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Dielectric response of graded spherical particles of anisotropic materials

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Anisotropic dielectric response occurs naturally due to the presence of gradation, like in functionally graded materials or graded biological cells. However, these materials with locally anisotropic dielectric responses can have macroscopically isotropic responses. In treating graded particles of anisotropic materials, traditional isotropic gradation methods need to be modified. In this work, we developed a first-principles approach, as well as an anisotropic differential effective dipole approximation, for calculating the dipole moment of these particles. To this end, the two methods are shown in excellent agreement. As a result, these approaches offer convenient and effective ways to investigate the dielectric properties and optical responses of graded spherical particles of anisotropic materials, as well as the electrokinetic phenomena of biological cells. [DOI: 10.1063/1.1633648]

I. INTRODUCTION

The concept of functionally graded materials (FGM), which are heterogeneous materials with spatially varying material properties,[1] was first proposed to develop heat-resistant material for the propulsion system and airframe of space shuttles.[2] These materials have received much attention, both analytical and experimental, as one of the advanced heterogeneous composite materials in various engineering applications by using gradients in thermal,[3–5] electric,[6,7] and mechanical properties.[8–13] The main characteristic that distinguishes FGM from conventional composite materials is the tailoring of graded composition and microstructure in an intentional manner according to the distribution of properties needed to achieve the desired function.[14] In nature, there are also many graded materials, such as biological cells because of the inhomogeneous compartments inside the nuclei.[15]

Due to the existence of gradation in graded particles, anisotropic dielectric response occurs naturally. However, these particles with local anisotropy are often macroscopically isotropic.[16] In this sense, the existing isotropic gradation models[15,17,18] for describing the isotropic graded particles are no longer valid, and should be modified accordingly. In this work, we shall develop a first-principles approach[17,18] and an anisotropic differential effective dipole approximation (ADEDA), respectively, for calculating the dipole moment of such anisotropic graded particles. We will show that the two methods agree with each other, for the case of linear gradation profiles inside the particles. Note the first-principles approach is herein performed for linear gradation profiles, whereas the ADEDA is valid for *arbitrary* gradation profiles.

This paper is organized as follows. In Sec. II, we will solve the field equations analytically for anisotropic graded spherical particle and obtain the exact solution for a linear profile. In Sec. III, we use the exact solution to validate a proposed approximation theory. A discussion and conclusions are given in Sec. IV.

II. FORMALISM

Let us consider a graded spherical particle with radius a. We adopt the spherical coordinates for convenience. The graded spherical particle has a tangential permittivity in the plane orthogonal to the radial vector of the sphere $[\varepsilon_{\theta\theta}(r) = \varepsilon_{\phi\phi}(r)]$, and a radial permittivity $\varepsilon_{rr}(r)$. Both $\varepsilon_{\theta\theta}(r)$ and $\varepsilon_{rr}(r)$ will be prescribed by radial functions. In view of the symmetry, the anisotropic permittivity of the graded sphere can be expressed as tensor $\vec{\varepsilon}_c(r)$,[19] namely,

$$\vec{\varepsilon}_c(r)=\begin{pmatrix} \varepsilon_{rr}(r) & 0 & 0 \\ 0 & \varepsilon_{\theta\theta}(r) & 0 \\ 0 & 0 & \varepsilon_{\phi\phi}(r) \end{pmatrix}. \quad (1)$$

It is worth noting that the above form is in spherical coordinates, rather than in Cartesian coordinates.

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We assume that such anisotropic graded spheres are randomly embedded in an isotropic homogeneous host medium with permittivity ε_m. The composite is subject to an external uniform electric field $\mathbf{E_0}$ along the z axis. When the composite under consideration is in the dilute limit, the interaction among the inclusions can be neglected, and hence the effective dielectric response of the composite can be obtained from the response of a single inclusion under an effective electric field $\overline{E}$. In this case, the constitutive relation between the displacement and the electric field reads $\mathbf{D}_c = \overleftrightarrow{\varepsilon}_c(r)\cdot\mathbf{E}_c$ for the anisotropic graded spherical inclusion, and $\mathbf{D}_m = \varepsilon_m \mathbf{E}_m$ for the host medium. The Maxwell equations read $\nabla\cdot\mathbf{D}=0$, $\nabla\times\mathbf{E}=0$, and hence $\mathbf{E}=-\nabla\Phi$, where Φ is an electric potential. The Laplace equation for the electric potential Φ is given by

$$\nabla\cdot(\overleftrightarrow{\varepsilon}_c(r)\cdot\nabla\Phi)=0. \tag{2}$$

In spherical coordinates, Eq. (2) can be rewritten as

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\varepsilon_{rr}(r)\frac{\partial\Phi}{\partial r}\right)+\frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\,\varepsilon_{\theta\theta}(r)\frac{\partial\Phi}{\partial r}\right)+\frac{1}{r^2\sin^2\theta}\frac{\partial}{\partial\psi}\left(\varepsilon_{\theta\theta}(r)\frac{\partial\Phi}{\partial\psi}\right)=0. \tag{3}$$

If the applied electric field is along the z axis, the potential Φ is independent of the angle ψ. Taking $\Phi=R(r)\Theta(\theta)$, we obtain the ordinary differential equation for the radial function $R(r)$,

$$\frac{1}{\varepsilon_{\theta\theta}(r)}\frac{d}{dr}\left(r^2\varepsilon_{rr}(r)\frac{dR}{dr}\right)-n(n+1)R=0. \tag{4}$$

In what follows, we will consider the gradation profile as linear radial functions,[17]

$$\varepsilon_{rr}(r)=\varepsilon(0)+g(r/a),\quad \varepsilon_{\theta\theta}(r)=\varepsilon(0)+h(r/a), \tag{5}$$

where g and h are two constants, and $\varepsilon(0)$ denotes the permittivity at $r=0^+$. In view of Eq. (5), we can rewrite the radial equation [Eq. (4)] as

$$\frac{d^2R}{dr^2}+\left[\frac{2}{r}+\frac{g}{a}\frac{1}{\varepsilon(0)+gr/a}\right]\frac{dR}{dr}-\frac{n(n+1)R}{r^2}\frac{\varepsilon(0)+hr/a}{\varepsilon(0)+gr/a}=0. \tag{6}$$

We investigate the case of small slopes, namely, $|g/[a\varepsilon(0)]|<1$, and define further a new variable $\hat{r}=gr/[a\varepsilon(0)]$. Consequently, Eq. (6) reduces to

$$\frac{d^2R}{d\hat{r}^2}+\left[\frac{2}{\hat{r}}+\frac{1}{1+\hat{r}}\right]\frac{dR}{d\hat{r}}-\frac{n(n+1)R}{\hat{r}^2}\frac{1+\frac{h}{g}\hat{r}}{1+\hat{r}}=0. \tag{7}$$

On the other hand, we suppose the power series solution as

$$R_n(\hat{r})=\sum_{k=0}^{\infty}C_k^n\hat{r}^{k+\rho}. \tag{8}$$

Accordingly, the second-order derivative of the power series solution is given by

$$\frac{d^2R_n(\hat{r})}{d\hat{r}^2}=\sum_{k=0}^{\infty}(k+\rho)(k+\rho-1)C_k^n\hat{r}^{k+\rho-2}. \tag{9}$$

The substitution of Eq. (9) into Eq. (7) yields

$$\sum_{k=0}^{\infty}C_k^n[(k+\rho)(k+\rho-1)+2(k+\rho)-n(n+1)]\hat{r}^{k+\rho-2}+\sum_{k=0}^{\infty}C_k^n[(k+\rho)(k+\rho-1)+3(k+\rho)-n(n+1)h/g]\hat{r}^{k+\rho-1}=0. \tag{10}$$

The coefficient of each term should vanish, and hence the lowest term satisfies

$$[\rho(\rho+1)-n(n+1)]C_0^n=0. \tag{11}$$

To our interest, Eq. (11) represents a characteristic equation for Eq. (7) (differential equation). Solving it, we obtain $\rho=n$, or $-(n+1)$. Similarly, the recursion relation can also be found from Eq. (10),

$$C_{k+1}^n=-\frac{(k+n)(k+n+2)-n(n+1)h/g}{(k+n+1)(k+n+2)-n(n+1)}C_k^n. \tag{12}$$

As $|g/[a\varepsilon(0)]|<1$ (namely, the normal permittivity is a linear radial function with small slopes), we obtain

$$\lim_{k\to\infty}\left|\frac{C_k^n}{C_k^{n+1}}\right|\to 1. \tag{13}$$

Therefore, within the sphere, the power series solution is convergent indeed.

So far, we can respectively write the potentials in the host medium and the sphere as

$$\begin{aligned}\Phi_m(r,\theta)&=B_0+\sum_{n=0}^{\infty}[B_nr^n+D_nr^{-(n+1)}]P_n(\cos\theta),\\ \Phi_c(r,\theta)&=A_0+\sum_{n=0}^{\infty}A_n\sum_{k=0}^{\infty}C_k^n\left(\frac{gr}{a\varepsilon(0)}\right)^{k+1}P_n(\cos\theta).\end{aligned} \tag{14}$$

As $r\to\infty$, the potential should be given by the far field case, $\Phi_m=-E_0r\cos\theta$. Accordingly, we obtain

$$\begin{aligned}\Phi_m(r,\theta)&=-E_0r\cos\theta+\frac{D_1}{r^2}\cos\theta,\\ \Phi_c(r,\theta)&=A_1\sum_{k=0}^{\infty}C_k^1\left(\frac{gr}{a\varepsilon(0)}\right)^{k+1}\cos\theta.\end{aligned} \tag{15}$$

With the appropriate boundary conditions under consideration, we obtain the coefficients A_1 and D_1, respectively, as

$$\begin{aligned}A_1&=\frac{-3\varepsilon_m}{(\varepsilon(0)+g)\upsilon_2+2\varepsilon_m\upsilon_1}E_0a^3,\\ D_1&=\frac{(\varepsilon(0)+g)\upsilon_2-\varepsilon_m\upsilon_1}{(\varepsilon(0)+g)\upsilon_2+2\varepsilon_m\upsilon_1}E_0a^3,\end{aligned} \tag{16}$$

where

$$\upsilon_1=\sum_{k=0}^{\infty}C_k^1a^2\left(\frac{g}{\varepsilon(0)}\right)^{k+1},$$

$$v_2=\sum_{k=0}^{\infty} C_k^1 a^2(k+1)\left(\frac{g}{\varepsilon(0)}\right)^{k+1}.$$

The local electric field inside the anisotropic graded particle can be derived from the gradient of the corresponding potential Φ_c. As a result, we obtain

$$\mathbf{E}_c=-A_1\sum_{k=0}^{\infty} C_k^1\left(\frac{g}{a\varepsilon(0)}\right)^{k+1} r^k[(k+1)\cos\theta\hat{\mathbf{e}}_r-\sin\theta\hat{\mathbf{e}}_\theta], \tag{17}$$

which is independent of $\hat{\mathbf{e}}_\psi$. Then, the corresponding displacement becomes

$$\mathbf{D}_c=-A_1\sum_{k=0}^{\infty} C_k^1\left(\frac{g}{a\varepsilon(0)}\right)^{k+1} r^k \times[\varepsilon_{rr}(k+1)\cos\theta\hat{\mathbf{e}}_r-\varepsilon_{\theta\theta}\sin\theta\hat{\mathbf{e}}_\theta]. \tag{18}$$

As a result, the displacement along the z axis within the particle is given by

$$D_{cz}=-A_1\sum_{k=0}^{\infty} C_k^1\left(\frac{g}{a\varepsilon(0)}\right)^{k+1} r^k \times[\varepsilon_{rr}(k+1)\cos^2\theta-\varepsilon_{\theta\theta}(\cos^2\theta-1)]. \tag{19}$$

The sum of the polarization within the particle is just the dipole moment of the particle, namely,

$$\int_{\Omega_c}(D_{cz}-\varepsilon_m E_{cz})dV=4\pi\varepsilon_m b a^3 E_0, \tag{20}$$

where b is the dipole factor of the anisotropic graded spherical particle, and Ω_c the volume occupied by the particle. From Eq. (20), we obtain the expression for b,

$$b=\frac{(\varepsilon(0)-\varepsilon_m)v_1+g v_3+2h v_4}{(\varepsilon(0)+g)v_2+2\varepsilon_m v_1}, \tag{21}$$

where

$$v_3=\sum_{k=0}^{\infty} C_k^1 a^2\frac{k+1}{k+4}\left(\frac{g}{\varepsilon(0)}\right)^{k+1},$$

$$v_4=\sum_{k=0}^{\infty} C_k^1 a^2\frac{1}{k+4}\left(\frac{g}{\varepsilon(0)}\right)^{k+1}.$$

When $g=h$ (i.e., isotropic graded spherical particles), the dipole factor b degenerates to[17]

$$b=\frac{(\varepsilon(0)-\varepsilon_m)v_1+g v_5}{(\varepsilon(0)+g)v_2+2\varepsilon_m v_1},$$

where

$$v_5=\sum_{k=0}^{\infty} C_k^1 a^2\frac{k+3}{k+4}\left(\frac{g}{\varepsilon(0)}\right)^{k+1}.$$

III. COMPARISON WITH ANISOTROPIC DIFFERENTIAL EFFECTIVE DIPOLE APPROXIMATION

Alternatively, we present an anisotropic differential effective dipole approximation (ADEDA), which is a numerical method for the analysis of the dielectric property of anisotropic graded particles with *arbitrary* gradation profiles. We may regard the gradation profile as a multishell construction. In details, we establish the dielectric profile gradually by adding shells. Let us start with an infinitesimal isotropic spherical core with permittivity $\varepsilon(0)$, and keep on adding shells with both tangential and normal permittivity profiles $\varepsilon_{\theta\theta}(r)$ and $\varepsilon_{rr}(r)$ at radius r, until $r=a$ is reached. At radius r, we have an inhomogeneous particle, and further regard such an inhomogeneous particle as an effective *homogeneous* one, which has the dipole factor

$$b(r)=\frac{\overline{\varepsilon}(r)-\varepsilon_m}{\overline{\varepsilon}(r)+2\varepsilon_m}, \tag{22}$$

where $\overline{\varepsilon}$ is the effective permittivity of the effective *homogeneous* particle with radius r. Then, we add to the particle a shell with infinitesimal thickness Δr, with permittivies $\varepsilon_{\theta\theta}(r)$ and $\varepsilon_{rr}(r)$. The dipole factor should change according to the dipole factor of one shell anisotropic composite inclusion,[19] that is,

$$\tilde{b}(r+\Delta r)=\frac{(\delta_1(r)\varepsilon_{rr}(r)+\overline{\varepsilon}(r))(\delta(r)\varepsilon_{rr}(r)-\varepsilon_m)+(\overline{\varepsilon}(r)-\delta(r)\varepsilon_{rr}(r))(\delta_1(r)\varepsilon_{rr}(r)+\varepsilon_m)\rho}{(\delta_1(r)\varepsilon_{rr}(r)+\overline{\varepsilon}(r))(\delta(r)\varepsilon_{rr}(r)+2\varepsilon_m)+(\overline{\varepsilon}(r)-\delta(r)\varepsilon_{rr}(r))(\delta_1(r)\varepsilon_{rr}(r)-2\varepsilon_m)\rho}, \tag{23}$$

with $\rho=[(r+\Delta r)/r]^{2\delta(r)+1}$, where $\delta(r)=[-1+(1+8\varepsilon_{\theta\theta}(r)/\varepsilon_{rr}(r))^{1/2}]/2$, and $\delta_1(r)=1+\delta(r)$. Let us write further $\Delta b(r)=b(r+\Delta r)-b(r)$, and take the limit $\Delta r\to 0$, then the desired correction $\Delta b(r)$ is infinitesimal accordingly. Thus, one can obtain a differential equation as

$$\frac{db(r)}{dr}=\frac{1}{3r\varepsilon_{rr}(r)\varepsilon_m}[(1-b(r))^2\varepsilon_{rr}r^2\delta(r)(1+\delta(r)) -(1+b(r)-2b(r)^2)\varepsilon_{rr}(r)\varepsilon_m -(1+2b(r))^2\varepsilon_m^2], \tag{24}$$

where $0<r<a$. Therefore, the dipole factor of an anisotropic graded spherical particle can be calculated by solving the first-order differential equation [Eq. (24)]. This differential equation can be integrated, at least numerically, as long as the gradation profiles $[\varepsilon_{\theta\theta}(r)$ and $\varepsilon_{rr}(r)]$ and the initial condition $[b(0)]$ are given. Note, as mentioned earlier, the ADEDA [Eq. (24)] is valid for arbitrary gradation profiles.

Since we obtained the exact expression [Eq. (21)] for the dipole factor of anisotropic graded spherical particles, we are able to investigate the dielectric property of these particles. In Fig. 1, the dipole factor (b) is plotted as a function of the

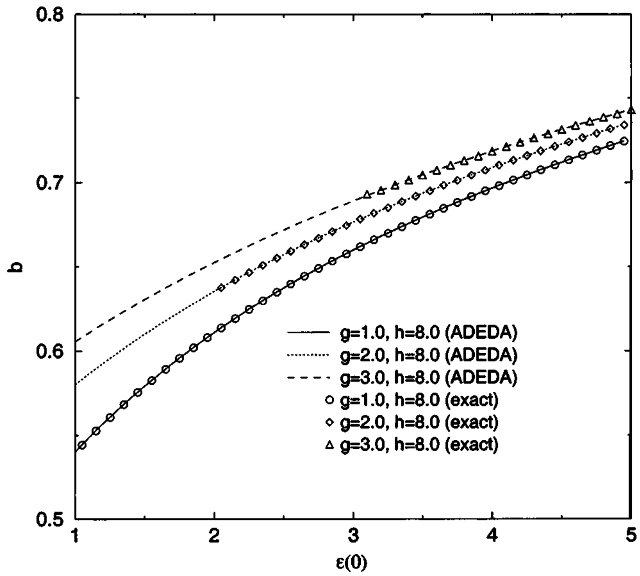

FIG. 1. The dipole factor b as a function of $\varepsilon(0)$ (permittivity of the spherical core with radius $r=0^+$), for the linear gradation profile at various slopes g and h. (a) Exact results predicted by the first-principles approach (symbol); (b) results obtained from the ADEDA (line). Note that the exact results are available only for $|g/[a\varepsilon(0)]|<1$.

initial value $[\varepsilon(0)]$ for various slopes (g and h). For the numerical calculations, we set $h>g$. That is, we assume the radial permittivity is smaller than the tangential permittivity, which is a physical assumption in realistic particles with local anisotropy due to the existence of gradation. In Fig. 1 (symbols), it is shown that b increases monotonically as the initial value $\varepsilon(0)$ increases. Meanwhile, increasing slope g leads to increasing b as well.

It is instructive to compare the first-principles approach with the ADEDA. We evaluate the ADEDA by considering linear gradation profiles [Eq. (5)] too. The numerical integration has been done by the fourth-order Runge–Kutta algorithm with step size 0.01, which guarantees accurate numerics. As shown in Fig. 1, the excellent agreement between the two methods is shown indeed. Thus, we would say that the ADEDA is a very good approximation for anisotropic graded spherical particles. The fact that ADEDA shows very good agreement with the first-principles approach is encouraging as ADEDA allows us to treat arbitrary graded profiles in realistic problems, such as optical properties of anisotropic graded materials, as well as electrokinetic behaviors of biological cells.

IV. DISCUSSION AND CONCLUSION

Our theory may be applied to optical properties of spherical particles of anisotropic graded materials, e.g., by discussing their surface plasma resonance effect. Preliminary results showed that a large figure of merit is achievable in the high-frequency region, where the optical absorption is quite small, by tuning the gradation profiles.

In addition, it is of particular interest to use the present theory to investigate the electrokinetic behaviors like electrorotation and dielectrophoresis of biological cells. Preliminary results showed that the presence of gradation inside the particles can lower the characteristic frequency, at which the electrorotation velocity reaches maximum.

In summary, we present a first-principles approach to calculate the dipole moment of anisotropic graded particles, in the presence of an external electric field. In fact, besides the above-used linear gradation profile, the power-law profile can also be applied to predict exact results.[17] For these gradation profiles, ADEDA is able to show excellent agreement, and ADEDA is valid for arbitrary gradation profiles.

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