Correlated discrete transfer of single electrons in ultrasmall tunnel junctions

by K. K. Likharev

Recent theoretical and experimental studies have revealed a new family of effects taking place in very small tunnel junctions at low temperatures. The effects have a common origin, the correlated discrete tunneling of single electrons and/or Cooper pairs resulting from their electrostatic ("Coulomb") interaction. This paper presents a brief review of the single-electron part of the family, including discussion of the background physics, methods of theoretical description of the new effects, experimental results, and possible applications of the new effects in analog and digital electronics.

1. Introduction

Soon after our meeting at the Tunneling at Low Temperatures Conference in Leuven, Belgium (August 1985), where the basic concepts of what we now call "Single-Electronics" had been reported for the first time, Rolf Landauer sent me a copy of his paper [1] published in 1962, with the following note: "Written at a time when I did not yet understand the role of $e^2/2C$. Nevertheless, not really wrong."

Not only was this paper quite correct for its purpose, it was also one of the cornerstones of our present-day understanding of the dynamics of and fluctuations in tunnel junctions. In what follows we show that the equations of Reference [1] can be used as a basis for analysis of the correlated single-electron tunneling, provided that minor changes reflecting the electrical charging effects are made. This modification was absolutely unimportant for the relatively large tunnel junctions available in the sixties, and can be neglected even for most devices studied nowadays. However, for extremely small (submicron) junctions cooled to very low (helium) temperatures, the modification becomes necessary and reveals a new physical picture of electron tunneling.

Some roots of this picture can be traced to the same year, 1962, in which Landauer's paper appeared. Experimenting with metallic granular thin films, Neugebauer and Webb [2] found that the dc conductance of the films was suppressed very substantially at low temperatures. They identified the suppression mechanism as electric charging of the grains by discrete electrons tunneling from the neighboring grains through the separating energy barriers, a concept we now discuss in more detail.

It is well known that electron transfer through a thin energy barrier is a discrete event leading to a change of the charge of the junction electrodes by $\pm e$. In theory, this discreteness is expressed by the celebrated tunnel Hamiltonian [3]

$$H_T = H_+ + H_-, \qquad H_+ = \sum_{k_1,k_2} T_{k_1k_2} c^{\dagger}_{k_2} c_{k_1}, \qquad H_- = H^{\dagger}_{+}, \tag{1}$$

presenting a sum of the independent contributions from transfers of single electrons from states (k_1) inside one electrode to states (k_2) inside the other one. In background electronics, the most recognized consequence of the discreteness is the shot noise arising in tunnel junctions at bias voltages V exceeding $k_B T/e$. In fact, according to the Schottky formula

$$\mathcal{S}_I(\omega) = \frac{1}{2\pi} eI, \tag{2}$$

the fluctuations of the current I are proportional to the charge unit e transferred during a single tunneling event.

In large-area junctions the discreteness hardly leads to any other effects. In a junction with a very small area $\mathcal{S}$ and hence a very small capacitance C between the electrodes, the electrostatic ("Coulomb") energy $E_Q = e^2/2C$ associated with its recharging by a single electron can become comparable to the scale $k_B T$ of the masking thermal fluctuations. For example, for a typical granular thin film ($d \sim 30$ nm) the grain capacitance C is less than 10^{-17} F, so that the relation $E_Q \gg k_B T$ can be satisfied at helium temperatures. In this situation, transfer of a single electron from one grain to another with a smaller value of C becomes highly improbable, at least at low-driving electric fields.

Despite experimental work with granular thin films (see, e.g., the recent Reference [4] and references therein), a quantitative description of the "Coulomb blockade of tunneling" in these structures apparently has not yet been developed because of their random structure. A simpler picture of the effect can be presented for other granular structures first studied by Zeller and Giaever [5] (see also experiments [6, 7]). The structure presents a set of separated metallic grains embedded into a tunnel barrier between two metallic electrodes. Here the Coulomb blockade leads to suppression of the tunnel current at voltages below the threshold value $V_t \sim e/C$ (see Section 4). For these structures, a quite complete physical understanding of the discrete tunneling was achieved [5] and a quantitative theory was developed [8].

The authors of these reknowned works did not, however, pay attention to the remarkable possibilities arising in systems where the *discrete* transfer of charge through energy barriers coexists with its *(quasi)continuous* transfer along the usual metallic conductors. Though the first notice of this possibility appeared only recently (1985) [9], it has already resulted in the rapid development of theory for the new effects [10–16], several suggestions concerning their practical applications [10, 15, 17], and, very recently, their first experimental observations [18–20].

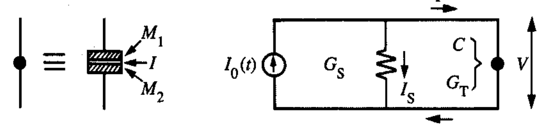

Figure 1

Notation for a small tunnel junction presented in this paper (left) and an equivalent circuit of the current-biased junction (right).

Because of this rapid progress, the only survey available (included as a supplementary chapter 16 to the monograph [21]) is already far from being complete. The present paper is an attempt to give a brief but up-to-date review of the field.

2. Single junction: Theoretical background

We demonstrate methods of description of coherent single-electron tunneling using the simplest example: a single current-biased tunnel junction. Taking into account a possible nonvanishing conductance of the current source (for the sake of generality of the treatment), one arrives at the circuit shown in **Figure 1**. Its analysis can be started with the following Hamiltonian:

$$H = \mathcal{E}(Q) + H_T + H_1\{k_1\} + H_2\{k_2\} + H_S\{k_S\} - I\phi, \tag{3}$$

where H_T is expressed by Equation (1), while

$$\mathcal{E}(Q) = \frac{Q^2}{2C}, \qquad \phi = \int V dt, \qquad V = Q/C,$$

$$I = I_0(t) - I_S\{k_S\}. \tag{4}$$

The operator Q of the electric charge of the junction can be expressed via the same creation and annihilation operators as H_T:

$$Q = -\frac{e}{2}\left(\sum_{k_1} c^{\dagger}_{k_1} c_{k_1} - \sum_{k_2} c^{\dagger}_{k_2} c_{k_2}\right) + \text{const.}, \tag{5}$$

so that H_T and Q do not commute. One can readily prove that the following commutation relations are valid for an arbitrary function $F(Q)$:

$$H_\pm F(Q) = F(Q \pm e) H_\pm. \tag{6}$$

The Hamiltonians H_1, H_2, and H_S describe the energy of the internal degrees of freedom $\{k_1\}$, $\{k_2\}$, and $\{k_S\}$ of the two electrodes of the junction and of the "shunt" G_S, respectively. The last term in Equation (3) describes the interaction of the junction with the current I (Figure 1).

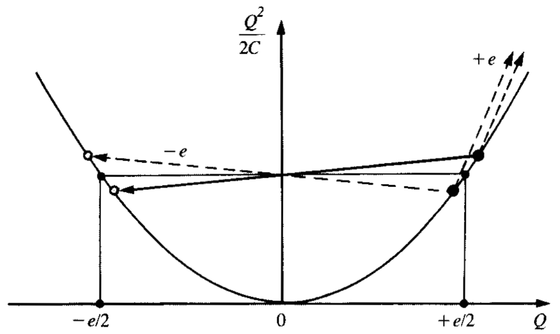

Figure 2

Change of the charging energy $\mathcal{F}$ of a small junction resulting from tunneling of a single electron, for two values of its initial charge Q. Solid-line arrow shows the only energy-advantageous event. From [10], reprinted with permission.

As long as the tunnel barrier transparency and the current I are not too large, they can be considered as perturbations, and one can write an explicit time-evolution equation for the density matrix $p(Q, Q', t)$ traced over the sets $\{k_1\}$, $\{k_2\}$, and $\{k_S\}$—see [10] for details. Now let the following limitation be satisfied as well:

$$G_S, G_T \ll R_Q^{-1}, \qquad R_Q \equiv h/4e^2 \approx 6.45\ \mathrm{k\Omega}. \tag{7}$$

Physically this condition means that the typical energy $\hbar G/C$ ($G \approx \max[G_S, G_T]$) of the quantum fluctuations in our system is much less than the charging energy scale E_Q. In this "classical" limit all off-diagonal elements of the density matrix vanish, and its diagonal elements are proportional to the classical probability density $\sigma(Q, t)$ obeying a very simple "master" equation [9, 10]

$$\frac{\partial\sigma}{\partial t} = -I_0(t)\frac{\partial\sigma}{\partial Q} + F_T + F_S, \tag{8a}$$

$$F_T(Q) = \Gamma^+(Q-e)\sigma(Q-e) + \Gamma^-(Q+e)\sigma(Q+e) - [\Gamma^+(Q) + \Gamma^-(Q)]\sigma(Q), \tag{8b}$$

$$F_S = \frac{G_S}{C}\frac{\partial}{\partial Q}\left(Ck_BT\frac{\partial\sigma}{\partial Q} + \sigma Q\right). \tag{9}$$

The rates $\Gamma^\pm$ of tunneling of a single electron increasing (+) or decreasing (−) the initial charge Q can be expressed as follows:

$$\Gamma^\pm(Q) = \frac{1}{e}I\left(\frac{\Delta\mathcal{F}^\pm}{e}\right)\left[1 - \exp\left(-\frac{\Delta\mathcal{F}^\pm}{k_BT}\right)\right]^{-1}, \tag{10}$$

where $I(V)$ is the dc I–V curve of the same junction biased by a dc voltage V, while $(-\Delta\mathcal{F}^\pm)$ is the change of the free energy due to the tunneling event; in our present case

$$\Delta\mathcal{F}^\pm \equiv \mathcal{F}(Q) - \mathcal{F}(Q \pm e) = \pm\frac{e}{C}(Q \pm e/2). \tag{11}$$

Note that Equations (8) (with $I_0 = F_S = 0$) correspond exactly to the basic equation (3.1) of Landauer's paper [1]! This is not very surprising, because the subject of that paper was the same discrete single-electron tunneling as here. The only new feature, which appeared in 1985, is a small shift ($\pm e/2$) of the arguments in Equation (11) arising from a strict account of the charging effects. We show, however, that this minor difference can lead to some major new results.

3. Single junction: The SET oscillations

Consider the simplest case of an unshunted tunnel junction ($G_S = 0$, $F_S = 0$, $I = I_0$). Let the initial charge Q be localized near some point Q_0 inside the range

$$-\frac{e}{2} < Q < +\frac{e}{2}. \tag{12}$$

Then, if the temperature is low enough,

$$T \ll T_0, \qquad T_0 \equiv \frac{e^2}{2k_BC}, \tag{13}$$

one can readily be convinced that all components of F_T vanish, so that no tunneling happens! This is the "Coulomb blockade of tunneling" [10], already mentioned in the Introduction. In our present simple case, the physical origin of this effect is especially clear (**Figure 2**): Within the range of Equation (12) the tunneling of even a single electron would lead to an increase of the charging energy $Q^2/2C$, and thus, in the low-temperature limit of Equation (13), such an event is virtually impossible.

According to Equation (8a), the narrow probability "packet" describing the initial distribution of Q will move with a small velocity

$$\dot{Q}_0 = I(t) \tag{14}$$

toward an edge of the range (12)—see **Figure 3(a)**. After the edge is reached (say, $Q_0 > e/2$), the rate $\Gamma^-(Q)$ becomes nonvanishing and the last term of Equation (8b) leads to a rapid ($\Delta t \sim \tau_T \equiv C/G_T$) decay of the packet. Simultaneously a new packet grows up at the point $Q_0' = Q_0 - e$, near the left edge but inside the Coulomb blockade range [**Figure 3(b)**]. It is evident that this process describes just the tunneling of a single electron through the junction (see the middle column in Figure 3).

Now, all terms in F_T vanish again, the new probability packet moves through the range (12) according to Equation (14) [**Figure 3(c)**], and then the whole process repeats periodically, with the average frequency

$$f_S = \bar{I}/e. \tag{15}$$

Charge Q and voltage $V = Q/C$ of the junction therefore perform relaxation-type oscillations with the same frequency. The physics of these "single-electron tunneling" (SET) oscillations [9] is very clear from Figure 3: Due to the Coulomb blockade of tunneling, the electric charge supplied by the bias current $I_0(t)$ accumulates on the junction until its threshold value $e/2$ is reached. Then a single electron is transferred; due to a very small capacitance C of the junction, this event results in a considerable change of the junction voltage ($\Delta V = e/C$). In the low-temperature limit [Equation (13)] this change is larger than a swing $k_B T/e$ of the thermal fluctuations, and thus affects essentially possible tunneling of the other electrons (more exactly, reduces drastically the probability of these secondary events). Thus, a time correlation of the tunneling events is established; in the present case the correlation takes the form of the coherent SET oscillations with the frequency (15) [22]. This particular process is reminiscent of water dripping from a leaking tap—see the right-hand column in Figure 3.

The whole picture presented above depends crucially upon whether the electric charge Q measured in units of e can really take fractional values, in particular those less than unity. In order to answer this question, one should remember that Q is essentially the surface charge of the junction electrodes forming the capacitor C. Of course, the tunneling, as a *discrete* process, can change Qe only by integer numbers, as expressed by Equation (10b). However, the current flow I through the usual metallic conductors is a *continuous* process, and thus can change Q by any amount, at least on the scale of e. In order to understand it, let us consider a minor shift Δx of the current carriers inside current leads connected to the junction. This shift results in a proportional increase ΔQ of the surface charge Q. The carrier motion in the usual (long) conductors is virtually not quantized, and hence $Q_e \equiv \int I dt$ is not quantized. (More exactly, quantization of the motion leads to a quantization scale of Q much less than e [23].) Below we see that recent experiments confirm the concept of the possible coexistence of discrete and continuous charge transfer in one system.

The oversimplified picture of the SET oscillations presented above is valid only in the limit of low dc bias current I, low temperatures T [Equation (13)], low conductance G_T [Equation (9)], and vanishing conductance G_S. At the present time, quite a complete understanding of the role of the listed factors has been reached; we give only a brief summary of these results.

Increasing the bias current I beyond $\sim 0.1 e/\tau_T$ leads to a gradual suppression of the amplitude of the SET oscillations (**Figure 4**); at larger currents the only trace of the Coulomb blockade is an offset of linear asymptotes of the dc I–V curve by e/C [9, 10]. Increasing the temperature beyond T_0 [Equation (13)] [13, 14] and G_T beyond R_Q^{-1} [11, 16] gives qualitatively the same effect [24]. Note that the very fact of coherent (monochromatic) oscillations and the fundamental

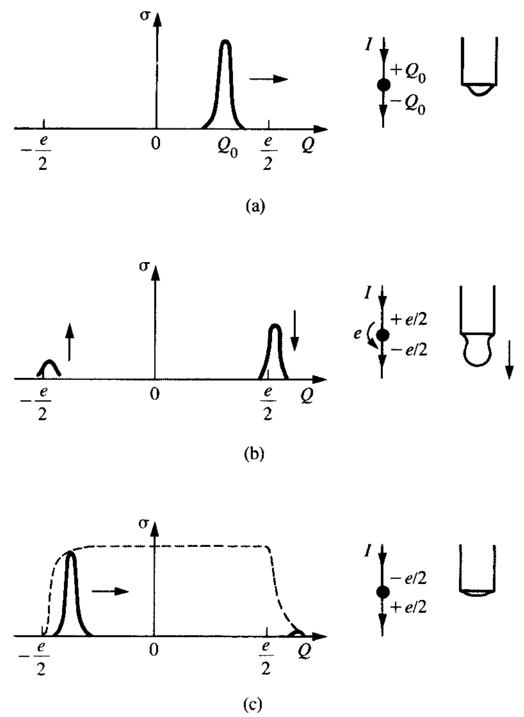

Figure 3

Time evolution of the probability density $\sigma(Q, t)$ in process of the SET oscillation (left column), a scheme of the corresponding single-electron transfer (middle), and its drip analogy (right). From [10], reprinted with permission.

relation (15) are not affected by these factors. On the contrary, a nonvanishing metallic ("shunt") conductance G_S [**Figure 5(a)**] leads to a broadening of the oscillation linewidth [**Figure 5(b)**].

One should also mention a series of works [10–16] analyzing the dynamics of a similar system with a considerable Josephson coupling of the electrodes. In this case, the SET oscillations can coexist with the "Bloch" oscillations [25] of the frequency

$$f_B = \bar{I}/2e. \tag{16}$$

Note, however, that the Bloch oscillations are not just a replica of the SET oscillations, despite their possible similar interpretation as "dipping" of the Cooper pairs [21, 25]. The reason is that in contrast to the single electrons, the Cooper pairs form a coherent superconducting condensate inside each electrode, so that their transfer constitutes a quantum

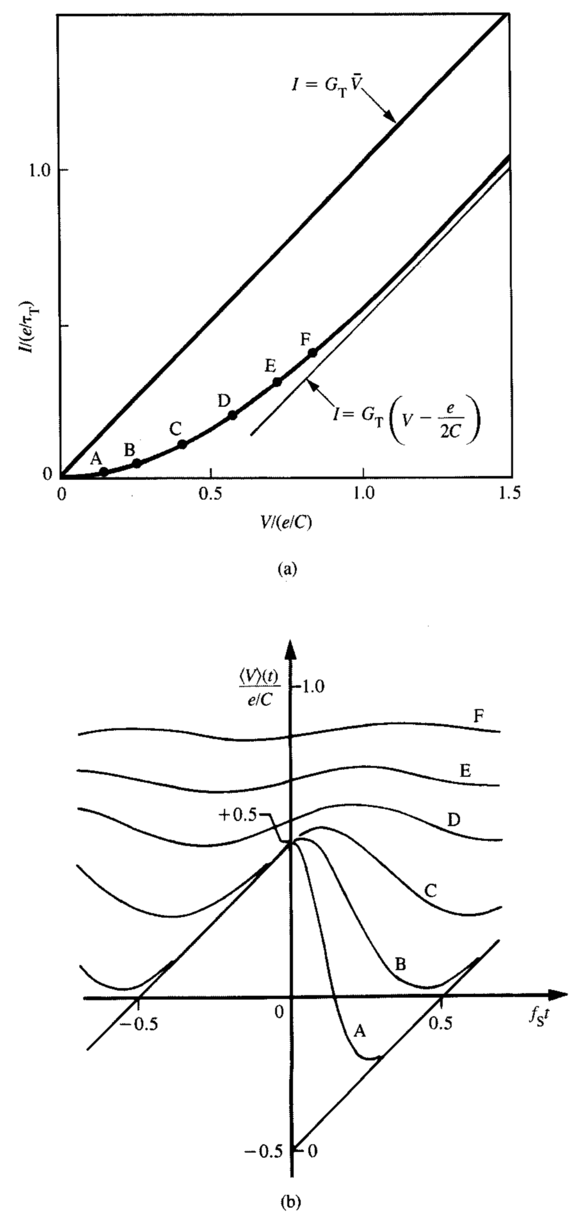

Figure 4

The SET oscillations in an unshunted tunnel junction with normal-metal electrodes in the low-temperature limit: (a) dc I–V curve. (b) Oscillation waveforms for several points of the curve. From [9, 10], reprinted with permission.

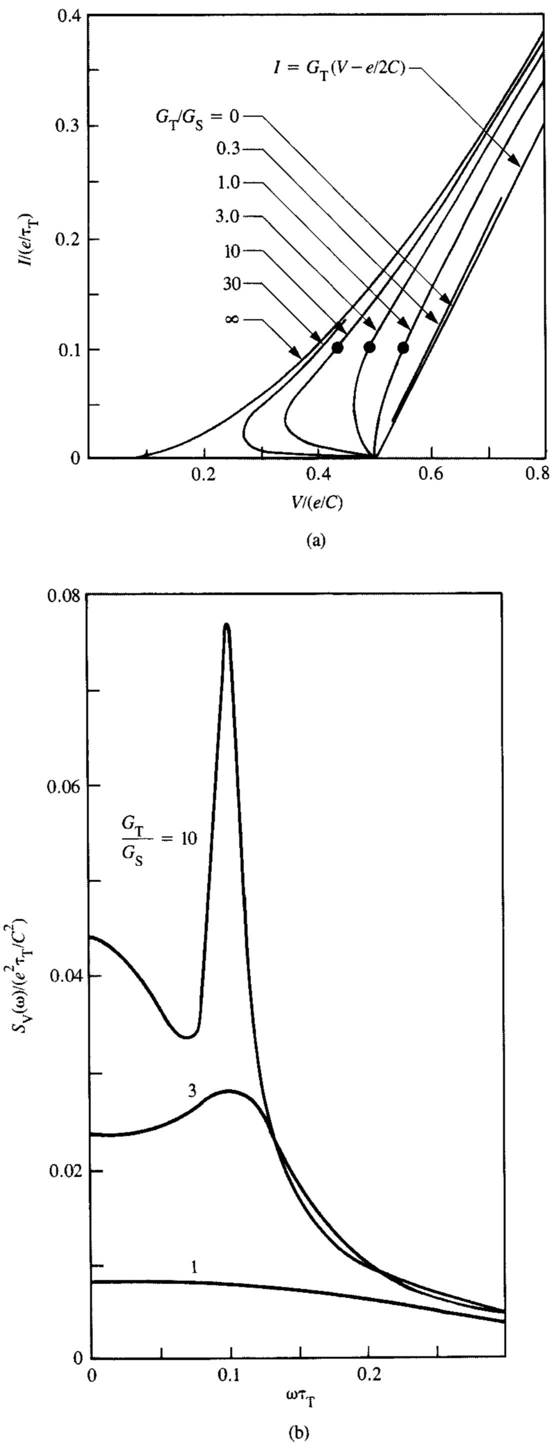

Figure 5

The SET oscillations in an externally shunted junction (normal metals, low temperatures): (a) dc I–V curves for several values of G_S. (b) Frequency spectra of the junction voltage for $I_0 = 0.1e/\tau_T$. From [14], reprinted with permission.

Table 1 Estimates of the main parameters of the single-electron tunneling junctions (after [15, 17]).

Level of fabrication technology	*Junction area* S (nm^2)	*Junction capacitance** C (aF)	*Temperature limit* T_0 (K)	*Voltage scale* $V_t = \frac{e}{2C}$ (mV)	*Current scale*† $I_t = \frac{V_t}{R_T}$ (nA)	*Time scale* $\tau_T = R_T C$ (ps)
State-of-the-art junctions [28]	100 × 100	300	3	0.25	2.5	30
Record junctions [19]	30 × 30	30	30	2.5	25	3
Nanolithography limits [29, 30]	10 × 10	3	300	25	250	0.3

* Estimated as $C_0 S$, where $C_0 = 3 \times 10^{-6}$ F/cm^2, a typical value for the soft metal oxide barriers.
† R_T is accepted to equal 100 kΩ, a value compatible with Equation (7).

process rather than the nearly classical process of the SET oscillations. This difference results in several important peculiarities of the Bloch oscillations [10–16].

To end our survey of theoretical results, we should mention an analysis of the single-electron tunneling in the zero-bias limit [12]. The main result of the analysis is that an arbitrarily small nonvanishing metallic conductance G_S (say, that of a measuring instrument) results in a qualitative change of the tunnel junction statistics and therefore a difference of its impedance and fluctuation properties from those calculated earlier [26, 27] disregarding the conductance.

Turning to the experimental situation, one should note that the main problem in the observation of the SET oscillations is fulfillment of Equation (13). First of all, the junction itself should be very small and/or the temperature should be rather low—see **Table 1**. Another problem is that the capacitance C apparently includes a contribution C_L of the leads attached to the junction; in a common junction geometry this contribution is much larger than the values given in Table 1. A possible way to get rid of C_L is to place a large resistance $R_S \gg R_Q$ fixing the bias current in the very near vicinity of the junction; in this case the capacitance of the more distant leads is not essential [25].

Recently, Büttiker and Landauer made a very interesting conjecture [31]. The theory described above does not take into account details of the electron transfer through the barrier, including the nonvanishing time τ_0 of the tunneling and the time τ_p of the restoration of electroneutrality inside the electrode after this event. For typical junctions available at present, the shortest time scale of our theory, τ_T (see Table 1), is much larger than both τ_0 and τ_p ($\sim 10^{-14}$ s), so that the model described above seems quite adequate for a small junction by itself. However, general principles of relativity require the tunneling event to be independent of environmental details located beyond the "horizon" radius $r \approx c(\tau_0 + \tau_p)$ of order 10^{-4} cm. Büttiker and Landauer supposed that for this reason the farther parts of the leads would not contribute to C_L. If this conjecture is true, C_L is restricted to a value of order 10^{-15} F, quite acceptable for the first experiments with the SET oscillations.

Nevertheless, two reported attempts to observe this effect in structures comprising few junctions gave negative results [19, 32] (no cutoff resistance R_S was used in these experiments). The only claims of possible observations of the SET (or Bloch) oscillations came from two groups experimenting with granular superconducting thin films [33, 34]. Irradiating the films with microwaves, the authors of those works have observed the appearance of the "voltage steps" in the dc I–V curves, separated by equal current intervals $\Delta I \propto f$. Such behavior, with ΔI equal to either ef or $2ef$, really does follow from the theory of the SET and Bloch oscillations—see, e.g., **Figure 6** [10] (like the well-known Josephson–Shapiro current steps, these voltage steps are due to the phase-locking of the SET oscillations by the external microwave signal and its harmonics). However, the constant $n \equiv \Delta I/ef$ in the experiments [33] was as high as $\sim 10^5$. Probably this behavior is due to some complex dynamics of the granular film resulting from the single-electron tunneling. It seems appropriate to remind the reader that relaxation oscillations with the index $n' \equiv V/\phi_0 f$ are routinely generated in complex Josephson-junction structures (see, e.g., [35]).

Somewhat more convincing evidence of the coherent oscillations with frequency expressed by Equation (15) or (16) has come from similar experiments by Yoshihiro and coworkers [34]; here up to 60 voltage steps with $n = 12$ have been observed. The rms deviation of n from the above value, averaged over the ensemble of the steps, was as small as $\sim 1\%$ for several different values of $f \sim 10$ GHz.

Nevertheless, an urgent need for new experiments, preferably with single junctions (including those without the Josephson coupling) is evident.

4. Two junctions: Sub-electron-charge control of the dc current

A more certain confirmation of the basic ideas of the correlated single-electron tunneling was obtained just recently [18–20] from experiments with a slightly more complex system comprising two small tunnel junctions connected in series [**Figure 7(a)**]. Its analysis [5, 8, 15] can be started with the Hamiltonian which is a generalization of

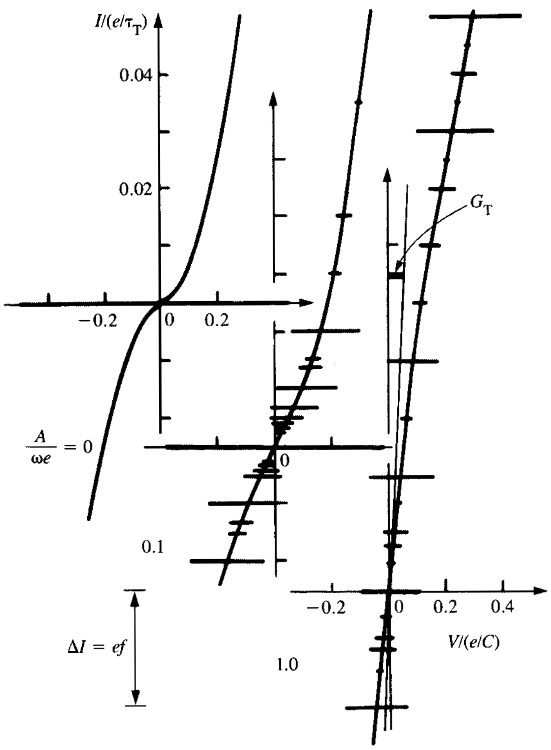

Figure 6

dc I–V curves of the small junction (normal metals, low temperatures, no shunting) irradiated by microwaves of frequency $f = 0.02\tau_T^{-1}$ for several values of the microwave current amplitude A. Note shifts of the origin. From [10], reprinted with permission.

Equation (3),

$$H = \mathcal{F}(n_1, n_2) + H_{T1} + H_{T2} + \sum_{i=1}^{3} H_i\{k_i\}, \tag{17}$$

where the free energy $\mathcal{F}$ can be presented as

$$\mathcal{F}(n_1, n_2) = \frac{Q^2}{2C_\Sigma} - e\left(n_1 \frac{C_2}{C_\Sigma} + n_2 \frac{C_1}{C_\Sigma}\right)V + \text{const.},$$

$$Q \equiv e(n_1 - n_2) + Q_0, \qquad Q_0 \equiv \int I_0 dt,$$

$$C_\Sigma \equiv C_1 + C_2. \tag{18}$$

The tunnel Hamiltonians H_{T1} and H_{T2} can be presented similarly to Equation (1); H_i describes the electrodes, and $n_{1,2}$ are the numbers of electrons already transferred through the junctions [Figure 7(a)].

Carrying out calculations similar to those of Section 2, one arrives again at Equations (8) and (10), with $F_S = 0$ and the following changes:

1. F_T is a sum of contributions describing two junctions.
2. Equation (11) is generalized as follows:

$$\Delta\mathcal{F}_i^{\pm} = \mathcal{F}(n_i, n_j) - \mathcal{F}(n_i \pm 1, n_j) \qquad j = 3 - i, \tag{19}$$

where i is the number of the junction whose tunneling rate is being considered.

Equations (10) and (19) show that in the low-temperature limit (13) (with the replacement $C \rightarrow C_\Sigma$), the tunneling is completely blocked as soon as $\Delta\mathcal{F}_i^{\pm}$ are negative for both junctions. According to Equation (18), the corresponding region has a very simple shape [Figure 7(b)]. One can see that the Coulomb blockade can be lifted by injection of the charge $Q_0 = e(n + ½)$ into the middle electrode.

Let us start with the case $Q_0 = \text{const.}$ (i.e., $I_0 = 0$). In this case a nonvanishing current can flow through both of the

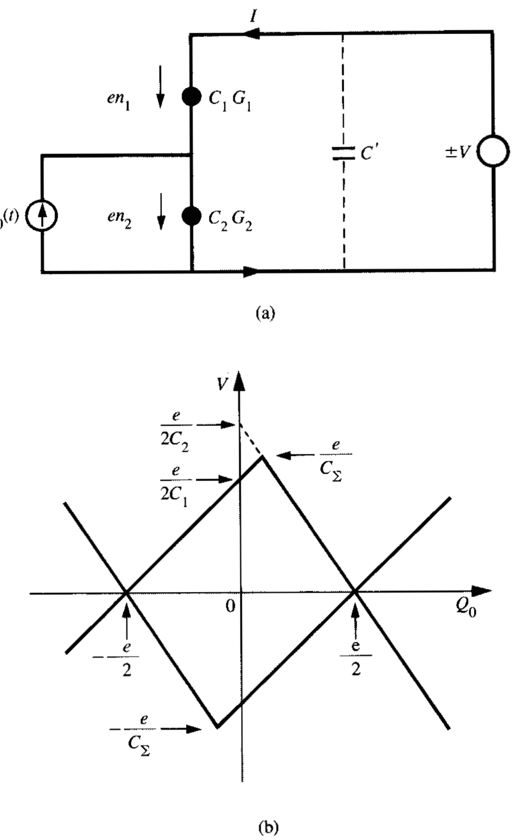

Figure 7

A system allowing control of the dc current I by subelectron changes of the electron charge $Q_0 = \int I_0 dt$: (a) Equivalent circuit. (b) Threshold curves limiting the region of the Coulomb blockade. From [10], reprinted with permission.

junctions outside the Coulomb blockade range, but no coherent SET oscillations take place in the system! The reason is that the system is voltage-biased rather than current-biased, so that a single electron-tunneling event does not change V and does not affect the consequent tunneling events in the same junction. However, tunneling events in two junctions are *mutually* correlated: As soon as an electron enters the middle electrode through one of the junctions, it acquires a large probability rate Γ for leaving the electrode almost immediately through the other junction.

This mutual correlation results in a strong influence of Q_0 on the tunneling dynamics, even outside the Coulomb blockade range. **Figure 8** shows a typical family of the dc I–V curves for several values of Q_0. (As Q_0 increases further, the picture is reproduced e-periodically.) One can see a very characteristic staircase pattern superimposed over the background Coulomb-blockade curve [cf. Figure 4(a)].

Each step of the staircase corresponds to a change of the average charge of the middle electrode by e. Generally the staircase has two periods, $\Delta V_1 = e/C_1$ and $\Delta V_2 = e/C_2$; in the simplest case, $G_i \ll G_j$, only one period, $\Delta V = e/C_i$, is developed.

This behavior was observed clearly in recent experiments. In order to obtain a system equivalent to that shown in Figure 7(a) (with $I_0 = 0$), Kuzmin and Likharev [18] have used a granular system very similar to that studied in the early experiments [5–7]. However, the authors of [18] have contrived to isolate completely all grains inside the barrier except (supposedly, the largest) one, and study this single object rather than the whole ensemble of grains with random parameters. This single grain was connected with the junction electrodes by the tunnel junctions, with $C_1 \approx C_2 \approx 3 \times 10^{-17}$ F and $G_2 \gg G_1 \sim 10^{-6}\ \Omega^{-1}$, so that (at helium temperatures) both conditions (7) and (13) were satisfied. **Figure 9** shows the dc I–V curve of one of the samples together with its first derivative. One can see oscillations with a very constant period ΔV [36]. The phase of these oscillations was stable at helium temperatures but could be shifted to a new value by heating the junction to room temperatures for few minutes **[Figure 10(a)]**. Comparison with calculations using Equations (8), (10), (18), and (19) **[Figure 10(b)]** allows one to interpret these shifts as changes of the parameter Q_0.

Within the simplest models of the tunnel barrier, this parameter (in the absence of I_0) is fixed by capacitances C_1, C_2 and the work functions φ_i of the electrodes [5, 8, 15]:

$$Q_0 = e^{-1}[C_1(\varphi_1 - \varphi_2) - C_2(\varphi_2 - \varphi_3)]. \quad (20)$$

However, it is physically evident that even a small (atomic-scale) shift of a single charged impurity inside one of the tunnel barriers of the structure should lead to a considerable change of Q_0. Hence, the results described above show that Q_0 is perfectly stable at helium temperatures (although the early experiments [6] hint at a

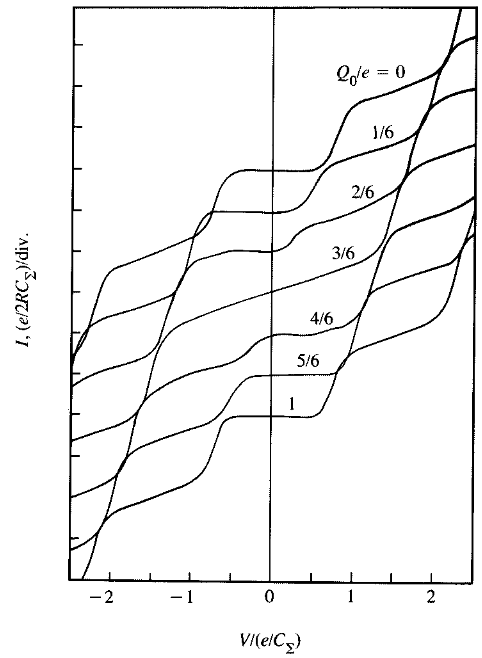

Figure 8

dc I–V curves of the circuit shown in Figure 7(a) for several values of the injected charge ($G_1 << G_2$, $C_1 = 2C_2$, $T = 0.1T_0$, normal-metal electrodes).

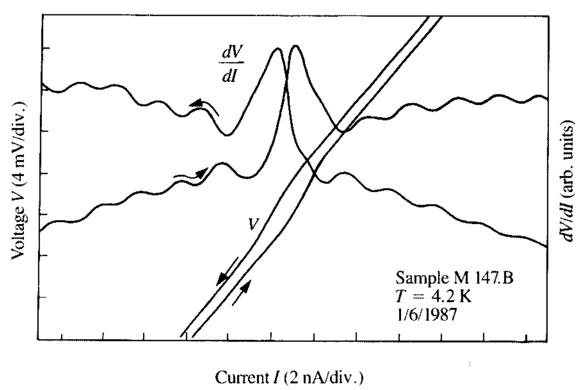

Figure 9

The dc voltage V and differential resistance $R_d \equiv dV/dI$ of a system comprising an In grain embedded between two Pb-alloy electrodes, as functions of the dc bias current I passed between the electrodes. A hysteresis of the I–V curve and general slopes of the $R_d(V)$ dependences are due to some imperfections of the electronics used in the measurements. From [18], reprinted with permission.

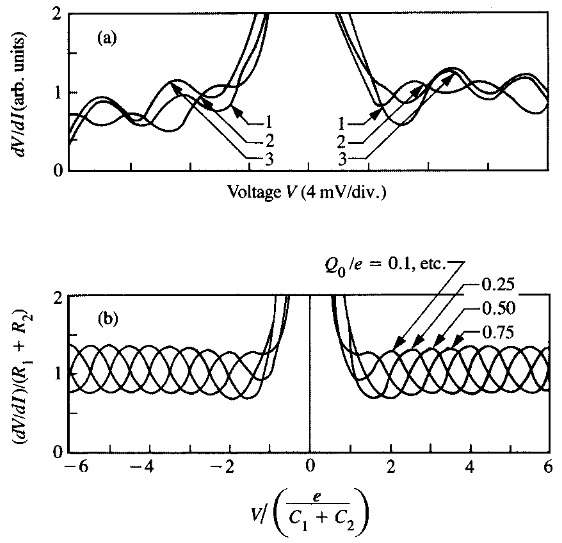

Figure 10

Differential resistance as a function of the dc voltage V: (a) Results of three sequential experiments with the same structure as in Figure 9, specimens allowed to reach 300 K between successive measurements. (b) Results of calculations for the S-I-N-I-S structure with experimental values $T = 4.2$ K, $C_1 = 3.2 \times 10^{-17}$ F, $G_1 << G_2$, $\Delta(T) = 1.2$ meV, the fitted value $C_2/C_1 = 1$, and for several values of Q_0. From [18], reprinted with permission.

possibility of a very slow relaxation of Q_0 even in these conditions), but relaxes rapidly to an equilibrium value at room temperatures. It has been demonstrated [18] that this value can be reproduced with an accuracy better than $\sim 0.1e$.

A possibility for controlling Q_0 at helium temperatures was demonstrated in the experiment by Fulton and Dolan [19], who used a unique thin-film microstructure [**Figure 11(a)**] with tunnel junctions as small as $\sim 10^{-11}$ cm^2. The capacitance C_0 was very small ($\sim 10^{-3}\, C_{1,2}$), but it nevertheless allowed one to change Q_0 by changing the voltage U [in this structure, $Q_0 = C_0U$ + const.; see Equation (28) below]. **Figure 11(b)** shows a family of the experimental dc I–V curves for five successive values of U, shifting Q_0 by increments of $\sim e/6$. Although the system differs slightly from that used to calculate the plots of Figure 8 (in particular, superconductivity of the electrodes provided an additional energy-gap structure in the experiment), the similarity of these results is apparent.

To end our discussion of the system shown in Figure 7(a), we should note that a nonvanishing current I_0 should induce there the SET oscillations with the frequency $f_S = \bar{I}_0/e$. If a sufficient Josephson coupling of the electrodes is provided, the SET/Bloch oscillations coexist with the usual Josephson oscillations, with the frequency $f_J = (2e/h)\bar{V}$. Mutual phase-locking of these oscillations leads to quantization of the ratio $\bar{V}/I_0$ in units of R_Q, similar to that arising at the quantized Hall effect. These predictions [37] are still to be confirmed experimentally.

5. Multijunction structures

Analysis of more complex systems of small tunnel junctions is aided by the fact that Equation (19) admits a ready generalization. It can be proved using the simple quantum-mechanical Golden Rule approach [21]. [This approach is, of course, less strict than the direct quantum-statistical methods used for the simpler systems; in particular, the limitation (7) could be hardly obtained in this way. Within this limit, however, both of the methods yield similar results.]

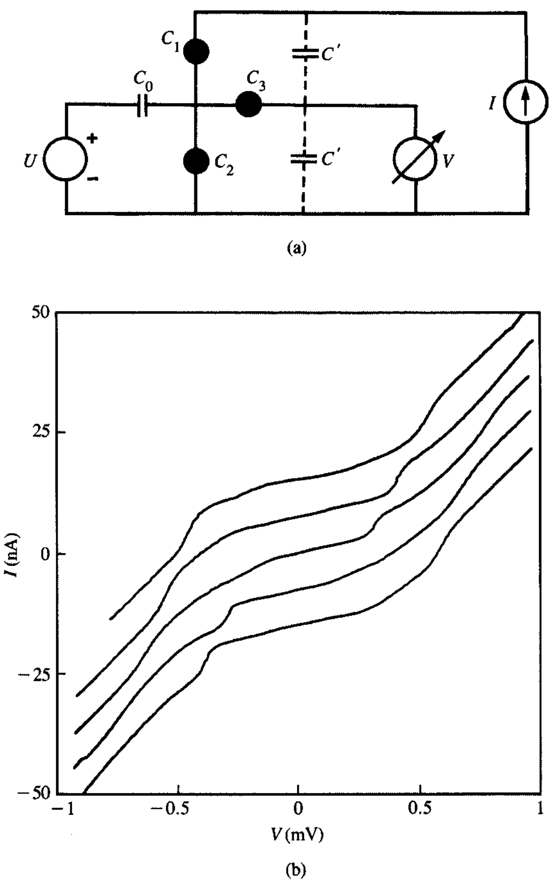

Figure 11

The experiment by Fulton and Dolan: (a) Equivalent circuit of their tunnel structure. (b) dc I–V curves for five sequential values of U, changing Q_0 by $\sim e/6$ each time. From [19], reprinted with permission.

Consider an arbitrary structure comprising N small junctions with the free energy $\mathcal{F}(n_1, \cdots, n_k, \cdots, n_N)$ where n_k is the number of electrons passed through the kth junction. According to the Golden Rule, the rate $\Gamma_k^{\pm}$ of the event $n_k \rightarrow n_k \pm 1$ can be written as

$$\Gamma_k^{\pm} = \frac{2\pi}{\hbar} \sum_{\mathrm{i,f}} |H_{\mathrm{T}_k}|^2_{\mathrm{i,f}} f(E_\mathrm{i})[1 - f(E_\mathrm{f})]$$

$$\times\, \delta\{[\mathcal{F}(n_1, \cdots, n_k, \cdots, n_N) + E_\mathrm{i}]$$

$$- [\mathcal{F}(n_1, \cdots, n_k \pm 1, \cdots, n_N) + E_\mathrm{f}]\}, \quad (21)$$

where E is the energy of the internal degrees of freedom of the system, indices i and f denote initial and final states of the degrees, and $f(E)$ is the Fermi function. But according to the standard theory of tunneling (see, e.g., [3]), a current I flowing through the same junction biased by a dc voltage V can be expressed in a similar way:

$$I(V) = I^{+}(V) - I^{-}(V),$$

$$I^{\pm}(V) = e\frac{2\pi}{\hbar} \sum_{\mathrm{i,f}} |H_{\mathrm{T}_k}|^2_{\mathrm{i,f}} f(E_\mathrm{i})$$

$$\times\, [1 - f(E_\mathrm{f})]\delta\{E_\mathrm{i} - (\mp eV + E_\mathrm{f})\}. \quad (22)$$

Combining Equations (21) and (22), one arrives at Equation (10), with

$$\Delta\mathcal{F}_k^{\pm} = \mathcal{F}(n_1, \cdots, n_k, \cdots, n_N)$$

$$- \mathcal{F}(n_1, \cdots, n_k \pm 1, \cdots, n_N). \quad (23)$$

The energy $\mathcal{F}$ can be readily calculated for even relatively complex structures because for this purpose all the junctions can be replaced by just their capacitances C_k.

On the other hand, *synthesis* of scientifically interesting and/or practically promising structures is facilitated by a deep (although incomplete) analogy between the coherent single-electron tunneling in small junctions and the macroscopic quantum effects in "large" superconducting junctions. More exactly, the two groups of the effects are related by the following electromagnetic duality transformations [15, 21]:

$$Q \leftrightarrow \phi \qquad \text{(in particular, } e \leftrightarrow \phi_0\text{)},$$

$$V \leftrightarrow I,$$

$$C \leftrightarrow L,$$

$$R \leftrightarrow G \qquad \text{(in particular, } R_\mathrm{Q} \leftrightarrow R_\mathrm{Q}^{-1}\text{)}, \quad (24)$$

series connection $\leftrightarrow$ parallel connection.

For example, these transformations relate the SET oscillations to the Josephson oscillations, and the system analyzed in Section 4 [Figure 7(a)] to the dc SQUID. The reader has a chance to see that these analogies are really prominent.

As an example of application of the duality [Equation (24)] to multijunction systems, **Figure 12** shows the

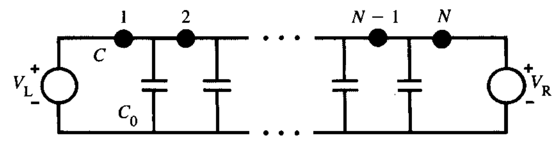

Figure 12

A single-electron analog of a long Josephson junction.

"Coulomb" analog of the long Josephson junction (more exactly, of its discrete version—see Chapter 8 of [21]). Listed below are the basic properties of this array, qualitatively similar to those of its Josephson-junction analog.

In a long array, single-electron "solitons" and "antisolitons" with the electrical charges $\pm e$ and the "size" (expressed in number of junctions)

$$n = 2\left[\operatorname{arccosh}\left(1 + \frac{C_0}{2C}\right)\right]^{-1} \quad (25)$$

can exist. In order to insert a train of the solitons into the junction from one of its edges, the corresponding driving voltage (either V_L or V_R) should exceed the threshold value

$$V_\mathrm{t} = \frac{e}{2C_0}\left[1 - \left(1 + \frac{C_0}{2C} + \left\{\frac{C_0^2}{4C^2} + \frac{C_0}{C}\right\}^{1/2}\right)^{-1}\right]. \quad (26)$$

If $V_\mathrm{R} = V_\mathrm{L} \equiv V$, this insertion leads to a static soliton array which can be either commensurate or incommensurate with the junction array. The site-filling factor as a function of V shows a typical devil's-staircase pattern very similar to that observed in other systems (see, e.g., [38]). At $V_\mathrm{L} \neq V_\mathrm{R}$ the arising longitudinal electric field can induce a viscous flow of the soliton array along the junction array, qualitatively similar to the quantized flux flow in overdamped long Josephson junctions and type-II superconductors.

Unfortunately, because of lack of space we must leave a detailed discussion of this and some other interesting systems for further publications.

6. Possible applications

Virtually all suggested applications of the coherent single-electron transfer require the very low probability p of undesired tunneling events during the time τ of operation. According to Equations (10) and (11) this probability can be roughly estimated as

$$p \sim \frac{\tau}{\tau_\mathrm{T}} \exp\left\{-\frac{E_\mathrm{Q}}{k_\mathrm{B}T}\right\} = \frac{\tau}{\tau_\mathrm{T}} \exp\left\{-\frac{T_0}{T}\right\}. \quad (27)$$

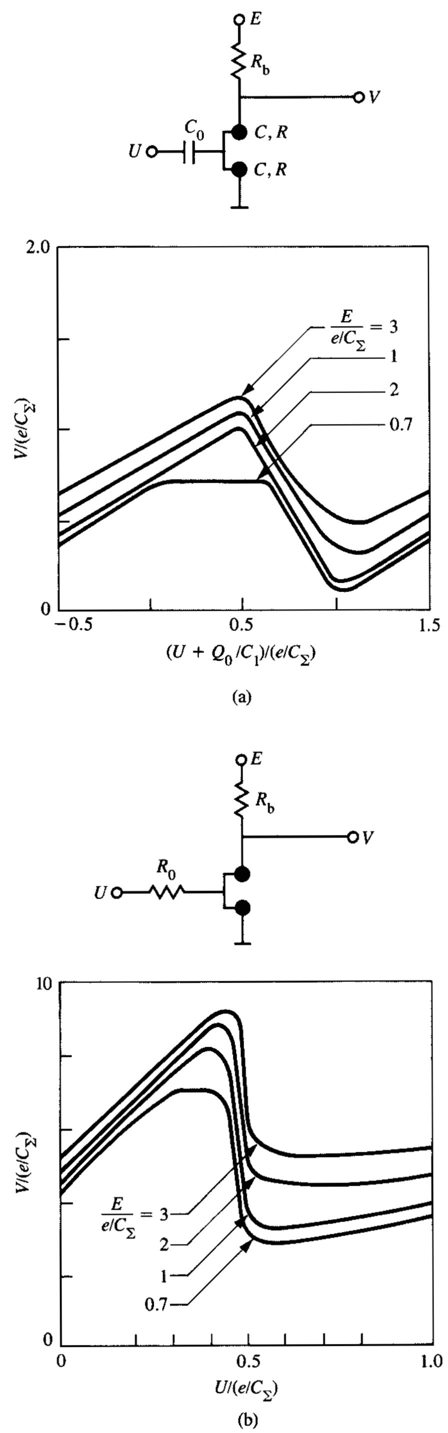

Figure 13

Two types of single-electron transistors: (a) Capacitive SET; (b) resistive SET. Shown are invertor/amplifier stages and control characteristics of the stages for several values of the supply voltage E. The other parameters are $C_0 = 2C$, $R_b = R_0 = 10R$, $T = 10^{-2}T_0$. From [15], reprinted with permission.

For typical values $p \sim 10^{-30}$, $\tau \sim 10^5$ s, and $\tau_T \sim 10^{-12}$ s, one obtains the following estimate of the upper operation temperature: $T \approx T_0/100$. Table 1 shows that even using very advanced lithography one cannot hope to raise T well above the helium level. This is why it is reasonable to discuss only the applications promising some unique capabilities.

Quantum metrology Voltage steps arising in the dc I–V curve of a microwave-irradiated small tunnel junction (Figure 6) can be used for design of a fundamental standard of the dc current [9, 10, 25] quite similar in structure to the dc voltage standard based on the Josephson effect. This direct way can be easier than an alternative one using a combination of the Josephson effect and the quantized Hall effect. Moreover, if both ways were realized in one laboratory, a "quantum metrology triangle" [25] could be closed, enabling one to find possible solid-state or quantum-electrodynamical corrections to the corresponding fundamental relations among frequency, current, and voltage.

Supersensitive electrometry The two-junction device described in Section 4 [Figure 7(a)] can be used to measure extremely small (subelectron) variations of the "external" electric charge Q_0 injected into its middle electrode. In the first experiments [18] a charge resolution better than at least 3×10^{-2} $e/\text{Hz}^{1/2}$ has been registered; presumably the resolution in the experiments of [19] was even better [Figure 11(b)] although no estimate has been given by the authors. Calculations [15] (neglecting the $1/f$ noise contribution) predict the resolution limit of order 10^{-6} $e/\text{Hz}^{1/2}$ for a junction area $\mathcal{S} \approx 3 \times 10^{-12}$ cm^2. In any case, the resolution can be much better than that of the commercially available electrometers ($\gtrsim 10^2$ $e/\text{Hz}^{1/2}$; see, e.g., [39]).

A foreseeable practical problem here is an extremely small input conductance ($C_i \sim C_\Sigma = C_1 + C_2$) of the device limited by the operation temperature—see Equation (13). A similar problem in their magnetic analogs (the SQUIDs) was solved by using the superconducting dc transformers, which apparently have no analog in electrostatics [this is one of the manifestations of incompleteness of the analogy expressed by Equation (24)]. If this problem can be solved or avoided in some way, the supersensitive electrometer could become a new unique tool in science and technology.

Digital microelectronics When reproducible fabrication of large arrays of small tunnel junctions becomes a reality, coherent single-electron tunneling could enable one to design at least two new types of digital VLSI circuits.

The circuitry of the first type could employ "single-electron transistors" (SETs) [10, 15, 17] based on the two-junction system studied in Section 4. **Figure 13** shows two possible structures of this type, the "capacitive SET" (a) and the "resistive SET" (b), used in the simplest amplifier/invertor stages. In order to describe the dynamics of the C-SET, the replacements [15]

$$C_2 \to C_2 + C_0, \qquad C_\Sigma \to C_\Sigma + C_0,$$
$$Q_0 \to Q_0 + C_0 U \tag{28}$$

should be made in all formulas of Section 4. The R-SET can be analyzed using the original equations of Section 4 supplemented by the additional relations [15]

$$\dot{Q}_0 = (U - \langle V_2 \rangle)/R_0, \qquad \langle V_2 \rangle = (\langle Q \rangle + C_1 V)/C_\Sigma, \tag{29}$$

where $\langle V_2 \rangle$ is the ensemble average of the voltage across junction 2.

Figure 13 shows the typical results of such analysis of the SETs, their control characteristics. One can see that the voltage gain $K_V \equiv |\partial V/\partial U|$ can be larger than unity in both circuits. In the C-SET, K_V cannot be larger than C_0/C_1, while in the R-SET the gain can be quite large. Another advantage of the R-SET is its well-defined threshold $U_t = e/2C_\Sigma$, while that of the C-SET includes the parameter Q_0, which is dependent on the relaxation inside the barrier (Section 4). On the other hand, an evident advantage of the C-SET is its small input admittance, which vanishes at low frequencies, while in the R-SET this parameter is close to $R_0^{-1} \neq 0$. A preliminary analysis shows, nevertheless, that both types of single-electron transistor can be used to compose logic and memory circuits very similar in design to those produced using the semiconductor FETs.

The second type of digital circuitry can use single electrons, Coulomb-blocked inside small metallic electrodes, to represent digital bits. The electrons can be passed from one electrode to the other by their discrete transfer through small tunnel junctions. Such a transfer induces a short pulse of current with the area

$$\int I dt \lesssim e \approx 0.16 \ \mu\text{A} \times \text{ps} \tag{30}$$

in the electrodes; the pulses can be used for processing the information. For example, **Figure 14(a)** shows the simplest stage capable of reproduction/regeneration of the pulses. A one-dimensional array of such stages forms a neuristor-type line which can provide transfer of a single-electron soliton (presenting a single bit) with a constant velocity.

Design of more complex logic/memory circuits of this "single-electron logic" family [17] is facilitated by the fact that the duality transformations (24) relate these circuits to those of the recently developed [40, 41] and tested [42] RSFQ logic family based on the Josephson effect. **Figure 14(b)** shows a simple logic/memory stage using the RSFQ ideas. In the absence of input pulses (A, B, T), the information is stored in the form of an extra charge $Q = \pm e/2$ of its middle electrode. The information can be read out by the clock pulse T [of the standard form (30)], which also restores the cell into its "0" state ($Q \approx -e/2$). The

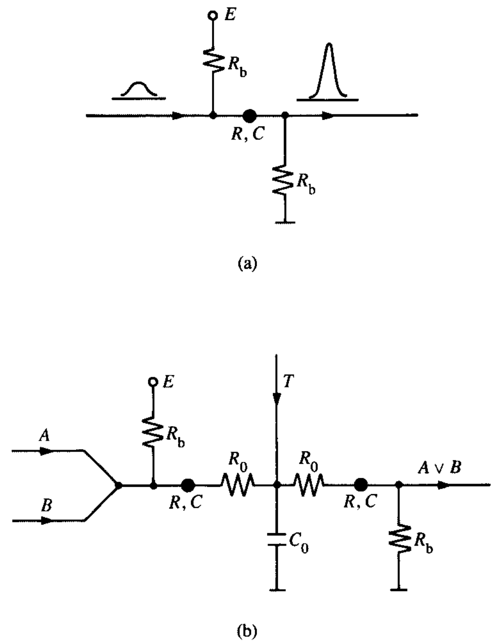

Figure 14

The simplest logic circuits of the single-electron logic family: (a) Buffer/amplifier; (b) timed-OR gate. From [17], reprinted with permission.

first of the pulses (A, B) induces transfer of a single electron through the left junction, changing the state of the cell to "1." The second pulse (if it comes) produces no effect, so that the circuit performs the timed-OR function. Other logic functions can also be readily performed [47].

General discussion Although the applied field of "Single-Electronics" [21] is still in a very early state of development, some general features of this new generation of the electronic circuits are already clear [15]. Their obvious drawbacks include the following:

- The need for advanced lithography, with a feature size of the order of 30 nm or less (see Table 1).
- High impedance ($|Z| \sim R_T \gg R_Q \approx 6$ kΩ), much larger than that of the present-day superconducting microstrip lines (these lines are seemingly the only way to carry picosecond pulses with small attenuation and distortion). This matching problem can be at least partly solved by using the increase of the kinetic inductance of the lines, which decreases in proportion to their cross-sectional area.

A list of advantages of the single-electronic devices is nevertheless impressive:

- Major parameters of the devices are not dependent on the junction resistance, i.e., on transparency of the tunnel barriers. This feature promises margins for the barrier thickness and composition that are much wider than those for, say, the Josephson-junction devices.
- The parameters are not affected by magnetic flux trapped in superconducting electrodes and ground plane, again contrasting well with the Josephson-junction circuits.
- The devices can combine high operation speeds with extremely low power consumption (see Table 1). Moreover, most useful characteristics are improved as the junction size decreases. The size of the junction is not limited by the underlying physics until extremely small dimensions, of the order of one nanometer (determined by the sum of the barrier thickness and the doubled Debye screening length of the electrodes), have been reached. Thus the single-electronic circuits are capable of an extremely large-scale integration, hardly attainable even by advanced nanolithography [29, 30].

The last fact is reminiscent of an intriguing possibility of using single molecules as the basic elements of the electronic circuits. In this "molecular electronic device" field (see, e.g., [43, 44]) several plausible ways to store and transfer the digital information have been suggested. However, the only way considered for rapid processing of the information was resonant tunneling (see [43, pp. 51, 121]) requiring an exact trimming of the tunnel barriers and quantum wells employed. Such accuracy would not be necessary if the single-electron transfer were used. For example, a molecular analog of the R-SET [Figure 13(b)] could be composed of just three electroactive (conducting) macromolecules separated by two gaps transparent for the electron tunneling, with wide margins for their transparencies.

Realization of the molecular single-electron circuits by self-assembly methods would be a great step into the future. Of course, this possibility (if feasible at all) would require a lot of effort.

7. Conclusion

The coexistence of two types of electric conduction, the continuous current flow in metals or semiconductors, and the discrete single-electron transfer through the tunnel barriers, makes possible a new group of effects in structures with very small tunnel junctions at low temperatures (see Table 1). The main feature of all these effects is a high degree of time correlation between single-electron tunneling events, either the successive tunnelings in the same junction, or the near-simultaneous tunnelings in different junctions, or both. The effects present several new possibilities for applied electronics, including a new type of microwave generation, control of a nonvanishing dc current by subelectron electric charges, and processing of digital information bits in the form of single electrons.

Several effects of the new group have been observed experimentally, and there is hardly any doubt at present of the correctness of the basic concepts of their theory. Some important problems, however, do remain in the theory:

1. It is not yet clear what constitutes the conditions of the coherent single-electron transfer through non-tunnel-type "weak links" [45], i.e., the metallic-conducting microshorts connecting two bulk metals. One possible guess is that the volume of the weak link should contain not more than one electron, i.e., $na^3 \lesssim 1$, where n is the conduction-electron concentration.
2. Experimental realization of the coherent SET oscillations could be greatly facilitated by placing an element with metallic resistance $R_S \gg R_Q = \pi\hbar/2e^2$ very close to the tunnel junction (see Section 3). However, R_Q is also a resistance scale of the metal-nonmetal transition due to localization (see, e.g., [46]). One may wonder whether this transition imposes a fundamental ban on the realization of such a resistor while retaining the continuous character of its conductivity.
3. Finally, there exists no detailed theory of the discrete tunneling event itself, including an important sequential stage of the charge relaxation inside the electrodes (i.e., of the restoration of their internal electroneutrality). In such a theory, not only the time τ_0 of tunneling itself [31] but also the time τ_p (see Section 3) should appear in a natural way. This theory would in particular resolve the problem of the relativistic cutoff of the capacitance C_L raised by Büttiker and Landauer [31].

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K. K. Likharev *Department of Physics, Moscow State University, Moscow 119899 GSP, U.S.S.R.* Dr. Likharev received his Candidate of Sciences degree from Moscow State University in 1969, and his

Doctor of Sciences degree from the Higher Attestation Committee of the U.S.S.R. in 1979. Since 1969, he has held permanent research positions at Moscow State University (1969, Junior Scientist; 1973, Senior Scientist; 1987, Major Scientist). As a guest scientist, he has also carried out research work at the State University of New York, Stony Brook, and at the University of California, Berkeley. Dr. Likharev is the author of several books, including the monograph *Dynamics of Josephson Junctions and Circuits*, published both in Russian and English (Gordon and Breach Publishers, New York, 1986). His current scientific interests are in physics and in applications of ultrasmall tunnel junctions and high-T_c superconductivity.