Spectroscopy of Energy-Level Splitting between Two Macroscopic Quantum States of Charge Coherently Superposed by Josephson Coupling

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We study Cooper-pair tunneling in a voltage-biased superconducting single-electron transistor under microwave irradiation. By tracing the peak positions of a photon-assisted Josephson-quasiparticle current as a function of the microwave frequency, we observe an energy-dispersion curve in the quasicharge space. This shows that energy-level splitting occurs between two macroscopic quantum states of charge coherently superposed by Josephson coupling. [S0031-9007(97)04097-0]

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Charging effect in systems of small Josephson junctions has been studied extensively in recent years and has given us many new insights into the quantum conjugate properties of phase and number [1,2]. In this work, with the aid of charging energy, we restrict the freedom of charge number in a small Josephson junction system—that is, we break a translational symmetry in the number space—and observe evidence of coherent superposition of two charge states. This is a macroscopic two-level system where, due to the charging effect, a single Cooper-pair tunneling event is accompanied with the entire redistribution of the polarized charge on the junction capacitors in the device. We also demonstrate the spectroscopic measurement of Josephson energy in a small Josephson junction, which would not be possible, in principle, in a Josephson junction with a macroscopic size.

In two isolated superconducting electrodes connected by a large Josephson junction with negligible charging energy, the phase difference φ at the junction is a good quantum number. The number n of excess electrons on one electrode, which is conjugate with φ, is uncertain, and many number states $|n\rangle$ with different n are coherently superposed. Here, Josephson energy E_J represents the *band width in the phase space,* as shown in a one-dimensional tight-binding Hamiltonian

$$H = -\frac{E_J}{2} \sum_n{}' \{|n\rangle\langle n+2| + |n+2\rangle\langle n|\} = -E_J \cos\varphi \,, \tag{1}$$

where the sum $\sum'$ includes only n for the same parity. Let us consider only the $2e$ (Cooper-pair) tunneling process between even-n states for the moment.

When the size of the junction is decreased so that the charging energy becomes dominant, the degeneracy between the different number states is lifted and the fluctuation of n is suppressed. However, in a superconducting single-electron box (S-SEB) circuit [left-hand side of Fig. 1(a)] [3], the electrostatic potential of the box electrode can be controlled by the gate voltage V_g. When the total induced charge (or quasicharge) Q_0 on the box is an odd integer m—that is, $Q_0 \equiv C_g V_g + Q_b = me$—the two number states, $|m-1\rangle$ and $|m+1\rangle$, again degenerate. Here C_g is the gate capacitance and Q_b is the background charge. Cooper-pair tunneling occurs between the

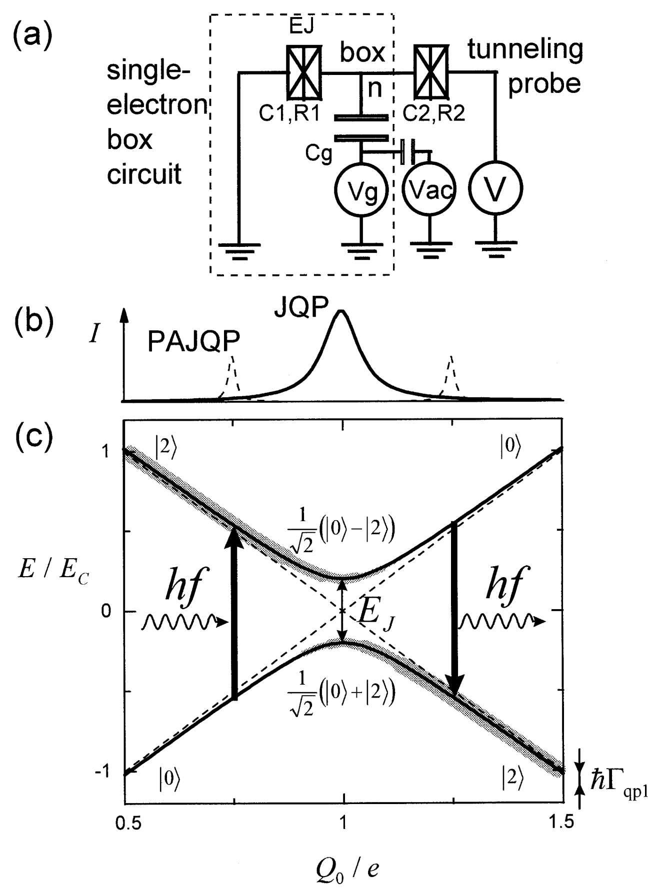

FIG. 1. (a) Schematic circuit diagram of an S-SET, which consists of an S-SEB and a probe junction. (b) Schematic I-Q_0 curve at a fixed V illustrating JQP (solid) and PAJQP (dashed) current peaks. (c) Energy diagram illustrating energy levels (solid curves) of an S-SEB as a function of Q_0 in the presence of the probe. Dashed lines show those for the case without Josephson coupling. Large arrows show photon-assisted processes which produce PAJQP current. Shadows around the energy levels illustrate level broadening due to the quasiparticle tunneling dissipation.

two states in resonance. For simplicity, we will restrict ourselves to the resonance between two states $|0\rangle$ and $|2\rangle$ at $m = 1$ without losing generality. The effective Hamiltonian can be written as the sum of a charging energy term and a Josephson coupling term,

$$H = E_C(\hat{n} - Q_0/e)^2 - \frac{E_J}{2}\{|0\rangle\langle 2| + |2\rangle\langle 0|\}, \qquad (2)$$

where $E_C \equiv e^2/2C_\Sigma$, C_Σ is the total capacitance of the box, and $\hat{n}$ is the number operator of excess electrons in the box. The resulting eigenstates, due to coherent superposition of the two states defined by the macroscopic number n, form two energy bands *in the quasicharge space* separated with a *band gap* of E_J [1].

Such coherent superposition between two different charge states has been observed both in an S-SEB and in related devices. Rounding of the steps in the expectation value of the charge number in an S-SEB has been measured by using a single-electron transistor (SET) electrometer [4]. Bloch oscillation in a current-biased small Josephson junction [5] and supercurrent in a superconducting single-electron transistor (S-SET) [6] have also indicated coherence between different charge states. Those measurements, however, focused on the ground-state properties of the coherently superposed systems, leaving the excited state unexplored. In this Letter, we report on our investigation of the excitation gap, where we measured the Josephson-quasiparticle (JQP) current [7–9] and photon-assisted JQP (PAJQP) current [10] through an S-SET at finite bias voltages. Recently, another group has also studied the excited state, but by a different method [11]. We can also find related physics in semiconductor systems, such as coupled double quantum dots [12] and wells [13].

The basic concept of our measurement is to detect the charge state in the S-SEB by using an additional superconducting tunneling probe [14]. As shown in Fig. 1(a), an S-SET can be considered as an S-SEB with a tunnel junction attached to the box. We can selectively detect charge state $|2\rangle$ in the S-SEB by tuning the chemical potential of the probe electrode so that two quasiparticles can sequentially tunnel out to the probe electrode from the state $|2\rangle$ ($|2\rangle \rightarrow |1\rangle \rightarrow |0\rangle$), while also preventing any tunneling from the state $|0\rangle$. The state is reset to $|0\rangle$ after the detection of the state $|2\rangle$, so cyclic charge transport, which involves one Cooper-pair tunneling and two quasiparticle tunnelings, is possible if Cooper-pair tunneling is possible on the S-SEB side for the transition $|0\rangle \rightarrow |2\rangle$. As a result, a JQP current is observed [7]. Thus, qualitatively, the JQP current we observe tells us whether there is Cooper-pair tunneling for the transition $|0\rangle \rightarrow |2\rangle$ and at what rate it occurs. Since state $|1\rangle$ is not stable against the quasiparticle tunneling at the probe junction, we do not have to worry here about the so-called poisoning effect of the resonance [6]. For the same reason, the parity of the charge number is irrelevant in this discussion, though we have considered only even-n states for resonance so far. Note that here we ignore other tunneling processes, such as quasiparticle tunneling on the S-SEB side and Cooper-pair tunneling at the probe junction. We can choose the bias condition so that the superconducting gap Δ in the density of states and the nondissipative nature of Cooper-pair tunneling, respectively, make the contributions from these two other tunneling processes negligibly small, at least at low temperatures and with little environmental coupling. For details, see Refs. [7–9,15].

Figure 1(c) illustrates the energy levels of the S-SEB as a function of Q_0 in the presence of the probe. From now on we will write $Q_0 = C_g V_g + C_2 V + Q_b$ to include the contribution of the finite probe voltage V, where C_2 is the capacitance of the probe junction. In the plot, an n-independent term $(Q_0 - e)^2/2C_\Sigma$ in energy is subtracted. Two bare charge states $|0\rangle$ and $|2\rangle$ (shown as dashed lines) have an electrostatic energy difference $\delta E(Q_0) \equiv 4E_C(Q_0/e - 1)$. Because of the Josephson coupling between the two charge states, the total energy difference is

$$\Delta E(Q_0) = \sqrt{\delta E(Q_0)^2 + E_J^2}, \qquad (3)$$

which gives an energy gap of E_J at the resonance. The JQP current in an I-Q_0 curve [the solid curve in Fig. 1(b)] has a peak at the resonant point where the Cooper-pair tunneling $|0\rangle \rightarrow |2\rangle$ occurs most frequently. The peak can be used as a marker of the resonance.

Microwave irradiation induces photon-assisted Cooper-pair tunneling [10]. The photon-assisted process which involves absorption or emission of a single photon is depicted in Fig. 1(c) as a large arrow. It opens another channel for the transition $|0\rangle \rightarrow |2\rangle$ and results in a PAJQP current that has a peak when the photon energy hf matches $\Delta E(Q_0)$ [the dashed curves in Fig. 1(b)]. Thus, by measuring the Q_0 shift of the PAJQP peak relative to the JQP peak as a function of microwave frequency f, we can trace the energy-gap dispersion relation in Fig. 1(c).

Concerning the peak width, we have to take into account the effect of decoherence. As long as we consider the ideal case described above, the only decoherence source is the first of the two sequential quasiparticle tunnelings (which have rates Γ_{qp1} and Γ_{qp2}, respectively) at the probe junction [8]. Hence, the state $|2\rangle$ is broadened by its lifetime $1/\Gamma_{qp1}$ as depicted in Fig. 1(c) as shadows around the energy levels. To clearly observe the energy gap E_J due to the coherent superposition, we must ensure that the broadening is smaller than the gap; that is, $E_J > \hbar\Gamma_{qp1}$ [12].

Our Al S-SET was fabricated by electron-beam lithography and a shadow evaporation technique. Since the Josephson energy on the S-SEB side $E_J = h\Delta/8e^2R_1$ [16] and the first quasiparticle tunneling rate at the probe junction $\Gamma_{qp1} \sim 2(\Delta + E_C)/e^2R_2$ [9] are both inversely proportional to the tunnel junction resistance, the ratio $E_J/\hbar\Gamma_{qp1}$ is always less than unity for S-SETs with nearly symmetric junction resistances, $R_1 \simeq R_2$. Hence, to realize $E_J > \hbar\Gamma_{qp1}$, we had to make an S-SET with asymmetric junction resistances $R_1 \ll R_2$. To do this, we made the two junctions with different areas and used three-angle shadow evaporation so that the barrier thickness of the smaller junction could be increased by using an additional oxidation process. Judging from the I-V-V_g measurements both in the normal and superconducting states, the capacitances of the junctions and the gate are $C_1 = 490 \pm 10$ aF, $C_2 = 20.8 \pm 0.4$ aF, $C_g = 2.66 \pm 0.03$ aF, the total resistance is $R_1 + R_2 \simeq 23$ MΩ, $E_C = 155 \pm 5$ μeV, and $\Delta \simeq 200$ μeV. We could not estimate accurately the asymmetry in the junction resistances from the measurements. Nevertheless, much of the evidence presented later indicates that we are in the desired regime $E_J > \hbar\Gamma_{\text{qp1}}$. In addition, though it is not shown explicitly in Fig. 1(a), the junction on the S-SEB side is split into two to form a SQUID loop with an area of about 1 μm^2 which allows us to control its effective Josephson energy with a small external magnetic field.

In our experiment, the sample was voltage biased, and the dc current and voltage were measured with battery-powered preamplifiers. The sample was mounted in a shielded copper box on the mixing chamber of a dilution refrigerator (~30 mK) and all the dc-measurement lines were filtered with copper-powder microwave filters at the entrance of the box. Microwave irradiation for photon-assisted transport experiments was introduced to the sample box by a CuNi coaxial cable. We connected this line, though it did not seem to couple exclusively, to the gate electrode of the sample via a −50-dB attenuator at the top of the cryostat and a commercial capacitive coupler at the entrance of the box that eliminated dc coupling. Although the coupling strength of the microwave to the sample was not uniform over the entire frequency range, we confirmed that the peak position of the photon-assisted current is insensitive to the microwave power, at least in the low-power regime used in this experiment.

Figure 2 shows I-Q_0 curves at $V = 730$ μV with microwave irradiation of 32.2 GHz for three E_J values from a maximum ($\phi/\phi_0 = 0$) to a minimum ($\phi/\phi_0 = 1/2$) controlled through a magnetic field, where ϕ is the magnetic flux in the SQUID loop and $\phi_0 \equiv h/2e$. Because of the possible asymmetry in the SQUID, we are not sure about the size of the residual E_J in the minimum case. The width of the JQP peak decreased rapidly as E_J decreased, which implies that the width was mostly determined by E_J as expected in the case where $E_J > \hbar\Gamma_{\text{qp1}}$; otherwise the peak width would be proportional to $\hbar\Gamma_{\text{qp1}}$ and independent of E_J [8,9]. On the other hand, the PAJQP peaks had a nearly E_J-independent width that was much narrower than that of the JQP peak. The width of the PAJQP peak should be on the same order as the level broadening $\hbar\Gamma_{\text{qp1}}$, at least when $hf \gg E_J$ [12]. Thus, the large difference in width between the JQP and the PAJQP peaks is additional evidence that $E_J > \hbar\Gamma_{\text{qp1}}$. This is in contrast to the case where $E_J < \hbar\Gamma_{\text{qp1}}$, where JQP and PAJQP peaks have similar widths [10,12]. In addition, there is a $2e$-periodic current shoulder marked as a parity effect in Fig. 2. This originated from the tunneling of a single residual quasiparticle in the ground state of the odd-n state, and is inversely proportional to the volume of the box electrode [17,18]. By taking into account the volume of the box electrode (~600 × 50 × 20 nm^3), fitting of the shoulder height (~3 pA) also indicates that there is a huge asymmetry in the junction resistances. It also allows us to estimate R_1 to be ~10 kΩ.

In Fig. 3, we show the positions of automatically detected peaks in I-Q_0 curves as a function of the microwave frequency for (a) the minimum E_J and (b) the maximum E_J. To compensate for the background charge fluctuation during the measurement, Q_0 is shifted so that the position of the JQP peak is adjusted to $Q_0/e = 1$ for each frequency. With the minimum E_J, PAJQP peaks appear on the dashed lines which represent $hf = \delta E(Q_0)$. Note that here we used only experimentally determined capacitances to calculate the relation and used no fitting parameters. For the maximum E_J, the dispersion curve shows a nonlinear behavior. This clearly indicates energy-level splitting due to Josephson coupling between two charge states and is approximately fitted with a calculated curve $hf = \Delta E(Q_0)$ for $E_J = 50$ μeV [Fig. 3(b)]. Since the minimum energy gap corresponds to E_J, these results demonstrate the first spectroscopic measurement of Josephson energy. According to the Ambegaokar-Baratoff relation [16], the obtained E_J corresponds to $R_1 \sim 13$ kΩ, which is consistent with our estimation above.

While this explanation of the peak positions works well, several findings remain that were not expected from the theories [8,12]. Asymmetry which increased with E_J was observed in the PAJQP peak height between the photon-absorption side and the photon-emission side. This could be partly explained by taking into account the Q_0 dependence of quasiparticle tunneling rates, but the asymmetry in the experiment still seems too large. Even without the microwave irradiation, the JQP peak itself has an asymmetric shape that is broader on the

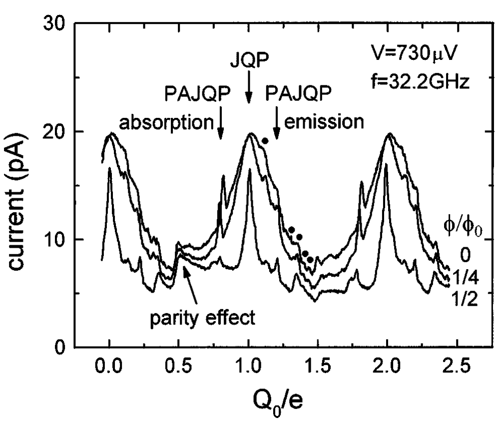

FIG. 2. I-Q_0 curves at $V = 730$ μV under 32.2 GHz microwave irradiation for three E_J values from the maximum ($\phi/\phi_0 = 0$) to the minimum ($\phi/\phi_0 = 1/2$). The curves are not offset. Black dots denote unspecified peaks whose positions did not depend on the microwave frequency.

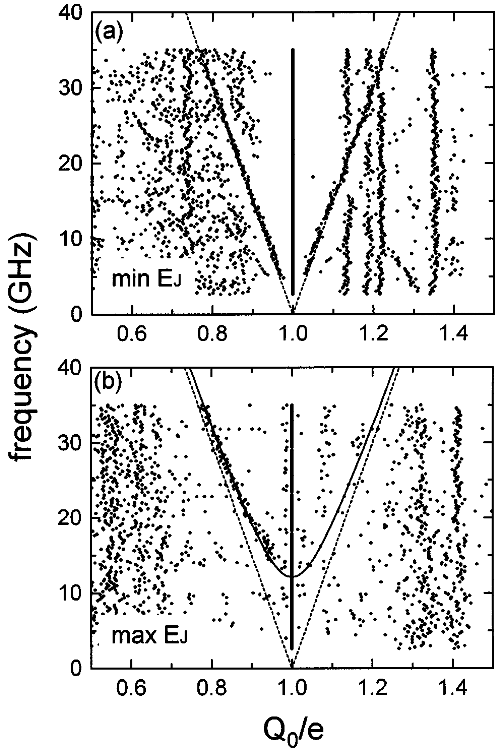

FIG. 3. Positions of peaks (dots) in I-Q_0 curves at $V = 730\ \mu$V as a function of the frequency of irradiated microwaves with (a) the minimum E_J and (b) the maximum E_J. For each frequency, Q_0 is shifted so that the position of the JQP peak is adjusted to $Q_0/e = 1$ in order to compensate for the background charge fluctuation. The solid curve is a calculated dispersion curve $hf = \sqrt{\delta E(Q_0)^2 + E_J^2}$ with $E_J = 50\ \mu$eV. The dashed lines show those with $E_J = 0$.

emission side. Furthermore, the observed JQP peak width ($\sim$250 μeV in energy scale [19]), is much larger than the expected value $E_J\sqrt{2 + \Gamma_{\mathrm{qp1}}/\Gamma_{\mathrm{qp2}}} \sim 90\ \mu$eV [8,12] for $E_J = 50\ \mu$eV. It may be possible to account for these discrepancies by introducing inelastic transitions (relaxation and excitation) between the energy levels in Fig. 1(c). The relaxation would broaden the JQP peak on the emission side, whereas the excitation would increase current on the absorption side. In addition, the JQP peak has several unknown peaks, only on the emission side, whose positions do not depend on the microwave frequency but differ from sample to sample. These peaks are denoted in Fig. 2 by black dots, as well as in Fig. 3, as frequency-independent peak positions. An energy-dependent inelastic transition rate may explain these peaks, since such peaks are likely when the transitions are due to coupling with an environment with an energy-dependent spectrum. Although they cannot be clearly seen in Fig. 3(b), we observed that the positions of these peaks shifted according to Eq. (3) as E_J increased.

The width of the observed PAJQP peak $\sim$16 μeV is also too large compared with $\hbar\Gamma_{\mathrm{qp1}} \sim 0.2\ \mu$eV at $V = 730\ \mu$V calculated by using $R_2 \sim 23$ MΩ. Though the origin of the extra broadening is not understood, the observed width could be an important measure of the quantum coherence time in our two-level system, if the decoherence is the main contributor to the broadening.

In summary, in our study of Cooper-pair tunneling in a voltage-biased superconducting single-electron transistor under microwave irradiation, we observed shifts of photon-assisted Josephson-quasiparticle current peaks as a function of the microwave frequency, and obtained an energy-dispersion curve in the quasicharge space. This shows that energy-level splitting occurs between two macroscopic quantum states of charge coherently superposed by Josephson coupling.

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