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Effective medium theory for dispersions of dielectric ellipsoids

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Abstract

A material composed of a mixture of distinct homogeneous media can be considered as a homogeneous one at a sufficiently large observation scale. In literature, a widely dealt topic is that of calculating the overall permittivity of a dispersion of dielectric spheres in a homogeneous matrix: the well-known Maxwell formula solves the problem in the case of very diluted suspensions. Moreover, this relationship has been adapted, to yield correct results even if the dispersion in not strongly diluted, by means of the so-called Bruggeman's procedure. In this paper, we apply this technique to perform a complete study on the equivalent permittivity of a dispersion of ellipsoids. The obtained solutions allow us to evaluate the effects of the shape of the inclusions on the overall electric behaviour of the mixture. In particular we find explicit expressions, which give the equivalent permittivity of the medium in terms of the eccentricities of the embedded ellipsoids and of some stoichiometric parameters. The treatment is carried out both for aligned ellipsoidal inclusions and for randomly oriented ellipsoids. In particular, new explicit relationships have been derived for dispersions of generally shaped randomly oriented ellipsoids.

Keywords: Heterogeneous media; Dielectric homogenisation; Mixture theory

1. Introduction

A widely dealt topic concerning the physical behaviour of heterogeneous materials (mixtures) is that of calculating their permittivity starting from the knowledge of the

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permittivity of each medium composing the mixture as well as of the structural properties of the mixture itself (percentage of each medium, shapes and relative positions of the single parts of the various media). Clearly, it concerns with isotropic linear media, which combine to form linear mixtures. We find in literature a large number of approximate analytical expressions for the effective permittivity of composed media as a function of the permittivity of its homogeneous constituents and some stoichiometric parameter [1]. Each of these relationships should yield correct results for a particular kind of microstructure or, in other words, for a well defined morphology of the composite material.

From the historical point of view we review some formulas describing a mixture composed by two linear isotropic components; one of the most famous is the Maxwell formula for a strongly diluted suspension of spheres (three-dimensional case) [1,2]

$$c\frac{\varepsilon_1-\varepsilon_2}{2\varepsilon_1+\varepsilon_2}=\frac{\varepsilon_1-\bar{\varepsilon}}{2\varepsilon_1+\bar{\varepsilon}}, \tag{1}$$

where ε_1 is the permittivity of the suspending medium, ε_2 is the permittivity of the embedded spheres, c the volume fraction of the medium 2 and $\bar{\varepsilon}$ is the equivalent permittivity of the mixture. A similar equation holds true for a mixture of parallel circular cylinders (two-dimensional case) [1]

$$c\frac{\varepsilon_1-\varepsilon_2}{\varepsilon_1+\varepsilon_2}=\frac{\varepsilon_1-\bar{\varepsilon}}{\varepsilon_1+\bar{\varepsilon}}. \tag{2}$$

An alternative model is provided by the differential method, which derives from the mixture characterisation approach used by Bruggeman [3]. In this case, the relations should maintain the validity also for less diluted suspensions; the three-dimensional case (not diluted dispersions of spheres) leads to the formula

$$1-c=\frac{\varepsilon_2-\bar{\varepsilon}}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\bar{\varepsilon}}\right)^{1/3} \tag{3}$$

and the two-dimensional case (parallel cylindrical inclusions) to the following:

$$1-c=\frac{\varepsilon_2-\bar{\varepsilon}}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\bar{\varepsilon}}\right)^{1/2}. \tag{4}$$

This procedure is based on the following considerations: suppose that the effective permittivity of a composite medium is known to be $\bar{\varepsilon}$. Now, if a small additional volume of inclusions is embedded in the matrix, the change in the permittivity is approximated to be that which would arise if an infinitesimal volume of inclusions were added to a uniform, homogeneous matrix with permittivity $\bar{\varepsilon}$. This leads, in the simpler and most studied case, to a differential equation with solution given by Eq. (3) (in the 3D case) or Eq. (4) (in the 2D case) [3].

As we have already pointed out, Eqs. (1)–(4) represent approximations obtained working on mixtures of spheres (3D) or cylinders (2D) and thus they should be corrected and extended to be able to describe dispersions of ellipsoids.

The first papers dealing with mixtures of ellipsoids were written by Fricke [4,5] dealing with the electrical characterisation of inhomogeneous biological tissues containing spheroidal particles: he found out some explicit relationships that simply were an extension of the Maxwell formula to the case with ellipsoidal inclusions.

In current literature Maxwell's relation for spheres and Fricke's expressions for ellipsoids are the so-called Maxwell–Garnett Effective Medium Theory results [6,7]: both theories hold on under the hypothesis of the very low concentration of the dispersed component. Once more, we observe that the classical approximation for spheres, given by Eq. (1), has been derived in different contexts by Maxwell [2], Maxwell-Garnett [8], Wagner [9] and Bottcher [10] as a generalised Clausius–Mossotti–Lorentz–Lorenz relation.

In recent literature some applications of the Bruggeman procedure to mixtures of ellipsoids have been shown [11–13] in connection with the problem of characterising the dielectric response of water-saturated rocks. In their works the authors have shown that the Bruggeman method applied to this specific problem leads to results in good agreement with the empirical Archie's law [14], which describes the dependence of the d.c. conductivity of brine-saturated sedimentary rocks on porosity.

In general, the electrical (thermal, elastic and so on) properties of composite materials are strongly microstructure dependent. The relationships between microstructure and properties may be used for designing and improving materials, or conversely, for interpreting experimental data in terms of microstructural features. Ideally, the aim is to construct a theory that employs general microstructural information to make some accurate property predictions. A simpler goal is the provision of property for different class of microstructures. A great number of works have been devoted to describe the relationship between microstructure and properties: in [15] 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 the Hashin–Shtrikman's variational analysis [16], 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–Fricke 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 [17].

In this work, we briefly review the Bruggeman procedure to analyse the behaviour of a dispersion of dielectric ellipsoids obtaining a complete set of explicit results, which should be correct for any volume fraction of the inclusions. Dealing with embedding of ellipsoids we may consider two particular case: in the first one all the ellipsoids are aligned and thus they generate an anisotropic overall behaviour of the composite medium. This kind of mixture is discussed for example in [11]. In the other case all the embedded ellipsoids are randomly oriented and then the whole heterogeneous medium will be isotropic. This case is considered in [12] only when the embedded inclusions are ellipsoids of rotation. The major achievement of the present work is given by the explicit theory concerning the case of dispersions of generally shaped (three different axes) randomly oriented ellipsoids: some new mixing formulas have been derived for such kind of dispersion.

It must be underlined that from a merely mathematical standpoint, the problem of calculating the mixture permittivity is identical to a number of others, for instance to that regarding permeability (in a magnetostatic situation), conductivity (in d.c. condition), thermal conductivity (in a steady-state thermal regime) and so on. Therefore, each theoretical formula predicts the effective value of any thermal, magnetic or electrical specific quantities.

2. Characterisation of dispersions of aligned ellipsoids

The theory is based on the following result, which describes the behaviour of an ellipsoidal particle (ε_2) embedded in a homogeneous medium (ε_1). Let the axes of the ellipsoid be a_x, a_y and a_z (aligned with axes x, y, z of the reference frame) and let a uniform electrical field $\bar{E}_0 = \left(E_{0x}, E_{0y}, E_{0z}\right)$ applied to the structure. Then, according to Stratton [18] a uniform electrical field appears inside the ellipsoid and it can be computed as follows. We define the function

$$R(s) = \sqrt{\left(s + a_x^2\right)\left(s + a_y^2\right)\left(s + a_z^2\right)} \tag{5}$$

and the depolarisation factors along each axes

$$L_x = \frac{a_x a_y a_z}{2}\int_0^{+\infty}\frac{\mathrm{d}s}{\left(s + a_x^2\right)R(s)}, \quad L_y = \frac{a_x a_y a_z}{2}\int_0^{+\infty}\frac{\mathrm{d}s}{\left(s + a_y^2\right)R(s)},$$
$$L_z = \frac{a_x a_y a_z}{2}\int_0^{+\infty}\frac{\mathrm{d}s}{\left(s + a_z^2\right)R(s)}. \tag{6}$$

We may observe that $L_x + L_y + L_z = 1$. Therefore, the electrical field inside the ellipsoid is given, in components, by [18]

$$E_{ix} = \frac{E_{0x}}{1 + L_x(\varepsilon_2 - \varepsilon_1)/\varepsilon_1}, \quad E_{iy} = \frac{E_{0y}}{1 + L_y(\varepsilon_2 - \varepsilon_1)/\varepsilon_1},$$
$$E_{iz} = \frac{E_{0z}}{1 + L_z(\varepsilon_2 - \varepsilon_1)/\varepsilon_1}. \tag{7}$$

This is the main result that plays an essential role in the further development of the theory. Now, we are ready to consider a dispersion of aligned ellipsoids (ε_2) embedded in a homogeneous medium (ε_1): each inclusion has the axes a_x, a_y and a_z aligned with the axes x, y, and z of the reference frame, respectively (see Fig. 1). Moreover, let c be the volume fraction of the embedded ellipsoids. To begin, we consider a diluted dispersion ($c \ll 1$) and thus we may evaluate the average value of the electrical field over the mixture volume by means of the following relationships:

$$\langle E_x \rangle = cE_{ix} + (1 - c)E_{0x}, \quad \langle E_y \rangle = cE_{iy} + (1 - c)E_{0y},$$
$$\langle E_z \rangle = cE_{iz} + (1 - c)E_{0z}. \tag{8}$$

This means that we do not take into account the interactions among the inclusions because of the very low concentration: each little ellipsoid behaves as a single one in

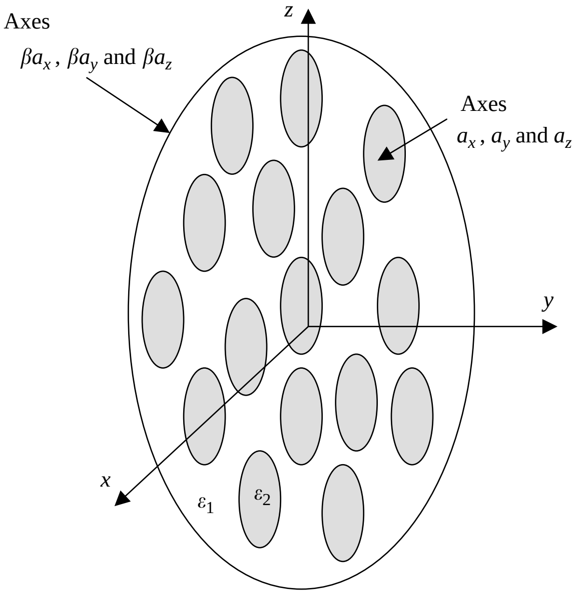

Fig. 1. Structure of a dispersion of aligned ellipsoids. The external surface of the mixture is a great ellipsoid with the same shape of the inclusions.

the whole space. Once more, to derive Eq. (8), we approximately take into account a uniform electrical field $\bar{E}_0 = \left(E_{0x}, E_{0y}, E_{0z}\right)$ in the space outside the inclusions. To define the mixture we are going to characterise, we consider a greater ellipsoid, which contains all the other ones. This ellipsoid represents the external surface of the composite materials. This ellipsoid has, by definition, the same shape of the inclusions and then the axes are given by βa_x, βa_y and βa_z, where β is a positive constant (it is aligned to the embedded ellipsoids, see Fig. 1). As one can simply verify, the depolarisation factors of this ellipsoid are the very same of each inclusion contained in the mixture. Moreover, we may observe that the overall behaviour of the mixture is anisotropic because of the alignment of the ellipsoidal particles. So, if we define the equivalent principal permittivity of the mixture along the axes x, y and z as ε_x, ε_y and ε_z, we may write down these expressions for the average value of the components of the electrical field inside the whole mixture

$$\langle E_x \rangle = \frac{E_{0x}}{1 + L_x(\varepsilon_x - \varepsilon_1)/\varepsilon_1}, \quad \langle E_y \rangle = \frac{E_{0y}}{1 + L_y(\varepsilon_y - \varepsilon_1)/\varepsilon_1},$$
$$\langle E_z \rangle = \frac{E_{0z}}{1 + L_z(\varepsilon_z - \varepsilon_1)/\varepsilon_1}. \tag{9}$$

These expressions are derived considering the whole mixture as a single inclusion in the whole space and then we have used the basic result given by Eq. (7). Now, by substituting Eq. (7) into Eq. (8) and by drawing a comparison with Eq. (9) we may find expressions for ε_x, ε_y and ε_z, which are the equivalent principal permittivities of the whole composite material

$$\begin{aligned}\varepsilon_j &= \varepsilon_1 + c(\varepsilon_2-\varepsilon_1)\frac{1}{1+(1-c)L_j(\varepsilon_2-\varepsilon_1)/\varepsilon_1} \\ &= \varepsilon_1 + c(\varepsilon_2-\varepsilon_1)\frac{1}{1+L_j(\varepsilon_2-\varepsilon_1)/\varepsilon_1} + O(c^2),\end{aligned} \tag{10}$$

where $j = x,\ y,\ z$. This result holds true only for very diluted dispersions of aligned ellipsoids. Eq. (10) has the same degree of approximation of Eqs. (1) and (2) (spheres and cylinders) and plays the same role in the development of the theory. Each of the three independent relations, which appears in Eq. (10), may be recast in the unified form $\varepsilon = F(\varepsilon_1, \varepsilon_2, c)$ where ε represents ε_x, ε_y or ε_z. We use this very simple form to review Bruggeman's procedure that is the method to find a second mixture relationship considering a first theory describing the composite material (actually function F). This second theory is usually more efficient than the first one even if the mixture is not strongly diluted. In Bruggeman's scheme the initial low concentration is gradually increased by infinitesimal additions of the dispersed component [3]. We start from $\varepsilon = F(\varepsilon_1, \varepsilon_2, c)$ for a mixture where c is the volume fraction of ellipsoids: we consider a unit volume of the mixture ($1\,\mathrm{m}^3$) and we add a little volume $\mathrm{d}c_0$ of inclusions. Therefore, we consider another mixture between a medium with permittivity ε (volume equals to $1\,\mathrm{m}^3$) and a second medium (ε_2) with volume $\mathrm{d}c_0$. In these conditions the volume fraction of the second medium will be $\mathrm{d}c_0/(1+\mathrm{d}c_0) \approx \mathrm{d}c_0$. So, using the original relation for the mixture we can write: $\varepsilon + \mathrm{d}\varepsilon = F(\varepsilon, \varepsilon_2, dc_0)$. In the final composite material, with the little added volume $\mathrm{d}c_0$, the matrix (ε_1) will have effective volume $1-c$ and the dispersed medium (ε_2) will have effective volume $c + \mathrm{d}c_0$.

The original volume fraction of the second medium is $c/1$ and the final one is $(c+\mathrm{d}c_0)/(1+\mathrm{d}c_0)$; so, it follows that the variation of the volume fraction of inclusions obtained by adding the little volume $\mathrm{d}c_0$ is simply given by $\mathrm{d}c = (c+\mathrm{d}c_0)/(1+\mathrm{d}c_0) - c/1 = \mathrm{d}c_0(1-c)/(1+\mathrm{d}c_0) \approx \mathrm{d}c_0(1-c)$.

Therefore, we have $\varepsilon + \mathrm{d}\varepsilon = F(\varepsilon, \varepsilon_2, \mathrm{d}c/(1-c))$. With a first-order expansion we simply obtain

$$\varepsilon + \mathrm{d}\varepsilon = F(\varepsilon, \varepsilon_2, 0) + \left.\frac{\partial F(\varepsilon, \varepsilon_2, c)}{\partial c}\right|_{c=0} \frac{\mathrm{d}c}{1-c}$$

and taking into account the obvious relation $\varepsilon = F(\varepsilon, \varepsilon_2, 0)$ we obtain the differential equation

$$\frac{\mathrm{d}\varepsilon}{\mathrm{d}c} = \frac{1}{1-c}\left.\frac{\partial F(\varepsilon, \varepsilon_2, c)}{\partial c}\right|_{c=0}.$$

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 three expressions given in Eq. (10) obtaining the following differential equations ($j = x,\ y,\ z$):

$$\frac{\mathrm{d}\varepsilon_j}{\mathrm{d}c} = \frac{1}{1-c}(\varepsilon_2 - \varepsilon_j)\frac{1}{1+L_j(\varepsilon_2-\varepsilon_j)/\varepsilon_j}. \tag{11}$$

These equations may be easily solved with the auxiliary conditions $\varepsilon_j(c=0)=\varepsilon_1(j=x,\ y,\ z)$ obtaining the results

$$1-c=\frac{\varepsilon_2-\varepsilon_j}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\varepsilon_j}\right)^{L_j}\quad (j=x,y,z). \tag{12}$$

These are the final expressions, which characterise a dispersion of aligned ellipsoids, obtained by means of Bruggeman's approach. The depolarisation factors are given by Eq. (6). We may derive some simplified version of this result in some limiting cases. First, we consider a case with $a_z\to\infty$: the ellipsoids degenerate to parallel elliptic cylinders. We may define the eccentricity of the elliptic base of these cylinders as $e=a_y/a_x$. So, the expressions for the depolarisation factors can be evaluated as follows:

$$\begin{aligned}
L_x&=\frac{e}{2}\int_0^{+\infty}\frac{\mathrm{d}\xi}{(\xi+1)^{3/2}(\xi+e^2)^{1/2}}=\frac{e}{e+1},\\
L_y&=\frac{e}{2}\int_0^{+\infty}\frac{\mathrm{d}\xi}{(\xi+e^2)^{3/2}(\xi+1)^{1/2}}=\frac{1}{e+1},\\
L_z&=0
\end{aligned}\tag{13}$$

and therefore, Eq. (12) yields the simplified results

$$\begin{aligned}
&1-c=\frac{\varepsilon_2-\varepsilon_x}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\varepsilon_x}\right)^{e/(e+1)},\quad 1-c=\frac{\varepsilon_2-\varepsilon_y}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\varepsilon_y}\right)^{1/(e+1)},\\
&\varepsilon_z=c\varepsilon_2+(1-c)\varepsilon_1.
\end{aligned}\tag{14}$$

The first two equations describe the principal permittivities in the directions (axes x and y) orthogonal to the cylinders and the third one defines the principal permittivity along the axes of the cylinders (z-axis). If $e=1$ the first two equations reduce, as expected, to Eq. (4). The third relation, as we expect, is an exact result describing a parallel connection of capacitors (the interfaces are aligned with the electrical field).

A second case deals with a mixture of inclusions shaped as ellipsoids of rotation; we consider $a_x=a_y$ and thus we define the eccentricity as $e=a_z/a_x=a_z/a_y$. The depolarisation factors may be computed in closed form as follows and the results depend on the shape of the ellipsoid; it is prolate (of ovary or elongated form) if $e>1$ and oblate (of planetary or flattened form) if $e<1$:

$$\begin{aligned}
L_x&=L_y=\frac{e}{2}\int_0^{+\infty}\frac{\mathrm{d}\xi}{(\xi+1)^2(\xi+e^2)^{1/2}}\\
&=\begin{cases}\dfrac{e}{4p^3}\left[2ep+\ln\dfrac{e-p}{e+p}\right] & \text{if } e>1,\\[2ex] \dfrac{e}{4q^3}\left[\pi-2eq-2\,\mathrm{arctg}\,\dfrac{e}{q}\right] & \text{if } e<1,\end{cases}
\end{aligned}$$

$$L_z = \frac{e}{2}\int_0^{+\infty} \frac{\mathrm{d}\xi}{(\xi+1)(\xi+e^2)^{3/2}} = \begin{cases} \dfrac{1}{2p^3}\left[e\ln\dfrac{e+p}{e-p} - 2p\right] & \text{if } e>1, \\ \dfrac{1}{2q^3}\left[2q - e\pi + 2e\,\mathrm{arctg}\,\dfrac{e}{q}\right] & \text{if } e<1, \end{cases} \tag{15}$$

where $p = \sqrt{e^2-1}$ and $q = \sqrt{1-e^2}$.

We may verify that $2L_x + L_z = 1$ for any value of e. Eq. (12) combined with Eq. (15) describes the complete behaviour of a dispersion of aligned ellipsoids of rotation (oblate or prolate).

For the sake of completeness, we show the complete expressions for the depolarisation factors in the case of generally shaped ellipsoids. The results have been expressed in terms of the elliptic integrals and have been derived under the assumptions: $0<a_x<a_y<a_z$, $0<e=a_x/a_y<1$ and $0<g=a_y/a_z<1$. The final expressions follow:

$$\begin{aligned} L_x &= \frac{1}{1-e^2} - \frac{e}{(1-e^2)\sqrt{1-e^2g^2}}E(v,q), \\ L_y &= \frac{e(1-e^2g^2)}{(1-e^2)(1-g^2)\sqrt{1-e^2g^2}}E(v,q) - \frac{eg^2}{(1-g^2)\sqrt{1-e^2g^2}}F(v,q) - \frac{e^2}{1-e^2}, \\ L_z &= \frac{eg^2}{(1-g^2)\sqrt{1-e^2g^2}}[F(v,q)-E(v,q)]. \end{aligned} \tag{16}$$

Here the quantities v and q are defined by

$$v = \text{arcsen}\sqrt{1-e^2g^2}, \quad q = \sqrt{\frac{1-g^2}{1-e^2g^2}} \tag{17}$$

and the elliptic integrals are defined below [19]

$$\begin{aligned} F(v,q) &= \int_0^v \frac{\mathrm{d}\alpha}{\sqrt{1-q^2\sin^2\alpha}} = \int_0^{\text{sen}\,v} \frac{\mathrm{d}x}{\sqrt{(1-x^2)(1-q^2x^2)}}, \\ E(v,q) &= \int_0^v \sqrt{1-q^2\sin^2\alpha}\,\mathrm{d}\alpha = \int_0^{\text{sen}\,v} \frac{\sqrt{1-q^2x^2}}{\sqrt{1-x^2}}\mathrm{d}x. \end{aligned} \tag{18}$$

Once again, we have $L_x + L_y + L_z = 1$. In Fig. 2 one can find the three plots of the depolarising factors versus the eccentricities e and g: it can be noted that for a sphere ($e=1$ and $g=1$) the relation $L_x = L_y = L_z = 1/3$ holds true. As one can see in Fig. 2, when $e\approx 1$ and $g\approx 1$, i.e. when we deal with spheroidal particles, the behaviour of the depolarising factors is quite linear and so it can be described by the following approximate relationships: $L_x \cong (11-4e-2g)/15$, $L_y \cong (2e+5-2g)/15$ and $L_z \cong (2e+4g-1)/15$. These expressions represent the equations of the tangent planes to the surfaces $L_x(e,g)$, $L_y(e,g)$ and $L_z(e,g)$ at the point $e=1$, $g=1$. They are

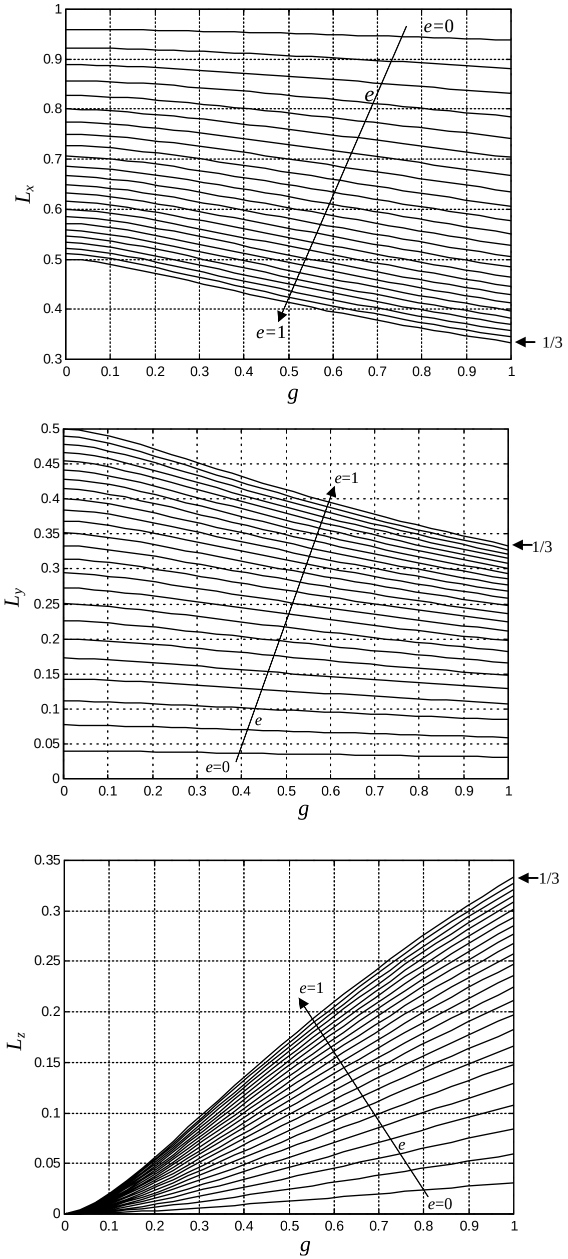

Fig. 2. Plots of the depolarisation factors as function of the eccentricities e and g. The three plots have been parameterised by the eccentricity e (we have used 25 uniformly spaced values for e ranging from 0 to 1).

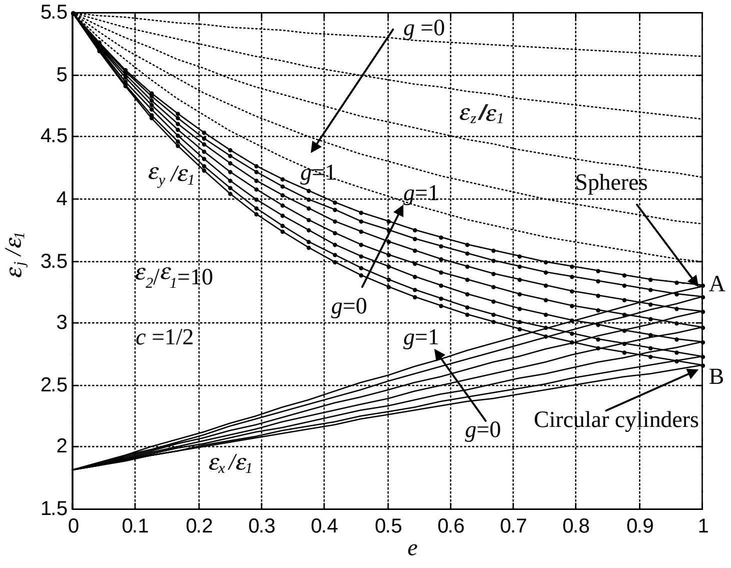

Fig. 3. Principal permittivities of a mixture of aligned ellipsoids (x-axis: continuous lines, y-axis: continuous dotted lines, z-axis: dotted lines). The three families of curves have been plotted versus the eccentricity e and parameterised by g. The values correspond to Eq. (12) and they have been computed with the assumptions $\varepsilon_2/\varepsilon_1 = 10$ and $c = 1/2$.

useful to evaluate the depolarisation factors of spheroidal particles (with high degree of accuracy for e, $g > 0.8$) without using elliptic integrals. Finally, we observe that Eq. (12) combined with Eq. (16) allows the characterisation of a mixture of aligned generally shaped ellipsoids. In Fig. 3 the behaviours of the three principal permittivities as function of the eccentricities e and g are shown. The plot concerns the case with $\varepsilon_2/\varepsilon_1 = 10$ and $c = 1/2$ and describes the three relative permittivities $\varepsilon_x/\varepsilon_1, \varepsilon_y/\varepsilon_1$ and $\varepsilon_z/\varepsilon_1$. In the point A the values of the eccentricities are $e = 1$ and $g = 1$ and thus it corresponds to a mixture of spheres: in such case the values of the depolarisation factors are $L_x = L_y = L_z = 1/3$ and the three principal permittivities are obviously given by Eq. (3). In the point B we have $e = 1$ and $g = 0$ and then it corresponds to a mixture of parallel circular cylinders: the values of the depolarisation factors are $L_x = L_y = 1/2$ and $L_z = 0$, the two permittivities along the axes x and y are given by Eq. (4) and the permittivity along the axes z is given by $\varepsilon_z = c\varepsilon_2 + (1 - c)\varepsilon_1$ (parallel of capacitors).

3. Characterisation of dispersions of randomly oriented ellipsoids

To begin, we are interested in the electrical behaviour of a single ellipsoidal inclusion (ε_2) arbitrarily oriented in the space and embedded in a homogeneous medium (ε_1). We define three unit vectors, which indicate the principal directions of the ellipsoids in the space: they are referred to as $\bar{n}_x, \bar{n}_y$ and $\bar{n}_z$ and they are aligned with the axes a_x, a_y and a_z of the ellipsoid, respectively. By using Eq. (7), we may compute the electrical field inside the inclusion, induced by a given external uniform electric field

$$\bar{E}_i = \frac{(\bar{E}_0 \cdot \bar{n}_x)\bar{n}_x}{1 + L_x(\varepsilon_2 - \varepsilon_1)/\varepsilon_1} + \frac{(\bar{E}_0 \cdot \bar{n}_y)\bar{n}_y}{1 + L_y(\varepsilon_2 - \varepsilon_1)/\varepsilon_1} + \frac{(\bar{E}_0 \cdot \bar{n}_z)\bar{n}_z}{1 + L_z(\varepsilon_2 - \varepsilon_1)/\varepsilon_1}. \tag{19}$$

This result simply derives from the sum of the three contributes to the electrical field along each axes. This expression may be written in explicit form, as follows:

$$E_{i,q} = \sum_k^{x,y,z} E_{0,k} \sum_j^{x,y,z} \frac{n_{j,k} n_{j,q}}{1 + L_j(\varepsilon_2 - \varepsilon_1)/\varepsilon_1}, \tag{20}$$

where $n_{j,k}$ is the kth component of the unit vector $\bar{n}_j (j = x, y, z)$.

For the following derivation, we are interested in the average value of the electrical field inside the ellipsoid over all the possible orientations of the ellipsoid itself and then we have to compute the average value of the quantity $n_{j,k}\ n_{j,q}$. Performing the integration over the unit sphere (by means of spherical coordinates) we obtain, after some straightforward computations

$$\langle n_{j,k} n_{j,q} \rangle = \tfrac{1}{3}\delta_{k,q} \quad \forall j. \tag{21}$$

Therefore, the average value of the electrical field (inside the randomly oriented inclusion), given by Eq. (20), may be written as

$$\langle E_{i,q} \rangle = \frac{E_{0,q}}{3} \sum_j^{x,y,z} \frac{1}{1 + L_j(\varepsilon_2 - \varepsilon_1)/\varepsilon_1}. \tag{22}$$

Now, we are ready to consider a mixture of randomly oriented ellipsoids. In Fig. 4 one can find the structure of the composite material: we consider a given number of randomly oriented ellipsoids (ε_2) embedded in a homogeneous matrix (ε_1). We may define, for example, the volume of the mixture by means of a sphere which contains all the ellipsoidal inclusions and which represents the external surface of the heterogeneous material. Once more, let c be the volume fraction of the embedded ellipsoids. The average value of the electrical field over the mixture (inside the sphere) is approximately computed by using Eq. (22)

$$\langle \bar{E} \rangle = (1 - c)\bar{E} + c\frac{\bar{E}_0}{3} \sum_j^{x,y,z} \frac{1}{1 + L_j(\varepsilon_2 - \varepsilon_1)/\varepsilon_1}. \tag{23}$$

Then, we define ε as the equivalent permittivity of the whole mixture (which is isotropic because of the randomness of the orientations) 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 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 ellipsoids and V_{o} as the volume of the remaining space among the inclusions (so that $V = V_{\mathrm{e}} \cup V_{\mathrm{o}}$). The

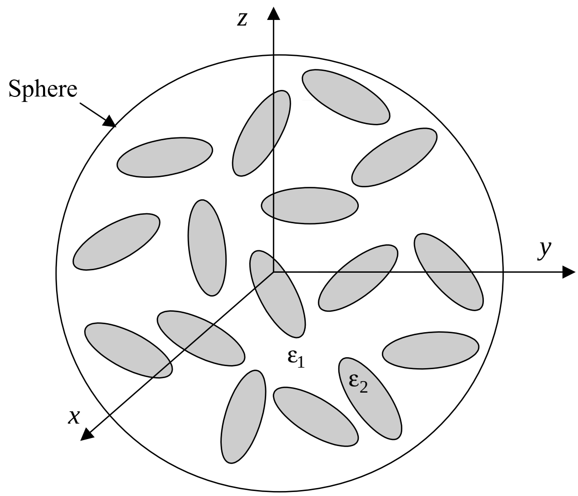

Fig. 4. Structure of a dispersion of randomly oriented ellipsoids. The external surface of the mixture is a sphere.

average value of $\bar{D}(\bar{r}) = \varepsilon(\bar{r})\bar{E}(\bar{r})$ is evaluated as follows:

$$\begin{aligned}\langle \bar{D} \rangle &= \frac{1}{V}\int_V \varepsilon(\bar{r})\bar{E}(\bar{r})\,\mathrm{d}\bar{r} = \frac{1}{V}\varepsilon_1 \int_{V_o} \bar{E}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\varepsilon_2 \int_{V_e} \bar{E}(\bar{r})\,\mathrm{d}\bar{r} \\ &= \frac{1}{V}\varepsilon_1 \int_{V_o} \bar{E}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\varepsilon_1 \int_{V_e} \bar{E}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\varepsilon_2 \int_{V_e} \bar{E}(\bar{r})\,\mathrm{d}\bar{r} - \frac{1}{V}\varepsilon_1 \int_{V_e} \bar{E}(\bar{r})\,\mathrm{d}\bar{r} \\ &= \varepsilon_1 \langle \bar{E} \rangle + c(\varepsilon_2 - \varepsilon_1)\langle \bar{E}_i \rangle. \end{aligned} \tag{24}$$

Drawing a comparison between Eqs. (22) and (24) we may find a complete expression, which allows us to estimate the equivalent permittivity ε and its first order expansion with respect to the volume fraction c:

$$\begin{aligned}\varepsilon &= \varepsilon_1 + \frac{\frac{1}{3}c(\varepsilon_2 - \varepsilon_1)\sum_j^{x,y,z} \varepsilon_1/(\varepsilon_1 + L_j(\varepsilon_2 - \varepsilon_1))}{1 + c\left[\frac{1}{3}\sum_j^{x,y,z} \varepsilon_1/(\varepsilon_1 + L_j(\varepsilon_2 - \varepsilon_1)) - 1\right]}, \\ &= \varepsilon_1 + \frac{1}{3}c\varepsilon_1(\varepsilon_2 - \varepsilon_1)\sum_j^{x,y,z} \frac{\varepsilon_1}{\varepsilon_1 + L_j(\varepsilon_2 - \varepsilon_1)} + \mathrm{O}(c^2). \end{aligned} \tag{25}$$

This result concerns the characterisation of a very diluted dispersion of randomly oriented ellipsoids with given shape (i.e. with fixed depolarisation factors L_j or eccentricities e and g). As before, to adapt this relationship to arbitrarily diluted composite materials we use Bruggeman's procedure, which leads to the following differential equation:

$$\frac{\mathrm{d}\varepsilon}{\mathrm{d}c} = \frac{1}{1-c}\varepsilon(\varepsilon_2 - \varepsilon)\frac{1}{3}\sum_j^{x,y,z} \frac{1}{\varepsilon + L_j(\varepsilon_2 - \varepsilon)}. \tag{26}$$

The solution of this equation depends on the values of the depolarisation factors showing the relationship between the overall permittivity and the shape of the ellipsoidal inclusions. We search for the solution in two particular cases: a dispersion of ellipsoids of rotation and a dispersion of generally shaped ellipsoids. In the first case we have $L_x = L_y$ and $2L_x + L_z = 1$ and thus only one factor completely defines the shape of the inclusions. If we use L_x as parameter, Eq. (26) reduces, after some straightforward computations, to the following one:

$$\frac{\mathrm{d}c}{1-c} = \left[\frac{1}{\varepsilon_2-\varepsilon} + \frac{3L_x(1-2L_x)}{(2-3L_x)\varepsilon} + \frac{2(3L_x-1)^2}{[(1+3L_x)\varepsilon+(2-3L_x)\varepsilon_2](2-3L_x)}\right]\mathrm{d}\varepsilon. \quad (27)$$

The integration of the above partial fraction expansion, with the condition $\varepsilon(c = 0) = \varepsilon_1$, yields the final result

$$1-c = \frac{\varepsilon_2-\varepsilon}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\varepsilon}\right)^{3L(1-2L)/(2-3L)} \times \left[\frac{(1+3L)\varepsilon_1+(2-3L)\varepsilon_2}{(1+3L)\varepsilon+(2-3L)\varepsilon_2}\right]^{2(3L-1)^2/(2-3L)(1+3L)}, \quad (28)$$

where $L = L_x$ is given by Eq. (15) and represents the depolarisation factor along the directions orthogonal to the principal axes of each inclusion. For convenience, we report here the complete expressions of L for prolate and oblate ellipsoidal particles

$$L = \begin{cases} \dfrac{e}{4p^3}\left[2ep + \ln\dfrac{e-p}{e+p}\right] & \text{if } e>1 \ \text{(prolate ellipsoids)}, \\ \dfrac{e}{4q^3}\left[\pi - 2eq - 2\,\mathrm{arctg}\,\dfrac{e}{q}\right] & \text{if } e<1 \ \text{(oblate ellipsoids)}, \end{cases} \quad (29)$$

where $p = \sqrt{e^2-1}$ and $q = \sqrt{1-e^2}$.

Eqs. (28) and (29) completely solve the electrical characterisation of a dispersion of randomly oriented ellipsoids of rotation. We may observe that, if $e = 1$ (spherical inclusions) we have $L = 1/3$ and thus Eq. (28) reduce to Eq. (3), which characterises dispersions of dielectric spheres. If $e \to 0$ we deal with a mixture of random oriented strongly oblate (lamellae or penny shaped) inclusions (in this case $L = 0$) and Eq. (28) degenerates to the following one:

$$\varepsilon = \varepsilon_2 \frac{3\varepsilon_1 + 2c(\varepsilon_2-\varepsilon_1)}{3\varepsilon_2 - c(\varepsilon_2-\varepsilon_1)} \quad \text{(Lamellae)}. \quad (30)$$

Finally, if $e \to \infty$ $(L \to 1/2)$ the inclusions become strongly prolate ellipsoids (circular cylinders or rods) randomly distributed in the space and Eq. (28) reduces to Eq. (31) below

$$1-c = \frac{\varepsilon_2-\varepsilon}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_2+5\varepsilon_1}{\varepsilon_2+5\varepsilon}\right)^{2/5} \quad \text{(Rods)}. \quad (31)$$

In Figs. 5 and 6 results obtained by using Eq. (28) are shown. In Fig. 5 a three dimensional plot of $\varepsilon/\varepsilon_1$ is represented (for $\varepsilon_2/\varepsilon_1 = 10$) showing the dependence on c and on $\mathrm{Log}_{10}(e)$. In Fig. 6 the ratio $\varepsilon/\varepsilon_1$ is plotted versus the logarithm of the eccentricity for $\varepsilon_2/\varepsilon_1 = 10$ and for different values of the volume fraction c (ranging from 0 to 1). The result concerning the case with $c = 1/2$ is evidenced and a comparison with corresponding classical Maxwell (Eq. (1)) and Bruggeman

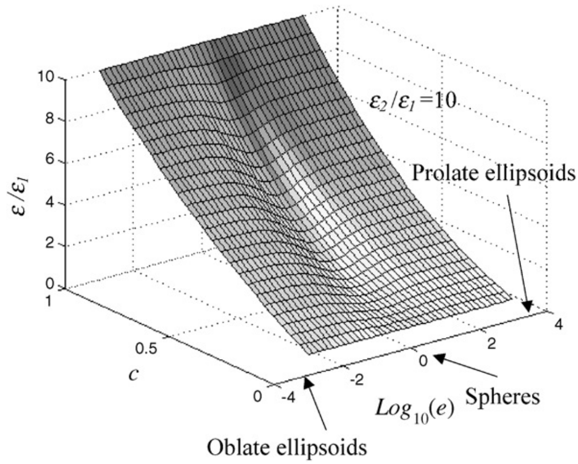

Fig. 5. Results obtained for a mixture of randomly oriented ellipsoids of rotation (Eq. (28)). The surface $\varepsilon/\varepsilon_1$ versus the volume fraction c and $\mathrm{Log}_{10}(e)$ is shown. It corresponds to $\varepsilon_2/\varepsilon_1 = 10$.

(Eq. (31)) formulas is drawn. Furthermore, the three particular limiting cases are evidenced: strongly oblate ellipsoids (lamellae or penny shaped inclusions), spheres and strongly prolate ellipsoids (circular cylinders or rods).

To complete our study we analyse a mixture of ellipsoids with three independent axes. This analysis represents the main result of the work and generalises the previous mixing formulas to the case of a dispersion of randomly oriented general ellipsoids. For such generally shaped ellipsoids the relation $L_x + L_y + L_z = 1$ holds true and therefore we may use two factors (L_x and L_y) as parameters which completely define the shape of the inclusions (they are given by Eq. (16)). To simply integrate Eq. (26) we define the following quantities depending on these depolarisation factors:

$$\begin{aligned}
A &= 3L_x^2L_y + 3L_xL_y^2 - 3L_xL_y,\\
B &= L_x + L_y - 6L_x^2L_y^2 - 8L_xL_y - 4L_x^3L_y - 4L_xL_y^3\\
&\quad + 11L_x^2L_y + 11L_y^2L_x - 3L_x^2 - 3L_y^2 + 4L_x^3 + 4L_y^3 - 2L_x^4 - 2L_y^4,\\
C &= 4L_x^3L_y + 4L_xL_y^3 - 2L_xL_y - 2L_x^2L_y - 2L_y^2L_x + 6L_x^2L_y^2\\
&\quad + 2L_x^2 + 2L_y^2 - 4L_x^3 - 4L_y^3 + 2L_x^4 + 2L_y^4,\\
D &= L_xL_y - L_x - L_y + L_x^2 + L_y^2.
\end{aligned} \tag{32}$$

Therefore, the differential equation (Eq. (26)) may be recast, after some straightforward calculations, in the following simplified form, which represents a partial fractions expansion:

$$\frac{\mathrm{d}c}{1-c} = \left[\frac{1}{\varepsilon_2 - \varepsilon} + \frac{A}{D\varepsilon} + \frac{B\varepsilon + C\varepsilon_2}{D[(D-1)\varepsilon^2 - 2(D+1)\varepsilon\varepsilon_2 + D\varepsilon_2^2]}\right]\mathrm{d}\varepsilon. \tag{33}$$

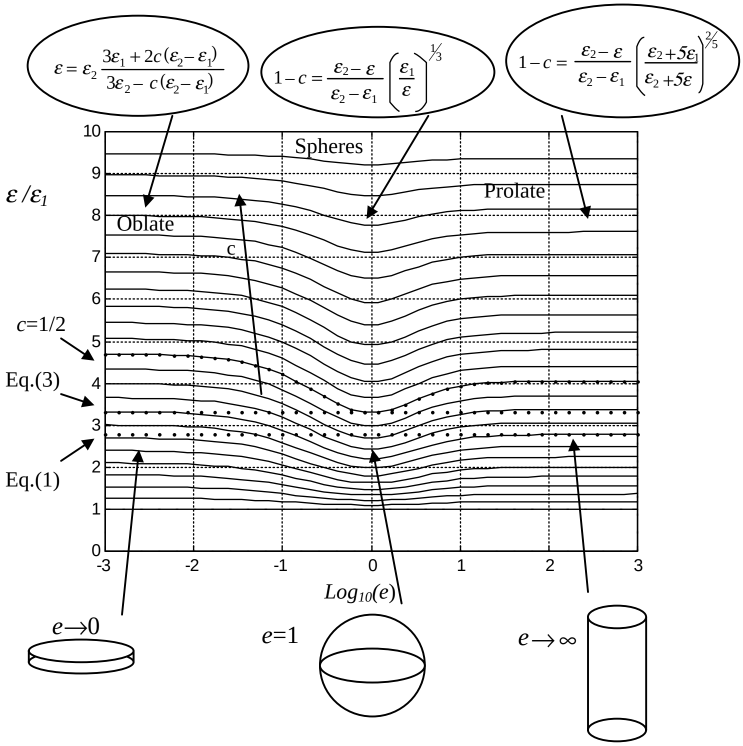

Fig. 6. Same as Fig. 5. The family of curves is parameterised by the volume fraction c. The three limiting cases of strongly oblate ellipsoids (penny shaped), spheres and strongly prolate ellipsoids (rods) are shown. The line concerning the case $c = 1/2$ is evidenced (dotted) to draw a comparison with classical results given by Eq. (1) (Maxwell) and Eq. (3) (Bruggeman).

By means of a lengthy but straightforward integration we have found the solution as follows:

$$1 - c = \frac{\varepsilon_2 - \varepsilon}{\varepsilon_2 - \varepsilon_1}\left(\frac{\varepsilon_1}{\varepsilon}\right)^{A/D}\left[\frac{(D-1)\varepsilon_1^2 - 2(D+1)\varepsilon_1\varepsilon_2 + D\varepsilon_2^2}{(D-1)\varepsilon^2 - 2(D+1)\varepsilon\varepsilon_2 + D\varepsilon_2^2}\right]^{\frac{B}{2D(D-1)}}$$
$$\times\left[\frac{\sqrt{1+3D}\varepsilon_2 + (D-1)\varepsilon - (D+1)\varepsilon_2}{\sqrt{1+3D}\varepsilon_2 + (D-1)\varepsilon_1 - (D+1)\varepsilon_2}\right.$$
$$\left.\times\frac{\sqrt{1+3D}\varepsilon_2 - (D-1)\varepsilon_1 + (D+1)\varepsilon_2}{\sqrt{1+3D}\varepsilon_2 - (D-1)\varepsilon + (D+1)\varepsilon_2}\right]^{\frac{B(D+1)+C(D-1)}{2D(D-1)\sqrt{1+3D}}}. \quad (34)$$

We may note that all the parameters A, B, C, and D depend on the depolarisation factors and thus, recalling Eq. (16) we deduce that they depend only on the two eccentricities e and g. So, the complete model describes the equivalent permittivity as function of the following parameters: the permittivities of the two involved materials ε_2 and ε_1, the volume fraction c of the embedded ellipsoids and the characteristic eccentricities e and g. Finally, $\varepsilon = \varepsilon(\varepsilon_1, \varepsilon_2, c, e, g)$.

Some particular cases may be taken into consideration: if we deal with a mixture of elliptic lamellae the depolarisation factors assume the values $L_x = 1$, $L_y = 0$, $L_z = 0$ and Eq. (34) reduces to Eq. (30) for any value of the eccentricity of the elliptical base of the inclusions (and for any shape of the contour of the lamellae). Another interesting limiting case is that concerning a mixture of elliptic cylinders randomly oriented in the space: for such inclusion the depolarisation factors are given by Eq. (13). If we use these values in Eqs. (32) and (34) we obtain the following mixing formula:

$$\begin{aligned} 1 - c = {}& \frac{\varepsilon_2 - \varepsilon}{\varepsilon_2 - \varepsilon_1}\left[\frac{(e^2+3e+1)\varepsilon_1^2 + 2(e^2+e+1)\varepsilon_1\varepsilon_2 + e\varepsilon_2^2}{(e^2+3e+1)\varepsilon^2 + 2(e^2+e+1)\varepsilon\varepsilon_2 + e\varepsilon_2^2}\right]^{\frac{(1+e^2)}{(2(e^2+3e+1))}} \\ & \times \left[\frac{(1+e)\sqrt{e^2-e+1}\,\varepsilon_2 + (e^2+e+1)\varepsilon_2 + (e^2+3e+1)\varepsilon_1}{(1+e)\sqrt{e^2-e+1}\,\varepsilon_2 + (e^2+e+1)\varepsilon_2 + (e^2+3e+1)\varepsilon}\right. \\ & \left. \times \frac{(1+e)\sqrt{e^2-e+1}\,\varepsilon_2 - (e^2+e+1)\varepsilon_2 - (e^2+3e+1)\varepsilon}{(1+e)\sqrt{e^2-e+1}\,\varepsilon_2 - (e^2+e+1)\varepsilon_2 - (e^2+3e+1)\varepsilon_1}\right]^{\frac{(e^2-3e+1)(1+e)}{2(e^2+3e+1)\sqrt{e^2-e+1}}}, \end{aligned} \tag{35}$$

where e represents the eccentricity of the base of the elliptic cylinders (ε_2) randomly embedded in the homogeneous matrix (ε_1). Two particular sub-cases can be shown for this latter relationship: if $e = 1$ we are describing a mixture of circular cylinders and then Eq. (35) degenerates to Eq. (31); if $e = 0$ the elliptic cylinders degenerate to sheets and thus Eq. (35) reduces to Eq. (30) describing dispersions of lamellae.

The irrational equation (Eq. (34)) has been numerically solved and a typical result is shown in Fig. 7 where one can deduce the effects of the shape of the inclusions on the effective macroscopic dielectric constant of the material. We have considered $\varepsilon_2/\varepsilon_1 = 10$, $c = 1/2$ and we have plotted the values of $\varepsilon/\varepsilon_1$ in terms of the eccentricities e and g of the embedded ellipsoids. The limiting cases of interest have been clearly indicated.

In our models the fraction volume ranges from 0 to 1 and this fact needs some explications: obviously, if one requires that the inclusions (actually ellipsoids) are not overlapping, the volume fraction can not reaches the unit value. By means of simple computer simulations we may verify that, placing inclusions at random in a given volume, they become overlapping when the volume fraction is greater than a given threshold. Nevertheless, we may think that Bruggeman's procedure continues to yield good results even for higher values of the volume fraction, when the inclusions become strongly overlapped. This fact derives from several comparisons between theoretical results for mixtures of spheres and experimental measurements, which can be found in literature [1]. The typical application of the Bruggeman procedure has given results in agreement with experimental measurements in various fields

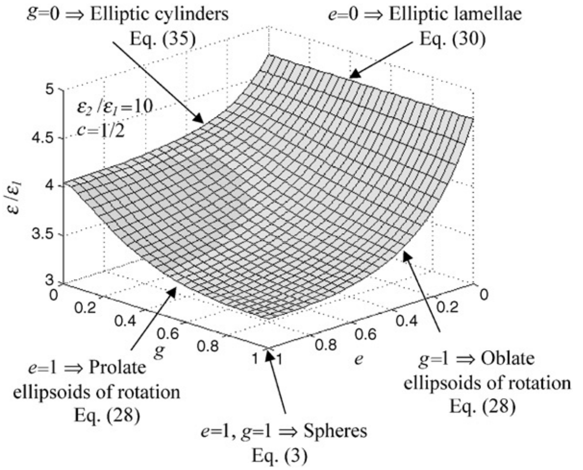

Fig. 7. Results for a mixture of randomly oriented and generally shaped ellipsoids (Eq. (32)). The surface $\varepsilon/\varepsilon_1$ versus the eccentricities e and g is shown with the assumptions $\varepsilon_2/\varepsilon_1 = 10$ and $c = 1/2$.

ranging from characterisation of dielectric mixtures of spheres [1,3,11] to characterisation of micro-mechanical dispersions from the elastic point of view (in this case the results are very satisfactory over the complete range $0<c<1$ [20,21]).

4. Conclusions

We have applied Bruggeman's procedure to a dispersion of ellipsoids and we have studied the resulting relationships for the equivalent permittivity. When the above procedure is implemented for evaluating the aforesaid permittivity, a sensible dependence of the results on the shape of the inclusions (actually on the eccentricities) is shown. The closed form analysis of a dispersion of ellipsoids offers a simple but clear example of the dependence of the macroscopic behaviour of composite materials on the microstructure or microscopic morphology. The results may suggest a hint for explaining why sometimes there are some inconsistencies between the standard mixture formulae and corresponding experiments: typically, standard formulae are based on the Maxwell relation for a mixture of spheres and do not take into account any different shapes of the inclusions, which may be present in actual heterogeneous media. Once more, our results show a dependence of the effective property on the eccentricities of the inclusions that is absolutely not negligible and thus they should be always taken into account when not spherical inclusions are embedded in a matrix. Finally, an explicit new relationship has been derived for the case of a dispersion of randomly oriented generally shaped ellipsoids.

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