Full Counting Statistics of Multiple Andreev Reflections

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We derive the full distribution of transmitted particles through a superconducting point contact of arbitrary transparency under voltage bias. The charge transport is dominated by multiple Andreev reflections. The counting statistics is a multinomial distribution of processes, in which multiple charges ne ($n = 1, 2, 3, \dots$) are transferred through the contact. For zero temperature we obtain analytical expressions for the probabilities of the multiple Andreev reflections. The current, shot noise, and high current cumulants in a variety of situations can be obtained from our result.

The complete understanding of the electronic transport in mesoscopic systems requires information that goes beyond the analysis of the current. This explains the great attention devoted in the last few years to current fluctuations in these systems [1]. An important goal is to obtain the full current distribution. This was realized by Levitov and co-workers [2], who borrowed the concept of full counting statistics (FCS) for photons and adapted it to electrons in mesoscopic systems. FCS gives the probability $P(N)$ that N charge carriers pass through a conductor in the measuring time. From the knowledge of these probabilities one can easily derive not only the conductance and noise, but all the cumulants of the current distribution. Since the introduction of FCS for electronic systems, the theory has been sophisticated and applied to many different contexts (for a recent review, see [3]). In particular, one of the authors and Nazarov have shown that, based on a Keldysh-Green function method, one can calculate in a unified manner the FCS of all contacts involving superconducting elements [4,5].

In the context of superconductivity it was shown in Ref. [6] that the FCS of a normal metal-superconductor point contact is a binomial distribution of pairs of electrons, which proceed in the superconductor as Cooper pairs. However, the FCS in the basic situation of a point contact between two superconductors out of equilibrium has not been investigated yet. In this system the transport properties for voltages V below the superconducting gap Δ are dominated by coherent multiple Andreev reflections (MAR) [7]. Recently, the microscopic theory of MAR [8] has provided new insight into this problem and has allowed the calculation of properties beyond the current such as the shot noise [9]. The predictions of this theory have been quantitatively tested in an impressive series of experiments in atomic-size contacts [10–12]. In particular, the analysis of the shot noise [9,12] has suggested that the current at subgap energies proceeds in "giant" shots, with an effective charge $q \sim e(1 + 2\Delta/|eV|)$. However, the question of whether the charge in these contacts is indeed transferred in big

chunks can be rigorously resolved only by the analysis of the FCS. This leads us to the central question addressed in this Letter: what is the FCS of MAR?

The answer, which we derive below, is that the statistics is a *multinomial* distribution of multiple charge transfers. Technically, we find that the cumulant generating function (CGF) for a voltage V has the form

$$S(\chi) = \frac{t_0}{h} \int_0^{eV} dE \ln\left[1 + \sum_{n=-\infty}^{\infty} P_n(E, V)(e^{in\chi} - 1) \right], \quad (1)$$

where t_0 is the measuring time. The CGF is related to the FCS by $P(N) = \int_{-\pi}^{\pi}(d\chi/2\pi) \exp[S(\chi) - iN\chi]$. The different terms in the sum in Eq. (1) correspond to transfers of multiple charge quanta ne at energy E with the probability $P_n(E, V)$, which can be seen by the $(2\pi/n)$ periodicity of the accompanying χ-dependent counting factor. This result proves that the charges are indeed transferred in large quanta. Below we find for any kind of superconducting junction explicit expressions for $P_n(E, V)$, which is just the probability of an nth-order MAR. In this process a quasiparticle injected at energy E is $n - 1$ times Andreev reflected to be transmitted at energy $E + neV$, resulting in a transfer of n electron charges. This is illustrated in Fig. 1 for BCS superconductors.

To arrive at these conclusions, we consider a voltage-biased superconducting point contact, i.e., two superconducting electrodes linked by a constriction, which is shorter than the coherence length and is described by a transmission probability T. To obtain the FCS in our system of interest we make use of the Keldysh-Green function approach to FCS introduced by Nazarov and one of the authors [4,5]. The FCS of superconducting constrictions has the general form [5]

$$S(\chi) = \frac{t_0}{h} \text{Tr} \ln\left[1 + \frac{T}{4}[\{\check{G}_1(\chi), \check{G}_2\}_\otimes - 2] \right]. \quad (2)$$

Here $\check{G}_{1(2)}$ denote matrix Green functions of the left and the right contact. The symbol $\otimes$ implies that the products of the Green functions are convolutions over
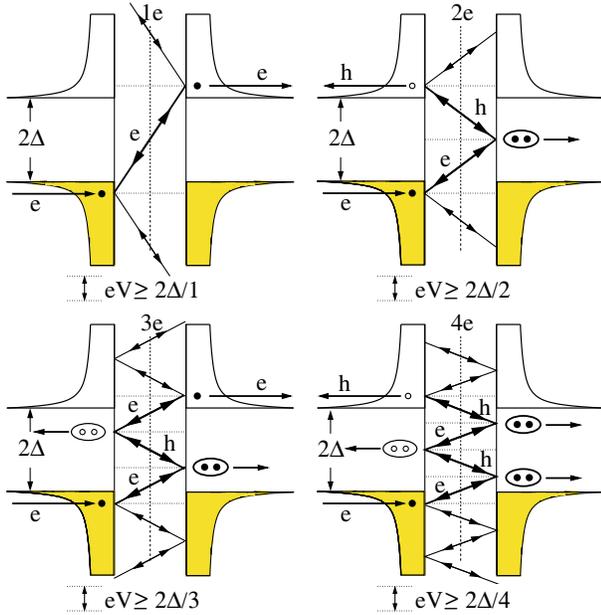
FIG. 1 (color online). Schematic representation of the MARs for BCS superconductors with gap Δ. We have sketched the density of states of both electrodes. In the upper left panel we describe the process in which a single electron tunnels through the system overcoming the gap due to a voltage $eV \geq 2\Delta$. The other panels show MARs of order $n = 2, 3, 4$. In these processes an incoming electron at energy E undergoes at least $n - 1$ Andreev reflections to finally reach an empty state at energy $E + neV$. In these MARs a charge ne is transferred with a probability, which for low transparencies goes as T^n. At zero temperature they have a threshold voltage $eV = 2\Delta/n$. The arrows pointing to the left in the energy trajectories indicate that a quasiparticle can be normal reflected. The lines at energies below E and above $E + neV$ indicate that after a detour a quasiparticle can be backscattered to finally contribute to the MAR of order n.

the internal energy arguments, i.e., $(G_1 \otimes G_2)(E, E') = \int dE_1 G_1(E, E_1) G_2(E_1, E')$. The trace runs not only over the Keldysh-Nambu space, but also includes integration energy. For a superconducting contact at finite bias voltage the CGF depends on time and Eq. (2) is integrated over a long measuring time t_0, much larger than the inverse of the Josepshon frequency.

Let us now describe the Green functions entering Eq. (2). The counting field χ is incorporated into the matrix Green function of the left electrode as follows:

$$\check{G}_1(\chi, t, t') = e^{-i\chi\check{\tau}_K/2}\check{G}_1(t, t')e^{i\chi\check{\tau}_K/2}. \quad (3)$$

Here $\check{G}_1(t, t')$ is the reservoir Green function in the absence of the counting field and $\check{\tau}_K = \hat{\sigma}_3\bar{\tau}_3$ a matrix in Keldysh($\hat{\ }$)-Nambu($\bar{\ }$) space. We set the chemical potential of the right electrode to zero and represent the Green functions by $\check{G}_1(t, t') = e^{ieVt\bar{\tau}_3}\check{G}_S(t - t')e^{-ieVt'\bar{\tau}_3}$ and $\check{G}_2(t, t') = \check{G}_S(t - t')$. Here, we have not included the dc part of the phase, since it can be shown that it drops from the expression of the dc FCS at finite bias. $\check{G}_S$ is the Green function of a superconducting reservoir (we consider the

case of a symmetric junction), which reads

$$\check{G}_S(E) = \begin{pmatrix} (\bar{A} - \bar{R})f + \bar{R} & (\bar{A} - \bar{R})f \\ (\bar{A} - \bar{R})(1 - f) & (\bar{R} - \bar{A})f + \bar{A} \end{pmatrix}. \quad (4)$$

Here $\bar{R}(\bar{A})(E)$ are retarded and advanced Green functions of the leads and $f(E)$ is the Fermi function. Advanced and retarded functions in (4) have the Nambu structure $\bar{R}(\bar{A}) = g^{R,A}\bar{\tau}_3 + f^{R,A}\bar{\tau}_1$ fulfilling the normalization condition $f^2 + g^2 = 1$. They depend on energy and the superconducting order parameter Δ.

In Eq. (2) the matrix appearing inside the logarithm has an infinite dimension in energy space. In the case of N-N or N-S contacts such a matrix is diagonal in this space, which makes almost trivial the evaluation of the FCS. In the S-S case at finite bias this is no longer true, which introduces an enormous complication.

We now tackle the problem of how the functional convolution in Eq. (2) can be treated. The time dependence of the Green functions leads to a representation of the form $\check{G}(E, E') = \sum_n \check{G}_{0,n}(E)\delta(E - E' + neV)$, where $n = 0, \pm 2$. Restricting the fundamental energy interval to $E - E' \in [0, eV]$ allows one to represent the convolution as a matrix product, i.e., $(G_1 \otimes G_2)(E, E') \rightarrow (\check{G}_1\check{G}_2)_{n,m}(E, E') = \sum_k (G_1)_{n,k}(E, E')(G_2)_{k,m}(E, E')$. We write the CGF as $S(\chi) = (t_0/h)\text{Tr}\ln\check{Q}$, where $\check{Q} = 1 + (\sqrt{T}/2)[\check{G}_1(\chi) - \check{G}_2]$ [13]. The trace in this new representation is written as $\int_0^{eV} dE\sum_n \text{Tr}\ln(\check{Q})_{nn}$. In this way the convolution is reduced to matrix algebra for the infinite-dimensional matrix $\check{Q}$. Still, the task to compute $\text{Tr}\ln\check{Q}$ is nontrivial. However, noting that $\text{Tr}\ln\check{Q} = \ln\det\check{Q}$, it is obvious at this stage that $\det\check{Q}$ has the form of a Fourier series in χ, which allows us to write the CGF as follows:

$$S(\chi) = \frac{t_0}{h}\int_0^{eV} dE\ln\left[\sum_{n=-\infty}^{n=\infty} P_n'(E, V)e^{in\chi}\right]. \quad (5)$$

Keeping in mind the normalization $S(0) = 0$, it is clear that one can rewrite this expression in the form anticipated in Eq. (1), where the probabilities are given by $P_n(E, V) = P_n'(E, V)/\sum_{n=-\infty}^{n=\infty} P_n'(E, V)$. Of course, one has still to extract the expression of these probabilities from the determinant of $\check{Q}$, which is a nontrivial task. It turns out that $\check{Q}$ has a block-tridiagonal form, which allows one to use a standard recursion technique. We define the following 4×4 matrices:

$$\check{F}_{\pm n} = \check{Q}_{\pm n, \pm n} - \check{Q}_{\pm n, \pm n \pm 2}\check{F}_{\pm n \pm 2}^{-1}\check{Q}_{\pm n \pm 2, \pm n}; \qquad n \geq 2;$$

$$\check{F}_0 = \check{Q}_{0,0} - \check{Q}_{0,-2}\check{F}_{-2}^{-1}\check{Q}_{-2,0} - \check{Q}_{0,2}\check{F}_2^{-1}\check{Q}_{2,0}, \quad (6)$$

where $\check{Q}_{n,m}(E) = \check{Q}(E + neV, E + meV)$. With these definitions, $\det\check{Q}$ is given by $\det\check{Q} = \prod_{j=-\infty}^{\infty} \det\check{F}_{2j}$. In practice, $\det\check{F}_n = 1$ if $|n| \gg \Delta/|eV|$. This reduces the problem to the calculation of determinants of 4×4 matrices.

In the zero-temperature limit one can work out this idea analytically to obtain the following expressions for the probabilities:

$$P'_n(E, V) = \sum_{l=0}^{n-1} K_{-n+l-2,l+2} J_{-n+l}(E) \left[\prod_{k=-n+l+1}^{l-1} (T/4)|f^A_k|^2 \right] J_l(E); \qquad n \geq 1;$$

$$P'_0(E, V) = K_{0,0} \left[Z^R_0 \left(1 + \frac{\sqrt{T}}{2}(g^R_0 - g^A_{-1}) - \frac{T}{4}(f^A_{-1})^2 B^A_{-2} \right) - \frac{T}{4}(f^R_0)^2 \right] [R \leftrightarrow A]. \tag{7}$$

Here, we have used the shorthand $g^{A,R}_n(E) \equiv g^{A,R}(E + neV)$, and we defined

$$Z^\alpha_{\pm n} = 1 \pm \frac{\sqrt{T}}{2}(g^\alpha_{\pm(n+1)} - g^\alpha_{\pm n}) - \frac{T}{4}(f^\alpha_{\pm(n+1)})^2 B^\alpha_{\pm(n+2)}; \qquad n \geq 0, \tag{8}$$

where $\alpha = R, A$, $K_{n,m} = (\prod_{j=1}^\infty \det\check{F}_{n-2j})(\prod_{j=1}^\infty \det\check{F}_{m+2j})$, and the different functions can be expressed as follows:

$$(B^\alpha_{\pm n})^{-1} = 1 \pm \frac{\sqrt{T}}{2}(g^\alpha_{\pm n} - g^\alpha_{\pm(n-1)}) - \frac{T}{4}(f^\alpha_{\pm n})^2/Z^\alpha_{\pm n}; \qquad \det\check{F}_{\pm n} = \prod_{\alpha=A,R} \left[Z^\alpha_{\pm n}\left(1 \pm \frac{\sqrt{T}}{2}(g^\alpha_{\pm n} - g^\alpha_{\pm(n-1)}) \right) - \frac{T}{4}(f^\alpha_{\pm n})^2 \right];$$

$$J_{\pm n} = \frac{\sqrt{T}}{2}(g^A_{\pm n} - g^R_{\pm n}) \left[Z^R_{\pm n} Z^A_{\pm n} - \frac{T}{4}|f^A_{\pm n}|^2 \right] \mp \frac{T}{4}(f^A_{\pm n} - f^R_{\pm n})[f^R_{\pm n} Z^A_{\pm n} + f^A_{\pm n} Z^R_{\pm n}]. \tag{9}$$

Notice that, since at zero temperature the charge flows only in one direction, only the P_n with $n \geq 0$ survive. Let us remark that $P_n(E, V)$ are positive numbers bounded between 0 and 1. Although at a first glance they look complicated, they can be easily computed and provide the most efficient way to calculate the transport properties of these contacts. In practice, to determine the functions $B^{A,R}_n$ and $\det\check{F}_n$, one can use the boundary condition $B^{A,R}_n = \det\check{F}_n = 1$ for $|n| \gg \Delta/|eV|$. For perfect transparency ($T = 1$) the previous expressions greatly simplify and the probabilities $P_n(E, V)$ can be written as

$$P_n = \sum_{l=0}^{n-1}(1 - |a_{-n+l}|^2) \left[\prod_{k=-n+l+1}^{l-1} |a_k|^2 \right](1 - |a_l|^2), \tag{10}$$

where $a(E)$ is the Andreev reflection coefficient defined as $a(E) = -if^R(E)/[1 + g^R(E)]$, and $a_n = a(E + neV)$.

In view of Eqs. (7)–(9) the probabilities P_n can be interpreted in the following way. P_n is the probability of a MAR of order n, where a quasiparticle in an occupied state at energy E is transmitted to an empty state at energy $E + neV$. The typical structure of the leading contribution to this probability consists of the product of three terms. First, J_0 gives the probability to inject the incoming quasiparticle at energy E. The term $\prod_{k=1}^{n-1}(T/4)|f^A_k|^2$ describes the cascade of $n - 1$ Andreev reflections, in which an electron is reflected as a hole and vice versa, gaining an energy eV in each reflection. Finally, J_n gives the probability to inject a quasiparticle in an empty state at energy $E + neV$. In the tunnel regime $P_n(E, V) = (T^n/4^{n-1})\rho_0\rho_n \prod_{k=1}^{n-1}|f^A_k|^2$, $\rho(E)$ being the reservoir density of states. This interpretation is illustrated in Fig. 1, where we show the first four processes for BCS superconductors. The product of the determinants in the expression of P'_n [see Eq. (7)] describes the possibility that a quasiparticle makes an excursion to energies below E or above $E + neV$ [14]. In the tunnel regime this possibility is very unlikely and at perfect transparency is forbidden. As can be seen in Eq. (10),

for $T = 1$ the quasiparticle can move only upwards in energy due to the absence of normal reflection.

From the knowledge of the FCS one can get a deep insight into the different transport properties by analyzing the role played by every process. For instance, in Fig. 2 we show the contribution to the dc current of the individual processes, i.e., $I_n = (2e/h) \int dE\, n P_n$, for the case of BCS superconductors of gap Δ. In this case $f^{A,R} = i\Delta/[(E \mp i\delta)^2 - \Delta^2]$, where $\delta = 0^+$, and $g^{A,R}$ follows from normalization. As can be seen in Fig. 2, a MAR of order n has a threshold voltage $eV = 2\Delta/n$, below which it cannot occur. The opening of MARs at these threshold voltages is the origin of the pronounced subgap structure visible in the different transport properties (see Fig. 3). Notice also that at low transmission the MAR of order n dominates the transport for voltages $[2\Delta/n, 2\Delta/(n - 1)]$, while at high transparencies several MARs give a significant contribution at a given voltage. This naturally explains why the effective charge is quantized only in the tunnel regime [9,12].
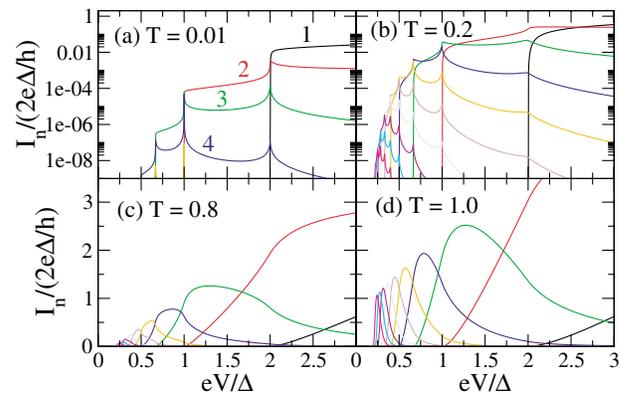

FIG. 2 (color online).   Current contribution of processes $n = 1, \ldots, 10$, from right to left, as a function of voltage for BCS superconductors of gap Δ. The different panels correspond to different transmissions. Notice the logarithmic scale in the upper panels.
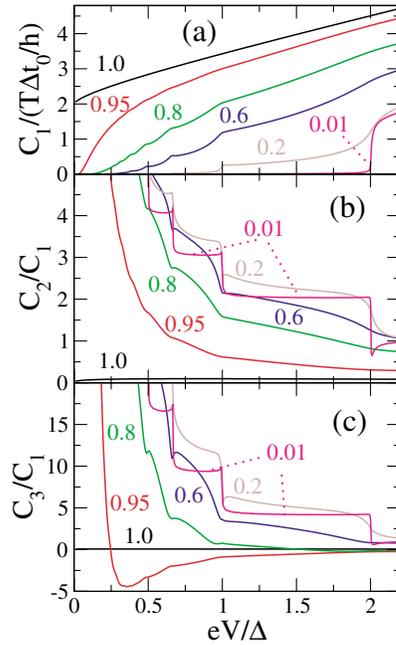
FIG. 3 (color online). (b),(c) Second and third cumulant at zero temperature for a contact between BCS superconductors. Both are normalized to the first cumulant (the average current) shown in panel (a). The transmissions are indicated in the plots.

From the CGF one can easily calculate the cumulants of the distribution and in turn many transport properties. Of special interest are the first three cumulants $C_1 = \overline{N}$, $C_2 = \overline{(N - \overline{N})^2}$, and $C_3 = \overline{(N - \overline{N})^3}$, which correspond to the average, width, and skewness of the distribution, respectively. From the fact that the FCS is a multinomial distribution, it follows that at zero temperature these cumulants can be expressed in term of the probabilities $P_n(E, V)$ as $C_n(V) = (t_0/h) \int_0^{eV} dE C_n(E, V)$, where

$$C_1(E, V) = \sum_{n=1}^{\infty} n P_n; \qquad C_2(E, V) = \sum_{n=1}^{\infty} n^2 P_n - C_1^2(E, V);$$

$$C_3(E, V) = \sum_{n=1}^{\infty} n^3 P_n - C_1(E, V)[C_1^2(E, V) + 3C_2(E, V)].$$

The first two cumulants are simply related to the dc current, $I = (2e/t_0)C_1$, and to the zero-frequency noise $S_I = (4e^2/t_0)C_2$. In Fig. 3 we show C_2 normalized by C_1, which reproduces the results for the shot noise reported in the literature [9]. In this figure we also show C_3. This cumulant determines the shape of the distribution, and it is attracting considerable attention [15,16] because it contains information on nonequilibrium physics even at temperatures larger than the voltage. As seen in Fig. 3, at low transmissions $C_3 = q^2 C_1$, where $q(V) = 1 + \text{Int}(2\Delta/eV)$ is the charge transferred in the MAR which dominates the transport at a given voltage. This relation is a striking example of the general relation conjectured in Ref. [15]. For higher transmissions this cumulant is negative at high voltage as in the normal state, where $C_3 = T(1 - T) \times$

$(1 - 2T)$, but it becomes positive at low bias. This sign change is due to the reduction of the MAR probabilities at low voltage. After the sign change there is a huge increase of the ratio C_3/C_1, which is a signature of the charge transfer in large quanta. Finally, at $T = 1$ the cumulants C_n (with $n > 1$) do not completely vanish due to the fact that at a given voltage different MARs give a significant contribution, and therefore their probability is smaller than 1 [see Fig. 2(d)].

In summary, we have demonstrated that in superconducting contacts at finite voltage the charge transport is described by a multinomial distribution of multiple charge transfers. This proves that in the MAR processes the charge is transmitted in large quanta. We have obtained analytically the MAR probabilities at zero temperature, from which all transport properties are easily computed. Our result constitutes the culmination of the recent progress in the understanding of MARs, which are a key concept in mesoscopic superconductivity.

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