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# Even-odd symmetry breaking in the NSN Coulomb blockade electrometer

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We have measured at low temperature the current through a submicrometer superconducting island connected to two normal metal leads by ultra-small tunnel junctions. As the bias voltage is lowered below the superconducting gap of this Coulomb blockade electrometer, the current changes from being e-periodic with gate charge to 2e-periodic. We interpret the 2e-periodic current at low voltages as a manifestation of a sequence of Andreev reflections which shuttles two electrons at a time through the island. This process can only exist if the island favors a state with a definite parity of the number of conduction electrons.

## 1. Introduction

It is well known that systems with a small number of particles, like atomic nuclei, behave in a very different way if the number of particles is even or odd [1]. Nevertheless, one is inclined to believe that the macroscopic properties of a large isolated metallic electrode or "island" should not depend on the even or odd character of the number of its conduction electrons. There is one case, however, where the oddness or evenness of electron number would in principle show up at the macroscopic level. According to the BCS theory, electrons in a superconductor tend to form Cooper pairs. But an isolated superconductor with an odd number of electrons cannot have all its electrons paired. In the ground state, one electron would remain as a quasi-particle excitation, which has an energy corresponding to the superconducting energy gap. On the contrary, if the number of electrons were even, the ground state would be free of such excitation. This simple picture may not be applicable to a real superconducting electrode:

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the presence of only one quasiparticle state located within  $k_{\rm B}T$  of the Fermi level would restore the even-odd symmetry at an arbitrary low temperature T. A very small but finite density of states below the superconducting gap would be undetectable in usual tunneling experiments. In this paper, we report novel experimental results supporting the simple picture of even-odd symmetry breaking in an isolated superconductor, at least when time scales of the order of an hour are considered.

# 2. The NSN Coulomb blockade electrometer

Our experiment uses the property, well established experimentally [2] and theoretically [3], that a tunneling current passing through an island in the Coulomb blockade regime (i.e.  $k_B T \ll E_c = e^2/2C_\Sigma$ ,  $C_\Sigma$  being the total island capacitance) depends on the total energy difference between the ground states of the island differing by one charge carrier. We connect the island to two normal metal leads via tunnel junctions (insets of fig. 1). A voltage difference V is applied to the leads. The potential of the island and hence, its number of electrons, can be

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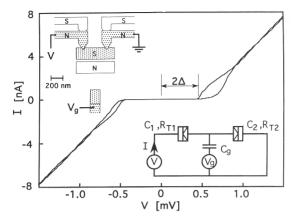


Fig. 1. Current through NSN Coulomb blockade electrometer as a function of bias voltage for two values of the gate voltage giving a minimum and a maximum gap for positive voltage. Arrow indicates the value of  $2\Delta/e = 470~\mu V$ . Lower inset shows electrical schematic for the electrometer, where the boxed symbols represent ultrasmall tunnel junctions. Upper inset depicts device layout. Normal and superconducting metallic regions are indicated by N and S respectively. The shaded regions indicated metal used for the NSN electrometer. The white regions are extraneous copies inherent to the double-angle shadow mask evaporation technique. Their sole effect is to contribute to the capacitances between the active shaded regions. The gate is actually 3  $\mu m$  away from the island.

varied independently of the potential of the leads by applying a "gate" voltage  $V_{\rm g}$  to a metallic finger capacitively coupled to the island.

If the island were in the normal state, it would have an equilibrium energy at V=0 given by  $E_n=E_{\rm c}(n-C_{\rm g}V_{\rm g}/e)^2$  where  $C_{\rm g}$  is the gate capacitance between finger and island (see fig. 2(a)). Normally at temperature T such that  $k_{\rm B}T\ll E_{\rm c}$ , there is no conduction at vanishingly small voltages V, but for gate voltages satisfying  $C_{\rm g}V_{\rm g}=(n+1/2)e$ , one has  $E_n=E_{n+1}$  and conduction is restored (fig. 2(b)): an electron can enter the central electrode through the left junction and leave through the right junction without changing the energy of the system. Because of this gate-voltage-controlled conduction, we can refer to the normal two-junction device as the NNN Coulomb blockade electrometer.

When the island is in the superconducting state (NSN Coulomb blockade electrometer), we expect the energy of the island to be given by  $E_n = E_c(n - C_oV_g/e)^2 + \Delta p_n$  where  $\Delta$  is the super-

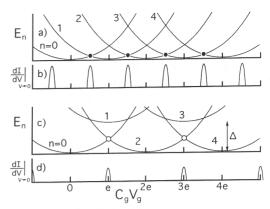


Fig. 2. Island total energy as a function of gate voltage  $V_{\rm g}$  for several values of the number n of excess electrons on the island, in the normal (a) and superconducting (c) states. In an ideal superconductor with gap  $\Delta$  the minimum energy for odd n is  $\Delta$  above the minimum energy for n. Black dots in (a) mark lowest energy level crossings where the transfer of an electron into and out from the island is energetically possible. The lowest energy level crossings in (c) are marked by a white dot to indicate that the simultaneous transfer of two electrons into and out from the island is energetically possible. Conductance in the vicinity of zero bias voltage as a function of gate voltage is shown in the normal (b) and superconducting state (d).

conducting energy gap and where  $p_n$  takes, in the ideal case, the values 1 or 0 if n is odd or even, respectively (see fig. 2(c)). We now suppose that  $C_{\Sigma}$ , while small enough to make the Coulomb energy  $E_c$  much greater than the thermal fluctuation  $k_BT$ , is sufficiently large to make the Coulomb energy less than the superconducting gap:  $\Delta > E_c$ . Consequently, as  $V_g$  is increased, the equality  $E_{2k+2} = E_{2k}$ , which takes place at  $C_g V_g = (2k+1)e$ , occurs before the equality  $E_{2k+1} = E_{2k}$ . In the superconducting state one thus expects the conduction at vanishing voltage to be restored when  $C_g V_g = (2k + 1)e$  (fig. 2(d)). This conduction would be provided by a sequence of two two-electron tunneling processes and should be 2e-periodic in gate charge Q = $C_{\sigma}V_{\sigma}$ . In the first process two normal electrons from the low voltage lead simultaneously tunnel into the island and form a Cooper pair. In the second process, which is the reverse of the first, a Cooper pair tunnels out of the island and form two electrons in the high voltage lead. This sequential 2e-transfer process is identical to the

Andreev reflection [4] of an electron in the first normal lead followed by the Andreev reflection of a hole in the second normal lead.

A 2e-periodicity of the sub-gap current as a function of gate charge has been found in the experiments of Geerligs et al. [5]. Tuominen et al. [6] and Haviland et al. [7] on the SSS Coulomb blockade electrode in which all electrodes are superconducting. However, in these experiments, the current as a function of gate voltage shows a lot of structure which is not completely understood. The fact that both the island and leads are superconducting complicates the interpretation of the results: both quasiparticle and Josephson tunneling have to be taken into account. In these experiments the 2e-periodicity cannot be unambiguously assigned to the properties of the superconducting island by itself. This is why we have chosen to investigate a Coulomb blockade electrometer in which the island only is superconducting. This case has been considered theoretically by Averin and Nazarov [8] but with emphasis on co-tunneling processes of single electrons rather than on twoelectron processes.

## 3. Sample

The sample was made using the now standard shadow-mask e-beam lithography technique [9]. During the first evaporation we deposited a 350 Å layer of aluminum to form a 0.23  $\mu$ m  $\times$ 0.86 µm superconducting electrode (see fig. 1). This layer was oxidized in 0.1 Torr O<sub>2</sub> during 5 min. The second evaporation consisted of a 50 Å thick buffer layer of aluminum followed immediately by a 550 Å thick layer of gold. The buffer layer was found useful for the adhesion of the gold layer. Due to the close contact to the gold layer, the aluminum buffer layer should stay normal by the proximity effect [10]. The sample was mounted in a shielded copper box thermally anchored to a dilution refrigerator. Measurements of the current I as a function of the bias voltage V and the gate voltage  $V_{\rm g}$  were made through carefully filtered coaxial leads [11].

#### 4. Results

We first performed large scale measurements of I versus V for different gate voltages in order to characterize the electrometer and to verify that the proximity effect was indeed taking place in the aluminium buffer layer. In fig. 1 we show the I(V) characteristics for gate charges Q = $C_{\alpha}V_{\alpha}$  giving a minimum and a maximum gap for positive bias voltage. A sharp current rise is seen at  $2\Delta$  in the minimum gap curve, and not at  $4\Delta$ like in the fully superconducting Coulomb blockade transistor [12]. This result is well explained by the theory of the NNN Coulomb blockade transistor [3], suitably modified to take into account the density of states of the superconducting island. For example this modified theory predicts that a tunnel event on the left junction changing n = 0 into n = 1 will occur for

$$\left(\frac{C_2 + C_g}{C_{\Sigma}}\right)V - \Delta/e > \left(e/2 - C_gV_g\right)/2C_{\Sigma}$$

while a tunnel event on the right junction changing n = 1 into n = 0 will occur for

$$\left(\frac{C_1}{C_{\Sigma}}\right)V - \Delta/e > (C_{\rm g}V_{\rm g} - e/2)/2C_{\Sigma} ,$$

hence the minimum gap for  $eV=2\Delta$ . Here  $C_1$  and  $C_2$  denote the capacitance of the left and right junctions, respectively. The data of fig. 1 therefore confirms the destruction of the superconducting state of the buffer aluminum layer due to the gold layer. The NSN Coulomb blockade transistor theory also explains the modulation of the current rise with gate voltage as displayed by a measurement of V versus  $V_{\rm g}$  taken at constant I (data not shown). From the fit of the data we obtain the tunnel resistances  $R_1=R_2=65~{\rm k}\Omega$ ,  $E_{\rm c}/e=127~{\rm \mu V}$ ,  $C_1=0.35~{\rm fF}$ ,  $C_2=0.28~{\rm fF}$ ,  $C_{\rm g}=7.2~{\rm aF}$  and  $\Delta/e=235~{\rm \mu V}$ . The value of  $C_{\rm g}$  is consistent with our previous results on NNN electrometers.

A further check on the NSN behavior of the sample was obtained by a detailed measurement of the smooth rise of I versus V for maximum gap (see fig. 3). We attribute this rounding of the

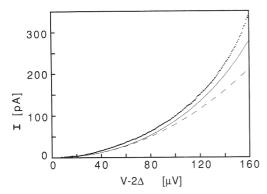


Fig. 3. Fine scale current-voltage characteristic in the vicinity of the voltage threshold for maximum gap. Solid line is the result of the calculation of the co-tunneling rate, including both  $(V-2\Delta)^2$  and  $(V-2\Delta)^3$  terms. Dashed line shows the contribution of the leading  $(V-2\Delta)^2$  term alone.

Coulomb blockade to inelastic co-tunneling events [13] in which one electron, while passing from one normal lead to the other, creates two quasiparticles in the island. This process can only occur for  $V > 2\Delta$ . We calculate at maximum gap and T=0 a co-tunneling current  $I_{\rm ct}=(R_{\rm K}/\pi R_1R_2)(C_{\rm S}/e)^2(\delta V^2\Delta/e+2\delta V^3/3\pi)$ , where  $\delta V=V-2\Delta/e$  and  $R_{\rm K}=h/e^2$ . The leading  $\delta V^2$  dependence is characteristic of a process involving the simultaneous transfer of an electron between two normal reservoirs and the breaking of a Cooper pair in an intermediate island. Our data points

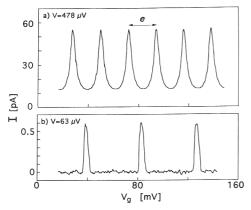


Fig. 4. Current through NSN transistor as a function of gate voltage for two values of bias voltage (a)  $eV \approx 2\Delta$  (b)  $eV \approx \Delta/4$ . In (a) double arrow marked "e" indicates gate voltage increment necessary to place one extra electron on the island.

presented in fig. 3 agree well with the theoretical curve (full line) in which all parameters were taken from previous measurements.

In addition to the e-periodic features observed at voltages of the order of or above  $2\Delta$ , we observed a 2e-periodic current for subgap bias voltages. We display this 2e-periodicity in fig. 4 where we show the current I as a function of gate voltage  $V_{\rm g}$  for  $V=478\,\mu{\rm V}$  ( $eV\simeq2e/\Delta$ ) and  $V=63\,\mu{\rm V}$  ( $eV\simeq\Delta/4$ ) and at 35 mK. Note that the 2e-periodic peaks are positioned half-way between two adjacent e-periodic peaks, as the theory presented graphically on fig. 2 predicts.

# 5. Coulomb blockade of Andreev reflection

Fig. 5 examines another aspect of the 2e-periodic feature by showing the current-voltage characteristics at gate charges 0, e and 1.06e. A linear I(V) around V=0 for Q=e is in agreement with our discussion of fig. 2(d). The data at Q=1.06e show that a small gap opens up near V=0 when Q departs from e. This gap can be interpreted as evidence for the Coulomb blockade of Andreev reflection. This blockade occurs when it is energetically unfavorable for the superconducting island to accept a pair of electrons. The opening of this gap is a behavior which is quite different from what is observed in SSS electrometers [5-7]. It does not point to a

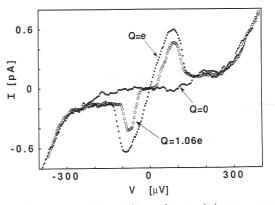


Fig. 5. Fine scale current-voltage characteristics near zero bias voltage for  $Q=C_{\rm g}V_{\rm g} \mod 2e=0$ , e and 1.06e. The Q=1.06e data display a small Coulomb gap.

coherent transport of electrons through the island but to an incoherent process where a voltage threshold for each of the two junctions must be exceeded for sequential tunneling to occur. The sequence of two Andreev reflections which we believe is responsible for the zerovoltage conductance does not require voltages greater than  $\Delta$  since no quasiparticles are excited. Its rate is 2e-periodic in Q because the intermediate state involves a charging of the island with 2e and its rate is proportional to the applied voltage. We have calculated (see Appendix) the zero-bias conductance at Q = e to be  $f^2(E_c/\Delta)R_K/8M_{\text{eff}}(R_1^2 + R_2^2)$  where  $f(x) = (2/\pi)\cos^{-1}(-x)(1-x^2)^{-1/2}$  and  $M_{\text{eff}}$  is the number of effective conduction channels through the junction. Our observed conductance of  $(100 \,\mathrm{M}\Omega)^{-1}$  corresponds to  $M_{\mathrm{eff}} \simeq 100$ , which we think is a reasonable order of magnitude considering the granularity of the electrode films [14]. The 2e-shuttling at Q = e through the island should be suppressed at bias voltages approaching  $2(\Delta - E_c)/e$  because single electrons can enter the island. This explains why the 2e-periodic current as a function of V forms a peak.

As we increased the temperature, the zerobias conductance associated with the peak for Q = e in fig. 5 remained approximately constant. However, the height of the peak decreased linearly with temperature and disappeared completely at  $T_0 = 130 \text{ mK}$ . This temperature dependence can be understood by replacing in the calculation of the population of the odd state the even-odd energy difference  $\Delta$  by the even-odd free energy difference  $F(T) = \Delta - k_B T \ln N_{eff}$ where  $N_{\rm eff}$  is the effective number of quasiparticle states [6]. At zero bias voltage and Q =e, the odd state becomes significantly populated when  $F(T) = E_c$  and we thus predict  $T_0 =$ 140 mK, in good agreement with our data. Our results differ from those of Tuominen et al. [6] who observed the vanishing of their 2e-periodic component at  $T_0 \simeq 300 \text{ mK}$ , which they interpret as the solution of F(T) = 0. There is probably no contradiction: our 2e-periodic tunneling mechanism can operate only if the island is in an even state whereas their unknown mechanism can perhaps operate whatever the parity of the island

electron number, the two parities corresponding to two 2*e*-periodic  $I(V_g)$  curves dephased by *e*.

# 6. Time scale of even-odd symmetry breaking

In fig. 6 we show a succession of  $I - V_g$  curves taken similarly to that of fig. 4(b). Each trace took 200 s, and successive traces were displaced downward slightly. Apart from a slow drift which we can attribute to the relaxation of the background charge [3], we have observed abrupt e-shifts of the curves which occurred intermittently on a time scale of several hours. A possible model to explain this data involves the infrequent tunneling of conduction electrons from the bulk of the island to localized states within the insulator at the surface of the island. The empty conduction state left by this event is immediately filled by an electron tunneling through one of the junctions. The filled localized state polarizes the island with charge e, thus giving an apparent shift of e in Q. This process can conceal the even-odd symmetry breaking if the data are accumulated on time scales much longer than the typical time of tunneling into the localized state. This time scale could be sensi-

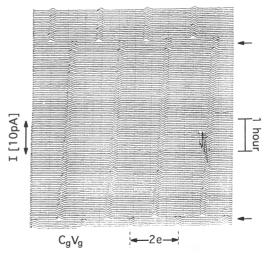


Fig. 6. Time evolution of  $I-V_{\rm g}$  characteristic. Repeated traces are shifted downward with time. Arrows indicate the position of e-shifts.

tively dependent on the material properties of the superconducting island.

## 7. Conclusion

We have presented the first experimental data on an NSN electrometer. At low bias voltage, this system has one dominant conduction mechanism, a sequence of two Andreev reflections. This 2e-electron tunneling mechanism reveals the even-odd symmetry breaking of the superconducting island by the 2e-periodicity of the current versus gate charge. The dependence of the data both with bias voltage and temperature agree quantitatively with the predictions based on this mechanism. Furthermore, our observation of random e-shifts in the gate charge demonstrates that even-odd symmetry breaking in a superconducting island is a concept which depends on the time scale of the observation. Finally, this experiment shows that Coulomb blockade may be used in disordered mesoscopic SN systems [15] to discriminate Andreev reflection on a background of single electron tunneling since at Q = eonly 2-electron conduction processes are allowed.

## Acknowledgements

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## **Appendix**

We calculate the current associated with the process depicted in fig. 7, in which two electrons from the left side of the NS junction combine to

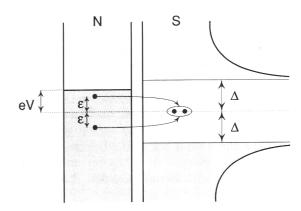


Fig. 7. Two-electron tunneling process involving a normal metal (N) and a superconductor (S) separated by a tunnel barrier. Two electrons on the N side tunnel coherently to form a Cooper pair on the S side. This process is equivalent to the Andreev reflection on the tunnel barrier of an electron in the N side into a hole.

form a Cooper pair on the right side. We start from the tunnel Hamiltonian

$$H_{t} = H + H^{\dagger} , \qquad (A1)$$

where H can be expressed in terms of electron operators c and  $c^{\dagger}$ :

$$\boldsymbol{H} = \sum_{mpk\sigma} t_{mkp} c_{mp\sigma}^{\dagger} c_{mk\sigma} . \tag{A2}$$

In the sum, m denotes the transverse (i.e. parallel to the barrier) mode index, p the longitudinal (i.e. perpendicular to the barrier) momentum on the right side, k the longitudinal momentum on the left side and  $\sigma$  the spin index. We assume that the tunnel matrix element  $t_{mkp}$  is independent of spin and that its modulus is independent of longitudinal momenta:  $|t_{mkp}| = t_m^2$ . It is well known that in the case where both sides are normal, this hamiltonian treated as a perturbation leads to a voltage independent one-electron tunneling conductance

$$G_{\rm NN}^{(1)} = R_{\rm T}^{-1} = \frac{2e^2}{h} \sum_{m} \mathcal{T}_{m} ,$$
 (A3)

where

$$\mathcal{T}_m = 4\pi^2 \rho_{lm} \rho_{rm} t_m^2 \,, \tag{A4}$$

where  $\rho_{1,rm}$  denotes the longitudinal density of state on the left (respectively right) side of the barrier. A Landauer approach to the tunneling conductance leads directly to eq. (A3) and identifies  $\mathcal{T}_m$  as the transmission coefficient of the barrier for longitudinal mode m.

We now take into account that fact that the right side is superconducting with a gap  $\Delta$  and compute the rate  $\Gamma$  of two-electron tunneling following the method of Schrieffer and Wilkins [16]. We start with Fermi's Golden Rule:

$$\Gamma = \frac{2\pi}{\hbar} \sum_{F} |\langle F|H^{(2)}|I\rangle|^2 \delta(E_F - E_I). \tag{A5}$$

The final state  $|F\rangle$  differs from the initial state  $|I\rangle$  by the emptying of two electron states on the left side with opposite spins, same mode index and with longitudinal momenta k' and k'' such that |k'| > |k''|. Since we take for the right side a BCS ground state with constant chemical potential, the extra Cooper pair which is formed as a result of two-electron tunneling does not lead to a change of state:

$$|F\rangle = c_{mk''-\sigma} c_{mk'\sigma} |I\rangle . \tag{A6}$$

The effective matrix element for two-electron tunneling is given by

$$\langle \mathbf{F}|H^{(2)}|\mathbf{I}\rangle = \mathcal{M}_{mk''k'\sigma}$$

$$= \sum_{M} \frac{\langle \mathbf{F}|H|M\rangle\langle M|H|\mathbf{I}\rangle}{E_{\mathbf{I}} - E_{\mathbf{F}}}.$$
(A7)

There are two types of intermediate states  $|M\rangle$  depending on whether electron k' or electron k'' tunnels first:

$$|\mathbf{M}\rangle = \gamma_{mp\sigma}^{\dagger} c_{mk'\sigma} |\mathbf{I}\rangle \tag{A8}$$

$$|\mathbf{M}\rangle = \gamma^{\dagger}_{m-p-\sigma} c_{mk''-\sigma} |\mathbf{I}\rangle \tag{A8'}$$

The operator  $\gamma_{mp\sigma}^{\dagger}$  affects the right side by destroying a pair  $(mp\sigma, m-p-\sigma)$  and creating a quasiparticle in mode m with longitudinal momentum p and spin  $\sigma$ . We now proceed by expressing H in terms of  $\gamma$ 's and  $\gamma^{\dagger}$ 's. We use the Bogoliubov transformation [17]

$$c_{p\sigma}^{\dagger} = u_{p\sigma} \gamma_{p\sigma}^{\dagger} + v_{p\sigma} \gamma_{-p-\sigma}^{\phantom{\dagger}} , \qquad (A9)$$

where the *u*'s and the *v*'s verify the symmetry relations  $u_{p\sigma} = u_{-p\sigma} = u_{p-\sigma} = u_{-p-\sigma}$  and  $v_{p\sigma} = v_{-p\sigma} = -v_{p-\sigma} = -v_{-p-\sigma}$ . Using these relations and the commutation properties of the *c*'s, we find

$$\mathcal{M}_{mk''k'\sigma} = \sum_{p} t_{mpk'} t_{m-pk''} u_{mp\sigma} v_{mp\sigma}$$

$$\left(\frac{1}{E_{mp} + \epsilon_{mk'} - eV} + \frac{1}{E_{mp} + \epsilon_{mk''} - eV}\right), \quad (A10)$$

where  $E_{mp}$  and  $\epsilon_{mk}$  denote quasiparticle and electron energies on the right and left side, respectively. We can now replace the sum over momenta p in eq. (A10) by an integral over the energy  $\zeta$  of the one-particle states participating in the BCS ground state:  $E_{mp}^2 = \Delta^2 + \zeta_{mp}^2$ . We also make use of the relation  $2u_{mp\sigma}v_{mp\sigma} = \Delta/E_{mp}$ . Finally, in view of the delta function in eq. (A5) we are only interested in  $\mathcal{M}_{mk''k'\sigma}$  such that  $\epsilon_{mk'} + \epsilon_{mk''} - 2eV = 0$  and it is useful to introduce the variable  $\epsilon = \epsilon_{mk'} - eV$  (see fig. 7). We get

$$|\mathcal{M}_{mk''k'}|^2 = \rho_{rm}^2 t_m^4 \left( \int_{-\infty}^{+\infty} \frac{\Delta \, \mathrm{d}\zeta}{\Delta^2 + \zeta^2 - \epsilon^2} \right)^2. \tag{A11}$$

The sum over F in eq. (A5) can now be replaced by an integral over the energy  $\epsilon$  in the interval [0,eV]. Since we are only interested in the zero voltage conductance we can drop the  $\epsilon^2$  term in eq. (A11) and we get  $\pi$  for the integral. Taking into account the two possibilities for  $\sigma$ , we finally obtain the expression for the two-electron tunneling conductance  $G_{\rm NS}^{(2)} = \lim_{V \to 0} 2e\Gamma/V$ 

$$G_{\rm NS}^{(2)} = 16\pi^4 \frac{e^2}{h} \sum_m \rho_{1m}^2 \rho_{rm}^2 t_m^4 = \frac{e^2}{h} \sum_m \mathcal{T}_m^2$$
. (A12)

Our perturbative result is identical, in the small  $\mathcal{T}_m$  limit, with the more general result by Beenakker for Andreev reflection in NS systems with arbitrary transmission on the N side [18]

$$G_{\rm NS}^{(2)} = \frac{4e^2}{h} \sum_{m} \frac{\mathcal{T}_m^2}{(2 - \mathcal{T}_{m})^2}.$$
 (A13)

If we assume that there are  $M_{\rm eff}$  modes that contribute to the sum in eq. (A12) with equal transmission coefficient  $\mathcal{T}$ , we get a relation between the two-electron tunnel conductance and the normal electron tunnel resistance:

$$G_{\rm NS}^{(2)} = \frac{h/e^2}{4M_{\rm eff}R_{\rm T}^2} \,. \tag{A14}$$

We can now generalize eq. (A14) to the case of the NSN Coulomb blockade electrometer with gate voltage  $V_{\rm g} = e/C_{\rm g}$ . By analogy with the NNN Coulomb blockade electrometer with gate voltage  $V_{\rm g} = e/2C_{\rm g}$  for which  $G_{\rm NNN}^{(1)} = (R_{\rm T_1} + R_{\rm T_2})^{-1}/2$ , we are tempted to write  $G_{\rm NSN}^{(2)} = (h/8e^2)M_{\rm eff}^{-1}(R_{\rm T_1}^2 + R_{\rm T_2}^2)^{-1}$ , where the G's refer to the zero-voltage conductances of the electrometer and the R's to the tunnel resistances the junctions (we assume that the capacitances of the two junctions are equal). However, we must take into account the fact that the island is not at constant potential during each of the two-electron tunneling processes. The  $E_{mp}$  term in the energy denominators of eq. (A10) must be replaced by  $E_{mp} - E_{c}$  since the quasiparticle in the virtual intermediate state lowers the electrostatic energy of the island by the Coulomb energy  $\dot{E}_{\rm c}$  (see fig. 2(c)). In eq. (A11) the integral becomes in the limit of small  $\epsilon$ 

$$\int_{-\infty}^{+\infty} \frac{\Delta \,\mathrm{d}\zeta}{\sqrt{\Delta^2 + \zeta^2} (\sqrt{\Delta^2 + \zeta^2} - E_c)} = \pi f(E_c/\Delta)$$

$$= \frac{2\cos^{-1}(-E_c/\Delta)}{\sqrt{1 - (E_c/\Delta)^2}}.$$
(A15)

The final expression for the zero-voltage conductance of the electrometer at  $V_g = e/C_g$  is thus

$$G_{\rm NSN}^{(2)} = \frac{h}{e^2} \frac{f^2(E_{\rm c}/\Delta)}{8(R_{\rm T_1}^2 + R_{\rm T_2}^2)} \,. \tag{A16}$$

This expression diverges when  $E_{\rm c}$  tends towards  $\Delta$ . This, however, is an artefact of the perturbation theory which is also found in the theory of co-tunneling [13]. At  $E_{\rm c} = \Delta$ , the two-electron tunneling process continuously merges into a one-electron tunneling process and the conduct-

ance stays finite. In our experiment  $E_c/\Delta \simeq 0.54$  and the value of the  $f^2$  factor is only 2.59.

## Note added in proof

A recent theoretical work by F.W.J. Hekking, L.I. Glazman, K.A. Matveev and R.I. Shekhter (preprint) extends our expression (A16) to non-zero temperature and bias voltage and finds good agreement with our experimental results.

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