## **Coulomb Blockade and Coherent Single-Cooper-Pair Tunneling in Single Josephson Junctions**

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We have measured the current-voltage characteristics of small-capacitance single Josephson junctions at low temperatures ( $T \le 0.04$  K), where the strength of the coupling between the single junction and the electromagnetic environment was controlled with one-dimensional arrays of dc SQUIDs. We have clearly observed Coulomb blockade of Cooper-pair tunneling and even a region of negative differential resistance, when the zero-bias resistance of the SQUID arrays is much higher than the quantum resistance  $h/e^2 \approx 26 \text{ k}\Omega$ . The negative differential resistance is evidence of coherent single-Cooper-pair tunneling in the single Josephson junction.

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Small-capacitance superconducting tunnel junctions provide a novel system for studying the interplay between the Josephson phase and the charge on the junction electrode. These are quantum mechanically conjugate variables, and their behavior is influenced by dissipation, such as discrete tunneling of quasiparticles, coupling to an electromagnetic environment, etc. The simplest and the most fundamental example is the single junction. Currentvoltage (*I*-*V*) characteristics of single junctions have been the subject of extensive theoretical investigations [1,2]. Experimentally, however, the observation of charging effects such as "Coulomb blockade" has been considered to be extremely difficult in single junctions because a high-impedance environment is necessary, and special care should be taken with the measurement leads [1]. For this reason, thin-film resistors [3] and tunnel-junction arrays [4,5] were employed for the leads, and an increase of differential resistance around  $V = 0$  was reported.

We use one-dimensional arrays of dc superconducting quantum interference devices (SQUIDs) for the leads. The advantage of this SQUID configuration is that the effective impedance of array can be varied *in situ* by applying an external magnetic field perpendicular to the substrate. Thus, the zero-bias resistance of the SQUID arrays at low temperatures can be varied over several orders of magnitude. This phenomenon has been extensively studied in terms of the superconductor-insulator transition [6]. The single junction in our samples, on the other hand, does not have a SQUID configuration, and therefore its parameters are practically independent of the external magnetic field. This enables us to study the *same* single junction in different environments. This type of experiment has not yet been reported to the best of our knowledge. We show that the *I*-*V* curve of the single junction is indeed sensitive to the state of the environment. Furthermore, we can induce a transition to a Coulomb blockade of the single junction when the zero-bias resistance of the SQUID arrays is much higher than the quantum resistance  $R_K = h/e^2 \approx 26 \text{ k}\Omega$ .

In addition to Coulomb blockade, we have clearly observed a region of negative differential resistance in the *I*-*V* curve. Negative differential resistance has been reported in one-dimensional arrays [6] and in twodimensional arrays [7], however, clear observation in single junctions has not yet been reported in the literature.

In the context of the theory of current-biased single Josephson junctions [1,2], the negative differential resistance appears as a result of coherent tunneling of single Cooper pairs. The onset of the coherent tunneling is characterized by the local voltage maximum in the low-current part of the *I*-*V* curve, or blockade voltage  $V_b$ . The value of  $V_b$  is suppressed as the ratio  $E_J/E_C$  is increased. Here, *E<sub>J</sub>* is the Josephson energy and  $E_C = e^2/2C$ , where *C* is the capacitance of the single junction. We show that the  $E_J/E_C$  dependence of our measured  $V_b$  is consistent with the theoretical prediction.

A scanning electron micrograph of an  $Al/Al_2O_3/Al$ sample is shown in Fig. 1. A single junction with the area of  $0.1 \times 0.1 \ \mu m^2$  is in the center of Fig. 1. On each side of the single junction there are two leads enabling four-point measurements of the single junction. A part of each lead close to the single junction consists of an array of dc SQUIDs. The area of each junction in the leads is  $0.3 \times 0.1 \mu m^2$  and the effective area of the SQUID loop is  $0.7 \times 0.2 \ \mu \text{m}^2$ . All of the samples have the same configuration, except for the number *N* of junction pairs in each lead. In Fig. 1,  $N = 17$  is shown, however, we measured the samples with larger *N*, up to 225. The tunnel junctions



FIG. 1. A scanning electron micrograph of a sample.

are fabricated on a  $SiO<sub>2</sub>$  substrate using electron-beam lithography and a double-angle-evaporation technique [8]. The oxidation conditions, or the thickness of the  $Al_2O_3$ layer, determines the tunnel resistance of the junctions. The samples are characterized by the normal-state resistance  $R_n$  of the single junction, the normal-state resistance *rn* per junction pair of the SQUID arrays, and *N*.

The samples were measured in a  ${}^{3}$ He- ${}^{4}$ He dilution refrigerator mainly at the base temperature  $(0.02-0.04 \text{ K})$ . The temperature was determined by measuring the resistance of a ruthenium-oxide thermometer fixed at the mixing chamber. To measure the resistance of the thermometer, we used an ac resistance bridge (RV-Elekroniikka, AVS-47). The samples were placed inside a copper rf-tight box which was thermally connected to the mixing chamber. All the leads entering the rf-tight box were low-pass filtered by 1 m of thermocoax cable [9]. We measured the *I*-*V* curve of the single junction in a four-point configuration, where the potential difference was measured through one pair of SQUID-array leads with a high-input-impedance instrumentation amplifier based on two operational amplifiers (Burr-Brown OPA111BM) and a preamplifier [Stanford Research Systems (SRS) SR560]. Through the other pair of SQUID-array leads, the bias was applied, and the current was measured with a current preamplifier (SRS SR570). When the voltage drop at the SQUID arrays was much larger than that at the single junction, the single junction was practically current biased. The SQUID arrays could be measured in a two-point configuration (same current and voltage leads) on the same side of the single junction. Note that the two arrays are connected in series and that current does not flow through the single junction. In order to obtain  $R_n$ and  $r_n$ , we measured at  $T = 2-4$  K (above the superconducting transition temperature of Al).

The effective Josephson energy between adjacent islands in the SQUID arrays is proportional to  $|\cos(\pi BA/\Phi_0)|$ , where *B* is the external magnetic field applied perpendicular to the substrate, *A* is the effective area of the SQUID loop, and  $\Phi_0 = h/2e = 2 \times 10^{-15}$  Wb is the superconducting flux quantum. Figure 2 shows the zero-bias resistance  $r_0$  as a function of *B*, of two SQUID arrays on the same side of the single junction. The arrays have  $r_n = 1.1 \text{ k}\Omega$  and  $N = 225$ . In our samples with  $A = 0.14 \ \mu \text{m}^2$ ,  $B A/\Phi_0$  becomes 1/2 at  $B = 7.4 \text{ mT}$ . Thus,  $r_0$  should have the first local maximum at 7.4 mT, as observed in Fig. 2. The value of  $r_0$  at the local maximum is more than  $10^7$  times larger than that at  $B = 0$ . This means that we can tune the electromagnetic environment for the single junction over a wide range. Henceforth we indicate the magnetic field as the frustration  $f = BA/\Phi_0$ .

In Figs. 3–5, we show some results on a sample with  $R_n = 17 \text{ k}\Omega$ ,  $r_n = 1.4 \text{ k}\Omega$ , and  $N = 65$ . The *I*-*V* curves for the single junction at several frustrations are shown in Fig. 3. The vertical broken lines represent the superconducting energy gap  $\pm 2\Delta$ , which is  $\pm 0.34$  meV for Al



FIG. 2. Zero-bias resistance of two SQUID array leads connected in series for a sample with  $r_n = 1.1 \text{ k}\Omega$  and  $N = 225$ .

[10]. The features within the gap are dependent on *f* especially in the vicinity of  $V = 0$ . A close-up of the region denoted by the boxes with dotted lines is shown in Fig. 4. As *f* is varied, the single-junction *I*-*V* curve shown in Fig. 4 develops a Coulomb blockade. We reemphasize that the Josephson energy of the single junction is independent of *f*, because it does not have a SQUID configuration



FIG. 3. Current-voltage characteristics of the single junction for the sample with  $R_n = 17 \text{ k}\Omega$ ,  $r_n = 1.4 \text{ k}\Omega$ , and  $N = 65$ . From top to bottom, the frustration  $f = BA/\Phi_0$  is 0, 0.22, 0.41, 0.45, and 0.49, respectively. The origin of the current axis is offset for each curve for clarity. The vertical broken lines and the boxes with dotted lines represent the superconducting energy gap  $\pm 2\Delta$  and the region that will be closed up in Fig. 4, respectively.



FIG. 4. Current-voltage  $(I-V)$  curves of the single junction for the same sample as in Fig. 3. From top left to bottom right, the frustration *f* is increased from 0.43 to 0.49 in steps of 0.01. The origin of each curve is offset for clarity. For the labeled curves, the *I*-*V* characteristics of the leads at the same *f* are shown in Fig. 5.

and the field  $f\Phi_0/A$  applied here is much smaller than the critical field for Al films ( $\approx 0.1$  T). The electromagnetic environment for the single junction (the SQUID array), however, is strongly varied with *f*. Figures 3 and 4 demonstrate that the low-bias region of the single-junction *I*-*V* curve is indeed sensitive to the environment.

The *I*-*V* curves of the two SQUID-array leads connected in series at  $f = 0.43$   $(r_0 = 0.61$  M $\Omega)$ , 0.46  $(r_0 =$ 3.2 M $\Omega$ ), and 0.49  $(r_0 = 43 \text{ M}\Omega)$  are shown in Fig. 5. The *I*-*V* curves of the leads are nonlinear, and in general the SQUID array cannot be described by a liner impedance model [6]. However, we may characterize the environment by their  $r_0$ . Coulomb blockade is visible only when  $r_0 \gg R_K$ , which is consistent with the theoretical conditions for the clear observation of Coulomb blockade in single junctions [11]. For an arbitrary linear environment characterized by  $Z_e(\omega)$ , Re[ $Z_e(\omega)$ ]  $\gg R_K$  is required for the Coulomb blockade of single-electron tunneling and  $\text{Re}[Z_e(\omega)] \gg R_K/4$  for that of Cooper-pair tunneling [11]. However, as seen in Fig. 5, our SQUID arrays are a nonlinear environment. It is interesting to note that at  $f = 0.46$  (labeled "b"), the *I*-*V* curve of the leads is still "Josephson-like" (differential resistance is



FIG. 5. Current-voltage curves of the two SQUID-array leads connected in series for the same sample as in Figs. 3 and 4. From *a* to *c*, the frustration is 0.43, 0.46, and 0.49, respectively.

lower around  $V = 0$ , while that of the single junction is already "Coulomb-blockade-like." This feature becomes more distinct in samples with larger *N*.

The region of negative differential resistance seen in Fig. 4 when Coulomb blockade is well developed, is related to coherent tunneling of single Cooper pairs according to the theory [1,2] of a current-biased single Josephson junction in an environment with sufficiently high impedance. For sufficiently low current, the electrical conduction of single Josephson junctions is governed by stochastic quasiparticle tunneling, and the *I*-*V* characteristic is highly resistive. As the current is increased, coherent single-Cooper-pair tunneling, or "Bloch oscillation" dominates, decreasing the mean voltage. As a result, the *I*-*V* curve has a region of negative differential resistance, or "back-bending" in the low-current part.

Theoretically, the value of the local voltage maximum, or blockade voltage  $V_b$ , is a function of the ratio  $E_J/E_C$ , and given by

$$
V_b \approx \begin{cases} 0.25e/C & \text{for } E_J/E_C \ll 1, \\ \delta_0/e & \text{for } E_J/E_C \gg 1, \end{cases}
$$
 (1)

as  $T \rightarrow 0$ , where

$$
\delta_0 = \frac{e^2}{C} 8 \left( \frac{1}{2\pi^2} \right)^{1/4} \left( \frac{E_J}{E_C} \right)^{3/4} \exp \left[ - \left( 8 \frac{E_J}{E_C} \right)^{1/2} \right] \quad (2)
$$

is the half-width of the lowest energy band [2]. When quasiparticle tunneling is neglected,  $V_b$  becomes larger [12],

$$
V_b \approx \begin{cases} e/C & \text{for } E_J/E_C \ll 1, \\ \pi \delta_0/e & \text{for } E_J/E_C \gg 1. \end{cases}
$$
 (3)

The calculation of  $V_b$  for arbitrary  $E_J/E_C$  has also been done for the case of no quasiparticle tunneling [13].



FIG. 6. Blockade voltage  $V_b$  divided by  $e/C$  as a function of  $E_I/E_C$ . From top to bottom, the curves represent the theoretical predictions of Refs. [12], [13], and [2], respectively. For the calculation of the solid symbols (lower data set) 45 fF/ $\mu$ m<sup>2</sup> is assumed, and for the open symbols (upper data set) 200 fF/ $\mu$ m<sup>2</sup>. The solid boxes and the open boxes represent the samples with the nominal junction area of 0.01  $\mu$ m<sup>2</sup> in Ref. [3].

In Fig. 6, we compare the measured  $V_b$  with the above predictions. We used  $E_J = h\Delta/8e^2R_n$ , and for  $E_C$  we estimated *C* from the junction area. A value of specific capacitance  $c_s = 45 \pm 5$  fF/ $\mu$ m<sup>2</sup> [14], which was obtained for the junctions with 3  $\times$  28  $\mu$ m<sup>2</sup> and 7  $\times$  54  $\mu$ m<sup>2</sup>, has been frequently employed [3,5,6]. Uncertainty in  $c_s$ , however, seems to be much larger when the junction area is on the order of 0.01  $\mu$ m<sup>2</sup> or smaller.

In the following experimental literature on small tunnel junctions, *C* was estimated from the offset voltage in the normal-state *I*-*V* curve. Fulton and Dolan measured samples with three junctions that share a common electrode, and obtained 0.20-0.23 fF for  $(0.03 \pm 0.01 \ \mu \text{m})^2 \times 3$  [15], i.e.,  $c_s = 42 - 192 \text{ fF}/\mu \text{m}^2$ . Geerligs *et al.* reported  $c_s \approx 110 \text{ fF}/\mu \text{m}^2$  for twodimensional  $(190 \times 60)$  junction arrays with the areas of 0.01 or 0.04  $\mu$ m<sup>2</sup> [7]. More recently, Penttilä *et al.* studied resistively shunted single Josephson junctions with the area of  $0.15 \times 0.15 \ \mu \text{m}^2$  [16]. The estimated *C* of their eight samples ranged between 0.8 and 6.6 fF, or  $c_s = 36 - 293$  fF/ $\mu$ m<sup>2</sup>.

Motivated by the above survey, in Fig. 6 we plot the experimental  $V_b$  vs  $E_J/E_C$ , where two different values of *cs* have been used to determine *C*. The lower data set uses 45 fF/ $\mu$ m<sup>2</sup> and the upper data set 200 fF/ $\mu$ m<sup>2</sup>. The data from Ref. [3] for the samples with the same nominal junction area  $(0.1 \times 0.1 \mu m^2)$  as in this work are also plotted. The measured  $V_b$  agrees with the theoretical predictions qualitatively. We also find a quantitative agreement when  $c_s \approx 200 \text{ fF}/\mu \text{m}^2$  is taken for 0.01  $\mu \text{m}^2$ . This apparently large junction capacitance may be partly explained by distributed capacitance of the SQUID arrays.

In summary, we have studied the current-voltage characteristics of single Josephson junctions biased with SQUID arrays. We have demonstrated that the Coulomb blockade in the single junction is indeed sensitive to the state of the environment. The region of negative differential resistance has been clearly observed in the *I*-*V* curve of the single junction, when the zero-bias resistance of the SQUID arrays is much higher than  $R_K$ , which is clear evidence of coherent single-Cooper-pair tunneling in the single Josephson junction.

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