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Magnetic structure of $ErNi_2B_2C$

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Neutron-diffraction techniques have been used to study the magnetic structure of the superconductor $ErNi_2B_2C$ ($T_c = 11$ K). The experimental results, obtained on *single* crystals, show that below approximately 7 K this compound is in an incommensurate modulated antiferromagnetic state, with wave vector $0.553\mathbf{a}^*$, that coexists with superconductivity. The field dependence of the magnetic structure of this compound was studied at 5.5 K as a function of an externally applied magnetic field both along the b and c axes of the tetragonal structure.

I. INTRODUCTION

The study of the physical properties of the recently discovered[1–4] superconducting rare-earth nickel boride-carbides, RNi_2B_2C (where R stands for a rare-earth element), may provide a better understanding of the competition between superconductivity and magnetism. The layered structure of these compounds is reminiscent of that of the high-T_c oxide superconductors and consists of RC layers separated by Ni_2B_2 sheets. Superconductivity is observed[2–5] in these compounds not only for the nonmagnetic rare-earth elements but also for the magnetic rare-earth elements (Tm,Er,Ho). Electronic band-structure calculations[6–8] for these compounds suggest that these materials are conventional superconductors.

Of particular interest for the heavy rare-earth nickel boride carbides are the resistive and upper critical field anomalies observed in measurements on powder samples by Eisaki *et al.*,[5] and single-crystal measurements by Canfield *et al.*[9] In a small temperature interval around 5 K the Ho compound was found to be reentrant under zero[5] or a finite but very small (20 G) (Refs. 9 and 10) externally applied magnetic field. Similar, but less pronounced, resistance anomalies are observed below 2.5 K and around 5.5 K for the Tm and Er compounds, respectively, but only under a modest magnetic field.

In a previous study[11] we reported the results of neutron-diffraction measurements on single crystals of $HoNi_2B_2C$. The experimental results[11] show that below approximately 4.7 K the Ho atoms are ordered in a simple antiferromagnetic structure: the moments are aligned ferromagnetically in each layer, with the magnetic moments of two consecutive layers aligned in opposite directions. The most interesting result of that study was that in the 4.7–6 K temperature range a transient modulated structure, characterized by two incommensurate wave vectors $\mathbf{k} = 0.585\mathbf{a}^*$ and $\mathbf{k}' = 0.915\mathbf{c}^*$, was observed. This suggests[11] that the pair breaking associated with the transient magnetic structure may be responsible for the deep minimum in H_{c2} and the almost reentrant behavior observed in this compound at approximately 5 K.

To obtain more information about the possible relationship between the magnetic structure and the H_{c2} and resistive anomalies in these compounds we performed a systematic neutron-diffraction study of the magnetic structure of $ErNi_2B_2C$. In this paper we report the results of these experiments.

II. EXPERIMENTAL DETAILS

Single crystals of $ErNi_2B_2C$ were grown at the Ames Laboratory by the high-temperature flux growth technique,[9] using boron depleted in the heavily neutron absorbing B^{10} nuclei. Single-crystal platelets extracted from the flux were examined by x rays and neutrons and were found to be single crystals of high quality with the c axis perpendicular to their flat surface. Magnetization measurements as a function of temperature and magnetic field were performed in a Quantum Design superconducting quantum interference device magnetometer on single crystals from the same batch as those used in the neutron-diffraction experiments.

The neutron-diffraction experiments were performed using the triple-axis spectrometer HB1A at the HFIR reactor of the Oak Ridge National Laboratory. Pyrolytic graphite reflecting from the (002) planes was used as monochromator and analyzer, and a pyrolytic graphite filter was used to minimize the $\lambda/2$ contamination of the incident beam. All measurements were performed with a constant incident neutron energy of 14.7 meV. The crystal used in the present experiment was of high quality with a mosaic spread characterized by a full width at half maximum (FWHM) of 0.09° measured by making use of a perfect germanium crystal as analyzer. The measured mosaic spread was found to increase by almost a factor of 2 between 7 and 1.7 K because of magnetostriction. Measurements over the 1.7–300 K temperature range were taken for two different crystal orientations, namely, with the scattering plane coincident with the a-b or a-c planes. The field dependence of the magnetic structure at 5.5 K was studied up to a field of 14 kG applied either along the tetragonal c axis or the basal plane b axis.

III. RESULTS AND DISCUSSION

In the absence of an applied field, between 300 K and approximately 7 K only nuclear reflections (hkl) with $h+k+l=2n$ are observed. As the temperature is decreased below approximately 7 K additional diffraction peaks start to develop in rows parallel to the reciprocal a axis (or the equivalent b axis of the tetragonal structure). The observed addi-

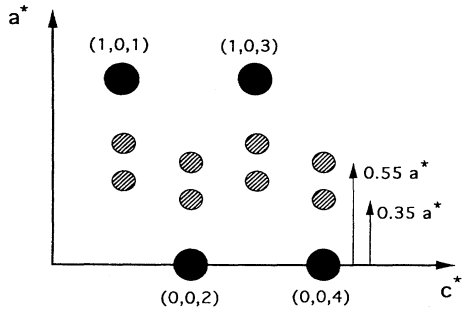

FIG. 1. Reciprocal space plane showing the points associated with nuclear scattering (filled circles), and the first- and third-order satellites (shaded circles) associated with the modulation wave vector $\mathbf{k}=0.553a^*$.

tional diffraction peaks are pairs of first- and higher-order satellites to each allowed nuclear reflection (hkl) with $h+k+l=2n$, with a fundamental incommensurate wave vector of (0.553, 0, 0). Figure 1 shows the first- and third-order satellites observed in the $(h0l)$ scattering plane. Fifth-, seventh-, and ninth-order satellites were also observed. The intensity of the observed satellites increases with decreasing temperature and approaches saturation below 3 K (Fig. 2). No other magnetic phase transition was observed down to approximately 1.7 K, the lowest temperature reached in the present experiment.

The above experimental observations imply that between 300 K and approximately 7 K the Er moments are not ordered, since in this temperature range, only nuclear and critical magnetic scattering is observed. Between approximately 7 K and 1.7 K, the lowest temperature reached in the present experiments, ordering of the moments is observed. In this temperature range the Er^{3+} moments are modulated along the a axis (or the equivalent b axis) with a wave vector $\mathbf{k}=0.553\mathbf{a}^*$, nearly doubling the chemical unit cell in the basal plane. The observations of satellites to all nuclear reflections, including (000), in both $\mathbf{a}^*$ and $\mathbf{b}^*$ directions shows that the structure is a *transverse* spin wave with a propagation vector along a^* (or b^*). Furthermore, the observation of higher-order harmonics shows that the modulation is not purely sinusoidal, but squared. The nuclear and magnetic structure factors obtained from the observed intensities are subject to large uncertainties due to secondary extinction effects. The extinction problem is particularly severe in the present experiments because of the high perfection of the crystals (FWHM$\simeq$0.1°) and the fact that as a result of magnetostriction, the mosaic of the crystals changes substantially as the temperature is reduced below the magnetic ordering temperature (see Sec. II). Nevertheless, given these large uncertainties, the moment obtained ($\simeq 8\mu_B$) by assuming that no moment is induced on the nickel atoms is in reasonable agreement with the results obtained by Sinha *et al.*,[12] on powder samples.

The magnetic ordering in $ErNi_2B_2C$ is similar to one of the transient modulated structures observed in $HoNi_2B_2C$ in the temperature region between 4.7 and 6 K. The magnetic wave vector along the a^* (b^*) in the Er compound (0.553) is only slightly smaller than that observed (0.585) for the Ho compound. In $HoNi_2B_2C$, however, additional satellites are observed corresponding to a squared modulation of the magnetic moments along the c axis, and no squaring of the magnetic structure along the a axis was observed in zero applied field. Since this modulated structure characterized by $\mathbf{k}=0.553\mathbf{a}^*$ coexists with superconductivity in $ErNi_2B_2C$, it is tempting to attribute the almost reentrant behavior of $HoNi_2B_2C$ at 5 K to the magnetic modulated structure char-

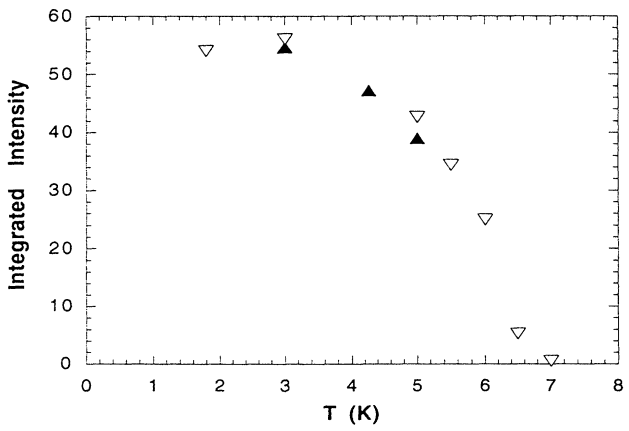

FIG. 2. Temperature dependence of the (1.447, 0, 0) satellite for increasing (filled triangles) and decreasing (open triangles) temperature.

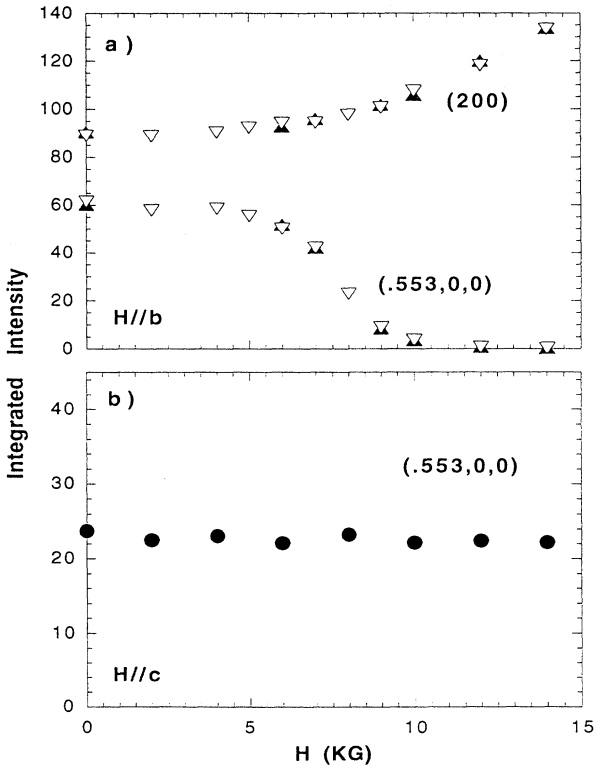

FIG. 3. (a) Field dependence at 5.5 K of the (0.553, 0, 0) satellite and (200) nuclear reflection for increasing (filled triangles) and decreasing (open triangles) fields along the b axis. (b) Field dependence of the (0.553, 0, 0) satellite reflection at 5.5 K for increasing field along the c axis.

acterized by $\mathbf{k}' = 0.915\mathbf{c}^*$ observed in this compound.

The magnetic structure of $ErNi_2B_2C$ has been also studied at 5.5 K as a function of a field applied either along the b or c axis. The results obtained with the crystal oriented in the $(h0l)$ plane and the field applied parallel to the b axis are summarized in Fig. 3(a). It is seen that the intensity of the satellites decreases with increasing magnetic field and becomes negligible at approximately 10 kG. In addition, an increase in the intensity of the nuclear Bragg peaks was observed. With the crystal oriented so that the $(hk0)$ plane is in the scattering plane and the field along the c axis, on the other hand, no change in the magnetic structure is seen for fields up to 14 kG [Fig. 3(b)]. These observations are consistent with bulk measurements on single crystals of $ErNi_2B_2C$ which show that T_N is strongly field dependent for a field applied in the basal plane, but rather insensitive to a field applied along the tetragonal c axis; actually, these measurements show that for a field of 10 kG applied parallel to the b axis, the ordering temperature T_N for the Er moments is below 5.5 K. Thus, for fields exceeding approximately 5 kG applied along the b axis the magnetic moments are not completely ordered, as shown (Fig. 3) by the decrease in the intensity of the magnetic satellite, and as a result, a moment is induced by the applied field causing an increase in the nuclear intensities.

In summary, the magnetic structure of $ErNi_2B_2C$ is found to be different from that observed in the corresponding Ho compound. In particular, in Er, the moments below 6 K order in an incommensurate modulated structure with the wave vector parallel to $\mathbf{a}^*$, whereas in Ho an incommensurate modulated structure, characterized by two wave vectors, one along a^* (with magnitude only slightly larger than that observed in the Er compound) and one along $\mathbf{c}^*$, is observed only between 4.7 and 6 K. This may be the origin of the observed differences in the magnetotransport properties of these compounds. In particular, it is tempting to attribute the almost reentrant behavior of Ho in the vicinity of 5 K to the pair breaking associated with the transient modulated structure with the wave vector along c^*. This conclusion is also suggested by band theoretical calculations[13] of the generalized susceptibility $\chi(q)$ of $LuNi_2B_2C$ (without matrix elements). In these calculations, the generalized susceptibility for wave vectors along $\mathbf{a}^*$ has a pronounced peak for a wave vector of approximately 0.6, a value close to the values observed for Ho (0.585) and Er (0.553). Since the calculations were performed for $LuNi_2B_2C$ it is natural to assume that the modulated structure with the wave vector along $\mathbf{a}^*$ and magnitude close to 0.6 is a common feature of these compounds and can be attributed to a nesting of their Fermi surfaces. Clearly, calculations with matrix elements may provide a better understanding of the magnetic ordering in these compounds.

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