Statistical model of charge transport in colloidal quantum dot array ⋆

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Abstract: A new statistical model of charge transport in colloidal quantum dot arrays is proposed. It takes into account Coulomb blockade forbidding multiple occupancy of nanocrystals and influence of energetic disorder of interdot space. The model explains power law current transients and the presence of memory effect. The fractional differential analogue of the Ohm law is found phenomenologically for nanocrystal arrays. The model combines ideas that were considered as conflicting by other authors: the Scher-Montroll idea about power law distribution of waiting times in localized states for disordered semiconductors is applied with taking into account Coulomb blockade, Novikov's condition about asymptotical power law distribution of time intervals between successful current pulses in conduction channels is fulfilled, carrier injection blocking predicted by Ginger and Greenham takes place.

Keywords: quantum dot array, stable Lévy laws, fractional derivatives, memory

1. INTRODUCTION

Researchers appealing to a discrete electron spectrum of quantum dots (QDs) often call them by "artificial atoms". An array of identical semiconductor QDs can be considered as artificial solid. Fundamental conceptions of solid state physics can be studied on the base of such systems. Understanding of charge and spin transport processes in QD arrays could lead to applications in spintronics and quantum computation. Despite the sufficient progress in synthesis, description of charge transport in QD arrays is not satisfactory [Novikov (2003); Novikov et al. (2005); Morgan et al. (2002)].

In many samples of colloidal QD arrays (in the lateral geometry), power law decay of current

$$I(t) \propto t^{-\alpha}, \quad 0 < \alpha < 1, \qquad (1)$$

is observed after applying of a large constant voltage $V(t) = V_0 l(t)$ [Morgan et al. (2002); Novikov (2003)], where $l(t)$ is the Heaviside step function. The exponent α is less than 1 and in the general case its value depends on nanocrystal size and temperature. Novikov et al. (2005) assert that (1) is not a bias current, it is a true current from source to drain due to the integral of Eq. (1) is charge and it tends to infinity

$$Q = \int_0^\infty I(t)dt \to \infty.$$

The observed non-exponential relaxation of current can be explained by time dependence of the state of the system. Ginger & Greenham (2000) proposed decreasing of charge flow due to suppression of injection from the contact. This suppression arises because electrons trapped in a nanocrystal prevent transport of other electrons through this QD, flow is jammed. Morgan et al. (2002) explain power law decay of current $I(t)$ by presentation of non-equilibrium electrons distributed over QD array as the Coulomb glass.

Novikov et al. (2005) proposed the model based on a stationary random process as authors assert. An array consists of $N \gg 1$ identical independent channels operating in the parallel regime. Each channel opens in random time moments and conducts a current pulse. These channels are completely characterized by the distribution of waiting times T between successful pulses. Authors Novikov et al. (2005) postulated that this distribution has a heavy tail of the power law kind

$$\Psi(t) = \mathsf{P}(T > t) \propto t^{-\nu}, \qquad 0 < \nu < 1, \qquad t \to \infty. \quad (2)$$

The mean value of such random variable diverges and this fact provides specific statistical properties of the process. In particular, memory effects arise.

The model satisfactorily explains power law current transients and power law noise spectrum but it does not reveal the physical mechanisms of the process, they postulate the distribution (2). As Novikov et al. (2005) assert, the base of the model is the stationary stochastic process and this stationarity contradicts to the time dependence of the state of the system, in particular, to the hypothesis about injection blocking from the contact that must occur due to the charge balance conditions. Many additional questions arise. What is the nature of this channels, why does the distribution between successful pulses has power law asymptotics, why are current pulses discrete and identical in values? Furthermore, if memory of the process is explained in frameworks of the latent variable conception [Uchaikin (2008)], how is this approach combined with the stationarity of the process? The goal of the present paper is to solve the contradictions listed above.

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Further, a new statistical model that describes power law relaxation of current and memory phenomena is proposed. It is shown that the basic random process is nonstationary. The model leads on the one hand to the idea of charge injection blocking, and it conforms to Novikov's model on the other.

2. CHARGE TRANSPORT BLOCKING IN THE MODIFIED SCHER-MONTROLL MODEL

To answer the questions listed in the Introduction we use a modification of the Scher-Montroll model. The classical version of this model explains successfully mean features of dispersive transport in disordered semiconductors [Scher (1975)]. Novikov (2003) provides arguments that the standard Scher-Montroll model does not describe power law decay of current in QD arrays. The model predicts unlimited accumulation of charge in a sample if an injection rate from the contact is constant. Injection blocking takes place in the modified model taking into account the Coulomb blockade effect.

Coulomb interaction is long range and it leads to collective phenomena of charge distribution over a sample Novikov (2003). These effects become apparent in the case of small values of voltage u. To study transport in arrays without taking into account the long range character of Coulomb interaction one has to apply large u. In the experiments described in Ref. Morgan et al. (2002); Drndic et al. (2002), values of voltage between source and drain were large (of the order 100 V) and they correspond to several hundred meV between neighboring QDs that is of the order of the interdot Coulomb energy and the nanocrystal charging energy.

In our model a QD array is represented as two or three dimensional lattice, QDs are situated in points of this lattice. The last can be considered as a set of parallel one-dimensional nanocrystal rows (conduction channels). Electrons perform one-sided random walk in the direction opposite to the applied field. The proposed model is qualitative and reflects main statistical properties of the process without long-range correlations. Nevertheless it allows to interpret power law decay of current, the presence of memory in nanocrystal arrays, to substantiate charge injection blocking, and it is agree with Novikov's phenomenological model.

The main idea proposed in Refs. Novikov (2003); Novikov et al. (2005) for explanation of power law current transients concludes in the assumption that time intervals between successful current pulses in conduction channels are independent random variables with distribution having heavy power law tails. The authors say nothing about nature and shape of these channels. In the present model, channels are associated with one-dimensional nanocrystal rows in ordered array. Let us show that if sojourn times in QDs are distributed according to the asymptotic power law with the exponent $0 < \nu < 1$, then time intervals between successful electron jumps from array to drain in one row have the same power law asymptotics in distribution.

Tunnelling from one nanocrystal to another, electrons follow each other. Coulomb repulsion between electrons allows no multiple occupancy of nanocrystals. Let at the moment t_j, j-th electron of some channel has jumped from array to drain. Let us find a distribution of the time interval $\theta = t_{j+1} - t_j$ between exits of this electron (j) and the next one ($j+1$) in the channel.

The next carrier ($j+1$) can be trapped in any nanocrystal of the channel except the last QD. Let p_n are probabilities to occupy the n-th QD at the moment t_j, where n is a number of nanocrystal in the channel. For the times $t > t_j$, in front of the ($j+1$)-th carrier there are no non-equilibrium electrons trapped in the channel. In other words the carrier will not be influenced by Coulomb repulsion from the side of electrons going ahead. Random walk of the carrier will not be blocked. The exit time of the ($j+1$)-th electron counted since the moment t_j is summed up of sojourn times in nanocrystals which the carrier has to visit before leaving the array,

$$T = \tau_n' + \sum_{k=n+1}^{N} \tau_k. \tag{3}$$

Here τ_k is a sojourn time in the k-th QD. The stroke of the time τ_n' signifies that the ($j+1$)-th electron has spent part of its waiting time in the n-th QD till the moment of exit of the j-th carrier.

It is known, that if two random variables having distributions with identical power law asymptotics are summed up, then the distribution of a resultant variable has asymptotics of the same order. Indeed, the asymptotic form ($\lambda \to 0$) of the Laplace transformation of a PDF with heavy power law tail is as follows,

$$\widehat{\psi}(\lambda) = \int_0^\infty e^{-\lambda t}\psi(t)\,dt \sim 1-(\lambda/c)^\mu, \quad 0<\alpha<1, \quad \lambda\to 0,$$

where c is a scale constant. Distribution of sum of two random variables is the convolution of their distributions. The Laplace transformation of the convolution of two functions is product of their Laplace images. For PDFs with identical power law asymptotics, we have

$$\left(1-\frac{\lambda^\mu}{c_1^\mu}\right)\left(1-\frac{\lambda^\mu}{c_2^\mu}\right) = 1-\left(\frac{\lambda}{b}\right)^\mu + \frac{\lambda^{2\mu}}{(c_1c_2)^\mu} \sim 1-(\lambda/b)^\mu,$$

$$\lambda\to 0, \quad b = \left(\frac{1}{c_1^\mu}+\frac{1}{c_2^\mu}\right)^{-1/\mu}.$$

If distributions of sojourn times τ_k and τ_n' are asymptotical power laws with the exponent $0<\nu<1$, the random variable (3) has a distribution with asymptotics of the same order. The time $\tau_n' = \tau_n - \theta$, where θ is the exit time of the j-th electron counted since the moment of trapping of the ($j+1$)-th electron into n-th nanocrystal. The random time θ has some PDF $p_\theta(t)$. Then

$$P(\tau_n' > t) = \int_0^\infty P(\tau_n > t+t')\;p_\theta(t')\;dt' \sim$$

$$\sim \frac{c^{-\nu}}{\Gamma(1-\nu)}\int_0^\infty (t+t')^{-\nu}p_T(t')\;dt' \sim \frac{(ct)^{-\nu}}{\Gamma(1-\nu)}, \quad t\to\infty.$$

Thus, the hypothesis about sojourn times distributed according to asymptotical power law conforms to Novikov's model assuming power law distributions of intervals between successful current pulses in conduction channels.

3. POWER-LAW DECAY OF CURRENT

Distribution of number of pulses in some channel is as follows,

$$p_n = \mathsf{P}(N(t)=n) = \mathsf{P}(N(t)<n+1) - \mathsf{P}(N(t)<n) = \\ = P(T_{n+1}>t) - P(T_n>t),$$

where $T_n = \sum_{i=0}^{n} T_i$. According to the generalized limit theorem (for more details, see Uchaikin & Zolotarev (1999)),

$$P(T_n<t) \sim G_+(cn^{-1/\nu}t;\nu), \qquad t\to\infty.$$

Here $G_+(t;\nu)$ is a distribution function of stable random variables. Thus, we have

$$p_n \sim G_+(cn^{-1/\nu}t;\nu) - G_+(c(n+1)^{-1/\nu}t;\nu) \sim \\ \sim \nu^{-1}n^{-1-1/\nu}ct\ g_+(cn^{-1/\nu}t;\nu),$$

where $g_+(t;\nu)$ is the stable density. The current is determined by the expression

$$i(t) = \frac{d\langle Q\rangle}{dt} = eZ\frac{d}{dt}\sum np_n \sim \\ \sim eZ\frac{d}{dt}\left[\nu^{-1}(ct)^{\nu}\int\limits_0^{\infty}\xi^{-1/\nu}g_+(\xi^{-1/\nu};\nu)d\xi\right] = \\ = eZ\nu c(ct)^{\nu-1}\int\limits_0^{\infty}s^{-\nu}g_+(s;\nu)ds = \frac{eZc^{\nu}}{\Gamma(\nu)}\ t^{\nu-1}, \\ t\gg c^{-1}, \quad 0<\nu<1,$$

where Z is the number of channels.

Thus, the exponent α of power-law decay of current is connected with the model parameter ν by the relation $\alpha = 1-\nu$. The results of Monte Carlo simulation confirming analytical calculations are presented in Sec. 5.

4. PHYSICAL FOUNDATIONS

Different physical mechanisms leading to asymptotical power-law distribution of waiting times are known (see Sibatov & Uchaikin (2009) and references therein). In most cases, such behavior is assumed to relate to disorder of a medium. Due to disordered structure of interdot space, energetic disorder always exists in colloidal QD arrays even in the case of ideal arrangement of nanocrystals in the coordinate space. Tunnelling probabilities from one nanocrystal to another are determined by height and weight of dividing energy barrier.

Random waiting time τ in some quantum dot is characterized by the probability

$$\mathrm{P}\{\tau>t\} = \exp(-t/\theta), \tag{4}$$

where the parameter θ represents the mean sojourn time in this nanocrystal if the next one is empty. According to the Zommerfeld-Bete quasi-classical formula for tunneling **?**:

$$\theta = \beta[\exp(\gamma d\sqrt{W})-1], \tag{5}$$

where d is a distance to a neighboring lattice point, W is a work function of electron transfer from one QD to another, the parameter β is inversely proportional to electric field intensity. As we see, the parameter θ depends exponentially on width d and height W of the dividing barrier, which have dispersion due to disorder. This leads to sufficient spread of θ values. After averaging over the QD ensemble, the mean value $\langle\theta\rangle$ can diverge. Following the work Sibatov & Uchaikin (2009), we choose the gamma density to model the distribution of the quantity $y = d\sqrt{W}$, the exponential density is a special case.

After averaging over y values, the PDF of θ has the form of asymptotical power law dependence multiplied by a slowly varying function Sibatov & Uchaikin (2009):

$$p_\theta(t) \propto \left(\ln\frac{t}{\beta}\right)^{-1+\frac{\langle d\sqrt{W}\rangle^2}{\mathrm{D}[d\sqrt{W}]}}\left(\frac{t}{\beta}\right)^{-1-\frac{\langle d\sqrt{W}\rangle}{\gamma\mathrm{D}[d\sqrt{W}]}}.$$

Thus the power law asymptotics is characterized by the parameter

$$\nu = \frac{\left\langle d\sqrt{W}\right\rangle}{\gamma\mathrm{D}[d\sqrt{W}]},$$

where $\mathrm{D}[d\sqrt{W}]$ is the square of fluctuations of the quantity $d\sqrt{W}$. As shown in Sibatov & Uchaikin (2009), the waiting time distribution has the same power law asymptotics. The mean waiting time diverges in the case $\nu<1$, in other words when spread of the quantity y is large enough, $\mathrm{D}[d\sqrt{W}] > \gamma^{-1}\left\langle d\sqrt{W}\right\rangle$.

5. MONTE CARLO SIMULATION

Electrons jump in one direction if the electric field is strong enough, in other words they perform one-dimensional one-sided random walk. Let $i=1,2,...N$ are the lattice point numbers. According to the reasonings of the previous section, the simulation scheme can be realized in the following way. The set of random jump rates μ_j is generated for all electrons trapped in QDs of the array. The probability of jump during a small time dt is determined by the product $\mu_j dt$. In addition, the set of random variables γ_j uniformly distributed in the interval (0,1) is generated. If the relation $\gamma_j<\mu_j dt$ is satisfied and the next QD is empty, the j-th electron jumps from the i-th QD to the $(i+1)$-th one and then new jump rate is generated for this electron. If the relation is not satisfied or the next QD contains trapped electron, then the electron stays put.

The jump rates μ_j must be distributed with the following PDF

$$\rho(\mu) = \frac{\nu}{\mu}\left(\frac{\mu}{\mu_{\max}}\right)^{\nu}, \qquad 0<\mu<\mu_{\max}.$$

Indeed, the sojourn time in a chosen QD before jump is distributed according to the exponential law

$$\mathsf{P}(T_j>t) = \exp(-\mu_j t),$$

and after averaging over the QD ensemble, we obtain the distribution of waiting times with power law tails

$$\mathsf{P}(\tau>t) = \langle\exp(-\mu_j t)\rangle = \\ = \int\limits_0^{\mu_{\max}}\rho(\mu)\exp(-\mu t)d\mu \sim \Gamma(\nu+1)(\mu_{\max}t)^{-\nu}, \quad t\to\infty.$$

When electrons perform jump into the drain, the current pulse is registered. Observed current is averaged over all channels of the nanocrystal array. The current calculated in such way is presented in Fig. 1, a in reduced coordinates. It decays according to the power law with the exponent $\alpha = 1-\nu$. Numeric calculations of the distribution of waiting times T between successful current pulses (Fig. 1, b) confirm the analytical results obtained in Sec. 2.

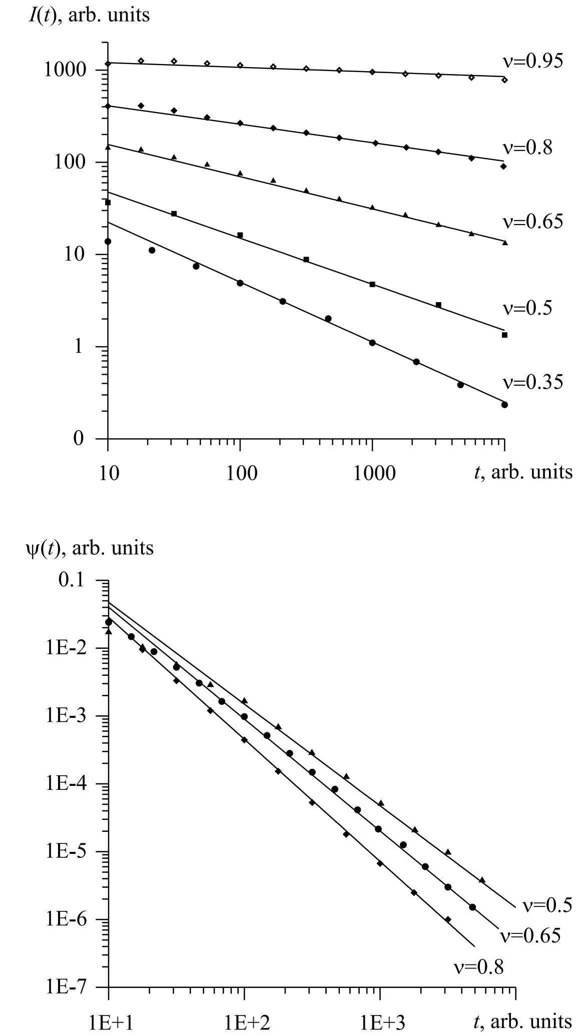

Fig. 1. a) Simulated decay of current (points), lines are power law dependencies with the exponent $-\alpha$, where $\alpha = 1-\nu$. b) PDFs of waiting times between successful current pulses in a channel. Slopes correspond to the exponents $-1-\nu$. Points are the result of Monte Carlo simulation.

6. RELATION BETWEEN CURRENT AND VOLTAGE

Now, we shall obtain the expression connecting current and voltage in QD arrays from the empirical law (1). An analogous derivation was for the first time performed by Westerlund (1991) for dielectrics. Note, that this derivation is true in the case of independence of the parameter α on voltage.

The current (1) is the response to the voltage step. Any voltage signal can be presented in the form of superposition of steps: $u(t) \approx \sum_i \Delta u_i \, 1(t - i\Delta t)$. Consequently, we have

$$i(t) \propto \lim_{\Delta t \to 0} \sum_i \Delta u_i (t - t_i)^{-\alpha} =$$

$$= u(0)\; t^{-\alpha} + \int_0^t \frac{du(t')}{dt'}(t-t')^{-\alpha} dt' = \frac{d}{dt} \int_0^t \frac{u(t')}{(t-t')^{\alpha}} dt'.$$

It is known that the operator

$${}_0\mathsf{D}_t^{\alpha} u(t) = \frac{1}{\Gamma(1-\alpha)} \frac{d}{dt} \int_0^t \frac{u(t')}{(t-t')^{\alpha}} dt'$$

is the fractional Riemann-Liouville derivative of the order $0 < \alpha < 1$. Note, the initial time moment $t = 0$ implies that $u = 0$ in the interval $(-\infty, 0)$. If we are not attached to some initial moment, the Riemann-Liouville derivative has to be replaced by the Weil derivative Uchaikin (2008), in other words we have to take $-\infty$ instead 0 as the lower limit of integration. In this case, current and voltage are related through the fractional differential relation

$$i(t) = K_{\alpha}\; {}_{-\infty}\mathsf{D}_t^{\alpha} u(t). \tag{6}$$

When $\alpha \to 0$, this relation represents the Ohm law for a conductor with conductivity K_0, when $\alpha \to 1$, the relation coincides with the expression for ideal dielectric with capacity K_1.

The parameter K_α can be easy determined from experimental data. If current decays according to the power law $i(t) \approx At^{-\alpha}$ after applying of the step voltage $u(t) = u_0 \; 1(t)$, then the constant K_α is connected with the parameter A by the relationship

$$K_\alpha = A\Gamma(1-\alpha), \qquad 0 < \alpha < 1.$$

The fractional differential analogue of Ohm's law indicates that QD arrays are attractive due to their small sizes as an element base for PID-controllers of fractional order becoming more and more popular [Tang et al. (2009); Si-Ammour et al. (2009)].

7. MEMORY EFFECT

In Ref. [Fischbein & Drndic (2005)], conductivity switching in CdSe nanocrystal arrays was studied experimentally (in the field-effect transistor geometry). Authors found out that arrays show hereditary behavior, and they assumed possible application of QD arrays as memory elements. Memory in arrays can be erased electrically or optically and is rewritable.

In the previous section, the expression relating current and voltage in arrays is obtained phenomenologically. This expression contains the fractional Weil derivative. It is known the fractional differential operator is nonlocal. In other words, it is not determined by function behavior in a vicinity of some point, but depends on function values in some interval, in our case in $(-\infty, t]$. Therefore, the relationship (6) assumes the presence of memory in the system. In Refs. [Uchaikin & Uchaikin (2005, 2007)], the memory regeneration phenomenon was predicted on the base of the fractional differential current-voltage relationship for dielectrics in the case when α is close but less than 1. In Ref. [Uchaikin (2008)], authors report about observation of this phenomenon in the oil capacitor.

Main features of the phenomenon are as follows. Charging-discharging process is studied: a constant voltage is applied to the capacitor during some time θ, the capacitor is charged, then the current signal is registered during the discharging process. The charging time θ is varied. It is found out that the current signal depends on a prehistory of the system. For α close to 1, the relaxation goes according to an exponential law at initial time interval, relative differences between curves corresponding different charging times are small, i. e. signals coincide, but since some time moment the relaxation turns into a long-term power-law regime and differences between curves become visible. In other words a memory about the system prehistory is regenerated.

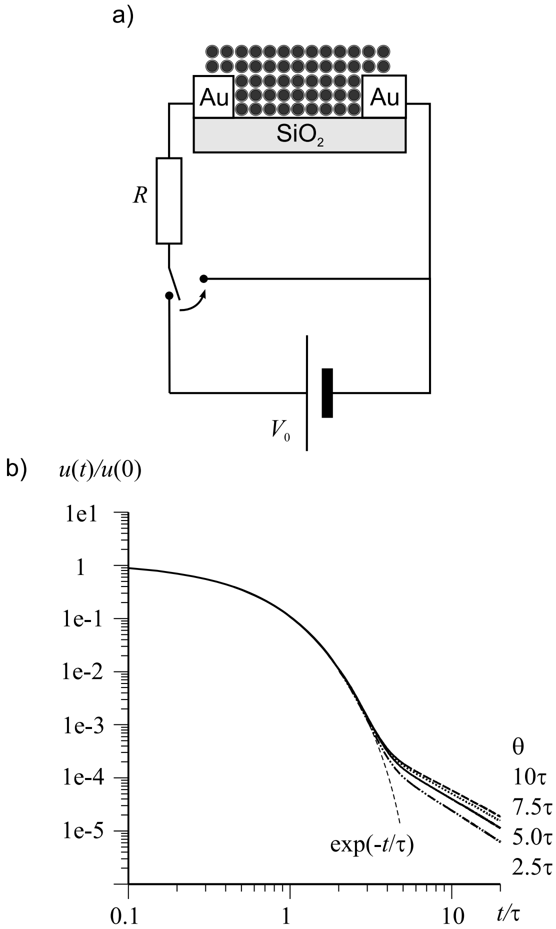

Fig. 2. a) The scheme for study of the memory phenomenon in nanocrystal arrays. b) Solutions to the equation (8) corresponding to different charging times $\theta/\tau = 10$, 7.5, 5.0, 2.5 and $\alpha = 0.998$.

The scheme for study of the memory phenomenon in a nanocrystal array can be realized as shown in Fig. 2, a. Taking into account an active resistance, we write the circuit equation

$$i(t)R + u(t) = V(t), \tag{7}$$

where $V(t)$ is a known time dependence of the external voltage that can be presented in the form

$$V(t) = V_0[l(t+\theta) - l(t)].$$

Here $l(t)$ is the step unit function. Current and voltage are related through Eq. (6). The circuit equation can be rewritten as follows

$$_{-\infty}D_t^\alpha f_\alpha(t) + \tau^{-\alpha} f_\alpha(t) = V(t), \tag{8}$$

where $\tau = (K_\alpha R)^{1/\alpha}$, $f_\alpha(t) = \tau^\alpha u(t)$.

The Green function of this equation (. ?) has the form

$$G_\alpha(t) = t^{\alpha-1} E_{\alpha,\alpha}\left(-(t/\tau)^\alpha\right). \tag{9}$$

Here

$$E_{\alpha,\beta}(x) = \sum_{j=0}^{\infty} \frac{x^j}{\Gamma(\alpha j + \beta)}$$

is the two-parametric Mittag-Leffler function.

Solution to Eq. (8) can be presented in the form [?]

$$f_\alpha(t) = \tau^\alpha V_0 \left[E_\alpha\left(-\frac{t^\alpha}{\tau^\alpha}\right) - E_\alpha\left(-\frac{(t+\theta)^\alpha}{\tau^\alpha}\right)\right], \tag{10}$$

where

$$E_\alpha(x) = \sum_{j=0}^{\infty} \frac{x^j}{\Gamma(\alpha j + 1)}$$

is the one-parametric Mittag-Leffler function. The solutions (10) for $\alpha = 0.998$ and for different times θ are presented in Fig. 2 b.

The following question arises, how can we reach values of α close to 1. In Ref. Morgan et al. (2002), dependencies $\alpha(u)$ and $\alpha(T)$ of the parameter on voltage and temperature are obtained experimentally for CdSe nanocrystal arrays. These investigations show that at voltages large enough (approximately > 100 V) α depends weakly on u. Remind that the relationship (6) and other ones following from it were obtained in the assumption about weak dependence of α on voltage. Dependencies $\alpha(T)$ indicate that the parameter increases with temperature approximately for $T > 200$ K, for temperatures $T > 250$ K it becomes close to 1.

8. CONCLUSION

The statistical model of charge transport in colloidal quantum dot arrays is proposed. The model neglects by long-range correlations conditioned by Coulomb interaction. It is justified for voltages large enough. Correlations in the model arise due to taking into account the Coulomb blockade effect forbidding trapping of more than 1 non-equilibrium electron by a QD. The model is in essence the modified Scher-Montroll model. The standard Scher-Montroll model is usually applied to dispersive transport in disordered semiconductors and dielectrics. A distinction is in the absence of independence of electron trajectories in the new model. Power-law asymptotics in the waiting time distribution is a consequence of spread of interdot energy barriers. This spread is related to energetic disorder of interdot space. A spread of jump rates increases if arrangement of nanocrystals in the matrix is not ordered.

The fractional differential current-voltage relationship is obtained phenomenologically. It represents the analogue of Ohm's law. Due to non-locality of the fractional derivative operator, the relation describes a process with power law memory. From this relationship it is follows that the memory regeneration phenomenon has to be observed in quantum dot arrays for values of α close, but less than 1. Due to their small sizes, QD arrays are perspective as elements for PID-controllers of fractional orders, it is possible their application as memory elements.

The proposed model is simple and does not take into account some particular qualities of the process, for example long-range correlations are neglected. Nevertheless it is in accordance with experimental data obtained at the corresponding voltages and with phenomenological models proposed earlier. Thereby, it allows to solve contradictions indicated in Novikov (2003); Novikov et al. (2005). The model takes into account two important aspects of the process: 1) energetic disorder of interdot space; 2) inhibition of multiple occupancy of QDs due to the Coulomb blockade. Agreement with experiments and other models indicates adequacy of these two positions at voltage values large enough . At the same time taking into account long-range Coulomb interaction is quite possible in this phenomenological model, at least in numerical simulation.

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