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Order and disorder in heterogeneous material microstructure: Electric and elastic characterisation of dispersions of pseudo-oriented spheroids

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Abstract

The paper deals with the electrical and elastic characterisation of dispersions of pseudo-oriented ellipsoids of rotation: it means that we are dealing with mixtures of inclusions of different eccentricities and arbitrary non-random orientational distributions. The analysis ranges from parallel spheroidal inclusions to completely random oriented inclusions. A unified theory covers all the orientational distributions between the random and the parallel ones. The electrical and micro-mechanical averaging inside the composite material is carried out by means of explicit results which allows us to obtain closed-form expressions for the macroscopic or equivalent dielectric constants or elastic moduli of the overall composite materials. In particular, this study allows us to affirm that the electrical behaviour of such a dispersion of pseudo-oriented particles is completely defined by one order parameter which depends on the given angular distribution. Moreover, the elastic characterisation of this heterogeneous material depends on two order parameters, which derive from the orientational distribution. The theory may be applied to characterise media with different shapes of the inclusions (i.e. spheres, cylinders or planar inhomogeneities) yielding a set of procedures describing several composite materials of great technological interest.

Keywords: Composite materials; Electric and elastic homogenisation; Mixture theory and order parameters

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1. Introduction

In recent years the characterisation of heterogeneous materials has attracted an ever increasing interest. A central problem, of considerable technological importance, is to evaluate the effective electric and elastic properties governing the behaviour of a composite material on the macroscopic scale. At present, it is well known that it does not exist a universally applicable mixing formula giving the effective properties of the heterogeneous materials as some sort of average of the properties of the constituent materials. In fact, the details of the micro-geometry can play a crucial role in determining the overall properties, particularly when the crystalline grains have highly anisotropic behaviour or when there is a large difference in the properties of the constituent materials. Therefore, the elastic and electrical properties of composite materials are strongly microstructure dependent. The main goal in the study of materials is to understand and classify the relationships between the internal structure of materials and their properties. The relationship between microstructure and properties may be used for designing and improving materials, or conversely, for interpreting experimental data in terms of micro-structural features. Ideally, the aim is to construct a theory that employs general micro-structural information to make some accurate property predictions. A simpler goal is the provision of property for different class of microstructures. A great number of theoretical formulas have been proposed to describe the behaviour of composite materials. A disadvantage of some approximated results is that they do not correspond to a priori known microstructure; this kind of results may be interpreted and classified only by means of comparison with numerical or experimental data. A different class of theories is rigorously based on realistic microstructures. These are the classical Hashin–Shtrikman variational bounds [1,2], which provide an upper and lower bound for composite materials, and the expansions of Brown [3] and Torquato [4,5] which take into account the spatial correlation function of the phases.

Dispersions or suspensions of inclusions in a homogeneous matrix give a particular example of heterogeneous materials: these media have been extensively studied both from the electrical and the elastic point of view. One of the first attempts to characterise electrical dispersions of spheres is that of Maxwell [6], which found out a famous formula for a strongly diluted suspension of spheres. A better model has been provided by the differential scheme, which derives from the mixture characterisation approach used by Bruggeman [7] and extensively described by Van Beek [8]. In this case the relations should maintain the validity also for less diluted suspensions of spheres. To understand the effect of different shape of the inclusions, ellipsoidal shaped particles have been considered: the first attempt was made by Fricke [9,10] dealing with the electrical characterisation of inhomogeneous biological tissues containing spheroidal particles. Many related results are summarised in [11,12]. In recent literature some applications of the Bruggeman differential procedure to mixtures of ellipsoids have been performed [13–15] in connection with the problem of characterising the dielectric response of water-saturated rocks. A complete review of the Bruggeman theory for ellipsoidal inclusions has been developed in Ref. [16].

Dealing with elastic characterisation of dispersions (see [17] for a theoretical introduction) some similar works have been developed: an exact result exists for such a material composed by a very dilute concentration of spherical inclusions dispersed in a solid matrix. This result is attributed to numerous authors [18,19]. To adapt the dilute formulas to the case of any finite volume fraction the differential method is applied both for spherical or cylindrical inclusions [20] and for ellipsoidal particles [21].

In this work we are dealing with a dispersion of spheroids (ellipsoids of rotations) embedded in a homogeneous matrix. A particular attention is devoted to the analysis of the effects of the orientational distribution of the particles inside the composite material. The limiting cases of the present theory are represented by all the particles aligned with a given direction (order) and all the particles randomly oriented (disorder). We take into account all the intermediate configurations between order and disorder with the aim to characterise a material with particles partially aligned. In Fig. 1 one can find some orientational distributions between the upon described limiting cases. We consider a given orthonormal reference frame and we take as preferential direction of alignment the z-axis. Each particle embedded in the matrix is not completely random oriented. The orientation is described by the following statistical rule: the principal axis of each particle forms with the z-axis an angle θ which follows a given probability density $f(\theta)$ defined in $[0\ \pi]$. The orientation of each particle is statistically independent from the orientation of the other particles. If $f(\theta) = \delta(\theta)$ (where δ is the Dirac delta function) we have all the particles with $\theta = 0$ and therefore they are all oriented with the z-axis. If $f(\theta) = (1/2)\sin\theta$ all the particles are uniformly random oriented in the space over all the possible orientations. Any other statistical distributions $f(\theta)$ define a transversely isotropic (uniaxial) material. In the following sections we develop a complete analysis of the combined effects of the shape (eccentricity) of the particles and of the state of order/disorder. This analysis allows us to evaluate the overall electric and elastic properties of the heterogeneous material. In particular, from the point of view of the shape of the particles, the so-called depolarisation factor L is the parameter that intervenes to characterise the medium. From the point of view of the state of order we verified the following property: for the electrical characterisation of the composite medium the state of order acts on the overall dielectric constant only by means of a parameter S, which takes into account the average value of the second Legendre polynomial, $S = \langle P_2(\cos\theta)\rangle_\theta$. On the other hand, the elastic moduli of the material depend on the state of order through two parameters that are defined as follows: $S = \langle P_2(\cos\theta)\rangle_\theta$ and $T = \langle P_4(\cos\theta)\rangle_\theta$. They correspond to the average values of the Legendre polynomial of order two and four, computed by means of the density probability $f(\theta)$. The results may be applied to describe the physical behaviour of heterogeneous materials starting from the knowledge of the physical properties (permittivity, elastic moduli) of each medium composing the mixture as

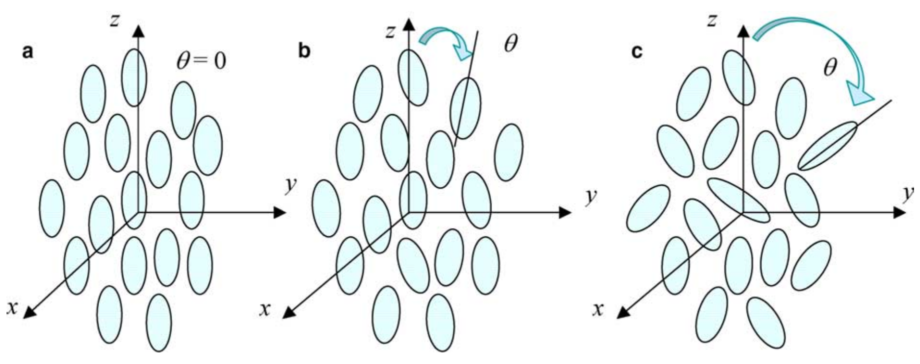


Fig. 1. Structure of a dispersion of pseudo-oriented ellipsoids. One can find some orientational distributions ranging from order to disorder. The two-phase material is described by the electric and/or elastic response of each phase, by the state of order and by the volume fraction of the inclusions.

well as of the structural properties of the mixture itself i.e. shape of the inclusions and state of order of the orientations (L and S for the electrical case, L, S and T for the elastic case).

2. Dielectric theory for pseudo-oriented inclusions

The theory is based on the following preliminary 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 $\overline{E}_0 = (E_{0x}, E_{0y}, E_{0z})$ applied to the structure. Then, according to Stratton [22] or Landau and Lifshitz [23] a uniform electrical field appears inside the ellipsoid and it can be computed as follows. We define the depolarisation factors along each axes:

$$L_k = \frac{a_x a_y a_z}{2} \int_0^{+\infty} \frac{\mathrm{d}s}{(s + a_k^2)\sqrt{(s + a_x^2)(s + a_y^2)(s + a_z^2)}} \tag{1}$$

Here and in the rest of the paper the index k ranges over the three symbols x, y and z. Explicit expressions of the depolarisation factors are given in literature [16] in terms of elliptic integrals. However, in the present work we are dealing with ellipsoids of rotation and the formulas are simplified and reported in the sequel. We may simply observe that $L_x + L_y + L_z = 1$. On the basis of these considerations, the electrical field inside the ellipsoidal inclusion is given, in components, by the following relations described, for example, by Stratton [22]:

$$E_{i,k} = \frac{\varepsilon_1 E_{0k}}{\varepsilon_1 + L_k(\varepsilon_2 - \varepsilon_1)} \tag{2}$$

Actually, 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 $\bar{n}_z$ (defined in a given reference frame) and they are aligned with the axes a_x, a_y and a_z of the ellipsoid, respectively. In the following the three unit vectors will be described by a given statistical distribution in order to model the microstructure of the heterogeneous material. By using Eq. (2), we may compute the electrical field inside the inclusion, induced by a given external uniform electric field:

$$\overline{E}_i = \frac{\varepsilon_1(\overline{E}_0 \cdot \bar{n}_x)\bar{n}_x}{\varepsilon_1 + L_x(\varepsilon_2 - \varepsilon_1)} + \frac{\varepsilon_1(\overline{E}_0 \cdot \bar{n}_y)\bar{n}_y}{\varepsilon_1 + L_y(\varepsilon_2 - \varepsilon_1)} + \frac{\varepsilon_1(\overline{E}_0 \cdot \bar{n}_z)\bar{n}_z}{\varepsilon_1 + L_z(\varepsilon_2 - \varepsilon_1)} \tag{3}$$

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 (component by component), as follows:

$$E_{i,q} = \varepsilon_1 \sum_k^{x,y,z} E_{0,k} \sum_j^{x,y,z} \frac{n_{j,k} n_{j,q}}{\varepsilon_1 + L_j(\varepsilon_2 - \varepsilon_1)} \tag{4}$$

where $n_{j,k}$ is the kth component of the unit vector $\bar{n}_j$ ($j = x, y, z$). From now on, we are interested in the behaviour of an ellipsoid of rotation and therefore we use the simplified notation $L_x = L_y = L$ and $L_z = 1 - 2L$. It means that L is the depolarisation factor along the unit vectors

$\bar{n}_x$ and $\bar{n}_y$ and $1-2L$ is the depolarising vector along the axis $\bar{n}_z$. We may use spherical coordinates ψ, φ and ϑ to write down explicit expressions for the unit vectors:

$$\begin{cases} \bar{n}_x = (\cos\psi\cos\varphi - \sin\psi\sin\varphi\cos\theta, -\cos\psi\sin\varphi - \sin\psi\cos\varphi\cos\theta, \sin\psi\sin\theta) \\ \bar{n}_y = (\sin\psi\cos\varphi + \cos\psi\sin\varphi\cos\theta, -\sin\psi\sin\varphi + \cos\psi\cos\varphi\cos\theta, -\cos\psi\sin\theta) \\ \bar{n}_x = (\sin\varphi\sin\theta, \sin\theta\cos\varphi, \cos\theta) \end{cases} \tag{5}$$

For the following derivations, we are interested in the average value of the electrical field inside the ellipsoid over the possible orientations of the ellipsoid itself and then we have to compute the average value of the quantity $n_{j,k}n_{j,q}$. The two angles ψ and φ are statistical independent from each other and distributed following a uniform probability density in the range $[0\ 2\pi]$. Performing the integration over the unit sphere, by means of spherical coordinates, we obtain, after some straightforward computations, the first step of the averaging procedure:

$$\begin{cases} \langle n_{x,x}n_{x,x}\rangle_{\psi,\varphi} = \langle n_{x,y}n_{x,y}\rangle_{\psi,\varphi} = \langle n_{y,x}n_{y,x}\rangle_{\psi,\varphi} = \langle n_{y,y}n_{y,y}\rangle_{\psi,\varphi} = \frac{1}{4}(1+\cos^2\theta) \\ \langle n_{x,z}n_{x,z}\rangle_{\psi,\varphi} = \langle n_{y,z}n_{y,z}\rangle_{\psi,\varphi} = \langle n_{z,x}n_{z,x}\rangle_{\psi,\varphi} = \langle n_{z,y}n_{z,y}\rangle_{\psi,\varphi} = \frac{1}{4}(1-\cos^2\theta) \\ \langle n_{z,z}n_{z,z}\rangle_{\psi,\varphi} = \cos^2\theta \end{cases} \tag{6}$$

Here, the symbol $\langle\ \rangle_{\psi,\varphi}$ represents the average value over the angles ψ and φ. The terms that not appear in the previous Eq. (6) are all zero.

The angle θ is statistical independent from the others and distributed following an arbitrary probability density $f(\theta)$, which defines the degree of ordering of the medium, ranging from perfect order ($f(\theta)=\delta(\theta)$), to complete disorder ($f(\theta)=(1/2)\sin\theta$). The statistical distribution of the angle θ is well described by the following order parameter S, which takes into account the average value of the second Legendre polynomial:

$$S = \langle P_2(\cos\theta)\rangle_\theta = \left\langle \frac{3}{2}\cos^2(\theta) - \frac{1}{2}\right\rangle_\theta = \int_0^\pi \left(\frac{3}{2}\cos^2(\theta) - \frac{1}{2}\right) f(\theta)\,\mathrm{d}\theta \tag{7}$$

where θ is the angle that the particle (its versor $\bar{n}_z$) makes with the preferential direction given by the axis z of the main reference frame (the symbol $\langle\ \rangle_\vartheta$ represents the average value over the angle ϑ).

By means of the definition of such order parameter we may perform the final averaging over the tilting angle θ:

$$\begin{cases} \langle n_{x,x}n_{x,x}\rangle_{\psi,\varphi,\theta} = \langle n_{x,y}n_{x,y}\rangle_{\psi,\varphi,\theta} = \langle n_{y,x}n_{y,x}\rangle_{\psi,\varphi,\theta} = \langle n_{y,y}n_{y,y}\rangle_{\psi,\varphi,\theta} = \frac{2}{3}(S+2) \\ \langle n_{x,z}n_{x,z}\rangle_{\psi,\varphi,\theta} = \langle n_{y,z}n_{y,z}\rangle_{\psi,\varphi,\theta} = \langle n_{z,x}n_{z,x}\rangle_{\psi,\varphi,\theta} = \langle n_{z,y}n_{z,y}\rangle_{\psi,\varphi,\theta} = \frac{2}{3}(1-S) \\ \langle n_{z,z}n_{z,z}\rangle_{\psi,\varphi,\theta} = \frac{2S+1}{3} \end{cases} \tag{8}$$

Here, the symbol $\langle\ \rangle_{\psi,\varphi,\vartheta}$ represents the average value over the angles ψ, φ and ϑ; for sake of simplicity, from now on the indication of the angles on which the averaging is performed will be omitted. Therefore, the average value of the electrical field (inside the randomly oriented inclusion), given by Eq. (4), may be written as:

$$\begin{cases} \langle E_{i,x}\rangle = \frac{E_{0,x}}{3}\left[\dfrac{\varepsilon_1(S+2)}{\varepsilon_1+L(\varepsilon_2-\varepsilon_1)}+\dfrac{\varepsilon_1(1-S)}{\varepsilon_1+(1-2L)(\varepsilon_2-\varepsilon_1)}\right] \\ \langle E_{i,y}\rangle = \frac{E_{0,y}}{3}\left[\dfrac{\varepsilon_1(S+2)}{\varepsilon_1+L(\varepsilon_2-\varepsilon_1)}+\dfrac{\varepsilon_1(1-S)}{\varepsilon_1+(1-2L)(\varepsilon_2-\varepsilon_1)}\right] \\ \langle E_{i,z}\rangle = \frac{E_{0,z}}{3}\left[\dfrac{2\varepsilon_1(1-S)}{\varepsilon_1+L(\varepsilon_2-\varepsilon_1)}+\dfrac{\varepsilon_1(2S+1)}{\varepsilon_1+(1-2L)(\varepsilon_2-\varepsilon_1)}\right] \end{cases} \tag{9}$$

Now, we are ready to consider a mixture of pseudo-oriented ellipsoids. In Fig. 1 one can find the structure of the composite material with various degrees of order: we consider a given number of randomly oriented ellipsoids (ε_2) embedded in a homogeneous matrix (ε_1). 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 given by:

$$\langle\overline{E}\rangle = (1-c)\overline{E}_0 + c\langle\overline{E}_i\rangle \tag{10}$$

Then, we define $[\varepsilon]$ as the equivalent permittivity tensor of the whole mixture by means of the relation $\langle\overline{D}\rangle = [\varepsilon]\langle\overline{E}\rangle$ [23]; to evaluate $[\varepsilon]$ 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_0 as the volume of the remaining space among the inclusions (so that $V = V_e \cup V_0$). The average value of $\overline{D}(\overline{r}) = \varepsilon(\overline{r})\overline{E}(\overline{r})$ is evaluated as follows:

$$\begin{aligned} \langle\overline{D}\rangle &= \frac{1}{V}\int_V \varepsilon(\overline{r})\overline{E}(\overline{r})\,\mathrm{d}\overline{r} = \frac{1}{V}\varepsilon_1\int_{V_0}\overline{E}(\overline{r})\,\mathrm{d}\overline{r} + \frac{1}{V}\varepsilon_2\int_{V_e}\overline{E}(\overline{r})\,\mathrm{d}\overline{r} \\ &= \frac{1}{V}\varepsilon_1\int_{V_0}\overline{E}(\overline{r})\,\mathrm{d}\overline{r} + \frac{1}{V}\varepsilon_1\int_{V_e}\overline{E}(\overline{r})\,\mathrm{d}\overline{r} + \frac{1}{V}\varepsilon_2\int_{V_e}\overline{E}(\overline{r})\,\mathrm{d}\overline{r} - \frac{1}{V}\varepsilon_1\int_{V_e}\overline{E}(\overline{r})\,\mathrm{d}\overline{r} \\ &= \varepsilon_1\langle\overline{E}\rangle + c(\varepsilon_2-\varepsilon_1)\langle\overline{E}_i\rangle \end{aligned} \tag{11}$$

Note that $\langle\overline{D}\rangle$ and $\langle\overline{E}\rangle$ are not parallel vectors because of the presence of the average value of the internal electric field given by Eq. (9). Drawing a comparison between Eqs. (9)–(11) we may find complete expressions, which allows us to estimate the equivalent permittivity tensor $[\varepsilon]$:

$$[\varepsilon] = \begin{bmatrix} \varepsilon_\perp & 0 & 0 \\ 0 & \varepsilon_\perp & 0 \\ 0 & 0 & \varepsilon_\parallel \end{bmatrix} \tag{12}$$

where the longitudinal and transversal permittivities are given by:

$$\begin{cases} \varepsilon_{\perp} = \varepsilon_1 + c(\varepsilon_2 - \varepsilon_1) \dfrac{\dfrac{1}{3}\left[\dfrac{\varepsilon_1(S+2)}{\varepsilon_1 + L(\varepsilon_2 - \varepsilon_1)} + \dfrac{\varepsilon_1(1-S)}{\varepsilon_1 + (1-2L)(\varepsilon_2 - \varepsilon_1)}\right]}{1 - c + \dfrac{c}{3}\left[\dfrac{\varepsilon_1(S+2)}{\varepsilon_1 + L(\varepsilon_2 - \varepsilon_1)} + \dfrac{\varepsilon_1(1-S)}{\varepsilon_1 + (1-2L)(\varepsilon_2 - \varepsilon_1)}\right]} \\ \varepsilon_{\parallel} = \varepsilon_1 + c(\varepsilon_2 - \varepsilon_1) \dfrac{\dfrac{1}{3}\left[\dfrac{2\varepsilon_1(1-S)}{\varepsilon_1 + L(\varepsilon_2 - \varepsilon_1)} + \dfrac{\varepsilon_1(2S+1)}{\varepsilon_1 + (1-2L)(\varepsilon_2 - \varepsilon_1)}\right]}{1 - c + \dfrac{c}{3}\left[\dfrac{2\varepsilon_1(1-S)}{\varepsilon_1 + L(\varepsilon_2 - \varepsilon_1)} + \dfrac{\varepsilon_1(2S+1)}{\varepsilon_1 + (1-2L)(\varepsilon_2 - \varepsilon_1)}\right]} \end{cases} \tag{13}$$

For some following applications it is interesting to take into consideration the first order expansion of the previous results:

$$\begin{cases} \varepsilon_{\perp} = \varepsilon_1 + c(\varepsilon_2 - \varepsilon_1)\dfrac{1}{3}\left[\dfrac{\varepsilon_1(S+2)}{\varepsilon_1 + L(\varepsilon_2 - \varepsilon_1)} + \dfrac{\varepsilon_1(1-S)}{\varepsilon_1 + (1-2L)(\varepsilon_2 - \varepsilon_1)}\right] + \mathrm{O}(c^2) \\ \varepsilon_{\parallel} = \varepsilon_1 + c(\varepsilon_2 - \varepsilon_1)\dfrac{1}{3}\left[\dfrac{2\varepsilon_1(1-S)}{\varepsilon_1 + L(\varepsilon_2 - \varepsilon_1)} + \dfrac{\varepsilon_1(2S+1)}{\varepsilon_1 + (1-2L)(\varepsilon_2 - \varepsilon_1)}\right] + \mathrm{O}(c^2) \end{cases} \tag{14}$$

This result concerns the characterisation of a very diluted dispersion of randomly oriented ellipsoids with given shape (i.e. with fixed depolarisation factor L) and given state of order (i.e. fixed S). To adapt this relationship to arbitrarily diluted composite materials we use the Bruggeman's procedure: actually, the application of this method as follows is only an approximation that in literature is often considered and it is known as asymmetrical or differential effective medium approximation [7,8,16]. Anyway, it leads to the following differential equations:

$$\begin{cases} \dfrac{\mathrm{d}\varepsilon_{\perp}}{\mathrm{d}c} = \dfrac{1}{1-c}\varepsilon_{\perp}(\varepsilon_2 - \varepsilon_{\perp})\dfrac{1}{3}\left[\dfrac{(S+2)}{\varepsilon_{\perp} + L(\varepsilon_2 - \varepsilon_{\perp})} + \dfrac{(1-S)}{\varepsilon_{\perp} + (1-2L)(\varepsilon_2 - \varepsilon_{\perp})}\right] \\ \dfrac{\mathrm{d}\varepsilon_{\parallel}}{\mathrm{d}c} = \dfrac{1}{1-c}\varepsilon_{\parallel}(\varepsilon_2 - \varepsilon_{\parallel})\dfrac{1}{3}\left[\dfrac{2(1-S)}{\varepsilon_{\parallel} + L(\varepsilon_2 - \varepsilon_{\parallel})} + \dfrac{(2S+1)}{\varepsilon_{\parallel} + (1-2L)(\varepsilon_2 - \varepsilon_{\parallel})}\right] \end{cases} \tag{15}$$

The solution of these equations depends on the values of the depolarisation factor and of the order parameter showing the relationship between the overall permittivities and the microstructure. Expressions in Eq. (15) reduce, after some straightforward computations, to the following ones:

$$\begin{cases} \dfrac{\mathrm{d}c}{1-c} = \left[\dfrac{1}{\varepsilon_2 - \varepsilon_{\perp}} + \dfrac{3L(1-2L)}{P\varepsilon_{\perp}} + \dfrac{(1-3L)^2(2+S)(1-S)}{P[(3-P)\varepsilon_{\perp} + P\varepsilon_2]}\right]\mathrm{d}\varepsilon_{\perp} \quad \text{where } P = 2 - 3L + S - 3SL \\ \dfrac{\mathrm{d}c}{1-c} = \left[\dfrac{1}{\varepsilon_2 - \varepsilon_{\parallel}} + \dfrac{3L(1-2L)}{Q\varepsilon_{\parallel}} + \dfrac{2(1-3L)^2(2S+1)(1-S)}{Q[(3-Q)\varepsilon_{\parallel} + Q\varepsilon_2]}\right]\mathrm{d}\varepsilon_{\parallel} \quad \text{where } Q = 2 - 3L - 2S + 6SL \end{cases} \tag{16}$$

The integration of the above partial fraction expansions, with the conditions $\varepsilon_{\perp}(c=0) = \varepsilon_1$, and $\varepsilon_{\parallel}(c=0) = \varepsilon_1$, yields the final result:

$$\begin{cases} 1-c=\dfrac{\varepsilon_2-\varepsilon_\perp}{\varepsilon_2-\varepsilon_1}\left(\dfrac{\varepsilon_1}{\varepsilon_\perp}\right)^{\frac{3L(1-2L)}{2-3L+S-3SL}} \\ \quad\times\left[\dfrac{(1+3L-S+3SL)\varepsilon_1+(2-3L+S-3SL)\varepsilon_2}{(1+3L-S+3SL)\varepsilon_\perp+(2-3L+S-3SL)\varepsilon_2}\right]^{\frac{(1-3L)^2(2+S)(1-S)}{(2-3L+S-3SL)(1+3L-S+3SL)}} \\ 1-c=\dfrac{\varepsilon_2-\varepsilon_\parallel}{\varepsilon_2-\varepsilon_1}\left(\dfrac{\varepsilon_1}{\varepsilon_\parallel}\right)^{\frac{3L(1-2L)}{2-3L-2S+6SL}} \\ \quad\times\left[\dfrac{(1+3L+2S-6SL)\varepsilon_1+(2-3L-2S+6SL)\varepsilon_2}{(1+3L+2S-6SL)\varepsilon_\parallel+(2-3L-2S+6SL)\varepsilon_2}\right]^{\frac{2(1-3L)^2(2S+1)(1-S)}{(2-3L-2S+6SL)(1+3L+2S-6SL)}} \end{cases} \tag{17}$$

This is the main result for the electrical characterisation presented in this work. We define the eccentricity e as the ratio $e=a_z/a_x=a_z/a_y$. The depolarisation factor L may be computed in closed form as follows and the result 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 $e<1$ [16,23]:

$$L=\frac{e}{2}\int_0^{+\infty}\frac{\mathrm{d}\xi}{(\xi+1)^2(\xi+e^2)^{1/2}}=\begin{cases}\dfrac{e}{4\left(\sqrt{e^2-1}\right)^3}\left[2e\sqrt{e^2-1}+\ln\dfrac{e-\sqrt{e^2-1}}{e+\sqrt{e^2-1}}\right] & \text{if } e>1\\ \dfrac{e}{4\left(\sqrt{1-e^2}\right)^3}\left[\pi-2e\sqrt{1-e^2}-2\,\mathrm{arctg}\dfrac{e}{\sqrt{1-e^2}}\right] & \text{if } e<1\end{cases} \tag{18}$$

The factor L assumes some characteristic values in correspondence to special shapes of the particles: for spheres $L=1/3$, for cylinders $L=1/2$ and for lamellae or penny shaped inclusions $L=0$. Moreover, the parameter S assume special values in some particular conditions of degree of order: $S=1$ with perfect order, $S=0$ with complete disorder and $S=-1/2$ when all the particles have the axes of rotation orthogonal to the z-axis of the main reference frame (all particles are lying randomly in planes perpendicular to the z-axis). Such particular values of the parameters S and L in Eq. (17) generate a series of analytical results, which are summarised below.

If $S=0$ we are in a state of complete disorder and we obtain a simplified result where $\varepsilon_\perp=\varepsilon_\parallel=\varepsilon$:

$$1-c=\frac{\varepsilon_2-\varepsilon}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\varepsilon}\right)^{\frac{3L(1-2L)}{2-3L}}\left[\frac{(1+3L)\varepsilon_1+(2-3L)\varepsilon_2}{(1+3L)\varepsilon+(2-3L)\varepsilon_2}\right]^{\frac{2(3L-1)^2}{(2-3L)(1+3L)}} \tag{19}$$

Eq. (19) solves the electrical characterisation of a dispersion of randomly oriented ellipsoids of rotation [16]. We may observe that, if $e=1$ (spherical inclusions) we have $L=1/3$ and thus Eq. (19) reduce to the following famous one, which characterises dispersions of dielectric spheres:

$$1-c=\frac{\varepsilon_2-\varepsilon}{\varepsilon_2-\varepsilon_1}\left(\frac{\varepsilon_1}{\varepsilon}\right)^{\frac{1}{3}} \tag{20}$$

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. (19) reduces to Eq. (21) below:

$$1 - c = \frac{\varepsilon_2 - \varepsilon}{\varepsilon_2 - \varepsilon_1}\left(\frac{\varepsilon_2 + 5\varepsilon_1}{\varepsilon_2 + 5\varepsilon}\right)^{\frac{2}{5}} \tag{21}$$

Finally, 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. (19) degenerates to the following one:

$$\varepsilon = \varepsilon_2 \frac{3\varepsilon_1 + 2c(\varepsilon_2 - \varepsilon_1)}{3\varepsilon_2 - c(\varepsilon_2 - \varepsilon_1)} \tag{22}$$

If $S = 1$ we are in a state of complete order and all the ellipsoids are perfectly aligned. The system in Eq. (17) reduces to the simpler one, where each permittivity directly depends on the correspondent depolarisation factor:

$$\begin{cases} 1 - c = \dfrac{\varepsilon_2 - \varepsilon_\perp}{\varepsilon_2 - \varepsilon_1}\left(\dfrac{\varepsilon_1}{\varepsilon_\perp}\right)^{L} \\ 1 - c = \dfrac{\varepsilon_2 - \varepsilon_\parallel}{\varepsilon_2 - \varepsilon_1}\left(\dfrac{\varepsilon_1}{\varepsilon_\parallel}\right)^{1-2L} \end{cases} \tag{23}$$

If $S = -1/2$ all the particles have the axes of rotation orthogonal to the z-axis of the main reference frame; it means that all the particles are lying randomly in planes perpendicular to the z-axis. Eq. (17) with the assumption $S = -1/2$ leads to the result:

$$\begin{cases} 1 - c = \dfrac{\varepsilon_2 - \varepsilon_\perp}{\varepsilon_2 - \varepsilon_1}\left(\dfrac{\varepsilon_1}{\varepsilon_\perp}\right)^{\frac{2L(1-2L)}{1-L}}\left[\dfrac{(1+L)\varepsilon_1 + (1-L)\varepsilon_2}{(1+L)\varepsilon_\perp + (1-L)\varepsilon_2}\right]^{\frac{(1-3L)^2}{(1-L)(1+L)}} \\ 1 - c = \dfrac{\varepsilon_2 - \varepsilon_\parallel}{\varepsilon_2 - \varepsilon_1}\left(\dfrac{\varepsilon_1}{\varepsilon_\parallel}\right)^{L} \end{cases} \tag{24}$$

We may observe that, if $e = 1$ (spherical inclusions) we have $L = 1/3$ and thus Eq. (24) reduce to Eq. (20) as expected. If $e \to \infty$ ($L \to 1/2$) the inclusions become circular cylinders lying randomly in planes perpendicular to the z-axis and Eq. (24) reduces to Eq. (25) below:

$$\begin{cases} 1 - c = \dfrac{\varepsilon_2 - \varepsilon_\perp}{\varepsilon_2 - \varepsilon_1}\left(\dfrac{3\varepsilon_1 + \varepsilon_2}{3\varepsilon_\perp + \varepsilon_2}\right)^{\frac{1}{3}} \\ 1 - c = \dfrac{\varepsilon_2 - \varepsilon_\parallel}{\varepsilon_2 - \varepsilon_1}\left(\dfrac{\varepsilon_1}{\varepsilon_\parallel}\right)^{\frac{1}{2}} \end{cases} \tag{25}$$

Finally, if $e \to 0$ we deal with a mixture of random oriented lamellae with the plane parallel to the z-axis (in this case $L = 0$) and Eq. (24) degenerates to the following one:

$$\begin{cases} \varepsilon_\perp = \varepsilon_2 \dfrac{2\varepsilon_1 + c(\varepsilon_2 - \varepsilon_1)}{2\varepsilon_2 - c(\varepsilon_2 - \varepsilon_1)} \\ \varepsilon_\parallel = c\varepsilon_2 + (1-c)\varepsilon_1 \end{cases} \tag{26}$$

We are conscious that Eq. (17), the main result, has been derived with a not rigorous mathematical procedure because of the application of the differential method of Bruggeman to this type of not isotropic material. Nevertheless, the expressions are interesting because, although approximately, describe in explicit form the interaction between the degree of order and the shape of particles embedded in the microstructure. However, we have solved Eq. (17) numerically and the results are shown in Fig. 2. More precisely, in Fig. 2a one can find the results when $\varepsilon_2/\varepsilon_1 = 10$, in Fig. 2b the results when $\varepsilon_2/\varepsilon_1 = 1/10$; in both cases we have considered a volume fraction $c = 1/3$ and we have plotted the longitudinal and transversal permittivities versus the order parameters S and decimal logarithm of the eccentricity $\log_{10}e$. We may observe that the effect of the order/disorder has opposite behaviour for prolate and oblate particles. This analysis has immediate applications to the field of the liquid crystals. The use of the order parameter in such

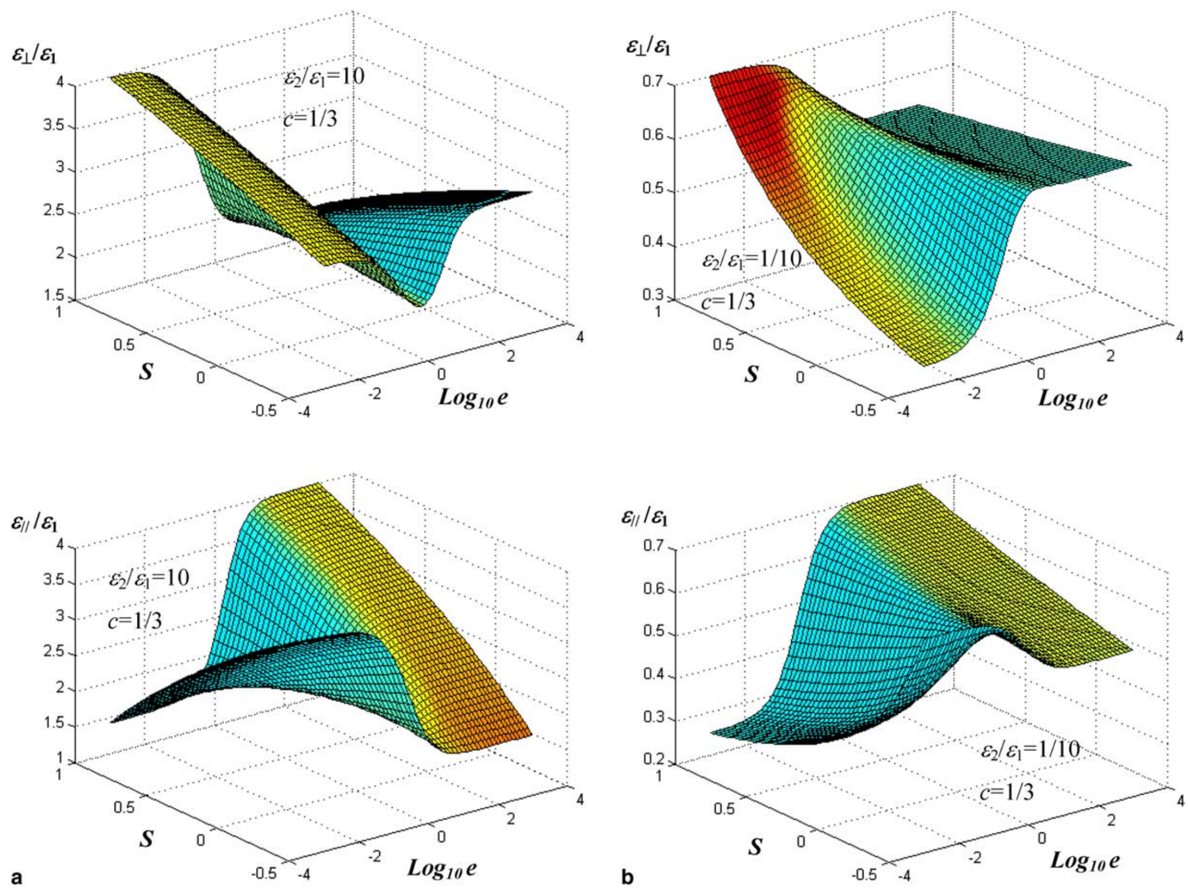


Fig. 2. Transversal and longitudinal dielectric permittivities versus the order parameters S and decimal logarithm of the eccentricity $\log_{10}e$. In (a) one can find the results when $\varepsilon_2/\varepsilon_1 = 10$, in (b) the results when $\varepsilon_2/\varepsilon_1 = 1/10$; in both cases we have considered a volume fraction $c = 1/3$.

field can be found in [24]. Our microstructure describes a material positionally disordered, but orientationally ordered, which corresponds to a nematic phase in liquid crystals [25,26]. So, the theory may be applied to a better understanding of the anisotropic optical behaviour of such materials.

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), electrical conductivity (in the stationary case), thermal conductivity (in a steady-state thermal regime) and so on. In other words, all the transport properties in heterogeneous materials follow the same mixing law when we are referring to the same microstructure, except for the elastic moduli that will be treated in the next section.

3. Elastic theory

The elastic properties of two-phase materials depend on the geometrical nature of the mixture (microstructure) and on the volume fraction of the two media. Such a composite material can be thought as a heterogeneous solid continuum that bonds together two homogeneous continua: each part of the media has a well-defined sharp boundary. The bonding at the interfaces remains intact in our models when the whole mixture is placed in an equilibrated state of infinitesimal elastic strain by external loads or constraints. In the present case, the boundary conditions require that both the vector displacement and the stress tensor be continuous across any interfaces. Each separate homogeneous region is characterised by its stiffness tensor, which describes the stress–strain relation. If both materials are linear and homogeneous this relation is given by:

$$\mathbf{T}_{ij} = \mathbf{L}^{s}_{ijkl}\mathbf{E}_{kl} \quad s = 1,2 \tag{27}$$

where $\mathbf{T}$ is the stress tensor (3×3 sized), $\mathbf{E}$ is the strain tensor (3×3 sized) and $\mathbf{L}$ is the constant stiffness tensor, which depends on the medium considered ($s = 1,2$). For isotropic media this latter is written, for example in terms of the bulk and shear constants, as follows:

$$\mathbf{L}^{s}_{ijkl} = k_s\delta_{ij}\delta_{kl} + 2\mu_s\left(\delta_{ik}\delta_{jl} - \frac{1}{3}\delta_{ij}\delta_{kl}\right) \quad s = 1,2 \tag{28}$$

where k_s and μ_s are the bulk and shear moduli of the sth medium ($s = 1,2$) and δ_{nm} is the Kronecker's delta. To solve a mixture problem consists in finding the equivalent macroscopic stiffness tensor for the whole composite material. We start with some definitions used to simplify the problem. Instead of describing the strain with the complete symmetric tensor we adopt a vector, which contains the six independent elements in a given order; the same approach is used for the stress T means transposed):

$$\widehat{\mathbf{E}} = [E_{11}\ E_{22}\ E_{33}\ E_{12}\ E_{23}\ E_{13}]^{\mathrm{T}}; \quad \widehat{\mathbf{T}} = [T_{11}\ T_{22}\ T_{33}\ T_{12}\ T_{23}\ T_{13}]^{\mathrm{T}} \tag{29}$$

Adopting this notation scheme the stiffness four-index tensor for the isotropic components is represented by a simpler matrix with six rows and six columns:

$$\widehat{\mathbf{L}}^{s}=\begin{bmatrix} k_s+\frac{4}{3}\mu_s & k_s-\frac{2}{3}\mu_s & k_s-\frac{2}{3}\mu_s & 0 & 0 & 0 \\ k_s-\frac{2}{3}\mu_s & k_s+\frac{4}{3}\mu_s & k_s-\frac{2}{3}\mu_s & 0 & 0 & 0 \\ k_s-\frac{2}{3}\mu_s & k_s-\frac{2}{3}\mu_s & k_s+\frac{4}{3}\mu_s & 0 & 0 & 0 \\ 0 & 0 & 0 & 2\mu_s & 0 & 0 \\ 0 & 0 & 0 & 0 & 2\mu_s & 0 \\ 0 & 0 & 0 & 0 & 0 & 2\mu_s \end{bmatrix} \quad s=1,2 \tag{30}$$

so that the stress–strain relations became $\widehat{\mathbf{T}}=\widehat{\mathbf{L}}^{1}\widehat{\mathbf{E}}$ in the matrix and $\widehat{\mathbf{T}}=\widehat{\mathbf{L}}^{2}\widehat{\mathbf{E}}$ inside each inclusion. At this point, to begin the strain computation we take into consideration a single ellipsoidal isotropic inclusion (medium 2) embedded in a isotropic matrix (medium 1); we suppose that the matrix is placed in an equilibrated state of infinitesimal constant elastic strain by external loads and then the inclusion is added to the matrix reaching a corresponding state of strain, which is well described by the Eshelby theory [27,28]. In particular it is important to notice that the internal strain is constant (all the entries are constant) if the external or bulk strain is constant. The Eshelby theory allows us to write down a relationship between the internal and original strain when they are constant (or uniform) in the space. Accordingly with the Eshelby theory [27,29] the relationship between the original external strain and the induced internal strain is given by:

$$\widehat{\mathbf{E}}_i=\left\{\mathbf{I}-\widehat{\mathbf{S}}\left[\mathbf{I}-(\widehat{\mathbf{L}}^{\mathbf{1}})^{-1}\widehat{\mathbf{L}}^{\mathbf{2}}\right]\right\}^{-1}\widehat{\mathbf{E}}_0=\widehat{\mathbf{A}}\widehat{\mathbf{E}}_0 \tag{31}$$

where $\mathbf{I}$ is the identity matrix with size 6×6, $\widehat{\mathbf{E}}_i$ is the internal strain, $\widehat{\mathbf{E}}_0$ is the original external strain, $\widehat{\mathbf{L}}^{1}$ and $\widehat{\mathbf{L}}^{2}$ are the stiffness tensor of the matrix and the inclusion respectively and $\widehat{\mathbf{S}}$ is the Eshelby tensor, which depends on the eccentricity e of the ellipsoid of rotation and on the Poisson ratio $v=(3k_1-2\mu_1)/\ [2(3k_1+\mu_1)]$ of the matrix. Here, we remember that the general structure of $\widehat{\mathbf{S}}$ is given by:

$$\widehat{\mathbf{S}}=\begin{bmatrix} s_{1111} & s_{1122} & s_{1133} & 0 & 0 & 0 \\ s_{1122} & s_{1111} & s_{1133} & 0 & 0 & 0 \\ s_{3311} & s_{3311} & s_{3333} & 0 & 0 & 0 \\ 0 & 0 & 0 & s_{1111}-s_{1122} & 0 & 0 \\ 0 & 0 & 0 & 0 & s_{1313} & 0 \\ 0 & 0 & 0 & 0 & 0 & s_{1313} \end{bmatrix} \tag{32}$$

where the symmetries are evident and correctly describe the ellipsoid of rotation which has two equivalent axes and a third one with different behaviour. In Table 1 one can find the complete expressions of all the entries of the tensor defined in Eq. (32). Moreover, see Appendix A for some special cases of the Eshelby tensor. The depolarisation factor L, which appears inside expressions in Table 1, may be computed by means of Eq. (20) and the result depends on the eccentricity e.

Matrix $\widehat{\mathbf{A}}$ is simply defined by Eq. (31). We remember that Eq. (31) is written taking into account a particular reference frame with axes aligned to the three principal directions of the

Table 1
List of the complete expressions of all the entries of the Eshelby tensor defined in Eq. (32)

s_{1111}	$\frac{1}{8}\frac{-3e^2+13L-4e^2L+8Lve^2-8Lv}{(e^2-1)(-1+v)}$
s_{1122}	$-\frac{1}{8}\frac{e^2+L-4e^2L+8Lve^2-8Lv}{(e^2-1)(-1+v)}$
s_{1133}	$-\frac{1}{2}\frac{2e^2L-e^2+L+2Lve^2-2Lv}{(e^2-1)(-1+v)}$
s_{3311}	$\frac{1}{2}\frac{-L+e^2-2e^2L-2ve^2+2v+4Lve^2-4Lv}{(e^2-1)(-1+v)}$
s_{3333}	$-\frac{2e^2-1-4e^2L+L-ve^2+v+2Lve^2-2Lv}{(e^2-1)(-1+v)}$
s_{1313}	$-\frac{1}{2}\frac{e^2L+2L-1+Lve^2-Lv-ve^2+v}{(e^2-1)(-1+v)}$

It corresponds to spheroids with eccentricity e and depolarisation factor L. The symbol v represents the Poisson ratio of the matrix.

embedded ellipsoid. In these conditions matrix $\widehat{\mathbf{A}}$ has the following mathematical form (which derives from the corresponding form of the Eshelby tensor, Eq. (32)):

$$\widehat{\mathbf{A}} = \begin{bmatrix} t_{1111} & t_{1122} & t_{1133} & 0 & 0 & 0 \\ t_{1122} & t_{1111} & t_{1133} & 0 & 0 & 0 \\ t_{3311} & t_{3311} & t_{3333} & 0 & 0 & 0 \\ 0 & 0 & 0 & t_{1111}-t_{1122} & 0 & 0 \\ 0 & 0 & 0 & 0 & t_{1313} & 0 \\ 0 & 0 & 0 & 0 & 0 & t_{1313} \end{bmatrix} \tag{33}$$

Obviously, by using Eqs. (31) and (32), it could be possible to write down the explicit expressions giving each element t_{ijkl} as function of k_1, k_2, μ_1, μ_2, e but these formulas are very complicated and not particularly useful at this stage of the work. Moreover, all the coefficients t_{ijkl} that not appear in Eq. (33) are always zero. With the aim of analysing the behaviour of a mixture of pseudo-oriented ellipsoids, we need to evaluate the average value of the internal strain inside the ellipsoid over all its possible orientations or rotations in the space (in agreement with the given orientational distribution). To perform this averaging over the rotations we name the original reference frame with the letter B and we consider another generic reference frame that is named with the letter F.

The relation between these bases B and F is described by means of a generic rotation matrix $\mathbf{R}(\psi,\theta,\varphi)$ where ψ, θ and φ are the Euler angles; we may consider this matrix as the product of three elementary rotations along the axes z, x and z respectively:

$$\mathbf{R}(\psi,\theta,\varphi) = \begin{bmatrix} \cos\psi & -\sin\psi & 0 \\ \sin\psi & \cos\psi & 0 \\ 0 & 0 & 1 \end{bmatrix} \cdot \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos\theta & -\sin\theta \\ 0 & \sin\theta & \cos\theta \end{bmatrix} \cdot \begin{bmatrix} \cos\varphi & -\sin\varphi & 0 \\ \sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{bmatrix} \tag{34}$$

The angle that defines the pseudo-orientational distribution is θ. Therefore the following relations hold on between the different frames: $\mathbf{E}_i^B = \mathbf{R}\mathbf{E}_i^F\mathbf{R}^{\mathrm{T}}$ for the internal strain and $\mathbf{E}_0^B = \mathbf{R}\mathbf{E}_0^F\mathbf{R}^{\mathrm{T}}$ for the bulk strain (here the subscript T means transposed). These expressions have been written with standard notation for the strain (3×3 sized matrix). They may be converted in our notation defining a matrix $\widehat{\mathbf{M}}(\psi, \theta, \varphi)$, 6×6 sized, which acts as a rotation matrix on our strain vectors: so, we may write $\widehat{\mathbf{E}}_i^B = \widehat{\mathbf{M}}\widehat{\mathbf{E}}_i^F$ inside the ellipsoid and $\widehat{\mathbf{E}}_0^B = \widehat{\mathbf{M}}\widehat{\mathbf{E}}_0^F$ outside it. The entries of the matrix $\widehat{\mathbf{M}}$ are completely defined by the comparison between the relations $\mathbf{E}_i^B = \mathbf{R}\mathbf{E}_i^F\mathbf{R}^{\mathrm{T}}$ and $\widehat{\mathbf{E}}_i^B = \widehat{\mathbf{M}}\widehat{\mathbf{E}}_i^F$ and by considering the notation adopted for the strain. Eq. (31) is written on the frame B and therefore it actually reads $\widehat{\mathbf{E}}_i^B = \widehat{\mathbf{A}}\widehat{\mathbf{E}}_0^B$; this latter may be reformulated on the generic frame F simply obtaining:

$$\widehat{\mathbf{E}}_i^F = \left\{\widehat{\mathbf{M}}(\Psi, \theta, \varphi)^{-1}\widehat{\mathbf{A}}\widehat{\mathbf{M}}(\Psi, \theta, \varphi)\right\}\widehat{\mathbf{E}}_0^F \tag{35}$$

The first average value of the strain inside the inclusion may be computed by means of the following integration over all the possible rotations over the angles φ and ψ (they are uniformly distributed over the whole range $[0\ 2\pi]$):

$$\left\langle\widehat{\mathbf{E}}_i\right\rangle_{\Psi,\varphi} = \frac{1}{4\pi^2}\int_0^{2\pi}\int_0^{2\pi}\left\{\widehat{\mathbf{M}}(\Psi, \theta, \varphi)^{-1}\widehat{\mathbf{A}}\widehat{\mathbf{M}}(\Psi, \theta, \varphi)\right\}\mathrm{d}\varphi\,\mathrm{d}\Psi\widehat{\mathbf{E}}_0 \tag{36}$$

By means of a very long but straightforward integration we have obtained an explicit relation between the external strain $\widehat{\mathbf{E}}_0(=\widehat{\mathbf{E}}_0^F)$ and the first average value $\langle\widehat{\mathbf{E}}_i\rangle_{\Psi,\varphi}$ inside the randomly oriented ellipsoid:

$$\langle\widehat{\mathbf{E}}_i\rangle_{\Psi,\varphi} = \begin{bmatrix} b_{1111} & b_{1122} & b_{1133} & 0 & 0 & 0 \\ b_{1122} & b_{1111} & b_{1133} & 0 & 0 & 0 \\ b_{3311} & b_{3311} & b_{3333} & 0 & 0 & 0 \\ 0 & 0 & 0 & b_{1111}-b_{1122} & 0 & 0 \\ 0 & 0 & 0 & 0 & b_{1313} & 0 \\ 0 & 0 & 0 & 0 & 0 & b_{1313} \end{bmatrix}\widehat{\mathbf{E}}_0 = \widehat{\mathbf{B}}(\theta)\widehat{\mathbf{E}}_0 \tag{37}$$

where the parameters b_{ijkl} depend on the coefficients t_{ijkl} and the angle θ, which is, still now, undefined. The results are summarised in Table 2 where the explicit expressions of all the coefficients b_{ijkl} are shown.

Finally, now we may perform the second averaging over the angle θ described by an arbitrary probability density $f(\theta)$ defined on the range $[0, \pi]$:

$$\langle\widehat{\mathbf{E}}_i\rangle_{\Psi,\varphi,\theta} = \int_0^{\pi} f(\theta)\widehat{\mathbf{B}}(\theta)\,\mathrm{d}\theta\widehat{\mathbf{E}}_0 = \widehat{\mathbf{C}}\widehat{\mathbf{E}}_0 = \begin{bmatrix} c_{1111} & c_{1122} & c_{1133} & 0 & 0 & 0 \\ c_{1122} & c_{1111} & c_{1133} & 0 & 0 & 0 \\ c_{3311} & c_{3311} & c_{3333} & 0 & 0 & 0 \\ 0 & 0 & 0 & c_{1111}-c_{1122} & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{1313} & 0 \\ 0 & 0 & 0 & 0 & 0 & c_{1313} \end{bmatrix}\widehat{\mathbf{E}}_0 \tag{38}$$

Table 2
List of the complete expressions of all the entries of the tensor $\widehat{\mathbf{B}}(\theta)$ defined in Eq. (37)

b_{1111}	$\left(\frac{1}{2}\cos(\theta)^2-\frac{3}{4}\cos(\theta)^4+\frac{1}{4}\right)t_{1313}+\left(-\frac{3}{4}\cos(\theta)^2+\frac{3}{8}\cos(\theta)^4+\frac{3}{8}\right)t_{3333}+\left(-\frac{3}{8}\cos(\theta)^4+\frac{1}{4}\cos(\theta)^2+\frac{1}{8}\right)t_{3311}$ $+\left(-\frac{3}{8}\cos(\theta)^4+\frac{1}{4}\cos(\theta)^2+\frac{1}{8}\right)t_{1133}+\left(\frac{3}{8}+\frac{1}{4}\cos(\theta)^2+\frac{3}{8}\cos(\theta)^4\right)t_{1111}$
b_{1122}	$\left(-\frac{1}{4}+\frac{1}{2}\cos(\theta)^2-\frac{1}{4}\cos(\theta)^4\right)t_{1313}+\left(-\frac{1}{4}\cos(\theta)^2+\frac{1}{8}\cos(\theta)^4+\frac{1}{8}\right)t_{3333}+\left(-\frac{1}{4}\cos(\theta)^2+\frac{3}{8}-\frac{1}{8}\cos(\theta)^4\right)t_{3311}$ $+\left(-\frac{1}{4}\cos(\theta)^2+\frac{3}{8}-\frac{1}{8}\cos(\theta)^4\right)t_{1133}+\left(-\frac{1}{4}\cos(\theta)^2+\frac{1}{8}\cos(\theta)^4+\frac{1}{8}\right)t_{1111}+t_{1122}\cos(\theta)^2$
b_{1133}	$(\cos(\theta)^4-\cos(\theta)^2)t_{1313}+\left(\frac{1}{2}\cos(\theta)^2-\frac{1}{2}\cos(\theta)^4\right)t_{3333}+\left(\frac{1}{2}-\cos(\theta)^2+\frac{1}{2}\cos(\theta)^4\right)t_{3311}$ $+\left(\frac{1}{2}\cos(\theta)^4+\frac{1}{2}\cos(\theta)^2\right)t_{1133}+\left(\frac{1}{2}-\frac{1}{2}\cos(\theta)^2\right)t_{1122}+\left(\frac{1}{2}\cos(\theta)^2-\frac{1}{2}\cos(\theta)^4\right)t_{1111}$
b_{3311}	$(\cos(\theta)^4-\cos(\theta)^2)t_{1313}+\left(\frac{1}{2}\cos(\theta)^2-\frac{1}{2}\cos(\theta)^4\right)t_{3333}+\left(\frac{1}{2}\cos(\theta)^4+\frac{1}{2}\cos(\theta)^2\right)t_{3311}$ $+\left(\frac{1}{2}-\cos(\theta)^2+\frac{1}{2}\cos(\theta)^4\right)t_{1133}+\left(\frac{1}{2}-\frac{1}{2}\cos(\theta)^2\right)t_{1122}+\left(\frac{1}{2}\cos(\theta)^2-\frac{1}{2}\cos(\theta)^4\right)t_{1111}$
b_{3333}	$(-2\cos(\theta)^4+2\cos(\theta)^2)t_{1313}+(-\cos(\theta)^4+\cos(\theta)^2)t_{3311}+(-\cos(\theta)^4+\cos(\theta)^2)t_{1133}$ $+(1-2\cos(\theta)^2+\cos(\theta)^4)t_{1111}+t_{3333}\cos(\theta)^4$
b_{1313}	$\left(\frac{1}{2}-\frac{3}{2}\cos(\theta)^2+2\cos(\theta)^4\right)t_{1313}+(-\cos(\theta)^4+\cos(\theta)^2)t_{3333}+(\cos(\theta)^4-\cos(\theta)^2)t_{3311}$ $+(\cos(\theta)^4-\cos(\theta)^2)t_{1133}+\left(-\frac{1}{2}+\frac{1}{2}\cos(\theta)^2\right)t_{1122}+\left(\frac{1}{2}\cos(\theta)^2-\cos(\theta)^4+\frac{1}{2}\right)t_{1111}$

We define two order parameters S and T as follows:

$$S=\langle P_2(\cos\theta)\rangle_\theta=\left\langle\frac{3}{2}\cos^2(\theta)-\frac{1}{2}\right\rangle_\theta=\int_0^\pi\left(\frac{3}{2}\cos^2(\theta)-\frac{1}{2}\right)f(\theta)\,\mathrm{d}\theta \tag{39}$$

$$\begin{aligned}T=\langle P_4(\cos\theta)\rangle_\theta&=\left\langle\frac{35}{8}\cos^4(\theta)-\frac{15}{4}\cos^2(\theta)+\frac{3}{8}\right\rangle_\theta\\&=\int_0^\pi\left(\frac{35}{8}\cos^4(\theta)-\frac{15}{4}\cos^2(\theta)+\frac{3}{8}\right)f(\theta)\,\mathrm{d}\theta\end{aligned} \tag{40}$$

They correspond to the average values of the Legendre polynomial of order two and four, computed by means of the density probability $f(\theta)$ defined on the range $[0,\pi]$. In Table 3 one can find the complete expressions of all the entries of the tensor $\widehat{\mathbf{C}}$ defined in Eq. (38). Once again, the coefficients c_{ijkl} could be explicitly written in terms of k_1, k_2, μ_1, μ_2, e, S and T; we prefer to recall the main stages of the procedure applied to obtain them. Firstly, we evaluate the matrix $\widehat{\mathbf{A}}$ by means of Eq. (31) (using the pertinent Eshelby tensor) and then we apply Eq. (38) and Table 3 to average the internal strain over the possible orientation of the ellipsoid (micro-mechanical averaging technique). In other words we may say that the matrix $\widehat{\mathbf{C}}$ represents the average value of $\widehat{\mathbf{A}}$ over all the possible rotations of the inclusion. We wish to point out that the expressions given in Table 3 are extremely convenient to perform the micro-mechanical averaging because it removes the problem of the integral evaluation and allows us to obtain results in closed form.

The two order parameters S and T defined in Eqs. (39) and (40) are subjected to the following constraints: $-1/2<S<1$ and $-3/7<T<1$. A point in the S–T plane, as indicated in Fig. 3,

Table 3
List of the complete expressions of all the entries of the tensor $\widehat{\mathbf{C}}$ defined in Eq. (38) in terms of the order parameters S and T

c_{1111}	$\left(-\frac{2}{21}S+\frac{4}{15}-\frac{6}{35}T\right)t_{1313}+\left(-\frac{2}{7}S+\frac{1}{5}+\frac{3}{35}T\right)t_{3333}+\left(-\frac{3}{35}T-\frac{1}{21}S+\frac{2}{15}\right)t_{3311}$ $+\left(-\frac{3}{35}T-\frac{1}{21}S+\frac{2}{15}\right)t_{1133}+\left(\frac{8}{15}+\frac{8}{21}S+\frac{3}{35}T\right)t_{1111}$
c_{1122}	$\left(-\frac{2}{15}+\frac{4}{21}S-\frac{2}{35}T\right)t_{1313}+\left(-\frac{2}{21}S+\frac{1}{15}+\frac{1}{35}T\right)t_{3333}+\left(-\frac{5}{21}S+\frac{4}{15}-\frac{1}{35}T\right)t_{3311}$ $+\left(-\frac{5}{21}S+\frac{4}{15}-\frac{1}{35}T\right)t_{1133}+\left(-\frac{2}{21}S+\frac{1}{15}+\frac{1}{35}T\right)t_{1111}+t_{1122}\left(\frac{2}{3}S+\frac{1}{3}\right)$
c_{1133}	$\left(\frac{8}{35}T-\frac{2}{21}S-\frac{2}{15}\right)t_{1313}+\left(\frac{1}{21}S+\frac{1}{15}-\frac{4}{35}T\right)t_{3333}+\left(\frac{4}{15}-\frac{8}{21}S+\frac{4}{35}T\right)t_{3311}$ $+\left(\frac{4}{35}T+\frac{13}{21}S+\frac{4}{15}\right)t_{1133}+\left(\frac{1}{3}-\frac{1}{3}S\right)t_{1122}+\left(\frac{1}{21}S+\frac{1}{15}-\frac{4}{35}T\right)t_{1111}$
c_{3311}	$\left(\frac{8}{35}T-\frac{2}{21}S-\frac{2}{15}\right)t_{1313}+\left(\frac{1}{21}S+\frac{1}{15}-\frac{4}{35}T\right)t_{3333}+\left(\frac{4}{35}T+\frac{13}{21}S+\frac{4}{15}\right)t_{3311}$ $+\left(\frac{4}{15}-\frac{8}{21}S+\frac{4}{35}T\right)t_{1133}+\left(\frac{1}{3}-\frac{1}{3}S\right)t_{1122}+\left(\frac{1}{21}S+\frac{1}{15}-\frac{4}{35}T\right)t_{1111}$
c_{3333}	$\left(-\frac{16}{35}T+\frac{4}{21}S+\frac{4}{15}\right)t_{1313}+\left(-\frac{8}{35}T+\frac{2}{21}S+\frac{2}{15}\right)t_{3311}+\left(-\frac{8}{35}T+\frac{2}{21}S+\frac{2}{15}\right)t_{1133}$ $+\left(\frac{8}{15}-\frac{16}{21}S+\frac{8}{35}T\right)t_{1111}+t_{3333}\left(\frac{8}{35}T+\frac{4}{7}S+\frac{1}{5}\right)$
c_{1313}	$\left(\frac{1}{7}S+\frac{16}{35}T+\frac{2}{5}\right)t_{1313}+\left(-\frac{8}{35}T+\frac{2}{21}S+\frac{2}{15}\right)t_{3333}+\left(\frac{8}{35}T-\frac{2}{21}S-\frac{2}{15}\right)t_{3311}$ $+\left(\frac{8}{35}T-\frac{2}{21}S-\frac{2}{15}\right)t_{1133}+\left(-\frac{1}{3}+\frac{1}{3}S\right)t_{1122}+\left(-\frac{5}{21}S+\frac{7}{15}-\frac{8}{35}T\right)t_{1111}$

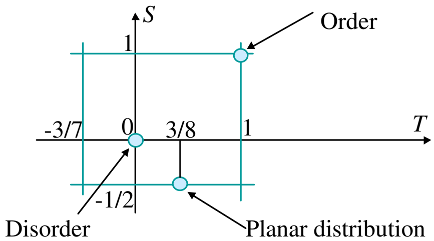


Fig. 3. The degree of orientational order is represented by a point in the S–T plane. Three particular cases of state of order can be observed: if $S = T = 1$ we are in the state of order, if $S = T = 0$ we are in the state of disordered and if $S = -1/2$ and $T = 3/8$ all particles are lying randomly in planes perpendicular to the z-axis.

represents the degree of orientational order. Three particular cases of state of order can be taken into consideration: if $S = T = 1$ we are in the state of order (particles aligned), if $S = T = 0$ we are in the state of disorder (particles randomly oriented) and, finally, if $S = -1/2$ and $T = 3/8$ all particles are lying randomly in planes perpendicular to the z-axis. These three cases are indicated in Fig. 3. The results displayed in Table 3 may be simplified in such particular cases. If $S = T = 1$ the averaging procedure over rotations is not important because of the alignment of the particles and this relationship holds on among the coefficients: $c_{ijkl} = t_{ijkl}$. If $S = T = 0$ we are dealing with randomly oriented particles and the relations reduce to overall isotropic behaviour:

$$\begin{cases} c_{1111} = c_{3333} = \frac{4}{15}t_{1313} + \frac{1}{5}t_{3333} + \frac{2}{15}t_{3311} + \frac{2}{15}t_{1133} + \frac{8}{15}t_{1111} \\ c_{1122} = c_{1133} = c_{3311} = -\frac{2}{15}t_{1313} + \frac{1}{15}t_{3333} + \frac{4}{15}t_{3311} + \frac{4}{15}t_{1133} + \frac{1}{15}t_{1111} + \frac{1}{3}t_{1122} \\ c_{1313} = c_{1111} - c_{1122} \end{cases} \tag{41}$$

If $S = -1/2$ and $T = 3/8$ all the particles have the axes of rotation orthogonal to the z-axis of the main reference frame and therefore they are lying randomly in planes perpendicular to the z-axis. The results of Table 3 reduce to the following:

$$\begin{cases} c_{1111} = \frac{1}{4}t_{1313} + \frac{3}{8}t_{3333} + \frac{1}{8}t_{3311} + \frac{1}{8}t_{1133} + \frac{3}{8}t_{1111} \\ c_{3333} = t_{1111} \\ c_{1122} = -\frac{1}{4}t_{1313} + \frac{1}{8}t_{3333} + \frac{3}{8}t_{3311} + \frac{3}{8}t_{1133} + \frac{1}{8}t_{1111} \\ c_{1133} = \frac{1}{2}t_{3311} + \frac{1}{2}t_{1122} \\ c_{3311} = \frac{1}{2}t_{1133} + \frac{1}{2}t_{1122} \\ c_{1313} = \frac{1}{2}t_{1313} - \frac{1}{2}t_{1122} + \frac{1}{2}t_{1111} \end{cases} \tag{42}$$

Still now, we have considered a single ellipsoidal particle and we have calculated the averaged internal strain $\langle \widehat{\mathbf{E}}_i \rangle = \langle \widehat{\mathbf{E}}_i \rangle_{\Psi,\varphi,\theta}$ when it is pseudo-randomly placed in a matrix with a given constant strain $\widehat{\mathbf{E}}_0$; from now on, we have to deal with an ensemble of inclusions (see Fig. 1) pseudo-oriented and distributed in the solid matrix with order parameters S and T. We consider a low value of the volume fraction of the dispersed component so that we may neglect the interactions among the inclusions. Therefore, each ellipsoidal particle behaves as a single one in the whole space. As before, we define c as the volume fraction of the inclusions. We may compute the average value of the elastic strain over the whole heterogeneous material by means of the relation:

$$\langle \widehat{\mathbf{E}} \rangle = (1-c)\widehat{\mathbf{E}}_0 + c\langle \widehat{\mathbf{E}}_i \rangle = \left[(1-c)\mathbf{I} + c\widehat{\mathbf{C}}\right]\widehat{\mathbf{E}}_0 \tag{43}$$

where we have considered the strain outside the inclusions approximately constant and identical to the bulk strain $\widehat{\mathbf{E}}_0$. Moreover, we define $\widehat{\mathbf{L}}_{eq}$ as the equivalent stiffness tensor of the whole mixture (which is anisotropic because of the pseudo-randomness of the orientations of the inclusions) by means of the relation $\langle \widehat{\mathbf{T}} \rangle = \widehat{\mathbf{L}}_{eq} \langle \widehat{\mathbf{E}} \rangle$; to evaluate $\widehat{\mathbf{L}}_{eq}$ we compute the average value $\langle \widehat{\mathbf{T}} \rangle$ of the stress 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_0 as the volume of the remaining space among the inclusions (so that $V = V_e \cup V_0$). The average value of $\widehat{\mathbf{T}} = \widehat{\mathbf{L}}(\bar{r})\widehat{\mathbf{E}}$ over the volume of the whole material is evaluated as follows ($\widehat{\mathbf{L}}(\bar{r}) = \widehat{\mathbf{L}}^1$ if $\bar{r} \in V_0$ and $\widehat{\mathbf{L}}(\bar{r}) = \widehat{\mathbf{L}}^2$ if $\bar{r} \in V_e$):

$$
\begin{aligned}
\langle\widehat{\mathbf{T}}\rangle &= \frac{1}{V}\int_V \widehat{\mathbf{L}}(\bar{r})\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} = \frac{1}{V}\widehat{\mathbf{L}}^1\int_{V_0}\widehat{\mathbf{E}}(\bar{r})d\bar{r} + \frac{1}{V}\widehat{\mathbf{L}}^2\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} \\
&= \frac{1}{V}\widehat{\mathbf{L}}^1\int_{V_0}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\widehat{\mathbf{L}}^2\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} + \frac{1}{V}\widehat{\mathbf{L}}^1\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} - \frac{1}{V}\widehat{\mathbf{L}}^1\int_{V_e}\widehat{\mathbf{E}}(\bar{r})\,\mathrm{d}\bar{r} \\
&= \widehat{\mathbf{L}}^1\langle\widehat{\mathbf{E}}\rangle + c(\widehat{\mathbf{L}}^2 - \widehat{\mathbf{L}}^1)\langle\widehat{\mathbf{E}}_i\rangle = \widehat{\mathbf{L}}^1\langle\widehat{\mathbf{E}}\rangle + c\left(\widehat{\mathbf{L}}^2 - \widehat{\mathbf{L}}^1\right)\widehat{\mathbf{C}}\widehat{\mathbf{E}}_0
\end{aligned} \tag{44}
$$

Drawing a comparison between Eqs. (43) and (44) we may find a complete expression, which allows us to estimate the equivalent stiffness tensor $\widehat{\mathbf{L}}_{\mathrm{eq}}$:

$$
\widehat{\mathbf{L}}_{\mathrm{eq}} = \widehat{\mathbf{L}}^1 + c\left(\widehat{\mathbf{L}}^2 - \widehat{\mathbf{L}}^1\right)\widehat{\mathbf{C}}\left[(1-c)\mathbf{I} + c\widehat{\mathbf{C}}\right]^{-1} \tag{45}
$$

It is a very long but straightforward task to verify that the general form of $\widehat{\mathbf{L}}_{\mathrm{eq}}$ is given by the following expression, in perfect agreement with transversely isotropic composites:

$$
\widehat{\mathbf{L}}_{\mathrm{eq}} = \begin{bmatrix}
k+m & k-m & l & 0 & 0 & 0 \\
k-m & k+m & l & 0 & 0 & 0 \\
l & l & n & 0 & 0 & 0 \\
0 & 0 & 0 & 2m & 0 & 0 \\
0 & 0 & 0 & 0 & 2p & 0 \\
0 & 0 & 0 & 0 & 0 & 2p
\end{bmatrix} \tag{46}
$$

The parameters here involved are known as Hill parameters [30]. A transversely isotropic material is always described by five elastic moduli as indicated. At the end of this procedure, they are depending on k_1, k_2, μ_1, μ_2, S, T, L (or the eccentricity e) and on the volume fraction c. Some particular cases follow. For spheres the averaging technique over all the orientations is not necessary but it may be used as check of the procedure. Anyway, the final result is given by the following relationships, which explicitly give the bulk modulus and the shear modulus of the overall isotropic composite material:

$$
\begin{aligned}
k_{\mathrm{eq}} &= \frac{k_1(4\mu_1 + 3k_2) + 4c\mu_1(k_2 - k_1)}{4\mu_1 + 3k_2 - 3c(k_2 - k_1)} = k_1 + \frac{4\mu_1 + 3k_1}{4\mu_1 + 3k_2}(k_2 - k_1)c + \mathrm{O}(c^2) \\
\mu_{\mathrm{eq}} &= \mu_1\frac{[(1-c)\mu_1 + c\mu_2](9k_1 + 8\mu_1) + 6\mu_2(k_1 + 2\mu_1)}{\mu_1(9k_1 + 8\mu_1) + 6[(1-c)\mu_2 + c\mu_1](k_1 + 2\mu_1)} \\
&= \mu_1 + \frac{5\mu_1(4\mu_1 + 3k_1)(\mu_2 - \mu_1)}{\mu_1(9k_1 + 8\mu_1) + 6\mu_2(k_1 + 2\mu_1)}c + \mathrm{O}(c^2)
\end{aligned} \tag{47}
$$

This is the well known result obtained by several authors in the earlier literature as described in [18,19]. When we adopt the Eshelby tensor for spheres (see Appendix A), the complete procedure furnishes, as results, Eq. (47) independently on the values of the order parameters S and T. A particular attention will be given to fibrous materials where the fibre may be considered in three different arrangements: randomly oriented in the space ($S = T = 0$) , aligned along a given direction ($S = T = 1$) and lying randomly in planes perpendicular to a given direction ($S = -1/2$ and $T = 3/8$). If we consider the characteristic Eshelby tensor (see Appendix A) for cylinders and we apply the previously outlined procedure with $S = T = 0$ (randomly oriented cylinders in the space), in closed form, we obtain, after a long and tedious algebraic computation, the explicit results:

$$k_{\mathrm{eq}} = k_1 + \frac{\mu_2 + 3\mu_1 + 3k_1}{\mu_2 + 3\mu_1 + 3k_2}(k_2 - k_1)c + \mathrm{O}(c^2) \tag{48}$$

$$\mu_{\mathrm{eq}} = \mu_1 + \frac{1}{5}\frac{(\mu_2-\mu_1)\left\{\begin{array}{l}64\mu_1^4+63\mu_1^3k_2+184\mu_1^3\mu_2+156\mu_1^2k_2\mu_2+72\mu_1^2\mu_2^2+90k_1k_2\mu_1\mu_2+\\120k_1\mu_1^2\mu_2+81k_1\mu_1^2k_2+36k_1\mu_2^2\mu_1+21k_2\mu_1\mu_2^2+9k_1k_2\mu_2^2+84k_1\mu_1^3\end{array}\right\}}{(\mu_1+\mu_2)(\mu_2+3k_2+3\mu_1)(3\mu_1k_1+\mu_1^2+3k_1\mu_2+7\mu_1\mu 2)}c+\mathrm{O}(c^2)$$

Here, we have reported, for sake of brevity, only the first order approximations instead of the complete expressions, which are very complicated. The medium is overall isotropic and we have reported bulk and shear modulus. These are relations that hold on for a fibrous material where each fibre or rod is randomly oriented in the space. In Fig. 4 one can find the plots of the elastic moduli versus the volume fraction of fibres. We may observe that the overall Hill parameters for an isotropic medium are given by: $k = k_{\mathrm{eq}} + (1/3)\mu_{\mathrm{eq}}$, $l = k_{\mathrm{eq}} - (2/3)\mu_{\mathrm{eq}}$, $n = (1/2)k_{\mathrm{eq}} + (2/3)\mu_{\mathrm{eq}}$ and $p = m = \mu_{\mathrm{eq}}$.

We may analyse a system of parallel cylinders by considering $S = T = 1$ and adopting the Eshelby tensor of cylindrical particles; we obtain a transversely isotropic material described by the following Hill parameters in the limit of very low values of the volume fraction:

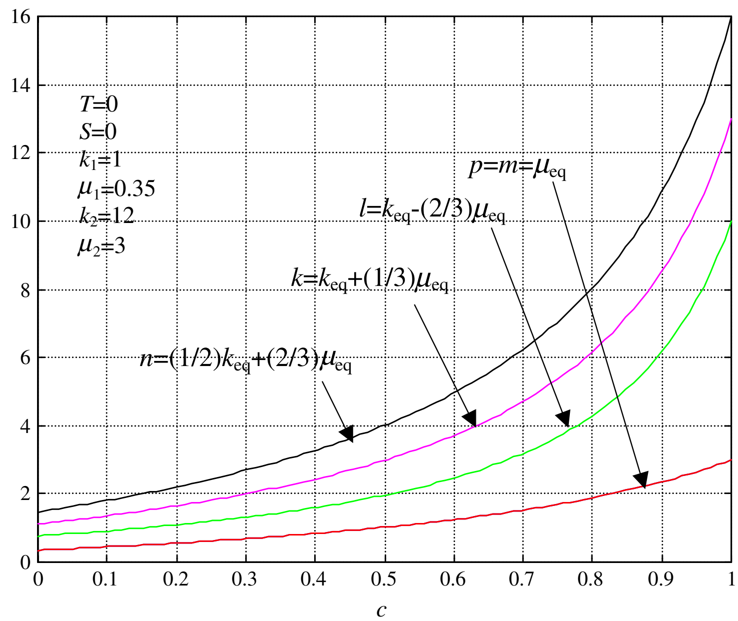


Fig. 4. Plots of the Hill parameters for a fibrous material where each fibre or rod is completely random oriented in the space. The elastic moduli are given in terms of the volume fraction of fibres. In this case $S = T = 0$.

$$
\begin{aligned}
k &= \left(\frac{1}{3}\mu_1 + k_1\right) - \frac{1}{3}\frac{(4\mu_1 + 3k_1)(\mu_1 + 3k_1 - \mu_2 - 3k_2)}{3\mu_1 + \mu_2 + 3k_2}c + \mathrm{O}(c^2) \\
m &= \mu_1 - 2\frac{\mu_1(4\mu_1 + 3k_1)(\mu_1 - \mu_2)}{\mu_1^2 + 3k_1\mu_1 + 7\mu_2\mu_1 + 3k_1\mu_2}c + \mathrm{O}_{(}c^2) \\
l &= \left(k_1 - \frac{2}{3}\mu_1\right) + \frac{1}{3}\frac{(4\mu_1 + 3k_1)(2\mu_1 - 3k_1 + 3k_2 - 2\mu_2)}{3\mu_1 + \mu_2 + 3k_2}c + \mathrm{O}(c^2) \\
n &= \left(\frac{2}{3}\mu_1 + \frac{1}{2}k_1\right) \\
&\quad - \frac{1}{6}\frac{-3k_1\mu_1 + 15k_1\mu_2 + 15\mu_1k_2 - 27k_2\mu_2 - 16\mu_2\mu_1 + 16\mu_1^2 + 9k_1^2 - 9k_2k_1}{3\mu_1 + \mu_2 + 3k_2}c + \mathrm{O}(c^2) \\
p &= \mu_1 - 2\frac{(\mu_1 - \mu_2)\mu_1}{\mu_1 + \mu_2}c + \mathrm{O}(c^2)
\end{aligned} \tag{49}
$$

These are relations that hold on for a fibrous material where each fibre is aligned to a given direction in the space. In Fig. 5 the behaviour of the elastic moduli versus the volume fraction of fibres is shown. Finally, we may take into account a mixture of cylinders lying randomly in planes perpendicular to the z-axis; in this case $S = -1/2$ and $T = 3/8$ and the results are:

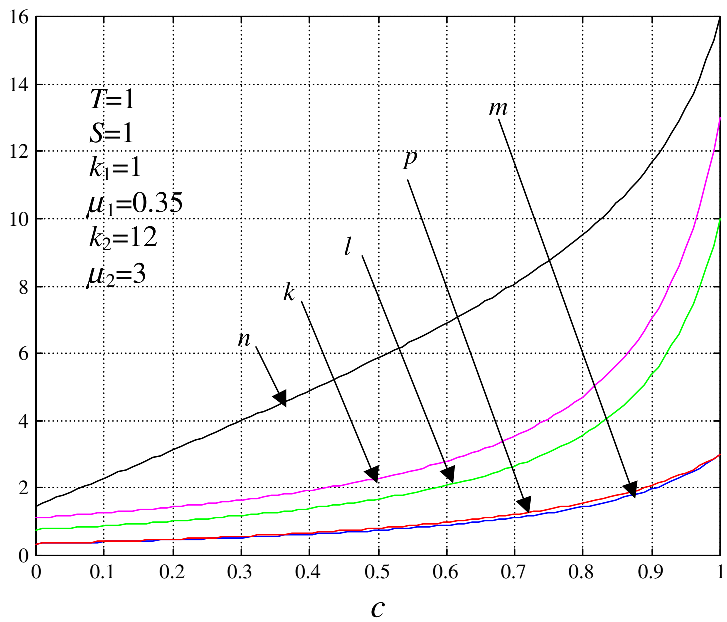


Fig. 5. Plots of the Hill parameters for a fibrous material where each fibre is aligned to a given direction in the space. The behaviour of the elastic moduli versus the volume fraction of fibres is shown. This case corresponds to $S = T = 1$.

$$
\begin{aligned}
k &= \left(\frac{1}{3}\mu_1 + k_1\right) - \frac{1}{12}(-108k_2\mu_2k_1^2 + 108k_1^3\mu_2 + 108k_1^3\mu_1 + 72k_1^2\mu_2^2 \\
&\quad + 108k_1^2\mu_1^2 + 76\mu_1^4 - 450\mu_1k_2\mu_2k_1 - 45\mu_1^2k_2k_1 + 156k_1\mu_2\mu_1^2 \\
&\quad - 246\mu_1^2k_2\mu_2 + 138\mu_1k_1\mu_2^2 - 189\mu_1k_2\mu_2^2 - 81k_2\mu_2^2k_1 + 396k_1^2\mu_2\mu_1 \\
&\quad - 52\mu_2^2\mu_1^2 - 24\mu_2\mu_1^3 + 90k_1\mu_1^3 + 51\mu_1^3k_2 - 108k_2k_1^2\mu_1)/((3\mu_1 + 3k_2 + \mu_2) \\
&\quad \times (\mu_1^2 + 3k_1\mu_1 + 7\mu_2\mu_1 + 3k_1\mu_2))c + \mathrm{O}(c^2) \\
m &= \mu_1 - \frac{1}{8}(\mu_1 - \mu_2)(60\mu_1^4 + 57\mu_1^3k_2 + 304\mu_2\mu_1^3 + 126k_1\mu_1^3 + 264\mu_1^2k_2\mu_2 \\
&\quad + 117\mu_1^2k_2k_1 + 192k_1\mu_2\mu_1^2 + 148\mu_2^2\mu_1^2 + 66\mu_1k_1\mu_2^2 + 63\mu_1k_2\mu_2^2 \\
&\quad + 144\mu_1k_2\mu_2k_1 + 27k_2\mu_2^2k_1)/((\mu_1 + \mu_2)(3\mu_1 + 3k_2 + \mu_2) \\
&\quad \times (\mu_1^2 + 3k_1\mu_1 + 7\mu_2\mu_1 + 3k_1\mu_2))c + \mathrm{O}(c^2) \\
l &= \left(k_1 - \frac{2}{3}\mu_1\right) + \frac{1}{6}(4\mu_1 + 3k_1)(19\mu_1^3 - 6\mu_2\mu_1^2 - 3k_1\mu_1^2 + 24\mu_1^2k_2 \\
&\quad + 24\mu_1k_2\mu_2 - 18k_1^2\mu_1 + 18\mu_1k_2k_1 - 42k_1\mu_2\mu_1 - 13\mu_2^2\mu_1 + 18k_2\mu_2k_1 \\
&\quad - 18k_1^2\mu_2 - 3k_1\mu_2^2)/((3\mu_1 + 3k_2 + \mu_2)(\mu_1^2 + 3k_1\mu_1 + 7\mu_2\mu_1 + 3k_1\mu_2))c + \mathrm{O}(c^2) \\
n &= \left(\frac{2}{3}\mu_1 + \frac{1}{2}k_1\right) - \frac{1}{6}(4\mu_1 + 3k_1)(19\mu_1^3 - 6\mu_2\mu_1^2 + 6k_1\mu_1^2 + 15\mu_1^2k_2 \\
&\quad - 39\mu_1k_2\mu_2 + 9k_1^2\mu_1 - 9\mu_1k_2k_1 + 21k_1\mu_2\mu_1 - 13\mu_2^2\mu_1 - 9k_2\mu_2k_1 \\
&\quad + 9k_1^2\mu_2 - 3k_1\mu_2^2)/((3\mu_1 + 3k_2 + \mu_2)(\mu_1^2 + 3k_1\mu_1 + 7\mu_2\mu_1 + 3k_1\mu_2))c + \mathrm{O}(c^2) \\
p &= \mu_1 - \frac{\mu_1(\mu_1 - \mu_2)(5\mu_1^2 + 6k_1\mu_1 + 11\mu_2\mu_1 + 6k_1\mu_2)}{(\mu_1 + \mu_2)(\mu_1^2 + 3k_1\mu_1 + 7\mu_2\mu_1 + 3k_1\mu_2)}c + \mathrm{O}(c^2)
\end{aligned} \tag{50}
$$

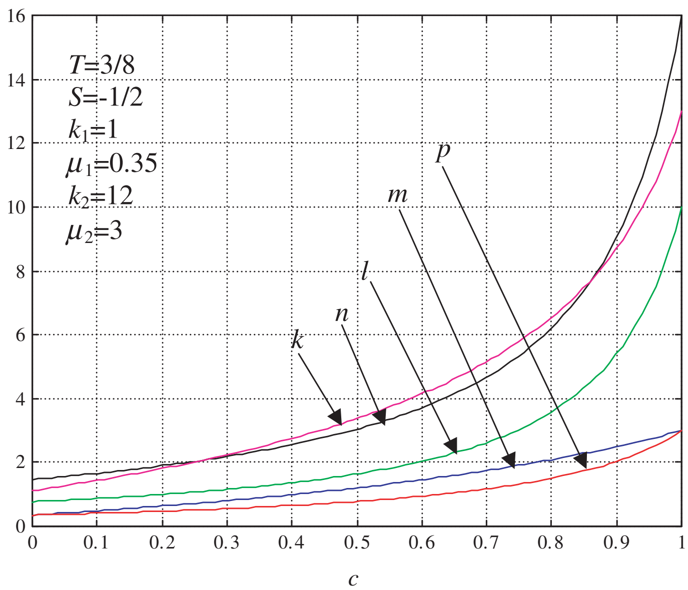


Fig. 6. Hill parameters for a mixture of fibres lying randomly in planes perpendicular to the z-axis; in this case $S = -1/2$ and $T = 3/8$. The results are shown in terms of the volume fraction.

In Fig. 6 the behaviour of the elastic moduli is reported versus the volume fraction of fibres. Finally, we have developed a software code that implements the complete procedure furnishing the five elastic moduli of the overall composite materials formed by non-randomly included spheroids. At the end of this procedure, as above said, they are depending on k_1, k_2, μ_1, μ_2, S, T, c and the eccentricity e: in Fig. 7 one can find several simulations describing different states of order of the material. For each orientational distribution (fixed S and T) we have plotted the five Hill parameters versus the eccentricity value of the embedded spheroids. This is done maintaining the same materials for inclusions and matrix and the same volume fraction of the inhomogeneities.

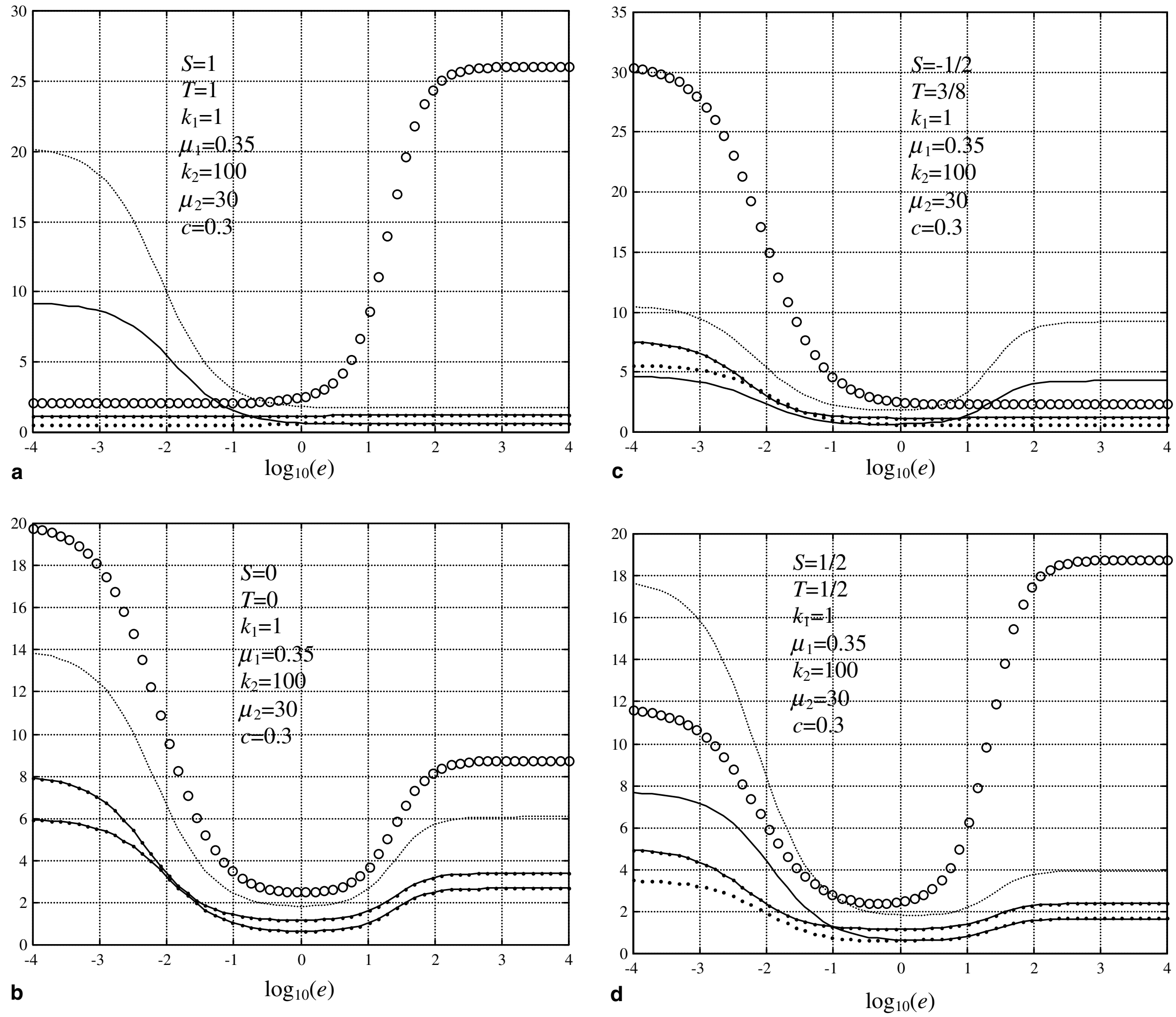


Fig. 7. Several results for the Hill parameters are reported in various states of order/disorder. Each plot corresponds to the indicated couple of order parameters S and T. The following lines describe the Hill parameters: continuous line $\rightarrow m$, dotted line $\rightarrow p$, circles $\rightarrow n$, dashed line $\rightarrow k$ and dotted continuous line $\rightarrow l$. In all cases we have considered the same materials for inclusions and matrix and the same volume fraction of the inhomogeneities.

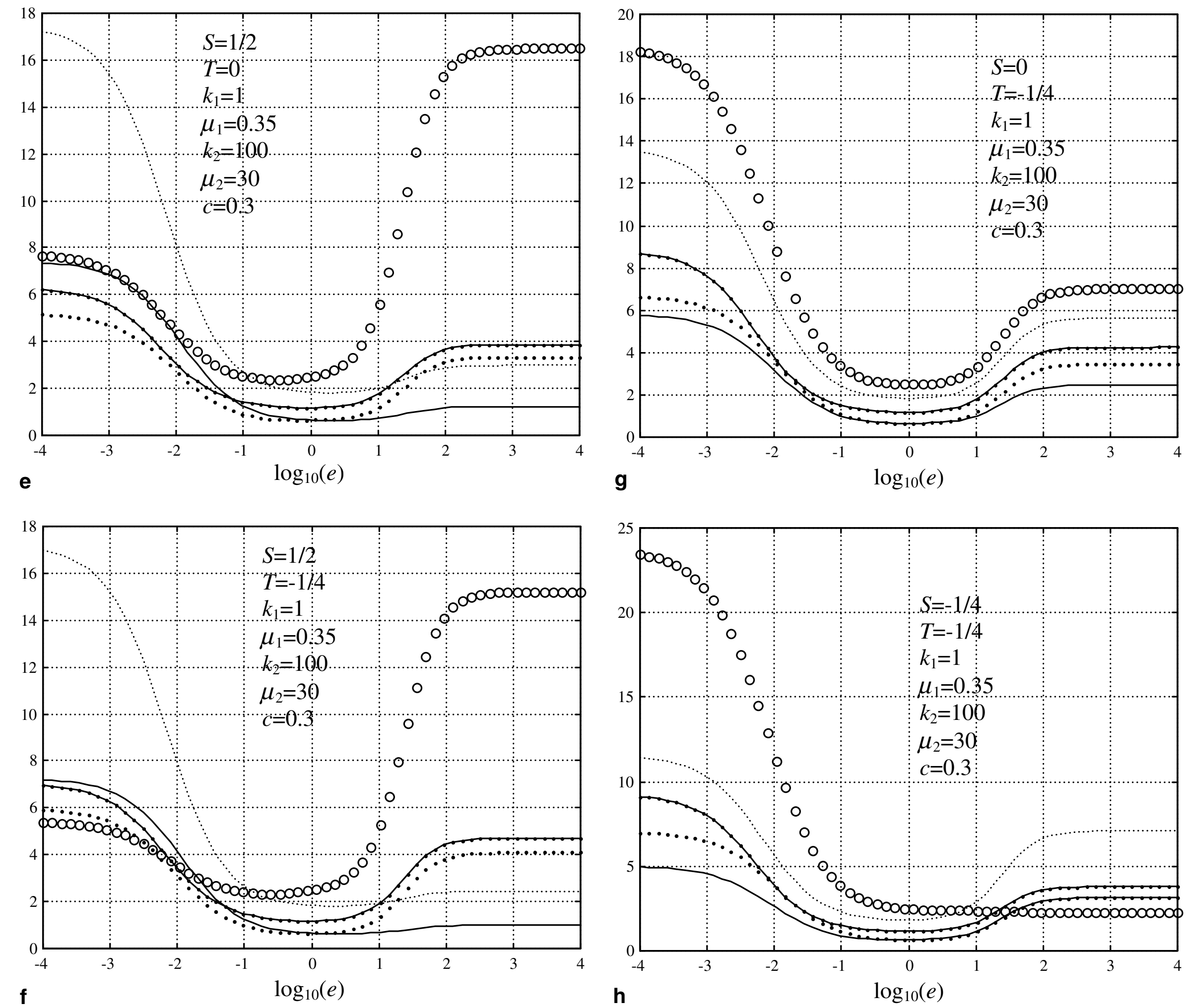
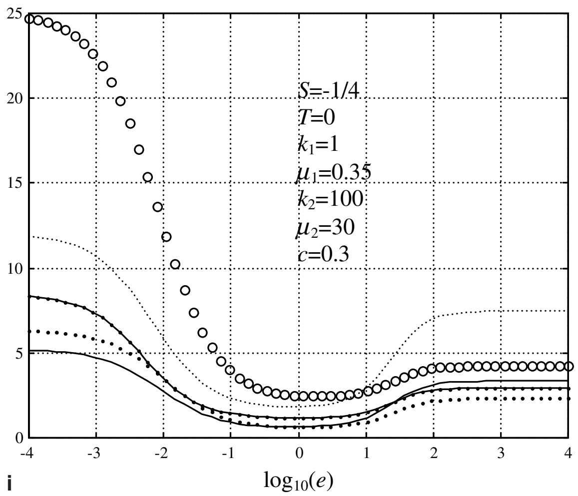


Fig. 7 (*continued*)

4. Conclusions

In this work we have analysed the effects of the orientational order/disorder of not spherical particles in composite or heterogeneous materials. As result of this analysis we have found the correct definition of some order parameters in such a way to predict the macroscopic electric and elastic properties as function of the state of microscopic order. In particular, for the electric characterisation we have found out new explicit relationships that allow us the computation of the permittivity tensor in terms of the shape of the embedded particles and the order parameter. On the other hand, from the point of view of the elastic properties of the overall medium, we have delineated and applied a complete procedure which takes into account any given orientational distribution of ellipsoids in the matrix. The theory can find many applications to real physical



i

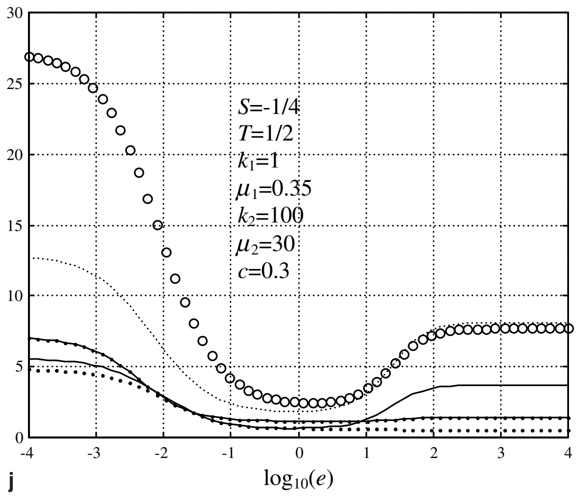


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Fig. 7 (*continued*)

situations ranging from technological aspects of composite materials to optical characterisation of nematic liquid crystals and to tissues modelling in biophysics: for example, the mechanical properties of bone, influenced by the trabecular architecture, can be investigated with these models.

Appendix A. Eshelby tensor for special cases

In this appendix we review the mathematical form of the Eshelby tensor [27] for special shapes of ellipsoids, which is used in the main text to describe the homogenisation procedure. We follow the conventions adopted in Mura [29]. It depends on the Poisson ratio $\nu = (3k_1 - \mu_1)/[2(3k_1 + \mu_1)]$ of the matrix.

For spheres we have:

$$\widehat{\mathbf{S}} = \begin{bmatrix} \frac{1}{15}\frac{7-5v}{1-v} & \frac{1}{15}\frac{5v-1}{1-v} & \frac{1}{15}\frac{5v-1}{1-v} & 0 & 0 & 0 \\ \frac{1}{15}\frac{5v-1}{1-v} & \frac{1}{15}\frac{7-5v}{1-v} & \frac{1}{15}\frac{5v-1}{1-v} & 0 & 0 & 0 \\ \frac{1}{15}\frac{5v-1}{1-v} & \frac{1}{15}\frac{5v-1}{1-v} & \frac{1}{15}\frac{7-5v}{1-v} & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{2}{15}\frac{4-5v}{1-v} & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{2}{15}\frac{4-5v}{1-v} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{2}{15}\frac{4-5v}{1-v} \end{bmatrix} \tag{A.1}$$

For cylinders:

$$\widehat{\mathbf{S}} = \begin{bmatrix} \frac{1}{8}\frac{5-4v}{1-v} & \frac{1}{8}\frac{4v-1}{1-v} & \frac{1}{2}\frac{v}{1-v} & 0 & 0 & 0 \\ \frac{1}{8}\frac{4v-1}{1-v} & \frac{1}{8}\frac{5-4v}{1-v} & \frac{1}{2}\frac{v}{1-v} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{4}\frac{3-4v}{1-v} & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{1}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{2} \end{bmatrix} \tag{A.2}$$

And finally, for planar inclusions:

$$\widehat{\mathbf{S}} = \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ \frac{v}{1-v} & \frac{v}{1-v} & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix} \tag{A.3}$$

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