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Anomalous magnetoresistance of YNi_2B_2C superconducting single crystals

Rambis K Chu†, W K Chu, Quark Chen, Z H Zhang and J H Miller Jr
Department of Physics and Texas Center for Superconductivity, University of Houston, Houston, TX 77204-5932, USA
E-mail: rambis@uh.edu

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Abstract. Measurements are reported on the temperature and field dependence of the resistivity of YNi_2B_2C single crystals ($T_c = 15.3$ K and $H_{c2}(5.5\text{ K}) \sim 4.7$ T), between T_c + 1 K and T_c + 145 K in magnetic fields up to 8 T. The in-plane magnetoresistance (MR), i.e., $\Delta\rho(H, T) = \rho(H, T) - \rho(0, T)$, changes from negative to positive as the temperature decreases. This sign-reversal occurs at $T = 80$ K when $H = 4$ T. A negative MR supports the presence of magnetically related scattering, and could result from suppression of spin fluctuations, often associated with nearly magnetic metals, by an external field. Our results are in quantitative agreement with the predicted dependence of short-range magnetic moment on temperature and applied field.

The discovery of quaternary borocarbide intermetallic compounds RNi_2B_2C (R = rare earth) has aroused considerable excitement due to their high transition temperature compared to most intermetallic superconductors [1, 2] (e.g., $T_c = 16.6$ K for Lu:1221 and 15.5 K for Y:1221). In contrast to the cuprate superconductors, electronic band structure calculations show that Ni 3d bands are the primary contributors to the density of states of RNi_2B_2C compounds, so superconductivity is attributed to the conventional electron–phonon mechanism [3–9]. Meanwhile, most recent studies have focused on the magnetic rare earth compounds, e.g., R = Er, Ho and Dy, because of the appearance of two microscopic ordering states, *viz*., superconductivity and antiferromagnetism below a certain temperature $T(T < T_N < T_c)$, where T_N is the Néel temperature [10–13]. For instance, recent inelastic neutron scattering measurements of polycrystalline and single-crystalline samples report that incommensurate antiferromagnetic phases exist along the (100) direction, with $\boldsymbol{q}_a = 0.585\boldsymbol{a}^*$ and $\boldsymbol{q}_a = 0.553\boldsymbol{a}^*$ for Er:1221 and Ho:1221, respectively [10, 14–16]. A long-wavelength spiral magnetic structure gradually develops below T_c, and the superconductivity is suppressed as the rare earth magnetic moments become increasingly ordered. Then, a sudden lock-in transition to an incommensurate antiferromagnetic structure of alternating ferromagnetic sheets of rare earth atoms occurs, accompanied by reentrant superconductivity. However, neutron powder diffraction [16], NMR and magnetic-susceptibility measurements on single-crystalline samples [17, 18] have found no observable magnetic moments or antiferromagnetic spin correlations in the Ni sublattice in Y:1221 samples. For the paramagnetic Y:1221 compound, which possesses the second highest T_c in single-phase form, the possible existence of short-range magnetic correlations and their potential effects on transport properties still remain to be settled. In this

† Corresponding author.

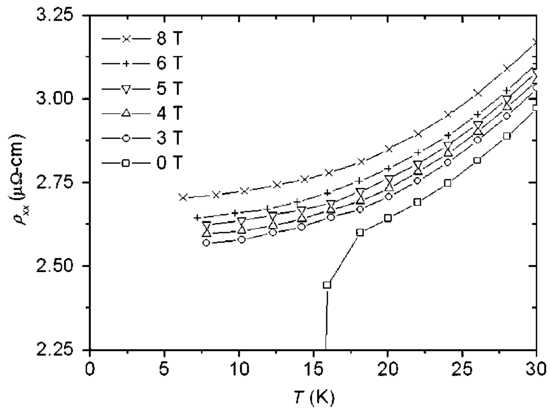
regard, a determination of the carrier scattering response to a magnetic field has profound implications in connection with such correlations. In order to shed light on this issue, we present the results of our systematic normal-state magnetoresistance (MR) study of Y:1221 single crystals. Unexpectedly, we have discovered a sign-change of MR as the temperature decreases. The details of this effect, and its possible explanation, will be discussed.

Studies of magnetoresistance (MR), or the dependence of resistivity on applied magnetic field, $\Delta\rho(H,T) = \rho(H,T) - \rho(0,T)$, can be a powerful tool to probe transport properties of disordered metals or alloys [19]. Some of the carriers' characteristic times, such as the relaxation rates of spin–orbit and inelastic scattering arising from electron–electron correlations, can be extracted first hand, without requiring any additional fitting parameters. In the nearly magnetic or strongly paramagnetic metals, a negative MR is induced by an external magnetic field, which suppresses the amplitudes of short-range spin fluctuations that act as scattering centres for the conducting electrons at low temperatures. In addition, negative MR can occur in the spin-disordered metals due to a reduction of the s–d scattering rate by a magnetic field. In fact, a negative longitudinal MR has been observed in Ho:1221 single crystals [20]. This suggests that spin-disordered scattering by Ho ions could be significant. In this work, we present the results of transverse MR measurements of YNi_2B_2C single crystals ($H \perp I \parallel a$-axis), at temperatures ranging from T_c+1 K to T_c+145 K. An unusual sign-change from negative to positive at 80 K has been observed for the first time. Our observation supports the hypothesis that YNi_2B_2C is a spin-fluctuation metal, as suggested by a recent NMR study [21]. Above the characteristic spin-fluctuation temperature, $T_{sf} \sim 80$ K, the observed negative MR can be understood in terms of scattering from localized magnetic moments. Below T_{sf}, the observed positive MR is plausible due to the topology of the Fermi surface.

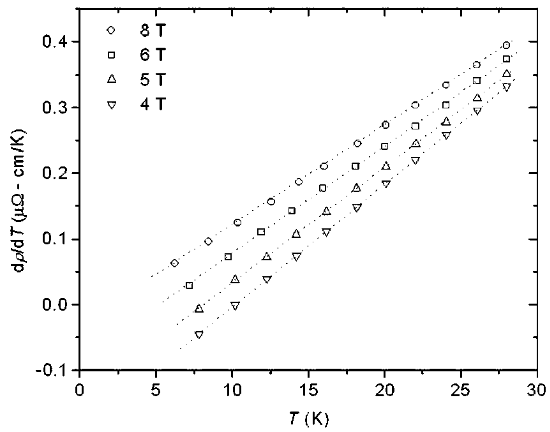
The samples used in these measurements were synthesized using the floating-zone method, with arc-melted polycrystalline stoichiometric ingots as seeds. Details of this method are described elsewhere [22]. The shiny, as-grown boule is about 80 mm in length and 60 mm in diameter, with the (100) direction along its symmetric axis. Rectangular prism a-oriented crystals were carefully trimmed out. Typically, their dimensions were $3 \times 0.5 \times 0.025$ mm^3. X-ray diffraction measurements were carried out in order to verify their orientations, and in all cases, the a-axis was within 1° of the normal to the longest dimension. Structural perfection was determined by ion channelling at room temperature. Its minimum yield, $\chi_{min} \sim 3\%$, demonstrated the high quality of the specimen.

The sample selected for the MR measurements was suitably oriented to within $\sim 0.1^\circ$ of its ab-plane being perpendicular to the field direction. A uniform transverse field, up to 8 T, was generated by a split-coil superconducting magnet. The temperature of the specimen was controlled between 2 K and 300 K with an accuracy of ± 0.01 K. The field strength was monitored by a Hall probe located below the sample holder. The platelet was attached to a thermometer/heater sapphire platform with a small amount of Apiezon grease, and copper wires with a diameter of 0.003 inches were attached to the platelet in a four-terminal configuration using Epotek H20E silver epoxy. The contact resistance was $\sim 1\ \Omega$. Measurements were performed with a dc current bias $I_a = 50$ mA generated by a Keithley 220 programmable current source, and both polarities were used to subtract the offsets in the contacts and the pre-amplifiers. Each data point was measured at least twice with a HP34420A digital nanovoltmeter and the readings provided eight steady digits of accuracy.

Figure 1(a) summarizes the MR data for $H = 0$, 3, 4, 5, 6 and 8 T from 5 to 30 K. In this temperature range, the in-field resistivity is similar to the prediction for a Fermi liquid, with the relation $\rho(H,T) = \rho_0(0) + \rho_0(H) + aT^2$, where the aT^2 term accounts for electron–electron correlations with a $\sim 10^{-11}\ \Omega$ cm K^{-2}. The parameters $\rho_0(0)$ and $\rho_0(H)$ are the zero-field and field-dependent residual resistivity, respectively. Matthiessen's rule of additivity is violated



(a)



(b)

Figure 1. (a) The temperature-dependent resistivity (in $\mu\Omega$ cm) plot between 5 and 30 K in fixed fields $H = 0$, 3, 4, 5, 6 and 8 T. (b) $\mathrm{d}\rho/\mathrm{d}T$ against T with a fit assuming $\rho = \rho_0 + aT^2$ for $H = 4$, 5, 6 and 8 T. The coefficient a is of the order of 10^{-11} Ω cm K^{-2}.

as we extrapolate to the zero-temperature limit, suggesting that the extra scattering is, in fact, magnetic. Figure 1(b) shows the low-temperature $\mathrm{d}\rho/\mathrm{d}T$ fit from 5 to 30 K. Only the high-field data have been considered in order to minimize the possible contributions of superconducting fluctuations. We found the T^2 coefficient to be 0.1, 0.075, 0.05 and 0.025 $\mu\Omega$ cm K^{-1} for $H = 4$, 5, 6 and 8 T respectively. We have further performed a series of MR measurements at various temperatures (i.e. $T = 16$, 20, 40, 60, 80, 120 and 160 K), with the applied field being swept from 0 to 8 T. The results are shown in figure 2. A couple of galvanomagnetic features can be inferred. (i) In the high-field limit, or $\omega_c\tau \gg 1$, YNi_2B_2C is a compensated metal, which means that the total number of electrons is equal to the total number of holes, or the MR would most likely saturate, at least at low temperatures. (ii) Not all the cyclotron orbits are closed, or the MR would obey Kohler's rule or $\Delta\rho$ would be proportional to H^2 [23, 24]. At low temperatures (i.e. $T = 16$, 20 and 40 K), $\Delta\rho$ is positive and increases monotonically with respect to H, as commonly encountered when the magnetoresistance arises from geometric effects on the Fermi surface. At $T = 60$ K, a discernible negative-to-positive crossover is observed around 1.2 T, where $\Delta\rho$ is zero. As the temperature increases, the region of negative

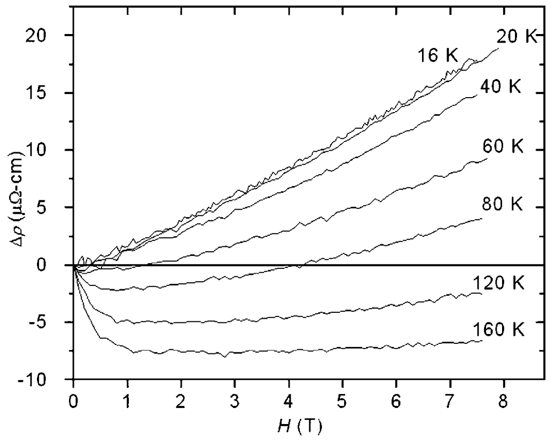
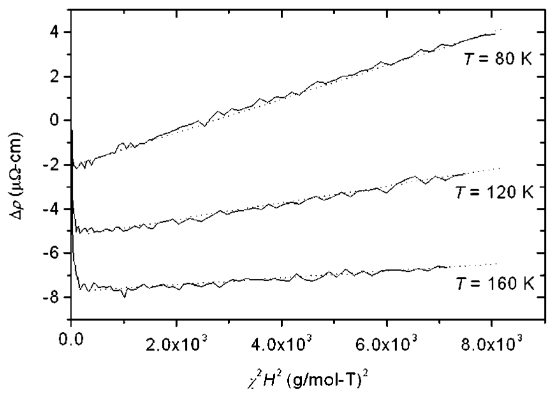


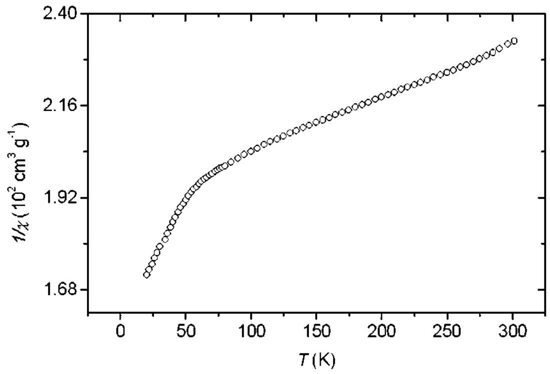
Figure 2. $\Delta\rho = \rho(H, T) - \rho(0, T)$ is plotted as a function of applied magnetic field for all temperatures. Note that beyond the low-field range, the gradients are decreasing as the temperature rises.

MR extends to a much higher field. For $T > 60$ K, the general features of MR consist of a small low-field region ($0 \leqslant H \leqslant 0.75$ T) with a negative slope and a large high-field region ($H \geqslant 0.75$ T) with a positive slope. The minimum dip is more pronounced at $T = 80$ K, and negative-to-positive crossover is now shifted to $H = 4$ T. As the temperature continues to rise ($T = 120$ and 160 K), the MR becomes negative within our measurement range, and $\Delta\rho(T = 120\text{ K})$ is greater than $\Delta\rho(T = 160\text{ K})$. In addition, the MR sharply decreases in the low-field region, and it eventually flattens to a small positive slope in the high-field region. For instance, at $T = 160$ K, the minimum virtually does not exist, and the slope is steep within the low-field region and is approximately equal to $-7.5\ \mu\Omega$ cm in the high-field region. As far as the origin of magnetoresistance is concerned, the s–d scattering typically found in ferromagnetic metals and alloys cannot account for the sign-reversal and negative MR at high temperatures. The following explanations are more plausible. A short-range magnetic transition exists and the 'ordering state' instead exists at high temperatures, or the thermal energy is of the same order of magnitude as, or even less than, this magnetic energy. In the following, we shall quantitatively discuss its origin in some detail.

It is well known that short-range magnetic correlations can occur in nearly magnetic metals (strongly paramagnetic metals), and this correlation is believed to cause local susceptibility enhancements and localized spin fluctuations [25]. It has also been pointed out that these enhancements and fluctuation effects only exist over a mean lifetime τ_{sf}, which is the time of the spin memory. This spin fluctuation lifetime may be related to thermal fluctuations, and gives a characteristic spin fluctuation temperature, $T_{sf} = h/k_B\tau_{sf}$, where k_B is Boltzmann's constant [26]. The localized spins fluctuate rapidly compared to their thermal motion below T_{sf}, and therefore the material macroscopically appears to be non-magnetic. However, above T_{sf}, the spin polarizations may persist longer than the thermally induced spatial fluctuations of the moment. If this is the case, the magnetic behaviour of the material is indistinguishable from that of a localized magnetic moment. Considering the localized spin fluctuations from the narrow d-band of the Ni atoms in the YNi_2B_2C matrix, the observed negative in-plane magnetoresistance above $T_{sf} = 80$ K (from them, $\tau_{sf}{\sim}10^{-14}$ s, which is of the same order as that of elemental ferromagnets in non-magnetic hosts [27]) can be analysed in terms of local moment theory [28]. The mean-field approximation is applied in this theory, and one



(a)



(b)

Figure 3. (a) Magnetoresistance $\Delta\rho$ versus $(\chi H)^2$ for $T = 80$, 120 and 160 K. (b) dc magnetic susceptibility χ versus temperature T for Y:1221 samples.

can separate the field-dependent resistivity $\rho(H, T)$ into two components: $\rho \uparrow \propto 1 + cM$ and $\rho \downarrow \propto 1 - cM$ depending on whether the spin of each scattered conduction electron is parallel or antiparallel to the spin of the local moment. Then the resistivity is additive if parallel, and $1/\rho(H, T) = 1/\rho \uparrow + 1/\rho \downarrow$ so the magnetoresistance is

$$\Delta\rho \equiv \rho(H, T) - \rho(0, T) \sim -M^2 = -(\chi H)^2 \quad (1)$$

where M is the magnetization. Therefore, $\Delta\rho$ is a quantity which would be proportional to the susceptibility of localized spins, which also depend on $1/T$. Figure 3(a) shows that the in-plane transverse magnetoresistance is proportional to the square of the magnetization at $T = 80$, 120 and 160 K by using χ from the temperature-dependent dc magnetic susceptibility measurements shown in figure 3(b). The magnetoresistance $\Delta\rho$ and the differential magnetoresistance coefficient $\mathrm{d}(\Delta\rho)/\mathrm{d}M^2$ seem to decrease as the temperature increases. This can be understood as follows: when T is larger than T_{sf}, the thermally induced spatial fluctuations become dominant, and the spin polarization lifetime becomes longer. Therefore, the conduction electrons are subjected to less scattering out of the localized moments. As the field continues to increase, the classical Lorentz force acts on the conduction electrons contributing positively in the form of Kohler's rule $\Delta\rho/\rho_0 = F(H/\rho_0)$, where F is a function that depends on the nature of the metal itself. In fact, it is reasonable to assume that the MR consists of a negative correlation component and a positive normal component, which

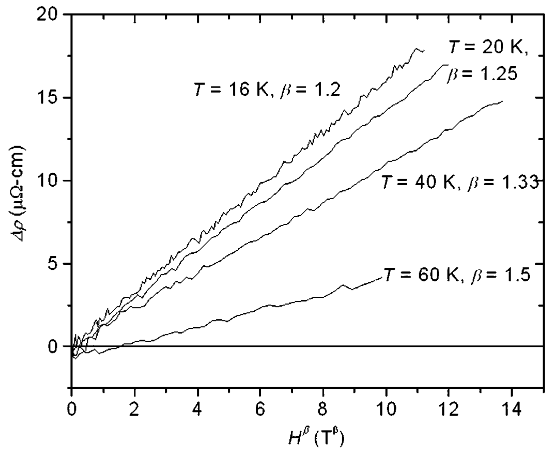


Figure 4. $\Delta\rho$ variation of H^β at low temperatures scales with different values of β.

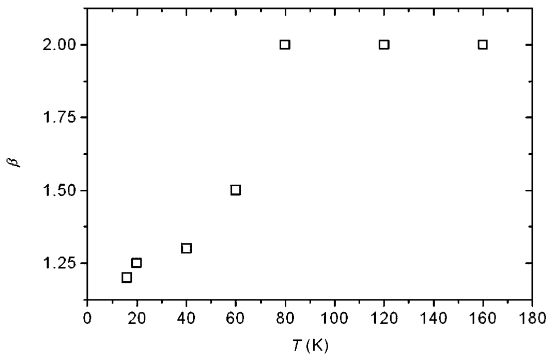


Figure 5. The β exponent as a function of T (i.e. $T = 16$, 20, 40, 60, 80, 120 and 160 K): a phase transition appears between 60 and 80 K.

complement each other as functions of H and T. In figure 4, we show the magnetoresistance below T_{sf} scales with H^β where $\beta = 1.2$, 1.25, 1.33 and 1.5 for $T = 16$, 20, 40 and 60 K, respectively. As far as this explanation is concerned, if the number of open cyclotron orbits on the Fermi surface is small, the field dependence may exhibit a power law somewhere between linear and quadratic. Effects of this sort were first observed experimentally on the high-field magnetoresistance of metals [29, 30]. Figure 5 shows the β exponent as a function of T, where a phase transition can be recognized from 60 to 80 K. It is apparent that the material may undergo a transition from a non-magnetic phase to one characterized by the formation of short-range localized moments, as described above in terms of the spin-fluctuation scenario. In addition, it is worth pointing out that a similar negative MR has been observed in a number of metallic glasses and amorphous alloys. Its existence is mainly due to weak localization (WL), which is caused by long-range quantum interference of a carrier scattered by defects in the system at liquid helium temperature range. As the temperature increases, the phase-coherence length decreases and WL is suppressed. Therefore, we can most likely rule out this possibility in our case.

In recapitulation, detailed measurements of $\Delta\rho$ ($0 < H < 8$ T, $16 < T < 160$ K) have been performed on YNi_2B_2C crystals for applied fields parallel to the a-axis.

A striking temperature-dependent MR, including a sign-reversal at a possible spin-fluctuation temperature, $T_{sf} = 80$ K, has been observed. Below 80 K, the spins fluctuate rapidly compared to their thermal motion and the material appears to be non-magnetic, and $\Delta\rho$ is positive. Above 80 K, the thermal fluctuations are rapid so that the spin polarizations appear to survive long enough to exhibit local moments, and $\Delta\rho$ is negative. Despite the fact that YNi_2B_2C is considered non-magnetic, $\Delta\rho$ reveals its dependence on the magnetization above T_{sf}. This dependence is consistent with the formation of localized moments, possibly from Ni ions. This further implies that the 3d electrons are not totally quenched, as thought originally, and they are both spatially and temporally correlated. Furthermore, the temperature dependence of the MR sign-change infers that YNi_2B_2C is a spin-fluctuation superconductor, in which 3d electronic correlations in the presence of a magnetic field play a significant role.

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