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MAXWELL GARNETT RULE FOR DIELECTRIC MIXTURES WITH STATISTICALLY DISTRIBUTED ORIENTATIONS OF INCLUSIONS

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Abstract—An analytical model of an effective permittivity of a composite taking into account statistically distributed orientations of inclusions in the form of prolate spheroids will be presented. In particular, this paper considers the normal Gaussian distribution for either zenith angle, or azimuth angle, or for both angles describing the orientation of inclusions. The model is an extension of the Maxwell Garnett (MG) mixing rule for multiphase mixtures. The resulting complex permittivity is a tensor in the general case. The formulation presented shows that the parameters of the distribution law for orientation of inclusions affect the frequency characteristics of the composites, and that it is possible to engineer the desirable frequency characteristics, if the distribution law is controlled.

1. INTRODUCTION

The problem of homogenization of different types of heterostructures from electromagnetic point of view has acquired much attention in the recent 10–15 years due to increased practical needs of design and application of novel composite materials with desirable frequency characteristics. Many papers have been published considering effective permittivity and/or permeability formulations for different kinds of composite media. Any linear particulate composite material can be considered as a particular case of a generalized multiphase bi-anisotropic medium with inclusions whose shape can be approximated by ellipsoids. A host material, as well as inclusions, can be dielectric, magnetic, magneto-dielectric, or conductive. These materials can

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be intrinsically isotropic or anisotropic. In addition, inclusions may possess anisotropy of shape (unless they are spherical), and in the general case, the axes of this anisotropy do not necessarily coincide with crystallographic or structural axes of their constituent material. Inclusions inside the host matrix may be arranged in an orderly manner, or they may be randomly dispersed. This spatial distribution may be homogeneously random, or form some inhomogeneities, like layers or clusters. As for orientations of inclusions, their main axes may be all aligned, or dispersed statistically, either with equal probability, or according to some distribution function.

This paper is not aimed at the review of all existing numerous theories and models of homogenization for all possible combinations of host/inclusions types, though it is worth mentioning a number of key publications, for example, [1–7], describing different homogenization procedures for the generalized bi-anisotropic mixtures. In particular, in these works, the most widely used mixing rules, such as Maxwell Garnett (MG) [8] and Bruggeman [9], are applied to treat mixtures with random, statistically equal positions of inclusions in 3D space. Extraction of effective electromagnetic properties for ellipsoidal inclusions with arbitrary, yet fixed deterministic orientation in 3D space, when all inclusions are aligned, has also been studied, both analytically and numerically [10–22]. However, the problem of statistical distribution of angles of orientation of ellipsoidal inclusions is lacking attention.

The present paper is aimed at the development of a simple for practical engineering applications model that will allow for calculating an effective permittivity for a composite with *statistically distributed angles of orientation* of ellipsoidal inclusions. The materials of both inclusions and matrix are assumed to be linear, isotropic, and homogeneous. The host is considered to be a dielectric, while inclusions may be either dielectric or conducting. Both host and inclusion materials may be frequency-dispersive. Anisotropy of inclusions is due only to their ellipsoidal shape. The concentration of inclusions is considered to be well below the corresponding percolation threshold. At the same time, 3D spatial distribution of all inclusions is considered to be homogeneous. With regards to wavelengths of the electromagnetic fields inside inclusions and host matrix, they are assumed to be long compared to the sizes of inclusions, so that quasistatic conditions within inclusions will be fulfilled. Then, within these limitations, the Maxwell Garnett rule may be applied.

The Maxwell Garnett (MG) mixing rule is a convenient model for predicting electromagnetic properties of composites containing two or more phases [6, 8, 23]. This model takes into account the frequency characteristics of materials comprising these composites [24–26]. In our paper [27], it has been shown that the MG formulation can be used at optical frequencies to predict frequency characteristics of composites containing conductive inclusions (nanorods). In [27], several subtle effects, such as the skin effect in conducting inclusions, the Drude frequency dependence of metals, the effect of the mean free path in small-size conducting inclusions, as well as the dimensional resonances in the nanorods, have been taken into account.

The statistical distribution of the aspect ratio of the inclusions also influences the electromagnetic (in particular, optical) characteristics of the composites. An analytical model that takes into account the statistical distribution of aspect ratios of the nanorods is presented in [28]. In [29], the two-dimensional statistical distribution of both aspect ratio and conductivity of inclusions have been considered, and some examples of computations for complex permittivity of composites at microwave frequencies have been given.

The objective of this paper is to extend the multiphase Maxwell Garnett (MG) mixing theory to the case of statistically distributed inclusion orientation. The necessity of having this model is dictated by practical scenarios when synthesizing composite materials with desirable dielectric responses. For example, in real composites, because of the technological processes, such as extrusion, the orientation of the nanorods typically is not uniformly distributed in three dimensions. There is always some bias in the angles of orientation of the inclusions. For this reason, it is important to include the statistical distribution of the angles of orientation in a composite model.

In many practical cases, to achieve higher volume loading of inclusions, higher packing density, and higher percolation threshold, it is favorable to have inclusions, such as fibers or rods, all aligned. However, it is difficult to achieve perfect alignment, and typically there is some "cone" of distribution around the most probable direction. Also, even in mixtures with nearly homogenous 3D random distribution of ellipsoidal inclusion orientation, there may be some slight statistical bias in orientation due to electrostatic interactions, or some kind of mechanical stress. All these examples demonstrate the necessity of taking into account distribution function models of orientation angles.

Section 2 of the present paper describes a mathematical model of the composite including the statistical distribution of inclusion orientation. Section 3 specifies effective permittivity tensors for some particular cases. The conclusions are summarized in Section 4.

2. MATHEMATICAL FORMULATION

2.1. Effective Permittivity Tensor for Oriented Inclusions

The objective of this section is to write the known Maxwell Garnett rule in a form convenient for further introduction of statistical distributions of angles of orientation of ellipsoidal inclusions. For this purpose, the multiphase Maxwell Garnett formulation is used. In this formulation, phases may differ by inclusion materials, and by the shapes of the inclusions. In addition, each phase of inclusions may have different distribution of spatial orientation.

Consider an individual dielectric ellipsoidal inclusion with permittivity ε_i in a base material with permittivity ε_b. Assume that both base and inclusion materials are linear, homogeneous, and crystallographically isotropic.

The polarizability tensor for an ellipsoidal particle is diagonal

$$\overleftrightarrow{\alpha} = \begin{pmatrix} \alpha_x & 0 & 0 \\ 0 & \alpha_y & 0 \\ 0 & 0 & \alpha_z \end{pmatrix}, \tag{1}$$

and has the following components [23]

$$\begin{aligned} \alpha_x &= \frac{4\pi}{3} c_x c_y c_z (\varepsilon_i - \varepsilon_b) \cdot \frac{\varepsilon_b}{\varepsilon_b + N_x(\varepsilon_i - \varepsilon_b)}; \\ \alpha_y &= \frac{4\pi}{3} c_x c_y c_z (\varepsilon_i - \varepsilon_b) \cdot \frac{\varepsilon_b}{\varepsilon_b + N_y(\varepsilon_i - \varepsilon_b)}; \\ \alpha_z &= \frac{4\pi}{3} c_x c_y c_z (\varepsilon_i - \varepsilon_b) \cdot \frac{\varepsilon_b}{\varepsilon_b + N_z(\varepsilon_i - \varepsilon_b)}, \end{aligned} \tag{2}$$

where $c_{x,y,z}$ denote the corresponding semi-axes of the ellipsoidal inclusion, and the corresponding depolarization form factors are N_x, N_y, and N_z. It is known that the relative permittivity tensor is related to the electric susceptibility tensor $\overleftrightarrow{\chi}^e$ through

$$\overleftrightarrow{\varepsilon}_i^e = \overleftrightarrow{I} + \overleftrightarrow{\chi}^e, \tag{3}$$

where $\overleftrightarrow{I} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$ is the unity tensor. The suseptibility tensor is

$$\overleftrightarrow{\chi}^e = \overleftrightarrow{\alpha}/\varepsilon_0. \tag{4}$$

The external susceptibility and permittivity tensors for an individual inclusion are both diagonal

$$\overleftrightarrow{\chi}^e = \begin{pmatrix} \chi_{xx} & 0 & 0 \\ 0 & \chi_{yy} & 0 \\ 0 & 0 & \chi_{zz} \end{pmatrix} \text{ and } \overleftrightarrow{\varepsilon}_i^e = \begin{pmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{pmatrix}. \tag{5}$$

Assuming that the inclusion particle is a spheroid with the semi-axes $c_x = c_y$, while c_z is different, the components of the tensor $\overleftrightarrow{\varepsilon}_i^e$ can be obtained from (2)–(4) as

$$\begin{aligned} \varepsilon_x = \varepsilon_y &= 1 + \frac{4\pi}{3\varepsilon_0} c_x^2 c_z (\varepsilon_i - \varepsilon_b) \cdot \frac{\varepsilon_b}{\varepsilon_b + N_x(\varepsilon_i - \varepsilon_b)}; \\ \varepsilon_z &= 1 + \frac{4\pi}{3\varepsilon_0} c_x^2 c_z (\varepsilon_i - \varepsilon_b) \cdot \frac{\varepsilon_b}{\varepsilon_b + N_z(\varepsilon_i - \varepsilon_b)}. \end{aligned} \tag{6}$$

It should be mentioned that for ellipsoidal inclusions the values c_x, c_y, and c_z should be positive finite numbers. Any disk or cylinder should be approximated by a corresponding closest ellipsoidal or spheroidal shape with finite axes, so that corresponding depolarization factors would be positive and finite as well.

If the inclusion is a rod with an aspect ratio $a = l/d$ (length/diameter), then its "semi-axes" are $c_x = c_y = d$, and $c_z = l$. The corresponding form factors in (6) can be calculated through their aspect ratio as, for example, in [23, 28].

Recalling that the volume fraction of n inclusions is defined as

$$f_i = \frac{nV_i}{V_\Sigma}, \tag{7}$$

where $V_\Sigma = 1$ is the total unit volume of the mixture, and that we have only one inclusion ($n = 1$), the volume fraction coincides with the volume of this inclusion. This is the volume of an individual spheroid that is very close to that of the corresponding cylinder

$$f_i = V_i \approx \frac{\pi \cdot d^2 l}{4}, \tag{8}$$

and the components (6) of the external permittivity tensor $\overleftrightarrow{\varepsilon}_i^e$ can be written as

$$\begin{aligned} \varepsilon_x = \varepsilon_y &= 1 + \frac{16 f_i}{3\varepsilon_0} (\varepsilon_i - \varepsilon_b) \cdot \frac{\varepsilon_b}{\varepsilon_b + N_x(\varepsilon_i - \varepsilon_b)}; \\ \varepsilon_z &= 1 + \frac{16 f_i}{3\varepsilon_0} (\varepsilon_i - \varepsilon_b) \cdot \frac{\varepsilon_b}{\varepsilon_b + N_z(\varepsilon_i - \varepsilon_b)}. \end{aligned} \tag{9}$$

The tensors $\overleftrightarrow{\chi}^e$ and $\overleftrightarrow{\varepsilon}_i^e$ are defined in the coordinate system (xyz) so that the major axis of the ellipsoid coincides with the axis z. However, for a mixture of inclusions, it is convenient to use a different coordinate system, where the external electric field is, for example, polarized in the $+z$-direction: $\vec{E}_e = E_e \hat{z}$, while the major axis of the inclusion is along the z' direction of the rotated coordinate system $(x'y'z')$, as is shown in Figure 1. Then, to rewrite the tensor $\overleftrightarrow{\varepsilon}_i^e$ in this new coordinate system $(x'y'z')$, one must accomplish rotation on the angles θ and φ, which are the angles between the major ellipsoid axis and the positive direction $+z$ along the axis z. In the coordinate system $(x'y'z')$, the new external permittivity tensor is

$$\overleftrightarrow{\varepsilon}_i^{new} = A_{rot} \cdot \overleftrightarrow{\varepsilon}_i^{e} \cdot A_{rot}^{-1}, \tag{10}$$

where A_{rot} is the rotation operator matrix, and A_{rot}^{-1} is its inverse matrix. The resultant new tensor of the effective permittivity can be expressed in a compact form as

$$\overleftrightarrow{\varepsilon}_i^{new} = \varepsilon_x \overleftrightarrow{I} + (\varepsilon_z - \varepsilon_x)\overleftrightarrow{W}, \tag{11}$$

where the matrix associated with the transformation of the coordinate systems is

$$\overleftrightarrow{W} = \begin{pmatrix} \cos^2\varphi\sin^2\theta & \cos\varphi\sin\varphi\sin^2\theta & \cos\varphi\cos\theta\sin\theta \\ \cos\varphi\sin\varphi\sin^2\theta & \sin^2\varphi\sin^2\theta & \sin\varphi\cos\theta\sin\theta \\ \cos\varphi\cos\theta\sin\theta & \sin\varphi\cos\theta\sin\theta & \cos^2\theta \end{pmatrix}. \tag{12}$$

Then the corresponding components of the new external permittivity tensor can be substituted in the Maxwell Garnett mixing formula. A tensor that will be used in the Maxwell Garnett formulation is analogous to the coefficient η_i in the isotropic case, as in [28],

$$\overleftrightarrow{\eta}_i = \frac{\overleftrightarrow{\varepsilon}_i^{new}}{\varepsilon_b} - \overleftrightarrow{I}. \tag{13}$$

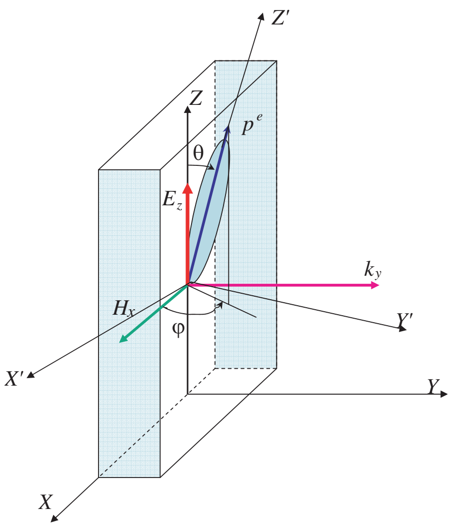

Figure 1. An individual electric dipole orientation with respect to the vectors of the plane wave normally incident upon the composite layer.

This tensor can be also represented in the following form through the components of the external permittivity tensor of inclusions as

$$\overleftrightarrow{\eta}_i = \left(\frac{\varepsilon_x}{\varepsilon_b} - 1\right) \cdot \overleftrightarrow{I} + \left(\frac{\varepsilon_z - \varepsilon_x}{\varepsilon_b}\right) \cdot \overleftrightarrow{W}, \tag{14}$$

or

$$\overleftrightarrow{\eta}_i = \eta_i \cdot \overleftrightarrow{I} + \beta_i \cdot \overleftrightarrow{W}, \tag{15}$$

where

$$\eta_i = \frac{\varepsilon_x}{\varepsilon_b} - 1 \tag{16}$$

and

$$\beta_i = \frac{\varepsilon_z - \varepsilon_x}{\varepsilon_b} \tag{17}$$

are scalar coefficients depending on the dielectric contrast of the inclusion with respect to the base material background.

The tensor $\overleftrightarrow{\eta}_i$ can be written then as

$$\overleftrightarrow{\eta}_i = \begin{pmatrix} \eta_i + \beta_i \cos^2\varphi \sin^2\theta & \beta_i \cos\varphi \sin\varphi \sin^2\theta & \beta_i \cos\varphi \cos\theta \sin\theta \\ \beta_i \cos\varphi \sin\varphi \sin^2\theta & \eta_i + \beta_i \sin^2\varphi \sin^2\theta & \beta_i \sin\varphi \cos\theta \sin\theta \\ \beta_i \cos\varphi \cos\theta \sin\theta & \beta_i \sin\varphi \cos\theta \sin\theta & \eta_i + \beta_i \cos^2\theta \end{pmatrix}. \tag{18}$$

Let us represent the Maxwell Garnett permittivity of isotropic multiphase mixtures [5, 24] in the following form

$$\varepsilon_{ef} = \varepsilon_b + \frac{\frac{1}{3}\sum\limits_{i=1}^{n}\left\{\varepsilon_b f_i \eta_i \sum\limits_{k=1}^{3} \frac{1}{1+N_{ik}\eta_i}\right\}}{1 - \frac{1}{3}\sum\limits_{i=1}^{n}\left\{f_i \eta_i \sum\limits_{k=1}^{3} \frac{N_{ik}}{1+N_{ik}\eta_i}\right\}}, \tag{19}$$

where N_{ik} are the corresponding depolarization factors for inclusions of the i-th type, and indices $k = 1, 2, 3$ correspond now to indices x, y, z for depolarization factors.

Quite formally, it is valid to substitute scalar values (ε_i, η_i, and unity) in (19) by their tensors analogs ($\overleftrightarrow{\varepsilon}_i^{new}$, $\overleftrightarrow{\eta}_i$, and $\overleftrightarrow{I}$). Denominators of fractions in tensor form should be substituted by multiplication by the corresponding inverse matrices.

$$\begin{aligned} \overleftrightarrow{\varepsilon}_{ef} \;=\; & \varepsilon_b \overleftrightarrow{I} + \frac{1}{3}\sum_{i=1}^{N} \varepsilon_b f_i \left[\sum_{k=1}^{3} \overleftrightarrow{\eta}_i \cdot \left(\overleftrightarrow{I} + \overleftrightarrow{\eta}_i N_{ik}\right)^{-1}\right] \\ & \cdot \left\{\overleftrightarrow{I} - \frac{1}{3}\sum_{i=1}^{N} f_i \left[\sum_{k=1}^{3} N_{ik} \overleftrightarrow{\eta}_i \cdot \left(\overleftrightarrow{I} + \overleftrightarrow{\eta}_i N_{ik}\right)^{-1}\right]\right\}^{-1}. \end{aligned} \tag{20}$$

If there is a diphasic mixture, then the summation $\sum_{i=1}^{N}$ is omitted, and (20) simplifies to

$$\overleftrightarrow{\varepsilon}_{ef} = \varepsilon_b \overleftrightarrow{I} + \frac{1}{3}\varepsilon_b f_i \sum_{k=1}^{3} \overleftrightarrow{\eta}_i \cdot \left(\overleftrightarrow{I} + \overleftrightarrow{\eta}_i N_{ik}\right)^{-1} \cdot \left[\overleftrightarrow{I} - \frac{1}{3} f_i \sum_{k=1}^{3} N_{ik} \overleftrightarrow{\eta}_i \cdot \left(\overleftrightarrow{I} + \overleftrightarrow{\eta}_i N_{ik}\right)^{-1}\right]^{-1}. \quad (21)$$

This is a 3×3-component tensor in the general case, since it is determined by the 3×3-component tensor $\overleftrightarrow{W}$, which depends on the angles θ and φ. The angle dependence is contained within the components of the tensor $\overleftrightarrow{\eta}_i$ (18). Herein, both θ and φ are statistically distributed. Assume that they are statistically independent of each other, and then their distributions can be considered separately.

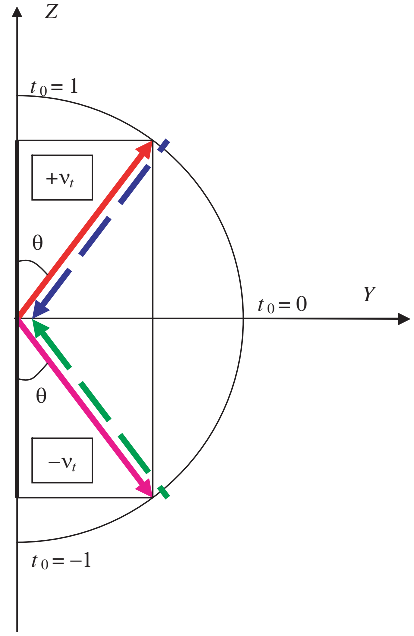

Figure 2. Diagram for calculating probability density of orientation with respect to the angle θ.

2.2. Inclusion Orientation Statistics

The model in this section takes into account the statistical distribution of the orientation of inclusions.

Some attempts to take into account the distribution of angles of orientation of the dipole moments in composites were done in [30–33]. In these publications, the dipole moments were magnetic moments, associated with high-anisotropy hexagonal ferrite single-domain independent particles. These magnetic moments were distributed arbitrarily in the range of angles $0 \leq \theta \leq 180°$. The total magnetic susceptibility of the composite depends on the relative number of particles in each of two non-interacting sets of particles with opposite preliminary magnetization. The probability density is comprised of two partial probabilities, corresponding to two oppositely directed sets of magnetic dipoles.

Herein, let us use a similar approach to analyze orientations of electric dipoles, corresponding to dielectric (or conducting) inclusions in the form of prolate spheroids. The effective permittivity that takes into account the statistical distribution of inclusion orientation will obviously be a tensor, since the presence of any preferred orientation leads to anisotropy of the material properties.

First, consider the dipole's orientation with respect to the angle θ, as is shown in Figure 2. It is convenient to introduce an auxiliary parameter $t = |\cos\theta|$. The inclusion dipoles are oriented within some limits $t \in [t_{\min} \ldots t_{\max}] \subset [0 \ldots 1]$. Let us separate inclusions depending on whether they are oriented upwards (in the 1st quadrant), or downwards (in the 4th quadrant), and introduce two partial probabilities: $q^+(t)$ for dipoles directed upwards, and $q^-(t)$ for dipoles directed downwards. We do not consider the other two quadrants −2nd and 3rd, because it is the spatial angle φ that is responsible for orientations in these quadrants. For the dipoles in the 1st quadrant, the angle $\theta \in [0 \ldots \pi/2]$ counts from the positive z direction. For the dipoles in the 4th quadrant, symmetrically, the angle $\theta \in [0 \ldots \pi/2]$ counts from the negative z direction. Next, let us introduce a coefficient $\nu_t = \{0, 1/2, 1\}$, which shows the dominating direction. If $\nu_t = 0$, the direction upwards is dominating, and if $\nu_t = 1$, the direction downwards is dominating. When $\nu_t = 1/2$, orientations in the 1st and 4th quadrants are equally probable. Then the total probability of orientation with respect to the angle θ is

$$p(t) = q^+(t) \cdot (1 - \nu_t) + q^-(t) \cdot \nu_t. \qquad (22)$$

Assume that the partial orientations are distributed according to normal Gaussian law

$$q^{+}(t) = \frac{1}{\sqrt{2\pi}\sigma_t^{+}} \exp\left(-\frac{t - t_0^{+}}{2(\sigma_t^{+})^2}\right);$$
$$q^{-}(t) = \frac{1}{\sqrt{2\pi}\sigma_t^{-}} \exp\left(-\frac{t - t_0^{-}}{2(\sigma_t^{-})^2}\right), \quad (23)$$

where $\sigma_t^{\pm}$ are the standard deviations (they may be different in the general case), and $t_0^{\pm}$ correspond to the mean angles of orientation $\theta_0^{\pm}$ (they also may be different in the general case, leading to asymmetry of distribution).

When the coefficient responsible for the positive/negative direction is $\nu_t = 1/2$, the probability density is calculated as an average of partial probabilities

$$p(t) = \frac{q^{+}(t) + q^{-}(t)}{2}. \quad (24)$$

This happens, when the orientations are close to the (xy) plane from the both positive and negative sides of the axis z. In the general case, this distribution may be asymmetrical with respect to the plane (xy). The inclusions are oriented with a symmetrical probability density close to the (xy) plane, only if $\sigma_t^{+} = \sigma_t^{-} = \sigma_t$; $t_0^{\pm} = 0$, and the corresponding mean angle is $\theta_0^{+} = \theta_0^{-} = \pi/2$. In this case, the probability density is

$$p(t) = \frac{1}{\sqrt{2\pi}\sigma_t} e^{-\frac{t}{2\sigma_t^2}}. \quad (25)$$

If the distribution of inclusion orientation is uniform within some the limits $t \in [t_{\min}^{\pm} \dots t_{\max}^{\pm}]$, then the partial probability densities are

$$q^{\pm}(t) = \frac{1}{\left|t_{\max}^{\pm} - t_{\min}^{\pm}\right|}. \quad (26)$$

Now let us consider the general case of an arbitrary bias in the angle φ counted counterclockwise starting from the positive direction of the axis x. Let us introduce another auxiliary variable $u = \cos\varphi$, where $u \in [-1 \dots 1]$. Since $u = \cos\varphi$ has the same values in the upper and lower half-planes of the plane (xy), it is reasonable to divide all the inclusions into two groups:

a) with the electric dipoles oriented in the 1st and 2nd quadrants — for $\varphi \in [0 \dots \pi]$, and
b) with the electric dipoles oriented in the 3rd and 4th quadrants — for $\varphi \in [\pi \dots 2\pi]$, and consider probabilities of orientation within these groups separately. The schematically these orientations are shown in Figure 3.

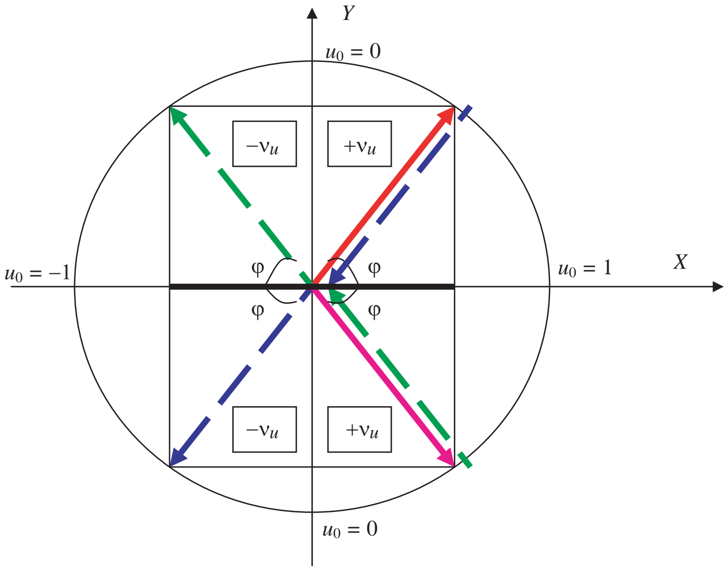

Figure 3. Diagram for calculating probability density of orientation with respect to the angle φ.

Let us introduce a parameter $\nu_u = \{0, 1/2, 1\}$, which shows a dominating direction — top half-plane or bottom half-plane on the plane (xy). If $\nu_u = 0$, the orientation is preferably in the upper half-plane, if $\nu_u = 1$, it is mainly in the lower half-plane, and if $\nu_u = 1/2$, the orientations are equally probable in both half-planes. Then the total probability of orientation with respect to the angle φ can be represented analogously to (22) as

$$p(u) = q^{+}(u) \cdot (1 - \nu_u) + q^{-}(u) \cdot \nu_u, \tag{27}$$

where the partial Gaussian probability densities are

$$\begin{aligned} q^{+}(u) &= \frac{1}{\sqrt{2\pi}\sigma_u^{+}} \exp\left(-\frac{u - u_0^{+}}{2(\sigma_u^{+})^2}\right); \\ q^{-}(u) &= \frac{1}{\sqrt{2\pi}\sigma_u^{-}} \exp\left(-\frac{u - u_0^{-}}{2(\sigma_u^{-})^2}\right), \end{aligned} \tag{28}$$

where $\sigma_u^{\pm}$ are the standard deviations, that may be different in the general case, and $u_0^{\pm}$ correspond to the mean angles of orientation $\varphi_0^{\pm}$ (they also may be different in the general case, leading to asymmetry of distribution).

When the coefficient responsible for the orientation in the upper/lower half-planes of (xy) is $\nu_u = 1/2$, the probability density is calculated as an average of two partial probabilities

$$p(u) = \frac{q^+(u) + q^-(u)}{2}. \tag{29}$$

In the general case, this distribution may be asymmetrical with respect to the x axis, depending on the values $\sigma_u^\pm$ and $u_0^\pm$. However, if $\nu_u = 1/2$, $\sigma_u^+ = \sigma_u^- = \sigma_u$, and $u_0^+ = u_0^- = 0$, this means that $\varphi_0^\pm = \pm\pi/2$, the majority of the dipoles are aligned along the y axis in the positive or negative directions. In this case, the probability density is

$$p(u) = \frac{1}{\sqrt{2\pi}\sigma_u} \exp\left(-\frac{u}{2\sigma_u^2}\right). \tag{30}$$

If $\nu_u = 1/2$, $\sigma_u^+ = \sigma_u^- = \sigma_u$, while $u_0^+ = 1$ and $u_0^- = -1$, this means that $\varphi_0^+ = 0$ and $\varphi_0^- = \pi$, so that the majority of dipoles are oriented close to the axis x, then the probability density is

$$p(u) = \frac{1}{\sqrt{2\pi}\sigma_u} \exp\left(-\frac{u}{2\sigma_u^2}\right) \cosh\left(\frac{1}{2\sigma_u^2}\right). \tag{31}$$

For any arbitrary $u_0^+ = -u_0^-$, the probability density is

$$p(u) = \frac{1}{\sqrt{2\pi}\sigma_u} \exp\left(-\frac{u}{2\sigma_u^2}\right) \cosh\left(\frac{u_0}{2\sigma_u^2}\right). \tag{32}$$

Consider the general case of arbitrarily biased statistical distributed of both angles θ and φ, shown schematically by a cone of orientations in Figure 4. The double probability density of orientation is

$$p(t, u) = p(t) \cdot p(u), \tag{33}$$

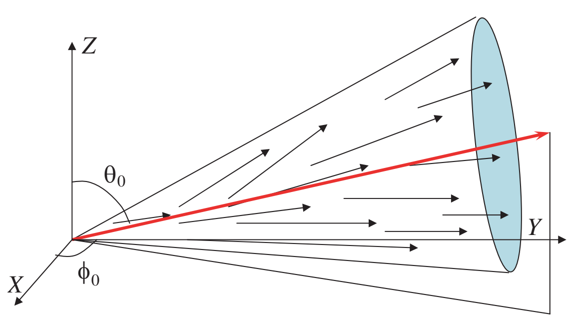

Figure 4. A cone of orientation of a dipole moments in a composite medium.

provided that the distributions with respect θ and φ are statistically independent, and the probabilities $p(t)$ and $p(u)$ are determined by (25) and (30), respectively.

3. EFFECTIVE PERMITTIVITY FOR SOME PARTICULAR CASES OF ORIENTED INCLUSIONS

The general tensor expression (20) for the effective permittivity of a composite depends on the angles of inclusion orientation, φ and θ, through the components of the tensor $\overleftrightarrow{\eta}_i$ (18). Let us consider statistics of orientations for some particular cases of inclusion alignment.

For definiteness, assume that the Poynting vector $\vec{\Pi}_{inc} = \Pi_{inc} \cdot \hat{y}$ (or, equivalently, the propagation vector $\vec{k}_{inc} = k_{inc} \cdot \hat{y}$.) is normally incident on the planar surface of the composite dielectric layer (xz). Also, assume that the electric field vector of this incident wave is polarized in the z-direction: $\vec{E}_{inc} = E_{inc} \cdot \hat{z}$. There are the following cases.

a) **Completely aligned case.** If all the inclusions are oriented in the same direction with the fixed angles θ and φ, then the resultant effective permittivity is determined by the general formulas (20) and (18), and no statistics are included.

b) **Isotropic case.** If all the inclusions within a homogeneous isotropic dielectric base are randomly (evenly) oriented in 3D space, then the integration over the angles $\theta \in [0 \ldots \pi]$ and $\varphi \in [0 \ldots 2\pi]$ of the angle-dependent components in (18) will yield zeros, and the tensor $\overleftrightarrow{\eta}_i$ will become diagonal with all three components equal

$$\overleftrightarrow{\eta}_i = \begin{pmatrix} \eta_i & 0 & 0 \\ 0 & \eta_i & 0 \\ 0 & 0 & \eta_i \end{pmatrix}, \tag{34}$$

that is, just a scalar value $\eta_i = \frac{\varepsilon_x}{\varepsilon_b} - 1$.

c) **Fixed angle $\boldsymbol{\theta}$, but arbitrary, homogeneously distributed, random angle $\boldsymbol{\varphi}$.** Then the tensor $\overleftrightarrow{\eta}_i$ becomes diagonal with two components being equal, and the third component being different:

$$\overleftrightarrow{\eta}_i = \begin{pmatrix} \eta_i & 0 & 0 \\ 0 & \eta_i & 0 \\ 0 & 0 & \eta_i + \beta_i \cos^2\theta \end{pmatrix}. \tag{35}$$

If the angle θ is statistically distributed within the limits $[\theta_{\min} \ldots \theta_{\max}]$, the probability density functions with respect to

θ (or equivalently, $x = |\cos\theta_i|$) should be considered, and the volume fraction function is then

$$f_i(x) = nV_i p(x), \tag{36}$$

where $p(x)$ is the orientation probability density function defined in a manner similar to (22), V_i is the volume of an inclusion (all the inclusions herein are assumed to be of the same volume), and n is the total number of inclusions per unit volume (concentration). The limits of integration $[x_{\min} \dots x_{\max}]$ depend on the range of the possible angles $[\theta_{\min} \dots \theta_{\max}]$. The standard deviation σ_x also determines the range of possible angles if this is a Gaussian distribution. Then the limits of integration will be approximately

$$\begin{aligned} x_{\min} &= x_0 - 3\sigma_x; \\ x_{\max} &= x_0 + 3\sigma_x. \end{aligned} \tag{37}$$

d) **In-plane random distribution.** If the angle θ is always equal only to 0, but the inclusions are oriented randomly in a uniform distribution with respect to the angle φ, then the $\overleftrightarrow{\eta}_i$ tensor is diagonal

$$\overleftrightarrow{\eta}_i = \begin{pmatrix} \eta_i & 0 & 0 \\ 0 & \eta_i & 0 \\ 0 & 0 & \eta_i + \beta_i \end{pmatrix}. \tag{38}$$

and there is no need of integration.

f) **Arbitrarily biased and statistically distributed angles $\boldsymbol{\varphi}$ and $\boldsymbol{\theta}$.** The double probability density of orientation is $p(t,u)$. Then the volume fraction of inclusions with orientation (θ,φ) in terms of (t,u) is

$$f_i(t,u) = nV_i p(t,u). \tag{39}$$

Hence, the effective permittivity (20) with statistically distributed inclusions orientations can be represented as

$$\overleftrightarrow{\varepsilon}_{ef} = \varepsilon_b \overleftrightarrow{I} + \frac{1}{3} \int_{t_{\min}}^{t_{\max}} \int_{u_{\min}}^{u_{\max}} \varepsilon_b f_i(t,u) \sum_{k=1}^{3} \overleftrightarrow{\eta}_i(t,u) \cdot \left(\overleftrightarrow{I} + \overleftrightarrow{\eta}_i(t,u) N_{ik}\right)^{-1}$$
$$\times \left[\overleftrightarrow{I} - \frac{1}{3} f_i(t,u) \sum_{k=1}^{3} N_{ik} \overleftrightarrow{\eta}_i(t,u) \cdot \left(\overleftrightarrow{I} + \overleftrightarrow{\eta}_i(t,u) N_{ik}\right)^{-1}\right]^{-1} du dt. \tag{40}$$

4. CONCLUSION

This analytical model allows for taking into the account the statistical distribution of orientations of elongated (prolate) ellipsoids (or spheroids) in a homogeneous and isotropic dielectric base. The inclusions are assumed to be also made of a homogeneous and isotropic dielectric. This model extends Maxwell Garnett multiphase formulations. Obviously, if there is some biased orientation of the spheroid major axis, the mixture is anisotropic with the effective permittivity described by a tensor. This anisotropy is induced by the anisotropy of shape of inclusions (different depolarization factors along the major and minor axes of the spheroid) and their orientation. Different particular cases of possible inclusion orientation are considered. In particular, the normal Gaussian distributions for either zenith angle θ, or azimuth angle φ, or for both angles θ and φ describing orientation of inclusions have been introduced in the multiphase Maxwell Garnett formulation for ellipsoidal inclusions. Though prolate spheroids were considered in this paper, the formulation is also valid for disk-like oblate spheroids. Then the zenith angle θ will be defined relative to the normal to the "disk" plane, and the azimuth angle φ will be considered in the "disk" plane.

A similar approach can be used for treating inclusions of isotropic form (spheres), but made of intrinsically crystallographically anisotropic materials, such as ferroelectric particles. Another case is a combination of crystallographic anisotropy of inclusions with anisotropy of shape, when the material is textured. Also, the base dielectric with anisotropic permittivity can be considered using the same approach.

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