Single-Electron Charging of a Superconducting Island

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We have calculated the quasiparticle current through a superconducting island in the Coulomb blockade regime. The current depends strongly on the parity of the total number of free electrons in the island. This dependence reflects the difference between ground-state properties of the superconductor with even and with odd number of electrons.

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Quantization of the electric charge Q of an isolated conductor in units of electron charge e, $Q=en$, where n is the number of excess electrons in the conductor, has been well known since the classic experiments of Millikan [1]. Recently it was realized that charge quantization leads to a variety of new physical phenomena in systems of tunnel junctions formed between conductors of small electric capacitance C [2,3]. These phenomena are based on the existence of the energy gaps of the order of the characteristic charging energy $e^2/2C$ between states with different n, which fix the number of electrons in the conductors. The gaps can be varied continuously, for example, by external voltage, so that one can control tunneling of single electrons to or from the conductors.

In conductors that are adequately described by the approximation of noninteracting electrons, the charge quantization coexists in a simple way with the quantization of electron energies [4]. Electron tunneling via a conductor with a fixed number of electrons enables one to observe its single-particle energy spectrum. A remarkable example of such a spectroscopy is given by the observation of the energy spectrum of a semiconductor quantum dot in the integer quantum Hall effect regime [5].

Even more interesting questions arise when charge quantization coexists with electron-electron interactions inside the conductor, for example, when it is a superconductor. From simple considerations (well known in the context of pairing in nuclei—see, e.g., Ref. [6]) based on the BCS theory, it follows that the ground-state energy E_0 of a superconducting island with fixed number N of free electrons depends on the parity of this number even when it is macroscopically large [7,8]. Since the number of paired electrons can only be even, for odd N one electron is necessarily in the unpaired state, so that the E_0 for odd N is larger than the E_0 for even N by the superconducting gap Δ.

This means that electron transport properties of a small superconducting island should depend on the parity of the total number of free electrons in it. Here we consider such an island between bulk normal electrodes that form two tunnel junctions in series [Fig. 1(a)]. Specifically, we calculate the current through this structure in the Coulomb blockade regime, i.e., at small bias voltages V [2,3]. In this regime the small electric capacitance C_Σ of the middle electrode provides a Coulomb energy barrier $\simeq E_C=e^2/2C_\Sigma$ for electron transfer in either of the tunnel junctions, so that the tunneling current is suppressed. However, the zero-current state is metastable due to quantum fluctuations of the charge that allow simultaneous transfer of electrons in both junctions without a charging of the middle electrode. There are two channels of such a quantum decay of the Coulomb blockade: inelastic [9–11], which leads to the creation of electron-hole excitations in the middle electrode, and elastic [12,13]. The rates γ_{in} and γ_{el} of these co-tunneling processes determine the current through the double-junction system below the Coulomb blockade threshold, and were calculated for systems with normal electrodes [9,13]. The inelastic process dominates in systems with not too small electrodes, and it has been observed in both metallic [10] and semiconductor junctions [11].

The superconductivity strongly modifies the co-tunneling processes, in that the energy gap Δ creates an additional tunnel barrier which depends on the parity of the total number N of free electrons in the superconductor. When N is even, inelastic tunneling cannot take place at voltages below $2\Delta/e$ because of the energy gap for creation of excitations, and only elastic tunneling is allowed. For odd N a quasiparticle exists unavoidably in

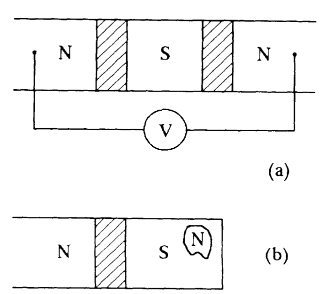

FIG. 1. The two systems under consideration: (a) ideal superconducting electrode between normal bulk electrodes forming two tunnel junctions in series; (b) superconducting "single-electron box" with small normal inclusion. Dashed regions denote tunnel barriers.

the superconductor and opens the inelastic channel as well. However, even for odd N the inelastic tunneling is suppressed in comparison to the normal metal, so that $\gamma_{\rm in}$ and $\gamma_{\rm el}$ are of the same order of magnitude.

The total number N of free electrons in the superconducting electrode depends on the electrostatic energy of the structure [2,3]:

$$U=\frac{Q^2}{2C_\Sigma}-\frac{eV}{C_\Sigma}(C_1n_2+C_2n_1)\,,\qquad Q=en-\varphi C_\Sigma,\;\; n=n_1-n_2,\;\; C_\Sigma=C_1+C_2\,. \tag{1}$$

Here n_j is the number of electrons that have tunneled through the jth junction, $C_{1,2}$ are junction capacitances, and φ is the potential difference between the middle and external electrodes at vanishing bias voltage V. In the Coulomb blockade regime N depends only on φ and its parity can be found by minimization of the ground-state energy $E(N)$ of the superconductor (including the charging energy) with respect to N:

$$E(N)=\frac{[e(N-N_0)-\varphi C_\Sigma]^2}{2C_\Sigma}+E_0(N)\,,\qquad E_0(N)=\begin{cases}\Delta & \text{for odd } N,\\ 0 & \text{for even } N,\end{cases} \tag{2}$$

where N_0 is the number of electrons in the superconductor at $\varphi=0$. The energy (2) as a function of φ is shown in Fig. 2. Different parabolas in this figure correspond to different n, and the lowest-lying curve at a given φ corresponds to the equilibrium number of electrons in the superconductor. One can see that when $\Delta<E_c$ and φ increases, N increases by 1 and changes from odd to even at $\varphi=\varphi_p^-$ and from even to odd at $\varphi=\varphi_p^+$, where

$$\varphi_p^\pm C_\Sigma/e=2p+N_0(\mathrm{mod}2)\pm\tfrac{1}{2}(1+\Delta/E_c)\,,\qquad p=0,\pm1,\ldots. \tag{3}$$

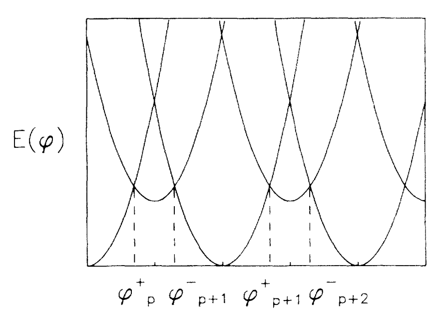

FIG. 2. Ground-state energy (2) of the superconducting island as a function of externally induced potential difference φ between the island and bulk external electrodes. The upper and lower sets of parabolas correspond, respectively, to odd and even numbers of electrons in the island. The lowest parabola at a given φ determines the ground-state energy. The intersections of parabolas give the φ values at which the number of electrons in the island is changed.

N can only be even if $\Delta>E_c$, and it is changed by 2 when φ passes through $\varphi_p=[2p+1+N_0(\mathrm{mod}2)]e/C_\Sigma$.

From now on, the calculations proceed along the same lines as in Ref. [13]. Besides the electrostatic energy (1), the Hamiltonian of the double-junction system includes the Hamiltonians of the normal external electrodes, $H_{1,2}$, the superconducting middle electrode, H_0, and the terms H_{T1}, H_{T2} describing tunneling between these electrodes:

$$H_{Tj}=H_j^++H_j^-\,,\quad H_j^-=(H_j^+)^\dagger\,,\qquad H_j^+=\sum_{k,m}T_{mk}^{(j)}(u_kb_k^\dagger+v_kb_k)c_m\,. \tag{4}$$

Here $c_m^\dagger, c_m$ and $b_k^\dagger, b_k$ are the creation and annihilation operators of, respectively, electrons in external electrodes and quasiparticles in the middle electrode, and u_k, v_k are the usual BCS factors [14]:

$$u_k^2=\tfrac{1}{2}[1+\varepsilon_k/(\Delta^2+\varepsilon_k^2)^{1/2}],\quad v_k^2=1-u_k^2\,.$$

We calculate the current associated with inelastic tunneling at small voltages, $V>2\Delta/e$, when only existing quasiparticles can participate in the tunneling. As we discussed above, in the φ region where the superconductor has an even number of electrons [see Eq. (3)], there are no quasiparticles in it at small temperatures, $T\ll\Delta$, and inelastic tunneling is suppressed. By contrast, in the φ region with an odd number of electrons, one quasiparticle should exist even at $T\ll\Delta$ and the rate of inelastic tunneling $\gamma_{\rm in}$ is nonvanishing. We assume that $\gamma_{\rm in}$ is much smaller than the energy relaxation rate in the superconductor, so that the quasiparticles (excited by the inelastic tunneling) relax into the energy state ε_q at the bottom of the energy spectrum, i.e., $\varepsilon_q=\Delta$, and $u_q^2=v_q^2=\frac{1}{2}$. The rate $\gamma_{\rm in}$ is given by the sum of the partial rates of electron transfer between the energy states $\varepsilon_m\to\varepsilon_{k,q}\to\varepsilon_n$ [9,13]. The matrix elements of these transitions are (for clarity we omit spin indices)

$$\langle M\rangle=\frac{T_{qm}^{(1)}T_{kn}^{(2)}v_qv_k}{E_1-\Delta-\varepsilon_m}+\frac{T_{km}^{(1)}T_{qn}^{(2)}u_qu_k}{E_1+(\varepsilon_k^2+\Delta^2)^{1/2}-\varepsilon_m}+\frac{T_{qm}^{(1)}T_{kn}^{(2)}v_qv_k}{E_2+(\varepsilon_k^2+\Delta^2)^{1/2}+\varepsilon_n}+\frac{T_{km}^{(1)}T_{qn}^{(2)}u_qu_k}{E_2-\Delta+\varepsilon_n}\,. \tag{5}$$

Here E_j is the change of electrostatic energy (1) due to electron transfer in the jth junction, and the tunneling amplitudes $T_{mk}^{(j)}$ were chosen to be real. The first term describes the process in which an electron jumps into the state ε_q, so that the unpaired electron in this state becomes paired, and then another electron jumps out of the middle electrode creating a quasiparticle in some other state ε_k. Other terms in Eq. (5) can be interpreted similarly.

Next, we make two assumptions. First, we assume that one can average over the state ε_q (e.g., due to finite temperature, $T\gg\delta$), so that the tunneling rates between this state and external electrodes can be directly related to the normal junction conductances $G_{1,2}$. Second, we take into account that the contribution to $|\langle M\rangle|^2$ of the terms that contain the products $T^{(1)}_{km}T^{(1)}_{qm}$ is smaller by a factor δ/Δ than that of those that contain only absolute values of tunneling matrix elements [13], and can be neglected. Under these assumptions one gets

$$\gamma_{\rm in}=\frac{\hbar G_1G_2\delta}{4\pi e^4}\int d\varepsilon_m\, d\varepsilon_n\, d\varepsilon_k\, f(\varepsilon_m)[1-f(\varepsilon_n)]$$
$$\times\left[u_k^2u_q^2\left(\frac{1}{E_1+(\varepsilon_k^2+\Delta^2)^{1/2}-\varepsilon_m}+\frac{1}{E_2+\Delta+\varepsilon_n}\right)^2\right.$$
$$\left.+v_k^2v_q^2\left(\frac{1}{E_1-\Delta-\varepsilon_m}+\frac{1}{E_2+(\varepsilon_k^2+\Delta^2)^{1/2}+\varepsilon_n}\right)^2\right]\delta(\varepsilon_n-\varepsilon_m+(\varepsilon_k^2+\Delta^2)^{1/2}-\Delta-eV)\,, \tag{6}$$

where $f(\varepsilon)$ is the Fermi distribution function, and δ denotes the average spacing between spin-degenerate levels of the middle electrode.

At small temperatures, $T\ll eV$, and voltages, $eV\ll E_1,E_2$, the integrals in Eq. (6) can be calculated explicitly:

$$\gamma_{\rm in}=\frac{\hbar G_1G_2\delta}{16\pi e^4}\Delta^2\left[\left(\frac{1}{E_1-\Delta}+\frac{1}{E_2+\Delta}\right)^2+\left(\frac{1}{E_1+\Delta}+\frac{1}{E_2-\Delta}\right)^2\right]$$
$$\times\{(1+z)\sqrt{z(2+z)}-\ln[1+z+\sqrt{z(2+z)}]\},\quad z\equiv eV/\Delta\,. \tag{7}$$

In particular, at $eV\ll\Delta$, Eq. (7) shows that $\gamma_{\rm in}\propto V^{3/2}$.

In contrast to inelastic tunneling, the rate $\gamma_{\rm el}$ of elastic tunneling in the double-junction system is not sensitive to the total number of electrons in the middle electrode. In the equations for $\gamma_{\rm el}$, the superconductivity of this electrode only changes the amplitude of the charge propagation, which now reads [compare to Eq. (11b) of the first paper in Ref. [13]]

$$F(\varepsilon,\varepsilon_m,\varepsilon_n)=\frac{u^2(\varepsilon)}{E_1+(\varepsilon^2+\Delta^2)^{1/2}-\varepsilon_m}-\frac{v^2(\varepsilon)}{E_2+(\varepsilon^2+\Delta^2)^{1/2}+\varepsilon_n}\,. \tag{8}$$

As in the normal system $\gamma_{\rm el}$ depends on the character of the classical electron motion in the middle electrode. In particular, when this motion is diffusion with coefficient D, and the time of diffusion across the electrode is short, $L^2/D\ll\hbar/\Delta$ (in other terms, the characteristic dimensions L of the electrode are small compared to the superconducting coherence length $\xi_0\equiv\sqrt{D\hbar/\Delta}$), one gets

$$\gamma_{\rm el}=\frac{\hbar G_1G_2\delta}{4\pi e^4}\int d\varepsilon_m\, d\varepsilon_n\, d\varepsilon\, f(\varepsilon_m)[1-f(\varepsilon_n)]|F(\varepsilon,\varepsilon_m,\varepsilon_n)|^2\delta(\varepsilon_n-\varepsilon_m-eV)\,. \tag{9}$$

Since the current associated with inelastic tunneling (7) vanishes as $V^{3/2}$ at $V\to 0$, the zero-bias conductance of the double-junction system is determined solely by elastic tunneling (9). At small temperatures, $T\ll\Delta$, this conductance is

$$G_{\rm el}=\frac{\hbar G_1G_2\delta}{8\pi e^2}\left[\sum_{j=1,2}\frac{1}{E_j(E_j^2-\Delta^2)}\{2E_j^2-\Delta^2[1+g(E_j/\Delta)]\}+\frac{2\Delta}{E_1-E_2}[g(E_2/\Delta)-g(E_1/\Delta)]-\frac{\pi}{2}\left(\frac{\Delta}{E_1}+\frac{\Delta}{E_2}\right)^2\right], \tag{10}$$

where

$$g(z)=\begin{cases}\dfrac{1}{z(z^2-1)^{1/2}}\ln\dfrac{1+z+(z^2-1)^{1/2}}{1+z-(z^2-1)^{1/2}}, & \text{for } z>1,\\[2ex] \dfrac{1}{z(1-z^2)^{1/2}}\left[\dfrac{\pi}{2}-\arctan\left(\dfrac{1+z}{1-z}\right)^{1/2}\right], & \text{for } z<1.\end{cases}$$

Equation (10) describes, in particular, the conductance due to electron tunneling through the energy barrier provided solely by the superconducting gap, when the charging energy is negligible, $E_1=E_2=0$:

$$G_{\rm el}=\hbar G_1G_2\delta/8e^2\Delta\,. \tag{11}$$

A comparison of Eqs. (7) and (10) shows that in contrast to the system with normal electrodes, $\gamma_{\rm in}$ for a superconducting middle electrode is of the same order of magnitude as $\gamma_{\rm el}$, and it is much smaller than in the normal system. As a result, the total tunneling rate through the superconducting electrode is smaller than the tunneling rate in the normal system by at least a factor of δ/Δ, which is of the order of 10^{-4} for typical experimental parameters [10,15,16].

The above results can be applied to the "single-electron box" [15,16], which is a metallic island connected to a bulk external electrode by one tunnel junction [Fig. 1(b)]. When the island is superconducting, the number N of electrons in it should change in accordance with Eq. (3) as a function of the island potential φ. However, experiments [15,16] show that N changes periodically in φ, as in the normal system with $\Delta=0$. This means that the superconductivity is not ideal [15], i.e., there is a finite subgap density of states.

One of the plausible models that can account for subgap states is a small normal inclusion in the superconductor [Fig. 1(b)]. (Another possibility, which we do not discuss here, is magnetic impurities.) Let us assume that the normal region is separated from the superconductor by a barrier with low transparency $|T|^2 \ll 1$. In this case one can describe electron tunneling to the island as the elastic tunneling process considered above, and calculate the tunneling rate from Eqs. (8)-(10). An electron tunnels to the normal region of the island through the energy barrier associated with the superconducting energy gap. If the total tunnel conductance G_2 between the normal inclusion and the superconductor is large, $G_2 \gg R_Q^{-1}$, where $R_Q = \pi\hbar/2e^2$, the charging energy of the system depends only on the total charge of the island, and it does not change when the electron tunnels inside the island. Hence, in Eqs. (8) and (9) one should take $E_2=0$ and $eV=-E_1$, where E_1 is electrostatic energy change due to the electron tunneling to the island, $E_1<0$.

As above, the rate Γ of tunneling to the island depends on the character of the electron motion in it. Under the conditions of Eq. (9), we get

$$\Gamma(\varepsilon) = \frac{\hbar G_1 G_2 \delta}{8e^4} \frac{\varepsilon}{(\Delta^2-\varepsilon^2)^{1/2}}, \tag{12}$$

where $\varepsilon \equiv -E_1 > 0$. This tunneling can be interpreted not only as a tunneling into the normal region, but also as a direct tunneling to the superconductor, where the normal inclusion induces a finite subgap density of states $\rho(\varepsilon)$:

$$\rho(\varepsilon) = \frac{e^2}{G_1\delta}\frac{d\Gamma(\varepsilon)}{d\varepsilon} = \frac{\hbar G_2}{8e^2}\frac{\Delta^2}{(\Delta^2-\varepsilon^2)^{3/2}}, \quad |\varepsilon| < \Delta. \tag{13}$$

Making use of Eq. (13) one can extend the calculations of $\Gamma(\varepsilon)$ to the situation when the external electrode of the single-electron box [Fig. 1(b)] is also a superconductor.

The density of states (13) is smaller than the density of states above the gap by a factor δ/Δ. Nevertheless, it is sufficiently large to account for the "normal" behavior of the islands in experiments with a single-electron box, since the corresponding tunneling time Γ^{-1}, with Γ given by Eq. (12), is still microscopically small for typical parameters [15,16]. In order to observe odd-even variations of the ground-state energy (2) of a superconducting island, the subgap tunneling time should be comparable to the time of the experiment. This condition can be satisfied if the distance d between the normal inclusion and the tunnel barrier is much larger than the coherence length ξ_0. In this case, one can show [substituting Eq. (8) into Eq. (11) of Ref. [13]] that Γ is exponentially small, $\Gamma \propto \exp\{-d/\xi_0\}$, so that the tunneling time Γ^{-1} can be macroscopically large.

In conclusion, we have calculated the rate of electron tunneling via virtual states of a superconducting island in the Coulomb blockade regime. This process can be responsible for a finite subgap tunneling rate into the superconductor with normal inclusion. For ideal superconductors, the tunneling rate depends on the parity of the total number of free electrons in the superconductor.

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