Coulomb Blockade of Two-Electron Tunneling

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We study the Coulomb blockade in a superconducting grain, connected to two normal electrodes by tunnel junctions. At small bias, the conductance of this system is due to electrons passing in *pairs* through the grain. The linear conductance is periodic in the gate voltage. The period and the conductance activation energy are determined by the charge $2e$, rather than e. At resonance the current first grows linearly with the applied bias and then drops as the quasiparticle transport channel opens up.

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Electron transport through conductors of small electric capacitance has been studied both experimentally and theoretically in great detail during the past years [1]. The simplest example of such a system consists of a small conducting grain, coupled to two macroscopic leads by tunnel barriers. The grain is linked capacitively to the leads (capacitors C_l, C_r in Fig. 1) as well as to a gate electrode (capacity C_g). The latter allows one to control the number N of electrons on the grain by the gate voltage V_g; transport is achieved if a bias voltage $V \equiv V_l - V_r$ is applied to the leads. For almost any value of V_g, the ground state of the grain is nondegenerate, and variations of its charge in the course of electron tunneling increase the electrostatic energy. This is why electron tunneling through a small grain is suppressed (Coulomb blockade). However, at certain values of V_g which form a periodic set with period e/C, the ground state is degenerate and the blockade is lifted (here $C = C_l + C_r + C_g$). At these values of V_g, the electrostatic energies of the system with N and $N+1$ electrons on the grain are equal. These two numbers are inevitably of different parity, which raises the question how the Coulomb blockade is modified if the grain is in the superconducting state and the electrons form Cooper pairs.

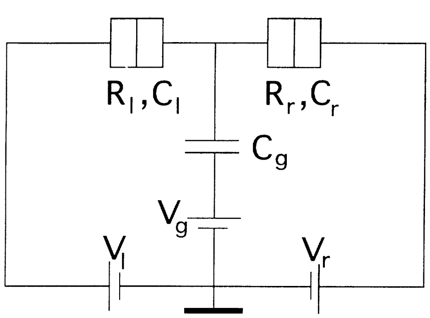

FIG. 1. Schematic representation of the double junction system.

In analogy with the pairing theory for nucleons [2], it was suggested recently [3,4] that the ground state of a superconducting grain favors even numbers of electrons, if the superconducting gap Δ exceeds the characteristic charging energy $E_c = e^2/2C$; see Fig. 2. Under these conditions the ground state of the system may no longer have the degeneracy $N \leftrightarrow N+1$. As a result, the single-electron tunneling between the leads and the superconducting grain is suppressed at low temperatures. In this paper we propose another mechanism of the electron transfer, namely the two-electron tunneling, which is not suppressed at low temperatures. This mechanism gives the resonant contribution to the tunnel current at some particular values of the gate voltage, when the ground state has the degeneracy $2n \leftrightarrow 2n+2$; see Fig. 2. The resonances are periodic in V_g with a period $2e/C_g$. The origin of these resonances is similar to that for a nonsuperconducting system [5,6], but the period is twice as large as in the normal state of the grain. The period is

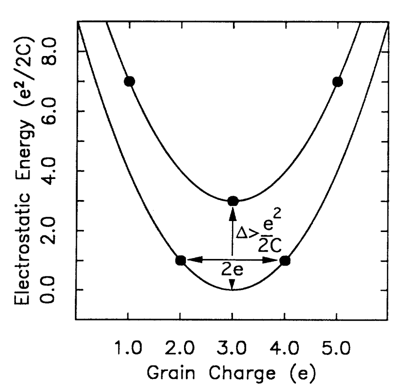

FIG. 2. Total energy of the grain as a function of the grain charge in the superconducting state. There is a degeneracy between states with an even number of electrons N and $N+2$. The state with $N+1$ electrons is shifted up by an amount $\Delta > E_c$.

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doubled because not one but two electrons are involved in the sequential (incoherent) transitions through the grain. Two electrons first tunnel into the grain and become a part of the superconducting condensate. Then, *another* pair of electrons tunnels from the condensate into the opposite lead, thus returning the grain to its initial state and finishing an elementary event of charge transfer. Because tunneling through the grain occurs as a sequence of two incoherent steps, there is no suppression of conductance due to destructive interference typical for elastic cotunneling [4,7]. As a result, two-electron tunneling provides the major mechanism of transport through a small superconducting grain. We find a conductance which is typically 2 orders of magnitude larger than the one due to cotunneling mechanisms [4]. Besides, since sequential tunneling is possible only at the two-electron degeneracy points, it is sensitive to resonant conditions. Away from these points, the conductance is exponentially small at low temperature with an activation energy twice as large as in the normal state. Transport through a superconducting grain is also suppressed if Δ is brought below $e^2/2C$, e.g., by applying a magnetic field. The doubling of activation energy and suppression of conductance at $\Delta < e^2/2C$ give other possibilities to demonstrate the $2e$ nature of charge transfer, in addition to the doubling of the period in V_g which was recently observed by Eiles, Martinis, and Devoret [8].

The process of tunneling of two electrons into the superconducting grain is very similar to Andreev reflection from the boundary between a normal metal and a superconductor. In the case of a tunnel junction connecting a normal metal with a superconductor, Andreev reflection provides a nonzero subgap conductance at small bias, when quasiparticle tunneling is suppressed [9]. We derive the amplitude of the Andreev process for tunneling into a small grain and find that charging *enhances* this amplitude when the energy E_c approaches Δ.

Charging energy reduces the threshold bias voltage V at which the Andreev process is replaced by quasiparticle tunneling into the grain. In some interval of V above the threshold only one extra electron is allowed in the grain. Being injected into a quasiparticle state, only this particular electron can leave the grain, because all other electrons are bound in pairs and belong to the condensate. This reduces the possible current through the grain strongly. The "odd" electron becomes trapped in the grain, thereby blocking the two-electron channel and thus causing a drop in the I-V characteristic. Further increase of the bias eventually allows for pair breaking and the current through the grain rapidly increases. The theory presented in this paper can explain the nonmonotonic I-V characteristic observed in Ref. [8].

In order to find the amplitude of the two-electron tunneling through a barrier separating the normal metal lead and the superconducting grain, we start with the Hamiltonian

$$\hat{H} = \hat{H}_N + \hat{H}_S + \hat{H}_T. \quad (1)$$

Here, $\hat{H}_N$ and $\hat{H}_S$ describe the lead and the superconducting grain, respectively; $\hat{H}_T$ is the tunnel Hamiltonian, which describes the transfer of electrons through the barrier. It can be conveniently expressed in terms of quasiparticle operators $\hat{\gamma}, \hat{\gamma}^\dagger$ for the superconductor, and electron operators $\hat{a}, \hat{a}^\dagger$ for the normal metal:

$$\hat{H}_T = \sum_{\mathbf{k},\mathbf{p},\sigma} \{ t_{\mathbf{kp}} \hat{a}^\dagger_{\mathbf{k},\sigma} (u_{\mathbf{p},\sigma} \hat{\gamma}_{\mathbf{p},\sigma} + v_{\mathbf{p},\sigma} \hat{\gamma}^\dagger_{-\mathbf{p},-\sigma}) + t^*_{\mathbf{kp}} (u_{\mathbf{p},\sigma} \hat{\gamma}^\dagger_{\mathbf{p},\sigma} + v_{\mathbf{p},\sigma} \hat{\gamma}_{-\mathbf{p},-\sigma}) \hat{a}_{\mathbf{k},\sigma} \}. \quad (2)$$

Here, $t_{\mathbf{kp}}$ are the tunnel matrix elements which we take to be spin independent, and $u_{\mathbf{p},\sigma}, v_{\mathbf{p},\sigma}$ are the BCS coherence factors [10]; the sum is taken over momenta $\mathbf{k}, \mathbf{p}$ and spin $\sigma = \uparrow, \downarrow$.

We calculate now the amplitude of tunneling into the grain of two electrons from states $\mathbf{k}\uparrow$, $\mathbf{k}'\downarrow$ in the lead. This amplitude $A_{\mathbf{k}\uparrow\mathbf{k}'\downarrow}$ should be found in second-order perturbation theory in $\hat{H}_T$ [11]. At temperature k_BT and applied bias voltage eV smaller than the superconducting gap Δ, the final state after tunneling cannot contain quasiparticles. Hence, the only allowed tunneling process produces an additional Cooper pair in the grain. This second-order process consists of two steps. First, one of the two electrons tunnels into the grain and forms a virtual state with a quasiparticle. Then the second one tunnels into the grain and couples with the quasiparticle to form a Cooper pair. The resulting amplitude can be expressed in terms of tunnel matrix elements and coherence factors entering the Hamiltonian (2):

$$A_{\mathbf{k}\uparrow\mathbf{k}'\downarrow} = \sum_{\mathbf{p}} t^*_{\mathbf{kp}} t^*_{\mathbf{k}'-\mathbf{p}} u_{\mathbf{p}} v_{\mathbf{p}} \left\{ \frac{1}{E_c - \varepsilon_p + \xi_k} + \frac{1}{E_c - \varepsilon_p + \xi_{k'}} \right\}. \quad (3)$$

Here the spin dependence of the coherence factors was dropped after using the relation $v_{\mathbf{p},\uparrow} = -v_{-\mathbf{p},\downarrow}$. The denominators in (3) contain the energy of the virtual state counted from the energy of the initial state. Because the transfer of an electron into a quasiparticle state on the grain is accompanied by a change of the grain charge, these denominators include the Coulomb energy term $E_N - E_{N+1} = E_c$ along with the standard electron and quasiparticle energies ξ_k and $\varepsilon_p = \sqrt{\Delta^2 + \xi_p^2}$. We introduced the electrostatic energy E_N for N electrons on the grain,

$$E_N = \frac{(Ne)^2}{2C} + \frac{Ne}{C}(C_l V_l + C_r V_r + C_g V_g),$$

where the indices l, r, and g refer to the left, right, and gate electrode, respectively (see Fig. 1). We finally neglect the electron energies $\xi_k \approx \xi_{k'} \approx 0$ at small $k_BT, eV \ll E_c < \Delta$; substitution of E_c for $E_N - E_{N+1}$ is valid near the resonance ($E_N \simeq E_{N+2}$). After replacing the product of tunnel matrix elements by the average over directions of $\mathbf{p}$, the summation in (3) can be performed, and we find

$$A_{\mathbf{k}\uparrow\mathbf{k}'\downarrow} = -\frac{2\nu_g \Delta \langle t^*_{\mathbf{kp}} t^*_{\mathbf{k}'-\mathbf{p}}\rangle_{\mathbf{p}}}{\sqrt{\Delta^2 - E_c^2}} \arctan\sqrt{\frac{\Delta + E_c}{\Delta - E_c}}, \quad (4)$$

where ν_g is the grain density of states; brackets $\langle\cdots\rangle_{\mathbf{p}}$ denote averaging over the directions of $\mathbf{p}$.

The rate Γ_l for the scattering of two electrons from the left metal into the grain can be expressed as

$$\Gamma_l(\epsilon_l) = \frac{2\pi}{\hbar}\sum_{\mathbf{k},\mathbf{k}'} |A_{\mathbf{k}\uparrow\mathbf{k}'\downarrow}|^2 f_l(\xi_k) f_l(\xi_{k'}) \delta(\xi_k + \xi_{k'} - \epsilon_l). \quad (5)$$

It contains the Fermi functions f_l for electrons with energies ξ_k, $\xi_{k'}$ in the left metal; energy conservation implies that the sum of these energies equals the energy ϵ_l needed to transfer two electrons from the left lead to the superconductor: $\epsilon_l = E_{N+2} - E_N - 2eV_l$. At zero charging energy, formula (5) gives the known [9] rate of two-electron tunneling between a bulk superconductor and a normal metal. Charging of the superconducting grain affects this rate in two ways. First, the "strip" of energies ϵ_l from which normal electrons tunnel into the grain is modified. Second, the amplitude $A_{\mathbf{k}\uparrow\mathbf{k}'\downarrow}$ itself depends on E_c. Note that Coulomb energy enhances the tunneling, and $A_{\mathbf{k}\uparrow\mathbf{k}'\downarrow}$ diverges [12] while E_c approaches Δ; see (4). The rate of tunneling from the right normal lead $\Gamma_r(\epsilon_r)$ is obtained from (5) by replacing indices $l \to r$.

The dependence of rates $\Gamma_l(\epsilon_l)$, $\Gamma_r(\epsilon_r)$ on energies is similar to that found for a normal system [6],

$$\Gamma_i(\epsilon_i) = \frac{2\pi}{\hbar}\gamma_i \frac{\epsilon_i}{\exp(\epsilon_i/k_BT) - 1}, \qquad i = l, r. \quad (6)$$

The only difference is in the definition of energy ϵ_i: it corresponds now to the transfer of *two* electrons through a junction in each completed act of tunneling. The dimensionless parameter γ_i results from the averaging of $|A_{\mathbf{k}\uparrow\mathbf{k}'\downarrow}|^2$ over the directions of momenta:

$$\gamma_i = \frac{1}{N_i}\frac{G_i^2}{(2\pi e^2/\hbar)^2}\frac{4\Delta^2}{\Delta^2 - E_c^2}\left\{\arctan\sqrt{\frac{\Delta + E_c}{\Delta - E_c}}\right\}^2. \quad (7)$$

Here G_i is the conductance of the left ($i = l$) or right ($i = r$) tunnel junction. The factor $1/N_i$ accounts for the particular geometry of the junction,

$$N_i = \langle |t_{\mathbf{kp}}|^2\rangle^2_{\mathbf{kp}} / \langle |\langle t_{\mathbf{kp}} t_{\mathbf{k}'\mathbf{p}}\rangle_{\mathbf{p}}|^2\rangle_{\mathbf{kk}'}.$$

For a point tunnel junction $t_{\mathbf{kp}}$ is a constant and $N_i = 1$. For a wide junction of area S_i, parameter N_i has the meaning of the number of electron modes effectively penetrating the barrier, $N_i \sim k_F^2 S_i$.

Equation (6) allows us to derive the linear conductance G through a superconducting grain. At low temperatures $k_BT \sim |E_N - E_{N+2}| \ll E_c, (\Delta - E_c)/\ln(\nu_g\Delta)$ (the latter energy is related to thermally induced quasiparticles, see below) and small bias $eV \ll k_BT$ we find

$$G = \frac{4\pi e^2}{\hbar}\frac{\gamma_l\gamma_r}{\gamma_l + \gamma_r}\frac{2eC_g(V_g - V_g^{(N)})/Ck_BT}{\sinh\{2eC_g(V_g - V_g^{(N)})/Ck_BT\}}. \quad (8)$$

Here $V_g^{(N)} = -(N+1)e/C_g$ is the gate voltage at which the resonance is reached. The dependence of G on temperature and V_g is very similar to the one for a normal-state grain [6]. However, according to (8) the period in V_g and the conductance activation energy at fixed value of $V_g - V_g^{(N)}$ are twice as large as in the normal grain case. Measurements of the period in V_g and of the activation energy of the conductance can be used to demonstrate the two-electron nature of the charge transport. The first type of measurement was performed recently [8].

The linear conductance (8) vanishes at zero temperature if the gate voltages $V_g \neq V_g^{(N)}$; two-electron tunneling occurs only at a bias above certain threshold. In the vicinity of a resonance the nonlinear I-V characteristic is determined by the transitions between two adjacent charge states N, $N+2$. The current is determined by the rates $\Gamma_l(\epsilon_l)$ and $\Gamma_r(-\epsilon_r)$ of these transitions,

$$I = 2e\frac{\Gamma_l(\epsilon_l)\Gamma_r(-\epsilon_r)}{\Gamma_l(\epsilon_l) + \Gamma_r(-\epsilon_r)}. \quad (9)$$

From the expression for the rates (6) we see that the current (9) is nonzero only if $\epsilon_l < 0$, such that the grain can be charged by two electrons, and simultaneously $\epsilon_r > 0$, which enables the decharging process. At a bias voltage $V = V_l - V_r$ this implies

$$V_r - \frac{C_l}{C_g}V < V_g - V_g^{(N)} < V_l + \frac{C_r}{C_g}V.$$

In a symmetric setup ($C_l = C_r$, $\gamma_l = \gamma_r = \gamma$) with $V_r \equiv 0$, the current resonance has the form

$$I(V_g) = \frac{8\pi e^2}{\hbar}\frac{\gamma}{V}\left[\frac{V^2}{4} - \frac{C_g^2}{C^2}\left(V_g - V_g^{(N)} - \frac{V}{2}\right)^2\right]. \quad (10)$$

Note that both the width and the height $I_{\max} = (2\pi e^2/\hbar)\gamma V$ of the peak $I(V_g)$ depend linearly on bias voltage, which is true not only for the symmetric setup. This agrees with the experimental results obtained by Eiles, Martinis, and Devoret [8].

According to (10), at resonance the current is proportional to the applied bias. This proportionality holds as long as electrons can only traverse the grain in pairs. At larger bias, when quasiparticle tunneling into the grain is allowed, the transport mechanism changes abruptly. [For instance, in a circuit with $V_r = 0$, Fig. 1, the threshold voltage at which this change occurs is $V_{\text{th}} = (\Delta - E_c)C/(C_g + C_r)e$.] At voltages above the threshold, a single electron can tunnel into the grain with a rate of the order of $w_{\text{qp}} \sim (G_l/e)[V^2 - V_{\text{th}}^2]^{1/2}$ (cf. [10]; we assume that the excess of V above the threshold is small). The escape rate of this quasiparticle from the grain into the opposite lead (r) is quite different. The reason is that *any* electron with an above-threshold energy in the

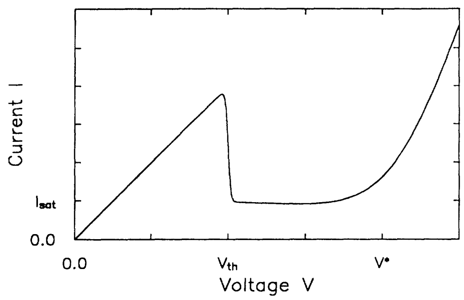

FIG. 3. Overall I-V characteristic for the double junction system with a superconducting grain. As a result of two-electron tunneling, the current starts linearly in V. At voltages $V > V_{\rm th}$, a quasiparticle can get trapped in the grain which reduces the current to the value $I_{\rm sat}$. At large enough bias $V > V^*$, transport is achieved by the usual quasiparticle current.

normal lead is available for tunneling into the grain, but only *one* unpaired electron can tunnel out of the grain. The escape rate $w_{\rm esc}$ is determined by the tunnel width of a discrete energy level available for the "odd" electron in the grain. Although the widths fluctuate from level to level, we can estimate the typical width $\hbar w_{\rm esc}$ through its relation to the conductance: $G_r \sim e^2\nu_g w_{\rm esc}$. Comparison of $w_{\rm qp}$ with $w_{\rm esc}$ shows that $w_{\rm qp}$ is the larger one if the bias exceeds the threshold voltage by a value as little as $1/\nu_g e$, corresponding to the spacing between discrete levels in the grain. At higher bias, the probability of having an odd number of electrons $N+1$ is the dominant one. Current through the grain is determined by the escape rate of the quasiparticle and saturates at the low level $I_{\rm sat} \sim G_r/\nu_g e$. The density of states of the grain is proportional to its volume: the larger the grain the smaller the current $I_{\rm sat}$. The current drop at the threshold voltage is determined by the ratio of $I_{\rm sat}$ to the two-electron current (10) and is substantial if the junction conductances are not too low: $G_{l,r} \gtrsim (e^2/h)(S_{l,r}/S_g)$. Here S_g and $S_{l,r}$ are the cross-sectional areas of the grain and the junctions, respectively. For the geometry of [8] the restriction on $G_{l,r}$ requires $1/G_{l,r} \lesssim 10$ MΩ, which was definitely satisfied in the experiment. We believe that the described mechanism of switching from two-electron processes to the "escape" current can explain the minimum in I-V characteristic observed by Eiles, Martinis, and Devoret [8].

At even higher bias, electrons belonging to the condensate can tunnel from the grain, leaving behind excitations. As a result, the usual quasiparticle transport channel opens up, and at $eV > eV^* \equiv (\Delta + E_c)C/C_l$ current starts to grow rapidly with bias. The overall I-V characteristic is sketched in Fig. 3.

Nonzero temperature makes the switching into the state with an odd number of electrons on the grain easier. Thus, at finite T the threshold voltage decreases approximately according to the following formula:

$$V_{\rm th} = \frac{C}{(C_g + C_r)e}[\Delta - E_c - k_B T \ln(\nu_g \Delta)],$$

and vanishes at $k_B T \simeq (\Delta - E_c)/\ln(\nu_g\Delta)$.

In conclusion, electron transport through a small superconducting grain under the conditions of Coulomb blockade is studied. We show that the major mechanism of transport at small bias is Andreev reflection [9] modified significantly by a finite charging energy. The dependence of the linear conductance on gate voltage and temperature is clearly related to the process of charge transfer by electron *pairs*. The nonlinear I-V characteristic may show a pronounced minimum due to a very specific mechanism of quasiparticle trapping in the grain that changes the parity of the number of electrons in the grain. Trapping is possible only due to the combined effect of superconductivity and Coulomb blockade.

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[12] In a narrow region $|E_c - \Delta| \sim (\hbar G/e^2)\Delta$ the higher-order perturbation theory should be employed.