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Superconductivity and Magnetism in Quaternary Borocarbides

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Quaternary borocarbide superconductors have attracted attention and are of current interest due to a variety of reasons. Some of them exhibit high T_c's for intermetallics, including the material which shows highest known T_c for intermetallics. Though the structure of RNi_2B_2C (R = Sc, Y, rare earth, Th, U) is effectively a layered structure like that of high T_c cuprates, rare earth ions influence the superconducting properties of these materials. Coexistence of superconductivity and magnetism has been found in these materials with magnetic ordering temperatures much higher than those of earlier known magnetic superconductors which implies stronger coupling of rare earth moment to conduction electrons. These features make quaternary borocarbides an altogether different class of materials. Being quaternary, and with the possibility of having more than one rare earth-carbon layer in the structure, this family offers the possibility of finding many new materials with wide ranging properties. Highlights of recent developments in this new subject of quaternary borocarbides are reviewed here.

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1. INTRODUCTION

Discovery of superconductivity (SC) at an elevated temperature (T_c ~12-15K) in multiphase Y-Ni-B-C system,[1,2] subsequent finding of SC in single phase RNi_2B_2C (R = Y, Ho, Er, Tm, Lu)[3,4] and finding of record T_c for intermetallics in Y-Pd-B-C system (~23 K)[5] brought forth an intense activity in the field of intermetallic superconductors[6–11] resulting in the new field, *superconductivity and magnetism in quaternary borocarbides.*[12] Some of the reasons for the excitement are: high T_c's of the superconducting materials with T_c reaching to 16.5K in $LuNi_2B_2C$[3] – that too for materials with large proportion of Ni, which had not been known till then –; finding of SC with record high temperatures for intermetallics in Y-Pd-B-C system ($T_c \sim 23$ K);[5] the structure of RNi_2B_2C (body centered tetragonal)[4] falling in a class which is in between those of layered high T_c cuprates and cluster superconductors like RRh_4B_4 (R = rare earth), being quaternary providing more opportunity for new materials – a large number of materials have been found in this family, including those based on Sc, U and Th;[13,14] they provide the entire range of physical properties – some of the members exhibit SC, some magnetism and a few coexistence of SC and magnetism with high coexistence temperatures and with very interesting features[15] *etc.* Some of the developments in this field can be found in Refs. 6-17. Here we review some of the recent developments in this topical field.

2. MATERIALS

RT_2B_2C (R = Sc, Y, rare earth, Th, U; T = Ni, Rh, Pd, Ir, Pt), (1221 phase), which form bulk of the presently known quaternary borocarbides, crystallize in a filled variation of the well known $ThCr_2Si_2$ type tetragonal structure with C atoms in the rare earth planes and Ni_2B_2 layers separated by planes of R-C.[4,18] It had been realized that one could have a homologous series of materials $(RC)_m(Ni_2B_2)_n$ out of this structure by inserting more sheets of R-C layers between the Ni_2B_2 layers.[4,19,20] Only two superconductors (LuNiBC and $La_3Ni_2B_2N_3$) have been found so far with increased R-C layers. LuNiBC with two R-C layers (1111 phase) shows a T_c of 2.9 K.[21] Unlike the case of RNi_2B_2C, increase of T_c has been achieved in RNiBC system with partial substitution of elements.[22,23] Upon partial substitution of Cu for Ni, T_c has been increased to as high as 8.9 K in $YNi_{1-x}Cu_xBC$ and to 6.6 K in $LuNi_{1-x}Cu_xBC$.[23] One must bear in mind that with the structures of these homologous series being similar, when any new material is found with T_c lower than that of corresponding 1221 phase, there is a necessity to ensure that the observed SC is not due to a 1221 phase with a depressed T_c arising due to introduction of substituents. Systematic *increase* of T_c with *increasing* Cu substitution is one of the reasonable evidences of SC arising from 1111 phase.

In the class of materials with three layers of R-C, only one compound, that too a nitride, $La_3Ni_2B_2N_3$, is known so far and it shows SC with $T_c \sim 12.5$ K.[19] This material was believed to be an unstable system under ambient conditions. A recent work[24] has shown that this compound is indeed a stable one, and that the instability arises due to the presence of LaN impurity (which is hygroscopic). Detailed studies have shown that

$La_3Ni_2B_2N_3$ is a hard type-II phonon mediated weak- to medium-coupling BCS superconductor.[24] One compound with four R-C layers, Y_2NiBC_2, has been reported but it does not show SC down to 4.2 K.[20]

In the context of new materials, the current finding of SC with a T_c as high as 24 K, Y-Ni-B-C system[25] in a sample prepared using powder metallurgical method, is an exciting result and is a new record for T_c in intermetallics. It may be pointed out that in one of our early studies on YNi_2B_2C, the possibility of presence a superconducting phase in Y-Ni-B-C system with T_c around 23 K had been indicated from the results of microwave absorption in the material.[26] It is important to note that boron and carbon being light elements, the quantitative determination of their content in different phases of a multiphase material often involves considerable uncertainty. While, the finding of a SC phase with 24 K in Y-Ni-B-C seems to be a definite result, the phase identification may have to await further investigations and confirmation.

3. SUPERCONDUCTING PHASE IN Y-Pd-B-C SYSTEM

The structure of the phase which is responsible for SC with T_c ~23 K in Y-Pd-B-C is still not resolved satisfactorily, as it has not been possible to prepare single phase samples. Large diamagnetic signal is observed in the multiphase material with nominal composition $YPd_5B_3C_{0.3}$.[5] We observed two superconducting transitions (T_c ~22 K and 10 K),[27] though weak in strength, in multiphase YPd_4BC_x ($0.2 < x < 1$) which suggests that in this family more than one phase supports SC. There have been suggestions that the structure is $LuNi_2B_2C$ type tetragonal structure[28-31] but with a chemical composition deficient in boron (YPd_2BC)[28] or with off-stoichiometry in carbon ($YPd_2B_2C_x$).[29,30] We have prepared a number of Y-Pd-B-C compositions, examined them for structure and SC and have attempted to determine quaternary phase diagram of Y-Pd-B-C.[17] A cubic phase (a~4.14Å) is found in the series $YPd_2B_{1+x}C$, but SC is very weak in these samples.[17] Since SC vanishes annealing in Y-Pd-B-C samples,[5] ideally, the superconducting phase has to be searched in rapidly quenched samples. A work in this direction has recently been reported.[32] A cubic phase has been identified in the rapidly quenched melt spun multiphase sample with the nominal composition YPd_2B_2C. A correlation has also been observed that on annealing, along with the decrease of diamagnetic signal, the line in x-ray diffraction pattern corresponding to cubic phase also decreases. From these it was concluded that the 23 K phase is YPd_2B_2C, but with a fcc structure (a = 4.15 Å).[32] However, no microscopic element analysis have been carried out to corroborate the composition of the phase. In our opinion, the composition and structure of superconducting phase in Y-Pd-B-C is still an open question and more effort is needed to solve the problem. A solution to this problem may point fresh directions in this important area.

4. COEXISTENCE OF SUPERCONDUCTIVITY AND MAGNETISM

The superconducting, magnetic and coexistence properties of many of the RNi_2B_2C (R = Y, rare earth) have been investigated by large number of groups both in polycrystalline and single crystalline forms, using a variety of techniques.[6–11,13–17,33] Some of the recent and important results are mentioned here. Most of the experimental results on YNi_2B_2C show that it is a type-II moderately strong-coupling BCS superconductor.[34] The superconducting parameters are similar to that of conventional metallic superconductors.[35] rf-SQUID effect from Josephson junctions arsing from the natural grain boundary junctions present in polycrystalline YNi_2B_2C has also been observed.[36] Though the structure is anisotropic (lattice parameter ratio, c/a ~3), the experimentally observed superconducting properties are almost isotropic[37] which is also consistent with band structure calculations.[38] It has been shown in a recent theoretical work[39] that there is a strong similarity at a fundamental level between these borocarbides and high T_c cuprates and a simple one band t-J model has been proposed as an appropriate model to explain the electronic properties of the Ni-borocarbides. An interesting prediction of the model is that a replacement of Lu by a divalent ion such as Ca should result in a Mott insulating state.[39]

One of the recent exciting results has been the observation, in inelastic neutron scattering experiment on YNi_2B_2C, of appearance of an intense new peak (Q = 0.525, 0, 8; E = 4.75 meV) on the onset of SC.[40] The intensity of this mode has also been found to decrease on application of an external magnetic field, confirming the association of this mode with SC.[40] Further, it is found[40] that the Q vector direction nearly matches with that of the incommensurate vectors of magnetic ordering observed in RNi_2B_2C family. This may throw further light on the mechanism of SC in these borocarbides.

The coexistence temperatures of RNi_2B_2C (R = Er, Ho, Dy) (T_c = 10.5 K, 8 K, 6 K; and T_N = 7 K, 8 K, 11 K, respectively)[41,42] are higher than that known for any other magnetic superconductor. ($TmNi_2B_2C$ is the remaining magnetic superconductor of the family, with T_c ~11 K and T_N ~1.5 K). Such high ordering temperatures imply that magnetic order is mediated through conduction electrons by RKKY-type exchange interactions suggesting the likelihood of very intimate interaction between SC and magnetism. Indeed, the magnetic relaxation rate found in ^{166}Er Mössbauer

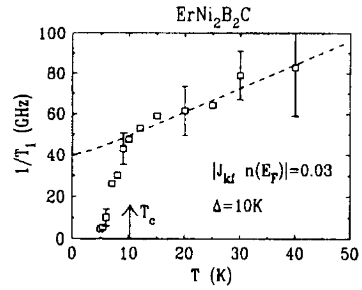

Fig. 1. Temperature dependence of Er^{3+} paramagnetic relaxation rate deduced from ^{166}Er Mössbauer spectra (From Ref. 43).

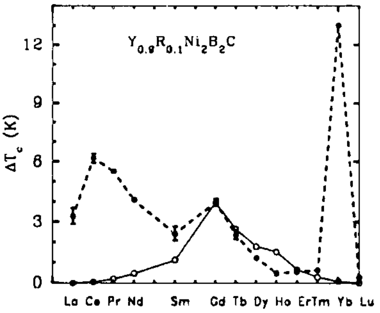

Fig. 2. Depression of T_c in YNi_2B_2C by 0.1 atomic fraction substitution of Y by different rare earths. •– experimental data, o– as expected for de-Gennes scaling. The lines are guide to eye.

investigations of $ErNi_2B_2C$ exhibit a significant change on the onset of SC (Fig. 1).[43] This suggests that conduction electrons which are responsible for the magnetic relaxation contribute to the formation of superconducting state.[43] Such an observation has not been seen before in any magnetic superconductor and is being investigated further. ^{161}Dy Mössbauer isomer shift results in $DyNi_2B_2C$ show that s-electron density at Dy site is similar to non metallic systems supporting that superconductivity arises from Ni_2B_2 layers.[44]

Magnetic structures of the magnetic superconductors of the quaternary borocarbides have been well investigated.[45] Amongst the magnetic superconductors, $HoNi_2B_2C$ and $DyNi_2B_2C$ are of particular interest, the former due to $T_c \approx T_N$ and the latter due to $T_c < T_N$.

In $DyNi_2B_2C$, (~11 K), SC occurs in an already magnetically ordered lattice ($T_c \sim 6.5$ K, $T_N \sim 11$ K).[46–49] Only two other such systems are known: $Er_2Fe_3Si_5$ ($T_c \sim 0.9$ K, $T_N \sim 2.5$ K) and $Tb_2Mo_3Si_4$ ($T_c \sim 2$ K, $T_N \sim 19$ K). It may be pointed out that though some heavy fermion superconductors with $T_c < T_N$, such as UPd_2Al_3, are known, the magnetic ion, however, has a much reduced magnetic moment in the ordered state in such cases.[50] Therefore, the magnetic superconductor $DyNi_2B_2C$ with relatively high T_c and T_N is an ideal material for investigation of coexistence phenomenon. For example, one would expect that SC in this material would be suppressed even by non magnetic impurities and has, indeed, been found to be the case. Substitution of 0.1 atomic fraction of Lu, is found to depress T_c

of $DyNi_2B_2C$ from 6.5 K to $\sim$ 3.5 K.[51] This may be viewed as pair breaking due to destruction of translational symmetry or creation of magnetic holes. $DyNi_2B_2C$ is a potential material to look for gapless SC due to nonmagnetic ions in a superconducting antiferromagnet.[15]

In the case of $HoNi_2B_2C$, T_c and T_N being very close, ($\sim$8 K), coexistence of SC and magnetic order seems to become very delicate. It exhibits a double reentrance behaviour in the absence of an externally applied magnetic field.[41] Such a phenomenon has not been seen in any other magnetic superconductor. Neutron investigations of $HoNi_2B_2C$ have shown[45] that in the temperature range 8 K$-$5 K (i.e. close to the onset of magnetic order) an incommensurate *c*-axis spiral magnetic spin structure develops which gives raise to a ferromagnetic component along *c*-direction resulting in the tendency to quench SC. However, at temperatures below 5 K, the spins settle down to a commensurate antiferromagnetic structure, restoring SC. A small incommensurate *a*-axis modulation of the spin structure, the intensity of which is sample dependent,[45] has also been observed. Recent results from neutron and x-ray scattering studies on $HoNi_2B_2C$ show that the magnetic structure of $HoNi_2B_2C$ in 5$-$7 K region is quite complex[52] having some components attributed to strain. Some of these may be due to effects of off-stoichiometry in the sample.

Superconducting properties of quaternary borocarbides appear to be sensitive to stoichiometry. In the NMR investigations of YNi_2B_2C, two ^{11}B resonance lines were observed below T_c.[53] Position of one of these is temperature independent and corresponds to that occurring in normal state of the material, while that of the other shifts with temperature and corresponds to superconducting state. This unshifted line below T_c was suggested to arise from possible boron/carbon vacancies or site interchange.[53] Our positron annihilation results of YNi_2B_2C also suggest vacancies at the carbon site.[54] As remarked earlier, it is difficult to determine the stoichiometry or site occupancy of B and C through conventional techniques.

Effect of stoichiometry is found to be stronger in the case of the magnetic superconductors, $DyNi_2B_2C$ and $HoNi_2B_2C$. Reentrance behaviour in $HoNi_2B_2C$ is found to be sample dependent. Detailed investigations of composition dependence of superconductivity and magnetism in $HoNi_2B_2C$, show that under variation of stoichiometry, if there is a slight reduction in lattice volume, the material tends to exhibit reentrance behaviour with SC getting extinguished on further reduction of lattice volume.[55] The lattice volume dependence of SC property is further borne out from the fact that on application of pressure, a non reentrance sample, shows reentrance behaviour.[55,56] In the case of $DyNi_2B_2C$, T_c being less than T_N, SC in the material is even more sensitive to stoichiometry.[46,49] In fact, because of this, finding of SC in $DyNi_2B_2C$ was delayed. These aspects warrant that one has to be careful and ensure stoichiometry and homogeneity of the samples in investigating the coexistence properties, particularly those of $HoNi_2B_2C$ and $DyNi_2B_2C$. A systematic study has shown that there is an optimum annealing temperature at which impurity phases are a minimum and that optimum annealing temperatures are different for different members of RNi_2B_2C series.[17]

Another recent exciting result in the field has been the observation of square flux lattice in the magnetic superconductor $ErNi_2B_2C$[57] as against the usual hexagonal flux lattice. The flux lattice has also been observed to rotate away from the direction of applied field on development of magnetic order. This is perhaps the first data to show the direct interaction of flux lattice and magnetic order. The authors have left the origin of the square vortex as an open question. Very likely, the origin seems to be linked to the very structure of the lattice as very recently square flux lattice has been observed as well, in the non magnetic superconducting member of this family YNi_2B_2C[58] and also in the other magnetic superconductor, $HoNi_2B_2C$.[58]

5. ANOMALOUS SUPPRESSION OF SUPERCONDUCTIVITY IN $YbNi_2B_2C$

The reduction in T_c of the magnetic superconductors RNi_2B_2C (R = Tm, Er, Ho, Dy) seems to follow a systematic similar to that of deGennes scaling.[41] It must be remarked that the well known A-G theory on depression of T_c due to pair breaking by magnetic ions predicts the depression to follow deGennes scaling is applicable only for dilute paramagnetic substitutions. Therefore, it is surprising that deGennes scaling like depression of T_c is seen in these concentrated magnetic systems. Whatever be the mechanism, as there is a systematic of T_c with respect to rare earth, one would expect $YbNi_2B_2C$ to exhibit SC around 12 K. We have been successful in synthesizing single phase $YbNi_2B_2C$ by a sintering route.[49,59] Our studies show that though Yb in the material is in trivalent state (as inferred from lattice parameter, magnetic susceptibility and L_{III} edge investigations), very surprisingly no SC is seen down to 2 K.[49,59] Our resistivity and specific heat studies show that the material is a dense Kondo system which evolves into heavy fermion state at low temperature with a coefficient of electronic specific heat of at least ~200 mJ/mol.K^2.[59] Thus, strong hybridization of 4f-electrons with conduction electrons takes place in $YbNi_2B_2C$. No magnetic order is seen in this material down to 70 mK.[60] Very recent investigations on single crystals of $YbNi_2B_2C$ show results[61] which are consistent with our results from polycrystalline samples. It must also be recalled that Yb ions can support SC and in fact, among the members of RMo_6S_8 series, $YbMo_6S_8$ has the highest T_c (~9 K). Considering that heavy fermion superconductors are known, further studies are needed to understand the exact cause of suppression of SC in this material which in turn would indirectly add to the knowledge of mechanism of SC in these quaternary borocarbides. We have also been successful in synthesizing the related material YbNiBC.[62] Interestingly, our preliminary studies through susceptibility and specific heat show that YbNiBC orders magnetically around 4 K which is a relatively high ordering temperature for a Yb-based system.

To enhance the understanding of SC in these borocarbides, we have carried out a systematic study of substitution of Y in YNi_2B_2C by dilute rare earth ions.[63] Our results (Fig. 2) show that while for heavier rare earths, the depression nearly follows deGennes scaling, it does not follow for the lighter rare earths. Indeed, the depression by La and Ce are rather large. While the depression by Ce could be understood in terms of the hybridization effects of 4*f*-electrons with conduction electrons, as Ce is found to be in VF state in this series of materials,[4,10] the large depression by La is intriguing. $LaNi_2B_2C$ being a non superconductor is an equally important aspect of the superconducting behaviour of these family of materials. Structural parameters and bond angles may play a crucial role in sustaining SC.[18] The most important observation in our partial substitution studies is the anomalous depression of T_c in YNi_2B_2C by Yb (Fig. 2). Substitution of 0.1 atomic fraction of Y by Yb in YNi_2B_2C depresses T_c by more than 12 K which is the largest suppression observed for Yb substitution in an intermetallic system.[64] This once again demonstrates very strong hybridization of *4f*-electrons of Yb with conduction electrons in the borocarbides. Thus, Yb-systems of this family promises to be a very interesting and important ones.

6. SUMMARY

Quaternary borocarbides have continued to retain their importance since their discovery nearly three years ago. Surprising results have been continually coming up. Some of such results of recent findings are: finding of anomalous suppression of SC in $YbNi_2B_2C$, anomalously large depression of T_c by even dilute substitution of Yb in YNi_2B_2C, observation of a 24K superconducting phase in Y-Ni-B-C system, observation of square vortex flux lattice in RNi_2B_2C (R = Y, Er, Ho) *etc.* There are still problems to be solved, such as, unambiguous identification of the superconducting phase in Y-Pd-B-C system. Many more replacements of elements in the composition are yet to be tried out and multinaries may not be far away. The field is rich in potential for new materials and physics.

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