Chapter 5

Effective Coefficients of Nonlinear Composites

In this chapter, we first confirm the general formulae of the effective nonlinear coefficients which have been derived in Chapter 4. We will apply these formulae to a simple case of nonlinear dielectric composite consisting of dilute weakly nonlinear cylindrical inclusions randomly dispersed in a linear host medium, and compare the results with the effective nonlinear coefficients which will be obtained by using the method of Gu and Yu [6]. After that, we will apply our formulae to a more complicated nonlinear composite consisting of dilute linear cylindrical inclusions randomly dispersed in nonlinear host medium.

5.1 The case of nonlinear inclusions in a linear host medium

We consider a simple case of nonlinear composite consisting of dilute weakly nonlinear cylindrical inclusions randomly dispersed in a linear host medium. A single inclusion model is used to determined the two dimension electrostatic potentials in the inclusion (ϕ^i) and host medium (ϕ^m). In this case $\chi_m = \eta_m = 0$ and the electric potentials can also be solved exactly since both potentials ϕ^i and ϕ^m satisfy Laplace equations. The potential in the inclusion can be written in the same form as Eq. (3.31)

$$\phi^i(r, \theta) = Cr\cos\theta, \tag{5.1}$$

where C is a constant coefficient. Eq. (5.1) implies that the electric field is uniform inside the inclusion, $\nabla\phi^i = C\hat{x}$. The potential ϕ^m for the host medium is similar to Eq. (3.32)

$$\phi^m(r, \theta) = (-E_0 r + Br^{-1})\cos\theta, \tag{5.2}$$

which automatically satisfies the boundary condition at remote distance and at the inclusion center. The constants B and C can be determined from boundary conditions Eqs. (3.10) and (3.11). We find that

$$Ba^{-1} - Ca = E_0 a, \tag{5.3}$$

and

$$\eta_i C^4 + \chi_i C^3 + \varepsilon_i C + \varepsilon_m B a^{-2} = -\varepsilon_m E_0. \tag{5.4}$$

Eliminating B, we obtain the equation for C:

$$\eta_i C^5 + \chi_i C^3 + \varepsilon C + 2E_0 \varepsilon_m = 0, \tag{5.5}$$

where $\varepsilon = \varepsilon_i + \varepsilon_m$. We therefore conclude that the results for electric potential are essentially the same as those of a linear composite. They differ only by the integration constants. In order to solve for C, we write C as a power series of E_0. We note that $C = -\frac{2\varepsilon_m E_0}{\varepsilon} = -cE_0$ when $\chi_i = \eta_i = 0$ as shown in Eqs. (3.31) and (3.32). We find

$$\begin{aligned} C = & -cE_0 + \left(\frac{\chi_i}{\varepsilon}\right) c^3 {E_0}^3 - \left[3\left(\frac{\chi_i}{\varepsilon}\right)^2 - \left(\frac{\eta_i}{\varepsilon}\right)\right] c^5 {E_0}^5 \\ & + \left[12\left(\frac{\chi_i}{\varepsilon}\right)^3 - 8\left(\frac{\chi_i}{\varepsilon}\right)\left(\frac{\eta_i}{\varepsilon}\right)\right] c^7 {E_0}^7 \\ & - \left[55\left(\frac{\chi_i}{\varepsilon}\right)^4 - 55\left(\frac{\chi_i}{\varepsilon}\right)^2\left(\frac{\eta_i}{\varepsilon}\right) + 5\left(\frac{\eta_i}{\varepsilon}\right)^2\right] c^9 {E_0}^9 + \dots . \end{aligned} \tag{5.6}$$

We also find that $B = (C + E_0)a^2$ which reduces to $-bE_0$ when $\chi_i = \eta_i = 0$. We find

$$\begin{aligned} B = & -bE_0 + \left(\frac{\chi_i}{\varepsilon}\right) a^2 c^3 {E_0}^3 - \left[3\left(\frac{\chi_i}{\varepsilon}\right)^2 - \left(\frac{\eta_i}{\varepsilon}\right)\right] a^2 c^5 {E_0}^5 \\ & + \left[12\left(\frac{\chi_i}{\varepsilon}\right)^3 - 8\left(\frac{\chi_i}{\varepsilon}\right)\left(\frac{\eta_i}{\varepsilon}\right)\right] a^2 c^7 {E_0}^7 \\ & - \left[55\left(\frac{\chi_i}{\varepsilon}\right)^4 - 55\left(\frac{\chi_i}{\varepsilon}\right)^2\left(\frac{\eta_i}{\varepsilon}\right) + 5\left(\frac{\eta_i}{\varepsilon}\right)^2\right] a^2 c^9 {E_0}^9 + \dots . \end{aligned} \tag{5.7}$$

For $\eta_i = 0$, Eqs. (5.6) and (5.7) are the same as the results of Gu and Yu [6]. Substituting C and B into Eqs. (5.1) and (5.2) respectively, we obtain the electric potentials in the inclusion and host medium as follows:

$$\phi^i(r,\theta) = \Big\{ -cE_0 + \Big(\frac{\chi_i}{\varepsilon}\Big)c^3{E_0}^3 - \Big[3\Big(\frac{\chi_i}{\varepsilon}\Big)^2 - \Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^5{E_0}^5 + \Big[12\Big(\frac{\chi_i}{\varepsilon}\Big)^3 - 8\Big(\frac{\chi_i}{\varepsilon}\Big)\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^7{E_0}^7 - \Big[55\Big(\frac{\chi_i}{\varepsilon}\Big)^4 - 55\Big(\frac{\chi_i}{\varepsilon}\Big)^2\Big(\frac{\eta_i}{\varepsilon}\Big) + 5\Big(\frac{\eta_i}{\varepsilon}\Big)^2\Big]c^9{E_0}^9 + \dots \Big\} r\cos\theta, \quad (5.8)$$

and

$$\phi^m(r,\theta) = -E_0 r\cos\theta + \Big\{ -bE_0 + \Big(\frac{\chi_i}{\varepsilon}\Big)a^2c^3{E_0}^3 - \Big[3\Big(\frac{\chi_i}{\varepsilon}\Big)^2 - \Big(\frac{\eta_i}{\varepsilon}\Big)\Big]a^2c^5{E_0}^5 + \Big[12\Big(\frac{\chi_i}{\varepsilon}\Big)^3 - 8\Big(\frac{\chi_i}{\varepsilon}\Big)\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]a^2c^7{E_0}^7 - \Big[55\Big(\frac{\chi_i}{\varepsilon}\Big)^4 - 55\Big(\frac{\chi_i}{\varepsilon}\Big)^2\Big(\frac{\eta_i}{\varepsilon}\Big) + 5\Big(\frac{\eta_i}{\varepsilon}\Big)^2\Big]a^2c^9{E_0}^9 + \dots \Big\} r^{-1}\cos\theta. \quad (5.9)$$

We will use these results to compute the effective coefficients up to the ninth order of this system. It should be noted that the terms with factors E_0, E_0^3, E_0^5, E_0^7 and E_0^9 in Eq. (5.9) are the zeroth, the first, the second, the third and the fourth-order potentials, respectively.

5.1.1 Effective coefficients by using the perturbation expansion method

We first determine the first-order effective coefficient, ε_e, from Eq. (4.9). However, as pointed out by Bergman [18], if the host medium has a large but finite volume $V = \pi R^2 L$, we therefore have to be careful when we calculate the quantities which arise from the host medium because there will actually be some corrections to these equations. These corrections are of two types: (1) small corrections, of relative order R^{-2}, that appear everywhere; (2) large corrections, of relative order 1 or more, that appear only near the surface of the composite. By using the zeroth-order electric potential, Eq. (4.9) becomes

$$\varepsilon_e = \frac{1}{VE_0^2}\Big[\int_0^L\int_0^{2\pi}\int_0^a \varepsilon_i|\nabla\phi_0^i|^2 r dr d\theta dz + \int_0^L\int_0^{2\pi}\int_a^R \varepsilon_m|\nabla\phi_0^m|^2 r dr d\theta dz\Big] + \frac{2\varepsilon_m(-b_0)V_i}{V}, \quad (5.10)$$

where the last term is the surface contribution term [19] and $V_i = \pi a^2 L$ is the volume of the inclusion. One finds, in the dilute limit ($V_i \ll V_m$),

$$\varepsilon_e = \varepsilon_m + 2\varepsilon_m p_i \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_i + \varepsilon_m}, \tag{5.11}$$

where $p_i = \frac{V_i}{V}$ is the inclusion volume fraction. Similarly, we can calculate the third-order effective coefficient, χ_e, from Eq. (4.11) by using the zeroth-order electric potential. In this simple case, there is no surface term in Eq. (4.11) because $\chi_m = 0$. Then Eq. (4.11) becomes

$$\chi_e = \frac{1}{VE_0^4}\Big[\int_0^L \int_0^{2\pi} \int_0^a \chi_i |\nabla\phi_0^i|^4 r dr d\theta dz\Big]. \tag{5.12}$$

We obtain χ_e in dilute limit ($V_i \ll V_m$) as

$$\chi_e = \chi_i p_i c^4. \tag{5.13}$$

We continue to calculate the high-order effective coefficients, η_e, δ_e and μ_e from Eqs. (4.13), (4.15) and (4.17), respectively. Again there are no surface terms in these equations because $\chi_m = \eta_m = 0$. Then Eqs. (4.13), (4.15) and (4.17) become, respectively,

$$\begin{aligned} \eta_e &= \frac{1}{VE_0^6}\Big[\int_0^L \int_0^{2\pi} \int_0^a \Big(\varepsilon_i |\nabla\phi_1^i|^2 + 4\chi_i |\nabla\phi_0^i|^2 \; \nabla\phi_0^i \cdot \nabla\phi_1^i + \eta_i |\nabla\phi_0^i|^6\Big) r dr d\theta dz \\ &+ \int_0^L \int_0^{2\pi} \int_a^R \varepsilon_m |\nabla\phi_1^m|^2 r dr d\theta dz\Big], \end{aligned} \tag{5.14}$$

$$\begin{aligned} \delta_e &= \frac{1}{VE_0^8}\Big[\int_0^L \int_0^{2\pi} \int_0^a \Big(2\varepsilon_i \nabla\phi_1^i \cdot \nabla\phi_2^i + 4\chi_i |\nabla\phi_0^i|^2 \; \nabla\phi_0^i \cdot \nabla\phi_2^i \\ &+ 2\chi_i |\nabla\phi_0^i|^2 |\nabla\phi_1^i|^2 + 4\chi_i (\nabla\phi_0^i \cdot \nabla\phi_1^i)^2 + 6\eta_i |\nabla\phi_0^i|^4 \; \nabla\phi_0^i \cdot \nabla\phi_1^i\Big) r dr d\theta dz \\ &+ \int_0^L \int_0^{2\pi} \int_a^R 2\varepsilon_i \nabla\phi_1^i \cdot \nabla\phi_2^i \; r dr d\theta dz\Big], \end{aligned} \tag{5.15}$$

and

$$\mu_e = \frac{1}{VE_0^{10}}\Big[\int_0^L\int_0^{2\pi}\int_0^a \Big(\varepsilon_i|\nabla\phi_2^i|^2 + 2\varepsilon_i\nabla\phi_1^i\cdot\nabla\phi_3^i + 4\chi_i|\nabla\phi_0^i|^2\,\nabla\phi_1^i\cdot\nabla\phi_2^i$$
$$+ 4\chi_i|\nabla\phi_0^i|^2\,\nabla\phi_0^i\cdot\nabla\phi_3^i + 4\chi_i|\nabla\phi_1^i|^2\,\nabla\phi_0^i\cdot\nabla\phi_1^i$$
$$+ 8\chi_i(\nabla\phi_0^i\cdot\nabla\phi_1^i)(\nabla\phi_0^i\cdot\nabla\phi_2^i) + 6\eta_i|\nabla\phi_0^i|^4\,\nabla\phi_0^i\cdot\nabla\phi_2^i$$
$$+ 12\eta_i|\nabla\phi_0^i|^2\,(\nabla\phi_0^i\cdot\nabla\phi_1^i)^2 + 3\eta_i|\nabla\phi_0^i|^4|\nabla\phi_1^i|^2\Big)rdrd\theta dz$$
$$+ \int_0^L\int_0^{2\pi}\int_a^R \Big(\varepsilon_m|\nabla\phi_2^m|^2 + 2\varepsilon_m\nabla\phi_1^m\cdot\nabla\phi_3^m\Big)rdrd\theta dz\Big]. \quad (5.16)$$

By using the zeroth, the first, the second and the third-order electric potentials from Eqs. (5.8) and (5.9), η_e, δ_e and μ_e can be calculated easily by using the mathematica program. Then we obtain the fifth, the seventh and the ninth-order effective nonlinear coefficients in dilute limit ($V_i \ll V_m$) as

$$\eta_e = -3\chi_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^6 + \eta_i p_i\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^6, \quad (5.17)$$

$$\delta_e = 12\chi_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)^2\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^8 - 8\eta_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^8, \quad (5.18)$$

and

$$\mu_e = -55\chi_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)^3\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^{10} + 55\eta_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)^2\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^{10}$$
$$-5\eta_i p_i\Big(\frac{\eta_i}{\varepsilon_i+\varepsilon_m}\Big)\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^{10}. \quad (5.19)$$

Our results for η_e, δ_e and μ_e in Eqs. (5.17)-(5.19) for $\eta_i = 0$ agree with those of Liu and Li [9] using decoupling approximation method.

5.1.2 Effective coefficients by using the method of Gu and Yu

In order to confirm our formulae for the seventh and the ninth-order effective nonlinear coefficients, Eqs. (4.15) and (4.17), we also follow and slightly extend the method of Gu and Yu [6] for effective coefficients to deal with nonlinear composite media

$$\frac{1}{V}\int_V \Big[\mathbf{D} - \mathbf{D}^m\Big]dV = \langle\mathbf{D}\rangle - \langle\mathbf{D}^m\rangle, \quad (5.20)$$

where V is the volume of the composite, $\langle \mathbf{D} \rangle$ and $\langle \mathbf{D}^m \rangle$ denote, respectively, the average electric displacement and average electric displacement in the host medium region with

$$\langle \mathbf{D} \rangle = \varepsilon_e \mathbf{E}_0 + \chi_e {E_0}^2 \mathbf{E}_0 + \eta_e {E_0}^4 \mathbf{E}_0 + \delta_e {E_0}^6 \mathbf{E}_0 + \mu_e {E_0}^8 \mathbf{E}_0. \tag{5.21}$$

Since the integrand on the left-hand side of Eq. (5.20) is zero in domain of the host medium (V_m), on applying electric displacement $\mathbf{D}$ from Eq. (2.2) and by using the average electric displacement $\langle \mathbf{D} \rangle$ definition of effective coefficients from Eq. (5.21), hence Eq. (5.20) becomes

$$\begin{aligned} \frac{1}{V} \int_{V_i} \Big[(\varepsilon_i - \varepsilon_m) \mathbf{E}^i + (\chi_i - \chi_m) |\mathbf{E}^i|^2 \, \mathbf{E}^i + (\eta_i - \eta_m) |\mathbf{E}^i|^4 \, \mathbf{E}^i \Big] dV \\ = (\varepsilon_e - \varepsilon_m) \mathbf{E}_0 + (\chi_e - \chi_m) {E_0}^2 \mathbf{E}_0 + (\eta_e - \eta_m) {E_0}^4 \mathbf{E}_0 \\ + (\delta_e - \delta_m) {E_0}^6 \mathbf{E}_0 + (\mu_e - \mu_m) {E_0}^8 \mathbf{E}_0. \end{aligned} \tag{5.22}$$

Because the electric field in the inclusion is uniform, the average electric field is the same as the electric field in the inclusion. We also use $\langle \mathbf{E} \rangle = \frac{1}{V} \int_V \mathbf{E}(\mathbf{x}) d^3x$ is equal to the external applied electric field ($\mathbf{E}_0$) for the boundary condition that $\mathbf{E}$ is uniform on the composite surface as shown in Appendix B. So Eq. (5.22) is valid for low inclusion concentration which the single inclusion model has been assumed.

For the case of linear medium ($\chi_m = \eta_m = \delta_m = \mu_m = 0$), then Eq. (5.22) becomes

$$\begin{aligned} \frac{1}{V} \int_{V_i} \Big[(\varepsilon_i - \varepsilon_m) \mathbf{E}^i + \chi_i |\mathbf{E}^i|^2 \, \mathbf{E}^i + \eta_i |\mathbf{E}^i|^4 \, \mathbf{E}^i \Big] dV \\ = (\varepsilon_e - \varepsilon_m) \mathbf{E}_0 + \chi_e {E_0}^2 \mathbf{E}_0 + \eta_e {E_0}^4 \mathbf{E}_0 + \delta_e {E_0}^6 \mathbf{E}_0 + \mu_e {E_0}^8 \mathbf{E}_0. \end{aligned} \tag{5.23}$$

By using the expansion for ϕ^i from Eq. (5.8), the electric field in the inclusion, $-\nabla \phi^i$, is

$$
\begin{aligned}
\mathbf{E}^i \;=\; & \Big\{cE_0 - \Big(\frac{\chi_i}{\varepsilon}\Big)c^3{E_0}^3 + \Big[3\Big(\frac{\chi_i}{\varepsilon}\Big)^2 - \Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^5{E_0}^5 \\
& - \Big[12\Big(\frac{\chi_i}{\varepsilon}\Big)^3 - 8\Big(\frac{\chi_i}{\varepsilon}\Big)\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^7{E_0}^7 \\
& + \Big[55\Big(\frac{\chi_i}{\varepsilon}\Big)^4 - 55\Big(\frac{\chi_i}{\varepsilon}\Big)^2\Big(\frac{\eta_i}{\varepsilon}\Big) + 5\Big(\frac{\eta_i}{\varepsilon}\Big)^2\Big]c^9{E_0}^9 + \dots\Big\}\hat{x}. \qquad (5.24)
\end{aligned}
$$

Substituting $\mathbf{E}^i$ from Eq. (5.24) into Eq. (5.23), we obtain

$$
\begin{aligned}
& \frac{1}{V}\int_{V_i}\Big[(\varepsilon_i - \varepsilon_m)\Big\{cE_0 - \Big(\frac{\chi_i}{\varepsilon}\Big)c^3{E_0}^3 + \Big[3\Big(\frac{\chi_i}{\varepsilon}\Big)^2 - \Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^5{E_0}^5 - \Big[12\Big(\frac{\chi_i}{\varepsilon}\Big)^3 \\
& \quad - 8\Big(\frac{\chi_i}{\varepsilon}\Big)\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^7{E_0}^7 + \Big[55\,\Big(\frac{\chi_i}{\varepsilon}\Big)^4 - 55\Big(\frac{\chi_i}{\varepsilon}\Big)^2\Big(\frac{\eta_i}{\varepsilon}\Big) + 5\Big(\frac{\eta_i}{\varepsilon}\Big)^2\Big]c^9{E_0}^9 + \dots\Big\} \\
& \quad + \chi_i\Big\{c^3E_0^3 - 3\Big(\frac{\chi_i}{\varepsilon}\Big)c^5{E_0}^5 + \Big[12\Big(\frac{\chi_i}{\varepsilon}\Big)^2 - 3\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^7{E_0}^7 \\
& \quad - \Big[55\,\Big(\frac{\chi_i}{\varepsilon}\Big)^3 - 30\Big(\frac{\chi_i}{\varepsilon}\Big)\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^9{E_0}^9 + \dots\Big\} + \eta_i\Big\{c^5{E_0}^5 - 5\Big(\frac{\chi_i}{\varepsilon}\Big)c^7{E_0}^7 \\
& \quad + \Big[25\Big(\frac{\chi_i}{\varepsilon}\Big)^2 - 5\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^9{E_0}^9 + \dots\Big\}\Big]dV \\
= \;& (\varepsilon_e - \varepsilon_m)E_o + \chi_e{E_0}^3 + \eta_e{E_0}^5 + \delta_e{E_0}^7 + \mu_e{E_0}^9. \qquad (5.25)
\end{aligned}
$$

Comparing the quantities both sides of Eq. (5.25) with the same power of E_0, the effective coefficients are determined.

To first order (of E_0), we have

$$
\varepsilon_e - \varepsilon_m = \frac{1}{V}\int_{V_i} c(\varepsilon_i - \varepsilon_m)dV, \qquad (5.26)
$$

and

$$
\varepsilon_e = \varepsilon_m + 2\varepsilon_m p_i \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_i + \varepsilon_m}, \qquad (5.27)
$$

where $p_i = \frac{V_i}{V}$ is the inclusion volume fraction.

To third order (of E_0^3), we have

$$
\chi_e = \frac{1}{V}\int_{V_i}\Big[-(\varepsilon_i - \varepsilon_m)\Big(\frac{\chi_i}{\varepsilon}\Big)c^3 + \chi_i c^3\Big]dV, \qquad (5.28)
$$

and

$$
\chi_e = \chi_i p_i c^4. \qquad (5.29)
$$

To fifth order (of E_0^5), we have

$$\eta_e = \frac{1}{V}\int_{V_i}\Big[(\varepsilon_i-\varepsilon_m)\Big[3\Big(\frac{\chi_i}{\varepsilon}\Big)^2-\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^5-3\chi_i\Big(\frac{\chi_i}{\varepsilon}\Big)c^5+\eta_i c^5\Big]dV, \quad (5.30)$$

and

$$\eta_e = -3\chi_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^6+\eta_i p_i\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^6. \quad (5.31)$$

To seventh order (of E_0^7), we have

$$\begin{aligned}\delta_e &= \frac{1}{V}\int_{V_i}\Big[-(\varepsilon_i-\varepsilon_m)\Big[12\Big(\frac{\chi_i}{\varepsilon}\Big)^3-8\Big(\frac{\chi_i}{\varepsilon}\Big)\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^7\\ &+ \chi_i\Big[12\Big(\frac{\chi_i}{\varepsilon}\Big)^2-3\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^7-5\eta_i\Big(\frac{\chi_i}{\varepsilon}\Big)c^7\Big]dV, \quad (5.32)\end{aligned}$$

and

$$\delta_e = 12\chi_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)^2\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^8-8\eta_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^8. \quad (5.33)$$

To ninth order (of E_0^9), we have

$$\begin{aligned}\mu_e &= \frac{1}{V}\int_{V_i}\Big[(\varepsilon_i-\varepsilon_m)\Big[55\Big(\frac{\chi_i}{\varepsilon}\Big)^4-55\Big(\frac{\chi_i}{\varepsilon}\Big)^2\Big(\frac{\eta_i}{\varepsilon}\Big)+5\Big(\frac{\eta_i}{\varepsilon}\Big)^2\Big]c^9-\chi_i\Big[55\Big(\frac{\chi_i}{\varepsilon}\Big)^3\\ &- 30\Big(\frac{\chi_i}{\varepsilon}\Big)\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^9+\eta_i\Big[25\Big(\frac{\chi_i}{\varepsilon}\Big)^2-5\Big(\frac{\eta_i}{\varepsilon}\Big)\Big]c^9\Big]dV, \quad (5.34)\end{aligned}$$

and

$$\begin{aligned}\mu_e = &-55\chi_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)^3\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^{10}+55\eta_i p_i\Big(\frac{\chi_i}{\varepsilon_i+\varepsilon_m}\Big)^2\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^{10}\\ &-5\eta_i p_i\Big(\frac{\eta_i}{\varepsilon_i+\varepsilon_m}\Big)\Big(\frac{2\varepsilon_m}{\varepsilon_i+\varepsilon_m}\Big)^{10}. \quad (5.35)\end{aligned}$$

We see that the results of δ_e and μ_e following the method of Gu and Yu [6] using the average electric displacement definition of effective coefficient, Eqs. (5.33) and (5.35), are the same as our results, Eqs. (5.18) and (5.19). These confirm our general formulae, Eqs. (4.15) and (4.17).

For the calculation of effective coefficients up to the ninth order by using the method of Gu and Yu [6], we can see that the ninth-order effective nonlinear coefficient (the last term of the right hand side of Eq. (5.23)), μ_e, is the coefficient of E_0^9. Therefore the electric potentials up to the fourth order are required in Eq. (5.25). It is more concise and less complicated to use our formulae, Eqs. (4.15) and (4.17), because it requires only the electric potentials up to the third order as also seen from Eqs. (5.15) and (5.16). We conclude that base on the electric potentials up to the same order, our formulae Eqs. (4.15) and (4.17), give more accurate results of high-order effective nonlinear coefficients than those obtained by using the method of Gu and Yu [6].

5.1.3 Application

We now apply our results, Eqs. (5.17), (5.18) and (5.19), to predict the response of the composite consisting of dilute weakly nonlinear cylindrical inclusions randomly dispersed in a linear host medium to an external uniform field. We consider a case of the inclusion which the $\mathbf{D}$ and $\mathbf{E}$ relation obey $\mathbf{D}^i = \varepsilon_i \mathbf{E}^i + \eta_i |\mathbf{E}^i|^4 \mathbf{E}^i$ with $\chi_i = 0$ and the host medium has the linear coefficient ε_m. In this case δ_e from Eq. (5.18) becomes zero and the relative fifth-order and the relative ninth-order coefficients, η_e/η_i and $\mu_e E_0^4/\eta_i$ are determined.

In Fig. 5.1, we report the relation between η_e/η_i and $\varepsilon_m/\varepsilon_i$ for inclusion packing fractions $p_i = 0.05$ and 0.1. It shows that as $\varepsilon_m/\varepsilon_i < 0.6$ the fifth-order effective nonlinear coefficient is negligible. The relative ninth-order effective nonlinear coefficient ($\mu_e E_0^4/\eta_i$) is plotted against $\varepsilon_m/\varepsilon_i$ for $\eta_i E_0^4/\varepsilon_i = 0.1$, satisfying the weakly nonlinear property, as shown in Fig. 5.2. This indicates that the less important of the ninth-order effective nonlinear coefficient occurs as $\varepsilon_m/\varepsilon_i$ decreasing and it approaches zero for $\varepsilon_m/\varepsilon_i < 0.7$. From these results, we can conclude that the effective higher-order nonlinear coefficients cannot be ignored, especially for $\varepsilon_m/\varepsilon_i > 1$ and p_i approaches 0.1.

In Fig. 5.3, we shows the difference of the ninth-order effective nonlinear coefficient (μ_e) calculated by the method of Gu and Yu [6] and our method (Eq. (4.17)) by using the electric potential (Eqs. (5.8) and (5.9)) up to only the third order. We can see that the value of μ_e calculated by our method (Eq. (4.17)) and the method of Gu and Yu [6] is equal to each other at $\varepsilon_m/\varepsilon_i = 1$. In the range $0.1 < \varepsilon_m/\varepsilon_i < 1.4$, the value of μ_e calculated by the method of Gu and Yu [6] and our method (Eq. (4.17)) are very close to each other. The value of μ_e obtained from the method of Gu and Yu [6] and our method (Eq. (4.17)) highly different to each other, especially for $\varepsilon_m/\varepsilon_i > 1.4$ and p_i approaches 0.1.

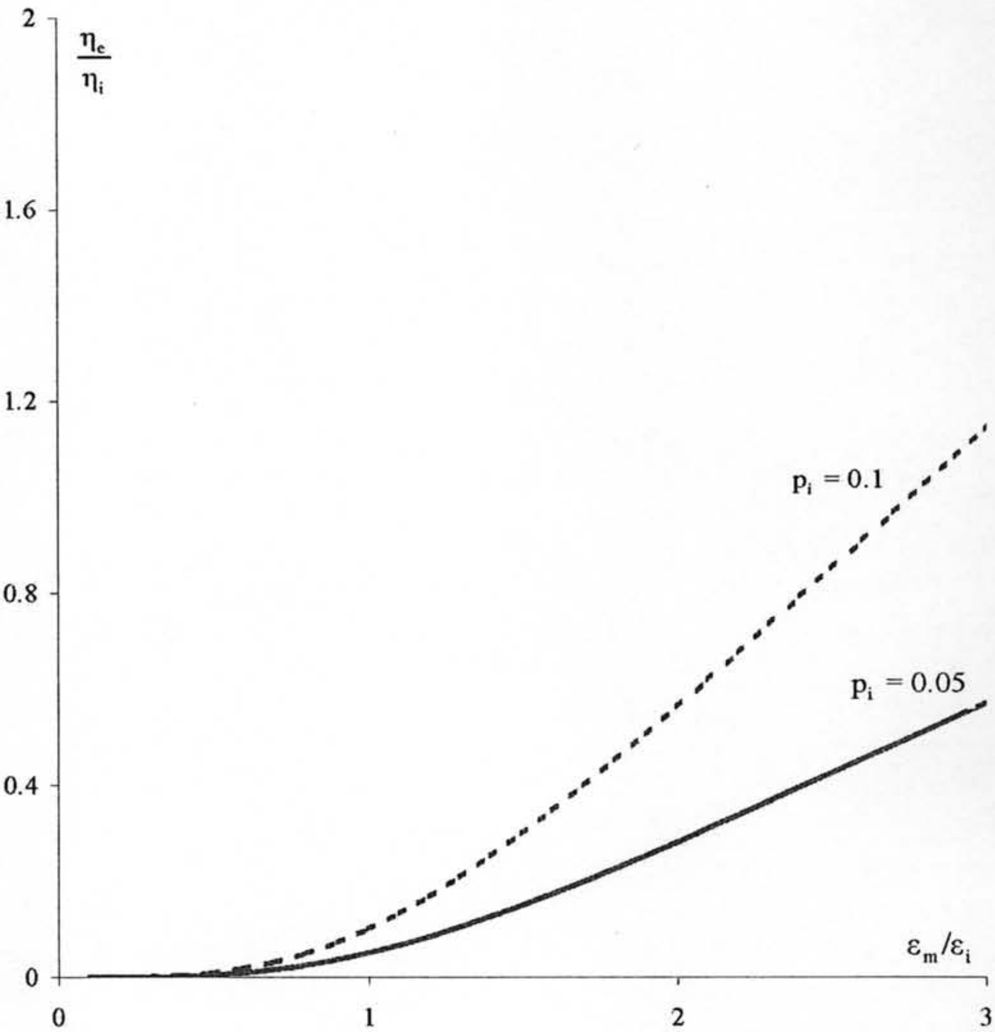

Figure 5.1: The relative fifth-order nonlinear coefficient for packing fraction $p_i = 0.05$ and 0.1.

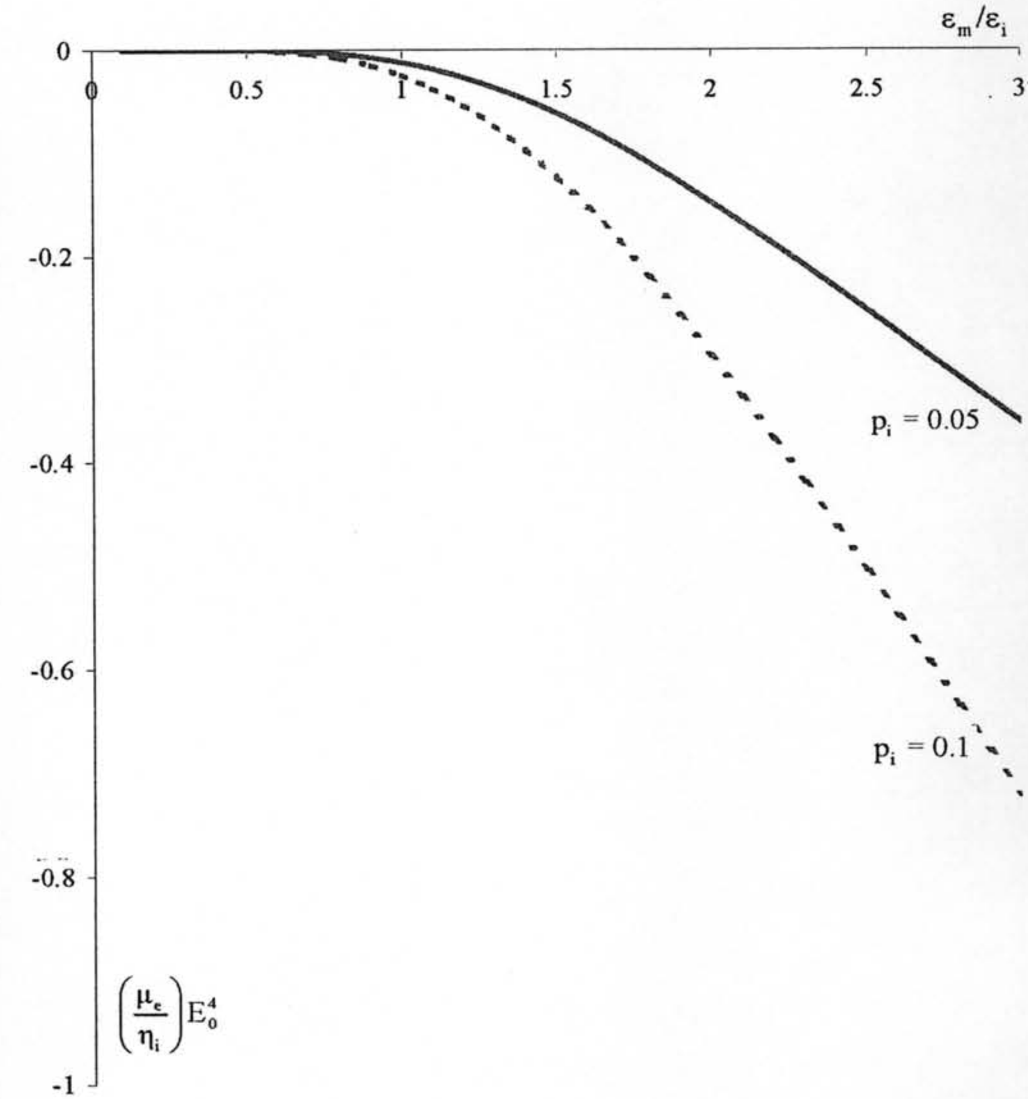

Figure 5.2: The relative ninth-order nonlinear coefficient for packing fraction $p_i = 0.05$ and 0.1.

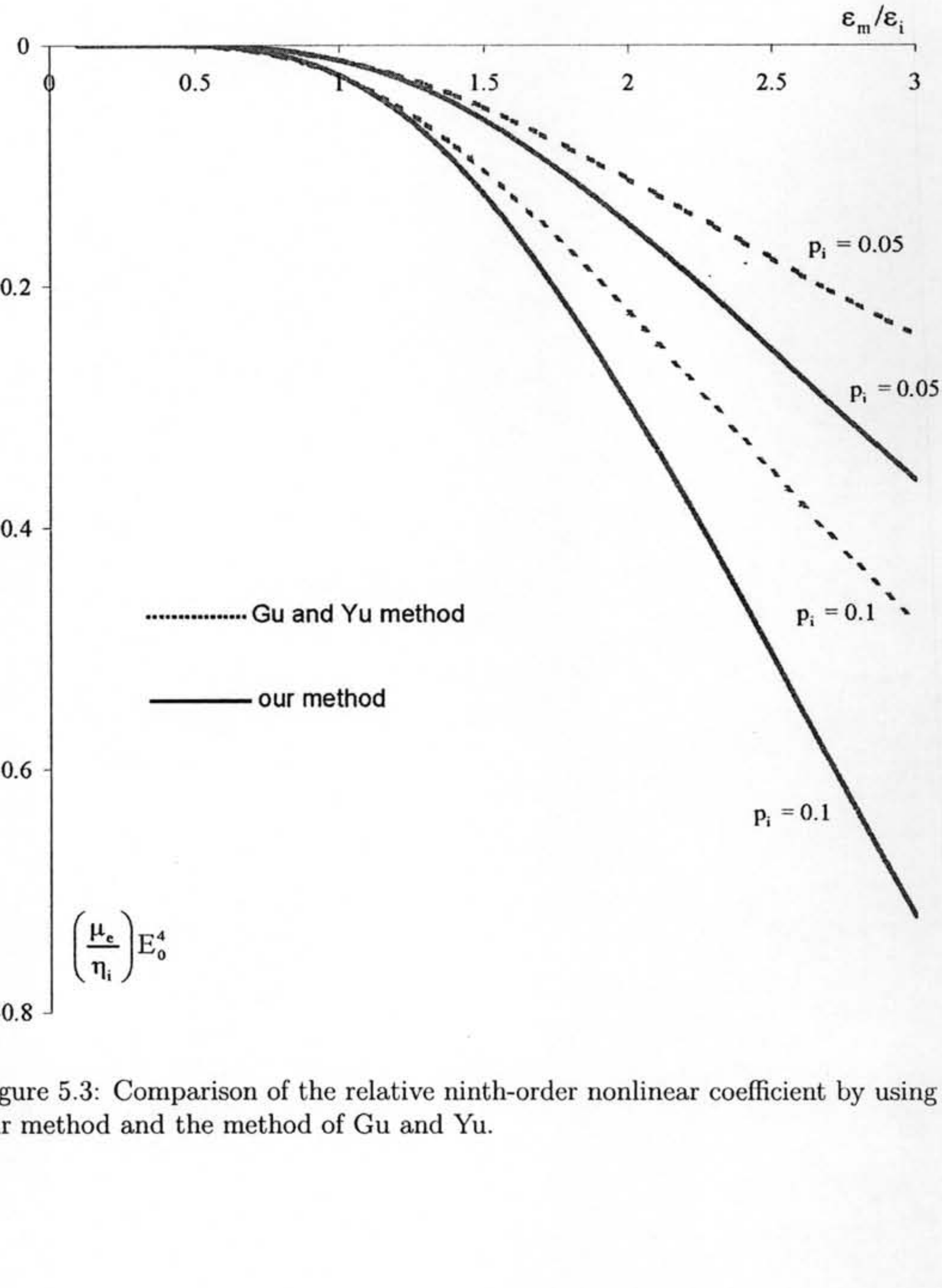

Figure 5.3: Comparison of the relative ninth-order nonlinear coefficient by using our method and the method of Gu and Yu.

5.2 The case of linear inclusions in a nonlinear host medium

In this case, we will use the solutions of electric potential up to the third order which has been obtained in chapter 3 to calculate the effective nonlinear coefficients up to the ninth order. For the first-order effective coefficient ε_e, the result is the same as Eq. (5.11). We first calculate the third-order effective coefficient from Eq. (4.11). For $\chi_i = 0$, Eq. (4.11) becomes

$$\chi_e = \frac{1}{VE_0^4}\Big[\int_0^L\int_0^{2\pi}\int_a^R \chi_m|\nabla\phi_0^m|^4 r dr d\theta dz\Big] + \frac{4\chi_m(-b_0)V_i}{V}, \tag{5.36}$$

where the last term is the surface term [19] and $b_0 = ba^{-2} = \frac{\varepsilon_m-\varepsilon_i}{\varepsilon_i+\varepsilon_m}$ is a factor related to the induced dipole moment due to the applied far electric field. Substituting ϕ_0^m from Eq. (3.32), we obtain the third-order effective coefficient in dilute limit as

$$\chi_e = \chi_m + \chi_m p_i\Big[-1-4b_0+4b_0^2+\frac{1}{3}b_0^4\Big]. \tag{5.37}$$

Our result for χ_e in Eq. (5.37) agrees with that of Bergman, Eq. (3.2) of Ref. [18] for $d=2$.

We continue to calculate the high-order effective nonlinear coefficients, η_e and δ_e from Eqs. (4.13) and (4.15), respectively. We consider a case of the nonlinear medium has only the third-order nonlinearity which the $\mathbf{D}$ and $\mathbf{E}$ relation obey $\mathbf{D}^m = \varepsilon_m\mathbf{E}^m + \chi_m|\mathbf{E}^m|^2\mathbf{E}^m$ and the inclusion has the linear coefficient ε_i. For $\chi_i = 0$ and $\eta_i = 0$, Eqs. (4.13) and (4.15) become, respectively,

$$\begin{aligned}\eta_e &= \frac{1}{VE_0^6}\Big[\int_0^L\int_0^{2\pi}\int_a^R\Big(\varepsilon_m|\nabla\phi_1^m|^2+4\chi_m|\nabla\phi_0^m|^2\;\nabla\phi_0^m\cdot\nabla\phi_1^m\Big)r dr d\theta dz \\ &+ \int_0^L\int_0^{2\pi}\int_0^a \varepsilon_i|\nabla\phi_1^i|^2 r dr d\theta dz\Big],\end{aligned} \tag{5.38}$$

and

$$\delta_e = \frac{1}{VE_0^8}\Big[\int_0^L\int_0^{2\pi}\int_a^R\Big(2\varepsilon_m\nabla\phi_1^m\cdot\nabla\phi_2^m + 4\chi_m|\nabla\phi_0^m|^2\;\nabla\phi_0^m\cdot\nabla\phi_2^m + 2\chi_m|\nabla\phi_0^m|^2|\nabla\phi_1^m|^2 + 4\chi_m(\nabla\phi_0^m\cdot\nabla\phi_1^m)^2\Big)rdrd\theta dz + \int_0^L\int_0^{2\pi}\int_0^a 2\varepsilon_i\nabla\phi_1^i\cdot\nabla\phi_2^i\; rdrd\theta dz\Big]. \quad (5.39)$$

By using the zeroth, the first and the second-order electric potentials in the inclusion and host medium from Eqs. (3.31), (3.32), (3.52), (3.55), (3.74) and (3.75), η_e and δ_e can be calculated by using the mathematica program. Then we obtain the analytic form solution of the fifth and the seventh-order effective nonlinear coefficients in dilute limit as

$$\eta_e = -\frac{209p_i\varepsilon_i^7\chi_m^2}{5(\varepsilon_i+\varepsilon_m)^8} - \frac{93p_i\varepsilon_i^8\chi_m^2}{10\varepsilon_m(\varepsilon_i+\varepsilon_m)^8} - \frac{271p_i\varepsilon_i^6\varepsilon_m\chi_m^2}{5(\varepsilon_i+\varepsilon_m)^8} + \frac{97p_i\varepsilon_i^5\varepsilon_m^2\chi_m^2}{15(\varepsilon_i+\varepsilon_m)^8} + \frac{108p_i\varepsilon_i^4\varepsilon_m^3\chi_m^2}{(\varepsilon_i+\varepsilon_m)^8} + \frac{1847p_i\varepsilon_i^3\varepsilon_m^4\chi_m^2}{15(\varepsilon_i+\varepsilon_m)^8} + \frac{557p_i\varepsilon_i^2\varepsilon_m^5\chi_m^2}{15(\varepsilon_i+\varepsilon_m)^8} - \frac{119p_i\varepsilon_i\varepsilon_m^6\chi_m^2}{5(\varepsilon_i+\varepsilon_m)^8} - \frac{529p_i\varepsilon_m^7\chi_m^2}{30(\varepsilon_i+\varepsilon_m)^8}, \quad (5.40)$$

and

$$\delta_e = \frac{29621p_i\varepsilon_i^9\chi_m^3}{45(\varepsilon_i+\varepsilon_m)^{11}} + \frac{55p_i\varepsilon_i^{11}\chi_m^3}{\varepsilon_m^2(\varepsilon_i+\varepsilon_m)^{11}} + \frac{12643p_i\varepsilon_i^{10}\chi_m^3}{45\varepsilon_m(\varepsilon_i+\varepsilon_m)^{11}} + \frac{41477p_i\varepsilon_i^8\varepsilon_m\chi_m^3}{45(\varepsilon_i+\varepsilon_m)^{11}} + \frac{32654p_i\varepsilon_i^7\varepsilon_m^2\chi_m^3}{45(\varepsilon_i+\varepsilon_m)^{11}} - \frac{1274p_i\varepsilon_i^6\varepsilon_m^3\chi_m^3}{9(\varepsilon_i+\varepsilon_m)^{11}} - \frac{9758p_i\varepsilon_i^5\varepsilon_m^4\chi_m^3}{9(\varepsilon_i+\varepsilon_m)^{11}} - \frac{11102p_i\varepsilon_i^4\varepsilon_m^5\chi_m^3}{9(\varepsilon_i+\varepsilon_m)^{11}} - \frac{26281p_i\varepsilon_i^3\varepsilon_m^6\chi_m^3}{45(\varepsilon_i+\varepsilon_m)^{11}} + \frac{3407p_i\varepsilon_i^2\varepsilon_m^7\chi_m^3}{45(\varepsilon_i+\varepsilon_m)^{11}} + \frac{10321p_i\varepsilon_i\varepsilon_m^8\chi_m^3}{45(\varepsilon_i+\varepsilon_m)^{11}} + \frac{1451p_i\varepsilon_m^9\chi_m^3}{15(\varepsilon_i+\varepsilon_m)^{11}}, \quad (5.41)$$

which are the new results of this work.

We now apply our results of χ_e (Eq. (5.37)), η_e (Eq. (5.40)) and δ_e (Eq. (5.41)) to predict the response of weakly nonlinear composite consisting of dilute linear cylindrical inclusion randomly dispersed in a nonlinear host medium. χ_e,

η_e and δ_e from Eqs. (5.37), (5.40) and (5.41) can be rewritten in the form as

$$\frac{\chi_e}{\chi_m} = 1 - p_i + \frac{p_i}{(\varepsilon_r+1)}\Big[4\varepsilon_r - 4\Big] + \frac{p_i}{(\varepsilon_r+1)^2}\Big[4\varepsilon_r^2 - 8\varepsilon_r + 4\Big] + \frac{p_i}{(\varepsilon_r+1)^4}\Big[\frac{\varepsilon_r^4}{3} - \frac{4\varepsilon_r^3}{3} + 2\varepsilon_r^2 - \frac{4\varepsilon_r}{3} + \frac{1}{3}\Big], \tag{5.42}$$

$$\frac{\eta_e E_0^4}{\varepsilon_m} = \frac{p_i\chi_r^2}{(\varepsilon_r+1)^8}\Big[-\frac{93\varepsilon_r^8}{10} - \frac{209\varepsilon_r^7}{5} - \frac{271\varepsilon_r^6}{5} + \frac{97\varepsilon_r^5}{15} + 108\varepsilon_r^4 + \frac{1847\varepsilon_r^3}{15} + \frac{557\varepsilon_r^2}{15} - \frac{119\varepsilon_r}{5} - \frac{529}{30}\Big], \tag{5.43}$$

and

$$\frac{\delta_e E_0^6}{\varepsilon_m} = \frac{p_i\chi_r^3}{(\varepsilon_r+1)^{11}}\Big[55\varepsilon_r^{11} + \frac{12643\varepsilon_r^{10}}{45} + \frac{29621\varepsilon_r^9}{45} + \frac{41477\varepsilon_r^8}{45} + \frac{32654\varepsilon_r^7}{45} - \frac{1274\varepsilon_r^6}{9} - \frac{9758\varepsilon_r^5}{9} - \frac{11102\varepsilon_r^4}{9} - \frac{26281\varepsilon_r^3}{45} + \frac{3407\varepsilon_r^2}{45} + \frac{10321\varepsilon_r}{45} + \frac{1451}{15}\Big], \tag{5.44}$$

where $\varepsilon_r = \frac{\varepsilon_i}{\varepsilon_m}$ and $\chi_r = \frac{\chi_m E_0^2}{\varepsilon_m}$.

In Fig. 5.4, we report the relation between χ_e/χ_m and $\varepsilon_i/\varepsilon_m$ for inclusion packing fractions $p_i = 0$, 0.05 and 0.1. It shows that the relative third-order effective nonlinear coefficient is decreasing as $\varepsilon_i/\varepsilon_m$ increasing from 0 to $\simeq 0.4$ and increasing as $\varepsilon_i/\varepsilon_m > 0.4$. We can see that all three curves pass a common point at $\varepsilon_i/\varepsilon_m \simeq 1.6$ and χ_e/χ_m is minimum at $\varepsilon_i/\varepsilon_m \simeq 0.4$ for $p_i = 0.05$ and 0.1. For $p_i = 0$, $\chi_e/\chi_m = 1$ (or $\chi_e = \chi_m$), as expected. The relative fifth-order effective nonlinear coefficient ($\eta_e E_0^4/\varepsilon_m$) is plotted against $\varepsilon_i/\varepsilon_m$ as shown in Fig. 5.5 for $\chi_m E_0^2/\varepsilon_m = 0.1$, satisfying the weakly nonlinear property. This indicates that η_e is maximize for $p_i = 0.05$ and 0.1, at $\varepsilon_i/\varepsilon_m \simeq 0.8$. All three curves have two common points at $\varepsilon_i/\varepsilon_m \simeq 0.5$ and 1.3 with $\eta_e = 0$. In Fig. 5.6, the relative seventh-order effective nonlinear coefficient is plotted against $\varepsilon_i/\varepsilon_m$ for $\chi_m E_0^2/\varepsilon_m = 0.1$, satisfying the weakly nonlinear property. It shows that δ_e is minimum at $\varepsilon_i/\varepsilon_m \simeq 0.7$. All three curves have two common points at $\varepsilon_i/\varepsilon_m \simeq 0.6$ and 1 with $\delta_e = 0$. From these results, we can conclude that the effective higher-order nonlinear coefficients cannot be ignored, especially for p_i approaches 0.1. These results show that the high-order effective nonlinear coefficients are important as pointed out by many authors for experimental works [3,4,13,14]. For example, high order optical nonlinearities of C_{60}– and C_{70}–toluene solutions were observed by Koudoumas *et al.* [13]. In their work, high order nonlinear coefficients depend on the laser intensity, corresponding to E_0^2, and the concentration of the sample. The value of the effective nonlinear coefficients of the sample up to the ninth-order have been reported.

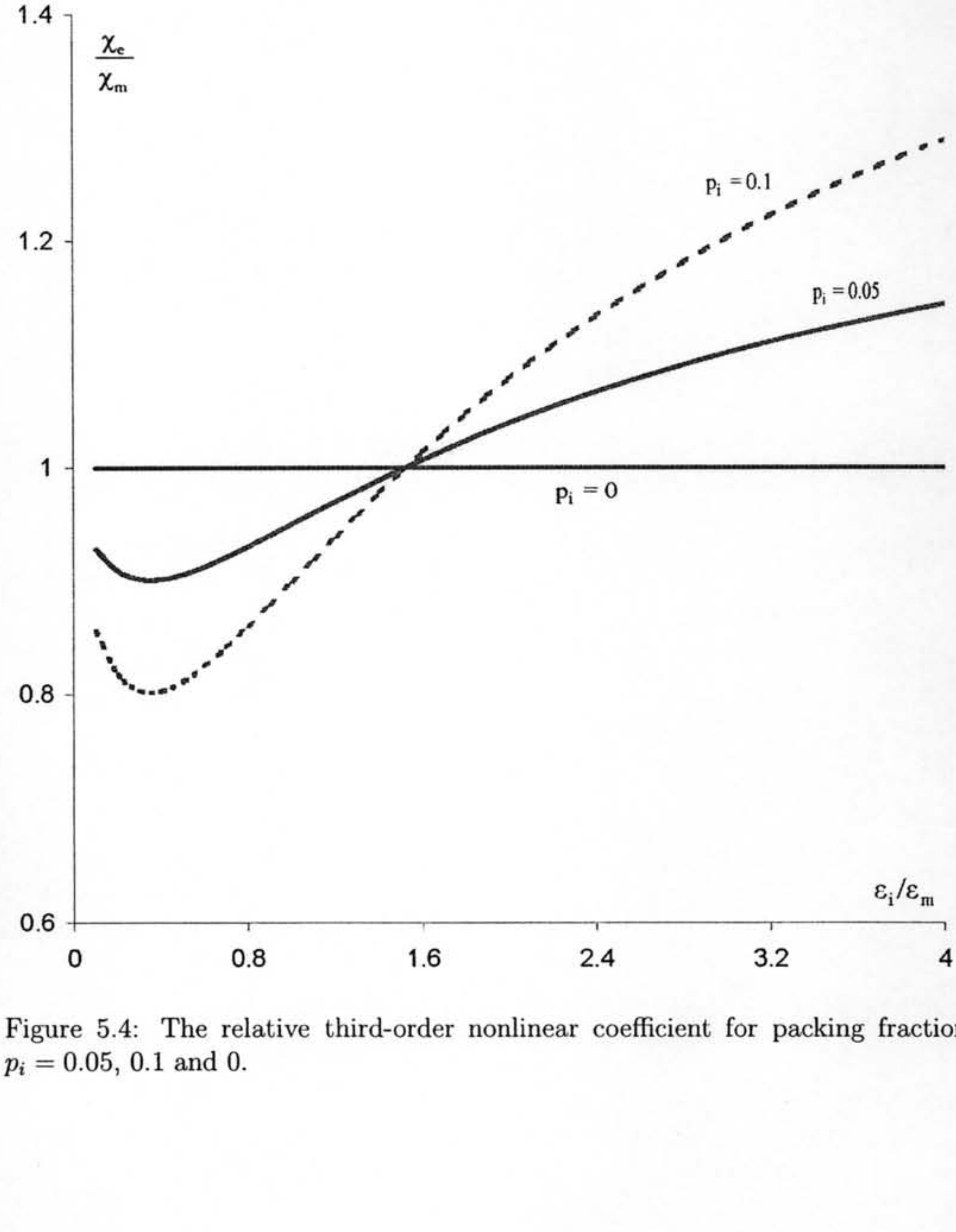

Figure 5.4: The relative third-order nonlinear coefficient for packing fraction $p_i = 0.05$, 0.1 and 0.

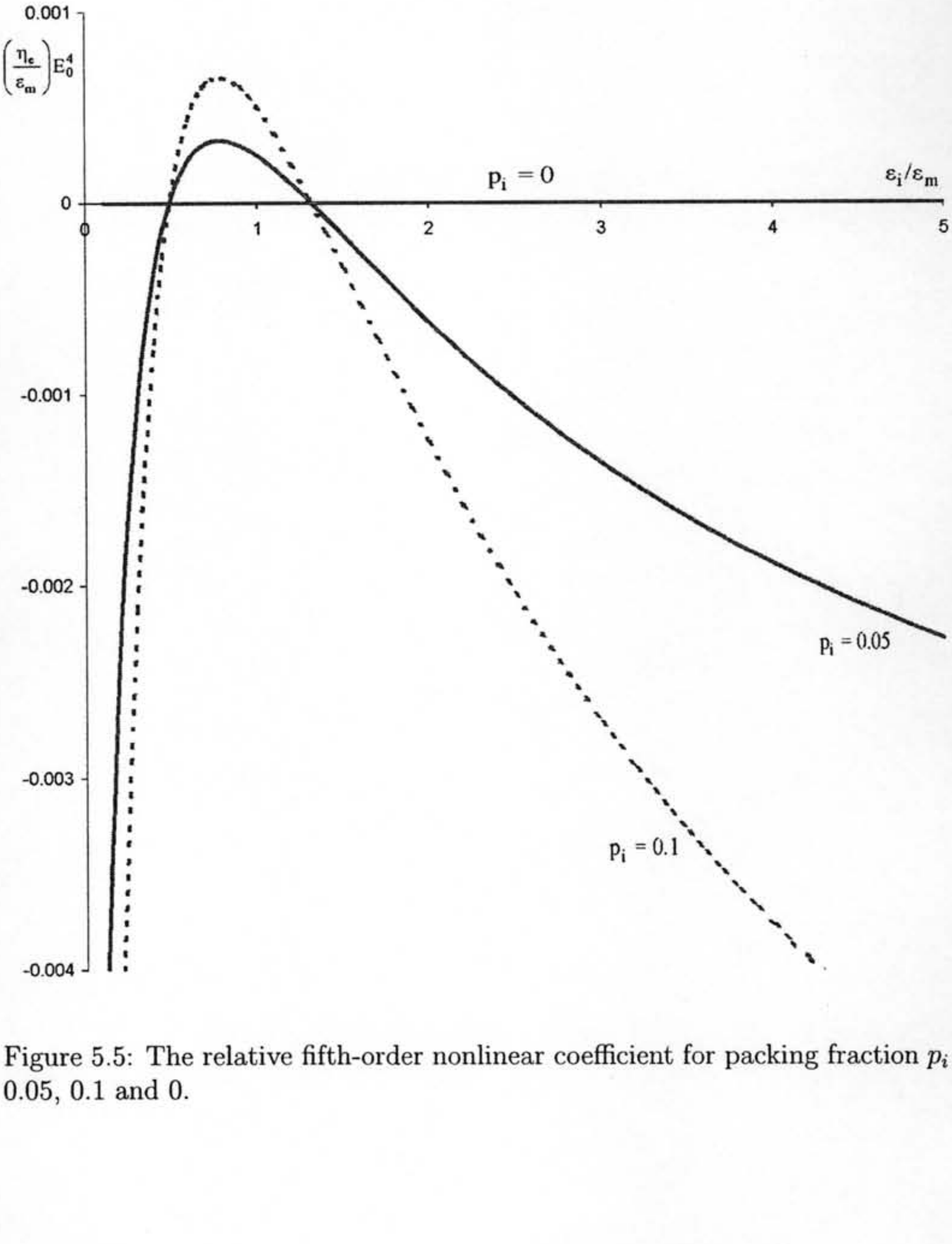

Figure 5.5: The relative fifth-order nonlinear coefficient for packing fraction $p_i =$ 0.05, 0.1 and 0.

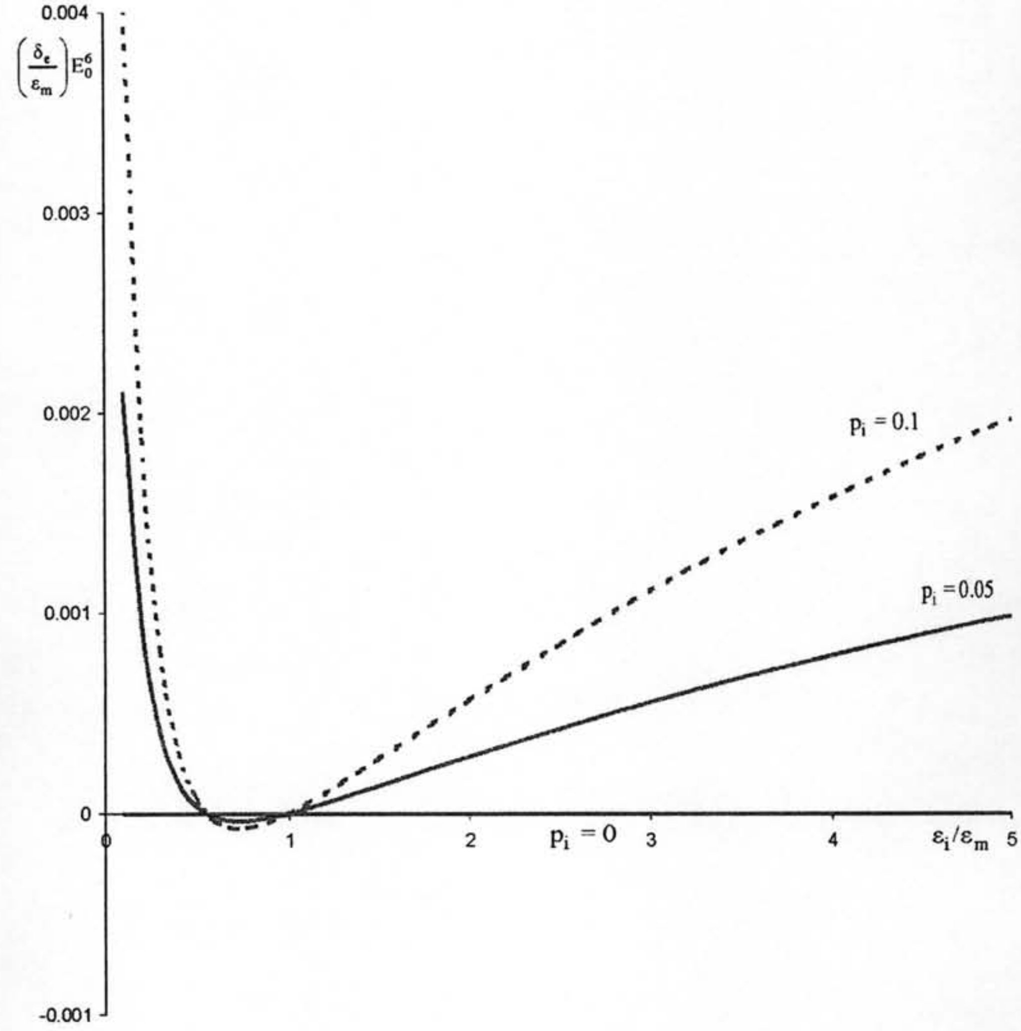

Figure 5.6: The relative seventh-order nonlinear coefficient for packing fraction $p_i = 0.05$, 0.1 and 0.