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NONMAXWELL RELAXATION IN DISORDERED MEDIA: PHYSICAL MECHANISMS AND FRACTIONAL RELAXATION EQUATIONS

V.E. Arkhincheev
Buryat Science Center, Siberian Branch of the Russian Academy of Sciences, 670047 Ulan-Ude, Russian Federation
and
The Abdus Salam International Centre for Theoretical Physics, Trieste, Italy.

Abstract

The problem of charge relaxation in disordered systems has been solved. It is shown, that due to the inhomogeneity of the medium the charge relaxation has a non-Maxwell character. The two physical mechanisms of a such behavior have been founded. The first one is connected with the "fractality" of conducting ways. The second mechanism of nonexponential non-Maxwell behavior is connected with the frequency dispersion of effective conductivity of heterogeneous medium, initially consisting of conducting phases without dispersion. The new generalized relaxation equations in the form of fractional temporal integro-differential equations are deduced.

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1 Introduction

As is known the charge relaxation in the conducting medium with e conductivity σ is described by the following system of the equations:

$$\frac{\partial \rho(\vec{r},t)}{\partial t} + div(\sigma E(\vec{r},t) = o \tag{1}$$

$$div\Big(E(\vec{r},t)\Big) = 4\pi\rho(\vec{r},t) \tag{2}$$

In the case of homogeneous isotropic system the relaxation equation follows from this system.

$$\frac{\partial \rho(\vec{r},t)}{\partial t} + 4\pi\sigma\rho(\vec{r},t) = 0 \tag{3}$$

The solution of this relaxation has exponential Maxwell form:

$$\rho(\vec{r},t) = \rho(\vec{r},0)exp\Big(-\frac{t}{\tau}\Big) \tag{4}$$

where $\tau = (4\pi\sigma)^{-1}$ is the Maxwell relaxation time.

What will be in the case of disordered media? Does the Maxwell exponential law conserve in the disordered media? At first it seems that the relaxation law conserves its exponential form with replacement the medium conductivity value for its effective value: $\sigma \leftrightarrow \sigma_{eff}$ But as it will be shown below that it is not so. The charge relaxation in the disordered system has nonexponential non-Maxwell behavior in a general case.

Furthermore the nonexponential behavior is observed in many experiments. Usually the following typical non-Maxwell dependencies are observed:

$$\rho(\vec{r},t) = \rho(\vec{r},0)exp\Big[-(\frac{t}{\tau})^{\nu}\Big] \tag{5}$$

It is the so-called empirical Kalraushe law or, in other words, the fractional-exponential law. The following dependency is a power law [1],[2]:

$$\rho(\vec{r},t) = \rho(\vec{r},0)\Big(\frac{\tau}{t}\Big)^{\gamma} \tag{6}$$

Here the values of the exponents ν and γ are changed as: $0 < \nu < 1$, and $0 < \gamma < 1$.

The aim of this paper is to study a charge relaxation in disordered systems and to understand the physical mechanisms, which lead to such nonexponential behavior, and deduce the generalized relaxation equations, describing the nonexponential relaxation The structure of the paper is as follows. The miscroscopic models of disordered media: comb model of percolation clusters and heterogeneous two-phase media such as layered and random structures are considered in paragraph 2. On the basis of these models the physical mechanisms of nonexponential charge relaxation are established. The generalized relaxation equations of fractional order are deduced in paragraph 3.

2 Physical mechanisms of non-Maxwell relaxation

To understand the physical mechanisms of nonexponential and non-Maxwell charge relaxation we will consider the following microscopical models of disordered systems: comb model structure and heterogeneous two-phase media (layered and random inhomogeneous medium).

We will show that in the case of the comb model the charge relaxation has a nonexponential character and it is connected with "fractality" of conducting percolation ways. The non-Maxwell relaxation is obtained in the case of the heterogeneous medium also. This mechanism is connected with appearance of "surface" charge at the interface between phases. The value of this "surface" charge is changed in the alternative electric field. It results in the emergence of the frequency dispersion of the effective conductivity of heterogeneous medium is appeared.

2.1 Relaxation on comb model of the percolation clusters

In this paragraph we consider the two-dimensional comb model of the percolation clusters. This model consists of one-dimensionlal axis (backbone) with fingers of infinite lengths, which are perpendicular to the axis. Firstly this model is introduced to study anomalous character of random walks in disordered systems [3]. This model allows to take into account the influence of the impasses -"dead ends" of the percolation clusters for transport. In Refs. [4], [5] the exact solution of the diffusion problem on the comb model was given. The feature of the transport transfer in the considered model is that the movement in the X-direction is possible only along the axis of the structure (Y=0). This means that the the transfer coefficient σ_{xx} differs from zero only at $y = 0$:

$$\sigma_{xx} = \sigma_1 \delta(y) \tag{7}$$

Here $\delta(y)$ is the Dirac delta-function, i.e. $X-$ component of the current is equal to:

$$J_x = \sigma_{xx} E_x = \sigma_1 \delta(y) E_x \tag{8}$$

The conductivity along fingers is considered as usual: $\sigma_{yy} = \sigma_2$. Thus the charge relaxation on the comb structure is described by the following conductivity tensor:

$$\hat{\sigma} = \begin{pmatrix} \sigma_1 \delta(y) & 0 \\ 0 & \sigma_2 \end{pmatrix}$$

From the continuity equation (1) with the above tensor conductivity and Poisson equation (2) one obtains the equation for an electric potential φ:

$$\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\frac{\partial \varphi}{\partial t} + 4\pi\left(\sigma_1 \delta(y)\frac{\partial^2}{\partial x^2} + \sigma_2 \frac{\partial^2}{\partial y^2}\right)\varphi = 0 \tag{9}$$

Let us make Fourier transformations with respect to time and on coordinate X. So we obtain that the Green function in the mixed (ω, k, y)- representation is described by the equation:

$$\left(-k^2 + \frac{\partial^2}{\partial y^2}\right) i\omega\varphi(\omega, k, y) + 4\pi\left(-\sigma_1 \delta(y) k^2 + \sigma_2 \frac{\partial^2}{\partial y^2}\right)\varphi(\omega, k, y) = 0 \tag{10}$$

Let us search the solution as:

$$\varphi(\omega, k, y) = \phi(\omega, k) exp(-\lambda|y|) \tag{11}$$

Inserting the solution (11) in the equation (10) one finds the parameter λ:

$$\lambda = |k| \frac{i\omega}{i\omega + 4\pi\sigma_2} \tag{12}$$

and the expression for the function ϕ:

$$\phi(\omega, k) = \frac{1}{2\lambda(i\omega + 4\pi\sigma_2) + 4\pi\sigma_1 k^2} \tag{13}$$

So one knows the electric potential (11), it is easy to establish the corresponding Green function of the equation for a concentration:

$$G(\omega, k, y) = \frac{2\lambda\delta(y) + k^2 - \lambda^2}{2\lambda(i\omega + 4\pi\sigma_2) + 4\pi\sigma_1 k^2} exp(-\lambda|y|) \tag{14}$$

One can write this result (14) as a sum of the two parts: "wire" concentration G_l and "volume" G_v:

$$G(\omega, k, y) = G_l(\omega, k) + G_v(i\omega, k, y) \tag{15}$$

where

$$G_l(\omega, k) = \frac{2\lambda\delta(y)}{2\lambda(i\omega + 4\pi\sigma_2) + 4\pi\sigma_1 k^2} \tag{16}$$

and

$$G_v(\omega, k, y) = \frac{k^2 - \lambda^2}{2\lambda(i\omega + 4\pi\sigma_2) + 4\pi\sigma_1 k^2} exp(-\lambda|y|) \tag{17}$$

Let us in a more details consider the time evolution of the "wire" concentration. As it follows from (15) it describes by the following equation:

$$[2\lambda(i\omega + 4\pi\sigma_2) + 4\pi\sigma_1 k^2]\rho_l(\omega, k, 0) = 0 \tag{18}$$

In the case ($\omega << \sigma_2$) it is simplified:

$$[\sqrt{4\pi i\sigma_2\omega} + 4\pi\sigma_1|k|]\rho_l(\omega, k, 0) = 0 \tag{19}$$

Correspondingly, in the (k, t)-representation the generalized relaxation of fractional temporal order is obtained:

$$\left(\frac{\partial^{\frac{1}{2}}}{\partial t^{\frac{1}{2}}} + 4\pi\tilde{D}|k|\right)\rho_l(k, t) = 0 \tag{20}$$

where $\tilde{D} = 2\frac{\sigma_1}{\sqrt{\sigma_2}}$

After some calculations one obtains the gaussain evolution of the one-dimensional charge density along axis of the structure:

$$G_l(x, t) = \frac{exp(-\frac{x^2}{4\tilde{D}t})}{2\sqrt{\pi\tilde{D}t}} \tag{21}$$

As another example we shall find the expression for the Green function, averaged on the coordinate y:

$$\hat{G}(\omega,k)=\int_{-\infty}^{\infty}G(\omega,k,y)dy=\frac{k^2}{\lambda^2[2\lambda(i\omega+4\pi\sigma_2)+4\pi\sigma_1k^2]} \tag{22}$$

At the $\sigma_2=0$ this Green function has a simple form:

$$\hat{G}(\omega,k)=\frac{1}{2|k|(i\omega+2\pi\sigma_1|k|)} \tag{23}$$

that is it is described the following equation:

$$\left(\frac{\partial}{\partial t}+2\pi\sigma_1|k|\right)\hat{\rho}(k,t)=0 \tag{24}$$

One can see that it essentially differs from (20) and for (x,t)- representation one obtains a simple formulae.

$$\hat{G}(x,t)=\frac{Vt}{x^2+(Vt)^2} \tag{25}$$

A similar expression is obtained when the charges are located in the plane and the field has all three components [6], [7].

In the general case and in the usual (x,t)-representation it is equal to:

$$\begin{aligned}\hat{G}(x,t) &= \frac{Vt}{x^2+(Vt)^2}exp(-2\pi\sigma_2t)+\frac{\sigma_2t}{\pi}\int_0^{\infty}\frac{V\tau}{x^2+(V\tau)^2}I_0\left(\frac{\sigma_2\sqrt{t^2-\tau^2}}{2}\right)\\ &+I_1\left(\frac{\sigma_2\sqrt{t^2-\tau^2}}{2}\right)\frac{\tau}{\sqrt{t^2-\tau^2}}d\tau\end{aligned} \tag{26}$$

Here I_0, I_1 are the modified Bessel functions, $V=2\pi\sigma_1$.

So in this paragraph the study of the relaxation on comb structure allows to establish the first mechanism of nonexponential non-Maxwell behavior. It is connected with that the electric field has a three-dimensional components, but relaxation of charge is possible only along some conducting lines (percolation ways).

2.2 Relaxation in heterogeneous two-phase media: effective medium aproximation

To consider charge relaxation in the heterogeneous two-phase conducting media let us transform the above system equation (1) and (2) to the form of the direct current equations:

$$div\left(\vec{j}(\vec{r},t)\right)=0,\ \vec{j}=(\sigma+\frac{i\omega}{4\pi})\vec{e} \tag{27}$$

Consequently, the conductivity at the frequency ω is described by the expression:

$$\sigma(\omega)=(\sigma+\frac{i\omega}{4\pi}) \tag{28}$$

Accordingly similar equations are hold in an inhomogeneous media for the averaged quantities $\vec{J}=<\vec{j}>,\ \vec{E}=<\vec{e}>$:

$$div\vec{J}=0,\ div\vec{E}=4\pi<\rho>, J=\sigma_{eff}\vec{E} \tag{29}$$

The effective conductivity of the medium σ_{eff} depends both the medium parameters and the frequency. At this approach all information of the medium inhomogeneity is contained in the effective conductivity. Thus according to (29) to describe charge relaxation in an inhomogeneous medium within the considered approximation it is enough to know the frequence dependency of its effective conductivity.

2.2.1 Layered structures

Consider inhomogeneous medium obtained by the random alternation of layers with equal size and different conductivities σ_1 and σ_2. Firstly we study the cherge relaxation along layers.

A) Along layers

In this case due to boundary conditions the electric field is homogeneous $\vec{E} = const$ and the averaging over random placement of phases is carried easily. So in this case:

$$\sigma_{eff} =< \sigma > + \frac{i\omega}{4\pi} \tag{30}$$

where $< \sigma >= x\sigma_1 + (1-x)\sigma_2$, and x is the concentration of the first phase. So the corresponding equation for the averaged concentration is following:

$$(i\omega + 4\pi\sigma) < \rho >= 0 \tag{31}$$

The Green's function of equation (31) is:

$$G^{\|}(\omega) = \frac{G_0}{(i\omega + 4\pi < \sigma >)} \tag{32}$$

Transforming this expression to the t-representation one obtains:

$$G^{\|}(t) = G_0 \; exp[-4\pi x\sigma_1 - 4\pi(1-x)\sigma_2] \tag{33}$$

B) Across layers

In the case of charge relaxation relaxation across the layers the current is constant $\vec{J} = const$ and the resistance averages out. Therefore, the effective conductivity of the medum equals:

$$\sigma_{eff} = \left(\frac{x}{\sigma_1 + \frac{i\omega}{4\pi}} + \frac{1-x}{\sigma_2 + \frac{i\omega}{4\pi}} \right)^{-1} \tag{34}$$

while the Green's function is:

$$G^{\perp} = \left(\frac{i\omega + 4\pi[\sigma_1(1-x) + \sigma_2 x]}{(i\omega + 4\pi\sigma_1)(i\omega + 4\pi\sigma_2)} \right) \tag{35}$$

Consequently, after necessary calculations one obtains in the t-representation:

$$G^{\|}(t) = G_0 \Big[x\exp(-4\pi\sigma_1) - (1-)\exp(-4\pi\sigma_2) \Big] \tag{36}$$

2.2.2 Randomly inhomogeneous two-phase medium

In the case of randomly inhomogeneous medium we will use the effective medium approximation to find the frequency dispersion of the conductivity of the medium, obtained by the random mixture of two phases. Two-dimensional system is considered for a simplicity. As well known in this approximation the effective conductivity of this medium is equal to [8]:

$$\sigma_{eff} = (\sigma_1 - \sigma_2)\epsilon + [(\sigma_1 - \sigma_2)^2\epsilon^2 + \sigma_1\sigma_2]^{\frac{1}{2}} \tag{37}$$

where $\epsilon = \frac{(x-x_c)}{x_c}$ is the deviation from the percolation threshold x_c. (In two-dimensional case $x_c = \frac{1}{2}$). As described above in the effective medium aproximation the conductivity of each phase at the frequency ω is described by the expression (28): $\sigma + \frac{i\omega}{4\pi}$. Consequently from (28) and (37) the following expression for the frequency dependence of the effective conductivity of a two-phase medium is obtained:

$$\sigma_{eff}(\omega) = (\sigma_1 - \sigma_2)\epsilon + [(\sigma_1 - \sigma_2)^2\epsilon^2 + \left(\sigma_1 + \frac{i\omega}{4\pi}\right)\left(\sigma_2 + \frac{i\omega}{4\pi}\right)]^{\frac{1}{2}} \tag{38}$$

Let us consider some limitings cases.

A) At the percolation threshold $\epsilon = 0$

In this case the low-frequency generalization of the well known Dykhne result [9] is obtained from (38):

$$\sigma_{eff}(\omega; \epsilon = 0) = [\left(\sigma_1 + \frac{i\omega}{4\pi}\right)\left(\sigma_2 + \frac{i\omega}{4\pi}\right)]^{\frac{1}{2}} \tag{39}$$

So the corresponding Green's function has a following form:

$$< G(t;0) >= I_0(2\pi(\sigma_1 - \sigma_2)t)exp[-2\pi(\sigma_1 + \sigma_2)t] \tag{40}$$

where $I_0(x)$ is a modified Bessel function ($\sigma_2 < \sigma_1$). Using the known asymptotic expression for a Bessel function the long-time asymptotic is obtained:

$$< G(t;0) > \sim \frac{exp(-4\pi\sigma_2 t)}{[(\sigma_1 - \sigma_2)t]^{\frac{1}{2}}} \tag{41}$$

It is the expected result. At large times the time relaxation is determined by the badly conductiving phase.

B) Strongly inhomogeneous medium ($\sigma_2 << \sigma_1$)

In this case we will use the expansion (38) in the form:

$$\sigma_{eff}(\omega; \epsilon) \sim (\sigma_1 - \sigma_2)|\epsilon|(1 \pm 1) + \frac{1}{2}\frac{(\sigma_1 + \frac{i\omega}{4\pi})(\sigma_2 + \frac{i\omega}{4\pi})}{(\sigma_1 - \sigma_2)|\epsilon|} \tag{42}$$

for $\frac{\sigma_2}{\sigma_1} << \epsilon^2 << 1$. It is valid far from percolation threshold and a low frequency limit. In this approximation one obtains in t-representation following expression:

$$< G(t;0)^{\pm} > \sim exp([-4\pi\sigma_2 t - 8\pi\sigma_1|\epsilon|(1\pm)1] - exp(-4\pi\sigma_1 t) \tag{43}$$

where +(-) sign refers to the situation above (below) the percolation threshold. Thus above percolation threshold the charge relaxation time depends on the deviation from threshold: $t_\epsilon \ (\sigma_1|\epsilon|^2)^{-1}$. In the case of the dielectric -metal mixture and below threshold a charge, which placed in the metal cluster, doesn't spread:

$$< \rho(t) > \sim \rho_0[1 - exp(-4\pi\sigma_1 t)] \tag{44}$$

One can expect this result, because the metal cluster has a finite size.

In the general case the averaged Green's function at considered approximation equals:

$$< G(\omega)) > = \left[(\sigma_1 - \sigma_2)\epsilon + [(\sigma_1 - \sigma_2)^2\epsilon^2 + \left(\sigma_1 + \frac{i\omega}{4\pi}\right)\left(\sigma_2 + \frac{i\omega}{4\pi}\right)]^{\frac{1}{2}}\right]^{-1} \tag{45}$$

Using the identity

$$\int_0^\infty \exp(-\alpha\tau)d\tau = \frac{1}{\alpha} \tag{46}$$

one can transform this equation in a more useful intergral form. After integration over ω one calculates the Green's function in the t-represntation:

$$\begin{aligned} < G(t;\epsilon) > \ &= \ I_0\Big(2\pi[\sigma_1 - \sigma_2][1 - 4\epsilon^2]t\Big)\exp[-2\pi(\sigma_1 + \sigma_2)t] - 4\pi\epsilon(\sigma_1 - \sigma_2) \\ &\times \int_{-\infty}^{t} \exp[-4\pi(\sigma_1 - \sigma_2)\epsilon\tau]I_0(2\pi[\sigma_1 - \sigma_2][(1 - 4\epsilon^2)(t^2 - \tau^2)]^{\frac{1}{2}})d\tau \end{aligned} \tag{47}$$

The physical reason, which lead to nonexponential non-Maxwell relaxation in the hetetogeneous many-phase medium, is connected with frequency dispersion of the effective conductivity.

3 Fractional relaxation equations

Let's return to the usual relaxation equation:

$$\frac{\partial\rho(\vec{r},t)}{\partial t} + 4\pi\sigma\rho(\vec{r},t) = 0 \tag{48}$$

One can see that to describe the nonexponential non-Maxwell evolution of charge relaxation it is nessesary to change it. Which ways are possible ? We believe that there are two possibilities of transforming this simple equation in according with physical mechanisms described above.

The first of them - "fractality" of conducting ways. In this case the electric field is present in all space, but charge can be spread only along some conductiving ways. These conducting ways do not get penetrated in all space and they formed the so called "fractal" space less than Euclead space. We call it as "fractality". The similar reason is discussed also for low-dimensional systems [6], [7]. As it was shown above for comb model of percolation clusters it is necessary to write fractional temporal relaxation equation:

$$\left(\frac{\partial^\nu}{\partial t^\nu} + 4\pi\tilde{\sigma}\right)\rho(k,t) = 0 \tag{49}$$

Before the equation in the same form of fractional temporal equation was deduced for regular fractals [10].

For a value $\nu = \frac{1}{2}$ the solution of fractional temporal equation may be written in the obvious form, using the identity (47) :

$$< G(t) >= \int_0^\infty \frac{exp(-\frac{\tau^2}{4t} - 4\tilde{\sigma}t)}{2\sqrt{\pi t}} d\tau \tag{50}$$

At big times the power law asymptotics is followed from the formulae (51):

$$< \rho(t) > \sim \frac{\rho_0}{(\tilde{\sigma}t)^{\frac{1}{2}}} \tag{51}$$

In the case of heterogeneous two-phase medium the change of the relaxation law from exponential to nonexponential nonMaxwell relaxation law is connected with the frequency dispersion of the effective conductivity. Let us emphasize that the initial conductivities of the phases σ_1, σ_2 are independent on frequency. But the frequency dependency arises in the composite material, consisting of random mixture of the phases. At the interface of the phases the "surface" charge appears and when the electric field is changed in time this charge is changed also. It is the reason of the frequency dependency of the effective conductivity $\sigma_{eff}(\omega)$. So the relaxation equation in this case should have the form in the ω-representation:

$$\Big(i\omega + 4\pi\sigma_{eff}(i\omega)\Big)\rho(\omega) = 0 \tag{52}$$

Near percolation threshold [11] the singular power dependency of effective conductivity σ_{eff} on frequency ω is possible:

$$\sigma_{eff}(\omega) = \sigma(\omega)^\mu \tag{53}$$

where $o < \mu < 1$

Consequently, another fractional temporal relaxation equation is obtained:

$$\Big(\frac{\partial}{\partial t} + 4\pi\check{\sigma}\frac{\partial^\mu}{\partial t^\mu}\Big)\rho(r,t) = 0 \tag{54}$$

One can obtain the fractional-exponential Karlraushe law (5) from (53) using the saddle point method.

So it is shown that two types of the generalized relaxation equations in the form of the fractional temporal equations are possible. They correspond to two physical mechanisms, which lead to the nonexponential non-Maxwell relaxation.

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