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THERMAL HOPPING OF SINGLE CHARGES IN STRONG DISSIPATION LIMIT

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Macroscopic charge quantization is analyzed in a case when thermal activation across trapping barriers dominates over tunneling, and barrier heights are comparable with charging and thermal energies. Barrier suppression, image force due to finite electron charge, and contribution of non-stable charge states are considered. Presented effects may be in part responsible for recent experimental data on high-temperature single-electronics in semiconductor and granulated systems.

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Macroscopic charge quantization in solids [1] allows for manipulation of discreet electrons. It is widely believed to be a consequence of Coulomb blockade of tunneling. But macroscopic charge quantization is no less basic quantum phenomena, than quantum tunneling. Both can co-exist in one and the same system, but also can manifest themselves each one independently. This was understood in the early years of "single-electronics" [2]. But since then most (if not all) of theoretical and experimental efforts were aimed at case of charge quantization in tunneling systems.

The main reason for this was experimental availability of trapping barriers formed of (1) metal oxides and (2) breaks in 2D electron gas. In these cases barrier height is of the order of 10 eV. This excludes any mechanism for electron transport but quantum tunneling through this barrier at any reasonable temperatures. Recall that $k_B \times 300$ K $\simeq 26$ meV only.

Experimental studies of charge quantizing systems formed of the base of semiconductors [3] and composite materials [4] have attracted much attention recently. In these systems trapping barrier height may become comparable with typical temperatures and with typical charging energies. And this makes essential the thermal contribution to electron transport, as long as transport via non-stable charge states.

In the present letter we analyze simple model applicable to the case of charge quantizing systems, where thermally activated hopping of discreet electrons dominate over quantum tunneling. We ague that traditional elimination of non-stable intermediate charge states (due to their low probability weight) may lead to essential loss of accuracy and non-physical results.

1. Macroscopic charge quantization with and without tunneling. Consider conducting piece of matter (island) with full capacitance C_Σ, isolated from the environment with a trapping barrier(s). We want total charge q of the island be macroscopically observable at the scale of electron charge e. In other words,

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q should be a "good" quantum number. For this it necessary, that charging energy or spacing $\Delta E = e^2/C_\Sigma$ between Fermi levels corresponding to charge states with $\Delta q = 1$, obey the relation $\Delta E \gg \delta E$, where δE is maximal quantum line width of charge state. Another obvious requirement being $k_B T \ll \Delta E$. Minimal lifetime $\tau_{life} = \hbar/\delta E$ of charge state is related to the maximum of its decay rate Γ: $\tau_{life} = 1/\Gamma$. So, good charge quantisation means $\Delta E \gg \hbar\Gamma$.

After recharging with $\Delta q = 1$, system should dissipate exactly the energy ΔE. Otherwise it will not settle at the final charge state of the reference island and will form coherent state involving two or more charge states. The duration of this dissipation is in fact *a time τ of switching*. Dissipation is needed for the whole macroscopic system to sense switching. As $\tau \simeq \hbar/\Delta E$ we have alternative relation for good charge quantization: $\tau \ll \tau_{life}$: electron stays most of the time (τ_{life}) on a certain island, and only rarely and quickly (spending time τ) changes place.

In case of dominating tunneling (with tunnel resistance R_{tun}) through the trapping barrier [2]: $\Gamma_{tum} \simeq R_{tun} C_\Sigma$. Leading to the condition of good quantization: $R_{tun} \gg \hbar/e^2 \simeq 4$ kΩ.

Evidently, R_{tun} may go far beyond this point, leading to nearly full suppression of tunnel transport. It may happen when trapping barrier becomes wide enough. At the same time height of the trapping barrier may remain comparable with $k_B T$ scale, which allows for thermal activation of charge carriers above the barrier, according to Arrhenius law [5]:

$$\Gamma = \begin{cases} \nu \exp(-\beta e B), & B > 0 \\ \nu, & B \leq 0 \end{cases}, \qquad (1)$$

where $\beta \equiv 1/k_B T$, eB is trapping barrier height, and ν is "attempt frequency". With frequency ν electrons hit the surface facing trapping barrier.

Traditionally, expression (1) was restricted to the case of low temperatures and high barriers: $\beta e B \gg 1$ (sure only case $B > 0$ was allowed). This is done to keep Γ within relation $\hbar\Gamma \ll \Delta E$. If we go this way, we get very small currents (below experimental resolution), which makes this system less interesting. Let us try go in another direction.

Let us try keep Γ small due to the smallness of ν. Really, $\nu \simeq vnA$, where v is Fermi speed, n is charge carrier concentration in a reference island, and $A = \Delta y \Delta z$ is area of the interface with the trapping barrier. Later we will encounter a case when trapping barrier becomes damped $B \leq 0$. In this pathological case $\Gamma \simeq \nu$. So, $\tau_{life} \simeq 1/\nu$. And switching time $\tau \simeq \Delta x/v$ is just a time needed to pass barrier domain with length Δx with Fermi speed v. Condition $\tau \ll \tau_{life}$ leads to the very simple relation:

$$n \ll 1/(\Delta x \Delta y \Delta z). \qquad (2)$$

Sure $\Delta x \Delta y z \Delta$ is just a volume of the trapping barrier, and relation (2) is just wanting that the total number of charges, injected into the barrier region be much less than 1 at every moment of the system's history. If so, we evidently have discrete (in units of e) switching of charge states *at any temperatures*. With 3D semiconductors we should have in mind barriers with dimensions: $\Delta x \simeq 1$ nm, $\Delta y \simeq 10$ nm, $\Delta z \simeq 10$ nm. So we need a system of semiconductor with $n \ll 10^{19}$ cm^{-3}.

Similarly, in case of 2D electron gas system we have a relation for sheet carrier concentration n_{2D}:

$$n_{2D} \ll 1/\Delta x \Delta y.$$

For barriers with $\Delta x \simeq 1$ nm, $\Delta y \simeq 10$ nm, we need $n_{2D} \ll 10^{13}$ cm^{-2}, to be compared with typical value for 2D electron gas in heterostructures $n_{2D} \simeq 10^{11}$ cm^{-2}.

Finally, to make use of Eq. (1), we need to assume, that Boltzman distribution of single-particle states above Fermi level (corresponding to each charge state of the island) is immediately restored after charge switching. Then successive charge switching events are *not quantum-coherent*. And there is no contribution of hot electrons to the thermal activated events. If so, density matrix of the system is diagonal, and we can make use of the simple detailed balance version of the kinetic equation. Really, duration of dissipation after switching is $\tau \simeq \hbar/\Delta E$. And we already restricted ourselves to the case $\tau \ll \tau_{life}$. We call this case a *strong dissipation limit*.

2. Model system under study. Consider double-barrier structure (Fig. 1) with reference island N (with self-capacitance C_0) carrying full charge ne, isolated with barriers B1 and B2 (each with capacitance C) from two terminals (L and R) with fixed voltages $\pm V$, and with a gate terminal (mutual capacitance C_g) with voltage V_g. We assume that bare (for $n = 0$, $V = 0$, $V_g = 0$) potential profile (as seen by infinitesimal probe charge δe) is like shown on Fig. 1. Triangular form of barriers is not important for our illustration, but greatly simplifies presentation of results.

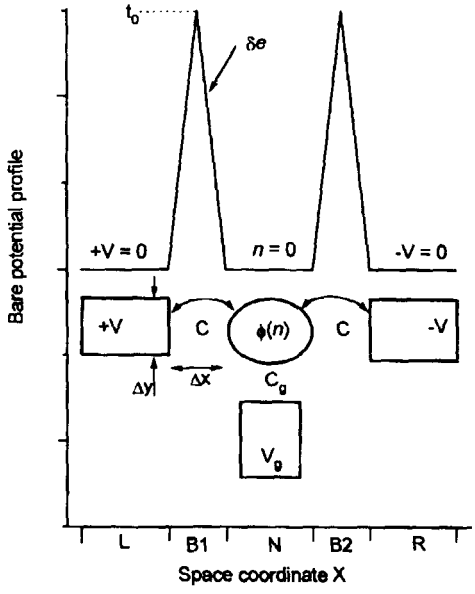

Fig.1. Bare potential profile accross double barrie structure as seen by probe charge δe

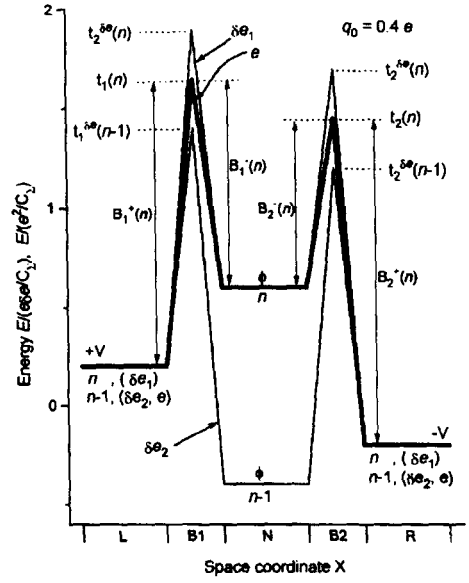

Fig.2. Renormalization of potential due to barrier suppression and finite electron charge

Then charge q_0, induced onto the island N, and potential φ of the island N are:

$$q_0 = C_g V_g, \quad \varphi(n) = (n + q_0)\, e/C_\Sigma, \tag{3}$$

where $C_\Sigma \equiv 2C + C_g + C_0$.

We assume that trapping barriers are insulators, so electrostatic field due to non-zero V and φ penetrates into barrier linearly and adds with the bare potential,

when seen by probe charge δe_1 (case of charge n on the island N, see Fig. 2) and δe_2 (case of charge $n-1$ on the island N):

$$t_{1,2}^{\delta e}(n) = t_0 + \frac{1}{2}\left(V_{1,2} + \varphi(n)\right),$$

where $V_{1,2} = \pm V$.

On Fig. 2 labels n and $n-1$ in the regions L, N, R correspond to the charge state of the island N while moving of probe charge along the curve.

Consider how potential profile changes due to finite value of electron charge e. Really, begin moving e from L rightward, when charge of the island N is $n-1$. Then electron e first sees potential according to curve labeled with δe_2. But when charge e finishes its motion through the barrier region B1 and lands onto the island N, it sure sees potential according to the curve labeled with δe_1, as charge of the island N is now n. Sure when inside region B1, electron sees something in between of curves δe_2 and δe_1. Accurate consideration [6] of multiple electrostatic images of the electron in conducting mirrors N and L, bring us to very simple result. Barrier height $t_k(n)$, renormalized due to finite value of e, is just:

$$t_{1,2}(n) = \frac{1}{2}\left(t_{1,2}^{\delta e}(n-1) + t_{1,2}^{\delta e}(n)\right),$$

where n refers to the charge state of the island N when the electron *under consideration* is landed on island N. Actual profile of potential, measured by finite charge e, becomes parabolic rather than linear, between points of lowest and highest potential, but only the highest point value matters for dynamics of thermal activation.

When additional electron is inside the regions B1 or B2, charge of the island N does not have definite value. It is switching.

So we get trapping barriers for the process of increasing (B^+) of n, and decreasing (B^-) of n through the corresponding barrier B1 or B2 (Fig. 2):

$$B_{1,2}^{+}(n) = t_{1,2}(n) - V_{1,2};$$

$$B_{1,2}^{+}(n) = t_{1,2}(n) - \varphi(n).$$

3. Detailed balance. Following Eq. 1, we have:

$$\Gamma(n-1 \to n) \equiv \Gamma^{+}(n),\ \Gamma(n \to n-1) \equiv \Gamma^{-}(n),\ \Gamma^{\pm}(n) \equiv \Gamma_1^{\pm}(n) + \Gamma_2^{\pm}(n)$$

and the detailed balance equations to solve for values of statistical weighs $p(n)$, with which each charge state n (Fig. 3) is contributing to the electron transport through the island:

$$\begin{array}{l} p(n)\,\Gamma^{+}(n+1) = p(n+1)\,\Gamma^{-}(n+1), \quad n_{min} \le n \le n_{max} - 1; \\ \sum_n p(n) = 1 \end{array} \tag{4}$$

Solving this, we get current via the barrier k:

$$I_k = (-1)^{k-1} e \sum_n \left[p(n)\,\Gamma_k^{+}(n+1) - p(n+1)\,\Gamma_k^{-}(n+1)\right]$$

as a function of voltages V, V_g, barrier height t_0, and temperature T, as plotted on Figs. 4-6. Sure $I_1 = I_2$.

4. Stable and non-stable charge states. We call a charge state n stable (Fig. 3) when all (4) barriers trapping it are positive: $B_k^{\pm}(n) > 0$. Stable states fall into interval: $n_{min} \le n \le n_{max}$, where:

$$\begin{aligned} n_{min} &= \text{floor}\left[-\left(\frac{2Ct_0}{e} - \frac{V_1 C}{e} - \frac{1}{2}\right) - q_0\right] + 1, \\ n_{max} &= \text{ceiling}\left[\frac{2Ct_0}{e} - \frac{V_2 C}{e} - \frac{1}{2} - q_0\right] - 1. \end{aligned} \tag{5}$$

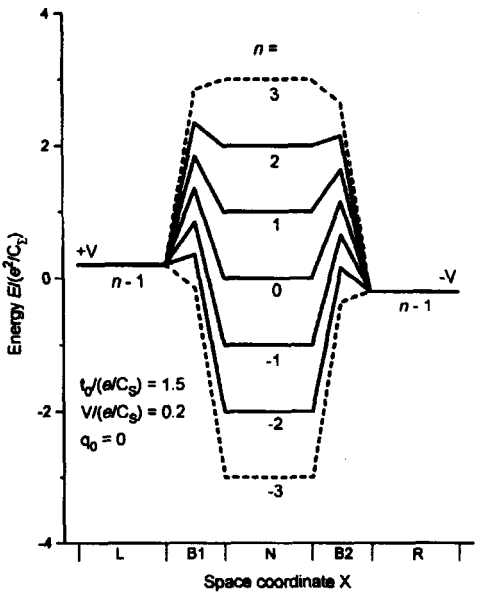

Fig.3. Reeman branching of Fermi levels, reffering to diffent charge states. Stable states are shown with solid curves, non-stable (but stationary) – with dashed curves

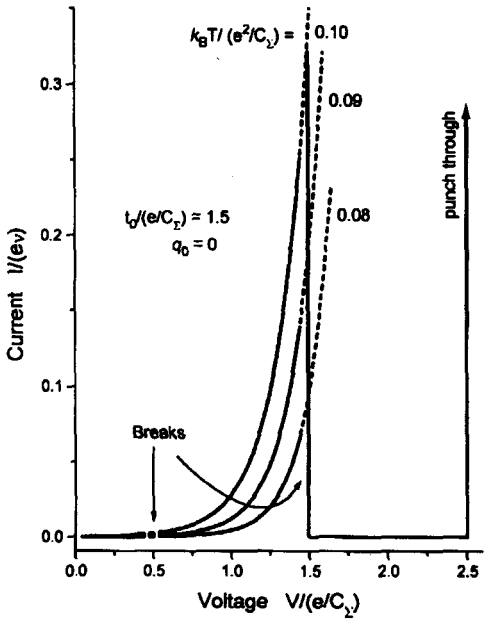

Fig.4. IV curve. Solid curves - contribution of stable states. Dashed curves – full current. "Breaks" refer to at least one barrier small compared to k_BT

When say V is large enough, there may be no stable states at all. States $n = \pm 3$ on Fig. 3 are not stable. But our reasoning in Section (1) ensures, that these non-stable states are still *stationary*, that is they have definite (from quantum point of view) energy and charge.

When plotting full current (dotted curves) on Figs.4-6, we took into account all charge states with non zero weight $p(n)$ according to (4). Contribution of stable states is given with solid curves. Note that each time, when certain state n becomes non-stable according to (5), solid curves exhibit sharp jumps. Most ultimately it is seen on Fig. 4, where at $V > 3/2\,(e/C_\Sigma)$, there are no stable states at all, and "stable" contribution to current show "Coulomb blockade", until at $V = 5/2\,(e/C_\Sigma)$, all the barriers for all states are suppressed and punch trough is predicted. While full current exhibits exponential growth approaching this point.

Following classical tradition [5], we omitted from the solid (stable) curves regions (breaks) where at least one trapping barrier is small compared to k_BT.

5. Discussion. We employed the simplest model to demonstrate, that charge quantizing systems can still be analyzed in unusual (but important for experiment) parameter range of barrier heights compared with charging and thermal energies, and voltage scales; where thermal activation dominates over tunneling. Sure this model is primitive, and does not take into account a number of important things. To mention but a few, change of capacitance with voltages and temperature, dependence of the potential profiles on temperature, influence of finite thermalization time upon switching event, influence of dissipation within the barrier, more realistic shape of barriers and its dependence upon voltages and temperature, coherent mixing of different charge states (that is quantum correction to our

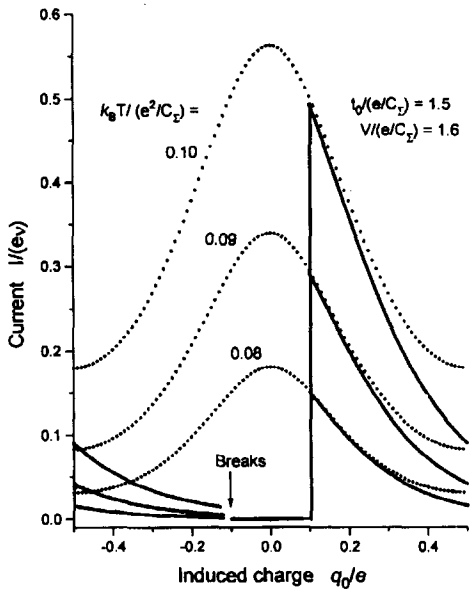

Fig.5. Current vs gate voltage (or induced charge). Region around $q_0 = 0$ corresponds to "Coulomb blockade" of current borned be stable states

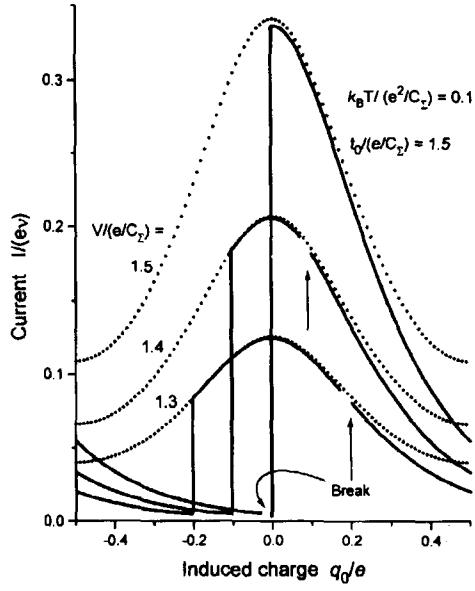

Fig.6. Each jump of "stable" curve corresponds to transision of a certain state n from stable to non-stable status

semi-classical results). Still it outlines qualitative predictions for experiment, like exponential IV curve (simingly seen in [7]) and punch through, with conservation of celebrated single-electron transistor effect. And it gives guidelines for treating non-stable charge states, which was forbidden in traditional theory.

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