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Effective permittivity of materials containing graded ellipsoidal inclusions

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Abstract. We prove a generic theorem stating the equivalence between a graded dielectric ellipsoid (with gradation along a family of internal confocal ellipsoids) and an anisotropic homogeneous ellipsoid. We then describe a procedure to obtain the three principal permittivities of the effective ellipsoid for any given dielectric gradation profile. Finally, we apply a multiscale approach to homogenize dispersions of ellipsoidal graded particles.

PACS. 77.22.Ch Permittivity (dielectric function) – 77.84.Lf Composite materials – 85.50.-n Dielectric, ferroelectric, and piezoelectric devices

1 Introduction

Functionally graded composite materials are characterized by internal gradients of composition or structure. Their physical properties are directly affected by the design of such gradients and they are matter of considerable interest in disciplines as diverse as tribology, geology, microelectronics, optoelectronics, biomechanics, fracture mechanics, and nanotechnology. In particular, within the large class of dielectric structures and metamaterials, graded systems have attracted much attention because they offer advantages over traditional composite materials [1–3]. For instance, a graded composition across an interface (either continuous or by discrete steps) can be used to redistribute thermal stresses or electric fields and to reduce stress concentrations at the intersection between an interface and a free surface [4].

The prediction of the effective macroscopic properties of graded media is typically based on the so-called homogenization procedures: a graded structure is ideally substituted by a homogeneous equivalent one, effectively describing the physical response of the overall system [5,6]. There have been a number of attempts in the above direction. A first-principles approach has been developed in order to obtain the electric potential and the electric field for spherical particles having a radially varying dielectric constant [7]. Moreover, a nonlinear differential effective dipole approximation has been developed to analyze the third-order nonlinear susceptibility of graded spherical particles [8]. As a further generalization, a model for second- and third-harmonic generation in random composites of graded spherical particles has been developed [9,10]. Furthermore, the optical properties of compositionally graded films have been obtained with analytical and numerical calculations [11]. Also the conductivity of heterogeneous media with graded anisotropic spherical inhomogeneities has been investigated by means of an energy equivalence principle [12]. A recent application of the transformation field method allows to calculate the effective properties of graded composites having arbitrary periodical structure [13]. The same method has been applied to estimate the effective permittivity of an anisotropic graded granular composite having inclusions of arbitrary shape and arbitrary anisotropic grading profile [14]. Finally, the effective property has been investigated theoretically in graded elliptical cylindrical composites consisting of inhomogeneous graded elliptical cylinders and an isotropic matrix by means of the elliptical cylindrical coordinates [15]. From the experimental point of view we remember that many different ceramic dielectric systems (typically for microwave applications) have been produced and analyzed [16–18].

So far theoretical models have been successfully developed only for specific geometries (i.e. typically for spherical or cylindrical particles), while general results for arbitrarily shaped inclusions are still lacking. In this work we present a thorough theory for graded ellipsoidal particles: in particular, we prove a generic theorem stating the equivalence between a graded dielectric ellipsoidal inclusion and an anisotropic homogeneous ellipsoid with effective dielectric permettivities. We then develop a procedure

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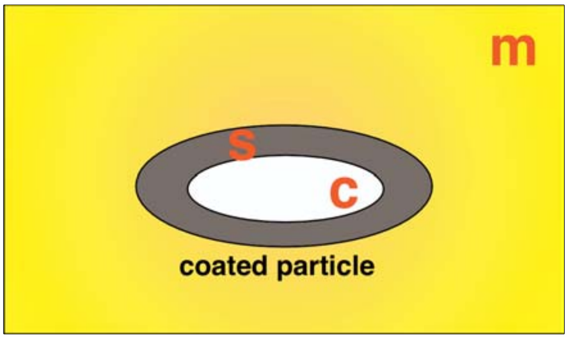

Fig. 1. Coated dielectric ellipsoidal particle with isotropic external shell (s) and internal anisotropic core (c) embedded into a given isotropic matrix (m).

to obtain the three principal permittivities of the effective ellipsoid when the dielectric gradation function is given.

The ellipsoidal shape does represent the most general geometry suitable for many practical applications: in particular, it allows us to analyze two important limiting cases, namely the spherical and the cylindrical ones. Under this respect, the present investigation provides a very general conceptual framework, including those specific cases previously investigated. Our development proceeds through a two-step procedure: firstly, we homogenize a simple coated ellipsoidal particle made of an anisotropic core and of an isotropic external shell; then, we adopt a limiting process to investigate the properties of the graded ellipsoidal inclusion. Once this problem is solved, the homogenization proceeds by averaging over the volume of a dispersion of graded particles [19–21]. This generates an effective mean field theory: such an approach, in previous works, has been successfully introduced both in the linear case [22–24] and in the nonlinear one [25,26]. The two-step procedure is commonly referred to as multiscale approach. In our case the multiscale homogenization first solves the problem inside each graded particle and then it copes with the overall dispersion of inclusions. A similar approach has been applied to the elastic properties of composite or cracked materials [27–30].

The structure of the paper is the following: in Section 2 we analyze the properties of a single coated ellipsoidal particle, in Section 3 we obtain the results describing a graded ellipsoidal inclusion and in Section 4 we present some applications to dispersions of graded inhomogeneities.

2 Coated dielectric ellipsoidal particle

We consider the structure represented in Figure 1 where a dielectric coated particle is embedded into a homogeneous matrix. The core is made of an anisotropic material with principal directions of the permittivity tensor aligned with the geometrical principal directions of the internal ellipsoid (semi-axes a_{c_1}, a_{c_2}, a_{c_3}). The core principal permittivities are ϵ_{3k} for $k = 1, 2, 3$. The shell is formed by an isotropic material with permittivity ϵ_2 (semi-axes a_{s_1}, a_{s_2}, a_{s_3}). Finally, the embedding homogeneous matrix is isotropic with permittivity ϵ_1. The two ellipsoids have the same foci and, therefore, they are both described by the following family of confocal ellipsoids

$$\frac{x_1^2}{a_{s_1}^2+\xi}+\frac{x_2^2}{a_{s_2}^2+\xi}+\frac{x_3^2}{a_{s_3}^2+\xi}=1. \tag{1}$$

If $\xi = 0$ equation (1) describes the external shell (semi-axes a_{s_1}, a_{s_2}, a_{s_3}) while if $\xi = \xi_c$ it describes the surface of the internal core (semi-axes a_{c_1}, a_{c_2}, a_{c_3}). Therefore, we have $a_{s_i}^2 + \xi_c = a_{c_i}^2$. We assume that the external semi-axes are ordered as follows: $0 < a_{s_3} < a_{s_2} < a_{s_1}$. So, inside the particle we always have $-a_{s_3}^2 < \xi < 0$. A given value of ξ in this range represents an ellipsoid placed inside the composite particle. It is also useful to introduce the volume fraction $c = (a_{c_1}a_{c_2}a_{c_3})/(a_{s_1}a_{s_2}a_{s_3})$ of the core into the whole inclusion.

We prove the following property: under the effect of a uniform electric field, the inclusion composed by an anisotropic core of permittivities ϵ_{3k} (for k=1,2,3) with volume fraction c and by an isotropic confocal shell of permittivity ϵ_2 (see Fig.1 for details) is exactly equivalent to an anisotropic homogeneous ellipsoid with principal permittivities

$$\tilde{\epsilon}_k=\epsilon_2\frac{[(\epsilon_{3k}-\epsilon_2)\,L_{ck}+\epsilon_2]+c\,(\epsilon_{3k}-\epsilon_2)\,(1-L_{sk})}{[(\epsilon_{3k}-\epsilon_2)\,L_{ck}+\epsilon_2]-c\,(\epsilon_{3k}-\epsilon_2)\,L_{sk}} \tag{2}$$

where L_{ck} and L_{sk} are the depolarization factors of the core and of the shell, respectively

$$L_{ck}=\frac{a_{c_1}a_{c_2}a_{c_3}}{2}\int_0^{+\infty}\frac{dt}{\left(a_{c_k}^2+t\right)\sqrt{\prod_{i=1}^{3}(a_{c_i}^2+t)}}$$

$$L_{sk}=\frac{a_{s_1}a_{s_2}a_{s_3}}{2}\int_0^{+\infty}\frac{dt}{\left(a_{s_k}^2+t\right)\sqrt{\prod_{i=1}^{3}(a_{s_i}^2+t)}}. \tag{3}$$

We also remark that L_{ck} and L_{sk} can be written in terms of elliptic integrals as discussed in Appendix A.

To prove this property we consider the structure represented in Figure 1 under the effect of an externally applied uniform electric field $\boldsymbol{E}_0 = (E_{01}, E_{02}, E_{03})$. The potentials ϕ_c in the core, ϕ_s in the shell and ϕ_m in the matrix can be expressed by [31]

$$\phi_c = C_k x_k \tag{4}$$

$$\phi_s = S_k x_k + T_k x_k\int_\xi^{+\infty}\frac{dt}{R\left(a_{s_k}^2+t\right)} \tag{5}$$

$$\phi_m = -E_{0k}x_k + Q_k x_k\int_\xi^{+\infty}\frac{dt}{R\left(a_{s_k}^2+t\right)} \tag{6}$$

where the summation over k has been implicitely assumed, $R(t) = \sqrt{(a_{s_1}^2+t)(a_{s_2}^2+t)(a_{s_3}^2+t)}$ and the variable ξ is defined by equation (1). The unknown coefficients C_k, S_k, T_k and Q_k can be found by forcing the continuity of the electric potential

$$\begin{aligned} \phi_c &= \phi_s \quad \text{at } \xi = \xi_c \\ \phi_s &= \phi_m \quad \text{at } \xi = 0 \end{aligned} \tag{7}$$

and of the normal component of the electric displacement

$$\begin{aligned} \epsilon_{3k} \frac{\partial \phi_c}{\partial x_k} n_k &= \epsilon_2 \frac{\partial \phi_s}{\partial n} \quad \text{at } \xi = \xi_c \\ \epsilon_2 \frac{\partial \phi_s}{\partial n} &= \epsilon_1 \frac{\partial \phi_m}{\partial n} \quad \text{at } \xi = 0 \end{aligned} \tag{8}$$

where n_k are the components of the normal unit vector of the surface $\xi = \xi_c$ and $\partial/\partial n$ is the directional derivative taken in the direction normal to the surface $\xi = \xi_c$ or $\xi = 0$. Equations (7) and (8) drive to

$$\begin{aligned} C_k &= \frac{\epsilon_1 \epsilon_2}{Z_k} E_{0k} \\ S_k &= \frac{\epsilon_1 \left[(\epsilon_{3k} - \epsilon_2) L_{ck} + \epsilon_2 \right]}{Z_k} E_{0k} \\ T_k &= -\frac{1}{2} \frac{\epsilon_1 a_{c_1} a_{c_2} a_{c_3} (\epsilon_{3k} - \epsilon_2)}{Z_k} E_{0k} \\ Q_k &= \frac{a_{s_1} a_{s_2} a_{s_3}}{2 Z_k} \left\{ c (\epsilon_{3k} - \epsilon_2) \left[(\epsilon_2 - \epsilon_1) L_{sk} - \epsilon_2 \right] \right. \\ &\quad \left. - (\epsilon_2 - \epsilon_1) \left[(\epsilon_{3k} - \epsilon_2) L_{ck} + \epsilon_2 \right] \right\} E_{0k} \end{aligned} \tag{9}$$

where

$$\begin{aligned} Z_k &= c (\epsilon_{3k} - \epsilon_2)(\epsilon_2 - \epsilon_1) L_{sk} (L_{sk} - 1) \\ &\quad - \left[(\epsilon_2 - \epsilon_1) L_{sk} + \epsilon_1 \right] \left[(\epsilon_{3k} - \epsilon_2) L_{ck} + \epsilon_2 \right]. \end{aligned} \tag{10}$$

It is important to remark that the external field is completely controlled by the coefficients Q_k, representing the effect of the inhomogeneity on the electric potential in the surrounding matrix.

Our aim is to work out a procedure to define an effective homogeneous inclusion having the same dielectric properties of a coated particle. We therefore consider the following substitutions in equation (9) for Q_k: $\epsilon_{3k} \to \tilde{\epsilon}_k$ (effective permettivities), $a_{c_k} = a_{s_k}$ and $L_{ck} = L_{sk}$ (for $k = 1, 2, 3$), thus obtaining

$$Q_k = \frac{a_{s_1} a_{s_2} a_{s_3}}{2} \frac{\tilde{\epsilon}_k - \epsilon_1}{(\tilde{\epsilon}_k - \epsilon_1) L_{sk} + \epsilon_1} E_{0k}. \tag{11}$$

These coefficients describe the behavior of the external field for an anisotropic ellipsoid of semi-axes a_{s_k} and permittivities $\tilde{\epsilon}_k$ placed in a matrix with dielectric constant ϵ_1 (without shell). By drawing a comparison between equation (9) and equation (11) we obtain an equation for the effective permittivities of the composite inclusion

$$\begin{aligned} &\frac{1}{Z_k} \left\{ c (\epsilon_{3k} - \epsilon_2) \left[(\epsilon_2 - \epsilon_1) L_{sk} - \epsilon_2 \right] \right. \\ &\qquad \left. - (\epsilon_2 - \epsilon_1) \left[(\epsilon_{3k} - \epsilon_2) L_{ck} + \epsilon_2 \right] \right\} \\ &\qquad\qquad = \frac{\tilde{\epsilon}_k - \epsilon_1}{(\tilde{\epsilon}_k - \epsilon_1) L_{sk} + \epsilon_1} \end{aligned} \tag{12}$$

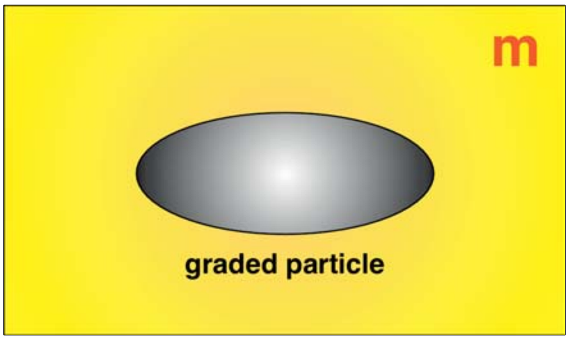

Fig. 2. Functionally graded inclusion with arbitrary permittivity profile $\epsilon(\xi)$ for $-a_{s_3}^2 < \xi < 0$.

Finally, by solving the above equation for $\tilde{\epsilon}_k$ we obtain the expression shown in equation (2). We remark that the result given in equation (2) does not depend on ϵ_1 and depends only on the internal properties of the particle.

This theorem plays a crucial role in the following development of the theory.

3 Graded ellipsoidal inclusion

The property given in equation (2) allows us to extend our formalism to the case of functionally graded particles with arbitrary permittivity profile $\epsilon(\xi)$ for $-a_{s_3}^2 < \xi < 0$ (see Fig. 2). The following result holds for graded dielectric inclusions: under the effect of a uniform electric field, the graded ellipsoidal particle with permittivity profile $\epsilon(\xi)$ (in the entire range $-a_{s_3}^2 < \xi < 0$) is exactly equivalent to an homogeneous anisotropic ellipsoid with principal permittivities given by $\epsilon_k(0)$ where the functions $\epsilon_k(\xi)$ are solutions of the following differential Riccati equations

$$\begin{aligned} \frac{d\epsilon_k(\xi)}{d\xi} &= -\frac{\left[\epsilon_k(\xi) - \epsilon(\xi) \right]^2}{2\epsilon(\xi) \left(a_{s_k}^2 + \xi \right)} - \frac{1}{R} \frac{dR}{d\xi} \left[\epsilon_k(\xi) - \epsilon(\xi) \right] \\ \epsilon_k\left(-a_{s_3}^2 \right) &= \epsilon\left(-a_{s_3}^2 \right) \end{aligned} \tag{13}$$

where $R(\xi) = \sqrt{(a_{s_1}^2 + \xi)(a_{s_2}^2 + \xi)(a_{s_3}^2 + \xi)}$. The values $\epsilon_k(s)$ (for a given s in the entire range $-a_{s_3}^2 < s < 0$) represent the effective principal permittivities of the ellipsoid defined by the bounds $-a_{s_3}^2 < \xi < s$.

To prove this property we take into consideration an infinitesimal ellipsoidal layer $(\xi, \xi + d\xi)$ in the functionally graded inclusion. The idea consists in applying the property derived in Section 2 to the composite particle made of the core $(-a_{s_3}^2, \xi)$ and of the coating $(\xi, \xi + d\xi)$. We suppose that the region $(-a_{s_3}^2, \xi)$ has been homogenized by obtaining the effective principal permittivities $\epsilon_k(\xi)$ (anisotropic core in this conceptual scheme). The infinitesimal ellipsoidal layer $(\xi, \xi + d\xi)$ is characterized by the value $\epsilon(\xi)$ of the profile permittivity (isotropic shell in this conceptual scheme). Since the effective principal permittivities $\epsilon_k(\xi + d\xi)$ of the larger region $(-a_{s_3}^2, \xi + d\xi)$ is straightforwardly obtained by equation (2), we get

$$\epsilon_k(\xi + d\xi) = \epsilon(\xi) \frac{N_\xi}{D_\xi} \tag{14}$$

where

$$N_\xi = [\epsilon_k(\xi) - \epsilon(\xi)]\, L_k(\xi) + \epsilon(\xi) + c\,[\epsilon_k(\xi) - \epsilon(\xi)]\,[1 - L_k(\xi + d\xi)] \quad (15)$$

$$D_\xi = [\epsilon_k(\xi) - \epsilon(\xi)]\, L_k(\xi) + \epsilon(\xi) - c\,[\epsilon_k(\xi) - \epsilon(\xi)]\, L_k(\xi + d\xi). \quad (16)$$

In this case the volume fraction c is given (up to the first order in $d\xi$) by the relationship

$$c = \frac{R(\xi)}{R(\xi + d\xi)} = \frac{R(\xi)}{R(\xi) + \frac{dR(\xi)}{d\xi} d\xi} = \frac{1}{1 + \frac{1}{R(\xi)}\frac{dR(\xi)}{d\xi} d\xi} = 1 - \frac{1}{R(\xi)}\frac{dR(\xi)}{d\xi} d\xi. \quad (17)$$

In order to obtain equation (17) we made use of the quantity $R(\xi) = \sqrt{(a_{s_1}^2 + \xi)(a_{s_2}^2 + \xi)(a_{s_3}^2 + \xi)}$, which represents the product of the three semi-axes of the ellipsoid with a given ξ: c is the ratio between the volumes or, equivalently, the ratio betwen the corresponding values of R. Moreover, the depolarization factors $L_k(\xi)$ of the ellipsoid defined by ξ are given by

$$L_k(\xi) = \frac{R(\xi)}{2} \int_0^{+\infty} \frac{ds}{R(\xi + s)\left(a_{s_k}^2 + \xi + s\right)}. \quad (18)$$

From equation (14) we may build the difference quotient for the variable $\epsilon_k(\xi)$

$$\frac{\epsilon_k(\xi + d\xi) - \epsilon_k(\xi)}{d\xi} = \frac{\epsilon(\xi)}{d\xi}\frac{N_\xi}{D_\xi} - \frac{\epsilon_k(\xi)}{d\xi}. \quad (19)$$

By performing the limit $d\xi \to 0$ and by using equation (17) for the volume fraction c, we obtain the first form of the differential equation

$$\frac{d\epsilon_k}{d\xi} = \frac{(\epsilon_k - \epsilon)\left\{(\epsilon_k - \epsilon)\frac{dL_k}{d\xi} - \frac{1}{R}\frac{dR}{d\xi}\left[(\epsilon_k - \epsilon) L_k + \epsilon\right]\right\}}{\epsilon}. \quad (20)$$

Now, the derivative of the depolarization factors $L_k(\xi)$ can be found as described in Appendix B

$$\frac{dL_k(\xi)}{d\xi} = \frac{1}{R(\xi)}\frac{dR(\xi)}{d\xi} L_k(\xi) - \frac{1}{2\left(a_{s_k}^2 + \xi\right)}. \quad (21)$$

Finally, the substitution of equation (21) in equation (20) allows us to obtain the final result shown in equation (13).

This property, proved for an arbitrarily shaped ellipsoid, can be applied to any geometry, including spheres or cylinders. If we consider a sphere ($a_{s_1} = a_{s_2} = a_{s_3} = R$) we have for symmetry reasons $\epsilon_1 = \epsilon_2 = \epsilon_3 = \epsilon_{eq}$; moreover, we may use the change of variable $R^2 + \xi = r^2$ in equation (13) obtaining the following differential equation for the effective permettivity in terms of the radius r

$$\frac{d}{dr}\left[r\epsilon_{eq}(r)\right] = 2\epsilon(r) - \frac{\epsilon_{eq}(r)^2}{\epsilon(r)};\ \epsilon_{eq}(0) = \epsilon(0) \quad (22)$$

Such a result agrees with the so-called Tartar formula, obtained in earlier literature [8]. Similarly, for a circular cylinder we have $a_{s_1} \to +\infty$ and $a_{s_2} = a_{s_3} = R$; the effective permittivities are $\epsilon_1 = \epsilon_\parallel$ (longitudinal permittivity) and $\epsilon_2 = \epsilon_3 = \epsilon_\perp$ (transversal permittivity). Once again, we may use the change of variable $R^2 + \xi = r^2$ in equation (13) obtaining the following results for the effective permettivities in terms of the radius r

$$\frac{d}{dr}\left[r\epsilon_\perp(r)\right] = \epsilon_\perp(r) + \epsilon(r) - \frac{\epsilon_\perp(r)^2}{\epsilon(r)};\ \epsilon_\perp(0) = \epsilon(0)$$

$$\epsilon_\parallel(r) = \frac{2}{r^2}\int_0^r \eta\epsilon(\eta)\, d\eta. \quad (23)$$

We remark that the transversal permittivity is the solution of a Riccati differential equation, while the longitudinal permittivity is equal to the average value of the permittivity function $\epsilon(r)$ over a section of the circular cylinder.

4 Dispersions of ellipsoidal graded inclusions

In this Section we apply the formal results derived in Section 3 to a dispersion of functionally graded ellipsoidal particles, i.e. to the most common situation found in material science.

According to the standard homogenization approach, we begin by considering a single functionally graded particle with a given permittivity profile and by solving the differential equations for the principal permittivities of the effective homogeneous inclusion. To this aim, the fourth-order Runge Kutta algorithm [32] was implemented to integrate the differential problem given in equation (13) with step size 1/1000. It was verified that this step size guarantees accurate numerical results. We have taken into consideration a functionally graded ellipsoid ($a_{s_1} = 3, a_{s_2} = 2.3, a_{s_3} = 2$) with a power law $\epsilon(\xi) = \epsilon_A + (\epsilon_B - \epsilon_A)(1 + \xi/a_{s_3}^2)^n$ where the exponent n can assume different values. Such a dielectric profile has been considered for different reasons: firstly, it is generic enough to fit many composition gradients used in real applications; secondly, in recent literature several exact solutions have been proposed for spheres and cylinders, having a similar dielectric profile varying along the radius of the particles: these solutions have been adopted for validating some homogenization procedures [7,8]. We remark that the quantity $1 + \xi/a_{s_3}^2$ has the role of a sort of *dimensionless radius* ranging from 0 (center of the particle) to 1 (external surface of the particle). In Figure 3 we report our results for an increasing power law with $\epsilon_A = 1, \epsilon_B = 10$ and in Figure 4 for a decreasing power law with $\epsilon_A = 10, \epsilon_B = 1$. In both cases we have computed the solution for $n = 1/4, 1/2, 1, 2, 4$.

In Figure 5 we show the results for a periodic profile of the dielectric constant $\epsilon(\xi) = \epsilon_0 + \Delta\epsilon \cos[2\pi\nu(1 + \xi/a_{s_3}^2)]$ with $\epsilon_0 = 1, \Delta\epsilon = 0.9$ and spatial frequency $\nu = 2$ or $\nu = 8$. This kind of periodicity is useful to model composite materials of smooth multishell inclusions [4]. We

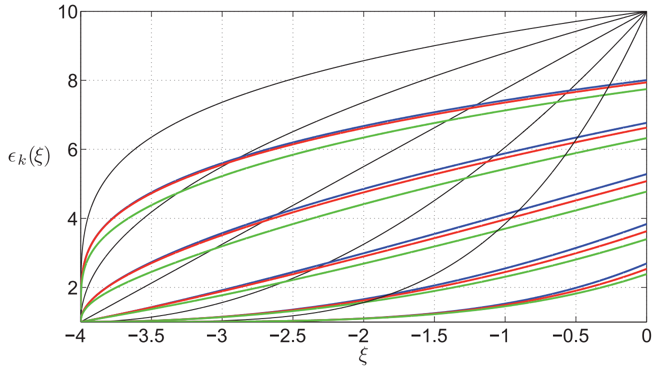

Fig. 3. (Color online) Solutions for a functionally graded ellipsoid ($a_{s_1} = 3, a_{s_2} = 2.3, a_{s_3} = 2$) with a increasing power law $\epsilon(\xi) = \epsilon_A + (\epsilon_B - \epsilon_A)(1 + \xi/a_{s_3}^2)^n$ where $\epsilon_A = 1, \epsilon_B = 10$ and $n = 1/4, 1/2, 1, 2, 4$. The black lines correspond to the profiles $\epsilon(\xi)$ and the colored lines correspond to $\epsilon_1(\xi)$ (blu), $\epsilon_2(\xi)$ (red) and $\epsilon_3(\xi)$ (green).

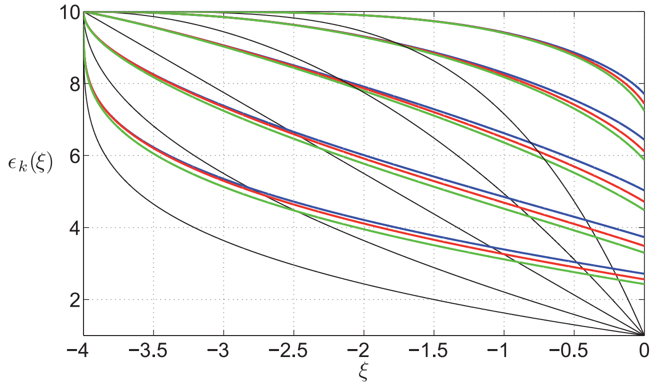

Fig. 4. (Color online) Solutions for a functionally graded ellipsoid ($a_{s_1} = 3, a_{s_2} = 2.3, a_{s_3} = 2$) with a decreasing power law $\epsilon(\xi) = \epsilon_A + (\epsilon_B - \epsilon_A)(1 + \xi/a_{s_3}^2)^n$ where $\epsilon_A = 10, \epsilon_B = 1$ and $n = 1/4, 1/2, 1, 2, 4$. The black lines correspond to the profiles $\epsilon(\xi)$ and the colored lines correspond to $\epsilon_1(\xi)$ (blu), $\epsilon_2(\xi)$ (red) and $\epsilon_3(\xi)$ (green).

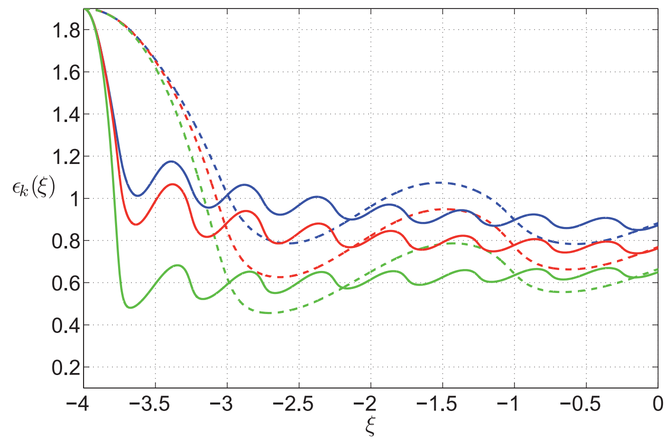

Fig. 5. (Color online) Solutions for a functionally graded ellipsoid ($a_{s_1} = 3, a_{s_2} = 2.3, a_{s_3} = 2$) with a periodic law $\epsilon(\xi) = \epsilon_0 + \Delta\epsilon \cos[2\pi\nu(1 + \xi/a_{s_3}^2)]$ where $\epsilon_0 = 1, \Delta\epsilon = 0.9$, $\nu = 2$ (dashed lines) and $\nu = 8$ (continuous lines). The curves correspond to $\epsilon_1(\xi)$ (blu), $\epsilon_2(\xi)$ (red) and $\epsilon_3(\xi)$ (green).

observe that, although the electric behavior of the particle is isotropic at any point (graded but scalar permettivity), the effective electric behavior of the whole inclusion is anisotropic because of the geometrical anisotropy in the spatial gradation of the dielectric constant. This means that the graded ellipsoidal geometry generates effective dielectric anisotropy, independently of the isotropy of the local electric behavior of the material. This is a remarkable feature of the system under investigation which can actually be proved for any kind of dielectric profile and any ratio among the axis length of the graded ellipsoid.

We move now to the case of a population of functionally graded ellipsoidal particles. We assume that each particle has semi-axes a_{s_1}, a_{s_2} and a_{s_3} and a given permittivity profile $\epsilon(\xi)$ for $-a_{s_3}^2 < \xi < 0$. We consider that all the particles are randomly oriented in a given matrix with scalar permittivity ϵ_1 and with a given volume fraction f. Each of these particles can be substituted by an homogeneous anisotropic inclusion with principal permittivities $\epsilon_1(0)$, $\epsilon_2(0)$ and $\epsilon_3(0)$. These values can be obtained with the first part of the homogenization procedure, i.e. with the integration of equation (13). The second part of the homogenizing theory consists in evaluating the effective dielectric constant ϵ_{eff} of the entire dispersion. A generalization of the Maxwell-Garnett theory may be used in order to take into account the anisotropic character of the embedded particles [23–25]

$$\begin{aligned}\epsilon_{eff} &= \epsilon_1 + \frac{\frac{1}{3} f \sum_{k=1}^{3} \frac{\epsilon_1 [\epsilon_k(0) - \epsilon_1]}{\epsilon_1 + L_{sk}[\epsilon_k(0) - \epsilon_1]}}{1 - f + \frac{1}{3} f \sum_{k=1}^{3} \frac{\epsilon_1}{\epsilon_1 + L_{sk}[\epsilon_k(0) - \epsilon_1]}} \\ &= \epsilon_1 + \frac{1}{3} f \sum_{k=1}^{3} \frac{\epsilon_1 [\epsilon_k(0) - \epsilon_1]}{\epsilon_1 + L_{sk}[\epsilon_k(0) - \epsilon_1]} + O\left(c^2\right)\end{aligned} \tag{24}$$

where the depolarization factors L_{sk} of the ellipsoids are defined in equation (3). Typically, the Maxwell-Garnett approximation only works for strongly dilute dispersions. Therefore, a further generalization is given by the Bruggeman differential scheme [21,23], which leads to the differential equation

$$\frac{d\epsilon_{eff}}{df} = \frac{1}{1-f} \epsilon_{eff} \frac{1}{3} \sum_{k=1}^{3} \frac{\epsilon_k(0) - \epsilon_{eff}}{\epsilon_{eff} + L_{sk}[\epsilon_k(0) - \epsilon_{eff}]} \tag{25}$$

with the intial condition $\epsilon_{eff}(f = 0) = \epsilon_1$ (permettivity of the matrix). The solution of this equation represents the second step in the multiscale homogenization procedure.

We take in consideration a graded ellipsoid ($a_{s_1} = 3, a_{s_2} = 2.3, a_{s_3} = 2$) with a power law $\epsilon(\xi) = \epsilon_A + (\epsilon_B - \epsilon_A)(1 + \xi/a_{s_3}^2)^n$ where $\epsilon_A = 10, \epsilon_B = 1$. In Figures 6 and 7 we have shown the effective permittivity ϵ_{eff} (for $\epsilon_1 = 1$ and $\epsilon_1 = 10$) in terms of the volume fraction f and the gradation exponent n. We can observe the effects of the gradation on the effective properties of the overall mixture. In our simulations we have considered only static permittivities, but we point out that our methodology can be also applied to the case of frequency dependent permittivities.

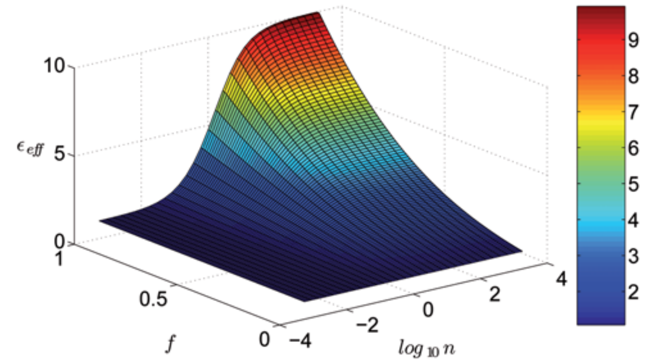

Fig. 6. (Color online) Application of the Bruggeman scheme, equation (25), to a disperion of functionally graded ellipsoids ($a_{s_1} = 3, a_{s_2} = 2.3, a_{s_3} = 2$) with profile $\epsilon(\xi) = \epsilon_A + (\epsilon_B - \epsilon_A)(1 + \xi/a_{s_3}^2)^n$ where $\epsilon_A = 10, \epsilon_B = 1$ embedded in a matrix of permittivity $\epsilon_1 = 1$. The solution ϵ_{eff} has been shown in terms of the volume fraction $0 < f < 1$ and of the exponent $\log_{10} n$.

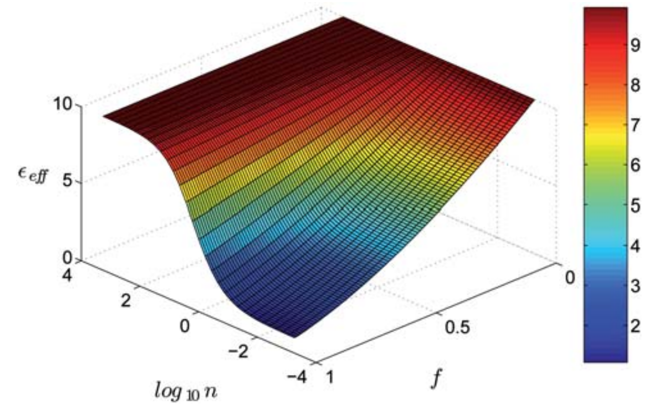

Fig. 7. (Color online) Application of the Bruggeman scheme, equation (25), to a disperion of functionally graded ellipsoids ($a_{s_1} = 3, a_{s_2} = 2.3, a_{s_3} = 2$) with profile $\epsilon(\xi) = \epsilon_A + (\epsilon_B - \epsilon_A)(1 + \xi/a_{s_3}^2)^n$ where $\epsilon_A = 10, \epsilon_B = 1$ embedded in a matrix of permittivity $\epsilon_1 = 10$. The solution ϵ_{eff} has been shown in terms of the volume fraction $0 < f < 1$ and of the exponent $\log_{10} n$.

For example, it is possible to consider Drude-like dielectric constant with ξ-dependent plasma frequency and/or ξ-dependent damping coefficient. The graded Drude dielectric function is very useful to analyze the properties of metal-dielectric composites [8].

5 Conclusion

In this work we have developed a theory for homogenizing a graded dielectric ellipsoid with arbitrary shape and arbitrary dielectric profile. To obtain the results we have firstly analyzed the simple case of a coated ellipsoidal particle and then we have approached the problem of the gradation by means of a limiting process. Such a procedure leads to a system of three Riccati differential equations, which can be solved with standard numerical methods. Moreover, we have described a multiscale procedure to obtain the effective permittivity of a dispersion of randomly oriented graded ellipsoidal particles, embedded in a given matrix. The proved equivalence between a graded particle and an effective anisotropic homogeneous one allows us to apply the classical mixture theories to the case of functionally graded inclusions.

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Appendix A: Depolarization factors

We consider the core ellipsoid with semi-axes $a_{c_1} > a_{c_2} > a_{c_3} > 0$ and the shell ellipsoid with semi-axes $a_{s_1} > a_{s_2} > a_{s_3} > 0$, both aligned to the reference frame under consideration. For the core we define two aspect ratios as $0 < e_c = a_{c_3}/a_{c_2} < 1$ e $0 < g_c = a_{c_2}/a_{c_1} < 1$. Similarly, for the shell we define two aspect ratios as $0 < e_s = a_{s_3}/a_{s_2} < 1$ e $0 < g_s = a_{s_2}/a_{s_1} < 1$. The depolarization factors L_{ck} and L_{sk}, defined in equation (3), depend on such aspect ratios as follows [23]

$$\begin{aligned}
L_{\alpha 1} &= \frac{e_\alpha g_\alpha^2}{(1-g_\alpha^2)\sqrt{1-e_\alpha^2 g_\alpha^2}}\left[\mathcal{F}(v_\alpha, q_\alpha) - \mathcal{E}(v_\alpha, q_\alpha)\right] \\
L_{\alpha 2} &= \frac{e_\alpha\left(1-e_\alpha^2 g_\alpha^2\right)}{(1-e_\alpha^2)(1-g_\alpha^2)\sqrt{1-e_\alpha^2 g_\alpha^2}}\mathcal{E}(v_\alpha, q_\alpha) \\
&\quad - \frac{e_\alpha g_\alpha^2}{(1-g_\alpha^2)\sqrt{1-e_\alpha^2 g_\alpha^2}}\mathcal{F}(v_\alpha, q_\alpha) - \frac{e_\alpha^2}{1-e_\alpha^2} \\
L_{\alpha 3} &= \frac{1}{1-e_\alpha^2} - \frac{e_\alpha}{(1-e_\alpha^2)\sqrt{1-e_\alpha^2 g_\alpha^2}}\mathcal{E}(v_\alpha, q_\alpha) \qquad (26)
\end{aligned}$$

where $\alpha = c$ or s (core or shell) and the quantities v_α e q_α are defined as follows

$$\begin{aligned}
v_\alpha &= \arcsin\sqrt{1-e_\alpha^2 g_\alpha^2} \\
q_\alpha &= \sqrt{(1-g_\alpha^2)/(1-e_\alpha^2 g_\alpha^2)}. \qquad (27)
\end{aligned}$$

The functions $\mathcal{F}(v,q)$ and $\mathcal{E}(v,q)$ are incomplete elliptic integrals of the first and second kind, respectively [33,34]

$$\begin{aligned}
\mathcal{F}(v,q) &= \int_0^v \frac{d\alpha}{\sqrt{1-q^2\sin^2\alpha}} = \int_0^{\sin v} \frac{dx}{\sqrt{(1-x^2)(1-q^2x^2)}} \\
\mathcal{E}(v,q) &= \int_0^v \sqrt{1-q^2\sin^2\alpha}\,d\alpha = \int_0^{\sin v} \frac{\sqrt{1-q^2x^2}}{\sqrt{1-x^2}}dx. \qquad (28)
\end{aligned}$$

Appendix B: Derivative of $L_k(\xi)$

We start with the definition of the depolarization factors $L_k(\xi)$ given in equation (18) and we develop the differentiation with respect to ξ. The application of the product rule leads to the first form

$$\frac{dL_k(\xi)}{d\xi} = \frac{R'(\xi)}{2}\int_0^{+\infty}\frac{ds}{R(\xi+s)\left(a_{s_k}^2+\xi+s\right)}$$
$$-\frac{R(\xi)}{2}\int_0^{+\infty}\left[\frac{R'(\xi+s)}{R^2(\xi+s)\left(a_{s_k}^2+\xi+s\right)}\right.$$
$$\left.+\frac{1}{R(\xi+s)\left(a_{s_k}^2+\xi+s\right)}\right]ds \qquad (29)$$

where $R'(\xi) = \frac{dR(\xi)}{d\xi}$ and $R'(\xi+s) = \frac{dR(\xi+s)}{d\xi} = \frac{dR(\xi+s)}{ds}$. Now we define the integral $I(\xi)$ as follows (it is the first part of the second integral in Eq. (29))

$$I(\xi) = \int_0^{+\infty}\frac{dR(\xi+s)}{ds}\frac{1}{R^2(\xi+s)\left(a_{s_k}^2+\xi+s\right)}ds. \qquad (30)$$

It can be rearranged by means of an integration by parts obtaining the equality

$$I(\xi) + \int_0^{+\infty}\frac{ds}{R(\xi+s)\left(a_{s_k}^2+\xi+s\right)} = \frac{1}{R(\xi)\left(a_{s_k}^2+\xi\right)}. \qquad (31)$$

Finally, by using the above equation (31) in equation (29) we obtain the final result as reported in equation (21). It is interesting to note that equation (21) with the Cauchy initial conditions $L_k(0) = L_{sk}$ is a linear differential equation (with variable coefficients) equivalent to the definition of $L_k(\xi)$ given in equation (18).

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