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Anomalous magnetic scattering in tetragonal RB_2C_2 (R = rare earth) observed by means of neutron diffraction

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Abstract

Detailed neutron diffraction experiments have been performed on tetragonal $R^{11}B_2C_2$ (R = rare-earth) compounds which show characteristic antiferroquadrupolar orderings. TbB_2C_2, ErB_2C_2 and HoB_2C_2, which have magnetic long-periodic states, show anomalous magnetic diffuse scattering with flat intensity. The anomalous diffuse scattering exists in a small region in the reciprocal space, surrounded by satellite peaks, due to the long-periodic magnetic structures. The profile and temperature dependence of the anomalous diffuse scattering indicate that the origin cannot be understood on the basis of magnetic short-range correlations.

1. Introduction

RB_2C_2 (R: rare-earth) compounds, which have the tetragonal LaB_2C_2-type crystal structure [1–3], show diverse and anomalous magnetic and electric quadrupolar behaviours caused by competition between antiferromagnetic (AFM) and antiferroquadrupolar (AFQ) interactions [4, 5]. Of the RB_2C_2 system, TbB_2C_2 and HoB_2C_2 show particularly characteristic AFQ orderings. HoB_2C_2 exhibits an AFQ ordering at $T_Q = 4.5$ K under zero magnetic field, even though a magnetic ordered state exists between T_Q and $T_N = 5.9$ K ($>T_Q$) [5–7]. The AFQ ordering in HoB_2C_2 was also confirmed by resonant x-ray scattering experiments by Matsumura *et al* [8]. On the other hand, TbB_2C_2 shows an AFM ordering at $T_N = 21.7$ K, but no AFQ ordering under zero magnetic field has been found so far. However, Kaneko *et al* [9, 10] reported that there exist AFQ ordered states under magnetic fields in TbB_2C_2. Since the internal and external magnetic fields lift the degeneracy of the ground states, the AFQ orderings in TbB_2C_2 and HoB_2C_2 are unique phenomena.

We think that the magnetic long-periodic states which were found in HoB_2C_2 and TbB_2C_2 are important for investigation of the anomalies in RB_2C_2. The magnetic ordered state in TbB_2C_2 below T_N has a long-periodic magnetic component with the magnetic moments in

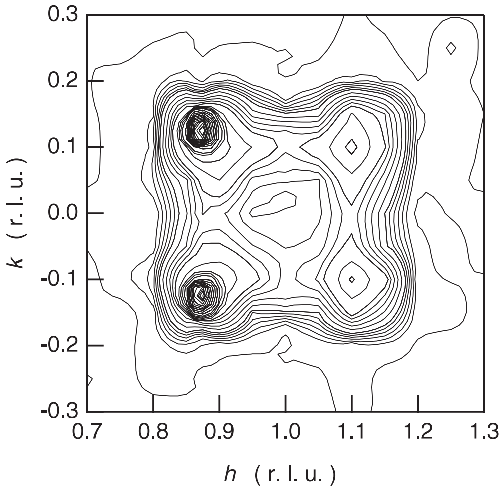

Figure 1. A contour map of the anomalous magnetic scattering at $T = 7$ K around the $(1, 0, 0)$ position in $Tb^{11}B_2C_2$ observed for KSD. Details have been reported in [11].

the c-plane as an appendant component of a very dominant $\boldsymbol{k} = (1, 0, 1/2)$ type of AFM structure with a small $\boldsymbol{k}' = (0, 0, 1/2)$ component; the propagation vector of the long-periodic component is $\boldsymbol{k}_L = (1 + \delta, \delta, 0)$, where $\delta = 0.13$ [9]. On the other hand, the long-periodic magnetic phase in HoB_2C_2 exists between T_N and T_Q [7, 12]. The propagation vector in the intermediate phase is $\boldsymbol{k}_L = (1 + \delta, \delta, \delta')$, where $\delta = 0.112$ and $\delta' = 0.04$ [7]. Note that ErB_2C_2, which shows no AFQ ordering, has a long-periodic magnetic state as well; the propagation vector in ErB_2C_2 is $\boldsymbol{k}_L = (1 + \delta, \delta, 0)$, where $\delta = 0.112$ [13]. The close similarity of the periodicities probably indicates that the long periodicity is based on some characteristics common to all members of the RB_2C_2 system. Thus, it is important to compare the long-periodic states with each other to understand characteristics of RB_2C_2.

The long-periodic magnetic structure phases in HoB_2C_2 and TbB_2C_2 are characterized by anomalous broad magnetic scattering with a trapezium-type profile as well as satellite peaks with long-periodic structures. Figure 1 shows the anomalous magnetic diffuse scattering observed in TbB_2C_2 at $T = 7$ K around the $(1, 0, 0)$ position [11]. The profile of the magnetic diffuse scattering cannot be understood in terms of magnetic short-range correlation or critical phenomena around T_N. Details of the anomalous scattering in TbB_2C_2 have been reported in [11]. The same trapezium-type diffuse scattering was also observed in the long-periodic state in HoB_2C_2 [12]. The anomalous magnetic scattering is thought to be an important feature in the RB_2C_2 system because of its similarity to those in HoB_2C_2 and TbB_2C_2; however, the origin of the anomalous magnetic scattering, especially the roles of AFQ interactions, is unknown as yet.

In ErB_2C_2 which also has a long-periodic magnetic structure phase between $T_N = 15.9$ K and $T_t = 13.0$ K, a similar broad magnetic scattering was observed in powder diffraction experiments [13]. Since ErB_2C_2 shows no AFQ ordering, comparing the broad magnetic scattering in ErB_2C_2 with those in HoB_2C_2 and TbB_2C_2 is important for understanding the possible role of AFQ interactions in the anomalous scattering. The authors, therefore, performed neutron diffraction experiments on a single-crystalline sample of $Er^{11}B_2C_2$ to clarify the diffuse scattering observed in the powder diffraction experiments.

For sample preparation, mixtures of 99.9% pure Er, 99.5%-enriched ^{11}B and 99.999% pure C were melted by the conventional argon arc technique; natural boron was replaced to

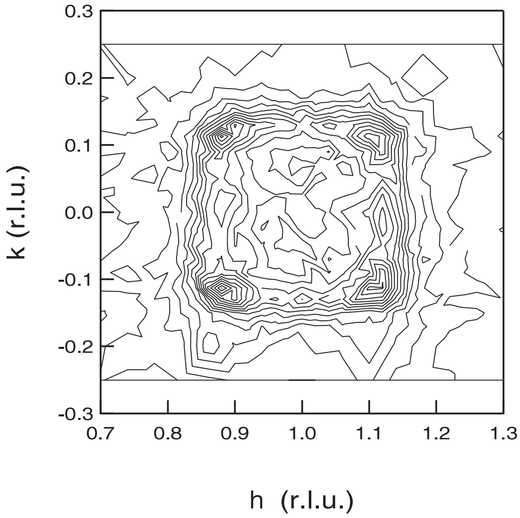

Figure 2. A contour map of the magnetic scattering at $T = 15.0$ K ($\sim T_N$) around the $(1, 0, 0)$ position in the $(h, k, 0)$ plane in $Er^{11}B_2C_2$ observed for KSD. There exists a square region surrounded by satellite positions, $(1+\delta, \pm\delta, 0)$ and $(1-\delta, \pm\delta, 0)$, with a nearly flat region around $(1, 0, 0)$. At this temperature, no satellite peak due to the long-periodic structure was detectable.

avoid its strong absorption effect. The single crystals were grown by the Czochralski method using a tri-arc furnace.

We performed neutron diffraction experiments on the Kinken neutron diffractometer KSD installed at the reactor JRR-3M in Japan Atomic Energy Research Institute, Tokai. A neutron beam with $\lambda = 1.52$ Å was obtained from the 3 1 1 reflection of the Ge monochromator. The collimation condition was 12′–open–sample–30′. The single-crystalline sample was mounted at the cold head of a closed-cycle He-gas refrigerator with the *c*-plane horizontal.

2. Results

In the experiments, anomalous broad magnetic scattering was also observed even in $Er^{11}B_2C_2$; the characteristics of the anomalous scattering are basically the same as those observed in the AFQ ordering compounds HoB_2C_2 and TbB_2C_2. Figure 2 shows a contour map of the intensity of the magnetic scattering in ErB_2C_2 around the $(1, 0, 0)$ position at $T = 15.0$ K; no magnetic satellite peak was observed at that temperature. As seen in figure 2, the profile of the magnetic diffuse scattering in ErB_2C_2 is nearly the same as that in TbB_2C_2 shown in figure 1 except for the existence of satellite peaks for TbB_2C_2 at $T = 7$ K; the nearly flat region was also observed in ErB_2C_2. The flat region indicates that the anomalous magnetic scattering shown in figure 2 cannot be understood in terms of magnetic short-range correlations either. With increasing temperature, anomalous diffuse scattering develops above 13 K $\sim T_t$, where satellite peaks also develop, and has its intensity maximum at about 15 K $\sim T_N$, where the satellite peaks disappear. Note that the anomalous diffuse scattering remains up to about 20 K.

The important features of the magnetic long-periodic states in RB_2C_2 (R = Tb, Ho and Er) which were found by our neutron diffraction experiments are as follows:

(1) there exists anomalous magnetic scattering in the square region around the $(1, 0, 0)$ reciprocal-lattice position with nearly flat intensity surrounded by the magnetic satellite peaks;

(2) diffuse-type magnetic scattering components were also observed around and between the satellite positions;
(3) these magnetic scattering components show complicated temperature dependence and persist up to $\sim 2T_N$ or $2T_Q$;
(4) with increasing temperature, the intensity of the anomalous magnetic scattering in ErB_2C_2 shows a maximum at T_N, where the satellite peaks due to the long-periodic structure disappear, while those for HoB_2C_2 and TbB_2C_2 show no obvious anomaly at the temperatures where satellite peaks due to long periodicity disappear.

Details of the characteristics of the anomalous diffuse scattering have been reported in [11, 12].

The fact that ErB_2C_2 shows the same type of anomalous magnetic scattering as HoB_2C_2 and TbB_2C_2 indicates that AFQ interactions are not necessarily required to understand the origin of the anomalous magnetic scattering. Moreover, the anomalous magnetic scattering is a characteristic common to all of the RB_2C_2 compounds with long-periodic magnetic structures. From this result and the closely similar periodicity, the authors conclude that characteristics found for the long-periodic magnetic states of HoB_2C_2, TbB_2C_2 and ErB_2C_2 are based on a property common to all members of the RB_2C_2 system, such as a structure of the Fermi surface and/or a common characteristic of the magnetic interactions in RB_2C_2.

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