Longitudinal magnon in the tetrahedral quantum spin system $Cu_2Te_2O_5Br_2$ near quantum criticality

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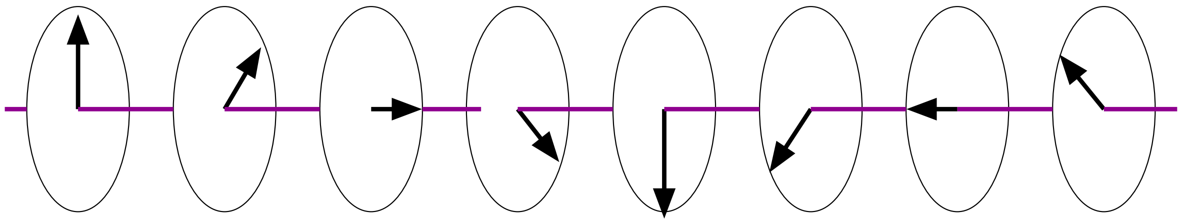
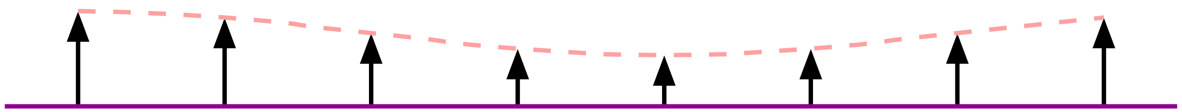
Lemmens *et al.*, PRL '01

Gros *et al*, PRB '03

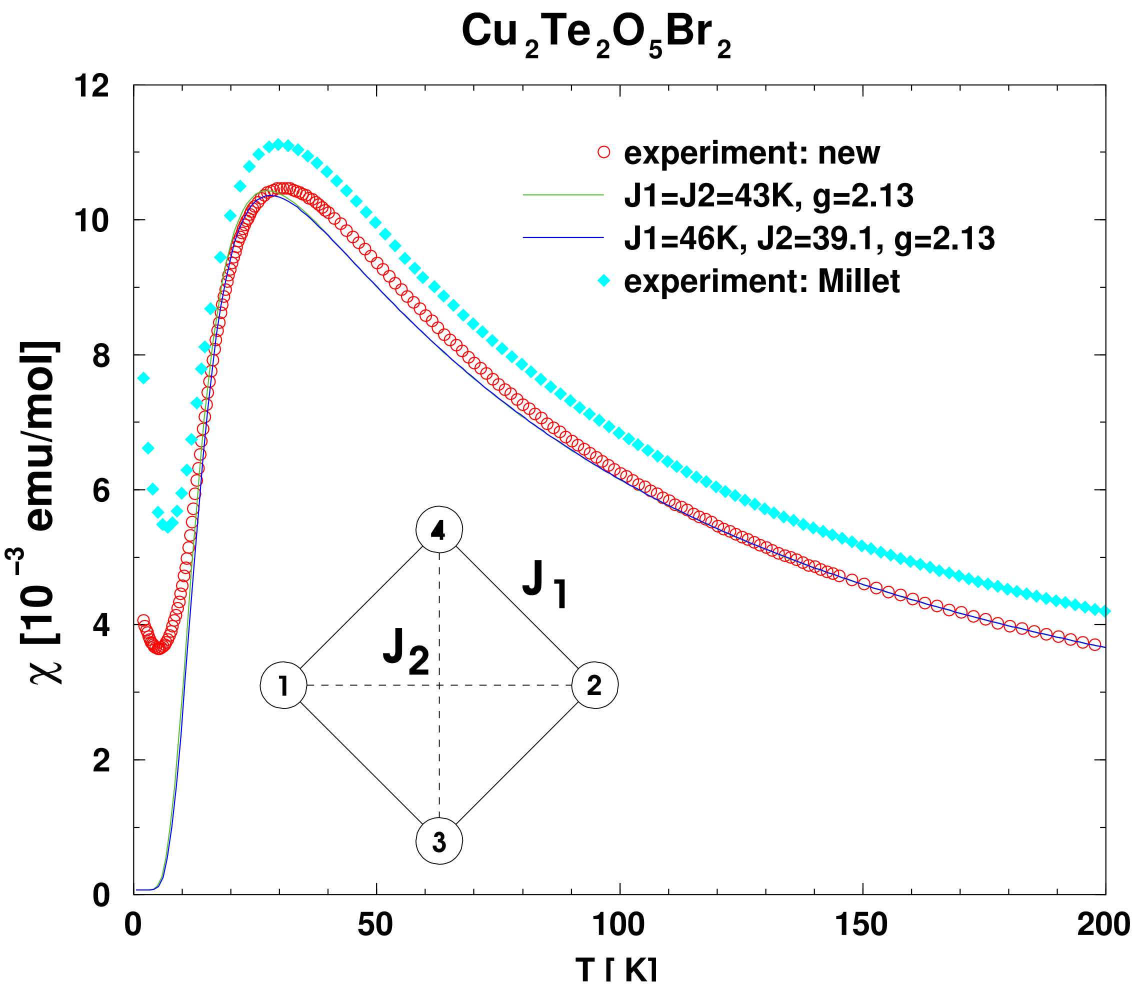


transversal magnon

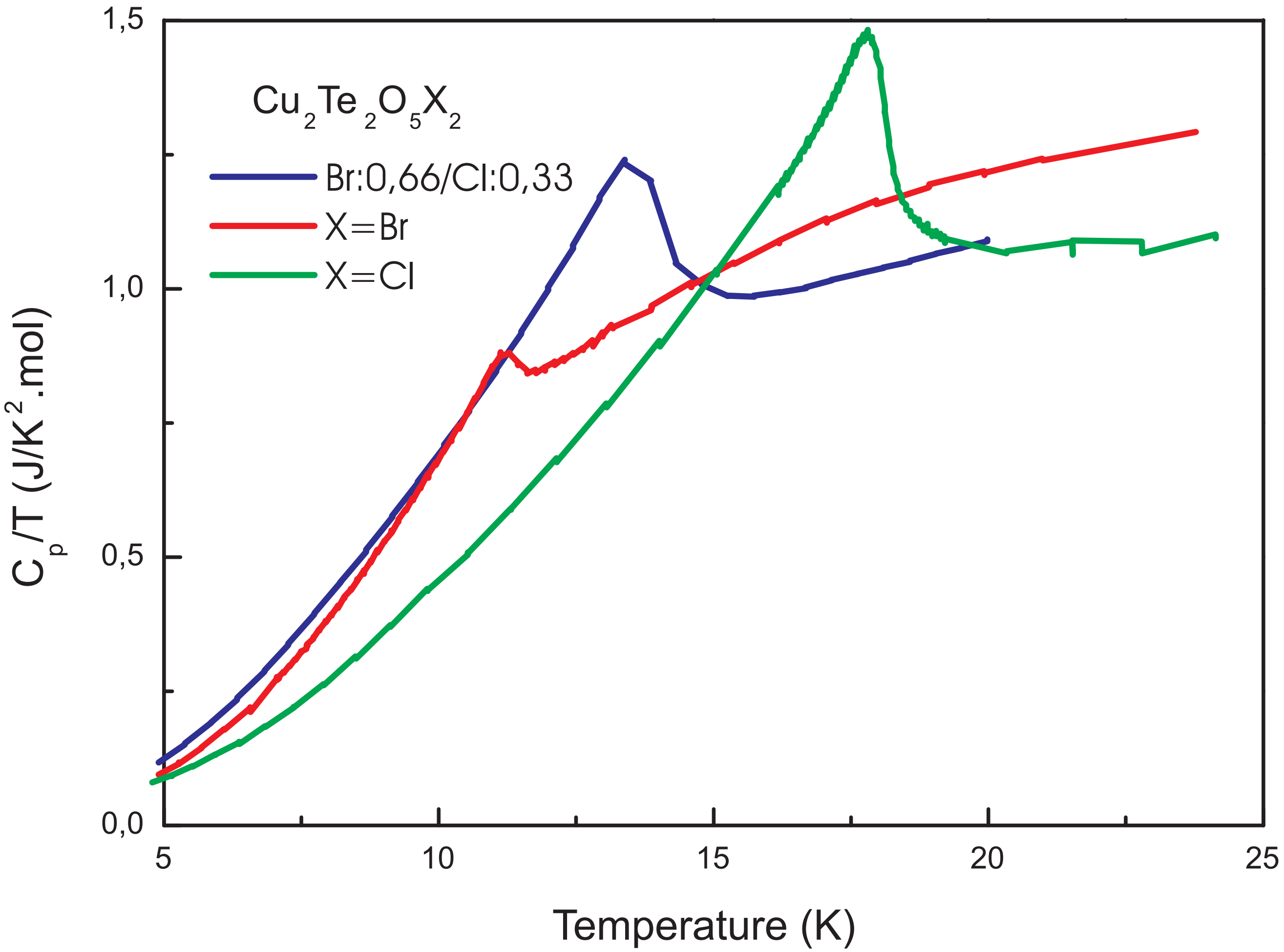


longitudinal magnon

Susceptibility of spin-1/2 tetrahedrons

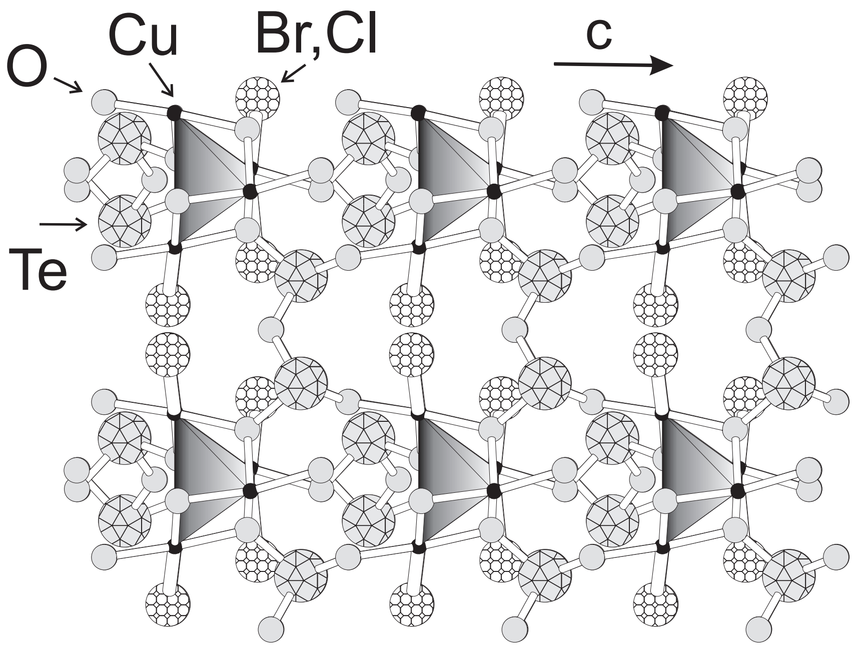


Phase transitions



- $T_N^{(Br)} = 11.4\,\mathrm{K}$
- $T_N^{(Cl)} = 18.2\,\mathrm{K}$
- $J \approx 40 - 46\,\mathrm{K}$

Coupled spin-1/2 tetrahedrons



decoupled: spin-singlet ground-state

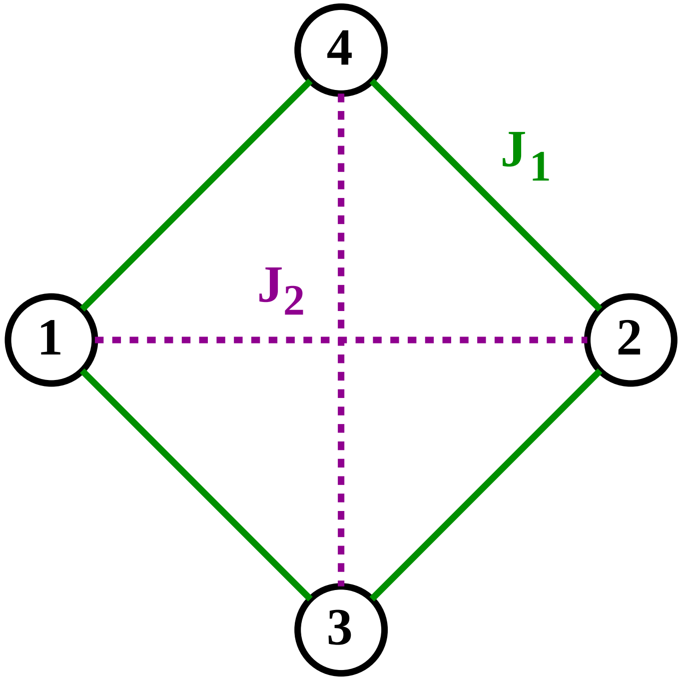
strongly coupled: antiferromagnet

$\Rightarrow$ $\boxed{\textbf{quantum-phase-transition}}$

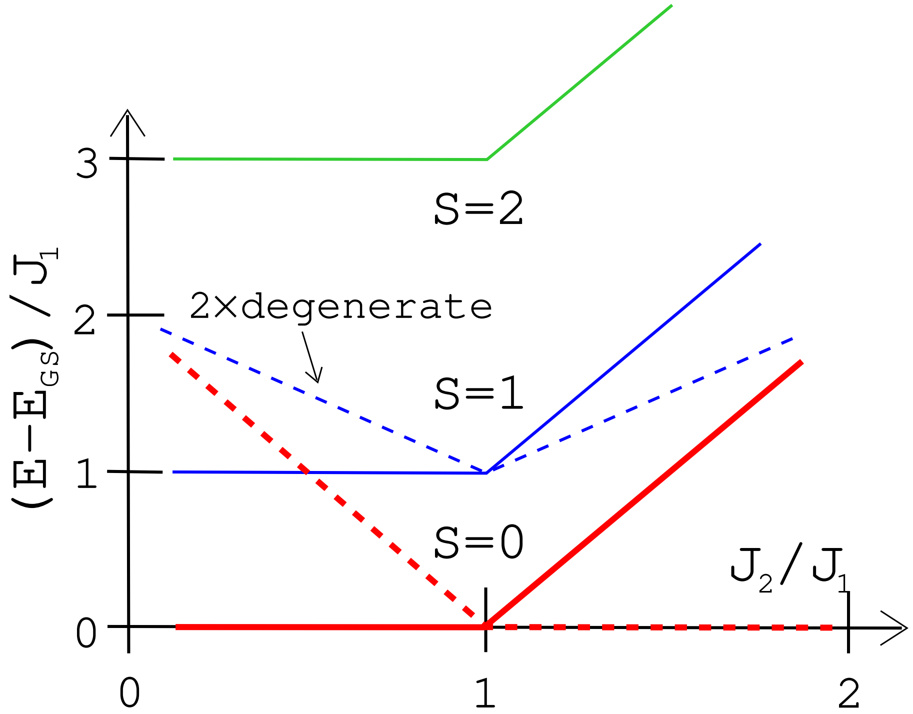
intra-tetrahedral states

$(J_1 < J_2)$

$$H_t = J_1\left[(\mathbf{S}_1+\mathbf{S}_2)\cdot(\mathbf{S}_3+\mathbf{S}_4)\right] + J_2\big(\mathbf{S}_1\cdot\mathbf{S}_2+\mathbf{S}_3\cdot\mathbf{S}_4\big)$$



- ground-state/excited singlet: ψ_{s1}, ψ_{s2}
- excited triplets: ψ^{α}_{t1}, ψ^{α}_{t2}, ψ^{α}_{t3}, $(\alpha = x, y, z)$
- excited quintuplet neglected

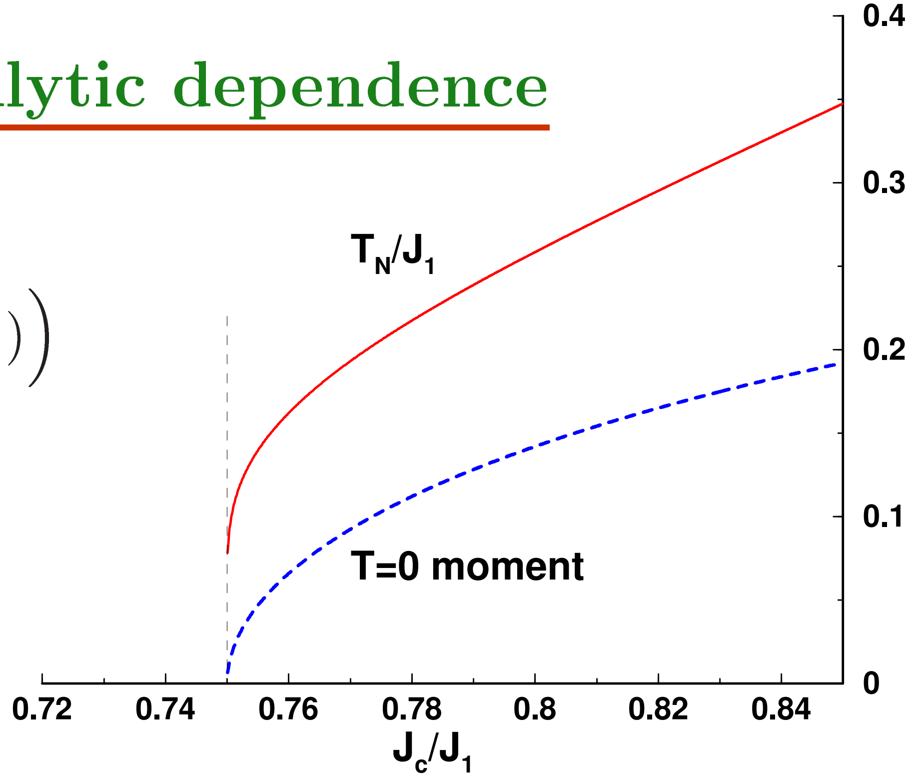


Molecular-field theory

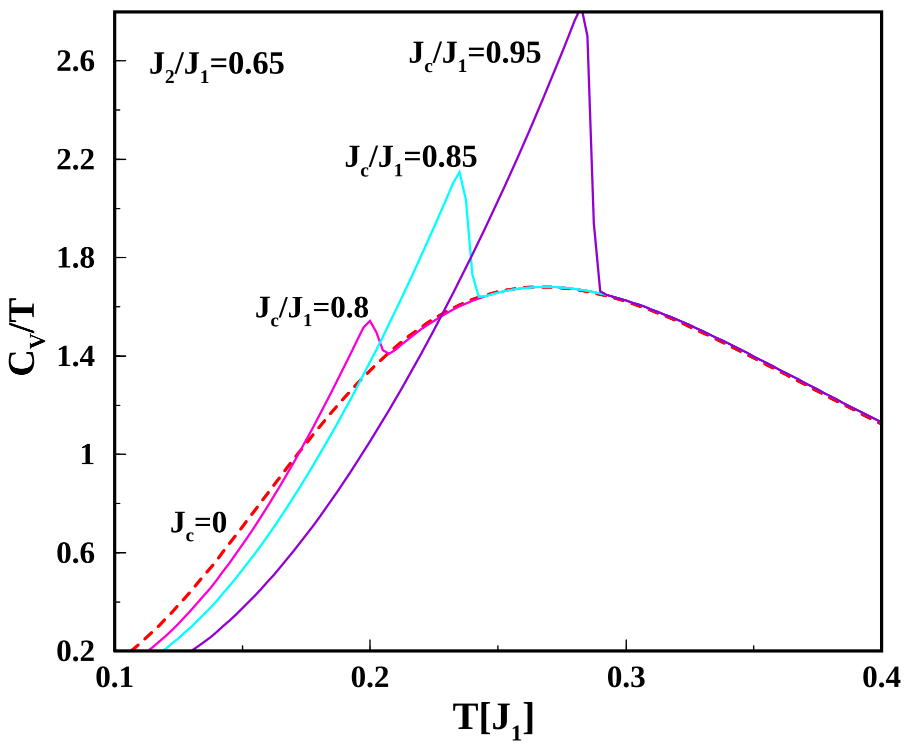
- molecular-field: $M = \langle S_1^z + S_2^z - S_3^z - S_4^z \rangle / 4$
- hamiltonian: $H_{MF} = H_t \; - \; J_c M \left(S_1^z + S_2^z - S_3^z - S_4^z \right)$
- quantum-phase transition: $\boxed{J_c^{(qc)} = 0.75 J_1}$

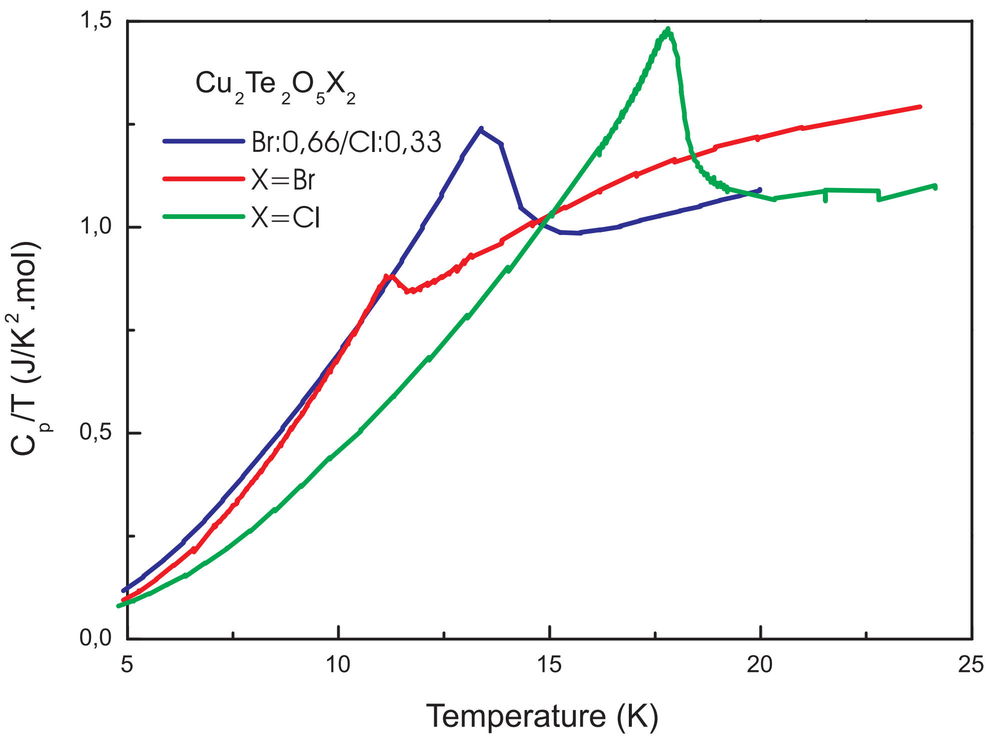
Néel-temperature: non-analytic dependence

$$T_N \simeq \Delta E_{s2} \log^{-1} \left(J_c^{(qc)} / (J_c - J_c^{(qc)}) \right)$$

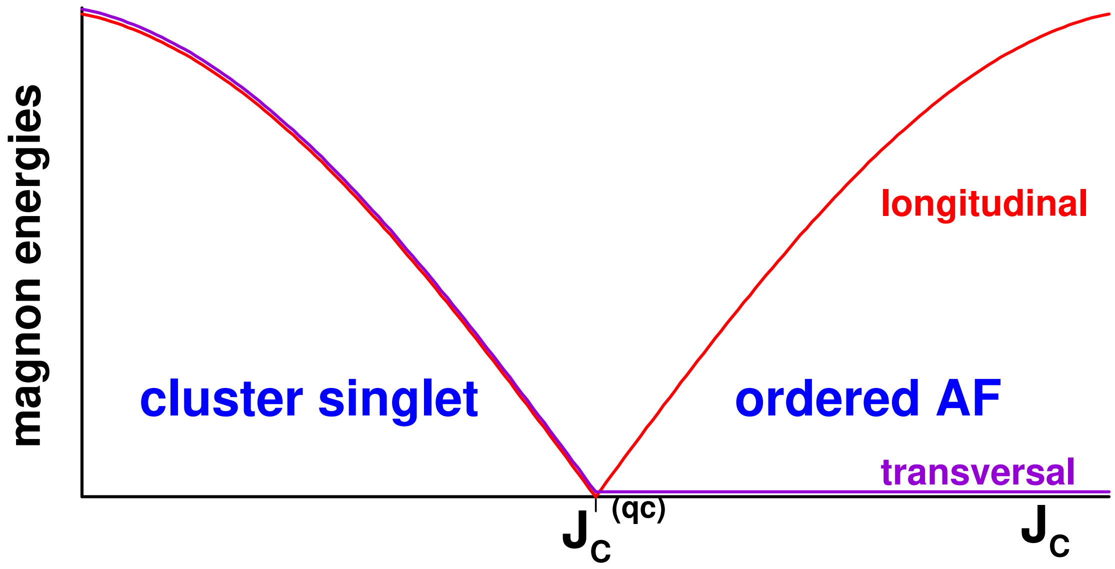


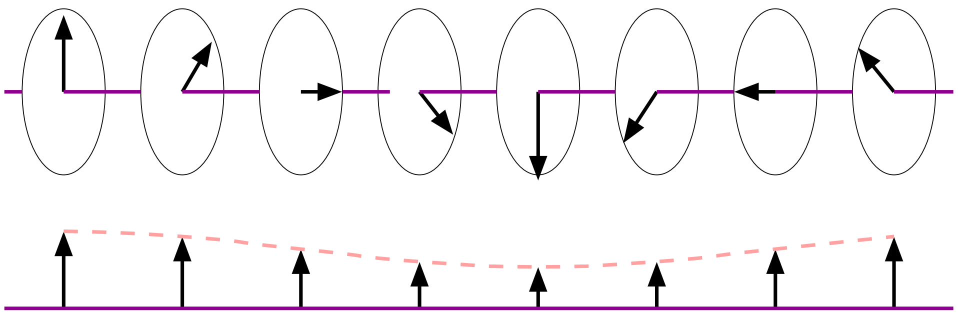
Phase transition near criticality





Transversal and longitudinal magnons



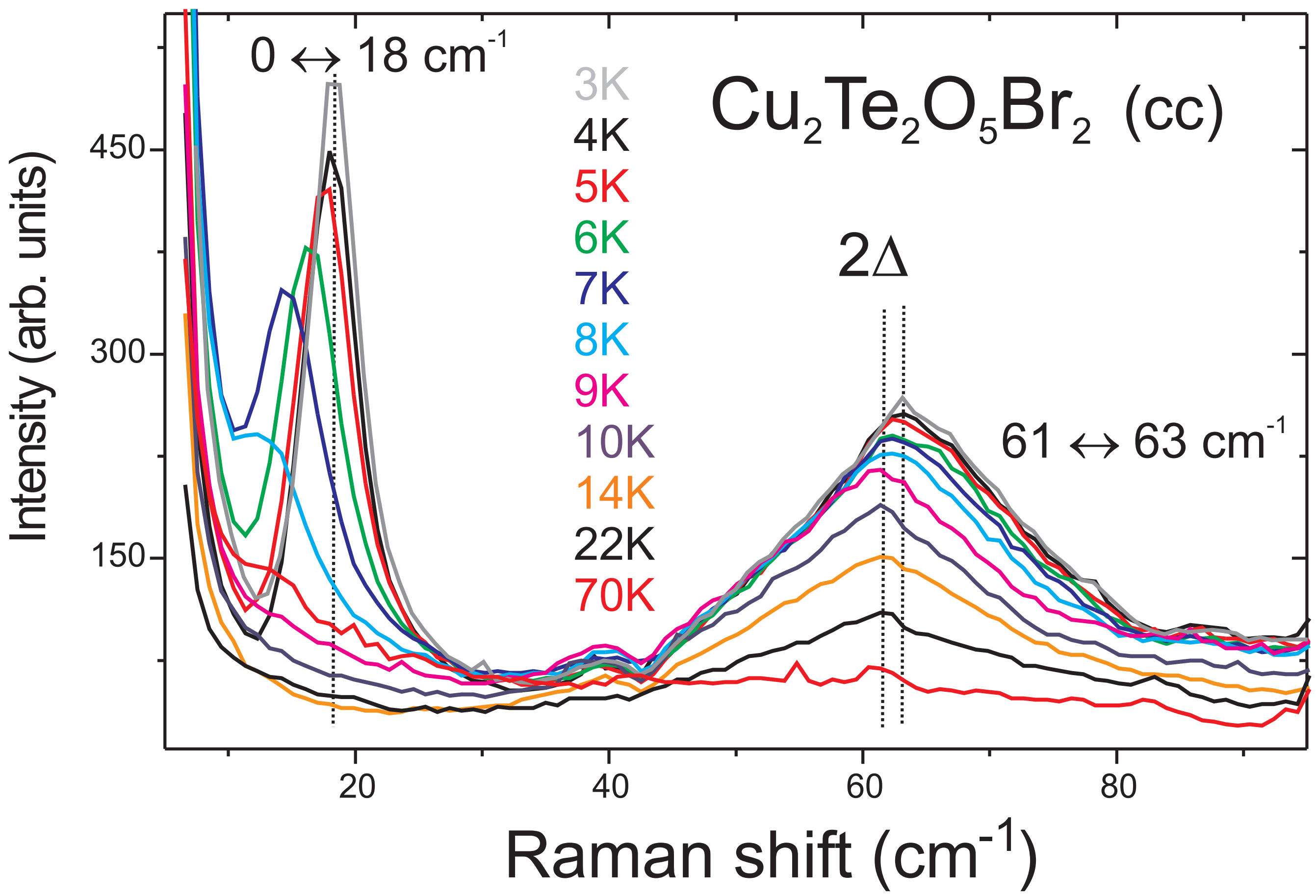


transversal magnon

longitudinal magnon

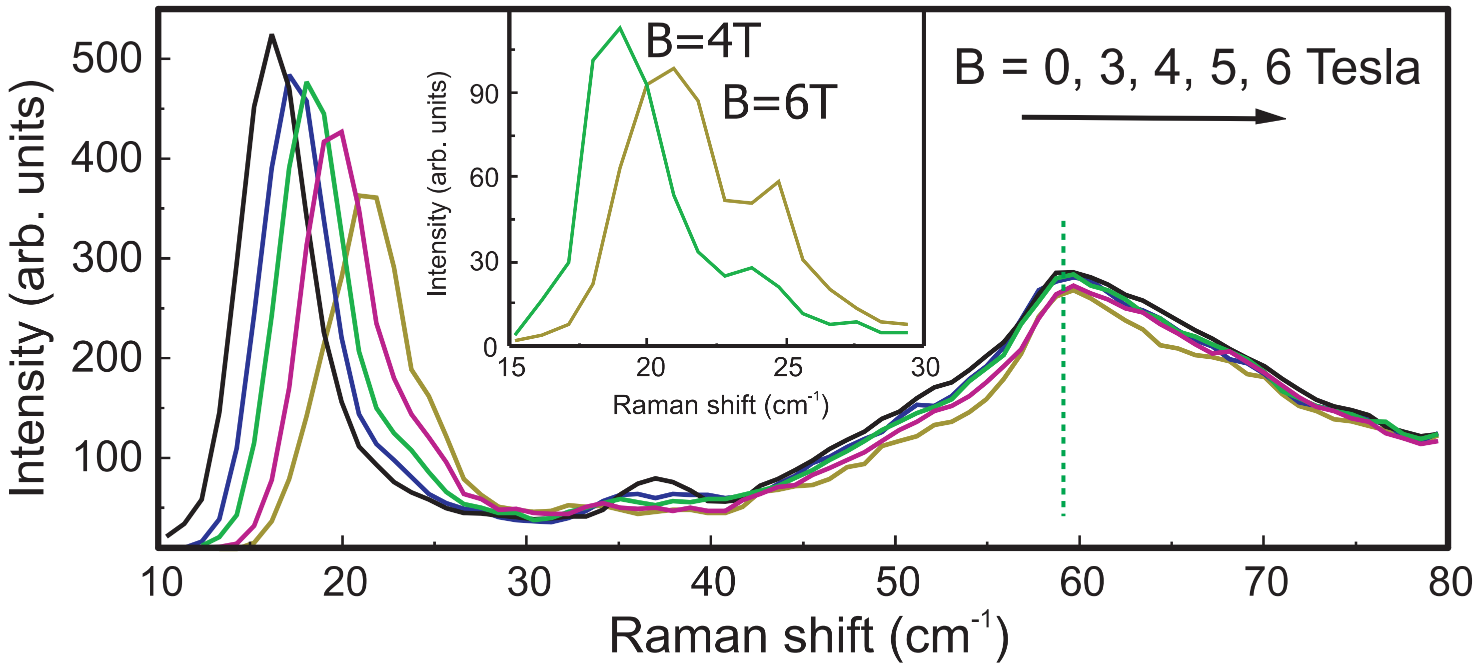
longitudinal magnon becomes soft at quantum-critical point

Raman-scattering



soft mode at $0 \leftrightarrow 18\,\mathrm{cm}^{-1}$: longitudinal magnon

Excited singlet



- ◇ longitudinal magnon shifts with external field
- ◇ new mode at $23.2\,\mathrm{cm}^{-1}$ does not shift

$\Rightarrow$ excited intra-tetrahedral singlet

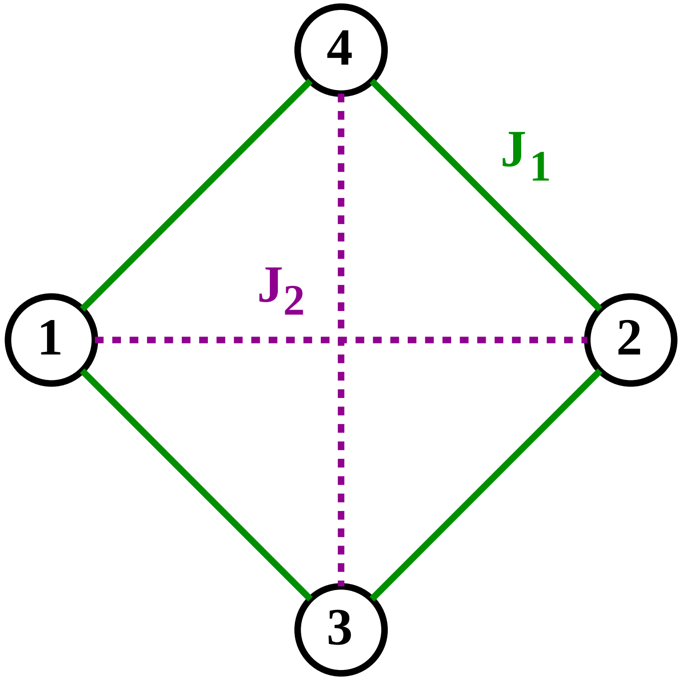
$\Rightarrow$ Parameters: $J_1 \sim 47\,\mathrm{K}, \quad J_2/J_1 \sim 0.66, \quad J_c/J_1 \sim 0.85$

Intra-tetrahedral states

$(J_1 < J_2)$

$$H_t = J_1\left[(\mathbf{S}_1 + \mathbf{S}_2)\cdot(\mathbf{S}_3 + \mathbf{S}_4)\right]$$

$$+ J_2\left(\mathbf{S}_1\cdot\mathbf{S}_2 + \mathbf{S}_3\cdot\mathbf{S}_4\right)$$



◇ ground-state/excited singlet: ψ_{s1}, ψ_{s2}

$$\psi_{s1} = \frac{-1}{\sqrt{3}}\left[t^0_{12}t^0_{34} - t^+_{12}t^-_{34} - t^-_{12}t^+_{34}\right] \;, \qquad \psi_{s2} = s_{12}s_{34}$$

◇ excited triplets: ψ^α_{t1}, ψ^α_{t2}, ψ^α_{t3}, $(\alpha = x, y, z)$

$$\psi^0_{t1} = \frac{1}{\sqrt{2}}\left[t^+_{12}t^-_{34} - t^-_{12}t^+_{34}\right], \qquad \psi^\alpha_{t2} = s_{12}t^\alpha_{34}, \qquad \psi^\alpha_{t2} = t^\alpha_{12}s_{12}$$

Longitudinal magnon

- supperposition of ground-state singlet
  and excited triplet

$$
\begin{aligned}
|\varphi\rangle &= \cos\varphi\,|\psi_{s1}\rangle + \sin\varphi\,|\psi^0_{t1}\rangle && \text{ground-state (with moment)} \\
|\tilde{\varphi}\rangle &= \sin\varphi\,|\psi_{s1}\rangle - \cos\varphi\,|\psi^0_{t1}\rangle && \text{longitudinal mangon}
\end{aligned}
$$

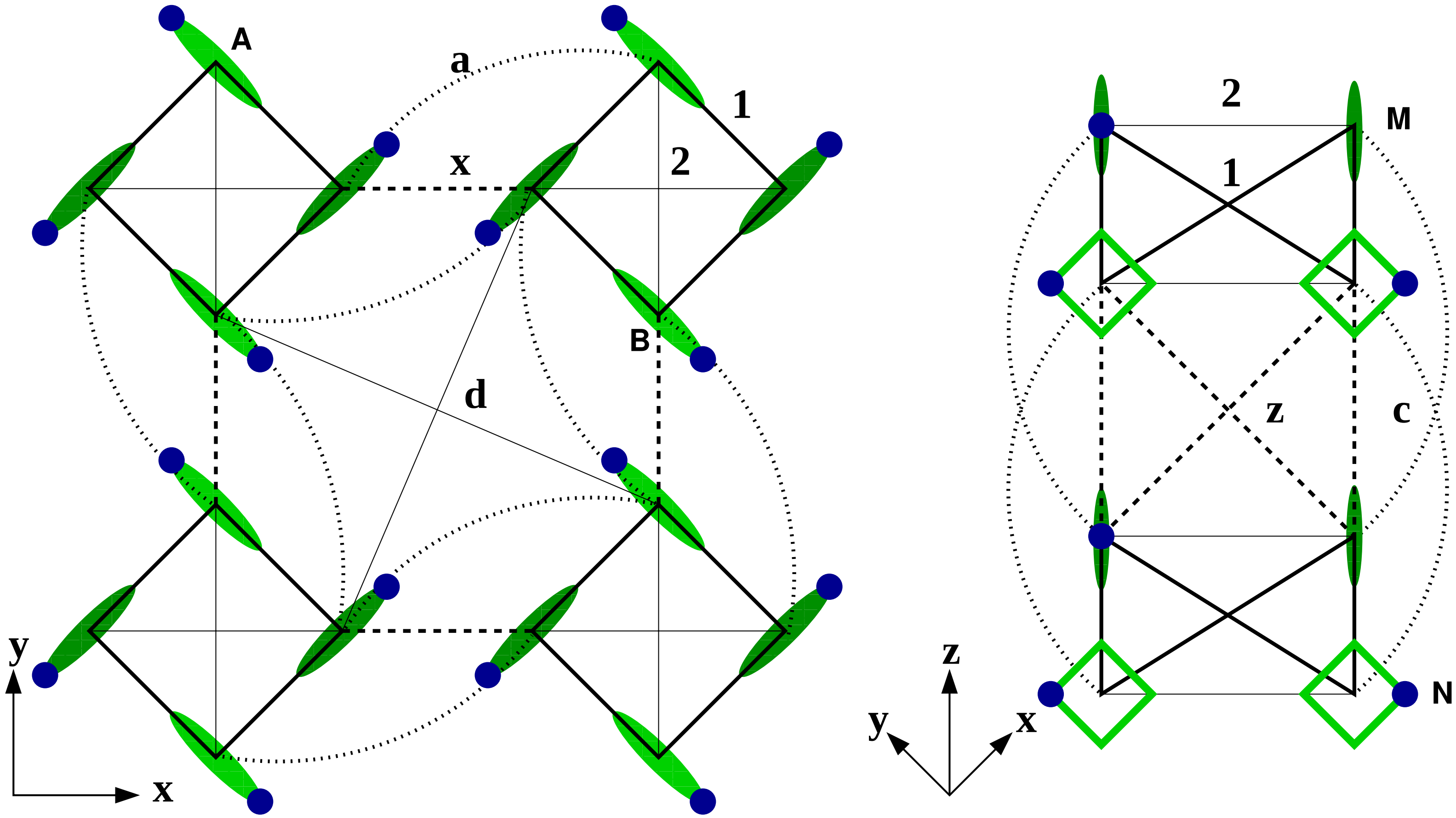
- eigen-energies

$$\Delta E_{\varphi,\tilde{\varphi}} = \frac{J_1}{2}\left[1 \mp \sqrt{1 + 32M^2J_c^2/(3J_1^2)}\right]$$

- self-consistency condition

$$M = \frac{e^{-\beta E_\varphi} - e^{-\beta E_{\tilde{\varphi}}}}{Z}\sqrt{\frac{2}{3}}\frac{\tan\varphi}{1+\tan^2\varphi}$$

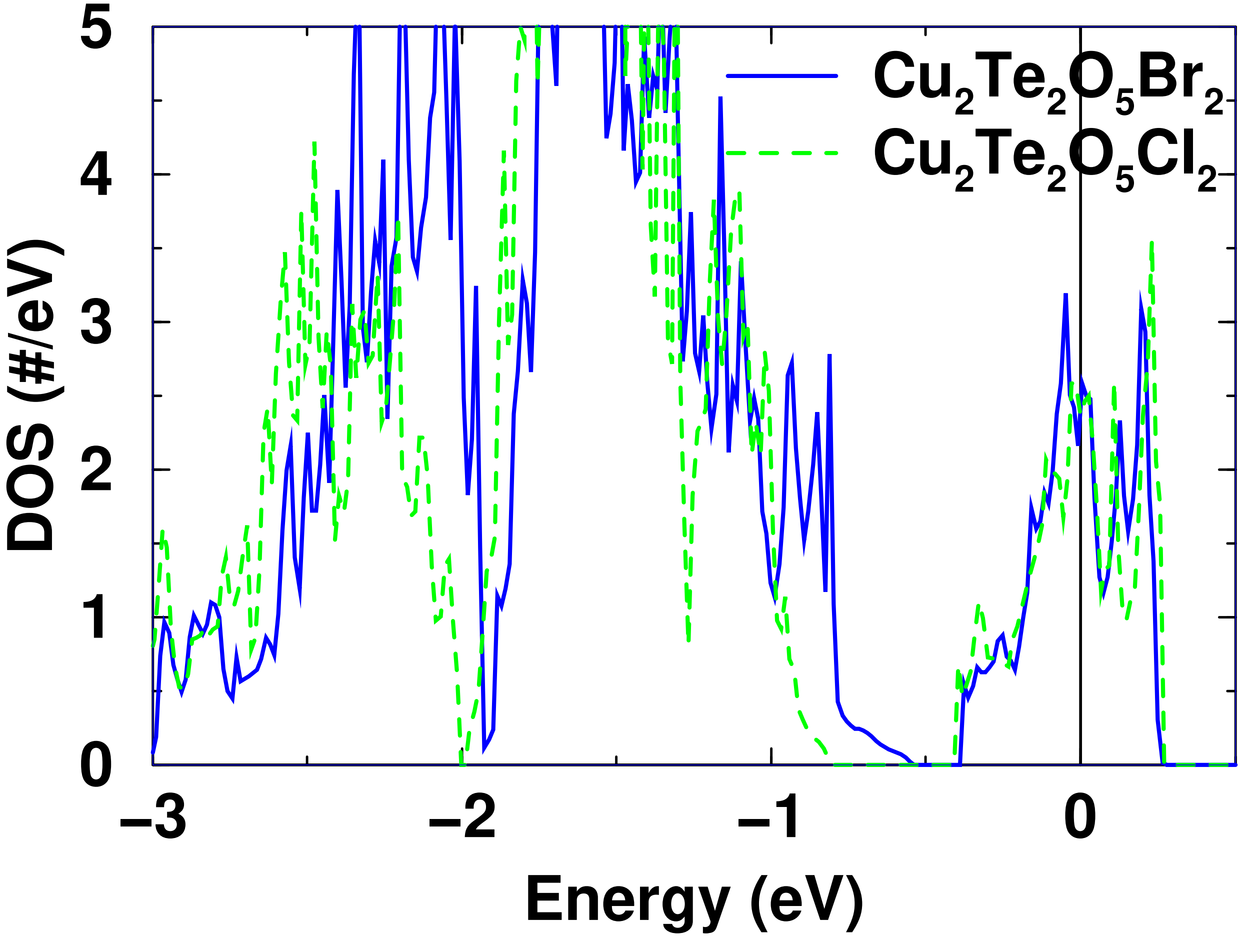
Schematic crystal structure



◇ important kinetic exchange pathes

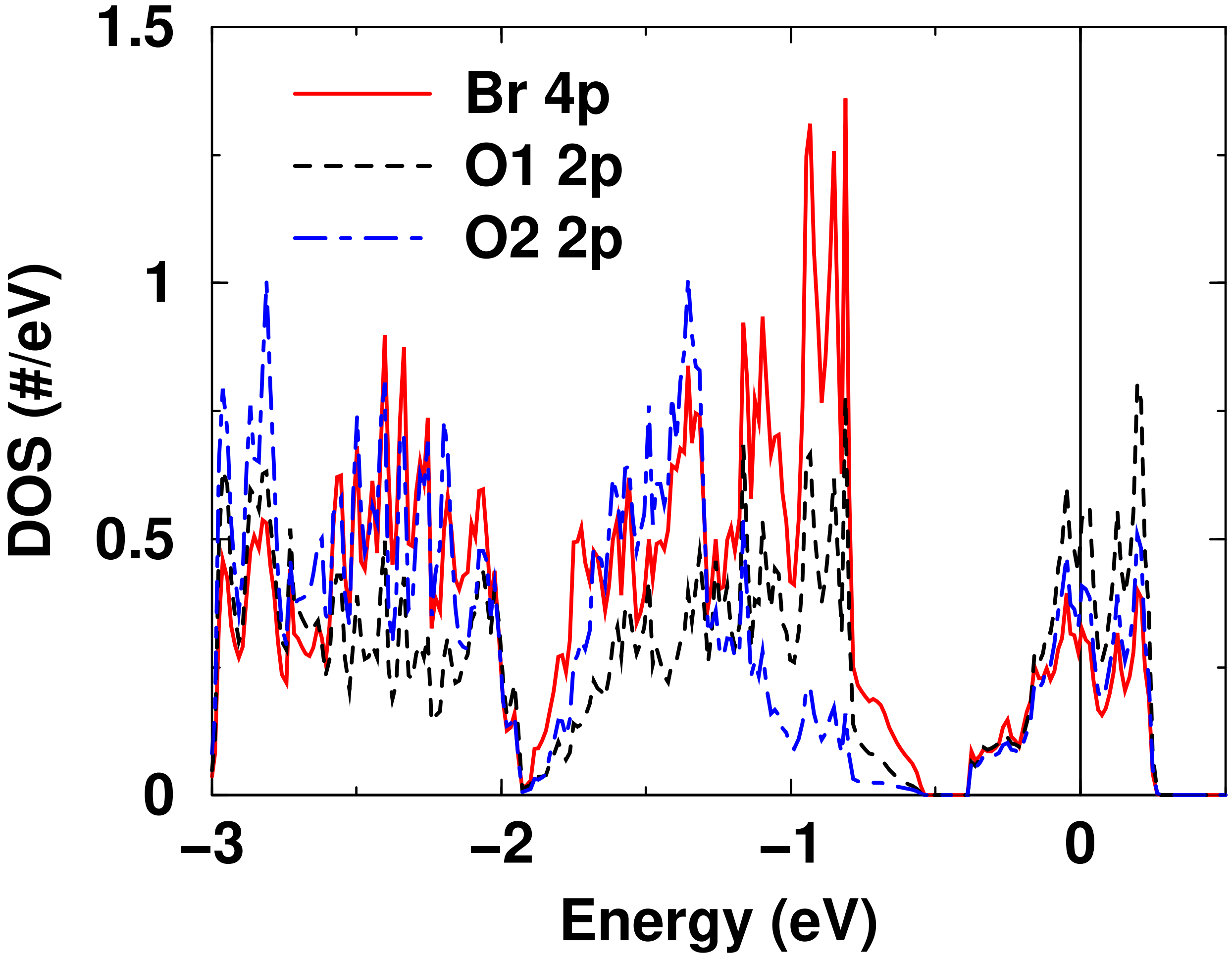
Cu-3d density of states

[LAPW,LMTO]



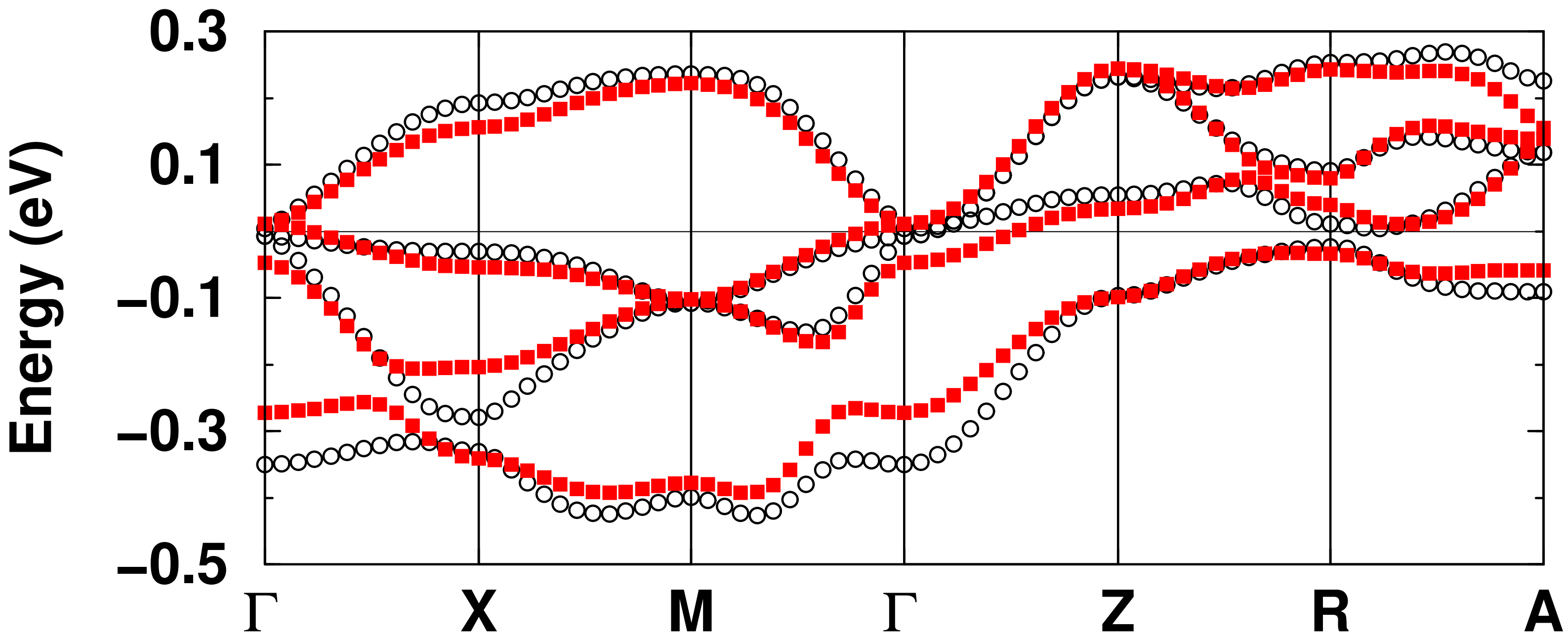
Oxygen density of states

[LAPW,LMTO]



Tight-binding fit

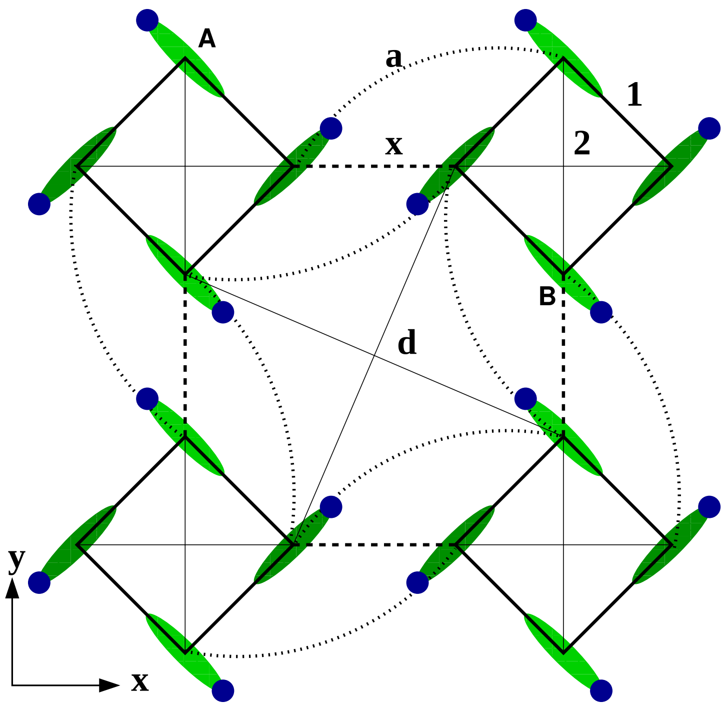
[downfolding]

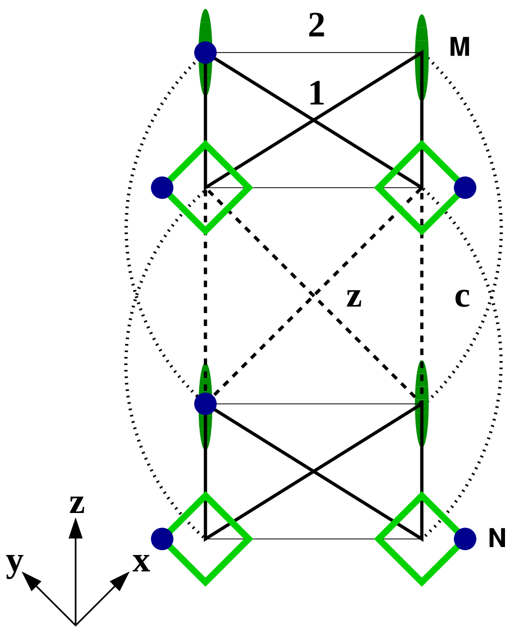


- ◇ four Cu-ions per unit cell
- ◇ half-filled + U ⇒ insulator

Downfolding parameters

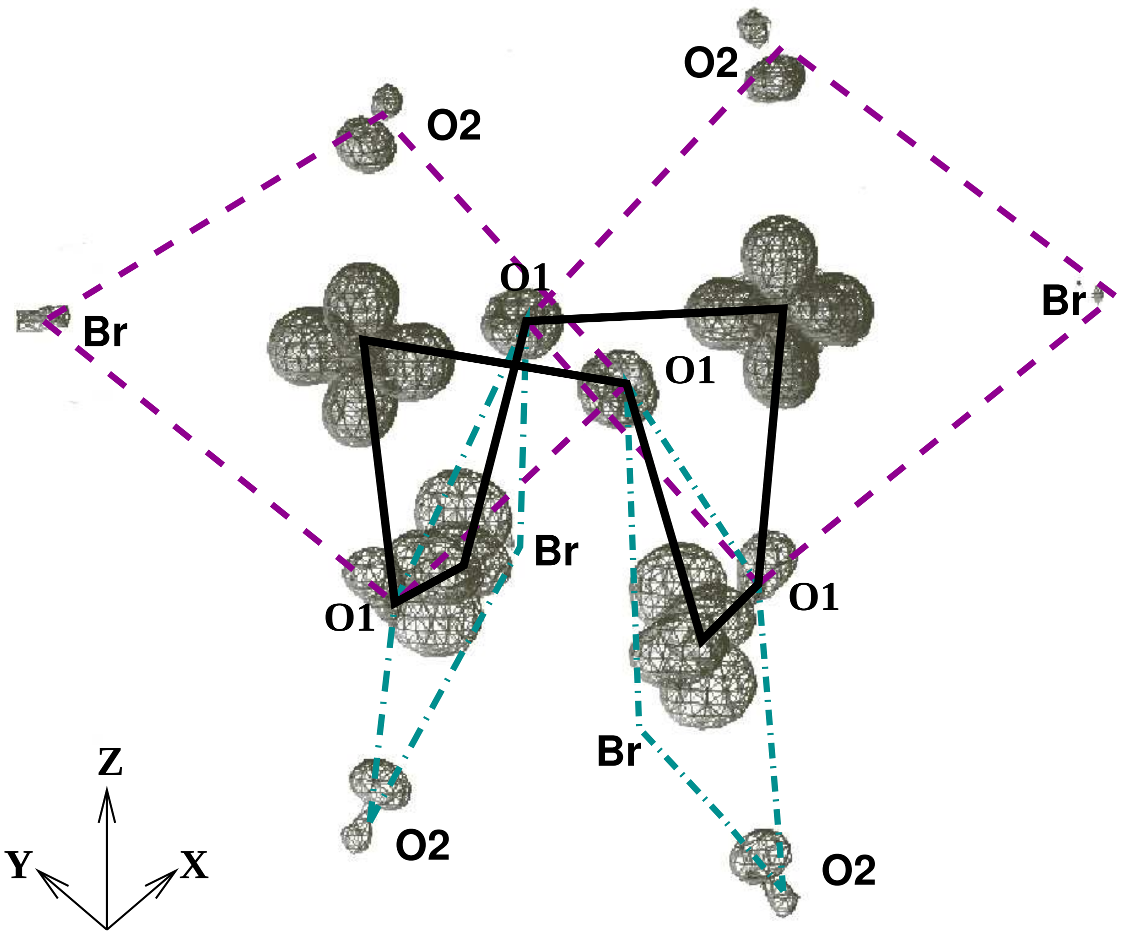
	Cu_{Br}	Cu_{Cl}		Cu+Br	Cu+Cl
t_1	80	98	t_1	155	181
t_2	4	0	t_2	-156	-132
t_x	-16	-10	t_x	-10	-14
t_z	11	12	t_z	34	33
t_a	-30	-29	t_a	5	8
t_c	-48	-45	t_c	-26	-19
t_v	-24	-23	t_v	9	8
t_d	-73	-80	t_d	8	8
t_r	-35	-48	t_r	-62	-72





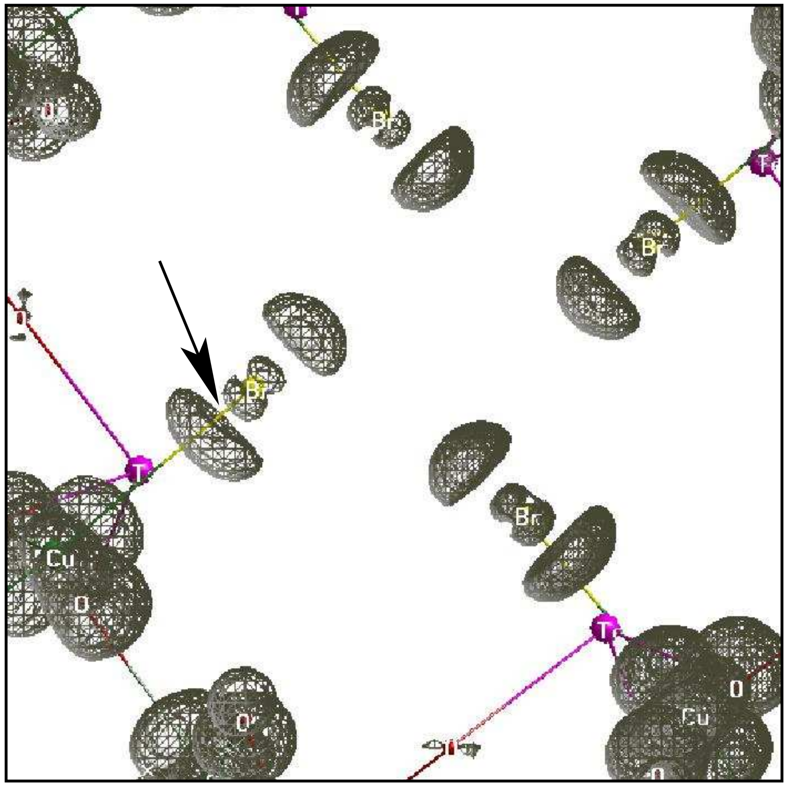
Position of Cu-3d orbital

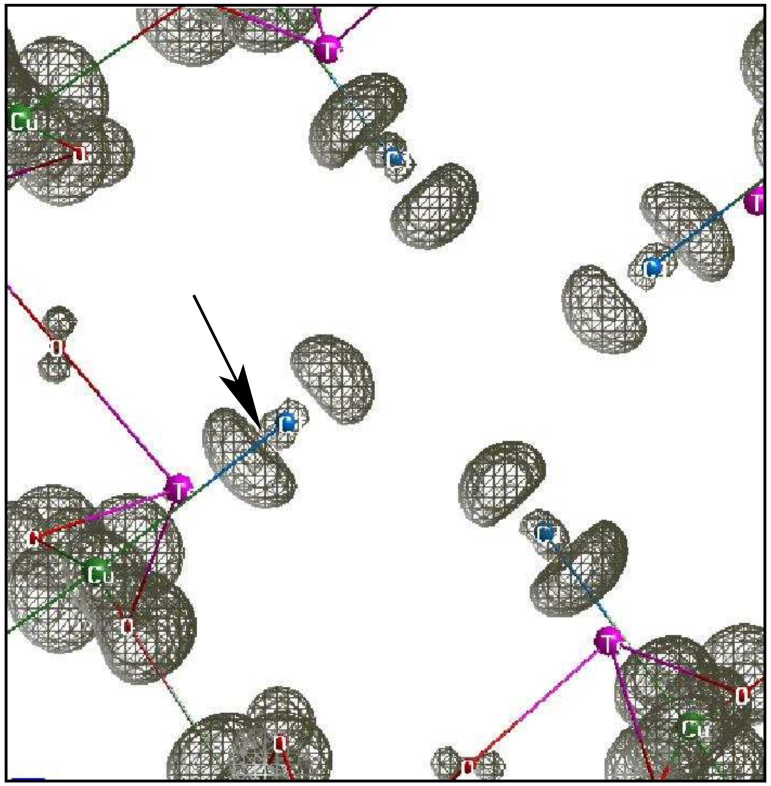
[electron density]



Position of Cl-3p/Br-4p orbital

[electron density]





◇ strong halogen-Cu hybridization

⇒ halogen-ions mediate Cu-Cu exchange