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Probing spin correlations with phonons in the strongly frustrated magnet: ZnCr2O4[1]

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Geometrically frustrated magnets can resist magnetic ordering and remain in a strongly correlated paramagnetic state well below the Curie-Weiss temperature. The spin-lattice coupling can play an important role in relieving the frustration in these systems. In ZnCr2O4, an excellent realization of the Heisenberg antiferromagnet on the pyrochlore network, a lattice distortion relieves the geometrical frustration through a spin- Peierls-like phase transition at Tc=12.5 K with a lowering of the symmetry from cubic to tetragonal. Conversely, spin correlations strongly influence the elastic properties of a frustrated magnet. By using infrared spectroscopy and published data on magnetic specific heat, we demonstrate that the frequency of the Cr optical phonon triplet in ZnCr2O4 tracks the nearest-neighbor spin correlations above Tc.* Below Tc, the triplet splits into a singlet and a doublet, separated by 11 cm- 1. This splitting gives a direct measurement of the spin- Peierls order parameter. From analysis of the ion displacements in the phonon modes we can conclude that direct Cr-Cr exchange dominates in ZnCr2O4. These experiments result in a clear understanding of spin-phonon coupling in ZnCr2O4 in contrast to other oxide magnets. Recent ab initio calculations** confirm the magnetic origin of both the phonon splitting in ZnCr2O4 and the frequency shifts in the ferromagnetic insulating spinel CdCr2S4. *A.B. Sushkov et al., Phys. Rev. Letters 94 (2005) 137202. ** C.J. Fennie and K.M. Rabe, cond-mat/0508136 and Mar06 APS Meeting.

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