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Magnetic ordering affected by multipolar interactions in $Ho_{1-x}Tb_xB_2C_2$

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Abstract

We performed a powder neutron diffraction experiment on $Ho_{1-x}Tb_xB_2C_2$ ($x = 0.2$, 0.4, 0.6 and 0.8) to compare the magnetic properties of HoB_2C_2 and TbB_2C_2. It is found that the magnetic satellite peaks described as $\boldsymbol{k} = [1 \pm \delta_1, \pm\delta_1, \pm\delta_2]$ get larger even in the (antiferromagnetic + antiferroquadrupolar) phase for $x = 0.2$, 0.4 and 0.6. And a satellite peak around 000 newly emerges for $x = 0.4$, 0.6 and 0.8.

1. Introduction

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 [1]. The magnetic structure in the AFM phase is a long-periodic one described by a propagation vector $\boldsymbol{k} = [1 \pm \delta_1, \pm\delta_1, \pm\delta_2]$ ($\delta_1 = 0.11$, $\delta_2 = 0.04$). And magnetic diffuse components around $\boldsymbol{k}$ are observed [2]. TbB_2C_2 only undergoes an AFM ordering at $T_N = 21.7$ K [3]. An AFQ ordering transition is induced by external magnetic fields. The propagation vectors in the main AFM component are described as $\boldsymbol{k}_0 = [0, 1, 1/2]$, $\boldsymbol{k}_1 = [0, 0, 1/2]$ and $\boldsymbol{k}_2 = [1 \pm \delta, \pm\delta, 0]$ ($\delta = 0.13$) [3]. And the diffuse components are also observed around $\boldsymbol{k}_2$-vector satellite peaks like for the AFM phase of HoB_2C_2.

The anomalous magnetic properties in the AFM phases of HoB_2C_2 and TbB_2C_2 are quite similar as regards the long-periodic magnetic structure and diffuse component. These magnetic anomalies are possibly due to the influence of quadrupolar interactions. Therefore we carried out specific heat measurements on $Ho_{1-x}Tb_xB_2C_2$ to compare the interactions in the AFM phases of HoB_2C_2 and TbB_2C_2. Our result is that T_Q is hardly dependent on the concentration from $x = 0.0$ to 0.6 while the peak height of the specific heat at T_Q gradually decreases [4]. This result indicates that the Tb^{3+} ions seem to be cooperative with the AFQ ordering in HoB_2C_2, although TbB_2C_2 itself shows no AFQ ordering under zero magnetic field. It seems that the Tb^{3+} ions play some role in the AFQ ordering in HoB_2C_2. Then we a performed powder neutron diffraction experiment on $Ho_{1-x}Tb_xB_2C_2$ to study the effect of Tb^{3+} substitution on the (AFQ + AFM) ordering phase in HoB_2C_2.

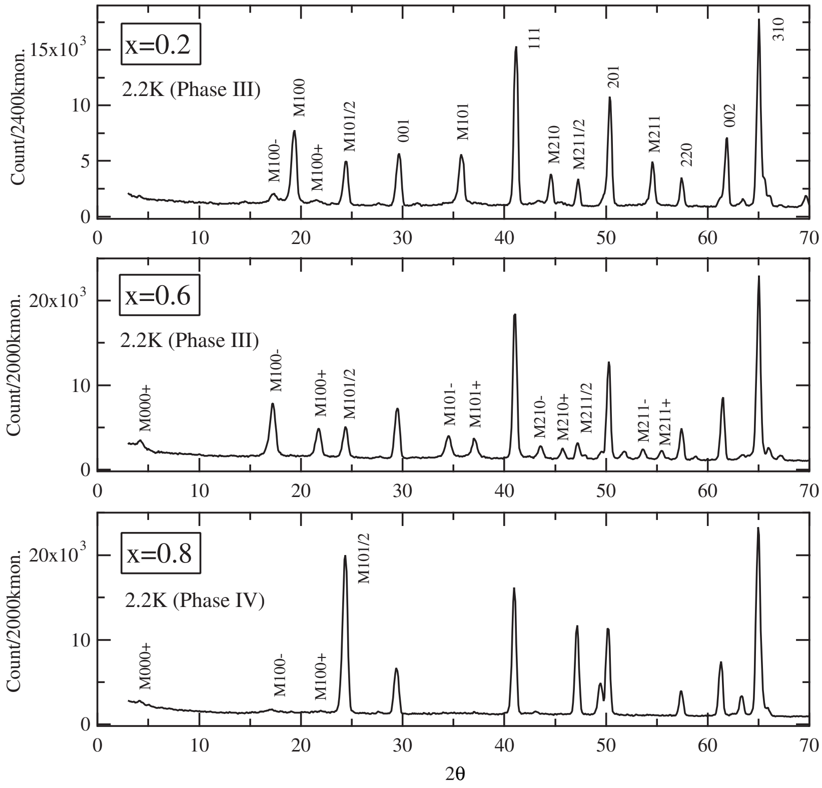

Figure 1. Powder neutron diffraction patterns for $Ho_{1-x}Tb_xB_2C_2$ at 2.2 K.

2. Experimental details

We synthesized $Ho_{1-x}Tb_x{}^{11}B_2C_2$ ($x = 0.2$, 0.4, 0.6 and 0.8) by the conventional argon arc technique. To ensure homogeneity, each ingot was turned over and remelted several times. 99.95% enriched ^{11}B was used instead of natural B to decrease the neutron absorption by the samples. We performed neutron powder diffraction experiments on the powder diffractometer for high efficiency and high resolution measurements HERMES, installed at the JRR-3M reactor in Japan [5].

3. Results and discussion

Figure 1 shows powder neutron diffraction patterns for $x = 0.2$, 0.6 and 0.8 at 2.2 K. The crystal structures of the samples are confirmed to be LaB_2C_2-type tetragonal ones. The diffraction pattern for $x = 0.2$ is nearly the same as that of HoB_2C_2, which is described by the four propagation vectors $\boldsymbol{k}_0 = [1, 0, 0]$, $\boldsymbol{k}_1 = [0, 1, 1/2]$, $\boldsymbol{k}_2 = [0, 0, 1/2]$ and $\boldsymbol{k}_3 = [0, 0, 0]$ [2]. However, satellite peaks described by $\boldsymbol{k} = [1 \pm \delta_1, \pm\delta_1, \pm\delta_2]$ emerge in $x = 0.2$. Moreover, the intensities of the satellite peaks for $x = 0.6$ become much larger than those for $x = 0.2$, while the original Bragg peaks described by $\boldsymbol{k} = [1, 0, 0]$ almost disappear for $x = 0.6$. Not only the M100 satellite peaks but also the M101, M210 and M211 satellite peaks become sharper and clearer as x increases. For $x = 0.8$ at 2.2 K, all satellite peaks become small. The

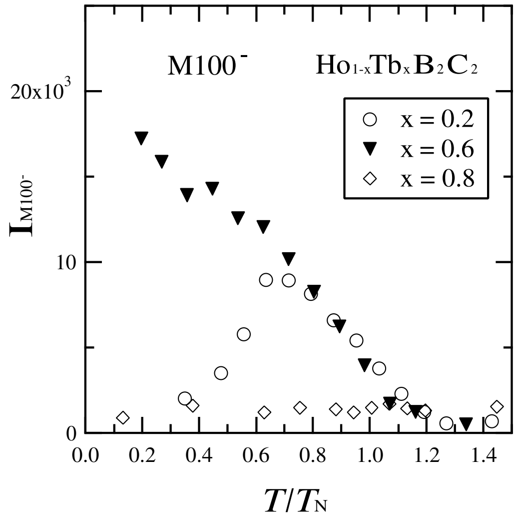

Figure 2. Integrated intensities of M100$^-$ for $x = 0.2$, 0.6 and 0.8.

powder pattern for $x = 0.8$ at 2.2 K is nearly the same as that of the AFM phase of TbB_2C_2 which is described mainly by three propagation vectors $\boldsymbol{k}_0 = [1, 0, 1/2]$, $\boldsymbol{k}_1 = [0, 0, 1/2]$ and $\boldsymbol{k}_2 = [1 \pm \delta, \pm\delta, 0]$. This is consistent with the results of specific heat measurements in showing that the ground state phase of $Ho_{1-x}Tb_xB_2C_2$ is (AFQ + AFM) for $x = 0.2$–0.6 and AFM for $x = 0.8$.

Specific heat measurements clarified that the ground state is the (AFQ + AFM) phase for $x = 0.0$–0.6 for the $Ho_{1-x}Tb_xB_2C_2$ system. The propagation vectors for the (AFQ + AFM) phases of HoB_2C_2 and TbB_2C_2 were reported to be a combination of $\boldsymbol{k}_0 = [1, 0, 0]$, $\boldsymbol{k}_1 = [0, 1, 1/2]$, $\boldsymbol{k}_2 = [0, 0, 1/2]$ and $\boldsymbol{k}_3 = [0, 0, 0]$. Nevertheless, the magnetic structure of the (AFQ + AFM) phase of HoB_2C_2 is described by four commensurate propagation vectors; those for $x = 0.2$ and 0.6 are long-periodic ones because $\boldsymbol{k}_0 = [1, 0, 0]$ is transformed gradually to the incommensurate vector $\boldsymbol{k} = [1 \pm \delta_1, \pm\delta_1, \pm\delta_2]$. This indicates that the magnetic structure of the (AFQ + AFM) phase of HoB_2C_2 changes to a longitudinal one on substituting Tb^{3+} for Ho^{3+}. The long-periodic magnetic component becomes larger as x increases and it has its maximum at $x = 0.6$.

Figure 2 shows the temperature dependence of the integrated intensities of M100$^-$. The instrumental factors which were estimated from the 111 nuclear Bragg peak were corrected. In the correction, we took account of the difference in nuclear scattering length b_n between Ho and Tb for each compound. As T/T_N decreases, the intensity for $x = 0.2$ increases gradually and decreases below $T/T_N = 0.7$. This behaviour is nearly the same as that for HoB_2C_2. However, the intensity for $x = 0.6$ continues to increase down to the lowest temperature, which indicates that the Tb^{3+} substitution enhances the longitudinal magnetic component under T_N.

A 000 satellite peak is newly discovered for $x = 0.4$, 0.6 and 0.8. This satellite peak remains even above T_N for each sample. This 000 satellite possibly originates from an impurity phase, but at present we have no information about this.

The diffuse component around $\boldsymbol{k} = [1 \pm \delta_1, \pm\delta_1, \pm\delta_2]$ satellite peaks is difficult to observe in powder patterns of $Ho_{1-x}Tb_xB_2C_2$. Single-crystal neutron scattering experiments on $Ho_{1-x}Tb_xB_2C_2$ are now in progress in order to make precise observations of diffuse components.

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