Revision of the quasiclassical boundary resistance of the metal/superconductor interface

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Transparency of the metal/superconductor interface is discussed in a frame of the Kupriyanov-Lukichev (K-L) quasiclassical boundary conditions. It is shown that the original K-L boundary transparency is expressed via the single-channel Landauer conductance. It is a source of dramatic discrepancy between the theory and experiments on the superconducting proximity effect. The improved multi-channel derivation is proposed for the boundary conditions, which rehabilitates consistency of the quasiclassical approach to the boundary transparency problem, and eliminates at least an order of magnitude of discrepancy between the theory and the experiment. The influence of the interface roughness and wave function symmetry mismatch at the boundary on the interface transparency is discussed. Warning is made to theories that use the quasiclassical boundary conditions with the interface transparency parameters close to unity.

Since works by Eilenberger, Larkin and Ovchinnikov, and Usadel, the quasiclassical theory is main calculation approach in physics of superconductivity. Most of calculations utilize the Usadel theory with the Kupriyanov-Lukichev (K-L) boundary conditions because the short mean-free path approximation matches most experiments with conventional superconductors (note that the short coherence-length, high-temperature superconductors refer mainly to a clean-limit case). As long as the boundary resistance, which enters the K-L boundary conditions, is considered as a phenomenological fitting parameter, the Usadel theory gives adequate description of contact phenomena that has been demonstrated, for example, for the Josephson effect by Golubov et al and for the proximity effect by Buzdin. However, it was pointed out by the Salerno group that problems immediately arise when one tries to reproduce the best-fit boundary resistance (or transparency of the interface), obtained from proximity-effect experiments, making use of the physical parameters of materials in the contact. The authors employed the band offset (Fermi momenta mismatch) model for the quantum-mechanical transmission coefficient,

\[ D = \frac{4v_{1z}v_{2x}}{(v_{1z} + v_{2x})^2}, \]  

which is plausible approximation as far as the free-electron model is used as a background for the quasiclassical theory (\( v_{1z} \) is a projection of the electron Fermi velocity of the \( i \)-th metal on the direction perpendicular to the interface). The quantum-mechanical transmission coefficient \( D \) is related to the boundary resistance \( R_b \) via parameter \( \gamma_b \) as follows

\[ \gamma_b = \frac{\sigma_2 AR_b}{\xi_2} = \frac{2l_2}{3\xi_2} \left( \frac{x_2D}{1-D} \right)^{-1}, \]  

where \( A \) is an area of the contact, \( \sigma_2 = e^2p_2^2l_2/3\pi^2\hbar^3 \) is the normal-phase conductivity, \( \xi_2 = (hD^2/2\pi T_c)^{1/2} \) is the coherence length, \( l_2 \) and \( p_2 \) are the mean free path and the Fermi momentum of electrons, \( D_2 = v_2l_2/3 \) is the conduction electron diffusion coefficient, \( T_c \) is the superconducting transition temperature, and, finally, \( x_2 = \cos\theta_2 \), where \( \theta_2 \) is the quasiparticle incidence angle. The subscript “2” refers to a metal #2. The parameter \( \gamma_b \) varies between zero (virtual interface between identical metals) and physical infinity (an ultimate case of highly resistive insulating barrier at the interface). Following Aarts et al, the Salerno group characterized the interface by the transparency parameter,

\[ T = \frac{1}{1 + \gamma_b}, \]

which varies between 0 and 1. From the fitting of their data on the proximity effect in Nb/Pd system, the authors obtained \( T_{\text{exp}}(\text{Nb/Pd}) \approx 0.46 \), whereas their estimation from Eqs. (1) and (2) gives \( T_{\text{theor}}(\text{Nb/Pd}) \approx 0.98 \) using the Fermi velocities \( v_{\text{Nb}} = 2.73 \times 10^7 \, \text{cm/s} \) and \( v_{\text{Pd}} = 2.00 \times 10^7 \, \text{cm/s} \). For Nb/Cu and Nb/Ag couples the Salerno group obtained \( T_{\text{exp}}(\text{Nb/Cu}) \approx 0.30 \) as compared with \( T_{\text{theor}}(\text{Nb/Cu}) \approx 0.5 \) \( (v_{\text{Cu}} = 1.57 \times 10^8 \, \text{cm/s}) \); and \( T_{\text{exp}}(\text{Nb/Ag}) \approx 0.33 \) as compared with \( T_{\text{theor}}(\text{Nb/Ag}) \approx 0.55 \) \( (v_{\text{Ag}} = 1.39 \times 10^8 \, \text{cm/s}) \). Similar low interface transparencies have been derived from proximity experiments on Nb/Al couples: \( T_{\text{exp}}(\text{Nb/Al}) \approx 0.2-0.25 \) \( (T_{\text{theor}} \approx 0.8) \) and Nb/CuNi couple: \( T_{\text{exp}}(\text{Nb/CuNi}) \approx 0.2-0.35 \). One may derive from the above data that there is systematic and drastic discrepancy between the experimentally established transparency of the S/N interface and the quasiclassical theory which is applied to reproduce the transparency data. The discrepancy looks even more dramatic if we recognize that one needs unrealistic difference in the free-electron Fermi energies, one hundred times and more, to reproduce the boundary resistance, say, Nb/Cu couple. The interface roughness or mismatch of the wave functions symmetry at the interface can not compensate so dra-
tic misfit between the experiment and the theory. Then, one may come to a conclusion that there is global inconsistency in the quasiclassical approach when applied to description of the contact phenomena.

In this paper we show that source of the discrepancy is a very approximate treatment of the boundary resistance, which has been made upon derivation of the K-L boundary conditions. We develop further the K-L approach and propose new derivation of the boundary conditions for the Usadel equations which relaxes mainly the above mentioned discrepancy. We argue that approximation of highly transparent interface (the quasiclassical superconducting pairing function is continuous across the interface), which was oftenly used in calculations of the contact phenomena, is hardly (if ever possible) to realize in an experiment with conventional superconductors.

We start from the Zaitsev boundary conditions for the Eilenberger equations following Kupriyanov and Lukichev:

\[ g_{a1} = g_{a2} = g_{a}, \]  

\[ g_{a} \left\{ R \left(1 - g_{a}g_{a}\right) + \frac{D}{4} \left(g_{a1} - g_{a2}\right)^{2} \right\} = \frac{D}{4} \left(g_{a1} - g_{a2}\right) \left(g_{a1} + g_{a2}\right), \]  

where \( g_{a(i)} \) are the symmetric (antisymmetric) Green functions, and \( R = 1 - D \) is the reflection coefficient. The boundary condition ensures continuity of the charge current at the interface, while the second one, Eq. 5, relates the quasiparticles flux to a drop of their density at the interface, and depends on transparency of the interface. Using the explicit matrix structure of the Eilenberger functions close to the transition temperature \( T_{c} \), we arrive at the basic for our discussion, linearized Zaitsev boundary condition

\[ 2Rf_{a} = D \left(f_{a2} - f_{a1}\right), \]  

where \( f_{a} \) is the anomalous pairing Green function, which describes superconductivity in the system. We have to notice here that equation has been derived without any approximation concerning transparency of the interface. To proceed further we need also the linearized Eilenberger equations for the normal metal which read:

\[ l \cos \theta_{2} \frac{df_{a2}}{dz} + f_{a2} = 0, \]  

\[ l \cos \theta_{2} \frac{df_{a2}}{dz} + f_{a2} = \langle f_{a2} \rangle, \]

where the angular brackets mean averaging over the solid angle. The original logics of K-L derivation is as follows (see also review, Ch. 4): using a constraint, \( \langle \cos \theta f_{a} \rangle = \text{const.}(z) \), obtained after the solid-angle averaging of equation 8, and the antisymmetric function \( f_{a2} \approx -l_{2} \cos \theta_{2} (dF_{2}/dz) \), found from Eq. 7 at \( z \gg l_{2} \) in the lowest order on anisotropy, we obtain from Eq. 4 the first linearized boundary condition at \( z = 0 \),

\[ l_{1}p^{2}_{f1} \frac{df_{1}}{dz} = l_{2}p^{2}_{f2} \frac{df_{2}}{dz}, \]

where \( F_{i} = \langle f_{i} \rangle \) is the isotropic anomalous Usadel function.

To derive the second boundary condition we divide at first both sides of equation \( 9 \) by \( 2R \):

\[ f_{a} = \frac{D}{2R} (f_{a2} - f_{a1}). \]

Then, we substitute the approximate antisymmetric function, utilized above, for the left-hand side, replace the symmetric Eilenberger functions in the right-hand side by their Usadel counterparts, \( f_{si} \rightarrow F_{si} \), multiply \( 10 \) by \( \cos \theta_{2} \) and average over the angle theta. This procedure yields the second linearized boundary condition,

\[ \xi_{2}g_{b} \frac{df_{2}}{dz} = F_{1} - F_{2}, \]

with \( \gamma_{b} \) given by Eq. 2. Equations 9 and 11 are well known linearized Kupriyanov-Lukichev boundary conditions for the Usadel functions. It has been shown by Lambert et al. that they are valid only at low transparencies of the S/N interface. Why does it so despite of the generic Zaitsev BC are valid for arbitrary transparency?

To answer the question let us have a look at the physical meaning of the linearized Zaitsev BC which is parental for the K-L one, Eq. 11. It has a sense of the boundary condition for a particular trajectory determined by the angles \( \varphi \) and \( \theta \). The language of trajectories is suitable when there is a continuous domain of quasiparticle incidence angles which accommodates macroscopic number of trajectories. In a case of a very few traversing trajectories it is more suitable to use a language of quantum-mechanical conductance channels. Then, the single-channel Landauer conductance (in units of the conductance quantum \( e^{2}/\pi h \)) can be immediately recognized in the ratio \( D/R \) which enters the right-hand side of equation 10. Subsequent derivation procedure involves integration over the incidence angles of the single-channel Landauer conductance. Implication of the single-channel conductance to BC is crucial for understanding the source of insufficiency of the original K-L approximation reproduced above.

Indeed, the integrated single-channel Landauer conductance matches physical expectation, that conductance of the interface is zero when the transparency coefficient \( D \) is zero (boundary resistivity is infinite). Alternatively, the conductance is infinite (boundary resistivity
is zero) when the contacting materials are identical, and the interface is virtual. On the other hand, practically in all experiments with superconducting heterocontacts we have many channels for conductance through the interface. From the physical point of view one may expect that the multi-channel conductance should enter the correct formulation of the boundary condition. Then, we have to answer on a next question: is the angle-integrated, single-channel Landauer conductance represent correctly the multi-channel conductance? The answer “no” is given at the end of the Section II of the paper by Büttiker, Imry, Landauer and Pinhas (hereafter will be referred as BILP). In the Section IV of their paper the authors derive the multi-channel conductance through the interface, valid at arbitrary transparency:

\[
G = \frac{2e^2}{\pi h} \left( \sum_{i=1}^{N} D_i \right),
\]

where

\[
g_l = \sum_{i=1}^{N} (v_i^l)^{-1}, \quad g_r = \sum_{i=1}^{N'} (v_i^r)^{-1},
\]

and \(v_i\) are velocities of quasiparticles moving to the left (l) and to the right (r) over \(N\) and \(N'\) channels of conductance, respectively. For a flat interface of macroscopic area the summations over the conductance channels can be replaced by integration over the incidence angles: \(\sum_{i=1}^{N_a} \rightarrow \int d\varphi \alpha \sin \theta_a\), and the integration runs over the semi-sphere (SS) in the direction of the particles velocity. Dependence of the multi-channel conductance on the Fermi-energies of contacting metals and roughness of the interface between them has been analyzed in detail by García and Stoll.

To make evident relevance of a sheet conductance \(\sigma_{KL}\) (conductance per unit square) of an interface for formulation of BC we rewrite Eqs. (9) and (10) in terms of conserving fluxes:

\[
\sigma_1 \frac{dF_1}{dz} = \sigma_2 \frac{dF_2}{dz},
\]

(14)

\[
\sigma_2 \frac{dF_2}{dz} = \sigma_{KL} (F_1 - F_2),
\]

(15)

where

\[
\sigma_{KL} = \sigma_{KL} = \left( \frac{e^2}{h} \right) \left( \frac{p_F^2}{\pi^2 h^2} \right) \left( 2\pi \frac{\langle x_2 D \rangle}{2(1 - D)} \right)
\]

is nothing else but the sheet conductance of the interface in the Kupriyanov-Lukichev approximation. However, as we deduced from the above analysis, the ultimate formulation of BC has to be expressed via the multi-channel sheet conductance (at the current status, with the last parentheses in Eq. replaced by the parentheses from Eq. 12). Now, the main direction of logics is to find an analogue or a good approximation of the multichannel conductance remaining within the quasiclassical formalism.

As the first step let us look what will happen if we withdraw transformation of the original, particular trajectory Zaitsev BC into the single-channel BC form. Starting from Eq. (10) we replace: (1) the antisymmetric Eilenberger function \(f_a\) in the left-hand side by the approximate expression used upon derivation of Eq. (9); (2) the symmetric Eilenberger functions in the right-hand side by their Usadel counterparts. Then, we have in an output

\[
-2(1 - D)l_2 \cos \theta_2 \frac{dF_2}{dz} = D(F_1 - F_2).
\]

(17)

The procedure of the angular averaging described after Eq. (10) yields BC with the sheet conductance

\[
\sigma_{GL} = \left( \frac{e^2}{h} \right) \left( \frac{p_F^2}{\pi^2 h^2} \right) \left( 2\pi \frac{\langle x_2 D \rangle}{2(1 - 3(1/2D))} \right).
\]

(18)

The behavior of \(\sigma_{GL}^{GT1}\) on a ratio of the Fermi energies \(R_F = \epsilon_{F2}/\epsilon_{F1} \leq 1\) is given by the dash line in Fig. 1. It is easy to see by comparison with Eq. (10) that the main problem of K-L sheet conductance at high transparency of interface - the denominator, \(1 - D\), is close to zero in the main domain of integration angles - is relaxed already in Eq. (15). However, the first step does not take into account properly the influence of other conducting channels (or trajectories crossing the interface) on propagation of an electron along a particular trajectory.

When there is a single conductance channel through the interface, all other bulk conducting channels approaching the interface remain unperturbed. In our particular case of the S/N junction the superconducting order parameter parameter is flat near the impenetrable interface with the only an atomic-size hole which accommodates one conductance channel. In the case of the transparent interface, the other conductance channels induce mutual accomodation of the pairing function wave function and the order parameter from the both sides of the interface (see, for example, Fig. 3 of Ref. (2)) creating gradients. Thus, the next step of approaching the true multichannel formulation could be consideration of the Eilenberger function gradients.

Careful look at the derivation procedure shows that upon replacement of the symmetric Eilenberger functions in the right-hand side of (11) by their Usadel limits we have lost in Eq. (17) gradient terms of the same order that we have in the left-hand side of this equation. In the dirty limit, the electron mean free path is the shortest length compared with the superconducting or normal state coherence lengths. Then, in the spirit of Ref. (14),
we expand the Eilenberger functions $f_{si}$ in powers of the mean free path along the particular trajectory, keeping the gradient terms,

$$f_{si} \simeq \langle f_{si} \rangle + l_{z1} \frac{df_{si}}{dz} = F_i + l_{z1} \frac{dF_i}{dz},$$

and substitute them into the right-hand side of Eq. 10. After the angular averaging procedure we arrive at the second BC (15) with the sheet conductance as follows:

$$\sigma_{GT2} = \left( \frac{e^2}{h} \right) \left( \frac{\rho_2^2}{\pi^2 h^2} \right) \times \left( 2\pi \left( 1 - \frac{4}{\pi} \langle x_2 D \rangle - \frac{3}{2} \approx \frac{2}{\hbar} \langle x_1 x_2 D \rangle \right) \right).$$

Our sheet conductance (20) is the next step to approximate the multichannel BILP sheet conductance (12) in the gradient approximation. The two terms in the denominator of the big parentheses in (20) are one-by-one counterparts of the similar terms, which we will have in (12) after replacement of $R_i$ by $1 - D_i$ in the denominator. We believe (from considerations of energies involved in the transmission) that the result holds for the general, non-linear BC as well. Dependence of the sheet conductance (20) on $R_F$ is displayed by the thick solid line in Fig. 1. It can be seen that the GT2 curve goes much closer to the BILP sheet conductance (dotted curve) than our first approximation given by Eq. 13.

The boundary conditions (13) and (15) can be expressed via the experimentally measured bulk resistivities of the contacting metals, $\rho_1 = \sigma_1^{-1}$ and $\rho_2 = \sigma_2^{-1}$, and the sheet resistance of the boundary, $A_{R_b} = \sigma_\parallel^{-1}$.

We believe that our approach gives consistent quasiclassical approximation to the multichannel BILP sheet conductance of the interface. The improvement is drastic in the region of high transparencies: our sheet conductance $\sigma_{GT2}$ is more than order of magnitude lower that the K-L one. Correspondingly, our sheet resistance of the interface, $A_{R_b} = (\sigma_{GT2})^{-1}$, in the region of $R_F \lesssim 1$ is more than order of magnitude higher than the K-L one. There are also other physical reasons that may influence the boundary resistance. García and Stoll have shown (Fig. 2 of Ref. [18]) that the interface roughness increases boundary resistance, i.e. $\gamma_\parallel$. At the Fermi energy ratio $R_F = 0.4$ this increase is about 20-60% for different models of the interface roughness. The reduced overlapping and symmetry mismatch of the wave functions may also essentially suppress transparency of the interface. Other physical aspect is that only the propagating (dispersive) part $E_{prop}$ of the total electron energy, $E_{tot} = E_{prop} + E_{rot}$, computed in the free electron model, contributes to the transport across the interface. So the narrow $d$-electron dispersion band may effectively produce reduced $R_F$ ratios as soon as the localized rotational contribution $E_{prop}$ is subtracted from the total energy of the $d$-state.

In a consequent way, one may think that light metals, for which the approximation of free electrons is well justified, could be candidates for the very transparent interface between them. If aluminum is chosen as a superconductor ($\varepsilon_F(Al) = 11.7$ eV) then the series of noble metals as a counter-electrode (from $\varepsilon_F(Ag) = 5.5$ eV to $\varepsilon_F(Cu) = 7$ eV) corresponds to $R_F = 0.4 - 0.6$, which implies essentially reduced interface transparency. It seems that perfect interface transparency can not be realized in an experiment. In the view of the intrinsically reduced transparency of the interface between any two metals we conclude that our quasiclassical boundary resistance can be used in the full range of experimentally attainable interface transparencies. On the other hand, all theories which essentially exploit perfect transparency of the interface have to be re-examined to check survival of predicted effects against the reduced transparency of the interface.

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**Figure Captions**

Fig. 1. Dependence of the normalized interface conductances on the Fermi energy ratio $R_F$. The scale of the ordinate is logarithmic.
