Single Cooper-Pair Tunneling in Small-Capacitance Junctions

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We present observations of charging effects for Cooper pairs in short linear arrays of small-capacitance Josephson junctions. Current-voltage characteristics show a Coulomb gap for Cooper-pair tunneling when the charging energy exceeds the Josephson coupling energy. In a double junction a zero-voltage current is observed that is modulated by a gate voltage applied to the metal island between the junctions. For longer arrays a crossover from Coulomb blockade of Copper-pair tunneling to a supercurrent is observed when the ratio of Josephson coupling to charging energy is increased.

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In a tunnel junction with superconducting electrodes, a tunneling matrix element of magnitude $E_J/2$ couples states differing in junction charge by a Cooper pair.[1] The Josephson coupling energy E_J is determined by the junction resistance R_n and the BCS gap Δ of the superconducting metal: $E_J = h\Delta/8e^2R_n$. For a junction of large capacitance, at zero bias voltage states differing by a large number of Copper pairs are nearly degenerate in energy. Therefore, there is a large uncertainty in the charge on the junction. The eigenstates of the junctions are described by the phase difference φ of the superconducting electrodes,[2] conjugate to the charge of the junction. However, when the junction capacitance is reduced to a value where the typical energy of charging by a single Cooper pair becomes of order E_J the degeneracy of charge states is lifted, even for states differing by only one Cooper pair. The junction state is then well described by the charge, and single Cooper-pair tunneling is an accurate concept to describe the conduction. Conventionally for normal-metal tunnel junctions, the charging energy is expressed in units $E_C = e^2/2C$. Recently, submicron fabrication techniques have progressed to a level where junctions can be fabricated that have $E_C \gtrsim E_J$. This opens the possibility to investigate the tunneling of individual Cooper pairs, and thus study the basic theory of Josephson junctions.

For normal-metal tunnel junctions, a description in terms of the junction charge is allowed provided that R_n is larger than about $\hbar/e^2$. In linear arrays of normal junctions with small capacitance the discreteness of charge transfer appears in several charging effects.[3,4] First, the current-voltage (I-V) characteristic shows a threshold voltage for conduction, the Coulomb gap, with a magnitude $(n-1)e/2C$ for an array of n junctions. Second, by capacitively applying a gate voltage V_g to the metal island (with capacitance C_g) between two junctions, the I-V curve can be changed. This change has a periodicity e in the "gate charge" C_gV_g induced on the island, reflecting the equivalence of island charges $(C_gV_g - ne)$ that differ by an integer times e. Most reported experiments on junctions with superconducting electrodes also only show single-electron effects, because of a very small ratio E_J/E_C.

Few experimental results have been published where the interaction of charging effects with Josephson coupling was notable (E_J of order E_C). Iansiti *et al.*[5] published experiments on small junctions which were interpreted with theory based on macroscopic quantum phenomena[6] for a small Josephson junction, i.e., a description in φ space. Fulton *et al.*[7] published experiments on a double superconducting junction and pointed out some aspects of charging effects for Cooper-pair tunneling to interpret their results. Their device is quite similar to ours. However, they did not report on the low-voltage region that we focus on in this Letter. Likharev and Zorin[8] and Averin and Likharev[4] have theoretically treated aspects of the double superconducting junction that are relevant for the present work. In this Letter we present current-voltage characteristics of small linear arrays of aluminum tunnel junctions. For low E_J, these show direct charging effects for Cooper pairs; for increasing E_J, a crossover to more classical behavior occurs.

Figure 1 shows I-V curves of a double Al-AlO$_x$-Al junction with $R_n = 58$ kΩ, and a capacitance derived from the Coulomb gap of about 1 fF ($E_J/E_C = 0.13$), in the normal and superconducting states. In both cases the I-V curves for two different gate voltages are shown. The junctions, with an area of 0.01 μm^2, were patterned by e-beam lithography and produced by shadow evaporation on an oxidized silicon substrate.[9] A junction is formed of two aluminum strips, of width 100 nm and thicknesses 20 and 40 nm, overlapping for about 100 nm. The two junctions are 1 μm apart. The sample was thermally anchored to the mixing chamber of a dilution refrigerator. The leads to the junctions were filtered by low-pass filters which were also thermally anchored to the mixing chamber. Normal-state measurements were performed in a magnetic field to suppress superconductivity. The inset of Fig. 1(a) shows the device and measurement layout. In the normal state [Fig. 1(a)] a Coulomb gap of about 70 μV is visible, which can be completely suppressed with the gate voltage.

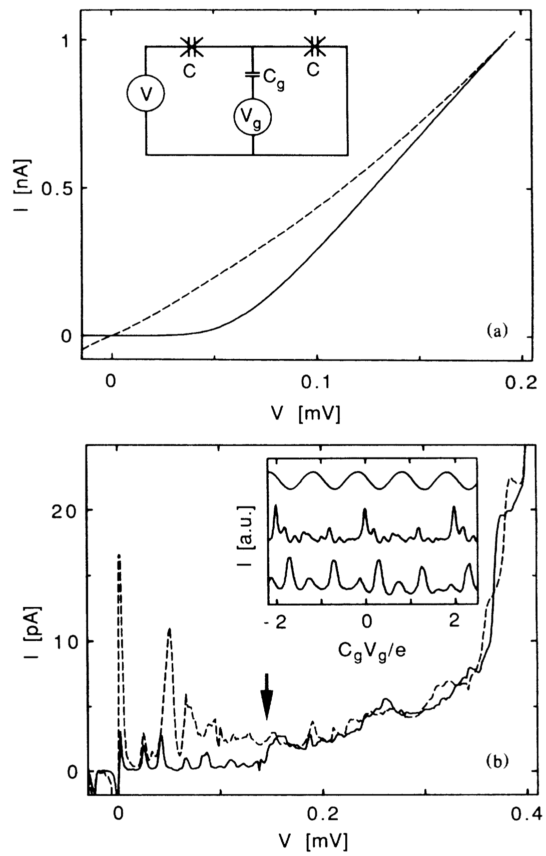

FIG. 1. *I-V* curves of a double junction with $R_n = 58$ kΩ ($E_J/E_C = 0.13$) for two different values of the gate voltage V_g at 10 mK. (a) In the normal state, realized by applying a magnetic field of 2 T. The Coulomb gap with a maximum value of about 70 μV (solid curve) can be suppressed with the gate voltage (dashed curve). Inset: Device and measurement layout. The junctions are denoted by crossed capacitor symbols, $C_g \approx 0.01C$. (b) In the superconducting state a Cooper-pair gap of about 150 μV arises (arrow). Coulomb gap and supercurrent are strongly dependent on gate voltage (compare solid and dashed curves). Inset: *I*-V_g curves for the normal state (top), the current peak at 20 μV (middle), and the supercurrent at $V = 0$ (bottom).

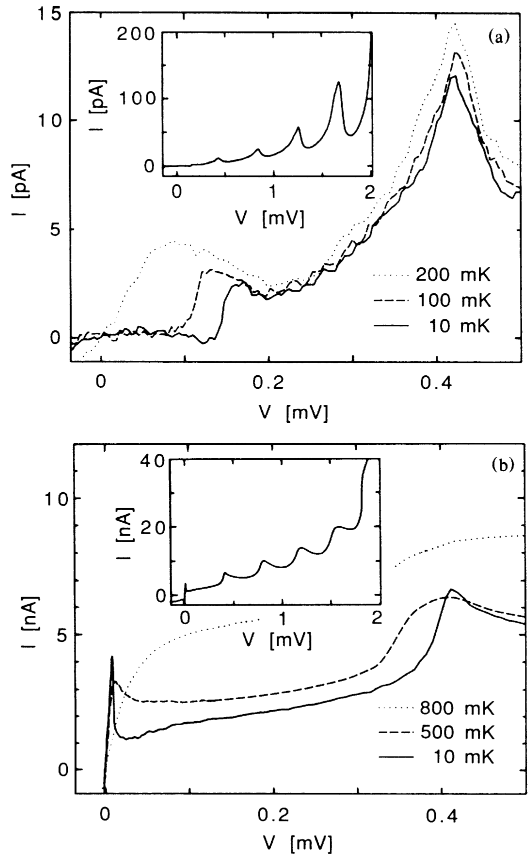

FIG. 2. *I-V* curves of linear arrays of five junctions. Insets: *I-V* curves at 10 mK on a larger scale. (a) $E_J/k_B \approx 0.13$ K and $E_C/k_B \approx 0.45$ K. A clear Cooper-pair gap arises. (b) $E_J/k_B \approx 1.4$ K and $E_C/k_B \approx 0.9$ K. At low temperature the resistance in the origin is zero (the finite slope in this figure is caused by the two-wire measurement method).

In the superconducting state [Fig. 1(b)] the curves show a current peak at zero voltage. In the following we will call this a supercurrent. We also see current peaks at multiples of about 20 μV, and for a voltage about equal to $2\Delta/e$ (0.4 mV for aluminum). Fulton *et al.* have previously considered the peak at $2\Delta/e$.[7] Here we want to emphasize two novel features. First, the current peaks in the first 150 μV, including the supercurrent, can be largely suppressed with the gate voltage, a clear indication of charging effects for Cooper pairs. Gate-voltage experiments will be described in more detail below. The second new feature in Fig. 1(b) is the voltage gap of about 150 μV (indicated by the arrow) that is visible if the supercurrent is suppressed with the gate voltage. The width of 150 μV is twice as large as the Coulomb gap in the normal state. This doubled width indicates Coulomb blockade of Cooper-pair tunneling as the origin of the gap. We will therefore call it a Cooper-pair gap. Figure 2(a) shows *I-V* curves for a linear array of five junctions, with $R_n = 60$ kΩ and $C \approx 2$ fF ($E_J/E_C = 0.3$). This device also exhibits a Cooper-pair gap, equal to about 2 times the normal-state Coulomb gap. With increasing temperature the gap first decreases in width and then changes into a supercurrentlike feature. Omission of the low-pass filters on the mixing chamber caused the high-temperature *I-V* curve to persist at the lowest temperature, thus hiding the Cooper-pair gap. On a larger scale (inset) four current peaks of increasing height are visible at voltages around multiples of $2\Delta/e$. In Fig. 2(b) we show the *I-V* curve of an array of five junctions with high E_J ($R_n = 5.5$ kΩ) and $C = 1$ fF ($E_J/E_C = 1.5$). Instead of a Cooper-pair gap, at low temperatures a supercurrent arises. On a large scale, we find again four current peaks at multiples of $2\Delta/e$, and negative differential resistance regions. These results were reproduced in the other samples that we examined. We have observed a Cooper-pair gap such as shown in Fig. 2(a) in arrays of five junctions with E_J/E_C up to 0.43. We have also examined other double junctions with small E_J. For these, as in Fig. 1(b), generally this gap was difficult to discern between the structure (resonances) in the *I-V* curve.

Three I-V_g curves for the double junction are given in the inset of Fig. 1(b). The height of the supercurrent is periodic in the gate voltage, with the same period e/C_g as in the normal state. The current just outside the Cooper-pair gap also oscillates with this single-electron period. It is important to note that if the voltage bias is increased, the curves invert. At a gate voltage where the supercurrent and the current just outside the Cooper-pair gap are at a maximum, the current near the first BCS gap and the current in the normal state (for arbitrary bias) are at a minimum. For the first two 20-μV resonances in Fig. 1(b) a doubled modulation period was observed, corresponding to $2e$ periodicity in the gate charge.

Since several of the concepts of single-electron tunneling in small junctions[4] are also useful to describe Cooper-pair tunneling,[7,8] we will first shortly discuss the extensively verified theory for normal-metal tunnel junctions. At zero temperature the single-electron tunneling rate is proportional to the change ΔE in the relevant (Gibbs) free energy, the sum of the capacitive energies and the work performed by the voltage sources. For a single voltage-biased junction $\Delta E = -eV$, and hence a Coulomb gap (or Cooper-pair gap) does not arise. In an array of junctions charge transfer occurs via intermediate states, where the charge resides on an electrode between the junctions. For low voltage, these states are higher in energy (by an amount of order E_C) than the initial state, so that tunneling is blocked and a Coulomb gap arises. With an externally applied gate voltage, the Coulomb gap can be suppressed. In two serially coupled junctions with island charge $e/2$, the energy change of a tunneling step is always smaller than zero for all finite voltages. Therefore, no threshold voltage for conduction is observed. For n serially coupled junctions, this complete suppression of the Coulomb gap is usually impossible due to random offset charging of the junctions,[4,9] e.g., by trapped charges near a junction barrier.

One essential difference between Cooper-pair tunneling (in the following abbreviated to CPT) and single-electron tunneling is the dependence of the tunnel rate on the energy change. Generally, Cooper pairs can only tunnel nondissipatively. Therefore, dc conduction by CPT can only be obtained if $\Delta E = 0$ for the tunneling event. If $\Delta E \neq 0$, an oscillating charge state is obtained, comparable to the ac supercurrent for a large capacitance junction under voltage bias. Coherent Cooper-pair tunneling across more than one junction (e.g., an array) can be usefully described as tunneling across an equivalent single junction with a smaller Josephson coupling.[8,10] Each intermediate tunneling step with energy change ΔE_i contributes to the decrease of the coupling by a factor of order $E_J/\Delta E_i$. In the present small-capacitance junctions, the intermediate states will typically differ by an amount of order E_C in energy. Obviously, the coherent transfer of Cooper pairs through an array of junctions is therefore strongly dependent on the ratio E_J/E_C and the number of junctions. Only for a gate charge e in a double junction at small voltage will the coupling between initial and final states be of order E_J.

We can now proceed to discuss the results for the double junction of Fig. 1 in the superconducting state. If a gate charge e is induced on the central electrode, at zero drive voltage the energy change for CPT is zero for either of the junctions. Therefore, a supercurrent develops as an equivalent of the complete suppression of the Coulomb gap in the normal state for a gate charge $e/2$. This is the situation of the dashed curve. One might expect that the height of the supercurrent is periodic in the gate voltage with period $2e/C_g$.[4] However, the observed periodicity is e/C_g because all states differing in gate charge by a multiple of e are equivalent due to the possibility of quasiparticle tunneling. This is true even if the number of quasiparticles is very small. States with gate charge equal to a multiple of e will relax by quasiparticle tunneling to the lower-energy state of island charge 0, which suppresses CPT. We propose that the supercurrent is at a maximum for all gate charges equal to an integer times e because occasionally a quasiparticle tunneling event produces the situation with island charge e, and thus catalyzes conduction by CPT. The supercurrent is limited by the duration of this situation, which only lasts until relaxation to the gate charge 0 occurs again. The probability of a tunneling event from island charge 0 to e is strongly dependent on temperature. Indeed, in our experiments the supercurrent was found to increase strongly for increasing temperature. At a gate charge e the Coulomb gap in the normal state is maximized, which explains the inversion of the current versus gate-voltage characteristics and in this way confirms conduction by CPT.

The other *I-V* curve of Fig. 1(b) (solid line) corresponds to the situation with a noninteger gate charge on the central island. Current by coherent CPT through both junctions is now smaller by a factor of about E_J/E_C. Inside the Cooper-pair gap, for voltages larger than the normal-state Coulomb gap, conduction also takes place by quasiparticle tunneling with a very small rate, proportional to the subgap conductance. At the Cooper-pair gap, Cooper pairs are mixed across one junction ($\Delta E = 0$) so that the quasiparticle tunneling events across that junction can be replaced by CPT. Therefore, CPT across this junction alternates with quasiparticle tunneling across the other, resulting in an increase of current. Fulton *et al.*[7] have shown that a similar process accounts for the current peak around the BCS gap voltage. They explained that for such a voltage CPT across one junction alternates with quasiparticle tunneling with a higher rate across the other. Because the energy gain of the quasiparticles is larger than 2Δ, the tunneling rate is in that case determined by the normal-state resistance.

For the interpretation of the current peaks at small nonzero voltages we use again the equivalence of a double junction to a single junction with coupling dependent on the gate charge. For a single junction under voltage bias, current resonances arise if the ac Josephson frequency $2eV/\hbar$ is in resonance with an environmental mode.[11] These resonances cause the current peaks in Fig. 1(b). Experiments have shown that the resonant modes are specific for our experimental circuit. They cause current peaks at the same voltages in the I-V curve of a single high-E_J junction. It is puzzling that in contrast to the situation for the supercurrent here a $2e$ periodicity in the gate charge is observed.

In Fig. 2(a) for the array of low-E_J/E_C junctions we observe a Cooper-pair gap as in Fig. 1(b). However, now Josephson coupling across the five junctions will generally be attenuated by a factor of order $(E_J/E_C)^4$. Because of random offset charging it is not possible to obtain a higher coupling using a gate charge. This is the reason for the absence of a supercurrent (similar to the impossibility in the normal state to suppress the Coulomb gap in this array completely) and for the absence of the 20-μV resonances. Again, at the Cooper-pair gap, the current increases because of the possibility of CPT alternating with quasiparticle tunneling. The current peaks at multiples of the BCS gap are, as for the double junction, probably a result of the combination of CPT with quasiparticle tunneling with a rate determined by the normal-state resistance. Finally, in the case of the array of Fig. 2(b), $E_J = 1.4E_C$. Therefore, all states with Cooper pairs on the central islands are mixed and there is strong coupling across the array. At zero voltage the Cooper pairs can transfer coherently through the complete chain.

In conclusion, we have observed features of localization of the charge on Josephson junctions due to small capacitance. The charge transfer unit is $2e$, but quasiparticles also play a role. For $E_J > E_C$, coherent mixing of the Cooper-pair states can occur despite the still appreciable charging energy, resulting in a supercurrent. In a double junction coherent Cooper-pair tunneling is modulated by charging of the central island through a gate voltage.

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