Neutron diffraction and theoretical model studies of the field induced magnetic phases of the $ErNi_2B_2C$ superconductor

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Introduction

Magnetism and superconductivity are two basic properties of materials that have been studied intensively for many years. While a solid understanding of magnetism has been developed during the last fifty years, there are significant basic properties that remain to be explained in superconductivity, as exemplified by the mechanism underlying high-temperature superconductivity. Superconductivity and static magnetic order are generally considered as competing phases. Back in the fifties and sixties, it was observed that superconductivity was strongly suppressed or extinguished by substitution of magnetic impurities at a 1% level [1, 2]. However, in the seventies it was revealed that true long-range magnetic order co-existed in Chevrel phase superconductors like RMo_6S_8 and the related RRh_4B_4 (R = rare earth) [3]. Here it is argued that the detrimental effect of the rare earth ions is avoided, because they form a fully ordered sublattice that is isolated from the conduction electrons. Furthermore, the ordering sets in at rather low temperature (approximately 1 K) and has a modulated structure that averages out on a length scale of the superconducting order parameter. In the late eighties all attention was given to the high-temperature superconductors where highly interesting, but so far unraveled, interactions exist between the magnetic and the superconducting electrons or holes within the same copper-oxide planes.

The rare-earth nickel boron-carbides RNi_2B_2C have attracted much attention after their discovery in 1994 [4, 5], because superconductivity and antiferromagnetism co-exist for R = Dy, Ho, Er and Tm at comparable temperatures $6\ K \le T_c \le 11\ K$ and $1.7\ K \le T_N \le 11\ K$ [6]. Here, T_N is the Néel temperature. Above this temperature, the antiferromagnetic order in a crystal is lost, similarly to the superconducting properties that are lost above the critical temperature T_c. $ErNi_2B_2C$, in particular, is interesting because magnetization studies have indicated weak ferromagnetism below $T_c = 2.3\ K$ in zero-field and because several magnetic phases develop in an applied magnetic field [7]. Single-crystal neutron-diffraction studies in zero field [8] have corroborated these results and confirmed the earlier neutron powder-diffraction findings [6] that the magnetic phases are characterized by a transversely polarized spin-density wave with an ordering vector (wave vector) $Q \approx 0.55\ a^*$ (or b^*, where a^* and b^* are reciprocal lattice vectors) and with the spins lying in the basal plane of the tetragonal crystal structure (see Fig. 1). If $Q = 0.5\ a^*$ was a stable configu-

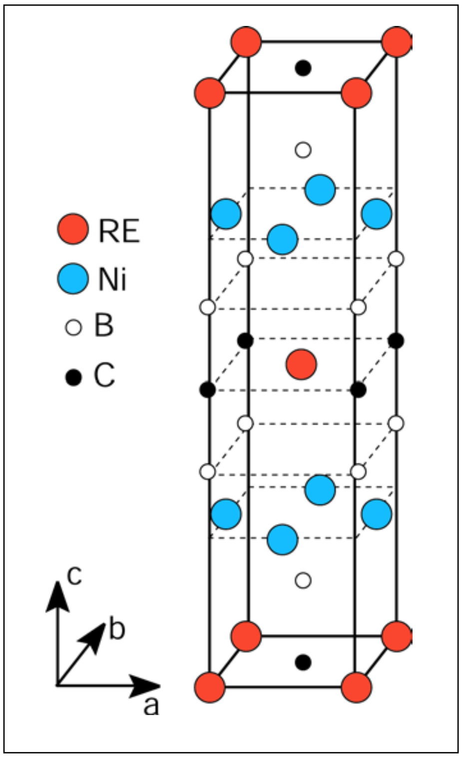

Fig. 1: Crystal structure of the rare-earth borocarbides

ration, the magnetic structure would consist of ferromagnetic sheets, and since the unit cell contains two Er-ions, the stacking sequence would be double layers: up-up-down-down etc. However, nesting at the Fermi surface results in phases with $Q \geq 0.55\ a^*$ and introduces phase slips in the stacking of the commensurate sequences.

Recently, a mean-field model has been established and shown to account for most of the observed experimental data, including the transition from weak ferromagnetism to antiferromagnetism at $2.3\ \mathrm{K}$ shown in Fig. 2 [9]. The model suggests that the magnetic transitions result from a series of structures with ordering vectors $Q = n/m\ a^*$ (or b^*) with $0.55 \leq n/m \leq 0.60$. The present neutron-diffraction study aims to establish the modulation vectors of the stable magnetic phases and to compare the results with the predictions of the mean-field model. A detailed account of the model is published in ref. 9, and the experimental results and model analyses are presented in ref. 10.

Results and discussion

Two stable structures have been identified with ordering vector $Q = 11/20\ a^*$ (or b^*) in zero field [9]. Above $T_c = 2.3\ \mathrm{K}$ there is an equal number of up and down spins, but below T_c one of the spin directions becomes more populated, leading to weak ferromagnetism as visualized in Fig. 2. At low temperatures the structures are squared-up and not sinusoidal as they are close to T_N.

A field applied along [0 1 0] results in the formation of two different domains. Since the spins are Ising like, the most favorable domain has $Q = n/m\ a^*$. When increasing the field H more up-spins are formed and phase transitions between structures with different commensurate modulation vectors result, as shown in Fig. 3a. The transverse domain stays at $Q \approx 16/29\ b^* \approx 0.55\ b^*$ and decreases gradually in intensity as the field is increased. The H-T-phase diagram in Fig. 3a agrees well with data derived from bulk measurements [7, 11].

When applied along [0 0 1] the critical field for the antiferromagnetic phase is as high as $\sim 17\ \mathrm{T}$ at $1.8\ \mathrm{K}$. The phase diagram, studied in fields up to $12\ \mathrm{T}$ (the magnetic torque detached the crystal at higher fields), is shown in Fig. 3c. A gradual change of Q from $\approx 11/20$ to $\approx 5/9\ a^*$ (or b^*) results when the field and/or the temperature is increased.

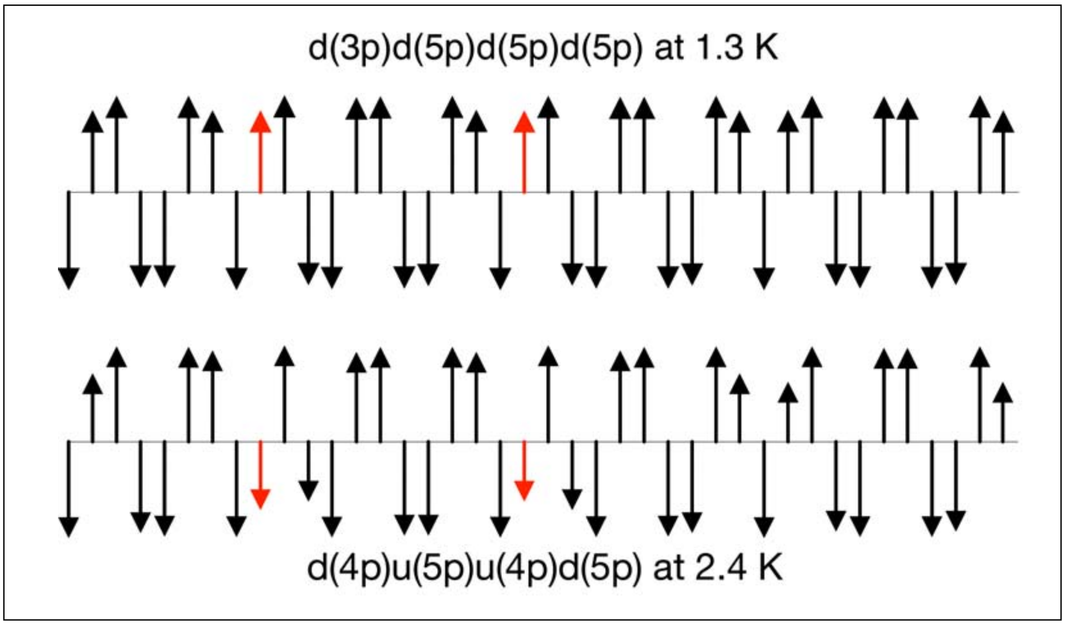

Fig. 2: One period of 40 layers of the $Q = 11/20$ zero-field structures calculated below the Curie temperature at $1.3\ \mathrm{K}$, and in the antiferromagnetic phase at $2.4\ \mathrm{K}$ (the red arrows indicate those layers in which the moments are reversed at the transition).

Studies performed with the field along the [1 1 0] direction reveal a series of ordering vectors $Q = Q_p + Q_\delta$ as a function of field and temperature. The principal ordering vector Q_p along a^* or b^* ranges from $Q_p \approx 11/20$ to $\approx 18/31$ reciprocal lattice units rlu, as shown in Fig. 3b. A peculiarity is a small, but clearly observable, rotation of the principal ordering vector Q_p by an orthogonal component $Q_\delta \approx -0.005\ b^*$, which occurs close to and above the superconducting critical field, H_{c2}.

The mean-field model has been a valuable tool in the interpretation of the neutron diffraction and the bulk magnetization data. It accounts for the stability of many of the observed phases, but not for the small finite Q_δ. This small rotation shows that the exchange interaction has anisotropic components.

Acknowledgements
This work is supported by the EU Commission under contract HPRI-CT-2001-00138 on neutron-diffraction experiments at BENSC, the Danish Technical Research Council via the Framework Program on Superconductivity, and the Danish Natural Science Research Program via DANSCATT.

[1] M.B. Maple, Applied Physics **9**, 173 (1976)
[2] A.A. Abrikosov and L.P. Gorkov, Sov. Phys. JETP **12**, 1243 (1961).
[3] M.B. Maple and O. Fischer (edt.): *Superconductivity in Ternary Compounds, Topics in Current Physics*, Vols. I and II, (Springer, Berlin, 1982).
[4] R. Nagarajan et al. Phys. Rev. Lett. **72**, 274 (1994).
[5] R.J. Cava et al. Nature **367**, 254 (1994).
[6] J.W. Lynn et al. Phys. Rev. B **55**, 6584 (1997).
[7] P.C. Canfield et al., Physica C **262B**, 249 (1996).
[8] S.-M. Choi et al., Phys. Rev. Lett. **87**, 107001 (2001).
[9] J. Jensen, Phys. Rev. B **65**, 140514 (2002).
[10] A. Jensen et al., Phys. Rev. B **69**, 104527 (2004).
[11] S.L. Budko and P.C. Canfield, Phys. Rev. B **61**, R14932 (2000).

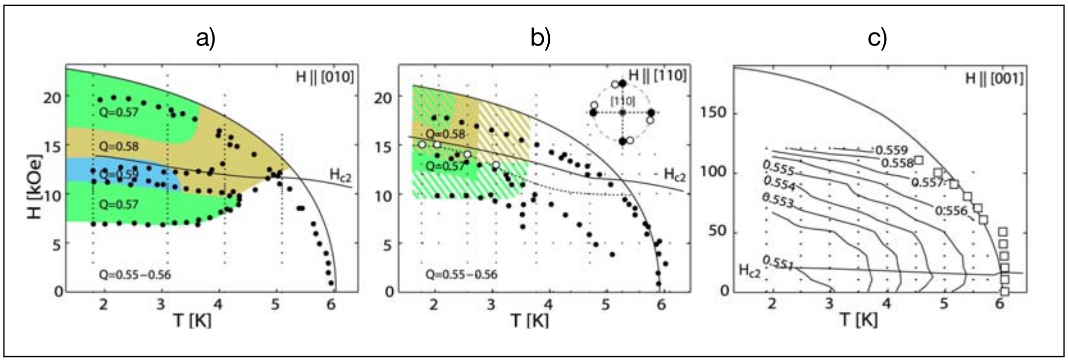

Fig. 3: Magnetic phase diagrams of $ErNi_2B_2C$ for the magnetic field applied along a) [0 1 0] , b) [1 1 0] and c) [0 0 1]. Small black dots mark the (T, H) points where the measurements were made. The outermost boundary lines in a) and b) interpolate points where the magnetic intensity disappears. In c) the boundary line is the Néel temperature determined by the mean-field model. A fine solid line marks the superconducting critical field H_{c2}, and the solid circles indicate the magnetic phase lines derived from magnetization measurements [11]. Four commensurate phases are presented in the phase diagram a) with H || [0 1 0]. In b), H || [1 1 0], coloured areas indicate the existence of one of the three phases, and the striped areas represent coexistence of two phases. The modulation vector has a finite transverse component above the dashed line, and the open circles indicate that the orthogonal component is zero at lower field. The insert shows schematically how the magnetic reflections are rotated in reciprocal space. In c), with H || [0 0 1], the contours indicate that Q increases continuously as function of H and T; the open squares represent the Néel temperature derived from the data.