Third–Order Perturbation for Weakly Nonlinear Dielectric Composites of Spherical Inclusions

Mayuree Natenapit*, Akkawuth Chitranondh and Chaivej Thongboonrithi

Department of Physics, Faculty of Science, Chulalongkorn University, Bangkok 10330, Thailand

ABSTRACT: The field equations and the boundary conditions up to the third–order perturbation expansion for electrostatic potential of weakly nonlinear dielectric composites are developed. We consider the electric displacement and the electric field relation which includes the third and the fifth-order nonlinear coefficients. The electric potential up to the third order of a composite consisting of dilute weakly nonlinear spherical inclusions embedded randomly in a linear dielectric host medium is obtained and analyzed. This paper provides the basis for further studies of more complicated nonlinear materials including higher–order nonlinear effective coefficients.

Key words: third–order perturbation, weakly nonlinear dielectric, electrostatic potential in composite media

INTRODUCTION

The effective properties of nonlinear composites have been studied in attempts to design materials with desirable properties.(1-4) In this research the electric response to an external electric field (E_0) of nonlinear composite materials, consisting of weakly nonlinear spherical inclusions randomly embedded in a linear medium, are investigated. Because of the complex nonlinear partial differential equation describing the electrostatic potential can not be solved exactly. Thus, we have to estimate the solution by using an approximation method. There are several types of approximation by which the electric potential can be determined, such as the variational method, the decoupling technique and the expansion perturbation method.(5-8) Second order perturbation method (6, 7) has been widely used to obtain a closed form solution for weakly nonlinear composites. However, there are some errors existing in the continuity of the normal component of electric displacement.

In this research, the third-order perturbation expansion and the single inclusion model were used to obtain the electrostatic potential in the dilute inclusions and host medium. We first develope the theory describing field equations and the boundary conditions in the theory section, then it is applied to solve for the electrostatic potential in the electrostatic potential section. The results are analyzed in the analysis of the electrostatic potential section for error by considering the boundary conditions at the inclusion surface for various material nonlinear coefficients. The conclusions of our work are given in the last section.

THEORY

We consider nonlinear dielectric composites with the following relation between the electric displacement $(\mathbf{D}^i)$ and the electric field $(\mathbf{E}^i)$ in the inclusion

$$\mathbf{D}^i = \varepsilon_i \mathbf{E}^i + \chi_i \left|\mathbf{E}^i\right|^2 \mathbf{E}^i + \eta_i \left|\mathbf{E}^i\right|^4 \mathbf{E}^i , \qquad \ldots (1)$$

Correspondence to: e-mail: mayuree@sc.chula.ac.th, mayuree@hotmail.com

Where ε_i, χ_i and η_i are linear, nonlinear dielectric coefficients in the third and the fifth-orders, respectively. The weakly nonlinear properties of the inclusions are that $\eta_i \left|\vec{E}^i\right|^4 << \varepsilon_i$ and $\chi_i \left|\vec{E}^i\right|^2 << \varepsilon_i$.

The basic equations describing the electric fields are[9]

$$\nabla \cdot \boldsymbol{D} = 0, \quad \text{… (2)}$$

and

$$\nabla \times \boldsymbol{E} = 0. \quad \text{… (3)}$$

The electrostatic potential in the inclusion $\left(\boldsymbol{u}^i\right)$ and host medium $\left(\boldsymbol{u}^m\right)$ are expanded up to the third order as follows:

$$\boldsymbol{u}^i = \boldsymbol{u}_0^i + \lambda \boldsymbol{u}_1^i + \lambda^2 \boldsymbol{u}_2^i + \lambda^3 \boldsymbol{u}_3^i, \quad \text{… (4)}$$

$$\boldsymbol{u}^m = \boldsymbol{u}_0^m + \lambda \boldsymbol{u}_1^m + \lambda^2 \boldsymbol{u}_2^m + \lambda^3 \boldsymbol{u}_3^m, \quad \text{… (5)}$$

where λ is the expansion parameter.

From Eq.(3), $\boldsymbol{E} = -\nabla \boldsymbol{u}$, the electric field can also be written in the form of perturbation expansion as

$$\boldsymbol{E}^\alpha = -\left[\nabla \boldsymbol{u}_0^\alpha + \lambda \nabla \boldsymbol{u}_1^\alpha + \lambda^2 \nabla \boldsymbol{u}_2^\alpha + \lambda^3 \nabla \boldsymbol{u}_3^\alpha\right], \quad \text{… (6)}$$

or

$$\boldsymbol{E}^\alpha = \boldsymbol{E}_0^\alpha + \lambda \boldsymbol{E}_1^\alpha + \lambda^2 \boldsymbol{E}_2^\alpha + \lambda^3 \boldsymbol{E}_3^\alpha ; \alpha = \boldsymbol{m}, \boldsymbol{i}. \quad \text{… (7)}$$

For convenience, we define

$$\boldsymbol{G}^\alpha = \left|\vec{\boldsymbol{E}}^\alpha\right|^2 = \left(\vec{\nabla} \boldsymbol{u}^\alpha\right) \cdot \left(\vec{\nabla} \boldsymbol{u}^\alpha\right) \quad \text{… (8)}$$

to denote the square of the electric fields in each region. From Eqs. (7) and (8), we obtain

$$\boldsymbol{G}^\alpha = \boldsymbol{G}_0^\alpha + \lambda \boldsymbol{G}_1^\alpha + \lambda^2 \boldsymbol{G}_2^\alpha + \lambda^3 \boldsymbol{G}_3^\alpha, \quad \text{… (9)}$$

where

$$\boldsymbol{G}_0^\alpha = \left|\vec{\nabla} \boldsymbol{u}_0^\alpha\right|^2, \quad \text{… (10.a)}$$

$$\boldsymbol{G}_1^\alpha = 2\nabla \boldsymbol{u}_0^\alpha \cdot \nabla \boldsymbol{u}_1^\alpha, \quad \text{… (10.b)}$$

$$\boldsymbol{G}_2^\alpha = \left|\vec{\nabla} \boldsymbol{u}_1^\alpha\right|^2 + 2\vec{\nabla} \boldsymbol{u}_0^\alpha \cdot \vec{\nabla} \boldsymbol{u}_2^\alpha, \quad \text{… (10.c)}$$

$$\boldsymbol{G}_3^\alpha = 2\nabla \boldsymbol{u}_0^\alpha \cdot \nabla \boldsymbol{u}_3^\alpha + 2\nabla \boldsymbol{u}_1^\alpha \cdot \nabla \boldsymbol{u}_2^\alpha. \quad \text{… (10.d)}$$

The relation between the electric displacement and the electric field in the inclusion and host medium are

$$\vec{\boldsymbol{D}}^i = \varepsilon_i \vec{\boldsymbol{E}}^i + \chi_i \boldsymbol{G}^i \vec{\boldsymbol{E}}^i + \eta_i \left(\boldsymbol{G}^i\right)^2 \vec{\boldsymbol{E}}^i, \quad \text{… (11)}$$

and

$$\boldsymbol{D}^m = \varepsilon_m \boldsymbol{E}^m. \quad \text{… (12)}$$

Substituting $\boldsymbol{E}^i$ from Eq. (6) and $\boldsymbol{G}^i$ from Eq. (9) in Eq. (11), we obtain

$$\boldsymbol{D}^i = -\varepsilon_i \nabla \boldsymbol{u}_0^i - \lambda\left(\varepsilon_i \nabla \boldsymbol{u}_1^i + \beta_i \boldsymbol{G}_0^i \nabla \boldsymbol{u}_0^i\right) - \lambda^2 \left(\varepsilon_i \nabla \boldsymbol{u}_2^i + \beta_i \left(\boldsymbol{G}_0^i \nabla \boldsymbol{u}_1^i + \boldsymbol{G}_1^i \nabla \boldsymbol{u}_0^i\right) + \gamma_i \left(\boldsymbol{G}_0^i\right)^2 \nabla \boldsymbol{u}_0^i\right)$$
$$-\lambda^3 \left(\varepsilon_i \nabla \boldsymbol{u}_3^i + \beta_i \left(\boldsymbol{G}_0^i \nabla \boldsymbol{u}_2^i + \boldsymbol{G}_1^i \nabla \boldsymbol{u}_1^i + \boldsymbol{G}_2^i \nabla \boldsymbol{u}_0^i\right) + \gamma_i \left(\left(\boldsymbol{G}_0^i\right)^2 \nabla \boldsymbol{u}_1^i + 2\boldsymbol{G}_0^i \boldsymbol{G}_1^i \nabla \boldsymbol{u}_0^i\right)\right), \quad \ldots (13)$$

where $\beta_i = \dfrac{\chi_i}{\lambda}$ and $\gamma_i = \dfrac{\eta_i}{\lambda^2}$. Comparing Eqs. (12) and (13) with

$$\boldsymbol{D}^\alpha = \boldsymbol{D}_0^\alpha + \lambda \boldsymbol{D}_1^\alpha + \lambda^2 \boldsymbol{D}_2^\alpha + \lambda^3 \boldsymbol{D}_3^\alpha, \quad \ldots (14)$$

yields

$$\boldsymbol{D}_0^\alpha = -\varepsilon_\alpha \nabla \boldsymbol{u}_0^\alpha, \quad \ldots (15)$$

$$\boldsymbol{D}_1^\alpha = -\varepsilon_\alpha \nabla \boldsymbol{u}_1^\alpha - \beta_\alpha \boldsymbol{G}_0^\alpha \nabla \boldsymbol{u}_0^\alpha, \quad \ldots (16)$$

$$\boldsymbol{D}_2^\alpha = -\varepsilon_\alpha \nabla \boldsymbol{u}_2^\alpha - \beta_\alpha \left(\boldsymbol{G}_0^\alpha \nabla \boldsymbol{u}_1^\alpha + \boldsymbol{G}_1^\alpha \nabla \boldsymbol{u}_0^\alpha\right) - \gamma_\alpha \left(\boldsymbol{G}_0^\alpha\right)^2 \nabla \boldsymbol{u}_0^\alpha, \quad \ldots (17)$$

and

$$\boldsymbol{D}_3^\alpha = -\varepsilon_\alpha \nabla \boldsymbol{u}_3^\alpha - \beta_\alpha \left(\boldsymbol{G}_0^\alpha \nabla \boldsymbol{u}_2^\alpha + \boldsymbol{G}_1^\alpha \nabla \boldsymbol{u}_1^\alpha + \boldsymbol{G}_2^\alpha \nabla \boldsymbol{u}_0^\alpha\right) - \gamma_\alpha \left(\left(\boldsymbol{G}_0^\alpha\right)^2 \nabla \boldsymbol{u}_1^\alpha + 2\boldsymbol{G}_0^\alpha \boldsymbol{G}_1^\alpha \nabla \boldsymbol{u}_0^\alpha\right). \quad \ldots (18)$$

Substituting Eqs. (15)-(18) in Eq. (2), we obtain

$$\nabla^2 \boldsymbol{u}_0^i = 0, \quad \ldots (19)$$

$$\varepsilon_i \nabla^2 \boldsymbol{u}_1^i + \beta_i \left(\boldsymbol{G}_0^i \nabla^2 \boldsymbol{u}_0^i + \nabla \boldsymbol{G}_0^i \cdot \nabla \boldsymbol{u}_0^i\right) = 0, \quad \ldots (20)$$

$$\varepsilon_i \nabla^2 \boldsymbol{u}_2^i + \beta_i \left(\boldsymbol{G}_0^i \nabla^2 \boldsymbol{u}_1^i + \nabla \boldsymbol{u}_1^i \cdot \nabla \boldsymbol{G}_0^i + \boldsymbol{G}_1^i \nabla^2 \boldsymbol{u}_0^i + \nabla \boldsymbol{u}_0^i \cdot \nabla \boldsymbol{G}_1^i\right)$$
$$+\gamma_i \left(\left(\boldsymbol{G}_0^i\right)^2 \nabla^2 \boldsymbol{u}_0^i + 2\boldsymbol{G}_0^i \nabla \boldsymbol{u}_0^i \cdot \nabla \boldsymbol{G}_0^i\right) = 0, \quad \ldots (21)$$

and

$$\varepsilon_i \nabla^2 \boldsymbol{u}_3^i + \beta_i \left(\boldsymbol{G}_0^i \nabla^2 \boldsymbol{u}_2^i + \nabla \boldsymbol{u}_2^i \cdot \nabla \boldsymbol{G}_0^i + \boldsymbol{G}_1^i \nabla^2 \boldsymbol{u}_1^i + \nabla \boldsymbol{u}_1^i \cdot \nabla \boldsymbol{G}_1^i + \boldsymbol{G}_2^i \nabla^2 \boldsymbol{u}_0^i + \nabla \boldsymbol{u}_0^i \cdot \nabla \boldsymbol{G}_2^i\right)$$
$$+\gamma_i \left(\left(\boldsymbol{G}_0^i\right)^2 \nabla^2 \boldsymbol{u}_1^i + 2\boldsymbol{G}_0^i \nabla \boldsymbol{u}_1^i \cdot \nabla \boldsymbol{G}_0^i + 2\boldsymbol{G}_0^i \boldsymbol{G}_1^i \nabla^2 \boldsymbol{u}_0^i + 2\boldsymbol{G}_0^i \nabla \boldsymbol{u}_0^i \cdot \nabla \boldsymbol{G}_1^i + 2\boldsymbol{G}_1^i \nabla \boldsymbol{u}_0^i \cdot \nabla \boldsymbol{G}_0^i\right) = 0. \quad \ldots (22)$$

Eqs. (19)-(22) are the equations describing the electric potential in the inclusion of the zeroth to the third order which have to be solved. Similarly for the host medium, the equations of the zeroth to the third-order potential are

$$\nabla^2 \boldsymbol{u}_0^m = 0, \quad \ldots (23)$$

$$\nabla^2 \boldsymbol{u}_1^m = 0, \quad \ldots (24)$$

$$\nabla^2 \boldsymbol{u}_2^m = 0, \quad \ldots (25)$$

and

$$\nabla^2 \boldsymbol{u}_3^m = 0, \quad \ldots (26)$$

which are Laplace equations.

ELECTROSTATIC POTENTIAL

In order to solve for each order electrostatic potential, the following boundary conditions are imposed:

1. the electrostatic potential in the host medium at remote distance $(r \to \infty)$.
2. the electrostatic potential at the center of the inclusion $(r = 0)$.
3. the continuity of the tangential of electric field at the inclusion surface $(r = R)$.
4. the continuity of the normal component of electric displacement at $r = R$.

From the above boundary conditions, we have

$$u_0^m(r \to \infty) = -E_0 r \cos\theta, \quad u_j^m(r \to \infty) = 0, \ \text{j = 1, 2, 3} \qquad \dots (27)$$

$$u_j^i(r = 0, \theta) = \textit{finite}, \ \text{j = 0, 1, 2, 3} \qquad \dots (28)$$

$$\left.\frac{\partial u_j^i}{\partial \theta}\right|_{r=R} = \left.\frac{\partial u_j^i}{\partial \theta}\right|_{r=R}, \ \text{j = 0, 1, 2, 3} \qquad \dots (29)$$

$$\varepsilon_i \left.\frac{\partial u_0^i}{\partial r}\right|_R = \varepsilon_m \left.\frac{\partial u_0^m}{\partial r}\right|_R, \qquad \dots (30.1)$$

$$\varepsilon_i \left.\frac{\partial u_1^i}{\partial r}\right|_R + \beta_i G_0^i \left.\frac{\partial u_0^i}{\partial r}\right|_R = \varepsilon_m \left.\frac{\partial u_1^m}{\partial r}\right|_R, \qquad \dots (30.2)$$

$$\varepsilon_i \left.\frac{\partial u_2^i}{\partial r}\right|_R + \beta_i \left(G_0^i \left.\frac{\partial u_1^i}{\partial r}\right|_R + G_1^i \left.\frac{\partial u_0^i}{\partial r}\right|_R \right) + \gamma_i \left(G_0^i\right)^2 \left.\frac{\partial u_0^i}{\partial r}\right|_R = \varepsilon_m \left.\frac{\partial u_2^m}{\partial r}\right|_R, \qquad \dots (30.3)$$

$$\varepsilon_i \left.\frac{\partial u_3^i}{\partial r}\right|_R + \beta_i \left(G_0^i \left.\frac{\partial u_2^i}{\partial r}\right|_R + G_1^i \left.\frac{\partial u_1^i}{\partial r}\right|_R + G_2^i \left.\frac{\partial u_0^i}{\partial r}\right|_R \right) + \gamma_i \left(\left(G_0^i\right)^2 \left.\frac{\partial u_1^i}{\partial r}\right|_R + 2G_0^i G_1^i \left.\frac{\partial u_0^i}{\partial r}\right|_R \right) = \varepsilon_m \left.\frac{\partial u_3^m}{\partial r}\right|_R. \qquad \dots (30.4)$$

The zeroth-order potential

We first determine the zeroth–order electrostatic potential, $u_0^m(r, \theta)$, from Eq. (23) of which the general solution is

$$u_0^m(r, \theta) = \sum_{l=0}^{\infty} \left(A_{0l}^m r^l + B_{0l}^m r^{-(l+1)} \right) P_l(\cos\theta), \qquad \dots (31)$$

where $P_l(\cos\theta)$ is Legendre polynomial of order l.

From the boundary condition in Eq. (27), we obtain

$$u_0^m(r, \theta) = -E_0 r \cos\theta + \sum_{l=0}^{\infty} B_{0l}^m r^{-2} P_l(\cos\theta). \qquad \dots (32)$$

For linear host medium, u_0^i also satisfies Laplace equation, Eq. (19) with the general solution

$$u_0^i(r, \theta) = \sum_{l=0}^{\infty} \left(A_{0l}^i r^l + B_{0l}^i r^{-(l+1)} \right) P_l(\cos\theta). \qquad \dots (33)$$

From the boundary condition in Eq. (28), the coefficient B_{0l}^{i} is zero so

$$u_0^i(r,\theta)=\sum_{l=0}^{\infty}A_{0l}^{i}r^{l}P_l(\cos\theta). \qquad \ldots (34)$$

Using the boundary condition in Eq. (29) which can be written as $u_0^i(R,\theta)=u_0^m(R,\theta)$, the coefficients B_{0l}^{m} in Eq. (32) and A_{0l}^{i} in Eq. (34) can be solved. The solutions are

$$A_{0l}^{i}=B_{0l}^{m}R^{-(l+2)}, \text{ for } l\neq 1 \qquad \ldots (35)$$

and

$$A_{01}^{i}=-E_0+B_{01}^{m}R^{-3}. \qquad \ldots (36)$$

From the boundary condition in Eq. (30.1), and by using Eqs. (35)-(36), we obtain

$A_{0l}^{i}=B_{0l}^{m}=0$ for $l\neq 1$,

$$B_{01}^{m}=E_0R^3\left[\frac{\varepsilon_i-\varepsilon_m}{\varepsilon_i+2\varepsilon_m}\right], \qquad \ldots (37)$$

and

$$A_{01}^{i}=-E_0+E_0\left\{\frac{\pounds_i-\pounds_m}{\pounds_i+2e_m}\right\}. \qquad \ldots (38)$$

The zeroth–order electric potential in the host medium and inclusion are

$$u_0^m(r,\theta)=-E_0\left(r-br^{-2}\right)\cos\theta, \qquad \ldots (39)$$

where

$$b=R^3\left[\frac{\varepsilon_i-\varepsilon_m}{\varepsilon_i+2\varepsilon_m}\right], \qquad \ldots (40)$$

and

$$u_0^i(r,\theta)=-E_0cr\cos\theta, \qquad \ldots (41)$$

where

$$c=\frac{3\varepsilon_m}{\varepsilon_i+2\varepsilon_m}. \qquad \ldots (42)$$

The first-order potential

The general solution of Eq. (24) is

$$u_1^m(r,\theta)=\sum_{l=0}^{\infty}\left(A_{1l}^{m}r^{l}+B_{1l}^{m}r^{-(l+1)}\right)P_l(\cos\theta). \qquad \ldots (43)$$

Imposing the first boundary condition in Eq. (27), $u_1^m(r\rightarrow\infty)=0$, we get $A_{1l}^{m}=0$.

In the nonlinear region, we consider Eq. (20) which is reduced to $\nabla^2u_1^i=0$ after u_0^i and

G_0^i terms are substituted using Eqs. (41) and (42). The general solution u_1^i is in the same form as Eq. (43) and after imposing the second boundary condition, Eq. (28), we get

$$u_1^i(r,\theta)=\sum_{l=0}^{\infty}A_{1l}^i r^l P_l(\cos\theta)\,. \qquad \ldots(44)$$

By using the third boundary condition in Eq. (29), we get

$$A_{1l}^i R^l = B_{1l}^m R^{-(l+1)}. \qquad \ldots(45)$$

Then imposing the fourth boundary condition in Eq. (30.2), we obtain

$$A_{1l}^i = B_{1l}^m = 0,\ \text{for } l\neq 1 \qquad \ldots(46)$$

and

$$\varepsilon_i B_{11}^m R^{-3}-\beta_i(E_0c)^3=\varepsilon_m(-2B_{11}^m R^{-3}). \qquad \ldots(47)$$

From Eq. (45) for $l=1$,

$$A_{11}^i = B_{11}^m R^{-3}, \qquad \ldots(48)$$

and by using Eqs. (47) and (48), the coefficients B_{11}^m and A_{11}^i are determined and the first-order potential in the host medium and inclusion are obtained as

$$u_1^m(r,\theta)=\frac{\beta_i(E_0c)^3R^3}{\varepsilon_i+2\varepsilon_m}r^{-2}\cos\theta, \qquad \ldots(49)$$

and

$$u_1^i(r,\theta)=\frac{\beta_i(E_0c)^3}{\varepsilon_i+2\varepsilon_m}r\cos\theta\,. \qquad \ldots(50)$$

The second-order potential

The general solution of Eq. (25) is

$$u_2^m(r,\theta)=\sum_{l=0}^{\infty}\left(A_{2l}^m r^l+B_{2l}^m r^{-(l+1)}\right)P_l(\cos\theta)\,. \qquad \ldots(51)$$

Imposing the boundary condition in Eq. (27), $u_2^m(r\to\infty)=0$, we get $A_{2l}^m=0$.

In the nonlinear region, we consider Eq. (21) which is reduced to $\nabla^2u_2^i=0$ after u_0^i, u_1^i, G_1^i and G_0^i terms are substituted using Eqs. (41), (42) and (50). The general solution of u_2^i is the same form as Eq. (51) and after imposing the second boundary condition, Eq. (28), we get

$$u_2^i(r,\theta)=\sum_{l=0}^{\infty}A_{2l}^i r^l P_l(\cos\theta)\,. \qquad \ldots(52)$$

By using the third boundary condition in Eq. (29), we get

$$A_{2l}^i R^l = B_{2l}^m R^{-(l+1)}\,. \qquad \ldots(53)$$

We again impose the fourth boundary condition in Eq. (30.3), hence

$$\boldsymbol{A}_{2l}^{i}=\boldsymbol{B}_{2l}^{m}=0,\ \text{for}\ \boldsymbol{l}\neq 1 \qquad \ldots (54)$$

and

$$\varepsilon_i \boldsymbol{B}_{21}^{m}\boldsymbol{R}^{-3}+\frac{3\beta_i^2\left(\boldsymbol{E}_0\boldsymbol{c}\right)^5}{\varepsilon_i+2\varepsilon_m}-\gamma_i\left(\boldsymbol{E}_0\boldsymbol{c}\right)^5=\varepsilon_m\left(-2\boldsymbol{B}_{21}^{m}\boldsymbol{R}^{-3}\right). \qquad \ldots (55)$$

From Eq. (53) for $\boldsymbol{l}=1$,

$$\boldsymbol{A}_{21}^{i}=\boldsymbol{B}_{21}^{m}\boldsymbol{R}^{-3}, \qquad \ldots (56)$$

and by using Eqs. (55) and (56), the coefficients $\boldsymbol{B}_{21}^{m}$ and $\boldsymbol{A}_{21}^{i}$ are determined and the second-order potential in the host medium and inclusion are obtained as

$$\boldsymbol{u}_2^{m}\left(\boldsymbol{r},\theta\right)=\left[\frac{-3\beta_i^2}{\left(\varepsilon_i+2\varepsilon_m\right)^2}+\frac{\gamma_i}{\left(\varepsilon_i+2\varepsilon_m\right)}\right]\cdot\left(\boldsymbol{E}_0\boldsymbol{c}\right)^5\boldsymbol{R}^3\boldsymbol{r}^{-2}\,\boldsymbol{cos}\theta, \qquad \ldots (57)$$

and

$$\boldsymbol{u}_2^{i}\left(\boldsymbol{r},\theta\right)=\left[\frac{-3\beta_i^2}{\left(\varepsilon_i+2\varepsilon_m\right)^2}+\frac{\gamma_i}{\left(\varepsilon_i+2\varepsilon_m\right)}\right]\cdot\left(\boldsymbol{E}_0\boldsymbol{c}\right)^5\boldsymbol{r}\,\boldsymbol{cos}\theta\ . \qquad \ldots (58)$$

The third-order potential

The third-order potential satisfying Eq. (26) and the boundary condition Eq. (27),

$\boldsymbol{u}_3^{m}\left(\boldsymbol{r}\to\infty\right)=0$, is

$$\boldsymbol{u}_3^{m}\left(\boldsymbol{r},\theta\right)=\sum_{\boldsymbol{l}=0}^{\infty}\boldsymbol{B}_{3l}^{m}\boldsymbol{r}^{-(l+1)}\boldsymbol{P}_l\left(\boldsymbol{cos}\theta\right). \qquad \ldots (59)$$

Similarly Eq. (22) is reduced to $\nabla^2\boldsymbol{u}_3^{i}=0$ after $\boldsymbol{u}_0^{i}$, $\boldsymbol{u}_1^{i}$, $\boldsymbol{u}_2^{i}$, $\boldsymbol{G}_0^{i}$, $\boldsymbol{G}_1^{i}$ and $\boldsymbol{G}_2^{i}$ terms are substituted using Eqs. (41), (42), (50), (58), (10.a), (10.b) and (10.c). After imposing the second boundary condition, Eq. (28), to $\boldsymbol{u}_3^{i}\left(\boldsymbol{r},\theta\right)$ we get

$$\boldsymbol{u}_3^{i}\left(\boldsymbol{r},\theta\right)=\sum_{\boldsymbol{l}=0}^{\infty}\boldsymbol{A}_{3l}^{i}\boldsymbol{r}^{l}\boldsymbol{P}_l\left(\boldsymbol{cos}\theta\right). \qquad \ldots (60)$$

By using the third boundary condition in Eq. (29), we get

$$\boldsymbol{A}_{3l}^{i}\boldsymbol{R}^{l}=\boldsymbol{B}_{3l}^{m}\boldsymbol{R}^{-(l+1)}. \qquad \ldots (61)$$

Imposing the fourth boundary condition in Eq. (30.4), we get

$$\boldsymbol{A}_{3l}^{i}=\boldsymbol{B}_{3l}^{m}=0,\ \text{for}\ \boldsymbol{l}\neq 1 \qquad \ldots (62)$$

and

$$\varepsilon_i B_{31}^{m}R^{-3}-\beta_i\left[\frac{-3\gamma_i}{\varepsilon_i+2\varepsilon_m}+\frac{12\beta_i^2}{\left(\varepsilon_i+2\varepsilon_m\right)^2}\right]\left(E_0c\right)^7+\frac{5\beta_i\gamma_i}{\varepsilon_i+2\varepsilon_m}\left(E_0c\right)^7=-2\varepsilon_m B_{31}^{m}R^{-3}. \qquad \ldots (63)$$

From Eq. (61) for $\boldsymbol{l}=1$,

$$\boldsymbol{A}_{31}^{i}=\boldsymbol{B}_{31}^{m}\boldsymbol{R}^{-3}, \qquad \ldots (64)$$

and by using Eqs. (63) and (64), $\boldsymbol{B}_{31}^{m}$ and $\boldsymbol{A}_{31}^{i}$ are determined and the third order potential in the host medium and inclusion are obtained as

$$u_3^m(r,\theta)=\left[\frac{12\beta_i^3}{(\varepsilon_i+2\varepsilon_m)^3}-\frac{8\beta_i\gamma_i}{(\varepsilon_i+2\varepsilon_m)^2}\right]R^3(E_0c)^7 r^{-2}\cos\theta\,, \quad \ldots (65)$$

and

$$u_3^i(r,\theta)=\left[\frac{12\beta_i^3}{(\varepsilon_i+2\varepsilon_m)^3}-\frac{8\beta_i\gamma_i}{(\varepsilon_i+2\varepsilon_m)^2}\right](E_0c)^7 r\cos\theta\,. \quad \ldots (66)$$

Combining all the solutions, the zeroth up to the third-order potential, in Eqs. (4) and (5), we obtain

$$u^i(r,\theta)=\{-E_0c+\frac{\chi_i}{\varepsilon_i+2\varepsilon_m}(E_0c)^3-\left[\frac{3\chi_i^2}{(\varepsilon_i+2\varepsilon_m)^2}-\frac{\eta_i}{\varepsilon_i+2\varepsilon_m}\right](E_0c)^5$$
$$+\left[\frac{12\chi_i^3}{(\varepsilon_i+2\varepsilon_m)^3}-\frac{8\chi_i\eta_i}{(\varepsilon_i+2\varepsilon_m)^2}\right](E_0c)^7\}r\cos\theta\,, \quad \ldots (67)$$

and

$$u^m(r,\theta)=-E_0r\cos\theta+\{E_0b+\frac{\chi_iR^3}{\varepsilon_i+2\varepsilon_m}(E_0c)^3-\left[\frac{3\chi_i^2R^3}{(\varepsilon_i+2\varepsilon_m)^2}-\frac{\eta_iR^3}{\varepsilon_i+2\varepsilon_m}\right](E_0c)^5$$
$$+\left[\frac{12\chi_i^3R^3}{(\varepsilon_i+2\varepsilon_m)^3}-\frac{8\chi_i\eta_iR^3}{(\varepsilon_i+2\varepsilon_m)^2}\right](E_0c)^7\}r^{-2}\cos\theta\,, \quad \ldots (68)$$

where $\boldsymbol{c}=\dfrac{3\varepsilon_m}{\varepsilon_i+2\varepsilon_m}$ and $\boldsymbol{b}=\dfrac{\boldsymbol{R}^3(\varepsilon_i-\varepsilon_m)}{(\varepsilon_i+2\varepsilon_m)}$.

Note that our results in Eqs. (67) and (68) agree with those of Yu *et al.*[2, 7] obtained by using the power series method. Therefore, the third-order field equations and the boundary conditions in Sec. II are applicable and useful for further studies of more complicated nonlinear materials such as nonlinear materials with nonlinear spherical or cylindrical inclusions embedded in different nonlinear materials.

ANALYSIS OF THE ELECTROSTATIC POTENTIAL

Since we used the perturbation expansion method, which is an approximate means, to help solve the electrostatic potential, the results were analyzed as to whether the continuity condition of the electric displacement in normal direction on the inclusion surface $\left(\boldsymbol{D}_n^i/\boldsymbol{D}_n^m=1\right)$ are satisfied. From Eq. (1),

$$\frac{\boldsymbol{D}^i}{\varepsilon_i}=\acute{\boldsymbol{E}}^i+\boldsymbol{k}\left|\acute{\boldsymbol{E}}^i\right|^2\acute{\boldsymbol{E}}^i+\boldsymbol{p}\left|\acute{\boldsymbol{E}}^i\right|^4\acute{\boldsymbol{E}}^i\,, \quad \ldots (69)$$

where $\boldsymbol{k}=\dfrac{\chi_i}{\varepsilon_i}$ and $\boldsymbol{p}=\dfrac{\eta_i}{\varepsilon_i}$ are relative nonlinear coefficients which denote the nonlinearity of the inclusions.

The ratio of $\dfrac{D_n^i}{D_n^m}=\left[\dfrac{E_n^i+k\left|\acute{E}^i\right|^2E_n^i+p\left|\acute{E}^i\right|^4E_n^i}{E_n^m}\right]\left(\dfrac{\varepsilon_i}{\varepsilon_m}\right)$ for varying $\varepsilon_i/\varepsilon_m$ are determined for various values of $\boldsymbol{k}$ and $\boldsymbol{p}$ in unit of $\boldsymbol{E}_0^{-2}$ and $\boldsymbol{E}_0^{-4}$, respectively. It was found that more deviation of $\boldsymbol{D}_n^i/\boldsymbol{D}_n^m$ from one occurs with increasing nonlinearity ($\boldsymbol{k}$ and $\boldsymbol{p}$ are larger), as expected. Furthermore it was observed that the deviation of $\boldsymbol{D}_n^i/\boldsymbol{D}_n^m$ from one was reduced in the range of very large or small values of $\varepsilon_i/\varepsilon_m$ which is the case of high contrast materials.

We also note that the electrostatic potential, including the third–order potential, was generally more accurate than the result having only the first and the second-orders. This is the reason we expanded the electrostatic potential up to the third order. From all cases of our study, the perturbation method is valid for weakly nonlinear inclusion with the relative nonlinear coefficients in which the third and the fifth order ($\boldsymbol{k}$ and $\boldsymbol{p}$) are less than 0.4.

From Eq. (67), if χ_i and η_i are negative the electric field inside the inclusion is greater than the corresponding field in the linear case and the nonlinear electric response is enhanced.

CONCLUSIONS

We developed the field equations and the boundary conditions based on the third–order perturbation expansion for weakly nonlinear composite materials with the electric displacement and the electric field relation including the fifth-order nonlinear coefficient. The simple case of dilute nonlinear spherical inclusions embedded randomly in a linear host medium subjected to a uniform external electric field was considered. We obtained the electrostatic potential in the inclusion and host medium which are required to predict the effective property of the composite materials. Furthermore, the results were analyzed for validity. The electric response enhancement was also predicted for negative values of the third and the fifth-order nonlinear coefficients. The third–order field equation and the boundary condition given in this work are useful for further studies of more complicated composite materials of which higher–order effective nonlinear properties can not be ignored.[10-12]

ACKNOWLEDGMENT One of the authors (C.T.) thanks Graduate School Chulalongkorn University for partial support of this work.

REFERENCES

1. Liao, H. B., Wen, W. and Wong, G. K. L. (2003) "Preparation and optical characterization of Au/SiO_2 composite films with multilayer structure" *J. Appl. Phys.* **93**, 4485-4488.
2. Yu, K. W., Wang, Y. C., Hui, P. M. and Gu, G. Q. (1993) "Effective conductivity of nonlinear composites of spherical particles: A perturbation approach" *Phys. Rev. B* **47**, 1782-1787.
3. Yu, K. W. and Gu, G. Q. (1993) "Effective conductivity of nonlinear composites. II. Effective-medium approximation" *Phys. Rev. B* **47**, 7568-7571.
4. Zeng, X. C., Bergman, D. J., Hui, P. M. and Stroud, D. (1988) "Effective-medium theory for weakly nonlinear composites" *Phys. Rev. B* **38**, 10970-10973.
5. Tongsri, J. and Natenapit, M. (2005) "Effective nonlinear coefficient of strongly nonlinear spherical dielectric composites" *The First Graduate Congress of Mathematics and Physical Science*, PB04, Bangkok, Thailand.
6. Lee, H. C., Yuen, K. P. and Yu, K. W. (1995) "Percolative conduction in two-component strongly nonlinear composites" *Phys. Rev. B* **51**, 9317-9320.
7. Yu, K. W., Hui, P. M. and Stroud, D. (1993) "Effective dielectric response of nonlinear composites" *Phys. Rev. B* **47**, 14150-14156.
8. Yu, K. W., Hui, P. M. and Lee, H. C. (1996) "Decoupling approximation for strongly nonlinear composites" *Phys. Lett. A* **210**, 115-120.
9. Jackson, J. D. (1999) *Classical Electro-dynamics*, Third Ed. John Wiley & Sons, Inc. New York.
10. Hui, P. M. (1993) "Higher order nonlinear response in dilute random composites" *J. Appl. Phys.* **73**, 4072-4073.
11. Liu, X. and Li, Z. (1995) "High order nonlinear susceptibilities of composites medium" *Solid State Commun*. **96**, 981-985.
12. Marckmann, C. J., Ren, Y., Genty, G. G. and Kristensen, M. (2002) "Strength and symmetry of the third-order nonlinearity during poling of glass waveguides" *IEEE Photo. Tech. Lett*. **14**, 1294-1296.

Received: June 23, 2006
Accepted: September 6, 2006