Effective AC response of a nonlinear spherical coated composite*

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An effective nonlinear response of a nonlinear composite with spherical coated inclusions randomly embedded in a host medium under the action of an external AC electric field, $\boldsymbol{E}_a = \boldsymbol{E}_1 \sin(\omega t) + \boldsymbol{E}_3 \sin(3\omega t)$, is investigated using a perturbation method. The local potentials of the composite at higher harmonics are given both in the region of local inclusion particles and in the local host region under the external AC electric field. All effective nonlinear responses of the composite and the relationship between the effective nonlinear responses at the fundamental frequency and third harmonics are also studied for spherical coated inclusion in a dilute limit.

Keywords: nonlinear composite, effective nonlinear response, coated composite, AC electric field

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1. Introduction

Recently, many researchers have studied the effective responses of nonlinear composites under different external electric fields, and they pointed out that the effective responses of composites were dependent not only on the physical properties and the volume fraction of inclusions, but also on the particle shape and the applied external field.[1−5] The studies also showed that if an external alternating current (AC) electric field was applied to a nonlinear composite, there were many components of local field at harmonics when the constitutive relations of composite were nonlinear.[6] For example, Levy *et al.*[2,6] and Hui *et al.*[7] derived the high order effective dielectric responses of nonlinear composites under an external electric field. Yu and Wang *et al.*[8] discussed the effective conductivity of a nonlinear composite of spherical particles using a perturbation approach. Gu *et al.*[9] and Wei *et al.*[10−12] presented the nonlinear effective response of a weakly cubic nonlinear composite at harmonic frequencies of external AC electric field. Gu *et al.*[13] considered the effect of nonhomogeneity on conductivity of a nonlinear composite medium. Gu and Yu[14] studied the effective conductivity of the composite medium with layered inclusions. Chen and Wei *et al.*[15] investigated the effect of an applied AC electric field on the effective response of a nonlinear spherical coated composite. Chen *et al.*[16] obtained the effective nonlinear AC response of a composite with spherical particles under the action of an external sinusoidal field, $\boldsymbol{E}_a = \boldsymbol{E}_1 \sin(\omega t) + \boldsymbol{E}_3 \sin(3\omega t)$. Shen *et al.*[17,18] dealt with the effective response of a nonlinear composite under the joint action of external AC and DC electric fields $\boldsymbol{E}_{\rm app} = \boldsymbol{E}_a(1 + \sin(\omega t))$, and so on.

In this paper, we study the effective response of a nonlinear spherical coated composite under an external AC field

$$\boldsymbol{E}_a = \boldsymbol{E}_1 \sin(\omega t) + \boldsymbol{E}_3 \sin(3\omega t). \tag{1}$$

Compared with the structures studied in Refs. [15]–[18], here the structure of the nonlinear composite is very complicated, and the third harmonic component is contained in the external sinusoidal field, so the effective responses will contain the nonlinear interactions between the fundamental wave and the third harmonic component of the external AC electric field, and between the two inclusion layers, i.e.

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the inclusion core particle and the coated shell layer. Furthermore, the case under our consideration will be closer to the circumstances of the practical application.

Now the composite is assumed to be homogenous and isotropic granular and coated spherical particles are randomly embedded in a host medium. The constitutive relations between current density $\boldsymbol{J}$ and electric field $\boldsymbol{E}$ in the inclusion and the host regions have the following forms:

$$\boldsymbol{J}^{\alpha} = \sigma_{\alpha}\boldsymbol{E} + \chi_{\alpha}|\boldsymbol{E}|^2\boldsymbol{E}, \quad \text{in} \quad \Omega_{\alpha}, \alpha = \mathrm{i,c,h}, \tag{2}$$

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. The symbol Ω_{α} denotes the region occupied by the α-type material. The real linear and real nonlinear dielectric constants, σ_{α} and χ_{α}, are independent of the frequency of applied field, and the corresponding governing equations satisfy $\boldsymbol{\nabla}\cdot\boldsymbol{J} = 0$ and $\boldsymbol{\nabla}\times\boldsymbol{E} = 0$, which are the reasonable assumptions for a low-frequency process. The boundary conditions of the coated composite are the continuities of the potentials and the normal components of the current density on the surface of the core, coated layer and host. At infinity, the potential must match that of the external applied field.

2. Perturbation method of the nonlinear AC process

We consider weakly nonlinear composites with coated spherical inclusions randomly embedded in a host, in which the nonlinear terms in the constitutive equations are smaller than the linear terms. We assume that the concentric spherical particle has a core with radius a and coated shell radius b. For the external field

$$\boldsymbol{E}_a = \boldsymbol{E}_1\sin(\omega t) + \boldsymbol{E}_3\sin(3\omega t), \tag{3}$$

which is applied to the nonlinear composites along the $\hat{z}$ direction, the electric potential at point $\boldsymbol{x}$ can be written as

$$\begin{aligned}&\Phi^{\alpha}(\boldsymbol{x},t)\\ &= \Phi_1^{\alpha}(\boldsymbol{x})\sin(\omega t) + \Phi_3^{\alpha}(\boldsymbol{x})\sin(3\omega t)\\ &\quad + \cdots \Phi_5^{\alpha}(\boldsymbol{x})\sin(5\omega t) + \ldots, \text{in } \Omega_{\alpha}, \alpha = \mathrm{i,c,h},\end{aligned} \tag{4}$$

where $\boldsymbol{E}_1 = \varepsilon_1\hat{z}$ and $\boldsymbol{E}_3 = \varepsilon_3\hat{z}$.

For weakly nonlinear composites, we can use the perturbation method to deal with the potentials.[19] The nonlinear susceptibility χ_{h} in the host medium is chosen as the perturbation parameter, so the potentials $\Phi_s^{\alpha}(\boldsymbol{x})$ can be expanded with parameter χ_{h} into the following forms:

$$\begin{aligned}&\Phi_s^{\alpha}(\boldsymbol{x})\\ &= \Phi_s^{0\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}\Phi_s^{1\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}^2\Phi_s^{2\alpha}(\boldsymbol{x}) + \cdots\\ &\quad \text{in} \quad \Omega_{\alpha}, \alpha = \mathrm{i,c,h}, \quad s = 1,3,5,\ldots.\end{aligned} \tag{5}$$

Using the electric field equation $\boldsymbol{E}_{\alpha} = -\nabla\Phi_{\alpha}(\boldsymbol{x},t)$ and substituting potentials (4) and (5) into the current density (2), we obtain perturbation expansions for $\boldsymbol{J}$ in the inclusion and host regions:

$$\begin{aligned}&\boldsymbol{J}_s^{\alpha}(\boldsymbol{x})\\ &= \boldsymbol{J}_s^{0\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}\boldsymbol{J}_s^{1\alpha}(\boldsymbol{x}) + \chi_{\mathrm{h}}^2\boldsymbol{J}_s^{2\alpha}(\boldsymbol{x}) + \cdots,\\ &\quad \text{in} \quad \Omega_{\alpha}, \alpha = \mathrm{i,c,h}, \quad s = 1,3,5,\ldots,\end{aligned} \tag{6}$$

where $\boldsymbol{J}_s^{k\alpha}(\boldsymbol{x})$ is a function of potentials $\Phi_s^{k\alpha}(\boldsymbol{x})$.

The governing equation

$$\boldsymbol{\nabla}\cdot\boldsymbol{J}(\boldsymbol{x},t) = 0$$

will be satisfied if each term in Eq. (6) satisfies its corresponding governing equation. Therefore we obtain a set of equations as follows:

$$\begin{aligned}&\boldsymbol{\nabla}\cdot\boldsymbol{J}_s^{k\alpha}(\boldsymbol{x}) = 0,\\ &\text{in} \quad \Omega_{\alpha}, \alpha = \mathrm{i,c,h}, \ s = 1,3,5,\ldots,\\ &\quad k = 0,1,2,3,\ldots.\end{aligned} \tag{7}$$

The boundary condition of the potentials and the current density are, respectively as follows.

1) At the interface between the inclusion core and inside layer of the coated shell, $\partial\Omega_{\mathrm{i}}$,

$$\begin{aligned}&\Phi_s^{k\mathrm{i}}(\boldsymbol{x}) = \Phi_s^{k\mathrm{c}}(\boldsymbol{x}), \quad \hat{\boldsymbol{n}}\cdot\boldsymbol{J}_s^{k\mathrm{i}}(\boldsymbol{x}) = \hat{\boldsymbol{n}}\cdot\boldsymbol{J}_s^{k\mathrm{c}}(\boldsymbol{x}),\\ &\text{on} \quad \partial\Omega_{\mathrm{i}}, s = 1,3,5,\ldots, \quad k = 0,1,2,3,\ldots,\end{aligned} \tag{8}$$

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

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

$$\begin{aligned}&\Phi_s^{k\mathrm{c}}(\boldsymbol{x}) = \Phi_s^{k\mathrm{h}}(\boldsymbol{x}), \quad \hat{\boldsymbol{n}}\cdot\boldsymbol{J}_s^{k\mathrm{c}}(\boldsymbol{x}) = \hat{\boldsymbol{n}}\cdot\boldsymbol{J}_s^{k\mathrm{h}}(\boldsymbol{x}),\\ &\text{on} \quad \partial\Omega_{\mathrm{c}}, s = 1,3,5,\ldots, k = 0,1,2,3,\ldots.\end{aligned} \tag{9}$$

3) At infinity

$$\left.\frac{\partial\Phi_1^{k\mathrm{h}}}{\partial x}\right|_{x\to\infty} = \begin{cases}-\varepsilon_1, & \text{if} \quad k = 0,\\ 0, & \text{if} \quad k \neq 0;\end{cases} \tag{10}$$

$$\left.\frac{\partial\Phi_3^{k\mathrm{h}}}{\partial x}\right|_{x\to\infty} = \begin{cases}-\varepsilon_3, & \text{if} \quad k = 0,\\ 0, & \text{if} \quad k \neq 0.\end{cases} \tag{11}$$

3. Potential in coated nonlinear spherical composites

From Eqs. (6) and (7), we obtain the governing equations of the zeroth-order potentials at frequencies ω and 3ω as follows:

$$\sigma_{\rm i}\nabla^2\Phi_s^{0\rm i}=0, \qquad s=1,3, \quad \text{in} \quad \Omega_{\rm i}, \tag{12}$$

$$\sigma_{\rm c}\nabla^2\Phi_s^{0\rm c}=0, \qquad s=1,3, \quad \text{in} \quad \Omega_{\rm c}, \tag{13}$$

$$\sigma_{\rm h}\nabla^2\Phi_s^{0\rm h}=0, \qquad s=1,3, \quad \text{in} \quad \Omega_{\rm h}. \tag{14}$$

With boundary conditions (8)–(11), the zeroth-order potentials in the inclusion and host regions can be derived from governing equations (12)–(14). At the fundamental frequency and the third harmonic frequency, the zeroth-order potentials are respectively

$$\begin{aligned}&\Phi_1^{0\rm i}(r,\theta)\\ &=-A_0\varepsilon_1 r\mathrm{P}_1(\cos\theta), \qquad (r\le a),\end{aligned} \tag{15}$$

$$\begin{aligned}&\Phi_1^{0\rm c}(r,\theta)\\ &=-(B_0r+Cr^{-2})\varepsilon_1\mathrm{P}_1(\cos\theta), \ (a\le r\le b),\end{aligned} \tag{16}$$

$$\begin{aligned}&\Phi_1^{0\rm h}(r,\theta)\\ &=-(r+Dr^{-2})\varepsilon_1\mathrm{P}_1(\cos\theta), \qquad (r\ge b),\end{aligned} \tag{17}$$

and

$$\begin{aligned}&\Phi_3^{0\rm i}(r,\theta)\\ &=-A_0\varepsilon_3 r\mathrm{P}_1(\cos\theta), \qquad (r\le a),\end{aligned} \tag{18}$$

$$\begin{aligned}&\Phi_3^{0\rm c}(r,\theta)\\ &=-(B_0r+Cr^{-2})\varepsilon_3\mathrm{P}_1(\cos\theta), \ (a\le r\le b),\end{aligned} \tag{19}$$

$$\begin{aligned}&\Phi_3^{0\rm h}(r,\theta)\\ &=-(r+Dr^{-2})\varepsilon_3\mathrm{P}_1(\cos\theta), \qquad (r\ge b),\end{aligned} \tag{20}$$

where $\mathrm{P}_1(\cos\theta)=\cos\theta$ is the first-order Legendre function, and the coefficients are

$$A_0=\frac{9\sigma_{\rm c}\sigma_{\rm h}}{(\sigma_{\rm i}+2\sigma_{\rm c})(\sigma_{\rm c}+2\sigma_{\rm h})+2(\sigma_{\rm i}-\sigma_{\rm c})(\sigma_{\rm c}-\sigma_{\rm h})(a/b)^3}, \tag{21}$$

$$B_0=\frac{3\sigma_{\rm h}(\sigma_{\rm i}+2\sigma_{\rm c})}{(\sigma_{\rm i}+2\sigma_{\rm c})(\sigma_{\rm c}+2\sigma_{\rm h})+2(\sigma_{\rm i}-\sigma_{\rm c})(\sigma_{\rm c}-\sigma_{\rm h})(a/b)^3}, \tag{22}$$

$$C=a^3\frac{3\sigma_{\rm h}(\sigma_{\rm c}-\sigma_{\rm i})}{(\sigma_{\rm i}+2\sigma_{\rm c})(\sigma_{\rm c}+2\sigma_{\rm h})+2(\sigma_{\rm i}-\sigma_{\rm c})(\sigma_{\rm c}-\sigma_{\rm h})(a/b)^3}\triangleq a^3C_0, \tag{23}$$

$$D=b^3\frac{(\sigma_{\rm i}+2\sigma_{\rm c})(\sigma_{\rm h}-\sigma_{\rm c})+(\sigma_{\rm c}-\sigma_{\rm i})(2\sigma_{\rm c}+\sigma_{\rm h})(a/b)^3}{(\sigma_{\rm i}+2\sigma_{\rm c})(\sigma_{\rm c}+2\sigma_{\rm h})+2(\sigma_{\rm i}-\sigma_{\rm c})(\sigma_{\rm c}-\sigma_{\rm h})(a/b)^3}\triangleq b^3D_0. \tag{24}$$

With the zeroth-order potentials at the fundamental frequency and the third harmonics, the governing equations of the first-order potentials both at the fundamental frequency and third harmonics take the following forms:

$$\begin{aligned}\sigma_\alpha\nabla^2\Phi_1^{1\alpha}=&-\beta_\alpha\nabla\left[\nabla\Phi_1^{0\alpha}\left\{\frac{3}{4}|\nabla\Phi_1^{0\alpha}|^2-\frac{1}{2}(\nabla\Phi_1^{0\alpha}\cdot\nabla\Phi_3^{0\alpha})+\frac{1}{2}|\nabla\Phi_3^{0\alpha}|^2\right\}\right.\\ &\left.+\nabla\Phi_3^{0\alpha}\left\{-\frac{1}{4}|\nabla\Phi_1^{0\alpha}|^2+(\nabla\Phi_1^{0\alpha}\cdot\nabla\Phi_3^{0\alpha})\right\}\right],\end{aligned} \tag{25}$$

and

$$\sigma_\alpha\nabla^2\Phi_3^{1\alpha}=-\beta_\alpha\nabla\left[\nabla\Phi_1^{0\alpha}\left\{-\frac{1}{4}|\nabla\Phi_1^{0\alpha}|^2+(\nabla\Phi_1^{0\alpha}\cdot\nabla\Phi_3^{0\alpha})\right\}+\nabla\Phi_3^{0\alpha}\left\{\frac{1}{2}|\nabla\Phi_1^{0\alpha}|^2+\frac{3}{4}|\nabla\Phi_3^{0\alpha}|^2\right\}\right], \tag{26}$$

where $\beta_\alpha=\chi_\alpha/\chi_{\rm h}$ and $\alpha=$ i, c, h.

In the core, coated layer and host regions, the general solutions of the first-order potentials at the fundamental frequency and the third harmonics can be exactly solved with the boundary conditions. The solutions are

$$\Phi_1^{1\rm i}(r,\theta)=[A_1^{1\rm i}r\mathrm{P}_1(\cos\theta)+A_1^{3\rm i}r^3\mathrm{P}_3(\cos\theta)](\varepsilon_1^3-\varepsilon_1^2\varepsilon_3+2\varepsilon_1\varepsilon_3^2), \qquad (r\le a), \tag{27}$$

$$\begin{aligned}\Phi_1^{1\rm c}(r,\theta)=&[(A_1^{1\rm c}r+C_1^{1\rm c}r^{-2}+C_1^{2\rm c}a^6r^{-5}+C_1^{3\rm c}a^9r^{-8})\mathrm{P}_1(\cos\theta)+(A_1^{3\rm c}r^3+D_1^{1\rm c}r^{-2}\\ &+D_1^{2\rm c}r^{-4}+D_1^{3\rm c}r^{-5}+D_1^{4\rm c}r^{-8})\mathrm{P}_3(\cos\theta)](\varepsilon_1^3-\varepsilon_1^2\varepsilon_3+2\varepsilon_1\varepsilon_3^2), \quad (a\le r\le b),\end{aligned} \tag{28}$$

$$\begin{aligned}\Phi_1^{1\rm h}(r,\theta)=&[(C_1^{1\rm h}r^{-2}+C_1^{2\rm h}r^{-5}+C_1^{3\rm h}r^{-8})\mathrm{P}_1(\cos\theta)+(D_1^{1\rm h}r^{-2}+D_1^{2\rm h}r^{-4}+D_1^{3\rm h}r^{-5}\\ &+D_1^{4\rm h}r^{-8})\mathrm{P}_3(\cos\theta)](\varepsilon_1^3-\varepsilon_1^2\varepsilon_3+2\varepsilon_1\varepsilon_3^2), \quad (r\ge b),\end{aligned} \tag{29}$$

$$\Phi_3^{1\mathrm{i}}(r,\theta) = [A_3^{1\mathrm{i}} r \mathrm{P}_1(\cos\theta) + A_3^{3\mathrm{i}} r^3 \mathrm{P}_3(\cos\theta)](\varepsilon_1^3 - 6\varepsilon_1^2\varepsilon_3 - 3\varepsilon_3^3), \qquad (r \le a), \tag{30}$$

$$\begin{aligned}\Phi_3^{1\mathrm{c}}(r,\theta) = {} & [(A_3^{1\mathrm{c}} r + C_3^{1\mathrm{c}} r^{-2} + C_3^{2\mathrm{c}} a^6 r^{-5} + C_3^{3\mathrm{c}} a^9 r^{-8})\mathrm{P}_1(\cos\theta) + (A_3^{3\mathrm{c}} r^3 + D_3^{1\mathrm{c}} r^{-2} \\ & + D_3^{2\mathrm{c}} r^{-4} + D_3^{3\mathrm{c}} r^{-5} + D_3^{4\mathrm{c}} r^{-8})\mathrm{P}_3(\cos\theta)](\varepsilon_1^3 - 6\varepsilon_1^2\varepsilon_3 - 3\varepsilon_3^3), \quad (a \le r \le b),\end{aligned} \tag{31}$$

$$\begin{aligned}\Phi_3^{1\mathrm{h}}(r,\theta) = {} & [(C_3^{1\mathrm{h}} r^{-2} + C_3^{2\mathrm{h}} r^{-5} + C_3^{3\mathrm{h}} r^{-8})\mathrm{P}_1(\cos\theta) + (D_3^{1\mathrm{h}} r^{-2} + D_3^{2\mathrm{h}} r^{-4} \\ & + D_3^{3\mathrm{h}} r^{-5} + D_3^{4\mathrm{h}} r^{-8})\mathrm{P}_3(\cos\theta)](\varepsilon_1^3 - 6\varepsilon_1^2\varepsilon_3 - 3\varepsilon_3^3), \quad (r \ge b),\end{aligned} \tag{32}$$

where $\mathrm{P}_3(\cos\theta) = (5\cos 3\theta + 3\cos\theta)/8$ is the third-order Legendre function.

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

$$A_1^{1\mathrm{i}} = \frac{(3/4)\beta_\mathrm{i} A_0^3 (F_1\sigma_\mathrm{c} + 2F_2\sigma_\mathrm{h}) + 3\beta_\mathrm{c} F_2 (F_3 B_0^3 + F_4 B_0^2 C_0 + F_5 B_0 C_0^2 + F_6 C_0^3) + 3\sigma_\mathrm{c} F_7}{(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})(a/b)^3}, \tag{33}$$

$$A_1^{1\mathrm{c}} = \frac{(3/4)\beta_\mathrm{i} A_0^3 G_1 + 3\beta_\mathrm{c}(G_2 B_0^3 - G_3 B_0^2 C_0 + G_4 B_0 C_0^2 - G_5 C_0^3) + 3\sigma_\mathrm{c}(\sigma_\mathrm{i} + 2\sigma_\mathrm{c}) G_6}{\sigma_\mathrm{c}[(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})(a/b)^3]}, \tag{34}$$

$$C_1^{2\mathrm{c}} = -\frac{6\beta_\mathrm{c}}{5\sigma_\mathrm{c}} B_0 C_0^2, \quad C_1^{3\mathrm{c}} = \frac{\beta_\mathrm{c}}{2\sigma_\mathrm{c}} C_0^3, \quad A_3^{1\mathrm{i}} = -\frac{1}{3} A_1^{1\mathrm{i}}, \quad A_3^{1\mathrm{c}} = -\frac{1}{3} A_1^{1\mathrm{c}}, \tag{35}$$

$$C_3^{2\mathrm{c}} = -\frac{1}{3} C_1^{2\mathrm{c}}, \quad C_3^{3\mathrm{c}} = -\frac{1}{3} C_1^{3\mathrm{c}}, \tag{36}$$

where

$$\begin{aligned}
F_1 &= 1 + 2\left(\frac{a}{b}\right)^3, \quad F_2 = 1 - \left(\frac{a}{b}\right)^3, \quad F_3 = \frac{1}{2}(\sigma_\mathrm{c} - \sigma_\mathrm{h}), \quad F_4 = \frac{9}{10}(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}), \\
F_5 &= \frac{3}{5}\left\{\frac{1}{2}\left[5\left(\frac{a}{b}\right)^3 - 1\right]\sigma_\mathrm{c} - \left[4\left(\frac{a}{b}\right)^3 + 1\right]\sigma_\mathrm{h}\right\}, \\
F_6 &= \frac{1}{5}\left\{\left[\left(\frac{a}{b}\right)^6 + \left(\frac{a}{b}\right)^3 + 2\right]\sigma_\mathrm{c} + \left[5\left(\frac{a}{b}\right)^6 + 5\left(\frac{a}{b}\right)^3 + 4\right]\sigma_\mathrm{h}\right\}, \\
F_7 &= -\frac{3}{4} + \frac{27}{10} D_0 - \frac{9}{10} D_0^2 + \frac{6}{5} D_0^3, \quad G_1 = 2\sigma_\mathrm{c}(\sigma_\mathrm{c} - \sigma_\mathrm{h})\left(\frac{a}{b}\right)^3, \\
G_2 &= \frac{1}{4}\left[\sigma_\mathrm{c}(\sigma_\mathrm{i} + 2\sigma_\mathrm{c}) + 2\sigma_\mathrm{c}(\sigma_\mathrm{h} - \sigma_\mathrm{c})\left(\frac{a}{b}\right)^3\right], \quad G_3 = \frac{9}{10}\sigma_\mathrm{c}(\sigma_\mathrm{i} + 2\sigma_\mathrm{h})\left(\frac{a}{b}\right)^3, \\
G_4 &= \frac{1}{5}(\sigma_\mathrm{c} - \sigma_\mathrm{h})(4\sigma_\mathrm{i} + 5\sigma_\mathrm{c})\left(\frac{a}{b}\right)^3 + \frac{1}{10}(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(8\sigma_\mathrm{h} - 5\sigma_\mathrm{c})\left(\frac{a}{b}\right)^6, \\
G_5 &= \frac{1}{15}\left[(\sigma_\mathrm{c} - \sigma_\mathrm{h})(5\sigma_\mathrm{i} - 2\sigma_\mathrm{c})\left(\frac{a}{b}\right)^3 + (\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(5\sigma_\mathrm{h} + \sigma_\mathrm{c})\left(\frac{a}{b}\right)^9\right], \\
G_6 &= -\frac{1}{4} + \frac{9}{10} D_0 - \frac{3}{10} D_0^2 + \frac{2}{5} D_0^3.
\end{aligned}$$

4. Effective response

The general formula for effective conductivity of a weakly nonlinear composite was formulated in Ref. [13]. In the present paper we deal with the effective nonlinear AC response of a spherical coated composite in the dilute limit; thus we use the method previously proposed for DC nonlinear response,[19]

$$\frac{1}{V}\int_V [\boldsymbol{J}(\boldsymbol{x},t) - (\sigma_\mathrm{h}\boldsymbol{E}(\boldsymbol{x},t) + \chi_\mathrm{h}|\boldsymbol{E}(\boldsymbol{x},t)|^2\boldsymbol{E}(\boldsymbol{x},t)]\,\mathrm{d}V = \overline{\boldsymbol{J}}(t) - [\sigma_\mathrm{h}\overline{\boldsymbol{E}}(t) + \chi_\mathrm{h}|\overline{\boldsymbol{E}}(t)|^2], \tag{37}$$

where V denotes the volume of composite $V = \Omega_\mathrm{i} + \Omega_\mathrm{c} + \Omega_\mathrm{h}$. $\overline{\boldsymbol{J}}(t)$ and $\overline{\boldsymbol{E}}(\mathrm{t})$ are the spatially averaged current density and electric field strength. Because the value of integration on the left-hand side of Eq. (37) is zero in the host region, equation (37) is simplified to

$$\frac{1}{V}\int_{\Omega_\mathrm{i}+\Omega_\mathrm{c}} [\boldsymbol{J}(\boldsymbol{x},t) - \sigma_\mathrm{h}\boldsymbol{E}(\boldsymbol{x},t) + \chi_\mathrm{h}|\boldsymbol{E}(\boldsymbol{x},t)|^2\boldsymbol{E}(\boldsymbol{x},t)]\,\mathrm{d}V$$

$$= \overline{\boldsymbol{J}}(\mathrm{t}) - \left[\left\{\sigma_{\mathrm{h}}\overline{\boldsymbol{E}}_1 + \chi_{\mathrm{h}}\left[\frac{3}{4}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_1 - \frac{1}{2}(\overline{\boldsymbol{E}}_1\cdot\overline{\boldsymbol{E}}_3)\overline{\boldsymbol{E}}_1 + \frac{1}{2}|\overline{\boldsymbol{E}}_3|^2\overline{\boldsymbol{E}}_1 - \frac{1}{4}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_3 + (\overline{\boldsymbol{E}}_1\cdot\overline{\boldsymbol{E}}_3)\overline{\boldsymbol{E}}_3\right]\right\}\sin\omega t$$
$$+ \left\{\sigma_{\mathrm{h}}\overline{\boldsymbol{E}}_3 + \chi_{\mathrm{h}}\left[-\frac{1}{4}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_1 + (\overline{\boldsymbol{E}}_1\cdot\overline{\boldsymbol{E}}_3)\overline{\boldsymbol{E}}_3 + \frac{1}{2}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_3 + \frac{3}{4}|\overline{\boldsymbol{E}}_3|^2\overline{\boldsymbol{E}}_3\right]\right\}\sin(3\omega t) + \ldots\Big]. \tag{38}$$

Since the constitutive relation is cubic nonlinear, under the external AC electric field $\boldsymbol{E}_1\sin(\omega t)+\boldsymbol{E}_3\sin(3\omega t)$, the current density includes many harmonic components. Thus the effective AC response of composite can be defined as follows:

$$\begin{aligned}\overline{\boldsymbol{J}}(t) &= \overline{\boldsymbol{J}}_{\omega}\sin\omega t + \overline{\boldsymbol{J}}_{3\omega}\sin 3\omega t + \ldots\\ &= [\sigma^*_{\omega,\omega}\overline{\boldsymbol{E}}_1 + \sigma^*_{3\omega,\omega}\overline{\boldsymbol{E}}_3 + \chi^*_{\omega^3,\omega}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_1 + \chi^*_{\omega^2(3\omega),\omega}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_3 + \chi^*_{\omega(3\omega),\omega}|\overline{\boldsymbol{E}}_3|^2\overline{\boldsymbol{E}}_1\\ &\quad + \chi^*_{(3\omega)^3,\omega}|\overline{\boldsymbol{E}}_3|^2\overline{\boldsymbol{E}}_3 + \ldots]\sin\omega t + [\sigma^*_{\omega,3\omega}\overline{\boldsymbol{E}}_1 + \sigma^*_{3\omega,3\omega}\overline{\boldsymbol{E}}_3 + \chi^*_{\omega^3,,3\omega}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_1\\ &\quad + \chi^*_{\omega^2(3\omega),3\omega}|\overline{\boldsymbol{E}}_1|^2\overline{\boldsymbol{E}}_3 + \chi^*_{\omega(3\omega)^2,3\omega}|\overline{\boldsymbol{E}}_3|^2\overline{\boldsymbol{E}}_1 + \chi^*_{(3\omega)^3,3\omega}|\overline{\boldsymbol{E}}_3|^2\overline{\boldsymbol{E}}_3 + \ldots]\sin 3\omega t + \ldots,\end{aligned} \tag{39}$$

where $\sigma^*_{\omega,\omega}$, $\sigma^*_{3\omega,\omega}$, $\chi^*_{\omega^3,\omega}$, $\chi^*_{\omega^2(3\omega),\omega}$, $\chi^*_{\omega(3\omega)^2,\omega}$ and $\chi^*_{(3\omega)^3,\omega}$ are the linear and the nonlinear effective conductivities at the fundamental frequency, and $\sigma^*_{\omega,3\omega}$, $\sigma^*_{3\omega,3\omega}$, $\chi^*_{\omega^3,,3\omega}$, $\chi^*_{\omega^2(3\omega),3\omega}$, $\chi^*_{\omega(3\omega)^2,3\omega}$ and $\chi^*_{(3\omega)^3,3\omega}$ are the linear and the nonlinear effective conductivities at the third harmonic 3ω.

The electric field at position $\boldsymbol{x}$ is generally given by

$$\boldsymbol{E}^{\alpha}(x,t) = \boldsymbol{E}^{\alpha}_1\sin(\omega t) + \boldsymbol{E}^{\alpha}_3\sin(3\omega t) + \boldsymbol{E}^{\alpha}_5\sin(5\omega t) + \ldots\ . \tag{40}$$

Substituting Eqs. (40) and (39) into Eq. (38), we obtain the AC effective responses of the nonlinear composite at the fundamental frequency and the third harmonics respectively,

$$\sigma^*_{\omega,\omega} = \sigma_{\mathrm{h}} + (\sigma_{\mathrm{i}}-\sigma_{\mathrm{h}})A_0 f_{\mathrm{i}} + (\sigma_{\mathrm{c}}-\sigma_{\mathrm{h}})B_0 f_{\mathrm{c}}, \tag{41}$$

$$\sigma^*_{3\omega,\omega} = 0, \tag{42}$$

$$\begin{aligned}\chi^*_{\omega^3,\omega} &= \frac{3}{4}\chi_{\mathrm{h}} + \frac{3}{4}\left[(\chi_{\mathrm{i}}-\chi_{\mathrm{h}})A_1^3 f_{\mathrm{i}} + (\chi_{\mathrm{c}}-\chi_{\mathrm{h}})\left(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_0^3\right)f_{\mathrm{c}}\right]\\ &\quad - \chi_{\mathrm{h}}\left[(\sigma_{\mathrm{i}}-\sigma_{\mathrm{h}})A_1^{1\mathrm{i}}f_{\mathrm{i}} + (\sigma_{\mathrm{c}}-\sigma_{\mathrm{h}})\left(A_1^{1\mathrm{c}} - \frac{a^3}{b^3}C_1^{2\mathrm{c}} - \frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_1^{3\mathrm{c}}\right)f_{\mathrm{c}}\right],\end{aligned} \tag{43}$$

$$\begin{aligned}\chi^*_{\omega^2(3\omega),\omega} &= -\frac{3}{4}\chi_{\mathrm{h}} - \frac{3}{4}\left[(\chi_{\mathrm{i}}-\chi_{\mathrm{h}})A_1^3 f_{\mathrm{i}} + (\chi_{\mathrm{c}}-\chi_{\mathrm{h}})\left(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_0^3\right)f_{\mathrm{c}}\right]\\ &\quad - \chi_{\mathrm{h}}\left[(\sigma_{\mathrm{i}}-\sigma_{\mathrm{h}})A_1^{1\mathrm{i}}f_{\mathrm{i}} + (\sigma_{\mathrm{c}}-\sigma_{\mathrm{h}})\left(A_1^{1\mathrm{c}} - \frac{a^3}{b^3}C_1^{2\mathrm{c}} - \frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_1^{3\mathrm{c}}\right)f_{\mathrm{c}}\right],\end{aligned} \tag{44}$$

$$\begin{aligned}\chi^*_{\omega(3\omega)^2,\omega} &= \frac{3}{2}\chi_{\mathrm{h}} + \frac{3}{2}\left[(\chi_{\mathrm{i}}-\chi_{\mathrm{h}})A_1^3 f_{\mathrm{i}} + (\chi_{\mathrm{c}}-\chi_{\mathrm{h}})\left(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_0^3\right)f_{\mathrm{c}}\right]\\ &\quad - 2\chi_{\mathrm{h}}\left[(\sigma_{\mathrm{i}}-\sigma_{\mathrm{h}})\,A_1^{1\mathrm{i}}f_{\mathrm{i}} + (\sigma_{\mathrm{c}}-\sigma_{\mathrm{h}})\left(A_1^{1\mathrm{c}} - \frac{a^3}{b^3}C_1^{2\mathrm{c}} - \frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_1^{3\mathrm{c}}\right)f_{\mathrm{c}}\right],\end{aligned} \tag{45}$$

$$\chi^*_{(3\omega)^3,\omega} = 0, \tag{46}$$

$$\sigma^*_{\omega,3\omega} = 0, \tag{47}$$

$$\sigma^*_{3\omega,3\omega} = \sigma_{\mathrm{h}} + (\sigma_{\mathrm{i}}-\sigma_{\mathrm{h}})A_0 f_{\mathrm{i}} + (\sigma_{\mathrm{c}}-\sigma_{\mathrm{h}})B_0 f_{\mathrm{c}}, \tag{48}$$

$$\begin{aligned}\chi^*_{\omega^3,3\omega} &= -\frac{1}{4}\chi_{\mathrm{h}} - \frac{1}{4}\left[(\chi_{\mathrm{i}}-\chi_{\mathrm{h}})A_1^3 f_{\mathrm{i}} + (\chi_{\mathrm{c}}-\chi_{\mathrm{h}})\left(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_0^3\right)f_{\mathrm{c}}\right]\\ &\quad - \chi_{\mathrm{h}}\left[(\sigma_{\mathrm{i}}-\sigma_{\mathrm{h}})A_3^{1\mathrm{i}}f_{\mathrm{i}} + (\sigma_{\mathrm{c}}-\sigma_{\mathrm{h}})\left(A_3^{1\mathrm{c}} - \frac{a^3}{b^3}C_3^{2\mathrm{c}} - \frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_3^{3\mathrm{c}}\right)f_{\mathrm{c}}\right],\end{aligned} \tag{49}$$

$$\begin{aligned}\chi^*_{\omega^2(3\omega),3\omega} &= \frac{3}{2}\chi_{\mathrm{h}} + \frac{3}{2}\left[(\chi_{\mathrm{i}}-\chi_{\mathrm{h}})A_1^3 f_{\mathrm{i}} + (\chi_{\mathrm{c}}-\chi_{\mathrm{h}})\left(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_0^3\right)f_{\mathrm{c}}\right]\\ &\quad + 6\chi_{\mathrm{h}}\left[(\sigma_{\mathrm{i}}-\sigma_{\mathrm{h}})A_3^{1\mathrm{i}}f_{\mathrm{i}} + (\sigma_{\mathrm{c}}-\sigma_{\mathrm{h}})\left(A_3^{1\mathrm{c}} - \frac{a^3}{b^3}C_3^{2\mathrm{c}} - \frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_3^{3\mathrm{c}}\right)f_{\mathrm{c}}\right],\end{aligned} \tag{50}$$

$$\chi^*_{(3\omega)^3,3\omega} = \frac{3}{4}\chi_{\mathrm{h}} + \frac{3}{4}\left[(\chi_{\mathrm{i}}-\chi_{\mathrm{h}})A_1^3 f_{\mathrm{i}} + (\chi_{\mathrm{c}}-\chi_{\mathrm{h}})\left(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_0^3\right)f_{\mathrm{c}}\right]$$

$$+ 3\chi_{\rm h}\left[(\sigma_{\rm i}-\sigma_{\rm h})A_3^{1{\rm i}}f_{\rm i}+(\sigma_{\rm c}-\sigma_{\rm h})\left(A_3^{1{\rm c}}-\frac{a^3}{b^3}C_3^{2{\rm c}}-\frac{a^3}{b^3}\left(1+\frac{a^3}{b^3}\right)C_3^{3{\rm c}}\right)f_{\rm c}\right], \quad (51)$$

$$\chi^*_{\omega(3\omega)^2,3\omega}=0, \quad (52)$$

where $f_{\rm i}$ and $f_{\rm c}$ are the volume fractions of the inclusion core and the coated shell, respectively. The quantity a^3/b^3 can be expressed by the volume fraction as $a^3/b^3 = f_{\rm i}/(f_{\rm i}+f_{\rm c})$. The coefficients $A_0, B_0, C_0, A_1^{1{\rm i}}, A_1^{1{\rm c}}, C_1^{2{\rm c}}$ and $C_1^{3{\rm c}}$ are given in Eqs. (21)–(23), (33)–(36) respectively.

From Eqs. (15)–(24), (27)–(36), (41)–(52), it is noticed that the effective response contains the nonlinear interactions between the fundamental wave and the third harmonic component of the external AC electric field, and between the two inclusion layers, i.e. the inclusion core particle and the coated shell layer.

5. Conclusion

In this paper, an effective nonlinear response of a weakly nonlinear composite with spherical coated particles randomly embedded in a host medium is analysed under the action of an external field, $\boldsymbol{E}_1\sin(\omega t)+\boldsymbol{E}_3\sin(3\omega t)$, using the perturbation method, and the effective conductivities at harmonics are obtained in a dilute limit. The results show that the effective response contains the nonlinear interactions between the fundamental wave and the third harmonic component of the external AC electric field, and between the two inclusion layers.

Considering the coefficients appearing in the effective response, we obtain some simple relationships between the effective nonlinear responses at the fundamental frequency and the third harmonics

$$\chi^*_{\omega^3,3\omega}=-\frac{1}{3}\chi^*_{\omega^3,\omega}, \quad \chi^*_{\omega^2(3\omega),3\omega}=2\chi^*_{\omega^3,\omega},$$
$$\chi^*_{(3\omega)^3,3\omega}=\chi^*_{\omega^3,\omega}, \quad \chi^*_{\omega^2(3\omega),\omega}=-\chi^*_{\omega^3,\omega},$$
$$\chi^*_{\omega(3\omega)^2,\omega}=2\chi^*_{\omega^3,\omega}, \quad \chi^*_{(3\omega)^3,3\omega}=-3\chi^*_{\omega^3,3\omega},$$
$$\chi^*_{\omega^2(3\omega),3\omega}=2\chi^*_{(3\omega)^3,3\omega}.$$

These relations can be used to estimate the effective nonlinear response at third harmonics if the effective nonlinear response at the fundamental frequency is known. In addition, in the case where $\varepsilon_3 = 0$ or $\sigma_{\rm c}=\sigma_{\rm h}$ and $\chi_{\rm c}=\chi_{\rm h}$, our results are exactly the same as those derived by Chen *et al.*[15,16] Moreover, although our discussion is restricted to the effective response of a nonlinear composite in a simple external sinusoidal electric field, $\boldsymbol{E}_1\sin(\omega t)+\boldsymbol{E}_3\sin(3\omega t)$, the method can be easily extended to the effective response of a nonlinear composite in an AC field with many more harmonic frequencies.

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