Equivalent permittivity tensor in anisotropic random media

Stefano Giordano*

Department of Biophysical and Electronic Engineering, University of Genoa, Via Opera Pia 11 A, 16145 Genoa, Italy

Abstract

An explicit result for the equivalent (or effective) permittivity tensor is derived for dispersions of anisotropic (biaxial) particles embedded in an isotropic matrix. The overall medium has a positional disorder but an orientational order and it exhibits a uniaxial behaviour. Each crystal is partially oriented along a given director (preferential direction) and the degree of orientation is taken into account by a suitable order parameter. The equivalent permittivity tensor of the whole heterogeneous material is described in terms of the order parameter, the volume fraction of the dispersed phase, the principal values of the permittivity tensor of the crystals and the components of the director along which the crystals are preferentially aligned.
© 2005 Elsevier B.V. All rights reserved.

Keywords: Anisotropic media; Composite materials

1. Introduction

A widely investigated topic in material science is that of calculating the permittivity or other specific quantities of heterogeneous media starting from the knowledge of the properties of each medium composing the mixture and from the microstructure of the composite medium itself. We find in literature a large number of methods to evaluate, both at numerical and theoretical level, the effective permittivity of composed media, as a function of the permittivity of its homogeneous constituents and some stoichiometric parameter [1,2]. From the historical point of view, one of the most famous results is the Maxwell formula for a strongly diluted suspension of spheres [1–3]. An alternative model is provided by the differential method, which derives from the mixture characterisation approach used by Bruggeman [4,5]. A great number of works have been devoted to describe the relationship between microstructure and properties. In [6] a functional unifying approach has been applied to better understand the intrinsic mathematical properties of a general mixing formula. A fundamental result is given by Hashin-Shtrik-

man's variational analysis [7], which provides an upper and lower bound for composite materials, irrespective of the microstructure. In particular, for a two-phase material, these bounds are given by two expressions of the Maxwell type. Finally, a method to find the relation between the spatial correlation function of the dispersed component and the final properties of the material is derived from the Brown expansion [8]. In this work we analyse the dielectric behaviour of dispersions of anisotropic inclusions. In particular we take into account spherical anisotropic objects embedded in a homogeneous matrix. Because of the anisotropy of the embedded particles the most relevant aspect in the microstructure is the orientation of the spherical particles themselves. We take into account a partial orientational order described by an order parameter S, which has the following meaning. When $S = 0$, the orientation of the particles is completely random, generating an overall isotropic medium. When $S = 1$, all the particles are aligned along a given director allowing the formation of a uniaxial material. Finally, when $S = -1/2$, all the particles are lying randomly in planes perpendicular to the given director. In this work we take into account all the intermediate configurations between order and disorder with the aim to characterise a material with particles partially aligned. As we will explain later, the order parameter S can assume all the fractional values between

---

*Tel.: + 39 010 3532899; fax: + 39 010 3532753.

E-mail addresses: giorda@dibe.unige.it, stefgiord14@libero.it.

$-1/2$ and 1. The three cases $S = 0$, $S = 1$ and $S = -1/2$ are simply presented as representative values on a continuum of values. We present, as result, the effective permittivity tensor of such a material depending on S. This analysis has immediate application to the field of the liquid crystals. Actually, our microstructure describes a material positionally disordered, but orientationally ordered, which corresponds to a nematic phase in liquid crystals [9,10]. The level of ordering is reflected in the macroscopic properties. For example, from an optical point of view, we may observe that the optical axis is given by the direction of orientation of the single crystals.

2. Single anisotropic inclusion

We first consider a single anisotropic sphere (characterised by its permittivity tensor $\boldsymbol{\varepsilon}$ [11]) embedded in an isotropic medium with permittivity ε_0. If we expose the system to a uniform electric field $\bar{E}_0$, a perturbation to this field appears both inside and outside the sphere. We are interested in the electric field which is present inside the sphere after the application of the external field. In an arbitrary reference frame, we may evaluate the internal electric field $\bar{E}_s$ and the internal electric displacement $\bar{D}_s$ using [12]

$$\bar{E}_s = 3\varepsilon_0[\boldsymbol{\varepsilon} + 2\varepsilon_0\mathbf{I}]^{-1}\bar{E}_0 \quad \text{and} \quad \bar{D}_s = 3\varepsilon_0\boldsymbol{\varepsilon}[\boldsymbol{\varepsilon} + 2\varepsilon_0\mathbf{I}]^{-1}\bar{E}_0. \tag{1}$$

Here $\mathbf{I}$ represents the identity tensor of order three. These fields are uniform within the sphere volume, and determined by the exciting field and the anisotropic permittivity tensor. Note that the internal electrical field is not necessarily parallel to the external applied field. In this equation, the anisotropy is completely general. It covers not only uniaxial and biaxial materials, but also gyrotropic media in which the permittivity tensor contains an antisymmetric part. However, in the following we consider a symmetric permittivity tensor in order to apply diagonalisation by means of a suitable orthogonal matrix (rotation matrix $\mathbf{R}$):

$$\boldsymbol{\varepsilon} = \mathbf{R}\begin{bmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{bmatrix}\mathbf{R}^{-1} = \mathbf{R}\,\boldsymbol{\varepsilon}_d\mathbf{R}^{-1}, \tag{2}$$

where ε_x, ε_y, ε_z are the three principal permittivities of the anisotropic sphere; the rotation matrix $\mathbf{R}$ represents the angular position (orientation) of the sphere with respect to a reference frame with axis parallel to the principal directions of the permittivity tensor.

If we consider an arbitrary reference frame in the space, we may thus calculate the internal electric field and electric displacement using the following relations:

$$\bar{E}_s = 3\varepsilon_0[\mathbf{R}\,\boldsymbol{\varepsilon}_d\mathbf{R}^{-1} + 2\varepsilon_0\mathbf{I}]^{-1}\bar{E}_0$$
$$= 3\varepsilon_0\mathbf{R}[\boldsymbol{\varepsilon}_d + 2\varepsilon_0\mathbf{I}]^{-1}\mathbf{R}^{-1}\bar{E}_0, \tag{3}$$

$$\bar{D}_s = 3\varepsilon_0\mathbf{R}\,\boldsymbol{\varepsilon}_d\mathbf{R}^{-1}[\mathbf{R}\,\boldsymbol{\varepsilon}_d\mathbf{R}^{-1} + 2\varepsilon_0\mathbf{I}]^{-1}\bar{E}_0$$
$$= 3\varepsilon_0\mathbf{R}\,\boldsymbol{\varepsilon}_d[\boldsymbol{\varepsilon}_d + 2\varepsilon_0\mathbf{I}]^{-1}\mathbf{R}^{-1}\bar{E}_0. \tag{4}$$

In our scheme, the rotation matrix $\mathbf{R}$ is a random one describing the distribution of the orientations of the spheres embedded in the hosting isotropic medium. In order to simplify successive computations, we define $\mathbf{A}_d = [\boldsymbol{\varepsilon}_d + 2\varepsilon_0\mathbf{I}]^{-1}$. It follows that the average values of the electric field and the displacement vectors (over the sphere dispositions) are given by the relationships:

$$\langle \bar{E}_s \rangle = 3\varepsilon_0\langle \mathbf{R}\mathbf{A}_d\mathbf{R}^{-1} \rangle \bar{E}_0 \quad \text{and}$$
$$\langle \bar{D}_s \rangle = 3\varepsilon_0\langle \mathbf{R}\,\boldsymbol{\varepsilon}_d\mathbf{A}_d\mathbf{R}^{-1} \rangle \bar{E}_0. \tag{5}$$

We may now define the statistical distribution of the orientations to calculate the above-indicated average values. The entire material that we wish to characterise is positionally disordered, but partially orientationally ordered. This means that the centres of the spheres are randomly distributed inside the medium but the orientation of principal axis of the permittivity tensor of each particle follow some statistical rules. We consider a single anisotropic (in general biaxial) sphere with a reference frame aligned along the principal directions of the permittivity tensor. In this frame we consider a given unit vector $\bar{w}$ (director), which helps to define the orientational order (see Fig. 1). Thus, all the spheres are oriented, but do not generate a perfect alignment among all these directors. As we will explain later, we can define an order parameter (the average value of the second Legendre polynomial):

$$S = \left\langle \frac{3}{2}\cos^2\vartheta - \frac{1}{2} \right\rangle_\vartheta, \tag{6}$$

where ϑ is the angle each particle (each unit vector $\bar{w}$) makes with the preferential direction given by the ξ-axis of a global reference frame (the symbol $\langle\,\rangle_\vartheta$ represents the average value over the angle ϑ). The meaning of this parameter can be described as follows. If $S = 1$, the alignment is perfect ($\vartheta = 0$), if $S = 0$ the alignment is completely random and if $S = -1/2$ all the particles (the corresponding unit vectors $\bar{w}$) are lying randomly in planes perpendicular to the director ($\vartheta = \pi/2$). We take into account all the intermediate configurations between order and disorder with the aim to characterise a material with particles partially aligned with
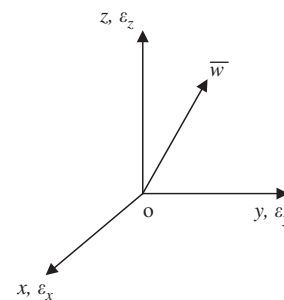

Fig. 1. Principal directions and permittivities of the spherical particles used in the composite material. An arbitrary unit vector $\bar{w}$ (director) is indicated and represents the privileged direction of alignment among the particles.

an arbitrary degree of order (see Fig. 2 for some examples of degree of order in the entire composite material). Once the direction of orientation is taken for a single sphere, it may be arbitrarily (randomly) rotated along the director. This means that we subdivide the complete rotation process in two elementary steps:

(1) first, we create non-perfect alignment by randomly fixing the angle ϑ ($\vartheta = 0$ means perfect alignment),
(2) secondly, we randomly rotate the sphere around the director, in order to establish a single preferential direction in the behaviour of the entire medium.

To perform the calculation, we explain in more detail the relevant phases. We begin with the first phase and for the given unit vector $\bar{w}$ we define the associated anti-symmetric matrix **w** as follows:

$$\bar{w} = \begin{bmatrix} w_x \\ w_y \\ w_z \end{bmatrix} \Rightarrow \mathbf{w} = \begin{bmatrix} 0 & -w_z & w_y \\ w_z & 0 & -w_x \\ -w_y & w_x & 0 \end{bmatrix}. \tag{7}$$

This definition will be used to better describe the generation of the tilting angle ϑ. To this aim, we take into account an arbitrary unit vector orthogonal to $\bar{w}$ and we name it $\bar{w}_\perp$; moreover, we complete the base with the unit vector $\bar{w} \wedge \bar{w}_\perp$. For example, we may assume the following components for the unit vector $\bar{w}_\perp$:

$$w_{\perp x} = \frac{w_y}{\sqrt{w_x^2 + w_y^2}}, \quad w_{\perp y} = -\frac{w_x}{\sqrt{w_x^2 + w_y^2}}, \quad w_{\perp z} = 0. \tag{8}$$

Therefore, on the plane formed by the vectors $\bar{w}_\perp$ and $\bar{w} \wedge \bar{w}_\perp$ we may take into account the arbitrary unit vector $\bar{v}$ orthogonal to $\bar{w}$, as sketched in Fig. 3. Thus, the following
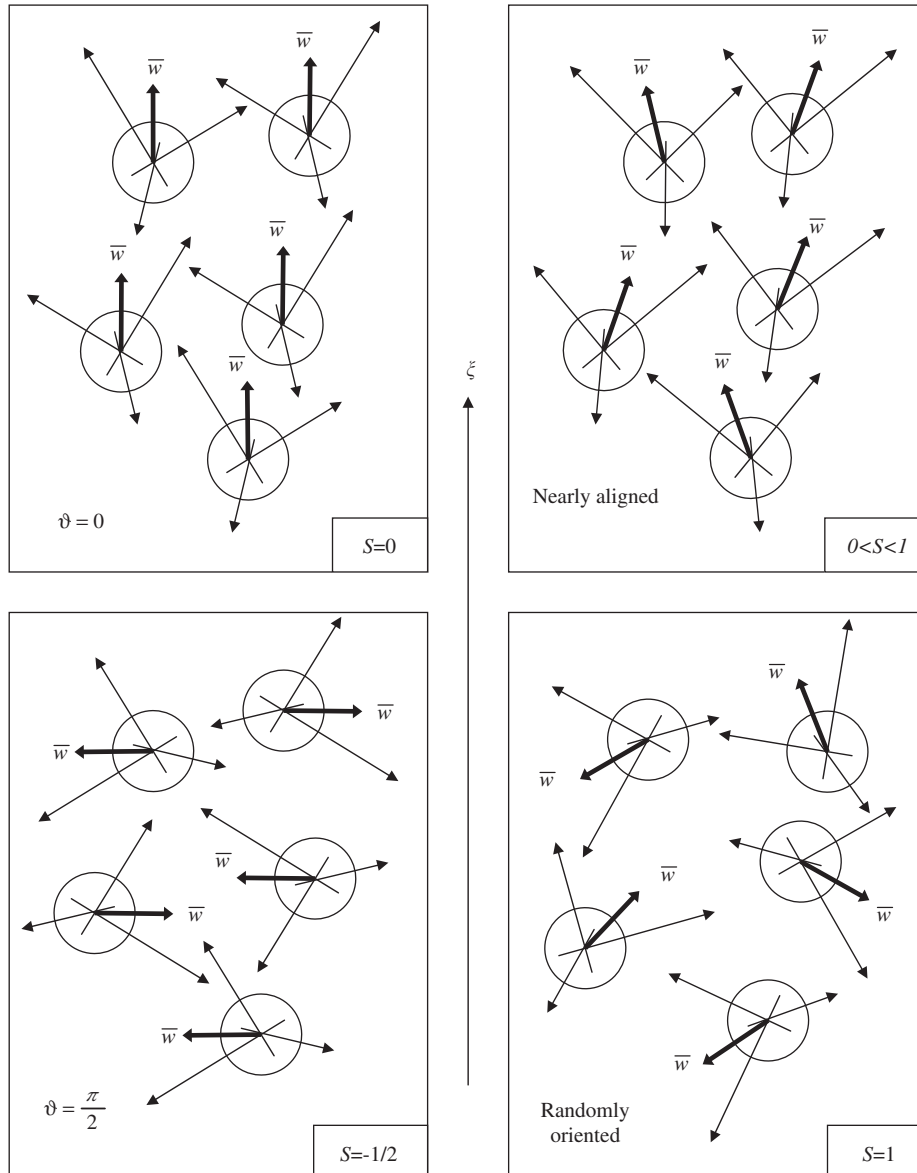

Fig. 2. Structure of a dispersion of pseudo-oriented anisotropic spheres. One can find some orientational distributions ranging from order to disorder. The two-phase material is described by the electric response of each phase, by the state of order and by the volume fraction of the inclusions.
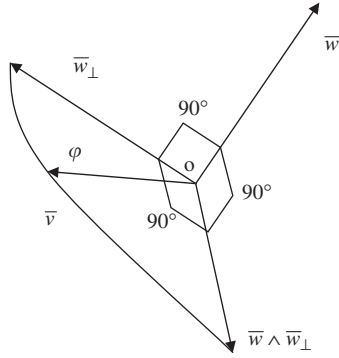
Fig. 3. Scheme of the vectors considered on the plane perpendicular to the director $\bar{w}$. Given the unit vector $\bar{w}$, we form a base by means of the unit vector $\bar{w}_\perp$ and the cross product $\bar{w} \wedge \bar{w}_\perp$; an arbitrary unit vector $\bar{v}$ perpendicular to $\bar{w}$ is defined as a linear combination of $\bar{w}_\perp$ and $\bar{w} \wedge \bar{w}_\perp$ and identified by using the angle φ.

relation holds:

$$\bar{v} = \cos\varphi\,\bar{w}_\perp + \sin\varphi\,\bar{w} \wedge \bar{w}_\perp, \tag{9}$$

where the angle φ is considered as a random variable uniformly distributed over the entire range $[0, 2\pi]$. According to the rule expressed in Eq. (7), we may define the anti-symmetric matrix $\mathbf{v}$ associated with the vector $\bar{v}$:

$$\mathbf{v} = \cos\varphi \begin{bmatrix} 0 & -w_{\perp z} & w_{\perp y} \\ w_{\perp z} & 0 & -w_{\perp x} \\ -w_{\perp y} & w_{\perp x} & 0 \end{bmatrix}$$
$$+ \sin\varphi \begin{bmatrix} 0 & -(w_x w_{\perp y} - w_y w_{\perp x}) & w_z w_{\perp x} - w_x w_{\perp z} \\ w_x w_{\perp y} - w_y w_{\perp x} & 0 & -(w_y w_{\perp z} - w_z w_{\perp y}) \\ -(w_z w_{\perp x} - w_x w_{\perp z}) & w_y w_{\perp z} - w_z w_{\perp y} & 0 \end{bmatrix}. \tag{10}$$

The first rotation of the orthonormal base is made around the unit vector $\bar{v}$ (which is stochastic for the presence of φ) with an angle ϑ; this rotation generates an inclination angle ϑ with respect to the director, as shown in Fig. 4.

The well-known Euler theorem states that each rotation may be described by a unit vector ($\bar{v}$ in our case) and an angle (ϑ in our case) and the relative rotation matrix is given by

$$\mathbf{R}_1 = \exp(\mathbf{v}\vartheta) = \mathbf{I} + \mathbf{v}\sin\vartheta + \mathbf{v}^2(1 - \cos\vartheta),$$
$$\mathbf{R}_1^{-1} = \exp(-\mathbf{v}\vartheta) = \mathbf{I} - \mathbf{v}\sin\vartheta + \mathbf{v}^2(1 - \cos\vartheta). \tag{11}$$

The first rotation thus fixes the orientation of the particles; now the spheres may rotate around the directors, and the relative angle of rotation ψ is considered to be randomly distributed over the range $[0, 2\pi]$ (see Fig. 5). The rotation matrix of this second phase is given by

$$\mathbf{R}_2 = \exp(\mathbf{w}\psi) = \mathbf{I} + \mathbf{w}\sin\psi + \mathbf{w}^2(1 - \cos\psi),$$
$$\mathbf{R}_2^{-1} = \exp(-\mathbf{w}\psi) = \mathbf{I} - \mathbf{w}\sin\psi + \mathbf{w}^2(1 - \cos\psi). \tag{12}$$

The combination of the two rotations defines a statistical medium where a privileged direction is given by the unit vector

$\bar{w}$; the orientational order is described by the random variable ϑ and in particular by the order parameter S (Eq. (6)), as we will verify later on. The statistical description of the orientation of each particle is given by the final rotation matrix $\mathbf{R} = \mathbf{R}_2 \mathbf{R}_1$, where (I) the unit vector $\bar{w}$ is a given and fixed director, (II) the unit vector $\bar{v}$ is given by Eqs. (8) and (9), (III) the angles φ and ψ are uniform random variables over the interval $[0, 2\pi]$, (IV) the angle ϑ is a random variable of which we know the expectation S (the order parameter). Under these hypotheses we may calculate the following mean value useful for evaluating the average electric field inside the particles (see Eq. (5)):

$$\langle \mathbf{R}\mathbf{A}_d\mathbf{R}^{-1} \rangle_{\varphi,\psi} = \langle \mathbf{R}_2\mathbf{R}_1\mathbf{A}_d\mathbf{R}_1^{-1}\mathbf{R}_2^{-1} \rangle_{\varphi,\psi}$$
$$= \frac{1}{4\pi^2} \int_0^{2\pi} \int_0^{2\pi} \mathbf{R}_2\mathbf{R}_1\mathbf{A}_d\mathbf{R}_1^{-1}\mathbf{R}_2^{-1}\, d\varphi\, d\psi$$
$$= \frac{1}{4\pi^2} \int_0^{2\pi} \int_0^{2\pi} [\mathbf{I} + \mathbf{w}\sin\psi + \mathbf{w}^2(1 - \cos\psi)]$$
$$\cdot [\mathbf{I} + \mathbf{v}\sin\vartheta + \mathbf{v}^2(1 - \cos\vartheta)]$$
$$\cdot \mathbf{A}_d[\mathbf{I} - \mathbf{v}\sin\vartheta + \mathbf{v}^2(1 - \cos\vartheta)]$$
$$\cdot [\mathbf{I} - \mathbf{w}\sin\psi + \mathbf{w}^2(1 - \cos\psi)]\, d\varphi\, d\psi. \tag{13}$$

Here the symbol $\langle \rangle_{\varphi,\psi}$ denotes "average over the uniformly distributed angles φ and ψ". A long but straightforward integration performed taking into account the definition of $\mathbf{v}$, given in Eqs. (10) and (8), allows us to write down a first important result

$$\langle \mathbf{R}\mathbf{A}_d\mathbf{R}^{-1} \rangle_{\varphi,\psi} = \left(\frac{3}{2}\cos^2\vartheta - \frac{1}{2} \right) \left[\mathbf{A}_d + \mathbf{A}_d\mathbf{w}^2 + \mathbf{w}^2\mathbf{A}_d \right.$$
$$\left. - \frac{1}{2}\mathbf{w}\mathbf{A}_d\mathbf{w} + \frac{3}{2}\mathbf{w}^2\mathbf{A}_d\mathbf{w}^2 \right]$$
$$+ \frac{1}{2}\sin^2\vartheta\, \mathrm{tr}(\mathbf{A}_d)\mathbf{I}. \tag{14}$$

Finally, performing the last averaging over the angle ϑ over the range $[0, \pi]$:

$$\langle \mathbf{R}\mathbf{A}_d\mathbf{R}^{-1} \rangle_{\varphi,\psi,\vartheta}$$
$$= S\left[\mathbf{A}_d + \mathbf{A}_d\mathbf{w}^2 + \mathbf{w}^2\mathbf{A}_d - \frac{1}{2}\mathbf{w}\mathbf{A}_d\mathbf{w} + \frac{3}{2}\mathbf{w}^2\mathbf{A}_d\mathbf{w}^2 \right]$$
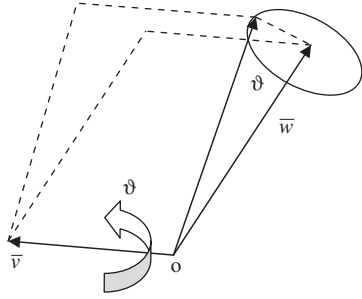$$+ \frac{1}{3}(1 - S)\, \mathrm{tr}(\mathbf{A}_d)\mathbf{I}, \tag{15}$$

Fig. 4. First rotation (described by the matrix $\mathbf{R}_1$) is obtained by means of rotation by an angle ϑ around the unit vector $\bar{v}$; correspondingly, the unit vector $\bar{w}$ is tilted at the same angle ϑ. In this way, partial alignment of the particles is obtained.
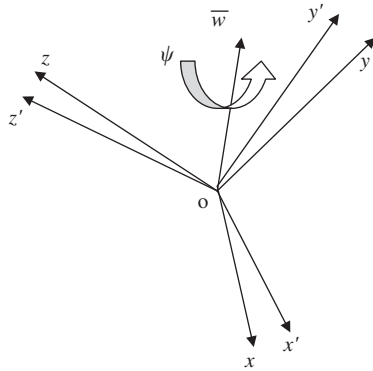

Fig. 5. Second rotation (described by the matrix $\mathbf{R}_2$) is obtained by performing a random rotation around the director $\bar{w}$ by an angle ψ. As a consequence, the particle is partially aligned along the director without restriction regarding the rotational position around the director.

where the order parameter S is defined by Eq. (6). It is interesting to note that if $S = 0$, we obtain the standard relation $\left\langle \mathbf{R}\mathbf{A}_d\mathbf{R}^{-1} \right\rangle_{\varphi,\psi,\vartheta} = \frac{1}{3}\mathrm{tr}(\mathbf{A}_d)\mathbf{I}$ which corresponds to isotropic orientation of the sphere, and if $S = 1$ we have the new result $\left\langle \mathbf{R}\mathbf{A}_d\mathbf{R}^{-1} \right\rangle_{\varphi,\psi,\vartheta} = \mathbf{A}_d + \mathbf{A}_d\mathbf{w}^2 + \mathbf{w}^2\mathbf{A}_d - \frac{1}{2}\mathbf{w}\mathbf{A}_d\mathbf{w} + \frac{3}{2}\mathbf{w}^2\mathbf{A}_d\mathbf{w}^2$ which corresponds to perfect alignment of the sphere to the director. Therefore, the average electric field inside an embedded sphere is given by

$$\left\langle \bar{E}_s \right\rangle = 3\varepsilon_0 S\left[\mathbf{A}_d + \mathbf{A}_d\mathbf{w}^2 + \mathbf{w}^2\mathbf{A}_d - \frac{1}{2}\mathbf{w}\mathbf{A}_d\mathbf{w} + \frac{3}{2}\mathbf{w}^2\mathbf{A}_d\mathbf{w}^2 \right]\bar{E}_0 + \varepsilon_0(1-S)\,\mathrm{tr}(\mathbf{A}_d)\bar{E}_0. \quad (16)$$

A similar relation holds on for the electrical displacement with $\varepsilon_d\mathbf{A}_d$ substituted for $\mathbf{A}_d$ (see Eq. (5)). The statistical distribution of orientations generates an overall material that is uniaxial from a macroscopic point of view, and the principal axis is given by the director $\bar{w}$. Therefore, we are interested in the average value of the electric field and the electric displacement when the applied field $\bar{E}_0$ is parallel or perpendicular to the unit vector $\bar{w}$. We start this computation taking into account the case with $\bar{E}_0$ parallel to $\bar{w}$. In this condition, we have $\bar{w} \wedge \bar{E}_0 = 0$ which means $\mathbf{w}\bar{E}_0 = 0$ (the matrix $\mathbf{w}$ acts on the components of the electric field in a

manner similar to a cross product $\bar{w}\wedge$). By applying this consideration to Eq. (16), we obtain the simplified relationship

$$\left\langle \bar{E}_s \right\rangle = 3\varepsilon_0 S[\mathbf{I} + \mathbf{w}^2]\mathbf{A}_d\bar{E}_0 + \varepsilon_0(1-S)\,\mathrm{tr}(\mathbf{A}_d)\bar{E}_0. \quad (17)$$

Taking into account the definition of $\mathbf{w}$ (Eq. (7)), it is easy to verify that the following formula is also true:

$$[\mathbf{I} + \mathbf{w}^2]_{ij} = w_i w_j \quad i,j = x, y, z. \quad (18)$$

Therefore, from Eq. (17) and from the relations $E_{0,k} = E_0 w_k$ (where $E_0 = |E_0|$), $\mathbf{A}_{d,j} = [\varepsilon_j + 2\varepsilon_0]^{-1}$, $[\mathbf{A}_d]_{jk} = \mathbf{A}_{d,j}\delta_{jk}$ we deduce that

$$\begin{aligned} \left\langle \bar{E}_s \right\rangle_i &= 3\varepsilon_0 S\sum_{j,k} w_i w_j \mathbf{A}_{d,j}\delta_{jk}E_{0,k} + \varepsilon_0(1-S)\sum_k \mathbf{A}_{d,k}E_{0,i} \\ &= 3\varepsilon_0 S\sum_k w_i w_k \mathbf{A}_{d,k}E_0 w_k + \varepsilon_0(1-S)\sum_k \mathbf{A}_{d,k}E_{0,i} \\ &= \varepsilon_0\sum_k \frac{3S w_k^2 + 1 - S}{\varepsilon_k + 2\varepsilon_0}E_{0,i}. \end{aligned} \quad (19)$$

This result and a similar one for the electric displacement can be summed up as follows for the case where $\bar{E}_0$ is parallel to $\bar{w}$:

$$\bar{E}_0\|\bar{w} \Rightarrow \begin{cases} \left\langle \bar{E}_s \right\rangle = \varepsilon_0\sum_j \dfrac{3S w_j^2 + 1 - S}{\varepsilon_j + 2\varepsilon_0}\bar{E}_0 = \alpha_\|\bar{E}_0, \\[2ex] \left\langle \bar{D}_s \right\rangle = \varepsilon_0\sum_j \dfrac{\varepsilon_j(3S w_j^2 + 1 - S)}{\varepsilon_j + 2\varepsilon_0}\bar{E}_0 = \beta_\|\bar{E}_0. \end{cases} \quad (20)$$

We can now begin the computation that takes into account the case of an applied electric field perpendicular to the director $\bar{w}$. In this case the following relation holds: $\bar{w} \wedge (\bar{w} \wedge \bar{E}_0) = -\bar{E}_0$ or, in other terms, $\mathbf{w}^2\bar{E}_0 = -\bar{E}_0$ (recall that the matrix $\mathbf{w}$ acts on the components of the electric field as works the cross product $\bar{w}\wedge$). Eq. (16), under this hypothesis, can be rewritten as

$$\begin{aligned} \left\langle \bar{E}_s \right\rangle &= 3\varepsilon_0 S\left[\mathbf{w}^2\mathbf{A}_d - \frac{1}{2}\mathbf{w}\mathbf{A}_d\mathbf{w} - \frac{3}{2}\mathbf{w}^2\mathbf{A}_d \right]\bar{E}_0 \\ &\quad + \varepsilon_0(1-S)\,\mathrm{tr}(\mathbf{A}_d)\bar{E}_0 \\ &= -\frac{3}{2}\varepsilon_0 S[\mathbf{w}^2\mathbf{A}_d + \mathbf{w}\mathbf{A}_d\mathbf{w}]\bar{E}_0 \\ &\quad + \varepsilon_0(1-S)\,\mathrm{tr}(\mathbf{A}_d)\bar{E}_0. \end{aligned} \quad (21)$$

A tedious but straightforward calculation allows us to observe that, if $\bar{E}_0 \perp \bar{w}$ (i.e. $\bar{E}_0 \cdot \bar{w} = 0$), the following relationship holds:

$$[\mathbf{w}^2\mathbf{A}_d + \mathbf{w}\mathbf{A}_d\mathbf{w}]\bar{E}_0 = -\sum_k \mathbf{A}_{d,k}(1 - w_k^2)\bar{E}_0. \quad (22)$$

Summing up, Eqs. (21) and (22) lead to the following relation for the internal average electric field and the analogous one for

the electric displacement:

$$\bar{E}_0 \perp \bar{w} \Rightarrow \begin{cases} \langle \bar{E}_s \rangle = \varepsilon_0 \sum_j \dfrac{\frac{3}{2}S(1-w_j^2)+1-S}{\varepsilon_j + 2\varepsilon_0} \bar{E}_0 = \alpha_\perp \bar{E}_0, \\[3mm] \langle \bar{D}_s \rangle = \varepsilon_0 \sum_j \dfrac{\varepsilon_j \left[\frac{3}{2}S(1-w_j^2)+1-S \right]}{\varepsilon_j + 2\varepsilon_0} \bar{E}_0 = \beta_\perp \bar{E}_0. \end{cases}$$

(23)

3. Effective permittivity tensor

Until now, we have analysed the average electric behaviour of a single spherical particle embedded in a random medium with a given statistical orientation in both principal directions, e.g. parallel and perpendicular to $\bar{w}$. We now consider a random medium where many particles, as described above (see Fig. 2), are embedded in an isotropic matrix having permittivity ε_0. The volume fraction of the dispersed phase will be indicated by c, where $0 < c < 1$, and at the beginning it will be considered very small (indicatively $c < 0.2$). Thus, we are dealing with a very diluted dispersion. This means that we do not take into account the interactions among the inclusions because of the very low concentration of spheres. For example, if we are dealing with N spheres of radius R in a cube of size l the volume fraction is given by $c = 4\pi R^3 N/(3l^3)$ and the low concentration condition is $N \ll 3l^3/(4\pi R^3)$. We now search for the electric behaviour of the overall dispersion of anisotropic spheres. Of course, from a macroscopic point of view, the permittivity tensor of such an overall medium will describe a uniaxial behaviour. The following considerations are valid both in the direction along $\bar{w}$ and in the directions orthogonal to $\bar{w}$. Specifically, we define ε ($\varepsilon_\parallel$ or $\varepsilon_\perp$) as the equivalent permittivity of the entire mixture by means of the relation $\langle \bar{D} \rangle = \varepsilon \langle \bar{E} \rangle$. To evaluate ε, we may compute the average value of the electric field and of displacement vector inside the random material. We also define V as the total volume of the mixture, V_e as the total volume of the embedded spheres, and V_o as the volume of the remaining space among the inclusions (so that $V = V_e \cup V_0$). The average value of the electric field is assumed to be the following:

$$\langle \bar{E} \rangle = c \langle \bar{E}_s \rangle + (1-c)\bar{E}_0 = (c\alpha + 1 - c)\bar{E}_0.$$

(24)

Here α and β stand for $\alpha_\parallel, \beta_\parallel$ or $\alpha_\perp, \beta_\perp$ depending on the direction under consideration. Eq. (24) means that, as we have above stated, we do not take into account the interactions among the inclusions because of the very low concentration of spheres; each sphere behaves as a single one in the entire space. Once more, we have approximately taken into account a uniform electric field $\bar{E}_0$ in the space outside the inclusions. This is the sole approximation introduced in this work and it corresponds to the classic approximation adopted to derive the standard Maxwell

mixing formula [1–3]. The average value of $\bar{D}(\bar{r}) = \varepsilon(\bar{r})\bar{E}(\bar{r})$ is evaluated as follows:

$$\begin{aligned} \langle \bar{D} \rangle &= \frac{1}{V} \int_V \varepsilon(\bar{r})\bar{E}(\bar{r})\, d\bar{r} \\ &= \frac{1}{V}\varepsilon_0 \int_{V_o} \bar{E}(\bar{r})\, d\bar{r} + \frac{1}{V} \int_{V_e} \bar{D}(\bar{r})\, d\bar{r} \\ &= \frac{1}{V}\varepsilon_0 \int_{V_o} \bar{E}(\bar{r})\, d\bar{r} + \frac{1}{V}\varepsilon_0 \int_{V_e} \bar{E}(\bar{r})\, d\bar{r} \\ &\quad + \frac{1}{V} \int_{V_e} \bar{D}(\bar{r})\, d\bar{r} - \frac{1}{V}\varepsilon_0 \int_{V_e} \bar{E}(\bar{r})\, d\bar{r} \\ &= \varepsilon_0 \langle \bar{E} \rangle + c\langle \bar{D}_s \rangle - c\varepsilon_0 \langle \bar{E}_s \rangle \\ &= \varepsilon_0 \langle \bar{E} \rangle + c(\beta - \varepsilon_0 \alpha)\bar{E}_0. \end{aligned}$$

(25)

Drawing a comparison between Eqs. (24) and (25) and recalling the definition of equivalent permittivity $\langle \bar{D} \rangle = \varepsilon \langle \bar{E} \rangle$, we may now find a complete expression that allows us to estimate the equivalent permittivity ε in the given direction and its first-order expansion with respect to the volume fraction c:

$$\varepsilon = \varepsilon_0 + c\frac{\beta - \varepsilon_0 \alpha}{1 - c + c\alpha} \cong \varepsilon_0 + c(\beta - \varepsilon_0 \alpha).$$

(26)

By using Eq. (20), we characterise the heterogeneous material along the director $\bar{w}$:

$$\begin{aligned} \varepsilon_\parallel &= \varepsilon_0 + c\frac{\varepsilon_0 \sum_j \frac{(\varepsilon_j - \varepsilon_0)(3Sw_j^2 + 1 - S)}{\varepsilon_j + 2\varepsilon_0}}{1 - c + c\varepsilon_0 \sum_j \frac{3Sw_j^2 + 1 - S}{\varepsilon_j + 2\varepsilon_0}} \\ &\cong \varepsilon_0 \left[1 + c \sum_j \frac{(\varepsilon_j - \varepsilon_0)(3Sw_j^2 + 1 - S)}{\varepsilon_j + 2\varepsilon_0} \right]. \end{aligned}$$

(27)

Similarly, by using Eq. (23), we find the permittivity in the directions orthogonal to $\bar{w}$:

$$\begin{aligned} \varepsilon_\perp &= \varepsilon_0 + c\frac{\varepsilon_0 \sum_j \frac{(\varepsilon_j - \varepsilon_0)[\frac{3}{2}S(1-w_j^2)+1-S]}{\varepsilon_j + 2\varepsilon_0}}{1 - c + c\varepsilon_0 \sum_j \frac{\frac{3}{2}S(1-w_j^2)+1-S}{\varepsilon_j + 2\varepsilon_0}} \\ &\cong \varepsilon_0 \left[1 + c \sum_j \frac{(\varepsilon_j - \varepsilon_0)[\frac{3}{2}S(1-w_j^2)+1-S]}{\varepsilon_j + 2\varepsilon_0} \right]. \end{aligned}$$

(28)

Eqs. (27) and (28) are the main results of this work and represent the principal permittivities of an anisotropic uniaxial material formed by biaxial spheres positionally disordered but orientationally ordered. These relationships have been expressed in terms of the order parameter S, the director $\bar{w}$, the principal values of the permittivity tensor of the constituent crystals and their corresponding volume fraction. As we will show later on, these expressions represent a generalisation of the well-known standard Maxwell mixing formula for isotropic spheres in isotropic matrix. Some particular limiting cases follow:

If the spheres are made of isotropic material, we have $\varepsilon_x = \varepsilon_y = \varepsilon_z = \varepsilon_m$ and, as expected, both Eqs. (27) and (28) reduce to the classical Maxwell formula, independently on

the value of S [1–3]:

$$\varepsilon_\parallel = \varepsilon_\perp = \varepsilon_0 + 3c \frac{\varepsilon_0(\varepsilon_m - \varepsilon_0)}{(1-c)(\varepsilon_m + 2\varepsilon_0) + 3c\varepsilon_0}$$

$$\cong \varepsilon_0 \left[1 + 3c \frac{\varepsilon_m - \varepsilon_0}{\varepsilon_m + 2\varepsilon_0} \right]. \tag{29}$$

In this latter case the orientation of the spheres does not affect the electric behaviour and, in effect, the components of the director $\bar{w}$ do not appear in the results.

If $S = 0$, the microstructure is completely disordered, and from a macroscopic point of view, the resulting material is exactly isotropic with permittivity given, in the dilute limit, by

$$\varepsilon_\parallel = \varepsilon_\perp \cong \varepsilon_0 \left[1 + c \sum_j \frac{\varepsilon_j - \varepsilon_0}{\varepsilon_j + 2\varepsilon_0} \right]. \tag{30}$$

It can be useful to write this formula in terms of the invariants of the permittivity tensor:

$$\varepsilon_\parallel = \varepsilon_\perp \cong \varepsilon_0 \left[1 + c \frac{3I_3 + 3\varepsilon_0 I_2 - 12\varepsilon_0^3}{I_3 + 2\varepsilon_0 I_2 + 4\varepsilon_0^2 I_1 + 8\varepsilon_0^3} \right], \tag{31}$$

where the three invariants are defined as follows:

$$I_1 = \mathrm{tr}\,\boldsymbol{\varepsilon},$$
$$I_2 = \frac{1}{2}[(\mathrm{tr}\,\boldsymbol{\varepsilon})^2 - \mathrm{tr}(\boldsymbol{\varepsilon}^2)],$$
$$I_3 = \det\boldsymbol{\varepsilon}. \tag{32}$$

Conversely, if $S = 1$ all the anisotropic particles are perfectly aligned along the director and we obtain from Eqs. (27) and (28) the simpler results

$$\varepsilon_\parallel = \varepsilon_0 + c \frac{\varepsilon_0 \sum_j \frac{3w_j^2(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0}}{1 - c + c\varepsilon_0 \sum_j \frac{3w_j^2}{\varepsilon_j + 2\varepsilon_0}}$$

$$\cong \varepsilon_0 \left[1 + c \sum_j \frac{3w_j^2(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0} \right], \tag{33}$$

$$\varepsilon_\perp = \varepsilon_0 + c \frac{\varepsilon_0 \frac{3}{2} \sum_j \frac{(1-w_j^2)(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0}}{1 - c + c\varepsilon_0 \frac{3}{2} \sum_j \frac{1-w_j^2}{\varepsilon_j + 2\varepsilon_0}}$$

$$\cong \varepsilon_0 \left[1 + c \frac{3}{2} \sum_j \frac{(1-w_j^2)(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0} \right]. \tag{34}$$

If $S = -1/2$, all the anisotropic particles are lying randomly on planes perpendicular to the director and we obtain from Eqs. (27) and (28) the results

$$\varepsilon_\parallel = \varepsilon_0 + c \frac{\varepsilon_0 \frac{3}{2} \sum_j \frac{(1-w_j^2)(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0}}{1 - c + c\varepsilon_0 \frac{3}{2} \sum_j \frac{1-w_j^2}{\varepsilon_j + 2\varepsilon_0}}$$

$$\cong \varepsilon_0 \left[1 + c \frac{3}{2} \sum_j \frac{(1-w_j^2)(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0} \right], \tag{35}$$

$$\varepsilon_\perp = \varepsilon_0 + c \frac{\varepsilon_0 \frac{3}{4} \sum_j \frac{(1+w_j^2)(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0}}{1 - c + c\varepsilon_0 \frac{3}{4} \sum_j \frac{1+w_j^2}{\varepsilon_j + 2\varepsilon_0}}$$

$$\cong \varepsilon_0 \left[1 + c \frac{3}{4} \sum_j \frac{(1+w_j^2)(\varepsilon_j - \varepsilon_0)}{\varepsilon_j + 2\varepsilon_0} \right]. \tag{36}$$

From previous results given by Eqs. (34) and (35), it is easy to observe the relationship that $\varepsilon_\perp(S = 1) = \varepsilon_\parallel(S = -1/2)$.

This relation derives from a most general property of our solutions: $\varepsilon_\perp(S = x) = \varepsilon_\parallel(S = -x/2)$, valid for any value of x, which immediately follows from Eqs. (27) and (28).

Finally, results often useful in practical applications are obtained by letting $\varepsilon_x = \varepsilon_y$ (uniaxial particles) partially aligned along the optical axis given by $w_x = 0$, $w_y = 0$ and $w_z = 1$. This case corresponds, for example, to a nematic liquid crystal where each uniaxial molecule is modelled by a spherical anisotropic particle. In any case, if we apply the general solutions given in Eqs. (27) and (28) to the present case we may obtain the final relationships:

$$\varepsilon_\parallel = \varepsilon_0 + c \frac{\varepsilon_0 \left[2\frac{\varepsilon_x - \varepsilon_0}{\varepsilon_x + 2\varepsilon_0}(1 - S) + \frac{\varepsilon_z - \varepsilon_0}{\varepsilon_z + 2\varepsilon_0}(1 + 2S) \right]}{1 - c + c\varepsilon_0 \left[2\frac{1-S}{\varepsilon_x + 2\varepsilon_0} + \frac{1+2S}{\varepsilon_z + 2\varepsilon_0} \right]}$$

$$\cong \varepsilon_0 \left[1 + 2c \frac{\varepsilon_x - \varepsilon_0}{\varepsilon_x + 2\varepsilon_0}(1 - S) + c \frac{\varepsilon_z - \varepsilon_0}{\varepsilon_z + 2\varepsilon_0}(1 + 2S) \right], \tag{37}$$

$$\varepsilon_\perp = \varepsilon_0 + c \frac{\varepsilon_0 \left[2\frac{\varepsilon_x - \varepsilon_0}{\varepsilon_x + 2\varepsilon_0}(1 + \frac{1}{2}S) + \frac{\varepsilon_z - \varepsilon_0}{\varepsilon_z + 2\varepsilon_0}(1 - S) \right]}{1 - c + c\varepsilon_0 \left[2\frac{1+S/2}{\varepsilon_x + 2\varepsilon_0} + \frac{1-S}{\varepsilon_z + 2\varepsilon_0} \right]}$$

$$\cong \varepsilon_0 \left[1 + 2c \frac{\varepsilon_x - \varepsilon_0}{\varepsilon_x + 2\varepsilon_0}\left(1 + \frac{1}{2}S\right) + c \frac{\varepsilon_z - \varepsilon_0}{\varepsilon_z + 2\varepsilon_0}(1 - S) \right]. \tag{38}$$

These particular results are in perfect agreement with those obtained in reference [13], which uses a complicated statistical technique from the standpoint of statistical continuum mechanics.

Fig. 6 shows plots of the equivalent permittivities $\varepsilon_\parallel$ and $\varepsilon_\perp$ versus the order parameter S for the case $-1/2 < S < 1$ using the parameters: $w_x = 0$, $w_y = 0$, $w_z = 1$, $\varepsilon_x = 15$, $\varepsilon_y = 15$, $\varepsilon_z = 3$ and $\varepsilon_0 = 1$. Six curves are presented for $\varepsilon_\parallel$ and for $\varepsilon_\perp$ which correspond to six different values of the volume fraction c starting from $c = 0$ and reaching the limiting value $c = 1$. One can observe the validity of the relation $\varepsilon_\perp(S = 1) = \varepsilon_\parallel(S = -1/2)$.

4. Differential effective medium theory

The differential effective medium theory, sometimes called the Bruggeman's procedure, is a method for finding a second mixture relationship by considering a first theory describing the composite material. This second theory is usually more efficient than the first one, even if the mixture is not strongly diluted. In Bruggeman's scheme [4] the initially low concentration is gradually increased by infinitesimal additions of the dispersed component [1,5].
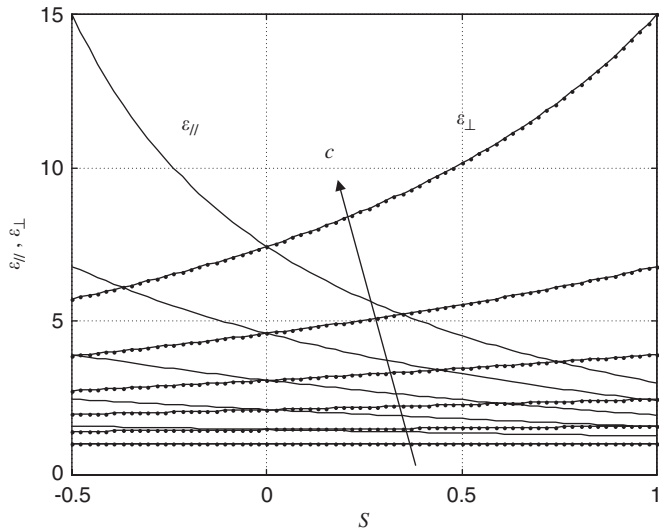
Fig. 6. Plots of the equivalent permittivities $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ given by Eqs. (37) and (38) versus the order parameter S ($-1/2 < S < 1$) with the values $w_x = 0$, $w_y = 0$, $w_z = 1$, $\varepsilon_x = 15$, $\varepsilon_y = 15$, $\varepsilon_z = 3$ and $\varepsilon_0 = 1$. Six curves are presented, and they correspond to six different values of the volume fraction c starting from $c = 0$ and reaching the limiting value $c = 1$.

In other words we may say that the differential procedure is a method that allows us to take into account approximately the interactions among the particles when the volume fraction is not very low. Of course, the limiting value $c = 1$ is never reached because the higher admissible values for the volume fraction correspond to uniform packing of touching spheres in the medium (for example $c = \pi/6$ for cubic packing of spheres). Typically, the mixing laws reported in literature and the results here presented are not correct for these extreme cases because of the strong interaction of the multipole moment terms among the spheres. Moreover, we may say that Eqs. (27) and (28) of the previous section are valid for values of c up to 0.1–0.2 and that results of the differential theory (described in this section) are valid for values of c up to 0.4–0.5. This restriction on the volume fraction of the mixture is very important but there are no direct restrictions on the size of the spheres embedded in the matrix. Anyway, we start from the simple mixing rule $\varepsilon = F(\varepsilon_0, \{e_k\}, c)$ where c is the volume fraction of embedded phase in the matrix (ε_0) and ε is the equivalent permittivity. In this work the function F is explicitly given in the final relations Eqs. (27) or (28). The set of parameters $\{e_k\}$ defines the microstructure and the material of the embedded particles. In our case, these parameters are given by w_x, w_y, w_z, ε_x, ε_y, ε_z plus the order parameter S. The standard differential procedure furnishes another relation describing the mixture for higher values of the volume fraction, and it is a solution of the following differential equation: $d\varepsilon/dc = (1/(1-c))$ $(\partial F(\varepsilon, \{e_k\}, c)/\partial c)|_{c=0}$ (see Refs. [1,4,5] for details). This equation, when the function F is given, defines a new function, which should better describe the mixture when it

is not strongly diluted. We may apply the method to the two expressions given in Eqs. (27) and (28), thereby obtaining the following differential equations:

$$\frac{d\varepsilon_{\parallel}}{dc} = \frac{1}{1-c} \frac{\partial \varepsilon_{\parallel}(c = 0, \varepsilon_0 = \varepsilon_{\parallel})}{\partial c},$$

$$\frac{d\varepsilon_{\perp}}{dc} = \frac{1}{1-c} \frac{\partial \varepsilon_{\perp}(c = 0, \varepsilon_0 = \varepsilon_{\perp})}{\partial c}. \tag{39}$$

Calculating the pertinent partial derivatives from Eqs. (27) and (28) result in

$$\left.\frac{\partial \varepsilon_{\parallel}}{\partial c}\right|_{c=0} = \varepsilon_0 \sum_j \frac{(\varepsilon_j - \varepsilon_0)(3Sw_j^2 + 1 - S)}{\varepsilon_j + 2\varepsilon_0},$$

$$\left.\frac{\partial \varepsilon_{\perp}}{\partial c}\right|_{c=0} = \varepsilon_0 \sum_j \frac{(\varepsilon_j - \varepsilon_0)[\frac{3}{2}S(1 - w_j^2) + 1 - S]}{\varepsilon_j + 2\varepsilon_0}. \tag{40}$$

We then can obtain the final system of ordinary differential equations representing the differential effective medium
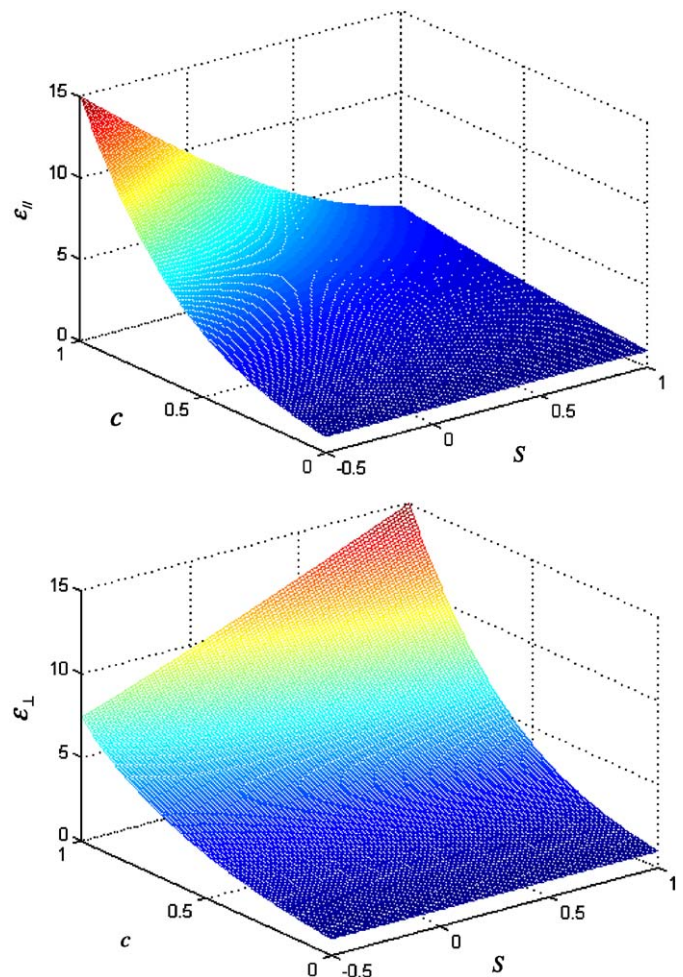

Fig. 7. Plots of the surfaces describing the behaviour of the equivalent permittivities $\varepsilon_{\parallel}$ (Fig. 7(a)) and $\varepsilon_{\perp}$ (Fig. 7(b)) given by the differential effective medium theory. The equivalent permittivities have been represented in terms of the order parameter S ($-1/2 < S < 1$) and the volume fraction c ($0 < c < 1$), with the same parameters used to obtain the plots of Fig. 6.

theory for the permittivity tensor in anisotropic random media:

$$
\begin{cases}
\dfrac{d\varepsilon_\parallel}{dc} = \dfrac{\varepsilon_\parallel}{1-c} \sum_j \dfrac{(\varepsilon_j - \varepsilon_\parallel)(3Sw_j^2 + 1 - S)}{\varepsilon_j + 2\varepsilon_\parallel}, \\[2mm]
\varepsilon_\parallel(c = 0) = \varepsilon_0, \\[2mm]
\dfrac{d\varepsilon_\perp}{dc} = \dfrac{\varepsilon_\perp}{1-c} \sum_j \dfrac{(\varepsilon_j - \varepsilon_\perp)[\frac{3}{2}S(1 - w_j^2) + 1 - S]}{\varepsilon_j + 2\varepsilon_\perp}, \\[2mm]
\varepsilon_\perp(c = 0) = \varepsilon_0.
\end{cases}
\tag{41}
$$

An example of a solution of such system is given in Fig. 7, where one can find the plots of the equivalent permittivities $\varepsilon_\parallel$ (Fig.7(a) and $\varepsilon_\perp$ (Fig. 7(b)) versus the order parameter S ($-1/2 < S < 1$) and the volume fraction c ($0 < c < 1$), with the same parameters used in Fig. 6: $w_x = 0$, $w_y = 0$, $w_z = 1$, $\varepsilon_x = 15$, $\varepsilon_y = 15$, $\varepsilon_z = 3$ and $\varepsilon_0 = 1$. These plots show two contours that describe the behaviour of the equivalent permittivities $\varepsilon_\parallel$ (Fig. 7(a)) and $\varepsilon_\perp$ (Fig. 7(b)) in terms of the parameters S and c. The differential system given in Eq. (41) could be solved in closed form because it involves the integration of rational functions. The explicit solutions are given by very complicated formulas, which provide no further understanding beyond the numerical solutions represented in Fig. 7. Hence, these relative expressions are not reported here.

5. Conclusions

We have set down the general equations for describing the tensor behaviour of an anisotropic random media in the dilute limit and by means of the differential scheme. The microstructure of the composite material is one of the most interesting, appearing, for example, in nematic liquid crystals and in other partially ordered physical systems. Distributions of anisotropic particles embedded in isotropic matrices have completely translational disorder but partial orientational order. The degree or orientation of the particles along a given director is described by a suitable order parameter. In both the dilute limit and in the differential scheme, the results are given by the equivalent permittivities of the overall uniaxial material; i.e. a permittivity along the director and a permittivity in the direction perpendicular to the director itself. These properties of the whole heterogeneous material are expressed in terms of the order parameter, the volume fraction of the dispersed phase, the principal values of the permittivity tensor of the crystals and the components of the director along which the crystals are preferentially aligned. These results have been found with a geometrical statistical approach and represent a strong generalisation of the standard mixing rules well known in the theory of composite materials.

References

[1] L.K.H. Van Beek, Dielectric behaviour of heterogeneous systems, Prog. Dielectric 7 (1967) 71–114.
[2] A. Sihvola, Electromagnetic Mixing Formulas and Applications, The Institution of Electrical Engineers, London, 1999.
[3] J.C. Maxwell, A Treatise on Electricity and Magnetism, Clarendon Press, Oxford, 1881.
[4] D.A.G. Bruggeman, Dielektrizitatskonstanten und Leitfahigkeiten der Mishkorper aus isotropen Substanzen, Ann. Phys. (Leipzig) 24 (1935) 636–664.
[5] S. Giordano, Effective medium theory for dispersions of dielectric ellipsoids, J. Electrostat. 58 (2003) 59–76.
[6] B. Bianco, M. Parodi, A unifying approach for obtaining closed-form expressions of mixtures permittivities, J. Electrostat. 15 (1984) 183–195.
[7] Z. Hashin, S. Shtrikman, A variational approach to the theory of the effective magnetic permeability of multiphase materials, J. Appl. Phys. 33 (1962) 3125–3131.
[8] W.F. Brown, Solid mixture permittivities, J. Chem. Phys. 23 (1955) 1514–1517.
[9] P.G. de Gennes, The Physics of Liquid Crystals, Clarendon Press, London, 1975.
[10] S. Chandrasekhar, Liquid Crystals, Oxford University Press, London, 1974.
[11] L.D. Landau, E.M. Lifshitz, Electrodynamics of Continuous Media, second ed., Pergamon Press, London, 1984.
[12] A. Cherepanov, A. Sihvola, Internal electric field of anisotropic and bianisotropic spheres, J. Electromagnetic Waves Appl. 10 (1996) 79–91.
[13] T. Nagatani, Effective permittivity in random anisotropic media, J. Appl. Phys. 51 (1980) 4944–4949.