Magnetic pair breaking in superconducting $HoNi_2B_2C$ studied on a single crystal by thermal conductivity in magnetic fields

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The magnetic field dependence of the thermal conductivity κ of a $HoNi_2B_2C$ single crystal (parallel to [110]; $2<T/\mathrm{K}<10$; $\mu_0H\leq 0.4$ T) is reported. It exhibits a characteristic change in the slope of $\kappa(T)$ at significantly lower temperatures (or lower fields) than the resistively measured critical temperature $T_c(H)$ (or the upper critical field $H_{c2}(T)$), thus pointing to an enhanced fraction of quasiparticles due to strong magnetic pair breaking. This demands a careful interpretation of $\kappa(T,H)$ data for distinct superconductors. Different scenarios for the occurrence of small energy gaps below and above the magnetic-ordering temperatures are discussed including multiband aspects of the superconductivity. In particular, we suggest an improved multiband approach for the commensurate antiferromagnetic phase: whereas a nearly isotropic band unaffected by the rare-earth magnetism shows a BCS-like gap, a second band is strongly affected by the competition between superconductivity and magnetism leading to a second smaller gap. The latter band with relatively large Fermi velocities dominates the thermal conductivity.

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I. INTRODUCTION

Rare-earth nickel borocarbides have provided much insight into superconducting and magnetic phenomena during recent years.[1] One of the most interesting compounds among them is $HoNi_2B_2C$ due to the occurrence of magnetic ordering in the superconducting state connected with a near-reentrant behavior. The competing interplay between magnetism and superconductivity in general has been studied since about 50 years.[2,3] In the early theoretical work this competition has been considered in the framework of the suppression of superconductivity by paramagnetic impurities,[4] including the behavior of the thermal conductivity, κ.[5] There gapless superconductivity occurs which cannot be described by a standard BCS-like gap due to the related induced damping effects. For $HoNi_2B_2C$, some hints for such a state have been derived from early point-contact spectroscopy (PCS) data[6] for a region below the onset of superconductivity. Furthermore, the zero-field behavior of κ has been ascribed to gapless superconductivity, too.[7] However, recent PCS measurements below about 5 K (Ref. 8) yield evidence for an isotropic BCS-like gap but with a value of the critical temperature, $\tilde{T}_c$, smaller than the usually observed $T_c\approx 8.5$ K what might indicate gapless superconductivity in a range just above that $\tilde{T}_c$ but a superconducting state with a small gap cannot be ruled out. Besides the nature of this state, it has to be investigated how the superconductivity is influenced by a rather complex multiband-type electronic structure. The first step in this direction has been undertaken by the theoretical description of the upper critical fields $H_{c2}(T)$ within a simple effective isotropic two-band model for the nonmagnetic borocarbides.[9] In the corresponding measurements strongly coupled slow electrons are probed whereas transport, penetration depth, and tunneling measurements probe mostly fast electrons. Thus, only a combined analysis of rather different measurements allows a deep enough insight into the whole electronic structure of these compounds. The possible role of such multiband effects also in their magnetic counterpart compounds, in particular in $HoNi_2B_2C$, is much less understood. Here, different subgroups of conduction electrons are additionally *differently* affected by various coexisting magnetic phases. In this context it should be mentioned that the multiband picture becomes more extended, if the electrons involved in the nonsuperconducting magnetic and/or gapless subsystems (called simply "bands") are counted separately from those bearing the superconductivity. Furthermore, it has to be considered that in $HoNi_2B_2C$ strong antiferromagnetic fluctuations are present well above the Néel temperature, T_N, as observed, e.g., in specific-heat[10] and thermal-expansion[11] studies. Generally, a rich variety of phenomena might be expected for multiband superconductors. Recent examples are the $\kappa(T)$ dependence of the multiband d–wave superconductor $Ce_{1-x}La_xCoIn_5$ (Refs. 12 and 13) and the gapless superconducting state in a quasi-two-dimensional multiband model system[14] or in high enough magnetic fields.[15]

Measurements of κ are a powerful tool to investigate the scattering mechanisms of different excitations, i.e., of electronic, phononic, and magnetic character. In the superconducting state, additional information can be obtained about the fraction of uncondensed quasiparticles. For zero field, κ has been investigated in several studies on $HoNi_2B_2C$ (Refs. 7 and 16–19); Ref. 18 includes a review for several rare-earth nickel borocarbides. However, an interesting field dependence of the thermal conductivity present at finite but low magnetic fields has been overlooked hitherto. Thus, to the best of our knowledge, only a single measurement in a magnetic field with a single value of 0.5 T on a *polycrystalline* sample in the temperature range from 4.2 to 15 K has been reported for $HoNi_2B_2C$.[17] However, this field is too large for the observation of a weak additional superconducting "band" which we have detected below 0.3 T and analyzed in the present work. Measurements of magnetic field dependences of the thermal conductivity for single crystals of magnetic borocarbides are missing in general, too. A systematic study of thermal conductivity $\kappa(T)$ for a (high-quality) single crystal of $HoNi_2B_2C$ down to 2 K and especially on its field-dependence provides the main motivation for the present work.

II. EXPERIMENTAL DETAILS

Here, we report simultaneous measurements of the thermal conductivity κ and the electrical resistivity, ρ, by standard two-thermometer-one-heater and four-point methods, respectively, using a physical property measurement system (Quantum design). These data have been taken in a continuous mode, i.e., a stepwise heating overlaying the slowly rising temperature in constant magnetic fields. Additional scans have been taken at constant temperature with the field changed in small steps. Our analysis is restricted to the range below 10 K and below the metamagnetic transition at about 0.4 T. A $HoNi_2B_2C$ single crystal with dimensions of about $7\times2\times1$ mm^3 with the long side parallel to [110] has been grown by the floating-zone method[20] and annealed at 1000 °C for 72 h. After finishing a scan, it has been heated up to at least 10 K and then cooled down in zero applied magnetic field. In the vicinity of T_N, κ differs in its values measured starting at 10 and at 80 K, probably due to remaining partial magnetic order above T_N (typical for magnetic borocarbides, see Müller *et al.* in Ref. 1) modified by the magnetic history. In the latter case, the development of such partial order leads to enhanced scattering and to a continuous decrease of κ with increasing H. This phenomenon will be discussed elsewhere together with a more detailed analysis of the magnetic transitions. A hysteresislike behavior of $\rho(T)$ has been observed also for the closely related compound $DyNi_2B_2C$.[21]

III. RESULTS AND DISCUSSION

The measurements of $\rho(T)$ of $HoNi_2B_2C$ which agree with a previous report[22] are shown in the inset of Fig. 1. For clarity, the data for several selected fields, only, are plotted; the ratio of its zero-field values at 300 and 9 K amounts to 13. In zero applied field (and similarly for $\mu_0H=0.05$ T), the superconductivity persists in the whole range below $T_c(0)$ $=8.6$ K. For 0.1 T, reentrant behavior is found near 5 K whereas for $\mu_0H\geq0.2$ T the superconductivity is suppressed above T_N. The detrimental influence of the magnetic scattering on the transport in this region is clearly seen by the large increase in $\rho(T)$ at T_N for 0.4 T.

Figure 1 shows $\kappa(T)$ measurements for three typical magnetic fields. For clarity, further field values are not included in the plot. The zero-field behavior of κ agrees fairly with previous observations[7,16–19] except the very sharp and large maximum near 5 K. One reason for its observation might be the high experimental resolution due to the small heater power required with a typical temperature difference of 20 mK at 4 K. Moreover, it should be noted that the present report is the first one with the field parallel to [110]. Furthermore, the higher crystal quality leads to somewhat higher values of κ than in the mentioned previous studies. The lower values measured on polycrystals are connected with the quite large anisotropy in κ (Ref. 19) due to the contribution of the smaller c-axis conductivity as well as with additional scattering at grain boundaries. Despite a minor hysteresis near T_N, all details of $\kappa(T)$, including the sharp peaks at T_N, can be repeated also for measurements with falling temperature. As previously shown by Cao *et al.*[17] superconductivity and magnetic structure are strongly affected by external fields exceeding 0.5 T. Here we focus on a systematic study in weaker fields below which metamagnetic structures do not occur.

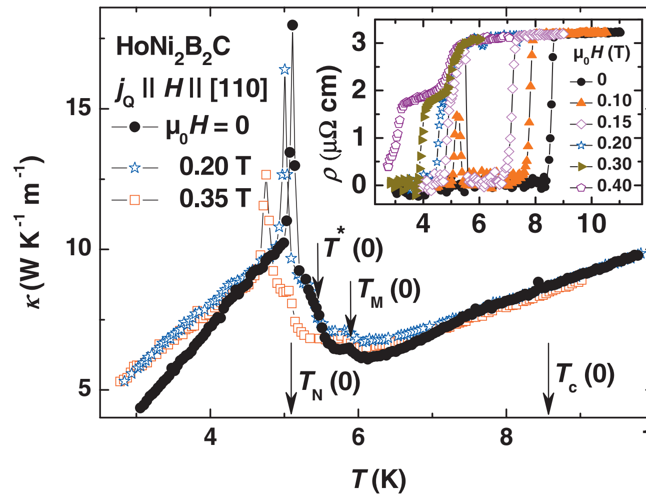

FIG. 1. (Color) Temperature-dependent thermal conductivity κ of $HoNi_2B_2C$ for three different values of the applied field H (parallel to [110] and to the heat current j_Q). The arrows mark the zero-field magnetic phase transitions (see text) and (not visible in $\kappa(T)$) T_c. The inset shows the simultaneously measured resistivity $\rho(T,H)$ for several fixed values of H. In the resistivity measurements shown in the inset, the same directions of the applied field and the electric current as in the thermal-conductivity measurements have been examined.

The increase in $\kappa(T)$ with falling temperature can be ascribed to a reduced scattering on magnetic fluctuations due to the onset of long-range magnetic order enhancing the heat transport by both, electrons and phonons. Possible microscopic reasons[23,24] will be discussed elsewhere. In addition, there is a tetragonal-to-orthorhombic lattice phase-transition[25] (very likely of the first order, see, e.g., Ref. 26) caused by the strong magnetoelastic coupling. Thus, below T_N a significant anisotropy occurs in the previously tetragonal basal plane. Hence, a comparison between different measurements requires information about both, crystallographic directions and crystal quality. The occurrence of three features in $\kappa(T)$ at T_N, T^*, and T_M confirms the known magnetic phase transitions.[23] With increasing fields up to 0.35 T, T_M is only weakly field dependent, whereas T^* and T_N are moderately shifted to lower T in line with previous studies of $\rho(T)$,[22] magnetization,[22] and specific heat.[26]

The superconducting transition, $T_c(H)$, as determined by $\rho(T,H)$ does not have any impact on $\kappa(T)$ in the two ranges above and below T_N. For these ranges, $\kappa(T)$ is shown at enlarged scales in two separate panels in Fig. 2. A change in the slope of $\kappa(T)$ can be observed well below $T_c(H)$ in all fields up to a certain one at a field-dependent temperature $T_d=T_d(H)$, for both ranges. This enhanced drop below a tem-

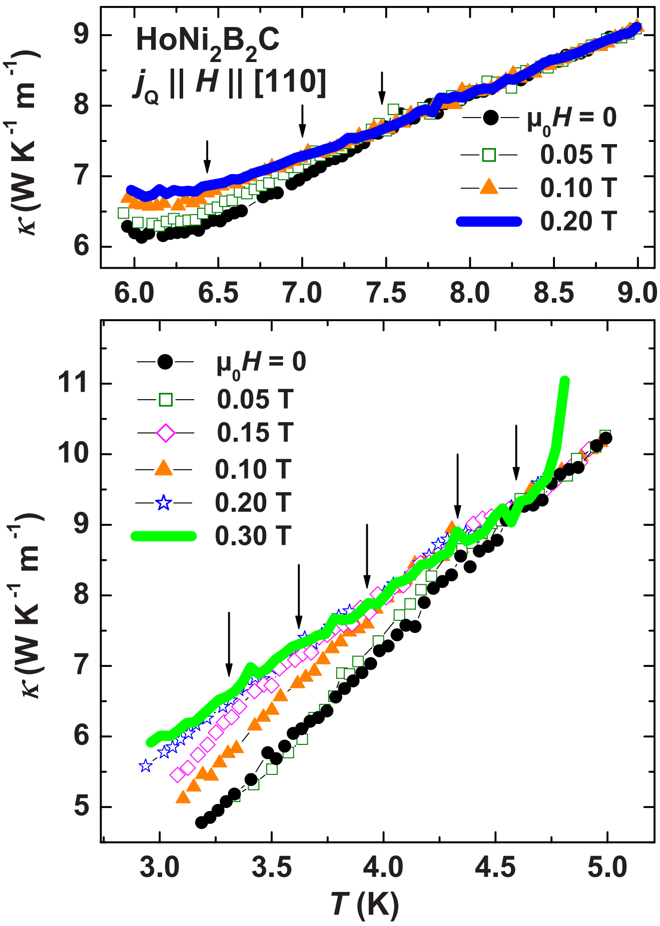

FIG. 2. (Color online) Thermal conductivity of $HoNi_2B_2C$ for selected fields in the two temperature regions above and below the magnetic transitions (upper and lower panel, respectively). The arrows mark $T_d(H)$, the onset of deviations of the low-field data from those in the highest field shown here (0.2 T and 0.3 T in the upper and the lower panel, respectively).

perature T_d is indicated by an arrow for each field curve. Remarkably, in the range above $T_d(0)$ all the data for different fields lie on top each other. Thus, for this choice of H and T, the influence of the applied field on the magnetic contribution to κ (as it might arise from magnetic structures or crystalline-electric-field contributions) is minor. Consequently, the changes at T_d are of electronic origin as will be discussed below.

Figure 3 shows additional scans at constant temperatures. Again, the corresponding field $H_d(T)$ below which $\kappa(H)$ becomes field dependent is significantly lower than the resistively determined upper critical field $H_{c2}(T)$ where the latter is not shown for these measurements.

Similar behavior (there denoted as H_s) has been found for the unconventional multiband superconductor URu_2Si_2 and interpreted as virtual H_{c2} of the smaller gap.[27] No change in the slope of $\kappa(H)$ presented here could be resolved at 8.0 K. In contrast, studies on the nonmagnetic YNi_2B_2C (Ref. 16) and $LuNi_2B_2C$ (Ref. 28) revealed $H_d(T)=H_{c2}(T)$ which had been ascribed to the change in the electronic contribution to $\kappa(H)$. In the latter study, a clear indication for the lower critical field $H_{c1}(T)$ was found with $\kappa(H)$ being constant for

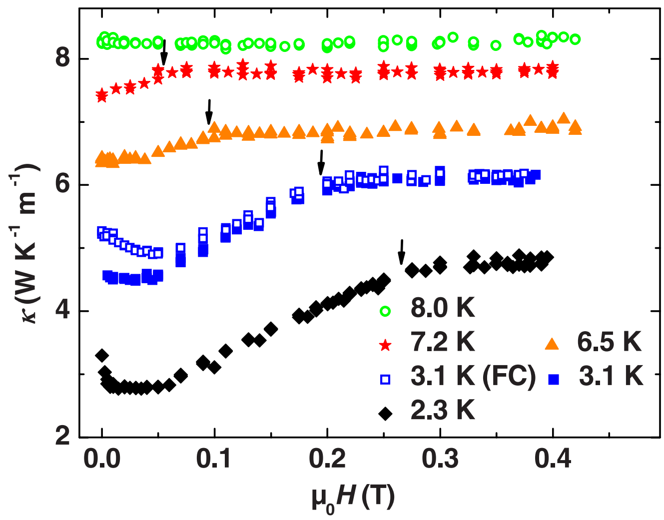

FIG. 3. (Color online) Thermal conductivity of $HoNi_2B_2C$: several selected results at constant temperature, field parallel [110]. The arrows indicate the field $H_d(T)$ at which the slope of $\kappa(H)$ changes significantly; FC means field cooled (otherwise zero-field cooled).

$H<H_{c1}$, and $\kappa(H)$ just above $H_{c1}(T)$ was rapidly decreasing with increasing field due to an enhanced scattering of phonons on flux vortices. For $HoNi_2B_2C$, both these features could not be distinguished unambiguously. As shown in Fig. 3 for 3.1 K, the low-field behavior of κ seems to depend on the magnetic history of the crystal and needs further investigation. These problems do, however, not concern the difference between H_d and H_{c2}.

The characteristic fields (or temperatures) from all scans described above are collected in the H-T diagram of Fig. 4. From the present $\rho(T)$ data alone, it is not possible to distinguish unambiguously between the two incommensurate magnetic transitions whose existence, however, has clearly been

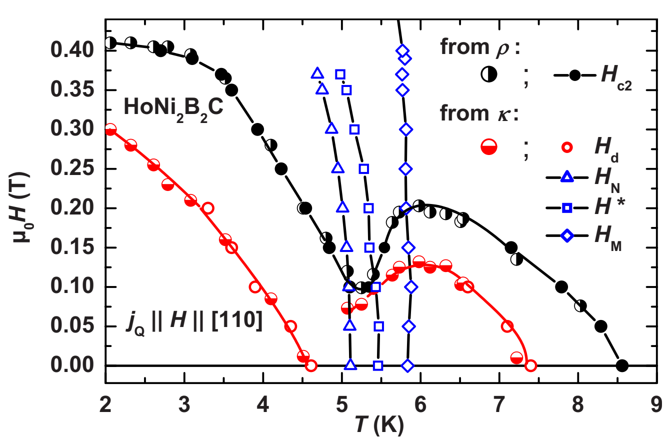

FIG. 4. (Color online) Temperature dependence of characteristic fields of $HoNi_2B_2C$ for heat current and H parallel to [110]. Open and filled symbols: measurement at constant H; half-filled symbols: constant T; for their meaning (see the text). The full lines are guides for the eye; the values of $H_d(T)$ in the vicinity of H_N are not assured. Above 0.4 T, further metamagnetic phases occur (not shown here).

verified by neutron scattering and specific-heat data.[23,26] The temperature dependencies of H_N, H^*, and also that of H_M (corresponding to T_M) agree with resistively determined results[22] where the corresponding characteristic temperatures have been denoted as T_N, T_2, and T_1, respectively. A more detailed discussion of the magnetic transitions will be given elsewhere including the influence of their dependence on the crystallographic direction as observed in Ref. 26.

The $H_{c2}(T)$ dependence is in line with previous reports suggesting a scenario of multiband coexistence of superconductivity and magnetism.[29] Its flattening below 3 K is probably caused by the vicinity of metamagnetic transitions. The temperature dependence of the characteristic field H_d described above shows remarkable similarities with $H_{c2}(T)$ for $3 < T/\mathrm{K} < 5$ where, however, $H_d < H_{c2}$ holds for all temperatures. The lower value of H_d can be considered as a manifestation of a frequently observed magnetic-field-induced coexistence of gapless and paired electrons at $H_d \leq H < H_{c2}$.[15] The experimental resolution of H_d in the range around 5 K is hampered by large changes in $\kappa(H)$ due to the magnetic transitions. Thus, no definite conclusion about a possible vanishing of H_d at H_N or H^* can be drawn. It should be noted that similar kinks in zero-field values of $\kappa(T)$ as denoted by T_d in Fig. 2 can be found in previous data.[18,19] In particular, for an unannealed crystal,[19] the value of $T_c - T_d$ is strongly reduced but also T_c is reduced. The very strongly annealing-dependent properties of the superconducting state of $HoNi_2B_2C$ have been a subject of many investigations[29,30] and point probably to a very weak superconducting state with a special vulnerability against crystallographic disorder. Such a state is typical for the usual coexistence of antiferromagnetism and single-band superconductivity[31] where the resulting electron pairing is modified by electron scattering on the magnetic structure. This subtle balance is destroyed even by nonmagnetic impurities similarly as the standard effective single-band superconductivity in nonmagnetic materials by magnetic impurities.[31]

To gain more insight into the meaning of T_d, the ratio between the electronic contribution, κ_e, in the superconducting (s) state and in the normal (n) state measured at 0.25 T above T_N is plotted in Fig. 5 where the temperature range is restricted to $T > T_M$. The contributions to κ from phonons and magnetic excitations have been separated using the Lorenz number, $L(T) = \kappa(T)\rho(T)/T$, divided by $L_0 = 2.44 \times 10^{-8}\ \mathrm{W}\Omega/\mathrm{K}^2$, set as the unity for the electronic part.

For comparison, we also include experimental results for $SmRh_4B_4$ (Ref. 32) which could be very well fitted by a theoretical description based on the pair-breaking effects of paramagnetic impurities.[5] Numerical results from this approach have been plotted for a certain impurity concentration for which gapless superconductivity was predicted to occur between about $0.7T_c$ and T_c.[5] Noteworthy, the onset of gaplessness does not create any structure in $\kappa_e^{(s)}/\kappa_e^{(n)}$. A small change in its slope for $SmRh_4B_4$ near $0.6T_c$ should rather be ascribed to characteristic changes in the electronic density of states. The strongly enhanced $\kappa_e^{(s)}$ of $HoNi_2B_2C$ and its kinklike structure (Fig. 5) point to an unusually high fraction of unpaired electrons that shows the limits of the paramagnetic-impurity approach adopted to a system with strong antiferro-

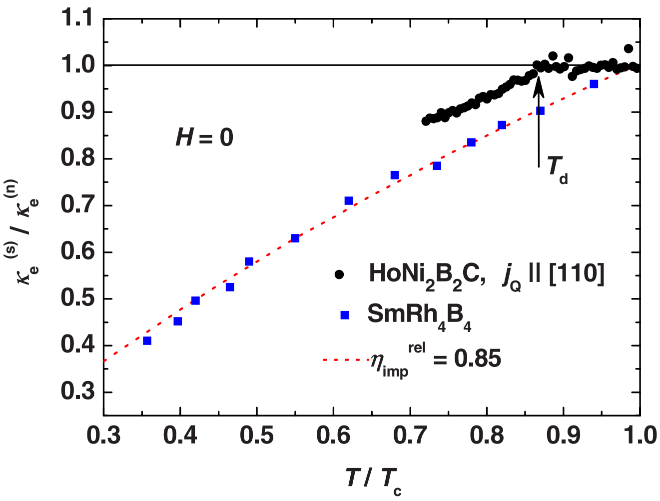

FIG. 5. (Color online) Zero-field values of $\kappa_e^{(s)}/\kappa_e^{(n)}$ of $HoNi_2B_2C$ as a function of the normalized temperature (circles). Additionally, data for $SmRh_4B_4$ (Ref. 32) are included (squares). The dashed line corresponds to a magnetic-impurity concentration of 0.85 (related to a critical one with vanishing T_c) in the paramagnetic-impurity picture.[5] The arrow marks the change in the slope for $HoNi_2B_2C$.

magnetic fluctuations which have been considered for the description of other properties.[31] The magnetic fluctuations might be regarded as more effective pair breakers. A linear fit to the data between T_d and T_c gives an extrapolation to $T = 0$ of $\kappa_e^{(s)}/\kappa_e^{(n)} = 0.97 \pm 0.05$ which (despite the scatter in the data) supports the assumption of a small but not vanishing fraction of condensed electrons in this range. The occurrence of the high fraction of unpaired electrons between T_d and T_N gives a strong indication for a special electronic state which is possibly connected with an additional structural transition at T_d. In principle, the similarities between the data for $HoNi_2B_2C$ and the paramagnetic-impurity picture point to related pairing mechanisms, most probably including a small-gap or gapless state in line with recent PCS results.[8] A full quantitative description of the magnetic pair breaking above T_N needs, however, further analysis. An alternative scenario for the occurrence of seemingly two different H_{c2} values (Fig. 4) might be a full decoupling of the two bands with different superconducting properties due to vanishing interband scattering (see also Ref. 33), in some sense comparable to the spatial separation of two superconducting phases in $PrOs_4Sb_{12}$ samples as proposed in Ref. 34.

At temperatures below the magnetic phase transitions, the observation of a BCS-like gap with a reduced T_c at first glance seems to contradict the occurrence of T_d in this range. It should be noted, however, that in zero applied field the normalized surface resistance shows a minimum at 4 K,[35] near $T_d(0)$. An explanation for the observed kink in the thermal conductivity is offered by a multiband approach as studied in its simplest isotropic two-band version for $LuNi_2B_2C$ and YNi_2B_2C.[9] In $HoNi_2B_2C$, below T_N, the experimental $H_{c2}(T)$ and the PCS results have been well explained in a picture of the multiband coexistence of superconductivity and magnetism. In this picture the superconductivity is dominated by one band with a BCS-Eliashberg-type[36] single gap

$\Delta(0) \approx 1$ meV.[29] However, in addition our $\kappa(T,H)$ data point to a *further* group of electrons which also participates in the superconductivity. It exhibits relatively large Fermi velocities and therefore dominates the heat transport in the normal state. This group seems to be characterized by a strong competition between superconductivity and magnetism which leads probably to a small additional energy gap not yet detected directly. The consideration of the London penetration depth $\lambda_L(0)$ and of the closely related so-called R check[37] might be helpful to elucidate the presence of that second group (band) of electrons. The coupling between the two bands under consideration needs further investigation. A general analysis of the competition between superconductivity and magnetic ordering in the borocarbides has been presented recently;[38] however, the authors ignore such multiband electronic effects being obviously important in these superconductors. Expanding the $\kappa(T,H)$ measurements down to lower temperatures would be very helpful for a deeper insight into the pairing mechanism. In particular, the low-field behavior of $\kappa_e^{(s)}/\kappa_e^{(n)}$ should be investigated. Large values for this ratio have been observed, at low temperatures, in unusually small fields also for other multiband superconductors such as $PrOs_4Sb_{12}$ (Ref. 39) and MgB_2.[40] The latter study further points to some similarities with the behavior of $LuNi_2B_2C$ (Ref. 41) and its large κ at about $0.85H_{c2}$. The present study reveals a similar behavior, however now in the vicinity of T_c and H_{c2}. It should be noted that, in general, the occurrence of $H_d(T)$ might, without additional knowledge about H_{c2}, cause a misleading determination of the latter from $\kappa(H,T)$. The observation of possibly related behaviors of other superconductors in recent studies should be noted in this context. The increasing deviation of T_c determined by ρ from that determined by κ with increasing H observed for URu_2Si_2 (Ref. 42) and there connected with a vortex-lattice melting might illustrate this situation. Noteworthy, in $CeIrIn_5$, the occurrence of a similar feature in $\kappa(T)$ as denoted by H_d in the present work seems to depend on the crystallographic direction.[43] Thus, special care has to be taken in interpreting $\kappa(H,T)$ for a multiband and/or magnetic superconductor.

IV. SUMMARY

The thermal conductivity, κ, of $HoNi_2B_2C$ shows distinct changes at the magnetic phase transitions where the sharp and large maximum at T_N seems to be connected with peculiarities of the magnetic orders and the magnetic excitations. The resistively determined T_c is not visible in $\kappa(T)$ which, however, features a characteristic kink at $T_d < T_c$. Such a kinklike structure has also been observed at constant T in $\kappa(H)$ and points, above T_N, to a mechanism akin to the pair breaking by paramagnetic impurities probably including a small-gap (or even gapless) superconducting state. Although the unusually large fraction of uncondensed electrons restricts the validity of this simple description, it might explain at least qualitatively the missing kink at T_c. A further analysis needs to involve the pair breaking by antiferromagnetic fluctuations. Alternatively, a decoupling of two bands due to a reduced interband scattering might occur. Below T_N, the description of the experimental data requires at least a two-band picture with a dominant group of electrons showing standard BCS-Eliashberg-type behavior with a clear single gap whereas a second group of electrons dominates $\kappa(T,H)$. This additional band is probably characterized by a strong competition between superconductivity and magnetic ordering resulting in a small energy gap not yet detected for instance by point-contact spectroscopy. Consequently, $\kappa(T,H)$ can provide additional information about the properties of multiband and/or magnetic superconductors, thus requiring a careful interpretation of the experimental data. In general, thermal-conductivity measurements are found to be very helpful to achieve, step by step, a more complete picture of the superconductivity in the title compound including also more weakly coupled electrons. This way they may contribute also to achieve a unified description of all magnetic and nonmagnetic borocarbides in future. In this context we strongly encourage analogous studies on single crystals of the other magnetic borocarbide superconductors $DyNi_2B_2C$, $ErNi_2B_2C$, and $TmNi_2B_2C$. Different types of the interplay between superconductivity and magnetism make $HoNi_2B_2C$ one of the most challenging compounds to get more insight into this complex competition and the related various superconducting and magnetic orderings.

Note added. In preparing the present version of our paper we have learned about a similar work for the Fe-based superconductor LaFeOP by Yamashita *et al.*[44] There the multigap structure of this compound has been studied by thermal-conductivity measurements in a low external magnetic field, too. The authors also found a characteristic field denoted there as $H_S \approx 0.035$ T below the upper critical field H_{c2} (somewhat analogously to our H_d reported above). Yamashita *et al.*[44] interpreted their H_S as a "virtual upper critical field" that controls the field dependence of the smaller gap of a "passive" band whose superconductivity is most likely induced by the proximity effect of the "active" bands with a primary gap. The gap ratio of the large and the small gaps is roughly estimated as $\Delta_L/\Delta_S \sim \sqrt{H_{c2}^c/H_S} \sim 6$. With $H_{c2}^c = 1$ T they arrive at $\Delta_S \sim 2$ K. In our case due to the competing magnetism and residual strong-coupling effects for the "active" band,[8] the situation is more complex.

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