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Multipole polarizability of a graded spherical particle

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Abstract. We have studied the multipole polarizability of a graded spherical particle in a nonuniform electric field, in which the conductivity can vary radially inside the particle. The main objective of this work is to access the effects of multipole interactions at small interparticle separations, which can be important in non-dilute suspensions of functionally graded materials. The nonuniform electric field arises either from that applied on the particle or from the local field of all other particles. We developed a differential effective multipole moment approximation (DEMMA) to compute the multipole moment of a graded spherical particle in a nonuniform external field. Moreover, we compare the DEMMA results with the exact results of the power-law graded profile and the agreement is excellent. The extension to anisotropic DEMMA will be studied in an Appendix.

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1 Introduction

In functionally graded materials (FGM), the materials or microstructure properties may vary spatially to meet the specific needs in various engineering applications [1]. The material or microstructure gradients in FGM lead to properties quite distinct from those of the homogeneous materials and conventional composite materials [1,2]. A unique advantage of using FGM is that one can tailor the materials properties via the design of the gradients. Over the past few years, there have been overwhelming attempts, both theoretical and experimental, of studying the responses of FGM to mechanical, thermal, and electric loads and for different microstructure in various systems [1–8]. Such FGM may exist in Nature or they can be made artificially. For instance, graded morphogen profiles can exist in a cell layer [4]. Graded structure may be produced by using various approaches, such as a three-dimensional X-ray microscopy technique [7], deformation under large sliding loads [8], and adsorbate-substrate atomic exchange during growth [6]. Moreover, it has been reported recently that the control of a compatibility factor can facilitate the engineering of FGM [5].

There have been various theoretical attempts to treat the composite materials of homogeneous inclusions [9] as well as multi-shell inclusions [10–13]. These established theories for homogeneous inclusions, however, cannot be applied to graded inclusions directly. To this end, we have recently developed a first-principles approach for calculating the effective response of dilute composites of graded cylindrical inclusions [14] as well as graded spherical particles [15]. More recently, the spectral representation of graded composites has also been established [16]. These theories essentially assumed that the particles are sufficiently far apart so that it is possible to neglect contributions from higher-order multipoles and these approaches are only valid for dilute composites. As the particles become closer, the local field is extremely inhomogeneous on the surface of the particles. To this end, the electromagnetic response of a system of homogeneous (non-graded) spherical particles has been studied extensively in the literature [17,18]. In this work, we aim at studying the multipole polarizability of a graded spherical particle in a nonuniform field which occurs naturally for a system of interacting particles. The electrostatic boundary-value problem of a graded spherical particle will be solved to obtain exact analytic results for the power-law graded profile. For arbitrary graded profiles, we will develop

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a differential effective multipole moment approximation (DEMMA) to compute the multipole moment of a graded spherical particle to capture the multipole response of the particle in a nonuniform field.

The plan of the paper is as follows. In Section 2, we will solve the boundary-value problem of a graded spherical particle in a nonuniform electric field. In this way, the exact analytic expression for the multipole polarizability is obtained for the case of a power-law conductivity profile. In Section 3, we derive the DEMMA for the multipole polarizability of a graded spherical particle of an arbitrary graded profile. In Section 4, we compare the DEMMA results with the exact results of first-principles approach. Discussion of our approaches will be given in Section 5. The generalization to anisotropic DEMMA will be discussed.

2 First-principles approach

We consider a graded spherical inclusion of radius a subjected to a nonuniform electric field of a point charge placed on the z-axis. For electrical conductivity, the constitutive relations read $\boldsymbol{J} = \sigma_i(r)\boldsymbol{E}$ and $\boldsymbol{J} = \sigma_m\boldsymbol{E}$ respectively in the graded spherical inclusion and the host medium, where $\sigma_i(r)$ is the conductivity profile of the graded spherical inclusion and σ_m is the conductivity of the host medium. Moreover,

$$\boldsymbol{\nabla}\cdot\boldsymbol{J} = 0, \qquad \boldsymbol{\nabla}\times\boldsymbol{E} = 0.$$

To this end, $\boldsymbol{E}$ can be written as the gradient of a scalar potential Φ, $\boldsymbol{E} = -\boldsymbol{\nabla}\Phi$, leading to a partial differential equation:

$$\boldsymbol{\nabla}\cdot[\sigma(r)\boldsymbol{\nabla}\Phi] = 0 \tag{1}$$

where $\sigma(r)$ is the dimensionless conductivity profile, while $\sigma(r) = \sigma_i(r)/\sigma_m$ in the inclusion, and $\sigma(r) = 1$ in the host medium. Without loss of generality, we may also let $a = 1$.

In spherical coordinates, the electric potential Φ satisfies

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\sigma(r)\frac{\partial\Phi}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\,\sigma(r)\frac{\partial\Phi}{\partial\theta}\right) + \frac{1}{r^2\sin^2\theta}\frac{\partial}{\partial\varphi}\left(\sigma(r)\frac{\partial\Phi}{\partial\varphi}\right) = 0. \tag{2}$$

We place a point charge of magnitude q on the z-axis at a distance R from the center of the graded sphere. Thus Φ is independent of the azimuthal angle φ. If we write $\Phi = f(r)\Theta(\theta)$ to achieve separation of variables, we obtain two distinct ordinary differential equations. For the radial function $f(r)$,

$$\frac{d}{dr}\left(r^2\sigma(r)\frac{df}{dr}\right) - l(l+1)\sigma(r)f = 0, \tag{3}$$

where l is an integer, while $\Theta(\theta)$ satisfies the Legendre equation [9]. The potential can be obtained by solving equation (3). Exact analytic results can be obtained for a power-law profile [14,15], linear profile [14], and exponential profile [19–21].

Let us consider the case in which the conductivity profile of the particle has a power-law dependence on the radius, $\sigma(r) = cr^k$, with $k \geq 0$ where $0 < r \leq 1$. Then the radial equation becomes

$$\frac{d^2f}{dr^2} + \frac{k+2}{r}\frac{df}{dr} - \frac{l(l+1)f}{r^2} = 0. \tag{4}$$

As equation (4) is a homogeneous equation, it admits a power-law solution [15],

$$f(r) = r^s. \tag{5}$$

Substituting it into equation (4), we obtain the equation $s^2 + s(k+1) - l(l+1) = 0$ and the solution is

$$s^k = \frac{1}{2}\left[-(k+1) \pm \sqrt{(k+1)^2 + 4l(l+1)}\right]. \tag{6}$$

The potentials in the inclusion and the host medium are given, respectively, by

$$\Phi_i(r,\theta) = \sum_{l=0}^{\infty} A_l r^{s_+^k(l)} P_l(\cos\theta),$$
$$\Phi_m(r,\theta) = \frac{q}{r_q} + \sum_{l=0}^{\infty} B_l r^{-(l+1)} P_l(\cos\theta). \tag{7}$$

The positive root of equation (6) has been chosen for nonsingular solution inside the inclusion. Meanwhile, the potential functions should satisfy the boundary conditions, as follow:

$$\Phi_i(r,\theta)\,|_{r=1} = \Phi_m(r,\theta)\,|_{r=1}\,,$$
$$\sigma(r)\frac{\partial\Phi_i(r,\theta)}{\partial r}\,|_{r=1} = \frac{\partial\Phi_m(r,\theta)}{\partial r}\,|_{r=1}\,. \tag{8}$$

The potential due to a point charge located at a distance R from the center of the particle can be rewritten in a multipole expansion in the vicinity of the particle surface [9]:

$$\frac{q}{r_q} = q\sum_{l=0}^{\infty}\frac{r^l}{R^{l+1}}P_l(\cos\theta). \tag{9}$$

From equation (8), we obtain a set of simultaneous linear equations

$$A_l = \frac{q}{R^{l+1}} + B_l, \qquad A_l = \frac{l\sigma_m}{\sigma_i(1)s_+^k(l)}\left(\frac{q}{R^{l+1}} - \frac{l+1}{l}B_l\right).$$

Solving the above equations, we obtain the coefficients:

$$A_l = \frac{q}{R^{l+1}}\frac{2l+1}{l(F_l+1)+1},$$
$$B_l = -\frac{q}{R^{l+1}}\frac{l(F_l-1)}{l(F_l+1)+1}, \quad l \geq 1, \tag{10}$$

$$H_l' = \frac{(1-\sigma_m/\sigma_i(r))\sigma_i(r)(l'\sigma_i(r)+l\bar{\sigma}_i(r))+\rho\sigma_i(r)[l'(\bar{\sigma}_i(r)-\sigma_i(r))+l\sigma_m(\bar{\sigma}_i(r)/\sigma_i(r)-1)]}{[1+l'\sigma_m/(l\sigma_i(r))]\sigma_i(r)(l'\sigma_i(r)+l\bar{\sigma}_i(r))+l'\rho\sigma_i(r)(\sigma_m-\sigma_i(r)+\bar{\sigma}_i(r)-\sigma_m\bar{\sigma}_i(r)/\sigma_i(r))} \quad (14)$$

where

$$F_l = \frac{\sigma_i(1)}{\sigma_m}\frac{s_+^k(l)}{l}, \quad l \geq 1. \quad (11)$$

The coefficients B_l are just proportional to the multipole response of the graded spherical particle to the applied multipole field. We thus identify the multipole factor

$$H_l = \frac{l(F_l-1)}{l(F_l+1)+1}, \quad l \geq 1. \quad (12)$$

From equations (11) and (12), the quantity F_l can be identified as the l-dependent equivalent conductivity of the graded particle (see Eq. (16) in Sect. 3 below). In the case of a homogeneous sphere, $k=0$, $s_+(l)=l$, $F_l=\sigma_i/\sigma_m$, one recovers the well known result [17]:

$$H_l = \frac{l(\sigma_i-\sigma_m)}{l(\sigma_i+\sigma_m)+\sigma_m}, \quad l \geq 1.$$

For a uniform field, however, the dipole factor of Dong et al. [15] recovers.

3 Differential effective multipole moment approximation

In this section, we develop the differential effective multipole moment approximation (DEMMA) for a graded spherical particle and hence compute the multipole factor. To establish the differential effective multipole moment theory, we mimic the graded profile by a multi-shell construction [22], i.e., we build up the conductivity profile gradually by adding shells. We start with an infinitesimal spherical core of conductivity $\sigma_i(0^+)$ and keep on adding spherical shells of conductivity given by $\sigma_i(r)$ at radius r, until $r=a$ is reached. At radius r, we have an inhomogeneous sphere with certain multipole moment. We further replace the inhomogeneous sphere by a homogeneous sphere of the same multipole moment and the graded profile is replaced by an effective conductivity $\bar{\sigma}_i(r)$. Thus,

$$H_l(r) = \frac{l(\bar{\sigma}_i(r)-\sigma_m)}{l(\bar{\sigma}_i(r)+\sigma_m)+\sigma_m}. \quad (13)$$

Next, we add to the sphere a spherical shell of infinitesimal thickness dr, of conductivity $\sigma_i(r)$. The resulting multipole factor H_l' will change according to [18]

see equation (14) above.

with $l'=l+1$ and $\rho=[r/(r+\mathrm{d}r)]^{2l+1}$.

Of course, the equivalent conductivity $\bar{\sigma}_i(r)$, being related to $H_l(r)$, should also change by the same token. Let us write $H_l'=H_l+\mathrm{d}H_l$, and take the limit d$r\to 0$, we obtain a differential equation:

$$\begin{aligned}\frac{\mathrm{d}H_l(r)}{\mathrm{d}r} = &-\frac{1}{(2l+1)r\sigma_m\sigma(r)}\\ &\times[(H_l(r)+l+H_l(r)l)\sigma_m+(H_l(r)-1)l\sigma(r)]\\ &\times[(H_l(r)+l+H_l(r)l)\sigma_m\\ &-(H_l(r)-1)(l+1)\sigma(r)]. \end{aligned} \quad (15)$$

Thus the multipole factor of a graded spherical particle can be calculated by solving the above differential equation with a given graded profile $\sigma_i(r)$. Solving $H_l(r)$ from equation (15) gives numerical results for the multipole factor. The nonlinear first-order differential equation can be integrated if we are given the graded profile $\sigma_i(r)$ and the initial condition $H_l(r=0)$. In general, it works for arbitrary graded profiles $\sigma_i(r)$.

The substitution of the relation equations (13) into (15) yields the differential equation for the equivalent conductivity $\bar{\sigma}_i(r)$,

$$\frac{\mathrm{d}\bar{\sigma}_i(r)}{\mathrm{d}r} = \frac{[\sigma_i(r)-\bar{\sigma}_i(r)][(l+1)\sigma_i(r)+l\bar{\sigma}_i(r)]}{r\sigma_i(r)}. \quad (16)$$

The $l=1$ limit of equation (16) is a special case of the Tartar formula, derived for assemblages of spheres with varying radial and tangential conductivities [23].

4 Numerical results

Figure 1 displays the comparison between the DEMMA (Eq. (15)) and the first-principles approach (Eq. (12)), for the model power-law graded profiles $\sigma_i(r)=cr^k$. Here H_l is plotted as a function of c, for different k. As c increases, the multipole factor H_l is caused to increase monotonically, for different multipole order l. The effect of l can also increase the H_l slightly. Nevertheless, increasing k causes the H_l to decrease. Interestingly, the excellent agreement between the two methods have been shown in this figure. In fact, the DEMMA is valid for arbitrary gradation profiles, as mentioned above. However, exact analytic results obtained from the first-principles approach often lack except for a few specific graded profiles like the power-law profiles. In view of the good comparison shown in this figure, one can safely use the DEMMA to treat other graded profiles which might be not possible, or difficult, to be solved by using the first-principles approach. To this end, the good agreement between DEMMA results and the exact results can be understood by the fact that equation (11) indeed solves equation (16) for the power-law profile. Moreover, the approximate DEMMA approach has been shown to be exact for spherical particles [24].

In Figure 2, we show the equivalent conductivity $\bar{\sigma}_i(r=a)$ of a graded spherical particle, according to the DEMMA (Eq. (16)), for the same model graded profile

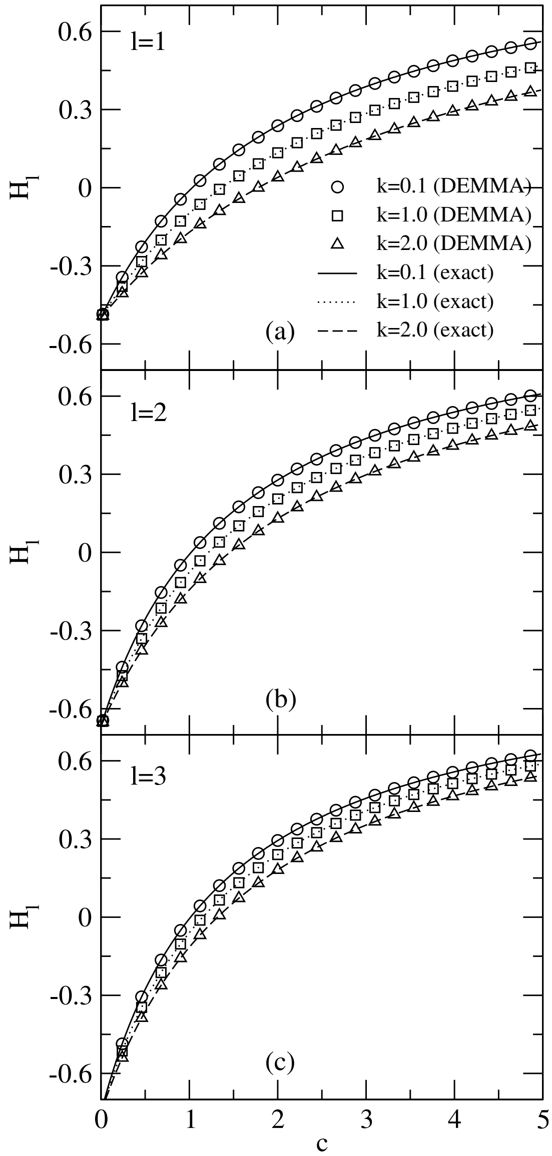

Fig. 1. Comparison of multiple factor H_l, between the DEMMA (Eq. (15)) and the first-principles approach (Eq. (12)). The graded profile is $\sigma_i(r) = cr^k$ and $\sigma_m = 1$. Excellent agreement between the two methods have been shown.

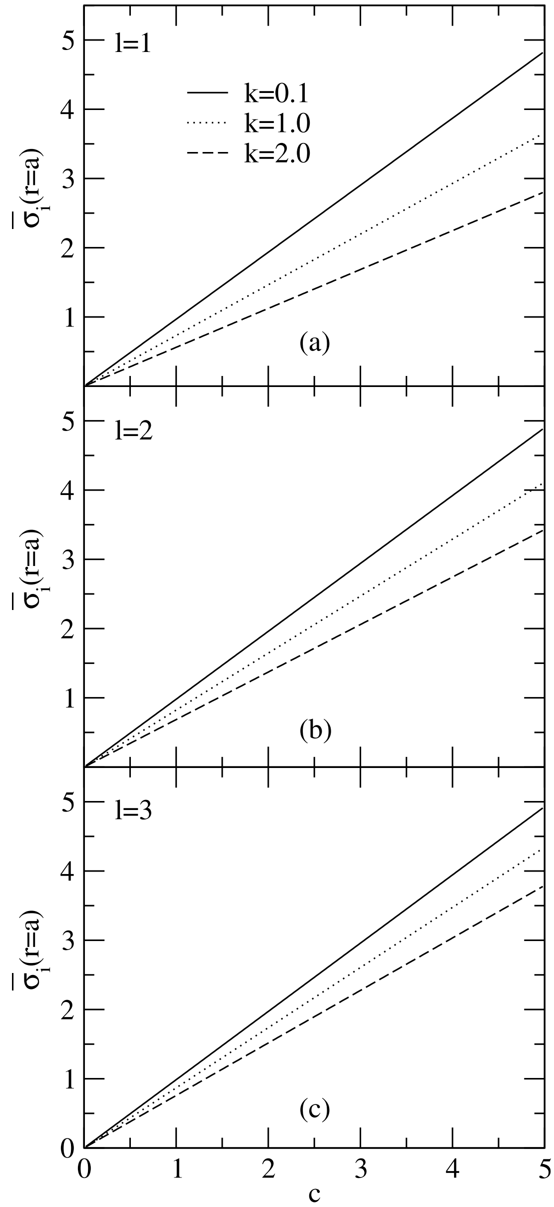

Fig. 2. The equivalent conductivity $\bar{\sigma}_i(r = a)$ of a graded spherical particle, according to the DEMMA (Eq. (16)). Here a denotes the radius of the particle, which is set to unity throughout the paper. The graded profile is $\sigma_i(r) = cr^k$.

$\sigma_i(r) = cr^k$. Equation (16) shows that $\bar{\sigma}_i(r = a)$ depends only on the multipole order l and the conductivity gradation profile $\sigma_i(r)$. Here a denotes the radius of the particle, which has been set to be unity throughout the paper. Similarly, increasing c or l causes the equivalent conductivity to increase. However, opposite behavior is obtained for increasing k.

In fact, the parameter k in the graded profile $\sigma_i(r) = cr^k$ measures the degree of inhomogeneity in the graded particle. From Figures 1 and 2, it is concluded that the presence of inhomogeneity in the particle can affect the electrical properties of the particles significantly. In other words, once an inhomogeneous particle is used in reality, its effect of inhomogeneity should be taken into account.

5 Discussion and conclusion

Here a few comments are in order. In this work, we have developed a differential effective multipole moment approximation (DEMMA) to compute the multipole moment of a graded spherical particle. We compared the DEMMA results with the exact results of the power-law profile and the agreement is excellent. Note that an exact solution is very few in composite research and to have one yields much insight. Such solutions should be useful as benchmarks.

We are now in a position to propose some applications of the present theory. As the multipole response is

$$H_l' = \frac{(\sigma_i{}^{\|}(r)u_- - \bar{\sigma}_i(r)l)(\sigma_i{}^{\|}(r)u_+ - \sigma_m l) - \rho_l(\sigma_i{}^{\|}(r)u_+ - \bar{\sigma}_i(r)l)(\sigma_i{}^{\|}(r)u_- - \sigma_m l)}{(\bar{\sigma}_i(r)l - \sigma_i{}^{\|}(r)u_+)(\sigma_i{}^{\|}(r)u_- + \sigma_m l')\rho_l - (\bar{\sigma}_i(r)l - \sigma_i{}^{\|}(r)u_-)(\sigma_i{}^{\|}(r)u_+ + \sigma_m l')}, \quad (A.3)$$

sensitive to the graded profile of the particles as well as to the structure of the nonuniform field source, there is a potential application to ac electrokinetics of graded colloidal particles [25]. The similar approach can be applied to electrorheological (ER) fluids [26,27] because the particles in ER fluids can have very dense structures locally. The local electric fields are extremely inhomogeneous near the particles so that multipole effects can play an important role. In this regard, we can also study the interparticle force between graded particles [28] in ER fluids.

In the other topics, we may attempt the similar calculation of the multipole response of a graded metallic sphere in the nonuniform field of an oscillating point dipole at optical frequency. The graded Drude dielectric function can be adopted [29]. When the oscillating source is placed close enough to the graded metallic sphere, higher-order multipole response can be excited. The approach may also be applied to the electroencephalogram of the human brain by regarding the brain as a graded anisotropic conducting sphere [30]. In this way, the forward problem can be analyzed. The derivation of the anisotropic DEMMA will be given in Appendix A.

In summary, we have studied the multipole response of a graded spherical particle in a nonuniform field. We developed a DEMMA to compute the multipole moment of a graded spherical particle. Moreover, we compare the DEMMA results with the exact results of the power-law profile and the agreement is excellent.

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Appendix A: Anisotropic differential effective multipole moment approximation

Let us consider a graded spherical particle with radius a. We adopt the spherical coordinates for convenience. The graded spherical particle has a tangential conductivity in the plane orthogonal to the radial vector of the sphere $\sigma_i^{\perp}(r)$ and a radial conductivity $\sigma_i{}^{\|}(r)$. Both $\sigma_i^{\perp}(r)$ and $\sigma_i{}^{\|}(r)$ will be prescribed by radial functions, $0 < r \leq a$. In view of the symmetry, the anisotropic conductivity of the graded sphere can be expressed as a tensor form $\overleftrightarrow{\sigma}_i\ (r)$, namely,

$$\overleftrightarrow{\sigma}_i\ (r) = \sigma_i{}^{\|}(r)\hat{r}\hat{r} + \sigma_i{}^{\perp}(r)\hat{\theta}\hat{\theta} + \sigma_i{}^{\perp}(r)\hat{\varphi}\hat{\varphi}. \quad (A.1)$$

Next, we present an anisotropic differential effective multipole moment approximation (ADEMMA), which, similar to DEMMA, is a numerical method for the analysis of the electric property of anisotropic graded particles with arbitrary gradation profiles. Similarly, we may regard the gradation profile as a multishell construction. In detail, we establish the electric profile gradually by adding shells. Let us start with an infinitesimal isotropic spherical core with conductivity $\sigma_i(0^+)$, and keep on adding shells with both tangential and normal conductivity profiles $\sigma_i^{\perp}(r)$ and $\sigma_i{}^{\|}(r)$ 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 with an effective conductivity $\bar{\sigma}_i(r)$, which has the multipole factor

$$H_l(r) = \frac{l(\bar{\sigma}_i(r) - \sigma_m)}{l(\bar{\sigma}_i(r) + \sigma_m) + \sigma_m}. \quad (A.2)$$

Then, we add to the particle a shell with infinitesimal thickness dr, with conductivities $\sigma_i{}^{\perp}(r)$ and $\sigma_i{}^{\|}(r)$. Its multipole factor H_l' should change according to the multipole factor of a single-coated particle [31]

See equation (A.3) above.

with $u_{\pm} = [-1 \pm \sqrt{1 + 4l(1+l)\sigma_i^{\perp}(r)/\sigma_i{}^{\|}(r)}]/2$, $\rho_l = [r/(r + \mathrm{d}r)]^{u_+ - u_-}$, and $l' = 1 + l$. Let us write further $H_l' = H_l + \mathrm{d}H_l$, and take the limit d$r \to 0$, we obtain a differential equation

$$\frac{\mathrm{d}H_l(r)}{\mathrm{d}r} = -\frac{1}{(1+2l)r\sigma_m\sigma_i{}^{\|}(r)}[l\sigma_m - u_-\sigma_i{}^{\|}(r) + H_l(r)(l'\sigma_m + u_-\sigma_i{}^{\|}(r))] [l\sigma_m - u_+\sigma_i{}^{\|}(r) + H_l(r)(l'\sigma_m + u_+\sigma_i{}^{\|}(r))]. \quad (A.4)$$

Thus the multipole factor of an anisotropic graded spherical particle $H_l(r = a)$ can be calculated by solving the nonlinear first-order differential equation (Eq. (A.4)) which can be integrated, at least numerically if we are given the graded profiles $\sigma_i^{\perp}(r)$ and $\sigma_i{}^{\|}(r)$ and the initial condition $H_l(r = 0)$. The substitution of equation (A.2) into equation (A.4) yields the differential equation for the equivalent conductivity

$$\frac{\mathrm{d}\bar{\sigma}_i(r)}{\mathrm{d}r} = \frac{(1+l)\sigma_i{}^{\|}(r)\sigma_i{}^{\perp}(r) - \sigma_i{}^{\|}(r)\bar{\sigma}_i(r) - l\bar{\sigma}_i(r)^2}{r\sigma_i{}^{\|}(r)}. \quad (A.5)$$

Equations (A.4) and (A.5) can respectively reduce to equations (19) and (20), as long as there is $\sigma_i^{\perp}(r) = \sigma_i{}^{\|}(r) = \sigma_i(r)$. The substitution of $l = 1$ into equation (A.5) reduces to the Tartar formula, derived for assemblages of spheres with varying radial and tangential conductivities [23]. Last but not least, it is also instructive to extend the first-principles approach of referernce [20] to multipole response and compare with the ADEMMA results.

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