

SPECIFIC HEAT OF THE TETRAGONAL ANTIFERROMAGNET $TbB_2C_2^*$

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Specific heat measurements of antiferromagnet TbB_2C_2 with $T_N = 21.7\text{ K}$ were carried out under magnetic fields up to 8 T applied along the [1 0 0] and [1 1 0] directions. The application of magnetic fields in TbB_2C_2 leads to increase of the transition temperature in both directions. In case for $H \parallel [1 1 0]$, the transition temperature reaches 31.8 K under 8 T which indicates that magnetic fields anomalously stabilize the antiferromagnetic ordered state. The obtained field dependence of the transition temperature is quite anisotropic between the [1 0 0] and [1 1 0] directions, which is similar to the antiferroquadrupolar compounds DyB_2C_2 and HoB_2C_2 .

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1. Introduction

Rare earth intermetallic compounds RB_2C_2 with the tetragonal LaB_2C_2 -type structure [1, 2] show various magnetic properties which originate in relatively strong antiferroquadrupolar (AFQ) interactions. DyB_2C_2 which is the first material exhibiting the AFQ order among tetragonal rare earth compounds has an AFQ order transition at $T_Q=24.7\text{ K}$ and an antiferromagnetic (AFM) transition successively at $T_N=15.3\text{ K}$ [3]. T_Q of DyB_2C_2 is about ten times higher than those of other AFQ compounds reported so far. An AFQ order in HoB_2C_2 is realized at $T_Q=4.5\text{ K}$ below an AFM order at $T_N=5.9\text{ K}$, in other words, HoB_2C_2 undergoes the AFQ order transition in the magnetic ordered state [4]. As a result, the AFM ordered state, called phase IV, appears between T_N and T_Q where many characteristic magnetic

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properties were observed [5, 6]. Owing to the existence of AFQ order, the $H-T$ magnetic phase diagrams of DyB_2C_2 and HoB_2C_2 have commonly unusual characteristics; the coexistent phases of AFQ and AFM ordered states are slightly stabilized by applied magnetic fields and the phase diagrams are quite anisotropic between $H \parallel [100]$ and $[110]$.

An isostructural compound TbB_2C_2 is an antiferromagnet with $T_N=21.7\text{ K}$ [7]. The magnetic structure can be described basically by $\mathbf{k}_2 = (011/2)$; the magnetic moments which lie in the basal c -plane couple antiferromagnetically between the corner and the face center of the unit cell, and the coupling along the $[001]$ direction is antiferromagnetic as well. Besides, the propagation vectors of $\mathbf{k}_4 = (001/2)$ and $\mathbf{k}_L = (1\pm\delta\pm\delta 0)$ ($\delta = 0.13$) are also required. Note that $\mathbf{k}_L = (1\pm\delta\pm\delta 0)$ component in TbB_2C_2 accompanies the characteristic diffuse scattering which is quite resemblant to that observed in phase IV of HoB_2C_2 [5, 8]. In addition to the characteristic magnetic structure, it is remarkable that the magnetization process under high fields exhibits similar behavior to those of the AFQ compound DyB_2C_2 and HoB_2C_2 . Based on this result, it is highly probable that AFQ interactions strongly affect in TbB_2C_2 as well. Therefore, the aim of this work is to clarify the magnetic properties of TbB_2C_2 under magnetic fields by means of specific heat measurements.

2. Experimental

For sample preparation, we used the stoichiometric amounts of constituents, Tb of 99.9%, B of 99.8% and C of 99.999% in purity. The compound was synthesized through the conventional argon arc technique. Single crystalline samples of TbB_2C_2 were grown by the Czochralski method using a tri-arc furnace. Specific heats of TbB_2C_2 were measured by using conventional relaxation method. Measurements were carried out in the temperature range from 0.5 K to 60 K and under magnetic fields up to 8 T. The specific heat of LaB_2C_2 was also measured in the same temperature range under $H = 0$ to estimate the lattice contribution to the specific heat.

3. Results and discussion

Fig. 1 shows magnetic specific heats of TbB_2C_2 under various magnetic fields applied along the $[100]$ direction. The magnetic contribution to the specific heat is obtained by subtracting the specific heat of LaB_2C_2 from that of TbB_2C_2 . A clear λ -type anomaly was observed at $T_N=21.7\text{ K}$ under $H=0$, which is well consistent with the results reported before [7]. As field increases, T_N exhibits monotonous increase and corresponding anomaly becomes slightly broad. Under 4 T, the transition temperature takes the

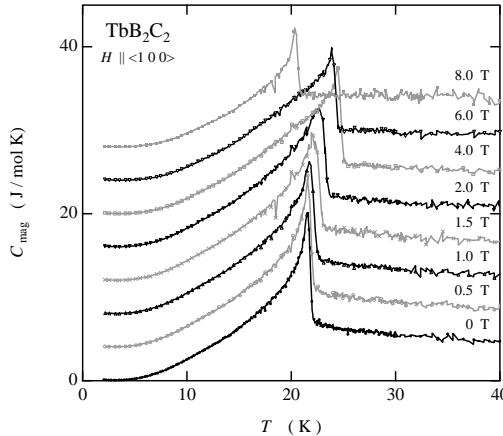


Fig. 1. Magnetic specific heats of TbB_2C_2 for $H \parallel [100]$. Curves under magnetic fields are vertically shifted for the clarity.

maximum of 25.2 K and suddenly decreases to 16.6 K under 8 T. In the magnetization curve for $H \parallel [100]$ at 4.2 K, the magnetic transitions were observed at 7.6 T and 8.6 T. This result indicates that the phase boundary corresponding to the anomaly in the specific heat closes between 7 to 9 T for $H \parallel [100]$. The anomalous increase of T_N with increasing magnetic fields is further remarkable in case of the field applied along the $[110]$ direction as shown in Fig. 2. Up to 8 T, the anomaly is still clear and the transition temperature keeps increasing to 31.8 K under 8 T, that is, the AFM transition temperature increases almost 10 K by the application of magnetic fields.

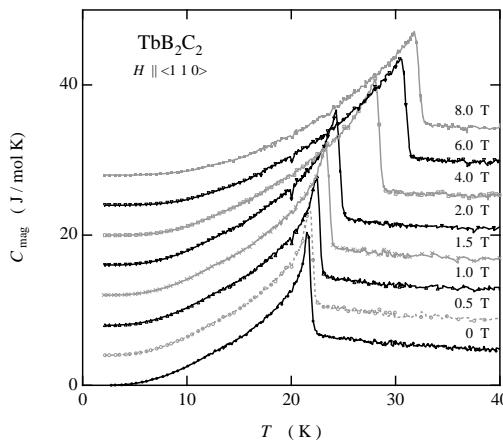


Fig. 2. Magnetic specific heats of TbB_2C_2 for $H \parallel [110]$. Curves under magnetic fields are vertically shifted for the clarity.

The unusual behavior of T_N increasing by applying magnetic fields cannot be explained only by AFM interactions. With respect to the increase of T_N , the similar behavior was also observed in the AFQ ordered phases of DyB_2C_2 and HoB_2C_2 . Furthermore, the magnetic field dependence of the transition temperature in TbB_2C_2 is quite anisotropic between [100] and [110], which is identical to that in DyB_2C_2 and HoB_2C_2 as well. Based on these facts, it is supposed that the AFQ order is realized in TbB_2C_2 under magnetic fields. Actually, our recent neutron diffraction experiments indicate that the magnetic structure of TbB_2C_2 under magnetic fields exhibits a characteristic coupling angle between the magnetic moments along the [001] direction which appears in the coexistent phase of AFM and AFQ order in DyB_2C_2 and HoB_2C_2 [9, 10]. One of the typical AFQ ordered compound CeB_6 also exhibits the similar increase of transition temperature with increasing fields. In case of CeB_6 , a theoretical calculation succeeded to explain the anomalous stability under magnetic fields by taking account octupolar interactions [11]. Therefore, we strongly suggest that TbB_2C_2 is the first material which shows the field-induced AFQ order and octupolar moment may play a more important role than DyB_2C_2 and HoB_2C_2 .

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