REFERENCES

[1] J. Thongsee and M. Natenapit. Effective nonlinear coefficients of strongly nonlinear dielectric composites. J. Appl. Phys. **101** (2007): 24103.

[2] L. Gao, Y. Y. Huang and Z. Y. Li. Effective medium approximation for strongly nonlinear composite media with shape distribution. Phys. Lett. A **306** (2003): 337.

[3] Y. F. Chen, K. Beckwitt, F. W. Wise, B. G. Aitken, J. S. Sanghera and I. D. Aggarwal. Measurement of fifth- and seventh-order nonlinearities of glasses. J. Opt. Soc. Am. B **23** (2006): 347.

[4] E. L. Filho, C. B. Araujo and J. J. Rodrigues. High-order nonlinearities of aqueous colloids containing silver nanoparticles. J. Opt. Soc. Am. B **24** (2007): 2948.

[5] H. B. Liao, W. W. Wen and G. K. Wong. Preparation and optical characterization of Au/SiO_2 composite films with multilayaer structure. J. Appl. Phys. **93** (2003): 4485.

[6] G. Q. Gu and K. W. Yu. Effective conductivity of nonlinear composites. Phys. Rev. B **46** (1992): 4502.

[7] K.W. Yu, Y.C. Wang, P.M. Hui and G.Q. Gu. Effective conductivity of nonlinear composites of spherical particles: A perturbation approach. Phys. Rev. B **47** (1993): 1782.

[8] K.W. Yu, P.M. Hui and D. Stroud. Effective dielectric response of nonlinear composites. Phys. Rev. B **47** (1993): 14150.

[9] X. Y. Liu and Z. Y. Li. High order nonlinear susceptibilities of composite medium. Solid State Comm. **96** (1995): 981.

[10] X. C. Zeng, D. J. Bergman, P. M. Hui and D. Stroud. Effective-medium theory for weakly nonlinear composites. Phys. Rev. B **38** (1988): 10970.

[11] L. Gao. Spectral representation theory for higher-order nonlinear response in random composites. Phys. Lett. A **322** (2004): 250.

[12] L. P. Gu, L. Gao and Z. Y. Li. Spectral representation theory for higher order nonlinear responses in random composites with arbitary nonlinearity. phys. stat. sol. (b) **241** (2004): 1115.

[13] E. Koudoumas, F. Dong, S. Couris and S. Leach. High order nonlinear optical response of fullerene solutions in the nanosecond regime. Opt. Comm. **138** (1997): 301.

[14] O. Ormachea. Comparative analysis of multi-wave mixing and measurements of higher-order nonlinearities in resonant media. Opt. Comm. **268** (2006): 317.

[15] M. Natenapit, A. Chitranondh and C. Thongboonrithi. Third-order perturbation for weakly nonlinear dielectric composites of spherical inclusion. J. Sci. Res. Chula. Univ. **31** (2006): 105.

[16] D. W. Jordan and P. Smith. Nonlinear Ordinary Differential Equations. Oxford: Clarendon Press, 1977.

[17] M. H. Nayfeh and M. K. Brussel. Electricity and Magnetism. New York: Wiley, 1985.

[18] D. J. Bergman. Nonlinear behavior and $1/f$ noise near a conductivity threshold: Effects of local microgeometry. Phys. Rev. B **39** (1989): 4598.

[19] K. W. Yu and G. Q. Gu. Effective conductivity of strongly nonlinear composites: variational approach. Phys. Lett. A **205** (1995): 295.

APPENDICES

Appendix A

Particular Solutions for ϕ_1^m, ϕ_2^m and ϕ_3^m

In this research, the electric potential equations for the host medium region (ϕ_1^m, ϕ_2^m and ϕ_3^m), Eqs. (3.34), (3.59) and (3.89), are in the form as

$$\nabla^2\phi(r,\theta) = \sum_j f_j(r)\cos(j\theta). \tag{A-1}$$

Consider (A-1) by using the cylindrical coordinates in two dimensions, r and θ, we obtain

$$\Big[\frac{1}{r}\frac{\partial}{\partial r}(r\frac{\partial}{\partial r}) + \frac{1}{r^2}\frac{\partial^2}{\partial \theta^2}\Big]\phi(r,\theta) = \sum_j f_j(r)\cos(j\theta). \tag{A-2}$$

Let the particular solution of $\phi(r,\theta)$ be the summation of

$$\phi_p(r,\theta) = R_j(r)\cos(j\theta). \tag{A-3}$$

By substituting (A-3) into (A-2), we obtain

$$\begin{aligned}\Big[\frac{1}{r}\frac{d}{dr}\Big(r\frac{dR_j(r)}{dr}\Big) - \frac{j^2}{r^2}R_j(r)\Big]\cos(j\theta) &= f_j(r)\cos(j\theta),\\ \frac{1}{r}\frac{d}{dr}\Big(r\frac{dR_j(r)}{dr}\Big) - \frac{j^2}{r^2}R_j(r) &= f_j(r).\end{aligned} \tag{A-4}$$

From considering the electric potential equations, Eqs. (3.34), (3.59) and (3.89), we find that $f_j(r)$ is in the form as

$$f_j(r) = C_j r^k, \tag{A-5}$$

where C_j is the constant coefficient of r^k and k is the power of r.
Substituting (A-5) into (A-4), we obtain

$$\frac{d^2R_j(r)}{dr^2} + \frac{1}{r}\frac{dR_j(r)}{dr} - \frac{j^2}{r^2}R_j(r) = C_j r^k. \tag{A-6}$$

We let the solution of $R_j(r)$ be

$$R_j(r) = \alpha_j r^{k+2}, \tag{A-7}$$

where α is the constant coefficient which have to be solved.

Substituting (A-7) into (A-6), we obtain

$$\alpha_j = \frac{C_j}{(k+2)(k+1)+(k+2)-j^2}. \tag{A-8}$$

Since the constant C_j, k, and j are known, then α_j can be obtained. The particular solution of (A-1) can be solved by substituting (A-7) and (A-8) into (A-3).

From α_j in (A-8), we see that the problem of solving the particular solution occurs when

$$(k+2)(k+1)+(k+2)-j^2 = 0, \tag{A-9}$$

or

$$k+2 = \pm j. \tag{A-10}$$

We cannot use (A-7) and (A-8) to solve the particular solution when the relation between k and j is in the form of (A-10). Next, we will solve this problem by considering (A-4) again.

From $d(\ln r) = \frac{1}{r}dr$, (A-4) can be rewritten as

$$\frac{d^2R_j(r)}{d(\ln r)^2} - j^2R_j(r) = r^2 f_j(r) = r^2(C_j r^k). \tag{A-11}$$

We let $x = \ln r$ and $r = e^x$. So (A-11) becomes

$$\frac{d^2R'_j}{dx^2} - j^2R'_j = C_j e^{\pm jx}, \tag{A-12}$$

which the trial solution is

$$R'_j = \pm\frac{C_j}{2j}xe^{\pm jx}. \tag{A-13}$$

Substituting $x = \ln r$, we obtain

$$R_j(r) = \pm\frac{C_j}{2j}r^{\pm j}\ln r. \tag{A-14}$$

Next, we will apply these formulae to solve the particular solution for the electric potential in the host medium region, $\phi^m_{1p}(r,\theta)$, $\phi^m_{2p}(r,\theta)$ and $\phi^m_{3p}(r,\theta)$.

The particular solution for the first-order electric potential

We first to calculate the particular solution for the first order electric potential in the host medium region from Eq. (3.34) which is

$$\nabla^2\phi_1^m(r,\theta) = -\frac{1}{\varepsilon_m}\Big[(8b^2r^{-5} - 4b^3r^{-7})\cos\theta - 4br^{-3}\cos 3\theta\Big]E_0^3, \tag{A-15}$$

where ε_m is in unit of β_m.

For convenient, we rewrite (A-15) as

$$\nabla^2\phi_1^m(r,\theta) = -\frac{1}{\varepsilon_m}\Big[f_1 + f_2 + f_3\Big]E_0^3, \tag{A-16}$$

where

$$f_1 = 8b^2r^{-5}\cos\theta, \tag{A-17}$$

$$f_2 = -4b^3r^{-7}\cos\theta, \tag{A-18}$$

$$f_3 = -4br^{-3}\cos 3\theta. \tag{A-19}$$

From (A-3), (A-7) and (A-8), the particular solution of $\phi_1^m(r,\theta)$ is in the form

$$\phi_{1p}^m(r,\theta) = -\frac{1}{\varepsilon_m}\Big[\alpha_1 r^{-3}\cos\theta + \alpha_2 r^{-5}\cos\theta + \alpha_3 r^{-1}\cos 3\theta\Big]E_0^3. \tag{A-20}$$

From (A-8), the coefficients α_1, α_2 and α_3 can be calculated as follows:

$$\begin{aligned}\alpha_1 &= \frac{8b^2}{(-5+2)(-5+1)+(-5+2)-1^2}\\ &= b^2, \end{aligned}\tag{A-21}$$

$$\begin{aligned}\alpha_2 &= \frac{-4b^3}{(-7+2)(-7+1)+(-7+2)-1^2}\\ &= -\frac{1}{6}\,b^3, \end{aligned}\tag{A-22}$$

$$\begin{aligned}\alpha_3 &= \frac{-4b}{(-3+2)(-3+1)+(-3+2)-3^2}\\ &= \frac{1}{2}\,b. \end{aligned}\tag{A-23}$$

Substituting (A-21)-(A-23) into (A-20), we obtain the particular solution of $\phi_1^m(r,\theta)$ as

$$\phi_{1p}^m(r,\theta) = -\frac{1}{\varepsilon_m}\Big[(b^2r^{-3} - \frac{1}{6}\,b^3r^{-5})\cos\theta + \frac{1}{2}\,br^{-1}\cos 3\theta\Big]E_0^3. \tag{A-24}$$

The particular solution for the second-order electric potential

We consider the second-order electric potential equation for the host medium region from Eq. (3.59) which is

$$\begin{aligned}\nabla^2\phi_2^m &= -\frac{1}{\varepsilon_m}\Big[\Big(\frac{16bb_1}{r^5}-\frac{20b^2}{r^5}-\frac{12b^2b_1}{r^7}+\frac{24bb_2}{r^7}+\frac{68b^3}{\varepsilon_m r^7}-\frac{24b^2b_2}{r^9}-\frac{76b^4}{\varepsilon_m r^9}\\&+\frac{56b^5}{3\varepsilon_m r^{11}}\Big)\cos\theta+\Big(-\frac{4b_1}{r^3}+\frac{8b}{\varepsilon_m r^3}+\frac{48bb_2}{r^7}+\frac{32b^3}{\varepsilon_m r^7}-\frac{24b^2b_2}{r^9}\\&-\frac{44b^4}{3\varepsilon_m r^9}+\frac{2b^5}{\varepsilon_m r^{11}}\Big)\cos 3\theta+\Big(-\frac{6b}{\varepsilon_m r^3}-\frac{24b_2}{r^5}-\frac{12b^2}{\varepsilon_m r^5}\Big)\cos 5\theta\Big]E_0^5,\end{aligned} \tag{A-25}$$

where ε_m is in unit of β_m.

The computation of $\phi_{2p}^m(r,\theta)$ is far more tedious because there are many terms in the right hand side of (A-25), but similar to that of $\phi_{1p}^m(r,\theta)$. We omit the details of calculation and give the result of $\phi_2^m(r,\theta)$ as

$$\begin{aligned}\phi_{2p}^m(r,\theta) &= -\frac{1}{\varepsilon_m}\Big[(b_5r^{-3}+b_6r^{-5}+b_7r^{-7}+b_8r^{-9})\cos\theta\\&+(b_9r^{-1}+b_{10}r^{-5}+b_{11}r^{-7}+b_{12}r^{-9})\cos 3\theta\\&+(b_{13}r^{-1}+b_{14}r^{-3})\cos 5\theta\Big]E_0^5,\end{aligned}$$

where

$$\begin{aligned}b_5 &= 2bb_1-\frac{5b^2}{2\varepsilon_m},\\ b_6 &= -\frac{b^2b_1}{2}+bb_2+\frac{17b^3}{6\varepsilon_m},\\ b_7 &= -\frac{b^2b_2}{2}-\frac{19b^4}{12\varepsilon_m},\\ b_8 &= \frac{7b^5}{30\varepsilon_m},\\ b_9 &= \frac{b_1}{2}-\frac{b}{\varepsilon_m},\\ b_{10} &= 3bb_2+\frac{2b^3}{\varepsilon_m},\\ b_{11} &= -\frac{3b^2b_2}{5}-\frac{11b^4}{30\varepsilon_m},\end{aligned}$$

$$
\begin{aligned}
b_{12} &= \frac{b^5}{36\varepsilon_m}, \\
b_{13} &= \frac{b}{4\varepsilon_m}, \\
b_{14} &= \frac{3b_2}{2} + \frac{3b^2}{4\varepsilon_m}.
\end{aligned}
$$

The particular solution for the third-order electric potential

We consider the third-order electric potential equation for the host medium region from Eq. (3.89) which is

$$
\begin{aligned}
\nabla^2\phi_3^m(r,\theta) &= -\frac{1}{\varepsilon_m}\Big[(g_1r^{-5} + g_2r^{-7} + g_3r^{-9} + g_4r^{-11} + g_5r^{-13} + g_6r^{-15})\cos\theta \\
&+ (g_7r^{-3} + g_8r^{-5} + g_9r^{-7} + g_{10}r^{-9} + g_{11}r^{-11} + g_{12}r^{-13} \\
&+ g_{13}r^{-15})\cos 3\theta + (g_{14}r^{-3} + g_{15}r^{-5} + g_{16}r^{-7} + g_{17}r^{-9} \\
&+ g_{18}r^{-11} + g_{19}r^{-13} + g_{20}r^{-15})\cos 5\theta + (g_{21}r^{-3} + g_{22}r^{-5} \\
&+ g_{23}r^{-7})\cos 7\theta\Big]E_0^7, \qquad \text{(A-26)}
\end{aligned}
$$

where ε_m is in unit of β_m and

$$
\begin{aligned}
g_1 &= 8b_1^2 + 16bc_1 + \frac{8b^2}{\varepsilon_m^2} - \frac{20b_5}{\varepsilon_m}, \\
g_2 &= -12bb_1^2 + 24b_1b_2 - 12b^2c_1 + 24bc_2 - \frac{4b^3}{\varepsilon_m^2} + \frac{52b^2b_1}{\varepsilon_m} \\
&+ \frac{60bb_2}{\varepsilon_m} + \frac{64bb_5}{\varepsilon_m} - \frac{60b_6}{\varepsilon_m} - \frac{12b^2b_9}{\varepsilon_m} - \frac{4b_{10}}{\varepsilon_m}, \\
g_3 &= -48bb_1b_2 + 144b_2^2 - 24b^2c_2 + \frac{100b^4}{\varepsilon_m^2} - \frac{128b^3b_1}{\varepsilon_m} + \frac{24b^2b_2}{\varepsilon_m} \\
&- \frac{52b^2b_5}{\varepsilon_m} + \frac{144bb_6}{\varepsilon_m} - \frac{120b_7}{\varepsilon_m} + \frac{64bb_{10}}{\varepsilon_m} - \frac{12b_{11}}{\varepsilon_m}, \\
g_4 &= -144bb_2^2 - \frac{832b^5}{3\varepsilon_m^2} + \frac{112b^4b_1}{3\varepsilon_m} - \frac{192b^3b_2}{\varepsilon_m} - \frac{112b^2b_6}{\varepsilon_m} + \frac{256bb_7}{\varepsilon_m} \\
&- \frac{200b_8}{\varepsilon_m} - \frac{40b^2b_{10}}{\varepsilon_m} + \frac{120bb_{11}}{\varepsilon_m} - \frac{24b_{12}}{\varepsilon_m}, \\
g_5 &= \frac{146b^6}{\varepsilon_m^2} + \frac{56b^4b_2}{\varepsilon_m} - \frac{192b^2b_7}{\varepsilon_m} + \frac{400bb_8}{\varepsilon_m} - \frac{60b^2b_{11}}{\varepsilon_m} + \frac{192bb_{12}}{\varepsilon_m}, \\
g_6 &= -\frac{21b^7}{\varepsilon_m^2} - \frac{292b^2b_8}{\varepsilon_m} - \frac{84b^2b_{12}}{\varepsilon_m},
\end{aligned}
$$

$$
\begin{aligned}
g_7 &= -4c_1 + \frac{16b_9}{\varepsilon_m} - \frac{4b_{13}}{\varepsilon_m}, \\
g_8 &= \frac{12bb_1}{\varepsilon_m} - \frac{12b_5}{\varepsilon_m} + \frac{24bb_9}{\varepsilon_m} - \frac{24bb_{13}}{\varepsilon_m}, \\
g_9 &= 48b_1b_2 + 48bc_2 + \frac{20b^3}{\varepsilon_m^2} + \frac{32b^2b_1}{\varepsilon_m} + \frac{24bb_5}{\varepsilon_m} \\
&\quad - \frac{24b_6}{\varepsilon_m} + \frac{8b^2b_9}{\varepsilon_m} - \frac{32b_{10}}{\varepsilon_m} - \frac{24b^2b_{13}}{\varepsilon_m}, \\
g_{10} &= -48bb_1b_2 - 24b^2c_2 + 40bc_3 + \frac{124b^4}{3\varepsilon_m^2} - \frac{56b^3b_1}{3\varepsilon_m} + \frac{168b^2b_2}{\varepsilon_m} \\
&\quad - \frac{4b^2b_5}{\varepsilon_m} + \frac{64bb_6}{\varepsilon_m} - \frac{40b_7}{\varepsilon_m} + \frac{88bb_{10}}{\varepsilon_m} - \frac{80b_{11}}{\varepsilon_m} - \frac{40b^2b_{14}}{\varepsilon_m}, \\
g_{11} &= -60b^2c_3 - \frac{70b^5}{\varepsilon_m^2} + \frac{4b^4b_1}{\varepsilon_m} - \frac{276b^3b_2}{\varepsilon_m} - \frac{12b^2b_6}{\varepsilon_m} + \frac{120bb_7}{\varepsilon_m} \\
&\quad - \frac{60b_8}{\varepsilon_m} - \frac{72b^2b_{10}}{\varepsilon_m} + \frac{144bb_{11}}{\varepsilon_m} - \frac{144b_{12}}{\varepsilon_m}, \\
g_{12} &= \frac{104b^6}{3\varepsilon_m^2} + \frac{76b^4b_2}{\varepsilon_m} - \frac{24b^2b_7}{\varepsilon_m} + \frac{192bb_8}{\varepsilon_m} - \frac{136b^2b_{11}}{\varepsilon_m} + \frac{216bb_{12}}{\varepsilon_m}, \\
g_{13} &= -\frac{46b^7}{9\varepsilon_m^2} - \frac{40b^2b_8}{\varepsilon_m} - \frac{216b^2b_{12}}{\varepsilon_m}, \\
g_{14} &= -\frac{12b_9}{\varepsilon_m} + \frac{48b_{13}}{\varepsilon_m}, \\
g_{15} &= -24c_2 + \frac{4b^2}{\varepsilon_m^2} - \frac{12bb_1}{\varepsilon_m} - \frac{24bb_9}{\varepsilon_m} + \frac{56bb_{13}}{\varepsilon_m} + \frac{32b_{14}}{\varepsilon_m}, \\
g_{16} &= \frac{10b^3}{\varepsilon_m^2} - \frac{40b_{10}}{\varepsilon_m} + \frac{40b^2b_{13}}{\varepsilon_m} + \frac{80bb_{14}}{\varepsilon_m}, \\
g_{17} &= 120bc_3 + \frac{8b^4}{\varepsilon_m^2} + \frac{120b^2b_2}{\varepsilon_m} + \frac{40bb_{10}}{\varepsilon_m} - \frac{60b_{11}}{\varepsilon_m} + \frac{8b^2b_{14}}{\varepsilon_m}, \\
g_{18} &= -36bb_2^2 - 40b^2c_3 - \frac{4b^5}{\varepsilon_m^2} - \frac{64b^3b_2}{\varepsilon_m} - \frac{4b^2b_{10}}{\varepsilon_m} \\
&\quad + \frac{96bb_{11}}{\varepsilon_m} - \frac{84b_{12}}{\varepsilon_m}, \\
g_{19} &= \frac{12b^4b_2}{\varepsilon_m} - \frac{12b^2b_{11}}{\varepsilon_m} + \frac{168bb_{12}}{\varepsilon_m}, \\
g_{20} &= -\frac{24b^2b_{12}}{\varepsilon_m}, \\
g_{21} &= -\frac{24b_{13}}{\varepsilon_m}, \\
g_{22} &= -\frac{14b^2}{\varepsilon_m^2} - \frac{64bb_{13}}{\varepsilon_m} - \frac{40b_{14}}{\varepsilon_m}, \\
g_{23} &= -60c_3 - \frac{5b^3}{\varepsilon_m^2} - \frac{48bb_2}{\varepsilon_m} - \frac{4b^2b_{13}}{\varepsilon_m} - \frac{40bb_{14}}{\varepsilon_m}.
\end{aligned}
$$

From (A-26), the terms $g_8 r^{-5}\cos 3\theta$ and $g_{16} r^{-7}\cos 5\theta$ have $k+2=-j$. Then the particular solution of these parts have to be solved by using (A-14). The computation of $\phi^m_{3p}(r,\theta)$ is far more tedious because there are many terms in the right hand side of (A-26), but similar to that of $\phi^m_{1p}(r,\theta)$ and $\phi^m_{2p}(r,\theta)$. We omit the details of calculation and give the results of $\phi^m_{3p}(r,\theta)$ as

$$\begin{aligned}\phi^m_{3p}(r,\theta) &= -\frac{1}{\varepsilon_m}\Big[(b_{15}r^{-3}+b_{16}r^{-5}+b_{17}r^{-7}+b_{18}r^{-9}+b_{19}r^{-11}\\ &\quad +b_{20}r^{-13})\cos\theta+(b_{21}r^{-1}+b_{22}r^{-3}\ln r+b_{23}r^{-5}+b_{24}r^{-7}\\ &\quad +b_{25}r^{-9}+b_{26}r^{-11}+b_{27}r^{-13})\cos 3\theta+(b_{28}r^{-1}+b_{29}r^{-3}\\ &\quad +b_{30}r^{-5}\ln r+b_{31}r^{-7}+b_{32}r^{-9}+b_{33}r^{-11}+b_{34}r^{-13})\cos 5\theta\\ &\quad +(b_{35}r^{-1}+b_{36}r^{-3}+b_{37}r^{-5})\cos 7\theta\Big]E_0^7, \end{aligned} \tag{A-27}$$

where $b_{15}=\frac{1}{8}\,g_1$, $b_{16}=\frac{1}{24}\,g_2$, $b_{17}=\frac{1}{48}\,g_3$, $b_{18}=\frac{1}{80}\,g_4$, $b_{19}=\frac{1}{120}\,g_5$, $b_{20}=\frac{1}{168}\,g_6$, $b_{21}=-\frac{1}{8}\,g_7$, $b_{22}=-\frac{1}{6}\,g_8$, $b_{23}=\frac{1}{16}\,g_9$, $b_{24}=\frac{1}{40}\,g_{10}$, $b_{25}=\frac{1}{72}\,g_{11}$, $b_{26}=\frac{1}{112}\,g_{12}$, $b_{27}=\frac{1}{160}\,g_{13}$, $b_{28}=-\frac{1}{24}\,g_{14}$, $b_{29}=-\frac{1}{16}\,g_{15}$, $b_{30}=-\frac{1}{10}\,g_{16}$, $b_{31}=\frac{1}{24}\,g_{17}$, $b_{32}=\frac{1}{56}\,g_{18}$, $b_{33}=\frac{1}{96}\,g_{19}$, $b_{34}=\frac{1}{144}\,g_{20}$, $b_{35}=-\frac{1}{48}\,g_{21}$, $b_{36}=-\frac{1}{40}\,g_{22}$ and $b_{37}=-\frac{1}{24}\,g_{23}$.

Appendix B

Proof of the Equivalence of Nonlinear Coefficient Definitions

We let the composite volume be V and a uniform external electric field ($\mathbf{E}_0$) is applied by fixing the electric potential on the composite surface (ϕ_S) at $-\mathbf{E}_0 \cdot \mathbf{x}$ for $\mathbf{x}$ on S. We will first show that the space average electric field inside the medium is equals $\mathbf{E}_0$.

We write the space average of the ith cartesian component of $\mathbf{E}$ as follows:

$$\begin{aligned} \langle E_i \rangle &= \frac{1}{V}\int_V E_i(\mathbf{x})\, d^3x, && \text{(B-1)} \\ &= -\frac{1}{V}\int_V \nabla_i \phi\, d^3x, && \text{(B-2)} \\ &= -\frac{1}{V}\int_V \boldsymbol{\nabla}\cdot(\hat{x}_i \phi)\, d^3x. && \text{(B-3)} \end{aligned}$$

By using the divergence theorem, we obtain

$$\langle E_i \rangle = -\frac{1}{V}\oint_S \hat{x}_i \cdot \hat{n}\phi_S\, d^2x. \qquad \text{(B-4)}$$

By the divergence theorem and $\phi_S = -(\mathbf{E}_0 \cdot \mathbf{x})_S$, we have

$$\begin{aligned} \frac{1}{V}\int_V \boldsymbol{\nabla}\cdot \hat{x}_i(\mathbf{E}_0\cdot\mathbf{x})\, d^3x &= \frac{1}{V}\oint_S (\mathbf{E}_0\cdot\mathbf{x})_S \hat{x}_i \cdot \hat{n}\, d^2x, \\ &= -\frac{1}{V}\oint_S \hat{x}_i \cdot \hat{n}\phi_S\, d^2x. && \text{(B-5)} \end{aligned}$$

The right hand side of (B-4) and (B-5) are the same, therefore

$$\begin{aligned} \langle E_i \rangle &= \frac{1}{V}\int_V \boldsymbol{\nabla}\cdot \hat{x}_i(\mathbf{E}_0\cdot\mathbf{x})\, d^3x, \\ &= \frac{1}{V}\int_V E_{0i}\, d^3x, \\ &= E_{0i}, && \text{(B-6)} \end{aligned}$$

and $\langle \mathbf{E} \rangle = \mathbf{E}_0$.

In these equations, $\hat{x}_i$ is a unit vector in the ith direction, $\hat{n}$ is a unit normal to the composite boundary surface (S) and E_{0i} is the ith component of $\mathbf{E}_0$.

One definition of effective coefficients is to relate the electrostatic energy of the composite to that of the homogeneous medium with effective coefficients by the equation

$$W = \int_V \mathbf{D} \cdot \mathbf{E} \, d^3x = V\big[\varepsilon_e E_0^2 + \chi_e E_0^4 + \eta_e E_0^6 + \delta_e E_0^8 + \mu_e E_0^{10}\big]. \tag{B-7}$$

To relate this form to other definition, Eq. (4.2), we write W as

$$\begin{aligned} W &= \int_V \mathbf{D} \cdot \mathbf{E} \, d^3x, & \text{(B-8)} \\ &= -\int_V \mathbf{D} \cdot \boldsymbol{\nabla}\phi \, d^3x, & \text{(B-9)} \\ &= -\int_V \Big[\boldsymbol{\nabla} \cdot (\mathbf{D}\phi) - \phi \boldsymbol{\nabla} \cdot \mathbf{D}\Big] \, d^3x. & \text{(B-10)} \end{aligned}$$

For $\boldsymbol{\nabla} \cdot \mathbf{D} = 0$, we get

$$\begin{aligned} W &= -\int_V \boldsymbol{\nabla} \cdot (\mathbf{D}\phi) \, d^3x, & \text{(B-11)} \\ &= -\oint_S \hat{n} \cdot \mathbf{D}\phi_S \, d^2x. & \text{(B-12)} \end{aligned}$$

By using the divergence theorem and replacing ϕ_S by $-(\mathbf{E}_0 \cdot \mathbf{x})_S$, we get

$$\begin{aligned} W &= \oint_S \hat{n} \cdot \mathbf{D}(\mathbf{E}_0 \cdot \mathbf{x})_S \, d^2x, \\ &= \int_V \boldsymbol{\nabla} \cdot \mathbf{D}(\mathbf{E}_0 \cdot \mathbf{x}) \, d^3x, \\ &= \int_V \Big[\boldsymbol{\nabla} \cdot \mathbf{D}(\mathbf{E}_0 \cdot \mathbf{x}) + \mathbf{D} \cdot \boldsymbol{\nabla}(\mathbf{E}_0 \cdot \mathbf{x})\Big] \, d^3x. & \text{(B-13)} \end{aligned}$$

For $\boldsymbol{\nabla} \cdot \mathbf{D} = 0$ which is our case, that has no free charge, and $\mathbf{D} \cdot \boldsymbol{\nabla}(\mathbf{E}_0 \cdot \mathbf{x}) = \sum_i D_i \frac{\partial}{\partial x_i}(\mathbf{E}_0 \cdot \mathbf{x}) = \mathbf{D} \cdot \mathbf{E}_0$, we obtain

$$\begin{aligned} W &= \int_V \mathbf{D} \, d^3x \cdot \mathbf{E}_0, & \text{(B-14)} \\ &= V\langle \mathbf{D} \rangle \cdot \mathbf{E}_0. & \text{(B-15)} \end{aligned}$$

Equating (B-7) and (B-15), we obtain the coefficients of $\mathbf{E}_0$

$$\langle \mathbf{D} \rangle = \varepsilon_e \mathbf{E}_0 + \chi_e {E_0}^2 \mathbf{E}_0 + \eta_e {E_0}^4 \mathbf{E}_0 + \delta_e {E_0}^6 \mathbf{E}_0 + \mu_e {E_0}^8 \mathbf{E}_0, \qquad \text{(B-16)}$$

which is the other equivalent defining equation, Eq. (4.2), for effective coefficients.

Appendix C

Proof of $\int_V \varepsilon \nabla \phi_0 \cdot \nabla \phi_n d^3x = 0$

Consider

$$\int_V \varepsilon \nabla \phi_0 \cdot \nabla \phi_n \, d^3x = -\int_V \mathbf{D}_0 \cdot \nabla \phi_n \, d^3x, \tag{C-1}$$

$$= -\int_V \Big[\nabla \cdot (\phi_n \mathbf{D}_0) - \phi_n (\nabla \cdot \mathbf{D}_0)\Big] \, d^3x. \tag{C-2}$$

For $\nabla \cdot \mathbf{D}_0 = 0$, we get

$$\int_V \varepsilon \nabla \phi_0 \cdot \nabla \phi_n \, d^3x = -\int_V \nabla \cdot (\phi_n \mathbf{D}_0) \, d^3x, \tag{C-3}$$

$$= -\oint_S \phi_{nS} \mathbf{D}_{0S} \cdot \hat{n} \, d^2x. \tag{C-4}$$

Since $\phi_{0S} = -\mathbf{E}_0 \cdot \mathbf{x}$ for $\mathbf{x}$ on S and $\phi_{nS} = 0$ for $n = 1,\ 2,\ 3, \ldots$, then the right hand side of (C-4) is zero for $n \neq 0$, and

$$\int_V \varepsilon \nabla \phi_0 \cdot \nabla \phi_n \, d^3x = 0. \tag{C-5}$$

Vitae

Mr. Chaivej Thongboonrithi was born on January 22, 1981 in Nakornprathom province, Thailand. He received his B.Sc. in Physics from Chulalongkorn University in 2004. Since June 2004, he has enrolled in the M.Sc. program in physics at Chulalongkorn University. He served as a teaching assistant in the fundamental physics laboratory courses at Chulalongkorn University during the academic years 2005–2007. He also used to work as a scientific translator for a private company during the year 2007.

Publications

1. M. Natenapit, A. Chitranondh and C. Thongboonrithi, "Third-order perturbation for weakly nonlinear dielectric composites of spherical inclusion," J. Sci. Res. Chula. Univ. **31** (2006): 105.

2. C. Thongboonrithi and M. Natenapit, "Ninth-order effective nonlinear coefficients of weakly nonlinear dielectric composites," (2007) (submitted to the Thai Journal of Physics).

Conference presentations

1. C. Thongboonrithi and M. Natenapit, "Seventh-order effective nonlinear coefficient of weakly nonlinear conducting composites," THE SECOND MATHEMATICS AND PHYSICAL SCIENCES GRADUATE CONGRESS, National University of Singapore, Singapore (12-14 December 2006).

2. C. Thongboonrithi and M. Natenapit, "Ninth-order effective nonlinear coefficients of weakly nonlinear dielectric composites," SIAM PHYSICS CONGRESS 2008, Khao Yai, Nakhon Ratchasima, Thailand (20-22 March 2008).