Neutron scattering on magnetic materials

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- Revisit an old system, $LaCoO_3$, in order to understand magnetic properties.
- Find new magnetic structure formation resulting from the incompatible symmetries of two competing magnetic phases.
- Result: the spin incommensurability and two phase competition effectively reduce the number of free charge carriers.
- Neutron experiments carried out at SPINS of the NCNR.

List of collaborators:

Danny Phelan (UVa) – part of his thesis

Kazuya Kamazawa, Mike Hundley – on the transport measurements

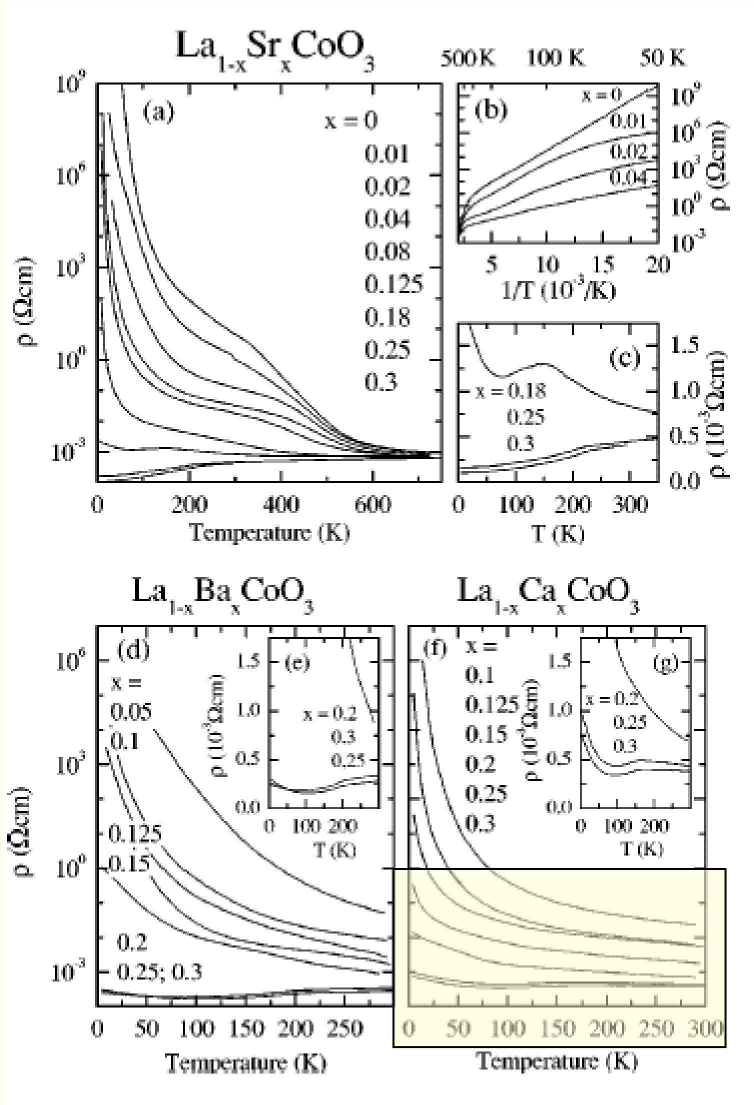
S.-H. Lee, S. Rosenkranz – on the single crystal work

Y. Qiu, J. Copley – on the inelastic powder measurements

J. F. Mitchell, J. L. Sarrao, Y. Moritomo and K. Yamada – on the sample growth

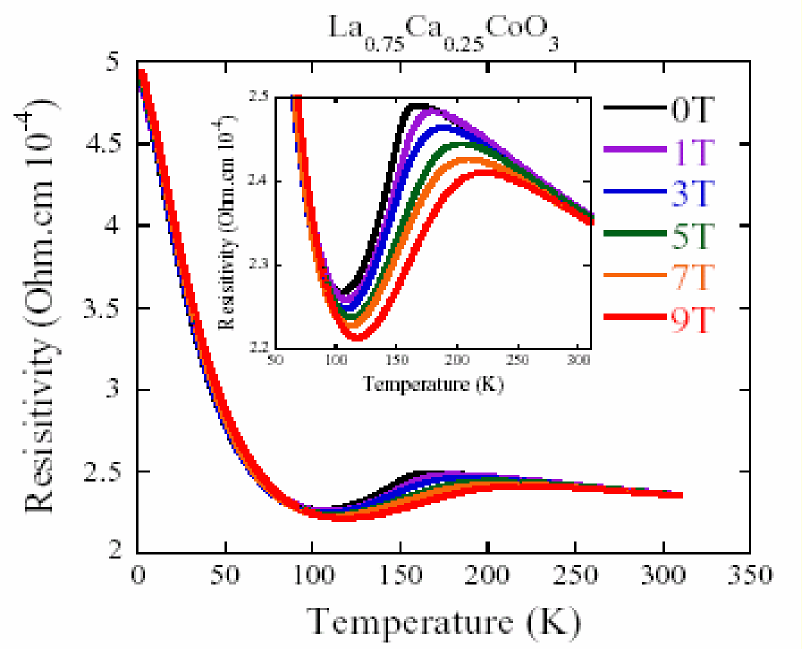
Y. Motome

Is there anything unusual about the transport properties?

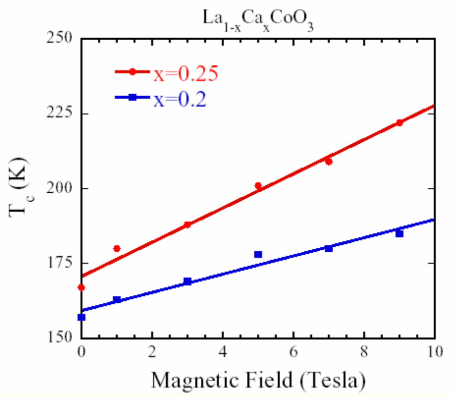


Kriener et al., PRB 69, 094417 (2004)

Is there anything unusual about the transport properties?

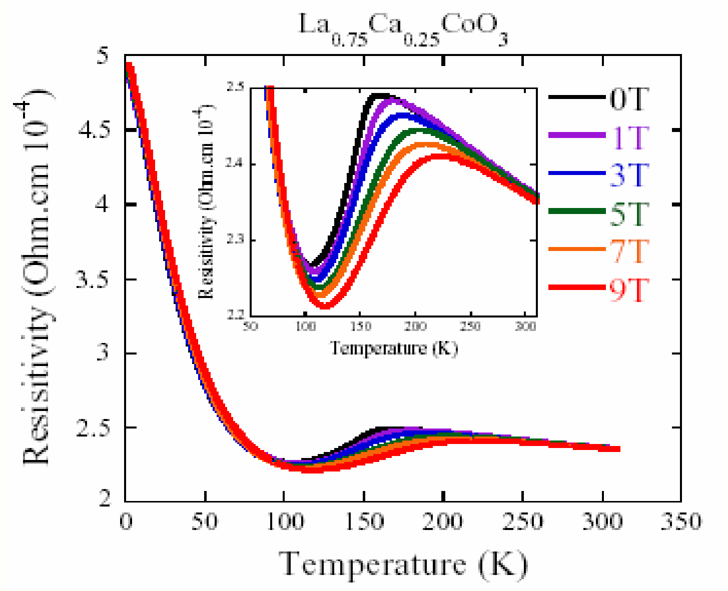


Residual resistivity below 100 K



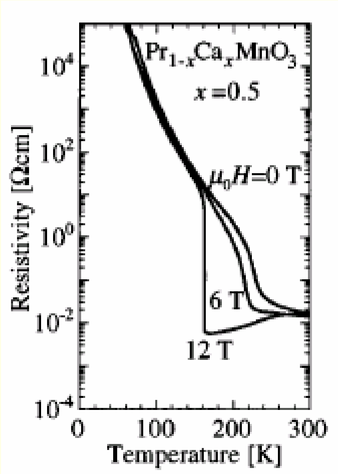
T_C increases as expected with field.

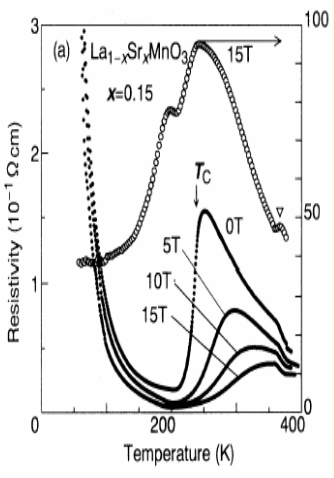
Is there anything unusual about the transport properties?



Upturn of resistivity – not typical of a good metal

Similar to …



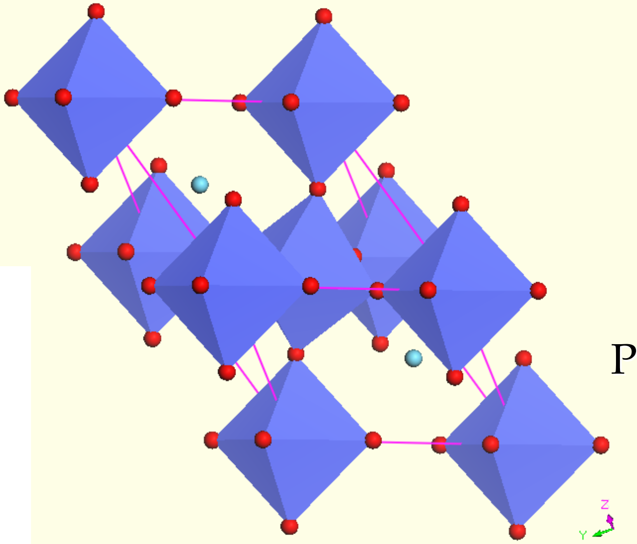


Tomioka et al., PRB 53, R1689 (1996)

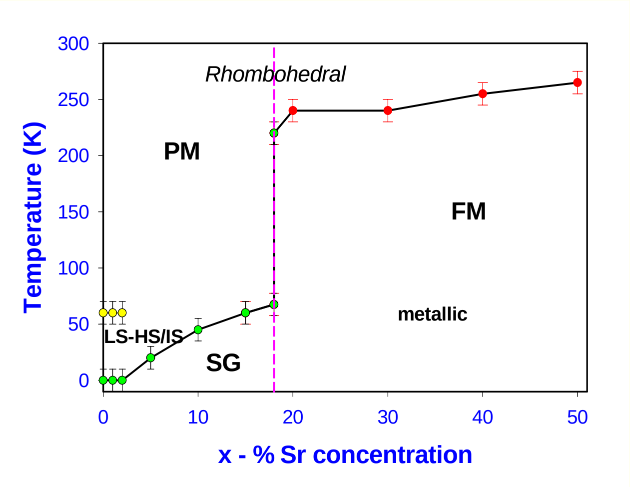
Urushibara et al., PRB 51, 14103 (1995)

What about the magnetoresistive effect?

- Metal-insulator and magnetic transitions are observed as a function of charge doping.
- MR is about 4 orders of magnitude smaller in cobaltites than in manganites. No sharp changes observed at T_c.



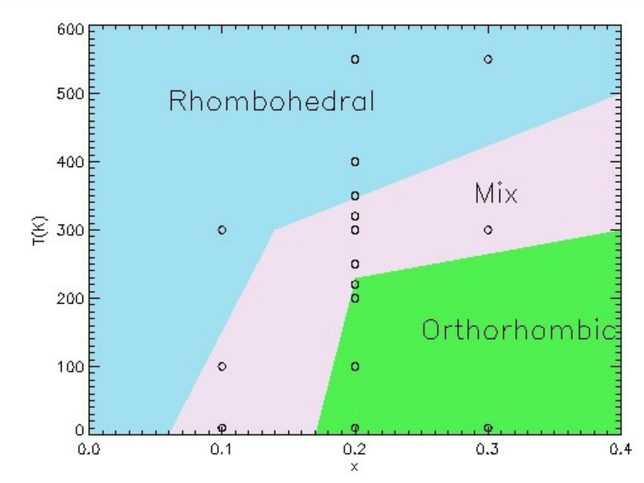
Sr-case



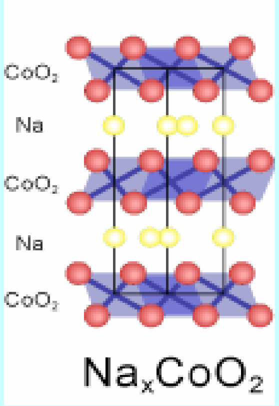
Louca et al., PRB **60**, R10378 (1999).

Ca-case

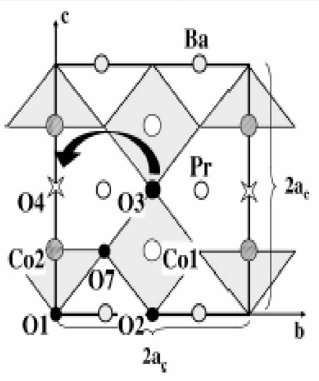
Phelan & Louca, unpublished data



How topological spin fluctuations influences charge transport in superconductor

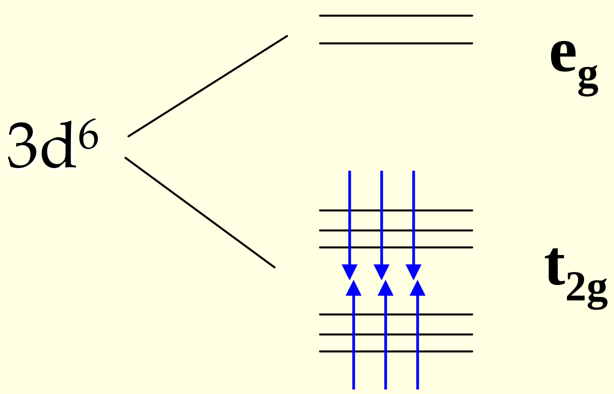


The octahedral and bipyramidal Co-O units in layered cobaltites are important in thermoelectric properties

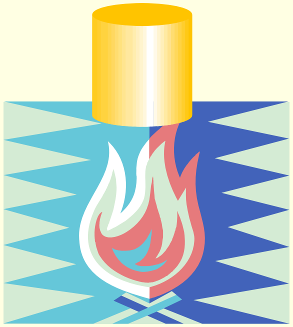


- What truly happens in the perovskite cobaltites?
  - How do they fit in the larger class of strongly correlated e$^-$ systems?
- For this, we look at the magnetic structure.

The ground state in $LaCoO_3$ is not magnetic



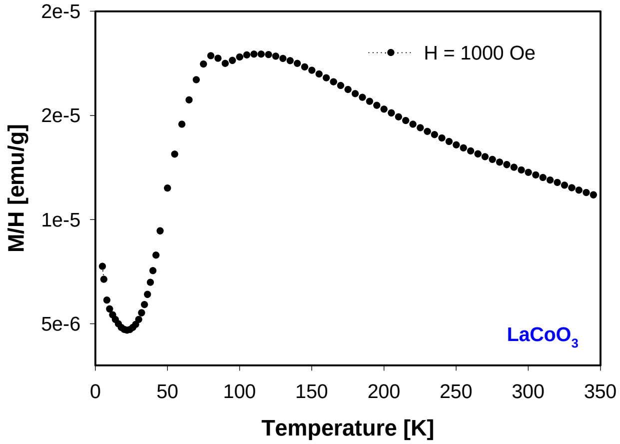
Total spin, S = 0



Octahedral field splitting

Co^{3+} - $3d^6$

But a transition occurs



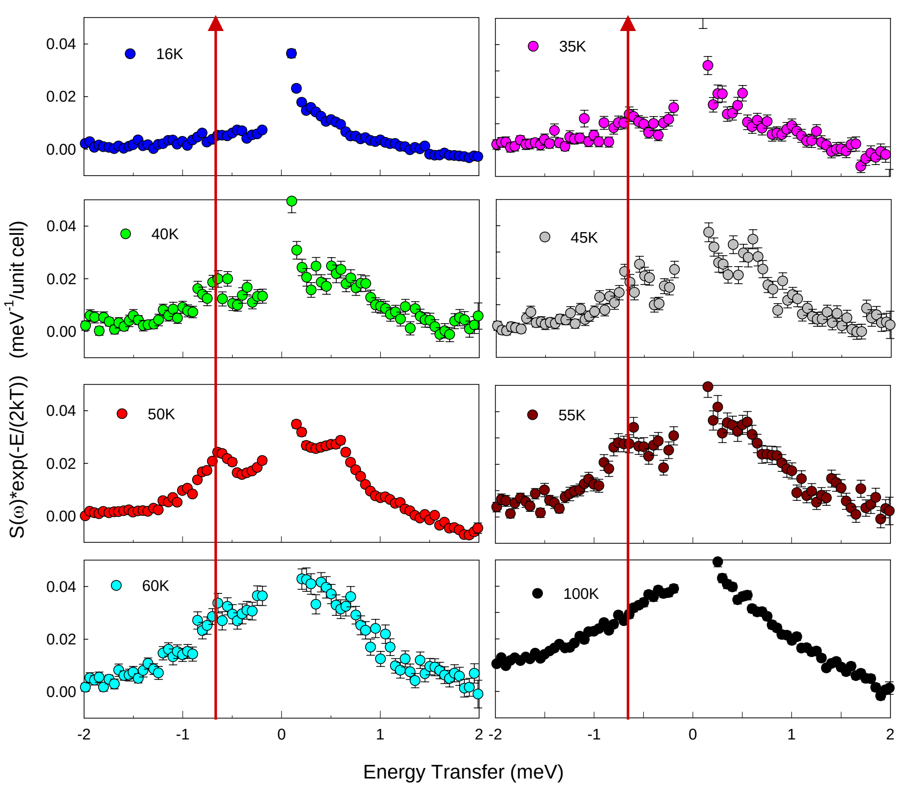
Low spin ⟷ Intermediate ⟷ High Spin

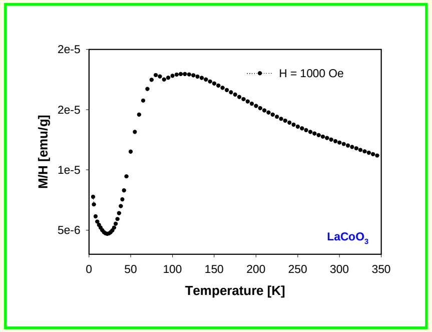
$t_{2g}{}^6 e_g{}^0$ $t_{2g}{}^5 e_g{}^1$ $t_{2g}{}^4 e_g{}^2$

Jahn-Teller active state

Magnetic component: 3 important contributions

1) Inelastic low energy excitations

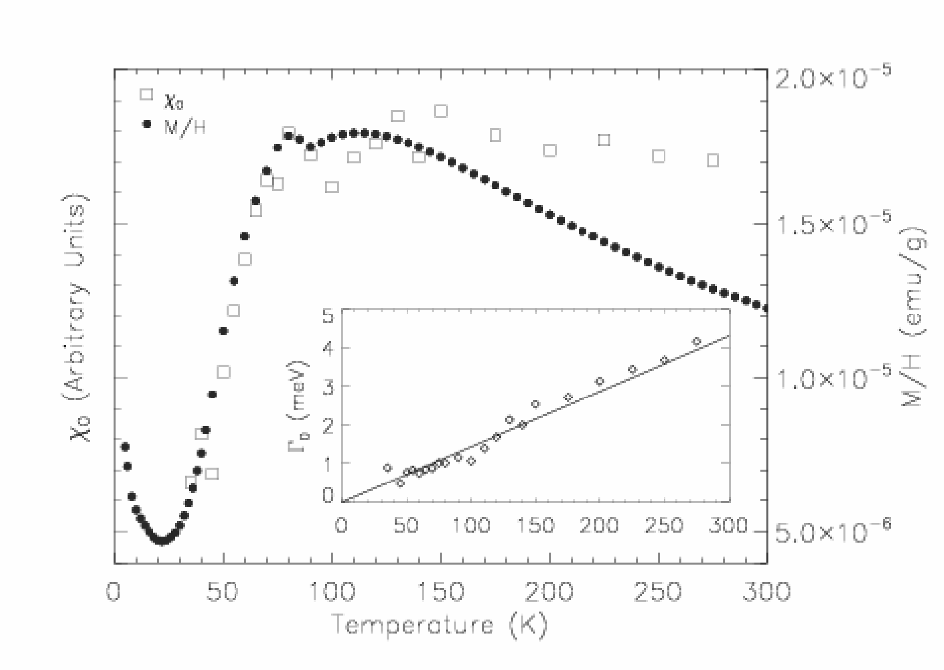




- Low energy magnetic excitation present. E_{char} ~ 0.6 meV.
- Superposed on the quasi-elastic signal due to increased paramagnetism. Together they get thermally enhanced with increasing temperature.

Inelastic intensity follows χ_{bulk}

$$\chi''(\hbar\omega)=\frac{\chi_0\Gamma_0\omega}{\omega^2+\Gamma_0^2}+\frac{\chi_1\Gamma_1\left|\omega\pm\omega_0\right|}{\left(\omega\pm\omega_0\right)^2+\Gamma_1^2}$$



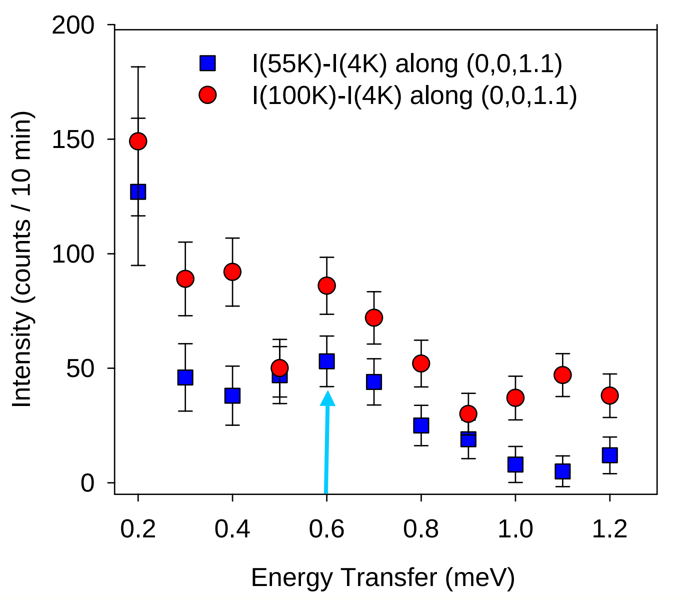
The first term (inelastic continuum intensity) contains χ_o, and is compared to the bulk susceptibility.

Origin of intensity is magnetic.

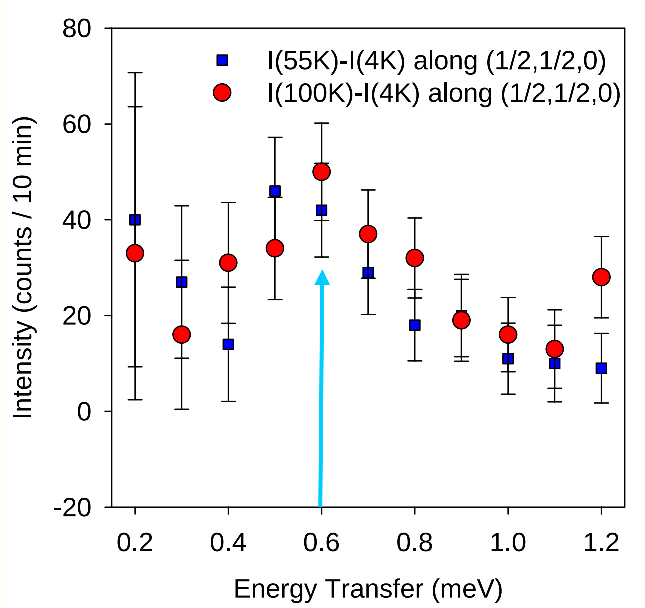
Phelan, Louca, Rosenkranz, Lee et al., PRL **96**, 027201 (2006).

The other two components

2) Ferromagnetic

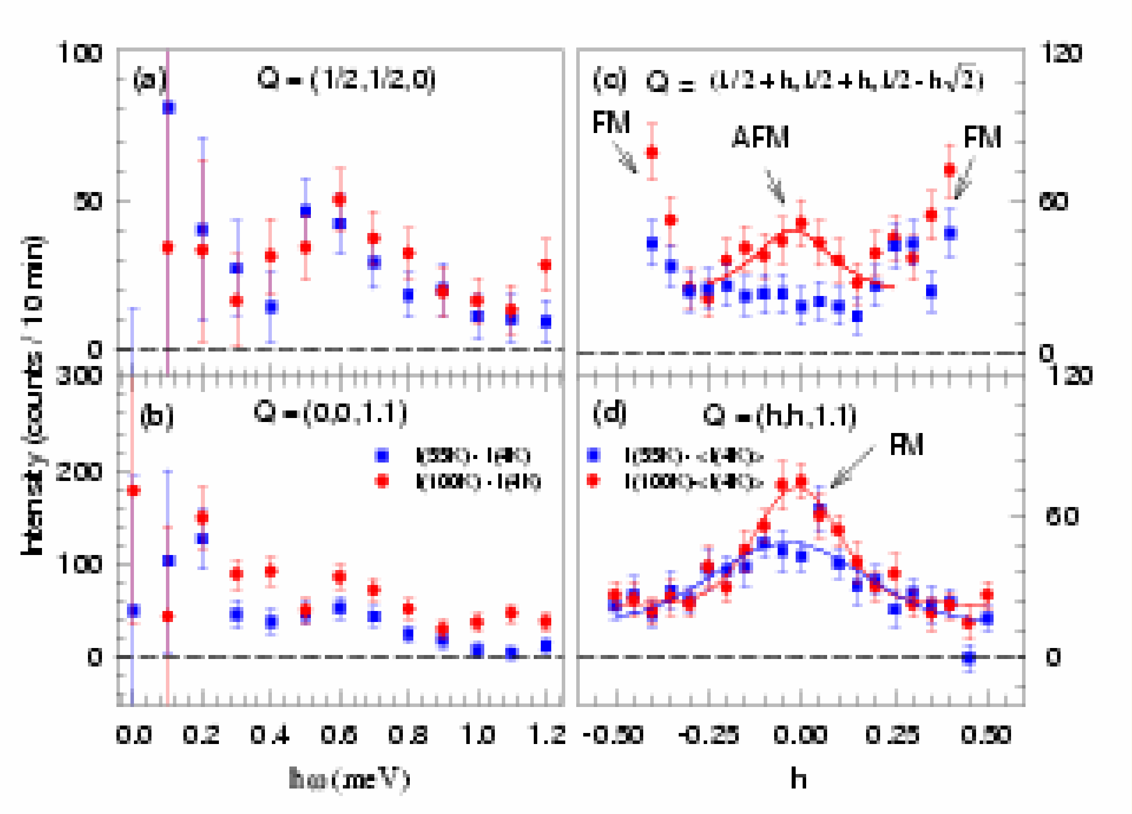


3) Antiferromagnetic!!!



- Constant Q scans at several points including (001), $\frac{1}{2}\frac{1}{2}0 \; and \; \frac{1}{2}\frac{1}{2}\frac{1}{2}$
- The energy excitation is present even at 100 K (washed out in the powder measurement).
- Excitation present at ferromagnetic and anti-ferromagnetic points although it is stronger at (001).

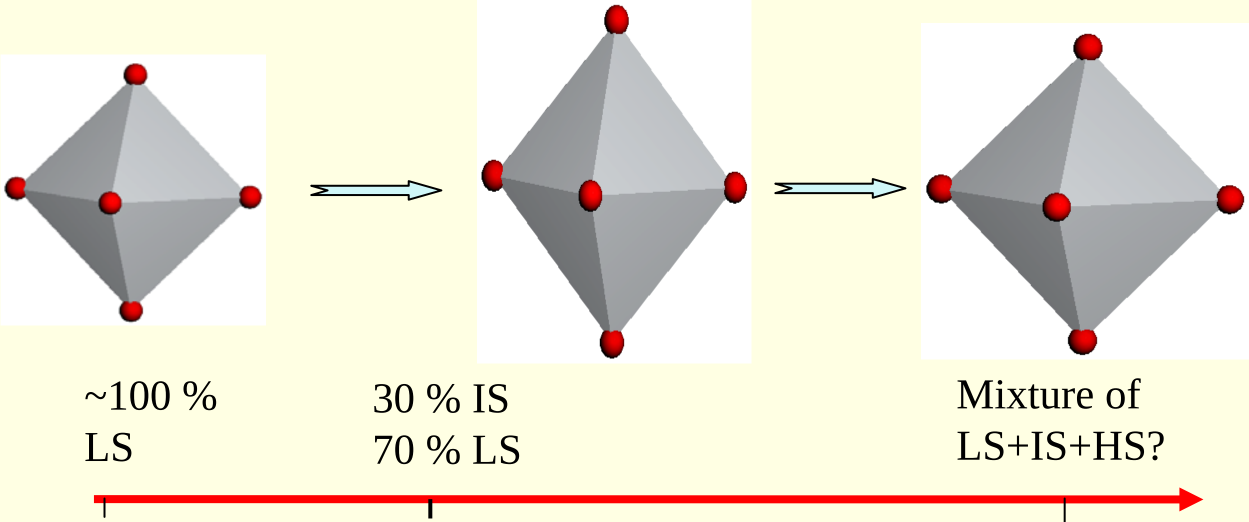
Strong ferromagnetism and weaker antiferromagnetism



DYNAMIC!

- A constant “background” is observed in both: Q-independent component due to paramagnetic fluctuations
- The PM signal hardly changes with temperature but the correlations between the ions become stronger.

Changes in the local atomic structure



~100 %
LS

30 % IS
70 % LS

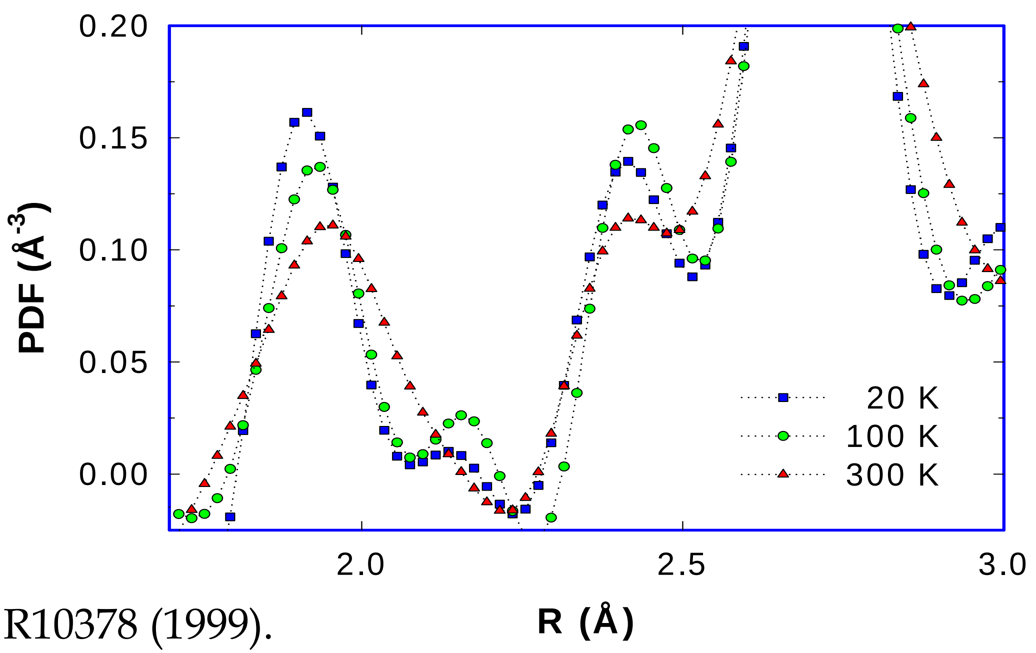
Mixture of
LS+IS+HS?

0 K

(Transition)

100 K

300 K



Louca et al., PRB **60**, R10378 (1999).

What does this mean for $LaCoO_3$?

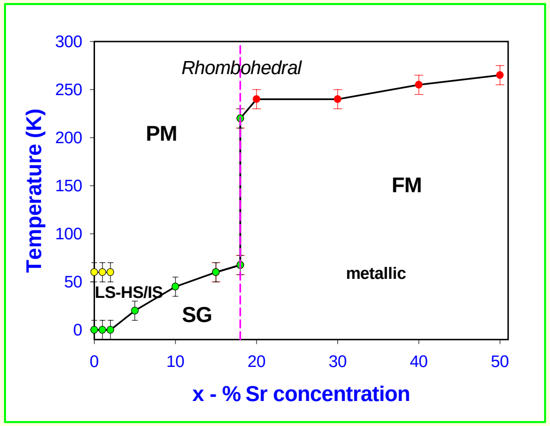
Possible scenarios: S = 0 ground singlet splits to an S=1 or an S=2

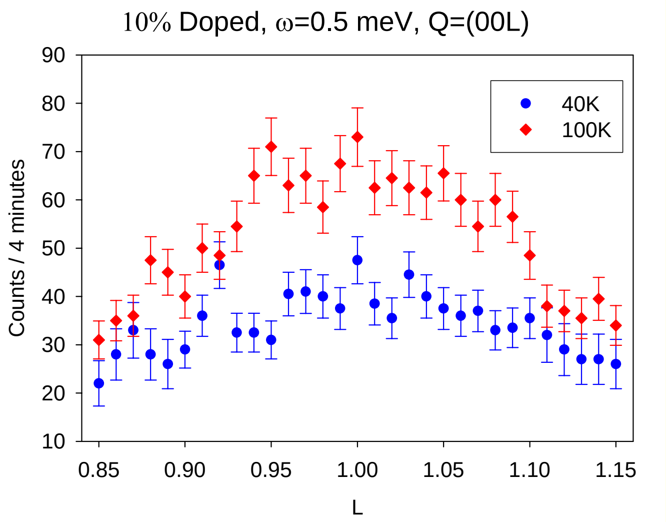
Theoretical arguments by Khomskii et al	Based on a value of g > 2 attributed the transition to an S = 2 state.
Experimental evidence by NMR, ESR and neutron	IS state – JT active Couples to orbital ordering

Observed: Dynamic magnetic correlations that have both FM and AFM characteristics (ξ = 3.6 Å)

Concluded: Orbital ordering is short-range and occurs in many directions. It is dynamic just like the magnetic correlations

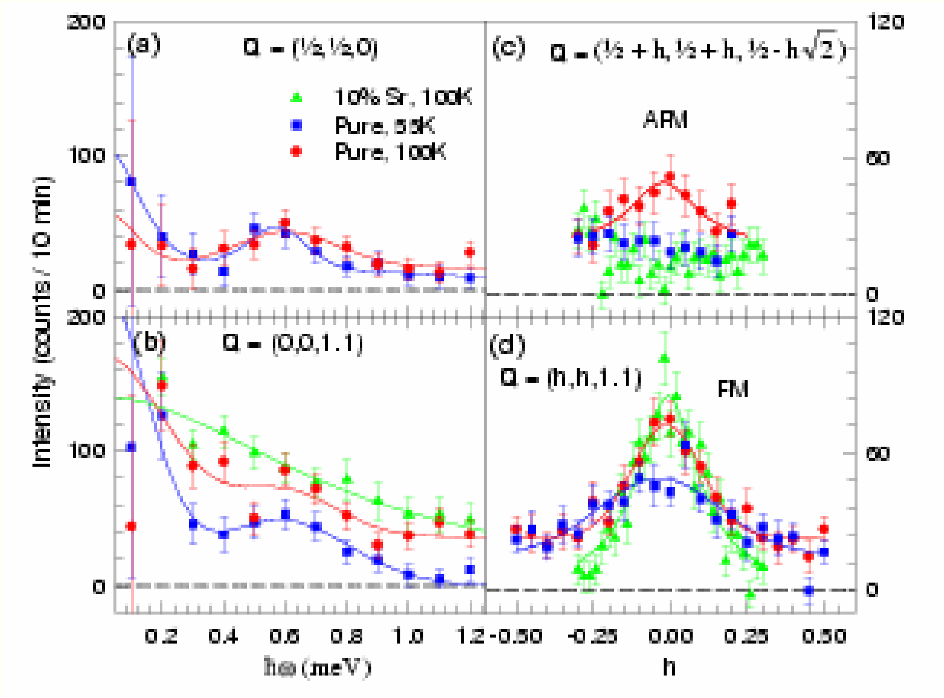
Introducing charges to the lattice changes the magnetic dynamics





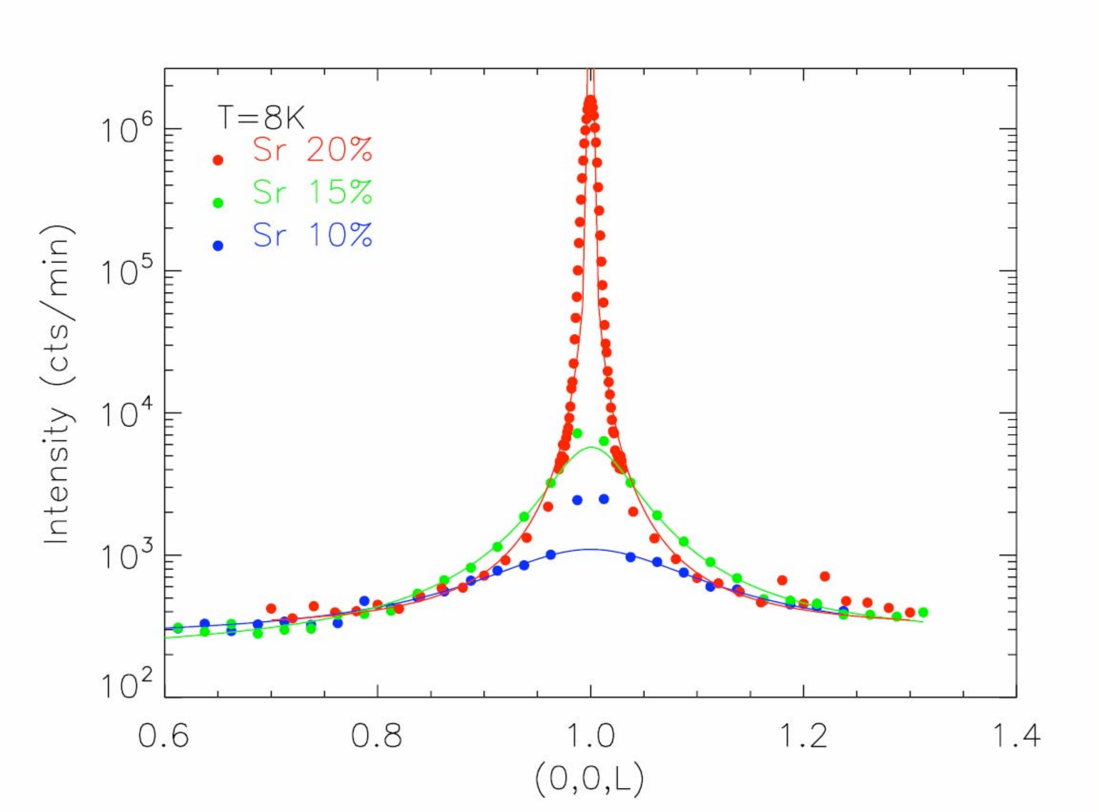
Dynamic magnetic signal is suppressed with cooling

Introducing charges to the lattice changes the magnetic dynamics



- The 0.6 meV mode (due to single ion effect) disappears.
- The AFM correlations are absent with Sr doping.
- Dynamic FM correlations are also suppressed.

Magnetic correlations become static and spatially isotropic

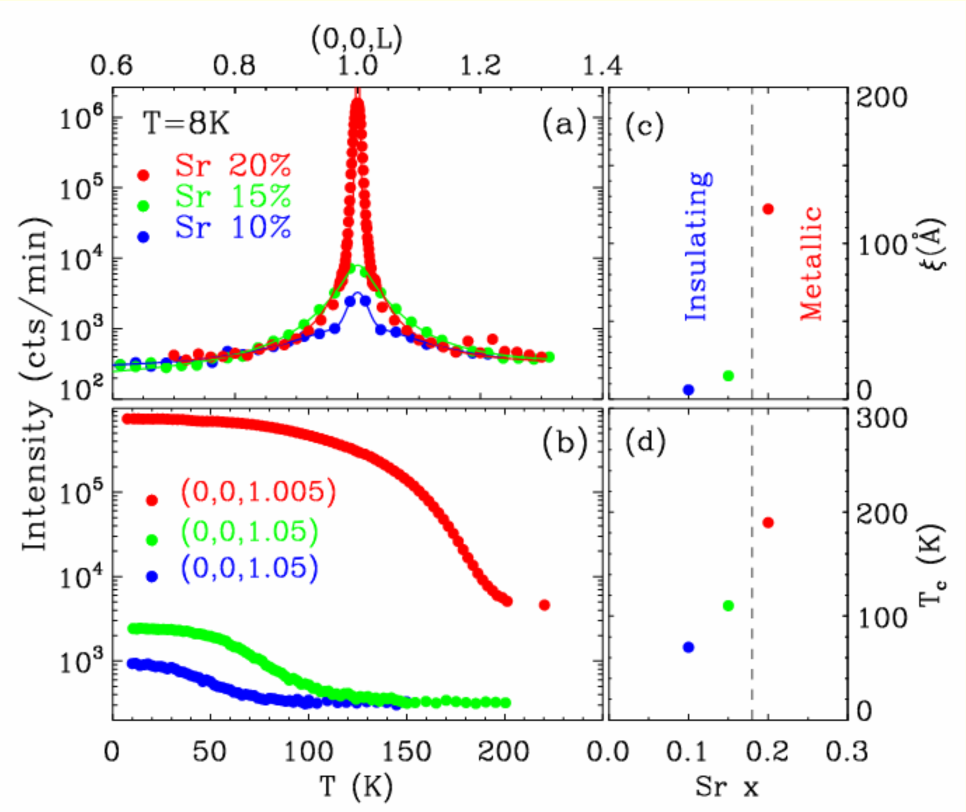


Measurements at the FM peak (001) reveal a circular object.

Scans centered around (001)

Circular object is isotropic in all directions

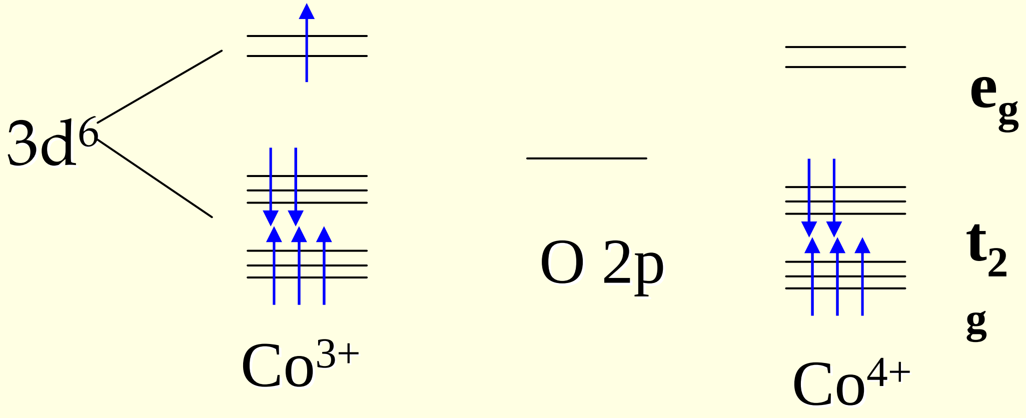
Correlation length increases with charge



- This is slow at first in the spin glass phase
- At 20 %, it is over 100 Å but still finite. The clusters increase in size in metallic state

Phelan, Louca et al., PRL **96**, 027201 (2006).

Correlation length increases with charge



3d^6

Co^{3+}

O 2p

Co^{4+}

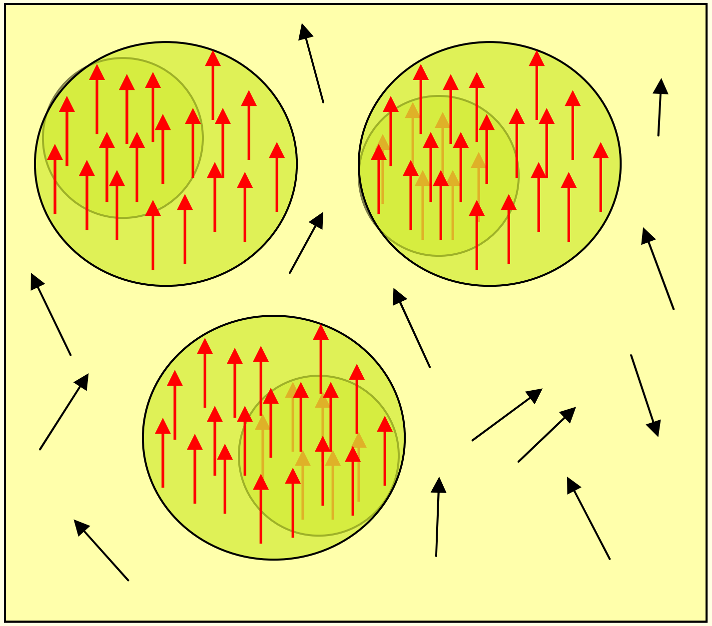
e_g

t_2

g

Double-exchange ferromagnetic coupling mediated by oxygen

Ferromagnetic bubbles expand with *x*
At the percolation concentration, conductivity happens



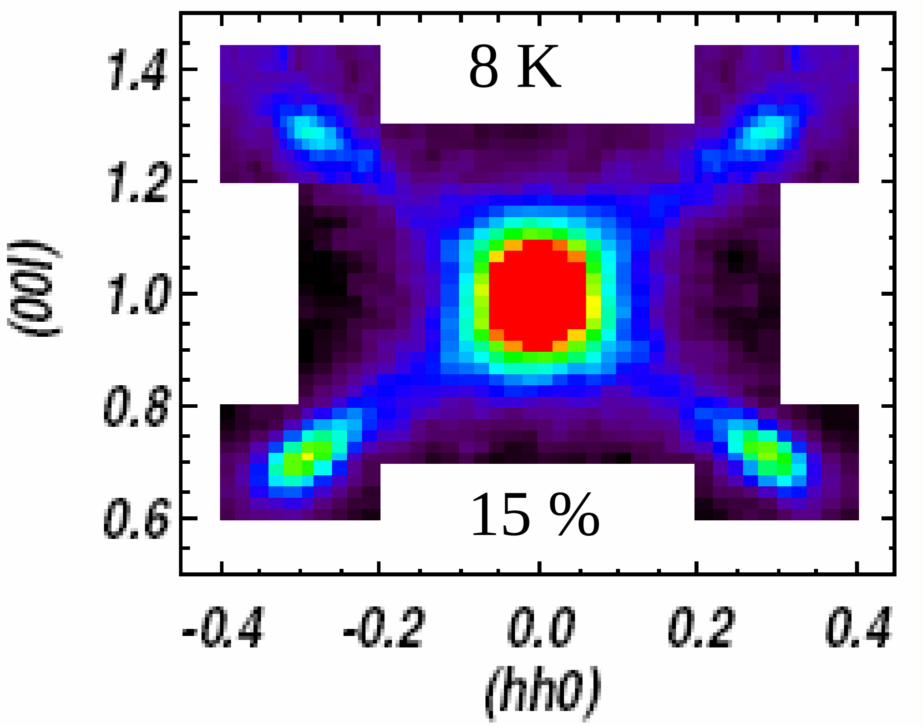
New competing state

Double exchange FM vs incommensurability

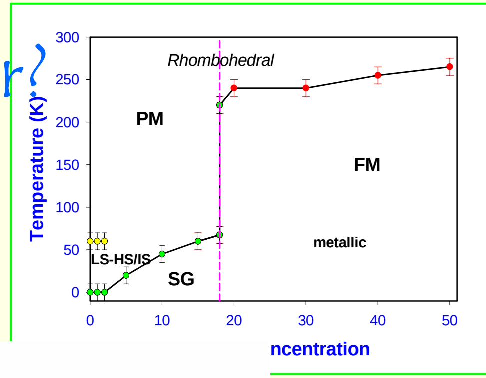
3 important contributions from elastic scattering

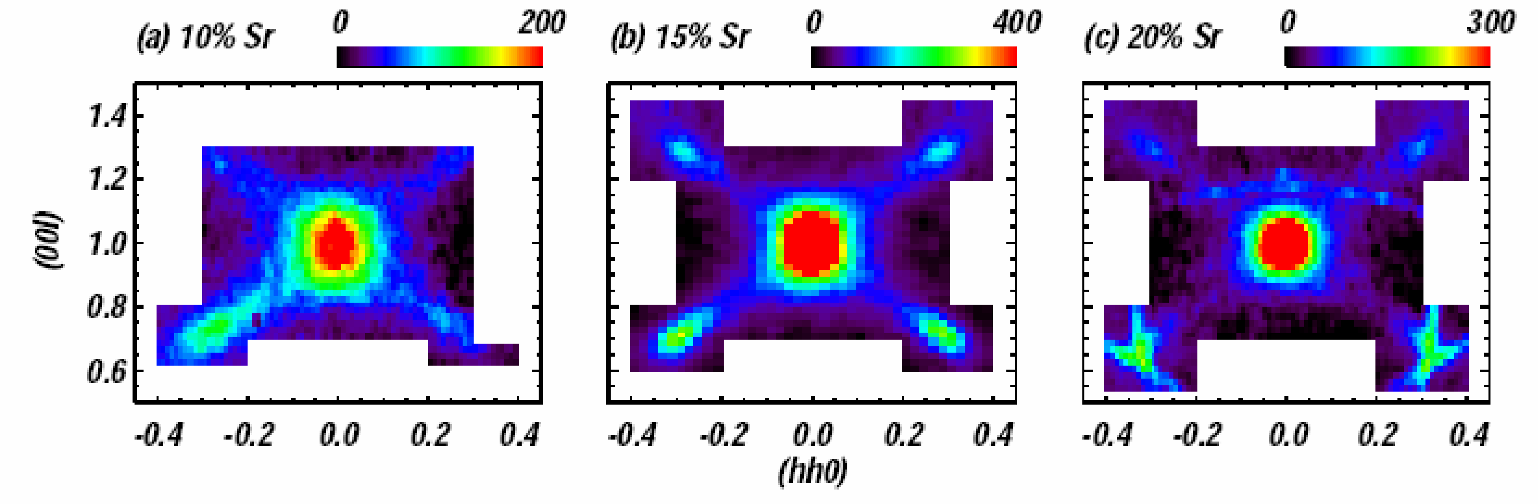
1. The isotropic feature centered at (001) due to FM correlations
2. An x-shaped pattern of weak diffuse intensity
3. Satellite peaks at the four corners.

hhl scans



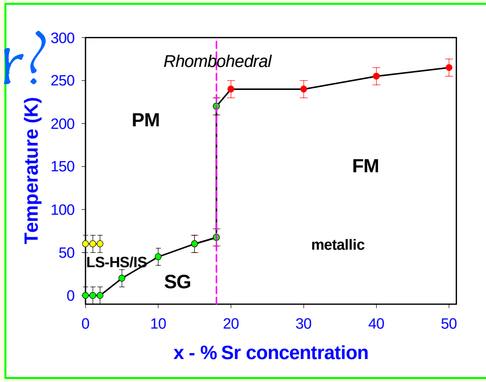
When does the superstructure appear?

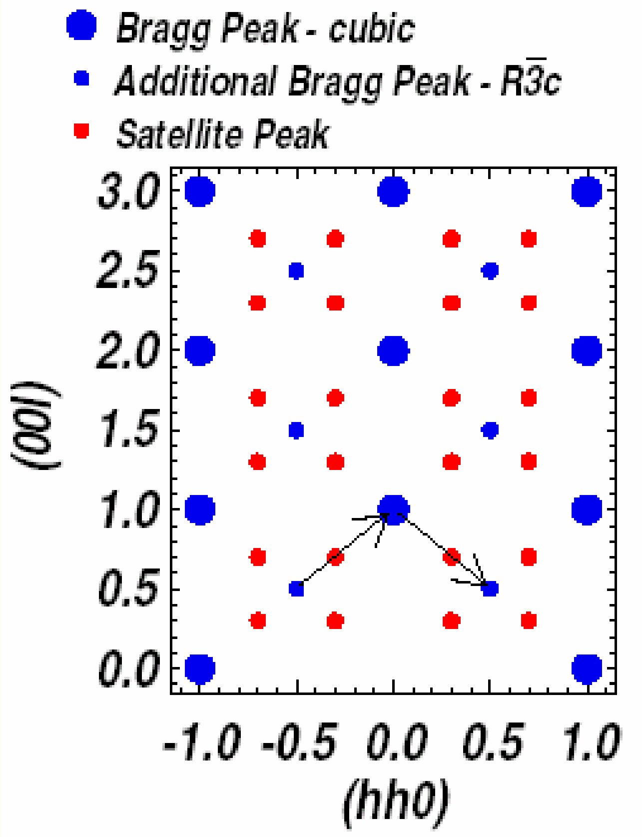




- Superlattice reflections appear at incommensurate positions **along (111) direction**.
- The positions of the peaks change with charge doping.
- What is the origin of this ordering?

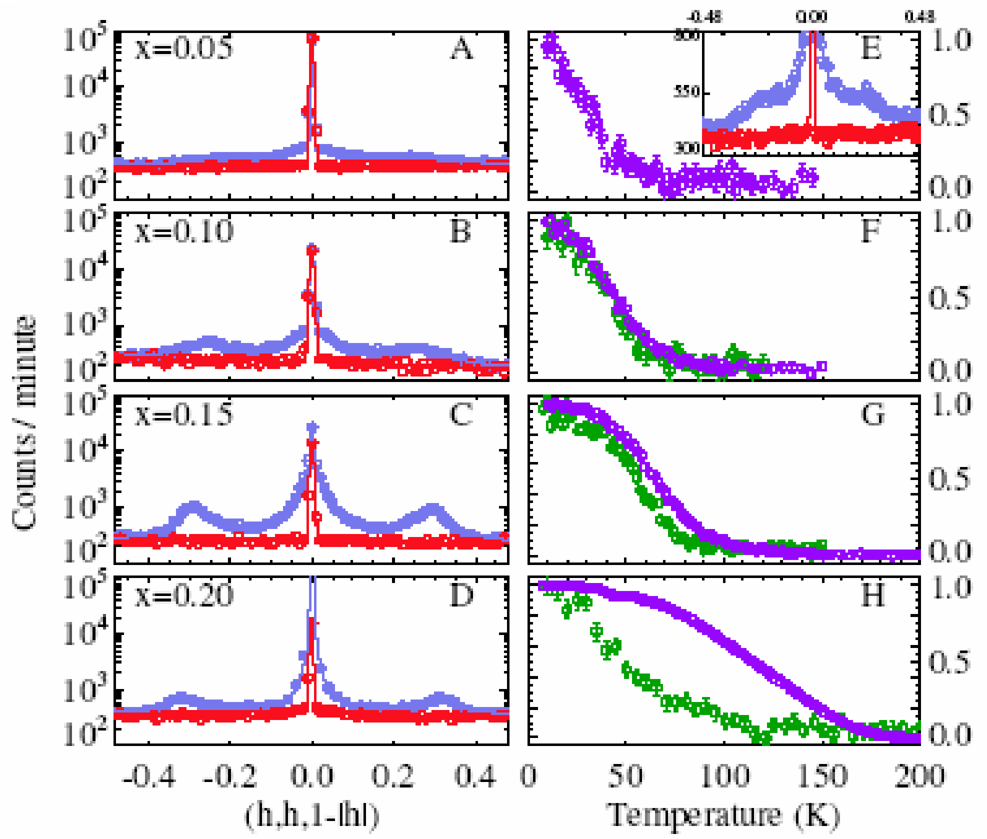
When does the superstructure appear?





Two ordering temperatures:

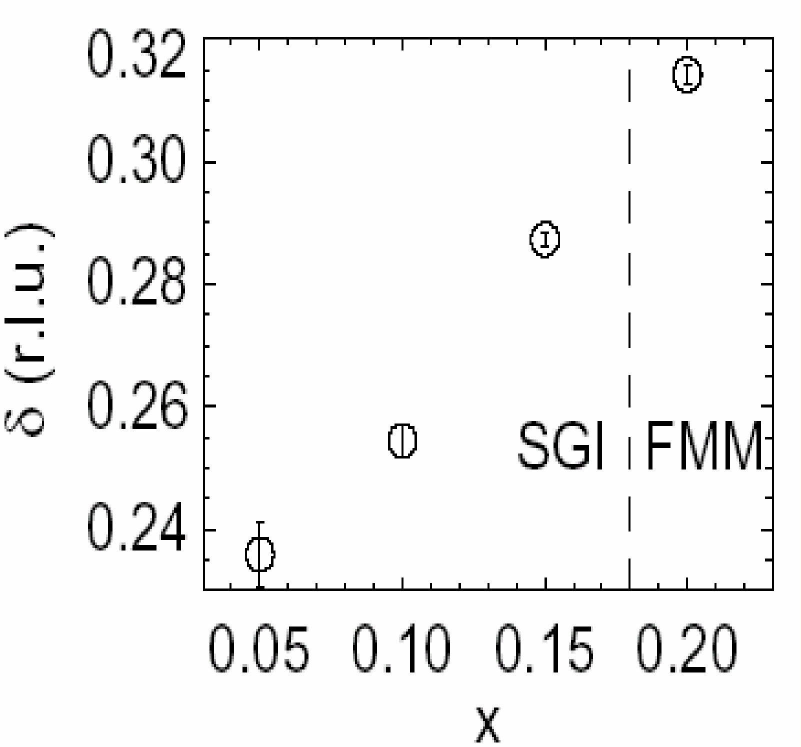
Cuts along (111)

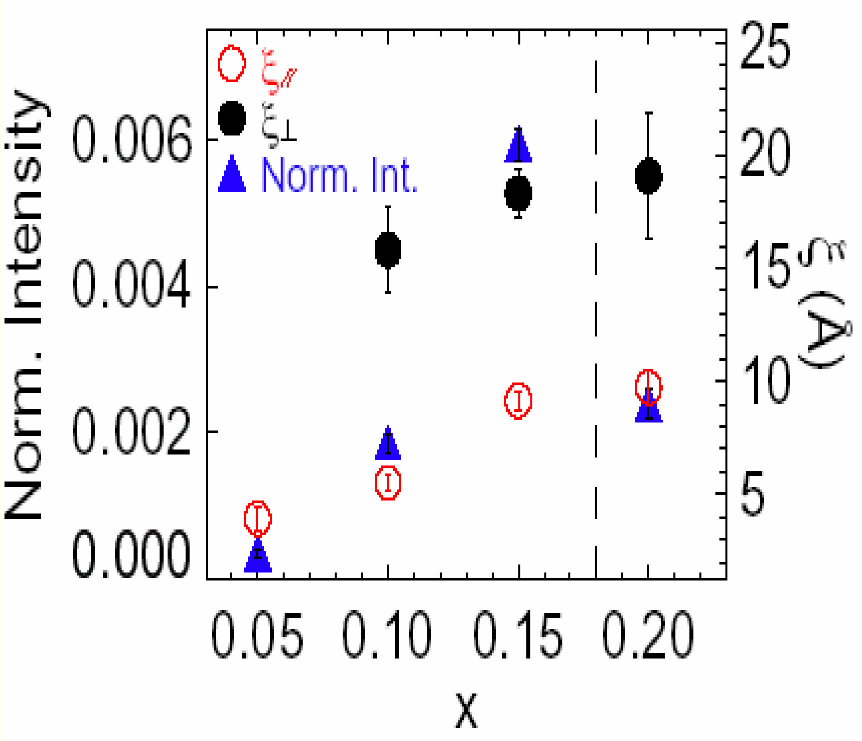


Onset temperature for FM or SG ordering occurs first Secondary ordering follows

- Peaks are absent above the long-range transition.
- The order parameter of the secondary spin ordering deviates more with the IM transition.
- Incommensurate peaks are magnetic as they follow the form factor dependence.

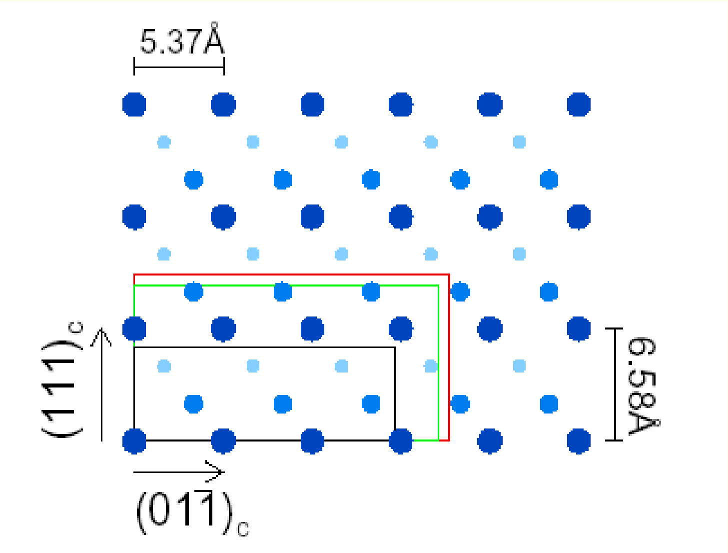
How the incommensurability varies with x

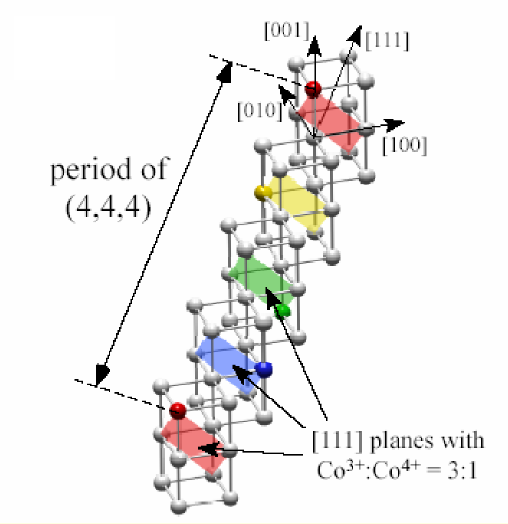




- Correlation length is longer in the perpendicular direction to (111) than in the parallel direction.
- Normalized intensity drops with the IM transition showing that peaks get weaker.

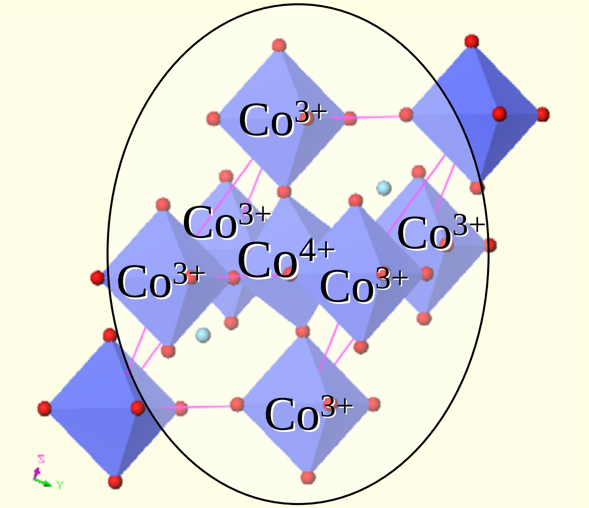
Physical representation of the ordering units



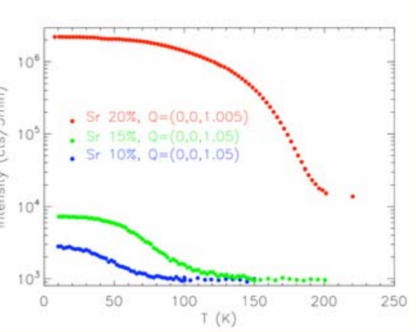


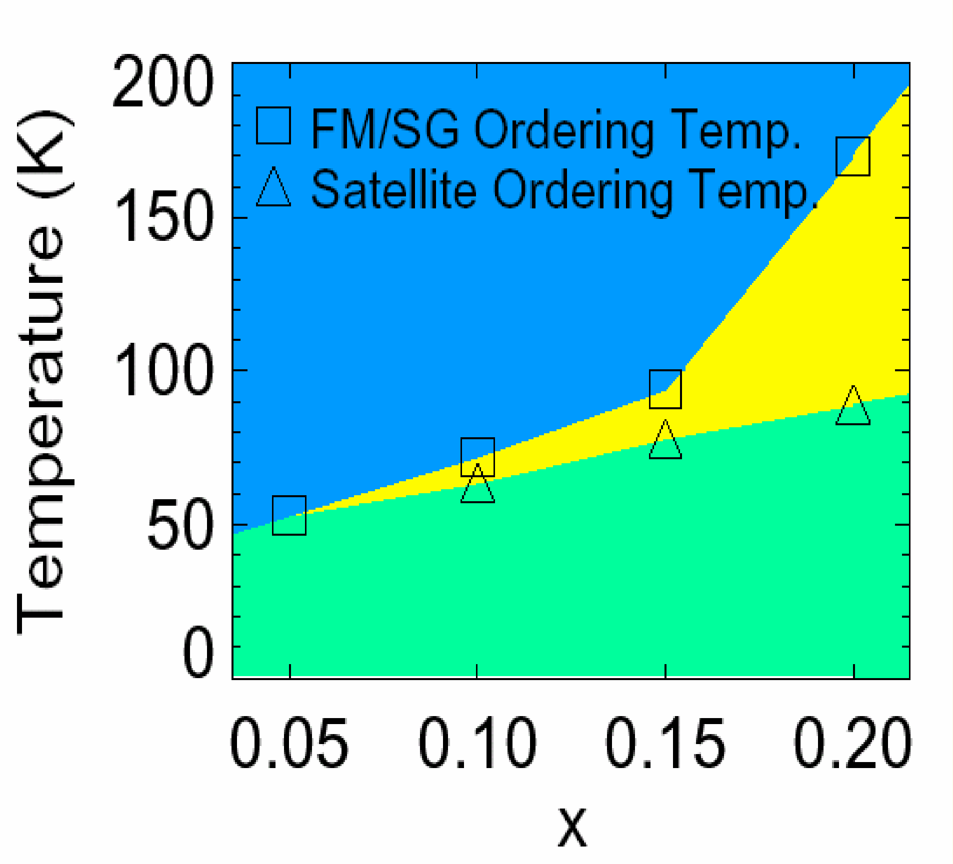
Spatial extent of the correlations

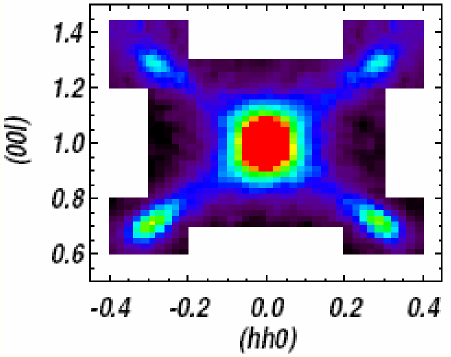
Self organization of 7-site clusters or extended polarons ordering along (111)

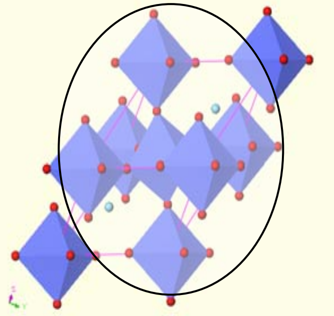


New phase diagram









Consequences of ordering:
-spin-charge localization
-plateau of the resistivity

In conclusion

- Two magnetic phases coexist and compete in the perovskite cobaltites.
- If the competition between the two is strong, they can phase separate.
- The incompatibility in the symmetry of the two does not allow the secondary spin ordering to extend to long-range.
- The possible charge localization might explain why the resistivity is not very low in the metallic phase and why the MR is small.
- At the same time, the charges are not fully localized and no long-range charge ordering is observed.
- The existence and organization of such structures appears to be a common feature in strongly correlated electron systems.

The End