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Neutron scattering study on the antiferroquadrupolar order of DyB_2C_2 under magnetic fields

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Abstract

Antiferroquadrupolar (AFQ) ordering in DyB_2C_2 has been investigated by means of neutron scattering under applied magnetic fields along [110] up to 10 T. In the AFQ ordered phase at 16 K, antiferromagnetic reflections indexed as $00\frac{1}{2}$ and $11\frac{1}{2}$ are induced by a magnetic field of 1 T. These induced magnetic reflections disappear at 2 T, and develop again, increasing in intensity with increasing field, above 3 T. As temperature increases, the integrated intensity at 4 T shows an anomalous decrease toward ∼13 K in addition to a disappearance at T_Q, while that at 8 T decreases normally toward T_Q. We observed this curious behaviour of the induced magnetic reflections within the AFQ ordered phase.

1. Introduction

RB_2C_2 (R = rare earth) compounds, which exhibit various magnetic properties, have been attracting a great deal of interest, because the interactions between quadrupolar moments representing the asphericity of the 4f charge distribution play significant roles in the physical phenomena. In particular, DyB_2C_2 is the first compound with a tetragonal symmetry to show an antiferroquadrupolar (AFQ) ordering [1–3]. This compound undergoes successive transitions from a paramagnetic phase (I) to an AFQ ordered phase (II) at $T_Q = 24.7$ K, and further into a phase of coexistence (III) of the AFQ and antiferromagnetic (AFM) orders at $T_N = 15.3$ K. Below T_N, a complicated magnetic structure occurs, which is represented by four propagation vectors: $\boldsymbol{k}_1 = (1, 0, 0)$, $\boldsymbol{k}_2 = (1, 0, \frac{1}{2})$, $\boldsymbol{k}_3 = (0, 0, 0)$, $\boldsymbol{k}_4 = (0, 0, \frac{1}{2})$. This structure can be understood as resulting from the effects of competitive coexistence of AFM and AFQ interactions.

Neutrons can disclose the AFQ ordering via the observation of field-induced antiferromagnetic peaks, reflecting the existence of distinct easy axes in the two AFQ sublattices, although neutrons do not directly couple to quadrupolar moments. We have

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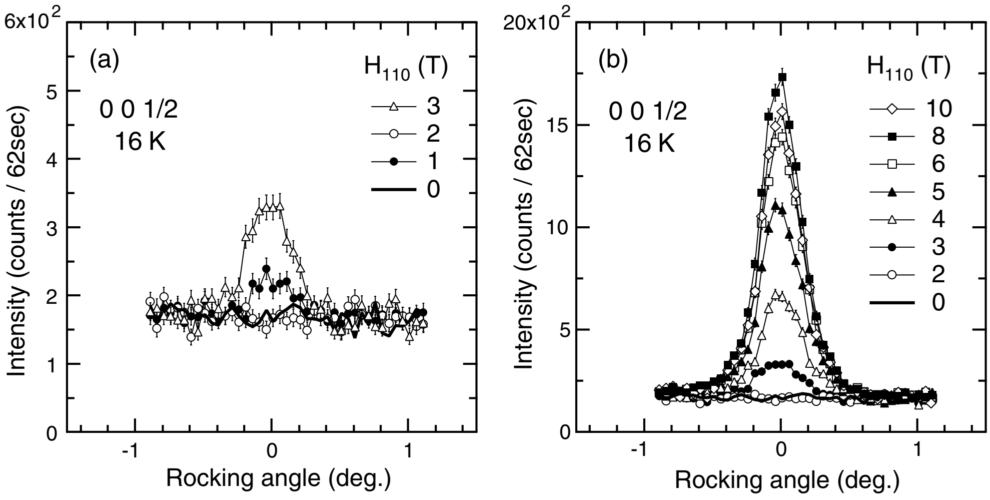

Figure 1. Rocking curves for the field-induced $00\frac{1}{2}$ AFM Bragg peak for (a) lower magnetic fields below 3 T and (b) fields up to 10 T applied along the $[1\bar{1}0]$ direction at 16 K.

indeed revealed AFQ order through neutron scattering measurements under magnetic fields for $\boldsymbol{H} \parallel [100]$ in the previous work [4]. The purpose of this paper, therefore, is to provide new information about the AFQ ordered phase in DyB_2C_2 by means of neutron scattering measurements under magnetic fields for $\boldsymbol{H} \parallel [110]$.

2. Experiments

The single crystals of $Dy^{11}B_2C_2$ were grown by the Czochralski method using a tetra-arc furnace. Since natural boron is a strong neutron absorber, B in the sample was replaced with enriched ^{11}B isotopes whose absorption cross-section is negligibly small. The sample used in the present experiment was a rectangle of dimensions $4 \times 4 \times 1$ mm^3.

Neutron scattering experiments under magnetic fields were performed on the triple-axis spectrometer TAS-2 installed at the thermal neutron guide of JRR-3M of the Japan Atomic Energy Research Institute (JAERI). The scattering measurements were made with an initial energy of 14.7 meV. Neutrons with this energy (wavelength: 2.359 Å) were obtained from the (002) reflection of the pyrolytic graphite (PG) monochromator. A guide–(monochromator)–80′–(sample)–40′–(analyser)–80′ arrangement was used for the horizontal collimation. A PG filter was placed before the sample to avoid $\lambda/2$ contamination. The scattering plane was chosen to be the [110]–[001] lattice plane in order to facilitate looking for the (hhl)-type reflections, since the propagation vector of the AFQ ordered structure which is consistent with the magnetic structure below T_N is expected to be $\boldsymbol{k} = (0, 0, \frac{1}{2})$ or $(1, 0, \frac{1}{2})$. Magnetic fields were applied vertically to the scattering plane, i.e. along the $[1\bar{1}0]$ direction, up to 10 T, using a He-free-type 10 T superconducting magnet developed by JAERI [5, 6].

3. Results

In the AFQ ordered phase at 16 K in zero field and the paramagnetic phase at 30 K for 8 T, only the nuclear reflections were recognized within the experimental accuracy. The application of

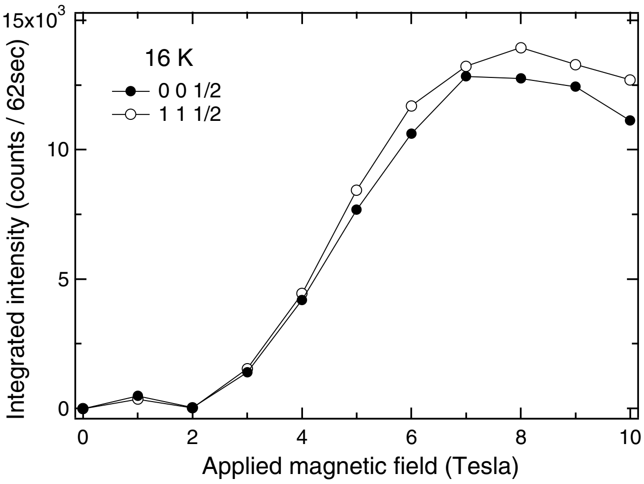

Figure 2. The applied magnetic field dependences of the integrated intensities of the induced AFM $00\frac{1}{2}$ and $11\frac{1}{2}$ Bragg peaks. The background components were subtracted from the data.

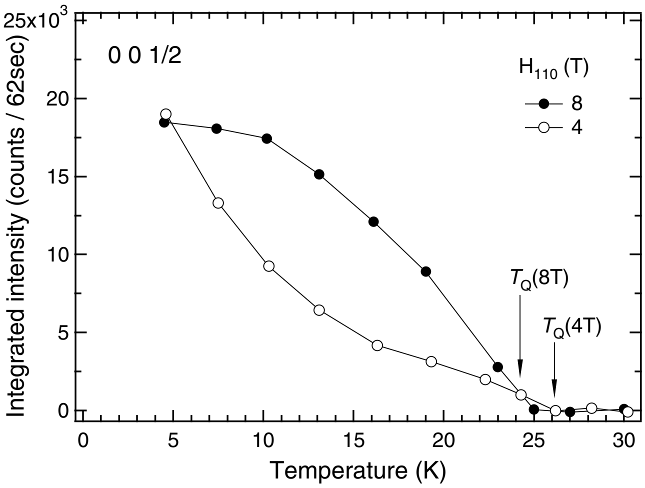

Figure 3. The temperature dependences of the integrated intensities of the induced AFM $00\frac{1}{2}$ Bragg peak at 4 and 8 T. The background components were subtracted from the data. The allows us to identify the transition temperatures determined from specific heat measurements [1].

magnetic fields up to 10 T causes the appearance of new $00\frac{1}{2}$ and $11\frac{1}{2}$ reflections in addition to the nuclear ones at 16 K. These peaks are attributed to AFM components of Dy moments induced by applied magnetic fields restricted by the underlying AFQ order. The rocking curves for $00\frac{1}{2}$ taken at 16 K for lower magnetic fields, below 3 T, and those from 0 up to 10 T are shown in figures 1(a) and (b), respectively. As can be seen from figure 1(a), the $00\frac{1}{2}$ reflection appears, has a maximum at 1 T, disappears at 2 T, and then increases in intensity above 3 T again. As shown in figure 1(b), the intensity of the induced AFM $00\frac{1}{2}$ Bragg peak grows rapidly with increasing field up to 8 T, and slightly decreases at 10 T. Figure 2 shows the applied magnetic field dependences of the integrated intensities of the induced AFM $00\frac{1}{2}$ and

$11\frac{1}{2}$ Bragg peaks. The background components were subtracted from the data. The $11\frac{1}{2}$ and $00\frac{1}{2}$ reflections show similar behaviour. The intensity of the $00\frac{1}{2}$ reflection becomes larger than that of the $11\frac{1}{2}$ reflection as the magnetic field increases. Indoh *et al* made it clear that new phase boundaries exist in the AFQ ordered phase along [110], by recent specific heat measurements [7]. It seems likely that the disappearance of the integrated intensities at 2 T results from crossings of these new phase boundaries. Figure 3 shows the temperature dependences of the integrated intensities of the induced AFM $00\frac{1}{2}$ Bragg peak at 4 and 8 T. The background components were subtracted from the data. The arrows indicate the AFQ transition temperatures T_Q determined by specific heat measurements [1]. As temperature increases, the integrated intensity at 4 T shows a rapid decrease toward $\sim$13 K in addition to a disappearance at T_Q, while that at 8 T decreases normally toward T_Q. This anomalous decrease in the low-temperature region at 4 T is also possibly due to the new phase boundary found by Indoh *et al* [7]. However, the present results are not complete enough to allow us to conclude that there is phase transition within the AFQ ordered phase, since there is insufficient information on the induced AFM reflections to allow us to distinguish between induced magnetic structures.

In summary, we confirmed that the AFM reflections are due to the field-induced magnetic moments on the AFQ structures for $\boldsymbol{H} \parallel [110]$. Although the curious behaviour of the integrated intensities was observed in the AFQ ordered phase, we need further investigation before we can understand these mechanisms.

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