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## Magnetic order in HoNiBC and ErNiBC

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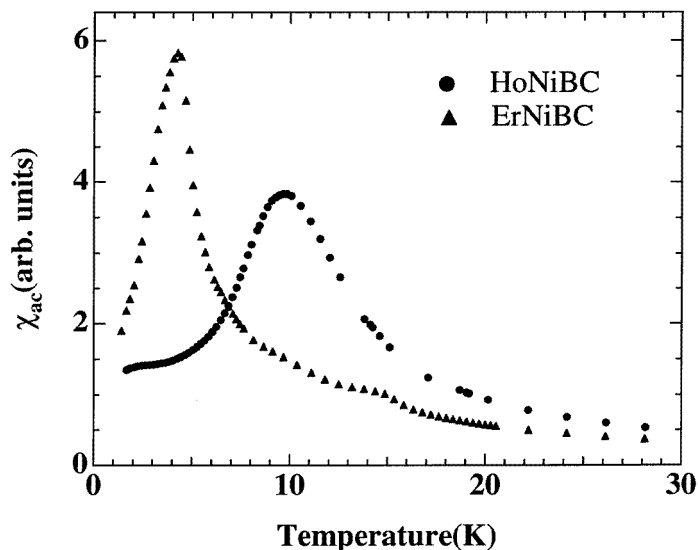
**Abstract.** Neutron diffraction measurements have been performed to study the nature of magnetic ordering in the compounds HoNiBC and ErNiBC. HoNiBC exhibits antiferromagnetic ordering of the Ho moments with a simple commensurate magnetic structure below  $T_N = 10$  K. The magnetic unit cell, doubled along the  $c$ -direction with respect to the chemical unit cell, consists of ferromagnetic planes antiferromagnetically coupled along the  $c$ -axis. On the other hand, the Er moments in ErNiBC show ferromagnetic ordering with a Curie temperature of 4.5 K. In contrast to the case for the structurally related superconducting counterparts (HoNi<sub>2</sub>B<sub>2</sub>C and ErNi<sub>2</sub>B<sub>2</sub>C) no modulated magnetic structures were observed for any temperatures above 1.4 K.

### 1. Introduction

The RNi<sub>2</sub>B<sub>2</sub>C (R = Y or rare earth) compounds have been studied very extensively [1–16] due to their interesting properties which include superconductivity with relatively high  $T_c$ s for magnetic as well as non-magnetic rare earths (16.5 K for Lu and 11 K for Tm), magnetic ordering of the R moments and the coexistence of magnetic ordering with superconductivity. In addition, they have a layered structure [17], resembling that of the high- $T_c$  copper oxide superconductors, with alternate layers of R–C and Ni<sub>2</sub>–B<sub>2</sub>. Even though RNi<sub>2</sub>B<sub>2</sub>C compounds form for most of the rare-earth ions, superconductivity is shown only by the compounds with R = Y, Dy, Ho, Er, Tm, and Lu [1, 5–7]. The gradual decrease in  $T_c$  from Tm (11 K) to Dy (6.5 K) is taken as an indication of the degree of interaction between the R magnetic moments and the conduction electrons. These compounds belong to a homologous series of compounds having the general formula (RC)<sub>*n*</sub>(NiB)<sub>*m*</sub> [18] where  $n$  and  $m$  are integers. The quaternary RNiBC ( $n = 2, m = 2$ ) compounds form with a structure similar to that of RNi<sub>2</sub>B<sub>2</sub>C but with an additional R–C layer in between the Ni<sub>2</sub>–B<sub>2</sub> layers, and are metallic but do not show superconductivity. The Bravais lattice of (RC)<sub>*n*</sub>(NiB)<sub>*m*</sub> changes from the body-centred tetragonal structure in RNi<sub>2</sub>B<sub>2</sub>C (space group  $I4/mmm$ ), to a simple tetragonal structure for RNiBC (space group  $P4/nmm$ ) [17, 18]. Band-structure calculations [19, 20] show that the density of states (DOS) at the Fermi level is less for LuNiBC than for LuNi<sub>2</sub>B<sub>2</sub>C compound. This reduced DOS is thought to be the primary reason for the absence of superconductivity in RNiBC compounds.

Even though the RNiBC compounds do not show superconductivity, they exhibit magnetic ordering of the R moments [21]. This provides an interesting comparison with the magnetic properties of the structurally related RNi<sub>2</sub>B<sub>2</sub>C compounds. Furthermore, since RNiBC may form as impurity phases in RNi<sub>2</sub>B<sub>2</sub>C compounds, it is of importance to have information about the magnetic nature of the RNiBC compounds in analysing

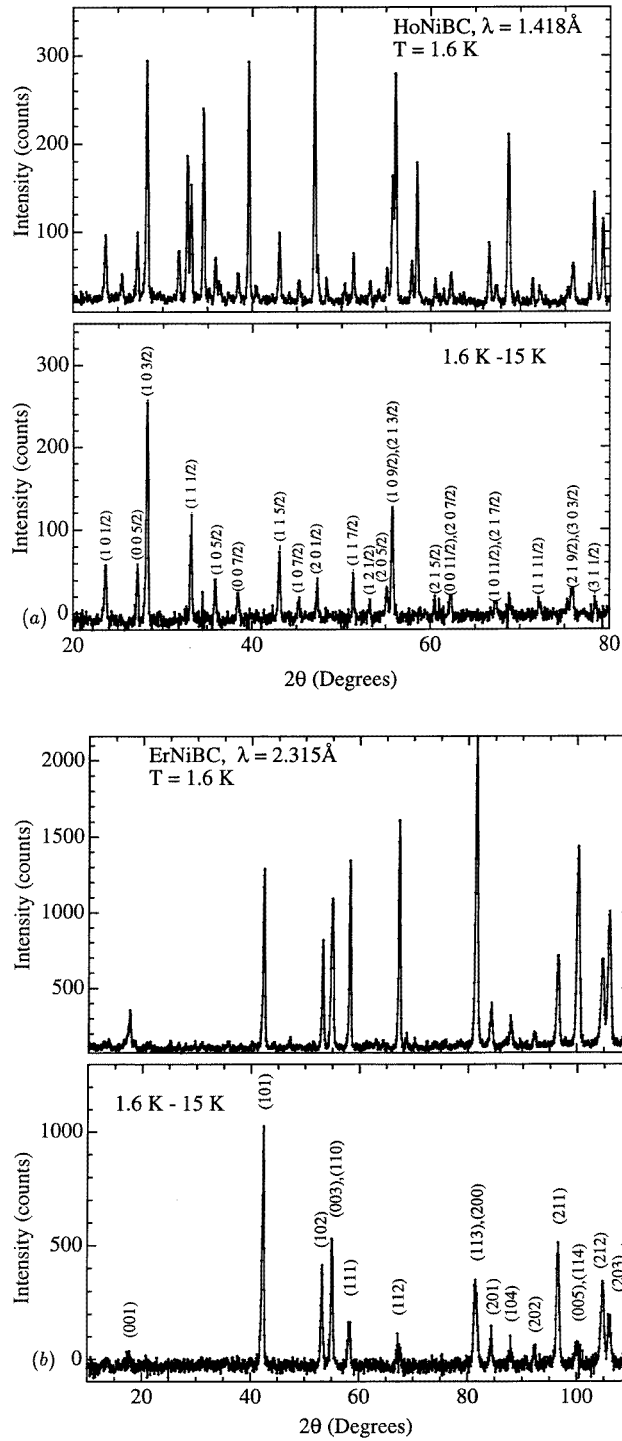
the magnetic properties of the  $\text{RNi}_2\text{B}_2\text{C}$  compounds. In this paper we present neutron diffraction measurements on  $\text{HoNiBC}$  and  $\text{ErNiBC}$  and compare with the corresponding 1221 compounds. Our results show that the  $\text{HoNiBC}$  compound exhibits antiferromagnetic ordering of the Ho moments with a simple commensurate magnetic structure below  $T_N = 10$  K. In contrast, the Er moments in  $\text{ErNiBC}$  show ferromagnetic ordering with a Curie temperature of 4.5 K. No modulated magnetic structures were observed at any temperature above 1.4 K in either of the compounds.



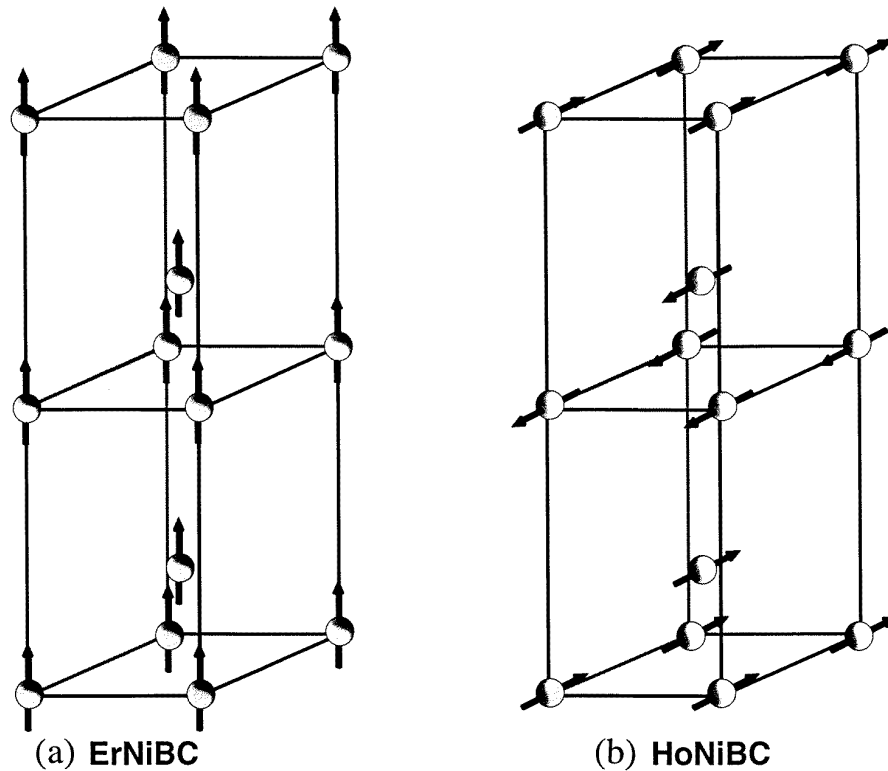
**Figure 1.** The temperature variation of the real part of the ac susceptibility for  $\text{HoNiBC}$  and  $\text{ErNiBC}$ . The peaks appearing at 10 K and 4.5 K correspond to magnetic ordering of the Ho and Er moments.

## 2. Experimental details

The samples were prepared by the standard arc-melting method and subsequent annealing. Stoichiometric amounts of Ho(99.9%), Er(99.9%), Ni(99.99%),  $^{11}\text{B}$ (99.5%) and C(99.999%) were melted several times under a flowing argon atmosphere on a water-cooled copper hearth, with the melted button turned over between meltings. The isotope  $^{11}\text{B}$  was used to prepare these samples to avoid the large absorption of neutrons by the naturally occurring  $^{10}\text{B}$ . The samples were then wrapped in tantalum foil, sealed in an evacuated quartz tube and annealed at 1000 °C for 16 hours. X-ray diffraction of the samples showed no peaks corresponding to any impurity phase. The ac susceptibility of the samples was measured by the standard mutual inductance method down to 1.4 K. Neutron diffraction experiments on powdered samples in the temperature range 1.4–15 K were performed using the TAS3 multidetector powder neutron diffractometer at Risø National Laboratory and the HB4 instrument at Oak Ridge National Laboratory using incident wavelengths of 2.315 Å and 1.418 Å, respectively. Diffraction from a powdered sample of  $\text{Al}_2\text{O}_3$  at room temperature was used to calibrate the wavelength and to provide a zero correction for the diffraction angle.



**Figure 2.** The magnetic diffraction Bragg peaks at 1.6 K for (a) HoNiBC ( $\lambda = 1.418 \text{ \AA}$ ) and (b) ErNiBC ( $\lambda = 2.315 \text{ \AA}$ ).



**Figure 3.** The magnetic structures of the ordered rare-earth moments: (a) ErNiBC—ferromagnetic; and (b) HoNiBC—antiferromagnetic.

### 3. Results, discussion and conclusions

Figure 1 shows the ac susceptibility of HoNiBC and ErNiBC. The peaks appearing at 10 K and 4.5 K correspond to magnetic ordering of the Ho and Er moments in HoNiBC and ErNiBC, respectively. Neutron diffraction data at 15 K, well above the magnetic ordering in HoNiBC and ErNiBC, were used as a reference to index all of the nuclear peaks. The lattice parameters obtained by indexing the nuclear peaks are  $a = 3.555 \pm 0.006 \text{ \AA}$ ,  $c = 7.537 \pm 0.010 \text{ \AA}$  for HoNiBC and  $a = 3.542 \pm 0.001 \text{ \AA}$ ,  $c = 7.547 \pm 0.004 \text{ \AA}$  for ErNiBC. In order to obtain the magnetic contributions, the 15 K data were subtracted from the data obtained at lower temperatures. Below the ordering temperature ( $T_N = 10 \text{ K}$ ), the magnetic peaks for HoNiBC (figure 2(a)) can be indexed if the chemical unit cell is doubled along the  $c$ -axis, as illustrated in figure 3(b). The magnetic structure consists of ferromagnetic planes of Ho moments coupled antiferromagnetically along the  $c$ -direction. However, the magnetic Bragg peaks of ErNiBC were observed at the same positions as the nuclear Bragg peaks, corresponding to a ferromagnetic ordering of the Er moments (figure 3(a)). From a comparison of the intensities for the magnetic (using the form factor for free ions [22]) and nuclear peaks, the Ho and Er magnetic moments at 1.6 K were estimated to be  $7.7 \pm 0.1 \mu_B$  and  $6.6 \pm 0.1 \mu_B$  and with angles,  $\Phi = 73^\circ \pm 4$  and  $\Phi = 0^\circ$  between the direction of the magnetic moment and the  $c$ -axis for HoNiBC and ErNiBC, respectively. The observed magnetic moments are smaller than the free-ion moments

**Table 1.** Comparison of the experimental ( $I_{\text{obsv}}$ ) and calculated ( $I_{\text{calc}}$ ) intensities of the magnetic Bragg peaks for HoNiBC for the neutron wavelength,  $\lambda = 1.418 \text{ \AA}$ . The intensities are normalized to 1000 with respect to the maximum observed intensity. ( $\chi^2 = 2.139$ .)

$2\theta$	( $hkl$ )	$I_{\text{obsv}}$	$\Delta I_{\text{obsv}}$	$I_{\text{calc}}$
23.62	(1 0 1/2)	286	21	300
27.21	(0 0 5/2)	239	20	256
28.25	(1 0 3/2)	1000	29	994
33.21	(1 1 1/2)	434	23	427
35.88	(1 0 5/2)	169	17	200
38.45	(0 0 7/2)	134	19	63
43.04	(1 1 5/2)	296	20	249
45.29	(1 0 7/2)	91	19	63
47.36	(2 0 1/2)	130	19	169
51.34	(1 1 7/2)	169	17	203
53.23	(1 2 1/2)	76	17	76
55.12	(2 0 5/2)	160	19	116
55.73	(1 0 9/2)	541	26	506
	(2 1 3/2)			
60.49	(2 1 5/2)	50	13	79
62.31	(0 0 11/2)	119	17	131
	(2 1 7/2)			
67.26	(1 0 11/2)	96	21	50
72.09	(1 1 11/2)	80	17	59
75.9	(2 1 9/2)	214	20	200
	(3 0 3/2)			
78.35	(3 1 1/2)	69	26	76
81.44	(2 0 11/2)	64	16	81
84.59	(1 1 13/2)	117	17	114
	(3 1 5/2)			
94.2	(1 0 15/2)	156	39	139
	(3 0 9/2)			
	(3 2 3/2)			

(Ho =  $10.4 \mu_B$ , Er =  $9.4 \mu_B$ ); this is considered to be due to strong crystalline-electric-field effects in these compounds. A comparison of the calculated and fitted intensities for the magnetic peaks in the two compounds is given in tables 1 and 2. In our fitting, no observable magnetic moments could be associated with the Ni sites within the resolution of our experimental data in either of these compounds.

RNiBC compounds are structurally related to the RNi<sub>2</sub>B<sub>2</sub>C compounds except for as regards an additional R–C layer in between the conducting Ni<sub>2</sub>–B<sub>2</sub> layers. However, in addition to the absence of superconductivity in RNiBC, the magnetic behaviour is also different. HoNi<sub>2</sub>B<sub>2</sub>C [8–11] shows AFM ordering with a commensurate structure only at low temperatures ( $T < 5 \text{ K}$ ), while a modulated, incommensurate structure is observed between 5 K and 8 K [8–10]. For HoNiBC, no modulation in the magnetic structure is observed down to 1.6 K. The magnetic structure is commensurate and the unit cell is doubled along the  $c$ -axis. In the case of ErNi<sub>2</sub>B<sub>2</sub>C [12, 13], a modulation along the  $a$ -direction is present below the AFM ordering temperature down to the lowest temperatures. However, in ErNiBC the dominant magnetic interactions are different and the compound undergoes a ferromagnetic ordering. In all of these compounds, the planes undergo ferromagnetic ordering of the rare-earth moments. It is the subtle changes in the interactions along the  $c$ -direction which give rise to different magnetic behaviour indicating the dominance of the

**Table 2.** Comparison of the experimental ( $I_{\text{obsv}}$ ) and calculated ( $I_{\text{calc}}$ ) intensities of the magnetic Bragg peaks for ErNiBC for the neutron wavelength,  $\lambda = 2.315 \text{ \AA}$ . The intensities are normalized to 1000 with respect to the maximum observed intensity. ( $\chi^2 = 0.819$ .)

$2\theta$	( $hkl$ )	$I_{\text{obsv}}$	$\Delta I_{\text{obsv}}$	$I_{\text{calc}}$
17.63	(001)	5	36	2
42.32	(101)	1000	34	1019
53.22	(102)	449	35	439
55.02	(003)	479	33	513
	(110)			
58.25	(111)	212	33	212
67.34	(112)	178	48	123
81.56	(113)	579	51	543
	(202)			
84.27	(201)	149	38	124
88.02	(104)	82	45	81
92.52	(202)	110	29	92
96.58	(211)	680	45	631
100.29	(005)	176	64	41
	(114)			
104.73	(212)	524	54	531
106.06	(203)	271	44	274

interplane over the intraplane interactions. The modulation of the magnetic ordering in the (Ho,Er)Ni<sub>2</sub>B<sub>2</sub>C compounds will be determined by the nesting effects on the Fermi surface, giving rise to a marked peak in  $\chi(\mathbf{q})$  at the modulation wave vector, and the type and magnitude of the inter- and intraplane interactions [23]. Both HoNi<sub>2</sub>B<sub>2</sub>C and ErNi<sub>2</sub>B<sub>2</sub>C exhibit an  $a$ -modulated structure, although for the Ho compound this only exists over a limited range of temperature ( $5 < T < 6.5$  K). HoNi<sub>2</sub>B<sub>2</sub>C also has a  $c$ -modulation in the magnetic structure for a large temperature range ( $5 < T < 8.5$  K). For HoNiBC and ErNiBC, the nesting effects have disappeared along with the reduction of the DOS at the Fermi level and the magnetic order shows no incommensurate features. The different ordering schemes are related to the differences in the interplane interactions in these compounds.

In conclusion, we have established the nature of magnetic ordering in HoNiBC and ErNiBC compounds from neutron diffraction measurements. Even though they are structurally related to the respective RNi<sub>2</sub>B<sub>2</sub>C compounds, the magnetic interactions in these compounds are different. HoNiBC exhibits antiferromagnetic ordering of the Ho moments with a simple commensurate magnetic structure below  $T_N$ . The Er moments in ErNiBC undergo a ferromagnetic transition with a Curie temperature of 4.5 K. No modulated magnetic structures were observed at any temperatures above 1.4 K for either of the compounds. Even though the absence of superconductivity in RNiBC can simply be related to the reduction in the density of states at the Fermi level, the difference in magnetic interactions between the RNiBC and RNi<sub>2</sub>B<sub>2</sub>C compounds can be assumed to arise from the difference in the interplane interactions and the changes in the band structure of these compounds.

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