

Single Cooper pair pump

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Abstract

We have operated a Cooper pair pump, a linear array of superconducting tunnel junctions in which single Cooper pairs are moved under the influence of ac signals applied to two gate electrodes. The pump is based on Coulomb blockade of charge tunneling. Because of the small junction capacitance precisely one Cooper pair is transferred per ac cycle. The current-voltage characteristics of this device show current plateaus close to $2ef$, where f is the frequency of the ac voltages. Deviations are explained in terms of Zener tunneling, Cooper pair co-tunneling and sporadic quasiparticle tunneling.

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Introduction

Several experiments have demonstrated that it is possible to exploit charging effects to move single electrons one by one through a linear array of small metal tunnel junctions [1,2]. In those experiments the tunneling of single electrons was controlled by the application of ac signals so that one electron was passed through the device per drive cycle. This leads to a current to frequency relation $I = ef$. Current control at a level of single charge carriers is achieved by the Coulomb blockade effect.

For example the single electron pump as described by Pothier et al. [2] transferred single electrons per cycle of two phase-shifted ac voltages. In our experiment we operate a similar device both in the superconducting state and in the normal state to investigate whether such control is possible for the superconducting charge carriers, Cooper pairs. The pump has been made of aluminum tunnel junctions which have superconducting electrodes at the experimental temperature, but can be brought to the normal state by applying a magnetic field. An experiment like this can show whether a Cooper pair behaves as a real localized charge carrier and is not merely a useful theoretical construction.

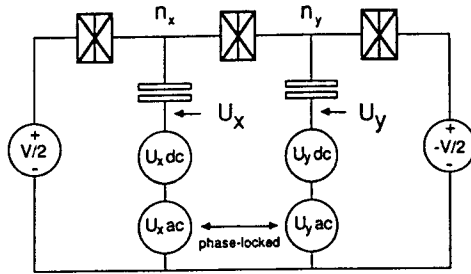
In this paper we focus on the new effects that were observed in the charge pump in the superconducting state. For high frequency driving voltages we observe a current which is significantly lower than $2ef$. The deviation is in good agreement with the expected rate of Zener tunneling of the system when it is swept too fast through the bias conditions at which a Cooper pair can tunnel. The operation of the device is affected by quasiparticle tunneling, although this appears less important than expected. A further deviation from the simple behavior is due to coincident tunneling of Cooper pairs through two junctions.

Operating principle

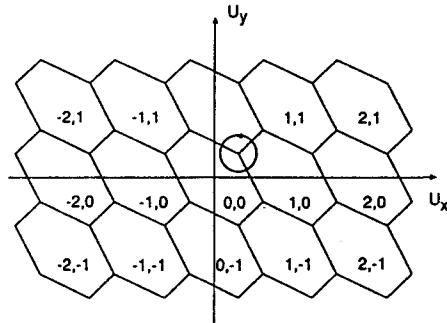
The control of charge transfer at the level of single carriers is based on Coulomb blockade of tunneling [3]. If tunneling of a charge carrier would increase the electrostatic energy of a circuit of tunnel junctions, it will not occur at sufficiently low temperature. For tunnel junction capacitances, C , below about 1 fF the change in energy during tunneling - of the order of $E_C = e^2/2C$ - is significantly larger than thermal energies available in a dilution refrigerator.

The charge pump [2] is shown schematically in Fig. 1a. It is a linear array of three tunnel junctions. The two superconducting islands between the three junctions are capacitively coupled to gate electrodes [4]. Because of the isolation by tunnel barriers, the charge on these islands will be quantized. If for given values of the bias voltage, V , and the two gate voltages, U_x and U_y , the system relaxes to the lowest possible energy state by one or more tunneling events, we can induce an arbitrary number of excess charge carriers on either of the islands. So, by adjusting the voltages one can cause a single electron or Cooper pair to tunnel through any of the three junctions.

a



b



c

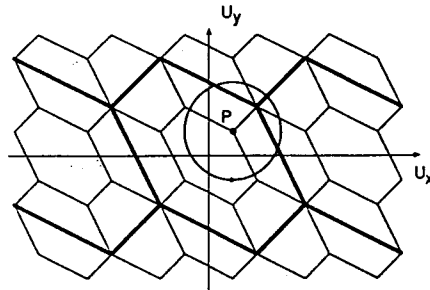


FIGURE 1. a. Schematic picture of the single Cooper pair pump. The crossed boxes denote the tunnel junctions. Two alternating gate voltages will move charges (quasi-)adiabatically one by one through the tunnel junctions. **b.** The lowest energy charge configuration as a function of the gate voltages U_x and U_y . Following the circle one electron is transferred through the device. **c.** The lowest energy charge configuration in the superconducting state. The thick lines give the cell boundaries in the superconducting state. The thin lines give the cell boundaries where a quasiparticle can tunnel.

One possible cycle which pumps an electron through the device starts by raising the voltage on the left gate. When the voltage across the leftmost junction exceeds a threshold voltage [5] one electron can tunnel from the lead onto the left island. The probability for tunneling is proportional to the voltage in excess of this threshold. If one then decreases the voltage on the left gate while increasing the voltage on the right gate, an electron will tunnel to the right island when there is sufficient voltage drop across the middle junction. Finally the voltage on the right gate is decreased and an electron tunnels through the rightmost junction into the lead on the right side. If the amplitudes of the alternating gate voltages are within the proper range, the Coulomb blockade insures that only one electron is transferred.

In the superconducting state, in principle one Cooper pair can be transferred in a similar cycle, but a tunneling event is fundamentally different. Since Cooper pairs are condensed in a common ground state, a Cooper pair cannot increase its kinetic energy like an electron can. Therefore a tunneling event between two electrodes is possible only if the resulting change in electrostatic energy is zero (or alternatively is sufficient to create an excitation) [6]. If the gate voltages are changed *slowly*, a Cooper pair will tunnel precisely as the junction threshold voltage is passed, keeping the system adiabatically in the lowest energy charge configuration. If the bias conditions of the system are swept too fast compared to the coupling constant for Cooper pair tunneling, Zener tunneling can occur [7,8]. In our situation this means that no Cooper pair is transferred and the system is excited to a higher energy charge configuration.

When ac voltages of appropriate frequency are applied to the gates, the above described cycle is repeated continuously and charge carriers are pumped one by one through the device. The result is a current proportional to the frequency and the charge unit; $I=ef$ for electrons and $I=2ef$ for Cooper pairs. This can be obtained within a certain bias voltage range around zero. The voltage at each gate should have the same frequency but be $\pi/2$ out of phase. Changing the phase difference by π reverses the direction of the current. To obtain a measurable current, of order pA's, the frequency must be in the MHz range.

The operation of the device can be represented as a cycle in U_x - U_y space. For fixed bias voltage, the total energy of the device is a function of the gate voltages and the charge configuration, i.e. the excess charge on the two islands. Fig. 1b gives for zero bias voltage the U_x - U_y plane divided into cells with the lowest energy charge configuration in the normal state. The charge configuration is denoted by (n_x, n_y) , the number of excess charges in unit e . Each hexagonal cell in this honeycomb pattern represents the gate voltage range in which a certain charge configuration minimizes the electrostatic energy. Neighboring cells have a charge configuration that differs by the tunneling of a single electron. At the boundaries between cells the energies of two charge configurations are equal.

To transfer an electron through the pump, the elliptic trajectory created by the alternating gate voltages should lead through three cells which yield the correct succession of charge states. This means that the trajectory should encircle one of the triple points in Fig. 1b (trajectories

encircling more triple points can also work). An example is the circle drawn in the figure. One cycle in the counter-clockwise direction starting in (0,0) forces the device through the charge configurations (1,0), (0,1) and back to (0,0) with the result that one electron is transferred through the device. Encircling the triple point in the clockwise direction will transfer one electron in opposite direction. For the same circling direction two nearest-neighbor triple points pump current in opposite direction. A trajectory around a triple point is realized by applying to the gates a superposition of dc voltages corresponding to a triple point and two phase-shifted ac voltages to encircle the triple point. For non-zero bias voltage V the cell boundaries are translated, causing the triple points to deform into triangles inside which no charge configuration is stable (Fig. 6b). When the gate voltage bias is inside these triangles, a dc current flows through the device even without rf modulation. However, controlled pumping is still realized if the modulation ellipse lies completely outside this triangle.

In the superconducting state the unit of charge which tunnels through a junction is $2e$, neglecting possible quasiparticle excitations. This means that the lowest energy charge configuration as a function of gate voltages again yields a honeycomb pattern, that is twice as large as in the normal state. It is drawn in Fig. 1c in thick lines, while the quasiparticle energy diagram is given in thin lines (similar to Fig. 1b). As in the normal state, the transfer of a charge carrier, in this case a Cooper pair, is performed by following a trajectory around a triple point. An example is the indicated circle.

Since the electron tunneling rate increases proportionally to the excess voltage over a junction, the tunneling rate is proportional to the distance from a cell boundary (in a direction in which the charge configuration is unstable). The tunneling electron will arrive above the Fermi level and subsequently relax. In contrast, a Cooper pair will tunnel on the crossing of a cell boundary.

For proper operation in the superconducting state it is essential that the probability of quasiparticle tunneling is very low on the time scale of one rf cycle. The presence of quasiparticles gives rise to the possibility that the island charge changes with e instead of $2e$. As can be verified in Fig. 1c the trajectory for the transfer of a Cooper pair leads into regions where the system is unstable against quasiparticle tunneling. In theory the resistance for quasiparticles R_{qp} should increase exponentially with decreasing temperature, approximately as [9, 10]:

$$R_{qp} = R_n \exp \frac{\Delta(0)(1/T - 1/T_c)}{k_B} \quad (1)$$

where $\Delta(0)$ is the BCS gap at $T=0$, T_c is the critical temperature of the superconducting electrodes, and R_n is the normal state resistance of the junction. The tunneling rate is of the order of

$$\Gamma_{qp} \approx (R_{qp} C)^{-1} \quad (2)$$

At the experimental temperatures (below 50 mK) according to eq. (1) R_{qp} should be so high that one would not expect quasiparticle tunneling to occur on the experimental time scales. However, other experiments [11, 12] have shown that even at the lowest temperature quasiparticle tunneling events still occur at a noticeable rate. The reason for the existence of quasiparticle excitations down to very low temperature is not yet clear to us.

Experiments

The fabrication technique for the present devices is described in ref. 10. They are patterned with e-beam lithography in a 3-layer resist. The junctions are fabricated by oblique angle evaporation. The layout is close to the schematic of Fig. 1a. In the pump for which results are presented here, the guarding of the gate electrodes was not very effective. Consequently, in addition to the gate capacitance drawn in Fig. 1a, there is a significant cross-capacitance (about 80-90%) from the gate to the other island. The junctions have an area of approximately $(60 \text{ nm})^2$. The junction capacitance is 0.35 fF, derived from the Coulomb gap voltage offset in the current voltage (I-V) characteristic. The homogeneity of the junction parameters is probably very good.

The normal state resistance of the junctions is 85 k Ω , yielding a Josephson coupling energy E_J of about 90 mK. E_J is the magnitude of the tunneling matrix element for the transfer of one Cooper pair [13]. It is usually determined from the critical current of the junction. Since the critical current in this array is suppressed by the charging effects, the quoted value is calculated with the relation $E_J = \hbar \Delta / 8e^2 R_n$. This is justified by results on larger aluminum junctions [10]. The BCS gap Δ of our aluminum films is 0.20 meV. The E_J/E_C ratio of the junctions is 0.03. For successful control of single Cooper pair tunneling with charging effects E_J should be much smaller than E_C [11]. This is clearly the case for our device. Also, one likes to have $E_J > k_B T$ in the experiment to minimize the influence of thermal fluctuations on Cooper pair tunneling. Measurements were done in a dilution refrigerator below 50 mK. Low-pass filters, thermally anchored to the mixing chamber, were used on all electrical leads. The gate voltages were in addition attenuated at 1.4 K and at the mixing chamber. All measurements were performed in a two-wire configuration, with active current measurement. The measurement electronics were optically decoupled from the rest of the equipment in the laboratory.

The procedure of operating the pump in the normal state is described in more detail elsewhere [2]. The triple points in gate voltage space (Fig. 1b) are found by applying a very small bias voltage and sweeping the dc gate voltages. As discussed above, the bias voltage will open the triple points into small triangles. Without rf modulation, dc current is only transferred inside the triangles, so that current peaks will indicate the position of the triple points. No current will flow far away from the triple points as long as the device is biased at a voltage low compared to e/C . Alternatively, an rf gate voltage modulation of small amplitude can be applied while

sweeping the dc gate voltage. In that case, positive or negative current peaks (at $I = \pm ef$) will indicate the position of the triple points. Ideally the pattern of Fig. 1b is reproduced. When cross-capacitance is high, this pattern is strongly deformed. However, it is still a simple procedure to bias on the triple points.

When the triple points have been found, we analyze the operation in terms of I-V characteristics as a function of rf parameters. The phase difference between the two rf gate voltages needs to be adjusted to obtain pumping in either direction. The required phase difference is determined experimentally. This is necessary since the phase relation outside the cryostat may differ from the phase relation at the device. Also, due to the cross capacitance the phase difference for an approximately circle-like modulation will be close to π instead of $\pi/2$. In Fig. 2 we show one period of the current versus phase difference in normal and superconducting state. In both cases two clear peaks are present, one positive and one negative. These are the peaks where plateaus are found in the I-V characteristics.

I-V characteristics in the normal state are presented in Fig. 3 for frequencies of 10 and 15 MHz. Plateaus at positive and negative current are found for the appropriate choice of the phase difference. The dashed lines are at the expected current values $\pm ef$. The rf amplitude was adjusted for maximum flatness of the current plateau. The plateaus are more rounded than e.g. for the turnstile of Ref. 1. This is at least partly due to electron co-tunneling (quantum leakage)[14]. Because of the intended operation in the superconducting state, the junction resistance was chosen lower (closer to the quantum resistance h/e^2) than is usual or desirable for single-electronic devices. We will return to the effects of co-tunneling later.

After removing the magnetic field to bring the aluminum to the superconducting state, and increasing the rf amplitude, again current plateaus were found. The procedure to find the triple points as it was used in the normal state did not work in the superconducting state. No current peaks were found at all on sweeping the gate voltages. We assert that this is due to quasiparticle tunneling. On time scales associated with dc voltage sweep rate, the superconducting triple points cannot be biased at because they are not stable against quasiparticle tunneling. Therefore in the experiment the gate voltages were kept fixed at the normal state triple point and only the rf amplitude was increased to get a modulation trajectory like the one given in Fig. 1c.

I-V characteristics in the superconducting state are given in Fig. 4 for frequencies between 2 and 20 MHz. Just as in the normal state, positive and negative current plateaus are obtained by appropriate choice of the phase difference between the two rf gate voltages. In contrast to the normal case in which the plateau goes smoothly through the I-axis, in the superconducting case we observe a step in the plateau at $V=0$. The dotted lines in Fig. 4 indicate the expected values $I=\pm 2ef$ (for 2 to 10 MHz). For the lower frequencies the higher of the two levels in a plateau (in absolute value) is close to $2ef$. At high frequencies even the highest plateau is significantly lower. For a frequency of 10 MHz, Fig. 5 gives the dependence of the I-V characteristics on the amplitude. The current at small bias voltage does not vary much, whereas wide flat plateaus are only obtained within a narrow amplitude range.

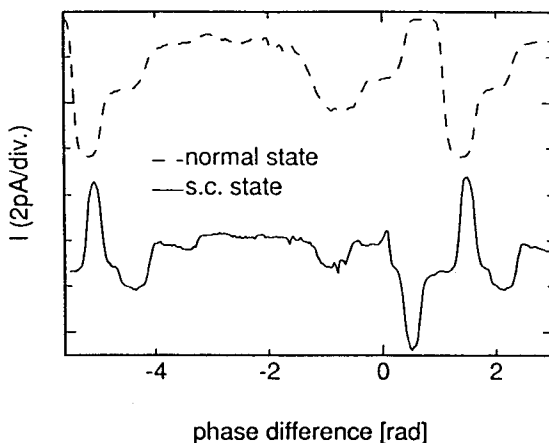


FIGURE 2. The current versus phase difference between the two gate voltages, in normal and superconducting (s.c.) state for an ac frequency of 20 MHz. ($V=0$)

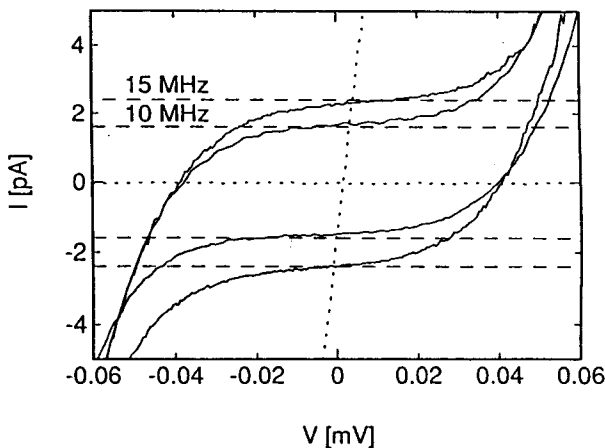


FIGURE 3. Current-voltage characteristics at 10 and 15 MHz in the normal state. The dotted curve is the I-V curve without r_f applied, the horizontal one with the gate voltage adjusted in the middle of a cell (maximal Coulomb gap) and the other with the gates adjusted at a triple point (minimal Coulomb gap). The positive and negative plateaus are obtained by choosing the appropriate phase difference. The dashed lines are at the expected values of $I = e f$. $T < 50$ mK

These are the basic results of the operation of the charge pump in the superconducting state. They indicate that this device indeed pumps single Cooper pairs per rf cycle. In the next section we will consider the most prominent features of the results. We will argue that the step in the current plateau is basically the same effect as the rounding of the plateaus in the normal state due to quantum leakage. We will also show that the deviation of the current level from $2ef$ is in agreement with the theory of Zener tunneling. Finally we will discuss the effects of quasiparticle tunneling in the superconducting state and explain why no $2e$ -periodic behavior is found in dc measurements.

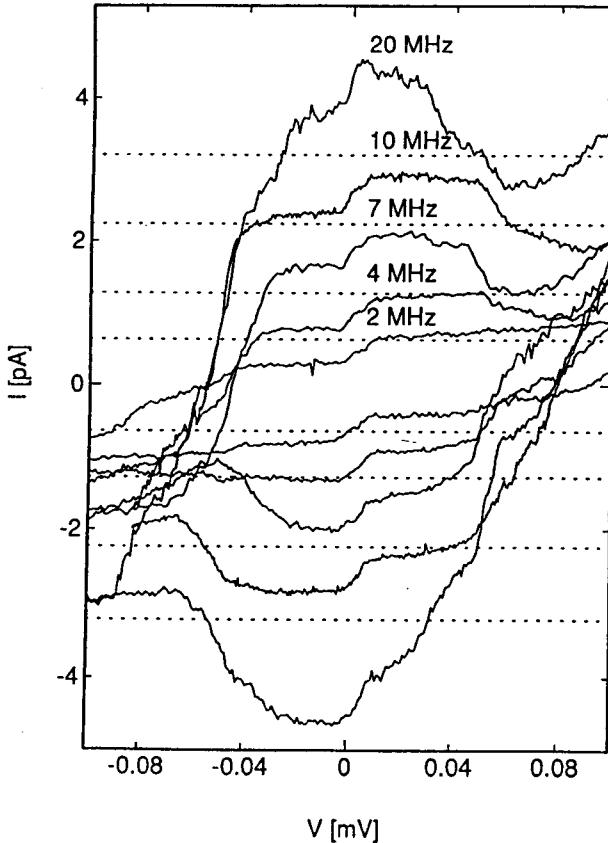


FIGURE 4. I-V characteristics for frequencies between 2 and 20 MHz in the superconducting state. The dashed lines are at the expected values $I = \pm 2ef$. $T < 50$ mK

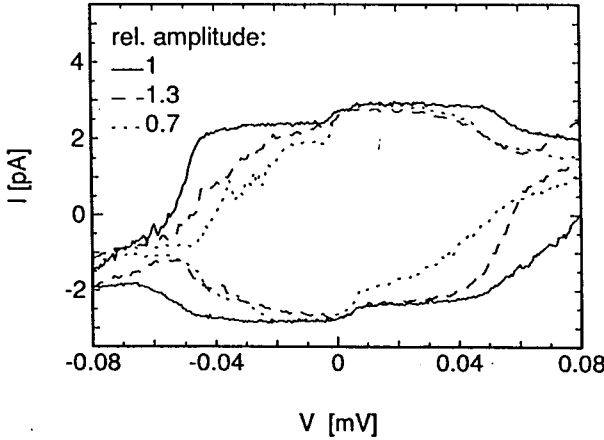


FIGURE 5. I-V characteristics at $f = 10$ MHz for three rf amplitudes, given relative to the value that gives maximal flatness of the plateau. This flatness depends strongly on amplitude.

Discussion of the experimental results

a. co-tunneling

The step at zero bias voltage in the current plateaus separates a level which is (at low frequency) close to $2ef$ from a level which is lower. On the lower level the sign of the current is opposite to the bias voltage, i.e. the modulation tries to pump Cooper pairs against a driving voltage. The character of the feature (two relatively flat levels, with a rather abrupt change) suggests a relation with a topological change. Indeed, the topology of the U_x - U_y stability diagram changes at $V=0$ if we take into account not only single Cooper pair tunneling events but also co-tunneling of Cooper pairs through two junctions in one event [15].

The change of charge configuration by Cooper pair tunneling through a given junction, can alternatively occur by two Cooper pair tunneling events in *opposite direction* in the two other junctions. This process is similar to co-tunneling in the normal state [14]. In the normal state the rate of co-tunneling increases continuously with bias voltage, causing a gradual rounding of the I-V curve. The different nature of Cooper pair tunneling is visible from the fact that co-tunneling results in a step. Like single Cooper pair tunneling, these co-tunneling events require a resonance of initial and final state. Thus the bias conditions can be represented by lines in the U_x - U_y plane, which are parallel to the lines for the single tunneling events. At $V=0$ the resonance conditions coincide. For finite bias voltage the resonance lines are separated. Fig. 6 schematically shows this change. In Fig. 6b the thick lines represent the single Cooper pair

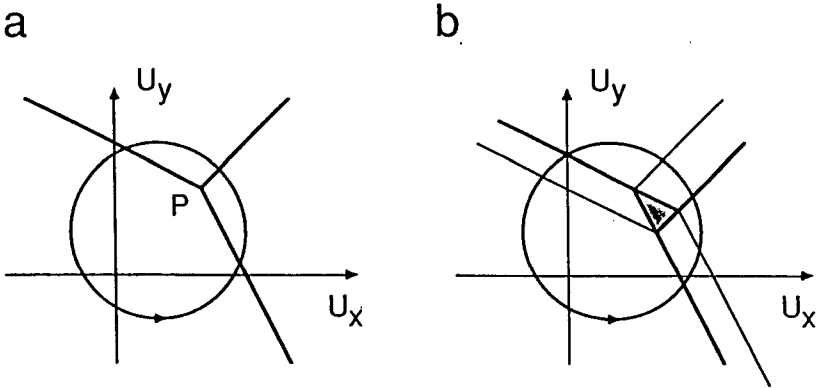


FIGURE 6. **a.** One triple point of Fig. 1c and the circle which corresponds to the transfer of one Cooper pair. The bias voltage is zero. **b.** If there is a small, positive bias voltage, the lines where a tunneling event occurs move and they meet in triangles instead of triple points. The indicated circle still results in the transfer of a Cooper pair. The thick lines represent the resonance condition for a single Cooper pair tunneling event, the thin lines for a co-tunneling event.

tunneling events and the thin lines co-tunneling. The distance between the lines is proportional to eV , in particular, the lines change place when the voltage changes sign. Hence, for a given circling direction the sign of the bias voltage determines which event can occur first. The co-tunneling resonance condition is met first if the circling direction in combination with single tunneling would yield a current opposite to the driving voltage.

A cycle with only co-tunneling events yields a current $4ef$ in a direction opposite to the current based on the single tunneling events. In the experiment, the drop to the lower plateau is much smaller than $6ef$. Thus, if our interpretation is correct, the co-tunneling events would need to have a reduced probability. The coupling for co-tunneling is indeed very small; because of the intermediate state with energy $\approx E_C$ it is reduced to about $E_J(E_J/E_C)$. In fact, simple Zener tunneling theory would predict that at the experimental modulation frequencies the probability of missing the double jumps is virtually 1. However, Cooper pair co-tunneling probably occurs as an inelastic transition induced by the coupling to the electromagnetic environment [16]. For the experimental parameters these are most likely thermally induced transitions. In the two devices examined so far, for the one with the smallest E_J (the smallest gap) there was a significantly larger current step in the plateau, even reversing the sign of the current. In this sample thermal equilibrium (equal occupation of the two possible charge states) is probably reached faster.

From Fig. 4 we observe that the size of the step is roughly independent of frequency. Qualitatively this is consistent with a transition rate which is significant only in a limited window around resonance. The time spent in this window and thus the relative magnitude of the effect decreases as $1/f$.

A qualitative analysis of the effects of co-tunneling is complicated by the fact that the relevant characteristics of the electromagnetic environment was not very well known in the experiments. In addition, co-tunneling is probably difficult to study separately from other disturbing effects, like the ones we will discuss in the following sections.

b. Zener tunneling

Around the resonance conditions for Cooper pair tunneling, the energy eigenstates of our array are perturbed from the electrostatic value by the Josephson coupling. When driving the system through these regions, a Zener transition can occur with a probability dependent on the drive speed. In the event of a Zener transition a Cooper pair tunneling is missed, and unless an inelastic transition occurs later in the cycle, the cycle will not contribute to the current. In Fig. 7 we show the experimentally determined level of the current plateau as a function of the rf frequency. Here we have determined the plateau current at low bias voltage, of the same sign as the current. In this way we might expect to remove as much as possible the effects of co-tunneling and quasiparticle tunneling. The dashed line in Fig. 7 follows $I=2ef$. At higher modulation frequencies the measured current deviates from this line.

The probability of Zener tunneling of an undamped system with coupling E_J driven by a rate of change $\dot{E}$ of the unperturbed (electrostatic) energy is approximately [8]:

$$P_Z = \exp \frac{-E_J^2 \pi}{4\hbar \dot{E}} \quad (3)$$

In the charge pump $\dot{E}$ depends on the rf frequency and amplitude. Generally $\dot{E}$ will be different for the three transitions in a cycle. Since it appears exponentially in eq. 3, one transition will likely dominate the total effect of Zener tunneling. This should then yield a current:

$$I = 2ef(1 - P_Z) \quad (4)$$

where P_Z is the Zener tunneling probability for the dominant crossing.

Since we do not know the attenuation, $\dot{E}$ is not known, but we know that $\dot{E} \propto f$. In Fig. 7 we have fitted eqs. 3,4 to the measurement by varying $\dot{E}/f$. The fitted value of $\dot{E}/f$ is equal to the actual value for the middle transition $((0,2) \leftrightarrow (2,0))$ in Fig. 1c, and twice as large as for the

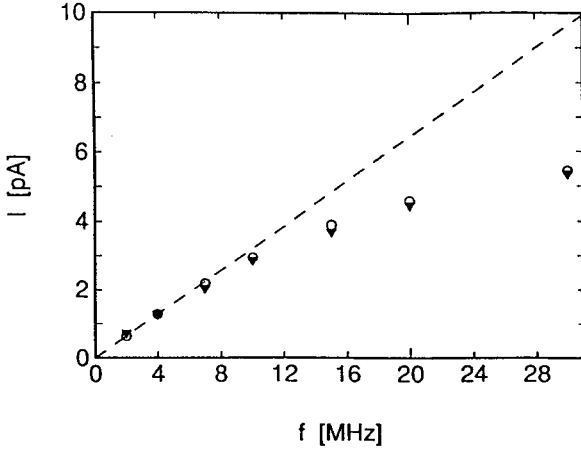


FIGURE 7. Current of the highest plateau versus frequency (triangles) compared to the expected value of $I=2ef$ (dashed line). The deviation at higher frequency is probably due to Zener tunneling. The open circles denote the calculated current for Zener tunneling according to eqs. (3-4) (see text). $\dot{E}/f$ is fitted for best agreement with experiment.

other two transitions. The circle in Fig. 1c is probably a reasonable approximation of the actual modulation in the experiment.

c. quasiparticle tunneling

Quasiparticle tunneling might be expected to disturb the operation of the charge pump. During part of the gate voltage modulation cycle, the charge state of the system is unstable against quasi-particle tunneling. The impossibility to find $2e$ -periodic behavior in dc measurements (e.g. when trying to find the $2e$ honeycomb pattern, but also in other experiments [11,12]) shows that these transitions indeed occur on experimental time scales.

If the gate voltage bias is not too far from the lowest energy configuration, the quasiparticle tunneling rate is of the order of eq.(2) which is extremely low. However, after a Zener transition, a quasiparticle tunneling event may decrease the electrostatic energy by an amount in excess of 2Δ . In that case the tunneling rate suddenly increases to a value of the order of $(R_n C)^{-1} \approx 20$ GHz [6]. We estimate that Zener transitions are a more important source of quasiparticle tunneling than a finite subgap resistance, for all but the lowest frequencies at which we have measured.

Quasiparticle tunneling events cause translations of the effective modulation ellipse over a distance e in the U_x - U_y plane. As a result the gate voltage modulation will occasionally jump and encircle other triple points. Some of those triple points will pump current in the opposite

("wrong") direction. It is not simple to estimate the rates and effects of quasiparticle tunneling. They will strongly depend on the precise bias parameters. At least the presence of quasiparticles is consistent with the experiments, where the plateaus are only obtained after quite careful tuning of rf phase difference and amplitude. Since we observe a pumped current which is close to $2ef$, and the deviation seems to be explained by Zener tunneling, apparently it is possible to reduce the effect of quasiparticle jumps considerably.

In order to evaluate the effects of quasiparticle tunneling, computer simulations could be very helpful. However, for direct comparison with experiment it would be necessary to have a much better knowledge of the experimental parameters than is presently the case.

Conclusions

We conclude that we have observed experimental evidence for the controlled transfer of Cooper pairs in a charge pump device based on Coulomb blockade. This is shown by current plateaus in the I-V characteristic close to $I = 2ef$. The Cooper pair pumping is disturbed by several factors of which we have identified Zener tunneling, Cooper pair co-tunneling, coupling to the electromagnetic environment at finite temperature and quasiparticle transitions.

This system or similar devices are very promising for further study of these effects.

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