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Detailed balance in single-charge traps

Rüdiger Bauernschmitt and Yuli V. Nazarov

Institut für Theoretische Festkörperphysik, Universität Karlsruhe, Engesserstrasse 7, Postfach 6980, D-7500 Karlsruhe 1, Germany

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We propose a model to explain recent experimental results concerning the charge trapping in a system of ultrasmall tunnel junctions. Degradation processes in the system are assumed to generate the strange single-electron jumps observed. The model predicts many spectacular features of charge trapping so that it can easily be verified.

Recent advances in nanotechnology allow one to fabricate systems of ultrasmall tunnel junctions exhibiting a pronounced Coulomb-blockade behavior. The state of such a system is characterized by the integer numbers of charges stored at the small metal islands constituting the system.[1] Depending on the layout and control parameters some of these charge states may be metastable. This means that all the single-electron tunneling processes that can change the state increase the charging energy. If only single-electron processes were allowed, at zero temperature the system would be in this state forever. We refer to such systems as to single-charge traps (SCT).

More complex tunneling processes involving two or more electrons[2] get a SCT to the ground state even at zero temperature. The rate of these cotunneling processes is much smaller than the typical single-electron rates providing long lifetime of the metastable states. If N electrons have to be involved into the process the suppression is proportional to $(R_T/R_K)^{N-1}$, R_T being a typical resistance of the tunnel junction, $R_K = 2\pi\hbar/e$. Therefore a typical SCT (Ref. 3) (Fig. 1) consists of the store island and the chain of N junctions connecting the island with the bulk electrode. The larger N is, the longer the lifetimes are. At small but finite temperatures the system is switched between the possible metastable states. If we accept the cotunneling mechanism of switching two simple conclusions can be drawn: (i) Since the system is in thermodynamic equilibrium the rates of switching must obey the detailed balance relation. (ii) The cotunneling rates must be strongly suppressed if the islands are superconductive[6] since the superconducting gap cuts the number of states available for tunneling.

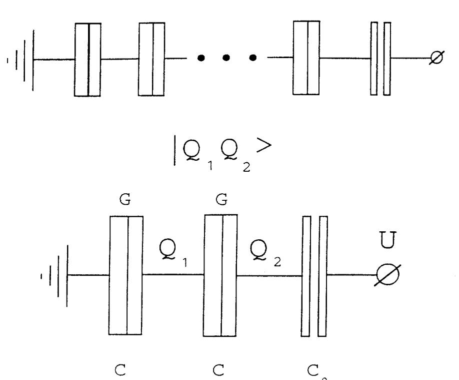

FIG. 1. Single-charge traps.

The SCT with two and four junctions was investigated.[4,5] In both experiments the switching rates observed were in the range 0.1–10 s^{-1}. Nevertheless the results seem to disagree with the cotunneling mechanism of switching. First, no quantitative agreement was obtained. Second, the reported difference between the rates at the normal and superconductive state is marginal compared with the theoretical predictions. The third, and most important point, is the indication on the absence of detailed balance in SCT.[5] This fact implies that the switching rate is determined by some unknown nonequilibrium processes which supply the energy required to switch between metastable states.

There are three ways to bring the energy into the system. First, a high-frequency electromagnetic noise could enter the cooled sample from the room-temperature environment. A sophisticated system of filters is implemented to attenuate that. The second possibility relates to the back influence of a measuring device on the system measured.[7] In fact, the SET electrometer is used for measuring. It is biased by a finite voltage that could supply energy larger than kT. In this case the switching rates would depend strongly on this voltage. That was not observed experimentally. The remaining possibility is to gain energy from an intrinsic source such as the degradation and slow relaxation processes going on in any real system. It is natural to relate these processes to so-called background charges.[9] These charges are located in the dielectric material of the junctions and are supposed to jump rarely and stochastically between discrete positions. They induce an extra voltage drop on the junctions thus influencing the integer charge dynamics. The background charge jump between the neighboring positions is very similar to a switching of a famous Anderson two-level system[10] (TLS) and in fact can be described in the same language. The TLS are thought to be responsible for $1/f$ noise in many systems[11] including the systems of small tunnel junctions.[8]

Every TLS is characterized by an energy difference ΔE between the levels. Those with ΔE of the order of T are jumping back and forth producing $1/f$ noise. Since the rates of TLS switching are supposed to be distributed in a wide range, at any time there are TLS with $\Delta E \gg T$ that did not have time to switch. If there is an interaction between the TLS and the SCT, this energy ΔE may be used to release a single charge kept in the SCT.

We will describe such an interaction with the following Hamiltonian:

$$\begin{aligned} H &= H_{\rm TLS} + H_{\rm SC} + H_{\rm int}, \\ H_{\rm TLS} &= -\Delta E|n_1 = 0\rangle\langle n_1 = 0| \\ &\quad + \left\{ \sum_k T_k^{(1)} b_k^\dagger |n_1 = 0\rangle\langle n_1 = 1| + \text{H.c.} \right\}, \\ H_{\rm SC} &= E_{th}|n_2 = 1\rangle\langle n_2 = 1| \\ &\quad + \left\{ \sum_k T_k^{(2)} a_k^\dagger |n_2 = 0\rangle\langle n_2 = 1| + \text{H.c.} \right\}, \\ H_{\rm int} &= e\delta V|n_1 = 0\rangle\langle n_1 = 0| \otimes |n_2 = 1\rangle\langle n_2 = 1|, \\ &\quad \delta V \ll E_{\rm th}. \end{aligned} \tag{1}$$

Here the TLS may either be in the excited ($n_1 = 1$) or in the ground ($n_1 = 0$) state. We consider the transition between some (meta)stable state ($n_2 = 0$) and an excited state ($n_2 = 1$) with one electron having passed a certain junction. The switching of the TLS induces a voltage difference δV at this junction. The operators a_k, b_j describe the microscopic degrees of freedom related to the corresponding tunneling process. They allow for the dissipation. In particular, a_k originates an electron-hole pair with electron and hole placed on opposite sides of the junction. The origin of b_j depends on the nature of the TLS. In the simplest case switching corresponds to the transition of one electron from the localized state in the insulator to the extended state in the metal. Then the b_j are just annihilation operators of electrons in these extended states. Actually we do not have to know much about a, b because they enter the result only in certain combinations. For example, if one calculates the rates without taking into account the interaction between the systems one obtains:

$$\begin{aligned} \Gamma_{\rm TLS} &= 2\pi \sum_j |T_j|^2 \delta(E_j - \Delta E) \equiv \Gamma_{\rm TLS}(\Delta E), \\ \Gamma_{\rm SC} &= 2\pi \sum_k |T_k|^2 \delta(E_k - E) \equiv \Gamma_{\rm SC}(E) \\ &= (G_T/e^2) E \Theta(E), \end{aligned} \tag{2}$$

G_T being the conductivity of the junction.

The process which we are interested in is as follows. Initially the TLS is in the excited state whereas the SCT is in the (meta)stable state. Finally the TLS is in the ground state. We consider the rate of cotunneling from the state $n_1 = 1, n_2 = 0$ to the state $n_1 = 0, n_2 = 1$ plus excitations k, j.

The amplitude of transition is given by

$$\mathrm{M}_{k,j} = T_j \frac{1}{-\Delta E + E_j} T_k + T_k \frac{1}{E_{th} + E_k} T_j. \tag{3}$$

The rate of interest is a sum of rates over j, k. We replace the summation by integration with making use of (2) and obtain

$$\begin{aligned} \gamma_{an} = \frac{1}{2\pi} \int & d\varepsilon_1 d\varepsilon_2 \Gamma_{\rm TLS}(\varepsilon_1) \Gamma_{\rm SC}(\varepsilon_2) \\ & \times \left(\frac{1}{-\Delta E + \varepsilon_1} + \frac{1}{E_{th} + \varepsilon_2} \right)^2 \\ & \times \delta(-\Delta E + \varepsilon_1 + E_{\rm th} + \varepsilon_2 + e\delta V). \end{aligned} \tag{4}$$

Two things simplify (4) greatly. First, we assume that the ΔE for different TLS are distributed in a region which is much larger than the Coulomb energy scale. In this case $\varepsilon_1 \approx \Delta E \gg E_C$ allows us to forget about the energy dependence of $\Gamma_{\rm TLS}$. Second, we use the fact that $e\delta V \ll E_{\rm th}$. It yields

$$\begin{aligned} \gamma_{an} &= \Gamma_{\rm TLS} \frac{1}{2\pi} \int_0^\infty \frac{d\varepsilon (e\delta V)^2}{(E_{\rm th} + \varepsilon)^4} \Gamma_{SC}(\varepsilon) \\ &\equiv \Gamma_{\rm TLS} \frac{G_T \hbar}{12\pi} (\delta V / E_{\rm th})^2. \end{aligned} \tag{5}$$

It is worth it to stress the universality of the expression obtained. In fact it does not depend on the detailed properties of the TLS. The important dependence on the parameters of the SCT is incorporated in $E_{\rm th}$. One can use the following simple phenomenological expression to desribe concrete experiments:

$$\gamma_{an} = A / E_{\rm th}^2, \tag{6}$$

where A is a phenomenological constant which may depend on the junction. It is given by the sum of $\Gamma_{\rm TLS}(e\delta V)^2$ over *all* the TLS in the sample.

In the superconducting state (5) should be modified taking into account the density of states in the superconductor. In this case

$$\Gamma_{\rm SC}(\varepsilon) = I_{\rm ss}(\varepsilon/e)/e, \tag{7}$$

$I_{\rm ss}(V)$ being the superconductor-superconductor tunneling characteristic.[12] Since $I_{\rm ss} < I_{nn}$ the anomalous rate is suppressed in the superconducting state. Nevertheless, if the superconducting gap Δ is comparable with charging energy the rate is still of the same order of magnitude because the TLS supply enough energy to generate quasiparticle excitations. This is in contrast to the cotunneling rate which is completely suppressed if the energy difference between the initial and final states is less than 2Δ.

At long time scales, the SCT switches many times between the different metastable states. The average charge is determined by the relative populations of the states and below we concentrate on the evaluation of these quantities. At low temperatures the anomalous processes are dominant. The populations are determined by quotients of their rates. In this regime we do not have to know the constant A in (6) for a quantitative description. At rising temperature the normal tunnel rates become more important and a crossover in the system properties takes place. The experimental observation of the crossover allows to us estimate A.

Let the energy difference between metastable and nonmetastable states be large compared to kT. Then we can describe the system in the restricted basis of metastable states. We introduce effective rates $\tilde{\gamma}_{i\to f}$ for the switching between them. Transitions from a metastable state i to a nonmetastable state I lead to higher charging energy of the system. They happen with anomalous rates $\gamma_{i\to I}$ as described above and with normal tunnel rates $\Gamma_{i\to I}$ which are thermally reduced following the detailed balance relation $\Gamma_{i\to I} = \Gamma_{I\to i}e^{-(E_I-Ei)/kT}$. The rates for transitions from the intermediate state I to any other state are only taken into acccount if the charging energy is lowered. They determine a probability $p_{I\to f}$ that by consecutive (co)tunneling the metastable state f is reached from I. In these terms the effective rates read

$$\tilde{\gamma}_{i\to f} = \sum_I (\gamma_{i\to I} + \Gamma_{I\to i}e^{-(E_I-Ei)/kT})p_{I\to f}. \quad (8)$$

They allow us to write down the master equation for the populations P_i of the metastable states,

$$\frac{d}{dt}P_i = \sum_{j\neq i}(-P_i\tilde{\gamma}_{i\to j} + P_j\tilde{\gamma}_{j\to i}). \quad (9)$$

Its stationary solution allows us to calculate the mean charge on the store island. This quantity can easily be observed in experiment because it corresponds to the potential measured by the SET electrometer.[13]

We concentrate on the simplest SCT shown in Fig. 1. It consists of two equal tunnel junctions with capacitances C and one capacitor C_0 in series with a voltage source U.

The expression for the electrostatic energy of the system is

$$E = \frac{e^2}{2C(1+2\eta)}\{\ (1+\eta){Q_1}^2 + 2{Q_2}^2 + 2Q_1Q_2 \\ -2\eta U(Q_1+2Q_2)\} \quad (10)$$

with $\eta = C_0/C$, Q_i the charge on island i divided by e and U the bias voltage in units e/C.

At a given bias voltage there is a certain number of metastable states $|0,Q_m\rangle$ in such a system. The integers Q_m lie in the window

$$\eta U - \frac{1+\eta}{2} < Q_m < \eta U + \frac{1+\eta}{2}. \quad (11)$$

We consider the case of two metastable states $|0,0\rangle$ and $|0,1\rangle$, which is relevant for $\eta = 1$ and U satisfying $0 < U < e/C$. Using (8) we find the effective switching rates $\tilde{\gamma}_{i\to f}$:

$$\tilde{\gamma}_{0\to 1} = (\gamma_{0\to A} + \Gamma_{0\to A})\frac{\Gamma_{A\to 1}}{\Gamma_{A\to 0}+\Gamma_{A\to 1}} \\ +(\gamma_{0\to B} + \Gamma_{0\to B})\frac{\Gamma_{B\to 1}}{\Gamma_{B\to 0}+\Gamma_{B\to 1}}, \quad (12)$$

$$\tilde{\gamma}_{1\to 0} = (\gamma_{1\to A} + \Gamma_{1\to A})\frac{\Gamma_{A\to 0}}{\Gamma_{A\to 1}+\Gamma_{A\to 0}} \\ +(\gamma_{1\to B} + \Gamma_{1\to B})\frac{\Gamma_{B\to 0}}{\Gamma_{B\to 1}+\Gamma_{B\to 0}}. \quad (13)$$

The indices 0 and 1 denote the states $|0,0\rangle$ and $|0,1\rangle$, A and B denote the two relevant intermediate states $|1,0\rangle$ and $|-1,1\rangle$, A stands for the state with the lower of the two energies.

In equilibrium the ratio $\rho = \tilde{\gamma}_{1\to 0}/\tilde{\gamma}_{0\to 1}$ is equal to the ratio of the populations P_0/P_1 of the metastable states. For ρ we find

$$\rho = \frac{\Pi_1 + e^{(E_1-E_A)/kT}}{\Pi_0 + e^{(E_0-E_A)/kT}} \quad (14)$$

with

$$\Pi_{1,0} = \frac{C_A\gamma_{1,0\to A}/\Gamma_{A\to 1,0} + C_B\gamma_{1,0\to B}/\Gamma_{B\to 1,0}}{C_A + C_B e^{-(E_B-E_A)/kT}}, \quad (15)$$

$$C_{A,B} = \frac{\Gamma_{A,B\to 1}\Gamma_{A,B\to 0}}{\Gamma_{A,B\to 0}+\Gamma_{A,B\to 1}}. \quad (16)$$

The behavior of ρ as a function of the applied voltage is governed by the competition of the $\Pi_{1,0}$ and the exponential terms in (14). At $T = 0$ the exponential terms become zero and hence $\rho = \Pi_1/\Pi_2$ in contrast to $\rho = \infty$ if all rates follow detailed balance. In the opposite limit, for large T, we can neglect $\Pi_{1,0}$ and find the classical detailed balance behavior. There is a sharp crossover between the two limits. It takes place in a narrow interval around T^* which can be estimated as $T^* \sim E_c/\ln(\Gamma/\gamma)$, Γ and γ being the orders of magnitude of the normal and the anomalous rates.

Figures 2 and 3 illustrate the behavior previously described. Figure 2 shows a plot of the equilibrium population of $|0,1\rangle$ as a function of the applied voltage at different T. The population is equal to the mean value of the charge in the trap in units e.

If all participating rates followed detailed balance, at $T = 0$, P_1 would jump from 0 to 1 at $U = 0.5e/C$. As $T \sim E_c$ this steplike behavior is washed out.

In our model, on the other hand, we get the sharpest

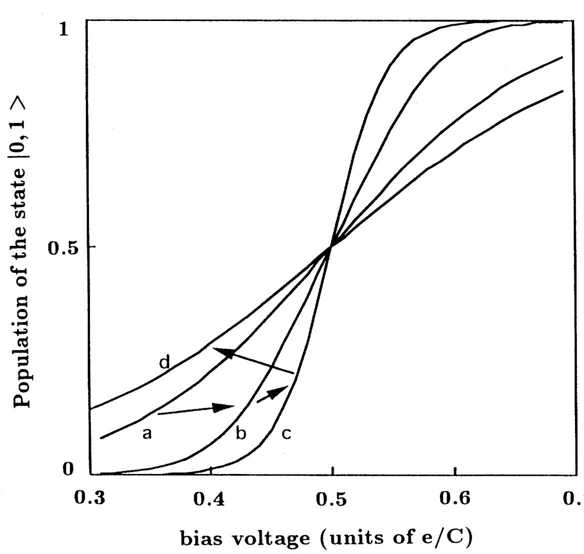

FIG. 2. The population P_1 of the metastable state $|0,1\rangle$ as a function of the bias voltage at different temperatures. T in units of E_c: *a*, 0; *b*, 0.02; *c*, 0.05; *d*, 0.15. The arrows indicate increasing temperature. $C/C_0 = 1$, $\gamma/\Gamma = 10^{-8}$.

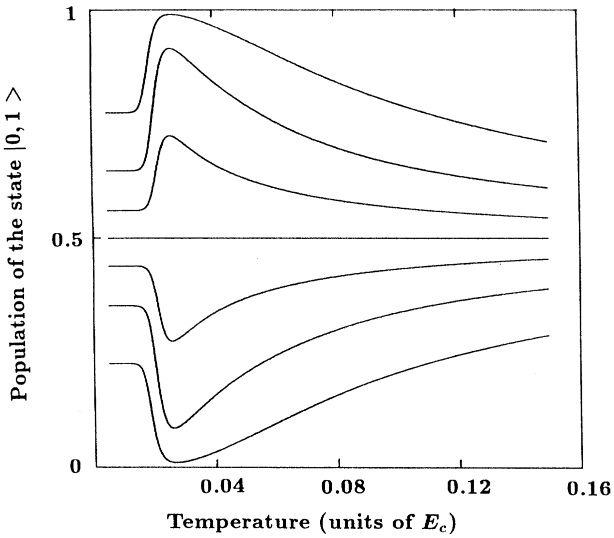

FIG. 3. P_1 as a function of temperature at different bias voltages. Voltage in units of e/C from below: 0.4, 0.45, 0.48, 0.5, 0.52, 0.55, 0.6.

switching between 0 and 1 at a finite temperature T' (curve a, Fig. 2). For higher temperatures we enter the detailed balance regime and the curve becomes washed out in the usual way (curve d). For lower temperatures we enter the anomalous regime (curve b) and approach the curve for $T = 0$ (curve a) which again becomes smoother.

Thus we predict a reentrant temperature behavior. Experimental evidence for such a behavior would confirm our model. In Fig. 3 the population P_1 is plotted versus T at different bias voltages. The crossover between the anomalous and the detailed balance behavior shows up in the sudden change of P_1 in a narrow T region.

In our simple model we considered so far only first-order electron tunneling. In a more realistic model the cotunneling rates of direct transitions between the metastable states will compete with the effective rates calculated above. As cotunneling follows the detailed balance relation it tends to restitute the Boltzmann distribution such that it would destroy the described anomalous behavior. As the anomalous rates $\gamma_{i\to f}$ scale with the junction conductance G, whereas the cotunneling rates scale for two junctions with G^2, our model becomes correct for small G.

If we are going to describe more complex SCT, we shall consider more complex anomalous processes. If the SCT consists of $N \gg 1$ junctions, the relevant scenario of switching may be as follows. First we transfer a charge from the store island to the middle of the chain. This is a higher-order anomalous process involving $N/2$ electrons gaining energy from a TLS switching. Then the charge jumps down to the bulk electrode completing the SCT switching.

The anomalous rate can be estimated as $\Gamma_{\mathrm{TLS}}(e\delta V/E_C)^2(G_T R_K)^{N/2}$. It competes with the rate of a direct cotunneling process which is of the order of $E_C(G_T R_K)^N$. Thus the dynamics of switching is always governed by the anomalous processes in the limit of small G_T and large N.

The minimal switching rates observed are of the order of inverse seconds.[5] From the estimations given above we can extract the number of TLS switchings per second which would provide such a rate. It yields 10^5 s^{-1} which does not seem too high. A highly doped silicon substrate used in the experiments contains many localized charges and their migration may give rise to the effect.

In conclusion, we propose a model to explain the unusual features observed in the slow dynamics of single-charge traps. The degradation processes were shown to generate anomalous charge jumps. Those may govern the slow dynamics of the system breaking the detailed balance rules. The universality of the model allows for the detailed predictions to be checked experimentally. In particular, an unexpected reentrant temperature behavior was predicted. In general, the anomalous jumps change drastically the behavior of single-electron systems at a long time scale and all the present concepts should be revised to take this into account.

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