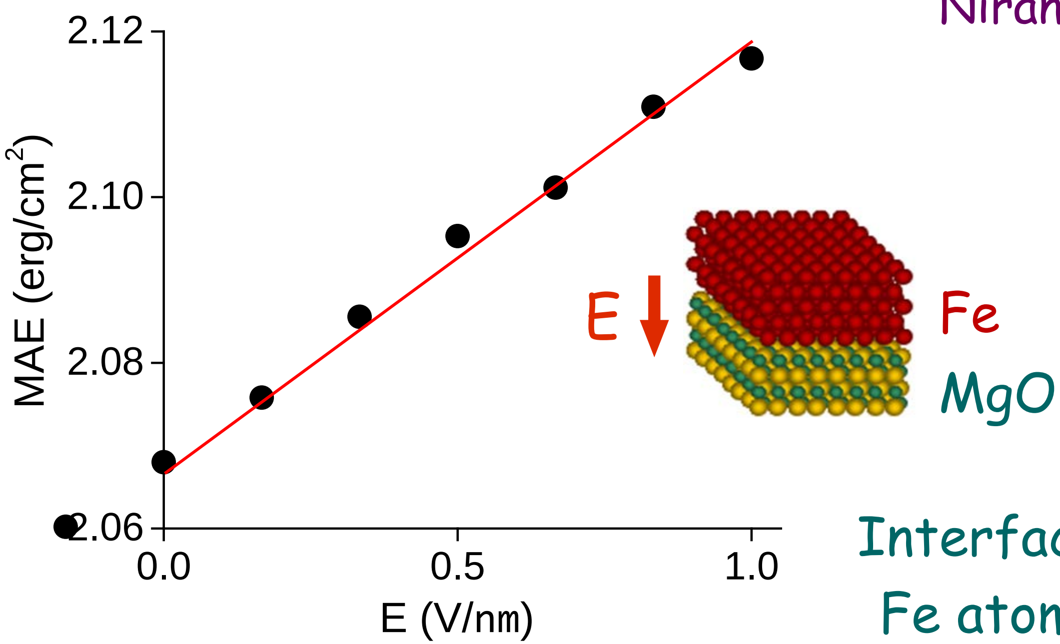
$E_{surface} \approx - K_S \cos^2\theta$ - surface

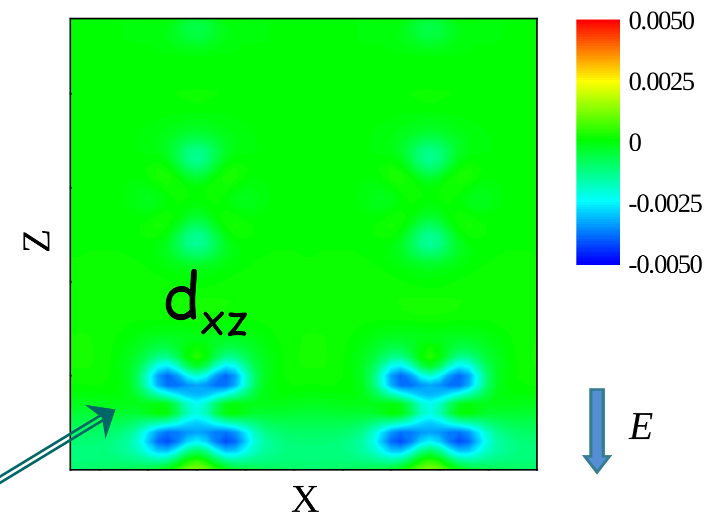
$$K \propto \sum_{o,u} \frac{|\langle o|L_z|u\rangle|^2 - |\langle o|L_x|u\rangle|^2}{\varepsilon_u - \varepsilon_o}$$ within 2nd order perturbation theory

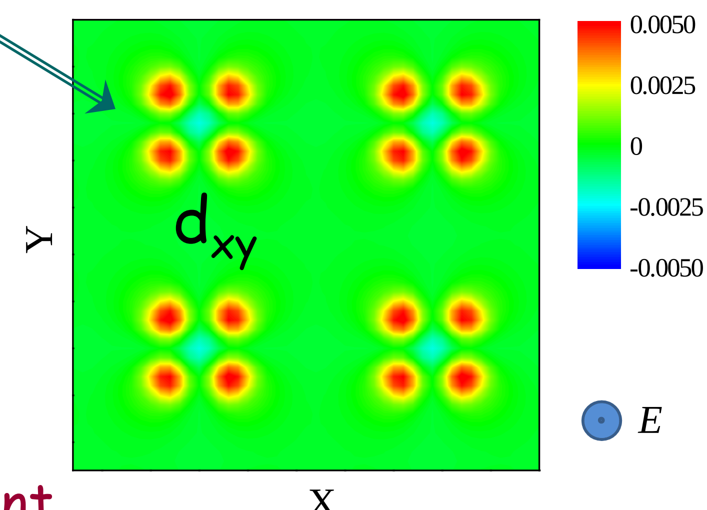
- sensitive to orbital population

Fe/MgO(100) Interface

Niranjan et al., APL 96, 222504 (2010)





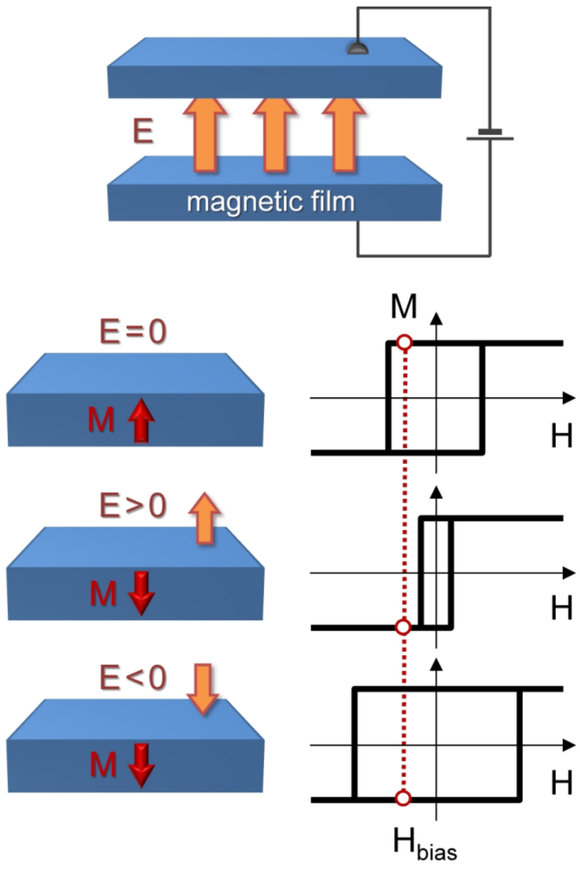


- Large perpendicular magnetic anisotropy (PMA): K ~ 2 erg/cm^2
- Increases with field away from interface
- Effect due to change in occupation of Fe-3d orbitals (d_{xz},d_{yz} $\rightarrow$ d_{xy})

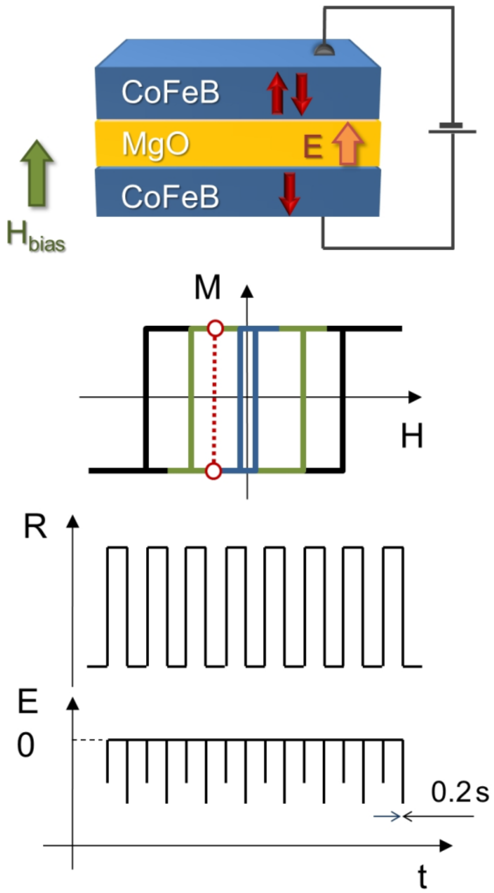
$\beta_s \approx$ 0.05 pJ/Vm – consistent with experiment

Toggle Switching of Magnetization

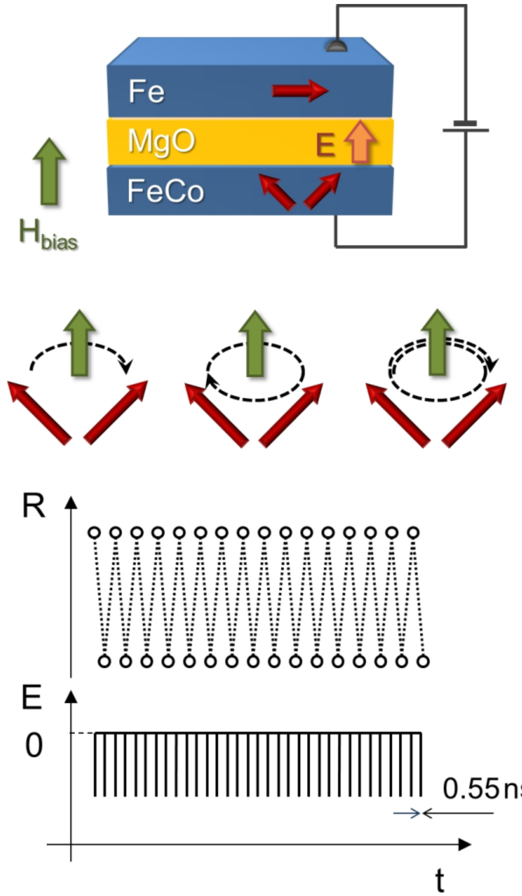
Tsymbal,
Nat. Mater. 11, 12 (2012)



Wang et al.,
Nat. Mater. 11, 64 (2012)



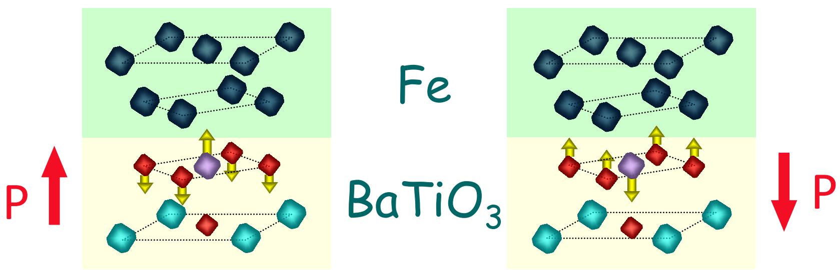
Shiota et al.,
Nat. Mater. 11, 39 (2012)



- ❑ Significant interest from industry

Ferroelectric Control of Magnetic Interface Anisotropy

Lukashev et al., JPCM 24, 226003 (2012)

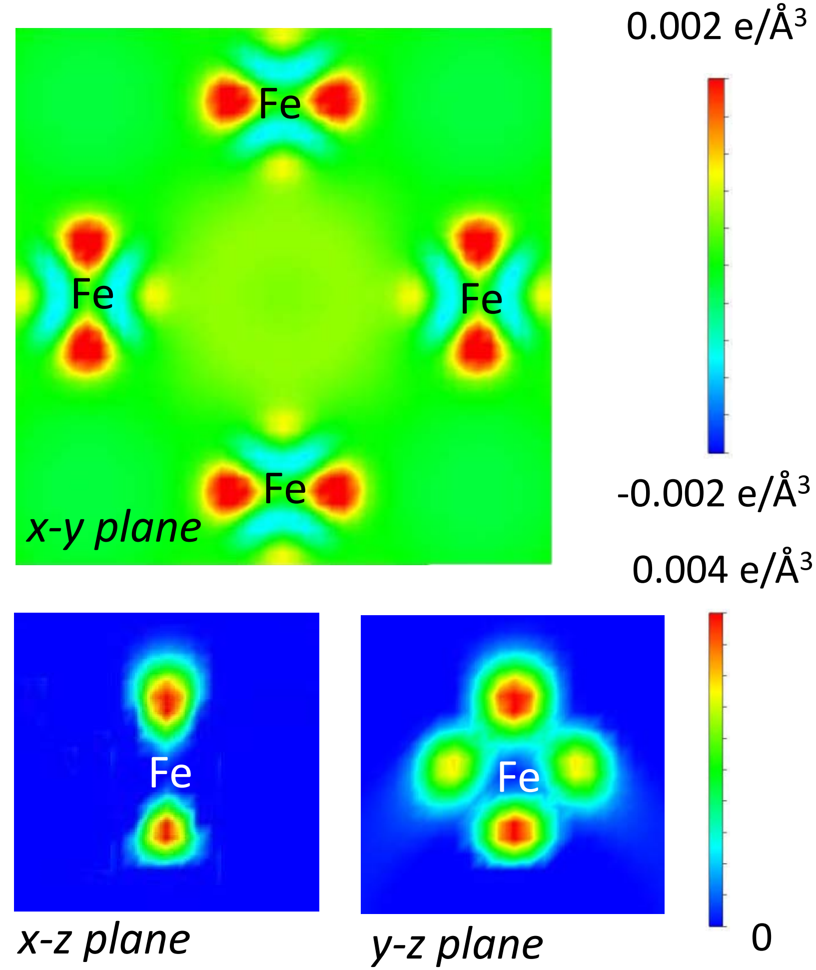


K ≈ 1.3 erg/cm² K ≈ 1.0 erg/cm²

- Large perpendicular anisotropy
- Large change with polarization switching
- Effect due to the relative change in 3d orbitals occupation
- Consistent with Fe/MgO

$E_c \sim 0.1$ V/nm $\Rightarrow \beta_s \sim 3$ pJ/Vm

Induced charge density

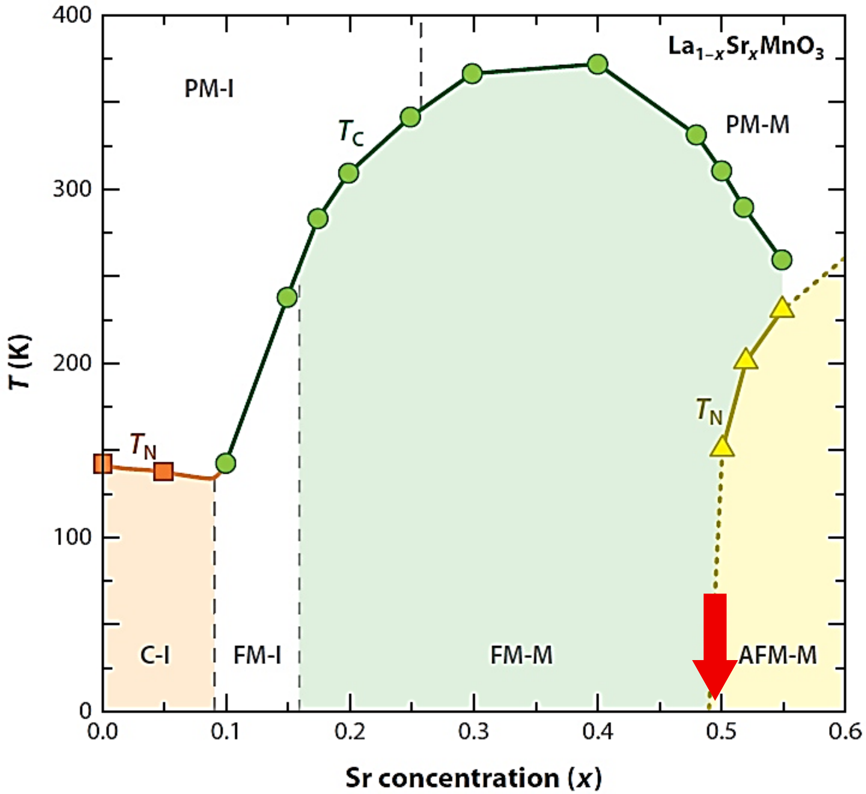


- VCMA effect enhanced by two orders in magnitude!

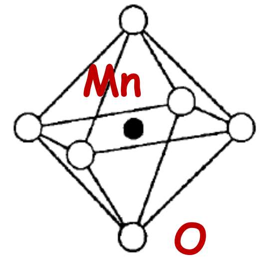
Electrostatic Doping of Magnetic Perovskites: $La_{1-x}A_xMnO_3$

- Magnetic perovskites: $La_{1-x}A_xMnO_3$ (A^{2+} = Ca, Sr, Ba)

$La_{1-x}Sr_xMnO_3$



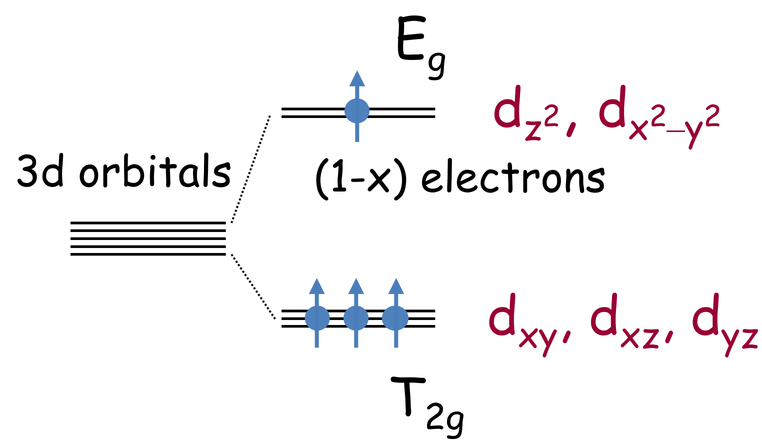
Hole doping of $LaMnO_3$:



$LaMnO_3$: E_g^1

$La_{1-x}Sr_xMnO_3$: E_g^{1-x}

$SrMnO_3$: E_g^0



- Transition from FM to AFM order near x ~ 0.5

Electrostatic Doping of Magnetic Perovskites: $La_{1-x}A_xMnO_3$

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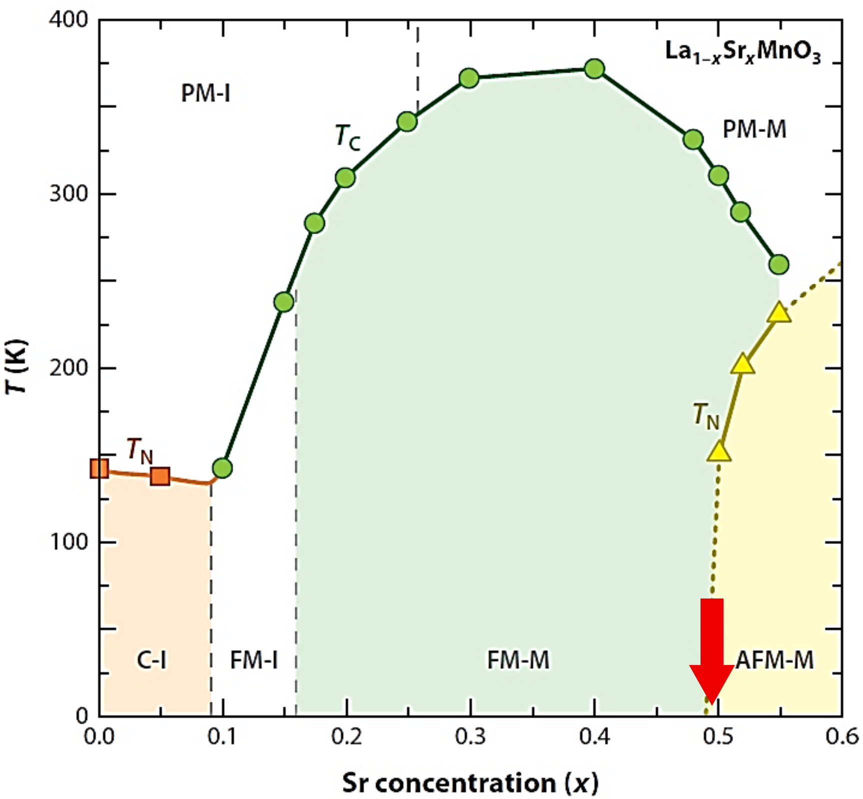
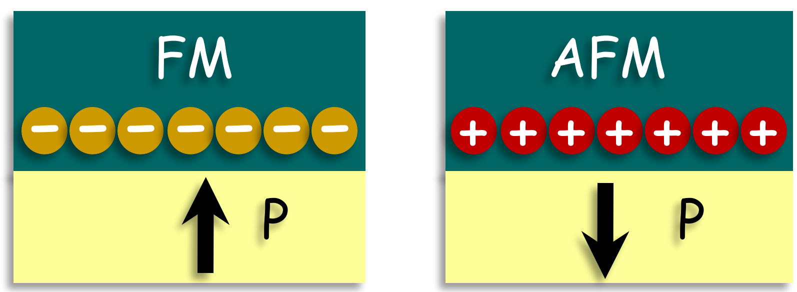
$La_{1-x}Sr_xMnO_3$



Changing magnetic order by "electrostatic doping" at an interface with a ferroelectric?

$La_{0.5}Sr_{0.5}MnO_3$

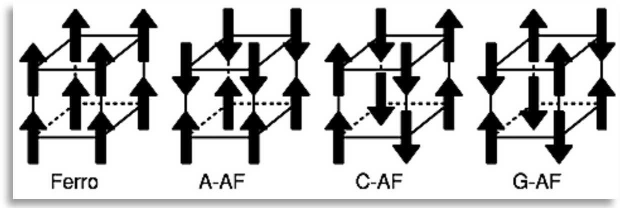


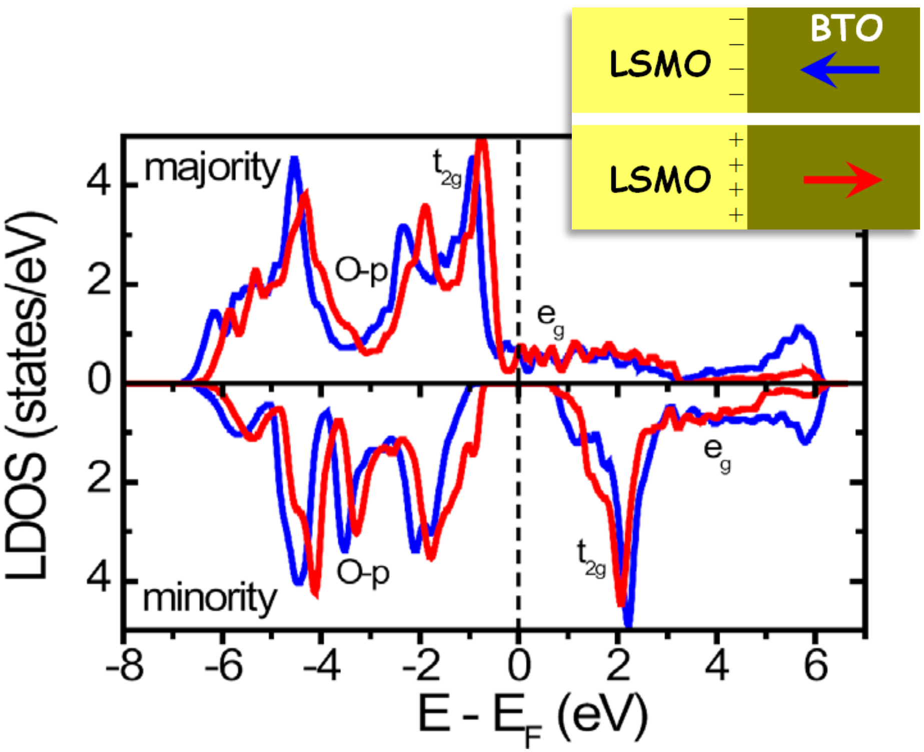
$BaTiO_3$

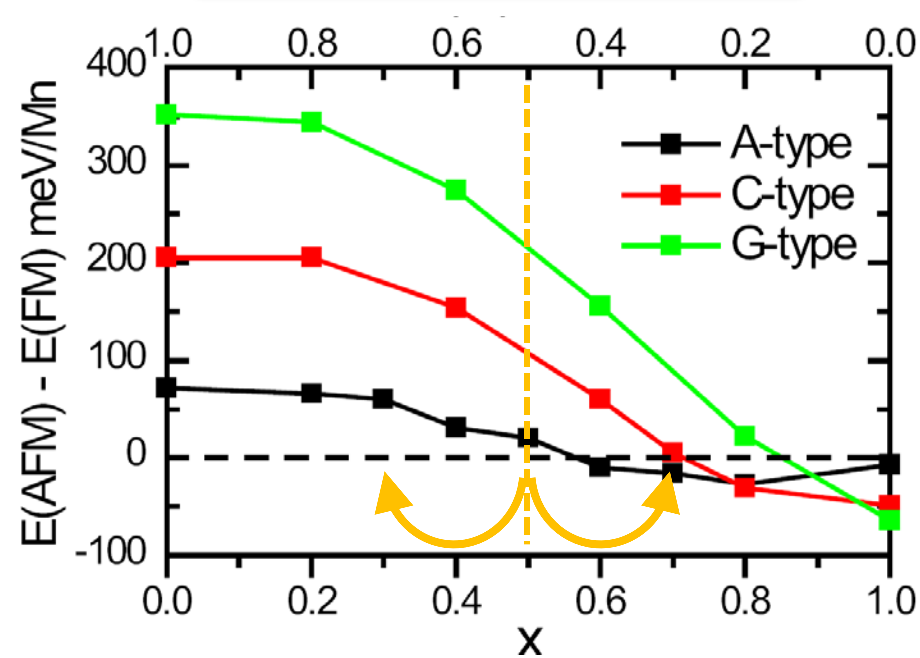
- Transition from FM to AFM order near x ~ 0.5

Interface Electronic Structure of $La_{0.5}Sr_{0.5}MnO_3$

Burton et al., PRB 80, 174406 (2009)



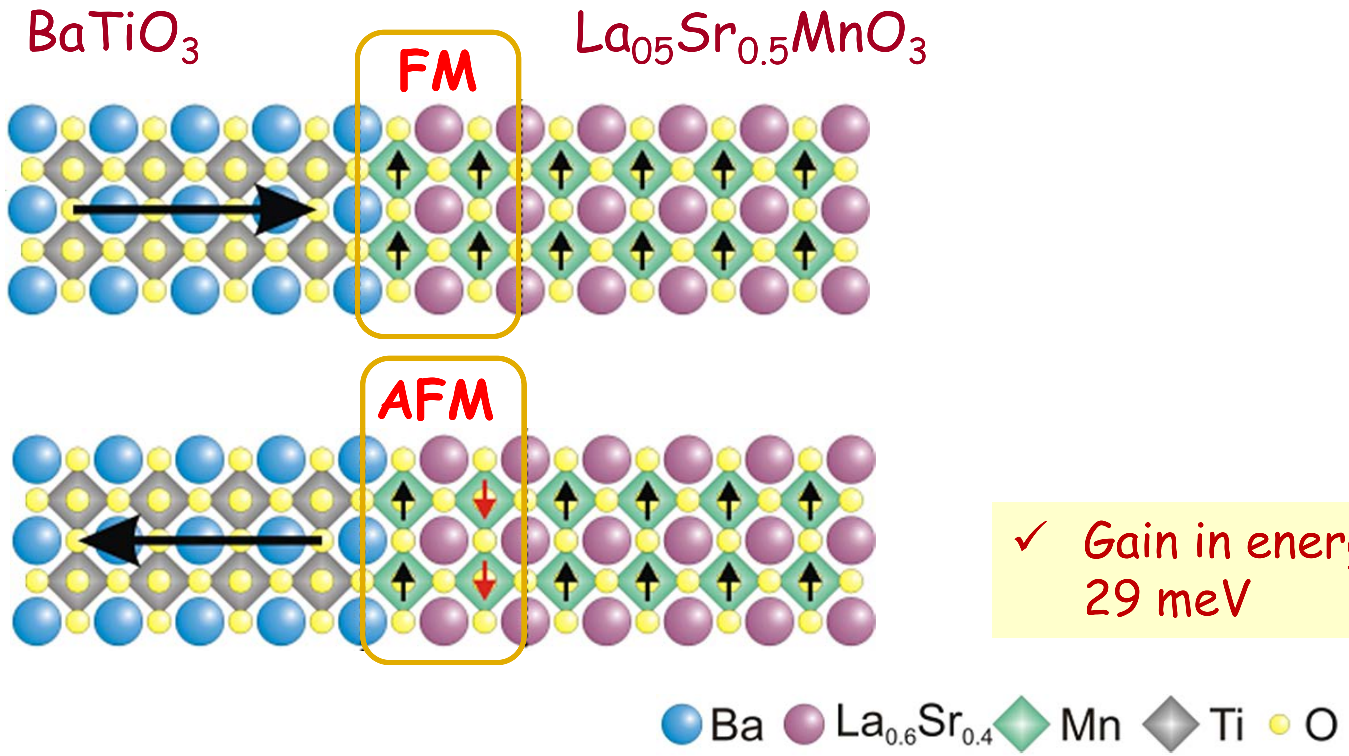




- Accumulation or depletion of electronic charge controls majority-spin e_g state population and thus magnetic order

Interface Magnetic Reconstruction

Burton et al., PRB 80, 174406 (2009)

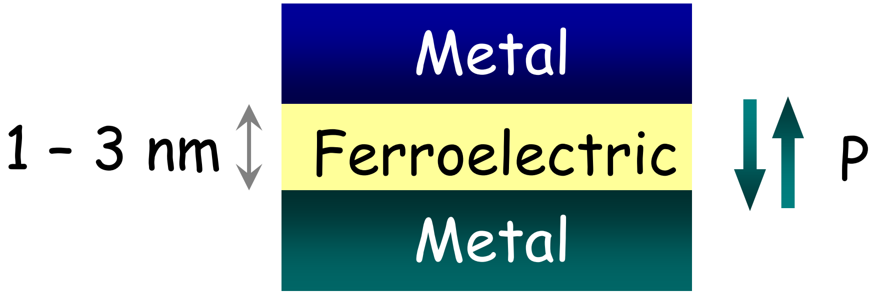


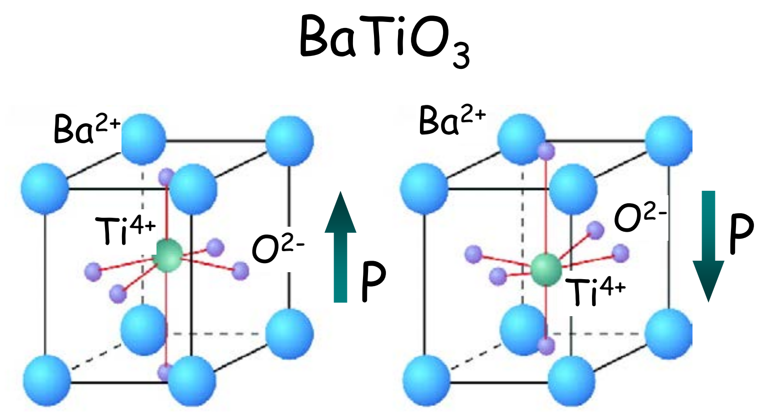
✓ Gain in energy: 29 meV

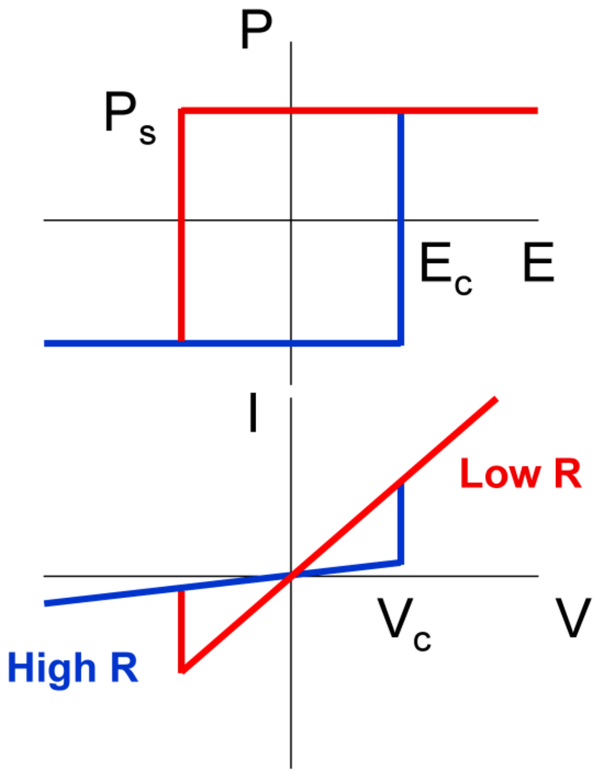
- Change in the local magnetic order from FM to AFM induced by ferroelectric polarization reversal

Ferroelectric Tunnel Junction (FTJ)

Tsymbal et al., Science 313, 181 (2006); MRS Bulletin 37, 138 (2012)

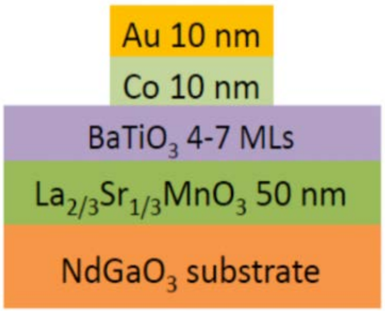




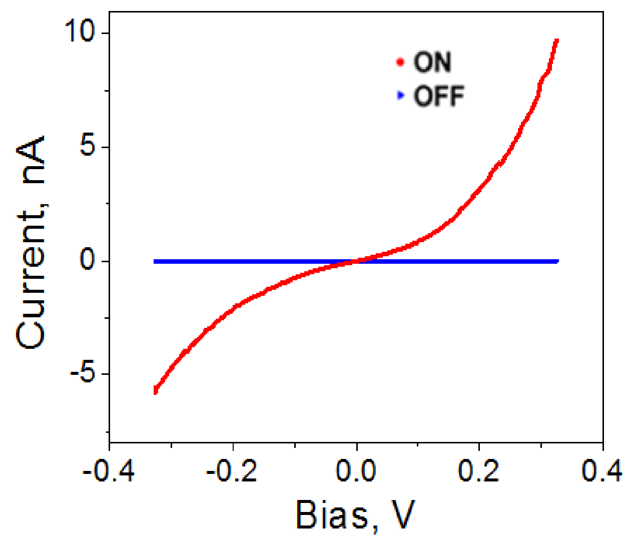


- Tunneling Electroresistance (TER) effect: resistive switching of FTJ at coercive electric field

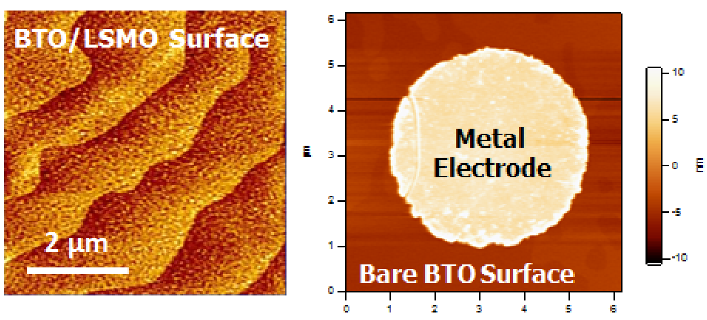
Reliable FTJ Devices



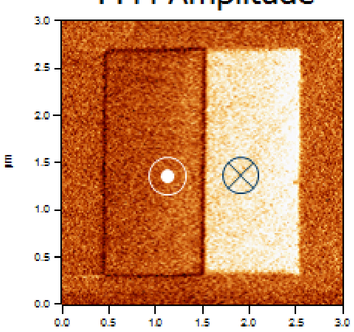
Kim et al.,
Nano Lett. 12, 5697 (2012)



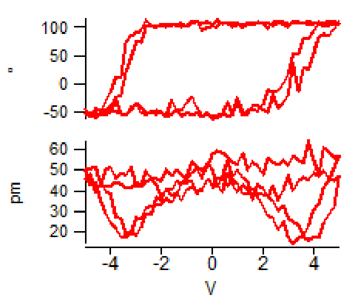
AFM Topography



PFM Amplitude



PFM Hysteresis



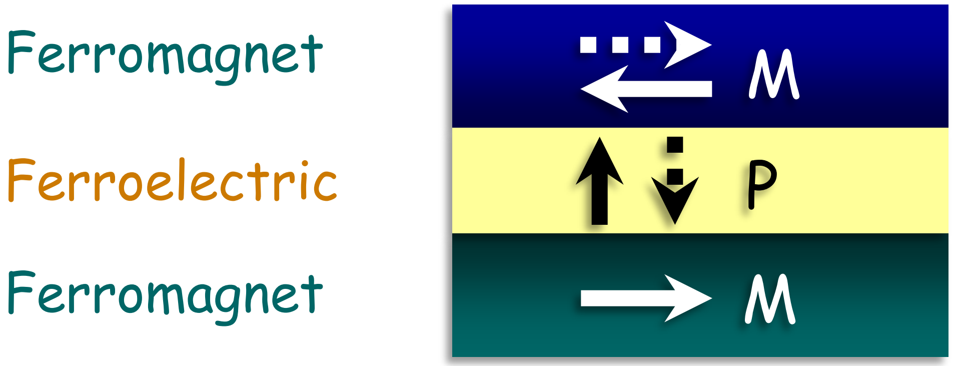
- ✓ FTJs with ON/OFF resistance ratio exceeding 10^3 at room temperature

Multiferroic Tunnel Junction (MFTJ)

Zhuravlev et al, APL 87, 222114 (2005); PRB 81, 104419 (2010)

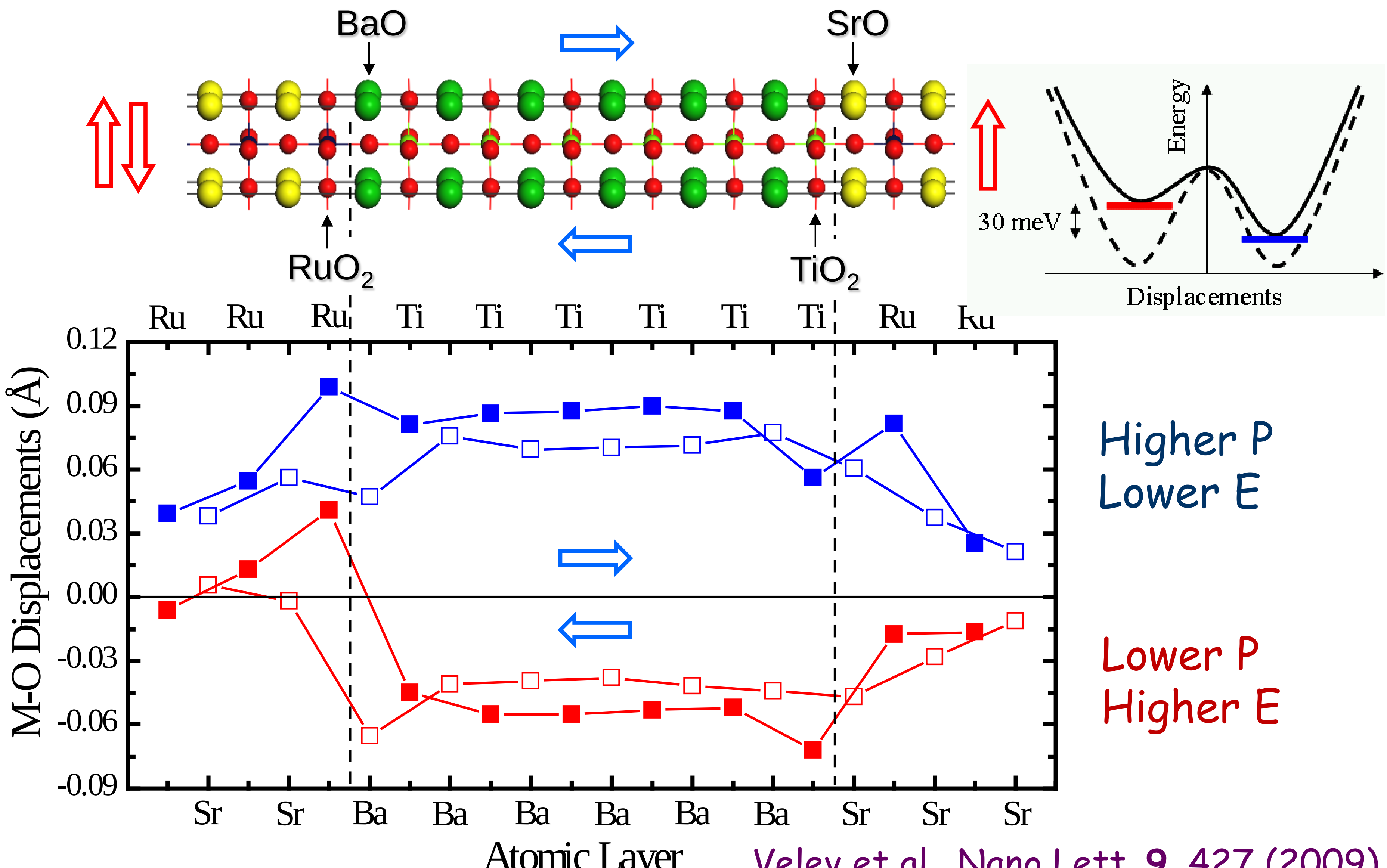
- Magnetic tunnel junction with a ferroelectric barrier

MFTJ = MTJ + FTJ



- MFTJ combines TMR with TER making a four-state resistive device
- Effect of ferroelectric polarization on transport spin polarization
- Multifunctional properties

$SrRuO_3/BaTiO_3/SrRuO_3$ Multiferroic Tunnel Junction (MFTJ)



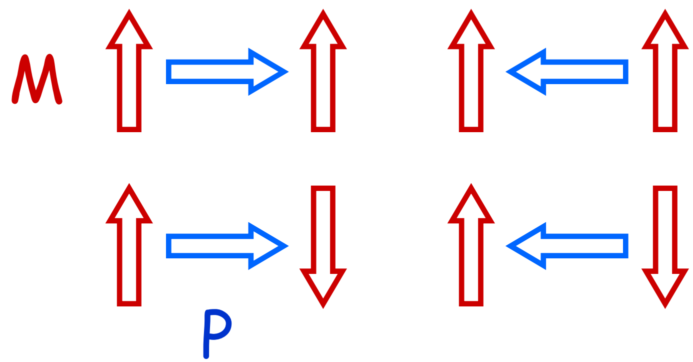
Higher P
Lower E

Lower P
Higher E

Velev et al., Nano Lett. **9**, 427 (2009)

Co-existence of TER and TMR

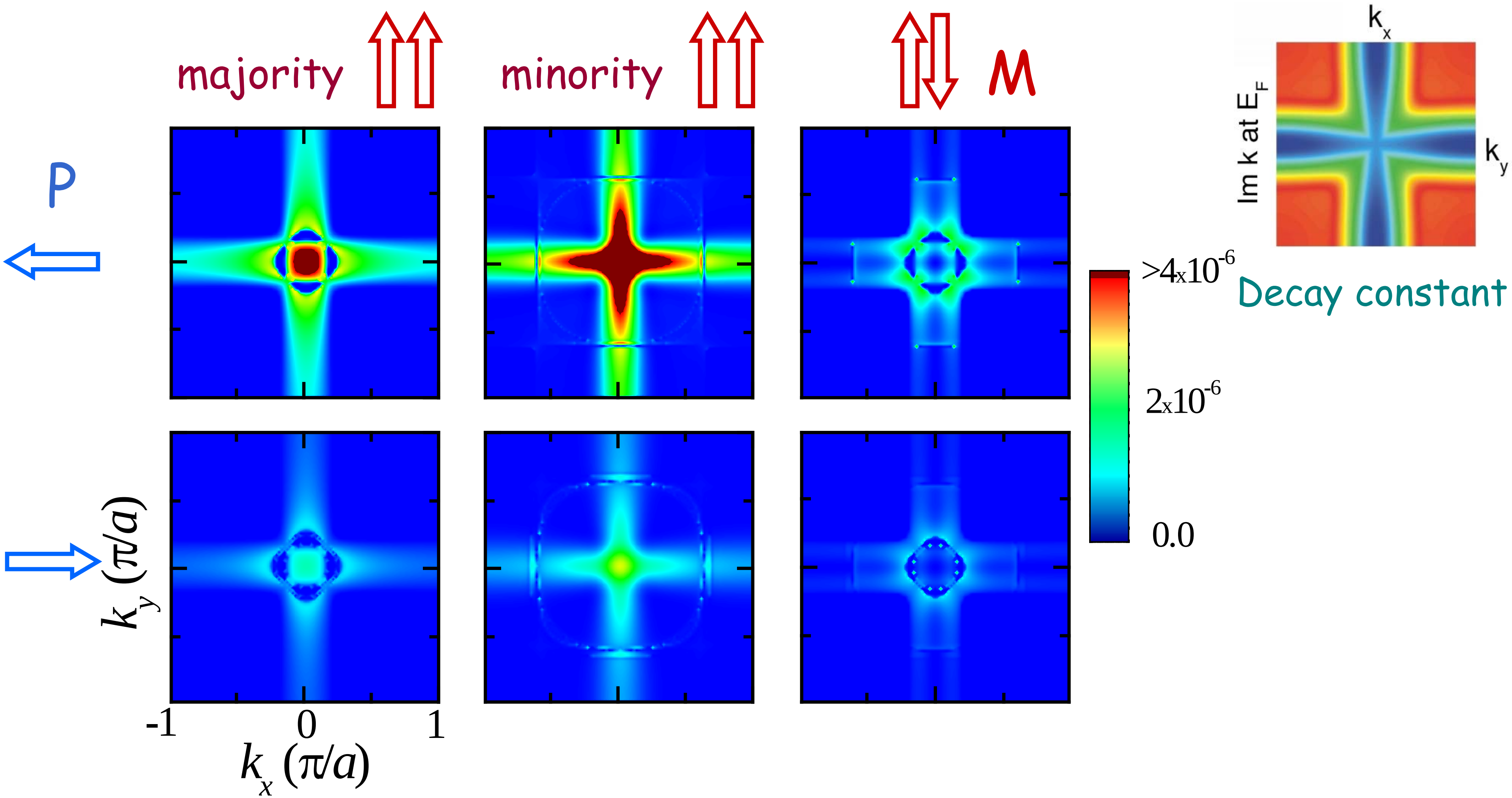
Velev et al., Nano Lett. **9**, 427 (2009)



Four conductance states in $SrRuO_3/BaTiO_3/SrRuO_3$ MFTJ

G ($10^{-7}e^2/h$)	↑↑	↑↓	**TMR (%)**
→	3.76	0.83	350
←	11.82	1.69	590
TER (%)	210	100	

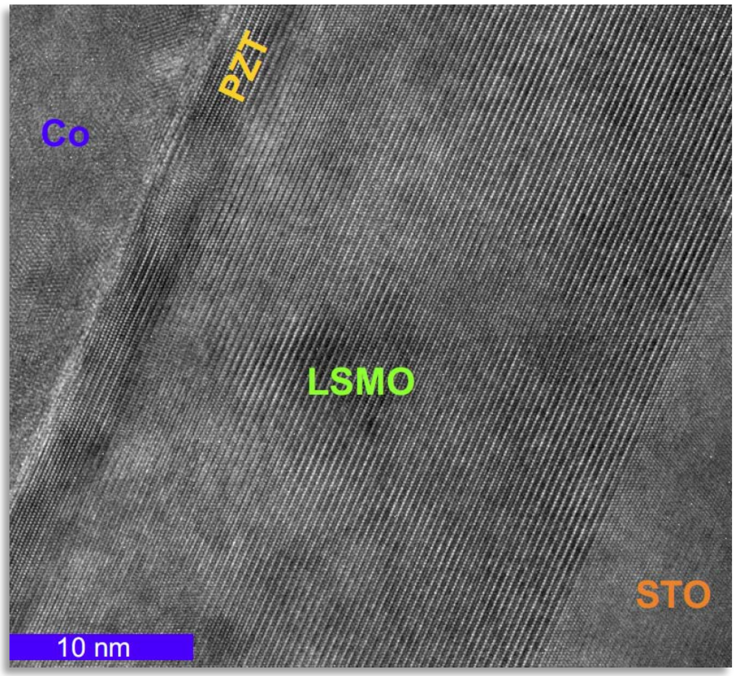
$K_{||}$-resolved Transmission

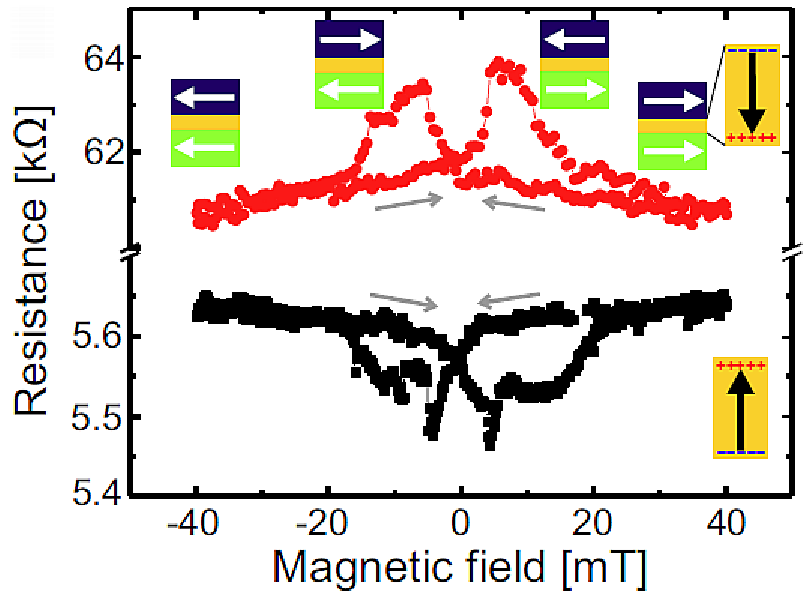


- TMR – selection rules for electronic transmission
- TER – attenuation constant in the barrier

Ferroelectric Control of Spin Polarization

Pantel et al., Nature Mater. 11, 289 (2010)

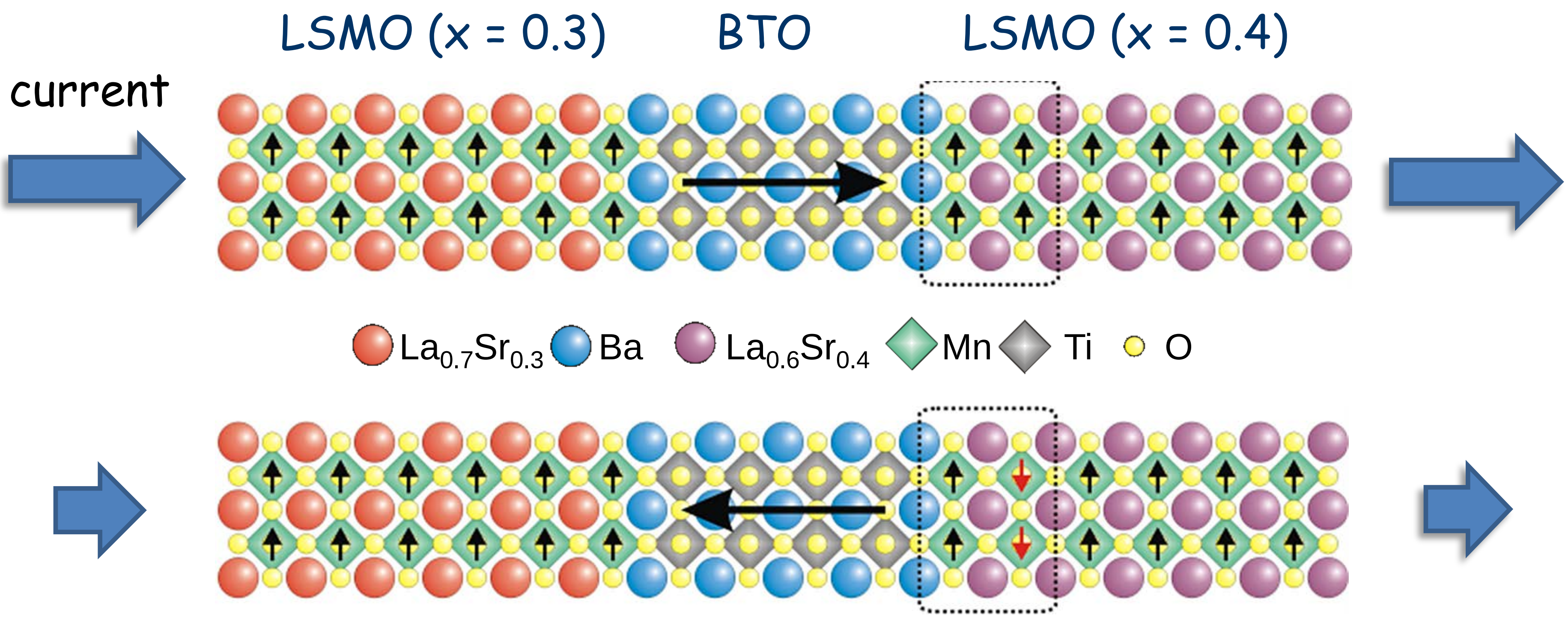




- Coexistence of TMR and TER
- Change of TMR sign with ferroelectric polarization reversal

Magnetoelectrically Driven TER Effect

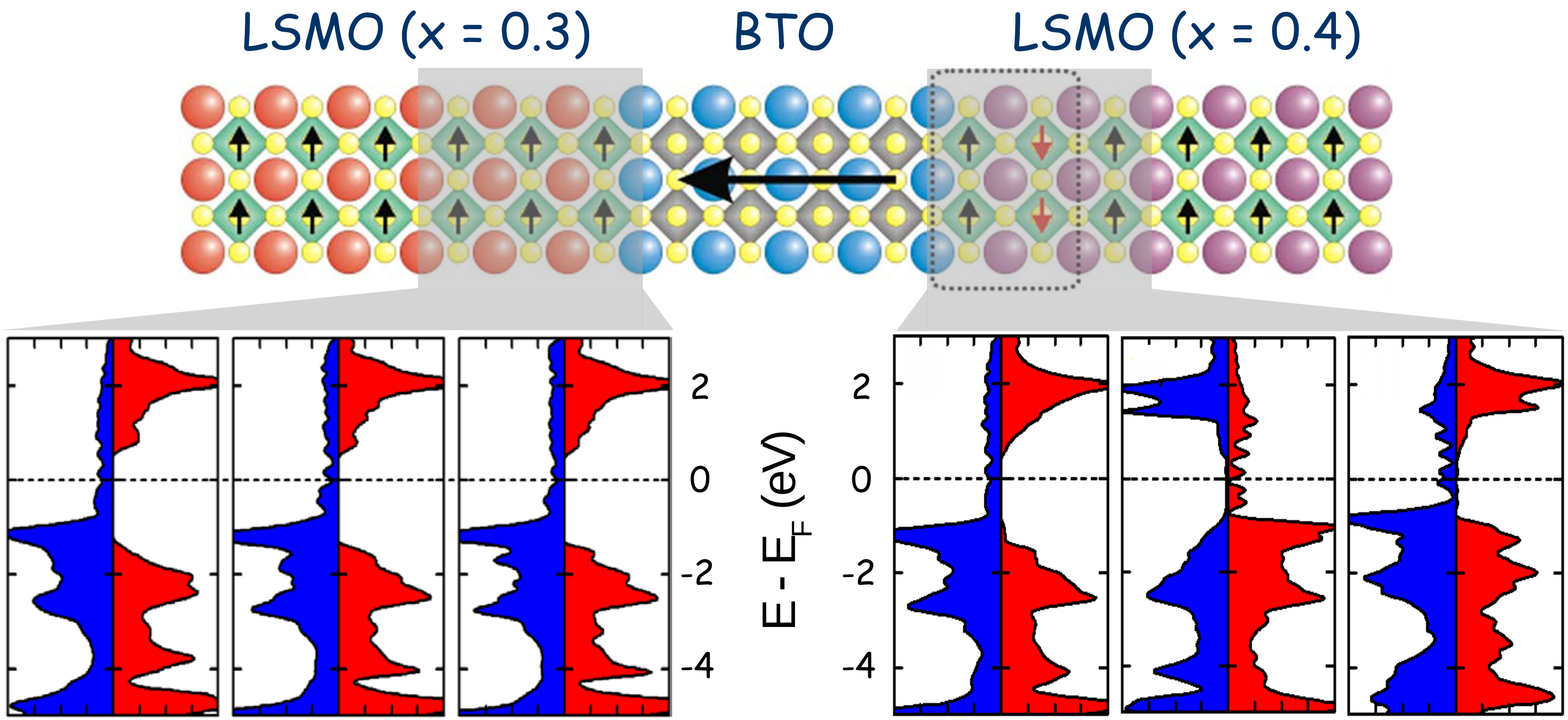
Burton et al., PRL 106, 157203 (2011)



- More than 20 fold change in resistance !

Electrically-Controlled Atomic Spin Valve

Burton et al., PRL 106, 157203 (2011)



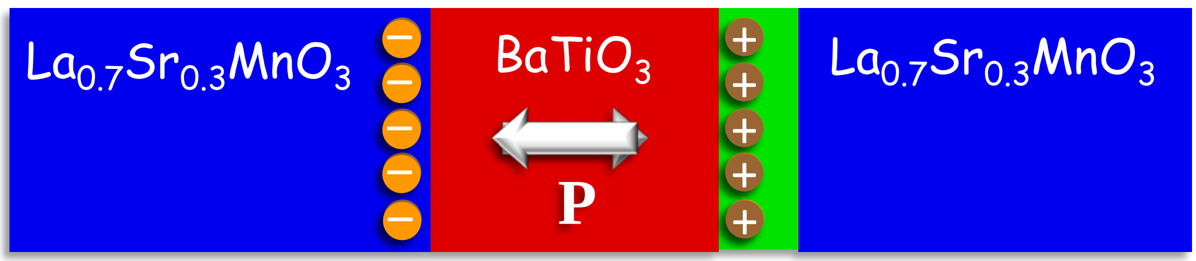
Layer and spin resolved density of states

- Polarization driven magnetic moment reversal acts as a spin valve enhancing dramatically resistance for antiparallel state

Experiments

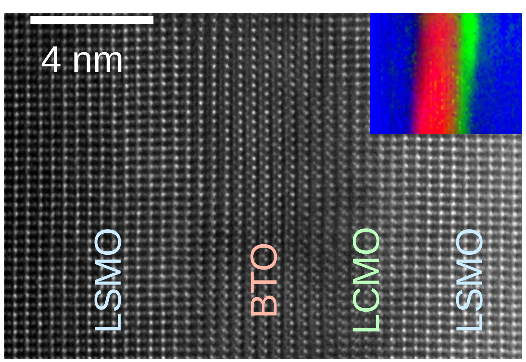
Yin et al., Nature Mater. 12, 397 (2013)

AFM Insulator
FM Metal

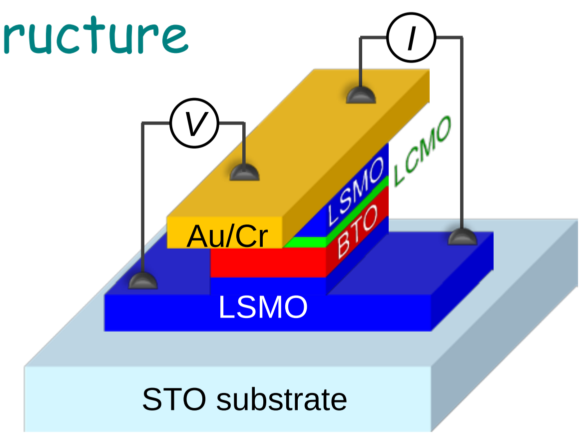


$La_{0.5}Ca_{0.5}MnO_3$
~1nm

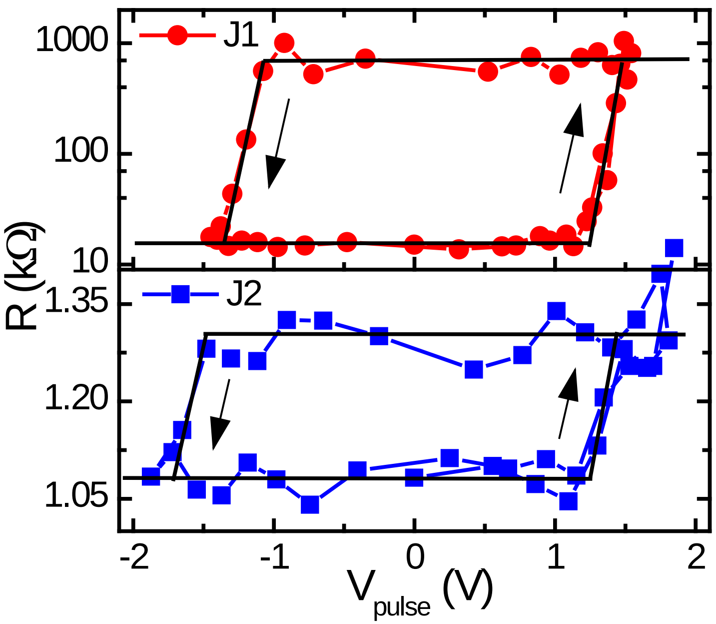
HRTEM & EELS



Device structure



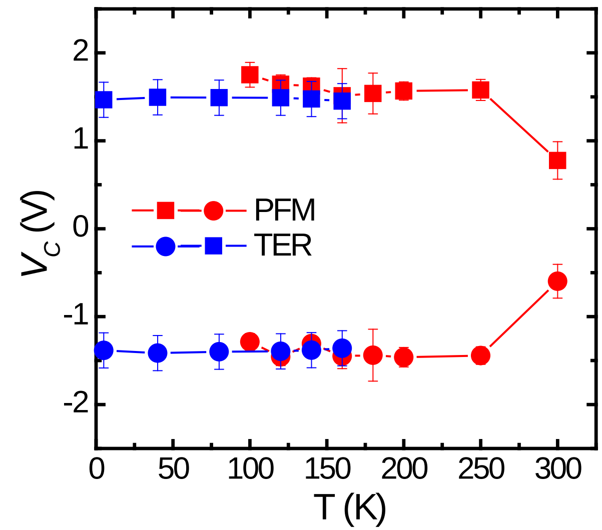
Experimental Result: ~10,000% TER

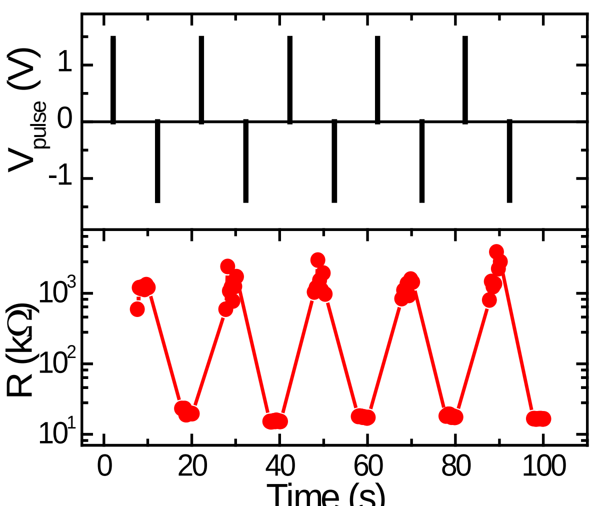


with LCMO interlayer
TER ~ 10,000%

without LCMO interlayer
TER ~ 40%

PFM data



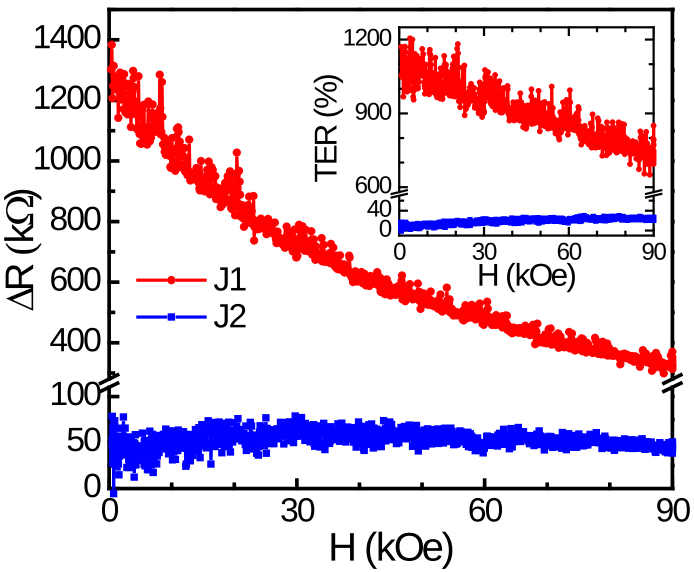


- TER associated with FE polarization switching
- Effect is reversed when LCMO is deposited on bottom interface
- Reproducible switching behavior

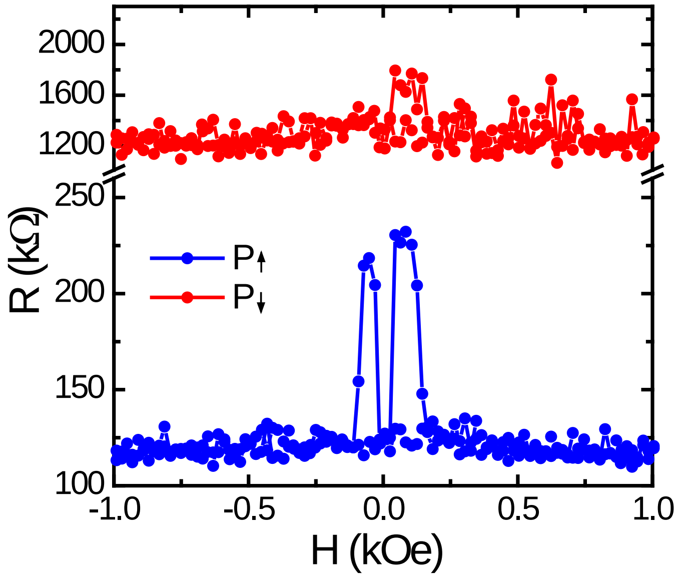
Magnetoresistance

Yin et al., Nature Mater. 12, 397 (2013)

High field data

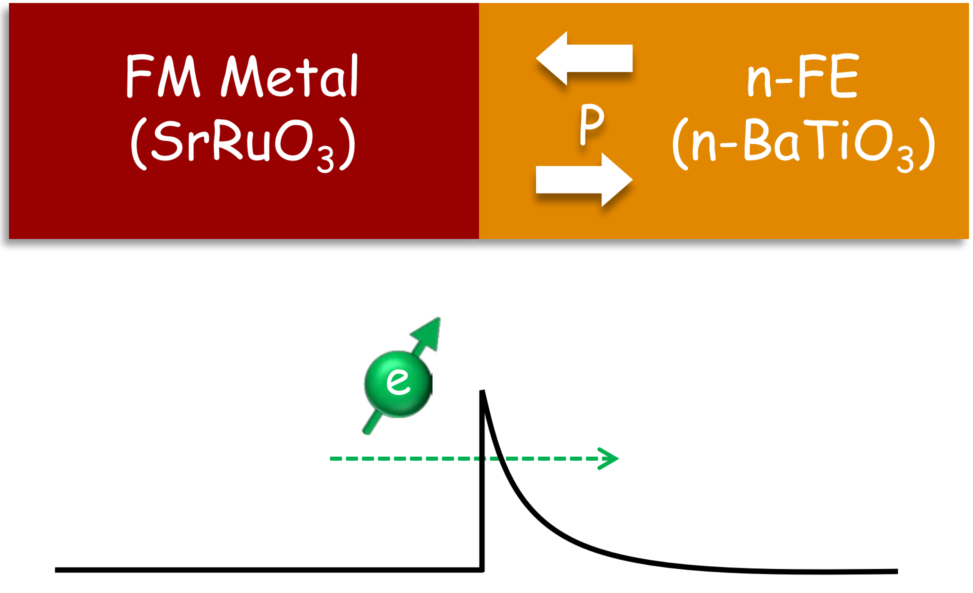


TMR



- Consistent with the magnetic reconstruction mechanism

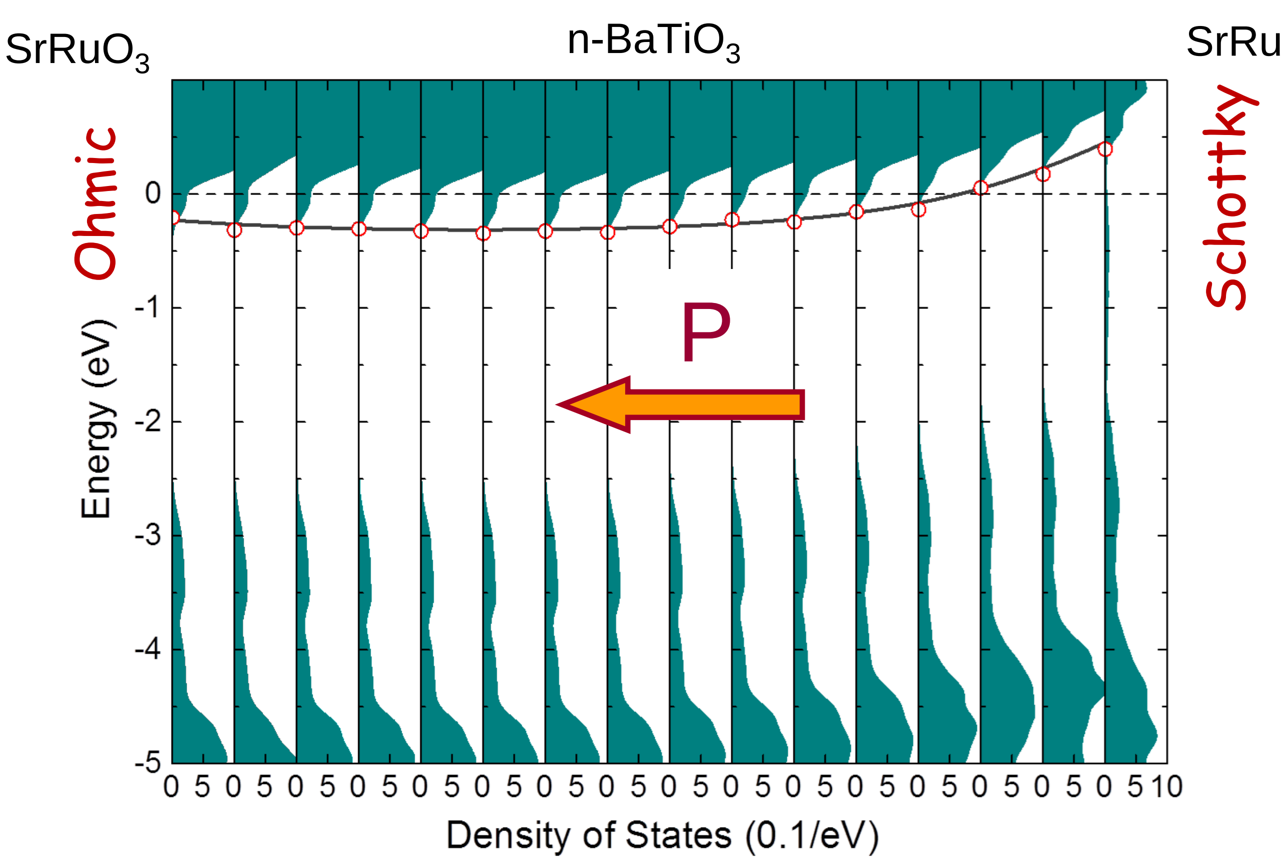
Metal/n-Ferroelectric Heterojunction

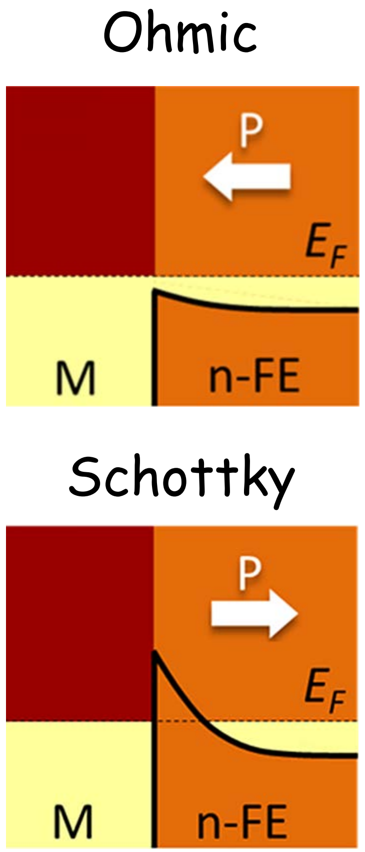


- Normal Metal:
  Ferroelectric polarization dependent resistance
- Ferromagnetic Metal:
  Ferroelectric polarization dependent spin-polarization

Band Bending across n-$BaTiO_3$

Liu et al., PRB 88, 165139 (2013)





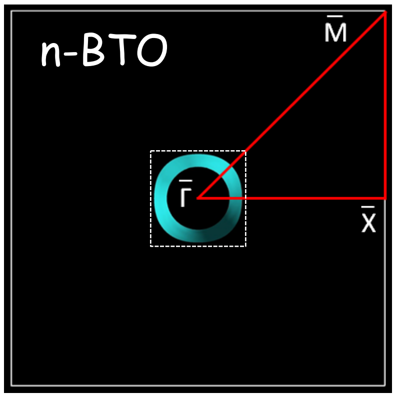
- Shottky or Ohmic contact depending on polarization orientation

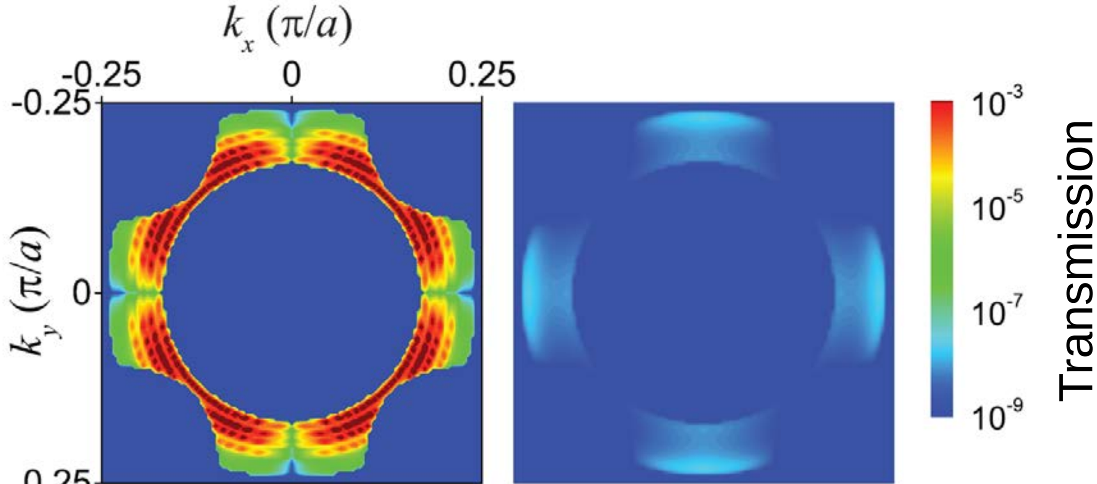
Transmission

Liu et al., PRB 88, 165139 (2013)

Fermi surfaces

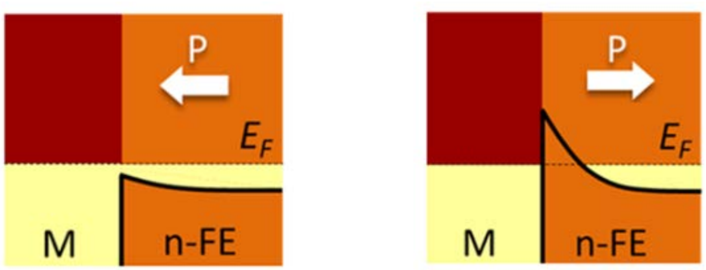
(view along z-direction)





RA = 5.5×10^2 Ωμm² RA = 3.78×10^7 Ωμm²

Ohmic Schottky

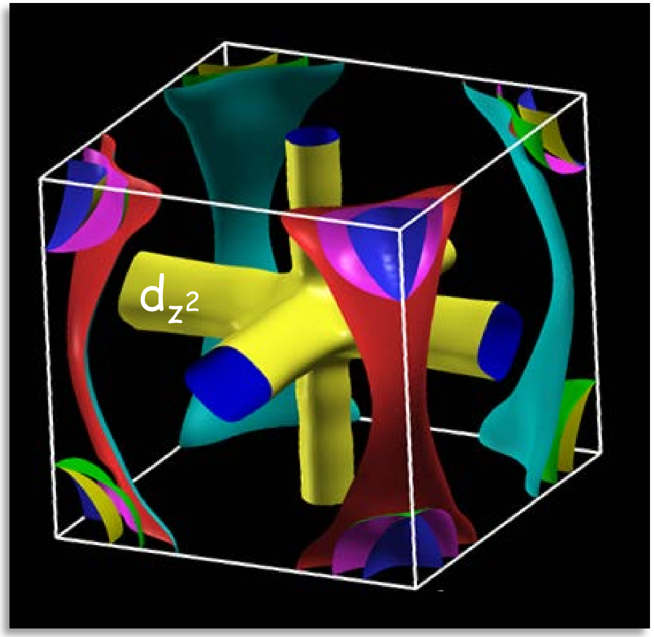


- 10^5 change in the interface resistance by switching ferroelectric polarization

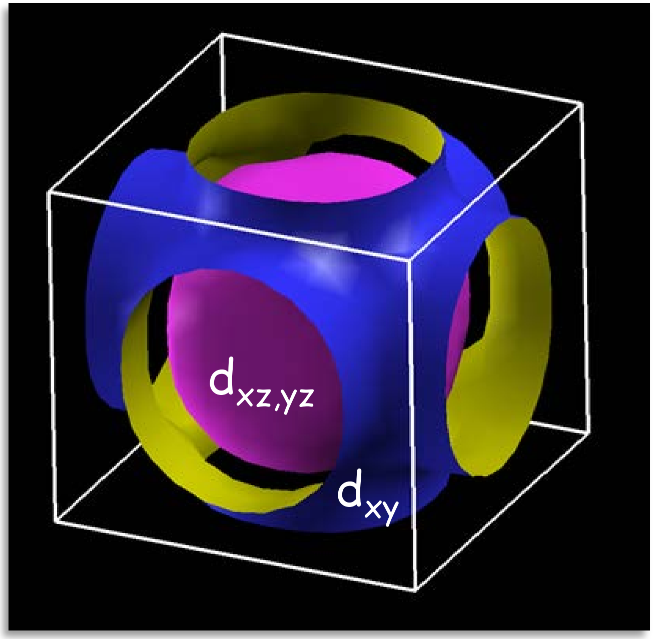
Fermi Surface of $SrRuO_3$

- $SrRuO_3$ is a ferromagnet below 160K
- Spin-dependent electric band structure ($1.2\mu_B$/f.u.)

Majority spin



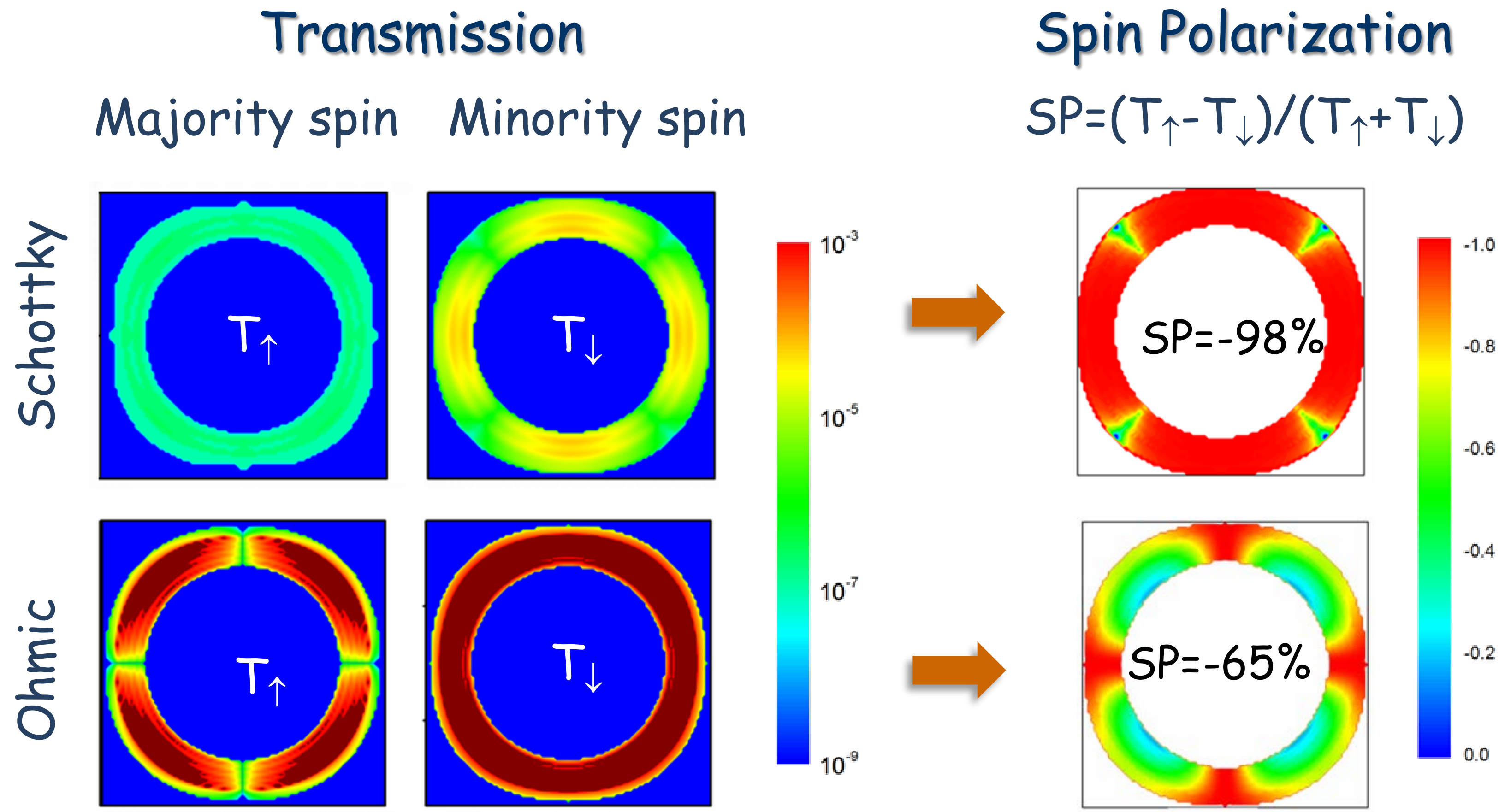
Minority spin



- Spin-dependent transmission is expected across $SrRuO_3$/n-$BaTiO_3$ interface

Transport Spin-Polarization

Liu et al., PRL 114, 046601 (2015)

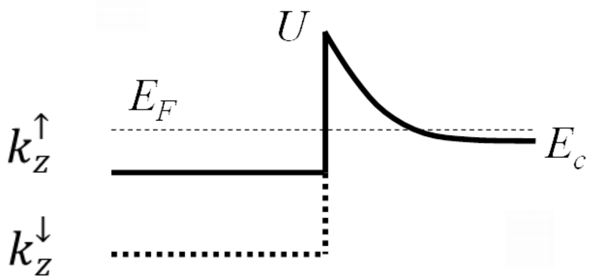


- Significant change in the transport spin-polarization with ferroelectric polarization reversal

Origin of Spin-Polarization Change

Liu et al., PRL 114, 046601 (2015)

A simple model:
Tunneling across a Schottky barrier

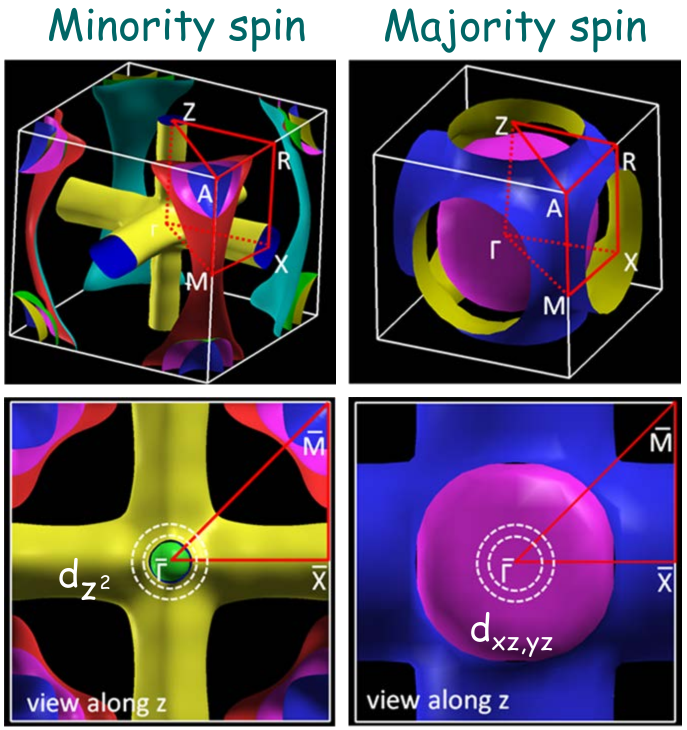


$$P_s = \left(\frac{k_z^\uparrow - k_z^\downarrow}{k_z^\uparrow + k_z^\downarrow}\right)\left(\frac{\kappa^2 - \gamma^2 k_z^\uparrow k_z^\downarrow}{\kappa^2 + \gamma^2 k_z^\uparrow k_z^\downarrow}\right)$$

κ - attenuation constant

similar to Slonczewski, PRB 39, 6995 (1989)

Fermi surface of $SrRuO_3$



$k_z^\uparrow \approx 0.079$ Å^{-1} $k_z^\downarrow \approx 0.634$ Å^{-1}

- Negative spin-polarization due to large minority-spin Fermi wave vector
- Change in the spin-polarization with ferroelectric polarization switching due to change in the barrier height
- Change in sign of the spin polarization for small electron doping

Conclusions

- Ferroelectric polarization provides a new degree of freedom to control materials properties relevant to spintronics
  - Interface magnetization
  - Interface magnetic order
  - Magnetocrystalline surface anisotropy
  - Electron and spin tunneling
  - Spin injection
- Switchable ferroelectric polarization at the interface with a magnetic metal forms a non-volatile spin-dependent state interesting for device applications

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