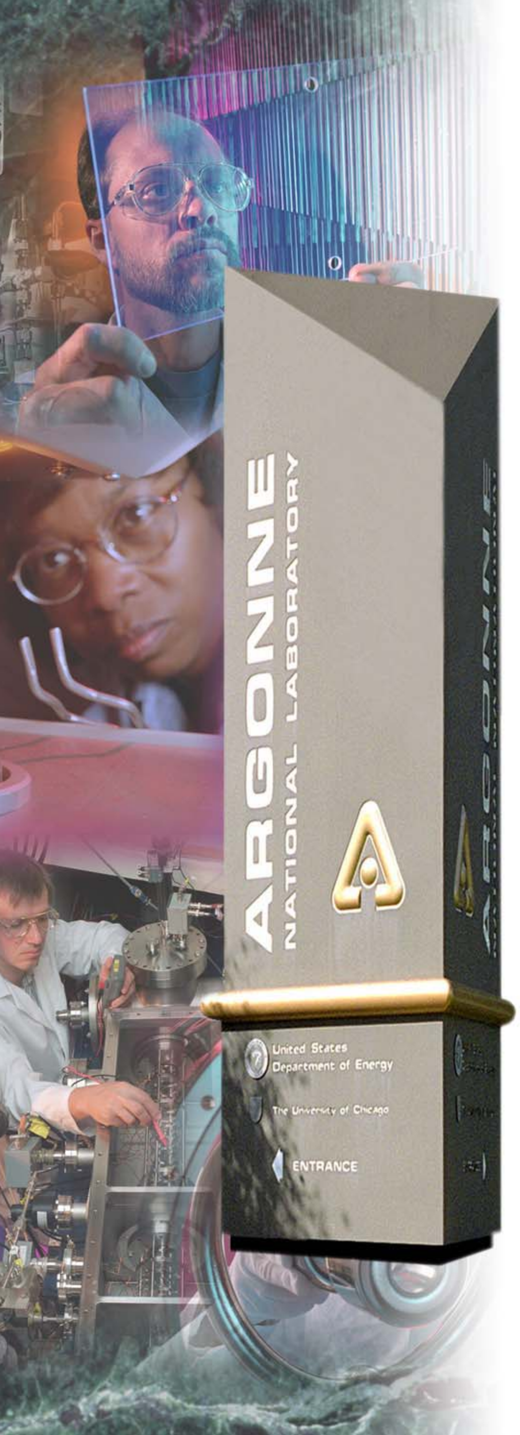

Surface Magnetic and Electronic Properties of Layered Manganite Single Crystals

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* Materials Science Division # Advanced Photon Source

Argonne National Laboratory

Office of Science
U.S. Department of Energy

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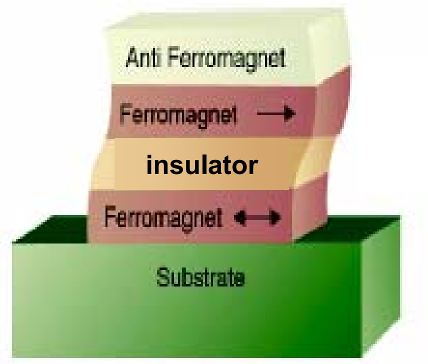
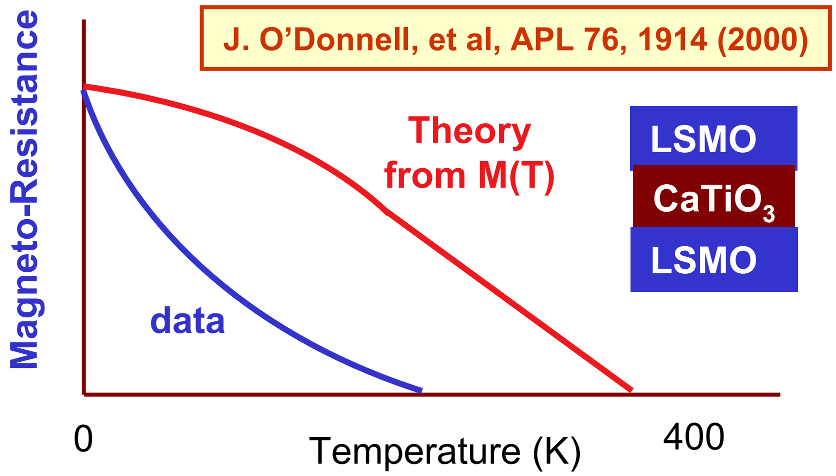
Spintronics

Concept: use electron spin (as well as the electronics' counterpart - charge) to process and transmit information



- **High degree of spin polarization (at surfaces)**
- **Transmission across interfaces without depolarization**

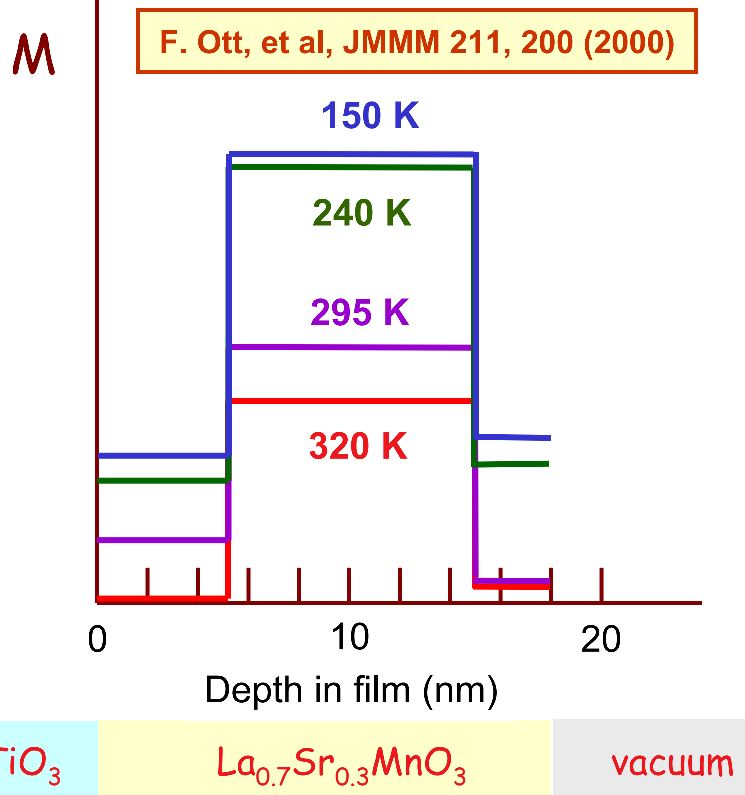
Unfortunately,
this magneto-resistance
can decrease dramatically
at higher temperatures



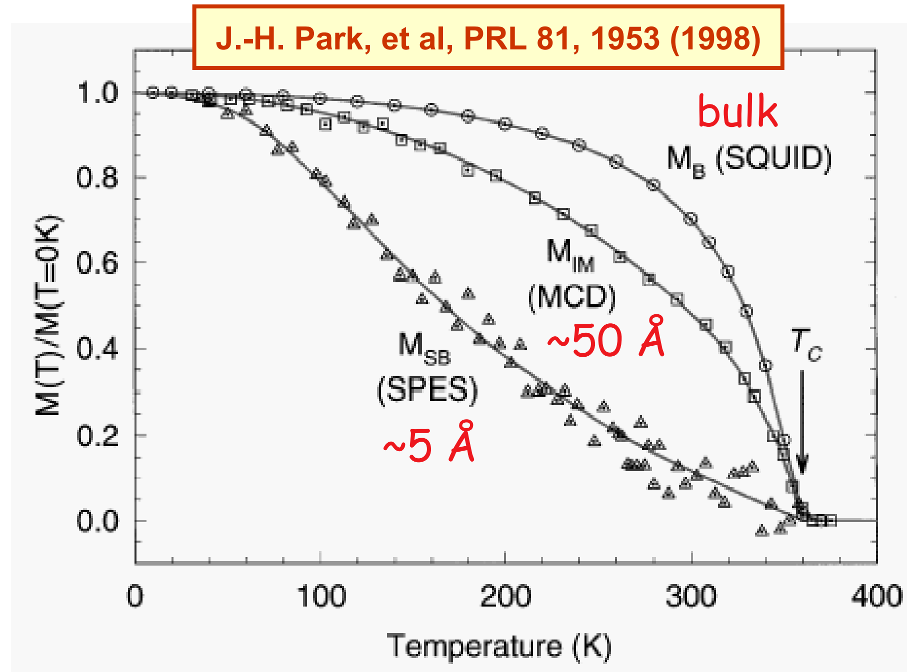
Possible reasons: Spin-flip scattering at interfaces; **Loss of surface spin polarization**

Surface Spin Polarization

Polarized neutron reflectometry



Magnetic 'dead' layers at both interfaces



The spin polarization at interfaces in all studied half-metallic oxides decreases much faster than M(T)
J.D.M. Coey and C.L. Chien, *MRS Bulletin*, 720 (2003)

Why does spin polarization decrease at surfaces?

Why Does Spin Polarization Decrease at Surfaces?

Chemical disorder
- loss of oxygen
- cation segregation

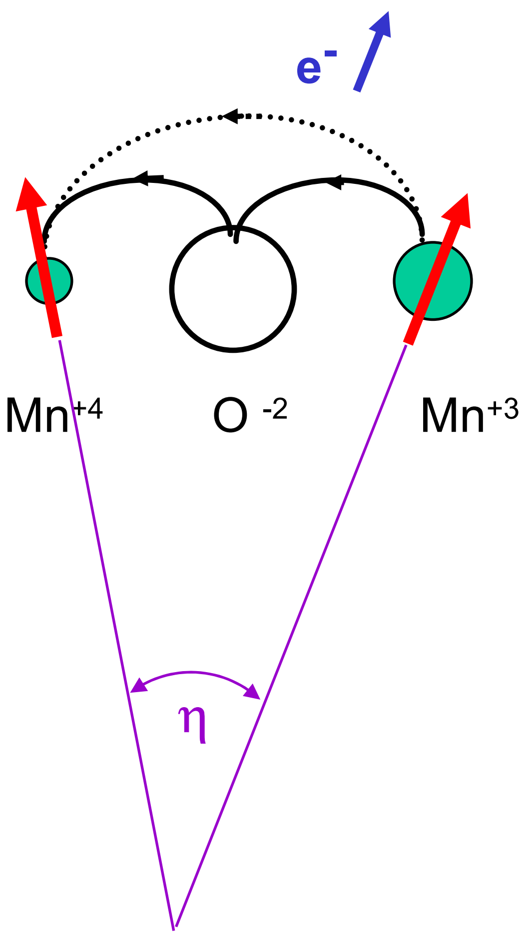
Physical disorder
- roughness
- surface reconstruction, strain

To evaluate these, we need to understand the mechanism of FM spin polarization

Doped Manganites: FM and Conduction by Double Exchange

In $La_{1-x}Sr_xMnO_3$, there are x ions of Mn^{+4} and 1-x ions of Mn^{+3}

This leads to the possibility of conductivity by double exchange



Hund's rule favors ↗ aligned with ↗

Hopping leads to FM exchange

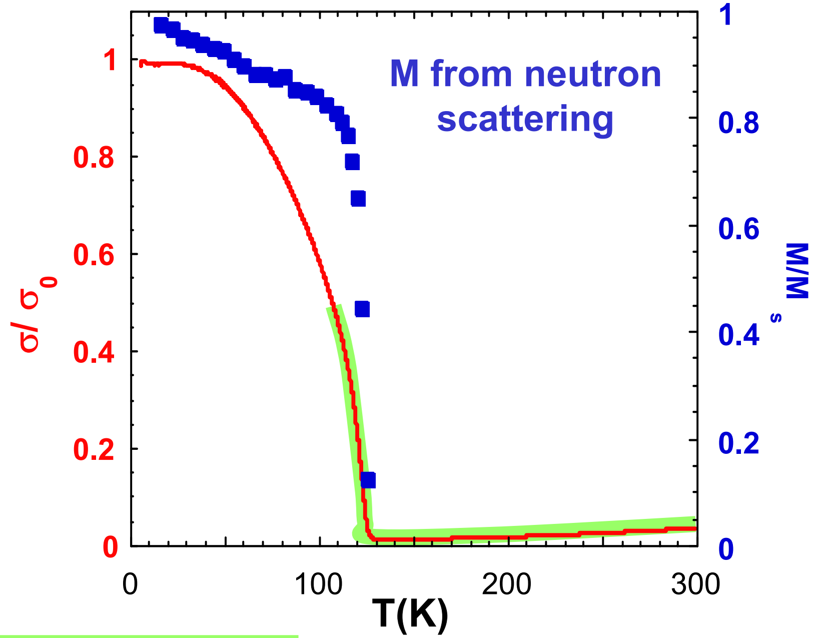
alignment of ↖ and ↗ lowers the total energy by increasing hopping rate

FM leads to higher conductivity~ $\cos^2(\eta/2)$

Doped Manganites Exhibit Double Exchange

Spontaneous magnetization below T_C is symbiotic with a metal insulator transition (MIT)

This suggests that conductivity and magnetization are a result of double-exchange



double-exchange *cannot* explain high temperature conductivity in a simple way, since for a paramagnet

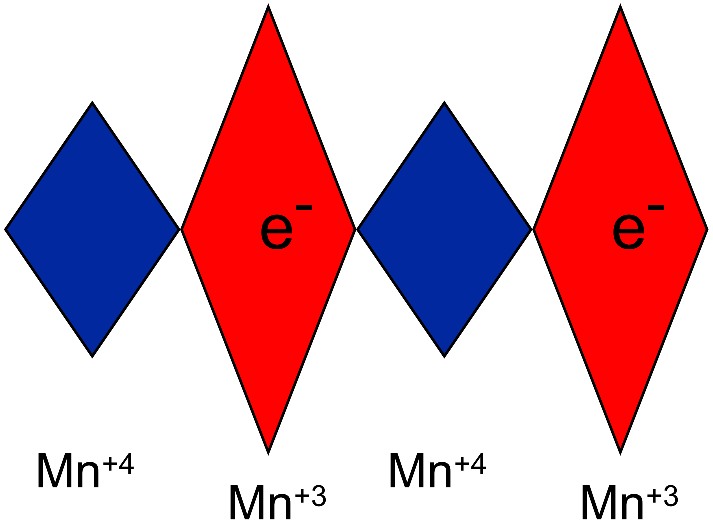
$$\sigma_{DE}/\sigma_o \sim \langle \cos^2(\eta/2) \rangle = 0.5$$

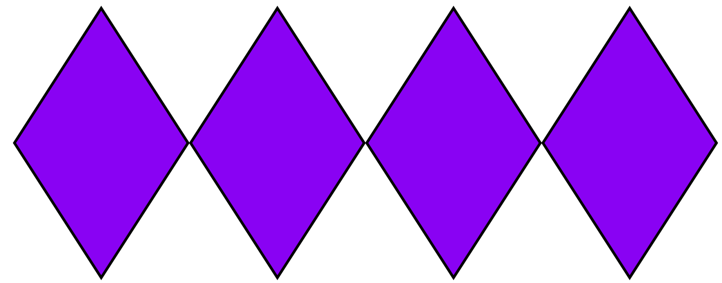
Electron localization mechanism is needed

*A.J. Millis, P.B. Littlewood and B.I. Shraiman, PRL **74**, 5144 (1995)*

Polarons

Suggested that polarons play a role in transport for $T>T_C$





Mn^{+3} is Jahn-Teller active

metallic (mixed-valence) state

Insulator above T_C (localized charges thermally activated)

Metal below T_C (delocalized charges in Bloch waves)

Why Does Spin Polarization Decrease at Surfaces?

Chemical disorder
loss of oxygen
cation segregation

Physical disorder
roughness
surface reconstruction, strain

Electron localization at surface
due to one or more of the above

Tunneling can address the metallic behavior of surfaces through the density of states, but . . .

To profile surface spin polarization, we need x-ray magnetic scattering and absorption

Could the free surface promote Mn^{+3}/Mn^{+4} ordering to better accommodate the strain of Jahn-Teller distortions ?

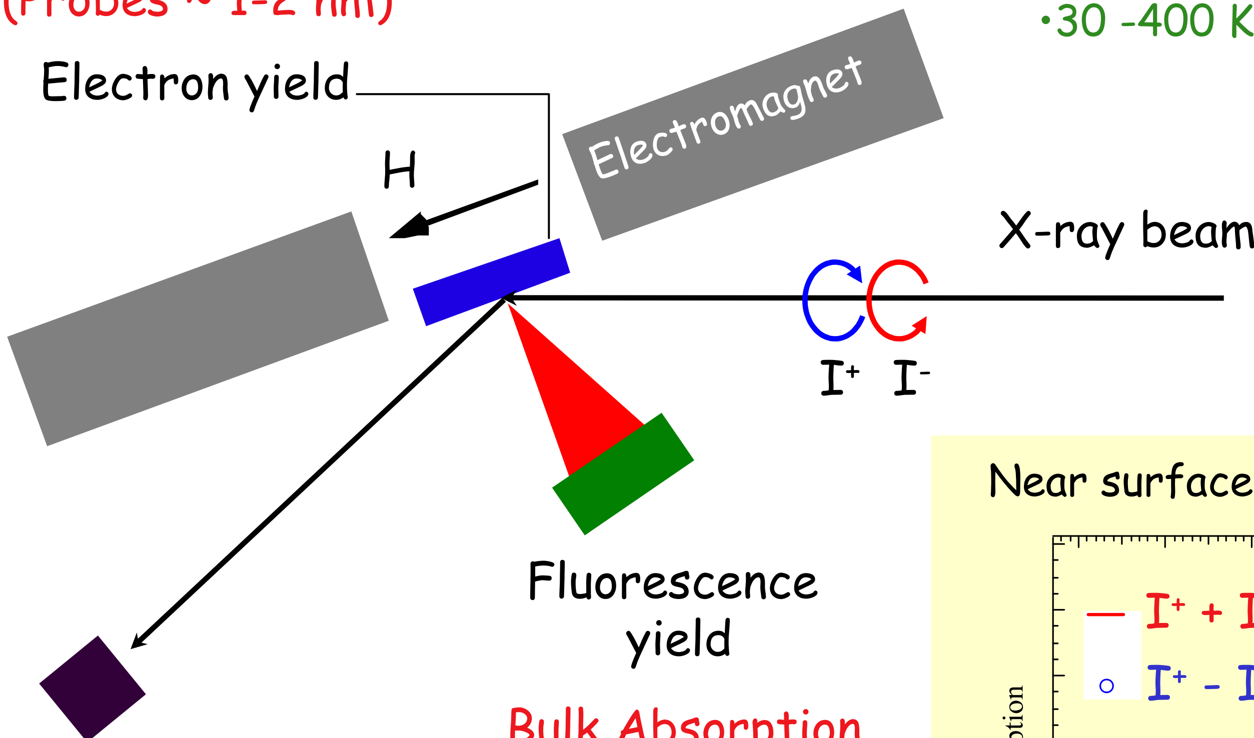
For Mn^{+3}/Mn^{+4} ordering, we need element-selective x-ray scattering and absorption

Fortunately, both are available at APS 4ID-C

X-ray Probes

Near Surface Absorption
(Probes ~ 1-2 nm)

- Applied fields to 7 T
- 30 -400 K sample temperature

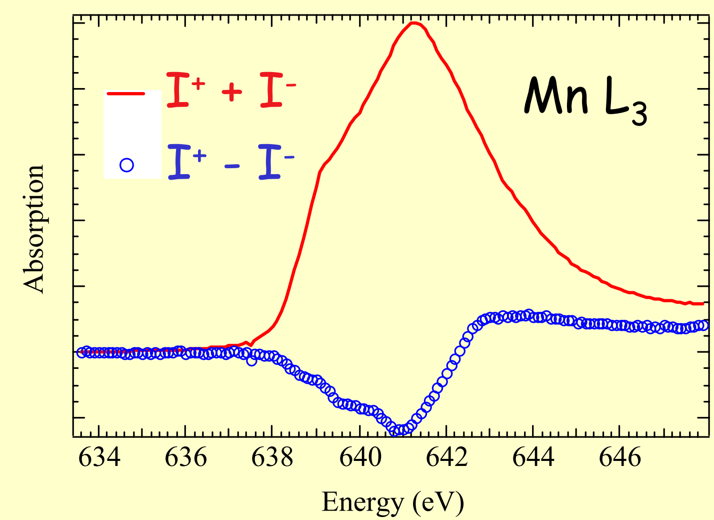


Bulk Absorption
(Probes ~ 20 nm)

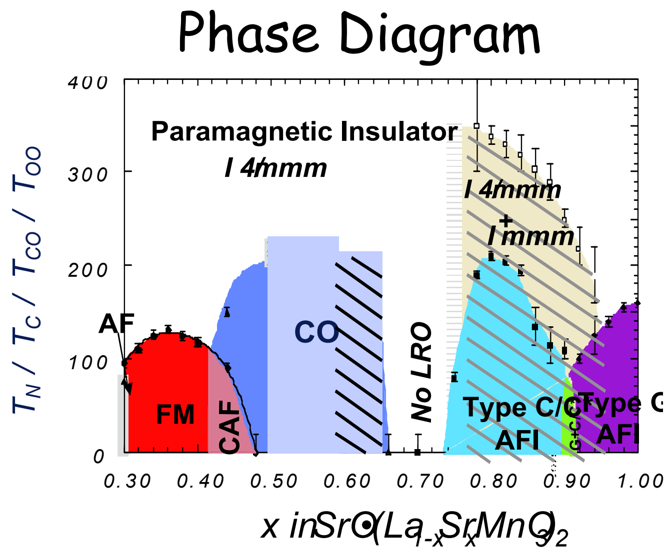
Reflectivity = magnetic resonant scattering

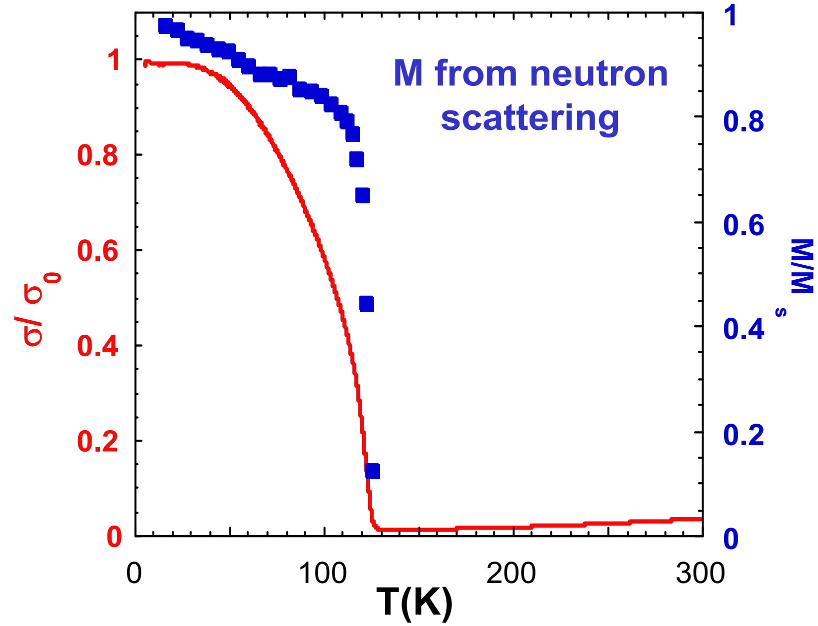
Surface and Interfaces

Near surface Mn L edge absorption

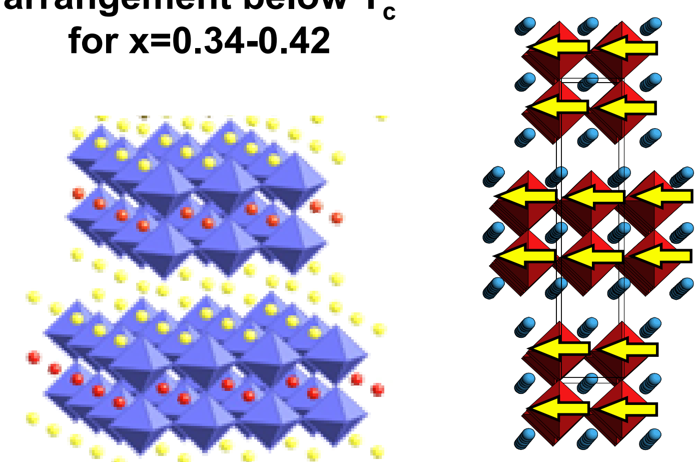


Bilayer Manganites -- $La_{2-2x}Sr_{1+2x}Mn_2O_7$





Structure and spin arrangement below T_c for x=0.34-0.42

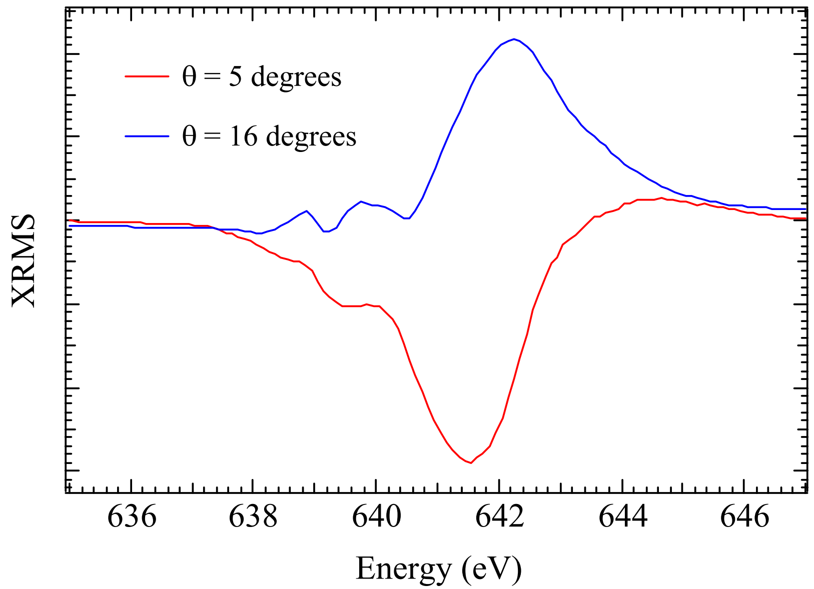


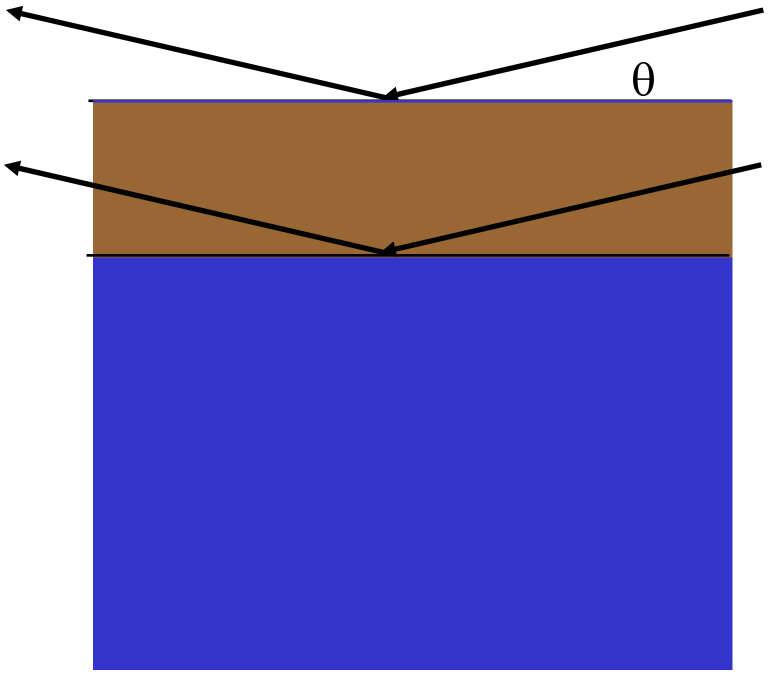
Manganites exhibit high degree of spin polarization in the bulk

What about their surfaces ?

Angle-Dependent X-Ray Resonant Magnetic Scattering

XRMS = Reflection $I^+ - I^-$



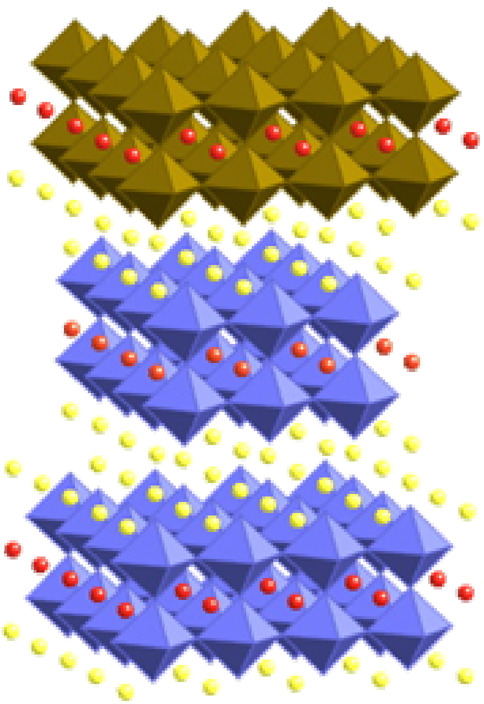


Sign change with angle ⟶ Interference

Chemical and magnetic surfaces are not coincident !

Modeling XRMS

Continuum electrodynamic modeling of XRMS



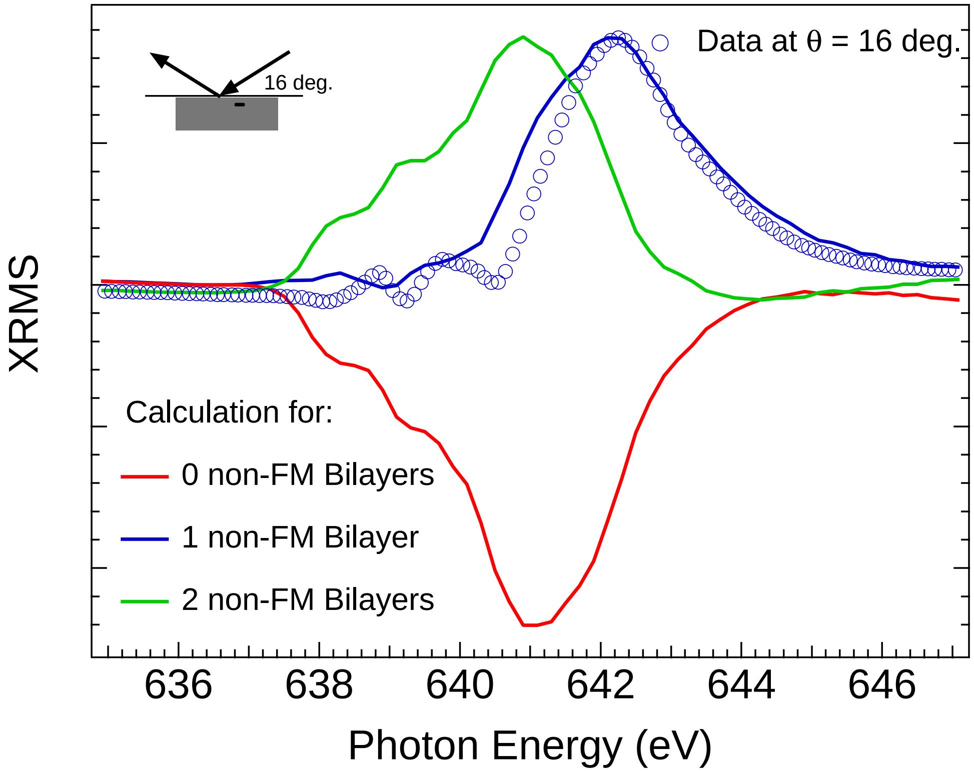
Structure

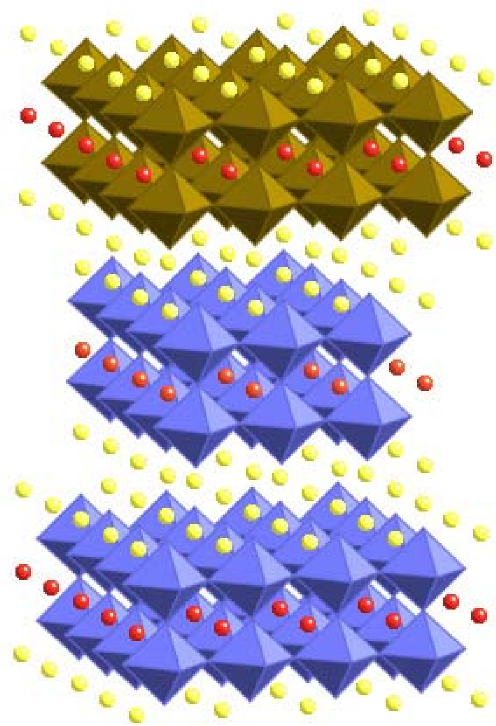
MnO_2 (5Å)
LaSrO (5Å)
MnO_2 (5Å)
LaSrO (5Å)
MnO_2 (5Å)

- Determine dielectric constants from absorption
- Input structure
- Solve Maxwell's equations

Thickness of Non-Magnetic Surface Layer

XRMS is very sensitive to magnetic profile !

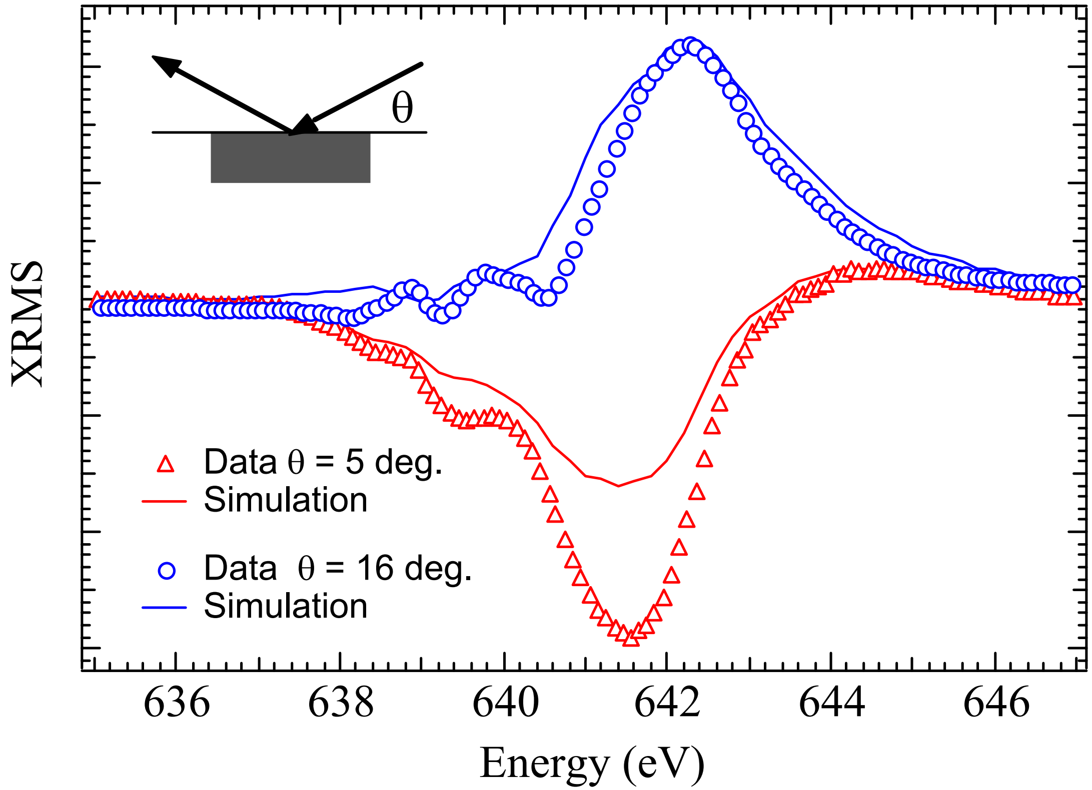




Top bilayer *alone* is non-FM

Non-Magnetic Surface Bilayer

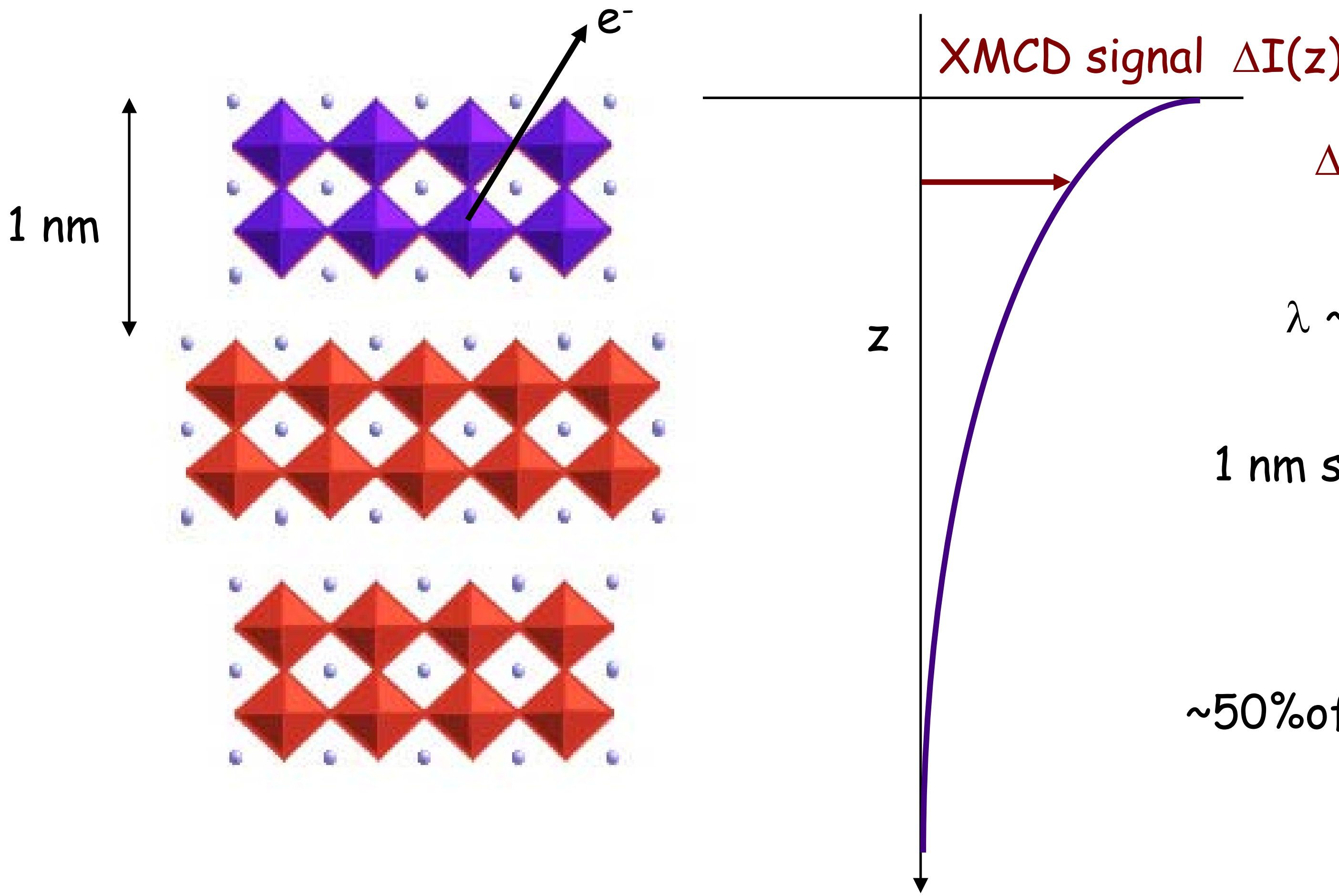
The angular dependence of XRMS is also only consistent with the top bilayer *alone* being magnetically inactive !



What about x-ray absorption ?

Depth Dependent of X-ray Magnetic Circular Dichroism (XMCD)

Depth dependent escape of secondary electrons



$\Delta I \sim e^{-z/\lambda}$

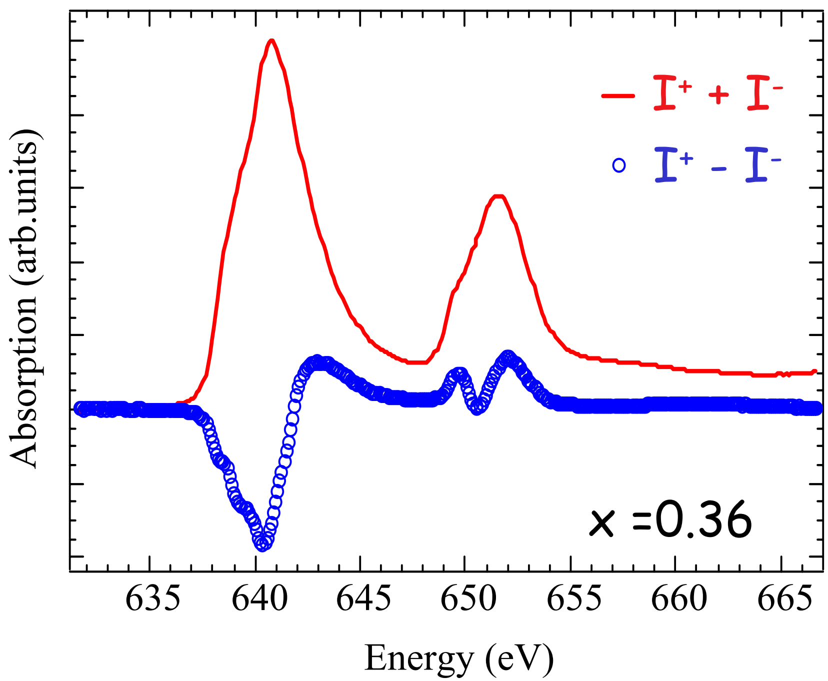
$\lambda \sim 1.5$ nm

1 nm surface layer

~50%of XMCD signal

Surface Magnetization vs. Temperature

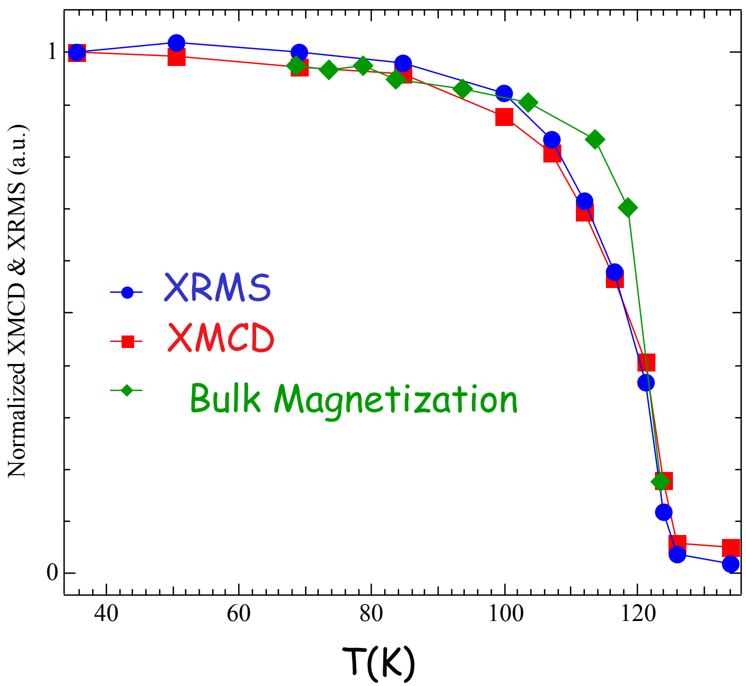
Quantify average near-surface magnetism (top ~1-2 nm)



surface magnetization =

normalized XMCD = $\{I^+ - I^-\}/\{I^+ + I^-\}$

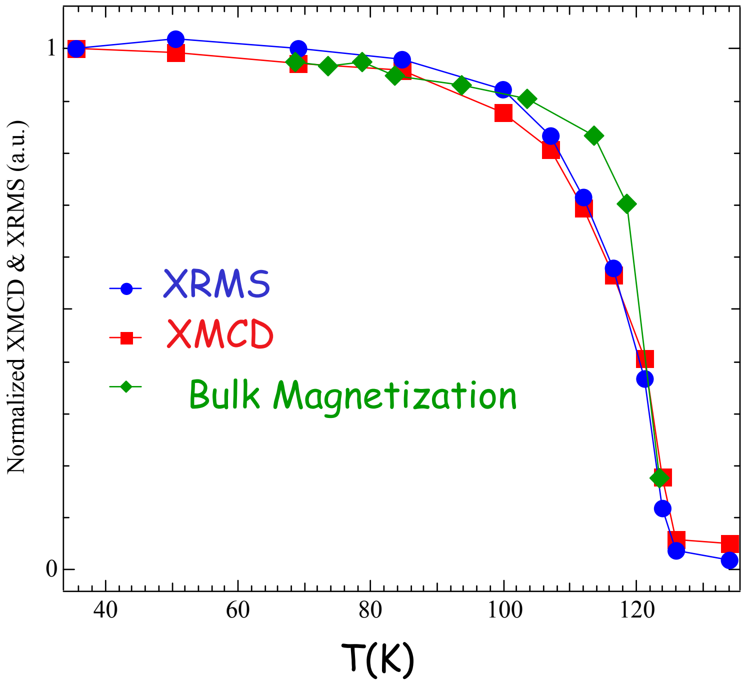
Surface magnetization XRMS, XMCD vs.temperature



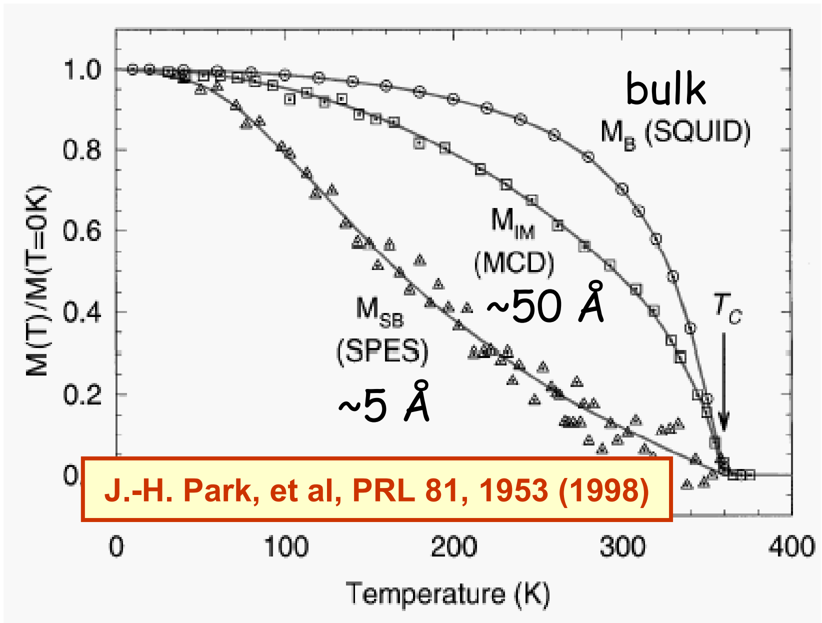
Both show bulk like M vs. T !

Magnetic Profile vs. Temperature

Bilayer Manganites



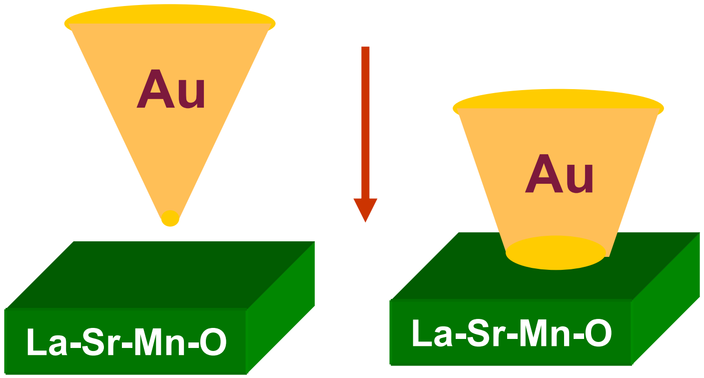
Cubic Perovskite Manganites



Bulk spin polarization in 2nd bilayer slight reduction only very close to T_C

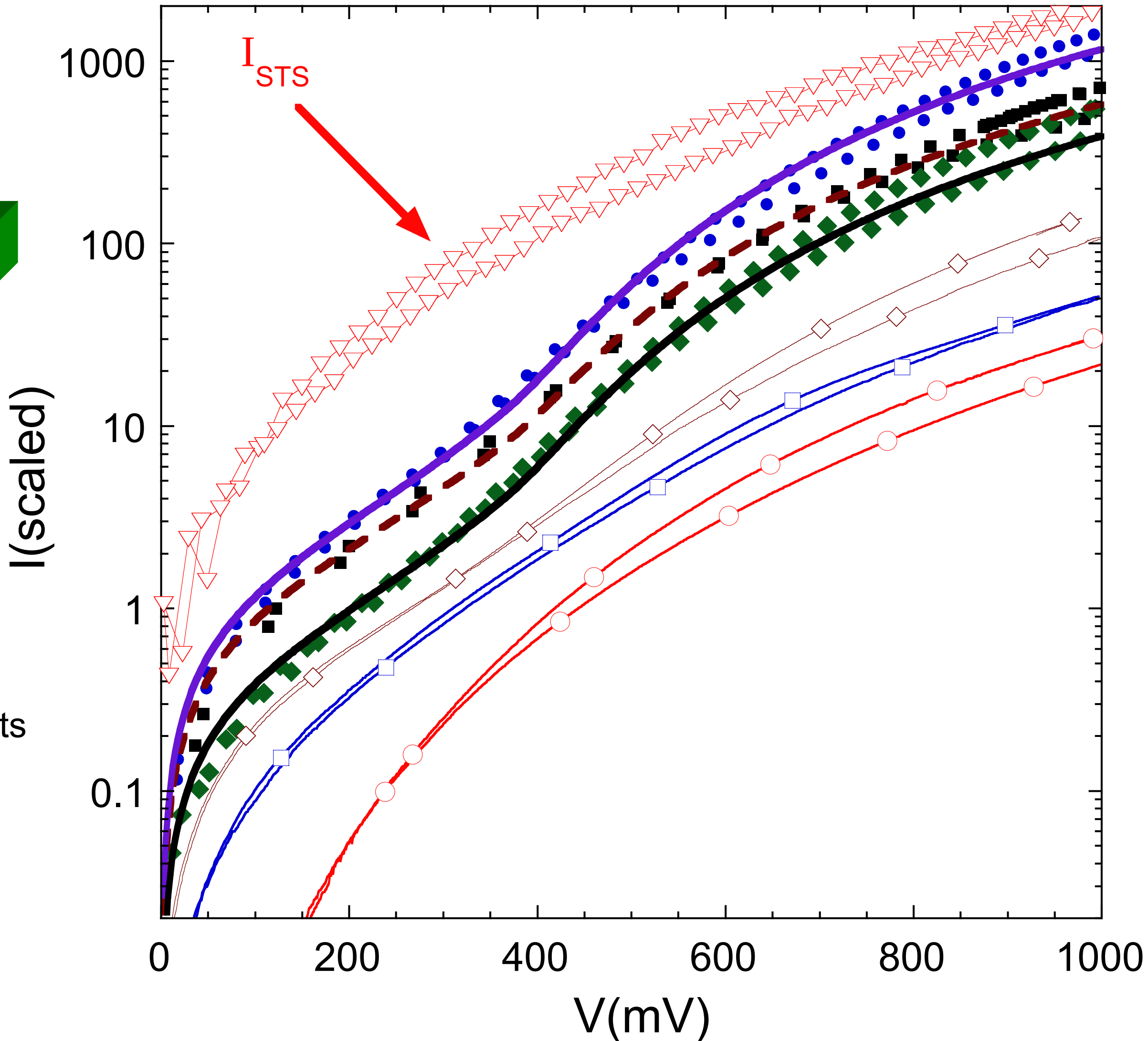
Significant reduction over most of the temperature range

Electronic Properties from Point-Contact Tunneling



Need barrier on surface of La-Sr-Mn-O crystal

Initial soft contact
Initial soft contact
After 2 hard contacts
Limiting behavior after ~10 hard contacts
Fits to tunneling calculation for ϕ=375 meV and t=1.4-1.5 nm

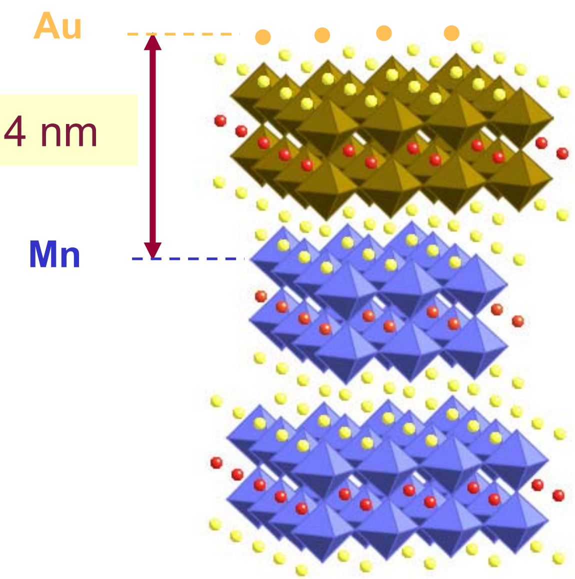


Intrinsic Insulating Surface Bilayer

Data shows the surface bilayer, *alone*, is an intrinsic insulator with a 375 meV bandgap

The 1.4-1.5 nm thick tunneling barrier is consistent with atomic dimensions

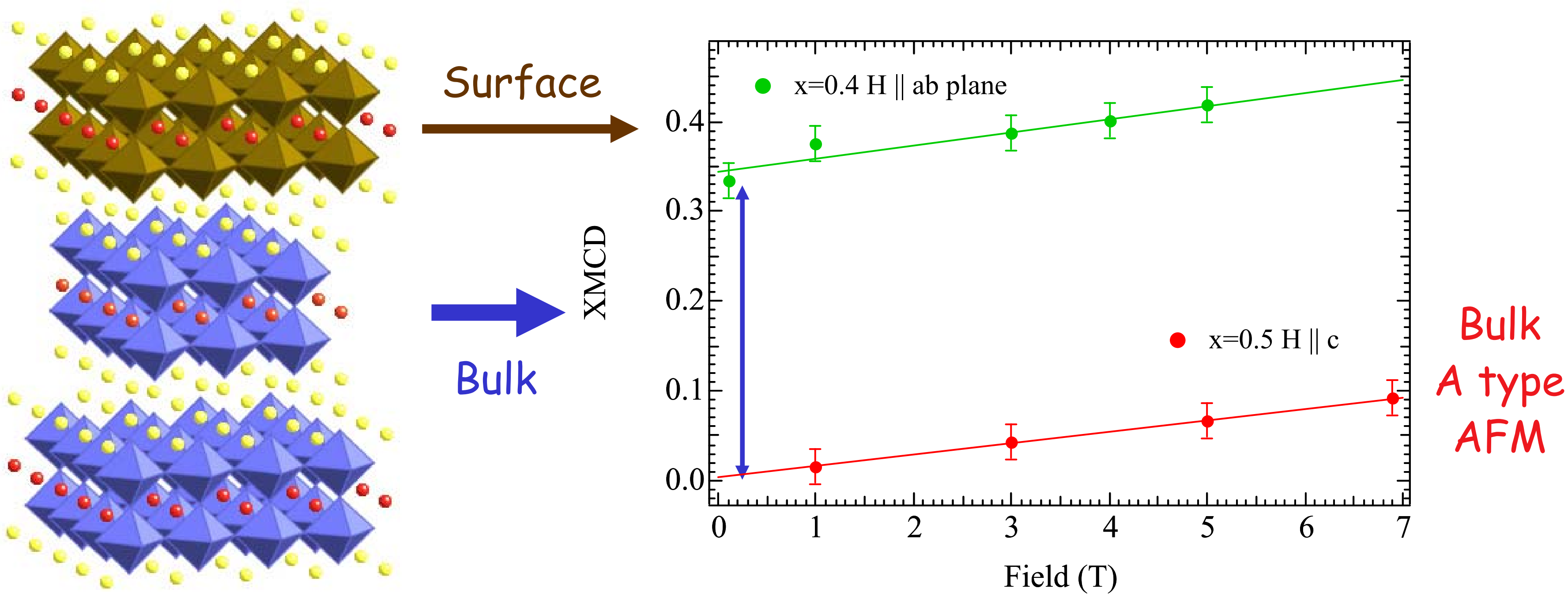
That the surface bilayer is an insulator which exhibits no long-range FM order is entirely consistent with DE



The abrupt changes in *only* the topmost bilayer is likely due to the weaker electronic and magnetic coupling between bilayers.

What is the Nature of Surface Magnetic Order?

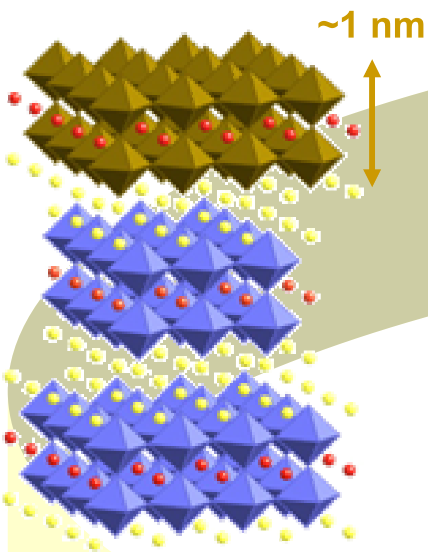
Probe surface order with near surface XMCD in high field



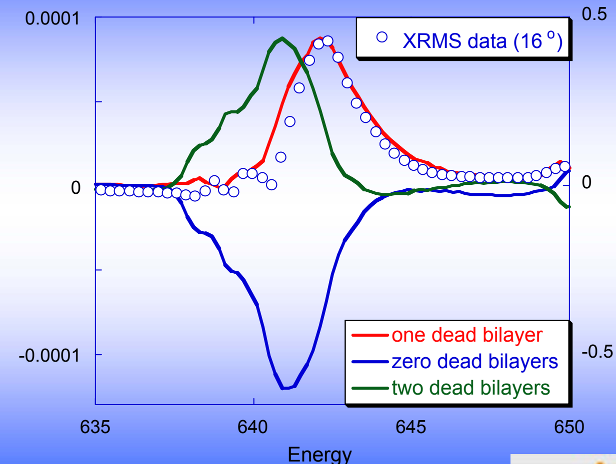
Linear slope from surface bilayer consistent with AFM superexchange

Self-Assembled Nanoskins: Pathway to Ideal Magnetic Tunnel Junctions

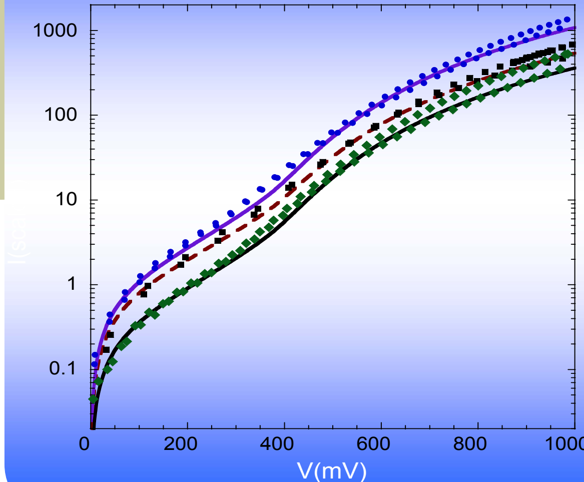
bilayer manganite



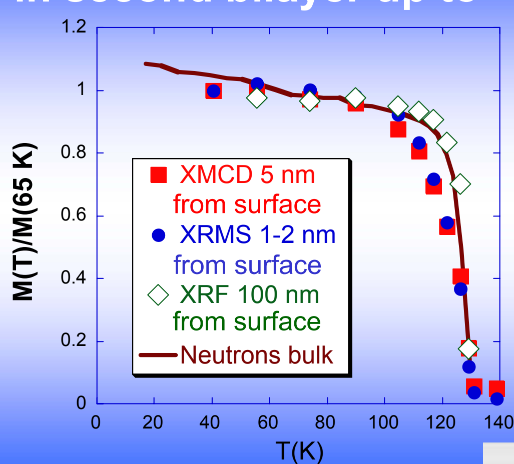
surface bilayer, alone, is non-ferromagnetic



tunnel barrier fit only by an insulating surface bilayer

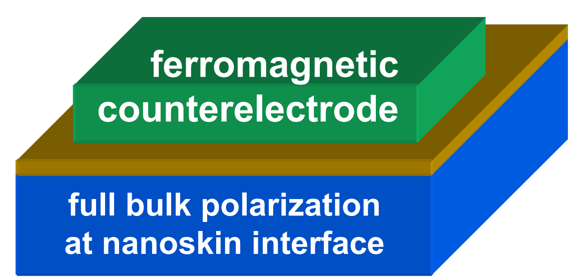


Full bulk spin polarization in second bilayer up to ~ T_C



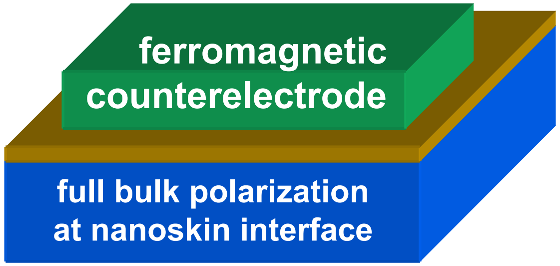
Ideal Magnetic Tunnel Junction:

intrinsic nanoskin barrier



Future Directions

- Fabricate magnetic tunnel junctions
  on layered manganites



- Seek the detailed mechanism of electron localization at surface

  study surface electronic properties using element-selective
  x-ray scattering and absorption

  explore oxygen edge as well as manganese

- Take-home message: Another example of the power of coupling synchrotron x-rays with other techniques to address complex science issues

THE END

Transport of Spin from Device to Device

One promising approach:

NATURE | VOL 401 | 7 OCTOBER 1999

Coherent transport of electron spin in a ferromagnetically contacted carbon nanotube

Kazuhito Tsukagoshi*‡, Bruce W. Alphenaar* & Hiroki Ago†