Experimental Evidence for Parity-Based 2e Periodicity in a Superconducting Single-Electron Tunneling Transistor

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(Received 12 June 1992)

We present experimental current-voltage (I-V) measurements on an Al-Al_2O_3-Al single-electron tunneling transistor in the superconducting state. We observe a variety of features which result from Cooper-pair tunneling processes. At low bias voltages and low temperatures we find that the I-V curve is $2e$ periodic with respect to the gate-induced charge. Remarkably, this periodicity persists up to 300 mK, a behavior which we interpret as strong evidence of an incremental free energy which depends on whether the number of electrons on the superconducting island is even or odd.

PACS numbers: 74.50.+r, 73.40.Gk, 73.40.Rw

A number of recent experimental and theoretical efforts have examined single-electron charging effects in small-capacitance tunnel-junction systems [1]. One such system is the single-electron tunneling (SET) transistor. This device is comprised of a small metallic "island" connected to two leads through small-capacitance tunneling junctions and electrostatically coupled to a gate electrode (Fig. 1). In the *normal* state, the current-voltage (I-V) curve is typically nonlinear [2], a result of the large characteristic charging energy, $E_C = e^2/2C_\Sigma$, involved in the tunneling of single electrons ($C_\Sigma = C_1 + C_2 + C_g$). Moreover, the I-V curve is an e-periodic function of the gate-induced charge, $Q_0 = C_g V_g$. This reflects the tendency for the charging energy, $U = Q^2/2C_\Sigma$, to be minimized by keeping the charge variable, $Q = en + Q_0$ (n is the excess charge which has tunneled onto the island), in a range $|Q| < e/2$ by way of single-e transitions in Q (i.e., $\Delta n = \pm 1$). Our focus here is to present experimental results on the SET transistor when the electrodes are *superconducting*. In this case, two additional energies become important: the gap in the quasiparticle density of states, Δ, and the Josephson energy of a single junction, E_J. We observe a variety of unique I-V features that involve Cooper-pair tunneling processes. The most striking of these is a $2e$-periodic Q_0 dependence that persists to surprisingly high temperatures. As we will show, this behavior strongly suggests that the free energy of the system depends on the parity of the total number of electrons on the superconducting island. In other words, the incremental change in free energy for adding an electron to the island depends on whether an even or an odd number of electrons is present.

We fabricate our Al-Al_2O_3-Al samples using electron-beam lithography. Typical junction areas are 60 nm×60 nm and island dimensions are 60 nm wide×2.2 μm long×23 nm thick. The junctions are spaced 1.8 μm apart and the gate electrode is 0.6 μm from the island. Measurements are made in a dilution refrigerator capable of temperatures down to 13 mK. The I-V curves are measured using a four-probe, voltage-biased configuration. The current is measured using a low-input-impedance current preamplifier. To help maintain a low-noise environment, the measurement circuit was equipped with both conventional low-pass and microwave filters. In addition, we used battery-powered voltage sources, and

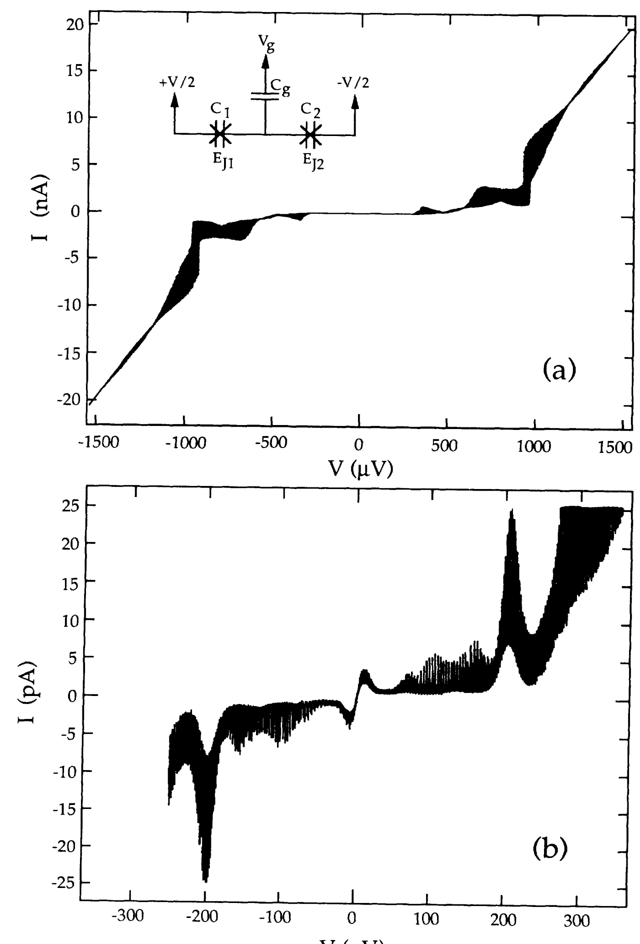

FIG. 1. $I(V,V_g)$ data for a superconducting SET transistor at $T \approx 15$ mK, and zero magnetic field. The current is measured while slowly sweeping V and quickly sweeping V_g, thus filling in the envelope of all possible $I(V)|_{V_g}$ curves. (a) Large current and voltage scale. (b) Small current and voltage scale. (Note current-scale change from nA to pA.) Inset: The bias configuration for the device.

performed the experiment in a shielded room.

Figure 1(a) shows I-V data at 15 mK on large current and voltage scales for a sample with $E_C = 180$ μeV, $E_J = 30$ μeV, $\Delta(0) = 234$ μeV, and $C_g = 41$ aF [3]. In our case, $C_1 \approx C_2$ (≈ 220 aF), $R_{n1} \approx R_{n2}$ (≈ 25 kΩ), and $C_g \ll C_\Sigma$. Since E_J is not negligible compared to E_C for this sample, I-V features resulting from Cooper-pair tunneling are evident. At voltages above $\sim 2\Delta/e$, current can be produced by Josephson-quasiparticle (JQP) cycles, which involve a combination of both Cooper-pair and quasiparticle tunneling events [4,5]. The peaks at 360 and 720 μV (e/C_Σ and $2e/C_\Sigma$, respectively) shown in Fig. 1(a) can be attributed to JQP cycles [6]. These features are e periodic with respect to Q_0 (see below), like those seen in the normal state.

Figure 1(b) shows I-V data on smaller current and voltage scales. Here the voltage is too small ($V \ll 2\Delta/e$) for JQP current to be substantial. We believe that these features arise from Cooper-pair tunneling accompanied by dissipation in the series external impedance [7]. The peak near zero voltage bias corresponds to a "supercurrent." (We use the conventional term "supercurrent" even though it is not a zero-voltage current of the type usually associated with the dc Josephson effect.) The height of the supercurrent peak is a strong function of Q_0, that is, it is a supercurrent whose magnitude is "tunable" with respect to the gate voltage. This dependence was predicted theoretically by others [8]. Other major peaks are seen at 100 and 200 μV whose heights are also strongly Q_0 dependent. In our experiment, the external environment is quite complicated and its impedance is not known in detail, so at this point no attempt has been made to compare the low-voltage I-V curves with theory. Rather, we focus on the I-Q_0 dependence.

The most striking features exhibited by our superconducting SET transistor are shown in the I-Q_0 curves of Figs. 2 and 3. Figure 2 shows that for *all* voltages below 195 μV the current clearly exhibits $2e$ periodicity with respect to Q_0. To our knowledge, the first report of $2e$ periodicity in a SET transistor was by Geerligs *et al.* [9], in which two current peaks (at 20 and 40 μV) in the I-V curve were reported to be $2e$ periodic. Our work is the first to report $2e$ periodicity over a broad range of voltages and temperatures. Figure 2 shows that the $2e$ periodicity is most pronounced for low bias voltages and gradually becomes masked in e periodicity as the voltage is increased into the JQP region, where quasiparticle tunneling is abundant. $2e$ periodicity at low voltage has been predicted at temperatures sufficiently low that Cooper-pair tunneling becomes the sole charge-transfer mechanism [7,8]. From this perspective, e periodicity is expected when the quasiparticle tunneling rate becomes appreciable. Our estimate below shows that this should occur at ~ 100 mK. In surprising contrast to this expectation, the I-Q_0 curves of Fig. 3 show that the $2e$ periodicity at low bias voltages persists to a temperature $T^* \approx 300$ mK. Although this persistence to such high temperatures cannot be explained by a quasiparticle tunneling *rate* argument, below we will obtain a consistent *equilibrium* interpretation of the data in Fig. 3 by (i) incorporating a free energy which depends on the parity of the total number of electrons in the superconducting island, and (ii) predicting the temperature dependence of this free energy by

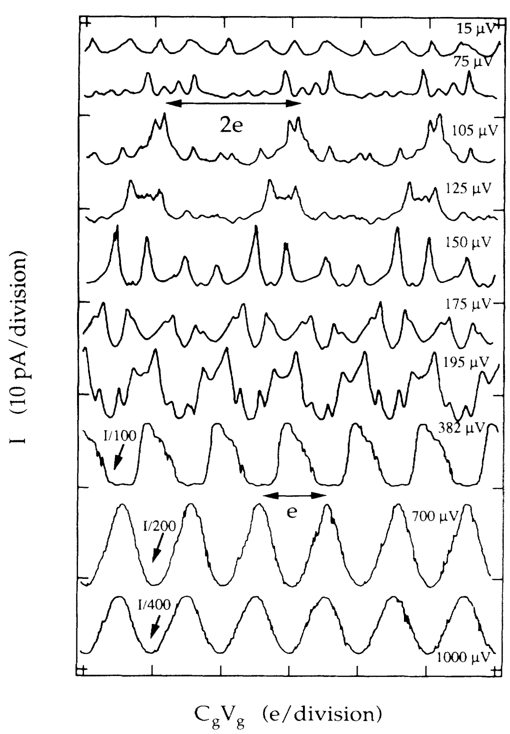

FIG. 2. I-Q_0 curves (displaced arbitrarily for clarity) at 15 mK for various bias voltages (V).

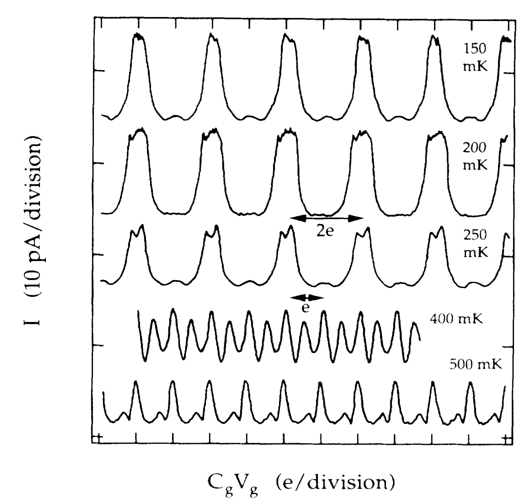

FIG. 3. I-Q_0 curves at $V \approx 100$ μV bias for various temperatures. Other $I(V,V_g)$ data not presented show that the $2e$ periodicity vanishes near 300 mK.

using a statistical mechanical analysis.

The charging energy, $U=Q^2/2C_\Sigma=(ne+Q_0)^2/2C_\Sigma$, is inherently important to tunneling processes in a superconducting SET transistor [1]. When plotted versus Q_0, the charging energy is represented by a series of parabolas [Fig. 4(a)]. Each parabola is for a fixed n and is displaced from its neighbors along the Q_0 axis by e. As long as single-e tunneling events ($n\to n\pm 1$) occur in the experimental time scale, the charging energy is minimized by keeping $|Q|\le e/2$. Thus, the actual charging energy is e periodic in Q_0. However, at sufficiently low temperatures and low voltages single-e (quasiparticle) tunneling events are extremely rare, and only $2e$ (Cooper pair) tunneling events remain. Consequently, only $\Delta n=\pm 2$ transitions occur on the experimental time scale, and the diagram becomes effectively $2e$ periodic in Q_0 [as indicated by the double arrows in Fig. 4(a)], resulting in an I-Q_0 curve with $2e$ periodicity. In this picture, quasiparticle tunneling masks the underlying $2e$ periodicity if the tunneling rate is larger than the Q_0 sweep rate. For the data shown in Fig. 3, the main contribution to the quasiparticle tunneling rate is from the tunneling of thermally excited quasiparticles. (Quasiparticle tunneling from pair breaking need not be considered, since $eV\ll 2\Delta$ and $E_C<2\Delta$.) At a given temperature, the transition rate for $n\to n\pm 1$ should be approximately $\Gamma=E_C/e^2R_{qp}\approx(1/2R_nC_\Sigma)\exp(-\Delta/k_BT)$, where R_{qp} is the subgap resistance. Using a characteristic Q_0 sweep rate of 1 sec^{-1}, we estimate that quasiparticle tunneling should mask the $2e$ periodicity for temperatures above 110 mK. From this perspective it is quite surprising that we observe $2e$ periodicity up to much higher temperatures (≈ 300 mK), where the tunneling rate would be $\sim 10^7$/sec.

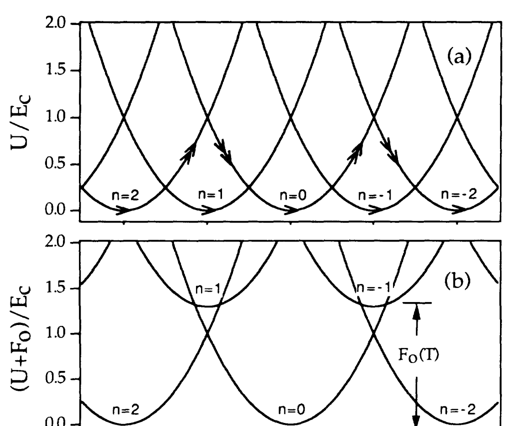

FIG. 4. (a) Charging-energy curves for various charge states. The single arrows show the e-periodic dependence when quasiparticle tunneling is substantial. The double arrows show the $2e$-periodic dependence when quasiparticle tunneling is suppressed. (b) Charging energy plus the parity-dependent energy F_0 (shown for $T=0$ K). We have arbitrarily assumed n is even when the actual number of electrons on the island is even.

To explain this striking disparity, we propose a more complete model in which the system free energy depends on whether the number of electrons on the superconducting island is even or odd. Recently, Averin and Nazarov [10] and Matveev [11] considered this parity dependence of the island's superconducting $T=0$ ground-state energy when calculating the system energy. They predicted that the ground-state energy is higher by an amount $F_0=\Delta$ when the number of electrons on the island is odd (due to the presence of an unpaired electron) compared to when the number is even (all electrons paired). With such a parity-dependent energy, the system free energy is *inherently* $2e$ periodic in Q_0 [Fig. 4(b)], and I-Q_0 should also be $2e$ periodic. However, as the temperature is increased, F_0 is diminished by entropy contributions, as shown below, and the effect fades out.

The grand canonical partition function for excitations above the ground state for a superconducting particle can be separated into two parts, Z_{even} and Z_{odd}, corresponding to an even and an odd number of excitations, respectively. The quantity $F_0(T)$ is the difference in the grand potential calculated for these two cases, that is, $F_0=k_BT\ln(Z_{\text{even}}/Z_{\text{odd}})$. Approximating all relevant excitation energies as Δ, we can separate Z_{even} and Z_{odd} algebraically, with the result

$$F_0(T)=k_BT\ln\frac{(1+e^{-\Delta/k_BT})^{N_{\text{eff}}}+(1-e^{-\Delta/k_BT})^{N_{\text{eff}}}}{(1+e^{-\Delta/k_BT})^{N_{\text{eff}}}-(1-e^{-\Delta/k_BT})^{N_{\text{eff}}}},$$

$$N_{\text{eff}}=2V\rho(0)\int_\Delta^\infty \exp[-(\varepsilon-\Delta)/k_BT]\frac{\varepsilon}{(\varepsilon^2-\Delta^2)^{1/2}}d\varepsilon, \tag{1}$$

where V is the island volume, $\rho(0)$ is the normal-metal density of states (including spin) per unit volume at the Fermi energy, and ε is the excitation energy. N_{eff} is the effective number of states available for excitation, which at low temperatures becomes $N_{\text{eff}}\approx 2\sqrt{2}V\rho(0)$

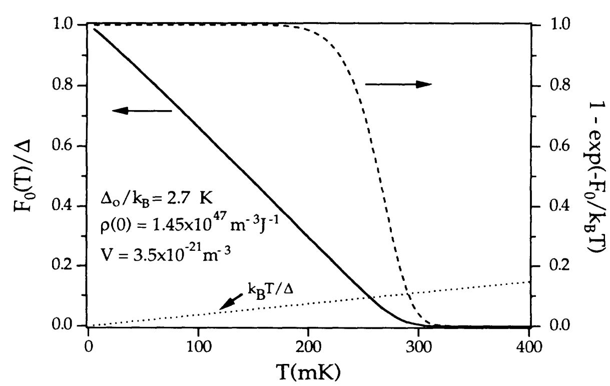

FIG. 5. The parity-dependent energy $F_0(T)$ and the thermal energy k_BT, both normalized to Δ, and the factor $[1-\exp(-F_0/k_BT)]$ vs temperature as calculated for our sample parameters. Note that the latter factor drops rapidly when $F_0(T)\approx k_BT$.

$\times(\Delta k_B T)^{1/2} \approx 10^4$ for our sample. This $F_0(T)$ is plotted in Fig. 5. At low temperatures [where $N_{\rm eff}\exp(-\Delta/k_B T) \ll 1$], F_0 reduces to the simple form

$$F_0(T) \approx \Delta - k_B T \ln(N_{\rm eff}) . \tag{2}$$

This relation clearly shows that F_0 decreases to ~ 0 at a temperature determined by the criterion $k_B T^* = \Delta/\ln(N_{\rm eff})$. Using our sample parameters, we find that $T^* \approx 300$ mK, much below T_c yet much higher than the 110 mK estimate above from the tunneling *rate* argument. This T^* corresponds to the temperature at which the thermal average number of quasiparticles on the island, $N_{\rm qp} = N_{\rm eff}\exp(-\Delta/k_B T)$, becomes of order 1. Since the statistical probability of the system having an odd number of electrons is smaller than that for an even number by a factor of $\exp(-F_0/k_B T)$, we expect the strength of the $2e$ periodicity to vary as $1-\exp(-F_0/k_B T)$. Figure 5 shows that this factor drops quickly to zero in the temperature range in which we observe the disappearance of the $2e$ periodicity in the I-Q_0 curves, that is, near 300 mK, in good agreement with our data shown in Fig. 3. This model predicts a $2e$-periodic I-Q_0 dependence even in the presence of non-negligible quasiparticle tunneling, so long as the system remains near equilibrium. We have made further measurements which show that T^* decreases linearly with H^2 in a magnetic field applied parallel to the plane of the film. This agrees with the parity model since Δ is expected to decrease linearly with H^2 over the range of fields used [12]. To summarize, the quasiparticle rate picture predicts $2e$ periodicity only when the system is out of equilibrium with respect to $n \to n \pm 1$ transitions, while the more complete *parity* model is an *equilibrium* picture that predicts $2e$ periodicity for a small island as long as F_0 is substantial compared to $k_B T$.

We measured a similar sample [$E_C = 78$ μeV, $E_J = 54$ μeV, $\Delta(0) = 186$ μeV] which exhibited only e periodicity. Others have also reported e periodicity in dc measurements at low temperatures where $2e$ periodicity was expected to dominate [9,13]. Thus, it appears that observability of $2e$ periodicity can be hindered by subtle experimental nonidealities. Our analysis points out that once one or more thermally excited quasiparticles are present on the island, F_0 is suppressed very nearly to zero. This suggests that excess quasiparticles (from nonidealities such as normal inclusions [10]) could also diminish F_0, making its manifestation unobservable. Excess quasiparticles also increase the quasiparticle tunneling rate above that expected from the form involving $R_{\rm qp}$. While such mechanisms may *hinder* the observability of $2e$ periodicity, they cannot *cause* it. Thus, those samples that *do* exhibit $2e$ periodicity must reflect the absence of such detrimental effects. We believe that this is evidenced in our experiment.

In conclusion, we have observed features related to Cooper-pair tunneling processes in a superconducting SET transistor, including $2e$-periodic I-Q_0 curves. We have shown that the persistence of $2e$ periodicity to relatively high temperatures is explained by a model that includes a parity-dependent free energy.

The authors would like to give special thanks to P. A. Bobbert for helpful conversations pointing out the possibility of an odd-even energy, and to D. V. Averin and Yu. V. Nazarov for a preprint of Ref. [10]. The authors would also like to acknowledge helpful input from L. I. Glazman, A. M. Goldman, A. E. Hanna, and K. A. Matveev. This research was supported in part by JSEP Grant No. N00014-89-J-1023, ONR Grant No. N00014-89-J-1565, and NSF Grant No. DMR-89-12927.

[1] For a review see D. V. Averin and K. K. Likharev, in *Mesoscopic Phenomena in Solids,* edited by B. L. Altshuler, P. A. Lee, and R. A. Webb (North-Holland, Amsterdam, 1991).

[2] T. A. Fulton and G. J. Dolan, Phys. Rev. Lett. **59**, 109 (1987).

[3] We determine C_Σ from the measured asymptotic voltage offset, $V_{\rm off} = e/C_\Sigma = 360$ μV. The individual junction resistance is found from the series combination $R_{n\Sigma} = 2R_n = 50$ kΩ. The superconducting gap Δ is determined from the single-quasiparticle threshold $V_t = 4\Delta/e$ for $Q_0 = e/2$. We calculate the Josephson coupling energy from the relation $E_J = h\Delta/8e^2R_n$. The gate-to-island capacitance is found from the I-V_g voltage period e/C_g in the normal state (which is obtained by applying a 2.4-T magnetic field).

[4] A. Maassen van den Brink, G. Schön, and L. J. Geerligs, Phys. Rev. Lett. **67**, 3030 (1991).

[5] T. A. Fulton, P. L. Gammel, D. J. Bishop, and L. N. Dunkleberger, Phys. Rev. Lett. **63**, 1307 (1989).

[6] M. T. Tuominen, J. M. Hergenrother, T. S. Tighe, and M. Tinkham (unpublished).

[7] A. Maassen van den Brink, A. A. Odintsov, P. A. Bobbert, and G. Schön, Z. Phys. B **85**, 459 (1991).

[8] K. K. Likharev and A. B. Zorin, Jpn. J. Appl. Phys. Suppl. 3, **26**, 1407 (1987); also *Mesoscopic Phenomena in Solids* (Ref. [1]), p. 213.

[9] L. J. Geerligs, V. F. Anderegg, J. Romijn, and J. E. Mooij, Phys. Rev. Lett. **65**, 377 (1990).

[10] D. V. Averin and Yu. V. Nazarov, preceding Letter, Phys. Rev. Lett. **69**, 1993 (1992).

[11] K. A. Matveev (private communication).

[12] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975); reprinted by Krieger, Florida, 1985, p. 269.

[13] P. Lafarge *et al.*, Z. Phys. B **85**, 327 (1991); L. J. Geerligs *et al.*, Z. Phys. B **85**, 349 (1991).