Superconductivity and magnetism in $DyNi_2B_2C$ single crystals

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Superconductivity has been observed in $DyNi_2B_2C$ single crystals at 6.4 K. $DyNi_2B_2C$ also orders antiferromagnetically at ~ 10.5 K and is a unique member of RNi_2B_2C compounds (R = Y or rare earth) in that the onset of superconductivity takes place in a magnetically ordered state. The crystals exhibit anomalous field-cooled magnetization in low applied fields of 1–5 Oe. Specific-heat measurements on the crystals give further evidence for the bulk nature of the magnetic order. The upper critical field H_{c2} measured parallel and perpendicular to the c axis of the crystals, shows little or no anisotropy. $H_{c2}(0)$ for $DyNi_2B_2C$ is deduced to be 0.7 T.

I. INTRODUCTION

The RNi_2B_2C compounds, where R = Y or rare earth, forms for most of the R ions and superconductivity above 4 K has been reported for R = Y, Lu, Tm, Er, and Ho,[1] with the highest T_c of 16.5 K being observed for $LuNi_2B_2C$. There is a considerable degree of interaction between the rare-earth ions and the conduction electrons in these compounds which brings about a reduction in T_c for the compounds containing magnetic rare earths, and for some members of the series there is a coexistence of magnetism and superconductivity. $HoNi_2B_2C$ and $ErNi_2B_2C$ exhibit antiferromagnetic order below T_c and the exact nature of ordering of the Ho and Er moments has been well established from neutron diffraction studies.[2–6] There is evidence from specific-heat measurements that $TmNi_2B_2C$ also orders magnetically below 1.5 K.[7]

We have recently shown that another member of this series, $DyNi_2B_2C$, can be made superconducting with a T_c onset at ~ 6.5 K, provided the stoichiometry is maintained close to the nominal composition, 1:2:2:1.[8] In the same paper, we have showed that phase purity is crucial to the observation of superconductivity in this compound and that the onset of superconductivity takes place in a magnetically ordered state. Evidence for magnetic order in $DyNi_2B_2C$ comes from our magnetic susceptibility, resistivity,[8] and neutron powder diffraction experiments.[9] Magnetic susceptibility measurements on our arc-melted ingots show an anomaly at 10.5 K (Ref. 8) and resistivity measurements on these samples show a change in slope at the same temperature. Preliminary analyses of our powder neutron diffraction data on $DyNi_2B_2C$ show the presence of antiferromagnetic peaks below 10 K. A detailed analysis of the exact nature of this magnetic ordering will be presented shortly.[9]

The observation of superconductivity in a magnetically ordered state in our arc-melted samples of $DyNi_2B_2C$ and the importance of phase purity motivated us to grow single crystals of this compound. Here we report on electrical resistance, magnetic susceptibility, magnetization, and upper critical field measurements for single crystals of $DyNi_2B_2C$, carried out to confirm our earlier findings on arc-melted ingots. We also present specific-heat measurements performed on a single-crystal specimen which provide conclusive evidence for the bulk nature of the magnetic order in $DyNi_2B_2C$.

II. EXPERIMENTAL DETAILS

Single crystals of $DyNi_2B_2C$ were grown by the flux technique using Ni_2B as flux.[10] Several sizable, good quality single-crystal platelets were obtained. The crystals were characterized by x-ray Laue photographs as having their c axis pointing out of the plane of the platelets. Crystals measuring $\sim 2\times 2\times 0.1$ mm^3 were used for the measurements. A standard four-probe dc method was used for the resistance measurements. Heat-capacity data were collected using a relaxation method between 2 and 20 K on a 4 mg single-crystal specimen. The upper critical field, H_{c2}, was determined by resistance measurements in magnetic fields of up to 1 T. A current of 20 mA was injected along the ab plane of the crystals while the magnetic field was applied parallel or perpendicular to the ab plane. In both geometries the current flow was perpendicular to the direction of the applied field. The dc magnetization was measured from 2 to 300 K using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design).

III. RESULTS AND DISCUSSION

Figure 1 shows the results of the dc magnetization measurements on a single crystal of $DyNi_2B_2C$, in a field $H=5$ Oe, applied parallel to the c axis of the crystal. From the zero-field-cooled (ZFC) magnetization curve, the magnetic shielding is estimated to be 100% of that expected for perfect diamagnetism. However, on cooling in the same field, the field-cooled (FC) magnetization shows an anomalous behavior, with positive values below T_c. Cooling in a smaller field (e.g., 1 Oe) produces an increase in the magnitude of the positive signal below T_c. This behavior was also seen in our arc-melted ingots.[8] There appears to be a strong paramagnetic contribution in the superconducting state due to the ordering of the Dy moments above T_c. Paramagnetic Meissner effect has

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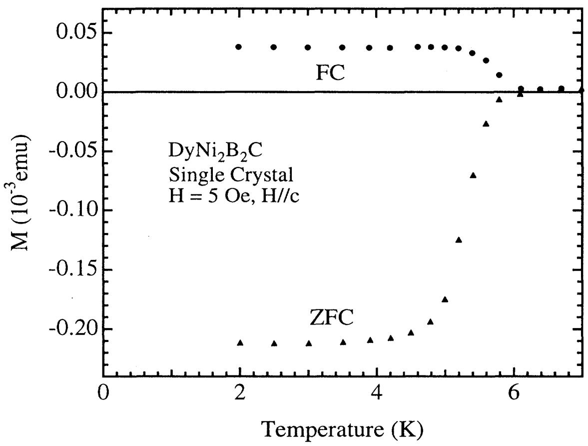

FIG. 1. ZFC and FC magnetization for a single crystal of $DyNi_2B_2C$ measured in a field of 5 Oe applied parallel to the c axis, showing the superconducting transition. An unusual positive value for the field-cooled magnetization in small applied fields is observed below T_c.

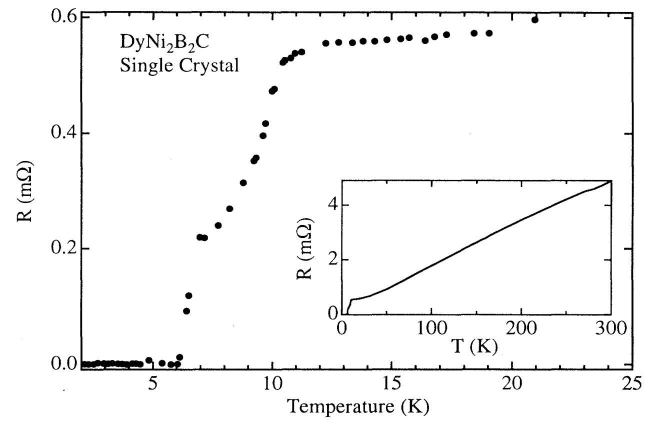

FIG. 3. Resistance versus temperature curve for single-crystal $DyNi_2B_2C$ showing the superconducting transition at 6.4 K. The knee seen at ~10.5 K is associated with the onset of antiferromagnetic order. The inset shows the full curve between 1.2 and 300 K.

been observed previously in the high-T_c cuprates for small applied fields.[11] This was attributed to spontaneous orbital currents and the weak-link Josephson junctions present in the high-T_c materials. The origin of the paramagnetic Meissner effect seen here for applied fields of up to 10 Oe is more likely to be due to the presence of spin magnetization and is currently the subject of further investigation. The dc magnetic susceptibility for the same crystal of $DyNi_2B_2C$ between 8 and 20 K measured in an applied field of 100 Oe is shown in Fig. 2. The anomaly at ≈ 10.5 K in susceptibility is associated with the magnetic ordering of the Dy moments.

The results of the resistance measurements are shown in Fig. 3. The crystals show a sharp drop in resistivity at ~10.5 K, corresponding to the antiferromagnetic ordering temperature, T_N. This knee is present for all the crystals and is more pronounced for the single crystals than for the arc-melted samples.[8] All the crystals show a sharp superconducting transition, but the T_c varies slightly in different crystals, ranging from 6.0 to 6.4 K. This may be due to a slight variation in the carbon stoichiometry which is known to change the superconducting transition in the polycrystalline samples.[8] Application of a magnetic field produces a parallel shift in the resistive transition to lower temperatures with increasing field. The superconducting transition is broadened only slightly by the field, with a ΔT_c of 0.6 K in a field of 0.5 T. For an applied field of 0.6 T, T_c shifts below 2 K, for both $H \| c$ and $H \perp c$. However, the application of a magnetic field has little or no effect on the position of the knee at 10.5 K. The temperature dependence of the upper critical field (H_{c2}) determined from these measurements is shown in Fig. 4. H_{c2} was determined from the onset of a resistive signal at the foot of the transition using an electric field criterion of 2 μV/cm. Using a

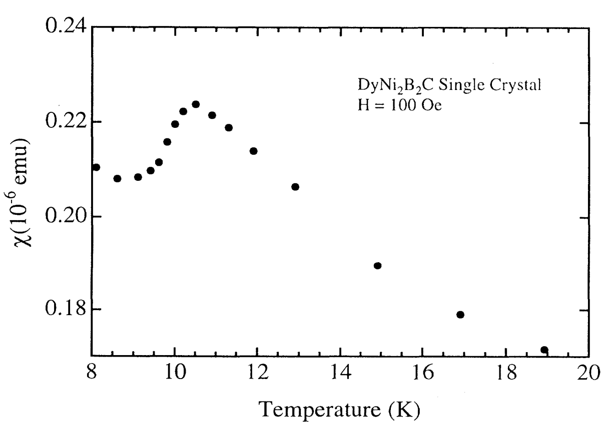

FIG. 2. Temperature dependence of the magnetic susceptibility for $DyNi_2B_2C$ single crystal, measured in an applied field of 100 Oe parallel to the c axis.

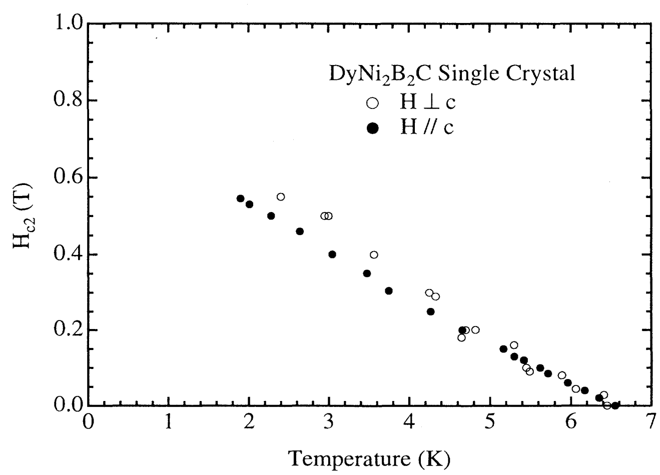

FIG. 4. The upper critical field H_{c2} as a function of temperature for a $DyNi_2B_2C$ single crystal for an applied field H both parallel and perpendicular to the c axis of the crystal.

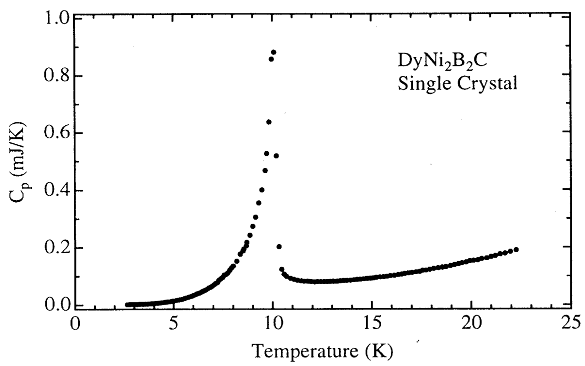

FIG. 5. Specific-heat data for a single-crystal specimen of $DyNi_2B_2C$. The peak at 10.2 K corresponds to the antiferromagnetic ordering of the Dy moments. Within the resolution of our measurement, there is no observable discontinuity at T_c.

different criterion, e.g., the temperature of the midpoint or the onset of the transition, does not alter the results significantly because of the parallel shift of the transition with field. Our measurements show that there is little or no anisotropy in $H_{c2}(T)$ for $DyNi_2B_2C$. It can be seen that H_{c2} is a nearly linear function of temperature near T_c with a slope $(dH_{c2}/dT)_{T=T_c}=-0.1$ T/K for $H\|c$ and $H\perp c$. Extrapolating the H_{c2} versus temperature curve to 0 K, we estimate $H_{c2}(0)$ to be 0.7 T. This gives a coherence length $\xi(0)$ for $DyNi_2B_2C$ of 220 Å. The upper critical field values are comparable with those of other RNi$_2$B$_2$C samples containing magnetic rare-earth ions [Er, $H_{c2}(0)\sim1.9$ T; Ho, $H_{c2}(0)\sim0.4$ T (Ref. 12)] and very different from the RNi$_2$B$_2$C materials containing nonmagnetic ions [Y, $H_{c2}(0)\sim9$ T; Lu, $H_{c2}(0)\sim9$ T (Refs. 12 and 13)]. H_{c2} is strongly suppressed by the presence of magnetic ordering in $DyNi_2B_2C$ and this underlines the interplay between magnetism and superconductivity in these materials.

Figure 5 shows the heat capacity of a single crystal of $DyNi_2B_2C$, which showed a T_c of 6.4 K from susceptibility and resistance measurements. The peak in the heat capacity at 10.2 K corresponds to the long-range antiferromagnetic ordering of the Dy moments and confirms the bulk nature of the transition. No feature associated with the superconducting transition could be resolved in our measurements either in the crystal or in arc-melted ingots. Any jump at the superconducting transition temperature, 6.4 K, would be swamped by the low-temperature tail of the magnetic transition. The heat capacity of YNi_2B_2C shows a large jump $(\Delta C/C_{total})$ at T_c of 40% with a γ of 18 mJ mol^{-1} K^{-2} and $TmNi_2B_2C$ shows a resolvable discontinuity at T_c of about 10%, with a γ of 18 mJ mol^{-1} mol^{-1} K^{-2}.[7] $HoNi_2B_2C$, however, does not show any discontinuity at T_c.[14] In the case of YNi_2B_2C, the jump at T_c is easily measurable as a discontinuity over the phonon background, whereas the discontinuity observed at T_c in $TmNi_2B_2C$ becomes smaller because of the large Schottky contribution in this temperature region.[7] The Schottky contribution in the case of $DyNi_2B_2C$ is also expected to be large and the fact that the superconducting transition occurs in a magnetically ordered state and only a few degrees kelvin below T_N makes it difficult to resolve any discontinuity at T_c. The resolution of our data on $DyNi_2B_2C$ suggests an upper limit of $\Delta C/C_{total}\sim2\%$ for the jump at T_c, if present, and assuming a BCS ratio $(\Delta C/\gamma T_c)$ of 1.43, this would limit the electronic coefficient γ to less than 8 mJ mol^{-1} K^{-2} for $DyNi_2B_2C$. Further analysis of the specific-heat data is in progress.

IV. CONCLUSIONS

Superconductivity has been observed in single crystals of $DyNi_2B_2C$ corroborating our previous results on arc-melted polycrystalline ingots. The crystals exhibit a superconducting transition at 6.4 K, preceded by antiferromagnetic order at ~10.5 K, making this compound a unique member of the borocarbide superconductors. Specific-heat measurements on single-crystal $DyNi_2B_2C$ have clearly established the bulk nature of the antiferromagnetic order at 10.2 K. The $H_{c2}(T)$ shows little or no anisotropy. $H_{c2}(0)$ is 0.7 T for H parallel and perpendicular to the c axis of the crystal. The anomalous field-cooled magnetization exhibited by these crystals in low fields is the subject of further investigation.

Note added in proof. A very recent publication[15] has also reported superconductivity and magnetism in single crystals of $DyNi_2B_2C$, which is in good agreement with the results presented here.

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