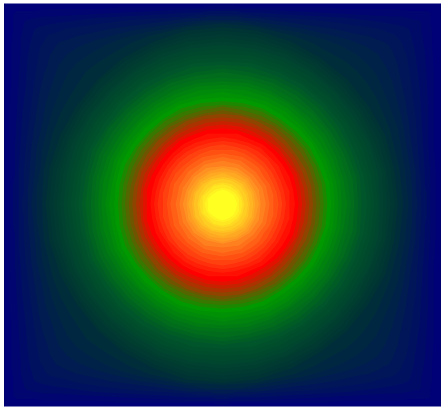 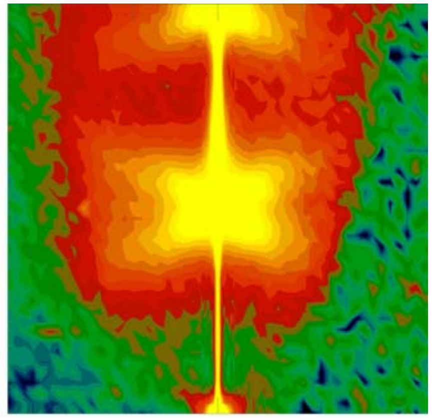 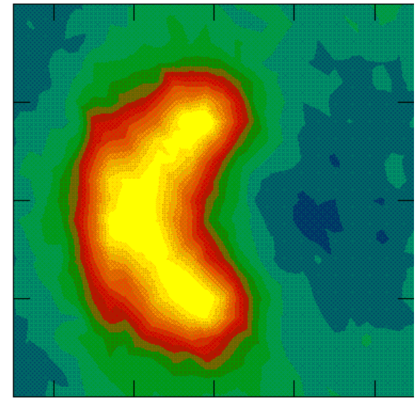

Competing Charge, Orbital, and Magnetic Order in Layered CMR Manganites

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Lida Vasiliu-Doloc (NIU/APS), Sunil Sinha (UCSD/APS), Jeffrey Lynn (NIST)

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Outline

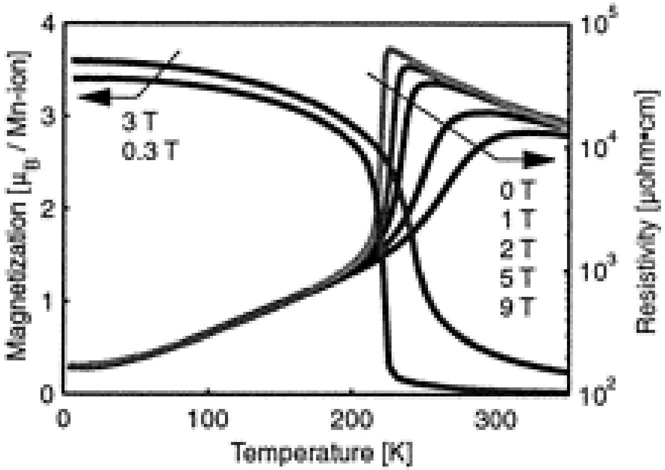
1. CMR in Bilayer Manganites
2. Magnetic Correlations
   - Conventional 2-D Ferromagnetism
3. Probing the structure of small polarons
   - Quantitative Analysis of Huang Scattering
4. Polaron Correlations
   - Structural Refinement of Orbital Stripes
   - Evidence for Competing Charge/Orbital Order

Measurements

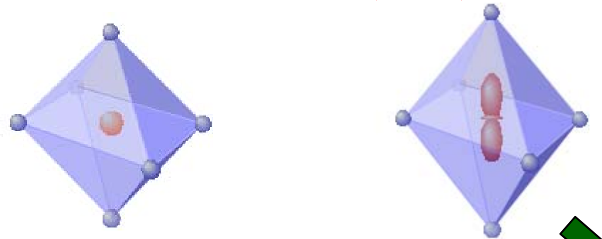
- Neutrons: NIST, IPNS, LANSCE
- X-rays: APS (SRI-CAT, BESSRC)

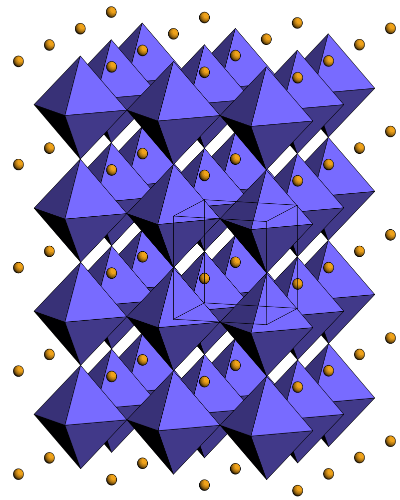
Colossal Magnetoresistance

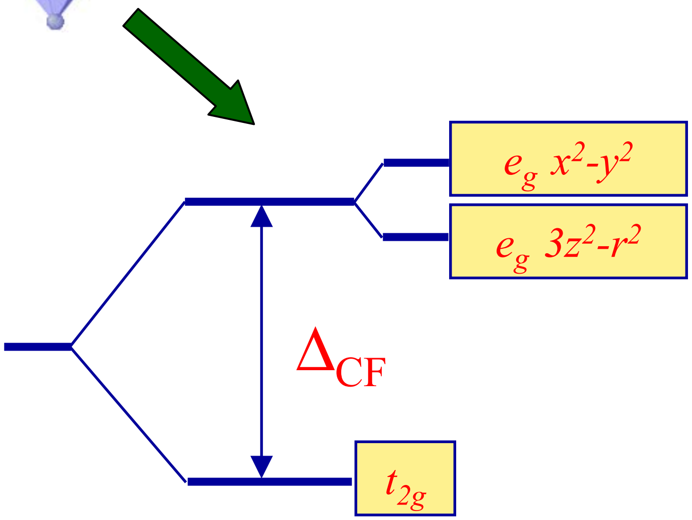
- The largest CMR effects are observed in mixed-valence manganites.
  - *e.g.* $La_{1-x}Ca_xMnO_3$, $La_{1-x}Sr_xMnO_3$
- The CMR is largest just above the ferromagnetic transition temperature



$x\,Mn^{4+} + (1-x)\,Mn^{3+}$



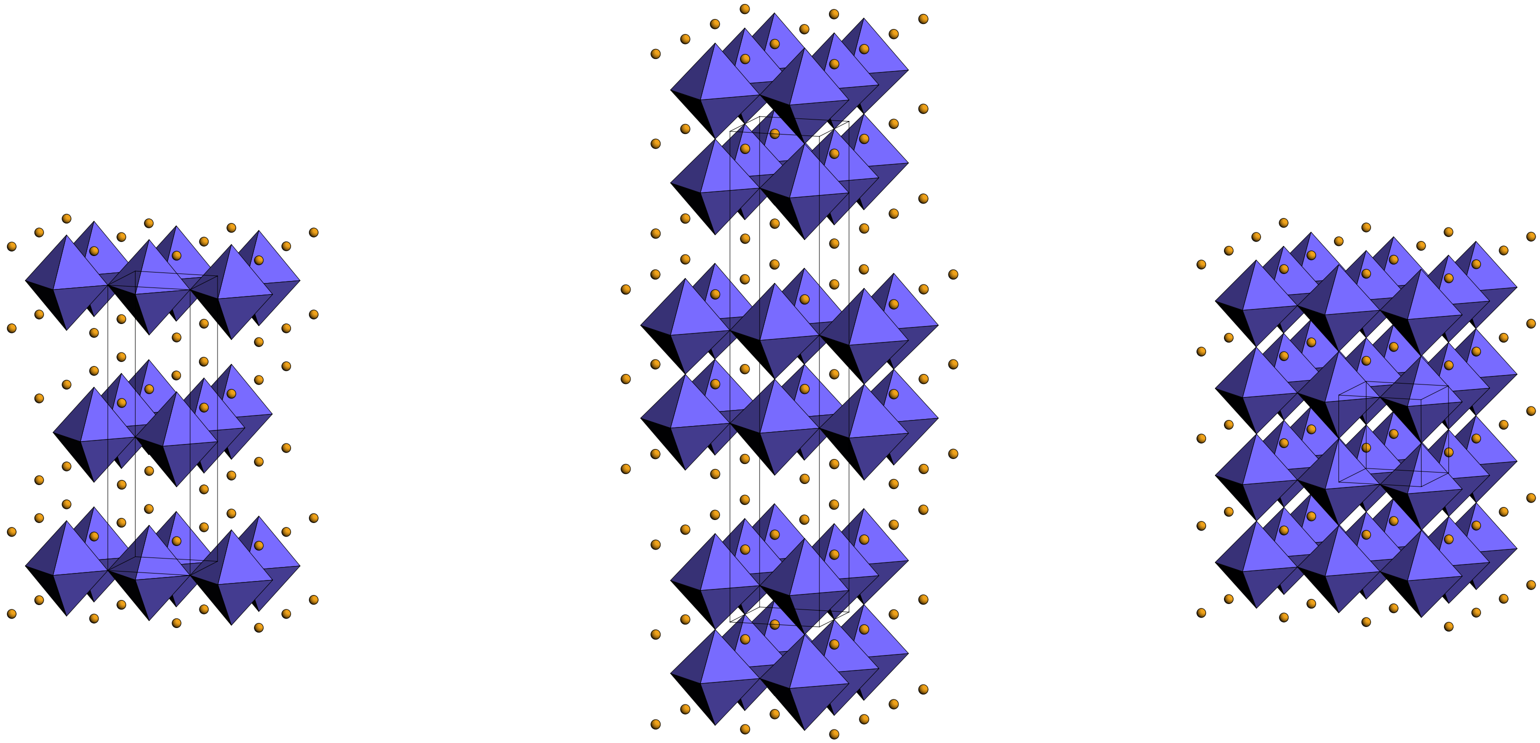




Millis, Littlewood and Shraiman [PRL 74, 5144 (1995)] showed that double exchange was not enough to explain CMR
i.e. Electrons are localized by local lattice distortions

Jahn-Teller Polarons

Naturally Layered Manganites

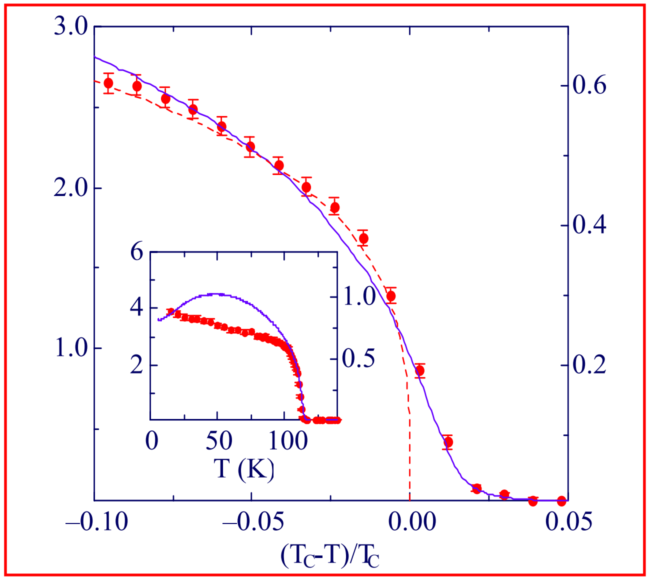


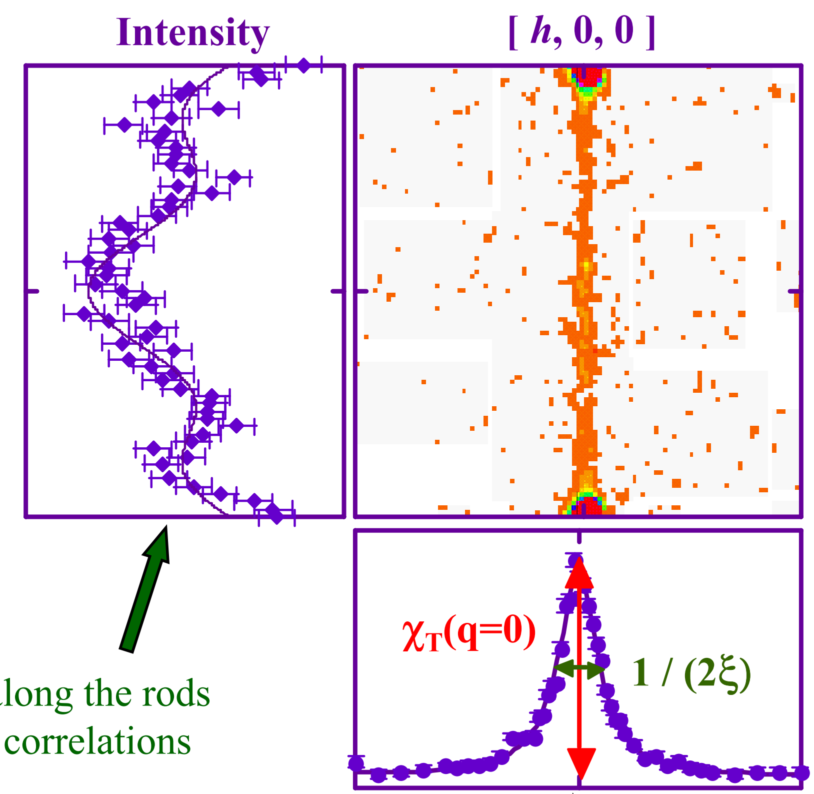
$n=1$ $n=2$ $n=\infty$

$SrO\ (La_{1-x}Sr_xMnO_3)_n$

Magnetic Correlations above T_C

x=0.4 Bilayer Manganite



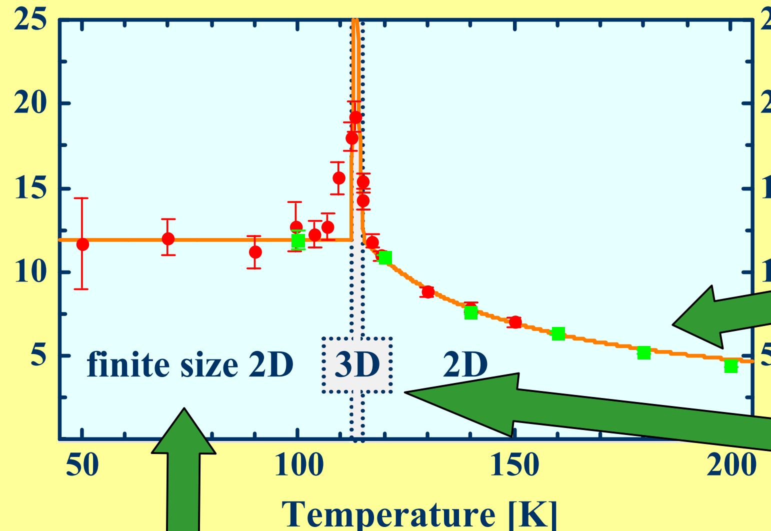


Intensity-variation along the rods
intra-bilayer spin correlations

Intensity-variation across the rods
in-plane spin correlations

R. Osborn, S. Rosenkranz, D. N. Argyriou, L. Vasiliu–Doloc, J. W. Lynn, S. K. Sinha, J. F. Mitchell, K. E. Gray, and S. D. Bader
Physical Review Letters **81**, 3964 (1998).

Two-Dimensional Correlation Length



$\xi/a \ll L_{eff} \sim \sqrt{J_1/J_3}$: 2D correlations

$$\xi/a = \xi_0 \, exp\,(b/\sqrt{t_{KT}}) \quad t_{KT} = \frac{T - T_{KT}}{T_{KT}}$$

observed : $\xi_0 = 0.3 \pm 0.1$, $b = 2.1 \pm 0.2$, $T_{KT} = 64 \pm 4$ K
theoretical : $\xi_0 \sim 1$, $b \sim 1.9$

$\xi/a \sim L_{eff}$: cross-over to 3D scaling

$$\xi/a = C_0^{+-} |t|^{-\nu} \qquad t = \frac{T - T_C}{T_C}$$

observed : $\xi_{co}/a \sim 3.2$
calculated : $= 11$

Renormalization Group Theory : $T_C = T_{KT} * (1 + b^2 / \ln^2(\xi_{co}))$
observed : $T_C = 113.2 \pm 0.1$ K
calculated : $T_C \sim 109$ K

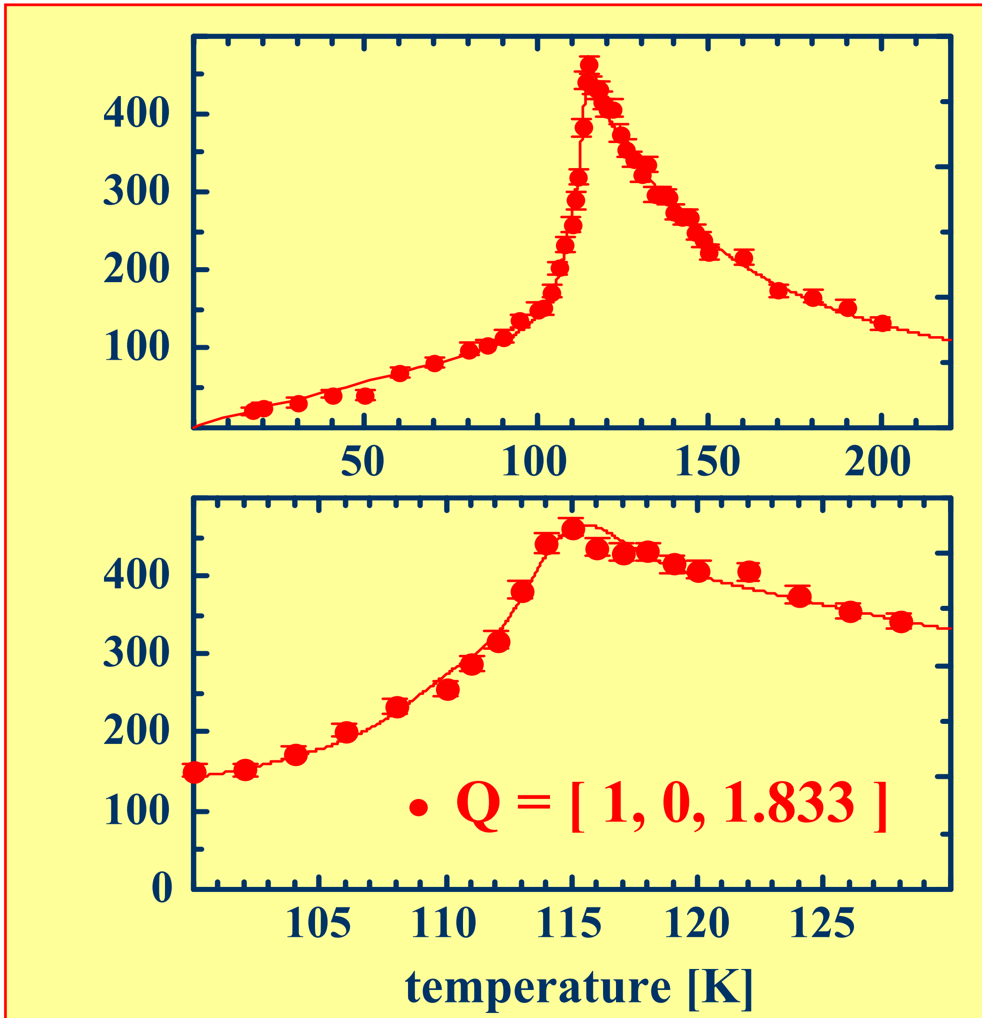
$T < T_C$: effective finite size 2D

$$\xi/a = \xi_{2D} \sim \sqrt{J_1/J_3}$$

observed : $\xi_{2D} \sim 3.2$
calculated : $= 11$

Quasi-2D single-layer Kosterlitz-Thouless model

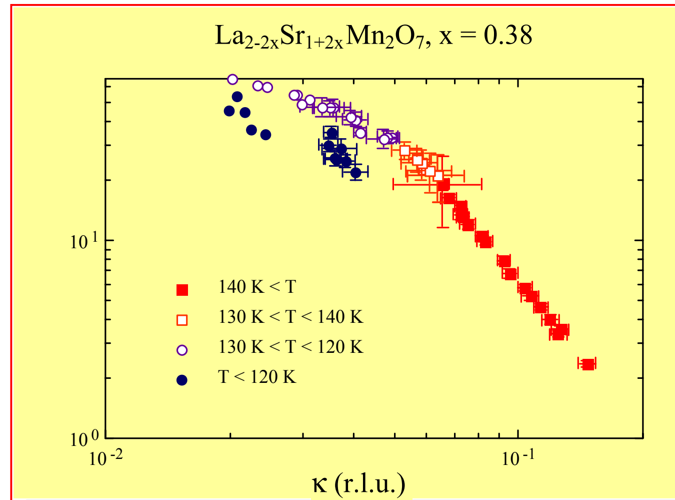
Two-Dimensional Magnetic Susceptibility



$T > T_C$: $\chi_T(0) = C \exp(B / \ t_{KT})$

observed: B = 3.9

theoretical: $B = b^*(2 - \eta) = 3.7$

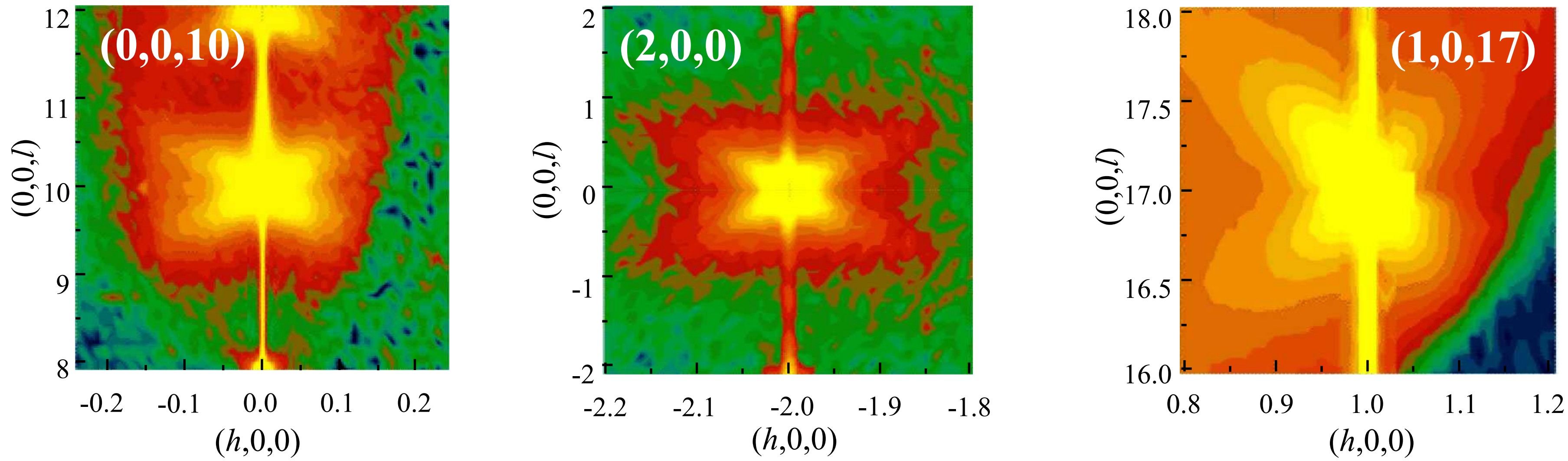


cf **K_2CuF_4, Rb_2CrCl_4**

K. Hirakawa *et al*, J. Phys. Soc. Jpn. **51**, 2151 (1982).
S. T. Bramwell *et al*, J. Phys. Soc. Jpn. **64**, 3066 (1995).

What about polarons?

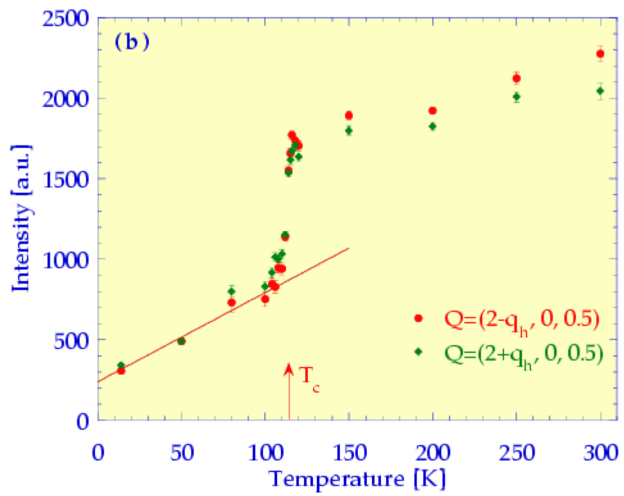
Diffuse Scattering from Polarons

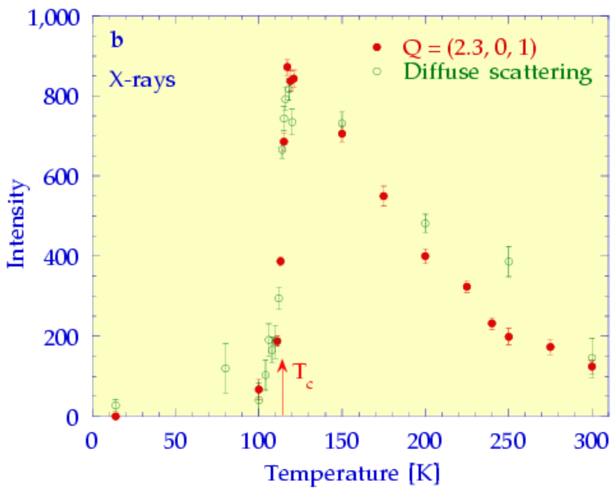


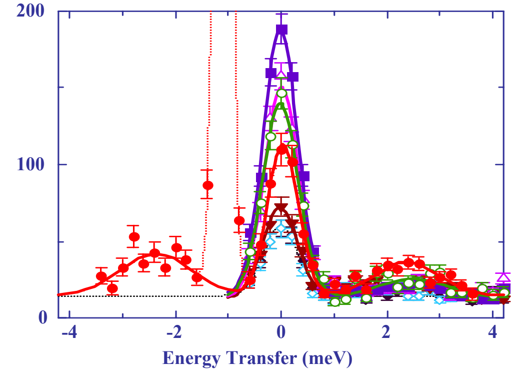
Jahn-Teller Distortion + Strain Field → **Huang Scattering**

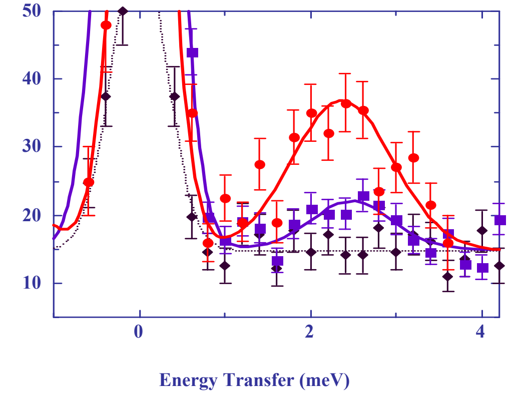
L. Vasiliu-Doloc, S. Rosenkranz, R. Osborn, S. K. Sinha, J. W. Lynn, J. Mesot, O. Seeck, and J. F. Mitchell, Physical Review Letters **83**, 4393 (1999).

Temperature Dependence of Diffuse Scattering









There is a dramatic increase in the intensity of diffuse scattering at T_C.

Inelastic neutron scattering shows that this increase is due to static diffuse scattering, ***not*** phonons (*i.e.* not Thermal Diffuse Scattering).

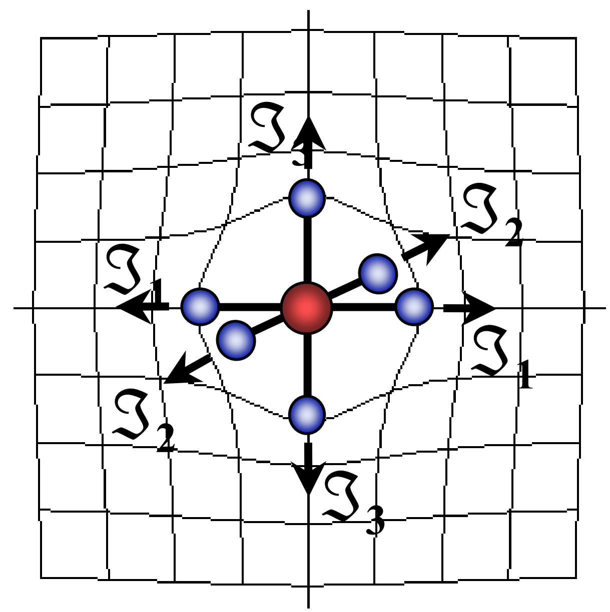
Modeling Huang Scattering

$$I(\mathbf{Q}) = \sum_{m,n} e^{i\mathbf{Q}\cdot(\mathbf{R}_m - \mathbf{R}_n)} f_m f_n e^{-W_m} e^{-W_n} \left\langle (\mathbf{Q}\cdot\mathbf{u}_m)(\mathbf{Q}\cdot\mathbf{u}_n) \right\rangle$$

$$I_{POL}(\mathbf{Q}) = N\left|F_{\mathbf{G}}\right|^2 \sum_{\alpha,\beta,\gamma,\delta} Q_\beta Q_\delta \left(\sum_{j,j'} \frac{\varepsilon_{\alpha,\mathbf{q},j}\, \varepsilon^*_{\beta,\mathbf{q},j}\, \varepsilon^*_{\gamma,\mathbf{q},j'}\, \varepsilon_{\delta,\mathbf{q},j'}}{\omega^2_{\mathbf{q},j}\, \omega^2_{\mathbf{q},j'}} \right) \sum_{m,n} \Im_{m,\alpha} \Im_{n,\gamma}\, e^{i\mathbf{q}\cdot(\mathbf{R}_m - \mathbf{R}_n)}$$

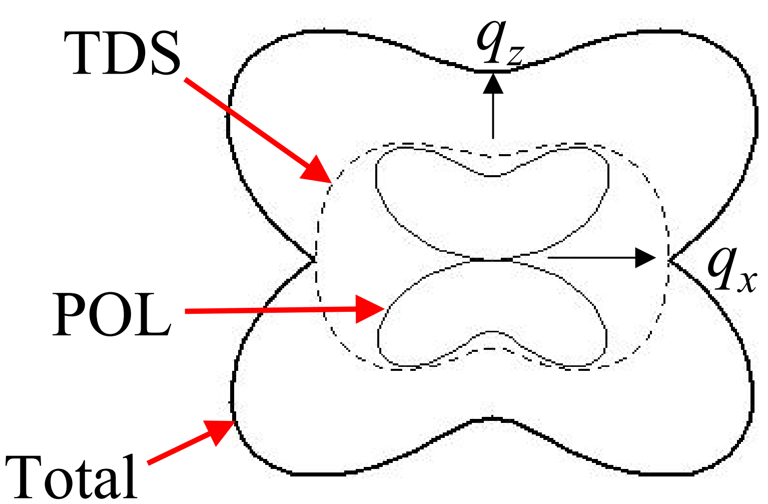
$$I_{TDS}(\mathbf{Q}) = N\left|F_{\mathbf{G}}\right|^2 \left(\frac{kT}{2M}\right) \sum_{\beta,\delta} Q_\beta Q_\delta \left(\sum_{j} \frac{\varepsilon^*_{\beta,\mathbf{q},j}\, \varepsilon_{\delta,\mathbf{q},j}}{\omega^2_{\mathbf{q},j}} \right)$$

$$u_{m,\delta} = \int \frac{d^3q}{\left(\frac{2\pi}{a}\right)^3} \sum_{\beta} \left(\sum_{j} \frac{\varepsilon^*_{\beta,\mathbf{q},j}\, \varepsilon_{\delta,\mathbf{q},j}}{\omega^2_{\mathbf{q},j}} \right) \sum_{n} \Im_{n,\beta}\, e^{i\mathbf{q}\cdot(\mathbf{R}_m - \mathbf{R}_n)}$$

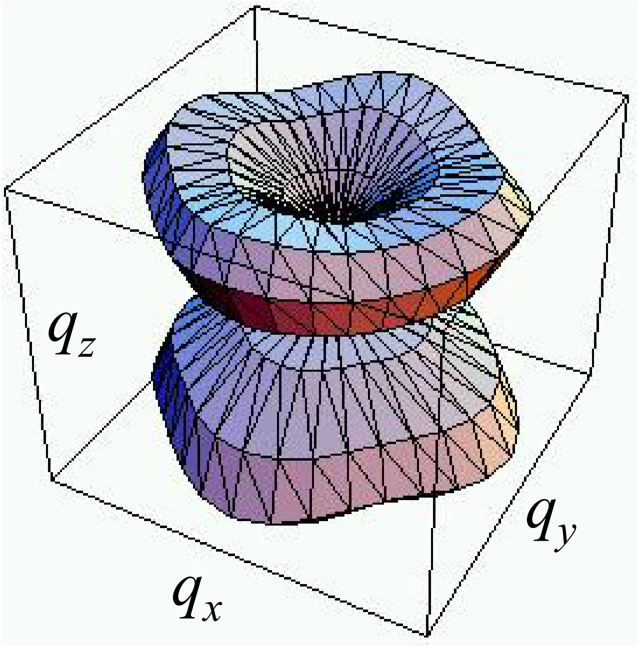


Polarons + TDS

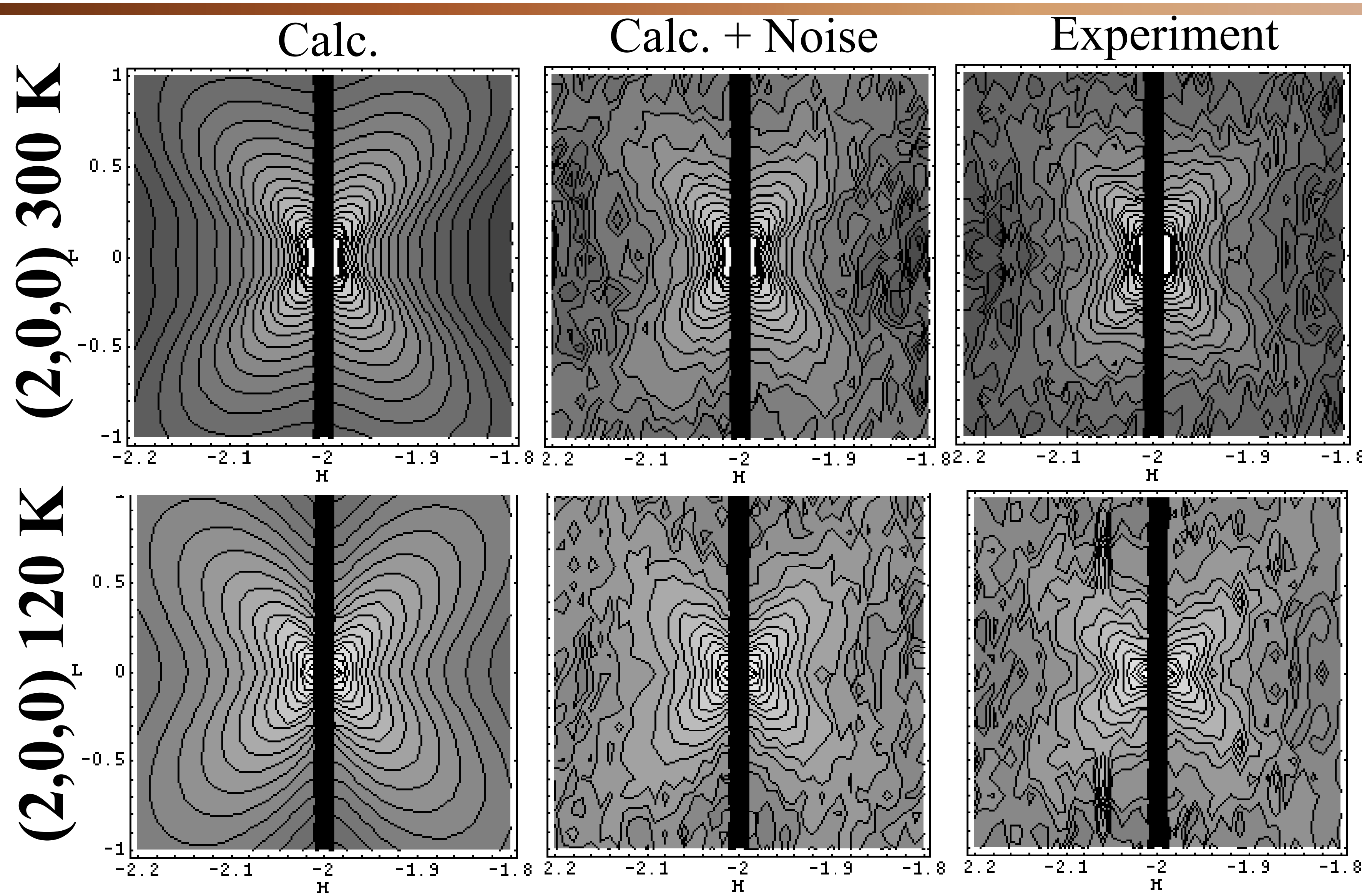
2D iso-intensity curve around (0, 0, 10) in the (q_x, 0, q_z) plane.



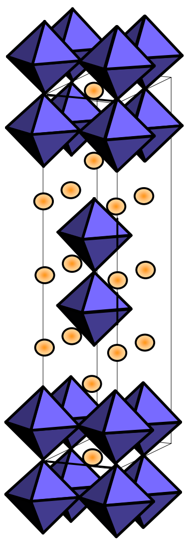
Polaronic 3D iso-intensity surface around (0, 0, 10).



Temperature Dependence

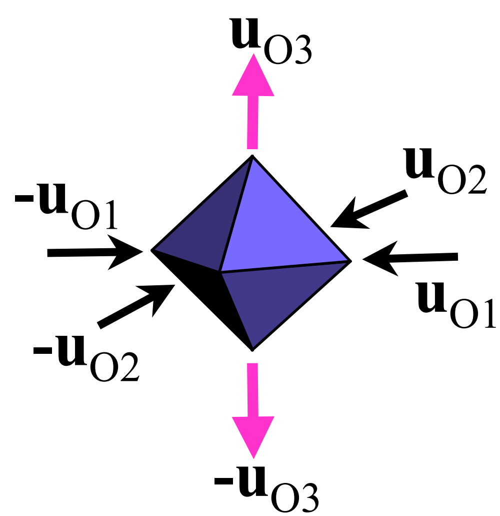


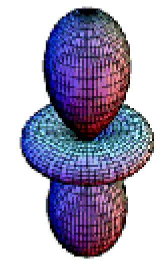
Orbital Polarization



300 K

$$\begin{pmatrix} u_{O1\_x} \\ u_{O2\_y} \\ u_{O3\_z} \end{pmatrix} = \begin{pmatrix} -0.012 \\ -0.012 \\ 0.074 \end{pmatrix} \text{Å}$$

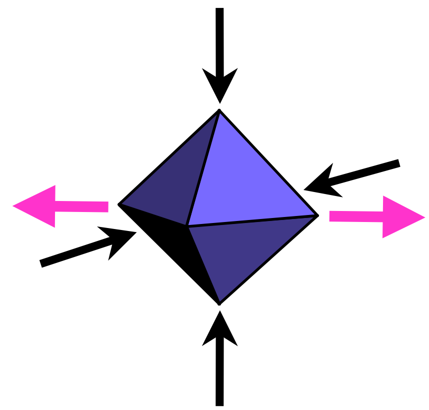


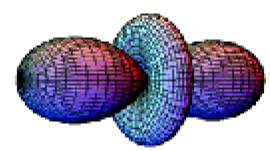


$d(3z^2-r^2)$

120 K

$$\begin{pmatrix} u_{O1\_x} \\ u_{O2\_y} \\ u_{O3\_z} \end{pmatrix} = \begin{pmatrix} 0.083 \\ -0.022 \\ -0.023 \end{pmatrix} \text{Å}$$

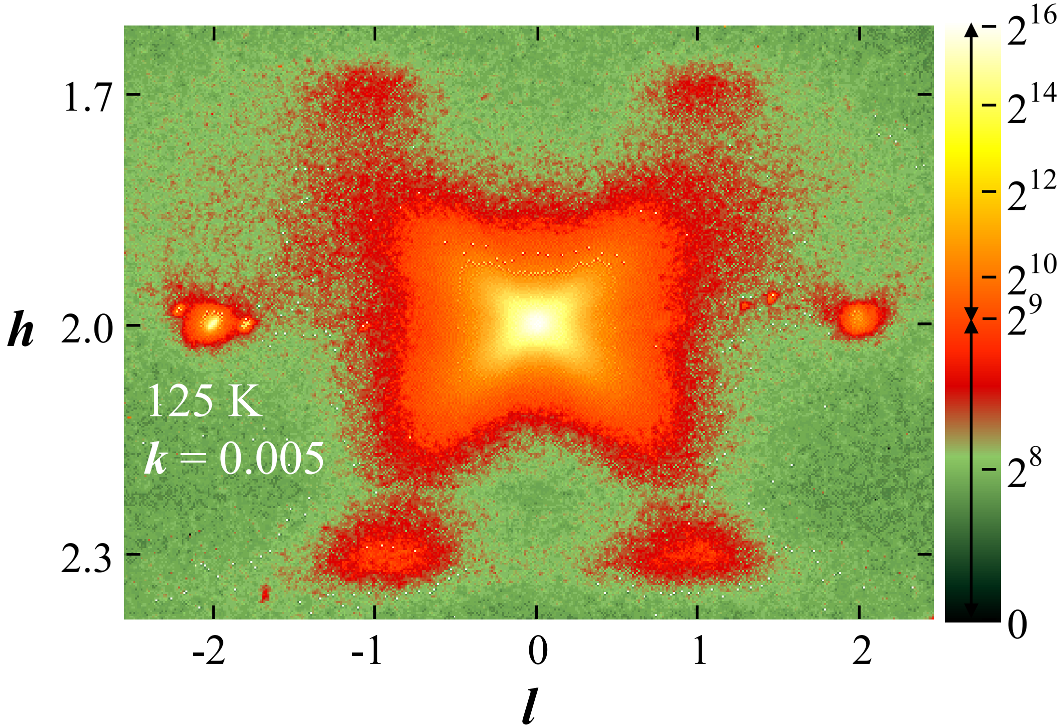




$d(3x^2-r^2)$

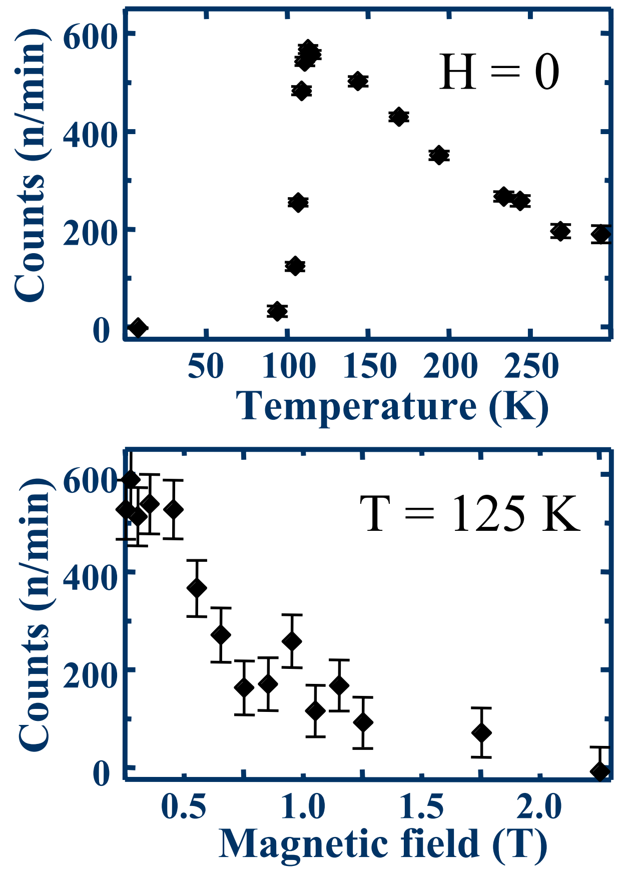
Evidence of Polaron Correlations

CCD x-ray data: 115 keV



BESSRC: 11 ID-C

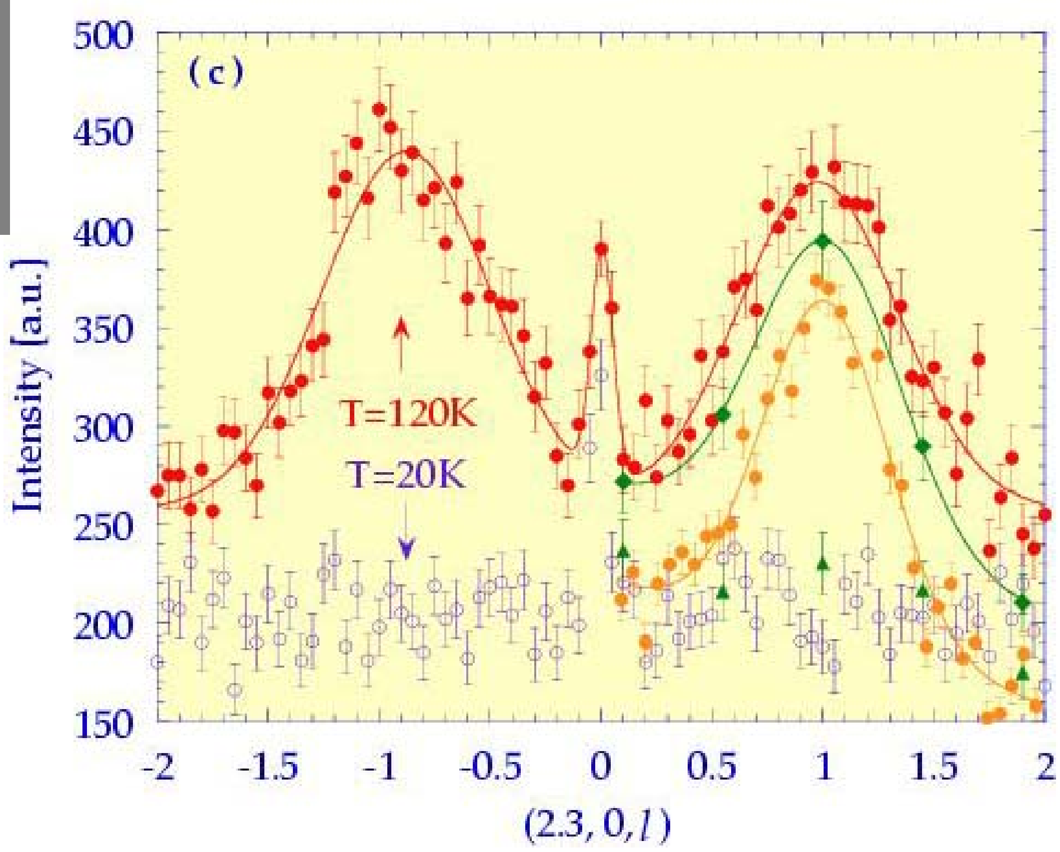
3-axis neutron data



Polarization Analysis of Polaron Correlations

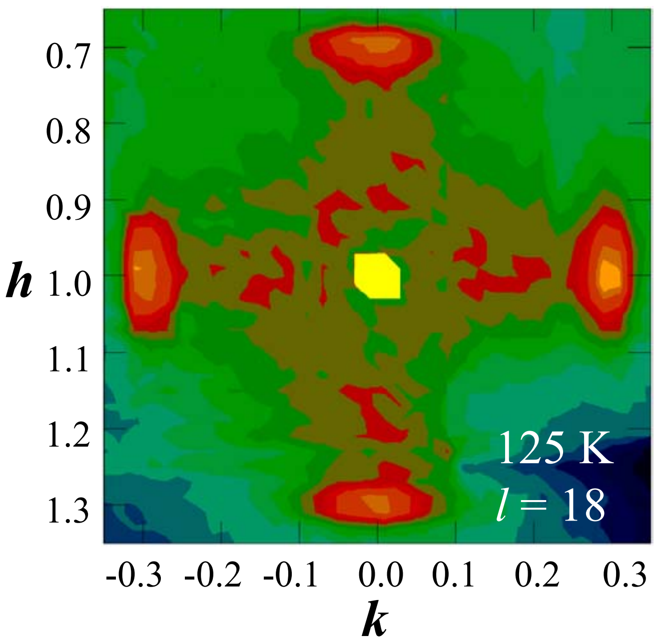
Neutron Polarization analysis shows that all the scattering is non-spin-flip

There is no magnetic component to the polaron correlations.

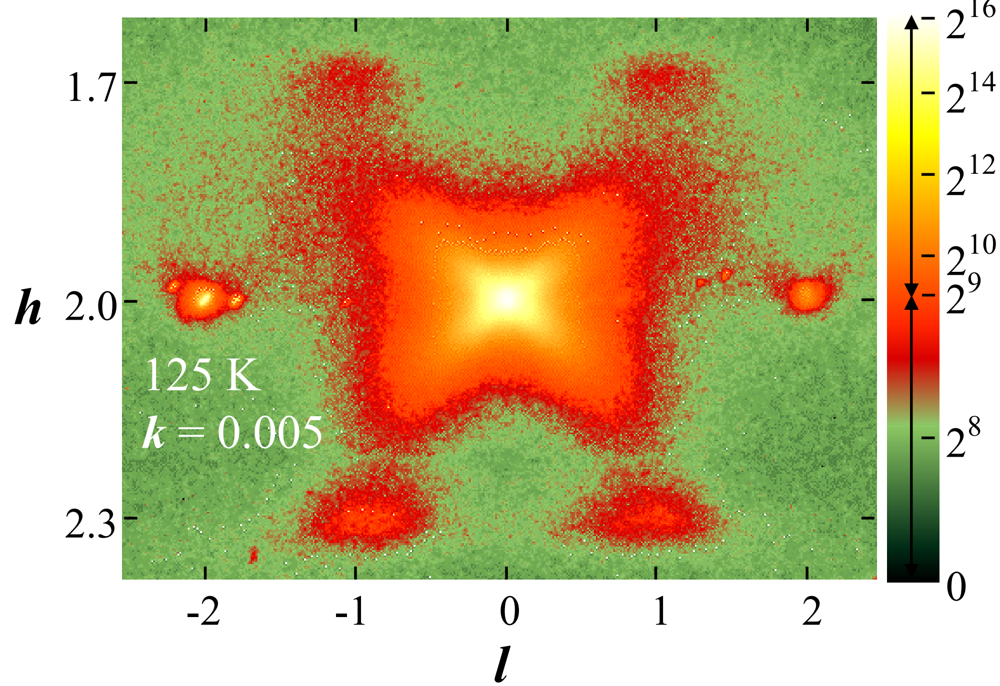


Modeling of Diffuse Satellites

x-ray SCD data (36 keV)



X-ray SCD data (115 keV)



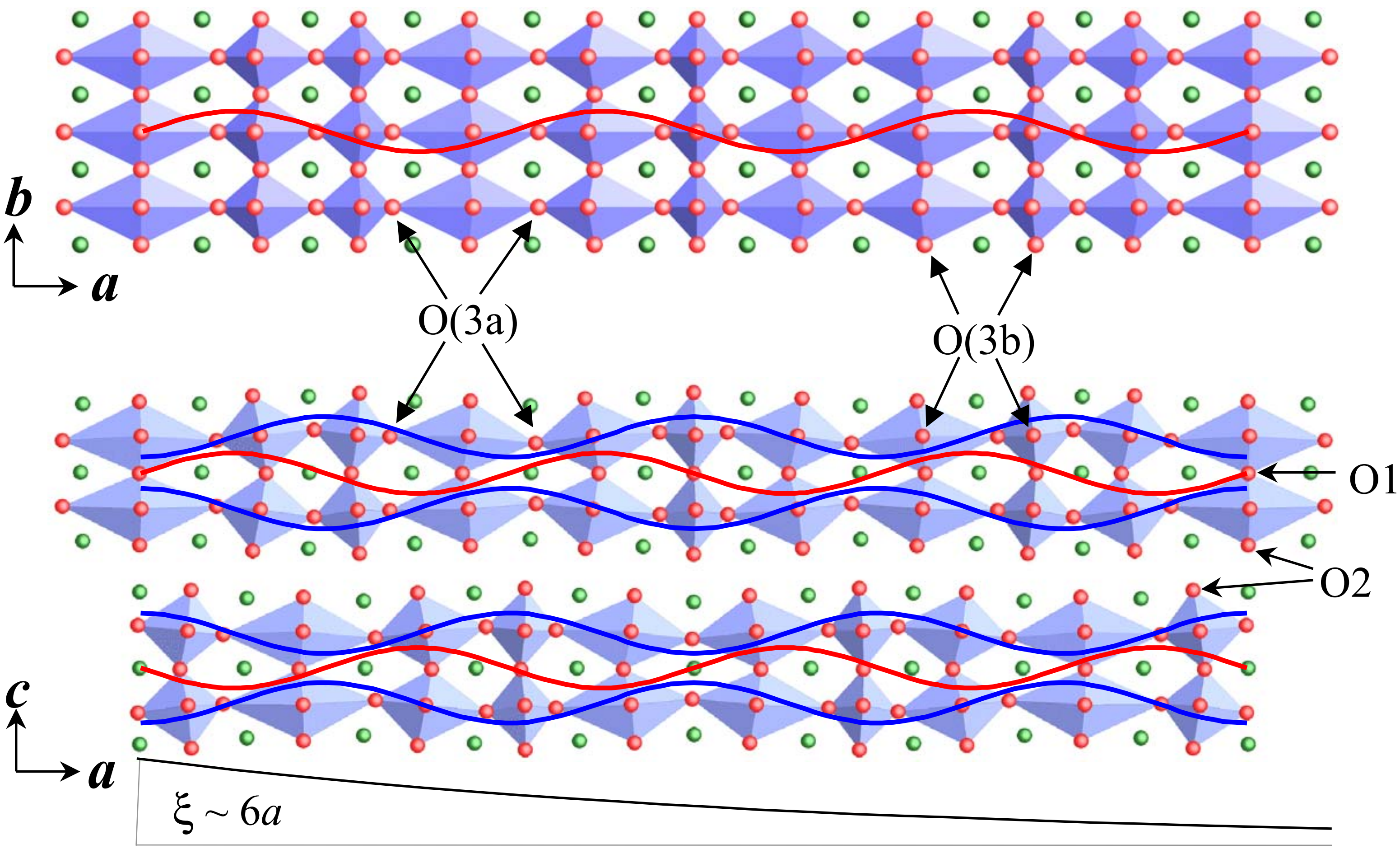
- 1D modulation along (1 0 0)
- $\mathbf{q}$ = (0.3 0 ±1) or (0.3 0 0)
- 109 unique 1st-order satellites

$$I(hklm) \propto |F|^2 = \left| \pi(\mathbf{G} + m\mathbf{q}) \cdot \sum_n \mathbf{u}_n f_n e^{i\mathbf{G}\cdot\mathbf{r}_n} \right|^2$$

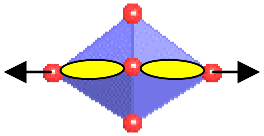
$$\mathbf{G} = (hkl) \qquad \mathbf{u}_n = \mathbf{u}_n^c + i\mathbf{u}_n^s$$

$$\Delta\mathbf{r}_n = \mathbf{u}_n^s \sin(\mathbf{q}\cdot\mathbf{r}_n) + \mathbf{u}_n^c \cos(\mathbf{q}\cdot\mathbf{r}_n)$$

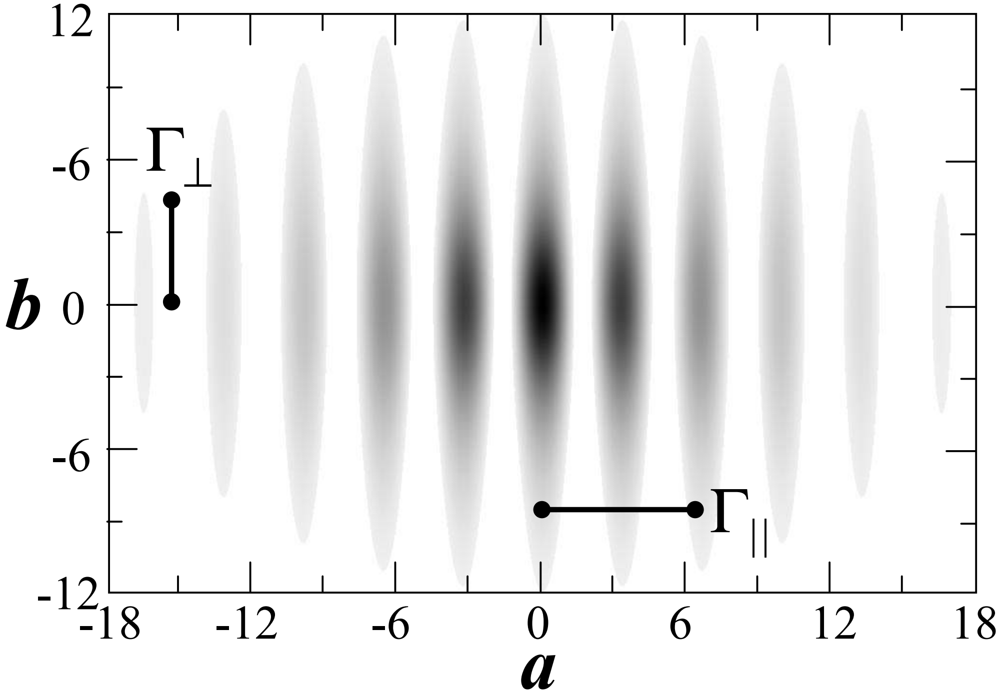
Cooperative Jahn-Teller Distortions



B. J. Campbell, R. Osborn, D. N. Argyriou, L. Vasiliu-Doloc, J. F. Mitchell, S. K. Sinha, U. Ruett, C. D. Ling, Z. Islam, and J. W. Lynn, Physical Review B **65**, 014427 (2001).



Correlated Polaronic Fluid



q ≈ **(0.3, 0, 0)**

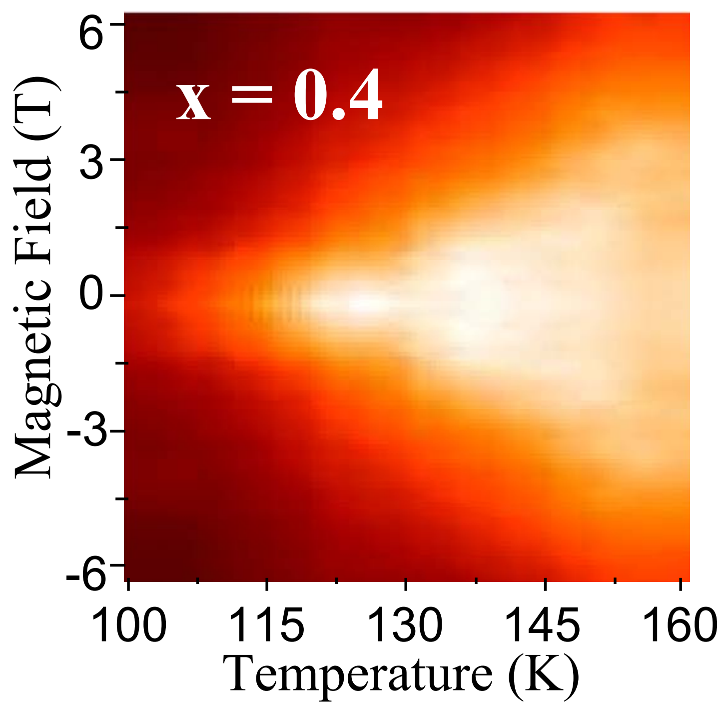
$\xi_x \approx 6\ a \approx 23$ Å

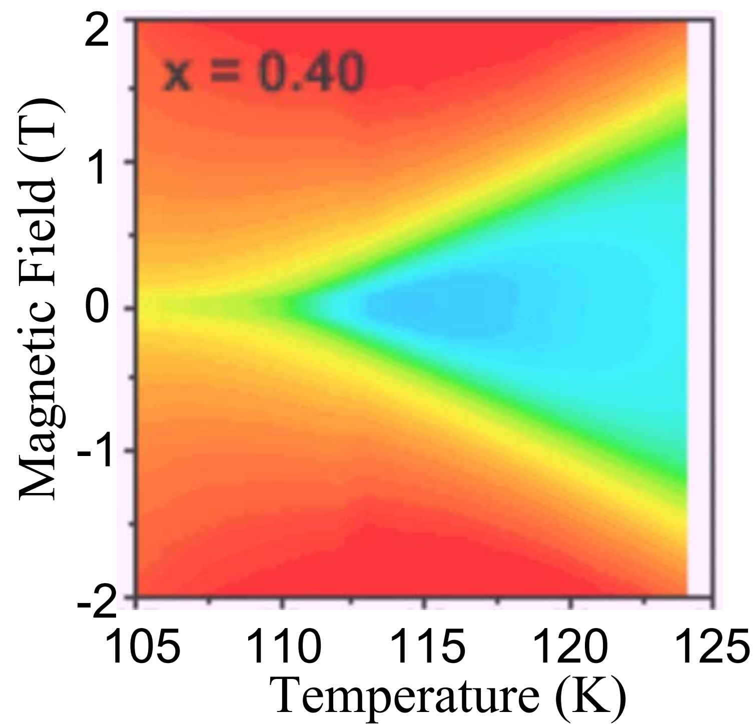
$\xi_y \approx 4\ a \approx 15$ Å

$\xi_z \approx c/2 \approx 10$ Å

displacement-displacement correlation function → probability amplitude

Polaron Melting in a Magnetic Field



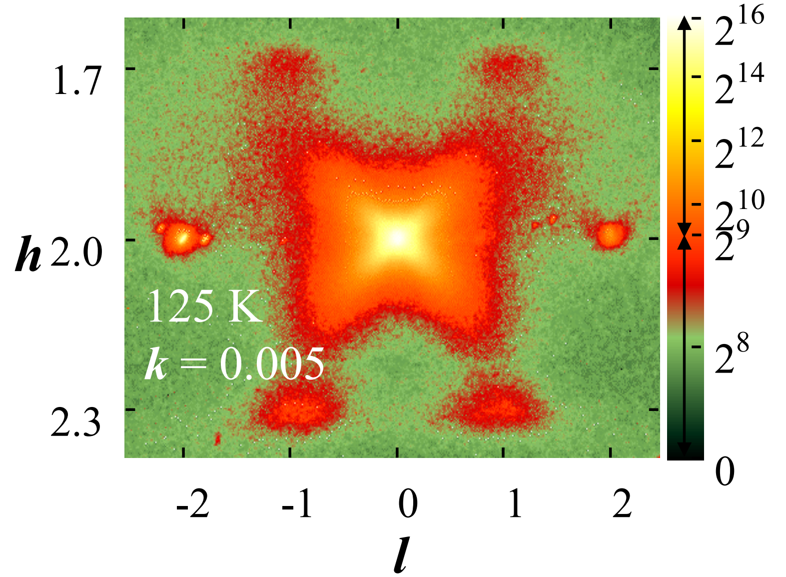


Diffuse satellite intensity at **Q** = (2.3,0,1). Triple-axis neutron scattering data.

Effective magnetic J-coupling from magnetization data.

Summary

- By combining neutron and x-ray scattering, we have established the existence of short-range charge/orbital ordering in the paramagnetic phase of layered CMR manganites.
- This has now been seen in CMR perovskites and is recognized as an important component in optimally-doped CMR compounds.
  - **Shimomura *et al* (PRL 1999)**
  - **Adams *et al* (PRL 2000), Dai *et al* (PRL 2000)**
  - **Kiriyukin *et al* (PRB 2002)**



- The systems respond to frustration induced by competing interactions by forming nanoscale defect structures.
- Transition metal oxides provide a valuable laboratory for tuning the balance of these interactions.
- New pulsed neutron source developments promise to enhance our ability to probe nanoscale phenomena.