Specific heat of single crystal of spin ice compound Dy$_2$Ti$_2$O$_7$

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Abstract

Dy$_2$Ti$_2$O$_7$ is regarded as a good example of geometrically frustrated spin systems exhibiting spin ice behavior. We report specific heat $C_p$ of single crystalline Dy$_2$Ti$_2$O$_7$ in the characteristic magnetic-field direction [100] down to 0.35 K and up to 5 T. We observed a single peak in $C_p$ with the peak temperature dependent on the field strength. For this direction, the component of the magnetic moment parallel to the field is equivalent for every spin in each tetrahedron. Thus in high fields, this peak is expressed as a Schottky peak characterized by the Zeemann splitting; in lower fields, however, the peak becomes representative of the energy of spin-spin interactions.

Key words: Dy$_2$Ti$_2$O$_7$; Spin Ice; Specific heat; Pyrochlore

Recently, pyrochlore oxides $A_2B_2O_7$ have been recognized as important systems to study geometrical frustration. In Dy$_2$Ti$_2$O$_7$ the "spin ice" behavior has clearly been observed in the specific heat [1]. This material is thought to have macroscopically degenerate ground state because of the residual entropy and absence of long-range magnetic ordering down to 50 mK [1,2].

The macroscopically degenerate ground state of these materials can be represented by analogy with water ice. [3] In Dy$_2$Ti$_2$O$_7$, the $A$ site ions, Dy$^{3+}$, constitute a corner-shared tetrahedral network. Because of the crystal-field effect, magnetic moments of the $A$ site ions exhibit Ising anisotropy along the local [111] direction. Furthermore, the effective nearest-neighbor ferromagnetic (FM) interaction makes the spin configuration of each tetrahedron to be 2-spins-in and 2-spins-out in the ground state. This constraint is referred to as the ice rule, and leads the ground state of this compound to be macroscopically degenerate with Pauling’s entropy $(1/2)R\ln(3/2)$ as the residual entropy [4].

Ramirez et al. [1] used polycrystalline sample of Dy$_2$Ti$_2$O$_7$ in their study of specific heat down to 0.25 K. There are three peaks in $C_{spin}(T)/T$ under magnetic field above 1.5 T. The peak temperatures are 0.34 K, 0.47 K and 1.12 K and independent of the field strength. In zero field, they found the residual entropy consistent with the expectation for the spin ice. They also found that the residual entropy readily disappears in magnetic field [1]. Because of the Ising anisotropy of Dy ions, the spin responses are different in the magnetic field along different directions. Therefore, it is important to investigate the physical properties of Dy$_2$Ti$_2$O$_7$ by using single crystals, in order to understand the origins of the specific heat peaks.

In this paper, we report the specific heat of single crystalline Dy$_2$Ti$_2$O$_7$ in the magnetic field. We focus on the data in the [100] field direction and examine the origin of the peak in the specific heat.

We grew single crystals of Dy$_2$Ti$_2$O$_7$ and Eu$_2$Ti$_2$O$_7$ by the floating zone method with an infrared furnace equipped with two elliptical mirrors. The details of the crystal growth were reported previously [2]. We measured specific heat between 0.35 and 40 K in fields up to 5 T by a relaxation method using a commercial calorimeter (Quantum Design, model PPMS). The sample size was approximately $2.0 \times 2.0 \times 0.1\text{mm}^3$ and the mass was about 1.5 mg. In order to avoid

Preprint submitted to LT23 Proceedings 15 June 2002
field inhomogeneity due to demagnetization effect, we prepared a thin slice of a crystal with the (110) plane as the flat plane.

For Dy$_2$Ti$_2$O$_7$ the specific heat is expressed as,

$$C_{\text{total}} = C_{\text{phonon}} + C_{\text{spin}}$$

(1)

$C_{\text{total}}$, $C_{\text{phonon}}$ and $C_{\text{spin}}$ represent the total specific heat, the phonon contribution and the spin contribution, respectively. In order to estimate the phonon contribution, we measure specific heat of Eu$_2$Ti$_2$O$_7$ because this material is also insulating, nonmagnetic, and with the lattice parameter almost the same as that of Dy$_2$Ti$_2$O$_7$.

The energy gap for each Dy$^{3+}$ ion between the nearly two-fold degenerate ground-state and the first exited state under crystal field is estimated to be approximately 150 K [5]. This permits us to consider only the nearly two-fold degenerate ground-state below 40 K. The energy scale of the nearest-neighbor FM interaction is estimated to be $J_{\text{eff}} = 1.2$ K in Dy$_2$Ti$_2$O$_7$ [6].

Along the [100] field direction, the components parallel to the field of four spins in each tetrahedron are equivalent. The value of $\cos \theta$ between the spin direction and the field direction is $1/\sqrt{3}$. Along this field direction the Zeemann energy at 1 T is expressed as $(1/\sqrt{3})g_IJ\mu_0\mu_B H_{\text{ext}} = 3.8$ K. Thus, in fields higher than about 0.3 T the Zeemann energy becomes larger than the energy scale of the nearest-neighbor FM interaction. Therefore, we expect a Schottky-like specific heat with the characteristic energy changing from the internal FM interaction to the Zeemann energy given by the external field. In addition, we expect that there are no field independent peaks.

In Fig. 1 we show the specific heat of single-crystalline Dy$_2$Ti$_2$O$_7$ down to 0.35 K under magnetic fields along the [100] direction. We observed only one peak in the specific heat in this field direction. With increasing field, we observed a changeover from the Schottky-like peak originating from effective FM interactions in lower fields.

In conclusion, we reported the specific heat of single crystalline Dy$_2$Ti$_2$O$_7$ down to 0.35 K under magnetic fields along the [100] direction. We observed only one peak in the specific heat in this field direction. With increasing field, we observed a changeover from the Schottky-like peak originating from effective FM interactions to the Zeemann energy associated with Ising spins Dy$^{3+}$ determined by the external field.

We acknowledge helpful discussion with M. Gingras and D. Yanagishima. H.F. is grateful for the financial support from JSPS Research Fellowships for Young Scientists.

References