# Theoretical Physics Institute University of Minnesota

TPI-MINN-94/39-T November '94

## Andreev Spectroscopy of Josephson Coupling

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#### Abstract

It is shown that the low energy spectrum of mesoscopic superconductors coupled by Josephson interaction can be probed by two-electron tunneling from a normal electrode. The Andreev reflection in the NS junction of a normal-superconductor-superconductor double junction (NSS transistor) provides a unique spectroscopic tool to probe the coherent Cooper pair tunneling and the energy spectrum of the Josephson (SS) junction. The ground state properties are reflected in a resonant structure of the linear conductance; excited states with an energy as low as the Josephson coupling energy lead to a threshold in the nonlinear I-V characteristic.

74.50.+r, 74.20.Fg, 72.10.Bg

Typeset using REVT<sub>E</sub>X

The quantum states of a mesoscopic conductor (grain) connected by weak links to particle reservoirs are significantly influenced by charging effects. The charging energy restricts tunneling of electrons between the grain and the reservoirs. At a discrete, periodic set of values of the electrostatic potential of the grain, which can be tuned continuously by means of a gate, states with different grain charges may have the same electrostatic energy. In the case of normal metals, many-body effects lead to a non-analytical variation of the grain charge with gate voltage in the vicinity of these degeneracy points [1,2]. However, this behavior develops only at very low temperatures, and has not been observed experimentally so far. The chances for the observation of coherent charge transfer are larger if the grain and particle reservoirs are in the superconducting state. Here the nature of the degeneracy depends on the relative magnitude of the charging energy  $E_C$  and the superconducting gap  $\Delta$ . If  $\Delta < E_C$  the degeneracy occurs between states having N and N + 1 electrons on the grain. In the opposite regime, states with the numbers N and N+2 may be degenerate and coherent Cooper pair tunneling may become observable [3]. The crossover between the two regimes was studied by measurements of the charge in an electron box [4] and of the switching current in a Josephson double-junction [5].

The possibility of superpositions of different charge states in a superconductor gives rise to a number of interesting macroscopic quantum effects [6–9]. Measurements of the island charge at low temperature and of the critical Josephson current provide information about the ground state energy of the system [3], but leave inaccessible the energy spectrum and structure of excited states [6–8]. On the other hand, for  $\Delta > E_C$ , the interplay between the Josephson coupling and charging leads to quantization of this spectrum. At the points of charge degeneracy the spacing between the corresponding discrete levels is of the order of the Josephson coupling energy  $E_J$ , which for a weak link is much smaller than the gap  $\Delta$ . This hierarchy of energies should allow one to resolve the low-energy spectrum, if a suitable spectroscopic tool is found.

In this paper we show that the I-V characteristic of a double tunnel junction connecting a superconducting lead with a normal lead through a mesoscopic superconducting grain, provides the needed spectroscopic tool. In the low-bias regime, the dominant mechanism of transport in the NS junction between the grain and the normal electrode is due to Andreev reflection. Under these conditions we find that the linear conductance is sensitive to the superposition of different charge states in the wave functions of the Josephson junction formed by the grain with the superconducting electrode. Specifically, near a degeneracy point we find that the conductance has a resonant shape as a function of the electrostatic potential of the grain. This resonance is directly related to the amplitudes of the states with N and N + 2 electrons on the grain involved in the superposition. In addition, the nonlinear I-V characteristic displays a threshold voltage that reveals the existance of an excited state with an energy on the order of  $E_J$ . At the threshold voltage, this state will be engaged in electron transport, and as a result the conductance of the system will increase. The value of the threshold voltage enables one to identify the energy of the excited state.

We consider a NSS transistor with a small superconducting island coupled via tunnel junctions to a normal left (L) electrode and a superconducting right (R) electrode. A gate at potential  $V_g$  is coupled capacitively to the island, and a bias voltage  $\pm V/2$  is applied symmetrically to the electrodes (V > 0). The charging energy of the transistor depends on the number of electrons N on the island, and also on the number of electrons  $N_R - N_L$  which have passed through the transistor,

$$E_{ch}(N_L, N, N_R) = E_C \left(\frac{Q_g}{e} - N\right)^2 - \frac{(N_R - N_L)}{2} eV .$$
 (1)

Here  $C = C_L + C_R + C_g$  is the total capacitance of the island, i.e., the sum of the two junction capacitances and the gate capacitance, and  $E_C = e^2/2C$  is the charging energy corresponding to a single electron. The effect of the gate voltage is contained in the external charge  $Q_g = C_g V_g + (C_L - C_R)V$ . All the properties of the transistor are 2*e*-periodic in the external charge. We, therefore, can restrict ourselves to the range  $0 \le Q_g \le 2e$ .

The states of the system  $|N_L, N, N_R\rangle$  are characterized by the number of charges in the electrodes and the island,  $N_L, N_R$ , and N, and furthermore, if present, by the energies of the excitations in the leads and the island. We start from a reference state without quasiparticle excitations, which we denote by  $|0,0,0\rangle$  with charging energy  $E_{ch}(0,0,0) = Q_g^2/2C$ . Other states participating in the two-electron transfer (Andreev reflection in the left junction and pair tunneling in the Josephson junction) are  $|0,2,-2\rangle$ ,  $|-2,0,2\rangle$ , and  $|-2,2,0\rangle$ . Their energies can be found from Eq. (1). The Andreev reflection process involves an intermediate state  $|-1_k, 1_p, 0\rangle$ , where one electron has been transferred through the NS junction onto the grain. The energy of this state is  $E_{kp} = E_{ch}(-1,1,0) - \xi_k + \epsilon_p$ , where  $\xi_k$  and  $\epsilon_p$  are the quasiparticle energies in the normal and the superconducting grain respectively.

The Josephson coupling, which is characterized by the energy scale  $E_J$ , mixes charge states differing by multiples of Cooper pairs in the two superconducting electrodes. If  $Q_g$ takes a value near e, and the bias V is small, the states  $|0,0,0\rangle$  and  $|0,2,-2\rangle$  are nearly degenerate and hence get mixed strongly. Similarly  $|-2,0,2\rangle$  and  $|-2,2,0\rangle$  are mixed. Near the degeneracy points the eigenstates are a superposition of two states

$$\psi_0 = \alpha |0, 0, 0\rangle + \beta |0, 2, -2\rangle,$$
  

$$\psi_1 = -\beta |0, 0, 0\rangle + \alpha |0, 2, -2\rangle$$
(2)

with energies

$$E_{0(1)} = E_C + \frac{(Q_g - e)^2}{2C} + \frac{eV}{2} - (+)\frac{1}{2}\sqrt{\delta E_{ch}^2 + E_J^2} .$$
(3)

Here the coefficients are

$$\alpha^{2} = 1 - \beta^{2} = \frac{1}{2} \left[ 1 + \frac{\delta E_{ch}}{\sqrt{\delta E_{ch}^{2} + E_{J}^{2}}} \right] , \qquad (4)$$

and we introduced the difference in charging energy,  $\delta E_{ch} \equiv E_{ch}(0,2,-2) - E_{ch}(0,0,0)$ , which is  $\delta E_{ch} = 4E_C \left(\frac{Q_g}{e} - 1\right) + eV$ . The coefficient  $\alpha$  is close to unity if the charging energy of the state  $|0,0,0\rangle$  lies below that of  $|0,2,-2\rangle$ , i.e. for  $\delta E_{ch} > 0$ , and vanishes in the opposite limit, while  $\beta$  shows the complementary behavior. The Josephson mixing of the other two states leads to the following eigenstates

$$\psi_0' = \alpha |-2, 0, 2\rangle + \beta |-2, 2, 0\rangle , \psi_1' = -\beta |-2, 0, 2\rangle + \alpha |-2, 2, 0\rangle .$$
(5)

The coefficients  $\alpha$  and  $\beta$  are the same as for the first pair, and the corresponding energies are

$$E'_{0(1)} = E_{0(1)} - 2eV . (6)$$

In the above consideration we neglected the effect of charging on the Josephson coupling constant that leads to a relatively small enhancement [8,10] of  $E_J$ .

At low bias voltages the dominant process for charge transfer across the NS junction is Andreev reflection. Generalizing the expression derived in Ref. [11] for a NSN transistor, we can write the amplitude for this second order tunneling process between the states  $\psi_0$  and  $\psi'_0$  as

$$A_{k,k'}(\psi_0 \to \psi'_0) = \alpha \beta \sum_p t^*_{kp} t^*_{k'-p} u_p v_p \\ \times \left( \frac{1}{E_0 - E_{kp}} + \frac{1}{E_0 - E_{k'p}} \right).$$
(7)

In this process two electrons from the states  $\mathbf{k}, \uparrow$  and  $\mathbf{k}', \downarrow$  of the normal electrode tunnel into the grain through a junction. The latter is characterized by the tunneling Hamiltonian with matrix elements  $t_{kp}$ . We suppressed the spin indices, and used the relation  $v_{p,\uparrow} = -v_{p,\downarrow}$  between the coefficients of the Bogoliubov transformation. The energies of the virtual intermediate states  $E_{kp}$  and  $E_{k'p}$  where **one** electron has been transferred to the island enter the denominators. The summation in Eq. (7) can be performed and, for  $\xi_k, \xi_{k'} \to 0$ , yields the result

$$A_{k,k'}(\psi_0 \to \psi'_0) = \alpha \beta \frac{\pi}{2} \nu F_0 \langle t^*_{kp} t^*_{k'-p} \rangle_{\dot{p}} .$$

$$\tag{8}$$

This expression involves the density of states of the island  $\nu$ , and an average over the directions of the momenta  $\hat{p}$ . We introduced the function

$$F_{0} \equiv \frac{4}{\pi} \frac{\Delta}{\sqrt{\Delta^{2} - [E_{ch}(-1,1,0) - E_{0}]^{2}}} \times \arctan \sqrt{\frac{\Delta - E_{ch}(-1,1,0) + E_{0}}{\Delta + E_{ch}(-1,1,0) - E_{0}}}.$$
(9)

The rate for the Andreev reflection process is obtained by the golden rule. After summation over the initial states  $\mathbf{k}$  and  $\mathbf{k}'$  one finds for low temperatures [11]

$$, \quad (\psi_0 \to \psi'_0) = \frac{2\pi}{\hbar} (\alpha\beta)^2 \frac{(G_n R_K)^2}{16\pi^2 N_{eff}} F_0^2 (E_0 - E'_0) \Theta(E_0 - E'_0) . \tag{10}$$

Here  $G_n$  is the normal state conductance of the NS junction,  $R_K = h/e^2 \approx 25.8k\Omega$  is the quantum resistance ( $G_n R_K \ll 1$ ), and  $N_{eff}$  is the effective number of parallel channels in the tunnel junction [11,12]. These parameters are conveniently absorbed in the definition of the Andreev conductance,  $G_A = G_n^2 R_K / N_{eff}$ . If the applied bias V is below some threshold

voltage  $V_{th}$ , the only transition possible at low temperatures is the Andreev reflection between the states  $\psi_0$  and  $\psi'_0$ . The resulting current  $I_{res} = 2e$ ,  $(\psi_0 \rightarrow \psi'_0)$ , due to the overlap of the functions  $\alpha$  and  $\beta$ , shows a pronounced structure as a function of gate charge, typical for resonant tunneling. This is most clearly seen in the dependence of the linear conductance on the gate charge

$$G_{res}(Q_g) = G_A \frac{E_J^2}{16E_C^2 (Q_g/e - 1)^2 + E_J^2} \frac{F_0^2}{4} .$$
(11)

The result is illustrated by the lower set of curves in Fig. 1. The width of the resonance is characterized by  $E_J$ . On this energy scale, as long as  $\Delta - E_C \gtrsim E_J$ , the function  $F_0$  can be considered constant.

Eq. (11) was derived under the assumption that the energy  $\Delta + E_{ch}(-1, 1, 0)$  of the intermediate state lies above  $E_0$ . The resonant shape changes drastically if the superconducting gap is lowered, such that these two energies can coincide. This occurs at two values of charge,  $Q_g = e \pm \delta Q_g^*$ , where  $\delta Q_g^*$  is

$$\frac{\delta Q_g^*}{e} = \frac{1}{2} \sqrt{\left(1 - \frac{\Delta}{E_C}\right)^2 - \left(\frac{E_J}{2E_C}\right)^2} \,. \tag{12}$$

If  $Q_g$  lies within the window  $e - \delta Q_g^* \leq Q_g \leq e + \delta Q_g^*$ , the ground state is no longer composed of even-charge states; rather the state  $|-1, 1_p, 0\rangle$  with one electron transferred through the NS junction has the lowest energy [4,13]. In the odd state the current is low; the maximal conductance  $G_{res}(e)$ , see Eq. (11), is not reached. Instead, within the abovementioned charge window, the conductance assumes the low value  $G \sim G_A (G_n R_K)^2$  determined by higher order tunneling processes through both junctions [14].

Even at  $\Delta > E_C - E_J/2$ , Andreev reflection leads solely to transitions between the states  $\psi_0$  and  $\psi'_0$  only, as long as the bias voltages are below a certain threshold. This threshold voltage is determined by the condition  $V_{th} = (E_1 - E_0)/2e$ . Its smallest value is  $V_{th} = E_J/2e$ . If the junction capacitances are equal,  $C_L = C_R$ , we find

$$eV_{th} = \frac{4}{3}E_C\left(\frac{Q_g}{e} - 1\right) + \sqrt{\left[\frac{8}{3}E_C\left(\frac{Q_g}{e} - 1\right)\right]^2 + \frac{1}{3}E_J^2}.$$
 (13)

At higher voltages,  $V > V_{th}$ , Andreev reflection processes can also lead to transitions between the other states introduced above. In addition to the transition,  $(\psi_0 \rightarrow \psi'_0)$  we find

$$, (\psi_{0} \to \psi_{1}') = \alpha^{4} \frac{G_{A}}{4e^{2}} F_{0}^{2} \left[ 2eV - (E_{1} - E_{0}) \right] \Theta (V - V_{th}) ,$$
  

$$, (\psi_{1} \to \psi_{0}') = \beta^{4} \frac{G_{A}}{4e^{2}} F_{1}^{2} \left[ 2eV + (E_{1} - E_{0}) \right] ,$$
  

$$, (\psi_{1} \to \psi_{1}') = (\alpha\beta)^{2} \frac{G_{A}}{4e^{2}} F_{1}^{2} 2eV .$$
(14)

The function  $F_1$  is defined similar to  $F_0$ , but with the energy of the initial state  $E_0$  replaced by  $E_1$ . Both  $F_0$  and  $F_1$  are approximately constant on the energy scale  $E_J$  if  $\Delta - E_C \gtrsim E_J$ . A master equation yields the probabilities  $W_1$ ,  $W_0$  for the system to be in the excited and ground state, respectively. I.e.,

$$W_{1} = \frac{, (\psi_{0} \to \psi_{1}')}{, (\psi_{0} \to \psi_{1}') + , (\psi_{1} \to \psi_{0}')} \Theta(V - V_{th}),$$
  

$$W_{0} = 1 - W_{1}.$$
(15)

The current then is

$$I = 2e[, (\psi_0 \to \psi'_0) +, (\psi_0 \to \psi'_1)]W_0 + 2e[, (\psi_1 \to \psi'_1) +, (\psi_1 \to \psi'_0)]W_1.$$
(16)

The threshold dependence of the probability  $W_1$  on  $V - V_{th}$  allows one to determine the Josephson energy  $E_J$ . This is illustrated with the help of Fig. 1. As long as the applied bias eV is smaller than  $E_J/2$ , which is the smallest value of  $eV_{th}$ , the gate voltage dependence of the differential conductance G = dI/dV shows the resonance (11) (two lower curves in Fig. 1). As soon as  $eV > E_J/2$ , the probability  $W_1$  can become nonzero, and an additional channel for charge transfer opens up at two values of  $Q_g$  (cf. Eq. (13)). This leads to a stepwise change of the conductance (two upper curves in Fig. 1). The smallest bias voltage at which these jumps occur enables one to determine  $E_J$ . The magnitude  $\delta G_{\pm}$  of the two jumps depends on the applied bias. If  $C_L = C_R$  we find

$$\delta G_{\pm}(V) = G_A \times \left\{ 1 \pm \left[ 1 + \frac{1}{2} \left( \frac{E_J}{2eV} \right)^2 \right] \sqrt{1 - \left( \frac{E_J}{2eV} \right)^2} \right\} \frac{F_1^2}{2}.$$
(17)

Another way to detect the Josephson energy is a measurement of the differential conductance as a function of the bias voltage V at a fixed gate-potential, illustrated in Fig. 2. Depending on the value of  $Q_g$ , a jump occurs at a certain threshold  $V_{th}$ , cf. Eq. (13); the magnitude  $\delta G(V_{th})$  is given by Eq.(17).

The results obtained above can be summarized with the help of Fig. 3, where we plotted the ratio I/V as a function of both V and  $Q_g$ . The superposition of the macroscopically different charge states of the grain lead to the current resonance as a function of the gatecharge near the degeneracy point  $Q_g = e$ . The sudden increase of the current at a certain threshold bias reflects the presence of a low-lying state with an energy  $\sim E_J$ , which, when it is excited, opens up an additional transport channel.

Spectroscopy of the Josephson coupling is only possible as long as real single electron tunneling processes are suppressed. Similar to the case of an NSN junction [11], the Andreev reflection can get "poisoned" once such processes become possible. Poisoning occurs at a certain threshold bias  $V_{poison}$ , at which the energy of the odd-charge state is lower than  $E_0$ . If the applied bias eV exceeds  $eV_{poison}$  by an amount as little as the level spacing of the grain, the probability  $W_o$  for the system to be in an odd charge state is approximately unity and current will drop substantially. Therefore, for the spectroscopy of the Josephson coupling, the condition  $V_{th} < V_{poison}$  must be met. At resonance  $Q_g \simeq e$  this condition is satisfied if  $\Delta > E_C + E_J/2$ .

The resonance structure in the  $I(Q_q)$ -dependence and the threshold structure in the I(V)-dependence is most pronounced if the temperature is smaller than the width of these features, which is given by the Josephson coupling energy  $E_J$ . In addition to intrinsic thermal fluctuations there exist fluctuations due to the external electrodynamic environment. They persist down to the lowest temperatures, causing voltage fluctuations proportional to the resistance of the external circuit. Therefore, a low impedance environment creates the most favorable conditions for the observation of the effects discussed here. It is also essential that there are no quasiparticles in the grain. This requires a sufficiently large superconducting gap  $\Delta > E_C + E_J/2$ , a perfect BCS density of states, and not too large bias voltages. In exchange for these restrictions we have a well controlled theory and unambiguous predictions for experiments. We can mention that coherent tunneling of Cooper pairs plays a role in a number of effects. Some examples are the gate voltage dependence of the critical current of SSS transistors [5,10], and the resonant Cooper pair tunneling at finite bias voltage [9,15]. The latter effect is controlled by quasiparticle-induced dissipation or influence of the environment. The spectroscopy of coherent mixing by Andreev reflection offers significant advantages over these examples. It is more straightforward than inferring the critical current from the runaway value [5], and requires only a direct measurement of the *I-V* characteristic in a convenient regime of subgap voltages. In contrast to the experiments of Haviland et al. [15], the suggested method does not rely on the fluctuations due to the hardly controllable electrodynamic environment of the junctions.

In conclusion, we studied the low energy spectrum of a NSS transistor in the presence of charging effects. The charging energy restricts the fluctuations of the number of electrons on the grain. In the vicinity of a degeneracy point, only two charge states with N and N + 2 electrons on the grain arise if  $\Delta > E_C$ . The Josephson coupling between the grain and the superconducting electrode leads to two new eigenstates that are superpositions of these, macroscopically different, charge states of the grain. The separation between the corresponding two discrete energy levels is on the order of the Josephson coupling energy  $E_J$ . We have shown how Andreev reflection in the NS-junction can probe the eigenstates. First, the resulting linear conductance at low bias shows a resonance as a function of the gate-voltage near the degeneracy point. This resonance is directly related to the amplitudes of the charge states involved in the ground state wave function. Second, a finite bias applied to the junction allows one to engage the second discrete energy level into electron transport. The corresponding threshold bias gives direct information about the discrete energy spectrum of a mesoscopic Josephson junction.

This work is supported by "Sonderforschungsbereich 195" of the Deutsche Forschungsgemeinschaft, by NSF Grant No. DMR-9423244, and by the Netherlands Organization for Scientific Research (NWO). One of the authors (LG) acknowledges the hospitality of the University of Karlsruhe where part of this work has been performed.

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#### FIGURES

FIG. 1. Differential conductance  $G/G_A$  of a NSS transistor as a function of the gate charge  $Q_g/e$ , for various values of  $eV/E_C = .047, .049, .051, .053$  (from bottom to top, curves off-set for clarity). The parameters are:  $E_J/E_C = 0.1$ ,  $C_L = C_R$ , and  $\Delta$  is large compared to  $E_C$ . The two lower curves ( $eV < E_J/2$ ) correspond to the resonance (11), the upper curves ( $eV > E_J/2$ ) show how threshold processes affect this resonance.

FIG. 2. Differential conductance  $G/G_A$  as a function of bias voltage  $eV/E_C$ , for various values of  $Q_g/e = .99, 1.0, 1.01$  (from bottom to top, curves off-set), parameters as in Fig. 1. A jump occurs at  $V = V_{th}$ , Eq. (13). Its magnitude is given by Eq. (18):  $\delta G_+$  if  $Q_g/e > 1 - eV_{th}/4E_C$ , and  $\delta G_-$  in the opposite limit.

FIG. 3. Dimensionless conductance  $I/(G_A V)$  as a function of both  $eV/E_C$  and  $Q_g/e$ . Parameters as in Fig. 1.