Crossed Andreev reflection and charge imbalance in diffusive NSN structures

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We formulate a microscopic theory of non-local electron transport in three-terminal diffusive normal-superconducting-normal (NSN) structures with arbitrary interface transmissions. At low energies ε we predict strong enhancement of non-local spectral conductance g_{12} ∝ 1/ε due to quantum interference of electrons in disordered N-terminals. In contrast, non-local resistance R_{12} remains smooth at small ε and, furthermore, is found to depend neither on parameters of NS interfaces nor on those of N-terminals. At higher temperatures R_{12} exhibits a peak caused by the trade-off between charge imbalance and Andreev reflection. Our results are in a good agreement with recent experimental observations and can be used for quantitative analysis of future experiments.

In hybrid NS structures quasiparticle current flowing in a normal metal is inevitably converted into that of Cooper pairs inside a superconductor. For quasiparticle energies above the superconducting gap ε > Δ this conversion is accompanied by electron-hole (or charge) imbalance which relaxes inside a superconductor at a typical inelastic length usually denoted as Λ_{Q}. As a result, at temperatures near the critical one T_{C} an electric field penetrates into a superconductor causing resistance enhancement for NS structures under consideration.

At subgap energies ε < Δ the physical picture becomes entirely different. In this case quasiparticle-to-Cooper-pair current conversion is provided by the mechanism of Andreev reflection. A quasiparticle enters the superconductor from the normal metal at a length of order of the superconducting coherence length ξ_{S}, forms a Cooper pair together with another quasiparticle, while a hole goes back into the normal metal. Due to this process subgap conductance of the NS structure remains non-zero down to T = 0K. Furthermore, in the presence of disorder this subgap conductance can be greatly enhanced at low energies due to quantum interference effects.

Further interesting effects may occur in three-terminal NSN structures. Provided the distance between two N-terminals is smaller than or comparable with ξ_{S}, electrons penetrating into the superconductor from the first N-terminal may form Cooper pairs with electrons from the second N-terminal. Then a hole goes into the second N-metal making the charge transfer effectively non-local. This important phenomenon of non-local (or crossed) Andreev reflection (CAR) enables direct experimental realization of entanglement between electrons from spatially separated N-terminals.

CAR was detected and investigated in several recent experiments by measuring the non-local resistance of multiterminal NSN systems. The authors observed a rich structure of different features many of which are still waiting for their theoretical interpretation. Note that not only CAR but also other physical processes contribute to the non-local conductance g_{12} thus making this interpretation rather complicated. For instance, the contribution of elastic cotunneling (EC) to g_{12} exactly cancels that of CAR in the lowest order in NS interface transmissions and at subgap energies. This cancellation is lifted either in higher orders in barrier transmissions or in the presence of interactions, e.g., with an effective external environment, or under external ac bias.

Another important issue is the effect of disorder in metallic terminals which needs to be analyzed for adequate interpretation of experimental results. Although CAR in disordered NSN structures was already addressed in a number of theoretical works, in various physical limits, we believe that general analysis of this issue is still missing in the literature. For instance, the role of disorder-induced electron interference in the non-local subgap transport, the effect of high NS barrier transmissions on CAR as well as some other features remain unclear. Yet another important unresolved problem is to describe an interplay between CAR and non-local charge imbalance. It was demonstrated both experimentally and theoretically that such interplay may result in a large non-local resistance peak which occurs at temperatures slightly below the critical one. It was conjectured that the behavior of this peak is controlled by the charge imbalance length Λ_{Q} parametrically exceeding the length scale ∝ ξ_{S} relevant for CAR.

In this work we develop a general theory of non-local electron transport in diffusive NSN structures which enables one to clarify the above issues and to formulate predictions to be tested in future experiments.

Model and basic equations. In what follows we will analyze a multiterminal diffusive NSN structure schematically shown in Fig. 1. Two normal terminals N_{1} and N_{2} with resistances r_{N_{1}} and r_{N_{2}} and electric poten-
tials $V_1$ and $V_2$ are connected to a superconducting electrode of length $L$ with normal state (Drude) resistance $r_L$ and electric potential $V = 0$ via small NS barriers with resistances $R_1$ and $R_2$ which can be expressed via channel transmissions $T_{1,n}$ and $T_{2,n}$ of these barriers as $1/R_{1(2)} = (e^2/\pi) \sum \sigma T_{1(2),n}$. For the sake of definiteness in Fig. 1 we chose specific geometry directly related, e.g., to experiments\textsuperscript{22} where the superconductor was fabricated in the form of a rather thin strip. NS barriers are located at the points $r_{1,2} = (x_{1,2}, 0, 0)$ and the corresponding segments of a superconducting strip have normal state resistances $r_{x_1}$ and $r_{L-x_2}$, see Fig. 1.

Our analysis is based on the quasiclassical Usadel equations for the Green-Keldysh matrix functions $\hat{G}$. In the absence of interactions these equations read\textsuperscript{22}

$$iD \nabla (\hat{G} \nabla \hat{G}) = [\hat{\Sigma}, \hat{G}], \quad \hat{G}^2 = 1, \quad (1)$$

where $[\hat{a}, \hat{b}] = \hat{a} \hat{b} - \hat{b} \hat{a}$, $D$ is the diffusion constant and

$$\hat{G} = \left( \begin{array}{cc} \hat{G}^R & \hat{G}^K \\ 0 & \hat{G}^R \end{array} \right), \quad \hat{\Sigma} = \Im \left( \begin{array}{cc} \varepsilon + eV & \Delta \\ -\Delta^* & -\varepsilon + eV \end{array} \right) \quad (2)$$

are $4 \times 4$ matrices in Keldysh\textsuperscript{\textregistered}Nambu space, $\varepsilon$ is the quasiparticle energy, $\Delta(T)$ is the superconducting order parameter which will be considered real further below and $V$ is the electric potential.

Far from the interfaces between metals the quasiclassical Green functions $\hat{G}$ coincide with their bulk equilibrium values. Deep in the superconductor they read

$$\hat{G}^{R,A}_S = \pm \frac{\hat{\tau}_3 \varepsilon + i \hat{\tau}_2 \Delta}{\sqrt{(\varepsilon + i \delta)^2 - \Delta^2}}, \quad \hat{G}^K = (\hat{G}^R_S - \hat{G}^A_S)n(\varepsilon), \quad (3)$$

where $n(\varepsilon) = \tanh(\varepsilon/2T)$ and $\hat{\tau}_i$ are Pauli matrices. In the normal terminals far from the tunnel barriers one has

$$\hat{G}^{K,1,2} = \left( \begin{array}{cc} \tanh \frac{\varepsilon + eV}{2T} & 0 \\ 0 & -\tanh \frac{\varepsilon - eV}{2T} \end{array} \right), \quad (4)$$

while the retarded and advanced Green functions $\hat{G}^{R,A}_{1,2}$ are set by the first Eq.\textsuperscript{3} with $\Delta = 0$. In the vicinity of the barriers the Green functions deviate from the above equilibrium values and should be determined from Eqs.\textsuperscript{1} supplemented by appropriate boundary conditions describing electron transfer across metallic interfaces. For diffusive superconductors one finds\textsuperscript{23}

$$A_{1,2} \sigma_1 \hat{G}_{1,2} \sigma_2 \hat{G}_{1,2} = A_{1,2} \sigma \sigma \hat{G}_S \sigma \sigma \hat{G}_S \quad (5)$$

for the first interface and similarly for the second one. Here $A_{1,2}$ are the barrier cross sections and $\sigma_{S,1,2}$ are Drude conductivities of S- and N-terminals.

Having derived the Green-Keldysh functions $\hat{G}$ one can easily evaluate the current density $j$ in our system with the aid of the standard relation

$$j = -\frac{\sigma}{8e} \int \tr[\hat{\tau}_3 (\hat{G} \nabla \hat{G})^K] d\varepsilon. \quad (6)$$

Non-local spectral conductance. The above general formalism enables one to describe electron transport at arbitrary barrier transmissions $T_{1,n}$ and $T_{2,n}$. Here we only assume that both NS barriers are sufficiently small to provide $R_{1,2} > r = \max(r_L, r_{N1}, r_{N2})$. This condition allows to effectively linearize Eqs.\textsuperscript{1} and express the solution of linearized Usadel equations via the diffusion $D^{rr'}(\omega)$ and the Cooperon $C^{rr'}(\omega)$. The diffusion satisfies the following diffusion equation

$$\left(-i\omega + \frac{1}{\tau_{Q^*}} - D \nabla^2\right) D^{rr'}(\omega) = \delta(r - r'), \quad (7)$$

while Cooperon is the solution of Eq.\textsuperscript{2} with effective charge imbalance relaxation time $\tau_{Q}$ replaced by dephasing time $\tau_{\rho}$. At $T \sim T_c$ $\tau_Q$ depends on the electron inelastic relaxation time $\tau_{in}$ as $\tau_Q \sim \tau_{in} T/\Delta(T)$.

Let us employ the standard representation of the Keldysh function $\hat{G}^K = \hat{G}^R \hat{h} - \hat{h} \hat{G}^A$ with $\hat{h} = f_L S_1 + f_T \hat{\tau}_3$, where $f_L$ and $f_T$ are respectively symmetric and antisymmetric in energy parts of the distribution function.

Combining the above expression for $\hat{G}^K$ with Eq.\textsuperscript{6} we define the current across the first barrier

$$I_1 = \frac{1}{2e} \int d\varepsilon g_1(\varepsilon)[n^N_{1} (\varepsilon, r_1) - f^N_{r'}(\varepsilon, r_1)]. \quad (8)$$

The spectral conductance $g_1(\varepsilon)$ is expressed via the functions $\hat{G}^R, \hat{G}^A$. Solving Eq.\textsuperscript{1} for $\hat{G}^R$ and keeping terms up to the first order in $r/R_{1,2}$, we find

$$g_1(\varepsilon) = g_1^{BTK}(\varepsilon) + \frac{\theta(\Delta - |\varepsilon|)\Delta^2}{\Omega^2} \Re C_{1r}^{r'1}(2\varepsilon) \quad (9)$$

$$+ \frac{\Delta^2}{\Omega^2} \sum_{j=1,2} \Re C_{1r}^{r'j}(2W(\varepsilon)) \frac{2e^2}{2e^2 N_j R_j},$$

where $W(\varepsilon) = i\Omega = iv\Delta^2 - \varepsilon^2$ for $|\varepsilon| < \Delta$, $W(\varepsilon) = |\Omega| \sign(\varepsilon)$ for $|\varepsilon| > \Delta$, and $g_1^{BTK}(\varepsilon)$ is defined by the standard expression\textsuperscript{9}

$$g_1^{BTK}(\varepsilon) = \frac{e^2}{\pi} \sum_n \left[ \frac{2 T_{1,n,\theta}^2 (\Delta - |\varepsilon|)\Delta^2}{T_{1,n,\theta}^2 + (2 - T_{1,n,\theta})^2 \Omega^2} \right. \quad (10)$$

$$+ \left. \frac{2 T_{1,n,\theta} (|\varepsilon| - \Delta) |\varepsilon|}{T_{1,n,\theta}^2 + (2 - T_{1,n,\theta})^2 \Omega^2} \right].$$

Note that the terms $\propto C_{S,1}$ in Eq.\textsuperscript{4} are evaluated in the limit $T_{1,n,2,n} \ll 1$ where they only matter as compared to $g_1^{BTK}(\varepsilon)$ provided $R_{1,2} > r$. The Cooperon term $\propto C_1$ describes enhancement of Andreev conductance by electron interference in diffusive N-meta\textsuperscript{15,16} while the term $\propto C_S$ accounts for broadening of the density of states in the superconductor. The spectral conductance $g_2(\varepsilon)$ is given by Eq.\textsuperscript{6} with interchanged indices $1 \leftrightarrow 2$.

Our next step is to solve the kinetic equation for the distribution function $f_T$. In the limit $r/R_{1,2} \rightarrow 0$ this solution is trivial: $f_f^R(\varepsilon) = 0$ and $f_f^{N_j}(\varepsilon, r_j) = h(\varepsilon, V_j) \equiv$
\[ (\tanh(\varepsilon + eV_j)/2T) - \tanh(\varepsilon - eV_j)/2T)/2 \] j = 1, 2. In the first order in \( r/R_{1,2} \) the function \( f_T^S \) is determined from the diffusion equation

\[
(2\tilde{\Omega} - D\nabla^2) f_T = \sum_{j=1,2} \frac{g_j(\varepsilon)h(\varepsilon, V_j)}{2e^2N_S K(\varepsilon)} \delta(\mathbf{r} - \mathbf{r}_j),
\]

where \( K(\varepsilon) = \theta(\Delta - |\varepsilon|)/\Delta^2/\Omega^2 - \theta(|\varepsilon| - \Delta)e^2/\Omega^2, \) and \( \tilde{\Omega} = \theta(\Delta - |\varepsilon|)\Omega \). Resolving Eq. (11) and substituting the result into Eq. (8), we obtain

\[
I_1(V_1, V_2) = \int d\varepsilon [g_{11}(\varepsilon)h(\varepsilon, V_1) - g_{12}(\varepsilon)h(\varepsilon, V_2)],
\]

(12)

and similarly for the current \( I_2 \). The last two terms in the local conductance \( g_{11}(\varepsilon) \) describe partial conductance suppression respectively due to local charge imbalance inside the superconductor and due to non-equilibrium quasiparticles in the normal metal. At energies \( |\varepsilon| > \Delta \) Eq. (11) accounts for the effect of non-local charge imbalance which yields non-zero contribution to \( g_{12}(\varepsilon) \) already in the lowest order in \( 1/R_{1}R_{2} \). In contrast, at subgap energies this lowest order contribution vanishes identically manifesting the well known cancellation between EC and CAR terms. This cancellation is lifted in higher orders in barrier transmissions. Accordingly, the full expression for \( g_{12}(\varepsilon) \) does not vanish also for \( |\varepsilon| < \Delta \) and describes non-trivial interplay between CAR and direct electron transfer in the presence of disorder.

Eq. (12) for the non-local spectral conductance together with Eqs. (4), (10) and (13) is the central result of this work. Note that this result is not specific to particular geometry of Fig. 1 but applies for other diffusive NSN structures as well.

Here, we will only analyze the system with effectively quasi-one-dimensional superconducting and normal wires, as shown in Fig. 1. Assuming \( x_2 > x_1 \) we obtain

\[
\begin{align*}
D_S^{x_1 x_2} &= \frac{\sinh[k(L - x_2)] \sinh kx_1}{kS_1D_S \sinh(kL)}, \\
C_S^{x_1 x_2} &= \frac{\tanh\left(\sqrt{-\omega^2 + 1/\tau_{Q'}}D_S L\right)}{S_1D_S \sqrt{-\omega^2 + 1/\tau_{Q'}}/D_S}.
\end{align*}
\]

where \( j = 1, 2 \). Here \( S_{1,2} \) and \( D_{1,2} \) are respectively effective cross sections and diffusion coefficients of the corresponding terminals and \( k = \sqrt{-\omega^2 + 1/\tau_{Q'}}/D_S \). Substituting Eqs. (15) into (13) - (14) we arrive at the conductance matrix describing the system in Fig. 1.

FIG. 2: Non-local spectral conductance \( g_{12}(\varepsilon) \) (normalized by \( G_0 = r_{11}r_{22}/r_{12}R_1R_2 \)) for diffusive NSN structures. We set \( L = 10\sqrt{2}\xi_S(0), x_1 = 0.45L, \) and \( x_2 = 0.55L. (a) \) The case of two identical barriers with resistances \( R_1 = R_2 = \pi/e^2N_{ch}\tau \) (\( N_{ch} \) is the number of channels and \( \tau \) is the barrier transmission) and for \( R_{1N} = R_{2N} = 0. \) (b) The case of two tunnel barriers with \( T_{1n}, T_{2n} \ll 1 \) and for \( L_1 = L_2 = L_N, R_{1n} = R_{2n}, R_3 = R_2, r_{1N}\xi_N/L_1R_1 = 0.0025 \) and \( \sqrt{R_L\xi_S/}L_1R_1 = 0.005. \)

Zero-bias anomaly. Let us first analyze the tunneling limit \( T_{1n}, T_{2n} \ll 1. \) In this case at subgap energies the term \( g_1^{BTK}(\varepsilon) \) can be neglected and for \( E_{1,2} \equiv D_{1,2}/L_{1,2}^2 \ll |\varepsilon| < \Delta \) we obtain

\[
\begin{align*}
g_{11}(\varepsilon) &= \frac{\Delta^2}{\Omega^2} \left[ \frac{r_{1S}(\varepsilon)}{2R_1^2} + \frac{r_{2S}(\varepsilon)}{R_1R_2} e^{-|\varepsilon - \xi_S(\varepsilon)|/\xi_S} \right], \\
g_{12}(\varepsilon) &= \frac{\Omega^2}{2\Delta^2} r_{1S}(\varepsilon)g_{11}(\varepsilon)g_{22}(\varepsilon) e^{-|\varepsilon - \xi_S(\varepsilon)|/\xi_S}.
\end{align*}
\]

Here \( r_{1S}(\varepsilon) = r_L\xi_S(\varepsilon)/L \) and \( r_{1S}(\varepsilon) = r_{N1}\xi_L(\varepsilon)/L_1 \) are Drude resistances of the segments of S- and N-metals with respective lengths \( \xi_S(\varepsilon) = \sqrt{D_S/2\Omega} \) and \( \xi_L(\varepsilon) = \sqrt{D_{1,2}/|\varepsilon|} \).

At small energies the local spectral conductance diverges as \( g_{11}(\varepsilon) \propto 1/\sqrt{\varepsilon} \) which is just well known disorder-induced zero-bias anomaly. For the non-local conductance this divergence turns out to be even stronger, \( g_{12}(\varepsilon) \propto 1/\varepsilon, \) since quantum interference in both diffusive normal metals simultaneously enhances non-local electron transport in our system. Thus, we predict a sharp low energy peak in the non-local conductance which occurs in the presence of disorder in the N-terminals, see also Fig. 2. Accordingly, the differential conductance \( G_{12}(V_2, T) = -\partial I_1/\partial V_2 \) increases as \( G_{12} \propto 1/\max(eV_2, T) \) with decreasing voltage and temperature.

Eq. (16) applies down to \( \varepsilon \sim E_1 \) and for even smaller energies \( r_{1S}(\varepsilon) \) should be substituted by \( 2r_{N1} \). Then for \( r_{N1}, r_{N2} \gg r_{1S}(\varepsilon) \) we get

\[
g_{12}(0) = G_{12}(0, 0) = \frac{r_{1S}r_{N1}r_{N2}}{2R_1^2R_2} e^{-|x_2-x_1|/\xi_S}.
\]

We also note that in the case of strongly asymmetric barriers \( R_{1,2} \ll R_1 \) the dominating contribution to \( g_{12} \) scales as \( \propto 1/R_1R_2^3 \) rather than \( \propto 1/R_1^3R_2^3 \).

Turning to the case of high barrier transmissions \( T_{1n}, T_{2n} \ll 1 \) we observe that in this case \( g_1 \) is dominated by \( g_1^{BTK} \) while other contributions can be neglected. In particular, for fully open barriers at subgap
FIG. 3: (a) Non-local resistance $R_{12}(T)$ (20), normalized by $R_0 = r_{s1} r_L - x_2 / r_L$, at different barrier transmissions $\tau$ and at $T = \tau \rightarrow \infty$. We chose $R_1 = R_2 = \pi e^2 N_{A,h} \tau = h/2e^2$ and set $L = 20 \sqrt{2} \xi_s(0)$, $x_1 = 9 \sqrt{2} \xi_s(0)$, $x_2 = 11 \sqrt{2} \xi_s(0)$, $r_L = 200 \Omega$ and $r_{N1} = r_{N2} = 0$. (b) $R_{12}(T)/R_0$ at different $|x_2 - x_1|$. Other parameters are the same as in Fig. 2b.

energies we obtain $g_{1,2} = g_{1,2}^{BTK} = 2/R_{1,2}$ and, hence,

$$g_{12}(\varepsilon) = \frac{\Omega^2}{\Delta^2} \frac{2r_{s\xi}(\varepsilon)}{R_1 R_2} e^{-|x_2 - x_1|/\xi_s(\varepsilon)}, \quad |\varepsilon| < \Delta. \tag{19}$$

Non-local resistance and charge imbalance peak. Let us now define non-local linear resistance

$$R_{12}(T) = \frac{G_{12}(0,T)}{G_{11}(0,T) G_{22}(0,T) - G_{12}(0,T)^2} \tag{20}$$

Combining this equation with Eq. (14), at $T \ll \Delta$ we arrive at a very simple and universal formula

$$R_{12} = \left( r_{s\xi} / 2 \right) e^{-|x_2 - x_1|/\xi_s}. \tag{21}$$

It is remarkable that independently of both barrier and $N$-terminal parameters the subgap non-local resistance is set only by the normal state resistance $r_{s\xi}$ of the superconducting wire segment of length $\xi_S$ and by the distance between the barriers measured in units of $\xi_S$. At low $T$ the dependence $R_{1,2} \approx r_0 \exp[-|x_2 - x_1|/\xi_S(0)]$ was observed in experiments with $r_0 \approx 0.56 \Omega$. For the parameters we estimate $r_0 = r_{s\xi}/2$ in the range of one $\Omega$. A similarly good agreement is found between Eq. (21) and experimental results.

The temperature dependence of $R_{12}(T)$ is depicted in Fig. 3. In the tunneling limit it exhibits a well pronounced peak which originates from the competition between charge imbalance and Andreev reflection. The maximum value of the non-local resistance $R_{12}$ is reached at $T^* = 2\Delta / \ln(R_1 R_2/\xi_s^2)$ and reads

$$R_{12}(T^*) \approx \frac{\alpha r_{x1}}{\lambda} \sqrt{\frac{1 - |x_2 - x_1|}{\lambda}} \left( \frac{r_{s\xi} + r_{1}(T^*)}{R_1} + \frac{r_{s\xi} + r_{2}(T^*)}{R_2} \right)^2, \tag{22}$$

where $\lambda = \alpha L$ and $\alpha = 1 - r_{x1}/r_1$. Thus, the peak resistance $R_{12}(T^*)$ decreases linearly with increasing distance $|x_2 - x_1|$ between the barriers. This behavior agrees well with recent observations. Furthermore, with the parameters we estimate $\lambda$ to be of order a micron in agreement with experimental findings. Such values of $\lambda$ appear lower than typical values of the charge imbalance relaxation length $\lambda Q = \sqrt{D_S T Q^*}$. The latter length scale is expected to gain importance only for $\lambda Q < \lambda$.

In summary, we developed a microscopic theory of non-local electron transport in diffusive NSN systems which accounts for non-trivial interplay between crossed Andreev reflection, disorder, quantum interference and non-local charge imbalance. Our results can be directly used for quantitative analysis of future experiments.

This work was supported in part by RFBR grant 09-02-00886. D.S.G. and M.S.K. also acknowledge support respectively from DFG Center for Functional Nanostructures (CFN) and from the Dynasty Foundation.

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