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

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Abstract

A new spin $S=1/2$ antiferromagnet on the kagomé lattice, $\{\text{Cu}_3(\text{titmb})_2(\text{OCOCH}_3)_6\cdot\text{H}_2\text{O}\}$ (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 $S=1/2$ 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 $S=1/2$ KHA, theories predict that the ground state is in a disordered quantum spin liquid with a small spin gap to the excited states cite1,2. The spin gap estimated to be of the order of 1/20 of the exchange interaction constantcite2. The ground state of an $S=1/2$ KHA may be described by a quantum dimmer mode cite3.

In this paper, we report the results of heat capacity measurements made on a new $S=1/2$ KHA, $\{\text{Cu}_3(\text{titmb})_2(\text{OCOCH}_3)_6\cdot\text{H}_2\text{O}\}$ in which Cu$^{2+}$ has $S=1/2$. The anisotropy in the $g$-tensor of copper (II) ion is smallcite4 so that Heisenberg model can be applied to the exchange interaction among the moments.

The compound $\{\text{Cu}_3(\text{titmb})_2(\text{OCOCH}_3)_6\cdot\text{H}_2\text{O}\}$ has the hexagonal structure with the lattice parameters, $a=15.539\text{Å}$ and $c=21.149\text{Å}cite4$. The crystal structure consists of Cu-CH$_3$COO 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 $\{\text{Cu}_3(\text{titmb})_2(\text{OCOCH}_3)_6\cdot\text{H}_2\text{O}\}$ 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 $\{\text{Cu}_3(\text{titmb})_2(\text{OCOCH}_3)_6\cdot\text{H}_2\text{O}\}$ 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
Fig. 1. The temperature dependence of the total heat capacity of \( \{ \text{Cu}_{3}(\text{titmb})_{2}(\text{OCOCH}_3)_6 \} \cdot \text{H}_2\text{O} \) 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 \( \{ \text{Cu}_{3}(\text{titmb})_{2}(\text{OCOCH}_3)_6 \} \cdot \text{H}_2\text{O} \) after subtracting the lattice heat capacity. The solid curves denote the corresponding theoretical results.

A 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 \( \beta=0.0149 \) 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 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.

In conclusion, we have observed two peak feature in the heat capacity of a new \( S = \frac{1}{2} \) KHA \( \{ \text{Cu}_{3}(\text{titmb})_{2}(\text{OCOCH}_3)_6 \} \cdot \text{H}_2\text{O} \). The higher temperature peak is explained as due to a short range magnetic ordering in two dimension. The appearance of the low temperature peak gives evidence for the presence of a spin gap in this \( S = \frac{1}{2} \) Kagomé lattice antiferromagnet.

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References