Combined Single-Electron and Coherent-Cooper-Pair Tunneling in Voltage-Biased Josephson Junctions

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Small-capacitance tunnel junctions show single-electron effects and, in the superconducting state, the coherent tunneling of Cooper pairs. We study these effects in a system of two voltage-biased Josephson junctions in series. Novel features show up in the *I-V* characteristics, in particular, pronounced structures at subgap voltages. These are due to combined quasiparticle and pair tunneling and to higher-order Cooper-pair tunneling processes. Several of them have been observed in experiments.

In small capacitance Josephson junctions the combination of coherent Cooper-pair tunneling and single-electron tunneling yields interesting nonclassical features in the *I-V* characteristics [1,2]. Unless a high-Ohmic resistor is placed close to the junction [3] or systems of junctions are considered [4], the junction is effectively voltage biased by the capacitance of the leads. In this Letter we consider the simplest system of voltage-biased Josephson junctions where Coulomb effects can be observed. It consists of two junctions [5] as shown in Fig. 1. This system has been studied by Fulton *et al.* [6] for currents in the range of $I \approx e/R_n C$ (≈ 1 nA). In these experiments, as well as in those we report, the junctions have ideal quasiparticle tunneling properties with a subgap conductance orders of magnitude smaller than the normal-state value $1/R_n$. The Josephson coupling energy E_J is lower than the scale for the charging energy E_C, but the superconducting gap is larger, $\Delta > E_C$. For voltages above $4\Delta/e$ the tunneling is characterized by normal-state properties and Josephson effects are negligible. At lower voltages, but above $2\Delta/e$, a current peak is found [6] due to a "Josephson-quasiparticle" cycle: the resonant tunneling of a Cooper pair across one junction followed by two quasiparticles tunneling across the other. In this process the energy gain is large enough that the quasiparticles tunnel with rates given by the normal-state conductance. Recently, on a smaller current scale (1–100 pA, i.e., still larger than typical subgap currents) much more structure has been observed in the *I-V* characteristics [7]. In this Letter we present further experimental results and give a theoretical interpretation based on the quantum-mechanical description of Josephson junctions.

The Hamiltonian of the system shown in Fig. 1 consists of the Coulomb energies associated with the charges on the capacitances, the work done on the voltage source (*V*), and the Josephson coupling energies depending on the phase differences ϕ_i. We also account for a gate voltage U coupled to the central island via a capacitance C_0. The overall phase difference, which is fixed by the applied voltage $\phi_2 + \phi_1 = 2eVt/\hbar$, is a classical variable. However, the phase of the island $\phi \equiv (\phi_2 - \phi_1)/2$ and the island charge Q are quantum-mechanical variables $Q = (\hbar/i)\partial/\partial(\hbar\phi/2e)$. We define $C_\Sigma \equiv C_0 + C_1 + C_2$, and for later use $E_{C_\Sigma} \equiv e^2/2C_\Sigma$. Then,

$$H_0 = \frac{Q^2}{2C_\Sigma} + \frac{Q}{C_\Sigma}\left[\frac{C_1 - C_2}{2}V + C_0 U\right]$$
$$- E_{J1}\cos\left[\frac{eV_t}{\hbar} - \phi\right] - E_{J2}\cos\left[\frac{eVt}{\hbar} + \phi\right]. \quad (1)$$

The term linear in Q can be removed from H_0 by a unitary transformation $\hat{\psi}(\phi,t) \equiv \psi(\phi,t)\exp(iq_0\phi/2)$. The "quasicharge" $q_0 = C_0 U/e + (C_1 - C_2)V/2e$ is a polarization charge induced by the gate voltage and an asymmetry of the capacitances C_1 and C_2. (Charged impurities polarizing the island lead to a further shift of q_0.) Next to the Hamiltonian we have to specify the states of the system. Since Cooper-pair and quasiparticle tunneling change the island charge only by integer multiples of e, the plane waves $\exp(iq\phi/2)$, with q an integer, provide a basis.

The Hamiltonian depends on time. Since voltages of interest are of the order of the charging energy $eV \approx E_{C_\Sigma}$ and typical frequencies are not small, an adiabatic approximation is not sufficient. On the other hand, the time dependence is periodic and Floquet's theorem applies: The solutions of Schrödinger's equation can be written as $\hat{\psi}(\phi;t) = \exp(-i\Omega t)u(\phi,t)$, where $u(\phi,t)$ depends periodically on time. After Fourier expansion we have

$$\hat{\psi}(\phi,t) = \sum_{q,n} g_{q,n} \exp\left[i(q_0+q)\frac{\phi}{2} - i\left(\Omega + \frac{neV}{\hbar}\right)t\right]. \quad (2)$$

These "time-Bloch" states are superpositions of states with different energy (which is not conserved in the voltage-driven system). Furthermore, they are superpositions of states with different charges, $(q_0+q)e$. The time-dependent Schrödinger equation reduces to an eigenvalue
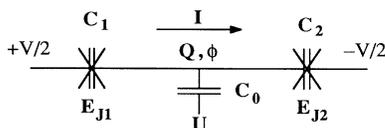

FIG. 1. The circuit of two Josephson junctions.

problem for the "quasienergy" $\omega \equiv \hbar \Omega / E_{C_\Sigma}$:

$$\omega g_{q,n} = [(q+q_0)^2 - 2nv]g_{q,n} - \frac{1}{2}\frac{E_{J1}}{E_{C_\Sigma}}(g_{q+2,n+1}+g_{q-2,n-1}) - \frac{1}{2}\frac{E_{J2}}{E_{C_\Sigma}}(g_{q+2,n-1}+g_{q-2,n+1}) . \tag{3}$$

We defined $v \equiv VC_\Sigma/e$. Except for an overall phase factor, the coefficients $g_{q,n}$ can be chosen real. By solving the eigenvalue problem we treat the Josephson coupling nonperturbatively. We have done this numerically, taking into account a finite number of coefficients with $|q| \le q_m$, $|n| \le n_m$, but we checked at all times the convergence of this procedure. For typical values $E_J/E_{C_\Sigma} \approx 0.2$ the restriction to $q_m = 5$ and $n_m = 4$ yields good results. From the resulting $(2q_m+1)(2n_m+1)$ eigenvectors only $2q_m + 1$ are needed for a basis of solutions. The other eigenvectors are copies of these basic ones corresponding to a different choice of the "time Brillouin zone." For the basic eigenvectors the choice $|\Sigma_{q,n}ng_{q,n}^2| < \frac{1}{2}$ minimizes truncation effects.

Without dissipation, a stationary gate voltage leads to an ac supercurrent. A dc current can flow due to quasiparticle tunneling. We can include it by using the tunnel

Hamiltonian of the system (see Ref. [2]) or, since we do not want to rederive Josephson effects, by using the following model [8]:

$$H = H_0 + H_{\text{bath}} + H_T ,$$

$$H_{\text{bath}} = \sum_k \sum_{j=1,2} \varepsilon(k)c_{kL_j}^\dagger c_{kL_j} + \{L \leftrightarrow R\} , \tag{4}$$

$$H_T = \sum_{j=1,2} \frac{a_j^{1/2}}{2N(0)}\exp(-i\phi_j/2)\sum_{k,k'}c_{kL_j}^\dagger c_{k'R_j} + \text{H.c.}$$

The c's describe quasiparticles in the two electrodes L,R for each junction $j=1,2$ (which are assumed to be independent). Their densities of states depend on the gap Δ, which leads to the step in the conductance. The parameters $a \equiv 2\hbar/\pi e^2 R_n$ and $N(0)$ refer to normal-state properties. From H_T we can calculate the quasiparticle transition rates between different time-Bloch states, in the way that the "golden rule" is derived. They are

$$W_{i \to f} = \sum_s (W_{1,i \to f}^{+,s} + W_{1,i \to f}^{-,s} + W_{2,i \to f}^{+,s} + W_{2,i \to f}^{-,s}) ,$$

$$W_{1,i \to f}^{\pm,s} = \frac{1}{R_1 C_\Sigma}\text{Th}[\omega_f - \omega_i + 2v(s \mp \tfrac{1}{2})]\left(\sum_{q,n}g_q^f{}_{\pm 1,n+s}g_{q,n}^i\right)^2 , \tag{5}$$

$$W_{2,i \to f}^{\pm,s} = \frac{1}{R_2 C_\Sigma}\text{Th}[\omega_f - \omega_i + 2v(s \mp \tfrac{1}{2})]\left(\sum_{q,n}g_q^f{}_{\mp 1,n+s}g_{q,n}^i\right)^2 .$$

We defined a function $\text{Th}[x] \equiv x[\exp(xE_{C_\Sigma}/kT) - 1]^{-1}$. The four terms in (5) account for quasiparticle tunneling from right to left or vice versa in both junctions. The voltage source provides s quanta of energy, similarly as in photon-assisted tunneling. Since the states are superpositions with different Cooper-pair charge, a quasiparticle transition between such states in general is accompanied by a transfer of Cooper pairs as well. The total charge transferred (averaged over both junctions) is $Q_1^{+,s} = Q_2^{+,s} = (\frac{1}{2} - s)e$ and $Q_1^{-,s} = Q_2^{-,s} = (-\frac{1}{2} - s)e$. The ac supercurrent flow averages to zero since no mechanism correlates its phase with quasiparticle tunneling. We account for the step in the quasiparticle conductance from the subgap value to $1/R_n$ by choosing different prefactors in the transition rates $W_{j,i \to f}^{\pm,s}$, depending on whether the energy difference between initial and final state, i.e., the argument of $\text{Th}[x]$, is smaller or larger than 2Δ.

Because of the quasiparticle transitions the time evolution is stochastic and can be described by a master equation for the probability to be in a certain eigenstate

$$\frac{\partial P_n}{\partial t} = \sum_{k \ne m} (P_k W_{k \to n} - P_n W_{n \to k}) . \tag{6}$$

The stationary solution $\partial P_n/\partial t = 0$ is sufficient to calculate the dc current

$$I = \sum_{i,f,s}\sum_{j=1,2\pm} P_i W_{j,i \to f}^{\pm,s} Q_j^{\pm,s} . \tag{7}$$

In the normal state our program reproduces known properties of single-electron tunneling. In the supercon-

ducting state we reproduce the results of Ref. [6] on the "large" current scale $e/R_\Sigma C_\Sigma$ (where $R_\Sigma = R_{n1} + R_{n2}$). But the quantum description outlined above contains much more information which shows up on a smaller scale. In Figs. 2 and 3 we compare measured and calcu-
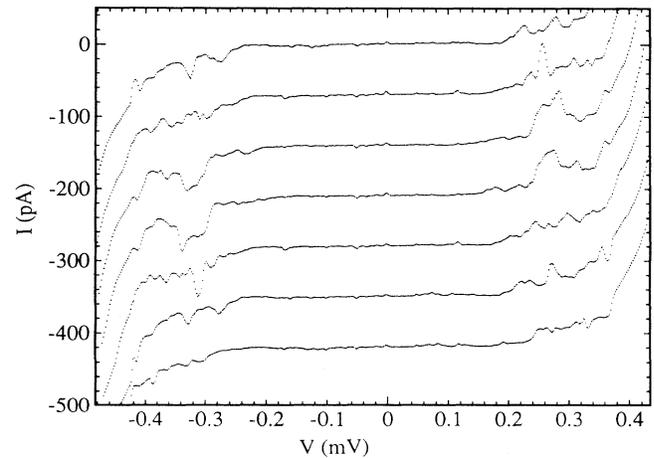

FIG. 2. The measured I-V characteristics of the circuit shown in Fig. 1. The different curves correspond to gate voltages UC_0/e between 0 and $\frac{1}{2}$. The parameters are $\Delta = 0.2$ meV $= 2.2$ K, $C_1 \approx C_2 \approx 0.47fF$ (i.e., $E_{C_\Sigma} = 1$ K), $R_1 \approx R_2 \approx 41$ kΩ ($E_{J1} \approx E_{J2} \approx 0.18$ K), $T = 10$ mK.

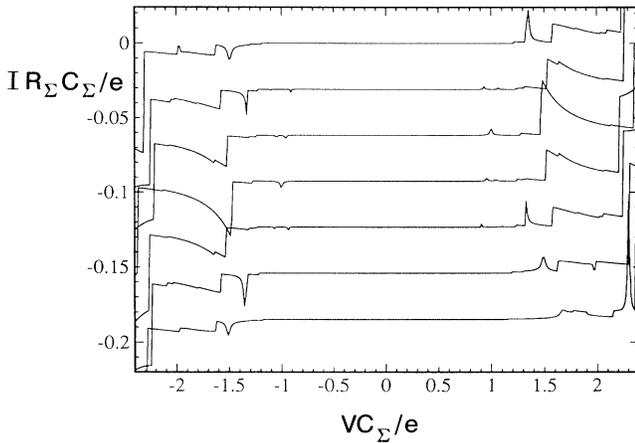FIG. 3. Calculated I-V characteristics for the same parameters as in Fig. 2.

FIG. 4. I-V characteristics for different values of q_0. The parameters are the same as in Fig. 2. The current is clipped at $IR_\Sigma C_\Sigma/e = 0.05$.

lated I-V characteristics for various values of the gate voltage U. Both agree qualitatively and, as far as the scales are concerned, quantitatively well. The parameters have been estimated from independent measurements in the normal state. For the quasicharge q_0, which depends on U and V and possibly trapped charges, we had to choose a reference value, and we allowed for a 10% asymmetry in the capacitances. The subgap conductance in the experiment is so small that in the calculation we set it to zero; i.e., no transitions take place if the energy differences involved are less than 2Δ. Nevertheless, a current flows a subgap voltages.

Because of the dependence of the I-V characteristics to q_0 we present them in Fig. 4 as a 3D plot. Here we only show results on the small current scale and voltages below the gap. The results of Fig. 2 represent cuts through this landscape. We notice a wealth of peaks and ridges in the I-V characteristic, which arise due to *cycles* composed of quasiparticle and Cooper-pair tunneling processes between different states. If each of these transitions is fast the resulting current is large. For the sake of the discussion we treat the Josephson coupling as a perturbation. Then Fig. 5(a) is helpful. Solid lines indicate where, depending on the island charge Q and the bias voltage v, first-order Cooper-pair tunneling processes are on resonance, broken lines indicate this for second-order processes. Lines with positive (negative) slope refer to tunneling in junction 1 (2). The horizontal broken lines at $Q = \pm 2e$ belong to processes where two Cooper pairs enter or leave the central island through different junctions. The grey area, bounded by $|Q/e| + v/2 = \Delta/E_{C_\Sigma} + \frac{1}{2}$, indicates where a quasiparticle transition can occur with the normal-state conductance. The dashed area, bounded by $|Q/e| + 3v/2 = \Delta/E_{C_\Sigma} + \frac{1}{2}$, shows where—in addition to the grey area—a process with a quasiparticle tunneling in one junction and a Cooper pair in the other [see Fig. 5(b)] can occur with the normal-state conductance. In such a $3e$ process, the island charge is changed by $\pm e$,
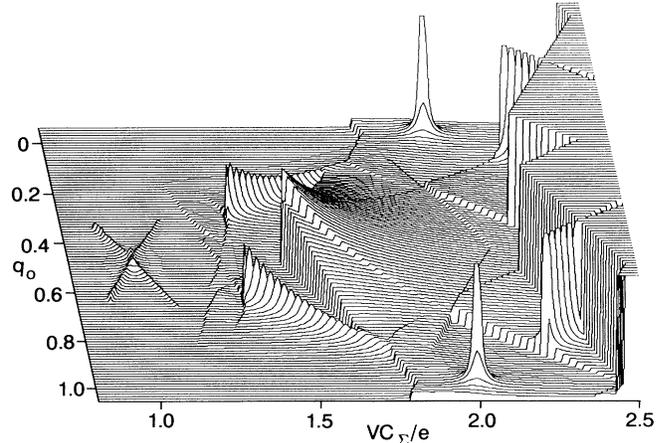
but $3eV/2$ source energy is being put into the system.

For illustration we discuss two examples: (i) The sharp peak at $q_0 = 0$, $v = 2$ arises since at this bias voltage and quasicharge both junctions are on resonance for Cooper-pair transitions (first- or second-order transitions at $Q = 0$ or $Q = \pm e$, respectively). None of the low-charge states are metastable. The higher charge states which can be reached by these Cooper-pair transitions decay freely by quasiparticle tunneling, which leads to current producing cycles. For higher bias voltages, further mechanisms can prevent trapping in a metastable state. Then the current is already enhanced if only one second-order transition is on resonance, leading to "ridges" which appear in the right-hand side of Fig. 4. (ii) Next we consider the shoulder at $q_0 = 0.5$, $v \approx 1.5$ (i.e., $eV = 4\Delta/3$). The rise of the current occurs where both the $Q = e/2$ and the $Q = -e/2$
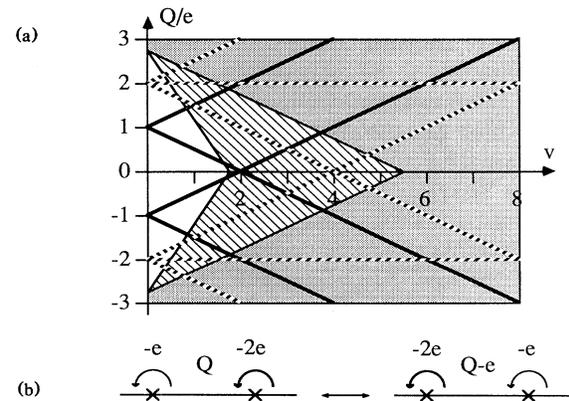

FIG. 5. (a) A map of the states, characterized by the island charge Q and the voltage $v \equiv VC_\Sigma/e$, where Cooper-pair tunneling is on resonance where quasiparticles can tunnel. (b) An illustration of the $3e$ process discussed.

state can undergo the $3e$ transition shown in Fig. 5(b). Such a transition is an off-resonance phenomenon, giving rise to a shoulder rather than a peak. The probability for this process increases when Cooper-pair mixing across the junctions is stronger, i.e., closer to the resonance lines in Fig. 5(a). This explains why the current decreases with increasing bias voltage. The fine structure on the hill can be traced to the onset of another $3e$ process in which both the Cooper pair and the quasiparticle tunnel in the same junction, thus leading to a $\pm 3e$ change of the island charge. These processes involve higher, less populated charge states. Hence they are less pronounced.

Some further comments can be made: (i) Also at $T=0$ and for zero quasiparticle conductance there is no fundamental threshold voltage below which no current is transported. Processes involving $5e$, $7e$, etc. provide higher multiples of $eV/2$ source energy to excite a quasiparticle. However, these processes are of higher order in E_J/E_C. (ii) The observation of these novel processes is possible only if the subgap conductance is low and conventional processes at low bias voltage are suppressed. (iii) The experiments of Ref. [7] show further structures which we cannot interpret within the present model. Some of them may arise due to the coupling to the nonideal environment. On the other hand, some of the sharp current peaks predicted here can be missed or can be smeared out due to effects not contained in our model. (iv) Voltage-biased double junctions have also been discussed in Ref. [9] in an approach which differs from ours. There much less structure in the I-V characteristic is produced, in part because the step structure in the quasiparticle conductance is not included.

In summary, we can say that our analysis describes several novel structures in the I-V characteristic of voltage-biased circuits of Josephson junctions at low voltages and currents, some of which have been observed in experiments. We account for the applied voltage by an explicit time dependence in the Hamiltonian, which leads us to time-Bloch functions. An alternative approach, without time dependence but more degrees of freedom, has recently been formulated in Ref. [10]. There we also discuss the effect of an environmental impedance. Future work should include time-dependent gate voltages and extend the analysis to larger systems.

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[1] D. V. Averin and K. K. Likharev, in *Mesoscopic Phenomena in Solids*, edited by B. L. Altshuler *et al.* (North-Holland, Amsterdam, 1991), Chap. 6.

[2] G. Schön and A. D. Zaikin, Phys. Rep. **198**, 237 (1990).

[3] J. M. Martinis and R. L. Kautz, Phys. Rev. Lett. **63**, 1507 (1989); A.N. Cleland, J. M. Schmidt, and J. Clarke, Phys. Rev. Lett. **64**, 1565 (1990).

[4] L. J. Geerligs *et al.*, Phys. Rev. Lett. **63**, 326 (1989); P. Delsing *et al.*, *ibid.* **63**, 1180 (1989).

[5] K. K. Likharev and A. B. Zorin, Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 1407 (1987).

[6] T. A. Fulton *et al.*, Phys. Rev. Lett. **63**, 1307 (1989).

[7] L. J. Geerligs *et al.*, Phys. Rev. Lett. **65**, 377 (1990); L. J. Geerligs, thesis, 1990 (unpublished).

[8] F. Guinea and G. Schön, J. Low Temp. Phys. **69**, 219 (1987).

[9] V. Ya. Aleshkin and D. V. Averin, Physica (Amsterdam) **165 & 166B**, 949 (1990).

[10] A. Maassen van den Brink *et al.*, Z. Phys. B (to be published).