Hall-effect in LuNi$_2$B$_2$C and YNi$_2$B$_2$C borocarbides: a comparative study

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The Hall effect in LuNi$_2$B$_2$C and YNi$_2$B$_2$C borocarbides has been investigated in normal and superconducting mixed states. The Hall resistivity $\rho_{xy}$ for both compounds is negative in the normal as well as in the mixed state and has no sign reversal below $T_c$ typical for high-$T_c$ superconductors. In the mixed state the behavior of both systems is quite similar. The scaling relation $\rho_{xy} \sim \rho_{xx}^\beta$ ($\rho_{xx}$ is the longitudinal resistivity) was found with $\beta = 2.0$ and 2.1 for annealed Lu- and Y-based compounds, respectively. The scaling exponent $\beta$ decreases with increasing degree of disorder and can be varied by annealing. This is attributed to a variation of the strength of flux pinning. In the normal state weakly temperature dependent Hall coefficients were observed for both compounds. A distinct nonlinearity in the $\rho_{xy}$ dependence on field $H$ was found for LuNi$_2$B$_2$C in the normal state below 40 K, accompanied by a large magnetoresistance (MR) reaching $+90\%$ for $H = 160$ kOe at $T = 20$ K. At the same time for YNi$_2$B$_2$C only linear $\rho_{xy}(H)$ dependences were observed in the normal state with an approximately three times lower MR value. This difference in the normal state behavior of the very similar Lu- and Y-based borocarbides seems to be connected with the difference in the topology of the Fermi surface of these compounds.

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I. INTRODUCTION

Investigation of the Hall effect in the normal and superconducting mixed states gives an important information about the electronic structure and the vortex dynamics of the investigated materials. The nature of both of them is not settled yet for the superconducting quaternary borocarbides $R$Ni$_2$B$_2$C ($R=\text{Y, rare earth}$) studied. Despite the fact that the borocarbides have a strongly anisotropic, layered tetragonal crystal structure, their electronic properties indicate three-dimensionality showing only a little anisotropy. Borocarbides based on magnetic rare earths show a wide range of competing effects between superconductivity and magnetism, see, e.g., Ref. 10. One of the interesting features of some borocarbides ($R$=Er, Lu, Y) is the vortex lattice (VL) with unusual square symmetry observed in the mixed state for magnetic fields $H$ directed along tetragonal $c$-axis at $H \gtrsim 1$ kOe. Square symmetry of VL can be connected with the anisotropy of the upper critical magnetic field $H_{c2}(T)$ observed in the $ab$-plane for LuNi$_2$B$_2$C. Practically no anisotropy of $H_{c2}(T)$ was found for YNi$_2$B$_2$C although this compound is very similar to LuNi$_2$B$_2$C. The reason for the difference in the behavior of these two borocarbides is still unclear.

To our knowledge, no data on the Hall effect for LuNi$_2$B$_2$C and only few for some other borocarbides are known so far. Namely, normal state Hall coefficients $R_H$ were found to be negative and only weakly temperature dependent for polycrystalline borocarbides based on $R = \text{Y}, \text{Ho}, \text{La}, \text{Er}$ and Gd. A negative but strongly temperature dependent Hall coefficient was found for the heavy-fermion-like compound YbNi$_2$B$_2$C. No sign reversal of the Hall resistivity $\rho_{xy}$ in the mixed state typical for high-$T_c$ superconductors was observed in YNi$_2$B$_2$C prepared under high pressure. The mixed-state Hall effect was not yet systematically studied for borocarbides. Since the mixed state Hall effect may depend on the peculiarities of the vortex lattice, it is of interest to investigate it for LuNi$_2$B$_2$C and YNi$_2$B$_2$C with an anomalous square VL. Also it is interesting to compare the results on the Hall effect in the normal and in the mixed states for Lu- and Y-based borocarbides, having substantially different types of anisotropy of the upper critical field.

The Hall effect in the superconducting mixed state, which was studied up to now mainly for high-$T_c$ superconductors, has recently attracted a considerable attention and should be described in more detail. The magnetic field penetrates into a type-II superconductor by magnetic induction, the motion of vortex lines induces a macroscopic electric field $\mathbf{E}$ given by the relation:

$$\mathbf{F} = \frac{1}{c} \mathbf{j} \times \mathbf{B},$$

(1)

where $\mathbf{j}$ is the transport current density and $\mathbf{B}$ is the magnetic induction. The motion of vortex lines induces a macroscopic electric field $\mathbf{E}$ given by the relation:

$$\mathbf{E} = \frac{1}{c} \mathbf{v}_L \times \mathbf{B},$$

(2)
where $v_L$ is the velocity of vortex motion. The vortex motion along the Lorentz force (parallel to $j$) gives the dissipative field ($E \parallel j$) and leads to the flux-flow resistivity. At the same time the vortex motion along the direction of transport current results in the Hall electric field ($E \perp j, B$). Thus, the Hall effect is a sensitive test of vortex dynamics in the investigated material. On the other hand, normal carriers in the vortex core, experiencing a Lorentz force, can also give a contribution to the mixed state Hall effect by the usual mechanism.

Two unexpected effects have been experimentally found for high-$T_c$ superconductors: (i) a sign reversal of the Hall resistivity $\rho_{xy}$ below $T_c$ and (ii) a striking scaling relationship between $\rho_{xy}$ and the longitudinal resistivity $\rho_{xx}$ in the superconducting transition region, $\rho_{xy} \sim \rho_{xx}^\beta$.

Sign reversal of the Hall resistivity $\rho_{xy}$ has been observed experimentally over a range of temperatures and magnetic fields below $T_c$ for several types of high-$T_c$ superconductors, e.g., YBa$_2$Cu$_3$O$_{7-\delta}$, Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$, Tl$_2$Ba$_2$CaCu$_2$O$_{8+\delta}$, $L_2-x$Ce$_x$CuO$_4$ ($L = \text{Nd, Sm}$), YBa$_2$Cu$_3$O$_{7-y}$, PrBa$_2$Cu$_3$O$_{7-y}$ superlattices (YBCO, BSCCO, TBCCO, LCCO, YBCO/ PBCO respectively), as well as for some conventional superconductors: In-Pb alloys, V, Nb, Pb (see Ref. 29), Mo$_3$S$_4$, $2H$ – NbSe$_2$. This Hall effect anomaly cannot be understood within the framework of the classical Bardeen-Stephen and Nozières-Vinen theories of vortex motion predicting the same sign of the Hall voltage for both the superconducting and the normal state. Recently several models based on different approaches have been proposed for the description of this effect (see, e.g., Refs. 34–39 and references therein), but the origin of this phenomenon remains a controversial problem. Meanwhile, the sign reversal of $\rho_{xy}$ below $T_c$ is expected to be a universal property, but its existence seems to depend crucially on the peculiarities of the electronic structure. Experimentally the pronounced influence of the doping level on the sign of the Hall voltage close to $T_c$ was observed for various high-$T_c$ cuprates. The sign reversal of the Hall effect disappears for heavily underdoped and strongly overdoped regimes.

Scaling behavior, $\rho_{xy} \sim \rho_{xx}^\beta$, in the superconducting mixed state was observed for the first time by Luo et al. for an YBCO thin film ($\beta = 1.7$). The same relationship was also found for several types of high-$T_c$ superconductors: YBCO single crystals ($\beta \approx 1.7$), BSCCO ($\beta \approx 2$), TBCCO ($\beta \approx 2$), LCCO ($\beta \approx 0.8$), and YBCO/PBCO superlattices ($\beta \approx 1.7$). In a recent investigation of superconducting indium thin films scaling with $\beta$ value $2 \pm 3$ was observed. Theoretically, Dorsey and Fisher have interpreted the observed behavior in the framework of glassy scaling near a vortex-glass transition. In their model, assuming the existence of a vortex-glass transition in a three-dimensional vortex system, the region where scaling behavior should be observed is restricted to a narrow region near the vortex-glass transition. However, it should be mentioned that scaling behavior was observed far beyond the possible vortex-glass transition. A phenomenological model, based on an entirely different approach, has been proposed by Vinokur et al. who have calculated the effect of pinning on the Hall resistivity. In their model the Hall conductivity $\sigma_{xy} \approx \rho_{xy}/\rho_{xx}^2$ ($|\rho_{xy}| \ll \rho_{xx}$) is independent of disorder and the scaling law $\rho_{xy} \sim \rho_{xx}^\beta$ is believed to be a general feature of any vortex state with disorder-dominated dynamics. Therefore, the value of $\beta = 2$ should not depend on the degree of disorder. On the other hand, Wang, Dong, and Ting (WDT) recently modified their earlier work, based on the normal core model proposed by Bardeen and Stephen. They developed a theory for the Hall effect including both pinning and thermal fluctuations. In the WDT theory scaling and sign reversal of $\rho_{xy}$ are explained by specially taking into account the backflow current of vortices due to pinning. Thereby, $\beta$ changes from 2 to 1.5 as the pinning strength increases. Controversial experimental results have been reported on the influence of disorder on the mixed-state Hall effect. For irradiated YBCO samples, $\beta$ was found to be $1.5 \pm 0.1$ compared to $2 \pm 0.2$ for unirradiated ones, in accordance with WDT (see also Refs. 26–28). However, no influence of disorder on the scaling exponent was observed for TBCCO irradiated by heavy ions. In that case, $\beta = 1.85$ holds even after irradiation (see also Ref. 40). A strong influence of pinning on the Hall effect in the mixed state was observed for YBCO single crystals. At the same time it was pointed out in Ref. 40 that pinning effects cannot be the only reason for the Hall anomaly for YBCO single crystals. All these controversial results show that more work is necessary for better understanding of the mixed-state Hall effect and the influence of disorder on it.

Very recently Wang and Mak 49 have interpreted the anisotropy of $H_{c2}(T)$ observed for borocarbenes in terms of a three-dimensional version of $\delta_{xx}$-$\delta_{yy}$ superconductivity. Possible $d$ wave nature of superconductivity for borocarbenes gives an additional motivation for further study of their electronic properties. In the present study we have investigated the Hall effect in the normal as well as in the mixed state for LuNi$_2$B$_2$C and YNi$_2$B$_2$C compounds prepared under the same conditions. The results for LuNi$_2$B$_2$C have been briefly reported in Ref. 53.

II. EXPERIMENTAL DETAILS

Polycrystalline LuNi$_2$B$_2$C (in the following denoted as PC AN) and YNi$_2$B$_2$C samples were prepared by arc-melting in Ar atmosphere and subsequent careful annealing at 1100 °C, as described in more detail in Ref. 54. The phase purity of the samples was checked by X-ray diffraction on a Philips PW 1820 system with CuK$_\alpha$ radiation. The reflections revealed practically a single phase. The lattice parameters were $a=3.464$ Å, $c=10.635$ Å for LuNi$_2$B$_2$C and $a=3.528$ Å, $c=10.546$ Å for YNi$_2$B$_2$C.
Bar-shaped samples were cut from the ingots. Typical dimensions of the samples were $3 \times 1 \times 0.3$ mm$^3$. Hall contacts with typical misalignment of less than 0.1 mm were used (this is essential because the maximum of the Hall voltage does not exceed several tens of nanovolts). At each point the Hall voltage was measured for two inverse directions of the magnetic field. Most measurements of the Hall effect and of the ac-susceptibility were done in magnetic fields up to 50 kOe using a Lake Shore model 7225 susceptometer with Keithley 182 nanovoltmeter and PAR-5209 Lock – in amplifier. Some measurements in magnetic fields up to 160 kOe were performed using an Oxford Teslatron system. The values of electrical current were 10-20 mA for dc measurements and 1 mA for ac ones. The magnetoresistance (MR) was measured by the standard four-probe method. For comparison some measurements were performed on an unannealed LuNi$_2$B$_2$C sample (denoted as PC UNAN) which has a considerably higher degree of disorder.

### III. RESULTS AND DISCUSSION

#### A. Resistivity and upper critical magnetic field

The experiments in the present work have been performed on polycrystalline samples (to the best of our knowledge, no data on the Hall-effect for single crystalline borocarbides are reported so far). For characterization of our samples the results on resistivity, upper critical field and magnetoresistance for them will be compared with data known for single crystals.

The temperature dependencies of the longitudinal resistivity $\rho_{xx}(T)$ for the annealed LuNi$_2$B$_2$C and YNi$_2$B$_2$C samples are depicted in Fig. 1. The $\rho_{xx}(T)$ curves obtained at $H=50$ kOe and 160 kOe are also shown. The resistivity of both compounds exhibits a weak temperature dependence just above superconducting transition temperature $T_c$. Both samples have a rather sharp superconducting transition, a low resistivity at low temperatures and high values of $T_c$ and the residual resistance ratio RRR ($\text{RRR} = \rho_{xx}(300 \text{ K})/\rho_{xx}(17 \text{ K})$). These parameters are compared in Table II with those reported recently for LuNi$_2$B$_2$C and YNi$_2$B$_2$C single crystals. It should be pointed out, that the resistivity of borocarbides is practically isotropic. The small difference ($\approx 2\%$) between the in-plane resistivity $\rho_{xx}$ and the resistivity along the $c$-axis $\rho_c$ observed for YNi$_2$B$_2$C single crystals at $T=15\div 300$ K is well within the experimental uncertainty. Thus, it is reasonable to compare the values of resistivity for polycrystalline and single crystalline borocarbides.

More precisely, the value of $\rho_{xx}(17 \text{ K})$ for the unannealed polycrystalline LuNi$_2$B$_2$C sample is 2.7 $\mu\Omega$cm, which is close to that for LuNi$_2$B$_2$C single crystals (1.6$\div$2.5 $\mu\Omega$cm, Refs. [5,6,7]), see Table II. The value of RRR is 41 for our LuNi$_2$B$_2$C sample, which is significantly higher than those observed for single crystals (23$\div$27, see Refs. [5,6,7]). Also the value of $T_c=16.7$ K is slightly higher than that reported for single crystals (15.8$\div$16.5 K, see Refs. [5,6,7]). The width of the superconducting transition $\Delta T_c$ (determined from zero field ac-susceptibility curve, see Fig. 3) is 0.27 K which is close to the values 0.2$\div$0.25 K typical for single crystals. The PC UNAN LuNi$_2$B$_2$C sample has a lower $T_c$ (14.7 K), a wider superconducting transition and a more than one order of magnitude higher value of $\rho_{xx}(17 \text{ K})$.

For the polycrystalline YNi$_2$B$_2$C sample the values of $\rho_{xx}(17 \text{ K})$, RRR, $T_c$ and $\Delta T_c$ are also comparable with the results reported for YNi$_2$B$_2$C single crystals [5,6,7]. At the same time our YNi$_2$B$_2$C sample has an approximately two times lower RRR value than LuNi$_2$B$_2$C prepared under the same conditions.

The results for the resistivity $\rho_{xx}(300 \text{ K})$ collected in Table II show surprisingly large discrepancies even for the single crystals. Thus an intersection of the $\rho_{xx}(T)$ dependences can be recognised for the two YNi$_2$B$_2$C single crystals [5,6,7]. These facts could be naturally understood taking into account the large uncertainty in geometrical factor used to determine the value of $\rho_{xx}$ from the experimentally measured resistance especially for small single crystals. To clarify this, linear $\rho_{xx}(T)$ dependences for two imaginary samples of the same imaginary compound with different RRR values (10 and 4 for samples A and B, respectively) have been plotted in Fig. 2. Mattissen’s
rule is expected to be valid for them, i.e. the difference in resistivities does not depend on temperature. Let us suppose, that the measurements of $\rho_{xx}(T)$ give the "true" values for the sample A and underestimate it by 1/3 of its "true" value for the sample B (e.g., due to the uncertainty in the dimensions of the sample, final width of the contacts, etc.). In that case for the sample B the obtained ("measured") $\rho_{xx}(T)$ curve (denoted as B' on Fig. 2 and having the same value of RRR as the curve B) will cross the curve for the sample A. This example illustrates that an uncertainty of geometrical factor of $\approx 20 \pm 30\%$ could explain the difference in room temperature resistivities and surprising intersection of the $\rho_{xx}(T)$ dependences for the two YNi$_2$B$_2$C single crystals. A strong support for this explanation is that, for single crystals of different quality known, one would expect close resistivity values rather at room temperature than at low temperatures. We conclude that the quality of the borocarbide samples compared in Table I can be mainly judged from the RRR data, whereas resistivity values are strongly influenced by the uncertainty in the geometrical factor used for determination of $\rho_{xx}$. Noteworthy, both room and low temperature resistivities for our LuNi$_2$B$_2$C sample are close to those reported for YNi$_2$B$_2$C single crystal which has a similar value of RRR as our policrystalline LuNi$_2$B$_2$C sample (see Table I).

Superconducting transitions determined from ac-susceptibility measurements are shown in Fig. 3 for different magnetic fields. Temperature dependences of upper critical magnetic fields $H_{c2}(T)$ are depicted on Fig. 3 for the annealed and unannealed LuNi$_2$B$_2$C samples. (The value of $H_{c2}$ was determined, similar as in Ref. 54, by the extrapolation of the ac-susceptibility curve to zero susceptibility value, see Fig. 3A.) For comparison the data from Ref. 54 for a LuNi$_2$B$_2$C single crystal (SCR) with $H \parallel <110>$ are also shown. The upward curvature (UC) in the $H_{c2}(T)$ dependence is clearly visible near $T_c$. Note that, in accordance with Ref. 54, the UC region is more pronounced and $|\partial H_{c2}/\partial T|$ is higher for the annealed PC AN sample. This suggests that this sample is close to the clean limit in terms of traditional theory of type-II superconductors. It is of interest to compare $|\partial H_{c2}/\partial T|$ values for different samples determined from the approximately linear parts of the $H_{c2}(T)$ dependences ($H=20\div50$ kOe). The value of $|\partial H_{c2}/\partial T|=6.8$ kOe/K, obtained for the annealed LuNi$_2$B$_2$C sample, is in good agreement with those determined for single crystals: 6.7 kOe/K (calculated by arithmetic averaging of $|\partial H_{c2}/\partial T|$ data reported in Ref. 54 for three directions of $H$, $\parallel <100>$, $\parallel <110>$, and $\parallel <001>$), 6.4 kOe/K (reported for $H \parallel <001>$ for another single crystal in Ref. 55) and 6.2 kOe/K (calculated by arithmetic averaging of the data reported in Ref. 54 for $H \parallel <100>$ and $\parallel <001>$). The $H_{c2}(T)$ dependence for our YNi$_2$B$_2$C sample is similar to that observed for the LuNi$_2$B$_2$C PC AN one and also is in good agreement with the results reported for YNi$_2$B$_2$C single crystals.

The value of $|\partial H_{c2}/\partial T|$ for our YNi$_2$B$_2$C sample is 6.0 kOe/K, see Table I.

The low resistivity just above $T_c$, high RRR and $T_c$ values, narrow superconducting transitions, pronounced UC region in the $H_{c2}(T)$ dependences and X-ray diffraction results give evidence for a good quality of our annealed LuNi$_2$B$_2$C and YNi$_2$B$_2$C samples.

FIG. 2. Temperature dependence of the resistivity for two imaginary samples (A and B) with different RRR values (10 and 4, respectively). Line B' corresponds to the underestimated by 1/3 of $\rho$ for sample B and has the same value of RRR as line B. See text for details.

FIG. 3. Temperature dependence of the real part of the ac-magnetic susceptibility for the LuNi$_2$B$_2$C (A) and YNi$_2$B$_2$C (B) samples in several magnetic fields. The upper critical field $H_{c2}$ was determined by linear extrapolation of the ac-susceptibility curve to zero susceptibility value, as shown in the upper part of the figure. Lines are guides for the eye.
FIG. 4. Temperature dependence of the upper critical field $H_{c2}$ for three LuNi$_2$B$_2$C samples. Open symbols - the results of Ref. 8 for single crystal. Solid symbols - the results for PC AN and PC UNAN polycrystalline samples (see text). Lines are guides for the eye.

B. Normal state Hall effect

The temperature dependences of the Hall resistivity, $\rho_{xy}(H)$, for LuNi$_2$B$_2$C PC AN and PC UNAN samples as well as for YNi$_2$B$_2$C sample in the normal and in the mixed states are shown in Fig. 5. First of all it should be emphasized that the Hall resistivity of all the samples is negative at $3.3 \text{ K} \leq T \leq 300 \text{ K}$, and has no sign reversal below $T_c$.

In the normal state, a pronounced nonlinearity in the $\rho_{xy}(H)$ dependences is evident at $T < \sim 40 \text{ K}$ for both LuNi$_2$B$_2$C samples. Linear $\rho_{xy}(H)$ dependences extrapolated from the low fields region are also shown in Fig. 5 by dashed lines. The deviation from linear $\rho_{xy}(H)$ dependence increases with lowering temperature. The anomaly is more pronounced for the annealed sample, although it is also distinctly seen for the unannealed one. More clearly the nonlinearity in the $\rho_{xy}(H)$ dependences can be seen in the insets of Fig. 5A and B where some results obtained in high magnetic fields (up to 160 kOe) are presented. It should be underlined that no nonlinearity in the $\rho_{xy}(H)$ dependence was observed for our YNi$_2$B$_2$C sample (see the inset of Fig. 5C). Earlier, linear $\rho_{xy}(H)$ dependences have been reported for YNi$_2$B$_2$C and YbNi$_2$B$_2$C samples prepared under high pressure. No indications of nonlinear $\rho_{xy}(H)$ dependences have been observed for Y-, Ho-, Gd- and La-based borocarbides. Thus, the nonlinearity in the $\rho_{xy}(H)$ dependence, found for LuNi$_2$B$_2$C samples with essentially different quality, can be considered as an intrinsic and specific property of the Lu-based borocarbide.

A nonlinear and even nonmonotonous $\rho_{xy}(H)$ dependence has been found earlier for the heavy fermion superconductor UBe$_1$_$_3$ and have been interpreted in the framework of a two-band model. In this model, at low fields, the light carriers with high mobilities give the prevalent contribution to the Hall effect, whereas at high fields the contribution of the heavier carriers having lower
mobilities is more significant. Very recently a similar two-band model has been used to interpret the results on the transport properties of Nd$_{2-x}$Ce$_x$CuO$_4$ epitaxial thin films. In an entirely different type of multiband models the existence of at least two bands with significantly different Fermi-velocities was found to be very essential for the quantitative description of $H_{c2}(T)$ curves with sizable UC for the Lu- and Y-based borocarbides. Several groups of carriers with different effective masses have been directly observed for YNi$_2$B$_2$C in dHvA experiments. Thus, some kind of a two-band model may be applicable for understanding of the nonlinear $\rho_{xy}(H)$ dependence found by us for LuNi$_2$B$_2$C borocarbide.

In Fig. 6 the temperature dependences of the Hall coefficients $R_H(T) = \rho_{xy}(T,H)/H$ at $H=50$ kOe are shown for the annealed LuNi$_2$B$_2$C and YNi$_2$B$_2$C samples. Below $\sim 60$ K the $R_H(T)$ dependence for LuNi$_2$B$_2$C shows a considerable deviation from the dotted line describing $R_H(T)$ at $H=50$ kOe for higher temperatures. This deviation is obviously connected with the nonlinearity in $\rho_{xy}(H)$ curves shown in Fig. 3A. At low temperatures the values of the Hall coefficient on the dotted line in Fig. 3 (obtained by the extrapolation of the high temperature $R_H(T)$ curve) coincide with the values of the low-field Hall coefficient, calculated at low temperatures using the low-field asymptotes for the $\rho_{xy}(H)$ curves (shown in Fig. 3A by dashed line). Only weak temperature dependences were observed for the low field Hall coefficients of LuNi$_2$B$_2$C and YNi$_2$B$_2$C borocarbides. This is in agreement with the observation of a weak $R_H(T)$ dependence for YNi$_2$B$_2$C in Refs. 15, 17. Weak $R_H(T)$ dependences were reported also for La-, Ho- and Gd-based borocarbides. Below $\sim 60$ K, the $R_H(T)$ curve obtained for LuNi$_2$B$_2$C at $H=50$ kOe exhibits a pronounced temperature dependence connected with the nonlinearity found for $\rho_{xy}(H)$ at low temperatures. Noteworthy, that a strong decrease of the Hall coefficient was found with increasing temperature for YbNi$_2$B$_2$C borocarbide having moderate heavy-fermion-like behavior.

Values of $R_H$ obtained in this work for LuNi$_2$B$_2$C and YNi$_2$B$_2$C are comparable with those earlier reported for YNi$_2$B$_2$C, but they are about five times (LuNi$_2$B$_2$C) or ten times (YNi$_2$B$_2$C) smaller than the value resulting from band structure calculations for LuNi$_2$B$_2$C (3 \cdot 10^{-5} m^2/C=3\cdot10^{-4} \mu\Omega cm/Oe). These deviations may be caused by correlation effects in borocarbides. The estimation of the carrier density from the $R_H$ value at $T=300$ K, by using a single band model which is a rough approximation, gives 1.5 and 2.4 carriers per unit cell for Lu- and Y-based borocarbides, respectively. (The estimation of the carrier density for YNi$_2$B$_2$C prepared under high pressure gives 0.6 carriers per unit cell, i.e. about four times smaller than present result and values reported in Refs. 15, 17. Probably this difference is connected with high sensitivity of the Hall coefficient for method of sample preparation.)

C. Normal state magnetoresistance

In Fig. 4 the field dependence of the longitudinal resistivity $\rho_{xx}(H)$ is shown for the annealed LuNi$_2$B$_2$C and LuNi$_2$B$_2$C samples. The values of magnetoresistance...
for the annealed LuNi$_2$B$_2$C sample at $T = 20$ K are as high as 25% and 90% for $H = 50$ kOe and 160 kOe, respectively (see also Fig. 1). At the same time considerably smaller values of MR were observed at these fields (10% and 33%, respectively) for the YNi$_2$B$_2$C sample, prepared under the same conditions as the Lu-based one, see Table 1. It should be underlined, that a magnetoresistance of only $\approx 7.3\%$ was observed, at $H=45$ kOe and $T = 20$ K, for a LuNi$_2$B$_2$C single crystal with RRR=25 ($H$ parallel to the tetragonal $c$-axis) i.e. the value of MR for our LuNi$_2$B$_2$C polycrystalline sample is about 3.5 times higher, than that of this single crystal. The value of MR (7.5%) reported in Ref. 9 for YNi$_2$B$_2$C single crystal with RRR=18 ($H \parallel c$) is comparable with that found for investigated YNi$_2$B$_2$C sample. High values of MR can be considered as an additional indication of the high quality of our annealed samples, because the value of MR for the unannealed sample is approximately 40 times smaller (see Table 1). It should be also noted that the impurities of magnetic rare earths, the concentration of which could be greater in Lu-based sample than in Y-based one, can not lead to the increase of MR, because the introduction of magnetic ions to nonmagnetic substance (e.g., borocarbide lattice) gives the negative contribution to MR due to decrease of spin-disorder scattering in a magnetic field, see, e.g., Refs. 5, 9.

A possible reason for the very large positive MR in LuNi$_2$B$_2$C and for the significantly larger MR of the polycrystalline sample compared to the single crystal in Ref. 9 is the formation of open orbits on the Fermi surface of that compound for $H \perp c$. (In principle the possibility of the formation of open orbits for borocarbides was pointed out in band structure calculations. In Ref. 9, e.g., it was claimed that one part of the Fermi surface is a cylinder along the $c$-axis.) It is well known that open orbits can lead to large values of MR $\propto H^2$, whereas closed orbits should give rise to saturation of magnetoresistance for large $H$. In that case, for polycrystals, the averaging of MR should lead to a practically linear $\rho_{xx}(H)$ dependence (so called Kapitza’s law). In accordance with this, the observed $\rho_{xx}(H)$ dependences for our LuNi$_2$B$_2$C PC AN sample follow approximately a linear law, see Fig. 1. The MR($H$) dependence for polycrystals, in the case of open orbits for some directions of $H$, should be stronger than that observed for single crystals for $H \parallel c$ where only closed orbits could be expected. Therefore, the significantly larger MR found for the LuNi$_2$B$_2$C polycrystals, in comparison with that observed for the single crystal for $H \parallel c$, can be considered as an indication for the open-orbits formation in LuNi$_2$B$_2$C for $H \perp c$. Investigation of the MR in high fields for LuNi$_2$B$_2$C single crystals with the two configurations (i) $J \parallel c$ and $H \perp c$ and (ii) $J \perp c$ and $H \parallel c$ are necessary to check this conclusion.

The nonlinear $\rho_{xy}(H)$ dependence and the large MR, found in this study, as well as the anisotropy of $H_{c2}$ in the $ab$-plane earlier reported for LuNi$_2$B$_2$C, may be caused by the same reason, namely, by peculiarities of its electronic structure. It should be underlined, that all these anomalies are absent for YNi$_2$B$_2$C. (For YNi$_2$B$_2$C a linear $\rho_{xy}(H)$ dependence and a substantially smaller MR can be seen in Fig. 1 and only a very small anisotropy of $H_{c2}(T)$ were reported in Refs. 8, 9). The differences in the properties of these very similar compounds should be connected with difference between their electronic structure. As has been noted in Ref. 9 the Fermi surface topology of the borocarbides is very sensitive to the position of the Fermi level, which may be slightly different for the two cases, Lu and Y, due to, e.g., different lattice constants. From the obtained results the formation of open orbits seems to be easier in case of LuNi$_2$B$_2$C in comparison with YNi$_2$B$_2$C. Nevertheless only the comparative study of LuNi$_2$B$_2$C and YNi$_2$B$_2$C single crystals (e.g., investigation of the angular dependence of MR in high fields) can give definitive verification of the proposed model.

Theoretically, it is more convenient to describe the behavior of the Hall effect in terms of the conductivity tensor rather than by the resistivity one, see, e.g., Ref. 9. As shown in the inset of Fig. 4, the nonlinearity in the dependence on magnetic field of the Hall conductivity, $\sigma_{xy}(H)$, in the normal state for LuNi$_2$B$_2$C is even more pronounced than the nonlinearity in the Hall resistivity curve $\rho_{xy}(H)$ ($\sigma_{xy} \propto \rho_{xy}/\rho_{xx}$, $\rho_{xx} \gg |\rho_{xy}|$). It is interesting to note that $\sigma_{xy}$ for LuNi$_2$B$_2$C becomes practically independent of the magnetic field for $H = 80\div 160$ kOe, at $T = 4.5\div 20$ K (see Fig. 4A). The nonlinear $\rho_{xy}(H)$ dependence and the large MR of LuNi$_2$B$_2$C are probably closely connected and result in a practically constant $\sigma_{xy}(H)$ at high magnetic fields. The reason why $\sigma_{xy}$ is independent of $H$ for high fields, resulting in $\rho_{xy} \sim \rho_{xx}^2$ in the normal state, is not yet understood. (It is noteworthy that $\rho_{xy} \sim \rho_{xx}^2$ scaling in the normal state was earlier observed for the superconducting heavy fermion compound UBe$_{13}$ 8). At the same time the Hall conductivity of YNi$_2$B$_2$C has only a slight nonlinearity at $T= 20$ K (see the inset of Fig. 4B). Only at $T = 4.5$ K and 3.3 K some tendency for saturation in $\sigma_{xy}(H)$ dependences was observed in high fields.

**D. Mixed state Hall effect**

In the mixed state, the variation of the Hall resistivity with magnetic field for both compounds can be described as follows: below $T_c$ in low fields there is $\rho_{xy}=0$ as can be seen from the $\rho_{xy}(H)$ curves at, e.g., $T=10$ K (Fig. 3A-C). At higher fields (in the region close to the resistive superconducting transition) the Hall resistivity increases in the absolute value and gradually reaches the $\rho_{xy}(H)$ curve obtained in the normal state at temperatures slightly higher than $T_c$. For YNi$_2$B$_2$C the normal state $\rho_{xy}(H)$ dependence is very close to linear (see the
curve obtained at \( T = 40 \, \text{K} \) in Fig. 3C. At the same time, for both LuNi\(_2\)B\(_2\)C samples the normal state \( \rho_{xy}(H) \) dependences have a nonlinearity with negative curvature. This nonlinearity, as it was pointed out above, is more pronounced for the annealed sample, see Fig. 3A and B. The Hall resistivity curve \( \rho_{xy}(H) \) in the mixed state shifts with increasing temperature to lower magnetic fields similar to the behavior usually observed for the longitudinal resistivity curve \( \rho_{xx}(H) \). Simultaneously the \( \rho_{xy}(H) \) and \( \rho_{xx}(H) \) transitions are shown in the insets of Fig. 3 for all samples. Their comparison is discussed below.

For LuNi\(_2\)B\(_2\)C as well as YNi\(_2\)B\(_2\)C no sign reversal of \( \rho_{xy}(H) \), typical for high-\( T_c \) superconductors, was observed below \( T_c \). The sign of the Hall resistivity is negative in the mixed as well as in the normal state. It should be noted, that for high-\( T_c \) superconductors not only the hole-like materials, but also the electron-like ones usually experience sign reversal of the Hall effect (see, e.g., Ref. 25). The behavior of the two LuNi\(_2\)B\(_2\)C samples with significantly different quality (the RRR value for the PC UNAN is only \( \approx 3 \)) in the mixed state is quite similar. Also the behavior of YNi\(_2\)B\(_2\)C in the mixed state is similar to that of LuNi\(_2\)B\(_2\)C. Therefore, the absence of the sign reversal of the Hall effect seems to be an intrinsic property of the investigated borocarbides. This result has been obtained on polycrystalline samples, but, as has been discussed above, the anisotropy of the electronic properties of borocarbides is small, and the quality of our annealed samples is high. Therefore, this conclusion should remain true also for the borocarbide single crystals. (For high-\( T_c \) superconductors, having considerably higher anisotropy in electronic properties, sign reversal in the Hall effect was observed usually for both poly- and single crystalline samples of the same system.)

In order to understand the absence of sign reversal in the \( \rho_{xy} \) for the investigated borocarbides, the following physical picture of the Hall effect in the mixed state can be used: there are two contributions to the Hall conductivity \( \sigma_{xy} \) in the superconducting state:

\[
\sigma_{xy} = \sigma_n + \sigma_{sc} \tag{4}
\]

where \( \sigma_n \) is connected with normal quasiparticles that experience a Lorentz force in the vortex core (it is expected to be proportional to \( H \)) and \( \sigma_{sc} \) is an anomalous contribution connected with the motion of vortices parallel to the electrical current density \( j \). In Refs. 33 and 36 it was claimed that \( \sigma_{sc} \sim 1/H \) and could have a sign opposite to that of \( \sigma_n \). Therefore, at low magnetic fields, the \( \sigma_{sc}(H) \) term is more at higher fields \( \sigma_n(H) \) will be dominant. If \( \sigma_{sc} \) has a different sign than \( \sigma_n \), it is possible to observe a sign reversal in the Hall effect at \( T < T_c \). Eq. (4) was verified and the term \( \sigma_{sc} \sim 1/H \) was observed, e.g., for YBCO 34. For LuNi\(_2\)B\(_2\)C and YNi\(_2\)B\(_2\)C the Hall conductivity decreases with increasing \( H \), as can be clearly seen in the insets of Fig. 7 for the \( \sigma_{xy} \) vs. \( H \) curves at \( T < T_c \). At the same time it should be pointed out, that observed \( \sigma_{xy} \)
vs. $H$ dependences seem to change more rapidly, than $1/H$. A similar behavior was observed for cuprates, see, e.g., Refs. [41]. Therefore, the mechanism of the mixed-state Hall effect connected with vortex motion seems to work for borocarbides as well. In the theory the sign of the Hall effect in the mixed state is determined by the energy derivative $\partial N(0)/\partial \mu$ of the density of states $N(0)$ averaged over the Fermi surface. For a complicated Fermi surface which has electron-like and hole-like parts the signs of $\sigma_{xy}$ in the normal and in the mixed states may be different. In the phenomenological theory, based on a Ginsburg-Landau equation and its gauge invariance, the sign of the Hall conductivity is determined by $\partial ln T_c/\partial \mu$, where $\mu$ is the chemical potential.

In any case, the sign of the Hall effect in the mixed state depends on the details of the band structure (see also Ref. [8]). From our results it follows, that, contrary to the case of high-$T_c$ superconductors, the signs of $\sigma_n$ and $\sigma_{xx}$ for borocarbides are the same (see the insets of Fig. 7). This seems to be the reason for the absence of sign reversal in the Hall effect in these borocarbides.

In the mixed state two regions concerning the behavior of $\rho_{xy}$ and $\rho_{xx}$ can be distinguished. At low magnetic fields both $\rho_{xy}$ and $\rho_{xx}$ vanishes. For higher fields it is clearly seen that the scaling behavior $|\rho_{xy}| = A \rho_{xx}^\beta$ holds for all three samples (see Fig. 5). The values of $\beta$ are $2.0 \pm 0.1$ and $2.1 \pm 0.1$ for Lu- and Y-based annealed samples, respectively. It decreases to $1.7 \pm 0.1$ for unannealed LuNi$_2$B$_2$C sample having a one order of magnitude higher resistivity at $T = 17$ K. This may be connected with an increase of pinning strength for the PC UNAN due to the considerably larger concentration of defects leading to the larger resistivity of this sample. The decrease of the scaling exponent with increasing pinning strength was obtained in the WDT theory [44], taking into account the backflow current of vortices due to the effects of pinning. Another manifestation of the pinning effects, predicted by the WDT model, can be seen in the insets of Fig. 5 where the $\rho_{xy}(H)$ and $\rho_{xx}(H)$ curves in the superconducting transition region are simultaneously shown. For decreasing fields, $\rho_{xx}$ vanishes at a definitely lower values of $H$ than $|\rho_{xy}|$ for all three samples. The same behavior was described by WDT taking into account the effect of pinning. In accordance with Ref. [44], the Hall resistivity can be observed only in the flux flow regime of superconducting transition. Prior to the flux flow, the longitudinal resistivity may become finite due to flux creep at finite temperature, while the Hall resistivity is still zero. (Vortices in the flux creep regime are pinned, and they are only able to creep along the $j \times B$ direction assisted by thermal activation. Creep of flux lines in this direction, in accordance with Eq. 4, does not lead to a Hall voltage.) From the obtained results it is obvious, that pinning effects are considerably important for the mixed state Hall effect in the investigated borocarbides. However, as for high-$T_c$ cuprates (see, e.g., Refs. [40,41]) not only pinning effects will govern the mixed-state Hall effect in borocarbides.

IV. CONCLUSIONS

We have investigated the Hall effect for LuNi$_2$B$_2$C and YNi$_2$B$_2$C borocarbides in the normal and in the superconducting mixed state. A negative and only slightly temperature-dependent low field Hall coefficient was found for both compounds above $T_c$. The value of the Hall coefficient $R_H$ is about one order of magnitude smaller, than that resulting from band structure calculations. A pronounced nonlinearity in the field dependence of the Hall resistivity, $\rho_{xy}(H)$, was found for LuNi$_2$B$_2$C in the normal state below 40 K accompanied by a very large positive magnetoresistence MR. An only linear $\rho_{xy}(H)$ dependence was observed for YNi$_2$B$_2$C. The possibility of open-orbits formation on the Fermi surface for $H \perp c$ is pointed out for LuNi$_2$B$_2$C contrary to YNi$_2$B$_2$C. Measurements of the angular dependence of MR in high magnetic fields for Lu- and Y-based single crystals are necessary to check the proposed model.

In the mixed state the behavior of Lu- and Y-based borocarbides is quite similar. Scaling behavior, $\rho_{xy} \sim \rho_{xx}^\beta$, was observed but no sign reversal, typical for high-$T_c$ superconductors, was found for them. The scaling exponent $\beta$ is $2.0 \pm 0.1$ for the annealed LuNi$_2$B$_2$C sample, it decreases to 1.7 $\pm$ 0.1 for the unannealed one, which, in accordance with the WDT theory, can be attributed to the increase of the pinning strength.

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|                  | Polycrystals | Single crystals |
|------------------|--------------|-----------------|
|                  | Lu | Y | Lu | Lu | Lu | Lu | Y | Y | Y |
| $\rho_{xx}(300\text{ K})$ ($\mu\Omega\text{cm}$) | 110 | 50 | 134 | 68 | 47 | 36 | 108 | 67 | 36 |
| $\rho_{xx}(17\text{ K})$ ($\mu\Omega\text{cm}$) | 2.7 | 2.3 | 43 | 2.5 | 1.9 | 1.6 | 1.7 | 2.5 | 3.8 | 2.1 |
| RRR              | 41 | 22 | 3.1 | 27 | 25 | 23 | 43 | 18 | 17 |
| $T_c$ (K)        | 16.7 | 15.5 | 14.7 | 16.5 | 16.1 | 16.0 | 15.8 | 15.7 | 15.6 | 15.7 |
| $\Delta T_c$ (K) | 0.27 | 0.44 | 0.6 | 0.2 | 0.25 | 0.2 | 0.25 | 0.2 | 0.25 |
| $|\partial H_{c2}/\partial T|$ (kOe/K) | 6.8 | 6.0 | 5.8 | 6.4 | 6.2 | 6.7 | 7.1 | 6.1 |
| MR(20 K,50 kOe) (%) | 25 | 10 | 0.7 | 7.3$^c$ | 7.3$^c$ |
| MR(20 K,160 kOe) (%) | 90 | 33 | 2.3 | 7.5$^d$ | 7.5$^d$ |

$^a$Unanneled (PC UNAN) LuNi$_2$B$_2$C sample.

$^b$For details of the $|\partial H_{c2}/\partial T|$ determination for different samples, see text.

$^c$The value obtained at $T=20$ K and $H=45$ kOe ($H \parallel c$).

$^d$The value obtained at $T=15$ K and $H=45$ kOe ($H \parallel c$).