

Influence of disorder on superconductivity in non-magnetic rare-earth nickel borocarbides

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Abstract. The effect of substitutional disorder on the superconducting properties of $\text{YNi}_2\text{B}_2\text{C}$ was studied by partially replacing yttrium and nickel by Lu and Pt, respectively. For the two series of $(\text{Y, Lu})\text{Ni}_2\text{B}_2\text{C}$ and $\text{Y}(\text{Ni, Pt})_2\text{B}_2\text{C}$ compounds, the upper critical field $H_{c2}(T)$ and the specific heat $c_p(T, H)$ in the superconducting mixed state have been investigated. Disorder is found to reduce several relevant quantities such as T_c , the upper critical field $H_{c2}(0)$ at $T = 0$ and a characteristic positive curvature of $H_{c2}(T)$ observed for these compounds near T_c . The $H_{c2}(T)$ data point to the clean limit for (Y, Lu) substitutions and to a transition to the quasi-dirty limit for (Ni, Pt) substitutions. The electronic specific heat contribution $\gamma(H)$ exhibits significant deviations from the usual linear $\gamma(H)$ law. These deviations reduce with growing substitutional disorder but remain even in the quasi-dirty limit which is reached in the $\text{Y}(\text{Ni}_{1-x}, \text{Pt}_x)_2\text{B}_2\text{C}$ samples for $x = 0.1$.

Keywords. Superconductivity; nickel borocarbides; disorder.

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1. Introduction

The rare-earth nickel borocarbide family $\text{RNi}_2\text{B}_2\text{C}$ ($\text{R} = \text{Y}$, rare-earth) [1,2] show interesting physical properties, especially by the interplay between superconductivity and magnetism in these compounds. Also the non-magnetic members of this family exhibit some features unexpected for ordinary s-wave superconductors. One of them is the unusual shape and the strong disorder dependence of the upper critical field $H_{c2}(T)$ [3].

High-quality borocarbide single crystals were found to be in the clean limit, e.g. their mean free path exceeds the coherence length of the superconductor. Therefore, $H_{c2}(T)$ is mainly determined by the electronic structure and high values of H_{c2} require clean samples, whereas for dirty limit superconductors H_{c2} increases with increasing impurity scattering. A characteristic feature of borocarbide compounds is their unusual $H_{c2}(T)$ dependence showing a positive curvature near T_c . This behavior which cannot be explained in the framework of a conventional single band model can be described successfully in

the framework of a two-band model [4] assuming two groups of electrons with different Fermi velocities. The magnitude of T_c , of $H_{c2}(0)$ at $T = 0$ and of the positive curvature of $H_{c2}(T)$ in RNi_2B_2C ($R = Y, Lu$) can be suppressed by controlled disorder created in $(Y, Lu)Ni_2B_2C$ compounds by isoelectronic substitutions at the R site. Nevertheless, the case of dirty limit was not reached even for the maximum disordered $(Y, Lu)Ni_2B_2C$ samples [5]. A higher degree of disorder can be realized by isoelectronic substitutions at the Ni site. In a $Y(Ni_{1-x}, Pt_x)_2B_2C$ single crystal with $x = 0.2$, dirty limit behavior was reported for $H_{c2}(T)$ and for the field dependence of the electronic specific heat parameter $\gamma(H)$ in the mixed state [6]. Thus, $\gamma(H) \propto H$ has been found for this single crystal, while a square-root law $\gamma(H) \propto \sqrt{H}$ was observed for a pure YNi_2B_2C single crystal and for polycrystalline $LuNi_2B_3C$ [7].

In the present paper, a series of $Y(Ni, Pt)_2B_2C$ compounds was investigated in order to analyze the transition from superconductivity in the clean to dirty limit for the first time in a broad range of Pt concentrations. Additionally, the closely related $(Y, Lu)Ni_2B_2C$ system in which isoelectronic substitutions are expected to produce much weaker disorder than those in the Ni–B network was also investigated for the sake of comparison.

2. Experimental details

Polycrystalline $Y_xLu_{1-x}Ni_2B_2C$ samples with $x = 0, 0.25, 0.5, 0.75, 1$ and $Y(Ni_{1-x}, Pt_x)_2B_2C$ samples with $x = 0, 0.05, 0.1, 0.15, 0.2, 0.5$ and 0.75 were prepared by a standard arc-melting technique. Powders of the elements were weighed in the stoichiometric compositions with a surplus of 10 wt% boron to compensate for the loss of boron caused by the arc melting. The powder was pressed into pellets which were melted under argon gas on a water-cooled copper plate in the arc furnace. To get homogeneous samples, they were turned over and melted again four times. After the melting procedure, the solidified samples were homogenized at $1100^\circ C$ for ten days.

The superconducting properties of the samples were determined by susceptibility, resistivity and specific heat measurements. The midpoint value of the normal-state resistivity and the onset temperature of the superconducting transition of the susceptibility measurement were used to determine the superconducting transition temperature T_c and the upper critical field $H_{c2}(T)$. Both methods were found to lead to the same result. The specific heat was measured in the range $4.2 K \leq T \leq 20 K$ and for magnetic fields $H \leq 8 T$ using a quasi-adiabatic step heating technique.

3. Results and discussion

3.1 Upper critical field $H_{c2}(T)$

$H_{c2}(T)$ data for both pure compounds of the $(Y_xLu_{1-x})Ni_2B_2C$ system [5,8] and for the sample with $x = 0.5$ are shown in figure 1. For these clean limit superconductors the impurity content plays a crucial role for obtaining high H_{c2} values. Therefore, the highest H_{c2} values are observed for $LuNi_2B_2C$, which has a better sample quality and a higher residual resistivity ratio $RRR = \rho(300 K)/\rho_n = 44$ than YNi_2B_2C with $RRR = 21$. By replacing Y partially by Lu and, thus, introducing substitutional disorder at the rare-earth site, the upper

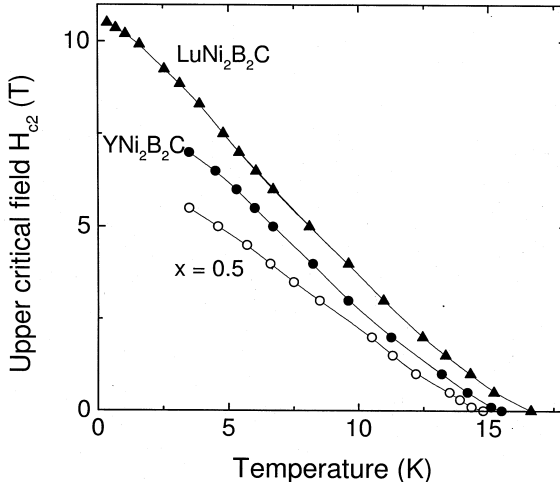


Figure 1. Temperature dependence of the upper critical field for $Y_xLu_{1-x}Ni_2B_2C$ with $x = 0, 1$ and 0.5 .

critical field is reduced and the lowest H_{c2} values are found for the compound with $x \sim 0.5$ (see figure 1). The $H_{c2}(T)$ dependence of non-magnetic borocarbides with its pronounced positive curvature near T_c can be described in a wide temperature range $0.3 \leq T/T_c \leq 1$ by the simple expression

$$H_{c2}(T) = H_{c2}^*(0)(1 - T/T_c)^{1+\alpha}. \quad (1)$$

The parameter $\alpha < 1$ measures the positive curvature near T_c and $H_{c2}^*(0) \sim 1.1 \div 1.2 H_{c2}(0)$ gives an upper bound for the true value of $H_{c2}(0)$ at $T = 0$.

The evolution of the three parameters T_c , H_{c2}^* and α in the investigated $(Y_xLu_{1-x})Ni_2B_2C$ series as a function of the Y concentration is shown in figure 2. Qualitatively, these three parameters vary in a similar way. They achieve their highest values for the pure samples and their lowest ones at about 50%, that means for the largest degree of disorder. In figure 2 the residual resistivity ratio is shown, too. It is strongly reduced by introducing a small degree of disorder. The reason for the broad plateau observed for $0.15 < x < 0.8$ is not understood up to now. A quantitative analysis shows that the sensitivity to the lattice-size disorder is most pronounced for the absolute value of $H_{c2}^*(0)$, somewhat less for α and weakest for T_c . Therefore, the parameter $H_{c2}^*(0)$ can be considered as the most sensitive measure of the perfection of the clean limit superconductor under consideration.

The temperature dependence of the upper critical field can be described in the framework of the two-band model [4] mentioned above. Using this model, the transition from the $H_{c2}(T)$ dependence with its typical positive curvature near T_c in the clean limit to the parabolic-like $H_{c2}(T)$ curve with a negative curvature near T_c in the dirty limit also can be explained [9] as shown in figure 3. Starting from the clean limit, the magnitude of $H_{c2}(0)$ and the positive curvature of $H_{c2}(T)$ near T_c are suppressed with increasing impurity scattering rate γ_{imp} . The positive curvature becomes small at above $\gamma_{imp} \sim 80$, i.e., in the quasi-dirty limit. Further increase of the impurity scattering rate approaching the

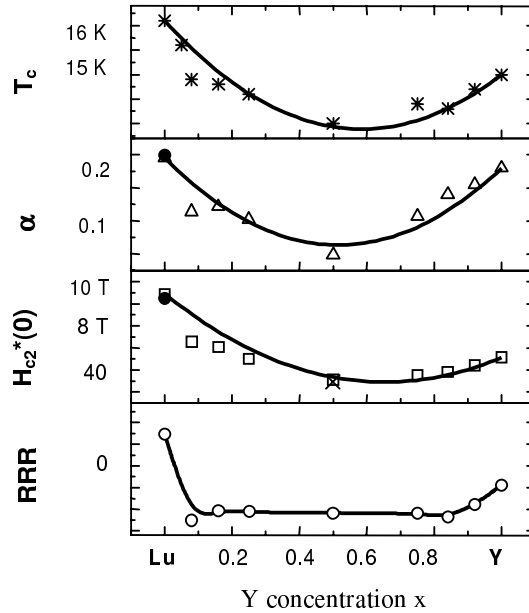


Figure 2. Concentration dependence of the superconducting transition temperature T_c , of the upper critical field parameters α describing the positive curvature of $H_{c2}(T)$ and $H_{c2}^*(0)$ (see eq. (1)) and of the resistivity ratio RRR of polycrystalline $Y_xLu_{1-x}Ni_2B_2C$.

dirty limit mainly affects the magnitude of $H_{c2}(0)$ which now begins to increase with increasing scattering rate. The comparison with the experimental data for $(Y_xLu_{1-x})Ni_2B_2C$ (see figure 2) shows that the parameter α for the positive curvature of $H_{c2}(T)$ remains positive even for the most disordered sample which indicates that this sample is not yet in the quasi-dirty limit.

It is well-known that the Ni–B network is responsible for the superconductivity in the borocarbides. Therefore, it is not surprising that $H_{c2}(T)$ is much more affected by disorder on the Ni site than by disorder in the rare-earth subsystem. This is shown in figure 4 where the influence of Lu impurities (in $(Y_xLu_{1-x})Ni_2B_2C$ compounds) and Pt impurities (in $Y(Ni_{1-x},Pt_x)_2B_2C$ compounds) on $H_{c2}(0)$ is compared. It is clearly seen that $H_{c2}(0)$ is much stronger suppressed by Pt ions on Ni sites than by Lu ions on Y sites. For the $Y(Ni_{1-x},Pt_x)_2B_2C$ compounds, the transition from clean to the quasi-dirty limit is observed at a Pt concentration of 10%, where $H_{c2}(0)$ has its lowest value. It should be noted that in the quasi-dirty limit, the positive curvature of $H_{c2}(T)$ disappears.

3.2 Specific heat

Consistent $H_{c2}(T)$ results were obtained from resistance and specific heat measurements for the investigated $(Y, Lu)Ni_2B_2C$ and $Y(Ni, Pt)_2B_2C$ series. From the specific heat $c_p(T, H)$, the electronic specific heat parameter $\gamma(H)$ in the mixed state was derived. In figure 5, normalized $\gamma(H)$ data for $Y(Ni, Pt)_2B_2C$ are shown. Large deviations from the

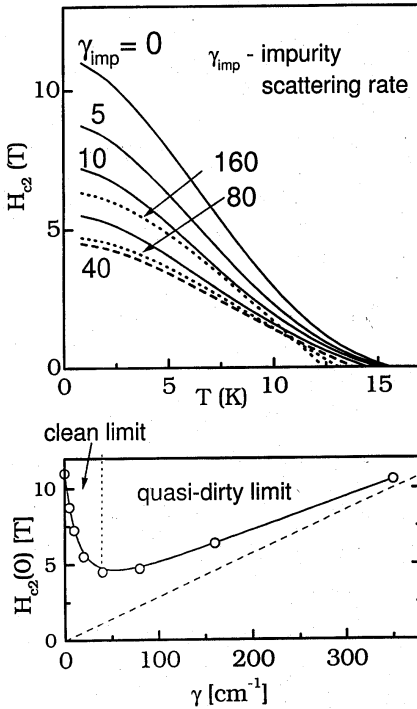


Figure 3. Temperature dependence of the upper critical field calculated within the two-band model [4] for several impurity scattering rates (upper panel). The calculated $H_{c2}(0)-\gamma_{imp}$ dependence in the lower panel illustrates the transition from the clean to the dirty limit. Dotted line: $H_{c2}(0)-\gamma_{imp}$ dependence within the dirty limit.

linear $\gamma(H)$ law expected in the dirty limit are observed, especially for the $\text{YNi}_2\text{B}_2\text{C}$ compound. These deviations become smaller for the Pt-doped samples in the quasi-dirty limit. However, they do not disappear as shown in figure 5. This peculiarity of the physics of the vortex state in the quasi-dirty limit is not understood so far. Also the basic understanding of the vortex physics in the clean limit is still far from a satisfactory level. A non-linear H dependence close to $\gamma(H) \propto \sqrt{H}$ has been reported not only in some unconventional superconductors with gap nodes in the quasi-particle spectrum of the vortex state such as $\text{YBa}_2\text{Cu}_3\text{O}_7$ [10] and the heavy fermion superconductor UPt_3 [11], but also in some clean s-wave superconductors such as CeRu_2 [12] and NbSe_2 [6,13]. Attempts to explain the unusual $\gamma(H)$ dependence of borocarbides include a shrinking of the vortex core with increasing applied field [6,13], field-induced gap nodes [12] and d-wave symmetry [14].

The $\gamma(H)$ law in conventional s-wave superconductors is linear because all quasi-particles are confined within the vortex core of radius ξ (coherence length). Therefore, the quasi-particle density of states, $N(H)$, is proportional to the number of vortices which scales with the magnetic field resulting in $N(H) \propto \xi^2 H$. Experimental data for the microwave surface impedance in the vortex state of $\text{YNi}_2\text{B}_2\text{C}$ [15] are consistent only with a linear $N(H)$ dependence. This means that the number of quasiparticles within the core is field-independent since the flux flow dissipation causing the microwave

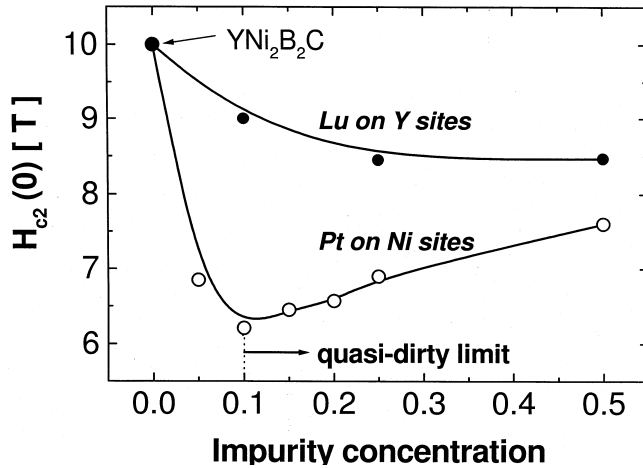


Figure 4. Suppression of the upper critical field $H_{c2}(0)$ of $\text{YNi}_2\text{B}_2\text{C}$ by Lu impurities on Y sites ($\text{Y}_x\text{Lu}_{1-x}\text{Ni}_2\text{B}_2\text{C}$ – filled circles) and by Pt impurities on Ni sites ($\text{Y}(\text{Ni}_{1-x}\text{Pt}_x)_2\text{B}_2\text{C}$ – open circles). In the latter case, the quasi-dirty limit is reached at $x = 0.1$.

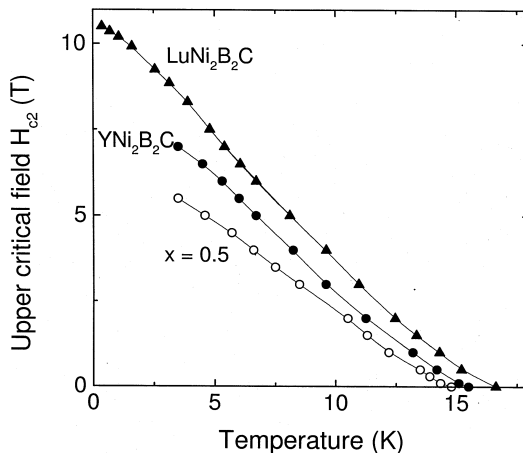


Figure 5. Magnetic field dependence of the specific heat contribution $\gamma(H)$ of the vortex core electrons in the mixed state normalized by the Sommerfeld parameter γ_N and $H_{c2}(0)$ for $\text{Y}(\text{Ni}_{1-x}\text{Pt}_x)_2\text{B}_2\text{C}$. The lines are guides for the eye and the dotted straight line corresponds to the usual linear $\gamma(H)$ law for s-wave superconductors in the dirty limit.

surface impedance mainly comes from the quasi-particles localized in the cores. This result excludes the scenario of the core shrinking with magnetic field as an origin of non-linear $\gamma(H)$ and indicates that delocalized quasi-particle states around the vortex cores, similar to d-wave superconductors, are responsible for the non-linear $\gamma(H)$ dependence of $\text{YNi}_2\text{B}_2\text{C}$

[15]. The presence of such delocalized quasiparticles has been verified in $\text{YNi}_2\text{B}_2\text{C}$ by investigations of the thermal conductivity [16].

4. Summary

Weak disorder effects have been found in $(\text{Y, Lu})\text{Ni}_2\text{B}_2\text{C}$ by isoelectronic substitutions of Y by Lu. A moderate suppression of characteristic features which are typical for the clean limit have been observed for the non-linearity of $\gamma(H)$, for the magnitude of the upper critical field $H_{c2}(0)$, the curvature exponent α of $H_{c2}(T)$ and T_c . Stronger disorder effects are observed in $\text{Y}(\text{Ni}_{1-x}, \text{Pt}_x)_2\text{B}_2\text{C}$ by isoelectronic substitutions of Ni by Pt. We deduce a transition from clean to quasi-dirty limit at a Pt concentration of $x \sim 0.1$, where $H_{c2}(0)$ has a minimum. A nearly vanishing positive curvature of $H_{c2}(T)$ is observed for $x > 0.1$, i.e., in the quasi-dirty limit. At the same time, a non-linear $\gamma(H)$ dependence is found in the quasi-dirty limit. Thus, the vortex physics remains rather complex even in the quasi-dirty limit.

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