Heat Capacity of a New S=1/2 Antiferromagnet on the Kagomé Lattice

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Abstract

A new spin $S = \frac{1}{2}$ antiferromagnet on the kagomé lattice, $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$ {titmb=1,3,5-tris(imidazol-1-ylmethyl)-2,4,6 trimethylbenzene} has been grown and we measured the heat capacity at low temperatures by a relaxation method. We have found two-peak structure in the temperature dependence of the heat capacity. The higher temperature peak is explained as due to a short-range magnetic ordering in two-dimension. The lower temperature peak suggests the presence of an energy gap in this $S = \frac{1}{2}$ kagomé antiferromagnet.

Key words: kagomé lattice; spin gap ; heat capacity ; two dimensional magnet

Spin systems with strong geometrical frustration exhibit interesting low energy properties. The two dimensional Kagomé Heisenberg antiferromagnet (KHA) is an example of such systems. For an S=$\frac{1}{2}$ KHA, theories predict that the ground state is in a disordered quantum spin liquid with a small spin gap to the excited statescite1,2. The spin gap estimated to be of the order of 1/20 of the exchange interaction constant[2]. The ground state of an S=$\frac{1}{2}$ KHA may be described by a quantum dimmer mode[3].

In this paper, we report the results of heat capacity measurements made on a new S=$\frac{1}{2}$ KHA, $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$ in which Cu^{2+} has $S = \frac{1}{2}$. The anisotropy in the g-tensor of copper (II) ion is small[4] so that Heisenberg model can be applied to the exchange interaction among the moments.

The compound $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$ has the hexagonal structure with the lattice parameters, a=15.539Åand c=21.149Å[4]. The crystal structure consists of Cu-CH_3COO infinite two dimensional Kagomé network which extends in the ab plane. These layers are well separated from each other by large titmb molecules and so the exchange interaction between the layers is expected to be much smaller than that within an ab layer.

Polycrystalline samples of $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$ were prepared by spontaneous assembly from the titmb ligand and copper (II) acetate in methanol solution. Details are described in ref. [4]. The material, titmb= 1,3,5-tris(imidazol-1-ylmethyl)-2,4,6-trimethylbenzene, was purchased from the Wako Pure Chemical Industries, Ltd. Heat capacity was measured by a relaxation method using a Quantum Design PPMS microcalorimeter in the temperature range between 0.4K and 20K. A sample of about 5mg in weight was attached to the sample platform with a small amount of Apiezon N grease.

Figure 1 shows the measured heat capacity, C, including the contribution of the lattice, as a function of temperature for the designated magnetic fields (H). The heat capacity of $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$ exhibited no sharp peaks down to 0.4K, which evidences the absence of long-range magnetic order in this temperature region. In zero field C deceases with decreasing temperature and shows an upturn below 1 K. With the application of external magnetic field a

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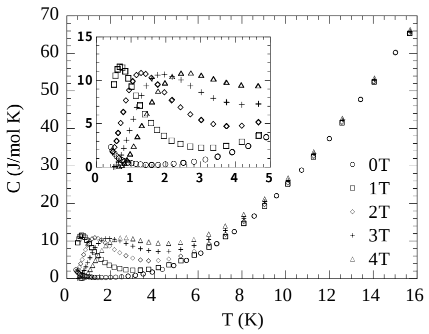

Fig. 1. The temperature dependence of the total heat capacity of $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$ in zero and applied magnetic fields. The inset shows the low temperature part.

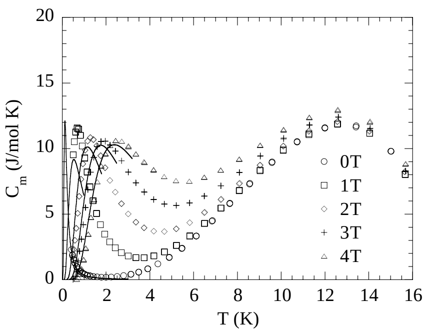

Fig. 2. The temperature dependence of the magnetic part of the heat capacity in $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$ after subtracting the lattice heat capacity. The solid curves denote the corresponding theoretical results.

peak appears whose position moves to high temperature side with increasing H. Also the peak width becomes broader as H is increased. In order to get the magnetic part of the heat capacity, C_m, we have subtracted the lattice heat capacity, C_l, from C. Here, we assumed that the C_l varies with temperature, T, as $C_l = \beta T^3$ with β=0.0149 and was independent of magnetic fields. We show in Fig. 2 the temperature dependence of magnetic heat capacity of this compound, after subtracting the lattice heat capacity, for the designated magnetic fields. We see a broad peak at about 13 K in addition to the low temperature peak already seen in the raw data. The former broad maximum reflects the entropy change associated with a short range antiferromagnetic ordering in low dimension[5]. On the other hand, the appearance of a peak in C_m at the low temperature suggests the presence of an energy gap.

We have analyzed the temperature and magnetic field dependence of C_m based on a simple energy level scheme, namely, a singlet ground state and first excited triplet with a spin gap, Δ. The result of the analysis is shown with the solid lines in Fig. 2. We were able to reproduce the experiment rather nicely with $\Delta/k_B =$ 0.37 K and g=2.20. The g value obtained in this study is consistent with that reported before[4]. Theory predicts that there are many $S_{tot} = 0$ states between the singlet ground state and the lowest excited triplet[2]. The introduction of these states to the analysis will improve the agreement between theory and experiment. By comparing the position of the high temperature peak in C_m with the results of the numerical calculations[6,7], we have the value $J/k_B \approx 19$ K for the nearest neighbor exchange interaction. From the numerical calculation[2] the lower bound is set to $\Delta \approx J/20$ for the spin gap. In the present case, this gives $\Delta/k_B \approx 1$ K, which is larger than that obtained experimentally ($\Delta/k_B = 0.37$ K).

In conclusion, we have observed two peak feature in the heat capacity of a new $S = \frac{1}{2}$ KHA $\{Cu_3(titmb)_2(OCOCH_3)_6\}\cdot H_2O$. The higher temperature peak is explained as due to a short range magnetic ordering in two dimension. The observation of the lower temperature peak gives evidence for the presence of a spin gap in this $S = \frac{1}{2}$ Kagomé lattice antiferromagnet.

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