MAGNETIC STRUCTURES IN RNi_2B_2C (R = Ho, Er) SUPERCONDUCTORS

C. STASSIS, A. I. GOLDMAN, P. DERVENAGAS, J. ZARESTKY, P. C. CANFIELD, B. K. CHO, D. C. JOHNSTON, and B. STERNLIEB*
Ames Laboratory and Department of Physics and Astronomy, Iowa State University, Ames, IA 50011
*Physics Department, Brookhaven National Laboratory, Upton, NY 11973

ABSTRACT

Single crystal neutron diffraction techniques have been employed to study the evolution of magnetic structures in RNi_2B_2C compounds in an attempt to understand the relationship between magnetic ordering and superconductivity in several members of this series. For $HoNi_2B_2C$, below the superconducting transition (T_c = 8 K), an incommensurate magnetic structure characterized by two wave vectors (0.585 a* and 0.915 c*) is found in a narrow temperature range between 4.7 K and 6 K. This is the same temperature range where bulk measurements find a deep minimum in the upper critical field, H_{c2}. Below 4.7 K, $HoNi_2B_2C$ is a simple collinear anteferromagnet. $ErNi_2B_2C$ (T_c = 11 K) orders in an incommensurate modulated antiferromagnetic state characterized by an ordering wave vector 0.553 a* below 7 K, which coexists with superconductivity.

Many recent studies[1-10] of the interplay between superconductivity and magnetism have focused on the newly discovered RNi_2B_2C series (R = Rare Earth element).[1-4] The structure of these compounds is body-centered tetragonal and consists of R-C planes separated, along the c-axis, by Ni_2B_2 sheets. The layered structure is similar to that of the $ThCr_2Si_2$ compounds and it is reminiscent of the high-T_c oxide superconductors. Members of the RNi_2B_2C series such as $HoNi_2B_2C$, $ErNi_2B_2C$, and $TmNi_2B_2C$ are of particular interest here since bulk measurements indicated the onset of magnetic ordering at temperatures below the superconducting transition. For the superconductors $HoNi_2B_2C$ (T_c = 8 K) and $ErNi_2B_2C$ (T_c = 11 K), anomalies in the resistivity and upper critical field have been observed both in powder[5] and single crystal studies.[6] $HoNi_2B_2C$, in fact, exhibits reentrant,[5] or nearly reentrant,[6] behavior in a narrow temperature range around 5 K. Electronic band structure calculations[8-10] for the $RENi_2B_2C$ compounds suggest that these materials are conventional superconductors.

One of the first steps toward an understanding of the relationship between superconductivity and magnetic order in the RNi_2B_2C series is a systematic investigation of the types of magnetic structures present in these compounds. Single crystals of high quality and of sufficient size for neutron diffraction measurements were grown at the Ames Laboratory by the high-temperature flux growth technique and characterized by x-ray diffraction and magnetization measurements. For the samples discussed here, the boron was isotopically depleted in the heavily neutron absorbing ^{10}B nuclei. All samples crystallize as platelets of typical dimensions of 2×4×0.1 mm^3 with the c-axis of the tetragonal structure perpendicular to the flat face. For the Ho and Er compounds discussed in this paper, magnetization measurements were performed on single crystals from the same batch as those used in the neutron diffraction measurements using a Quantum Design SQUID magnetometer. For both compounds, the low-temperature normal

state susceptibility is highly anisotropic and suggests that the Rare Earth moment is constrained to lie in, or close to, the basal plane. Neutron diffraction measurements were performed using the triple axis spectrometers HB1A and HB2 at the HFIR of Oak Ridge National Laboratory and H7 at the HFBR of Brookhaven National Laboratory. Most measurements were performed at a constant incident energy of 14.7 meV using pyrolytic graphite reflecting from the (002) planes as the monochromator and analyzer, together with double pyrolytic graphite filters to minimize the $\lambda/2$ contamination of the incident beam. Typically, the samples were oriented so that the scattering plane was coincident with the a-c plane of the structure.

Above approximately 6 K, $HoNi_2B_2C$ is paramagnetic and only nuclear reflections (*hkl*) with $h+k+l=2n$ are observed in the neutron diffraction scans, consistent with the crystal structure (space group I4/mmm) of this compound.[3] As the temperature is decreased below about 6 K, but above approximately 5 K, three additional types of diffraction peaks start to develop, as shown in Figure 1.[11] Scattering develops at the positions of forbidden nuclear peaks (*hkl*) with

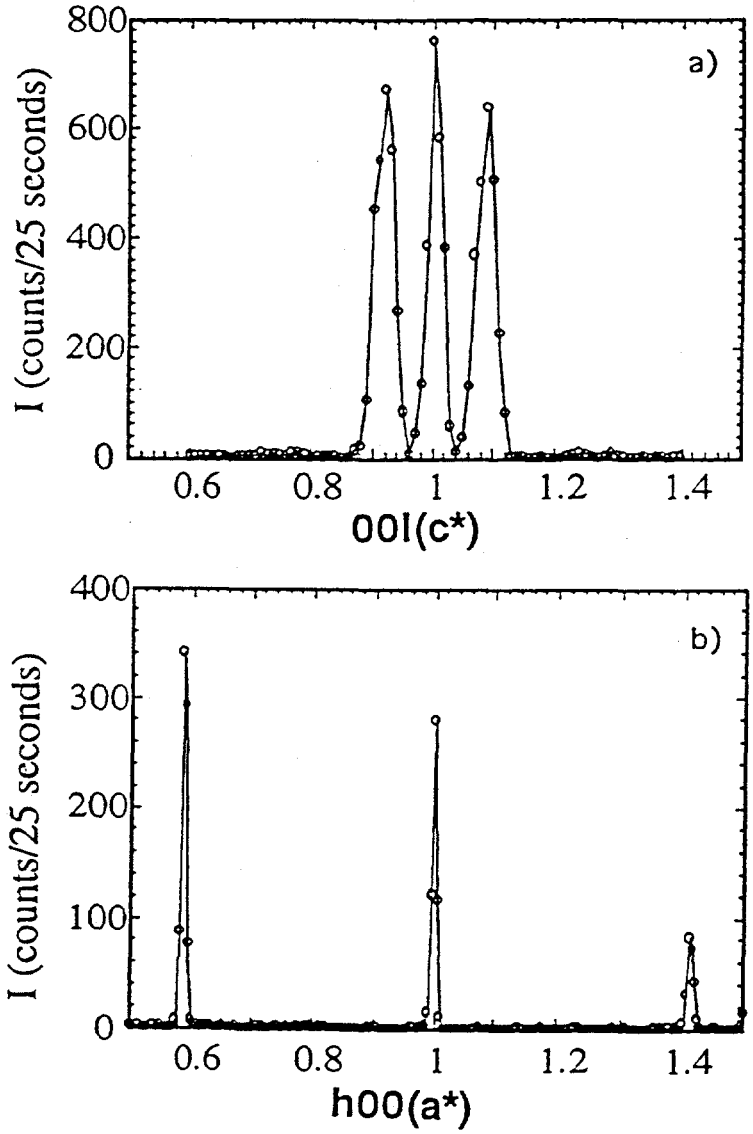

Figure 1. Neutron diffraction data along the a) 00l and b) h00 directions of $HoNi_2B_2C$ at T = 5.3 K. The lines in the figures are guides to the eye.

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$h+k+l=2n+1$ and, in addition, pairs of satellites to each nuclear reflection appear at wave vectors of 0.585 a* and 0.915 c* corresponding to additional incommensurate modulations along both the a-axis (or b-axis) and c-axis of the tetragonal structure. Third-order satellites of the modulation along the c-axis are also observed, indicating that the modulation is not purely sinusoidal, but squared. We point out here that it is not known, at this time, whether the modulations along the a- and c-axes are characteristic of a single domain or two, physically distinct magnetic structures.

Below approximately 5 K, Figure 2 shows a rather abrupt decrease in the intensity of both sets of incommensurate satellite peaks and a concomitant increase in the intensity of magnetic scattering associated with the commensurate antiferromagnetic structure. At low temperatures

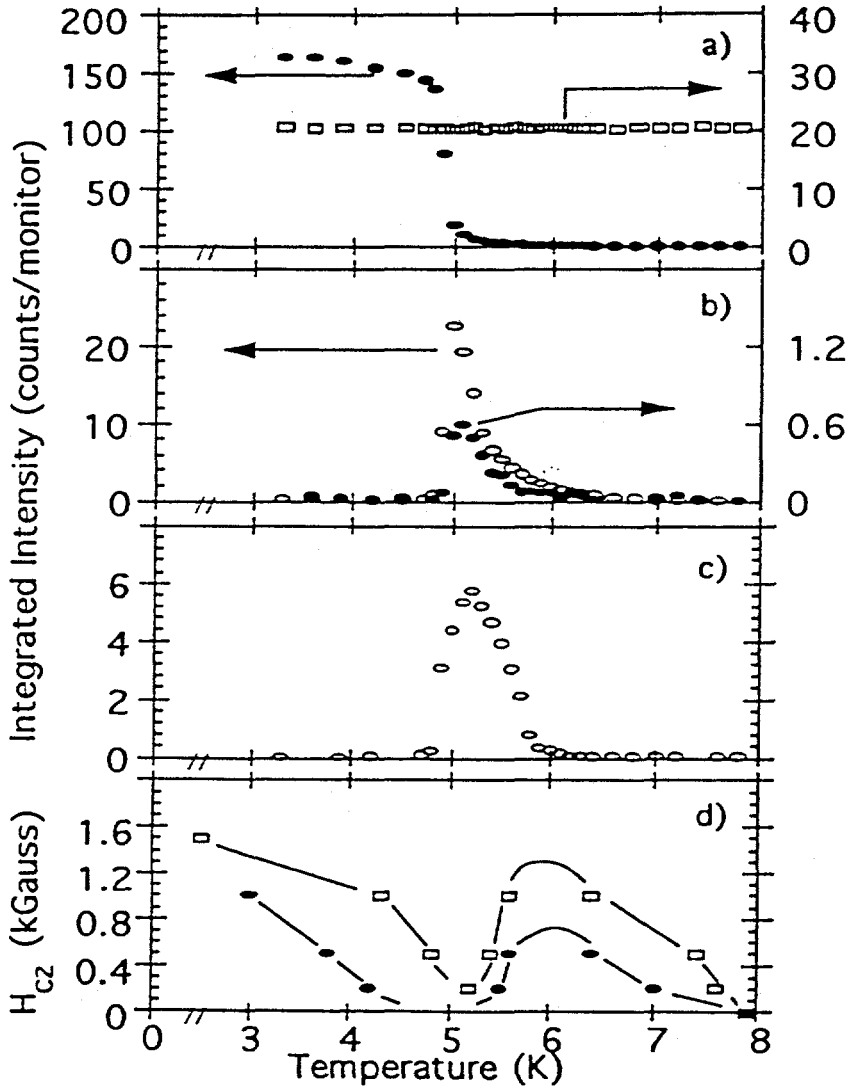

Figure 2. Temperature dependence of a) the (003) commensurate antiferromagnetic peak (filled ellipse) and the (101) nuclear Bragg peak (open rectangles); b) the first- (open ellipse) and third-order (filled ellipse) satellites associated with the modulation wave vector, K_1 = (0, 0, 2.915); c) the K_2 = (1.415, 0, 0) magnetic satellite; and d) the upper critical field, H_{c2} (Ref. 6) determined via temperature-dependent magnetization measured parallel (open rectangles) and perpendicular (filled ellipse) to the tetragonal c axis. Lines are intended as guides to the eye.

then, the magnetic structure is a commensurate antiferromagnet and the magnetic unit cell is the same as the chemical unit cell. The modulated magnetic structure(s) exist only in a very narrow temperature range between 4.7 K and 6 K. This is the same temperature range where there is a deep anomaly in the upper critical field (also shown in Figure 2). Indeed, for very small applied fields (about 20 G) in this temperature range, reentrant behavior has been observed in single crystal samples.[6] In powders on $HoNi_2B_2C$, reentrance in the absence of any applied field has been observed resistively.[5] In light of the data shown in Figure 2, then, it seems reasonable to attribute the deep minimum in H_{c2} to the pair-breaking interactions associated with the modulated magnetic structure in this temperature range. We also note that the strength of the interaction favoring the commensurate antiferromagnetic ground state in $HoNi_2B_2C$ seems close to that favoring the incommensurate modulation, and speculate that the energy associated with the stabilization of the superconducting ground state may indeed play a role in determining the ultimate low-temperature ground state of the coupled electron-local moment system.

Neutron powder diffraction measurements by Grigereit, et al.,[12] on $HoNi_2B_2C$, have yielded results similar to those reported for single crystal measurements.[11] These powder measurements, however, did not find evidence of the satellites at 0.585 a* described above and shown in Figure 1. The apparent absence of the a* satellites in their measurement is at odds with recent diffraction data[13] on powder samples produced at the Ames Laboratory, and is puzzling in light of recent results from both single crystal[14] and powder[15] neutron diffraction measurements on $ErNi_2B_2C$. As illustrated in Figure 3, single crystal measurements on the Er compound reveal that below approximately 7 K, magnetic satellites to the nuclear reflections develop at an incommensurate wave vector 0.553 a*, and persist down to the lowest temperature measured in the experiment (1.7 K). Interestingly, neither the satellites along c* nor scattering at the commensurate antiferromagnetic positions is found in $ErNi_2B_2C$.

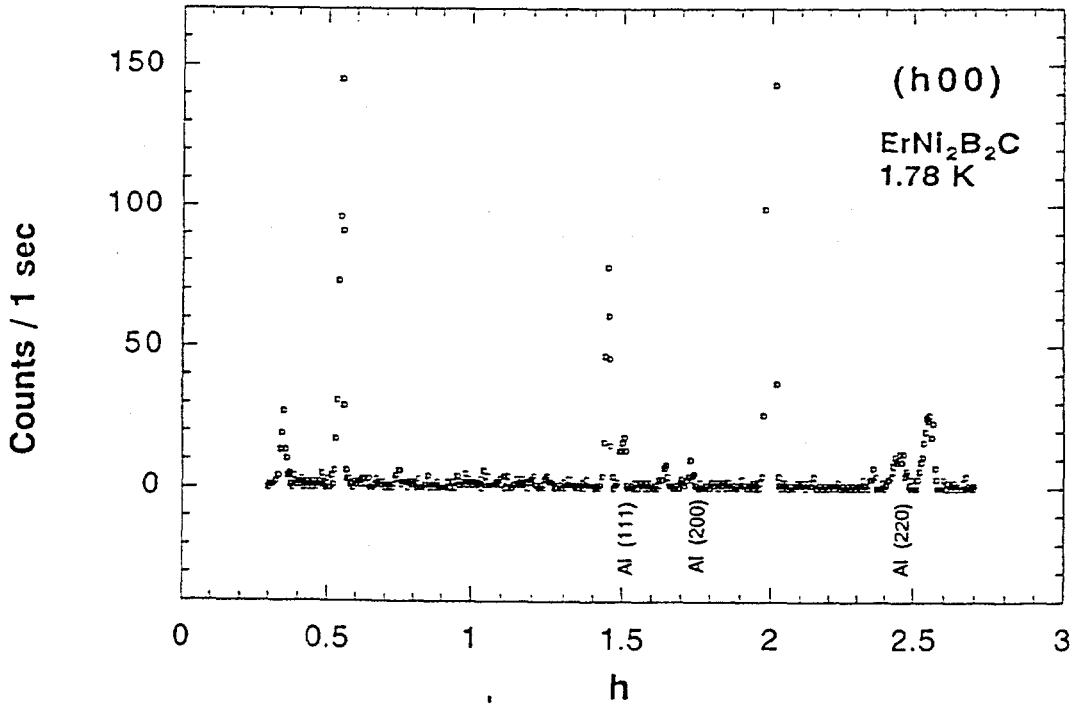

Figure 3. Neutron diffraction scan along the h00 direction of $ErNi_2B_2C$ at 1.8 K, showing the first and third order satellites associated with the modulation with wave vector 0.553 $\vec{a}$ *. Fifth, seventh, and ninth order satellites were also observed in scans with higher preset.

The common feature of a modulation along a* in both $ErNi_2B_2C$ and $HoNi_2B_2C$ is intriguing in light of recent band theoretical calculations of the generalized electronic susceptibility of $LuNi_2B_2C$ (without matrix elements).[16] These calculations suggest that a nesting feature along a* may be common to the normal state of all compounds in this family, since $\chi(q)$ exhibits a pronounced peak at a wave vector of approximately 0.6 a*. Neutron diffraction measurements on other members of the RNi_2B_2C family are underway to explore this issue and to further elucidate the relationship between superconductivity and magnetism in these novel compounds.

Acknowledgments

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