Vol 14 No 6, June 2005
1009-1963/2005/14(06)/1217-06

Chinese Physics

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Effective nonlinear AC response to composite with spherical particles*

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(Received 2 December 2004; revised manuscript received 12 January 2005)

An effective nonlinear alternative-current (AC) response to granular nonlinear-composite with spherical inclusions embedded in a host medium under the action of an external AC field is investigated by using a perturbation approach. The local potentials of composite at higher harmonics are derived both in a region of local inclusion particles and in a local host region under the action of a sinusoidal field $\boldsymbol{E}_1 \sin \omega t + \boldsymbol{E}_3 \sin 3\omega t$. An effective nonlinear-response to composite and the relationship between the effective nonlinear-responses at the fundamental frequency and the third harmonics are also studied for the spherical inclusions in a dilute limit.

Keywords: nonlinear composites, effective nonlinear response, third harmonics
PACC: 7220H, 7722C, 7320

1. Introduction

Nonlinear composite has attracted much attention both in basic research and in industrial applications in recent years.[1−5] The research on an effective response to nonlinear composite under the action of an alternative-current (AC) external field has been extensively carried out for decades because of its potential applications in engineering. Different theories have been developed for studying the effective nonlinear AC response to granular nonlinear-composite. Levy *et al*[3] considered the second-harmonic generation and the induced third-harmonic generation in a spherical nonlinear-composite. Hui *et al*[6] derived the effective dielectric response to nonlinear media. Gu and Wei *et al*[7−11] gave the effective nonlinear-response to cubic weakly-nonlinear-composites under the action of an external AC electric field with one fixed frequency. Wei *et al*[12] considered the effective nonlinear-response to cylindrical nonlinear-composites in a sinusoidal electric field with a fundamental frequency ω and the third harmonic frequency 3ω.

If an external AC electric field with harmonic frequencies is applied to a nonlinear composite in which the constitutive relation is nonlinear, there will appear many components at all harmonics in a local field. It means that the effective response depends on the external field. In this paper, we deal with the effective nonlinear-response to a granular nonlinear-composite containing spherical particles with their radii being a, randomly embedded in a host medium under the action of a simple external sinusoidal electric field

$$\boldsymbol{E}_a(\boldsymbol{x}, t) = \boldsymbol{E}_1 \sin(\omega t) + \boldsymbol{E}_3 \sin(3\omega t), \qquad (1)$$

where

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

The constitutive relations between current density $\boldsymbol{J}$ and electric field $\boldsymbol{E}$ in the inclusion and host regions are assumed to be

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

where i and h denote the quantities in inclusion particle (i) and host (h) regions, respectively. Ω_{α} represents the region occupied by the α-type of material. Assume that the linear and nonlinear conductive coefficients,

*Project supported by the National High Technology Development Program of China (Grant No 2002AA639270), and the National Natural Science Foundation of China (Grant Nos 40476062 and 10374026).

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σ_α and χ_α, are real and independent of the frequency of the applied field, which is valid for low-frequency processes in a quasi-electrostatic field described by the equation $\nabla \times \boldsymbol{E} = 0$. The corresponding governing equation satisfies $\nabla \cdot \boldsymbol{J} = 0$. Boundary conditions of the composite require the continuity of both the potentials and the normal components of current density at the interface between the inclusion and the host. At infinity the potential must match that of the applied field.

2. Perturbation method of nonlinear AC process

The weakly-nonlinear composite with spherical inclusions randomly embedded in a host medium is discussed, in which the nonlinear terms in the constitutive relations (2) are smaller than the linear terms. For cubic nonlinearity of the constitutive relation of composite, the sinusoidal AC electric field (1) induces the components with harmonic frequencies. Electric potentials at a point $\boldsymbol{x}$ are

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

The third-order conductivity χ_h in the host medium is chosen as a perturbation parameter to develop perturbation expansions for the electric fields and potentials.[13,14] Expansions for the electric potentials in different regions of composite are

$$\begin{aligned}\Phi_s^\alpha(\boldsymbol{x}) =& \Phi_s^{0\alpha}(\boldsymbol{x}) + \chi_\mathrm{h}\Phi_s^{1\alpha}(\boldsymbol{x}) + \chi_\mathrm{h}^2\Phi_s^{2\alpha}(\boldsymbol{x}) + \cdots, \\ &\text{in} \quad \Omega_\alpha, \quad \alpha = \mathrm{i}, \mathrm{h}, \quad s = 1,3,5,\cdots. \end{aligned} \tag{4}$$

Denoting

$$\chi_\alpha = \frac{\chi_\alpha}{\chi_\mathrm{h}}\chi_\mathrm{h} = \beta_\alpha\chi_\mathrm{h}$$

and substituting the potentials (3) and (4) into the current density (2), we obtain perturbation expansions for $\boldsymbol{J}$ in the inclusion and host regions

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

where $\boldsymbol{J}_s^{k\alpha}(\boldsymbol{x})$ is a function of potentials $\Phi_s^{k\alpha}(\boldsymbol{x})$. Since the perturbation current densities for the basic sinusoidal component and the third component are used frequently in the following discussion, we give their expressions here:

$$\boldsymbol{J}_1^{0\alpha}(\boldsymbol{x}) = -\sigma_\alpha\nabla\Phi_1^{0\alpha}(\boldsymbol{x}), \tag{6}$$

$$\boldsymbol{J}_3^{0\alpha}(\boldsymbol{x}) = -\sigma_\alpha\nabla\Phi_3^{0\alpha}(\boldsymbol{x}), \tag{7}$$

$$\begin{aligned}\boldsymbol{J}_1^{1\alpha}(x) =& -\sigma_\alpha\nabla\Phi_1^{1\alpha}(\boldsymbol{x}) - \beta_\alpha\Bigg[\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\Bigg\{\frac{3}{4}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 - \frac{1}{2}(\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\cdot\nabla\Phi_3^{0\alpha}(\boldsymbol{x})) \\ &+ \frac{1}{2}\left|\nabla\Phi_3^{0\alpha}(\boldsymbol{x})\right|^2\Bigg\} + \nabla\Phi_3^{0\alpha}(\boldsymbol{x})\Bigg\{-\frac{1}{4}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 + (\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\cdot\nabla\Phi_3^{0\alpha}(\boldsymbol{x}))\Bigg\}\Bigg], \end{aligned} \tag{8}$$

$$\begin{aligned}\boldsymbol{J}_3^{1\alpha}(\boldsymbol{x}) =& -\sigma_\alpha\nabla\Phi_3^{1\alpha}(\boldsymbol{x}) - \beta_\alpha\Bigg[\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\Bigg\{-\frac{1}{4}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 + (\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\cdot\nabla\Phi_3^{0\alpha}(\boldsymbol{x}))\Bigg\} \\ &+ \nabla\Phi_3^{0\alpha}(\boldsymbol{x})\Bigg\{\frac{1}{2}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 + \frac{3}{4}\left|\nabla\Phi_3^{0\alpha}(\boldsymbol{x})\right|^2\Bigg\}\Bigg]. \end{aligned} \tag{9}$$

The governing equation

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

will be satisfied if each term in Eq.(5) satisfies its corresponding governing equation. Therefore we obtain a set of equations

$$\begin{aligned}&\nabla\cdot\boldsymbol{J}_s^{k\alpha}(\boldsymbol{x}) = 0, \quad \alpha = \mathrm{i}, \mathrm{h}, \\ &s = 1,3,5,\cdots, \quad k = 0,1,2,\cdots. \end{aligned} \tag{10}$$

The corresponding boundary conditions of each term in perturbation potentials and current densities are, respectively, as follows:

At the interface between inclusion and host, $\partial\Omega_\mathrm{i}$,

$$\begin{aligned}&\Phi_s^{k\mathrm{i}}(\boldsymbol{x}) = \Phi_s^{k\mathrm{h}}(\boldsymbol{x}), \quad \hat{n}\cdot\boldsymbol{J}_s^{k\mathrm{i}}(\boldsymbol{x}) = \hat{n}\cdot\boldsymbol{J}_s^{k\mathrm{h}}(\boldsymbol{x}), \\ &\text{on} \quad \partial\Omega_\mathrm{i}, \quad s = 1,3,5,\cdots, \quad k = 0,1,2,\cdots. \end{aligned} \tag{11}$$

At infinity

$$\frac{\partial \Phi_1^{k\mathrm{h}}}{\partial x}\Big|_{x\to\infty} = \begin{cases} -\varepsilon_1, & \text{if } k=0, \\ 0, & \text{if } k\neq 0, \end{cases} \tag{12}$$

$$\frac{\partial \Phi_3^{k\mathrm{h}}}{\partial x}\Big|_{x\to\infty} = \begin{cases} -\varepsilon_3, & \text{if } k=0, \\ 0, & \text{if } k\neq 0. \end{cases} \tag{13}$$

3. Potential distributions in nonlinear composite

From Eqs.(6), (7) and (10), we obtain the governing equations of the zero-order potentials at frequencies ω and 3ω:

$$\sigma_{\mathrm{i}}\nabla^2\Phi_s^{0\mathrm{i}} = 0, \quad s=1,3, \quad \text{in} \quad \Omega_{\mathrm{i}}, \tag{14}$$

$$\sigma_{\mathrm{h}}\nabla^2\Phi_s^{0\mathrm{h}} = 0, \quad s=1,3, \quad \text{in} \quad \Omega_{\mathrm{h}}. \tag{15}$$

With boundary conditions (11), (12) and (13), the zero-order potentials in the inclusion and host regions can be derived from the governing equations (14) and (15). At the fundamental frequency and the third harmonic frequency, the zero-order potentials are

$$\Phi_1^{0\mathrm{i}}(r,\theta) = -A_0\varepsilon_1 r P_1(\cos\theta), \quad (r\le a), \tag{16}$$

$$\Phi_1^{0\mathrm{h}}(r,\theta) = -(r+Br^{-2})\varepsilon_1 P_1(\cos\theta), \quad (r\ge a), \tag{17}$$

$$\Phi_3^{0\mathrm{i}}(r,\theta) = -A_0\varepsilon_3 r P_1(\cos\theta), \quad (r\le a), \tag{18}$$

$$\Phi_3^{0\mathrm{h}}(r,\theta) = -(r+Br^{-2})\varepsilon_3 P_1(\cos\theta), \quad (r\ge a), \tag{19}$$

where $P_1(\cos\theta)=\cos\theta$ is the first-order Legendre function and the coefficients are

$$A_0 = \frac{3\sigma_{\mathrm{h}}}{\sigma_{\mathrm{i}}+2\sigma_{\mathrm{h}}}, \tag{20}$$

$$B = \frac{\sigma_{\mathrm{h}}-\sigma_{\mathrm{i}}}{\sigma_{\mathrm{i}}+2\sigma_{\mathrm{h}}}a^3 = a^3B_0. \tag{21}$$

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

$$\begin{aligned}\sigma_\alpha\nabla^2\Phi_1^{1\alpha}(\boldsymbol{x}) =& -\beta_\alpha\nabla\cdot\Big[\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\Big\{\frac{3}{4}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 - \frac{1}{2}(\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\cdot\nabla\Phi_3^{0\alpha}(\boldsymbol{x})) \\ &+\frac{1}{2}\left|\nabla\Phi_3^{0\alpha}(\boldsymbol{x})\right|^2\Big\} + \nabla\Phi_3^{0\alpha}(\boldsymbol{x})\Big\{-\frac{1}{4}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 + (\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\cdot\nabla\Phi_3^{0\alpha}(\boldsymbol{x}))\Big\}\Big]\end{aligned} \tag{22}$$

and

$$\begin{aligned}\sigma_\alpha\nabla^2\Phi_3^{1\alpha}(\boldsymbol{x}) =& -\beta_\alpha\nabla\cdot\Big[\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\Big\{-\frac{1}{4}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 + (\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\cdot\nabla\Phi_3^{0\alpha}(\boldsymbol{x}))\Big\} \\ &+\nabla\Phi_3^{0\alpha}(\boldsymbol{x})\Big\{\frac{1}{2}\left|\nabla\Phi_1^{0\alpha}(\boldsymbol{x})\right|^2 + \frac{3}{4}\left|\nabla\Phi_3^{0\alpha}(\boldsymbol{x})\right|^2\Big\}\Big],\end{aligned} \tag{23}$$

which are Poisson equations. Solutions of the first-order potentials at the fundamental frequency and the third harmonics in the inclusion and host regions are

$$\Phi_1^{1\mathrm{i}}(r,\theta) = (B_{11}^{\mathrm{i}} r P_1(\cos\theta) + B_{13}^{\mathrm{i}} r^3 P_3(\cos\theta))(\varepsilon_1^3 - \varepsilon_1^2\varepsilon_3 + 2\varepsilon_1\varepsilon_3^2), \quad r\le a, \tag{24}$$

$$\begin{aligned}\Phi_1^{1\mathrm{h}}(r,\theta) =& (\varepsilon_1^3 - \varepsilon_1^2\varepsilon_3 + 2\varepsilon_1\varepsilon_3^2)[(B_{11}^{\mathrm{h}} r^{-2} + B_{12}^{\mathrm{h}} r^{-5} + B_{13}^{\mathrm{h}} r^{-8})P_1(\cos\theta) \\ &+ (C_{11}^{\mathrm{h}} r^{-2} + C_{12}^{\mathrm{h}} r^{-4} + C_{13}^{\mathrm{h}} r^{-5} + C_{14}^{\mathrm{h}} r^{-8})P_3(\cos\theta)], \quad r\ge a,\end{aligned} \tag{25}$$

$$\Phi_3^{1\mathrm{i}}(r,\theta) = (T_{31}^{\mathrm{i}} r P_1(\cos\theta) + T_{33}^{\mathrm{i}} r^3 P_3(\cos\theta))(\varepsilon_1^3 - 6\varepsilon_1^2\varepsilon_3 - 3\varepsilon_3^3), \quad r\le a, \tag{26}$$

and

$$\begin{aligned}\Phi_3^{1\mathrm{h}}(r,\theta) =& (\varepsilon_1^3 - 6\varepsilon_1^2\varepsilon_3 - 3\varepsilon_3^3)[(T_{31}^{\mathrm{h}} r^{-2} + T_{32}^{\mathrm{h}} r^{-5} + T_{33}^{\mathrm{h}} r^{-8})P_1(\cos\theta) \\ &+ (D_{31}^{\mathrm{h}} r^{-2} + D_{32}^{\mathrm{h}} r^{-4} + D_{33}^{\mathrm{h}} r^{-5} + D_{34}^{\mathrm{h}} r^{-8})P_3(\cos\theta)], \quad r\ge a,\end{aligned} \tag{27}$$

where

$$P_3(\cos\theta)=\frac{5\cos 3\theta+3\cos\theta}{8}$$

is the third-order Legendre function and coefficients are as follows:

$$B_{11}^{\mathrm{i}}=\frac{6/5B_0^3-9/10B_0^2+27/10B_0-3/4+3/4A_0^3\beta_{\mathrm{i}}}{\sigma_{\mathrm{i}}+2\sigma_{\mathrm{h}}},$$

$$B_{13}^{\mathrm{i}}=\frac{a^{-2}}{3\sigma_{\mathrm{i}}+4\sigma_{\mathrm{h}}}\left(-\frac{27}{10}B_0^2+\frac{54}{55}B_0^3\right),$$

$$B_{11}^{\mathrm{h}}=\frac{a^3}{\sigma_{\mathrm{i}}+2\sigma_{\mathrm{h}}}\left[\frac{3}{4}(\beta_{\mathrm{i}}A_0^3-1)+\frac{27}{10}B_0+b_1B_0^2+b_2B_0^3\right],$$

(where $b_1=\dfrac{12\sigma_{\mathrm{i}}+15\sigma_{\mathrm{h}}}{10\sigma_{\mathrm{h}}}$ and $b_2=\dfrac{2\sigma_{\mathrm{h}}-5\sigma_{\mathrm{i}}}{10\sigma_{\mathrm{h}}}$),

$$B_{12}^{\mathrm{h}}=-\frac{6a^6}{5\sigma_{\mathrm{h}}}B_0^2,$$

$$B_{13}^{\mathrm{h}}=\frac{a^9}{2\sigma_{\mathrm{h}}}B_0^3,$$

$$C_{11}^{\mathrm{h}}=-\frac{9a^3}{10\sigma_{\mathrm{h}}}B_0,$$

$$C_{12}^{\mathrm{h}}=\frac{a^5}{3\sigma_{\mathrm{i}}+4\sigma_{\mathrm{h}}}(b_3B_0+b_4B_0^2+b_5B_0^3,$$

(where $b_3=\dfrac{9(3\sigma_{\mathrm{i}}+4\sigma_{\mathrm{h}})}{10\sigma_{\mathrm{h}}}$, $b_4=\dfrac{9(6\sigma_{\mathrm{i}}+5\sigma_{\mathrm{h}})}{10\sigma_{\mathrm{h}}}$, and $b_5=\dfrac{9(-15\sigma_{\mathrm{i}}+4\sigma_{\mathrm{h}})}{220\sigma_{\mathrm{h}}}$),

$$C_{13}^{\mathrm{h}}=-\frac{9a^6}{5\sigma_{\mathrm{h}}}B_0^2,$$

$$C_{14}^{\mathrm{h}}=\frac{9a^9}{44\sigma_{\mathrm{h}}}B_0^3,$$

$$T_{31}^{\mathrm{i}}=\frac{-2/5B_0^3+3/10B_0^2-9/10B_0+1/4-1/4A_0^3\beta_{\mathrm{i}}}{\sigma_{\mathrm{i}}+2\sigma_{\mathrm{h}}}=-\frac{1}{3}B_{11}^{\mathrm{i}},$$

$$T_{33}^{\mathrm{i}}=\frac{a^{-2}}{3\sigma_{\mathrm{i}}+4\sigma_{\mathrm{h}}}\left(\frac{9}{10}B_0^2-\frac{18}{55}B_0^3\right)=-\frac{1}{3}B_{13}^{\mathrm{i}},$$

$$T_{31}^{\mathrm{h}}=\frac{a^3}{\sigma_{\mathrm{i}}+2\sigma_{\mathrm{h}}}\left[\left(\frac{1}{4}-\frac{1}{4}\beta_{\mathrm{i}}A_0^3\right)-\frac{9}{10}B_0-b_6B_0^2+b_7B_0^3\right],$$

(where $b_6=\dfrac{5\sigma_{\mathrm{h}}+4\sigma_{\mathrm{i}}}{10\sigma_{\mathrm{h}}}$ and $b_7=\dfrac{5\sigma_{\mathrm{i}}-2\sigma_{\mathrm{h}}}{30\sigma_{\mathrm{h}}}$),

$$T_{32}^{\mathrm{h}}=\frac{2a^6}{5\sigma_{\mathrm{h}}}B_0^2,$$

$$T_{33}^{\mathrm{h}}=-\frac{a^9}{6\sigma_{\mathrm{h}}}B_0^3,$$

$$D_{31}^{\mathrm{h}}=\frac{3a^3}{10\sigma_{\mathrm{h}}}B_0,$$

$$D_{32}^{\mathrm{h}}=\frac{a^5}{3\sigma_{\mathrm{i}}+4\sigma_{\mathrm{h}}}(b_8B_0+b_9B_0^2+b_{10}B_0^3),$$

(where $b_8=-\dfrac{3(3\sigma_{\mathrm{i}}+4\sigma_{\mathrm{h}})}{10\sigma_{\mathrm{h}}}$, $b_9=-\dfrac{3(6\sigma_{\mathrm{i}}+5\sigma_{\mathrm{h}})}{10\sigma_{\mathrm{h}}}$, and $b_{10}=-\dfrac{3(15\sigma_{\mathrm{i}}-4\sigma_{\mathrm{h}})}{220\sigma_{\mathrm{h}}}$),

$$D_{33}^{\mathrm{h}}=\frac{3a^6}{5\sigma_{\mathrm{h}}}B_0^2,$$

$$D_{34}^{\mathrm{h}}=-\frac{3a^9}{44\sigma_{\mathrm{h}}}B_0^3.$$

4. Effective conductivity

In a dilute limit, the effective nonlinear-conductivity can be derived by using the general method previously proposed for DC nonlinear response.[14] Obviously, it is

$$\begin{aligned}&\frac{1}{V}\int_V[\boldsymbol{J}(\boldsymbol{x},t)-(\sigma_{\mathrm{h}}\boldsymbol{E}(\boldsymbol{x},t)+\chi_{\mathrm{h}}\left|\boldsymbol{E}(\boldsymbol{x},t)\right|^2\boldsymbol{E}(\boldsymbol{x},t))]\mathrm{d}V\\&=\bar{\boldsymbol{J}}(t)-[\sigma_{\mathrm{h}}\bar{\boldsymbol{E}}(t)+\chi_{\mathrm{h}}\left|\bar{\boldsymbol{E}}(t)\right|^2\bar{\boldsymbol{E}}(t))],\end{aligned}\tag{28}$$

where V denotes the volume of composite, $\bar{\boldsymbol{J}}(t)$ and $\bar{\boldsymbol{E}}(t)$ are the spatial averages of current density and electric field, respectively. At low particle concentrations, a reasonable assumption is that the average field of the composite can be simply regarded as the external field.[15] Because the integration on the left-hand side of Eq.(28) is zero in the host region, Eq.(28) is simplified into the form

$$\begin{aligned}&\frac{1}{V}\int_{\Omega_{\mathrm{i}}}[\boldsymbol{J}(\boldsymbol{x},t)-(\sigma_{\mathrm{h}}\boldsymbol{E}(\boldsymbol{x},t)+\chi_{\mathrm{h}}\left|\boldsymbol{E}(\boldsymbol{x},t)\right|^2\boldsymbol{E}(\boldsymbol{x},t))]\mathrm{d}V\\&=\bar{\boldsymbol{J}}(t)-\left[\left\{\sigma_{\mathrm{h}}\bar{\boldsymbol{E}}_1+\chi_{\mathrm{h}}\left[\frac{3}{4}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_1-\frac{1}{2}(\bar{\boldsymbol{E}}_1\cdot\bar{\boldsymbol{E}}_3)\bar{\boldsymbol{E}}_1+\frac{1}{2}\left|\bar{\boldsymbol{E}}_3\right|^2\bar{\boldsymbol{E}}_1-\frac{1}{4}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_3+(\bar{\boldsymbol{E}}_1\cdot\bar{\boldsymbol{E}}_3)\bar{\boldsymbol{E}}_3\right]\right\}\sin\omega t\\&+\left\{\sigma_{\mathrm{h}}\bar{\boldsymbol{E}}_3+\chi_{\mathrm{h}}\left[-\frac{1}{4}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_1+(\bar{\boldsymbol{E}}_1\cdot\bar{\boldsymbol{E}}_3)\bar{\boldsymbol{E}}_1+\frac{1}{2}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_3+\frac{3}{4}\left|\bar{\boldsymbol{E}}_3\right|^2\bar{\boldsymbol{E}}_3\right]\right\}\sin 3\omega t+\cdots\Big].\end{aligned}\tag{29}$$

For 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 because of the cubic nonlinearity in the constitutive relation. Thus the effective AC response of composite can be defined as follows:

$$\begin{aligned}\bar{\boldsymbol{J}}(t) =&\bar{\boldsymbol{J}}_\omega \sin\omega t + \bar{\boldsymbol{J}}_{3\omega} \sin 3\omega t + \cdots \\ =&[\sigma^*_{\omega,\omega}\bar{\boldsymbol{E}}_1 + \sigma^*_{3\omega,\omega}\bar{\boldsymbol{E}}_3 + \chi^*_{\omega^3,\omega}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_1 + \chi^*_{\omega^2(3\omega),\omega}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_3 + \chi^*_{\omega(3\omega)^2,\omega}\left|\bar{\boldsymbol{E}}_3\right|^2\bar{\boldsymbol{E}}_1 \\ &+ \chi^*_{(3\omega)^3,\omega}\left|\bar{\boldsymbol{E}}_3\right|^2\bar{\boldsymbol{E}}_3 + \cdots]\sin\omega t + [\sigma^*_{\omega,3\omega}\bar{\boldsymbol{E}}_1 + \sigma^*_{3\omega,3\omega}\bar{\boldsymbol{E}}_3 + \chi^*_{\omega^3,3\omega}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_1 \\ &+ \chi^*_{\omega^2(3\omega),3\omega}\left|\bar{\boldsymbol{E}}_1\right|^2\bar{\boldsymbol{E}}_3 + \chi^*_{\omega(3\omega)^2,3\omega}\left|\bar{\boldsymbol{E}}_3\right|^2\bar{\boldsymbol{E}}_1 + \chi^*_{(3\omega)^3,3\omega}\left|\bar{\boldsymbol{E}}_3\right|^2\bar{\boldsymbol{E}}_3 + \cdots]\sin 3\omega t + \cdots, \qquad (30)\end{aligned}$$

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 linear and 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 linear and nonlinear effective conductivities at the third harmonic 3ω. Electric fields at a position $\boldsymbol{x}$ are generally given by

$$\begin{aligned}\boldsymbol{E}^\alpha(\boldsymbol{x},t) =&\boldsymbol{E}^\alpha_1(\boldsymbol{x})\sin\omega t + \boldsymbol{E}^\alpha_3(\boldsymbol{x})\sin 3\omega t \\ &+ \boldsymbol{E}^\alpha_5(\boldsymbol{x})\sin 5\omega t + \cdots. \qquad (31)\end{aligned}$$

Substituting Eq.(31) in an inclusion region into the left-hand side of Eq.(29), effective responses at the fundamental frequency and the third harmonics can be obtained respectively.

For the fundamental frequency, effective responses are

$$\begin{aligned}\sigma^*_{\omega,\omega} =&\sigma_{\rm h} + (\sigma_{\rm i} - \sigma_{\rm h})A_0 f_{\rm i}, \\ \sigma^*_{3\omega,\omega} =&0, \\ \chi^*_{\omega^3,\omega} =&\frac{3}{4}\chi_{\rm h} + \left[\frac{3}{4}(\chi_{\rm i} - \chi_{\rm h})A_0^3 - \chi_{\rm h}(\sigma_{\rm i} - \sigma_{\rm h})B^{\rm i}_{11}\right] f_{\rm i}, \\ \chi^*_{\omega^2(3\omega),\omega} =&-\frac{3}{4}\chi_{\rm h} + \left[-\frac{3}{4}(\chi_{\rm i} - \chi_{\rm h})A_0^3 + \chi_{\rm h}(\sigma_{\rm i} - \sigma_{\rm h})B^{\rm i}_{11}\right] f_{\rm i}, \\ \chi^*_{\omega(3\omega)^2,\omega} =&\frac{3}{2}\chi_{\rm h} + \left[\frac{3}{2}(\chi_{\rm i} - \chi_{\rm h})A_0^3 - 2\chi_{\rm h}(\sigma_{\rm i} - \sigma_{\rm h})B^{\rm i}_{11}\right] f_{\rm i}, \\ \chi^*_{(3\omega)^3,\omega} =&0,\end{aligned}$$

where $f_{\rm i}$ is the volume fraction of the inclusion particles.

For the third harmonics, we have

$$\begin{aligned}\sigma^*_{\omega,3\omega} =&0, \\ \sigma^*_{3\omega,3\omega} =&\sigma_{\rm h} + (\sigma_{\rm i} - \sigma_{\rm h})A_0 f_{\rm i}, \\ \chi^*_{\omega^3,3\omega} =&-\frac{1}{4}\chi_{\rm h} + \left[-\frac{1}{4}(\chi_{\rm i} - \chi_{\rm h})A_0^3 - \chi_{\rm h}(\sigma_{\rm i} - \sigma_{\rm h})T^{\rm i}_{31}\right] f_{\rm i}, \\ \chi^*_{\omega^2(3\omega),3\omega} =&\frac{3}{2}\chi_{\rm h} + \left[\frac{3}{2}(\chi_{\rm i} - \chi_{\rm h})A_0^3 + 6\chi_{\rm h}(\sigma_{\rm i} - \sigma_{\rm h})T^{\rm i}_{31}\right] f_{\rm i}, \\ \chi^*_{\omega(3\omega)^2,3\omega} =&0, \\ \chi^*_{(3\omega)^3,3\omega} =&\frac{3}{4}\chi_{\rm h} + \left[\frac{3}{4}(\chi_{\rm i} - \chi_{\rm h})A_0^3 + 3\chi_{\rm h}(\sigma_{\rm i} - \sigma_{\rm h})T^{\rm i}_{31}\right] f_{\rm i}.\end{aligned}$$

5. Discussion

It is noticed that there exist some differences between the effective nonlinear-response to nonlinear composite in an external field containing the third harmonics and the effective nonlinear-response to nonlinear composite in an external field $\boldsymbol{E}_1 \sin\omega t$. The former contains the interaction-components of the applied field components $\boldsymbol{E}_1$ and $\boldsymbol{E}_3$, so in this case the effective nonlinear-response to nonlinear composite is much more complicated than that in the simple external field $\boldsymbol{E}_1 \sin\omega t$.[8,9,16]

Now we consider a specific case. When assumption $\varepsilon_3 = 0$ is satisfied, the zero-order and first-order potentials at the fundamental frequency and the third harmonics are

$$\Phi_1^{0\mathrm{i}}(r,\theta) = -A_0\varepsilon_1 r P_1(\cos\theta), \quad (r \le a),$$
$$\Phi_1^{0\mathrm{h}}(r,\theta) = -(r + Br^{-2})\varepsilon_1 P_1(\cos\theta), \quad (r \ge a),$$
$$\Phi_3^{0\mathrm{i}}(r,\theta) = 0, \quad (r \le a),$$
$$\Phi_3^{0\mathrm{h}}(r,\theta) = 0, \quad (r \ge a),$$
$$\Phi_1^{1\mathrm{i}}(r,\theta) = (B_{11}^{\mathrm{i}} r P_1(\cos\theta) + B_{13}^{\mathrm{i}} r^3 P_3(\cos\theta))\varepsilon_1^3, \quad r \le a,$$
$$\begin{aligned}\Phi_1^{1\mathrm{h}}(r,\theta) =& \varepsilon_1^3[(B_{11}^{\mathrm{h}} r^{-2} + B_{12}^{\mathrm{h}} r^{-5} + B_{13}^{\mathrm{h}} r^{-8})P_1(\cos\theta) \\ &+ (C_{11}^{\mathrm{h}} r^{-2} + C_{12}^{\mathrm{h}} r^{-4} + C_{13}^{\mathrm{h}} r^{-5} \\ &+ C_{14}^{\mathrm{h}} r^{-8})P_3(\cos\theta)], \quad r \ge a,\end{aligned}$$
$$\Phi_3^{1\mathrm{i}}(r,\theta) = (T_{31}^{\mathrm{i}} r P_1(\cos\theta) + T_{33}^{\mathrm{i}} r^3 P_3(\cos\theta))\varepsilon_1^3, \quad r \le a,$$
$$\begin{aligned}\Phi_3^{1\mathrm{h}}(r,\theta) =& \varepsilon_1^3[(T_{31}^{\mathrm{h}} r^{-2} + T_{32}^{\mathrm{h}} r^{-5} + T_{33}^{\mathrm{h}} r^{-8})P_1(\cos\theta) \\ &+ (D_{31}^{\mathrm{h}} r^{-2} + D_{32}^{\mathrm{h}} r^{-4} + D_{33}^{\mathrm{h}} r^{-5} \\ &+ D_{34}^{\mathrm{h}} r^{-8})P_3(\cos\theta)], \quad r \ge a.\end{aligned}$$

The effective responses are

$$\sigma^*_{\omega,\omega} = \sigma_{\mathrm{h}} + (\sigma_{\mathrm{i}} - \sigma_{\mathrm{h}})A_0 f_{\mathrm{i}}, \tag{32}$$
$$\sigma^*_{3\omega,\omega} = 0, \tag{33}$$
$$\begin{aligned}\chi^*_{\omega^3,\omega} =& \frac{3}{4}\chi_{\mathrm{h}} + \left[\frac{3}{4}(\chi_{\mathrm{i}} - \chi_{\mathrm{h}})A_0^3 \right. \\ &\left. - \chi_{\mathrm{h}}(\sigma_{\mathrm{i}} - \sigma_{\mathrm{h}})B_{11}^{\mathrm{i}}\right] f_{\mathrm{i}},\end{aligned} \tag{34}$$
$$\chi^*_{\omega^2(3\omega),\omega} = 0, \quad \chi^*_{\omega(3\omega)^2,\omega} = 0, \quad \chi^*_{(3\omega)^3,\omega} = 0, \tag{35}$$
$$\sigma^*_{\omega,3\omega} = 0, \quad \sigma^*_{3\omega,3\omega} = 0, \tag{36}$$
$$\begin{aligned}\chi^*_{\omega^3,3\omega} =& -\frac{1}{4}\chi_{\mathrm{h}} + \left[-\frac{1}{4}(\chi_{\mathrm{i}} - \chi_{\mathrm{h}})A_0^3 \right. \\ &\left. - \chi_{\mathrm{h}}(\sigma_{\mathrm{i}} - \sigma_{\mathrm{h}})T_{31}^{\mathrm{i}}\right] f_{\mathrm{i}},\end{aligned} \tag{37}$$
$$\chi^*_{\omega^2(3\omega),3\omega} = 0, \quad \chi^*_{\omega(3\omega)^2,3\omega} = 0, \quad \chi^*_{(3\omega)^3,3\omega} = 0. \tag{38}$$

Obviously, the effective responses given by the expressions (32)–(38) are exactly the same as those derived by Wei *et al*[16] for effective responses in the simple external field $\boldsymbol{E}_1 \sin\omega t$. So the result of Wei *et al*[16] is just the one for a particular case of the present results.

6. Conclusions

In this paper, an effective nonlinear response to weakly-nonlinear composite with spherical particles randomly embedded in a host medium is analysed under the action of an AC external field $\boldsymbol{E}_1 \sin\omega t + \boldsymbol{E}_3 \sin 3\omega t$, and effective conductivities at harmonics are derived in a dilute limit. It is also deduced that the result of Wei *et al*[16] is the applicable one for our particular case. In application, our results are useful to estimate electric responses to electrorheological fluids.

The present results manifest some simple relations 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-responses at the third harmonics if the effective nonlinear-responses at the fundamental frequency have been known. In addition, although our discussion is restricted to the effective responses to 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 responses to nonlinear composite in an AC field with much more harmonic frequencies.

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