The interacting spin-1/2 tetrahedral system $Cu_2Te_2O_5X_2$ (X = Br, Cl). A mean-field/random phase approximation analysis

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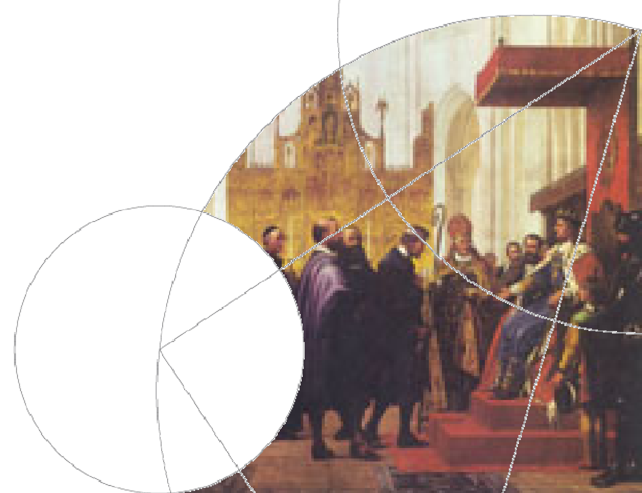
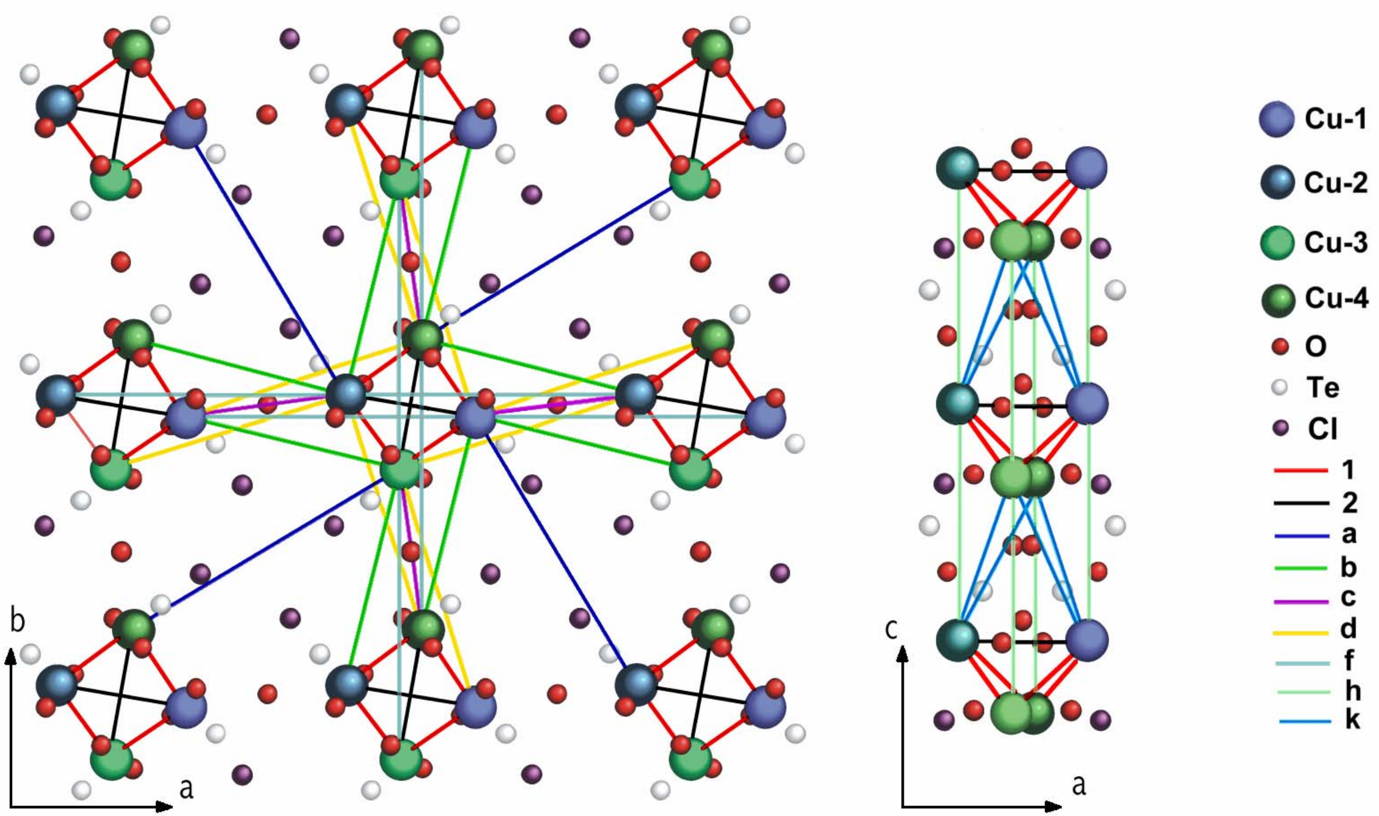


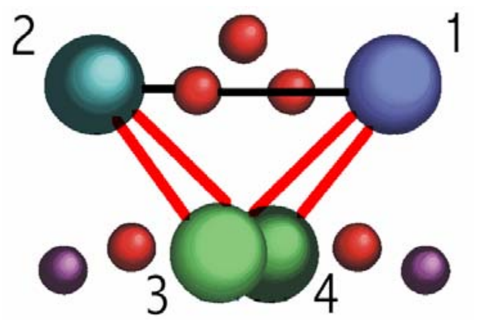
Crystal structure of $Cu_2Te_2O_5Cl_2$

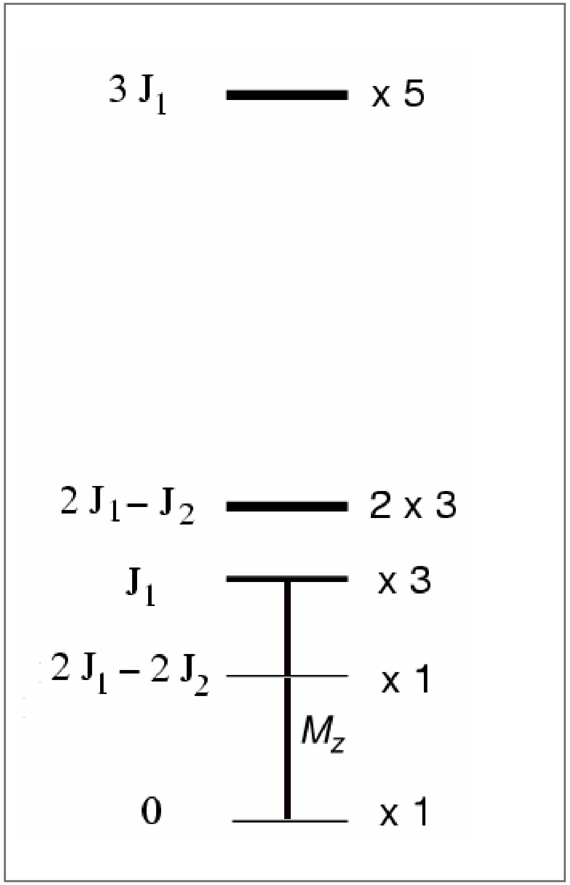


The $Cu_2Te_2O_5Cl_2$ crystal projected on the *ab* and *ac* planes.

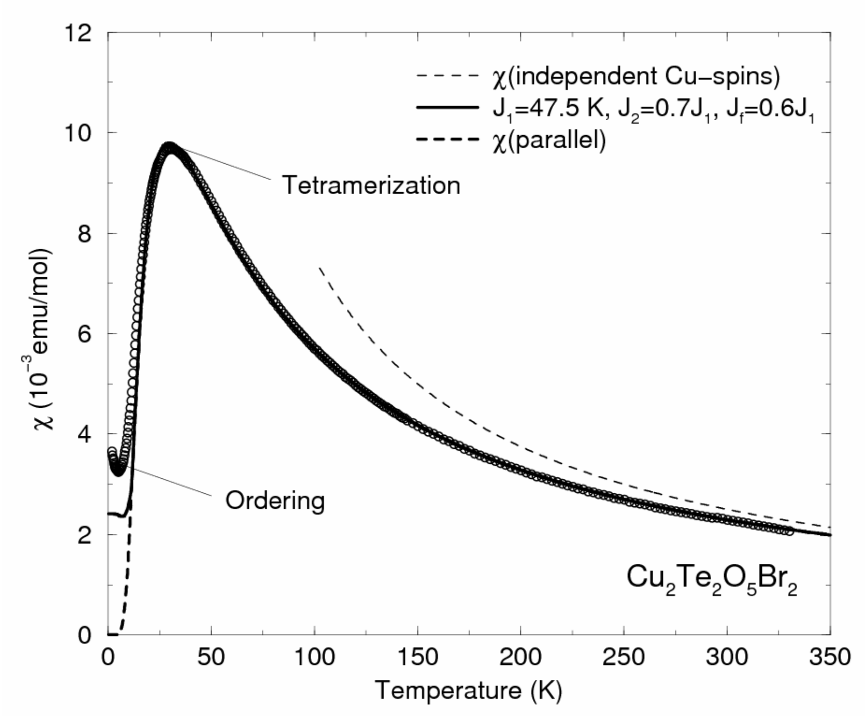
M. Johnsson, K.W. Törnroos, F. Mila, and P. Millet, Chem. Mater. **12**, 2853 (2000).

The tetrahedral unit





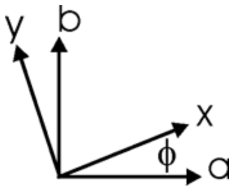
$$\mathcal{H}_t = J_1 \left(\mathbf{S}_1 + \mathbf{S}_2\right) \cdot \left(\mathbf{S}_3 + \mathbf{S}_4\right) + J_2 \left(\mathbf{S}_1 \cdot \mathbf{S}_2 + \mathbf{S}_3 \cdot \mathbf{S}_4\right)$$

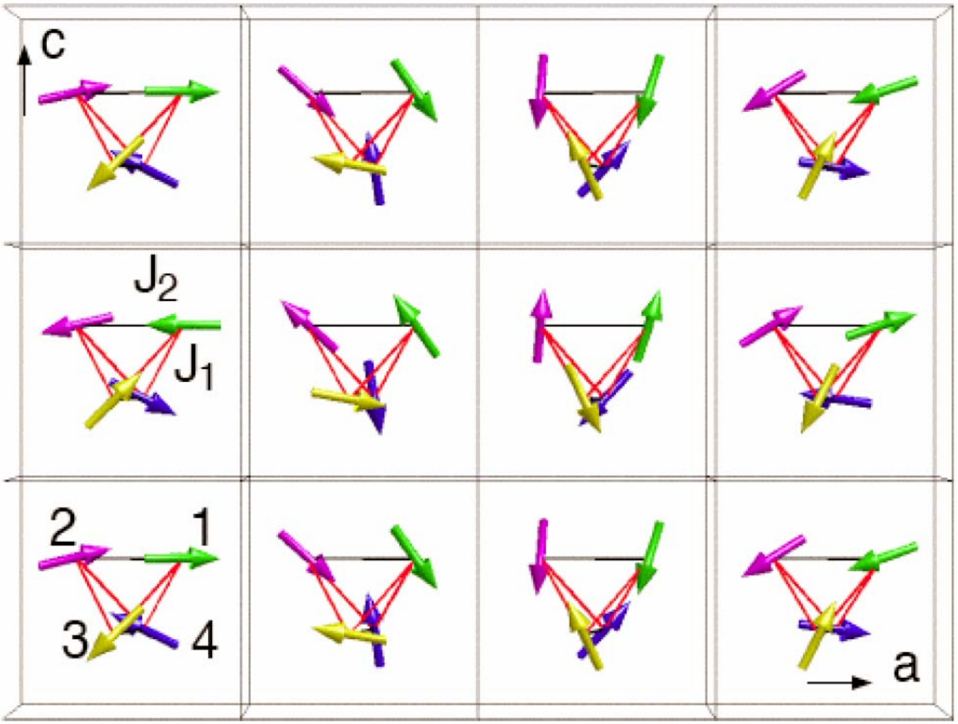


$$M_z = S_{1z} + S_{2z} - S_{3z} - S_{4z}$$

Magnetic ordering of $Cu_2Te_2O_5X_2$: Tilted helix

$$\langle \mathbf{S}_{\alpha i} \rangle = S_0 \left[\hat{\mathbf{x}} \cos(\mathbf{Q} \cdot \mathbf{R}_i + \psi_\alpha) + \hat{\mathbf{z}} \sin(\mathbf{Q} \cdot \mathbf{R}_i + \psi_\alpha) \right]$$





Structure at 2 K:

X = Cl, $T_N = 18.2$ K
$\mathbf{Q} = (-0.150, 0.422, 0.5)$
$S_0 = 0.44,\ \phi = 14°$
$\psi_2 = 13°, \psi_3 = -136°, \psi_4 = 154°$

X = Br, $T_N = 11.4$ K
$\mathbf{Q} = (-0.172, 0.356, 0.5)$
$S_0 = 0.20,\ \phi = 9°$
$\psi_2 = 22°, \psi_3 = -105°, \psi_4 = 134°$

Polarized and unpolarized neutron diffraction experiments:
O. Zaharko, H. Rønnow, J. Mesot, S. J. Crowe, D. McK. Paul, P. J. Brown, A. Daoud-Aladine, A. Meents, A. Wagner, M. Prester, and H. Berger, Phys. Rev. B **73**, 064422 (2006).

Mean-field model

$$\mathcal{H}_{\text{intra}} + \mathcal{H}_{\text{inter}} = \sum_i \left[\mathcal{H}(J_1, J_2) + \mathbf{D}_{\alpha\beta} \cdot \mathbf{S}_\alpha \times \mathbf{S}_\beta\right] + \frac{1}{2} \sum_{\alpha i, \beta j} J_{ij}^{\alpha\beta} \mathbf{S}_{\alpha i} \cdot \mathbf{S}_{\beta j}$$

Cl: $J_1 = 40.9$ K, $J_2 = J_1$, $D_1 = 0.03 J_1$, $\theta_D = 55.6°$

Br: $J_1 = 47.5$ K, $J_2 = 0.7 J_1$, $D_1 = 0.03 J_1$, $\theta_D = 33.2°$

	J_a/J_1	J_b/J_1	J_c/J_1	J_d/J_1	J_f/J_1	J_h/J_1	J_k/J_1
Cl:	0.8	−0.1	−0.1	−0.3	−0.03	0.1	−0.42
Br:	0.17	0.33	0.1	−0.35	−0.1	0.1	−0.22

		S_0	ϕ_α	θ_1	θ_3	ψ_2	ψ_3	ψ_4
Cl:	Calc.	0.436	14	−1.2	−0.1	3.9	−153.6	157.5
	Exp.	0.44	14			13	−136	154
Br:	Calc.	0.205	9	−1.9	−1.0	12.1	−159.2	171.3
	Exp.	0.20	9			22	−105	134

Magnetic excitations (RPA)

$$\overline{\overline{\chi}}(\alpha i, \beta j, \omega) = \overline{\overline{\chi}}^0_{\alpha\beta}(i,\omega)\delta_{ij} + \sum_{k,\gamma,\eta} \overline{\overline{\chi}}^0_{\alpha\gamma}(i,\omega)\, J^{\gamma\eta}_{ik}\, \overline{\overline{\chi}}(\eta k, \beta j, \omega)$$

$$\chi^0_{AB}(j,\omega) = \lim_{\epsilon\to 0^+}\left[\sum_{ab}^{E_a\neq E_b} \frac{\langle a\,|\,A\,|\,b\rangle\langle b\,|\,B\,|\,a\rangle}{E_b - E_a - \hbar(\omega + i\epsilon)}(n_a - n_b) + \frac{1}{k_B T}\left(\frac{i\epsilon}{\omega + i\epsilon}\right)^2 \left\{\sum_{ab}^{E_a = E_b} \langle a\,|\,A\,|\,b\rangle\langle b\,|\,B\,|\,a\rangle\, n_a - \langle A\rangle\langle B\rangle\right\}\right]$$

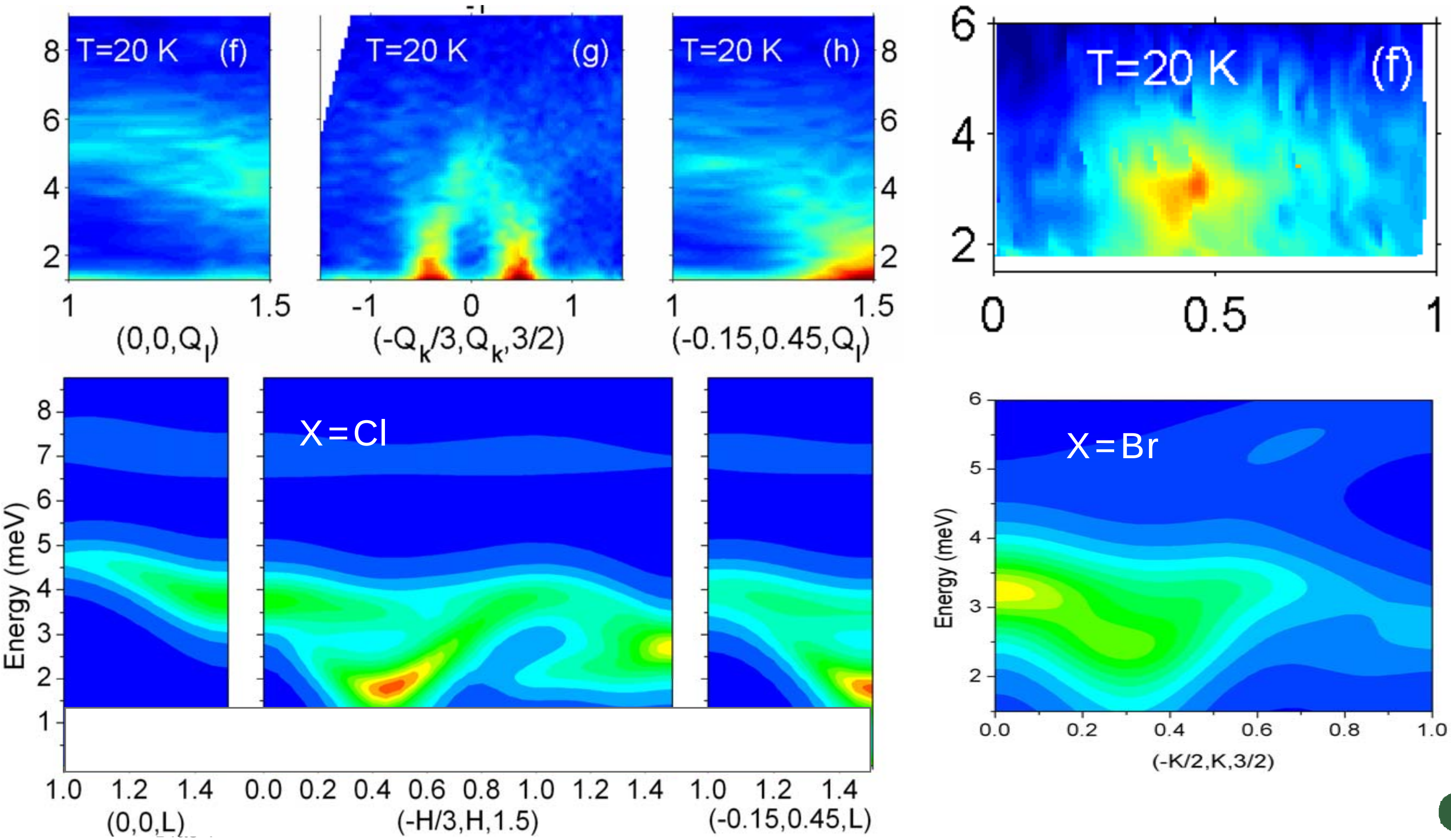
Paramagnetic phase:

$$\overline{\overline{\chi}}_{\alpha\beta}(\mathbf{q},\omega) = \overline{\overline{\chi}}^0_{\alpha\beta}(\omega) + \sum_{\gamma\eta} \overline{\overline{\chi}}^0_{\alpha\gamma}(\omega) J^{\gamma\eta}(\mathbf{q}) \overline{\overline{\chi}}_{\eta\beta}(\mathbf{q},\omega)$$

$$I(\mathbf{q},\omega) = \sum_{\xi\eta} \frac{\delta_{\xi\eta} - q_\xi q_\eta / q^2}{4\pi\left(1 - e^{-\hbar\omega/k_B T}\right)} \sum_{\alpha\beta} \mathrm{Im}\left[\chi^{\xi\eta}_{\alpha\beta}(\mathbf{q},\omega) e^{-i\,\mathbf{q}\cdot(\mathbf{r}_\alpha - \mathbf{r}_\beta)}\right]$$

Singlet-triplet excitations at 20 K

K. Prša, H. M. Rønnow, O. Zaharko, H. Berger, J. J. Chang, S. Streule, N. B. Christensen, J. Jensen, M. Jiménez-Ruiz, M. Prester, and J. Mesot (to be published).



Spin waves in the ordered phase

$$\overline{\overline{\chi}}(\alpha i, \beta j, \omega) = \overline{\overline{\chi}}^{0}_{\alpha\beta}(i, \omega)\delta_{ij} + \sum_{k,\gamma,\eta} \overline{\overline{\chi}}^{0}_{\alpha\gamma}(i, \omega)\, J^{\gamma\eta}_{ik}\, \overline{\overline{\chi}}(\eta k, \beta j, \omega)$$

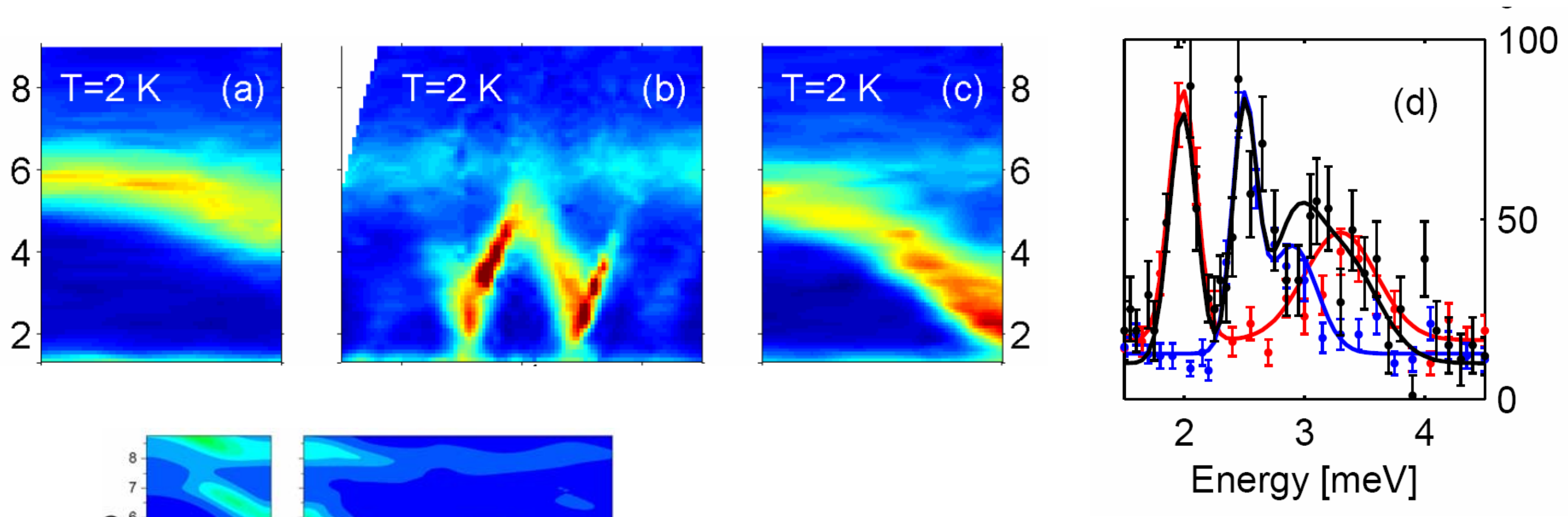
Commensurable structure with dimension $\tau_1 \times \tau_2 \times \tau_3$ unit cells

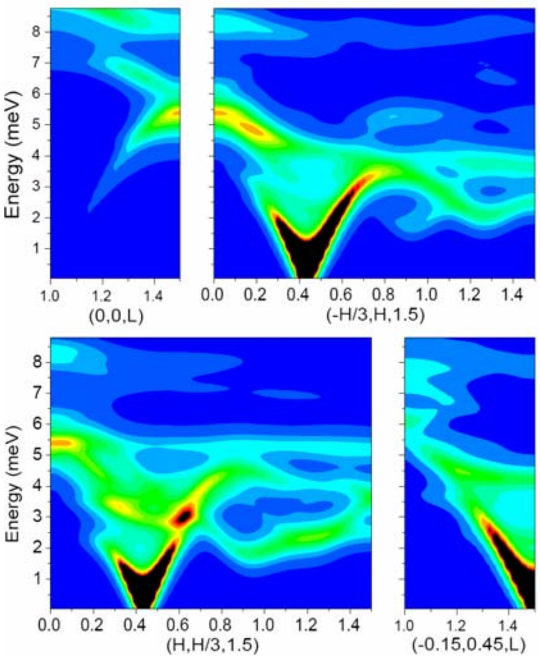
$$\mathbf{Q_p} = \left(\frac{p_1}{\tau_1}, \frac{p_2}{\tau_2}, \frac{p_3}{\tau_3}\right), \quad p_i = 0, 1, \ldots, \tau_i - 1$$

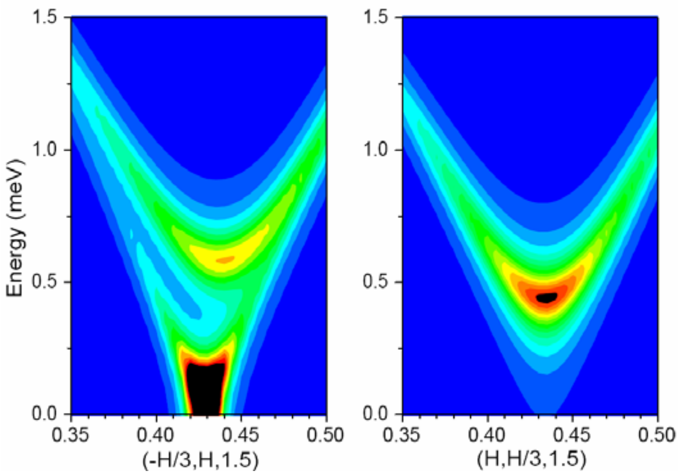
$$\overline{\overline{\chi}}_{\alpha\beta}(\mathbf{q}, \mathbf{Q_p}) = \overline{\overline{\chi}}^{0}_{\alpha\beta}(\mathbf{Q_p}) + \sum_{\mathbf{Q_s}} \sum_{\gamma\eta} \overline{\overline{\chi}}^{0}_{\alpha\gamma}(\mathbf{Q_p} - \mathbf{Q_s}) J^{\gamma\eta}(\mathbf{q} + \mathbf{Q_s}) \overline{\overline{\chi}}_{\eta\beta}(\mathbf{q}, \mathbf{Q_s})$$

$$I(\mathbf{q}, \omega) = \sum_{\xi\eta} \frac{\delta_{\xi\eta} - q_\xi q_\eta / q^2}{4\pi\left(1 - e^{-\hbar\omega/k_B T}\right)} \sum_{\alpha\beta} \mathrm{Im}\left[\chi^{\xi\eta}_{\alpha\beta}(\mathbf{q}, \mathbf{Q}_p = \mathbf{0}, \omega) e^{-i\,\mathbf{q}\cdot(\mathbf{r}_\alpha - \mathbf{r}_\beta)}\right]$$

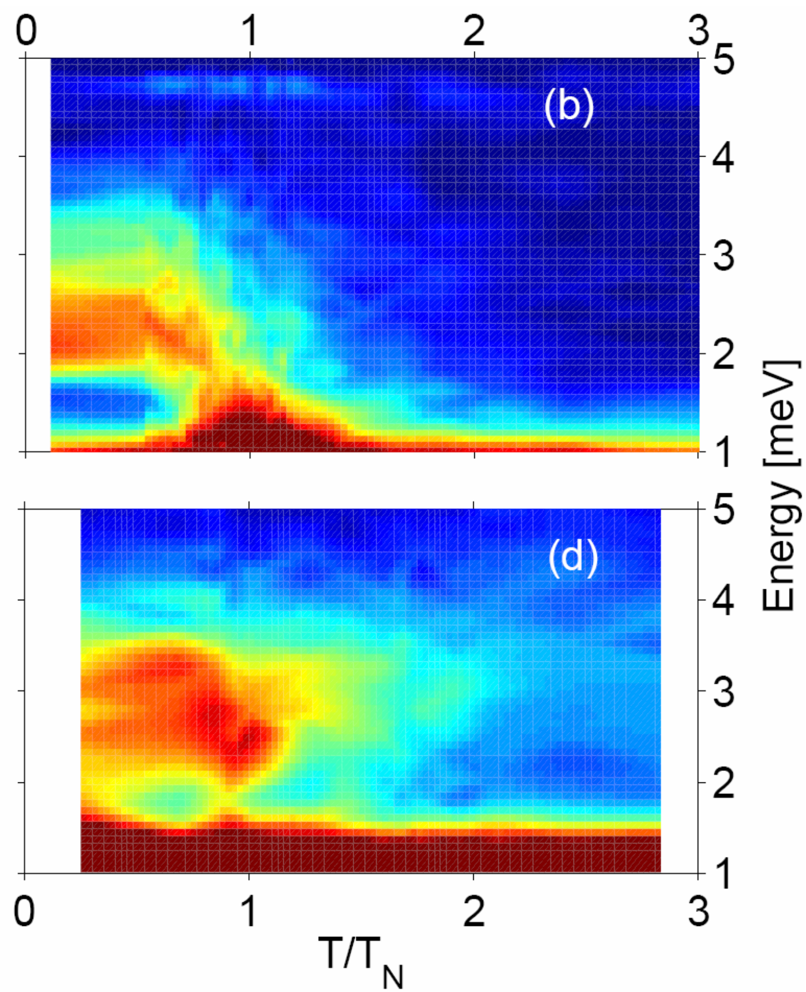
Spin waves in $Cu_2Te_2O_5Cl_2$ at 2 K

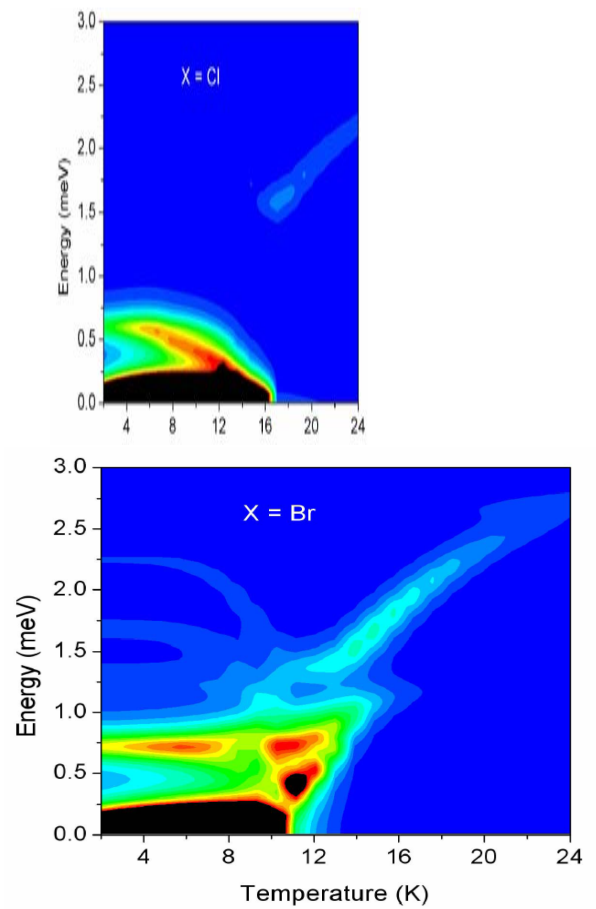






The excitation energy gap at **Q**





CONCLUSION

- The effective spin of the two systems is large (S_{eff} = 15/2) and the tetramerized spins are interacting in a 3D fashion.
- The cluster mean-field is able to account for the complex tilted helical ordering of the two compounds.
- The singlet-triplet excitations are clearly observable in the paramagnetic phase and behave as expected from theory.
- The DM anisotropy is anticipated to be weak, and the systems should show a Goldstone-like mode in their ordered phase.
- This is in accord with the "linear spin-wave theory" (RPA), but the experiments show a strongly gapped excitation near the magnetic Bragg points.
- This discrepancy is puzzling:
    - A shortcoming of the RPA?
    - 10-20 times larger anisotropy?
    - Experimental difficulties?
    - ?