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# ANTIFERROQUADRUPOLAR TRANSITION IN $Dy_{0.8}Gd_{0.2}B_2C_2$* **

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Temperature dependence of specific heat as well as magnetization reveals that $Dy_{0.8}Gd_{0.2}B_2C_2$ undergoes an antiferroquadrupolar transition at $T_Q = 17.5$ K below an antiferromagnetic one at $T_N = 20.0$ K. Gd substitution in $DyB_2C_2$ decreases $T_Q$ and increases $T_N$, and results in a transition sequence reverse to that in $DyB_2C_2$. The magnetization increases below $T_N$ and decreases below $T_Q$. The results are discussed as a manifestation of anomalous properties of the magnetic phase adjacent to the antiferroquadrupolar ordered phase.

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## 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 a successive antiferromagnetic (AFM) one at $T_N = 15.3$ K [2]. The AFQ order in $DyB_2C_2$ is the first example in tetragonal rare-earth compounds, and its transition temperature $T_Q$ is about ten times higher than those of other AFQ materials found to date. Furthermore, an iso-structural compound $HoB_2C_2$ has the AFQ ordering temperature at $T_Q = 4.5$ K below the AFM transition temperature $T_N = 5.9$ K, that is, the new phase named phase IV appears between $T_Q$ and $T_N$ [3]. This transition sequence resembles to that in $Ce_{0.75}La_{0.25}B_6$ [4]. This kind of phase adjacent to the AFQ phase attracts our interest because of its mysterious and controversial character. No evidence has been found for any magnetic order in phase IV of $Ce_{0.75}La_{0.25}B_6$, although antiferromagnetic reflections have been observed by neutron diffraction of the similar

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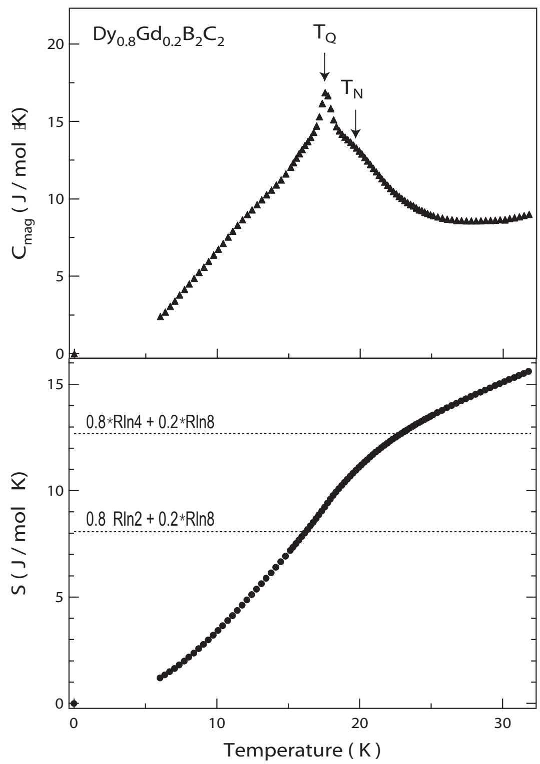


Fig. 1. Temperature dependence of the specific heat and entropy of $Dy_{0.8}Gd_{0.2}B_2C_2$.

phase in $HoB_2C_2$ [5, 6]. The present work provides experimental results of specific heat and magnetization of $Dy_{0.8}Gd_{0.2}B_2C_2$. The Gd substitution enhances the AFM interaction and weakens the AFQ interaction, and hence a transition sequence is expected to be the reverse of that in $DyB_2C_2$.

## 2. Experiments

Sample preparation was described elsewhere [2]. We used Dy and Gd of 99.9%, B of 99.8% and C of 99.999% in purity. The magnetizations were measured using a SQUID magnetometer and the specific heat was measured by a conventional relaxation method.

## 3. Results and discussion

Fig. 1 shows temperature dependence of the specific heat and entropy 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 20.0 K as shown by the arrows in the figure are considered to correspond the AFQ and AFM transition temperatures, respectively, because these temperatures are just on the extrapolated lines from the lower Gd concentrations than $x = 0.2$ [7]. This transition sequence is the reverse of that in $DyB_2C_2$ with $T_Q = 24.7$ K and $T_N = 15.3$ K [2]. The similar phenomenon has been observed in $Ce_{1-x}La_xB_6$ with $x = 0.25$ [4].

Fig. 2 shows temperature dependence of the magnetizations $M/H$ of $Dy_{0.8}Gd_{0.2}B_2C_2$ along three crystallographic axes. The arrows indicate the transition temperatures determined by the specific heat shown in Fig. 1. It is remarkable that, as the temperature decreases, the magnetization in the $c$-plane increases below $T_N$, decreases below $T_Q$ and increases again below 15 K.

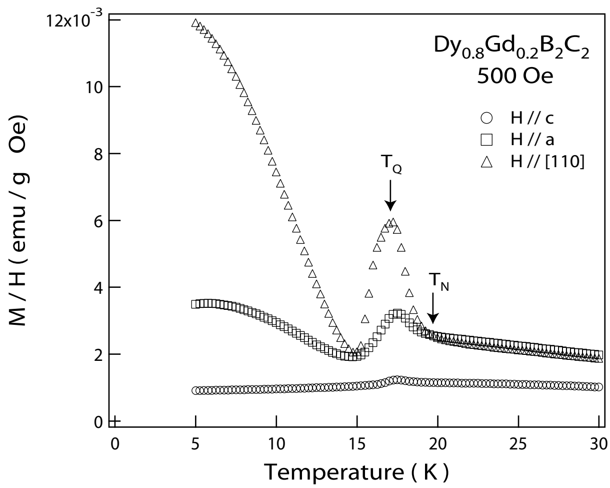


Fig. 2. Temperature dependence of the magnetizations of $Dy_{0.8}Gd_{0.2}B_2C_2$.

Below $T_Q$, $Dy_{0.8}Gd_{0.2}B_2C_2$ is in a coexistent phase of AFM and AFQ order which is similar to those in $HoB_2C_2$ below $T_Q$ and $DyB_2C_2$ below $T_N$, and then it is expected that spontaneous magnetizations appear [2, 3]. It is supposed that the Gd moments with $L = 0$ align antiferromagnetically so as to decrease the spontaneous magnetization. The anomalous change of magnetizations below $T_Q$ shown in Fig. 2 is attributable to different temperature dependence between the magnitudes of Dy and Gd moments, and a compensation temperature appears around 15 K.

The behavior that the magnetization increases below $T_N$ was also observed in phase IV of $Ce_{0.75}La_{0.25}B_6$ under uniaxial pressures along [0 0 1] [8]. Furthermore, $TbB_2C_2$ which is an antiferromagnet shows the similar anomalous increase of magnetization below $T_N$ [9, 10]. The largest increase appears along the [1 1 0] direction with a weak uniaxial pressure along $[1\,\bar{1}\,0]$ of $TbB_2C_2$ [11]. In addition, it is noted that the antiferromagnetic phase of

$TbB_2C_2$ is adjacent to an AFQ phase induced by magnetic fields [12]. The magnetizations in the $c$-plane of $HoB_2C_2$ show cusps at $T_N$ [2], that is, the magnetization of phase IV in $HoB_2C_2$ behaves in the similar way to that in $Ce_{0.75}La_{0.25}B_6$ under no uniaxial pressure. Consequently, the magnetic behavior of the phase between $T_Q$ and $T_N$ in $Dy_{0.8}Gd_{0.2}B_2C_2$ manifests some characteristics of the anomalous magnetic phase adjacent to the AFQ phase.

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