Charge relaxation in layered and randomly inhomogeneous two-phase materials in the effective-medium approximation

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Charge relaxation in layered structures is investigated, and a probabilistic interpretation of the results is given. The charge spreading problem in a randomly inhomogeneous two-phase medium is solved for arbitrary phase concentrations. It is shown on the basis of the scaling hypothesis that in strongly inhomogeneous media and near the percolation threshold the charge relaxes in a power-law manner in both the two- and three-dimensional cases.

1. INTRODUCTION

The variation of the nature of relaxation of an excess charge density ρ in inhomogeneous media, namely a 2D two-phase medium near the percolation threshold (with equal phase concentrations) in a comb structure, has been investigated in Ref. 1. It has been shown that the relaxation time in a randomly inhomogeneous medium with equal phase concentrations is determined by the conductivity of the poorly conducting phase, while in a metal-dielectric mixture the charge relaxes according to a power law. Charge spreading on a comb structure also has a non-Maxwellian character.

The purpose of the present work is further investigation of charge relaxation in inhomogeneous media. Layered and randomly inhomogeneous materials are considered in the effective-medium approximation. The space dimensionality is unimportant for this approximation. A generalization of the Maxwellian law is obtained for the case of layered structures, and a probabilistic interpretation of these results is given. The character of charge relaxation is established for a two-phase medium with arbitrary phase concentrations. This character differs substantially from Maxwellian in the general case. It is also shown on the basis of the scaling hypothesis that is strongly inhomogeneous (metal–dielectric) media the charge relaxes by a power law in the 2-D and 3-D cases.

Charge relaxation in a conducting medium is described by the system of equations

$$\frac{\partial \rho}{\partial t}+\operatorname{div}\mathbf{j}=0, \quad \operatorname{div}\mathbf{e}=4\pi\rho, \quad \mathbf{j}=\sigma\mathbf{e}. \tag{1}$$

The vectors **j** and **e** are the electric current and field, respectively. The permittivity of the medium is assumed equal to unity. This system can be transformed into equations for a constant current

$$\operatorname{div}\mathbf{j}=0, \quad \mathbf{j}=\left(\sigma+\frac{i\omega}{4\pi}\right)\mathbf{e}. \tag{2}$$

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

$$\sigma(\omega)=\sigma+i\omega/4\pi. \tag{3}$$

Similar equations also hold in an inhomogeneous medium for the averaged quantities $\mathbf{J}=\langle\mathbf{j}\rangle$ and $\mathbf{E}=\langle\mathbf{e}\rangle$:

$$\operatorname{div}\mathbf{J}=0, \quad \operatorname{div}\mathbf{E}=4\pi\langle\rho\rangle, \quad \mathbf{J}=\sigma_{eff}\mathbf{E}. \tag{4}$$

The effective conductivity of the medium σ_{eff} depends on both the frequency and the medium parameters. We emphasize that in this approach all the information concerning the medium inhomogeneity is contained in the effective conductivity.

Thus, according to (4), to describve charge relaxation in an inhomogeneous medium within the approximation considered it is sufficient to know the frequency dependence of its effective conductivity.

2. LAYERED STRUCTURES

Consider inhomgeneous structures obtained by random alternation of layers with different conductivities σ_1 and σ_2. The law of relaxation along the layers of the structure is determined quite simply. In this case the field is homogeneous, and the averaging is carried out easily:

$$\sigma_{eff}=\langle\sigma\rangle+\frac{i\omega}{4\pi}, \tag{5}$$

where $\langle\sigma\rangle=x\sigma_1+(1-x)\sigma_2$, and x is the concentration of the first phase. An equation is correspondingly obtained for the averaged concentration:

$$(i\omega+4\pi\langle\sigma\rangle)\langle\rho\rangle=0. \tag{6}$$

The Green's function of Eq. (6) is

$$G^{\parallel}(\omega)=(i\omega+4\pi\langle\sigma\rangle)^{-1}. \tag{7}$$

Transforming to the t-representation, we obtain

$$G^{\parallel}(t)=\exp\left[-4\pi\sigma_1 xt-4\pi\sigma_2(1-x)t\right]. \tag{8}$$

During charge spreading across the layered structure the current is constant and the resistance averages out. In this case, therefore, the effective-medium conductivity equals

$$\sigma_{eff}=\left(\frac{x}{\sigma_1+i\omega/4\pi}+\frac{1-x}{\sigma_2+i\omega/4\pi}\right)^{-1}, \tag{9}$$

while the Green's function is

$$G^{\perp}(\omega)=\frac{i\omega+4\pi[\sigma_1(1-x)+\sigma_2 x]}{(i\omega+4\pi\sigma_1)(i\omega+4\pi\sigma_2)}. \tag{10}$$

For computational convenience we represent expression (10) in the form

$$G^{\perp}(\omega)=\frac{i\omega+2\pi(\sigma_1+\sigma_2)+2\pi(\sigma_1-\sigma_2)(1-2x)}{[i\omega+2\pi(\sigma_1+\sigma_2)]^2-[2\pi(\sigma_1-\sigma_2)]^2}, \tag{11}$$

in which case we obtain in the t-representation

$$G^{\perp}(t)=x\exp(-4\pi\sigma_1 t)+(1-x)\exp(-4\pi\sigma_2 t). \tag{12}$$

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The results (8) and (12) have the following probabilistic interpretation. We denote the concentration decrease due to relaxation in the first phase by G_1, and in the second phase by G_2. Charge spreading along the conducting layers in the first and second phases can be treated as two independent events; therefore, by the theorem of multiplying independent events we have

$$G^{\parallel}(t)=G_1^{x}G_2^{(1-x)}. \tag{13}$$

Differently stated, the logarithm of the concentrations is averaged in this case.

According to (12), during charge relaxation across the layers the concentration decreases due to spreading over the first or second phase:

$$G^{\perp}(t)=xG_1+(1-x)G_2, \tag{14}$$

i.e., the concentration itself is averaged, while the decrease in charge density is described by the theorem of probability addition.

The expressions obtained by us for charge relaxation are valid for 3-D structures with 2-D dependence (nothing depends on the coordinate z). For charge spreading only in a plane and in a 3D field, relaxation has a non-Maxwellian character even in the homogeneous case.[2]

3. RANDOMLY INHOMOGENEOUS TWO-PHASE MEDIA

To find the frequency dependence of a two-phase medium with arbitrary phase concentrations we use the effective-medium approximation. For definiteness we consider the 2-D case. As well known, in this approximation the effective-medium conductivity is[3]

$$\sigma_{eff}=(\sigma_1-\sigma_2)\varepsilon+[(\sigma_1-\sigma_2)^2\varepsilon^2+\sigma_1\sigma_2]^{1/2}, \tag{15}$$

where $\varepsilon=(x-x_c)/x_c$ is the deviation from the percolation threshold $x_c=\frac{1}{2}$.

As noted above, the conductivity of each of the phases at the frequency ω is described by the expression $\sigma+i\omega/4\pi$. Consequently, from (3) and (15) we obtain the following expression for the frequency dependence of the effective conductivity of a two-phase medium:

$$\sigma_{eff}(\omega)=(\sigma_1-\sigma_2)\varepsilon+\left[(\sigma_1-\sigma_2)^2\varepsilon^2+\left(\sigma_1+\frac{i\omega}{4\pi}\right)\left(\sigma_2+\frac{i\omega}{4\pi}\right)\right]^{1/2}. \tag{16}$$

Correspondingly, the Green's function averaged over the randomly arranged phases equals

$$G=\left\{\left[(\sigma_1-\sigma_2)^2\varepsilon^2+\left(\sigma_1+\frac{i\omega}{4\pi}\right)\left(\sigma_2+\frac{i\omega}{4\pi}\right)\right]^{1/2}+(\sigma_1-\sigma_2)\varepsilon\right\}^{-1}. \tag{17}$$

Using the identity

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

following simple transformations and integration over ω we calculate the Green's function in the t-representation:

$$G(t)=\exp[-2\pi(\sigma_1+\sigma_2)t]\{I_0(2\pi[\sigma_1-\sigma_2][1-4\varepsilon^2]^{1/2})-4\pi\varepsilon(\sigma_1-\sigma_2)\int_0^{t}\exp[-4\pi(\sigma_1-\sigma_2)\varepsilon\tau]I_0(2\pi[\sigma_1-\sigma_2]\times[(1-4\varepsilon^2)(t^2-\tau^2)]^{1/2})\,d\tau, \tag{18}$$

where $I_0(x)$ is the modified Bessel function.

We stress that Eq. (18) describes charge relaxation in a two-phase medium for arbitrary phase concentrations. Near the percolation threshold $\varepsilon=0$ expression (18) transforms to the corresponding expression of Ref. 1, obtained by a different method, by means of the general Dykhne approach. For $\varepsilon=\pm\frac{1}{2}$ one obtains, as expected, a homogeneous medium with the usual Maxwellian relaxation law.

Equation (18) is quite unwieldy, and therefore it is interesting to consider some of its limits, more precisely the spreading above and below the percolation threshold in a strongly inhomogeneous medium ($\sigma_2\ll\sigma_1$). For this we use expansion (16) in the form

$$\sigma_{eff}(\omega)\sim(\sigma_1-\sigma_2)|\varepsilon|(1\pm1)+\frac{1}{2}\frac{(\sigma_1+i\omega/4\pi)(\sigma_2+i\omega/4\pi)}{(\sigma_1-\sigma_2)|\varepsilon|} \tag{19}$$

for $\sigma_2/\sigma_1\ll\varepsilon^2\ll1$. Equation (19) is valid at low frequencies, as well as far enough from the percolation threshold. In this approximation the Green's function is

$$G\sim\frac{8\pi(\sigma_1-\sigma_2)|\varepsilon|}{(i\omega+4\pi\sigma_1)(i\omega+4\pi\sigma_2)+2[4\pi(\sigma_1-\sigma_2)\varepsilon]^2(1\pm1)}, \tag{20}$$

i.e., in the t-representation one has

$$G^{\pm}(t)\propto\{\exp[-4\pi\sigma_2 t-8\pi\sigma_1\varepsilon^2(1\pm1)t]-\exp(-4\pi\sigma_1 t)\}, \tag{21}$$

where the $+$ ($-$) sign refers to the situation above (below) the percolation threshold.

Thus, above the percolation threshold the charge relaxes with a time $t_\varepsilon\sim1/\sigma_1\varepsilon^2$ that depends on the proximity to the percolation threshold. Below the percolation threshold and in the dielectric–metal mixture a chrage placed in the metallic region remains in it:

$$\rho(t)\sim\rho_0[1-\exp(-4\pi\sigma_1 t)]. \tag{21'}$$

4. STRONGLY INHOMOGENEOUS (METAL–DIELECTRIC) MEDIA AT THE PERCOLATION THRESHOLD

We treat relaxation in the 3-D case on the basis of the scaling hypothesis. According to this hypothesis the effective conductivity of a strongly inhomgeneous medium near the percolation threshold can be represented in the self-similar form

$$\sigma_{eff}=\sigma_1 h^S f(\varepsilon/h^{S/T}), \tag{22}$$

where the parameter $h=\sigma_2/\sigma_1\ll1$ is the ratio of the phase conductivities, and the quantity ε is the deviation from the percolation threshold. The asymptotic behavior of the function f is described by the exponents S, T, and Q of percolation theory:

$$f(z)=\begin{cases}|z|^{-Q}, & z\ll-1\\ 1, & |z|\ll1.\\ z^T, & z\gg1\end{cases} \tag{23}$$

The exponents S, T, and Q are connected by the exact relationship

$$T(1/S-1)=Q. \tag{24}$$

Using (3) and the asymptotic behavior of (22), we obtain the frequency dependence of the effective conductivity of a strongly inhomogeneous medium near the percolation threshold:

$$\sigma_{eff}(\omega)\propto\sigma_1^{(1-S)}(i\omega)^S. \tag{25}$$

The dependence (25) was also established in Ref. 4. Calculations similar to those performed above yield the charge relaxation law in metal–dielectric mixtures:

$$\rho(t)\propto\rho_0/(\sigma_1 t)^{1-S}. \tag{26}$$

In the 2D case Eq. (26) is confirmed by exact solution of the problem [1], while the exponent value $S=\frac{1}{2}$ was established in Ref. 5. A possible exponent value $S=\frac{2}{3}$ for the 3-D case is indicated in Ref. 6.

The power-law relaxation in a metal–dielectric mixture is a consequence of the absence of characteristic dimensions in the problem, since the correlation radius is infinite near the percolation threshold, and metallic phase inclusions are possible for all dimensions along which charge spreading occurs.

In conclusion, the following qualitative pattern can be given in strongly inhomogeneous media near the percolation threshold, as follows from our results. At short times $t\ll t_\varepsilon$, when the charge does not emerge beyond a unit cell of the size of the correlation radius, the charge spreads in power-law fashion. At later times, relaxation above the percolation threshold is exponential with a time t_ε that depends on the proximity to the percolation threshold, while below the percolation threshold only polarization of the medium takes place in a dielectric phase with metallic inclusions.

5. DISCUSSION

Thus, in the present article we have treated charge relaxation within the effective-medium approximation. It might seem that if the effective conductivity σ_{eff} of an inhomogeneous medium at zero frequency is known the answer must have a Maxwellian form: $\rho\sim\rho_0\exp(-4\pi\sigma_{\text{eff}}t)$. In real situations, as shown above, this is not the case.

The effective-medium method is widely used to describe inhomogeneous media. It yields, for example, the correct qualitative behavior for the effective conductivity of a two-phase system at arbitrary phase concentrations.[3] In the 2D case and near the percolation threshold the exact solution of the conductivity problem[5] demonstrates the validity of this approximation. Therefore, it can be expected that the effective-medium method also describes correctly the basic features of charge spreading in inhomogeneous media.

Let us discuss the results and the validity of the effective-medium approximation. In the simplest case of an inhomogeneous medium consisting of two half-planes with different conductivities (in a bicrystal) the exact solution of the problem coincides with the solution obtained by the effective-medium method. Relaxation along the boundary of the two phases is described by expression (8), while the spreading across the layers is described by Eq. (12) at a phase concentration $x=\frac{1}{2}$.

The decrease of the excess charge density in a percolation-type randomly inhomogeneous medium ($\sigma_2\ll\sigma_1$) above and below the percolation threshold, as well as at the threshold itself, can be explained as follows. Below the percolation threshold the entire charge in a dielectric phase with metallic inclusions goes from the metallic regions to the boundary, and therefore it is sufficient to estimate the charge variation with time at the boundary. The conservation law for the surface charge ρ_s is

$$\frac{\partial\rho_s}{\partial t}=j=\sigma(E-4\pi\rho_s). \tag{27}$$

Consequently, for charge relaxation below the threshold we have

$$\rho=\rho_0[1-\exp(-4\pi\sigma_1 t)], \tag{28}$$

where $\rho_0=E/4\pi$. This corresponds to the result (21′).

Above the percolation threshold charge spreads over an infinite metallic cluster (a set of percolating paths departing to infinity). Following a single time interval τ the charge in a conducting medium decreases by an amount $\sigma\tau\rho_0$, and accordingly after N steps we obtain an exponential decrease of the charge density:

$$\rho\sim\rho_0(1-\sigma\tau)^N\sim\rho_0\exp(-\sigma\tau N). \tag{29}$$

The number of steps N is proportional to the time t, but since the percolation paths are tortuous the charge spreads more slowly in a percolating system. To reach in a Euclidean space a distance R it is necessary to trace a tortuous percolation path of larger length $\mathscr{L}$. Due to the scale invariance of the system both lengths are measured in units of the correlation radius $L_c\approx|\varepsilon|^{-\nu}$. Therefore the number of steps is

$$N\sim tf(\varepsilon), \tag{30}$$

where the information about the structure of the percolation paths is contained in the functional dependence on the proximity to the percolation threshold, i.e., in $f(\varepsilon)$. In a two-phase medium $f(\varepsilon)=\varepsilon^2$.

The power-law behavior in a metal-dielectric mixture is also easily explained. Near the percolation threshold the correlation radius is infinite, and therefore metallic phase inclusions of all dimensions are possible. The absence of characteristic dimensions in the problem also leads to a power-law charge relaxation. We note also that for regular fractals of Serpinskii parquet type a similar power-law behavior was independently established in Ref. 7 by means of normalization-group transformations. (Metal–dielectric percolation systems are associated with random fractals.)

Thus, the estimates performed verify the validity of the effective-medium method in describing charge relaxation in inhomogeneous media. It is also noted that with increasing time the charge occupies an ever increasing part of the space, and the averaging is over ever increasing scales. Consequently, the results obtained within the effective-medium approximation are asymptotically exact in the limit of long times.

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