LA-UR- 09-01297

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Title: Non-uniform magnetization in LaAlO3/SrTiO3 Superlattices

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Intended for: Talk in Berlin

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Non-uniform magnetization in $LaAlO_3/SrTiO_3$ superlattices.

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Recently, Brinkman et al., [Nature **6**, 493 (2007)] reported magnetism induced at the interface between $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) inferred from transport measurements. They found the magnetization to be greatly enhanced at low temperatures (i.e., liquid He temperature) and by application of high (10+ T) fields. We report polarized neutron reflectometry measurements of the magnetization depth profile of two LAO/STO superlattices with the same number of bilayer repeats. For low temperatures and a field of 11 T, the intensities of the superlattice Bragg reflections for both samples exhibited a dependence upon neutron beam polarization. The spin dependence was much weaker at small field (and low temperature) and disappeared altogether at 11 T and 300 K. These observations demonstrate that the magnetization depth profile has the period of the LAO/STO superlattice. The neutron spin dependence was more pronounced for the sample with a thin LAO layer compared to one with a thick LAO layer, suggesting that the magnetism may be interfacial in origin.

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Non-uniform magnetization in $LaAlO_3/SrTiO_3$ Superlattices

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This work is funded by the Department of Energy

Emergent phenomena in LAO/STO

- Competition of order parameters, e.g. polarization and electric field, in $LaAlO_3$ and $SrTiO_3$ bilayers may lead to new emergent phenomena including:
  - Magnetism
  - Conductivity (2DEG)
  - Superconductivity

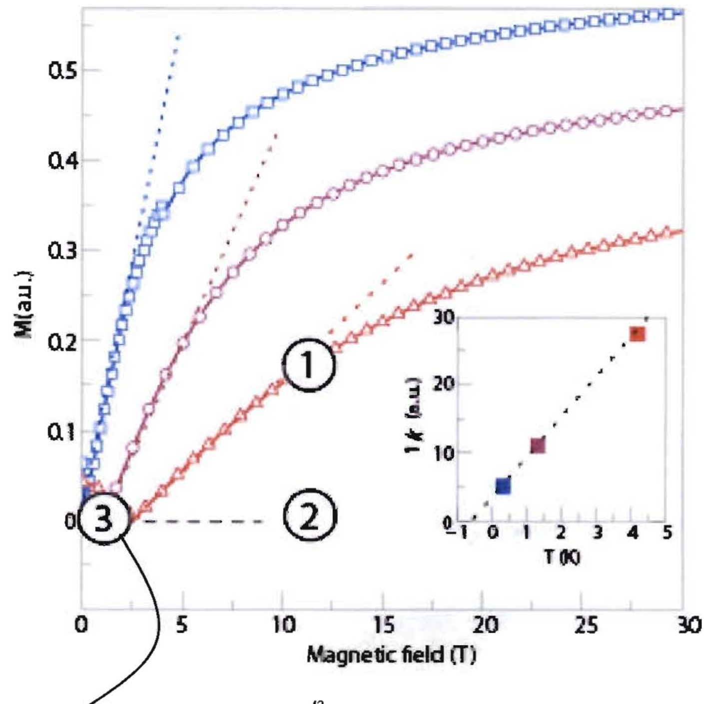
For a review see: R. Ramesh and D.G. Schlom, MRS Bulletin **33**, 1006 (2008) and articles therein.

Outline

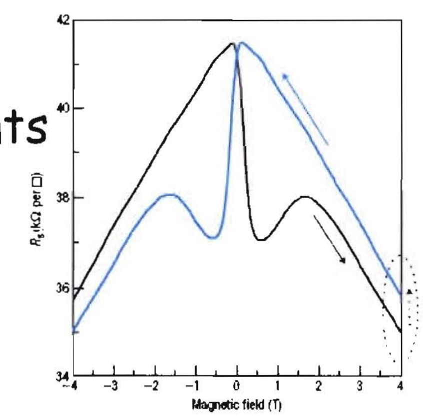
- Can the existence of magnetism in LAO/STO be confirmed?
- Why do we care? If true, we might have at hand, a mechanism for carrier induced ferromagnetism.
- How do we confirm magnetism? We look for its signature in LAO/STO superlattices with polarized neutron reflectometry.

Magnetic effects at the interface between non-magnetic oxides*

- 26 u.c. of LAO on TiO_2-terminated STO.
- Magnetization *derived* from resistance measurements.
- At 11 T and 4 K $M \sim 0.2 g\mu_B/\pi \sim 18$ emu/cm^3.
- Hysteresis is suggestive of domains.



Neutron measurements



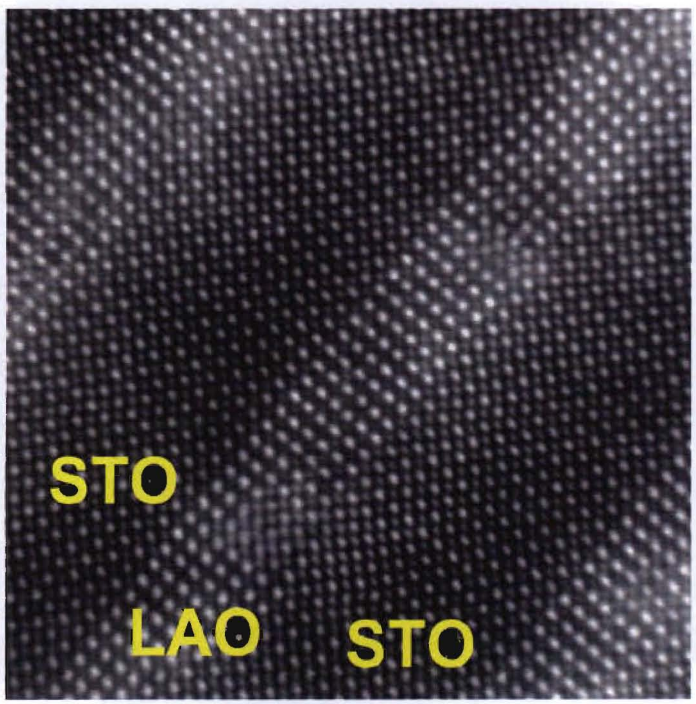
*A. Brinkman et al., Nature Materials **6**, 493 (2007).

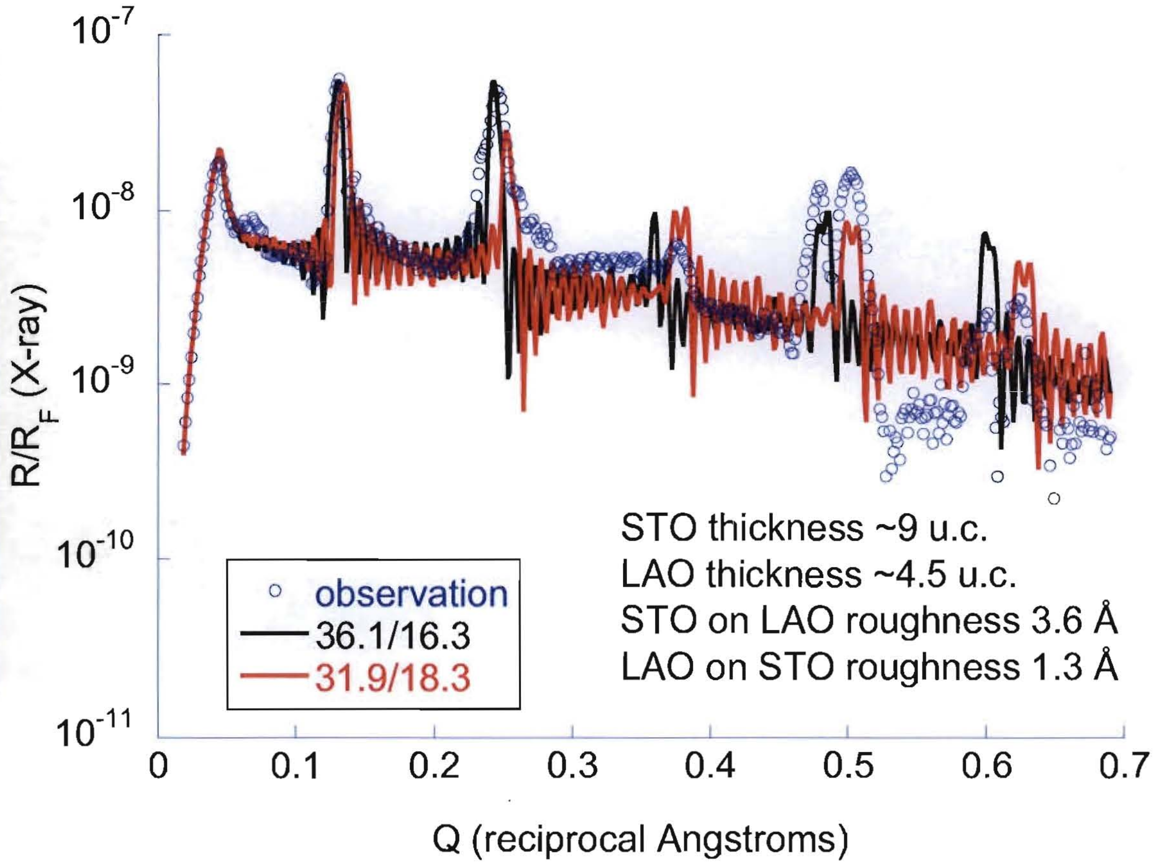
Sample fabrication

- LAO/STO superlattice grown in the group of J. Santamaria, Spain.
- Sample grown via high pressure pure oxygen sputtering technique at T = 900°C, P = 3 mbar (O_2) and 3 Å/min.
- Annealed at T = 500°C, P = 900 mbar (O_2) for 30 minutes.
- The STO substrate was TiO_2-terminated, and the STO layers *ought* to be too.
- My sample: $[LAO(4\ u.c.)/STO(12\ u.c.)]_{15}$

TEM and X-ray characterization

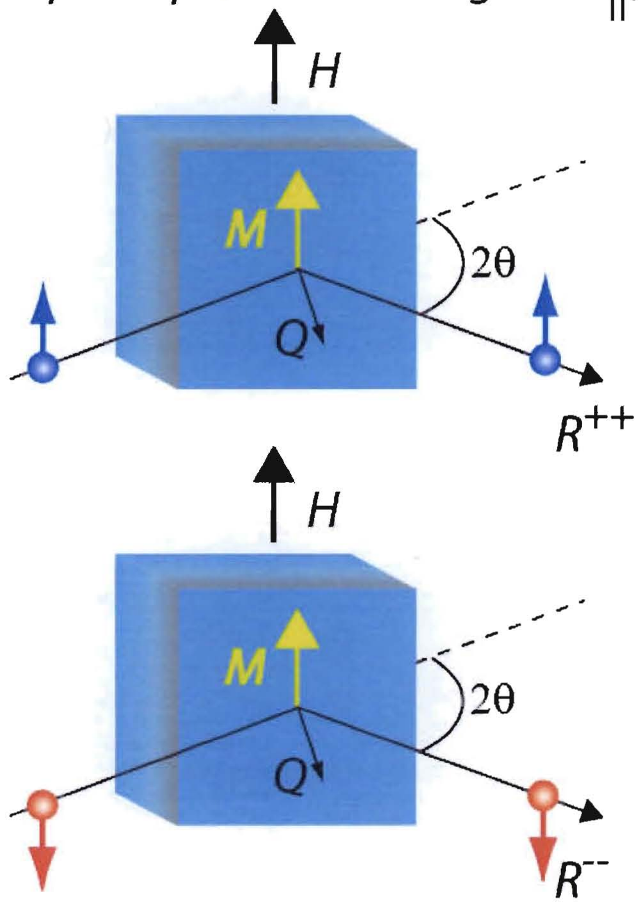
- Might the STO on LAO interface be rough because of non-integer u.c. LAO thickness?

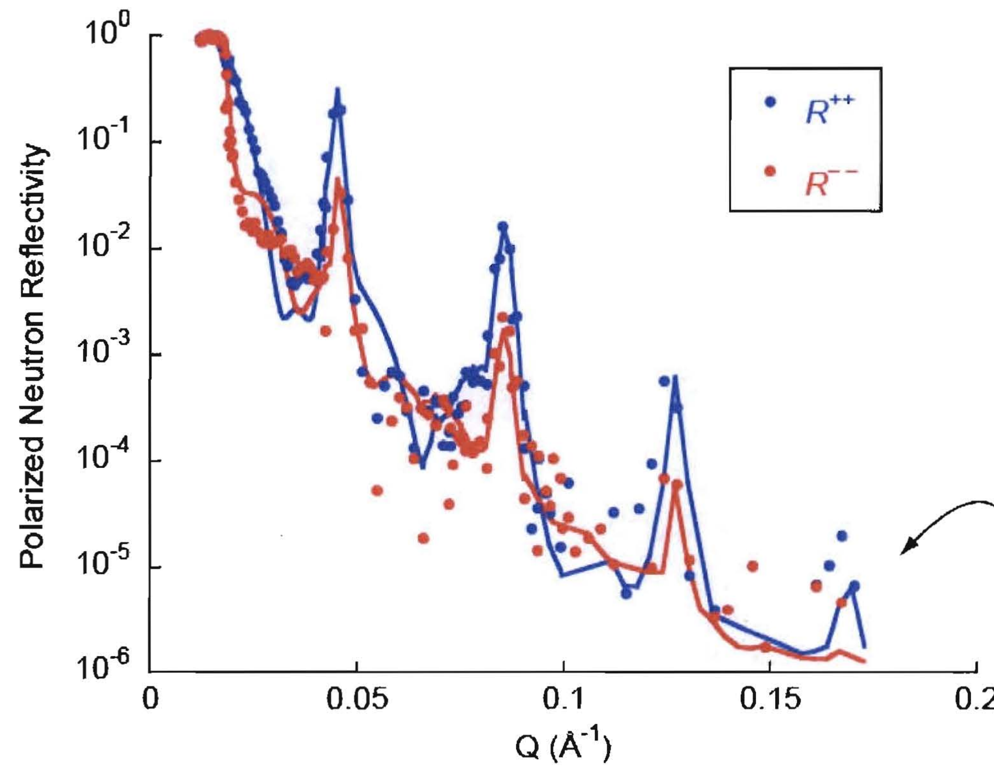




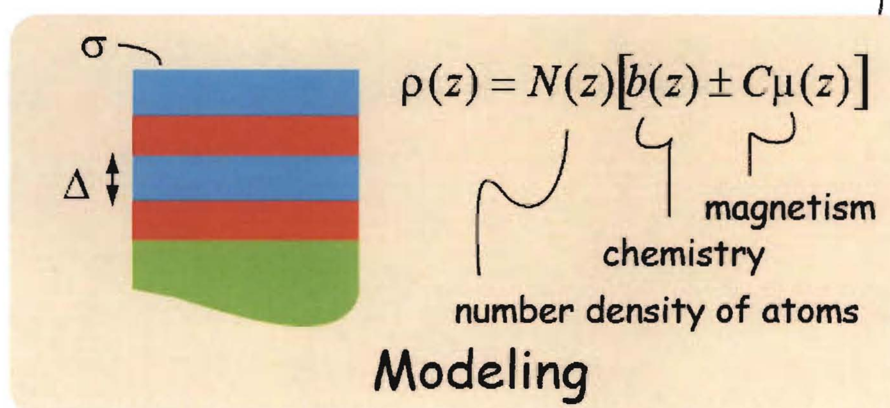
Polarized neutron reflectometry

non-spin-flip reflectivities give $\overline{M_{||}(Q)}$



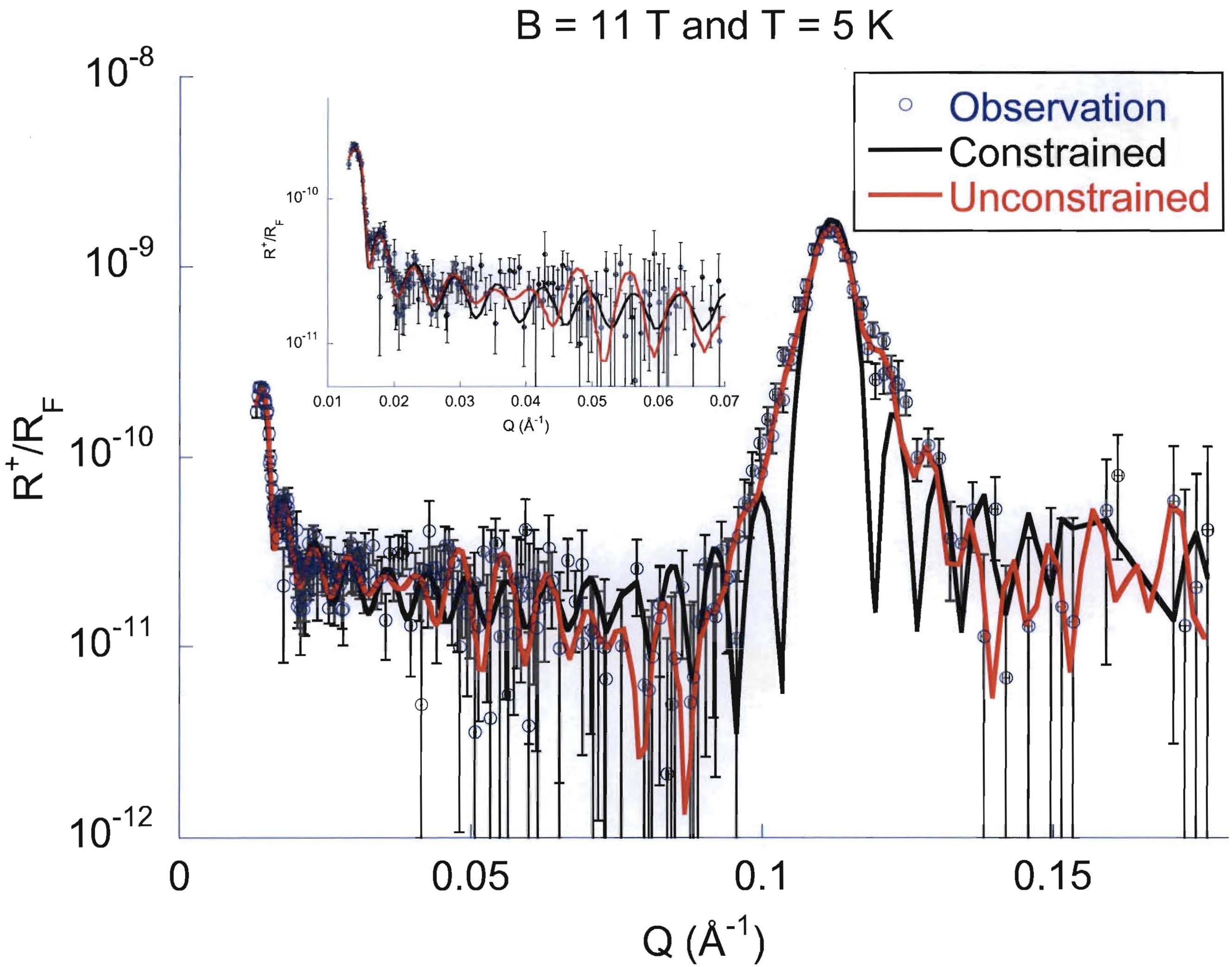


Measurement of the neutron scattering as a function of wavevector transfer (of order 0.1 A^{-1}) allows us to determine the structures and properties of materials that are non-uniform with nanometer resolution.



Why neutron scattering?

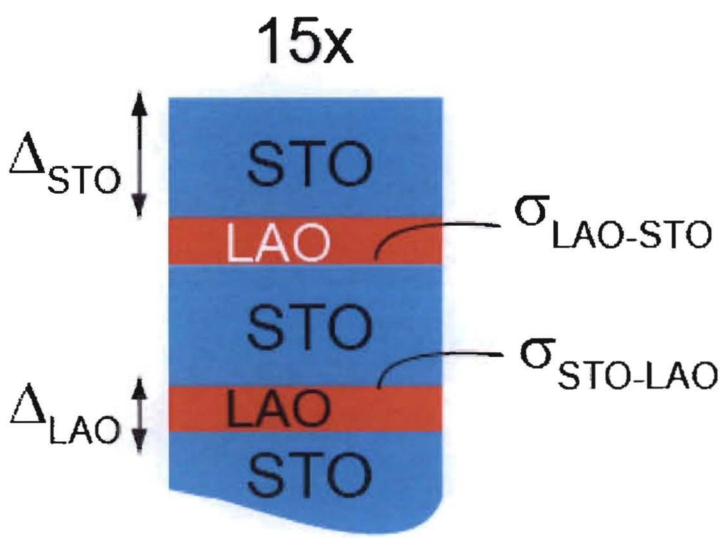
- Relatively strong, predictable, and well-understood interaction with magnetism.
- Scattering is directly related to L and S.
- Quantitative (complements MOKE, XMCD).
- Penetrating (there are probably better choices for surfaces).
- Compatible with many forms of sample environment.
- Statistical.
- Discriminating.



B = 11 T and T = 5 K
Observation
Constrained
Unconstrained
R^+/R_F
Q (Å^{-1})
10^{-8}
10^{-9}
10^{-10}
10^{-11}
10^{-12}
0
0.05
0.1
0.15
10^{-10}
10^{-11}
0.01
0.02
0.03
0.04
0.05
0.06
0.07

Modeling the nuclear structure

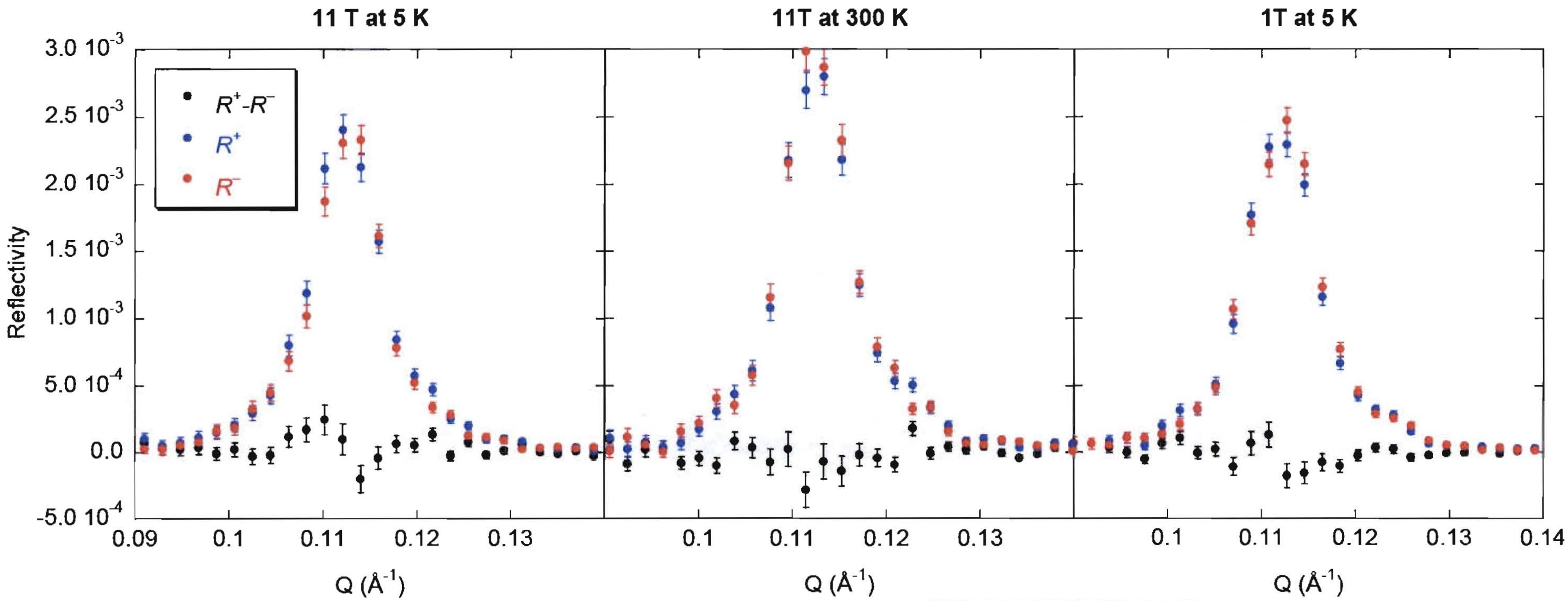
$$\rho(z) = N(z)\left[b(z) \pm C\mu(z)\right]$$



We conclude that PNR "sees" a less uniform sample than do X-rays.

- We ignore magnetism.
- We use literature Nb values for STO and LAO.
- We constrain all LAO-STO interfaces to have the same roughness, likewise for STO-LAO interfaces.
- "Thickness constrained" model: Δ_{LAO} = 19 Å and Δ_{STO} = 38 Å
- "Thickness unconstrained" model: Δ_{LAO} = 20±3 Å and Δ_{STO} = 38±3 Å, i.e., somewhat less than 1 unit cell variation.

At high B and low T, magnetization varies with the period of the superlattice.



+0.0005±0.0003

-0.0007±0.0003

-0.0004±0.0002

Measurement overcomes instrument bias and then some.

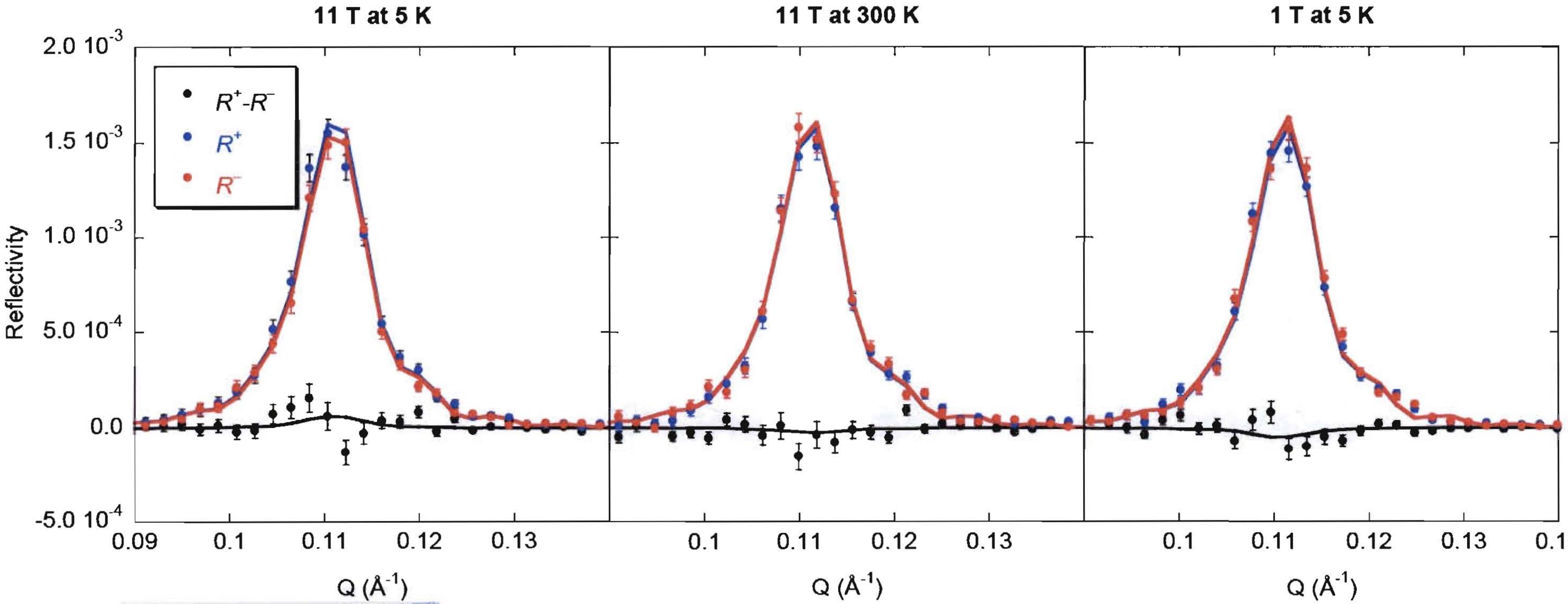
$$\int_{0.106}^{0.121} \left(R^{+} - R^{-}\right) dQ$$

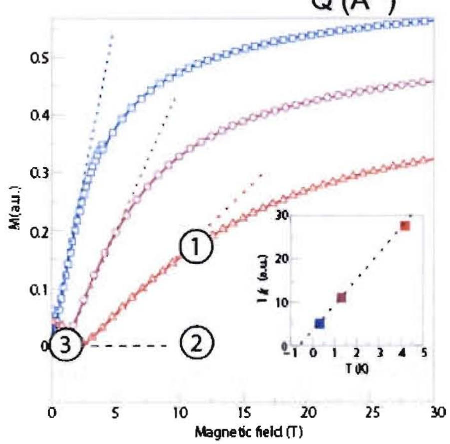
Control measurements show a negative instrument bias.

Modeling the magnetic structure

- We used the "thickness unconstrained" model for the nuclear structure of the superlattice. An adequate predictive model of the nuclear component of the Bragg reflection.
- We assumed the magnetization M within each LAO layer is uniform and the same for all LAO layers. Likewise for the STO layers (but $M_{LAO} \neq M_{STO}$).
  - To see non-uniform M *within* each layer, we would need measurements of more Bragg reflections.
- The spin dependence of the Bragg reflection tells us only the change of M, ΔM, (and its direction) across the LAO-STO interface. For example, a change from 5 to 0 emu/cm^3 or 105 to 100 emu/cm^3 affect the Bragg reflection the same way.
- The spin dependence of all the other PNR data tells us something about the magnitude of M (i.e., whether 5 or 105 emu/cm^3 was correct).

At high B and low T, LAO is magnetic, STO is not magnetic.



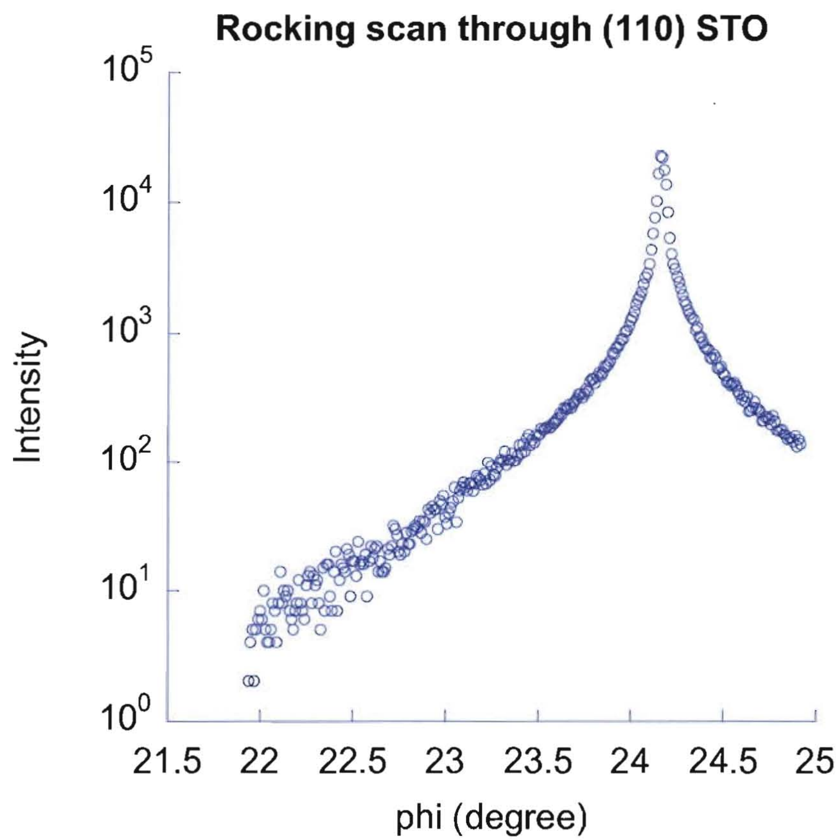


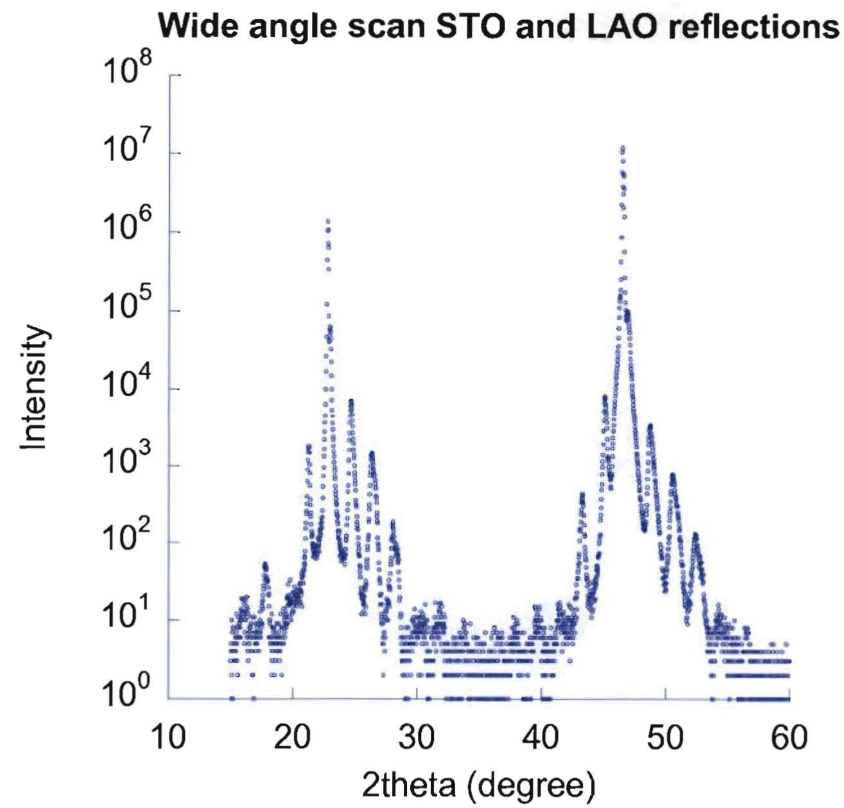
	B (T)	T (K)	M_{LAO} (emu/cm^3)
1	11	5	+5 (+7)
2	11	300	-4 (-6)
3	1	5	-5 (-2)

Conclusions

- The raw data show a positive spin dependence at the Bragg reflection that more than compensates for a negative spin dependence due to instrumental bias.
- We see a variation of magnetism in the sample that has the period of the superlattice.
- Modeling suggests the spin dependence is consistent with magnetization in the LAO layer of ~5-10 emu/cm^3, and no magnetization in the STO layer.
- We do not suggest the magnetism is necessarily ferromagnetism, nor do we suggest the magnetism is necessarily uniform within the LAO layer.
- We plan new measurements at lower temperatures with more samples, including a BTO/STO superlattice, using a new method (the Flipper égalitaire) to reduce instrument bias.

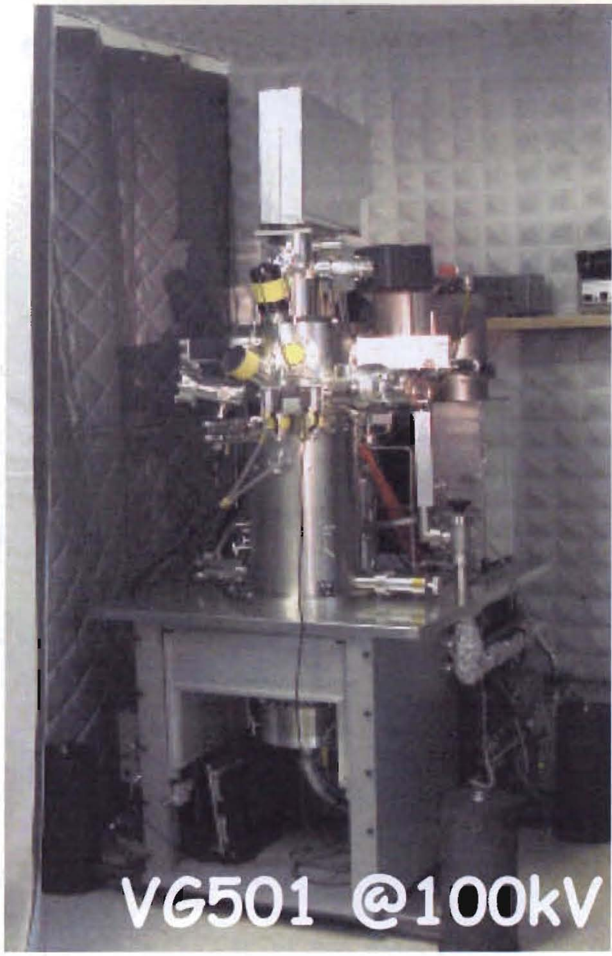
Wide angle X-ray characterization





Good crystalline quality.

What can we learn from the STEM?



Aberration corrected:
1.3 Å spatial resolution