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ASYMMETRIC EFFECTIVE MEDIUM APPROXIMATION FOR DESCRIBING THE OPTICAL CHARACTERISTICS OF RANDOMLY INHOMOGENEOUS MEDIA WITH DISCRETE INCLUSIONS

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ABSTRACT

The symmetric Bruggeman approximation, also known as the Effective Medium Approximation (EMA), is widely used in applications including the description of light scattering on inhomogeneous structures containing discrete inclusions. However, in the latter case, the natural asymmetry of the fillers topology, where discrete inclusions are mostly surrounded by the material of a simply connected matrix, is not taken into account. In this paper, two versions of asymmetric EMA are proposed for the case of a statistically isotropic medium containing discrete inclusions based on the difference in the structure of the fields inside and outside the inclusions. One of them does not differ too much from the usual EMA and leads to the same percolation threshold. For the second, the threshold value differs from the usual one even in the case of spherical particles. Expressions are given for the corresponding percolation thresholds in the model of randomly oriented elliptical particles. The proposed approximations are compared with the standard Maxwell Garnett and Bruggeman approximations for the case of silver particles in a dielectric matrix.

Keywords: effective parameters of randomly inhomogeneous media, homogenization, Bruggeman and Maxwell Garnett approximations, percolation threshold

1. INTRODUCTION

In describing the optical characteristics of macroscopically inhomogeneous media, both natural and artificial, various "mixing rules" are widely used, which make it possible to approximately replace a real small-scale compared to a wavelength randomly inhomogeneous medium with a homogeneous one with some effective parameters (see, for example, the review [1] and the literature cited there).

Of the large number of known models for calculating effective parameters, the Maxwell-Garnett approximation (MGA) and the Bruggeman effective medium approximation (EMA) are distinguished. The first of them is constructed for a model of a homogeneous medium with random inclusions, and the second considers a symmetric composite completely filled by randomly distributed particles with different macroscopic characteristics. In this case, the MGA is an asymmetric approximation in which one of the components is selected and plays the role of a matrix. As a result, the MGA, unlike the symmetric EMA, does not allow to describe the percolation threshold associated with the occurrence of "sticking together" of random inclusions in an infinite cluster upon an increase of their concentration, which is considered to be a drawback of this approximation. At low concentrations of inhomogeneities far from the threshold, both approximations give identical results. In this paper, we obtain simple asymmetric EMA modifications based on a model of a homogeneous medium with random scatterer, and at the same time allowing us to describe the occurrence of a percolation threshold.

2. DERIVATION OF ASYMMETRIC EMA EQUATIONS

Consider a homogeneous medium with a permittivity ε_0 containing statistically uniformly distributed particles with a permittivity ε_1 occupying a volume fraction f_1, so that the volume fraction of a particle-free medium is $f_0 = 1 - f_1$. Considering the medium to be small-scale, so that the particle sizes and the distances between them are small compared to the wavelengths of the radiation in question, we use the quasistatic approximation. In this approximation the electrical and magnetic properties of the medium can be described independently. For the case of a homogeneous external field E_{out} = const created by sources outside the medium (a strict setting of the boundary conditions can be found in [2]), the effective permittivity ε^* can be determined by the relation [1]

$$\varepsilon^* = \langle \varepsilon E \rangle_v / \langle E \rangle_v = (f_0 \langle \varepsilon_0 E_0 \rangle + f_1 \langle \varepsilon_1 E_1 \rangle) / \langle E \rangle, \quad (1)$$

$$\langle E \rangle = f_0 \langle E_0 \rangle + f_1 \langle E_1 \rangle. \quad (2)$$

Here angle brackets with index V mean averaging over "physically infinitesimal volume", small compared to the total volume of the medium, but containing a large number of particles,

$$\langle \ \rangle_v = \frac{1}{V}\int_V \ldots dr, \quad (3)$$

and the same brackets without an index are statistical averaging, which includes averaging over the volumes of each component (for simplicity, we do not introduce special notation for vector quantities).

The fields E_0 and E_1 included in (1) can be considered, respectively, as fluctuating fields outside and inside the "characteristic particle". All further approximations are associated with statistical hypotheses about the properties of these fields. For simplicity, here we restrict ourselves to the case of spherical particles (a generalization to the ellipsoid model can be easily obtained taking into account the results of [3]). In the simplest approximation, valid in the limit of highly rarefied media (formally $f_1 \rightarrow 0$), the field outside the particles E_0 is assumed to be equal to the field in their absence, $E_0 = E_{out}$, thereby completely neglecting the effect of scattering on E_0. In this case, the field inside the characteristic particle will be expressed through the external field E_0 by the known relation [4]

$$E_1 = \Lambda_{10} E_0, \ \Lambda_{10} = \frac{3\varepsilon_0}{\varepsilon_1 + 2\varepsilon_0}. \quad (4)$$

In this approximation, the angle brackets on the right side of (1) can be omitted, which after reducing by E_0 gives the usual MGA

$$\varepsilon^* = (f_0 \varepsilon_0 + f_1 \varepsilon_1 \Lambda_{10}) / (f_0 + f_1 \Lambda_{10}). \quad (5)$$

The use of EMA is associated with an attempt to take into account the mutual influence of scattering by particles, self-consistently considering as a "characteristic particle" a spherical particle located in a uniform average field $\langle E \rangle$ in an "effective medium" with a dielectric constant ε^*. Wherein

$$E_1 = \Lambda_{1*} \langle E \rangle, \ \Lambda_{1*} = \frac{3\varepsilon^*}{\varepsilon_1 + 2\varepsilon^*}. \quad (6)$$

In the usual Bruggeman approximation [5], it is assumed that in (1) in estimating the field outside the particles E_0, similar ratios should be used, i.e.

$$E_0 = \Lambda_{0*} \langle E \rangle, \ \Lambda_{0*} = \frac{3\varepsilon^*}{\varepsilon_0 + 2\varepsilon^*}, \quad (7)$$

leading to a well-known relation

$$f_0 \frac{\varepsilon_0 - \varepsilon^*}{\varepsilon_0 + 2\varepsilon^*} + f_1 \frac{\varepsilon_1 - \varepsilon^*}{\varepsilon_1 + 2\varepsilon^*} = 0. \quad (8)$$

Conditions (7) correspond to the aggregate topology, i.e. the case when the component with ε_0 consists of spherical particles. However, for the asymmetric case considered here with a distinguished medium and discrete inclusions, there is no reason to use (7) for the field between the particles, since the array of the medium has no direct connection with the spherical shape of the particles.

Within the framework of the self-consistent approximation, it suffices to assume that the field outside the particles E_0 is approximately equal to the average field, i.e. having kept (6) for particles, instead of (7) for the medium field, put in (1) $E_0 = \langle E \rangle$. Substituting this relation and (6) into (1), after simple transformations, instead of (8), we obtain

$$f_1 \frac{\varepsilon_1 - \varepsilon^*}{\varepsilon_1 + 2\varepsilon^*} + f_0 \frac{\varepsilon_0 - \varepsilon^*}{2\varepsilon^*} = 0. \quad (9)$$

A more reasonable approximation than (9), “intermediate” between EMA and MGA, is obtained when calculating the field inside the “effective particle” as an external field with respect to the particle if instead of the total average field <E> the average field between the particles <E_0> is taken. Then (1) and (2) give

$$\varepsilon^* = (f_0\, \varepsilon_0 + f_1\, \varepsilon_1\, \Lambda_{1*}) / (f_0 + f_1\, \Lambda_{1*}). \qquad (10)$$

This relation coincides in form with MGA (5), however, taking into account (6), it does not represent an explicit expression, but the equation for ε^*, which can be easily reduced to a form similar to (9)

$$f_1 \frac{\varepsilon_1 - \varepsilon^*}{\varepsilon_1 + 2\varepsilon^*} + f_0 \frac{\varepsilon_0 - \varepsilon^*}{3\varepsilon^*} = 0. \qquad (11)$$

In the next section, we compare some properties of the approximations considered here.

3. SOME CONSEQUENCES

All the approximations described in the previous section admit a formal transition to the case of complete filling of the medium with particles. $f_1 = 1$ when $\varepsilon^* = \varepsilon_1$. These approximations are valid in the general case of complex permittivities. The same expressions are preserved during the transition from the dielectric constant ε to the description of the medium conductivity σ, as well as when describing many other kinetic coefficients (see [6]).

It is easy to show (see [1]) that all forms of EMA (8) – (10) describe the occurrence of a percolation threshold. However, if approximation (9), like the usual Bruggeman approximation (8), gives the percolation threshold $f_{1c} = 1/3$, then equation (11) already has a different threshold value, namely $f_{1c} = 1/4$.

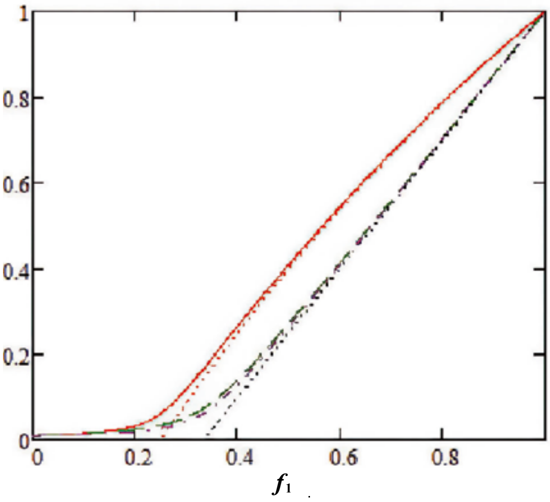

Fig. 1. Effective conductivity for a medium with conductivity σ_0 and particles with conductivity σ_1 at $\sigma_0/\sigma_1 = 0.01$ in models (8) – – –, (9) – - – and (10) ▬. The dots indicate the limiting values of these curves for a none-conducting medium $\sigma_0 = 0$

All approximations (8) – (10) lead to quadratic equations with respect to ε^*, which are easily solved. To illustrate, Fig. 1 shows the dependences of the effective conductivities of a weakly conducting medium with conducting particles in models (8) – (10). It can be seen from this figure that model (9) with a threshold $f_{1c} = 1/3$ gives a result that is qualitatively close to the usual Bruggeman model (8), while model (10) is noticeably different from (8) due to differences in the percolation thresholds.

As another example consider the case of silver nanoparticles in a matrix with refractive index $n = 1.5$. Fig. 2 shows the behaviour of real and imaginary parts of the complex refractive index $N = \sqrt{\varepsilon}$ for the volume silver according to the data taken

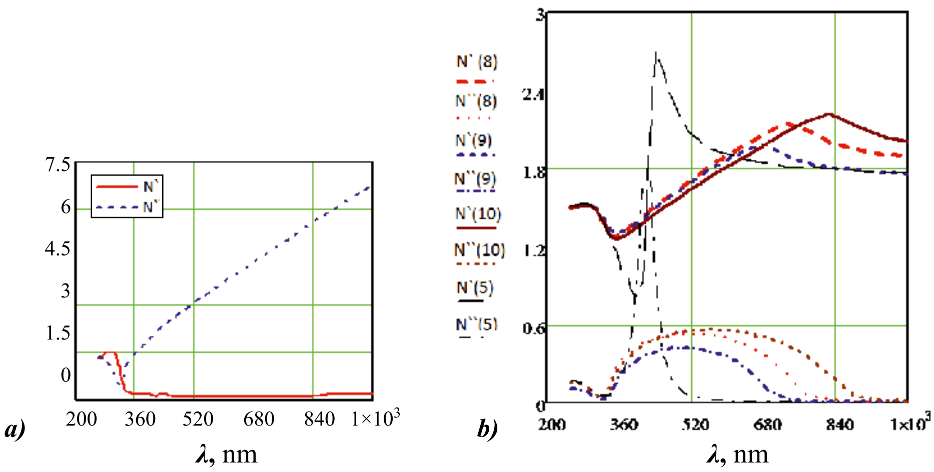

Fig. 2. The real and imaginary parts of the refractive index $N = N' + iN''$ (a) bulk *Ag* according to [7], and (b) effective refractive index in models MGA (5) and EMA (8) – (10) with $f_1 = 0.1$

from [7], as well as for the effective refractive index $N^* = \sqrt{\varepsilon^*}$ for a medium with silver nanoparticles in the MGA (5) and EMA (8) – (10) with filling $f_1 = 0{,}1$. It can be seen from these figures that all the considered models of the effective medium in this example noticeably differ from the MGA, and give qualitatively similar, but quantitatively different results. Moreover, if MGA describes the presence of a narrow plasmon resonance, then all EMA schemes give a broadened resonance with a maximum shifted to the red side ("red shift").

4. DISCUSSION

The examples considered above show that the proposed modifications give results that are qualitatively similar to the usual EMA with noticeable quantitative differences. It is not difficult to generalize these approximations to the case of randomly oriented elliptic particles. To do this, it suffices to replace the quantities Λ_{1*} and Λ_{10} by corresponding tensor expressions, supplementing the averaging symbols in (1) by averaging over random orientations, which reduces to calculating one third of the trace of the matrix [1]. Moreover, in the case of ellipsoids with a depolarization tensor L for model (10) the percolation threshold is equal to

$$f_{1c} = 1/\left(1 + \left\langle \frac{1}{\mathrm{L}} \right\rangle\right), \tag{12}$$

and for the model (9)

$$f_{1c} = 1/\left\langle \frac{1}{\mathrm{L}} \right\rangle, \tag{13}$$

where

$$\left\langle \frac{1}{\mathrm{L}} \right\rangle = \frac{1}{3}\mathrm{Sp}\,\frac{1}{\mathrm{L}}, \tag{14}$$

and the division by L is understood in the sense of matrix inversion. Expression (13) is also obtained by using a direct generalization of the standard form EMA (8) to the case of elliptic cells, if we only accept, as is usually done, that the points of the medium correspond to spherical cells. This expression is also preserved for models with fluctuating depolarization factors L, for which it is sufficient to supplement the right-hand side of (14) with statistical averaging over L.

When using effective parameters in applications, the question naturally arises about the conditions of applicability of certain models. Necessary conditions are the applicability of the quasistatic approximation. However, sufficient conditions cannot be indicated in the general case, since in real problems the particles are not strictly randomly distributed and can also have some complex internal structure, for which the statistical models under consideration can serve only as a rough approximation (a useful discussion of the lack of universal effective parameters for macroscopically inhomogeneous media can be found in [8]). Therefore, the choice of a particular model is usually justified only by comparing their results with specific real or numerical experiments. As the last example, we can mention the work [9], in which the classical EMA (8) is compared with the results of numerical calculations

5. CONCLUSION

In this note we have considered two variants of EMA for the case of a medium with discrete inclusions, which take into account the difference between the simply connected matrix topology and the topology of single particles. One of them does not differ much from the commonly used symmetric Bruggeman approximation and gives the same flow threshold $f_1 = 1/3$, while for the second this difference is more significant and corresponds to a lower threshold value $f_1 = 1/4$. Both approximations can be easily generalized to the case of randomly oriented elliptic particles. It can be expected that using these approximations will find useful applications in practical applications.

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