ANDREEV-REFLECTION AND POINT-CONTACT SPECTROSCOPY OF SUPERCONDUCTING RARE EARTH TRANSITION METAL BOROCARBIDES.

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1. Introduction

The recently discovered [1], superconducting compounds $\text{RNi}_2\text{B}_2\text{C}$, where $R$ is rare earths (Lu, Tm, Er, Ho and Dy) and Y, still remain a hot topic of intensive research. Their crystallographic structure resembles that of high-$T_c$ materials, albeit being 3D in electronic properties. The mechanism of Cooper pairing in these compounds is not known in detail although they are commonly believed to be mediated by the ordinary electron-phonon interaction, like A15. The boron isotope effect [2, 3] clearly points that the phonons are involved in superconductivity. Whether these compounds are $s$− or $d$−wave superconductors [4] is still under debate. The important subclass in these materials is presented by the magnetic compounds ($R=$Tm, Er, Ho, and Dy), with the antiferromagnetic (AFM) order [5] and weak ferromagnetism (Er) [6] coexisting with superconductivity.

The measurements of the quasiparticle density of states (DOS) by tunneling spectroscopy encounters experimental difficulties, especially these refer to the magnetic superconductors. To our knowledge, no information is available about the electron-quasiparticle-interaction (EQI) spectral function measured by tunneling spectroscopy in the superconducting state, which unambiguously allows one to determine the mechanism of Cooper pairing.

An investigation of point contacts between a normal metal and a superconductor can partially solve these problems. The superconducting energy gap and DOS can be measured by Andreev-reflection spectroscopy [7], and EQI spectral function can be obtained by inelastic point-contact
spectroscopy (PCS) both in the normal [9] and in the superconducting [10] states. Comparing the tunneling junction with point contacts, we can see that the boundary between $\text{RNi}_2\text{B}_2\text{C}$-superconductor and vacuum (dielectric) in tunnel devices constitutes much stronger discontinuity than the interface between $\text{RNi}_2\text{B}_2\text{C}$ and a normal metal in point contacts, since in the latter case there is a Fermi-sea of electrons at both sides around the interface with no additional dielectric layer introduced. This discontinuity may severely influence the measurements making them different from the bulk properties.

The schematic experimental setup is shown in Fig.1. A small noble metal (Ag, Cu) rod with sharp edges touches the $(a-b)$-plane edge of a flat $\text{RNi}_2\text{B}_2\text{C}$ single crystal or a sharp edge of a polycrystal with an a priori unknown orientation. Many spots can be tried both on the rod and along the $\text{RNi}_2\text{B}_2\text{C}$ edge for investigating the point contacts. The temperature and magnetic field can be varied, the latter, in case of a single crystal, is oriented either $\parallel$ or $\perp$ to the $a-b$ plane. The $I-V$ characteristics and their first, $R(V) = dV/dI(V)$, and second, $dR/dV(V)$, derivatives are recorded by means of a standard lock-in technique. The latter equals $dR/dV(V) = 2\sqrt{2}V_2/V_1^2$, where $V_2$ is the second harmonic and $V_1$ is the modulation rms voltage. The diameter of a point contact can be estimated through the Sharvin formula for a clean orifice $d = 27.5/\sqrt{R_0}$ where $d$ is in nm, the nominator is the average of the noble metal and $\text{RNi}_2\text{B}_2\text{C}$ [11], and $R_0$ is the zero-bias normal-state resistance of a contact in $\Omega$.

2. Superconducting energy gap measurements by Andreev-reflection spectroscopy.

2.0.1. BCS-like DOS in paramagnetic and antiferromagnetic state of $\text{RNi}_2\text{B}_2\text{C}$ ($R=\text{Dy}, \text{Tm}$)

Recently the temperature dependence of quasiparticle DOS has been measured in the DyNi$_2$B$_2$C-Ag point contacts [11] by means of Andreev-reflection spectroscopy. The $R(V)$-curves normalized by the normal-state $R(V)$ dependences are fitted very well with the Blonder-Tinkham-Klapwijk (BTK) theory [7] with a zero smearing parameter $\Gamma$ [8]. This evidences that the quasiparticle DOS is BCS-like in this material. The superconducting energy gap extrapolated to zero temperature equals $(3.63\pm0.05) \ k_B T_c$ which evidences that on the average DyNi$_2$B$_2$C is a moderately strong coupling superconductor. The only disagreement with the BTK theory is that the decrease of the differential resistance around zero bias in the superconducting state is typically a few percentage, instead of the order of one-half of a normal resistance [7]. This can be due either to microscopic inhomogeneity of the material under the contact where only a small part re-
veals the unperturbed superconducting properties, or to the intrinsically decreased Andreev reflection from an AFM superconductor with ferromagnetically ordered DyC planes. It should be stressed that DyNi$_2$B$_2$C is the only $R$Ni$_2$B$_2$C compound which has $T_c < T_N$. Hence, one might think that the molecular field due to the AFM order averages to zero on the scale of superconducting coherence length except of very narrow directions almost parallel to the $a-b$ planes.

The situation is different in TmNi$_2$B$_2$C where incommensurate spin wave ordering occurs at temperatures much below the superconducting transition ($T_c \gg T_m$) and the superconductivity develops in the paramagnetic state. Preliminary results [12] show that the overall behavior of $R(V)$-curves also fits the BTK predictions. That means that this compound also has the BCS form of quasiparticle DOS.

2.0.2. Fine structure in temperature dependence of superconducting energy gap in ErNi$_2$B$_2$C near $T_m$.

The overall temperature dependence of the Andreev-reflection spectra in ErNi$_2$B$_2$C (Fig.2) also follows the BTK model. In this material $T_c > T_m$ and around $T_m = 6.5$ K spin fluctuations depress the superconducting order parameter which manifested as the lowering of $H_{c2}$ near $T_m$ [13]. Recently, this depression has been measured using the temperature dependence of the superconducting penetration depth $\lambda(T)$ [14] which together with superconducting coherence length $\xi(T)$ permits determination of the temperature dependence of the thermodynamic critical field $H_c = \phi_0/[2\sqrt{2}\pi\xi(T)\lambda(T)]$ proportional to the superconducting order parameter (energy gap). Direct measurement of the superconducting energy gap by means of the Andreev reflection and corresponding BTK fitting (Fig.2) is shown in Fig.3. There is a noticeable dip in $\Delta(T)$ around $T_m$, but the overall dependence follows the BCS law shown with the solid line. For this particular contact measured on polycrystalline sample with unknown orientation, the extrapolated gap is 1.82 mV ($2\Delta_0 = 4.23$ $k_BT_c$), which is larger than the average value $\Delta_0/k_BT_c = 3.66 \pm 0.4$ [15], probably because of underestimated $T_c$.

Interestingly, the $\Gamma$-parameter drops around $T_m$ while it takes relatively large value at other temperatures. The barrier parameter $Z$ experiences no change around $T_m$. We emphasize that except the small anomaly around $T_m$, the quasiparticle density of states in superconducting state can be approximated by the BCS dependence both in paramagnetic and AFM states as follows from the BTK fits (Fig.2).

2.0.3. Two superconducting states of HoB$_2$Ni$_2$C.

In the Ho-compound, the superconducting transition temperature ($T_c = 8.7$ K) is close to that of the magnetic transition temperature ($T_m \sim 8.5$ K) into
the incommensurate helical state with wave vector $q^* = 0.91 c^*$. Moreover, the incommensurate $a$-axis modulation with $q^*_a = 0.55 a^*$ also exists over the same temperature range along with the spiral-AFM transition. Both of these states collapse near the commensurate AFM transition with $q^* = c^*$ and $T_N \approx 5$ K [5]. The Andreev-reflection spectra of HoNi$_2$B$_2$C vividly mirror these transformations (Fig.4) [16]. Below $T_c$ and down to $T_N$ the $R(V)$-spectra cannot be fitted by the BTK model. The shallow dip which appears around zero bias has an order of magnitude larger width than that expected for the given $T_c$. We emphasize that for $T_c \geq T \geq 6.5$ K the Andreev spectra are completely different from those in the paramagnetic state of Dy, Tm and Er compounds. Hence, in HoNi$_2$B$_2$C we observed the magnetic transition at the same temperature as the superconducting one, but not at $T_m = 6.5$ K as in the single crystals [17]. It is probable that the uncompensated internal magnetic field along the $a-b$ plane for the spiral structure leads to the gapless behavior of DOS in the temperature region discussed. Below the AFM transition ($T < T_N = 6.6$ K), the spectra reveal the ordinary BTK behavior. Interestingly, the temperature dependence of the spectra follows the BCS law albeit with a new $T_c^* \approx 6.6$ K. The BTK fits yield a zero-temperature energy gap $\Delta_0 = 1.04 \pm 0.06$ meV which gives $2\Delta_0/k_B T_c^* = 3.7 \pm 0.2$ for lower "$T_c^*", i.e. the same as for other RNi$_2$B$_2$C [18]. Using the upper $T_c = 8.5$ K, one obtains unreasonably low $2\Delta_0/k_B T_c = 2.8$.

3. Point-contact spectroscopy of electron-quasiparticle-interaction spectral function.

3.0.4. Principles of PCS.

The point-contact spectroscopy (PCS) involves studies of the nonlinearities of the $I-V$ characteristics of metallic constrictions in the normal state, with the size $d$ smaller than the inelastic electron mean free path (m.f.p.) [19, 20]. In contrast to a tunneling junction, an ideal point contact has no barrier. PCS has the advantage that the material is probed into the depth of the current spreading region, which is of the order of the constriction size. This size should not be too large in order not to violate the conditions of the spectroscopic regime of the current flow: $d \leq \min(l_e, \sqrt{l_{in} l_e})$, where $l_e$ and $l_{in}$ are the elastic and inelastic electron mean free paths, respectively.

In the ballistic regime ($d \leq l_e, l_{in}$), the second derivative of the $I-V$ characteristic, $dR/dV(V)$, is proportional to the electron-quasiparticle-interaction (EQI) spectral function analogous to the Eliashberg function for the electron-phonon-interaction (EPI):

$$\frac{d \ln R}{dV}(V) = \frac{4}{3} \frac{e d}{\hbar v_F} g_{PC}(\omega) \big|_{\hbar \omega = eV}; \quad (T \approx 0)$$
Here $g_{PC}(\omega)$ is the EQI or EPI spectral function with kinematic restrictions imposed by the contact geometry. The factor 4 (instead of 8 as in homocontacts) is due to weak EPI coupling and much greater $v_F$ for the noble metal, as compared to $R\text{Ni}_2\text{B}_2\text{C}$. Correspondingly, the noble-metal EPI function is not seen in the spectrum. For the same reason, in the two-band model of $R\text{Ni}_2\text{B}_2\text{C}$ [21], only the EQI spectral function for the band with the lowest Fermi velocity and largest EQI is seen in the spectrum.

In the spectroscopic regime no heating of the contact area occurs since the energy dissipation length, $\Lambda_\varepsilon$, is much larger than the contact size. However, if the contact size is large compared to $\Lambda_\varepsilon$, then there is local heating and the temperature rises up to $T_0 \simeq eV/3.63k_B$. The thermal feature in the PC spectra can be quite large if at a particular temperature $T_c$, a sharp increase in resistivity occurs like, for example, for the superconducting-normal or AFM-paramagnetic transitions. The voltage position of such transition on the $I - V$ curve should depend on the input power $V/R^2$, which means on the contact resistance.

3.0.5. Comparison between phonon DOS and EQI spectral function for superconducting $\text{YNi}_2\text{B}_2\text{C}$ and non-superconducting $\text{LaNi}_2\text{B}_2\text{C}$.

It is very instructive to compare the phonon density of states, measured by inelastic neutron scattering, and the EPI (EQI) spectral function, obtained by PCS, for superconducting and non-superconducting compounds (Fig.5). For these we choose the nonmagnetic superconducting compound $\text{YNi}_2\text{B}_2\text{C}$ and non-superconducting homologue $\text{LaNi}_2\text{B}_2\text{C}$ [22, 23]. The phonon spectra measured by neutrons contain three groups of peaks: low-frequency modes (0–30 meV), middle-frequency group (40–60 meV) and boron high-frequency modes (100–160 meV) [22]. For point-contact spectra, the high-frequency modes cannot be seen as separate peaks because of non-spectroscopic (thermal) regime of current flow where the m.f.p. becomes shorter than the contact size. For the same reason, the middle-size peaks are smeared, approaching the thermal regime. On the contrary, the low-frequency peaks are well resolved and can be compared with the phonon spectra. It should be noted that the neutron spectra are taken at room temperature while the PC spectra at liquid helium temperature. For non-superconducting $\text{LaNi}_2\text{B}_2\text{C}$ the positions of the low-frequency peaks are very close in both spectra (a) and (b) whereas for $\text{YNi}_2\text{B}_2\text{C}$ the whole low-frequency group in PC spectrum (d) is shifted appreciably below the neutron one (c). This substantial softening of the low-frequency peaks with decreasing temperature was thoroughly studied in Ref.[24] by neutron scattering. A special problem arises with the low-frequency peak at $\approx 4$ meV, which is not resolved yet. In neutron scattering, the emergence of this peak coincides with the transition to the superconducting state and it is observed
within a narrow solid angle of phonon wave vectors. In PCS it is seen both in the superconducting and sometimes in the normal state whereas in the latter case its intensity is greatly diminished by the magnetic field. Here we remind that in order to measure the PCS spectra the requisite magnetic field should be applied to suppress superconductivity. One should also take into account that the lowest-frequency peak is clearly seen in the EPI spectrum which is the phonon DOS weighted by the averaged EPI matrix element within a relatively large ($\sim 45^\circ$) solid angle.

Summarizing the PCS study of EQI function, one can argue that the low frequency modes are characteristic of the superconducting compounds while the behaviour of high energy part of the spectra does not differ appreciably for superconducting and non-superconducting compounds [25, 23, 18].

3.0.6. Low-frequency phonon and crystal-electric-field excitation peaks in $\text{ErNi}_2\text{B}_2\text{C}$

In the $\text{RNi}_2\text{B}_2\text{C}$ compound where magnetic and superconducting orders coexist (i.e. $R$-ion is magnetic), the new branches of excitations appear which can interact with an electron. These are the magnons at temperatures below the characteristic magnetic transition temperatures ($T_m, T_N$) and crystal-electric-field (CEF) excitations. Neutron scattering experiments and magnetization measurements show that the characteristic energies in the latter case are absent in the range 2-10 meV in Ho-, and possibly, Dy-compounds [26, 27]. In Fig.6 the PC EQI spectra are shown for two different ErNi$_2$B$_2$C-Ag point contacts. Along with the second harmonic curves 1,2, $V_2(V)$, taken in the magnetic field needed to destroy superconductivity, the Fig.6 shows (insets) the Andreev-reflection spectra $R(V)$ for the same junctions. These curves serve as a "passport" evidencing that there is an intact material under the contact. Both PC spectra have a large low-frequency peak at about 8-9 meV confirming that this low-frequency phonon feature is necessary for observation of the superconducting state. There is an additional structure at about 6 meV which most probably corresponds to the CEF-excitation determined in ErNi$_2$B$_2$C by neutrons [28, 29]. There is also a non-identified structure (maximum of $R(V)$) at zero bias which does not belong to superconductivity.

3.0.7. Low-energy peaks in $\text{HoNi}_2\text{B}_2\text{C}$ and $\text{DyNi}_2\text{B}_2\text{C}$ compounds.

The low-energy parts of the EQI spectra of HoNi$_2$B$_2$C and DyNi$_2$B$_2$C are shown in Fig.7. These compounds possess the same ground state (commensurate AFM order with magnetic moments aligned ferromagnetically along the [110] direction on the $RC$ planes [5]) though with different $T_N$. Their spectra are very similar in overall shape while the positions of peaks are different. The characteristic energies are hardly due only to the differences
between the phonon branches since the masses of the constituent atoms are nearly the same. Quite definitely, the magnetic excitations should be involved. This follows from the strong temperature and magnetic field dependences of the intensity and energy position in HoNi$_2$B$_2$C [25, 23]. It is difficult to interpret these dependences, since the magnetic phase diagram in HoNi$_2$B$_2$C is very complicated. Fortunately, it is simpler in DyNi$_2$B$_2$C. The unexpected temperature dependence of the lowest-peak for the latter compound is shown in Fig.8 [30]. At lowering the temperature, the lowest-frequency peak appears at about 15 K, much above the superconducting order $T_c = 6$ K and even the magnetic order $T_N = 10.6$ K. Its height and area grow linearly with $T$ (see the lower inset), which means that its width remains approximately constant. It can also be seen in the superconducting state as a strong feature superimposed on the rapidly changing background due to the superconducting DOS. The other phonon lines ($\epsilon V = 10 - 30$ meV in Fig.8) are smeared due to the large background in the PC spectra without any noticeable overall change of intensity with temperature. We are inclined to relate the lowest-lying peak with magnetic excitations (like the same peak at $\epsilon V = 3.5$ meV, Fig.7, in HoNi$_2$B$_2$C). It should not be the true magnon density of state peak, since it is observed above $T_N$ in DyNi$_2$B$_2$C, neither it is due to the CEF excitation, since no peak at this energy is expected. Its origin still remains a puzzle. Perhaps it may be attributed to the spin fluctuations which, judging from the neutron measurement, have a large tail at higher temperatures [5]. In HoNi$_2$B$_2$C, the lowest-frequency peak mentioned above exists starting at $T = 12$ K, which is also above the magnetic transition temperature $T_m = 8.5$ K in this material.

4. Discussion

In literature [31, 32] one can encounter a statement that at least the non-magnetic compounds $R$Ni$_2$B$_2$C (Y, Lu) are similar to A15 superconductors in that they have enhanced electron density of state at the Fermi energy and their Cooper-pairing mechanism involves phonons [2, 3]. Still, despite several attempts, no phonon structure was observed above the energy gap in the tunneling characteristics [35, 36, 37, 38]. In the tunneling characteristics of A15, a distinct phonon structure is observed with a proper value of the electron-phonon parameter $\lambda$ [40]. For magnetic $R$Ni$_2$B$_2$C, the situation is even worse, since the superconducting energy gap is not safely measured [35], to say nothing about the above the gap spectroscopic structure. This situation is disturbing and similar to that in high-$T_c$ superconductors where no structure is seen above the gap, yet [33, 34].

Contrary to tunneling, the PC spectra show strong electron-quasiparticles interactions both with phonons and magnetic excitations. In principle, they
can be studied both in normal and superconducting [10] states. Interpretation of these spectra is not straightforward, since several electron-quasiparticles interactions can interfere with each other. The magnetic structure can hinder the electron-phonon interaction responsible for Cooper pairing [41, 42]. On the other hand, the superconducting transition may lead to a change in magnetic order [43] since it affects the RKKY interaction. Even in the non-magnetic compounds, the proximity of the superconducting energy gap to nonadiabatic softening of phonon modes may lead to drastic changes in the phonon spectrum and electron-phonon interaction. Only the systematic study of PC spectra, both in non-magnetic and magnetic compounds in different magnetic fields and at various temperatures, can shed light on the peculiarities of Cooper pairing in these compounds. In this respect, the use of high quality single crystals is invaluable, since the properties of these compounds are often very anisotropic.

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Figure 1. Schematic presentation of a sample holder.

Figure 2. Differential resistance of polycrystalline ErNi$_2$B$_2$C-Ag point contact at different temperatures: $T = 4.2$, 4.6, 5.0, 5.6, 6.2, 6.8, 7.4, 8.2, and 9.0 K from bottom to top. The experimental curves (dots) are shifted vertically for clarity and normalized for each curve separately. The BTK fits are shown with thin straight lines. $H = 0$. Resistance in the normal state: $R_N = 16.5$ Ω.

Figure 3. (a) Temperature dependence of the superconducting energy gap in ErNi$_2$B$_2$C (solid square dots) determined by BTK fitting shown in Fig.2. The dashed vertical straight line points to the magnetic ordering temperature $T_m$. (b) and (c) are smearing parameter $\Gamma$ and barrier strength $Z$ for the same point contact, respectively. Note the dip of the gap at $T_m$.

Figure 4. Temperature dependence of Andreev-reflection spectra for HoNi$_2$B$_2$C. The curves represented by dots are shifted vertically for clarity after subtraction of a polynomial fit of the 8.6 K data [16]. Near each curve the temperature is indicated. $T_c$ and $T^*_c$ stand for superconducting transition temperature and the "BCS-like" transition temperature, respectively. BTK fits are shown as thin solid lines. $R_N = 2.65$ Ω. The Y-scale is shown by a vertical bar of 0.1 Ω. Note that the BCS-like Andreev-reflection spectra appear only below $T^*_c$.

Figure 5. Comparison of phonon densities of states measured by incoherent scattering of neutrons at room temperatures [22] (curves (a) and (c)) and electron-quasiparticle spectral functions obtained by point-contact spectroscopy at liquid helium temperatures [23] for non-superconducting LaNi$_2$B$_2$C and superconducting YNi$_2$B$_2$C, respectively. Note the substantial softening of low-frequency phonons in YNi$_2$B$_2$C due to lowering temperature[24].

Figure 6. The second harmonic signal for two different ErNi$_2$B$_2$C-Ag point contacts proportional to the EQI spectral function. Resistance of junctions 1 and 2 are 8.25 and 14.4 Ω, modulation voltages 0.975 and 1.94 mV, and magnetic field 1.95 and 2.6 T, respectively. Correspondingly, for each junction their Andreev-reflection spectra are presented in the inserts. Note the low-frequency peaks at energies below 10 meV and the feature at $eV \approx 6$ meV which is tentatively ascribed to crystal-field excitation.

Figure 7. Comparison between the PC spectra of HoNi$_2$B$_2$C and DyNi$_2$B$_2$C. The parameters are $R_N=2.3$ and 27 Ω, $T = 4.2$ and 1.8 K, and $H = 0.5$ and 0.65 T, for HoNi$_2$B$_2$C and DyNi$_2$B$_2$C, respectively.

Figure 8. The temperature dependence of the EQI PC spectra of DyNi$_2$B$_2$C with $R_N = 4.8$ Ω, $V_1 = 0.76$ mV. The temperatures are 6, 7, 9, 11, and 15 K for different curves, respectively. Either the area or the height defined as is shown in the upper inset for $T=6$ K are used for the intensity of the lowest-lying peak. The lower inset displays the temperature dependence of these parameters (squares - area, dots - heights) normalized at $T = 7$ K. Three more pairs of dots are added which correspond to the other contact (not shown) whose characteristic is presented in Ref.[30]. Note the onset temperature $\approx 15$ K which is substantially higher than $T_N = 10.5$ K and $T_c = 6$ K. $H = 0$. 
ErNi$_2$B$_2$C-Ag (polycrystal)
$R=16.5\ \Omega$

(b) $\Gamma$ [meV]

(c) $Z$

Yanson "Andreev-reflection..."
Fig.3/8
HoNi$_2$B$_2$C-Ag

$T_c = 8.6 \, K$

$T_c = 6.6 \, K$

Voltage [mV]

Resistance

0.1 Ω
ErNi$_2$B$_2$C-Ag

$V_2$ [µV]

Voltage [mV]

$H > H_{c2}$

$H=0$

Yanson "Andreev-reflection..."

Fig.6/8
Figure 7/8: Graph showing the relationship between voltage [mV] and the derivative of the natural logarithm of resistance with respect to voltage [1/V] for HoNi$_2$B$_2$C and DyNi$_2$B$_2$C.
