

Observation of Electron-Phonon Interaction with Soft Phonons in Superconducting RNi_2B_2C

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The point-contact (PC) spectra of the electron-phonon interaction (EPI) in the superconducting RNi_2B_2C ($R = Ho, Y$) compounds reveal strong EPI with low-energy (4–9 meV) phonons, unlike the case of nonsuperconducting $LaNi_2B_2C$. The PC-EPI spectral functions for all three compounds are determined and lower limits for the EPI interaction parameter λ estimated. Besides, an unusual suppression of the PC-EPI spectral intensity by the external magnetic field was observed for the superconducting compounds, unlike the nonsuperconducting one. [S0031-9007(97)02299-0]

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The recently discovered intermetallic compounds RNi_2B_2C ($R = \text{rare earth, Y}$) show a very rich interplay between superconductivity and magnetism with relatively high superconducting and magnetic critical temperatures [1,2]. Their crystal structure resembles that of the high- T_c superconductors although electronically they are rather three-dimensional metals. The magnetic moments of the Ho ions order ferromagnetically in the basal plane, exhibiting just above the Néel temperature (5 K) an incommensurate spiral structure along the a and c axes [3–5]. The inelastic neutron scattering reveals low-temperature softening of low-energy phonon branches in the nonmagnetic superconducting $LuNi_2B_2C$ [6] at about the same reduced wave vectors $\mathbf{a}^* \approx 0.55$ at which the spin-density wave appears in the magnetic isostructural compounds. Theoretical calculations show that there is an essential nesting of the Fermi surface at about this particular wave vector [7]. The fact that the superconducting transition temperature of the compounds containing magnetic rare earth ions are lower than those of nonmagnetic ones indicates that there is significant exchange coupling between the f electrons and the conduction electrons. All these observations point to the possible strong interaction between magnetic and vibrational degrees of freedom in these compounds, determining self consistently the resulting mechanism of Cooper pairing. Similar mixing of various interactions one may expect for the heavy-fermion and high- T_c superconductors.

Although it was proposed that these compounds fall into the class of common phonon-mediated superconductors there is disagreement as to which phonons play a major role. Mattheiss *et al.* [8] emphasize the role of high-energy ($\hbar\omega = 106$ meV) boron A_{1g} vibrations while Pickett and Singh [9] point on the importance of low-energy phonon branches. The first point of view finds confirmation in the observation of an appreciable boron-isotope effect in YNi_2B_2C [10] while the second goes

in line with the recent findings of strong phonon softening in $LuNi_2B_2C$ [6]. A recent comparative study of the normal-state transport and magnetic properties of nonsuperconducting $LaNi_2B_2C$ and superconducting YNi_2B_2C ($T_c = 15.4$ K) and $HoNi_2B_2C$ ($T_c = 8.7$ K) has revealed new anomalies in their temperature dependences which forced the authors to question the phonon-mediated pairing mechanism in these compounds [11].

In the present point-contact (PC) study, for the first time an attempt is made to determine the electron-phonon-interaction (EPI) Eliashberg functions for all three compounds yielding lower limits for the EPI parameter λ . We have found that the strong interaction of conduction electrons with soft phonons is what distinguishes the EPI in the superconducting materials from that in the nonsuperconducting $LaNi_2B_2C$. The strong suppression of the soft phonons spectral intensity by the external magnetic field, observed for both superconducting materials unlike for the nonsuperconducting one, proves the tight connection between the electron-phonon interaction and magnetism.

The point-contact spectroscopy enables one to determine directly the spectral function of the EPI as well as the interaction of conduction electrons with other elementary excitations in metals [12]. The method consists of measuring the nonlinear current-voltage characteristics of metallic (nontunneling) contacts with a characteristic size smaller than the inelastic electron mean free path $l_{in}(T, eV)$ at a given temperature T and voltage V . The differential resistance $R = dV/dI(V)$ is directly proportional to the energy-dependent scattering rate of the conduction electrons. Unlike the traditional elastic tunneling spectroscopy of superconductors [13] the measurements are carried out in the normal state and are equally effective for metals both with strong and weak EPI. In case of a ballistic regime of electron flow between dissimilar electrodes with strongly different Fermi velocities the second derivative of the I - V characteristic is directly proportional

to the PC EPI spectral function $g_{PC}(\omega) = \alpha_{PC}^2(\omega)F(\omega)$ of the metal with the smaller Fermi velocity v_F [12],

$$\frac{d \ln R}{dV}(V) = \frac{4}{3} \frac{ed}{\hbar v_F} g_{PC}(\omega)|_{\hbar\omega=eV}; \quad (T \approx 0). \quad (1)$$

The function $g_{PC}(\omega)$ is similar to the Eliashberg function with $\alpha_{PC}^2(\omega)$ being the averaged EPI matrix element with kinematic restrictions imposed by the contact geometry and $F(\omega)$ the phonon density of states. The contact diameter d is determined by the resistance at zero bias R_0 in the normal state via the Sharvin expression [12]. In case of copper $d \approx 30/\sqrt{R_0[\Omega]}$ nm, which we shall use for further estimates.

The investigated $\text{HoNi}_2\text{B}_2\text{C}$ and $\text{YNi}_2\text{B}_2\text{C}$ compounds were single crystals grown by a Ni_2B flux-growth technique [14] and the polycrystalline $\text{LaNi}_2\text{B}_2\text{C}$ was made by arc-melting and subsequent annealing [2]. The point contacts are made directly in the cryostat between a Cu or Ag bar and the compounds. For the single crystals we mostly investigated contacts against the ab -plane edges, because this contact orientation gave the best superconducting properties in the point-contact spectra [15]. For each of the compounds we have recorded spectra with either of the counter electrodes with essentially the same results. During one run many contacts at different sites of the crystal surface are investigated. Typical contact resistances are about 1Ω corresponding to the contact size of about 30 nm. This is of the same order of magnitude as the electron mean free path in the starting material. Thus one may expect the spectroscopic regime of current flow [$d \leq \min(l_{in}, \sqrt{l_{in}l_e})$, l_e being elastic scattering mean free path] not to be violated for the best contacts studied. Since the diameter of a contact is much smaller than a crystalline size, also in the case of polycrystalline $\text{LaNi}_2\text{B}_2\text{C}$ we probe the spectra of a single crystal though of unknown orientation. These spectra are reproducible. A magnetic field up to 10 T can be applied either along the ab plane or perpendicular to it.

In Fig. 1 the PC spectra are shown for all three compounds in the normal state (by applying the requisite magnetic field at 4.2 K). The experimental data are almost symmetric with respect to the applied bias voltage. The contacts chosen for Fig. 1 belong to the best according to the following criteria. First, the $R(V)$ curves in the superconducting state reveal the expected Andreev-reflection structure evidencing that the PC spectra refer to the superconducting phase of the material under the contact [15]. Second, the spectra shown correspond to the smallest overall increase of the differential resistance (of about a few tens of %), while the phonon-related structure remains clearly discernible from the monotonically rising background. The PC spectrum of $\text{HoNi}_2\text{B}_2\text{C}$ (Fig. 1) exhibits strong peaks at about 4 and 9 meV corresponding to the maxima of the soft phonon density of states expected from the dispersion curves observed in $\text{LuNi}_2\text{B}_2\text{C}$ by neutron scattering at low temperatures [6]. There is also an essen-

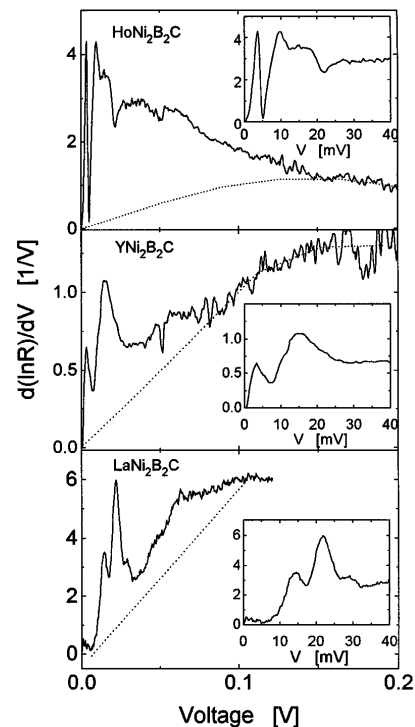


FIG. 1. The second derivatives $d(\ln R)/dV$ of the I - V curves ($R = dV/dI$) for contacts of $R\text{Ni}_2\text{B}_2\text{C}$ ($R = \text{Ho}, \text{Y}$) with Cu and of $\text{LaNi}_2\text{B}_2\text{C}$ with Ag, proportional to the electron-phonon-interaction spectral functions. The dotted lines stand for the supposed background. The contact parameters and measuring conditions are as follow. For $\text{HoNi}_2\text{B}_2\text{C}$: $R = 2.3 \Omega$, ($H \parallel c$) = 0.5 T; for $\text{YNi}_2\text{B}_2\text{C}$: $R = 16.6 \Omega$, ($H \perp c$) = 7 T; for $\text{LaNi}_2\text{B}_2\text{C}$: $R = 0.91 \Omega$, $H = 0$. The insets show the same data for the low bias voltages.

tial spectral intensity between 15 and 19 meV coinciding with a flattening of the phonon dispersion curves. The PC spectrum of Y compound also reveals a soft phonon peak at about the same energy (4 meV). The importance of the low-energy part of the EPI spectral function is strongly emphasized by the fact that the EPI spectrum of $\text{LaNi}_2\text{B}_2\text{C}$ *does not exhibit 4 and 9 meV soft phonon modes*. The observation of these features in the electron-phonon-interaction spectra of the superconducting compounds, unlike the nonsuperconducting $\text{LaNi}_2\text{B}_2\text{C}$, could be connected with differences in the electronic structure between members of the same homological row, such as the nesting of the Fermi surface mentioned above.

Because of the crystal field effects which force the Ho magnetic moments to lay in the (ab) plane [16], a field oriented along the (ab) plane influences the $\text{HoNi}_2\text{B}_2\text{C}$ magnetic structure much more than that parallel to the c axis. We have observed a strong suppression of the low-energy phonon structure in the PC spectra by the field of the order of a few tesla oriented parallel to the (ab) plane [Fig. 2(a)]. At higher fields, the remaining shallow maxima in the $d(\ln R)/dV(V)$ curves are almost field independent and *located at the same voltage positions*.

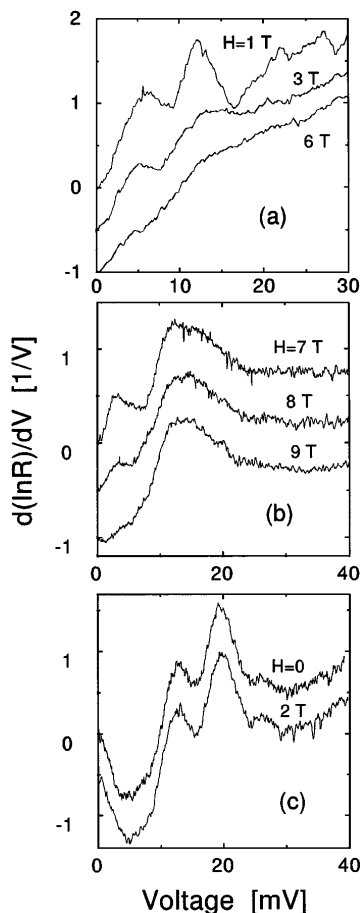


FIG. 2. Magnetic-field dependences of PC spectra with low-energy phonons: (a) $\text{HoNi}_2\text{B}_2\text{C}$ -Ag contact, $R = 0.62 \Omega$; (b) $\text{YNi}_2\text{B}_2\text{C}$ -Cu contact, $R = 16.6 \Omega$; $H \parallel (ab)$. (c) $\text{LaNi}_2\text{B}_2\text{C}$ -Cu contact, $R = 2.08 \Omega$. $T = 4.2 \text{ K}$. Curves shifted vertically by -0.5 with respect to each other.

The persistence of this spectral structure at higher fields proves that the observed structure is not due to the interaction with magnons since at high fields the magnetic order is destroyed. Surprisingly, in nominally nonmagnetic $\text{YNi}_2\text{B}_2\text{C}$ we also observe the field dependence for the intensity of the softest ($\sim 4 \text{ meV}$) phonon peak for fields higher than the upper critical field $H_{c2}(0) \approx 5\text{--}6 \text{ T}$ [Fig. 2(b)]. This may point to the possibility that a magnetic field will destroy the nesting feature that gives rise to the phonon softening. The rather complicated magnetic-field dependence of PC spectra is unusual for the EPI and shows the tight interaction between the vibrational and magnetic degrees of freedom. Taking advantage of the fact that the field oriented parallel to the c axis in $\text{HoNi}_2\text{B}_2\text{C}$ does not disturb the magnetic structure much, we used as small fields $\parallel c$ as only needed to suppress superconductivity in $\text{HoNi}_2\text{B}_2\text{C}$. For $\text{YNi}_2\text{B}_2\text{C}$, we applied the smallest requisite fields perpendicular to the c direction. There is no reason to expect magnetic anisotropy in this material [17], as also supported by our experiments on other crystal directions. These conditions correspond

to the observation of the most intense low-energy phonon structure achievable in our experiments. The PC spectra of $\text{LaNi}_2\text{B}_2\text{C}$ are essentially independent on magnetic field [Fig. 2(c)].

The spectra of all three compounds have a similar background which increases linearly with voltage with a saturating behavior for voltages above about 100 mV as shown by the dotted lines in Fig. 1. The spectroscopic condition [$d \leq \min(l_{\text{in}}, \sqrt{l_{\text{in}} l_e}]$] is probably not fulfilled at about 100 meV leading to a not well resolved high-energy boron vibrational mode in our spectra. Some of the spectra taken on $\text{HoNi}_2\text{B}_2\text{C}$ with the contact axis oriented parallel to the c direction show a wide maximum around 100 meV corresponding to the thermal regime [18], without any traces of superconductivity and low-energy EPI peaks. However, for the $\text{HoNi}_2\text{B}_2\text{C}$ spectrum in Fig. 1, any structure around 100 meV is absent with the presence of superconductivity in the contact area as revealed by the Andreev-reflection signal in the spectra. From these observations the high-frequency boron mode seems to be not vitally important for superconductivity.

The linear background signal at small energies cannot be described by the additional electron scattering on the nonequilibrium distribution of generated phonons leading to the usually observed energy integral of the phonon spectrum for the background. The low-energy background could be a signature of the importance of electron-electron scattering processes due to the exchange interaction in these materials.

Using Eq. (1) and subtracting the background we obtain the PC EPI spectral functions $g_{\text{PC}}(\omega)$ shown in Fig. 3 [19]. The integration of the curves in Fig. 3 gives PC EPI parameters $\lambda_{\text{PC}} = 2 \int g_{\text{PC}}(\omega) d\omega/\omega$. Previous experience with conventional superconductors has shown that in the case of large-Fermi-surface metals $\lambda_{\text{PC}} \approx \lambda$, where λ is the electron-phonon interaction parameter used

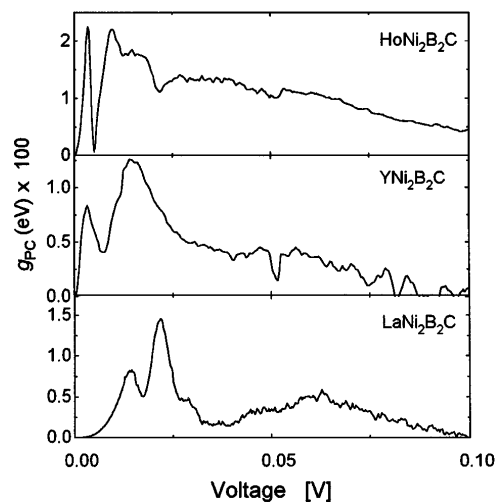


FIG. 3. Point-contact spectral functions g_{PC} of the electron-phonon interaction in $\text{HoNi}_2\text{B}_2\text{C}$, $\text{YNi}_2\text{B}_2\text{C}$, and $\text{LaNi}_2\text{B}_2\text{C}$.

in the strong-coupling theory of superconductivity. We cannot estimate the absolute value of λ_{PC} since the regime of current flow is not known. Supposing that to be the same for typical contacts we find the following inequality row based on many PC spectra investigated

$$\lambda_{LaNi_2B_2C} < \lambda_{YNi_2B_2C} < \lambda_{HoNi_2B_2C}. \quad (2)$$

The λ parameter for $LaNi_2B_2C$ is the smallest as expected, but that for $HoNi_2B_2C$ appears to be larger than for YNi_2B_2C though $T_{c,HoNi_2B_2C} < T_{c,YNi_2B_2C}$. This may be due to our underestimation of the EPI in YNi_2B_2C since we are forced to apply relatively large fields to suppress superconductivity in this compound, decreasing the intensity of the soft phonon mode, too. On the other hand, the enhancement of the soft phonon peaks in the PC EPI spectra by magnetism in $HoNi_2B_2C$ may be the cause of the enhanced λ in this material despite its T_c is lowered by the magnetic scattering. Further study of the field dependence of EPI spectra in the superconducting materials should clarify this point.

In conclusion, we have shown the effectiveness of point-contact spectroscopy in the study of the electron-phonon interaction spectral function both in superconducting and normal, magnetic and nonmagnetic, rare-earth nickel borocarbides. To our knowledge up to now this is the *only* experimental technique able to yield energy-resolved information on the electron-phonon interaction in these compounds. The strong EPI with soft phonon modes is clearly demonstrated to be important for the superconducting compounds and the first determination of EPI spectral functions with the estimates of the λ parameters are made. The magnetic-field dependence of the point-contact spectra points to an enhancement of the EPI by magnetism.

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- [1] R. Nagarajan *et al.*, Phys. Rev. Lett. **72**, 274 (1994).
 - [2] R. J. Cava *et al.*, Nature (London) **367**, 146 (1994).
 - [3] T. E. Grigereit *et al.*, Phys. Rev. Lett. **73**, 2756 (1994).
 - [4] A. I. Goldman *et al.*, Phys. Rev. B **50**, R9668 (1994).
 - [5] J. W. Lynn *et al.*, Phys. Rev. B **53**, 802 (1996).
 - [6] P. Dervenagas *et al.*, Phys. Rev. B **52**, R9839 (1995).
 - [7] J. Y. Rhee *et al.*, Phys. Rev. B **51**, 15 585 (1995).
 - [8] L. F. Mattheiss *et al.*, Solid State Commun. **91**, 587 (1994).
 - [9] W. E. Pickett and D. J. Singh, Phys. Rev. Lett. **72**, 3702 (1994).
 - [10] D. D. Lawrie and J. P. Frank, Physica (Amsterdam) **245C**, 159 (1995).
 - [11] I. R. Fisher *et al.*, Phys. Rev. B **52**, 15 086 (1995).
 - [12] A. V. Khotkevich and I. K. Yanson, *Atlas of Point Contact Spectra of Electron-Phonon Interactions in Metals* (Kluwer Academic, New York, 1995).
 - [13] W. L. McMillan and J. M. Rowell, in *Superconductivity*, edited by R. D. Parks (Dekker, New York, 1969), Vol. 1, p. 561.
 - [14] M. Xu *et al.*, Physica (Amsterdam) **227C**, 321 (1994).
 - [15] L. F. Rybaltchenko *et al.*, Physica (Amsterdam) **218B**, 189 (1996).
 - [16] B. K. Cho *et al.*, Phys. Rev. B **53**, 2217 (1996).
 - [17] E. Johnston-Halperin *et al.*, Phys. Rev. B **51**, 12 852 (1995).
 - [18] I. K. Yanson and O. I. Shklyarevskii, Sov. J. Low Temp. Phys. **12**, 509 (1986).
 - [19] The calibration of the vertical axis in Fig. 3 should be taken with caution and yields only a lower limit for the spectral intensity since we suppose the ballistic regime of current flow through the contacts. With $v_F = 2 * 10^7$ cm/sec we obtain $\lambda_{PC} = 0.03, 0.05,$ and 0.1 for La, Y, and Ho compounds, respectively. For the diffusion regime ($l_e \ll d$) these values may be substantially larger, since the contact diameter in Eq. (1) should be replaced by l_e .