Transport and superconducting properties of $R\mathrm{Ni}_2\mathrm{B}_2\mathrm{C}$ (R=Y, Lu) single crystals

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The in-plane resistivity, in-plane absolute thermopower, and upper critical field measurements are reported for single-crystal samples of $\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ and $\mathrm{LuNi}_2\mathrm{B}_2\mathrm{C}$ superconductors. The in-plane resistivity shows metallic behavior and varies approximately linearly with temperature near room temperature (RT) but shows nearly quadratic behavior in temperature at low temperatures. The $\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ and $\mathrm{LuNi}_2\mathrm{B}_2\mathrm{C}$ single-crystal samples exhibit large transverse magnetoresistance ($\approx 6-8$ % at 45 kOe) in the ab plane. The absolute thermopower $S(T)$ is negative from RT to the superconducting transition temperature T_c. Its magnitude at RT is a few times of the value for a typical good metal. $S(T)$ is approximately linear in temperature between $\approx$150 K and RT. Extrapolation to $T=0$ gives large intercepts (few μV/K) for both samples suggesting the presence of a much larger ''knee'' than would be expected from electron-phonon interaction renormalization effects. The upper critical fields for H parallel and perpendicular to the c axis and the superconducting parameters derived from it do not show any anisotropy for the $\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ single-crystal samples in agreement with magnetization and torque magnetometry measurements, but a small anisotropy is observed for the $\mathrm{LuNi}_2\mathrm{B}_2\mathrm{C}$ single crystals. The analysis shows that these are moderately strong-coupling type-II superconductors (similar to the A-15 compounds) with a value of the electron-phonon coupling parameter $\lambda(0)$ approximately equal to 1.2 for $\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ and 1.0 for $\mathrm{LuNi}_2\mathrm{B}_2\mathrm{C}$, the Ginzburg-Landau coherence length $\xi(0)$ approximately equal to 70 Å, and $H_{c2}(0)\sim 60-70$ kOe. The temperature dependence of the upper critical field shows a positive curvature near T_c in disagreement with the Werthamer, Helfand, Hohenberg, and Maki (WHHM) theory but in agreement with a recent solution of the Gor'kov equation using a basis formed by Landau levels (Bahcall); however, the data show a severe disagreement between the observed low-temperature behavior of $H_{c2}(T)$ and that predicted either by WHHM or Bahcall's expressions. [S0163-1829(97)06413-8]

INTRODUCTION

The latest discovery of superconductivity in intermetallic borocarbide compounds, namely, $\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ (Refs. 1 and 2) (T_c=15.6 K), multiphase Y-Pd-B-C (Ref. 2) (superconducting transition temperature, T_c=23.2 K), $R\mathrm{Ni}_2\mathrm{B}_2\mathrm{C}$ (Refs. 3–6) (Lu-Gd; T_c=16.6 K for Lu) has had enormous impact on superconductivity research during the last two years and has led to renewed interest in the search for high-temperature superconductivity in multielement intermetallic compounds and possibly exotic superconductivity mechanism(s) leading to T_c higher than 10 K. T_c values of these borocarbides are similar to those of the other well-known high-T_c intermetallic A-15 superconductors.[7] Two fascinating features of $R\mathrm{Ni}_2\mathrm{B}_2\mathrm{C}$ compounds are that they contain a large amount of nickel, a ferromagnetic metal usually detrimental to superconductivity, and that superconductivity is observed not only for the nonmagnetic rare-earth elements but also for the heavy magnetic rare earths like Tm, Er, Ho, and Dy having a large saturation magnetic moment. They exhibit a spectrum of very interesting physical properties depending upon the R atom; compounds with R=Y, Lu seem to be isotropic,[8] BCS-type[9] superconductors with relatively high T_c; R=Yb belongs to the heavy fermion system[10,11] and is not superconducting down to 0.34 K;[11] R=Tm-Dy exhibit the coexistence of superconductivity and magnetic order (generally antiferromagnetic) with additional effects due to anisotropy induced by crystalline electric fields.[5,12–17] Compounds with R=Tb and Gd are not superconducting at least above 0.5 and 1.4 K, respectively.[18–22]

The structure of these compounds is body-centered-tetragonal (space group $I4/mmm$) with alternating square-planar layers of rare-earth carbides and corrugated $\mathrm{Ni}_2\mathrm{B}_2$ sheets with a unit cell consisting of two formula units.[23] This is the same as the $\mathrm{ThCr}_2\mathrm{Si}_2$ structure with an additional carbon atom per rare-earth atom in the rare-earth layer. The structure has Ni-B sheets running parallel to the basal plane and seems to be highly anisotropic from the view point of the ratio of c/a which is approximately 3 for both $\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ and $\mathrm{LuNi}_2\mathrm{B}_2\mathrm{C}$ compounds since their lattice parameters are a=3.526 Å, c=10.534 Å [$\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ (Ref. 24)] and a=3.464 Å, c=10.631 Å [$\mathrm{LuNi}_2\mathrm{B}_2\mathrm{C}$ (Ref. 23)], respectively. These characteristics are somewhat similar to those of cuprate superconductors although the T_c for the borocarbides is much smaller than that of the cuprates. The electronic structure of transition-metal borides and carbides has generally been characterized by strong covalent bonding between the constituent elements,[25] but band-structure calculations on $\mathrm{LuNi}_2\mathrm{B}_2\mathrm{C}$ (Refs. 26–30) and $\mathrm{YNi}_2\mathrm{B}_2\mathrm{C}$ (Refs. 26, 31) show that these are thoroughly three-dimensional metals belonging to the family of conventional superconductors with a relatively high density of states at the Fermi level E_F. The states near the Fermi level are dominated by the Ni($3d$) character with some contributions from other atoms. Ni($3d$), and

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B($2p$) states hybridize by a very small amount around E_F, but the strength of such a hybridization is not so great as that of Cu-O hybridization within CuO_2 planes in high-T_c cuprates, and bridging carbon atoms provide strong interlayer interactions which lead to the three-dimensional structure.[30] The calculated density of states shows a peak at E_F arising from a relatively dispersionless energy band which is close to E_F.[26] No peak in the density of states at E_F has been observed[32,33] in photoemission studies on polycrystalline YNi_2B_2C, the absence of which has been attributed to the effects of electron correlations due to the presence of nickel, a $3d$-late transition metal, which gives rise to strong electron correlations. One might expect nickel to give rise to possible magnetism in these compounds, but no local magnetic moment on Ni atoms has been inferred either from neutron-diffraction measurements on polycrystalline samples down to 2 K (Ref. 21) or from NMR experiments[34] on single-crystal YNi_2B_2C although earlier NMR studies on polycrystalline YNi_2B_2C (Ref. 35) and muon spin-rotation measurements of the internal field in polycrystalline $TmNi_2B_2C$ (Ref. 36) suggest the existence of dynamically fluctuating moments on Ni atoms. Since Ni-derived $3d$ electrons are considered to be superconducting electrons in these compounds, the possibility of the existence of a localized moment on Ni atoms is not reasonable since it would lead to strong pair breaking if superconductivity is due to s-wave pairing. Boron in these compounds, it is speculated, gives rise to higher phonon frequencies because of its low mass, and consequently a high T_c. Both a band-structure calculation[28] and an isotope experiment[37] seem to confirm that the high-frequency optical phonons associated with B atoms play an important role in the superconductivity of these compounds. The role of carbon is not clear although it is absolutely essential since YNi_4B does not show superconductivity. A recent structural study[38] on polycrystalline YNi_2B_2C reported highly anisotropic thermal vibrations of C atoms and may have some bearing on the high T_c of these compounds.

Since the discovery of these borocarbide superconductors, extensive studies have been done on their superconducting properties and the interplay of superconductivity and magnetism in some of these compounds in the polycrystalline state as well as with single crystals.[39] A number of superconducting studies indicate that these are type-II superconductors with a small coherence length ($\approx$50–100 Å). Theoretical studies[26–31] as well as some experimental reports[24,40,41] indicate that these are moderately strong-coupling superconductors, and the observation of soft phonon modes for R=Lu (Ref. 42) seems to suggest the same, but recent tunneling measurements[9] show them to be weak-coupling BCS type. Many experimental results indicate that these compounds are the conventional phonon-mediated s-wave superconductors although some deviations are reported, namely absence of the coherent peak in the NMR relaxation rate below T_c,[43] T^3 dependence of the specific heat in a wide range of temperatures below T_c,[24] and anomalous non-BCS temperature dependence of H_{c1},$\lambda(T,H=0)$ and microwave impedance.[44]

Each family of superconductors so far discovered, including A-15 compounds, Chevrel compounds, cuprates, and fullerides, possesses characteristic structural and electronic features which have important bearing on the occurrence of superconductivity in them. Normal-state transport properties of superconductors provide important information which is expected to lead to an understanding of superconductivity in them. The recent availability of good single crystals of these borocarbides provides a unique opportunity to investigate the normal-state transport properties in well characterized samples without the granularity problems present in polycrystalline samples which could be detrimental to interpretation of results on transport properties. While there has been feverish activity on studies of the superconducting properties, and the interplay of magnetism and superconductivity in these borocarbides, there are hardly any reports on studies of the normal-state transport properties of these compounds except our preliminary reports[14,39,45] on single-crystal RNi_2B_2C, (R=Y, Lu-Gd), recent brief reports on thermopower[46] of polycrystalline YNi_2B_2C and $LuNi_2B_2C$, and the thermal conductivity of YNi_2B_2C and $HoNi_2B_2C$ single crystals.[47] In this paper we present results of detailed studies on in-plane electrical resistivity and thermopower in single crystals of YNi_2B_2C and $LuNi_2B_2C$ along with their superconducting properties. While YNi_2B_2C is a widely investigated compound, only a few reports have appeared on $LuNi_2B_2C$. These compounds do not show any sign of magnetic order or crystal-field effects which do influence the transport properties of the analogous compounds with other rare earths and make interpretation of their transport properties more difficult. Results are compared with the high-T_c cuprates and A-15 compound superconductors wherever appropriate.

EXPERIMENTAL DETAILS

Single crystals of YNi_2B_2C and $LuNi_2B_2C$ are grown by a Ni_2B flux method.[48] An arc-melted and well-annealed single-phase polycrystalline ingot of YNi_2B_2C ($LuNi_2B_2C$) compound is placed in an alumina crucible with an equal mass of Ni_2B on top of it and is melted in flowing pure argon gas at 1763 K. The melt is cooled to 1473 K at 10 K/h, followed by furnace cooling to room temperature (RT). Single crystals of YNi_2B_2C ($LuNi_2B_2C$) grow into the flux from the original polycrystalline compound. These crystals can be easily removed from the original polycrystalline ingot. The crystals are platelike having mostly irregular shapes with surfaces in the ab plane. The x-ray diffraction of these platelets show that they are single crystals of YNi_2B_2C ($LuNi_2B_2C$) with the c axis perpendicular to the flat surfaces. The as-grown single-crystal platelets usually have somewhat rough surfaces with a metallic shine and luster.

The flux-free single-crystal platelets are cut into rectangular parallelepiped shapes using a wire saw for the resistivity and thermoelectric power measurements. The typical size of these samples is 2.0 mm$\times$1.0 mm$\times$0.3 mm. The crystal surfaces are ground flat and then polished mechanically to remove the surface roughness and make them uniformly thick.

The resistivity is measured using a dc four-probe technique. Two 0.15 mm copper wires are attached to the sample as the current leads using indium solder. The voltage leads consist of two heavy formvar insulated 0.1 mm diameter copper wires which are attached to the sample with Epotek 410E silver epoxy.[49] The maximum error in the absolute resistivity value comes mainly from the uncertainties in the distance between two voltage leads due to the finite width of

the electrical contacts (since the sample length is very small) and the thickness measurement. Considering all the possible errors, the uncertainty in the absolute resistivity is no more than $\pm 10\%$ and much less for relative measurements. The resistance in zero applied magnetic field is measured from RT (300 K) to 4.2 K and in applied magnetic field (maximum 5 T) between 25 and 4.2 K. A double can He-4 cryostat equipped with a 6 T superconducting magnet is used for the measurements. More experimental details can be found in an earlier publication.[14]

Thermopower of the samples is measured using a differential technique with temperature gradient along the ab plane. Samples for thermopower measurements are cut from the same block of a single crystal of YNi_2B_2C ($LuNi_2B_2C$) from which samples for the resistance measurements were cut. The cryostat used for thermopower measurements is similar to the one described earlier[50] but with some improvements in its design and automatic data collection. The differential temperature across the sample is measured using a calibrated 0.075 mm diameter Au 0.07 % Fe/chromel thermocouple. The thermopower of each sample is measured against a 0.1 mm manganin wire precalibrated against a well annealed lead foil. Samples are kept in a 10^{-6} torr dynamic vacuum during the measurements. The absolute accuracy in the thermopower is estimated to be ± 0.1 μV/K.

RESULTS AND DISCUSSION

A. Normal-state resistivity

Room-temperature (RT, 300 K) in-plane resistivity ρ_{ab} of single crystals of YNi_2B_2C and $LuNi_2B_2C$ are measured to be 67.1 and 46.8 $\mu\Omega$ cm, respectively. The lower resistivity of single-crystal $LuNi_2B_2C$ samples is probably due to better growth of the crystal with less imperfections and/or defects. This is also reflected in the residual resistivity ratio (RR) as indicated later. These in-plane resistivity values are of the same order of magnitude as for some of binary alloys of the rare-earth and transition-metal intermetallic compounds/alloys[51] and some A-15 compounds.[7] Figure 1 displays the temperature dependence of ρ_{ab}, to be denoted ρ hereafter, of single-crystal samples of YNi_2B_2C and $LuNi_2B_2C$ from 300 to 4.2 K and in zero applied magnetic field H. The resistivity is clearly seen to be metallic, i.e., ρ decreases with a decrease in temperature. This decrease in ρ is approximately linear with T at higher temperatures (near RT) although a slight curvature towards the T axis is clearly visible pointing to a possibility of saturation of the resistivity at higher temperatures. This curvature is slightly more in the case of the YNi_2B_2C single-crystal (sc) sample. $[d\rho/dT]_{RT}$ values for sc-YNi_2B_2C and sc-$LuNi_2B_2C$ are 0.22 and 0.15 $\mu\Omega$ cm/K, respectively. As the temperature is lowered below $\approx$100 K, the resistivity becomes nonlinear and finally drops abruptly to zero at the superconducting transition temperatures 15.6 and 16.1 K, respectively, for sc-YNi_2B_2C and sc-$LuNi_2B_2C$ samples (to be referred to as Y and Lu samples hereafter for brevity wherever convenient), respectively. The superconducting transition temperature (T_c) value of YNi_2B_2C agrees well with the T_c values reported by others,[3,4,48,52,53] but T_c for $LuNi_2B_2C$ is slightly smaller than that observed by others.[3,40,54–56] The superconducting transition width for either sample is $\approx$0.25 K which is quite sharp, indicating the good quality (homogeneity) of both samples. The good quality of these crystals is further confirmed by a reasonably high resistivity ratio RR, defined as, $\rho(300)/\rho(T_{c,\text{onset}})$ i.e., RR(Y)=18 and RR(Lu)=25, where $\rho(T_{c,\text{onset}})$ of the Y and Lu samples are 3.8 and 1.9 $\mu\Omega$ cm, respectively. The lower RR value of the Y sample implies that the Y sample has more imperfections. The resistivity just above the superconducting transition temperature does not seem to become temperature independent for either sample, exhibiting a weak temperature dependence.

Based on the measured values of ρ(RT) and $[d\rho/dT]_{RT}$ of the Y and Lu samples in this work, and recently reported theoretical band-structure calculations,[27] estimates of the electron mean free path (l) and the transport electron-phonon coupling parameter (λ_{tr}) are made for these materials.

The resistivity can be written as

$$\rho^{-1}=\frac{2}{3}\,e^2N(0)\,\nu_F l, \tag{1}$$

where $N(0)$ is the band quasiparticle density of states at the Fermi level and ν_F is the Fermi velocity. The values of $N(0)$ for $LuNi_2B_2C$ and YNi_2B_2C have been calculated[27,31] and are 4.8 and 4.03 states/(eV unit cell), respectively. The Fermi velocity ν_F also has been calculated[27] to be 3.6×10^7 cm/s for $LuNi_2B_2C$. No such estimate of ν_F is available for YNi_2B_2C. Hence, ν_F of YNi_2B_2C is assumed to be the same as that of $LuNi_2B_2C$ since they have similar band structures. These numbers in Eq. (1) give l(Y)=110 Å and l(Lu)=190 Å at T=16 K, and l(Y)=6.1 Å and l(Lu)=7.6 Å at 300 K. The electron mean free path l at room temperature in these samples is of the order of their atomic spacing, and therefore, the semiclassical Boltzmann theory may not be strictly valid to describe the transport phenomena at room and higher temperatures. In addition, possible anharmonicity and Fermi smearing effects at higher temperature may be responsible for producing some flattening of the resistivity near room temperature. Since l(Y) is smaller than l(Lu) at RT, the relatively larger flattening of the resistivity of YNi_2B_2C near

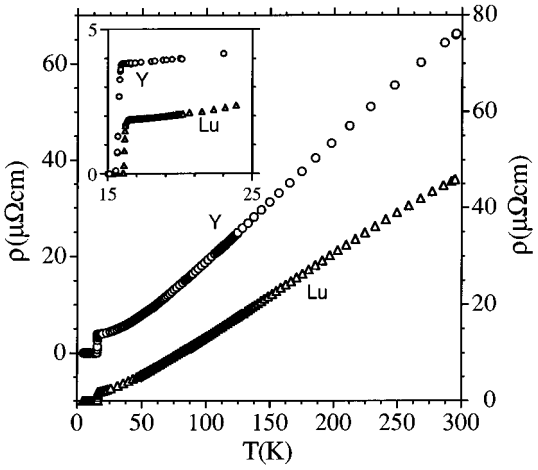

FIG. 1. Temperature dependence of the in-plane resistivity of single-crystal samples of YNi_2B_2C (open circles) and $LuNi_2B_2C$ (open triangles). The inset shows the resistivity variation near T_c.

TABLE I. Estimated values of the electron-phonon coupling parameter λ.

Sample	λ_{tr}	λ ($\mu^*=0.10$)	λ ($\mu^*=0.15$)	λ_γ
YNi_2B_2C	1.2	0.79	0.93	0.97
$LuNi_2B_2C$	0.97[a],0.78[b]	0.98	1.14	0.68[a],1.07[b]

[a]Estimated using measured values of $d\rho/dT$ or γ with the calculated band density of states $N(0)=4.8$ states/eV unit cell from Ref. 27.

[b]Estimated using measured values of $d\rho/dT$ or γ with the calculated band density of states $N(0)=3.88$ states/eV unit cell from Ref. 30.

room temperature in comparison with that of $LuNi_2B_2C$ is understandable. The resistivity flattening effect is truly very small here, and therefore, linearity between ρ and T near RT is essentially valid in this range.

In the Bloch-Grüneisen transport theory the temperature dependence of ρ is related to the electron-phonon coupling constant λ_{tr} by the relation[57]

$$\frac{d\rho}{dT}=\frac{8\pi^2}{\hbar\omega_p^2}k_B\lambda_{tr}, \tag{2}$$

where ω_p is the Drude plasma frequency which is related to $N(0)$ through the relation

$$\hbar^2\omega_p^2=4\pi e^2N(0)\nu_F^2. \tag{3}$$

Equation (2) assumes that residual resistivity and other contributions to ρ are negligibly small compared to the electron-phonon contribution described by the standard Boltzmann transport theory. Further, Eq. (2) is valid for $T>0.7\ \Theta_D$ where Θ_D is the Debye temperature.[57] The reported values of Θ_D for YNi_2B_2C are 489 K (Ref. 52) and 537 K,[24] and ≈350 K for $LuNi_2B_2C$.[40,54] The use of Eq. (2) to calculate λ_{tr} from ρ (RT) and $[d\rho/dT]_{RT}$ values seems to be reasonable for sc-$LuNi_2B_2C$, but it is only approximately correct for sc-YNi_2B_2C since RT (300 K)$<0.7\ \Theta_D$(Y). The plasma frequency of each compound can be calculated using Eq. (3) which gives 5.1 eV for sc-$LuNi_2B_2C$ and 4.7 eV for sc-YNi_2B_2C where $\nu_F=3.6\times10^7$ cm/s is assumed for both samples, and $N(0)=4.8$ and 4.03 states/(eV unit cell) for $LuNi_2B_2C$ (Ref. 27) and YNi_2B_2C,[31] respectively. A substitution of $[d\rho(\mathrm{Y})/dT]_{RT}=0.22\ \mu\Omega$ cm/K and $[d\rho(\mathrm{Lu})/dT]_{RT}=0.15\ \mu\Omega$ cm/K in Eq. (2) yields $\lambda_{tr}(\mathrm{Y})=1.2$ and $\lambda_{tr}(\mathrm{Lu})=0.97$ as listed in Table I. Another recent calculation[30] for Lu gives the value $N(0)=3.88$ states/(eV unit cell) which produces a smaller value for λ_{tr}. These values seem reasonable and represent a semiempirical measure of electron-phonon coupling constant λ which appears in the McMillan equation for the superconducting transition temperature T_c.[58] With the knowledge of T_c and the Debye temperature Θ_D of a superconductor, λ can be estimated from the McMillan equation

$$k_BT_c=\frac{\hbar\omega_{\log}}{1.2}\exp\left[-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right], \tag{4}$$

where $\omega_{\log}$ is taken to be $0.7\ \omega_{ph}$, ω_{ph} is regarded to be the same as the Debye frequency $\omega_D=k_B\Theta_D/\hbar$, and μ^* is Coulomb pseudopotential and usually taken between 0.1 and 0.15. Values of λ determined from Eq. (4) with $\mu^*=0.15$ are given in Table I. Closer agreement with λ_{tr} is found for $\mu^*=0.15$ for Y, but for Lu better agreement is found with $\mu^*=0.10$. It is not clear why this should be since the electronic and phonon structures of the two compounds should be very similar.

The electron phonon coupling parameter can also be estimated from the ratio of $N(0)|_{obs}/N(0)|_{band}=\gamma|_{obs}/\gamma|_{band}=(1+\lambda_\gamma)$, where γ is the specific-heat constant and λ_γ is the electron-phonon mass enhancement parameter which should be similar to λ. Movshovich *et al.*[52] reported $\gamma_{obs}=18.7$ mJ/(mol K^2) for sc-YNi_2B_2C, and the calculated value[31] of γ_{band} for YNi_2B_2C is 9.5 mJ/(mol K^2) with $N(0)$ ≈4.03 states/(eV unit cell). Similarly, γ_{obs} $LuNi_2B_2C$ (Ref. 40)=19 mJ/(mol K^2) and calculated $\gamma_{band}=11.3$ mJ/(mol K^2) using $N(0)=4.8$ states/(eV unit cell). The calculated value $N(0)=3.88$ states/(eV unit cell) for $LuNi_2B_2C$ (Ref. 30) gives $\gamma_{band}=9.15$ mJ/(mol K^2). The corresponding values of λ_γ are listed in Table I. There appears to be a particularly wide spread in the various estimates for Lu, but values for Y are reasonably consistent. A 10–15 % disagreement among the electron-phonon coupling parameters obtained from different procedures is commonly found in other superconductors.[57] In any case, the range of λ_{tr} or λ values are such that these compounds would be considered to be only moderately strong-coupling superconductors. A comparison of these compounds with some *A*-15 superconductors which have T_c near 15 K, i.e., Nb_3Sn (T_c≈17 K), V_3Si (T_c≈15 K) shows that λ values for Y(Lu)Ni_2B_2C superconductors are close to that of V_3Si (λ≈1.0) but much smaller than that of Nb_3Sn (λ≈1.8) or Nb_3Al (λ≈1.5).[7] Strong-coupling elemental superconductors Pb and Nb have λ≈1.2.[57]

A similarity between Y(Lu)Ni_2B_2C and *A*-15 high-T_c superconductors is also found in the temperature dependence of their resistivities at low temperatures, i.e., $1.25T_c<T<0.1\Theta_D$. Below 100 K, the resistivity of either borocarbide compound shows a nonlinearity with temperature and it does not decrease as rapidly as expected from the Bloch-Grüneisen theory. To determine the exact temperature dependence of $\rho(T)$, the low-temperature data was fitted to the expression

$$\rho(T)=\rho_0+AT^p \tag{5}$$

in the temperature interval $1.25T_c<T<0.1\Theta_D$ using a least-squares fit procedure, with the square of the correlation coefficient to determine the goodness of the fit. The temperature region above 20 K was chosen to minimize the superconducting fluctuation effects. Figure 2 shows the plot of ρ vs $T^{2.2}$ for the Y sample. The in-plane ρ_0, A, and p parameters obtained from the fit are 3.38 $\mu\Omega$ cm, 8.1×10^{-4} $\mu\Omega$ cm/K^p, and 2.2 for the sc-YNi_2B_2C, and 1.36 $\mu\Omega$ cm, 1.8×10^{-3} $\mu\Omega$ cm/K^p, and 2.0 for the sc-$LuNi_2B_2C$. Thus, the low-temperature in plane resistivity of these metallic sc-Y(Lu)Ni_2B_2C compounds seems to be anomalous in the temperature region $1.25T_c<T<0.1\Theta_D$ in a sense that its temperature dependence is different from the T^5 or T^3 dependence expected from the conventional theory and usually

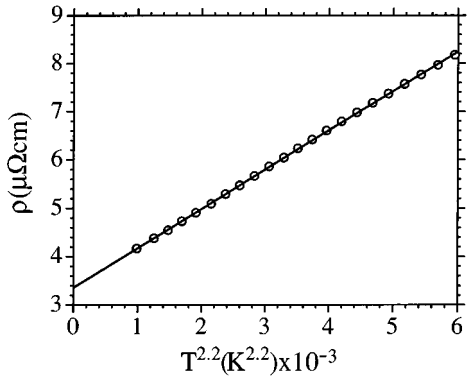

FIG. 2. Resistivity vs T^p, where $p=2.2$, for sc-YNi_2B_2C. The straight line drawn through the points represents a straight line least-sqaures fit.

observed for ordinary and transition metals. The exponent p, which is approximately equal to 2 for either of these borocarbide compounds, is similar to the one found[7,59] for $\rho(T)$ of disordered superconducting high-T_c A-15 compounds (Nb_3Sn, Nb_3Al, Nb_3Ge, V_3Si, etc.), i.e., $p=2$. The value of A is smaller for the Y sample than for the Lu sample as expected from the resistivity values. The T^2 behavior of ρ in A-15 compounds was initially suggested by Webb *et al.*[60] to be due to a non-Debye phonon spectrum $F(\omega)$ which was later discounted by Gurvitch[61] through a careful analysis of $\rho(T)$ data on disordered A-15 compounds. The possibility that the electron-electron interaction gives the T^2 dependence of $\rho(T)$ can be ruled out since the coefficient A is about two to three orders of magnitude larger than expected from this scattering mechanism.[59,60,62] The T^2 behavior is also observed in magnetic or nearly magnetic metals. It is possible that scattering from reported antiferromagnetic fluctuations on the Ni sites,[35,36] could contribute to this temperature dependence, but other experiments[21,34] do not confirm these reports. Gurvitch[63] has proposed that strong electron-phonon coupling is the necessary condition for observing $\rho_{\text{e-ph}}\ \alpha T^2$ in A-15 compounds, but it is not sufficient; the simultaneous presence of strong coupling and high disorder are required to produce the temperature exponent p equal to 2.0. A high disorder results in the breakdown of the electron-phonon momentum conservation law $\mathbf{k}\pm\overline{q}=\mathbf{k}'$ and may be responsible for the T^2 behavior of ρ in the A-15's. RNi_2B_2C (R=Y,Lu) have λ values similar to those of some of A-15 compounds, but the samples investigated here are single crystals, and therefore do not have large disorder. Therefore the possibility of the breakdown of the electron-phonon momentum conservation law does not arise. The present result $\rho(T)\alpha T^2$ for sc-Y(Lu)Ni_2B_2C at low temperatures must be due to some other mechanism(s). Recent photoemission studies[32] on YNi_2B_2C indicate that the Ni $3d$-derived conduction bands are narrower than the calculated ones, and are accompanied by a satellite. These results are indicative of the presence of electron-electron correlation effects. Such effects, as well as some other unidentified scattering mechanisms, may be responsible for the quadratic temperature dependence at low temperatures.

While no measurements on the c-axis electrical resistivity of any borocarbide superconductors RNi_2B_2C have been reported as yet, Sera *et al.*[47] have recently reported in-plane and the c-axis thermal conductivity measurements on RNi_2B_2C (R=Y,Ho). They find little anisotropy between the c and a axes at low temperatures. Assuming that the Wiedemann-Franz law is valid for these superconductors, it means that there will be little anisotropy between the in-plane and c-axis resistivities, unlike in high-temperature superconductors, although these borocarbide superconductors also have a layered structure.

Figure 3 shows the dependence of in-plane resistance of YNi_2B_2C at 25 K as a function of magnetic field (parallel to the c axis) up to 4.5 T. The change in the resistance at 4.5 T is about 6.5% which is quite large in comparison with normal metals.[64] Figure 3 clearly shows that the resistance varies nearly as H^2 for low field values, and seems to become linear with H at higher magnetic fields. A fit to data to $R(T,H)=R(T,0)+R1*H+R2*H^2$ shows that $R1/R2\approx 10$ indicating that there is a substantial contribution from the linear term in addition to the quadratic term. A fit to the power law $[R(T,H)-R(T,0)]/R(T,0)=AH^n$ gives $n=1.27$ indicating that the magnetic-field dependence of the magnetoresistance is closer to linear than quadratic in H. This fit is shown as an inset in Fig. 3. At lower temperature, i.e., $T=15$ K, we find $[R(T,4.5T)-R(T,0)]/R(T,0)\approx 7.5\%$ and 8.0% for H parallel and perpendicular to the c axis indicating a small anisotropy in the magnetoresistance. A similar amount of magnetoresistance has been observed in a YNi_2B_2C polycrystalline sample.[65] Magnetoresistance of the order of 40% at 8 T (with magnetic field perpendicular to the current) has been reported[55] in polycrystalline $LuNi_2B_2C$ with estimated electron mean free path $l=700$ Å. It is difficult to explain such a large electron mean free path in a polycrystalline sample compared to the value of about 200 Å in our single-crystal sample of $LuNi_2B_2C$. In contrast, we find a magnitude ($\Delta R/R\approx 7.3\%$ for $H=45$ kOe at 20 K) and field dependence for sc-$LuNi_2B_2C$ very similar to that shown in Fig. 3. Perhaps the grain boundaries in the polycrystalline sample in Ref. 55 consist of a different compound with very unusual properties.

When a magnetic field is applied, the resulting electron orbits may be closed or open depending upon the topology of the Fermi surface. The transverse magnetoresistance saturates for closed orbits but grows indefinitely as H^2 for open orbits.[64,66,67] A linear magnetoresistance at high fields has been observed for single crystals as well as polycrystalline materials.[68–71] While there is hardly any good understanding of the linear magnetoresistance, it has been shown theoretically to occur in polycrystals with open orbits,[66,72] due to thickness variation,[73] or also if the carrier transport takes place along two-dimensional skipping-orbit states.[74] Most recently Park and Kim[75] have shown that the interference of two open orbits modeled by two cylindrical Fermi surfaces gives rise to a linear transverse magnetoresistance when the magnetic field is slightly tilted from the direction perpendicular to the plane defined by the cylinder axes. Kim, Hwang, and Ihm[30] show that there are five electronic complex Fermi surfaces of $LuNi_2B_2C$, one of which is a cylindrical surface perpendicular to the c axis. Thus, the open electron orbits are expected in $LuNi_2B_2C$ (and YNi_2B_2C due to similarity between their band structures) which may lead to the linear magnetoresistance as predicted by Park and Kim and observed by us at high fields. A more careful study of

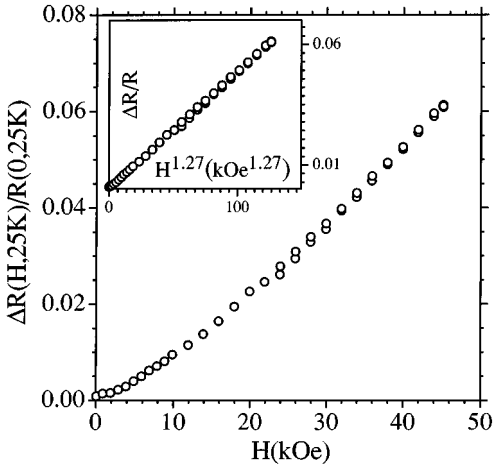

FIG. 3. Field dependence of the transverse magnetoresistance, $\Delta R/R(0,25\ \mathrm{K})=[R(H,25\ \mathrm{K})-R(0,25\ \mathrm{K})]/R(0,25\ \mathrm{K})$ for YNi_2B_2C with magnetic field parallel to the c axis. The inset shows the variation of the magnetoresistance with $H^{1.27}$.

magnetoresistance in these compounds is needed for a better understanding of the phenomenon.

B. Thermopower

The absolute thermopower, $S(T)$, measured with temperature gradient along the ab plane of the single crystals of YNi_2B_2C and $LuNi_2B_2C$, is shown in Fig. 4 as a function of temperature. $S(T)$ is negative for both the samples from RT to just above the superconducting transition temperature T_c where it rapidly drops to zero within the measurement accuracy. The superconducting transition temperatures determined this way are within ± 0.25 K of that determined by the resistivity measurements. The sharp fall of S to zero at T_c also confirms the good quality of the samples. The negative thermopower does not necessarily mean that the charge carriers in these compounds are electrons;[76] however, the band-structure calculations[26–31] indicate so. Hall effect measurements are needed to ascertain the nature of charge carriers.

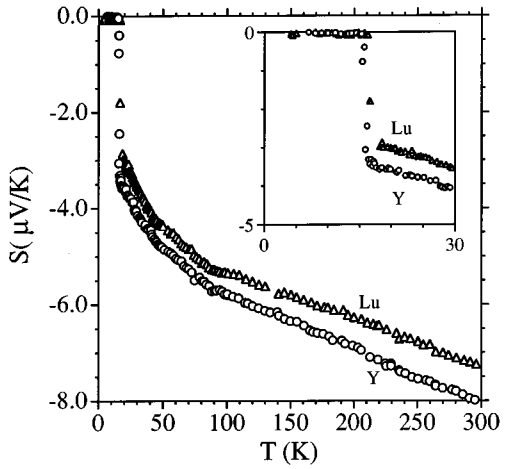

FIG. 4. Temperature dependence of the in-plane thermopower of single-crystal samples of YNi_2B_2C (open circles) and $LuNi_2B_2C$ (open triangles). The inset shows data close to T_c.

The thermopower of both samples is seen to be linear in T near room temperature within the measurement accuracy. Room-temperature thermopower $S(\mathrm{RT})$ and $[dS/dT]_{\mathrm{RT}}$ are -8.1 μV/K and -11.5 nV/K^2, and -7.3 μV/K and -10.4 nV/K^2 for YNi_2B_2C and $LuNi_2B_2C$ samples, respectively, which are nearly the same for both samples. The magnitude of $S(\mathrm{RT})$, reported here, is somewhat larger than the typical value associated with free electron/conventional metals, i.e., -1.28 μV/K for lead and 1.94 μV/K for gold, but it is approximately the same as for palladium[76] [$S(\mathrm{RT})=-10$ μV/K] and many high-T_c cuprate superconductors.[77]

The thermopower of conventional nonmagnetic metals consists of two contributions, a diffusion contribution and a phonon-drag contribution resulting from the transfer of phonon momentum to the electron gas. The diffusion contribution is proportional to temperature, while the phonon-drag contribution falls at low temperature as the phonons freeze out, and at high temperatures as the excess phonon momentum gets limited by phonon-phonon scattering. This usually results in a phonon-drag peak in conventional metals with T^3 dependence below $0.1\Theta_D$ and falls as T^{-1} above $\approx 0.3\Theta_D$. Figure 4 clearly shows that there are no structures (peak, etc.) in S vs T data from RT down to T_c, i.e., no obvious phonon-drag peak is present which one would expect,[76] if present, in the temperature region $0.1\Theta_D < T < 0.3\Theta_D$. Since these samples are single crystals and have reasonably high resistivity ratios one would have expected some signature of a phonon-drag peak in the data. However, the usual signature is totally absent, and S is similar to that in amorphous metals[78,79] and, except for sign, many high-T_c cuprate superconductors.[77]

It is obvious from the thermopower data that in addition to the diffusion thermopower, which is proportional to the temperature, there is(are) additional contribution(s) to the thermopower yielding the observed temperature dependence of S. Figure 5 shows data (S-bT) vs T for YNi_2B_2C and $LuNi_2B_2C$ single crystals, where b is the coefficient obtained by fitting the S vs T data to a straight line, i.e., $S(T)=a+bT$, in the linear region ($T\approx 100$ K to RT). (S-bT) represents contributions to the thermopower other than the diffusion thermopower, and as observed from Fig. 5, this contribution is negative and almost constant between 100 and 300 K for both samples; approximately -4.3 μV/K for the $LuNi_2B_2C$ sample and -4.8 μV/K for the YNi_2B_2C sample. Below 100 K, this contribution to the thermopower for each sample varies approximately as T^{-1}, until the superconducting transition temperature at which it rises abruptly to zero. Generally, the most important contribution to the thermopower of a metal, in addition to the diffusion thermopower, is the phonon-drag contribution. Any contribution from possible magnetic impurities is expected to be much smaller than that observed in Fig. 5. Recently, Trodahl[80] tried to explain the thermopower of high-T_c cuprate superconductors by including the phonon-drag contribution with the assumption that the phonon-phonon scattering in high-T_c cuprates remains weaker than phonon-electron scattering even at room temperature. He finds that the temperature dependence of the phonon-drag contribution to the thermopower of high-T_c cuprates is very similar to that shown for S-bT in Fig. 5, i.e., it is almost temperature independent between 100 K and RT, and this constant value represents

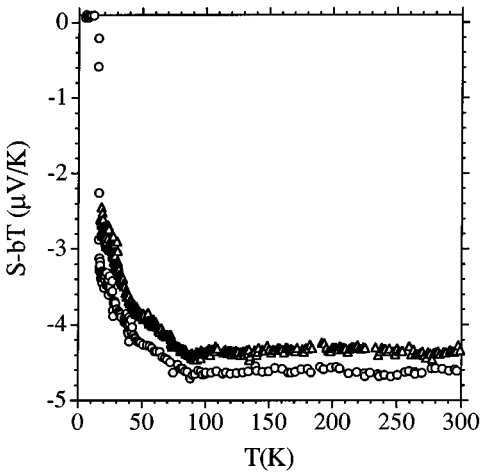

FIG. 5. Temperature dependence of $(S\text{-}bT)$ vs T for sc-YNi_2B_2C and sc-$LuNi_2B_2C$. The coefficient b is the slope of the straight line obtained by least-squares fitting of the data in the linear region (between 100 K and RT). $(S\text{-}bT)$ represents the phonon-drag contribution as a function of temperature assuming either no or comparatively very small other contributions to the total thermopower in comparison with the diffusion thermopower. Symbols are the same as in Fig. 4.

the saturation value of the phonon-drag thermopower. This kind of temperature dependence of the phonon-drag thermopower leads to a simple shift of the linear diffusion thermopower between 100 K and RT. This particular behavior relates to the layered nature of high-T_c cuprates. Borocarbides are also layered compounds; hence a similar phonon-drag contribution to the thermopower may not be unreasonable. Therefore, assuming that Fig. 5 represents the phonon-drag contribution to the total thermopower the saturation value of this contribution in sc-Y(Lu)Ni_2B_2C is estimated to be ≈ -4.5 μV/K.

The extrapolation of the data of Fig. 4 near room temperature, assuming a linear T dependence of S, does not pass through $S=0$ at $T=0$, and gives intercepts of -4.61 and -4.34 μV/K, respectively, for the Y and Lu samples. This result implies that, in the absence of superconductivity, there would be a low temperature ''knee'' in $S(T)$, similar to that produced by electron-phonon renormalization.[81] This ''knee'' seems to be present near 70 K for Y and 100 K for Lu samples. Electron-phonon renormalization would lead to an enhanced thermopower that is given by

$$S = S_b[1+\lambda(T)], \tag{6}$$

where $\lambda(T)$ is the electron-phonon mass enhancement parameter and S_b is the bare thermopower (without renormalization effects). In this expression certain corrections[82] have been assumed small and therefore neglected. Equation (6) can be rewritten as

$$\frac{S}{T} = \frac{S_b}{T}\,[1+\lambda(T)]. \tag{7}$$

A plot of S/T vs T should then give a measure of $\lambda(T)$, and $[S/T]_{T\Rightarrow 0}/[S/T]_{\mathrm{RT}}$ should approximate $1+\lambda(0)$.[79] Assuming that the S/T value just above T_c is $[S/T]_{T\Rightarrow 0}$ as an approximation, we find that $\lambda(0)>5$–6 which is absurdly high when compared to the estimated values of $\lambda(0)$ for Y (Lu)Ni_2B_2C by others[26,27,30,31] or to those obtained here or to the values observed for even very strong-coupling superconductors[57] (viz. Pb-Bi alloys). It may be mentioned that a recent μSR study[83] on YNi_2B_2C reports a mass-enhancement factor of 9.4 from penetration depth measurements. Recently Kaiser and Mountjoy[84] have explained the thermopower of high-T_c superconductors within the existing metallic diffusion-thermopower theory in terms of an anomalously large strong electron-phonon coupling (greater than 5), such as might arise from an anharmonic double-well potential[85] in $YBa_2Cu_3O_{7-\delta}$. It has been recently reported[86] that the electron-phonon coupling λ for a double-well potential could reach huge values, 5–50 or more. Whether large and anisotropic thermal vibrations of carbon atoms in the Y-C plane in YNi_2B_2C, as observed by Godart *et al.*,[38] would lead to such a situation should be studied theoretically.

In the presence of spin fluctuations, Eq. (7) is modified to

$$\frac{S}{T} = \frac{S_b}{T}\,[1+\lambda(T)+\lambda_{\mathrm{sf}}], \tag{8}$$

where λ_{sf} is the mass-enhancement parameter due to spin fluctuations. Kohara *et al.* and others[35] have reported the presence of antiferromagnetic spin fluctuations in polycrystalline YNi_2B_2C but a NMR study of single-crystal YNi_2B_2C by Suh *et al.*[34] shows no antiferromagnetic correlations. Even if such fluctuations were present, one would not expect λ_{sf} to have large values which could explain $[S/T]_{T\Rightarrow 0}/[S/T]_{\mathrm{RT}}\approx 5$–7.

Lee *et al.*[46] have reported $S(T)$ for polycrystalline YNi_2B_2C and $LuNi_2B_2C$. They find S(RT) values to be -5.1 and -4.7 μV/K for YNi_2B_2C and $LuNi_2B_2C$ polycrystalline samples, respectively. These are almost two-thirds of the values reported here for the single-crystal samples. $[dS/dT]$ is almost zero in their work in the temperature interval 150 K$<T<$RT. Thus, the present results on YNi_2B_2C and $LuNi_2B_2C$ single crystals are in disagreement, except for the sign of the thermopower, with those on polycrystalline Y(Lu)Ni_2B_2C. However, the single-crystal measurements are taken with ΔT along the ab plane. Polycrystalline samples would give an average of the thermopower along the ab plane and the c axis, appropriately weighted for the conductivities in those directions.[76] Since no measurement of thermopower is available along the c axis of these crystals, it is not possible to comment on it quantitatively. If this is the reason for the difference between S for single crystals and polycrystalline samples, the polycrystalline results indicate a possibility of a positive thermopower along the c axis of Y(Lu)Ni_2B_2C single crystals. Lee *et al.*[46] have suggested that the nonlinear behavior of the thermopower of Y(Lu)Ni_2B_2C polycrystalline samples may be due to the mixed-valence phenomenon like that found in $CeNi_x$,[87] and they found a good fit to the expression $S'=S-bT=AT/(B^2+T^2)$, where b, A, and B are constants. The data for single crystals presented here could not be well described by such a fit, and there is no reason to expect temperature-dependent valence fluctuations in these compounds. Possibly the difference may be attributed to a strikingly different behavior between the

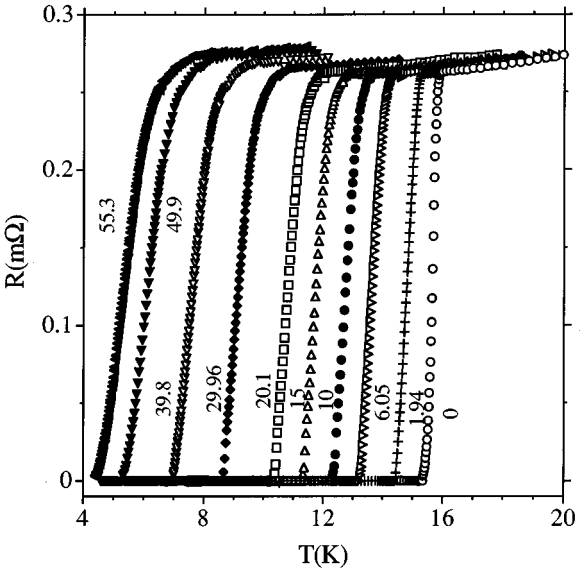

FIG. 6. Magnetic-field dependence of resistive transition to superconductivity for YNi_2B_2C with magnetic field perpendicular to the c axis. The field values are in kOe. Only a few transition curves are shown for the sake of clarity.

bulk crystals and the grain-boundary materials which are probably far from stoichiometry.

C. Superconductivity

The superconducting transition temperatures of YNi_2B_2C and $LuNi_2B_2C$ single-crystal samples are found to be 15.6 and 16.1 K, respectively, from the resistive transition measurements where T_c is defined as the intersection of the line drawn through the steepest part of the transition curve with the temperature axis. In order to determine the upper critical field $H_{c2}(T)$, resistance of the samples has been measured as a function of temperature in various fixed values of magnetic fields applied parallel and perpendicular to the c axis of both samples. Figure 6 shows the results for YNi_2B_2C sample with the applied magnetic field perpendicular to the c axis which clearly shows that the magnetic field *basically* shifts the resistive transition curve without affecting the transition width significantly at lower magnetic fields ($H<50$ kOe), i.e., the effect of H is only to decrease T_c of the sample. The large positive magnetoresistance can also be seen in this figure. Similar behavior of the resistive superconducting transition is observed for a magnetic field applied parallel to the c axis of YNi_2B_2C and for both field directions with $LuNi_2B_2C$. Magnetization measurements, i.e., M vs T for single-crystal YNi_2B_2C also show similar behavior for different applied magnetic fields.[8,48] This behavior of the superconducting transition in an applied magnetic field is in contrast with the high-T_c cuprate superconductors in which the transition width increases even in presence of a small external magnetic field, and where it increases enormously as the applied magnetic field is increased, leading to extensive tailing effects as R approaches zero.[88] In such a case it becomes difficult to define $H_{c2}(T)$; however, the sharp transition curves for sc-Y(Lu)Ni_2B_2C samples investigated here permit determination of $H_{c2}(T)$ unambiguously.

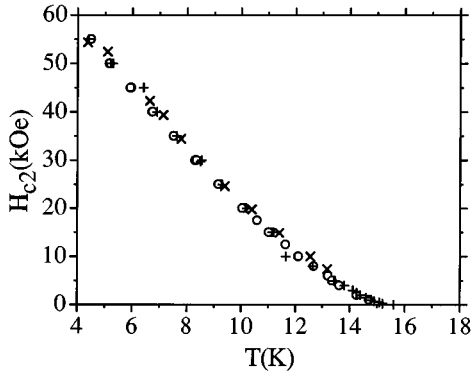

FIG. 7. Temperature dependence of the upper critical field H_{c2} for the YNi_2B_2C single-crystal sample with magnetic field parallel (open circles) and perpendicular ($\times$) to the c axis. The current density was $\approx$26 A/cm^2 and was along the ab plane. Data deduced from magnetization measurements (Ref. 8) with field normal to the c axis (crosses) are shown for comparison.

$T_c(H)$ is determined from the intersection of the straight line drawn through the steep superconducting transition region and the T axis which is then converted into $H_{c2}(T)$ for each sample. We shall refer to $H_{c2}^{\rm pl}(T)$ and $H_{c2}^{\rm pr}(T)$ as the upper critical fields parallel and perpendicular to the c axis of the crystal. Figures 7 and 8 show $H_{c2}^{\rm pl}(T)$ and $H_{c2}^{\rm pr}(T)$ vs T for sc-YNi_2B_2C and sc-$LuNi_2B_2C$ samples. The current was 50 mA (current density$\approx$26 A/cm^2) for the Y sample, and 10 mA ($\approx$3 A/cm^2) for the Lu sample in the ab plane of each. From the figures we note that the upper critical fields $H_{c2}^{\rm pl}(T)$ and $H_{c2}^{\rm pr}(T)$ of the Y sample show almost no anisotropy, while those of the Lu sample show a small anisotropy, i.e., $[H_{c2}^{\rm pr}(T)]_{\rm Lu}$ is larger than $[H_{c2}^{\rm pl}(T)]_{\rm Lu}$ and the difference becomes larger at lower temperatures. $[H_{c2}^{\rm pr}(T)]_{\rm Lu}$ is $\approx$15% higher than $[H_{c2}^{\rm pl}(T)]_{\rm Lu}$ at $T\approx$4.5 K. Absence of the anisotropy of $H_{c2}(T)$ of sc-YNi_2B_2C determined from the resistive transition in a magnetic field agrees well with reports based on magnetization measurements.[8,48] This observation is in contrast to the strong anisotropy seen in $H_{c2}(T)$ between in-plane and c-axis-aligned fields in cuprate high-T_c superconductors[88] which also have a layered structure.

It has been previously reported[14] that $H_{c2}(T)$ for $HoNi_2B_2C$ single-crystal samples is strongly current dependent, especially in the range of fields and temperature where reentrant superconductivity is observed. To determine if such a dependence exists for sc-YNi_2B_2C samples, $H_{c2}^{\rm pl}(T)$ and $H_{c2}^{\rm pr}(T)$ were obtained from the resistive transition curves recorded using two different current densities, i.e., 2.6 and 26 A/cm^2, along the ab plane which are shown in Fig. 9. A small current dependence is clearly observed becoming larger at lower temperatures; $H_{c2}(T)$ differs by $\approx$10% at the lowest temperature ($\approx$4.5 K) for the two current densities. It should be noted, however, that the anisotropy between $H_{c2}^{\rm pl}(T)$ and $H_{c2}^{\rm pr}(T)$, for a given current density, does not alter, and the superconducting transition temperature $T_c(H=0)$ is *exactly* the same for the two current densities. A similar current density dependence was observed with $LuNi_2B_2C$ samples. The observed current density dependence of $H_{c2}(T)$ for these samples remains unexplained and is definitely not due to a heating effect, as pointed out earlier.[14]

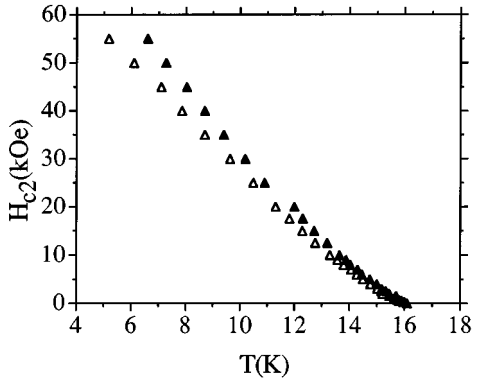

FIG. 8. Temperature dependence of the upper critical field H_{c2} for the $LuNi_2B_2C$ single-crystal sample with magnetic field parallel (open triangles) and perpendicular (filled triangles) to the c axis. The current density was ≈ 3 A/cm^2 and was along the ab plane.

$H_{c2}(T)$ vs T data in Figs. 7 and 8 show that $H_{c2}(T)$ does not intersect the T axis *linearly* but bends towards the higher T side giving a positive curvature to the $H_{c2}(T)$ curve, i.e., $d^2H_{c2}/dT^2>0$. Please note that determination of $H_{c2}(T)$ from magnetization measurements[8] on single-crystal YNi_2B_2C are in excellent agreement with those from resistance measurements as shown in Fig. 7. A similar positive curvature in $H_{c2}(T)|_{T\rightarrow T_c}$ has been reported in polycrystalline and single-crystal RNi_2B_2C compounds,[8,14,54,55,89,90] dichalcogenides,[91] amorphous superconductors,[92] A-15 superconducting compounds,[7] Ba-doped C_{60},[93] and in high-T_c cuprate superconductors.[94] The Werthamer, Helfand, Hohenberg, and Maki, (WHHM) theory,[95,96] which takes into account the orbital and paramagnetic effects of an external field as well as nonmagnetic and spin-orbit scatterings, is normally used to explain $H_{c2}(T)$ of superconducting alloys (type-II superconductors). The WHHM theory predicts a linear temperature dependence of $H_{c2}(T)$ near T_c, i.e., $H_{c2}(T)\propto(1-T/T_c)$, in either the ''clean'' or the ''dirty'' limit. This theory thus fails to explain the positive curvature of $H_{c2}(T)$ near T_c as observed in a large number of superconducting systems. Several explanations have been advanced to explain this behavior of $H_{c2}(T)$. Some of these are: (1) scattering by magnetic impurities,[97] (2) reduced dimensionality and disorder,[98,99] (3) strong electron-phonon coupling effects,[100] (4) bipolaron effect[101,102] and (5) two-component response[90] in granular polycrystalline superconductors (intra- and intergranular effects). In the case of high-T_c cuprate superconductors, $H_{c2}(T)$ is found to vary as $(1-T/T_c)^{3/2}$ near T_c if H_{c2} is derived from the resistance measurements or as $(1-T/T_c)$ if it is derived from magnetization measurements.[88] In high-T_c cuprate superconductors this peculiar temperature dependence of H_{c2} near T_c has been attributed to the flux flow in $R(T,H)$ measurements as evidenced by long resistive tails in the transition region, and to the twin boundaries limiting the spatial extent of the vortices in one direction.[88] Neither of these possible causes are present in $Y(Lu)Ni_2B_2C$ single-crystal samples; yet a positive curvature is observed in $H_{c2}(T)$ near T_c regardless of whether the applied magnetic field is parallel or perpendicular to the c axis. Reduced dimensionality and disorder cannot be applicable to either of the two samples investigated here since they behave like three-dimensional metals,[26,27] and they are single crystals with relatively low resistivities, excluding a high degree of disorder. Two-component response[90] may be applicable to the granular polycrystalline samples but not to single-crystal RNi_2B_2C samples. There seems to be no evidence of applicability of the bipolaron model to RNi_2B_2C superconductors, moreover in this model[101] $H_{c2}(0)$ goes to infinity unless limited by some additional effects like localization of bosons. The presence of a small amount of magnetic impurities is certainly a possibility but unless these are present in large quantities, $H_{c2}(T)$ is not expected to be affected substantially.[88] A possibility of distributed T_c in a given sample does not arise here as the samples are single crystals, and the resistive transitions are quite sharp.

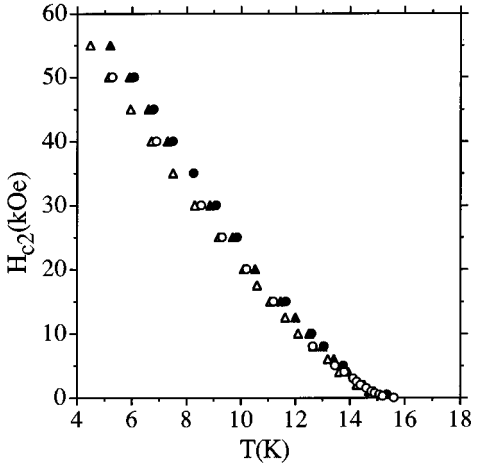

FIG. 9. The current dependence of $H_{c2}(T)$ for the YNi_2B_2C single-crystal sample. Circles and triangles represent data with magnetic field perpendicular and parallel, respectively, to the c axis. Open and filled circles/triangles are for 50 mA ($\approx$26 A/cm^2) and 5 mA ($\approx$2.6 A/cm^2) current through the sample, respectively.

The positive curvature in $H_{c2}(T)$ near T_c makes it difficult to apply the standard WHHM theory and its extensions to obtain certain characteristic parameters in the superconducting state of $Y(Lu)Ni_2B_2C$ single crystals. However, this theory seems to be the only one which would give certain characteristic superconducting parameters. To apply the theory, the $H_{c2}(T)$ vs T data is divided into two sections: (a) the lower temperature data which fits to a straight line yielding T_{ca} which turns out to be lower than the observed T_c (T_{ca} is determined by the intersection of this line with the T axis.); (b) The region near T_c which shows the positive curvature. Figures 7 and 8 show that the low-temperature data almost lie on a straight line. As a matter of fact one would not expect from the WHHM theory the $H_{c2}(T)$ data to lie on a straight line at such low temperatures ($T/T_c<0.7$). To obtain $[dH_{c2}/dT]_{T=T_c}$, the low-temperature $H_{c2}(T)$ data between $T=0.85T_c$ and $0.25T_c$ is fitted to a straight line. The goodness of the fit is measured by the square of the correlation coefficient ($r^2>0.997$). The slope obtained is not $[dH_{c2}/dT]_{T=T_c}$ but $[dH_{c2}/dT]_{T=T_{ca}}$. This slope is then used to calculate the upper critical field $H_{c2}(0)$, the Ginzburg-Landau coherence length $\xi(0)$ and/or the BCS coherence length $\xi_{0\mathrm{BCS}}$, the thermodynamic critical field $H_c(0)$

with the knowledge of experimentally determined H_{c1}, the Ginzburg-Landau parameter $\kappa(0)$, the magnetic-field penetration depth $\lambda(0)$, and the specific-heat constant γ, since all of these can be expressed in terms of $[dH_{c2}/dT]_{T=T_c}$ and/or the parameters derived from it. However, the exact relationship between these parameters and $[dH_{c2}/dT]_{T=T_c}$ depends upon whether the sample is ''clean'' ($l \gg \xi_{0\mathrm{BCS}}$) or ''dirty'' ($l \ll \xi_{0\mathrm{BCS}}$). The electron mean free path l has already been estimated for both the samples by the resistivity measurements. The BCS coherence length $\xi_{0\mathrm{BCS}}$ can be determined from the BCS relation:

$$\xi^{\Delta}_{0\mathrm{BCS}} = \frac{\hbar v_F}{\pi \Delta_o}, \tag{9}$$

where Δ_o is the energy gap at $T=0$ K, and the Δ superscript on $\xi_{0\mathrm{BCS}}$ indicates that it has been determined using the above relation. Recent break junction tunneling measurements[9] on polycrystalline YNi_2B_2C show that the energy gap of YNi_2B_2C and $LuNi_2B_2C$ obeys the BCS relation

$$2\Delta_o \approx (3.5 \pm 0.1) k_B T_C, \tag{10}$$

suggesting that these are BCS-type weak-coupling superconductors. Equation (9) gives $\xi^{\Delta}_{0\mathrm{BCS}} = 321$ Å for YNi_2B_2C and 311 Å for $LuNi_2B_2C$ where $v_F = 3.6 \times 10^7$ cm/s is taken from the band-structure calculations.[27] For both of these compounds, the electron mean free path l is smaller than $\xi^{\Delta}_{0\mathrm{BCS}}$, i.e., $[l/\xi^{\Delta}_{0\mathrm{BCS}}]$ is approximately equal to 0.34 for YNi_2B_2C and 0.61 for $LuNi_2B_2C$. These ratios indicate that the Y and Lu single-crystal samples may be considered as ''quasidirty.'' Thus, various standard formulas[95,103,104] valid for the ''dirty'' limit are used in the analysis:

$$H_{c2}(0) = 0.693 T_c \left[-\frac{dH_{c2}}{dT} \right]_{T=T_c}, \tag{11}$$

TABLE II. Superconducting parameters for single crystal YNi_2B_2C and $LuNi_2B_2C$ determined from critical field measurements assuming the dirty limit.

	YNi_2B_2C	$LuNi_2B_2C$
T_c K	15.6	16.1
H_{c1} Oer	369[a]	800[b]
$H^{\mathrm{pl}}_{c2}(0)$ kOe	65.2	61.5
$H^{\mathrm{pr}}_{c2}(0)$ kOe	65.1	69.5
$\xi^{\mathrm{pl}}(0)$ Å	71	73
$\xi^{\mathrm{pr}}(0)$ Å	71	69
$\xi^{\mathrm{pl}}_{0\ \mathrm{BCS}}$Å	64	39
$\xi^{\mathrm{pr}}_{0\ \mathrm{BCS}}$ Å	64	35
$H^{\mathrm{pl}}_{c}(0)$ kOe	2.71	4.18
$H^{\mathrm{pr}}_{c}(0)$ kOe	2.71	4.39
$\kappa^{\mathrm{pl}}(0)$	17	10.4
$\kappa^{\mathrm{pr}}(0)$	17	11.2
$\lambda^{\mathrm{pl}}(0)$ Å	1207	759
$\lambda^{\mathrm{pr}}(0)$ Å	1207	772

[a]References 82 and 105.
[b]Reference 55.

$$\xi(0) = 0.85(\xi_{0\mathrm{BCS}} l)^{1/2} = \sqrt{\frac{\phi_0}{2\pi H_{c2}(0)}}, \tag{12}$$

where ϕ_0 is the flux quantum. Other formulas used are

$$2\frac{H_{c1}}{H_{c2}} = \frac{\ln \kappa + 0.5}{\kappa^2}, \tag{13}$$

$$H_c(0) = \frac{H_{c2}(0)}{\sqrt{2}\kappa(0)}, \tag{14}$$

$$\lambda(0) = \kappa(0)\xi(0). \tag{15}$$

The slopes $[dH^{\mathrm{pl}}_{c2}/dT]_{T=T_{ca}}$ and $[dH^{\mathrm{pr}}_{c2}/dT]_{T=T_{ca}}$, instead of $[dH^{\mathrm{pl}}_{c2}/dT]_{T=T_c}$ and $[dH^{\mathrm{pr}}_{c2}/dT]_{T=T_c}$, determined as explained above, are used to calculate $H^{\mathrm{pl}}_{c2}(0)$ and $H^{\mathrm{pr}}_{c2}(0)$ from Eq. (11). $[dH^{\mathrm{pl}}_{c2}/dT]_{T=T_{ca}}$ and $[dH^{\mathrm{pr}}_{c2}/dT]_{T=T_{ca}}$ are -6.03 and -6.02 kOe/K, respectively, for the YNi_2B_2C sample, and -5.51 and -6.23 kOe/K, respectively, for the $LuNi_2B_2C$ sample. The superconducting parameters determined from substitution of these values in the above formulas are tabulated in Table II.

The values of various parameters obtained here agree reasonably well (within 10–15 %) with the earlier reported values for single crystals as well as polycrystalline YNi_2B_2C samples. It is clear that there is essentially no anisotropy in superconducting parameters of YNi_2B_2C single crystals with respect to the c axis. Earlier reports on the upper critical field measurements for $LuNi_2B_2C$ polycrystalline samples give $H_{c2}(0) = 90$ kOe by Takagi *et al.*[55] and 57.5 kOe by Kim, Kim, and Stewart.[54] The $H_{c2}(0)$ values determined here for sc-$LuNi_2B_2C$ are close to that of Kim, Kim, and Stewart and only two-thirds of the value reported by Takagi *et al.*, but the other parameters [$\xi(0)$, λ, and κ] agree within 10–20 % of the reported values by them. The anisotropy in various superconducting parameters with respect to the c axis is of the order of 10% in $LuNi_2B_2C$ in contrast to the absence of anisotropy for YNi_2B_2C.

The values of the BCS coherence length calculated from the slope of $H_{c2}(T)$, as done above, are very small, i.e., about 65 Å for YNi_2B_2C and about 35 Å for $LuNi_2B_2C$. These values are in severe disagreement with the BCS coherence length calculated from Eq. (9) in which the energy gap is taken from the tunneling measurements. Strong-coupling effects cannot explain this disagreement even though renormalization of the Fermi velocity by the factor $(1+\lambda)^{-1}$ will reduce the value from Eq. (9) by approximately a factor of 2. To match the value of $\xi^{\Delta}_{0\mathrm{BCS}}$ from Eq. (9) to that determined from the slope of the critical field would require a ratio of $2\Delta_0/k_B T_c = 9$–15 rather than the value 3.5 reported in Ref. 9, a clearly unreasonable number. Another possibility is that the theoretical value of v_F is off by a factor of 2–5. Based on the value of $\xi_{0\mathrm{BCS}}$ determined from the critical field the samples would be in the clean limit which would change the numerical factor 0.693 in Eq. (11) to 0.727 and the factor 0.85 in Eq. (12) to 0.74. This would increase the parameters calculated for Table II by almost 5% and allow calculation of the specific-heat coefficient γ from the formula given by Wiesman *et al.*[106] for clean superconductors, i.e., $l \gg \xi_0^{H_{c2}}$,

$$\gamma = 2.16\times 10^{-5}\left[-\frac{dH_{c2}}{dT}\right]_{T=T_c}\left[\frac{1}{\rho_0(1+1.3l/(\xi_0^{H_{c2}})^*}\right], \tag{16}$$

where $(\xi_0^{H_{c2}})^* = \xi_0^{H_{c2}}/(1+\lambda)$, and superscript H_{c2} means that the coherence length is determined from $[dH_{c2}/dT]_{T=T_{ca}}$, λ is the electron-phonon coupling parameter, γ is in ergs/(cm^3 K^2) and ρ_o is in Ω cm. With $l=l_{\rm tr}$ determined earlier from the resistivity just above T_c, and $\lambda=\lambda_{\rm tr}$, for YNi_2B_2C $\gamma^{\rm pr}=\gamma^{\rm pl}=22.8$ mJ/(mol K^2), and for $LuNi_2B_2C$, $\gamma^{\rm pl}=18.2$ mJ/(mol K^2) and $\gamma^{\rm pr}=18.6$ mJ/(mol K^2). These values agree very well with those reported recently, i.e., 18.7 mJ/(mol K^2) for YNi_2B_2C (Ref. 52) and 19 mJ/(mol K^2) for $LuNi_2B_2C$.[40]

While we have used $H_{c2}(T)$ data well below T_c to get $[dH_{c2}/dT]_{T=T_{ca}}$, the positive curvature in $H_{c2}(T)$ near T_c has not been explained. Recently Bahcall[107] has shown that solving the BCS-Gor'kov theory near $H_{c2}(T)$ directly by generalizing pairing between plane waves to pairing between many electronic Landau levels leads to significant deviations from the semiclassical theory for clean and isotropic superconductors. This approach lowers the value of the upper critical field and causes $H_{c2}(T)$ to vanish quadratically near T_c leading to a positive curvature, i.e., $d^2H_{c2}/dT^2>0$ near T_c, and indicates that a type-II superconductor converts to type I near T_c. Bahcall has given the following equation which describes the temperature dependence of $H_{c2}(T)$ near T_c:

$$1.61e^{-1/g}\sqrt{h}+0.727h-(1-t)=0, \tag{17}$$

where $h=H_{c2}^B(T)/H_{c2}^S(0)$, $H_{c2}^B(T)$ refers to the calculated $H_{c2}(T)$ from Bahcall's theory, $H_{c2}^S(0)$ is the upper critical field at $T=0$ K in the semiclassical theory, $g=V_oN(0)$ is the BCS coupling constant, and $t=T/T_c$. In the limit $g\to 0$, the semiclassical result, $H_{c2}^S(0)=H_{c2}(0)=-0.727T_c[dH_{c2}/dT]_{T=Tc}$, is obtained (for clean type-II superconductors). For nonzero values of g, the presence of the $\sqrt{h}$ term makes $H_{c2}(T)$ vanish quadratically near T_c, not linearly as in the WHHM theory. This theory leads to a smaller value of $H_{c2}(0)$ for nonzero g values, i.e., $g=0.4$ leads to an $\approx$25% reduction in $H_{c2}(0)$ obtained from the semiclassical (WHHM) theory. Equation (17) provides a good fit to the $H_{c2}(T)$ data between T_c and $T=0.75T_c$ for both Y and Lu samples. The fit is better for the Lu sample than for the Y sample. Figure 10 shows the fit for $H_{c2}^{\rm pl}(T)$ of the Lu sample, and a similarly good fit is obtained for $H_{c2}^{\rm pr}(T)$ of the Lu sample with $g^{\rm pl}\approx 0.40$ and $g^{\rm pr}=0.38$, respectively. The g values for the Y sample are $g^{\rm pl}=0.58$ and $g^{\rm pr}=0.57$. The g values suggest that YNi_2B_2C is a stronger coupling superconductor than $LuNi_2B_2C$ which is also reflected in the higher value of $\lambda_{\rm tr}$ for YNi_2B_2C than for $LuNi_2B_2C$. Bahcall finds $g=0.3$ gives a good fit between $H_{c2}(T)$ for Nb_3Sn (Ref. 7) and Eq. (17) near T_c. It may be noted that the g value for lead, which is a strong-coupling superconductor, is $\approx$0.4.[103] Thus, the obtained values of g for YNi_2B_2C and $LuNi_2B_2C$ place them in the strong-coupling superconductor category.

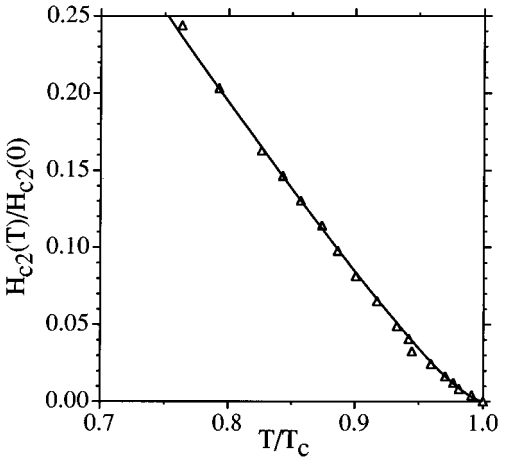

FIG. 10. Temperature dependence of $H_{c2}^{\rm pl}(T)/H_{c2}^{\rm pl}(0)$ vs T/T_c for the $LuNi_2B_2C$ single-crystal sample between $T/T_c=0.75$ and 1.0. The line drawn through the data is a least-squares fit to Eq. (17) given by Bahcall (Ref. 107) for $g=0.4$.

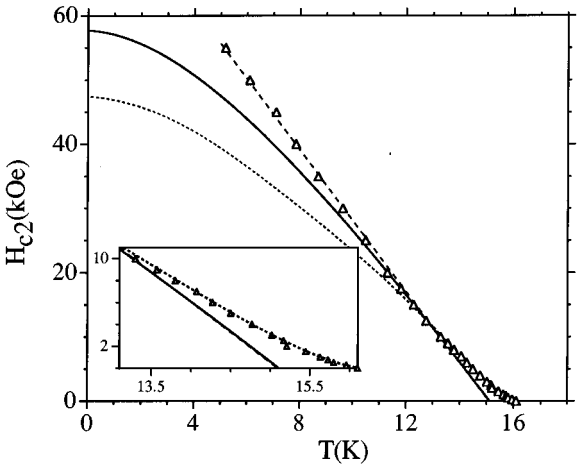

FIG. 11. Critical field $H_{c2}(T)$ as a function of temperature T for $LuNi_2B_2C$ single-crystal samples. The prediction of Bahcall's calculations (Ref. 107) with $g=0.4$ is represented by the dotted curve. The solid curve is the prediction of the WHHM theory in the ''dirty'' limit. The dashed line is a straight line fit to the low-temperature data used to determine the parameters for WHHM theory. The inset expands the data near T_c.

Neither this theory nor the WHHM theory adequately describes the low-temperature behavior of $H_{c2}(T)$ for either of the samples as shown in Fig. 11. Although Bahcall's theory can be fit well to the high-temperature data where the positive curvature is seen, it predicts much too low a value for $H_{c2}(T)$ at lower temperatures. It is difficult to compare the $H_{c2}(T)$ data with the WHHM theory because of the positive curvature it shows near T_c, but, if the linear region of the $H_{c2}(T)$ data is extrapolated to T_{ca} and that slope used with the WHHM theory with $T_c=T_{ca}$, the resulting curve does not describe the lower temperature data as shown in Fig. 11. The figure clearly shows that the data lie on a straight line for $T<0.8T_c$ with no indication of deviation from linear behavior down to $0.3T_c$. Thus, the WHHM theory does not seem to be valid for these samples, even at lower temperatures, i.e., the straight line behavior of the data extends far beyond the region predicted by the WHHM theory. Lower temperature measurements should be made to determine the

extent of the breakdown of theory. Since Bahcall's theory[107] predicts a lower value of $H_{c2}(0)$ than does WHHM (about 23% lower for $g \approx 0.4$) it gives even a worse description at low temperatures. Thus, the theoretical ideas need to be modified in order to explain the experimental $H_{c2}(T)$ results reported here for YNi_2B_2C and $LuNi_2B_2C$ single crystals as well as those reported in the literature for other RNi_2B_2C superconducting compounds.

SUMMARY AND CONCLUSIONS

Measurements of the in-plane resistivity, in-plane thermopower, and upper critical field $H_{c2}(T)$ for YNi_2B_2C and $LuNi_2B_2C$ single crystals are reported. The in-plane resistivities for both samples vary approximately linearly at room temperature and follow a power law T^p at low temperatures with $p=2.2$ and 2.0, respectively, for the Y and Lu samples for $1.25T_c < T < 0.1\Theta_D$, similar to that followed by the strong-coupling and disordered A-15 compounds. The analysis of the resistivity data shows that these are moderately strong-coupling superconductors with the electron-phonon parameter $\lambda_{tr}=1.2$ and 0.97 for the Y and Lu samples, respectively. These values agree within 10–15 % with the values obtained from the analysis of the specific-heat and superconducting transition temperature data. The Y sample shows a large and positive transverse magnetoresistance, and is about 8% ($H=45$ kOe) at 15 K. That for the Lu sample is comparable (about 7% at 20 K), not near the 40% value reported for a polycrystalline sample.[55] A very small anisotropy ($\approx 2\%$) is observed with respect to the direction of H parallel and perpendicular to the c axis of the crystal.

The absolute thermopower (S) as well as the slope $(dS/dT)_{RT}$ of both samples are negative from RT to T_c and their magnitudes are approximately the same. S varies approximately linearly near RT and an extrapolation to $T=0$ gives large intercepts (≈ -4 μV/K) suggesting that in the absence of superconductivity there would be a low-temperature ''knee'' in $S(T)$ similar to that produced by electron-phonon renormalization. The ratio of the highest value of S/T at low temperatures to that at RT indicates the possibility of a large value of the electron-phonon parameter $\lambda(0)$, 6–8, which does not agree with the values of $\lambda(0)$ estimated from other experimental measurements. The usual phonon-drag peak shape is not observed, but a large contribution beyond the diffusion term exists. The temperature dependence of this contribution is similar to the phonon-drag contribution to the thermopower calculated for high-temperature superconductors (layered compounds) by Trodahl.[86]

The resistive transition curves in external magnetic field applied parallel and perpendicular to the c axis have been used to determine the upper critical field $H_{c2}(T)$. A small current dependence of $H_{c2}(T)$ has been observed in the samples which becomes larger at lower temperatures. $H_{c2}(T)$ for both samples shows a positive curvature near T_c which is not predicted by the WHHM theory. Various characteristic superconducting parameters have been calculated using the slope of $H_{c2}(T)$ vs T in the temperature region $T < 0.85T_c$. The upper critical field $H_{c2}(0)$ is typically 65 kOe, the Ginzburg-Landau coherence length ≈ 70 Å, the BCS coherence length and the penetration depth $\lambda(0)$ are ≈ 65 and ≈ 1200 Å for YNi_2B_2C, and ≈ 35 and ≈ 800 Å for $LuNi_2B_2C$, respectively. These numbers show that both samples are type-II superconductors. A severe disagreement between the BCS coherence lengths obtained from the measurement of the energy gap[9] and from $H_{c2}(T)$ measurements is found which can be only somewhat improved if one takes into account the renormalization of the Fermi velocity. $H_{c2}(T)$ for the Y sample does not show any anisotropy with respect to the c axis in agreement with the magnetization measurements,[8] while the Lu sample shows an anisotropy in $H_{c2}(T)$ of about $\approx 10\%$. The positive curvature of $H_{c2}(T)$ near T_c can be explained in terms of a recent theoretical idea of Bahcall[107] who has calculated $H_{c2}(T)$ directly by generalizing pairing between plane waves to pairing between many electronic Landau levels in presence of a magnetic field within the framework of the BCS-Gor'kov theory. A fit between the experimental data near T_c and the theoretical expression given by Bahcall gives the BCS coupling constant $V_o N(0) \approx 0.4$ for $LuNi_2B_2C$ and ≈ 0.6 for YNi_2B_2C. These values place both samples in the category of strong-coupling superconductors. On the other hand, the low-temperature behavior (down to 4.2 K) of $H_{c2}(T)$ for both samples cannot be explained by either the WHHM or Bahcall's theory. $H_{c2}(T)$ at low temperature greatly exceeds that expected from the slope of the region linear in temperature and presents an interesting challenge to the theory.

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