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Ferromagnetism of $LaCoO_3$

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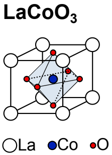
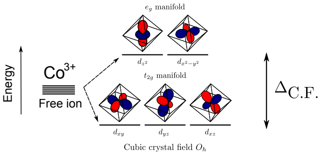
Intro: low-T physics of $LaCoO_3$ (cubic structure)

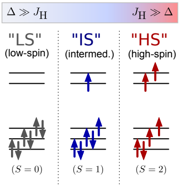
The compound is studied since 1960's, both experimentally and theoretically



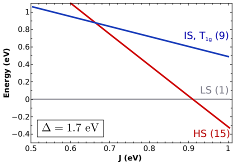
Orbital structure

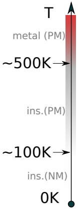


Relevant spin states



Tanabe-Sugano diagram



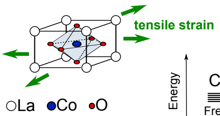


Important facts:

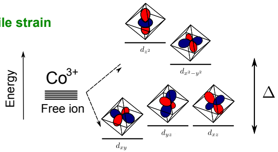
- ✓ at low T: insulator, sizeable charge gap
- ✓ nonmagnetic $\longrightarrow$ paramagnetic
- ✓ higher relevance of HS/IS among low-T excitations is still under debate
- ✓ "unusual" behavior at high magnetic fields (~ 100 T)
- ✓ proximity to [A] HS–LS spin-state ordering, [B] BEC of IS ($S = 1$) excitons
- ✓ short-range ferromagnetic correlations (no long-range order)

Intro: low-T physics of $LaCoO_3$ (stretched structure)

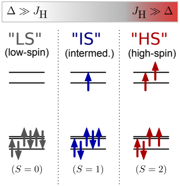
film $LaCoO_3$



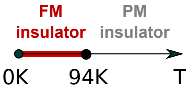
Orbital structure



Relevant spin states



Magnetic ordering



Important facts:

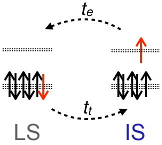
- ✓ insulator
- ✓ long-range ferromagnetic (FM) ordering with $T_C = 94\,\mathrm{K}$
- ✓ magnetic structure with a propagation vector (1/4, -1/4, 1/4)
- ✓ g factor $\approx 0.7\mu_\mathrm{B}$ per Co atom
- ✓ controversial predictions from DFT+U approaches:
  - candidate ..–HS–LS–HS–LS–.. with magnetic moments on HS sites
  - AFM/FM alignment depends on U; the FM couplings are too small

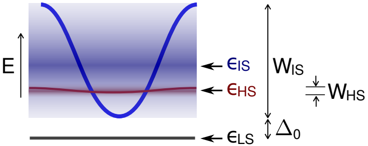
?? 1/4 structure, T_C, g factor and the physics behind

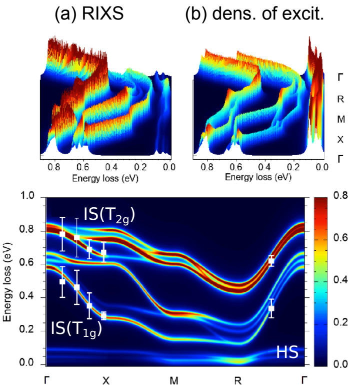
Intro: highly-dispersive IS, immobile HS (cubic structure)

LS state ≡ vacuum

IS state ≡ exciton (electron+hole)



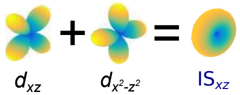




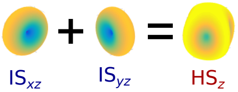
experiment + theory

[R.-P. Wang, A. Hariki, *et al.*, PRB **98**, 035149 (2018)]

$IS(T_{1g})$ exciton:

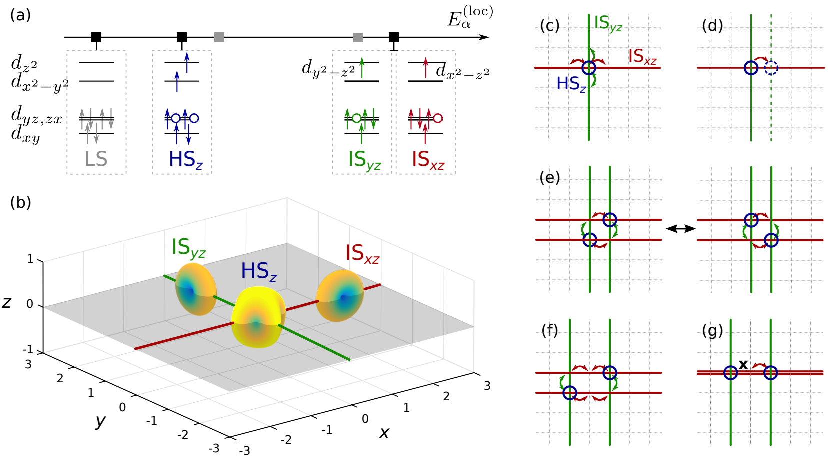


$HS(T_{2g})$ bi-exciton:



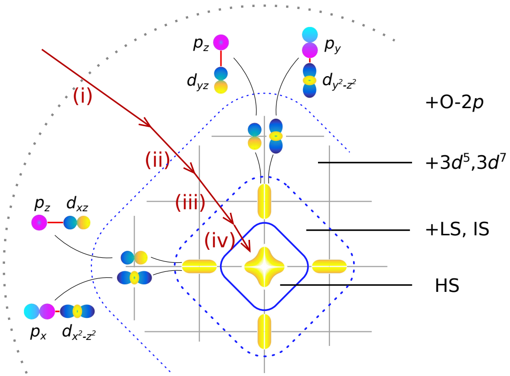
- ✓ $E_{HS} < E_{IS}$ ⇒ effective on-site attraction of IS
- ✓ account for formation/decay of HS lowers its energy!

{IS,HS}-boson picture of the film $LaCoO_3$



Q.: How we extract the hopping and interaction amplitudes for the real crystal?
A.: Series of well-established and logically-consistent approximations...

Methodology



(i) density functional theory (DFT)
[wien2k]

(ii) Wannier projection to d-only Fermi-Hubbard model, adding interaction in the Slater form
[wien2wannier, wannier90]

(iii) strong-coupling (t^2/U) expansion in the d^6 manifold
[ED, 2nd order pert. theory]

⇒ effective Bose-Hubbard model
treatment: exact diagonalization

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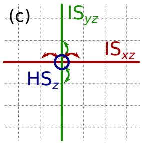
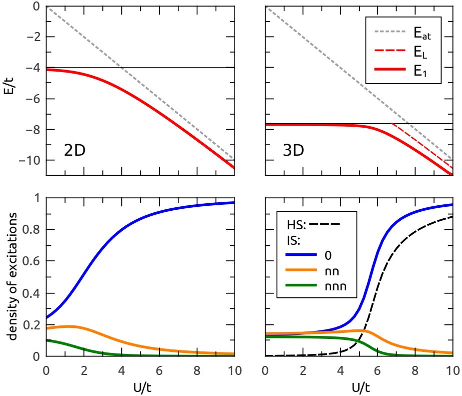
optional, but useful step:

(iv) reduction to the HS-only (spin-1/2) Ising model

level (iii): "isolated" HS states

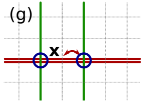


- one IS state per line (length $\sim$ 10 sites),
- known IS hopping amplitude t,
- local attraction U is determined by E_{HS}, E_{IS} (taken as tuneable parameter)

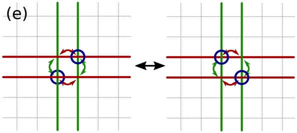


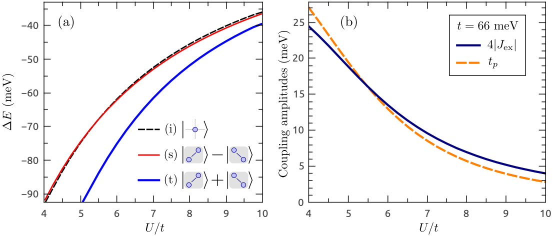
- realistic regime: $U/t \approx 8$
- energy of HS is lowered by nn fluctuations
- HS is surrounded by the IS "cloud" $\Rightarrow$ "dressed" state

Q.: What if we add excitations?
A.: Rather not on the same line:
(hard-core constraint $\rightarrow$ repulsion)



plaquettes with two "dressed" HS & their FM exchange

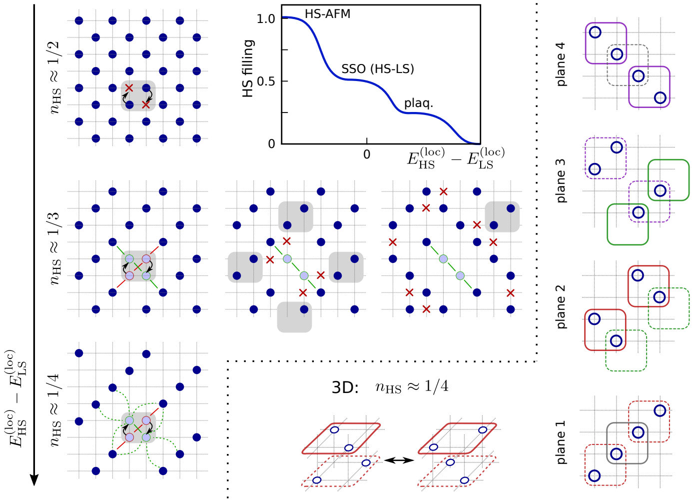




- effective FM coupling on the diagonal: $4J_{\rm ex} \approx -7\,$meV at $U/t = 8$ ($J_{\rm ex} < 0$ results from the spin-resolved analysis)
- diagonal ↑-↑ attraction $V = 4|J_{\rm ex}|$ is comparable to $T_c \approx 94\,$K
- the amplitude for the pair flip t_p is of the same order

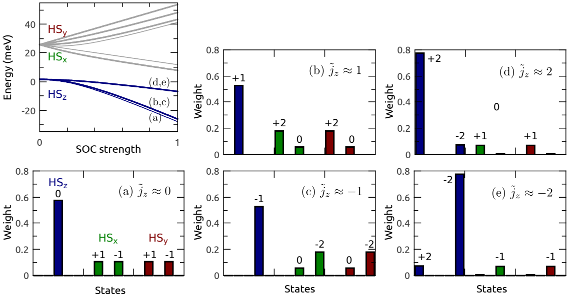
diagonal attraction & nn (nnn) repulsion along $x, y \Rightarrow$ stripe formation!

decorating lattice with the “dressed” HS



supporting remarks: role of SOC; g-factor estimates

Spin-orbit coupling is analyzed in the atomic limit



main effects of SOC:

- filters 3 out of 5 lowest spin states ($\tilde{j} = 0, \pm 1$);
- acts as a single-ion anisotropy term in spin space
- adds a portion of $\mathrm{HS}_{x,y}$ in the HS_z set

g factors in the subspace of HS with $\tilde{j} = 0, \pm 1$:

$$M_r = \mu_\mathrm{B}(2s_r + l_r) = \mu_\mathrm{B} g_r \tilde{j}_r$$

$g_\parallel \approx 2.73$
$g_\perp \approx 2.64$
(per HS atom)

Experiment:
$g_\mathrm{Co} \approx 0.7 \pm 0.1$

$$g_\mathrm{Co} \approx \frac{1}{4} g_\mathrm{HS}$$

Summary / Acknowledgements

Brief $\sum_{\text{slides}}$

- FM in the insulating film $LaCoO_3$ originates from the highly-fluctuative character of excitations;
- the lattice is filled by $1/4$ of HS; these form diagonal stripes (agrees with exp.-observed propagation vector)
- FM couplings on diagonals are of the same order as exp. T_C
- at $n_{\mathrm{HS}} = 1/4$, g factors fall close to exp. values
- the low-energy physics of film $LaCoO_3$ $\Leftrightarrow$ the (extended) Ising model

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Thank you for your attention!