Temperature dependence of even-odd electron-number effects in the single-electron transistor with a superconducting island

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A simple quasiequilibrium model is presented that accounts in some detail for the observed temperature dependence of the crossover from $2e$ to e periodicity (vs gate charge) in the current through a single-electron tunneling transistor with a mesoscopic superconducting island.

The single-electron tunneling transistor consists of a small metallic island weakly coupled to two bias leads by high-resistance, low-capacitance tunnel junctions, and capacitively coupled to a gate electrode by a capacitance C_g. The current I through the device for a given bias voltage V is a periodic function of the voltage V_g on the gate electrode. If the island is of normal metal, the period corresponds to a change in the gate charge $Q_0 = C_g V_g$ by a single electronic charge e, whereas if the island is superconducting, the period can be $2e$ or e, depending on the temperature and the bias voltage across the two tunnel junctions. Qualitatively, the period is $2e$ if as many electrons as possible on the superconducting island are paired; the period becomes e when at least one excess quasiparticle is present, whether by injection at high bias voltages or by thermal excitation as the temperature is raised. In this paper we present a simple model calculation which gives insight into how this crossover in period takes place as a function of temperature in the limit of low bias voltage, together with some illustrative experimental data.

To calculate the actual device current $I(V, V_g)$ theoretically, it is necessary to make a kinetic calculation,[1,2] solving a master equation to find the self-consistent steady-state nonequilibrium populations of all relevant states, and the resulting current. However, in the limit of low bias voltage, state populations will be near to the $V = 0$ *equilibrium* values for the same gate voltage V_g. At sufficiently low bias voltages, we expect the current through the device to be proportional to V with a coefficient[3] which is a function of V_g and T, dependent on the equilibrium populations. Thus, we expect that the *period* (e or $2e$) of the current will be determined by the *period* with which the populations vary with V_g, allowing us to use the periodicity of the equilibrium populations as a proxy for the periodicity of the current at low bias voltages. The enormous simplification which this entails is the motivation for pursuing this approach, even if it is limited to finding the period of $I(V_g)$, without being able to find its magnitude and wave form. It seems likely that an analytic prescription could be developed for calculating the linear response $dI/dV|_{V=0}$ as a function of V_g based on the knowledge of the equilibrium populations, but that remains for future work.

We start by recalling that if the island is in the *normal* state, with $V \approx 0$, the part of the electrostatic energy which depends on n, the number of excess electrons on the island, is given by

$$E = \frac{(Q_0 - ne)^2}{2C_\Sigma} \equiv E_c \left[\frac{Q_0}{e} - n \right]^2 . \tag{1}$$

Here $Q_0 = -C_g V_g + Q_{00}$ is the charge induced by the gate plus any intrinsic offset charge Q_{00} from charged impurities, e is the charge of the electron including its sign, C_Σ is the total capacitance of the island to the bias leads and the gate electrode, and $E_c \equiv e^2/2C_\Sigma$ characterizes the Coulomb charging energy. As is evident from the plot in Fig. 1(a), this expression is minimized if ne always takes on the value nearest to Q_0. Thus, as V_g is swept, n changes by unity every time Q_0 passes through a half-integral value. This leads to a variation of the populations, and hence of the current $I(V_g)$ at fixed bias V, which is e periodic.

If the island is a *superconductor*, the above results are modified by the electron pairing. If the total number N of conduction electrons on the island is even, the BCS ground state is fully paired; if N is odd, the ground state must include one quasiparticle above the energy gap Δ. To describe this distinction, Averin and Nazarov[4] introduced an explicit additive energy term, which has the value Δ in odd-N states, and zero in even-N states. As can be seen from Figs. 1(b) and 1(c), this has the effect of introducing a $2e$ periodicity in the energy level diagram, and hence in the populations of the various possible states. This in turn should be reflected in a $2e$ periodicity in the low-voltage current through the device at low temperatures, but at sufficiently high temperatures we expect to recover the e periodicity of the normal state. The objective of this paper is to clarify the nature of this transition from $2e$ to e periodicity with increasing temperature.

Although $2e$-periodic currents in an *SSS* transistor (i.e., one in which leads and island are both superconducting) had been reported earlier by Geerligs *et al.*,[5] this even-odd electron number effect on the tunnel current was first clearly demonstrated and interpreted by Tuominen *et al.*,[6,7] also using an *SSS* device. Their work showed that the $2e$ periodicity changed to an e periodicity upon warming through a temperature T^*, far below T_c, where $\Delta(T^*) \approx \Delta(0) \gg k_B T$ and the material is still

strongly superconducting. Empirically, T^* was found to be essentially the temperature at which a *single* thermal quasiparticle is excited in the whole sample. Some more recent data illustrating this effect in an *NSN* transistor are shown in Fig. 2. [The bias voltage (125 μV) used to obtain data with excellent signal/noise ratio is too large for this quasiequilibrium analysis to be strictly applicable, but it should still be qualitatively correct.] We now examine the physics which leads to the changeover from a 2e- to an e-periodic dependence at this particular T^*.

For simplicity, we restrict our attention to the case $\Delta > E_c$, for which Fig. 1(c) displays the relevant low-lying energy levels. For all values of Q_0, the ground state has an even number of electrons, and is *nondegenerate* (except at the level crossings where Q_0/e is an odd integer). In contrast, the lowest states with an *odd* number of electrons have a high statistical weight N_{eff}, because the quasiparticle states form a quasicontinuum above the energy gap. This $N_{\text{eff}} \approx 10^4$ is essentially[8] the total number of quasiparticle states within $k_B T$ above the gap in the entire island volume (typically $\sim 3 \times 10^{-15}$ cm^3). Taking account of the multiplicity of levels, the probability of

finding the system in one of these odd-n levels relative to the probability of being in the even-n ground state is $\sim N_{\text{eff}} e^{-\Delta E/kT}$, where $\Delta E(Q_0)$ is the energy difference from the even-n ground state up to the lowest odd-n state.[9] Since the average of $\Delta E(Q_0)$ over Q_0 is simply Δ, the heuristic criterion based on approximately one thermally excited quasiparticle suggests defining a zero-order estimate $k_B T_0^* = \Delta/\ln N_{\text{eff}}$. Upon inserting $N_{\text{eff}} \approx 10^4$ and the BCS value $\Delta = 1.76 k_B T_c$, one finds $T_0^* = (\Delta/k_B)/\ln(N_{\text{eff}}) \approx T_c/5$, in good agreement with experimental data.

Although this simple argument gives the correct answer, it is not clear exactly how it relates to the experiment, in which the periodicity of the measured current with Q_0/e is the issue, while the simple theoretical estimate above is obtained after *averaging* over Q_0. Physically, two distinct energies are competing with the thermal energy: the energy gap Δ opposes creation of *quasiparticles*, and the Coulomb energy E_c tries to make the *charge* or *electron* number match the gate charge Q_0. In the normal state, $\Delta = 0$, and Q_0 alone controls the number of electrons, subject to thermal rounding, as described above. In the present case, we assume $\Delta > E_c$, so that the ground state must contain the *even* number of electrons closest to that specified by Q_0. At $T > 0$, quasiparticles
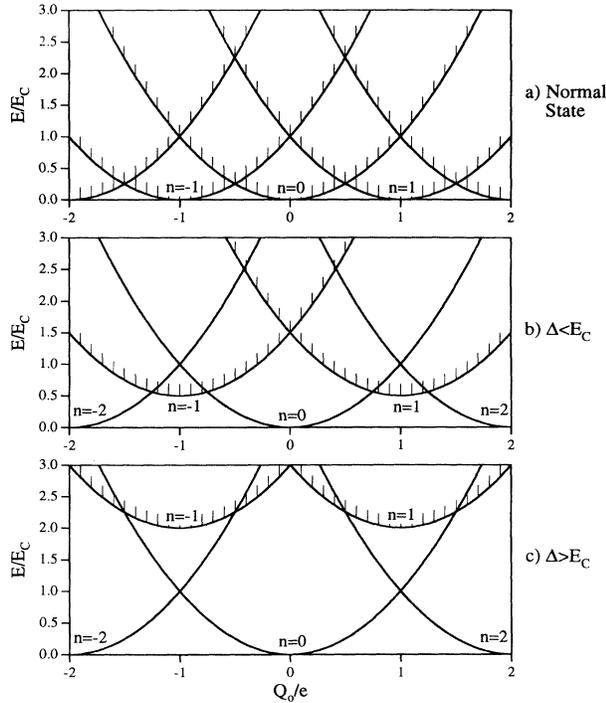

FIG. 1. The n-dependent part of the system energy E as a function of the gate charge Q_0 at $T=0$. (a) Normal island, showing e periodicity of energy levels; (b) superconducting island, with $\Delta < E_c$; (c) superconducting island, with $\Delta > E_c$. In both (b) and (c), the energy level structure shows 2e periodicity. At the lowest-lying degeneracy points where two adjacent parabolas cross, the value of n in the ground state changes, and charge transport can occur at $T=0$ without an energy barrier. Shading indicates schematically the presence of a continuum of low-lying quasiparticle states above the energy gap in the case of $n =$ odd, or in the normal state. [For simplicity, we assume that the parities of N and n are the same.]
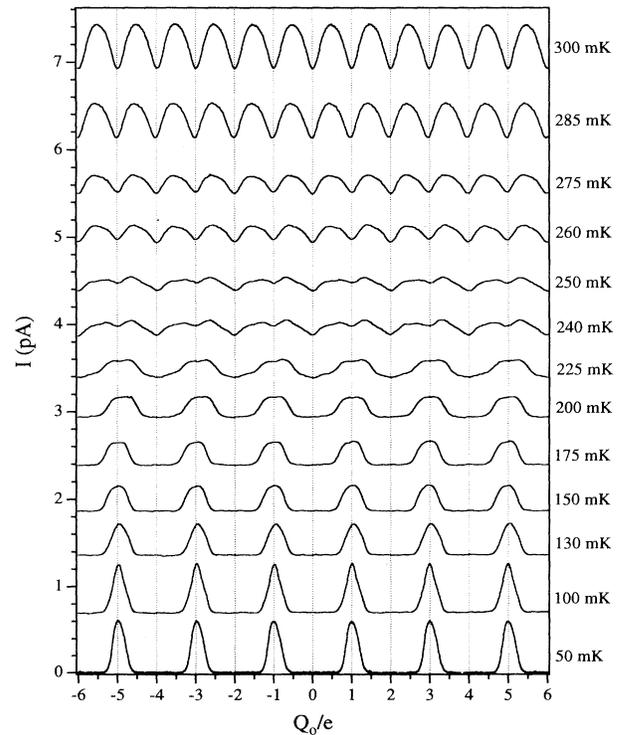

FIG. 2. Current through an *NSN* single-electron transistor with Al island vs gate charge Q_0 at temperatures from 50 to 300 mK (bias voltage $V = 125$ μV). Curves have been displaced upward successively for clarity. Note that the transition from 2e periodicity to e periodicity occurs in the rather narrow temperature range 240–270 mK near T^*, where the even-odd free energy difference F_0 is going to zero.

can be thermally excited. It is useful to separate the effect of E_c which enters only when comparing states differing in *charge* by $\pm e$, from the effect of Δ, which is present for any excitation. In cases where the charge is changed, Fig. 1(c) shows that E_c raises the energy of odd-n states relative to even-n states by an amount which is $2e$ periodic in Q_0, and which can be written explicitly as

$$E_0(Q_0) = (1 - 2|Q_0/e|)E_c \qquad (2)$$

if Q_0/e is restricted to the representative range $-1 < Q_0/e < +1$. This $E_0(Q_0)$ has the property that $E_0(Q_0) = -E_0(Q_0 + e)$, so that it averages to zero over Q_0, as stated above. The expression (2) can be extended periodically to describe arbitrarily large Q_0 values.

The precise physical criterion for the transition from $2e$ to e periodicity is that the probability of an *odd* number of electrons at Q_0 be the same as the probability of an *even* number of electrons at $(Q_0 + e)$ for all Q_0, just as would be the case in the normal state. To allow a simple analytic computation of these probabilities, we again replace the exact system of energy levels by a model in which, for $-1 \le (Q_0/e) \le 1$, the ground state is a nondegenerate state with $n = 0$, and the quasicontinuum of excited states above the gap is modeled by a single level with degeneracy N_{eff}, and $n = 1$ or -1 depending on the sign of Q_0. [These are the states indicated by the two lowest parabolas at $|Q_0| \le e$ in Fig. 1(c).] In the typical case where $k_B T \ll E_c$, we can ignore all *charge* states with n values other than these. We also assume that $k_B T \ll \Delta$, but because of the large statistical weight N_{eff} of the excited states, we need to take account of any number (so long as it is $\ll N_{\mathrm{eff}}$) of quasiparticle *excitations* above these even and odd ground states. We also assume the island is in (weak) tunneling contact with a particle reservoir at $V = 0$, so that the island can contain either an even or odd number of electrons.

By constructing forms in which either the even or odd terms cancel, we can then write down partial partition sums for even and odd n separately as

$$Z_{\mathrm{even}} = [(1 + e^{-\Delta/k_B T})^{N_{\mathrm{eff}}} + (1 - e^{-\Delta/k_B T})^{N_{\mathrm{eff}}}]/2 = Z_{\mathrm{even}}^0$$

$$\qquad (3a)$$

and

$$Z_{\mathrm{odd}} = e^{-E_0/k_B T}[(1 + e^{-\Delta/k_B T})^{N_{\mathrm{eff}}} - (1 - e^{-\Delta/k_B T})^{N_{\mathrm{eff}}}]/2$$

$$= e^{-E_0/k_B T} Z_{\mathrm{odd}}^0 , \qquad (3b)$$

where Z^0 refers to the corresponding partition sum with $E_0 = 0$. These model results should be quite accurate, so long as $k_B T \ll \Delta$. It follows that the relative probability of odd to even numbers of electrons will be

$$\frac{P_{\mathrm{odd}}(Q_0)}{P_{\mathrm{even}}(Q_0)} = \frac{Z_{\mathrm{odd}}}{Z_{\mathrm{even}}} = e^{-E_0/k_B T} \frac{Z_{\mathrm{odd}}^0}{Z_{\mathrm{even}}^0}$$

$$\equiv e^{-E_0/k_B T} e^{-F_0/k_B T} , \qquad (4)$$

where F_0 is the odd/even free energy difference introduced by Tuominen *et al.*,[6] namely

$$F_0 = k_B T \ln(Z_{\mathrm{even}}^0 / Z_{\mathrm{odd}}^0) . \qquad (5)$$

At low temperatures, the leading terms in the binomial expansions dominate, and we find

$$F_0 \approx \Delta - k_B T \ln N_{\mathrm{eff}} = k_B \ln N_{\mathrm{eff}}(T_0^* - T) . \qquad (5a)$$

In this approximation F_0 drops linearly to zero at the $T_0^* = \Delta/k_B(\ln N_{\mathrm{eff}})$ defined above, but if more terms in the expansion are included, $F_0(T)$ goes to zero only asymptotically, but very rapidly, as T exceeds T_0^*. A plot of $F_0(T)$ for typical parameters is shown in Fig. 3(a).

As noted above, the appropriate test for the presence of $2e$ periodicity is the ratio of probabilities $P_{\mathrm{odd}}(Q_0)/P_{\mathrm{even}}(Q_0 + e)$, which reduces to unity for all values of Q_0 if there is overall e periodicity. Recalling that $E_0(Q_0 + e) = -E_0(Q_0)$, after a little algebra one obtains

$$\frac{P_{\mathrm{odd}}(Q_0)}{P_{\mathrm{even}}(Q_0 + e)} = \frac{1 + e^{E_0/k_B T} e^{-F_0/k_B T}}{1 + e^{E_0/k_B T} e^{F_0/k_B T}} \qquad (6)$$

$$\approx 1 - \frac{2}{(1 + e^{-E_0(Q_0)/k_B T})} \left[\frac{F_0(T)}{k_B T} \right] , \qquad (6a)$$

where the second form holds only for $(F_0/k_B T) \ll 1$. Clearly this ratio of probabilities goes to unity for all Q_0 as F_0 goes to zero above T_0^*, while the departure of this ratio from unity below T_0^* gives a measure of the strength of the $2e$ periodicity. This analysis also shows that the crossover from $2e$ to e periodicity largely occurs in a small temperature range of width $\sim T_0^*/\ln N_{\mathrm{eff}}$ near T_0^*. For typical numbers, this is only about a 10% range, which is generally consistent with the data in Fig. 2. It is interesting that both T_0^* and the fractional width of the crossover region approach zero only as $1/\ln N_{\mathrm{eff}}$ as one approaches the thermodynamic limit. Accordingly, the limitation of the observability of the even-odd electron number effect to mesoscopic samples does not stem from the statistical factors involving Δ discussed here, but rather from the need to have $E_c > k_B T$ in order to be able to sweep the electron number by sweeping V_g (or Q_0). With respect to (6), in the macroscopic limit as $E_c/k_B T$ and hence $E_0/k_B T$ go to zero, the ratio of probabilities is independent of V_g and simply reflects the effect of the energy gap in favoring an even number of electrons; there is no observable modulation with period $2e$. Evidently, a treatment of the *observable* consequences of the even-odd parity effect must take account of E_c as well as Δ.

By taking account of the Q_0 dependence of $E_0(Q_0)$, one can get more detail by considering "point-wise periodicity" as distinct from the concept of overall periodicity considered above. For example, $E_0(Q_0) = \pm E_c$ for the even- and odd-integer values of Q_0/e, respectively, so that when comparing the current at Q_0 with that at $(Q_0 + e)$ and $(Q_0 + 2e)$, one finds pointwise $2e$ periodicity at integer values of Q_0/e. However, when Q_0/e is any *half* integer, $E_0(Q_0) = 0$, so that the current at this Q_0 is the same as that at $(Q_0 + e)$ and $(Q_0 + 2e)$, showing pointwise e periodicity. Thus pointwise $2e$ periodicity should be strongest and last to disappear at
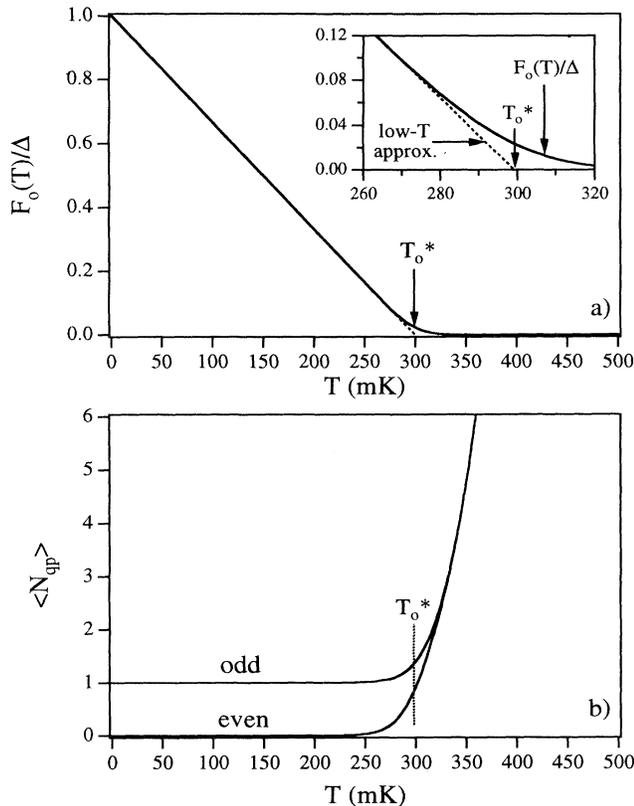
FIG. 3. (a) Normalized even-odd free energy difference $F_0(T)/\Delta$ vs temperature for typical parameters $N_{eff} \approx 15\,000$ and $\Delta/k_B = 2.84$ K, for which $T_0^* \approx 300$ mK. The inset shows a magnified view of the approach to zero near T_0^*. (b) Temperature dependence of the number of quasiparticles for odd and even total numbers of conduction electrons for the same parameters as in (a). Note the rapid convergence to the macroscopic limit just above T_0^*, where $\langle N_{qp} \rangle \sim 1$ for even as well as odd N.

As noted above, computer simulations of $I(Q_0)$ data involve solving a rather complex system of rate equations, rather than the purely equilibrium population analysis given here. Nonetheless, similar conclusions about the crossover from $2e$ to e periodicity are obtained from these much more difficult calculations. In these simulations, one assumes states with definite numbers of particles and rapid relaxation of quasiparticles between tunneling events so that the initial state for each event is a canonical ensemble for the appropriate number of particles. In this fixed-n case, the expectation value of the number of quasiparticles for odd and even n depends only on Δ and T, independent of E_c or Q_0. Plots of these numbers for typical parameters are shown in Fig. 3(b). This figure illustrates nicely that the crossover from $2e$ to e periodicity occurs in the temperature range in which the number of quasiparticles changes from 0 or 1, for even or odd n, respectively, to a rapidly rising value which is independent of the parity.

Although the exact form of the periodic current dependence $I(Q_0)$ depends on whether one is studying an SSS or an NSN system, at low bias voltages the period (e vs $2e$) should depend only on T/T^*. In fact, the T^* values observed with SSS devices by Tuominen et al.[6] and by Amar et al.[10] are very similar to that seen for the NSN device of Fig. 2.

According to Fig. 1(c), if the system follows the minimum energy state while Q_0 is swept, it always stays in states of even n, with two electrons entering or leaving at each crossing point of the parabolas. On the other hand, in Fig. 1(b), electrons enter one at a time, so that the ground state visits both even- and odd-n states, but the system is in even-n states for a greater fraction of the sweep. Lafarge et al.[11] used this effect to extract from their data the temperature dependence of $F_0(T)$, confirming that the dependence (5) expected from this analysis, and plotted in Fig. 3(a), is a good description.

integer values of Q_0/e. This prediction is confirmed in the data of Fig. 2, in that the current at integer values of Q_0/e (i.e., the bottoms of the variations) retains a $2e$-periodic variation at ~ 260 mK, whereas the current at half-integer values of Q_0/e (i.e., the tops of the variations) already appears pointwise e periodic at that temperature.

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