

Point-contact Andreev reflection spectroscopy of a magnetic superconductor $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$

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The Andreev reflection spectra $dI/dV(V)$ of the magnetic superconductor $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ have been investigated. Pronounced stimulation of superconductivity by an external magnetic field has been observed for the first time. The effect showed up as enhancement of the gap structure (and hence the gap itself) in the spectra and its shift towards higher voltages with an increasing field. In the intermediate fields the structure also behaved strangely: instead of the usual smooth decrease with a growing field, the gap features dropped abruptly near the critical point H_{c2} . Of interest is also the abnormally high relative gap value $2\Delta/k_B T_c \approx 4$ (as compared to conventional singlet superconductors) which was found for some contacts from a comparison of experimental spectra and the modified Blonder–Tinkham–Klapwijk theory. We attribute the features revealed in the point-contact spectroscopic investigations of $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ in a magnetic field to the triplet-type Cooper pairing in the compound because only in this case one can expect the stimulation of superconductivity in the stationary magnetic fields up to $\sim 0.7H_{c2}$.

PACS: **74.45.+c** Proximity effects; Andreev reflection; SN and SNS junctions;
74.70.Dd Ternary, quaternary, and multinary compounds (including Chevrel phases, borocarbides, etc.);
74.20.Rp Pairing symmetries (other than *s*-wave) .

Keywords: point contact, Andreev reflection spectroscopy, magnetic superconductors, triplet pairing.

Introduction

It is well known [1] that in many compounds antiferromagnetism (AFM) coexists readily with singlet superconductivity in a wide temperature region because the magnetic moments compensate each other appreciably at distances comparable to the superconducting coherence length. Theoretically [2–4], in ferromagnetic (FM) substances the FM state and singlet superconductivity can coexist in a limited temperature region because in structures with a disturbed regularity of magnetic moments a change in their relative orientation minimizes the total magnetic moment. With the advent of the microscopic theory of superconductivity many researchers pointed imme-

diately to a basic possibility of triplet conductivity, i.e., the Cooper pairing of electrons with parallel spins. According to the latest data, this type of ordering is expected in newly synthesized uranium-containing FM superconductors UGe_2 [5], URhGe [6], UCoGe [7] in which *5f*-electrons cause both types of cooperative phenomena. Convincing evidence in favor of the triplet Cooper pairing was obtained in direct experiments on the strontium ruthenate Sr_2RuO_4 [8], only layered perovskite that becomes superconducting without the presence of Cu.

These findings have drawn much attention to other magnetic compounds in which coexistence of superconductivity and FM could be possible on a microscopic scale. These were the families of rare-earth molybdenum chalco-

genides ReMo_6X_8 (Re is a rare-earth element and X is a chalcogene) and rare-earth rhodium borides ReRh_4B_4 which possess a wide diversity of magnetic and superconducting properties [1]. In some of these compounds FM and superconductivity coexist in a rather narrow temperature region below the Curie point. The effect was most pronounced in ErRh_4B_4 [9] in which superconductivity appears at ~ 8.7 K and persists after the FM transition, $T_C \sim 1.2$ K, down to ~ 0.8 K demonstrating thus a considerable (~ 0.4 K) region of ferromagnetism–superconductivity coexistence. A similar behavior was also observed in HoMo_6X_8 .

Of equal interest is another rare-earth rhodium boride DyRh_4B_4 which can exist in several phase modifications but only one of them (most complex technologically) can be superconducting. Employing special technologies, the authors of [10] succeeded in synthesizing and investigating this phase. Besides, the close atomic radii of Dy and Y made it possible to prepare a number of $\text{Dy}_{1-x}\text{Y}_x\text{Rh}_4\text{B}_4$ derivatives ($0 \leq x \leq 1$). It was found that in this system the critical temperature of the superconducting transition T_c changes smoothly from 4 to 10 K as x increases from 0 to 1.

The system with $x \geq 0.4$ was found to undergo two magnetic transitions: a FM transition during which the Curie temperature T_C decreased from ~ 40 to ~ 12 K as the index x changed from 0 to 0.4 and an AFM transition at $T < T_c$. Note that compounds with higher yttrium-content ($x > 0.4$) are not magnetic. The authors analyzed the magnetic and resistive characteristics of some samples and concluded that the triplet type pairing was quite possible at certain temperatures. Later [11,12] the first transition was identified as ferrimagnetic, in which case the magnetic structure consists of two sublattices with unequal and opposite directed magnetic moments. This however does not prohibit its coexistence with superconductivity.

A compound of this family ($\text{Dy}_{0.8}\text{Y}_{0.2}\text{Rh}_4\text{B}_4$) was used to form a point contact (PC) with Au, and the Andreev reflection spectra $dI/dV(V)$ [11–13] and the dependence $H_{c2}(T)$ were measured on it in a wide range of temperatures and magnetic fields. By analyzing the measured spectra the authors obtained the temperature, $\Delta(T)$, and magnetic field, $\Delta(H)$, dependences of the order parameter. They differed considerably from the classical dependences of conventional type II superconductors. The difference was particularly striking in $\Delta(H)$ at $T < T_N$ (T_N is a temperature of AFM transition). In our opinion, this deviation is in favor of the previous assumption [10] of the triplet mechanism of Cooper pairing in the system $\text{Dy}_{1-x}\text{Y}_x\text{Rh}_4\text{B}_4$. The analysis of the magnetic field characteristics of the $\text{Dy}_{0.8}\text{Y}_{0.2}\text{Rh}_4\text{B}_4$ compound prompts a similar conclusion.

In this work we have investigated a compound of somewhat different composition — $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$. The effect of the magnetic field upon the PC Andreev reflection spectra $dI/dV(V)$ was investigated mainly at 1.6 and 4.2 K. In a certain range $\sim (0.5\text{--}0.7)H_{c2}$ the magnetic

field was found to enhance superconductivity rather than suppress it. We attribute the effect to the spin-triplet type of pairing in this compound because superconductivity stimulation by a stationary magnetic field is only possible when spins of the electrons in pairs are oriented in parallel.

Experiment

The samples of $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ were prepared by arc-melting the starting components and subsequent annealing for several days. According to the x-ray phase and structural analyses, the resulting objects were single-phase polycrystals with the LuRu_4B_4 type structure (space group $I4/mmm$). The critical superconducting transition temperature was about 7.0 K (as counted off from the midpoint of the resistive transition) (Fig. 1). A partial substitution of Ru for Rh permitted synthesis under the normal pressure, which would be impossible otherwise. According to the electron microscopic analysis, the samples had a close-packed structure consisting of approximately equiaxial crystallites whose sizes varied from several units to several tens of micrometers. Many of the crystallites had submicron-thick layers at their boundaries which might be non-identified inclusions.

The PC Andreev reflection spectra, $dI/dV(V)$ -characteristics, of N–S contacts were investigated in a wide range of voltage biases much exceeding the gap sizes. This permitted us to control the excess (Andreev) current and to exclude unstable contacts from consideration. The spectra were taken on fresh fractures of small (2–3 mm across) samples broken off a bulk ingot. A counterelectrode was an Au wire sharpened mechanically and etched chemically.

The measurements were made mostly at 1.6 and 4.2 K in magnetic fields varying from zero to the critical value. A reasonable electrical and mechanical stability was achieved only on the contacts whose resistance R_N was within several tens of Ohms (R_N is the contact resistance in the normal state). Gauging the sizes of the $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ -based contacts is rather a challenge for the lack of infor-

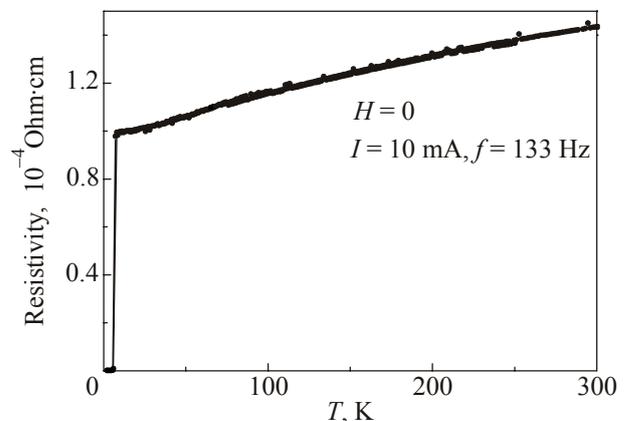


Fig. 1. The resistive transition of the $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ sample into the superconducting state.

mation about their basic properties in literature. We believe that the high excess current I_{exp} in the contacts selected indicated for spectroscopic conditions in our experiments, i.e., the contact sizes were smaller or at least comparable to the inelastic mean free path of electrons.

The PC spectra $dI/dV(V)$ were measured using the standard modulation method and synchronous detection with simultaneous computer recording. They were then processed in terms of the modified Blonder–Tinkham–Klapwijk (BTK) theory [14–16] which is practiced widely for parameterization of N–S point contacts. Despite some serious simplifications, the theory ensures adequate descriptions of the superconducting characteristics of conventional s -wave superconductors with an isotropic gap function $\Delta(k)$. Besides, the theory is efficient at estimating qualitatively the angular dependence $\Delta(k)$ in anisotropic single-crystalline or at least coarse-grained superconductors from directed PC spectroscopy data provided that the Fermi momenta in the contacting electrodes are significantly different. This is possible because the raster of the quasiparticles injected from the normal metal narrows considerably to the extent of the Fermi momenta ratio k_{FN}/k_{FS} [17,18]. The effect of narrowing is favored by the contact geometry (elongated channel) which we expect from our preparation technique. In addition to two basic parameters — gap Δ and barrier Z , characterizing the penetrability of the N–S boundary, the modified BTK theory includes the spectrum smearing parameter Γ which describes both the pair-breaking processes and the nonuniform distribution of Δ over the contact area.

Results and discussion

The typical magnetic field set of PC spectra $dI/dV(V)$ for the contact $\text{Au-Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ (normal resistance $R_N \approx 3.7 \Omega$) taken in various magnetic fields ($0-H_c2$) at 1.6 K is shown in Fig. 2. Similar sets were also registered within the temperature range ~ 1.6 – 2.0 K on the stable contacts permitting a complete cycle of measurement. They were little more than ten altogether. The unstable contacts also demonstrated similar spectra but they were influenced by electric and mechanical perturbing factors, which prohibited measuring a complete set.

The high quality of the investigated contacts is attested by the large excess (Andreev) currents I_{exc} that changed but little in the overgap region of voltages ($V \gg \Delta/e$). For the contacts whose spectra are illustrated in Figs. 2 and 5 I_{exc} makes about 50 and 80% of the BTK value for a one-dimensional model of a contact. It is obvious that I_{exc} of a three-dimensional contact should be higher but only slightly on account of the difference between the Fermi momenta in the contacting electrodes and the expected shape of the contact area (elongated channel).

We also measured temperature sets of spectra on several contacts in a zero magnetic field (not discussed here).

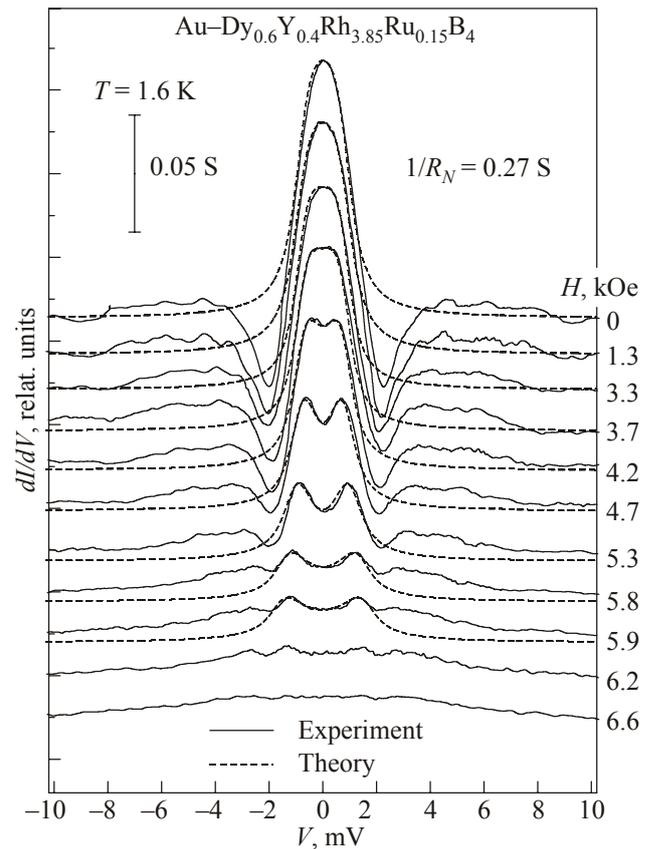


Fig. 2. Representative set of the Andreev spectra ($dI/dV(V)$) for a typical contact with $R_N \approx 3.7 \Omega$ exhibiting a considerable enhancement of the gap structure in a magnetic field at $T = 1.6$ K. The BTK fitting of the spectra is shown by dash curves. The magnetic field is specified at each curve. The fitting revealed the tendency of the dimensionless barrier parameter Z to grow with the field, kOe: 0.1 (0); 0.13 (2.63); 0.16 (3.29); 0.26 (3.95); 0.34 (4.48); 0.42 (5.21); 0.34 (6.06), the smearing parameter $\Gamma \approx \approx 0.1$ meV being invariant. For clearness, the curves are arbitrarily displaced vertically.

They had no features. The onset of the superconducting transition T_c^{on} evidenced by an appreciable zero-bias maximum in the curve $dI/dV(V)$ was within 6.7–6.9 K, which is slightly different from the corresponding value for a bulk sample (Fig. 1) and is further proof of the high quality of our contacts. The obtained T_c was about 1 K higher than T_c of $\text{Dy}_{0.8}\text{Y}_{0.2}\text{Rh}_4\text{B}_4$ [12]. This is because of the lower content of magnetic Dy and fits the data obtained in the first study of the electric and magnetic characteristics of the $\text{Dy}_{1-x}\text{Y}_x\text{Rh}_4\text{B}_4$ system [10].

It was rather hard to detect significant visual distinctions between the temperature PC spectra taken in a zero magnetic field and the spectra of conventional superconductors. However, the difference was drastic when the spectra were measured in a magnetic field near $T = 1.6$ K (Fig. 2). An example of a trivial spectrum is illustrated in Fig. 3 of Asen and Keck [19]. The magnetic field spectra of our contacts have two significant distinctions. Firstly,

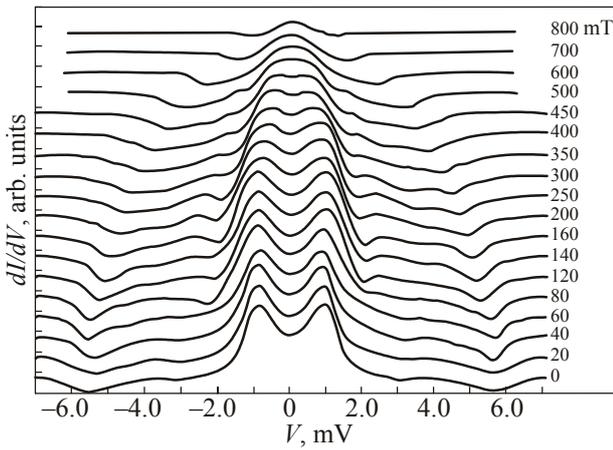


Fig. 3. Dynamic conductance versus the applied voltage for a Ta–Ag point contact ($R = 3.56 \Omega$, $T = 1.5 \text{ K}$) in different magnetic fields (0–899 mT) [19].

the spectra taken in a zero magnetic field have no double gap maxima near $V = 0$ that are expected when the contacting electrodes have different Fermi momenta or a thin dielectric layer appears at the N–S boundary [20] which is natural for conventional superconductors.

However, when nonconventional superconductors (cuprates, heavy-fermion compounds and more recent iron pnictides and chalcogenides) come into contact with a N-electrode whose k_{FN} is much higher, the correlation between the gap maxima intensity and the ratio k_{FN}/k_{FS} is rather weak, if any. This contradicts the classical theory. We also observed this in the PC spectra of $\text{EuAsFeO}_{0.85}\text{F}_{0.15}$ [21] in which the Fermi velocities v_F in the contacting electrodes differed up to eightfold. This should suggest a tunnel regime with high gap maxima and practically zero Andreev current. Nevertheless, these were pure Andreev-type spectra with weak gap maxima or without them at all. This discrepancy was first noted and interpreted by Deutscher and Nozieres [22] who assumed that the electron mass renormalization responsible for the effective Fermi velocity v_F is much weaker in the N–S contact area than that in the bulk material, which caused a significant departure of the gap structure of exotic superconductors from the classical BTK predictions [20].

The other and more essential distinction of our spectra measured at 1.6 K (Fig. 2) from classical ones is an enhancement of the gap structure in an increasing magnetic field. Initially, in a low magnetic field, the central maximum caused by the Andreev reflection is broadened. It should be emphasized that the width of this maximum is directly related to the magnitude of the gap in any of the existing models for the time being, which can be used to calculate the electrical characteristics of N–S contacts. At a certain moment classical double maxima form in the spectra, just like in the N–S contacts based on conventional superconductors. In this case, their position on the energy scale accurately determines the magnitude of the gap itself,

provided a small smearing of the spectra ($\Gamma \ll \Delta$). As the field grows further, the maxima intensity increases to a certain level and then the processes reverses ending in almost complete suppression of the maxima. The gap maxima voltages also grow up to a certain stable value which persists until the critical point H_{c2} is reached. This surprising behavior is clear evidence of superconductivity stimulation by a stationary magnetic field.

There is one more spectroscopy-unrelated feature in our PC spectra — dips of differential conductivity at voltages exceeding noticeably those of the gap. The dips account for the excessive resistance of the N–S boundary. The excessive resistance has been known for decades since [23,24] but its first adequate explanation appeared in [25] where it was attributed to disturbance of the balance between the chemical potentials of the Cooper pairs and normal quasiparticles due to significant current injection to the N–S structure. Later the interpretation was supported in numerous independent studies. Equalization of the potentials is commonly described simply and rigorously in terms of the relaxation times τ_Q of charge imbalance between the quasi-electron and quasi-hole branches in the energy excitation spectrum of superconductors (see, e.g., [26]). The equalization is achieved mainly through the interaction between nonequilibrium quasiparticles and phonons. The latter are rather scanty at low temperatures and low excitation energies $eV \ll \hbar\omega_D$ (ω_D is the Debye frequency), which accounts for the relatively long time of energy relaxation τ_E of quasiparticles (up to 10^{-9} s). In the hierarchy of characteristic relaxation times of superconductors τ_Q is significantly higher than τ_E , which makes the reason for the excessive resistance at the N–S boundary quite obvious. The problem was analyzed for N–S point contacts and an expression was derived to describe the excessive resistance in such structures [27].

As previously, we found the magnetic field dependence of the order parameter $\Delta(H)$ by matching our experimental spectra (Figs. 2 and 5) with the modified BTK theory including the smearing parameter Γ [15]. Usually, in the case of conventional superconductors the barrier parameter Z , estimated for the lowest-temperature zero-field $dI/dV(V)$ curve of each set of spectra, was practically invariant for curves measured in higher fields. This occurred to be improper for our contacts $\text{Au-Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ because magnetic field caused significant transformations in the gap structure, characterized to a large extent by the parameter Z (an example of Z -variations is illustrated in the caption to Fig. 2). This Z -growth can be explained by the electron mass increase in a magnetic field as it was multiply observed in U -based ferromagnetic superconductors [28]. Similarly, such phenomenon is quite possible in the magnetic compound studied here. So, the initial weakness of renormalization effects in the contact area (according to Deutscher and Nozieres [22]) could be compensated by the electron mass enhancement in a magnetic field. This

should result in an increase of the gap maxima intensity in the field as it really takes place in our experiments.

The Δ -values found from the BTK-analysis of the spectra in Fig. 2 are plotted as a function of the magnetic field $\Delta(H)$ in Fig. 4. This figure also carries two possible theoretical dependences $\Delta(H)$ commonly used for a comparison with experimental results. They were calculated for the bulk state [28,29] and the thin films in a parallel magnetic field [30] of conventional type II superconductors. Either of them is applicable to describe PC data depending on the geometry of the experiment (the relative orientation of the contact and magnetic field axes). An intermediate sort of dependence is also possible.

According to the analysis of PC spectra, most of the contacts measured in a zero magnetic field at $T \sim 1.6$ K characterized by the gap within 0.6–1.2 meV ($2\Delta/kT_c = 2.0$ –4.0 in reduced units). The upper limit of the range is indicative of an exotic character of the Cooper pairing in the compound investigated. In conventional superconductors the above ratio is close to 3.52 (in conformity with the BCS theory) and reaches 4 only in substances with a strong electron-phonon interaction (e.g., Hg and Pb). Moreover, in conventional superconductors a high characteristic ratio $2\Delta/k_B T_c$ is possible only in the nonmagnetic state. Meanwhile our compound contains rare-earth element Dy with a relatively large magnetic moment ($\sim 8\mu_B$). As is well known, the order parameter decreases rapidly when intrinsic magnetic moments or external fields affect the singlet superconductors. We observed an opposite effect in our experiments.

The PC spectra of $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ exhibited an anomalous behavior in a magnetic field in the whole

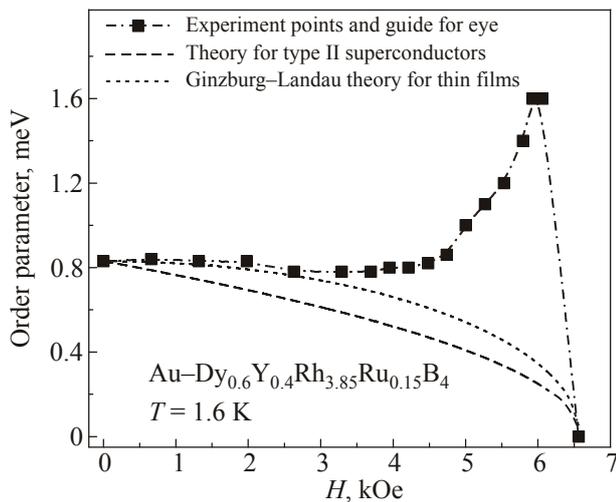


Fig. 4. The dependence of the order parameter upon the magnetic field $\Delta(H)$ at $T \approx 1.6$ K for the contact whose spectra are illustrated in Fig. 2. For comparison, two theoretical dependences (broken lines) are shown, which are possible in contacts based on conventional superconductors when the contact axis is along or perpendicular to the field.

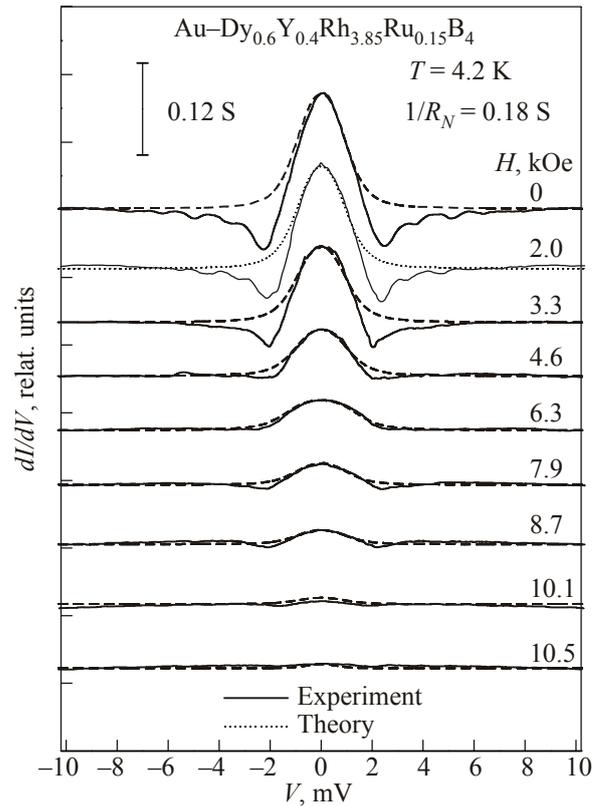


Fig. 5. A typical set of magnetic field PC spectra for one of the contacts with $R_N \approx 5.5 \Omega$ (solid lines). An acceptable coincidence of experimental and BTK-calculated spectra (broken lines) was obtained using invariant fitting parameters $Z \approx 0.1$ and $\Gamma \approx 0.1$ meV. For clearness, the curves are arbitrarily displaced vertically.

range of the temperatures used, $T = 1.6$ –4.2 K (spectra in Fig. 5 are measured at 4.2 K and generally we did not go above this temperature). It is therefore hardly reasonable to attribute the phenomenon observed to a magnetic transition below 4.2 K where some compounds of the $\text{Dy}_{1-x}\text{Y}_x\text{Rh}_4\text{B}_4$ family experience certain magnetic transformations. The magnetic field does not stimulate gap maxima in the spectra at 4.2 K (Fig. 5) but they always appear in the spectra at 1.6 K (Fig. 2). However, the fact that the central maximum in Fig. 5 does not become narrower with an increasing field (its width correlates directly with Δ) and decreases sharply near H_{c2} agree basically with the data at 1.6 K. The effect of the magnetic field at $T = 4.2$ K is seen more clearly in the dependence $\Delta(H)$ (Fig. 6) derived from the BTK analysis of the spectra in Fig. 5. The observed effect becomes weaker as the temperature increases (of Figs. 4 and 6).

We suggest that the anomalous behavior of a PC spectrum in a magnetic field is caused by the triplet-type Cooper pairing in the $\text{Dy}_{0.6}\text{Y}_{0.4}\text{Rh}_{3.85}\text{Ru}_{0.15}\text{B}_4$ compound. The concept makes it easy to explain the enhancement of the gap structure in the PC spectra. Indeed, when the electron spins of the Cooper pairs are parallel, the applied field

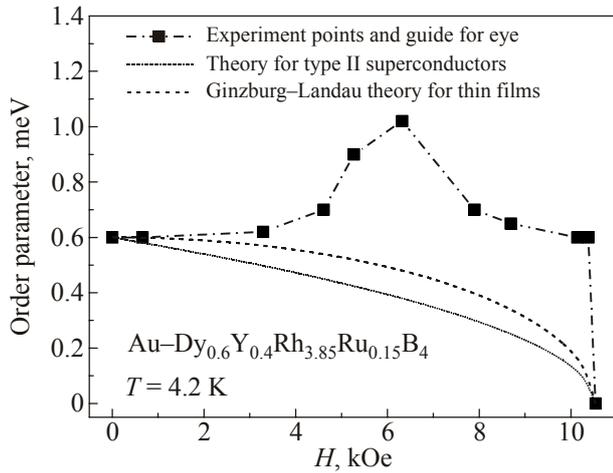


Fig. 6. The BTK-calculated dependence of the order parameter on the magnetic field for the spectra shown in Fig. 5. For comparison, the figure illustrates two theoretical dependences (broken lines) expected for contacts based on conventional singlet superconductors.

stabilizes the parallel orientation of them resulting in the enhance of superconducting parameters. In this case the pair potential (order/gap parameter) determining the energy of the electron coupling in the Cooper pairs can hold its intensity up to the critical point where superconductivity is destroyed by the orbital magnetic moments. It is precisely these moments are responsible for the progressive reduction of the Cooper pairs and hence the intensity of the gap features in an increasing magnetic field.

If superconductivity stimulation occurred only in weak fields, it could be attributed, within a singlet model of pairing, to a suppression of possible disturbance of the magnetic order at $H \ll H_{c2}$. This would enhance the condensate stability and somewhat increase the gap voltage. However, the assumption is hardly reasonable because the effect exists in a wide range of fields and no smooth decrease in Δ occurs near H_{c2} . Other possible factors (extraneous inclusions of different phase compositions or dielectric layers in the PC region) are meaningless for this consideration as the critical parameters of all the contacts were practically invariant. Moreover, as the BTK-estimates show, in some cases the excess current I_{exc} can reach $\sim 80\%$ of the corresponding theoretical value and never decreases below $\sim 25\%$.

It should be noted that the superconducting characteristics of the related compound $Dy_{0.8}Y_{0.2}Rh_4B_4$ [12,13] had some features that could be attributed to the triplet-type pairing, at least below the magnetic transition point near 3.5 K. But those PC spectra had no striking anomalies (like in our spectra) though the compounds have close elemental compositions. The lower content of Dy in our sample only reduces the magnetic effect and the partial substitution of Ru for Rh (for technical reason) can hardly influence its properties because these elements occupy neighboring positions in the periodic table and differ only in one electron in the 4d-shell.

It is obvious that further broader research by various techniques is necessary to clear up the origin of the strong anomalies in the PC spectra of $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ in a magnetic field and to substantiate the possibility of the triplet-type pairing in this compound.

Conclusions

1. The PC Andreev reflection spectra $dI/dV(V)$ have been investigated in N-S contacts based on the magnetic superconductor $Dy_{0.6}Y_{0.4}Rh_{3.85}Ru_{0.15}B_4$ in different magnetic fields, the critical temperature of the onset of the superconducting transition being $T_c^{on} = 6.7-6.9$ K.

2. When the magnetic field grows, the gap features of the spectra (and hence the gap/order parameter) do not shift towards lower energies, as in classical spectra; on the contrary, they move in the opposite direction and gain intensity. After reaching a maximum and the following loss of their intensity they still remain practically non-shifted on the energy axis up to the critical point H_{c2} where the superconducting state disappears in a stepwise manner.

3. We suggest that a triplet mechanism of Cooper pairing operates in the compound investigated because stimulation of superconductivity by an external stationary magnetic field is possible only in this case. The assumption permits a reasonable explanation of the high (up to 4) ratios $2\Delta/k_B T_c$ unusual for singlet magnetic superconductors.

4. The high Andreev current (up to $\sim 80\%$ of the BTK estimate for a one-dimensional case) in some contacts makes the presence of extraneous inclusions in the PC area, resulting in the destructive modification of the spectra, improbable. This is also supported by the close critical parameters of our point contacts and the bulk material.

5. To clear up the origin of the effect observed, it is necessary to have information about the electron and magnetic structures of the object studied. This calls for comprehensive investigations by various techniques of its electric and magnetic characteristics, including the PC properties, in a wide range of temperatures and magnetic fields.

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