Novel Magnetic Excitations in a Breathing Pyrochlore Antiferromagnet

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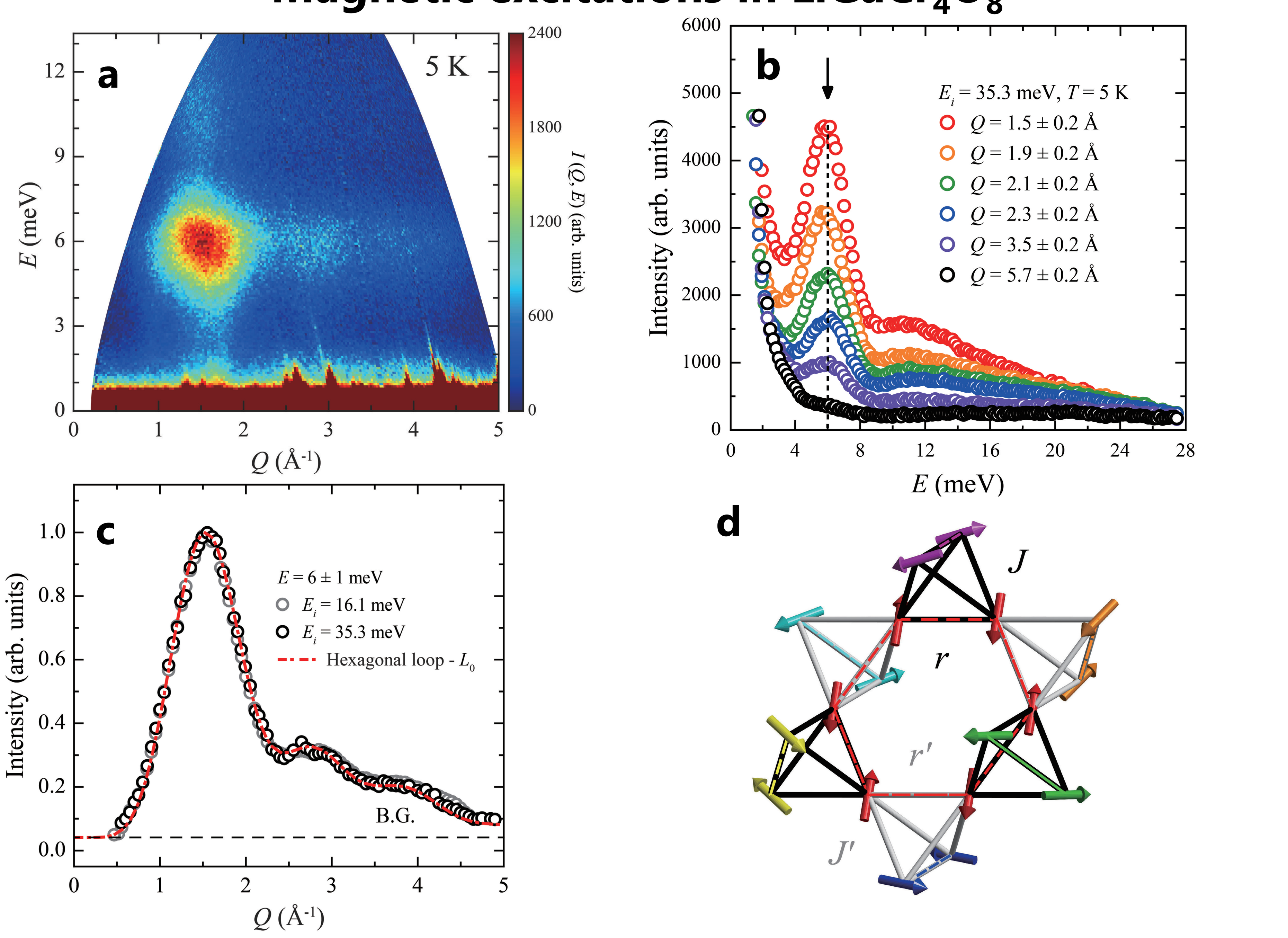
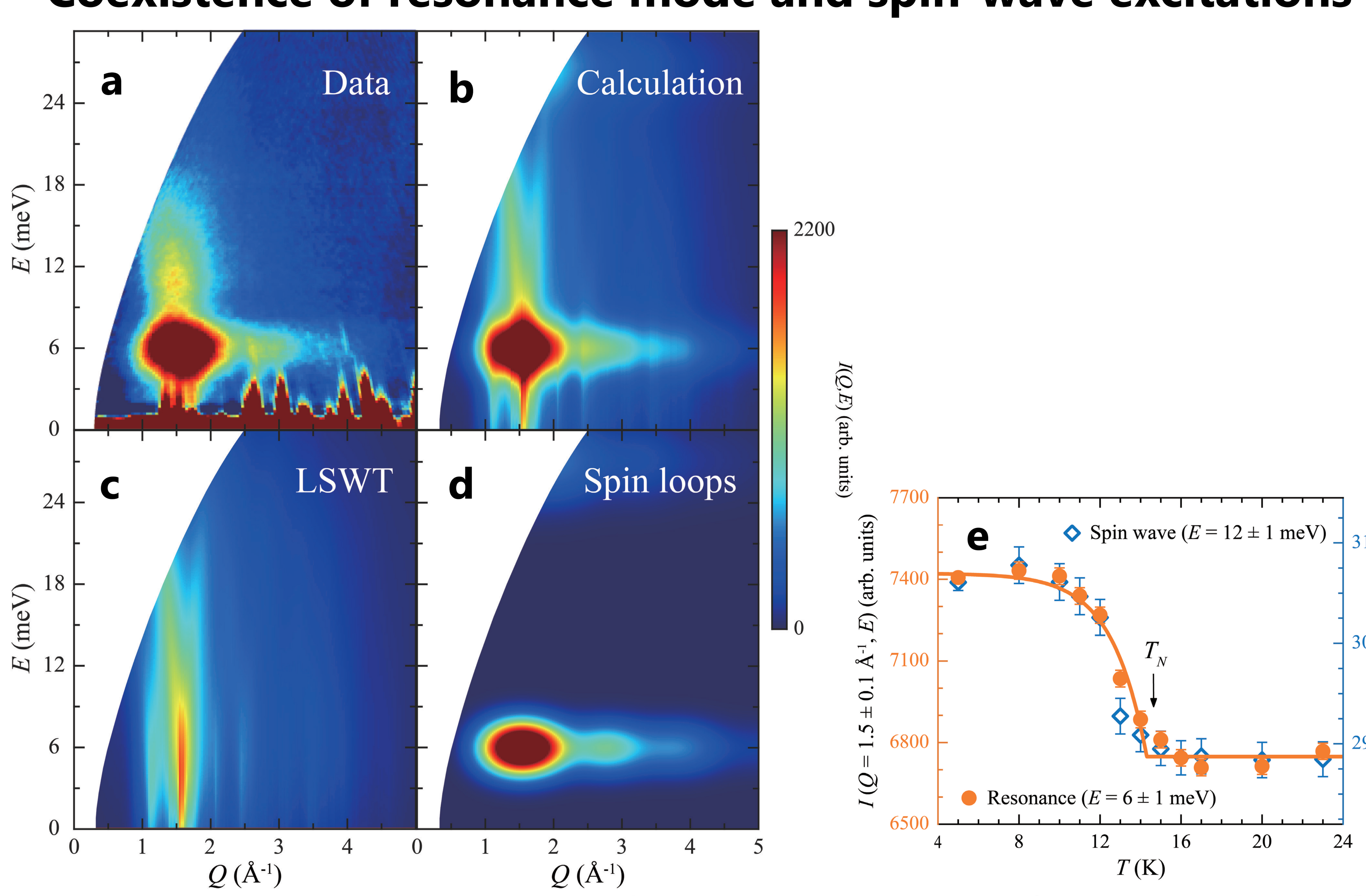
Abstract: **We report neutron scattering measurements of the spinel oxide $LiGaCr_4O_8$, in which magnetic ions Cr^{3+} form a breathing pyrochlore lattice. Our experiments reveal the coexistence of a nearly dispersionless resonance mode and dispersive spin-wave excitations in the magnetically ordered state, which can be quantitatively described by a quantum spin model of hexagonal loops and linear spin-wave theory with the same set of exchange parameters, respectively. Comparison to other Cr spinel oxides reveals a linear relationship between the resonance energy and lattice constant across all these materials, which is in agreement with our hexagonal loop calculations. Our results suggest a unified picture for spin resonances in Cr spinel oxides.**

Magnetic excitations in $LiGaCr_4O_8$



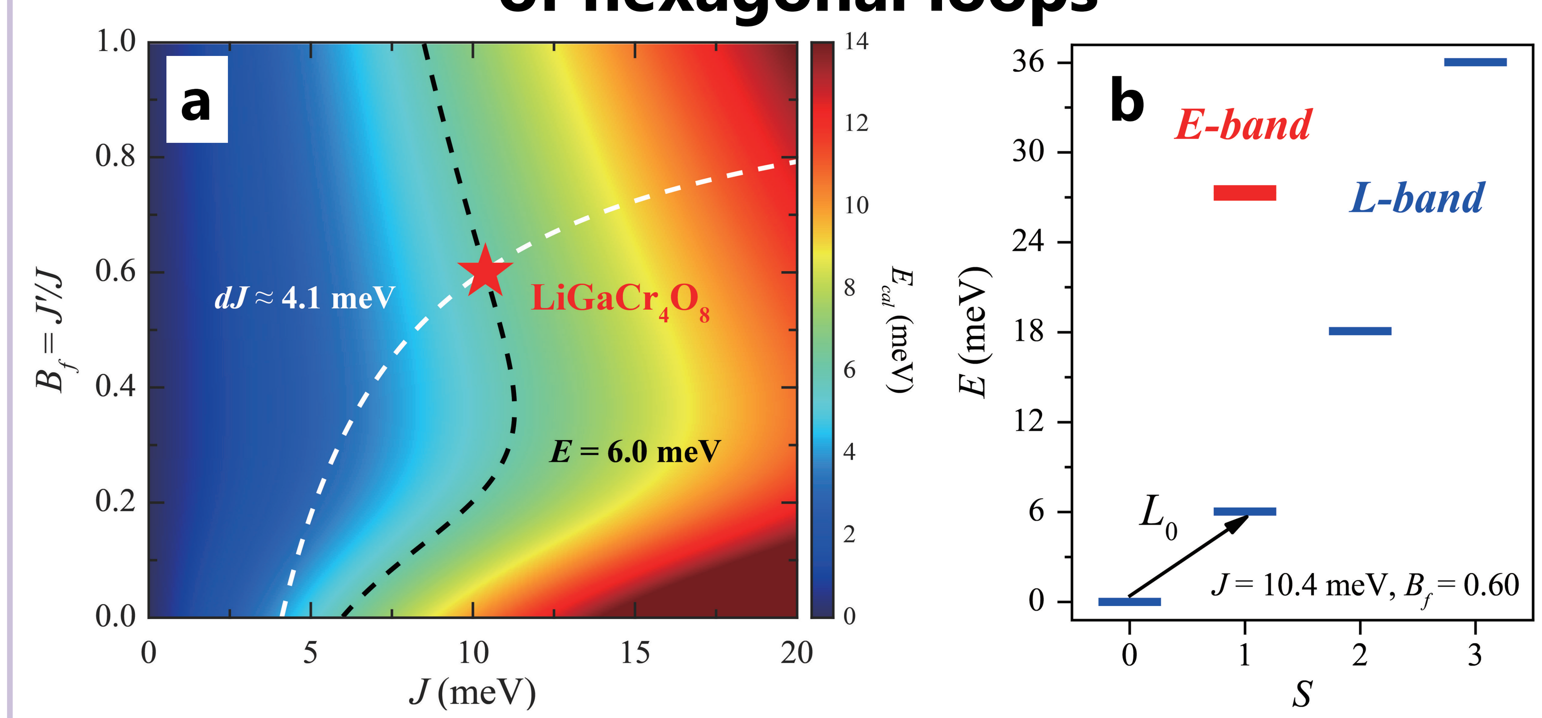
a, Magnetic excitation spectra of $LiGaCr_4O_8$ at 5 K with incident neutron energy E_i = 16.1 meV. **b**, Energy cuts at indicated Q. The arrow indicates the resonance mode. **c**, Q dependence of the spin resonance measured with different incident energies and the calculated spin excitation L_0 for hexagonal spin loops (red curve) with a constant background. **d**, The sketch of a hexagonal spin loop on the lattice of $LiGaCr_4O_8$. The hexagonal loop is formed by alternating bonds with bond length r and r', which correspond to AFM interactions J and J'.

Coexistence of resonance mode and spin-wave excitations



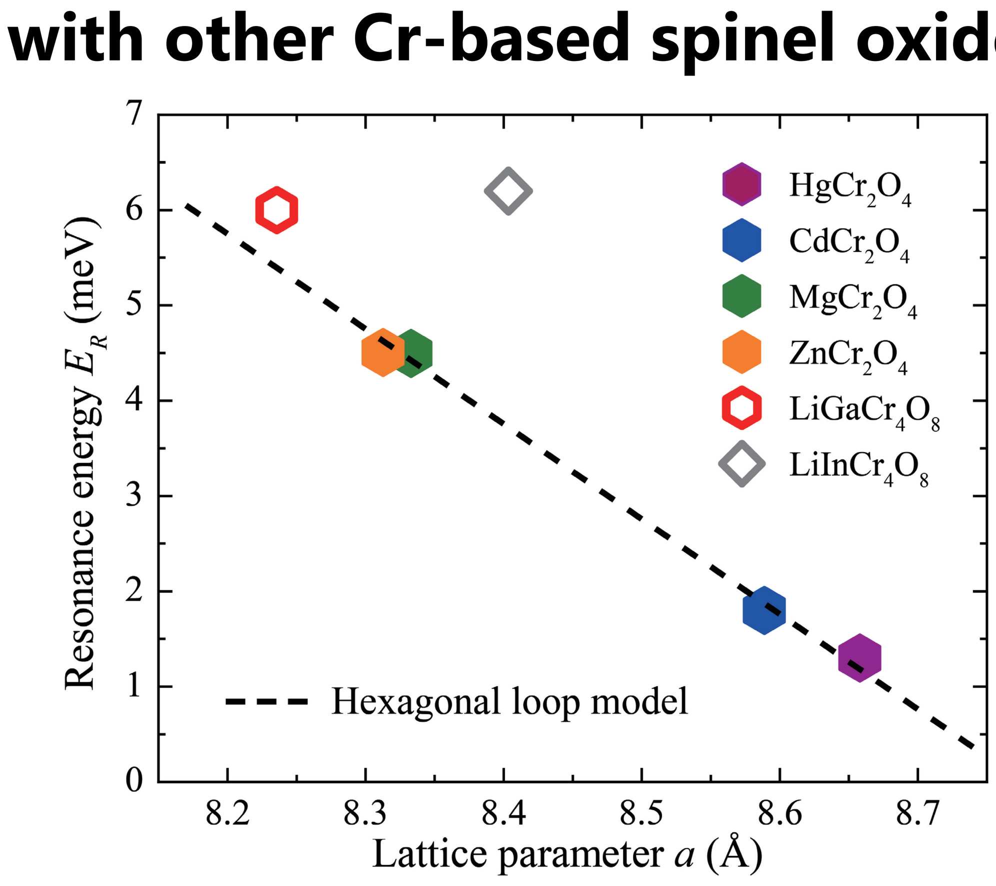
a, Magnetic excitation spectra of $LiGaCr_4O_8$ at 5 K with E_i = 35.5 meV. **b**, The total simulated scattering spectra combining the contributions from: **c**, Calculated spin-wave spectra and **d**, Calculated hexagonal spin loops' excitations. **e**, Temperature dependences of the spin resonance and spin wave. **The simultaneous development of spin-liquid-like loop excitations and spin-wave excitations below T_N suggests that the stable magnetic order not only promotes the long-range spin-wave propagation mode, but also meets the antiparallel conditions inside the hexagonal loop, which leads to spin loop excitations.**

Resonance mode in a quantum spin model of hexagonal loops



a, Calculated resonance energies versus magnetic interactions J and breathing factors $B_f = J'/J$. Combining the calculation with the resonance energy E = 6.0 meV from our experiments and the constraint of dJ = J-J' from the universal ratio of dJ/dr in chromium oxides yields J = 10.4 meV and B_f = 0.60. **b**, The calculated energy spectrum versus the total spin S of the hexagonal loop using the above parameter set. The blue and red symbols indicate L and E band spin levels.

Compare the resonance mode of $LiGaCr_4O_8$ with other Cr-based spinel oxides



The relationship between resonance energies E_R and lattice parameters a of Cr-based spinel oxides. The dashed line represents the calculated linear relation between E_R and a with dE_R/da = -10 meV/Å using our quantum hexagonal loop model with the fixed breathing factor B_f = 1 and the universal ratio of dJ/dr ≈ −40 meV/Å in chromium oxides. **This strongly indicates that the resonance modes in all other ACr_2O_4 are also associated with the hexagonal spin loops and the resonance energy provides an accurate measurement of the magnetic exchange interaction.** The resonance energy in $LiGaCr_4O_8$ would slightly deviate from the linear relationship owing to the presence of two different J and J' (B_f = 0.60). This also agrees with the data.

Reference

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