Charge Transfer between a Superconductor and a Hopping Insulator

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We develop a theory of the low-temperature charge transfer between a superconductor and a hopping insulator. We show that the charge transfer is governed by the coherent two-electron–Cooper pair conversion process *time-reversal reflection*, where electrons tunnel into a superconductor from the localized states in the hopping insulator located near the interface, and calculate the corresponding interface resistance. A specific feature of this problem is the interplay between the time-reversal reflection at the interface and transport through the percolation cluster. To allow for this interplay, we have generalized the connectivity criterion of the percolation theory to include surface effects. We show that the time-reversal interface resistance is accessible experimentally, and that in mesoscopic structures it can exceed the bulk hopping resistance.

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The transmission of the charge through the normal metal-superconductor interface occurs via the electron-hole conversion known as the Andreev reflection process: An electron incident from the metal side with an energy smaller than the energy gap in the superconductor is converted into a hole which moves backward with respect to the electron. The missing charge $2e$ (an electron has charge $-e$ and a hole $+e$) propagates as an electron pair into the superconductor and joins the Cooper pair condensate [1]. Correspondingly, a Cooper pair transfer from the superconductor is described as the Andreev reflection of a hole. This Andreev transport channel is characterized by the so-called Andreev interface contact resistance. Since transport current is introduced into a superconductor via normal leads, the Andreev reflection phenomenon is a foundation for most applications of superconductors (see Ref. [2] for a review).

There exists, however, an important experimental situation of the hopping insulator (HI) coupled to a measuring circuit via superconducting leads (see, for example, [3]), where the conventional Andreev reflection picture does not apply. The transport in hopping semiconductors occurs via localized (*nonpropagating*) single-particle states [4] with undefined momentum, and, therefore, a Cooper pair on the superconductor side cannot form. A single-particle transport through the interface is exponentially suppressed, $\propto e^{-\Delta/T}$, where Δ is the superconductor gap, the temperature T being measured in energy units; therefore, to explain the finite conductivity observed in experiments, one needs Andreev-type processes capable to facilitate two particle transfer through the hopping insulator-superconductor interface allowing for Cooper pair formation. The possibility of such a transfer through the hopping-superconductor interface was discussed in Ref. [5], but no quantitative theory of hopping transport-supercurrent conversion was presented.

In this Letter, we develop a theory for the transport through the HI-superconductor interface and derive the corresponding contact resistance. We show that the low-temperature charge transfer occurs via the correlated processes mediated by the *pairs* of hopping centers located near the interface. This process is close to the conventional Andreev electron-to-hole reflection into a normal metal, the exponential suppression of transport specific to a single-particle processes being lifted. Thus, despite the limitation in the number of coherent hopping centers that can carry Andreev transport, the resulting contact resistance can become low as compared to the resistance of the hopping insulator. However, in mesoscopic structures, the interface resistance can be comparable to or even exceed the hopping resistance. The proposed mechanism resembles the so-called crossed Andreev charge transfer [6], discussed recently in connection with a superconductor-dot entangler [7,8]. The difference is that in Refs. [7,8] the transport mediated by artificial quantum dots was considered. In our case, the transport occurs via randomly located sites in the HI, and the main problem one has to solve is finding the optimal configuration of the sites responsible for the charge transfer. Hereafter, we will refer to the proposed charge transfer mechanism as to the *time-reversal reflection*.

Let a superconductor (S) and an HI to occupy the adjacent 3D semispaces separated by a tunneling barrier (B). The presence of the barrier simplifies calculations which will be made in the lowest nonvanishing approximation in the tunneling amplitude T_0. This models the Schottky barrier usually presenting at a semiconductor-metal interface. In the linear response theory, the conductance is determined by the Kubo formula [9] for the susceptibility,

$$\chi(\omega) = i \int_0^\infty \langle[\hat{I}^+(t), \hat{I}(0)]\rangle e^{i\omega t} dt, \qquad (1)$$

as $\mathcal{G} = \lim_{\omega\to 0}\omega^{-1}\,\mathrm{Im}\chi(\omega)$. Here $\hat{I}(t)$ is defined as [10]:

$$\hat{I}(t) = iedT_0 \int d^2r[a^+(\mathbf{r}, t)b(\mathbf{r}, t) - \mathrm{H.c.}],$$

where $\mathbf{r}$ is the coordinate in the interface plane, $a^+(\mathbf{r}, t)$ and $b(\mathbf{r}, t)$ are creation and annihilation operators in the semiconductor and superconductor, respectively, and d is the electron localization length under barrier. The susceptibility $\chi(\omega)$ is calculated by analytical continuation of the Matsubara susceptibility [11],

$$\chi_M(\Omega) = \int_0^\beta \langle T_\tau I(\tau)I(0)\rangle e^{i\Omega\tau}d\tau. \quad (2)$$

Here T_τ means ordering in the imaginary time, $\beta \equiv 1/T$. In the expression for $\langle T_\tau I(\tau)I(0)\rangle$, one should expand to the second order with respect to the tunneling Hamiltonian,

$$H_T(\tau) = dT_0 \int d^2r[a^+(\mathbf{r}, \tau)b(\mathbf{r}, \tau) + \mathrm{H.c.}]. \quad (3)$$

Keeping only those second order terms that contain $\langle T_\tau b(\mathbf{r}, \tau)b(\mathbf{r}_0, 0)\rangle\langle T_\tau b^+(\mathbf{r}_1, \tau_1)b^+(\mathbf{r}_2, \tau_2)\rangle$ products and, thus, represent the time-reversal scattering in which we are interested, one arrives at the expression

$$\langle T_\tau \hat{I}(\tau)\hat{I}(0)\rangle = e^2|T_0|^4 \int d\tau_1 d\tau_2 \prod_i d^2r_i(A + B);$$
$$A(\{x_i\}) = F(x - x_0)F^+(x_1 - x_2)G(x_1, x)G(x_2, x_0),$$
$$B(\{x_i\}) = F(x - x_1)F^+(x_0 - x_2) \quad (4)$$
$$\times [G(x_0, x)G(x_2, x_1) - G(x_0, x_1)G(x_2, x)],$$

where $x \equiv \{\mathbf{r}, \tau\}$, $x_0 \equiv \{\mathbf{r}_0, 0\}$, $x_i \equiv \{\mathbf{r}_i, \tau_i\}$; $F(x - x') = \langle T_\tau b(\mathbf{r}, \tau)b(\mathbf{r}', \tau')\rangle$ is the anomalous Green's function in the superconductor, while $G(x, x') = -\langle T_\tau a(\mathbf{r}, \tau)a^+ \times (\mathbf{r}', \tau')\rangle$ is the Green's function in the HI. One can show that the Andreev-type process we are interested in is given by the first term of $B(\{x_i\})$ in Eq. (4). The relevant diagram is shown in Fig. 1. This diagram is similar to that considered by Hekking and Nazarov [12] in connection with tunneling transport between a superconductor and normal metal. However, here the normal Green's functions correspond to localized states that lead to significantly different results.

Using the Matsubara frequency representation, one obtains

$$\chi_M(\Omega) = 2Te^2|T_0|^4d^4 \int \prod_i d^2r_i \sum_{\omega_n} F(\mathbf{r} - \mathbf{r}_1, \omega_n)$$
$$\times F^+(\mathbf{r}_0 - \mathbf{r}_2, \omega_n)G(\mathbf{r}_0, \mathbf{r}, \omega_n - \Omega_m)$$
$$\times G(\mathbf{r}_2, \mathbf{r}_1, -\omega_n),$$

where $\Omega_m = 2\pi mT$ and $\omega_n = (2n + 1)\pi T$. The normal Green's functions can be expressed through the wave functions of the localized states, $\varphi_s(\mathbf{r}) = (\pi a^3)^{-1/2} \times \exp(-|\mathbf{r} - \mathbf{r}_s|/a)$, as

$$G(\mathbf{r}, \mathbf{r}', \omega_n) = \sum_s \frac{\varphi_s^*(\mathbf{r})\varphi_s(\mathbf{r}')}{i\omega_n - \varepsilon_s}. \quad (5)$$

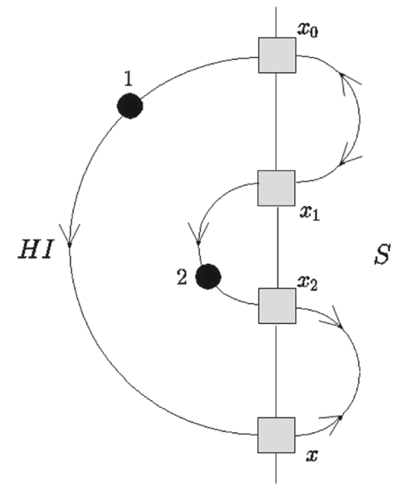

FIG. 1. The diagram describing the time-reversal reflection. Lines with one arrow correspond to the Green's functions in the HI. They are associated with either center 1 or center 2. Lines with two arrows correspond to anomalous Green's functions; see [11]. Squares correspond to matrix elements of the tunneling Hamiltonian (3).

We have assumed that, for all the sites under consideration, the voltage drops between the site and the superconductor are the same. This is true when the partial interface resistance due to a time-reversal pair is much larger than the typical resistance of the bond forming the percolation cluster. This situation resembles that considered by Larkin and Shklovskii for the tunnel resistance between the hopping conductors [13].

The anomalous Green's function $F(R, \omega_n)$ is

$$F(R, \omega_n) = \frac{\pi g_m \Delta}{2\sqrt{\Delta^2 + \omega_n^2}} \frac{\sin(Rk_F)}{Rk_F} e^{-R/\pi\xi\sqrt{\Delta^2 + w_n^2}/\Delta}. \quad (6)$$

Here $\xi_p = (p^2 - p_F^2)/2m$, $g_m = mp_F/\pi^2\hbar^3$ is the density of states in a metal, $k_F = p_F/\hbar$, while ξ is the coherence length in a superconductor. Since $F(R)$ oscillates with the period $2\pi/k_F$, integration over spatial coordinates along the interface yields the factor $a^4/k_F^6|\boldsymbol{\rho}_{ls}|^2$. Here $\boldsymbol{\rho}_{ls}$ is a projection of the vector $\mathbf{R}_{ls}$ connecting the centers on the interface plane. Note that the dependencies on R and ξ are similar to that given by Eq. (21) of Ref. [7] for the pair of the quantum dots near the superconducting interface. However, the latter equation does not specify the dependences of the transmission coefficients on the real physical parameters of the interface and the localized centers.

The summation over the Matsubara frequencies ω_n is standard,

$$T\sum_{\omega_n} f(\omega_n) = \oint \frac{d\varepsilon}{4\pi i} f(\varepsilon) \tanh\frac{\varepsilon}{2T}.$$

The contour of integration closes the cuts $|\varepsilon| > \Delta$ along the real axis. Upon analytical continuation, one arrives at the following expression for the conductance:

$$\mathcal{G} = \frac{\pi e^2 g_m^2 |T_0|^4 d^4}{2\hbar T k_F^6 a^2} \sum_{s \neq l} \frac{n(\varepsilon_s) n(\varepsilon_l) \Delta^2}{|\boldsymbol{\rho}_{ls}|^2 (\Delta^2 - \varepsilon_s^2)} e^{-2(z_s + z_l)/a} \times e^{-2|\boldsymbol{\rho}_{ls}|\sqrt{\Delta^2 - \varepsilon_s^2}/\pi\xi\Delta} \delta(\varepsilon_s + \varepsilon_l + U_c). \quad (7)$$

Here $n(\varepsilon) \equiv (e^{\varepsilon/T} + 1)^{-1}$ is the Fermi distribution, U_c is the energy of the intersite Coulomb repulsion, and the z axis is perpendicular to the interface. Equation (7) is based on the assumption that the energy conservation law can be satisfied. This is the case for macroscopic junctions with area $S \gg [ga\hbar\nu(T)]^{-1}$, where g is the effective density of states in the HI, while $\nu(T)$ is the typical hopping rate at temperature T. This inequality is fulfilled in macroscopic samples, although it can be violated in mesoscopic junctions or at very low temperatures. In that case, phonon-assisted processes may be important. For a macroscopic junction, $\sum_{l,s} \to g^2 \int d^3 r_l d^3 r_s d\varepsilon_l d\varepsilon_s$. Note that g is the density of states in the layer adjacent to the interface. Because of screening by the superconductor, it is not affected by the Coulomb gap and can be considered as constant. Since we are dealing with the pairs close to the interface, the Coulomb repulsion is suppressed by screening. This screening can be conveniently regarded as an interaction of the charged particle with its image having the opposite charge. Thus, the Coulomb correlations manifest themselves as the dipole-dipole interaction, and for $\rho_{sl} \gg a$ one arrives at $U_c = e^2 a^2/\kappa\rho_{sl}^3$. Requiring that $U_c < T$, one obtains a cutoff $\rho_{sl} \geq \rho_T \equiv a(e^2/\kappa a T)^{1/3}$. As a crude estimate, we take $d^4 \sim k_F^{-4}$, while $T_0 \approx T_p e^{-\Lambda}$, with $T_p \sim \varepsilon_F$. Bearing this in mind, one finds $g_m^2 T_p^2/k_F^6 \sim g_m^2 \varepsilon_F^2/k_F^6 \sim 1$. Since the ratio $T_p/(ak_F)^2$ is of the order of the typical energy of the localized state $\varepsilon_d \sim \hbar^2/ma^2$, one obtains

$$\frac{\mathcal{G}}{\mathcal{G}_n} \sim g a^3 \varepsilon_d \ln\left(\frac{\xi}{\rho_T}\right) e^{-2\Lambda}, \qquad \mathcal{G}_n \sim \frac{e^2}{\hbar} g a S \varepsilon_d e^{-2\Lambda}. \quad (8)$$

Here $\mathcal{G}_n$ is the conductance of a boundary between a normal metal and a HI, while S is the contact area. The product $gaS\varepsilon_d$ is nothing but the number of localized centers within the layer of a thickness a near the interface.

The above approach holds, as we have already mentioned, only if the resistance of the typical time-reversal resistor (TRR) is much larger then that of the critical hopping resistor $R_h = (h/e^2\gamma)e^{\zeta}$, where γ is a dimensionless factor depending on the mechanism of electron-phonon interaction and ζ is the hopping exponent [14], i.e., with the exponential accuracy, as long as $4\Lambda > \zeta$.

There are many realistic situations where the barrier strength Λ is not too large; the Schottky barrier at the natural interface [5] is certainly a case like that. Consequently, if $\zeta \gg 1$, i.e., if the system is far from the metal-to-insulator transition point, the procedure of summation over the localized states should be modified. Namely, the choice of the pairs facilitating the charge transfer depends on the structure of the bonds connecting critical pairs to the rest of percolation cluster.

According to the above considerations, the voltage drops mainly on the bond connecting percolation cluster in the HI with the critical TRR for which the distance between its pair components is less than the correlation length $\mathcal{L}$ of the backbone cluster. The incoherent electron transport can be ensured by a *single* bond connecting the cluster to any of the TRR sites. Thus, the ratio $\mathcal{G}/\mathcal{G}_n$ is the probability to find a TRR contacting the percolation cluster. To estimate this probability, we have to generalize the connectivity criterion of the percolation theory to allow for surface effects.

Let us consider the layer with the thickness of the typical hopping distance r_h near the interface where all the bonds of the backbone cluster necessarily have a site within this layer. The total number of states in this layer is $gSr_h\varepsilon_h$, where $\varepsilon_h = T\zeta$ is the width of the hopping band. This product can be estimated as $(\beta/8)(S/r_h)^2$, where β is a numerical constant [14]. For the case of Mott variable range hopping (VRH), $\beta \approx 20$.

The number of TRRs in this layer can be estimated as follows. Let us note first that the conserving energy $\varepsilon_s + \varepsilon_l + U_c$ from the δ function in Eq. (7) is associated with the band given by the broadening $\nu = \nu_0 \exp(-2r_d/a)$. Here r_d is distance to the nearest neighbor in HI. Indeed, the most natural source for the broadening of the resonance is coupling of the localized states. Second, since both electrons escape from the TRR through a single bond, the in-plane distance ρ_{sl} should not exceed the typical distance between the hopping sites $r_h = a\zeta/2 \ll \xi$. Keeping the exponential accuracy, we arrive at the following criterion that the resistance of TRR is less than the resistance of a typical hopping resistor:

$$4\Lambda + \ln\frac{T}{\nu_0} + \frac{\max(|\varepsilon_s|, |\varepsilon_l|)}{T} + \frac{2r_d}{a} + \frac{2(z_s + z_l)}{a} < \zeta. \quad (9)$$

One may consider this equation as a generalization of the "connectivity criterion" to include the TRR. Here we deal with the independent variables $\varepsilon_s, \varepsilon_l, r_d, z_s$, and z_l over which the averaging procedure should be done with an account of the restriction of Eq. (9). Thus, the number of the relevant TRRs is

$$8\pi^2 g^3 S \int d\varepsilon_l \int d\varepsilon_s \int_0^{\varepsilon_s} d\varepsilon_d \int r_d^2 dr_d \int dz_s dz_l \int_0^{r_h} \rho d\rho \times \Theta\left[\frac{\max(\varepsilon_s, \varepsilon_l)}{T} + \frac{2r_d}{a} + \frac{2(z_s + z_l)}{a} - \alpha\zeta\right], \quad (10)$$

where $\alpha \equiv 1 - [4\Lambda + \ln(T/\nu_0)]/\zeta < 1$. Let us now measure the energies in units of $\alpha\varepsilon_h$ and lengths in units of αr_h, where $\varepsilon_h \equiv T\zeta$. Again, the product $gr_h^3\varepsilon_h$ can be estimated as $\beta/8$, and we obtain the number of effective TRRs as $N_A \sim \mathcal{A}\alpha^7 S/r_h^2$, where

$$\mathcal{A} = 4\pi^2(\beta/8)^2 \int d\epsilon_l \int d\epsilon_s \int_0^{\epsilon_s} d\epsilon_d \int \eta_d^2 d\eta_d \int d\eta_s \int d\eta_l \times \Theta[\max(\epsilon_s, \epsilon_l) + (\eta_d + \eta_s + \eta_l) - 1] \approx 0.1. \quad (11)$$

Consequently, $\mathcal{G}/\mathcal{G}_n \sim \mathcal{A}\alpha^7 \ll 1$. One concludes that the difference between the "contact" resistances in normal and superconducting states is dominated by the contribution of TRRs. Since $\mathcal{G}_N \approx R_h^{-1}S/\mathcal{L}^2$,

$$\delta R \equiv \mathcal{G}_n^{-1} - \mathcal{G}^{-1} \approx -\mathcal{G}^{-1} = -R_h(\mathcal{L}^2/S\mathcal{A}\alpha^7). \quad (12)$$

Note that the interface resistance is of the order of the resistance of HI layer with the thickness $\sim\mathcal{L}/(\mathcal{A}\alpha^7) \gg \mathcal{L}$. Since $\mathcal{L} \sim a\zeta^2$, one concludes that for $\zeta > 10$ the interface resistance can be comparable to or even exceed the hopping resistance if the thickness of the sample (or of the contact) is $\lesssim 10\ \mu$m.

The resistance estimated above can be experimentally measured as a magnetoresistance in magnetic fields higher than the critical field for superconductivity (a similar effect for a quasiparticle channel was studied in Ref. [5]).

We were implicitly assuming so far that the variable range hopping occurs according to the Mott's law. This assumption certainly holds near the interface where the Coulomb gap is screened by a superconductor. However, in the bulk of HI, the Efros-Shklovskii (ES) law [14,15] can become the dominant hopping mechanism. Then the value of ζ in the connectivity criterion (9) will be controlled by the Coulomb gap $\zeta \to \zeta_{\rm ES} = (\beta_1 e^2/\kappa a T)^{1/2}$, where β_1 is a numerical constant [14,15]. In this case, it turns out that each bond of the ES backbone cluster finds some TRR ensuring charge transfer. Thus, in the limiting case of a weak tunneling interface barrier, the contact resistance will be the same for both normal metal and superconductor leads. This fact can be used to discriminate between Mott and ES laws in the situation when it is difficult to do so from temperature dependence.

Note that, in principle, the charge transfer involving double occupied localized states is possible. However, such a process would require an additional activation exponential factor $\propto e^{-U/T}$, where U is the on-site correlation energy. One can also consider processes where a double occupied center (so-called D^- center) serves as an intermediate state for the phonon-assisted two-electron tunneling. This channel can be neglected because (i) it involves an additional small preexponential factor due to phonon-assisted tunneling; (ii) for the corresponding 3-sites problem, one also has either the additional tunneling exponential $\propto e^{-4r_h/a}$ or a small probability to form a close triad of hopping sites.

To conclude, we have developed a theory of the low-temperature charge transfer between a superconductor and a HI and calculated the interface resistance. This resistance is dominated by time-reversal reflection processes involving localized states in the insulator. It is the time-reversal reflection process that allows the low-temperature measurements of hopping transport utilizing superconducting electrodes in the experimental setups. In the ES VRH regime, the corresponding interface resistance is small as compared to the bulk hopping resistance and is nearly equal to the resistance at the interface between the HI and normal metal. On the contrary, in the Mott hopping regime (relevant, in particular, for 2D gated structures), the interface resistance grows much larger and becomes commensurate (or even exceeds) to the bulk hopping resistance. This effect is especially pronounced in the mesoscopic samples. The contribution from the interface resistance can be detected by application of the external magnetic field: The relatively weak magnetic field will drive the superconductor into the normal state but will not affect the hopping transport, thus eliminating the time-reversal reflection process. This effect holds even in the case where the interface contribution is less than the typical resistance of the hopping system itself.

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