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Author(s):   Maisi, V. F. & Lotkhov, S. V. & Kemppinen, A. & Heimes, A. & Muhonen, J. T. & Pekola, Jukka

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Excitation of Single Quasiparticles in a Small Superconducting Al Island Connected to Normal-Metal Leads by Tunnel Junctions

V. F. Maisi,[1,2,*] S. V. Lotkhov,[3] A. Kemppinen,[2] A. Heimes,[4] J. T. Muhonen,[1,5] and J. P. Pekola[1]

[1]*Low Temperature Laboratory (OVLL), Aalto University School of Science, P.O. Box 13500, 00076 Aalto, Finland*
[2]*Centre for Metrology and Accreditation (MIKES), P.O. Box 9, 02151 Espoo, Finland*
[3]*Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany*
[4]*Institut für Theoretische Festkörperphysik, Karlsruher Institut für Technologie, Wolfgang-Gaede-Strasse 1,
D-76128 Karlsruhe, Germany*
[5]*Centre for Quantum Computation and Communication Technology, School of Electrical Engineering and Telecommunications,
University of New South Wales, Sydney, New South Wales 2052, Australia*
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We investigate the dynamics of individual quasiparticle excitations on a small superconducting aluminum island connected to normal metallic leads by tunnel junctions. We find the island to be free of excitations within the measurement resolution. This allows us to show that the residual heating, which typically limits experiments on superconductors, has an ultralow value of less than 0.1 aW. By injecting electrons with a periodic gate voltage, we probe electron-phonon interaction and relaxation down to a single quasiparticle excitation pair, with a measured recombination rate of 16 kHz. Our experiment yields a strong test of BCS theory in aluminum as the results are consistent with it without free parameters.

The quasiparticle excitations describing the microscopic degrees of freedom in superconductors freeze out at low temperatures, provided no energy exceeding the superconductor gap Δ is available. Early experiments on these excitations were performed typically close to the critical temperature with large structures so that N_S, the number of quasiparticle excitations, was high [1–9]. Later on, as the fabrication techniques progressed, it became possible to bring N_S close to unity to reveal the parity effect of electrons on a superconducting island [10–14]. In recent years, the tunneling and relaxation dynamics of quasiparticles, which we address in this Letter, have become a topical subject because of their influence on practically all superconducting circuits in the low temperature limit [15–22].

We study the quasiparticle excitations on a small aluminum island shown in Fig. 1(a). The island is connected via a thin insulating aluminum oxide layer to two normal metallic copper leads to form a single-electron transistor (SET) allowing quasiparticle tunneling. By measuring the tunnel current against source-drain bias voltage V_b and offset charge n_g of the island, we first show that the island can be cooled down to have essentially no quasiparticle excitations. Then we intentionally inject excitations to the superconductor and probe electron-phonon interaction, the inherent relaxation mechanism of a superconductor, down to a single quasiparticle pair.

The current I through the SET is governed by sequential tunneling of single quasiparticles and it exhibits Coulomb diamonds which overlap each other because of the superconductor energy gap [23,24], observed for our structure as a region bounded by the red sawtooths in Fig. 1(b). In the subgap regime, $|eV_b| < 2\Delta$, the current should be suppressed if there are no quasiparticle excitations present. Nonetheless, we observe a finite current which has a period twice as long in n_g as compared to the high bias region, a unique feature of a superconducting island due to Cooper
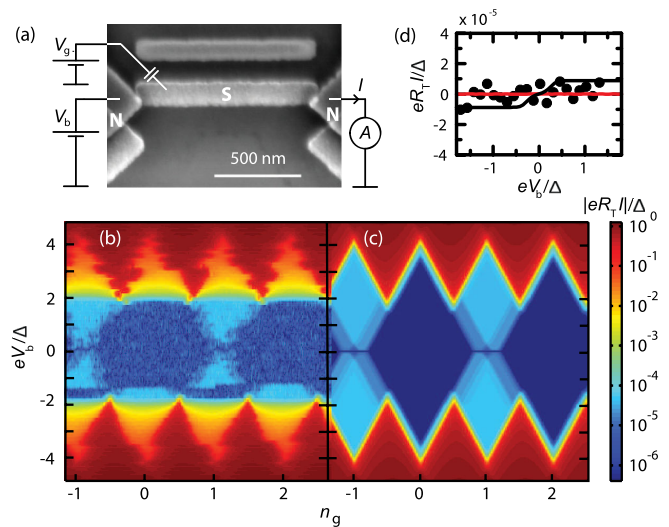

FIG. 1 (color online). (a) Scanning electron micrograph of the sample studied. It is biased with voltage V_b and a gate offset voltage V_g is applied to a gate electrode (not shown) to obtain a gate offset charge $n_g = C_g V_g / e$, where C_g is the gate-island capacitance. (b) Measured source-drain current I as a function of bias and gate voltages. (c) Calculated current based on sequential single-electron tunneling model. (d) Measured current at $n_g = 0$ is shown as black dots. Black line is calculated assuming a quasiparticle generation rate of 2 kHz, and the red line is calculated assuming a vanishing generation rate.

pairing of electrons. The current is caused by a single electron unable to pair in the condensate and hence remaining as an excitation. This parity effect has been observed in the past in similar structures [11–13] but typically with two-electron Andreev tunneling being the main transport process. We focus on devices where Andreev current is suppressed by large charging energy, $E_c > \Delta$, which makes two-electron tunneling energetically unfavorable compared to that of a single quasiparticle [24–26]. In this case, the transport is dominated by single-electron processes allowing simple and direct probing of the quasiparticle excitations without the interfering multielectron tunneling.

For a quantitative description of the transport characteristics, we performed a numerical simulation of the device operation shown in Fig. 1(c). To describe simultaneously the charging of the island with electrons and the excitations involved in the superconducting state, we assign the probability $P(N, N_S)$ for having N excess electrons and N_S quasiparticle excitations on the island. The time evolution of $P(N, N_S)$ is described by a master equation

$$\frac{d}{dt} P(N, N_S) = \sum_{N', N_S'} \Gamma_{N' \to N, N_S' \to N_S} P(N', N_S'), \quad (1)$$

where $\Gamma_{N' \to N, N_S' \to N_S}$ is the transition rate from N', N_S' to N, N_S. For $N' = N$, $N_S' = N_S$ we insert a rate $\Gamma_{N \to N, N_S \to N_S} = -\sum_{N', N_S'} \Gamma_{N \to N' \neq N, N_S \to N_S' \neq N_S}$, which is a sum of all rates out from the state (N, N_S). These rates are set by electron tunneling between the island and the leads, Cooper pair breaking, and recombination of quasiparticles.

Tunneling rates are calculated by the standard first order perturbation theory [23–26]. Electrons tunneling into the superconducting island to states with energy $E > \Delta$ in the semiconductor model [27] increase the quasiparticle number N_S and electron number N by one. Electrons tunneling to states $E < -\Delta$ increase N and decrease N_S by one by filling a hole. Similarly, electrons tunneling out from $E > \Delta$ decrease both N and N_S while electrons from states $E < -\Delta$ increase N_S and decrease N. For a detailed description of these rates, see the Supplemental Material [28].

The tunneling rates depend on N_S, which is the number of excited quasiparticle states on the superconducting island. The nonequilibrium quasiparticle distribution function $f_S(E)$ gives the probability that such a quasiparticle state with energy E is occupied. In general, f_S has a complicated form as a function of energy. However, due to the fact that quasiparticles are injected close to the gap, the resulting tunneling and recombination are not sensitive to the functional form of f_S. Because of symmetry, branch imbalance [3,27] is not created in our system and therefore we parametrize the quasiparticle number by an effectively increased temperature T_S and a Fermi distribution in the case of f_S. This gives the relation

$$N_S = \sqrt{2\pi} D(E_F) V \sqrt{\Delta k_B T_S} e^{-\Delta/k_B T_S}, \quad (2)$$

where $D(E_F) = 1.45 \times 10^{47}$ J^{-1} m^{-3} is the density of states in the normal state [29] and V the volume of the island. For the normal metallic leads we use Fermi distribution with $T_N = 60$ mK, equal to the base temperature of the cryostat.

For the steady-state represented by Fig. 1(c), we solve Eq. (1) with $(d/dt)P(N, N_S) = 0$ and calculate the current as an average of the tunneling rates weighted by the probabilities $P(N, N_S)$. The parameter values of sample A, $E_c = 240$ μeV, $\Delta = 210$ μeV, and tunneling resistances $R_{T1} = 220$ kΩ and $R_{T2} = 150$ kΩ for the two junctions were used in the simulations. They were determined from measurements in the high bias regime ($|eV_b| > 2\Delta$) and hence their values are independent of the subgap features.

The simulation of Fig. 1(c) reproduces the behavior observed in the experiments. The relaxation rate of a single quasiparticle excitation in or out from the island via tunneling is expected to be $\Gamma_{qp} \equiv \Gamma_{N+1 \to N, 1 \to 0} = [2e^2 R_T D(E_F) V]^{-1} = 190$ kHz, where we have used the measured dimensions for $V = 1.06$ μm $\times$ 145 nm $\times$ 25 nm. R_T^{-1} is the average of the two conductances R_{T1}^{-1} and R_{T2}^{-1}. This rate reflects the injection, $\propto R_T^{-1}$, in a system with the number of states per energy being $D(E_F)V$. From the fit in the subgap regime, we obtain $\Gamma_{qp} = 150$ kHz, consistent with the prediction. The value of Γ_{qp} affects only the value of current on the light blue plateau of Fig. 1(c), not the actual form or size of the terrace. As our simulation based on sequential tunneling reproduces all the features in the subgap regime, we confirm that the two-electron periodicity originates from single-electron tunneling and the operation is essentially free of multielectron tunneling processes. At odd integer values of n_g, the characteristic feature of Andreev tunneling would be a linear-in-V_b current at low bias voltages and a subsequent drop [12,30], which is absent in our data. The leakage current in the subgap region does not vanish even in the zero temperature limit but remains essentially the same as presented in Fig. 1. Therefore, all quasiparticle excitations cannot be suppressed at finite bias voltages by lowering the temperature, if n_g is close to an odd integer. At $|eV_b| > 2\Delta$, incoherent cotunneling is activated, which we do not consider here. Also in Fig. 1(b), there is a change of parity, $\Delta n_g = 1$, once during the measurement due to an unknown reason [12].

At even integer values of n_g, we have ideally no current flow as all electrons are paired. If Cooper pair breaking would take place on the island with rate $\Gamma_e \equiv \Gamma_{N \to N, 0 \to 2}$, we would obtain two quasiparticle excitations in the superconductor. One of these excitations can then relax by tunneling to the leads followed by tunneling of a new excitation to neutralize the offset charge, similarly to the odd n_g case described above. This cycle would continue until the two quasiparticles recombine to a Cooper pair. As the recombination rate $\Gamma_{rec} = \Gamma_{N \to N, 2 \to 0} = 16$ kHz, discussed below, is slower than the rates in the cycle, we have

several electrons tunneling through the device for each broken pair, hence amplifying the signal. The resulting current through the SET is then $I/e = 4\Gamma_{qp}\Gamma_e/(\Gamma_{rec} + \Gamma_e)$, where $4\Gamma_{qp}$ is the tunneling rate of the two quasiparticles to the forward direction for the two junctions and $\Gamma_e/(\Gamma_{rec} + \Gamma_e)$ the probability to be in the state with $N_S = 2$. Therefore, at low excitation rates, $\Gamma_e \ll \Gamma_{rec}$, on average $4\Gamma_{qp}/\Gamma_{rec} = 40$ electrons tunnel through the device for each broken pair. With this model, we obtain an upper bound $\Gamma_e = 2$ kHz for the pair breaking rate under our experimental conditions based on simulations shown in Fig. 1(d). This rate corresponds to energy absorption at less than $2\Gamma_e\Delta = 0.1$ aW power on the superconducting island. It is in agreement with expectations: The SET is protected against high frequency photons causing pair breaking with an indium sealed Faraday cage. Also, the pair breaking caused by phonons is expected to be orders of magnitude smaller than the determined upper bound.

Under constant biasing conditions, there is at most one quasiparticle present in the subgap regime at low temperatures. The nontunneling relaxation on the island is then not possible, since recombination would call for two excitations. Therefore, the static case can be described by pure tunneling without other relaxation processes. To study recombination of two quasiparticles into a Cooper pair, we injected intentionally more quasiparticles to the island. The injection was done by a periodic drive of the gate voltage. By changing n_g, we change the potential of the superconducting island and either pull quasiparticle excitations into the island when the potential is lowered or create hole-type excitations as potential is raised and quasiparticles tunnel out. The number of injected quasiparticles and the number of quasiparticles on the island can then be determined from the resulting current curves with the help of simulations. In the experiment we approach two different limits which we will discuss in the following: When the pumping frequency is high, N_S is large. Such a situation can be described by a *thermal model*, i.e., by an increased time-independent effective temperature of the superconducting island. In the opposite limit of low frequency, N_S is small. Then the thermal model fails and we have to account for the exact time-dependent number of quasiparticles.

In Fig. 2(a) we show the measured current for three different values of bias voltage V_b. The gate drive is sinusoidal around $n_g = 1/2$ with amplitude A_g expressed in units of e/C_g and frequency $f = 1$ MHz corresponding to fast pumping. Without accumulation of quasiparticles to the island, the current would show quantized plateaus with spacing ef, similar to the hybrid turnstile [23]. In the curves of Fig. 2 we show the amplitude region where the first plateau with $I = ef$ should form. The plateaulike regime within $0 < A_g < 1$ corresponds approximately to the amplitude range of a Coulomb diamond in the stability diagram of the SET. As the island has a surplus of
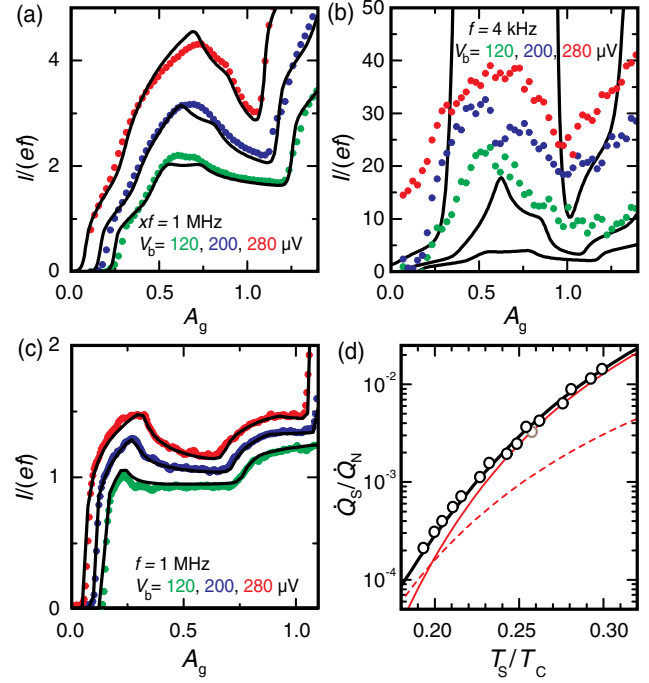

FIG. 2 (color online). (a),(b) Measured current of sample A against gate voltage amplitude A_g at $f = 4$ and 1000 kHz at bias voltage values $V_b = 120, 200, 280$ μV shown as blue, red, and green dots, respectively. Black lines show simulations assuming an elevated temperature on the superconducting island. (c) Similar measurement for sample B at $f = 1$ MHz. (d) The electron-phonon heat flux in the superconducting state normalized by that in the normal state extracted from the measurements (black circles). Temperature is expressed with respect to the critical temperature $T_C = \Delta/1.76k_B$. The theoretical result of Eq. (3) is shown by the black line. Solid and dotted red lines show the recombination and scattering part of Eq. (3) correspondingly. The open gray symbol is from sample B.

quasiparticles, i.e., it is heated up, the current at the plateau area is substantially higher than ef and even nonmonotonic. The nonmonotonic behavior arises as the system stays part of the cycle, towards large values of A_g, in the state ($N = 2, N_S = 0$), where no current flows (for the case $E_c \sim \Delta$), or Coulomb blockade forbids quasiparticle relaxation by tunneling at certain gate values (for $E_c \sim 2\Delta$). We now use the thermal model where the heat injection to the island by electron tunneling is balanced by electron-phonon interaction. The heat flux into the phonon bath is given by

$$\dot{Q}_{e\text{-ph}} = \frac{\Sigma V}{24\zeta(5)k_B^5} \int_0^\infty d\epsilon \epsilon^3 [n(\epsilon, T_S) - n(\epsilon, T_P)]$$
$$\times \int_{-\infty}^\infty dE n_S(E) n_S(E + \epsilon) \left(1 - \frac{\Delta^2}{E(E + \epsilon)}\right)$$
$$\times [f_S(E) - f_S(E + \epsilon)], \tag{3}$$

where Σ is the material constant for electron-phonon coupling, $\zeta(z)$ the Riemann zeta function, $n_S(E)$ the BCS

density of states, and $n(\epsilon, T) = \{\exp[\epsilon/(k_B T)] - 1\}^{-1}$ the Bose-Einstein distribution of the phonons at temperature T_P. Equation (3) is obtained by kinetic Boltzmann equation calculations [31,32]. For an alternative derivation, see Supplemental Material [28]. The simulations based on the thermal model are shown as black lines in Fig. 2(a). The electron-phonon coupling constant $\Sigma = 1.8 \times 10^9 \; \mathrm{W \, K^{-5} \, m^{-3}}$, used in simulations, was measured in the normal state, where $\dot{Q}_{e\text{-ph}} = \Sigma V (T_N^5 - T_P^5)$ and T_N is the electron temperature [33].

We expect the thermal model to be a good approach if $N_S \gg 1$. With the high frequency and large amplitude drive in Fig. 2(a), we have $N_S \sim 10$ quasiparticles present for $A_g \sim 1$, suggesting that the thermal model is adequate for these data. If the frequency is lowered to $f = 4$ kHz, shown in Fig. 2(b), the thermal model fails as N_S approaches unity. As a further proof of the overheating, we repeated the high frequency measurement using sample B with measured parameter values $E_c = 620 \; \mu\mathrm{eV}$, $\Delta = 270 \; \mu\mathrm{eV}$, $R_{T1} = 1800 \; \mathrm{k\Omega}$, $R_{T2} = 960 \; \mathrm{k\Omega}$, and $V = 800 \times 60 \times 15 \; \mathrm{nm}^3$. The result is shown in Fig 2(c). Again, the simulations (black lines) are able to reproduce all nontrivial features of the measured curves. As a summary of the thermal model fits, we repeated the measurement of Fig. 2(a) at different frequencies and determined by numerical simulations the temperature of the superconducting island and the heat injected into it based on the measured current. The results are shown in Fig. 2(d) as black circles. The results match well with the expected electron-phonon coupling of the superconductor, Eq. (3), presented as the solid black line.

Next, we show that a rigorous way to describe a low number of excitations is to consider them explicitly with Eq. (1). We measured the characteristics at four frequencies $f = 4$, 10, 100, and 1000 kHz at $V_b = 280 \; \mu\mathrm{V}$ and again at the first plateau region, shown in Fig. 3. The thermal model is presented now as solid gray lines. In numerical calculations based on Eq. (1), we keep track of the number of excitations during the cycle and take into account the recombination rates. For low temperatures, $T_P \ll T_S \ll \Delta/k_B$, the heat flux of Eq. (3) decomposes to recombination terms, proportional to $e^{-2\Delta/k_B T_S}$, and scattering terms, proportional to $e^{-\Delta/k_B T_S}$, yielding

$$\dot{Q}_{\mathrm{rec}} = \frac{\pi V \Sigma}{3\zeta(5)k_B^5}\left[k_B T_S \Delta^4 + \frac{7}{4}(k_B T_S)^2 \Delta^3 \right] e^{-2\Delta/k_B T_S}$$

$$\dot{Q}_{\mathrm{sc}} = V\Sigma T_S^5 e^{-\Delta/k_B T_S}. \qquad (4)$$

Their contributions are presented in Fig. 2(d), and the leading order terms are consistent with the lifetimes given in Ref. [5]. Whereas the scattering does not change the number of quasiparticles N_S, recombination leads to transitions $N_S \to N_S - 2$. We account for this process by including the recombination rate $\Gamma_{N\to N, N_S \to N_S - 2} = \dot{Q}_{\mathrm{rec}}(N_S)/2\Delta \approx \Sigma \Delta^2 N_S^2/[12\zeta(5)D(E_F)^2 k_B^5 V]$, $N_S \geq 2$,

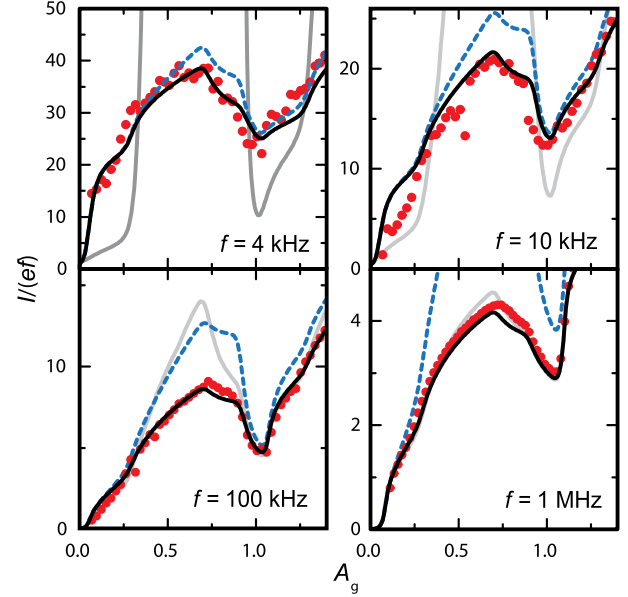FIG. 3 (color online).   Red circles show the measured current for $f = 4$–1000 kHz at $V_b = 280 \; \mu\mathrm{V}$. Black lines are simulations based on Eq. (1). Recombination rates are taken to match the heat flux in the regime where the thermal model applies [Eq. (3)]. Dotted blue lines are calculated with vanishing electron-phonon relaxation rate and solid gray lines with the thermal model.

where the relation between the effective temperature T_S in Eq. (4) and the exact quasiparticle number N_S is given by Eq. (2). The solid black lines of Fig. 3 are calculated with the same value of Σ as obtained in the normal state. Blue dotted lines show similar simulations where electron-phonon relaxation is disregarded.

With the simulations based on instantaneous quasiparticle number we can reproduce the experimental features precisely with no free parameters in the calculation. At the lowest frequency, $f = 4$ kHz, we have only one quasiparticle present for most of the time. Hence, the curves are not sensitive to the recombination. As frequency is increased, the simulations without electron-phonon relaxation deviate from the experimental data. For $f = 10$ kHz we probe the recombination rate of a single qp pair only, $\Gamma_{\mathrm{rec}} = \Gamma_{N\to N, 2\to 0} = 16$ kHz. We checked this by artificially changing the recombination rates for $N_S > 2$, without any significant difference in the curves. At higher frequencies, the recombination for $N_S > 2$ becomes significant as well. The results of the two models approach each other and the thermal model becomes valid.

In summary, a small superconducting island at low temperatures has allowed us to study the dynamics of single electronic excitations and their relaxation. Under quiescent conditions we found a vanishing Cooper pair breaking rate within the measurement resolution: based on the measurement noise, we obtained an upper limit of 2 kHz for this rate. On the other hand, by periodically

*ville.maisi@mikes.fi

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