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Shape-dependent effects of dielectrically nonlinear inclusions in heterogeneous media

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In this work the electrical response of a mixture composed of dielectrically nonlinear ellipsoids dispersed in a linear matrix is modeled. The inclusions may be randomly oriented. The aim is both to set up a methodology apt to deal with this kind of system and to use it to study the effect of marked nonsphericity of inclusions on the global behavior of a mixture. The results are quite interesting from both these points of view. The method here developed extends the Maxwell-Garnett theory [*A Treatise on Electricity and Magnetism* (Clarendon, Oxford, 1881)], which deals with dielectrically linear inclusions, and it allows, *inter alia*, to obtain a closed-form expression for the hypersusceptibility ratio of the mixture to the dispersed inclusions. These latter can range from cylinders to spheres, already present in the literature, to "penny-shaped" particles. The theoretical framework is based on the assumption that the dispersion is very dilute. We were able to show that in a specific case, when oblate particles such as elliptic lamellae are dispersed in a matrix having dielectric constant lower than the linear term of inclusion permittivity, a remarkable nonlinear effect occurs. This theory finds application in fields such as nonlinear optics and, more broadly, in many branches of material science. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2128689]

I. INTRODUCTION

In recent material science development, considerable attention has been devoted to electromagnetically nonlinear composite structures due to their applications, for instance, to integrated optical devices (such as optical switching and signal processing devices).[1–3] More specifically, intrinsic optical bistability has been extensively studied theoretically as well as experimentally with the help of mixture theory.[4,5] In all of these cases, a linear medium has been considered containing spherical inclusions randomly located, or at most spheroidal inclusions having fixed orientation.

Historically, the first studies characterizing mixtures, in terms of their constituting phase properties and the underlying microstructure, concerned linear inclusions immersed in linear matrices. In the current literature, Maxwell's relation for linear spheres[6,7] and Fricke's expressions for linear ellipsoids[8,9] form the so-called Maxwell-Garnett effective-medium theory:[10,11] both cases are derived under the hypothesis of very low concentration of the linear dispersed component. The so-called Bruggeman or differential technique can be then applied to generalize these results to the case of larger volume fractions.[12,13] A lot of work has also been devoted to describing the relationship between microstructure and subsequent macroscopic properties; for instance, Bianco and Parodi applied a functional unifying approach to capture the intrinsic mathematical properties of a general mixing formula.[14] A fundamental result is given by the Hashin-Shtrikman variational analysis,[15] which provides upper and lower bounds for composite materials, irrespective of their microstructure. Finally, the relation between the spatial correlation function of the dispersed component and the global properties of the material was derived via the Brown expansion.[16]

Recent progress in this field can be ascribed to Goncharenko *et al.*,[17] who dealt with dielectrically linear and nonlinear spheroidal inclusions of geometric factors probabilistically distributed. Then Lakhtakia and Mackay studied the size-dependent Bruggeman theory, which considers the effective particle dimension for nondilute dispersions.[18] Furthermore, a wide survey of mixture theory applications has been made by Mackay,[19] when he analyzed the peculiar properties exhibited by metamaterials. Important results concerning a dispersion of dielectrically nonlinear and graded parallel cylinders have been achieved by Wei and Wu.[20]

Our aim is to extend previous work and to explore the importance of inclusion shape in this context. This is achieved by analyzing the effect of either weak or strong nonsphericity of the particles on the nonlinear behavior of the whole composite material. To do this, we consider a dispersion of dielectrically nonlinear ellipsoidal particles, concisely stated as "nonlinear" throughout the paper, randomly oriented in a (dielectrically) linear matrix and we then develop a mathematical procedure to perform the needed averages of the electric quantities over all orientations of the inclusions.

This analysis leads to the nonlinear constitutive equation connecting the macroscopic electric displacement to the macroscopic electric field. It is worth pointing out that, as it

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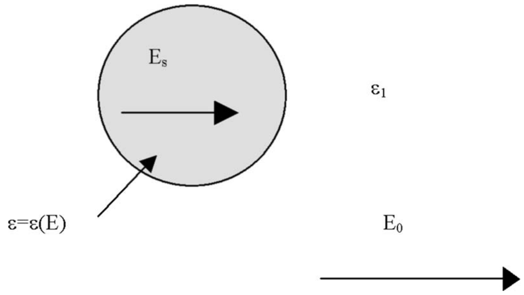

FIG. 1. Schematic of a dielectrically nonlinear sphere immersed in an external uniform field E_0: a uniform field E_s appears inside the particle.

is frequent in this field, the results of this work have been derived under electrostatic assumption, but they hold valid also in the low-frequency regime, as long as the wavelength is much larger than the largest dimension of the inclusion.

This work explores a wide range of inclusion shapes and presents a random orientation of the particles.

II. METHODOLOGICAL APPROACH

For the sake of clarity in exposition, we will describe how the approach tackles problems of increasing complexity.

A. Field perturbation due to one single nonlinear spherical inclusion in a uniform field

A nonlinear isotropic and homogenous sphere can be described from the electrical point of view by the constitutive equation

$$\mathbf{D} = \varepsilon(E)\mathbf{E}, \tag{1}$$

where $\mathbf{D}$ is the electric displacement inside the particle, $\mathbf{E}$ is the electric field, and the function ε depends only on the modulus E of $\mathbf{E}$. This latter property accounts for the fact that the medium inside the ellipsoid is isotropic and homogenous. Let us now place this inclusion in a linear matrix characterized by permittivity ε_1 (see Fig. 1) and let us calculate the field inside the spherical inclusion when a uniform external field $\mathbf{E}_0$ is applied to the system. If the particle were linear, in the dielectric sense, with permittivity ε_2, we would have, inside the sphere, a uniform Lorentz electric field E_s given by the well-known formula[21]

$$E_s = 3E_0\varepsilon_1/(2\varepsilon_1 + \varepsilon_2). \tag{2}$$

Conversely, if the sphere were electrically nonlinear, it is easy to prove that the internal field would satisfy the equation

$$E_s = 3E_0\varepsilon_1/[2\varepsilon_1 + \varepsilon(E_s)]. \tag{3}$$

This is true since the electric field E_s fulfilling Eq. (3) satisfies both Maxwell's laws and the boundary conditions at the inclusion surface as its linear counterpart (2) does when $\varepsilon_2 = \varepsilon(E_s)$. This very simple observation has, however, several interesting consequences in the field of nonlinear mixtures.

B. Field perturbation due to one single nonlinear ellipsoidal inclusion in a uniform field

Here we present a general solution to the problem of a nonlinear ellipsoidal particle embedded in a linear material. The theory is based on the following result derived for the linear case, which describes the behavior of one electrically linear ellipsoidal particle of permittivity ε_2 in a linear homogeneous medium of permittivity ε_1. Let the axes of the ellipsoid be l_x, l_y, and l_z (aligned with axes x, y, and z of the ellipsoid reference frame) and let a uniform electric field $\mathbf{E}_0=(E_{0x},E_{0y},E_{0z})$ be applied to the structure. Then, according to Stratton,[21] the electric field $\mathbf{E}_s=(E_{sx},E_{sy},E_{sz})$ inside the ellipsoid is uniform and it can be expressed as follows:

$$E_{si} = \frac{E_{0i}}{1 + L_i(\varepsilon_2/\varepsilon_1 - 1)}. \tag{4}$$

Here, and throughout the paper, the index i takes the x, y, and z values. The expressions for the depolarization factors L_i in the case of generally shaped ellipsoid can be found in the literature.[13] They can be expressed in terms of elliptic integrals under the assumption that $0<l_x<l_y<l_z$, and having defined eccentricities as follows: $0<e=l_x/l_y<1$ and $0<g=l_y/l_z<1$. The condition $L_x+L_y+L_z=1$ is always fulfilled.

Let us now generalize this result to the case where a dielectrically nonlinear ellipsoid is embedded in the linear matrix. The main result follows: the electric field inside the inclusion is uniform even in the nonlinear case and it may be calculated by means of the following system of equations:

$$E_{si} = \frac{E_{0i}}{1 + L_i[\varepsilon(E_s)/\varepsilon_1 - 1]}, \quad \forall\, i, \tag{5}$$

where, as before, $\mathbf{E}_0$ is a uniform electric field applied to the structure and $\mathbf{E}_s$, the unknown in the nonlinear system (5), is a uniform field as well. This property holds true due to the same reasons as in Sec. II A: if a solution of (5) exists, due to self-consistency, all the boundary conditions are fulfilled and the problem is completely analogous to its linear counterpart, treated by Stratton,[21] provided that $\varepsilon_2=\varepsilon(E_s)$.

C. Algorithm convergence and physical constraints

An interesting aspect related to the problem faced in this work shows up when one considers, for spherical inclusions, the nonlinear equation (3) and tries to solve it iteratively. This means that, in order to solve for E_s, one starts with a given initial value $E_s{}^0$, and one uses the successive approximations described by the iteration rule,

$$E_s{}^{n+1} = 3E_0\varepsilon_1/[2\varepsilon_1 + \varepsilon(E_s{}^n)]. \tag{6}$$

In what follows we provide a sufficient convergence criterion for a more general iterative scheme applicable to dielectrically nonlinear ellipsoids. It can be derived from the nonlinear system (5). The system is in the form $E_{si}=f_i(E_{sx},E_{sy},E_{sz})$ and the iteration rule takes the form

$$E_{si}{}^{n+1} = \frac{E_{0i}}{1 + L_i[\varepsilon(\|\mathbf{E}_s{}^n\|)/\varepsilon_1 - 1]}. \tag{7}$$

To investigate the behavior of this convergence process, we recall a well-known property holding for vector sequences: let $\mathbf{E}_s^*$ be a fixed point (or equilibrium point) for the sequence $\mathbf{E}_s^{n+1}=\mathbf{f}(\mathbf{E}_s^n)$ [so that $\mathbf{E}_s^*=\mathbf{f}(\mathbf{E}_s^*)$]; such a point is said to be locally asymptotically stable if the iterations lead to $\mathbf{E}_s^*$ for a suitable set of starting values $\mathbf{E}_s^0$. Then, the local asymptotic stability is guaranteed if the Jacobian matrix of $\mathbf{f}$, evaluated at $\mathbf{E}_s=\mathbf{E}_s^*$, has all the eigenvalues with an absolute value strictly less than 1.[22] We leave in Appendix A the details of the calculations that lead to the following sufficient convergence criterion: the iteration rule given by Eq. (7) is convergent to the exact internal electric field if the nonlinear material of the ellipsoid fulfills the condition $|(E/\varepsilon)(\partial\varepsilon/\partial E)|<1$.

It is quite intriguing to see how several physical properties correlate with this convergence condition; in this regard one can separate this condition in two different statements. The first one is $(E/\varepsilon)(\partial\varepsilon/\partial E)>-1$ that can be recast, by means of Eq. (1), to the form $\partial D/\partial E>0$. This relation expresses a general property of the permittivity function, always fulfilled in real materials.[23] The second condition is $(E/\varepsilon)(\partial\varepsilon/\partial E)<1$. This is surely satisfied whenever $\partial\varepsilon/\partial E<0$, therefore, for instance, in all materials where the Langevin model of dipole generation and orientation applies, such as water.[24] In a more general context, one can describe nonlinear dielectric materials by means of the so-called Kerr nonlinearity relation, often adopted in metamaterials study,[19]

$$\varepsilon(E)=\varepsilon_2+\alpha E^2, \tag{8}$$

which assumes that ε_2 and α are constant. The Kerr nonlinearity is termed *focusing* or *defocusing* according to the fact that $\alpha>0$ or $\alpha<0$, respectively.[25] It is straightforward to verify that the convergence condition $(E/\varepsilon)(\partial\varepsilon/\partial E)<1$ is always verified for defocusing Kerr nonlinearity and is verified only if $E_s^2<\varepsilon_2/\alpha$ (we remind the reader that here E_s is the modulus of the actual electric field inside the inclusion) in the case of focusing nonlinearity.

To test the iteration scheme, we considered a water ellipsoidal drop placed in a rigid linear homogeneous matrix not allowing for droplet deformation. A permittivity function that describes remarkably well the nonlinear water behavior and fulfills the above-mentioned conditions is the following:[26,27]

$$\varepsilon(E)=\left[\varepsilon(\infty)^2+\frac{\varepsilon(0)^2-\varepsilon(\infty)^2}{1+\gamma E^2}\right]^{1/2}, \tag{9}$$

where $\varepsilon(\infty)=6\varepsilon_0, \varepsilon(0)=80\varepsilon_0$, and $\gamma=6.49\times10^{-18}\ \mathrm{m^2/V^2}$ (ε_0 is the vacuum permittivity). These parameters fit well the water behavior at an absolute temperature of 298 °K and in a frequency range going from the static case to approximately 1 GHz.

In Fig. 2 one can find an example of iterative process applied to an ellipsoid, with depolarizing factors $L_x=0.1$, $L_y=0.3$, and $L_z=0.6$, that is immersed in a uniform electric field with components $E_{0x}=-5\times10^9$ V/m, $E_{0y}=5\times10^9$ V/m, and $E_{0z}=-3\times10^9$ V/m. The particle is described by the constitutive relation (9) and the linear matrix has permittivity $\varepsilon_1=35\varepsilon_0$. The first plot represents the convergence of the iterations described in Eq. (7), while the second one represents the behavior of the permittivity versus the iterations number. In all the tests we made, only few iterative steps were sufficient to reach good convergence.

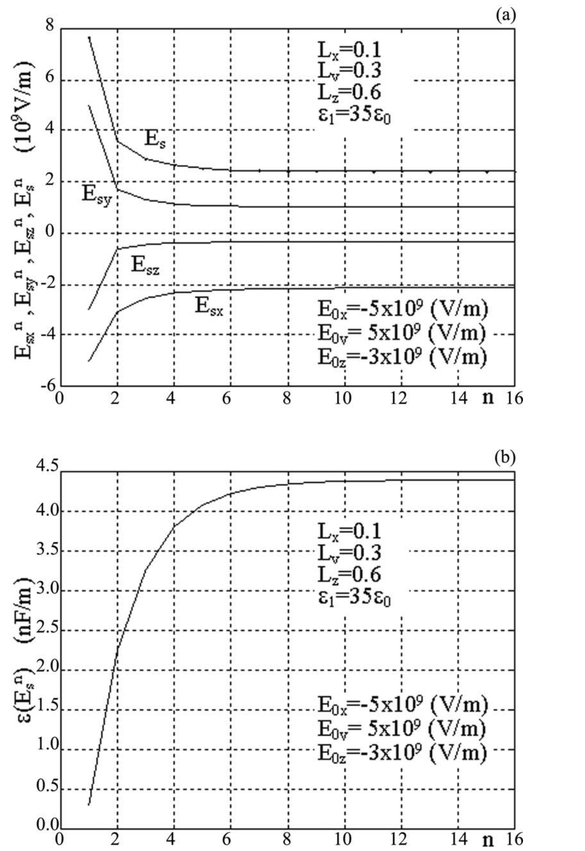

FIG. 2. Example of iterative process applied to an ellipsoid with specified depolarizing factors to the given uniform electric field. The particle is described by the nonlinear constitutive relation given in Eq. (9) (water) and it is embedded in a linear medium with permittivity ε_1. The first plot represents the convergence of the iterations described in Eq. (7), while the second one represents the behavior of the permittivity vs the iteration number.

D. Maxwell-Garnett mixtures of nonlinear ellipsoids

The aim of this subsection is to extend the results, holding for a single inclusion, to a mixture of randomly oriented nonlinear ellipsoids in a linear homogeneous matrix (see Fig. 3).

The permittivity of the inclusions is described by the isotropic nonlinear relation (8) and the linear matrix has permittivity ε_1; the overall permittivity function of the dispersion is expected to be isotropic because of the random orientation of the particles and therefore it can be expanded in a series with respect to the field modulus $\varepsilon(E)=\varepsilon_{\mathrm{eq}}+\beta E^2+\delta E^4+\cdots$, where the coefficients $\varepsilon_{\mathrm{eq}}$ (the subscript "eq" points out the equivalent character of the term), β, and δ depend on various parameters of the mixture such as the eccentricities of the ellipsoids, the volume fraction c of the included phase, and the permittivities ε_1, ε_2, and α. The homogenization procedure should provide the coefficients $\varepsilon_{\mathrm{eq}}$, β, and δ in terms of the mentioned parameters. In the technical literature, the coefficients α and β of the first nonlinear

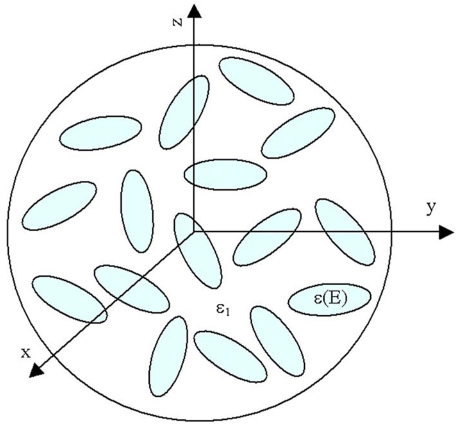

FIG. 3. (Color online) Structure of a dispersion of randomly oriented ellipsoids. We used a spherical boundary just as an example of a closed surface containing the mixture. This refers to the volume V defined in Sec. II D 2.

term of the expanded constitutive equations for inclusions and mixture, respectively, are often called hypersusceptibilities.[23]

The main achievement of this work is the derivation of a closed-form expression for the hypersusceptibility ratio β/α. This quantity is of interest inasmuch as it represents the amplification of the composite material nonlinear behavior with respect to that of the inclusions. The expression is derived under the assumption of a Kerr-like constitutive equation of the composite medium, is of the form $\varepsilon(E)=\varepsilon_{eq}+\beta E^2$, which neglects higher-order terms. All the computations are carried out under the same hypothesis underlying the linear Maxwell-Garnett theory,[11] that is, low concentration c of the dispersed phase.

1. Average electric field inside a single randomly oriented inclusion

To begin the analysis, we substitute Eq. (8), holding for a single ellipsoid, in Eq. (5),

$$E_{si}=\frac{\varepsilon_1 E_{0i}}{\varepsilon_1+L_i[\varepsilon_2-\varepsilon_1+\alpha(E_{sx}{}^2+E_{sy}{}^2+E_{sz}{}^2)]}. \tag{10}$$

This is an algebraic system of degree nine with three unknowns, namely, E_{sx}, E_{sy}, and E_{sz}. It might be hard, if not impossible, to be solved analytically, but we are interested, for our purposes, in just the first terms of a series expansion for the solution. To obtain it, we may symbolically use the iteration scheme defined in Eq. (7) or we may adopt the ansatz $E_{si}=\lambda_i E_{0i}+\mu_i E_{0i}{}^3$ and solve for λ_i and μ_i. For the sake of brevity, we omit here the simple but long calculation, which leads to the solution

$$E_{si}=\frac{\varepsilon_1 E_{0i}}{(1-L_i)\varepsilon_1+L_i\varepsilon_2}-\frac{\alpha\varepsilon_1{}^3 L_i E_{0i}}{[(1-L_i)\varepsilon_1+L_i\varepsilon_2]^2}\times\sum_j\frac{E_{0j}{}^2}{[(1-L_j)\varepsilon_1+L_j\varepsilon_2]^2}+O(\|\mathbf{E}_0\|^4). \tag{11}$$

We observe that the first term represents the classical Lorentz field appearing in a dielectrically linear ellipsoidal inclusion. The second term is the first nonlinear contribution, which is directly proportional to the inclusion hypersusceptibility α. To simplify the expressions, from now on, we will use the notation $a_i=(1-L_i)\varepsilon_1+L_i\varepsilon_2$.

To derive the mixture behavior, we need to calculate the electric field of a single nonlinear ellipsoidal inclusion arbitrarily oriented in space and embedded in a homogeneous medium with permittivity ε_1. In order to do this, we shall express Eq. (11) in the global framework of reference of the mixture. We define three unit vectors, indicating the principal directions of each ellipsoid in space: they are referred to as $\hat{n}_x$, $\hat{n}_y$, and $\hat{n}_z$, and they correspond to the axes l_x, l_y, and l_z of the ellipsoid. By using Eq. (11), we may compute the electric field, induced by a given external arbitrary uniform electric field, inside the inclusion (from now on we will omit the additional higher-order terms),

$$\mathbf{E}_s=\sum_i\left[\frac{\varepsilon_1\mathbf{E}_0\cdot\hat{n}_i}{a_i}-\frac{\alpha\varepsilon_1{}^3 L_i\mathbf{E}_0\cdot\hat{n}_i}{a_i^2}\sum_j\frac{(\mathbf{E}_0\cdot\hat{n}_j)^2}{a_j^2}\right]\hat{n}_i. \tag{12}$$

We shall now average it over all the possible orientations of the particle. The averaging method used is quite interesting and of general applicability but it is also a bit cumbersome, so we leave it in Appendix B. The result of the process is

$$\langle\mathbf{E}_s\rangle=\mathbf{E}_0(\varepsilon_1 M-\alpha\varepsilon_1{}^3 E_0{}^2 N), \tag{13}$$

where M and N depend on the linear term of the permittivities and on the geometry of the inclusions and are defined in Appendix B, Eq. (B9). We note that the average field inside the particle is aligned with the external field and thus the average behavior of the inclusion is isotropic; in contrast, from Eq. (12) it follows that the electric field inside the ellipsoid is not aligned with the external one when a given orientation is kept fixed for the particle itself.

2. Averaging process in a dilute mixture

If we now consider a mixture with a volume fraction $c \ll 1$ of randomly oriented, dielectrically nonlinear, ellipsoids embedded in a homogeneous matrix with permittivity ε_1, we can evaluate a different kind of average, the one of the electric field over all of the space occupied by the mixture. It can be done via the following relationship:

$$\langle\mathbf{E}\rangle=c\langle\mathbf{E}_s\rangle+(1-c)\mathbf{E}_0. \tag{14}$$

This means that we do not take into account the interactions among the inclusions because of the very low concentration: each ellipsoid behaves as an isolated one. Once more, to derive Eq. (14), we assume an approximately uniform electric field $\mathbf{E}_0$ in the space outside the inclusions.

To evaluate the equivalent constitutive equation, we compute the average value of the displacement vector inside the random material. V is defined as the total volume occupied by the mixture, V_e as the region occupied by the inclusions, and V_o as the remaining space (so that $V=V_e\cup V_o$). The average value of $\mathbf{D}(\mathbf{r})=\varepsilon\mathbf{E}(\mathbf{r})$ is evaluated as follows ($\mathbf{D}$ and $\mathbf{E}$ represent the local fields, and $\langle\mathbf{D}\rangle$ and $\langle\mathbf{E}\rangle$ their macroscopic counterparts):

$$
\begin{aligned}
\langle \mathbf{D} \rangle &= \frac{1}{|V|}\int_V \varepsilon \mathbf{E}(\mathbf{r}) d\mathbf{r} \\
&= \frac{1}{|V|}\varepsilon_1 \int_{V_o} \mathbf{E}(\mathbf{r}) d\mathbf{r} + \frac{1}{|V|}\int_{V_e} \varepsilon \mathbf{E}(\mathbf{r}) d\mathbf{r} \\
&= \frac{1}{|V|}\varepsilon_1 \int_{V_o} \mathbf{E}(\mathbf{r}) d\mathbf{r} + \frac{1}{|V|}\varepsilon_1 \int_{V_e} \mathbf{E}(\mathbf{r}) d\mathbf{r} \\
&\quad + \frac{1}{|V|}\int_{V_e} (\varepsilon - \varepsilon_1)\mathbf{E}(\mathbf{r}) d\mathbf{r} = \varepsilon_1 \langle \mathbf{E} \rangle + c\langle [\varepsilon(E_s) \\
&\quad - \varepsilon_1]\mathbf{E}_s \rangle.
\end{aligned}
\tag{15}
$$

It can be noted that the average value given by $\langle[\varepsilon(\|\mathbf{E}_s\|) - \varepsilon_1]\mathbf{E}_s\rangle$ is not available from the previous computations and it must be calculated *ex novo*, this is done in Appendix C, leading to the following result:

$$
\langle \mathbf{D} \rangle = \varepsilon_1 \langle \mathbf{E} \rangle + c\varepsilon_1(\varepsilon_2 - \varepsilon_1) M \mathbf{E}_0 + c\alpha\varepsilon_1^{\,4} P E_0^{\,2} \mathbf{E}_0, \tag{16}
$$

where P is defined in Appendix C, Eq. (C6). From Eqs. (13), (14), and (16) it follows that all the averaged vectorial quantities are aligned with $\mathbf{E}_0$, therefore, we can continue our computations with scalar quantities; moreover, from now on, we will leave out the average symbols $\langle\cdot\rangle$.

Equations (13), (14), and (16) may then be rewritten as

$$
\begin{aligned}
&E_s = \varepsilon_1 M E_0 - \alpha\varepsilon_1^{\,3} E_0^{\,3} N, \\
&E = cE_s + (1-c)E_0, \\
&D = \varepsilon_1 E + c\varepsilon_1(\varepsilon_2 - \varepsilon_1) M E_0 + c\alpha\varepsilon_1^{\,4} P E_0^{\,3}.
\end{aligned}
\tag{17}
$$

These are the main equations describing the overall mixture behavior. We shall recall the approximations introduced to obtain them: the second equation, dealing with the average value of the electric field, has been deduced under the hypothesis of low concentration. The first and third equations are exact from the volume fraction point of view, but they are approximated from the nonlinearity point of view: in fact, they account just for the first nonlinear terms. By solving system (17), we search for a relation between D and E characterizing the nonlinear mixture. By eliminating E_s from the first two relationships, we obtain

$$
E = (1 - c + c\varepsilon_1 M) E_0 - c\alpha\varepsilon_1^{\,3} N E_0^{\,3}. \tag{18}
$$

We now need to solve the previous equation with respect to E_0: for our purposes it is sufficient to obtain a series solution with two terms and thus we let $E_0 = \lambda E + \mu E^3$, we substitute it in Eq. (18), and we solve for the unknown coefficients; the result is

$$
E_0 = \frac{E}{(1 - c + c\varepsilon_1 M)} + \frac{c\alpha\varepsilon_1^{\,3} N E^3}{(1 - c + c\varepsilon_1 M)^4}. \tag{19}
$$

The final result is obtained by substituting Eq. (19) in the third equation of system (17) and neglecting the powers of E greater than 3,

$$
D = \varepsilon_1 E + c\varepsilon_1(\varepsilon_2 - \varepsilon_1) M \left[\frac{E}{(1 - c + c\varepsilon_1 M)} + \frac{c\alpha\varepsilon_1^{\,3} N E^3}{(1 - c + c\varepsilon_1 M)^4} \right] + c\alpha\varepsilon_1^{\,4} P \frac{E^3}{(1 - c + c\varepsilon_1 M)^3}. \tag{20}
$$

III. RESULTS

Our technique was applied to a dispersion of ellipsoids described by Eq. (8), with volume fraction c and embedded in a homogeneous matrix with permittivity ε_1; this has led to the nonlinear constitutive Eq. (20) for the composite medium in the form $\mathbf{D} = \varepsilon(E)\mathbf{E} = (\varepsilon_{eq} + \beta E^2)\mathbf{E}$, where

$$
\varepsilon_{eq} = \varepsilon_1 + \frac{c\varepsilon_1(\varepsilon_2 - \varepsilon_1) M}{(1 - c + c\varepsilon_1 M)} = \varepsilon_1 \frac{1 - c + c\varepsilon_2 M}{1 - c + c\varepsilon_1 M}, \tag{21}
$$

$$
\frac{\beta}{\alpha} = c\varepsilon_1^{\,4} \frac{P + c[(\varepsilon_2 - \varepsilon_1) M N + P(\varepsilon_1 M - 1)]}{(1 - c + c\varepsilon_1 M)^4}. \tag{22}
$$

The already mentioned quantities M, N, and P, defined in Appendixes B and C, depend only on geometrical factors (ellipsoid eccentricities) and on the linear terms of the permittivities.

Equation (21), giving the linear approximation for the permittivity, coincides with the Maxwell-Garnett formula for a dispersion of ellipsoids.[13] Equation (22) represents the mixture to inclusion hypersusceptibility ratio. The first-order expansion with respect to the volume fraction [from Eq. (20)] is

$$
\varepsilon(E) = \varepsilon_1 + [\varepsilon_1(\varepsilon_2 - \varepsilon_1) M + \alpha\varepsilon_1^{\,4} P E^2] c. \tag{23}
$$

The methodology was applied to examine the actual effects of particle nonsphericity on mixtures. In Figs. 4 and 5 we show plots of the properties of the overall mixture as a function of the eccentricities of the ellipsoids composing the mixture itself. In Fig. 4 one can see the plots of the quantities $\varepsilon_{eq}/\varepsilon_1$ and β/α versus the eccentricities, which define the shape of the particles, derived from Eqs. (21) and (22) with $\varepsilon_1 = 1$, $\varepsilon_2 = 10$, and $c = 1/5$. In Fig. 5 the same plots are derived with the following parameter values: $\varepsilon_1 = 10$, $\varepsilon_2 = 1$, and $c = 1/5$. In both cases we may observe that the amplification of the hypersusceptibility assumes the greatest values when dealing with planar nonlinear particles (elliptic lamellae). More surprisingly, hypersusceptibility ratio tends to assume its highest values (greater than 50) when $\varepsilon_1 > \varepsilon_2$.

In order to better capture the behavior of the system, the analysis of the simplified expression (23) can be quite useful, indeed. For instance, one can see that mixture electric behavior versus inclusion shape is mostly based on the parameters M and P. More precisely, the dependence of ε_{eq} on the inclusion shape acts through the M parameter, while the dependence of the ratio β/α on the inclusion shape acts through the P parameter.

Thus, peculiarities exhibited in correspondence of a given pair of eccentricities must follow the complex mathematical structure of P and M, which, in turn, depend only on geometrical factors and the linear permittivities ε_1 and ε_2. In particular, we observe that the difference $\varepsilon_{eq} - \varepsilon_1$ is pro-

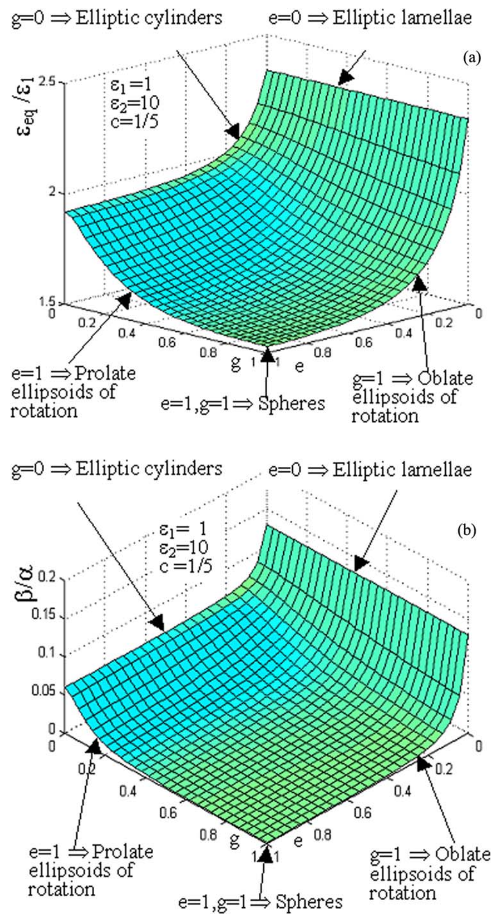

FIG. 4. (Color online) Plots of the surfaces $\varepsilon_{eq}/\varepsilon_1$, and β/α [Eqs. (21) and (22)] vs the eccentricities which define the shape of the particles with $\varepsilon_1=1$, $\varepsilon_2=10$, and $c=1/5$.

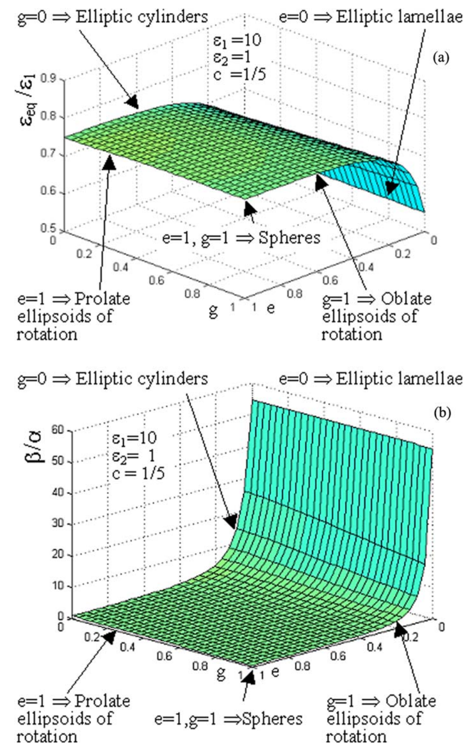

FIG. 5. (Color online) As for Fig. 4 but with $\varepsilon_1=10$, $\varepsilon_2=1$, and $c=1/5$. The amplification of the hypersusceptibility assumes the greatest values when $\varepsilon_1>\varepsilon_2$ and we are dealing with elliptic lamellae.

portional to $\varepsilon_2-\varepsilon_1$: this fact explains the different behaviors shown by ε_{eq} in Figs. 4(a) and 5(a). In agreement with this, in Fig. 4(a), where ε_2 is greater than ε_1, the penny-shaped particles lead to the highest value for ε_{eq}; consistently, in Fig. 4(b), where ε_2 is less than ε_1, the penny-shaped particles exhibit the lowest equivalent, e.g., effective, linear permittivity.

In a similar manner, we may observe that the ratio β/α is proportional to the fourth power of ε_1: this explains why the β/α behavior is so loosely dependent on $\varepsilon_2/\varepsilon_1$ [see Figs. 4(b) and 5(b)]; nonetheless, the amplification is very sensitive to large values of ε_1, as it is shown in Fig. 5(b).

Using Eqs. (21) and (22) we derived the electrostatic behavior in the following specific and meaningful cases.

A. Dispersion of spheres

If we use the depolarization factors for spherical objects ($L_x=L_y=L_z=1/3$) we obtain the simplified expression

$$\varepsilon(E)=\overbrace{\varepsilon_1\frac{2\varepsilon_1+\varepsilon_2-2c(\varepsilon_1-\varepsilon_2)}{2\varepsilon_1+\varepsilon_2+c(\varepsilon_1-\varepsilon_2)}}^{\varepsilon_{eq}}+\overbrace{\frac{81c\varepsilon_1^{\,4}\alpha E^2}{[2\varepsilon_1+\varepsilon_2+c(\varepsilon_1-\varepsilon_2)]^4}}^{\beta E^2}, \tag{24}$$

which corresponds to the following hypersusceptibility ratio, already derived by Yu *et al.*:[1]

$$\frac{\beta}{\alpha}=\frac{81c\varepsilon_1^{\,4}}{[2\varepsilon_1+\varepsilon_2+c(\varepsilon_1-\varepsilon_2)]^4}=c\left(\frac{\varepsilon_{eq}+2\varepsilon_1}{\varepsilon_2+2\varepsilon_1}\right)^4. \tag{25}$$

It is interesting to observe that Eqs. (24) and (25) are also true for $c=1$; in this case (very high volume fraction of inclusions) the procedure is not expected to be valid but nonetheless the result appears to be exact, at least for spheres. It follows that the first-order expansion of Eq. (21) for low values of the volume fraction, c, is

$$\varepsilon(E)=\varepsilon_1-3c\varepsilon_1\frac{\varepsilon_1-\varepsilon_2}{2\varepsilon_1+\varepsilon_2}+\frac{81c\varepsilon_1^{\,4}\alpha E^2}{(2\varepsilon_1+\varepsilon_2)^4}. \tag{26}$$

When dealing with spherical inclusions, the issue of particle orientation does not occur; we then have derived the following more accurate expression, via a long but straightforward calculation here omitted for the sake of brevity:

$$\begin{aligned}\varepsilon_{eq}(E)=\varepsilon_1&\frac{2\varepsilon_1+\varepsilon_2-2c(\varepsilon_1-\varepsilon_2)}{2\varepsilon_1+\varepsilon_2+c(\varepsilon_1-\varepsilon_2)}\\&+\frac{81c\varepsilon_1^{\,4}\alpha E^2}{[2\varepsilon_1+\varepsilon_2+c(\varepsilon_1-\varepsilon_2)]^4}\\&-\frac{2187c(1-c)\varepsilon_1^{\,6}\alpha^2E^4}{[2\varepsilon_1+\varepsilon_2+c(\varepsilon_1-\varepsilon_2)]^7}+\cdots.\end{aligned} \tag{27}$$

The first two right-hand-side terms are in perfect agreement with those appearing in Eq. (24). It is interesting to notice that the third term, a nonlinear perturbation of degree four, does not affect the exact results for $c=0$ nor $c=1$.

B. Dispersion of cylinders

We adopt the depolarization factors for strongly prolate particles ($L_x=L_y=1/2$, $L_z=0$) and we obtain the expression

$$\varepsilon(E)=\frac{\varepsilon_1[3(\varepsilon_1+\varepsilon_2)-3c(\varepsilon_1-\varepsilon_2)]-c\varepsilon_2(\varepsilon_1-\varepsilon_2)}{3(\varepsilon_1+\varepsilon_2)+2c(\varepsilon_1-\varepsilon_2)}+\frac{9}{5}c\alpha E^2 \times\frac{9\varepsilon_2^4+36\varepsilon_2^3\varepsilon_1+102\varepsilon_2^2\varepsilon_1^2+132\varepsilon_2\varepsilon_1^3+441\varepsilon_1^4}{[3(\varepsilon_1+\varepsilon_2)+2c(\varepsilon_1-\varepsilon_2)]^4} -\frac{18}{5}c^2\alpha E^2\frac{(3\varepsilon_2^2+10\varepsilon_1\varepsilon_2+23\varepsilon_1^2)(\varepsilon_2-\varepsilon_1)^2}{[3(\varepsilon_1+\varepsilon_2)+2c(\varepsilon_1-\varepsilon_2)]^4}. \quad (28)$$

The corresponding expansion to first order in the volume fraction c is

$$\varepsilon(E)=\varepsilon_1-c\frac{(5\varepsilon_1+\varepsilon_2)(\varepsilon_1-\varepsilon_2)}{3(\varepsilon_1+\varepsilon_2)}+\frac{1}{45}c\alpha E^2 \times\frac{9\varepsilon_2^4+36\varepsilon_2^3\varepsilon_1+102\varepsilon_2^2\varepsilon_1^2+132\varepsilon_2\varepsilon_1^3+441\varepsilon_1^4}{(\varepsilon_1+\varepsilon_2)^4}. \quad (29)$$

C. Dispersion of strongly oblate (planar) particles

We consider the depolarization factors for "penny-shaped" or strongly oblate particles ($L_x=1$, $L_y=L_z=0$) and we obtain the expression

$$\varepsilon(E)=\varepsilon_2\frac{3\varepsilon_1-2c(\varepsilon_1-\varepsilon_2)}{3\varepsilon_2+c(\varepsilon_1-\varepsilon_2)}+\frac{9}{5}c\alpha E^2\frac{9\varepsilon_1^4+12\varepsilon_2^2\varepsilon_1^2+24\varepsilon_2^4-2c(4\varepsilon_2^2+2\varepsilon_1\varepsilon_2+3\varepsilon_1^2)(\varepsilon_2-\varepsilon_1)^2}{[3\varepsilon_2+c(\varepsilon_1-\varepsilon_2)]^4}. \quad (30)$$

The corresponding expansion to first order in c is

$$\varepsilon(E)=\varepsilon_1-c\frac{(\varepsilon_1+2\varepsilon_2)(\varepsilon_1-\varepsilon_2)}{3\varepsilon_2}+\frac{1}{45}c\alpha E^2\frac{9\varepsilon_1^4+12\varepsilon_2^2\varepsilon_1^2+24\varepsilon_2^4}{\varepsilon_2^4}. \quad (31)$$

D. Nonlinear ellipsoids having linear permittivity term coincident with matrix dielectric constant

When $\varepsilon_1=\varepsilon_2$, we have $M=1/\varepsilon_1$, $P=1/\varepsilon_1^4$, and $N=1/(3\varepsilon_1^4)$ and we obtain from Eqs. (21) and (22) the following very simple relation:

$$\varepsilon(E)=\varepsilon_1+c\alpha E^2. \quad (32)$$

In this particular case the relation provides exact results also for $c=1$.

IV. CONCLUSIONS AND COMMENTS

In this work we developed the theoretical machinery which enables us to infer the electrical behavior of a mixture containing dielectrically nonlinear ellipsoidal inclusions, provided that the dielectric constant of the linear matrix, the concentration, and the eccentricities of the ellipsoidal inclusions are known. Our results are based on the assumption of randomly oriented inclusions. The results for specific cases of spheres, cylinders, and lamellae have been derived and placed in the context of the present literature. Besides, the broader applicability of the method gave us a more complete view over the influence that inclusion shape has on the global behavior. In particular, hypersusceptibility amplification was taken as a measure of this effect.

Our present activity is focused on extending the methodology to deal with higher-concentration values for the inclusions.

APPENDIX A: SUFFICIENT CONDITION FOR THE CONVERGENCE OF THE ITERATIVE PROCESS

We want to study the convergence behavior of system (7). We start by computing the derivatives $\partial f_i/\partial E_{sj}$, which appear in the Jacobian matrix; a straightforward calculation provides

$$\frac{\partial f_i}{\partial E_{sj}}=-\frac{\varepsilon_1 E_{sj}E_{0i}L_i}{E\{\varepsilon_1+L_i[\varepsilon(E)-\varepsilon_1]\}^2}\frac{\partial\varepsilon}{\partial E}, \quad (A1)$$

where, for the sake of brevity, we have used the notation $E=\|\mathbf{E}_s\|$.

All the derivatives, will, from now on, be evaluated at $\mathbf{E}_s=\mathbf{E}_s^*$ and this fact will be indicated with the symbol *. Therefore, recalling Eq. (5), we may write

$$J_{ij}^*=\left.\frac{\partial f_i}{\partial E_{sj}}\right|^*=-\frac{L_i}{\varepsilon_1}\frac{1}{E}\frac{\partial\varepsilon}{\partial E}E_{sj}\frac{E_{si}^2}{E_{0i}}. \quad (A2)$$

The Jacobian J may be written in an explicit form as follows:

$$J^*=\left.\frac{\partial f_i}{\partial E_{sj}}\right|^* =-\frac{1}{\varepsilon_1}\frac{1}{E}\frac{\partial\varepsilon}{\partial E}\begin{bmatrix} L_1\frac{E_{s1}^3}{E_{01}} & L_1E_{s2}\frac{E_{s1}^2}{E_{01}} & L_1E_{s3}\frac{E_{s1}^2}{E_{01}} \\ L_2E_{s1}\frac{E_{s2}^2}{E_{02}} & L_2\frac{E_{s2}^3}{E_{02}} & L_2E_{s3}\frac{E_{s2}^2}{E_{02}} \\ L_3E_{s1}\frac{E_{s3}^2}{E_{03}} & L_3E_{s2}\frac{E_{s3}^2}{E_{03}} & L_3\frac{E_{s3}^3}{E_{03}} \end{bmatrix}. \quad (A3)$$

It is easy to observe that the second and the third row in the Jacobian matrix are simply proportional to the first one. This implies that the rank of the matrix is one. Therefore, two eigenvalues assume the value zero, which is compatible with the convergence condition, while the third one is not yet known. This latter can be derived by subtraction from the trace of the matrix, remembering that the trace is the sum of the eigenvalues irrespective of the basis in which the matrix is expressed.[28] Thus, the sole eigenvalue different from zero has the absolute value given by the expression

$$|\lambda| = |\mathrm{tr}(J^*)| = \left| \frac{1}{\varepsilon_1}\frac{1}{E}\frac{\partial\varepsilon}{\partial E}\left(L_1\frac{E_{s1}^{\ 3}}{E_{01}} + L_2\frac{E_{s2}^{\ 3}}{E_{02}} + L_3\frac{E_{s3}^{\ 3}}{E_{03}} \right) \right|. \tag{A4}$$

The convergence or stability condition is given by $|\lambda|<1$ and we search for sufficient conditions to assure this statement. From Eq. (5) we derive the expression

$$\varepsilon(E) = \frac{\varepsilon_1 E_{0i} - (1-L_i)\varepsilon_1 E_{si}}{L_i E_{si}}. \tag{A5}$$

We may multiply and divide Eq. (A4) by $\varepsilon(E)$ obtaining, with the help of Eq. (A5),

$$\begin{aligned} |\lambda| &= \left| \frac{1}{\varepsilon_1}\frac{1}{E}\frac{\partial\varepsilon}{\partial E}\frac{1}{\varepsilon}\left(\sum_i \varepsilon L_i \frac{E_{si}^{\ 3}}{E_{0i}} \right) \right| \\ &= \left| \frac{1}{E}\frac{\partial\varepsilon}{\partial E}\frac{1}{\varepsilon}\left(\sum_i \frac{E_{0i}-(1-L_i)E_{si}}{E_{0i}} E_{si}^{\ 2} \right) \right| \\ &\leqslant \left| \frac{1}{E}\frac{\partial\varepsilon}{\partial E}\frac{1}{\varepsilon} \right| \sum_i \left| \frac{E_{0i}-(1-L_i)E_{si}}{E_{0i}} \right| E_{si}^{\ 2}. \end{aligned} \tag{A6}$$

Once again, from Eq. (5) we have $E_{si}=\varepsilon_1 E_{0i}/[(1-L_i)\varepsilon_1 + L_i\varepsilon(E)]$ where $1-L_i>0$ and $L_i>0$; it follows that E_{si} and E_{0i} have the same sign and we may write

$$0 \leqslant \frac{E_{si}}{E_{0i}} = \frac{\varepsilon_1}{(1-L_i)\varepsilon_1 + L_i\varepsilon(E)} \leqslant \frac{\varepsilon_1}{(1-L_i)\varepsilon_1} = \frac{1}{1-L_i}, \tag{A7}$$

from which we derive the property $(1-L_i)E_{si}/E_{0i}\leqslant 1$ or $1-(1-L_i)E_{si}/E_{0i}\geqslant 0$. Then, the absolute value appearing in the last sum of Eq. (A6) can be evaluated as follows:

$$\begin{aligned} \left| \frac{E_{0i}-(1-L_i)E_{si}}{E_{0i}} \right| &= \frac{E_{0i}-(1-L_i)E_{si}}{E_{0i}} \\ &= \frac{|E_{0i}|-(1-L_i)|E_{si}|}{|E_{0i}|} \leqslant 1. \end{aligned} \tag{A8}$$

Summarizing, the convergence condition simplifies as follows:

$$\begin{aligned} |\lambda| &\leqslant \left| \frac{1}{E}\frac{\partial\varepsilon}{\partial E}\frac{1}{\varepsilon} \right| \sum_i \left| \frac{E_{0i}-(1-L_i)E_{si}}{E_{0i}} \right| E_{si}^{\ 2} \\ &\leqslant \left| \frac{1}{E}\frac{\partial\varepsilon}{\partial E}\frac{1}{\varepsilon} \right| \sum_i E_{si}^{\ 2} = \left| \frac{E}{\varepsilon}\frac{\partial\varepsilon}{\partial E} \right| < 1. \end{aligned} \tag{A9}$$

APPENDIX B: ELECTRIC FIELD AVERAGING OVER ALL ORIENTATIONS

The expression for the internal electric field in (12) can be rewritten component by component, as follows:

$$E_{sk} = \sum_i \left[\frac{\varepsilon_1 E_{0l} n_{il}}{a_i} - \frac{\alpha\varepsilon_1^{\ 3} L_i E_{0l} n_{il}}{a_i^2} \sum_j \frac{E_{0q} n_{jq} E_{0p} n_{jp}}{a_j^2} \right] n_{ik}, \tag{B1}$$

where n_{jk} is the kth component of the unit vector $\hat{n}_j$, $(j=x,y,z)$ and we have considered the implicit sums of l, q, and p over 1, 2, and 3. For the following derivation, we are interested in the average value of the electric field $\mathbf{E}_s$ over all the possible orientations of the ellipsoid itself and then we have to compute the following:

$$\begin{aligned} \langle E_{sk}\rangle = \sum_i \Bigg[& \frac{\varepsilon_1 E_{0l}\langle n_{il} n_{ik}\rangle}{a_i} \\ & - \sum_j \frac{\alpha\varepsilon_1^{\ 3} L_i E_{0l} E_{0q} E_{0p} \langle n_{ik} n_{il} n_{jq} n_{jp}\rangle}{a_i^2 a_j^2} \Bigg]. \end{aligned} \tag{B2}$$

We are interested in the average values of the quantities $n_{il}n_{ik}$ and $n_{ik}n_{il}n_{jq}n_{jp}$. Performing the integration over the unit sphere (by means of spherical coordinates) we obtain, after some straightforward computations, the first result

$$\langle n_{il} n_{ik}\rangle = \frac{1}{3}\delta_{lk} \quad (i=x,y,\text{ and } z,\text{ not summed}). \tag{B3}$$

The determination of the average value of $n_{ik}n_{il}n_{jq}n_{jp}$ is a more complicated task. If $i=j$ we are dealing with a single unit vector, say, $\hat{n}=(n_1,n_2,n_3)$, and a long but straightforward integration leads to the result

$$\langle n_k n_l n_q n_p\rangle = \frac{1}{15}(\delta_{kl}\delta_{pq} + \delta_{pk}\delta_{ql} + \delta_{pl}\delta_{qk}). \tag{B4}$$

If $i\neq j$ we are dealing with two orthogonal unit vectors, say, $\hat{n}$ and $\hat{m}=(m_1,m_2,m_3)$; as before the integration provides

$$\langle m_k m_l n_q n_p\rangle = \frac{2}{15}\delta_{kl}\delta_{pq} - \frac{1}{30}(\delta_{pk}\delta_{ql} + \delta_{pl}\delta_{qk}). \tag{B5}$$

The results shown in Eqs. (B4) and (B5) may be used to obtain the requested average value of the quantity $n_{ik}n_{il}n_{jq}n_{jp}$: in fact, it is equals to Eq. (B4) if $i=j$ and it is given by Eq. (B5) if $i\neq j$.

In the end, we may write the final formula

$$\begin{aligned} \langle n_{ik} n_{il} n_{jq} n_{jp}\rangle = & \frac{1}{15}\left(2\delta_{kl}\delta_{pq} - \frac{1}{2}\delta_{pk}\delta_{ql} - \frac{1}{2}\delta_{pl}\delta_{qk} \right) \\ & - \frac{1}{15}\delta_{ij}\left(\delta_{kl}\delta_{pq} - \frac{3}{2}\delta_{pk}\delta_{ql} - \frac{3}{2}\delta_{pl}\delta_{qk} \right). \end{aligned} \tag{B6}$$

So, Eqs. (B3) and (B6) can be used to simplify Eq. (B2), which gives the average of the electric field inside the ellipsoid over all its possible orientations. The implicit sums that appear into this expression can be calculated as follows:

$$E_{0l}\langle n_{il}n_{ik}\rangle = \frac{1}{3}E_{0l}\delta_{lk} = \frac{1}{3}E_{0k},$$

$$\begin{aligned} &E_{0l}E_{0q}E_{0p}\langle n_{ik}n_{il}n_{jq}n_{jp}\rangle \\ &= \frac{1}{15}E_{0l}E_{0q}E_{0p}\left(2\delta_{kl}\delta_{pq} - \frac{1}{2}\delta_{pk}\delta_{ql} - \frac{1}{2}\delta_{pl}\delta_{qk}\right) \\ &\quad - \frac{1}{15}E_{0l}E_{0q}E_{0p}\delta_{ij}\left(\delta_{kl}\delta_{pq} - \frac{3}{2}\delta_{pk}\delta_{ql} - \frac{3}{2}\delta_{pl}\delta_{qk}\right) \\ &= \frac{1}{15}E_{0k}E_0^2(1+2\delta_{ij}). \end{aligned} \tag{B7}$$

Finally, after some straightforward calculation we obtain

$$\langle E_{sk}\rangle = E_{0k}\left[\frac{\varepsilon_1}{3}\sum_i \frac{1}{a_i} - \frac{\alpha\varepsilon_1{}^3E_0{}^2}{15}\left(\sum_i\sum_j \frac{L_i}{a_i^2a_j^2} + 2\sum_i \frac{L_i}{a_i^4}\right)\right]. \tag{B8}$$

By letting

$$M = \frac{1}{3}\sum_i \frac{1}{a_i},$$

$$N = \frac{1}{15}\left(\sum_i\sum_j \frac{L_i}{a_i^2a_j^2} + 2\sum_i \frac{L_i}{a_i^4}\right), \tag{B9}$$

we get the final formula for the average value of the internal electric field,

$$\langle \mathbf{E}_s\rangle = \mathbf{E}_0(\varepsilon_1 M - \alpha\varepsilon_1{}^3E_0{}^2N). \tag{B10}$$

APPENDIX C: $[\varepsilon(\|\mathbf{E}_S\|)-\varepsilon_1]\mathbf{E}_S$ AVERAGING OVER ALL ORIENTATIONS

We consider a single ellipsoidal nonlinear inclusion and we search for the average value of the quantity $[\varepsilon(\|\mathbf{E}_s\|)-\varepsilon_1]\mathbf{E}_s$ over all the possible orientations of the particle. From Eqs. (5) and (11) we obtain

$$\begin{aligned} &[\varepsilon(\|\mathbf{E}_s\|)-\varepsilon_1]E_{si} \\ &= \frac{\varepsilon_1}{L_i}(E_{0i}-E_{si}) \\ &= \frac{\varepsilon_1}{L_i}\left(E_{0i} - \frac{\varepsilon_1E_{0i}}{a_i} + \frac{\alpha\varepsilon_1{}^3L_iE_{0i}}{a_i^2}\sum_j \frac{E_{0j}{}^2}{a_j^2}\right); \end{aligned} \tag{C1}$$

therefore, in vector notation,

$$[\varepsilon(\|\mathbf{E}_s\|)-\varepsilon_1]\mathbf{E}_s = \sum_i \frac{\varepsilon_1}{L_i}\left[\mathbf{E}_0\cdot\hat{n}_i - \frac{\varepsilon_1\mathbf{E}_0\cdot\hat{n}_i}{a_i} + \frac{\alpha\varepsilon_1{}^3L_i\mathbf{E}_0\cdot\hat{n}_i}{a_i^2}\sum_j\frac{(\mathbf{E}_0\cdot\hat{n}_j)^2}{a_j^2}\right]\hat{n}_i. \tag{C2}$$

By taking the kth component in the global reference framework, we may write

$$[\varepsilon(\|\mathbf{E}_s\|)-\varepsilon_1]E_{sk} = \sum_i \frac{\varepsilon_1}{L_i}\left(E_{0l}n_{il} - \frac{\varepsilon_1E_{0l}n_{il}}{a_i} + \frac{\alpha\varepsilon_1{}^3L_iE_{0l}n_{il}}{a_i^2}\sum_j \frac{E_{0q}n_{jq}E_{0p}n_{jp}}{a_j^2}\right)n_{ik}, \tag{C3}$$

and averaging

$$\begin{aligned} &\langle[\varepsilon(\|\mathbf{E}_s\|)-\varepsilon_1]E_{sk}\rangle \\ &= \sum_i \frac{\varepsilon_1}{L_i}E_{0l}\left[\langle n_{il}n_{ik}\rangle - \frac{\varepsilon_1\langle n_{il}n_{ik}\rangle}{a_i} + \sum_j \frac{\alpha\varepsilon_1{}^3L_iE_{0q}E_{0p}\langle n_{ik}n_{il}n_{jq}n_{jp}\rangle}{a_i^2a_j^2}\right] \\ &= \sum_i \frac{\varepsilon_1}{L_i}\left[\frac{1}{3}E_{0k} - \frac{\varepsilon_1E_{0k}}{3a_i} + \sum_j \frac{\alpha\varepsilon_1{}^3L_iE_{0k}E_0{}^2(1+2\delta_{ij})}{15a_i^2a_j^2}\right] \\ &= \frac{\varepsilon_1E_{0k}}{3}\left(\sum_i \frac{1}{L_i} - \varepsilon_1\sum_i \frac{1}{a_iL_i}\right) \\ &\quad + \frac{\alpha\varepsilon_1{}^4E_{0k}E_0{}^2}{15}\left(\sum_i\sum_j \frac{1}{a_i^2a_j^2} + 2\sum_1 \frac{1}{a_i^4}\right). \end{aligned} \tag{C4}$$

The first term can be simplified observing that

$$\begin{aligned} &\varepsilon_1E_{0k}\frac{1}{3}\left(\sum_i \frac{1}{L_i} - \varepsilon_1\sum_i\frac{1}{a_iL_i}\right) \\ &= \varepsilon_1E_{0k}\frac{1}{3}\sum_i \frac{a_i-\varepsilon_1}{a_iL_i} \\ &= \varepsilon_1E_{0k}\frac{1}{3}\sum_i \frac{L_i(\varepsilon_2-\varepsilon_1)}{a_iL_i} = \varepsilon_1E_{0k}(\varepsilon_2-\varepsilon_1)M, \end{aligned} \tag{C5}$$

where M is defined in Eq. (B9).

The second term in Eq. (C4) may be simplified by defining the following parameter P:

$$P = \frac{1}{15}\left(\sum_i\sum_j \frac{1}{a_i^2a_j^2} + 2\sum_i \frac{1}{a_i^4}\right). \tag{C6}$$

Summing up, the balance equation for the displacement vector given in Eq. (15) may be recast in the form

$$\begin{aligned} \langle\mathbf{D}\rangle &= \varepsilon_1\langle\mathbf{E}\rangle + c\langle[\varepsilon(\|\mathbf{E}_s\|)-\varepsilon_1]\mathbf{E}_s\rangle \\ &= \varepsilon_1\langle\mathbf{E}\rangle + c\varepsilon_1(\varepsilon_2-\varepsilon_1)M\mathbf{E}_0 + c\alpha\varepsilon_1{}^4PE_0{}^2\mathbf{E}_0. \end{aligned} \tag{C7}$$

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