2e and e Periodic Pair Currents in Superconducting Coulomb-Blockade Electrometers

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We have measured the current as a function of gate charge Q in superconducting Coulomb blockade electrometers with charging energies $E_c \ll \Delta$, the superconducting gap. We find a large pair current which exhibits a clear transition from $2e$ to e periodicity at about 250 mK. To explain our data, we propose an equilibrium model in which the current is due entirely to Cooper pairs. The periodicity change results from rapid shifts in the instantaneous effective Q, caused by fluctuations in quasiparticle number. We find good agreement, and predict the loss of $2e$ periodic current for large E_c.

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A Coulomb-blockade electrometer can be thought of as a very small conducting island which is weakly coupled to the external world by means of two tunnel junctions. The device also has a capacitively coupled gate electrode, with capacitance C_g, which allows the island's potential to be altered by application of a voltage V_g. To ensure that the coupling is weak, and therefore that the number of electrons on the island is well defined, each junction must have a normal state tunneling resistance $R_n > h/4e^2$ and the total island capacitance C_Σ must be small enough that the elementary charging energy $E_c \equiv e^2/2C_\Sigma$ is much greater than k_BT, where T is the temperature. To use the electrometer, one applies a fixed voltage V across the series junctions (see inset to Fig. 1), and measures the current I passing through the island as a function of the gate charge $Q=C_gV_g$. The device characteristics are well understood when the island and leads are normal metal; current arises from the discrete tunneling of electrons onto the island and is periodic in Q with periodicity e [1,2].

The e periodicity in the normal state arises from the simple fact that the current is carried by individual electrons. Naively, one would expect a superconducting device to show a $2e$ periodicity because the current should then be carried by Cooper pairs. However, in several previous studies where $2e$ periodicity was expected, only e-periodic behavior was found [3-5]. Recently, $2e$ periodicity has been reported for entirely superconducting electrometers (SSS) [5-8], as well as electrometers in which the island is superconducting and the leads are normal (NSN) [9-12]. Tuominen *et al.* [6] found the current to be $2e$ periodic below ~300 mK, and explained the crossover to e periodicity above that temperature in terms of the disappearance of a parity-dependent free energy. Although their explanation gives the correct transition temperature, the complexity of their $I(Q)$ curves makes it difficult to interpret the crossover in detail. In this Letter, we present detailed measurements and discussion of the $2e$ to e periodicity transition in two SSS electrometers. In a notable departure from previously reported work, our data are taken on devices with $E_c \ll \Delta$, and show remarkably simple features that we explain using an equilibrium theory which incorporates E_c and Q as well as Δ and k_BT.

Our electrometers have an Al island and Al-Al_2O_3-Al tunnel junctions fabricated on unoxidized 100 mm^2 Si chips using e-beam lithography and double-angle evaporation [1]. Each island measures 1.8 μm×0.2 μm, and is typically 25 to 55 nm thick. The leads are ~0.1 μm wide and cross all or part of the island's width, enabling variations in junction capacitance. On each chip, we fabricate up to 12 electrometers with junction capacitances ranging from 1 to 3 fF. The Al leads extend over 100 μm in length before contacting Au pads; our design does not incorporate quasiparticle traps near the island [8].

We measure the devices in nominally zero magnetic field at temperatures as low as 47 mK on a dilution refrigerator inside a shielded room in an electromagnetically quiet subbasement. Wiring to the chip incorporates rf and microwave filters placed at room temperature, 1 K and the mK stage. For most measurements, we voltage-bias the devices and measure the current through the island using a current preamplifier [13]. The gap Δ/e is

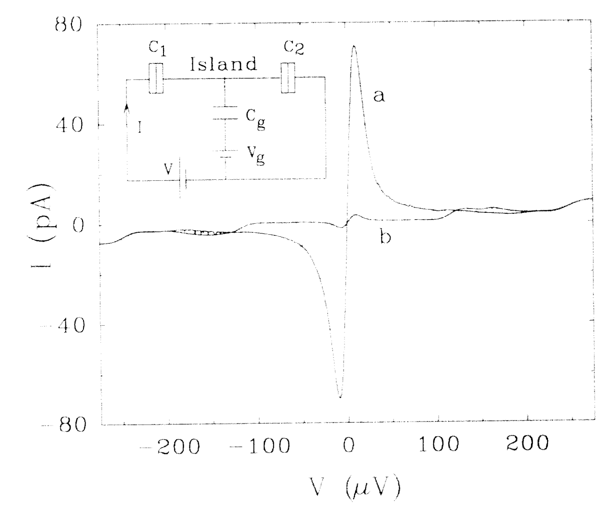

FIG. 1. I-V characteristics at low voltages for device 1. $T=50$ mK. Curve a corresponds to $Q=e$, while curve b is for $Q=0$. Inset shows device layout: C_g is the gate capacitance, C_1 and C_2 are capacitances of the two tunnel junctions, and $C_\Sigma = C_1 + C_2 + C_g$.

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taken to be 1/4th the voltage at which there is a sharp onset of quasiparticle current in the I-V curves. We deduce R_n and E_c from measurements in the normal state, produced by applying a 1.9 T magnetic field. For the two devices which we measured in detail, we find $E_c = 43$ μeV, $\Delta = 193$ μeV, $R_n \approx 58$ kΩ, and island volume $\Omega \approx 0.9 \times 10^{-20}$ m^3 for device 1; and $E_c = 31$ μeV, $\Delta = 186$ μeV, $R_n \approx 29$ kΩ, and $\Omega \approx 1.7 \times 10^{-20}$ m^3 for device 2.

Figure 1 shows low-voltage I-V curves of device 1 at 50 mK for fixed Q. In this so-called supercurrent region, the current peaks at $V \approx 10$ μV, rather than at zero voltage as occurs in Josephson junctions with $R_n \ll h/4e^2$. The peak current has a strong dependence on the gate charge Q and is $2e$ periodic, reaching a maximum when Q/e is an odd integer, and a minimum when Q/e is even. For device 1, the $2e$ periodicity persists to $V \approx 200$ μV, above this the system becomes e periodic and the $Q=0$ and $Q=e$ curves are indistinguishable. Device 2 has a higher peak current ($\sim$0.45 nA) and shows similar behavior.

Upon increasing the temperature, the $2e$ periodicity occurs over a steadily decreasing voltage range. At any voltage in this range, the $2e$ periodic current diminishes, being replaced by e periodic current above a well-defined temperature. For simplicity, we will focus on the most prominent $2e$ periodic feature, the current peak at 10 μV. Figure 2 shows $I(Q)$ curves at $V = 10$ μV for device 1 taken for T between 50 and 500 mK. At 50 mK (curve a), peaks occur only at odd values of Q/e, while at even Q/e the current is a minimum; the period is clearly $2e$. For $T > 260$ mK (curves h-j), current peaks of equal magnitude occur at odd as well as even values of Q/e, and the system is e periodic. The curves at intermediate temperatures (curves b-g) reveal a remarkably simple pattern. As T is increased, the transition from $2e$ to e periodicity is accomplished by a reduction in the peak height at odd Q/e and a concurrent emergence of peaks at even Q/e until the odd and even peaks become equal. As the temperature is raised further, both peaks grow but remain of identical heights, and e periodicity is maintained.

The clarity of our low-voltage $I(Q)$ curves, and the systematic manner in which they vary with temperature, suggest that an accurate comparison can be made with theory. At low bias voltages, the island is almost in equilibrium with its surroundings and it is appropriate to construct an equilibrium thermodynamic model. We regard the current through the island as a probe of the state of the system. To construct the model we assume: (1) the Josephson coupling energy can be neglected [14] when computing the energy of pairs and quasiparticles on the island, (2) the island is at a constant temperature T, (3) it has a fixed volume Ω, and (4) it can exchange pairs and quasiparticles with the leads. The last condition implies that the chemical potential μ is fixed, rather than the number of particles on the island. The island μ, and thus the average particle number, can be changed by varying V_g.

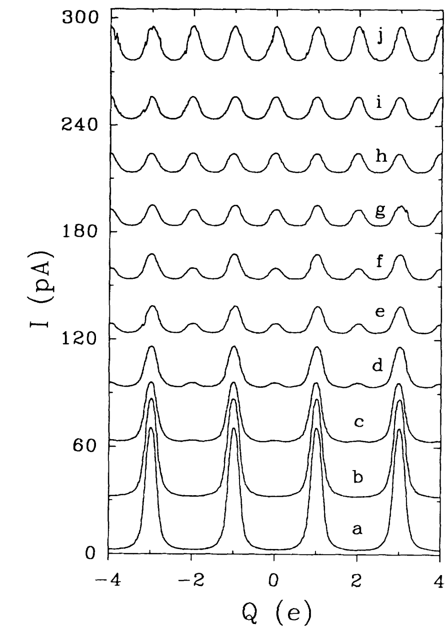

FIG. 2. Plots of current vs gate charge at $V = 10$ μV for device 1, taken at curves a, 50 mK; b, 170 mK; c, 205 mK; d, 220 mK; e, 234 mK; f, 238 mK; g, 250 mK; h, 264 mK; i, 300 mK; and j, 487 mK. For clarity, curves b-j have been displaced successively by 30 pA along the I axis. The bottom curve (a) has no current offset.

With these assumptions, the island will assume a thermodynamic state in which the Gibbs grand potential [15] $\Phi_F(\mu, \Omega, T) = F - \mu N$ is a minimum, where F is the Helmholtz free energy, and we can identify $N = 2N_p + N_{qp}$ as the number of excess electrons on the island. Given the grand partition function Z, Φ_F can be obtained using the relation $\Phi_F = -k_B T \ln(Z)$. When the island is superconducting, we find $Z = Z_e + Z_0$, where [16]

$$Z_e = [A_e \cosh(f) + A_0 \sinh(f)] \times \sum_{n=-\infty}^{\infty} \exp\left[-\frac{(Q+2ne)^2}{2C_\Sigma k_B T}\right], \qquad (1a)$$

$$Z_0 = [A_e \sinh(f) + A_0 \cosh(f)] \times \sum_{n=-\infty}^{\infty} \exp\left[-\frac{[Q+(2n+1)e]^2}{2C_\Sigma k_B T}\right], \qquad (1b)$$

and where

$$f = \Omega \int_0^\infty D(E) dE \exp(-E/k_B T),$$

$$A_e \approx 1 + \sum_{n=1}^{\infty} A_{2n} f(T/2n),$$

$$A_0 \approx \sum_{n=1}^{\infty} A_{2n+1} f(T/(2n+1)),$$

$A_n = (-1)^n (n^2 - 5n + 4) 2^{n-3}/n!$, $D(E) \approx E D(E_f)/(E^2 - \Delta^2)^{1/2}$ is the quasiparticles' density of states in the island, and $D(E_f)$ is the density of states in the normal metal at the Fermi energy E_f.

The sums can be evaluated numerically and converge rapidly for our experimental parameters. For T very small, $Z_0 \approx 0$ and no more than two terms are important in Z_e, those for which $2n \approx Q/|e|$ or $2(n+1) \approx Q/|e|$. Essentially, the island assumes a state in which there are no quasiparticles and the number of excess pairs N_p minimizes the charging energy $(Q+2N_p e)^2/2C_\Sigma$. This is expected because as T approaches zero, Φ_F approaches the charging energy $U-\mu N$; minimizing Φ_F means choosing the state with the least charging energy. Figure 3 shows a section of the plot of charging energy vs Q for the experimental parameters of device 1. Each parabola corresponds to a fixed number of pairs and quasiparticles on the island. We have shaded the parabolas with quasiparticles to indicate a continuum of energy levels above the curves; unlike pairs, quasiparticles can carry random thermal kinetic energy. Note that a state with N_{qp} quasiparticles has an energy of at least $N_{qp}\Delta$. At $T=0$, the system assumes a state on the lowest parabolas, i.e., with zero quasiparticles. As the temperature is increased, single-quasiparticle states at $Q=\pm e$ are the first to get populated, the excitation energy being the lowest there. From the grand partition function, we can obtain the probability at any fixed Q and T that the island has a given number of quasiparticles.

Current arises from transitions between states that differ in the number of pairs or quasiparticles, i.e., from transfer of charge on or off the island. The only transitions which are significant are those between states differing by ± 1 pair or quasiparticle. In fact at low temperatures, the quasiparticles' density is very low and, although we may not neglect the probability that the island has quasiparticles on it, we may neglect the *current* carried by quasiparticles at low voltages. Taking this into account, we can write

$$I(Q,T) \approx \sum_{n=-\infty}^{\infty} P_n(Q,T) I_n(Q,T) , \qquad (2)$$

where $P_n(Q,T)$ is the probability that the island has n electrons on it, and $I_n(Q,T)$ is the associated pair current. The individual pair currents $I_n(Q,T)$ have a noteworthy symmetry, $I_n(Q,T)$ equals $I_0(Q,T)$ for n even and $I_0(Q+e,T)$ for n odd. This symmetry arises from the periodicity evident in Fig. 3 and the fact that pair transitions only connect parabolas with the same number of quasiparticles. The current can then be written as

$$I(Q,T) \approx P_{\text{even}}(Q,T) I_0(Q,T) + P_{\text{odd}}(Q,T) I_0(Q+e,T) , \qquad (3)$$

where $P_{\text{even}}(Q,T) = Z_e/Z$, and $P_{\text{odd}}(Q,T) = Z_0/Z$ are, respectively, the probabilities of having an even or odd number of electrons on the island.

Our experimental $I(Q)$ curves can now be understood.

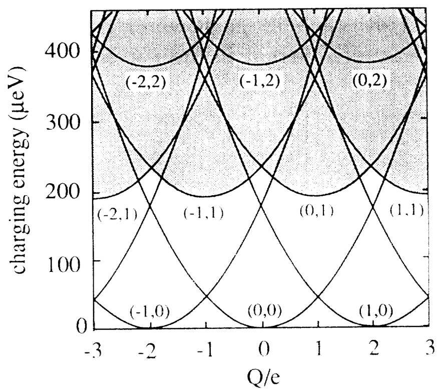

FIG. 3. Theoretical plot of the island's charging energy as a function of Q for the parameters of device 1: $\Delta = 193$ μeV, $E_c = 43$ μeV. Each parabola corresponds to a fixed number of pairs and quasiparticles (N_p, N_{qp}).

Equation (3) says that $I(Q)$ is a superposition of two currents, displaced with respect to each other along the Q axis by e, and weighted by the probabilities P_{even} and P_{odd}. If we assume that I_0 is practically independent of temperature, i.e., we assume $I_0(Q,T) \approx I_0(Q)$, then the temperature dependence of $I(Q)$ will come from the way P_{even} and P_{odd} change with temperature. At sufficiently low temperatures, $P_{\text{odd}} \approx 0$ and $P_{\text{even}} \approx 1$ for *all* Q; thus $I(Q, \text{low } T) \approx I_0(Q)$. Experimentally we obtain $I_0(Q)$ as the measured $I(Q)$ at $T \approx 50$ mK, where the quasiparticle population is expected to be negligible; we find that $I_0(Q)$ is $2e$ periodic. As the temperature is increased, P_{odd} becomes non-negligible, and the $I(Q)$ curve begins to get a contribution from the $P_{\text{odd}}(Q,T)$ term, seen as an e-shifted and reduced version of $I(Q)$ superposed on the original $I(Q)$.

To provide a detailed comparison, we plot in Fig. 4 the measured values of $I(e,T)$ and $I(0,T)$ alongside the predictions of Eq. (3), shown by solid lines. The theoretical plots use the experimentally determined E_c, Δ, and Ω for each device; there are no adjustable parameters. We find good agreement on the temperature for the $2e$ to e crossover. The theory also reproduces other details evident in the data, most significantly the minimum in the $Q=e$ current. We note, however, that above the transition temperature, the measured current falls below the theoretical value; this discrepancy probably arises from ignoring the Josephson energy and the possible temperature dependence in $I_0(Q,T)$.

We would like to mention some implications of our analysis. First, the analysis does not depend on the nature of the leads, except in that they act as good reservoirs for charge and energy exchange. However, NSN and SSS electrometers will have different characteristics because the functional form of $I_0(Q,T)$ does depend on the leads. Second, our model is valid only near zero voltage, where the island is in equilibrium with the leads. A quantitative understanding of the data at larger voltages will require a nonequilibrium model which goes beyond

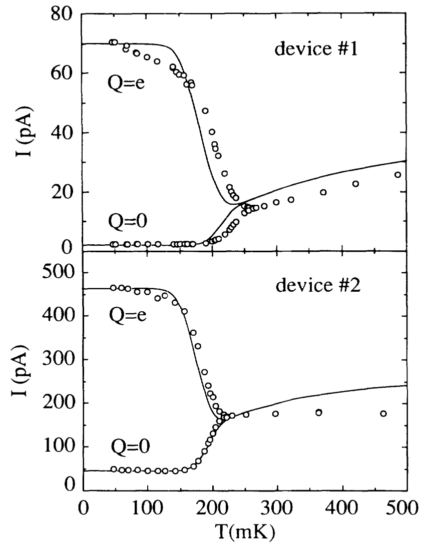

FIG. 4. The temperature dependence of electrometer current at $Q=e$ and $Q=0$ for devices 1 and 2. $V=10$ μV. Open circles are measured data, while the solid curves are computed theoretically.

the simple picture presented here. Third, we find that the temperature of the periodicity transition is determined only by the island gap Δ and volume Ω, and is almost independent of E_c. Thus the theory of Tuominen *et al.* [6], which does not incorporate E_c, gives the correct transition temperature. However, we find that the *observability* of the $2e$ periodic current requires that E_c not be too large. The temperature at which $I(e,T)$ starts to decrease significantly is roughly proportional to $\Delta - E_c$, the energy required to excite a quasiparticle at $Q=e$. This fact was also recognized by Eiles [5]. It follows that devices with large E_c (smaller junctions) are unsuitable for observing clear $2e$ periodicity. For example, our simulations show that for Al ($\Delta \sim 190$ μeV), when $C_\Sigma=0.5$ fF, the $Q=e$ current drops below 10^{-3} of its zero temperature value by $T=0.1$ K.

Finally, we note that other mechanisms can hide the $2e$ periodicity. We have found that degrading the electromagnetic shielding of our cryostat causes the $Q=e$ peak to drop by a factor of ~ 3, and a noticeable secondary peak to appear at $Q=0$ even at 50 mK. This points to the possibility that external noise sources can inject quasiparticles onto the island, mimicking the effect of higher temperatures. This could explain the secondary ($Q=0$) peaks seen in an NSN electrometer by Hergenrother, Tuominen, and Tinkham [12] and in some SSS devices by Eiles [5].

In conclusion, the low-voltage characteristics of superconducting electrometers can be understood within the framework of a simple equilibrium picture where, for some fraction $P_{\rm odd}(Q,T)$ of the total time, there is an odd number of quasiparticles present on the island. The increasing probability of having an odd number of quasiparticles causes the periodicity to change from $2e$ to e. Because of this, Cooper pair current need not *look* $2e$ periodic in the gate charge. In our model, E_c and Δ act somewhat independently: E_c sets the criterion for observability in that a strong $2e$ periodic current should be seen for $E_c \ll \Delta$, while Δ alone determines the temperature of the periodicity transition. By taking this into account, some of the apparently conflicting results from earlier experiments can now be understood.

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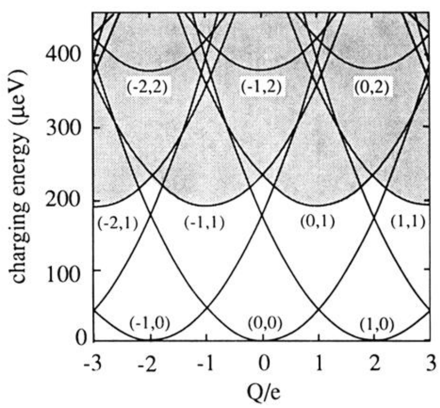

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