Magnetic non-uniformity in $(La_{0.4}Pr_{0.6})_{0.67}Ca_{0.33}MnO_3$ films and measurement of the strain-magnetization coupling coefficient

S. Singh[1,2], M. R. Fitzsimmons[1], T. Lookman[1], H. Jeen[3,4], M. A. Roldan,[5] M. Varela[3], and A. Biswas[4]

[1]Los Alamos National Laboratory, Los Alamos, NM 87545, USA

[2] Solid State Physics Division, Bhabha Atomic Research Center, Mumbai 400085, India

[3]Oak Ridge National Laboratory, Oak Ridge TN 37831 USA

[4]Department of Physics, University of Florida, Gainesville, FL 32611, USA

[5] GFMC. Dpto. Fisica Aplicada III, Universidad Complutense de Madrid, 28040 Madrid, Spain

Work supported by the OBES-DOE and the NSF.

Outline

- Motivation and background
- Sample preparation and characterization
- Possible evidence for phase separation
- Magnetic depth profile $\sigma = 0$ and consequences
- Magnetic depth profile $\sigma \neq 0$ and consequences
- Conclusions

Motivation

- To explore phase separation/co-existence in LPCMO thin films.
- To understand origin of low TMR (attributed to degraded interfacial magnetization).
- To understand the *exclusive* role of strain on magnetism.

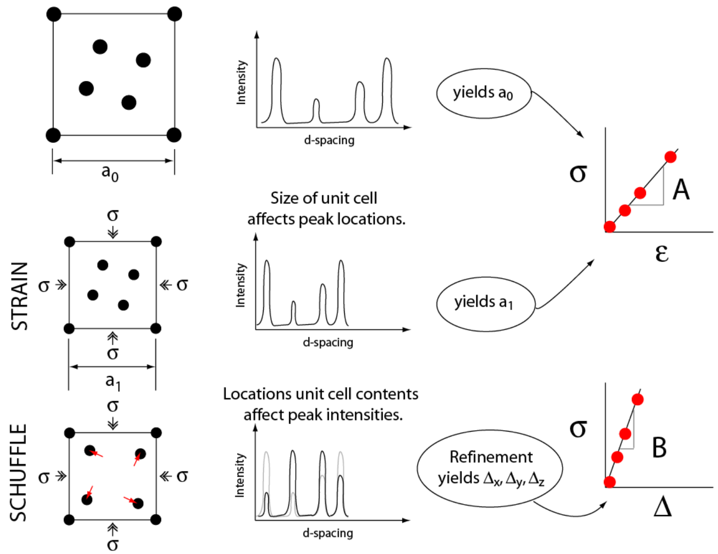
Motivation: Clarify the role of stress on ferromagnetism in manganite films, which is decidedly mixed.

Report	Compressive, strengthens FM	Tensile, weakens FM	Compressive, weakens FM	Tensile, strengthens FM
Bulk LCMO & pressure	✓			
Theory 1	✓	✓		
Theory 2			✓	✓
Thickness 1	✓	✓		
Thickness 2			✓	✓
Epi-strain	No effect	✓	No effect	
Chemical pressure	✓	✓		
Phase transformation				✓
Piezoeoelectric		✓		
Mechanical jigs	T_{MI} increases	T_{MI} decreases		

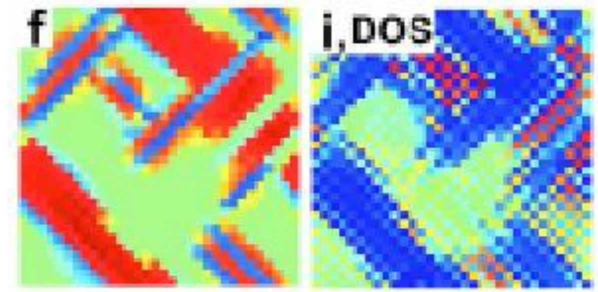
Films

Background

$$E_{structure} = E_{strain} + E_{atomic} + E_{coupled}$$
$$= A\varepsilon^2 + B\Delta^2 + C\Delta\varepsilon^2$$

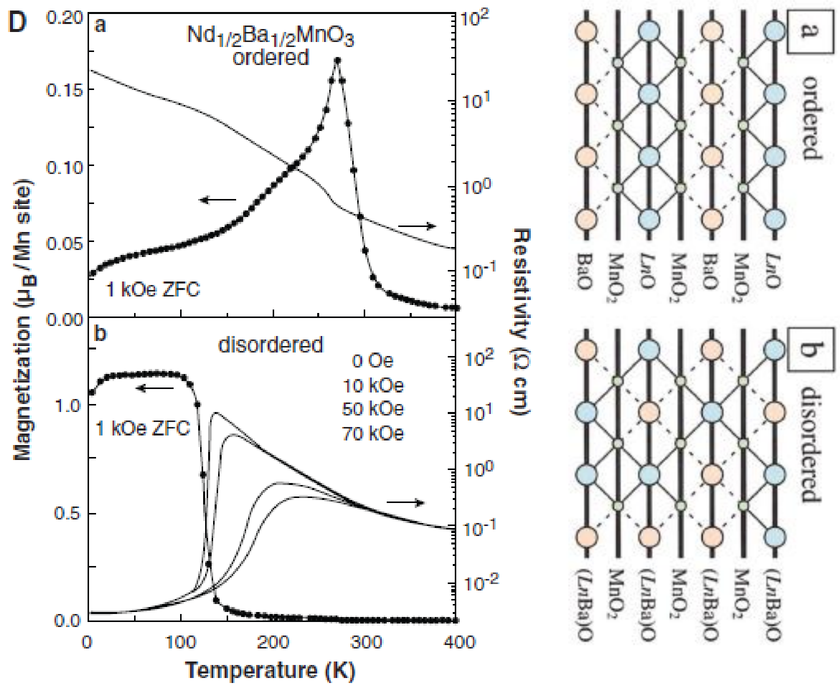


K.H. Ahn et al., Nature **428**, 401 (2004).



- Property sum and product rules are important at the nm scale. R.E. Newnham, D.P. Skinner and L.E. Cross, Mat. Res. Bull. **13**, 525 (1978).
- Also, quenched disorder [e.g., E. Dagotto, Science 309, 257 (2005)].
  - Random fluctuations of dopant density, strain fields, J-T distortions…
- Phase coexistence very sensitive to the environment.

Complexity in systems that are not “clean”.

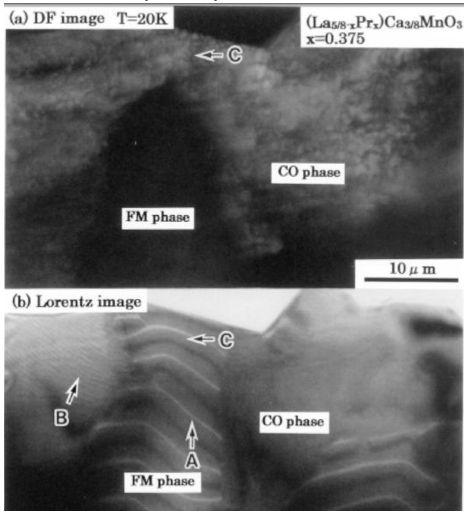


E. Dagotto, Science 309, 257 (2005).

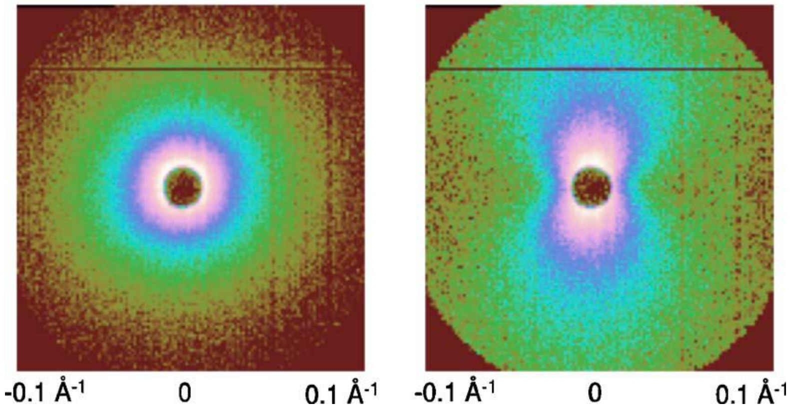
D. Akahoshi et al., PRL 90 1777203 (2003).

Experimental evidence for phase coexistence.

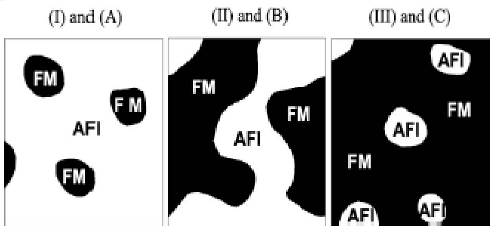
DF and Lorentz images of ***bulk*** LPCMO. (Mori)



Field dependence SANS data of ***bulk*** PCMO. (Saurel)

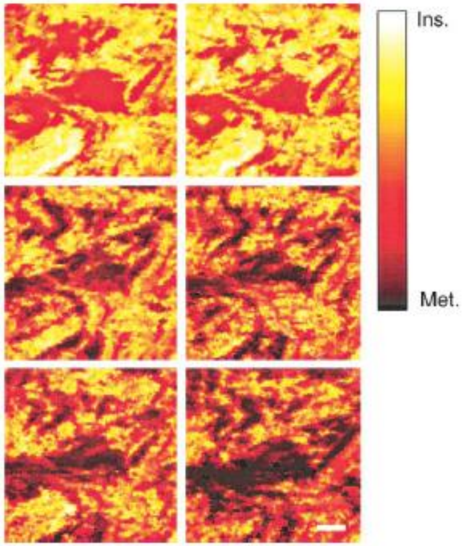


Relevant length scales vary from 100's nm to 10's of microns.

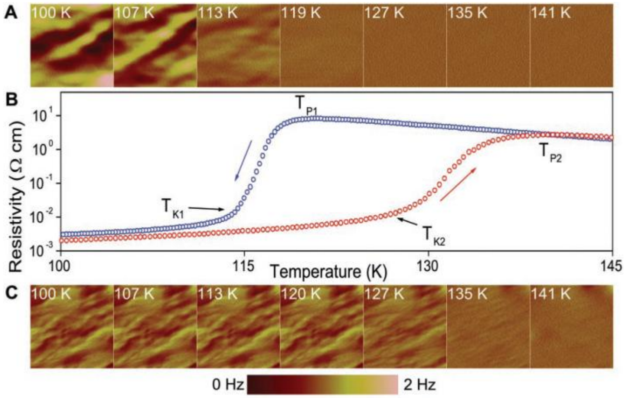


H increases ⟶

STM images of LCMO ***thin film*** from 0 to 9T. (Fäth)



MFM of LPCMO ***thin film***
Zhang et al., Science 298 805 (2002).



Compelling evidence

	Electronic phase separation?	Magnetic phase separation?
Bulk	Yes	Yes
Film	Yes	?

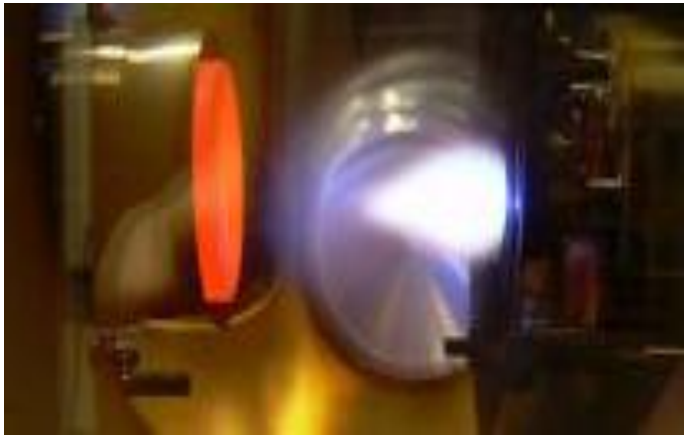
Sample preparation

- Samples grown by PLD (A. Biswas, UFL).
- Target composition: $(La_{0.4}Pr_{0.6})_{0.67}Ca_{0.33}MnO_3$
- (110) $NdGaO_3$ (NGO) substrates are 1cm by 1cm by 250μm.
- 30 nm thick (101) LPCMO single crystal films.
- Small epi-strain:

+0.4% || [001] NGO

+0.2% || [-110] NGO

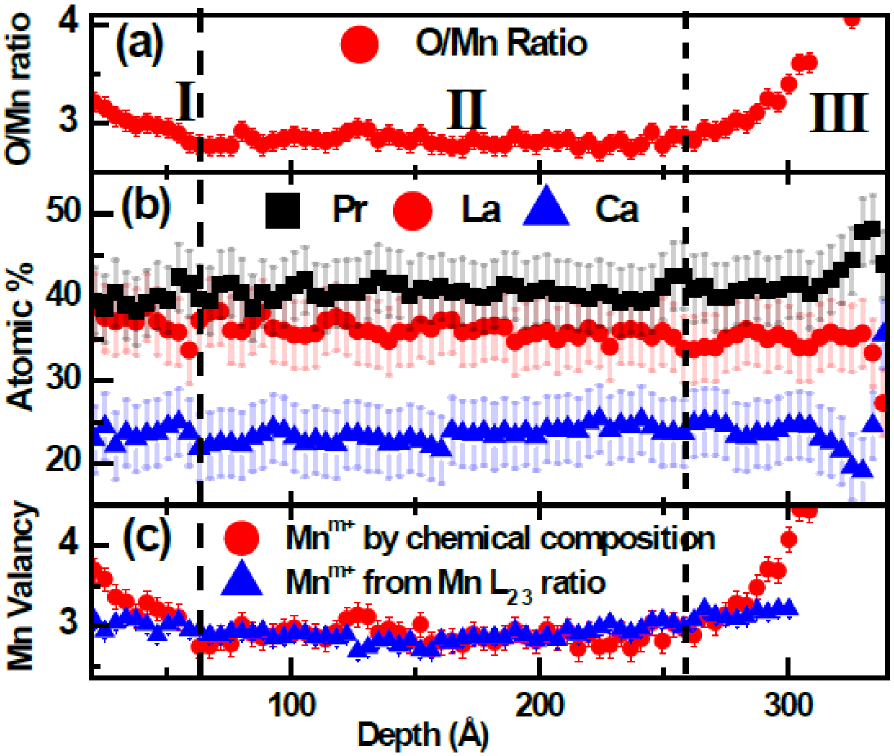
relative to bulk LPCMO.



Electron energy loss spectroscopy

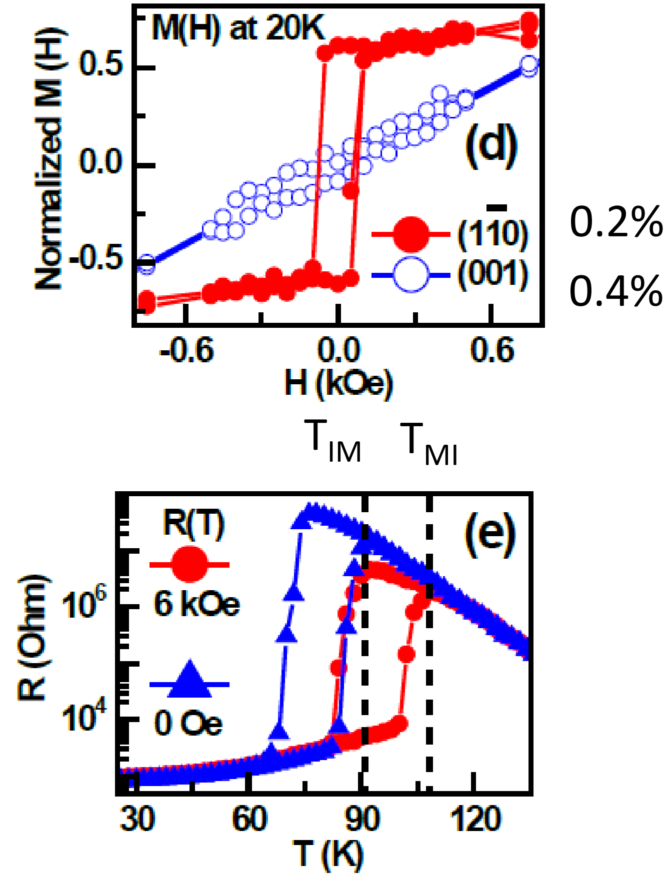
- Chemically nonuniform.
- Excess Mn^{4+} at surface and buried interface.
- Excess Mn^{4+} due to excess O, not Ca deficiency.

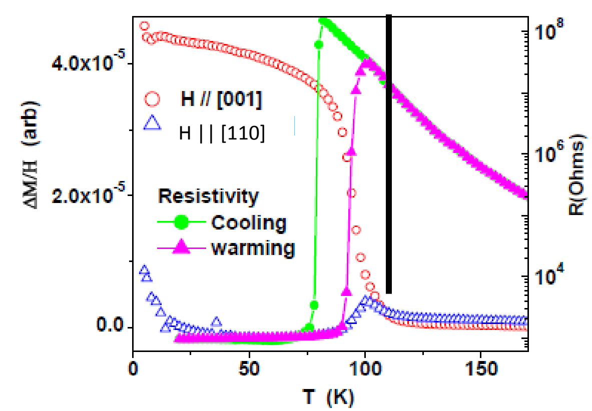
$(La_{0.5}Pr_{0.5})_{0.77}Ca_{0.23}MnO_3$



Magnetometry

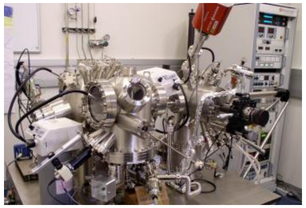
- Strong in-plane anisotropy.
- Field favors metallic phase.
- Metal-insulator transitions are not the same as the Curie temperature.

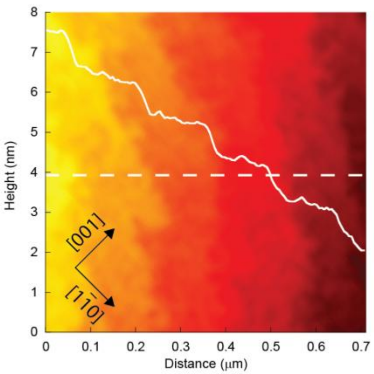


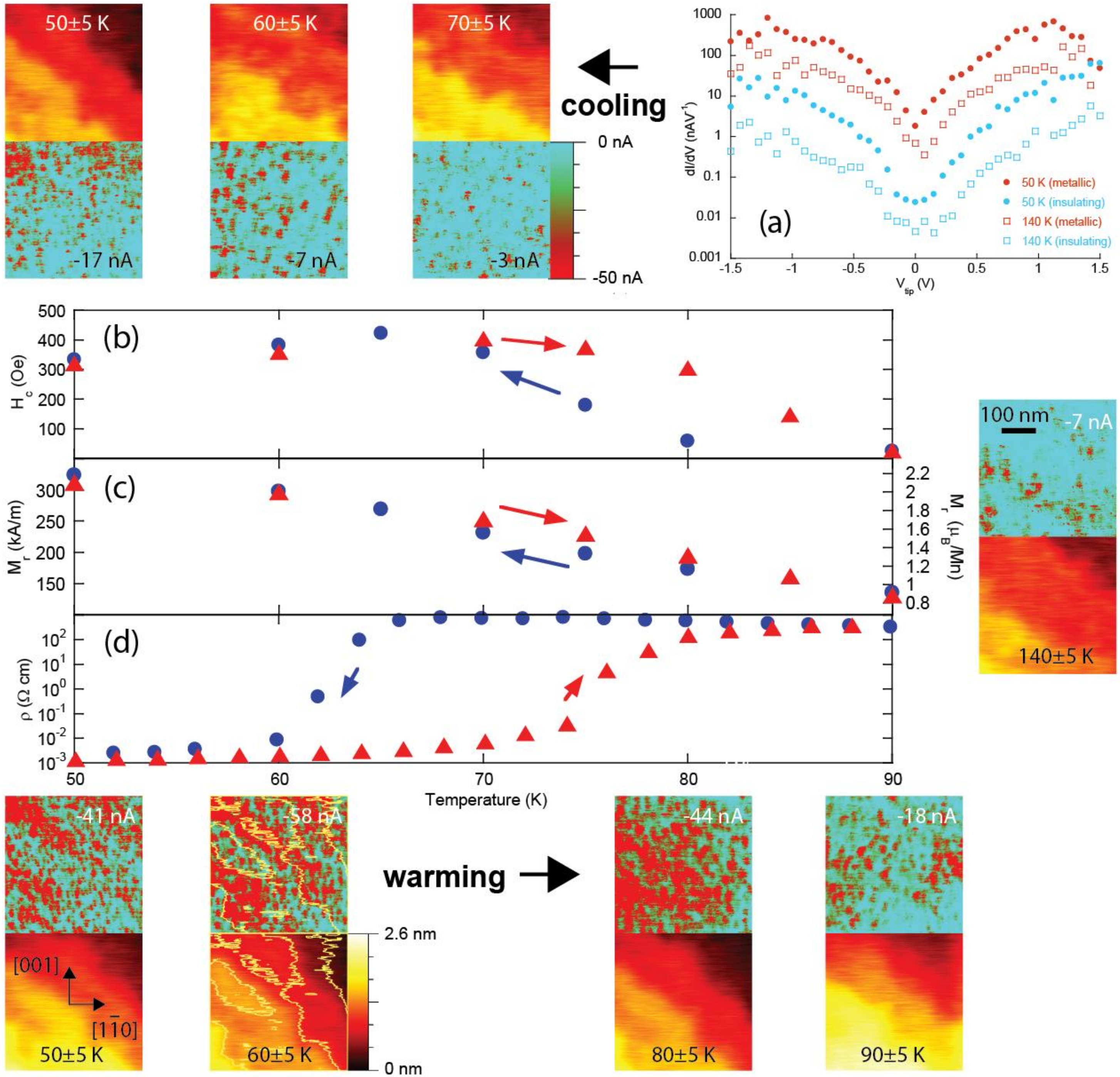


Performed at the ANL CINT facility (w/ J. Guest).

Temperature changed at 0.4K/min.

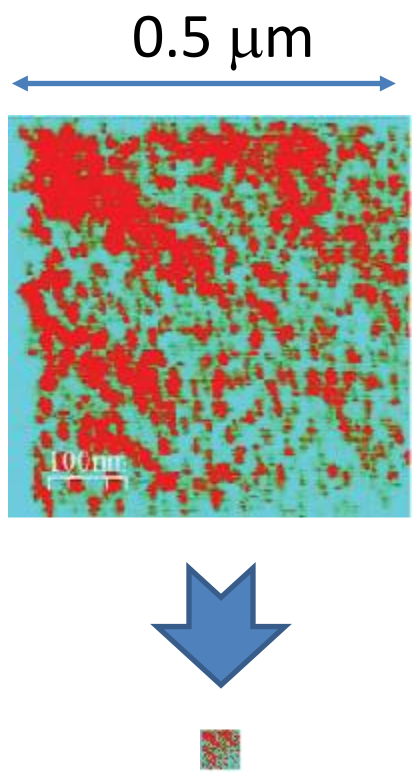




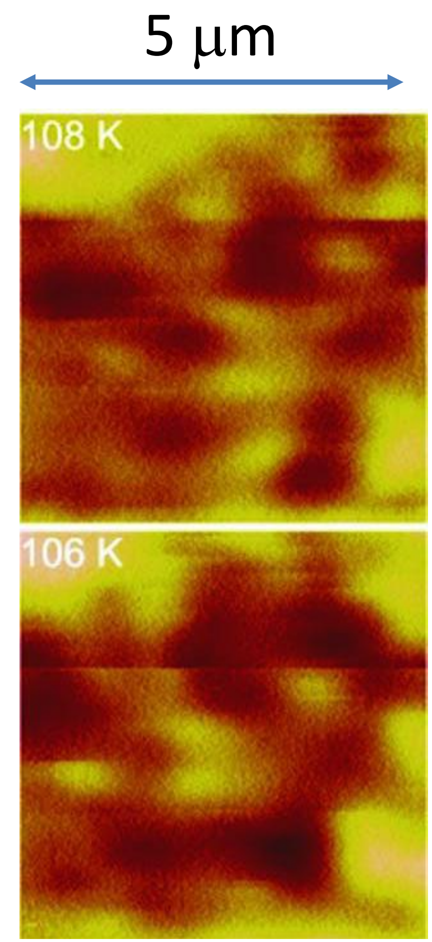


The length scales for electronic and magnetic texture do not match for films of the same nominal composition.

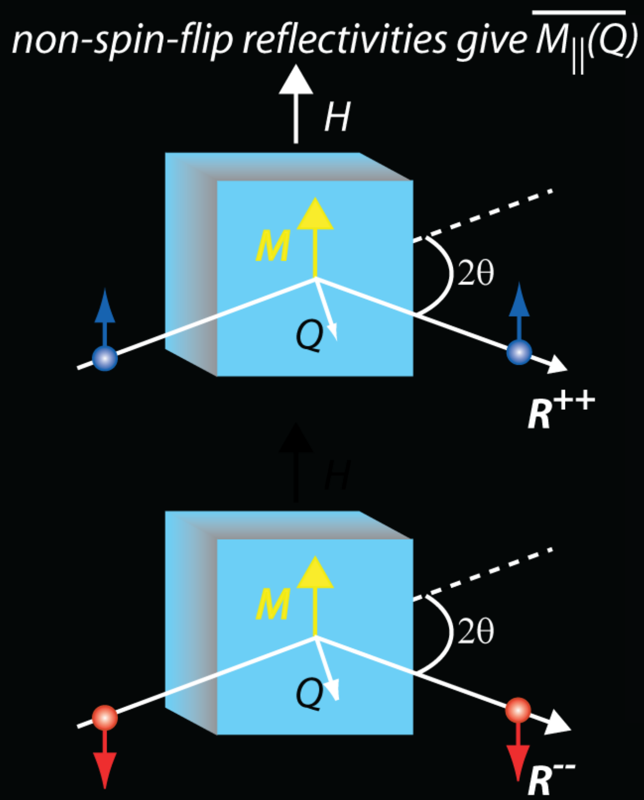
cAFM (this work)

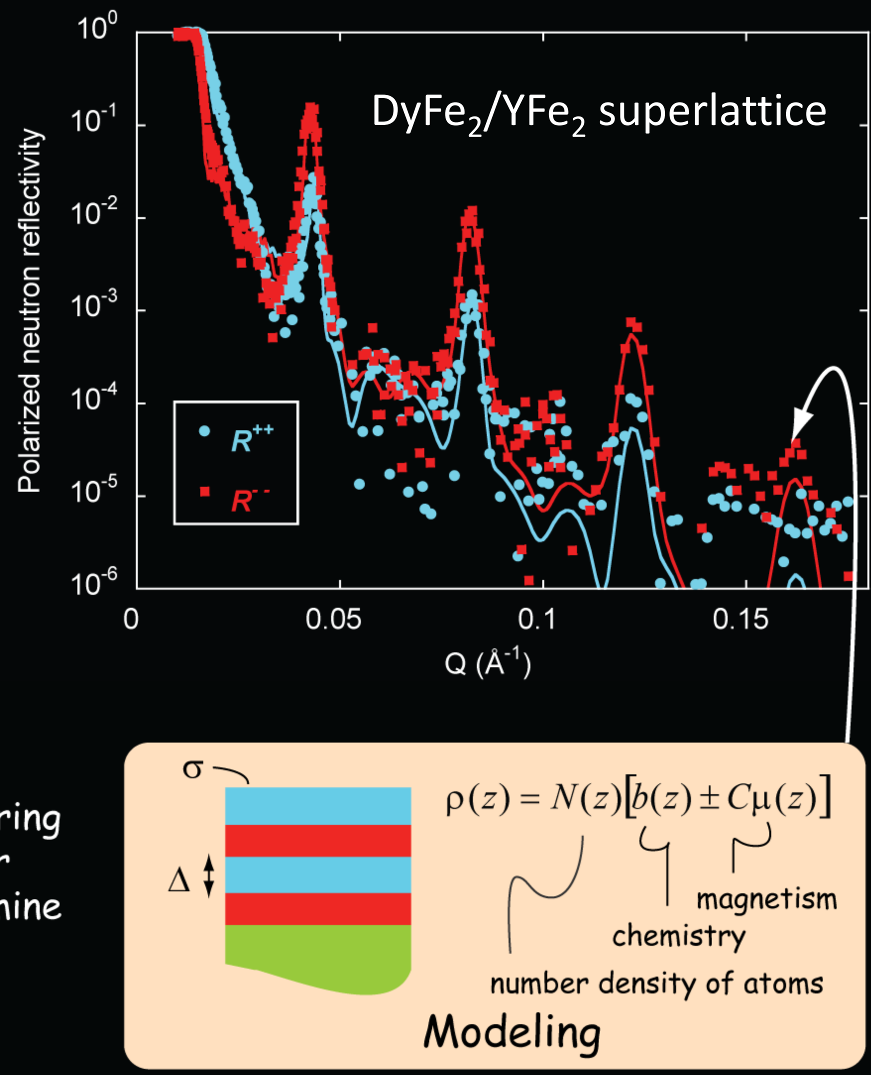


MFM (Lozanne, Phys. Today 1/3)



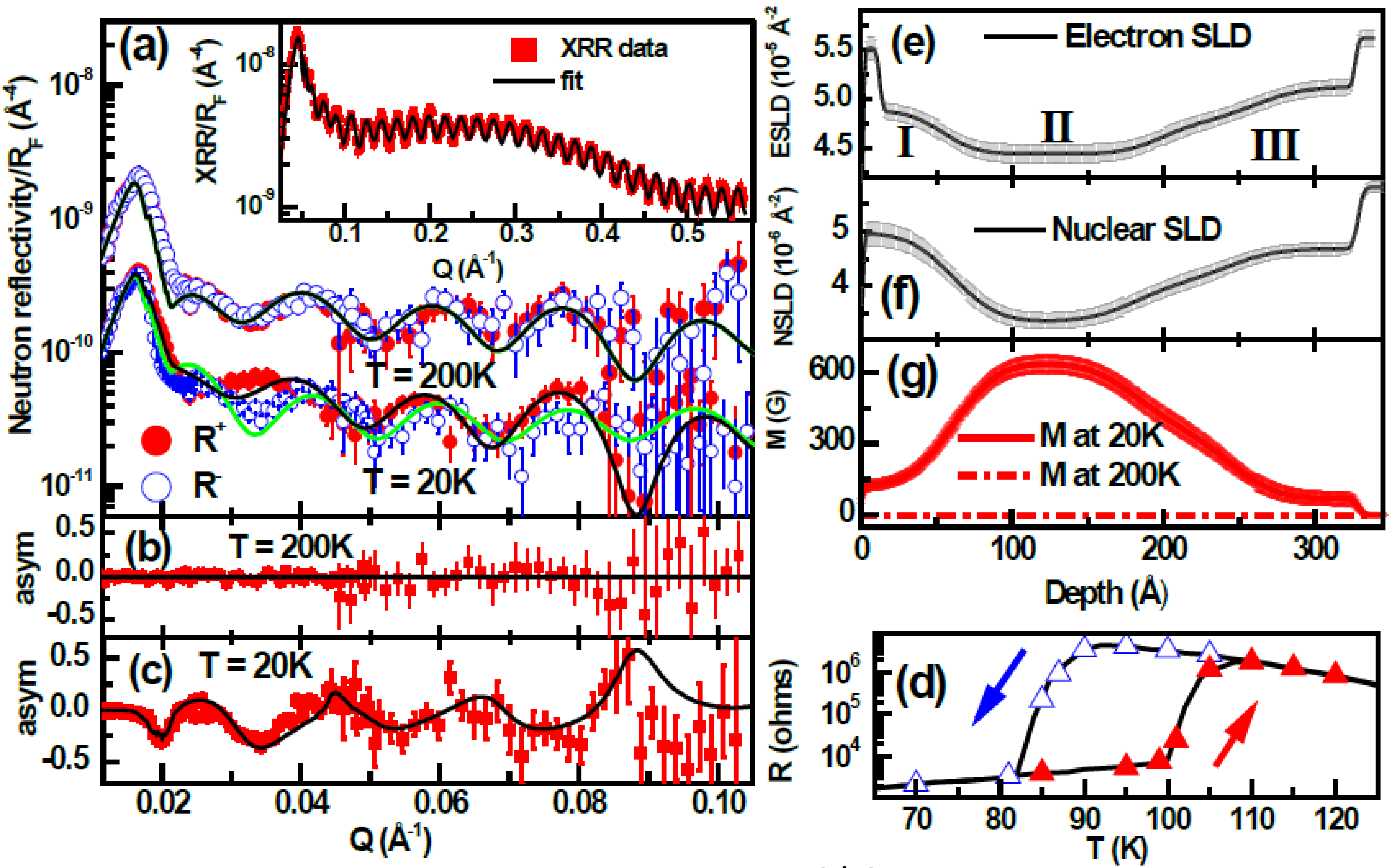
Polarized Neutron Reflectometry





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.

Chemical and magnetic depth profiles are non-uniform (σ = 0)



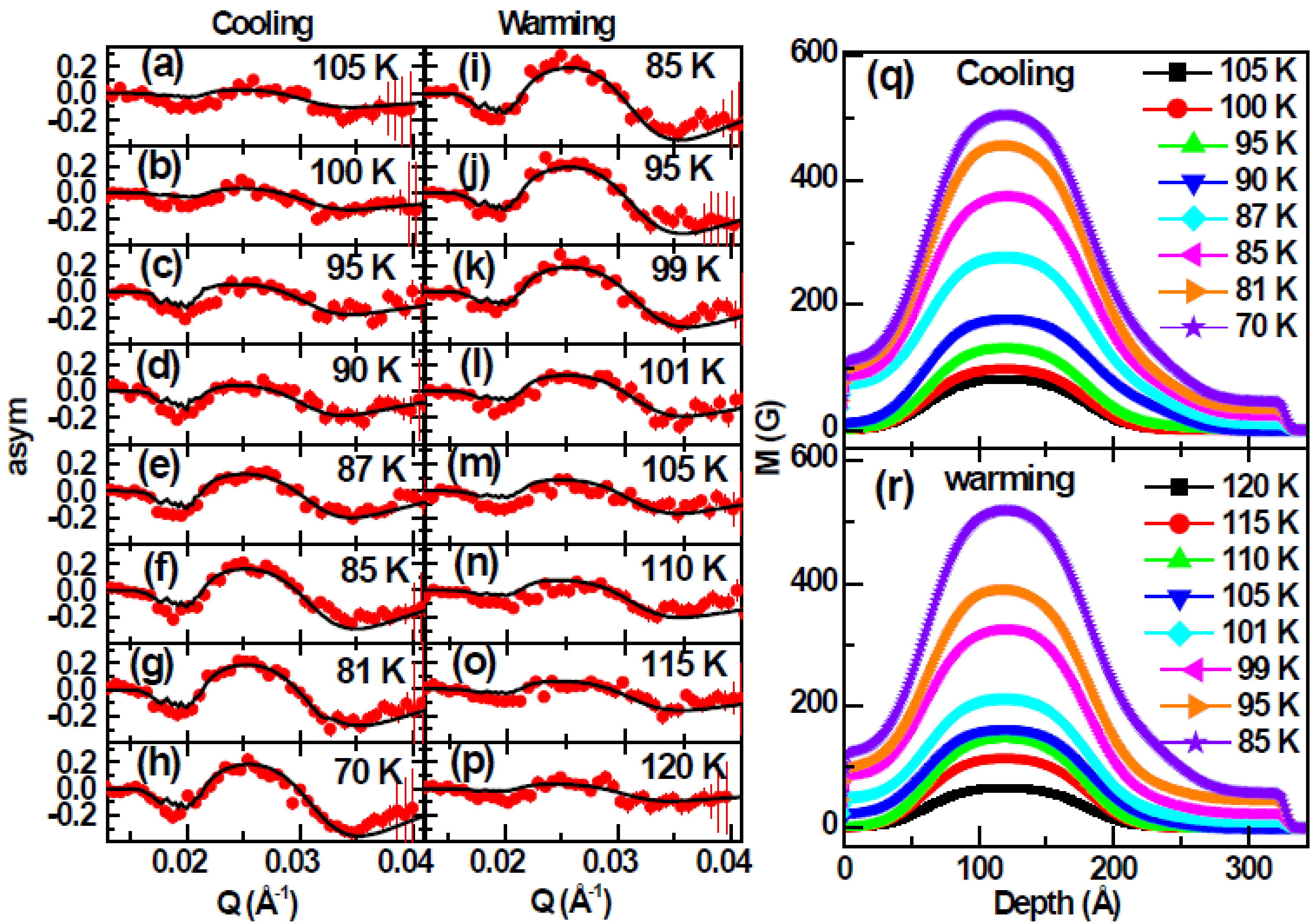
H = 6 kOe

S. Singh et al., PRL **108**, 077207 (2012)

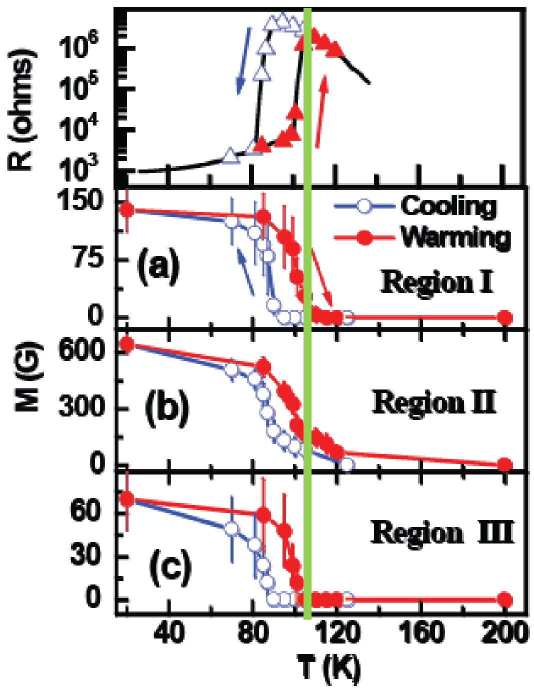
Temperature dependence of the saturation magnetization

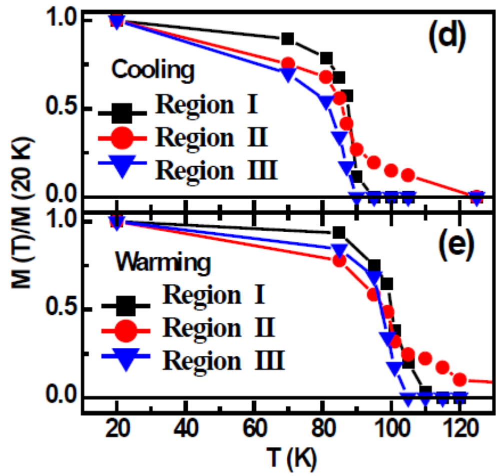
Spin asymmetry of the neutron reflectivity

Magnetization depth profiles



Saturation magnetization is less in Mn^{4+} rich regions.





H = 6 kOe

(1) M shows hysteresis similar to R.

(2) T_c of region II exceeds T_{MI}

(3) M_s suppressed in Mn^{4+} rich regions (more AF interactions?).

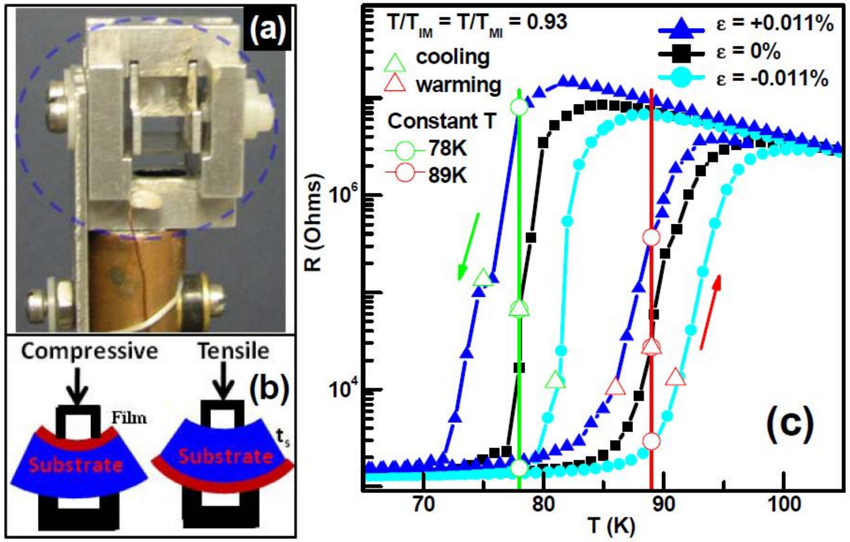
S. Singh et al., PRL **108**, 077207 (2012)

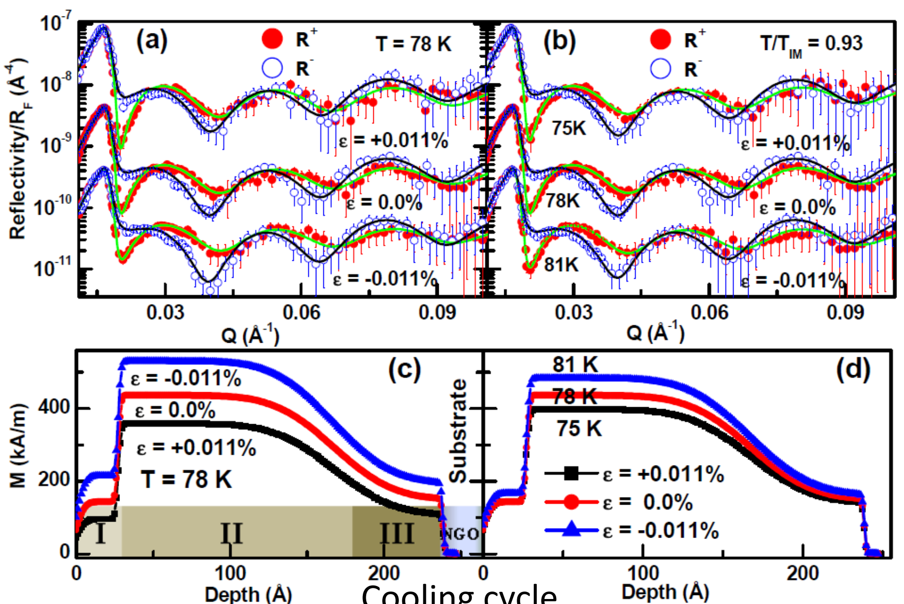
Compressive stress stabilizes T_{IM} and T_{MI} to higher T and increases M_s.

- 4 point mechanical jig produces $\varepsilon = \pm 0.011\%$.
- Neutron scattering and transport measured vs. σ, H and T.
- Collected data for constant T and constant $T/T_{IM,MI}$.
- Compressive stress ($-\varepsilon$) increases M_s (T_{MI} & T_{IM}).

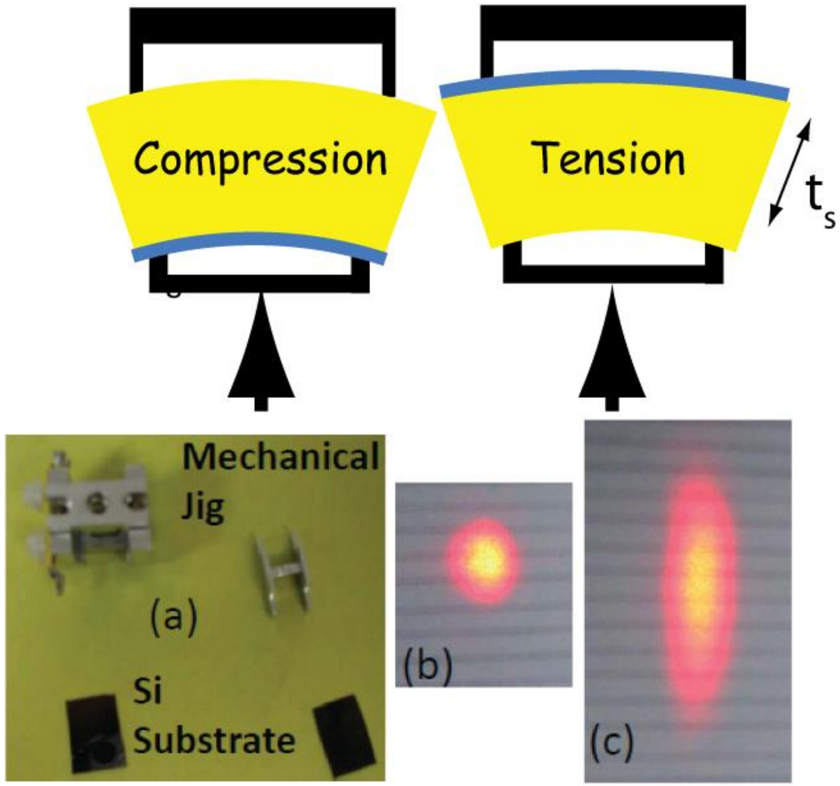
H = 6 kOe

http://arxiv.org/abs/1201.4001

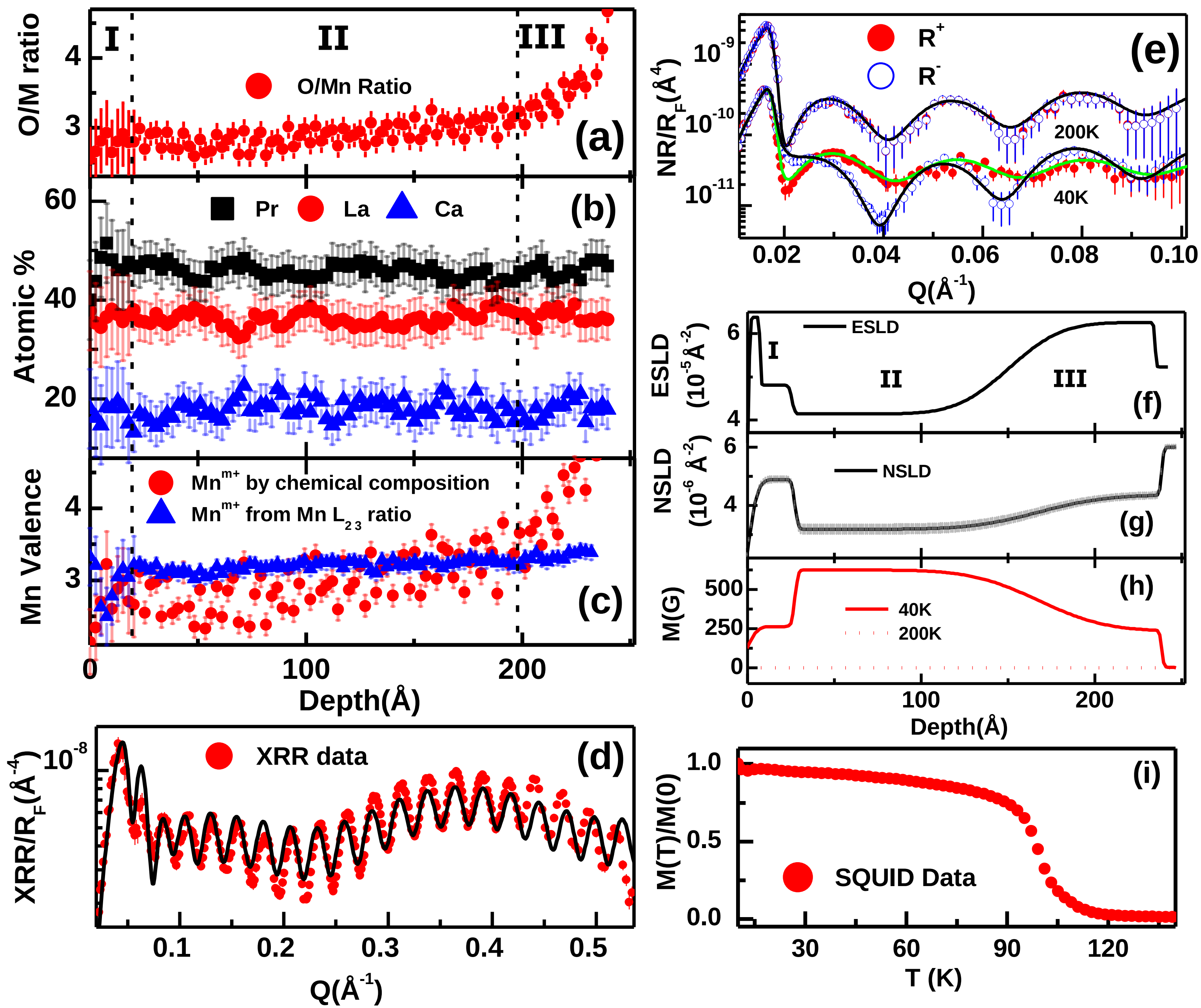




Cooling cycle

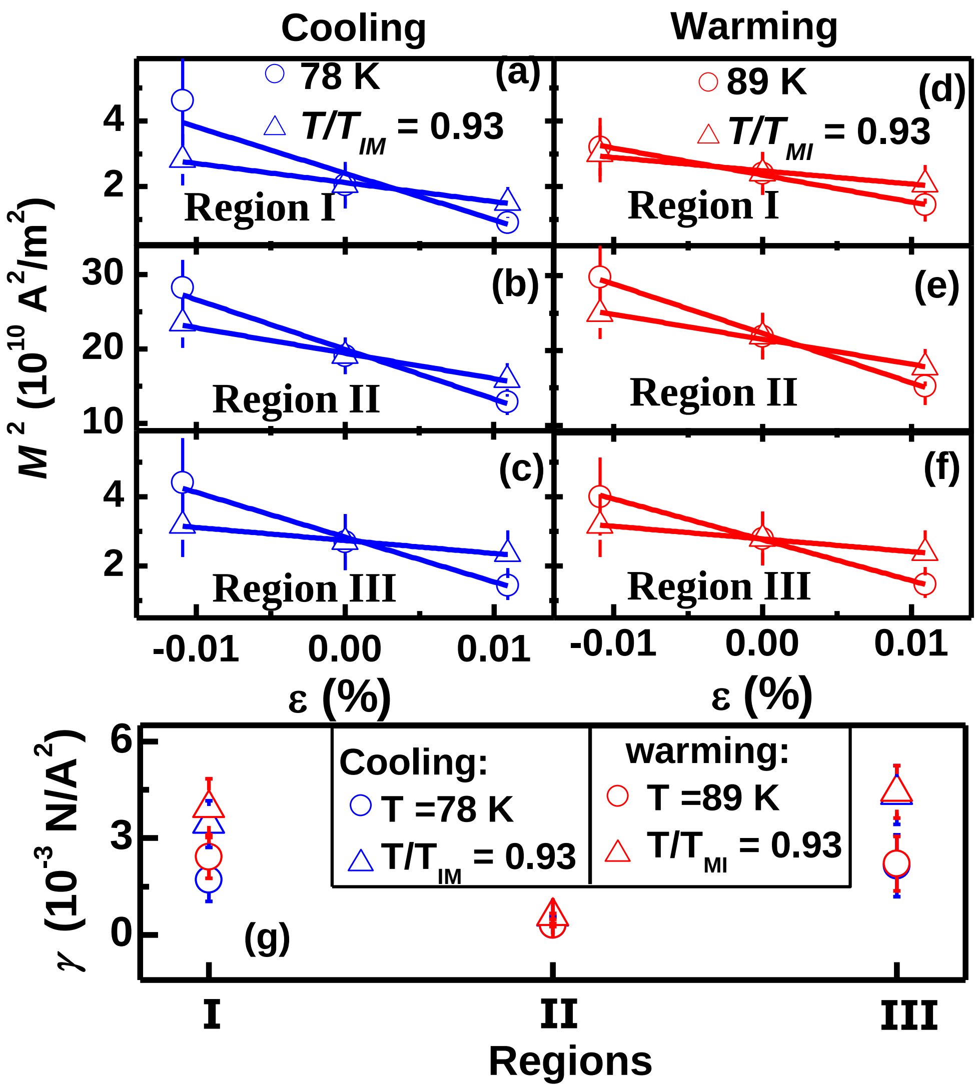

Compression
Tension
t_s
Mechanical
Jig
(a)
Si
Substrate
(b)
(c)

Characterization of the film: EELS, XRR, PNR



Magnetic depth profile & σ ≠ 0

- $F_C = \gamma \epsilon M_\varepsilon^2 + \frac{A}{2}\epsilon^2$
- $M^2 = M_0^2 - \frac{A}{\gamma}\epsilon$
- A = 200 GPa
- γ does not depend upon cooling or warming cycles.
- γ smallest for film bulk (least Mn^{4+}) implies strongest coupling.
- γ ~ 0.0003-0.0006 N/A^2



http://arxiv.org/abs/1201.4001

Conclusions

- Length scales of electronic and magnetic texture may differ.
- Length scale of electronic texture confined by terrace steps.
- The LPCMO films are neither chemically nor magnetically uniform with depth.
- Compressive *elastic* strain (-'ve ε)
  - Increases M_s.
  - Favors the ferromagnetic phase.
  - Increases the metal-insulator transition temperatures.
- Coupling between strain and ferromagnetism is strongest for the bulk film composition (i.e., not Mn^{4+} rich).
- Demonstrated several technical innovations that can be broadly applied to other systems, especially multiferroic, and piezomagnetic films.