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Large hysteresis effect and reentrant behavior in $DyNi_2B_2C$ at temperatures $T<2$ K

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Detailed measurements of resistivity ρ_{ab} as a function of temperature and magnetic field have been carried out on $DyNi_2B_2C$ single crystals. For $H\|[100]$, an anomalously large hysteresis effect of superconductivity and a reentrant behavior were observed in the low temperature range ($T<2$ K). This hysteresis effect is anisotropic in the a-b plane with a maximum value for $H\|[100]$, and almost zero for $H\|[110]$. The reentrant behavior and the large hysteresis of superconductivity indicate a strong interplay between magnetism and superconductivity in $DyNi_2B_2C$. [S0163-1829(98)50114-2]

The recently discovered RNi_2B_2C ($R=$Y and lanthanide)[1,2] compounds have attracted a great deal of attention and have been regarded as ideal materials for studies of the interplay between long-range magnetic order and superconductivity because of their high magnetic ordering temperatures and comparable magnetic and superconducting condensation energies.[3,4] For $R=$Tm,[5] Er,[6] Ho,[7,8] and Dy,[9,10] superconductivity coexists with antiferromagnetism with $T_C\approx10.8$, 10.5, 8.5, and 6.2 K and $T_N\approx1.5$, 5.8, 6.0, and 10.3 K, respectively. $DyNi_2B_2C$ is the unique member of the RNi_2B_2C system with $T_N>T_C$. At temperatures $T<T_N$, the magnetic moments of Dy are ordered ferromagnetically in each Dy-C layer with moments aligned along the [110] direction, and the moments in each two neighboring Dy-C layers are aligned in opposite directions.[4,11] Large anisotropy of magnetic and superconducting properties was observed in $DyNi_2B_2C$ single crystals.[9,10,12] In contrast to the studies on superconducting $HoNi_2B_2C$,[7,8,13] $ErNi_2B_2C$,[13,6] and $TmNi_2B_2C$,[13,5] no reentrant behavior or other anomalous features were observed in previous studies on $DyNi_2B_2C$ single crystals, indicating a perfect coexistence of antiferromagnetism and superconductivity. However, all earlier studies on $DyNi_2B_2C$ were carried out at temperatures $T>1.8$ K. In this paper, we report the observation of a large hysteresis effect of superconductivity in the low-temperature range from 50 mK to 2 K, with magnetic field applied along the crystallographic [100] axis of $DyNi_2B_2C$ single crystal.

Single crystals of $DyNi_2B_2C$ were grown using flux method with Ni_2B as flux.[14] Resistivity and susceptibility measurements as well as the x-ray Laue backscattering method were used for sample characterization. A standard ac four-probe method was adopted for the measurement of resistivity ρ_{ab} with a modulation current of 1 mA flowing in the a-b plane at 117 Hz. The dimension of the crystal for the measurements is about $0.6\times0.5\times0.06$ mm^3. The sample was oriented using the x-ray Laue backscattering method. Resistivity measurements were performed in an adiabatic demagnetization cryostat at temperatures between 50 mK and 14 K. Magnetic fields up to 5 T can be reached with a superconducting solenoid.

Figure 1 shows the temperature dependence of resistivity $\rho(T)$ under various fixed magnetic fields H applied along the crystallographic [100] axis. For zero magnetic field, the $\rho(T)$ curve exhibits a drop of resistivity at 10.3 K, which is attributed to a phase transition from paramagnetism to antiferromagnetism.[11] A sharp superconducting transition occurs with zero resistivity temperature $T_C^{\rho=0}=6.25$ K and superconducting transition width $\delta T_C\approx0.3$ K, indicating that the sample has a high homogeneity. With increasing field, T_C shifts to lower temperature with slightly broaded δT_C. The resistivity at the temperature of onset-superconducting transition is about 2.0 ± 0.2 $\mu\Omega$ cm. The sample displays a very large residual resistivity ratio (RRR), $\rho(300\text{ K})/\rho(T_C^{onset})=32$, indicating a high degree of perfection.

Figure 2 shows the field dependence of resistivity $\rho(H)$ with H increasing from 0 to 20 kOe and $H\|[100]$. For the curve of $T=4.2$ K, besides the superconducting transition occurring at $H_{C2}\approx2.7$ kOe (defined as the intersection of the straight line drawn through the steepest part of superconducting transition range with the H axis), two other steps can also be observed at $H_{M1}\approx7.0$ kOe and $H_{M2}\approx13$ kOe, respectively. In comparison with the reports on magnetization measurements in $DyNi_2B_2C$,[12,15] it is reasonable to attribute the steps at H_{M1} and H_{M2} to two field-induced metamagnetic

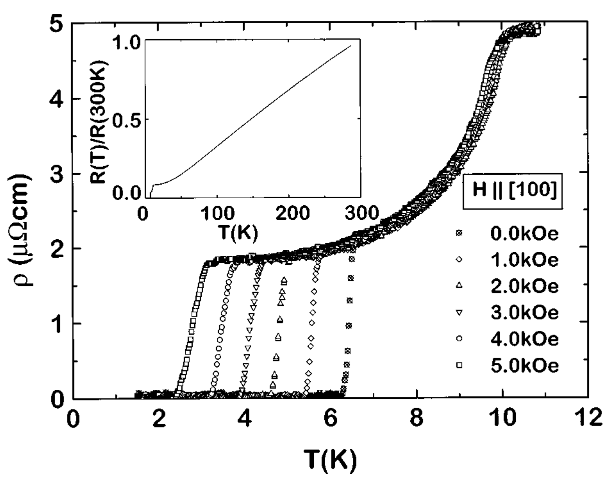

FIG. 1. Temperature dependence of resisitivity ρ_{ab} under various fixed fields applied along the crystallographic [100] axis for the $DyNi_2B_2C$ single crystal. Inset: Temperature dependence of zero-field normalized resistance in the temperature range between 4.2 and 300 K.

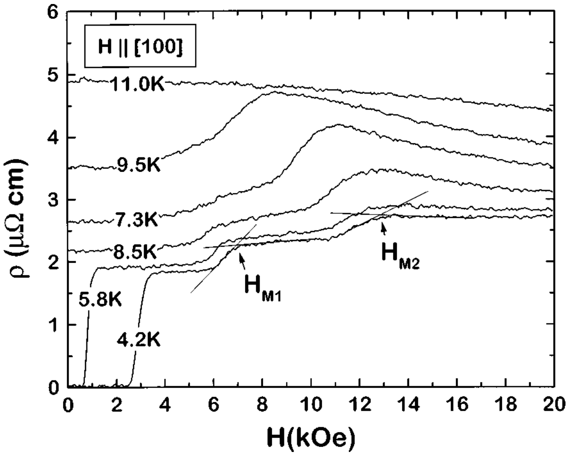

FIG. 2. Resisitivity $\rho_{ab}(H)$ with $H \| [100]$ at various fixed temperatures. Besides the superconducting transition, two field-induced metamagnetic phase transitions at H_{M1} and H_{M2} are also observed.

phase transitions. Both H_{M1} and H_{M2} shift to lower values with increasing temperature. Similar field-induced metamagnetic phase transitions were also observed in other magnetic RNi_2B_2C members.[7,8,16–18,6]

Figure 3 shows the curves of $\rho(H)$ isotherms ($H \| [100]$) at $T=1.7$ K, 0.85 K, and 50 mK, with H increasing from 0 to 20 kOe and then decreasing to zero. For $T=0.85$ K, as H increases, the superconducting transition occurs at $H_{C2} \approx 7.4$ kOe, followed by the metamagnetic phase transition at $H_{M2} \approx 15.5$ kOe. With decreasing field, H_{M2} shifts to a lower value of about 11 kOe, and the upper critical field also becomes smaller with $H_{C2} \approx 4$ kOe, and the shifting of upper critical field $\Delta H_{C2} = H_{C2}$ (field increasing) $-H_{C2}$ (field dreasing)≈ 3.4 kOe. No structure associated with H_{M1} can be resolved in both field-increasing and field-decreasing curves. For $T=50$ mK, an even larger hysteresis effect is observed. The field-increasing part of $\rho(H)$ shows a similar behavior as at 0.85 K, with slightly higher H_{C2} and H_{M2}. With decreasing field, the resistivity decreases continuously to zero at $H_{C2} \approx 1.4$ kOe, no anomalous feature associated with either metamagnetic transition can be clearly resolved above 1.4 kOe. The shifting of the upper critical field $\Delta H_{C2} \approx 6$ kOe, $\Delta H_{C2}/H_{C2}(0) \approx 80\%$. In the curve for $T=1.7$ K, the upper critical field H_{C2} exhibits a modest hysteresis with $\Delta H_{C2} = 0.9$ kOe.

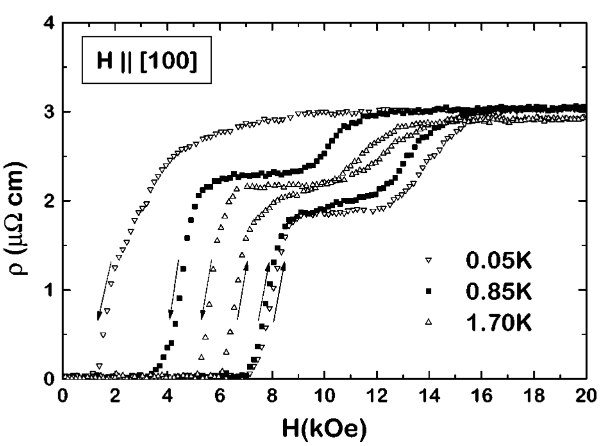

FIG. 3. Field dependence of resisitivity $\rho_{ab}(H)$ with $H \| [100]$ at temperatures $T=50$ mK, 0.85 K, and 1.7 K. Arrows indicate the sweeping direction of the external field.

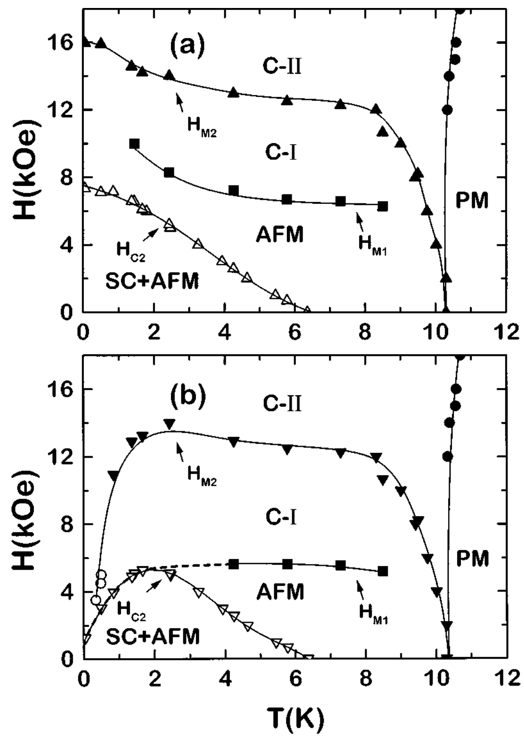

FIG. 4. (a) Phase diagram for $DyNi_2B_2C$ derived from the field-increasing part of $\rho_{ab}(H)$ with $H \| [100]$; (b) phase diagram derived from the field-decreasing part of $\rho_{ab}(H)$ curves. The solid lines drawn through the data points serve as a guide to the eye. The value of $H_{M1}(T)$ at temperatures below 4 K cannot be determined by our resistivity measurements. The dashed line is proposed to be a possible boundary between AFM and C-I (see text for details).

Based on $\rho(T)$ and $\rho(H)$ measurements with $H \| [100]$, the phase diagram is determined. Figures 4(a) and 4(b) show the results derived from the field-increasing and field-decreasing parts of $\rho(H)$ curves, respectively. The magnetic

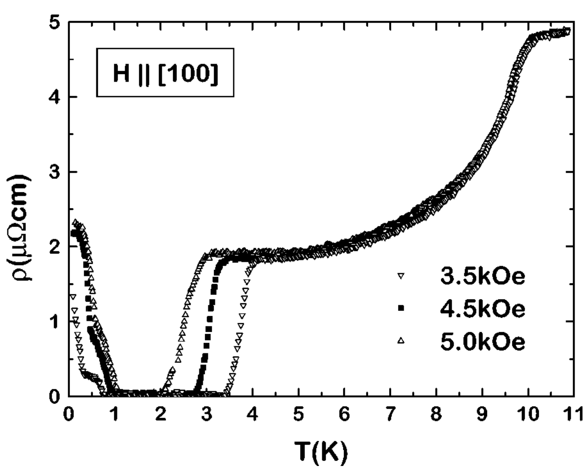

FIG. 5. Measurement of $\rho(T)$ with increasing temperature after a process with H increasing from 0 to 20 kOe at temperature $T=50$ mK, and then lowered to final fields $H_0=3.5$, 4.5, and 5 kOe, respectively.

phase below the H_{M1} curve is antiferromagnetic (AFM), the metamagnetic phase between curve H_{M1} and curve H_{M2} and the phase above curve H_{M2} shall be referred as C-I and C-II, respectively. Their natures of magnetic ordering cannot be determined by our ρ_{ab} measurement. The phase boundary between C-II and paramagnetism (PM) is derived from the data of $\rho(T)$. Under high fields $H>10$ kOe, the $\rho(T)$ curves display a clear kink around 10.5 K, whose position is almost field independent as indicated by the solid circles in Fig. 4. This kink cannot be observed in the case of $H\|[110]$.

In Fig. 4(a), the upper critical field $H_{C2}(T)$ exhibits no local minimum or other features as observed in $HoNi_2B_2C$, $ErNi_2B_2C$, and $TmNi_2B_2C$. Near T_C the curve of $H_{C2}(T)$ displays a positive curvature similar to that observed in other superconducting RNi_2B_2C members.[19]

In Fig. 4(b), the upper critical field $H_{C2}(T)$ derived from the field-decreasing part of $\rho(H)$ curves shows a parabolic-shaped behavior with a maximum of $H_{C2}^{max}\approx 5.3$ kOe at $T=2$ K. Comparing Figs. 4(a) and 4(b), one can see that the hysteresis of superconducting transition and metamagnetic phase transition H_{M2} becomes larger as temperature decreases from 2.0 K to 50 mK. The metamagnetic phase transition H_{M1} cannot be resolved in the field-decreasing part of $\rho(H)$ curves at low temperatures $T<4$ K, so we cannot determine the phase boundary between metamagnetic phase C-I and antiferromagnetic phase (AFM) at temperatures $T<4$ K. Above 2.0 K, H_{C2} and H_{M2} exhibit negligible hysteresis, while H_{M1} keeps a modest hysteresis effect with $\Delta H_{M1}\approx 1.5$ kOe at $T>4.0$ K.

In Fig. 4(b), the upper critical field $H_{C2}(T)$ determined from field-decreasing parts of $\rho(H)$ displays a maximum at $T=2$ K. Such a parabolic-shaped $H_{C2}(T)$ predicts a reentrant behavior. If we first increase H from 0 to 20 kOe at temperature $T=50$ mK, and then lower H down to a field H_0 between $H_{C2}(T=50\text{ mK})$ and H_{C2}^{max}, the sample will remain in the normal state, and afterwards when increasing temperature, the sample shall undergo a superconducting transition. Figure 5 shows the corresponding results with various final fields H_0. For $H_0=4.5$ kOe, with increasing temperature, the sample first undergoes a phase transition from normal state to superconducting state at $T_{C2}\approx 0.95$ K, and then returns to normal state at $T_{C1}\approx 2.8$ K. With lower H_0, T_{C1} becomes larger while T_{C2} becomes smaller. In addition, there exists a kink around 0.5 K in these $\rho(T)$ curves. As shown in Fig. 4(b), the open circles corresponding to the kink positions of $\rho(T)$ curves in Fig. 5 agree well with the extrapolation of H_{M2} deduced from $\rho(H)$ measurement. Hence we tend to believe that the kink observed in Fig. 5 may be associated with the phase transition from C-II to C-I.

No hysteresis effects can be observed for $H\|[001]$. In the case of $H\|[110]$, the hysteresis effect for superconductivity is almost zero. Detailed results of anisotropic superconducting and magnetic properties will be reported elsewhere.

The anomalous features of large hysteresis of superconductivity and the reentrant behavior indicate that a strong interplay between superconductivity and magnetism exists in $DyNi_2B_2C$. A full interpretation of such a behavior requires detailed measurements of magnetization and neutron diffraction at low temperatures $T<2$ K. Here we note that the hysteresis of metamagnetic phase transition from C-I to AFM at temperatures $T<2$ K might play an important role in determining the unusual behavior of superconductivity. Unfortunately the metamagnetic phase transition H_{M1} cannot be resolved in the field-decreasing part of $\rho(H)$ curves at low temperatures $T<4$ K. However, according to the magnetization measurement on the $DyNi_2B_2C$ single crystal at $T=1.8$ K reported by Tomy *et al.*,[12] large hysteresis of a similarly field-induced phase transition from AFM to an intermediate metamagnetic phase was observed with $\Delta H_{M1}\approx 5$ kOe. In comparison with the behavior of H_{M2} and H_{C2}, it is expected that the hysteresis effect of H_{M1} will also become larger with decreasing temperature. Due to this expected large hysteresis of H_{M1}, when decreasing field, the metamagnetic phase C-I possibly extends to lower-field superconducting range. The metamagnetic phase C-I possesses a ferromagnetic component with a value of about $3.5\mu_B$,[12,15] and this strongly suppresses the superconductivity, leading to the lowering of H_{C2} in the low temperature range $T<2$ K as shown in Fig. 4(b). Comparing the result of $M(H)$ reported by C. V. Tomy *et al.*[12] with our $\rho(H)$ measurement in a similar temperature range, we note that in their field-decreasing part of the $M(H)$ curve at $T=1.8$ K, the metamagnetic transition occurs at $H_{M1}\approx 5$ kOe, which is nearly the same as the upper critical field $H_{C2}=5.2\pm 0.2$ kOe determined from the field-decreasing part of $\rho(H)$ at 1.7 K, as shown in Fig. 2(b). Therefore one possibility is that the phase boundary between C-I and AFM in the low temperature range $T<2$ K is identical with that between the normal and the superconducting states, as indicated by dashed line in Fig. 4. With decreasing field, when the phase transition from C-I to AFM occurs at H_{M1}, the suppression of superconductivity by the metamagnetic phase C-I is also reduced, and the superconducting transition might occur at the same field. It is particularly interesting that the coexistence of superconductivity with the metamagnetic phase C-I can also not be ruled out by our resistivity measurements. In RNi_2B_2C, only $ErNi_2B_2C$ exhibits a possible coexistence of superconductivity and a weak-ferromagnetic phase (WFM) with a ferromagnetic component of $0.33\mu_B$ (Refs. 17 and 6) which is about ten times smaller than the ferromagnetic component of the metamagnetic phase C-I observed in $DyNi_2B_2C$. Detailed investigations by magnetization measurement at low temperatures $T<2$ K are in progress.

In summary, we have carried out detailed resistivity measurements as a function of temperature and field on $DyNi_2B_2C$ single crystals. The field dependence of resistivity $\rho(H)$ measured at temperatures $T<2$ K with $H\|[100]$ exhibits an anomalously large hysteresis, and the upper critical field $H_{C2}(T)$ derived from the field-decreasing part of the $\rho(H)$ curves displays a maximum at $T\approx 2$ K. We propose that the interaction between superconductivity and the metamagnetic phase (C-I) plays a dominant role in determining the low-temperature ($T<2$ K) superconducting properties. Detailed magnetization and neutron-scattering measurements will be needed for a complete explanation of the origin for such a low-temperature behavior.

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