Resonant inelastic x-ray scattering spectra of the dynamic Jahn-Teller Cu^{2+} center in $CuAl_2O_4$

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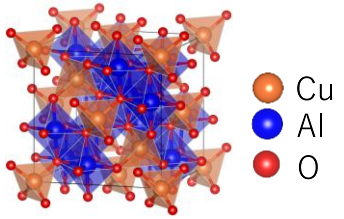
1. Magnetism of $CuAl_2O_4$

Magnetism of $CuAl_2O_4$:

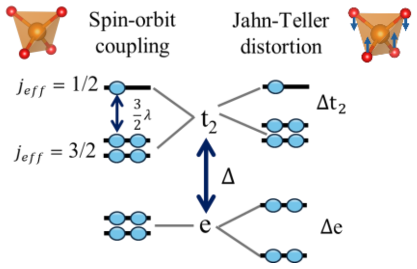
- Geometrically frustrated magnet
- Spin glass phase develops [1]

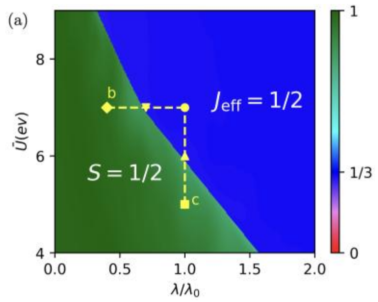


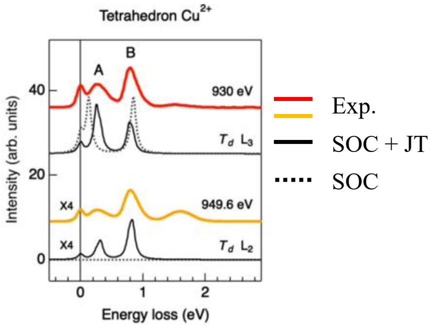
$CuAl_2O_4$

Electronic states of Cu^{2+} in CuO_4 tetrahedron:

Competition between the spin-orbit coupling (SOC) and Jahn-Teller (JT) interaction.







DFT+DMFT: $j_{eff} = ½$ SO coupled magnet [2]. The SOC suppresses the JT deformation.

Cu RIXS + LDA+U: SOC + nonnegligible JT deformation are necessary [3]. The latter disagrees with the x-ray diffraction data.

This work

Revealing the nature of the ground state of the Cu spinel magnet by reproducing the RIXS spectra based on the SOC + dynamic JT model.

2. Theory

Model Hamiltonian for a CuO_4 tetrahedron

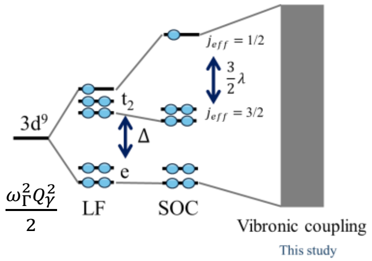
$$\hat{H} = \hat{H}_{LF} + \hat{H}_{SOC} + \hat{H}_0 + \hat{H}_{JT}$$

Ligand-field $\quad \hat{H}_{LF} = \Delta \hat{h}_{LF}$

SOC $\quad \hat{H}_{SOC} = -\lambda\, \hat{\boldsymbol{l}} \cdot \hat{\boldsymbol{s}}$

Harmonic oscillation $\quad \hat{H}_0 = \sum_\gamma -\frac{1}{2}\frac{\partial^2}{\partial Q_\gamma^2} + \frac{\omega_\Gamma^2 Q_\gamma^2}{2}$

Vibronic coupling



$$\hat{H}_{JT} = -V_E \begin{pmatrix} \hat{Q}_{Ez^2} & -\hat{Q}_{Ex^2-y^2} & 0 & 0 & 0 \\ -\hat{Q}_{Ex^2-y^2} & \hat{Q}_{Ez^2} & 0 & 0 & 0 \\ 0 & 0 & \frac{1}{2}\hat{Q}_{Ez^2} - \frac{\sqrt{3}}{2}\hat{Q}_{Ex^2-y^2} & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{2}\hat{Q}_{Ez^2} + \frac{\sqrt{3}}{2}\hat{Q}_{Ex^2-y^2} & 0 \\ 0 & 0 & 0 & 0 & -\hat{Q}_{Eu} \end{pmatrix} - V_T \begin{pmatrix} 0 & 0 & \frac{1}{2}\hat{Q}_{Tyz} & \frac{1}{2}\hat{Q}_{Tzx} & -\hat{Q}_{Txy} \\ 0 & 0 & -\frac{\sqrt{3}}{2}\hat{Q}_{Tyz} & \frac{\sqrt{3}}{2}\hat{Q}_{Tzx} & 0 \\ \frac{1}{2}\hat{Q}_{Tyz} & -\frac{\sqrt{3}}{2}\hat{Q}_{Tyz} & 0 & \frac{\sqrt{3}}{2}\hat{Q}_{Txy} & \frac{\sqrt{3}}{2}\hat{Q}_{Tzx} \\ \frac{1}{2}\hat{Q}_{Tzx} & \frac{\sqrt{3}}{2}\hat{Q}_{Tzx} & \frac{\sqrt{3}}{2}\hat{Q}_{Txy} & 0 & \frac{\sqrt{3}}{2}\hat{Q}_{Tyz} \\ -\hat{Q}_{Txy} & 0 & \frac{\sqrt{3}}{2}\hat{Q}_{Tzx} & \frac{\sqrt{3}}{2}\hat{Q}_{Tyz} & 0 \end{pmatrix}$$

$\times k$

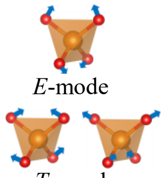
$\times k^2$

Q_γ Normal coordinates

g_Γ Vibronic coupling parameter

ω_Γ Frequencies ($\omega_E = 658\ \mathrm{cm}^{-1}, \omega_{T_2} = 419\ \mathrm{cm}^{-1}$) [4]

k: Reduction factor by d-p hybridization



Energy eigenstates of the model

$\hat{H}|\psi_\mu\rangle = E_\mu|\psi_\mu\rangle,$ ← Numerical diagonalization of our model H

$|\psi_\mu\rangle = \sum_k |\phi_k\rangle \otimes |\chi_{k\mu}\rangle,\quad |\chi_{k\mu}\rangle = \sum_{\boldsymbol{n}} |\boldsymbol{n}\rangle C_{n;k\mu}.$

Quantum entanglement of the spin-orbit and vibrational states

Cu L_2 and L_3 edge RIXS intensity [5]

$$\frac{d^2\sigma}{d\Omega dk} \propto \frac{\omega_I}{\omega_I}\left|\sum_X \frac{\left(\hat{\boldsymbol{p}}\cdot\epsilon_F^{(\alpha')}\right)FX\left(\hat{\boldsymbol{p}}\cdot\epsilon_I^{(\alpha)}\right)XI}{E_X - E_I - \hbar\omega_I - i\eta X}\right|^2 \delta(E_F + \hbar\omega_F - E_I - \hbar\omega_I)$$

$\hat{\boldsymbol{p}}$ Electron momentum operator

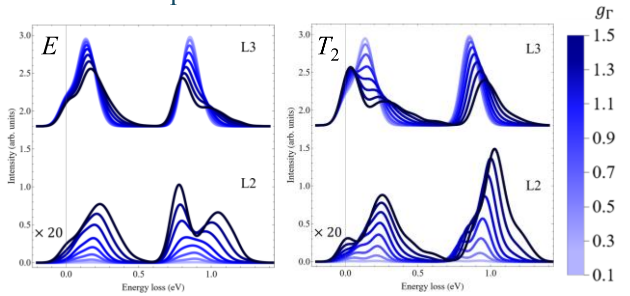
I Initial state } Energy eigenstates of our model H

F Final state

X Intermediate core-hole states

3. Results

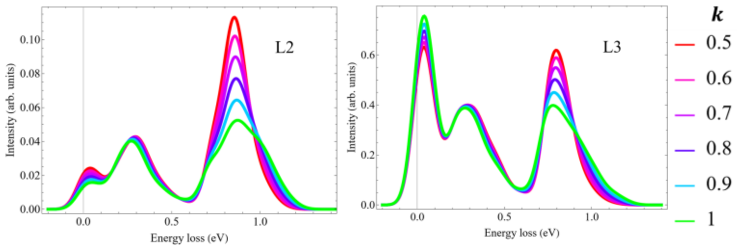
Simulated RIXS spectra



As g increases, new peaks emerge.

$\Delta = -0.72\ eV$

$\lambda = 0.1\ eV$



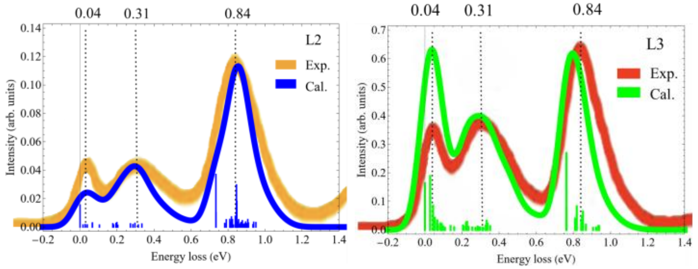
$\Delta = -0.57\ eV$

$\lambda = 0.1\ eV$

$g_E = 1.4$

$g_{T_2} = 1.4$

Comparison with Experimental RIXS spectra



$\Delta = -0.57\ eV$

$\lambda = 0.1\ eV$

$g_E = 1.4$

$g_{T_2} = 1.4$

$k = 0.5$

4. Conclusion

By concomitantly treating the SOC and dynamic JT effect, we reproduced the RIXS spectra of $CuAl_2O_4$. The compound is a $j_{eff} = ½$ spin-orbit coupled magnet.

References

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