SPECIFIC HEAT OF Y$_x$Lu$_{1-x}$Ni$_2$B$_2$C IN THE MIXED STATE

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The temperature and magnetic field dependences of the electronic specific heat $c_p$ in the superconducting mixed state as well as the upper critical field $H_{c2}(T)$ have been measured on a series of polycrystalline Y$_x$Lu$_{1-x}$Ni$_2$B$_2$C samples with the aim to study the influence of disorder. The electronic specific heat contribution $c_{esm} = \gamma(H) \cdot T$ in the mixed (vortex) state exhibits significant deviations from the usual $\gamma(H) \propto H$-law for conventional superconductors for all compositions resulting in a disorder dependent negative curvature of $\gamma \propto H^{1-\beta(x)}$. For the samples with substitutional disorder at the rare earth site $\beta$ and the positive curvature exponent $\alpha$ of $H_{c2}(T)$ are reduced but $\alpha, \beta > 0$ holds for all $x$, as distinct from the case of much stronger disorder due to iso-electronic partial Pt substitutions at the Ni site where $\alpha$ and $\beta$ rapidly vanish (Nohara et al. (1999) J. Phys. Soc. Jpn. 68 1078). $\alpha$ exhibits a clear minimum near $x = 0.5$. The magnitudes of $H_{c2}(T)$ and $T_c$, the curvatures of $H_{c2}(T)$ and $\gamma(H)$, as well as the Sommerfeld constant $\gamma_N$ in the normal state are reduced in a similar fashion for intermediate composition.

1. Introduction

The recent discovery of the rare earth transition metal borocarbide (nitride) family (RC)$_n$T$_2$B$_2$C(N), R=Y, Lu, Sc, Th, La; T=Ni, Pd, Pt; $n=1$ to 3 [1, 2] which contains superconductors with relatively high transition temperatures $T_c$ up to 23K has stimulated numerous studies of their thermodynamic and transport properties
in the superconducting as well as in the normal state. At first glance most of those results support a classification of these materials as typical intermetallic phonon mediated superconductors with a moderately strong coupling strength. However, clean RNi$_2$B$_2$C samples exhibit also some features unexpected for ordinary superconductors both in the superconducting and in the normal state. Among several peculiarities [3] we emphasize the unusual shape and the strong disorder dependence of the upper critical magnetic field $H_{c2}(T)$ and the $T^3$ dependence of the electronic specific heat $c_{es}(T)$ in the superconducting state compared with exponential behaviour for ordinary $s$-wave superconductors. According to Nohara et al. [4] isoelectronic transition metal substitution affects strongly the field dependence of the electronic specific heat contribution $c_{esm} = \gamma(H) \cdot T$ in the mixed state. While for an Y(Ni$_{0.8}$Pt$_{0.2}$)$_2$B$_2$C single crystal $\gamma(H) \propto H$, a marked sublinear law was observed for pure YNi$_2$B$_2$C single crystal and polycrystalline LuNi$_2$B$_2$C [5]

$$\frac{\gamma(H)}{\gamma_N} \propto \sqrt{H/H_{c2}(0)},$$

where $\gamma_N$ denotes the Sommerfeld constant in the normal state. Thus already small substitutions in the TB network (which is commonly believed to be the crucial locus for the superconductivity) affect significantly the superconducting properties, presumably due to the predominant role of the T-derived $d$ states in the mechanism of superconductivity. At the same time by isoelectronic substitutions in the RC charge reservoir much weaker disorder could be studied (see Fuchs et al., Drechsler et al., these Proc.). The remarkable $\gamma(H) \propto \sqrt{H}$-law for YNi$_2$B$_2$C and LuNi$_2$B$_2$C was regarded at first as evidence for $d$-wave pairing [5]-[7]. In this context it is interesting to study also the closely related Y$_x$Lu$_{1-x}$Ni$_2$B$_2$C system as an appropriate weak disorder case. Thus changing the composition $x$, more detailed information should be gained about the influence of weak disorder upon the field dependence of the specific heat $c_p(T, H)$, the shape and the magnitude of $H_{c2}(T)$, as well as about the nature of the pairing state.

2. Experimental Details

Polycrystalline Y$_x$Lu$_{1-x}$Ni$_2$B$_2$C samples were prepared by a standard arc melting technique. Powders of the elements were weighted in the stoichiometric compositions with a surplus of 10 wt.% boron to compensate the high losses of boron caused by the arc melting. The powder was pressed to pellets which were melted under argon gas on a water-cooled copper plate in an arc furnace. To get homogeneous samples, they were turned over and melted again four times. After the melting procedure the solidified samples were homogenised at 1100°C for ten days. The specific heat was measured in the range 4.2K $\leq T \leq$ 20K and for magnetic fields $H \leq$ 8T using a quasi adiabatic step heating technique. The heating pulses were generated by a strain gauge heater and $T$ was measured with an Au-Ge thin film resistor.
3. Results and Discussion

The specific heat $c_p/T$ vs. $T^2$ at $H = 0$ of a $Y_xLu_{1-x}Ni_2B_2C$ series is shown in Fig. 1. The corresponding curves for $H \leq 8T$ of the pure $Y$ sample ($x = 1$) are depicted in Fig. 2. Applying $H = 8T$ resulted in a shift of $T_c$ below the lowest measured temperature of 4.2K. Measurements in this field were used to analyze the normal state specific heat $c_p = \gamma_N T + \beta_D T^3$, where $\gamma_N$ is the Sommerfeld constant and $\beta_D T^3$ is the Debye contribution. The $\gamma_N$ was determined by extrapo-
the data reported by several groups [9]-[13]. To determine \( \gamma(H) \) ≤ \( T \approx 20.2 \text{mJ/molK} \), slightly exceed those derived from Eq. (1) as reported in Refs. [4, 5]. Anyhow, \( \beta \) has been extrapolated linearly to \( \gamma(H) )/\gamma_N \propto (H/H_{c2,\text{eff}}(0))^{1-\beta} \). Noteworthy, the case \( \beta = 0.31 \) to 0.35 and \( H/H_{c2,\text{eff}}(0) \leq 0.75 \) is not very distinct from the dirty (unitary) d-wave limit where \( \gamma(H) \propto H \ln H \) according to Ref. [17] (not shown here).

Figure 3. Magnetic field dependence of the specific heat contribution \( \gamma(H) \) of the vortex core electrons in the mixed state \( (H \leq H_{c2}) \) in units of the effective upper critical field \( H_{c2,\text{eff}}(0) \) shown in Fig. 4. The straight reference line corresponds to the usual s-wave dirty limit behaviour. The inset shows the curvature parameter \( \beta \) of \( \gamma(H) \) according to \( \gamma(H)/\gamma_N \propto (H/H_{c2,\text{eff}}(0))^{1-\beta} \). Noteworthy, the case \( \beta = 0.31 \) to 0.35 and \( H/H_{c2,\text{eff}}(0) \leq 0.75 \) is not very distinct from the dirty (unitary) d-wave limit where \( \gamma(H) \propto H \ln H \) according to Ref. [17] (not shown here).

lating the \( c_p/T \) vs \( T^2 \) curves to \( T = 0 \) in the normal state. This way we obtained \( \gamma_N = 20.4 \) \((x = 0)\), 19 \((x = 0.25)\), 18.3 \((x = 0.5)\), 18 \((x = 0.75)\) and 20.2mJ/molK \((x = 1)\) for our \( Y_x Lu_{1-x} Ni_2 B_2 C \) series in good agreement with the data reported by several groups [9]-[13]. To determine \( \gamma(H) \), the \( c_p/T \) vs. \( T^2 \) curves have been extrapolated linearly to \( T = 0 \) from the data in the range 4.2K ≤ \( T \leq 7 \)K or up to the onset of the transition to the normal state. The results are shown in Fig. 3. Apparently, for all \( Y_x Lu_{1-x} Ni_2 B_2 C \) samples \( \gamma(H) \) is a sublinear function on \( H \). Generalizing Eq. (1), the data were analyzed by the expression

\[
\gamma(H)/\gamma_N = (H/H_{c2,\text{eff}}(0))^{1-\beta(x)},
\]

where the fitting parameter \( \beta \) measures the negative curvature of \( \gamma(H) \) while \( H_{c2,\text{eff}}(0) \approx (0.8 \text{ to } 0.9)H_{c2}(0) \) yields a lower bound for the true \( H_{c2}(0) \) (see Fig. 4). We obtained \( \beta = 0.66 \), 0.42, 0.42, 0.41 and 0.56 from \( x = 0 \) to \( x = 1 \), respectively. The behaviour of \( \beta(x) \) is shown in the inset of Fig. 3. The curvature of \( \gamma(H) \) becomes maximal for the bordering cases \( x = 0; 1 \) and markedly smaller in between. Noteworthy, our curvatures for \( Lu Ni_2 B_2 C \) and \( YNi_2 B_2 C \) do slightly exceed those derived from Eq. (1) as reported in Refs. [4, 5]. Anyhow, \( \gamma(H) \) shows for all \( Y_x Lu_{1-x} Ni_2 B_2 C \) samples a clear \( H \) sublinear dependence. A \( \gamma(H) \approx \sqrt{H} \)-law as well as the \( c_{es}(T) \propto T^3 \) dependence raise the question whether an unconventional pairing mechanism is responsible for this peculiarity.
According to Ref. [6] $\gamma(H) \propto \sqrt{H}$ is a signature for a nodal order parameter with $d$-wave pairing (for which $\beta = 0.57$ has been predicted in Ref. [14]) while $\gamma(H) \propto H$ points to nodeless $s$-wave pairing in the dirty limit since there are conventional superconductors which exhibit also deviations from the $\gamma(H) \propto H$-law in the clean limit, e.g. V$_3$Si [15], NbSe$_2$[4] ($\beta = 0.33$), and CeRu$_2$[14]. Hence, the sublinear $\gamma(H)$ dependences observed here are not inevitably related to $d$-wave pairing [16].

Since the lattice constants vary linearly between $x = 0$ and $x = 1$, one might expect a maximum of $T_c$ around $x = 0.6$ according to an “universal” curve $T_c = T_c(d_{Ni-Ni})$ proposed by Lai et al. [18], where $d_{Ni-Ni}$ is the Ni-Ni distance. However, in accord with Refs. [8,9] a dip near $x = 0.7$ is observed ($T_c \approx 14.6$K at $x = 0.75$). A similar behaviour is found for $H_{c2}(0)$ and also for the curvature exponent $\alpha$ (see Fig. 4):

$$H_{c2}(T) = H_{c2}^* (1 - T/T_c)^{1+\alpha}, \quad \text{valid for} \quad 0.3 \leq T/T_c,$$

(3)

(distinct from a statement of a constant curvature [9]). $H_{c2}^* \approx 1.1 \div 1.2H_{c2}(0)$ gives an upper bound for the true value $H_{c2}(0)$ at $T = 0$. Ongoing from pure samples in the clean limit to the mixed specimens which remain still in the quasi-clean limit, all these quantities are somewhat reduced due to weak disorder at

Figure 4. Composition dependence of the transition temperature $T_c$ derived from the onset of the jump of $c_P(T)$ (upper panel), the Sommerfeld constant $\gamma_N$ (second panel), the upper (lower) bound for the upper critical field (third panel, see Eqs. (2,3) and text), the specific heat curvature exponent $\beta$ of $\gamma(H)$ (fourth panel), and the curvature exponent $\alpha$ of the upper critical field $H_{c2}$ (lower panel).
the R-sites as well as due to a slight reduction of the electron-phonon coupling constant $\lambda$ at intermediate $x$ deduced from the similar behavior of $\gamma_N$ [19, 20] (see Fuchs et al., these Proc.).

To summarize, we have shown that the $\gamma(H)$ curves of the pure specimens ($x = 0; 1$) of $Y_xLu_{1-x}Ni_2B_2C$ exhibit the highest negative curvatures. Weak disorder effects, caused by isoelectronic substitutions of Y by Lu, yield a decrease of the curvature without reaching the linear limit, which would be the hallmark for the dirty limit. Similar moderate suppressions which are typical for the quasi-clean limit have been found for the upper critical field, its curvature, and $T_c$.

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The temperature and magnetic field dependences of the electronic specific heat in the superconducting mixed state as well as the upper critical field have been measured on a (Y,Lu)Ni2B2C series to study the influence of disorder. The electronic specific heat in the vortex state shows deviations from the usual gamma (H) proportional to H-law for conventional superconductors for all samples resulting in a disorder dependent negative curvature of gamma proportional to H^1-beta). For samples with disorder at the R site beta and the positive curvature exponent alpha of H_{sub(c2)} are reduced but alpha, beta remain always positive, as distinct from much stronger disorder due to Pt substitutions at the Ni site where alpha, beta rapidly tend to 0. Alpha shows a minimum near x = 0.5. H_{sub(c2)}(0), Tc, the curvatures of H_{sub(c2)} and gamma(H), as well as the normal state Sommerfeld constant gamma sub(N) are reduced similarly.

Keywords: Specific heat, upper critical field, dirty limit, clean limit, Sommerfeld constant