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Generalized effective-field approximation for inhomogeneous medium with inclusions in multilayered shells

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An approach is proposed for calculating effective physical characteristics of a inhomogeneous medium with several levels of nesting of its microstructure — the generalized effective-field approximation. With the help of this approach, an expression is obtained for an effective permittivity tensor of an inhomogeneous medium with ellipsoidal inclusions in a multilayered shell, the boundaries of all layers of which are ellipsoids. The proposed approach allows to take into account probabilistic distributions of orientations and forms of inclusions, as well as the presence of several types of inclusions. Two cases of matrix composites are considered: 1) with spherical isotropic inclusions with a multilayered shell; 2) with ellipsoidal anisotropic inclusions with a multilayered shell. For an inhomogeneous medium with homogeneous inclusions, this approximation is shown to produce the same result as the generalized singular approximation.

Keywords: inhomogeneous medium, composite, matrix, inclusion, multilayered shell, generalized effective-field approximation, generalized singular approximation, effective permittivity.

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Introduction

A considerable part of heterogeneous materials is characterized by their nested structure, i. e. that mutual arrangement of homogeneous components, when the particles of some of them are completely immersed in the areas filled with other components. In particular, polycrystals have a structure with one level of nesting because the crystalline components present in the material in the form of crystallites are separated from each other by intergranular space, actually constituting a separate component in the material with a different, with respect to the crystal, structural state of atoms [1]. Also, structures with one level of nesting are matrix-type composites with homogeneous inclusions. In this case, a continuous component, called the matrix, completely surrounds the particles of other components (inclusions) [2,3].

If the inclusions in the composite are heterogeneous, for example, represent a homogeneous core in a single- or multilayer shell, then the material structure has several levels of nesting. The problem of predicting the properties of such materials is, on the one hand, more difficult compared to the similar problem for materials with a single nesting level, but, on the other hand, the use of heterogeneous inclusions provides an opportunity to improve the desired characteristics of the materials. The presence of the shell can significantly affect the physical properties of the individual inclusions and, consequently, the corresponding physical properties of the composite itself. For example, inclusions with dielectric core and metal shell have additional plasmon resonances compared to solid metal particles, and their frequency position depends both on the material and on the shape of the shell, in particular, on its thickness [4–8].This provides additional opportunities to control the frequency location of the plasmon resonances, and hence the properties of the matrix composite containing such inclusions.

However, even describing the properties of real matrix composites with homogeneous inclusions may require a model with more than one level of nesting. As studies [9–11] show, the contact between inclusions and matrix in matrix composites as well as between different inclusions is not perfect. In particularly, in the presence of heat flow, there is a temperature jump at the boundary between the different components of the heterogeneous medium, which indicates the presence of contact thermal resistance at the boundary of the components. To account for contact thermal resistance, a model of a composite with coated inclusions can be used. In this case, the presence of contact thermal resistance is modeled by a single-layer inclusion shell with specially selected thickness and thermal conductivity [12]. If a composite with heterogeneous inclusions consisting of a core with a single- or multilayer shell is considered, then to account for the non-ideal contact between the inclusions and the matrix, an additional layer in the shell should be introduced that will simulate the contact resistance.

The presence in composites of impurities associated with the peculiarities of technological processes of production also leads to the complication of modeles for calculating the characteristics of these materials. For example, a syntactic foam based on an organosilicon binder and glass microspheres contains a certain amount of water, which as a result worsens the dielectric characteristics of the material [13]. Part of the water contained in the composite may be present in the form of continuous films on the surface of the microspheres, that requires the introduction of an additional layer of inclusion shell to account for their influence on the characteristics of the material.

Thus, the development of methods for predicting the effective characteristics of heterogeneous materials having a microstructure with several nesting levels, i. e. containing inclusions with a multilayer shell, seems relevant. To date, there are a number of works devoted to the prediction of the effective properties of heterogeneous media with inclusions in the shell. For example, in [14] a generalization of the Maxwell–Garnett approximation to a matrix medium with single-type spherical inclusions in a single-layer spherical shell has been proposed. In [15], a general scheme of generalization of the effective medium approximation to the cases of media with heterogeneous inclusions is given and an example of a medium with spherical inclusions in a single-layer shell is considered. In [16] bilateral variational estimates of the effective permittivity of a spheroplastic with spherical inclusions in a single-layer shell were obtained. In [17] generalizations of the Maxwell–Garnett approximation to matrix composites with spherical inclusions with a multilayer shell are obtained. A generalized effective-field approximation field for calculating the effective characteristics of heterogeneous media with ellipsoidal inclusions with a single-layer shell is proposed in [18]. Special mention should be made of the work [19], which proposed an approach for calculating the effective thermal conductivity of a composite with unidirectional ellipsoidal inclusions in a multilayer shell.

In the present paper, the generalized effective-field approximation [18] is extended to the case of heterogeneous media with inclusions with a homogeneous core and a multilayer shell with homogeneous layers, with the boundaries of the inclusion cores and shells assumed ellipsoidal. The proposed approach has a high degree of generality and can be applied both to multicomponent composites and to polycrystals, it is also able to naturally take into account the probability distribution of orientations and shapes of inclusions. It is shown that in the absence of a shell for inclusions, this approach gives the same result as the generalized singular approximation [20]. We consider special cases of a matrix composite with single-type isotropic spherical inclusions with a multilayer shell, as well as a matrix composite with an isotropic matrix and anisotropic ellipsoidal inclusions with a multilayer shell.

1. Problem statement. Formalism using the comparison body and Green's function method. Effective field

Consider a sample of volume V with boundary S of a statistically homogeneous heterogeneous medium consisting of inclusions of a nested structure; let N — the number of all inclusions in the sample. A particular inclusion with number k is considered to consist of a homogeneous core $V_n^{(k)}$, which is surrounded by a shell having homogeneous layers $V_{n-1}^{(k)}, \ldots, V_1^{(k)}$, $k = \overline{1, N}$, where $V_{n-1}^{(k)}$ — the nearest layer to the core, $V_1^{(k)}$ — the outermost layer of the kth inclusion shell. The maximum number of homogeneous areas that make up a particular medium inclusion will be assumed to be n. If the inclusion contains fewer homogeneous areas, we will assume that the volumes of the missing areas are zero. The area occupied by all kth inclusion is denoted as $V^{(k)}$:

$$V^{(k)} = \bigcup_{i=1}^{n} V_i^{(k)}, \quad k = \overline{1, N}.$$

Let us also introduce the notation $\tilde{V}_j^{(k)}$ for the area consisting of the layer $V_j^{(k)}$ and all more inner layers of the kth inclusion shell, including the core:

$$\tilde{V}_j^{(k)} = \bigcup_{i=j}^{n} V_i^{(k)}, \quad k = \overline{1, N}. \tag{1}$$

The volumes of all areas will be denoted in the same way as the areas themselves. The dielectric characteristics of the core and each shell layer of each inclusion will be considered known, the tensors of permittivity of areas $V_i^{(k)}$ will be denoted as $\boldsymbol{\varepsilon}_i^{(k)}$, $i = \overline{1, n}$; $k = \overline{1, N}$. Let us also assume that there are no free charges and double electric layers in the medium.

Let's assume that a constant electric field of strength $\mathbf{E}_0$ be applied to the boundary of this sample of heterogeneous medium. The permittivity tensor of a given medium $\boldsymbol{\varepsilon}(\mathbf{r})$ is a random piecewise constant function of coordinates:

$$\boldsymbol{\varepsilon}(\mathbf{r}) = \boldsymbol{\varepsilon}_i^{(k)}, \ \mathbf{r} \in V_i^{(k)}, \ i = \overline{1, n}; \quad k = \overline{1, N}.$$

The $\boldsymbol{\varepsilon}^*$ tensor of the effective permittivity of a sample of a given medium is defined as an operator relating the average values of the vectors of electric induction and electric field strength over the sample volume:

$$\langle \mathbf{D} \rangle = \boldsymbol{\varepsilon}^* \langle \mathbf{E} \rangle. \tag{2}$$

To calculate $\boldsymbol{\varepsilon}^*$, consider the boundary problem for the scalar electric potential $\varphi(\mathbf{r})$ in this medium ($\mathbf{E} = -\nabla\varphi$):

$$\nabla \cdot \boldsymbol{\varepsilon}(\mathbf{r}) \nabla \varphi(\mathbf{r}) = 0, \quad \varphi\big|_S = -(\mathbf{E}_0 \cdot \mathbf{r}). \tag{3}$$

To solve the problem (3), it is reasonable to consider a similar problem for a homogeneous comparison body having the same dimensions and shape as the sample of heterogeneous medium [18,21]:

$$\nabla \cdot \boldsymbol{\varepsilon}^c \, \nabla \varphi^c(\mathbf{r}) = 0, \quad \varphi^c\big|_S = -(\mathbf{E}_0 \cdot \mathbf{r}), \tag{4}$$

where the index „c" indicates the values related to the comparison body. Let us introduce notations for the differences of the quantities related to problems (3) and (4):

$$\varphi'(\mathbf{r}) = \varphi(\mathbf{r}) - \varphi^c(\mathbf{r}), \quad \boldsymbol{\varepsilon}'(\mathbf{r}) = \boldsymbol{\varepsilon}(\mathbf{r}) - \boldsymbol{\varepsilon}^c,$$

then, subtracting (4) from (3), we obtain the boundary problem

$$\nabla \cdot \boldsymbol{\varepsilon}^c \nabla \varphi'(\mathbf{r}) = -\nabla \cdot \boldsymbol{\varepsilon}'(\mathbf{r}) \nabla \varphi(\mathbf{r}), \quad \varphi'\big|_S = 0. \tag{5}$$

By entering the Green's function $G(\mathbf{r}, \mathbf{r}_1)$

$$\nabla \cdot \boldsymbol{\varepsilon}^c \nabla G(\mathbf{r}, \mathbf{r}_1) = -\delta(\mathbf{r} - \mathbf{r}_1), \quad G(\mathbf{r}, \mathbf{r}_1)\big|_{\mathbf{r}_1 \in S} = 0,$$

the solution of problem (5) can be written as an integral [18]

$$\varphi'(\mathbf{r}) = \int\limits_V G(\mathbf{r}, \mathbf{r}_1) \big(\nabla \cdot \boldsymbol{\varepsilon}'(\mathbf{r}_1) \nabla \varphi(\mathbf{r}_1)\big) d\mathbf{r}_1. \tag{6}$$

If the sample is considered unbounded, then $G(\mathbf{r}, \mathbf{r}_1) = G(\mathbf{r}_1 - \mathbf{r})$, and (6) will

$$\varphi'(\mathbf{r}) = \int G(\mathbf{r}_1 - \mathbf{r}) \big(\nabla \cdot \boldsymbol{\varepsilon}'(\mathbf{r}_1) \nabla \varphi(\mathbf{r}_1)\big) d\mathbf{r}_1. \tag{7}$$

Integrating (7) by parts and taking the gradient from the left and right parts, we obtain

$$\mathbf{E}'(\mathbf{r}) = \int \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r}) \boldsymbol{\varepsilon}'(\mathbf{r}_1) \mathbf{E}(\mathbf{r}_1) d\mathbf{r}_1, \tag{8}$$

where $\nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r})$ — tensor of second derivatives of the Green's function, the upper index 1 of the Hamiltonian differential operator means differentiation by $\mathbf{r}_1$. Since $\mathbf{E}'(\mathbf{r}) = \mathbf{E}(\mathbf{r}) - \mathbf{E}^c$, where $\mathbf{E}^c = \text{const}$ — the electric field strength in the comparison body, from (8), we obtain the equation for the electric field strength in the sample of the inhomogeneous medium, which can be written as

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^c + \mathbf{Q}(\mathbf{r}) \big(\boldsymbol{\varepsilon}(\mathbf{r}) - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}), \tag{9}$$

where $\mathbf{Q}(\mathbf{r})$ — tensor integral operator whose action is defined by the formula

$$\mathbf{Q}(\mathbf{r}) \mathbf{f}(\mathbf{r}) = \int \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r}) \mathbf{f}(\mathbf{r}_1) d\mathbf{r}_1.$$

Due to the heterogeneous structure, the sample can be considered to consist of a finite number of inclusions. Note that if the heterogeneous medium is a matrix-type composite with inclusions, the matrix can also be considered to consist of individual grains without a shell. The shape of matrix particles in the following discussion is assumed to be ellipsoidal with fixed aspect ratio and orientation (a special case of the shape of the above particles — spherical). It should be noted that the area occupied by the matrix can be covered with non-overlapping particles of a given shape and orientation with any predetermined accuracy, using particles of different sizes. In this case, the smaller particles will fill the space left between the larger ones. The material characteristics of all these imaginary model particles should coincide with the material characteristics of the matrix regardless of their size, in contrast to the material characteristics of real inclusions, which may depend on the size of the latter.

So, let the current point $\mathbf{r}$ lie inside the kth inclusion. Let us decompose the operator $\mathbf{Q}(\mathbf{r})$ into external and internal components with respect to the k-inclusion:

$$\mathbf{Q}(\mathbf{r}) = \mathbf{Q}^{(k)}_{\text{ext}}(\mathbf{r}) + \mathbf{Q}^{(k)}_{\text{int}}(\mathbf{r}),$$

then (9) will take the form

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^c + \mathbf{Q}^{(k)}_{\text{ext}}(\mathbf{r}) \big(\boldsymbol{\varepsilon}(\mathbf{r}) - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}) + \mathbf{Q}^{(k)}_{\text{int}}(\mathbf{r}) \big(\boldsymbol{\varepsilon}(\mathbf{r}) - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}),$$

$$\mathbf{r} \in V^{(k)}. \tag{10}$$

Let us write out in detail the terms in (10) corresponding to the external and internal components of the operator $\mathbf{Q}(\mathbf{r})$:

$$\mathbf{Q}^{(k)}_{\text{ext}}(\mathbf{r}) \big(\boldsymbol{\varepsilon}(\mathbf{r}) - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r})$$

$$= \sum_{\substack{i=1 \\ i \neq k}}^{N} \int\limits_{V^{(i)}} \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r}) \big(\boldsymbol{\varepsilon}(\mathbf{r}_1) - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}_1) d\mathbf{r}_1.$$

$$\mathbf{Q}^{(k)}_{\text{int}}(\mathbf{r}) \big(\boldsymbol{\varepsilon}(\mathbf{r}) - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r})$$

$$= \sum_{j=1}^{n} \int\limits_{V_j^{(k)}} \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r}) \big(\boldsymbol{\varepsilon}_j^{(k)} - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}_1) d\mathbf{r}_1. \tag{11}$$

The first two terms in (10) can be called the effective field strength at a given point of the kth inclusion, which is formed as a result of the application of an external field to the composite sample and the presence of other inclusions in the sample:

$$\mathbf{E}^{(k)}_{\text{eff}}(\mathbf{r}) = \mathbf{E}^c + \mathbf{Q}^{(k)}_{\text{ext}}(\mathbf{r}) \big(\boldsymbol{\varepsilon}(\mathbf{r}) - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}), \;\; \mathbf{r} \in V^{(k)}. \tag{12}$$

Let's rewrite (10) with (11), (12) in mind:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}^{(k)}_{\text{eff}}(\mathbf{r}) + \sum_{j=1}^{n} \int\limits_{V_j^{(k)}} \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r})$$

$$\times \big(\boldsymbol{\varepsilon}_j^{(k)} - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}_1) d\mathbf{r}_1, \quad \mathbf{r} \in V^{(k)}.$$

2. Solving the problem in the generalized effective-field approximation for the case of ellipsoidal boundaries of nuclei and inclusion shell layers

Calculate the average intensities $\langle \mathbf{E} \rangle_j^{(k)}$ $(j = \overline{1, n})$ of the electric field in all layers of the shell and the core of the kth inclusion:

$$\langle \mathbf{E} \rangle_j^{(k)} = \langle \mathbf{E}_{\text{eff}} \rangle_j^{(k)} + \frac{1}{V_j^{(k)}} \sum_{i=1}^{n} \int\limits_{V_i^{(k)}} \Bigg(\int\limits_{V_j^{(k)}} \nabla^1 \otimes \nabla^1 G(\mathbf{r}_{1i} - \mathbf{r}) d\mathbf{r} \Bigg)$$

$$\times \big(\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c\big) \mathbf{E}(\mathbf{r}_{1i}) d\mathbf{r}_{1i}, \;\; j = \overline{1, n}, \tag{13}$$

where

$$\langle \mathbf{E}_{\text{eff}} \rangle_j^{(k)} = \frac{1}{V_j^{(k)}} \int_{V_j^{(k)}} \mathbf{E}_{\text{eff}}^{(k)}(\mathbf{r}) d\mathbf{r}, \quad j = \overline{1, n}$$

— average effective field strengths in the shell layers and core of the k-th inclusion. In (13) the order of integration over $\mathbf{r}$ and $\mathbf{r}_{1i}$ $(i = \overline{1, n})$ is changed in averaging; this is justified due to continuity or uniform convergence of corresponding integrals over $\mathbf{r}$ and $\mathbf{r}_{1i}$ in these areas [22].

Next, we will assume that the boundaries of the nuclei and all shell layers of all inclusions are ellipsoids. Then the areas $\tilde{V}_j^{(k)}$ $(j = \overline{1, n})$ defined by expression (1) are the insides of the corresponding ellipsoids. Let's introduce the designations:

$$\tilde{\mathbf{g}}_{j,i}^{(k)}(\mathbf{r}_1) = \int_{V_j^{(k)}} \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r}) d\mathbf{r}, \quad \mathbf{r}_1 \in V_i^{(k)},$$

$$\mathbf{g}_{j,i}^{(k)}(\mathbf{r}_1) = \int_{\tilde{V}_j^{(k)}} \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r}) d\mathbf{r}, \quad \mathbf{r}_1 \in V_i^{(k)},$$

$$j = \overline{1, n}; \quad i = \overline{1, n};$$

$$\mathbf{g}_j^{(k)}(\mathbf{r}_1) = \int_{\tilde{V}_j^{(k)}} \nabla^1 \otimes \nabla^1 G(\mathbf{r}_1 - \mathbf{r}) d\mathbf{r} = \int_{\tilde{V}_j^{(k)}} \nabla \otimes \nabla G(\mathbf{r}) d\mathbf{r},$$

$$\mathbf{r}_1 \in \tilde{V}_j^{(k)}, \quad j = \overline{1, n}. \tag{14}$$

All tensors $\mathbf{g}_j^{(k)}$ are independent of the choice of point $\mathbf{r}_1$ inside the corresponding ellipsoidal area $\tilde{V}_j^{(k)}$ [23]. For tensors $\mathbf{g}_{j,i}^{(k)}(\mathbf{r}_1)$ we have

$$\mathbf{g}_{j,i}^{(k)}(\mathbf{r}_1) = \mathbf{g}_j^{(k)}, \quad i \geq j. \tag{15}$$

For the tensors $\tilde{\mathbf{g}}_{j,i}^{(k)}(\mathbf{r}_1)$ we have

$$\tilde{\mathbf{g}}_{j,i}^{(k)}(\mathbf{r}_1) = \mathbf{g}_j^{(k)} - \mathbf{g}_{j+1}^{(k)}, \quad i = \overline{j+1, n}, \tag{16}$$

$$\tilde{\mathbf{g}}_{j,j}^{(k)}(\mathbf{r}_1) = \mathbf{g}_j^{(k)} - \mathbf{g}_{j+1,j}^{(k)}(\mathbf{r}_1), \tag{17}$$

$$\tilde{\mathbf{g}}_{j,i}^{(k)}(\mathbf{r}_1) = \mathbf{g}_{j,i}^{(k)}(\mathbf{r}_1) - \mathbf{g}_{j+1,i}^{(k)}(\mathbf{r}_1), \; i \leq j-1. \tag{18}$$

Given (14)−(18), expressions (13) will take the form

$$\langle \mathbf{E} \rangle_j^{(k)} = \langle \mathbf{E}_{\text{eff}} \rangle_j^{(k)} + \frac{1}{V_j^{(k)}} \left[\sum_{i=1}^{j-1} \int_{V_i^{(k)}} \left(\mathbf{g}_{j,i}^{(k)}(\mathbf{r}_{1i}) - \mathbf{g}_{j+1,i}^{(k)}(\mathbf{r}_{1i}) \right) \times (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) \mathbf{E}(\mathbf{r}_{1i}) d\mathbf{r}_{1i} + \int_{V_j^{(k)}} \left(\mathbf{g}_j^{(k)} - \mathbf{g}_{j+1,j}^{(k)}(\mathbf{r}_{1j}) \right) (\boldsymbol{\varepsilon}_j^{(k)} - \boldsymbol{\varepsilon}^c) \times \mathbf{E}(\mathbf{r}_{1j}) d\mathbf{r}_{1j} + \sum_{i=j+1}^{n} (\mathbf{g}_j^{(k)} - \mathbf{g}_{j+1}^{(k)}) (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) V_i^{(k)} \langle \mathbf{E} \rangle_i^{(k)} \right],$$

$$j = \overline{1, n},$$

or

$$\langle \mathbf{E} \rangle_j^{(k)} = \langle \mathbf{E}_{\text{eff}} \rangle_j^{(k)} + \frac{1}{V_j^{(k)}} \left[\sum_{i=1}^{j-1} \int_{V_i^{(k)}} \mathbf{g}_{j,i}^{(k)}(\mathbf{r}_{1i}) (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) \mathbf{E}(\mathbf{r}_{1i}) d\mathbf{r}_{1i} - \sum_{i=1}^{j} \int_{V_i^{(k)}} \mathbf{g}_{j+1,i}^{(k)}(\mathbf{r}_{1i}) (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) \mathbf{E}(\mathbf{r}_{1i}) d\mathbf{r}_{1i} + \sum_{i=j}^{n} \mathbf{g}_j^{(k)} \times (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) V_i^{(k)} \langle \mathbf{E} \rangle_i^{(k)} - \sum_{i=j+1}^{n} \mathbf{g}_{j+1}^{(k)} (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) V_i^{(k)} \langle \mathbf{E} \rangle_i^{(k)} \right], \tag{19}$$

$$j = \overline{1, n}.$$

Using (19), find the expression for the average field strength in k-th inclusion

$$\langle \mathbf{E} \rangle^{(k)} = \sum_{j=1}^{n} f_j^{(k)} \langle \mathbf{E} \rangle_j^{(k)} = \langle \mathbf{E}_{\text{eff}} \rangle^{(k)} + \frac{1}{V^{(k)}} \left[\sum_{j=1}^{n} \sum_{i=1}^{j-1} \int_{V_i^{(k)}} \mathbf{g}_{j,i}^{(k)}(\mathbf{r}_{1i}) (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) \mathbf{E}(\mathbf{r}_{1i}) d\mathbf{r}_{1i} - \sum_{j=1}^{n} \sum_{i=1}^{j} \int_{V_i^{(k)}} \mathbf{g}_{j+1,i}^{(k)}(\mathbf{r}_{1i}) (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) \mathbf{E}(\mathbf{r}_{1i}) d\mathbf{r}_{1i} + \sum_{j=1}^{n} \mathbf{g}_j^{(k)} \sum_{i=j}^{n} (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) V_i^{(k)} \langle \mathbf{E} \rangle_i^{(k)} - \sum_{j=1}^{n} \mathbf{g}_{j+1}^{(k)} \sum_{i=j+1}^{n} (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) V_i^{(k)} \langle \mathbf{E} \rangle_i^{(k)} \right], \tag{20}$$

where $\langle \mathbf{E}_{\text{eff}} \rangle^{(k)}$ — the average effective field strength in the kth inclusion:

$$\langle \mathbf{E}_{\text{eff}} \rangle^{(k)} = \sum_{j=1}^{n} f_j^{(k)} \langle \mathbf{E}_{\text{eff}} \rangle_j^{(k)},$$

$f_j^{(k)}$ — the relative volume fractions of the shell and core layers in the kth inclusion:

$$f_j^{(k)} = \frac{V_j^{(k)}}{V^{(k)}}, \quad j = \overline{1, n}.$$

It is natural to assume in (20) that

$$\mathbf{g}_{n+1,i}^{(k)}(\mathbf{r}_{1i}) \equiv 0, \quad i = \overline{1, n}; \quad \mathbf{g}_{n+1}^{(k)} = 0. \tag{21}$$

Note that in (20) in the first double sum the internal sum at $j = 1$ is zero. Also in the second and fourth double sums, in force (21), the external summation can be carried out up to $(n-1)$. Further, it is easy to check that the first two double sums compensate each other completely, and in the third and fourth double sums most of the summands also „cancel„each other, so for the average field strength in the kth inclusion we obtain the following expression:

$$\langle \mathbf{E}\rangle^{(k)} = \langle \mathbf{E}_{\text{eff}}\rangle^{(k)} + \mathbf{g}_1^{(k)} \sum_{i=1}^{n} f_i^{(k)} (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c) \langle \mathbf{E}\rangle_i^{(k)}. \quad (22)$$

The average field strength in each particular shell layer of the k-th inclusion is related to the average field strength in the core of the same inclusion by the linear relation

$$\langle \mathbf{E}\rangle_j^{(k)} = \boldsymbol{\lambda}_{jn}^{(k)} \langle \mathbf{E}\rangle_n^{(k)}, \quad j = \overline{1, n-1}, \quad (23)$$

where $\boldsymbol{\lambda}_{jn}^{(k)}$ — tensor operator that depends on the geometric and material characteristics of this inclusion, the comparison body, and the characteristics and arrangement of the other inclusions in the sample. By substituting (23) into the equation obtained by equating the right-hand side (22) and the middle part (20), we express $\langle \mathbf{E}\rangle_n^{(k)}$ through $\langle \mathbf{E}_{\text{eff}}\rangle^{(k)}$:

$$\langle \mathbf{E}\rangle_n^{(k)} = \boldsymbol{\lambda}_{n0}^{(k)} \langle \mathbf{E}_{\text{eff}}\rangle^{(k)}, \quad (24)$$

where

$$\boldsymbol{\lambda}_{n0}^{(k)} = \left[\sum_{i=1}^{n} f_i^{(k)} \big(\mathbf{I} - \mathbf{g}_1^{(k)} (\boldsymbol{\varepsilon}_i^{(k)} - \boldsymbol{\varepsilon}^c)\big) \boldsymbol{\lambda}_{in}^{(k)} \right]^{-1}, \quad (25)$$

and obviously $\boldsymbol{\lambda}_{nn}^{(k)} = \mathbf{I}$.

Using (22) and (24), we connect the average field strength in the kth inclusion with the average effective field strength in it:

$$\langle \mathbf{E}\rangle^{(k)} = \left(\sum_{i=1}^{n} f_i^{(k)} \boldsymbol{\lambda}_{in}^{(k)} \right) \boldsymbol{\lambda}_{n0}^{(k)} \langle \mathbf{E}_{\text{eff}}\rangle^{(k)}.$$

For the average field strength in the sample we obtain

$$\langle \mathbf{E}\rangle = \sum_{k=1}^{N} \frac{V^{(k)}}{V} \langle \mathbf{E}\rangle^{(k)} = \frac{1}{V} \sum_{k=1}^{N} V^{(k)} \left(\sum_{i=1}^{n} f_i^{(k)} \boldsymbol{\lambda}_{in}^{(k)} \right) \times \boldsymbol{\lambda}_{n0}^{(k)} \langle \mathbf{E}_{\text{eff}}\rangle^{(k)}.$$

Let us introduce an operator $\boldsymbol{\Lambda}^{(k)}$ linking the average effective field strengths in the kth inclusion and in the whole sample:

$$\langle \mathbf{E}_{\text{eff}}\rangle^{(k)} = \boldsymbol{\Lambda}^{(k)} \langle \mathbf{E}_{\text{eff}}\rangle, \quad k = \overline{1, N}, \quad (26)$$

and $\langle \mathbf{E}_{\text{eff}}\rangle$ is calculated similarly $\langle \mathbf{E}\rangle$:

$$\langle \mathbf{E}_{\text{eff}}\rangle = \sum_{k=1}^{N} \frac{V^{(k)}}{V} \langle \mathbf{E}_{\text{eff}}\rangle^{(k)}.$$

Then the expression for $\langle \mathbf{E}\rangle$ can be written as

$$\langle \mathbf{E}\rangle = \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle \langle \mathbf{E}_{\text{eff}}\rangle, \quad (27)$$

where

$$\left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle = \frac{1}{V} \sum_{k=1}^{N} V^{(k)} \left(\sum_{i=1}^{n} f_i^{(k)} \boldsymbol{\lambda}_{in}^{(k)} \right) \boldsymbol{\lambda}_{n0}^{(k)} \boldsymbol{\Lambda}^{(k)}.$$

From (27), express the average effective field strength in the sample through the average field strength in it

$$\langle \mathbf{E}_{\text{eff}}\rangle = \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle^{-1} \langle \mathbf{E}\rangle. \quad (28)$$

Using (23), (24), (26), (28), let us express the average field strengths in the layers of the shell and in the core of k-th inclusion through the average field strength in the sample

$$\langle \mathbf{E}\rangle_j^{(k)} = \boldsymbol{\lambda}_{jn}^{(k)} \boldsymbol{\lambda}_{n0}^{(k)} \boldsymbol{\Lambda}^{(k)} \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle^{-1} \langle \mathbf{E}\rangle,$$

$$j = \overline{1, n}, \quad k = \overline{1, N}.$$

For the average value of the electric induction in the kth inclusion, we have

$$\langle \mathbf{D}\rangle^{(k)} = \sum_{i=1}^{n} f_i^{(k)} \boldsymbol{\varepsilon}_i^{(k)} \langle \mathbf{E}\rangle_i^{(k)} = \left(\sum_{i=1}^{n} f_i^{(k)} \boldsymbol{\varepsilon}_i^{(k)} \boldsymbol{\lambda}_{in}^{(k)} \right) \boldsymbol{\lambda}_{n0}^{(k)} \boldsymbol{\Lambda}^{(k)} \times \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle^{-1} \langle \mathbf{E}\rangle, \quad k = \overline{1, N}. \quad (29)$$

Using (29), let's calculate the average induction in the sample

$$\langle \mathbf{D}\rangle = \sum_{k=1}^{N} \frac{V^{(k)}}{V} \langle \mathbf{D}\rangle^{(k)} = \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\varepsilon}_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle \times \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle^{-1} \langle \mathbf{E}\rangle, \quad (30)$$

where

$$\left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\varepsilon}_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle = \frac{1}{V} \sum_{k=1}^{N} V^{(k)} \left(\sum_{i=1}^{n} f_i^{(k)} \boldsymbol{\varepsilon}_i^{(k)} \boldsymbol{\lambda}_{in}^{(k)} \right) \times \boldsymbol{\lambda}_{n0}^{(k)} \boldsymbol{\Lambda}^{(k)}.$$

From (2) and (30) the expression for the effective dielectric characteristic tensor of a given sample of heterogeneous material follows

$$\boldsymbol{\varepsilon}^* = \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\varepsilon}_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \boldsymbol{\Lambda} \right\rangle^{-1}. \quad (31)$$

The formula (31) is exact, but the practical calculation $\boldsymbol{\varepsilon}^*$ of it requires knowledge of the operators $\boldsymbol{\lambda}_{in}$ $(i = \overline{1, n})$ and $\boldsymbol{\Lambda}$ for each of the sample inclusions, which is a gigantic complexity due to the huge number of inclusions and their mutual influence on each other. The statistical homogeneity of the material and the smallness of each inclusion compared to the entire material sample justify the representation of the $\mathbf{\Lambda}$ operator as

$$\mathbf{\Lambda} = \mathbf{I} + \delta\mathbf{\Lambda},$$

where $\delta\mathbf{\Lambda}$ — the fluctuation additive resulting from the irregular distribution of inclusions over the sample volume, their differences in size, shape, material characteristics, and orientation of crystallographic axes. In turn, the operators $\boldsymbol{\lambda}_{in}$ can be represented as

$$\boldsymbol{\lambda}_{in} = \boldsymbol{\lambda}_{in}^0 + \delta\boldsymbol{\lambda}_{in}, \quad i = \overline{1, n},$$

where $\boldsymbol{\lambda}_{in}^0$ — tensor relating the average field strengths in the shell layers and the core of an inclusion identical to the given one but in solitude in the comparison medium with a uniform applied field; $\delta\boldsymbol{\lambda}_{in}$ — correction due to the influence of other inclusions.

As a first simplifying assumption, let us assume that

$$\mathbf{\Lambda} \approx \mathbf{I},$$

i.e. we will neglect the fluctuation of the effective field. In this case, for the tensor $\boldsymbol{\varepsilon}^*$ we obtain the expression

$$\boldsymbol{\varepsilon}^* = \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\varepsilon}_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \right\rangle \left\langle \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in} \right) \boldsymbol{\lambda}_{n0} \right\rangle^{-1}, \quad (32)$$

which can be called a generalized effective-field approximation for $\boldsymbol{\varepsilon}^*$ medium consisting of inclusions with multilayer shells.

As a second simplifying assumption, we may assume that the average field strengths in the shell and core of each inclusion are related in the same way as those of a single inclusion with the same parameters, placed in an infinite comparison medium with a uniform applied field, i.e.

$$\boldsymbol{\lambda}_{in} \approx \boldsymbol{\lambda}_{in}^0, \quad i = \overline{1, n}. \quad (33)$$

Note that the tensor components $\mathbf{g}_1^{(k)}$ included in expression (25) to calculate $\boldsymbol{\lambda}_{n0}^{(k)}$ $(k = \overline{1, N})$ in the coordinate system associated with the axes of kth inclusion can be calculated by the formula [20]:

$$g_{ij}^{(k)} = -\frac{1}{4\pi} \int_0^{\pi} \int_0^{2\pi} \frac{n_i n_j}{n_s \varepsilon_{sl}^c n_l} \sin\vartheta \, d\vartheta \, d\varphi, \quad (34)$$

where ε_{sl}^c, $s, l = 1, 2, 3$ — components of the tensor $\boldsymbol{\varepsilon}^c$ in this coordinate system; $n_1 = (a_1^{(k)})^{-1} \cos\varphi \sin\vartheta$, $n_2 = (a_2^{(k)})^{-1} \sin\varphi \sin\vartheta$, $n_3 = (a_3^{(k)})^{-1} \cos\vartheta$ — the components of the normal (non unit) to the ellipsoid surface which is the boundary of kth inclusion; $a_1^{(k)}$, $a_2^{(k)}$, $a_3^{(k)}$ — its half-axes.

Averaging in (32) is performed over all inclusions in the sample. If all inclusions in a sample — are of the same type in terms of their material characteristics and differ only in their shapes and orientations in space, then the angular brackets in (32) should be interpreted as an averaging of the corresponding tensor quantities associated with the inclusions over the shapes and orientations of the inclusions, taking their probability distributions into account.

It should be noted that the approximate value for $\boldsymbol{\varepsilon}^*$ obtained with expression (32) depends on the comparison medium parameter — tensor $\boldsymbol{\varepsilon}^c$ of its permittivity, and in case of heterogeneous matrix medium also on a vector parameter, characterizing the shape and orientation of grains composing the matrix. By varying the values of these parameters, we can obtain different types of approximations for the tensor $\boldsymbol{\varepsilon}^*$, and their specific values should be chosen based on the features of the structure of the heterogeneous medium. For example, for a matrix medium with a volume fraction of inclusions not exceeding 0.4, it is reasonable to choose the matrix as the comparison medium, for a heterogeneous medium such as a statistical mixture the medium itself with effective permittivity, i.e. use the idea of self-consistency [20].

As for the choice of the shape of the matrix particles, for a macroscopically isotropic heterogeneous medium of the matrix type, it seems to be necessary to consider their shape as spherical. In the case of anisotropic media, the choice of matrix particle shape requires separate consideration and is not discussed in this paper. It should be noted, however, that accepting the matrix as a comparison medium automatically removes the question of choosing the form of its particles due to the structure of the obtained expression (25) for the tensor $\boldsymbol{\lambda}_{n0}^{(k)}$, which fully agrees with the physical sense of this situation, when the particles of matrix „merges“ with comparison medium.

3. Some particular applications of the generalized effective-field approximation

3.1. Heterogeneous medium consisting of homogeneous inclusions

Let us first consider some of the limiting cases when the heterogeneous medium consists of homogeneous inclusions.

3.1.1. The core is present, but all layers of the shell are absent

Let's assume that all inclusions have a core, but all shell layers are missing, i.e. $f_n^{(k)} = 1$, $f_i^{(k)} = 0$, $i = \overline{1, n-1}$. Then by formula (25), given that $\boldsymbol{\lambda}_{nn}^{(k)} = \mathbf{I}$, we have

$$\boldsymbol{\lambda}_{n0}^{(k)} = \left(\mathbf{I} - \mathbf{g}_1^{(k)}(\boldsymbol{\varepsilon}_n^{(k)} - \boldsymbol{\varepsilon}^c)\right)^{-1}, \quad k = \overline{1, N},$$

and since the core occupies the entire volume of the inclusion, $\mathbf{g}_1^{(k)} = \mathbf{g}_n^{(k)} = \mathbf{g}^{(k)}$ — the tensor associated with this kth inclusion and used in the generalized singular approximation [20], and (32) takes form

$$\boldsymbol{\varepsilon}^* = \langle \boldsymbol{\varepsilon}_n [\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon}_n - \boldsymbol{\varepsilon}^c)]^{-1} \rangle \langle [\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon}_n - \boldsymbol{\varepsilon}^c)]^{-1} \rangle^{-1},$$

coinciding with the formula for $\boldsymbol{\varepsilon}^*$ in the generalized singular approximation [20].

3.1.2. All inclusions have no core and no shell layers, except for one layer

Suppose, for example, that for kth inclusion there is only j_kth layer, i.e. $f^{(k)}_{j_k} = 1$, $f^{(k)}_i = 0$, $i \neq j_k$. In this case $V^{(k)} = \tilde{V}^{(k)}_{j_k}$, $\mathbf{g}^{(k)}_1 = \mathbf{g}^{(k)}_{j_k} = \mathbf{g}^{(k)}$,

$$\begin{aligned} \boldsymbol{\lambda}^{(k)}_{n0} &= \left[\left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)}_{j_k} - \boldsymbol{\varepsilon}^c)\right) \boldsymbol{\lambda}^{(k)}_{j_k n} \right]^{-1} \\ &= (\boldsymbol{\lambda}^{(k)}_{j_k n})^{-1} \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)}_{j_k} - \boldsymbol{\varepsilon}^c)\right)^{-1}, \end{aligned}$$

and for the tensor $\boldsymbol{\varepsilon}^*$ we have

$$\begin{aligned} \boldsymbol{\varepsilon}^* &= \left\langle \boldsymbol{\varepsilon}^{(k)}_{j_k} \boldsymbol{\lambda}^{(k)}_{j_k n} (\boldsymbol{\lambda}^{(k)}_{j_k n})^{-1} \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)}_{j_k} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle \\ &\times \left\langle \boldsymbol{\lambda}^{(k)}_{j_k n} (\boldsymbol{\lambda}^{(k)}_{j_k n})^{-1} \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)}_{j_k} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle^{-1} \\ &= \left\langle \boldsymbol{\varepsilon}\left(\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle \left\langle \left(\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle^{-1}, \end{aligned} \quad (35)$$

where

$$\left\langle \boldsymbol{\varepsilon}\left(\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle = \frac{1}{V} \sum_{k=1}^{N} V^{(k)} \boldsymbol{\varepsilon}^{(k)}_{j_k} \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)}_{j_k} - \boldsymbol{\varepsilon}^c)\right)^{-1},$$

$$\left\langle \left(\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle = \frac{1}{V} \sum_{k=1}^{N} V^{(k)} \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)}_{j_k} - \boldsymbol{\varepsilon}^c)\right)^{-1}.$$

Expression (35) also coincides with the formula for $\boldsymbol{\varepsilon}^*$ in the generalized singular approximation.

3.1.3. For each particular inclusion, the material characteristics of all shell layers and of core are the same

Let us consider the case, when for each particular inclusion, the material characteristics of all layers of the shell and the core are the same, i.e. $\boldsymbol{\varepsilon}^{(k)}_1 = \boldsymbol{\varepsilon}^{(k)}_2 = \ldots = \boldsymbol{\varepsilon}^{(k)}_n = \boldsymbol{\varepsilon}^{(k)}$. Then $\mathbf{g}^{(k)}_1 = \mathbf{g}^{(k)}$

$$\boldsymbol{\lambda}^{(k)}_{n0} = \left[\sum_{i=1}^{n} f^{(k)}_i \boldsymbol{\lambda}^{(k)}_{in} \right]^{-1} \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)} - \boldsymbol{\varepsilon}^c)\right)^{-1},$$

for $\boldsymbol{\varepsilon}^*$, we also end up with an expression in the generalized singular approximation:

$$\begin{aligned} \boldsymbol{\varepsilon}^* &= \left\langle \boldsymbol{\varepsilon}^{(k)} \left(\sum_{i=1}^{n} f^{(k)}_i \boldsymbol{\lambda}^{(k)}_{in} \right) \left[\sum_{i=1}^{n} f^{(k)}_i \boldsymbol{\lambda}^{(k)}_{in} \right]^{-1} \right. \\ &\quad \left. \times \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle \\ &\quad \times \left\langle \left(\sum_{i=1}^{n} f^{(k)}_i \boldsymbol{\lambda}^{(k)}_{in} \right) \left[\sum_{i=1}^{n} f^{(k)}_i \boldsymbol{\lambda}^{(k)}_{in} \right]^{-1} \right. \\ &\quad \left. \times \left(\mathbf{I} - \mathbf{g}^{(k)}(\boldsymbol{\varepsilon}^{(k)} - \boldsymbol{\varepsilon}^c)\right)^{-1} \right\rangle^{-1} \\ &= \langle \boldsymbol{\varepsilon}\left(\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c)\right)^{-1} \rangle \langle \left(\mathbf{I} - \mathbf{g}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^c)\right)^{-1} \rangle^{-1}. \end{aligned}$$

Thus, in the limiting cases of homogeneous inclusions, expression (32) coincides with the expression for $\boldsymbol{\varepsilon}^*$ in the generalized singular approximation [20]. Note, that in all the considered limiting cases, no simplifying assumptions were made about the type of operators $\boldsymbol{\lambda}_{in}$, $i = \overline{1, n}$.

3.2. Composite consisting of isotropic matrix and of identical inclusions with isotropic spherical core and shell with isotropic spherical layers

Let's assume that ε_m — permittivity of the matrix, ε_i, $i = \overline{1, n}$ — permittivity of the shell layers, including the core, of each inclusion; $a_1 > a_2 > \ldots > a_n$ — radii of spheres which are the boundaries of shell and core. Let's assume that f — the total volume fraction of all inclusions in the medium. Let's take the comparison medium as isotropic: $\boldsymbol{\varepsilon}^c = \varepsilon^c \mathbf{I}$. By direct calculation according to formula (34) we obtain that

$$\mathbf{g}^{(k)}_1 = -(3\varepsilon^c)^{-1} \mathbf{I}, \quad k = \overline{1, N}. \quad (36)$$

Tensor operators $\boldsymbol{\lambda}_{in}$, $i = \overline{1, n}$, will be taken in approximation (33), to find the tensor $\boldsymbol{\lambda}^0_{in}$ form, we calculate the average electric field strength in all layers of the shell of an isolated inclusion in an infinite comparison medium with a uniform applied field $\mathbf{E}_0$.

Let's assume that $\mathbf{k}$ — orth in the direction of $\mathbf{E}_0$, then the field potential in ith layer of the shell will have the form [24]:

$$\varphi_i(\mathbf{r}) = A_i(\mathbf{k} \cdot \mathbf{r}) + B_i \frac{(\mathbf{k} \cdot \mathbf{r})}{r^3}, \quad i = \overline{1, n-1}, \quad (37)$$

where A_i, B_i — constants, which are calculated using boundary conditions. The field in the inclusion core is uniform, its potential

$$\varphi_n(\mathbf{r}) = A_n(\mathbf{k} \cdot \mathbf{r}), \quad A_n = \text{const}. \quad (38)$$

For the average field strength in the ith layer of an insulated inclusion shell, by definition, we have (V_i — volume occupied by the ith layer of the inclusion shell):

$$\langle \mathbf{E}_i \rangle = (V_i)^{-1} \iiint\limits_{V_i} \mathbf{E}_i dV = (V_i)^{-1} \iiint\limits_{V_i} (-\nabla\varphi_i) dV. \quad (39)$$

Let us change in (39) from the volume integral to the integral over the total surface of the ith layer of the shell using the gradient theorem [25]:

$$\langle \mathbf{E}_i \rangle = (V_i)^{-1} \left[-\oiint\limits_{S_i} \varphi_i \mathbf{n}_i dS + \oiint\limits_{S_{i+1}} \varphi_i \mathbf{n}_{i+1} dS \right], \quad (40)$$

where S_i, S_{i+1} — spheres that are the outer and inner boundaries of the ith layer of the shell, respectively, $\mathbf{n}_i$ and $\mathbf{n}_{i+1}$ — the outer unit normals to them. Given (37) and the fact that on the sphere $\mathbf{n} = \mathbf{r}/r$, for the first of the integrals in (40), we have the expression

$$\oiint\limits_{S_i} \varphi_i \mathbf{n}_i dS = \oiint\limits_{S_i} \left(A_i + B_i \frac{1}{r^3} \right) \frac{(\mathbf{k}\cdot\mathbf{r})}{r} \mathbf{r} dS. \quad (41)$$

Parameterize S_i with spherical angles θ, ψ (θ — the angle between vectors $\mathbf{k}$ and $\mathbf{r}$):

$$\mathbf{r}\big|_{S_i} = \begin{pmatrix} a_i \sin\theta\cos\psi \\ a_i \sin\theta\sin\psi \\ a_i\cos\theta \end{pmatrix}, \ 0 \le \theta \le \pi, \ 0 \le \psi < 2\pi, \ r\big|_{S_i} = a_i,$$

then

$$\frac{(\mathbf{k}\cdot\mathbf{r})}{r} = \cos\theta, \quad dS = a_i^2 \sin\theta d\theta d\psi.$$

Let us substitute these expressions in (41):

$$\oiint\limits_{S_i} \varphi_i \mathbf{n}_i dS = \int\limits_0^{\pi} d\theta \int\limits_0^{2\pi} d\psi \left(A_i + B_i \frac{1}{a_i^3} \right) a_i^3 \begin{pmatrix} \sin\theta\cos\psi \\ \sin\theta\sin\psi \\ \cos\theta \end{pmatrix} \times \sin\theta\cos\theta.$$

Obviously, the first two components of the integral are zero, and for the third we have (axis z is directed along $\mathbf{k}$):

$$\left(\oiint\limits_{S_i} \varphi_i \mathbf{n}_i dS \right)_z = \frac{4\pi}{3} (A_i a_i^3 + B_i).$$

Similarly, for the second integral in (40), the first two components are also zero, and for the third, we obtain

$$\left(\oiint\limits_{S_{i+1}} \varphi_i \mathbf{n}_{i+1} dS \right)_z = \frac{4\pi}{3} (A_i a_{i+1}^3 + B_i).$$

With account of the fact that

$$V_i = \frac{4\pi}{3} (a_i^3 - a_{i+1}^3),$$

as a result, for the average electric field strength in the ith layer of the shell, we have the following value:

$$\langle \mathbf{E}_i \rangle = -A_i \mathbf{k}, \quad i = \overline{1, n-1}. \quad (42)$$

From (38), the expression for the average electric field strength in the core follows evidently

$$\langle \mathbf{E}_n \rangle = -A_n \mathbf{k}. \quad (43)$$

From (42), (43), we have an expression for the tensors $\boldsymbol{\lambda}_{in}^0$:

$$\boldsymbol{\lambda}_{in}^0 = \frac{A_i}{A_n} \mathbf{I}, \quad i = \overline{1, n}. \quad (44)$$

Substituting (36) and (44) into (25), we find

$$\boldsymbol{\lambda}_{n0} = \frac{3\varepsilon^c A_n}{\sum\limits_{i=1}^{n} f_i (2\varepsilon^c + \varepsilon_i) A_i} \mathbf{I}. \quad (45)$$

The particles of the matrix will be considered spherical without a shell, for them

$$f_i^{(m)} = 0, \ i = \overline{1, n-1}; \quad f_n^{(m)} = 1; \quad \boldsymbol{\lambda}_{n0}^{(m)} = \frac{3\varepsilon^c}{(2\varepsilon^c + \varepsilon_m)} \mathbf{I}. \quad (46)$$

Taking into account the composition of the medium in question, the averaging in (32) is reduced to the arithmetic mean with weights equal to the volume fractions of the medium components:

$$\boldsymbol{\varepsilon}^* = \left[(1-f)\varepsilon_m \boldsymbol{\lambda}_{n0}^{(m)} + f \left(\sum_{i=1}^{n} f_i \varepsilon_i \boldsymbol{\lambda}_{in}^0 \right) \boldsymbol{\lambda}_{n0} \right] \times \left[(1-f)\boldsymbol{\lambda}_{n0}^{(m)} + f \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}_{in}^0 \right) \boldsymbol{\lambda}_{n0} \right]^{-1}. \quad (47)$$

Obviously, in this case, the effective permittivity of an heterogeneous medium will be a scalar quantity, the expression for which is obtained after substituting (44)–(46) into (47) and elementary algebraic transformations:

$$\varepsilon^* = \varepsilon_m \frac{\sum\limits_{i=1}^{n} f_i [(1-f) 2\varepsilon^c \varepsilon_m + f(2\varepsilon^c + \varepsilon_m)\varepsilon_i] A_i}{\sum\limits_{i=1}^{n} f_i [(1-f)(2\varepsilon^c + \varepsilon_i) + f(2\varepsilon^c + \varepsilon_m)\varepsilon_i] A_i}. \quad (48)$$

If we take a matrix as the comparison medium, i.e. $\varepsilon^c = \varepsilon_m$, then (48) will take shape

$$\varepsilon^* = \varepsilon_m \frac{\sum\limits_{i=1}^{n} f_i [2\varepsilon_m (1-f) + \varepsilon_i (1+2f)] A_i}{\sum\limits_{i=1}^{n} f_i [\varepsilon_m (2+f) + \varepsilon_i (1-f)] A_i}. \quad (49)$$

In the case of balls without a shell ($n = 1$, $f_1 = 1$), the classical Maxwell–Garnett formula is obtained from formula (49):

$$\varepsilon^* = \varepsilon_m \frac{2\varepsilon_m + \varepsilon_1 + 2f(\varepsilon_1 - \varepsilon_m)}{2\varepsilon_m + \varepsilon_1 - f(\varepsilon_1 - \varepsilon_m)}.$$

3.3. Composite consisting of isotropic matrix and identical inclusions with anisotropic ellipsoidal core and multilayer shell with anisotropic ellipsoidal layers

Let's assume that ε_m — permittivity of the matrix, $\boldsymbol{\varepsilon}_i$, $i = \overline{1, n}$, the permittivity tensors of the shell layers, including the core, of each inclusion; $a_{i,1}, a_{i,2}, a_{i,3}$, $i = \overline{1, n}$ — the half-axes of the surfaces ellipsoids $S_i^{(k)}$, which are the boundaries of the shell layers and the core of k-th inclusion. Let's assume that f — the total volume fraction of all inclusions in the medium. All inclusions will be considered equally oriented in space.

We will assume that the sizes and orientations of the geometric axes of the surfaces-ellipsoids $S_i^{(k)}$, $i = \overline{1, n}$, are coordinated with the tensors $\boldsymbol{\varepsilon}_i$, $i = \overline{1, n}$, in a certain way. In particularly, for the ith shell layer, the images of its inner $S_{i+1}^{(k)}$ and outer $S_i^{(k)}$ boundaries are confocal ellipsoids under the linear transformation T_i, which eliminates the anisotropy of its dielectric properties. The initial Cartesian coordinate system in which the geometric parameters of the inclusions are given, denote as $x^1x^2x^3$. The coordinate transformation associated with the transformation T_i has the form

$$\mathbf{r} = \mathbf{T}_i\mathbf{r}_i',$$

where $\mathbf{r} = (x^1x^2x^3)^T$, $\mathbf{r}_i' = (x_i^{1'}x_i^{2'}x_i^{3'})^T$ — vector-columns of coordinates of the current point in the original $x^1x^2x^3$ and new $x_i^{1'}x_i^{2'}x_i^{3'}$ coordinate systems; $\mathbf{T}_i$ — matrix of this transformation. We denote the ellipsoid surface images $S_i^{(k)}$ and $S_{i+1}^{(k)}$ under the transformation T_i as $S_i'^{(k)}$ and $S_{i+1}'^{(k)}$; denote their half-axes as $a_{i,1'}, a_{i,2'}, a_{i,3'}$ and $a_{i+1,1'}, a_{i+1,2'}, a_{i+1,3'}$. The matrix $\mathbf{T}_i$ of this transformation is related to the permittivity tensor $\boldsymbol{\varepsilon}_i$ of the ith shell layer by the relation [26]:

$$\boldsymbol{\varepsilon}_i = \mathbf{T}_i\mathbf{T}_i^T. \tag{50}$$

Condition (50) ambiguously determines the transformation T_i (with accuracy to an arbitrary rotation around the origin), so in addition to (50), we will assume that the axes of the system $x_i^{1'}x_i^{2'}x_i^{3'}$ are directed along the axes of ellipsoids $S_i'^{(k)}$, $S_{i+1}'^{(k)}$. The confocality of $S_i'^{(k)}$ and $S_{i+1}'^{(k)}$ means that there exists a parameter $t_i' > 0$, that

$$t_i' = a_{i,i'}^2 - a_{i+1,i'}^2, \quad i' = 1', 2', 3'.$$

Let's take the comparison medium as isotropic: $\boldsymbol{\varepsilon}^c = \varepsilon^c\mathbf{I}$. Tensor operators $\boldsymbol{\lambda}_{in}$, $i = \overline{1, n}$, will be taken in approximation (33), to find the tensor species $\boldsymbol{\lambda}_{in}^0$, we calculate the average electric field strength in all layers of the shell of an isolated inclusion in an infinite comparison medium with a uniform applied field $\mathbf{E}_0$.

Consider the ith layer of the kth inclusion, which we will consider isolated in an infinite comparison environment. The electric field potential in it has the form (the upper index denoting the inclusion number is omitted everywhere, because the dependence will be the same for all inclusions) [26]

$$\varphi_i = \big((\boldsymbol{\beta}_i + \mathbf{N}_{i,0}'(\xi_i')\boldsymbol{\alpha}_i)\mathbf{E}_0, \mathbf{r}\big),\ \mathbf{r} \in V_i,\ 0 \le \xi_i' \le t_i',$$

where $\boldsymbol{\alpha}_i$, $\boldsymbol{\beta}_i$ — constant tensors of rank 2; $\mathbf{N}_{i,0}'(\xi_i')$ — tensor function, in coordinate system $x^1x^2x^3$ calculated by the formula

$$\mathbf{N}_{i,0}'(\xi_i') = (\mathbf{T}_i^{-1})^T\mathbf{N}_i'(\xi_i')\mathbf{T}_i^{-1}, \tag{51}$$

$\mathbf{N}_i'(\xi_i')$ — the same tensor function in the coordinate system $x_i^{1'}x_i^{2'}x_i^{3'}$, which has a diagonal form with principal components:

$$N_{i,i'}'(\xi_i') = \frac{a_{i+1,1'}a_{i+1,2'}a_{i+1,3'}}{2}\int\limits_{\xi_i'}^{+\infty}\frac{du}{[u + a_{i+1,i'}^2]\tilde{R}_u},$$

$$i' = 1', 2', 3'; \quad 0 \le \xi_i' \le t_i';$$

$$\tilde{R}_u = \left[(u + a_{i+1,1'}^2)(u + a_{i+1,2'}^2)(u + a_{i+1,3'}^2)\right]^{1/2}.$$

Notice, that

$$\mathbf{N}_{i,0}'(0) = \mathbf{L}_{i+1,0}'^{(ext)}, \quad \mathbf{N}_{i,0}'(t_i') = \tilde{v}_{i+1}\mathbf{L}_{i,0}'^{(int)},$$

where $\tilde{v}_{i+1}$ — the relative volume fraction of the volume inside the surface $S_{i+1}^{(k)}$ in the volume inside the surface $S_i^{(k)}$, i. e.

$$\tilde{v}_{i+1} = \frac{a_{i+1,1}a_{i+1,2}a_{i+1,3}}{a_{i,1}a_{i,2}a_{i,3}},$$

$\mathbf{L}_{i,0}'^{(int)}$ — tensor of generalized geometric factors of the ellipsoid $S_i^{(k)}$ considering the anisotropy of dielectric properties in the nearest shell layer inside $S_i^{(k)}$ in coordinate system $x^1x^2x^3$; $\mathbf{L}_{i+1,0}'^{(ext)}$ — tensor of generalized geometric factors of the ellipsoid $S_{i+1}^{(k)}$ considering the anisotropy of dielectric properties in the nearest shell layer outside $S_{i+1}^{(k)}$ in the $x^1x^2x^3$ coordinate system [26]. Similarly (51), there is a relation of these tensors to the same tensors in the coordinate system $x_i^{1'}x_i^{2'}x_i^{3'}$:

$$\mathbf{L}_{i,0}'^{(int)} = (\mathbf{T}_i^{-1})^T\mathbf{L}_i'^{(int)}\mathbf{T}_i^{-1}, \quad \mathbf{L}_{i+1,0}'^{(ext)} = (\mathbf{T}_i^{-1})^T\mathbf{L}_{i+1}'^{(ext)}\mathbf{T}_i^{-1}.$$

Using a procedure similar to the calculation of the average field strength in a one-layer [27] shell, we obtain the following expression for the average field strength in the ith layer of the shell:

$$\langle\mathbf{E}_i\rangle = \left[-\boldsymbol{\beta}_i + \frac{\tilde{v}_{i+1}}{1 - \tilde{v}_{i+1}}\big(\mathbf{L}_{i+1,0}'^{(ext)} - \mathbf{L}_{i,0}'^{(int)}\big)\boldsymbol{\alpha}_i\right]\mathbf{E}_0. \tag{52}$$

The field inside the core is uniform, its intensity can be written as $\mathbf{E}_n = -\boldsymbol{\beta}_n\mathbf{E}_0$, where $\boldsymbol{\beta}_n$ — a constant rank 2 tensor, so the average field strength in the core

$$\langle\mathbf{E}_n\rangle = -\boldsymbol{\beta}_n\mathbf{E}_0. \tag{53}$$

From (52), (53) the form of tensors $\boldsymbol{\lambda}_{in}^0$ follows:

$$\boldsymbol{\lambda}_{in}^0 = \left[\boldsymbol{\beta}_i - \frac{\tilde{v}_{i+1}}{1 - \tilde{v}_{i+1}}\big(\mathbf{L}_{i+1,0}'^{(ext)} - \mathbf{L}_{i,0}'^{(int)}\big)\boldsymbol{\alpha}_i\right]\boldsymbol{\beta}_n^{-1},\ i = \overline{1, n}, \tag{54}$$

and one should naturally assume that $\tilde{v}_{n+1} = 0$. Substituting (54) into (25), we obtain

$$\boldsymbol{\lambda}_{n0} = \boldsymbol{\beta}_n \left[\sum_{i=1}^{n} \big(\mathbf{I} - \mathbf{g}_1(\boldsymbol{\varepsilon}_i - \boldsymbol{\varepsilon}^c) \big) \times \left[f_i \boldsymbol{\beta}_i - v'_{i+1} \big(\mathbf{L}'^{(ext)}_{i+1,0} - \mathbf{L}'^{(int)}_{i,0} \big) \boldsymbol{\alpha}_i \right] \right]^{-1}, \quad (55)$$

where v'_{i+1} — volume fraction of the volume inside the surface $S^{(k)}_{i+1}$ to the volume of the whole inclusion, i.e.

$$v'_{i+1} = \frac{a_{i+1,1} a_{i+1,2} a_{i+1,3}}{a_{1,1} a_{1,2} a_{1,3}}, \quad i = \overline{0, n-1}; \quad v'_{n+1} = 0.$$

Here, it has been taken into account that

$$f_i = \frac{a_{i,1} a_{i,2} a_{i,3} - a_{i+1,1} a_{i+1,2} a_{i+1,3}}{a_{1,1} a_{1,2} a_{1,3}} = (1 - \tilde{v}_{i+1}) v'_i, \quad i = \overline{1, n}.$$

As in the considered example 3.2, the matrix can be considered to consist of spherical particles without a shell, for them (see (46))

$$f_i^{(m)} = 0, \ i = \overline{1, n-1}; \quad f_n^{(m)} = 1; \quad \boldsymbol{\lambda}^{(m)}_{n0} = \frac{3\varepsilon^c}{(2\varepsilon^c + \varepsilon_m)} \mathbf{I}.$$

Taking into account the identity of all inclusions and their identical orientation in space, the averaging in (32) is reduced to the calculation of the arithmetic mean with weights equal to the volume fractions of the components, i.e. to calculate $\boldsymbol{\varepsilon}^*$ we use a formula similar to (47) and substituting (46) in it we obtain

$$\boldsymbol{\varepsilon}^* = \left[(1-f)\varepsilon_m \frac{3\varepsilon^c}{(2\varepsilon^c + \varepsilon_m)} \mathbf{I} + f \left(\sum_{i=1}^{n} f_i \boldsymbol{\varepsilon}_i \boldsymbol{\lambda}^0_{in} \right) \boldsymbol{\lambda}_{n0} \right] \times \left[(1-f) \frac{3\varepsilon^c}{(2\varepsilon^c + \varepsilon_m)} \mathbf{I} + f \left(\sum_{i=1}^{n} f_i \boldsymbol{\lambda}^0_{in} \right) \boldsymbol{\lambda}_{n0} \right]^{-1},$$

where $\boldsymbol{\lambda}^0_{in}$, $i = \overline{1, n}$, and $\boldsymbol{\lambda}_{n0}$ are calculated by formulas (54), (55) respectively.

Conclusion

The main results of this work are the generalized effective-field approximation proposed therein for calculating the effective characteristics of an heterogeneous medium consisting of inclusions with a multilayer shell, as well as the expression (32) for the effective permittivity tensor of such a medium obtained with its help. This approximation has a high degree of generality and can be applied to multicomponent media and allows taking into account the texture and probabilistic distribution of inclusion shapes. It is shown that in the limiting cases of media with homogeneous inclusions, the approximation proposed in this paper gives the same results as the generalized singular approximation [20].

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Conflict of interest

The authors declare that they have no conflict of interest.

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