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Specific heat and disorder in the mixed state of non-magnetic borocarbides

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Abstract. – The temperature and magnetic-field dependence of the specific heat $c_p(T, H)$ in the superconducting (sc) mixed state as well as the upper critical field $H_{c2}(T)$ have been measured for polycrystalline $Y_xLu_{1-x}Ni_2B_2C$ and $Y(Ni_{1-y}Pt_y)_2B_2C$ samples. The linear-in-T electronic specific-heat contribution $\gamma(H) \cdot T$ exhibits significant deviations from the usual linear-in-H law resulting in a disorder-dependent negative curvature of $\gamma(H)$. The $H_{c2}(T)$ data point to the quasi-clean limit for (Y, Lu)-substitutions and to a transition to the quasi-dirty limit for (Ni, Pt)-substitutions. The $\gamma(H)$-dependence is discussed in the unitary d-wave as well as in the quasi-clean s-wave limits. From a consideration of $\gamma(H)$ data only, d-wave pairing cannot be ruled out.

Introduction. – The rare-earth (R) transition metal (T) borocarbide family RCT_2B_2 ($R = Y, Lu$; $T = Ni, Pd, Pt$) contains superconductors with relatively high transition temperatures T_c up to 23 K [1,2]. The coexistence of superconductivity and magnetism for members of this family, where R are magnetic rare-earth ions, has stimulated numerous studies of their thermodynamic properties in the sc and in the normal state. At first glance, most of those results support a classification of these materials as intermetallic phonon-mediated superconductors with a moderately strong coupling strength. However, clean RNi_2B_2C samples exhibit also some features unexpected for ordinary s-wave superconductors. We emphasize the unusual shape and the strong disorder dependence of the upper critical field $H_{c2}(T)$ and a nearly T^3-scaling of the electronic specific heat $c_{es}(T)$ in the sc state compared with exponential behaviour for ordinary s-wave superconductors [3].

According to Nohara *et al.* [4] the isoelectronic T-substitution does affect strongly the field dependence of the linear-in-T electronic specific-heat contribution $\gamma(H) \cdot T$ in the mixed state.

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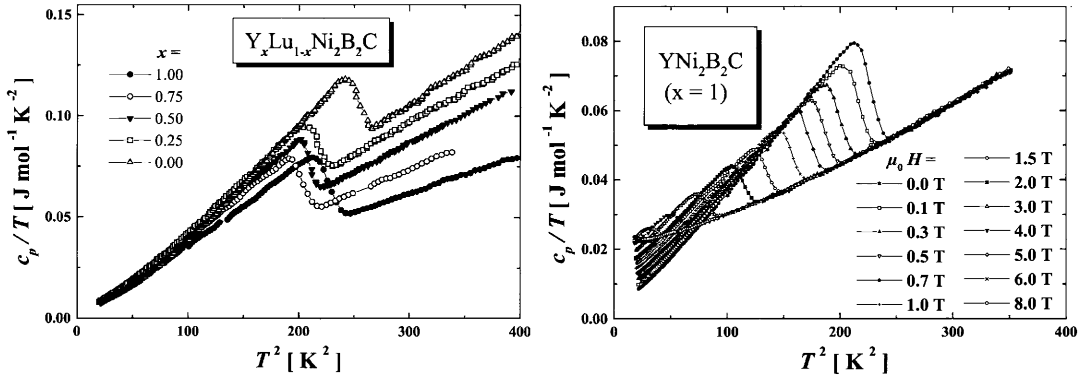

Fig. 1 – Zero-magnetic-field specific heat $c_p(T)/T$ *vs.* T^2 of the $Y_xLu_{1-x}Ni_2B_2C$ series (left panel) and specific heat $c_p(T,H)/T$ *vs.* T^2 of YNi_2B_2C for various magnetic fields (right).

Thus, for an $Y(Ni_{0.8}Pt_{0.2})_2B_2C$ single crystal, $\gamma(H) \propto H$ has been found, while a square-root law was observed for a pure YNi_2B_2C single crystal and for polycrystalline $LuNi_2B_2C$ [5]:

$$\gamma(H)/\gamma_{\mathrm{N}} \propto \sqrt{H/H_{\mathrm{c2}}(0)}, \qquad (1)$$

where γ_{N} is the Sommerfeld constant in the normal state. Although the observed $\gamma(H) \propto \sqrt{H}$-law for YNi_2B_2C and $LuNi_2B_2C$ was regarded initially as evidence for d-wave pairing [5, 6], the disorder-related transition from a $\sqrt{H}$ to a linear-in-H dependence was subsequently used to rule out d-wave superconductivity in non-magnetic borocarbides [4]. However, to the best of our knowledge, systematic investigations of this problem in a broader concentration range for $Y(Ni_{1-y}Pt_y)_2B_2C$ are lacking. Since isoelectronic substitutions in the RC charge reservoir are expected to produce much weaker disorder than those in the TB network, we studied also the closely related $Y_xLu_{1-x}Ni_2B_2C$ system for the sake of comparison [7]. By changing both compositions, x and y, deeper insight should be gained on how the disorder does affect the field dependence of the specific heat $c_p(T,H)$, the shape, and the magnitude of $H_{\mathrm{c2}}(T)$, as well as the nature of the pairing state. It has recently been pointed out that possibly an unconventional mechanism is responsible for superconductivity in borocarbides [8].

Experimental details. – Polycrystalline $Y_xLu_{1-x}Ni_2B_2C$ with $x = 0$, 0.25, 0.5, 0.75, 1, and $Y(Ni_{1-y}Pt_y)_2B_2C$ samples with $y = 0$, 0.05, 0.1, 0.15, 0.2, 0.25, 0.5, and 0.75 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 losses of boron during arc melting. The powder was pressed to pellets which were melted in 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 between $4.2\,\mathrm{K} \leq T \leq 20\,\mathrm{K}$ increasing the temperature after the samples were cooled down from $T > T_{\mathrm{c}}$ in applied fields $\mu_0 H \leq 8\,\mathrm{T}$ using a quasi-adiabatic step heating technique [9]. The upper critical field $H_{\mathrm{c2}}(T)$ was determined by taking $T_{\mathrm{c}}(H)$ from the onset of the jump of c_p in the particular field.

Results and discussion. – To illustrate typical specific-heat behaviour, the c_p/T *vs.* T^2 data at $H = 0$ of the $Y_xLu_{1-x}Ni_2B_2C$ series and the corresponding curves for $\mu_0 H \leq 8\,\mathrm{T}$ of the pure Y sample ($x = 1$) are shown in fig. 1. Measurements at 8 T were used to analyse the normal-state specific heat $c_p = \gamma_{\mathrm{N}} T + \beta_{\mathrm{D}} T^3$, where $\beta_{\mathrm{D}} T^3$ is the Debye contribution. The Sommerfeld values γ_{N} were determined by extrapolating the c_p/T *vs.* T^2 curves of the high

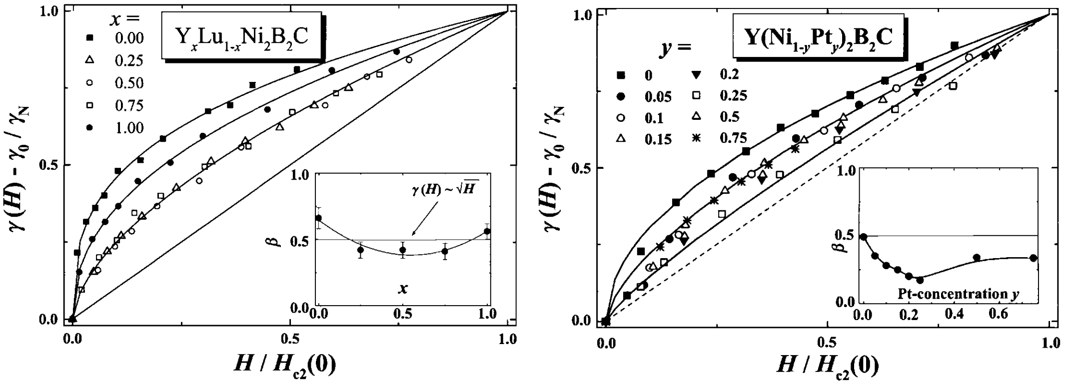

Fig. 2 – Magnetic-field dependence of the specific-heat contribution $\gamma(H)$ of the vortex core electrons in the mixed state ($H \leq H_{c2}$) normalized by γ_N and $H_{c2}(0)$ (see fig. 4) for $Y_xLu_{1-x}Ni_2B_2C$ (left panel) and $Y(Ni_{1-y}Pt_y)_2B_2C$ (right panel). Residual $\gamma_0 = \gamma\,(H = 0)$ values have been subtracted. The lines are fits according to eq. (2) and the straight reference line corresponds to the usual linear-in-H s-wave dirty-limit behaviour. The insets show the curvature parameters $\beta(x)$ and $\beta(y)$ as defined in eq. (2).

field data in the normal state to $T \to 0$. By integrating specific-heat differences between the sc and normal state $(c_s(T, 0\text{ tesla}) - c_n(T, 8\text{ tesla}))/T$ from T_c down to a temperature $T < T_c$ the entropy conservation was checked resulting in the vanishing entropy difference between sc and normal state $S_s - S_n$ for $T \to 0$ and for $T \to T_c$ and in a minimum in between. In this way we obtained $\gamma_N = 20.4$ ($x = 0$), 19.0 ($x = 0.25$), 18.3 ($x = 0.5$), 18.0 ($x = 0.75$), and $20.2\,\mathrm{mJ/mol\,K^2}$ ($x = 1$) for our $Y_xLu_{1-x}Ni_2B_2C$ series in good agreement with the data reported previously by several groups [10–16] and $\gamma_N = 20.2$ ($y = 0$), 20.2 ($y = 0.05$), 18.4 ($y = 0.1$), 16.4 ($y = 0.15$), 16.2 ($y = 0.2$), 16.9 ($y = 0.25$), 15.3 ($y = 0.5$), and $15.0\,\mathrm{mJ/mol\,K^2}$ ($y = 0.75$) for the $Y(Ni_{1-y}Pt_y)_2B_2C$ series. The $\gamma(H)$-values obtained in the same way as the Sommerfeld values γ_N are represented in fig. 2. The entropy is conserved in applied fields, too. For all samples $\gamma(H)$ is a sublinear function of H. At first generalizing eq. (1), the data were analysed by the expression

$$\frac{\gamma(H) - \gamma_0}{\gamma_N} = \left[H/H_{c2}(0)\right]^{1-\beta}, \qquad (2)$$

where $\gamma_0 = \gamma(H = 0)$ specifies the linear-in-T contribution observed in the zero field and β measures the sublinearity (*i.e.* a negative curvature) of $\gamma(H)$. $H_{c2}(0)$ is the field where $\gamma(H)$ reaches γ_N. We obtained $\beta = 0.66$, 0.42, 0.42, 0.41, and 0.56 ongoing from $x = 0$ to $x = 1$ for $Y_xLu_{1-x}Ni_2B_2C$ and $\beta = 0.46$, 0.35, 0.25, 0.25, 0.21, 0.17, 0.34, and 0.33 ongoing from $y = 0$ to $y = 0.75$ for $Y(Ni_{1-y}Pt_y)_2B_2C$, with uncertainties of $\Delta\beta/\beta \leq 10\%$ due to the small residual γ_0 and due to the procedure used to determine $\gamma(H)$, as mentioned above. For all our samples but $Y(Ni_{0.75}Pt_{0.25})_2B_2C$ and $Y(Ni_{0.5}Pt_{0.5})_2B_2$, residual values $\gamma_0 \leq 1.5\,\mathrm{mJ/mol\,K^2}$ are observed. For $y = 0.25$ and 0.5 we find $\gamma_0 = 3.4$ and $3.3\,\mathrm{mJ/mol\,K^2}$, respectively.

The dependence of $\beta(x)$ is shown in the inset of fig. 2 and in fig. 4 (left panel). β reaches the largest values for the bordering cases $x = 0$ and 1 and becomes markedly smaller in between. We note that our curvatures for $LuNi_2B_2C$ and YNi_2B_2C exceed slightly the value of $\beta = 0.5$ suggested by eq. (1) and that reported in refs. [4,5]. To the best of our knowledge, the strong sublinearities for $\gamma(H)$, measured by the exponent β, of the borocarbides under consideration are the largest reported so far for any superconductor except for the recently

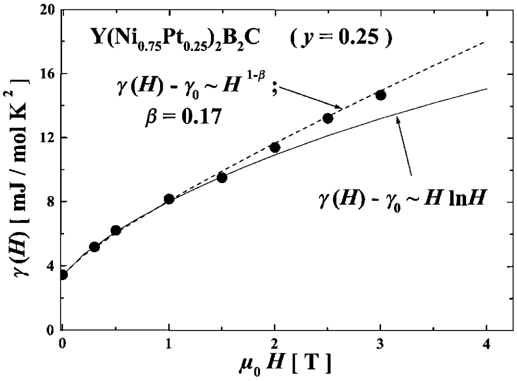

Fig. 3 – Magnetic-field dependence of $\gamma(H)$ for $\mathrm{Y(Ni_{0.75}Pt_{0.25})_2B_2C}$. The solid line is a fit according to eq. (3). The dashed line is a fit according to eq. (2) with $\beta = 0.17$.

discovered $\mathrm{MgB_2}$ ($\beta = 0.77$) [17]. The $\beta(y)$ behaviour is depicted in the inset of fig. 2 and in fig. 4 (right panel). The curvature parameter β is significantly reduced with increasing Pt concentrations y for $y \leq 0.25$ and $\beta(y)$ exhibits a *finite* minimum at about $y = 0.25$ which is at variance with the linear law for an $\mathrm{Y(Ni_{0.8}Pt_{0.2})_2B_2C}$ single crystal reported in ref. [4]. We attribute that observation to a stronger disorder compared with our samples. Larger Pt concentrations ($y > 0.25$) reveal even more pronounced β values, *e.g.* $y = 0.5$; $\beta = 0.34$ and β saturates at this value.

The observed $\gamma(H) \propto H^{1-\beta}$-law with $\beta \approx 0.5$ raises the question whether an unconventional pairing mechanism is responsible for this peculiarity since, according to ref. [18], $\gamma(H) \propto \sqrt{H}$ is a signature for a nodal order parameter with d-wave symmetry (a somewhat larger value $\beta = 0.59$ has been found in ref. [19]) while $\gamma(H) \propto H$ is usually expected for superconductors with isotropic s-wave order parameter. According to refs. [20, 21], Volovik's clean-limit d-wave approach can be generalized to describe also strong impurity scattering. Then at low magnetic fields $H \ll H_{c2}(0)$ the specific-heat coefficient $\gamma(H)$ follows an $H \ln H$-dependence:

$$\gamma(H) = \gamma_0 + \gamma_{\mathrm{N}} D\left(\frac{H}{H_{c2}(0)}\right) \ln\left[\frac{\pi}{2a^2}\left(\frac{H_{c2}(0)}{H}\right)\right], \tag{3}$$

where a and D are constants. Such a behaviour was observed for various disordered high-T_c cuprates and considered as evidence for d-wave superconductivity in the unitary scattering limit [22, 23]. At the same time its applicability to non-magnetic borocarbides under consideration was disclaimed [22]. However, some of our data can be described equally well by eq. (3) for $H/H_{c2}(0) \leq 0.3$ as well as by eq. (2) using intermediate values for β (0.15 to 0.35). This is shown in fig. 3: obviously, the $H \ln H$ behaviour is not very distinct from the power law at low fields $\mu_0 H \leq 1.5\,\mathrm{T}$. At higher fields the $H \ln H$-dependence may deviate since it was derived for low fields only [20]. The existence of a non-negligible γ_0 is a feature predicted for a d-wave order parameter in the unitary limit [24] (large $\gamma_0 \approx 3.3\,\mathrm{mJ/mol\,K^2}$ are obtained for $y = 0.25$ and $y = 0.5$). Hence, d-wave pairing cannot be ruled out in non-magnetic borocarbides by considering $\gamma(H)$ data only. While the deviation from the linearity of $\gamma(H)$ is frequently ascribed to a shrinking of the vortex cores with magnetic field and to vortex core interactions [4, 25, 26], recent investigations support the assumption of delocalized quasiparticle states around the vortex core to be responsible for this feature, in a similar way as in d-wave superconductors [27]. However, there are several conventional, but anisotropic s-wave superconductors which also exhibit deviations from the $\gamma(H) \propto H$ law in the clean

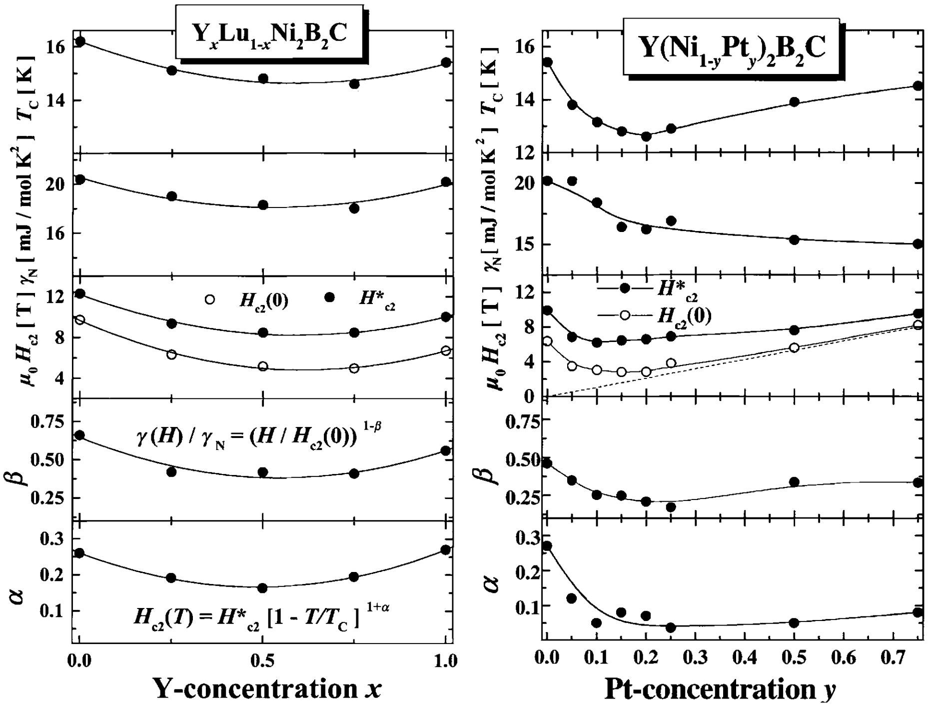

Fig. 4 – Composition dependence of the transition temperature T_c (upper panels), the Sommerfeld constant γ_N (second-row panels), the upper bound for the upper critical field H^*_{c2} according to eq. (4) and $H_{c2}(0)$ according to eq. (2) (third-row panels; see text for more details), the specific-heat curvature exponent β of $\gamma(H)$ according to eq. (2) (fourth-row panels), and the curvature exponent α of the upper critical field H_{c2} according to eq. (4) (lower panels) determined for $Y_xLu_{1-x}Ni_2B_2C$ (left) and for $Y(Ni_{1-y}Pt_y)_2B_2C$ (right). The lines are guides to the eye.

limit, *e.g.* V_3Si [28], $NbSe_2$ [4] ($\beta = 0.33$), and $CeRu_2$ [19,29]. Remarkably, a sublinear $\gamma(H)$ behaviour has been reported also for the novel "medium-T_c" superconductor MgB_2 [17,30]. In this general context, recent ultrahigh-resolution photoemission spectroscopy measurements suggest that a highly anisotropic gap might be responsible for the above-mentioned peculiarities in clean Ni borocarbides [31]. By introducing disorder due to Pt substitution ($y = 0.2$), a complete isotropization of the gap was observed. The highly anisotropic gap function is corroborated by magnetic-field–dependent thermal-conductivity data for $LuNi_2B_2C$ in the mixed state [8]. The gap minimum $\Delta_{\min}$ was reported to be at least 10 times smaller than the gap maximum Δ_0, $\Delta_{\min} \leq \Delta_0/10$, and possibly going to zero at nodes [8]. Calculations of the density of states (DOS) at the Fermi level, $N(0)$, in the mixed state with interacting vortices revealed a $H^{0.67}$ ($\beta = 0.33$) dependence of $\gamma(H)$ for anisotropic s-wave superconductors [19].

Like $\gamma(H)$, the upper critical field $H_{c2}(T)$ can be described also by a simple scaling law [32]:

$$H_{c2}(T) = H^*_{c2}(1 - T/T_c)^{1+\alpha}, \quad \text{valid for } 0.3 \leq T/T_c. \tag{4}$$

Our values of the upper critical field $H_{c2}(0) \approx 0.9H^*_{c2}$ are reduced due to R-site substitution. A similar behaviour was found for the pronounced positive curvature of $H_{c2}(T)$ near T_c, which is measured by the exponent α in eq. (4), in contrast to the opposite statement of a nearly constant curvature [10]. The unusual positive curvature of $H_{c2}(T)$ near T_c observed here can be explained for superconductors in the clean limit by a significant dispersion of the

Fermi velocities using, *e.g.*, an effective two-band model [33]. T_c and γ_N are reduced to a smaller extent, which has been ascribed to a slight reduction of the electron-phonon coupling constant λ at intermediate x [34, 35]. For T_c a dip near $x = 0.7$ is observed, in accordance with refs. [10, 32] ($T_c \approx 14.6\,\mathrm{K}$ at $x = 0.75$). The dirty-limit region is not reached (which would be represented by vanishing α and increasing $H_{c2}(0)$ with increasing disorder [11, 12]).

In the case of Pt substitutions in the investigated range $0 < y < 0.75$, the values of T_c, β, and $H_{c2}(0)$ are reduced, too. As for R-substitutions, those sc properties exhibit minima at intermediate composition while the Sommerfeld constant γ_N and the curvature parameter α of $H_{c2}(T)$ depend monotonously on y. For $y < 0.2$ a strong decrease of α with increasing y is observed, but for $y > 0.2$ an increase of α does not occur (see fig. 4). This behaviour of H_{c2} suggests that the quasi-dirty limit has been reached at about $y \approx 0.2$ since $H_{c2}(0)$ increases linearly with y above $y \approx 0.2$, while the curvature of $H_{c2}(T)$ measured by α remains strongly reduced. In the dirty-limit case $H_{c2}(0)$ is expected to increase linearly with the degree of disorder [11]. The results obtained for $\mathrm{Y(Ni_{1-y}Pt_y)_2B_2C}$ show that the deviations from the linearity of $\gamma(H)$ measured by β are not correlated with the field exponent α. While α almost vanishes, β does increase for $y > 0.25$. Thus, here the behaviour of the specific heat in the vortex state even in the quasi-dirty limit remains rather complex.

To summarize, the deviations from the linear $\gamma(H)$ behaviour we observe for the pure specimens of $\mathrm{Y_xLu_{1-x}Ni_2B_2C}$ ($x = 0; 1$) are only exceeded for the recently discovered $\mathrm{MgB_2}$ superconductor. Weak disorder effects caused by isoelectronic substitutions of Lu by Y yield a reduction of the $\gamma(H)$-nonlinearity without reaching the standard linear behaviour. Similar moderate suppressions of characteristic features which are typical for the quasi-clean limit have been found for the upper critical field $H_{c2}(0)$, the curvature exponent α, γ_N, and T_c. Stronger disorder effects are caused by isoelectronic substitutions of Ni by Pt. From the behaviour of $H_{c2}(T)$ we deduce a transition from clean to quasi-dirty limit caused by isoelectronical substitutions at the T-site. The quasi-dirty limit is concluded from the nearly vanishing curvature of $H_{c2}(T)$ and from the approximately linear increase of $H_{c2}(0)$ with y for $y \geq 0.2$. At the same time there the sublinearity of $\gamma(H)$ remains and does even increase. Hence, a simple monotonous relationship between α and β, as one might expect by considering the results on $\mathrm{Y_xLu_{1-x}Ni_2B_2C}$ only, does not hold in the quasi-dirty limit. In the case of intermediate deviations from the linearity of $\gamma(H)$ ($\beta = 0.15$–0.35), our results on specific heat at low magnetic fields are discussed in the context of a dirty d-wave model on the one hand and within the framework of the conventional s-wave picture in the quasi-clean limit on the other hand. At low fields the $H \ln H$-dependence of $\gamma(H)$ predicted for d-wave pairing in the dirty (unitary) limit is not very distinct from the $H^{1-\beta}$ behaviour which favours s-wave superconductivity in the quasi-clean limit. Thus, considering results on $\gamma(H)$ a possible unconventional pairing mechanism in borocarbide superconductors cannot be ruled out.

Additional Remark. – Recently, the sublinear H-dependence of $\gamma(H)$ has been addressed theoretically for a clean s-wave two-band superconductor [36]. It was found that β depends sensitively on the ratio of the two gaps of the strongly and weakly coupled bands. That appealing picture proposed for $\mathrm{MgB_2}$ might be transferred also to borocarbides under consideration. Then the two-band character manifests itself by two unusual curvature exponents α and β.

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