

# **Ferromagnetism and “Polaron” Percolation in Novel Magnetic Materials**

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The University of Tennessee**

# Outline

- **Electronic Phase Separation in Complex Oxides**

*PRL (in press)*

- **Formation and Percolation of Polarons in Dilute Doped Magnetic Semiconductors**

*APL 86, 152507 (05); PRB (in press)*

- **Surface States Mediated Ferromagnetic Coupling in Magnetic Nanodot assembly**

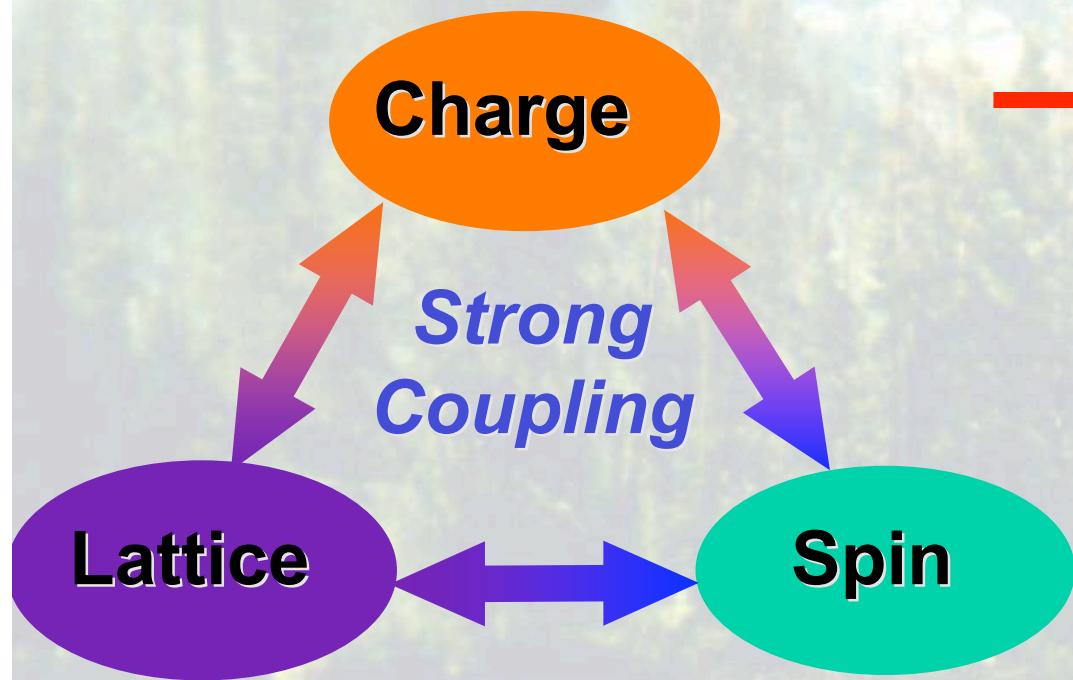
*PRL 92, 237201 (04); PRL 95, 27201 (05); PRL (in press)*

# I. Electronic Phase Separation in Complex Oxides

Visualization of Doped Holes (Small  
Polarons) with Atomic Resolution in  
 $(La_{5/8-0.3}Pr_{0.3})Ca_{3/8}MnO_3$

# Electronic Phase Separation and Inhomogeneity

## Strongly Correlated TMO



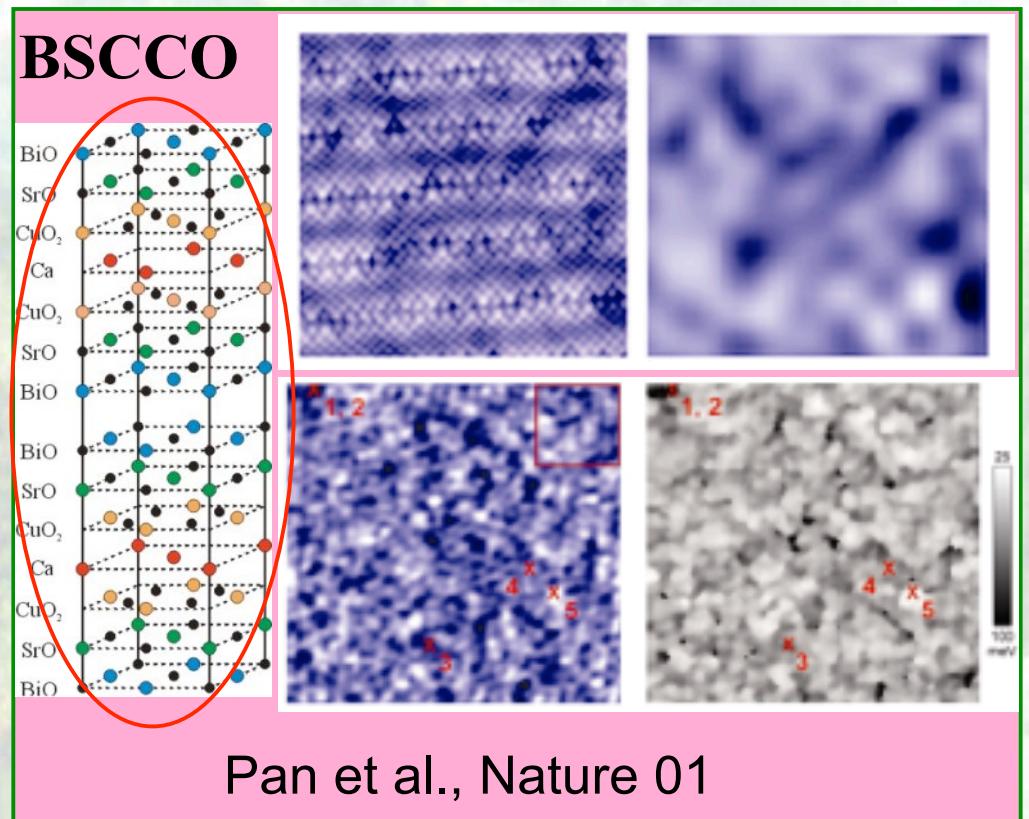
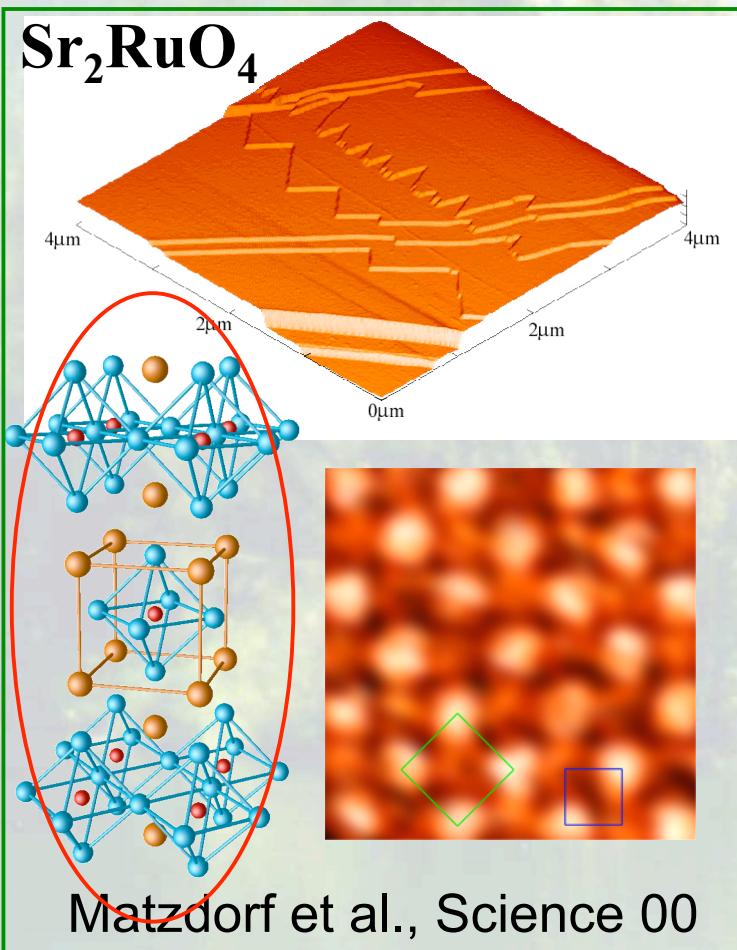
- High  $T_c$  superconductivity
- CMR
- Multiferroic



- Phase Separation & Inhomogeneity

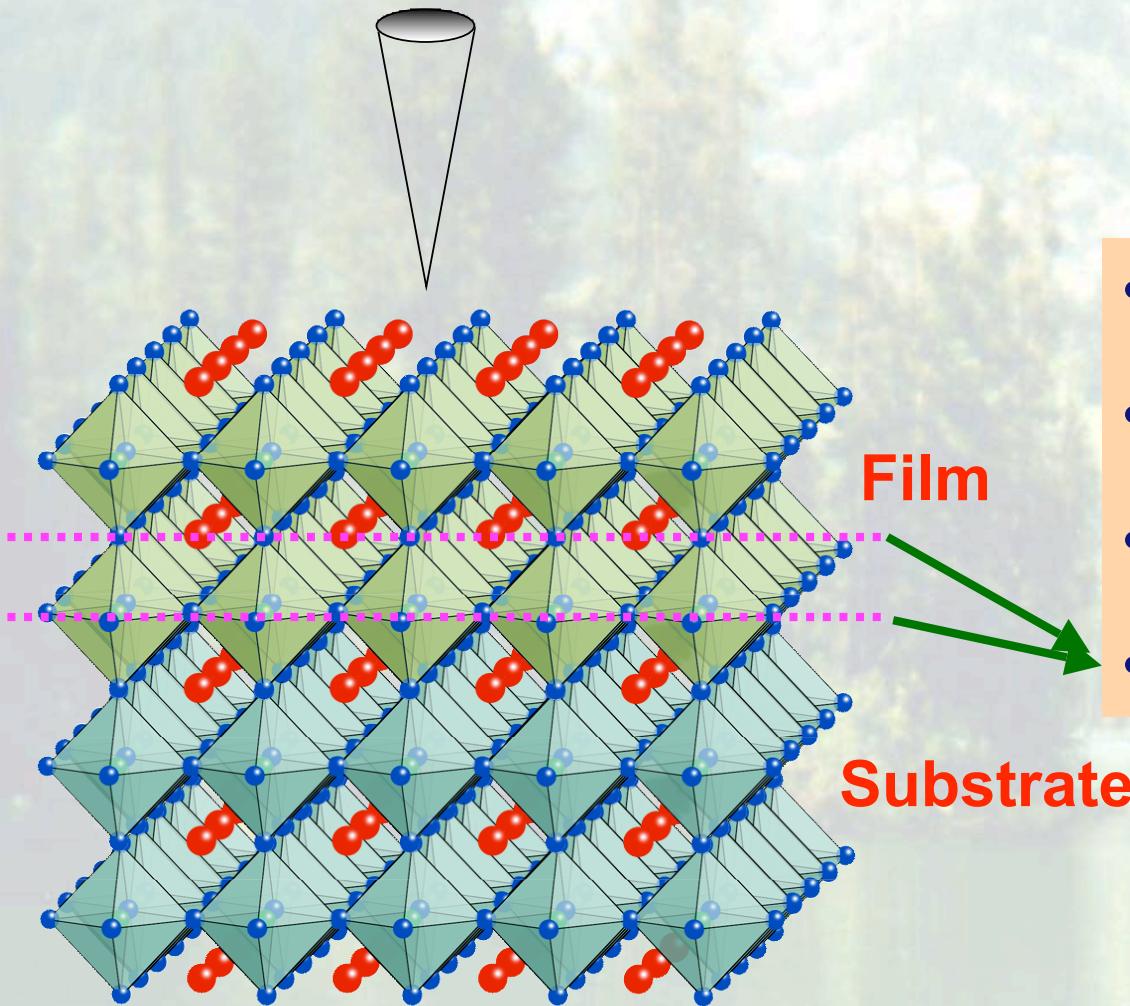
Role of dopants and defects?

# Scanning Tunneling Microscopy and Spectroscopy



**Layered TMO**

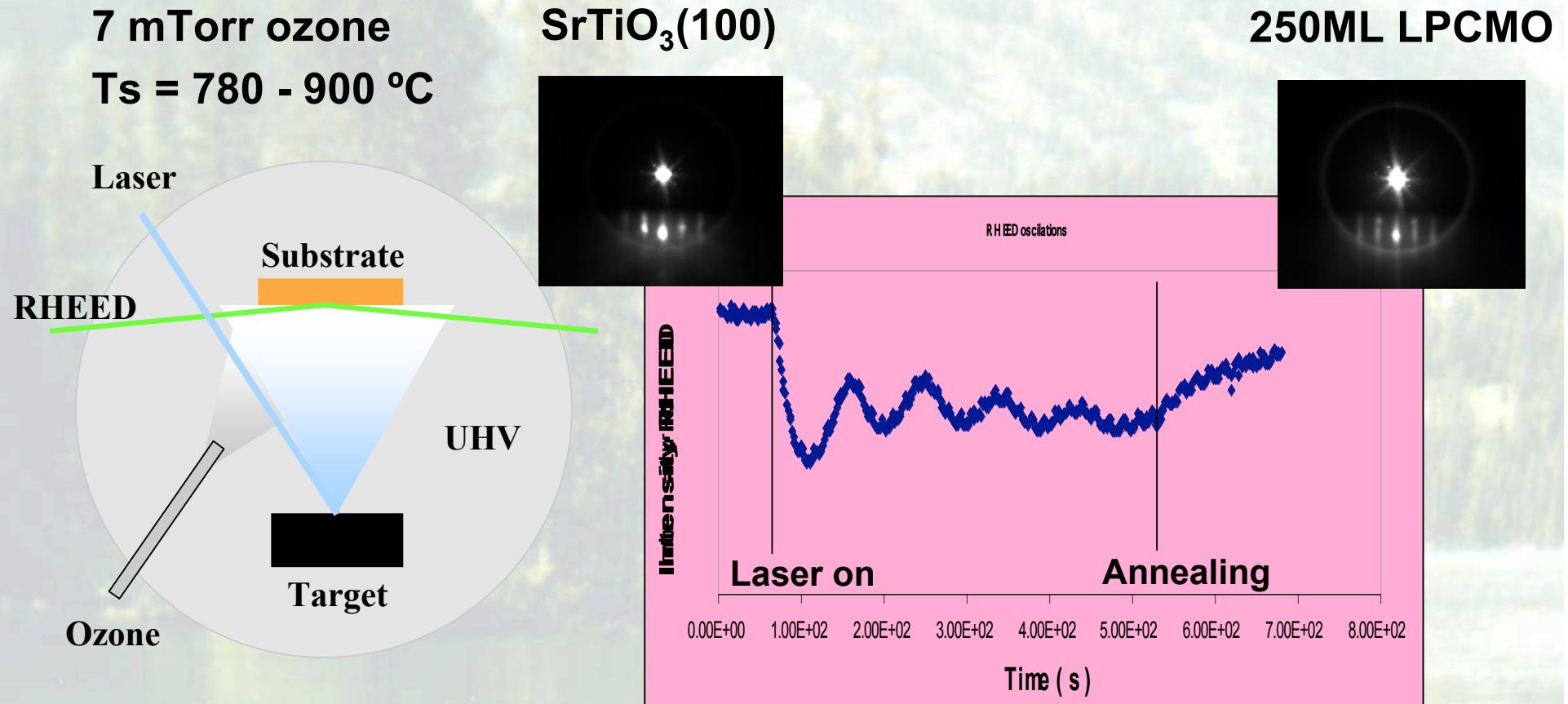
# **3D TMO : In-situ Growth of High-quality, Epitaxial Thin Films**



**Control of**

- Clean Surface
- Thickness
- Lattice Strain
- Surface Termination

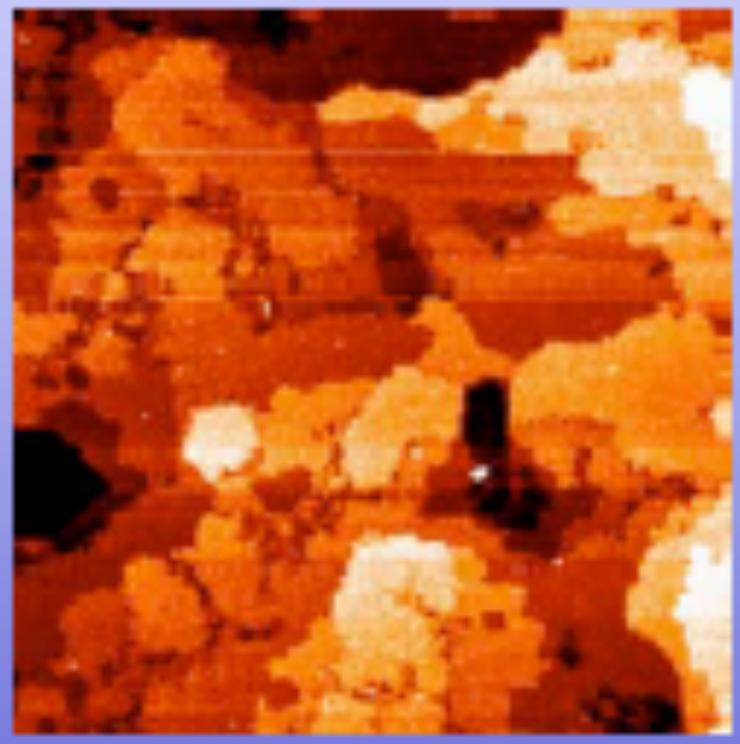
# In-situ Growth by Laser-MBE



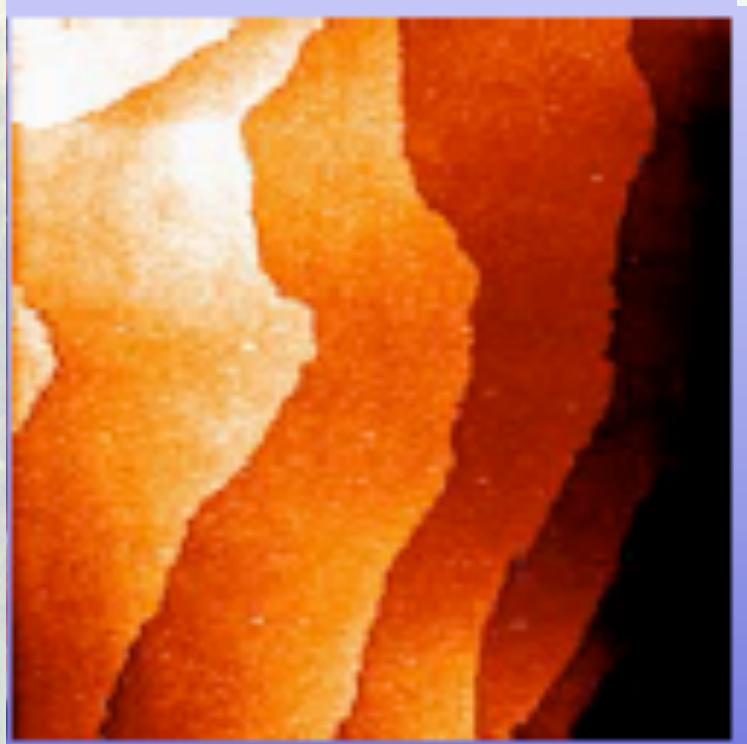
In-situ characterization ( $< 1 \times 10^{-10}$  torr) with MOKE,  
4-probe, LEED/AES, and VT STM/AFM

# Surface Topography of Different Growth Mode

Layer-by-layer Growth  
Mode  
 $T_s=780\text{ }^\circ\text{C}$



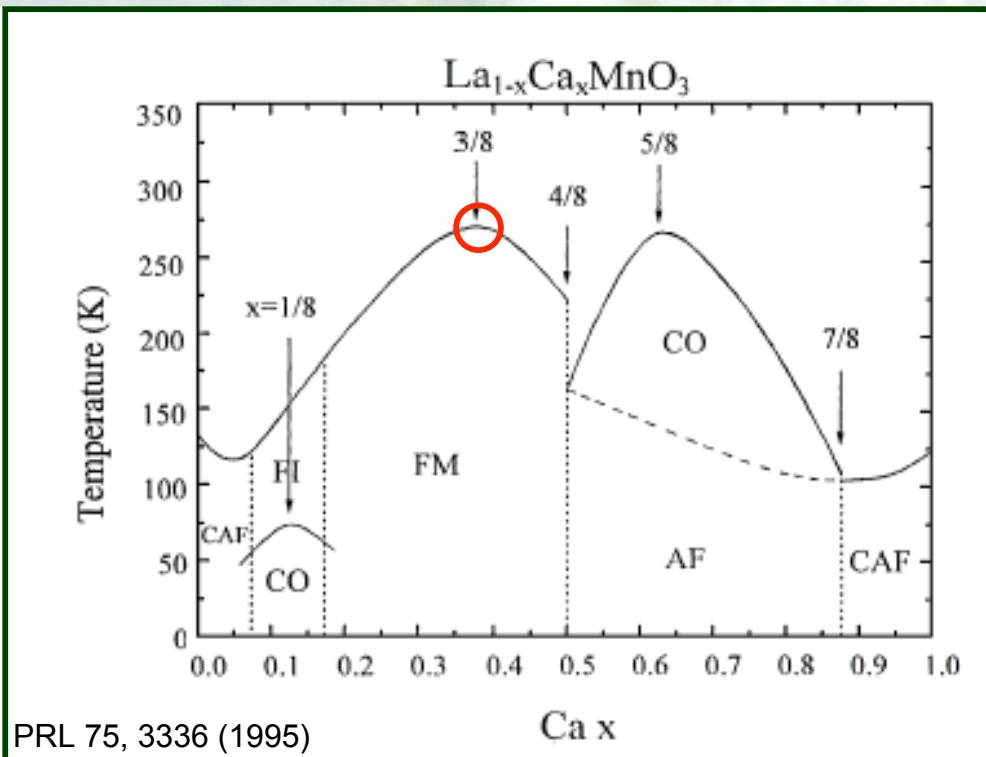
Step-flow Growth Mode  
 $T_s=850\text{ }^\circ\text{C}$



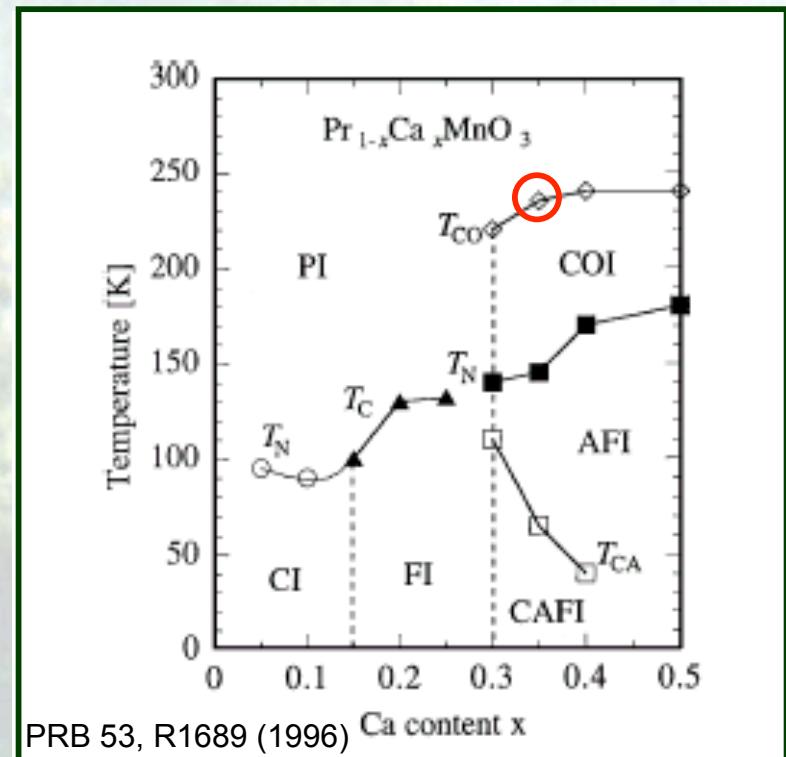
$(\text{La}_{5/8-y}\text{Pr}_y)\text{Ca}_{3/8}\text{MnO}_3$ ,  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ ,  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$

LaMnO<sub>3</sub>

# Mixing FM Metal and CO Insulator



PRL 75, 3336 (1995)



PRB 53, R1689 (1996)

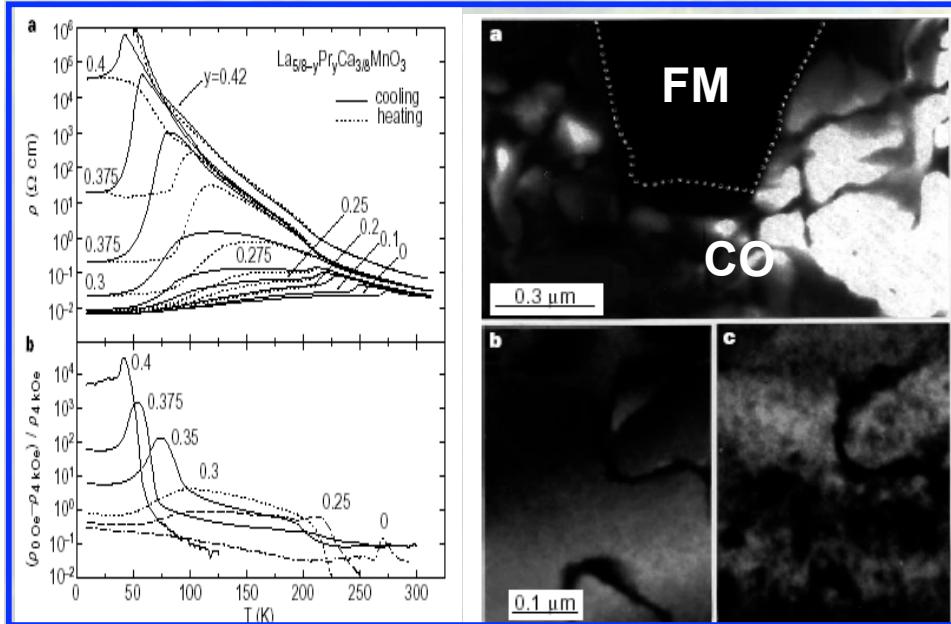
$\text{La}_{5/8}\text{Ca}_{3/8}\text{MnO}_3 T_c = 275 \text{ K}$

$\text{Pr}_{5/8}\text{Ca}_{3/8}\text{MnO}_3 T_{co} = 220 \text{ K}$

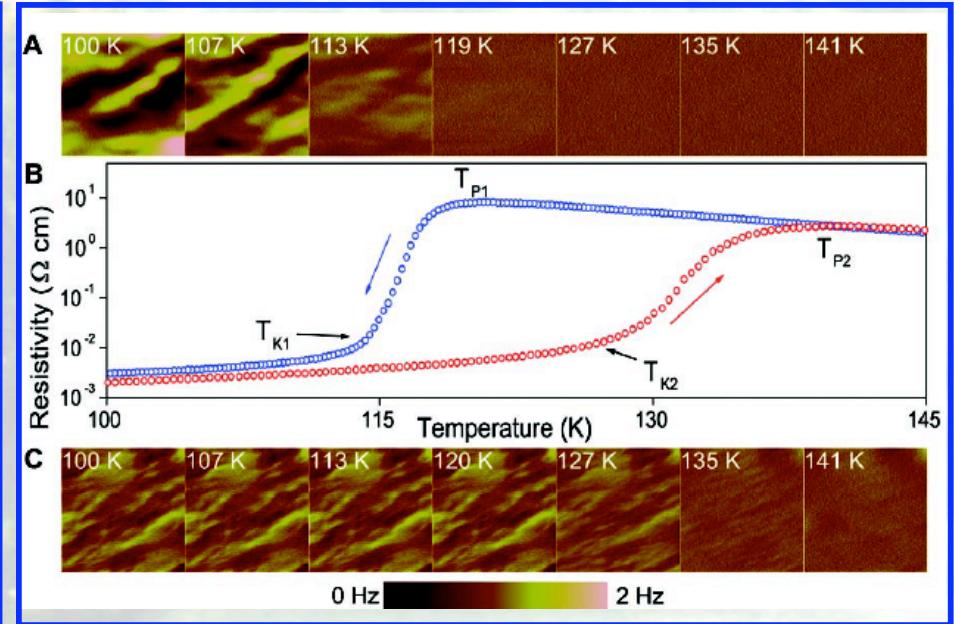
$(\text{La}_{5/8-y}\text{Pr}_y)\text{Ca}_{3/8}\text{MnO}_3$

# Large-scale Phase Separation in LPCMO

TEM



MFM

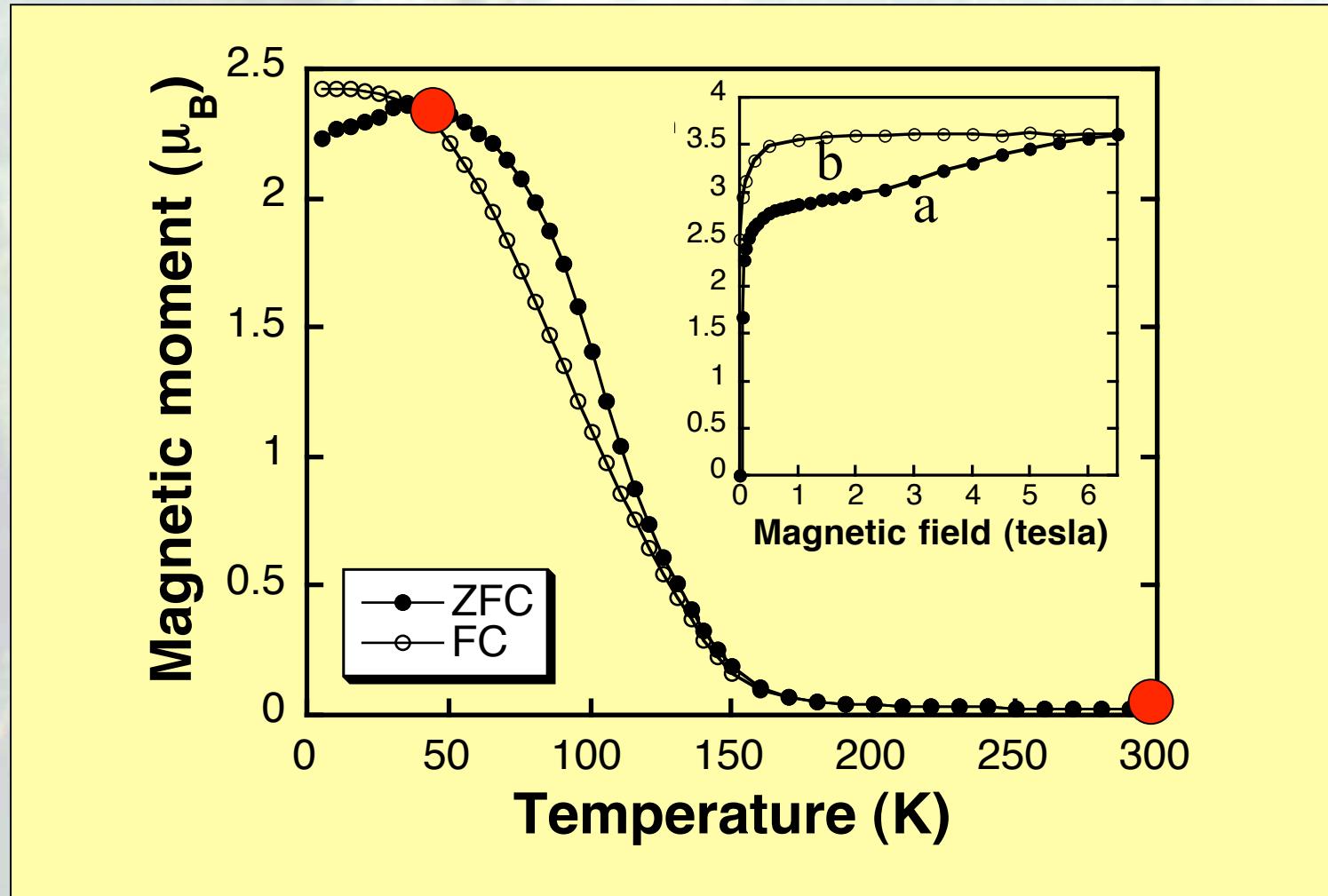


Uehara et al, Nature 399, 560 (1999)

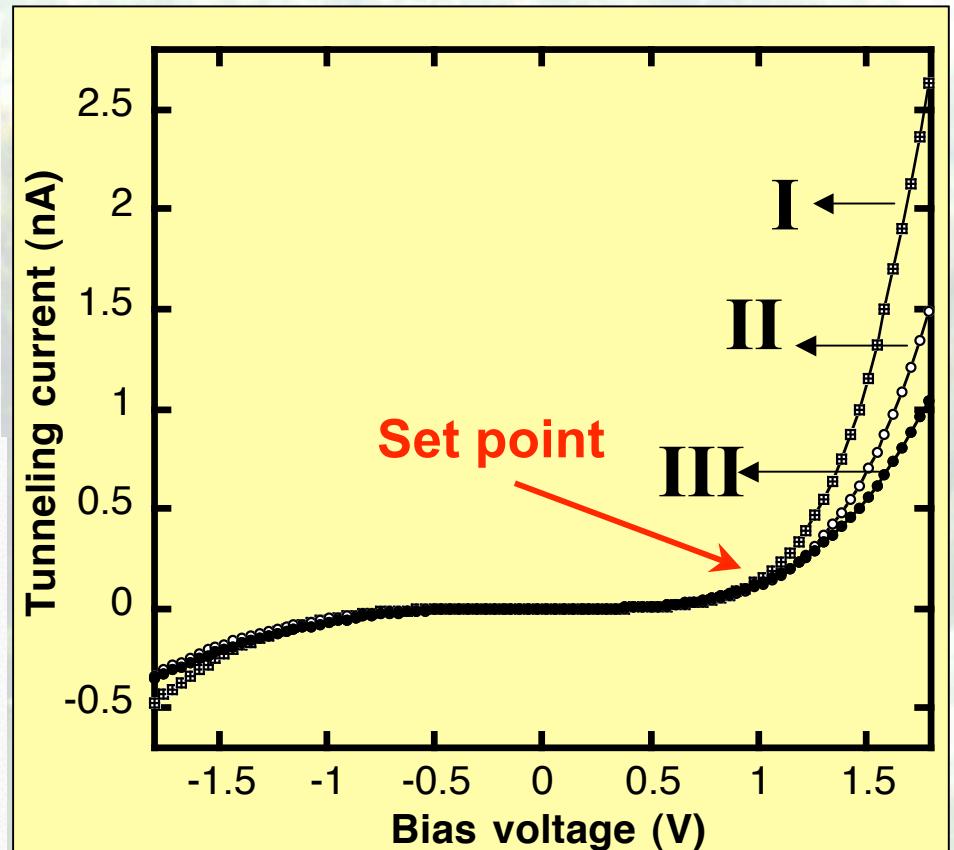
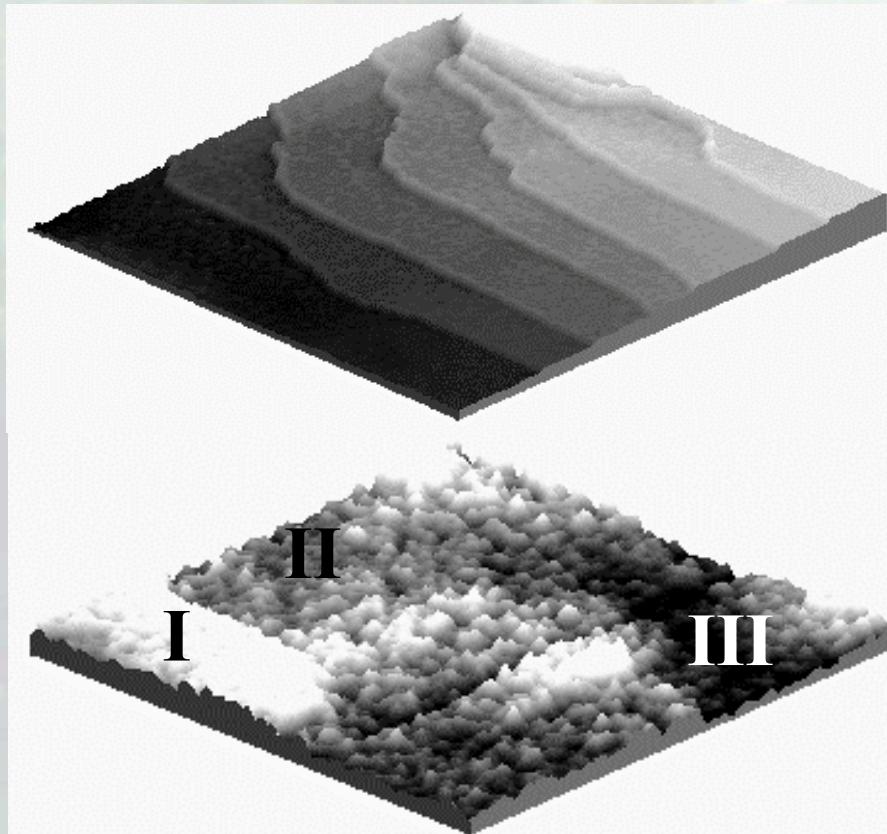
Zhang et al, Science 298, 805 (2002)

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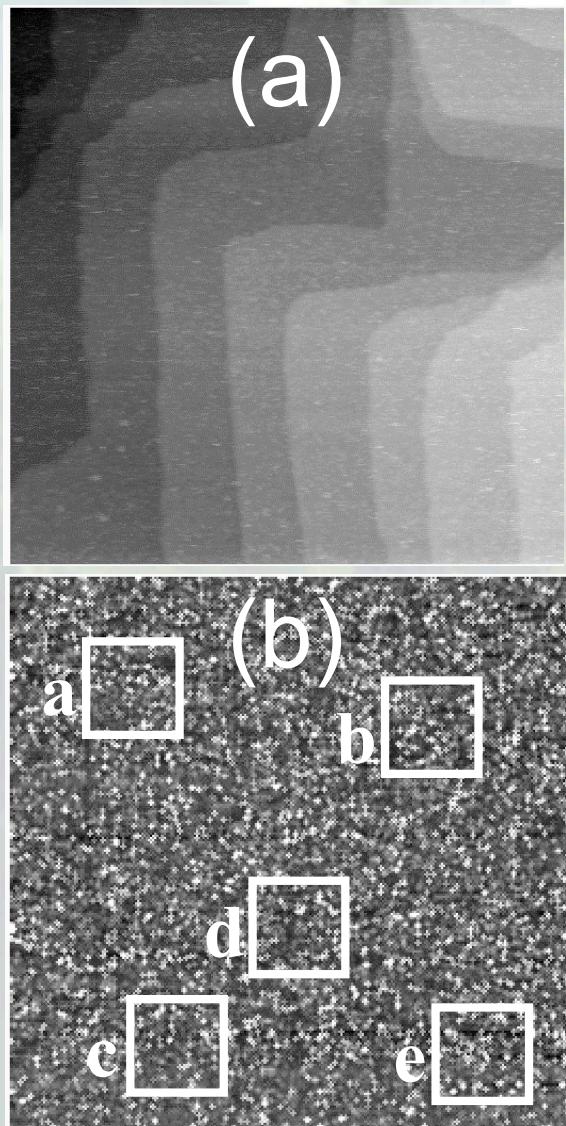
# Magnetic Properties by SQUID: 120 nm LPCMO



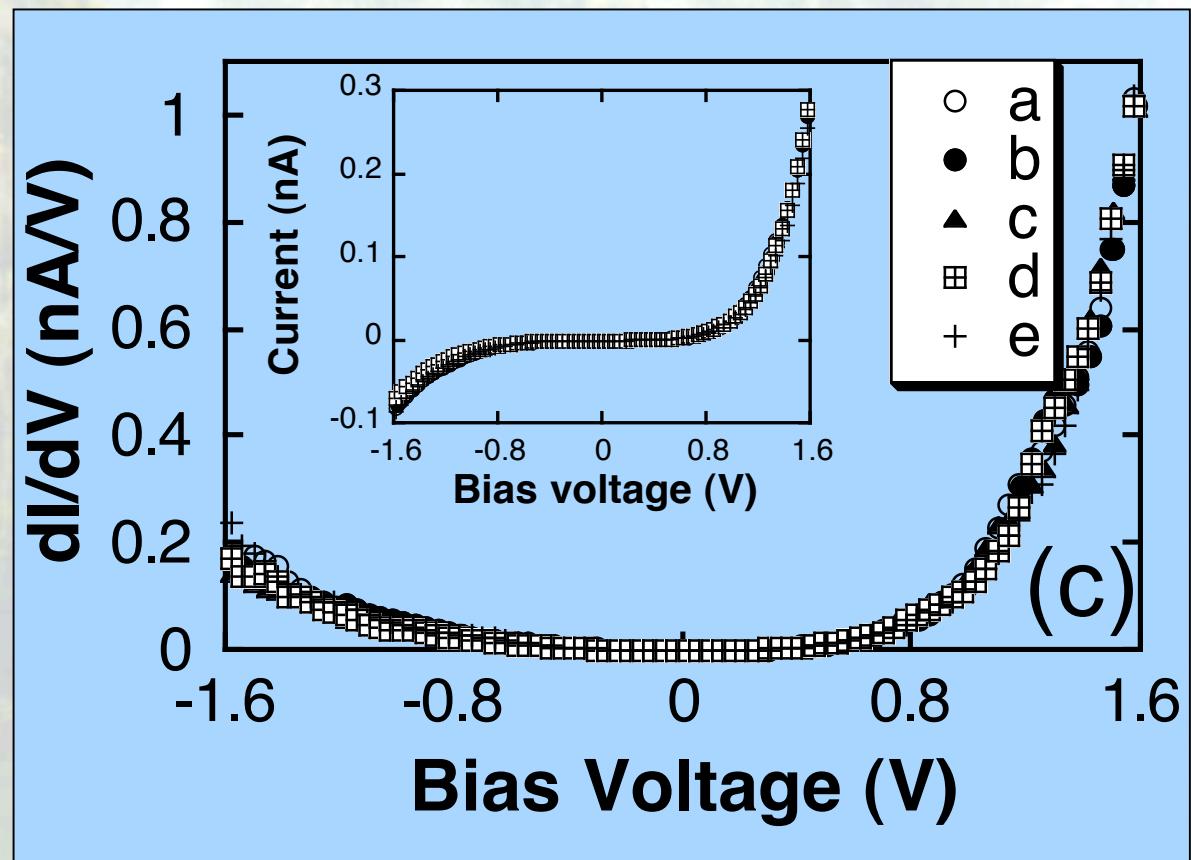
# Phase Separation in LPCMO Film at 60 K



**320 nm x 320 nm**



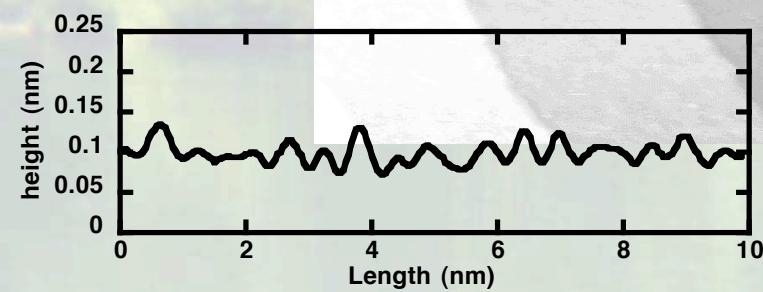
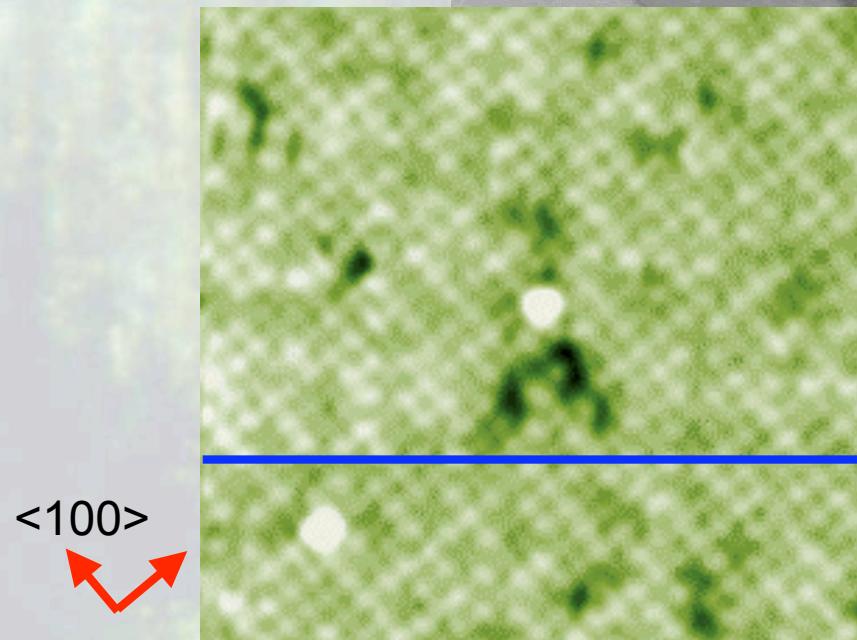
## STM and STS Above T<sub>c</sub> (RT)



# Atomic-scale Electronic Inhomogeneity above Tc

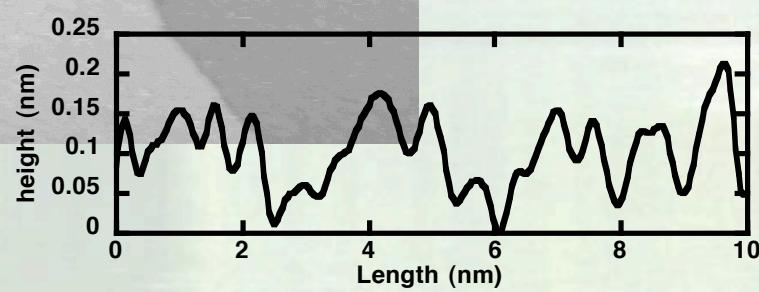
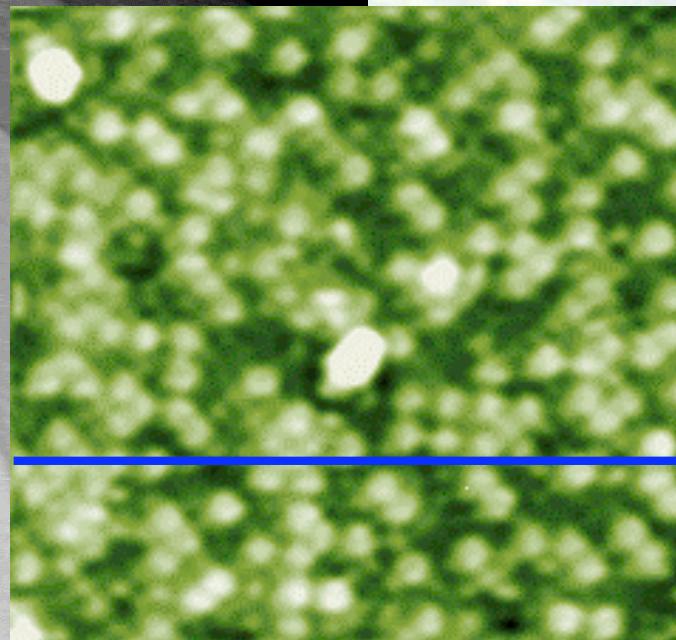
Occupied State Image

$V_{\text{bias}} = 1.5\text{V}$ ,  $I_t = 20\text{ pA}$



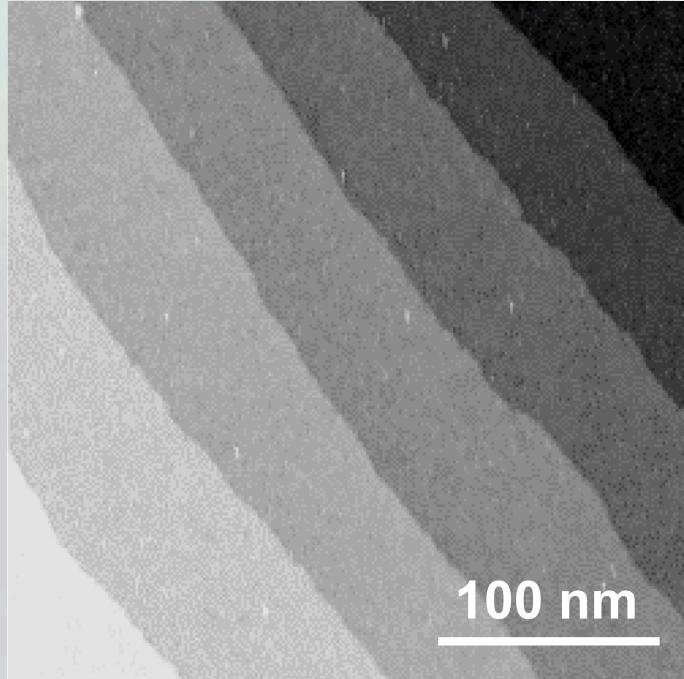
Empty State Image

$V_{\text{bias}}$   ,  $I_t = 50\text{ pA}$

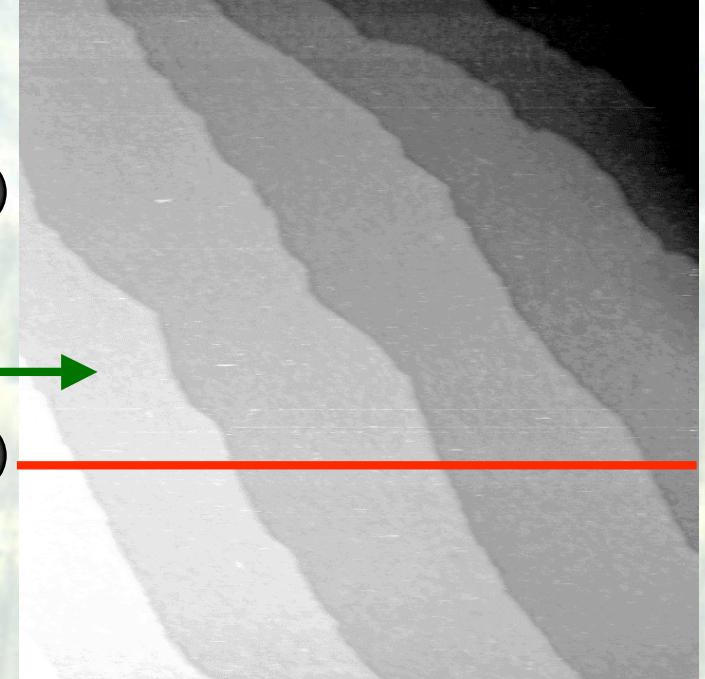
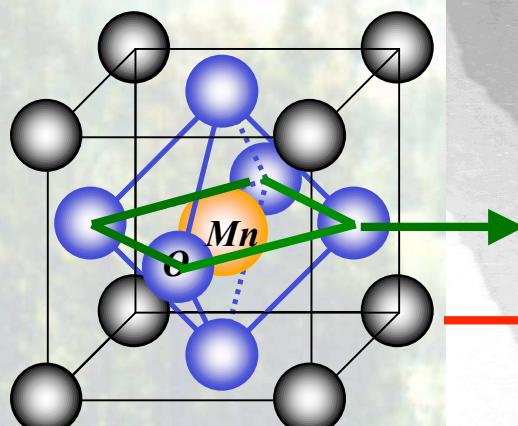


# Growth Unit Cell by Unit Cell

STO substrate



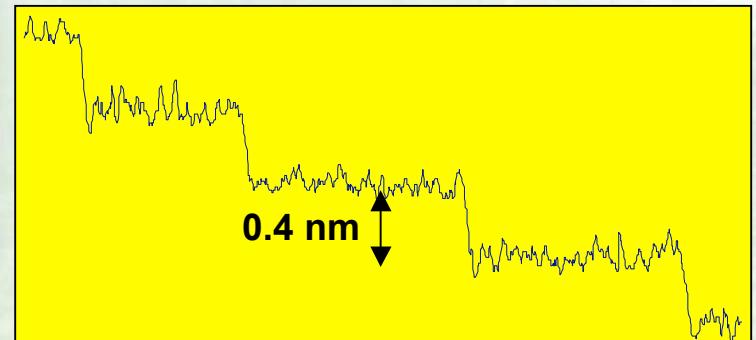
LPCMO Film



Ti-O termination

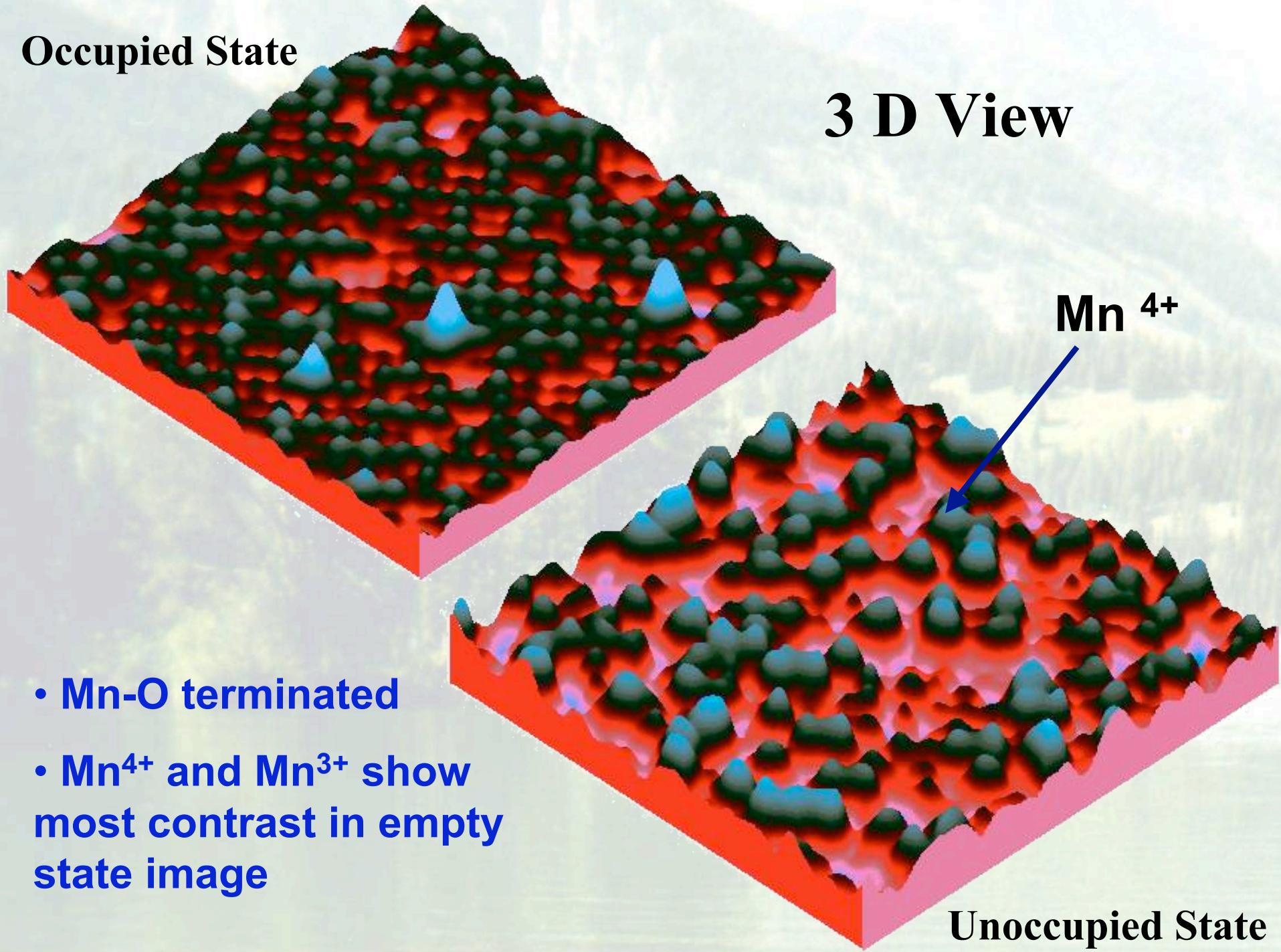
Kawasaki et al., Science (94)

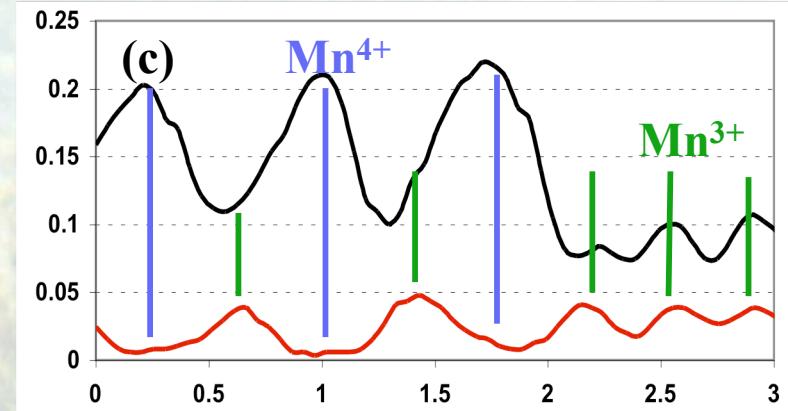
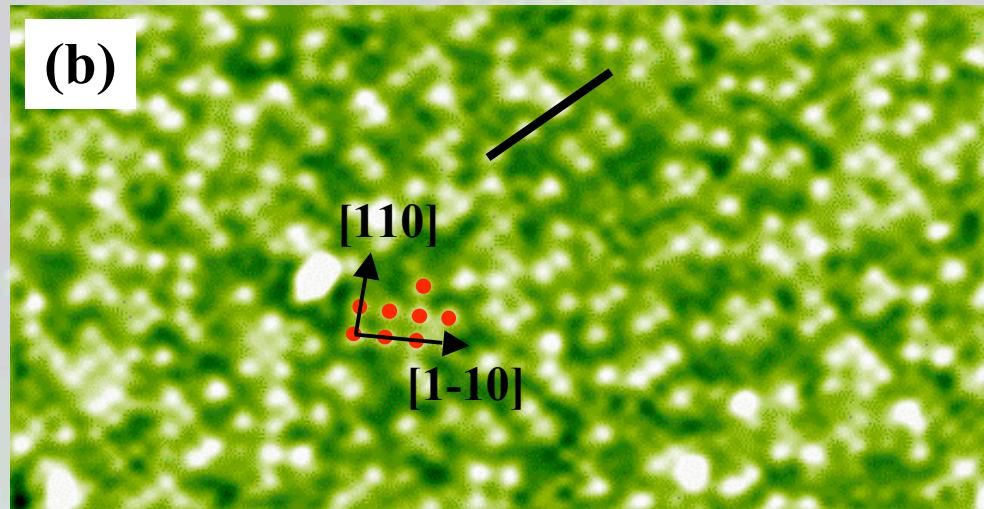
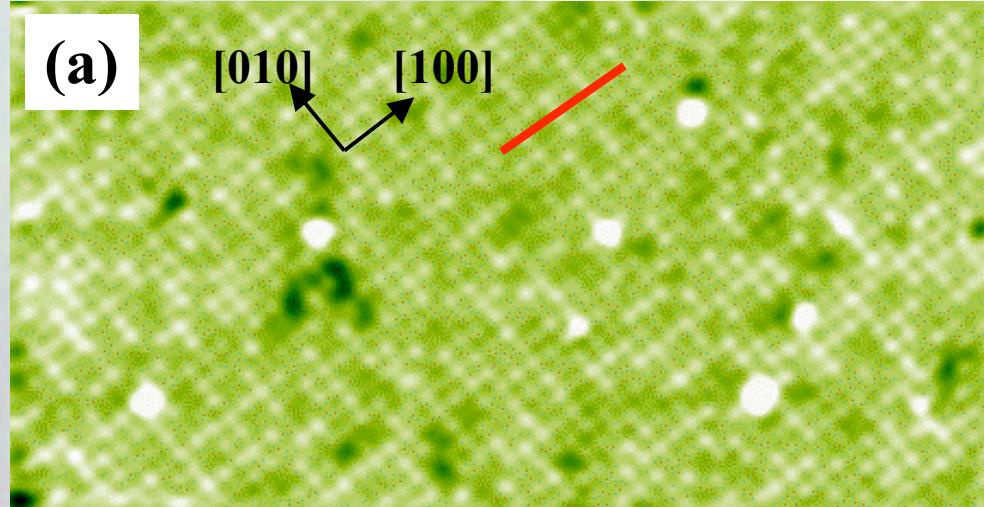
Mn-O termination



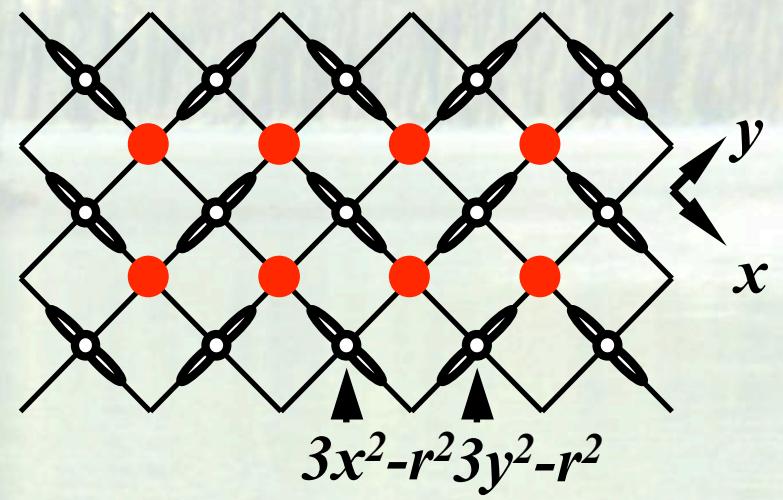
Occupied State

3 D View



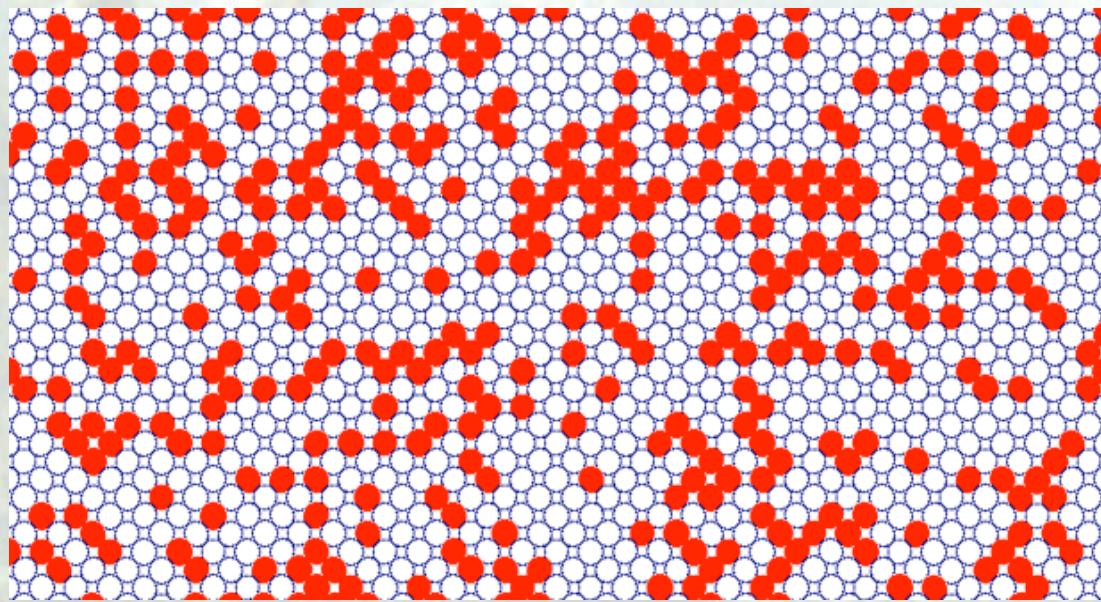


**(d) Charge Order Structure**

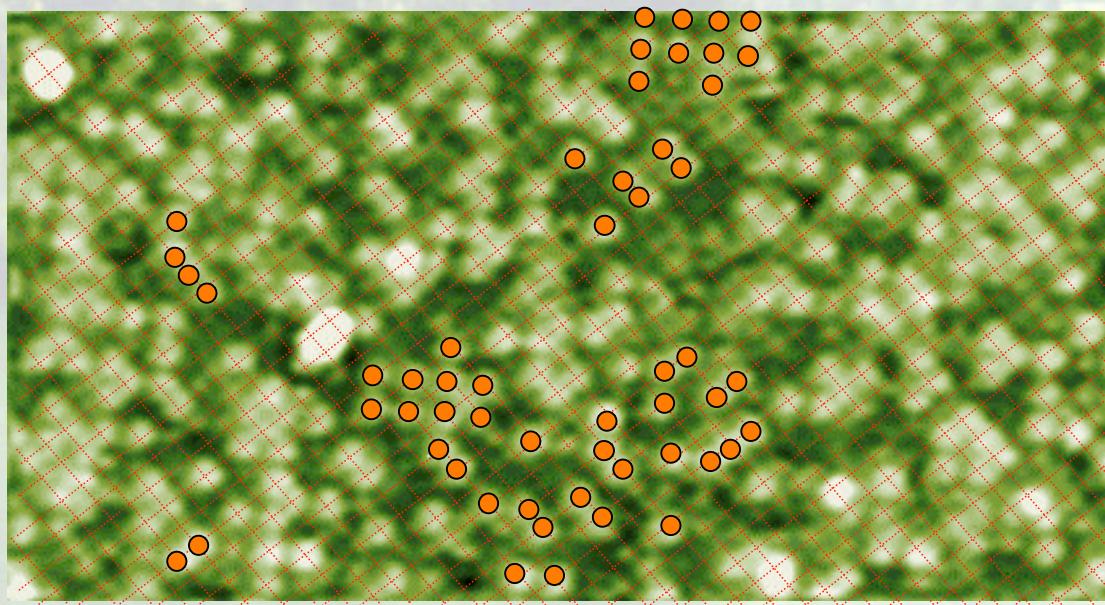


# Random Distribution vs. Experimental Data

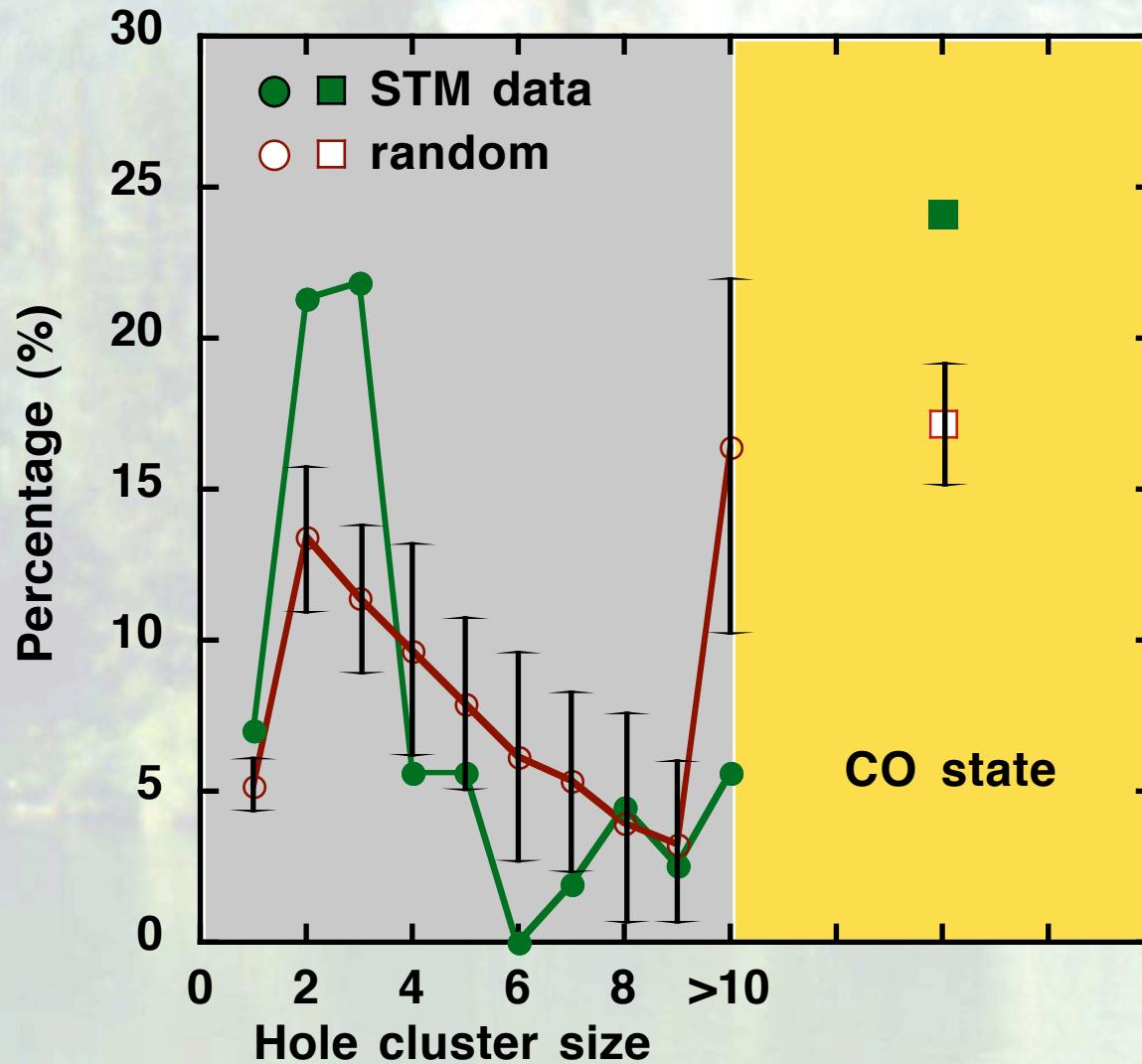
random



STM



# Correlated Polarons



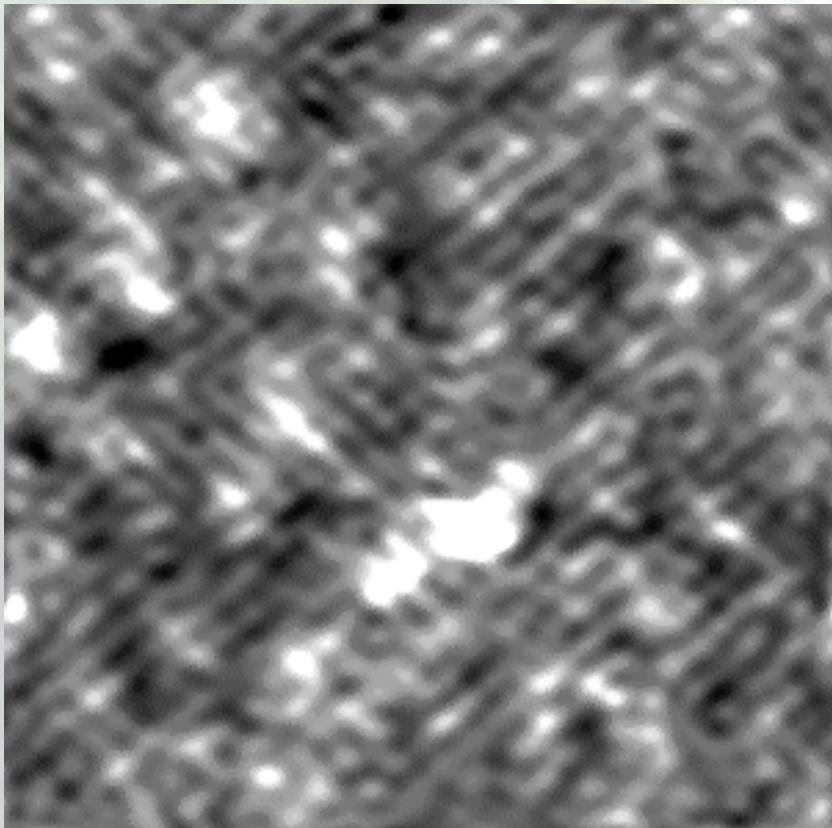
**STM data**

- prefer charge order
- prefer short range polaron correlation

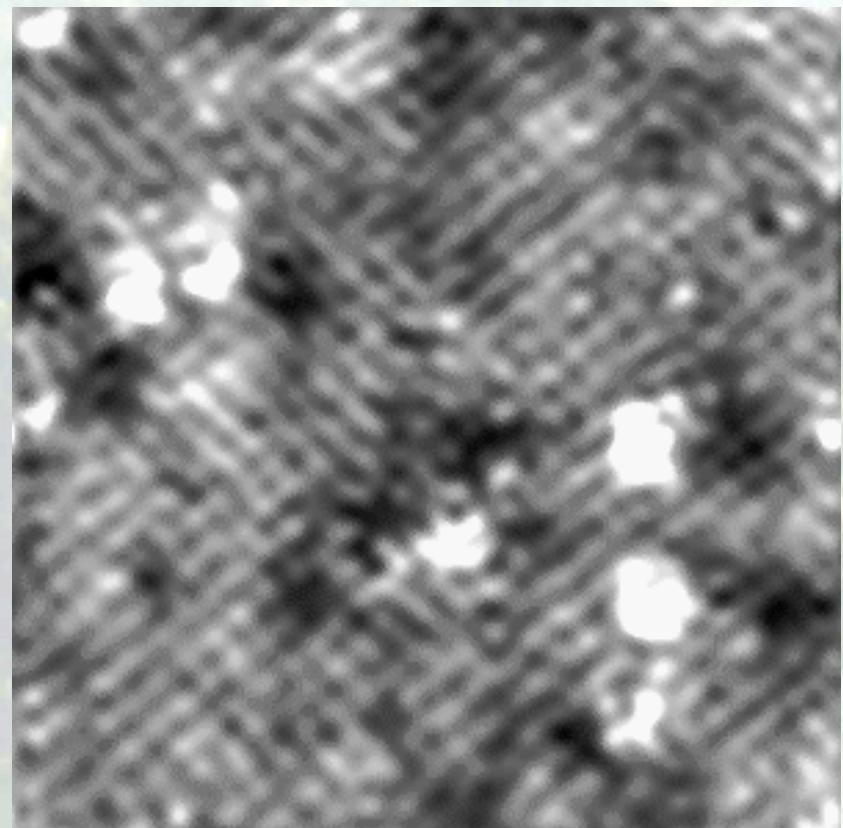
PRL (in press)

# Undoped LaMnO<sub>3</sub>/Nb-SrTiO<sub>3</sub> (001)

Occupied State ( $V_b=1.513\text{ V}$ )

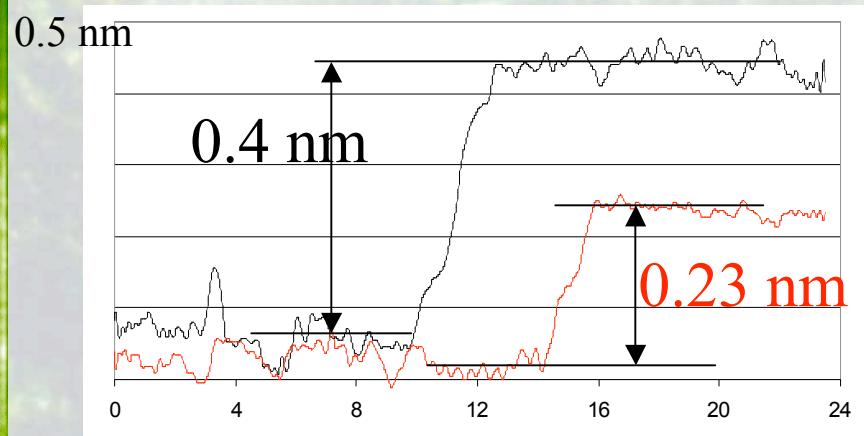
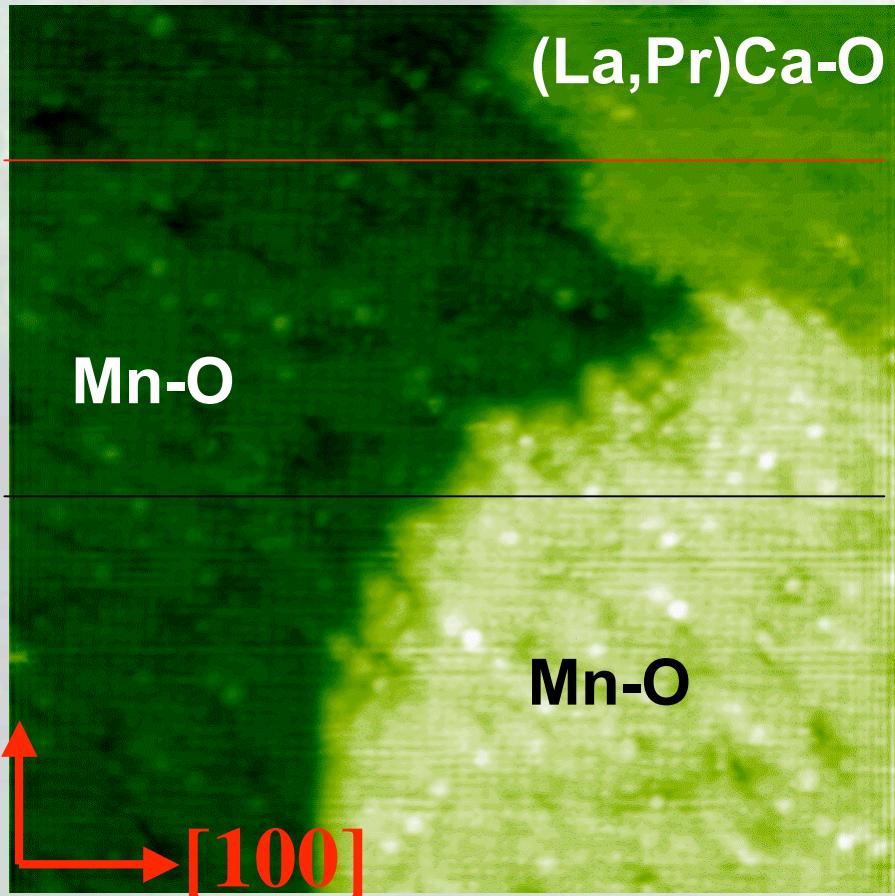


Empty State ( $V_b=-1.544\text{V}$ )

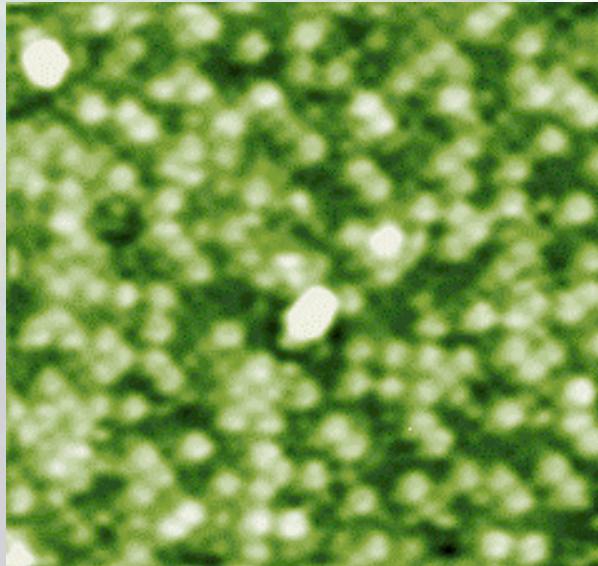


18x18 nm,  $I_t = 0.030\text{ nA}$

# Termination Layer Control

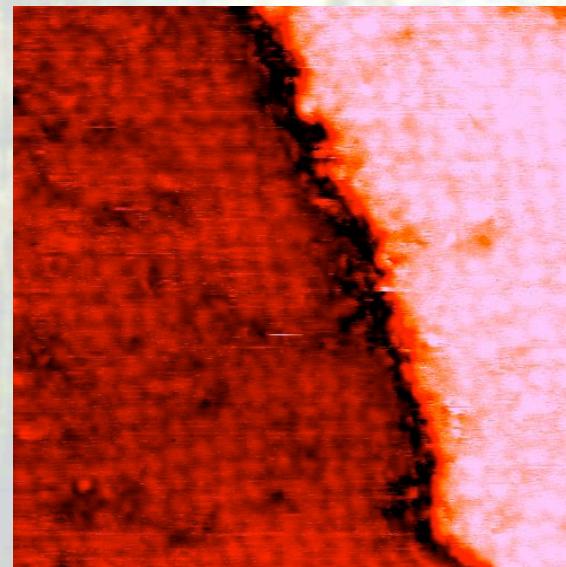


LPCMO: CO cluster

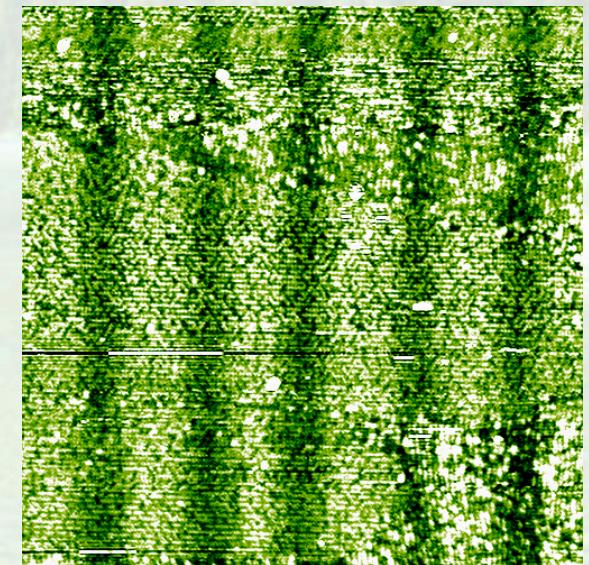


## Above T<sub>c</sub>: Paramagnetic Insulating

LCMO: Ca ordering

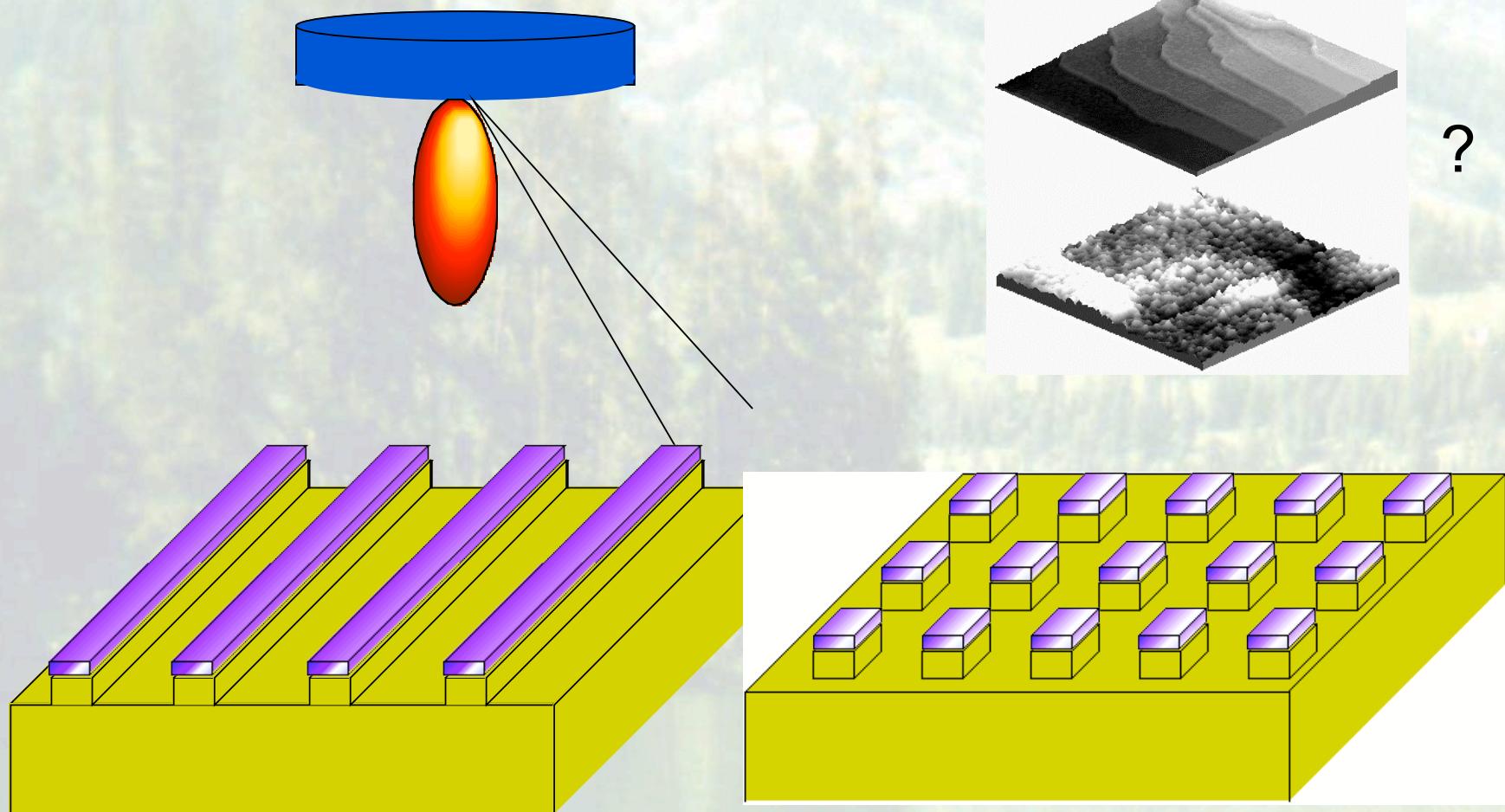


LSMO: Stripe phase



T dependence  
Field dependence  
Termination layer control

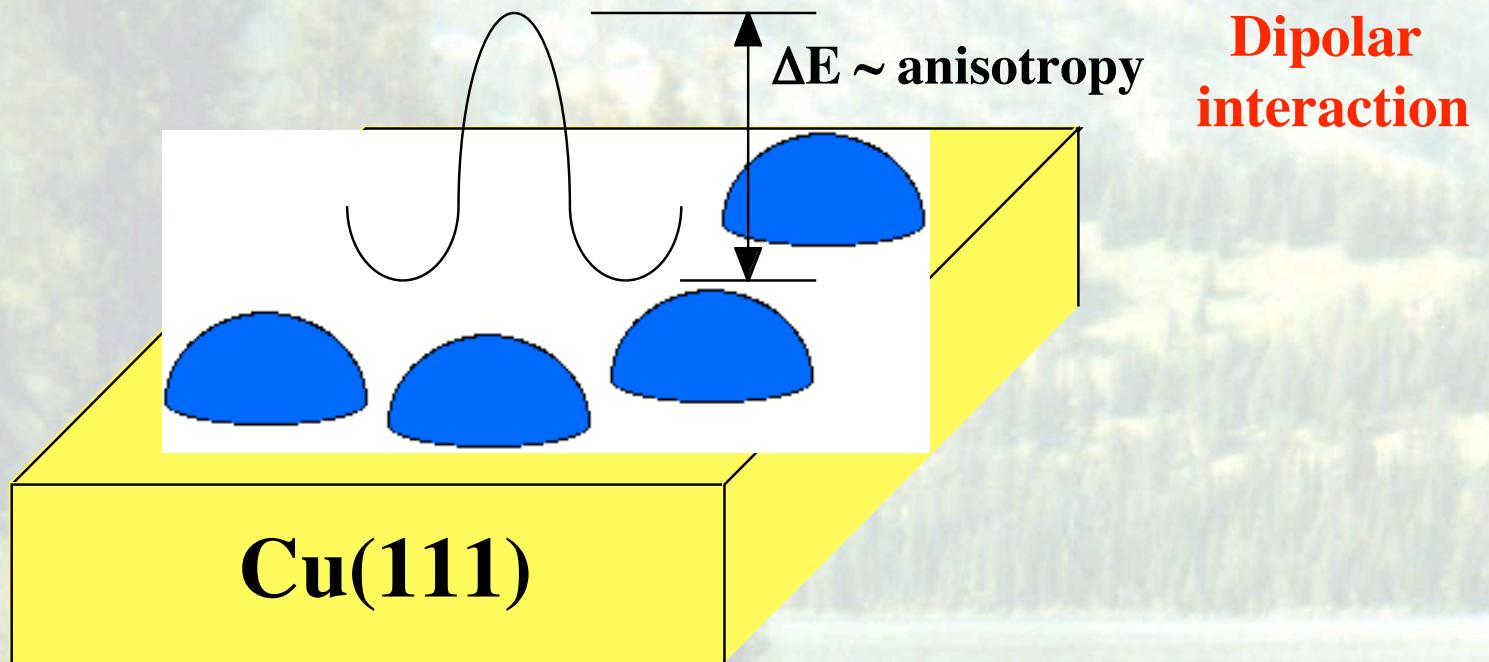
# Spatially Confined TMO Nanostructures



## **II. Superferromagnetic Nanodot Assemblies**

**Surface States Mediated Ferromagnetic  
Coupling Between Nanodots**

# Superferromagnetic Nanodot Assemblies



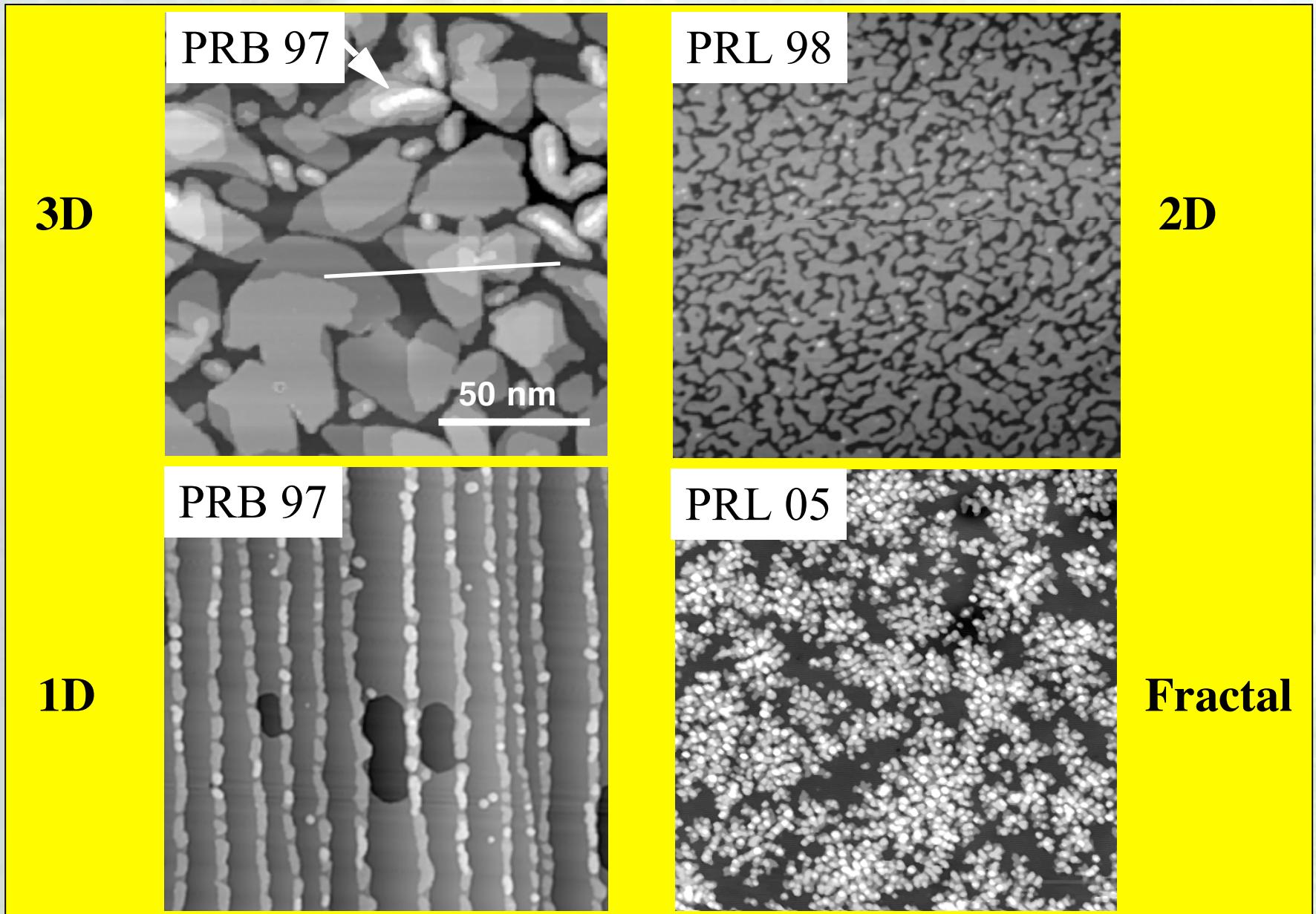
Superparamagnetism

$$M(t) = M_0 e^{-t/\tau}$$

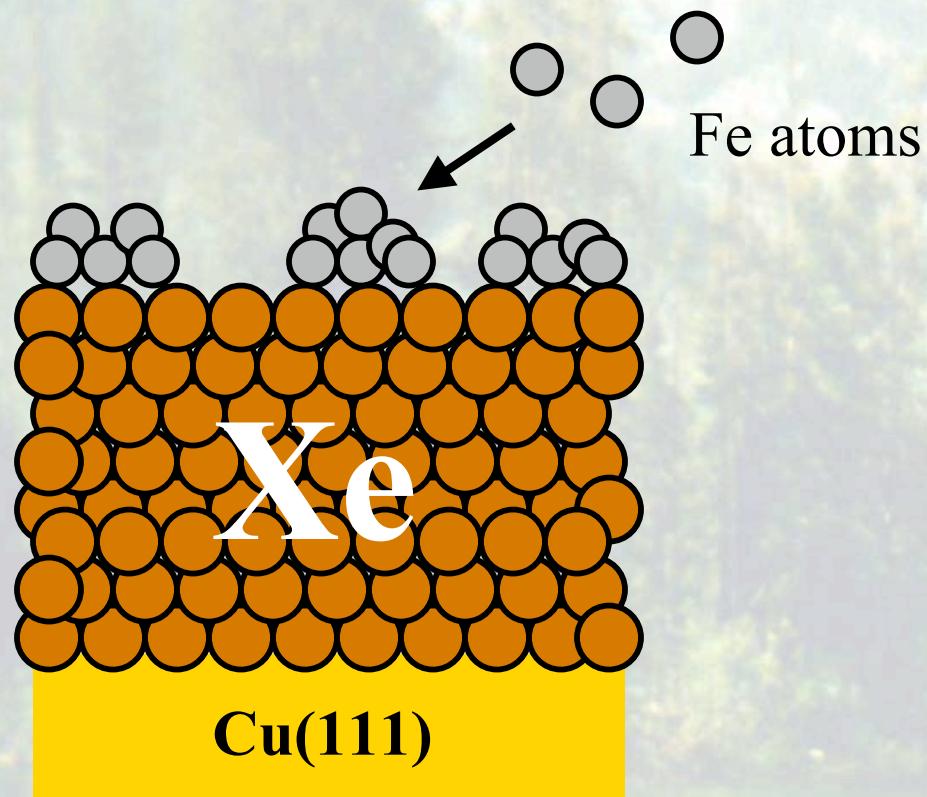
$$1/\tau = f_0 e^{-\alpha}, \quad \alpha = \frac{\Delta E}{k_B T}$$

Blocking  
temperature  $T_B$

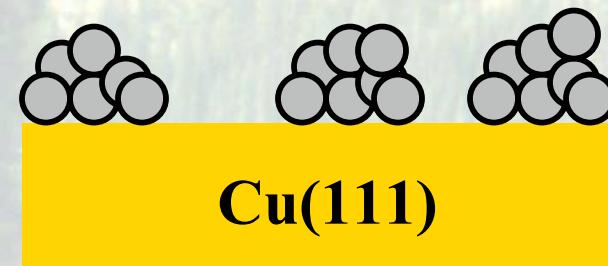
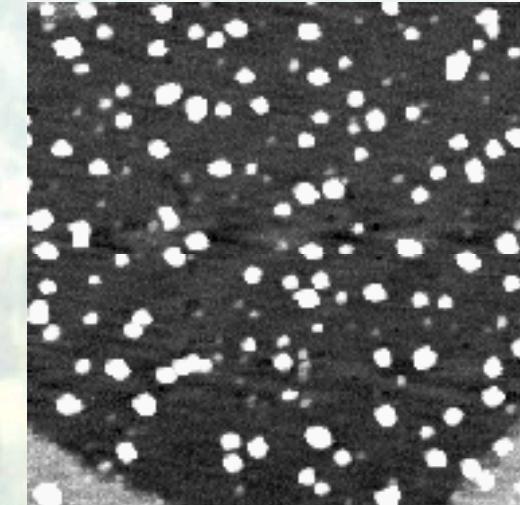
# Fe on Cu(111): Direct Deposition



# Buffer Layer Assisted Growth

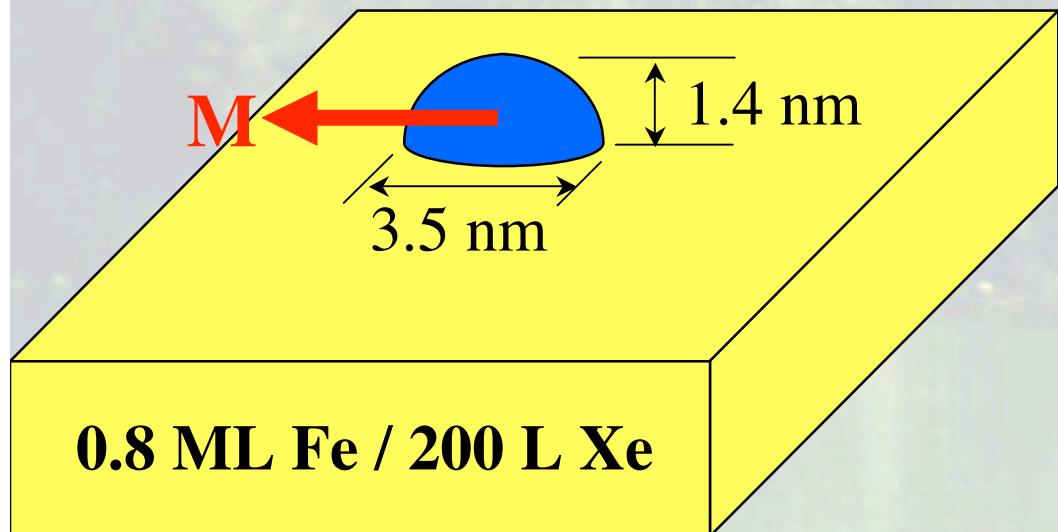
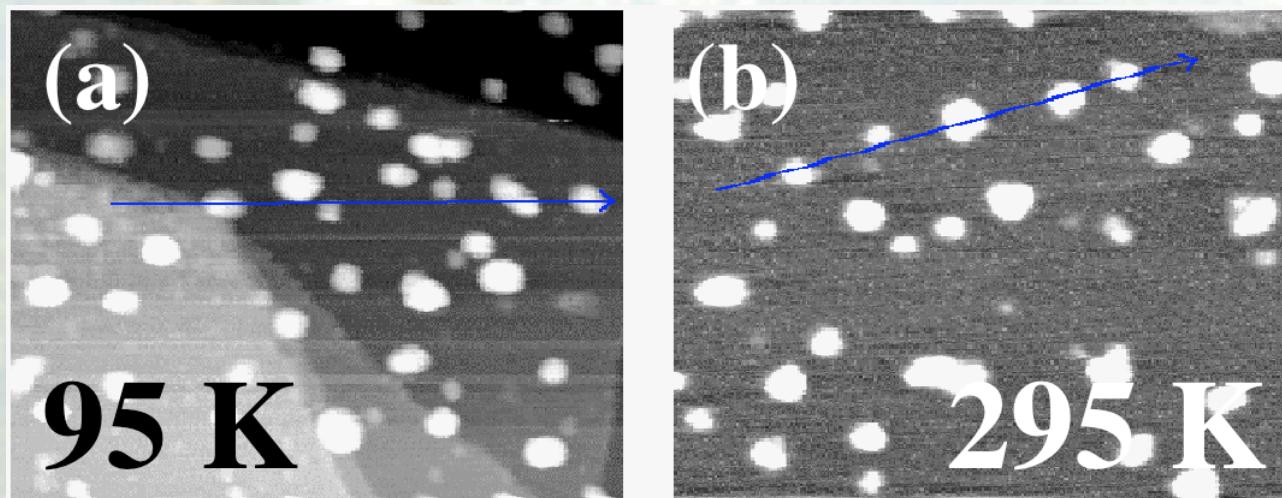


30 K

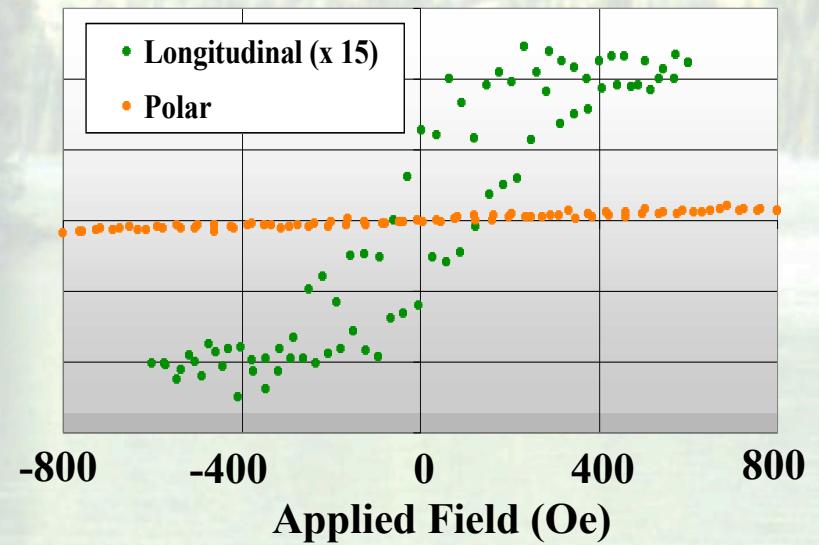


90 K

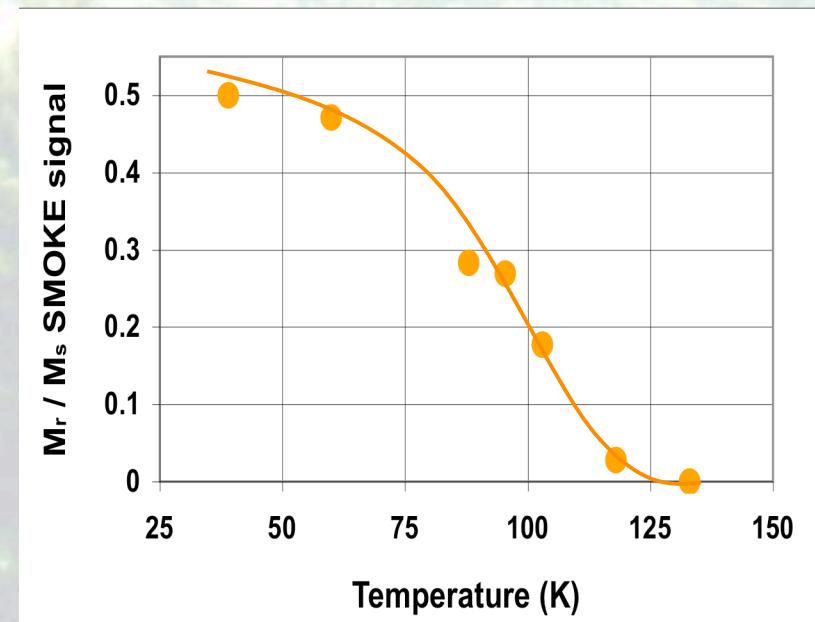
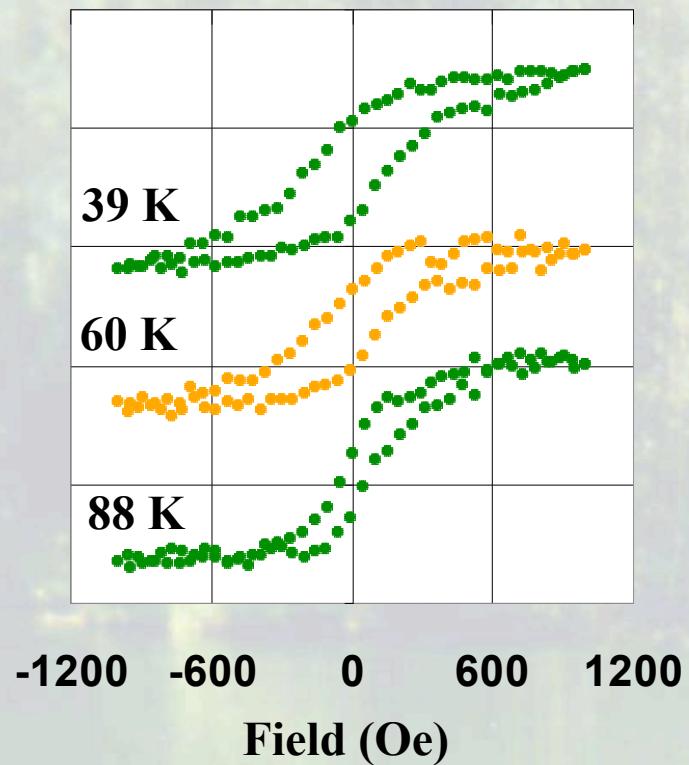
# Morphologically Stable



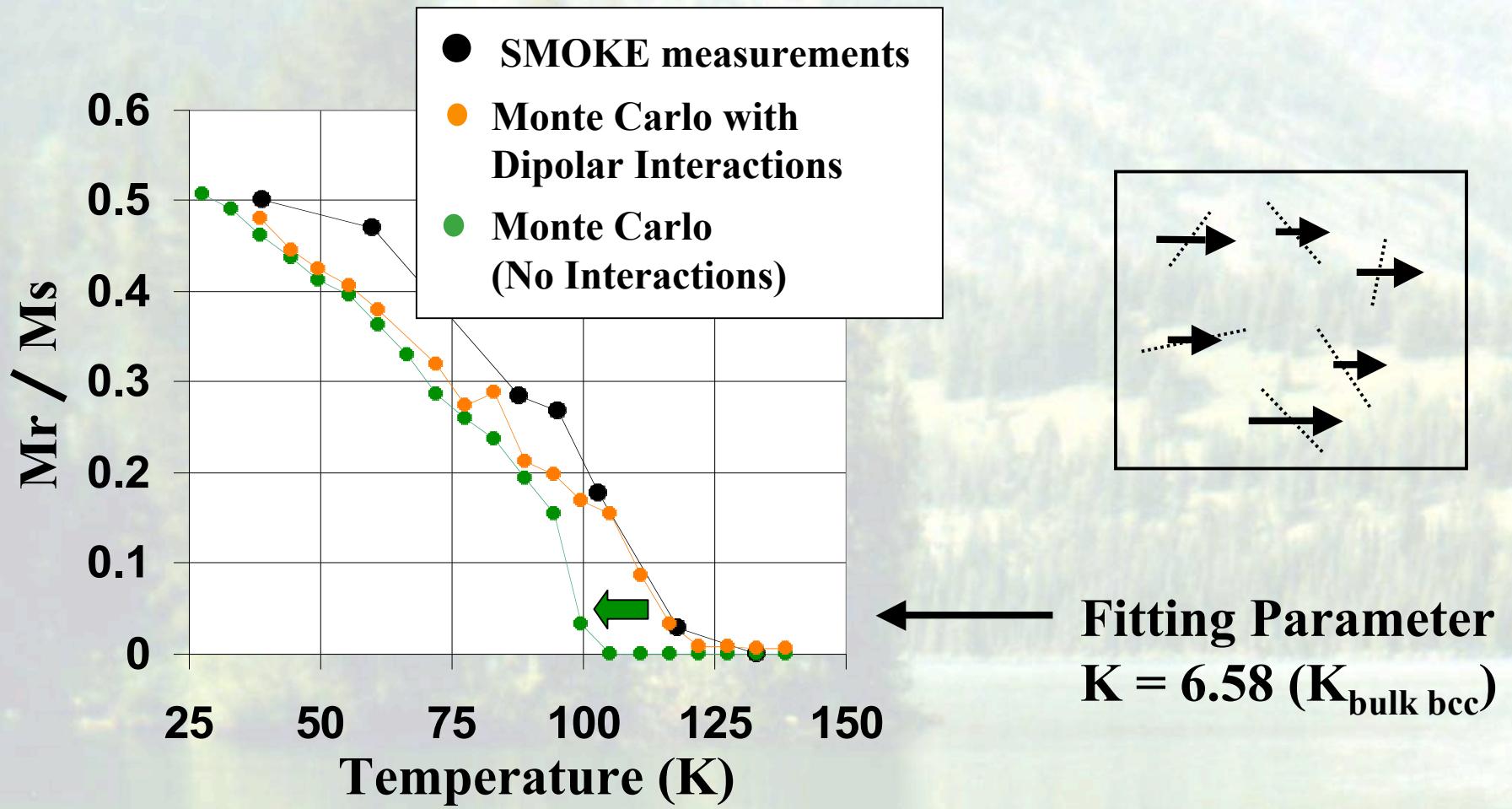
$T_B \sim 2$  K



# Unusually High “Blocking Temperature”



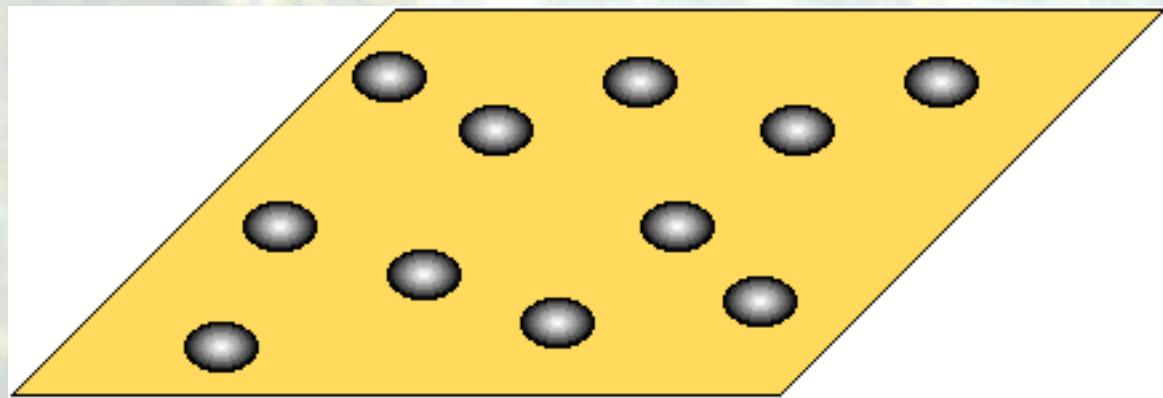
# Monte Carlo Simulations



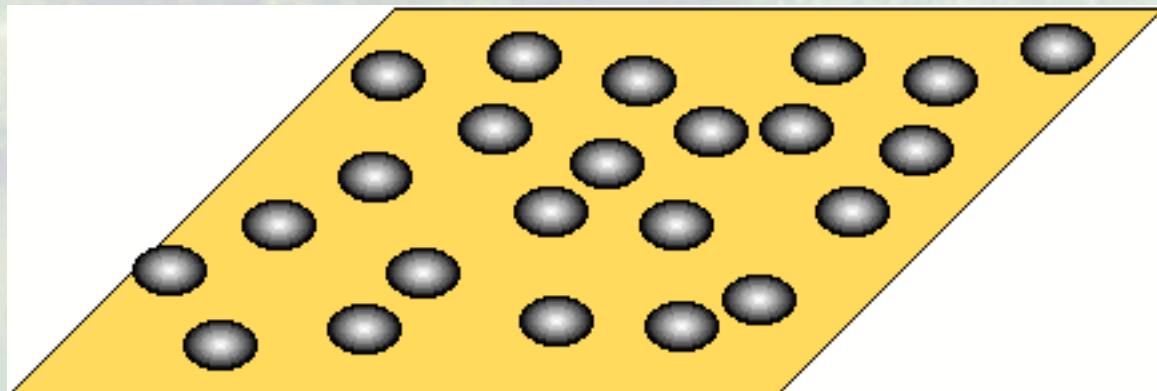
Dipolar interaction is not the major driving force!

H.K. Lee, T.C Schulthess, D.P. Landau

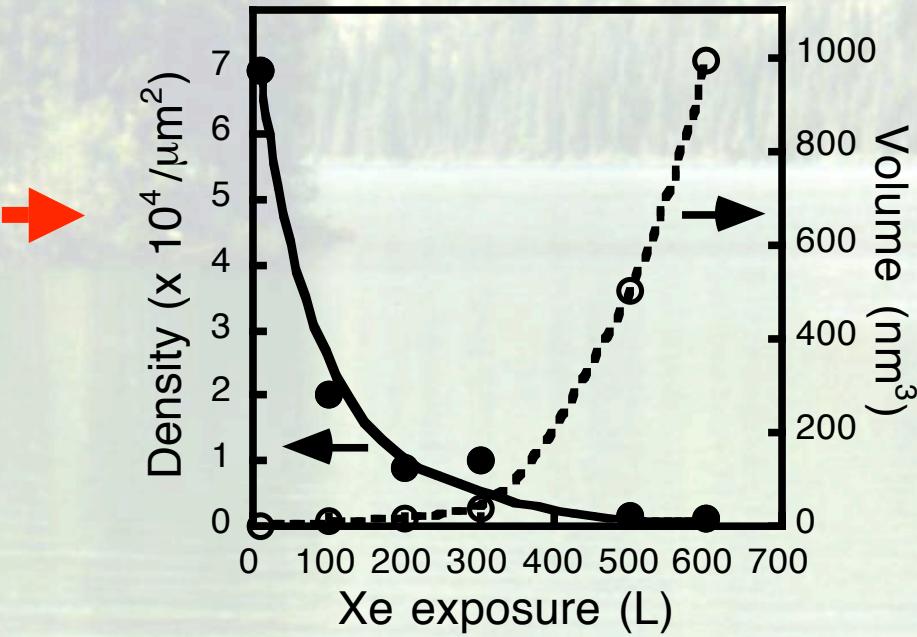
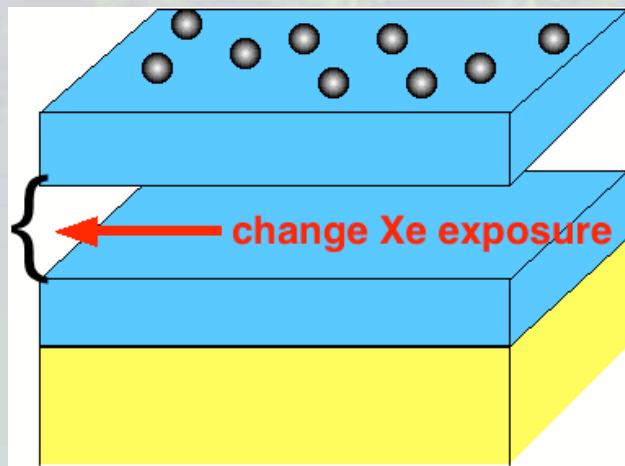
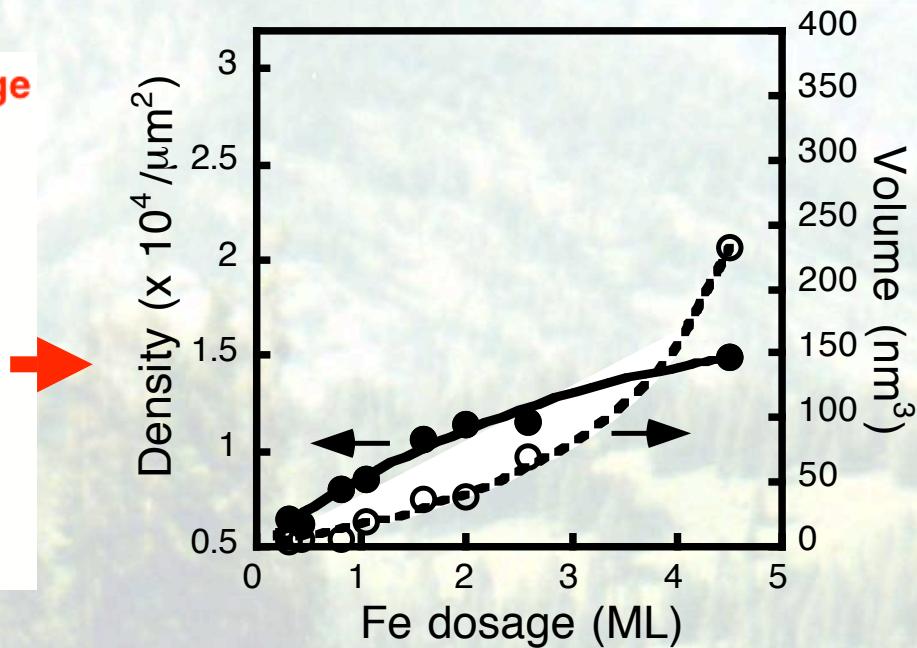
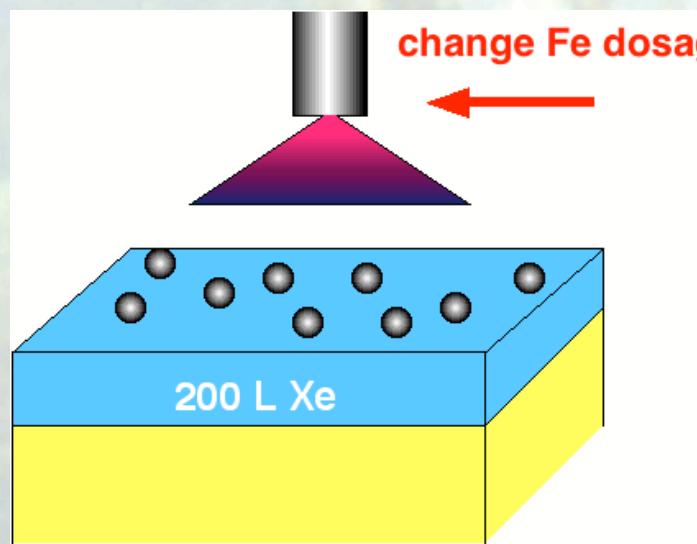
# To Study the Role of Interactions...



Vs.

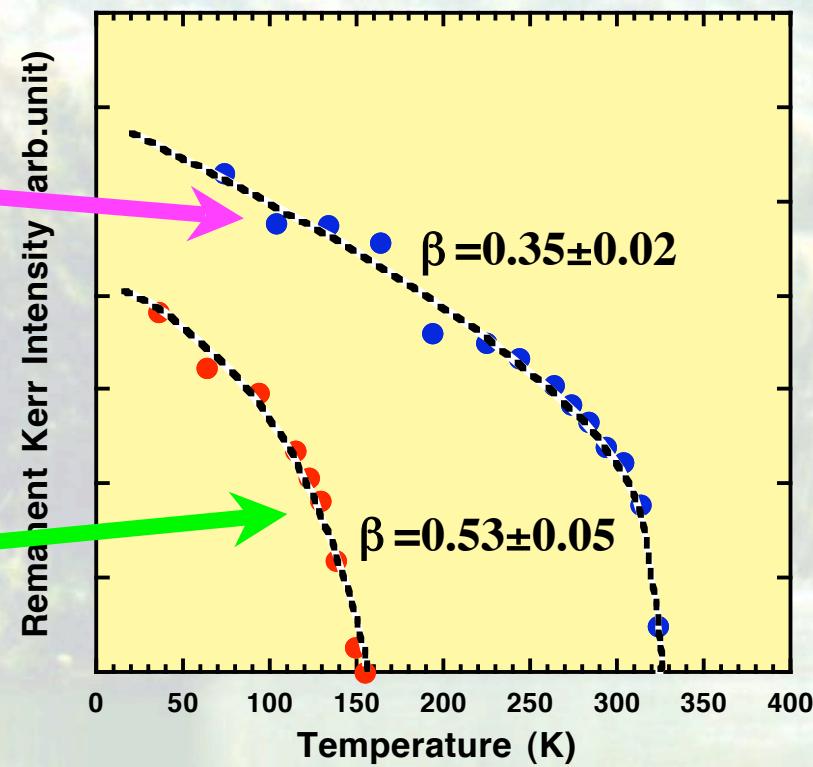
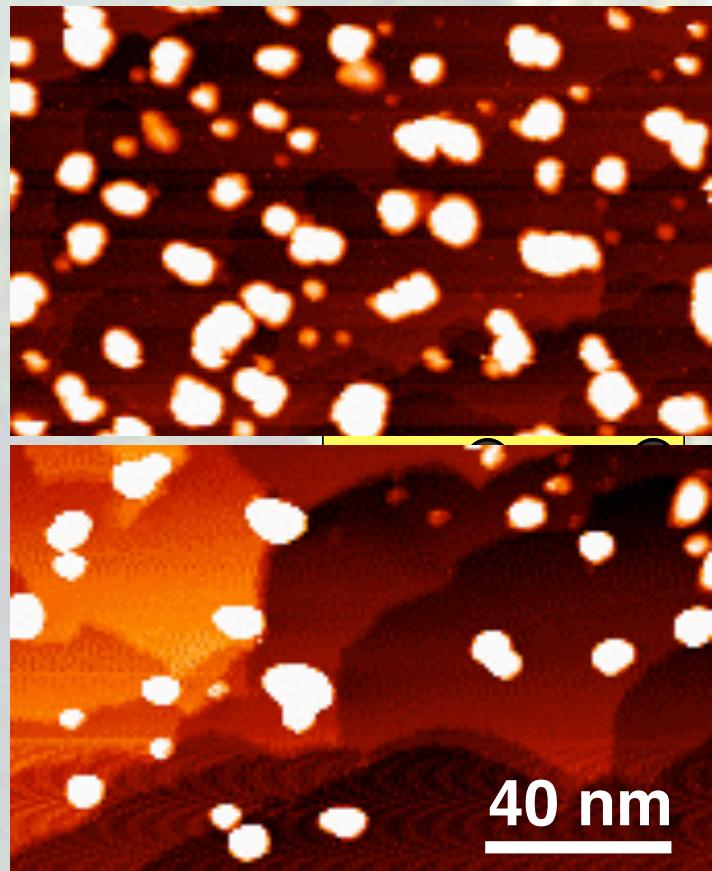


# Control the Size and Density



# Magnetic Behavior

Fixed Dot Size  
Varying Spacing

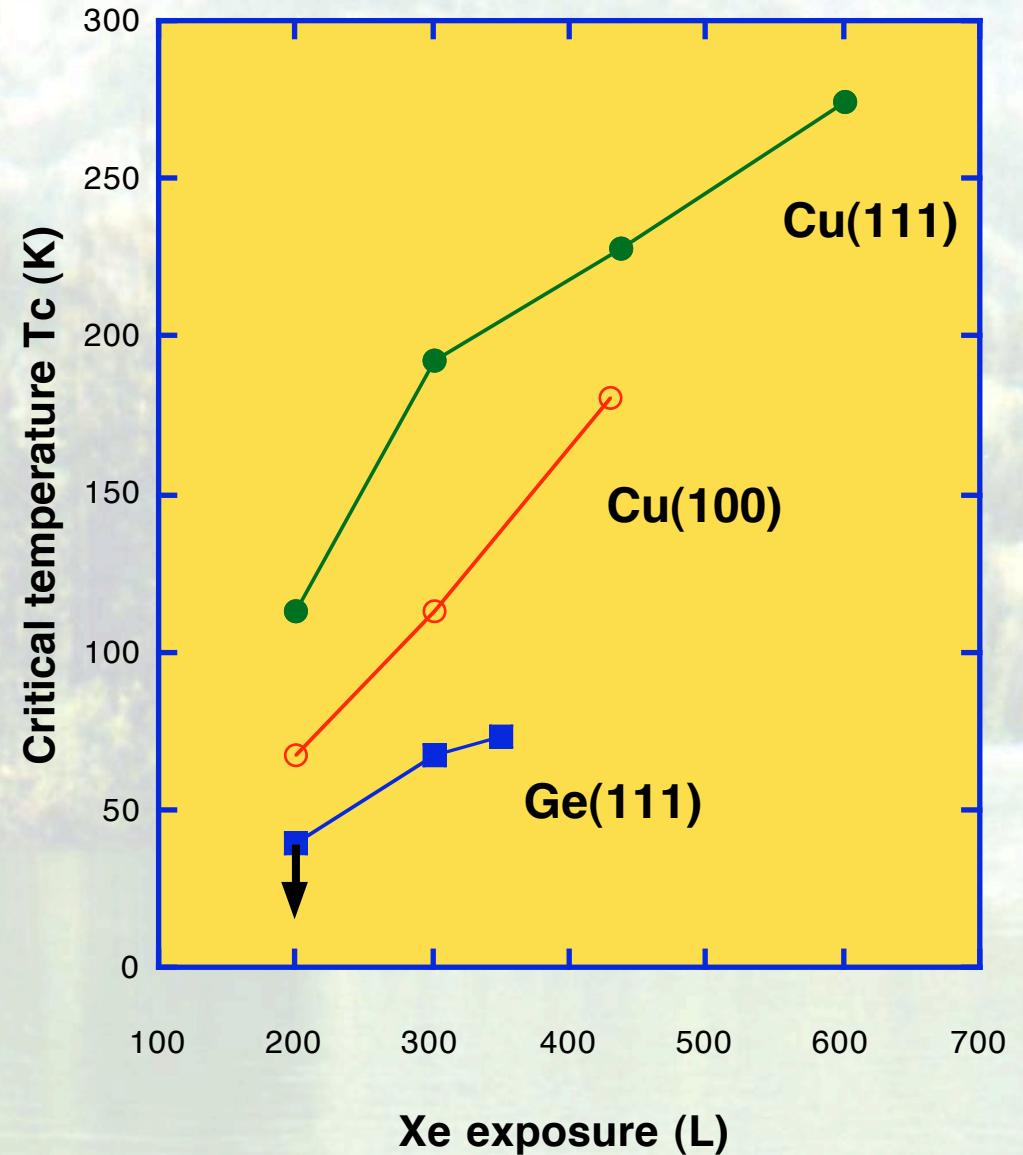
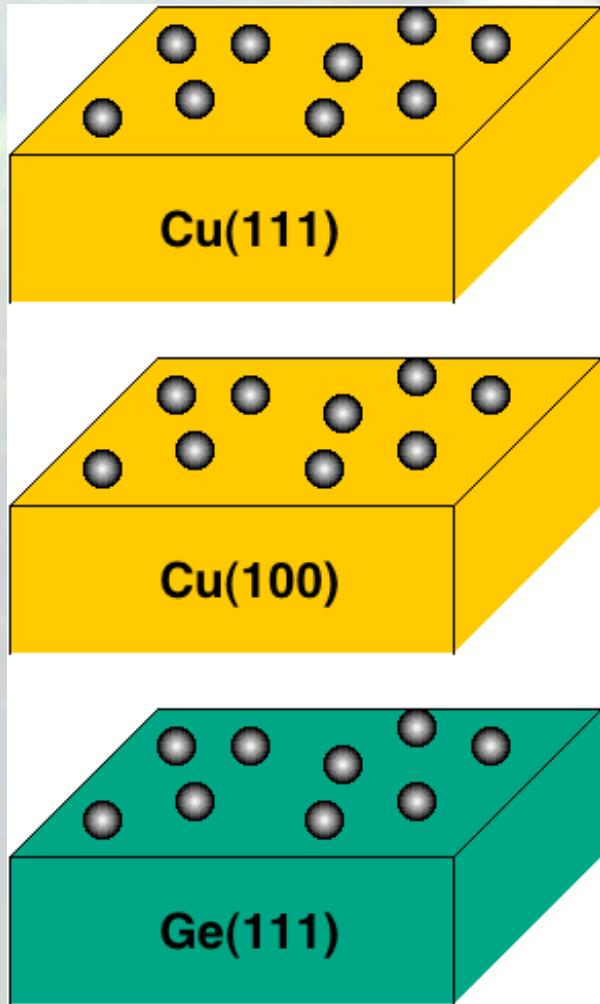


dot-dot *interactions* help stabilize magnetic order in the system

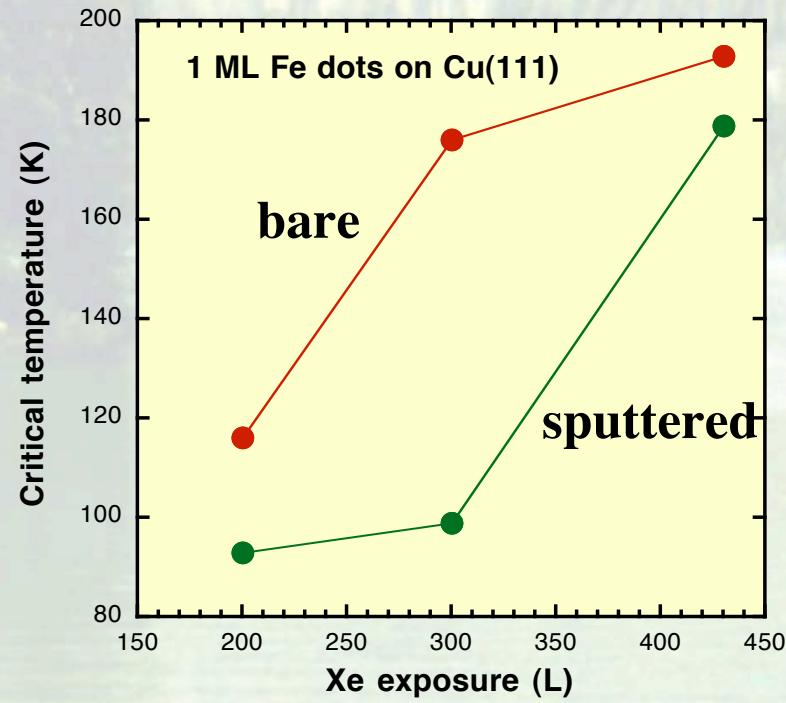
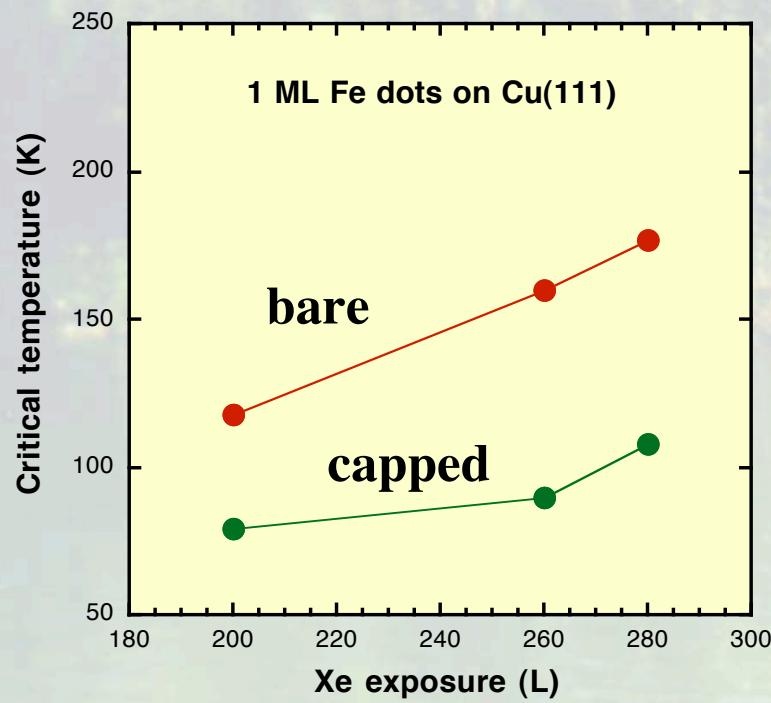
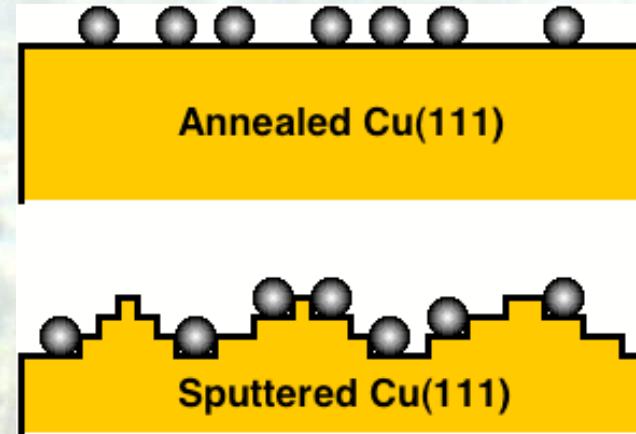
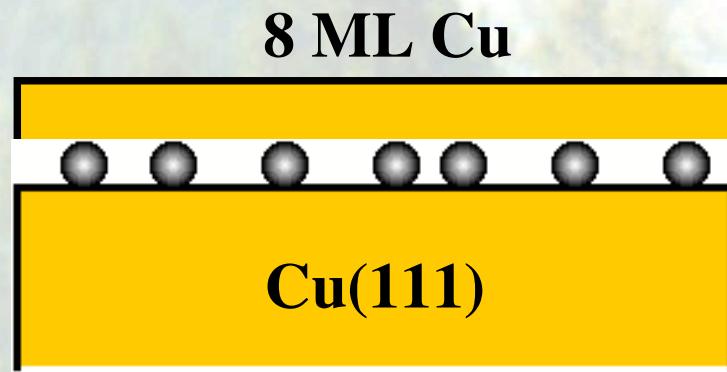
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J.Pierce et al., PRL 92, 237201 (2004)

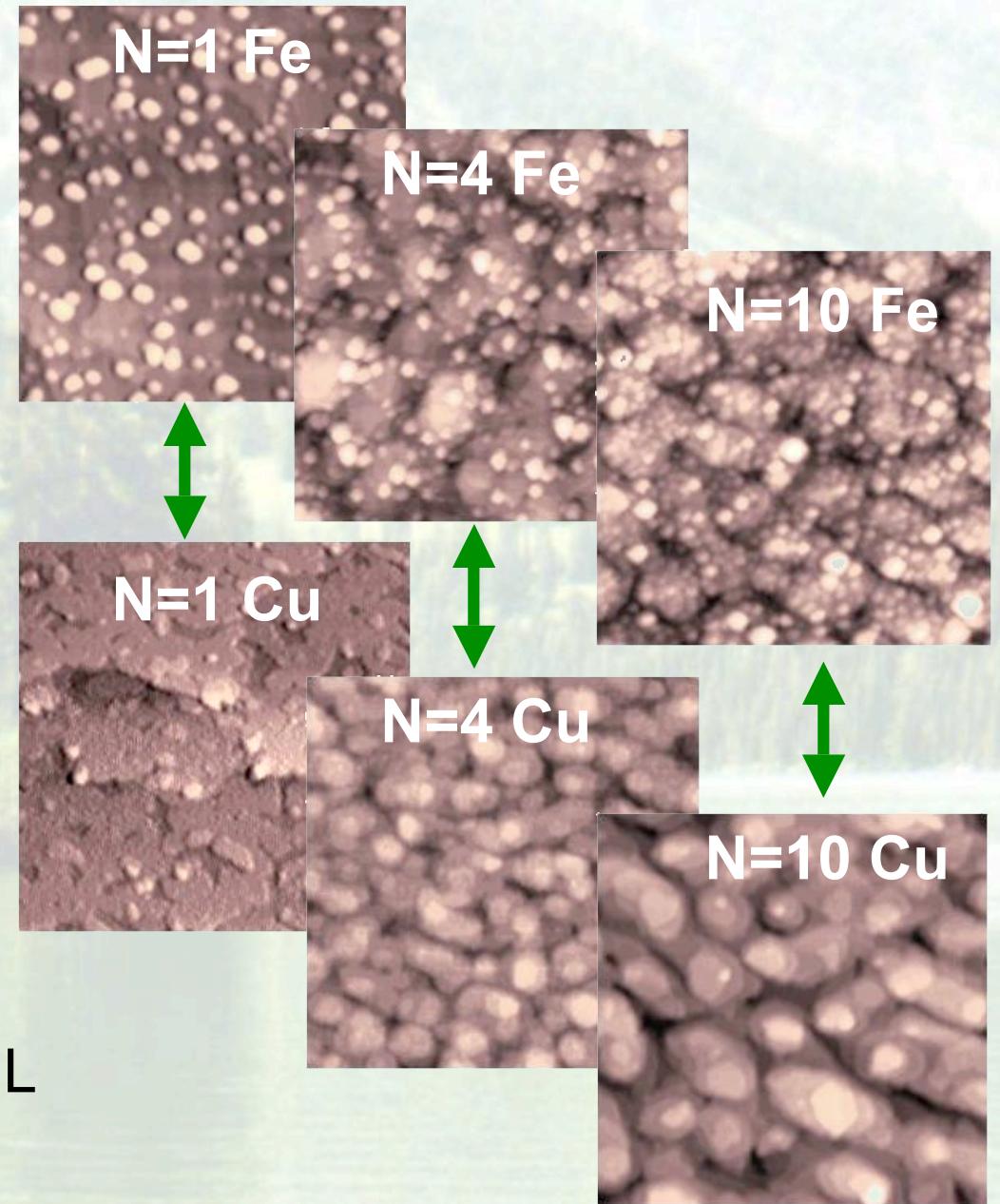
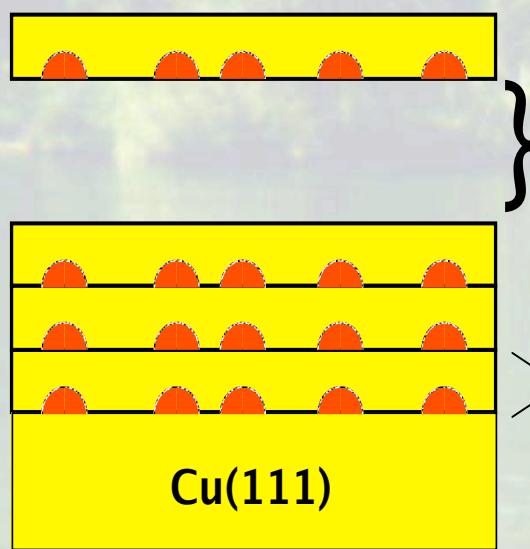
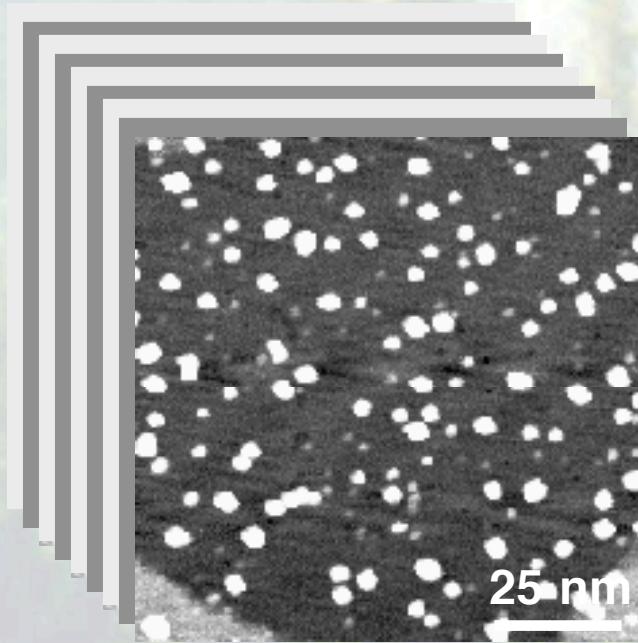
# Density of States is Critical



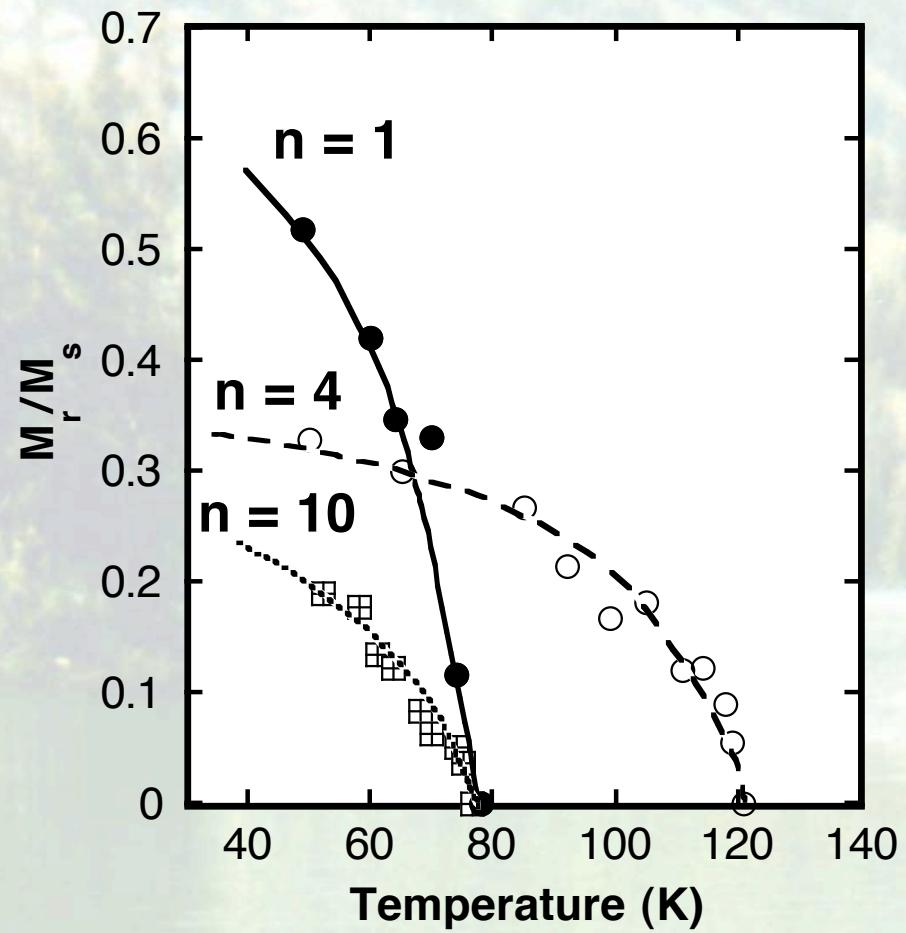
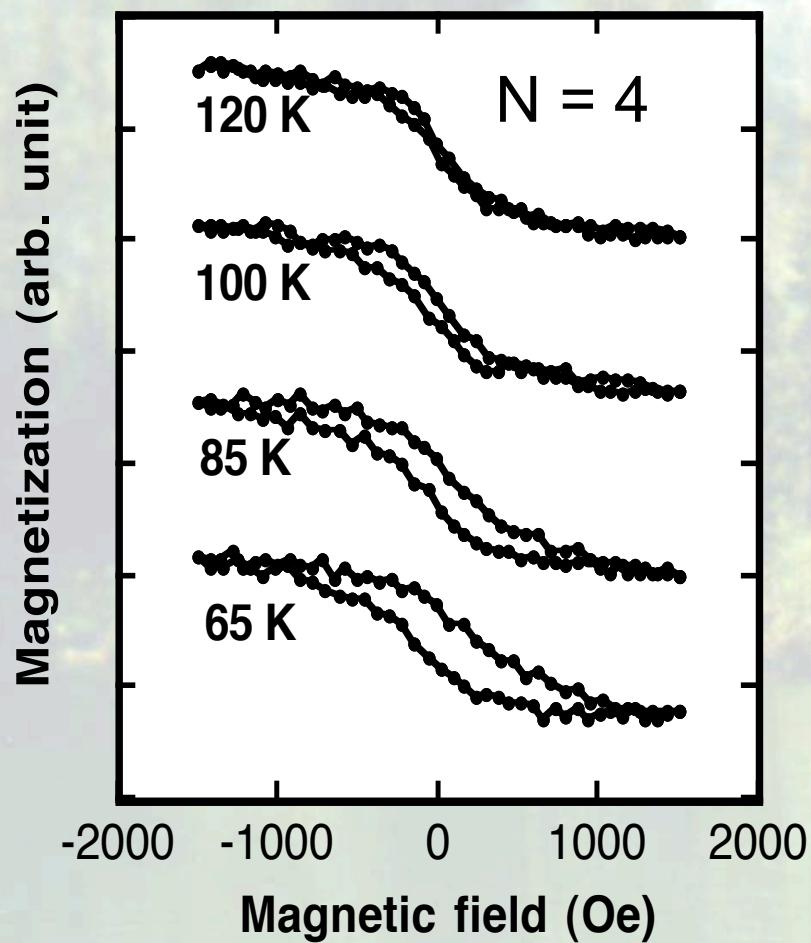
# Role of Surface States



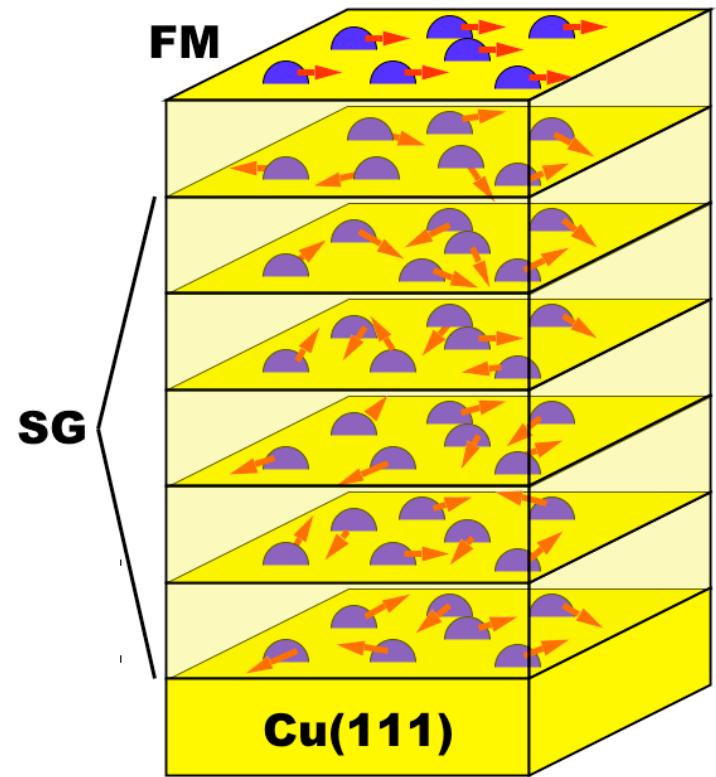
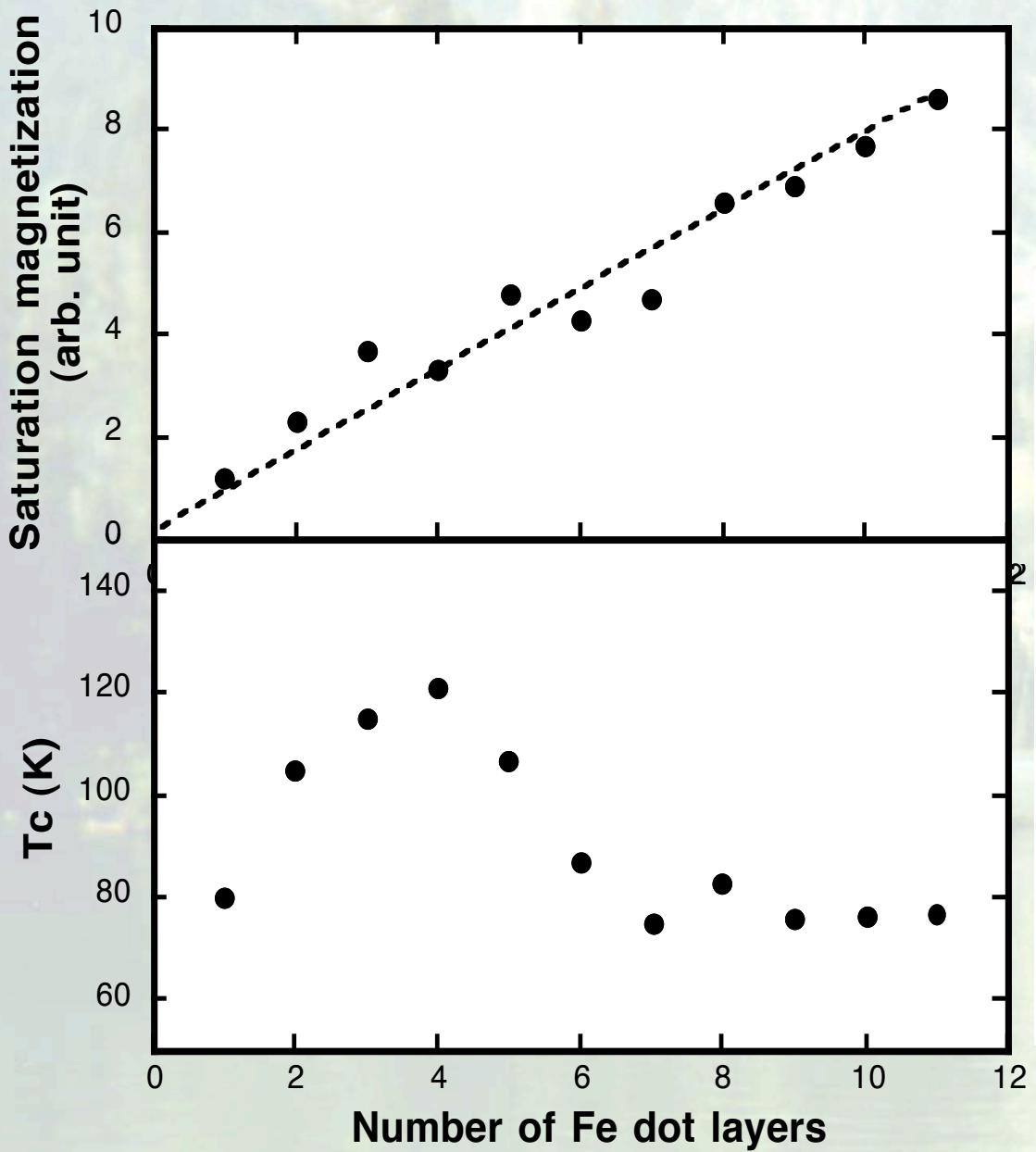
# Growth of Multilayer Fe Dots



# Magnetism of Multilayer Fe Dots



# Ferromagnetism vs. Spin Glass



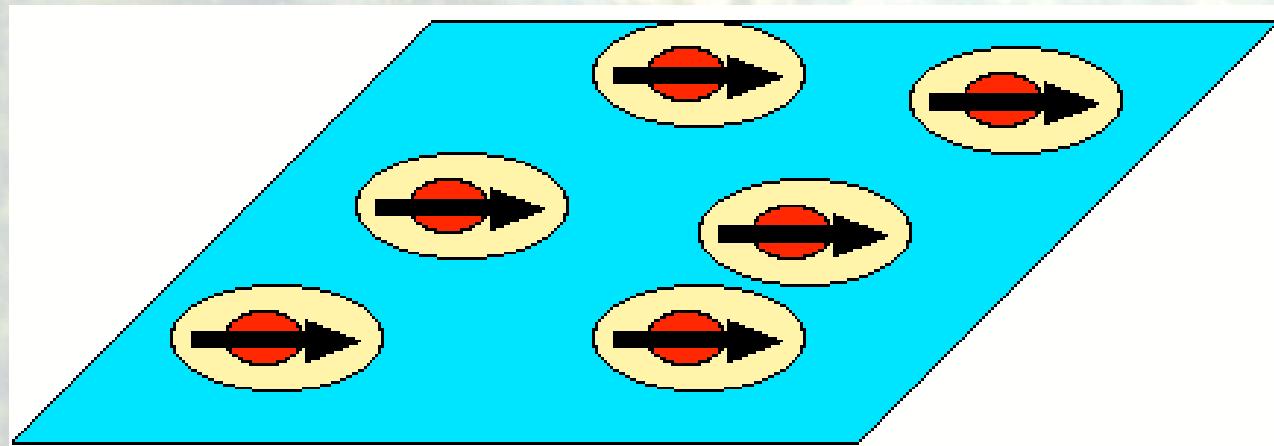
PRL (in press)

# Conclusion

Fe/Cu(111) dots

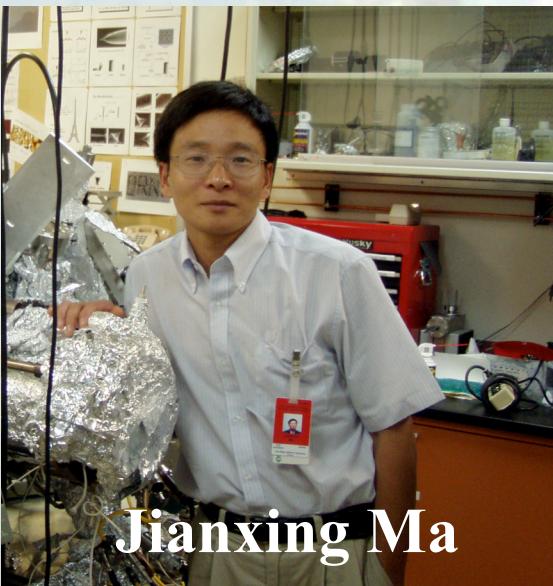
$Mn_xGe_{1-x}$  DMS

LPCM<sub>O</sub> Films



**High resolution magnetic imaging is critically needed!**

## Manganites



Jianxing Ma

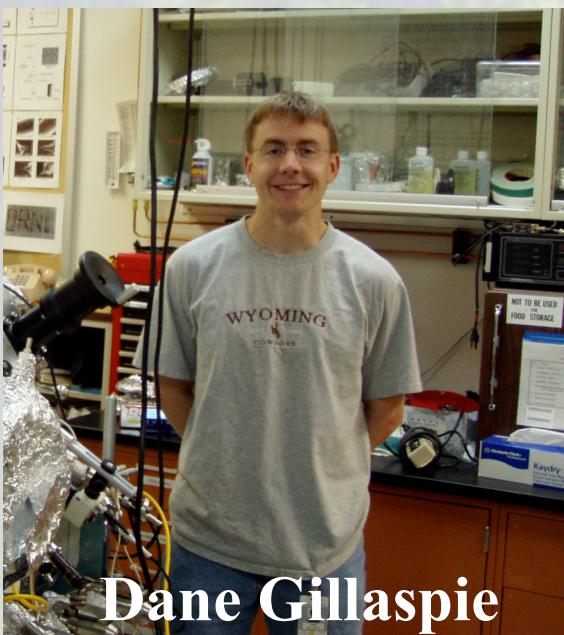
## Fe Nanodots



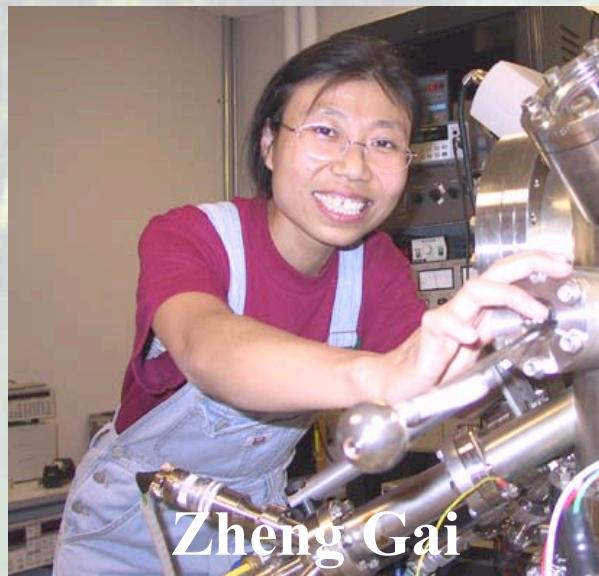
Maria  
Torija



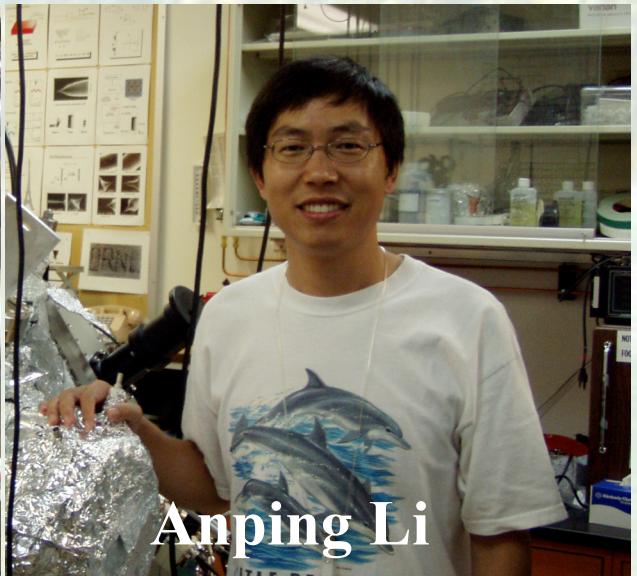
John Pierce



Dane Gillaspie

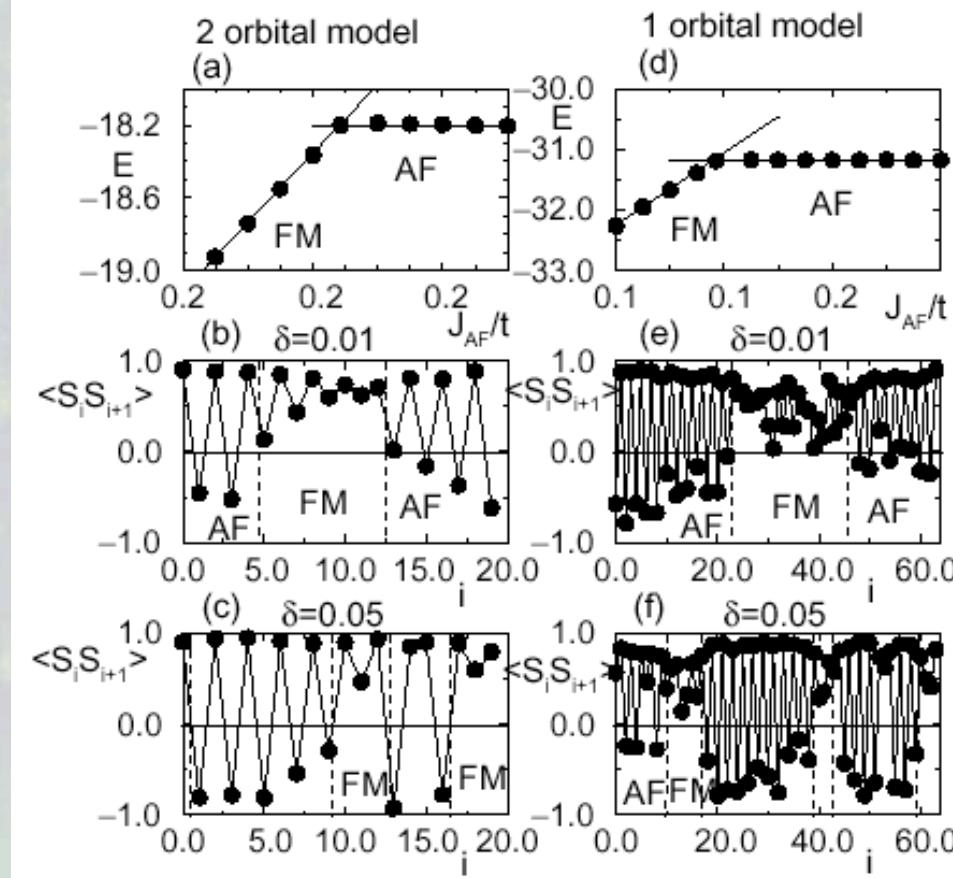


Zheng Gai



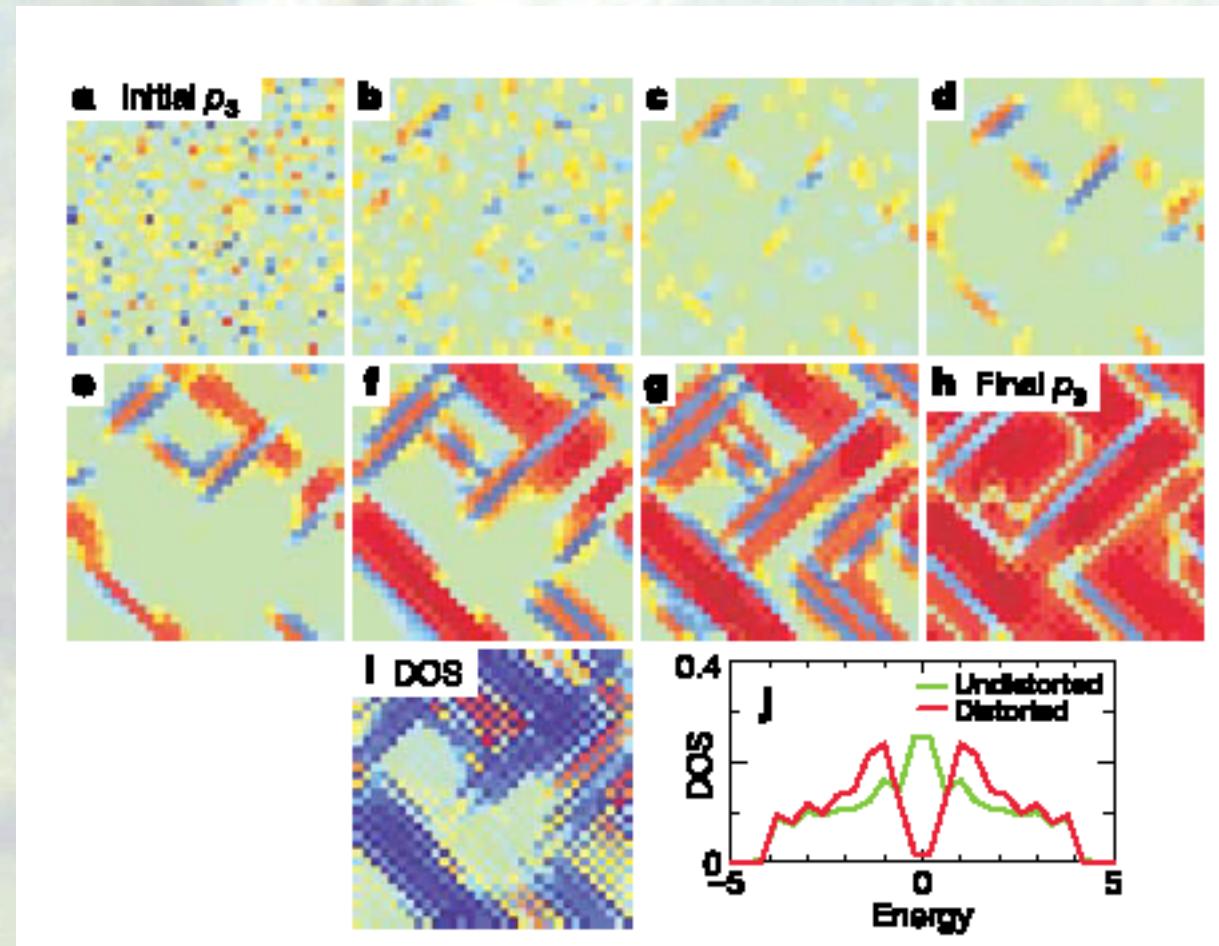
Anping Li

# Disorder-induced Phase Separation



Moreo et al., PRL 84, 5568 (2000)

# Polaron and Elastic Strain



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