Journal of Alloys and Compounds 262–263 (1997) 22–27

Quaternary borocarbides: the new and exciting magnetic superconductors

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Abstract

Quaternary borocarbide superconductors $RENi_2B_2C$ (RE = Y, Dy, Ho, Er, Tm and Lu) have several features which make them ideal candidates to study one of the most exciting and intriguing phenomena of condensed matter, viz., the interplay of superconductivity and long range magnetic order. Not only their superconducting transition temperatures are the highest among the intermetallic superconductors, their magnetic ordering temperatures are also the highest among all the known magnetic superconductors. $DyNi_2B_2C$ presents a very interesting situation; superconductivity sets in an already magnetically ordered lattice. Other examples of this interplay in the borocarbide family will be presented. Absence of superconductivity in $YbNi_2B_2C$ is rather anomalous; it points to the 4f-conduction electron hybridization. © 1997 Elsevier Science S.A.

Keywords: Borocarbides; Quaternary; High-T_c superconductivity; Magnetism; Interplay

1. Introduction

Discovery of superconductivity in the multiphase quaternary borocarbide system Y-Ni-B-C [1,2] is a major event in condensed matter. It took place nearly 7 years after the Bednorz–Muller discovery of High-T_c superconductivity and injected fresh life into superconductivity research. One of the most exciting features of borocarbide superconductors is the co-existence/interplay of superconductivity and magnetism occurring at temperatures much higher than observed ever before.

This discovery came about as a result of our interest and efforts, spanning over the last 2 decades, in the phenomena of valence fluctuations. Among a variety of mixed valence compounds, we investigated in several Ni-containing materials. Members of the series of ternary borides $RENi_4B$ (RE = rare earth) were studied as part of this program. We observed (see Fig. 1), in samples of nominal composition of YNi_4B [1], a weak but reproducible signal of superconductivity at T_c as high as 12 K. This was an exciting result as no binary or ternary superconducting phases of the elements Y, Ni and B are known with such high T_c. We, thus, had a new superconducting phase with T_c as high as 12 K.

In our efforts to investigate the origin of this weak superconducting signal, we prepared alloys YNi_4BC_x in which carbon had been introduced. A dramatic enhancement of the superconducting properties took place in these alloys. Several samples of the compositions $YNi_xB_yC_z$, with varying relative proportions of Ni, B, and C, were studied. Superconducting sample (resistivity and diamagnetism shown in Fig. 2) $YNi_2B_3C_{0.2}$ exhibited a sizeable specific heat anomaly at ~ 13 K, thereby establishing bulk superconductivity in the quaternary system Y-Ni-B-C [2]. This pioneering and seminal discovery [1,2] laid down the foundation of the new and exciting field of quaternary borocarbide superconductors.

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PII S0925-8388(97)00323-X

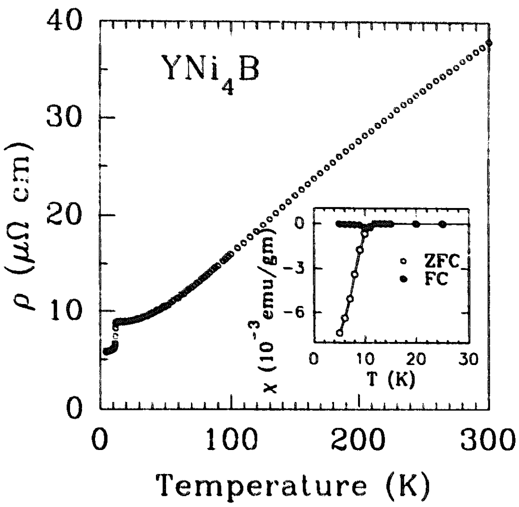

Fig. 1. Superconductivity at ~ 12 K in samples of nominal composition YNi_4B. There is a sharp drop in resistivity and the material exhibits diamagnetism (inset).

Subsequently, T_c as high as 23 K [3] and two superconducting transitions with $T_c \approx 22$ K and 10 K [4] were reported in multiphase samples of the Pd-containing quaternary system Y-Pd-B-C. Some of our samples of the composition YNi_2B_2C showed, in microwave absorption studies [5], a feature suggesting the existence of a superconducting phase with $T_c \approx 23$ K. Subsequently, possibility of a superconducting transition at $T_C \sim 24$ K was indicated [6] in samples of the quaternary system Y-Ni-B-C. These samples were synthesized using powder metallurgy under high temperature — high pressure conditions.

2. Structure of the $RENi_2B_2C$-phase

Fig. 3 shows a unit cell of the structure [7] of the superconducting single phase $LuNi_2B_2C$ ($T_c \sim 16.5$ K [8]) belonging to the quaternary system RE-Ni-B-C.

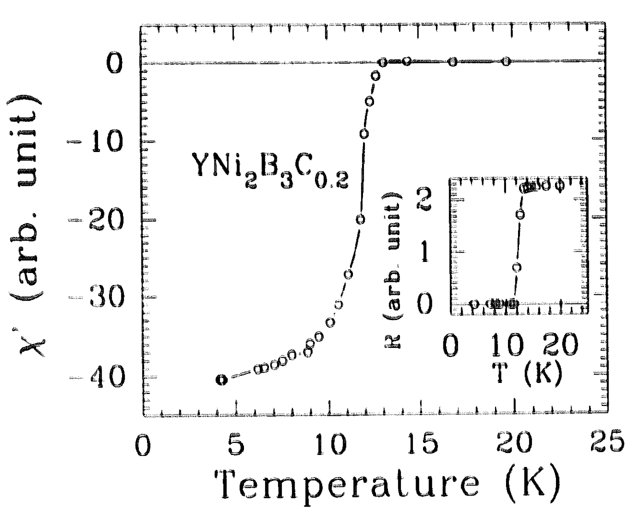

Fig. 2. Superconductivity in $YNi_2B_3C_x$. The diamagnetic response and the drop in resistivity (inset) start at $T \sim 13$ K.

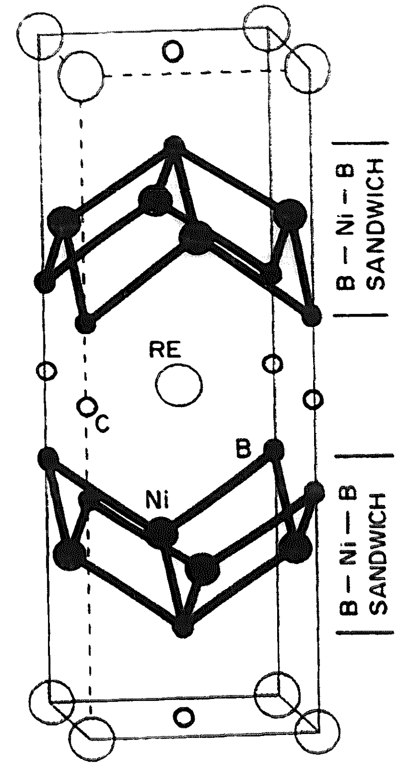

Fig. 3. Unit cell of the structure of $RENi_2B_2C$. See text for details.

This is a filled-in version of the well known tetragonal $ThCr_2Si_2$-structure (space group I4/mmm). It was confirmed [9] that YNi_2B_2C and several other members of the series $RENi_2B_2C$ (RE = Ho, Er and Tm) have the same structure. Highly anisotropic vibrations of C-atoms were inferred from the X-ray diffraction [9]. The structure is essentially a stack of infinite RE-C sheets and Ni_2B_2 ('B-Ni-B sandwiches') layers arranged in an alternating sequence along the c-axis. The short B–C bonds provide contact between RE-C planes and Ni_2B_2-layers; they are also responsible for the conduction path along the c-axis.

It is to be recalled that in the ternary superconductors such as $RERh_4B_4$ and $REMo_6S_8$, co-existence of superconductivity and magnetism and relatively high T_c have been regarded [10a,b] to arise because their respective structure contains clusters of transition metal atoms. Borocarbides have no such clusters but they exhibit high T_c values and high T_N values (see below). Therefore, clusters are not a crucial and essential ingredient for high T_c and the co-existence phenomena in intermetallics.

3. Superconductivity and magnetism in $RENi_2B_2C$

Borocarbides are particularly important as they have rather high T_c values and T_N values, see Table 1. In fact, the co-existence occurs at the highest ever reported temperature. Further, $RENi_2B_2C$ series is

Table 1
Superconducting (T_c) and anti-ferromagnetic transition (T_N) temperatures in superconducting quaternary Ni-containing borocarbides

$RENi_2B_2C$	T_c/K	T_N/K	Remarks
$DyNi_2B_2C$	6[a]	11[b]	$T_c < T_N$
$HoNi_2B_2C$	8[c]	8.5[d]	$T_c \sim T_N$
$ErNi_2B_2C$	11[c]	6.5[e]	$T_c > T_N$
$TmNi_2B_2C$	10.6[c]	1.5[f]	$T_c > T_N$
$YbNi_2B_2C$	$T_c < 2$[g]	$T_N < 2$[g]	

[a] [12–15].
[b] [11,12].
[c] [8].
[d] [11,25].
[e] [11,29].
[f] 22a, 22b, 35, 37].
[g] [15,40,41].

unique as its members have all the conceivable combinations of T_c and T_N, namely, $T_c > T_N$, $T_c \sim T_N$ and $T_c < T_N$. In this paper, we focus on some of the phenomena arising due to the interplay of superconductivity and magnetism in these materials.

3.1. $DyNi_2B_2C$: ($T_c \sim 6$ K, $T_N \sim 11$ K; $T_c < T_N$)

From the magnetic susceptibility [11,12] and specific heat [12] measurements, it was shown that $DyNi_2B_2C$ undergoes an antiferromagnetic ordering at $T_N \approx 11$ K. Superconductivity at a fairly high T_c (~ 6 K) was reported both in single crystal [13,14] as well as polycrystal samples of $DyNi_2B_2C$ [12,15]. Fig. 4 [15] shows ac-susceptibility of a polycrystalline sample of $DyNi_2B_2C$ where one clearly sees the occurrence of magnetic transition at ≈ 11 K and superconducting transition at ≈ 6 K. As T_c is less than T_N, one observes here superconductivity in an already magnetically ordered lattice. Such materials are rather rare:

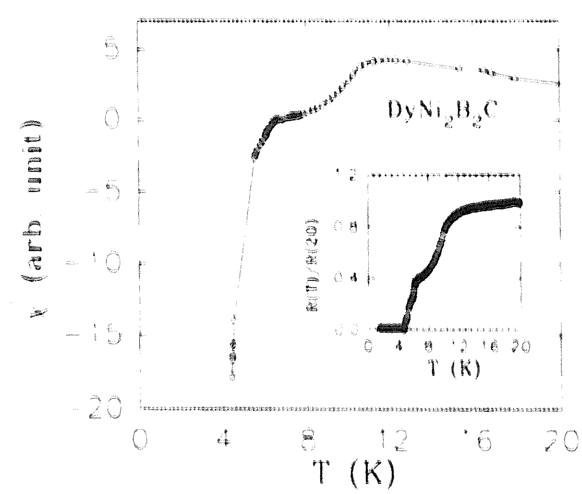

Fig. 4. AC-susceptibility and resistivity (inset) of a poly-crystalline sample of $DyNi_2B_2C$ showing clearly super-conducting and magnetic transitions.

only two other stoichiometric compounds ($Er_2Fe_3Si_5$: $T_c \approx 1$ K and $T_N \approx 2.9$ K [16]; and $Tb_2Mo_3Si_4$: $T_c \approx 1.5$ K and $T_N \approx 19$ K [17]) are known. $DyNi_2B_2C$ has the highest T_c among them.

Only a few materials with $T_c < T_N$ are known inspite of the fact that there is no fundamental reason why many more such materials should not exist. However, in view of the fact that non-magnetic impurities give rise to pair-breaking — and therefore depression of T_c — in anti-ferromagnetic superconductors [18a–d], it is conceivable that there are many anti-ferromagnetic superconductors; their T_c is, however, suppressed beyond observation by non-magnetic impurities which, presumably, may always be present in small concentrations and are hard to detect. It must be pointed out that non-magnetic impurities do not appreciably affect T_c in conventional isotropic superconductors (Anderson's theorem [19]). The following two observations are very instructive in this regard: (i) superconductivity in $DyNi_2B_2C$ was not observed down to 2 K in the earlier studies [11]. Perhaps, some extrinsic/intrinsic disorder caused the depression of T_c in these studies (ii) T_c in $Dy_{1-x}Lu_xNi_2B_2C$ [20] gets depressed strongly by non-magnetic Lu-ions.

One possible approach, as it emerges from what has been said above, to identify magnetic superconductors with $T_c < T_N$ is to examine more closely such anti-ferromagnets whose non-magnetic analogues exhibit superconductivity. One can easily find several such examples from literature [21]. These materials should be prepared so as to minimize the effects of non-magnetic impurities.

The basic message is: observation of superconductivity in $DyNi_2B_2C$ at T_c less than T_N is a clear assurance of the existence of many more of such materials and that the two earlier known materials with $T_c < T_N$ (see above) are not a freak of nature. As has already been stressed [21], such materials are significant for investigating phenomena related to coexistence of superconductivity and magnetism, such as gapless superconductivity in anti-ferromagnetic superconductors, atomic-level Meissner shielding, nature of pairing in the anti-ferromagnetic state and so on.

Magnetic structure of $DyNi_2B_2C$, as determined from neutron diffraction studies [22a,b; 23a,b] is quite simple. Ferromagnetic sheets of Dy-moments (confined in the a–b plane with no component along the c-axis) are stacked anti-ferromagnetically along the c-axis. ^{161}Dy-Mossbauer absorption studies in $DyNi_2B_2C$ show [24] that Dy-spins undergo a first order magnetic transition at ~ 11 K and that the Dy-C layers are insulating. The Ni_2B_2-layers are then the carriers of supercurrent in $RENi_2B_2C$ superconductors.

3.2. $HoNi_2B_2C$ ($T_c \sim 8$ K, $T_N \sim 8.5$ K; $T_c \sim T_N$)

A remarkable feature of $HoNi_2B_2C$ is that it exhibits double re-entrant superconducting transition (DRST) [11,25]; it is the first material in which such a behavior has been observed in zero applied field. Coming from high temperature, $HoNi_2B_2C$ orders magnetically (c-axis spiral [25]) at ≈ 8.5 K and goes into the superconducting state (S_1-state) almost at the same temperature ($T_c \approx 8$ K). It re-enters the normal state at ≈ 5 K, and regains superconductivity (S_2-state) as the material is further cooled below 5 K. There is a modification in the nature of the magnetic ordering as the material passes from the S_1-state to S_2-state. Magnetic ordering is incommensurate in the S_1-state whereas it is commensurate in the S_2-state [22a,b; 25]. Drastic reduction of the superconducting fluid density $\rho_{sup}(T)$ [26] in the normal state, sandwiched between the two superconducting states S_1 and S_2, is consistent with DRST in $HoNi_2B_2C$. Double re-entrant superconducting transition is very sensitive to the chemical composition (sample dependent) and the heat treatment [27a,b; 28a,b]. Fig. 5 shows ac-$\chi(T)$ of two samples I and II of $HoNi_2B_2C$ having nearly the same nominal composition. Sample II undergoes a transition at $T_c = 8$ K whereas sample I exhibits DRST. We believe that non-magnetic impurities, which are hard to detect and are likely to be present, are responsible for this sample-dependent behavior of $HoNi_2B_2C$.

3.3. $ErNi_2B_2C$ ($T_c \sim 10.6$ K, $T_N \sim 7$ K; $T_c > T_N$)

Co-existence of superconductivity and magnetism

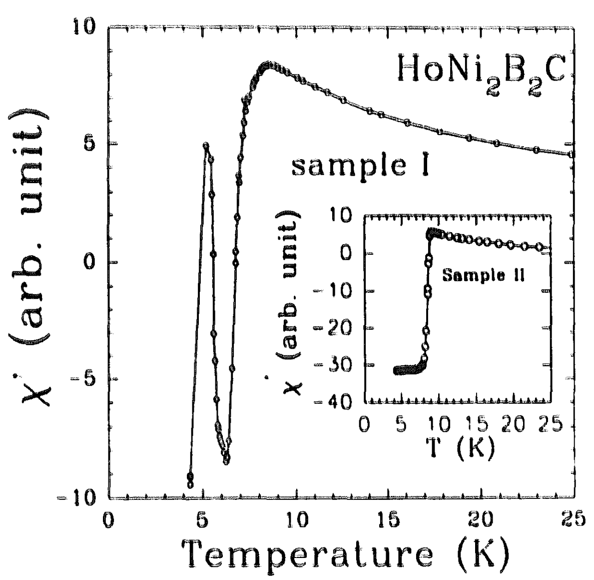

Fig. 5. Diamagnetic response of two polycrystalline samples of $HoNi_2B_2C$. Sample II shows a superconducting transition at 8 K whereas sample I clearly shows the double re-entrant superconducting transition (Z. Hossain, Chandan Mazumdar, R. Nagarajan and L. C. Gupta, unpublished results).

in $ErNi_2B_2C$ was observed through specific heat and resistivity studies [29]. With its $T_c > T_N$, this material provides a complementary combination of T_c and T_N with respect to that in $DyNi_2B_2C$. Neutron diffraction studies, both in polycrystal as well as in single crystal samples, show that Er-magnetic moments undergo an incommensurate anti-ferromagnetic ordering [30,31]. The Er-moments are ordered in a transversely polarized planar sinusoidal structure propagating along the a(b)-axis with Er-moments parallel to the b(a)-axis. No c-component was observed in these measurements. Though speculations [32a,b] have been made on the possibility of $ErNi_2B_2C$ undergoing a transition to a weakly ferromagnetic state below 2.3 K, the existing data [32a,b] do not conclusively prove it. Implications of such a state with respect to a spontaneous vortex phase in a magnetic superconductor have been pointed out [33].

From the Mossbauer studies of ^{166}Er in $ErNi_2B_2C$ [34] it was shown that Er-spins undergo a first order magnetic transition at $T_N = 6$ K into an incommensurate magnetic phase and the sinusoidal modulation gets fully squared up at low temperatures ($T = 1.4$ K). Er-spin relaxation rate exhibits an anomaly at $T_c = 10.6$ K, suggesting that the band electrons are exchange-coupled with the 4f-moments and are involved in the formation of the superconducting state. In fact, high T_N values in borocarbides also imply that the RKKY-interaction is the dominant exchange interaction between the RE- and the conduction electron spins. In this respect, borocarbides are very different from the ternary superconductors $RERh_4B_4$ $REMo_6S_8$, wherein it is usually assumed that the superconducting electrons (derived from Rh- and Mo-metal atoms) do not have appreciable interaction with the RE-spins.

3.4. $TmNi_2B_2C$ ($T_c = 11$ K, $T_N = 1.5$ K; $T_c > T_N$)

$TmNi_2B_2C$ has the smallest ratio $T_N/T_c = 1.5/11$ among all the magnetic superconducting quaternary borocarbides. The magnetic structure of this material is quite different from that of the three borocarbide magnetic superconductors, namely, magnetic moments are oriented along the c-axis [22a,b] whereas they are perpendicular to the c-axis in $RENi_2B_2C$ (RE = Dy, Ho and Er).

$H_{c2}(T)$ is highly anisotropic below ~ 6 K, $H_{c2}(\perp c) \approx 2\, H_{c2}(\| c)$ [35]. This is consistent with the anisotropy of magnetic susceptibility in the normal state, namely, $\chi_{H\| c} > \chi_{H\perp c}$. These observations suggest that the magnetic pair-breaking is responsible for the overall anisotropy of H_{c2}. Specific heat measurements [36] have been interpreted to suggest that the Tm-C sheets order ferromagnetically (strong ferromagnetic interaction within the Tm-C sheet) and that these ferromagnetic sheets are weakly coupled anti-ferromagnetically. Two-dimensional-ferromagnetic magnons, arising due to the ferromagnetically aligned sheets of Tm-moments, contribute a large gamma term to the low temperature specific heat [36].

Mu-SR measurements have been carried out on $TmNi_2B_2C$ [37] down to 100 mK to learn about the dynamics of Tm- and Ni-moments. The Tm-spins are correlated, at the Mu-SR time scale, at temperatures as high as $T > 10\ T_N$. The rapid 2D-correlations in the normal phase are slowed down by the onset of superconductivity. Anti-ferromagnetic fluctuations associated with Ni-moments have been studied in YNi_2B_2C by NMR techniques with divergent conclusions drawn with respect to their existence [38] and the absence [39]. We believe that magnetic fluctuations of Ni-spins in borocarbide superconductors, non-magnetic as well as magnetic, is an open and important question considering that Ni-electrons form Cooper pairs. Anti-ferromagnetic fluctuations may play a role in the mechanism of superconductivity.

3.5. $YbNi_2B_2C$ — a non-magnetic normal heavy fermion

Yb-ions are quite close to being trivalent in $YbNi_2B_2C$ [15,40]. From the pattern of variation of T_c in $RENi_2B_2C$ [21], $YbNi_2B_2C$ should superconduct at $T_c \sim 12$ K. Remarkably, $YbNi_2B_2C$ does not superconduct down to 2 K [15,40] nor does it order magnetically. Single crystal studies have also confirmed this behavior of $YbNi_2B_2C$ [41]. Specific heat studies show that a heavy fermion state evolves at low temperatures [40,41] suggesting an enhanced hybridization of the localized Yb-4f and the itinerant conduction electrons which may be responsible for the anomalous depression of T_c in $YbNi_2B_2C$.

4. Summary

To summarize, we have described here briefly the events that led to the discovery of superconductivity in multiphase samples of the quaternary system Y-Ni-B-C. Tetragonal structure of $RENi_2B_2C$ does not have clusters which used to be considered essential for high T_c in intermetallics. We commented briefly upon some of the features of borocarbides, for example, highest T_N in the superconducting state, all possible combinations of T_c and T_N: $T_c > T_c$, $T_c \sim T_c$, and $T_c < T_c$ and anomalous suppression of T_c due to the hybridization of the 4f- and conduction electrons. Mossbauer studies $ErNi_2B_2C$ suggest that the conduction electrons which are exchange-coupled with RE-spins are also involved in the formation of the superconducting state. This raises some basic questions with respect to the very survival of superconductivity under such circumstances, and one is motivated to take a fresh look at the co-existence phenomena. These materials provide us with an opportunity to investigate new and yet unexplored phenomena [21,42] associated with interplay of superconductivity and magnetism. Borocarbides have great potential to generate new physics; they have led to the pathway to identify new superconducting materials, possibly with even more exciting properties.

Acknowledgements

Several of our studies described here are a part of our collaborative program with laboratories within and outside India. Work in borocarbides at TIFR is being carried out in collaboration with R. Nagarajan, S.K. Dhar and Z. Hossain.

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