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Elastic point contact spectroscopy of electron-phonon interaction in superconductive lead

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A method for experimental determination of the spectral function of electron-phonon interaction (SF EPI) in superconductors with strong coupling has been developed. The method is based on analysis of non-linear current-voltage characteristics of point heterocontacts superconductor-normal metal. The nonlinearities arise in the elastic component of the current through the hetero-contact due to dependence on energy of the complex function of the energy gap in superconductor, and they have a similar nature as ones observed in superconducting tunnel contacts. For the purpose of present study the lead as a typical superconductor with strong EPI was chosen. In order to obtain the SE EPI the Eliashberg equation was solved numerically following the McMillan-Rowell method. Our solution is in good agreement with the results of elastic tunnel spectroscopy of superconductors as well as with the data provided by the inelastic point contact spectroscopy in the normal state.

Keywords: superconductivity, electron-phonon interaction, point-contact spectroscopy, lead.

Развит метод экспериментального определения спектральной функции электрон-фононного взаимодействия (СФ ЭФВ) для сверхпроводников с сильной связью. Данный метод основан на анализе нелинейностей вольт-амперной характеристики точечного гетероконтакта сверхпроводник-нормальный металл, которые возникают в упругой компоненте тока через гетероконтакт вследствие зависимости комплексной функции энергетической щели сверхпроводника от энергии квазичастиц и имеют общую природу с наблюдаемыми для сверхпроводящих туннельных контактов. Объектом исследования выбран свинец как типичный сверхпроводник с сильным ЭФВ. Полученная путем численного решения уравнений Элиашберга по методу Макмиллана-Роуэлла СФ ЭФВ согласуется с результатами упругой туннельной спектроскопии сверхпроводников и неупругой микроконтактной спектроскопии в нормальном состоянии.

Ключевые слова: сверхпроводимость, электрон-фононное взаимодействие, микроконтактная спектроскопия, свинец.

Розвинено метод експериментального визначення спектральної функції електрон-фононної взаємодії (СФ ЕФВ) для надпровідників із сильним зв'язком. Даний метод заснований на аналізі нелінійностей вольт-амперної характеристики точкового гетероконтакту надпровідник-нормальний метал, які виникають у пружній компоненті струму через гетероконтакт внаслідок залежності комплексної функції енергетичної щілини надпровідника від енергії квазічастинок і мають спільну природу із такими, що спостерігаються для надпровідних тунельних контактів. Об'єктом дослідження обрано свинець як типовий надпровідник з сильною ЕФВ. Отримана шляхом чисельного розв'язання рівнянь Еліашберга за методом Макмілана-Роуела СФ ЕФВ узгоджується з результатами пружною тунельної спектроскопії надпровідників і непружної мікроконтактної спектроскопії у нормальному стані.

Ключові слова: надпровідність, електрон-фононна взаємодія, мікроконтактна спектроскопія, свинець.

Formulation of the problem

Method of inelastic point contact spectroscopy of the electron-phonon interaction (EPI) in metals [1,2] that was proposed in [3] and later was theoretically substantiated in [4] is based on analysis of small nonlinearity of the point contact current-voltage characteristic (IVC) caused by generation of the nonequilibrium phonons within the contact. The method makes it possible to determine the spectral function of the EPI (SF EPI) that is regarded to be the most detailed characteristic of the electron-phonon system. In the case of the superconducting point contacts (S-c-S) and the point heterocontacts superconductor-normal metal (S-c-N) the theory of the inelastic point contact spectroscopy EPI in superconductors with weak EPI [5,6] had demonstrated its validity throughout the cycle of works [7] where for the first time manifestations of the EPI-related effects were observed experimentally in the characteristics of superconducting point contacts. At the same time the experimental studies [7,8] revealed several discrepancies with predictions of the theory [5,6] that were

not without a reason attributed to the strong EPI in [8] the superconductor (Pb) used in the mentioned experiments. Later on the theory of conductivity of the S-c-N contacts containing a strong-EPI superconductor [9] provided a proof that the elastic component of the current through the contact is a nonlinear one and that the nonlinearities were caused by dependence of the function of the energy gap on the energy in a very similar way to the elastic tunnel spectroscopy of the EPI in superconductors [10,11].

The normalized differential conductivity of the S-c-N point contact in the clean limit (i.e. the "Knudsen limit" that is called sometimes the ballistic current regime) equals in accordance to [9]

$$G(\omega)=\frac{\sigma_S}{\sigma_N}=1+\left|\frac{\Delta(\omega)}{\omega+\sqrt{\omega^2-\Delta^2(\omega)}}\right|^2,\ \text{eV}=\hbar\omega \quad (1)$$

and it differs from the normalized conductivity of a tunnel contact [10,11]

$$N(\omega)=\frac{\sigma_S}{\sigma_N}=\mathrm{Re}\frac{\omega}{\sqrt{\omega^2-\Delta^2(\omega)}},\quad \text{eV}=\hbar\omega \qquad (2)$$

which is the tunnel density of states. In (1,2) σ_S, σ_N are the contact differential conductivities for the superconducting and the normal state respectively of the superconducting electrode that is made of a metal under investigation, $\hbar\omega$ is the energy of quasiparticles, e is the elementary charge, and V is the biasing voltage applied to the contact.

In the context of the method of elastic tunnel spectroscopy the normalized differential conductivity is employed for reconstruction of the SF EPI by means of numerical solution of the Eliashberg equations of the phonon model of superconductivity.

The mentioned above considerations provide a ground for formulation of the goal of the current research. Namely the basic idea of it is in the use of the point contact experimental data instead of the data associated with the tunnel effect for reconstruction of the EPI function by solving the Eliashberg equations.

Method of reconstruction of the EPI function

Calculations of the SF EPI g(ω) are based on numerical solution of the system of two integral Eliashberg equations at T = 0 for an isotropic superconductor [12,13]:

$$\Delta(\omega)=\frac{1}{Z(\omega)}\int_{\Delta_0}^{\omega_c}\mathrm{Re}\left\{\frac{\Delta(\omega')}{\left[\omega'^2-\Delta^2(\omega')\right]^{1/2}}\right\}* \qquad (3)$$

$$*\left[K^+(\omega,\omega')-\mu^*\right]d\omega',$$

$$\left[1-Z(\omega)\right]\omega=\frac{1}{Z(\omega)}\int_{\Delta_0}^{\omega_c}\mathrm{Re}\left\{\frac{\omega'}{\left[\omega'^2-\Delta^2(\omega')\right]^{1/2}}\right\}*$$

$$*K^-(\omega,\omega')d\omega',$$

where the cores are

$$K^{\pm}(\omega,\omega')=\int_0^{\infty}g(\omega'')\left(\frac{1}{\omega''+\omega'+\omega+iO^+}\pm\right.$$

$$\left.\pm\frac{1}{\omega''+\omega'-\omega-iO^+}\right)d\omega''.$$

Here Δ(ω) is the complex function of the energy gap, Z(ω) is the complex renormalization function, μ^* is the coulomb pseudopotential, and ω_c is the cut-off frequency. The latter is usually taken as $\omega_c \approx 5\omega_{max}$, where ω_{max} is the highest frequency of the single-phonon part the EPI spectrum.

We used the McMillan-Rowell method [10,14] for our search of the solution of the equations (3). This iteration method includes two nested cycles called the outer and the inner iteration loops. The input parameters for the inner loop are the experimentally determined half-width of the energy gap Δ_0, the seed value for the coulomb pseudopotential μ^*, and a reasonable zero-approximation $g^{(0)}(\omega)$ for the g(ω) function (see for example [12]). These parameters are set to the Eliashberg equations, and the solution for the function Δ(ω) that is a complex one is corrected in accordance with the following condition $\Delta(\Delta_0) = \Delta_0$. After that the value μ^* is used again along with the $g^{(0)}(\omega)$ for getting the next approximation for the Δ(ω), and so on. The iteration process in the internal loop is stopped when the difference between the last Δ(ω) and next to the last one riches experimentally sound limit. The function Δ(ω) obtained in the inner loop is employed for the use in the outer loop for calculations of the normalized conductivity of a contact that is along with the experimentally measured normalised conductivity serves in turn for further correction of the initial zero approximation $g^{(0)}(\omega)$ for the EPI function. The latter procedure is based on the minimization by means of the variation method of the functional derivative $\partial[\sigma_S(\omega)/\sigma_N(\omega)]/\partial g(\omega)$ [14]. The EPI function $g^{(1)}(\omega)$ of the first approximation produced in the outer iteration loop then feeds the inner loop for further adjustment of the Δ(ω). The function $g^{(2)}(\omega)$ is then found in the same way, and the process lasts until the limit of experimental accuracy for the contact conductivity is reached. The corrections of the g(ω) are made within the interval ω_1 to ω_{max}, where $\hbar\omega_1 = \Delta_0 + \delta$. In practice $\delta \approx$ 1-2 meV. For the region $0 < \omega < \omega_1$

the approximation ~ ω^2 is in effect.

Measurement technique and experimental data

Lead was chosen for this study as a superconductor with one of the greatest values of the EPI constant λ = 1.55. It should be noted that the mercury only among the other metallic elements demonstrates the greater λ = 1.6 [10]. That is why results on the tunnel effect in lead are traditionally used in development of computer programs for reconstruction the SF EPI g(ω) by the McMillan-Rowell method [10,14] or by the competitive D'yachenko method [11,15] (see the references in [16]). The tabulated tunnel data are available from [1,17] (in [1] a table of the function g(ω) [18] computed by D'yachenko method is given), and can be employed for tests.

In our experiments the point contacts were connected in the 4-wire scheme way to the measurement setup operating in the current-source mode for both the biasing and the modulation signals. The dependencies

$$V_1(V) = \frac{i}{\sqrt{2}} \frac{dV}{dI}(V),$$

and

$$V_2(V) = \frac{i^2}{4\sqrt{2}} \frac{d^2V}{dI^2}(V)$$

were measured. Here V_1 and V_2 are detected effective voltages corresponding to the first and the second harmonic of the modulation current with the amplitude i. These dependencies are proportional to the first and to the second derivatives of the IVC as the functions of the applied voltage bias V. For the conventional normal state point contacts such the dependencies are called the point contact spectra, and they routinely serve for reconstruction of the point contact EPI functions $g_{pc}(\omega)$ [1,19].

The sliding-type point contacts [1] were produced in the course of the experiment. The superconducting sample electrodes were spark-cut from the bulk polycrystalline high-purity C-000 lead. For making the normal counter-electrodes that needed for fabrication of the heterocontacts we used high-quality single crystals of Ru ($\rho_{300K}/\rho_{4.2K} \approx$ 2500). While making the Pb/Ru contacts the geometrical axis of the contact was oriented along the $[10\bar{1}0]$ direction in the Ru-electrodes to ensure provision of the most "hard" EPI spectrum in Ru [7] that does not overlap with the EPI spectrum of Pb (Fig.1).

Prior to mounting to the mechanical cell for point contact fabrication [20] and putting into He-atmosphere of the cryostat the surface of the electrodes was exposed to electro-chemical etching. All the measurements were carried out at the temperature T = 1.5 K. For that temperature the energy gap 2Δ in the superconducting lead perfectly matches the value $2\Delta_0$ at T = 0. In the case of necessity in order to create the normal-state point contacts the superconductivity in the lead electrode was suppressed by the external constant magnetic field. The magnitude of the resistance of the point contact in the normal state R_0 was defined as R at $V \to 0$ using the initial linear region of the IVC. The typical values for the resistance in the normal state were about several Ohm.

Being one of the key input parameters for the task of solution of the Eliashberg equation (3) solution the value Δ_0 was determined experimentally by localizing the subharmonics of the gap at the IVC derivatives of the homocontacts Pb/Pb (Fig.2).

Subharmonics of the gap seen at the IVCs of the point contacts S-c-S are caused by the multiple processes of Andreev reflection (see for instance [2,10]), and manifest themselves as peculiarities at the derivatives of the IVC located at the voltages V = 2Δ/en, where n = 1,

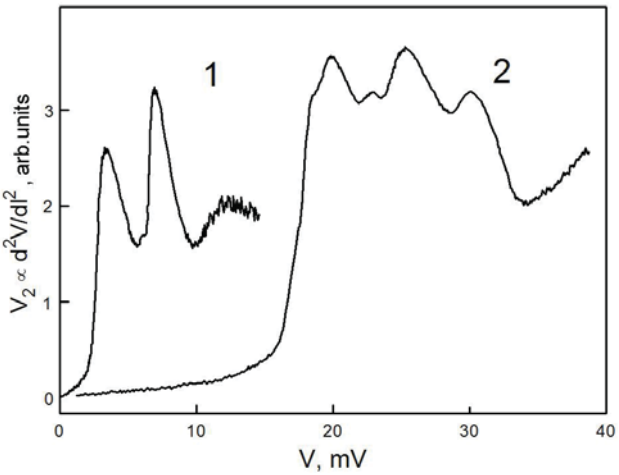

Fig.1. Inelastic EPI point contact spectra of lead (1) and ruthenium (2) captured at T = 1.5 K for the point contacts at the normal state. The superconductivity of lead is supressed by external constant magnetic field.

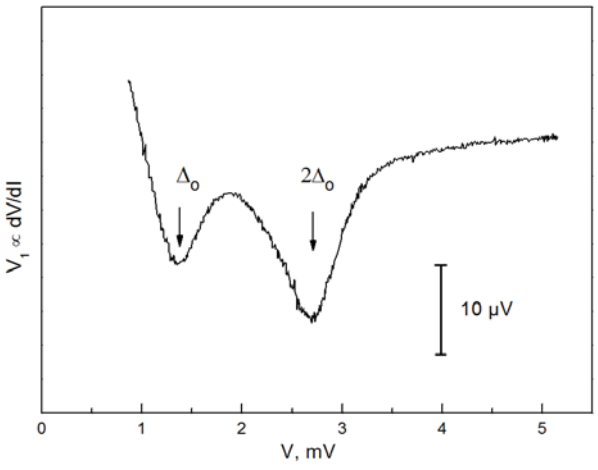

Fig.2. The subharmonic 2Δ/n (n = 1, 2, …) gap structure at the first derivative of the IVC of the superconductive lead point contact at T = 1.5 K. R_0 = 5.8 Ohm.

2, 3, … . Such the peculiarities correspond to minima at the dependencies of the differential resistance R(V) = dV/dI(V) vs voltage. We found the value Δ_0 = 1.35 meV for the superconducting lead. This figure precisely matches the one that was obtained earlier in the experiment on the tunnel contacts made of the lead taken from the same piece of metal [21].

It should be underlined that measurement of the energy gap using the subharmonic gap structure at the IVS derivatives is a direct experimental method. While the Blonder-Tinkham-Klapwijk method (BTK) [22] that is called Andreev spectroscopy requires cumbersome numerical calculations for retrieving the value of the Δ the accuracy of the direct experimental sub-harmonics based method is limited only by the precision of the voltmeter used for measurements of the voltage applied to the contact.

In the course of preparations for the numerical calculations employing the McMillan-Rowell method the dependencies $V_2(V)$ for the point heterocontacts Pb/Pu were measured for the case of the superconducting and the normal state of the Pb-electrode.

Results of the numeric calculations and discussion

The problem of determination of the SF EPI from the Eliashberg equations on the basis of point contact experimental data can be split into few tasks. The solution of them in context of the present work is described below.

In accordance to the theory [9] the differential conductivity of the S-c-N contact (1) exhibits maxima, and the differential resistance accordingly shows minima in the vicinity of the characteristic phonon frequencies that are shifted by the value of Δ_0 to higher energies in comparison to the phonon density of states (PDOS) F(ω) and the SF EPI. Therefore at the second derivatives $d^2V/dI^2(V)$ certain characteristic peculiarities in the form of the derivatives of a functional minimum should be seen. Lead has the face-centred cubic crystalline structure with one atom per the unit cell. Its PDOS is comparatively simple with its two-peak F(ω) structure. One of the peaks corresponds to the transversal acoustic oscillations and the other peak is due to the longitudinal ones. In order to make a comparison with the experimental results the first and the second derivatives of the S-c-N point contact IVC were calculated using the relationship (1) and the tables [17] of the real and imaginary parts of the Δ(ω) for Pb. The computations revealed minima of the contacts' differential resistance in the vicinity of the T- and L-maximum of the F(ω) and the EPI spectrum in lead. The amplitude of the calculated minima makes up few percent of the R_0 (Fig.3).

The numerically derived second derivative of the IVC (1 at the Fig.4) clearly demonstrates a non-monotonic structure as a result of the non-trivial form of the Δ(ω) and reproduces all the thin details of the experimental curve 3 at the Fig.4. At the same time a BCS-based calculation

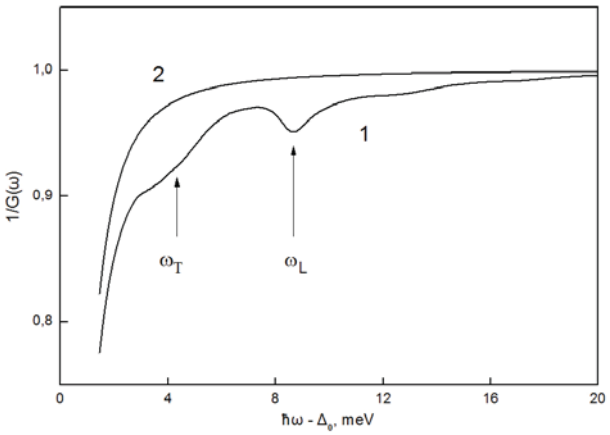

Fig.3. The calculated from the relationship (1) normalized differential resistance R ≡ 1/G of the S-c-N point contact:
1 – with the use of the function Δ(ω) for Pb [17];
2 – in accordance to the BCS model Δ(ω) = Δ_0 = const.

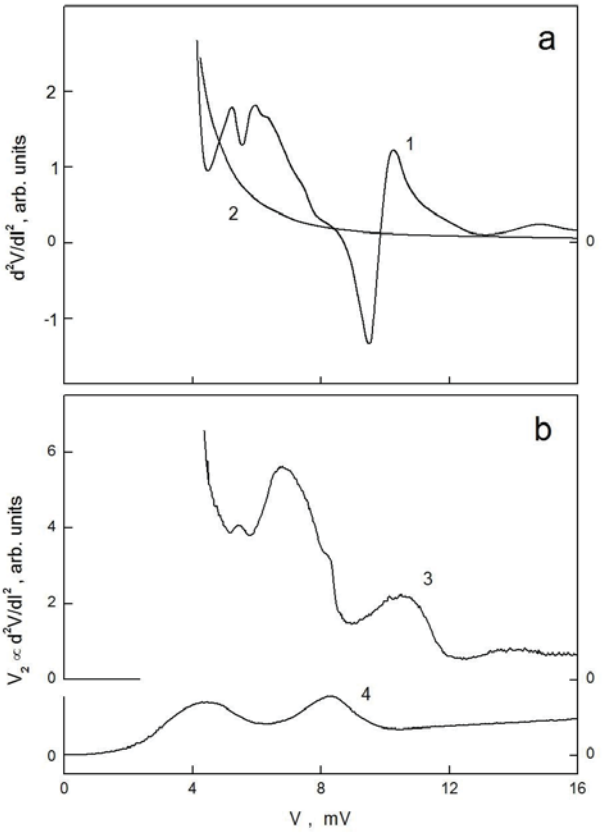

Fig.4. Comparison of the calculated (panel a) and experimental (panel b) IVC second derivatives:
1 – numerical calculations for the S-c-N contact following relationship (1) with the use of the function Δ(ω) for Pb [17]; 2 – the same calculation in the BCS model Δ(ω) = Δ_0 = const. 3, 4 – graphs $V_2(V)$ μ $d^2V/dI^2(V)$, captured at T = 1.5 K for the heterocontact Pb/Ru in the case of the superconducting (1) and the normal (2) state of thee lead electrode (the y-scale for 1, 2 is the same). R_0 = 6.5 Ohm.

produces featureless smooth dependencies $1/G(\omega) \propto dV/dI(V)$ and $d^2V/dI^2(V)$ (see the curve 2 at Fig.3 and 2 at Fig.4).

The well-known Nernst-Lindemann model when the PDOS is shaped by two characteristic phonon frequencies only provides a sound choice for the zero-approximation of the $g^{(0)}(\omega)$ in our case. In accordance with this model we assume presence of only two maxima of the T- and the L-type that in turn means complete degeneration of the oscillatory T-branches. The maxima are modelled by isosceles triangles of equal height. Every of the tree branches of the phonon spectrum contributes equally to the $g^{(0)}(\omega)$, and therefore for observation of the normalization criterion the area associated with the T-maximum is two times greater than the area of the L-maximum. In our modelling of the zero-approximation EPI function $g^{(0)}(\omega)$ we set locations for the T- and L- maxima as 4.4 and 8.5 meV respectively while the maximal EPI spectrum frequency was taken 10 meV. The latter value can be estimated as $\omega_{max} \approx 2 < \omega^2 >^{1/2}$, where the mean square phonon frequency $< \omega^2 >$ is calculated using the relationship [23] for the Debye temperature $\Theta_D = \frac{3}{2} < \omega^2 >^{1/2}$ which is derived from the theory of the momentums of frequency distributions.

In order to achieve better accuracy of the final product which is the EPI function we obtain the input dependence of the normalized differential resistance vs voltage by numerical integration of the measured second derivative of the IVC. Such a trick is often used in the course of handling the data of tunnel experiments [10]. The magnitude of the conductivity is normalized to the conductivity of the same point contact in the normal state when the superconductivity in the sample is destroyed by the external constant magnetic field. Prior to the normalization the inelastic contribution is removed from the superconducting state data. For that purpose we use the values $V_2(V)$ μ $d^2V/dI^2(V)$ for the contact in the normal state (4 at Fig.4) with the obvious substitution of the argument V to $V + \Delta_0$ [6,24].

To avoid the divergence in the transcendental equations (3) in the course of the calculations we produced expansions of the integrand into series in small parameter Δ^2/ω^2 as recommended in [25]. Following [26] the correction of the coulomb pseudopotential μ^* has not been made. At every step the value $\mu^* = 0.13$ [10] was adopted.

The resultant complex function $\Delta^{(2)}(\omega)$ has already been not far from the tabulated one [17]. Visibly good agreement of the $g(\omega)$ takes place if we compare it with the known functions for the film tunnel contacts [10,17] as well as with the results of the inelastic point contact spectroscopy in the normal state. At the Fig.5 the calculated EPI function $g(\omega)$ is presented along with the EPI point contact function $g_{pc}(\omega)$.

The latter function is derived from the point contact spectrum $V_2(V)$ of homocontact Pb/Pb in the normal state (1 at Fig.1) as an output product of the modern version of the

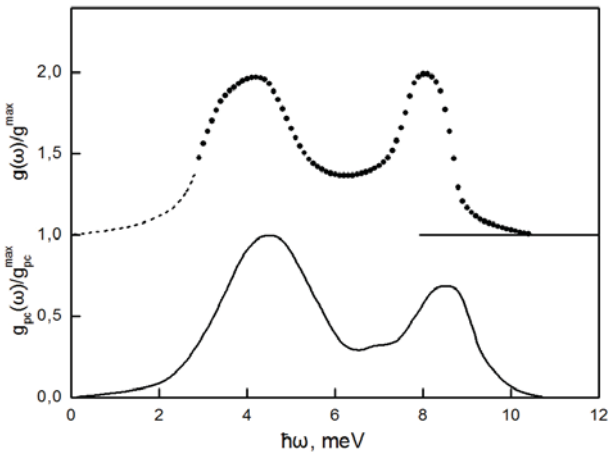

Fig.5. The spectral EPI functions in lead: the thermodynamic EPI function $g(\omega)$ derived by McMillan-Rowell method from the Pb/normal-metal point contact data, and the point contact EPI function $g_{pc}(\omega)$ reconstructed using Pb point contact spectrum in the normal state. For the $g(\omega)$ the beginning of the coordinate axis is shifted by 1 upwards.

program [19]. At the Fig.5 the EPI functions are displayed for comparison in relative units (being normalized to theirs maximal values). However the absolute values of the $g(\omega)$ function can be easily recovered following [25,27] by means of adjustment to the known transition temperature T_c = 7.19 K in lead (for instance by employing the McMillan formula).

The matching of the $g(\omega)$ function to its tunnel analogue clearly depends on the number of iterations in every cycle of the algorithm of solution of the Eliashberg equations. However the different relative intensities of the $g(\omega)$ and $g_{pc}(\omega)$ maxima at the Fig.5 are explained by presence of the form-factor for the $g_{pc}(\omega)$ [1] under the integral, and also by the temperature and instrumental smearing [7] of the sharper L-maximum at 8.5 meV in the EPI point contact spectrum in lead (1 at Fig.1) which was used as a starting position in the process of reconstruction of the $g_{pc}(\omega)$.

Conclusion

The primary results of the current work can be presented in the form of short statements.

1. For the first time the method of elastic point contact spectroscopy has been employed as a practically suitable tool for reconstruction of the spectral function of the EPI in superconductors by means of numerical solution of the system of Eliashberg equations. The proposed approach is based on use of experimental data for the conductivity of the point contact but not on the tunnel data.

2. In a short perspective the proposed approach is about to complement the arsenal of methods for investigation of new superconductors with high values of critical magnetic fields that are not always achievable in the laboratory environment.

3. The method of reconstruction of the SF EPI may be critically important in those cases when fabrication of the high-quality tunnel contacts turns out to be difficult or impossible and it becomes advantageous eliminating many problems of investigation the high-purity superconductors in the form of bulk samples or high-quality single crystals.

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