Possible self-selection of tunnel channels with reproducible electrical parameters

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Coulomb correlations may cause self-selection of quasi-one-dimensional current channels in a specially prepared granulated material. The characteristic resistance R_0 of a single channel is much larger than the resistance quantum R_K. It depends on bulk properties of the material and on the macroscopic geometry of the sample, and may be tuned by nearby gate. Total resistance of the sample becomes quantized in units of R_0. Values of R_0 and control voltages may be *reproducible* from sample to sample. The present experimental status of this idea and its possible applications to single-electron devices operating at up to room temperature are discussed. © *1995 American Institute of Physics.*

When typical size (and typical capacitance C) of a conducting piece scales down, it generates new energy scale $E_C \equiv e^2/C \gg k_B T$ well above thermal noise (e is electron charge). Providing a weak interface to the outer world here gives decisive role to *macroscopic charge quantization.*[1,2] E_C may range from $k_B \cdot 1$ K (aluminum island between tunnels made with e-beam lithography[2]—numerously demonstrated experimental result) to $k_B \cdot 10^3$ K (our hypothetical claim for naturally grown metallic islands, experimentally approved for grains within polysilicon channel[3]).

1. Charge quantization with one-junction isolation

Let us consider (Fig. 1) the simplest system which allows for changing the quantized charge $q = en$ of metallic island Z. Let the total capacitance of the island Z be C (the sum of the tunnel and the nontunnel capacitances) and the tunnel resistance of the junction be R. We want the energy of the island to be sensitive to the number of electrons residing on the island. So we should compare the *level spacing* ΔE (energy difference for the states with $|\Delta q| = e$) with the quantum *linewidth* δE of the energy level and $k_B T$ scale:

$$\delta E \ll |\Delta E|, \quad k_B T \ll |\Delta E|. \tag{1}$$

The linewidth δE is expressed in terms of the *lifetime* τ_l of an excited charge state: $\delta E \simeq \hbar/\tau_l$. The typical voltage across the junction is $V \simeq -\Delta E/e$. The average tunnel current is $\langle I \rangle = e/\tau_l$. By definition of tunnel resistance: $R \simeq V/\langle I \rangle = (-\Delta E/e)/(e/\tau_l)$. Hence $\tau_l \simeq e^2 R/(-\Delta E)$ and $\delta E \simeq -\hbar \Delta E/e^2 R$. Estimate $\Delta E \simeq -E_C$ gives typical lifetime $\tau_l \simeq RC$, and with Eqs. (1) leads to the conditions ($R_K \equiv 2\pi\hbar/e^2$):

$$R \gg R_K/2\pi = 4.1 \ \text{k}\Omega, \quad C \ll e^2/(k_B T). \tag{2}$$

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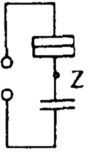

FIG. 1. Single-electron box. Divided rectangle denotes tunnel junction with tunnel resistance R. Voltage source is on the left.

The Al/AlO_x/Al tunnel junctions fabricated by electron-beam lithography with a tunnel barrier thickness $d=2$ nm and area $A=50\times 50$ nm^2 have[4] $R\simeq 100$ kΩ. The tunnel resistance R cannot be raised further because this would strongly decrease the reproducibility of the samples. Island capacitance C here is on the order of the tunnel capacitance. Hence $E_C\simeq k_B\cdot 10$ K and with $T\ll 10$ K we expect the required charge quantization.

The tunneling electron should dissipate certain energy $E_{\rm diss}$. Otherwise, it will not settle on the target island and will form a coherent state involving two or more islands. The duration of this dissipation is in fact *a time τ of tunneling*. Dissipation is needed for the whole macroscopic system to sense tunneling. Since $\tau\simeq\hbar/E_{\rm diss}$ and $E_{\rm diss}\simeq|\Delta E|$, Eqs. (2) lead to the relation $\tau\ll\tau_l$: the electron stays most of the time (τ_l) on a certain island, and only rarely and quickly (spending a time τ) changes place. For typical lithography parameters[4] we have $\tau\simeq\hbar/(e^2/C)=1\times 10^{-12}$ s and $\tau_l\gtrsim RC=2\times 10^{-10}$ s, which illustrates the accuracy of Eqs. (1) in such systems.

Let i be the number of "internal" islands with a fixed charge en_i, x be the number of "external" islands with fixed voltages Φ_x, and N_x be the number of electrons which enter the external island at some initial moment. The total energy of the system comprising electrostatic field of the islands and external voltage sources is:[1,5]

$$E(n,N,\Phi)=(1/2)e^2\sum_{ii'} n_iC_{ii'}^{-1}n_{i'}+e\sum_{ix} n_iD_{ix}\Phi_x+e\sum_{x} N_x\Phi_x. \qquad (3)$$

This energy (along with the thermal contribution) can be spent for dissipation while tunneling. Equation (3) gives the Coulomb spectrum of the Fermi levels of the system as a function of its electrostatic state.

2. Charge quantization with tunnel-chain isolation

Poor accuracy of charge quantization in a single tunnel device can be improved if we replace a single tunnel with a chain of $N\geqslant 2$ tunnel junctions with approximately equal capacitances C (see Fig. 2). Today, lithographic technology can minimize stray capacitance of the islands between junctions: $C_s\ll C$. If the employed voltages V and X are of the scale of E_C/e, the system in Fig. 2 exhibits a useful property. Its electrostatic energy, given by Eq. (3), is never minimized when a nonzero number of electrons occupies islands *between* junctions. Only the electron residing on either Z or $+V$ electrodes forms a (quasi-) stable state. The electron sitting on Z sees between Z and $+V$ a number of energetically unfavorable positions between junctions and thus it is isolated with a multiple tunnel barrier from another nearest stable position on $+V$. This complex barrier

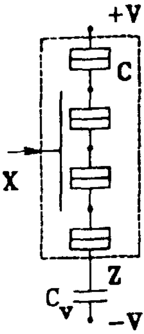

FIG. 2. Island Z isolated with chain of $N=4$ junctions. Both states $n_z=0$ and $n_z=1$ minimize energy equation (3) and are separated with multiple tunnel barrier. The transition $0\rightarrow 1$ can be forced by applying (and then removing) the negative signal (voltage or charge) to gate X which is capacitively coupled to the chain of junctions.

may be suppressed (allowing change of n_z) either by changing the voltage V (which we associate with a dc voltage source) or by applying negative voltage to gate X, or (most important here) by simply removing the electron charge e from gate X. Beginning with $n_z=0$ and holding $V=\text{const}$, we can switch to $n_z=1$ just by manipulating the gate signal. A method for processing and storing one-bit information was proposed in Ref. 6.

The device (consisting of $N=7$ Al/AlO_x/Al junctions with $R\sim 300$ kΩ, size $\sim 50\times 70$ nm^2, fabricated with e-beam lithography) shown in Fig. 2 was tested in Ref. 7. The trapping energy barrier heights was $k_B\cdot 3.5$ K and typical $V\simeq 1$ mV, n_z was steady for over two hours at 50 mK.

3. Quasi-stable states. Quantum and thermal noise

Tunnel chain provides much better isolation and the charge states of the node Z become quasi-stable. In Sec. 1 we stressed that it is important to avoid coherent mixing of discrete charge states. Let us determine how this contributes to the decay of quasi-stable states. Consider the device in Fig. 2 with $N=2$ in a state $n_z=1$ in such a bias configuration that $E(n_z=1)>E(n_z=0)$. Simple one-electron tunneling is still suppressed since the position on the intermediate island between tunnels has the energy $E(n_{\text{int}}=1,\ n_z=0)>E(n_{\text{int}}=0,\ n_z=1)$. Thus, a new complex potential barrier involving the forbidden state $n_{\text{int}}=1$ is formed (it is evidently no ordinary tunnel barrier of the junctions). But these states, $n_z=1$ and $n_{\text{int}}=1$, can form a coherent mixture of states which will be able to decay, adding one electron to $+V$ and thus subtracting it from Z. The mixture decays with the right energy balance $\Delta E<0$ and we can expect it to have the regular decay probability $1/\tau_l$. But the probability for this nonstationary mixture to find itself near the energy $E(n_{\text{int}}=1,\ n_z=0)$ is[8] $\delta E/\Delta E$. For the escape time τ_Q of the trapped electron due to quantum coherency mixing we therefore have: $1/\tau_Q\simeq(\delta E/\Delta E)(1/\tau_l)$. Recalling Sec. 1, we find $\tau_Q\simeq(2\pi R/R_K)\tau_l$. When $N>2$, coherent mixture should include approximately $N-1$ forbidden states before it will be able to decay regularly. By induction we obtain $\tau_Q(N)=(2\pi R/R_K)^{N-1}\tau_l$.

For realization[7] of an aluminum trap this gives $\tau_Q(7)\simeq 10^{13}$ s, which is still very little on the error scale of modern computers.[5] Increasing N and E_C parameters gives a more pronounced macroscopic charge quantization.

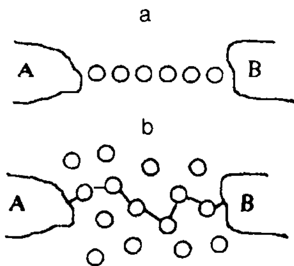

FIG. 3. a—Ideal chain of nanoscale granules between conducting terminals A and B would be nice but cannot be realized. Gate electrode (not shown) could be placed on top and across the chain or positioned on the side. b—In composite media of metallic balls stuffed into an insulator electrons select a quasi-1D channel of balls.

Another reason for increasing N and E_C is the classic expression for thermally activated escape time $\tau_T \simeq \tau_t \exp(E_{tr}/k_B T)$. For the N trap the trapping barrier height is $E_{tr} \simeq E_C N^2 C_v/(C+NC_v)$. Therefore, τ_T grows with N and E_C/T. Even for an exotic $T=50$ mK the system[7] has $\tau_T \simeq 10^{18}$ s.

4. Self-organization of 1D ball current chains in naturally grown composite media

We have seen above that only multi-junction isolation of a metallic island can lead to practical single-electron devices. Analysis[9] shows that increasing N and E_C brings the desired accuracy (with each junction defined lithographically) provided that uniformity of the tunnels is high, and that there are no charged contaminations in the vicinity of the reference nodes and junction chains. Aluminum technology employed in Ref. 6 is near the best possible E_C and uniformity values provided the e-beam lithography is used to make *each* tunnel in a chain. The same applies to the approach [2] with gated Schottky barriers that separate the 100-nm lakes of 2D electron gas (2DEG) in GaAs/AlGaAs heterostructures. Both approaches leave us within 10-mK operating temperature range. Current tendency is to leave the e-beam patterning only for wiring leads and for outlining the general shape of chains. Junctions and islands within these outlined chains are provided by more "natural" means before or after the e-beam stage.

This seems to be a way to increase the operating temperature without loss of the charge quantization accuracy. Consider a single metallic ball with a radius $r=2$ nm, which is incorporated into a medium like SiO_2 with a typical dielectric constant $\varepsilon=5$. In this case the Coulomb scale would be $E_C=e^2/(2\cdot 4\pi\varepsilon\varepsilon_0 r)=k_B\times 10^3$ K. Of course, any tunnel barrier touching this ball will increase its capacitance and decrease E_C, but later we will see how this effect can be minimized.

There was an old idea[2] to grow chains of such granules between some conductors (Fig. 3a) by means of a self-assembly of such chains. The only obstacle was the lack of realization of such a self-assembly process.

Today, however, we have a well-established technology[10] which provides us with a precisely controllable medium of conducting balls stuffed into an insulator. We can accordingly transform an old idea of self-assembly *at the stage of system construction* into the idea of self-organization of a 1D ball current channel *at the stage of current transport* through a disordered composite medium. The electrical properties of this channel are reproducible and depend only on the material parameters of the medium and on its

overall geometry. To obtain this feature, technology should carefully control the means radius r of the balls, the form and the width Δr of the distribution of the radius, the mean minimal separation l between immediate neighbors, and the thickness T, length L, and width W of this composite film that contains balls.

Let us examine Fig. 3b. Let $\Delta r \simeq r/2$. Since l is also dispersed, the energy E_C needed to charge a ball with one extra electron essentially changes from ball to ball. The probability to populate a ball is $\sim \exp(-E_C)$, which is a sensitive function of E_C. At low temperatures and with a small voltage applied between A and B, we evidently have Coulomb blockade of electron transport. As the voltage increases toward the threshold of the Coulomb blockade, we expect that the threshold will be lifted first due to formation of a single shortest chain of balls with lowest E_C and separated by smallest l.

The mean l must be chosen in such a way that it leads to a typical tunnel resistance between balls $R \simeq 100$ kΩ. In Sec. 1 we saw that this is enough for a good localization of the electron on a particular ball. Looking at the reference parameters given in Sec. 1, we might say that to obtain $R(r,l) \simeq 100$ kΩ with $r \simeq 2$ nm, we should have $l \simeq 1$ nm. To estimate the volume concentration of metal in our composite material, we assume that balls form a cubic lattice. This assumption leads to the value $\rho_V \simeq (4\pi/3)r^3/(2r+l)^3 \simeq 0.2$, which can be easily provided by technology.[10] But sure residual contaminations in the medium between balls will form localized centers which will facilitate electronic transport[11] and reduce the effective R. To improve R reduction we need to increase l, say, an order of magnitude, i.e., up to $l \simeq 10$ nm. The corresponding metal volume concentration is $\rho_V \simeq 0.02$, which is even easier to obtain. At the same time, this value of l justifies our assumption made above that full capacitance of the ball in such a medium can be only a few percent more than the capacitance of a single ball.

If a channel deviates too far from a straight line, its total resistance will go too high and a less resistive (and straighter) channel will emerge. We can therefore estimate the number of balls in our channel as $N \simeq L/l$. Then its total resistance is $R_0 \simeq RL/l$. Recall that now we have $r \simeq l$, which means that the self-capacitance of the ball is no less than the tunnel capacitance. This makes irrelevant the estimates for the trapping barrier height made in Sec. 4. Here the complex barrier heights is on the order of $E_{tr} \gtrsim E_{C\,\min}$, where $E_{C\,\min} \simeq e^2/[2 \cdot 4\pi\varepsilon\varepsilon_0(r+\Delta r)]$ is the typical charging energy of the most energetically favorable balls.

It seems reasonable to choose the width and thickness of the film is such a way that $W \simeq 3l$ and $T \simeq 3l$, in order to ensure that electrons have enough choice to select the proper 1D ball chain. But this choice should not be too rich to ensure that formation of exactly one channel be the most probable occurrence near the threshold.

The total Coulomb threshold for transport voltage is $V_{th} \simeq E_{tr}/e$. With an increase in the transport voltage beyond the typical value of $V_{smea} \simeq E_{C\,\max}/e$ with $E_{C\,\max} \simeq e^2/[2 \cdot 4\pi\varepsilon\varepsilon_0(r-\Delta r)]$, the single-channel picture will be evidently smeared out with multiple-channel net current that flows on top of all the barriers. The latter regime was carefully analyzed long ago.[12] For voltages V such that $V_{th} < V < V_{smea}$ we expect gradual transition from one-channel current to net current. For more narrow but finite range of voltages V above V_{th} and for finite range of gate voltages we can expect that the

same single channel persists. The resistance in this case depends on the gate voltage. Note that for $\Delta r \simeq r/2$ we have $V_{\text{smea}} \simeq 3V_{\text{th}}$.

Estimates above give only the mean statistical value of the electrodynamic parameters which characterize the 1D ball channel. We can estimate fundamental deviations from these values, noting that the number N of balls in the channel inevitably fluctuates at least as $\Delta N=1$ from sample to sample. We see that the total resistance of the channel has an error $\simeq \Delta N/N$ and can be kept within 10% with the choice $N=10$. But the threshold V_{th} and the typical gate voltages needed to modulate the current do *not* depend on ΔN.

Two mechanisms contribute to the formation of a 1D ball channel with proper tunnel resistance between the balls.

- Electrons prefer balls with lowest charging energy E_C.
- If two balls are too close together so that the resistance $R \simeq R_K$, then electrons do not distinguish these balls. Such a pair should be regarded as a composite ball within a channel. We see that even if the technology accidentally places some of the balls too close to each other, proper values $R \gg R_K$ will be found anyway.

5. Quasi-1D ball current channel versus quasi-1D ballistic current channel

Note that the whole picture is based on energy balance consideration and should *not* be sensitive to the nature of electron transport between balls. This transport may be a mixture[11] of direct tunneling between balls, resonant tunneling via localized centers, thermally activated jumps, etc. We just need to maintain conditions (such as the parameters r and l), ensuring the absence of coherent mixing of electrons that reside on different balls. Our parameter R may have a more general meaning of typical value of differential resistance at low voltages for voltage-biased pair of balls. That is why we avoid using the name "tunnel channel" in favor of "ball current channel."

The self-selected 1D ball channel has a resistance much higher than the resistance quantum $R_0 \gg N \cdot R_K$, and it can be modulated by external gate because of the modulating trapping barrier height. The channel itself (as a geometrical entity) stays intact within the finite gate voltage range.

Recall that quasi-1D ballistic current channel in mesoscopic samples has the resistance R_K, and that the external gate can change only the number of such channels that can fit into the sample. This leads to quantization of the longitudinal resistance. Recall also that electron wave function preserves coherency along the whole length of the ballistic channel and electrochemical potential has the same value along it.

In our ball current channel wave function coherency is intentionally broken at each inter-ball spacing. After each tunneling or hopping event the electron thermalizes on the new ball and settles down at the new Fermi level characteristic of the given charge state of the system.

6. Experimental status of the 1D ball channel idea

Until now, the idea of self-selection of quasi-1D ball channel was demonstrated only in the nonmetallic domain. But studies in different media justify the opinion that it is not the details of the structure, but the geometry and capacitance that govern this phenomenon.[11]

The authors of Ref. 13 used a GaAs wafer randomly δ-doped with Si donors (5×10^{12} cm^{-2}) within a few atomic layers. This thin Si-doped layer is about 10 nm below the 2DEG layer which in turn is about 20 nm below the GaAs surface. To outline the chain shape a strip was left (by regular e-beam lithography means) in 2D electron gas which was 150 nm wide and 200 nm long. Adjusting the voltage applied to the side gate, it is possible to force the formation of lakes in 2DEG which are $\simeq 10$ nm in size and are exactly on top of each donor. Lakes are isolated with barriers of typical tunnel resistance $R \simeq 100$ kΩ. Self-selection of lakes with reference to lowest charging energy E_C organizes a valley: one-dimensional chain of up to ten lakes through which a tunnel current can flow.

Fabricated devices similar to that shown in Fig. 2 (regarded as memory cells) demonstrated escape time of several hours at a much higher temperature, $T=4.2$ K. Unfortunately, this approach is (evidently) heavily bounded with unpredictable background charges of the lakes. To combat this circumstance, the authors of Ref. 13 employed n-electrons-per-bit coding with n up to 40.

To outline the tunnel chain the authors of Ref. 3 deposited 4-nm-thick, 100-nm-long, 100-nm-wide strip of amorphous Si. Subsequent crystallization produced vertical cylinder crystals 4 nm high and 10 nm in diameter. Similar to Sec. 4, a valley of 1D chain of up to 15 grains was formed. Isolated from the valley with a multiple barrier, there always exists another grain with low E_C, which represent the most deep swamp. Discrete electrons enter or leave the swamp following appropriate gate signal. But the tunnel current flowing through the valley is greatly influenced by the charge state n_{sw} of the swamp. $|\Delta n_{sw}|=1$ can change the current from 60 pA to <0.1 pA with a transport voltage of 20 mV even at room temperature. Both such states survived for no less than one hour at $T=300$ K. Switching to any of $0 \leqslant n_{sw} \leqslant 5$ states following the gate signal was demonstrated. The authors called their device "read-only memory for mobile computers/communicators."

The authors of Ref. 3 have been heavily criticized for a too straightforward interpretation of the mechanism involved. The basic issue of what is the physical reason for electron localization on polysilicon grains seems very far from being understood. This paper is nonetheless important because it demonstrates: 1) self-selective nature of ball chains and 2) definite ability to control channel resistance with external gate.

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