Macroscopic conductivity of random inhomogeneous media. Calculation methods

A G Fokin

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Abstract. The macroscopic conductivity of inhomogeneous media such as polycrystals, composites, etc., is discussed. One of the major global parameters of a randomly inhomogeneous medium (RIM), the effective (macroscopic) conductivity tensor (ECT), is considered. Based on functional analysis ideas, particularly the projector and orthogonal reduced field concepts, a method for obtaining bounds on ECT is developed. A criterion is given by which a position of each iteration solution relative to the preceding one is determined. It is shown that previous model ECT results fall within, and are in fact special cases, of the scheme proposed by the author. The importance of the auxiliary, zero-fluctuation parameter σ^{c} in constructing convergent series for quantities of interest is established. Extended versions of the Hashin-Shtrikman variational principles and Keller's theorem are obtained. The structural parameters introduced for describing an RIM are expressed, in the proposed method, in terms of the *n*-point probabilities (*n*-point interactions) for the random local conductivity field. The piecewise uniform 'polarised' field approximation is combined with classical energy theorems to obtain the ECT bounds best achievable within the npoint RIM description.

A G Fokin Moscow Institute of Elecronic Technology (Technology University), K498 MIET, 103498 Moscow, Russia Tel. (7-095) 532 99 63 Fax (7-095) 530 22 33

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1. Introduction

The use of materials with complex structure (ceramics, composites, polycrystals, emulsions, etc.) in science and technology poses a number of specific problems for which the inhomogeneity of the medium is responsible. Inhomogeneities are understood to be deviations of local values of material characteristics [the specific conductivity tensor $\sigma = \sigma(\mathbf{r})$ in the case of conduction] of the medium from the prescribed values. We shall designate these deviations as follows:

$$\sigma' \equiv \sigma - \sigma^{\rm c} \,, \tag{1.1}$$

where σ^c is the specific conductivity tensor (conductivity tensor) of an auxiliary medium (a comparison medium) differing from the inhomogeneous one only in material properties. In the case of a random inhomogeneous medium it is a random tensor field.

Problems of macroscopic description of random inhomogeneous medium (RIM) draw an incessant attention of scientists [1-52]. This is, first, because these media differ essentially from homogeneous and regular inhomogeneous ones and, thus, they present additional mathematical interest and, second, because the results can be put to practical use. One of the major problems is to find macroscopic (effective) characteristics of RIM based on information available.

The solution of this problem is sought in three main lines of inquiries:

(1) calculation of approximate values;

(2) evaluation of bounds in which effective characteristics of the medium lie;

(3) search of exact solutions for real media and for model structures.

To describe macroscopical conduction of an RIM the effective conductivity tensor $\hat{\sigma}^*$ is used. It is specified by the equation:

$$\langle \mathbf{J} \rangle = \langle \sigma \mathbf{E} \rangle \equiv \hat{\sigma}^* \langle \mathbf{E} \rangle \,, \tag{1.2}$$

where angular brackets mean averaging over the ensemble of realisations, which is the same as averaging over the volume V of the medium provided that specific conditions are satisfied [8, 14, 20, 30, 31, 37]. Generally $\hat{\sigma}^*$ is an integral operator whose kernel $\sigma^*(\mathbf{r}_1, \mathbf{r}_2)$ is introduced by the equation:

$$\langle \mathbf{J}(\mathbf{r}_1) \rangle \equiv \int \sigma^*(\mathbf{r}_1, \mathbf{r}_2) \langle \mathbf{E}(\mathbf{r}_2) \rangle \, \mathrm{d}\mathbf{r}_2 \,.$$
 (1.3)

However, if

(1) the characteristic size of RIM is large enough in comparison with the scale of inhomogeneity of the average field $\langle \mathbf{E} \rangle$, the free path of charge carriers, and the characteristic size of the region the average over which represents the ensemble average;

(2) the RIM is statistically uniform, i.e. *n*-point probabilities of the random field $\sigma(\mathbf{r})$ are invariant under translation;

(3) the boundary conditions on the surface S bounding the volume V of the medium in question are macroscopically homogeneous [38]:

$$\varphi(\mathbf{r}) = -\mathbf{r} \cdot \langle \mathbf{E} \rangle, \quad \mathbf{r} \in S, \tag{1.4}$$

for the Dirichlet problem or

$$J_n(\mathbf{r}) = \mathbf{n} \cdot \langle \mathbf{J} \rangle, \quad \mathbf{r} \in S, \tag{1.5}$$

for the Neumann problem (here $\langle \mathbf{E} \rangle$ and $\langle \mathbf{J} \rangle$ are constant vectors) and the electric field intensity \mathbf{E} is related to the current density \mathbf{J} through the Ohm law; then the operator $\hat{\sigma}^*$ is local [12]:

$$\hat{\sigma}^* = \sigma^* \hat{I}, \quad \sigma^*(\mathbf{r}_1, \mathbf{r}_2) = \sigma^* \delta(\mathbf{r}_1 - \mathbf{r}_2), \quad (1.6)$$

where σ^* is the effective (macroscopic) conductivity tensor (ECT).

The simplest exact solutions were obtained by Wiener in 1912 (see Refs [2, 9]). In our notation they take the form:

$$\sigma_{11}^* = \langle \sigma \rangle, \quad \sigma_{33}^* = \langle \rho \rangle^{-1}; \quad \sigma \rho = 1, \qquad (1.7)$$

$$\langle \Gamma \rangle \equiv \sum_{a=1}^{N} v_a \Gamma_a , \qquad \sum_{a=1}^{N} v_a = 1 ; \qquad \Gamma \equiv \sigma, \rho , \qquad (1.8)$$

$$\sigma_{ij}^{*} = \sigma_{11}^{*} \delta_{ij} + (\sigma_{33}^{*} - \sigma_{11}^{*}) \delta_{i3} \delta_{j3}, \quad \sigma_{ij}(\mathbf{r}) = \sigma(\mathbf{r}) \delta_{ij}.$$
(1.9)

Here σ^* is the ECT of an unbounded layered medium with the following properties:

(1) planes between the plane-parallel layers are orthogonal to the third axis;

(2) the conductive properties of a layer are described by the tensor σ (1.9) for one of N homogeneous isotropic components;

(3) the thickness of a layer and the number of a component, on which σ depends, are random numbers.

The values σ_{11}^* and σ_{33}^* constitute bounds

$$\langle \rho \rangle^{-1} \equiv \sigma_{\mathbf{W}}^{-} \leqslant \sigma^{*} \leqslant \sigma_{\mathbf{W}}^{+} \equiv \langle \sigma \rangle; \quad \sigma_{ij}^{*} \equiv \sigma^{*} \delta_{ij}, \quad (1.10)$$

in which the effective conductivity σ^* of a macroisotropic mixture of homogeneous and isotropic components lies. The bounds in (1.10) take into account the information on RIM presented by one-point probabilities [15, 29].

The Keller theorem [13] and the Dykhne symmetry transform [20] have given impetus to investigations of symmetry properties of inhomogeneous medium. A number of interesting results have been obtained and structures for which exact solutions exist have been found [19, 22, 25, 26, 28, 29, 32, 33, 37, 39, 40].

Bruggeman have found [2] self-consistent solutions for two- and three-dimensional macroisotropic mixtures of two homogeneous isotropic components. These solutions are often used for interpretation of measurements as well as in theory of percolation [7, 11, 24, 27, 32, 33, 38, 39].

I M Lifshits with his co-workers developed a method of calculation for static [4] and dynamic [6] effective characteristics in theory of elasticity. This method proposes to solve differential equations coefficients of which are random tensor fields. The solution of the problem is presented as a series, each term of which describes interaction of the appropriate multiplicity. The method of the random field theory is fruitful in calculation of approximate values as well as in selection of auxiliary fields used in variational methods [8, 11, 12, 14, 21, 30-37, 41].

Based on the classical energy inequalities Hashin and Shtrikman [9, 10, 38] developed a variational method for calculating the bounds σ_{HS}^{\pm} which, unlike the bounds σ_{W}^{\pm} in (1.10) found by Wiener, account for the additional statistic information on RIM contained in two-point probabilities. Three-particle interactions were partly incorporated by Beran [14]. This made it possible to further narrow the bounds in the case of two-dimensional [22] and three-dimensional [19] macroisotropic mixtures of two homogeneous isotropic components.

Below the symbols $\sigma_{(n)}^{\pm}$ are used to denote the best bounds for σ^* calculated with regard for interactions of multiplicity $k \leq n$. In other words the statistic information on RIM is presented by *n*-point probabilities. In this notation the Wiener and Hashin–Shtrikman bounds are

$$\sigma_{\mathrm{W}}^{\pm} \equiv \sigma_{(1)}^{\pm}, \qquad \sigma_{\mathrm{HS}}^{\pm} \equiv \sigma_{(2)}^{\pm}. \tag{1.11}$$

Note that only recently it has been clearly understood that the problems of description are common for different transport (and similar) phenomena at the macroscopic level. Previously, each particular problem was solved (and often it is solved nowadays) by a specific method developed especially for it. It is safe to say that the diversity of phenomena and structures of RIM gave rise to the diversity of methods, applications, and models.

As an example of structures that can be described macroscopically within the scope of a single approach to RIM, a polycrystal and a composite are shown schematically in Fig. 1. The regions marked by dashed lines qualitatively present the microstructure of an RIM. In the first case the grain (region) of inhomogeneity is a crystallite and in the second case it is an isotropic ellipsoidal inclusion. The orientation of crystalophysical axes of the crystallite (Fig. 1a) or principal axes of the ellipsoid (Fig. 1b) governs the





response of the grain of inhomogeneity to an outer field. Arrows indicate vectors $\langle \mathbf{J} \rangle$ in macroscopical and \mathbf{J} in microscopical descriptions of RIM. In view of the boundary conditions the vector \mathbf{J} changes jumpwise from one grain to another. The electric field intensity \mathbf{E} may be presented similarly.

Some terms of statistical physics can be very useful in solving the problem of how macroscopic material characteristics $\hat{\sigma}^*$ from (1.2) of inhomogeneous medium depend on the random field of respective microcharacteritics $\sigma(\mathbf{r})$. In particular, it is convenient to call the overall bulk of information about the medium (see above) the microstate of this medium (system). The latter depends on the topological properties of the space of RIM and the material properties of the substance filling the space. The internal structure of the medium is given by the spatial distributions of inhomogeneities, i.e., by their sizes and shapes (when interfaces are clearly defined) and by the proper distribution functions. Similarly, material characteristics of the substance are described. In terms of state the transition from $\sigma(\mathbf{r})$ to $\hat{\sigma}^*$ is reduced to the description of the macrostate of the medium on the basis of the available information about its microstates. Fluctuations of the random field $\sigma(\mathbf{r})$ can be expressed in terms of fluctuations of the corresponding microparameters, which, in turn, determine the changes in correlating macroparameters.

The spread of values of the random field $\sigma(\mathbf{r})$ (the difference between the minimum and the maximum) is one of the main factors on which the intensity of fluctuations depends. Below I use the parameter $\varkappa \equiv \sigma^-/\sigma^+$, where $\sigma^- \equiv \inf \sigma$ and $\sigma^+ \equiv \sup \sigma$, as a measure for the spread of conductivity [see also (6.15b) and (6.14b)]. In the case when the notion of the grain of inhomogeneity is valid, i.e., there are regions in which $\sigma(\mathbf{r}) = \text{const}$, the value of $\sigma(\mathbf{r})$ changes jumpwise on the interface between two grains, and σ^- and σ^+ are, respectively, the least and greatest principal (eigen) values of the tensor $\sigma(\mathbf{r})$. One-phase polycrystals and mechanical mixtures (composites) of two isotropic components are the most popular materials for investigations. In the first case $\varkappa = \sigma_1/\sigma_3$ ($\sigma_1 \leq \sigma_2 \leq \sigma_3$), where σ_k is the principal

value of the tensor of conductivity of crystallite. In the second case $\varkappa = x$ if $\sigma_1/\sigma_2 \equiv x < 1$ and $\varkappa = 1/x$ if x > 1. Here σ_1 and σ_2 are scalar conductivities of the first and second components, respectively. If the parameter \varkappa takes a value near the limits of the interval $\varkappa \in [0, 1]$, then fluctuations of conductivity are said to be small $(1 - \varkappa \ll 1)$ or large $(\varkappa \ll 1)$. In the last case \varkappa can be presented in the form $\varkappa \equiv 10^{-\nu}$, where $\nu \ge 1$. In this case the region $v \in [1, 3]$ is realised in the system, the state of which is far from the phase transition (or from a similar state in a sense) [1, 53-58]. Under specific conditions depending on other parameters (for example, on volume concentrations of components) the further increase of v $(v \ge 10)$ can lead to what we shall call gigantic fluctuations. Such a situation takes place in binary systems [7, 20, 28] (composites provide an example of such a system [59, 60]). Different anomalous effects in conductive inhomogeneous media near the percolation threshold [24, 28, 51, 57, 60-67)and those induced by a magnetic field [28, 46, 57, 60, 61, 64-66, 68 - 74] draw a great attention.

The spatial distribution of inhomogeneities is another factor on which the intensity of fluctuations of conductivity depends in a random inhomogeneous medium, but information about the real distribution is too scarce and hard to gain. The simplest and most widely used part of it is information about the sizes and shapes of grains of inhomogeneities or spatial scales of correlations [30, 75]. According to available data [53, 76-81] the mean size of a grain of inhomogeneity (for a polycrystal or a composite) varies from 1 to $10^3 \,\mu m$ depending on the system. The mean size (diameter) is obtained by averaging over the sizes of grains in all directions. In the absence of mechanical texture (anisotropy of distribution of sizes of grain in Euler angles) the mean (effective) grain has the shape of a ball (spatial problem) or that of a circle (plane problem) [21, 23, 37]. How to use information on distribution of grains in sizes and shapes to describe macroscopic properties of an RIM presents an interesting problem [82], the adequate solution of which has not been found yet. Note that the ellipsoidal shape of a grain has been accounted for in calculations of different macroscopic characteristics of an RIM since, supposedly, Fricke [1].

The existence of correlations between the structure (microstate) of inhomogeneous medium and its macroscopical properties poses a number of important practical problems: the choice of optimal structure of an RIM to provide the desired parameters of materials [31, 48, 51, 56, 58, 64, 73, 74, 76–79, 84–88], adequate model description of different systems being investigated (artificial as well as natural) [1, 53, 62-92], etc.

For calculation of σ^* and $\sigma^{\pm}_{(n)}$ this paper develops the method proposed in Refs [27, 29, 34–37, 83]. This method takes advantages of the method of random field theory [4, 6, 8, 11, 12, 21, 30] as well as those of the variational method [9, 10, 14, 19, 22, 31, 38].

In Sections 2–4 the method of integral equations and projecting operators (projectors) is developed to calculate the physical fields **J** and **E** and the ECT for inhomogeneous media. Introduction of the reduced material characteristics $\bar{\sigma}$, $\bar{\rho}$ and physical fields **j**, **e** (Section 3) makes it possible to use two (mutually dual) iterative procedures to solve one boundary-value problem. These procedures become independent with the appropriate choice of the auxiliary parameters σ^c and ρ^c . In Section 4 the sufficient convergence conditions (SCC) are obtained for mutually dual iterative procedures using the projector formalism. In Section 5 general properties of ECT are studied. In Section 6 various variants of construction of bounds for $\sigma_{(n)}^{\pm}$ are considered when there is only a limited information on an RIM. Isolation of local (singular) component of projectors leads to a singular approximation (Section 7). Analytical results of the latter section are used in other methods (for example, in variational methods). In Section 8 variational principles are formulated on the basis of classical energy theorems. With the restrictions on the parameters σ^{c} and ρ^{c} they are reduced to the generalised Hashin-Shtrikman variational principles (Section 8.3). In Section 8.4 the Beran approximation is considered. In Section 9 I consider in detail how piecewise uniform 'polarised' fields τ and η can be applied in construction of bounds for $\sigma_{(n)}^{\pm}$. In Section 10 exact solutions are reviewed: (1) completely symmetric Dykhne media [20]; (2) self-consistent solutions; (3) new solutions in the Dykhne-Mendelson form. The generalised Keller theorem [13] is obtained. Section 11 is dedicated to the study of role of structural parameters characterising the interactions between inhomogeneities. In Sections 10-12 the case of the mixture of two homogeneous isotropic components is considered in detail. The correlation between statistic properties of RIM and density dependence of parameter *j* describing the contribution of three-particle interactions into $\sigma_{(3)}^{\pm}$ is established. In Section 12 Miller's models of asymmetric and symmetric cell materials are analysed [19].

2. Transition to integral equations

Equations of steady current in the absence of an outer magnetic field are

$$\nabla \times \mathbf{E} = 0, \qquad (2.1)$$

$$\nabla \cdot \mathbf{J} = 0. \tag{2.2}$$

Equations (2.1) and (2.2) can be rewritten in the form:

$$\mathbf{E} = -\nabla \varphi \,, \tag{2.3}$$

$$\mathbf{J} = \nabla \times \boldsymbol{\Psi} \,. \tag{2.4}$$

Here φ and ψ are, respectively, scalar and vector potentials. To solve (2.1) and (2.2) together, the material equation — the Ohm law — is used in one of two forms:

$$\mathbf{J} = \sigma \mathbf{E}, \qquad J_k = \sigma_{ki} E_i; \qquad \sigma = \sigma(\mathbf{r}), \qquad (2.5)$$

$$\mathbf{E} = \rho \mathbf{J}, \qquad E_i = \rho_{ik} J_k; \qquad \rho = \rho(\mathbf{r}), \qquad (2.6)$$

where the random tensor fields σ and ρ are related by the equations:

$$\sigma \rho = I, \qquad \sigma_{ik} \rho_{ki} = \delta_{ij} \,. \tag{2.7}$$

In addition, the boundary conditions must be given for the tangent component of the field \mathbf{E} and for the normal component of the field \mathbf{J} on the surface S bounding the volume V of the medium.

Two mutually dual schemes of solutions are possible for Eqns (2.1) and (2.2):

(1) σ -scheme based on presentations (2.3) and (2.5);

(2) ρ -scheme based on presentations (2.4) and (2.6).

The first scheme is simpler and, as a result, more popular, though both schemes are equal in all other respects. However, it can be shown that each scheme can be translated into the other. This will be shown below by the example of the σ -scheme.

In σ -scheme Eqns (2.1) and (2.2) are reduced with regard for (2.3) and (2.5) to the Laplace equations:

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

(2.8)

with the boundary conditions

$$\varphi(\mathbf{r}) = \varphi_0, \quad \mathbf{r} \in S_1; \quad J_n(\mathbf{r}) = J_0, \quad \mathbf{r} \in S_2,$$
$$J_n \equiv \mathbf{n} \cdot \mathbf{J}, \quad S_1 \cup S_2 = S, \quad (2.9)$$

where **n** is a unit vector of the external normal to the surface S bounding V.

To solve the problem posed by (2.8) and (2.9) we shall introduce the Green's function for the Poisson equation with uniform boundary conditions as in Ref. [93]:

$$L^{c}G(\mathbf{r},\mathbf{r}') = -\delta(\mathbf{r}-\mathbf{r}'), \qquad L^{c} = \nabla \cdot \sigma^{c} \nabla; \qquad \mathbf{r},\mathbf{r}' \in V,$$
(2.10)

$$\begin{aligned} \mathbf{G}(\mathbf{r}, \mathbf{r}') &= 0, \quad \mathbf{r} \in \mathbf{S}_1; \quad \mathbf{n} \cdot \sigma^* \mathbf{V} \mathbf{G}(\mathbf{r}, \mathbf{r}') = 0, \\ \mathbf{r} \in \mathbf{S}_2; \quad \mathbf{r}' \in V. \end{aligned}$$
(2.11)

Here index 'c' marks the quantities pertaining to the comparison medium (see Section 1), macroscopically identical to the RIM in question and different only in local values of the material characteristics.

The operator L^{c} (2.10) associated with the comparison medium is treated in the same way as in perturbation theory. Consequently, it complies with the same ordinary restrictions as those imposed on an unperturbed operator:

(1) the solution to the problem (2.8), (2.9) in the form

$$L^{c}\varphi^{c} = 0, \qquad L^{c} = \nabla \cdot \sigma^{c}\nabla, \qquad \mathbf{r} \in V,$$
 (2.12)

$$\varphi^{\mathrm{c}}(\mathbf{r}) = \varphi_0, \quad \mathbf{r} \in S_1; \quad J_n^{\mathrm{c}}(\mathbf{r}) = J_0, \quad \mathbf{r} \in S_2 \quad (2.13)$$

is known for L^c ;

(2) The perturbation operator $L' \equiv L - L^c$ is small in some sense and, hence, the perturbation series is convergent (see Section 4).

It is not difficult to see that (2.8) and (2.12) lead to the equation

$$L^{c}\varphi' = -L'\varphi, \quad \mathbf{r} \in V;$$

$$L' = \nabla \cdot \sigma'\nabla, \quad \varphi' \equiv \varphi - \varphi^{c},$$
(2.14)

with the boundary conditions

$$\varphi' = 0$$
, $\mathbf{r} \in S_1$; $J'_n = 0$, $\mathbf{r} \in S_2$; $\mathbf{J}' \equiv \mathbf{J} - \mathbf{J}^c$.
(2.15)

following from (2.9) and (2.13).

Let us consider the field $\delta \phi \equiv \phi'$ in the volume V of the comparison medium. Then the right-hand side of Eqn (2.14) presents a source generating the field $\delta \phi$. For the potential $\delta \phi$ the intensity $\delta \mathbf{E}$ and the current density $\delta \mathbf{J}$ are

$$\varphi' = \delta \varphi \Rightarrow \delta \mathbf{E} = -\nabla \delta \varphi = \mathbf{E}', \quad \delta \mathbf{J} = \sigma^c \delta \mathbf{E} \neq \mathbf{J}'.$$
(2.16)

In view of the boundary conditions (2.15) the potential $\delta \varphi$ vanishes on the part S_1 of the surface S, while the normal component of the vector $\delta \mathbf{J}$ is different from zero on the part S_2 of the surface S.

Introducing the 'polarised' current vector $\mathbf{T} \equiv \mathbf{J} - \sigma^{c} \mathbf{E} = \sigma' \mathbf{E}$ we can represent the problem (2.14)–(2.16) in the form:

$$L^{c}\delta\phi = -g, \quad g = -\nabla \cdot \mathbf{T}, \quad \mathbf{T} = \sigma' \mathbf{E}, \quad \mathbf{r} \in V,$$

$$(2.17)$$

$$\delta\phi = 0, \quad \mathbf{r} \in S_{1}; \quad \delta J_{n} = \mathbf{n} \cdot \delta \mathbf{J} = -\mathbf{n} \cdot \mathbf{T}, \quad \mathbf{r} \in S_{2}.$$

$$(2.18)$$

According to the generalised Green's formula [93] we have

$$\begin{split} \mathrm{d} V \big[G(\mathbf{r}, \mathbf{r}_1) L^{\mathrm{c}} \delta \phi(\mathbf{r}) &- \delta \phi(\mathbf{r}) L^{\mathrm{c}} G(\mathbf{r}, \mathbf{r}_1) \big] \\ &+ \oint \, \mathrm{d} \mathbf{S} \cdot \big[G(\mathbf{r}, \mathbf{r}_1) \delta \mathbf{J}(\mathbf{r}) + \delta \phi(\mathbf{r}) \sigma^{\mathrm{c}} \nabla G(\mathbf{r}, \mathbf{r}_1) \big] = 0 \end{split}$$

Whence, with regard for (2.10), (2.11), (2.17) and (2.18), we obtain the perturbed potential [35]:

$$\delta \phi(\mathbf{r}_1) = \int G(\mathbf{r}_2, \mathbf{r}_1) g(\mathbf{r}_2) \, \mathrm{d} V(\mathbf{r}_2) + \oint G(\mathbf{r}_2, \mathbf{r}_1) \mathbf{T}(\mathbf{r}_2) \cdot \mathrm{d} \mathbf{S}(\mathbf{r}_2) \,.$$
(2.19)

Transforming the surface integral into the volume integral in (2.19) we finally have:

$$\varphi'(\mathbf{r}_1) = \int \mathbf{T}(\mathbf{r}_2) \cdot \nabla^{(2)} G(\mathbf{r}_2, \mathbf{r}_1) \, \mathrm{d} V(\mathbf{r}_2) ;$$

$$\nabla^{(a)} \equiv \frac{\partial}{\partial \mathbf{r}_a} , \qquad a = 1, 2 . \qquad (2.20)$$

According to (2.16) and (2.20) the perturbed field \mathbf{E}' is:

$$\mathbf{E}'(\mathbf{r}_1) = -\int \mathbf{T}(\mathbf{r}_2) \cdot \nabla^{(2)} \nabla^{(1)} G(\mathbf{r}_2, \mathbf{r}_1) \, \mathrm{d}V(\mathbf{r}_2) \,. \tag{2.21}$$

Following [21, 27, 34, 35] we introduce the tensor integral operator \hat{Q} , the kernel $Q(\mathbf{r}_1, \mathbf{r}_2)$ of which is related to the Green's function $G(\mathbf{r}_1, \mathbf{r}_2)$ by the equation:

$$Q(\mathbf{r}_1, \mathbf{r}_2) = -\nabla^{(1)} \otimes \nabla^{(2)} G(\mathbf{r}_1, \mathbf{r}_2), \qquad (2.22)$$

or by components:

$$Q_{ij}(\mathbf{r}_1, \mathbf{r}_2) = -\nabla_i^{(1)} \nabla_j^{(2)} G(\mathbf{r}_1, \mathbf{r}_2) \,. \tag{2.23}$$

Then solution (2.21) can be rewritten in the form [27, 34, 35]

$$\mathbf{E} = \mathbf{E}^{c} + \widehat{Q}\mathbf{T} = \mathbf{E}^{c} + \widehat{X}\mathbf{E}, \qquad \widehat{X} \equiv \widehat{Q}\sigma', \qquad \mathbf{T} = \sigma'\mathbf{E}.$$
(2.24)

Thus, the problem of determination of the field \mathbf{E} from the differential equation (2.8) with the boundary conditions (2.9) is reduced to solution of a nonuniform integral equation of second kind [93]. The solution of this equation yields the relation between the unknown field \mathbf{E} and the known field \mathbf{E}^{c} .

It is easy to see that in view of (2.1) and (2.10) \hat{Q} complies with the equations:

$$\nabla \times \widehat{Q} = \widehat{O}, \quad \nabla \cdot \widehat{P} = \widehat{O}, \tag{2.25}$$

$$-\widehat{P} \equiv \sigma^{c}\widehat{I} + \sigma^{c}\widehat{Q}\sigma^{c}, \quad -\widehat{Q} = \rho^{c}\widehat{I} + \rho^{c}\widehat{P}\rho^{c}; \quad \sigma^{c}\rho^{c} = I$$
(2.26)

where $\widehat{I}(I)$ is a unit operator (tensor) of rank 2.

Expressing \widehat{Q} via \widehat{P} we arrive after simple manipulations from (2.24) at the equation [27, 35]:

$$\mathbf{J} = \mathbf{J}^{c} + \widehat{P}\mathbf{H} = \mathbf{J}^{c} + \widehat{Y}\mathbf{J}, \qquad \widehat{Y} \equiv \widehat{P}\rho',$$
$$\mathbf{H} = \rho'\mathbf{J} = -\rho^{c}\mathbf{T}.$$
(2.27)

The equation has the same form as (2.24) but it makes possible to express the unknown field **J** through the known field **J**^c. Here **H** is the 'polarised' intensity vector similar to the vector **T**. Equations (2.24) and (2.27) are equivalent and they will be used on equal terms.

Besides (2.24) and (2.27) we shall need the equations [37]

$$\mathbf{E} = \langle \mathbf{E} \rangle + \widehat{R}\widehat{X}\mathbf{E}, \qquad \mathbf{J} = \langle \mathbf{J} \rangle + \widehat{R}\widehat{Y}\mathbf{J}, \qquad (2.28)$$

where the isolation operator \widehat{R} of a random component of a field *F* is specified by the relation [27]:

$$\left(\widehat{R}F\right)^{n+1} \equiv F(\widehat{R}F)^n - \left\langle F(\widehat{R}F)^n \right\rangle, \quad n \ge 0.$$
(2.29)

Unlike (2.24) and (2.27) Eqns (2.28) do not include the fields \mathbf{E}^{c} and \mathbf{J}^{c} , respectively. This makes the calculation of ECT simpler.

Transition from (2.24) and (2.27) to (2.28) is necessary to solve the Dirichlet and Neumann problems when the equations [37]

$$\overline{Q}\,\tilde{\mathbf{f}}_{1} = 0\,, \qquad \left\{\mathbf{e}^{c} - \langle \mathbf{e} \rangle\right\} = 0\,, \qquad \tilde{\mathbf{f}}_{1} \in \,\vec{\mathcal{H}}\,, \tag{2.30a}$$

$$\overline{P}\,\tilde{\mathbf{f}}_2 = 0\,, \qquad \left\{\mathbf{j}^{\mathrm{c}} - \langle \mathbf{j} \rangle\right\} = 0\,, \qquad \tilde{\mathbf{f}}_2 \in \,\vec{\mathcal{H}} \tag{2.30b}$$

are valid. Here we used the notation of (3.2) and (3.25) and \mathbf{f}_a are constant vectors. For a homogeneous comparison medium and boundary conditions (1.4) and (1.5) from (2.30) it follows that

$$\mathbf{e}^{c} = \langle \mathbf{e} \rangle, \quad \overline{\widehat{Q}} \mathbf{f}_{1} = \overline{\widehat{Q}} \widehat{R} \mathbf{f}_{1}, \quad \mathbf{f}_{1} \in \overrightarrow{\mathcal{H}},$$
 (2.31a)

$$\mathbf{j}^{c} = \langle \mathbf{j} \rangle, \qquad \widehat{\overline{P}} \mathbf{f}_{2} = \widehat{\overline{P}} \widehat{R} \mathbf{f}_{2}, \qquad \mathbf{f}_{2} \in \vec{\mathcal{H}}, \qquad (2.31b)$$

in the first case and the second case, respectively. Here $\langle {\bf f}_a \rangle = {f {f f}}_a.$

3. Projective operators

Here we consider symmetric (Hermitian) operators, i.e., those, for which the equation $\hat{S}^{(+)} = \hat{S}$ is satisfied [93]. The index (+) represents the conjugation operation:

$$(\mathbf{E}_1, \widehat{S} \mathbf{E}_2) \equiv (\widehat{S}^{(+)} \mathbf{E}_1, \mathbf{E}_2).$$
(3.1)

Here we define the inner product $(\mathbf{E}_1, \mathbf{E}_2)$ of two vector functions \mathbf{E}_1 and \mathbf{E}_2 in a Hilbert space $\hat{\mathcal{H}}$ as

$$(\mathbf{E}_1, \mathbf{E}_2) \equiv \int \langle \mathbf{E}_1 \cdot \mathbf{E}_2 \rangle \, \mathrm{d}v \equiv \left\{ \langle \mathbf{E}_1 \cdot \mathbf{E}_2 \rangle \right\}, \quad \mathrm{d}v \equiv \frac{\mathrm{d}V}{V}, (3.2)$$

where braces mean averaging over volume.

The natural question arises how the statistical average (1.2) is related to the volume one (3.2). Usually the ergodic hypothesis (or theorem) is applied [8, 14, 20, 30, 31, 37, 94]. The field is said to be ergodic (or self-averaging [20, 95-98]) if there exists the limit

$$\lim_{V \to \infty} \left\langle \left\{ \xi'' \right\}^2 \right\rangle = 0, \quad \left\{ \xi'' \right\} = \left\{ \xi \right\}''; \quad f'' \equiv f - \left\langle f \right\rangle, \ (3.3)$$

The limit is denoted by

$$\lim_{V \to \infty} \{\xi\} = \left\langle \{\xi\} \right\rangle \tag{3.4}$$

and presents the convergence in mean square. Instead of (3.4) we shall write [8]

$$\{\xi\} = \left\langle \{\xi\} \right\rangle = \left\{ \langle\xi\rangle \right\},\tag{3.5}$$

where the equality is understood to be the limit (3.4). In the case of a statistically uniform field the definition of (3.4) becomes simpler:

$$\lim_{V \to \infty} \{\xi\} = \{\langle\xi\rangle\} = \langle\xi\rangle = \text{const}, \qquad (3.6)$$

and the statistical average is full. Similarly, instead of (3.6) we shall write

$$\{\xi\} = \langle\xi\rangle \,. \tag{3.7}$$

The equivalence of two ways of averaging makes it easier to extract the statistical information because the full information is contained in a single realisation of the statistical field $\xi(\mathbf{r})$ and it can be extracted from a single sample. Frequently, the limit $V \rightarrow \infty$ in (3.6) is unnecessary and equality (3.7) is valid with a good accuracy for a finite V.

In the case of real-valued field conjugation in (3.1) is reduced to transposition. The operator \hat{Q} introduced in (2.22) and (2.23) can be presented in view of (3.1) and (3.2) in the form [35]:

$$\widehat{Q} = -\nabla \widehat{G} \otimes \nabla^{(+)} \,, \tag{3.8}$$

where \hat{G} is an integral operator, the kernel of which — the Green's function $G(\mathbf{r}_1, \mathbf{r}_2)$ (2.10), (2.11) — satisfies, in the strength of the reciprocity theorem, the equation [30, 93]

$$G(\mathbf{r}_1, \mathbf{r}_2) = G(\mathbf{r}_2, \mathbf{r}_1), \qquad (3.9)$$

equivalent to the symmetry condition for the operator \widehat{G} : $\widehat{G}^{(+)} = \widehat{G}$.

It is easy to show that \widehat{Q} is a symmetric operator. In fact, applying the operator (+) to both sides of (3.8) and considering the equations $(\widehat{S}^{(+)})^{(+)} = \widehat{S}$, $(\widehat{S}_1 \widehat{S}_2)^{(+)} = \widehat{S}_2^{(+)} \widehat{S}_1^{(+)}$, and $\widehat{G}^{(+)} = \widehat{G}$ we can write:

$$\widehat{Q}^{(+)} = -(\nabla \widehat{G} \otimes \nabla^{(+)})^{(+)} = -(\nabla^{(+)})^{(+)} \widehat{G}^{(+)} \otimes \nabla^{(+)} = \widehat{Q}.$$
(3.10)

Similarly, it follows from (2.26) that

$$\widehat{P}^{(+)} = \widehat{P}.$$

Let us consider the equation

$$\int \mathbf{E}' \cdot \mathbf{J}' \, \mathrm{d}V = \int \varphi' \nabla \cdot \mathbf{J}' \, \mathrm{d}V - \oint \, \mathrm{d}\mathbf{S} \cdot \mathbf{J}' \varphi' \,, \qquad (3.11)$$

in which the surface integral vanishes by the boundary conditions (2.15). In turn the volume integral on the right-hand side in (3.11) is zero by virtue of the equation

$$\nabla \cdot \mathbf{J}' = 0; \quad \mathbf{J}' = \sigma^{c} \mathbf{E}' + \mathbf{T}$$
(3.12)

derived from (2.17). In terms of inner products the equation (3.11) has the form:

$$(\mathbf{E}', \mathbf{J}') = 0. \tag{3.13}$$

Considering Eqns (2.24) and (2.27) and the fact that \widehat{P} and \widehat{Q} are Hermitian, we have

$$(\widehat{Q}\mathbf{T},\widehat{P}\mathbf{H}) = (\mathbf{T},\widehat{Q}^{(+)}\widehat{P}\mathbf{H}) = (\mathbf{T},\widehat{Q}\widehat{P}\mathbf{H}) = (\mathbf{H},\widehat{P}\widehat{Q}\mathbf{T})(3.14)$$

Equalities (3.14) enable us to introduce important relations for the operators \hat{P} and \hat{Q} . Substituting the operator \hat{P} defined by (2.26) into (3.14) and expressing the field **H** through **T** by means of (2.27) we find

$$(\mathbf{T}, \widehat{Q}\mathbf{T}) + (\mathbf{T}, \widehat{Q}\sigma^{c}\widehat{Q}\mathbf{T}) = 0.$$
(3.15)

The equivalent operator equation is [34, 35]:

$$\widehat{Q} + \widehat{Q}\sigma^{c}\widehat{Q} = \widehat{O}.$$
(3.16)

Equation (3.16) implies that \widehat{Q} is negative in the sense of the inequality $(\mathbf{T}, \widehat{Q}\mathbf{T}) \leq 0$ because the integral (quadratic) form $(\mathbf{E}', \sigma^c \mathbf{E}')$ is positive [99]. Similarly, (3.14) leads to the equation:

$$\widehat{P} + \widehat{P}\rho^{\,c}\widehat{P} = \widehat{O}\,.\tag{3.17}$$

Equation (3.17) implies that \widehat{P} is negative, i.e., $(\mathbf{H}, \widehat{P}\mathbf{H}) \leq 0$, because the integral form $(\mathbf{J}', \rho^c \mathbf{J}')$ is positive.

In the above consideration we used two fields **E** and $\mathbf{J} = \sigma \mathbf{E}$. They belong to the space $\vec{\mathcal{H}}$ but differ by the dimension factor σ . It is, however, desirable to get away with the factor and use fields of the same dimension. This can be attained by multiplying **E** and **J** by the symmetric positive tensors $\sqrt{\sigma^c}$ and $\sqrt{\rho^c}$, respectively. The representation $\sigma^c = (\sqrt{\sigma^c})^2$ is unique because σ^c is positive and symmetric [99].

Now we shall introduce notation for the aforementioned fields and operators:

$$\mathbf{j} \equiv \sqrt{\rho^{c}} \mathbf{J}, \quad \mathbf{e} \equiv \sqrt{\sigma^{c}} \mathbf{E}; \quad \sqrt{\sigma^{c}} \sqrt{\rho^{c}} = I, \qquad (3.18)$$
$$\widehat{\overline{P}} \equiv -\sqrt{\rho^{c}} \widehat{\overline{P}} \sqrt{\rho^{c}}, \quad \widehat{\overline{P}}^{(+)} = \widehat{\overline{P}};$$
$$\widehat{\overline{Q}} \equiv -\sqrt{\sigma^{c}} \widehat{\overline{Q}} \sqrt{\sigma^{c}}, \quad \widehat{\overline{Q}}^{(+)} = \widehat{\overline{Q}}. \qquad (3.19)$$

In view of (3.18) and (3.19) Eqns (2.24), (2.27) and (2.28) take the form:

$$\mathbf{e} = \mathbf{e}^{c} - \widehat{\overline{Q}}\overline{\sigma}'\mathbf{e}, \qquad \sigma' \equiv \sqrt{\sigma^{c}}\overline{\sigma}'\sqrt{\sigma^{c}}, \qquad \mathbf{e}^{c} \equiv \sqrt{\sigma^{c}}\mathbf{E}^{c},$$
(3.20)

$$\mathbf{j} = \mathbf{j}^{c} - \widehat{\overline{P}} \overline{\rho}' \mathbf{j}, \qquad \rho' \equiv \sqrt{\rho^{c}} \overline{\rho}' \sqrt{\rho^{c}}, \qquad \mathbf{j}^{c} = \mathbf{e}^{c}, \quad (3.21)$$

$$\mathbf{e} = \langle \mathbf{e} \rangle - \widehat{R}\overline{\widehat{Q}}\overline{\sigma}'\mathbf{e}, \qquad \mathbf{j} = \langle \mathbf{j} \rangle - \widehat{R}\overline{\widehat{P}}\rho'\mathbf{j}, \qquad (3.22)$$

the two operator equations in (2.26) are reduced to one:

$$\widehat{\overline{P}} + \widehat{\overline{Q}} = \widehat{I}, \qquad (3.23)$$

and, instead of (3.16) and (3.17) we have [34, 35]:

$$\hat{\overline{Q}}^2 = \hat{\overline{Q}}, \quad \hat{\overline{P}}^2 = \hat{\overline{P}}.$$
 (3.24)

On the strength of (3.23) and (3.24) the positive symmetric operators $\overline{\overline{Q}}$ and $\overline{\overline{P}}$ possess all the properties of projective operators (projectors) [99]. Thus, the space $\overline{\mathcal{H}}$ can be presented as the sum

$$\vec{\mathcal{H}} = \vec{\mathcal{H}}_1 \oplus \vec{\mathcal{H}}_2 \tag{3.25}$$

of two orthogonal subspaces $\vec{\mathcal{H}}_1$ and $\vec{\mathcal{H}}_2$, elements of which are the vectors $\overline{\overline{Q}} \mathbf{f}$ and $\overline{\overline{P}} \mathbf{f}$, respectively. The projectors $\overline{\overline{Q}}$ and $\overline{\overline{P}}$ comply with the inequalities [99]:

$$\widehat{O} \leqslant \overline{\widehat{Q}} \leqslant \widehat{I}, \quad \widehat{O} \leqslant \overline{\widehat{P}} \leqslant \widehat{I}.$$
 (3.26)

The left-hand values are reached when $\vec{\mathcal{H}}_1$ and $\vec{\mathcal{H}}_2$ consist of a single zero element and the right-hand values are reached when $\vec{\mathcal{H}}_1$ or $\vec{\mathcal{H}}_2$ is the same as $\vec{\mathcal{H}}$.

Similar to (2.24), (2.27), and (2.28) we shall rewrite (3.20), (3.21), and (3.22) in the form

$$\mathbf{e} = \mathbf{e}^{c} + \widehat{X}\mathbf{e}, \quad \widehat{\overline{X}} \equiv -\widehat{\overline{Q}}\overline{\sigma}', \quad \overline{\sigma}' = \overline{\sigma} - I, \quad (3.27)$$

$$\mathbf{j} = \mathbf{j}^{c} + \overline{Y}\mathbf{j}, \quad \overline{Y} \equiv -\overline{P}\overline{\rho}', \quad \overline{\rho}' = \overline{\rho} - I,$$
 (3.28)

$$\mathbf{e} = \langle \mathbf{e} \rangle + \widehat{R} \,\overline{\widehat{X}} \mathbf{e} \,, \qquad \mathbf{j} = \langle \mathbf{j} \rangle + \widehat{R} \,\overline{\widehat{Y}} \mathbf{j} \,. \tag{3.29}$$

According to (3.23) the reduced fields e and j can be decomposed into sums of mutually orthogonal components belonging to the subspaces $\vec{\mathcal{H}}_1$ and $\vec{\mathcal{H}}_2$. It turns out that the fields e' and j' belong exclusively to $\vec{\mathcal{H}}_1$ and $\vec{\mathcal{H}}_2$, respectively. This fact enables us to write instead of (3.27) and (3.28):

$$\mathbf{e}' = \overline{\overline{X}}\mathbf{e} = \ \vec{\mathcal{E}}_1 + \overline{\overline{X}}\mathbf{e}', \qquad \overline{\overline{X}}^k \mathbf{e}^c \equiv \ \vec{\mathcal{E}}_k \in \vec{\mathcal{H}}_1, \quad k \ge 1, (3.30)$$

$$\mathbf{j}' = \widehat{\overline{Y}}\mathbf{j} = \vec{\mathcal{J}}_1 + \widehat{\overline{Y}}\mathbf{j}', \qquad \widehat{\overline{Y}}^k \mathbf{j}^c \equiv \vec{\mathcal{J}}_k \in \vec{\mathcal{H}}_2, \quad k \ge 1, (3.31)$$

where the operator \overline{X} maps the subspace \mathcal{H}_1 onto itself and the operator \widehat{Y} does it with the subspace \mathcal{H}_2 . Thus, in terms of Refs [99, 100] Eqns (3.30) and (3.31) can be treated as functional equations in subspaces \mathcal{H}_1 and \mathcal{H}_2 , respectively.

In conclusion of this section we consider properties of the operator \widehat{R} (2.29). Along with \widehat{R} we introduce the statistical averaging operator \widehat{M} which is related to \widehat{R} by the equation:

$$\widehat{R} + \widehat{M} = \widehat{I}, \qquad \widehat{M}F \equiv \langle F \rangle.$$
 (3.32)

It is easy to see that the operators \widehat{R} and \widehat{M} obey the relations:

$$\widehat{R}^2 = \widehat{R}, \quad \widehat{R}^{(+)} = \widehat{R}; \quad \widehat{M}^2 = \widehat{M}, \quad \widehat{M}^{(+)} = \widehat{M};$$

$$\widehat{R}\widehat{M} = \widehat{M}\widehat{R} = \widehat{O}, \qquad (3.33)$$

$$\widehat{O} \leqslant \widehat{R} \leqslant \widehat{I}, \qquad \widehat{O} \leqslant \widehat{M} \leqslant \widehat{I},$$

$$(3.34)$$

which are similar to (3.24) and (3.26). Therefore, \hat{R} and \hat{M} are projective operators. Thus, the space $\hat{\mathcal{H}}$ can be represented as a sum

$$\vec{\mathcal{H}} = \vec{\mathcal{H}}^R \oplus \vec{\mathcal{H}}^M, \qquad (3.35)$$

where

$$\vec{\mathcal{H}}^R = \vec{\mathcal{H}}_1^R \oplus \vec{\mathcal{H}}_2^R, \qquad \vec{\mathcal{H}}^M = \vec{\mathcal{H}}_1^M \oplus \vec{\mathcal{H}}_2^M, \qquad (3.26)$$

of two orthogonal subspaces $\vec{\mathcal{H}}^R$ and $\vec{\mathcal{H}}^M$, elements of which are, respectively, random $\hat{R}\mathbf{f}$ ($\langle \hat{R}\mathbf{f} \rangle = 0$) and regular $\hat{M}\mathbf{f}$ vectors. Each of the subspaces $\vec{\mathcal{H}}_1$ and $\vec{\mathcal{H}}_2$ can also be presented in the form of (3.35):

$$\vec{\mathcal{H}}_a = \vec{\mathcal{H}}_a^R \oplus \vec{\mathcal{H}}_a^M; \quad a = 1, 2.$$
(3.37)

It follows from Eqns (3.29) that

$$\mathbf{e}'' = \widehat{R}\overline{\widehat{X}}\mathbf{e} = \mathbf{e}_1 + \widehat{R}\overline{\widehat{X}}\mathbf{e}'', \qquad (\widehat{R}\overline{\widehat{X}})^k \langle \mathbf{e} \rangle \equiv \mathbf{e}_k \in \vec{\mathcal{H}}_1^R, \quad k \ge 1,$$
(3.38)

$$\mathbf{j}'' = \widehat{R}\widehat{Y}\mathbf{j} = \mathbf{j}_1 + \widehat{R}\widehat{Y}\mathbf{j}'', \qquad (\widehat{R}\widehat{Y})^k \langle \mathbf{j} \rangle \equiv \mathbf{j}_k \in \mathcal{H}_2^R, \qquad k \ge 1$$
(3.39)

Without further comments we write similar relations for fields of the type of $\hat{M}\mathbf{f}$. Applying the operator \hat{M} to both sides of Eqns (3.27) and (3.28) we have:

$$\langle \mathbf{e}' \rangle = -\overline{\widehat{Q}}(\widehat{\overline{\sigma}}^* - \widehat{I})\mathbf{e}^{\mathsf{c}} - \overline{\widehat{Q}}(\widehat{\overline{\sigma}}^* - \widehat{I})\langle \mathbf{e}' \rangle, \quad \langle \mathbf{e}' \rangle \in \vec{\mathcal{H}}_1^M, (3.40)$$

$$\langle \mathbf{j}' \rangle = -\overline{P}(\hat{\rho}^* - \widehat{I})\mathbf{j}^{c} - \overline{P}(\hat{\rho}^* - \widehat{I})\langle \mathbf{j}' \rangle, \qquad \langle \mathbf{j}' \rangle \in \mathcal{H}_2^M. (3.41)$$

In the further investigation the leading part belong to fields (3.30), (3.31), (3.38), and (3.39).

4. Convergence conditions for perturbation method in theory of inhomogeneous media

The functional equations (3.38) in \mathcal{H}_1 and (3.39) in \mathcal{H}_2 or (3.29) in \mathcal{H} are solved when the operators \widehat{A} and \widehat{B} of the forms [37, 99]

$$\widehat{A} = (\widehat{I} - \widehat{R}\,\widehat{\overline{X}})^{-1}\,,\qquad \widehat{B} = (\widehat{I} - \widehat{R}\,\widehat{\overline{Y}})^{-1} \tag{4.1}$$

are found. Given the convergence condition, these operators can be presented as Neumann series:

$$\widehat{A} = \sum_{k=0}^{\infty} (\widehat{R}\,\widehat{\overline{X}})^k \,, \qquad \widehat{B} = \sum_{k=0}^{\infty} (\widehat{R}\,\widehat{\overline{Y}})^k \,. \tag{4.2}$$

In this case the fields e and j are given, with regard for (3.38) and (3.39), by the expansions:

$$\mathbf{e} = \widehat{A} \langle \mathbf{e} \rangle = \sum_{k=0}^{\infty} \mathbf{e}_k , \quad \mathbf{e}'' = \widehat{A} \mathbf{e}_1 = \sum_{k=1}^{\infty} \mathbf{e}_k , \quad \mathbf{e}_0 \equiv \langle \mathbf{e} \rangle , (4.3)$$
$$\mathbf{i} = \widehat{B} \langle \mathbf{i} \rangle = \sum_{k=0}^{\infty} \mathbf{i}_k , \quad \mathbf{i}'' = \widehat{B} \mathbf{i}_1 = \sum_{k=1}^{\infty} \mathbf{i}_k , \quad \mathbf{i}_0 \equiv \langle \mathbf{i} \rangle , (4.4)$$

called perturbation series (iterative series).

Below we examine a uniform (in norm) convergence for series (4.2)-(4.4). We shall say [99] that a sequence

$$\widehat{A}_{(n)} \equiv \sum_{k=0}^{n} (\widehat{R}\overline{\overline{X}})^{k}$$

 $\overline{k=0}$

is convergent to \widehat{A} in norm if $\|\widehat{A}_{(n)} - \widehat{A}\| \to 0$ as $n \to \infty$. As is known, the notion of norm is directly related to that of inner product (3.2). By definition we have [93, 99, 100]

$$\|\widehat{S}\| = \sup_{\mathbf{f}\neq 0} \frac{\|S\mathbf{f}\|}{\|\mathbf{f}\|}, \quad \|\mathbf{f}\|^2 \equiv (\mathbf{f}, \mathbf{f}), \quad \mathbf{f} \in \vec{\mathcal{H}}.$$
(4.5)

According to the Banach theorem [99, 100] the operator $(\widehat{I} - \widehat{RX})$ has a continuous inverse operator \widehat{A} of the form of

 $\left\|\widehat{R}\widehat{\overline{X}}\right\| \leqslant k_1 < 1.$

holds for the norm of the operator \widehat{RX} . The necessary and sufficient convergence conditions for the series \widehat{A} from (4.2) is that the inequality

 $\left\| (\widehat{R}\widehat{\overline{X}})^n \right\| \leqslant k_1 < 1 \,.$

holds for an $n \ge 1$ [100]. Since the projectors have a unit norm [99], the sufficient convergence condition (SCC) for the series \widehat{A} from (4.2) can be written in the form:

$$\|\widehat{R}\widehat{\widehat{X}}\| \leq \|\widehat{R}\| \|\widehat{\widehat{X}}\| = \|\widehat{\widehat{X}}\| \leq \|\widehat{\widehat{Q}}\| \|\overline{\sigma}'\| = \|\overline{\sigma}'\| \leq k_1 < 1.$$
(4.6)

The SCC for the series \widehat{B} from (4.2) is

$$\|\widehat{R}\,\widehat{\overline{Y}}\| \leq \|\widehat{R}\,\|\|\widehat{\overline{Y}}\| = \|\widehat{\overline{Y}}\| \leq \|\widehat{\overline{P}}\,\|\|\bar{\rho}\,'\| = \|\bar{\rho}\,'\| \leq k_2 < 1.$$

$$(4.7)$$

By rewriting inequalities (4.6) and (4.7) in operator (tensor) form we have [34, 37]

$$0 \leq |\bar{\sigma}'| \leq k_1 I, \quad -k_1 I \leq \bar{\sigma}' \leq k_1 I, \quad 0 \leq k_1 < 1,$$
(4.8)

$$0 \leq |\bar{\rho}'| \leq k_2 I, \quad -k_2 I \leq \bar{\rho}' \leq k_2 I, \quad 0 \leq k_2 < 1,$$
(4.9)

where the absolute value of the operator (tensor) \hat{s} satisfies [44] the relations $|\hat{s}| \equiv \sqrt{\hat{s}^2} \ge \pm \hat{s}$.

In the initial notation the SCC of (4.8) and (4.9) for the series from (4.2) have the form:

$$-k_1 \sigma^{\mathsf{c}} \leqslant \sigma' \leqslant k_1 \sigma^{\mathsf{c}}, \qquad (1-k_1) \sigma^{\mathsf{c}} \leqslant \sigma \leqslant (1+k_1) \sigma^{\mathsf{c}},$$

$$(4.10a)$$

$$-k_2 \rho^{\mathsf{c}} \leqslant \rho' \leqslant k_2 \rho^{\mathsf{c}}, \qquad (1-k_2) \rho^{\mathsf{c}} \leqslant \rho \leqslant (1+k_2) \rho^{\mathsf{c}}.$$

$$(4.10b)$$

It is easy to see that by virtue of the equations

$$\bar{\sigma}' = \bar{\sigma} - I = \sqrt{\bar{\sigma}} (I - \bar{\rho}) \sqrt{\bar{\sigma}} = -\sqrt{\bar{\sigma}} \bar{\rho}' \sqrt{\bar{\sigma}} ,$$

$$\sigma \rho = \sigma^{c} \rho^{c} = I$$
(4.11)

the relations are valid

$$\bar{\sigma}' \gtrless 0 \Leftrightarrow \bar{\rho}' \lessgtr 0. \tag{4.12}$$

for the same reference medium $(\sigma^c \rho^c = I)$ and definite (negative or positive) $\bar{\sigma}'$ and $\bar{\rho}'$.

It is safe to say that (4.8) and (4.9) are also SCC for the series:

$$\hat{a} = (\widehat{I} - \overline{\widehat{X}})^{-1} = \sum_{k=0}^{\infty} \widehat{\overline{X}}^k, \quad \hat{b} = (\widehat{I} - \overline{\widehat{Y}})^{-1} = \sum_{k=0}^{\infty} \widehat{\overline{Y}}^k, \quad (4.13)$$

which appear in the course of solution of the functional equations (3.27) and (3.28) or (3.30) and (3.31). Note that series (4.13) are convergent slower than series (4.2) by virtue of the inequalities

$$\|\widehat{R}\widehat{\overline{X}}\| \le \|\widehat{\overline{X}}\|, \quad \|\widehat{R}\widehat{\overline{Y}}\| \le \|\widehat{\overline{Y}}\|$$

$$(4.14)$$

which follow from (4.6) and (4.7).

Operators (4.1) and (4.2) are related (mutually) by the equations [27]

$$\widehat{A} = \widehat{a} \langle \widehat{a} \rangle^{-1}, \qquad \widehat{B} = \widehat{b} \langle \widehat{b} \rangle^{-1}.$$
(4.15)

Thanks to them we can pass from one expansion to the other.

Since \hat{A} and \hat{B} are independent of the parameters σ^{c} and ρ^{c} [34], the choice of them is governed solely by inequalities (4.10). To determine the interval, in which the values of σ^{c} and ρ^{c} lie, such that the convergence conditions for series (4.2) are satisfied, we rewrite (4.10) in the form:

$$\sigma(1+k_1)^{-1} \leq \sigma^c \leq \sigma(1-k_1)^{-1}, \quad 0 \leq k_1 < 1,$$
 (4.16a)

$$\rho(1+k_2)^{-1} \le \rho^{c} \le \rho(1-k_2)^{-1}, \quad 0 \le k_2 < 1.$$
(4.16b)

Obviously, only the left-hand sides of the inequalities (4.16) impose severe restrictions on the values of σ^{c} and ρ^{c} .

In many papers on the subject the question of convergence of series the authors use is not considered explicitly. Usually, it is postulated that the required small parameter exists [4, 8, 11, 12, 14, 32, 33]. However, quantitative estimates for this parameter are based, as a rule, on intuitive suggestions. As a result, the restrictions imposed on the small parameter are either extremely severe or insufficient for the series to be convergent. The first group is presented by the inequalities [4, 8, 14, 33]:

$$\sup_{\mathbf{r}} \left| \bar{\sigma}'(\mathbf{r}) \right| \ll 1; \quad \left| \left\langle \left(\bar{\sigma}_0'' \right)^n \right\rangle \right| \ll 1, \quad n \ge 2.$$
(4.17)

The inequality from Ref. [32] (see notation in (6.17))

$$\left\langle \bar{\sigma}_{0}^{\prime\prime} \,\overline{Q} \bar{\sigma}_{0}^{\prime\prime} \right\rangle < I; \quad \left\langle \bar{\sigma}_{0} \right\rangle \equiv I \Rightarrow \sigma^{c} = \left\langle \sigma \right\rangle$$

$$(4.18)$$

is, as is shown in Ref. [36], the simplest and it is weaker than the others in the infinite chain of inequalities representing the convergence conditions for the series \hat{A} from (4.2) and \hat{a} from (4.13).

If the SCC is satisfied in one of the forms (4.6), (4.8), (4.10a), and (4.16a) for the series \widehat{A} or in one of the forms (4.7), (4.9), (4.10b), and (4.16b) for the series \widehat{B} , then \widehat{A} and \widehat{B} can be presented in forms (4.1) and (4.15) and expanded in Neumann series (4.2).

Figure 2a shows the plots of the functions $\xi = \xi(x)$ (where $\xi \equiv \sigma^c/\sigma_2$, $x \equiv \sigma_1/\sigma_2$) which specify the characteristic regions of the parameter σ^c in the case of a mixture of two isotropic components. The symbolic presentations of the functions used in constructions ($\xi = 1/2$, $\xi = 1$, $\xi = 2$, $\xi = x/2$, $\xi = x$, $\xi = 2x$, $\xi = |1 - x|/2$, $\xi = 2x/|1 - x|$) are given around the perimeter of the square.

The values of ξ above the bold solid line form a set of values of ξ for which the SCC in the form of (4.16a) is satisfied. The values of ξ below the bold dashed line form a set of values of ξ for which the SCC in the form of (4.16b) is satisfied. The two series (5.3) and (5.4) are convergent when the value of ξ belongs to both above sets. The region with slanting cross-hatching (or the prohibited region) contains the values of ξ for which the SCC is not satisfied neither in the form of (4.16a) nor in the form of (4.16b).

The values of ξ above the thin solid line form a set of values of ξ for which the series from (5.3) is of constant signs while the series from (5.4) is of alternating signs. The values of ξ below the thin broken line form a set of values of ξ for which the series from (5.4) is of constant signs while the series from



Figure 2.

(5.3) is of alternating signs. The series from (5.3) is convergent if it is of alternating signs and the value of ξ belongs to the region below the thin broken line and above the bold solid line. The last region is cross-hatched horizontally. Similarly, the region is defined, in which the series from (5.4) is convergent, of constant sign, or of alternating sign. The series from (5.4) is convergent if it is of alternating signs and the value of ξ belongs to the vertically cross-hatched region.

Along with SCC (4.8) and (4.9) we shall consider weaker restrictions [see (6.17)]

$$\left\langle \bar{\sigma}^{\,\prime\prime} \, \overline{Q} \, \bar{\sigma}^{\,\prime\prime} \right\rangle \leqslant I, \qquad \left\langle \bar{\rho}^{\,\prime\prime} \, \overline{P} \, \bar{\rho}^{\,\prime\prime} \right\rangle \leqslant I.$$

$$(4.19)$$

In the case of a mixture of two isotropic components they take the form:

$$\xi \ge c|1-x|, \quad c^2 \equiv \overline{Q}v_1v_2, \quad v_1+v_2 = 1, \quad (4.20a)$$

$$\xi \leqslant \frac{x/d}{|1-x|}, \quad d^2 \equiv \overline{P}v_1v_2, \quad \overline{P} + \overline{Q} = 1.$$
 (4.20b)

The values of ξ above the dash-dot line in Fig. 2a form a set of values of ξ , for which the criterion (4.20a) is satisfied provided that $\overline{Q} = 1$ and $v_1 = 1/2$, when $c = c_{\text{max}} = 1/2$ and the area S_1 of the part of the square between the bold solid line and the

dash-dot line is at maximum. The values of ξ below the dotted line form a set of values of ξ , for which the criterion (4.20b) is satisfied provided that $\overline{P} = 1$ and $v_1 = 1/2$ when $d = d_{\text{max}} = 1/2$ and the area S_2 of the part of the square between the bold dashed and dotted line is at minimum. The values of ξ from the set S_1 satisfy the criterion (4.20a) and do not satisfy the SCC (4.16a). Therefore, these values cannot be used to construct a convergent series (5.3). Similarly, the values of ξ from the region S_2 violate the SCC (4.16b) and, hence, they cannot be used to construct a convergent series (5.4).

In Figure 2b the bold lines bound the region, in which series from (5.3) and (5.4) are of fixed signs. The thin lines represent the values $\xi = \langle \bar{\sigma} \rangle$ (solid line) and $\xi = \langle \bar{\rho} \rangle^{-1}$ (dashed line) for $v_2 = 1/3$. The dash-dot line and the dotted line have the same sense as in Fig. 2a. but when $v_2 = \overline{Q} = 1/3$ and $v_1 = \overline{P} = 2/3$. The values $\xi = \langle \bar{\sigma} \rangle$ from the left-hand triangle and $\xi = \langle \bar{\rho} \rangle^{-1}$ from the right-hand triangle in the prohibited region cannot be used to construct convergent series (5.3) and (5.4).

It is seen from Fig. 2b that the values of $\sigma^{c} = \langle \sigma \rangle$ (the line $\xi = \langle \bar{\sigma} \rangle$) satisfy the convergence criterion (4.18) from Ref. [32] everywhere. However, a part of this line crosses the left-hand triangle of the prohibited region and this means that the SCC (4.16a) is not satisfied there. The same refers to the case $\xi = \langle \bar{\rho} \rangle^{-1}$.

The choice $\sigma^c = \langle \sigma \rangle$ (or $\rho^c = \langle \rho \rangle$) is natural but in no way related to the SCC. The magnitude of σ^c itself has the sense of zero fluctuation level which is arbitrary to a certain degree. Rigorous [i.e. consistent with the SCC of series (5.3)] choice of σ^c allows one to obtain the convergent series for any fluctuations of the field $\sigma(\mathbf{r})$. The question of the choice of ρ^c — the zero fluctuation level for the field $\rho(\mathbf{r})$ dual to the field $\sigma(\mathbf{r})$ — is tackled in a similar way.

Existing methods to compute $\hat{\sigma}^*$ (ECT), both model and strictly analytical, reduce as a rule the computation of $\hat{\sigma}^*$ to the expansion of the ECT in corresponding small parameter whose existence, however, does not ensure the convergence of the series investigated. For that, conditions obtained by a correct procedure [for example, the SCC in the form (4.8) and (4.9)] are needed. It is not always that a natural choice of statistically mean value of the material characteristics (σ, ρ) for zero fluctuation level is the best one. Moreover, as shown above (see also Fig. 2b), such a choice of σ^{c} and ρ^{c} may lead to divergence of series (5.3) and (5.4). Since the curves $\xi = \langle \bar{\sigma} \rangle$ and $\xi = \langle \bar{\rho} \rangle^{-1}$ lie in the region where signs of the odd terms of series (6.11) are not known in a general case, the location of even boundaries (6.13) with respect to $\bar{\sigma}^*$ also remains undetermined. Therefore Aleksandrov's assumption [101] that a series similar to (5.3) is alternating in signs at $\sigma^{c} = \langle \sigma \rangle$ is not of general character and is realised only in particular cases. The condition of small fluctuations, $1 - \varkappa \ll 1$ (see the Introduction), which ensures the convergence of series (5.3)and (5.4), is commonly used [102] to represent the ECT as the sum of first three terms (account for pair interactions).

5. Effective conductivity tensor

Using the definition (1.2) and the Ohm law (2.5) and (2.6) we can write for the presented material characteristics $\bar{\sigma}$ (3.20) and $\bar{\rho}$ (3.21):

$$\langle \mathbf{j} \rangle = \langle \bar{\sigma} \mathbf{e} \rangle \equiv \hat{\bar{\sigma}}^* \langle \mathbf{e} \rangle, \quad \hat{\sigma}^* = \sqrt{\sigma^c} \, \hat{\bar{\sigma}}^* \sqrt{\sigma^c}, \quad (5.1)$$

$$\langle \mathbf{e} \rangle = \langle \bar{\rho} \mathbf{j} \rangle \equiv \hat{\bar{\rho}}^* \langle \mathbf{j} \rangle, \qquad \hat{\rho}^* = \sqrt{\rho^c} \, \hat{\bar{\rho}}^* \sqrt{\rho^c} \,. \tag{5.2}$$

Substituting (4.3) and (4.4) and considering (4.2), (3.38), and (3.39), we find

$$\hat{\bar{\sigma}}^* = \left\langle \bar{\sigma} \widehat{A} \right\rangle = \sum_{k=0}^{\infty} \hat{\bar{\sigma}}^{(k)}; \qquad \hat{\bar{\sigma}}^{(k+1)} \equiv \left\langle \bar{\sigma}' (\widehat{R} \, \widehat{\bar{X}})^k \right\rangle,$$
$$k \ge 0; \qquad \hat{\bar{\sigma}}^{(0)} \equiv \widehat{I}, \qquad (5.3)$$

$$\hat{\bar{\rho}}^* = \left\langle \bar{\rho}\widehat{B} \right\rangle = \sum_{k=0}^{\infty} \hat{\bar{\rho}}^{(k)}; \qquad \hat{\bar{\rho}}^{(k+1)} \equiv \left\langle \bar{\rho}'(\widehat{R}\,\widehat{\overline{Y}})^k \right\rangle, k \ge 0; \qquad \hat{\bar{\rho}}^{(0)} \equiv \widehat{I}.$$
(5.4)

In general, $\hat{\sigma}^*$ and $\hat{\rho}^*$ are integral operators and various relations between them are understood in the sense of the appropriate integral forms of the type (3.1). If the conditions from which (1.6) and (3.7) follow are satisfied, then the statistical average is complete and $\bar{\sigma}^*$ and $\bar{\rho}^*$ are tensors. In this case the relations between the effective characteristics are understood in the sense of the appropriate quadratic form. In what follows we shall, as a rule, mean effective conductivity (σ^*) and resistance (ρ^*) tensors, for which Eqns (5.10) are satisfied.

Along with (5.3) and (5.4) other representations for $\hat{\sigma}^*$ and $\hat{\rho}^*$ can be helpful. Expressing \hat{A} and \hat{B} in the form (4.1) from (5.3) and (5.4) we have \dagger

$$\hat{\bar{\sigma}}^* = \widehat{I} + \left\langle (\bar{q}\widehat{I} + \overline{Q}\widehat{R})^{-1} \right\rangle, \quad \bar{q}\bar{\sigma}' \equiv I; \quad \hat{\bar{\sigma}}^* \Rightarrow \bar{\sigma}^*\widehat{I}, \ (5.5)$$

$$\hat{\bar{\rho}}^* = \widehat{I} + \left\langle (\bar{p}\widehat{I} + \overline{P}\widehat{R})^{-1} \right\rangle, \quad \bar{p}\bar{\rho}' \equiv I; \quad \hat{\bar{\rho}}^* \Rightarrow \bar{\rho}^*\widehat{I},$$
(5.6)

where the tensors \bar{p} and \bar{q} are related by the equations:

$$\bar{p} + \bar{q} = -I, \quad \sigma \rho = I.$$
 (5.7)

Introducing the tensors \bar{p}^* and \bar{q}^* and using (4.15) we find from (5.3) and (5.4)

$$\left(\hat{\bar{q}}^* + \hat{\overline{Q}}\right)^{-1} = \left\langle \left(\bar{q}\hat{I} + \hat{\overline{Q}}\right)^{-1} \right\rangle; \quad \hat{\bar{q}}^*(\hat{\sigma}^* - \hat{I}) \equiv \hat{I}, \quad (5.8)$$

$$\left(\hat{\bar{p}}^* + \widehat{\bar{P}}\right)^{-1} = \left\langle \left(\bar{p}\widehat{I} + \widehat{\bar{P}}\right)^{-1} \right\rangle; \quad \hat{\bar{p}}^*(\hat{\bar{\rho}}^* - \widehat{I}) \equiv \widehat{I}.$$
(5.9)

Whence it follows that

$$\bar{p}^* + \bar{q}^* = -I, \quad \sigma^* \rho^* = I.$$
 (5.10)

The representation of $\hat{\sigma}^*$ in the form (5.5) yields eventually the relation:

$$\delta\hat{\bar{\sigma}}^* = \left\langle \hat{A}^{(+)}\delta\bar{\sigma}\hat{A} \right\rangle.$$
(5.11)

It establishes the equivalence

$$\delta \bar{\sigma}^* \gtrless 0 \Leftrightarrow \delta \bar{\sigma} \gtrless 0, \quad \sigma^c = \text{const}$$
 (5.12)

between the changes $\delta \bar{\sigma}^*$ and $\delta \bar{\sigma}$ in effective and local conductivities for fixed outer conditions, including sources, boundary conditions and geometry. Similarly, it follows from (5.6) that

$$\delta\hat{\bar{\rho}}^* = \left\langle \hat{B}^{(+)}\delta\bar{\rho}\hat{B} \right\rangle,\tag{5.13}$$

$$\delta \bar{\rho}^* \ge 0 \Leftrightarrow \delta \bar{\rho} \ge 0, \quad \rho^c = \text{const}.$$
 (5.14)

[†] The property of locality of operators $\hat{\sigma}^*$ and $\hat{\rho}^*$ which is used further leads to some technical simplifications leaving unchanged the structure and sense of all relations which can be readily written in operator form.

Relations (5.11 - 5.14) may be interpreted simply in terms of energy quantities.

6. Consideration for multiparticle interactions in perturbation method

As a rule, the calculation of σ^* and ρ^* is based on a limited volume of statistical information about the RIM of interest. As a result the use of expansions (5.3) and (5.4) yields for them the approximate values

$$\Gamma_{(n)}^* \equiv \sum_{k=0}^n \Gamma^{(k)}, \qquad \Gamma \equiv \sigma, \rho; \qquad \widehat{\Gamma}^{(k)} \Rightarrow \Gamma^{(k)} \widehat{I}, \qquad (6.1)$$

in which all the *k*-particle interactions (where $k \le n$) between inhomogeneities are taken into account. Generally the quantities $\sigma_{(n)}^*$ and $\rho_{(n)}^*$ are unrelated by Eqn (5.10). Moreover, the position of $\Gamma_{(n)}^*$ relative to Γ^* remains uncontrolled. Therefore, it is important to establish the sign of the difference

$$\Gamma^* - \Gamma^*_{(q-1)} = \sum_{k=q}^{\infty} \Gamma^{(k)} \,. \tag{6.2}$$

To solve this problem the equalities [36, 37]

$$(\mathbf{e}_{k}, \bar{\sigma}' \mathbf{e}_{q-k-1}) = -(\mathbf{e}_{k}, \mathbf{e}_{q-k}) = \left(\langle \mathbf{e} \rangle, \bar{\sigma}^{(q)} \langle \mathbf{e} \rangle\right),$$
$$q-1 \ge k \ge 1,$$
(6.3)

$$(\mathbf{j}_k,\,ar{
ho}\,'\mathbf{j}_{q-k-1}) = -(\mathbf{j}_k,\,\mathbf{j}_{q-k}) = \left(\langle \mathbf{j}
angle,\,ar{
ho}\,^{(q)}\langle \mathbf{j}
angle
ight),$$

 $q-1 \ge k \ge 1 \tag{6.4}$

are used. These equalities provide for the transfer of indices in inner products and are consistent with the definitions (3.38) and (3.39), and (5.3) and (5.4). They are followed for q = 2k by the inequalities:

$$\Gamma^{(2k)} \leqslant 0, \quad k \ge 1, \tag{6.5}$$

which are corollaries of (3.26) and are independent of whether Γ' is of fixed or alternating signs. If we make the substitutions $\mathbf{e}_k \rightarrow \mathbf{e}_k + \mathbf{e}_{k+1}$ and $\mathbf{j}_k \rightarrow \mathbf{j}_k + \mathbf{j}_{k+1}$, then it follows from (6.3) and (6.4) for q = 2k that

$$\Gamma^{(2k)} + 2\Gamma^{(2k+1)} + \Gamma^{(2k+2)} \leqslant 0, \quad k \ge 1.$$
(6.6)

The summation of inequalities (6.6) over k from n to ∞ yields

$$\Gamma^{(2n)} + 2\sum_{k=2n+1}^{\infty} \Gamma^{(k)} \leq 0 \quad \text{or} \quad 2\sum_{k=2n}^{\infty} \Gamma^{(k)} \leq \Gamma^{(2n)} \leq 0.$$
(6.7)

Assuming q = 2n in (6.2) and considering (6.7) we have

$$\Gamma^* \leqslant \Gamma^*_{(2n-1)}, \quad n \ge 1; \quad \Gamma \equiv \sigma, \, \rho \,. \tag{6.8}$$

Thus, summing odd number of terms in (6.1) we arrive at the upper boundary for Γ^* . The combined use of (6.8) for σ and ρ yields two-sided boundaries both for σ^* and for ρ^* :

$$\left[\rho_{(2n-1)}^*\right]^{-1} \leqslant \sigma^* \leqslant \sigma_{(2n-1)}^* \,. \tag{6.9}$$

Whence we find for σ^* at n = 1

$$\sigma_{(1)}^{-} \equiv \langle \rho \rangle^{-1} \leqslant \sigma^* \leqslant \langle \sigma \rangle \equiv \sigma_{(1)}^{+} , \qquad (6.10)$$

where the notation is the same as in (1.11). The tensor inequalities (6.10) extend the result of Wiener (1.10) to media, local conductivities of which possess arbitrary symmetry properties and spatial distributions. Note that the calculation of bounds (6.9) is carried out without any additional restrictions on the parameters σ^{c} and ρ^{c} except (4.8) and (4.9).

If σ' and ρ' are of fixed signs [not necessarily coordinated as in (4.12)], then instead of (6.5) we have [36, 37]:

$$\Gamma^{(k)} \leq 0, \quad k \ge 1; \quad \Gamma' \equiv \Gamma - \Gamma^{c} \le 0,$$
 (6.11a)

$$\Gamma^{(2k)} \leqslant 0 \leqslant \Gamma^{(2k-1)}, \quad k \ge 1; \quad \Gamma' \ge 0.$$
(6.11b)

By virtue of inequalities (6.11) the position of $\Gamma_{(n)}^*$ (6.1) relative to Γ^* is controllable on each step of the iterative procedure. In fact, it is easy to obtain similar to (6.7) and (6.11) that

$$\sum_{k=n+1}^{\infty} \Gamma^{(k)} \leq 0, \qquad n \geq 0; \qquad \Gamma' \leq 0, \qquad (6.12a)$$

$$\sum_{k=2n+2}^{\infty} \Gamma^{(k)} \leqslant 0 \leqslant \sum_{k=2n+1}^{\infty} \Gamma^{(k)}, \quad n \ge 0; \qquad \Gamma' \ge 0.$$
 (6.12b)

Instead of (6.8) we find from (6.2), (6.12), and (4.16):

$$\Gamma^* \leqslant \Gamma^*_{(n)}, \quad n \ge 0; \quad \Gamma \leqslant \Gamma^c < \infty; \quad \sigma^c \rho^c \neq I, \ (6.13a)$$

$$\Gamma^*_{(2n)} \leqslant \Gamma^* \leqslant \Gamma^*_{(2n+1)}, \quad n \ge 0;$$

$$\frac{\Gamma}{2} < \Gamma^c \leqslant \Gamma; \quad \sigma^c \rho^c \neq I.$$

$$(6.13b)$$

Let us consider the case of (6.13a) when $\sigma' \leq 0$, $\rho' \leq 0$ and, as a result, series (5.3) and (5.4) are of constant signs. Their combined use yields two-sided bounds for Γ^* in the form:

$$\left[\boldsymbol{B}_{(n)}^{*}\right]^{-1} \leqslant \boldsymbol{\Gamma}^{*} \leqslant \boldsymbol{\Gamma}_{(n)}^{*}, \quad n \ge 0, \qquad (6.14a)$$

$$\Gamma^{c} = \Gamma^{+} \equiv \sup \Gamma = (\inf B)^{-1}; \quad B\Gamma = I, \quad \Gamma \equiv \sigma, \rho.$$
(6.14b)

In contrast to (6.9) these bounds are valid for each *n*. They are obtained thanks to additional restrictions on the parameters σ^{c} and ρ^{c} in the form of (6.11a) and (6.14b).

In the case of (6.13b) we have $\sigma' \ge 0$, $\rho' \ge 0$ and, as a result, series (5.3) and (5.4) are of alternating signs. Their combined use yields two-sided bounds for Γ^* in the form

$$\begin{bmatrix} B_{(2n-1)}^* \end{bmatrix}^{-1} \leqslant \Gamma^* \leqslant \Gamma_{(2n-1)}^*,$$

$$\Gamma_{(2n)}^* \leqslant \Gamma^* \leqslant \begin{bmatrix} B_{(2n)}^* \end{bmatrix}^{-1}; \quad n \ge 0,$$

$$\Gamma^c = \Gamma^- \equiv \inf \Gamma \equiv (\sup B)^{-1}; \quad B\Gamma = I, \quad \Gamma \equiv \sigma, \rho.$$

(6.15b)

In Figures 3a-c three possible schemes of how the two-sided boundaries $\Gamma_{(n)}^{\pm}$ for Γ^* can be narrowed are presented. These schemes present the iterative procedures (6.13)-(6.15). The quantities $\Gamma_{(n)}^*(\pm)$ and $B_{(n)}^*(\pm)$ stand for $\Gamma_{(n)}^*$ and $B_{(n)}^*$, which



are calculated using the parameters Γ^{\pm} and B^{\pm} , respectively. They are marked by points the numbering of which is consistent with the numbering of the corresponding values of $\Gamma_{(n)}^*$ and $[B_{(n)}^*]^{-1}$. Figure 3a shows that a single scheme is sufficient to

calculate two-sided bounds for Γ^* in the even approximation

provided that the two values Γ^{\pm} of the parameter Γ^{c} are given. As follows from Fig. 1b, c and (6.10), odd approximations yield two-sided bounds for any value of the parameter Γ^{c} , however Γ^{*} must be calculated by two schemes. A superficial comparison of schemes (6.14) and (6.15) does not allow one to define which scheme is more efficient. To this end a comprehensive analysis is required. However, inequalities (6.14a) are more handy because, by virtue of (6.13a) and (4.16), the restrictions on the parameter Γ^{c} are less severe than in the case of (6.13b).

For n = 0 we find from (6.14) and (6.15)

$$\Gamma_{(0)}^{-} \equiv \Gamma^{-} \leqslant \Gamma^{*} \leqslant \Gamma^{+} \equiv \Gamma_{(0)}^{+}, \qquad \Gamma \equiv \sigma, \rho.$$
(6.16)

According to (1.11) the subscript (0) implies that no statistical information about RIM is taken into account when the bounds $\Gamma_{(0)}^{\pm}$ are calculated. In the case of n = 1 the bounds (6.14) and (6.15) are the same as (6.10).

Prior to consideration of the bounds $\sigma_{(2)}^{\pm}$ we shall introduce the notation [37]:

$$\left\langle \bar{\sigma}^{\,\prime\prime} \, \widehat{\overline{Q}} \, \bar{\sigma}^{\,\prime\prime} \right\rangle \equiv \left\langle \bar{\sigma}^{\,\prime\prime} \, \overline{Q} \, \bar{\sigma}^{\,\prime\prime} \right\rangle, \qquad \left\langle \bar{\rho}^{\,\prime\prime} \, \widehat{\overline{P}} \, \bar{\rho}^{\,\prime\prime} \right\rangle \equiv \left\langle \bar{\rho}^{\,\prime\prime} \, \overline{P} \, \bar{\rho}^{\,\prime\prime} \right\rangle, (6.17)$$

$$0 \leq \overline{Q} \leq I, \quad 0 \leq \overline{P} \leq I; \quad \overline{P} + \overline{Q} = I,$$
 (6.18)

where the tensors \overline{Q} and \overline{P} are found by integration and expressed through the inner products of the type of (6.3) and (6.4). By definition (6.17), two-point probabilities of the random field $\sigma(\mathbf{r})$ should be given to calculate these tensors. The tensor relations (6.18) follow from the original operator relations (3.23) and (3.26). The only exception is (3.24), from which we have

$$\overline{Q} - \overline{Q}^2 = \sqrt{\overline{Q}} (1 - \overline{Q}) \sqrt{\overline{Q}} = \sqrt{\overline{Q}} \,\overline{P} \sqrt{\overline{Q}} \ge 0 \,,$$

$$\overline{P} - \overline{P}^2 \ge 0 \,. \tag{6.19}$$

Generally, the tensors \overline{Q} and \overline{P} depend on the parameters σ^c and ρ^c , respectively. If, however, the comparison medium is isotropic, then \overline{Q} and \overline{P} are purely geometric parameters. In this case \overline{Q} is referred to as a depolarisation tensor [1, 3, 16, 21, 37, 102].

In the approximation n = 2 we find from (5.3), (5.4), and (6.1) and (6.14), (6.15) with regard for (5.1) and (5.2) that

$$\sigma_{(2)}^* = \langle \sigma \rangle + \langle \sigma'' Q \sigma'' \rangle, \qquad \overline{Q} \equiv -\sqrt{\sigma^{c}} Q \sqrt{\sigma^{c}}; \qquad \sigma' \gtrless 0,$$
(6.20a)

$$\rho_{(2)}^* = \langle \rho \rangle + \langle \rho'' P \rho'' \rangle, \quad \overline{P} \equiv -\sqrt{\rho^{c}} P \sqrt{\rho^{c}}; \quad \rho' \ge 0.$$

If the parameters σ^c and ρ^c obey the inequalities $\Gamma' \ge 0$, then the values of $\sigma_{(2)}^*$ and $\rho_{(2)}^*$ are used according to the scheme (6.15). If not, they are used according to the scheme (6.14). In contrast to the case of n = 0, 1, the bounds (6.20) are not the best in the class of bounds n = 2, in which two-particle interactions are accounted for. These bounds were first calculated by Hashin and Shtrikman [9, 10, 38]; they will be considered later. The example presented (the case of n = 2) shows the role of the auxiliary parameters Γ^c . The arbitrary choice of Γ^c for calculation of $\Gamma_{(2)}^*$ can yield only one-sided bounds as this is the case in papers discussed by Aleksandrov in Ref. [101]; or it gives approximate values for Γ^* . The consideration of three-particle interactions is associated with cumbersome mathematical manipulations. Therefore, most papers, in which the effect has been studied, consider macroisotropic mixtures of two isotropic components. Beran has made one of the first attempts to use the statistical information that the case of n = 3 presents [14]. He proposed a simple modification of the variational method (see Section 8.4).

The approximate values of $\Gamma_{(3)}^*$ and $[B_{(3)}^*]^{-1}$ regardless of the choice of the parameters Γ^c and B^c give, according to (6.9), (6.14), and (6.15), two-sided bounds for Γ^* . The arbitrariness of the parameters σ^c and ρ^c is used below to obtain more strict bounds in the n = 3 approximation. From (5.3), (5.4), (6.1), and (3.19) we find

$$\sigma_{(3)}^* = \langle \sigma \rangle + 2 \langle \sigma'' \widehat{Q} \sigma'' \rangle + \langle \sigma'' \widehat{Q} \sigma \widehat{Q} \sigma'' \rangle, \qquad (6.21a)$$

$$\rho_{(3)}^* = \langle \rho \rangle + 2 \langle \rho'' \widehat{P} \rho'' \rangle + \langle \rho'' \widehat{P} \rho \widehat{P} \rho'' \rangle.$$
(6.21b)

Let us consider an isotropic comparison medium:

$$\sigma_{ij}^{c} = \sigma^{c} \delta_{ij}, \quad u\sigma^{c} \equiv 1; \quad \rho_{ij}^{c} = \rho^{c} \delta_{ij}, \quad v\rho^{c} \equiv 1.$$
(6.22a)

Unfortunately we are forced to accept the coincidence in notation for the tensors σ^{c} and ρ^{c} (6.20) and for the scalars σ^{c} and ρ^{c} (6.22a). It seems that the option is to use the presentation

$$u\sigma_{ij}^{c} \equiv \delta_{ij}, \quad v\rho_{ij}^{c} \equiv \delta_{ij}$$
 (6.22b)

instead of (6.22a). With regard for (6.22b) Eqn (6.21) can be rewritten as:

$$\sigma_{(3)}^{*}(u) = \langle \sigma \rangle - 2 \langle \sigma'' \overline{\widehat{Q}} \sigma'' \rangle u + \langle \sigma'' \overline{\widehat{Q}} \sigma \overline{\widehat{Q}} \sigma'' \rangle u^{2},$$

$$\widehat{Q} = -u \overline{\widehat{Q}},$$

$$\rho_{(3)}^{*}(v) = \langle \rho \rangle - 2 \langle \rho'' \overline{\widehat{P}} \rho'' \rangle v + \langle \rho'' \overline{\widehat{P}} \rho \overline{\widehat{P}} \rho'' \rangle v^{2},$$

$$\widehat{P} = -v \overline{\widehat{P}},$$

(6.23b)

where $\widehat{\overline{Q}}$ and $\widehat{\overline{P}}$ are independent of the parameters of the comparison medium and Eqns (6.23) themselves are understood in the sense of the appropriate equations for quadratic forms in the space $\widehat{\mathcal{H}}^M$ (3.36).

The arbitrariness of the auxiliary parameters u, v makes it possible to minimise the functions $\sigma_{(3)}^*(u)$ and $\rho_{(3)}^*(v)$. In notation of (4.3) and (4.4) this yields

$$\min(\mathbf{e}_{0}, \sigma_{(3)}^{*}\mathbf{e}_{0}) = \left(\mathbf{e}_{0}, \langle \sigma \rangle \mathbf{e}_{0}\right) - \frac{(\mathbf{f}_{1}, \mathbf{f}_{1})^{2}}{(\mathbf{f}_{1}, \sigma \mathbf{f}_{1})}, \quad \mathbf{f}_{1} \equiv \widehat{\overline{Q}} \sigma'' \mathbf{e}_{0},$$
(6.24a)

$$\min(\mathbf{j}_{0}, \boldsymbol{\rho}_{(3)}^{*}\mathbf{j}_{0}) = \left(\mathbf{j}_{0}, \langle \boldsymbol{\rho} \rangle \mathbf{j}_{0}\right) - \frac{\left(\mathbf{g}_{1}, \mathbf{g}_{1}\right)^{2}}{\left(\mathbf{g}_{1}, \boldsymbol{\rho}\mathbf{g}_{1}\right)}, \quad \mathbf{g}_{1} \equiv \widehat{\overline{P}} \boldsymbol{\rho}^{\,\prime\prime} \mathbf{j}_{0} \,.$$

$$(6.24b)$$

Combining (6.24a) and (6.24b) we can write similar to (6.9):

$$\left[\rho_{(3)}^{*}(v^{e})\right]^{-1} \leqslant \sigma^{*} \leqslant \sigma_{(3)}^{*}(u^{e}), \qquad \sigma^{*}\rho^{*} = I,$$
(6.25)

where u^e and v^e are the values of the parameters u and v, for which $\sigma^*_{(3)}(u)$ and $\rho^*_{(3)}(v)$ are at minimum. Whence it is seen that all the bounds in (6.9) can be improved for $n \ge 2$ in the spirit of (6.24).

The method presented here of consideration of multiparticle interactions on the basis of the perturbation series can be extended to any value of n. Only information about the appropriate *n*-point probabilities of the random field $\sigma(\mathbf{r})$ is needed to implement this program.

Bounds (6.24) and (6.25) for $\sigma' \leq 0$, $\rho' \leq 0$ can be calculated with the use of the generalised Schwartz inequality to evaluate each term of series (5.3) and (5.4) and the results of the subsequent summations.

7. Singular approximation

Often it proves to be expedient to extract the local part from the operator \hat{Q} (or \hat{P}) [see (5.5) and (1.6)] and to study its role in construction of solution of Eqns (3.20)–(3.22).

Following [103] we present $\overline{\overline{Q}}$ as the sum:

$$\widehat{\overline{Q}} = \widehat{\overline{Q}}^{s} + \widehat{\overline{Q}}^{f}, \quad \widehat{\overline{Q}}^{s} \equiv \overline{Q}\widehat{I},$$
(7.1)

of the local operator \overline{Q}^s and nonlocal operator \overline{Q}^f . The determination of the constant tensor \overline{Q} is an independent problem, the solution of which should be found in each particular case separately [103, 11, 16, 21, 23, 37]. Similar to (7.1) we write for \overline{P} :

$$\widehat{\overline{P}} = \widehat{\overline{P}}^{s} + \widehat{\overline{P}}^{f}, \qquad \widehat{\overline{P}}^{s} \equiv \overline{\overline{P}}\widehat{I}.$$
(7.2)

Below the tensors \overline{Q} and \overline{P} are defined according to (6.17)–(6.19).

One of the simplest methods to calculate the fields e and **j** and the effective characteristics $\bar{\sigma}^*$ and $\bar{\rho}^*$ with allowance for multiparticle interactions is the singular approximation (s-approximation) [21, 23, 104]:

$$\widehat{\overline{Q}} \to \widehat{\overline{Q}}^{s} = \overline{Q}\widehat{I}, \qquad \widehat{\overline{P}} \to \widehat{\overline{P}}^{s} = \overline{P}\widehat{I}.$$
 (7.3)

Substitution of (7.3) into (3.20)-(3.22) yields:

$$\mathbf{e} \to \mathbf{e}^{s} = \mathbf{e}^{c} - \overline{Q}\,\overline{\sigma}'\mathbf{e}^{s} = \langle \mathbf{e}^{s} \rangle - \overline{Q}\,\widehat{R}\overline{\sigma}'\mathbf{e}^{s}\,, \tag{7.4a}$$

$$\mathbf{j} \to \mathbf{j}^{s} = \mathbf{j}^{c} - \overline{P} \,\bar{\rho}' \mathbf{j}^{s} = \langle \mathbf{j}^{s} \rangle - \overline{P} \widehat{R} \bar{\rho}' \mathbf{j}^{s} \,. \tag{7.4b}$$

In the s-approximation the operators \widehat{A} and \widehat{B} (4.2) take the form:

$$\begin{split} \widehat{A} &\to A^{s}\widehat{I}, \qquad A^{s} = (I + \overline{Q}\widehat{R}\overline{\sigma}')^{-1} \\ &= (I + \overline{Q}\overline{\sigma}')^{-1} \langle (I + \overline{Q}\overline{\sigma}')^{-1} \rangle^{-1}, \quad (7.5a) \\ \widehat{B} &\to B^{s}\widehat{I}, \qquad B^{s} = (I + \overline{P}\widehat{R}\overline{\rho}')^{-1} \end{split}$$

$$= (I + \overline{P}\bar{\rho}')^{-1} \langle (I + \overline{P}\bar{\rho}')^{-1} \rangle^{-1} . \quad (7.5b)$$

Finally, instead of (5.5), (5.6) and (5.8), (5.9) we have

$$\bar{\sigma}^{s} = \langle \bar{\sigma}A^{s} \rangle, \quad (\bar{q}^{s} + \overline{Q})^{-1} = \langle (\bar{q} + \overline{Q})^{-1} \rangle, \quad (7.6a)$$

$$\bar{\rho}^{s} = \langle \bar{\rho}B^{s} \rangle, \quad (\bar{p}^{s} + \overline{P})^{-1} = \langle (\bar{p} + \overline{P})^{-1} \rangle.$$
 (7.6b)

Along with \overline{Q} and \overline{P} we use the tensors \overline{T} and \overline{S} specified by the equations [23, 105]

$$(I+\overline{T})\overline{Q} \equiv I, \quad (I+\overline{S})\overline{P} \equiv I,$$
(7.7)

Using them we find from (7.6):

$$(\sigma^{s} + T)^{-1} = \langle (\sigma + T)^{-1} \rangle, \quad T \equiv \sqrt{\sigma^{c}} \, \overline{T} \sqrt{\sigma^{c}}, \quad (7.8a)$$

$$\left(\rho^{s}+S\right)^{-1}=\left\langle\left(\rho+S\right)^{-1}\right\rangle, \quad ST\equiv I=\sigma^{s}\rho^{s}=\sigma^{c}\rho^{c}.$$
 (7.8b)

A scalar parameter of the type of T was introduced by Bruggerman in Ref. [2] to derive the calculation formulae for effective dielectric permittivity of a mixture of two isotropic components. Earlier Fricke [1] used a similar quantity for similar purposes.

The parametric dependence of σ^{s} on *T* makes it possible to obtain all the range of values of σ^{*} [2, 23, 37, 106]. In fact we find from (7.8) and (6.10) for T = 0 and S = 0:

$$\sigma^{s} = \langle \rho \rangle^{-1} \equiv \overline{\sigma_{(1)}}, \quad \sigma_{(1)}^{+} \equiv \langle \sigma \rangle = (\rho^{s})^{-1}.$$
(7.9)

The parameter T vanishes when $\sigma^c = 0$ or $\overline{T} = 0$ which, according to (7.7), is possible for $\overline{Q} = 1$. In another limit (S = 0) we have either $\rho^c = 0$ or $\overline{P} = 1$. In terms of the parameters σ^c and \overline{Q} this means that $\sigma^c = \infty$ or $\overline{Q} = 0$. Bounds (7.9) can be narrowed if there is a selection criterion for the parameters T and S, the substitution of which into (7.8) leads to suitable bounds for σ^* . Definition (6.17) makes it possible, in the case of the isotropic comparison medium (6.22b), to find the geometric parameter \overline{T} . In the s-approximation the issue of the appropriate choice of the parameter σ^c for calculation of the bounds $\sigma^{\pm}_{(n)}$ remains open.

Now we turn to considering variational method of calculation of $\sigma_{(n)}^{\pm}$, in which σ^{c} is found in a natural manner.

8. Variational methods of calculation of bounds for σ^*

Variational methods based on classical energy theorems often prove to be efficient in solution of different problems in theory of RIM.

8.1 Classical energy theorems

Taking into account (2.9) we introduce the functionals U_2 and U_1 in the form [37]:

$$U_2 = \int w_\sigma \, \mathrm{d}V + \int_{S_2} \varphi J_0 \, \mathrm{d}S \,, \qquad 2w_\sigma \equiv \mathbf{E} \cdot \sigma \mathbf{E} \,, \qquad (8.1a)$$

$$U_1 = -\int w_\rho \,\mathrm{d}V - \int_{S_1} \varphi_0 J_n \,\mathrm{d}S \,, \qquad 2w_\rho \equiv \mathbf{J} \cdot \rho \mathbf{J} \,. \quad (8.1b)$$

Here U_2 is the potential energy and $(-U_1)$ is a complementary energy, with $U_1 = U_2 = U$ if φ is the solution of problem (2.8), (2.9); $\rho \sigma = I$, $\mathbf{J} = \sigma \mathbf{E}$, $\mathbf{E} = -\nabla \varphi$.

According to the potential energy minimum theorem (principle) [107-109]

$$U_2 \leqslant U_2 \,, \tag{8.2a}$$

where \tilde{U}_2 is the value of U_2 on a virtual, piecewise continuously differentiable potential $\tilde{\varphi}$ obeying the relations

$$\widetilde{\mathbf{E}} = -\nabla \widetilde{\varphi}, \quad 2\widetilde{w}_{\sigma} = \widetilde{\mathbf{E}} \cdot \sigma \widetilde{\mathbf{E}}, \quad \mathbf{r} \in V;
\widetilde{\varphi} = \varphi_0, \quad \mathbf{r} \in S_1.$$
(8.3a)

However, $\tilde{\varphi}$ is not a solution to the problem (2.1), (2.2), and (2.9) because $\nabla \cdot \sigma \tilde{\mathbf{E}} \neq 0$ and the boundary conditions are not satisfied on the part S_2 of the surface S.

According to the complementary energy minimum theorem (principle) [107–109]

$$U_1 \geqslant \tilde{U}_1, \tag{8.2b}$$

where \widetilde{U}_1 is the value of U_1 on the virtual, piecewise continuously differentiable current density \widetilde{J} , for which the relations

$$\nabla \cdot \widetilde{\mathbf{J}} = 0, \qquad 2\widetilde{w}_{\rho} = \widetilde{\mathbf{J}} \cdot \rho \widetilde{\mathbf{J}}, \qquad \mathbf{r} \in V;$$

$$\widetilde{J}_{n} = J_{0}, \qquad \mathbf{r} \in S_{2}$$
(8.3b)

hold. However, $\tilde{\mathbf{J}}$ is not a solution to the problem (2.1), (2.2), and (2.9) because $\nabla \times \rho \tilde{\mathbf{J}} \neq 0$ and the boundary conditions are not satisfied on the part S_1 of the surface S.

Combining (8.2) we obtain two-sided bounds for the potential energy U of electric field in a medium:

$$\widetilde{U}_1 \leqslant U \leqslant \widetilde{U}_2 \,. \tag{8.4}$$

They can be used in both calculation schemes. Note that in σ -scheme the final results are expressed in terms of the field **E** while in ρ -scheme they are expressed in terms of **J**.

8.2 Extremal properties of energy functionals in $\vec{\mathcal{H}}$ -space

Let us consider the σ -scheme. Along with the field $\tilde{\varphi}$ from theorem (8.2a) we introduce the current density \tilde{J} , which is related to \tilde{E} from (8.3a) by the equation:

$$\widetilde{\mathbf{J}} = \sigma^{\mathrm{c}} \widetilde{\mathbf{E}} + \widetilde{\mathbf{T}}, \qquad (8.5)$$

where the vector $\widetilde{\mathbf{T}}$ of the 'polarised' current is added on the right-hand side in order that the field $\widetilde{\mathbf{J}}$ should satisfy the relations

$$\nabla \cdot \widetilde{\mathbf{J}} = 0, \quad \mathbf{r} \in V; \quad \widetilde{J}_n = J_0, \quad \mathbf{r} \in S_2.$$
 (8.6)

Kroener [110] used the term 'polarised' by analogy with the term in the electrostatic field theory to denote fields that Eshelby [111] introduced to solve the problem on an ellipsoidal inclusion. This idea have been finally shaped by Hill [107].

By solving the boundary-value problem (8.3a), (8.5), and (8.6) we have instead (2.24):

$$\widetilde{\mathbf{E}} = \mathbf{E}^{c} + \widehat{Q}\widetilde{\mathbf{T}}, \qquad \widetilde{\mathbf{T}} = \widetilde{\mathbf{J}} - \sigma^{c}\widetilde{\mathbf{E}} \neq \sigma'\widetilde{\mathbf{E}}.$$
(8.7)

With the transformations (3.18) - (3.20) we rewrite (8.5) and (8.7) in the form

$$\tilde{\mathbf{e}} = \mathbf{e}^{c} - \overline{\widehat{\mathcal{Q}}} \tilde{\boldsymbol{\tau}}, \quad \tilde{\mathbf{j}} = \tilde{\mathbf{e}} + \tilde{\boldsymbol{\tau}}, \quad \widetilde{\mathbf{T}} \equiv \sqrt{\sigma^{c}} \, \tilde{\boldsymbol{\tau}}.$$
(8.8)

To simplify further consideration we exclude the potential energy U^c of the comparison medium from inequality (8.4). Using then properties (3.4) we calculate the full average (over volume and over ensemble of realisations) of the energy characteristics of the RIM. To this end we introduce the notation:

$$\langle U - U^{c} \rangle \equiv V u', \quad \langle \widetilde{U}_{\alpha} - U^{c} \rangle \equiv V F_{\alpha}; \quad \alpha = 1, 2.$$
 (8.9)

Upon simple manipulations we have [36]:

$$2u' = (\mathbf{e}^{c}, \, \bar{\sigma}' \mathbf{e}) \,, \tag{8.10a}$$

Together with (5.1), (5.3), (5.5), (5.8) and (4.15) this yields

$$2u' = \left(\mathbf{e}^{\mathbf{c}}, \left(\hat{\bar{\sigma}}^* - \widehat{I}\right) \langle \mathbf{e} \rangle\right) = \left(\mathbf{e}^{\mathbf{c}}, \left(\hat{\bar{q}}^* + \widehat{\overline{Q}}\right)^{-1} \mathbf{e}^{\mathbf{c}}\right). \quad (8.10b)$$

By subtracting U^c from \widetilde{U}_2 , averaging over ensemble and using (8.8) with notations (8.9) and (3.30), we have

$$2F_2 = (\mathbf{e}^{\mathrm{c}}, \, \bar{\sigma}' \mathbf{e}^{\mathrm{c}}) + 2(\tilde{\mathbf{t}}, \, \vec{\mathcal{E}}_1) + (\tilde{\mathbf{t}}, \, \overline{\vec{\mathcal{Q}}} \bar{\sigma} \, \overline{\vec{\mathcal{Q}}} \tilde{\mathbf{t}}) \,. \tag{8.11}$$

Similarly, by subtracting U^c from \widetilde{U}_1 , averaging over ensemble and using the relations

$$\widetilde{\mathbf{j}} = \mathbf{j}^{c} - \overline{P} \widetilde{\mathbf{\eta}}, \quad \widetilde{\mathbf{e}} = \widetilde{\mathbf{j}} + \widetilde{\mathbf{\eta}}; \quad \widetilde{\mathbf{H}} \equiv \sqrt{\rho^{c}} \widetilde{\mathbf{\eta}}, \quad \widetilde{\mathbf{\eta}} = -\widetilde{\mathbf{\tau}},$$
(8.12)

which replace (8.8), with notation (8.9) and (3.31), we have

$$2F_1 = -(\mathbf{j}^c, \,\bar{\rho}\,'\mathbf{j}^c) - 2(\tilde{\mathbf{\eta}}, \,\vec{\mathcal{J}}_1) - (\tilde{\mathbf{\eta}}, \,\overline{\widehat{P}}\bar{\rho}\overline{\widehat{P}}\tilde{\mathbf{\eta}}) \,. \tag{8.13}$$

Thus, by virtue of (8.9), (8.11), and (8.13) we have

$$F_1 \leqslant u' \leqslant F_2 \tag{8.14}$$

instead of (8.4). The first and second functional (variational) derivatives of F_{α} are needed to study their extremal properties. In notation adopted in Ref. [30] the second derivatives of F_{α} take the form:

$$\frac{\delta}{\delta\tilde{\boldsymbol{\eta}}} \otimes \frac{\delta}{\delta\tilde{\boldsymbol{\eta}}} F_1 = -\widehat{\overline{P}}\overline{\rho}\widehat{\overline{P}} < 0, \qquad \frac{\delta}{\delta\tilde{\boldsymbol{\tau}}} \otimes \frac{\delta}{\delta\tilde{\boldsymbol{\tau}}} F_2 = \widehat{\overline{Q}}\overline{\sigma}\widehat{\overline{Q}} > 0.$$
(8.15)

With inequalities (8.15) the following variational principles can be formulated.

(1) A functional F_1 of the form

$$F_{1}[\boldsymbol{\eta}] = -\frac{1}{2} (\mathbf{j}^{c}, \, \bar{\rho}' \mathbf{j}^{c}) - (\boldsymbol{\eta}, \, \vec{\mathcal{J}}_{1}) - \frac{1}{2} (\boldsymbol{\eta}, \, \widehat{\overline{P}} \bar{\rho} \widehat{\overline{P}} \boldsymbol{\eta}) \,, \quad (8.16)$$

which is defined in the field of 'polarised' intensity η belonging to a Hilbert space $\vec{\mathcal{H}}$, is, provided that

$$\bar{p}\mathbf{\eta} = \mathbf{j}^{c} - \widehat{P}\mathbf{\eta} = \mathbf{j}, \quad \bar{p}\,\bar{\rho}\,' = I,$$
(8.17)

at maximum

$$F_{1}^{\max} = -\frac{1}{2} (\mathbf{j}^{c}, \bar{\rho}' \mathbf{j}^{c}) - \frac{1}{2} (\mathbf{\eta}, \ \vec{\mathcal{J}}_{1}) = -\frac{1}{2} (\mathbf{j}^{c}, \mathbf{\eta}), \quad (8.18)$$

equal to the true potential energy

$$u' = -\frac{1}{2} (\mathbf{j}^{c}, \, \bar{\rho}' \mathbf{j}) = -\frac{1}{2} (\mathbf{j}^{c}, \, (\bar{\rho}\widehat{I} + \widehat{\overline{P}})^{-1} \mathbf{j}^{c}) \,. \tag{8.19}$$

(2) A functional F_2 of the form

$$F_{2}[\tau] = \frac{1}{2} (\mathbf{e}^{c}, \, \bar{\sigma}' \mathbf{e}^{c}) + (\tau, \, \vec{\mathcal{E}}_{1}) + \frac{1}{2} (\tau, \, \widehat{\overline{\mathcal{Q}}} \bar{\sigma} \widehat{\overline{\mathcal{Q}}} \tau) \,, \qquad (8.20)$$

which is defined in the field of the 'polarised' current τ belonging to a Hilbert space $\vec{\mathcal{H}}$, is, provided that

$$\bar{q}\mathbf{\tau} = \mathbf{e}^{c} - \widehat{\overline{Q}}\mathbf{\tau} = \mathbf{e}, \quad \bar{q}\,\bar{\sigma}\,' = I,$$
(8.21)

at minimum

$$F_2^{\min} = \frac{1}{2} (\mathbf{e}^c, \, \bar{\sigma}' \mathbf{e}^c) + \frac{1}{2} (\tau, \, \vec{\mathcal{E}}_1) = \frac{1}{2} (\mathbf{e}^c, \tau) \,, \qquad (8.22)$$

equal to the true potential energy

$$u' = \frac{1}{2} (\mathbf{e}^{c}, \, \bar{\sigma}' \mathbf{e}) = \frac{1}{2} \left(\mathbf{e}^{c}, \, (\bar{q}\hat{I} + \widehat{\overline{Q}})^{-1} \mathbf{e}^{c} \right). \tag{8.23}$$

With the equality $\tau = -\eta$ (8.12) both variational principles can be formulated in terms of either τ or η .

If in principles 1 and 2 we impose certain restrictions on the approximating fields $\tilde{\tau}$ and $\tilde{\eta}$, then, upon their application, we obtain bounds F^{\pm} for u'. This case is considered later (see Section 9).

8.3 Generalisation of Hashin-Shtrikman variational principles

Another way to simplify the functionals F_{α} from (8.16) and (8.20) is to impose restrictions on the parameters σ^{c} and ρ^{c} of the comparison medium. Using Hill's idea [107] we transform inequalities (8.4) as:

$$-R_{1} \leq u' - M \leq R_{2}, \quad 2M[\tilde{\mathfrak{t}}] = (\tilde{\mathfrak{t}}, 2\mathbf{e}^{c} - \overline{Q}\tilde{\mathfrak{t}} - \bar{q}\tilde{\mathfrak{t}}),$$

$$(8.24a)$$

$$2R_{1} \equiv (\tilde{\mathbf{f}}, \bar{\rho}'\tilde{\mathbf{f}}), \quad 2R_{2} \equiv (\tilde{\mathbf{f}}, \bar{\sigma}'\tilde{\mathbf{f}}); \quad \tilde{\mathbf{f}} \equiv \tilde{\mathbf{e}} - \bar{q}\tilde{\mathfrak{t}}(8.24b)$$

Inequalities (8.24a) generalise the Hashin–Shtrikman variational principles [9, 10] to the case of boundary conditions (2.9). In addition, and this is more essential, the projective operator \widehat{Q} is present in the formulations of principles. Apart from clarification and simplification of previous results this makes it possible to obtain new results (with consideration of three-particle interactions) on the basis of principle 1 and 2 from Section 8.2. R_1 and R_2 , which are defined through tensors ρ' and σ' , have the opposite signs because of (4.12). If we restrict the choice of the parameters ρ^c and σ^c by the conditions $R_1 \leq 0$ and $R_2 \leq 0$, then, discarding R_1 or R_2 , we have from (8.24):

$$M^{-} \leq u', \quad R_{1} \leq 0, \quad \bar{\rho}' \leq 0 \leq \bar{\sigma}', \quad (8.25a)$$

$$u' \leq M^+$$
, $R_2 \leq 0$, $\bar{\sigma}' \leq 0 \leq \bar{\rho}'$, (8.25b)

where M^- and M^+ are the values of the functional M when the parameters ρ^c and σ^c obey the inequalities $R_1 \leq 0$ and $R_2 \leq 0$, respectively. As a result, bounds (8.25) are weaker than those in (8.14).

To study the extremal properties of M we write the first and second derivatives of the M functional from (8.24a)

$$\frac{\delta}{\delta\tilde{\tau}} M = \mathbf{e}^{c} - \left(\bar{q}\widehat{I} + \widehat{\overline{Q}}\right)\tilde{\tau}, \qquad \frac{\delta}{\delta\tilde{\tau}} \otimes \frac{\delta}{\delta\tilde{\tau}} M = -\left(\bar{q}\widehat{I} + \widehat{\overline{Q}}\right).$$
(8.26)

With regard for (8.8), equating the first derivative to zero leads, in contrast to (8.7), to the relations

$$\bar{q}\tilde{\mathbf{\tau}} = \mathbf{e}^{\mathbf{c}} - \overline{Q}\tilde{\mathbf{\tau}} = \tilde{\mathbf{e}} \Rightarrow \tilde{\mathbf{\tau}} = \bar{\sigma}'\tilde{\mathbf{e}}, \qquad (8.27)$$

equivalent to (2.24) and valid for the field \tilde{e} , for which (2.1), (2.2), and (2.9) are satisfied; i.e., because of the uniqueness of the solution, this field is the same as the field e. Given (8.27) the type of the extremum M is determined by the sign of the second derivative (8.26) written in the operator form.

Let $\bar{q} \ge 0$. It follows then from (3.23), (3.26), (4.12) and (5.5)–(5.7) that

$$\widehat{O} \leqslant \bar{q}\widehat{I} + \widehat{\overline{Q}} = -\left(\bar{p}\widehat{I} + \widehat{\overline{P}}\right), \qquad \bar{\rho}' \leqslant 0 \leqslant \bar{\sigma}', \qquad (8.28)$$

and, consequently, the extremum is a maximum. In the opposite case of $\bar{q} \leq 0$ we find instead of (8.28) that

$$\widehat{O} \leqslant \overline{p}\widehat{I} + \overline{\widehat{P}} = -\left(\overline{q}\widehat{I} + \overline{\widehat{Q}}\right), \quad \overline{\sigma}' \leqslant 0 \leqslant \overline{\rho}'.$$
(8.29)

Hence, the second derivative of the functional M (8.26) is positive and the extremum of interest is a minimum. It is easy to see that, given (8.27), the extremal value M_e coincides with the true potential energy u' (8.10). With regard for (8.27)– (8.29) it follows from (8.10) and (8.25) that

$$2M_{\rm e}^{\pm} = \left(\mathbf{\tau}, \, (\bar{q}\widehat{I} + \overline{\widehat{Q}})\mathbf{\tau}\right), \quad \bar{\sigma}' \leq 0, \quad \tilde{\mathbf{\tau}} = \mathbf{\tau}.$$
(8.30)

To summarise what has been said we shall formulate the variational principle.

(3) A functional M of the form

$$M = M[\mathbf{\tau}] \equiv \frac{1}{2} \left(\mathbf{\tau}, \, 2\mathbf{e}^{\,\mathbf{c}} - \widehat{\overline{Q}} \,\mathbf{\tau} - \bar{q} \,\mathbf{\tau} \right), \tag{8.31}$$

which is defined on the field of a 'polarised' current τ in a Hilbert space $\vec{\mathcal{H}}$, reaches its extremum when

$$\bar{q}\mathbf{\tau} = \mathbf{e}^{\,\mathbf{c}} - \widehat{\overline{Q}}\mathbf{\tau} = \mathbf{e}\,, \quad \bar{q}\,\bar{\sigma}\,' = I.$$
 (8.32)

Its extremal value M_e equal to the true potential energy u' is a minimum

$$M_{\rm e}^{+} = \frac{1}{2} \left(\mathbf{\tau}, \ (\bar{q}\hat{I} + \widehat{\overline{Q}})\mathbf{\tau} \right) = u' \leqslant 0 \,, \tag{8.33a}$$

if $\bar{\sigma}' \leq 0$, and it is a maximum

$$M_{\rm e}^{-} = \frac{1}{2} \left(\mathbf{\tau}, \, (\bar{q}\hat{I} + \widehat{\overline{Q}})\mathbf{\tau} \right) = u' \ge 0 \,, \tag{8.33b}$$

if $\bar{\sigma}' \ge 0$.

(4) A functional N of the form

$$N = N[\mathbf{\eta}] \equiv \frac{1}{2} \left(\mathbf{\eta}, \, 2\mathbf{j}^{c} - \widehat{\overline{P}}\mathbf{\eta} - \overline{p}\mathbf{\eta} \right), \qquad (8.34)$$

which is defined on the field of a 'polarised' intensity η in a Hilbert space $\vec{\mathcal{H}}$, reaches its extremum when

$$\bar{p}\mathbf{\eta} = \mathbf{j}^{c} - \widehat{\overline{P}}\mathbf{\eta} = \mathbf{j}, \quad \bar{p}\,\bar{\rho}\,' = I.$$
(8.35)

Its extremal value N_e equal to the true potential energy u' taken with the opposite sign is a minimum

$$N_{\rm e}^+ = \frac{1}{2} \left(\mathbf{\eta}, \, (\bar{p}\hat{I} + \widehat{P})\mathbf{\eta} \right) = -u' \leqslant 0 \,, \tag{8.36a}$$

if $\bar{\rho}' \leq 0$ and it is a maximum

$$N_{\rm e}^{-} = \frac{1}{2} \left(\mathbf{\eta}, \, (\bar{p}\widehat{I} + \widehat{\overline{P}})\mathbf{\eta} \right) = -u' \ge 0 \,, \tag{8.36b}$$

if $\bar{\rho}' \ge 0$.

Note that the fourth variational principle is an alternative to the third and is obtained by calculating bounds (8.4) in the ρ -scheme. In addition it, as well as the third, is weaker than the classical principles in Section 8.2 and, like (6.25), it leads to less severe bounds than (8.14).

In contrast to the familiar principles [9, 10] the above variational principles include projective operators $\overline{\hat{Q}}$ and $\overline{\hat{P}}$ and, as a result, their range of applicability can be extended with the use of the iterative procedure from Sections 4 and 6. The capabilities of the Hashin–Shtrikman method are essentially limited by the lack of the operators $\overline{\hat{Q}}$ and $\overline{\hat{P}}$.

Only the simplest problems for a RIM were solved by their method.

8.4 Beran's approximation

Beran was the first who found bounds for σ^* with regard for three-point probabilities [14, 31]. He calculated an effective dielectric permittivity ϵ^* of a statistically uniform and isotropic unbounded RIM with the boundary conditions equivalent to (1.4) or (1.5). His task was further simplified by calculating only the upper bound (8.4) in the first case and only the lower bound in the second case.

Functionals (8.1) take the form

$$U_1 = -\int w_\rho \, \mathrm{d}V - \int_S \varphi_0 J_n \, \mathrm{d}S \,,$$

$$U_2 = \int w_\sigma \, \mathrm{d}V \equiv W_\sigma \,; \quad S_2 = 0 \,, \qquad (8.37a)$$

when boundary conditions (1.4) are satisfied, and

$$U_{1} = -\int w_{\rho} \, \mathrm{d}V \equiv W_{\rho} ,$$

$$U_{2} = \int w_{\sigma} \, \mathrm{d}V + \int_{S} \varphi J_{0} \, \mathrm{d}V; \qquad S_{1} = 0 \qquad (8.37b)$$

in the case of (1.5). For the electric field energy

$$W = W_{\rho} = W_{\sigma} = \frac{1}{2} \int \mathbf{e} \cdot \mathbf{j} \, \mathrm{d}V \tag{8.38}$$

by virtue of (8.4) and in accordance with (8.37) we have

$$\widetilde{U}_1 \leqslant W_\sigma \leqslant \widetilde{U}_2 \equiv \widetilde{W}_\sigma \,, \tag{8.39a}$$

$$-\widetilde{W}_{\rho} \equiv \widetilde{U}_1 \leqslant -W_{\rho} \leqslant \widetilde{U}_2.$$
(8.39b)

Using the right Beran inequality from (8.39a) and the left Beran inequality from (8.39b) we formulate the variational minimum principles for the functionals

$$2\widetilde{W}_{\rho} \equiv \int \tilde{\mathbf{j}} \cdot \bar{\rho} \tilde{\mathbf{j}} \, \mathrm{d}V, \qquad 2\widetilde{W}_{\sigma} \equiv \int \tilde{\mathbf{e}} \cdot \bar{\sigma} \tilde{\mathbf{e}} \, \mathrm{d}V, \qquad (8.40)$$

from which the upper bounds

$$\hat{\rho}^* \leqslant \hat{\rho}_*^+, \quad \hat{\sigma}^* \leqslant \hat{\sigma}_*^+ \tag{8.41}$$

for the effective characteristics follow.

Inequalities (8.41) are obtained by solving different boundary-value problems and because of this the combined use of them to find two-sided bounds [14, 31] is equivalent to the assumption that $\hat{\sigma}^*$ is independent of boundary conditions. The last proposition is true only if the medium is unbounded and, hence, there are no boundary effects.

To find $\hat{\rho}_*^+$ and $\hat{\sigma}_*^+$ from (8.40) we must define virtual fields \tilde{j} and \tilde{e} , which satisfy (8.3b) for $S_2 = S$ and (8.3a) for $S_1 = S$, respectively. In notation of (4.3) and (4.4) the approximating Beran fields take the form:

$$\tilde{\mathbf{j}} = \mathbf{j}_0 + v\mathbf{j}_1, \quad \tilde{\mathbf{e}} = \mathbf{e}_0 + u\mathbf{e}_1, \quad (8.42)$$

where v and u are arbitrary scalar parameters. Substitution of (8.42) into (8.40) and subsequent minimisation of functions (6.23) yield the same bounds as those in (6.24) and (6.25) which we obtained in another way. Beran's method proposed

for a macroisotropic medium [14] was used to calculate σ_B^{\pm} in three-dimensional [9] and two-dimensional [22] macroisotropic mixtures of two homogeneous isotropic components. As for approximation (6.24), (6.25) it applies for an RIM, symmetry properties of which are arbitrary both in microscale and in macroscale.

9. Virtual polarised fields

The choice of virtual fields used to approximate the functionals in Section 8 affects essentially the solution pattern and accuracy of the solutions obtained with the use of variational principles.

We shall demonstrate this fact by the example of fields (8.42). Bounds (6.25) can be narrowed if, instead of (8.42), we take the fields

$$\tilde{\mathbf{j}} = \left(\widehat{I} - \overline{\widehat{P}}\overline{\rho}\,''g\right)\mathbf{j}_0\,, \qquad \tilde{\mathbf{e}} = \left(\widehat{I} - \overline{\widehat{Q}}\overline{\sigma}\,''f\right)\mathbf{e}_0\,, \tag{9.1}$$

where g and f are regular tensor fields found by varying functionals (8.40). Substituting (9.1) into (8.40) we write instead of (6.23):

$$2\widetilde{W}[g] = \left(\mathbf{j}_0, \left[\langle \bar{\rho} \rangle \widehat{I} + 2\hat{\bar{\rho}}^{(2)}g + g(\hat{\bar{\rho}}^{(3)} - \hat{\bar{\rho}}^{(2)})g\right]\mathbf{j}_0\right), \quad (9.2a)$$

$$2\widetilde{W}[f] = \left(\mathbf{e}_{0}, \left[\langle \bar{\sigma} \rangle \widehat{I} + 2\hat{\bar{\sigma}}^{(2)}f + f(\hat{\bar{\sigma}}^{(3)} - \hat{\bar{\sigma}}^{(2)})f\right]\mathbf{e}_{0}\right).$$
(9.2b)

Here we use the notation of (5.3) and (5.4). Extrema $\widetilde{W}[g]$ and $\widetilde{W}[f]$ are minima and they are reached when

$$g^{e}\widehat{I} = \left[\hat{\bar{\rho}}^{(2)} - \hat{\bar{\rho}}^{(3)}\right]^{-1}\hat{\bar{\rho}}^{(2)}, \quad f^{e}\widehat{I} = \left[\hat{\bar{\sigma}}^{(2)} - \hat{\bar{\sigma}}^{(3)}\right]^{-1}\hat{\bar{\sigma}}^{(2)}.$$
(9.3)

The resultant upper bounds are:

$$\hat{\bar{\rho}}^{+} = \langle \bar{\rho} \rangle \hat{I} - \langle \bar{\rho}^{\,\prime\prime} \, \hat{\overline{P}} \bar{\rho}^{\,\prime\prime} \rangle \langle \bar{\rho}^{\,\prime\prime} \, \hat{\overline{P}} \bar{\rho} \hat{\overline{P}} \bar{\rho}^{\,\prime\prime} \rangle^{-1} \langle \bar{\rho}^{\,\prime\prime} \, \hat{\overline{P}} \bar{\rho}^{\,\prime\prime} \rangle , \qquad (9.4a)$$

$$\hat{\bar{\sigma}}^{+} = \langle \bar{\sigma} \rangle \hat{I} - \left\langle \bar{\sigma}^{\,\prime\prime} \, \overline{Q} \bar{\sigma}^{\,\prime\prime} \right\rangle \left\langle \bar{\sigma}^{\,\prime\prime} \, \overline{Q} \bar{\sigma} \overline{Q} \bar{\sigma}^{\,\prime\prime} \right\rangle^{-1} \left\langle \bar{\sigma}^{\,\prime\prime} \, \overline{Q} \bar{\sigma}^{\,\prime\prime} \right\rangle. \tag{9.4b}$$

Using the Schwartz inequality [99]

$$\begin{aligned} \left| (\Psi_1, \Psi_2) \right|^2 &\leqslant (\Psi_1, \Psi_1) (\Psi_2, \Psi_2) \,, \\ \Psi_1 &\equiv \widehat{B}^{1/2} \mathbf{e}_0 \,, \qquad \Psi_2 \equiv \widehat{B}^{-1/2} \widehat{A} \mathbf{e}_0 \,; \\ \widehat{B} &\equiv \left\langle \bar{\sigma}^{\,\prime\prime} \, \overline{\widehat{Q}} \bar{\sigma} \overline{\widehat{Q}} \bar{\sigma}^{\,\prime\prime} \right\rangle \,, \qquad \widehat{A} \equiv \left\langle \bar{\sigma}^{\,\prime\prime} \, \overline{\widehat{Q}} \bar{\sigma}^{\,\prime\prime} \right\rangle \,, \end{aligned} \tag{9.5}$$

we find

$$\min(\mathbf{e}_{0}, \hat{\bar{\sigma}}_{(3)}^{*} \mathbf{e}_{0}) \geqslant (\mathbf{e}_{0}, \hat{\bar{\sigma}}^{+} \mathbf{e}_{0})$$
(9.6a)

from (6.24a) and (9.4b) and, similarly,

$$\min(\mathbf{j}_0, \hat{\bar{\rho}}^*_{(3)} \mathbf{j}_0) \ge (\mathbf{j}_0, \hat{\bar{\rho}}^+ \mathbf{j}_0)$$
(9.6b)

from (6.24b) and (9.4a). Bounds (9.4) are narrower than those of Beran [14].

Now we shall consider piecewise uniform random fields $\tilde{\tau}$ and $\tilde{\eta}$, which are used in various modifications of the Hashin–Shtrikman method [9, 10, 17, 18, 38, 107]. We shall examine only the variational principles from Section 8.2 because they are stronger than those from Section 8.3 and the principles from Section 8.4 are their special cases. To

simplify mathematical calculations we use inequality (2.31) since it yields bounds for σ^* without additional manipulations.

Thus, we solve the extremal problem for functionals F_1 (8.16) and F_2 (8.20) written in the form

$$2F_{1} = \left(\langle \mathbf{j} \rangle, \left(I - \bar{\rho}^{+}[f_{1}]\right) \langle \mathbf{j} \rangle\right),$$

$$2F_{2} = \left(\langle \mathbf{e} \rangle, \left(\bar{\sigma}^{+}[f_{2}] - I\right) \langle \mathbf{e} \rangle\right), \qquad (9.7)$$

$$\bar{\rho}^{+}[f_{1}] = \langle \bar{\rho} \rangle - \left\langle f_{1}^{\prime\prime} \overline{P} \bar{\rho}^{\prime\prime} \right\rangle - \left\langle \bar{\rho}^{\prime\prime} \overline{P} f_{1}^{\prime\prime} \right\rangle$$

$$+ \left\langle f_{1}^{\prime\prime} [\overline{P} + \overline{P} \bar{\rho}_{j}^{\prime} \overline{P}] f_{1}^{\prime\prime} \right\rangle, \qquad f_{1} = f_{1}(\bar{\rho}^{\prime}), \qquad (9.8a)$$

$$\bar{\sigma}^{+}[f_{2}] = \langle \bar{\sigma} \rangle - \left\langle f_{2}^{\prime\prime} \overline{Q} \bar{\sigma}^{\prime\prime} \right\rangle - \left\langle \bar{\sigma}^{\prime\prime} \overline{Q} f_{2}^{\prime\prime} \right\rangle$$

$$+ \left\langle f_2''[\overline{Q} + \overline{Q}\overline{\sigma}_j'\overline{Q}]f_2''\right\rangle, \quad f_2 = f_2(\overline{\sigma}'), \quad (9.8b)$$

where along with (4.18) and (6.17) we use the notation:

$$\langle f_1'' \,\overline{P} \overline{\rho} \overline{P} f_1'' \rangle \equiv \langle f_1'' [\,\overline{P} + \overline{P} \overline{\rho}_j' \,\overline{P}] f_1'' \rangle ,$$

$$\rho \equiv \sqrt{\rho^j} (I + \overline{\rho}_j') \sqrt{\rho^j} ,$$

$$\langle f_2'' \,\overline{Q} \overline{\sigma} \widehat{\overline{Q}} f_2'' \rangle \equiv \langle f_2'' [\,\overline{Q} + \overline{Q} \overline{\sigma}_j' \,\overline{Q}] f_2'' \rangle ,$$

$$\sigma \equiv \sqrt{\sigma^j} (I + \overline{\sigma}_j') \sqrt{\sigma^j} .$$

$$(9.9b)$$

Since tensors f_1 and f_2 are piecewise uniform and $\langle \mathbf{j} \rangle$ and $\langle \mathbf{e} \rangle$ are uniform, the statistical averages of (9.9) are tensors and their representation in the forms of the right-hand sides of inequalities (9.9) is possible because of arbitrariness of parameters ρ^c and σ^c . Thus, relations (9.9) give the ρ^j and σ^j values of the parameters ρ^c and σ^c , the substitution of which into, respectively, F_1 and F_2 result in (9.7) and (9.8). Definitions (9.9) can be rewritten in the form:

$$(g_1'', \bar{\rho}_j' g_1'') \equiv 0, \qquad g_1 \equiv \left(\widehat{\overline{P}} - \overline{P} \,\widehat{I}\right) f_1, \qquad (9.10a)$$

$$(g_2'', \overline{\sigma}_j'g_2'') \equiv 0, \quad g_2 \equiv (\overline{Q} - \overline{Q}\,\widehat{I})f_2.$$
 (9.10b)

Here index *j* indicates that the value of a statistical parameter is calculated with the use of three-point probabilities of the statistical field $\sigma(\mathbf{r})$. This parameter (or, generally, parameters) can be interpreted geometrically because it carries information about the pattern of the spatial distribution of inhomogeneities (in the amount that three-point probabilities present).

Varying the functionals F_1 and F_2 or, what is the same, the tensors $\bar{\rho}^+[f_1]$ and $\bar{\sigma}^+[f_2]$ over the virtual fields f_1 and f_2 , respectively, we obtain:

$$\bar{\rho}_{(3)}^{+} \equiv \bar{\rho}^{+}[f_{1}^{e}] = I + \langle f_{1}^{e} \rangle = I + \langle (\bar{p}_{j} + \overline{P}R)^{-1} \rangle, \quad \bar{p}_{j}\bar{\rho}_{j}' \equiv I,$$

$$(9.11a)$$

$$\bar{\sigma}_{(3)}^{+} \equiv \bar{\sigma}^{+}[f_{2}^{e}] = I + \langle f_{2}^{e} \rangle = I + \langle (\bar{q}_{j} + \overline{Q}\widehat{R})^{-1} \rangle, \quad \bar{q}_{j}\bar{\sigma}_{j}' \equiv I,$$

$$(9.11b)$$

Solution (9.11) has the form of (7.6) and, in contrast to the singular approximation, we have found relations (9.10) from which the parameters ρ^{j} and σ^{j} can be determined uniquely for two reference media $(\rho^{j}\sigma^{j} \ge I)$; substitution of these parameters into (9.11a, b) yields the bounds $\sigma_{(3)}^{\pm}$ for σ^{*} .

Using the 'reversibility' of the formulae of the s-approximation (7.8) we rewrite bounds (9.11) in the form:

$$\sigma_F^- \leqslant \sigma^* \leqslant \sigma_F^+, \quad \sigma_F^\pm \equiv \sigma^{\rm s}(\sigma_j^\pm), \quad \sigma^{\rm s} \equiv \sigma^{\rm s}(\sigma^{\rm c}); (9.12)$$

$$\sigma_j^+ \equiv \sigma^j, \quad \sigma_j^- \equiv (\rho^j)^{-1}; \quad \sigma_j^- \leqslant \sigma_j^+.$$
(9.13)

It can be shown [21, 37] that the variational principles from Section (8.3) result in the bounds

$$\sigma_{MN}^{-} \leqslant \sigma^{*} \leqslant \sigma_{MN}^{+}, \quad \sigma_{MN}^{\pm} \equiv \sigma^{s}(\sigma^{\pm})$$
(9.14)

which can be presented in the same form as bounds (9.12).

The parameters $\sigma^{c} = \sigma^{\pm}$ from (6.14b) and (6.15b) are related to σ_{i}^{\pm} by the inequalities

$$\sigma^- \leqslant \sigma_i^- \leqslant \sigma_i^+ \leqslant \sigma^+ \,. \tag{9.15}$$

This chain of inequalities follows from the inequalities

$$M^{-} \leq M - R_{1} = F_{1} \leq u' \leq F_{2} = M + R_{2} \leq M^{+},$$
 (9.16)

which, in turn, follow from (8.14), (8.24a) and similar inequalities in which the functional N is present.

Inequalities (9.15) permit one to find the limiting value for the structural parameter *j*. In the simplest case of a mixture of two homogeneous isotropic components from (9.15) we find [37, 83]:

$$j \in [0,1]; \quad \sigma_j^+ = \sigma_j^- = \begin{cases} \sigma^+, & j = j^+ = 1, \\ \sigma^-, & j = j^- = 0. \end{cases}$$
(9.17)

Introducing, similar to (1.11), (6.14b), (6.15b), and (6.16), the notation

$$\begin{split} \Gamma_{(0)}^{\pm} &\equiv \Gamma^{\pm} , \qquad \Gamma_{(1)}^{+} \equiv \langle \Gamma \rangle , \qquad \Gamma_{(1)}^{-} \equiv \langle \Gamma^{-1} \rangle^{-1} , \\ \Gamma_{(2)}^{\pm} &\equiv \Gamma^{s}(\Gamma^{\pm}) , \qquad \Gamma_{(3)}^{\pm} \equiv \Gamma^{s}(\Gamma_{j}^{\pm}) , \qquad \Gamma^{s} \equiv \Gamma^{s}(\Gamma^{c}) , \ (9.18) \end{split}$$

we write down the chain of narrowing bounds $\Gamma_{(n)}^{\pm}$ for Γ^*

$$\Gamma_{(0)}^{-} \leqslant \Gamma_{(1)}^{-} \leqslant \Gamma_{(2)}^{-} \leqslant \Gamma_{(3)}^{-} \leqslant \Gamma^{*} \leqslant \Gamma_{(3)}^{+} \leqslant \Gamma_{(2)}^{+} \leqslant \Gamma_{(1)}^{+} \leqslant \Gamma_{(0)}^{+};$$

$$\Gamma \equiv \sigma, \rho.$$
(9.19)

Here equalities are valid when the relative geometrical parameters take their limiting values.

Bounds (9.12) incorporate information about an RIM in the amount that three-point probabilities present (threeparticle interactions between inhomogeneities) and are the best in the piecewise uniform approximation for random fields of 'polarised' current $\tilde{\tau}$ and 'polarised' intensity $\tilde{\eta}$.

Bounds (9.12) can be narrowed down even further if *n*point probabilities (where $n \ge 4$) are taken into consideration. This can be done by introducing, according to (3.38), summands of the form $\bar{\sigma}' \mathbf{e}_k$ (where $k \ge 1$) into $\tilde{\tau}$ (and similarly into $\tilde{\eta}$).

10. Exact solutions

Apart from (1.7) only several exact solutions have been found for σ^* . Moreover, as in the case of (1.7), media the ECT of which are presented by simple analytical formulae are highly symmetric. This is due to the fact that the spatial distributions of inhomogeneities in such media correspond to specific limiting structures.

10.1 Completely symmetric media

For two-dimensional mixture of two isotropic components (inclusions of the same shape and size are distributed in the matrix so that their centres form a rectangular lattice the symmetry axes of which coincide with the axes of inclusions) the theorem has been proved [13] that the x component $\sigma_{xx}^*(\sigma_1, \sigma_2)$ of the ECT for the medium, the matrix of which is described by the conductivity σ_1 [see the notation in (1.9)] and the inclusions of which have the conductivity σ_2 , is related to the y component $\sigma_{yy}^*(\sigma_2, \sigma_1)$ for the medium, the conductivity of the matrix of which is σ_2 and that of the inclusions of which is σ_1 , by the equation:

$$\sigma_{xx}^*(\sigma_1, \sigma_2)\sigma_{yy}^*(\sigma_2, \sigma_1) = \sigma_1\sigma_2, \qquad (10.1)$$

when the inner structures of the media are the same. Mendelson [25] has shown that the Keller theorem is valid for milder restrictions on the geometry of inclusions and their spatial distribution. In another way similar relations have been obtained by Dykhne in Ref. [20], in which he presented a symmetry transform for the media from the Keller theorem. The case of nontextured polycrystalline media have been considered in Refs [20, 28, 39, 40, 112] (the Dykhne transform) and in Refs [113, 114] (the Keller theorem).

In what follows we shall use the generalised Keller theorem applicable for three-dimensional media. To modify inequalities (10.1) we introduce the notation:

$$\sigma_{i}^{*} \equiv \sigma^{*}(\sigma_{1}, \sigma_{2}; v_{1}, v_{2}; \overline{Q}_{ii}) \equiv \sigma^{*}(\overline{Q}_{ii}), \quad i \in [1, 3],$$

$$\tilde{\sigma}_{i}^{*} \equiv \sigma^{*}(\sigma_{2}, \sigma_{1}; v_{1}, v_{2}; \overline{P}_{ii}) \equiv \tilde{\sigma}^{*}(\overline{Q}_{ii});$$

$$\overline{P}_{ii} = 1 - \overline{Q}_{ii}, \quad \sum_{i=1}^{3} \overline{Q}_{ii} = 1,$$
(10.2)

where \overline{Q}_{ij} and \overline{P}_{ij} are components of the tensors \overline{Q} and \overline{P} , which are given by (6.17)–(6.19) and depend on inner geometry (structure) of the medium. In what follows we shall consider the isotropic comparison medium (6.22) when \overline{Q} has the meaning of the depolarisation tensor.

As is seen from (7.6) the tensor \overline{Q} affects essentially the value and symmetry properties of σ^{s} (similarly, in the case of ρ^{s}). The variation $\delta \overline{\sigma}^{s}$ caused by a change in the structure $(\delta \overline{Q} \neq 0 = \delta \sigma^{c})$ takes, by virtue of (7.6) and (6.18), the form

$$\delta \bar{\sigma}^{s} = \left\langle (\bar{q} + \overline{Q}\hat{R})^{-1}\hat{R}\delta \overline{P}\,\hat{R}(\bar{q} + \overline{Q}\hat{R})^{-1} \right\rangle, -\delta \overline{Q} = \delta \overline{P} \neq 0 = \delta \sigma^{c}.$$
(10.3)

Equation (10.3) means that σ^{s} increases with the structural parameter \overline{P} and reaches bounds (7.9) for limiting values of \overline{P} (6.18). Similarly, it follows from (5.5), (3.23), (7.1) and (7.2) that

$$\begin{split} \delta \hat{\sigma}^* &= \left\langle \left(\bar{q} \widehat{I} + \overline{\widehat{Q}} \widehat{R} \right)^{-1} \widehat{R} \, \delta \widehat{\widehat{P}} \, \widehat{R} \left(\bar{q} \widehat{I} + \overline{\widehat{Q}} \widehat{R} \right)^{-1} \right\rangle, \\ &- \delta \overline{\widehat{Q}} = \delta \overline{\widehat{P}} \neq \widehat{0} = \delta \sigma^c \widehat{I}; \\ \delta \widehat{\overline{Q}} &= \delta \overline{Q} \, \widehat{I} + \delta \widehat{\overline{Q}}^{\mathrm{f}}, \quad \delta \widehat{\overline{P}} = \delta \overline{P} \, \widehat{I} + \delta \widehat{\overline{P}}^{\mathrm{f}}. \end{split}$$
(10.4)

These relations imply that $\hat{\sigma}^*$, as well as $\bar{\sigma}^s$, increases with the parameter \overline{P} at a fixed $\hat{\overline{P}}^f$ and reaches bounds (7.9) for limiting values $\hat{\overline{P}}^s = \overline{P} \hat{I}$ (6.18) which are the same as the limiting values $\hat{\overline{P}}$ (3.26).

For a two-dimensional medium from (10.1) and (10.2) we have

$$\begin{aligned}
\sigma_{\chi\chi}^{*}(\sigma_{1},\sigma_{2}) &\equiv \sigma_{1}^{*} \equiv \sigma^{*}(\overline{Q}), \\
\sigma_{yy}^{*}(\sigma_{1},\sigma_{2}) &\equiv \sigma_{2}^{*} \equiv \sigma^{*}(\overline{P}); \quad \overline{Q} \equiv \overline{Q}_{11}, \\
\sigma_{yy}^{*}(\sigma_{2},\sigma_{1}) &\equiv \sigma^{*}(\sigma_{2},\sigma_{1};v_{1},v_{2};\overline{P}) \equiv \tilde{\sigma}_{1}^{*} \equiv \tilde{\sigma}^{*}(\overline{Q}), \\
\overline{P} &\equiv 1 - \overline{Q} = \overline{Q}_{22}.
\end{aligned}$$
(10.5)

In this notation Eqn (10.1) takes the form

$$\sigma^*(Q)\tilde{\sigma}^*(Q) = \sigma_1 \sigma_2. \tag{10.6}$$

According to (7.9) the solution σ^{s} from (7.6) satisfies the relation

$$\sigma^{c} \in [0, \infty] \Rightarrow \sigma^{s} \in \left[\langle \rho \rangle^{-1}, \langle \sigma \rangle \right].$$
(10.7)

This means that σ^{s} describes all possible real structures of inhomogeneous media. Therefore, it should comply with the Keller theorem in the form of (10.6).

For a mixture of two isotropic and homogeneous component we have from (7.6)-(7.8) with the use of the notation (1.8) and (1.9):

$$\sigma^{s} = \langle \sigma \rangle - \frac{\overline{Q} D_{\sigma}}{\overline{Q}[\sigma] + \overline{P} \sigma^{c}} = \frac{\overline{Q} \sigma_{1} \sigma_{2} + \overline{P} \sigma^{c} \langle \sigma \rangle}{\overline{Q}[\sigma] + \overline{P} \sigma^{c}} , \qquad (10.8a)$$

$$D_{\sigma} \equiv \left\langle \left(\sigma''\right)^{2} \right\rangle = v_{1}v_{2}(\sigma_{1} - \sigma_{2})^{2},$$

$$[\sigma] \equiv v_{1}\sigma_{2} + v_{2}\sigma_{1}, \qquad \langle \sigma \rangle \equiv v_{1}\sigma_{1} + v_{2}\sigma_{2}, \qquad (10.9a)$$

$$\sigma_a \equiv \sigma(\mathbf{r}_a), \quad \mathbf{r}_a \equiv \mathbf{r} \in V_a, \quad v_a \equiv \frac{V_a}{V}, \quad v_a \in [0, 1].$$
(10.9b)

With definition (10.2) we then have

$$\tilde{\sigma}^{s} = \frac{\overline{P}\sigma_{1}\sigma_{2} + \overline{Q}\tilde{\sigma}^{c}[\sigma]}{\overline{P}\langle\sigma\rangle + \overline{Q}\tilde{\sigma}^{c}}; \quad \sigma_{1} \leftrightarrow \sigma_{2} \Rightarrow [\sigma] \leftrightarrow \langle\sigma\rangle.$$
(10.8b)

Substituting (10.8) into (10.6) we obtain the equation [83]:

$$\sigma^{c}\tilde{\sigma}^{c} = \sigma_{1}\sigma_{2} \tag{10.10}$$

for the σ^{c} and $\tilde{\sigma}^{c}$ parameters of 'reciprocal' media (in the terminology adopted in Ref. [112]).

Let us show that one-dimensional structures, i.e. layered media, satisfy Eqn (10.6). Putting $\overline{P} = 0$ we have from (10.8) and (10.2) that the conduction in the normal direction is

$$\overline{P} = 0, \quad \overline{Q} = 1 \Rightarrow \sigma^* = \frac{\sigma_1 \sigma_2}{[\sigma]} = \langle \rho \rangle^{-1}, \quad \tilde{\sigma}^* = [\sigma].$$
(10.11a)

Similarly, putting $\overline{P} = 1$ we have from (10.8) and (10.2) that the conduction in the direction parallel to layers is

$$\overline{P} = 1, \quad \overline{Q} = 0 \Rightarrow \sigma^* = \langle \sigma \rangle, \quad \tilde{\sigma}^* = \frac{\sigma_1 \sigma_2}{\langle \sigma \rangle} = [\rho]^{-1}.$$
(10.11b)

Thus, 'one-dimensional' RIM complies with the Keller theorem. By comparing (10.11) with (9.18) and (9.19) we conclude that the best bounds $\sigma_{(n)}^{\pm}$ for n = 1 are consistent with (10.6). The case of n = 0 from (9.18) and (9.19) is trivial.

Now we shall consider the generalised Hashin–Shtrikman bounds $\sigma_{(2)}^{\pm}$ (9.18) and (9.19), for which according to (10.8) we write

$$\begin{aligned} \sigma_{(2)}^{+} &= \sigma_2 \frac{Q\sigma_1 + P\langle \sigma \rangle}{\overline{Q}[\sigma] + \overline{P}\sigma_2} , \qquad \tilde{\sigma}_{(2)}^{+} &= \sigma_1 \frac{P\sigma_2 + Q[\sigma]}{\overline{P}\langle \sigma \rangle + \overline{Q}\sigma_1} ; \\ \sigma^{\rm c} &= \sigma^+ \equiv \sigma_2 , \qquad (10.12a) \\ \sigma_{(2)}^{-} &= \sigma_1 \frac{\overline{Q}\sigma_2 + \overline{P}\langle \sigma \rangle}{\overline{Q}[\sigma] + \overline{P}\sigma_1} , \qquad \tilde{\sigma}_{(2)}^{-} &= \sigma_2 \frac{\overline{P}\sigma_1 + \overline{Q}[\sigma]}{\overline{P}\langle \sigma \rangle + \overline{Q}\sigma_2} ; \\ \sigma^{\rm c} &= \sigma^- \equiv \sigma_1 . \qquad (10.12b) \end{aligned}$$

Whence the equation

.

$$\sigma_{(2)}^{\pm}\tilde{\sigma}_{(2)}^{\pm} = \sigma_1\sigma_2 \tag{10.13}$$

follows. It means that in the case of n = 2 the best bounds obey (10.6). For $3\overline{Q} = 1$ the values $\sigma_{(2)}^{\pm}$ describe possible real three-dimensional structures which have been calculated in Ref. [9].

Bounds $\sigma_{(3)}^{\pm}$ (9.18) account for three-particle interactions, therefore a medium (or model) for which they are exact solutions must have three 'degrees of freedom', i.e., it must either be a mixture three components or have an additional parameter describing orientation of nonspherical inclusions. Substitution of the values σ_j^{\pm} and $\tilde{\sigma}_j^{\pm}$ into (10.10) in place of σ^c and $\tilde{\sigma}^c$, respectively, must result, as (10.10), in a relation between *j* and *j*, where *j* is the value of the parameter *j* for the 'reciprocal' medium (see Section 11).

The resultant generalisation (10.6) of the Keller theorem (10.1) extends its applicability to three-dimensional media. The use of the two-dimensional version (10.1) for evaluation of σ^* in three-dimensional media brings about the inequality [115]:

$$\sigma_{xx}^*(\sigma_1, \sigma_2) \sigma_{yy}^*(\sigma_2, \sigma_1) \ge \sigma_1 \sigma_2 . \tag{10.14}$$

This prompts the conclusion that in the three-dimensional case the theorem of the form of (10.1) does not exist.

Among the infinite set of two-dimensional RIM we shall distinguish media the macroscopic conductivity of which is invariant under permutation of components: $\sigma_1 \leftrightarrow \sigma_2$. The requirement that the statistical properties of components be identical [15, 29] restricts very essentially the class of media to be considered (see Section 12). Given the values of σ_1 and σ_2 such media are described by the same value of the macroscopic conductivity, which is equal according to (10.1) and (10.6) to [20, 25]:

$$\sigma^* = \tilde{\sigma}^* = \sqrt{\sigma_1 \sigma_2}$$
, $v_1 = v_2 = \overline{Q} = \overline{P} = \frac{1}{2}$. (10.15)

The unique solution (10.15) describes the conductive properties of two-dimensional homogeneous medium. The macroscopic conductivity of an RIM, the statistical properties of components of which are identical, is equal to (10.15). Using the terminology adopted in Refs [15, 26] we shall term such a RIM a completely symmetric medium.

10.2 Self-consistent solutions

The studies of conductive properties of different macroscopic systems (models) resulted in exact solutions for σ^* called selfconsistent. Often they are loosely called approximations of theory of effective medium [24, 28, 106, 112]. However, this term is normally applied to a homogeneous medium, the properties of which (for example, conduction) are equal to macroscopic properties of different realisations of the ensemble of inhomogeneous media.

The idea of self-consistency was first utilised by Bruggerman [2] in calculation of effective dielectric permittivity of three- and two-dimensional macroisotropic media. In these cases we have $\overline{Q} = 1/3$ and $\overline{Q} = 1/2$ in formulae (10.8), respectively.

By definition [2, 7, 27, 55, 83, 105] a self-consistent solution symbolised by σ^{SCS} is derived from (7.6) when $\sigma^{c} = \sigma^{SCS}$. It is easy to see that σ^{SCS} satisfies the equation

$$\left\langle \left(\bar{q} + \overline{Q}\widehat{R}\right)^{-1} \right\rangle = 0, \quad \sigma^{c} = \sigma^{SCS}.$$
 (10.16)

From this and from (10.8) for a mixture of two homogeneous and isotropic components we obtain the equation:

$$\overline{P}(\sigma^{\text{SCS}})^2 - b\sigma^{\text{SCS}} - \overline{Q}\sigma_1\sigma_2 = 0, \qquad b \equiv \overline{P}\langle\sigma\rangle - \overline{Q}[\sigma],$$
(10.17a)

the solution of which

$$2\overline{P}\sigma^{\rm SCS} = \Delta + b \,, \qquad \Delta^2 \equiv b^2 + 4\overline{P}\,\overline{Q}\sigma_1\sigma_2 \tag{10.18a}$$

determines the principal values of σ^{SCS} for different $\overline{P} = 1 - \overline{Q}$. Solution (10.18a) describes macroanisotropic self-consistent media and is the most general among the solutions of this kind. Anisotropy of macroscopic conduction occurs either because of a deviation of the shapes of grains from a sphere (in the statistical sense) or as a result of anisotropy of spatial distribution of grains [13, 16, 21–23, 37, 38, 83, 104, 105, 111, 112]. Note also that solution (10.18a) accounts implicitly for multiparticle interactions. Besides it is symmetric relative to characteristics of components (conductivities and concentrations). As a consequence, a medium described by the conductivity σ^{SCS} falls into the class of symmetric medium. Following [29] we shall term a medium symmetric if its macroscopic properties are invariant under inversion of components:

$$(\sigma_1, v_1) \leftrightarrow (\sigma_2, v_2) \,. \tag{10.19}$$

The inversion is understood to be spatial permutation of components $\sigma_1 \leftrightarrow \sigma_2$ with simultaneous exchange of their concentrations $v_1 \leftrightarrow v_2$. The invariance imposes severe restrictions on statistical properties of an RIM [15, 29, 112].

Now we shall consider a self-consistent solution for a 'reciprocal' medium. Instead of (10.17a) and (10.18a) we, respectively, write:

$$\overline{Q}(\tilde{\sigma}^{\rm SCS})^2 + b\tilde{\sigma}^{\rm SCS} - \overline{P}\sigma_1\sigma_2 = 0, \qquad (10.176)$$

$$2\overline{Q}\tilde{\sigma}^{\rm SCS} = \Delta - b; \qquad \tilde{b} = -b, \qquad \widetilde{\Delta} = \Delta. \qquad (10.18b)$$

By multiplying the left-hand and right-hand sides of (10.18a) and (10.18b) we have

$$\sigma^{\text{SCS}}\tilde{\sigma}^{\text{SCS}} = \sigma_1 \sigma_2 \,. \tag{10.20}$$

Like σ^{SCS} , the solution $\tilde{\sigma}^{SCS}$ is symmetric. Below we shall show that the media described by σ^{SCS} and $\tilde{\sigma}^{SCS}$ are, in a sense, limiting in the class of symmetric media (see Section 12). For each $\overline{P} = 1 - \overline{Q}$ there is only one completely symmetric solution of the form

$$v_1 = v_2 \Rightarrow \sigma^{\text{SCS}}(\sigma_1, \sigma_2) = \sigma^{\text{SCS}}(\sigma_2, \sigma_1) \equiv \sigma^{\text{CSM}}$$
 (10.21)

among self-consistent solutions such that it is invariant under permutation $\sigma_1 \leftrightarrow \sigma_2$. Media of this type have been considered by Frisch [15], Dykhne [20], Mendelson [25], Schulgasser [26], Fokin [29], Shvidler [39], and Balagurov [112]. With regard for (10.21) we find from (10.17a):

$$b = (\overline{P} - \overline{Q})\langle\sigma\rangle, \quad \langle\sigma\rangle = [\sigma] = \frac{\sigma_1 + \sigma_2}{2}.$$
 (10.22)

Substituting (10.22) into (10.18) we obtain the values

$$4\sigma^{\text{CSM}} = \left(\langle \sigma \rangle^2 + 8\sigma_1 \sigma_2\right)^{1/2} + \langle \sigma \rangle, \qquad v_2 = \frac{1}{2},$$
$$2\tilde{\sigma}^{\text{CSM}} = \left(\langle \sigma \rangle^2 + 8\sigma_1 \sigma_2\right)^{1/2} - \langle \sigma \rangle, \qquad \overline{P} = \frac{2}{3} \qquad (10.23a)$$

in the case of three-dimensional macroisotropic media and

$$\sigma^{\text{CSM}} = \tilde{\sigma}^{\text{CSM}} = \sqrt{\sigma_1 \sigma_2} \equiv \sigma^{\text{DM}}, \quad v_2 = \overline{P} = \frac{1}{2} \quad (10.23 \text{ b})$$

in the case of two-dimensional macroisotropic media, respectively.

10.3 New solutions in the Dykhne-Mendelson form

The examination of expressions (10.18) shows that the solution in the Dykhne–Mendelson form (10.23b), valid for completely symmetric media, is realised in a certain range of concentrations for any media from the class of symmetric media.

It can be shown that Eqns (10.23b) take place provided that

$$2\overline{P}\,\overline{Q}(\sigma^{\rm SCS} - \tilde{\sigma}^{\rm SCS}) = (\overline{Q} - \overline{P}\,)\Delta + b = 0\,, \qquad (10.24)$$

whence after simple manipulations we find

$$b = (\overline{P} - \overline{Q})\Delta, \quad \Delta = \sqrt{\sigma_1 \sigma_2}.$$
 (10.25)

If follows from (10.25) and the definition of (10.17a) that the concentrations

$$v_{2} = v_{0} \equiv \overline{P}v_{m} + \overline{Q}(1 - v_{m}) = \frac{\overline{Q} + \overline{P}\sqrt{x}}{1 + \sqrt{x}},$$

$$v_{0} \in [v_{m}, 1 - v_{m}], \qquad (10.26)$$

$$v_0 \in [v_{\mathrm{m}}, 1 - v_{\mathrm{m}}], \qquad (10.26)$$

$$v_{\rm m} \equiv \frac{\sqrt{x}}{1+\sqrt{x}}, \quad \frac{\sigma_1}{\sigma_2} \equiv x \in [0,1], \qquad (10.27)$$

and arbitrary values of \overline{P} and $\overline{Q} = 1 - \overline{P}$ satisfy the equations

$$\sigma^0 \equiv \sigma^{\text{SCS}} = \tilde{\sigma}^{\text{SCS}} = \sqrt{\sigma_1 \sigma_2} , \quad \overline{Q} \in [0, 1] . \quad (10.28)$$

Putting $\overline{Q} \equiv \overline{Q}_{11}$, $\overline{P} \equiv 1 - \overline{Q}_{11} = \overline{Q}_{22}$ and invoking the notation of (10.2) and (10.5) we then have

$$\sigma_{xx}^{\text{SCS}}(\sigma_1, \sigma_2) = \sigma_{yy}^{\text{SCS}}(\sigma_2, \sigma_1) = \sigma^0, \quad v_2 = v_0 = \frac{\overline{Q} + \overline{P}\sqrt{x}}{1 + \sqrt{x}}$$
(10.29a)

$$\sigma_{xx}^{SCS}(\sigma_2, \sigma_1) = \sigma_{yy}^{SCS}(\sigma_1, \sigma_2) = \sigma^0,$$

$$v_2 = v_0' \equiv \frac{\overline{Q} + \overline{P}/\sqrt{x}}{1 + 1/\sqrt{x}} = 1 - v_0,$$

$$\sigma_{xx}^{SCS}(\sigma_2, \sigma_1) \leq \sigma_{xx}^{SCS}(\sigma_1, \sigma_2) = \sigma^0$$

$$= \sigma_{yy}^{SCS}(\sigma_2, \sigma_1) \leq \sigma_{yy}^{SCS}(\sigma_1, \sigma_2),$$
(10.29b)

0

SCS /

$$v_2 = v_0 \gtrless \frac{1}{2} \gtrless \overline{P} \tag{10.30}$$

for a two-dimensional symmetric medium. It is easy to see that solutions (10.29a) and (10.29b) describe RIM for which (10.19) is valid. For $\overline{P} = \overline{Q}$ formulae (10.29) go into (10.23b).

For further analysis it is convenient to introduce the functions

$$R_{(n)}^{\pm} \equiv \frac{\sigma_{(n)}^{\pm} - \sigma_1}{\sigma_2 - \sigma_1} , \qquad \widetilde{R}_{(n)}^{\pm} \equiv \frac{\widetilde{\sigma}_{(n)}^{\pm} - \sigma_1}{\sigma_2 - \sigma_1} ,$$
$$R^0 \equiv \frac{\sigma^0 - \sigma_1}{\sigma_2 - \sigma_1} , \qquad R^{\text{DM}} \equiv \frac{\sigma^{\text{DM}} - \sigma_1}{\sigma_2 - \sigma_1} , \qquad (10.31)$$

where we use the notation of (9.18), (10.23b), and (10.28). Whence it follows

$$\begin{aligned} R_{(0)}^{-} &= 0 , \qquad R_{(0)}^{+} = 1 ; \qquad R_{(1)}^{-} = \frac{v_2 x}{v_1 + v_2 x} , \qquad R_{(1)}^{+} = v_2 , \\ \widetilde{R}_{(1)}^{-} &= \frac{v_1 x}{v_1 x + v_2} , \qquad \widetilde{R}_{(1)}^{+} = v_1 ; \qquad R^0 = \frac{\sqrt{x}}{1 + \sqrt{x}} = v_{\rm m} . \end{aligned}$$

$$(10.32)$$

Fig. 4 shows the plots of the functions from (10.31)–(10.33) when 5x = 1. Regions 1 and 2 are the ranges of the functions

$$\widetilde{R}^{\text{SCS}} \equiv \frac{\widetilde{\sigma}^{\text{SCS}} - \sigma_1}{\sigma_2 - \sigma_1} , \qquad R^{\text{SCS}} \equiv \frac{\sigma^{\text{SCS}} - \sigma_1}{\sigma_2 - \sigma_1}$$
(10.33)

respectively. The intersection of these sets is hatched. The horizontal line $(\overline{Q} \in [0, 1])$ in the hatched region is the set of values of \mathbb{R}^0 . The points *a* and *b* in this line represent solutions



Figure 4.

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(10.29a) and (10.29b) related by the transform $(\sigma_1, 1 - v_0) \leftrightarrow (\sigma_2, v_0)$; the DM point represents the Dykhne–Mendelson solution (10.23b).

If we remove the limitation (5x = 1) on the parameter x and vary it within the range $x \in [0, 1]$, then the hatched region extends to fill the triangle the sides of which are the diagonals of the square and the abscissa axis. The height of the triangle to the hypotenuse represents the range of the function R^{DM} (10.31).

The hatched region of the range $x \in [1, \infty]$, for which $\sigma_1 \ge \sigma_2$, is presented by the upper triangle, which is the image of the lower triangle after inversion relative to the centre of the square.

Thus, the set of all possible values of R^0 , for which solutions of the form (10.28) and (10.29) exist, occupies a half of the square (see Fig. 4). The second half of the square is occupied by the range of the function *R* obtained for solutions for which Eqn (10.28) is not satisfied. Finally, the range R^{DM} is presented by the unit segment of a vertical line going through the centre of the square.

Thus, we have shown that a solution of the form of (10.23b) is realised for any symmetric medium when the concentration is $v_2 = v_0$ (it is determined by the geometric parameter $\overline{P} = 1 - \overline{Q}$ and the ratio x of conductivities of the components). The form of functions (10.31)–(10.33) does not depend on the dimension of the RIM. Therefore, Fig. 4 is also applied in the three-dimensional case with an appropriate interpretation of the regions, curves, and points.

11. Structural parameters of inhomogeneous media

In solutions of different problems in theory of inhomogeneous media it frequently proves useful to introduce parameters to present the available statistical information about an RIM. These parameters, called structural or statistical, give an idea of the spatial distribution of inhomogeneities, i.e., the internal geometry of the medium.

Here these parameters are considered by the example of the simplest RIM which is a mixture of two homogeneous isotropic components. Following [15, 29, 116] we introduce a scalar indicator function f of the form

$$f(\mathbf{r}) = \begin{cases} 0, & \mathbf{r} \in V_1, \\ 1, & \mathbf{r} \in V_2, \end{cases} \quad V_1 \cup V_2 = V.$$
(11.1)

The region V_a which the *a*th component occupies is symbolised by the same letter as the volume of this region.

In the notation of (1.9), (4.18), (10.9), and (11.1) the random scalar field $\sigma(\mathbf{r})$, its average value $\langle \sigma \rangle$, and its fluctuation σ'' take the form

$$\sigma = \sigma_1 + (\sigma_2 - \sigma_1)f; \quad \sigma = \sigma(\mathbf{r}), \quad f = f(\mathbf{r}), \quad \mathbf{r} \in V,$$
(11.2a)
$$\langle \sigma \rangle = \sigma_1 + (\sigma_2 - \sigma_1)\langle f \rangle, \quad \sigma'' = (\sigma_2 - \sigma_1)f'',$$

$$f'' = f - \langle f \rangle.$$
(11.2b)

We assume that the scalar random field $f(\mathbf{r})$ is statistically uniform and ergodic [8, 14, 30, 31, 37]. Quantities with such properties are often called self-averaging [20, 95–98]. Then we can write by virtue of (3.7)

$$\langle f \rangle = v_2 \equiv v, \quad f''(\mathbf{r}) = \begin{cases} (-v_2), & \mathbf{r} \in V_1, \\ v_1, & \mathbf{r} \in V_2, \end{cases}$$
(11.3)

where v has the sense of geometrical probability that the point **r** falls into the region V_2 [15]. The equality of $\langle f \rangle$ and the volume concentration v_2 imparts an extensive meaning to this parameter. For the second-order central moment function we have [21, 30]

$$\langle f''(\mathbf{r}_1) f''(\mathbf{r}_2) \rangle = \Phi(\mathbf{r}) , \quad \mathbf{r} \equiv \mathbf{r}_1 - \mathbf{r}_2 ;$$

$$\Phi(0) \equiv D_f = v_1 v_2 .$$
(11.4)

Along with v we shall consider the tensor parameters \overline{P} and \overline{Q} [as in (10,5), (10.17), and (10.18) we shall also use \overline{P} and \overline{Q} to denote the principal values of these tensors], which according to the definitions of (6.17) take the form

$$\overline{P} \equiv \left(\overline{f}'', \widehat{\overline{P}}\overline{f}''\right), \quad \overline{Q} \equiv \left(\overline{f}'', \overline{\overline{Q}}\overline{f}''\right);$$
$$\overline{f} \equiv f D_f^{-1/2}, \quad D_f = \|f''\|^2$$
(11.5)

for the field $\bar{\sigma}$ (11.2a) [21, 37]. In the case of an ellipsoidal inclusion the depolarisation tensor \overline{Q} has been calculated by Osborn [3]. In contrast to v the statistical parameters \overline{P} and \overline{Q} have intensive meaning. As is seen from Section 9, they play an important part in determination of the numerical values and symmetry properties of the tensors $\sigma_{(n)}^{\pm}$ (where $n \ge 2$). If the effective (ensemble average) grain of inhomogeneity of an RIM can be presented by a three-axial ellipsoid, the problem is reduced to determination of three principal values of the tensors \overline{Q} and $\sigma_{(n)}^{\pm}$, respectively.

In order to calculate the bounds $\sigma_{(3)}^{\pm}$ (see Section 9) the tensor parameter *j* should be introduced to account for the statistical information that three-point probabilities present [these probabilities are given with the use of moments of the random field $f(\mathbf{r})$] [15, 29]. According to (9.10) and with regard for (11.5) and (6.19) the parameter *j* is specified by the relations:

$$j \equiv (g, fg); \quad g \equiv G \|G\|^{-1}, \quad \|g\| = 1; \quad j \in [0, 1],$$

$$G \equiv \left(\overline{\widehat{Q}} - \overline{Q}\widehat{I}\right) f'' = \left(\overline{P}\widehat{I} - \overline{\widehat{P}}\right) f'';$$

$$\|G\|^2 = (f'', \overline{P}\overline{Q}f'') \ge 0.$$
(11.6)

Note that in view of (6.22) the operators \overline{P} and \overline{Q} and the tensors \overline{P} and \overline{Q} are independent of the material characteristics of the initial comparison medium. Therefore, the statistical parameter *j* (of extensive meaning, as *v*) is also independent of σ^{c} (or ρ^{c}) and its properties reflect those features of spatial distribution of inhomogeneities, which are described in terms of three-point probabilities.

Using (11.6) for σ^{j} and ρ^{j} we get from (9.10) [37, 83]:

$$\Gamma^{j} = \Gamma_{1} + (\Gamma_{2} - \Gamma_{1})j, \quad j \in [0, 1]; \quad \Gamma \equiv \sigma, \rho. \quad (11.7)$$

The relation between *j* and the macrostructure of the RIM is also exhibited by the dependence of this parameter on \overline{P} (or on \overline{Q}). Generally, the principal axis of the tensors *j* (11.6) and \overline{P} and \overline{Q} (11.5) are not coincident, therefore, $j = j(\overline{Q})$ should be understood as a diagonal component of the tensor *j*, which corresponds to a given principal value of the tensor \overline{Q} .

It follows from (11.7) for σ_i^{\pm} (9.13) used in (9.12) that

$$\sigma_j^+ \equiv \sigma^j = \langle \sigma \rangle_j, \qquad \sigma_j^- \equiv (\rho^j)^{-1} = \frac{\sigma_1 \sigma_2}{[\sigma]_j},$$
$$\langle \sigma \rangle_j \equiv \sigma_1 + (\sigma_2 - \sigma_1)j, \quad [\sigma]_j \equiv \sigma_2 + (\sigma_1 - \sigma_2)j; \quad j \in [0, 1].$$
(11.8)

The parameters σ_j^{\pm} comply with relations (9.17) responsible for consistency between the next bounds $\sigma_{(3)}^{\pm}$ with the previous ones $\sigma_{(2)}^{\pm}$.

According to (9.18), (10.8), and (11.8) in the approximation of piecewise uniform 'polarised' fields the best bounds (9.12) take the form

$$\sigma_{(3)}^{+} = \frac{\overline{Q}\sigma_{1}\sigma_{2} + \overline{P}\langle\sigma\rangle_{j}}{\overline{Q}[\sigma] + \overline{P}\langle\sigma\rangle_{j}}, \qquad \sigma_{(3)}^{-} = \sigma_{1}\sigma_{2}\frac{\overline{Q}[\sigma]_{j} + \overline{P}\langle\sigma\rangle}{\overline{Q}[\sigma]_{j} + \overline{P}\sigma_{1}\sigma_{2}},$$
(11.9a)

and, similarly, for the 'reciprocal' media they are:

$$\tilde{\sigma}_{(3)}^{+} = \frac{\overline{P}\sigma_{1}\sigma_{2} + \overline{Q}[\sigma][\sigma]_{\tilde{j}}}{\overline{P}\langle\sigma\rangle + \overline{Q}[\sigma]_{\tilde{j}}}, \qquad \tilde{\sigma}_{(3)}^{-} = \sigma_{1}\sigma_{2}\frac{\overline{P}\langle\sigma\rangle_{\tilde{j}} + \overline{Q}[\sigma]}{\overline{P}\langle\sigma\rangle\langle\sigma\rangle_{\tilde{j}} + \overline{Q}\sigma_{1}\sigma_{2}},$$
(11.9b)

where \tilde{j} is the value of the parameter *j* for the 'reciprocal' medium.

For $\sigma_{(3)}^{\pm}$ and $\tilde{\sigma}_{(3)}^{\pm}$ to comply with theorem (10.6), the parameters σ_j^{\pm} and $\tilde{\sigma}_j^{\pm}$ should satisfy the equation (10.10), by virtue of which we have

$$\sigma_{(3)}^+\tilde{\sigma}_{(3)}^+ = \sigma_1\sigma_2 \Rightarrow \sigma_j^+\tilde{\sigma}_j^+ = \sigma_1\sigma_2 \Rightarrow j\tilde{j} = \frac{j\sigma_2 - j\sigma_1}{\sigma_2 - \sigma_1}, \quad (11.10a)$$

$$\sigma_{(3)}^{-}\tilde{\sigma}_{(3)}^{-} = \sigma_{1}\sigma_{2} \Rightarrow \sigma_{j}^{-}\tilde{\sigma}_{j}^{-} = \sigma_{1}\sigma_{2} \Rightarrow j\tilde{j} = \frac{j\sigma_{1} - \tilde{j}\sigma_{2}}{\sigma_{1} - \sigma_{2}} \quad (11.10b)$$

for the parameters j and \tilde{j} . These two relations can be fulfilled provided that

$$j = \tilde{j} = j^{\pm}, \quad j \in [0, 1].$$
 (11.12)

This condition converts the bounds $\sigma_{(3)}^{\pm}$ into $\sigma_{(2)}^{+}$ $(j = j^{+} = 1)$ and $\sigma_{(2)}^{-}$ $(j = j^{-} = 0)$ according to (9.17).

Thus, a medium with three 'degrees of freedom' from Section 10 does not obey the relations following from the generalised Keller theorem. This fact can be explained by the absence of an exact solution or by the two-component origin of the Keller theorem.

12. Asymmetric and symmetric media

To calculate *j* directly by means of formulae (11.6) the thirdorder central moment function $\Psi(\mathbf{R}_3)$ should be given [15, 29]:

$$\Psi(\mathbf{R}_3) \equiv \left\langle f''(\mathbf{r}_1) f''(\mathbf{r}_2) f''(\mathbf{r}_3) \right\rangle$$
$$= v_1 v_2 \left[v_1 \varphi_1(\mathbf{R}_3) - v_2 \varphi_2(\mathbf{R}_3) \right], \qquad (12.1)$$

where the functions φ_a (a = 1, 2) are expressed in terms of the relevant probabilities of the random field $f(\mathbf{r})$. Considering (12.1), (11.4) and the auxiliary equation

$$\langle f''(\mathbf{r}_1)f(\mathbf{r}_3)f''(\mathbf{r}_2)\rangle = v_2\Phi(\mathbf{r}) + \Psi(\mathbf{R}_3), \quad \langle f(\mathbf{r}_3)\rangle = v_2,$$

we present *j* in the form

$$j = v_1 j_1 + v_2 (1 - j_2), \qquad \overline{S}_a - \overline{S} \equiv j_a \in [0, 1]; \qquad a = 1, 2;$$
$$\overline{SP} = \overline{Q}. \qquad (12.2)$$

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Here S are defined according to (7.7) and S_a are geometrical parameters which are derived from (11.6) with the aid of the corresponding terms in (12.1). It is easy to see that the range of the parameter j_a is coincident with (11.6). This fact follows also from the asymptotic expansion method proposed by Miller [19]. Unfortunately, his own conclusions for asymmetric media are erroneous). In terms of the parameter *j* his results take the form

$$j^{\mathrm{M}} = v_1^2 j_1^{\mathrm{M}} + v_2 (1 - v_2 j_2^{\mathrm{M}}), \quad j_a^{\mathrm{M}} \in [0, 1].$$
 (12.3)

Since the regions of j_a (12.2) and $j_a^{\rm M}$ (12.3) are the same, the bounds for the parameters j and $j^{\rm M}$, on which the bounds $\sigma_{(3)}^{\pm}$ depend, are related by:

$$j^{-} \leq j_{M}^{-} \leq j_{M}^{+} \leq j^{+}; \quad j \in [0, 1],$$

$$j^{M} \in [v_{1}v_{2}, 1 - v_{1}v_{2}].$$
(12.4)

A constriction of the range of the structural parameter j^{M} in Miller's model results in contraction of the corresponding bounds σ_{M}^{\pm} in comparison with $\sigma_{(3)}^{\pm}$. Moreover, nonzero (in a general case) gaps Δ^{\pm} of the form [37]

$$\Delta^{-} \equiv \frac{\sigma_{\overline{M}}^{-} - \sigma_{\overline{(3)}}^{-}}{\sigma_{\overline{(3)}}^{+} - \sigma_{\overline{(3)}}^{-}}, \qquad \Delta^{+} \equiv \frac{\sigma_{\overline{(3)}}^{+} - \sigma_{\overline{M}}^{+}}{\sigma_{\overline{(3)}}^{+} - \sigma_{\overline{(3)}}^{-}}, \qquad (12.5)$$

appear and, as a result, σ_{M}^{\pm} become inconsistent with $\sigma_{(2)}^{\pm}$ in the sense of limit (9.17). Clearly, the structures that the parameter *j* describes when it belongs to the half-open intervals

$$j \in [j^-, j_{\rm M}^-] \cup (j_{\rm M}^+, j^+],$$
 (12.6)

are not covered by Miller's model of an asymmetric medium. Two-dimensional version of this model is realised in Ref. [22]. The inconsistencies in the model of an asymmetric medium have been revealed by Brown in Ref. [117] from the point of view of probability theory.

Now we turn to consideration of symmetric media (see Section 10), the macroscopic properties of which are invariant under inversion of components (10.19). The requirement of symmetry restricts the class of functions to be used as central moments of the random field f. In particular, instead of (12.1) we have [29]:

$$\Psi^{\rm SM}(\mathbf{R}_3) = v_1 v_2 (v_1 - v_2) \varphi(\mathbf{R}_3) \,. \tag{12.7}$$

In view of (12.7) we rewrite (12.2) in the form:

$$j^{\text{SM}} = v_2 + \bar{j}(v_1 - v_2), \quad \bar{j} \in [0, 1].$$
 (12.8)

Whence it follows that

$$j^{\text{SM}} \in [j_{\text{SM}}^{-}, j_{\text{SM}}^{+}]; \quad j_{\text{SM}}^{-} \equiv \min(v_1, v_2),$$

 $j_{\text{SM}}^{+} = \max(v_1, v_2).$ (12.9)

By setting $j_1^{M} = j_2^{M} \equiv \overline{j}^{M}$ in (12.3) we get:

$$j_{\rm M}^{\rm SM} = v_2 + \bar{j}^{\rm M}(v_1 - v_2), \quad v_1 + v_2 = 1,$$
 (12.10)

as in the case of (12.8). Thus, Miller's model yields correct results for a symmetric medium only in the case of two

components. In addition, the quadratic dependence is not present in (12.3) because of peculiar features of two-component medium. The application of Miller's model of a symmetric medium in the case of three components leads to the same difficulties as for the model of an asymmetric medium.

Let us consider several special cases. It follows from (12.8) for $v_1 = v_2$ that

$$j^{\text{CSM}} = \frac{1}{2},$$
 (12.11)

i.e. only one macrostructure of an homogeneous medium meets the Dykhne complete statistical symmetry requirement [20] (see Section 10.1).

We shall find the range j^{SCS} of the parameter j^{SM} , for which solutions are self-consistent (see Section 10.2). Using the definition (10.16) and the bounds $\sigma_{(1)}^{\pm}$ and σ^{\pm} we write from (6.10), (6.14)–(6.16):

$$\sigma_{1} \equiv \sigma^{-} \leqslant \langle \rho \rangle^{-1} \leqslant \sigma^{\text{SCS}} = \sigma^{\text{c}} \leqslant \langle \sigma \rangle \leqslant \sigma^{+} \equiv \sigma_{2} \,. \, (12.12)$$

Whence it is clear that the range σ^{c} , for which self-consistent solutions exist, is bounded by the values $\langle \rho \rangle^{-1}$ and $\langle \sigma \rangle$. Once these values are presented in the form of (11.7)

$$\langle \Gamma \rangle = \Gamma_1 + (\Gamma_2 - \Gamma_1) j^{\text{SCS}}; \quad \Gamma = \sigma, \rho, \qquad (12.13)$$

which is used to calculate the bounds $\sigma_{(3)}^{\pm}$ with account for three-particle interactions, they yield

$$j^{\text{SCS}} = v_2 \tag{12.14a}$$

for the media described by σ^{SCS} . For 'reciprocal' media (10.18b) instead of (12.14a) we have

$$\tilde{j}^{\text{SCS}} = v_1 = 1 - j^{\text{SCS}}$$
. (12.14b)

Relations (12.14b) follow from a more general equation

$$\tilde{j} = 1 - j, \tag{12.15}$$

which is obtained in the case of a mixture of two isotropic components when

$$\tilde{f} = 1 - f, \quad \tilde{f}'' = -f''.$$
 (12.16)

Note that by virtue of (12.6), (12.9), and (9.17) the generalised Hashin–Shtrikman solutions (10.12), to which the values j = 0 and j = 1 correspond, are not feasible in the classes of asymmetric Miller media (12.3) and symmetric media (12.8).

Figure 5 shows the range of the parameter j = j(v) for all the possible structures of RIM. The diagonals of the square indicate the range of j^{SM} for all the possible structures from the class of symmetric media. The hatched regions bounded by parabolas and horizontal sides of the square represent the structures which are not feasible within the scope of Miller's model of an asymmetric media. The solid diagonal of the square presents the range of j^{SCS} for the structures from the class of self-consistent media. The dashed diagonal of the square presents the range of \tilde{j}^{SCS} for the structures from the class of 'reciprocal' self-consistent media. The upper and lower sides of the square are two ranges of the parameter $j_{\text{GHS}} = 0$ and $j_{\text{GHS}}^+ = 1$ for the structures, which are feasible in the class of the generalised Hashin – Shtrikman solutions and leading, respectively, to lower $\sigma_{(2)}$ and upper $\sigma_{(2)}^+$ bounds. The point of intersection of diagonals $j^{\text{CSM}} = 1/2$ describes all possible implementations of RIM, which are macroscopically equivalent to Dykhne's completely symmetric media.



Figure 5.

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