Conductivity of random inhomogeneous media

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The problem of finding the effective conductivity tensor σ_* of a random inhomogeneous medium is solved. The tensor σ_* is obtained in the form of a series together with the convergence conditions. The boundary points $\sigma_\pm^{(n)}$ (here n is the multiplicity of the allowed interactions) of the region containing σ_* are also found. In the case of a mixture of two isotropic components (for $n\leqslant 3$), the effect of the topological properties of the components on the statistical parameters determining the structure of the medium is investigated. Finally, for a self-consistent medium a solution σ_{SC} is obtained that allows for macroscopic anisotropy. This solution is shown to describe the critical behavior near the percolation threshold.

1. INTRODUCTION

The problems that emerge when random inhomogeneous media are studied macroscopically have attracted the unflagging attention of researchers (see, e.g., Refs. 1–4). This is due, first, to the fact that such media differ substantially from homogeneous and regular-inhomogeneous media, which leads to additional mathematical difficulties, and, second, to the possibility of obtaining results that have important applications. A key problem is that of finding the macroscopic (effective) material characteristics of an inhomogeneous medium from a given body of statistical data. Solution of this problem follows three basic paths: (1) searching for exact solutions for real media and model structures, (2) determining the boundary points of the region containing the effective characteristics of a medium, and (3) calculating the approximate values.

The simplest exact solutions obtained by Wiener in 1912 (see, e.g., Refs. 5 and 6) have the form

$$\sigma_{ij}^*=\sigma_1^*\delta_{ij}+(\sigma_3^*-\sigma_1^*)\delta_{i3}\delta_{j3}, \tag{1.1}$$

$$\sigma_1^*=\langle\sigma\rangle,\quad \sigma_3^*=\langle\rho\rangle^{-1},\quad \sigma\rho\equiv 1, \tag{1.2}$$

where

$$\langle X\rangle\equiv\sum_{a=1}^{N}v_aX(a),\quad \sum v_a=1,$$

$$X_{ij}(a)=X(a)\delta_{ij},\quad X=\sigma,\rho.$$

Here σ_* is the effective conductivity tensor of a medium whose N isotropic components constitute randomly alternating plane-parallel layers with a normal parallel to the third axis, and v_a is the volume concentration of the ath component. The quantities σ_1^* and σ_3^* are also the boundary points of the region

$$\langle\rho\rangle^{-1}\equiv\sigma_-^W\leqslant\sigma_*\leqslant\sigma_+^W\equiv\langle\sigma\rangle \tag{1.3}$$

containing the effective conductivity σ_* of the macroisotropic mixture of N isotropic components.

Keller's theorem[7] and Dykhne's symmetry transformations[8] gave a new impetus to the study of symmetry properties of inhomogeneous media. This yielded several interesting results.[9–15] It also led to structures that allow for exact solutions.

The self-consistent solutions obtained by Bruggeman[5] for two- and three-dimensional macroisotropic mixtures of two isotropic components are often used to interpret the results of measurements and in percolation theory.[8,12,14,16,17]

Starting from classical energy relations, Hashin and Shtrikman[6] developed a variational method for obtaining the boundary points $\sigma_\pm^{HS}$, which, in contrast to the Wiener boundary points $\sigma_\pm^W$, allow for the statistical data contained in two-point probabilities. Partial allowance for three-point interaction, which was made by Beran,[18] led to additional narrowing of the region in the cases of three-dimensional[9] and two-dimensional[19] macroisotropic mixtures of two isotropic components.

Lifshitz together with co-workers suggested a method for calculating static[20] and dynamic[21] effective characteristics in problems of elasticity theory. This is based on solving differential equations whose coefficients are random tensor fields. The solution is represented in the form of a series each term of which describes interactions of respective multiplicity. The method of random-fields theory was found to be effective both in deriving approximate solutions and in selecting auxiliary fields used in variational methods.

To calculate σ_* and the boundary points $\sigma_\pm^{(n)}$ with $n\leqslant 3$ (the multiplicity of the allowed interactions is $k\in[0,n]$) of the region containing σ_*, we develop below a method[22] that uses the advantages of both the method of random-field theory[20–23] and the variational method.[6,9,18,19] We find that for $n\leqslant 2$ the boundary points $\sigma_\pm^{(n)}$ satisfy relationships that follow from Keller's theorem and Dykhne's transformations.[7,8,10,11,14,15] A self-consistent solution is obtained for the case of an arbitrarily shaped inclusion. Finally, the possibility of using this solution to describe anomalous phenomena near the percolation threshold is demonstrated.

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2. THE EFFECTIVE CONDUCTIVITY TENSOR

Let us consider the equations for a time-independent current:

$$\nabla\cdot\mathbf{j}=0,\quad \nabla\times\mathbf{E}=0. \tag{2.1}$$

Representing the field $\mathbf{E}$, according to (2.1), in the form

$$\mathbf{E}=-\nabla\varphi, \tag{2.2}$$

where φ is the scalar potential, and allowing for the material equation, Ohm's law

$$\mathbf{j}=\sigma\mathbf{E},\quad j_k=\sigma_{kj}E_j,\quad \sigma=\sigma(\mathbf{r}), \tag{2.3}$$

where σ is the random tensor field, we proceed from Eq. (2.1) to the Laplace equation

$$L\varphi=0,\quad L=\nabla\cdot\sigma\nabla=\nabla_i\sigma_{ij}\nabla_j,\quad \mathbf{r}\in V, \tag{2.4}$$

for an inhomogeneous medium, which is solved with the boundary conditions

$$\varphi(\mathbf{r})=\varphi_0,\quad \mathbf{r}\in S_1,\quad j_n(\mathbf{r})=j_0,\quad \mathbf{r}\in S_2,$$
$$j_n\equiv\mathbf{n}\cdot\mathbf{j},\quad S_1\cup S_2=S. \tag{2.5}$$

Here S is the surface forming the boundary of volume V, and $\mathbf{n}$ the unit outward normal to S.

Let us now introduce the Green function of the Poisson equation with homogeneous boundary conditions:

$$L_cG(\mathbf{r},\mathbf{r}')=-\delta(\mathbf{r}-\mathbf{r}'),\quad L_c=\nabla\cdot\sigma_c\nabla,\quad \mathbf{r},\mathbf{r}'\in V,$$
$$G(\mathbf{r},\mathbf{r}')=0,\quad \mathbf{r}\in S_1,\quad \mathbf{n}\cdot\sigma_c\nabla G(\mathbf{r},\mathbf{r}')=0, \tag{2.6}$$
$$\mathbf{r}\in S_2,\quad \mathbf{r}'\in V.$$

Here the subscript c marks quantities that refer to an auxiliary (or complementary) medium, which macroscopically (geometry in the large and the boundary conditions) is identical to the inhomogeneous medium considered and differs only in material characteristics. The operator L_c obeys the ordinary restrictions imposed on an unperturbed operator: (1) the solution to problem (2.4), (2.5) for L_c is known, and (2) the perturbation operator $L'\equiv L-L_c$ is in a certain sense small, which ensures the convergence of the perturbation series.

Using the mathematical tools developed in Ref. 22, we write the solution to problem (2.4), (2.5) for the field $\mathbf{E}$ specified by (2.2) in the form

$$\mathbf{E}=\hat{A}\langle\mathbf{E}\rangle,\quad \hat{A}=\sum_{k=0}^{\infty}(R\hat{Q}\sigma')^k,\quad \sigma'\equiv\sigma-\sigma_c. \tag{2.7}$$

Here $\hat{Q}$ is specified as follows:

$$\mathbf{X}\equiv\hat{Q}\mathbf{Y},\quad X_i(\mathbf{r}_1)=\int Q_{ij}(\mathbf{r}_1,\mathbf{r}_2)Y_j(\mathbf{r}_2)d^3r_2,$$
$$Q_{ij}(\mathbf{r}_1,\mathbf{r}_2)\equiv-\nabla_i^1\nabla_j^2G(\mathbf{r}_1,\mathbf{r}_2),\quad \nabla_i^\alpha\equiv\frac{\partial}{\partial r_{\alpha i}},\quad \alpha=1,2, \tag{2.8}$$

and the angle brackets stand for averaging over the realization ensemble, which under certain conditions[8,18,22] coincides with averaging over the volume. Finally, the operator R that isolates the random component of a field F satisfies the following relations:

$$(RF)^{n+1}\equiv F(RF)^n-\langle F(RF)^n\rangle,\quad n\geqslant 0. \tag{2.9}$$

To describe macroscopically the conducting properties of an inhomogeneous medium we use the effective conductivity tensor σ_* specified by the following relations:[6,8,15,17,18,20,22–24]

$$\langle\mathbf{j}\rangle=\langle\sigma\mathbf{E}\rangle\equiv\sigma_*\langle\mathbf{E}\rangle, \tag{2.10}$$

which in combination with (2.7) yields[22,24]

$$\hat{\sigma}_*=\langle\sigma\hat{A}\rangle,\quad \langle\hat{A}\rangle=\hat{I},\quad \hat{I}\mathbf{X}=\mathbf{X}. \tag{2.11}$$

Generally, $\hat{\sigma}_*$ is an integral operator, but if, firstly, the dimensions of the inhomogeneous medium are large compared to the inhomogeneity scale of the mean field $\langle\mathbf{E}\rangle$, the mean free path of the charge carriers, and the characteristic size of the region over which the averaging that replaces ensemble averaging is done; and secondly, the inhomogeneous medium possesses the property of statistical homogeneity, in view of which n-point probabilities of the random field $\sigma(\mathbf{r})$ are invariant under translations, then the operator $\hat{\sigma}_*$ possesses the property of locality, and the kernel of $\hat{\sigma}_*$ can be represented in the form

$$\sigma_*(\mathbf{r}_1,\mathbf{r}_2)=\sigma_*\delta(\mathbf{r}_1-\mathbf{r}_2),\quad \sigma_*=\text{const.} \tag{2.12}$$

In the limit of macrohomogeneous boundary conditions, we have instead of (2.5)

$$\varphi(\mathbf{r})=-\mathbf{r}\cdot\langle\mathbf{E}\rangle,\quad \text{or}\quad j_n(\mathbf{r})=\mathbf{n}\cdot\langle\mathbf{j}\rangle,\quad \mathbf{r}\in S, \tag{2.13}$$

where $\langle\mathbf{E}\rangle$ and $\langle\mathbf{j}\rangle$ are constant vectors.

In view of (2.12), combining (2.11) and (2.7) yields[22]

$$\hat{\sigma}_*=\sigma_c\hat{I}+\sum_{k=0}^{\infty}\langle\sigma'(R\hat{Q}\sigma')^k\rangle=\sigma_*\hat{I},\quad \sigma_*=\sum_{k=0}^{\infty}\sigma_k. \tag{2.14}$$

Macroscopically, the problem of describing an inhomogeneous medium is solved in a similar manner if, following (2.1), we write $\mathbf{j}$ in the form

$$\mathbf{j}=\nabla\times\boldsymbol{\psi}, \tag{2.15}$$

with $\boldsymbol{\psi}$ the vector potential. Allowing for the material equation

$$\mathbf{E}=\rho\mathbf{j},\quad E_i=\rho_{ik}j_k,\quad \rho\sigma\equiv I,\quad \rho=\rho(\mathbf{r}), \tag{2.16}$$

where ρ is the random-resistances tensor field, we proceed from (2.1) to the vector equation

$$M\boldsymbol{\psi}=0,\quad M_{ij}=e_{imk}e_{lnj}\nabla_m\nabla_n\rho_{kl}. \tag{2.17}$$

Here e_{imk} are the components of the Levi–Civita identity tensor. The procedure for solving Eq. (2.17) is similar to that described above. For the tensor ρ_* specified by the equations

$$\langle\mathbf{E}\rangle=\langle\rho\mathbf{j}\rangle\equiv\hat{\rho}_*\langle\mathbf{j}\rangle,\quad \hat{\rho}_*\hat{\sigma}_*=\hat{I}, \tag{2.18}$$

it leads to the expression[22]

$$\hat{\rho}_*=\rho_c\hat{I}+\sum_{k=0}^{\infty}\langle\rho'(R\hat{P}\rho')^k\rangle,\quad \rho_*=\sum_{k=0}^{\infty}\rho_k,$$
$$\rho_*\sigma_*=I, \tag{2.19}$$

similar to (2.14), where operator $\hat{P}$ is related to operator $\hat{Q}$ through the equation

$$-\hat{P}=\sigma_c\hat{I}+\sigma_c\hat{Q}\sigma_c. \qquad (2.20)$$

Usually σ_* and ρ_* are calculated on the basis of a limited body of information about the inhomogeneous medium, which yields the following approximate values for σ_* and ρ_*:

$$\sigma_*^{(n)}\equiv\sum_{k=0}^{n}\sigma_k,\quad \rho_*^{(n)}\equiv\sum_{k=0}^{n}\rho_k, \qquad (2.21)$$

which allow for k-particle interactions between the inhomogeneity regions ($k\in[0,n]$). Generally, $\sigma_*^{(n)}$ and $\rho_*^{(n)}$ are not related via (2.19). Moreover, their position in relation to σ_* and ρ_* often remains uncontrollable. Hence the importance of the problem of finding the boundary points $\sigma_\pm^{(n)}$ (with n the greatest multiplicity of the allowed interactions) of the region containing σ_*.

3. THE BOUNDARY POINTS $\sigma_\pm^n$ FOR σ_*

Studying the series in (2.7) leads to the following inequalities:[22]

$$\frac{1}{2}\sigma<\frac{\sigma}{1+k_1}\leqslant\sigma_c\leqslant\frac{\sigma}{1-k_1}<\infty,\quad 0\leqslant k_1\leqslant 1, \qquad (3.1)$$

which form the sufficient conditions for convergence of the series. Also, the terms in (2.7) satisfy the conditions

$$\sigma_k\leqslant 0,\quad k\geqslant 1,\quad \sigma'\equiv\sigma-\sigma_c\leqslant 0, \qquad (3.2a)$$

$$\sigma_{2k}\leqslant 0\leqslant\sigma_{2k-1},\quad k\geqslant 1,\quad \sigma'\geqslant 0, \qquad (3.2b)$$

in view of which the position of $\sigma_*^{(n)}$ (2.21) in relation to σ_* is controllable at each step of the calculation procedure. Indeed, combining (3.1) and (3.2) yields the following inequalities:

$$\sigma_*\leqslant\sigma_*^{(n)}\equiv\sum_{k=0}^{n}\sigma_k,\quad n\geqslant 0,\quad \sigma\leqslant\sigma_c<\infty, \qquad (3.3a)$$

$$\sigma_*^{(2n)}\leqslant\sigma_*\leqslant\sigma_*^{(2n+1)},\quad n\geqslant 0,\quad \tfrac{1}{2}\sigma<\sigma_c\leqslant\sigma. \qquad (3.3b)$$

In the case of (2.15) the sufficient conditions for convergence of the respective series are

$$\frac{1}{2}p<\frac{p}{1+k_2}\leqslant p_c\leqslant\frac{p}{1-k_2}<\infty,\quad 0\leqslant k_2<1. \qquad (3.4)$$

For the terms of the series in (2.19) instead of (3.2) we write

$$\rho_k\leqslant 0,\quad k\geqslant 1,\quad p'\equiv p-p_c\leqslant 0, \qquad (3.5a)$$

$$\rho_{2k}\leqslant 0\leqslant\rho_{2k-1},\quad k\geqslant 1,\quad \rho'\geqslant 0. \qquad (3.5b)$$

Thus, for $k\geqslant 1$ the series in (2.19) is of constant sign if $\rho'\leqslant 0$ and an alternating series if $\rho'\geqslant 0$. Combining (3.4) and (3.5) yields the following inequalities:

$$\rho_*\leqslant\rho_*^{(n)}\equiv\sum_{k=0}^{n}\rho_k,\quad n\geqslant 0,\quad \rho\leqslant\rho_c<\infty, \qquad (3.6a)$$

$$\rho_*^{(2n)}\leqslant\rho_*\leqslant\rho_*^{(2n+1)},\quad n\geqslant 0,\quad \tfrac{1}{2}\rho<\rho_c\leqslant\rho. \qquad (3.6b)$$

The inequalities (3.3a) and (3.6a) prove to be more convenient. Using them simultaneously yields two-sided boundaries for both σ_* and ρ_*:

$$[Y_*^{(n)}]^{-1}\leqslant X_*\leqslant X_*^{(n)},\quad XY=I,$$
$$X_*^{(n)}\equiv\sum_{k=0}^{n}X_k,\quad X_c\equiv\sup X,\quad X=\sigma,\rho. \qquad (3.7)$$

For a macroisotropic mixture of N isotropic components this implies

$$Y_0^{-1}\leqslant X_*\leqslant X_0,$$
$$X_0=X_c=\max X(a)\equiv X_m,\quad \text{for } n=0, \qquad (3.8a)$$
$$(Y_0+Y_1)^{-1}=\langle Y\rangle^{-1}\leqslant X_*\leqslant\langle X\rangle$$
$$=X_0+X_1$$
$$=\sum_{a=1}^{N}v_aX(a)\quad \text{for } n=1, \qquad (3.8b)$$

where $X(a)$ is the value of X for the ath component. The boundary points in (3.8a) and (3.8b) are the best, in the volume $n=0$ and $n=1$ respectively, statistical information about the medium and are denoted by $X_\pm^{(n)}$.

For the sake of convenience we use below the language of conductivities. Bearing in mind the above, instead of (3.8) we get

$$\rho_m^{-1}\equiv\sigma_-^{(0)}\leqslant\sigma_*\leqslant{}_+^{(0)}\equiv\sigma_m,\quad \sigma_m\rho_m\geqslant 1,\quad \text{for } n=0, \qquad (3.9a)$$

$$\langle\rho\rangle^{-1}\equiv\sigma_-^{(1)}\leqslant\sigma_*\leqslant\sigma_+^{(1)}\equiv\langle\sigma\rangle,\quad \text{for } n=1. \qquad (3.9b)$$

For $n=2$, according to (3.7), (2.14), and (2.19), we can write[22]

$$[\rho_*^{(2)}]^{-1}\leqslant\sigma_*\leqslant\sigma_*^{(2)},\quad \rho_c=\rho_m,\quad \sigma_c=\sigma_m,$$
$$\rho_*^{(2)}=\langle\rho\rangle-\bar{h}<(\rho'')^2\rho_c^{-1}, \qquad (3.9c)$$
$$\sigma_*^{(2)}=\langle\sigma\rangle-\bar{g}\langle(\sigma'')^2\rangle\sigma_c^{-1},\quad \bar{g}+\bar{h}=1,$$

where the geometric parameter $\bar{g}$ is a component of the depolarization tensor (for a macroisotropic medium $\bar{g}=d^{-1}$, where d is the dimensionality of the medium's space). Tensor $\bar{g}$ is calculated via two-point probabilities and is determined by the shape of the surface of the effective inhomogeneity grain.[23] When the inclusions in a homogeneous lattice are distributed at random, the effective grain is a sphere.

The boundary points in (3.9c) are not the best in the information volume $n=2$ that includes two-point probabilities. To narrow the region, Hashin and Shtrikman[6] developed a variational approach in which they introduced a functional that in our notation has the form

$$S[\boldsymbol{\tau}]=\tfrac{1}{2}(\mathbf{E}_c,\sigma_c\mathbf{E}_c)+\tfrac{1}{2}(\boldsymbol{\tau},2\mathbf{E}_c+\mathbf{E}'-q\boldsymbol{\tau}),$$

$$(\mathbf{x},\mathbf{y})\equiv\int\mathbf{x}\cdot\mathbf{y}\,dV,\quad q\sigma'\equiv I,\quad \boldsymbol{\tau}=\mathbf{j}-\sigma_c\mathbf{E}. \qquad (3.10)$$

Using (3.10) we obtain from the classical theorems for the energy U of an electric field

$$-M_2 \leqslant U-S[\tau] \leqslant M_1,$$

$$2M_1(\mathbf{F}_1,\sigma'\mathbf{F}_2), \quad 2M_2 \equiv (\mathbf{F}_2,\rho'\mathbf{F}_2), \tag{3.11}$$

$$\mathbf{F}_1 \equiv \rho_c \mathbf{F}_2 = \mathbf{E}-q\boldsymbol{\tau}.$$

Equality is achieved if we substitute the fields $\mathbf{E}$ and $\boldsymbol{\tau}$ that satisfy Eqs. (2.1) and (2.5), and, in view of this, are coupled by the equation $\mathbf{E}=q\boldsymbol{\tau}$.

The extremum of the functional S is an absolute minimum for $M_1 \leqslant 0$ and an absolute maximum for $M_2 \leqslant 0$.[6] For such cases Eq. (3.11) yields

$$U \leqslant S_+, \quad M_1 \leqslant 0, \quad \sigma' \leqslant 0 \leqslant \rho',$$

$$S_- \leqslant U, \quad M_2 \leqslant 0, \quad \rho' \leqslant 0 \leqslant \sigma', \tag{3.12}$$

where S_+ and S_- are the extreme values of $S[\tau]$ for $M_1 \leqslant 0$ and $M_2 \leqslant 0$, respectively.

When macrohomogeneous boundary conditions (2.13) are employed, by combining (3.12) with (3.9a) we find the best boundary points for the effective conductivity σ_* in the case of $n=2$ (Refs. 22 and 23):

$$\sigma_-^{(2)} \leqslant \sigma_* \leqslant \sigma_+^{(2)}, \quad \sigma_\pm^{(2)} \equiv \sigma_s(\sigma_\pm^c),$$

$$\sigma_s + \bar{t}\sigma_c \equiv \frac{1}{\langle (\sigma+\bar{t}\sigma_c)^{-1}\rangle}, \quad \bar{t} \equiv \frac{\bar{h}}{\bar{g}}, \tag{3.13}$$

$$\sigma_+^c \equiv \sigma_m, \quad \sigma_-^c \equiv \rho_m^{-1}.$$

The first to obtain the boundary points (3.13) in a different form for a macroisotropic mixture of N isotropic components ($d=3$) were Hashin and Shtrikman.[6]

Allowing for three-particle interactions involves cumbersome mathematical calculations, in view of which the majority of papers dealing with this effect consider macroisotropic mixtures of two isotropic components. One of the first to attempt to use statistical information of volume $n=3$ was Beran,[18] who suggested a simple modification of the variational approach.

Irrespective of the choice of parameters σ_c and ρ_c, the approximate values $\sigma_*^{(3)}$ and $\rho_*^{(3)}$ are, according to (3.3) and (3.6), the upper boundaries for ρ_c and ρ_*, respectively. The arbitrariness of parameters σ_c and ρ_c is used below to obtain more stringent boundaries in the case of $n=3$.

For a macroisotropic medium of two isotropic components Eq. (2.14) yields (here $\sigma_1 \leqslant \sigma_2$)

$$\sigma_*^{(3)}(u) = \langle\sigma\rangle - \bar{g}D_\sigma(2u-Ku^2), \quad u\sigma_c \equiv 1,$$

$$D_\sigma \equiv \langle(\sigma'')^2\rangle = v_1v_2(\sigma_1-\sigma_2)^2 \quad \sigma_a \equiv \sigma(a),$$

$$v_a \in [0,1], \quad a=1,2, \tag{3.14}$$

$$K \equiv \bar{g}[\sigma] + \bar{h}\langle\sigma\rangle_j, \quad [\sigma] \equiv v_1\sigma_2 + v_2\sigma_1,$$

$$\langle\sigma\rangle_j \equiv \sigma_1 + j(\sigma_2-\sigma_1).$$

Here the geometric parameter j describing three-particle interactions is defined as

$$j \equiv (\mathbf{y}, f\mathbf{y}), \quad f=f(\mathbf{r}) = \begin{cases} 0 \text{ if } \mathbf{r}\in V_1, \\ 1 \text{ if } \mathbf{r}\in V_2, \end{cases} \quad V_1 \cup V_2 = V, \tag{3.15}$$

$$\mathbf{y} = \mathbf{Y}(\mathbf{Y},\mathbf{Y})^{-1/2}, \quad \mathbf{Y} \equiv (\bar{\hat{Q}} - \bar{g}\hat{I})R\boldsymbol{\tau},$$

$$\bar{\hat{Q}} \equiv -\sigma_c^{1/2}\hat{Q}\sigma_c^{1/2},$$

and is related to the parameter J introduced in Ref. 22 through the following formula:

$$j = \frac{J-J_-}{J_+-J_-}, \quad v_1\bar{g} - v_2 \equiv J_- \leqslant J \leqslant J_+ \equiv v_1 - v_2\bar{g}. \tag{3.16}$$

The coordinate dependence of the piecewise homogeneous approximating field $\boldsymbol{\tau}$ is fully determined by the indicator function f defined in (3.15). From (3.15) and (3.16) it follows also that

$$j = \frac{(\bar{\hat{Q}}f'', f\bar{\hat{Q}}f'')}{\bar{g}\bar{h}\langle(f'')^2\rangle} - v_1\frac{\bar{g}}{\bar{h}}, \quad j\in[0,1], \quad v_a \in [0,1]. \tag{3.17}$$

The arbitrariness of the auxiliary parameter u makes it possible to minimize the function $\sigma_*^{(3)}(u)$ defined in (3.14). This yields

$$\min \sigma_*^{(3)} \equiv \sigma_+^{(3)} = \langle\sigma\rangle - \frac{\bar{g}D_\sigma}{\bar{g}[\sigma] + \bar{h}\langle\sigma\rangle_j}. \tag{3.18a}$$

Similarly, for $\rho_+^{(3)}$ we get

$$\min \rho_*^{(3)} \equiv \rho_+^{(3)} = \langle\rho\rangle = \frac{\bar{h}D_\rho}{\bar{h}[\rho] + \bar{g}\langle\rho\rangle_j}. \tag{3.18b}$$

The solutions (3.18a) and (3.18b) can be represented in the form (3.13):

$$\sigma_+^{(3)} = \sigma_s(\langle\sigma\rangle_j), \quad [\rho_+^{(3)}]^{-1} \equiv \sigma^{(3)} = \sigma_s(\langle\rho\rangle_j^{-1}). \tag{3.19}$$

Eqs. (3.19) and (3.13) yield for the limiting values of parameter j specified by (3.17)

$$\sigma_+^{(3)} = \sigma_-^{(3)} = \begin{cases} \sigma_-^{(2)} \text{ if } j=j_- \equiv 0, \\ \sigma_+^{(2)} \text{ if } j=j_+ \equiv 1. \end{cases} \tag{3.20}$$

Miller[9] suggested a model of a symmetric medium whose statistical properties were studied in Ref. 13. (The components of a symmetric medium have the same geometry and differ only in the values of conductivity and bulk density.) At $v_1=v_2$ such a medium satisfies the Dykhne equivalence criterion.[8] For a symmetric medium, as can be demonstrated,[13,22] the region of possible values of the parameter j narrows. Instead of (3.17) we have

$$0 \leqslant \min(v_1,v_2) \equiv j_-^{\rm SM} \leqslant j^{\rm SM} \leqslant j_+^{\rm SM} \equiv \max(v_1,v_2) \leqslant 1. \tag{3.21}$$

At $v_1=v_2$ this implies $j^{\rm SM}=1/2$, that is, only one structure in the inhomogeneous medium satisfies the Dykhne's requirement of total statistical symmetry.[8] From (3.20) and (3.21) we see that in nontrivial cases ($v_2 \neq 0,1$) the boundary points in (3.13) are unattainable for symmetric media. These points describe the conductive properties of an inhomogeneous medium formed by an ensemble of double-layer composite spheres ($\bar{g}=\frac{1}{3}$) filling the volume under consideration and leaving no empty space.[6] In view of what has been said, Hashin and Shtrikman's solution[6] is not

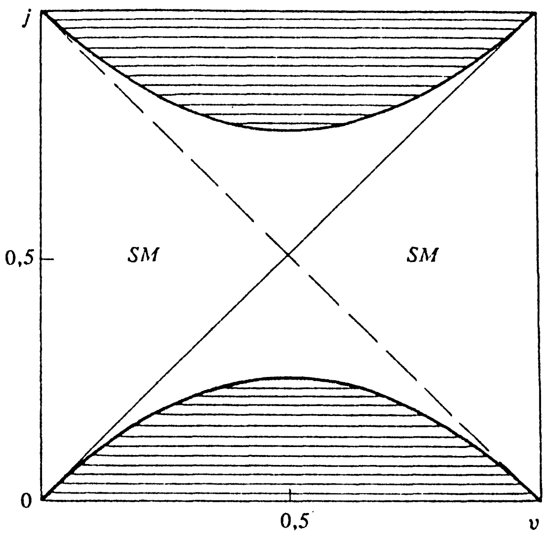

FIG. 1. The set of values of the geometric parameter j. The concentration dependence of parameter j^{SCM} for self-consistent media. The set of values of j^{SM} describing symmetric media.

realized in the class of symmetric media. Following Miller's terminology,[9] we will call such media asymmetric.

Figure 1 shows the range of values of the geometric parameter j realizing all possible structures of inhomogeneous media. The diagonals of the square specify the range of values of j describing various structures belonging to the class of symmetric media. The hatched regions bounded by parabolas and the horizontal sides of the square describe structures not realizable in the model of Miller, Beran, and Silnutzer.[9,19,22] The diagonal of the square depicted by a solid line specifies the set of values of j describing structures that belong to the class of self-consistent media (see below). The point of intersection of the diagonals corresponds to a unique solution, a random[8] or completely symmetric[11] Dykhne medium.

4. KELLER'S THEOREM AND DYKHNE'S TRANSFORMATIONS

Keller's theorem[7] has been proved for the case of a two-dimensional mixture of two isotropic components: inclusions of the same shape and size are distributed in the lattice in such a way that their centers form a rectangular lattice whose symmetry axes coincide with those of the inclusions. According to Keller,[7] the xth component $\sigma^*_{xx}(\sigma_1,\sigma_2)$ of the effective conductivity tensor $\sigma_*(\sigma_1,\sigma_2)$ of a medium whose lattice has conductivity σ_1 and whose inclusions have conductivity σ_2 is related to the yth component $\sigma^*_{yy}(\sigma_2,\sigma_1)$ of the tensor $\sigma_*(\sigma_2,\sigma_2)$ of a medium with a lattice conductivity σ_2 and an inclusion conductivity σ_1 by the formula

$$\sigma^*_{xx}(\sigma_1,\sigma_2)\sigma^*_{yy}(\sigma_2,\sigma_1)=\sigma_1\sigma_2,\quad \sigma_1\leqslant\sigma_2, \tag{4.1}$$

the intrinsic geometries of the media being the same. Dykhne[8] discovered a symmetry transformation that connects these media. Using Dykhne's method,[8] Balagurov[14] demonstrated that Eq. (4.1) is valid for less stringent restrictions imposed on the geometry of the inclusions. Mendelson[10] did the same by a different method.

We introduce the notation

$$\sigma^*_i\equiv\sigma_*(\bar g_{ii})=\sigma_*(\sigma_1,\sigma_2;v_1,v_2;\bar g_{ii}),\quad i\in[1,3],$$
$$\tilde\sigma^*_i\equiv\sigma_*(\sigma_2,\sigma_1;v_1,v_2;\bar h_{ii}),\quad \bar h_{ii}=1-\bar g_{ii},\quad \sum_{i=1}^{3}\bar g_{ii}=1, \tag{4.2}$$

where the $\bar g_{ij}$ are the components of the depolarization tensor $\bar g$ (Refs. 22 and 23) that determines the symmetry properties of tensor σ_* which can be shown to decrease as $\bar g$ increases:[25]

$$\frac{\partial\sigma_*}{\partial\bar g}\leqslant 0. \tag{4.3}$$

For a two-dimensional medium Eqs. (4.1) and (4.2) yield

$$\sigma^*_{xx}\equiv\sigma^*_1=\sigma_*(\bar g),\quad \sigma^*_{yy}\equiv\sigma^*_2=\sigma_*(\bar h),$$
$$\bar g\equiv\bar g_{11},\quad \bar h\equiv\bar g_{22}=1-\bar g, \tag{4.4}$$
$$\sigma^*_{yy}(\sigma_2,\sigma_1)\equiv\sigma_*(\sigma_2,\sigma_1;v_1,v_2;\bar g_{22})\equiv\tilde\sigma^*_1=\tilde\sigma_*(\bar g).$$

In this notation Eq. (4.1) assumes the form

$$\sigma_*(\bar g)\tilde\sigma_*(\bar g)=\sigma_1\sigma_2. \tag{4.5}$$

Solution σ_s specified in (3.13) obeys the following relationship:[5,23]

$$\sigma_c\in[0,\infty]\Rightarrow\sigma_s\in[\langle\rho\rangle^{-1},\langle\sigma\rangle], \tag{4.6}$$

which means that σ_s describes all possible real structures of inhomogeneous media. Hence, it must satisfy Keller's theorem (4.5). We write (3.13), as we did (3.18c), as

$$\sigma_s=\langle\sigma\rangle-\frac{\bar g D_\sigma}{\bar g[\sigma]+\bar h\sigma_c}=\frac{\bar g\sigma_1\sigma_2+\bar h\sigma_c\langle\sigma\rangle}{\bar g[\sigma]+\bar h\sigma_c}. \tag{4.7a}$$

Allowing here for the definition (4.2), we obtain

$$\tilde\sigma_s=\frac{\bar h\sigma_1\sigma_2+\bar g\tilde\sigma_c[\sigma]}{\bar h\langle\sigma\rangle+\bar g\tilde\sigma_c},\quad \sigma_1\leftrightarrow\sigma_2\Rightarrow[\sigma]\leftrightarrow\langle\sigma\rangle. \tag{4.7b}$$

Substituting (4.7a) and (4.7b) into (4.5), we arrive at the equality

$$\sigma_c\tilde\sigma_c=\sigma_1\sigma_2, \tag{4.8}$$

which the parameters σ_c and $\tilde\sigma_c$ of "mutual" media (in the terminology of Ref. 14) must obey.

Let us show that Keller's theorem in the form (4.5) is applicable to one-dimensional structures—layered media. Putting $\bar g=1$, we obtain from (4.7) and (4.2) a relation for the conductivity in the direction normal to the layers:

$$\bar g=1,\quad \bar h=0\Rightarrow\sigma_*=\frac{\sigma_1\sigma_2}{[\sigma]}=\langle\rho\rangle^{-1},\quad \tilde\sigma_*=[\sigma]. \tag{4.9a}$$

Similarly, putting $\bar g=0$, we obtain from (4.7) and (4.2) a relation for the conductivity in the direction parallel to the layers:

$$\bar g=0,\quad \bar h=1\Rightarrow\sigma_*=\langle\sigma\rangle,\quad \tilde\sigma_*=\sigma_1\sigma_2\langle\sigma\rangle^{-1}=[\rho]^{-1}. \tag{4.9b}$$

Thus, "one-dimensional" random inhomogeneous media satisfy Keller's theorem. Comparing (4.9) with (3.9b), we conclude that the best boundaries for $n=1$ agree with (4.5). The case of $n=0$ (3.9a) is trivial.

Let us now study the boundary points $\sigma_\pm^{(2)}$ (3.13), for which, according to (4.7), we write

$$\sigma_+^{(2)}=\sigma_2\frac{\bar{g}\sigma_1+\bar{h}\langle\sigma\rangle}{\bar{g}[\sigma]+\bar{h}\sigma_2},\quad \tilde{\sigma}_+^{(2)}=\sigma_1\frac{\bar{h}\sigma_2+\bar{g}[\sigma]}{\bar{h}\langle\sigma\rangle+\bar{g}\sigma_1},\qquad(4.10a)$$

$$\sigma_-^{(2)}=\sigma_1\frac{\bar{g}\sigma_2+\bar{h}\langle\sigma\rangle}{\bar{g}[\sigma]+\bar{h}\sigma_1},\quad \tilde{\sigma}_-^{(2)}=\sigma_2\frac{\bar{h}\sigma_1+\bar{g}[\sigma]}{\bar{h}\langle\sigma\rangle+\bar{g}\sigma_2}.\qquad(4.10b)$$

These yield the following equalities:

$$\sigma_\pm^{(2)}\tilde{\sigma}_\pm^{(2)}=\sigma_1\sigma_2,\qquad(4.11)$$

which imply that the best boundaries for the case of $n=2$ obey (4.5). At $\bar{g}=\frac{1}{3}$ the values $\sigma_\pm^{(2)}$ describe the possible real three-dimensional structures calculated in Ref. 6.

The boundary points $\sigma_\pm^{(3)}$ (3.19) allow for three-particle interactions. Hence, the model for which they are exact solutions must contain "three degrees of freedom," that is, must either be three-component or contain an additional parameter describing the orientation of nonspherical inclusions. In either case such a medium will not satisfy the restrictions of Keller's theorem, which is "linked" with two-component systems.

5. SELF-CONSISTENT MEDIUM

We consider below a particular case of a symmetric medium described by the effective conductivity tensor σ_* calculated via (3.13) with $\sigma_c=\sigma_*$. As can be shown,[23] in this case σ_* is the solution of the equation

$$\langle(\sigma-\sigma_*)(\sigma+\bar{t}\sigma_*)^{-1}\rangle=0.\qquad(5.1)$$

For a mixture of two isotropic components we obtain from Eq. (5.1) [or, which is simpler, from (4.7a)] the equation

$$\bar{t}\sigma_*^2-(\bar{t}\langle\sigma\rangle-[\sigma])\sigma_*-\sigma_1\sigma_2=0,\qquad(5.2)$$

whose solution

$$\sigma_{SC}\equiv\sigma_*=a+\sqrt{a^2+b},\quad 2\bar{t}a\equiv\bar{t}\langle\sigma\rangle-[\sigma],\quad \bar{t}b\equiv\sigma_1\sigma_2\qquad(5.3)$$

for different values of $\bar{t}$ determines the principal values of σ_{SC}. Solution (5.3) describes macroanisotropic self-consistent media and is the most general solution of this type obtained so far. The anisotropy in macroscopic conductivity is due either to the deviation (on the average) of the shape of inhomogeneity grains from spherical or to anisotropy in the spatial distribution of the grains.[14,23] For a macroisotropic medium ($\bar{t}=d-1$) the solution in the form (5.3) was obtained earlier by Bruggeman.[5] Later it was duplicated by various researchers (see, e.g., Refs. 16 and 17).

Solution (5.3) allows implicitly for many-particle interactions. Moreover, it is symmetric in its components, in view of which a medium described by σ_{SC} belongs to the class of symmetric media. Following Ref. 13, we call a medium symmetric if its properties are invariant under the inversion:

$$(\sigma_1,v_1)\leftrightarrow(\sigma_2,v_2).\qquad(5.4)$$

This invariance imposes fairly stringent restrictions on the statistical properties of the medium,[13,14] and, hence, according to (3.21), the range of values of parameter j^{SM} narrows.

Let us find the set of the values j^{SCM} of parameter j^{SM} that describe self-consistent media. Using the definition of σ_{SC} and Eq. (3.9b), we can write

$$\langle\rho\rangle^{-1}\leqslant\sigma_{SC}=\sigma_c\leqslant\langle\sigma\rangle,\quad \sigma_c\equiv\langle\sigma\rangle_j=\sigma_1+j(\sigma_2-\sigma_1),\qquad(5.5a)$$

which together with the reverse inequalities

$$\langle\sigma\rangle^{-1}\leqslant\rho_{SC}=\rho_c\leqslant\langle\rho\rangle,\quad \rho_c\equiv\langle\rho\rangle_j=\rho_1+j(\rho_2-\rho_1),\qquad(5.5b)$$

yields the following formula for j:

$$j=v_2\equiv j^{SCM}.\qquad(5.6)$$

In Fig. 1 the set of the values of j^{SCM} that describe self-consistent media is depicted by the solid diagonal of the square. If we express the solution σ_{SC} (5.3) in the form (5.5a), we find that the value of parameter j that must be substituted into (5.5a) to obtain (5.3) does not exceed v_2. The explanation is that solution (5.3) allows for interactions of any multiplicity, while the boundary points specified in (3.18) and (5.5) allow only for three-particle interactions.

Among the self-consistent solutions there is a unique one, totally symmetric, that is invariant under the interchange $\sigma_1\leftrightarrow\sigma_2$:

$$v_1=v_2\Rightarrow\sigma_{SC}(\sigma_1,\sigma_2)=\sigma_{SC}(\sigma_2,\sigma_1).\qquad(5.7)$$

Media of this type were considered by Frisch,[26] Dykhne,[8] Mendelson,[10] Schulgasser,[11] the present author,[13] Balagurov,[14] and Shvidler.[15] In Fig. 1 the point of intersection of the diagonals corresponds to a totally symmetric medium. Combining (5.7) with (5.3), we find that

$$\sigma_{SC}=a+\sqrt{a^2+b},\quad 4\bar{t}a\equiv(\bar{t}-1)(\sigma_1+\sigma_2),\quad \bar{t}b=\sigma_1\sigma_2.\qquad(5.8)$$

For a macroisotropic medium ($\bar{t}=d-1$) this yields

$$8\sigma_{SC}=\sigma_1+\sigma_2+\sqrt{(\sigma_1+\sigma_2)^2+32\sigma_1\sigma_2},\quad d=3,\qquad(5.9a)$$

$$\sigma_{SC}=\sqrt{\sigma_1\sigma_2},\quad d=2.\qquad(5.9b)$$

Expressing (5.9b) in the form (5.5a), we can write

$$\sigma_{SC}=\sqrt{\sigma_1\sigma_2}\equiv\sigma_1+R_*(\sigma_2-\sigma_1)\Rightarrow R_*=\frac{\sqrt{\sigma_1}}{\sqrt{\sigma_1}+\sqrt{\sigma_2}}.\qquad(5.10)$$

Here the substitution of R_* for j is due to allowing for all many-particle interactions in the exact solution (5.3). In Eq. (3.18a), where the structural parameter j is used in accordance with its definition (3.17), we have allowed only for three-particle interactions. The parameter R_* (5.10) is interpreted in a broader sense than j.

In terms of parameter R_* (5.10), the boundary points (3.9) and (3.19) assume the following form:

$$n=0:\quad 0\leqslant R_*\leqslant 1, \tag{5.11a}$$

$$n=1:\quad \frac{x}{1+x}\leqslant R_*\leqslant\frac{1}{2},\quad \frac{\sigma_1}{\sigma_2}\equiv x\in[0,1], \tag{5.11b}$$

$$n=2:\quad \frac{2x}{1+3x}\leqslant R_*\leqslant\frac{1+x}{3+x}, \tag{5.11c}$$

$$n=3:\quad \frac{x(3+x)}{(1-x)^2+8x}\leqslant R_*\leqslant\frac{1+3x}{4(1+x)}, \tag{5.11d}$$

where for $n\geqslant 1$ we have allowed for the following equations:

$$v_2=\bar{h}=j=\frac{1}{2},\quad R_*=\frac{\sqrt{x}}{1+\sqrt{x}}. \tag{5.12}$$

In the general case we can obtain in the same manner as we did in (5.11) a set of boundary points $R_\pm^{(n)}$ for successively narrowing regions of variation of parameter R_*.

6. CRITICAL BEHAVIOR OF THE CONDUCTIVITY OF SELF-CONSISTENT MEDIA NEAR THE PERCOLATION THRESHOLD

In papers on percolation theory (see, e.g., Refs. 8, 12, 14, 16, 17, and 26) systems whose conductivities are restricted by the inequality $x\ll 1$ are of special interest. The behavior of conductivity σ_* near the percolation threshold $v_2=v_c$ resembles the behavior of the order parameter in a second-order phase transition.[12]

Let us consider the asymptotic behavior of solution (5.3) for $x\ll 1$:

$$\frac{\sigma_*}{\sigma_2}\equiv\bar{\sigma}_*(x)=\bar{\sigma}_*(0)+Ax-Bx^2,\quad x\ll x_c, \tag{6.1a}$$

$$\bar{\sigma}_*(0)\equiv\frac{v_2-\bar{g}}{\bar{h}}H(v_2-\bar{g}),$$

$$A\equiv\frac{\bar{h}-v_2}{\bar{h}}H(v_2-\bar{g})+\frac{\bar{g}}{|v_2-\bar{g}|},$$

$$B\equiv\frac{v_1v_2\bar{g}}{|v_2-\bar{g}|^3},\quad H(y)=\begin{cases}1 \text{ if } y\geqslant 0,\\ 0 \text{ if } y<0,\end{cases}\quad 2x_c=\frac{(v_2-\bar{g})^2}{v_1v_2+\bar{g}\bar{h}}. \tag{6.1b}$$

We choose the statistical parameter $\bar{g}$, whose size and symmetry properties are determined by the nature of the spatial distribution of the regions occupied by the two components, to be the parameter that plays the important role in the description of the anomalous behavior of $\bar{\sigma}_*$. The depolarization tensor $\bar{g}$ is equal to the singular component of the second derivative of the Green function (2.6) calculated for the effective grain.[23]

The existence of a critical point $v_2=v_c$ at which the structure of the medium is essentially altered suggests that the parameter $\bar{g}$ determined by this structure depends on v_2 and that this dependence changes significantly at point $v_2=v_c$.

Let us introduce the notation

$$\bar{g}(v)=\bar{g}_+\equiv\frac{v-\xi^t}{1-\xi^t},\quad v_2\equiv v\geqslant v_c, \tag{6.2a}$$

$$\bar{g}(v)=\bar{g}_-(v)\equiv\frac{v}{1-\xi^q},\quad v\leqslant v_c,\quad |v-v_c|\equiv\xi\ll 1, \tag{6.2b}$$

where t and q are the critical exponents of percolation theory.[12] At $d=2$ and $t=q$ the functions $\bar{g}_\pm(v)$ are related thus:

$$\bar{g}_-(v_1)+\bar{g}_+(v)=1,\quad v_1+v_2=1,\quad d=2. \tag{6.3}$$

Combining (6.1) and (6.2), we arrive at the following expression for conductivity $\sigma_*(x)$ in the neighborhood of the critical point $v=v_c$:

$$\bar{\sigma}_*=\begin{cases}(v-\bar{g}_+)/(1-\bar{g}_+) \text{ if } v\geqslant v_c,\\ x\bar{g}_-/(\bar{g}_--v) \text{ if } v<v_c.\end{cases} \tag{6.4}$$

Another principal value of the effective conductivity tensor in the case of $d=2$ is obtained from (6.4) and (6.3) via the substitution

$$\bar{g}_+(v)\to\bar{h}_+(v)=1-\bar{g}_+(v)=\bar{g}_-(1-v),\quad d=2. \tag{6.5}$$

This implies that when $v>v_c$, the following clusters are formed: (1) a conductive ("metallic") cluster along the field, and (2) a nonconductive cluster (an "insulator") normal to the field. For the three-dimensional problem, the fact that

$$\bar{h}_{11}=\bar{g}_{22}+\bar{g}_{33},\quad \bar{g}_{11}\neq\bar{g}_{\alpha\alpha},\quad \alpha\neq 1,\quad d=3, \tag{6.6}$$

complicates the relationship between $\bar{g}_+$ and $\bar{g}_-$. However, in this case, too, with each conductive cluster we can associate a nonconductive one.

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