Magnetic properties of spin-orbital polarons in lightly doped cobaltates

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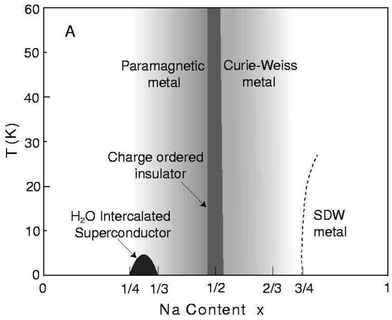
Les Houches 2006

What's so interesting about Na_xCoO_2?



M. L. Foo *et al.*, Phys. Rev. Lett **92**, 247001 (2004)

- Super-conducting when hydrated for $x \approx 0.3$
- Charge order at half-filling
- Large thermo-power for small hole concentration (large x)

für Festkörperforschung

What we are interested in.

- Layered structure: lattice spacing within the layers much smaller than between them
- Low temperatures: *A*-type anti-ferromagnetism for $0.7 < x < 1$: FM layers are AF coupled, *strong inter-plane coupling*
- Non-magnetic for $x = 1$.
- $0.7 < x < 1$: Susceptibility larger than can be explained by doped holes.
- Negative Curie-Weiss temperature: $\chi \propto 1/(T - \theta)$ with $\theta < 0$. Contradicts $\theta > 0$ inferred from coupling constants.

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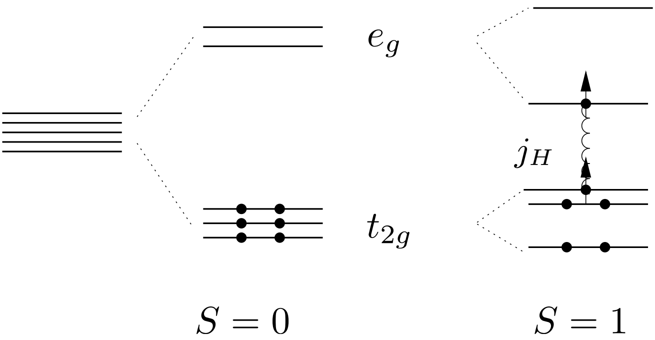
Doping-dependent orbital occupation

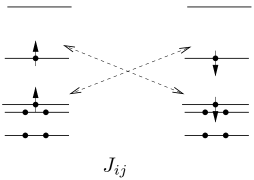
Without doping:
fully occupied t_{2g}
$S = 0$

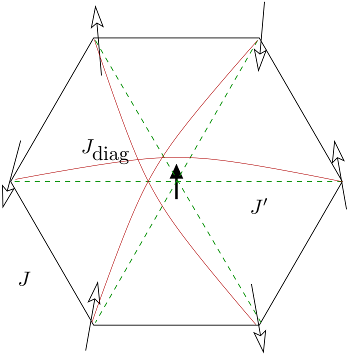
Next to a hole:
lattice is distorted
Hund's rule leads to $S = 1$



Resulting effective Spin Polaron

AF Exchange between $S = 1$





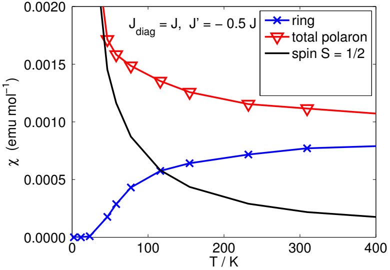
$$\mathcal{H} = J \sum_{i=1}^{6} \vec{S}_i \vec{S}_{i+1} + J_{\text{diag}} \sum_{i=1}^{3} \vec{S}_i \vec{S}_{i+3} + J' \sum_{i=1}^{6} \vec{s}_0 \vec{S}_i$$

$$J_{\text{diag}} \sim J \sim 10 - 30\text{meV}, \quad |J'| \lesssim J$$

G. Khaliullin, Prog. Theor. Phys. Suppl. **160**, 155 (2005)

Susceptibility

$$\chi(T) = \langle (S^z_{\text{tot}})^2 \rangle / T = \sum_l \frac{e^{-\frac{E_l}{k_b T}}}{ZT} \langle E_l | \Big(\sum_{i=0}^{6} S^z_i \Big)^2 | E_l \rangle$$



Small T: spin $1/2$
Large T: $S = $ 1-ring

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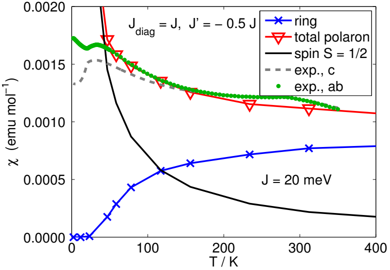

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Exp. data: S. Bayrakci *et al.*, Phys. Rev. B **69**, 100410(R) (2004)

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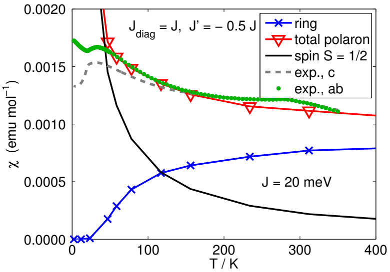
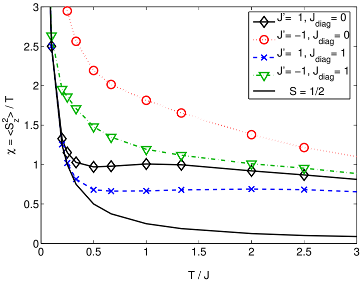




Small T: spin $1/2$
Large T: $S = 1$-ring

all parameters:
$\chi_{\mathrm{polaron}} \gg \chi_{S=1/2}$ for large T

Exp. data: S. Bayrakci *et al.*, Phys. Rev. B **69**, 100410(R) (2004)

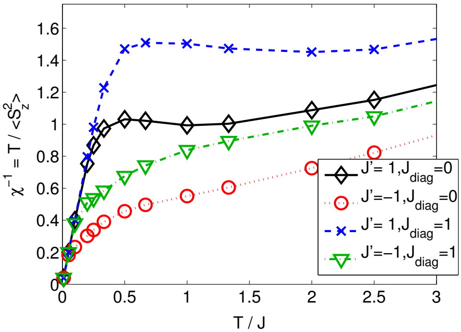
Inverse Susceptibility

Curie-Weiss law

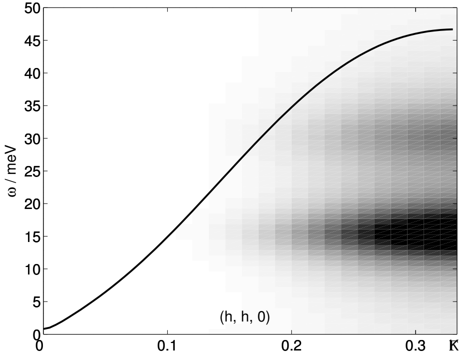
Large T: $\chi \sim 1/(T - \theta)$

Continue high-T behavior: Line would cross T-axis at $T = \theta < 0$.

FM $J' < 0$ gives better fit to experiment.



Dynamic Spin-Structure Factor $\chi''(\vec{q}, \omega)$



Gray-shades: Excitations of a polaron

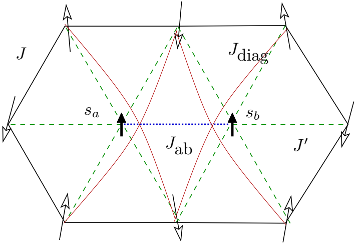
Line: Spin-Wave fit to measurements

S. Bayrakci *et al.*, Phys. Rev. Lett. **94**, 157205 (2005)

Spin waves are expected to scatter on polaron excitations.

Bi-polaron

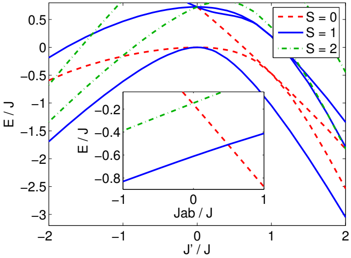
Polarons can move, they meet occasionally: bi-polaron



Direct coupling between the two holes with $S = 1/2$:
$J_{ab} \sim -J$ is FM

Polarons are dilute.
They rarely form bi-polarons, this reduces the coupling.

Indirect coupling via spin-clouds



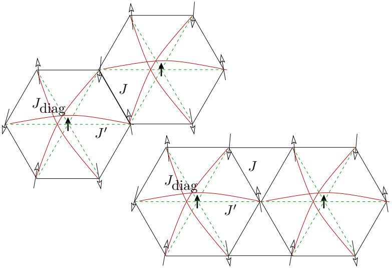
s_a polarizes the spin cloud, the spin cloud makes s_b parallel to s_a:
Lowest state is triplet, additional coupling $J_{\mathrm{eff}} = E_{\mathrm{triplet}} - E_{\mathrm{singlet}}$ is also FM.

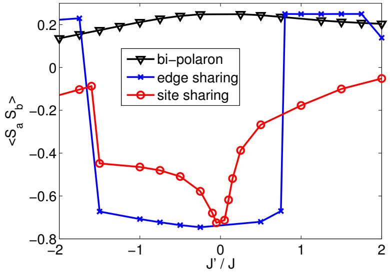
Total nearest-neighbor coupling is

- FM
- Rather weak because it only operates between the dilute polarons.

More interacting configurations (n.n.n.)

Spin clouds can mediate interaction between next nearest neighbor polarons.





Next nearest neighbor interaction is AF!

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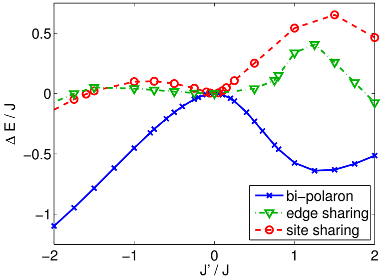
Total polaron coupling



n.n. coupling is FM

n.n.n. coupling is AF:

- $J' > 0$: $J_{\text{n.n.n.}} \sim -J_{\text{n.n.}}$
  strong n.n.n. coupling
- should therefore show up in spin waves as deviation from n.n. Heisenberg
- $J' < 0$: $J_{\text{n.n.n.}} \sim 0$
  almost only n.n. coupling

Spin waves can give information about J':
n.n.-Heisenberg-like suggests $J' < 0$.

Effective Spin-Polaron model for Na_xCoO_2

- Susceptibility:
  High temperature: Many spins with $S = 1$
  Small temperature: Rings of $S = 1$ freeze into $S_{\text{tot}} = 0$.
  Large susceptibility at high T, negative θ.
- Dynamic spin-structure factor:
  Largest weight at $\vec{q} = K = (1/3, 1/3, 0)$, $\omega \lesssim J$
- Polaron interaction via bi-polarons:
  Dilution reduces in-plane coupling. Effective n.n. interaction is FM.

M. Daghofer, P. Horsch, G. Khaliullin, Phys. Rev. Lett. **96**, 216404 (2006)