Dielectric responses of anisotropic graded granular composites having arbitrary inclusion shapes

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The transformation field method (TFM) originated from Eshelby's transformation field theory is developed to estimate the effective permittivity of an anisotropic graded granular composite having inclusions of arbitrary shape and arbitrary anisotropic grading profile. The complicated boundary-value problem of the anisotropic graded composite is solved by introducing an appropriate transformation field within the whole composite region. As an example, the effective dielectric response for an anisotropic graded composite with inclusions having arbitrary geometrical shape and arbitrary grading profile is formulated. The validity of TFM is tested by comparing our results with the exact solution of an isotropic graded composite having inclusions with a power-law dielectric grading profile and good agreement is achieved in the dilute limit. Furthermore, it is found that the inclusion shape and the parameters of the grading profile can have profound effect on the effective permittivity at high concentrations of the inclusions. It is pointed out that TFM used in this paper can be further extended to investigate the effective elastic, thermal, and electroelastic properties of anisotropic graded granular composite materials.

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I. INTRODUCTION

Graded granular composites have attracted much attention since they have advantages over nongraded composites in the design of engineering materials for specific needs.[1,2] In particular, because the bulk effective responses of graded granular composites can be controlled by varying the physical and microstructural properties or the constituent materials, we have found many applications in material science, such as electric, thermal, and mechanical materials.[3–8] Recently, a lot of methods were developed to predict the effective properties of graded granular composites. For instance, in the dilute limit, Gu and Yu[9] exactly derived the effective conductivities of the graded cylindrical composites having power-law, linear or exponential grading profiles on the inclusions. Based on hypergeometrical functions, Wei *et al.*[10,11] studied the effective ac permittivity of graded spherical colloidal suspensions and the effective dc permittivity of graded nonlinear cylindrical composites having inclusions with a power-law dielectric grading profile in the dilute limit. Yu *et al.*[12] developed a method called differential effective dipole approximation (DEDA) for the calculation of the effective dielectric responses of graded composites having spherical inclusions with radially graded dielectric profile. Later, DEDA was shown to be exact for spherical and cylindrical particles.[10,13] The DEDA method was also used by Dong *et al.*[14,15] and Huang *et al.*[16] to estimate the properties of linear and nonlinear graded composites. Recently, Duan *et al.*[17] proposed a differential replacement procedure to estimate the effective properties of anisotropic graded spherical composites on the basis of energy equivalency condition. Furthermore, Mejdoubi and Brosseau[18] used the finite-element method to discuss the controllable effective complex permittivity of functionally graded composites. However, in general, there are few attempts to find the effective responses of graded granular composites with complex structures, probably due to complicated boundary conditions, especially for anisotropic graded composites having arbitrary inclusion shape. Because the physical properties of the graded granular composites depend on the constitutive relations, physical properties and microstructures of their constituent materials in a complicated way, it is difficult to find a unified theoretical method for the estimation of their effective responses, although there are strong needs due to their potentially important applications in the engineering field. In this study, we aim at the development of a theoretical method to deal with the problem of estimating the effective responses of anisotropic graded granular composite materials.

In order to circumvent the difficulties caused by different inclusion shapes, a concept of transformation field in inclusion region proposed by Eshelby[19] to study the elastic field of infinite composites were further developed by Nemat-Nasser and Taya[20] to investigate the bulk effective elastic moduli of composites having periodic voids (periodic composites), where the transformation field will not be a constant in periodic composites. Later, the periodic composites were extensively studied by Fourier's transformation method, such as Tao and co-workers[21,22] and Shen *et al.*[23] had used Fourier method to investigate the electric conductivities and elastic moduli of periodic composites. Bergman and Dunn[24] had proposed an integral equation in Fourier space to estimate the bulk effective properties of periodic composites. Along this line, transformation field method (TFM) was significantly extended by Gu and co-workers[25–28] to calculate the effective conductivity, photonic dispersion relation, viscosity, and piezoelectric constants of periodic composites having complex inclusion structures including inclusion fractal geometry.[29,30] In addition, the finite-difference time-domain method can be developed to investigate the dielectric behavior of complex composites having inclusion fractal structures.[31] Recently, TFM was further enhanced to solving the effective response problem of isotropic graded composites having arbitrary inclusion shapes.[32] The aim of the present paper is to further extend the TFM to estimate the bulk effective dielectric response of anisotropic graded periodic granular composites having arbitrary inclusion shape and with grading dielectric profile. The basic idea of the transformation field is to find a field function so that the constitutive relationships of the inclusion and host materials can be expressed by a unified constitutive equation by matching the continuous electric displacement in interfaces of different phases. The local perturbation electric fields originated from the effects of the grading profile and the shape were expressed in terms of an appropriate field, i.e., transformation field. A unified constitutive equation was set up for the field, and a set of algebraic equations were established to determine the unknown transformation field. In this way, the differential equations of an anisotropic graded granular system were transformed to a set of algebraic equations of transformation field having simple integrals. Furthermore, formulas of the bulk effective dielectric responses were given by means of the transformation field.

In Sec. II, TFM will be derived for anisotropic graded composites without the limitation of inclusion shapes and dielectric profiles by introducing transformation field into inclusion region, and a set of algebraic equations is given for determining the transformation fields. In Sec. III, based on the solutions of transformation fields of Sec. II, the effective anisotropic dielectric constants of anisotropic graded composites are formulated. In Sec. IV, the examples of anisotropic graded composites are performed for discussing the effects of inclusion shapes and anisotropic graded profiles on effective anisotropic permittivity. Finally, a brief conclusion is given in Sec. V.

II. TRANSFORMATION FIELD METHOD

Consider the linear anisotropic dielectric constitutive relations of the host and the inclusion materials, respectively,

$$D_k^\alpha = \varepsilon_{kj}^\alpha E_j, \tag{1}$$

where the superscripts $\alpha=h,i$ denote quantities of the host and inclusion granular materials, respectively, and the subscripts $j,k=x,y,z$ represent quantities along x, y, and z directions, respectively. The electric displacement, electric field, and permittivity are denoted by D, E, and ε, respectively. The governing electrostatic equations are $\nabla\cdot D=0$ and $\nabla\times E=0$. Consider an anisotropic graded composite in which anisotropic graded granular inclusions are embedded in an anisotropic nongraded matrix. In this case, the dielectric property of the graded inclusion is a function of the spatial variable $\vec{x}$, namely $\varepsilon_{kj}^i=\varepsilon_{kj}^i(\vec{x})$.

For an anisotropic graded periodic composite in three-dimensional space, if an external uniform electric field E_j^0 is applied to the periodic composite along the j-coordinate direction, a perturbation electric field E will occur in the whole composite region since the graded inclusions will change the local electric fields. Thus, the constitutive relations of the host region Ω_h and the inclusion region Ω_i become $D_k^h=\varepsilon_{kj}^h(E_j^0+E_j)$ and $D_k^i=\varepsilon_{kj}^i(E_j^0+E_j)$, respectively, where E_j ($j=x,y,z$) denote the components of the perturbation electric field. In order to avoid matching the complicated boundary conditions, a transformation field E^* can be defined in the composite region such that its components obey the following relationships in the host and inclusion regions, respectively,

$$E_k^* = 0 \quad \text{in } \Omega_h, \tag{2}$$

$$\varepsilon_{kj}^h(E_j^0+E_j-E_j^*) = \varepsilon_{kj}^i(\vec{x})(E_j^0+E_j) \quad \text{in } \Omega_i. \tag{3}$$

Using Eqs. (2) and (3), a unified constitutive equation is obtained for the whole composite region $\Omega=\Omega_i+\Omega_h$,

$$D_k = \varepsilon_{kj}^h(E_j^0+E_j-E_j^*) \quad \text{in } \Omega. \tag{4}$$

Therefore, the governing equation in region Ω can be rewritten as the following form:

$$\nabla_k[\varepsilon_{kj}^h(E_j^0+E_j-E_j^*)] = 0. \tag{5}$$

For the periodic composite, the electric potential $\Phi(\vec{x})$ of the perturbation electrical field and the transformation field $E_k^*(\vec{x})$ can be expressed in terms of Fourier transformation components because of the periodicity,

$$\Phi(\vec{x}) = \sum_{|n|\neq 0}^{\infty} \Phi(\vec{\xi})\exp(i\vec{\xi}\cdot\vec{x}), \tag{6}$$

$$E_k^*(\vec{x}) = \sum_{|n|\neq 0}^{\infty} E_k^*(\vec{\xi})\exp(i\vec{\xi}\cdot\vec{x}), \tag{7}$$

$$E_k(\vec{x}) = -\nabla_k\Phi(\vec{x}) = -i\sum_{|n|\neq 0}^{\infty} \xi_k\Phi(\vec{\xi})\exp(i\vec{\xi}\cdot\vec{x}), \tag{8}$$

where the reciprocal vectors are $\vec{\xi}=(\xi_x,\xi_y,\xi_z)=2\pi\left(\frac{n_x}{l_x},\frac{n_y}{l_y},\frac{n_z}{l_z}\right)$, and l_x, l_y, and l_z are the lengths of the unit cell along the x, y, and z directions, respectively. The inverse Fourier transformations of the perturbation potential and the transformation field are, respectively,

$$\Phi(\vec{\xi}) = \frac{1}{V}\int_\Omega \Phi(\vec{x})\exp(-i\vec{\xi}\cdot\vec{x})d\vec{x}. \tag{9}$$

$$E_k^*(\vec{\xi}) = \frac{1}{V}\int_\Omega E_k^*(\vec{x})\exp(-i\vec{\xi}\cdot\vec{x})d\vec{x} = \frac{1}{V}\int_{\Omega_i} E_k^*(\vec{x})\exp(-i\vec{\xi}\cdot\vec{x})d\vec{x}. \tag{10}$$

where V denotes the volume of whole composite region. Substituting Eqs. (6)–(8) into Eq. (5), the following relationship between the perturbation electric potential and the transformation field is obtained in Fourier space,

$$\Phi(\vec{\xi}) = i\xi_k\varepsilon_{kj}^h E_j^*(\vec{\xi})/(\varepsilon_{rs}^h\xi_r\xi_s). \tag{11}$$

Substituting Eq. (11) into Eq. (8), the perturbation electric field can be expressed by the transformation field,

$$E_k(\vec{x}) = \sum_{|n|\neq 0}^{\infty} \xi_k \xi_t \varepsilon_{tl}^h E_l^*(\vec{\xi})/(\varepsilon_{rs}^h \xi_r \xi_s)\exp(i\vec{\xi}\cdot\vec{x}). \quad (12)$$

Thus, substituting Eqs. (7), (10), and (12) into Eq. (3), we obtained a set of integral equations for the unknown transformation field $E^*(\vec{x})$ as follows:

$$\varepsilon_{kj}^h E_j^*(\vec{x}) = [\varepsilon_{kj}^h - \varepsilon_{kj}^i(\vec{x})]E_j^0 + V^{-1}[\varepsilon_{kj}^h - \varepsilon_{kj}^i(\vec{x})]\sum_{|n|\neq 0}^{\infty} \xi_j \xi_t \varepsilon_{tl}^h \int_{\Omega_i} E_l^*(\vec{x}') \times \exp[i\vec{\xi}\cdot(\vec{x}-\vec{x}')]d\vec{x}'/(\varepsilon_{rs}^h \xi_r \xi_s), \quad (13)$$

where the subscripts $k=x,y,z$ denote the three integral equations of the unknown transformation field.

In order to solve these equations, the transformation field can be expressed in a series, for example, a power series as follows:

$$E_i^*(\vec{x}) = \sum_{\alpha,\beta,\gamma} C_i^{\alpha\beta\gamma}(x/l_x)^\alpha (y/l_y)^\beta (z/l_z)^\gamma, \quad (14)$$

where $C_i^{\alpha\beta\gamma}$ $(i=x,y,z)$ are the unknown coefficients. Substituting Eq. (14) into Eq. (13) and multiplying both sides of the resulting equations by a term $(x/l_x)^p(y/l_y)^q(z/l_z)^s$, and then integrating these equations over the inclusion region, we get a set of linear algebraic equations of the unknown coefficients $C_l^{\alpha\beta\gamma}$,

$$\varepsilon_{kl}^h \sum_{\alpha,\beta,\gamma} C_l^{\alpha\beta\gamma} A^{p+\alpha,q+\beta,s+\gamma} = (\varepsilon_{kl}^h A^{p,q,s} - \tilde{A}_{kl}^{p,q,s})E_l^0 + V^{-1}\sum_{\alpha,\beta,\gamma} C_l^{\alpha\beta\gamma} T_{kl}^{p,q,s,\alpha,\beta,\gamma} - V^{-1}\sum_{\alpha,\beta,\gamma} C_l^{\alpha\beta\gamma}\tilde{T}_{kl}^{p,q,s,\alpha,\beta,\gamma}, \quad (15)$$

where the superscripts $\alpha,\beta,\gamma=0,1,2,3,\ldots,N$, and N is called the approximation order of the transformation field. $p,q,s=0,1,2,3,\ldots,M$, and M is a parameter of the number of equations, which can be selected so that the equations are closed,

$$T_{kl}^{p,q,s,\alpha,\beta,\gamma} = \sum_{|n|\neq 0}^{\infty} \varepsilon_{kj}^h \xi_j \xi_t \varepsilon_{tl}^h G^{p,q,s}(\vec{\xi}) G^{\alpha,\beta,\gamma}(-\vec{\xi})/(\varepsilon_{uw}^h \xi_u \xi_w),$$

$$\tilde{T}_{kl}^{p,q,s,\alpha,\beta,\gamma} = \sum_{|n|\neq 0}^{\infty} \xi_j \xi_t \varepsilon_{tl}^h \tilde{G}_{kj}^{p,q,s}(\vec{\xi}) G^{\alpha\beta\gamma}(-\vec{\xi})/(\varepsilon_{uw}^h \xi_u \xi_w),$$

$$A^{pqs} = \int_{\Omega_i}\left(\frac{x}{l_x}\right)^p\left(\frac{y}{l_y}\right)^q\left(\frac{z}{l_z}\right)^s d\vec{x},$$

$$\tilde{A}_{kj}^{pqs} = \int_{\Omega_i}\left(\frac{x}{l_x}\right)^p\left(\frac{y}{l_y}\right)^q\left(\frac{z}{l_z}\right)^s \varepsilon_{kj}^i(\vec{x})d\vec{x},$$

$$G^{\alpha\beta\gamma}(\vec{\xi}) = \int_{\Omega_i}\left(\frac{x}{l_x}\right)^\alpha\left(\frac{y}{l_y}\right)^\beta\left(\frac{z}{l_z}\right)^\gamma \exp(i\vec{\xi}\cdot\vec{x})d\vec{x},$$

$$\tilde{G}_{kj}^{\alpha\beta\gamma}(\vec{\xi}) = \int_{\Omega_i}\left(\frac{x}{l_x}\right)^\alpha\left(\frac{y}{l_y}\right)^\beta\left(\frac{z}{l_z}\right)^\gamma \varepsilon_{kj}^i(\vec{x})\exp(i\vec{\xi}\cdot\vec{x})d\vec{x}.$$

Clearly, using Eq. (15), the transformation field can be found for the anisotropic graded granular composite with arbitrary numerical precision. Also, it is noted that the derivation does not rely on the shape of the inclusions, the grading profile, the spatial distribution, and the number of them in the unit cell. Therefore, once the unknown transformation field is obtained from Eq. (15), we can use it to estimate the bulk effective responses of any anisotropic graded composites.

III. EFFECTIVE RESPONSES OF ANISOTROPIC GRADED COMPOSITES

Considering the constitutive relations of an anisotropic linear composite, the effective permittivity ε_{jk}^e under an external electric field E_k^0 is defined as follows:

$$\varepsilon_{jk}^e \langle E_k^0 \rangle = \langle D_j \rangle, \quad (16)$$

where $\langle A\rangle = V^{-1}\int_\Omega A dV$. Note that the volumetric average of the perturbation electric field E_k is zero over the whole composite region due to the periodicity.[25–28] Thus, by taking the volumetric average over the unified constitutive Eq. (4), we have

$$\langle D_j \rangle = \varepsilon_{jk}^h \langle E_k^0 + E_k - E_k^* \rangle. \quad (17)$$

Combining Eqs. (16) and (17), the effective permittivity formula is obtained,

$$\varepsilon_{jk}^e E_k^0 = \varepsilon_{jk}^h [E_k^0 - \langle E_k^*(\vec{x})\rangle_i], \quad (18)$$

where $\langle A\rangle_i \equiv V^{-1}\int_{\Omega_i} A dV$. Because the transformation field [Eq. (15)] includes the interactions between the parameters of inclusion and host materials, such as the graded profile, the shape, and the spatial distribution and the number of the granular inclusions, Eq. (18) can be used to estimate the effective permittivity of composites having high concentrations of the inclusions. For example, if an external electric field E_z^0 is applied to the composite along the z direction, the effective anisotropic responses ε_{jz}^e can be calculated by the formulas, $\varepsilon_{jz}^e = \varepsilon_{jk}^h [E_k^0 - \langle E_k^*(\vec{x})\rangle_i]/E_z^0$ $(j=x,y,z)$. In the following section, the effective dielectric responses of two different anisotropic graded composites are discussed numerically.

IV. NUMERICAL DISCUSSION

In our TFM numerical examples of this section, we have considered the cubic periodic anisotropic graded composites having a unit cube cell in three-dimensional space, where the anisotropic graded inclusions (in our examples, the graded inclusion shapes will be of cubic and spherical) are located at the center of a unit cube cell of anisotropic host materials. The dimensionless length of unit cube cell is fixed at 10 so

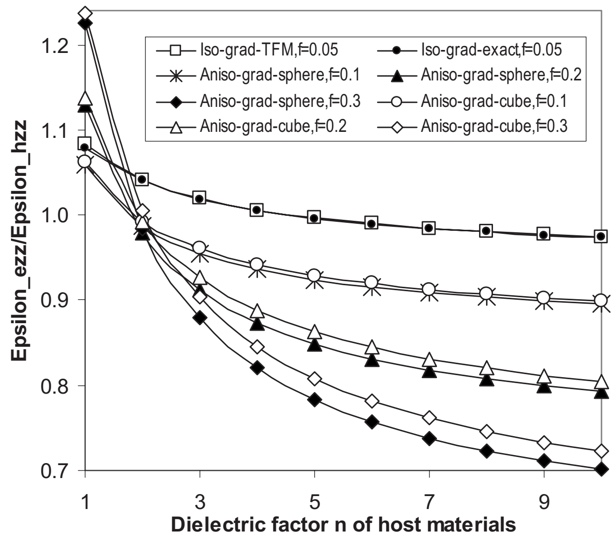

FIG. 1. Effective dielectric constant contrast $\varepsilon_{zz}^{e}/\varepsilon_{zz}^{h}$ versus the dielectric factor n of the host material at inclusion volume fractions $f=0.1$, 0.2, 0.3. The dielectric grading profiles of the anisotropic graded spherical and cubic inclusions are $\varepsilon_{xx}^{i}=|x|^{2}+1$, $\varepsilon_{yy}^{i}=|y|^{2}+2$, and $\varepsilon_{zz}^{i}=|z|^{2}+3$, respectively, and the anisotropic dielectric constants of the host material are $\varepsilon_{xx}^{h}=n$, $\varepsilon_{yy}^{h}=2n$, $\varepsilon_{zz}^{h}=3n$, where n is the dielectric factor of the host material. In this figure, "aniso-grad-sphere" and "aniso-grad-cube" denote the results of two kinds of anisotropic graded composites, namely, having graded anisotropic spherical and cubic inclusions, respectively, by means of a transformation field method. The phases "iso-grad-TFM" and "iso-grad-exact" denote the TFM results and the exact dilute solution in Ref. 33 for an isotropic graded spherical inclusion having dielectric grading profile of the form $\varepsilon^{i}=r^{2}$ embedded in an isotropic host material having isotropic dielectric constant $\varepsilon^{h}=n$, at an inclusion volume faction $f=0.05$.

that the graded permittivity and the volume fraction of inclusions have a large change in inclusion space and unit cube cell, respectively, because of the inclusion graded dielectric responses related to its space. To calculate the transformation fields with Eq. (15), in our numerical examples, the parameter M of the number of equations and the approximation order N of transformation field are fixed at 5, respectively, so that 648 coupled equations of transformation field coefficients are closed.

To test the validity of TFM for the calculation of the effective permittivity of graded composites, we have calculated the effective dielectric response of a cubic periodic composite by TFM, where an isotropic spherical inclusion of a power-law dielectric grading profile $\varepsilon^{i}=cr^{k}$ (where r is the radial variable of the spherical inclusion) is located at the center of the unit cube cell, and then compared the results with that of exact dilute solution of isotropic graded spherical inclusion embedded in an infinite isotropic matrix. Furthermore, we note that the effective response of spherical graded composites having an infinite isotropic matrix can be exactly derived from Ref. 33 for an inclusion power-law grading profile $\varepsilon^{i}=cr^{k}$. From Fig. 1, it is clear that good agreements are obtained in the dilute limit for some illustrative composite systems. In order to discuss the effects due to the volume fraction, the shape and the parameters of the grading profile of the anisotropic graded inclusion on the effective permittivity, we have considered two kinds of anisotropic graded periodic composites, namely, anisotropic graded spherical and cubic inclusions having anisotropic power-law dielectric grading profile embedded in a unit cube of anisotropic host material, respectively. In Fig. 1, it can be seen that the differences between the effective responses of the anisotropic graded cubic and spherical composites increase with increasing inclusion volume fraction for a given power k. The results also show that, at high concentrations, the inclusion shape affects the differences in the effective responses of the graded composite systems. In the dilute limit, as expected, the effects of the inclusion shape are smaller. Naturally, the effective responses will be affected by the parameters of the inclusion grading profile. In Fig. 2, the changes of the effective responses for the graded cubic and spherical composites with the power k are shown at a fixed volume fraction of the inclusions. It clearly shows that the differences in effective responses of the two anisotropic graded composites are enhanced as the value of the power of the grading profile increases. Thus, for the graded composites, the inclusion shape and its grading profile clearly change the effective responses of the graded composite system.

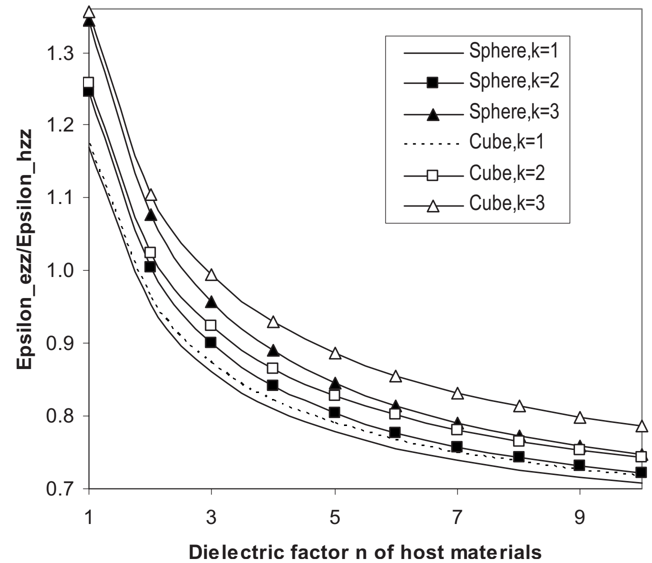

FIG. 2. Effective dielectric constant contrast $\varepsilon_{zz}^{e}/\varepsilon_{zz}^{h}$ versus the dielectric factor n of the host material at inclusion volume fraction $f=0.3$. The dielectric grading profiles of the anisotropic graded spherical and cubic inclusions are $\varepsilon_{xx}^{i}=|x|^{k}+1$, $\varepsilon_{yy}^{i}=|y|^{k}+2$, and $\varepsilon_{zz}^{i}=|z|^{k}+3$, respectively, and the anisotropic dielectric constants of the host material are $\varepsilon_{xx}^{h}=n$, $\varepsilon_{yy}^{h}=2n$, and $\varepsilon_{zz}^{h}=3n$, where n is the dielectric factor of the host material.

Furthermore, we also calculate the effective permittivity of an anisotropic composite system having anisotropic graded spherical inclusions embedded in an isotropic matrix for different volume fractions. Figure 3 indicates that the graded composites having anisotropic spherical inclusions with graded profiles along the x, y, or z directions clearly have anisotropic effective responses. However, it should be pointed out that for graded composites having radial-symmetric graded spherical inclusions embedded in isotropic

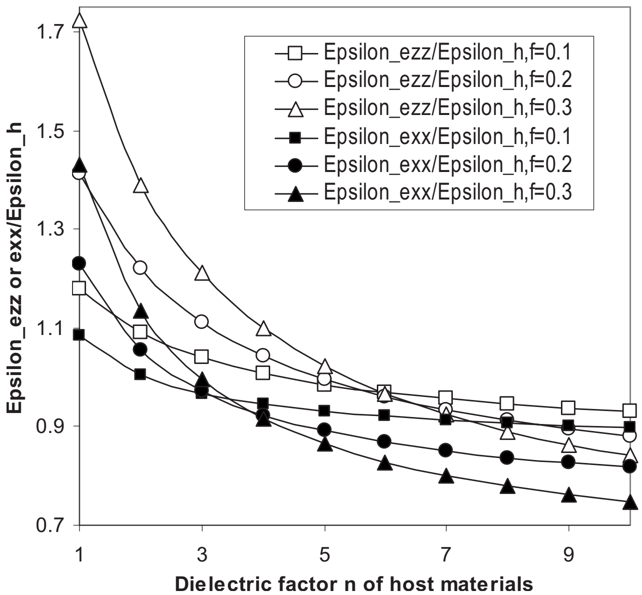

FIG. 3. Effective dielectric constant contrasts $\varepsilon_{zz}^{e}/\varepsilon^{h}$ and $\varepsilon_{xx}^{e}/\varepsilon^{h}$ of graded spherical composites versus dielectric factor n of the host material, where the dielectric grading profiles of the graded spherical inclusions are $\varepsilon_{xx}^{i}=|x|^2+1$, $\varepsilon_{yy}^{i}=|y|^2+2$, and $\varepsilon_{zz}^{i}=|z|^2+3$, respectively, and the dielectric constant of the isotropic host material is $\varepsilon^{h}=n$, where n is the dielectric factor of the host material, and the inclusion volume fractions are $f=0.1$, 0.2, 0.3, respectively.

matrix, in which the inclusion spheres have radial and tangential anisotropic grading profiles with respect to the radial variable, their effective responses are isotropic.[1,34]

V. CONCLUSIONS

For an anisotropic graded granular periodic composite having arbitrary inclusion shape and grading profile, a TFM is developed to investigate its effective permittivity. The complicated boundary-value problem originated from the arbitrary inclusion shape is overcome by introducing a transformation field in the whole composite region. A set of algebraic equations is set up for solving of the unknown transformation field, and formulas of the effective responses are derived. Furthermore, the effects of the volume fraction, the grading profile, and the shape of the granular inclusions on the effective responses are discussed for two different anisotropic graded composite systems (i.e., the graded cubic and graded spherical inclusion composites). At high inclusion concentrations, the inclusion shape is found to have profound effects on the effective responses. The numerical results also show that the inclusion grading profile can enhance the differences in effective responses between different graded composite systems. Also, the inclusion shape and the grading profile are two key factors for tailor-making of the effective bulk responses in the design of functionally graded composite materials. In addition, we note that TFM is applicable to estimate the effective dielectric responses of graded composites having high concentration, complicated inclusion grading profile, and arbitrary inclusion shapes if the order of the transformation field is taken high enough. In summary, we have proposed a method for the estimation of bulk effective responses of anisotropic graded granular periodic composites having arbitrary inclusion shape and grading profile. The method can be further extended to deal with the effective properties of other types of anisotropic graded random composites if we have a large random sample of anisotropic graded composites regarded as a unit cell, for example, graded elastic, graded piezoelectric or graded thermal transport coefficient composites. Moreover, it is also possible to develop TFM to investigate the effective responses of graded composites having nonlinear constitutive relations, by combining with the existing methods dealing with nonlinear composites.[35–38]

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