ANOMALOUS MAGNETIC PHASES IN $Ho_{1-x}Tb_xB_2C_2$*

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(Received July 10, 2002)

HoB_2C_2 and TbB_2C_2 show anomalous magnetic phases which are adjacent to antiferroquadrupolar ordered phases and supposedly affected by multipolar interactions. We carried out specific heat and magnetization measurements to complete magnetic phase diagrams of pseudo-binary $Ho_{1-x}Tb_xB_2C_2$. It is clarified that the interactions in the anomalous magnetic phases of HoB_2C_2 and TbB2C_2 are quite similar and Tb^{3+} ions are cooperative to the antiferroquadrupolar order in HoB_2C_2.

PACS numbers: 75.50.Ee, 75.90.+w

1. Introduction

Tetragonal LaB_2C_2-type [1] compounds RB_2C_2 show a series of interesting magnetic phenomena. HoB_2C_2 undergoes an antiferromagnetic (AFM) ordering at $T_N = 5.9$ K and an antiferroquadrupolar (AFQ) ordering at $T_Q = 4.5$ K [2]. By neutron diffraction experiments, Ohoyama *et al.* have revealed that its ground state magnetic structure is described with four propagation vectors of $\boldsymbol{k}_1 = [1\ 0\ 0]$, $\boldsymbol{k}_2 = [1\ 0\ 1/2]$, $\boldsymbol{k}_3 = [0\ 0\ 0]$, $\boldsymbol{k}_4 = [0\ 0\ 1/2]$ [3]. The magnetic moments neighboring along the [0 0 1]-axis are arranged by an angle of about 90 degrees by the AFQ interactions. Furthermore, they observed that the magnetic structure in the AFM phase between T_Q and T_N is a long-periodic one with a propagation vector $\boldsymbol{k}_1 = [1 \pm \delta_1\ \ \pm\delta_1\ \delta_2]$ ($\delta_1 = 0.11,\ \delta_2 = 0.04$) accompanied with diffuse components which appear in a broad reciprocal space between the satellites belonging to $\boldsymbol{k}_1$ [3,4].

While TbB_2C_2 shows only an AFM transition at $T_N = 21.7$ K, Kaneko *et al.*, have found that magnetic fields induce an AFQ order coexisting with an AFM order in TbB_2C_2 [5]. As same as the magnetic structure in the ground state of HoB_2C_2, the magnetic structure of the field-induced

* Presented at the International Conference on Strongly Correlated Electron Systems, (SCES 02), Cracow, Poland, July 10–13, 2002.
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(AFM+AFQ) state in TbB_2C_2 is described by four propagation vectors, $\boldsymbol{k}_1 = [1\ 0\ 0]$, $\boldsymbol{k}_2 = [1\ 0\ 1/2]$, $\boldsymbol{k}_3 = [0\ 0\ 0]$, $\boldsymbol{k}_4 = [0\ 0\ 1/2]$. The AFM ground state has a long-periodic magnetic structure with propagation vectors of $\boldsymbol{k}_0 = [0\ 1\ 1/2]$, $\boldsymbol{k}_1 = [0\ 0\ 1/2]$, $\boldsymbol{k}_2 = [1 \pm \delta\ \pm \delta\ 0]$ ($\delta = 0.13$) and diffuse components very similar to those in HoB_2C_2 [6]. There are some resemblances between HoB_2C_2 and TbB_2C_2 that the magnetic structures of the their AFM phases are described with very similar long periodicities and diffuse components. Then we carried out specific heat and magnetization measurements on the sigle crystalline samples of pseudo-binary $Ho_{1-x}Tb_xB_2C_2$ compounds in order to clarify whether the dominant interactions in those AFM phases are identical or not.

2. Experimental

We synthesized $Ho_{1-x}Tb_xB_2C_2$ ($0.2 \le x \le 0.8$) by a conventional argon arc technique. To ensure homogeneity, each ingot was turned over and re-melted several times. The single crystals of $Ho_{1-x}Tb_xB_2C_2$ were prepared by the Czochralski technique using a tetra-arc furnace. Specific heats were measured by using a conventional relaxation method and magnetic measurements were carried out using a SQUID magnetometer.

3. Results and discussion

Fig. 1 shows temperature dependence of the magnetic specific heats of $Ho_{1-x}Tb_xB_2C_2$ ($0.0 \le x \le 1.0$). While T_N increases gradually from 5.9 K to 21.7 K as x increases, T_Q hardly changes up to $x = 0.6$ and disappears

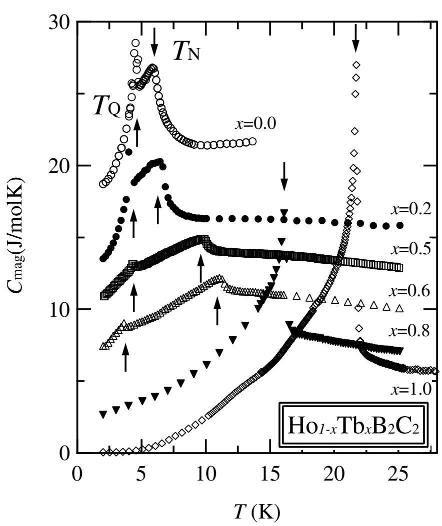

Fig. 1. Temperature dependence of the specific heats of $Ho_{1-x}Tb_xB_2C_2$.

above $x = 0.6$. As TbB_2C_2 undergoes no AFQ ordering without magnetic field [5], it is quite mysterious that the T_Q value hardly decreases up to $x = 0.6$. Furthermore, the anomalous specific heat at T_Q becomes smaller as x increases, which suggests that the Tb^{3+} ions do not participate the AFQ order consisting of the Ho^{3+} ions. Then we think that the Tb^{3+} ions are not destructive but supportive for the AFQ order consisting of Ho^{3+} ions by some reasons like quadrupolar and/or octupolar interactions, although TbB_2C_2 itself shows no AFQ ordering without magnetic field, that is, the AFQ order in HoB_2C_2 seems to be quite stable against the Tb substitution.

Fig. 2 represents the H–T magnetic phase diagrams ($H \parallel [1\,1\,0]$-axis) of $Ho_{1-x}Tb_xB_2C_2$ ($x = 0.0$, 0.4, 0.6, 1.0). The AFM phase (phase IV) becomes to extend to lower temperatures continuously as x increases. This

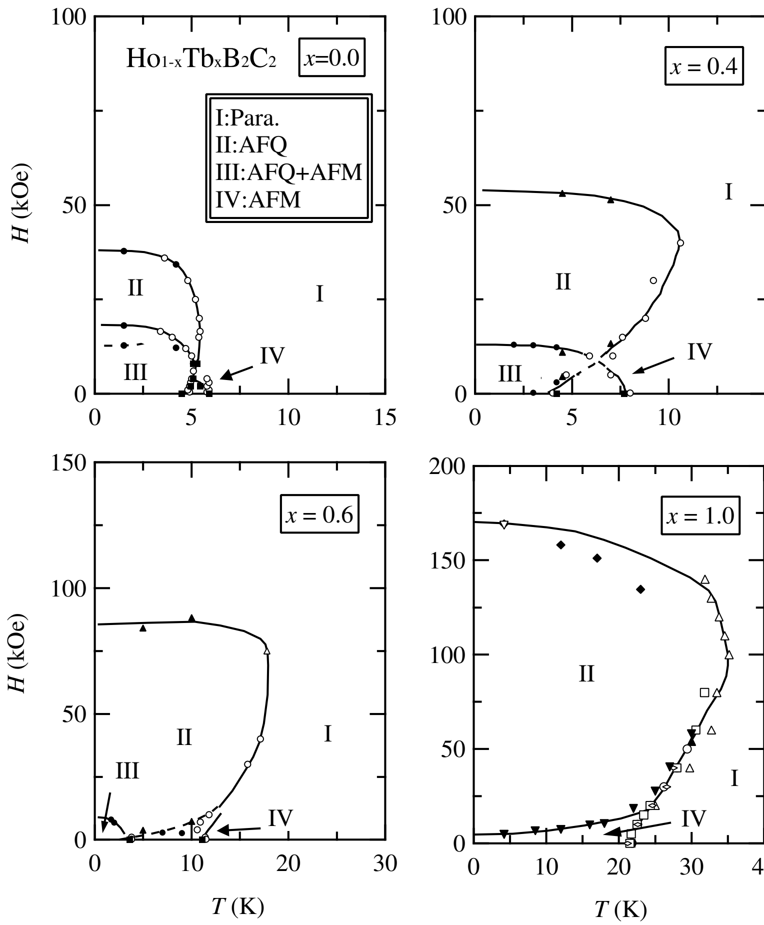

Fig. 2. Magnetic phase diagrams of $Ho_{1-x}Tb_xB_2C_2$.

fact implies that the interactions in phase IV of both HoB_2C_2 and TbB_2C_2 are very similar. Moreover, it is observed that the AFQ phase (phase II) goes on expanding and the (AFQ+AFM) phase (phase III) disappears gradually as x increases. Kaneko *et al.* have supposed that phase II in TbB_2C_2 is the AFQ one as same as HoB_2C_2 [5]. The supposition is just based on the similarity of magnetization processes of HoB_2C_2 and TbB_2C_2. By the present results on the H–T magnetic phase diagrams of $Ho_{1-x}Tb_xB_2C_2$, we confirm that phase II in TbB_2C_2 is the AFQ phase continuing to that in HoB_2C_2. It is noted that the AFM phase (phase IV) and the AFQ phase (phase II) are side by side without any intermediate phase, although phase IV and phase II are directly adjacent to each other in TbB_2C_2 but they are not in contact with each other in HoB_2C_2 [7].

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