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Effective dielectric response of graded composite materials containing anisotropic particles*

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The effective dielectric response of granular composites, in which spheroidal particles with graded shells are randomly distributed in a host matrix, is investigated. General expressions for the effective dielectric constant of the composites and partial resonant condition are obtained in the dilute limit by use of a quasi-static approximation. In particular, spheroidal particles with a power-law gradation profile in the shells are studied in detail. We find that, by adjusting the dielectric gradient profile in the shells, the shape and structure of particles, it is possible to enhance the effective dielectric constant of the composite and to realize partial resonance. Under the partial resonant conditions, the coated spheroidal particles with graded shells within the host matrix can be regarded as equivalent homogeneous spheroids embedded in the same host. The equivalent spheroids have the same dielectric constant as the original cores and semiaxes equal to those of the original shells: i.e., the partial resonant system behaves as if the cores of the particles were enlarged and the shells were absent.

Keywords: effective dielectric response, partial resonance, graded composite materials, geometric anisotropy

PACC: 7720, 7730, 7785

1. Introduction

The granular composites and suspensions have attracted much attention due to various applications.[1−14] In most granular composites and suspensions, there may exist inhomogeneous compartmentalization inside particles, or interfaces between the particles and the host matrix may not be very sharp and smooth due to the diffusion and surface roughness.[4−6] The most common models, which are used for studying the dielectric response of these granular composites, are shell-models.[6−11] Actually, the inhomogeneities mentioned above can be studied in the framework of graded materials. Graded materials with spatial variations in their composition or microstructure are abundant in nature such as biological cells. Due to potential and specific applications in physics and engineering, graded composite materials have attracted much attention and designed and prepared in laboratories in recent years.[15−17] A first-principles approach[18] and a differential effective dipole approximation (DEDA)[19,20] have been developed to investigate the dielectric or thermal response of graded spherical granular composites.

Of course, the effective dielectric response is sensitive to the microstructure of the composites. Geometric anisotropy is clearly a common phenomenon in most granular composites or suspensions, e.g. red blood cells are oblate spheroid in shape. Besides, the spheroidal granular systems can be considered as convenient model systems to study a variety of physical phenomena.[21,22] It should be also noted that the introduction of the shape of particles may be considered as one of ways to describe the interactions inside a composite.

In addition, partially resonant composites are of interest.[9−14,23,24] Based on the definition given by Nicorovici *et al*, a partial resonant system means a

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compound system, containing a component with a given volume fraction (f), whose properties are equivalent to those of a system with that component having a larger volume fraction.[9] Thus, the partial resonance can give rise to an enhancement or extension of the properties of enlarged component materials such as optical properties.[9−14] In hollow-particle composites, the partial resonance can contribute to the increase in porosity, which has potential applications in microelectronic packing.[25,26] In Ref.[23], Nicorovici *et al* demonstrated that the partial resonant model could predict the features in the transmission, reflectance and absorption spectra by comparing the results of model with experimental measurements on Ag–MgF_2-void cermets. The partial resonant structures are important for all-angle efficiency enhancement in light emitting diodes and photodetectors.[24] Partial resonant conditions for coated spherical and cylindrical particles have been studied.[9−13] Under the partial resonant conditions, a three-component granular composite has exactly the same effective dielectric response as a two-component composite composed of core material embedded in the same host as if the shells were absent. This occurs when the core dielectric constant $\varepsilon_{\rm c}$ and the shell dielectric constant $\varepsilon_{\rm s}$ satisfy the condition $\varepsilon_{\rm c} + (d-1)\varepsilon_{\rm s} = 0$, where $d = 2$ or 3 for cylindrical or spherical granular composites respectively. For the condition to be satisfied, one of their dielectric constants must be negative. In general, metals have a negative dielectric constant when the frequency of electric field is less than the plasma resonance frequency. For example, $\varepsilon_{\rm copper} = -48.05$ at $\lambda = 1.0\mu$m;[27] $\varepsilon_{\rm silver} = -1$ at $\lambda = 0.338\mu$m.[23] Recently, left-handed materials, which have a negative dielectric constant and negative permeability simultaneously, have received much attention due to their unique electromagnetic properties and potential applications.[28−30] Smith and co-workers experimentally reported that an array of conducting nonmagnetic split ring resonators and continuous thin wires had both a effective negative dielectric constant and a negative permeability.[29]

In this work, we investigate the effective dielectric response of graded spheroidal granular composites, in which particles have homogeneous cores and graded shells, and also investigate the partial resonant condition of the spheroidal particles whose polarized directions are parallel to the symmetric principal axes.

2. Formalism

Let us consider a dielectric composite system, in which spheroidal particles are randomly embedded in a host matrix, subjected to a uniform electric field. For describing the shape of spheroids, depolarization factors L_z and $L_{xy}(L_x = L_y \equiv L_{xy})$ are introduced, which are positive and satisfy the sum rule $L_z + 2L_{xy} = 1$.[31,32] For spherical particles, $L_z = L_{xy} = 1/3$. In general, L_z is strongly dependent on the shape of the spheroidal particles and is given by[32,33]
$L_z = \dfrac{1}{1-m^2} + \dfrac{m}{(m^2-1)^{3/2}} \ln(m + \sqrt{m^2-1})$ for prolate spheroid with the aspect ratio $m = \dfrac{c}{a} > 1$, while $L_z = \dfrac{1}{1-m^2} + \dfrac{m}{(1-m^2)^{3/2}} \arccos m$ for oblate spheroid with $m = \dfrac{c}{a} < 1$, where $a(= b), c$ are the semi-axes of a spheroid along the Cartesian axes. With a given aspect ratio m, one can uniquely determine a depolarization factor L_z. Thus we use L_z to characterize the shape of the spheroidal particles. For simplicity, we assume that the spheroidal particles possess an isotropic dielectric constant but with a uniaxial direction (along z-direction). We consider a composite of the suspended spheroidal particles with a low concentration and ignore the interaction among the particles.

2.1. Composites containing spheroidal particles with homogeneous shells

At first, we consider a homogeneous spheroidal particle system, in which spheroidal particles with a dielectric constant $\varepsilon_{\rm c}$ satisfy the equation $\dfrac{x^2}{a_{\rm c}^2} + \dfrac{y^2}{a_{\rm c}^2} + \dfrac{z^2}{c_{\rm c}^2} = 1$ and are embedded in the host matrix with a dielectric constant $\varepsilon_{\rm m}$. The system is subjected to a uniform electric field $\boldsymbol{E}_0$ along z-direction. In a quasi-static approximation, the induced dipole moment $\boldsymbol{p}$ along z-direction of each spheroid is given by solving the Laplace's equation and boundary conditions in ellipsoidal coordinate[31,32]

$$\boldsymbol{p} = 4\pi\varepsilon_{\rm m} b_z a_{\rm c}^2 c_{\rm c} \boldsymbol{E}_0,$$

where

$$b_z = \frac{1}{3} \frac{\varepsilon_{\rm c} - \varepsilon_{\rm m}}{\varepsilon_{\rm m} + L_z(\varepsilon_{\rm c} - \varepsilon_{\rm m})}. \quad (1)$$

It is dipole factor, which measures the degree of polarization for the particle in an external field. Next, we add a spheroidal shell with the dielectric constant $\varepsilon_{\rm s}$ to the spheroidal particle, and the overall semiaxes of the coated spheroid are $a_{\rm s}$, $a_{\rm s}$, $c_{\rm s}$. The shape of the

spheroidal shell is the same as that of the core, i.e. the shell and the core of the particle have the same depolarization factor. The dipole factor of the coated spheroid is given by[20,31,34]

$$b_{1z} = \frac{1}{3}\frac{(\varepsilon_s - \varepsilon_m) + 3x_1\lambda_1[\varepsilon_s + L_z(\varepsilon_m - \varepsilon_s)]}{[\varepsilon_m + L_z(\varepsilon_s - \varepsilon_m)] + 3x_1\lambda_1 L_z\,(1 - L_z)\,(\varepsilon_s - \varepsilon_m)}, \tag{2}$$

with $x_1 = \frac{1}{3}\frac{\varepsilon_c - \varepsilon_s}{\varepsilon_s + L_z(\varepsilon_c - \varepsilon_s)}$,

where structural parameter $\lambda_1 = \frac{a_c^2 c_c}{a_s^2 c_s}$, which represents the ratio of the core volume to the whole coated spheroid volume. It is easy to check that b_{1z} (Eq.(2)) reduces to b_z (Eq.(1)) when $\varepsilon_c = \varepsilon_s$. Thus the dipole factor remains unchanged if one adds a spheroidal shell with the same dielectric constant.

The dipole factor of the coated spheroid (Eq.(2)) can also be expressed by

$$b_{1z} = \frac{1}{3}\frac{\mu_1\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_z(\mu_1\varepsilon_c - \varepsilon_m)}, \tag{3}$$

where

$$\mu_1 = \frac{\gamma_0\{(1 - L_z)(1 - \lambda_1)\gamma_0 + [L_z + (1 - L_z)\lambda_1]\}}{\gamma_0[(1 - L_z) + \lambda_1 L_z] + L_z(1 - \lambda_1)}, \tag{4}$$

with dielectric factor $\gamma_0 = \frac{\varepsilon_s}{\varepsilon_c}$. It can be a complex constant.

By comparing the dipole factor (Eq.(3)) of the coated spheroid with that of simple homogeneous spheroid (Eq.(1)), we can find that these two expressions are similar in form. The only difference is that ε_c becomes $\mu_1\varepsilon_c$. Therefore, we can assume that the single-shell spheroidal particles within the host can be regarded as equivalent homogeneous spheroidal particles embedded in the same host. The equivalent homogeneous particles have the dielectric constant $\mu_1\varepsilon_c$ and semiaxes a_s, a_s, c_s equal to overall semiaxes of original coated spheroids. Here the equivalent homogeneous spheroids induce the same dipole moment as the original coated spheroids. The factor μ_1 is called the equivalent coefficient and determined by the dielectric factor γ_0, the structure parameter λ_1 and depolarization factor L_z.

We choose the electric field to be parallel to the z axis; however, this axis has no special property to distinguish it from the other principle axes. Therefore, the dipole factor along $x(y)$-direction $b_{1x(y)}$ and correspondingly equivalent coefficient μ_1', when the electric field is parallel to the x or y axis, respectively, are

$$b_{1x(y)} = \frac{1}{3}\frac{\mu_1'\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_{x(y)}(\mu_1'\varepsilon_c - \varepsilon_m)},$$

$$\mu_1' = \frac{\gamma_0\{(1 - L_{xy})(1 - \lambda_1)\gamma_0 + [L_{xy} + (1 - L_{xy})\lambda_1]\}}{\gamma_0[(1 - L_{xy}) + \lambda_1 L_{xy}] + L_{xy}(1 - \lambda_1)}.$$

The effective dielectric constant in the Maxwell–Garnett approximation (MGA) for a randomly oriented spheroidal particle system in the dilute limit is given by[33,35]

$$\varepsilon_{\text{eff}} = \varepsilon_m\left\{1 + \frac{f}{3}\left[\frac{\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_z(\varepsilon_c - \varepsilon_m)} + 2\frac{\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_{xy}(\varepsilon_c - \varepsilon_m)}\right]\right\}, \tag{5}$$

where ε_c and ε_m are the dielectric constants of the spheroids and the host matrix respectively, f represents the volume fraction of spheroidal particles, and ε_{eff} is the effective dielectric constant of the whole system. Now substituting the dielectric constants of these equivalent homogenous spheroids $\mu_1\varepsilon_c$ and $\mu_1'\varepsilon_c$ for ε_c in the second term and third term in Eq.(5) respectively, and substituting the volume fraction of these equivalent spheroids f'/λ_1 for f, we can obtain an expression for the effective dielectric constant of the randomly oriented coated spheroidal particle system in a dilute limit as follows:

$$\varepsilon_{\text{eff}} = \varepsilon_m\left\{1 + \frac{f'/\lambda_1}{3}\left[\frac{\mu_1\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_z(\mu_1\varepsilon_c - \varepsilon_m)} + 2\frac{\mu_1'\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_{xy}(\mu_1'\varepsilon_c - \varepsilon_m)}\right]\right\} \tag{6}$$

where f' is the volume fraction of the cores of the particles.

For a spheroidal particle, whose polarized direction is parallel to the symmetric principal axis, when μ_1=1, then $b_{1z} = b_z$, which means that the dipole factor of the coated spheroid is the same as that of the equivalent homogeneous spheroid composed of the core material. Namely, the coated spheroid induces the same dipole moment as that of the equivalent homogeneous spheroid, which has the same dielectric constant ε_c as that of the original core and semiaxes equal to those of the original shell. In this case, partial

resonance occurs and partial resonant condition of the coated spheroidal particle is obtained as

$$\gamma_0 = -\frac{L_z}{1-L_z},$$

i.e.

$$L_z\varepsilon_c + (1-L_z)\varepsilon_s = 0. \tag{7}$$

For a spherical (or cylindrical) particle, $L_z = \frac{1}{3}$ (or $L_z = \frac{1}{2}$), the condition can be simplified as $\varepsilon_c + 2\varepsilon_s = 0$ (or $\varepsilon_c + \varepsilon_s = 0$), which is the same as the well-known result in Refs.[9–11]. In Fig.1, we plot the dielectric constant of the core (ε_c) versus that of the shell (ε_s) for various depolarization factors L_z when partial resonant condition is satisfied. It is obvious that the relation between ε_c and ε_s is linear, and one dielectric constant is opposite to another dielectric constant in sign. In general, if the core of the spheroidal particle is metallic (nonmetallic), its shell must be oppositely nonmetallic (metallic). It is also shown that the ratio of ε_c to ε_s increases as the depolarization factor L_z decreases. When $L_z \to 0$ (needle spheroids), ε_c is much higher than ε_s; while $L_z \to 1$ (disc spheroids), the case is the contrary. Namely, to realize partial resonance of the needle (disc) spheroid, not only must the dielectric constant of the core be opposite to that of the shell in sign, but also the former must be much larger (smaller) than the latter in magnitude. This is very interesting for the disc spheroidal particles. The dielectric constant of the shells must be much larger than that of the cores in magnitude, but the particles in the system behave as if the shells were absent.

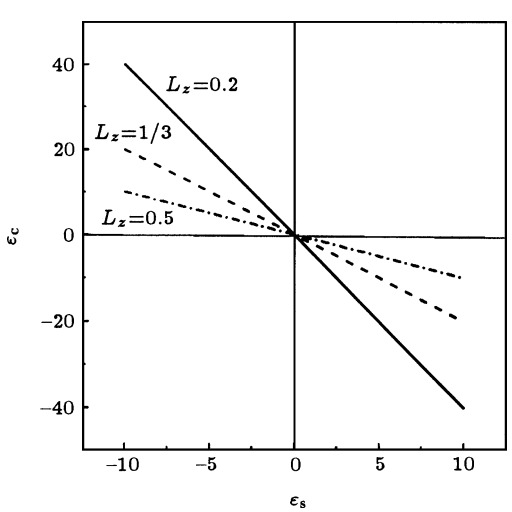


Fig.1. Dielectric constant of the core (ε_c) versus dielectric constant of the shell (ε_s) for various depolarization factors L_z under the partial resonant condition.

The above approach can be extended to the particles coated by two shells with different dielectric constants. If each spheroidal particle has two shells with dielectric constants ε_{s1} and ε_{s2} and semiaxes a_{s1}, a_{s1}, c_{s1}; a_{s2}, a_{s2}, c_{s2} respectively, we can also obtain the dipole factor by solving the Laplace's equation and boundary conditions,

$$b_{2z} = \frac{1}{3}\frac{\mu_2\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_z(\mu_2\varepsilon_c - \varepsilon_m)}, \tag{8}$$

where the equivalent coefficient μ_2 is

$$\mu_2 = \frac{\gamma_2 G - \gamma_2(1-L_z)H}{G + L_z H} \tag{9}$$

with

$$\begin{aligned} G =& \lambda_1\{[L_z + (1-L_z)\gamma_1][L_z\gamma_1 + (1-L_z)\gamma_2] \\ &+ \lambda_1 L_z(1-L_z)(-\gamma_1+\gamma_2)(-1+\gamma_1)\}, \\ H =& \lambda_2\{(-\gamma_1+\gamma_2)[L_z + (1-L_z)\gamma_1] \\ &+ \lambda_1(-1+\gamma_1)[(1-L_z)\gamma_1 + L_z\gamma_2]\}, \end{aligned} \tag{10}$$

and

$$\gamma_2 = \frac{\varepsilon_{s2}}{\varepsilon_c}, \quad \lambda_2 = \frac{a_c^2 c_c}{a_{s2}^2 c_{s2}}; \quad \gamma_1 = \frac{\varepsilon_{s1}}{\varepsilon_c}, \quad \lambda_1 = \frac{a_c^2 c_c}{a_{s1}^2 c_{s1}}.$$

It is easy to check that μ_2 reduces to μ_1 when $\varepsilon_{s2} = \varepsilon_{s1}$ or $\varepsilon_{s1} = \varepsilon_c$. When μ_2=1, the partial resonant condition of the coated spheroidal particles with double shells, whose polarized directions are parallel to the symmetric principal axes, can also be obtained.

2.2. Composites containing spheroidal particles with graded shells

We consider a graded spheroidal particle system. Each spheroidal particle can be parametrized by $x^2 + y^2 + \frac{z^2}{m^2} = r^2$, and the dielectric profile for the graded particle is given by $\varepsilon(r)$ where $a_c \le r \le a_s$, m is the aspect ratio, a_c and a_s are the lengths of the semi principle axes along x-(y) axis of the core and the shell. We mimic the graded profile by a multi-shell construction, i.e. we build up the dielectric profile by adding shells gradually. We start with a spheroidal core with the dielectric constant ε_c and semiaxis a_c, and keep on adding spheroidal shells with a dielectric constant given by $\varepsilon(r)$ at r, until $r = a_s$ is reached. At r, we have an inhomogeneous spheroid whose equivalent coefficient is given by $\mu(r)$. To make the exact expression of $\mu(r)$, we further replace the inhomogeneous spheriodal shell by a homogeneous spheriodal shell, and the graded dielectric profile is replaced by

an effective dielectric constant $\bar{\varepsilon}(r)$. Thus it is similar to the spheriodal particle with a single homogeneous shell. According to Eq.(4), we obtain $\mu(r)$ as

$$\mu(r) = \frac{\gamma\{(1-L_z)(1-\lambda)\gamma + [L_z + (1-L_z)\lambda]\}}{\gamma[(1-L_z) + \lambda L_z] + L_z(1-\lambda)}, \tag{11}$$

where

$$\gamma = \frac{\bar{\varepsilon}(r)}{\varepsilon_c} \quad \text{and} \quad \lambda = \frac{a_c^2 c_c}{mr^3} = \left(\frac{a_c}{r}\right)^3 (c_c = ma_c).$$

Next, we add to the spheroid another spheroidal shell with an infinitesimal thickness dr and the dielectric constant $\varepsilon(r)$. The increment in equivalent coefficient due to an infinitesimal addition is $d\mu$. Hence the equivalent coefficient can be written as $\mu + \mathrm{d}\mu$. Since the particle in the case is equivalent to double-shell construction, the equivalent coefficient should be changed according to Eqs.(9) and (10), correspondingly $\gamma_2 = \varepsilon(r)/\varepsilon_c$, $\lambda_2 = \left(\frac{a_c}{r+\mathrm{d}r}\right)^3$, $\gamma_1 = \gamma = \frac{\bar{\varepsilon}(r)}{\varepsilon_c}$, and $\lambda_1 = \lambda = \left(\frac{a_c}{r}\right)^3$. Of course, the effective dielectric constant $\bar{\varepsilon}(r)$, related to $\mu(r)$, should be also changed by the same mode. Let us write $\mu_2 = \mu + \mathrm{d}\mu$ and take the limit $\mathrm{d}r \to 0$ and keep to the first order in dr, we obtain a differential equation for the equivalent coefficient $\mu(r)$ as

$$\frac{\mathrm{d}\mu}{\mathrm{d}r} = \frac{3[\varepsilon(r) - \varepsilon_c\mu][(1-L_z)\varepsilon(r) + L_z\varepsilon_c\mu]}{\varepsilon_c\varepsilon(r)r}. \tag{12}$$

For spherical particles $\left(L_z = \frac{1}{3}\right)$, the equation reduces to

$$\frac{\mathrm{d}\mu}{\mathrm{d}r} = \frac{[\varepsilon(r) - \varepsilon_c\mu][2\varepsilon(r) + \varepsilon_c\mu]}{\varepsilon_c\varepsilon(r)r}. \tag{13}$$

Note that Eq.(13) is just the Tartar formula[36] and it is in agreement with the results in Refs.[13,19].

Therefore, the equivalent coefficient of the coated spheroidal particles with graded shells embedded in a host matrix can be calculated by solving the differential equation (12) with a given gradient profile $\varepsilon(r)$. The nonlinear first-order differential equation can be integrated at least numerically, if the graded profile $\varepsilon(r)$ and initial condition $\mu(r = a_c)$ are given. Once $\mu(r = a_s)$ is calculated, the spheroidal particles with graded shells in the whole system are equivalent to homogeneous spheroidal particles with dielectric constant $\mu\varepsilon_c$ and semiaxes a_s, a_s, c_s. The dipole moment induced by the equivalent homogeneous spheroids is equal to that of original graded spheroidal particles.

Similarly, when the electric field is parallel to $x(y)$ axis, the equivalent coefficient μ_1' can be also obtained. The expression is similar to Eq.(12) as long as L_z is replaced by L_{xy}. Therefore the effective dielectric constant of the graded spheroidal particle system can be also obtained as long as μ_1, μ_1' and λ_1 are replaced by $\mu(a_s)$, $\mu'(a_s)$ and $\left(\frac{a_c}{a_s}\right)^3$ respectively in Eq.(6).

When $\mu(a_s) = 1$, the partial resonance of spheroid particles whose polarized directions are parallel to the symmetric principal axes occurs. Similarly the spheroidal particles with graded shells in a host matrix can be regarded as equivalent homogenous spheroids with the dielectric constant ε_c and semiaxis a_s, a_s, c_s embedded in the same host. Namely, the property of the cores of the particles will extend to the shells as if the graded shells were absent.

2.3. Power-law dielectric gradation profile in the shells

We consider that the dielectric constant of the shell of spheroidal particles has a power-law profile $\varepsilon(r) = \varepsilon_s\left(\frac{r}{a_s}\right)^n (a_c < r < a_s)$. Obviously, ε_s is the dielectric constant of interfaces between particles and the host matrix. By solving the differential equation (12), the equivalent coefficient of the coated graded spheroidal particle system is obtained as follows:

$$\mu = \frac{k_1(3-6L_z+n)(q^s-1) + k_1 s(1+q^s) + 6(1-L_z)(1-q^s)q^n}{k_1[6L_z k_1(1-q^s) + (3-6L_z+n)(1-q^s)q^n + s(1+q^s)q^n]}, \tag{14}$$

with $s = \sqrt{9 + n(6-12L_z+n)}$, $q = \frac{a_c}{a_s}$ and $k_1 = \frac{\varepsilon_c}{\varepsilon_s}$.

Similarly, μ_1' can be also expressed as Eq.(14) as long as L_z is replaced by L_{xy}. According to Eq.(6), the effective dielectric constant of the randomly oriented graded spheroidal particle system in the dilute limit is obtained as follows:

$$\varepsilon_{\mathrm{eff}} = \varepsilon_m\left\{1 + \frac{f/\lambda}{3}\left[\frac{\mu\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_z(\mu\varepsilon_c - \varepsilon_m)} + 2\frac{\mu'\varepsilon_c - \varepsilon_m}{\varepsilon_m + L_{xy}(\mu'\varepsilon_c - \varepsilon_m)}\right]\right\}, \tag{15}$$

where f is the volume fraction of the cores and $\lambda =$

$\left(\frac{a_c}{a_s}\right)^3$. Namely, f/λ is the volume fraction of the whole particles. It shows that the dielectric gradation profile, the structure and shape of particles have great influences on the effective dielectric constant of the composite system.

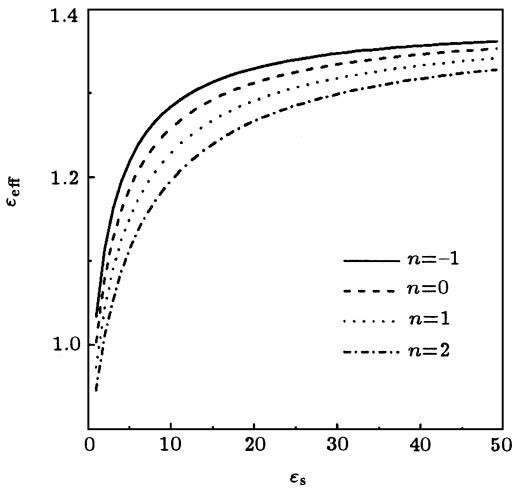


Fig.2. Effective dielectric constant $\varepsilon_{\rm eff}$ against $\varepsilon_{\rm s}$ for $n = -1$, $n = 0$, $n = 1$, and $n = 2$. Parameters: $\varepsilon_{\rm m} = 1$, $\varepsilon_{\rm c} = 5$, $f/\lambda = 0.1$, $q = 0.3$, and $L_z = 0.6$.

In Fig.2, we plot the effective dielectric constant $\varepsilon_{\rm eff}$ against $\varepsilon_{\rm s}$ for $n = -1$, $n = 0$, $n = 1$, and $n = 2$ when the structure and shape of particles are fixed. It is clear that $\varepsilon_{\rm eff}$ increases monotonically with $\varepsilon_{\rm s}$, while it decreases as the index n increases. It is attributed to the fact that average dielectric constant in the shell $\bar{\varepsilon}$ increases with $\varepsilon_{\rm s}$ and decreases with n. In Fig.3, the dependence of $\varepsilon_{\rm eff}$ on the structure and shape of particles is investigated. We plot $\varepsilon_{\rm eff}$ versus core–shell semiaxis ratio and depolarization factor when the dielectric gradation profile is given ($n = 1$ and $\varepsilon_{\rm s}$=10). From Fig.3(a), we see that $\varepsilon_{\rm eff}$ decreases with L_z firstly for prolate spheroids $\left(L_z < \frac{1}{3}\right)$ and reaches a minimum for spherical particles $\left(L_z = \frac{1}{3}\right)$ and it then increases with the further increase of L_z. Fig.3 (b) indicates that $\varepsilon_{\rm eff}$ increases smoothly as the core–shell semiaxis ratio (q) increases in a region of $q < 0.5$ but decreases dramatically with q in a region of $q > 0.5$. At $q = 0.5$, $\varepsilon_{\rm eff}$ reaches a maximum. Therefore by suitably adjusting the dielectric gradient profile in shells, the structure and shape of particles, it is possible to enhance the effective dielectric constant of the graded granular composite.

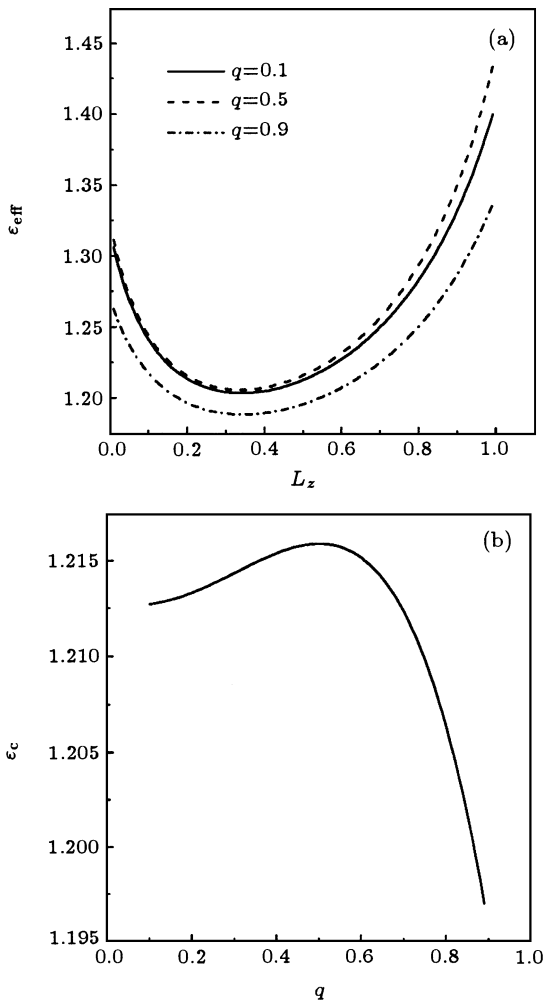


Fig.3. Dependence of $\varepsilon_{\rm eff}$ on structure parameter q and shape factor L_z. Parameters: $\varepsilon_{\rm m} = 1$, $\varepsilon_{\rm c} = 5$, $f/\lambda = 0.1$, $n = 1$ and $\varepsilon_{\rm s} = 10$. (a) Dependence of $\varepsilon_{\rm eff}$ on shape factor L_z for selected q : $q = 0.1$, $q = 0.5$ and $q = 0.9$. (b) Dependence of $\varepsilon_{\rm eff}$ on structure parameter q when $L_z = 0.5$.

If we take $\mu = 1$, the partial resonant condition of the coated graded spheroidal particles whose polarized direction is along the z-axis is obtained as follows:

$$\varepsilon_{\rm c} + k\varepsilon_{\rm s} = 0, \tag{16}$$

where k is related to n, L_z, and q, and $k = A + B$ with

$$A = \frac{(3 - 6L_z + n)(1 + q^n)(1 - q^s) - s(1 - q^n)(1 + q^s)}{12L_z(1 - q^s)},$$

$$B = \frac{\sqrt{144(1-L_z)q^n + \left[(3-6L_z+n)(1+q^n) + s(1-q^n)\dfrac{1+q^s}{1-q^s}\right]^2}}{12L_z}.$$

Note that if $n \to 0$, then $\varepsilon(r) \to \varepsilon_s$, the particles are equivalent to the coated particles with homogeneous shells, and Eq.(16) can be reduced to $L_z\varepsilon_c + (1-L_z)\varepsilon_s = 0$, which is the same as Eq.(7).

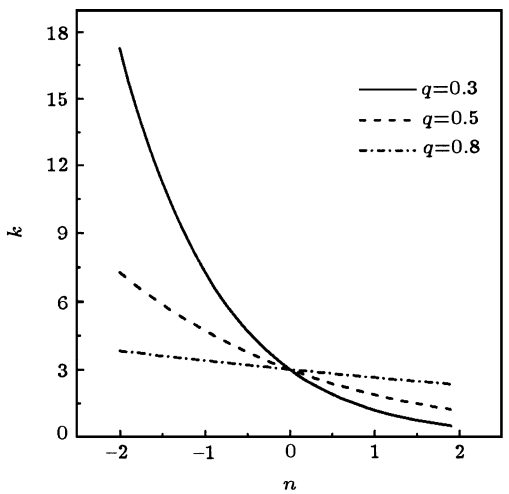


Fig.4. Dependence of coefficient k on n for $q = 0.3$, $q = 0.5$ and $q = 0.8$.

In Fig.4, we plot the coefficient k versus n for various core–shell semiaxis ratios q when the depolarization factor L_z is given. It indicates that coefficient k decreases dramatically as n increases when the core–shell semiaxis ratio q is small, while k decreases smoothly with n when q takes a large value. In other words, k is more dependent on n in the case of the thick shells than in the case of the thin shells. If the thickness of the shells closes to zero, i.e. $q \to 1$, k would be independent of n. It can be easily understood by the fact that the dielectric gradient profile in the shells plays a more important role when the shells are thicker. We also find that k decreases as q increases when $n < 0$, but it increases with q when $n > 0$. It is attributed to the fact that the average dielectric constant of the graded shells decreases as q increases when $n < 0$ and increases with q when $n > 0$. In Fig.5, we also plot the coefficient k against depolarization factor L_z, it is obvious that k decreases as L_z increases. When $L_z \to 0$ (needle spheroids), the value of k is large, while $L_z \to 1$ (disc spheroids), k is small. These indicate that to satisfy the partial resonant condition, the dielectric constant of the cores (ε_c) must be much higher than dielectric constant of the interfaces (ε_s) for needle spheroidal particles. But it is another story for the disc spheroidal particles. Figures 4, 5 also show that k is always positive. Thus we can conclude that to satisfy the partially resonant condition $\varepsilon_c + k\varepsilon_s = 0$, at the least the dielectric constant of the cores must be opposite to the dielectric constant of the shells of the particles in sign. That is to say, if the cores of the spheroidal particles are metallic (nonmetallic), the shells must be oppositely nonmetallic (metallic). These results are similar to those for spheroidal particles with homogeneous shells.

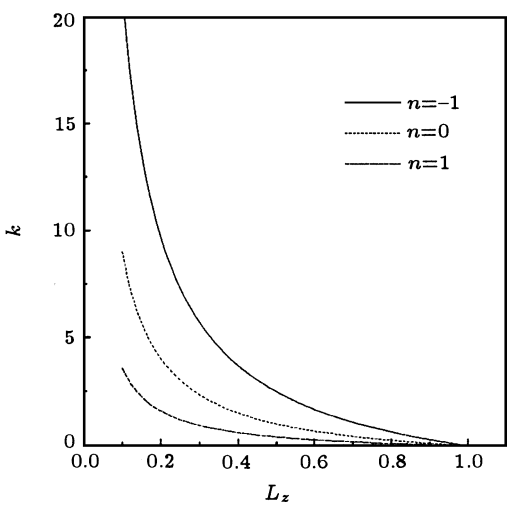


Fig.5. Dependence of coefficient k on shape factor L_z for $n = -1$, $n = 0$, and $n = 1$.

As a result, by suitably adjusting the core–shell semiaxis ratio q, dielectric gradient profile (ε_s, n), and depolarization factor L_z, it is feasible to realize the partial resonance of graded geometric anisotropic particles.

3. Conclusion

In summary, we have investigated the effective dielectric response of graded composites composed of geometric anisotropic particles in a quasi-static approximation. We develop the general expressions for the effective dielectric constant of the composite containing randomly oriented spheroidal particles with arbitrary graded shells in a dilute limit. Freyria *et al*[17] observed a graded cell response when they studied the

cell-implant interactions experimentally. The dielectric constant may change rapidly near the boundary of the cells. Thus we present and further study in detail a power-law gradation profile of the dielectric constant in the shells of particles. It is possible to enhance effective dielectric response by suitably adjusting the physical and geometrical parameters such as the dielectric gradient profile in the shells, the core–shell semiaxis ratio and depolarization factor of particles.

The partial resonance of graded spheroidal particles can be realized as long as the partial resonant condition is satisfied. Partial resonant condition depends on the geometric structure and physical properties of the particles. For the power-law dielectric gradient profile, partial resonant condition can be satisfied by suitably adjusting the geometric structure, the shape and gradient profile parameters of particles. Regardless of the geometric structure and shape of particles, there exists a common condition that the dielectric constant of the core must be opposite in sign to the dielectric constant of the shell. It indicates that, if the core of the spheroidal particle is metallic (nonmetallic), the shell must be oppositely nonmetallic (metallic).

The present formalism of the partial resonant condition is valid only for the particles whose polarized directions are parallel to the symmetric principal axes. When the direction of polarization of spheroidal particles is randomly oriented, the partial resonant condition will become more complicated. Work is in progress in this direction.

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