Vol. 34 (2003) ACTA PHYSICA POLONICA B No 2

SPECIFIC HEAT OF THE TETRAGONAL ANTIFERROMAGNET TbB_2C_2*

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(Received July 10, 2002)

Specific heat measurements of antiferromagnet TbB_2C_2 with $T_N = 21.7$ 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 $H||[1\,1\,0]$, 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.

PACS numbers: 75.30.Kz, 75.90.+w, 75.40.Cx

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 K and an antiferromagnetic (AFM) transition successively at T_N=15.3 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 K below an AFM order at T_N=5.9 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

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.

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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 $H\|[1\,0\,0]$ and $[1\,1\,0]$.

An isostructural compound TbB_2C_2 is an antiferromagnet with T_N=21.7 K [7]. The magnetic structure can be described basically by $\boldsymbol{k}_2 = (0\,1\,1/2)$; 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 $[0\,0\,1]$ direction is antiferromagnetic as well. Besides, the propagation vectors of $\boldsymbol{k}_4 = (0\,0\,1/2)$ and $\boldsymbol{k}_L = (1\pm\delta\pm\delta\,0)$ ($\delta = 0.13$) are also required. Note that $\boldsymbol{k}_L = (1\pm\delta\pm\delta\,0)$ 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 $[1\,0\,0]$ 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 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

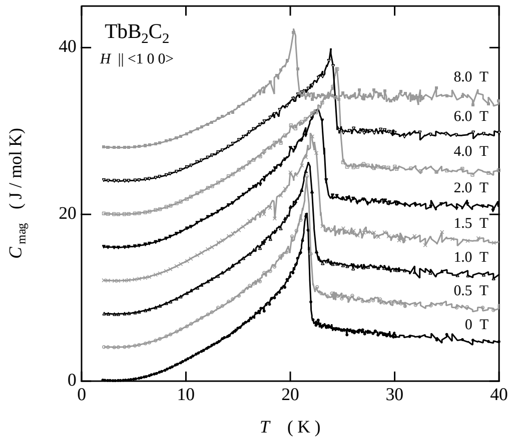

Fig. 1. Magnetic specific heats of TbB_2C_2 for $H \| [1\,0\,0]$. Curves under magnetic fields are vertically shifted for the clarity.

maximum of 25.2 K and suddenly decreases to 16.6 K under 8 T. In the magnetization curve for $H \| [1\,0\,0]$ at 4.2K, 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 $H \| [1\,0\,0]$. The anomalous increase of T_N with increasing magnetic fields is further remarkable in case of the field applied along the [1 1 0] 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.

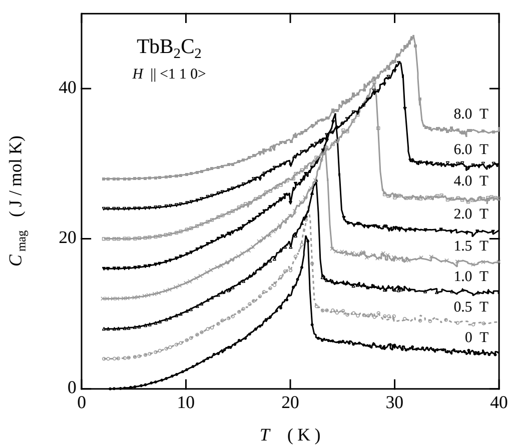

Fig. 2. Magnetic specific heats of TbB_2C_2 for $H \| [1\,1\,0]$. Curves under magnetic fields are vertically shifted for the clarity.

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 [1 0 0] and [1 1 0], 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 [0 0 1] 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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