



# Intrinsic Inhomogeneities in Doped Manganites and Related Oxides

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*NIST Center for Neutron Research*

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# Outline

- CMR systems
  - Natural GMR System:  $(La-Sr)CoO_3$
  - Avalanche Behavior at low T in a Phase-Separated Material
  - Ferromagnetic Droplet Formation
  - Polaron Formation and Nature of the Ferromagnetic Transition
  - Polaron Glass Phase
  - Effect of Disorder on  $T_C$  in  $(La-Ba)MnO_3$
  - Polaron Dynamics
  - “Polaron Glass” phase in YBCO

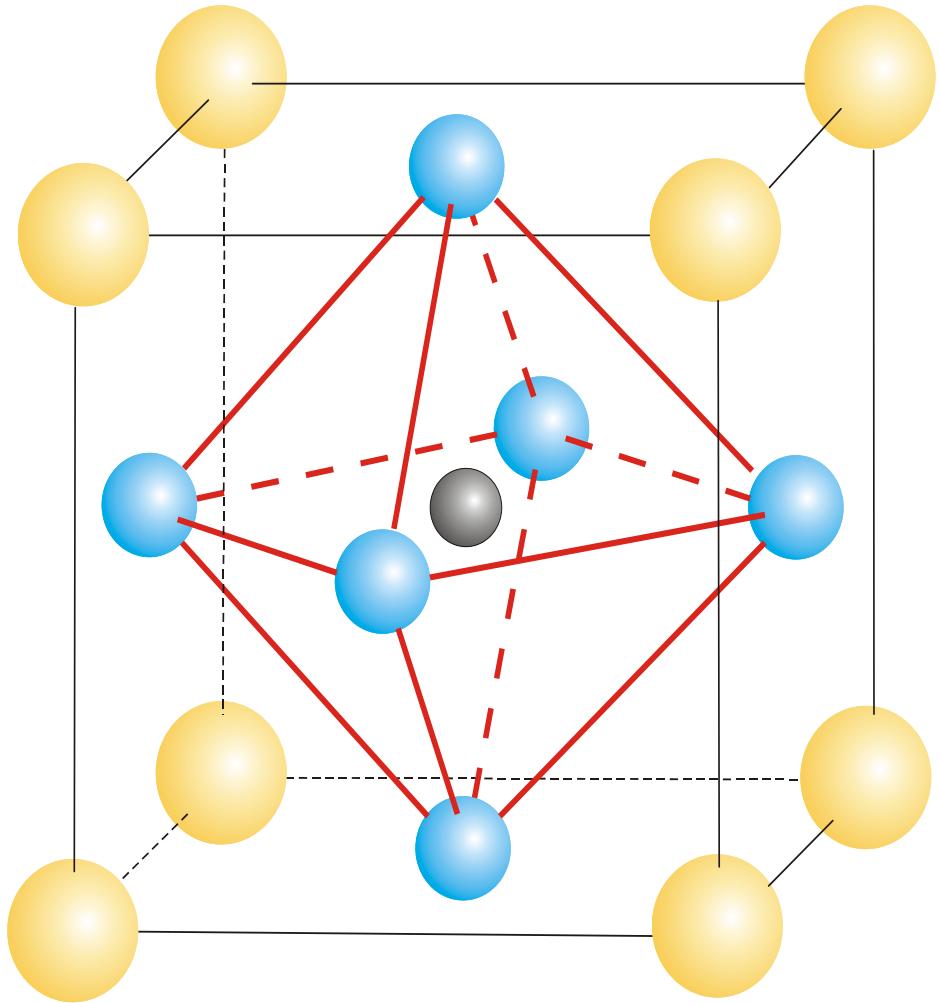
# (La-Sr)CoO<sub>3</sub> Natural GMR System

Inter-granular Giant Magnetoresistance in a  
Spontaneously Phase Separated Perovskite Oxide, J.  
Wu, J. W. Lynn, C. J. Glinka, J. Burley, H. Zheng,  
J. F. Mitchell, and C. Leighton,  
Phys. Rev. Lett. **94**, 037201 (2005)

# Systems under Investigation

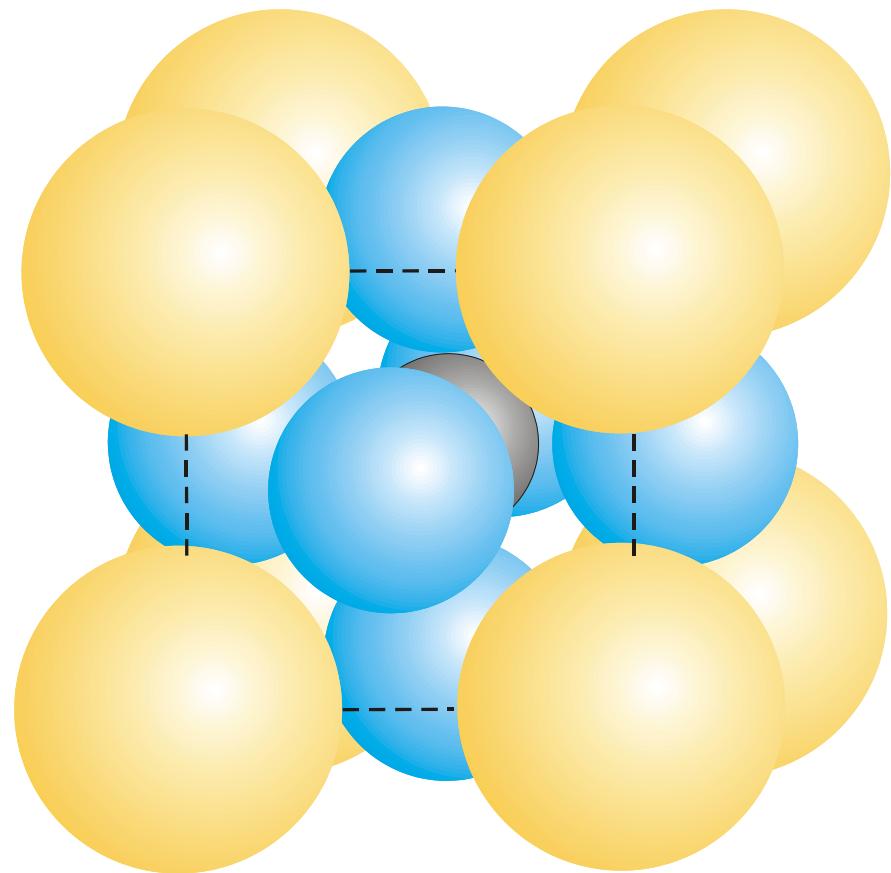
- CMR systems, Multiferroics ( $\text{La-CaMnO}_3$ ,  $\text{HoMnO}_3$ )
- Magnetic Superconductors (e.g.  $\text{RNi}_2\text{B}_2\text{C}$ ,  $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ )
- Heavy Fermion Materials ( $\text{CeRhIn}_5$ ,  $\text{PrOs}_4\text{Sb}_{12}$ , ...)
- Non-Fermi Liquids ( $\text{UCu}_{5-x}\text{Pd}_x$ ,  $\text{Sc}_{1-x}\text{U}_x\text{Pd}_3$ )
- Magnetic Order and Fluctuations in Cuprates (NCCO)
- Cobaltates ( $\text{Na}_x\text{CoO}_2$  (*just add water for Superconductivity*))
- Frustrated Magnets

# Ideal Perovskite Structure (Cubic)



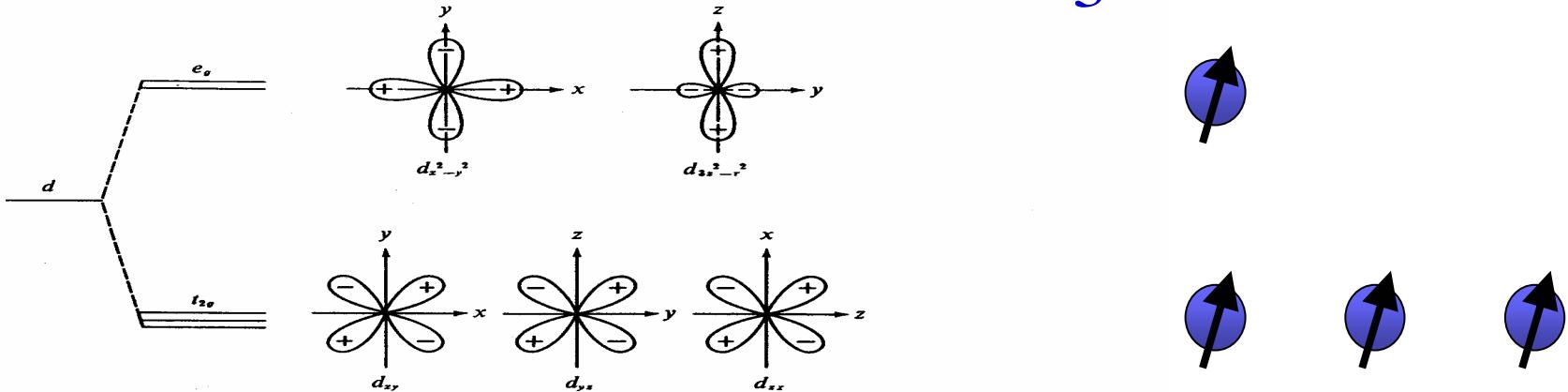
**LaMnO<sub>3</sub>**

# Ideal Perovskite Structure (Cubic)



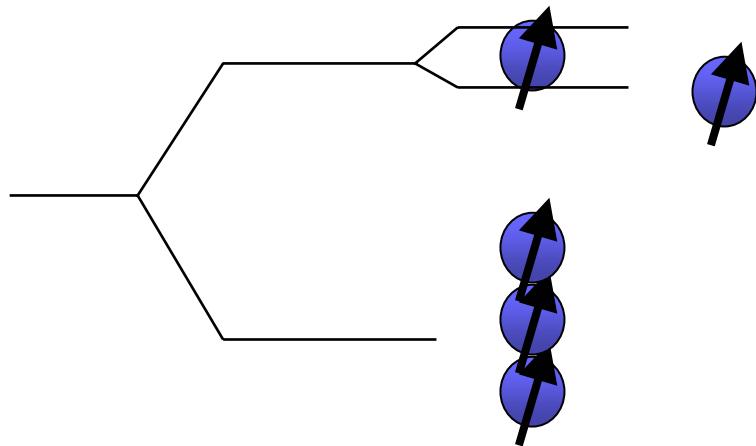
# *Basic Interactions*

## Mn<sup>3+</sup> in LaMnO<sub>3</sub>



Strong Hund's Rule Coupling  
(on-site electrons must be parallel)  
Jahn-Teller Distortion

# Basic Interactions

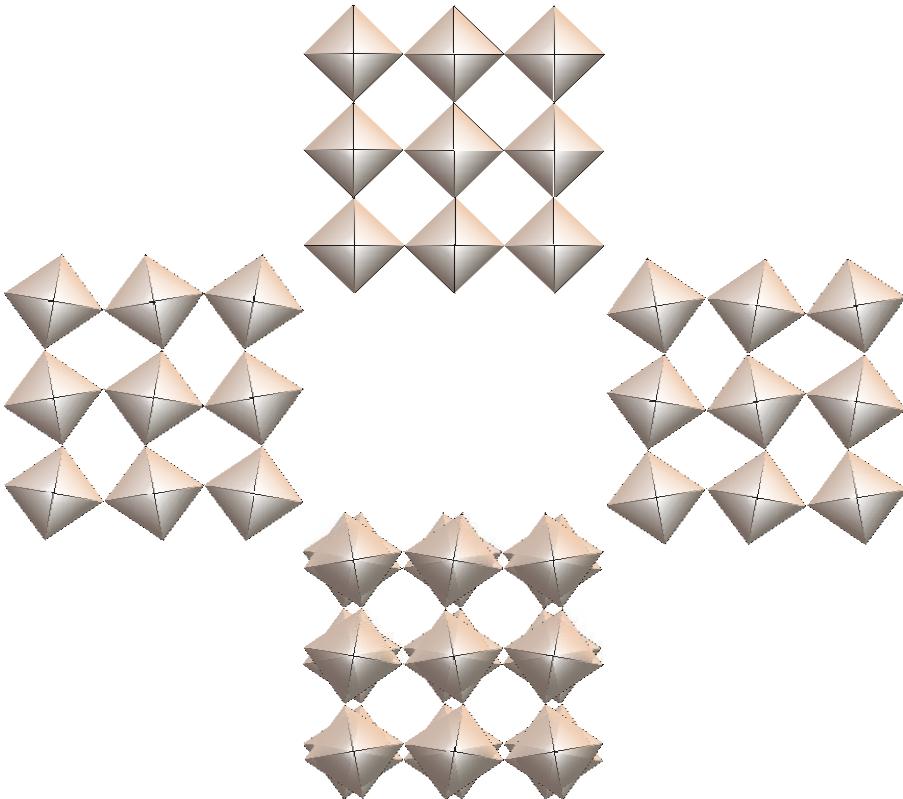


**Strong Hund's Rule Coupling**

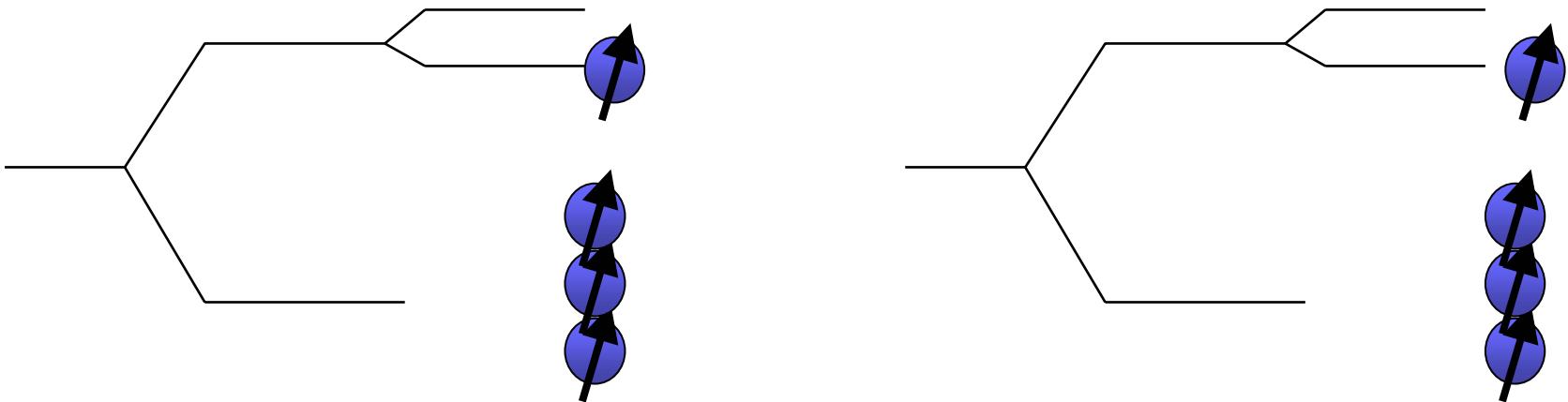
(on-site electrons must be parallel)

**Jahn-Teller Distortion** couples spins with lattice

# Tiling (Orthorhombic)

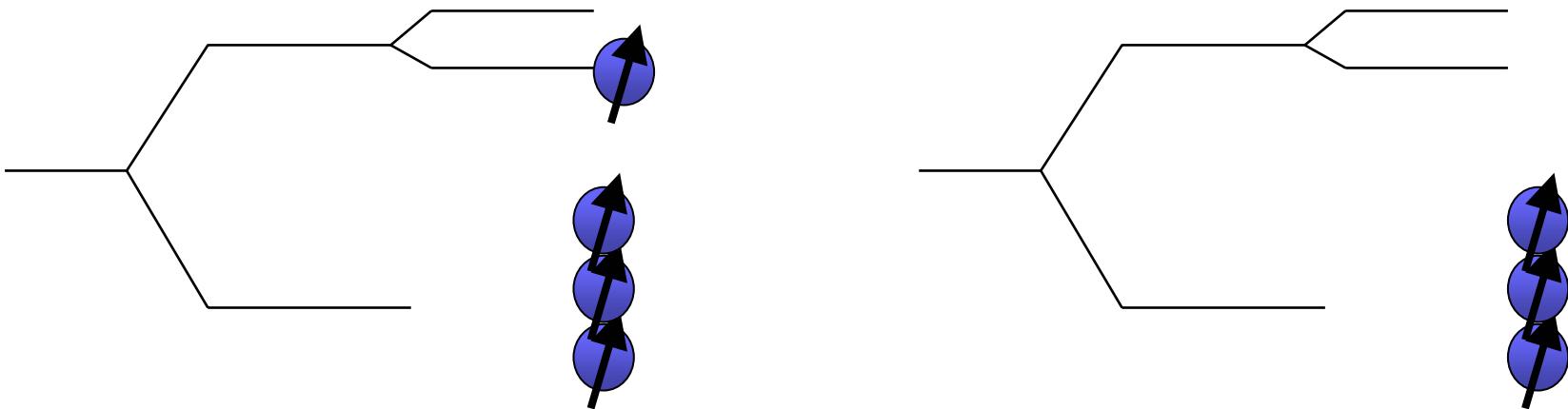


# Basic Interactions



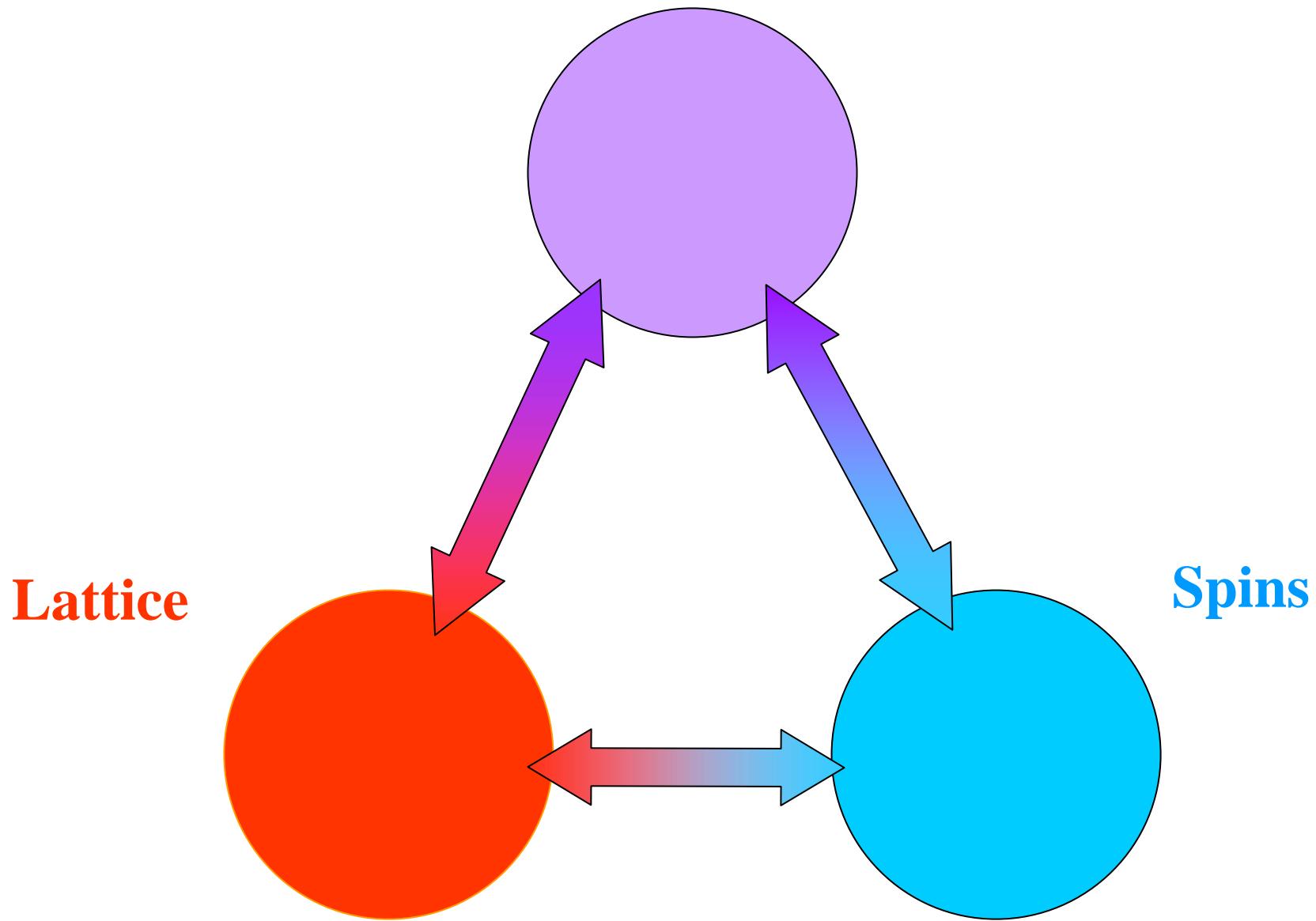
$e_g$  electron can only hop if core spins  
are parallel (& an empty site)  
Jahn-Teller Distortion couples spins  
with lattice

# Basic Interactions

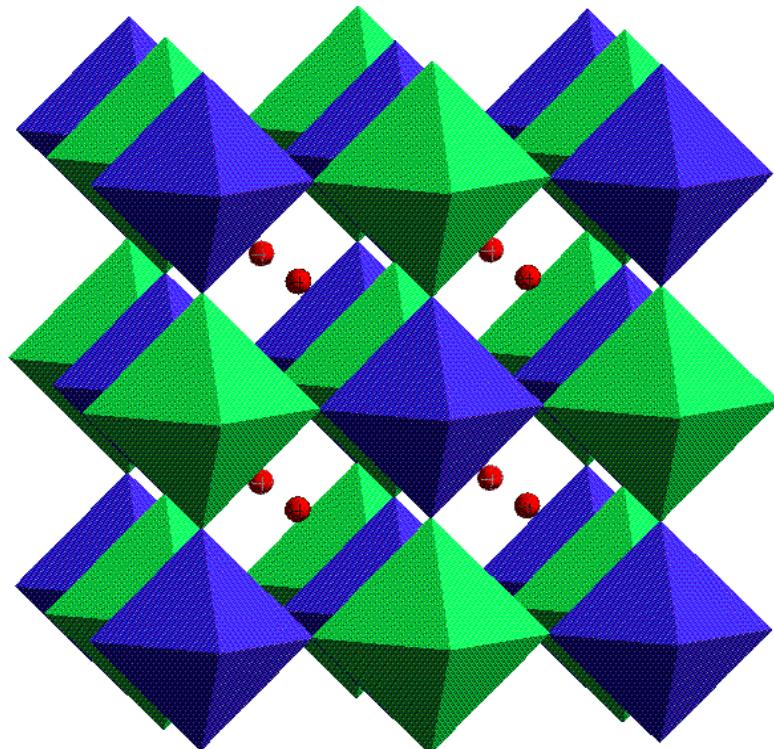


$e_g$  electron can only hop if core spins  
are parallel (& an empty site)  
Jahn-Teller Distortion couples spins  
with lattice

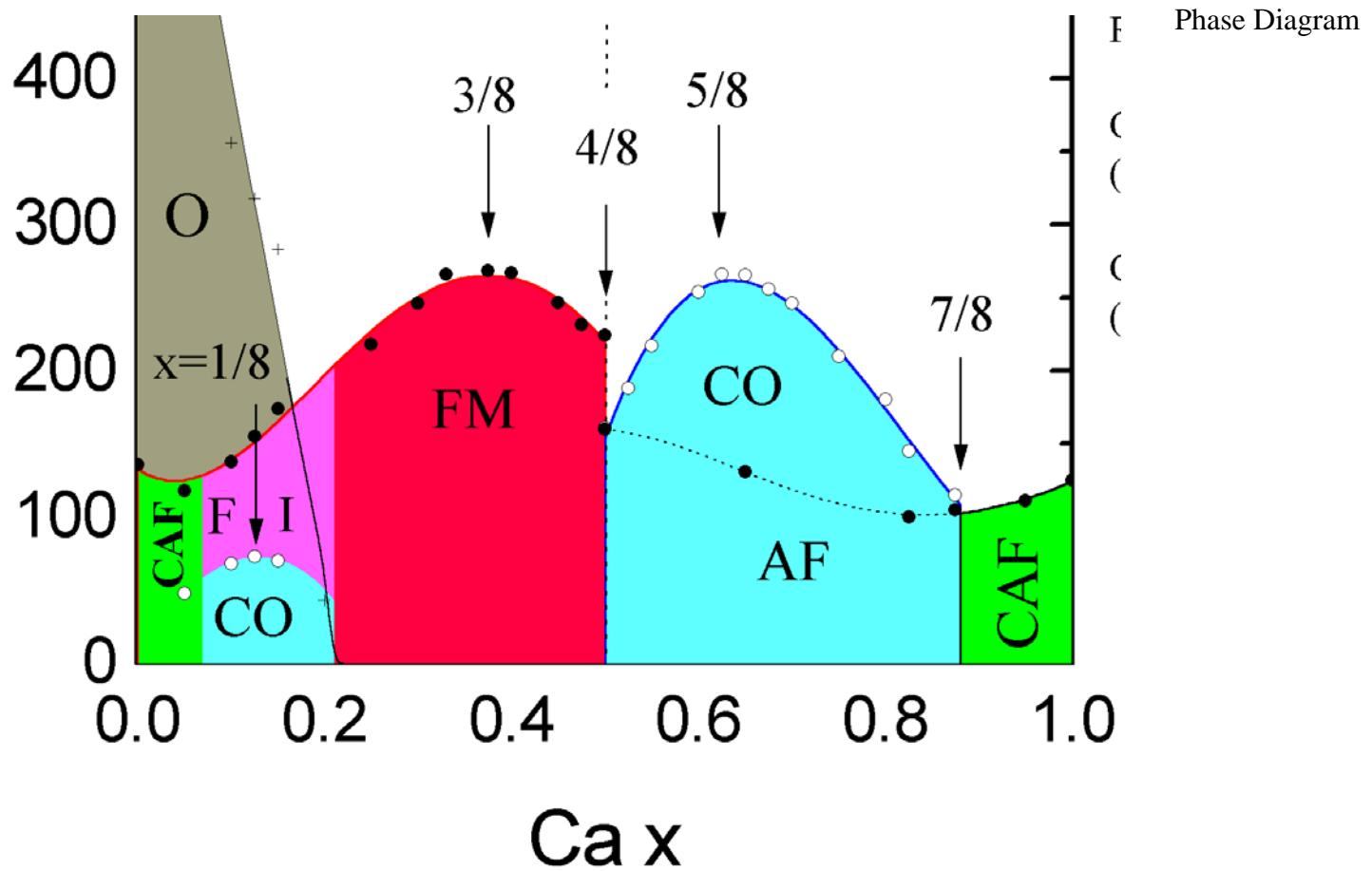
# Conductivity



# Doping of Structure



# $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$

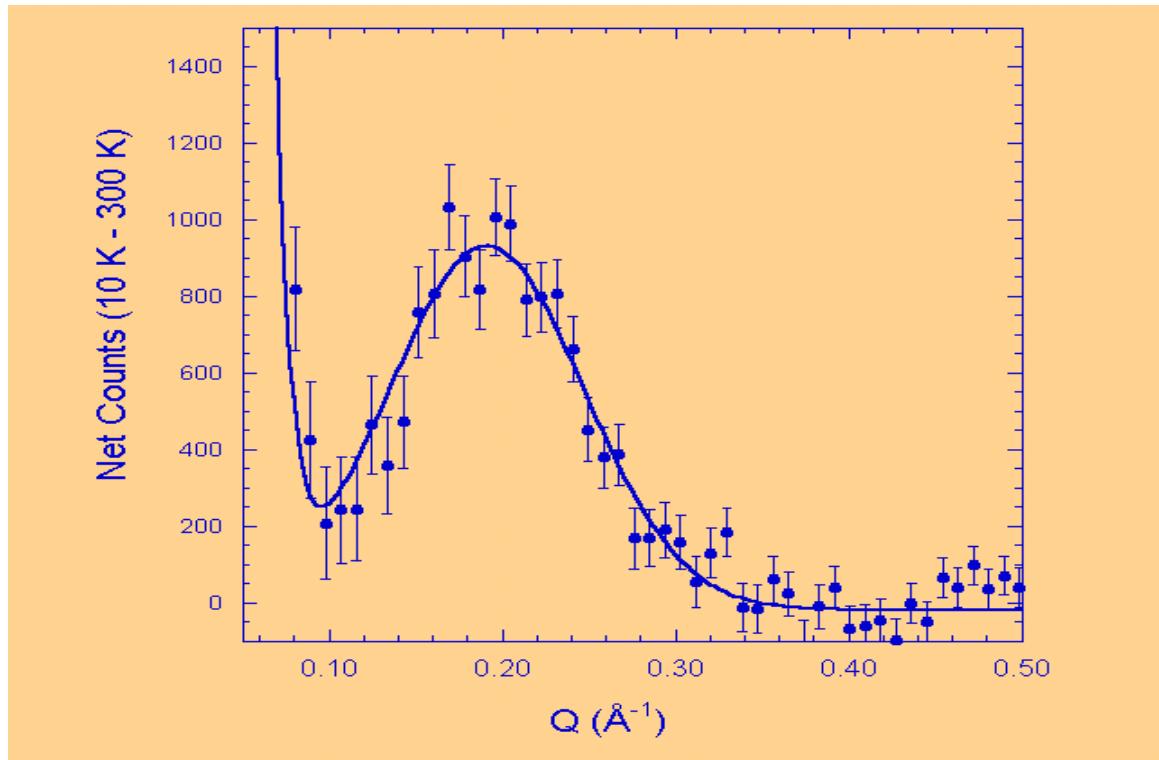


S-W. Cheong and C. H. Chen

*Colossal Magnetoresistance, Charge Ordering, and Related Properties of Manganese Oxides* (World Scientific, 1998),  
p. 241 (Ed. by Raveau and Rao)

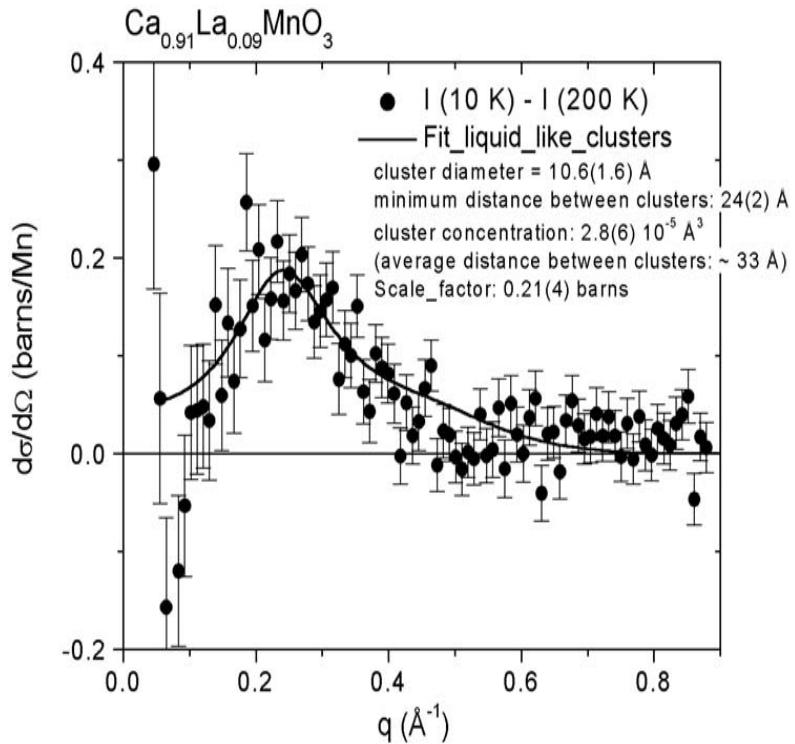
# Ferromagnetic Droplets

$\text{La}_{0.95}\text{Ca}_{0.05}\text{MnO}_3$



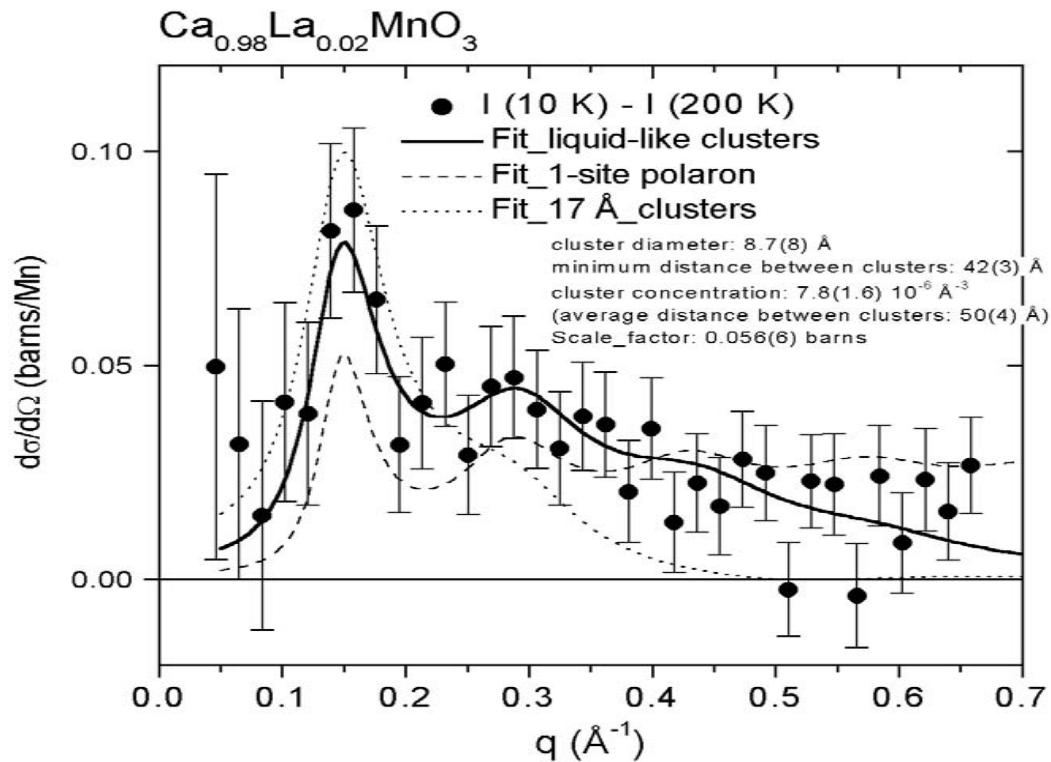
M. Hennion, *et al.* Phys. Rev. Lett. **81**, 1957 (1998)

# Ferromagnetic Clusters (Electron-doped)



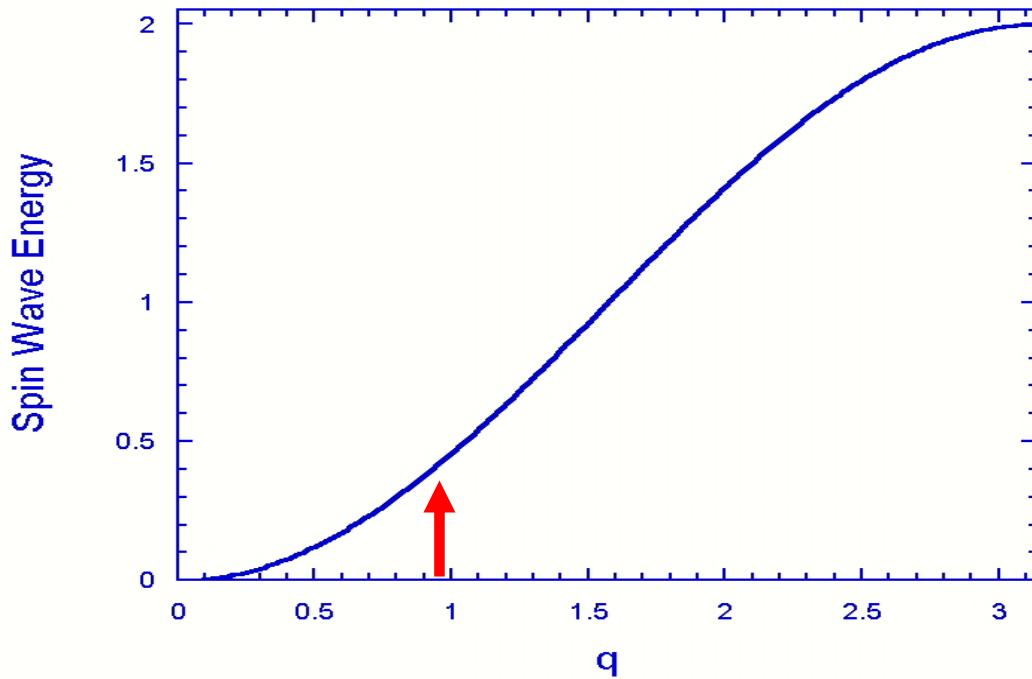
E. Granado, C. D. Ling, J. J. Neumeier, J. W. Lynn, and D. N. Argyriou, Phys. Rev. B**68**, 134440 (2003)

# Ferromagnetic Clusters



# Excitations

$$E_{sw} = 2JS(1 - \cos(aq))$$

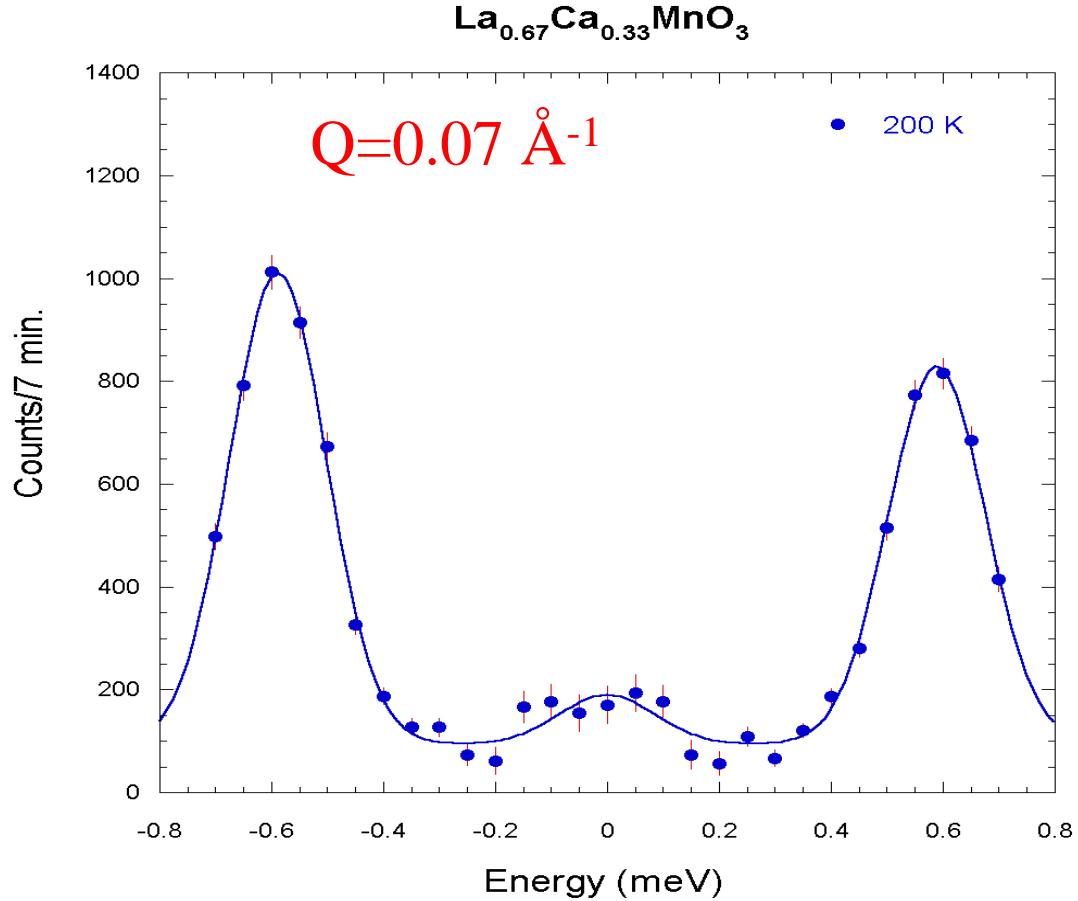


$$E_{sw} = 2JS(1 - \cos(aq)) \approx \Delta + D(T)q^2 \approx D(T)q^2$$

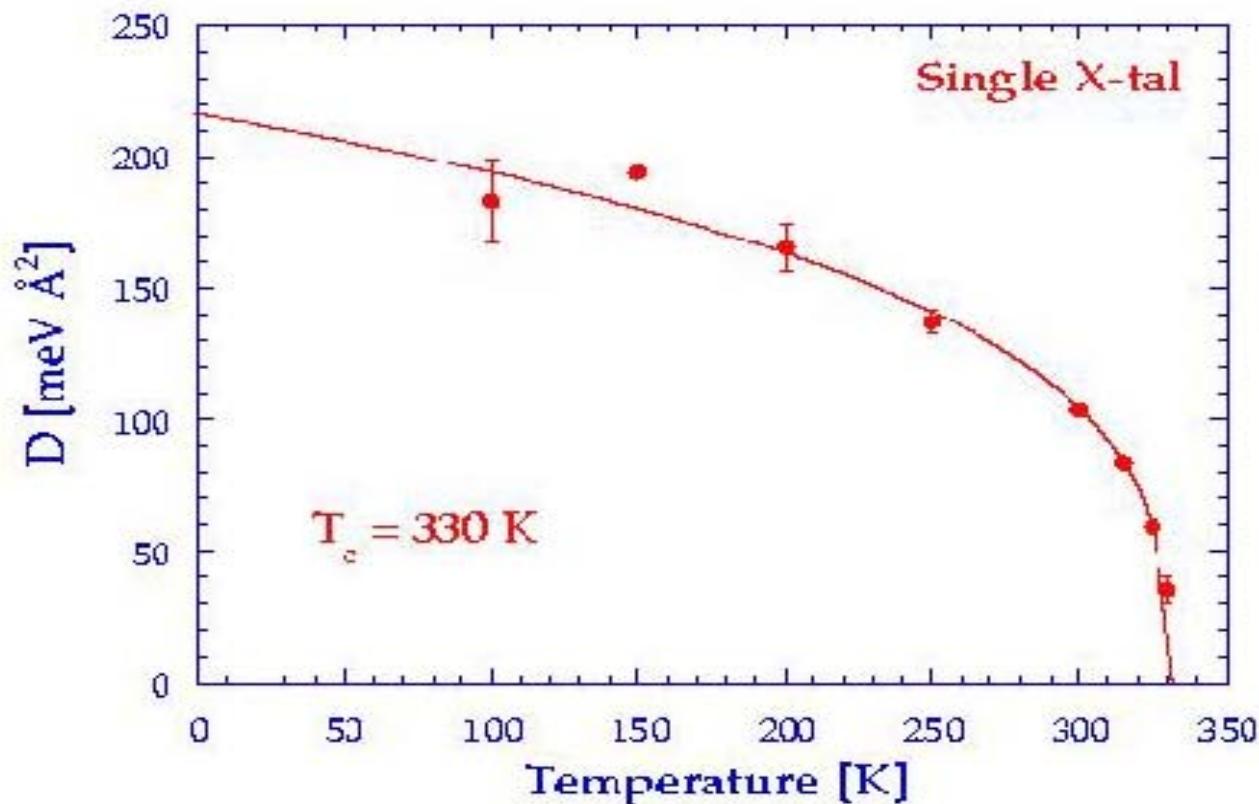
$$D(T) \sim M(T)$$

$$\Gamma \sim T^2 q^4$$

# Spin Dynamics

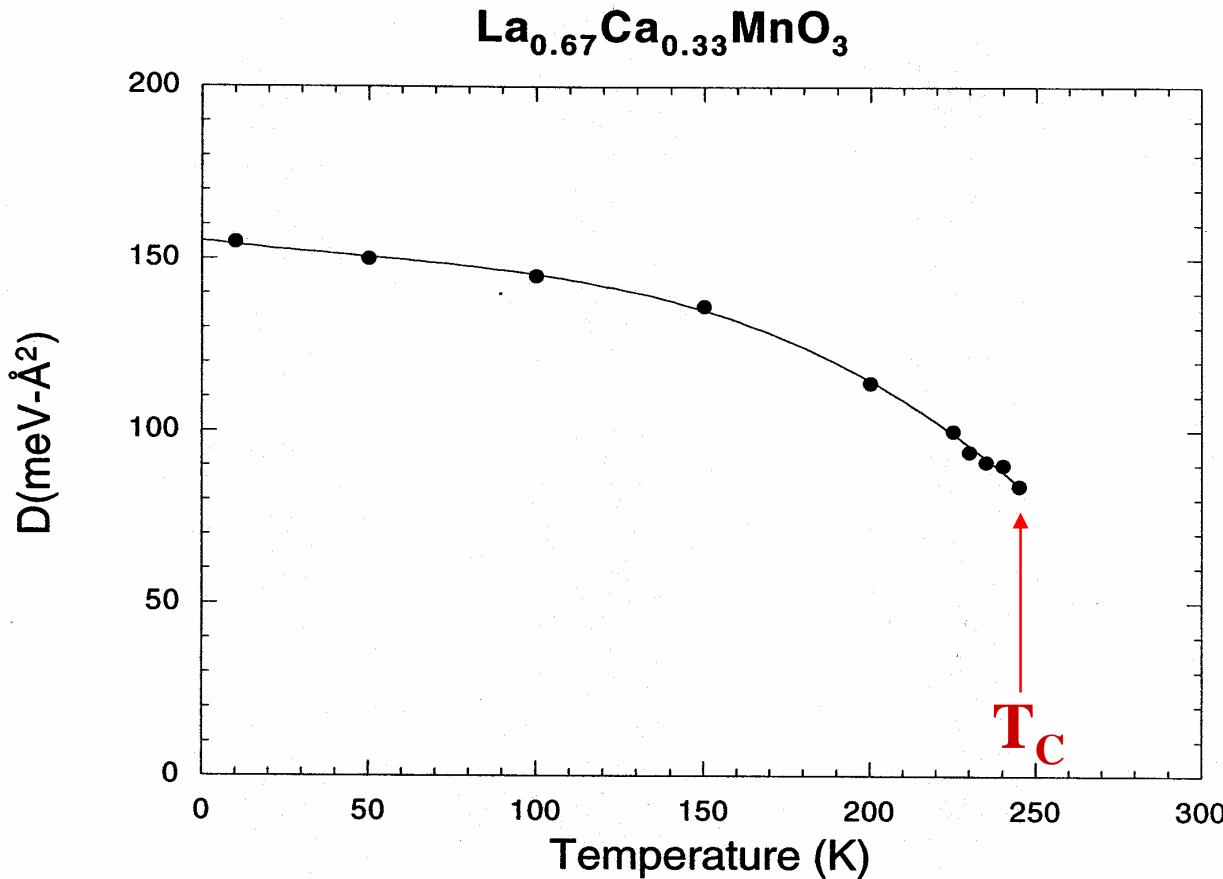
$$E_{\text{SW}} = D(T) Q^2$$


# $\text{La}_{0.8}\text{Ba}_{0.2}\text{MnO}_3$ $D(T)$



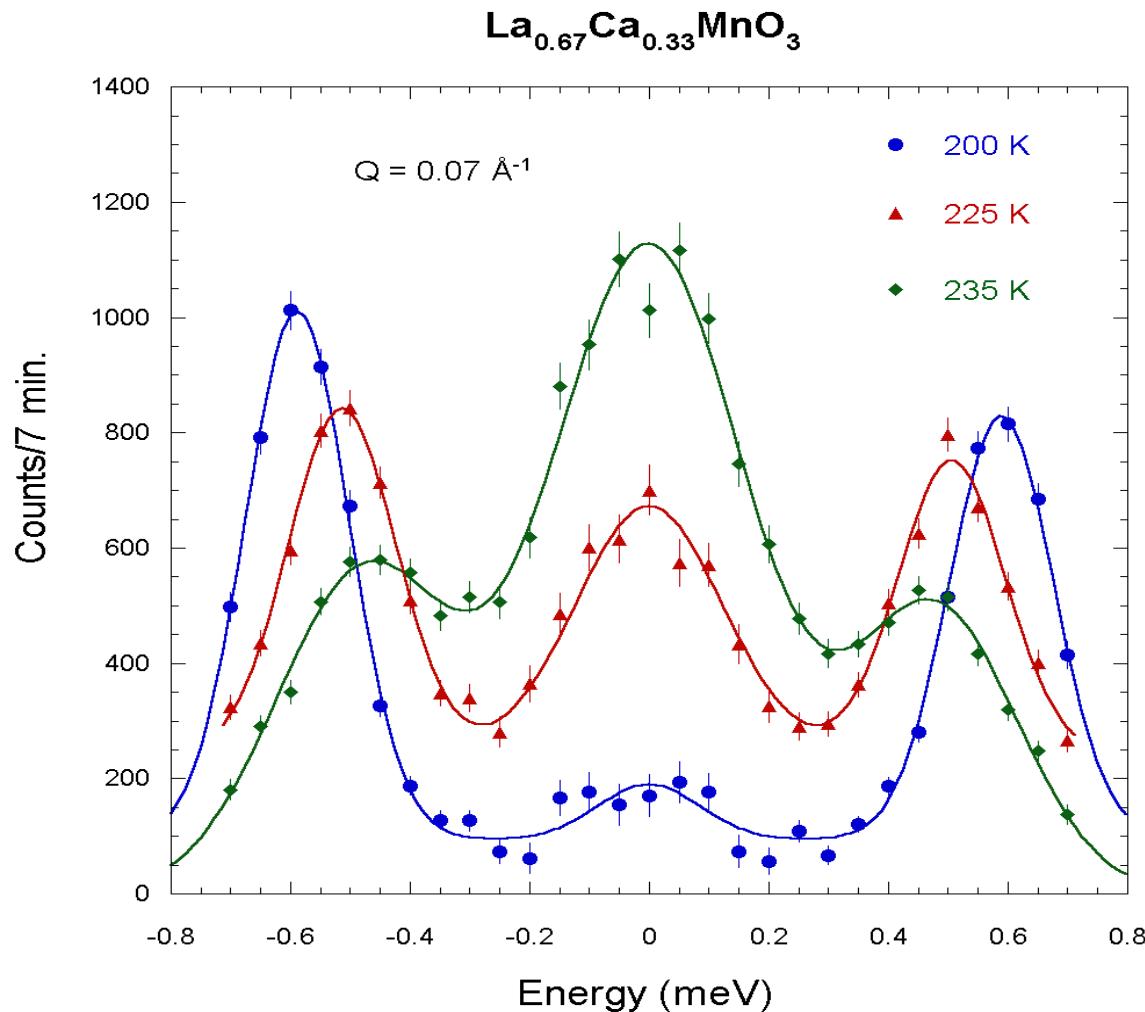
$$E_{\text{SW}} = D(T) Q^2$$

# $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$

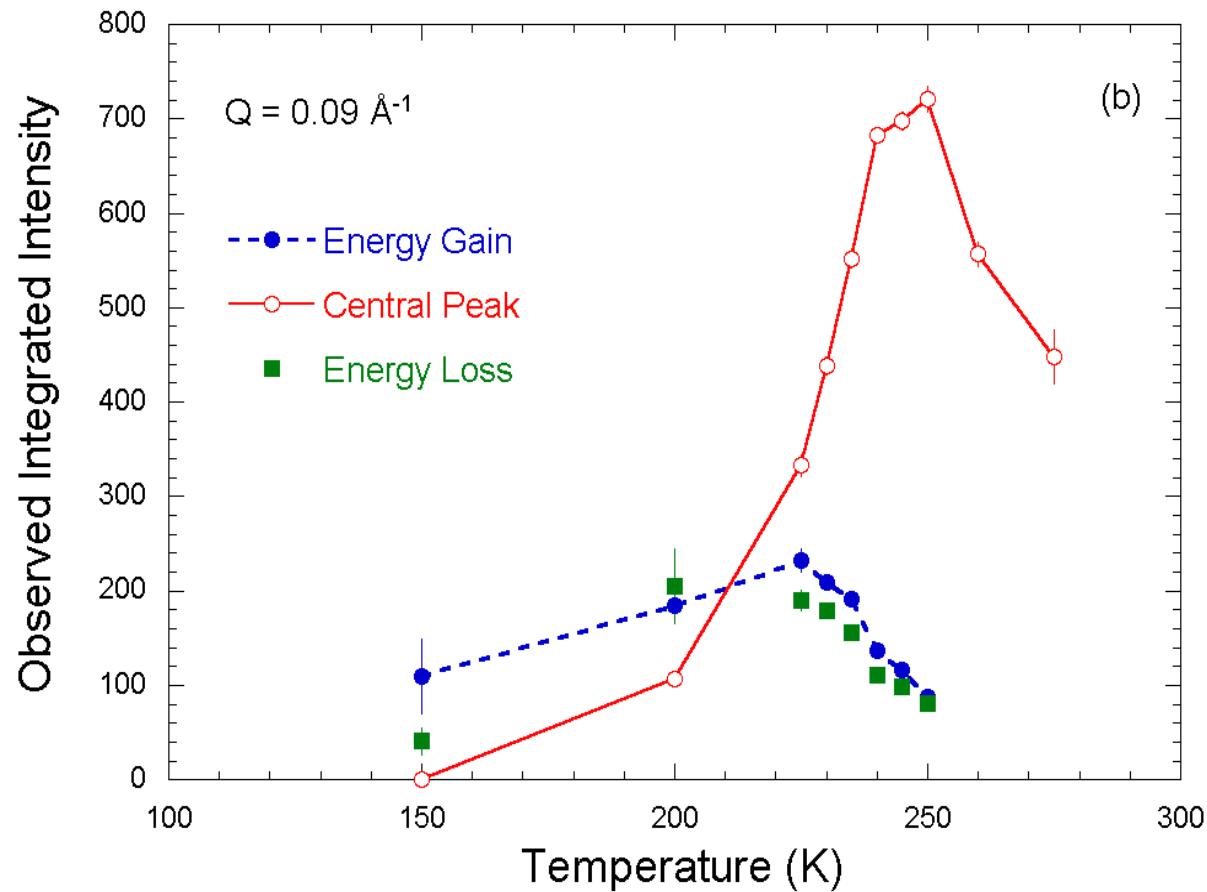


Phys. Rev. Lett. **76**, 4046 (1996)

# Spin waves

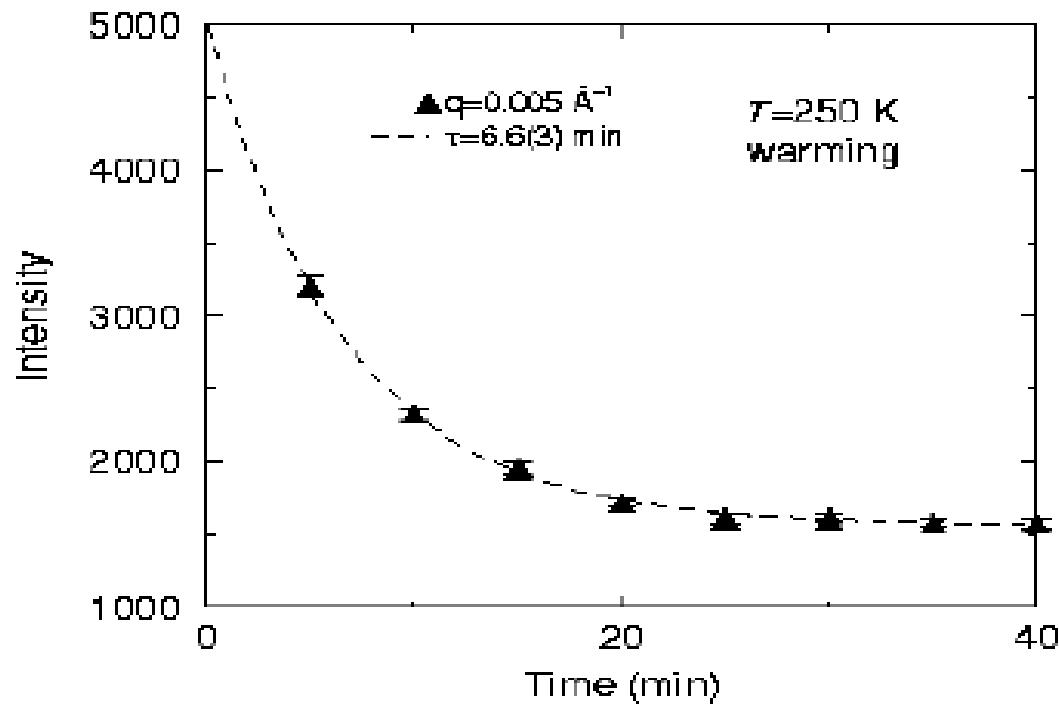


# CP Intensity

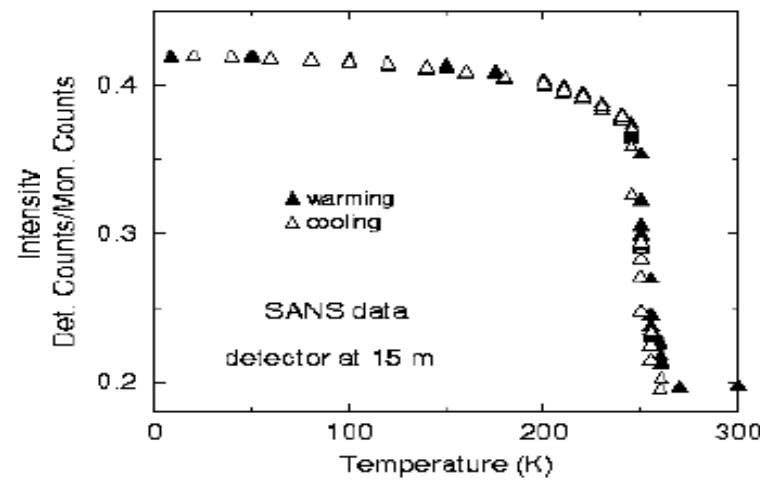
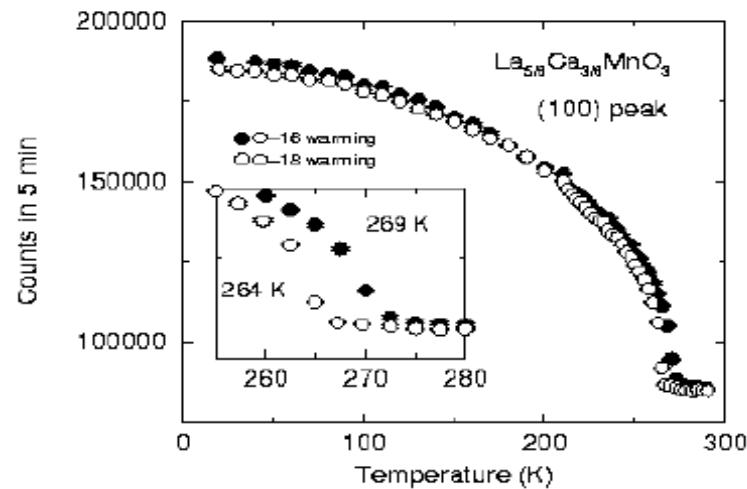


Phys. Rev. Lett. **76**, 4046 (1996).

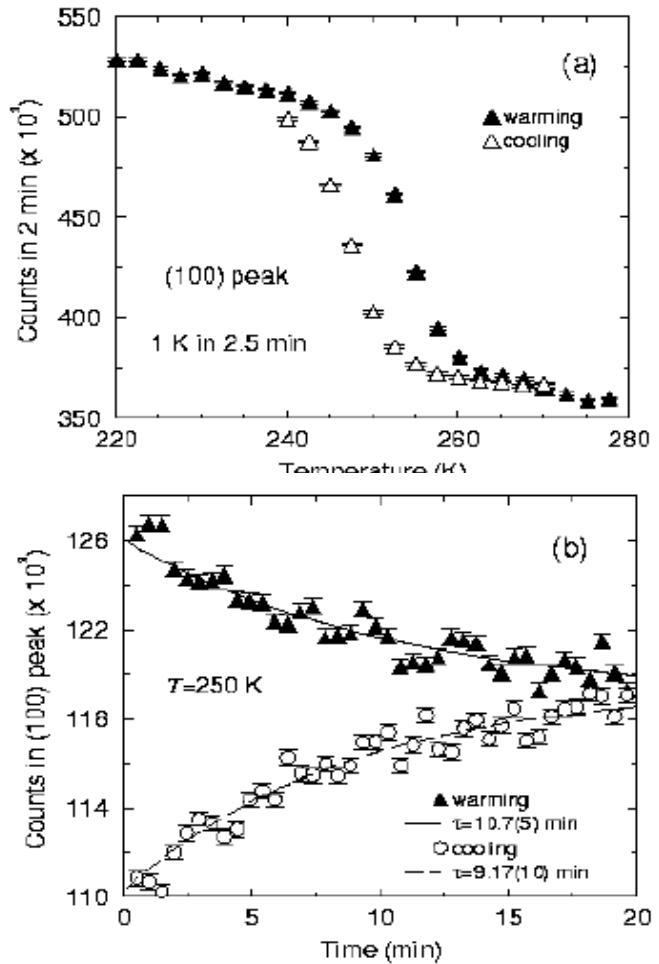
# Time Dependence



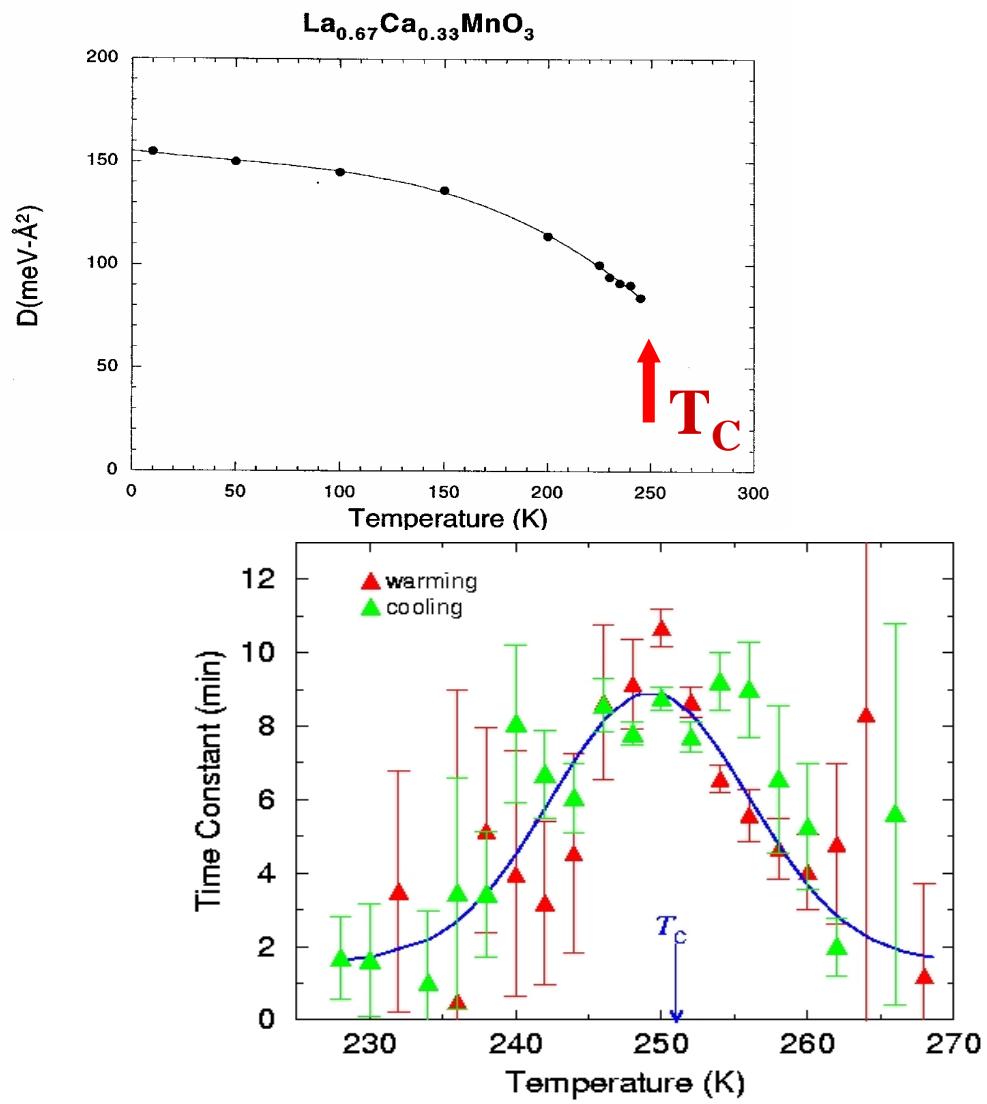
# *Time Dependence*



# *Time Dependence (Single Crystal)*

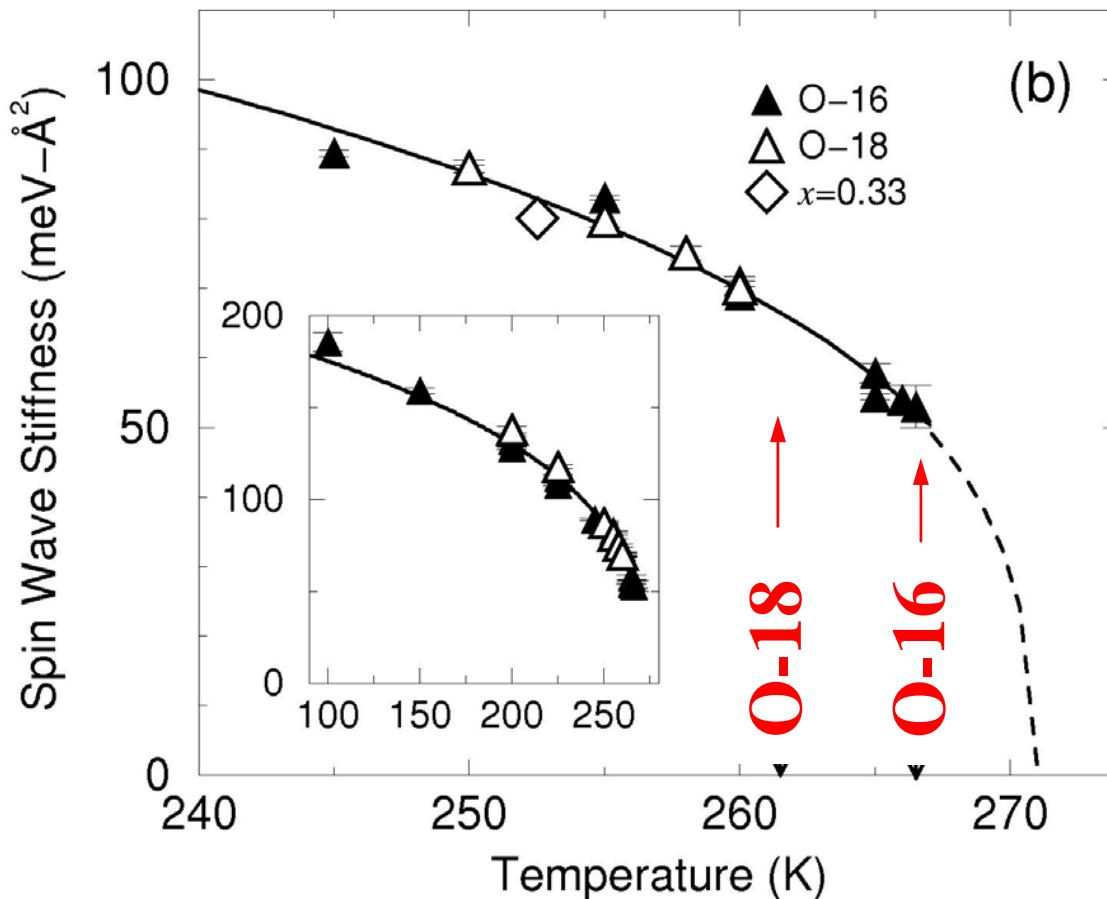


# *Discontinuous Transition*



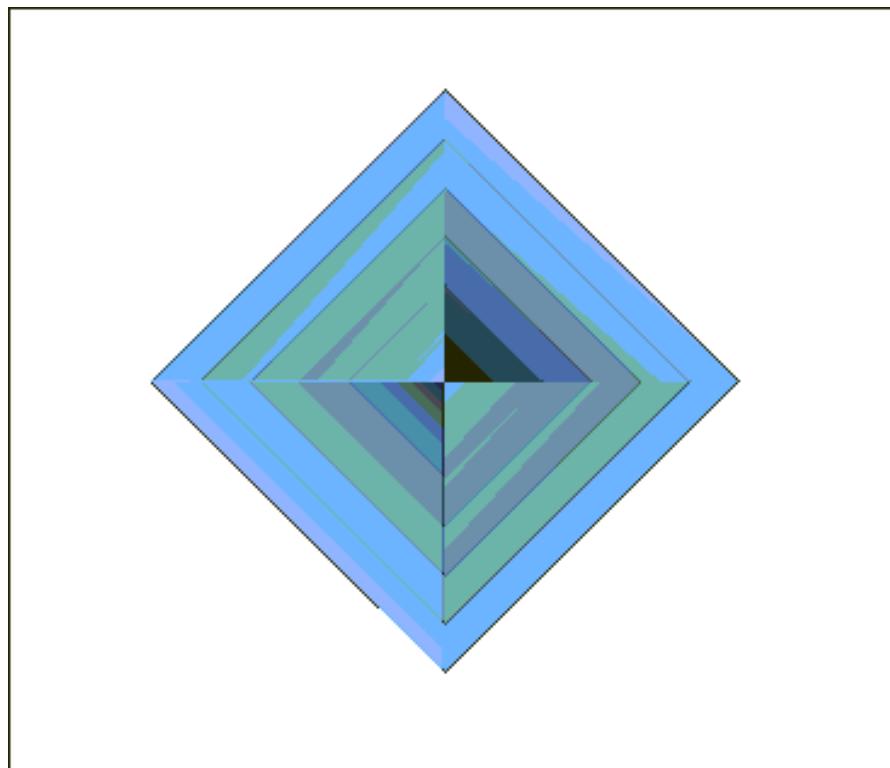
# $\text{La}_{5/8}\text{Ca}_{3/8}\text{MnO}_3$

## Isotope Dependence of $T_C$ & Spin Stiffness

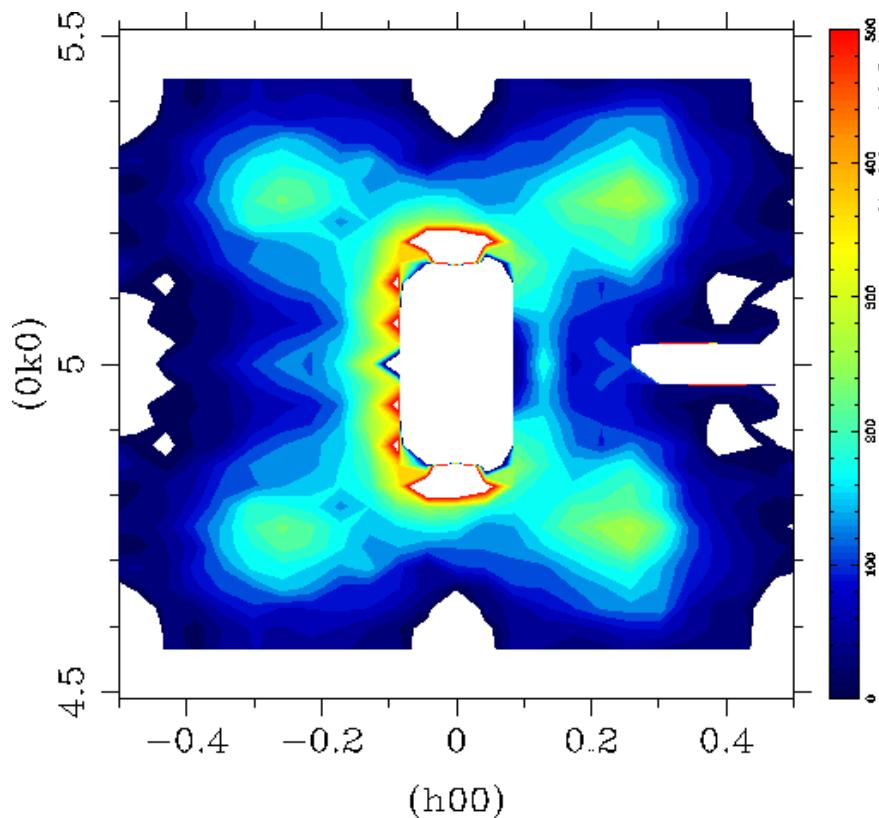


C. P. Adams, et al., Phys. Rev. B**70**, 134414 (2004)

# And there is coupling to the Lattice

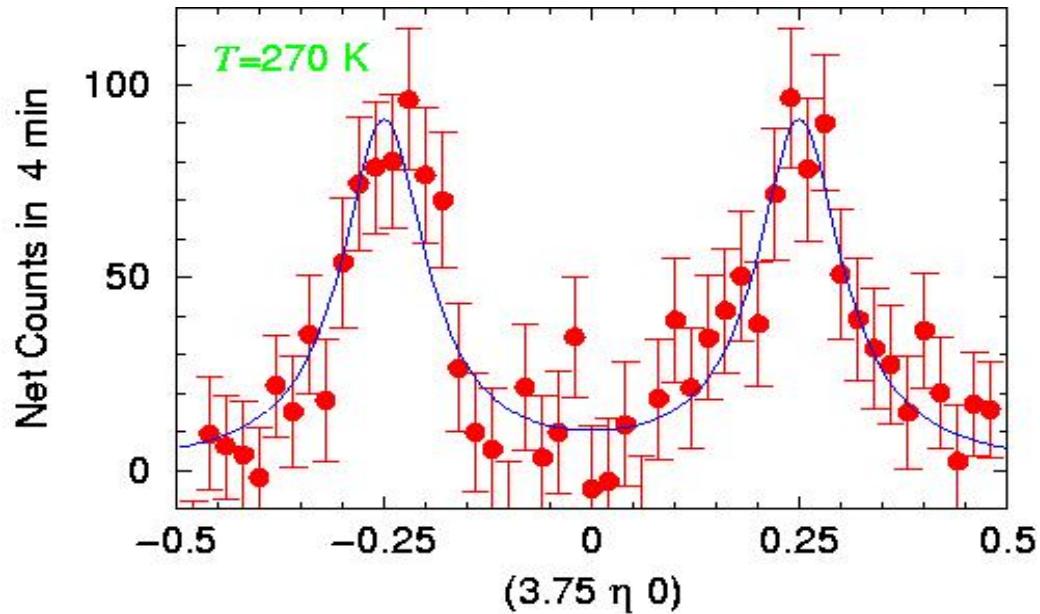


# Nanoscale Correlations in CMR Manganites



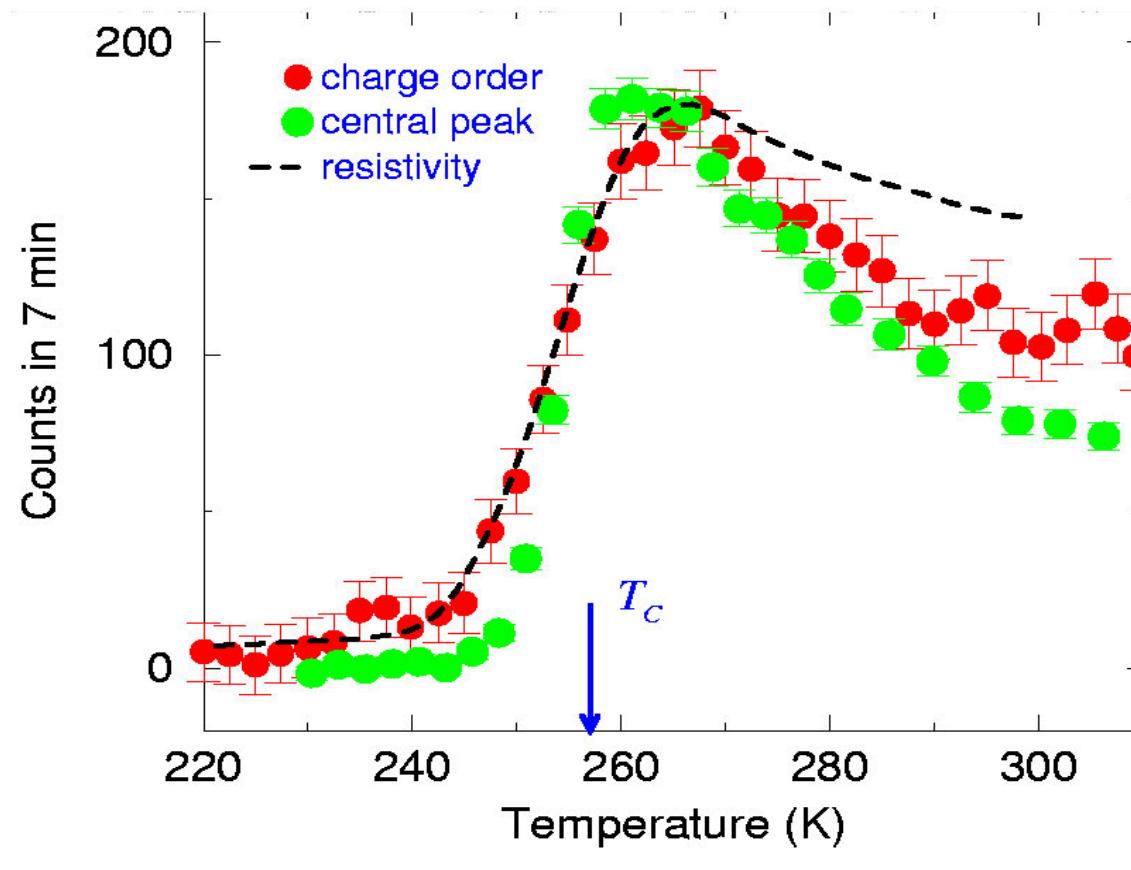
CE-type  $(1/4, 1/4, 0)$  peaks

# Polaron Peaks

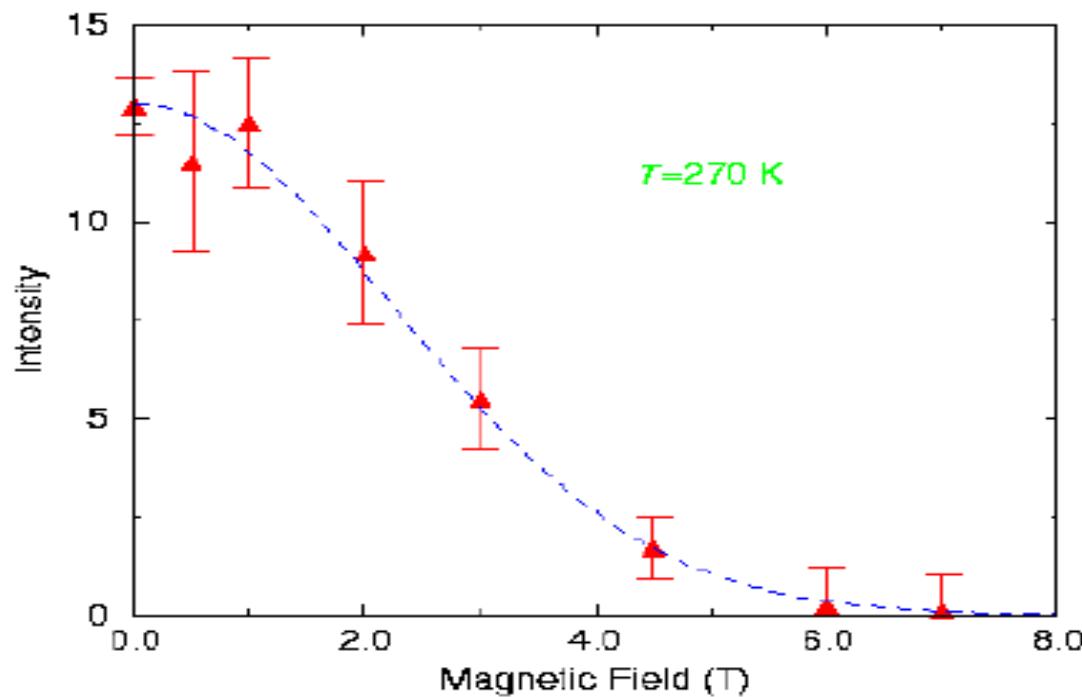


- C. P. Adams, J. W. Lynn, Y. M. Mukovskii, A. A. Arsenov, and D. A. Shulyatev, Phys. Rev. Lett. **85**, 3954 (2000).  
P. Dai, J. A. Fernandez-Baca, N. Wakabayashi, E.W. Plummer, Y. Tomioka, and Y. Tokura, Phys. Rev. Lett. **85**, 2553 (2000).

# T dependence

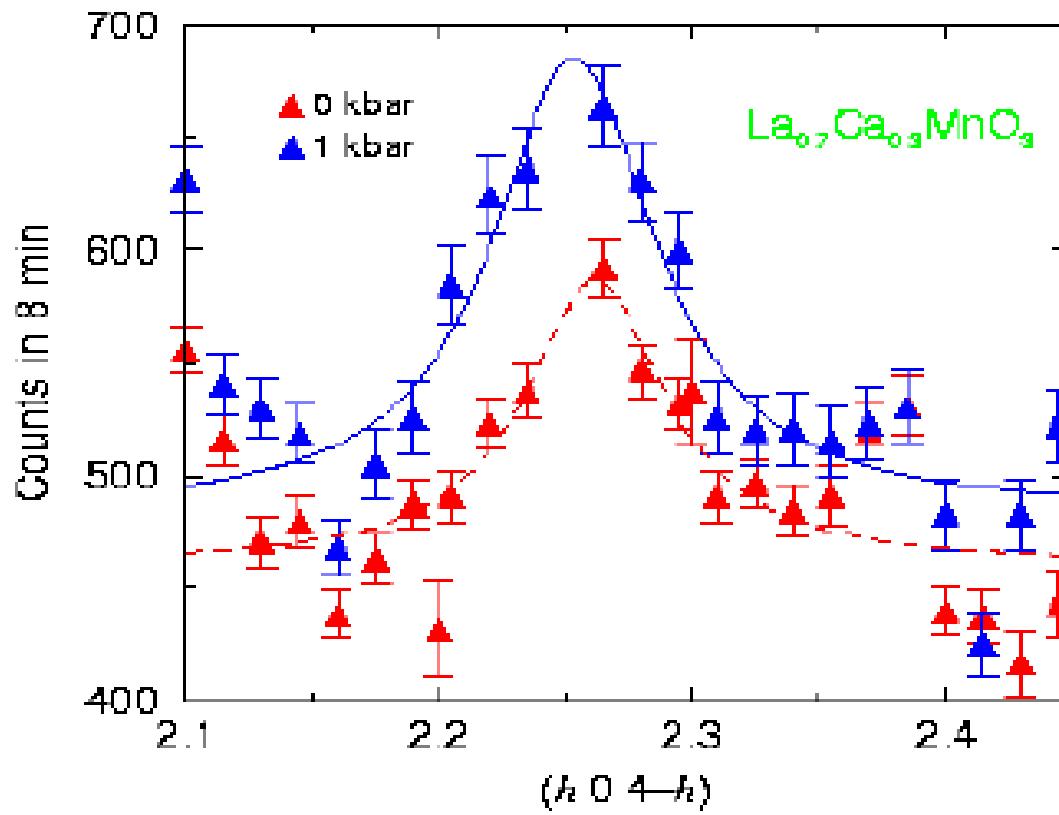


# $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ Field Dependence



J. Appl. Phys. **89**, 6846 (2001).

# $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ stress



C. P. Adams, et al., Phys. Rev. B**70**, 134414 (2004)

# T dependence

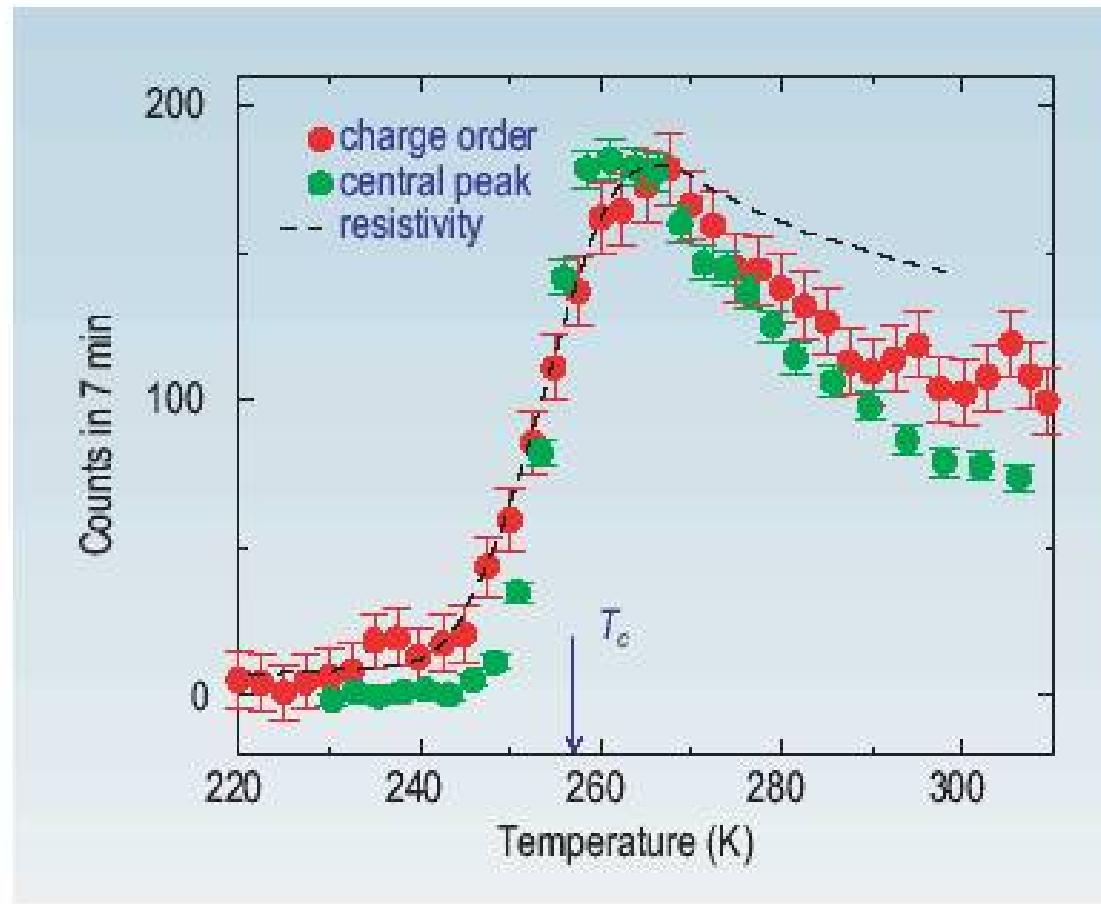


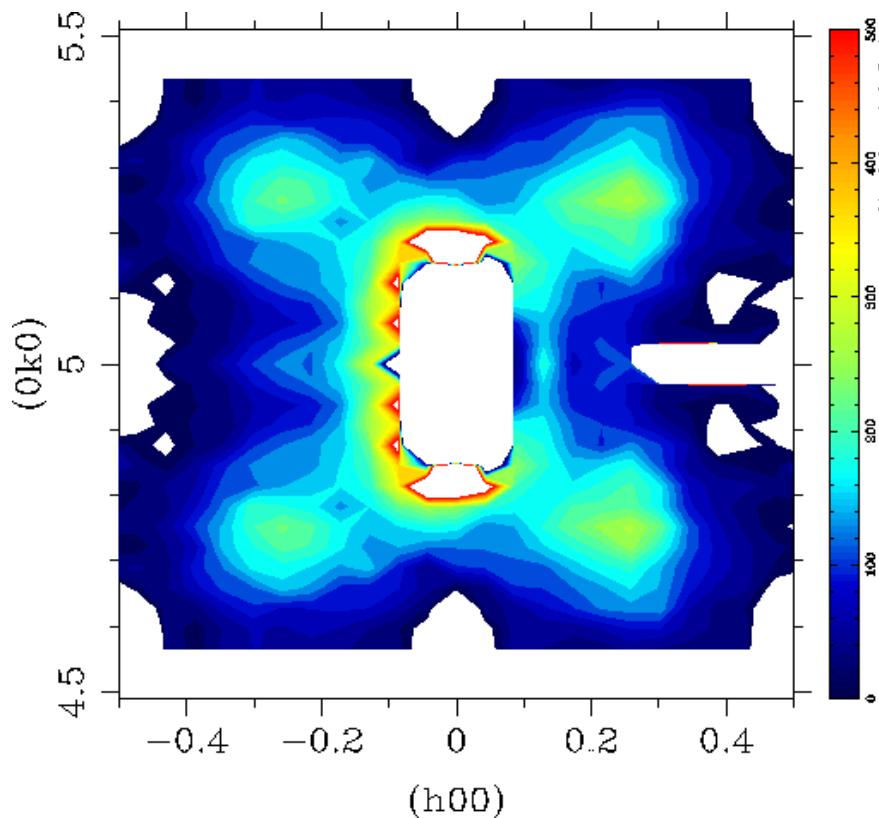
FIGURE 2. Temperature dependence of the intensity of the polaron peak at 3.75 0.25 0, compared to the magnetic quasielastic central peak, and the resistivity. The data have been scaled vertically so the peak heights match. The similarity of the data indicates a common physical origin.

# Single Polaron

vs.

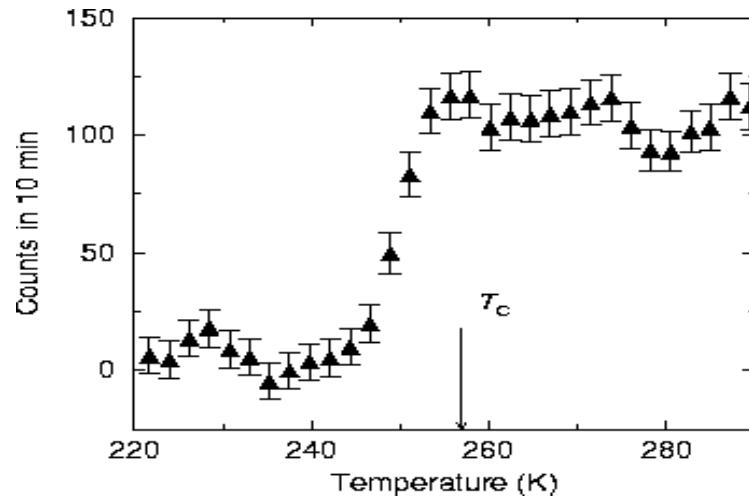
# Correlated Polarons

# Nanoscale Correlations in CMR Manganites

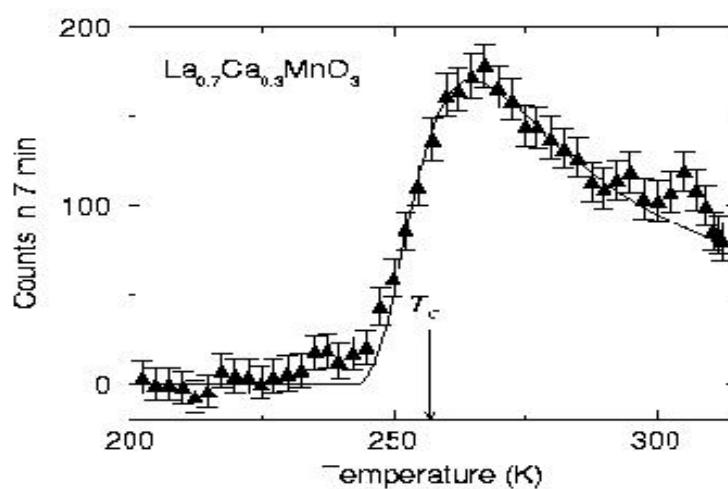


CE-type  $(1/4, 1/4, 0)$  peaks

# Two Types of Polaron Scattering



Single  
Polaron



Polaron  
Correlation  
Peak

# First-Order Nature of Transition

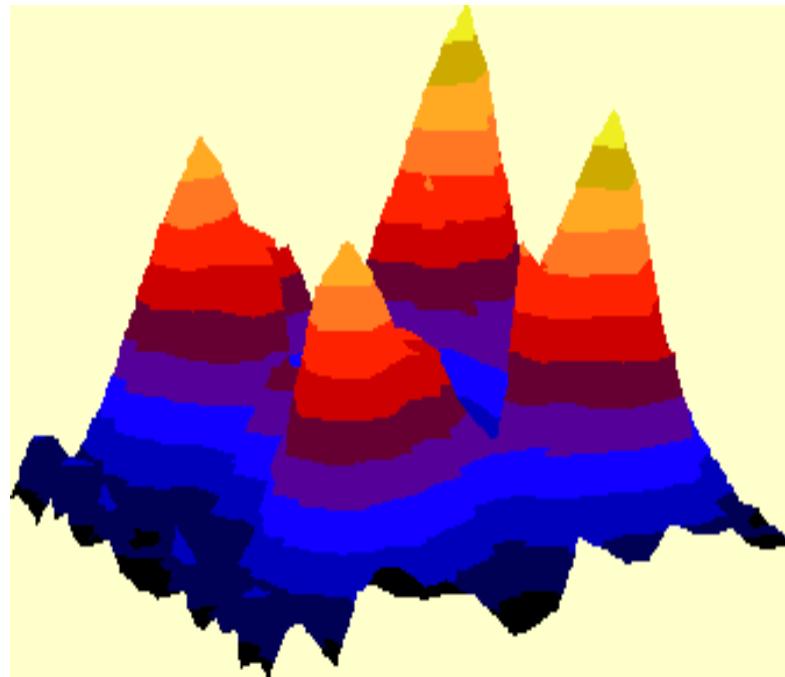
CMR: Polaron Formation Truncates  
Ferromagnetic Phase and enhances MR  
 $D(T)$  discontinuous

Two-phase coexistence

Long-Time relaxation of physical quantities  
near  $T_C$

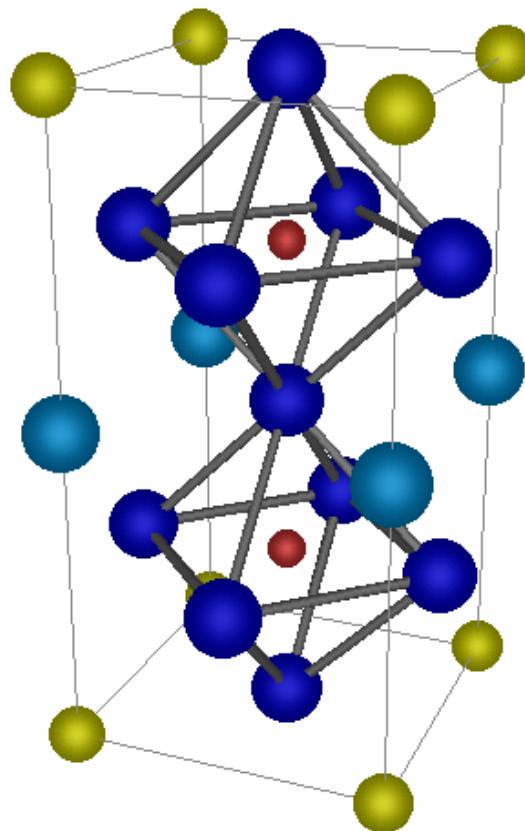
Single Polaron—diffuse (structural) scattering  
Correlated Polaron Peaks (structural)  
Quasielastic paramagnetic scattering

# Polaron “Mountains” in CMR Manganites

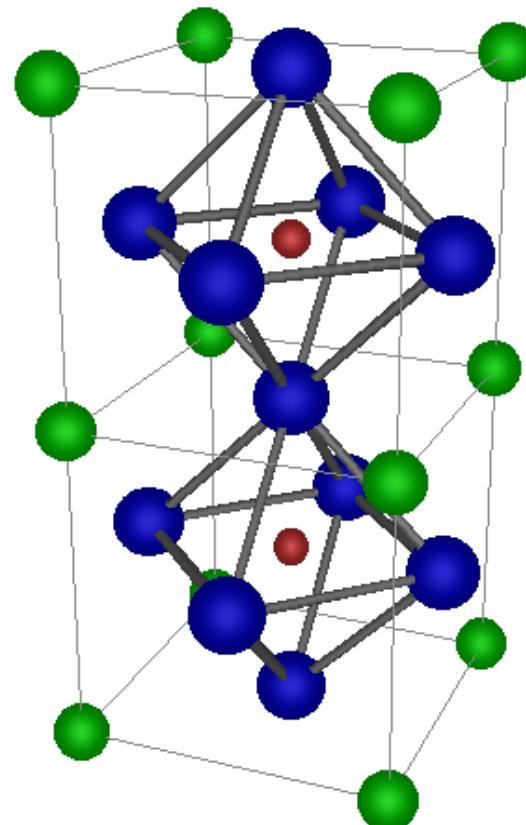


What Controls the Polaron Formation??

# $\text{La}_{0.5}\text{Ba}_{0.5}\text{MnO}_3$



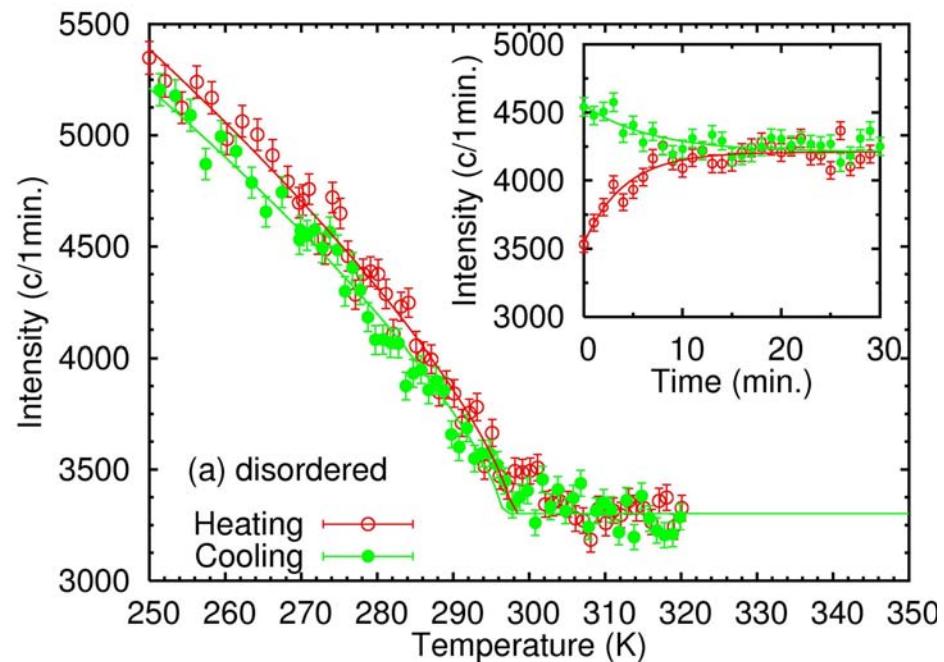
Chemically Ordered



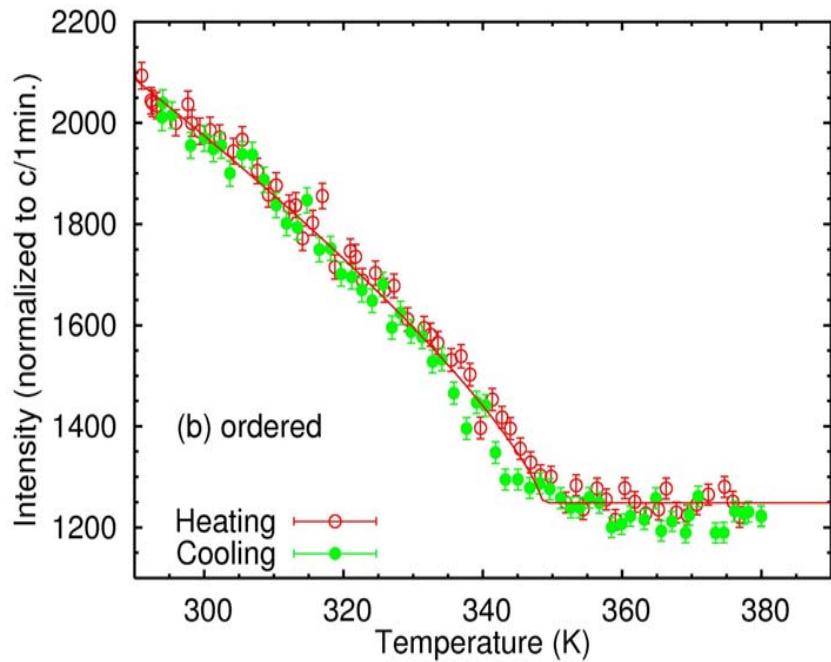
Chemically Disordered

# $\text{La}_{0.5}\text{Ba}_{0.5}\text{MnO}_3$ Order Parameters

Chemically Disordered

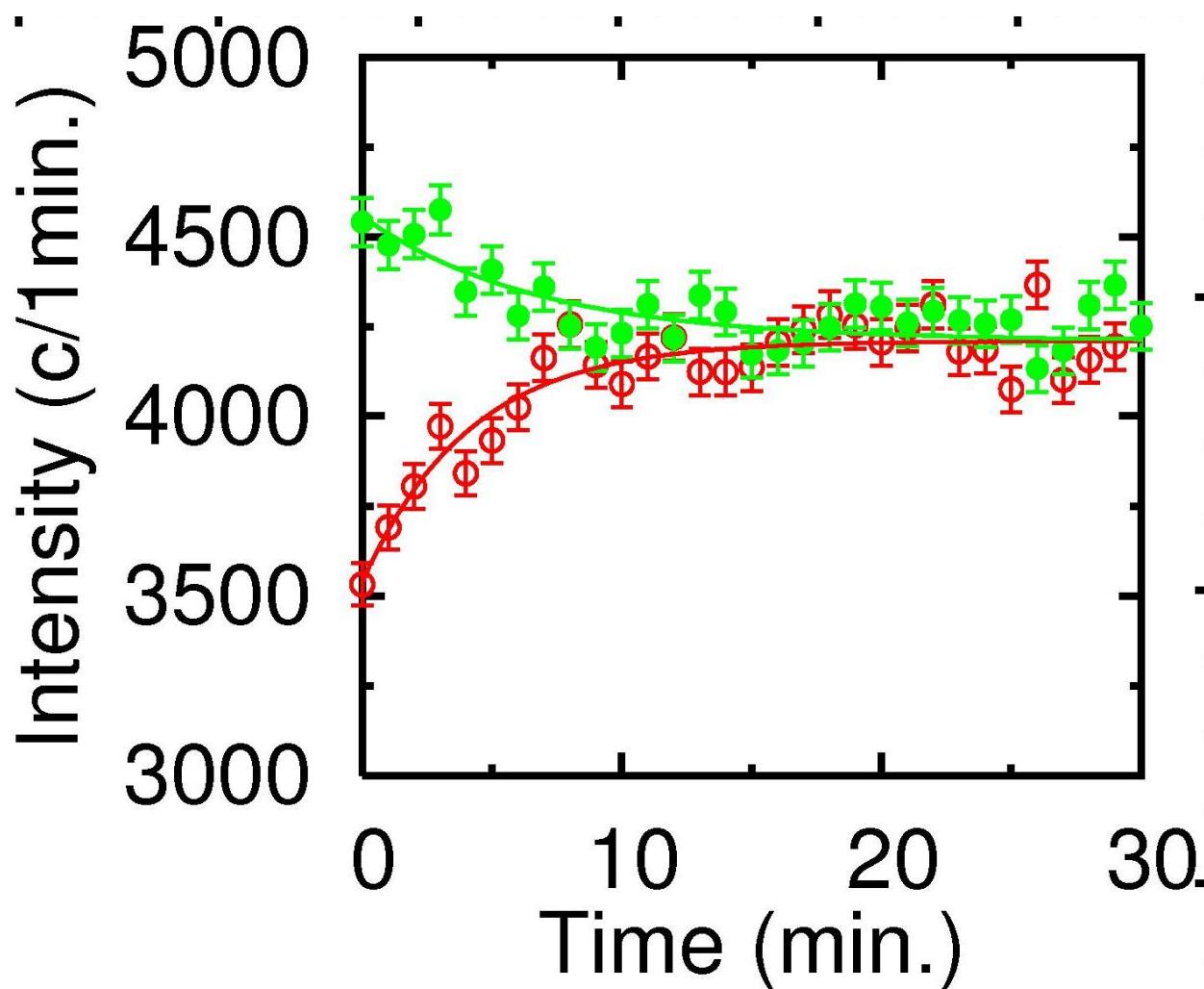


Chemically Ordered

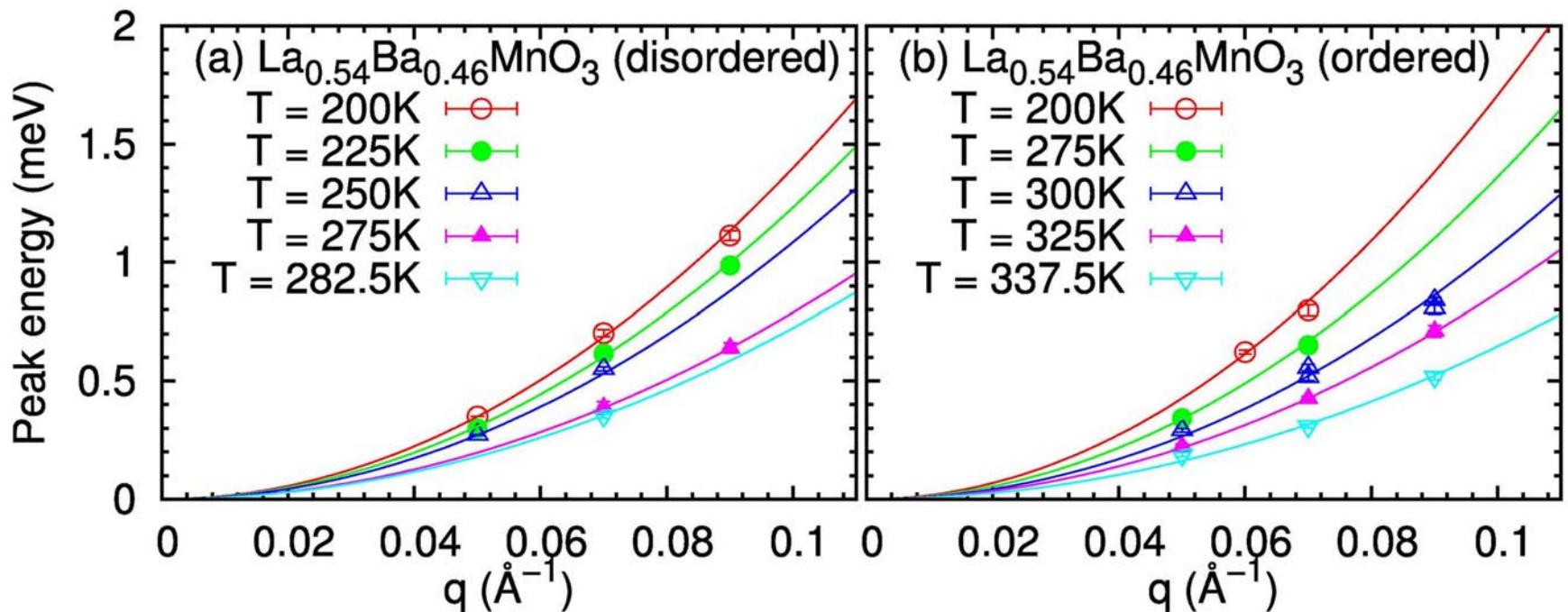


T. Sato, J. W. Lynn, and B. Dabrowski,  
Phys. Rev. Lett. **93**, 267204 (2004).

# Time Dependence



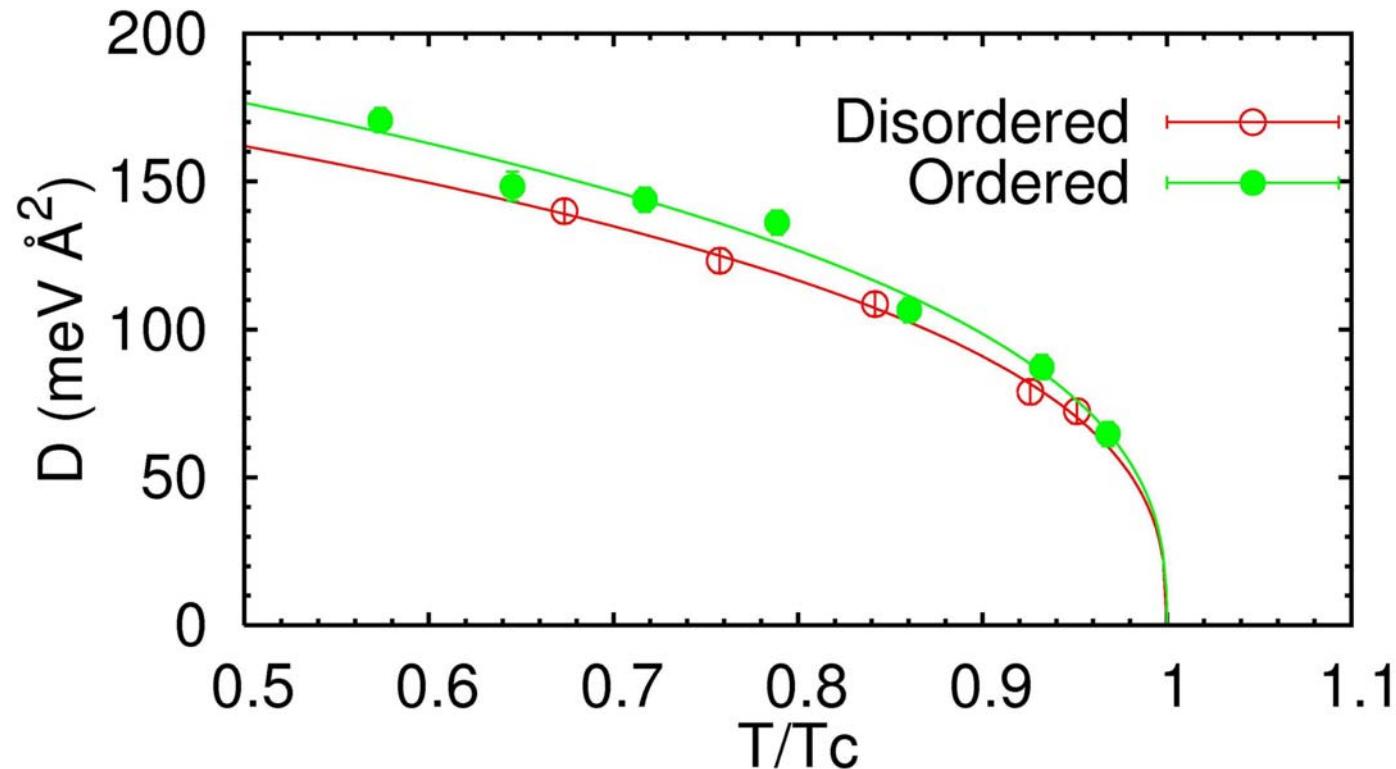
# $\text{La}_{0.5}\text{Ba}_{0.5}\text{MnO}_3$ Dispersion



Chemically Disordered

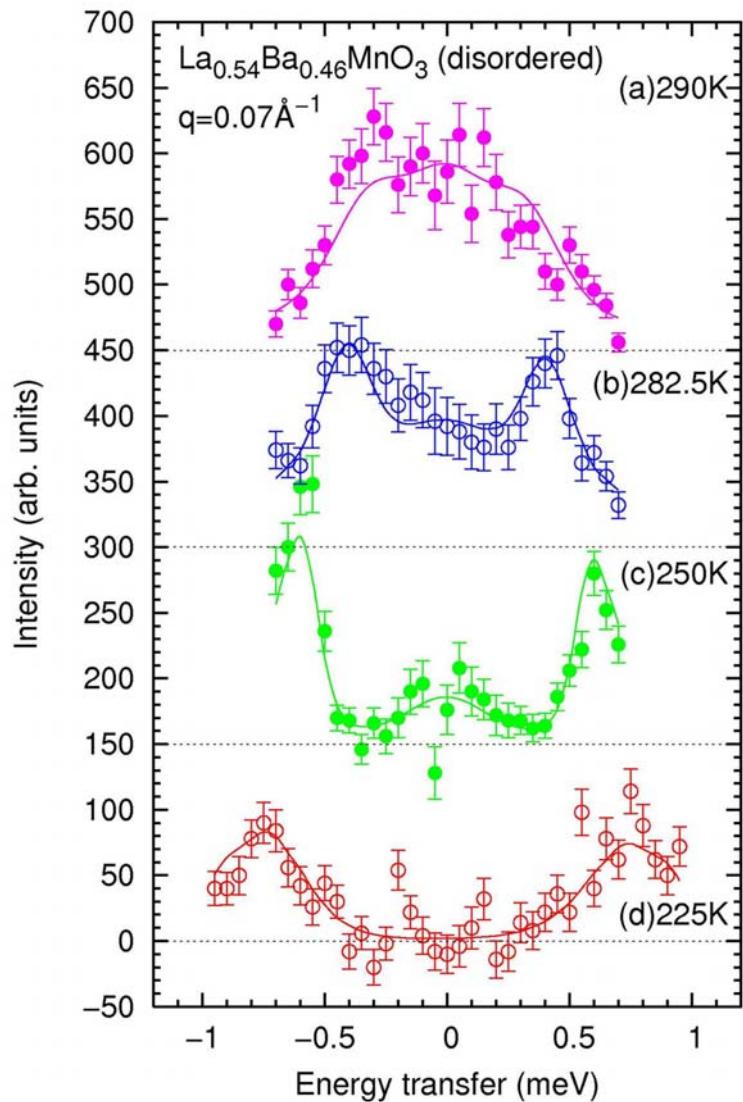
Chemically Ordered

# $\text{La}_{0.5}\text{Ba}_{0.5}\text{MnO}_3$ Exchange Energy

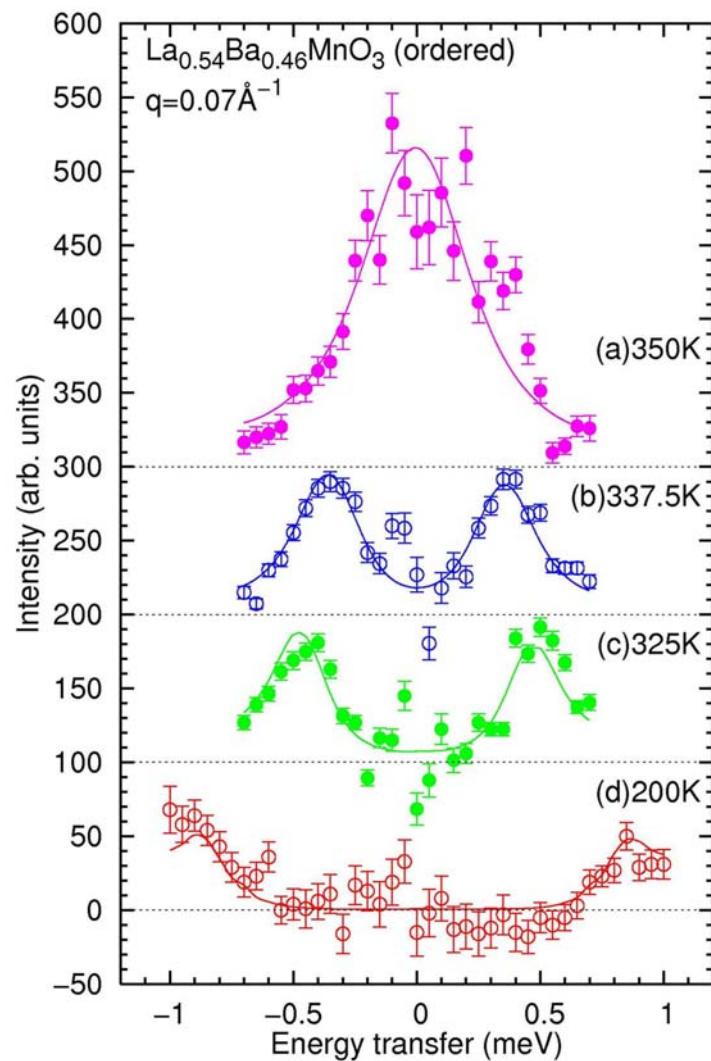


T. Sato, J. W. Lynn, and B. Dabrowski,  
Phys. Rev. Lett. **93**, 267204 (2004).

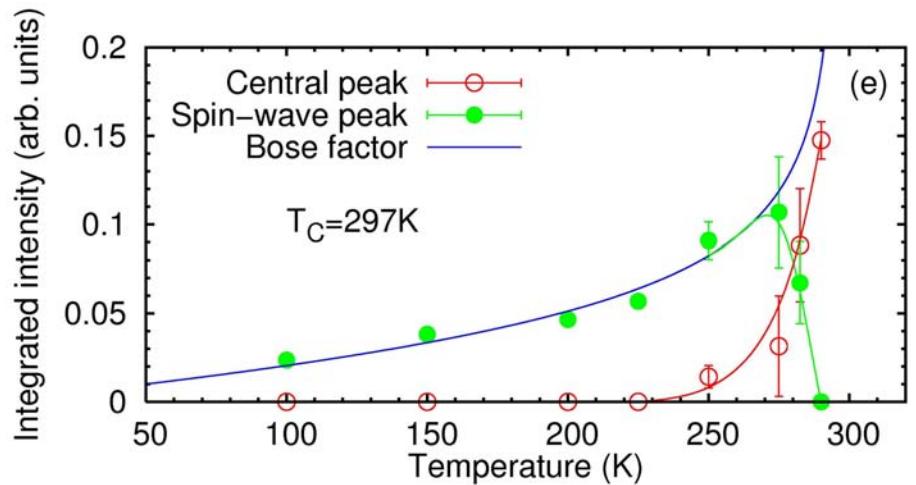
# Chemically Disordered



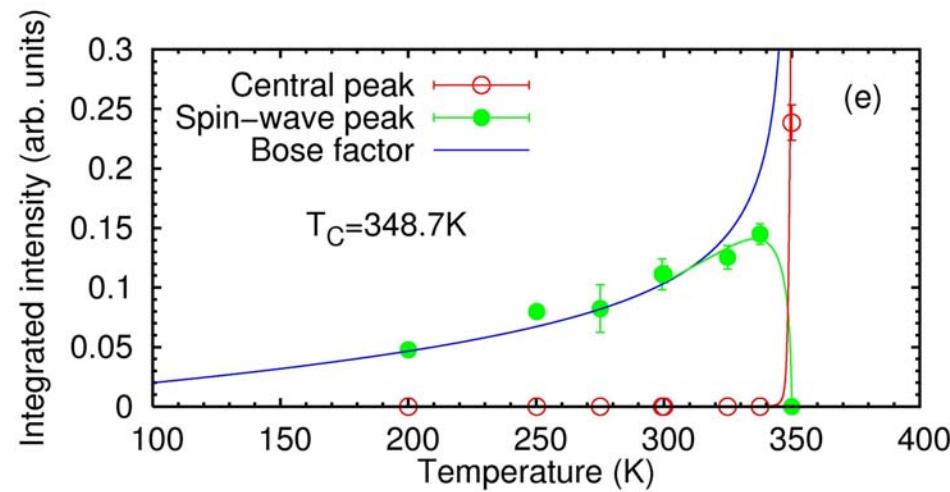
# Chemically Ordered



# $\text{La}_{0.5}\text{Ba}_{0.5}\text{MnO}_3$ Intensities



Chemically Disordered

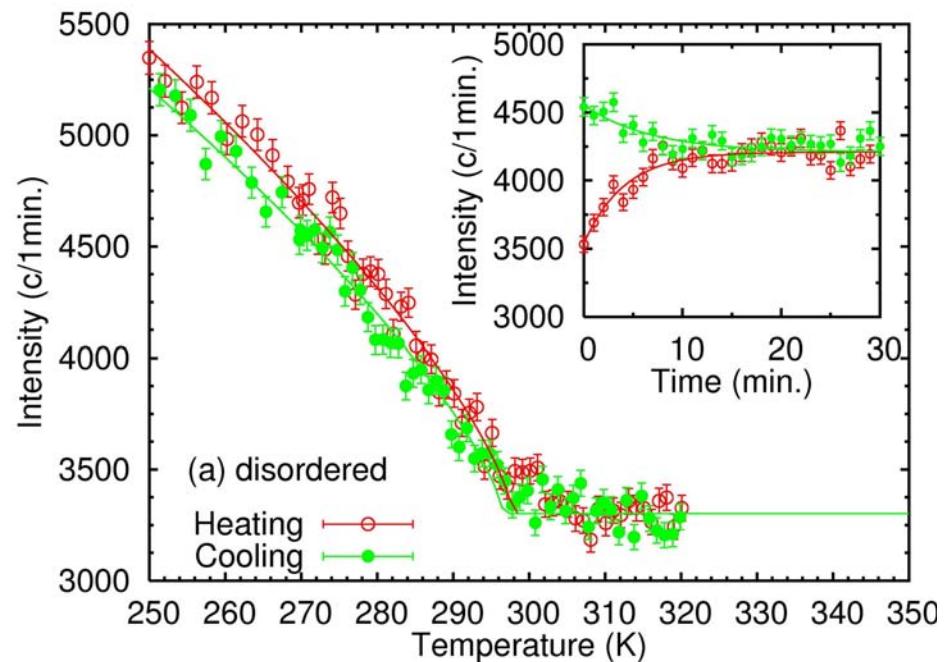


Chemically Ordered

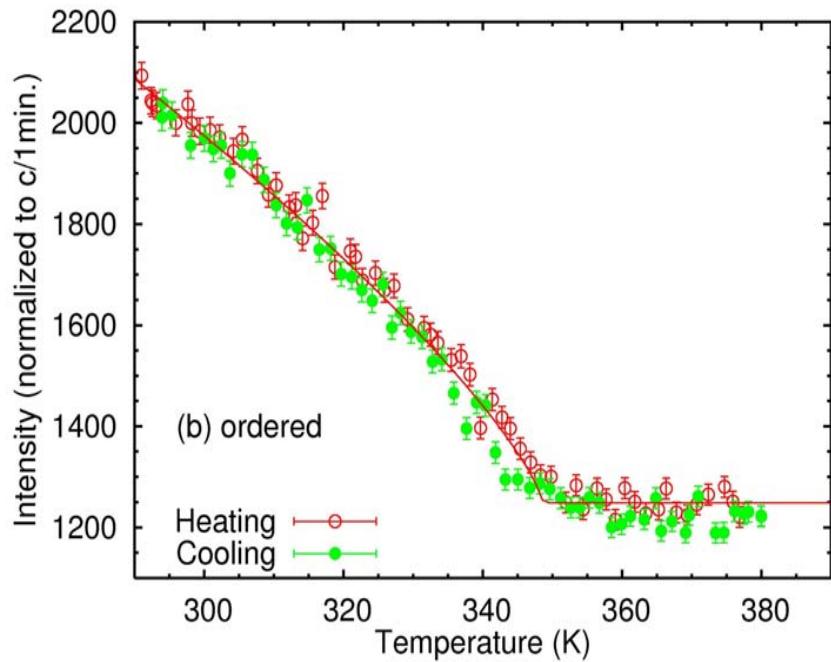
DisOrder assists Polaron Formation

# $\text{La}_{0.5}\text{Ba}_{0.5}\text{MnO}_3$ Order Parameters

Chemically Disordered



Chemically Ordered



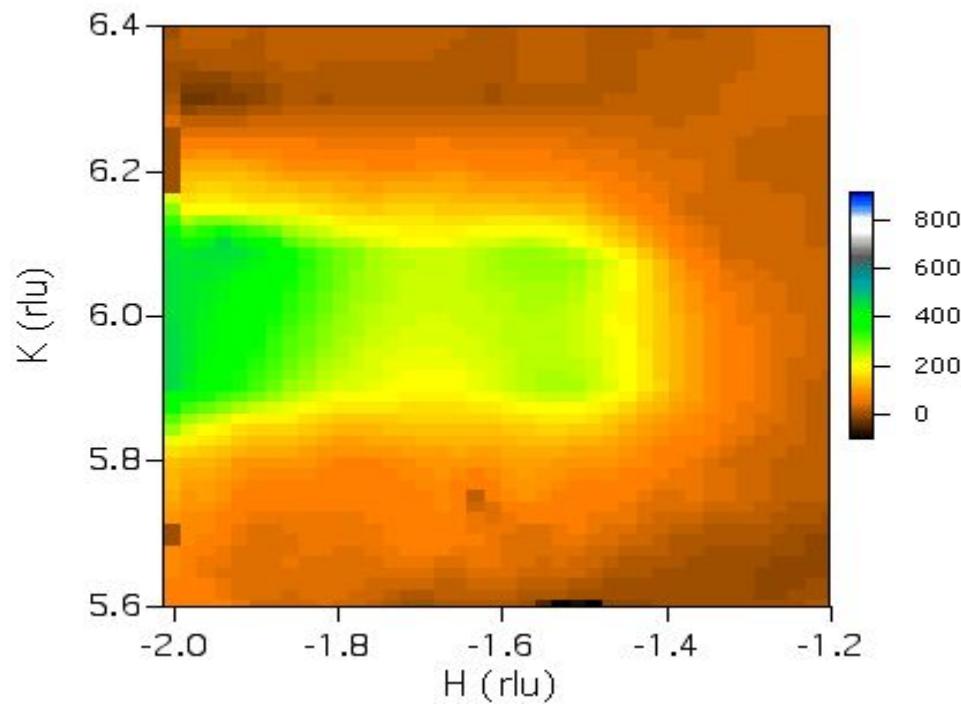
T. Sato, J. W. Lynn, and B. Dabrowski,  
Phys. Rev. Lett. **93**, 267204 (2004).

# What about the Dynamics of the Polarons?

# $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$

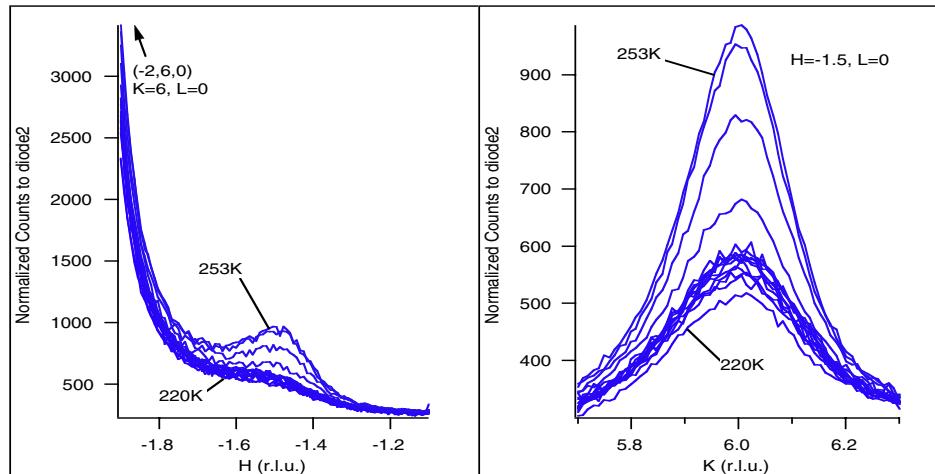
## X-ray Diffuse Map

I(300 K - 220 K)



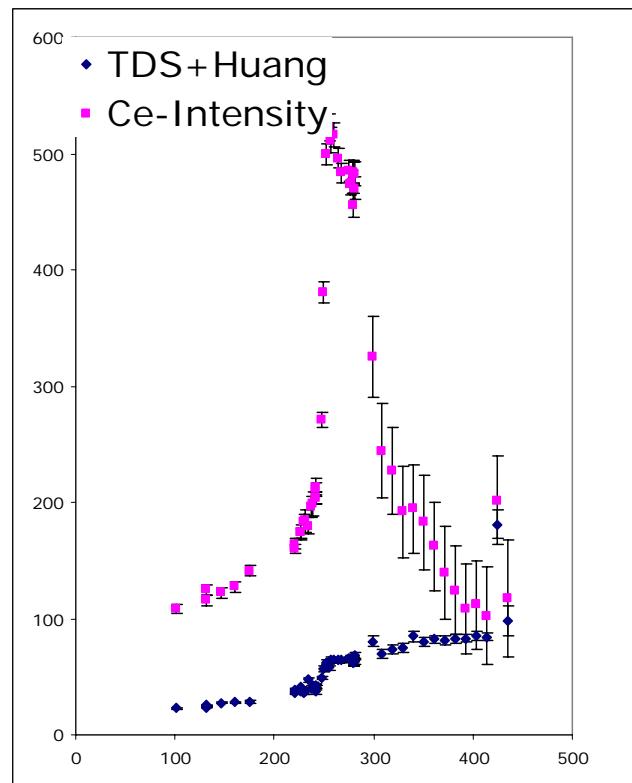
Advanced Photon Source (BESERC)  
115 keV

# X-ray Diffuse Scattering

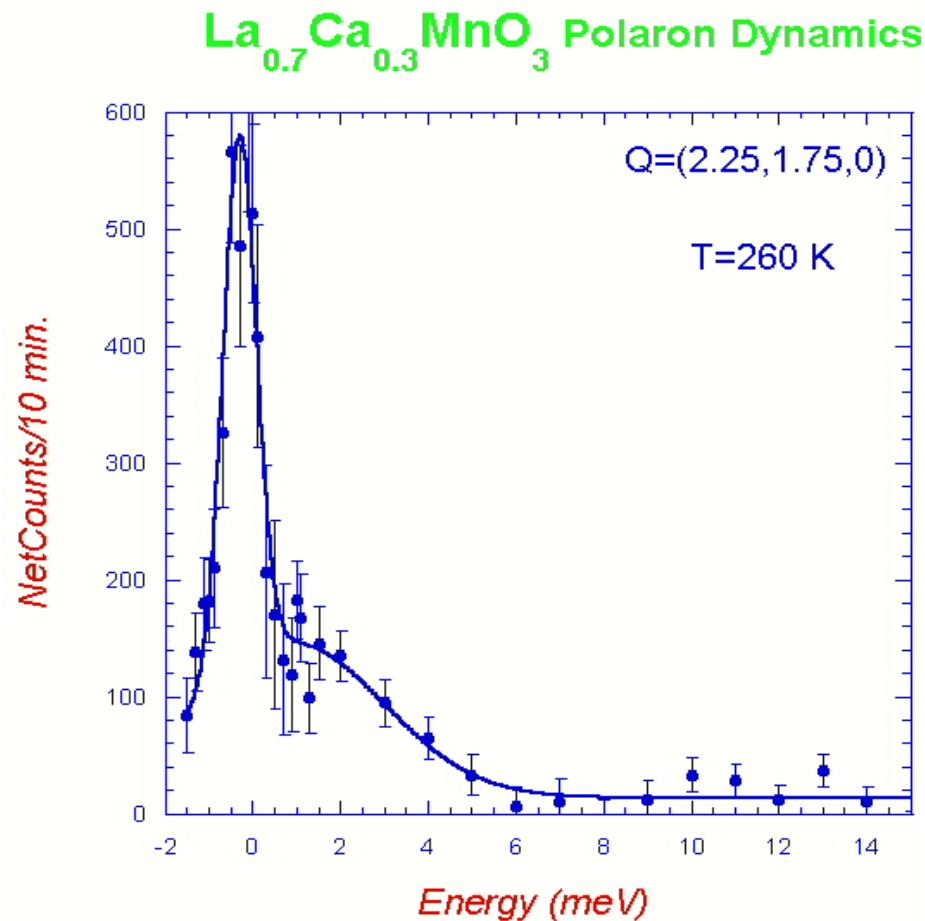


APS BESERC beam line  
(with D. Argyriou, Y. Ren, Y. Mukovskii)

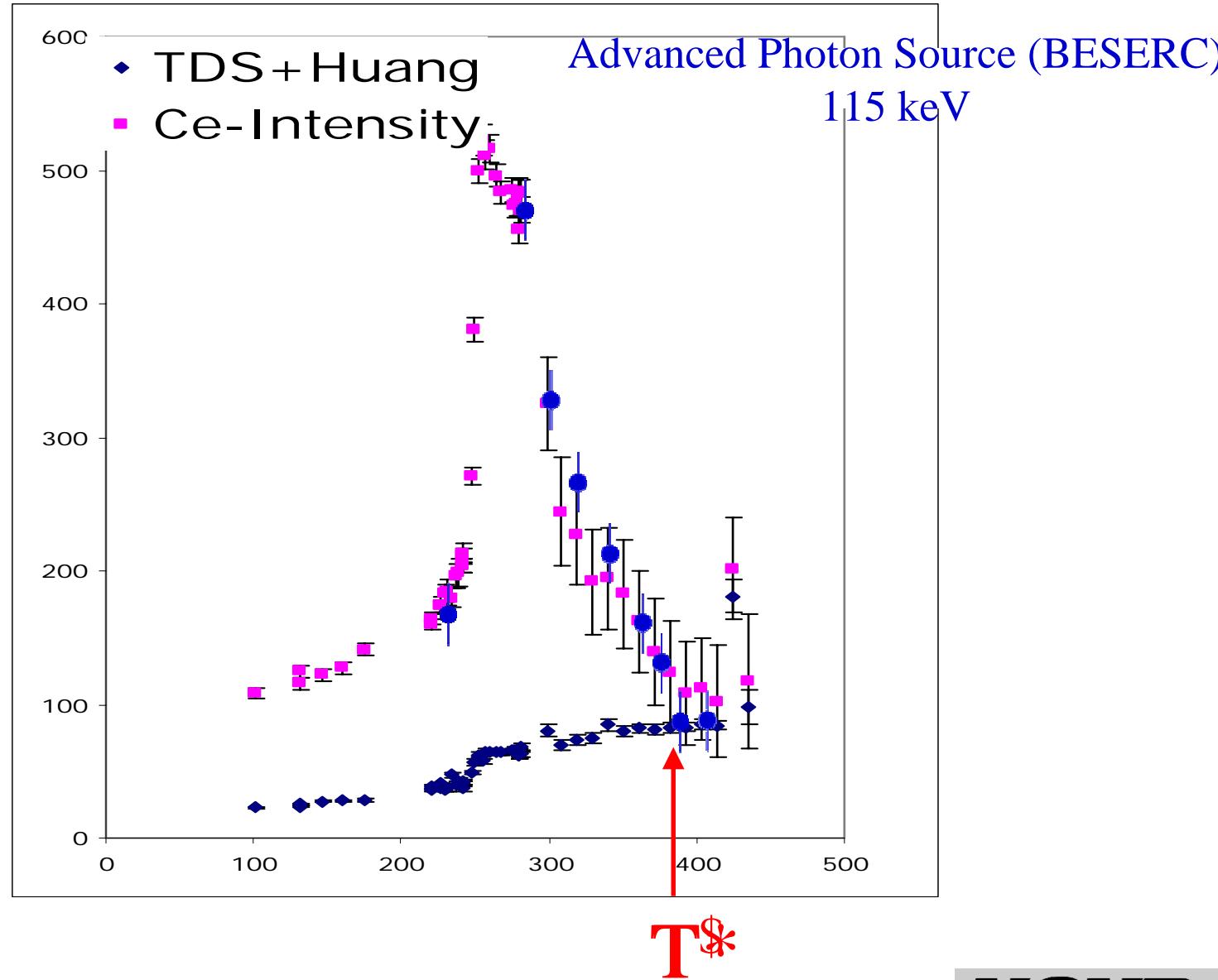
# X-ray Diffuse Scattering



# Polaron Dynamics

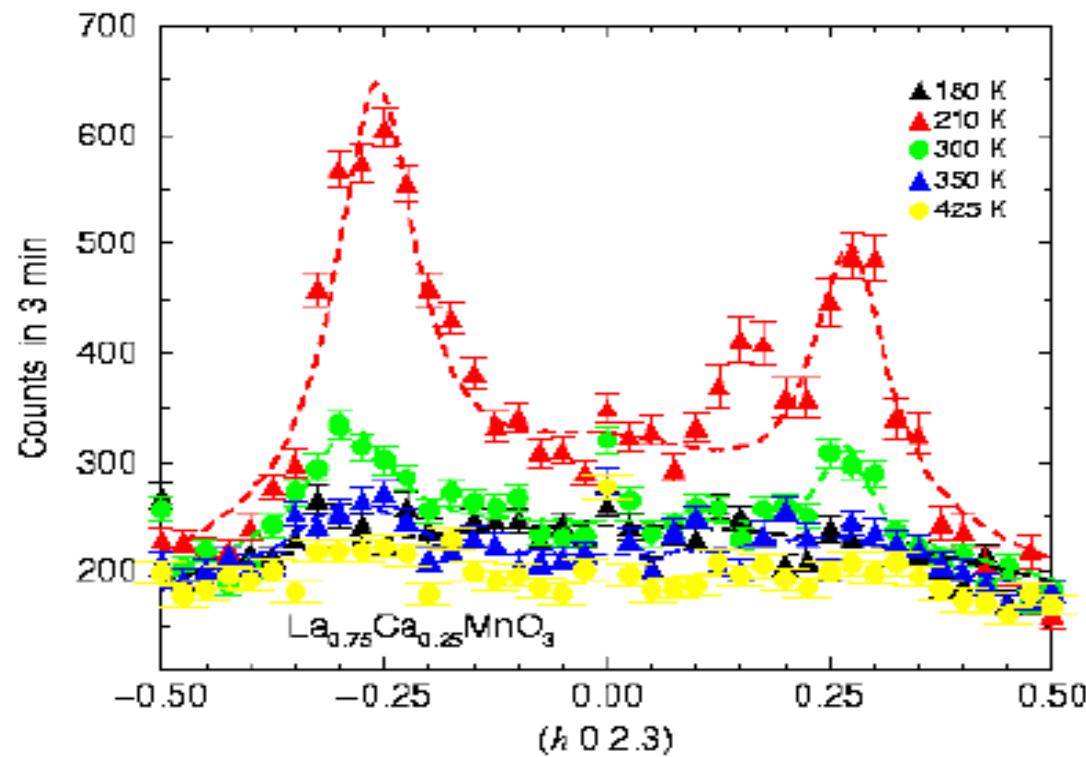


# X-ray Diffuse Scattering



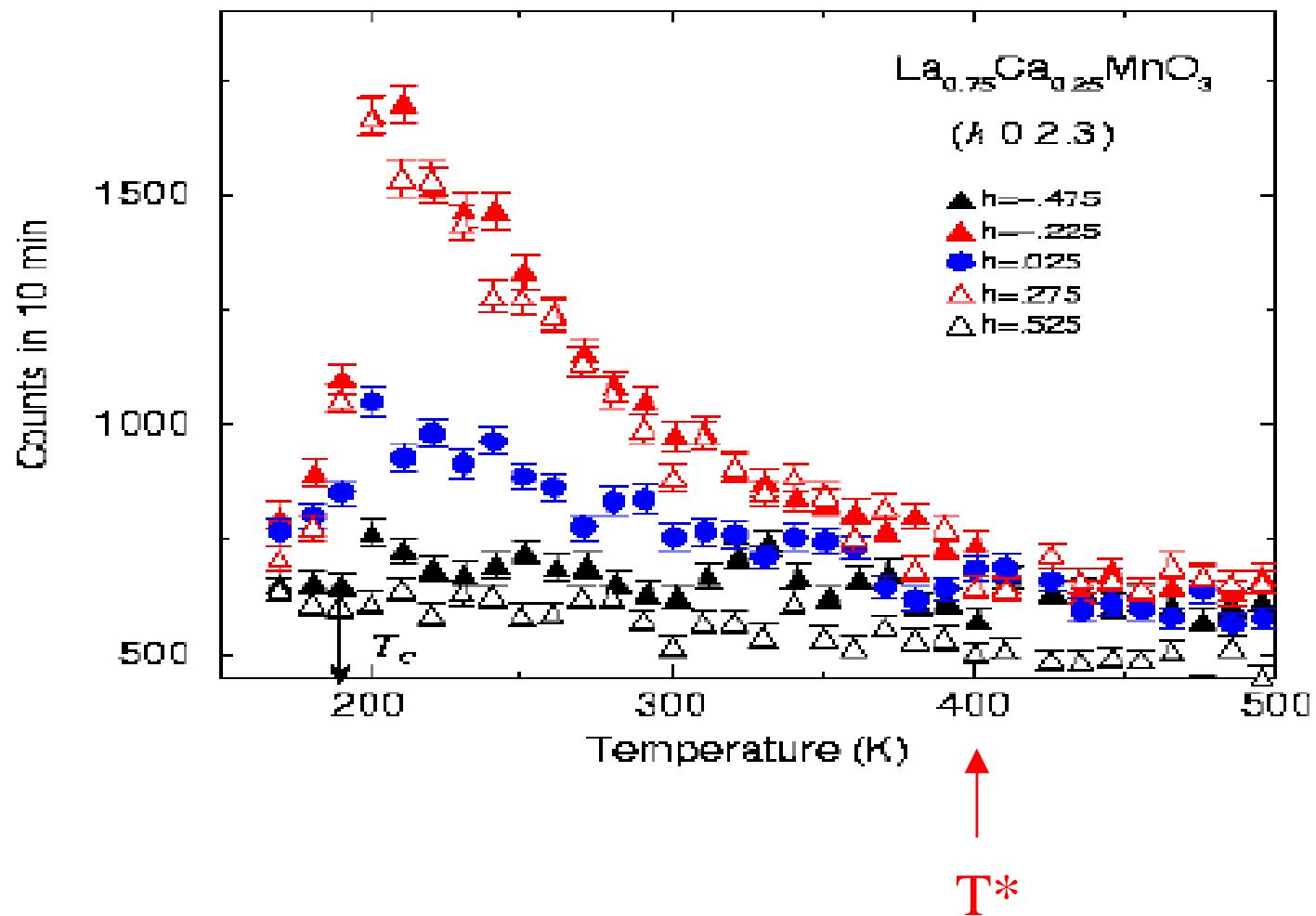
$T\$$

# LCMO $x=0.25$



With J. Fernandez-Baca, P. Dai

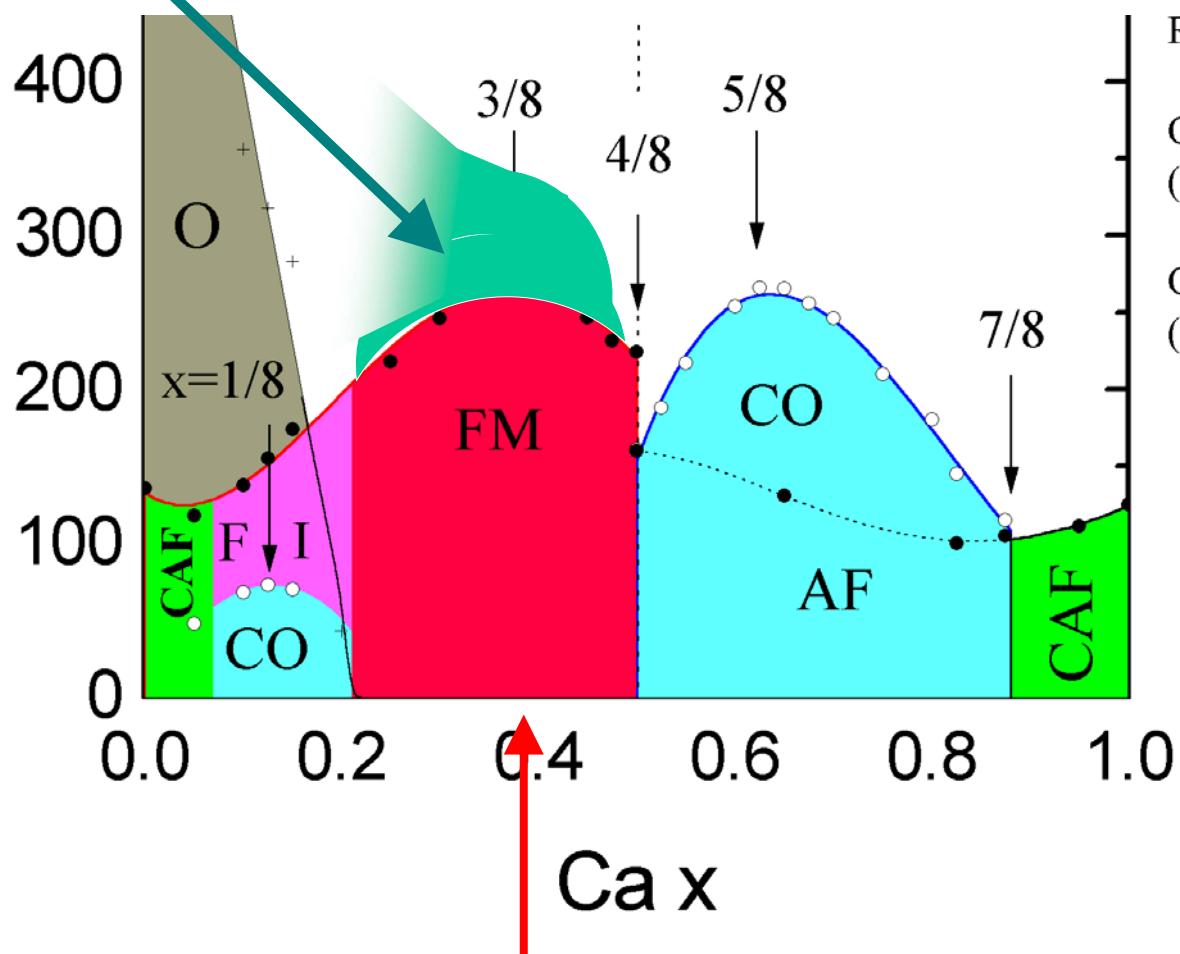
# T dependence



# $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$

Phase Diagram

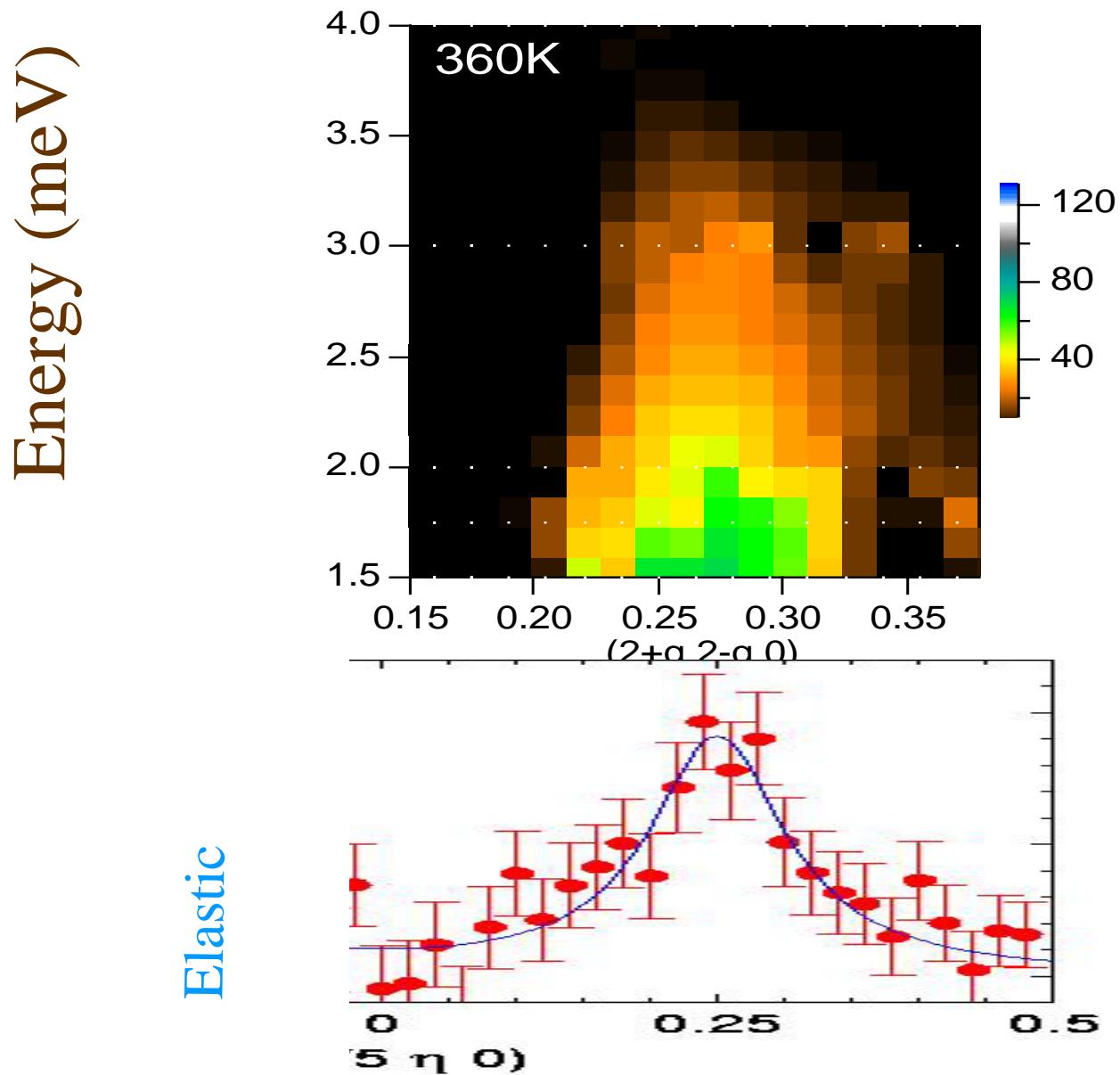
Polaron Glass



S-W. Cheong and C. H. Chen

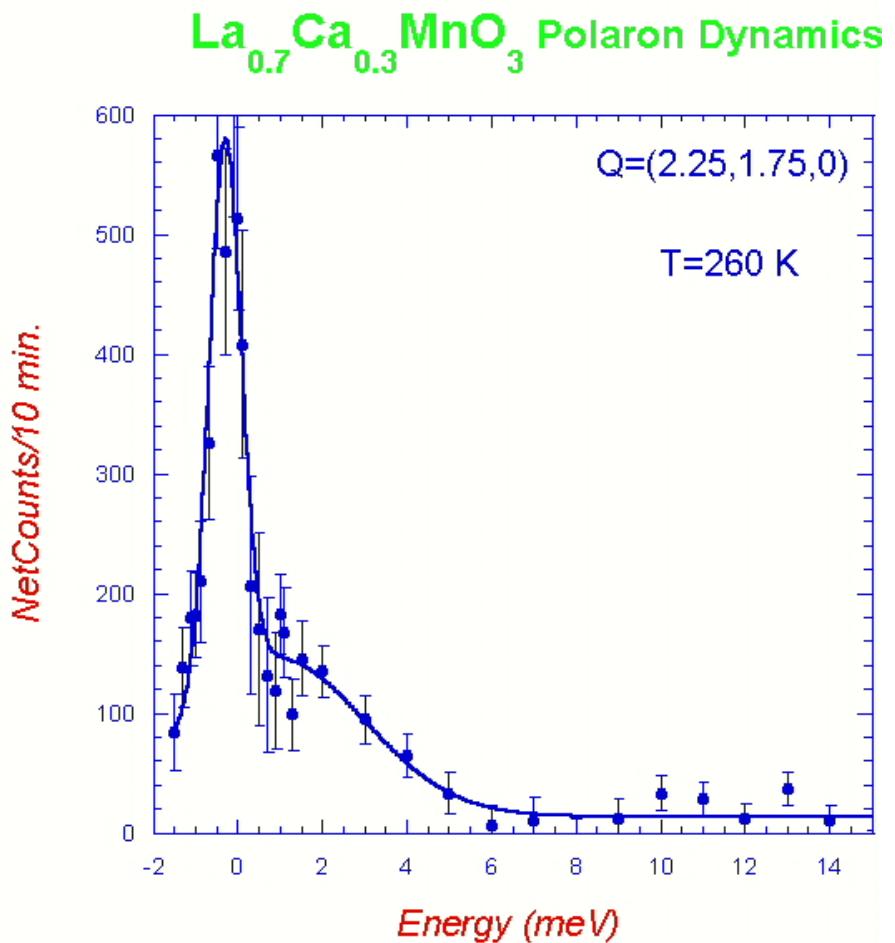
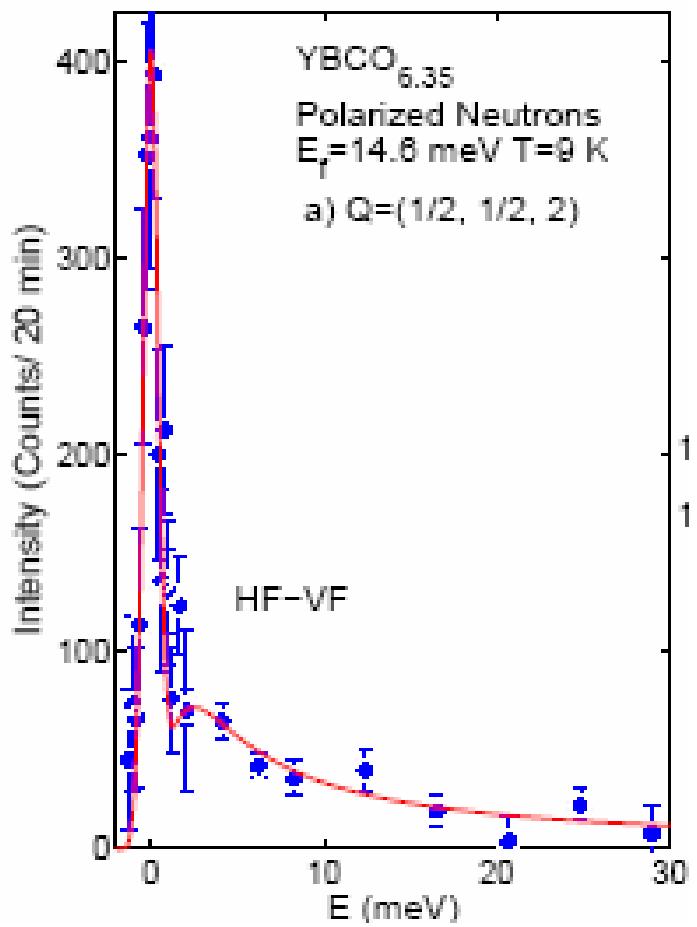
*Colossal Magnetoresistance, Charge Ordering, and Related Properties of Manganese Oxides* (World Scientific, 1998),  
p. 241 (Ed. by Raveau and Rao)

# Polaron Dynamics



D. N. Argyriou, *et al.*, Phys. Rev. Lett. 89, 036401(2002).

# Spin Glass in YBCO<sub>6.353</sub>



YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.353</sub> ( $T_c=18$  K)  
C. Stock, et al., cond-mat 0505083

# Nanoscale Correlations in Oxides

- Manganites
  - Spin, Charge, & Lattice coupled
  - *Highly* correlated electrons
  - Competing States—Delicate Balance
  - Ferromagnetic metal  $\Leftrightarrow$  Polaron Glass  $\Leftrightarrow$  Polaron Fluid
- Relaxor Ferroelectrics
  - $\text{PbTiO}_3$
  - $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  (PMN)
  - Nanopolar ferroelectric droplets
- Cuprates and Nickelates
  - Charge Stripes and Spin Stripes

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