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Observation of Andreev reflection in all-superconducting single-electron transistors

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We present the first experimental evidence for Andreev reflection in a single-electron transistor with both the leads and the island superconducting. In this process, two electrons tunnel to form a Cooper pair, or the reverse. It provides a second mechanism for two-electron transfer in addition to Cooper pair tunneling. Andreev reflection is found to contribute to several current cycles within the Coulomb blockade of single-electron tunneling. The observation of these cycles, however, requires that the charging energy $E_C<\Delta$.

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Charge transfer processes in single-electron transistors (SET's) have been the subject of much attention. In particular, within the Coulomb blockade of single-electron tunneling,[1,2] two-electron tunneling dominates and has been extensively examined. In devices with both superconducting leads and a superconducting island (SSS), the focus has been on combined Cooper pair and quasiparticle tunneling.[3,4] In SET's with superconducting islands and normal leads (NSN), experimental and theoretical investigations have concentrated on Andreev reflection (AR) at low bias voltages.[5–8] In this paper we report the first experimental evidence for Andreev reflection in *all-superconducting* SET's.

At a normal-superconductor (NS) interface, an electron from the normal metal can be Andreev reflected as a hole, forming a Cooper pair in the superconductor.[9,10] AR also occurs at the normal-insulator-superconductor (NIS) tunnel junctions of NSN SET's, providing the dominant low-bias current mechanism.[5–8] In superconductor-insulator-superconductor (SIS) junctions, such two-electron tunneling can produce current at $V=\Delta/e$ (for equal superconducting energy gaps Δ in the two electrodes), instead of the usual $V=2\Delta/e$ needed for single quasiparticle tunneling.[11,12] Multiple Andreev reflection can also occur in SIS structures, producing subharmonic gap structure at $V=2\Delta/ne$ for n an integer,[13,14] as well as enhanced shot noise.[15] As we show below, in all-superconducting SET's with the characteristic charging energy $E_C<\Delta$ (where $E_C=e^2/2C_\Sigma$ and $C_\Sigma=C_1+C_2+C_g$ is the total capacitance of the SET island), less energy is needed to transfer two electrons to the island via AR than to create two quasiparticles in single-electron tunneling. In this regime AR contributes to current features within the Coulomb blockade.

We consider here an SSS SET (see inset, Fig. 1) with the Josephson coupling $E_{Ji}=h\Delta/8e^2R_i\ll E_C$. In this limit, the number of excess electrons n on the island is well-defined, and the system is governed by the charging energy of the island $U(n)=(Q_0-ne)^2/2C_\Sigma$, where $Q_0=C_gV_g$ is the induced gate charge.[1,2] The equilibrium value of n will be that which minimizes the island's charging energy, i.e., the integer closest to Q_0/e. At low temperatures, sequential single-electron tunneling through such a device requires that the bias voltage provide enough energy to overcome both the superconducting energy gaps at the two SIS junctions and the change ΔU in the charging energy caused by changing the number of electrons on the island by ±1. There is thus a threshold voltage for single-electron tunneling, varying between $4\Delta/e$ and $4\Delta/e+2E_C/e$ depending on Q_0. Below this threshold voltage, single-electron transport is energetically forbidden and the system is in the Coulomb blockade of single-electron tunneling.

Within the blockade, thermally activated or higher-order processes are necessary for charge transport through the device. At elevated temperatures $T\lesssim T_c$, thermally excited quasiparticles can tunnel, producing current peaks when the singularities of the quasiparticle densities of states are aligned.[16–18] At lower temperatures, where thermal quasiparticles can be neglected, processes transferring two electrons are required. Inelastic cotunneling across the entire device through the energy barrier created by the island charging energy contributes to the current through SSS devices for voltages above $4\Delta/e$ but below the Coulomb blockade threshold.[19] At voltages below $4\Delta/e$, processes favoring the creation of Cooper pairs instead of quasiparticles dominate. In SSS SET's, subgap current mechanisms involving Cooper pair tunneling have been extensively studied,[3,4,20–23] while in NSN devices, AR has been shown to be the dominant current-producing process at low bias voltages.[5,6] We demonstrate here that AR also contributes to charge transport in all-superconducting SET's.

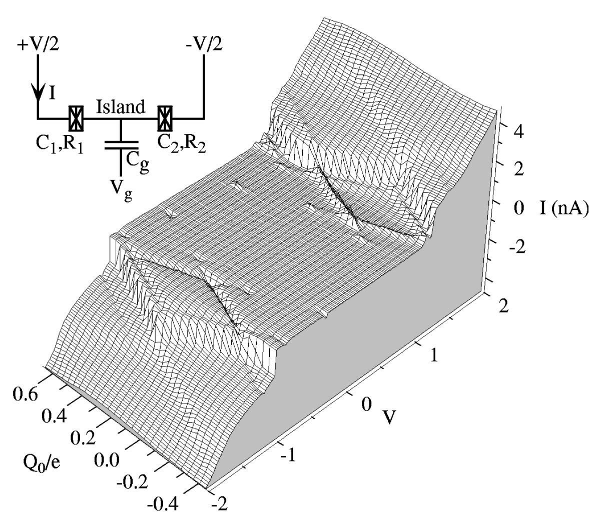

FIG. 1. Measured $I(V,Q_0)$ surface for a SSS SET. Inset: bias schematic for the device.

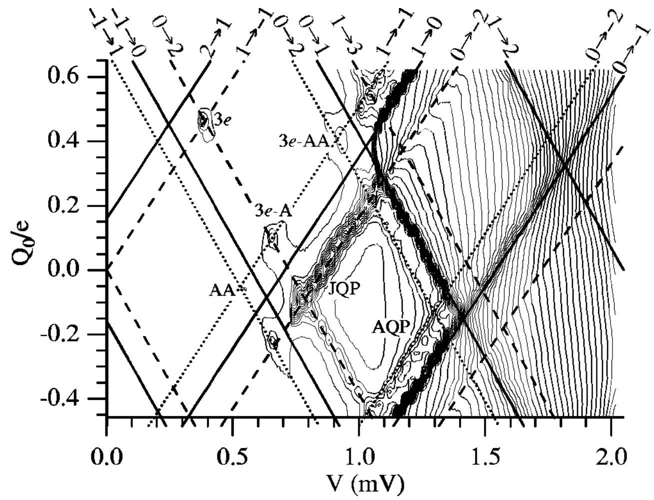

FIG. 2. Contour plot of the positive-bias region of Fig. 1. Solid lines are the *thresholds* for single-electron tunneling through junctions 1 (positive slope) and 2 (negative slope). Dotted lines are the *thresholds* for Andreev reflection. Dashed lines are where Cooper pair tunneling is *resonant*. The lines are labeled with the initial and final n values. The cycles producing the labeled features are listed in Table I, and the processes forming the cycles are illustrated in Fig. 3. The contour interval is 80 pA.

We fabricated our Al-AlO$_x$-Al SET samples with standard electron beam lithography and shadow evaporation techniques. Our measurements were made in a symmetric, four-point configuration in a dilution refrigerator at a mixing chamber temperature of $\approx$50 mK, although self-heating effects[24] elevated the electron temperature to $\approx$130 mK. Sample parameters could be inferred directly from the measured device response in the superconducting and normal states. For the sample discussed here, $\Delta=260$ μeV, $C_1=$ 191 aF, $C_2=158$ aF, $C_g=64$ aF, $R_1=114$ kΩ, $R_2=151$ kΩ, and $E_C=194$ μeV. Using the Ambegaokar-Baratoff relation,[25] we estimate $E_{J1}=7.4$ μeV and $E_{J2}=5.6$ μeV. The sample was thus in the regime where $E_{Ji} \ll E_C < \Delta$.

Figure 1 shows the current response of the device as a function of bias voltage and gate charge Q_0. We focus on the positive bias regime for the rest of our discussion, and assume $T=0$ so there are no thermally excited quasiparticles. The contour plot of the current response for positive bias voltages, shown in Fig. 2, accentuates the numerous current features found within the Coulomb blockade. The observed features are produced by cycles of tunneling processes through the two junctions. Each individual process—single quasiparticle tunneling, Cooper pair tunneling, and AR—has a characteristic dependence on the bias voltage and gate charge determined by the energetics of the transition. We can label each tunneling transition across a single junction by two parameters: the number m of electrons transferred and the number q of quasiparticles created. (We assume equal superconducting gaps for the island and both leads.) For $V>0$ and $T=0$, we need only consider transitions across junction 1 that change $n \to n-m$, and across junction 2, $n \to n+m$ (inset, Fig. 1). For a given transition, the bias voltage must supply sufficient energy to overcome the change ΔU in the charging energy and create the quasiparticles. This energy balance requirement for tunneling across junction i can be written as

$$m\kappa_i eV(Q_0)=U(n\mp m)-U(n)+q\Delta$$
$$=2mE_C\left[(-1)^{i-1}\left(\frac{Q_0}{e}-n\right)+\frac{m}{2}\right]+q\Delta\,, \tag{1}$$

where $\kappa_i=1-(C_i+C_g/2)/C_\Sigma$ is the fraction of the applied bias voltage appearing across junction i.

In the absence of thermally excited quasiparticles, single-electron tunneling across one of the SIS junctions in an SSS SET creates one quasiparticle on each side of the junction, each with minimum energy Δ. Equation (1) with $m=1$ and $q=2$ therefore gives the minimum or threshold voltage for this transition. The tunneling of Cooper pairs is a dissipationless process, significant only when the energies of the initial and final states are the same. The resonance conditions for this two-electron transition are given by Eq. (1) with $m=2$ and $q=0$. AR is characterized by the transfer of two electrons, accompanied by the formation of two quasiparticles. Either a Cooper pair tunnels, forming two quasiparticles on the other side of the junction, or two electrons tunnel, forming a Cooper pair on the other side of the junction and leaving behind two quasiparticle excitations. The AR thresholds are thus given by Eq. (1) with $m=2$ and $q=2$. Note that with AR, both quasiparticles are on the same side of the junction, unlike the case of single-electron tunneling.

These threshold and resonance conditions define families of lines for different n in the $V-Q_0$ plane. They are plotted in Fig. 2: solid lines for single-electron tunneling ($m=1$, $q=2$), dashed for Cooper pair tunneling ($m=2$, $q=0$), and dotted for AR ($m=2$, $q=2$). The lines are labeled with the initial and final n values. Transitions across junction 1 have a positive slope in the $V-Q_0$ plane; junction 2 transitions have a negative slope. The lines facilitate the identification of the processes constituting the cycles which produce the observed current features. We can divide the features into two groups. Cycles involving only the two-electron transfer processes of Cooper pair tunneling and AR link states with the same parity. The majority, however, involve island charge states of different parity and can be viewed as cycles between the charge states n and $n\pm1$.

Three processes, illustrated in Fig. 3, contribute to $n \to n \pm 1$ transitions. In addition to the tunneling of a single quasiparticle across one of the junctions, there is the $3e$ process.[4] In the limit $E_C \gg E_J$, this process can be viewed as a Cooper pair tunneling through one junction, followed by a quasiparticle tunneling through the other. Involving Cooper pair tunneling, the process only occurs in the vicinity of the Cooper pair resonances. This process, in which three electrons tunnel, changes the island charge by 1. For consistency in notation, we adopt the name J-e for this process. As discussed above, AR provides a second mechanism for transferring two electrons across a junction. We can thus define another process taking $n \to n \pm 1$ which we term A-e. This process is a combination of two individual transitions: AR across one junction is followed by a quasiparticle tunneling across the other.

From these three $n \to n \pm 1$ processes, six current-producing charging cycles can be formed, as summarized in Table I. Sequential single-electron tunneling through the two junctions occurs above the Coulomb blockade. The well-

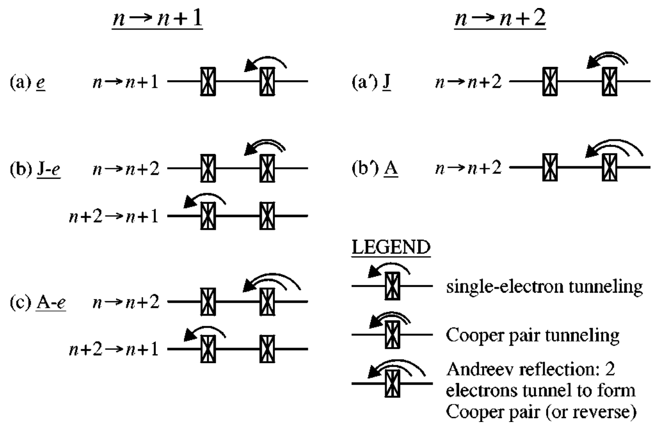

FIG. 3. The three processes changing the island charge by one, and the two parity conserving processes that change the island charge by two, illustrated for positive bias and increasing island charge. The current-producing cycles formed from these processes are listed in Table I.

studied JQP cycle consists of a J-e process combined with single-electron tunneling, i.e., Cooper pair tunneling through one junction followed by two quasiparticles tunneling through the other.[3] An alternative name for this cycle is therefore J-e-e, but we will adhere to the more established name JQP. (Although the term JQP has sometimes been used to describe any combination of Cooper pair and quasiparticle tunneling, we use the term here strictly for this particular cycle.) The JQP cycle is energetically favorable only if all of the three constituent transitions are. Thus the bias voltage for resonance Cooper pair tunneling must be above the threshold for the second particle transition, i.e., $V>(2\Delta+E_C)/e \approx 714\ \mu$V. [For $E_C<2\Delta/3$, the cycle also requires $V<(2\Delta+3E_C)/e$.[3,23]] Note that at $V=4E_C/e\approx 776\ \mu$V, the JQP cycle can proceed through resonant Cooper pair tunneling through either junction. Alternating J-e processes form the $3e$ peak at the intersection of the Cooper pair resonance curves at $V=2E_C/e\approx 388\ \mu$V. For this four-step cycle to be allowed, however, the Cooper pair resonances must occur at voltages above the thresholds for the two quasiparticle tunneling transitions, which requires $E_C>2\Delta/3$. These three large features, found along the single-electron threshold and Cooper pair resonance lines of Fig. 2, are all familiar from earlier investigations.[1,3,21–23] We focus here on the features lying along the dotted lines, originating in current-producing charging cycles involving Andreev reflection.

TABLE I. Named current features, the processes which compose the cycles producing them, and the bias conditions for which they occur (P = peak, T = threshold).

Feature	Cycle Components	Location
	$n\leftrightarrow n\pm 1$	
SET	e, e	T: $4\Delta\leq eV\leq 4\Delta+2E_C$
JQP	J-e, e	P: $2\Delta+E_C\leq eV$[a]
$3e$	J-e, J-e	P: $eV=2E_C$
AQP	A-e, e	T: $3\Delta+E_C\leq eV$
$3e$-A	J-e, A-e	P: $\Delta+2E_C\leq eV$
$3e$-AA	A-e, A-e	T: $2\Delta+2E_C\leq eV$
	$n\leftrightarrow n\pm 2$	
supercurrent	J, J	P: $eV\approx 0$
JA	J, A	P: $\Delta\leq eV$
AA	A, A	T: $2\Delta\leq eV$

[a] $2\Delta+E_C\leq eV\leq 2\Delta+3E_C$ for $E_C<2\Delta/3$ (Refs. 3 and 23).

The remaining three $n\rightarrow n\pm 1$ cycles all rely on AR through the A-e process. Together with single-electron tunneling, the A-e process forms the AQP cycle, which produces the current ridges seen just below the Coulomb blockade threshold in Fig. 2. This cycle is analogous to the JQP cycle: AR at one junction is followed by two quasiparticles tunneling through the other, and the cycle only becomes energetically favorable for voltages above both the AR threshold (dotted lines, Fig. 2) and the quasiparticle thresholds (solid lines), i.e., for $V\geq(3\Delta+E_C)/e\approx 974\ \mu$V. Alternating J-$e$ and A-e processes produce the peaks seen in Fig. 2 at the crossing of the Cooper pair resonances and AR thresholds at $V=(\Delta+2E_C)/e\approx 648\ \mu$V. Because of the similarity to the $3e$ cycle, we term this cycle $3e$-A. The features at the AR threshold crossings at $V=(2\Delta+2E_C)/e\approx 908\ \mu$V are caused by $3e$-AA cycles. In these cycles, *both* of the two-electron transfers occur through AR instead of Cooper pair tunneling. The $3e$-AA features would appear at half-odd integer values of Q_0/e for $C_1=C_2$.

The cycles involving fixed parity cannot include quasiparticle tunneling; they must instead rely solely on the two-electron transfer processes of Cooper pair tunneling and AR. At the Andreev threshold crossings near $V=2\Delta/e\approx 520\ \mu$V, we observe AA cycles formed from sequential AR through the two junctions. This cycle is similar to the sequential Cooper pair tunneling that produces supercurrent through SSS devices. If the junction capacitances were equal, the Andreev thresholds would intersect at integer values of Q_0/e. Though not seen in the data presented here, JA cycles, in which Cooper pair tunneling across one junction alternates with AR at the other, are also possible and are $2e$-periodic in Q_0.[26] The threshold for this process is at $V=\Delta/e$.

The major difference between AR in NSN and SSS SET's is that there is no minimum threshold in NSN systems, while in SET devices two quasiparticles of minimum energy Δ are involved. A single NIS junction has a finite Andreev conductance at $V=0$.[7,8] In contrast, for a single SIS junction at $T=0$ Andreev reflection requires $V>\Delta/e$. Consequently, whereas AR in NSN SETs can produce a finite conductance at zero bias if the Coulomb blockade of AR is tuned away by the gate charge,[5,6] in SSS devices Andreev processes only contribute to the current for $V>\Delta/e$.

This minimum voltage necessary for AR at SIS junctions is the sole difference between the Cooper pair resonance conditions and the Andreev thresholds. Any current cycle relying on Cooper pair tunneling for two-electron transport can also use AR, but at a voltage higher by Δ/e. Thus while the $3e$ peak is found at $V=2E_C/e$, the $3e$-A peak is at $V=(2E_C+\Delta)/e$ and the $3e$-AA peak is at $V=(2E_C+2\Delta)/e$. Similarly, the AA cycle has its threshold at $V=2\Delta/e$, compared to the JA cycle at $V=\Delta/e$ and the supercurrent at $V\approx 0$.

The spacings between the AQP ridges and the Coulomb blockade thresholds are intimately connected to the two energy scales Δ and E_C. Single-electron tunneling requires the creation of two quasiparticles, each with minimum energy Δ, per tunneling electron. In contrast, AR requires the creation of only one quasiparticle per tunneling electron, but increases the charging energy of the island. This difference in energies is reflected in the spacings between the AR and quasiparticle thresholds: for junction i they are separated in voltage by an amount δV_i given by $\kappa_i e \delta V_i = \Delta - E_C$. If $\Delta < E_C$, the quasiparticle tunneling thresholds are lower than the AR thresholds—less energy is needed to break a Cooper pair than to put an extra electron on the island. In this regime, AR processes will be difficult to observe against the large background of quasiparticle tunneling. Thus the AQP current ridge and the $3e$-A and $3e$-AA cycles should only be visible for $E_C < \Delta$. That a similar condition holds for the AA cycle is more subtle. Although there is no quasiparticle current cycle that is energetically favorable at the $2\Delta/e$ voltage threshold for the AA cycle, if $\Delta < E_C$ then single quasiparticle tunneling transitions are allowed which can interrupt or ''poison'' the AA cycle by taking the system out of the appropriate n states. For the $3e$-A cycle there is an additional constraint: for there to be a peak along the Cooper pair resonance curve, $E_C > 2\Delta/3$. Otherwise, the Cooper pair resonance will be below the threshold for the quasiparticle tunneling step of the cycle. (Note that this condition is the same as for a pronounced $3e$ peak.) The observation of the $3e$-A peaks therefore requires $2\Delta/3 < E_C < \Delta$.

In conclusion, we have observed empirical evidence for AR in all-superconducting SET's, in which two quasiparticles tunnel to form a Cooper pair (or the reverse). This two-electron transfer mechanism is identifiable by its unique $V-Q_0$ dependence, and it contributes to several current cycles within the Coulomb blockade of single-electron tunneling. The resulting current features should only be observable, however, for $E_C < \Delta$.

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