LOCAL SPIN RESONANCE AND SPIN-PEIERLS-LIKE PHASE TRANSITION IN $ZnCr_2O_4$

Most magnets order when the thermal energy drops below a microscopic energy scale for magnetic interactions. The topology of certain lattices can, however, reduce the energy gain associated with long range ordering thus favoring more entropic phases. Figure 1 shows one such lattice, the tetravalent site in the pyrochlore structure, that forms a network of corner-sharing tetrahedra. As shown in the figure, antiferromagnetically (AFM) interacting spins on the lattice cannot satisfy all their exchange interactions simultaneously. This phenomenon, called geometrical frustration, can lead to macroscopic classical ground state degeneracy and offers the possibility of qualitatively new states of matter. Theoretical studies have in fact shown that spins with nearest neighbor antiferromagnetic Heisenberg interactions on the pyrochlore lattice do not have a long range ordered phase at all.

Pyrochlore magnets studied experimentally so far exhibit a coninuous phase transition at a finite temperature, T_f, into a glassy phase with static short range correlations [1]. Spinel antiferromagnets, AB_2O_4, in which the octahedral B site forms the same magnetic lattice as in the pyrochlore structure, however behave quite differently. For instance, $ZnCr_2O_4$ exhibits a first order phase transition to a long-range ordered Néel phase at $T_c = 12.5$ K , much less than the Curie-Weiss temperature $|\theta_{CW}| = 393$ K. We have explored this ordered phase and the corresponding phase transition through inelastic neutron scattering [2].

Figure 2 provides an overview of our neutron scattering results as a color image of $\tilde{I}(Q, \omega)$ at three temperatures. For $T > T_c$, Figures 2a and 2b show a constant-Q ridge centered at $Q \approx 1.5$ Å^{-1} and extending beyond 10 meV. The ridge indicates quantum critical fluctuations of small AFM clusters, most likely antiferromagnetically correlated tetrahedra, and closely resembles those obtained in similar experiments on other frustrated AFM. For $T < T_c$, however, the low energy spectral weight concentrates into a sharp constant-energy mode centered at $\hbar\omega = 4.5$ meV $\approx |J| >> k_BT_c$. The wave vector dependence of this resonance intensity reveals that it is an excitation among antiferromagnetically correlated nearest neighbor spins. Though they can not be seen in Fig. 2c, there are in fact magnetic Bragg peaks in the elastic scattering channel (see Fig. 3b), which provide evidence for long-range order for $T < T_c$. It is unusual that excitations of such localized character exist in a long-range ordered phase. The resonance

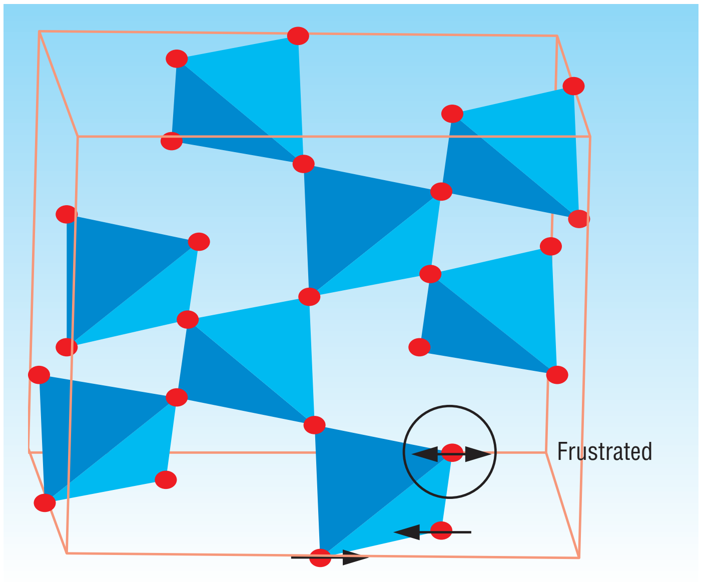

FIGURE 1. A network of corner-sharing tetrahedra. When two spins in a tetrahedron are aligned antiparallel to each other then the third spin can not satisfy its antiferromagnetic interaction with the other two spins simultaneously.

indicates the presence of weakly interacting spin clusters within the ordered phase, which is a key feature of geometrically frustrated magnets.

Theoretical work indicates that magnetic order cannot develop in an isotropic spin system with nearest neighbor antiferromagnetic Heisenberg interactions on the pyrochlore lattice. It is natural to ask what deviation from this model causes order to develop in $ZnCr_2O_4$? To answer the question we probed the temperature dependence of static and dynamic features of this system in the vicinity of the phase transition. Figure 3 shows that long range antiferromagnetic order (blue squares in frame (b)) and the local spin resonance (frame (a)) appear simultaneously in a spectacular first order transition. It also shows that magnetic ordering is accompanied by a cubic to tetragonal lattice distortion (red circles in frame (b)). The lattice distortion plays a crucial role in relieving frustration and allowing long-range order to develop. It is well known that exchange interaction between Cr^{3+} ions whose oxygen coordination octahedra share an edge are strongly dependent on the oxygen bond angles and hence the metal ion spacing. As a consequence the tetragonal strain $\epsilon_a > 0$ and $\epsilon_c < 0$ yields weaker AFM interactions between spins occupying the same basal plane and stronger AFM interactions between all other spin pairs. This reduces

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the energy of a particular long range ordered spin configuration with respect to the low energy degenerate manifold thus allowing the system to achieve long range order. The overall picture that emerges is that of two distinct phases in competition: a cubic cooperative paramagnet and a tetragonal long-range ordered antiferromagnet. Though the spin Hamiltonian has a lower expectation value in the latter phase, the lattice energy is greater and the entropy is lower in the tetragonal phase. The phase transition occurs when the free energy of the tetragonal low entropy phase drops below that of the disordered cubic paramagnet.

There are strong analogies between the phase transition in $ZnCr_2O_4$ and the spin-Peierls (SP) transition. In both cases the high T phase is nearly quantum critical and can lower its energy through a lattice distortion. In both cases the transition occurs from a strongly correlated paramagnet: $T_c << \theta_{CW}$, and in both cases low energy spectral weight is moved into a finite energy peak.

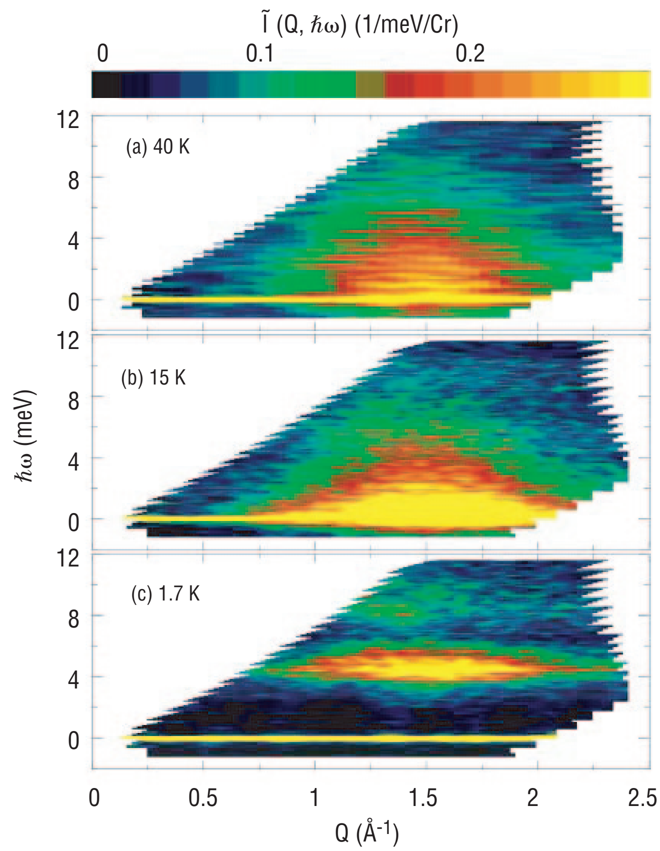

FIGURE 2. Contour maps of the magnetic neutron scattering intensity at temperatures spanning the phase transition at T_c = 12.5(5) K. The data were taken by utilizing a flat analyzer and two-dimensional position-sensitive detector at the SPINS spectrometer.

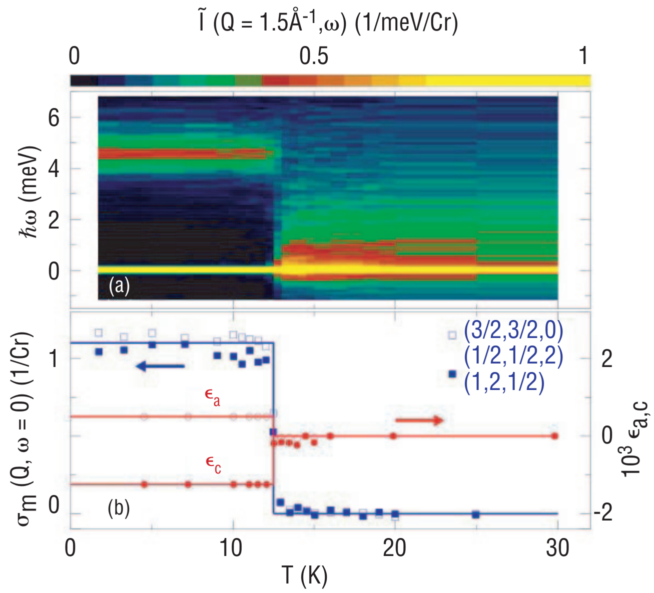

FIGURE 3. (a) Image of inelastic neutron scattering for Q = 1.5 Å^{-1}. (b) T-dependence of magnetic Bragg scattering from a powder, σ_m (blue squares), and of lattice strains measured n a single crystal (red circles).

There are also important differences between the two transitions. The low T phases are qualitatively different, the transition in $ZnCr_2O_4$ is a first order one, while the SP transition is second order, and the change in entropy at T_c plays an important role in $ZnCr_2O_4$, but not in a SP transition. The central idea that finite lattice rigidity can preclude a spin liquid at T = 0 however does carry over and should be relevant for any frustrated magnet when other symmetry breaking interactions are sufficiently weak.

REFERENCES

[1] J. S. Gardner, B. Gaulin, S.-H. Lee, C. Broholm, N. P. Raju, J. E. Greedan, Phys. Rev. Lett. **83**, 211 (1999).

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