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Neutron diffraction studies of $Ho_{1-x}Y_xNi_2B_2C$ compounds

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Abstract

Neutron diffraction measurements have been carried out to investigate the nature of magnetic ordering in $Ho_{1-x}Y_xNi_2B_2C$ ($x = 0, 0.1$ and 0.2) compounds. $HoNi_2B_2C$ shows a complex type of magnetic ordering below the superconducting transition, with a commensurate antiferromagnetic ordering of the Ho moments below 5 K and a modulation of the magnetic ordering along *a*- and/or *c*-direction between $5\text{ K} < T < 8.5\text{ K}$. The substitution of Y for Ho alters the commensurate nature of the Ho ordering. In $Ho_{0.9}Y_{0.1}Ni_2B_2C$, satellite peaks corresponding to *c*-modulation appear below T_N and exist down to 2 K, together with the principal Bragg peaks. However, when the Y concentration is increased to 20%, the principal Bragg peaks disappear and only the *c*-modulated satellite magnetic peaks are observed below T_N.

1. Introduction

The RNi_2B_2C superconductors with magnetic R ions (Tm, Er, Ho and Dy) [1–13] have been the subject of many recent studies due to their interesting magnetic properties in the superconducting state. They have a layered structure [14] which resembles many of the high temperature superconductors, with alternating layers of R–C and Ni_2–B_2. The compounds with R = Tm, Er, Ho and Dy are superconducting above 4 K [1, 11–12] and the nature of magnetic ordering in the superconducting state were established by susceptibility, specific heat and neutron diffraction measurements [1–13].

Of the RNi_2B_2C compounds, $HoNi_2B_2C$ (T_c of ~ 9 K) shows extremely interesting properties with anomalies around 5 K in the magnetisation measurements [2]. Specific heat measurements also reveal anomalous behaviour around this temperature [10]. Neutron diffraction measurements on powder samples [3, 6] as well as single crystals [4] confirm that these anomalies arise due to the unusual nature of the ordering of the Ho moments. Below 5 K, the compound shows commensurate AFM ordering of the Ho moments. Above this temperature ($5 < T < 8$ K), the commensurate nature changes over to a complex type of ordering with *a*- and/or *c*-modulations [3]. The existence of the *a*-modulation around 5–6.5 K is not observed in all the experiments and is thought to be dependent on the sample properties. However, the observed *a*-modulation coincides well with the temperature region where the anomalous behaviours occur.

We have investigated the nature of the modulations in this compound by neutron diffraction measurements in $Ho_{1-x}Y_xNi_2B_2C$ ($x = 0, 0.1$ and 0.2) compounds. By substituting Y for Ho, the magnetic ordering temperature

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of the Ho moments is expected to decrease and the T_c to increase. This in turn increases the interval between the T_c and T_N and is thus expected to change the nature of the magnetic modulations. Our results show that in $Ho_{0.9}Y_{0.1}Ni_2B_2C$, satellite peaks corresponding to c-modulation appears below 6 K and exist down to 2 K, below which only the commensurate magnetic ordering occurs. However, when the Y concentration is increased to 20%, the principal Bragg peaks is absent in the magnetic structure and only the c-modulated satellite peaks are observed at low temperatures.

2. Experiments

The samples were prepared by the standard arc-melting method using ^{11}B. The AC susceptibility was measured using a mutual inductance method. Neutron diffraction experiments on powdered samples were performed at Risoe National Laboratory and Oak Ridge National Laboratory.

3. Results, discussion and conclusion

Fig. 1 shows the magnetic susceptibility of $Ho_{1-x}Y_xNi_2B_2C$, $x = 0, 0.1, 0.2$, in an AC field of 3 Oe. As is evident from the figure, there is a slight increase in the T_c for the Y substituted samples. The peaks at 5 K for $x = 0$ and 3.5 K for $x = 0.1$ correspond to the anomalies associated with the magnetic ordering of Ho moments. The results show that the anomalous behaviour around the Ho ordering disappears for concentrations $x \geqslant 0.2$.

The diffraction data at 15 K, which is well above both T_c and T_N, was used as a reference to obtain the magnetic contributions by subtracting from the data obtained at lower temperatures. The 15 K data was also used to index all the nuclear peaks and the lattice parameters obtained from a fit to the peaks are listed in Table 1. For the $x = 0$ sample [3], the observed magnetic peaks at 1.6 K can be indexed on the basis of a body-centred unit cell for Ho, which has the same size as the chemical cell, with the antiferromagnetic (+ −) spin sequences at the positions of (0, 0, 0) and $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ with the indices, $h + k + l$, equal to odd numbers. A similar indexing can also be done for the $x = 0.1$ at 1.6 K. As the temperature increases above 2 K, satellite peaks appear for the $x = 0.1$ sample. Fig. 2 shows the (0 0 1) magnetic peak at $T = 3.5$ K, along with the satellite peaks. These satellite peaks correspond to a modulation of the magnetic order along the c-axis with a wave vector, $q_c = 0.09$ Å^{-1}. These peaks exists upto 6 K as shown in the inset of Fig. 2. The temperature at which the maximum of the intensity of the satellite peaks (3.5–4 K), agrees well with the AC susceptibility data where the anomaly occurs.

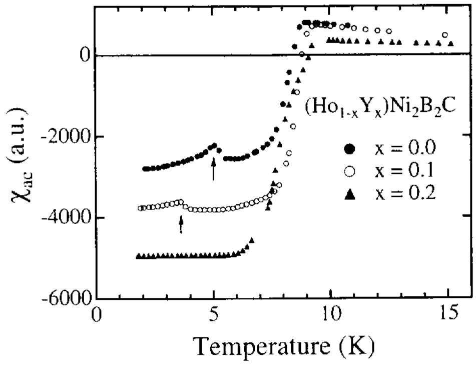

Fig. 1 AC susceptibility versus temperature for $Ho_{1-x}Y_xNi_2B_2C$ compounds, in applied AC filed of 3 Oe and a frequency of 113 Hz. The arrows indicate the re-entrant type behaviour.

Table 1
List of lattice parameters (a and c), magnetic moment of Ho at 1.6 K in the ordered state (μ), the orientation of the magnetic moment with respect to the c-axis (Φ) and the value of modulation vectors along the a-direction (q_a) and c-direction (q_c) for $Ho_{1-x}Y_xNi_2B_2C$ compounds (x = 0, 0.1, 0.2).

	$x = 0$	$x = 0.1$	$x = 0.2$
a (Å)	3.509 ± 0.001	3.521 ± 0.001	3.519 ± 0.001
c (Å)	10.528 ± 0.001	10.570 ± 0.003	10.548 ± 0.003
$\mu(\mu_B)$	8.9 ± 0.20	7.41 ± 0.08	3.21 ± 0.07
Φ	75 ± 5°	57 ± 1	90°
q_c (Å^{-1})	0.078 (136 Å)	0.09 (117 Å)	0.104 (101 Å)
q_a (Å^{-1})	0.414 (8.4 Å)	—	—

As the Y concentration is increased to $x = 0.2$, the principal magnetic Bragg peaks are absent down to the lowest temperatures (1.6 K) and only satellite peaks corresponding to a c-modulation appears. Fig. 3 shows a typical spectra corresponding to the magnetic peak (0 0 1). The satellite peaks can be indexed as $(h, k, l \pm q_c)$, where $h + k + l$ is odd and $q_c = 0.104$ Å^{-1}. From a comparison of the intensities of the magnetic peaks with the nuclear Bragg peaks, the magnetic moments and its alignments with the c-axis were estimated for all the compositions and are listed in Table 1.

In $HoNi_2B_2C$, the low temperature ($T < 5$ K) antiferromagnetic commensurate structure of the ordered Ho moments is able to coexist with superconductivity. However, at high temperatures ($5 < T < 8.5$ K) where the superconductivity also sets in, a correlation between the superconducting order parameter and the onset of

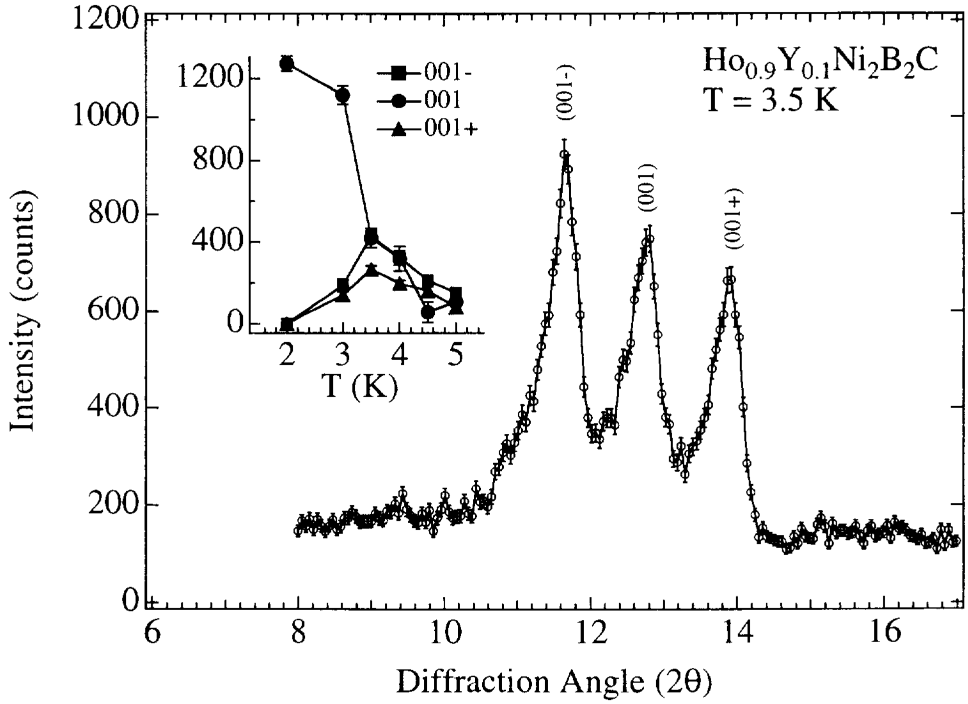

Fig. 2. The magnetic (0 0 1) peak with its satellites at 3.5 K for $Ho_{0.9}Y_{0.1}Ni_2B_2C$. The inset shows the variation of the integrated intensity of the (0 0 1) peak and its satellites with temperature.

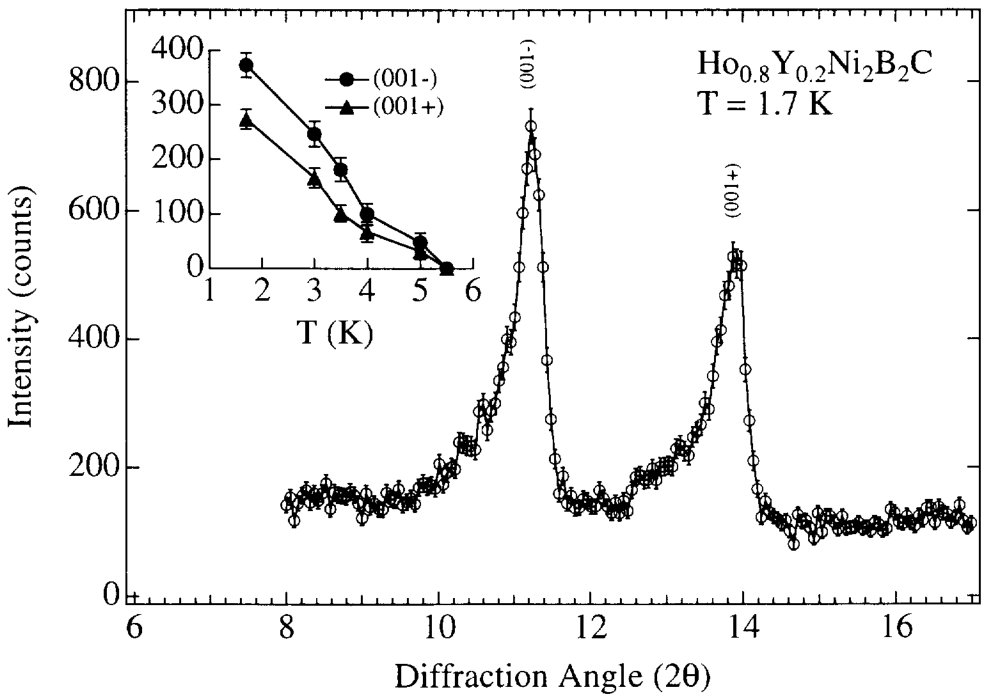

Fig. 3. The satellites of the magnetic (0 0 1) peak at 1.7 K for $Ho_{0.8}Y_{0.2}Ni_2B_2C$. Note the absence of the principal Bragg peak. The inset shows the variation of the integrated intensity of the satellites with temperature.

magnetic order may force the Ho moments to adapt a spiral configuration along the c-axis to coexist with superconductivity. This may be due to the build-up of ferromagnetic correlations in the a–b planes. When the non-magnetic Y is doped for the Ho, the correlations between the Ho moments are expected to be reduced. With $x = 0.1$, the interactions in the a–b interplanes are not strong enough to build a stable commensurate ordering down to 2 K. Increasing Y concentration to 20%, weakens the correlations of Ho moments in such a way that only the c-axis incommensurate peaks can be observed.

In conclusion, we have shown that substitution of Y for Ho in $HoNi_2B_2C$ alters the nature of Ho ordering. For $Ho_{0.9}Y_{0.1}Ni_2B_2C$, c-modulated peaks appear together with principal peaks down to 2 K, implying the nature of unstable magnetic ordering. The maximum peak intensity of the satellite peaks corresponds to the temperature at which anomalous behaviour in AC susceptibility occurs, as in the case of undoped $HoNi_2B_2C$. However, for $Ho_{0.8}Y_{0.2}Ni_2B_2C$, only c-modulated peaks without the principal Bragg peaks are be observed down to the lowest temperatures.

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