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Magnetoresistance study of some $RENi_2B_2C$ borocarbides

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Abstract

The influence of magnetic field up to 130 kOe on normal-state resistance was investigated at $1.5 \leq T \leq 300$ K for several $RENi_2B_2C$ compounds (*RE*=Lu, Yb, Tm, Ho) prepared using a 'high pressure–high temperature' technique. A positive magnetoresistance was observed for $LuNi_2B_2C$ based on nonmagnetic Lu^{3+}. However, for the compounds with localized magnetic moments (Ho^{3+}, Tm^{3+}, Yb^{3+}), the magnetoresistance was negative being qualitatively different for various $RENi_2B_2C$ compounds. Possible reasons for this difference are discussed. Some preliminary results on the Hall-effect of the $YbNi_2B_2C$ compound are also presented and compared with ones previously obtained for YNi_2B_2C. © 1998 Elsevier Science S.A.

Keywords: Borocarbides; Rare earth; Magnetoresistance

1. Introduction

The recent discovery [1,2] of a new quaternary superconducting system $RENi_2B_2C$ (RE=rare earth, Y) provides new possibilities for the investigation of interactions between superconductivity and magnetism [3]. The coexistence of superconductivity and magnetic ordering was observed for several borocarbides based on RE^{3+} ions with localized magnetic moments (RE=Tm, Er, Ho, Dy), see, e.g., [3,4].

$RENi_2B_2C$ exhibit a filled version of the $ThCr_2Si_2$ structure stabilized by carbon occupying positions in the RE layers [5]. The variations of superconducting transition temperature (T_c) and Neel temperature (T_N) for these compounds correlates with the de Gennes factor of a localized magnetic RE^{3+} moment [3]. Also the dependence of T_c on the size of RE^{3+} was pointed out [6]. $YbNi_2B_2C$ deviates from the observed correlations mentioned above, i.e. no superconducting and long-range magnetic ordering was observed for this compound at $T>0.34$ K [7,8]. The temperature dependence of the resistance of $YbNi_2B_2C$ has an anomalous drop at $T<40$ K not observed for the other $RENi_2B_2C$ compounds. These anomalies were connected with a moderate heavy fermion-like behaviour of $YbNi_2B_2C$ [7,8].

The investigation of magnetoresistance (MR) for the YNi_2B_2C compound has revealed vanishingly small field dependence of resistance at $H<130$ kOe [9]. It would be interesting to compare these results with those for borocarbides based on magnetic rare earths and particularly for the $YbNi_2B_2C$ compound including the behaviour of MR in sufficiently high magnetic fields. However only fragmentary data on normal-state MR were mentioned in several publications [10,11] devoted to some $RENi_2B_2C$ compounds (RE=Lu, Er, Ho) in relatively small magnetic fields. It is also interesting to study the Hall-effect for $YbNi_2B_2C$ and to compare the results with the data for YNi_2B_2C [9].

In this work, the results of a systematic study of normal-state magnetoresistance in magnetic fields up to 130 kOe are reported and discussed for several $RENi_2B_2C$ compounds (RE=Lu, Yb, Tm, Ho) based on either nonmagnetic or magnetic RE^{3+} ions.

2. Experimental details

The synthesis of $RENi_2B_2C$ polycrystalline samples was carried out using a 'high pressure–high temperature' technique in a 'toroid' high pressure cell by the method similar to the one used in Refs. [9,12]. No annealing process was performed after synthesis. Sample quality and crystal structure parameters were controlled at room temperature by X-ray diffraction. All the investigated samples were essentially single phase containing only small amounts (<5%) of REB_2C_2 impurity phase. The registered lines were indexed with a body centred tetragonal structure (space group *I4/mmm*). The obtained lattice

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Table 1
Lattice parameters a and c for several $RENi_2B_2C$

RE	a, Å	c, Å
Ho	3.512	10.492
Tm	3.477	10.621
Yb	3.485	10.621
Lu	3.461	10.622

constants (see Table 1) are in agreement with those reported in, e.g., [8].

Bar-shaped samples were cut from the ingots. Resistance measurements were performed by a standard four probe ac-technique at $1.5 \leq T \leq 300$ K and in magnetic fields up to 130 kOe. Signals were registered by PAR-5210 Lock-in amplifier. For superconducting samples at $T < T_c$, normal-state MR was determined at $H > H_{c2}(T)$.

Two of the investigated compounds (RE=Lu and Tm) were superconducting with T_c values of 15.5 and 8.5 K, respectively. These values are slightly lower than reported in Ref. [2] (16.6 and 10.5 K), a fact that may be connected with the method of sample preparation. High pressure synthesis may also lead to considerably higher values of resistivity. For example the resistivity values of $TmNi_2B_2C$ equal 128 μΩcm and 56 μΩcm at 300 and 12 K respectively. The values of resistivity for the other $RENi_2B_2C$ compounds investigated in this work are comparable with these values. As mentioned above, no annealing process was performed after high-pressure synthesis.

No signs of superconductivity were observed for our $HoNi_2B_2C$ and $YbNi_2B_2C$ samples, although the former compound was reported to be a superconductor with $T_c = 8$ K [2]. It should be pointed out that this compound is extremely sensitive to Ni stoichiometry. The sample with excess of Ni of only 0.3% does not show any trace of superconductivity [13]. One of the possible reasons for the lack of superconductivity for $HoNi_2B_2C$, in our case, may be connected with a slight deviation of the Ni concentration from the nominal composition during high pressure synthesis.

3. Results and discussion

Temperature dependencies of resistance $R(T)$ in zero magnetic field for all investigated $RENi_2B_2C$ compounds (RE=Lu, Yb, Tm, Ho) are shown in Fig. 1. For comparison $R(T)$ dependencies in maximum applied magnetic field $H = 130$ kOe are also shown. In accordance with previously published data the $R(T)$ dependencies in zero magnetic field exhibit metal-like behaviour.

As can be seen from Fig. 1, for superconducting compounds $RENi_2B_2C$ (RE=Lu, Tm) a magnetic field $H = 130$ kOe is sufficient to fully destroy the superconducting transitions at $T > 1.5$ K. The values of critical magnetic fields $H_{c2}(0)$ for these compounds, estimated from the shift of resistive transition in magnetic field, are approximately 90 and 16 kOe, respectively. In the case of $LuNi_2B_2C$ with nonmagnetic Lu^{3+}, the magnetic field not only destroys the superconductivity but also leads to a slight increase of the resistance in normal state. On the other hand for the compounds based on RE with localized magnetic moment, the increase of magnetic field decreases the normal state resistance.

An anomalous decrease of the resistance with lowering temperature was observed for $YbNi_2B_2C$ (at $T < 30$ K) as reported in Refs. [7,8] and for $HoNi_2B_2C$ (at $T \approx 6 \div 7$ K). In the case of $YbNi_2B_2C$, the anomaly persists in $H = 130$ kOe, see Fig. 1. However in magnetic field $H = 130$ kOe, the peculiarity for $HoNi_2B_2C$ disappears fully.

If the anomalous drop in resistance is connected with partial superconductivity of our $HoNi_2B_2C$ sample, one

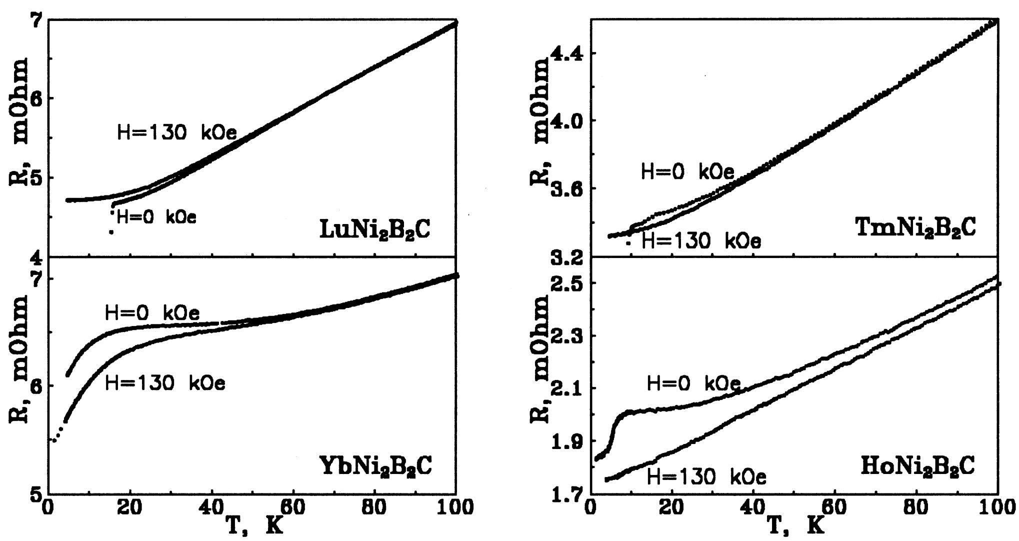

Fig. 1. Temperature dependencies of resistance in zero magnetic field and in H=130 kOe for several $RENi_2B_2C$ (RE=Lu, Yb, Tm, Ho) compounds.

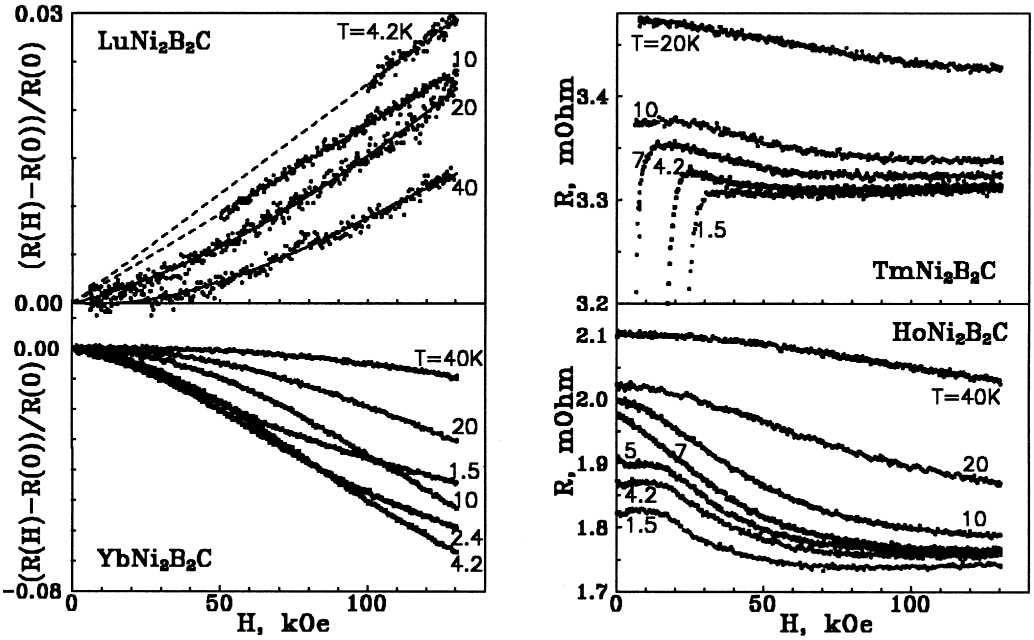

Fig. 2. Magnetoresistance of $RENi_2B_2C$ compounds (RE=Lu, Yb, Tm, Ho) for several temperatures (the absolute values of resistance are shown for RE=Tm and Ho). Numbers on figures denote temperature in K.

would expect positive magnetoresistance at $T<7$ K connected with the destruction of superconductivity by magnetic field. But as can be seen from Fig. 2, only negative magnetoresistance was observed. Also one would expect that this anomaly should be completely suppressed in $H>H_{c2}(0)=4\div8$ kOe [3,13]), but magnetic field $H=10$ kOe$>H_{c2}(0)$ only slightly shifts the observed anomaly. So it is reasonable to conclude that this anomaly is not connected with partial superconductivity of our sample, but rather with the reduction of spin disorder resistivity below T_N. The anomaly disappears fully in high magnetic field $H=130$ kOe eventually caused by field suppression of magnetic transition. It should be noted that for $DyNi_2B_2C$, a similar sharp decrease of resistance was associated with the AF transition at $T_N=10.3$ K, followed by a superconducting transition with $T_c=6.2$ K$<T_N$ [4]. Magnetic field $H=5$ kOe only weakly shifts the anomaly at T_N but fully suppresses a superconducting transition at $T>4.2$ K [4].

The results of normal-state MR study for all investigated compounds are presented in detail in Fig. 2. It is clearly seen that as mentioned above, only positive MR was observed for $LuNi_2B_2C$ with the nonmagnetic Lu^{3+} ion at $4.2\leq T\leq40$ K. The value of normalized MR is small enough and increases when temperature decreases. MR equals approximately 3% at $T=20$ K and $H=130$ kOe. To obtain the $(R(H)-R(0))/R(0)$ dependence at $T<T_c=15.5$ K, the observed $R(H)$ dependence at $H>H_{c2}(T)$ was extrapolated to $H=0$ using the simplest linear dependencies shown on Fig. 2 by dashed lines. The obtained $R(H)$ dependencies at $T<T_c=15.5$ K may not be pure normal state MR (some contribution from destruction of superconductivity is not excluded if the magnetic field only slightly exceeds $H_{c2}(T)$ value).

For all investigated compounds based on RE^{3+} ions with localized magnetic moments (RE=Yb, Tm, Ho), only negative MR was observed (Fig. 2) probably connected with the reduction of spin-disorder scattering in a magnetic field.

For $TmNi_2B_2C$ at $T<8$ K, in small magnetic fields, the completion of superconductivity destruction is clearly seen. Measurements at higher fields give the possibility of determining normal-state MR. Normal-state MR is negative at all temperatures but $R(H)$ dependencies begin to pass through a minimum at $H\approx60$ kOe as the temperature decreases below about 5 K. Similar $R(H)$ dependencies with a minimum were also observed at $T<5$ K for $HoNi_2B_2C$ compound. So at $T<5$ K and in sufficiently high magnetic fields ($H>60$ kOe) the resistance of Tm- and Ho-based compounds begins to rise with increase of H as it is in the case of $LuNi_2B_2C$ for the whole range of magnetic fields. Such behaviour may be connected with a sufficiently high extent of alignment of RE^{3+} magnetic moments in high magnetic fields at $T<5$ K and consequently with predominance of the nonmagnetic part of MR.

On the other hand, the resistance of $YbNi_2B_2C$ monotonically decreases with an increase of magnetic field up to $H=130$ kOe in the whole temperature interval (1.5÷40 K). The decrease of resistance at $H<50$ kOe increases in absolute value with lowering T, though in high magnetic fields MR decreases in absolute value as temperature lowers below 4 K, see Fig. 2. The difference in character of MR between $YbNi_2B_2C$ and $HoNi_2B_2C$ or $TmNi_2B_2C$ compounds may be connected with strong electron hybridization leading to the absence of superconductivity and heavy fermion-like behaviour pointed out for $YbNi_2B_2C$ [7,8]. From the results obtained, it is possible to suppose the essential difference of field dependence of magnetic moment in high magnetic fields between $YbNi_2B_2C$ and other borocarbides based on magnetic RE^{3+} ions for which heavy fermion-like behaviour was not observed.

It should be noted that for the $HoNi_2B_2C$ sample, rather complicated MR behaviour was observed at $H<25$ kOe and $T<10$ K, see Fig. 2, that is probably connected with several magnetic transitions in this compound at $T<7$ K [11,14].

Preliminary Hall-effect measurements of $YbNi_2B_2C$ performed at $1.8\leq T\leq 300$ K revealed that the Hall coefficient R_H is negative similar to YNi_2B_2C [9,15]. The estimated R_H gives $-4.0\ 10^{-12}$ Ωcm/Oe at $T=4.2$ K that corresponds to the one-band model Hall density of 2.0 per cell. Contrary to the relatively small temperature dependence of the Hall coefficient for YNi_2B_2C [9,15], the value of R_H for $YbNi_2B_2C$ decreases by approximately 3 times with increase of temperature from 4.2 to 300 K.

4. Conclusion

For $LuNi_2B_2C$ only positive MR was observed. At the same time MR was negative for all investigated compounds based on RE^{3+} ions with localized magnetic moments (Ho^{3+}, Tm^{3+}, Yb^{3+}) being qualitatively different for various RE^{3+}. In the case of RE=Ho and Tm, the $R(H)$ dependencies begin to pass through a minimum at $H=60$ kOe as the temperature decreases below about 5 K. On the other hand, the resistance of nonsuperconducting $YbNi_2B_2C$ monotonically decreases with increase of H in the whole temperature interval (1.5÷40 K). The difference in character of MR between $YbNi_2B_2C$ and $HoNi_2B_2C$ or $TmNi_2B_2C$ may be connected with heavy fermion-like behaviour pointed out for $YbNi_2B_2C$ [7,8]. The essential difference in field dependence of the magnetic moment in high magnetic fields between $YbNi_2B_2C$ and other magnetic borocarbides is supposed.

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References

[1] R. Nagarajan, C. Mazumdar, Z. Hossain, S.K. Dhar, K.V. Gopalakrishnan, L.C. Gupta, C. Godart, B.D. Padalia, R. Vijayaraghavan, Phys. Rev. Lett. 72 (1994) 274–277.
[2] R.J. Cava, H. Takagi, H.W. Zandbergen, J.J. Krajewski, W.F. Peck, T. Siegrist, B. Batlogg, R.B. Dover, R.J. Felder, K. Mizuhashi, J.O. Lee, H. Eisaki, S. Uchida, Nature 367 (1994) 252–253.
[3] H. Eisaki, H. Takagi, R.J. Cava, B. Batlogg, J.J. Krajewski, W.F. Peck, K. Mizuhashi, J.O. Lee, S. Uchida, Phys. Rev. B 50 (1994) 647–650.
[4] B.K. Cho, P.C. Canfield, D.C. Johnston, Phys. Rev. B 52 (1995) 3844–3847.
[5] T. Siegrist, H.W. Zandbergen, R.J. Cava, J.J. Krajewski, W.F. Peck, Nature 367 (1994) 254–256.
[6] C.C. Lai, M.S. Lin, Y.B. You, H.C. Ku, Phys. Rev. B 51 (1995) 420–423.
[7] S.K. Dhar, R. Nagarajan, Z. Hossain, E. Tominez, C. Godart, L.C. Gupta, R. Vijayaraghavan, Solid State Comm. 98 (1996) 985–989.
[8] A. Yatskar, N.K. Budraa, W.P. Beyermann, P.C. Canfield, S.L. Bud'ko, Phys. Rev. B 54 (1996) 3772–3775.
[9] V.N. Narozhnyi, V.N. Kochetkov, A.V. Tsvyashchenko, L.N. Fomicheva, J. Low Temp. Phys. 105 (1996) 1647–1652.
[10] I.R. Fisher, J.R. Cooper, K.R. Locherer, R.J. Cava, P.C. Canfield, J. Low Temp. Phys. 105 (1996) 1623–1628.
[11] K.D.D. Rathnayaka, D.G. Naugle, B.K. Cho, P.C. Canfield, Phys. Rev. B 53 (1996) 5688–5695.
[12] A.V. Tsvyashchenko, J. Less-Common Met. 99 (1984) L9.
[13] H. Schmidt, M. Weber, H. Braun, Physica C 256 (1996) 393–399.
[14] J.P. Hill, B.J. Sternlieb, D. Gibbs, C. Detlefs, A.I. Goldman, C. Stassis, P.C. Canfield, B.K. Cho, Phys. Rev. B 53 (1996) 3487–3491.
[15] I.R. Fisher, J.R. Cooper, R.J. Cava, Phys. Rev. B 52 (1995) 15086–15089.