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The nonlinear effective dielectric response of graded composites

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Abstract
The perturbation method is developed to deal with the effective nonlinear dielectric responses of weakly nonlinear graded composites, which consist of the graded inclusion with a linear dielectric function of spatial variables of inclusion material. For Kerr-like nonlinear graded composites, as an example in two dimensions, we have used the perturbation method to solve the boundary value problems of potentials, and studied the effective responses of nonlinear graded composites, where a cylindrical inclusion with linear dielectric function and nonlinear dielectric constant is randomly embedded in a homogeneous host with linear and nonlinear dielectric constants. For the exponential function and the power-law dielectric profiles of cylindrical inclusions, in the dilute limit, we have derived the formulae of effective nonlinear responses of both graded nonlinear composites.

1. Introduction

Graded composites have attracted much attention because the effective properties of graded composites are of wide application to electric, thermal, optical and mechanical engineering [1–10]. For a graded material, its physical properties vary continuously in space. In nature, this kind of graded material is abundant. On the other hand, graded composites are designed in laboratories by changing their composition or microstructure for the specific needs of an engineering design [6, 8, 10, 11]. For example, the dielectric constant, thermal conductivity and electric conductivity can be designed to vary along the radial direction of a cylindrical or spherical particle. The physical properties of graded composites are more important and more useful than those of homogeneous composites in material sciences. For instance, the mechanical properties of graded composites can be used to improve bonding strength, toughness and wear resistance [7–9]. However, conventional formulae of effective responses in homogeneous composites are not suitable for calculating the effective nonlinear properties of graded composites. Recently, many authors have devoted themselves to the study of effective properties of graded composites, and have proposed several methods. For example, Gu and Yu [12] have studied the effective electric conductivities of linear composites with graded inclusion, and derived the formulae of cylindrical graded composites for the linear and the power-law conductivity profiles exactly by using special functions. Dong *et al* [13] have investigated the dielectric response of graded particles of anisotropic materials by using the first-principles approach and effective dipole approximation method. Gao *et al* [14] have developed the nonlinear differential effective dipole approximation to derive the effective nonlinear response of graded composites. In fact, for weakly nonlinear composites with graded inclusion materials, the perturbation method can be developed to derive the effective response of nonlinear composites. The merit of this method is that the local potentials in graded nonlinear composite regions can be exactly derived by solving a set of linear order differential equations [15]. In this paper we will develop the perturbation method to deal with the effective dielectric responses of nonlinear graded composites with a unidirectional cylindrical inclusion, where the dielectric responses of the linear term of the cylindrical inclusions are exponent dielectric and power-law dielectric profiles, respectively.

Consider the Kerr-like nonlinear constitutive relations

$$D_\alpha = \varepsilon_\alpha E + \chi_\alpha |E|^2 E, \qquad \text{in } \Omega_\alpha \tag{1}$$

where the subscripts $\alpha = \mathrm{i}$ or h. We note that the quantities with subscripts i and h are the physical quantities of inclusion (i) and host (h) regions, respectively. D and E are the electric displacement field and the electric field, respectively. Ω_α ($\alpha = \mathrm{i}$, h) is the region occupied by the α-type material. ε and χ are the linear and nonlinear dielectric responses, respectively. For nonlinear cylindrical composites, the linear dielectric response $\varepsilon_\mathrm{i}(r)$ is a function of the space variable r of the cylindrical region (where r is the radial variable of cylindrical inclusion in cylindrical coordinates). The dielectric response, ε_h, χ_h and χ_i are constants. We assume that the usual electrostatic equations are satisfied: $\nabla \cdot D = 0$ and $\nabla \times E = 0$. The boundary conditions are the continuities of the potential Φ and electric displacement field D on the cylindrical surface, $\partial\Omega_\mathrm{i}$, $\hat{n} \cdot D_\mathrm{i} = \hat{n} \cdot D_\mathrm{h}$ and $\Phi_\mathrm{i} = \Phi_\mathrm{h}$, where $\hat{n}$ is the outward normal vector of inclusion surface. In this paper, for a cylindrical particle with dielectric function profiles, we study the nonlinear response of graded composites with two cases dielectric profiles, $\varepsilon_\mathrm{i}(r) = c\mathrm{e}^{\beta r}$ (c and β are constants) and $\varepsilon_\mathrm{i}(r) = c_k r^k$.

The paper is organized as follows. In next section, we will give the electric displacement field and potential equations of Kerr-like nonlinear composites by using the perturbation method. For both cases of cylindrical dielectric functions $\varepsilon_\mathrm{i}(r) = c\mathrm{e}^{\beta r}$ and $\varepsilon_\mathrm{i}(r) = c_k r^k$, we derive the zero order and the first order potentials of cylindrical nonlinear graded composites, respectively. In section 3, in the dilute limit, the effective nonlinear responses of the two types graded composites are derived. Furthermore we demonstrate that our results can be exactly reduced to the formulae of nonlinear homogeneous composites when we let $\beta \to 0$ and $k \to 0$, respectively. In section 4, some conclusions are given.

2. Electrostatic potentials of a graded composite

In order to solve equation (1), the perturbation method is applied to solve weakly nonlinear composites, $|\chi|E|^2/\varepsilon| \ll 1$. The nonlinear coefficient of the host material χ_h is chosen as the perturbation parameter. For the potential, $\Phi_\alpha(r, \varphi)$, we have

$$\Phi_\alpha(r, \varphi) = \sum_{k=0}^{\infty} \chi_\mathrm{h}^k \Phi_\alpha^k(r, \varphi) \qquad \text{in } \Omega_\alpha. \tag{2}$$

Using the relationship $E = -\nabla\Phi$ and substituting equation (2) into (1), we obtain

$$D_\alpha = \sum_{k=0}^{\infty} \chi_{\rm h}^k D_\alpha^k \qquad \text{in } \Omega_\alpha, \tag{3}$$

and

$$D_\alpha^0 = -\varepsilon_\alpha \nabla\Phi_\alpha^0, \tag{4}$$
$$D_\alpha^1 = -\varepsilon_\alpha \nabla\Phi_\alpha^1 - \beta_\alpha \nabla\Phi_\alpha^0 |\nabla\Phi_\alpha^0|^2, \tag{5}$$
$$D_\alpha^2 = -\varepsilon_\alpha \nabla\Phi_\alpha^2 - \beta_\alpha [\nabla\Phi_\alpha^1 |\nabla\Phi_\alpha^0|^2 + 2\nabla\Phi_\alpha^0 (\nabla\Phi_\alpha^0 \cdot \nabla\Phi_\alpha^1)], \tag{6}$$
$$\cdots\cdots\cdots\cdots$$

where $\beta_\alpha = \chi_\alpha/\chi_{\rm h}$. For the electrostatic problems, the electric displacements, D_α, satisfy $\nabla \cdot D_\alpha = 0$. Thus we obtain the expression $\nabla \cdot D_\alpha^k = 0$. Substituting equations (4)–(6) into this expression, we get a set of perturbation equations of potentials $\Phi_\alpha^k(r, \varphi)$.

$$\nabla \cdot (\varepsilon_\alpha \nabla\Phi_\alpha^0) = 0, \tag{7}$$
$$\nabla \cdot (\varepsilon_\alpha \nabla\Phi_\alpha^1) = -\nabla \cdot (\beta_\alpha \nabla\Phi_\alpha^0 |\nabla\Phi_\alpha^0|^2), \tag{8}$$
$$\nabla \cdot (\varepsilon_\alpha \nabla\Phi_\alpha^2) = -\nabla \cdot [\beta_\alpha \nabla\Phi_\alpha^1 |\nabla\Phi_\alpha^0|^2 + 2\beta_\alpha \nabla\Phi_\alpha^0 (\nabla\Phi_\alpha^0 \cdot \nabla\Phi_\alpha^1)], \tag{9}$$
$$\cdots\cdots\cdots.$$

The boundary conditions are obtained by the perturbation method, $\Phi_{\rm i}^k = \Phi_{\rm h}^k$ and $\hat{n} \cdot D_{\rm i}^k = \hat{n} \cdot D_{\rm h}^k$, on the cylindrical particle surface $\partial\Omega_{\rm i}$.

Case (a). Let us consider a cylindrical inclusion with a dielectric function $\varepsilon_{\rm i}(r) = c{\rm e}^{\beta r}$ and nonlinear dielectric constant $\chi_{\rm i}$ embedded in a homogeneous and isotropic host with linear and nonlinear dielectric constants $\varepsilon_{\rm h}$ and $\chi_{\rm h}$. The cylindrical particle has unit radius. If an external electric $E_{\rm app} = E_0\hat{x}$ is applied to the cylindrical composite along the $\hat{x}$ direction, the equations (7)–(9) can be reduced to two-dimensional problems in cylindrical coordinates (r, φ). For the zeroth order potentials $\Phi_\alpha^0(r, \varphi)$, the potentials $\Phi_{\rm h}^0(r, \varphi)$ in the host region can be obtained easily from equation (7): $\Phi_{\rm h}^0(r, \varphi) = -(r + Br^{-1})\cos(\varphi)$.

In the cylindrical region, using the variable separation method, we let the general solution of the zeroth order potential $\Phi_{\rm i}^0(r, \varphi)$ be the following form:

$$\Phi_{\rm i}^0(r, \varphi) = \sum_{n=0}^{\infty} A_n R_n(r)\cos(n\varphi), \tag{10}$$

where $R_n(r)$ is called the radial part. Substituting equation (10) into (7) of the inclusion region, we have the differential equation of the radial part,

$$\frac{1}{r}\frac{\rm d}{{\rm d}r}\left(r\,\frac{{\rm d}R_n(r)}{{\rm d}r}\right) + \beta\,\frac{{\rm d}R_n(r)}{{\rm d}r} - \frac{n^2}{r^2}R_n(r) = 0. \tag{11}$$

The authors of [16, 17] have given the solution of differential equation (11) by using the Kummer function $F(\lambda, \gamma, z)$ and the Frobenius series method [18]: $R_n(r) = (\beta r)^n F(n, 2n+1, -\beta r)$. Using the boundary conditions and considering the Kummer function $F(1, 3, -\beta r) = 2({\rm e}^{-\beta r} + \beta r - 1)/\beta^2 r^2$, we obtain the zeroth order potentials $\Phi_{\rm h}^0(r, \varphi)$ and $\Phi_{\rm i}^0(r, \varphi)$ in [17]:

$$\Phi_{\rm h}^0(r, \varphi) = -(r + Br^{-1})E_0\cos(\varphi), \qquad \text{in } \Omega_{\rm h}, \tag{12}$$
$$\Phi_{\rm i}^0(r, \varphi) = -AE_0\cos(\varphi)({\rm e}^{-\beta r} + \beta r - 1)/(\beta^2 r), \qquad \text{in } \Omega_{\rm i}, \tag{13}$$

where

$$A = 2\varepsilon_{\rm h}/(c{\rm e}^\beta v_1 + \varepsilon_{\rm h} v_2),$$

$$B = (\varepsilon_h v_2 - c\mathrm{e}^{\beta} v_1)/(c\mathrm{e}^{\beta} v_1 + \varepsilon_h v_2),$$
$$v_1 = (1 - \mathrm{e}^{-\beta} - \beta\mathrm{e}^{-\beta})/\beta^2,$$
$$v_2 = (\mathrm{e}^{-\beta} + \beta - 1)/\beta^2.$$

Substituting the zeroth order potential into equation (8), we will obtain the first order potentials $\Phi^1_\alpha(r, \varphi)$. Considering the two equations (12) and (13), we rewrite equation (8) in the form

$$\nabla^2\Phi^1_h = (8B^2r^{-5} - 4B^3r^{-7})\cos(\varphi)E^3_0/\varepsilon_h - 4Br^{-3}\cos(3\varphi)E^3_0/\varepsilon_h, \qquad \text{in } \Omega_h \tag{14}$$

$$\left(\beta + \frac{1}{r}\right)\frac{\partial\Phi^1_i(r,\varphi)}{\partial r} + \frac{\partial^2\Phi^1_i(r,\varphi)}{\partial r^2} + \frac{1}{r^2}\frac{\partial^2\Phi^1_i(r,\varphi)}{\partial\varphi^2} = -A^3E^3_0\beta_i[F_1(r)\cos(\varphi) + F_3(r)\cos(3\varphi)]/c \qquad \text{in } \Omega_i \tag{15}$$

where

$$F_1(r) = [(9\mathrm{e}^{-4\beta r} + \mathrm{e}^{-2\beta r})\beta^4r^4 + (33\mathrm{e}^{-4\beta r} - 15\mathrm{e}^{-3\beta r} + \mathrm{e}^{-2\beta r} - 3\mathrm{e}^{-\beta r})\beta^3r^3 + (56\mathrm{e}^{-4\beta r} - 58\mathrm{e}^{-3\beta r} - 4\mathrm{e}^{-2\beta r} + 6\mathrm{e}^{-\beta r})\beta^2r^2 + (50\mathrm{e}^{-4\beta r} - 102\mathrm{e}^{-3\beta r} + 54\mathrm{e}^{-2\beta r} - 2\mathrm{e}^{-\beta r})\beta r + 16\mathrm{e}^{-\beta r}(\mathrm{e}^{-\beta r} - 1)^3]/(4\beta^6r^7)$$

$$F_3(r) = [(3\mathrm{e}^{-4\beta r} - \mathrm{e}^{-2\beta r})\beta^3r^3 + (11\mathrm{e}^{-4\beta r} - 13\mathrm{e}^{-3\beta r} - \mathrm{e}^{-2\beta r} + 3\mathrm{e}^{-\beta r})\beta^2r^2 + (12\mathrm{e}^{-4\beta r} - 30\mathrm{e}^{-3\beta r} + 24\mathrm{e}^{-2\beta r} - 6\mathrm{e}^{-\beta r})\beta r - 2\mathrm{e}^{-\beta r}(1 - \mathrm{e}^{-\beta r})^3]/(4\beta^5r^6).$$

Because equation (14) is Poisson's equation, we write out the general solution of the host region with the boundary condition at infinity,

$$\Phi^1_h(r,\varphi) = -B_1r^{-1}\cos(\varphi)E^3_0 - B_3r^{-3}\cos(3\varphi)E^3_0 - (B^2r^{-3} - \tfrac{1}{6}B^3r^{-5})E^3_0\cos(\varphi)/\varepsilon_h - \tfrac{1}{2}Br^{-1}E^3_0\cos(3\varphi)/\varepsilon_h. \tag{16}$$

For equation (15), the general solution can be given by the sum of the solution $\Phi^{1g}_i(r,\varphi)$ of equation (15) without the source term and the particular solution $\Phi^{1p}_i(r,\varphi)$ of equation (15) because it is a linear differential equation. From equation (11), we have

$$\Phi^{1g}_i(r,\varphi) = -\sum_{n=0}^{\infty} A_n(\beta r)^n F(n, 2n+1, -\beta r)\cos(n\varphi)E^3_0. \tag{17}$$

From equation (15), the particular solution $\Phi^{1p}_i(r,\varphi)$ is given as

$$\Phi^{1p}_i(r,\varphi) = -[a(r)\cos(\varphi) + b(r)\cos(3\varphi)]A^3E^3_0. \tag{18}$$

Here we should note that we only give the useful coefficients of the particular solution for calculating the linear and nonlinear effective responses. Using Frobenius's method [18], we get the useful coefficients of function $a(r) = c^{-1}\beta_i\sum_{k=0}^{\infty} a_k r^{k+2}$ as follows:

$$a_k = \beta^{k+1}\sum_{n=0}^{k}(-1)^{n+1}A^0_{k-n}(k-n+2)(k-n)!/(k+3)!,$$
$$a_0 = \beta A^0_0/3,$$
$$A^0_k = \tfrac{1}{4}\sum_{i=1}^{5} A^i_k, \qquad (k = 0, 1, 2, 3, \ldots),$$
$$A^1_k = [9(-4)^{k+3} + (-2)^{k+3}]/(k+3)!,$$
$$A^2_k = (-1)^{k+4}[-3 - 15(3)^{k+4} + 33(4)^{k+4} + 2^{k+4}]/(k+4)!,$$
$$A^3_k = (-1)^{k+5}[6 - 4(2)^{k+5} - 58(2)^{k+5} + 56(4)^{k+5}]/(k+5)!,$$
$$A^4_k = (-1)^{k+6}[-2 + 54(2)^{k+6} - 102(3)^{k+6} + 50(4)^{k+6}]/(k+6)!,$$
$$A^5_k = -16(-1)^{k+7}[1 - 3(2)^{k+7} + 3^{k+8} - 4^{k+7}]/(k+7)!.$$

Thus the general solution of equation (15) is the following form:

$$\Phi_{\mathrm{i}}^{1}(r,\varphi) = -A_1\beta r F(1,3,-\beta r)\cos(\varphi)E_0^3 - A_3(\beta r)^3 F(3,7,-\beta r)\cos(3\varphi)E_0^3 \\ - [a(r)\cos(\varphi) + b(r)\cos(3\varphi)]A^3E_0^3. \tag{19}$$

Using the boundary conditions, we determine the constants of equations (18) and (19). The coefficients for determining the effective response are given below.

$$A_1 = [f(B) - a(1)A^3 - c\mathrm{e}^{\beta}a'(1)A^3 - \beta_{\mathrm{i}}A^3(\tfrac{3}{4}v_1^3 + \tfrac{1}{4}v_1v_2^2)]/[(c\mathrm{e}^{\beta} + \varepsilon_{\mathrm{h}})\beta F(1,3,-\beta) \\ - c\mathrm{e}^{\beta}\beta^2 F(2,4,-\beta)/3],$$

$$B_1 = A_1\beta F(1,3,-\beta) + a(1)A^3 - (B^2 - \tfrac{1}{6}B^3)/\varepsilon_{\mathrm{h}},$$

$$f(B) = \tfrac{3}{4}(1-B)^3 + \tfrac{1}{4}(1+B) - \tfrac{9}{4}B^2 + \tfrac{5}{12}B^3,$$

$$a(1) = c^{-1}\beta_{\mathrm{i}}\sum_{k=0}^{\infty} a_k,$$

$$a'(1) = c^{-1}\beta_{\mathrm{i}}\sum_{k=0}^{\infty}(k+2)a_k.$$

Up to now, we have given the zeroth and the first order potentials. It is known that the perturbation solutions of the first order are enough to predict the third order nonlinear effective response because of the perturbation solution $\Phi_{\alpha}^{k}(r,\varphi) \propto E_0^{2k+1}$.

Case (b). For the linear dielectric response of a cylindrical inclusion with the power-law profile $\varepsilon_{\mathrm{i}}(r) = c_k r^k$, we can deal with the weakly nonlinear problem of graded composites as the above process of *case (a)*. The zeroth order potentials are given in [12]:

$$\Phi_{\mathrm{i}}^{0}(r,\varphi) = -H_0 r^s\cos(\varphi)E_0, \qquad \text{in } \Omega_{\mathrm{i}}, \tag{20}$$

$$\Phi_{\mathrm{h}}^{0}(r,\varphi) = -(r + D_0 r^{-1})\cos(\varphi)E_0, \qquad \text{in } \Omega_{\mathrm{h}}, \tag{21}$$

where $s = (\sqrt{k^2+4} - k)/2$, $H_0 = 2\varepsilon_{\mathrm{h}}/(\varepsilon_{\mathrm{h}} + c_k s)$, $D_0 = (\varepsilon_{\mathrm{h}} - c_k s)/(\varepsilon_{\mathrm{h}} + c_k s)$. With the perturbation method, we have obtained the first order potential equation of the inclusion region from equation (8):

$$(k+1)\frac{\partial\Phi_{\mathrm{i}}^{1}(r,\varphi)}{\partial r} + r\frac{\partial^2\Phi_{\mathrm{i}}^{1}(r,\varphi)}{\partial r^2} + r^{-1}\frac{\partial^2\Phi_{\mathrm{i}}^{1}(r,\varphi)}{\partial\varphi^2} \\ = -\beta_{\mathrm{i}}H_0^3E_0^3[G_1(r)\cos(\varphi) + G_3(r)\cos(3\varphi)]/c_k, \tag{22}$$

where

$$G_1(r) = d_1 r^{3s-k-3},$$
$$G_3(r) = d_3 r^{3s-k-3},$$
$$d_1 = [\tfrac{1}{2}s(s-1) - \tfrac{1}{4}ks](3s^2+1) + \tfrac{1}{2}(s^2-1),$$
$$d_3 = (s^2-1)[\tfrac{1}{2}(s^2-s-1) - \tfrac{1}{4}ks].$$

In order to solve equation (22), the variable separation method is also applied to equation (22) without source. Letting $\Phi_{\mathrm{i}}^{0}(r,\varphi) = \sum_{n=0}^{\infty} H_n R_n(r)\cos(n\varphi)$ and substituting it into equation (22) without the source term, we have

$$r^2\frac{\partial^2 R_n}{\partial r^2} + (k+1)r\frac{\partial R_n}{\partial r} - n^2R_n = 0. \tag{23}$$

Clearly the solution of equation (23) has the form $r^{s(n)}$ with $s(n) = (\sqrt{k^2+4n^2} - k)/2$. Thus, the general solution of equation (23) is obtained. Considering the particular solution of equation (22) and the potential of the first order in the host region, which is the same form as equation (16), in both inclusion and host regions, we obtain the first order solution of potentials by solving equations (16) and (22):

$$\Phi_{\rm h}^1(r,\varphi) = -[D_1 r^{-1} + (D_0^2 r^{-3} - \tfrac{1}{6}D_0^3 r^{-5})/\varepsilon_{\rm h}]\cos(\varphi)E_0^3 - (D_3 r^{-3} + \tfrac{1}{2}D_0 r^{-1}/\varepsilon_{\rm h})\cos(3\varphi)E_0^3, \quad (24)$$

$$\Phi_{\rm i}^1(r,\varphi) = -(H_1 r^s + H r^{3s-k-2})\cos(\varphi)E_0^3 - (H_3 r^p + G r^{3s-k-2})\cos(3\varphi)E_0^3, \quad (25)$$

where $p = (\sqrt{k^2+36} - k)/2$. The coefficients of the first potentials for determining the effective response are obtained from the boundary conditions of the dielectric displacement and potential on the cylindrical surfaces:

$$H_1 = [1 - 2D_0 - D_0^3/3 - s\beta_{\rm i} H_0^3(1/4 + 3s^2/4) - \varepsilon_{\rm h} H - c_k(3s-k-2)H]/(c_k s + \varepsilon_{\rm h}),$$

$$H = \beta_{\rm i} H_0^3 d_1/(c_k d_2),$$

$$d_2 = 1 - (3s-k-2)(3s-2),$$

$$D_1 = H_1 + H - (D_0^2 - \tfrac{1}{6}D_0^3)/\varepsilon_{\rm h}.$$

3. Effective nonlinear dielectric response

At low concentration of graded cylindrical inclusion, we will use Landau's method previously proposed in [15] to calculate the effective response of graded nonlinear composites, where the unidirectional parallel grade cylindrical inclusions are dispersedly embedded in a host.

$$\frac{1}{V}\int_V [(\varepsilon_{\rm i}(r) - \varepsilon_{\rm h})E + (\chi_{\rm i} - \chi_{\rm h})|E|^2 E]\,{\rm d}V = \bar{D} - (\varepsilon_{\rm h}\bar{E} + \chi_{\rm h}|\bar{E}|^2\bar{E}) \quad (26)$$

where V is the volume of composites. $\bar{D}$ and $\bar{E}$ denote, respectively, the ensemble average displacement and electric fields. In order to calculate the effective nonlinear response of the graded composites, we first define the effective response of the graded composites when we regard the graded composites as an effective homogeneous medium,

$$\bar{D} = \varepsilon_{\rm e}\bar{E} + \chi_{\rm e}|\bar{E}|^2\bar{E}, \quad (27)$$

where $\varepsilon_{\rm e}$ and $\chi_{\rm e}$ are the linear and nonlinear effective dielectric response, respectively. Substituting equation (27) into (26) and considering the fact that the integration of the left-hand side of equation (26) is zero in the host region, we have

$$\frac{1}{V}\int_{\Omega_{\rm i}} [(\varepsilon_{\rm i}(r) - \varepsilon_{\rm h})E + (\chi_{\rm i} - \chi_{\rm h})|E|^2 E]\,{\rm d}V = (\varepsilon_{\rm e} - \varepsilon_{\rm h})\bar{E} + (\chi_{\rm e} - \chi_{\rm h})|\bar{E}|^2\bar{E}. \quad (28)$$

In the dilute limit, we can regard the external applied field $E_{\rm a} = E_0\hat{x}$ as the average electric field $\bar{E}$ because, in this case, the interactions among particles can be neglected. Hence the effective response of the graded composites will be obtained by using the potentials of a cylindrical inclusion region at low concentration limit. Here we should note that two widely used methods, the Maxwell–Garnett approximation (MGA) and the effective-medium approximation (EMA), were developed to calculate the nonlinear dielectric response [20–22]. The two methods can be extended to study the effective response of nonlinear graded composites for higher concentration. Here we discuss the effective responses in the dilute limit with average field $\bar{E} = E_{\rm a}$, although the concentration range of our method is smaller than that of the Levy and Bergman method [21, 22].

Case (a). For the exponent dielectric function profile, substituting the potentials of the cylindrical particle region into equation (28), we get the effective dielectric response,

$$\varepsilon_{\rm e} = \varepsilon_{\rm h} + f_{\rm i}A[c({\rm e}^{\beta} - \beta - 1)/\beta^2 - \varepsilon_{\rm h}({\rm e}^{-\beta} + \beta - 1)/\beta^2], \quad (29)$$

$$\chi_{\rm e} = \chi_{\rm h} + 2f_{\rm i}\chi_{\rm h}A_1[c({\rm e}^{\beta} - \beta - 1)/\beta - \varepsilon_{\rm h}({\rm e}^{-\beta} + \beta - 1)/\beta] + f_{\rm i}A^3\chi_{\rm h}[cT_1 - \varepsilon_{\rm h}a(1)] + \tfrac{1}{4}f_{\rm i}A^3(\chi_{\rm i} - \chi_{\rm h})T_2, \quad (30)$$

where f_i is the volume fraction of the cylindrical inclusion. We should note that in the above derivation of effective response we have applied the formulae $F(1,2,\beta) = (\mathrm{e}^\beta - 1)/\beta$ and $F(1,3,\beta) = 2(\mathrm{e}^\beta - \beta - 1)/\beta^2$. The coefficients T_1 and T_2 are

$$
\begin{aligned}
T_1 &= \sum_{k=0}^{\infty} (k+3)[P_{k+2}(1) - P_{k+2}(0)]a_k,\\
T_2 &= \sum_{k=1}^{\infty} \beta^{k-1}(s_k^1 + s_k^2 + s_k^3)/(k+1),\\
s_k^1 &= 8[3(-2)^{k+4} - 3(-1)^{k+4} - (-3)^{k+4}]/(k+4)!,\\
s_k^2 &= 8[-(-3)^{k+3} + (-2)^{k+3} + (-1)^{k+3}]/(k+3)!,\\
s_k^3 &= [-3(-3)^{k+2} + (-2)^{k+2} - (-1)^{k+2}]/(k+2)!,
\end{aligned}
$$

where the two values of $P_{k+2}(1)$ and $P_{k+2}(0)$ in the constant T_1 can be calculated by the function $P_k(x)$ if $x = 1$ and $x = 0$, respectively,

$$P_k(x) = \int x^k \mathrm{e}^{\beta x}\,\mathrm{d}x = \frac{\mathrm{e}^{\beta x}}{\beta^{k+1}}[(\beta x)^k - k(\beta x)^{k-1} + k(k-1)(\beta x)^{k-2} - \cdots + (-1)^k k!].$$

Next we will demonstrate that our results can be reduced to the effective nonlinear responses of homogeneous composites when the parameter $\beta \to 0$. In this case, let $\beta \to 0$; we have the following limits,

$$
\begin{aligned}
&\lim_{\beta\to 0} v_1 = \lim_{\beta\to 0} v_2 = 1/2,\\
&\lim_{\beta\to 0} a(1) = \lim_{\beta\to 0} a'(1) = 0,\\
&\lim_{\beta\to 0} B = (\varepsilon_\mathrm{h} - c)/(\varepsilon_\mathrm{h} + c) = b_0,\\
&\lim_{\beta\to 0} (\mathrm{e}^\beta - \beta - 1)/\beta^2 = 1/2,\\
&\lim_{\beta\to 0} (\mathrm{e}^{-\beta} + \beta - 1)/\beta^2 = 1/2,\\
&\lim_{\beta\to 0} A = 2T,\\
&\lim_{\beta\to 0} [P_{k+2}(1) - P_{k+2}(0)] = 1/(k+3),\\
&\lim_{\beta\to 0} T_1 = \lim_{\beta\to 0} a(1) = 0,\\
&\lim_{\beta\to 0} A_1\beta = [1 - 2b_0 - b_0^3/3 - \beta_\mathrm{i} T^3]/\sigma = T_0,\\
&\lim_{\beta\to 0} T_2 = 1/2,
\end{aligned}
$$

where the coefficients $b_0 = (\varepsilon_\mathrm{h} - c)/(c + \varepsilon_\mathrm{h})$, $T = 2\varepsilon_\mathrm{h}/\sigma$, $\sigma = \varepsilon_\mathrm{h} + c$. Substituting these limits into equations (29) and (30), we get the effective responses of the homogeneous nonlinear composites if $\varepsilon_\mathrm{i}(r) = c$ (i.e. $\beta \to 0$) from equations (29) and (30), respectively.

$$\varepsilon_\mathrm{e} = \varepsilon_\mathrm{h} + T f_\mathrm{i}(c - \varepsilon_\mathrm{h}), \tag{31}$$

$$\chi_\mathrm{e} = \chi_\mathrm{h} + f_\mathrm{i}\chi_\mathrm{h}(c - \varepsilon_\mathrm{h})T_0 + f_\mathrm{i}T^3(\chi_\mathrm{i} - \chi_\mathrm{h}), \tag{32}$$

where the coefficients T and T_0 are also, respectively, the coefficients c and b_3 of [19, 15]. Comparing equations (31) and (32) with that of the nonlinear response of weakly nonlinear homogeneous composites of [19, 15], we will see that our results are the same as those results of [19, 15] for $\varepsilon_\mathrm{i} = c$. In order to show the effects of the exponent dielectric function profile on the effective nonlinear dielectric response, in figure 1 we plot the effective nonlinear

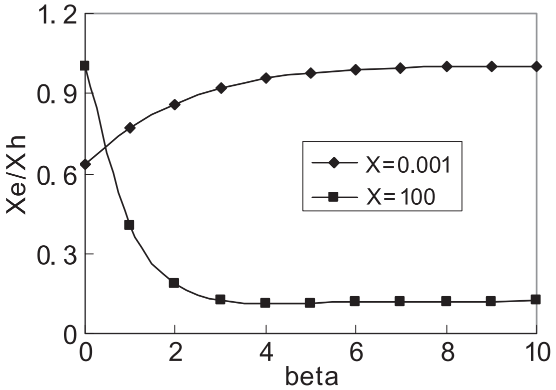

Figure 1. The effective nonlinear dielectric response contrast ratio, χ_e/χ_h, of the exponent dielectric function profile, $\varepsilon_i = c e^{\beta r}$, as a function of the parameter β for different contrast ratios, χ_i/χ_h, at volume fraction $f_i = 0.1$. In the figure, the symbols X and X_e/X_h are the quantities χ_i/χ_h and $(\chi_e/\chi_h)/\max(\chi_e/\chi_h)$, respectively, and the parameter ratio $c/\varepsilon_h = 1$.

dielectric response of graded cylindrical composites with dielectric profile $\varepsilon_i = c e^{\beta r}$ versus the parameter β at $f_i = 0.1$. Clearly, the effective nonlinear dielectric response contrast ratio, χ_e/χ_h, increases (or decreases) as the parameter β increases for small (or large) contrast ratio χ_i/χ_h.

Case (b). For the power-law dielectric function profile $\varepsilon_i(r) = c_k r^k$, substituting the potentials of the cylindrical inclusion into the left-hand side of equation (28), we have the formulae of effective responses,

$$\varepsilon_e = \varepsilon_h + f_i H_0[c_k(s+1)/(s+k+1) - \varepsilon_h], \tag{33}$$

$$\begin{aligned}\chi_e = \chi_h &+ f_i \chi_h H_1[c_k(s+1)/(k+s+1) - \varepsilon_h] + f_i \chi_h H[c_k(3s-k-1)/(3s-1) - \varepsilon_h] \\ &+ \tfrac{1}{4} f_i H_0^3(\chi_i - \chi_h)(3s^3 + s^2 + s + 3)/(3s-1),\end{aligned} \tag{34}$$

where the coefficients H, H_0 and H_1 are given in equations (20), (24) and (25). Here we demonstrate that our results can be reduced to the effective nonlinear response of weakly nonlinear composites when we take the limit $k \to 0$ (i.e. $\varepsilon_i = c_0$) for the power-law dielectric profile. Taking $k \to 0$ in equations (33) and (34), we have the following limits:

$$\lim_{k\to 0} H_0 = 2\varepsilon_h/(\varepsilon_h + c_0) = C,$$

$$\lim_{k\to 0}(s+1)/(s+k+1) = 1,$$

$$\lim_{k\to 0}(3s^3 + s^2 + s + 3)/(3s-1) = 4,$$

$$\begin{aligned}&\lim_{k\to 0}\{H_1[c_k(s+1)/(k+s+1) - \varepsilon_h] - H[c_k(3s-k-1)/(3s-1) - \varepsilon_h]\} \\ &\qquad = (c_0 - \varepsilon_h)(1 - 2M_0 - M_0^3/3 - \beta_i c^3)/(c_0 + \varepsilon_h) = b_3(c_0 - \varepsilon_h),\end{aligned}$$

where

$$M_0 = (\varepsilon_h - c_0)/(c_0 + \varepsilon_h), \qquad b_3 = (1 - 2M_0 - M_0^3/3 - \beta_i c^3)/(c_0 + \varepsilon_h),$$

$$C = 2\varepsilon_h/(c_0 + \varepsilon_h).$$

Substituting these limits into equations (33) and (34), we have the following effective response:

$$\varepsilon_e = \varepsilon_h + f_i C(c_0 - \varepsilon_h), \tag{35}$$

$$\chi_e = \chi_h + f_i \chi_h b_3(c_0 - \varepsilon_h) + f_i(\chi_i - \chi_h)C^3. \tag{36}$$

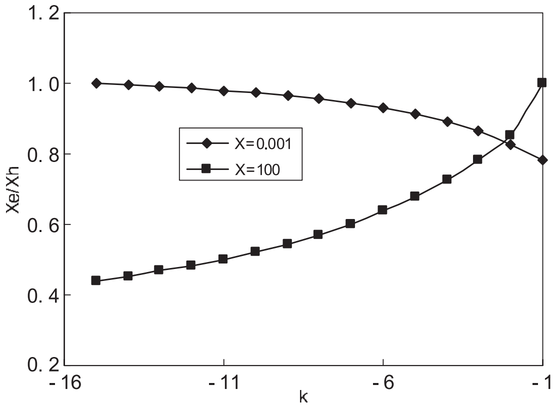

Figure 2. The effective nonlinear dielectric response contrast ratio, χ_e/χ_h, of the power-law dielectric function profile, $\varepsilon_i = c_k r^k$, as a function of the parameter k for different contrast ratios, χ_i/χ_h, at volume fraction $f_i = 0.1$. In the figure, the symbols X and X_e/X_h, are the quantities χ_i/χ_h and $(\chi_e/\chi_h)/\max(\chi_e/\chi_h)$, respectively, and the parameter ratio $c_k/\varepsilon_h = 1$.

Comparing equations (35) and (36) with the results of [19, 15], then we regain the effective response of weakly nonlinear homogeneous composites with $\varepsilon_i = c_0$. In figure 2, with equation (34), we plot the effective nonlinear dielectric response of graded cylindrical composites with dielectric profile $\varepsilon_i = c_k r^k$ versus the parameter k for $f_i = 0.1$. In contrast to the exponent dielectric profile, the effective nonlinear dielectric response contrast ratio, χ_e/χ_h, increases (or decreases) as the parameter k increases for large (or small) contrast ratio χ_i/χ_h.

4. Conclusion

We have developed the perturbation method to solve the problem of nonlinear graded composites. For weakly nonlinear graded composites, the local potentials are derived from the perturbation equations of graded composites of a cylindrical particle with exponent and power-law dielectric profiles. The effective nonlinear responses are obtained in the dilute limit. Furthermore, we have shown that our results can be exactly reduced to the effective response of nonlinear homogeneous composites. Our results can be extended to predict the effective nonlinear thermal conductivities of Kerr-like nonlinear graded composites without contact heat resistance on the inclusion surfaces when the linear order thermal conductivity of the cylindrical inclusion is an exponential function or a power-law function of radial distances of the inclusion. For spherical inclusion particles with exponent and power-law dielectric profiles, the perturbation method can also be used to deal with the effective nonlinear response of spherical nonlinear graded composites. The effective response of a graded weakly nonlinear composite is different form that of homogeneous weakly nonlinear composites. From our results, we can see that the linear dielectric function profiles of graded inclusions give rise to strong effects on the effective nonlinear response of graded composites. For higher concentration, the effective response of graded nonlinear composites can be studied by EMA and MGA methods combined with our potential solution of graded composites [21, 22]. For a complicated graded function, a number method is useful for estimating the effective nonlinear dielectric response, such as the finite element method [23]. Here we should note that some methods for dealing with nonlinear homogeneous composites may be developed to solve the effective nonlinear response of graded composites, such as the homotopy method for the weakly nonlinear problem [24, 25], the variational method for strongly nonlinear composites [26–28], Rayleigh method for periodic nonlinear graded composites [29], and so on.

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