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Field-induced quantum phase transitions in the spin-1/2 triangular-lattice antiferromagnet Cs₂CuBr₄

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Abstract. In classical magnetic spin systems, geometric frustration leads to a large number of states of identical energy. We report here evidence from magnetocaloric and related measurements that in Cs_2CuBr_4 — a geometrically frustrated Heisenberg S= 1/2 triangular antiferromagnet — quantum fluctuations stabilize a series of gapped collinear spin states bounded by first-order transitions at simple increasing fractions of the saturation magnetization for fields directed along the c axis. Only the first of these quantum phase transitions has been theoretically predicted, suggesting that quantum effects continue to dominate at fields much higher than previously considered.

1. Introduction

How will antiferromagnetically coupled spins orient themselves relative to their nearest neighbors? The answer depends on the detailed nature of the spin coupling, the geometric arrangement of the spins in the crystal lattice, and the strength of the applied magnetic field. When the spins are arranged in a 2D triangular lattice, as in Cs_2CuBr_4, the zero field ground state is degenerate, because the spins are unable to align opposite to all of their neighboring spins simultaneously [1, 2, 3].

Quantum fluctuations can lift this geometrically induced degeneracy, creating a variety of otherwise unexpected ground states and excitations. In particular, in the presence of an externally applied magnetic field, these fluctuations can stabilize remarkable collinear spin structures over a finite field region. For a spin $\frac{1}{2}$ Heisenberg antiferromagnet, the collinear arrangement of spins produces a plateau (or plateaus) in the magnetization M at $M/M_s = 1 - 2d/(u+d) = 1 - 2d/n$ where d is the number of down spins, u is the number of up spins, $n = u + d$, and M_s is the saturation magnetization.

For a triangular arrangement of spins like Cs_2CuBr_4 [4] or Cs_2CuCl_4 [5], theory predicts a magnetic-field induced quantum-phase transition into a *uud* ('up-up-down') collinear arrangement of two spins up and one spin down at $M = \frac{1}{3}M_s$ for a 3 spin unit cell [6]. Experimentally, however, Cs_2CuBr_4 is to date the only known $S = \frac{1}{2}$ triangular lattice
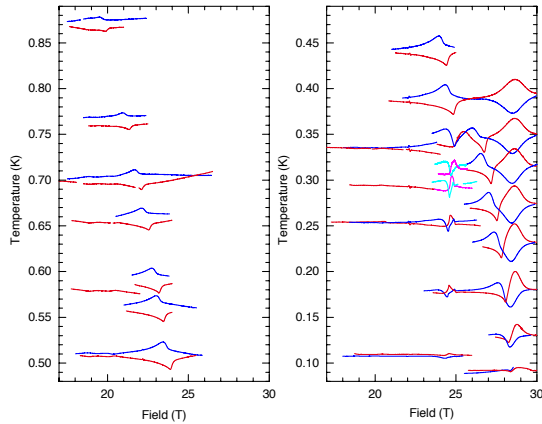
Figure 1. Magnetocaloric measurements as a function of magnetic field for a series of fixed temperatures, for fields directed along the crystallographic c axis. Red/blue traces represent field sweeps at $+0.2$ tesla/minute and -0.2 tesla/minute, respectively. Prominent features are seen just above the saturation field of 28.5 tesla, at the antiferromagnetic to paramagnetic phase boundary $H_c(T)$, and at two new field induced states around 22 tesla and $24 - 25$ tesla. The uud transition occurs at fields too low to be seen in this figure.

antiferromagnet in which this uud state occurs [4, 7]; the suppression of this quantum stabilized state with increasing in-plane anisotropy appears to prevent its formation in Cs_2CuCl_4 [8, 9]. The uud transition is unexpectedly first-order [4, 10], and does not appear when the magnetic field is directed perpendicular to the triangular layers [4]. The Dzyaloshinskii-Moriya interaction breaks the symmetry when the magnetic field is directed along the triangular layers, providing one possible explanation for the directional dependence and the first order nature of the transitions[9].

Interestingly, pulsed field magnetization measurements of Cs_2CuBr_4 [7] revealing the existence of a magnetization plateau at $\frac{1}{3}M_s$ also suggested the existence of an additional, theoretically unpredicted collinear state at $\frac{2}{3}M_s$. In this paper, we present a magnetocaloric and magnetic-torque measurements at temperatures down to 90 mK and in fields up to 30 tesla confirming the presence of additional quantum phase transitions in this material.

2. Measurements and Analysis

For the magnetocaloric experiments reported here, the applied field was directed along the crystallographic c axis [11], which is within the bc plane of the triangular spin lattice. The sample was thermally linked to a temperature controlled platform inside a miniature rotatable calorimeter [12] that was in turn weakly thermally linked to the cryogenic bath. The platform was held at a constant true temperature using a computer program that actively corrected for the magnetoresistance of the sensors [13].

When the magnetic field is swept at a constant rate, the temperature difference ΔT between the sample and the platform depends on the temperature dependence of the magnetization $(\partial M/\partial T)_H$, the field sweep rate $\dot{H}$, and the thermal conductance between the sample and the platform κ:

$$\Delta T = -\frac{T}{\kappa}\left[\left(\frac{\partial M}{\partial T}\right)_H + \frac{C_H}{T}\frac{d(\Delta T)}{dH}\right]\dot{H}. \qquad (1)$$

As shown in figure 1, reversing the field sweep direction reverses the sign of ΔT, creating an offset between the up and down sweeps that depends on $(\partial M/\partial T)_H$. In addition, we see a sharp deviation in ΔT when crossing the phase boundary between the lower field antiferromagnetic state and the higher field paramagnetic state. This feature moves out to higher field as we reduce the platform temperature, following the field and temperature dependence of the phase boundary. In the traces that extend out further in field, such as the trace at 0.385 K, a second broad feature appears near the saturation field of 28.5 tesla.

Closer inspection of the traces reveals additional transitions at fields below the antiferromagnetic to paramagnetic phase boundary. Most prominent are temperature independent transitions at 24.5 tesla and 25.0 tesla seen in the 0.335 K trace and below as the antiferromagnetic phase boundary shifts to higher field with decreasing temperature. Less prominent but also visible at 0.70 K and below are a pair of weak, second order transitions into and out of an additional magnetic phase at 18.8 and 20.4 tesla which we call the 'A' phase, plus a very narrow pair of first order transitions at 22.1 tesla which we call the 'B' phase.

In Figure 2, we compare the features identified in our magnetocaloric effect measurements with corresponding features in our measurements of the magnetic torque and magnetization. In all three measurements, features corresponding to the phase transitions into and out of the uud state at $\frac{1}{3}M_s$ are clearly visible, as are the higher field transitions discussed earlier.

In the magnetocaloric data shown in Figure 2a, we observe large but asymmetric temperature spikes upon entering and leaving the uud state for both field sweep directions. The shape of these spikes is due to two factors: the heat release due to the metastability of first-order phase transitions and the smaller latent heat release/absorption upon entering/leaving the uud state.

In the magnetic torque measurement shown in Figure 2b, the sample was first aligned along the c axis, then slightly rotated away towards the a axis so as to produce a measurable response. This particular trace is at too high a temperature to observe the $\frac{2}{3}M_s$ phase, but we again
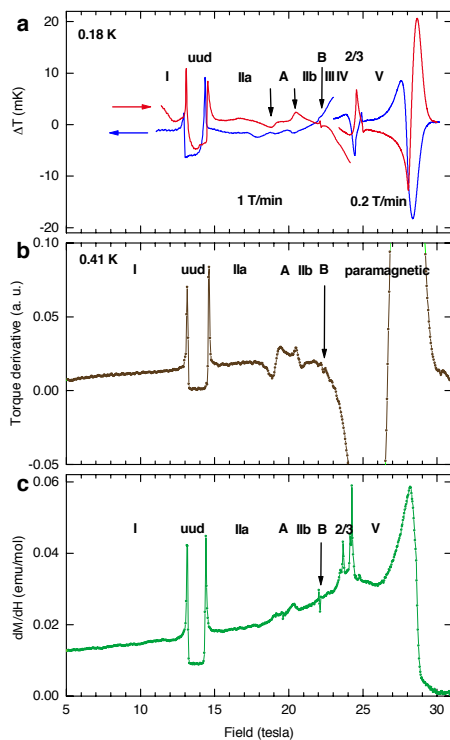
Figure 2. Comparison of magnetocaloric, magnetic torque, and magnetization measurements. Labels designate distinct field-induced states within the antiferromagnetic/paramagnetic phase boundary.
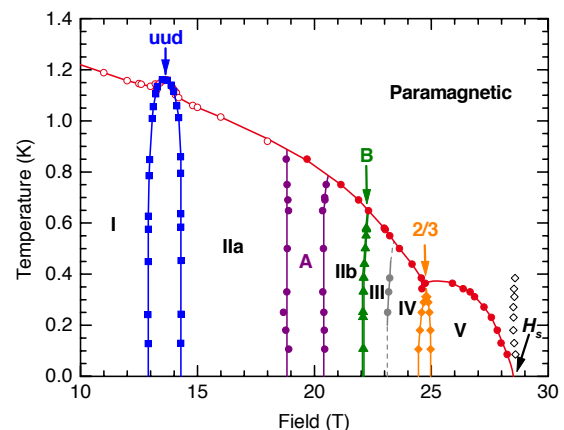
Figure 3. Magnetic phase diagram constructed from our magnetocaloric-effect measurements. Circles represent second-order phase boundaries; all other solid symbols represent first-order boundaries. The uud, B, and 2/3 phases are collinear states bounded by first-order phase transitions at $\frac{1}{3}$, $\frac{5}{9}$, and $\frac{2}{3}$ of the saturation magnetization M_s. The A phase at $\frac{1}{2}M_s$ is not collinear, and is bounded by second-order phase transitions. Roman numerals I, IIa and IIb, III, IV, and V indicate incommensurate phases. Open diamonds denote the features associated with the saturation field H_s and do not represent a phase boundary.

see prominent features at the phase boundaries to the $\frac{1}{3}M_s$, A and B phases, confirming their magnetic origin.

Finally, in Figure 2c we show the derivative of the magnetization with respect to field, taken from Ref. [7], to facilitate direct comparison with corresponding features in the magnetocaloric and magnetic torque measurements. Prominent features occur in both the magnetocaloric and magnetization measurements at the transitions into and out of the $\frac{1}{3}M_s$ or uud phase existing between 12.9 and 14.3 tesla, the A phase existing between 19 and 20.5 tesla, the very narrow B phase at 22 tesla, and the $\frac{2}{3}M_s$ phase [7] existing between 24 and 25 tesla.

3. Conclusions

A phase diagram constructed from our results is shown in Figure 3. Our results reveal a series of apparently collinear gapped spin states at $\frac{1}{3}$, $\frac{5}{9}$, and $\frac{2}{3}M_s$, suggesting that quantum effects continue to dominate at fields much higher than previously considered. The phase boundaries are nearly vertical, indicating that the phase diagram primarily reflects the zero-temperature energies rather than entropies of the different states. Suggested spin arrangements for these fractional states will be presented elsewhere.

As noted earlier, the field-induced quantum phase transitions between the collinear states and the adjacent incommensurate spin states are first-order. Thermodynamically, the magnetization plateaus accompanying these collinear states should occur in the field range corresponding to a cusp in the ground state energy $\mathcal{E}$ as a function of magnetization M. In the simplest case, the transitions to the collinear state are second order [14]. If, however, quantum fluctuations are strong enough to induce inflection points in $\mathcal{E}(M)$ near the cusp, then the transition can become first order. Animated graphs illustrating the evolution of the magnetization as a function of increasing field near a cusp in $\mathcal{E}(M)$ for both first and second order cases are included online with this paper. The field and magnetization values are arbitrary, and are not intended to be representative of the field values for any particular transition in Cs_2CuBr_4.

In this scenario, the first order nature of the the transitions to the collinear states could arise solely due to quantum fluctuations, independent of the presence of the Dzyaloshinskii-Moriya interaction. We are currently investigating the angular dependence of these field-induced phase transitions to better distinguish between fluctuation and spin-orbit contributions to the order of the transitions and the absence of these transitions for fields directed along the a axis.

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