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Detecting parity effect in a superconducting device in the presence of parity switches

E. T. Mannila,1,* V. F. Maisi,1,2 H. Q. Nguyen,3,4 C. M. Marcus,3 and J. P. Pekola1
1QTF Centre of Excellence, Department of Applied Physics, Aalto University, FI-00076 Aalto, Finland
2Division of Solid State Physics and NanoLund, Lund University, 22100 Lund, Sweden
3Center for Quantum Devices and Station Q Copenhagen, Niels Bohr Institute, University of Copenhagen, Copenhagen, Denmark
4Nano and Energy Center, Hanoi University of Science, VNU, 120401 Hanoi, Vietnam

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We present a superconducting device showing a clear parity effect in the number of electrons, even when there is, on average, a single nonequilibrium quasiparticle present and the parity of the island switches due to quasiparticles tunneling in and out of the device at rates on the order of 100 Hz. We detect the switching by monitoring in real time the charge state of a superconducting island connected to normal leads by tunnel junctions. The quasiparticles are created by Cooper pairs breaking on the island at a rate of a few kilohertz. We demonstrate that the pair breaking is caused by the backaction of the single-electron transistor used as a charge detector. With sufficiently low probing currents, our superconducting island is free of quasiparticles 97% of the time.

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In a superconductor, electrons participating in conduction form Cooper pairs. The minimum energy for an unpaired quasiparticle excitation is \( \Delta \), the superconducting gap, which leads to a free energy difference between states with an even and odd number of electrons in the absence of subgap states. The resulting parity effect is commonly observed in features periodic in \( 2e \), with \( e \) the electron charge, in the transport through an island or by measuring the average charge in an isolated box [1–4]. In thermal equilibrium, the parity effect disappears at temperatures where quasiparticles are excited, around 200 mK for typical micron-scale aluminum structures, as the free energy difference disappears. A clean \( 2e \) periodicity of Coulomb blockade is often taken to suggest a device free of quasiparticles [5–7].

In addition to suppressing the parity effect [8,9], quasiparticle excitations are generally detrimental for superconducting devices. In Josephson junction based qubits, quasiparticles tunneling across the junction cause decoherence [10,11]. For quantum computing using Majorana modes in superconductor-semiconductor hybrids, topological protection is only present when the total fermion parity of the system stays constant. The parity lifetime is a fundamental bound to the coherence time of such a qubit [12–14]. At low temperatures the quasiparticle density \( n_{qp} \) should be exponentially suppressed, and the parity lifetime consequently exponentially long. In practice, often a saturation of \( n_{qp} \) to values several orders of magnitude higher than in thermal equilibrium is observed in experiments on qubits [10,15,16], resonators [17–19], and quantum capacitance [20] and kinetic inductance detectors [21]. Another quantity related to \( n_{qp} \) is the poisoning time between successive quasiparticle tunneling events. Quasiparticle densities or poisoning times can be inferred from transport measurements [13,14,22–24] or qubit coherence times [25]. Even-to-odd transitions from quasiparticle tunneling can also be measured in real time with radio-frequency reflectometry [15,26–28] or in the parity-dependent frequency shift of transmon qubits [16,29]. In this work, we measure in real time quasiparticle tunneling and parity switching on a superconducting island, and observe a parity effect in the parity-dependent occupation probabilities and tunneling rates of charge states. The quasiparticles are created from the backaction of the charge detector [9]. This is a critical issue for Majorana qubit proposals incorporating charge readout [30,31].

We characterize the state of the superconducting island with the excess charge \( N \) and number of excitations \( N_S \) on the island, where \( N \) and \( N_S \) are integers of the same parity, following Ref. [23], which provides the quantitative details of the model. The relevant processes in our system are shown in Fig. 1(a). If there are two or more excitations on the island, they can recombine to a Cooper pair with rates \( \Gamma_{rec}(N_S) \). We include recombination via the electron-phonon coupling. Cooper pairs on the island can break, creating two excitations, with a rate \( \Gamma_{pb} \), assumed independent of the state of the island. The thermal electron-phonon pairbreaking rate is vanishingly small at the temperatures of the experiment, so this rate arises from nonequilibrium conditions. We directly detect the quasiparticle tunneling events between the superconducting island and normal metal leads at temperature \( T_N \), which change both \( N \) and \( N_S \) by one. If excess quasiparticles are present (the superconductor temperature \( T_S > T_N \)) but \( k_B T_S \ll \Delta \), the rate for quasiparticles tunneling out of the island \( \Gamma_{qp}(N_S) \equiv \Gamma(N \rightarrow N \pm 1, N_S \rightarrow N_S - 1) \) depends, for a range of energy gains, only on the quasiparticle density \( n_{qp} = \sqrt{2\pi D(E_F)\Delta k_B T_S e^{-\Delta/k_B T_S}} \) or \( N_S = n_{qp} V \) before the tunneling event as [32]

\[
\Gamma_{qp}(N_S) = \frac{N_S}{2e^2 R_T D(E_F) V}.
\]
Here, $R_T$ is the resistance of the tunnel junction, $k_B$ the Boltzmann constant, and $D(E_F) = 2.15 \times 10^{17} \text{ J}^{-1} \text{ m}^{-2}$ the normal density of states (including spin) at the Fermi level. In particular, a single quasiparticle in the island in volume $V = 550 \text{ nm} \times 2 \mu \text{m} \times 50 \text{ nm}$ and $R_T = 15.6 \text{ M} \Omega$ corresponds to $\Gamma_{ph} = 110 \text{ Hz}$. Tunneling events which increase $N_k$ are suppressed by the superconducting gap when $N$ is close to the gap offset $N_g$.

If $\Gamma_{ph}$ is zero in the model above, we recover the thermal equilibrium case. The free energies of the charge states $N$ of a superconducting island are $E = E_C(N - n_g)^2 + F(T_3) \times N$ mod 2, which includes, in addition to the contribution of the charging energy $E_C = c^2/2C_S$ with $C_S$ the total capacitance of the island, the free energy cost $F(T_3) \approx k_B T_3 \ln |D(E_F) / |V \Delta |$ of an unpaired excitation [1,3]. The approximation is valid when $k_B T_3 \ll \Delta$. These free energies are sketched in Fig. 1(b) against $n_g = C_S V_g / e$, where $V_g$ is the voltage applied to a gate electrode coupled via capacitance $C_S$. When $F(T_3) > E_C$, as in our devices below 120 mK, the ground state has even parity, and we expect to see only two-electron Andreev tunneling events. The states with odd $N$ should become significantly occupied only above the temperature $T_0 = \Delta / [k_B \ln |V D(E_F) / |V \Delta |] \approx 190 \text{ mK}$, where a single quasiparticle is thermally excited on the island.

Our device, shown in Fig. 2(a), is a single-electron transistor (SET) with a superconducting aluminum island connected to normal metal copper leads with aluminum oxide tunnel barriers a few nanometers thick. The capacitively coupled charge detector is another SET, but with a copper island and aluminum leads. The devices were fabricated with standard electron-beam lithography and three-angle evaporation on thermally oxidized silicon substrates. We have measured two similar devices, samples A and B. The energy gap $\Delta = 206 \mu \text{eV}$ (210 $\mu \text{eV}$), total tunnel resistance of the two junctions 70 $\text{M} \Omega$ (40 $\text{M} \Omega$), and the charging energy $E_C = 0.33 \Delta = 68 \mu \text{eV}$ (0.45$\Delta = 95 \mu \text{eV}$) of sample A (B) were determined by fitting the current-voltage characteristics as shown in Fig. 2(b).

For the electron counting experiments, the superconducting island was kept at zero bias. The island acts as a single-electron box connected to normal leads through the parallel resistance of the two junctions $R_T = 15.6 \text{ M} \Omega$ (8.9 $\text{M} \Omega$), with both devices having unequal tunnel junctions whose areas and resistances differ by a factor of 2. Sample A was measured at 60 mK in a DC measurement setup sketched in Fig. 2(a), where we directly record the amplified detector current $I_{det}$. Sample B was measured at 25 mK in a setup where the detector was used as an RF-SET [33,34].

Figures 2(c)–2(j) show real-time traces of the charge detector output at $n_g = 0 ([c,d,g,h])$ and $n_g = 1 ([e,f,i,j])$. In sample A [Figs. 2(c)–2(f)], three charge states are always occupied for a significant fraction of time, even though the charging energy ($E_C/k_B \approx 800 \text{ mK}$) is much larger than the bath temperature. Most of the transitions are single-electron
transitions. At \( n_g = 0 \) the state at \( I_{det} = 200 \) pA corresponding to \( N = 0 \) is more occupied than \( N = \pm 1 \) at 150 and 250 pA, while at \( n_g = 1 \) the state \( N = 1 \) (220 pA) has a lower occupation probability than \( N = 0 \) (170 pA) or \( N = 2 \) (270 pA). In sample B [Figs. 2(g)-2(j)], where using smaller detector currents is possible (see Supplemental Material [34]), the odd states are occupied with almost two orders of magnitude lower probability, suggesting a much lower density of nonequilibrium quasiparticles.

We measure time traces across a range of \( n_g \) and extract the occupation probabilities of each charge state [Fig. 3(a)]. At all values of \( n_g \) there are three or four charge states visible. This can be explained with a nonequilibrium quasiparticle population. Intuitively, if there is a quasiparticle with energy \( \Delta = 3E_C \) on the island, the energy cost of charging the island with an additional electron is possible to overcome. If \( n_g = 0.5 \), the charging part of the energy \( E_C(N - n_g)^2 \) is smaller than \( \Delta \) for the states \( N = -1, 0, 1, \) and 2, which are the states observed. However, even in this nonequilibrium situation, the most probable state has always even parity as in thermal equilibrium.

The tunneling rates \( \Gamma_{N \rightarrow M} \) of single-electron transitions [Figs. 3(b) and 3(c)] are determined from the time traces. For each transition, there is a range in \( n_g \) where the rate is independent of the energy gained in the transition, since the tunneling rates are dominated by excess quasiparticles in the superconductor [32]. The measured rates \( \Gamma_{N \rightarrow N \pm 1} \) are a weighted average of the rates \( \Gamma_{qp}(N_S) \) over the \( N_S \) states for a given \( N \) and thus directly proportional to a mean quasiparticle population \( N_S \). At the plateaus, \( \Gamma_{qp}(N_{S, odd}) = 160 \) Hz for odd \( N \) and \( \Gamma_{qp}(N_{S, even}) = 80 \) Hz for even \( N \), and thus the mean quasiparticle population depends on parity. The ratio \( \Gamma_{qp}(N_{S, odd})/\Gamma_{qp}(N_{S, even}) = (N_{S, odd})/(N_{S, even}) \approx 2 \) means that \( (N_{S, even}) \geq 0.5 \) and at least two quasiparticles must be present for 25% of the time in even charge states. To maintain such a quasiparticle population, quasiparticles must be generated either from Cooper pairs breaking or electrons tunneling from the leads with a total rate on the same order as with what they tunnel out or recombine. The expected recombination rate is 9.7 kHz for \( N_S = 2 \) and larger for more quasiparticles (assuming the electron-phonon coupling constant \( \Sigma = 1.8 \times 10^9 \) W K\(^{-2}\) m\(^{-3}\) [23]), two orders of magnitude larger than the measured tunneling rates. A Cooper pair breaking rate much larger than the tunneling rates is then needed to produce the observed excess quasiparticles, in contrast to models where quasiparticles tunnel in from the leads [8,39]. Any broken Cooper pair will, on average, recombine on the superconducting island before having time to tunnel out. The quasiparticle population on the island is determined by the competition between pair breaking and recombination, with the tunnel contacts only serving to probe the resulting quasiparticle density.

We calculate numerically the transition rates between different \( (N, N_S) \) states as in Ref. [23], which gives the quasiparticle tunneling and recombination rates and a corresponding rate equation, and solve for the steady-state occupation probabilities. The solid lines in Fig. 3(a) are the occupation probabilities for each charge state with any number of excitations, while the solid lines in Figs. 3(b) and 3(c) are the average rates between different charge states. To reproduce the significant occupation probabilities of odd charge states, we need to include a Cooper pair breaking rate \( \Gamma_{pb} = 4.6 \) kHz. We are able to reproduce quantitatively all the features in the transition rates and occupation probabilities with only \( \Gamma_{pb} \) as a free parameter in our model. Other parameters are either determined from independent measurements \((R_T, \Delta, E_C)\) or they are known literature values \([\Sigma, D(E_F)]\). Some of the transitions interpreted as two successive single-electron events might be two-electron Andreev events, which are not included in the model. Yet their influence to obtained results is weak: assuming successive transitions from \( N \) to \( N \pm 2 \) occurring within 1 ms to be Andreev events decreases the inferred single-electron tunnel rates only by a few percent.

The finite bandwidth of the detector (a few kilohertz) mostly causes the measured rates to underestimate the true rates at the quasiparticle-induced plateaus by 10%–20% [40] and does not affect our main conclusions. In Fig. 3, the simulated rates are corrected to account for finite bandwidth using the model of Ref. [40].

The time-averaged number of excitations from the simulations is \((N_S) = 0.86\) in even charge states and 1.6 in odd states. The equilibrium temperature where the parity effect is expected to disappear corresponds to a single quasiparticle being excited. It is somewhat against the common view that the parity effect is clearly visible even with a single nonequilibrium excitation present and quasiparticles continuously tunneling in and out of the device, as we demonstrate here. The ratio of the pair breaking and recombination rates \( \Gamma_{pb}/\Gamma_{rec} \) determines the quasiparticle density. However, we cannot determine these two rates independently: the agreement between simulations and experiment in Fig. 3 remains equally good if \( \Gamma_{pb} \) and the electron-phonon coupling constant \( \Sigma \) setting \( \Gamma_{rec} \) are scaled up or down but by the same factor.
We now turn to the origin of the Cooper pair breaking rate observed. We repeat the measurement of time traces versus $n_q$ in sample A at different $V_{b, det}$ between 385 and 560 $\mu$V and extract $\Gamma_{qp}$ from the quasiparticle-induced plateaus, shown as a function of $I_{det}$ in Fig. 4. The tunneling rate increases linearly with detector current, but $\Gamma_{qp}$ extrapolates to 0 at $I_{det} = 0$. Solid lines are calculated $\langle N_q \rangle$ for $N = 0$ (red) and $N = 1$ (blue) at $n_q = 0.5$ as a function of the Cooper pair breaking rate $\Gamma_{pb} = A_{det}/e$. Inset: $\Gamma_{qp}(\langle N_q \rangle)$ measured two weeks earlier extrapolates to 30 Hz at $I_{det} = 0$, which corresponds to $\Gamma_{pb} \approx 1$ kHz.

FIG. 4. Quasiparticle tunneling rates $\Gamma_{qp}$ in sample A corresponding to events out of charge states with even $\Gamma_{qp}(\langle N_q \rangle_{even})$, circles) and odd $\Gamma_{qp}(\langle N_q \rangle_{odd})$, triangles) parity. $\Gamma_{qp}$ corresponds to a mean quasiparticle number $\langle N_q \rangle$ according to Eq. (1). The rates decrease with decreasing detector current, and $\Gamma_{qp}$ extrapolates to less than 15 Hz with $I_{det} = 0$. Solid lines are calculated $\langle N_q \rangle$ for $N = 0$ (red) and $N = 1$ (blue) at $n_q = 0.5$ as a function of the Cooper pair breaking rate $\Gamma_{pb} = A_{det}/e$. Inset: $\Gamma_{qp}(\langle N_q \rangle_{even})$ measured two weeks earlier extrapolates to 30 Hz at $I_{det} = 0$, which corresponds to $\Gamma_{pb} \approx 1$ kHz.

In conclusion, we have observed a clear parity effect in the occupation probabilities and tunneling rates of the charge states of a superconducting island, even in the presence of a single nonequilibrium excitation and frequent parity switches. The excitations are generated by Cooper pairs breaking on the superconducting island, and the quasiparticles almost always recombine before tunneling out. The poisoning time or parity lifetime of the island—defined as the time between quasiparticle tunneling events—can be long, even though the island is still poisoned in the sense of quasiparticles being present. The Cooper pair breaking is caused by the backaction of the charge detector, which can be minimized by reducing the detector current. We expect that in future experiments, the statistics of electron counting yields access to the recombination and pair breaking rates independently of each other as in the spin-blockade studies [45–47], where the electron occupation preserving spin-flip rate was determined from the tunneling statistics.

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