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Magnetic phase diagrams of $Dy_{0.8}Gd_{0.2}B_2C_2$

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Abstract

Gd substitution in DyB_2C_2, which undergoes an antiferroquadrupolar (AFQ) ordering at $T_Q = 24.7$ K and an antiferromagnetic (AFM) one at $T_N = 15.3$ K, enhances the AFM interaction and weakens the AFQ interaction, and the transition sequence at 20% Gd is the reverse of that in DyB_2C_2. The AFQ transition occurs at $T_Q = 17.5$ K, below the AFM one at $T_N = 19.0$ K in $Dy_{0.8}Gd_{0.2}B_2C_2$. The present work provides some experimental results on the specific heat and magnetization of $Dy_{0.8}Gd_{0.2}B_2C_2$. The magnetic phase diagrams of $Dy_{0.8}Gd_{0.2}B_2C_2$ have some resemblance to those of HoB_2C_2 and $Ce_{0.75}La_{0.25}B_6$. There is an anomalous magnetic phase (named phase IV) that is adjacent to the AFQ phase in each compound.

1. Introduction

DyB_2C_2 with the tetragonal LaB_2C_2-type structure [1] undergoes an antiferroquadrupolar (AFQ) ordering at $T_Q = 24.7$ K and, successively, an antiferromagnetic (AFM) one at $T_N = 15.3$ K [2]. The AFQ order in DyB_2C_2 is the first example found in tetragonal rare-earth compounds, and its transition temperature T_Q is about ten times higher than those for other AFQ materials found to date. Gd substitution in DyB_2C_2 decreases T_Q and increases T_N, and this results in a transition sequence in $Dy_{0.8}Gd_{0.2}B_2C_2$ that is the reverse of that in DyB_2C_2—that is, the AFQ transition occurs at $T_Q = 17.5$ K, below the AFM one at $T_N = 19.0$ K in $Dy_{0.8}Gd_{0.2}B_2C_2$ [3]. This transition sequence is very similar to that in an isostructural compound HoB_2C_2 which has AFQ ordering at $T_Q = 4.5$ K, below the AFM transition temperature at $T_N = 5.9$ K—that is, the new phase named phase IV appears between T_Q and T_N [4]. This transition sequence also resembles that in $Ce_{0.75}La_{0.25}B_6$ [5]. This type of phase, which is adjacent to an AFQ phase, attracts our interest because of its mysterious and controversial character. Here, it is noted that the AFM phase of TbB_2C_2 is also adjacent to an AFQ phase which is induced by magnetic fields applied along the c-plane, and thus this phase has also been named phase IV [6]. The present work provides the magnetic phase diagrams of $Dy_{0.8}Gd_{0.2}B_2C_2$ obtained by means of specific heat and magnetization measurements.

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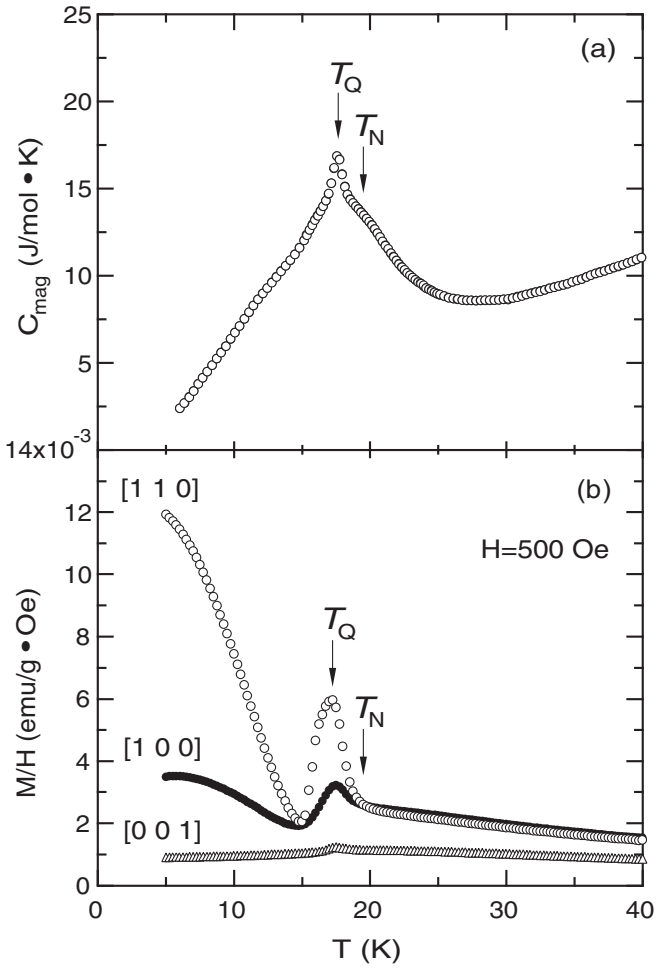

Figure 1. Temperature dependences of (a) the specific heat and (b) the magnetization of the single-crystalline $Dy_{0.8}Gd_{0.2}B_2C_2$ sample.

2. Experimental details

The sample preparation has been described elsewhere [2]. We used Dy and Gd of 99.9% purity, B of 99.8% purity, and C of 99.999% purity. The magnetizations were measured using a SQUID magnetometer for low magnetic fields below 5 T and a vibrating-sample magnetometer up to 14 T, and the specific heat was measured by a conventional relaxation method.

3. Results and discussion

Figure 1 shows the temperature dependences of the specific heat and magnetizations M/H of the single-crystalline $Dy_{0.8}Gd_{0.2}B_2C_2$ compound. A sharp anomaly at 17.5 K and a broadened peak around 19.0 K in the specific heats as shown in figure 1(a) are considered to correspond to the AFQ and AFM transition temperatures, respectively, because these temperatures are

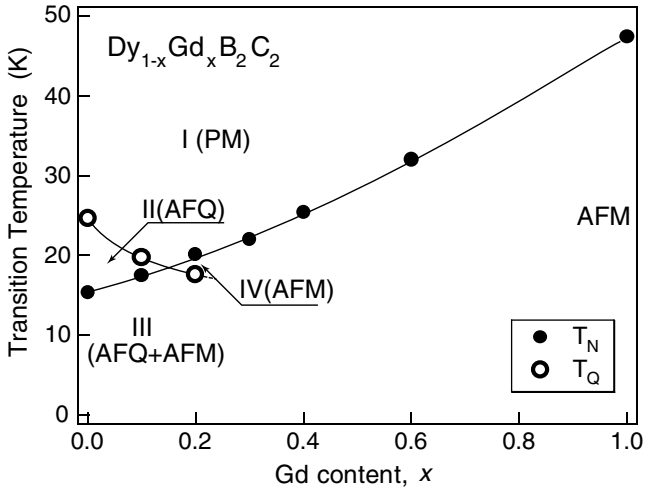

Figure 2. Concentration dependences of the AFQ (open circles) and AFM (closed circles) ordering temperatures, T_Q and T_N, of $Dy_{1-x}Gd_xB_2C_2$.

just on the extrapolated lines from the Gd concentrations lower than 20%. Figure 2 shows the concentration dependence of the AFM and AFQ transition temperatures of $Dy_{1-x}Gd_xB_2C_2$ which are determined by the specific heat [3]. As shown in the figure, the transition sequence of $Dy_{0.8}Gd_{0.2}B_2C_2$ is the reverse of that in DyB_2C_2 with $T_Q = 24.7$ K and $T_N = 15.3$ K [2]. Similar transition sequences occur in HoB_2C_2 [4] and $Ce_{0.75}La_{0.25}B_6$ [5]. It is noted that in HoB_2C_2 the specific heat anomaly at T_Q is very sharp and that at T_N is rather broad—that is, the behaviours of the specific heat are similar for $Dy_{0.8}Gd_{0.2}B_2C_2$ and HoB_2C_2.

The temperature dependence of the magnetization is peculiar in $Dy_{0.8}Gd_{0.2}B_2C_2$, as shown in figure 1(b), where the arrows indicate the transition temperatures determined by the specific heat. As the temperature decreases, the magnetization in the c-plane increases below T_N, decreases below T_Q, and increases again below 15 K. As is clear in figure 2, $Dy_{0.8}Gd_{0.2}B_2C_2$ is in a phase with coexistence of AFM and AFQ order (phase III) below T_Q which is similar to those below T_Q in HoB_2C_2 and below T_N in DyB_2C_2, and then spontaneous magnetizations are expected to appear [2, 4]. The spontaneous magnetization in phase III originates in the AFM structure forced on the non-collinear AFQ structure. The magnetization behaviour of $Dy_{0.8}Gd_{0.2}B_2C_2$ below T_Q is quite mysterious. These M–T curves in the c-plane seem to have some resemblance to the compensation behaviour of sublattice magnetizations in ferrimagnets where two different kinds of magnetic moment show different temperature dependences of the magnitudes of the moments—for example, in compounds that consist of rare earths and 3d transition metals. However, it is hard to apply this model simply to the present case, since the Gd moments randomly substitute for the Dy moments.

The magnetization below T_N in $Dy_{0.8}Gd_{0.2}B_2C_2$ increases as temperature decreases, although the magnetizations around T_N which is the higher-temperature boundary of phase IV show AFM-like cusps in both $Ce_{0.75}La_{0.25}B_6$ and HoB_2C_2. However, the magnetization increase below T_N has been observed in phase IV of $Ce_{0.75}La_{0.25}B_6$ under uniaxial pressures along [001] [7]. Furthermore, TbB_2C_2 shows a similar anomalous increase of the magnetic susceptibility below T_N [6, 8]. The largest increase appears along the [110] direction when a weak uniaxial pressure is applied along $[1\bar{1}0]$. Consequently, the anomalous increase of

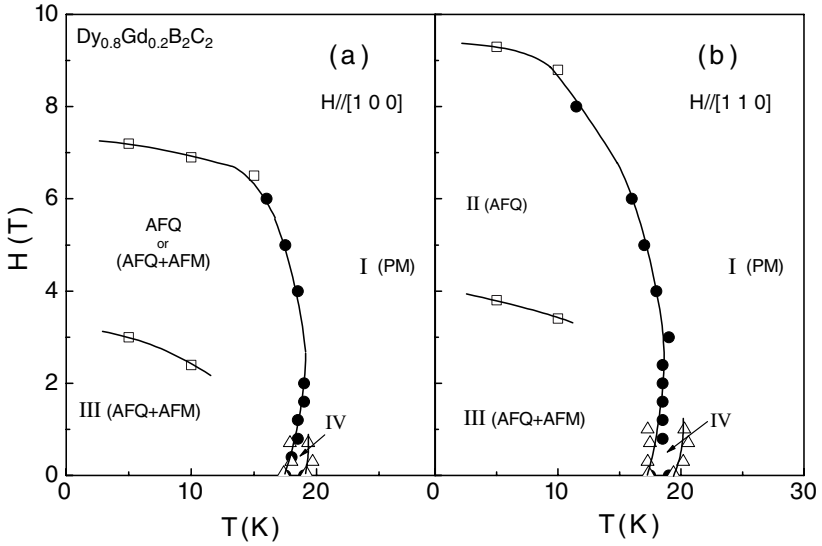

Figure 3. Magnetic H–T phase diagrams of $Dy_{0.8}Gd_{0.2}B_2C_2$ in the magnetic fields along (a) [100] and (b) [100] directions, where the open squares, open triangles, and closed circles are data from the M–H, M–H, and C_{mag} measurements, respectively.

M/H below T_N in $Dy_{0.8}Gd_{0.2}B_2C_2$ is similar to some extent to the behaviour in phase IV of $Ce_{0.75}La_{0.25}B_6$ and TbB_2C_2, although its physical origin remains unresolved.

As mentioned above, T_Q appears clearly in the specific heat. The transition at T_N is discernible as an inflection point in the magnetization, although it is obscure in the specific heat. Therefore, the magnetic H–T phase diagrams shown in figure 3 were constructed from three complementary sets of measurements—of the temperature dependence of the magnetization below 1.2 T, the magnetization process up to 14 T, and the specific heat below 8 T. Scaling with respect to magnetic field and temperature leads to the phase diagram of $Dy_{0.8}Gd_{0.2}B_2C_2$ for $H \parallel [110]$ shown in figure 3(b), which is similar to that for HoB_2C_2 [4], although the details around the regions where phase boundaries are close to each other and may cross are not clear from the present measurements. For $H \parallel [110]$, the extrapolated critical field at 0 K between phases II (AFQ) and I (paramagnetic, PM) decreases from ∼14.5 T for DyB_2C_2 to ∼9.5 T for $Dy_{0.8}Gd_{0.2}B_2C_2$, and the one between phases III (AFQ + AFM) and II increases from ∼3.5 T to ∼4.0 T. Linear relations of these critical fields with T_Q and T_N have been found for $Dy_{1-x}Y_xB_2C_2$ [9]. These changes are understandable, since the Gd substitution weakens the AFQ interaction and strengthens the AFM interaction.

The diagram for $H \parallel [100]$ shown in figure 3(a) is seemingly similar to that for $H \parallel [110]$. This similarity may suggest that the phase in the higher-field region is the AFQ phase (phase II). However, the higher critical field is ∼7.3 T which is close to the value ∼7.0 T for DyB_2C_2, where the phase in the higher-field region has been identified as another AFQ + AFM phase. It is noted that no pure AFQ phase (phase II) has been found in higher fields along the [100] direction in any of the compounds of TbB_2C_2, DyB_2C_2, and HoB_2C_2. At present, we cannot definitely identify the phase in the higher-field region for $H \parallel [100]$.

In efforts to observe more precise phase boundaries and to confirm and characterize each phase, experiments based on elastic constant measurement and the resonant x-ray scattering of $Dy_{0.8}Gd_{0.2}B_2C_2$ are now in progress.

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