Thermoelectric Effect in the Normal Conductor–Superconductor Junction: A BTK Approach

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The thermopower of the junction between normal conductor and s-wave superconductor has been investigated. For this purpose we have analyzed in detail a simple generalization of the Blonder–Tinkham–Klapwijk theory by taking into account explicitly an energy dependence of the density of states near the Fermi level. Both linear and nonlinear thermopowers have been calculated for 3D free electron gas, 3D Fermi liquid, and the case with Van Hove singularity in the vicinity of the Fermi level. In the linear regime, for all models, the thermopower as function of temperature has a clear maximum with its position and the value depending strongly on the junction barrier strength. In the nonlinear regime, we have found very large values of the thermopower (up to $8k_{\mathrm{B}}/e$) and strongly asymmetric behavior with respect to the change of the temperature gradient sign.

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1. Introduction

Tunneling spectroscopy between normal and superconducting (NS) materials is one of the most effective tools to study the nature of the superconducting state [1, 2]. Despite the existence of energy gap in superconductor (SC), quantum transport is still possible for energies below the gap value by means of reflection of the incident electron as a hole with the opposite charge and momentum, as predicted by Andreev [3, 4]. Currents flowing across the junction of an arbitrary transparency were theoretically analyzed in the seminal paper of Blonder, Tinkham, and Klapwijk (BTK) [5]. They have discussed a continuous transition from the tunneling limit to the metallic regime. The theory was shown to be successful in describing experimental data of the current–voltage characteristics in point contacts [6–8].

There were many extensions of this theory, related to incorporating different nature of superconducting state: d-wave symmetry of the order parameter [9], the Fulde–Ferrell–Larkin–Ovchinnikov state [10, 11], hole superconductivity [12], two-band superconductivity [13], dimensionality of the problem [14, 15], as well as different type of the non-superconducting material of the junction: ferromagnet [16], semiconductor [17], graphene [18]. The theory was also successfully adapted to the molecular junctions [19]. The main strength of the BTK approach is its simplicity and possibility of studying exotic features in a straightforward manner.

Nonetheless, the electrical conductance does not provide the full understanding of the system. For example, it is important to look into the thermodynamic properties of particular materials and their electrical response to the temperature gradient. In general, the problem of thermoelectric phenomenon in superconductors and in the superconductive junctions was studied extensively both theoretically [20–31] and experimentally [32–34] with an astonishing precision. Namely, the temperature and the voltage is measured to the order of mK and μV, respectively. Also, in majority of those papers the thermoelectric effect was analyzed using the Green function approach. Explicitly, the Eilenberger–Usadel equations [35] were used, as they seem to be the most effective modern tool in the description of thermoelectric properties of superconductive systems. However, despite the great progress achieved in the field during last two decades, the thermopower of such systems is still not fully understood [36]. In the words of the recent review [36]: "a significant advance in the theory is required before the thermopower of mesoscopic proximity-coupled systems is understood".

In view of the successes of the BTK theory to describe conductance of the system it is of interest to study the thermopower also within this intuitive approach. Although it was employed earlier in some special cases [12, 37] in its simple form leads to zero thermopower irrespectively of the temperature difference over the junction. Here we propose a simple extension of the theory to include thermodynamic properties of the quantum fluids on both sides of the junction, particularly non-trivial density of states naturally gives rise to the Seeback effect.

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Additionally, as in the case of conductance, this approach can be further generalized to take into account specific features of some exotic materials (e.g., those with non-standard band mass distribution, nontrivial gap symmetry, realistic dimensionality of the system).

Usually, the effect of density of states (DOS) energy dependence in the NS junctions current characteristics is ignored. Even though, it is negligible when conductance is studied, in the case of the thermopower it has turned out to be crucial. Therefore this modification to the BTK approach would complete the picture of the thermoelectric effect in planar NS junctions. The effect of the DOS in the similar manner was also considered earlier by Mazin [38] in the context of measuring the spin polarization of the ferromagnet in the ferromagnet–superconductor junction, and by Kupka [15] in the context of realistic three-dimensional geometry of NS junction.

Here we present the general framework which allows to obtain a nonzero Seebeck effect within the extended BTK approach, and applied to the simplest case of the normal metal–s-wave superconductor junction. Namely, we include explicitly the effect of the non-constant DOS around the Fermi level and apply it to the following selected situations: 3D free electron gas and 3D Fermi liquid. This inclusion gives rise to the nonzero thermoelectric effect, and results in large thermopower value across the junction. Particular set of parameters, leading to the large value of the thermopower, can possibly be realized in heavy fermion systems, where a small Fermi velocity implies a large value of dimensionless parameter modeling the barrier strength. Additionally, we have also discussed the effect of the logarithmic Van Hove singularity (VHS) present in the vicinity of the Fermi energy that gives possibility to study the thermopower also in the case of negative slope of DOS.

The paper is organized as follows. In Sect. 2 we discuss BTK theory with an explicit inclusion of energy dependent DOS, as well as refer to the previous works on the thermoelectric effect in NS junction. In Sect. 3 we derive the modified BTK formula for the case of 3D free electron gas and present results concerning the thermopower, both in the linear and the nonlinear regimes. Additionally we apply this approach to the Fermi liquid case. In Sect. 4 we extend our analysis to the case of DOS with VHS represented by a logarithmic function. Finally, in Sect. 5, we provide a brief summary.

2. Model and Approach

2.1. Modified Blonder–Tinkham–Klapwijk formalism

The BTK theory [5] describes electrical current through the NS junction. The starting point are the Bogoliubov–de Gennes equations [39] describing a two-component wave function $\begin{pmatrix} u \\ v \end{pmatrix}$ for s-wave superconductor with a constant superconducting gap Δ:

$$\begin{pmatrix} \mathcal{H} & \Delta \\ \Delta^* & -\mathcal{H}^\dagger \end{pmatrix} \begin{pmatrix} u \\ v \end{pmatrix} = E \begin{pmatrix} u \\ v \end{pmatrix}, \tag{1}$$

where the Hamiltonian for free electron gas reads $\mathcal{H} = -\frac{\hbar^2\nabla^2}{2m} - \mu(x) + V(x)$. The potential, utilized in the form $V(x) = K\delta(x)$, models the interfacial scattering. For convenience we also introduce dimensionless barrier strength $Z = K/\hbar v_{\mathrm{F}}$. In BTK formalism, one considers incident electron which is scattered by different processes at the interface with a finite transparency: it can be reflected, reflected as a hole (Andreev reflection — AR) or transmitted as a quasiparticle. For the states taking part in the tunneling processes, employing plane wave approximation, the corresponding multicomponent wave function is constructed. For simplicity we assume that all of particles have the same value of the momentum equal to the Fermi momentum. Applying the standard boundary conditions, i.e., by demanding continuity of the wave functions and their spatial derivatives at the interface, one can derive the probabilities of all the processes. Using those probabilities, following BTK, the net current formula is obtained in the following form [5]:

$$\begin{aligned} I_{\mathrm{NS}} = 2N(0)ev_{\mathrm{F}}\mathcal{A}\int_{-\infty}^{\infty} \Big\{ & [1 - B(E)]f^{\mathrm{N}}(E - eV) \\ & - A(E)f^{\mathrm{N}}(E + eV) \\ & - \big[1 - A(E) - B(E)\big] f^{\mathrm{S}}(E) \Big\} \mathrm{d}E, \end{aligned} \tag{2}$$

where $N(0)$ denotes DOS at the Fermi level, v_{F} is the Fermi velocity, $\mathcal{A}$ is the area of contact, and f^{N}, f^{S} are the Fermi distribution functions for the NC and SC, respectively. Functions $A(E)$ and $B(E)$ are the tunneling probabilities of the hole (Andreev) reflection and the usual reflection of the incident electron, respectively. For simplicity, we also define the transfer probability

$$\begin{aligned} T(E) \equiv\ & (1 - B)f^{\mathrm{N}}(E - eV) - Af^{\mathrm{N}}(E + eV) \\ & - (1 - A - B)f^{\mathrm{S}}(E). \end{aligned} \tag{3}$$

BTK approach has been generalized to realistic 3-dimensional geometry, where $T(E)$ depends also on the angle of incidence, as shown in a number of papers [14, 15]. However, the main contribution to the current is provided for angles of incidence close to zero, effectively similar to one-dimensional geometry (for justification, see Ref. [15] and references therein).

In the one-dimensional case, the product of the DOS ($\sim \partial k/\partial E_k$) and the charge carriers velocity ($\sim \partial E_k/\partial k$) is constant. In more general case, this assumption is well justified for calculating current–voltage characteristics, but it is insufficient for more subtle effects such as the thermoelectricity considered here. Also, it is important to include the effect of energy-dependent DOS in the study of spin-polarization of the ferromagnet in the case of the ferromagnet–superconductor junction [38].

In BTK approach [5], electrons contributing to the current are only those whose velocities are directed towards the junction i.e. those with $v_x > 0$. The current (2) through junction of area $\mathcal{A}$ can be rewritten in the spirit of the Landauer–Büttiker approach to the electronic transport as

$$I_{\rm NS} = e\mathcal{A}\sum_{k,\sigma} v_x T(E_k), \tag{4}$$

with $T(E_k)$ given by (3). In higher-dimension geometries, the energy dependence of DOS does not cancel out with that of the group velocity. In those cases, we have to take into account explicitly the product of DOS and velocity in the net current formula, i.e., start from the formula

$$I_{\rm NS} = 2e\mathcal{A}\int_0^\infty {\rm d}E T(E) \oint_{v_x>0} \frac{{\rm d}S(E)}{|\nabla_k E|} v_x. \tag{5}$$

In this formula we treat DOS rather as a "number of conductance channels". This concept is based on the assumption of not taking into account the fact that the hole retracing electron path is on the opposite side of the Fermi surface and therefore, feels different DOS. We also neglect the shift of DOS due to non-zero voltage over the junction, since it rather weakly influences the current characteristics. The resulting integral over the equi-energy surfaces can be solved by introducing the explicit energy dependence. This simple effect, in turn modifies the final BTK formula. In Sect. 3 we present our calculations of the thermopower taking into account the parabolic dispersion relation in the 3D free electron gas and linear dispersion relation around the Fermi energy for 3D Fermi liquid [40].

2.2. *Thermopower definition*

As said above, in the standard BTK approach there is no nonzero thermopower due to the symmetry in expression (2) irrespectively of the temperature difference. However, the assumptions introduced in more specific models give rise to the non-zero thermopower in NS junctions. The only works exploring thermoelectric properties of the NS junction via BTK formalism is that by Hirsch [12, 37]. He explored thermoelectric phenomena by including the energy dependent superconducting gap function [12] observed in the hole-superconductivity model, as well as by considering a more realistic rectangular barrier of finite width [37]. The latter case was also investigated in the case of ferromagnet–superconductor Andreev point contacts [41]. Here, the source of the thermoelectric effect in the NS junction is the energy dependence of v_x and of DOS, as expressed in (5). This feature does not alter much the conductivity, but gives rise to the thermoelectric phenomena.

By keeping the two electrodes at different temperature or applying voltage, we obtain a non-zero current through the junction. If both the voltage V and the temperature difference δT are small, we can write that the total current as

$$I_{\rm e} = L_{11} V + L_{12}\delta T. \tag{6}$$

The quantity that measures the thermoelectric effect is the thermopower or the Seebeck coefficient S, which is defined as the voltage driving to zero the current flowing in response to the temperature difference. This means that

$$S \equiv -\left(\frac{V}{\delta T}\right)_{\rm I=0} = \frac{L_{12}}{L_{11}}. \tag{7}$$

3. 3D Free-electron-gas case

For 3D free electron gas, the integral in (5) can be carried out analytically and is equal to

$$\oint_{v_x>0} \frac{{\rm d}S(E)}{|\nabla_k E|} v_x = \frac{k^2}{\hbar}\int_{\frac{-\pi}{2}}^{\frac{\pi}{2}} {\rm d}\theta \int_{\frac{-\pi}{2}}^{\frac{\pi}{2}} {\rm d}\phi \cos^2(\theta)\cos(\phi)$$
$$= \frac{2m\pi E}{\hbar^3}. \tag{8}$$

We have obtained linear energy dependence, which in turns modifies the BTK net-current formula in the following manner:

$$I_{\rm NS} = \frac{1+Z^2}{eE_{\rm F}R_{\rm N}}\int_{-E_{\rm F}}^{\infty} (E+E_{\rm F}) T(E)\,{\rm d}E. \tag{9}$$

As in the BTK papers [5], we have set the chemical potential equal to zero and thus, in the final result have shifted energies by the Fermi energy. Also, the current is normalized by the resistance of the NN junction [5] ($R_{\rm N}$). Let us note that in all numerical calculations discussed in the next sections we have taken the Fermi energy equal to $E_{\rm F} = 150\Delta$.

3.1. *Linear regime*

In Fig. 1 we draw schematically the NS junction. The black rectangle represents the barrier and the gray regions in it mark the regime of energies, for which the probability $A(E)$ is large ≈ 1. The dashed line show the chemical potential, taken as zero of energy. The bias is applied to the normal metal only. The black lines are the Fermi distributions on the cold and the warm sides of the junction. δT is the temperature difference across the junction.

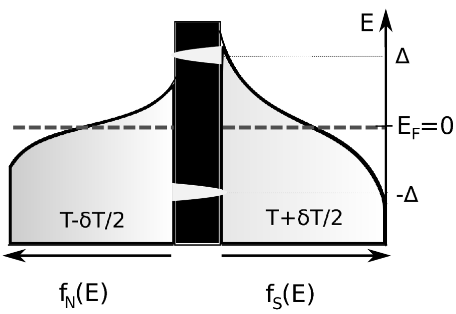

Fig. 1. Schematic diagram showing energetic properties of the NS junction in the limit of strong barrier, where the NC is colder than SC. The black rectangle represents the barrier. Heavy black lines show the Fermi functions. Dashed line denotes (common) Fermi energy, and thin lines on the right indicate positions of energy gap ($\pm\Delta$).

In the linear regime and very small temperature difference and voltage, δT, $V \to 0$, we expand the Fermi functions in the Taylor series, which to the first order are

$$f^{\mathrm{N}} = f_{T-\delta T/2}(E - eV) = f_T(E) - eV\frac{\partial f}{\partial E} + \frac{\delta T}{2T}E\frac{\partial f}{\partial E},$$

$$f^{\mathrm{S}} = f_{T+\delta T/2}(E) = f_T(E) - \frac{\delta T}{2T}E\frac{\partial f}{\partial E}. \tag{10}$$

Substituting those forms to Eq. (5) we obtain the explicit formula for the thermopower

$$S = \frac{\int \frac{\partial f}{\partial E}E(E+E_{\mathrm{F}})(1-B-A)\,\mathrm{d}E}{k_{\mathrm{B}}T\int \frac{\partial f}{\partial E}(E+E_{\mathrm{F}})(1-B+A)\,\mathrm{d}E}\frac{k_{\mathrm{B}}}{e}. \tag{11}$$

In Fig. 2 we present temperature dependence of the thermopower in the linear regime for selected values of the barrier strength, Z. The tendency is similar to that obtained by Hirsch [12]. Namely, with the increase of the barrier strength the thermopower increases. Also, for sufficiently high barrier strengths we have obtained a clear maximum of the thermopower as a function of temperature. The temperature dependence remains similar in the nonlinear regime (for larger temperature difference, δT), with slightly changed position of the thermopower maximum. What is worth mentioning the character of the thermopower temperature dependence for larger barrier strengths qualitatively agrees with the corresponding thermopower dependence calculated within the Usadel-equation framework [27].

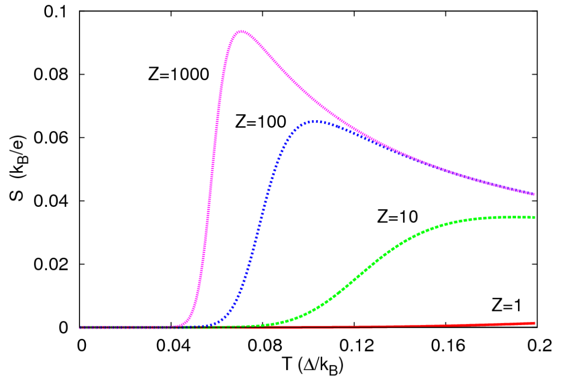

Fig. 2. Temperature dependence of the linear thermopower for selected barrier strength values, Z. The case of 3D free electron gas is considered.

The relevant energies of the junction are schematically presented as the diagram in Fig. 1. Probability of AR, $A(E)$, is equal to 1 for energies $\pm\Delta$, irrespectively of the value of the barrier strength [5]. Therefore, in the large barrier strength limit, $Z \to \infty$, the transport is almost blocked for all energies except for those close to $\pm\Delta$. Due to this circumstance, we obtain practically zero thermopower for temperatures less than $T = 0.05\Delta/k_{\mathrm{B}}$. In this limit, thermal excitations do not reach the upper channel of transmission around $+\Delta$ and there are fully occupied states on both sides of the junction for $E = -\Delta$. Once the temperature crosses this limit, we reach a maximum for the thermopower and subsequently the part T^{-1} starts dominating for all barrier strengths. The highest temperature shown in Fig. 2 is $T = 0.2\Delta/k_{\mathrm{B}}$, as we do not take into account the temperature dependence of the superconducting gap.

3.2. Nonlinear regime

In this section we present numerical results in the nonlinear regime. The thermopower depends on the barrier strength Z, temperature T, and the temperature difference δT. We have obtained asymmetric behavior of the thermopower (Fig. 3b–d) as a function of temperature difference δT, despite of the symmetric behavior of the zero-voltage current (Fig. 3a). For the positive sign of the temperature difference, we have obtained large values of the thermopower (up to $8k_{\mathrm{B}}/e$) in spite of a relatively small currents.

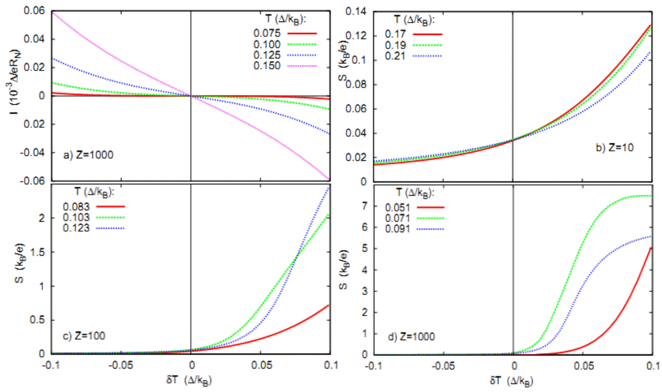

Fig. 3. Nonlinear zero-voltage current as a function of temperature difference, δT for selected values of the average temperature (T) in the system, for the barrier strength parameter $Z = 1000$ (part (a)) and nonlinear thermopower as a function of the temperature difference (δT) for selected temperatures (T) of the junction (around the temperature of the maximal linear thermopower, cf. Fig. 2) and selected values of barrier strength parameter $Z = 10, 100, 1000$ (parts (b), (c), and (d), respectively). 3D free electron gas is considered.

The asymmetry can be understood via a simple picture (cf. Fig. 1). For any temperature difference when at least one of the temperatures is high enough for thermal excitations to reach the transmission channel ($+\Delta$), a relatively large current is flowing through the junction. Consider the temperature difference of a positive sign, so the thermal smearing in SC is higher than in NC, such as thermal excitations in the former reach the energy equal to Δ. The voltage, applied to NC in order to have zero net current, shifts the Fermi energy in the NC with respect to the Fermi energy in the SC.

Large voltages are required to compensate for thermally induced currents. Therefore, we have a relatively high thermopower. The opposite happens for the negative sign of the temperature difference. In that situation even small shift of the Fermi energy in the NC results in the large change of the flowing current, and one gets small thermopower.

In a similar manner, we may argue about the behavior of the thermopower as a function of the barrier strength, observed clearly in the linear regime. With the increasing parameter Z, the main transmission channels around energies $\pm\Delta$ are narrower. For that reason, we have to shift the Fermi level by a larger voltage to have zero net current over the junction.

3.3. 3D Fermi-liquid case

It was shown first by Landau [42] that a more accurate description of electrons in metals is the theory of the Fermi liquid taking into account interaction between particles near the Fermi energy. This theory assumes linear dispersion relation in the vicinity of the Fermi level, i.e.

$$E_k - E_{\mathrm{F}} = \hbar v_{\mathrm{F}}(k - k_{\mathrm{F}}), \tag{12}$$

with the Fermi velocity $v_{\mathrm{F}} = \hbar k_{\mathrm{F}}/m^*$, where k_{F} is the Fermi momentum and m^* the enhanced effective mass. Since the Fermi liquid is considered to be more realistic description of electrons in metals, we have calculated analytically the product of the DOS and velocity within this model in three dimensions

$$\oint_{v_x>0} \frac{\mathrm{d}S(E)}{|\nabla_k E|} v_x = \frac{k^2}{\hbar} \int_{\frac{-\pi}{2}}^{\frac{\pi}{2}} \mathrm{d}\theta \int_{\frac{-\pi}{2}}^{\frac{\pi}{2}} \mathrm{d}\phi \cos^2(\theta)\cos(\phi)$$
$$= \frac{(E + E_{\mathrm{F}})^2 \pi}{\hbar^3 v_{\mathrm{F}}^2}. \tag{13}$$

The energy dependence is explicitly quadratic and different from the corresponding linear dependence for 3D free electron gas, cf. Eq. (8). The numerical results, however, both in linear and nonlinear regime differ only slightly from those shown for a 3D free electron gas and we do not present them here.

4. Effect of the Van Hove singularity

In Sects. 3 and 4 we have presented a consistent argument for the importance of the DOS shape on the thermopower in the 3D free electron gas, and in the 3D Fermi liquid. For the sake of completeness, we have investigated the thermopower within the model, in which we include in the integrand (5) the form of DOS with Van Hove singularity (VHS). We model the VHS by logarithmic function of energy with fitting parameters taken from the two-dimensional tight-binding model [43], namely:

$$N(E) = b_1 \ln\left|\frac{E}{b_2}\right|, \tag{14}$$

with $b_1 = -0.04687|t|^{-1}$, and $b_2 = 21.17796|t|$, where t is hopping, taken as $|t| = 100\Delta$.

This model corresponds to the assumption of constant velocity and realistic DOS. In the linear regime, thermopower can be expressed as

$$S = \frac{\int \frac{\partial f}{\partial E} E\left(\ln|\frac{E-\alpha}{b_2}|\right)(1 - B - A)\,\mathrm{d}E}{k_{\mathrm{B}}T \int \frac{\partial f}{\partial E}\left(\ln|\frac{E-\alpha}{b_2}|\right)(1 - B + A)\,\mathrm{d}E} \frac{k_{\mathrm{B}}}{e}, \tag{15}$$

where α denotes a relative shift (chosen arbitrarily) of the Fermi energy with respect to VHS. There were experiments in which the Fermi energy was found in a very close vicinity of the VHS [44, 45], or moved appreciably relative to it.

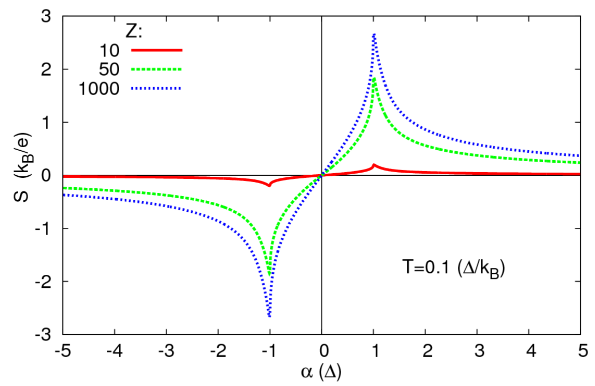

Fig. 4. The dependence of the linear thermopower on the relative shift of the Fermi energy with respect to Van Hove singularity α for a few values of barrier strength parameter Z. The average temperature in the system is taken as $T = 0.1\Delta/k_{\mathrm{B}}$.

The results of the linear thermopower as a function of parameter α for different values of barrier strengths for the fixed temperature $T = 0.1\Delta/k_{\mathrm{B}}$, are presented in Fig. 4. The maximum of the thermopower is well pronounced and fixed to the particular value of the shift of the Fermi energy with respect to the VHS, $\alpha = \Delta$. Therefore, we have studied the linear temperature dependence of the thermopower and nonlinear effects, particularly for $\alpha = \Delta$. The behavior of temperature dependence of the linear thermopower (not shown) is similar as that for 3D gas and for the Fermi liquid, but the values are larger than in the previous cases. The interesting feature of this model is that when the Fermi energy crosses the VHS, thermopower changes sign. This behavior of the thermopower is very different from that found by Hirsch who postulated universality of the thermopower sign for the hole theory of SC [12]. This observation seems to be very promising for the future experimental verification.

5. Conclusions

In this paper we have extended the Blonder, Tinkham, Klapwijk approach [5] to the tunneling spectroscopy between normal metal and superconducting materials of s-wave type. Our contribution is to go beyond the approximation of the constant product of the density of states (DOS) and the group velocity of charge carriers. Inspired by the papers of Mazin [38] and Kupka [15] we have introduced to the BTK current formula an explicit energy dependence of the product of DOS and the velocity. For the case of 3D free-electron gas, the product was found to be linear in energy, whereas within the Fermi liquid theory, quadratic. This modification virtually does not affect the current–voltage characteristics or conductance of the NS junction, but it gives rise to the thermoelectric effect. Previously only Hirsch [12, 37] examined the thermopower in NS junction via BTK approach in special cases by introducing either the energy-dependent superconducting gap function [12] (in the so-called model of the hole superconductivity) or by invoking the barrier with finite width [37].

Both linear and nonlinear regimes of thermopower were studied for the cases of free electron gas, the Fermi liquid, and for the model with the logarithmic Van Hove singularity. With the increase of the barrier strength the thermopower increases. We believe that heavy fermion systems are promising for the experimental verification of our theoretical predictions, since low values of the Fermi velocity should result in high value of the effective barrier strength.

We have found a maximum of the thermopower as a function of average junction temperature (Fig. 2). Behavior is qualitatively similar to that obtained via the Usadel–Eilenberger equations [27]. In the nonlinear regime, we have investigated additionally its dependence on the temperature difference, where we have found a spectacular asymmetry of the thermopower with respect to δT sign inversion (Fig. 3b–d), irrespectively of the fact that the zero-voltage current is completely symmetric (Fig. 3a). This can be attributed to the asymmetry of the system, where by applying the voltage one shifts the Fermi energy of the normal conductor with respect to the superconducting gap in the superconductor.

The approach can be applied also for the studies of 2D systems. The integral in (5) leads to either $\sqrt{E}$ or E dependence of the velocity times DOS function for 2D free electrons and the Landau–Fermi liquid, respectively.

Studies of the thermopower in the case of DOS with Van Hove singularity intuitively shows that the thermopower has different sign on the negative and positive slope of DOS (Fig. 4). Similar behavior of the thermopower can be expected for the Kondo insulators. In those systems one observes a sharp minimum in the DOS at the Fermi level which is sometimes considered to be a small semiconducting-like gap between the bands. Let us note that BTK theory predicts extremely large values of the barrier strength parameter Z in heavy fermion systems resulting from the small value of the Fermi velocity (this is also the case for large differences between the Fermi velocities on the both sides of the junction interface [1]). However, this would require an extension of the present approach to the situation with strongly correlated electrons [46–48].

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