ANOMALOUS MAGNETIC PHASES IN Ho_{1-x}Tb_xB_2C_2

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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 pseudobinary Ho_{1-x}Tb_xB_2C_2. It is clarified that the interactions in the anomalous magnetic phases of HoB_2C_2 and TbB_2C_2 are quite similar and Tb^{3+} ions are cooperative to the antiferroquadrupolar order in HoB_2C_2.

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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 \( k_1 = [1 0 0] \), \( k_2 = [1 0 1/2] \), \( k_3 = [0 0 0] \), \( k_4 = [0 0 1/2] \) [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 \( k_1 = [1 \pm \delta_1 \pm \delta_1 \delta_2] \) (\( \delta_1 = 0.11 \), \( \delta_2 = 0.04 \)) accompanied with diffuse components which appear in a broad reciprocal space between the satellites belonging to \( k_1 \) [3,4].

While TbB_2C_2 shows only an AFM transition at T_N = 21.7K, 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

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(AFM+AFQ) state in TbB$_2$C$_2$ is described by four propagation vectors, $k_1 = [1\ 0\ 0]$, $k_2 = [1\ 0\ 1/2]$, $k_3 = [0\ 0\ 0]$, $k_4 = [0\ 0\ 1/2]$. The AFM ground state has a long-periodic magnetic structure with propagation vectors of $k_0 = [0\ 1\ 1/2]$, $k_1 = [0\ 0\ 1/2]$, $k_2 = [1\pm\delta\ \pm\delta\ 0]$ ($\delta = 0.13$) and diffuse components very similar to those in HoB$_2$C$_2$ [6]. There are some resemblances between HoB$_2$C$_2$ and TbB$_2$C$_2$ 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 single crystalline samples of pseudo-binary Ho$_{1-x}$Tb$_x$B$_2$C$_2$ 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$_x$B$_2$C$_2$ (0.2 $\leq x \leq$ 0.8) by a conventional argon arc technique. To ensure homogeneity, each ingot was turned over and remelted several times. The single crystals of Ho$_{1-x}$Tb$_x$B$_2$C$_2$ 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$_x$B$_2$C$_2$ (0.0 $\leq x \leq$ 1.0). While $T_N$ increases gradually from 5.9K to 21.7K as $x$ increases, $T_Q$ hardly changes up to $x = 0.6$ and disappears

![Graph](image_url)

Fig. 1. Temperature dependence of the specific heats of Ho$_{1-x}$Tb$_x$B$_2$C$_2$. 

$C_{mag} (J/molK)$

$T_N$
above $x = 0.6$. As TbB$_2$C$_2$ 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$_2$C$_2$ itself shows no AFQ ordering without magnetic field, that is, the AFQ order in HoB$_2$C$_2$ 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$_x$B$_2$C$_2$ ($x = 0.0, 0.4, 0.6, 1.0$). The AFM phase (phase IV) becomes to extend to lower temperatures continuously as $x$ increases. This

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Fig. 2. Magnetic phase diagrams of Ho$_{1-x}$Tb$_x$B$_2$C$_2$. 
fact implies that the interactions in phase IV of both HoB$_2$C$_2$ and TbB$_2$C$_2$
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$_2$C$_2$
is the AFQ one as same as HoB$_2$C$_2$ [5]. The supposition is just based on the
similarity of magnetization processes of HoB$_2$C$_2$ and TbB$_2$C$_2$. By the
present results on the $H$-$T$ magnetic phase diagrams of Ho$_{1-x}$Tb$_x$B$_2$C$_2$, we
confirm that phase II in TbB$_2$C$_2$ is the AFQ phase continuing to that in
HoB$_2$C$_2$. 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$_2$C$_2$ but they are not in contact with each other in HoB$_2$C$_2$ [7].

REFERENCES

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