Effective response in nonlinear spherical coated composites under external AC and DC electric fields*

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By using the perturbation method, effective nonlinear direct current (DC) and alternating current (AC) responses of nonlinear composites with spherical coated inclusions randomly embedded in a host medium are studied under the action of an external electric field $\boldsymbol{E}_{\rm a} = \boldsymbol{E}_0 + \boldsymbol{E}_1 \sin\omega t + \boldsymbol{E}_3 \sin 3\omega t$ with different amplitudes and frequencies. The local potentials of composites at all harmonics are given in the inclusion particles and the host regions. All effective nonlinear responses to composites and the relationship between the effective nonlinear responses at all harmonics are also deduced for the spherical coated inclusions in a dilute limit.

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

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1. Introduction

The nonlinear response of composite media has been of much interest because of its important properties and potential application in engineering and in theoretical studies. Recently, many researchers have focused their attention on studying the effective responses of nonlinear composites under different external electric fields.[1−6] In general, the effective responses of composites are related not only to the intrinsic constitutive relations, physical properties of materials, particle shapes, and volume fractions of inclusions, but also to the applied external field.[7−23] In particular, if an external current (AC) electric field is applied to the nonlinear composites, there appear to be many components of a local field in harmonics when the constitutive relations of composites are nonlinear.[7,10] In addition, the local fields can also affect the nonlinear susceptibilities and the nonlinear optical response.[12] For example, Levy *et al.*[5] considered the second harmonic generation and the induced third harmonic generation in a spherical nonlinear composite; Hui *et al.*[10] have derived the effective dielectric response to nonlinear media; Gu *et al.*[11,12,] and Wei *et al.*[15−17] presented the nonlinear effective response of weakly cubic nonlinear composites to an external DC or simple AC electric field. Later, using the perturbation method, Wei *et al.*[18,19] dealt with the effective AC response and the effective AC and DC responses of nonlinear composites, respectively, under an external AC electric field $\boldsymbol{E}_{\rm a} = \boldsymbol{E}_1 \sin\omega t + \boldsymbol{E}_3 \sin 3\omega t$ with different amplitudes and frequencies and an external AC and a DC electric field $\boldsymbol{E}_{\rm app} = \boldsymbol{E}_{\rm a}(1+\sin\omega t)$ for the first time; Chen *et al.*[20] obtained the effective nonlinear AC response of composite with spherical particles under the action of an external sinusoidal field, $\boldsymbol{E}_{\rm a} = \boldsymbol{E}_1 \sin\omega t + \boldsymbol{E}_3 \sin 3\omega t$; Shen *et al.*[21,22] dealt with the effective response of the nonlinear composite under the actions of an external AC and a DC electric field, $\boldsymbol{E}_{\rm a} = \boldsymbol{E}_{\rm a}(1 + \sin\omega t)$; Hao *et al.*[23] studied the effective response of the nonlinear spherical coated composites under an external AC field $\boldsymbol{E}_{\rm a} = \boldsymbol{E}_1 \sin\omega t + \boldsymbol{E}_3 \sin 3\omega t$, and so on.

In this paper, we will devote our efforts to the investigation of effective nonlinear AC and DC responses of spherical coated composites under an external electric field $\boldsymbol{E}_{\rm a} = \boldsymbol{E}_0 + \boldsymbol{E}_1 \sin(\omega t) + \boldsymbol{E}_3 \sin(3\omega t)$ with different amplitudes and frequencies, where $\boldsymbol{E}_0 = \varepsilon_0 \hat{z}$, $\boldsymbol{E}_1 = \varepsilon_1 \hat{z}$, $\boldsymbol{E}_3 = \varepsilon_3 \hat{z}$.

Then we consider homogeneous and isotropic composites, where coated spherical particles are ran-

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domly embedded in a host medium. The constitutive relationship between electric displacement $\boldsymbol{D}$ and an electric field $\boldsymbol{E}$ are assumed to be in the form of

$$\boldsymbol{D}_\alpha = \sigma_\alpha \boldsymbol{E} + x_\alpha |\boldsymbol{E}|^2 \boldsymbol{E}, \quad \text{in } \Omega_\alpha,\ \alpha = \mathrm{i}, \mathrm{c}, \mathrm{h}, \tag{1}$$

where the subscripts $\alpha = \mathrm{i}, \mathrm{c}, \mathrm{h}$ denote the quantities of the inclusion core particle (i), coated shell layer material (c), and host regions (h), respectively; quantity Ω_α is the region occupied by the α-type of material. We assume that the linear dielectric constants σ_α and nonlinear conductive coefficients x_α are independent of the frequency of the applied field, which is valid for the low-frequency processes. The corresponding governing equations are $\boldsymbol{\nabla} \cdot \boldsymbol{D} = 0$ and $\boldsymbol{\nabla} \times \boldsymbol{E} = 0$. The boundary conditions of the coated composites are the continuities 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 processes

We take into account weakly nonlinear composites with coated spherical inclusion 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 a radius a and a coated shell radius b. The external field $\boldsymbol{E}_\mathrm{a} = \boldsymbol{E}_0 + \boldsymbol{E}_1 \sin \omega t + \boldsymbol{E}_3 \sin 3\omega t$ is applied to the composites along the $\hat{z}$-direction, where $\boldsymbol{E}_0 = \varepsilon_0 \hat{z}$, $\boldsymbol{E}_1 = \varepsilon_1 \hat{z}$, $\boldsymbol{E}_3 = \varepsilon_3 \hat{z}$. The electric potential at a point $\boldsymbol{x}$ can be expressed in the following form:

$$\begin{aligned}\phi_\alpha(\boldsymbol{x}, \omega, t) = {} & \phi_0^\alpha(\boldsymbol{x}) + \phi_1^\alpha(\boldsymbol{x}) \sin \omega t + \phi_2^\alpha(\boldsymbol{x}) \cos 2\omega t \\ & + \phi_3^\alpha(\boldsymbol{x}) \sin 3\omega t + \phi_4^\alpha(\boldsymbol{x}) \cos 4\omega t + \cdots \\ & \text{in } \Omega_\alpha \ \ \alpha = \mathrm{i}, \mathrm{c}, \mathrm{h}\end{aligned} \tag{2}$$

For weakly nonlinear composites, we can use the perturbation method to deal with the potentials.[11] 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:

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

Using the electric field equation $\boldsymbol{E}_\alpha(\boldsymbol{x}, \omega, t) = -\nabla \phi_\alpha(\boldsymbol{x}, \omega, t)$ and substituting the potentials (2) and (3) into Eq. (1), we obtain the perturbation expansions for the electric displacement $\boldsymbol{D}$ in the inclusion core particle, coated shell layer material, and host regions:

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

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

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

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 $\boldsymbol{\nabla} \cdot \boldsymbol{D}(\boldsymbol{x}, \omega, t) = 0$ is satisfied. Therefore we obtain a set of equations

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

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

At the interface between the inclusion core and inside layer of the coated shell $\partial\Omega_i$,

$$\begin{aligned}& \phi_k^{\mathrm{si}}(\boldsymbol{x}) = \phi_k^{\mathrm{sc}}(\boldsymbol{x}), \quad \hat{n} \cdot \boldsymbol{D}_k^{\mathrm{si}}(\boldsymbol{x}) = \hat{n} \cdot \boldsymbol{D}_k^{\mathrm{sc}}(\boldsymbol{x}), \\ & \text{on} \quad \partial\Omega_i, \ s = 0, 1, 2, 3, \ldots, \\ & \qquad k = 0, 1, 2, 3, \ldots.\end{aligned} \tag{7}$$

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

At the interface between the outside layer of the coated shell and host medium, $\partial\Omega_c$,

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

At infinity the potential satisfies

$$\frac{\partial \phi_k^{sh}(x)}{\partial x}\,|_{x \to \infty} = \begin{cases} -\varepsilon_0, & \text{if} \quad s = 0, k = 0, \\ -\varepsilon_1, & \text{if} \quad s = 0, k = 1, \\ -\varepsilon_3, & \text{if} \quad s = 0, k = 3, \\ 0, & \text{if} \quad s \neq 0 \quad \text{or if} \quad s = 0 \\ & \text{and} \quad k \neq 0, 1, 3. \end{cases} \tag{9}$$

3. Potentials in coated nonliear spherical composites

From Eqs. (5) and (6), we can obtain the governing equations of the zero-order potentials at the zero frequency, the first frequency, the second, the third,

and the higher harmonics as follows:

$$\sigma_\alpha \nabla^2 \phi_k^{0\alpha}(\boldsymbol{x}) = 0, \quad k = 0, 1, 3, \ldots, \quad \text{in} \quad \Omega_\alpha, \quad \alpha = \mathrm{i}, \mathrm{c}, \mathrm{h}. \tag{10}$$

With the boundary conditions (7)–(9), the zero-order potentials of the core, coated layer, and host regions can be derived from governing Eqs. (10), and they are

$$\phi_0^{0\mathrm{i}}(\boldsymbol{x}) = -A_0 \varepsilon_0 r \mathrm{P}_1(\cos\theta), \ (r \le a), \tag{11}$$

$$\phi_0^{0\mathrm{c}}(\boldsymbol{x}) = -(B_0 r + C r^{-2}) \varepsilon_0 \mathrm{P}_1(\cos\theta), \quad (a \le r \le b), \tag{12}$$

$$\phi_0^{0\mathrm{h}}(\boldsymbol{x}) = -(r + D r^{-2}) \varepsilon_0 \mathrm{P}_1(\cos\theta), \ (r \ge b), \tag{13}$$

$$\phi_1^{0\mathrm{i}}(\boldsymbol{x}) = -A_0 \varepsilon_1 r \mathrm{P}_1(\cos\theta), \ (r \le a), \tag{14}$$

$$\phi_1^{0\mathrm{c}}(\boldsymbol{x}) = -(B_0 r + c r^{-2}) \varepsilon_1 \mathrm{P}_1(\cos\theta), \quad (a \le r \le b), \tag{15}$$

$$\phi_1^{0\mathrm{h}}(\boldsymbol{x}) = -(r + D r^{-2}) \varepsilon_1 \mathrm{P}_1(\cos\theta), \ (r \ge b), \tag{16}$$

$$\phi_3^{0\mathrm{i}}(\boldsymbol{x}) = -A_0 \varepsilon_3 r \mathrm{P}_1(\cos\theta), \ (r \le a), \tag{17}$$

$$\phi_3^{0\mathrm{c}}(\boldsymbol{x}) = -(B_0 r + c r^{-2}) \varepsilon_3 \mathrm{P}_1(\cos\theta), \quad (a \le r \le b), \tag{18}$$

$$\phi_3^{0\mathrm{h}}(\boldsymbol{x}) = -(r + D r^{-2}) \varepsilon_3 \mathrm{P}_1(\cos\theta), \ (r \ge b), \tag{19}$$

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

$$A_0 = \frac{9\sigma_\mathrm{c}\sigma_\mathrm{h}}{(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})(\frac{a}{b})^3}, \tag{20}$$

$$B_0 = \frac{3\sigma_\mathrm{h}(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})}{(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})(\frac{a}{b})^3}, \tag{21}$$

$$C = \frac{3\sigma_\mathrm{h}(\sigma_\mathrm{i} - \sigma_\mathrm{c})a^3}{(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})(\frac{a}{b})^3} = a^3 C_0, \tag{22}$$

$$D = \frac{(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h}) + (\sigma_\mathrm{i} - \sigma_\mathrm{c})(2\sigma_\mathrm{c} + \sigma_\mathrm{h})(\frac{a}{b})^3}{(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_c + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})(\frac{a}{b})^3} b^3 = b^3 D_0. \tag{23}$$

The zero-order potentials with 2ω frequency equal zero because of their boundary conditions and governing equations.

For the advance derivation, we obtain the governing equations of the first-order potential at a different frequency as follows:

$$\sigma_\alpha \nabla^2 \phi_0^{1\alpha} + \beta_\alpha \nabla \cdot \Big[\Big(|\nabla\phi_0^{0\alpha}|^2 + \frac{1}{2}|\nabla\phi_1^{0\alpha}|^2 + \frac{1}{2}|\nabla\phi_3^{0\alpha}|^2 \Big) \nabla\phi_0^{0\alpha} + |\nabla\phi_0^{0\alpha} \cdot \nabla\phi_1^{0\alpha}| \nabla\phi_1^{0\alpha} + |\nabla\phi_0^{0\alpha} \cdot \nabla\phi_1^{0\alpha}| \nabla\phi_3^{0\alpha} \Big] = 0, \tag{24}$$

$$\sigma_\alpha \nabla^2 \phi_1^{1\alpha} + \beta_\alpha \nabla \cdot \Big[2|\nabla\phi_0^{0\alpha} \cdot \nabla\phi_1^{0\alpha}| \nabla\phi_0^{0\alpha} + \Big(|\nabla\phi_0^{0\alpha}|^2 + \frac{3}{4}|\nabla\phi_1^{0\alpha}|^2 - \frac{1}{2}|\nabla\phi_1^{0\alpha} \cdot \nabla\phi_3^{0\alpha}| + \frac{1}{2}|\nabla\phi_3^{0\alpha}|^2 \Big) \nabla\phi_1^{0\alpha} + \Big(-\frac{1}{4}|\nabla\phi_1^{0\alpha}|^2 + |\nabla\phi_1^{0\alpha} \cdot \nabla\phi_3^{0\alpha}| \Big) \nabla\phi_3^{0\alpha} \Big] = 0, \tag{25}$$

$$\sigma_\alpha \nabla^2 \phi_2^{1\alpha} + \beta_\alpha \nabla \cdot \Big[\Big(-\frac{1}{2}|\nabla\phi_1^{0\alpha}|^2 + |\nabla\phi_1^{0\alpha} \cdot \nabla\phi_3^{0\alpha}| \Big) \nabla\phi_0^{0\alpha} + \Big(-|\nabla\phi_0^{0\alpha} \cdot \nabla\phi_1^{0\alpha}| + |\nabla\phi_0^{0\alpha} \cdot \nabla\phi_3^{0\alpha}| \Big) \nabla\phi_1^{0\alpha} + \frac{1}{2}|\nabla\phi_0^{0\alpha} \cdot \nabla\phi_1^{0\alpha}| \nabla\phi_3^{0\alpha} \Big] = 0, \tag{26}$$

$$\sigma_\alpha \nabla^2 \phi_3^{1\alpha} + \beta_\alpha \nabla \cdot \Big[2|\nabla\phi_0^{0\alpha} \cdot \nabla\phi_3^{0\alpha}| \nabla\phi_0^{0\alpha} + \Big(-\frac{1}{4}|\nabla\phi_1^{0\alpha}|^2 + |\nabla\phi_1^{0\alpha} \cdot \nabla\phi_3^{0\alpha}| \Big) \nabla\phi_1^{0\alpha} + \Big(|\nabla\phi_0^{0\alpha}|^2 + \frac{1}{2}|\nabla\phi_1^{0\alpha}|^2 + \frac{3}{4}|\nabla\phi_3^{0\alpha}|^2 \Big) \nabla\phi_3^{0\alpha} \Big] = 0, \tag{27}$$

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

The solutions of the first order potential at zero frequency in the three regions respectively are as follows:

$$\phi_0^{1\mathrm{i}}(\boldsymbol{x}) = \big[A_0^{1\mathrm{i}} r \mathrm{P}_1(\cos\theta) + A_0^{3\mathrm{i}} r^3 \mathrm{P}_3(\cos\theta) \big] \Big(\varepsilon_0^3 + \frac{3}{2}\varepsilon_0\varepsilon_1^2 + \frac{3}{2}\varepsilon_0\varepsilon_3^2 \Big), \quad (r \le a), \tag{28}$$

$$\phi_0^{1\mathrm{c}}(\boldsymbol{x}) = \big[(A_0^{1\mathrm{c}} r + C_0^{1\mathrm{c}} r^{-2} + C_0^{2\mathrm{c}} a^6 r^{-5} + C_0^{3\mathrm{c}} a^9 r^{-8}) \mathrm{P}_1(\cos\theta) + (A_0^{3\mathrm{c}} r^3 + D_0^{1\mathrm{c}} r^{-2} + D_0^{2\mathrm{c}} r^{-4} + D_0^{3\mathrm{c}} r^{-5} + D_0^{4\mathrm{c}} r^{-8}) \mathrm{P}_3(\cos\theta) \big] \Big(\varepsilon_0^3 + \frac{3}{2}\varepsilon_0\varepsilon_1^2 + \frac{3}{2}\varepsilon_0\varepsilon_3^2 \Big), \quad (a \le r \le b), \tag{29}$$

$$\phi_0^{1\mathrm{h}}(\boldsymbol{x}) = \big[(C_0^{1\mathrm{h}} r^{-2} + C_0^{2\mathrm{h}} r^{-5} + C_0^{3\mathrm{h}} r^{-8}) \mathrm{P}_1(\cos\theta) + (D_0^{1\mathrm{h}} r^{-2} + D_0^{2\mathrm{h}} r^{-4} + D_0^{3\mathrm{h}} r^{-5} + D_0^{4\mathrm{h}} r^{-8}) \mathrm{P}_3(\cos\theta) \big] \Big(\varepsilon_0^3 + \frac{3}{2}\varepsilon_0\varepsilon_1^2 + \frac{3}{2}\varepsilon_0\varepsilon_3^2 \Big), \quad (r \ge b). \tag{30}$$

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

At the basic frequency:

$$\phi_1^{1\mathrm{i}}(\boldsymbol{x}) = \big[A_1^{1\mathrm{i}} r \mathrm{P}_1(\cos\theta) + A_1^{3\mathrm{i}} r^3 \mathrm{P}_3(\cos\theta)\big]\Big(3\varepsilon_0^2\varepsilon_1 + \frac{3}{4}\varepsilon_1^3 - \frac{3}{4}\varepsilon_1^2\varepsilon_3 + \frac{3}{2}\varepsilon_1\varepsilon_3^2\Big), \quad (r \le a), \tag{31}$$

$$\phi_1^{1\mathrm{c}}(\boldsymbol{x}) = \big[\big(A_1^{1\mathrm{c}} r + C_1^{1\mathrm{c}} r^{-2} + C_1^{2\mathrm{c}} a^6 r^{-5} + C_1^{3\mathrm{c}} a^9 r^{-8}\big)\mathrm{P}_1(\cos\theta) + \big(A_1^{3\mathrm{c}} r^3 + D_1^{1\mathrm{c}} r^{-2} + D_1^{2\mathrm{c}} r^{-4} + D_1^{3\mathrm{c}} r^{-5} + D_1^{4\mathrm{c}} r^{-8}\big)\mathrm{P}_3(\cos\theta)\big]\Big(3\varepsilon_0^2\varepsilon_1 + \frac{3}{4}\varepsilon_1^3 - \frac{3}{4}\varepsilon_1^2\varepsilon_3 + \frac{3}{2}\varepsilon_1\varepsilon_3^2\Big), \quad (a \le r \le b), \tag{32}$$

$$\phi_1^{1\mathrm{h}}(\boldsymbol{x}) = \big[\big(C_1^{1\mathrm{h}} r^{-2} + C_1^{2\mathrm{h}} r^{-5} + C_1^{3\mathrm{h}} r^{-8}\big)\mathrm{P}_1(\cos\theta) + \big(D_1^{1\mathrm{h}} r^{-2} + D_1^{2\mathrm{h}} r^{-4} + D_1^{3\mathrm{h}} r^{-5} + D_1^{4\mathrm{h}} r^{-8}\big)\mathrm{P}_3(\cos\theta)\big]\Big(3\varepsilon_0^2\varepsilon_1 + \frac{3}{4}\varepsilon_1^3 - \frac{3}{4}\varepsilon_1^2\varepsilon_3 + \frac{3}{2}\varepsilon_1\varepsilon_3^2\Big), \quad (r \ge b); \tag{33}$$

At the second harmonics:

$$\phi_2^{1\mathrm{i}}(\boldsymbol{x}) = \big(A_2^{1\mathrm{i}} r \mathrm{P}_1(\cos\theta) + A_2^{3\mathrm{i}} r^3 \mathrm{P}_3(\cos\theta)\big)\varepsilon_0^3, \quad (r \le a), \tag{34}$$

$$\phi_2^{1\mathrm{c}}(\boldsymbol{x}) = \big[\big(A_2^{1\mathrm{c}} r + C_2^{1\mathrm{c}} r^{-2} + C_2^{2\mathrm{c}} a^6 r^{-5} + C_2^{3\mathrm{c}} a^9 r^{-8}\big)\mathrm{P}_1(\cos\theta) + \big(A_2^{3\mathrm{c}} r^3 + D_2^{1\mathrm{c}} r^{-2} + D_2^{2\mathrm{c}} r^{-4} + D_2^{3\mathrm{c}} r^{-5} + D_2^{4\mathrm{c}} r^{-8}\big)\mathrm{P}_3(\cos\theta)\big]\Big(-\frac{3}{2}\varepsilon_0\varepsilon_1^2 + 3\varepsilon_0\varepsilon_1\varepsilon_3\Big), \quad (a \le r \le b), \tag{35}$$

$$\phi_2^{1\mathrm{h}}(\boldsymbol{x}) = \big[\big(C_2^{1\mathrm{h}} r^{-2} + C_2^{2\mathrm{h}} r^{-5} + C_2^{3\mathrm{h}} r^{-8}\big)\mathrm{P}_1(\cos\theta) + \big(D_2^{1\mathrm{h}} r^{-2} + D_2^{2\mathrm{h}} r^{-4} + D_2^{3\mathrm{h}} r^{-5} + D_2^{4\mathrm{h}} r^{-8}\big)\mathrm{P}_3(\cos\theta)\big]\Big(-\frac{3}{2}\varepsilon_0\varepsilon_1^2 + 3\varepsilon_0\varepsilon_1\varepsilon_3\Big), \quad (r \ge b); \tag{36}$$

At the third harmonics:

$$\phi_3^{1\mathrm{i}}(\boldsymbol{x}) = \big[A_3^{1\mathrm{i}} r \mathrm{P}_1(\cos\theta) + A_3^{3\mathrm{i}} r^3 \mathrm{P}_3(\cos\theta)\big]\Big(3\varepsilon_0^2\varepsilon_3 - \frac{1}{4}\varepsilon_1^3 + \frac{3}{2}\varepsilon_1^2\varepsilon_3 + \frac{3}{4}\varepsilon_3^3\Big), \quad (r \le a), \tag{37}$$

$$\phi_3^{1\mathrm{c}}(\boldsymbol{x}) = \big[\big(A_3^{1\mathrm{c}} r + C_3^{1\mathrm{c}} r^{-2} + C_3^{2\mathrm{c}} a^6 r^{-5} + C_3^{3\mathrm{c}} a^9 r^{-8}\big)\mathrm{P}_1(\cos\theta) + \big(A_3^{3\mathrm{c}} r^3 + D_3^{1\mathrm{c}} r^{-2} + D_3^{2\mathrm{c}} r^{-4} + D_3^{3\mathrm{c}} r^{-5} + D_3^{4\mathrm{c}} r^{-8}\big)\mathrm{P}_3(\cos\theta)\big]\Big(3\varepsilon_0^2\varepsilon_3 - \frac{1}{4}\varepsilon_1^3 + \frac{3}{2}\varepsilon_1^2\varepsilon_3 + \frac{3}{4}\varepsilon_3^3\Big), \quad (a \le r \le b), \tag{38}$$

$$\phi_3^{1\mathrm{h}}(\boldsymbol{x}) = \big[\big(C_3^{1\mathrm{h}} r^{-2} + C_3^{2\mathrm{h}} r^{-5} + C_3^{3\mathrm{h}} r^{-8}\big)\mathrm{P}_1(\cos\theta) + \big(D_3^{1\mathrm{h}} r^{-2} + D_3^{2\mathrm{h}} r^{-4} + D_3^{3\mathrm{h}} r^{-5} + D_3^{4\mathrm{h}} r^{-8}\big)\mathrm{P}_3(\cos\theta)\big]\Big(3\varepsilon_0^2\varepsilon_3 - \frac{1}{4}\varepsilon_1^3 + \frac{3}{2}\varepsilon_1^2\varepsilon_3 + \frac{3}{4}\varepsilon_3^3\Big), \quad (r \ge b), \tag{39}$$

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

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

$$A_0^{1\mathrm{i}} = \frac{\beta_\mathrm{i} A_0^3 (F_1\sigma_\mathrm{c} + 2F_2\sigma_\mathrm{h}) + \beta_\mathrm{c} F_2 (F_3 B_0^3 + F_4 B_0^2 C_0 + F_5 B_0 C_0^2 + F_6 C_0^3) + 3\sigma_\mathrm{c} F_7}{(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})\Big(\frac{a}{b}\Big)^3}, \tag{40}$$

$$A_0^{1\mathrm{c}} = \frac{\beta_\mathrm{i} A_0^3 G_1 + \beta_\mathrm{c}(G_2 B_0^3 - G_3 B_0^2 C_0 + G_4 B_0 C_0^2 - G_5 C_0^3) + 3\sigma_\mathrm{c}(\sigma_\mathrm{i} + 2\sigma_\mathrm{c}) G_6}{\sigma_\mathrm{c}\Big[(\sigma_\mathrm{i} + 2\sigma_\mathrm{c})(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}) + 2(\sigma_\mathrm{i} - \sigma_\mathrm{c})(\sigma_\mathrm{c} - \sigma_\mathrm{h})\Big(\frac{a}{b}\Big)^3\Big]}, \tag{41}$$

$$C_0^{2\mathrm{c}} = -\frac{8\beta_\mathrm{c}}{5\varepsilon_\mathrm{c}} B_0 C_0^2, \quad C_0^{3\mathrm{c}} = \frac{2\beta_\mathrm{c}}{3\varepsilon_\mathrm{c}} C_0^3, \quad C_1^{2\mathrm{c}} = C_0^{2\mathrm{c}}, \quad C_1^{3\mathrm{c}} = C_0^{3\mathrm{c}}, \quad A_1^{1\mathrm{i}} = A_0^{1\mathrm{i}}, \quad A_1^{1\mathrm{c}} = A_0^{1\mathrm{c}}, \tag{42}$$

where

$$F_1 = 1 + 2\Big(\frac{a}{b}\Big)^3, \quad F_2 = 1 - \Big(\frac{a}{b}\Big)^3, \quad F_3 = 2(\sigma_\mathrm{c} - \sigma_\mathrm{h}), \quad F_4 = \frac{18}{5}(\sigma_\mathrm{c} + 2\sigma_\mathrm{h}),$$

$$F_5 = \frac{6}{5}\Big\{\Big[5\Big(\frac{a}{b}\Big)^3 - 1\Big]\sigma_\mathrm{c} - 2\Big[4\Big(\frac{a}{b}\Big)^3 + 1\Big]\sigma_\mathrm{h}\Big\},$$

$$F_6 = \frac{1}{5}\Big\{\Big[71\Big(\frac{a}{b}\Big)^6 + 71\Big(\frac{a}{b}\Big)^3 + 17\Big]\sigma_\mathrm{c} + \Big[-20\Big(\frac{a}{b}\Big)^6 - 20\Big(\frac{a}{b}\Big)^3 + 34\Big]\sigma_\mathrm{h}\Big\},$$

$$F_7 = \Big(-1 + \frac{18}{5}D_0 - \frac{6}{5}D_0^2 - \frac{17}{5}D_0^3\Big),$$

$$G_1 = 2\sigma_\mathrm{c}(\sigma_\mathrm{c} - \sigma_\mathrm{h})\Big(\frac{a}{b}\Big)^3, \quad G_2 = \sigma_\mathrm{c}(\sigma_\mathrm{i} + 2\sigma_\mathrm{c}) + 2\sigma_\mathrm{c}(\sigma_\mathrm{h} - \sigma_\mathrm{c})\Big(\frac{a}{b}\Big)^3, \quad G_3 = \frac{18}{5}\sigma_\mathrm{c}(\sigma_\mathrm{i} + 2\sigma_\mathrm{h})\Big(\frac{a}{b}\Big)^3,$$

$$G_4 = \frac{4}{5}(\sigma_{\rm c}-\sigma_{\rm h})(4\sigma_{\rm i}+5\sigma_{\rm c})\Big(\frac{a}{b}\Big)^3 + \frac{2}{5}(\sigma_{\rm i}+2\sigma_{\rm c})(8\sigma_{\rm h}-5\sigma_{\rm c})\Big(\frac{a}{b}\Big)^6,$$
$$G_5 = \frac{2}{15}\Big(\sigma_{\rm c}-\sigma_{\rm h}\Big)(10\sigma_{\rm i}+71\sigma_{\rm c})(\frac{a}{b})^3 + \frac{1}{15}(\sigma_{\rm i}+2\sigma_{\rm c})(71\sigma_{\rm c}-20\sigma_{\rm h})\Big(\frac{a}{b}\Big)^9,$$
$$G_6 = \Big(-\frac{1}{3}+\frac{6}{5}D_0-\frac{2}{5}D_0^2-\frac{17}{15}D_0^3\Big).$$

4. Effective response

In the dilute limit, we derive the effective nonlinear response of spherical coasted nonlinear composites under external AC and DC electric fields using the effective response method previously proposed for a DC response,[11]

$$\frac{1}{V}\int_V \big[\boldsymbol{D}(\boldsymbol{x},\omega,t)-(\sigma_{\rm h}\boldsymbol{E}(\boldsymbol{x},\omega,t)+\chi_{\rm h}|\boldsymbol{E}(\boldsymbol{x},\omega,t)|^2\boldsymbol{E}(\boldsymbol{x},\omega,t))\big]\,{\rm d}v$$
$$=\bar{\boldsymbol{D}}(\omega,t)-\big[\sigma_{\rm h}\bar{\boldsymbol{E}}(\omega,t)+\chi_{\rm h}|\bar{\boldsymbol{E}}(\omega,t)|^2\bar{\boldsymbol{E}}(\omega,t)\big], \tag{43}$$

where V denotes the whole region, $V=\Omega_{\rm i}+\Omega_{\rm c}+\Omega_{\rm h}$. $\bar{\boldsymbol{D}}$ 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. (43) is zero in the host region, equation (43) is simplified as the following form:

$$\frac{1}{V}\int_{\Omega_{\rm i}+\Omega_{\rm c}} \big[\boldsymbol{D}(\boldsymbol{x},\omega,t)-(\sigma_{\rm h}\boldsymbol{E}(\boldsymbol{x},\omega,t)+\chi_{\rm h}|\boldsymbol{E}(\boldsymbol{x},\omega,t)|^2\boldsymbol{E}(\boldsymbol{x},\omega,t))\big]\,{\rm d}v$$
$$=\bar{\boldsymbol{D}}(\omega,t)-\big[\sigma_{\rm h}\bar{\boldsymbol{E}}(\omega,t)+\chi_{\rm h}|\bar{\boldsymbol{E}}(\omega,t)|^2\bar{\boldsymbol{E}}(\omega,t)\big]. \tag{44}$$

At low concentrations, we evaluate the effective response of the composites with a single particle potential under the external field.[11] For external AC and DC electric fields, because of cubic nonlinearity of the constitutive relation, the electric displacement includes many harmonics components. Thus we can define the effective response of the composites by regarding the nonlinear composites as an effective homogeneous medium.

$$\bar{\boldsymbol{D}}(\omega,t)=\bar{\boldsymbol{D}}_0+\bar{\boldsymbol{D}}_1\sin\omega t+\bar{\boldsymbol{D}}_2\cos 2\omega t+\bar{\boldsymbol{D}}_3\sin 3\omega t+\cdots, \tag{45}$$

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

$$\begin{aligned}\boldsymbol{E}_\alpha(\boldsymbol{x},\omega,t)&=\boldsymbol{E}_0^\alpha(\boldsymbol{x})+\boldsymbol{E}_1^\alpha(\boldsymbol{x})\sin\omega t+\boldsymbol{E}_2^\alpha(\boldsymbol{x})\cos 2\omega t+\boldsymbol{E}_3^\alpha(\boldsymbol{x})\sin 3\omega t+\cdots\\&=-[\nabla\phi_0^{0\alpha}(\boldsymbol{x})+\chi_{\rm h}\nabla\phi_0^{1\alpha}(\boldsymbol{x})+\cdots]-[\nabla\phi_1^{0\alpha}(\boldsymbol{x})+\chi_{\rm h}\nabla\phi_1^{1\alpha}(\boldsymbol{x})+\cdots]\sin\omega t\\&\quad-[\nabla\phi_2^{0\alpha}(\boldsymbol{x})+\chi_{\rm h}\nabla\phi_2^{1\alpha}(\boldsymbol{x})+\cdots]\cos 2\omega t-[\nabla\phi_3^{0\alpha}(\boldsymbol{x})+\chi_{\rm h}\nabla\phi_3^{1\alpha}(\boldsymbol{x})+\cdots]\sin 3\omega t+\cdots.\end{aligned} \tag{46}$$

Substituting Eqs. (45) and (46) into Eq. (44), we can derive the effective responses of nonlinear composites at the n-th harmonics, respectively:

$$\sigma^*_{0,0}=\sigma_{\rm h}+(\sigma_{\rm i}-\sigma_{\rm h})A_0f_{\rm i}+(\sigma_{\rm c}-\sigma_{\rm h})B_0f_{\rm c}, \tag{47}$$

$$\begin{aligned}\chi^*_{0,0}=&\chi_{\rm h}+\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3f_{\rm i}+(\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3+\frac{18}{5}B_0C_0^2\frac{a^3}{b^3}-\frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big]\\&-\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1\rm i}f_{\rm i}+(\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1\rm c}-\frac{a^3}{b^3}C_0^{2\rm c}-\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3\rm c}\Big)f_{\rm c}\Big],\end{aligned} \tag{48}$$

$$\begin{aligned}\chi^*_{0(\omega)^2,0}=&\frac{3}{2}\chi_{\rm h}+\frac{3}{2}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3f_{\rm i}+(\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3+\frac{18}{5}B_0C_0^2\frac{a^3}{b^3}-\frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big]\\&-\frac{3}{2}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1\rm i}f_{\rm i}+(\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1\rm c}-\frac{a^3}{b^3}C_0^{2\rm c}-\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3\rm c}\Big)f_{\rm c}\Big],\end{aligned} \tag{49}$$

$$\begin{aligned}\chi^*_{0(3\omega)^2,0}=&\frac{3}{2}\chi_{\rm h}+\frac{3}{2}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3f_{\rm i}+(\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3+\frac{18}{5}B_0C_0^2\frac{a^3}{b^3}-\frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big]\\&-\frac{3}{2}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1\rm i}f_{\rm i}+(\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1\rm c}-\frac{a^3}{b^3}C_0^{2\rm c}-\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3\rm c}\Big)f_{\rm c}\Big],\end{aligned} \tag{50}$$

$$\sigma^*_{\omega,\omega}=\sigma_{\rm h}+(\sigma_{\rm i}-\sigma_{\rm h})A_0f_{\rm i}+(\sigma_{\rm c}-\sigma_{\rm h})B_0f_{\rm c}, \tag{51}$$

$$\chi^*_{0^2(\omega),\omega} = 3\chi_{\rm h} + 3\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] - 3\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{52}$$

$$\chi^*_{(\omega^3),\omega} = \frac{3}{4}\chi_{\rm h} + \frac{3}{4}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] - \frac{3}{4}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{53}$$

$$\chi^*_{(\omega^2)3\omega,\omega} = -\frac{3}{4}\chi_{\rm h} - \frac{3}{4}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] + \frac{3}{4}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{54}$$

$$\chi^*_{\omega(3\omega)^2,\omega} = \frac{3}{2}\chi_{\rm h} + \frac{3}{2}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] - \frac{3}{2}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{55}$$

$$\sigma^*_{2\omega,2\omega} = \sigma_{\rm h} + (\sigma_{\rm i}-\sigma_{\rm h})A_0 f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})B_0 f_{\rm c}, \tag{56}$$

$$\chi^*_{0(\omega^2),2\omega} = -\frac{3}{2}\chi_{\rm h} - \frac{3}{2}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] + \frac{3}{2}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{57}$$

$$\chi^*_{0(\omega)3\omega,2\omega} = 3\chi_{\rm h} + 3\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] - 3\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{58}$$

$$\sigma^*_{3\omega,3\omega} = \sigma_{\rm h} + (\sigma_{\rm i}-\sigma_{\rm h})A_0 f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})B_0 f_{\rm c}, \tag{59}$$

$$\chi^*_{0^2(3\omega),3\omega} = 3\chi_{\rm h} + 3\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] - 3\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{60}$$

$$\chi^*_{(\omega^3),3\omega} = -\frac{1}{4}\chi_{\rm h} - \frac{1}{4}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] + \frac{1}{4}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{61}$$

$$\chi^*_{(\omega^2)3\omega,3\omega} = \frac{3}{2}\chi_{\rm h} + \frac{3}{2}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] - \frac{3}{2}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{62}$$

$$\chi^*_{(3\omega)^3,3\omega} = \frac{3}{4}\chi_{\rm h} + \frac{3}{4}\Big[(\chi_{\rm i}-\chi_{\rm h})A_0^3 f_{\rm i} + (\chi_{\rm c}-\chi_{\rm h})\Big(B_0^3 + \frac{18}{5}B_0C_0^2\frac{a^3}{b^3} - \frac{2}{5}\frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^3\Big)f_{\rm c}\Big] - \frac{3}{4}\chi_{\rm h}\Big[(\sigma_{\rm i}-\sigma_{\rm h})A_0^{1{\rm i}} f_{\rm i} + (\sigma_{\rm c}-\sigma_{\rm h})\Big(A_0^{1{\rm c}} - \frac{a^3}{b^3}C_0^{2{\rm c}} - \frac{a^3}{b^3}\Big(1+\frac{a^3}{b^3}\Big)C_0^{3{\rm c}}\Big)f_{\rm c}\Big], \tag{63}$$

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, $A_0^{1{\rm i}}$, $A_0^{1{\rm c}}$, $C_0^{2{\rm c}}$, $C_0^{3{\rm c}}$, $A_1^{1{\rm i}}$, $A_1^{1{\rm c}}$, $C_1^{2{\rm c}}$, and $C_0^{3{\rm c}}$ are given in Eqs. (20)–(23) and (40)–(42), respectively.

From Eqs. (11)–(23), (28)–(42), (47)–(63), it is noticed that the effective nonlinear response contains the nonlinear interactions between the direct current component, the fundamental wave, and the third harmonic components of the external AC and DC electric fields, and between the two inclusion layers, i.e. the inclusion core particle and the coated shell layer.

5. Conclusion and discussion

In this paper, effective nonlinear AC and DC responses of weakly nonlinear spherical coated composites, which are randomly embedded in a host medium,

are analyzed under an external field $\boldsymbol{E}_{\rm a} = \boldsymbol{E}_0 + \boldsymbol{E}_1 \sin\omega t + \boldsymbol{E}_3 \sin 3\omega t$ with different amplitudes and frequencies using the perturbation method, and the formulas of the effective nonlinear responses at harmonics are obtained in the dilute limit. Compared with the structures studied in Refs. [11]–[23], here the structure of the nonlinear composite is complicated, and the direct current component and the third harmonic component are contained in the external field, so the effective responses contain the nonlinear interactions between the direct current component, the fundamental wave, and the third harmonic components of the external AC and DC electric fields, and between the two inclusion layers, i.e. the inclusion core particle and the coated shell layer. Furthermore, our case is closer to the circumstances of the practical application and will have a bearing on the nonlinear dielectric response problem of nonlinear composites.

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 as follows:

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

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

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

$$\chi^*_{(\omega)^3,\omega} = \chi^*_{(3\omega)^3,3\omega} = \frac{3}{4}\chi^*_{0,0},$$

$$\chi^*_{(\omega)^2 3\omega,\omega} = -\frac{3}{4}\chi^*_{0,0}, \quad \chi^*_{(\omega)^3,3\omega} = -\frac{1}{4}\chi^*_{0,0}.$$

Clearly, with these relationships we can obtain a comprehensive understanding of the effective nonlinear response of the nonlinear composites.

In addition, if assuming $\varepsilon_0 = 0$, our results are exactly the same as those derived by Hao *et al.*[23]

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