SPECIFIC HEAT OF THE TETRAGONAL ANTIFERROMAGNET TbB$_2$C$_2$*

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Specific heat measurements of antiferromagnet TbB$_2$C$_2$ with $T_N = 21.7$K were carried out under magnetic fields up to 8T applied along the [100] and [110] directions. The application of magnetic fields in TbB$_2$C$_2$ leads to increase of the transition temperature in both directions. In case for $H||[110]$, the transition temperature reaches 31.8K under 8T which indicates that magnetic fields anomalously stabilize the antiferromagnetic ordered state. The obtained field dependence of the transition temperature is quite anisotropic between the [100] and [110] directions, which is similar to the antiferroquadrupolar compounds DyB$_2$C$_2$ and HoB$_2$C$_2$.

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1. Introduction

Rare earth intermetallic compounds RB$_2$C$_2$ with the tetragonal LaB$_2$C$_2$-type structure [1, 2] show various magnetic properties which originate in relatively strong antiferroquadrupolar (AFQ) interactions. DyB$_2$C$_2$ 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$_2$C$_2$ is about ten times higher than those of other AFQ compounds reported so far. An AFQ order in HoB$_2$C$_2$ is realized at $T_Q=4.5$ K below an AFM order at $T_N=5.9$ K, in other words, HoB$_2$C$_2$ 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$_2$C$_2$ and HoB$_2$C$_2$ have commonly un-
usual 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$_2$C$_2$ is an antiferromagnet with $T_N=21.7\,\text{K}$
[7]. The magnetic structure can be described basically by $k_2 = (011/2)$;
the magnetic moments which lie in the basal $c$-plane couple antiferromagneti-
cally 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 $k_1 = (001/2)$ and $k_1 = (1\pm\delta \pm 0\delta)$ ($\delta = 0.13$) are
also required. Note that $k_1 = (1\pm\delta \pm 0\delta)$ component in Tbb$_2$C$_2$ accompa-
nies the characteristic diffuse scattering which is quite resemblant to that
observed in phase IV of HoB$_2$C$_2$ [5,8]. In addition to the characteristic mag-
netic structure, it is remarkable that the magnetization process under high
fields exhibits similar behavior to those of the AFQ compound DyB$_2$C$_2$ and
HoB$_2$C$_2$. Based on this result, it is highly probable that AFQ interactions
strongly affect in Tbb$_2$C$_2$ as well. Therefore, the aim of this work is to
clarify the magnetic properties of Tbb$_2$C$_2$ under magnetic fields by means
of specific heat measurements.

2. Experimental

For sample preparation, we used the stoichiometric amounts of con-
stituents, Tb of 99.9%, B of 99.8% and C of 99.999% in purity. The com-
 pound was synthesized through the conventional argon arc technique. Single
crystalline samples of Tbb$_2$C$_2$ were grown by the Czochralski method using
a tri-arc furnace. Specific heats of Tbb$_2$C$_2$ were measured by using
conventional relaxation method. Measurements were carried out in the tem-
perature range from 0.5K to 60 K and under magnetic fields up to 8T. The
specific heat of LaB$_2$C$_2$ 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$_2$C$_2$ 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$_2$C$_2$ from
that of Tbb$_2$C$_2$. A clear $\lambda$-type anomaly was observed at $T_N = 21.7\,\text{K}$ un-
der $H = 0$, 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 4T, the transition temperature takes the
maximum of 25.2K and suddenly decreases to 16.6K under 8T. In the magnetization curve for $H \parallel [100]$ at 4.2K, the magnetic transitions were observed at 7.6 T and 8.6T. 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 8T, the anomaly is still clear and the transition temperature keeps increasing to 31.8K under 8T, that is, the AFM transition temperature increases almost 10K by the application of magnetic fields.

Fig. 1. Magnetic specific heats of TbB$_2$C$_2$ for $H \parallel [100]$. Curves under magnetic fields are vertically shifted for the clarity.

Fig. 2. Magnetic specific heats of TbB$_2$C$_2$ 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$_2$C$_2$ and HoB$_2$C$_2$. Furthermore, the magnetic field dependence of the transition temperature in TbB$_2$C$_2$ is quite anisotropic between [1 0 0] and [1 1 0], which is identical to that in DyB$_2$C$_2$ and HoB$_2$C$_2$ as well. Based on these facts, it is supposed that the AFQ order is realized in TbB$_2$C$_2$ under magnetic fields. Actually, our recent neutron diffraction experiments indicate that the magnetic structure of TbB$_2$C$_2$ 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$_2$C$_2$ and HoB$_2$C$_2$ [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$_2$C$_2$ is the first material which shows the field-induced AFQ order and octupolar moment may play a more important role than DyB$_2$C$_2$ and HoB$_2$C$_2$.

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