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The effect of magnetic ordering on the crystal field levels of $ErNi_2B_2C$

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Abstract

The quaternary compound $ErNi_2B_2C$ becomes superconducting with a critical temperature of 9 K. DC magnetisation indicates the presence of magnetic ordering of the Er moments at ≈ 7 K. Specific heat measurements show a Schottky anomaly around 45 K with a magnitude of 10 J mol K^{-1} which agrees well with our model for the crystal-field scheme and neutron scattering data. We observed changes in the widths and positions of peaks in the crystal-field spectra, indicating the build up of magnetic interactions with the f electrons and thus contributing to the crystal-field Hamiltonian.

1. Introduction

The newly discovered compounds with the composition $(RE)Ni_2B_2C$ (RE = rare-earth) show relatively high superconducting transition temperatures [1,2] despite the presence of magnetic rare earth ions (Ho, Er, Tm, etc.) and the 3d transition element Ni. Unlike the high T_c 123 superconducting oxides (where $T_c \approx 92$ K for most RE ions) [3] the rare-earth ions influence the superconductivity, with T_c decreasing from Tm to Dy and the nonmagnetic RE (Y, Lu) having the highest transition temperatures [2]. The variation of T_c with the magnetic ion suggests that there is a degree of interaction between paired conduction electrons and magnetic moments [5], similar to that observed in the $(RE)Rh_4B_4$ compounds [6]. Anomalous behaviour in the upper-critical-field measurements have been reported [5] in $HoNi_2B_2C$ ($T_c \approx 8$ K) showing reentrant behaviour at 5 K in zero field. Obviously, the rare-earth ions play an important role in these new materials and for this reason we have started to investigate the crystalline-electric-field level scheme for different rare-earth ions in the $(RE)Ni_2B_2C$ compound by inelastic neutron scattering. From the tetragonal symmetry of the RE site in $(RE)Ni_2B_2C$, the Hamiltonian is given by

$$H_{CEF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_4^4 O_4^4 + B_6^4 O_6^4$$

where B_m^n denotes the CEF parameters and O_m^n are the equivalent Stevens operators.

In this paper we present the results from experiments on $ErNi_2B_2C$ showing evidence of strong magnetic correlations around the Er site above and below T_N.

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2. Experimental details

Samples were prepared by the standard arc-melting technique. High purity constituents Er (> 99.9%), Ni (> 99.99%), ^{11}B (> 99.5%) and graphite flakes were melted on a water-cooled copper hearth under an argon atmosphere. The resulting buttons were turned over and remelted several times to insure good homogeneity. The ingots were then wrapped in tantalum foil, sealed in an evacuated quartz tube and annealed at (975–1050) °C for 12–16 h. X-ray diffraction showed some very weak impurity peaks in some of the samples. Magnetic-susceptibility measurements between 4 and 300 K were performed using a vibrating-sample magnetometer in fields up to 1.5 T. Specific-heat measurements between 20 and 300 K were performed in an adiabatic-sweep calorimeter on a 100 mg sample. The non-magnetic RE, YNi_2B_2C was used as a reference for the phonon contribution to the specific-heat data. The neutron inelastic scattering experiments were performed on the chopper spectrometer HET, at the pulsed neutron source ISIS, Rutherford Appleton Laboratory. Incident energies up to 60 meV were used to explore the crystal-field spectra of a 20 g polycrystalline sample of $ErNi_2B_2C$. The experiments were performed in either a closed-cycle refrigerator (12–300 K) or an orange cryostat (1.2–300 K).

3. Results and discussion

Fig. 1 shows the magnetic susceptibility of $ErNi_2B_2C$ in an applied field of 1 T. The susceptibility is Curie–Weiss-like above the superconducting transition temperature (T_c) with a μ_{eff} of $7.3\mu_B$ per Er ion (cf. $9.59\mu_B$ for free trivalent ion). The DC susceptibility versus temperature has a maximum at 7 K. This corresponds to the magnetic ordering of the Er moments, since the applied field is well above H_{c2} for this compound [5].

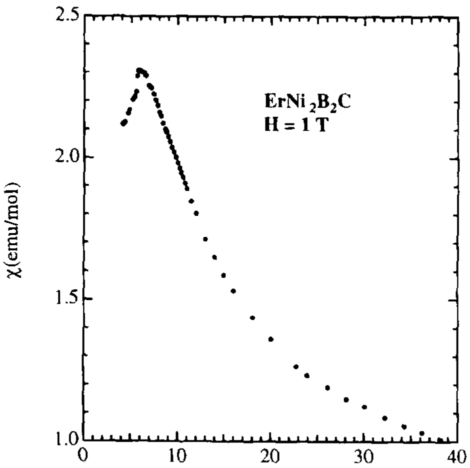

Fig. 1. DC magnetic susceptibility of $ErNi_2B_2C$ zero-field cooled then measured with a magnetic field of 1 T.

To discriminate between different possible crystalline electric fields (CEF) schemes, specific-heat measurements were also carried out on $ErNi_2B_2C$. The phonon contribution to the $ErNi_2B_2C$ specific heat was determined using the isostructural compound YNi_2B_2C, since the rare-earth ion, Y, is non-magnetic. Fig. 2 shows the specific-heat data after the phonon contribution has been subtracted. The calculated specific heat, from the best CEF level scheme to date, is also shown. The Schottky anomaly at 45 K has a magnitude of 10 J mol^{-1} K^{-1} this is reproduced well, in both magnitude and position by the predicted specific heat from the CEF model.

Preliminary calculations of the crystal-field parameters from neutron data give:

$$B^0{}_2 = (- 0.641 \times 10^{-1} \pm 0.6 \pm 10^{-3})\ \text{meV},$$

$$B^0{}_4 = (- 0.337 \times 10^{-3} \pm 0.7 \times 10^{-5})\ \text{meV},$$

$$B^0{}_6 = (0.826 \times 10^{-6} \pm 0.1 \times 10^{-7})\text{meV},$$

$$B^4{}_4 = (- 0.145 \times 10^{-2} \pm 0.1 \times 10^{-3})\text{meV},$$

$$B^4{}_6 = (- 0.281 \times 10^{-4} \pm 0.6 \times 10^{-6})\text{meV}.$$

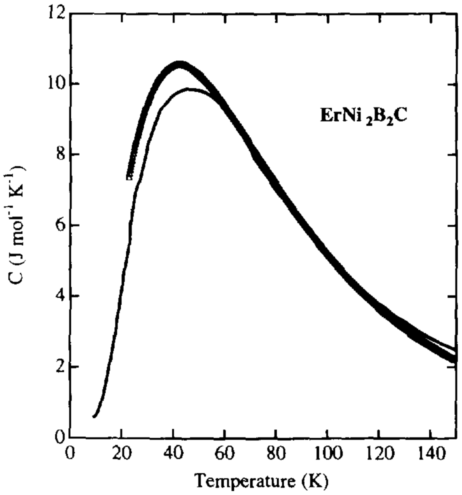

Fig. 2. Magnetic contribution to the specific heat of $ErNi_2B_2C$. Triangles indicate the experimental data while the estimate from the CEF model is the smooth curve. The specific heat of YNi_2B_2C was subtracted from the specific heat of $ErNi_2B_2C$ to remove the phonon contribution.

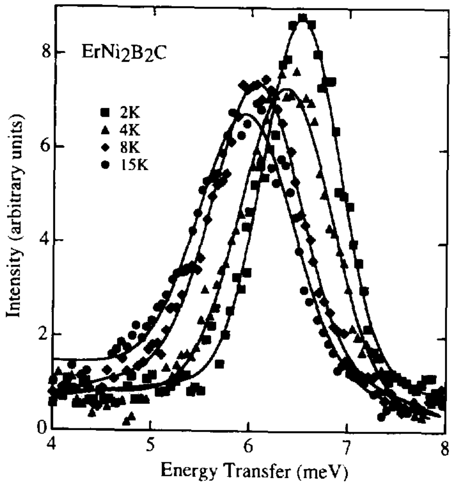

Fig. 3. A single CEF excitation of $ErNi_2B_2C$ at temperatures between 2 and 15 K. The shift in position and width is due to an extra mean-field contribution to the CEF hamiltonian from the build up of magnetic correlations between rare-earth ions.

Fig. 3 shows the well defined CEF peak at about 6.5 meV at four different temperatures above and below T_c and T_N. The crystal-field model predicts that this is a single excitation from the ground state to the first CEF level. As the temperature decreases, even above T_N, the peak centre shifts, indicating the presence of an exchange interaction with the f electrons. Recent neutron-diffraction results [7] have indicated that $ErNi_2B_2C$ orders with ferromagnetic planes antiferromagnetically coupled along the *c*-axis. Strong ferromagnetic correlations in the *a–b* plane could account for the shift above T_N, before even larger local effects dominate below T_N, causing a further increase in shift.

4. Conclusion

We present evidence from neutron-scattering data of a shift in the crystal-field levels due to magnetic correlations above and below T_N. We have also presented initial crystal-field parameters for the magnetic superconductor $ErNi_2B_2C$ which agree well with specific heat data.

References

[1] C. Mazumdar et al., Solid State Commun. 87 (1993) 413; R.Nagarajan et al., Phys. Rev. Lett. 72 (1994) 274.
[2] R.J. Cava et al., Nature 367 (1994) 252.
[3] L. Soderholm et al., Phys. Rev. B 43 (1991) 7923.
[4] R.J. Cava et al., Nature 367 (1994) 146.
[5] H. Eisaki et al., Phys. Rev. B 50 (1994) 647.
[6] W.A. Fertig et al., Phys. Rev. Lett. 38 (1977) 987.
[7] D.McK. Paul et al., Physica B 213&214 (1995) 107.