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THE EXTENSION OF THE MAXWELL GARNETT MIXING RULE FOR DIELECTRIC COMPOSITES WITH NONUNIFORM ORIENTATION OF ELLIPSOIDAL INCLUSIONS

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Abstract—This paper presents the extension of the Maxwell Garnett effective medium model accounting for an arbitrary orientation of ellipsoidal inclusions. The proposed model is shown to be asymptotically convergent to the Maxwell Garnett mixing rule for a homogenous distribution of inclusions. Subsequently, a special case of a thin composite layer with a two-dimensional distribution of inclusions is considered and a simplified Maxwell Garnett formula is formally derived. The proposed model is validated against the alternative theoretical calculations and measurements data.

1. INTRODUCTION

Recently, approximate modeling of macroscopic electromagnetic properties of mixtures has gained an increasing interest, mainly due to the growing applicability of polymer composites reinforced with conductive inclusions, such as carbon fibers or nanotubes. A large market for such composites can be found in manufacturing of electromagnetic shielding and absorbing materials exhibiting a competitive performance when compared to classical panels, like heavy metallic ones.

Host materials in such inhomogeneous compositions are usually made of polymers possessing advantageous properties, like low density, low permittivity, negligible losses, good mechanical processability and many others. As a popular example, epoxy resin [1], polyester [2], polystyrene [3], polyethylene [4], or polypropylene [5] can be recalled.

In many applications, inclusions dispersed in the polymer are made of conductive carbon-based fillers, such as carbon black (CB)

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powders, carbon fibers (CF) or carbon nanotubes (CNT). The advantage of the fibers and even more of the nanotubes is in their high aspect ratio and relatively large electrical conductivity, so that a small amount of such inclusions, even much below a percolation threshold [6], leads to a substantial change of electrical properties of a composite without a significant increase of an overall weight.

However, engineering of electromagnetic shields and absorbers based on carbon-reinforced composites requires quantitative knowledge of their electromagnetic properties, if one does not want to rely solely on costly cut-and-try experiments. The most straightforward way to approach the issue is to apply one of the known numerical electromagnetic techniques, like the finite element method or the finite difference time domain one. Unfortunately, brute-force electromagnetic modeling that represents microscopic details of a mixture is still prohibitively time-consuming to be applied in a real design cycle, mainly due to an extremely large ratio between an operating wavelength (e.g., 30 mm in X-band) and the smallest dimensions of carbon inclusions (diameters at the nanometer scale). In electromagnetic modeling, spatial discretization is usually determined by the operating wavelength, with practical recommendations of 10–20 spatial cells per wavelength that suppress the dominant numerical dispersion errors to 1–0.25%, respectively.

In the case of mixtures, discretization would need to be refined so as to appropriately capture tiny geometrical details of the inclusions. For the considered example, the refinement would be by a factor of roughly $1.5\,\mathrm{mm}/15\,\mathrm{nm} = 10^5$, increasing memory requirements by ca. 10^{15} and computing time by 10^{20}. This unfavorable scaling naturally stimulates a search for the effective (quasi-static) representation of electromagnetic properties of such composites.

There is a variety of mathematical models that aim to represent effective electromagnetic properties of mixtures. Most of them exhibit very stringent limitations that must be satisfied to achieve a reliable solution. One of the simplest approximations is known as the Maxwell Garnett mixing rule [7]:

$$\varepsilon_{\mathit{eff}} = \varepsilon_b + \frac{\frac{1}{3} f_i \left(\varepsilon_i - \varepsilon_b\right) \sum\limits_{k=1}^{3} \frac{\varepsilon_b}{\varepsilon_b + N_k(\varepsilon_i - \varepsilon_b)}}{1 - \frac{1}{3} f_i \left(\varepsilon_i - \varepsilon_b\right) \sum\limits_{k=1}^{3} \frac{N_k}{\varepsilon_b + N_k(\varepsilon_i - \varepsilon_b)}} \tag{1}$$

where $\varepsilon_b = \varepsilon\varepsilon_{b,r}$ denotes permittivity of a host material, $\varepsilon_i = \varepsilon\varepsilon_{i,r}$ is bulk permittivity of ellipsoidal inclusions, f_i is the volume fraction of inclusions, and N_k stands for so-called depolarization factors that can be calculated from the following integral [8, 9]:

$$N_k = 0.5 c_x c_y c_z \int_0^\infty \frac{dr}{\left(r + c_k^2\right)\sqrt{\left(r + c_x^2\right)\left(r + c_y^2\right)\left(r + c_z^2\right)}} \tag{2}$$

where $k = x, y, z$ denotes Cartesian coordinates, and c_x, c_y, c_z stand for semi-axes of an ellipsoidal inclusion.

There are also other widely recognized models representing effective permittivity of mixtures, such as Bruggeman [10], McLachlan [6, 11, 12], or differential mixing rule [13] methods. However, the advantage of the Maxwell Garnett model is that, for a given volume fraction of inclusions f_i, it explicitly provides effective permittivity of a mixture with no need of iterative calculations. However, there is a rigid requirement that, in the case of conducting inclusions, a mixture is far below the percolation threshold, understood as a transition between isolating and conducting properties [6, 12]. If inclusions are in the shape of spheroids with a large aspect ratio $a = l/d \gg 1$, where l is the length and d is the diameter of a spheroid, the percolation threshold is usually approximated as $p_c \sim 1/a$ [12]. It indicates that, with the increasing aspect ratio, the percolation threshold decreases and, in consequence, special attention must be paid whether the Maxwell Garnett model still provides a reliable solution. Another inherent limitation of the Maxwell Garnett formula is a quasi-static approximation requiring a distance between inclusions to be much smaller than the operating wavelength [8]. That requirement is usually satisfied in the microwave spectrum region, if one considers polymer composites reinforced with elongated carbon inclusions.

The Maxwell Garnett mixing rule, in one of its common versions, represents effective permittivity of a composite with randomly oriented ellipsoidal inclusions uniformly dispersed in a host material. Such

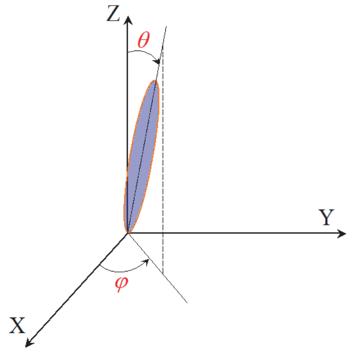


Figure 1. A single inclined spheroidal inclusion.

effective permittivity becomes isotropic, even though it contains strongly anisotropic ellipsoidal inclusions. However, it can happen that — due to some bias occurring in a mixing process — the orientation of inclusions is not purely random, contributing to anisotropy of the mixture. Let us consider, for instance, a very thin composite layer reinforced with carbon fibers, such as paint composites [14] or thin shielding screens. Due to a very small thickness of the processed composite, with respect to the fibers' average length, the orientation of those fibers is mostly two-dimensional. Referring to Figure 1, if a thin composite is laid in the xy-plane ($\theta = 90°$), carbon fibers are uniformly distributed within the range of $\varphi = 0, \dots, 360°$. However, due to the symmetry of the spheroidal inclusions only the half of space needs to be considered, that is, $\varphi = 0, \dots, 180°$. Consequently, such a composite exhibits uniaxial anisotropy with the properties along the z-axis being different from those in the xy-plane.

In order to represent effective permittivity of such anisotropic composite using the Maxwell Garnett approximation, a formula taking into account the orientation of inclusions has to be derived. In general, the problem of an arbitrary distribution of ellipsoidal inclusions was addressed many years ago [8, 15]. However, the authors did not proceed to solutions for any specific non-uniform distribution of the inclusions' orientation. Formally, such specific solutions could be derived based on Equation (18) in [8]. Yet, most authors do not follow this path and continue to use the "intuitive" coefficient of 1.5 [16, 1].

Lately, the paper approaching the Maxwell Garnett approximation of a dielectric mixture with statistically distributed orientation of inclusions has been published [17]. The authors start their investigation representing polarizability of a single ellipsoidal inclusion as a diagonal tensor that is further rotated by a given set of spherical angles φ and θ (see Figure 1). The obtained non-diagonal tensor is, subsequently, applied to represent effective permittivity of a composite reinforced with several arbitrarily oriented inclusions that occupy a particular volume fraction. Although the method introduced in [17] addresses the issue in an interesting way, the paper lacks computational examples validating the proposed method. However, a simple test shows that the solution as of [17] does not asymptotically converge to the well-established isotropic solution of the Maxwell Garnett mixing rule. Therefore, in this paper the alternative solution of the Maxwell Garnett formula for dielectric composites with the arbitrary non-uniform orientation of ellipsoidal inclusions will be derived. Additionally, the already mentioned "intuitive" coefficient of 1.5 will be verified.

In the next Section, the Maxwell Garnett effective permittivity formula for the given distribution of inclusions orientations will be formally derived and validated. Afterwards, that formula will be used to establish effective permittivity of a composite with a 2D distribution of inclusions.

2. FORMULA DERIVATION

The solution of the Laplace's equation [18, 19], derived in an ellipsoidal coordinate system for a single ellipsoid buried in a homogeneous dielectric host and aligned with one of Cartesian coordinates leads to a diagonal polarizability tensor. The diagonal coefficients of that tensor are given as (see Equation (10) in [8]):

$$\alpha_k = v_i \frac{\varepsilon_b \left(\varepsilon_i - \varepsilon_b\right)}{\varepsilon_b + N_k \left(\varepsilon_i - \varepsilon_b\right)} \tag{3}$$

where $k = x, y, z$ denotes Cartesian coordinates and v_i stands for an ellipsoid's volume.

The dipole moment of such a single ellipsoidal scattering obstacle may be represented by the following formula:

$$p_k = \alpha_k E_{e,k} = v_i \left(\varepsilon_i - \varepsilon_b\right) E_{i,k} \tag{4}$$

where E_e and E_i stand, respectively, for external and internal electric field components.

In a more general dyadic notation, polarizability can be represented in the following form (see Equation (45) in [19]):

$$\overleftrightarrow{\alpha} = v_i \varepsilon_b \left(\varepsilon_i - \varepsilon_b\right) \left[\varepsilon_b \overleftrightarrow{I} + \overleftrightarrow{L} \left(\varepsilon_i - \varepsilon_b\right)\right]^{-1} \tag{5}$$

where L is a depolarization dyadic which, in the case of an inclusion aligned with the Cartesian coordinates, has the following diagonal form (see Equation (46) in [19]):

$$\overleftrightarrow{L} = \begin{bmatrix} N_x & 0 & 0 \\ 0 & N_y & 0 \\ 0 & 0 & N_z \end{bmatrix} \tag{6}$$

Subsequently, the dipole moment of a single inclusion obtained from Equation (5) can be applied to evaluate effective permittivity of a composite with a given number n of such inclined ellipsoidal inclusions per unit volume. For that purpose, let us introduce an electric displacement vector written as:

$$\vec{D} = \overleftrightarrow{\varepsilon}_{\mathit{eff}} \vec{E}_e = \varepsilon_b \vec{E}_e + \vec{P} \tag{7}$$

where

$$\vec{P} = \sum_m n_m \vec{p}_m \tag{8}$$

is the polarization density and an index m iterates over all types (orientations) of inclusions dispersed in a unit volume of a composite.

In a rough approximation of the dipole moment tensor of a single inclusion dispersed in a mixture (see Equation (4)), it can be assumed that each inclusion is illuminated with the already introduced external electric field E_e. However, a more precise solution should account for the contribution of a field scattered from neighboring inclusions to a local field illuminating each inclusion in a mixture. Consequently, the local field E_L can be written in the following form [8]:

$$\vec{E}_L = \vec{E}_e + \frac{1}{\varepsilon_b}\overleftrightarrow{L}\vec{P} \tag{9}$$

leading to the modified polarizability (compare with Equation (4)):

$$\vec{p} = \overleftrightarrow{\alpha}\vec{E}_L \tag{10}$$

Introducing Equations (9), (10) to Equations (7), (8) with an additional assumption of a bi-phased composition, the following formula for effective permittivity of a mixture can be derived:

$$\overleftrightarrow{\varepsilon}_{\mathit{eff}} = \varepsilon_b\overleftrightarrow{I} + n\overleftrightarrow{\alpha}\left[\overleftrightarrow{I} - \frac{1}{\varepsilon_b}n\overleftrightarrow{\alpha}\overleftrightarrow{L}\right]^{-1} \tag{11}$$

where I represents a unit tensor.

Extension to a multiphase mixture requires slight modification of Equation (11):

$$\overleftrightarrow{\varepsilon}_{\mathit{eff}} = \varepsilon_b\overleftrightarrow{I} + \sum_m n_m\overleftrightarrow{\alpha}_m\left[\overleftrightarrow{I} - \frac{1}{\varepsilon_b}\sum_m n_m\overleftrightarrow{\alpha}_m\overleftrightarrow{L}_m\right] \tag{12}$$

In this Section, effective permittivity of a multi-phase mixture with ellipsoidal inclusions has been formally derived. In Section 3, that solution will be applied to account for a predefined distribution of inclusions.

3. PREDEFINED DISTRIBUTION OF INCLUSIONS

Equation (12) enables the consideration of a statistically distributed orientation of inclusions occupying, in total, a specified volume fraction $f_i = nv_i$, where n is the number of inclusions per unit volume. For that purpose, let the distribution of inclusions be given as follows:

$$n_m = p\left(\theta_m, \varphi_m\right)\sin\left(\theta_m\right)n \tag{13}$$

with the following scaling condition imposed:

$$\sum_{\varphi=0}^{2\pi}\sum_{\theta=0}^{\pi} p\left(\theta, \varphi\right)\sin\left(\theta\right) = 1 \tag{14}$$

where $\sin(\theta)$ is a Jacobian determinant accounting for a rectangular-to-spherical coordinate systems transformation.

Next, for each orientation of inclusions (θ_m, φ_m), both the polarizability tensor (see Equation (10)) and the depolarization dyadic (see Equation (6)) must be rotated and, subsequently, applied in Equation (12). Let us assume, hereafter, that the alignment of inclusions before rotation is along the z-axis ($\theta = 0°$ in Equation (1)) and that the inclusions are in the shape of spheroids, so the two of three semi-axes are equal $c_x = c_y$. Thus, taking advantage of the formulae applied in [17, Equations (11), (12)], the polarizability of a single inclined spheroid can be represented in the following way:

$$\overleftrightarrow{\alpha}_i^{new}(\theta,\varphi) = \alpha_x \overleftrightarrow{I} + (\alpha_z - \alpha_x)\overleftrightarrow{W} \tag{15}$$

where

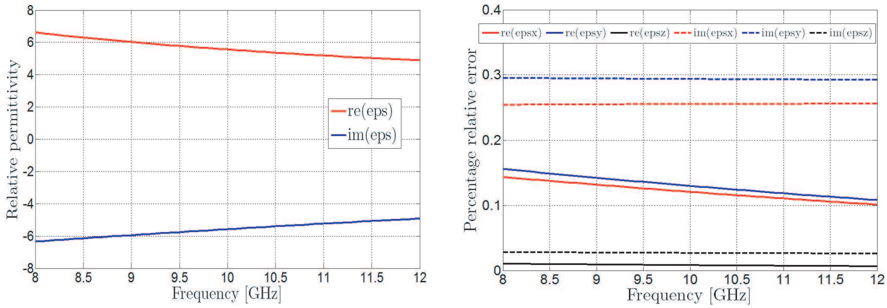
$$\overleftrightarrow{W} = \begin{bmatrix} \cos^2(\phi)\sin^2(\theta) & \cos(\phi)\sin(\phi)\sin^2(\theta) & \cos(\phi)\cos(\theta)\sin(\theta) \\ \cos(\phi)\sin(\phi)\sin^2(\theta) & \sin^2(\phi)\sin^2(\theta) & \sin(\phi)\cos(\theta)\sin(\theta) \\ \cos(\phi)\cos(\theta)\sin(\theta) & \sin(\phi)\cos(\theta)\sin(\theta) & \cos^2(\theta) \end{bmatrix} \tag{16}$$

is a rotation matrix.

In the next Section, computational tests of Equation (12), supplemented with the consideration given in this Section, will be undertaken to validate the formula against theoretical computations and measurements. The issue of the intuitive coefficient of 1.5, introduced in [16], referring to the 2D orientation of inclusions within a dielectric composite, will also be addressed.

4. COMPUTATIONAL TESTS

In the first test, effective permittivity of a mixture with randomly oriented inclusions will be computed using Equation (12) and, afterwards, compared against the well-known isotropic Maxwell Garnett formula (see Equation (1)). In order to focus on a practical case, the results published in [1] will be considered, where an absorbing screen manufactured in an epoxy resin reinforced with carbon fibers was investigated. Measurements published in [1] show that complex permittivity of epoxy is almost non-dispersive within X-band and equals ca. $\varepsilon_{eff} = 3.045 - j0.051$ (see Figure 3 in [1]). After [1], bulk conductivity of carbon fibers is expected to amount to $\sigma_f = 40\,\text{kS/m}$, while their aspect ratio is equal to $a = \text{length/diameter} = 4\,\text{mm}/7\,\mu\text{m} \cong 571.43$. Let us also assume that a total volume fraction amounts to $f_i = 0.028\%$ (as given in Table I in [1]). In the case of spheroidal inclusions aligned with the Cartesian coordinates, depolarization factors as given by Equation (2) amount to $N_x = N_y \cong 0.5$ and $N_z \cong 1.944\text{e-}5$.



(a) (b)

Figure 2. A Maxwell Garnett representation of effective permittivity of an isotropic mixture of carbon fibers dispersed in an epoxy resin. (a) Complex effective permittivity obtained with Equation (1). (b) A relative error of diagonal elements computation with Equation (12) as compared to Equation (1).

Figure 2(a) shows real and imaginary components of isotropic effective permittivity obtained with Equation (1) for the given composite. Next, the same mixture was computed iteratively with Equation (12). Figure 2(b) plots the relative error of diagonal elements computation, as compared to the reference results shown in Figure 2(a). In numerical computations of the effective permittivity tensor as given by Equation (12), an angular discretization step of 1° was taken for both φ and θ variables. Additionally, it is assumed that the probability density $p(\theta, \varphi)$ is constant and normalized according to Equation (14). As shown in Figure 2(b), the error of both real and imaginary parts of diagonal elements computation is on the level below 0.3%. Regarding non-diagonal elements of the tensor given by Equation (12), their values reach a negligible level of ca. 1e-17. The choice of the angular discretization step smaller than 1° yields even better accuracy level but at the cost of higher computational effort.

However, comparing Figure 2(a) with the measurement results published in [1, see Figure 7], it can be clearly seen that those results are different. Apparently, as pointed out in [1], the reason is that the processed composite layer is very thin, as compared to the average length of the applied carbon fibers. Thus, it can be expected that their orientation is mostly two-dimensional within the layer. To account for that, the authors of [1], after [16] applied the already mentioned intuitive coefficient of 1.5 rescaling the effective permittivity tensor in the following way:

$$\overleftrightarrow{\varepsilon}_{eff} = \begin{bmatrix} 1.5\varepsilon_{eff} & 0 & 0 \\ 0 & 1.5\varepsilon_{eff} & 0 \\ 0 & 0 & \varepsilon_b \end{bmatrix} \tag{17}$$

where the scalar ε_{eff} corresponds to the isotropic solution calculated with Equation (1).

Let us validate those premises using Equation (12). The probability density function $p(\theta, \varphi)$ is assumed to have a linear distribution in the xy-plane ($\theta = 90°$) and within the range $\varphi = 0°, \ldots, 360°$ with the angular step of $d\varphi = 1°$. It refers to the case of effective permittivity of a composite with carbon fibers randomly dispersed in the xy-plane.

Figure 3 shows the calculated complex permittivity (red line) compared with the measurement results (green line) taken from [1, Figure 7]. Additionally, effective permittivity calculated with the intuitive tensor given by Equation (17) (as taken from [1, Equation (6)]) is also shown (black dashed line). At first, it can be noticed that the way the coefficient 1.5 is applied does not lead to a correct representation of effective permittivity of the composite with 2D oriented inclusions. A closer insight into the results shown in Figure 3 shows that, excluding a bump obtained in the measurements around 9.5 GHz, the plot of a real part of Equation (17) is shifted up by ca. 1.495 while an imaginary part is well fitted when compared to the measurement data. On the contrary, the iterative solution of Equation (12) (red line) is much better fitted to the measurements (green line). However, if xx- and yy-diagonal elements

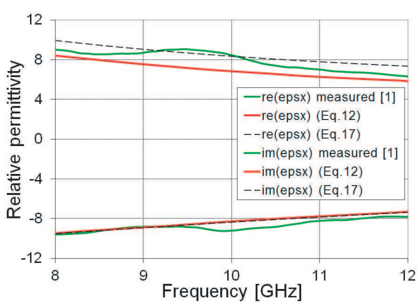


Figure 3. A Maxwell Garnett representation of effective permittivity of an epoxy resin with 2D-oriented carbon fibers.

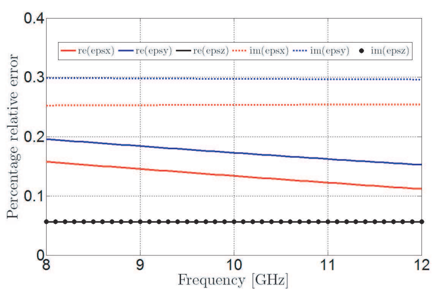


Figure 4. A relative error of complex effective permittivity of 2D-oriented carbon fibers dispersed in an epoxy resin computed with Equation (18) and compared to Equation (12).

in Equation (17), will be modified as follows:

$$\varepsilon_{eff\ x,y} = \varepsilon_b + 1.5 \frac{\frac{1}{3} f_i \left(\varepsilon_i - \varepsilon_b\right) \sum\limits_{k=1}^{3} \frac{\varepsilon_b}{\varepsilon_b + N_k(\varepsilon_i - \varepsilon_b)}}{1 - \frac{1}{3} f_i \left(\varepsilon_i - \varepsilon_b\right) \sum\limits_{k=1}^{3} \frac{N_k}{\varepsilon_b + N_k(\varepsilon_i - \varepsilon_b)}} \tag{18}$$

the obtained solution will fit exactly the iterative solution of Equation (12) (red line). For that reason, the plot of Equation (18) is omitted in Figure 3.

Unlike in Equation (17), only the "mixture part" is rescaled by one 1.5 in Equation (18), what seems to be reasonable, since it can be expected that the contribution of host's permittivity ε_b to total effective permittivity of a composite ε_{eff} should not depend on the specific alignment of inclusions. Moreover, if one considers an asymptotic problem when $\varepsilon_i = \varepsilon_b$, Equation (17) erroneously yields $\varepsilon_{eff\ x,y} = 1.5\varepsilon_b$ suggesting that the solution of the Maxwell Garnett formula is not asymptotically convergent to a single-phased case.

Figure 4 presents a relative error of effective permittivity computed with Equation (18) against the iterative solution of Equation (12). It can be seen that the validity of the newly defined simplified and non-iterative formula applicable for 2D-oriented ellipsoidal inclusions buried in a host dielectric has been proven.

The author carried out several tests for different composite definitions with the 2D orientation of inclusions and, in all cases, Equation (18) fits precisely the corresponding results generated with an iterative solution of Equation (12).

Concluding, it has been proven that Equation (12), together with Equations (13)–(16), provide the correct representation of effective permittivity of a composite with a predefined distribution of ellipsoidal inclusions' orientations. Additionally, a new simplified formula for effective permittivity of a composite with a 2D distribution of inclusions has been given (see Equation (18)).

5. CONCLUSION

To the best of author's knowledge, this is the first formally and experimentally validated extension of the Maxwell Garnett mixing rule accounting for an arbitrary statistical orientation of ellipsoidal inclusions. In addition, a simplified formula dedicated to the modeling of thin composite layers with two-dimensional distribution of inclusions has been derived. The author believes that those ready-to-use formulae are very useful to the modeling of dilute mixtures with a process-dependent inclusions' orientation.

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