Upper critical field peculiarities of superconducting YNi$_2$B$_2$C and LuNi$_2$B$_2$C

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We present new upper critical field $H_{c2}(T)$ data in a broad temperature region $0.3K \leq T \leq T_c$ for LuNi$_2$B$_2$C and YNi$_2$B$_2$C single crystals with well characterized low impurity scattering rates. The absolute values for all $T$, in particular $H_{c2}(0)$, and the sizeable positive curvature (PC) of $H_{c2}(T)$ at high and intermediate $T$ are explained quantitatively within an effective two-band model. The failure of the isotropic single band approach is discussed in detail. Supported by de Haas van Alphen data, the superconductivity reveals direct insight into details of the electronic structure.

The observed maximal PC near $T_c$ gives strong evidence for clean limit type II superconductors.

74.60.Ec, 74.70.Ad, 74.20.-z, 74.72Ny

FIG. 1. Experimental data for $H_{c2}(T)$ of LuNi$_2$B$_2$C (magnetic field $\vec{H}$ || the c-axis) compared with theoretical curves: (i) the isotropic single band (ISB) model with Fermi velocity $v_F=2.76 \cdot 10^7$ cm/s and various impurity scattering rates $\gamma_{imp}$ explained in the text and the legends and (ii) the two-band model (TBM) with $v_{Fi}$ ($i=1,2$) in units of $10^7$ cm/s.
field ac susceptibility \( \chi(T) \) with transition widths \( \Delta T_c = 0.2 \) K. The upper critical field \( H_{c2}(T) \) along the c-axis, shown in Figs. 1 and 2, has been measured resistively for fixed \( T \) adopting the midpoint criterion: \( \rho(H_{c2}, T) = 0.5\rho(0.7 T = 17 K) \approx 0.5\rho_n \). The transition width \( \Delta H = |H(\rho = 0.9\rho_n) - H(\rho = 0.1\rho_n)| \) increases up to 0.75 T (1.5 T for LuNi$_2$B$_2$C) at \( T < 0.5 \) K starting from a nearly constant value of 0.3 T at \( T > 4 \) K (8 K for LuNi$_2$B$_2$C). The low residual resistivity \( \rho(0) \approx 2.5\mu\Omega \) cm and the ratio \( \rho(300 K)/\rho_n = 43 \) (27 for LuNi$_2$B$_2$C), together with the observations of magnetoquantum oscillations \( \lambda \) indicate a high quality and a low impurity content of our samples. This suggests that we are in the clean limit in terms of the traditional theory of type II superconductors \cite{20}. In this limit one has to consider the electronic structure in more detail. We restrict ourselves to an effective two-band model \cite{21} which, especially on the simple BCS-level, has a long history \cite{22}. Due to the neglect of strong coupling effects, a BCS-like theory is not expected to describe real superconductors quantitatively. Such effects must be studied within the Eliashberg theory \cite{24,27}. To calculate \( H_{c2} \) we have solved numerically the corresponding linearized equations of Ref. 23,

\[
\bar{\omega}_i(n) = \omega_n + \pi T \sum_{j,m} \left[ \lambda_{i,j}(m-n) + \delta_{mn} (\gamma^m_{imp,i,j} + \gamma^m_{imp,j,i}) / 2 \pi T \right] \text{sgn}(\omega_m),
\]

\[
\bar{\Delta}_i(n) = \pi T \sum_{j,m} \lambda_{i,j}(m-n) - \mu^r \delta_{ij} \theta(\omega_n - |\omega_m|) + \delta_{mn} (\gamma^m_{imp,j,i} - \gamma^m_{imp,i,j}) / 2 \pi T |\chi_j(m)| \Delta_j(m),
\]

\[
\chi(n) = (2/\sqrt{\beta_1}) \int_0^\infty dq \exp(-q^2) \times \tan^{-1}(q/\sqrt{\beta_1} |\bar{\omega}_i(n)| + i\mu_B H_{c2} \text{sgn}(\omega_n)),
\]

\[
\beta_i = eH_{c2}/v_F^2/2,
\]

\[
\lambda_{i,j}(n) = \int_0^\infty d\omega \alpha_{ij}^2 F(\omega) / (\omega^2 + \omega_n^2).
\]

The bands at \( E_F \) are labeled by \( i,j \). Here \( \omega_n = 2\pi T (2n+1) \) are the Matsubara frequencies, \( \alpha_{ij}^2 F(\omega) \) and \( \Delta_i \), denote the spectral density, the superconducting order parameter of the \( i \)th band respectively. In our approach, as in any two-band model, two gaps, below and above the BCS-value of 3.5\( k_B T_c \), occur naturally. In general, interband coupling \( (i \neq j) \) mediated by phonons \( (\alpha_{ij}^2 F(\omega)) \) and impurities is important. Since there is no experimental evidence \cite{23,24} for the presence of magnetic impurities in high quality samples, we neglect the magnetic scattering rate \( \gamma_{imp} \). For the quantification of the non-magnetic counterpart \( \gamma_{imp} \approx 2 \pi T_D \), the Dingle temperatures, \( T_D \), measured by the de Haas-van Alphen (dHvA) effect are very suitable \cite{17,15}. The experimental values \( T_D = 28 K \) and 4K reveal \( \gamma_{imp} = 18 K \) and 25K for our YNi$_2$B$_2$C and LuNi$_2$B$_2$C single crystals, respectively, indicating that the clean limit is reached since \( \gamma_{imp} \leq 2\Delta_0 \approx 51K \) holds for both samples, where \( 2\Delta_0 \) denotes the smaller of the two gaps. Hence, the scattering by impurities can be neglected setting \( \gamma_{imp} = 0 \).

In the weak coupling limit of an ISB case, Eqs. (1-5) are equivalent to the well-known WHH-theory \cite{23}. Any anisotropy of \( H_{c2} \) can be described by a similar, but much more tedious, system of equations \cite{24}. Since the measured anisotropy is relatively weak, it will be ignored for the sake of simplicity. Therefore, only \( H_{c2}(T) \)'s will be compared with those computed for our isotropic models.

The standard ISB model \cite{23} describes quantitatively the renormalization of the physical properties of metals due to electron-phonon (el-phon) interaction. The input parameters of the ISB model are the density of states at \( E_F \), \( N(0) \), the Fermi velocity \( v_F \), the impurity scattering rate \( \gamma_{imp} \), the Coulomb pseudopotential \( \mu^r \), and the spectral function \( \alpha_{ij}^2 F(\omega) \) of the el-phon interaction. These quantities can be determined from a few experimental data: the normal state low-\( T \) electronic specific heat \( \gamma_S T \), the plasma frequency \( \omega_{pl} \) inferred from the optical conductivity, \( H_{c2}(0) \), \( T_c \) and its isotope exponent \( \alpha \), as well as the normal state low-\( T \) dc resistivity \( \rho(0) \approx \rho(T_c) \) which similarly to \( T_D \), gives a direct measure of the sample purity. We adopt for the Coulomb pseudopotential \( \mu^r = 0.1 \) and \( \omega_{pl} = 600 \) meV for the energy cutoff in Eq. (2). The total el-phon coupling constant, \( \lambda = 2 \int dq \alpha_{ij}^2 F(\omega) / \omega \), can be estimated from the boron isotope effect \( \Delta \approx 0.2 \) \cite{21} and the well-resolved phonon spectrum \cite{33}.

We first consider LuNi$_2$B$_2$C. To find a lower bound for \( \lambda \), we accounted for only the high-energy carbon phonons centered at 50 meV and the boron branch at 100 meV. Fitting the experimental \( \alpha_B \) and \( T_c \) values, we obtained the partial coupling constants \( \lambda_{1a0} = 0.31, \lambda_{2a0} = 0.22, \) and \( \lambda = \lambda_{1a0} + \lambda_{2a0} = 0.53 \), where the subscripts denote the corresponding phonon energies in meV. An upper bound of \( \lambda = 0.77 \) has been found using the Lu phonons centered near 9 meV \( (\lambda_0 = 0.34) \) and the same B band \( (\lambda_{100} = 0.43) \) as in the case before. In the following a wide averaged spectrum with \( \lambda = 0.65 \) \( (\lambda_{100} = 0.37, \lambda_{2a0} = 0.5, \lambda_a = 0.16) \) will be used which reproduces the experimental values of \( \alpha_B \) and \( T_c \). \( N(0) = 11.8 \) mJ/mol \( k_B^2 K^2 \) has been estimated from the experimental value \cite{33} of \( \gamma_S = 2 \pi^2 k_B^2 (1 + \lambda)N(0)/3 = 19.5 \) mJ/mol \( K^2 \). The value of \( v_F = 2.76 \) \( 10^7 \) cm/s follows from the experimental value \cite{33} of the plasma frequency \( \omega_{pl} = \sqrt{4 \pi^2 \nu_F^2 N(0)/3} = 4.0 \) eV. The analogous values for YNi$_2$B$_2$C are \( \lambda = 0.637, N(0) = 11.1 \) mJ/mol \( k_B^2 K^2 \), \( v_F = 3 \times 10^7 \) cm/s and \( H_{c2}(0) = 2T \), where the data of Refs. 8 and 34 have been used.

We solved Eqs. (1-5) with these parameter sets for two types of spectral densities \( \alpha_{ij}^2 F(\omega) \): (i) a wide-averaged spectrum and (ii) a single Einstein mode peaked at \( h\omega_{pl} = 42.4 \) meV chosen to yield the experimental \( T_c = 16.5 \) K for LuNi$_2$B$_2$C using the same value of \( \lambda = 0.65 \) as in the first case. The results are shown in Fig. 4. Note that in the intermediate coupling regime under consideration, as expected, \( H_{c2}(T) \) is insensitive to details of the shape of \( \alpha_{ij}^2 F(\omega) \) \cite{27} and, in the clean-limit case, also insen-
sitive to the actual value of the small scattering rates. Comparing the LuNi$_2$B$_2$C data with the ISB curves one clearly realizes strong deviations. In particular, there is a discrepancy of about 3 between experimental and ISB model values of $H_{c2}(0)$. For YNi$_2$B$_2$C the discrepancy reaches even a factor of 5. Therefore it makes no sense to discuss any details of the shape of $H_{c2}(T)$, such as the PC, resulting in deviations of the $H_{c2}(T)$ curves of the order 10 to 20 %, until the reason for the large failure to account for the magnitude of $H_{c2}(0)$ has been elucidated. We remind the reader how this serious difficulty was circumvented in previous studies. At first, frequently the quantity $h_{c2}(T) = -H_{c2}(T)/[T_c(dH_{c2}/dT)_{T=T_c}]$, describing the shape of the $H_{c2}(T)$ curve, has been considered, but the absolute values of $H_{c2}(T)$ have not been discussed at all [7]. At second, since within the ISB-model $H_{c2}(0)$ is a monotonically increasing function of the impurity content, in principle, large $H_{c2}(0)$-values might be obtained. To check this approach we calculated the impurity scattering rate $\gamma_{imp}$ which is required to increase $H_{c2}(0)$ up to the LuNi$_2$B$_2$C value of 7.6 T. Thus we obtain $\gamma_{imp} \approx 427$ K which would lead to $\rho(0) \approx 17 \mu\Omega cm$ which strongly deviates from the experimental value $\rho(0) \approx 2.5 \mu\Omega cm$. In this context we note that our data and those of Ref. 35 show dependencies just opposite to those predicted by the ISB-model: $H_{c2}(0)$ and $T_c$ increase when $\gamma_{imp}$ decreases! At third, a further empirical parameter, the clean limit coherence length, has been introduced in Ref. 35. However, this results in the overdetermination of the model parameter set and the consistency of the two values $v_F$ obtained using (i) a clean-limit coherence length and (ii) normal state data has not been checked. Thus the ISB approach fails to explain simultaneously the three values $\gamma_S=19.5$ mJ/mol K$^2$, $\hbar \omega_{pl}=4$ eV, $H_{c2}(0)=7.6$ T in the clean limit, and the four values $\gamma_S$, $\omega_{pl}$, $H_{c2}(0)$, $\rho=2.5 \mu\Omega cm$ in the dirty limit. In addition, the ISB model is also unable to explain the small gap values $2\Delta_0/k_BT_c < 3.5$ observed in microwave [18], tunneling [10], and dHvA [37] measurements. Furthermore the PC of $H_{c2}(T)$ near $T_c$ and the extended quasi-linear behavior of $H_{c2}(T)$ down to $T \approx 1$ to 2 K cannot be described within the ISB. The ISB model with a single $v_F$ contradicts the dHvA data which clearly show the presence of at least six different sections $F_0 \ldots F_5$ [18,19] with roughly two or three groups of $v_F$'s.

Turning to our TBM we solved Eqs. (1-5). For the sake of simplicity, the same phonon spectrum as in the ISB case discussed above has been adopted. We achieve an excellent agreement with the LuNi$_2$B$_2$C data (see Fig. 1) for $\lambda_1=0.51$, $\lambda_2=0.2$, $\lambda_{12}=0.4$, $\mu_1^1=\mu_2^1=0.1$, $v_{F1}=3.7 \times 10^5$ cm/s, $v_{F2}=0.96 \times 10^5$ cm/s. For YNi$_2$B$_2$C we used the following set: $\lambda_1=0.5$, $\lambda_2=0.2$, $\lambda_{12}=0.4$, $\mu_1^1=\mu_2^1=0.1$, $v_{F2}=3.8 \times 10^5$ cm/s, $v_{F1}=0.89 \times 10^5$ cm/s, $N(0)=11$ mJ/mol k$_B^2$K$^2$. Reproducing $T_c=15.6$ K, the adopted values of $N(0)$ agrees well with the LDA-value $[14]$ of 9.5 mJ/mol k$_B^2$K$^2$. The plasma frequency $\hbar \omega_{pl}=4.4$ eV is in accord with $\hbar \omega_{pl}=4.25$ eV obtained in Ref. 34. From the obtained $v_F$'s it is concluded that $\omega_{pl}$ is mainly related to the second weakly coupled band. Then transport, optical and tunneling data mainly exhibit the properties of that band whereas the strongly coupled band remains almost hidden. The calculated value of the penetration depth at 4.2 K, 100 nm, is in agreement with the data of Ref. 35. Our $\gamma_S=17.2$ mJ/mol K$^2$ should be compared with $\gamma_S=18.2$ mJ/mol K$^2$ reported in Ref. 8. Finally, we arrive at $H_{c2}(0)=9.4$ to 9.9 T in good agreement with our experimental value $H_{c2}(0)=10.6 \pm 0.2$ T. The experimental $H_{c2}(T)$ curve of YNi$_2$B$_2$C together with results of the TBM are shown in Fig. 2. The values $v_F \approx 4.2 \times 10^5$ cm/s and 0.7 to 1.3 $\times 10^5$ cm/s of the extremal orbits $F_2$ and $F_2/2$, respectively, derived from earlier dHvA data [18,19] on the same YNi$_2$B$_2$C single crystal do not deviate much from the parameters $v_{F1} \approx 3.8 \times 10^5$ cm/s and $v_{F1} \approx 0.8$ to 0.96 $\times 10^5$ cm/s introduced empirically in our approach. The remaining deviations might be due to natural differences between $v_F$'s on extremal orbits seen in the dHvA-experiments and the corresponding effective quantities of our TBM which contains implicitly information on the whole Fermi surface.

Further calculations within our TBM reveal that the PC of $H_{c2}(T)$ mainly depends upon the strength of the interband coupling ($\lambda_{12}$, $\lambda_{21}$) and to a lesser extent upon the ratio of the Fermi velocities and the intraband coupling strength ($\lambda_1$, $\lambda_2$). These findings may also be interpreted in terms of the flat Ni-derived band near $E_F$ and the dispersive bands crossing $E_F$ seen in the LDA-band structure [14]. The strong mixing of these bands might be viewed as the microscopic origin for significant interband coupling. Further work is required to clarify this point. For low $T < 4$ K, $H_{c2}(T)$ is very sensitive to the

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**FIG. 2.** Temperature dependence of $H_{c2}(T)$ for YNi$_2$B$_2$C. Experimental points for the magnetic field $\vec{H} \parallel$ c-axis. The Fermi-velocities of the two-band model (TBM) are given in units of $10^5$ cm/s. For the interaction constants see the text.
The actual $v_{F,2}/v_{F,1}$ ratio. This is illustrated by the additional curves shown in Fig. 2. The variation of $v_{F,1}$ and $v_{F,2}$ results in slightly higher and lower $H_{c2}(0)$ values, respectively. Our empirical parameter sets for LuNi$_2$B$_2$C and YNi$_2$B$_2$C differ almost only in their $v_{F,1}$ values which roughly scale with the Ni-Ni distance as $d_{N-Ni}^{-5}$. In this context the study of Sc(Th)Ni$_2$B$_2$C crystals having much reduced (increased) Ni-Ni distances is of interest.

Our analysis reveals that both compounds can be well described within an effective two-band model provided there are at least two groups of electrons having (i) significantly different Fermi-velocities, (ii) strong coupling in the small-$v_F$ band, as well as (iii) sizeable coupling between the small-$v_F$ and the large-$v_F$ band. This case differs from the situation considered in Refs. 24 and 26.

There, the stronger coupling is in the large-$v_F$ band and the curvature of $H_{c2}(T)$ near $T_c$ is negative. A PC would only appear at intermediate $T$ if the interband coupling and the impurity scattering are both weak. In contrast, in this region $H_{c2}(T)$ shows almost no curvature in our model. In other words, the result of Refs. 24 and 26 can be understood as an average over two weakly coupled superconductors, the first with a high $H_{c2}(0)$ but a low $T_c$ and the second one with a small $H_{c2}(0)$ but high $T_c$. In our case the isolated small-$v_F$ subsystem would have high values of $\lambda H_{c2}(0)$ and $T_c$. The values of $H_{c2}(0)$ and $T_c$ of the coupled system are reduced by the second large-$v_F$ subsystem with weak interaction parameters. In this case, which to the best of our knowledge has not been considered so far, the PC of the resulting $H_{c2}(T)$ near $T_c$ becomes a direct manifestation of that interband coupling. In our TBM, the PC of $H_{c2}(T)$, as well as $H_{c2}(0)$, and $T_c$ are suppressed by growing impurity content and the PC vanishes upon reaching the dirty limit with $T_c \approx 11$ K. Thus the observed maximal PC of $H_{c2}(T)$ near $T_c$ of multi-band systems of the type under consideration can be regarded as a direct measure of the high sample quality, as opposed to the widely spread belief attributing it simply to sample inhomogeneities. The latter scenario is excluded by the sharp transition in the $\chi(T)$ data.

To summarize, we have shown that the superconductivity of pure non-magnetic borocarbides should be described within a multi-band picture. The isotropic TBM gives a reasonable starting point toward the understanding of the mechanism of superconductivity in these compounds. Combined studies of quantum oscillations (dHvA) and $H_{c2}(T)$ unified by Eliashberg analysis are found to be valuable supplementary tools to elucidate the specific role of subgroups of electrons having small $v_F$'s and strong coupling to bosons. These electrons may be readily overlooked by other experimental techniques.

We thank O. Dolgov, D. Rainer, E. Maksimov, H. Braun, N. Schopohl, M. Golden, J. Fink, L. Schultz, H. Eschrig, and A. Gadun for discussions. This work was supported by the INTAS-93-2154 grant, the SFB 463, and the Deutsche Forschungsgemeinschaft.

\[ \text{Note that the term “two-band” should not be taken too literally. Within the Eliashberg theory a single anisotropic band model is equivalent to an isotropic multi-band model more suitable for theoretical studies.} \]

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