Vol 18 No 2, February 2009
1674-1056/2009/18(02)/0757-06

Chinese Physics B

© 2009 Chin. Phys. Soc.
and IOP Publishing Ltd

Effective response of nonlinear cylindrical coated composites under external AC and DC electric field

Shen Yu-Yan(沈玉艳), Chen Xiao-Gang(陈小刚)†, Cui Wei(崔 巍),
Hao Yan-Hua(郝艳华), and Li Qian-Qian(李倩倩)

College of Science, Inner Mongolia University of Technology, Hohhot 010051, *China*

(Received 12 July 2008; revised manuscript received 15 August 2008)

This paper uses the perturbation method to study effective response of nonlinear cylindrical coated composites. Under the external AC and DC electric field $\boldsymbol{E}_{\rm a}(1+\sin\omega t)$, the local potentials of composites at all harmonic frequencies are induced. An effective nonlinear response to composite is given for the cylindrical coated inclusions in the dilute limit.

Keywords: nonlinear composites, effective nonlinear response, the external AC and DC electric field

PACC: 7220H, 7722C, 7320

1. Introduction

Recently, nonlinear responses of composites have been widely studied because of their potential applications in engineering and theory.[1−6] The effective response of composites is not only dependent on the intrinsic constitutive relation, physical properties of materials, particle shape, and volume fraction, but also on the applied external field.[7−10] In particular, when an external AC electric field is applied to the nonlinear composites, a lot of works have focused on the effective nonlinear AC response at harmonics frequencies. For example, Levy *et al*[4] have investigated the nonlinear response at the higher harmonic frequencies. Hui *et al*[10] have studied the effective nonlinear response of nonlinear inhomogeneous composites with external AC field. Gu and Wei *et al*[11−14] have derived the nonlinear effective response of cubic weakly nonlinear composites at harmonic frequencies of external AC electric field. Chen and Wei *et al*[15] have investigated the effects of the applied AC electric field on the effective response of nonlinear spherical coated composites. Wei *et al*[16,17] have studied the effective response of nonlinear cylindrical coated composites under AC electric field. Chen *et al*[18] have investigated the effective nonlinear AC response to composite with spherical particles under the action of a sinusoidal field $\boldsymbol{E}_1\sin(\omega t)+\boldsymbol{E}_3\sin(3\omega t)$. In this paper, we will devote to the effective response of the nonlinear cylindrical coated composites under the external AC and DC electric field $\boldsymbol{E}_{\rm app}=\boldsymbol{E}_{\rm a}(1+\sin\omega t)$.

We consider homogeneous and isotropic composites, where coated cylindrical particles are randomly embedded in a host medium. The constitutive relations between electric displacement $\boldsymbol{D}$ and electric field $\boldsymbol{E}$ are assumed in the form

$$\boldsymbol{D}_\alpha=\varepsilon_\alpha\boldsymbol{E}+\chi_\alpha\left|\boldsymbol{E}\right|^2\boldsymbol{E},\quad \text{in}\quad \Omega_\alpha,\quad \alpha=\text{i},\text{c},\text{h}, \tag{1}$$

where the subscripts $\alpha=$ i, c, h denote the quantities in inclusion core particle (i), coated shell layer material (c), and host regions (h), respectively. Ω_α is the region occupied by the α-type material. Both real linear and nonlinear dielectric constant, ε_α and χ_α, are independent of the frequency of applied field, which is a valid assumption for the low-frequency processes. The corresponding governing equations satisfy $\nabla\cdot\boldsymbol{D}=0$ and $\nabla\times\boldsymbol{E}=0$. The boundary conditions of the coated composites are the continuity of the potentials and the normal components of the electric displacement on each interface of the core, coated layer, and host. At infinity the potential must match that of the external applied field.

2. Perturbation method for nonlinear AC and DC process

Let us take into account weakly nonlinear composites with coated cylindrical inclusions randomly embedded in a host medium, in which the nonlinear

†Corresponding author. E-mail: xiaogang_chen@imut.edu.cn

terms in the constitutive equations are smaller than the linear terms. We assume that the concentric cylindrical particle has a radius of core a and a coated shell radius b. The external field $\boldsymbol{E}_{\rm app} = \boldsymbol{E}_{\rm a}(1+\sin\omega t)$ with the finite frequency ω is applied to the composites along $\hat{x}$-direction, where $\boldsymbol{E}_{\rm a} = E_0\hat{x}$. The electric potentials at a point $\boldsymbol{x}$ can be expressed in the following form:

$$\begin{aligned}&\Phi_\alpha(\boldsymbol{x},\omega,t)\\ =\ &\Phi_0^\alpha(\boldsymbol{x})+\Phi_1^\alpha(\boldsymbol{x})\sin\omega t+\Phi_2^\alpha(\boldsymbol{x})\cos 2\omega t\\ &+\Phi_3^\alpha(\boldsymbol{x})\sin 3\omega t+\Phi_4^\alpha(\boldsymbol{x})\cos 4\omega t+\ldots\\ &\text{in}\quad \Omega_\alpha,\alpha=\mathrm{i},\mathrm{c},\mathrm{h}.\end{aligned}\tag{2}$$

For weakly nonlinear composites, we can develop the perturbation method to deal with the potentials.[11] The nonlinear susceptibility $\chi_{\rm h}$ in the host medium is chosen as the perturbation parameter. The potentials Φ_k^α can be expanded with the parameter $\chi_{\rm h}$ as:

$$\begin{aligned}&\Phi_k^\alpha(\boldsymbol{x})\\ =\ &\Phi_k^{0\alpha}(\boldsymbol{x})+\chi_{\rm h}\Phi_k^{1\alpha}(\boldsymbol{x})+\chi_{\rm h}^2\Phi_k^{2\alpha}(\boldsymbol{x})+\ldots,\\ &\text{in}\quad \Omega_\alpha,\alpha=\mathrm{i},\mathrm{c},\mathrm{h}.\end{aligned}\tag{3}$$

Using the electric field equation $\boldsymbol{E}_\alpha(\boldsymbol{x},\omega,t) = -\nabla\Phi_\alpha(\boldsymbol{x},\omega,t)$ and substituting the Eqs.(2) and (3) into Eq.(1), we get the perturbation expansions for the electric displacement $\boldsymbol{D}$ in inclusion, coated layer and host regions,

$$\begin{aligned}&\boldsymbol{D}_\alpha(\boldsymbol{x},\omega,t)\\ =\ &\boldsymbol{D}_0^\alpha(\boldsymbol{x})+\boldsymbol{D}_1^\alpha(\boldsymbol{x})\sin\omega t+\boldsymbol{D}_2^\alpha(\boldsymbol{x})\cos 2\omega t\\ &+\boldsymbol{D}_3^\alpha(\boldsymbol{x})\sin 3\omega t+\boldsymbol{D}_4^\alpha(\boldsymbol{x})\cos 4\omega t+\ldots\\ &\alpha=\mathrm{i},\mathrm{c},\mathrm{h}.\end{aligned}\tag{4}$$

The local electric displacement $\boldsymbol{D}_k^\alpha$ is written as

$$\begin{aligned}&\boldsymbol{D}_k^\alpha(\boldsymbol{x})\\ =\ &\boldsymbol{D}_k^{0\alpha}(\boldsymbol{x})+\chi_{\rm h}\boldsymbol{D}_k^{1\alpha}(\boldsymbol{x})+\chi_{\rm h}^2\boldsymbol{D}_k^{2\alpha}(\boldsymbol{x})+\ldots,\\ &\text{in}\quad \Omega_\alpha;\alpha=\mathrm{i},\mathrm{c},\mathrm{h};\ \ k=0,1,2,\ldots,\end{aligned}\tag{5}$$

where $\boldsymbol{D}_k^{s\alpha}(\boldsymbol{x})$ is a function of the potentials $\Phi_k^{s\alpha}(\boldsymbol{x})$. If each term of Eq.(5) satisfies its corresponding governing equation, the governing equation $\nabla\cdot\boldsymbol{D}(\boldsymbol{x},\omega,t)=0$, is satisfied. Therefore we obtain a set of equations

$$\begin{aligned}&\nabla\cdot\boldsymbol{D}_k^{s\alpha}(\boldsymbol{x})=0,\quad \alpha=\mathrm{i},\mathrm{c},\mathrm{h};\\ &s=0,1,2,3,\ldots;k=0,1,2,3,\ldots.\end{aligned}\tag{6}$$

The corresponding boundary conditions of each term in the perturbation potential and electric displacement are, respectively, as follows.

At the interfaces between the inclusion core and inside layer of the coated shell, $\partial\Omega_{\rm i}$,

$$\begin{aligned}&\Phi_k^{\rm si}(\boldsymbol{x})=\Phi_k^{\rm sc}(\boldsymbol{x}),\quad \hat{\boldsymbol{n}}\cdot\boldsymbol{D}_k^{\rm si}(\boldsymbol{x})=\hat{\boldsymbol{n}}\cdot\boldsymbol{D}_k^{\rm sc}(\boldsymbol{x}),\\ &\text{on}\ \ \partial\Omega_{\rm i}, s=0,1,2,3,...;\quad k=0,1,2,3,...,\end{aligned}\tag{7}$$

where $\hat{\boldsymbol{n}}$ is the surface normal of the core.

At the interface between the outside layer of the coated shell and host medium, $\partial\Omega_{\rm c}$,

$$\begin{aligned}&\Phi_k^{\rm sc}(\boldsymbol{x})=\Phi_k^{\rm sh}(\boldsymbol{x}),\quad \hat{\boldsymbol{n}}\cdot\boldsymbol{D}_k^{\rm sc}(\boldsymbol{x})=\hat{\boldsymbol{n}}\cdot\boldsymbol{D}_k^{\rm sh}(\boldsymbol{x})\\ &\text{on}\ \ \partial\Omega_{\rm c}, k=0,1,2,3,...; s=0,1,2,3,....\end{aligned}\tag{8}$$

At infinity the potentials satisfy

$$\left.\frac{\partial\Phi_k^{\rm sh}(\boldsymbol{x})}{\partial x}\right|_{x\to\infty}=\begin{cases}-E_0, & \text{if}\ \ k=0\ \ \text{and}\ \ s=1,\\ 0, & \text{if}\ \ k\neq 0\ \ \text{and}\ \ s\neq 1.\end{cases}\tag{9}$$

3. Potentials in coated cylindrical composites

For convenience, we only consider the two-dimensional nonlinear composite of coated cylinder inclusion embedded in a host. The inclusions are random and their axes are aligned in the $\hat{z}$-direction. We will derive the zero and first potential at harmonic because it is enough to calculate the effective cubic nonlinear response of Kerr composites.

The governing equations of the zero-order potential at the zero frequency in the media regions are Laplace equations:

$$\varepsilon_\alpha\nabla^2\Phi_0^{0\alpha}(\boldsymbol{x})=0,\quad \text{in}\ \ \Omega_\alpha,\alpha=\mathrm{i},\mathrm{c},\mathrm{h}.\tag{10}$$

With the boundary conditions we get the zero-order potentials at the zero frequency:

$$\Phi_0^{0\rm i}(\boldsymbol{x})=A_0(a_0+1)rE_0\cos\theta\quad (r\le a),\tag{11}$$

$$\Phi_0^{0\rm c}(\boldsymbol{x})=A_0(a_0r+a^2r^{-1})E_0\cos\theta\quad (a\le r\le b),\tag{12}$$

$$\Phi_0^{0\rm h}(\boldsymbol{x})=(-r+B_0b^2r^{-1})E_0\cos\theta\quad (r\ge b),\tag{13}$$

where

$$A_0=\frac{2(\varepsilon_{\rm i}-\varepsilon_{\rm c})\varepsilon_{\rm h}}{(\varepsilon_{\rm i}+\varepsilon_{\rm c})(\varepsilon_{\rm c}+\varepsilon_{\rm h})+(\varepsilon_{\rm i}-\varepsilon_{\rm c})(\varepsilon_{\rm c}-\varepsilon_{\rm h})\left(\frac{a}{b}\right)^2},\tag{14}$$

$$a_0=\frac{\varepsilon_{\rm i}+\varepsilon_{\rm c}}{\varepsilon_{\rm c}-\varepsilon_{\rm i}},\tag{15}$$

$$B_0 = A_0\left(a_0 + \left(\frac{a}{b}\right)^2\right) + 1. \tag{16}$$

In addition, the boundary conditions and the governing equations of the zero-order potential at first frequency ω are the same as the zero-order potential at zero frequency. Hence, we have $\Phi_0^{0\alpha} = \Phi_1^{0\alpha}$. For the zero-order potentials of the second, the third, and the higher harmonics, as their boundary-value problems correspond to the Laplace equations without source, these solutions equal zero.

At zero frequency, we get the governing equations of the first-order potentials at different frequencies,

$$\varepsilon_\alpha \nabla^2 \Phi_0^{1\alpha} + \beta_\alpha \nabla \cdot \left[\frac{5}{2}\nabla \Phi_0^{0\alpha} |\nabla \Phi_0^{0\alpha}|^2\right] = 0, \tag{17}$$

$$\varepsilon_\alpha \nabla^2 \Phi_1^{1\alpha} + \beta_\alpha \nabla \cdot \left[\frac{15}{4}\nabla \Phi_0^{0\alpha} |\nabla \Phi_0^{0\alpha}|^2\right] = 0, \tag{18}$$

$$\varepsilon_\alpha \nabla^2 \Phi_2^{1\alpha} + \beta_\alpha \nabla \cdot \left[-\frac{3}{2}\nabla \Phi_0^{0\alpha} |\nabla \Phi_0^{0\alpha}|^2\right] = 0, \tag{19}$$

$$\varepsilon_\alpha \nabla^2 \Phi_3^{1\alpha} + \beta_\alpha \nabla \cdot \left[-\frac{1}{4}\nabla \Phi_0^{0\alpha} |\nabla \Phi_0^{0\alpha}|^2\right] = 0, \tag{20}$$

where $\beta_\alpha = \chi_\alpha/\chi_{\rm h}$, $\alpha = {\rm i}, {\rm c}, {\rm h}$. The solutions of the first order potential at zero frequency in three regions are, respectively,

$$\Phi_0^{1{\rm i}}(\boldsymbol{x}) = (A_0^{1{\rm i}} r\cos\theta + A_0^{3{\rm i}} r^3\cos 3\theta)E_0^3 \quad (r \le a), \tag{21}$$

$$\begin{aligned}\Phi_0^{1{\rm c}}(\boldsymbol{x}) = {} & (A_0^{1{\rm c}} r + C_0^{1{\rm c}} r^{-1} + C_0^{2{\rm c}} a^4 r^{-3} + C_0^{3{\rm c}} a^6 r^{-5})E_0^3\cos\theta \\ & + (A_0^{3{\rm c}} r^3 + D_0^{1{\rm c}} r^{-3} + D_0^{2{\rm c}} r^{-1})E_0^3\cos 3\theta \quad (a \le r \le b),\end{aligned} \tag{22}$$

$$\Phi_0^{1{\rm h}}(\boldsymbol{x}) = (C_0^{1{\rm h}} r^{-1} + C_0^{2{\rm h}} r^{-3} + C_0^{3{\rm h}} r^{-5})E_0^3\cos\theta + (D_0^{1{\rm h}} r^{-3} + D_0^{2{\rm h}} r^{-1})E_0^3\cos 3\theta \quad (r \ge b), \tag{23}$$

In this paper, we study the effective response of nonlinear cylindrical coated composites under external AC and DC electric field, so we only give the coefficients of the potentials, which are useful for estimating the effective response:

$$A_0^{1{\rm i}} = \frac{A_0^3}{\varepsilon_{\rm i}+\varepsilon_{\rm c}}\left[2\varepsilon_{\rm c}F_1\frac{\varepsilon_{\rm c}-\varepsilon_{\rm h}}{\varepsilon_{\rm i}+\varepsilon_{\rm c}}\left(\frac{a}{b}\right)^2 - 1\right]\left[\frac{5}{2}\beta_{\rm i}(a_0+1)^3 + \beta_{\rm c}F_3\right] + \frac{2\varepsilon_{\rm c}}{\varepsilon_{\rm i}+\varepsilon_{\rm c}}F_1(F_2 + \beta_{\rm c}A_0^3F_4), \tag{24}$$

$$A_0^{1{\rm c}} = F_1\left\{F_2 + \frac{\varepsilon_{\rm c}-\varepsilon_{\rm h}}{\varepsilon_{\rm i}+\varepsilon_{\rm c}}A_0^3\left(\frac{a}{b}\right)^2\left[\frac{5}{2}\beta_{\rm i}(a_0+1)^3 + \beta_{\rm c}F_3\right]\right\} + F_1\beta_{\rm c}A_0^3F_4, \tag{25}$$

$$C_0^{2{\rm c}} = \frac{5\beta_{\rm c}A_0^3a_0}{2\varepsilon_{\rm c}}, \quad C_0^{3{\rm c}} = -\frac{5\beta_{\rm c}A_0^3}{12\varepsilon_{\rm c}}. \tag{26}$$

where

$$\begin{aligned}F_1 = {} & \frac{-(\varepsilon_{\rm i}+\varepsilon_{\rm c})}{(\varepsilon_{\rm i}+\varepsilon_{\rm c})(\varepsilon_{\rm c}+\varepsilon_{\rm h}) + (\varepsilon_{\rm i}-\varepsilon_{\rm c})(\varepsilon_{\rm c}-\varepsilon_{\rm h})\left(\frac{a}{b}\right)^2}, \\ F_2 = {} & \frac{5}{6}B_0^3 + 5B_0 + \frac{5}{2}, \quad F_3 = -\frac{5}{2}a_0^3 + 5a_0^2 + \frac{5}{6}, \\ F_4 = {} & -\frac{5}{12}\left(\frac{a}{b}\right)^6\left(1+\frac{\varepsilon_{\rm h}}{\varepsilon_{\rm c}}\right) - \frac{5}{2}a_0\left(\frac{a}{b}\right)^4\left(1-\frac{\varepsilon_{\rm h}}{\varepsilon_{\rm c}}\right) + \frac{5}{2}a_0\left(\frac{a}{b}\right)^2\left(1-\frac{\varepsilon_{\rm h}}{\varepsilon_{\rm c}}\right) \\ & -\frac{5}{12}\left(\frac{a}{b}\right)^2\left(1-\frac{\varepsilon_{\rm h}}{\varepsilon_{\rm c}}\right) - 5a_0^2\left(\frac{a}{b}\right)^2 + \frac{5}{2}a_0^3.\end{aligned}$$

As the above procedure for dealing with the first potentials at zero frequency, the first potentials at the basic frequency, the second, and the third harmonic frequencies are, respectively, as follows.

At the first potentials at the basic frequency,

$$\Phi_1^{1{\rm i}}(\boldsymbol{x}) = (A_1^{1{\rm i}} r\cos\theta + A_1^{3{\rm i}} r^3\cos 3\theta)E_0^3 \quad (r \le a), \tag{27}$$

$$\begin{aligned}\Phi_1^{1{\rm c}}(\boldsymbol{x}) = {} & (A_1^{1{\rm c}} r + C_1^{1{\rm c}} r^{-1} + C_1^{2{\rm c}} a^4 r^{-3} + C_1^{3{\rm c}} a^6 r^{-5})E_0^3\cos\theta \\ & + (A_1^{3{\rm c}} r^3 + D_1^{1{\rm c}} r^{-3} + D_1^{2{\rm c}} r^{-1})E_0^3\cos 3\theta \quad (a \le r \le b),\end{aligned} \tag{28}$$

$$\Phi_1^{1{\rm h}}(\boldsymbol{x}) = (C_1^{1{\rm h}} r^{-1} + C_1^{2{\rm h}} r^{-3} + C_1^{3{\rm h}} r^{-5})E_0^3\cos\theta + (D_1^{1{\rm h}} r^{-3} + D_1^{2{\rm h}} r^{-1})E_0^3\cos 3\theta \quad (r \ge b). \tag{29}$$

At the second harmonic frequency,

$$\Phi_2^{1\mathrm{i}}(\boldsymbol{x}) = (A_2^{1\mathrm{i}} r\cos\theta + A_2^{3\mathrm{i}} r^3\cos 3\theta)E_0^3 \quad (r \le a), \tag{30}$$

$$\Phi_2^{1\mathrm{c}}(\boldsymbol{x}) = (A_2^{1\mathrm{c}} r + C_2^{1\mathrm{c}} r^{-1} + C_2^{2\mathrm{c}} a^4 r^{-3} + C_2^{3\mathrm{c}} a^6 r^{-5})E_0^3\cos\theta + (A_2^{3\mathrm{c}} r^3 + D_2^{1\mathrm{c}} r^{-3} + D_2^{2\mathrm{c}} r^{-1})E_0^3\cos 3\theta \quad (a \le r \le b), \tag{31}$$

$$\Phi_2^{1\mathrm{h}}(\boldsymbol{x}) = (C_2^{1\mathrm{h}} r^{-1} + C_2^{2\mathrm{h}} r^{-3} + C_2^{3\mathrm{h}} r^{-5})E_0^3\cos\theta + (D_2^{1\mathrm{h}} r^{-3} + D_2^{2\mathrm{h}} r^{-1})E_0^3\cos 3\theta \quad (r \ge b). \tag{32}$$

At the third harmonic frequency,

$$\Phi_3^{1\mathrm{i}}(\boldsymbol{x}) = (A_3^{1\mathrm{i}} r\cos\theta + A_3^{3\mathrm{i}} r^3\cos 3\theta)E_0^3 \quad (r \le a), \tag{33}$$

$$\Phi_3^{1\mathrm{c}}(\boldsymbol{x}) = (A_3^{1\mathrm{c}} r + C_3^{1\mathrm{c}} r^{-1} + C_3^{2\mathrm{c}} a^4 r^{-3} + C_3^{3\mathrm{c}} a^6 r^{-5})E_0^3\cos\theta + (A_3^{3\mathrm{c}} r^3 + D_3^{1\mathrm{c}} r^{-3} + D_3^{2\mathrm{c}} r^{-1})E_0^3\cos 3\theta \quad (a \le r \le b), \tag{34}$$

$$\Phi_3^{1\mathrm{h}}(\boldsymbol{x}) = (C_3^{1\mathrm{h}} r^{-1} + C_3^{2\mathrm{h}} r^{-3} + C_3^{3\mathrm{h}} r^{-5})E_0^3\cos\theta + (D_3^{1\mathrm{h}} r^{-3} + D_3^{2\mathrm{h}} r^{-1})E_0^3\cos 3\theta \quad (r \ge b). \tag{35}$$

The coefficients are

$$C_1^{2\mathrm{c}} = \frac{3}{2}C_0^{2\mathrm{c}}, \quad C_1^{3\mathrm{c}} = \frac{3}{2}C_0^{3\mathrm{c}}, \quad A_1^{1\mathrm{i}} = \frac{3}{2}A_0^{1\mathrm{i}}, \quad A_1^{1\mathrm{c}} = \frac{3}{2}A_0^{1\mathrm{c}}. \tag{36}$$

4. Effective response

In the dilute limit, we derive the effective nonlinear response of cylindrical coated nonlinear composites under external AC and DC electric field by using the effective response method previously proposed for DC and AC response,[11−19]

$$\begin{aligned}&\frac{1}{V}\int_V [\boldsymbol{D}(\boldsymbol{x},\omega,t) - (\varepsilon_{\mathrm{h}}\boldsymbol{E}(\boldsymbol{x},\omega,t) + \chi_{\mathrm{h}}|\boldsymbol{E}(\boldsymbol{x},\omega,t)|^2\boldsymbol{E}(\boldsymbol{x},\omega,t))]\mathrm{d}v \\ &= \bar{\boldsymbol{D}}(\omega,t) - [\varepsilon_{\mathrm{h}}\bar{\boldsymbol{E}}(\omega,t) + \chi_{\mathrm{h}}|\bar{\boldsymbol{E}}(\omega,t)|^2\bar{\boldsymbol{E}}(\omega,t)],\end{aligned} \tag{37}$$

where V denotes the whole region, i.e. $V = \Omega_{\mathrm{i}} + \Omega_{\mathrm{c}} + \Omega_{\mathrm{h}}$. $\bar{\boldsymbol{D}}$ and $\bar{\boldsymbol{E}}$ are the spatial averages of electric displacement and electric field, respectively. Because the integration on the left-hand side of Eq.(37) is zero in the host region, equation (37) is simplified as

$$\begin{aligned}&\frac{1}{V}\int_{\Omega_{\mathrm{i}}+\Omega_{\mathrm{c}}} [\boldsymbol{D}(\boldsymbol{x},\omega,t) - (\varepsilon_{\mathrm{h}}\boldsymbol{E}(\boldsymbol{x},\omega,t) + \chi_{\mathrm{h}}|\boldsymbol{E}(\boldsymbol{x},\omega,t)|^2\boldsymbol{E}(\boldsymbol{x},\omega,t))]\mathrm{d}v \\ &= \bar{\boldsymbol{D}}(\omega,t) - [\varepsilon_{\mathrm{h}}\bar{\boldsymbol{E}}(\omega,t) + \chi_{\mathrm{h}}|\bar{\boldsymbol{E}}(\omega,t)|^2\bar{\boldsymbol{E}}(\omega,t)].\end{aligned} \tag{38}$$

For external AC and DC electric field, because of cubic nonlinearity of the constitutive relation, the electric displacement includes many harmonic components. Thus we can define the effective response of the composites by regarding nonlinear composites as an effective homogeneous medium,

$$\begin{aligned}\bar{\boldsymbol{D}}(\omega,t) = {}& \bar{\boldsymbol{D}}_0 + \bar{\boldsymbol{D}}_1\sin\omega t + \bar{\boldsymbol{D}}_2\cos 2\omega t \\ &+ \bar{\boldsymbol{D}}_3\sin 3\omega t + \ldots,\end{aligned} \tag{39}$$

where $\bar{\boldsymbol{D}}_n = \tilde{\varepsilon}_{n\omega}\boldsymbol{E}_{\mathrm{a}} + \tilde{\chi}_{n\omega}|\boldsymbol{E}_{\mathrm{a}}|^2\boldsymbol{E}_{\mathrm{a}} + \ldots$, $n = 0, 1, 2, 3, \ldots$; $\tilde{\varepsilon}_{n\omega}$ and $\tilde{\chi}_{n\omega}$ are the linear and the nonlinear effective dielectric response at the n-th harmonic frequency $n\omega$, respectively. The electric fields at position $\boldsymbol{x}$ are generally given by using Eq.(2),

$$\begin{aligned}&\boldsymbol{E}_\alpha(\boldsymbol{x},\omega,t) \\ &= \boldsymbol{E}_0^\alpha(\boldsymbol{x}) + \boldsymbol{E}_1^\alpha(\boldsymbol{x})\sin\omega t + \boldsymbol{E}_2^\alpha(\boldsymbol{x})\cos 2\omega t + \boldsymbol{E}_3^\alpha(\boldsymbol{x})\sin 3\omega t + \ldots \\ &= -[\nabla\Phi_0^{0\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}\nabla\Phi_0^{1\alpha}(\boldsymbol{x}) + \ldots] \\ &\quad -[\nabla\Phi_1^{0\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}\nabla\Phi_1^{1\alpha}(\boldsymbol{x}) + \ldots]\sin\omega t \\ &\quad -[\nabla\Phi_2^{0\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}\nabla\Phi_2^{1\alpha}(\boldsymbol{x}) + \ldots]\cos 2\omega t \\ &\quad -[\nabla\Phi_3^{0\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}\nabla\Phi_3^{1\alpha}(\boldsymbol{x}) + \ldots]\sin 3\omega t + \ldots.\end{aligned} \tag{40}$$

Substituting Eqs.(40) and (39) into Eq.(38), we can derive the effective responses of the nonlinear composites,

$$\tilde{\varepsilon}_0 = \varepsilon_{\mathrm{h}} - f_{\mathrm{i}}(\varepsilon_{\mathrm{i}} - \varepsilon_{\mathrm{h}})A_0(a_0 + 1) - f_{\mathrm{c}}(\varepsilon_{\mathrm{c}} - \varepsilon_{\mathrm{h}})A_0 a_0,$$

$$
\begin{aligned}
\tilde{\chi}_0 &= \frac{5}{2}\chi_{\rm h} - \frac{5}{2}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 - \frac{5}{2}f_{\rm c}(\chi_{\rm c}-\chi_{\rm h})A_0^3\left(a_0^3+2a_0\frac{a^2}{b^2}\right) - \chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_0^{1{\rm i}} \\
&\quad -\chi_{\rm h}f_{\rm c}(\varepsilon_{\rm c}-\varepsilon_{\rm h})\left(A_0^{1{\rm c}} - \frac{a^2}{b^2}C_0^{2{\rm c}} - \frac{a^2}{b^2}\left(1+\frac{a^2}{b^2}\right)C_0^{3{\rm c}}\right), \\
\tilde{\varepsilon}_\omega &= \varepsilon_{\rm h} - f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_0(a_0+1) - f_{\rm c}(\varepsilon_{\rm c}-\varepsilon_{\rm h})A_0a_0, \\
\tilde{\chi}_\omega &= \frac{15}{4}\chi_{\rm h} - \frac{15}{4}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 - \frac{15}{4}f_{\rm c}(\chi_{\rm c}-\chi_{\rm h})A_0^3\left(a_0^3+2a_0\frac{a^2}{b^2}\right) - \chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_1^{1{\rm i}} \\
&\quad -\chi_{\rm h}f_{\rm c}(\varepsilon_{\rm c}-\varepsilon_{\rm h})\left(A_1^{1{\rm c}} - \frac{a^2}{b^2}C_1^{2{\rm c}} - \frac{a^2}{b^2}\left(1+\frac{a^2}{b^2}\right)C_1^{3{\rm c}}\right), \\
\tilde{\varepsilon}_{2\omega} &= 0, \\
\tilde{\chi}_{2\omega} &= -\frac{3}{2}\chi_{\rm h} + \frac{3}{2}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 + \frac{3}{2}f_{\rm c}(\chi_{\rm c}-\chi_{\rm h})A_0^3\left(a_0^3+2a_0\frac{a^2}{b^2}\right) - \chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_2^{1{\rm i}} \\
&\quad -\chi_{\rm h}f_{\rm c}(\varepsilon_{\rm c}-\varepsilon_{\rm h})\left(A_2^{1{\rm c}} - \frac{a^2}{b^2}C_2^{2{\rm c}} - \frac{a^2}{b^2}\left(1+\frac{a^2}{b^2}\right)C_2^{3{\rm c}}\right), \\
\tilde{\varepsilon}_{3\omega} &= 0, \\
\tilde{\chi}_{3\omega} &= -\frac{1}{4}\chi_{\rm h} + \frac{1}{4}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 + \frac{1}{4}f_{\rm c}(\chi_{\rm c}-\chi_{\rm h})A_0^3\left(a_0^3+2a_0\frac{a^2}{b^2}\right) - \chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_3^{1{\rm i}} \\
&\quad -\chi_{\rm h}f_{\rm c}(\varepsilon_{\rm c}-\varepsilon_{\rm h})\left(A_3^{1{\rm c}} - \frac{a^2}{b^2}C_3^{2{\rm c}} - \frac{a^2}{b^2}\left(1+\frac{a^2}{b^2}\right)C_3^{3{\rm c}}\right),
\end{aligned}
$$

where $f_{\rm i}$ and $f_{\rm c}$ are the volume fraction of the inclusion core and the coated shell, respectively. The quantity a^2/b^2 can be expressed by the volume fraction, $a^2/b^2 = f_{\rm i}/(f_{\rm i}+f_{\rm c})$.

Considering the coefficients appearing in the effective response, we obtain these relations, $\tilde{\varepsilon}_\omega = \tilde{\varepsilon}_0$, $\tilde{\varepsilon}_{2\omega} = \tilde{\varepsilon}_{3\omega}$, $\tilde{\chi}_\omega = \frac{3}{2}\tilde{\chi}_0$, $\tilde{\chi}_{2\omega} = -\frac{3}{5}\tilde{\chi}_0$, $\tilde{\chi}_{3\omega} = -\frac{1}{10}\tilde{\chi}_0$. Clearly, with these relations we can get a comprehensive understanding of the effective nonlinear response of the nonlinear composites.

5. Conclusion

In this paper, an effective response of nonlinear coated composites, which randomly embedded in a host medium, is analysed under the external field $\boldsymbol{E}_{\rm app} = \boldsymbol{E}_{\rm a}(1+\sin\omega t)$ by using the perturbation method, and the formulae of the effective responses at harmonic frequencies are obtained in the dilute limit. In addition, we can derive the effective nonlinear response of the cylinder nonlinear composites without the coated shell from our results. In this case we assume that $\varepsilon_{\rm c} = \varepsilon_{\rm h}$ and $\chi_{\rm c} = \chi_{\rm h}$. Under this condition, the effective responses of the cylinder nonlinear composites are

$$
\begin{aligned}
\tilde{\varepsilon}_0 &= \varepsilon_{\rm h} - f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_0(a_0+1), \\
\tilde{\chi}_0 &= \frac{5}{2}\chi_{\rm h} - \frac{5}{2}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 \\
&\quad -\chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_0^{1{\rm i}}, \\
\tilde{\varepsilon}_\omega &= \varepsilon_{\rm h} - f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_0(a_0+1), \\
\tilde{\chi}_\omega &= \frac{15}{4}\chi_{\rm h} - \frac{15}{4}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 \\
&\quad -\chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_1^{1{\rm i}}, \\
\tilde{\varepsilon}_{2\omega} &= 0, \\
\tilde{\chi}_{2\omega} &= -\frac{3}{2}\chi_{\rm h} + \frac{3}{2}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 \\
&\quad -\chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_2^{1{\rm i}}, \\
\tilde{\varepsilon}_{3\omega} &= 0, \\
\tilde{\chi}_{3\omega} &= -\frac{1}{4}\chi_{\rm h} + \frac{1}{4}f_{\rm i}(\chi_{\rm i}-\chi_{\rm h})A_0^3(a_0+1)^3 \\
&\quad -\chi_{\rm h}f_{\rm i}(\varepsilon_{\rm i}-\varepsilon_{\rm h})A_3^{1{\rm i}},
\end{aligned} \tag{41}
$$

where $A_0(a_0+1) = -c, B_0 = -b_0, A_0^{1{\rm i}} = A_{1,0}^{{\rm i},1}, A_1^{1{\rm i}} = \frac{3}{2}A_0^{1{\rm i}}, A_2^{1{\rm i}} = -\frac{3}{5}A_0^{1{\rm i}}$, and $A_3^{1{\rm i}} = -\frac{1}{10}A_0^{1{\rm i}}$, if $\varepsilon_{\rm c} = \varepsilon_{\rm h}$ and $\chi_{\rm c} = \chi_{\rm h}$. The coefficients $c, b_0, A_{1,0}^{{\rm i},1}$ are given in Ref.[19]. So the result of Ref.[19] is a special case of our work. Moreover, we can use the method to deal with the effective responses of the nonlinear spherical coated composites under external AC and DC electric field.

References

[1] Stroud D and Hui P M 1988 *Phys. Rev.* B **37** 8719
[2] Blumenfeld R and Bergman D 1991 *Phys. Rev.* B **44** 7378
[3] Castameda P P, deBotton B and Li G 1992 *Phys. Rev.* B **46** 4387
[4] Levy O, Bergman D J and Stroud D G 1995 *Phys. Rev.* E **52** 3184
[5] Niu J S, Luo Y and Ma B K 2002 *Chin. Phys.* **11** 144
[6] Gu Y and Yu K W 2002 *Chin. Phys.* **11** 601
[7] Levy O and Bergman D J 1993 *J. Phys.*: *Condens. Matter* **5** 7095
[8] Joseph W H, Inguva R and Bowden C M 1989 *Phys. Rev.* A **40** 5729
[9] Castaneda P P 1997 *Phys. Lett.* A **224** 163
[10] Hui P M, Cheung P and Stroud D 1998 *J. Appl. Phys.* **84** 3451
[11] Gu G Q and Yu K W 1992 *Phys. Rev.* B **46** 4502
[12] Gu G Q, Hui P M and Yu K W 2000 *Physica* B **279** 62
[13] Wei E B, Song J B and Gu G Q 2002 *Physica* B **324** 322
[14] Wei E B, Song J B, Tian J W and Gu G Q 2003 *Phys. Lett.* A **309** 160
[15] Chen X G, Wei E B and Song J B 2004 *Commun. Theor. Phys.* **41** 77
[16] Wei E B, Tian J W and Song J B 2004 *Chin. Phys.* **13** 388
[17] Wei E B, Yang Z D and Gu G Q 2004 *J. Phys.* D: *Appl. Phys.* **37** 107
[18] Chen X G, Liang F C and Wei E B 2005 *Chin. Phys.* **14** 1217
[19] Wei E B, Song J B and Gu G Q 2004 *J. Appl. Phys.* **95** 1377