Heat Capacity and Magnetocaloric Effect of the Spin Dimer System Sr$_3$Cr$_2$O$_8$

A.A. Aczel (McMaster, Physics and Astronomy), G.M. Luke (McMaster, Physics and Astronomy), M. Jaime (LANL), and Y. Kohama (LANL)

Introduction
Spin dimer compounds possess non-magnetic ground states that are the direct product of spin-singlets. The recent interest in these systems is due to quantum phenomena associated with their magnetic excitations, called triplons. Upon application of a magnetic field larger than the critical field $H_{c1}$ necessary to close the spin gap, one can drive a spin dimer system to a regime characterized by long-range magnetic order [1]. In the simplest scenario, these systems become fully polarized above a second critical field $H_{c2}$ and so they can be tuned through two quantum critical points. If the kinetic energy terms dominate the potential energy terms in the U(1) invariant Hamiltonian of such a system, the ground state between $H_{c1}$ and $H_{c2}$ can alternatively be described as a Bose-Einstein condensate (BEC) of triplons. In ideal BEC systems, the (H,T) phase boundary follows the expression $T_c = A(H-H_{c1})^{2/d}$ close to the critical fields $H_{c1}$ and $H_{c2}$, where $d$ is the dimensionality of the system.

Experimental
Our group has succeeded in producing large single crystals of the new spin dimer system Sr$_3$Cr$_2$O$_8$ for the first time by the traveling solvent floating zone technique [2]. We measured the magnetic susceptibility of some of our crystals and found evidence of the spin gap behaviour we were expecting for this system. From this data, we were able to estimate the singlet-triplet gap to be 61.5(3) K. We then proceeded to collect heat capacity and magnetocaloric effect (MCE) data in Tallahassee using the 35 T resistive magnet.

Results and Discussion
The heat capacity measurements showed lambda-like anomalies above ~30.4 T; this provided unambiguous evidence for the field-induced order in this system (Fig 1a). We were able to map out a complete phase diagram for the $H \perp c$ orientation of Sr$_3$Cr$_2$O$_8$ through a combination of heat capacity, MCE, and magnetization measurements (Fig. 1b), some of which were performed using the pulsed fields at Los Alamos because of the high $H_{c1}$ value for this system.

Many MCE scans were performed at low temperatures so that we could determine a critical exponent for this system. The fitted data all must lie within the universal regime for the obtained critical exponent to be meaningful, and an earlier Monte Carlo simulation study [3] suggested that the universal regime extends no higher than $\sim 0.4T_{\text{max}}$, where $T_{\text{max}}$ is the maximum temperature of the magnetically-ordered regime. To ensure that we stayed within the universal regime, only the MCE data below $\sim 2.7$ K was fit to a power law of the form: $T_c = A(H-H_{c1})^\nu$. Our best fit, shown in the Fig 1b inset, yielded $\nu = 0.65(2)$ with a corresponding $H_{c1} = 30.40(1)$ T. The former is in agreement with the expected value of 2/3 for a 3D BEC universality class, strongly suggesting that Sr$_3$Cr$_2$O$_8$ is a realization of a new triplon BEC. Note that the maximum ordering temperature of 8 K for Sr$_3$Cr$_2$O$_8$ is the highest observed in any triplon BEC system where $H_{c2}$ is experimentally-accessible.

References

Figure 1: (a) Sr$_3$Cr$_2$O$_8$ heat capacity results, (b) Complete Sr$_3$Cr$_2$O$_8$ phase diagram determined via heat capacity, magnetocaloric effect, and magnetization measurements.