Thermal Properties of Geometrons in the Frustrated Spinel $ZnCr_2O_4$

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Introduction

Spinels of general formula $A^{2+}Cr_2^{3+}O_4$, A^{2+} being a non-magnetic ion, are one of the simplest systems in which to study the low-energy magnetic excitations of a 3D Heisenberg magnet in an environment of strong geometrical frustration. The flexibility of the spinel structure provides a unique way to tune the nearest-neighbor exchange coupling, J, and the frustration parameter, $f=\Theta_{CW}/T_N$, through chemical doping at the tetrahedral A^{2+} site. Moreover, neither the orbital degrees of freedom nor charge fluctuations are expected to be relevant in the Cr^{3+}:t_{2g}^3 of this large gap insulator. In the particular case of $ZnCr_2O_4$, it undergoes a structural distortion and develops a non-collinear coplanar magnetic order below $T_{N,S}$=12.5(5) K. Below $T_{N,S}$, the spectral weight of the spin-excitation spectrum condenses into two modes at 4.5 meV and 9.0 meV, corresponding to AF-hexamer and heptamer bond-spin structures [1]. These spin molecules play the role of the elementary excitations (geometrons) of the frustrated spin system. We have recently measured the thermal conductivity of $ZnCr_2O_4$ single crystals down to 50 mK in magnetic fields up to 14 T (see **Figure 1**).

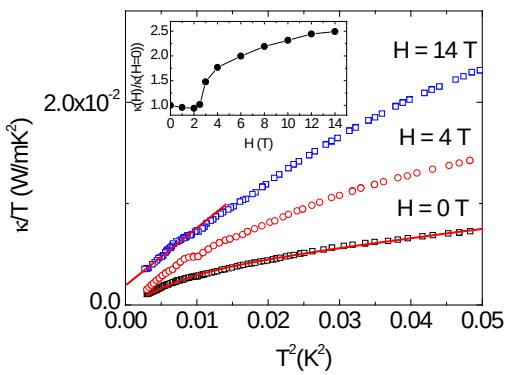

Figure 1: Field and temperature dependence of thermal conductivity below the magnetostructural transition. The solid lines represent fits to $\kappa(T) = \alpha\exp(-\Delta z/k_BT) + \beta T^3$

The results are puzzling. The zero field thermal conductivity is well fitted by $\kappa(T) = \alpha\exp(-\Delta z/k_BT) + \beta T^3$ in which the first term is a thermally-activated term for spin thermal conduction and the second one is the usual Debye phonon term. However, at higher fields a simple cubic term, plus a linear contribution are enough to describe the system at low temperatures. The origin of the linear term is unclear, as the samples are very good insulators. Hexameric and heptameric resonant molecular structures were reported in the ordered phase of $MgCr_2O_4$ and $ZnCr_2O_4$ 2] (clusters of highly frustrated spins or geometrons that act as independent quasiparticles). As the magnetic field destabilizes the spin-bond structure, spin frustration restores partially, so that gapped and gapless excitations may coexist in this structure. We have now synthesized single crystals of $ZnCr_2O_4$ and $MgCr_2O_4$, which show different Cr-Cr distances. This is a unique opportunity to study the thermodynamic properties of this new state that shows up after the broken-symmetry phase of the pyrochlore lattice is destabilized by a magnetic field. Higher magnetic fields must be applied to saturate the contribution of spin excitations to the thermal conductivity and specific heat, so they can be appropriately modeled.

Experimental, Results & Conclusion

Single crystals of $ZnCr_2O_4$ were measured using a dilution-refrigerator specific heat option in a 14 T Physical Properties Measurement System by Quantum Design. Below 500 mK a second time constant was observed in the data, and this time constant increased exponentially with decreasing temperature, making measurements impossible below 500 mK. This observation was robust during several attempts to remount the sample. It is most likely due to a poor thermal contact between the sample and the substrate (Kapitza resistance), which generally has to do with non-matching phonon frequencies. The internal thermal conductivity of the sample cannot account for this second tau, and neither can nuclear magnetic moments. This is an unusually high temperature in which to encounter such large Kapitza resistances. The best path forward is to press a pellet of the sample mixed with a conducting metal to improve thermal contact.

Acknowledgements

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References

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