Diluted and concentrated isoelectronic substitutional effects in superconducting $R_xY_{1-x}Ni_2B_2C$ compounds

J. Freudenberger, G. Fuchs, K.-H. Müller, K. Nenkov, S.-L. Drechsler, A. Kreyssig[+], H. Rosner[+], K. Koepernik[*], D. Lipp[+], and L. Schultz

Institut für Festkörper- und Werkstofforschung Dresden, Postfach 270016, D-01171 Dresden, Germany
[+] *TU Dresden, Physik, D-01062 Dresden, Germany*
[*] *MPI CPFS Dresden, D-01187 Dresden, Germany*

The suppression of superconductivity of pseudo-quaternary $R_xY_{1-x}Ni_2B_2C$ compounds in the dilute limit was found to be mainly caused by magnetic pair-breaking in the case of the heavy rare-earth elements (R = Tm, Er, Ho, Dy, Tb, Gd) and by lattice distortions in the case of the light rare-earth elements (R = Pr, Nd) resulting from the large difference of the ionic radii between the Y host ions and the R impurity ones.

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1. INTRODUCTION

The superconducting transition temperature T_c of pure quaternary RNi_2B_2C compounds with R = Y, Tm, Er, Ho, Dy has been found to decrease nearly linearly with increasing de Gennes factor $dG = (g-1)^2J(J+1)$, where g is the Landé factor and J is the total angular momentum of the R^{3+} Hund's-rule ground state [1,2]. A linear scaling with dG has also been reported for the antiferromagnetic ordering temperature T_N across the RNi_2B_2C series with heavy rare-earth elements R including Tb and Gd [2]. This correlation between T_c and T_N for different R points to the dominance of magnetic pair-breaking. In the absence of crystalline-electric field effects the suppression of T_c by non-interacting paramagnetic impurities can be described quantitatively by the Abrikosov-Gor'kov theory [3], where in the dilute limit of magnetic impurity concentrations the change of T_c: $\delta T_c = T_c^0 - T_c$ fol-

lows the linear de Gennes scaling, i.e. $\delta T_c \propto dG$. Naturally, this scaling law holds also for several pseudo-quaternary $R_x R'_{1-x}Ni_2B_2C$ compounds [4–7] with magnetic R and nonmagnetic R' rare-earth elements, using an *effective* de Gennes factor $DG = xdG(R)$. But in the case of pseudo-quaternary compounds T_c is influenced additionally by the difference of the ionic radii of R and R' reflecting some nonmagnetic disorder on the rare-earth site. This effect has shown to be responsible for the stronger depression rate $\delta T_c/(DG)$ of the $Ho_x R'_{1-x}Ni_2B_2C$ compounds with $R' = Lu$ compared with $R' = Y$ [7,9]. The influence of nonmagnetic disorder effects on T_c has also been studied in $Lu_x Y_{1-x}Ni_2B_2C$ compounds [8,10].

The aim of the present study is to study the relationship of magnetic pair-breaking and of nonmagnetic *isoelectronic* substitutional disorder effects on the superconducting transition temperature of pseudo-quaternary $R_x Y_{1-x}Ni_2B_2C$ compounds in the dilute limit of paramagnetic impurities as well as to consider the former effect for concentrated mixed crystals.

2. EXPERIMENTAL DETAILS

Polycrystalline $R_x R'_{1-x}Ni_2B_2C$ samples were prepared by a standard arc melting technique as described in Ref. 7. A powder X-ray diffractometer in Bragg-Brentano-geometry was used to verify the phase purity of $> 95\%$ as well as the lattice structure. The X-ray diffraction experiments were done on crushed powders using Co_{K_α} radiation. AC susceptibility measurements were utilized to determine T_c as well as the transition width ΔT_c.

3. RESULTS

The X-ray patterns of the investigated samples revealed the $LuNi_2B_2C$-type structure (space group I/4mmm) [11]. For the lattice parameters a and c of the $R_x Y_{1-x}Ni_2B_2C$ compounds a linear interpolation between the parent compounds was found. No significant fractions of phase impurities were detected. The superconducting transitions of $R_x Y_{1-x}Ni_2B_2C$ compounds show sharp widths of up to $\Delta T_c \simeq 0.3$ K. The superconducting transition temperatures T_c of $R_{0.05}Y_{0.95}Ni_2B_2C$ compounds are shown in Fig. 1 in dependence on the effective de Gennes factor. This small concentration of paramagnetic R ions ensures that the compounds are in the dilute limit. As shown in Fig 1, a nearly linear depression of T_c with increasing de Gennes factor expected from the Abrikosov Gor'kov theory [3] is observed in the case of the heavy rare-earth elements. For the light rare-earth elements Pr and Nd strong deviations from the straight line describing approximately the

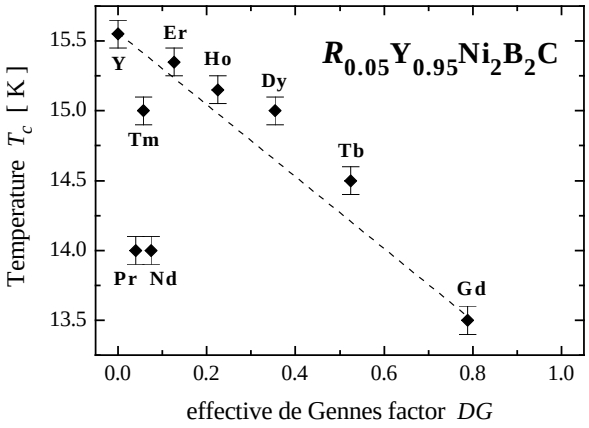

Fig. 1. Superconducting transition temperature T_c in dependence of the de Gennes factor $(g-1)^2J(J+1)$ for the $R_{0.05}Y_{0.95}Ni_2B_2C$ compounds.

experimental data for the heavy rare-earth elements are observed which can not be explained in the framework of the Abrikosov-Gor'kov theory.

In order to visualise the different influence of magnetic pair-breaking and disorder effects on T_c, the reduction of the superconducting transition temperatures, δT_c, of $R_{0.05}Y_{0.95}Ni_2B_2C$ samples and the depression rates $|\delta T_c/DG|$ are shown in Fig. 2 in dependence on the difference of the ionic radii, δr, between yttrium and the R ions.

The depression rates $|\delta T_c/DG|$ of $R_{0.05}Y_{0.95}Ni_2B_2C$ found for the heavy rare-earth elements R = Er, Ho, Dy, Tb and Gd are small and almost independent of δr. Hence, nonmagnetic disorder effects $\propto (\delta r)^2$ can be neglected and the variation of T_c observed for the heavy rare-earth elements is dominated by magnetic pair breaking. We attribute the enhanced depression rate for R = Tm, despite of $\delta r(Tm) \simeq -\delta r(Gd)$, to the fact that $TmNi_2B_2C$ is the only heavy rare-earth borocarbide compound with an magnetically easy c-axis [12,13].

The experimental data for the depression rates $|\delta T_c/DG|$ of $R_{0.05}Y_{0.95}Ni_2B_2C$ shown in Fig 2 can be described approximately by $|\delta T_c/DG| \simeq |\delta T_c/DG|_{\rm Gd}[1+a(\delta r)^2]$, where $|\delta T_c/DG|_{\rm Gd} = 2.5$ K is the depression rate for R = Gd and a is a fitting parameter. The $Gd_xY_{1-x}Ni_2B_2C$ compound is a convenient reference system for the study of pair-breaking by paramagnetic impurities in the family of pseudo-quaternary $R_xY_{1-x}Ni_2B_2C$ compounds, because Gd exhibits no crystalline electric field splitting of the ground-state multiplets. The second term in the relation above describes the increase of the depression rate in the case of large differences of the ionic radii between the Y host and the R impurity ion such as for R = Nd and Pr. The quadratic dependence of the depression rate $|\delta T_c/DG| \propto (\delta r)^2$

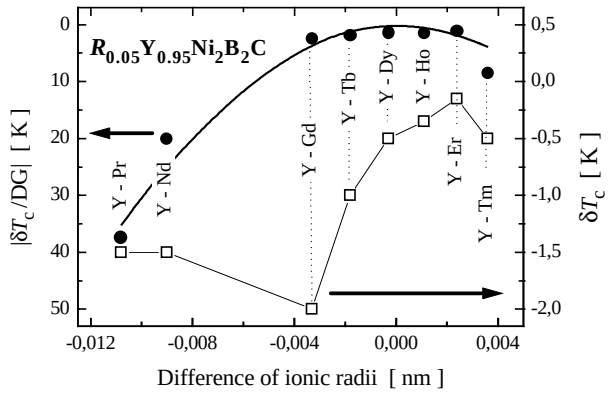

Fig. 2. Absolute reduction of the critical temperature δT_c (open squares) and the relative reduction of T_c normalized to the effective de Gennes factor DG (closed circles) in dependence on the host-impurity difference of the ionic radii.

upon the different ionic radii on the rare-earth site, found for large values of δr, suggests that local lattice strains are responsible for the variation of the depression rate.

4. ASPECTS OF CONCENTRATED NONMAGNETIC SUBSTITUTIONS

To supplement the given above analysis we performed electronic structure calculations for the electronic density of states $N(E_F, x)$ within the coherent potential approximation (CPA) for the $Lu_xY_{1-x}Ni_2B_2C$ system combined with a consideration of specific heat and the upper critical field $H_{c2}(T, x)$ data. Then from the empirically found linear relation [15] between T_c and the Sommerfeld constant γ of the electronic specific heat: $T_c \propto \gamma(x) = N(E_F, x)(1+\lambda(x))$, the empirical x-dependence of the electron-phonon interaction λ can be estimated. According to our CPA-calculations [16] the x-dependence of N is very smooth: it interpolates quadratically between the limiting values of N_Y=4.0 and N_{Lu}=4.2 states /(eV unit cell) at the level of 1.5 % suppression in between. Adopting a comparable suppression by the local distortions ignored in our approach, the remaining x-dependence of λ is much stronger: at about 12% interpolating again quadratically between λ_Y=1.05 and λ_{Lu}=1.09. This significant x-dependence explains together with a disorder induced partial mixing of different bands crossing the Fermi surface (increase of the effective Fermi velocity) also qualitatively the observed reductions of T_c, $H_{c2}(T)$ and of its positive curvature

near T_c [8,17] provided coupling to low-frequency phonons is taken into account. We speculate that the disorder discussed here affects the coupling to the soft anomalous phonons near 7 meV possibly related to nesting effects [17].

5. SUMMARY

The superconducting transition temperature of pseudo-quaternary $R_x Y_{1-x} Ni_2B_2C$ compounds with small concentration of paramagnetic rare-earth elements R is suppressed by magnetic pair-breaking and by nonmagnetic disorder effects on the rare-earth site due to the different ionic radii of the yttrium host and the R ions. Magnetic pair breaking was found to suppress T_c for the heavy rare-earth elements, whereas the strong decrease of T_c observed for the light rare-earth elements Pr and Nd is mainly caused by local lattice strains due to the large difference of the ionic radii, δr, between the Y host and the R ions resulting in a nearly quadratic dependence for the depression rate $|\delta T_c/DG| \propto (\delta r)^2$ upon δr.

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