Nonlinear effects in the high-frequency electric conductivity of two- and three-dimensional polycrystals

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The frequency dispersion of the electric conductivity of a polycrystal in a strong electric field is determined by a method which is a generalization of the classical percolation approach previously proposed [B. I. Shklovskii and A. L. Efros, Electronic Properties of Doped Semiconductors, Springer, 1984] to determine the static hopping conductivity. It is shown that the spatial distribution of the current fluctuations in the polycrystal forms a percolation structure with a frequency dependent correlation radius and with a modulation depth $\Delta^{t/s}$, where Δ is the variance of the heights of the intercrystalline barriers, and t and s are the critical exponents of the electric conductivity. For strong electric fields, a frequency-dependent nonlinearity criterion is obtained and the field dependence of the electric conductivity is plotted for the entire essential frequency range in an arbitrary number of dimensions.

1. INTRODUCTION

A simple model of a polycrystal was proposed in Ref. 2 for the case when the screening radius is much smaller than the average crystallite dimension and the electric conductivity is limited by intergranular barriers whose heights are distributed over a rather wide interval. In this model a polycrystal is set in correspondence in the ohmic regime with a regular lattice of dimensionality d, whose sites correspond to the internal regions of the crystallites, and the bonds correspond to the intercrystallite barriers modeled by conductivities of the form

$$\sigma=\sigma_0 e^{-\mu\Delta}+i\omega c_0, \tag{1}$$

where σ_0 is the conductivity of the inner part of the microcrystal, Δ is the variance of the intercrystalline barriers in units of the temperature T, μ is a random quantity uniformly distributed in the interval $\{0,1\}$, c_0 is the barrier capacitance (whose fluctuations we neglect), and ω is the frequency of the external electric field.

This model was used to analyze consistently the static conductivity in weak and strong electric fields,[2,3] the frequency dispersion of the conductivity in weak electric fields,[4] and to consider in addition nonlinear effects in the conductivity of a one-dimensional polycrystal.[5] What remained uninvestigated was the conductivity of two- and three-dimensional polycrystals in strong alternating electric fields. The quasi-equilibrium finite-cluster approximation,[6,7] which yields the dispersion of the conductivity in the ohmic regime, cannot be used for strong alternating electric fields. The point is that in such fields it is difficult to introduce a connectivity criterion that determines the size of the quasi-equilibrium cluster, since the barrier heights become dependent on the voltage drops across them.

A novel method is therefore proposed here for the calculation of the frequency dispersion of the conductivity. It differs from the quasi-equilibrium clusters and is essentially a direct generalization, to include alternating electric fields of any intensity, of the classical percolation approach proposed by Shklovskiĭ and Éfros,and also by Ambegaokar, Halperin, and Langer, for the solution of the problem of static hopping conduction.[8,9] The approach proposed here is based on an analysis of the spatial distribution of the current fluctuations in a polycrystal.

2. CONNECTION BETWEEN THE PERCOLATION APPROACH AND THE SPATIAL DISTRIBUTION OF CURRENT FLUCTUATIONS IN A POLYCRYSTAL

A prominent place in disordered systems is occupied by metal–dielectric mixtures consisting of two components having substantially different conductivities, σ_m of the metallic phase and σ_d of the dielectric. The usual method of modeling such randomly inhomogeneous objects is to study the conductivities of lattices with branches having random conductivities and distributed independently of one anther, with a density

$$P(\sigma)=x\delta(\sigma-\sigma_m)+(1-x)\delta(\sigma-\sigma_d). \tag{2}$$

The main feature of such a percolation system is the presence of a characteristic spatial scale—the correlation length $L(x-x_c)$, which can exceed by many times the minimum spatial scale a that is equal to the lattice period. The correlation radius is particularly large in a small vicinity of the concentration percolation threshold x_c—in the scaling region in which the metal–dielectric transition takes place. Since $L(x-x_c)$ is in this region the only parameter having the dimension of length and determining the concentration dependences, the conductivity of a metal–dielectric mixture can be described by the complex scaling function $\Psi(z)$ introduced in Ref. 10:

$$\sigma=\sigma_m h^s \Psi\left(\frac{x-x_c}{h^{s/t}}\right). \tag{3}$$

Here $h=\sigma_d/\sigma_m$ is the analog of the magnetic field in phase-transition theory, $\Psi(z)$ as a function of the complex variable z has as its asymptotes

$$\Psi(z)=\begin{cases} z^{-q}, & |z|\gg 1, \quad \pi-\dfrac{s}{t}\dfrac{\pi}{2}\leqslant \arg z\leqslant \pi+\dfrac{s}{t}\dfrac{\pi}{2}, \\ z^{t}, & |z|\gg 1, \quad -\dfrac{s}{t}\dfrac{\pi}{2}\leqslant \arg z\leqslant \dfrac{s}{t}\dfrac{\pi}{2}, \end{cases}$$

and the critical exponents q, s, and t satisfy the known rela-

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tion $q = t/s - t$, while their numerical values depend on the dimensionality of space.

The problem of the conductivity of a polycrystalline semiconductor has at first glance nothing in common with the structure of the percolation problem. The distribution density of the conductivities (1) differs from the density (2) primarily because the former does not contain a parameter on which the polycrystal conductivity can have a threshold dependence. The problem of the conductivity of a polycrystal has therefore likewise no correlation length having a singular dependence on this parameter. This means that to use percolation theory to determine the conductivity of a polycrystal it is necessary to introduce a parameter analogous to the metallic-phase concentration x in Eq. (2).

This can be done by calculating the distribution of the current fluctuations in the polycrystal by the effective-medium method. We single out an arbitrary branch having a conductivity σ and determine the current I flowing through it under the assumption that the remainder of the lattice is a homogeneous medium of conductivity g. Since this problem has been solved in detail by Kirpatrick,[11] we take the liberty of presenting directly the result

$$I=\langle V\rangle d\frac{\sigma g}{\sigma+(d-1)g}. \tag{4}$$

Here $\langle V\rangle = \langle E\rangle a$ is the average voltage drop on a length a,

$$g=\sigma_0 e^{-p\Delta}+i\omega c, \tag{5}$$

and the parameters p and c represent the real and imaginary parts of the conductivity of the homogeneous medium. Since μ is random, it follows that by fixing p and c we set each random distribution of the conductivities σ in correspondence with some distribution of the currents I; we proceed now to study this distribution. We begin with the static limit $\omega = 0$ and set the initial lattice in correspondence with an "effective" one having conductivities

$$\sigma^{\text{eff}}=\sigma_0 d e^{-p\Delta}\bar{\sigma}, \tag{6}$$

where

$$\bar{\sigma}=\frac{1}{1+(d-1)e^{(\mu-p)\Delta}} \tag{7}$$

is a dimensionless random quantity having a distribution density

$$P(\bar{\sigma})=\int_0^1 \delta\left(\bar{\sigma}-\frac{1}{1+(d-1)e^{(\mu-p)\Delta}}\right)d\mu. \tag{8}$$

Of greatest interest is the leading term of the expansion of $P(\bar{\sigma})$ in terms of Δ^{-1}, which is obtained by going in it to the limit $\Delta\to\infty$:

$$P_0(\bar{\sigma})=p\delta(\bar{\sigma}-1)+(1-p)\delta(\bar{\sigma}). \tag{9}$$

Comparison of (9) and (2) shows that the effective lattice introduced by us is a percolation system with conductivities $\sigma_m = \sigma_0 d e^{-p\Delta}$ and $\sigma_d = 0$. The parameter p plays the role of the density of the "metallic" phase that characterizes the spatial distribution of current fluctuations with amplitude $I\sim e^{-p\Delta}$. The current fluctuations can therefore be described by a correlation length that depends on the logarithm of the amplitude of these fluctuations.

This raises the question: how are the conductivities of the initial and effective lattices connected? To obtain the answer it suffices to note that Eq. (4) differs only by a constant factor from the equation for a series connection of conductors having the conductivities of a random element and of an homogeneous medium. The construction of an effective lattice from the initial one reduces therefore to adding a limiting resistor to each branch. Obviously, this procedure can only decrease the conductivity of each branch and of the system as a whole. The conductivity of the effective lattice can therefore be regarded as the lower-bound of the conductivity of the polycrystal. The conductivity of the effective lattice as a function of the parameter p, however, has an abrupt maximum

$$\langle\sigma^{\text{eff}}(p)\rangle=\begin{cases}\sigma_0 d e^{-p\Delta}(p-p_c)^t, & p>p_c, \quad (p-p_c)/p_c\ll 1,\\ 0, & p<p_c,\end{cases} \tag{10}$$

This means that the following estimate is valid for the polycrystal conductivity of interest to us:

$$\langle\sigma\rangle\geqslant\sigma_0 d\left(\frac{t}{\Delta}\right)^t\exp(-p_c\Delta-t). \tag{11}$$

The stationary distribution of the current has here a percolation character and can be described by a correlation length $L_c\sim a\Delta^{\nu}$ (ν is the critical exponent of the correlation radius).

3. RENORMALIZATION GROUP OF THE EFFECTIVE MEDIUM AND FREQUENCY DISPERSION OF THE POLYCRYSTAL CONDUCTIVITY

We proceed now to analyze the nonstationary situation ($\omega > 0$). To simplify the procedure, we introduce a dimensionless parameter $u = -\Delta^{-1}\ln(\omega\tau)$ (where $\tau = c_0/\sigma_0$) that varies in the range $0 < u < 1$ when the frequency of the external field runs through the interval $e^{-\Delta} < \omega\tau < 1$. In the new notation, the quantities of interest to us take the form

$$\sigma=\sigma_0(e^{-\mu\Delta}+ie^{-u\Delta}), \tag{12}$$

$$g=\sigma_0(e^{-p\Delta}+ice^{-u\Delta}), \tag{13}$$

$$\sigma^{\text{eff}}=d\frac{\sigma g}{\sigma+(d-1)g}=\sigma_0 d(\bar{\sigma}e^{-p\Delta}+i\bar{c}e^{-u\Delta}). \tag{14}$$

The parameter $\bar{c}$ has the meaning of the capacitance of an element of the effective medium.By analogy with the static case, we calulate the distribution densities of the real and imaginary parts of the conductivities of the effective-lattice elements

$$P(\bar{\sigma},\bar{c})=\int_0^1\delta[\bar{\sigma}-\bar{\sigma}(p,u,\mu,\Delta)]\delta[\bar{c}-\bar{c}(p,u,\mu,\Delta)]d\mu. \tag{15}$$

The functions $\bar{\sigma}(p,u,\mu,\Delta)$ and $\bar{c}(p,u,\mu,\Delta)$ are determined from relations (12)–(14), and just as in the static case we are interested here in the leading term of the expansion of the distribution density in powers of Δ^{-1}:

$$P_0(\bar{\sigma},\bar{c})=\lim_{\Delta\to\infty}P(\bar{\sigma},\bar{c}).$$

Obviously, the result of this limiting transition depends strongly on the relation between p and u. Simple calculation shows that for $p < u$

$$P_0(\bar\sigma,\bar c)=p\delta(\bar\sigma-1)\delta(\bar c-1)+(1-p)\delta(\bar\sigma)\delta\left(\bar c-\frac{1}{d-1}\right), \tag{16}$$

for $p=u$

$$P_0(\bar\sigma,\bar c)$$

$$=p\delta(\bar\sigma-1)\delta(\bar c-c)+(1-p)\delta\left(\bar\sigma-\frac{1}{(d-1)^2+[1+(d-1)c]^2}\right)$$

$$\times\delta\left(\bar c-\frac{(d-1)(1+c^2)+c}{(d-1)^2+[1+(d-1)c]^2}\right), \tag{17}$$

and for $p>u$

$$P_0(\bar\sigma,\bar c)=\{\theta(2u-p)[(2u-p)\delta(\bar\sigma-1)+(p-u)\delta(\bar\sigma-\infty)]+\theta(p-2u)u\delta(\bar\sigma-\infty)\}\delta(\bar c-c)+[(p-u)\delta(\bar\sigma-\infty)+(1-p)\delta(\bar\sigma)]\delta\left(\bar c-\frac{c}{1+(d-1)c}\right), \tag{18}$$

where $\theta(x)$ is the Heaviside step function.

Let us assume that the spatial distribution (16)–(18) of the conductivities can be characterized by a correlation length, and let us try to find the $L(p,u)$ dependence. To this end we consider the renormalization approximation of the distribution density $P(\sigma,\bar c)$, which has the meaning of averaging over the small-scale fluctuations under the scale transformation $L\to bL$, where $b>1$:

$$P_{n+1}(\bar\sigma,\bar c)=\hat W\{P_n(\bar\sigma,\bar c)\}. \tag{19}$$

The operator $\hat W$ reduces to averaging of the real and imaginary parts of the conductivity $G(\sigma_1^{\rm eff},\ldots,\sigma_N^{\rm eff})$ of the transformed cell (N is the number of elements in the cell).[12] An explicit form of this transformation can be obtained from the obvious relation

$$\bar\sigma e^{-p\Delta}+i\bar c e^{-u\Delta}=G[(\bar\sigma_1 e^{-p\Delta}+i\bar c_1 e^{-u\Delta}),\ldots,(\bar\sigma_N e^{-p\Delta}+i\bar c_N e^{-u\Delta})]$$
$$=e^{-p\Delta}\,\mathrm{Re}\,G[(\bar\sigma_1+i\lambda\bar c_1),\ldots,(\bar\sigma_N+i\lambda\bar c_N)]$$
$$+ie^{-u\Delta}\,\mathrm{Im}\,G[(\lambda^{-1}\bar\sigma_1+i\bar c_1),\ldots,(\lambda^{-1}\bar\sigma_N+ic_N)], \tag{20}$$

where $\lambda=e^{(p-u)\Delta}$. This is legitimate because the conductivity of the cell, regardless of its shape and dimensionality, is the ratio of two homogeneous functions, and the degree of the numerator is greater by unity than that of the denominator. It follows from (20), however, that the transformation (19) depends on the parameter λ, which has different limiting values as $\Delta\to\infty$, depending on the relation between p and u,

$$P_{n+1}(\bar\sigma,\bar c)$$

$$=\lim_{\Delta\to\infty}\int d\bar\sigma_1\,d\bar c_1\ldots d\bar\sigma_N\,d\bar c_N P_n(\bar\sigma_1,\bar c_1)\ldots P_n(\bar\sigma_N,\bar c_N)\delta\{\bar\sigma-\mathrm{Re}\,G[(\bar\sigma_1+i\lambda\bar c_1),\ldots,(\bar\sigma_N+i\lambda\bar c_N)]\}\delta\{\bar c-\mathrm{Im}\,G[(\lambda^{-1}\bar\sigma_1+i\bar c_1),\ldots,(\lambda^{-1}\bar\sigma_N+i\bar c_N)]\}. \tag{21}$$

To make the reasoning that follows more lucid, we turn to Fig. 1, which shows the region in which the functions $\langle\bar\sigma(p,u)\rangle\langle\bar c(p,u)\rangle$ of interest us to are defined. In the part of the square $OFCD$ above the diagonal OC we have $p<u$ and a large Δ the imaginary part of the current is exponentially small compared with the real. In this region we should therefore be interested only in the distribution density of the real part of the effective conductivity

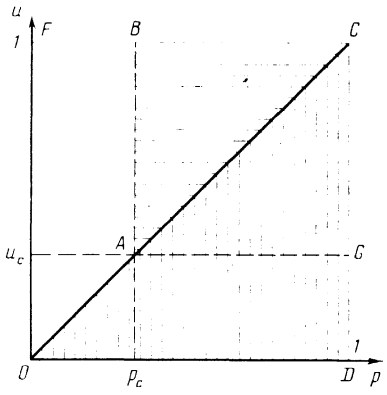

FIG. 1. Regions in which the real and imaginary parts of the effective conductivity are defined. The hatches indicate the directions in which $\mathrm{Re}\langle\sigma^{\rm eff}(p)\rangle$ changes in the upper half of the squares and $\mathrm{Im}\langle\sigma^{\rm eff}(u)\rangle$ in the lower half. The line AB and AG indicate the position of the singularity of $\langle\sigma^{\rm eff}(p,u)\rangle$.

$$P(\bar\sigma)=\int P(\bar\sigma,\bar c)\,d\bar c,$$

since it is just what we need to calculate the average conductivity

$$\langle\bar\sigma\rangle=\lim_{n\to\infty}\langle\bar\sigma_n\rangle$$
$$=\lim_{n\to\infty}\int\bar\sigma P_n(\bar\sigma,\bar c)\,d\bar c\,d\bar\sigma.$$

The structure of the transformation (21), however, is such that in this region of variation of the parameters p and u the density $P(\bar\sigma)$ is independently transformed in the limit $\Delta\to\infty$. This circumstances can be easily seen by putting $\lambda=0$ (for $p<u$ and $\Delta\to\infty$) and integrating (21) with respect to c. We obtain then

$$P_{n+1}(\bar\sigma)=\int d\bar\sigma_1\ldots d\bar\sigma_N P_n(\bar\sigma_1)\ldots P_n(\bar\sigma_N)\delta[\bar\sigma-G(\bar\sigma_1,\ldots,\bar\sigma_N)]. \tag{22}$$

The initial distribution density $P_0(\sigma)$ is obtained by integrating expressions (16) with respect to c

$$P_0(\bar\sigma)=p\delta(\bar\sigma-1)+(1-p)\delta(\bar\sigma) \tag{23}$$

and describes a percolation structure with a correlation radius $L_c\sim a|p_c-p|^{-\nu}$. The dependence of $\mathrm{Re}\langle\sigma^{\rm eff}(p)\rangle$ on p is given by Eq. (10).

Below the diagonal OC the real part of the current is exponentially small compared with the imaginary. Just as in the preceding case, we consider the distribution density of the imaginary part of the effective-medium conductivity

$$P(\bar c)=\int P(\bar\sigma,\bar c)\,d\bar\sigma,$$

which is needed to calculate

$$\langle c\rangle=\lim_{n\to\infty}\langle c_n\rangle=\lim_{n\to\infty}\int\bar c P_n(\bar\sigma,\bar c)\,d\bar\sigma\,d\bar c.$$

For $p>u$ the parameter $\lambda=\infty$ in the limit as $\Delta\to\infty$, and averaging over $\bar\sigma$ greatly simplifies the transformation (21):

$$P_{n+1}(\bar c)=\int d\bar c_1\ldots d\bar c_N P_n(\bar c_1)\ldots P_n(\bar c_N)\delta[\bar c-G(\bar c_1\ldots\bar c_N)]. \tag{24}$$

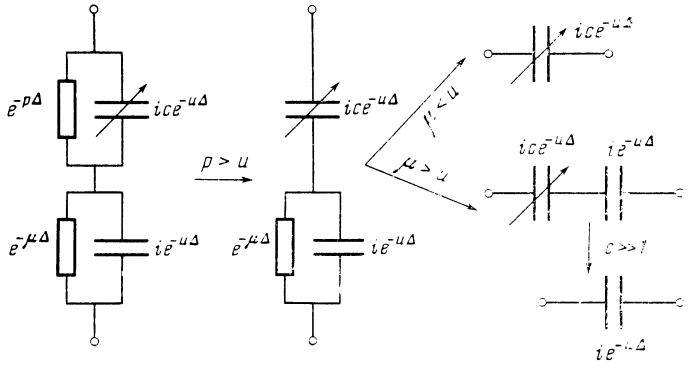

FIG. 2. Sequence of simplifications of the equivalent circuit of an element of an effective random medium for $\Delta \gg 1$ and $p > u$. Discarding the exponentially small terms of the imaginary part reveals in the effective lattice two "fractions": branches with concentration $(1-u)$ and with unity capacitance.

The initial distribution density can be obtained by averaging the distribution (18) over $\bar{\sigma}$:

$$P_0(\bar{c}) = u\delta(\bar{c}-c) + (1-u)\,\delta\left(\bar{c} - \frac{c}{1+(d-1)c}\right). \qquad (25)$$

If $c \gg 1$ this density describes a percolation system with radically differing component conductivities $\sigma_m = c$ and $\sigma_d = 1/(d-1)$. The concentration ratio of the components with σ_m and σ_d depends on the frequency.

To interpret Eq. (25), we must turn to Fig. 2, which shows a successive chain of simplifications of the diagram of an element of the effective medium by discarding conductivities whose contribution to the imaginary part of the current is exponentially small at the given combination of parameters ($p > u$, $c \gg 1$). It can be seen that the phase with the concentration u and with the conductivity c contains those initial-lattice elements where the relaxation frequency Γ exceeds the external-field frequency ω. This means that during the entire period $2\pi/\omega$ the microcrystals that make contact through such barriers are in equilibrium, i.e., they form a "quasi-equilibrium cluster." The connectivitiy condition that determines the spatial distribution of these clusters is exactly the same as obtained in Ref. 4 from physical considerations.

Our theory corroborates thus the method of quasi-equilibrium finite clusters. The electric conductivity of a metal–dielectric mixture with a distribution density (25) can be expressed with the aid of the scaling function $\Psi(z)$ defined in Eq. (3):

$$\mathrm{Im}\langle\sigma^{\mathrm{eff}}(u)\rangle = \sigma_0 \frac{d}{(d-1)^s} c^{1-s} e^{-u\Delta} \Psi\{(u-u_c)[(d-1)c]^{s/t}\}. \qquad (26)$$

The correlation radius of such a random system is obviously a function of the parameter u, i.e., of the frequency

$$L(\omega) \approx a\Delta^{\nu} \ln^{-\nu}(\omega/\omega_c).$$

When $p = u$ the distribution density (17) describes a two-component mixture with metallic- and dielectric-phase concentrations p and $1-p$, respectively, and with complex conductivities equal for $c \gg 1$ to

$$\sigma_m = 1 + ic, \qquad \sigma_d = \frac{1}{(d-1)^2 c^2} + \frac{i}{(d-1)}.$$

It is easy to verify that if $c \gg 1$ the complex number $z = (p-p_c)/h^{s/t}$ lands in the region where the function $\Psi(z)$ is defined, so that at $p = u$ the complex conductivity of the effective lattice is

$$\langle\sigma^{\mathrm{eff}}(p=u)\rangle = \sigma_0\, d e^{-p\Delta} \sigma_m h^s \Psi\left(\frac{p-p_c}{h^{s/t}}\right). \qquad (27)$$

We have thus the following situation (see Fig. 1): in the region OFC where $\mathrm{Re}\,\langle\sigma^{\mathrm{eff}}(p)\rangle$ is defined we have $\mathrm{Im}\,\langle\sigma^{\mathrm{eff}}(u)\rangle$ in the triangle OCD, while on the diagonal OC the function $\langle\sigma^{\mathrm{eff}}(p=u)\rangle$ is fully defined. Let us consider the condition for joining the functions constructed by us along the diagonal OC. To this end we turn to Fig. 3a, which shows $\mathrm{Re}\langle\sigma^{\mathrm{eff}}(p=u)\rangle$ and $\mathrm{Re}\langle\sigma^{\mathrm{eff}}(p=u-0)\rangle$, and to Fig 3(b), which shows $\mathrm{Im}\,\langle\sigma^{\mathrm{eff}}(p=u)\rangle$ and $\mathrm{Im}\langle\sigma^{\mathrm{eff}}(p=u+0)\rangle$ on the diagonal OC (the common factor $e^{-p\Delta}$ is omitted). It can be seen that the joining condition is satisfied exactly at $c = \infty$. Then, however, the frequency dependence of the electric conductivity acquires a logarithmic divergence at $\omega = \omega_c = \tau^{-1} e^{-p_c\Delta}$. This singularity (which is typical of the method of quasi-equilibrium finite clusters) can in our case be smoothed out by choosing for the parameter c a finite

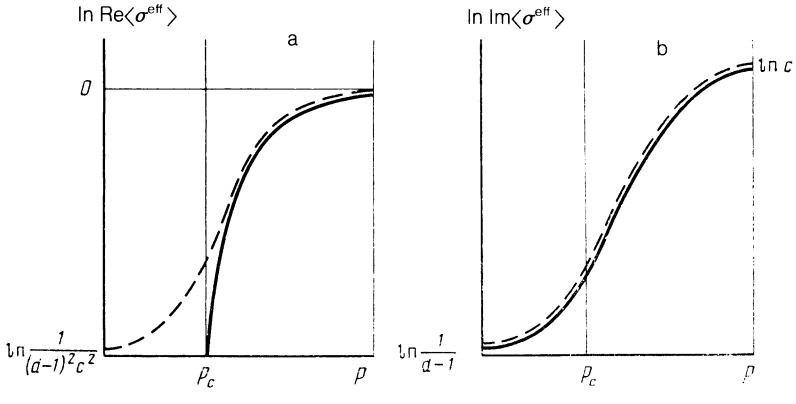

FIG. 3. Joining of the real (a) and imaginary (b) parts of the conductivity on the diagonal OC. The dashed curves are the dependences corresponding to the condition $p = u$.

value on the basis of simple physical consideration. From the definition of the quasi-nonequilibrium clusters it follows that the potentials of the sites belonging to one and the same cluster are equal. Such a formation has in an external field the electric properties of a metallic particle. Therefore as the frequency is lowered, $\omega \to \omega_c$, the dielectric constant of a metal–dielectric mixture with a metallic-phase concentration $x = \Delta^{-1} \ln(\omega/\omega_c)$ will increase like

$$\varepsilon(\omega) \approx \varepsilon_0 \Delta^q \ln^{-q}(\omega/\omega_c).$$

In view of the natural physical uncertainity of the boundary of a quasi-equilibrium cluster, this divergence should be "smeared-out" over a frequency region $|\ln(\omega/\omega_c)| \sim 1$. This, however, restricts the limiting value dielectric constant to $\varepsilon(\omega_c) \sim \varepsilon_0 \Delta^q$. Equating this value of that given by our solution $\varepsilon \propto c^{1-s}$, we obtain the dependence of c on Δ:

$$c \propto \Delta^{q/(1-s)} \propto \Delta^{t/s}. \tag{28}$$

It is easy to verify that in this case the condition for joining the real parts of the conductivity are likewise not violated. This estimate yields the depth of modulation of the spatial distribution of the current (its value coincides with c). In addition we obtain a nontrivial estimate of the tangent of the dielectric-loss angle in the region of the transition from the high-frequency to the static regime:

$$\operatorname{tg} \delta(\omega_c) \propto \Delta^{-t/s}. \tag{29}$$

Let us list some results. Our analysis has shown that the entire frequency range is divided by the frequency ω_c into two intervals. $\omega < \omega_c$ the real part of the conductivity is independent of frequency:

$$\operatorname{Re}\langle\sigma(\omega)\rangle = \sigma'(\omega) \approx \sigma_0 d \left(\frac{t}{\Delta}\right)^t \omega_c \tau. \tag{30}$$

If $\omega > \omega_c$, accordingly, the imaginary part of the conductivity is equal to

$$\operatorname{Im}\langle\sigma(\omega)\rangle = \sigma''(\omega) \approx \sigma_0 \frac{d}{d-1} \omega\tau \left(\frac{\Delta}{t}\right)^q \Psi\left[t^{-1} \ln\left(\frac{\omega}{\omega_c}\right)\right]. \tag{31}$$

The missing branches can be calculated by the Kramers–Kronig relations, which take in this situation the form[13]

$$\operatorname{Im}\langle\sigma(\omega)\rangle|_{\omega<\omega_c} = \left(1-\frac{\omega^2}{\omega_c^2}\right)^{1/2} \frac{2\omega}{\pi} \int_{\omega_c}^{\infty} d\omega' \left[\sigma''(\omega') \Big/ \left(\frac{\omega'^2}{\omega_c^2}-1\right)^{1/2} (\omega'^2-\omega^2)\right], \tag{32}$$

$$\operatorname{Re}\langle\sigma(\omega)\rangle|_{\omega>\omega_c} = \sigma_0 d \left(\frac{t}{\Delta}\right)^t \omega_c \tau - \left(\frac{\omega^2}{\omega_c^2}-1\right)^{1/2} \frac{2\omega}{\pi} \times \int_{\omega_c}^{\infty} d\omega' \left[\sigma''(\omega') \Big/ \left(\frac{\omega'^2}{\omega_c^2}-1\right)^{1/2} (\omega'^2-\omega^2)\right]. \tag{33}$$

As always, dispersion relations yield not the true conductivity but certain functions that satisfy formally Eqs. (32) and (33). In our case this circumstance is manifest in the form of unphysical jumps of the derivatives at $\omega = \omega_c$ (see Fig. 4). The onset of these singularities is due to the fact that in the course of the joining we continue the solutions obtained in the regions where λ is zero or infinite to the point where

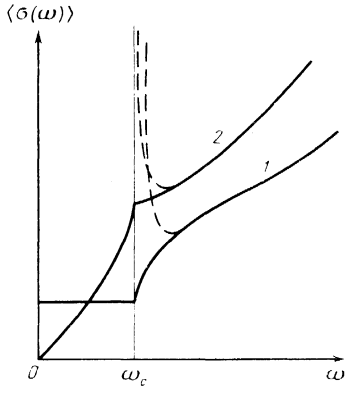

FIG. 4. General picture of the frequency dispersion of the electric conductivity. A dashed line shows the logarithmic divergence that arises in the approximation of quasi-equilibrium finte clusters; $1 - \operatorname{Re}\langle\sigma(\omega)\rangle$, $2 - \operatorname{Im}\langle\sigma(\omega)\rangle$.

$\lambda = 1$. The equation derived by us can therfore not be used in the immediate vicinity of ω_c. Greatest interest attaches thus to the asymptotic equations, which can be used in a large frequency interval

$$\langle\sigma(\omega)\rangle \approx \sigma_0 \frac{d}{d-1} \omega\tau \frac{\Delta^q}{\ln^q(\omega/\omega_c)} \left[i + \frac{\pi q}{2\ln(\omega/\omega_c)}\right], \quad \omega \gg \omega_c. \tag{34}$$

$$\langle\sigma(\omega)\rangle \approx \sigma_0 d \left[\left(\frac{t}{\Delta}\right)^t \omega_c\tau + i \frac{\Gamma(1-q)}{\pi(d-1)} \Delta^q \omega\tau\right], \quad \omega \ll \omega_c. \tag{35}$$

Relation (34) practically coincides with the one obtainable by the method of quasi-equilibrium finite clusters.[4]

4. LIMIT ON THE GROWTH OF THE SPATIAL SCALE OF CURRENT FLUCTUATIONS IN A STRONG ELECTRIC FIELD, AND NONLINEAR EFFECTS IN ELECTRIC CONDUCTIVITY

The question of the influence of a strong electric field on the electric conductivity of a polycrystal can also be investigated in the context of the approach described above. It is known that the field dependence of the conductivity of a polycrystal is due to the strong exponential nonlinearity of the current–voltage characteristics of the potential barriers that separate the crystallites:

$$J = J_0 \exp(-\mu\Delta) \operatorname{sh}\left(\frac{eV}{2T}\right). \tag{36}$$

Here V is the voltage drop across the barrier, J the current flowing through the barrier, and e the electron charge. In this case the connection between the barrier current and voltage is no longer determined by Ohm's law, but by the complicated nonlinear equation

$$\dot{v} + \Gamma(\mu) \operatorname{sh}(v) = j(t). \tag{37}$$

where

$$v = \frac{eV}{2T},$$

$$\Gamma(\mu) = \frac{J_0 e}{2Tc_0} \exp(-\mu\Delta) = \Gamma_M \exp(-\mu\Delta), \quad j(t) = \frac{eI(t)}{2Tc_0}.$$

Equation (37) must be linearized to derive from it a relation for the field-dependent barrier conductivity. Since the problem has no small parameter, we use, just as in Ref. 5,

the optimum-linearization method.[14] Assuming $j(t)$ to be a given function of the time, we replace the operator in the left-hand side of (37) by a linear one, by introducing a field-dependent effective relaxation frequency

$$\dot{v}+\gamma(\mu)v=j(t). \tag{38}$$

The parameter $\gamma(\mu)$ can be determined from the condition that the mean squared difference of the left-hand sides of (37) and (38) be a minimum:

$$\frac{d}{d\gamma(\mu)}\int_{t_1}^{t_2}[\Gamma(\mu)\operatorname{sh} v-\gamma(\mu)v]^2\,dt=0.$$

In the case of a periodic external signal the instants of time t_1 and t_2 must be chosen to satisfy the relation $t_2-t_1=2\pi/\omega$. We obtain thus the equation

$$\gamma(\mu)=\Gamma(\mu)\int_{t_1}^{t_2} v\operatorname{sh} v\,dt\Big/\int_{t_1}^{t_2} v^2\,dt. \tag{39}$$

Knowing $\gamma(\mu)$ we can represent the initial nonlinear system by a linear circuit in which each element contains a certain "linearized" electric conductivity

$$\sigma=\sigma_0[\gamma(\mu)+ie^{-u\Delta}]. \tag{40}$$

One must not forget, of course, that the integrals in the right-hand side of (39) contain the time dependence of the barrier voltage $v(t)$. In our approach, it is natural to find this function from expression (4) for the current, obtained in the effective-medium approximation:

$$v(t)=\frac{e\langle E\rangle a}{T}\operatorname{Re}\times\left(\frac{d(e^{-p\Delta}+ice^{-u\Delta})}{\gamma(\mu)+(d-1)e^{-p\Delta}+ie^{-u\Delta}[1+(d-1)c]}\right)e^{i\omega t}. \tag{41}$$

Substituting (41) in (39) we get for $\gamma(\mu)$ the transcendental equation

$$\frac{\gamma(\mu)}{\Gamma(\mu)}=\frac{2T}{e\langle E\rangle a\,dy}I_1\left[\frac{e\langle E\rangle a\,dy}{T}\right],$$
$$y=\left\{\frac{e^{-2p\Delta}+c^2e^{-2u\Delta}}{[\gamma(\mu)+(d-1)e^{-p\Delta}]^2+e^{-2u\Delta}[1+(d-1)c]^2}\right\}^{1/2}, \tag{42}$$

where $I_1(y)$ is a Bessel function of imaginary argument.

The problem of nonlinear electric conductivity of a polycrystal reduces thus to an analysis of Eq. (42) and of the ensuing renormalization-group equations of the linearized problem. In accord with the logic of our method, we must begin with a determination of the distributions of the real and imaginary parts of the effective conductivity

$$\sigma^{\text{eff}}=\sigma_0 d(\tilde{\sigma}e^{-p\Delta}+i\tilde{c}e^{-u\Delta})=\frac{\sigma_0 d(e^{-p\Delta}+ice^{-u\Delta})[\gamma(\mu)+ie^{-u\Delta}]}{\gamma(\mu)+(d-1)e^{-p\Delta}+ie^{-u\Delta}[1+(d-1)c]}, \tag{43}$$

$$P(\tilde{\sigma},\tilde{c})=\int_0^1\delta[\tilde{\sigma}-\tilde{\sigma}(\gamma(\mu),p,u,\Delta)]\delta[\tilde{c}-\tilde{c}(\gamma(\mu),p,u,\Delta)]\,d\mu. \tag{44}$$

We determine the $\gamma(\mu)$ dependence in the integral (44) from (42) and the functions $\tilde{\sigma}(\gamma(\mu),p,u,\Delta)$ and $\tilde{c}(\gamma(\mu),p,u,\Delta)$ from (43). To calculate the integral (44) we must change to a new integration variable

$$\eta=-\Delta^{-1}\ln\gamma(\mu),\ \eta(0)=-\Delta^{-1}\ln\gamma(0),\ \eta(1)=-\Delta^{-1}\ln\gamma(1).$$

The connection between the old and new variable can be obtained with the aid of (42). This connection becomes particularly simple $\Delta\gg 1$ if, as usual, we neglect the exponentially small terms. We have then for $p\leqslant u$

$$\mu=\eta+\theta(\eta-p)\frac{\alpha(E)}{\Delta},\quad \eta(0)=0,\quad \eta(1)=1-\frac{\alpha(E)}{\Delta}, \tag{45}$$

and for $p>u$

$$\mu=\eta+\theta(\eta-u)\frac{\alpha(E)}{\Delta},\quad \eta(0)=0,\quad \eta(1)=1-\frac{\alpha(E)}{\Delta}, \tag{46}$$

where the field dependence of the parameter $\alpha(E)$ is given by the simple given

$$\alpha(E)=\ln I_1\left[\frac{e\langle E\rangle ad}{T(d-1)}\right]-\ln\left[\frac{e\langle E\rangle ad}{2T(d-1)}\right]. \tag{47}$$

The meaning of $\alpha(E)$ is clear from the plot of (45) in Fig. 5. If $\mu\Delta$ is the unperturbed barrier height, then $\eta\Delta$ is the barrier height with allowance for the incident voltage $\alpha(E)$. It follows from (47) that at sufficiently high values of the argument

$$\alpha\sim\frac{e\langle E\rangle ad}{T(d-1)}. \tag{48}$$

By making the change of variables $\mu\to\eta$ we easily calculate the distribution density $P_0(\bar{\sigma},\bar{c})$, and the result differs from that in the linear case [Eqs. (16)–(18)] only in that the parameters p and u must now be replaced by p' and u':

$$p'=p+\frac{\alpha(E)}{\Delta},\quad u'=u+\frac{\alpha(E)}{\Delta}. \tag{49}$$

The argument of $\alpha(E)$, however, depends on the lattice period a, which increases after each renormalization-group transformation. In a strong electric field the number of parameters that determine the renormalization group of the effective medium is thus increased by unity.

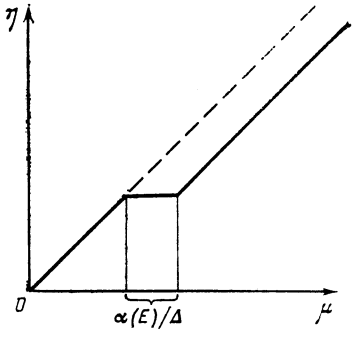

FIG. 5. Effective height of the barriers vs its unperturbed height in a strong alternating electric field. At $p>u$ the length $\alpha(E)$ of the horizontal "shelf" is proportional to the number of barriers that satisfy the "resonance" condition and are responsible for the absorption.

It is difficult to obtain the explicit form of the $\alpha(E)$ transformation after each enhancement. To clarify this tranformation in the region of small and large scales, however, it suffices to recall that the definition of the field $\langle E \rangle$ also contains the length, so that the scaling results depend on how we define the mean field acting in the system. In an infinite medium, at fixed p' and u', a natural definition of the mean field is

$$\langle E \rangle = \frac{\langle V_c \rangle}{L_c(p')}, \tag{50}$$

where $\langle V_c \rangle$ is the average voltage drop over a scale $L_c \sim a|p_c - p'|^{-\nu}$. In each enhancement we must multiply the parameter a by a quantity $b > 1$. At not too large a number k of iterations we have $ab^k \ll L_c$ and the renormalization approximation affects only the microscale, leaving L_c unchanged. The increase of $\alpha(E)$ after each renormalization is then given by

$$\alpha_k \approx \alpha\left(\frac{e\langle V_c \rangle}{T}\frac{ab^k}{L_c}\right),$$

where $\langle V_c \rangle = \langle E \rangle L_c$. Starting with $k^* = \ln L_c / \ln a$ there is left in the system only one spatial scale L_c, and the renormalization transformation leaves the parameter $\alpha(E)$ invariant:

$$\alpha' = \alpha\left(\frac{e\langle V_c \rangle}{TbL_c} bL_c\right) = \alpha_c\left(\frac{e\langle V_c \rangle}{T}\right) = \alpha_c\left(\frac{e\langle E \rangle L_c}{T}\right).$$

The limiting forms of the equations in (49) are therefore

$$p' = p + \frac{eEad}{2T\Delta|p_c - p'|^{\nu}(d-1)}, \quad p < u, \tag{51}$$

$$u' = u + \frac{eEad}{2T\Delta|u_c - u'|^{\nu}(d-1)}, \quad p > u. \tag{52}$$

These equations contain a fundamentally important effect, namely, the growth of the spatial scale of the fluctuations is limited in a strong electric field. In fact, consider the $p'(p)$ dependence plotted in Fig. 6. It can be seen that in a strong field that satisfies the condition $eEL_c > T$ first introduced by Shklovskiĭ[15] this function has a jump at a certain field-dependent value

$$p^* = p_c - \left[\frac{veEad}{2T\Delta(d-1)}\right]^{1/(\nu+1)}\left(\frac{\nu+1}{\nu}\right). \tag{53}$$

Consider the region $p < u$. From Fig. 7 it is clear that the jump of $p'(p)$ leads to a jumpwise change of the conductivity $\mathrm{Re}\langle\sigma^{\mathrm{eff}}(p)\rangle$ at $p = p^*$. Obviously, this jump leads to a shift

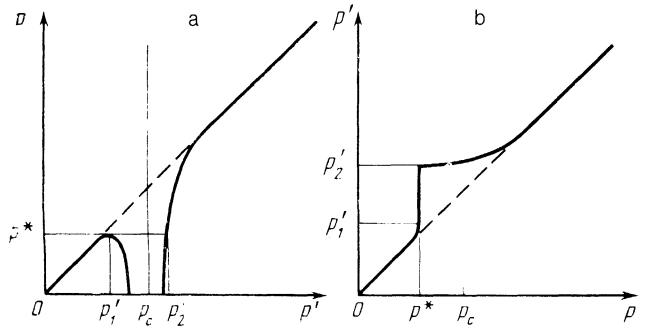

FIG. 6. Graphic solution of Eq. (51) (a) and the corresponding $p'(p)$ dependence (b).

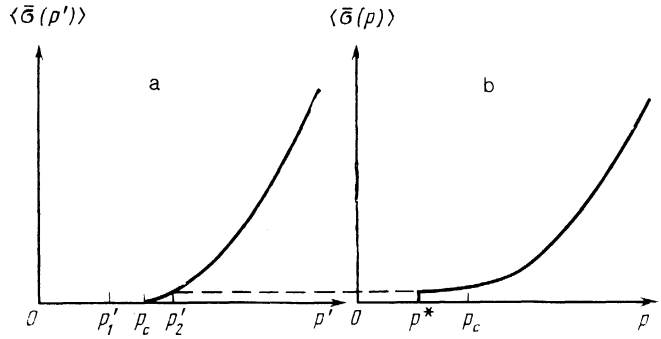

FIG. 7. Alternation of the character of the percolation phase transition by the jump of the function $p'(p)$.

of the maximum of $\mathrm{Re}\langle\sigma^{\mathrm{eff}}(p)\rangle$ with increase of the electric field and to the strong exponential field dependence predicted in Ref. 15 for the conductivity and confirmed in Ref. 16 by a direct computer calculation:

$$\mathrm{Re}\langle\sigma\rangle \approx \sigma_0 d\left[\frac{eEad}{2T\Delta(d-1)}\right]^{1/(\nu+1)} \times \exp\left\{-p_c\Delta + \left[\frac{eEa\Delta^{\nu}d}{2T(d-1)}\right]^{1/(\nu+1)}\left(\frac{\nu+1}{\nu}\right)\right\}. \tag{54}$$

The correlation radius of the current fluctuations in a polycrystal decreases with increase of the electric field like

$$L_c(E) \approx a\left[\frac{veEad}{2T\Delta(d-1)}\right]^{-\nu/(\nu+1)} \tag{55}$$

We consider now the region $p > u$. In strong fields we can put, without loss of generality, $c = \infty$. It turns out here that the limiting frequency ω_c is repalced by a higher frequency $\omega_c^* = \tau^{-1}\exp(-p^*\Delta)$. If $\omega < \omega_c^*$ the function $\mathrm{Re}\langle\sigma\omega\rangle = \sigma'(\omega)$ does not depend on the frequency and is described by Eq. (54). If $\omega > \omega_c^*$ the real and imaginary parts (32) and (33) of the conductivity, calculated using the Kramers–Kronig relations, can be expressed in terms of the parameter $t(\omega)$:

$$\mathrm{Re}\langle\sigma(\omega)\rangle \approx \sigma'(E) + \sigma_0\frac{d}{d-1}\omega\tau\Delta^q\frac{\pi q}{2t^{q+1}(\omega)}, \tag{56}$$

$$\mathrm{Im}\langle\sigma(\omega)\rangle \approx \sigma_0\frac{d}{d-1}\omega\tau\frac{\Delta^q}{t^q(\omega)}. \tag{57}$$

The value of $t(\omega)$ is determined from the simple transcendental equation

$$t(\omega) + \frac{\alpha^*}{t^{\nu}(\omega)} = \ln\left(\frac{\omega}{\omega_c}\right), \quad \alpha^* = \frac{eEa\Delta^{\nu}}{2T(d-1)}, \tag{58}$$

which follows directly from (52). Equation (56)–(58) shows that nonlinear effects should be observed in the high-frequency conductivity in the frequency interval

$$\omega_c \exp\left[(\alpha^*)^{1/(\nu+1)}\right] \ll \omega \ll \omega_c \exp(\alpha^*), \tag{59}$$

and the condition for the validity of the linear approximation is

$$\frac{eEa\Delta^{\nu}}{T\ln^{\nu}(\omega/\omega_c)} < 1. \tag{60}$$

This formula must be regarded as a direct generalization of Shklovskiĭ's[15] nonlinearity criterion for the static case to in-

clude the case of alternating fields. Interest attaches to the field dependence of the tangent of the dielectric-loss angle in the region of the transition from the high-frequency regime to the static one:

$$\operatorname{tg}\delta(\omega_c^*)\approx(d-1)\left[\frac{eEad}{2T\Delta(d-1)}\right]^{(t+q)/(\nu+1)} \qquad (61)$$

It can be seen that the absorption increases in strong electric fields. This effect is due to equalization of the heights of the "resonance" barriers—a phenomenon whose possible existence was pointed out by me first in Ref. 5. The general conclusion that the degree of inhomogeneity of a polycrystalline semiconductor is decreased in a strong alternating electric field has thus been fully confirmed for the two- and three-dimensional cases.

5. CONCLUSION

The problem solved in the present paper is an example of a direct application of the renormalization-group method to an investigation of a system of the "percolation" type. The very possibility of this approach is due to the percolation structure of the spatial distribution of the local current fluctuations in the volume of the sample—a fact that is almost obvious and is implicitly used in the classical percolation-theory method. The present paper emphasizes, apparently for the first time ever, the role of the percolative current structure in electric-conduction systems of the hopping type, and establishes the connection between the current-fluctuation amplitude and the dispersion of the logarithm of the electric conductivity [Eq. (28)]. It should be noted that the simplicity of the resultant renormalization group is due to the assumed large value of this parameter. This simplification leads to a certain loss of information, in particular to unphysical jumps of the derivatives at $\omega=\omega_c$. The accuracy of the calculations confirms fully the accuracy of the classical method of percolation theory.

In cases of ohmic conductivity, the results agree, within the accuracy of the method, with those that can be obtained by previously known methods.[1,6,7] For fields that vary strongly with time, however, the frequency and field dependences (56)–(61) differ substantially from those that can be assumed directly by generalizing the results of an investigation of the one-dimensional model.[5] This pertains in particular to the field dependence of $\tan\delta(\omega)$, which has in the two- and three-dimensional cases an entirely different form that in the one-dimensional one. An explanation of these differences is that the resistance of a one-dimensional polycrystal is proportional to the number of resonance barriers $\alpha(E)$ that are connected in series (since we are considering a one-dimensional case). When the number of dimensions of the space is increased, resonance barriers can become connected in parallel, and this changes the field dependence of the absorption.

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