NEUTRON SCATTERING AS A PROBE OF THE MAGNETIC, STRUCTURAL, AND SPIN DYNAMICAL PROPERTIES OF COLOSSAL MAGNETORESISTIVE MANGANITES

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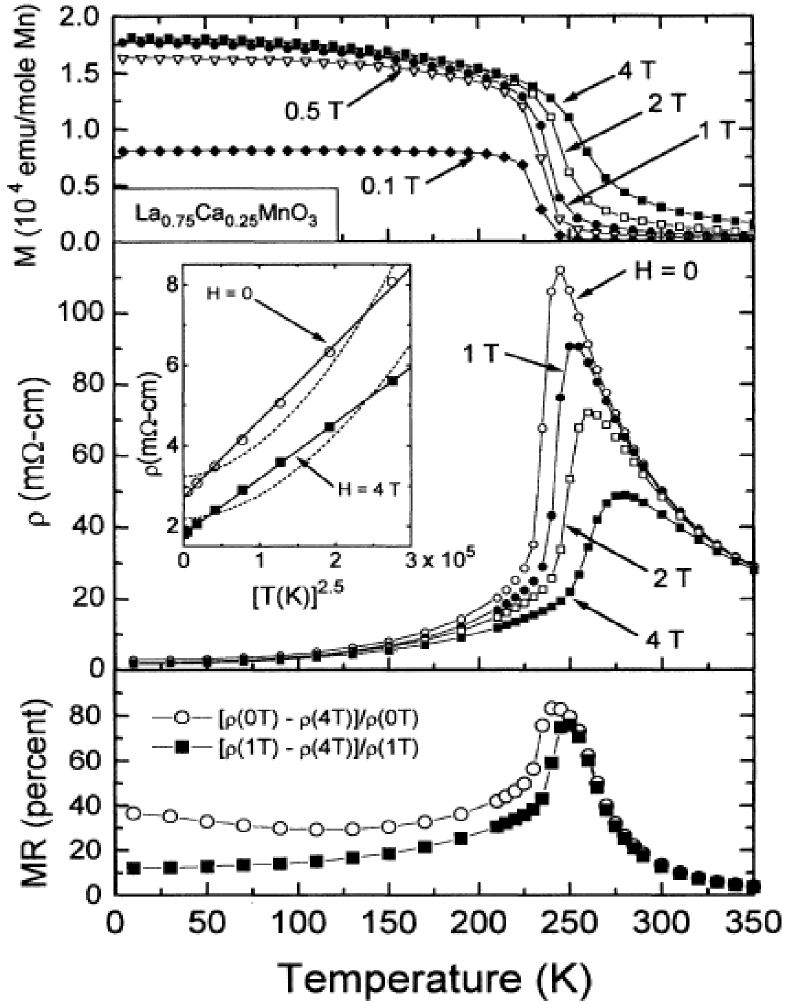
Colossal Magnetoresistance

$La_{1-x}Ca_xMnO_3$ and related materials

Phase transition between a ferromagnetic metal and a paramagnetic insulator

Applied magnetic field changes resistivity by orders of magnitude

Technological applications in commercial magnetic field sensors: hard drive read heads, etc.

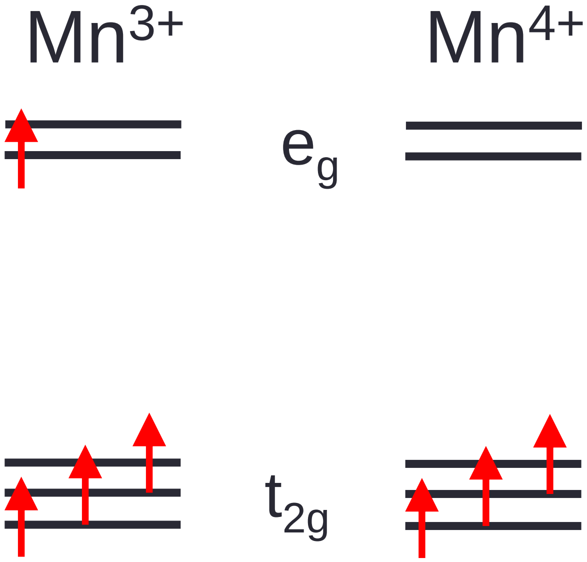


P. Schiffer, *et al.*, *PRL* **75** 3336 (1995)

$La_{1-x}Ca_xMnO_3$

- Perovskite structure
- Pseudo-cubic, $a = 3.88$ Å
- $x = 0.3$

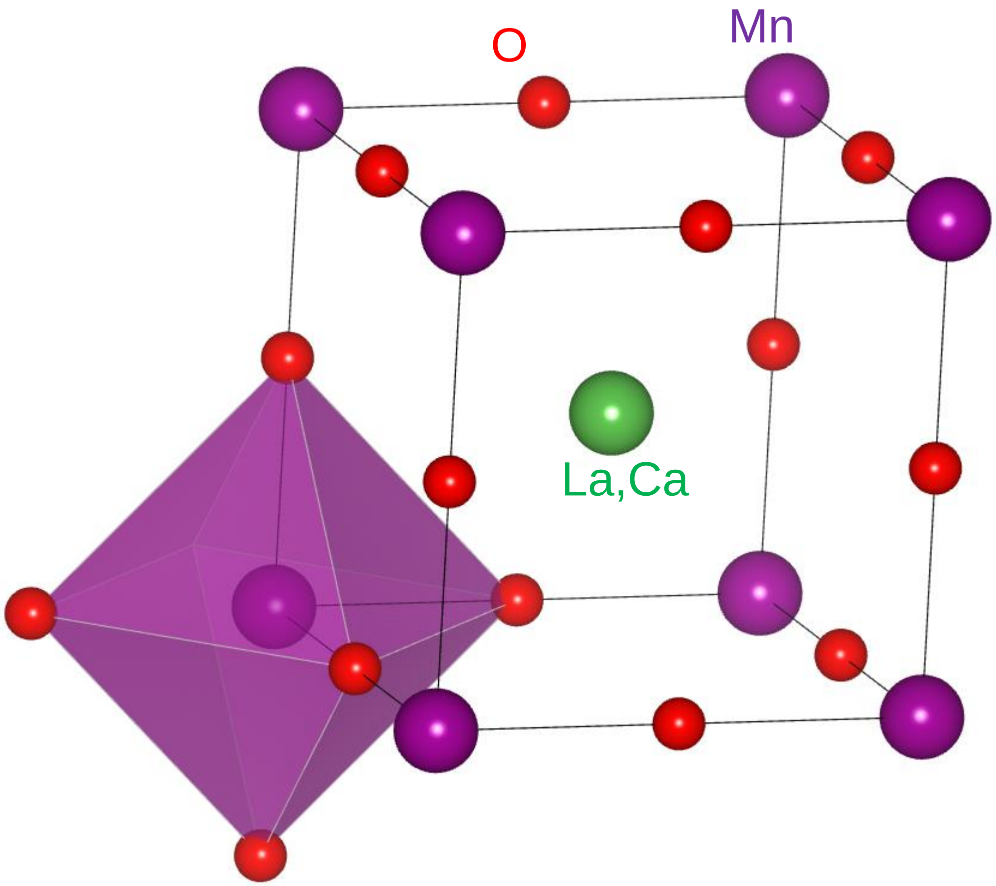
- Average Mn oxidation: $(3+x)^+$



Mn^{3+} Mn^{4+}

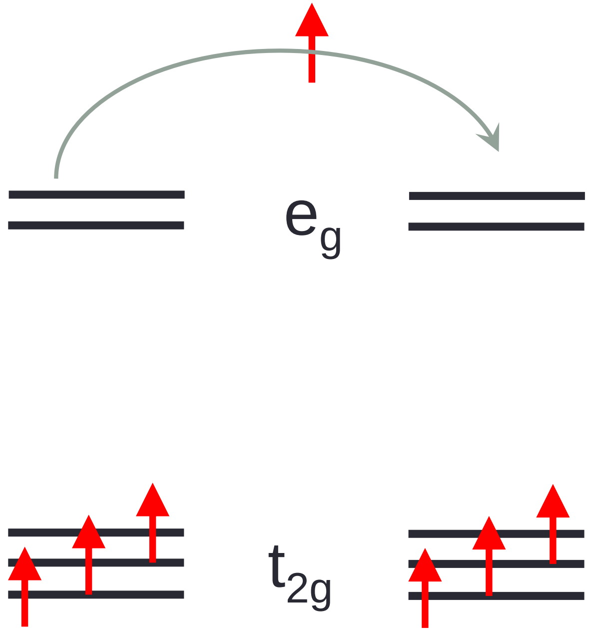
e_g

t_{2g}



Coupling between lattice, charge, and magnetic degrees of freedom

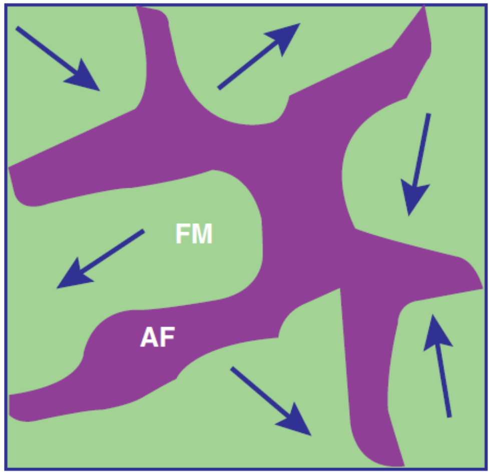
- charge ↔ magnetism
  - Zener double exchange



- Hund's Rules
  - All electrons on a site will have aligned magnetic moments
  - S=2 (Mn^{3+}) or S=3/2 (Mn^{4+})
  - An e_g conduction electron can hop between neighboring Mn sites only if they are ferromagnetically aligned
- Zener double exchange alone can not explain the magnitude of the CMR effect
  - Millis *et al*., *PRL* **74**, 5144 (1995)

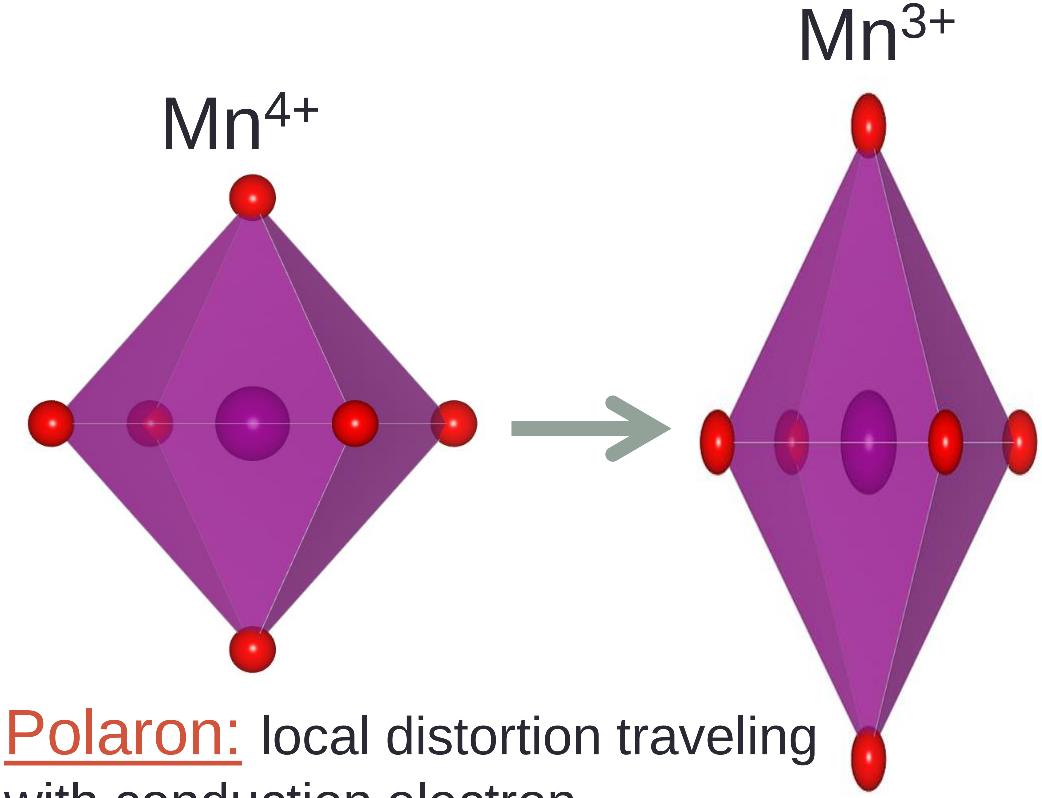
Coupling between lattice, charge, and magnetic degrees of freedom

- Zener double exchange alone can not explain the magnitude of the CMR effect
  - Electron-Phonon interaction
  - Large-scale phase coexistence



Dagotto, *Science* **309** 257 (2005)

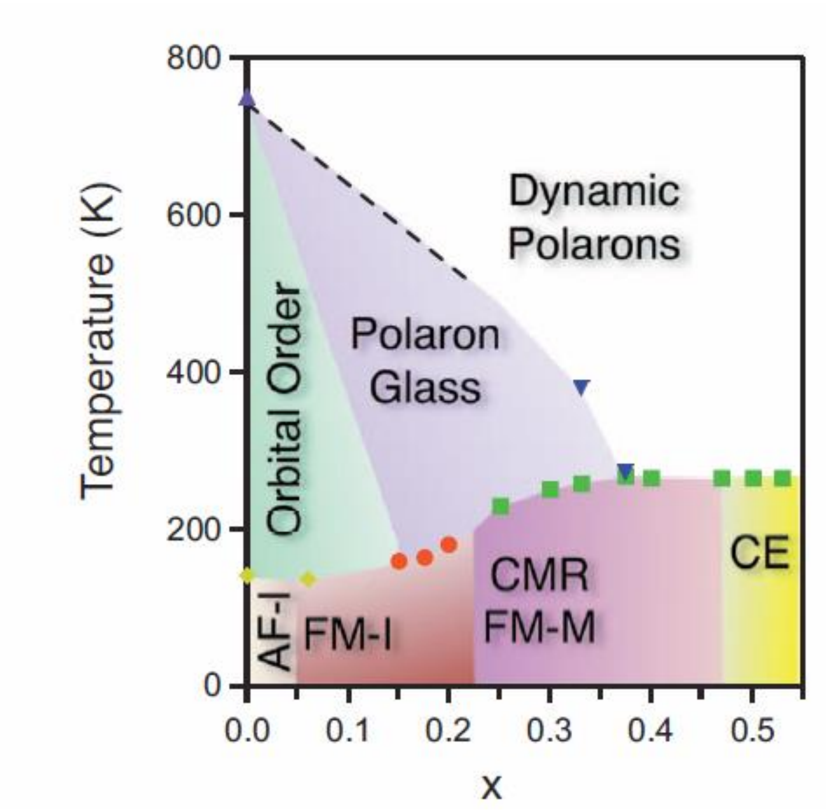
- charge ↔ lattice
  - Mn^{3+} is Jahn-Teller distorting



Polaron: local distortion traveling with conduction electron

Coupling between lattice, charge, and magnetic degrees of freedom

Phase diagram of $La_{1-x}Ca_xMnO_3$
J.W. Lynn, *et al.*, *PRB* **76**, 014437 (2007)



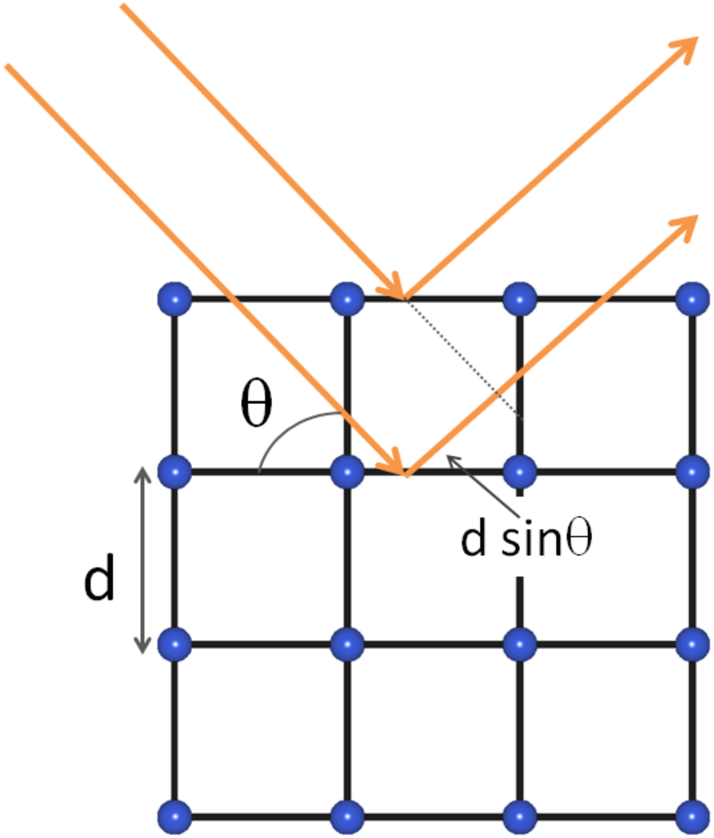
- Complex phase diagram driven

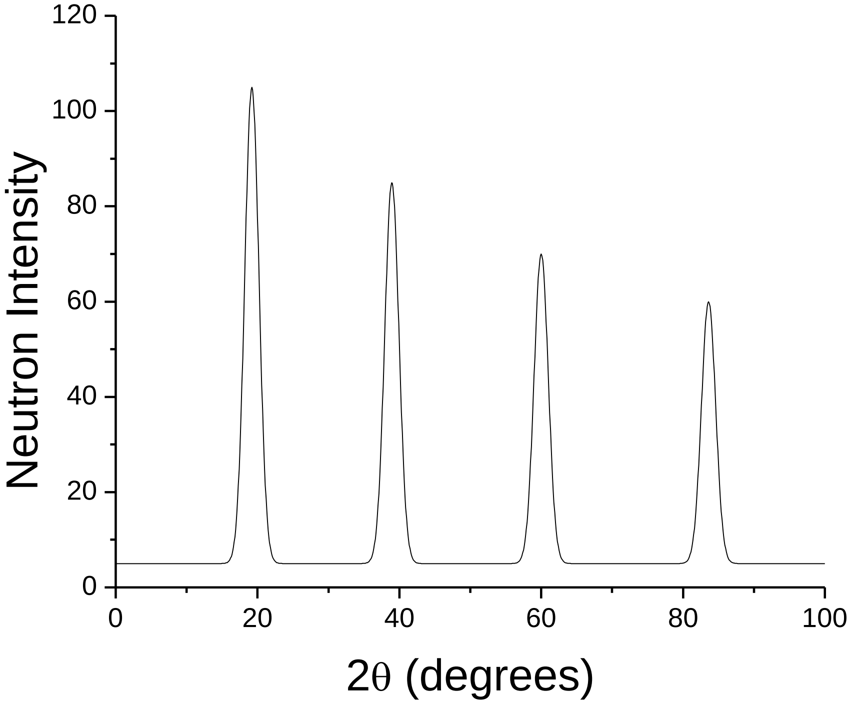
- Ideal for neutron scattering!
  - Structural (nuclear)
  - Magnetic
  - Static
    - Elastic Scattering / Diffraction
  - Dynamic
    - Inelastic Scattering / Spectroscopy
    - Well defined excitations as well as short-range correlations

Neutron Diffraction – Bragg’s Law

- Constructive interference when $2d \sin\theta = n\lambda$

$\lambda = h/p$



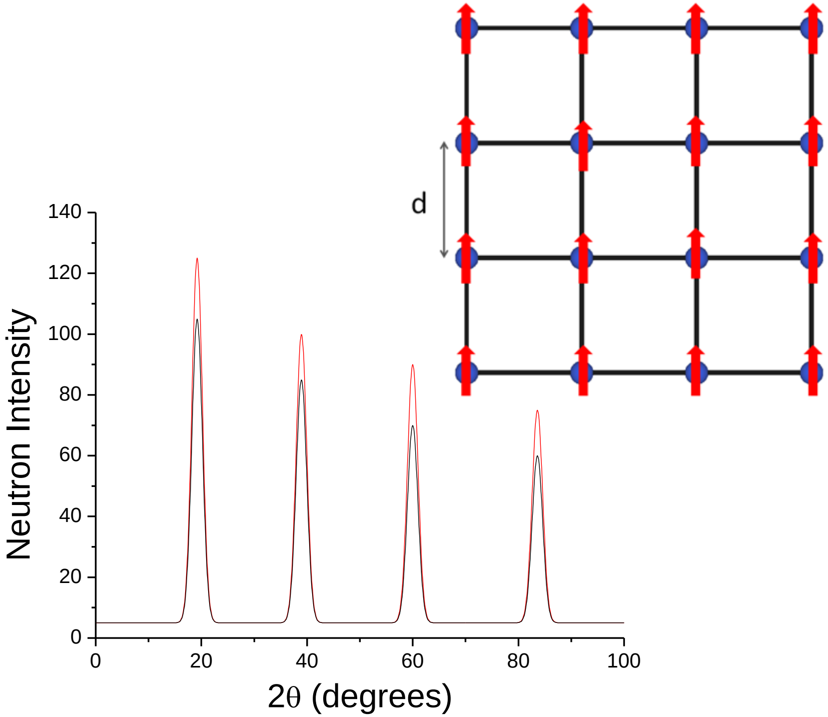


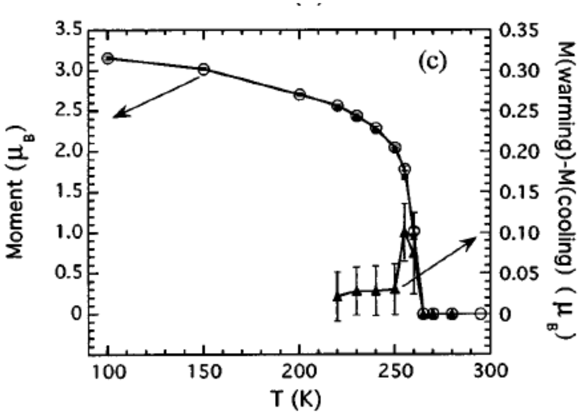
- “Where atoms are”

Ferromagnetic Order ($T < T_c$)

Neutrons have a magnetic moment

- Sensitive to ordered magnetic moments
- Ferromagnetic order
  - Magnetic peaks on top of nuclear peaks



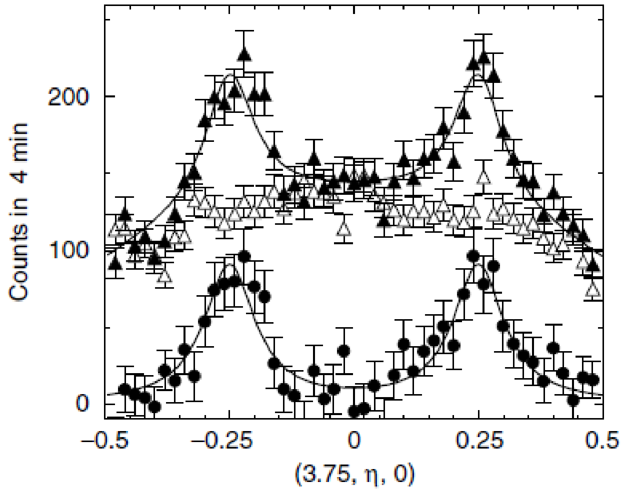


Q. Huang, *et al.*, *PRB* **58** 2684 (1998)

Ordered moment as a function of temperature

Static Polaron Correlations ($T > T_c$)

Peak at (¼ ¼ 0) positions

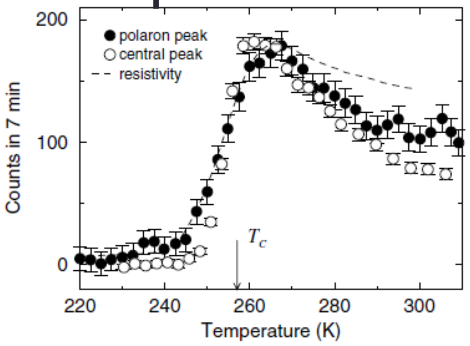


Polaron Peaks

C.P. Adams, *et al., PRL* **85** 3954 (2000)

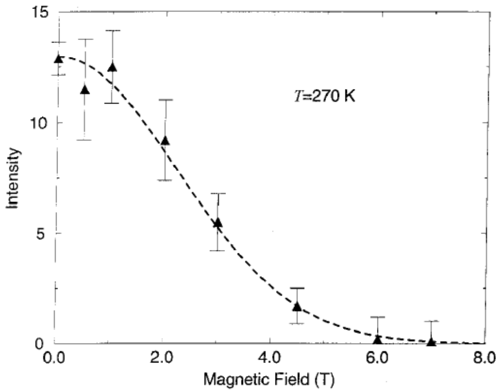
- Charge (Mn^{3+} vs. Mn^{4+}) and orbital order increases the unit cell by a factor of 4x4

- Temperature and field dependence



C.P. Adams, *et al., PRL* **85** 3954 (2000)

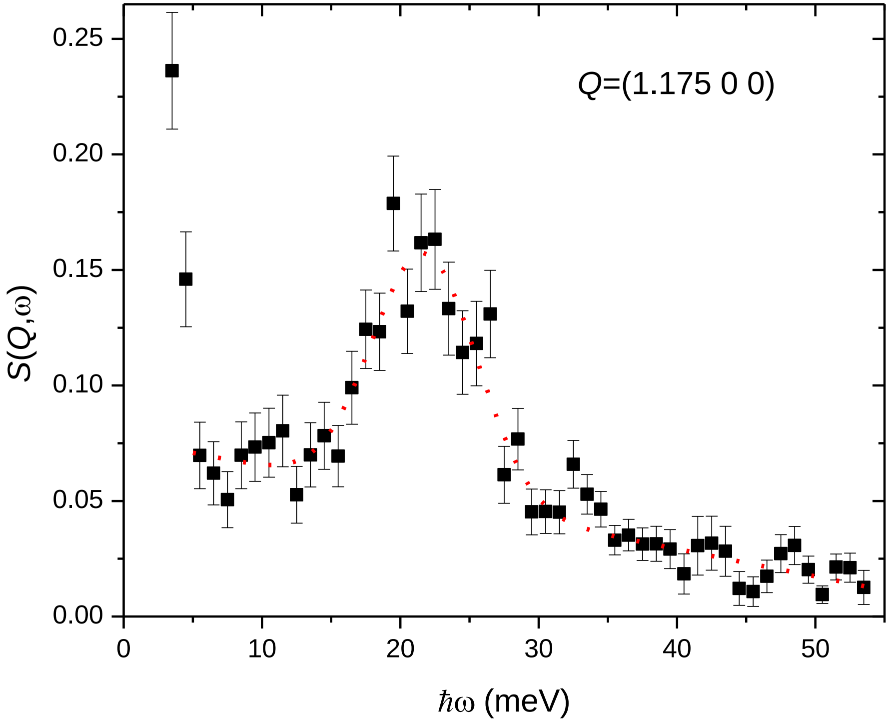
C.P. Adams, *et al., JAP* **89** 6846 (2001)

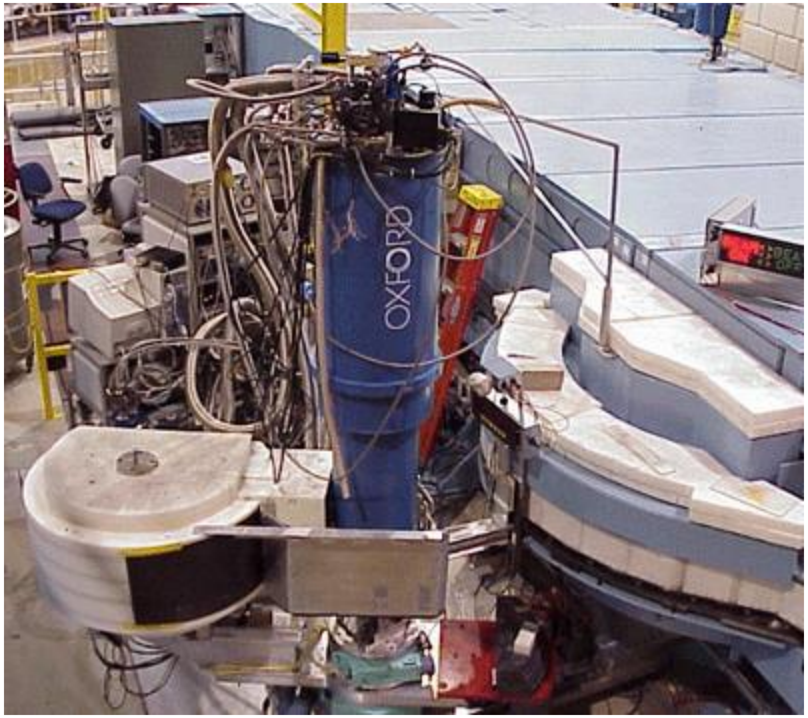


Polaron melting drives CMR?

Neutron Spectroscopy

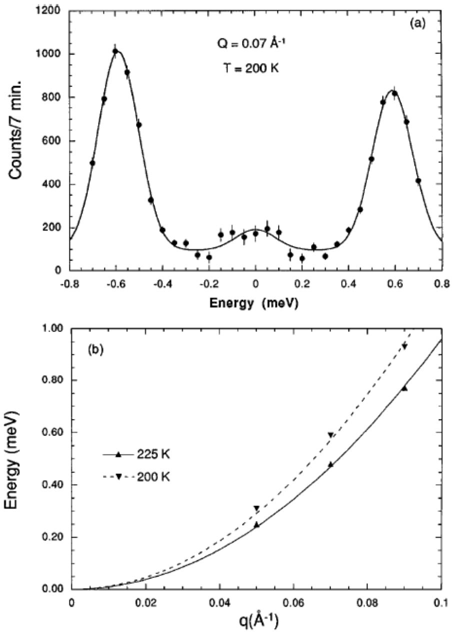
- “What atoms do”
  - Analyze the neutron incoming and outgoing energies





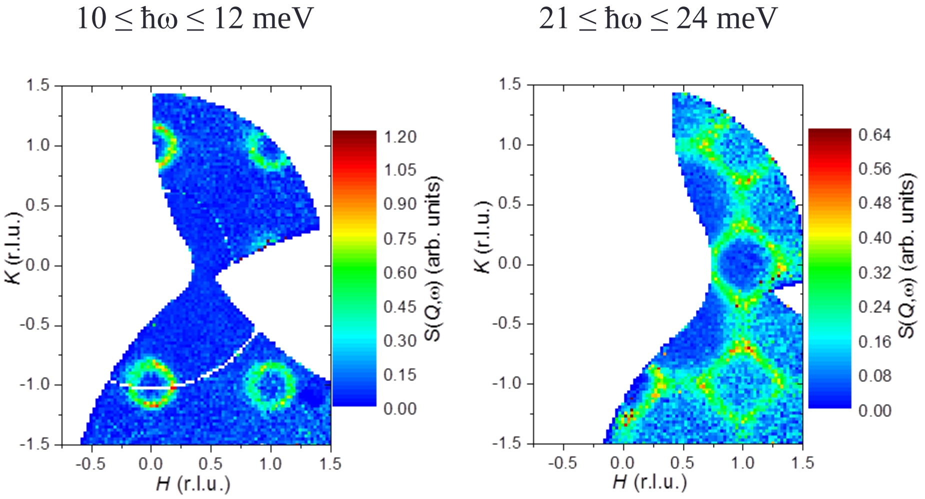
$$\textbf{Intensity} \propto \mathbf{S}^{\alpha\beta}(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\int dt\, e^{-i\omega t}\,\frac{1}{N}\sum_{\vec{R}\vec{R}'} e^{i\vec{Q}\cdot(\vec{R}-\vec{R}')} < S^{\alpha}_{\vec{R}}(t)\, S^{\beta}_{\vec{R}'}(0) >$$

Spin Waves ($T < T_c$)

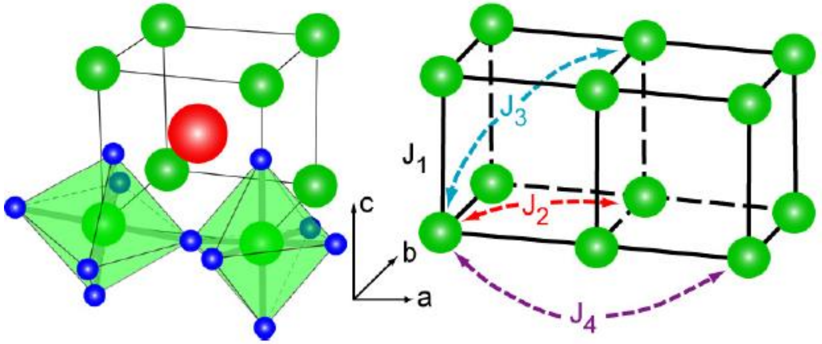


- Spin wave dispersion
  - Well defined excitations
  - ω varies with *q*
  - Gives information about exchange constants

$$\hat{H} = -\sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$



- Spin wave energy softens near zone boundary
  - 4th nearest neighbor exchange need to model dispersion

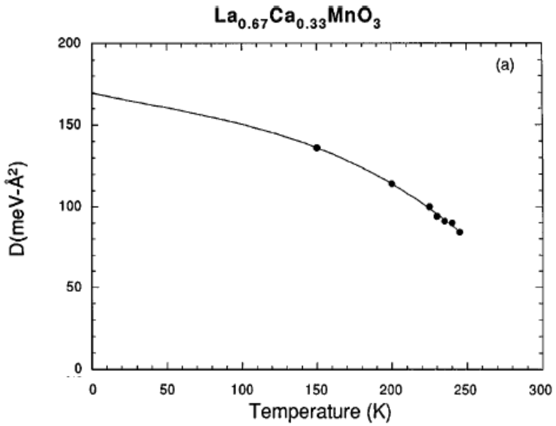


Zhang, *et al., JPCM* **19** 315204 (2007)

- *J* is very isotropic

Spin Waves ($T < T_c$)

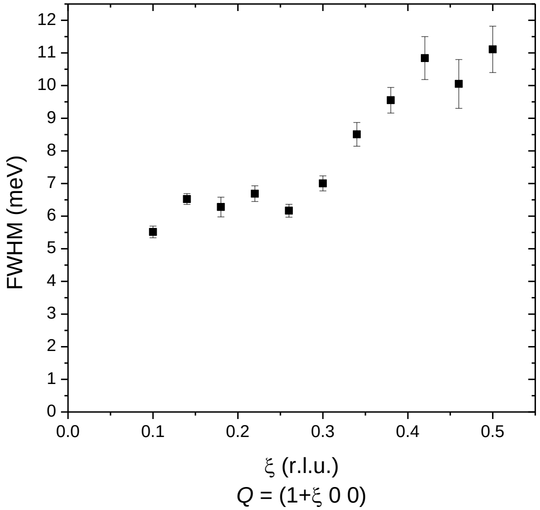
- Anomalous spin wave behavior



J.W. Lynn, *et al.*, *PRL* **76** 4046 (1996)

Spin wave stiffness vs. temperature

- Spin wave stiffness coefficient
  - Renormalizes at higher temperatures
  - But does not collapse at T_c like most ferromagnets

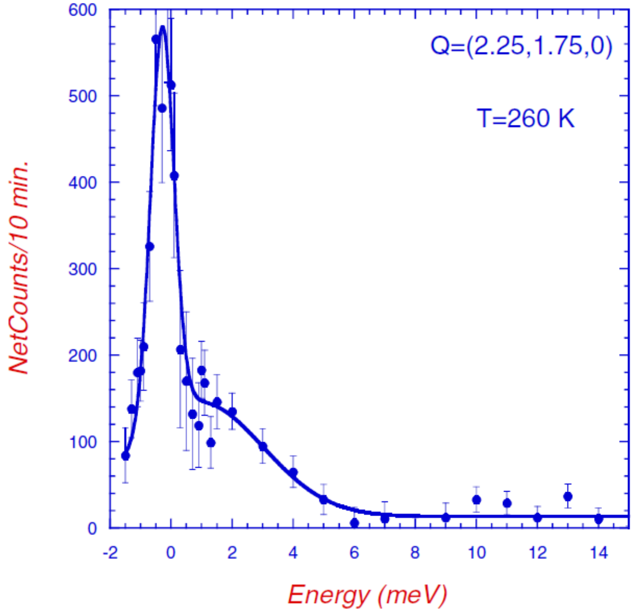


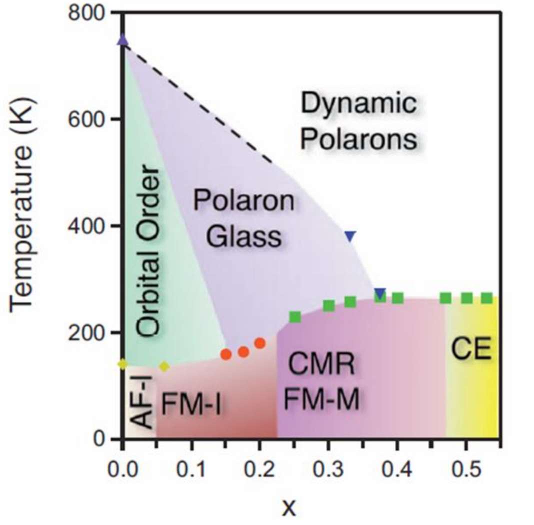
Spin wave broadening at higher energies near zone boundary

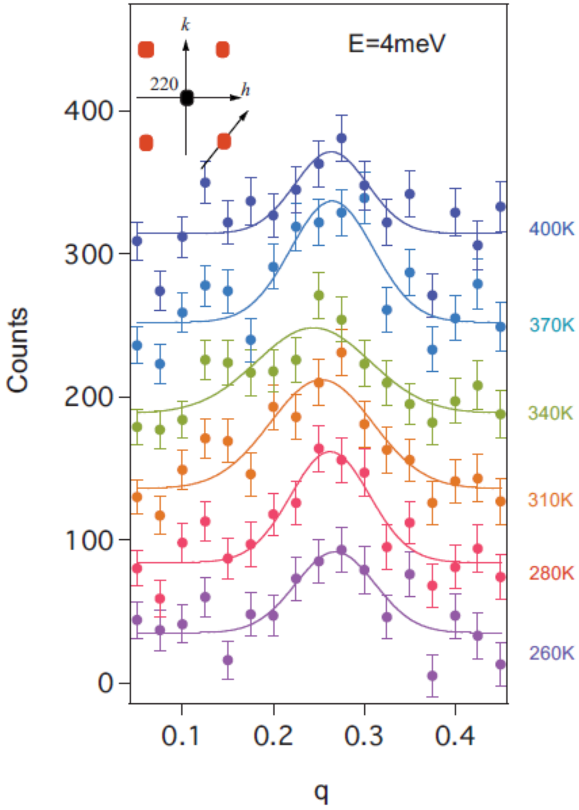
Dynamic Polaron Correlations ($T > T_c$)

- Broad features in neutron scattering experiments reveal short-range correlations

J.W. Lynn, *et al., PRB* **76** 014437 (2007)



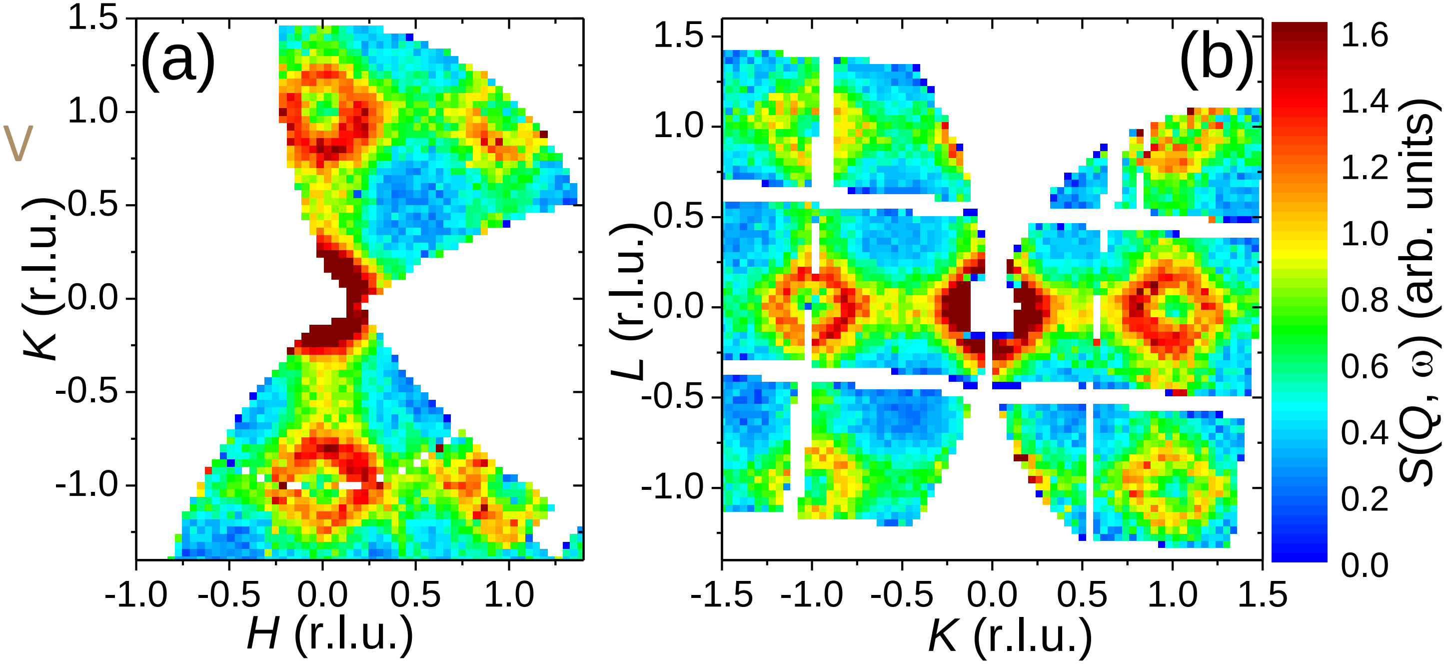




- Static and dynamic polarons observed from T_c up to 400 K
- Only dynamic polarons at higher temperatures

Paramagnetic correlations @ 265 K (1.03 T_c)

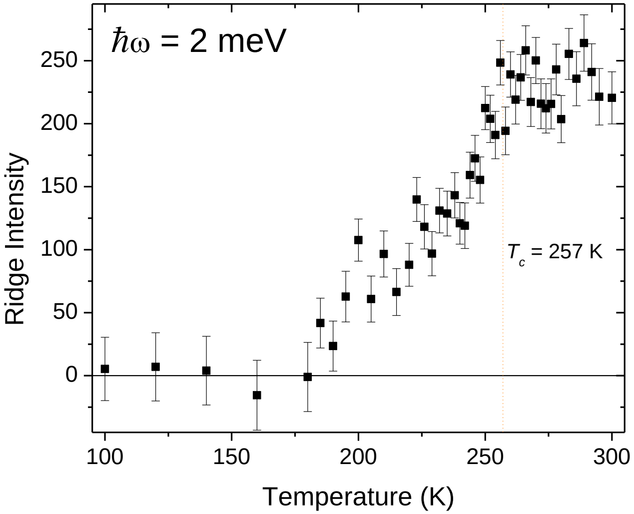
ARCS data
T = 265 K
3 meV ≤ ħω ≤ 5 meV

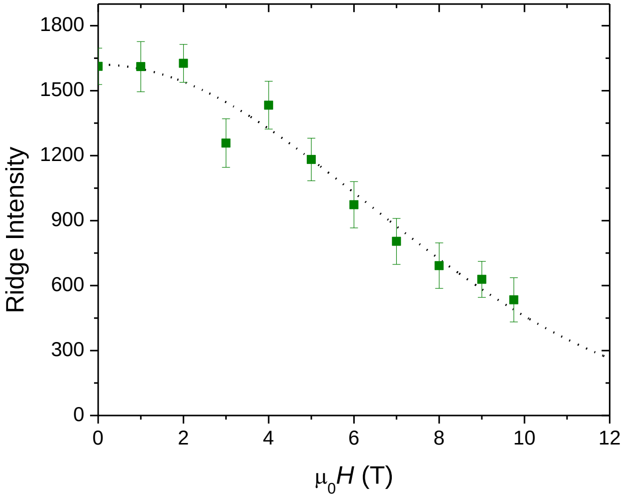


- Two components to the paramagnetic scattering
  - Rings surrounding Bragg positions
  - Ridges along (H 0 0) connecting the rings

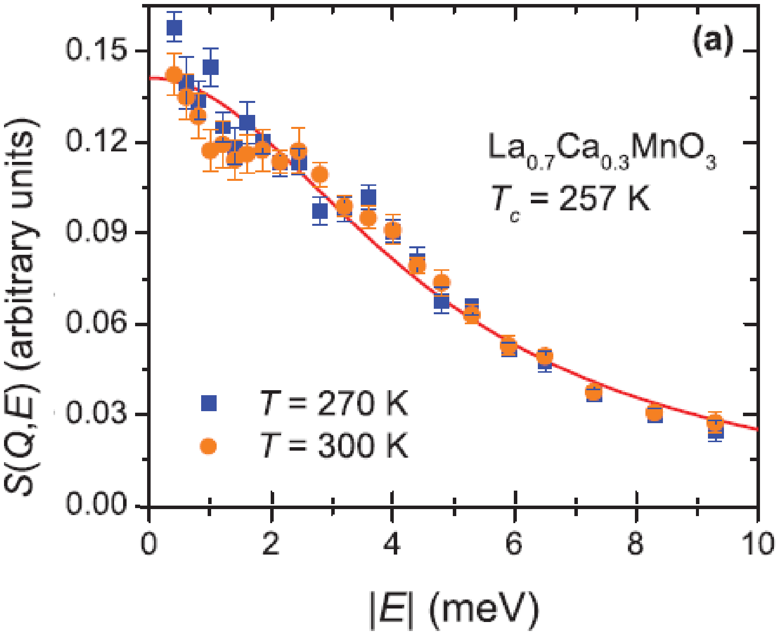
J.S. Helton, *et al., PRB* **85** 144401 (2012)

Paramagnetic ridges





Broad temperature and magnetic field dependence

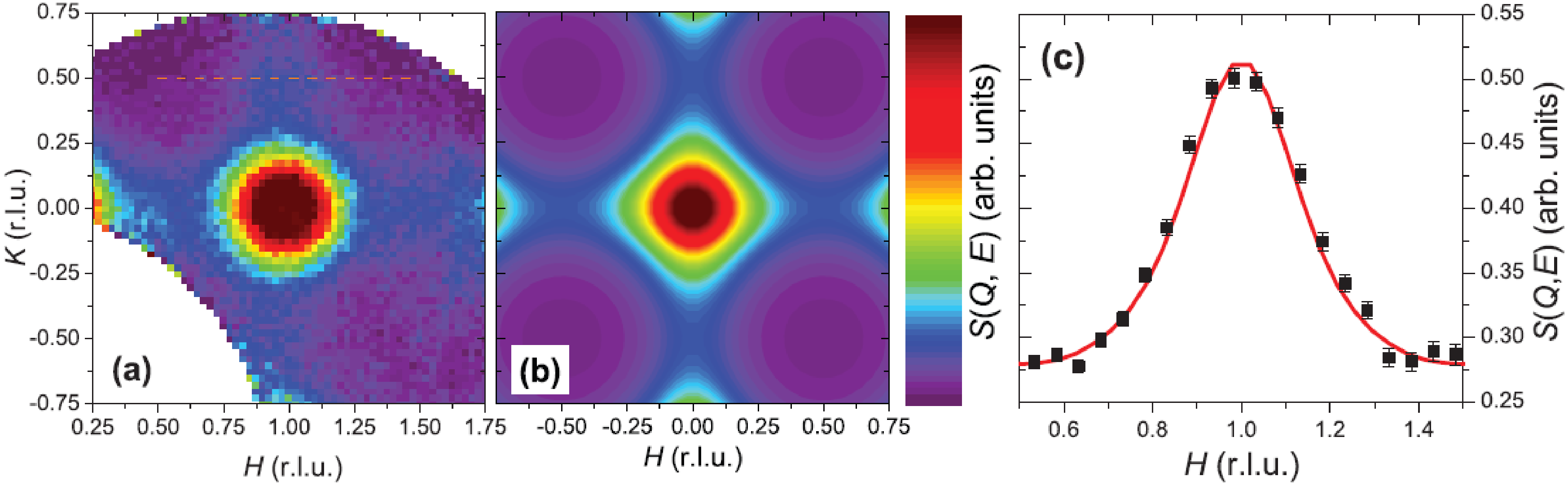


Quasielastic magnetic scattering

- Paramagnetic ridges are consistent with short-range ferromagnetic correlations dependent on number of nearest-neighbor ‘hops’ connecting two Mn ions

$$\langle S_i S_j \rangle \propto e^{-\frac{n}{\xi}} \qquad \xi = 0.7 \ \text{hops}$$

- Magnetic part of diffuse polarons
  - Conduction electron becomes self-trapped after small number of hops



J.S. Helton, *et al.*, *PRB* **90** 214411 (2014)

Conclusions

- The colossal magnetoresistance in $La_{1-x}Ca_xMnO_3$ and other materials stems in part from coupling between charge, magnetic, and lattice degrees of freedom
    - Ideal for study with neutron spectroscopy
- Below T_c
    - Long range ferromagnetic order with well-defined spin waves
    - Spin waves anomalously broaden and soften near zone edge
- Above T_c
    - Short range polaron correlations
        - Static and dynamic polarons at (¼ ¼ 0) positions
    - Magnetic ridges
        - Magnetic correlations – diffuse part of magnetic polarons
- Phase competition and coexistence at this transition may drive magnitude of the CMR effect