Probing Spin Correlations with Phonons in the Strongly Frustrated Magnet $ZnCr_2O_4$

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$ZnCr_2O_4$ represents a nearly ideal 3D geometrically frustrated Heisenberg spin 3/2 system on a pyrochlore lattice. The interaction between the Cr^{3+} ions is dominated by direct exchange. While the Curie-Weiss temperature is 390 K the system remains paramagnetic down to 12.5 K where it undergoes a structural phase transition and goes into a Neel antiferromagnetic state. It has been proposed spin-lattice interaction drives this phase transition. Ionic displacement modes that modulate the Cr-Cr distance breaks the frustration allowing the huge spin degeneracy of the ground state to be partially lifted by a mostly tetragonal static distortion with c < a,a.

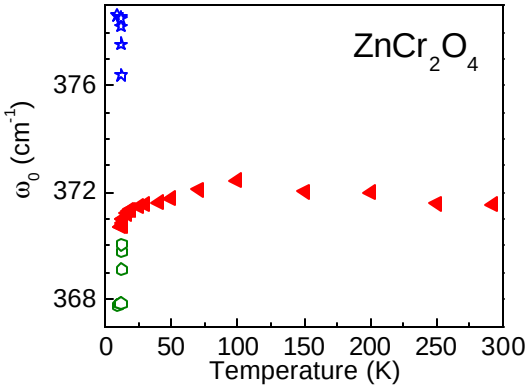

Fig. Resonance frequency of the $F_{1u}(3)$ phonon, coupled to spins. Phonon triplet (triangles) softens below 100 K and splits into a doublet (hexagons) and a singlet (stars) below Tc=12.5 K.

We have measured the temperature dependence of the phonon spectrum of $ZnCr_2O_4$. Only the one out of four infrared active phonons which strongly modulates the Cr-Cr coordinates shows spin-correlated temperature behavior (Fig.) [1]. We report two main observations: Softening of the phonon frequency in the spin liquid state below ~100 K and its splitting into two bands below Tc. The frequency shift due to the spin-phonon coupling gives a measure of the spin-spin correlation function $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle(T)$

The frequency shift due to spin-phonon coupling, $Dw(T) = l \langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle(T)$, where $\langle \boldsymbol{S}_i \cdot \boldsymbol{S}_j \rangle(T)$ is a spin-spin correlation function, gives a measure of l, the is a spin-phonon coupling constant. We extract $l = 6.2$ cm^{-1} by comparing the phonon softening in the paramagnetic state with the magnetic specific heat [2] of $ZnCr_2O_4$. The average value of the spin-Peierls order parameter in the low temperature phase is then determined from the phonon splitting. The results show that the ordered phase is more complicated than a pure tetragonal distortion of the lattice. The observed effects are in overall agreement with the picture of predominant spin-lattice interactions in strongly frustrated magnet $ZnCr_2O_4$. Supported by MRSEC/NSF No. DMR-0080008 and DMR-0348679

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[2] H. Martinho, N. O. Moreno, J. A. Sanjuro, C. Rettori *et al.*, Phys. Rev. B **64**, 024408 (2001).