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Neutron diffraction from $HoNi_2B_2C$

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Abstract

$RENi_2B_2C$ (RE = rare-earth) are quaternary compounds which exhibit a considerable degree of interaction between their superconducting and magnetic properties. The Ho variant is found to become superconducting at $T_c = 9$ K, but anomalous behaviour is observed in the low-field magnetic properties at $T < 7$ K (increasing susceptibility with decreasing temperature); at temperatures less than 5.6 K more conventional behaviour is regained. Neutron diffraction measurements demonstrate that at low temperatures ($T < 5$ K) the magnetic order consists of ferromagnetic planes antiferromagnetically coupled up the c-axis with a Ho moment of $8.9 \pm 0.2\ \mu_B$ canted at $15 \pm 5^\circ$ from the a–b plane. At high temperatures (5 K $< T <$ 8.5 K) satellites, together with the principal Bragg reflections, are observed which correspond to a modulation of the magnetic order with a wavelength of ~ 136 Å up the c-axis. In the intermediate temperature (5 K $< T <$ 7 K) range further Bragg reflections become evident. An additional modulation along the a-axis becomes important in this temperature range.

1. Introduction

Superconductivity in the RE–Ni–B–C system was first indicated by the investigations of phases present in samples of Y–Ni–B–C [1]. $RENi_2B_2C$ was identified as the appropriate compound for the superconducting material [2] and the structure was determined by X-ray diffraction using small single crystals [3]. The structure of these compounds consists of RE–C layers separated by Ni_2–B_2 layers. Although these materials resemble to some extent the layered structures of the cuprates, which exhibit high-temperature superconductivity, it is thought that superconductivity in these materials is due to a very strong electron–phonon interaction [4]. The $RENi_2B_2C$ compounds can be stabilised for a number of the heavy RE elements. For RE ions with magnetic moments there is considerable interaction between the tendency to superconductivity and magnetic ordering. In the study of the bulk properties for a range of $RENi_2B_2C$ compounds, as described in Ref. [5], the Ho compound was found to be reentrant in zero applied magnetic field at temperatures around 5 K where a pronounced dip in H_{c2} occurs. Fig. 1 presents our low field susceptibility measurements of a zero-field-cooled sample of $HoNi_2B_2C$ measured using a SQUID magnetometer. It should be noted that our samples of $HoNi_2B_2C$ have a higher T_c than those used in Ref. [5] and the dip structure is also shifted in temperature. We believe this is because we have better homogeneity and are nearer to the optimum carbon content. The anomalous superconducting behaviour in this material is directly linked to the nature of the magnetic ordering of the Ho moments. The neutron diffraction experiments described in this paper were performed to

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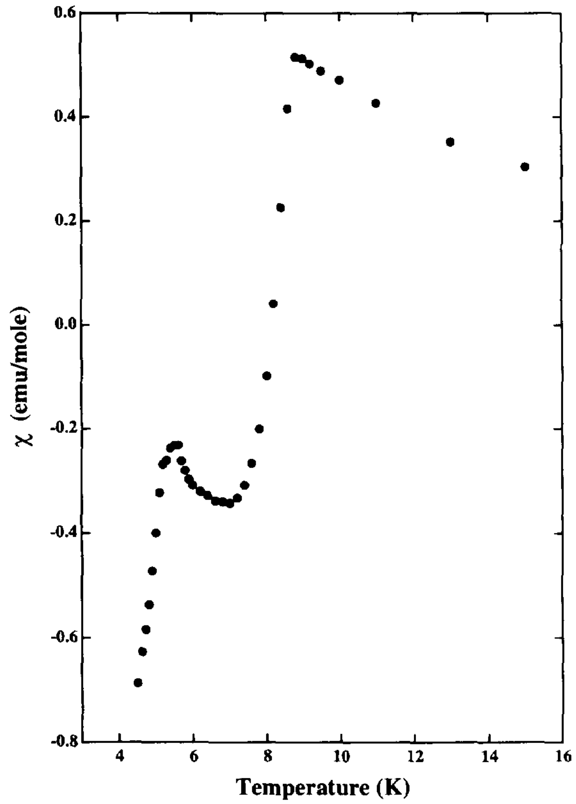

Fig. 1. Magnetic susceptibility of $HoNi_2B_2C$ cooled in zero field then measured with an applied magnetic field of 10 Oe.

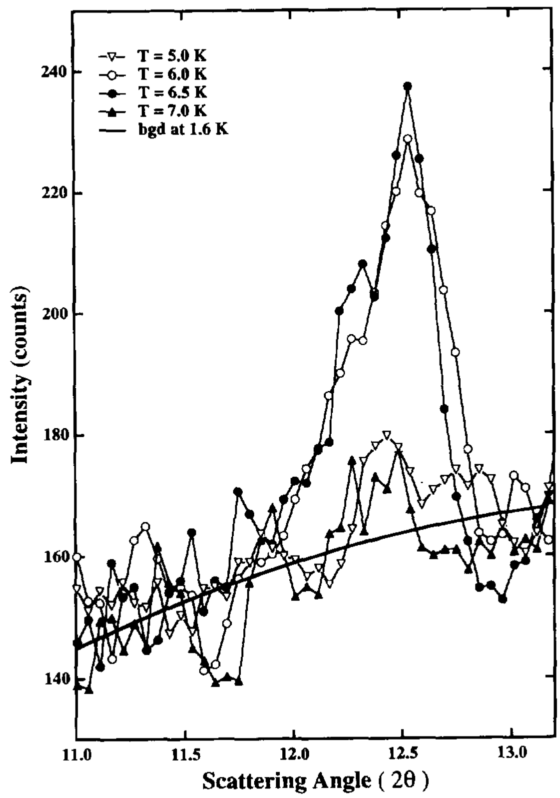

Fig. 2. One of the additional Bragg reflections, which correspond to a modulation along the *a*-axis, in the intermediate temperature range (5 K $< T <$ 7 K). This data was taken with a neutron wavelength of 1.47 Å.

investigate correlations between the anomalous superconducting properties and the magnetic ordering.

2. Experimental details and observations

The neutron diffraction experiments were performed using the TAS3 diffractometer at Risø National Laboratory and the HB4 instrument at Oak Ridge National Laboratory using incident wavelengths of 1.475 Å and 1.418 Å, respectively. The sample for these experiments was produced by arc-melting of the constituent elements followed by extensive annealing. ^{11}B was used to reduce the neutron absorption of the powdered sample. Background data were measured at 15 K and subtracted from the observations at lower temperatures to reveal the magnetic diffraction pattern.

At low temperatures ($T < 5.2$ K), the magnetic diffraction pattern can be indexed simply with the condition that $h + k + l$ is an odd integer. This implies that for this body-centred tetragonal material the magnetic cell is the same size as the chemical cell. A model with ferromagnetic planes antiferromagnetically coupled up the *c*-axis is capable of explaining the observed diffraction intensities, if the Ho moments are aligned at $75 \pm 5^\circ$ to the *c*-axis. A comparison of the intensity of the magnetic reflections to the nuclear reflections provides an estimate for the low-temperature magnitude of the Ho moment as $8.9 \pm 0.2\ \mu_B$. At high temperatures (5 K $< T <$ 8.5 K) the same set of principal Bragg reflections is observed but each reflection is accompanied by two satellites. A consideration of the positions of these satellites shows that this pattern corresponds to a modulation of the magnetic moment up the *c*-axis with a modulation wavelength of $\sim$ 136 Å, 13 unit cells in the *c*-direction. In the intermediate temperature range, where the anomalous magnetisation occurs, an additional set of Bragg reflections occurs. These peaks are only present if 5 K $< T <$ 7 K, as is demonstrated in Fig. 2. These reflections correspond to a modulation along the *a*-axis with a period of 2.4 unit cells for the Ho moments.

3. Conclusion and discussion

The low-temperature antiferromagnetic commensurate structure of the ordered Ho magnetic moments is capable of coexisting with the superconductivity. The magnitude of the ordered moment suggests that crystal-field effects are important in this compound. The interactions which lead to ferromagnetic planes of Ho ions are probably stronger than the antiferromagnetic coupling up the *c*-axis, since the susceptibility has ferromagnetic character at high temperatures [5], although crystal-field effects will also influence the magnitude and temperature dependence of the susceptibility.

It would appear that for this material the existence of a spiral modulation is adopted, to coexist with the superconducting state. In superconductors with a tendency to ferromagnetic order, e.g. $HoRh_4B_4$, a spiral state is often adopted in order to permit such coexistence. The high-temperature state probably has the Ho moment arranged on the surface of a cone with a cycloidal spiral in the *a–c* plane, this configuration permits Bragg reflections with the period of the spiral and the commensurate spacing.

In the region where the anomaly in magnetisation is observed, an additional modulation is observed. It is this modulation which appears to be the origin of the anomaly and the source of the pair breaking in this region of temperature.

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