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Hard-core bosonic DMFT study on spin-state transition in $LaCoO_3$

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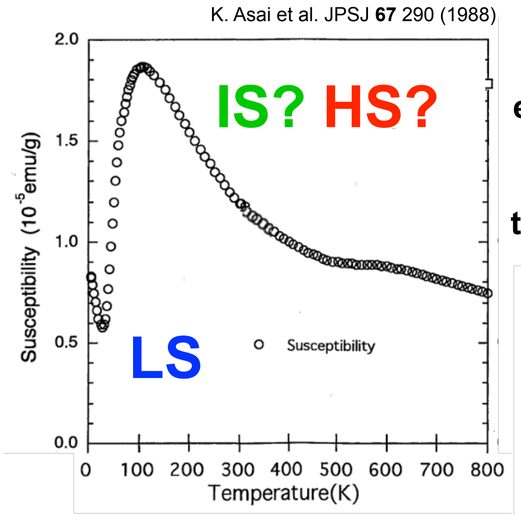
We study spin-state transition in $LaCoO_3$ using 'bosonic' dynamical mean field theory (B-DMFT). The spin excitations of $LaCoO_3$ can be viewed as a collection of mobile spin-triplet excitons which obey a hard-core (HC) constraint. One-particle DMFT self-energy is ill-defined in the HC constraint, leading a numerical difficulty to achieve a DMFT self-consistent condition. To solve this, 1) we adopt a Dyson-Mori formalism in computing the self-energy and 2) develop a continuous-time quantum Monte Carlo impurity numerical solver implementing a broken commutation relation by the HC constraint. To test our methodology for HB-DMFT, we calculate a phase diagram of two-dimensional two-band Hubbard model, allowing a staggered order of boson densities that corresponds to a spin-state order (SSO) in the electronic system. We discuss a future plan for an application of the HB-DMFT method to a more realistic model of $LaCoO_3$.

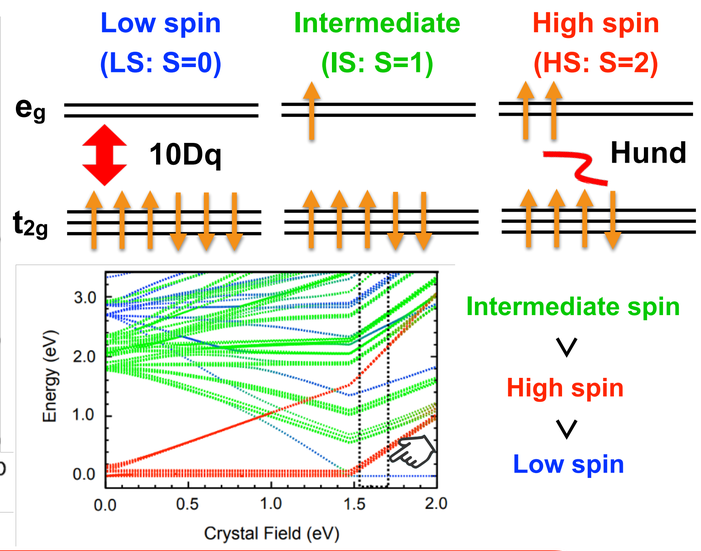
Introduction: $LaCoO_3$

Susceptibility

K. Asai et al. JPSJ **67** 290 (1988)



$Co^{3+}(3d^6)$: atomic picture

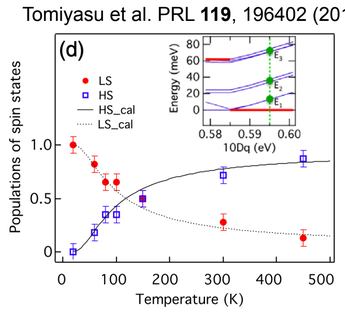


Question

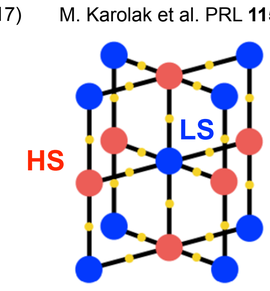
LS to HS thermal transition Co^{3+} in atomic picture?

Experiment vs Theory

Tomiyasu et al. PRL **119**, 196402 (2017)



M. Karolak et al. PRL **115**, 046401 (2015)



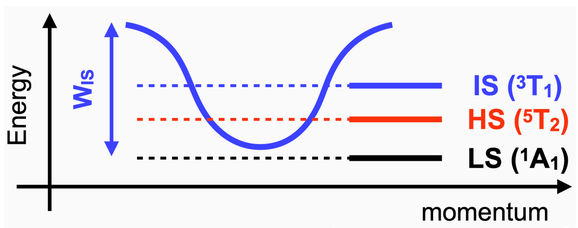
Messages

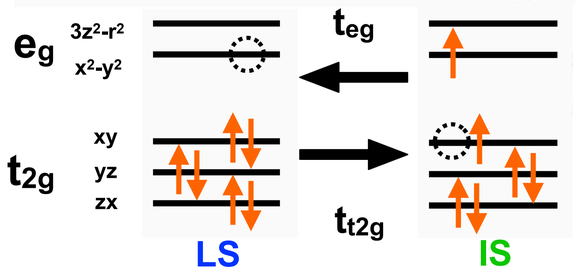
- No experimental evidence of HS-LS long-range ordering
- Spin-state transition beyond the atomic picture

Excitonic picture: mobility of IS

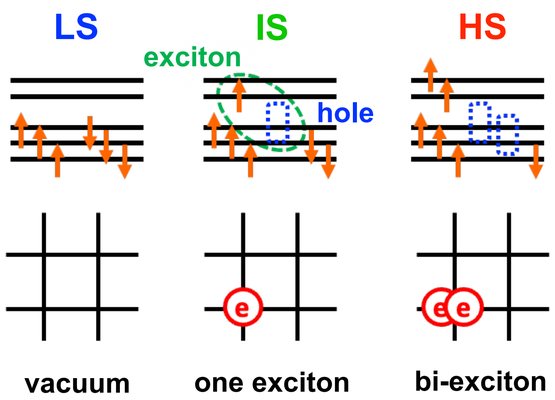
Mobility of intermediate spin

A. Sotnikov et al. Sci. Rep. **6**, 30510 (2016)

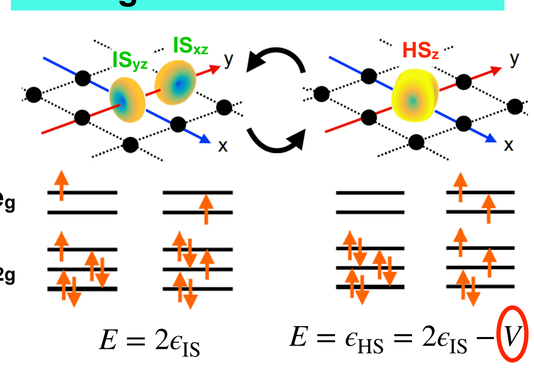




$Co^{3+}(3d^6)$: excitonic picture



Strong attractive interaction



$$E = 2\epsilon_{IS} \qquad E = \epsilon_{HS} = 2\epsilon_{IS} - V$$

Messages

- Large dispersion of IS
- Two IS excitons into one HS exciton (immobile bi-exciton)
- Strong attractive V in $LaCoO_3$

Theory: attractive bosonic Hubbard model

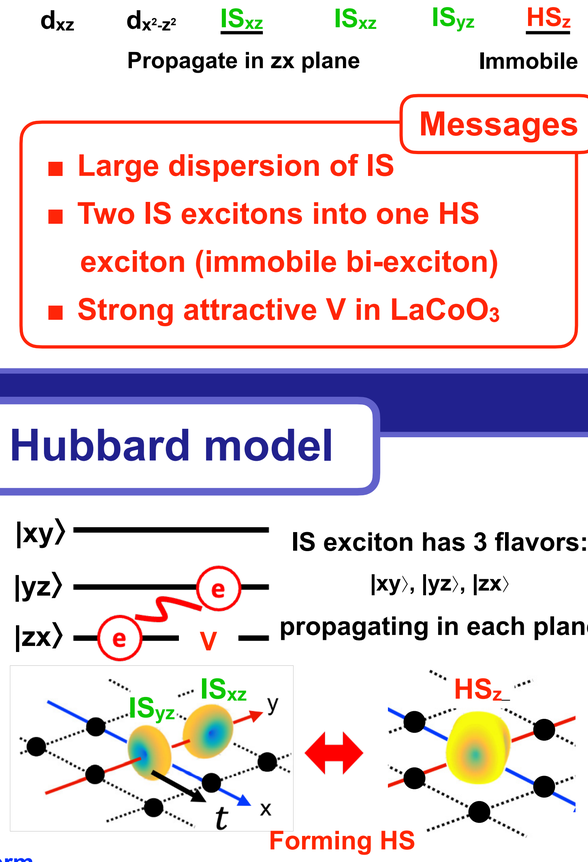
Attractive bosonic Hubbard model

$$\hat{H} = \epsilon_{IS} \sum_{\mathbf{i}} \sum_{\alpha} \hat{n}_{\mathbf{i},\alpha} - V \sum_{\mathbf{i}} \sum_{\alpha>\beta} \hat{n}_{\mathbf{i},\alpha}\hat{n}_{\mathbf{i},\beta}$$

On-site energy — Local attraction term

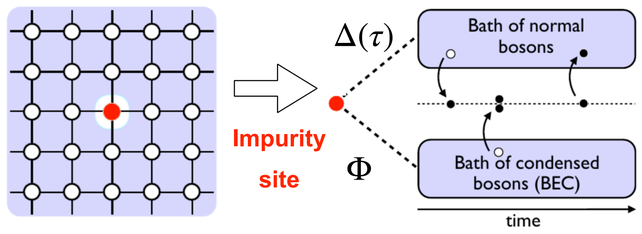
$$+t \sum_{\mathbf{i}} \sum_{\alpha,\beta} (1-\delta_{\alpha\beta}) \hat{b}^{\dagger}_{\mathbf{i}\pm\mathbf{e}_{\beta},\alpha} \hat{b}_{\mathbf{i},\alpha}$$

Flavor-diagonal and direction-dependent hopping term



Method: DMFT for hard-core bosons

BDMFT: Anderson impurity model



Hardcore constraint (HC)

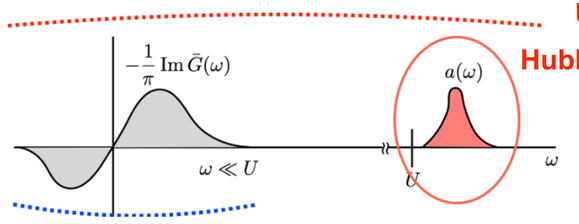
$$[b_i, b_j] = [b_i^{\dagger}, b_j^{\dagger}] = 0$$

$$[b_i, b_j^{\dagger}] = \delta_{ij}(1 - 2n_i)$$

Dynamic and kinematic HC constraint act in different Hilbert space

Broken sum rule of spectrum

$$-\frac{1}{\pi}\int_{-\infty}^{+\infty} \mathrm{Im}G(\omega)\mathrm{d}\omega = 1$$



$$-\frac{1}{\pi}\int_{-\infty}^{+\infty} \mathrm{Im}\bar{G}(\omega)\mathrm{d}\omega = \Gamma$$

Definition of rescaling factor Γ

$$\Gamma \equiv -\frac{1}{\pi}\int_{-\infty}^{\infty} d\omega \Im\bar{G}(\omega) = 1 - \int_{-\infty}^{\infty} d\omega a(\omega)$$

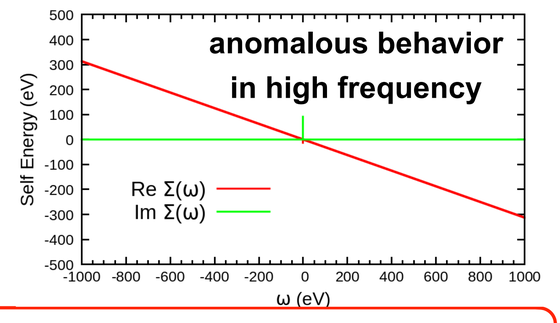
Green function for HC bosons

$$G(\omega) = \frac{1}{\omega - h_{\mathrm{loc}} - \Delta(\omega) - \Sigma(\omega)}$$

Break the sum rule

$$\bar{G}(\omega) = \frac{\Gamma}{\omega - h_{\mathrm{loc}} - \Delta(\omega) - \Sigma(\omega)}$$

Self energy (atomic model)



Messages

Problem on HB-DMFT:

- Self energy ill-defined with HC
- Numerical difficulty for achieving DMFT self-consistent condition

Solution for this problem:

- Use Dyson-Mori form of the GF
- Apply HC to the CT-QMC solver for auxiliary Anderson impurity model

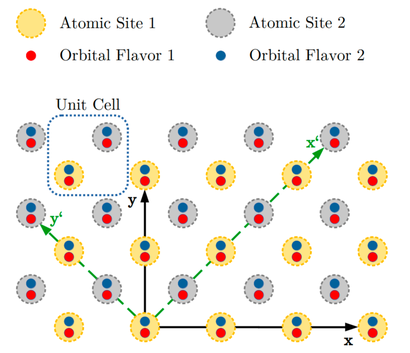
Result: calculation under spin-state order

2D 2-flavor Hubbard model

(Fixed: β = 40 eV^{-1}, t = 0.116 eV, ε_{IS} = 0.34 eV)

$$\hat{H} = \underbrace{\epsilon_{IS} \sum_{\mathbf{i}} \sum_{\alpha} \hat{n}_{\mathbf{i},\alpha}}_{\text{On-site energy}} \underbrace{+\frac{U}{2} \sum_{\mathbf{i}} \sum_{\alpha} \hat{n}_{\mathbf{i},\alpha}(\hat{n}_{\mathbf{i},\alpha} - 1)}_{\text{Dynamical HC constraint}}$$

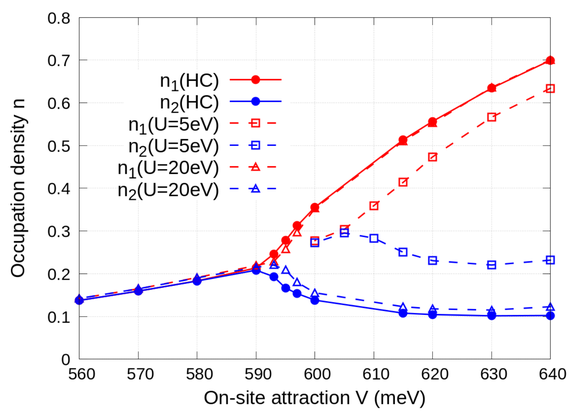
$$\underbrace{-V \sum_{\mathbf{i}} \hat{n}_{\mathbf{i},1}\hat{n}_{\mathbf{i},2}}_{\text{Local attraction term}} \underbrace{+t\left(\sum_{\mathbf{i},\delta_1} \hat{b}^{\dagger}_{\mathbf{i}\pm\delta_1,1}\hat{b}_{\mathbf{i},1} + \sum_{\mathbf{i},\delta_2} \hat{b}^{\dagger}_{\mathbf{i}\pm\delta_2,2}\hat{b}_{\mathbf{i},2}\right)}_{\text{Flavor-diagonal and direction-dependent hopping term}}$$



Question

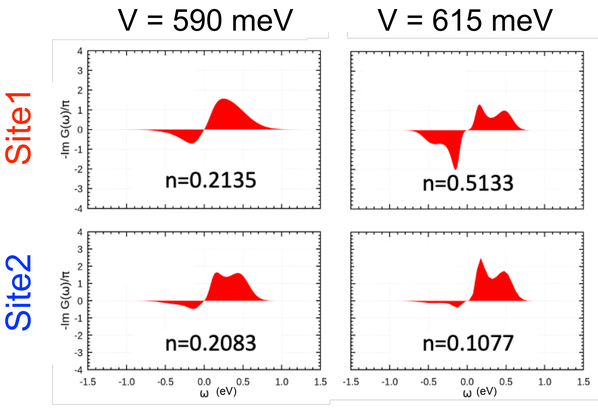
Spin-state order appears by the on-site attraction V (HS excitation)?

Occupation vs attraction V



- SSO appears spontaneously by V
- Explicit HC corresponds to U → ∞

Spectral density (with HC)



- Our methodology for HB-DMFT is implemented properly

Future work

Application of HB-DMFT to a realistic model of $LaCoO_3$ 1) including full IS degrees of freedom and 2) extracting parameters from ab-initio calculation