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Superconducting and magnetic properties of DyNi₂B₂C single crystals

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Abstract

We have investigated the superconducting and magnetic properties of DyNi₂B₂C single crystals for applied fields parallel and perpendicular to the *c*-axis. Magnetisation measurements show that this compound has highly anisotropic properties in both the normal and the superconducting state. The normal state susceptibility follows a Curie–Weiss temperature dependence for applied field perpendicular to the *c*-axis and a much weaker temperature dependence if the field is along the *c*-axis. For magnetic fields applied perpendicular to the *c*-axis, the magnetisation versus field curves show anomalies below T_N with clear discontinuities and eventual saturation for $H > 30$ kOe, while showing a normal paramagnetic behaviour when the field is applied along the *c*-axis.

1. Introduction

The discovery of RNi₂B₂C (R = rare earth) [1] compounds has opened the possibility of studying the interplay between superconductivity and long-range magnetic order. These compounds form for most of the *R* ions and show superconductivity for magnetic as well as nonmagnetic rare earth ions. The layered structure [2] of these compounds (tetragonal, space group I4/mmm) consists of R–C layers well-separated by the Ni₂–B₂ layers. However, a degree of interaction can be deduced from the fact that nonmagnetic rare earth ions (Y and Lu) have the highest T_c when compared to the magnetic rare earth ions, where T_c decreases from Tm to Ho. In fact, the coexistence of magnetic ordering of the Ho, Er and Tm moments below the superconducting transition temperature has been established from neutron diffraction and specific heat measurements [3–8]. DyNi₂B₂C orders with a T_N of 10.5 K. Recently, we have shown that bulk

superconductivity can be obtained in DyNi₂B₂C with a T_c of 6.5 K [9] if the stoichiometry of carbon is kept close to the nominal composition of 1.0. These results were later confirmed for single crystals [10]. DyNi₂B₂C is unique amongst the RNi₂B₂C compounds in that the superconducting transition takes place in a magnetically ordered state. Both T_c and T_N values are relatively high compared to the other compounds showing similar behaviour.

The availability of sizeable single crystals of RNi₂B₂C compounds has prompted us to study the anisotropic nature of the superconducting and magnetic properties of DyNi₂B₂C. The compound shows highly anisotropic properties both in the normal and superconducting states. For small applied fields, the compound shows a full shielding and a positive Meissner fraction when the field is applied parallel to the *c*-axis ($H//c$) but much smaller fractions when the field is applied perpendicular to the *c*-axis ($H\perp c$). The normal susceptibility follows Curie–Weiss law for $H\perp c$ while a weak temperature dependence is seen for $H//c$ with a pronounced maximum at around 100 K. For $H\perp c$, the magnetisation

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isotherms show anomalies below T_N with clear steps corresponding to metamagnetic states while a normal paramagnetic behaviour is observed for $H//c$.

2. Experiments

Polycrystalline samples of $\text{DyNi}_2\text{B}_2\text{C}$ were prepared by standard arc-melting [9]. Single crystals were prepared using Ni_2B as flux [11]. The crystals were characterised by X-ray Laue photographs and AC susceptibility measurements. The magnetisation versus field was measured using a VSM (Oxford Instruments) and the magnetic susceptibility using a SQUID (Quantum Design).

3. Results and discussion

$\text{DyNi}_2\text{B}_2\text{C}$ shows antiferromagnetic ordering at $T_N = 10.5$ K and superconductivity in a magnetically ordered state at $T_c = 6.5$ K [10]. In order to investigate the anisotropic magnetic and superconducting properties, the magnetisation was measured as a function of temperature and field for both $H//c$ and $H\perp c$. Fig. 1 shows the low field magnetisation of a $\text{DyNi}_2\text{B}_2\text{C}$ single crystal for an applied field of 10 Oe for both $H//c$ and $H\perp c$. For $H//c$ the ZFC magnetisation corresponds to 100% for the shielding fraction. However, when the crystal was cooled in the same field and the FC

magnetisation measured, the signal showed positive values, making it impossible to calculate the Meissner fraction. This anomalous paramagnetic Meissner signal was observed for fields $H < 40$ Oe, above which the Meissner signal becomes negative. For $H\perp c$, the magnetisation gives a smaller fraction for shielding (80%) and Meissner effect (20%) due to a strong paramagnetic contribution from the Dy moments.

Fig. 2 shows the anisotropic magnetisation of a $\text{DyNi}_2\text{B}_2\text{C}$ single crystal in the superconducting state ($T = 1.8$ K). For $H\perp c$ (Fig. 2(a)), the magnetisation shows anomalies with clear steps. These steps corresponds to different field-driven metamagnetic states for this compound. The magnetisation eventually shows a saturation for magnetic fields greater than 30 kOe. When the field is decreased, the magnetisation shows hysteresis. From the saturation value of the magnetisation, the magnetic moment of Dy is estimated to be $9.55\mu_B$, which compares well with the free ion moment of Dy. The steps and saturation observed in the magnetisation are temperature dependent and disappear above T_N . The magnetisation then shows a normal paramagnetic behaviour. However, no anomalies are observed in the magnetisation for $H//c$, as shown in Fig. 2(b). The upper critical field values at 1.8 K were estimated from the low field magnetisation to be $H_{c2} = 2$ and 2.5 kOe for $H\perp c$ and $H//c$, respectively. These values are smaller than those obtained from our resistance measurements [10] in magnetic fields, where very little anisotropy was observed in the upper critical fields.

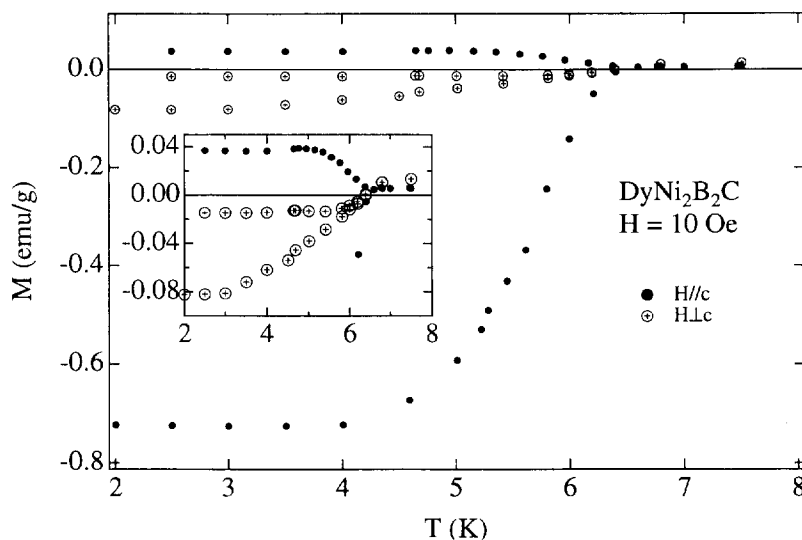


Fig. 1. Low field DC magnetisation, zero field cooled (ZFC) and field cooled (FC), for a single crystal of $\text{DyNi}_2\text{B}_2\text{C}$ for applied fields both parallel ($H//c$) and perpendicular ($H\perp c$) to the c -axis. The inset shows the magnetisation curves on an expanded scale.

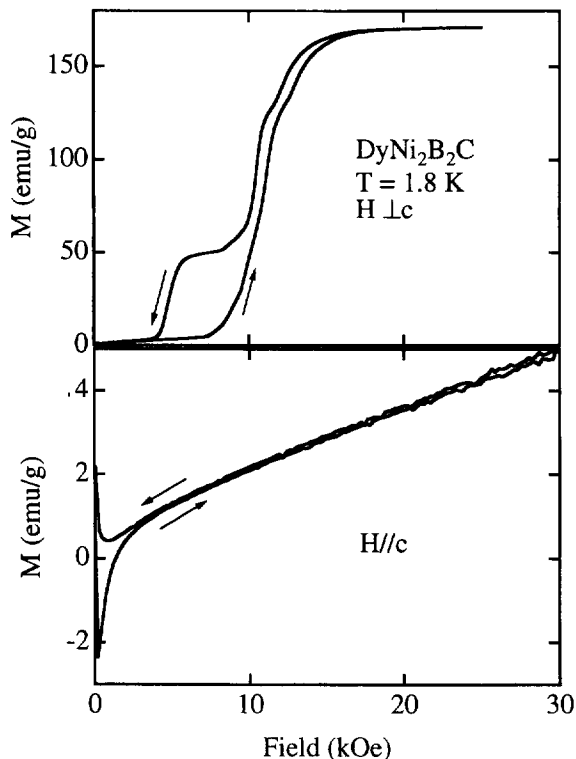


Fig. 2. The variation of magnetisation with applied fields for a $\text{DyNi}_2\text{B}_2\text{C}$ single crystal for (a) $H//c$ and (b) $H\perp c$ at 1.8 K. The arrows indicate the forward and reverse direction where the magnetic field is increased and decreased, respectively.

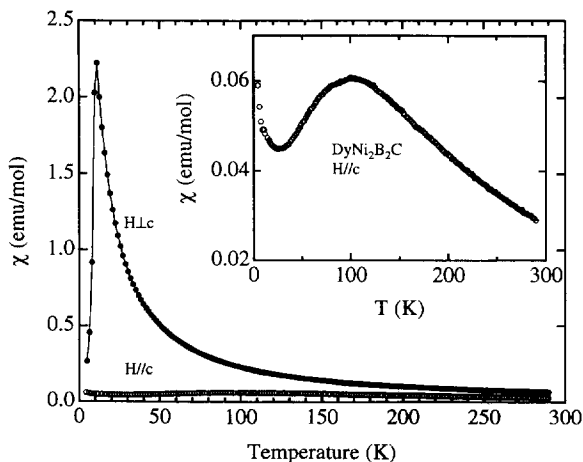


Fig. 3. The normal state magnetic susceptibility for a $\text{DyNi}_2\text{B}_2\text{C}$ single crystal for both $H//c$ and $H\perp c$ in an applied field of 5 kOe. The inset shows the anomalous susceptibility behaviour when the field is applied parallel to the c -axis.

The temperature dependence of the magnetic susceptibility of a $\text{DyNi}_2\text{B}_2\text{C}$ single crystal in a fields of 5 kOe, for both the directions of the applied field is shown in Fig. 3. The magnetic susceptibility follows a Curie–Weiss-like temperature dependence with a peak at the ordering temperature ($T_N = 10.5$ K) for $H\perp c$, whereas the paramagnetic susceptibility is much weaker for $H//c$ with a broad maximum at around 100 K. The anisotropy in magnetic susceptibility persists up to 300 K, but becomes less pronounced at higher temperatures. The anisotropy as well as the broad maximum in susceptibility may be due to CEF effects, as observed earlier in $\text{HoNi}_2\text{B}_2\text{C}$ single crystals [12]. The high-temperature tail of the susceptibility ($T > 200$ K) was fitted to a Curie–Weiss equation which gives the effective magnetic moment as $\mu_{\text{eff}} = 12.38\mu_B$ and $\theta_p = +16.4$ K for $H\perp c$ and $\mu_{\text{eff}} = 9.0\mu_B$ and $\theta_p = -3$ K for $H//c$. The difference in the θ_p values may also be a manifestation of the CEF effects in this compound. Detailed analysis of the CEF levels in this compound is in progress.

$\text{DyNi}_2\text{B}_2\text{C}$ is the only compound in the $\text{RNi}_2\text{B}_2\text{C}$ series which exhibits superconductivity ($T_c = 6.5$ K) after ordering magnetically ($T_N = 10.5$ K). The highly anisotropic magnetic properties displayed by this compound are probably due to the fact that the Dy moments are predominantly confined to the a - b plane. This suggestion is confirmed in the neutron diffraction measurements [13]. The observation of the paramagnetic Meissner effect for smaller applied fields needs further investigation. The anomalies in the magnetisation versus field curves suggest the formation of metamagnetic states with the Dy moments flipping to easy orientations before becoming aligned along the field direction to produce the saturated paramagnetism. The anisotropy in the normal state susceptibility indicates the presence of strong CEF effects in this material. The broad maximum in the susceptibility for $H\perp c$ around 100 K is a manifestation of these CEF effects, which in turn forces the moments to lie in the a - b plane. The magnetic properties of this compound resemble those of the $\text{HoNi}_2\text{B}_2\text{C}$ compound [12].

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