

Nonlinear susceptibilities of a granular composite medium

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1995 J. Phys.: Condens. Matter 7 6335

(<http://iopscience.iop.org/0953-8984/7/31/016>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.151

The article was downloaded on 12/05/2010 at 21:52

Please note that [terms and conditions apply](#).

Nonlinear susceptibilities of a granular composite medium

Jia-jun Wang† and Zhen-ya Li‡

† Department of Physics, Suzhou University, Suzhou 215006, People's Republic of China

‡ China Center of Advanced Science and Technology (World Laboratory), PO Box 8730, Beijing 100080, People's Republic of China

Received 10 August 1994, in final form 12 April 1995

Abstract. An effective-medium theory is developed for a composite medium consisting of nonlinear coated spherical grains embedded in a linear host. By means of a field-dependent T -matrix approach, exact expressions for the effective permittivity and nonlinear susceptibilities of the composite medium with coated spherical grains is obtained.

1. Introduction

The physical properties of a granular composite medium composed of grains embedded in a host medium have attracted much interest [1]. For the linear case, many methods such as the Maxwell-Garnett approximation, the effective-medium approximation and the coherent-potential approximation [2–5] have been developed for obtaining the effective macroscopic parameters of this inhomogeneous system. Recently, much interest has shifted to the nonlinear case. Many workers have devoted themselves to finding approximate methods for solving the nonlinear response of such a composite system [6–15]. The T -matrix approach [13–15] is one of those methods which was originally used for the linear case and then extended to the nonlinear case. Not only can it reproduce the results from other methods in [2–5], but also it is convenient to calculate the nonlinear response of the composite.

Agarwal and Dutta Gupta [14] calculate the third- and fifth-order nonlinear susceptibilities of a composite by use of the T -matrix formulations. Based on the T -matrix approach, Kothari [15] developed an effective-medium theory for nonlinear spherical particles embedded in a linear host. By introduction of a field-dependent T -matrix, an exact expression for the effective permittivity (dielectric function) can be obtained. For a small volume fraction of the grains, the effective linear permittivity and the contributions of all the higher-order nonlinear susceptibilities are presented.

In the present work, we shall generalize the T -matrix approach to the case of coated nonlinear spherical grains embedded in a linear host and present explicit expressions for the effective permittivity of this case. The coating shell takes into account the fact that owing to diffusion, etc, the interfaces between the nonlinear grains and the linear host are not sharp and there must exist interfacial shell layers, the dielectric properties of which are different from those of the grains and the host. We take, for simplicity, the coating shell to be a linear concentric shell, but the method used here can easily be generalized to a more complicated case.

2. Formalism

We consider a nonlinear composite system consisting of nonlinear grains with linear coating shells randomly distributed in a linear host. The radii of the concentric core (grain) and the coated sphere are r_0 and R , respectively. The ratio of r_0 to R is an important parameter, the third power of which is expressed by $\lambda (= (r_0/R)^3)$, indicating the volume fraction of the core in the whole coated grain. The permittivity of the linear shell and host are assumed to be ϵ_s and ϵ_0 , respectively, while the nonlinear core has the following permittivity:

$$\epsilon_c = \epsilon_1 + \frac{\alpha |\mathbf{E}_c|^2}{1 + \beta |\mathbf{E}_c|^2} \quad (1)$$

where the first term ϵ_1 gives the linear part and the second term gives the nonlinear contribution to ϵ_c with α and β constant. The field \mathbf{E}_c is the local field inside the nonlinear core. For a spherical grain with a concentric shell embedded in a linear dielectric host subjected to an applied field \mathbf{E}_0 along the z axis, in the case of dilute limit, the outermost radius is also far smaller than the average distance between coated grains, so that the higher-order multipole scattering, which is of order $(R/L)^3$ (where L denotes the average distance between grains), can be neglected; each coated spherical grain is excited by a uniform field; the field in the core and the shell can be easily obtained by solving the quasi-electrostatic Maxwell equations regardless of the interaction between grains and can be expressed as [16]

$$\mathbf{E}_c = A \mathbf{e}_z \quad (2)$$

$$\mathbf{E}_s = B \mathbf{e}_z + \frac{C}{r^3} \mathbf{e}_z - \frac{3C_z}{r^4} \mathbf{e}_r \quad (3)$$

where \mathbf{e}_z and \mathbf{e}_r are the unit vectors along the z axis and the vector radius, respectively, while

$$A = \frac{9\epsilon_0\epsilon_s}{D} E_0 \quad (4)$$

$$B = \frac{3\epsilon_0(2\epsilon_s + \epsilon_c)}{D} E_0 \quad (5)$$

$$C = \frac{3\epsilon_0(\epsilon_s - \epsilon_c)r_0^3}{D} E_0 \quad (6)$$

with

$$D = (2\epsilon_0 + \epsilon_s)(\epsilon_c + 2\epsilon_s) + 2\lambda(\epsilon_s - \epsilon_0)(\epsilon_c - \epsilon_s) \quad (7)$$

$$\lambda = (r_0/R)^3 \quad (8)$$

Using equation (1), equation (2) can be rewritten as

$$\mathbf{E}_c = K_1 \mathbf{E}_0 - \frac{K_2 |\mathbf{E}_c|^2 \mathbf{E}_c}{1 + \beta |\mathbf{E}_c|^2} \quad (9)$$

with

$$K_1 = \frac{9\epsilon_0\epsilon_s}{D_0} \tag{10}$$

$$K_2 = \frac{[2\epsilon_0 + \epsilon_s + 2\lambda(\epsilon_s - \epsilon_0)]\alpha}{D_0} \tag{11}$$

$$D_0 = (2\epsilon_0 + \epsilon_s)(\epsilon_1 + 2\epsilon_s) + 2\lambda(\epsilon_s - \epsilon_0)(\epsilon_1 - \epsilon_s). \tag{12}$$

Equation (9) has the solution given by

$$\mathbf{E}_c = K_1 S \mathbf{E}_0 \tag{13}$$

where S is a function of the applied field \mathbf{E}_0 and satisfies the following equation:

$$S = 1 - \frac{K_2 |\mathbf{E}_0|^2 |K_1|^2 |S|^2}{1 + (\beta + K_2) |\mathbf{E}_0|^2 |K_1|^2 |S|^2}. \tag{14}$$

From equations (2) and (3), we can easily obtain the volume-averaged electric field in the coated grains:

$$\begin{aligned} \langle \mathbf{E} \rangle = & \left(\lambda + \frac{1}{3\epsilon_s} (\epsilon_1 + 2\epsilon_s)(1 - \lambda) \right) K_1 \mathbf{E}_0 \\ & + \left[\frac{1 - \lambda}{3\epsilon_s} - \left(\lambda + \frac{1 - \lambda}{3} \frac{\epsilon_1 + 2\epsilon_s}{\epsilon_s} \right) \frac{K_2}{\alpha} \right] \frac{\alpha K_1 |K_1|^2 |S|^2 |\mathbf{E}_0|^2 \mathbf{E}_0}{1 + (\beta + K_2) |\mathbf{E}_0|^2 |K_1|^2 |S|^2}. \end{aligned} \tag{15}$$

At the same time, the spatially averaged displacement vector $\langle \mathbf{D} \rangle$ can also be evaluated from equations (2) and (3) together with the following relation:

$$\mathbf{D}_i = \epsilon_i \mathbf{E}_i \quad (i = c, s) \tag{16}$$

where \mathbf{D}_c and \mathbf{D}_s are the displacement vectors inside the core and the shell of the coated sphere, respectively; we have

$$\begin{aligned} \langle \mathbf{D} \rangle = & \left(\lambda\epsilon_1 + \frac{(\epsilon_1 + 2\epsilon_s)(1 - \lambda)}{3} \right) K_1 \mathbf{E}_0 \\ & + \left[\frac{1 + 2\lambda}{3} - \left(\lambda\epsilon_1 + \frac{(1 - \lambda)(\epsilon_1 + 2\epsilon_s)}{3} \right) \frac{K_2}{\alpha} \right] \frac{\alpha K_1 |K_1|^2 |S|^2 |\mathbf{E}_0|^2 \mathbf{E}_0}{1 + (\beta + K_2) |\mathbf{E}_0|^2 |K_1|^2 |S|^2}. \end{aligned} \tag{17}$$

On the other hand, using the T -matrix approach [15], the spatial average of the local electric field \mathbf{E} and the displacement vector \mathbf{D} can be expressed as follows;

$$\langle \mathbf{E} \rangle = (1 + \langle GT_L \rangle) \mathbf{E}_0 + \langle GT_{NL} \rangle |\mathbf{E}_0|^2 \mathbf{E}_0 \tag{18}$$

$$\langle \mathbf{D} \rangle = [\epsilon_0(1 + \langle GT_L \rangle) + \langle T_L \rangle] \mathbf{E}_0 + (\epsilon_0 \langle GT_{NL} \rangle + \langle T_{NL} \rangle) |\mathbf{E}_0|^2 \mathbf{E}_0 \tag{19}$$

where T_L and T_{NL} are the linear and nonlinear T -matrices, while G is an integral operator [1]:

$$GF = \int dr' \mathbf{G}(\mathbf{r}, \mathbf{r}') F(\mathbf{r}') \tag{20}$$

with \mathbf{G} , the Green function, satisfying the equation

$$\nabla \cdot \epsilon_0 \mathbf{G} = \delta(\mathbf{r} - \mathbf{r}') \mathbf{I} \quad (21)$$

and \mathbf{I} is the unit tensor.

Comparing equations (15) and (17) with equations (18) and (19), we can obtain the following relations:

$$\mathbf{I} + \langle GT_L \rangle = \left(\lambda + \frac{(1-\lambda)(\epsilon_1 + 2\epsilon_s)}{3\epsilon_s} \right) K_1 \quad (22)$$

$$\langle GT_{NL} \rangle = \left[\frac{1-\lambda}{3\epsilon_s} - \left(\lambda + \frac{1-\lambda}{3} \frac{\epsilon_1 + 2\epsilon_s}{\epsilon_s} \right) \frac{K_2}{\alpha} \right] \frac{\alpha K_1 |K_1|^2 |S|^2}{1 + (\beta + K_2) |\mathbf{E}_0|^2 |K_1|^2 |S|^2} \quad (23)$$

$$\langle T_L \rangle = \lambda(\epsilon_1 - \epsilon_0) K_1 + \frac{(\epsilon_1 + 2\epsilon_s)(1-\lambda)}{3} K_1 \left(1 - \frac{\epsilon_0}{\epsilon_s} \right) \quad (24)$$

$$\begin{aligned} \langle T_{NL} \rangle = & \left[\lambda + \frac{K_2 \lambda}{\alpha} (\epsilon_0 - \epsilon_1) + \frac{1-\lambda}{3} \left(1 - \frac{\epsilon_0}{\epsilon_s} \right) - \frac{(1-\lambda)(\epsilon_1 + 2\epsilon_s)}{3} \frac{K_2}{\alpha} \left(1 - \frac{\epsilon_0}{\epsilon_s} \right) \right] \\ & \times \frac{\alpha K_1 |K_1|^2 |S|^2}{1 + (\beta + K_2) |\mathbf{E}_0|^2 |K_1|^2 |S|^2}. \end{aligned} \quad (25)$$

The results given in equations (22)–(25) hold well for a single coated sphere inside the host medium. They can be generalized to the case of a colloidal medium provided that such features as size dispersion, and correlation effects arising from multipole scattering can be neglected. For a small volume fraction f of the nonlinear cores in the composite, the T -matrix can be written as the sum of the T -matrices of the individual particles. Quantities such as $\langle GT_L \rangle$, $\langle GT_{NL} \rangle$, $\langle T_L \rangle$ and $\langle T_{NL} \rangle$ corresponding to the colloidal case can be obtained by multiplying those of a single coated particle by f/λ .

Now, if we introduce the effective permittivity ϵ_{eff} as given by

$$\epsilon_{eff} = \bar{\epsilon} + \frac{C_1 \langle |\mathbf{E}|^2 \rangle}{1 + C_2 \langle |\mathbf{E}|^2 \rangle} \quad (26)$$

we can obtain the following results [15]:

$$\bar{\epsilon} = \epsilon_0 + \frac{\langle T_L \rangle}{1 + \langle GT_L \rangle} \quad (27)$$

$$\frac{C_1 P_0}{1 + C_2 Q_0 |\mathbf{E}_0|^2} = \langle T_{NL} \rangle - (\bar{\epsilon} - \epsilon_0) \langle GT_{NL} \rangle \quad (28)$$

with

$$\begin{aligned} P_0 = & (1 + \langle GT_L \rangle) |1 + \langle GT_L \rangle|^2 + \langle GT_{NL} \rangle^* (1 + \langle GT_L \rangle)^2 |\mathbf{E}_0|^2 + 2 \langle GT_{NL} \rangle |1 + \langle GT_L \rangle|^2 |\mathbf{E}_0|^2 \\ & + 2(1 + \langle GT_L \rangle) |\langle GT_{NL} \rangle|^2 |\mathbf{E}_0|^4 + \langle \langle GT_{NL} \rangle \rangle^2 (1 + \langle GT_L \rangle)^* |\mathbf{E}_0|^4 \\ & + \langle GT_{NL} \rangle |\langle GT_{NL} \rangle|^2 |\mathbf{E}_0|^6 \end{aligned} \quad (29)$$

$$Q_0 = |1 + \langle GT_L \rangle|^2 + \langle GT_{NL} \rangle^* (1 + \langle GT_L \rangle) |\mathbf{E}_0|^2 + \langle GT_{NL} \rangle (1 + \langle GT_L \rangle)^* |\mathbf{E}_0|^2 + |\langle GT_{NL} \rangle|^2 |\mathbf{E}_0|^4 \quad (30)$$

where $\bar{\epsilon}$ is the effective linear permittivity and ϵ_0 is the permittivity of the linear host.

We apply the above results to find the effective-medium parameters for the composite medium with a small volume fraction f of nonlinear grains (which are surrounded by a linear shell) embedded in it. Using equations (22) and (24) with equation (27), we have the linear part of the effective permittivity:

$$\bar{\epsilon} = \epsilon_0 + \frac{f K_1 (\epsilon_1 - \epsilon_0) + (f/3\lambda)(1 - \lambda)(\epsilon_2 + 2\epsilon_s)(1 - \epsilon_0/\epsilon_s) K_1}{1 + f(K_1 - 1/\lambda) + (f/3\lambda)(1 - \lambda)K_1(\epsilon_1 + 2\epsilon_s)/\epsilon_s}. \tag{31}$$

In the meanwhile, if we insert equations (23) and (25) multiplied by f/λ into equation (28), we obtain

$$\begin{aligned} \frac{C_1 P_0}{1 + C_2 Q_0 |E_0|^2} &= \frac{f}{\lambda} \left[\lambda + \frac{K_2 \lambda}{\alpha} (\bar{\epsilon} - \epsilon_1) + \frac{1 - \lambda}{3} \left(1 - \frac{\bar{\epsilon}}{\epsilon_s} \right) \left(1 - \frac{K_2}{\alpha} (\epsilon_1 + 2\epsilon_s) \right) \right] \\ &\times \frac{\alpha K_1 |K_1|^2 |S|^2}{1 + (\beta + K_2) |E_0|^2 |K_1|^2 |S|^2}. \end{aligned} \tag{32}$$

Using equations (22)–(25), we can rewrite P_0 and Q_0 as

$$\begin{aligned} P_0 &= \left[1 + f \left(K_1 - \frac{1}{\lambda} \right) + \frac{f(1 - \lambda) \epsilon_1 + 2\epsilon_s}{3\lambda \epsilon_s} K_1 \right] \\ &\times \left| 1 + f \left(K_1 - \frac{1}{\lambda} \right) + \frac{f(1 - \lambda) \epsilon_1 + 2\epsilon_s}{3\lambda \epsilon_s} K_1 \right|^2 \\ &\times \left| 1 + \frac{f}{\lambda N} \left(\frac{\alpha(1 - \lambda)}{3K_2 \epsilon_s} - \lambda - \frac{1 - \lambda}{3} \frac{\epsilon_1 + 2\epsilon_s}{\epsilon_s} \right) (1 - S) \right|^2 \\ &\times \left[1 + \frac{f}{\lambda N} \left(\frac{\alpha(1 - \lambda)}{3K_2 \epsilon_s} - \lambda - \frac{1 - \lambda}{3} \frac{\epsilon_1 + 2\epsilon_s}{\epsilon_s} \right) (1 - S) \right] \end{aligned} \tag{33}$$

$$\begin{aligned} Q_0 &= \left| 1 + f \left(K_1 - \frac{1}{\lambda} \right) + \frac{f(1 - \lambda) \epsilon_1 + 2\epsilon_s}{3\lambda \epsilon_s} K_1 \right|^2 \\ &\times \left| 1 + \frac{f}{\lambda N} \left(\frac{\alpha(1 - \lambda)}{3K_2 \epsilon_s} - \lambda - \frac{1 - \lambda}{3} \frac{\epsilon_1 + 2\epsilon_s}{\epsilon_s} \right) (1 - S) \right|^2 \end{aligned} \tag{34}$$

where

$$N = \frac{1}{K_1} \left[1 + f \left(K_1 - \frac{1}{\lambda} \right) + \frac{f(1 - \lambda) \epsilon_1 + 2\epsilon_s}{3\lambda \epsilon_s} K_1 \right]. \tag{35}$$

We make the following choices for the unknown parameters C_1 and C_2 :

$$\begin{aligned} C_1 &= \frac{f}{\lambda} \left[\lambda + \frac{\lambda K_2}{\alpha} (\bar{\epsilon} - \epsilon_1) + \frac{1 - \lambda}{3} \left(1 - \frac{\bar{\epsilon}}{\epsilon_s} \right) \left(1 - \frac{K_2}{\alpha} (\epsilon_1 + 2\epsilon_s) \right) \right] \\ &\times \alpha K_1 |K_1|^2 \left[1 + f \left(K_1 - \frac{1}{\lambda} \right) + \frac{f(1 - \lambda)(\epsilon_1 + 2\epsilon_s)}{3\lambda \epsilon_s} K_1 \right]^{-1} \\ &\times \left| 1 + f \left(K_1 - \frac{1}{\lambda} \right) + \frac{f(1 - \lambda)(\epsilon_1 + 2\epsilon_s) K_1}{3\lambda \epsilon_s} \right|^{-2} \end{aligned} \tag{36}$$

$$\begin{aligned}
 1 + C_2 Q_0 \frac{|\mathbf{E}_c|^2}{|K_1|^2 |S|^2} &= \frac{1}{|S|^2} [1 + (K_2 + \beta) |\mathbf{E}_c|^2] \\
 &\times \left| 1 + \frac{f}{\lambda N} \left(\frac{\alpha(1-\lambda)}{3K_2\epsilon_s} - \lambda - \frac{1-\lambda}{3} \frac{\epsilon_1 + 2\epsilon_s}{\epsilon_s} \right) (1-S) \right|^2 \\
 &\times \left[1 + \frac{f}{\lambda N} \left(\frac{\alpha(1-\lambda)}{3K_2\epsilon_s} - \lambda - \frac{(1-\lambda)(\epsilon_1 + 2\epsilon_s)}{3\epsilon_s} \right) (1-S) \right]. \quad (37)
 \end{aligned}$$

It should be mentioned here that the procedure of determination is self-consistent, because the final result of ϵ_{eff} does not depend on any arbitrary choices of C_1 and C_2 .

Using equations (28) and (32) together with equations (10)–(12), equation (36) can be expressed in the more compact form

$$C_1 = \frac{f\alpha}{N^2 |N|^2}. \quad (38)$$

On the other hand, from equation (37) we can obtain

$$C_2 |\mathbf{E}_c|^2 = \frac{1}{|N|^2} \left(1 + K_3 |\mathbf{E}_c|^2 - \left| \frac{1 + \beta |\mathbf{E}_c|^2}{1 + K_3 |\mathbf{E}_c|^2} \right|^2 \right) \quad (39)$$

where

$$K_3 = K_2 + \beta + \frac{K_2 f}{\lambda N} \left(\frac{\alpha(1-\lambda)}{3K_2\epsilon_s} - \lambda - \frac{1-\lambda}{3} \frac{\epsilon_1 + 2\epsilon_s}{\epsilon_s} \right). \quad (40)$$

In a composite medium when a small fraction of coated spherical grains are distributed in the host, the spatially averaged field over all the volume can be found by inserting $\langle GT_L \rangle$ and $\langle GT_{NL} \rangle$ for this case into equation (18), and we have

$$\langle \mathbf{E} \rangle = N \mathbf{E}_c \times \frac{1 + K_3 |\mathbf{E}_c|^2}{1 + \beta |\mathbf{E}_c|^2}. \quad (41)$$

With the help of the above relation, we can rewrite equation (26) as

$$\epsilon_{eff} = \bar{\epsilon} + \frac{C_1 |N|^2 |\mathbf{E}_c|^2}{1 + K_3 |\mathbf{E}_c|^2}. \quad (42)$$

For small-field values, equation (42) can be expanded into a power series of $|\langle \mathbf{E} \rangle|^2$. The result for the simple case $\beta = 0$ is

$$\epsilon_{eff} = \bar{\epsilon} + 4\pi \bar{\chi}^{(3)} |\langle \mathbf{E} \rangle|^2 + 4\pi \bar{\chi}^{(5)} |\langle \mathbf{E} \rangle|^4 + 4\pi \bar{\chi}^{(7)} |\langle \mathbf{E} \rangle|^6 + 4\pi \bar{\chi}^{(9)} |\langle \mathbf{E} \rangle|^8 \quad (43)$$

where

$$4\pi \bar{\chi}^{(3)} = C_1 \quad (44)$$

$$4\pi \bar{\chi}^{(5)} = -C_1 \eta_0 \quad (45)$$

$$4\pi \bar{\chi}^{(7)} = C_1 (\eta_1 + \eta_0^2) \quad (46)$$

$$4\pi \bar{\chi}^{(9)} = -C_1 (\eta_2 + 2\eta_1 \eta_0 + \eta_0^3) \quad (47)$$

with

$$\eta_0 = \frac{1}{|N|^2} (2K_3 + K_3^*) \tag{48}$$

$$\eta_1 = \frac{1}{|N|^4} (K_3^2 + (K_3^*)^2 + |K_3|^2) \tag{49}$$

$$\eta_2 = \frac{1}{|N|^6} [K_3^3 + K_3^2 K_3^* + K_3 (K_3^*)^2 + (K_3^*)^3]. \tag{50}$$

It should be pointed out that all the results here can be reduced to the corresponding results in [15] by letting $\lambda = 1$, e.g. equations (31) and (32) can be re-expressed as

$$\bar{\epsilon} = \epsilon_0 + \frac{fX(\epsilon_1 - \epsilon_0)}{1 + f(X - 1)} \tag{51}$$

$$\epsilon_{eff} = \bar{\epsilon} + \frac{\bar{A}|P|^2|E_L|^2}{1 + a_0|E_L|^2} \tag{52}$$

where

$$X = \frac{3\epsilon_0}{\epsilon_1 + 2\epsilon_0} \tag{53}$$

$$a = \frac{\alpha}{\epsilon_1 + 2\epsilon_0} \tag{54}$$

$$\bar{A} = \frac{f\alpha}{P^2|P|^2} \tag{55}$$

$$P = (1/X)[1 + f(X - 1)] \tag{56}$$

$$a_0 = a + \beta - f\alpha/P. \tag{57}$$

and E_L is the local field within the sphere. Equations (51) and (52) are identical with those of [15].

3. Results and calculations

The third-order nonlinear susceptibility $4\pi\bar{\chi}^{(3)}/\alpha$ corresponding to equation (44) and the local field E_c/E_0 within the nonlinear core as functions of the structural parameter λ are depicted in figure 1 and figure 2, respectively. Other parameters such as ϵ_0 , ϵ_s , ϵ_1 and F ($= f/\lambda$, the volume fraction of the coated grains) are given as shown in their figure captions. From figure 1, we can see that the nonlinear susceptibility exhibits a dependence on the structural parameter λ . For a given F (in the dilute limit), there appears a small peak at a critical value λ_0 , where the system has a relatively strong nonlinear response. In the region $\lambda < \lambda_0$, the nonlinear susceptibility will increase with increase in λ , while in the region $\lambda > \lambda_0$ the nonlinearity will show a slight decrease when the value of λ increases. When λ tends to 1, the thickness of the coating shell tends to zero and the nonlinear response corresponds to the uncoated case in [15]. On the other hand, a decrease in the local field within the nonlinear core together with the increase in λ can be seen from figure 2. This

indicates that the internal local field will increase when the thickness of the coating shell increases. Combining the phenomena shown in figure 1 and figure 2, we can discuss some physical features. It is well known that the macroscopic nonlinear response results from the following two factors: firstly the volume fraction of the nonlinear constituent; secondly the local field in the nonlinear material. From the variance of the two factors together with the structural parameter λ , we can understand the curve shown in figure 1. When λ is near the critical value λ_0 , the two factors show an optimum combination and the system has a relatively strong nonlinearity. In the region where $\lambda < \lambda_0$, the contribution of the first factor to the macroscopic nonlinearity is more important than the second factor. As a result, the nonlinear susceptibility increases with increase in the relative weight of the nonlinear core (i.e. with the increase in λ), although the local field in the nonlinear core decreases at the same time. When the value of λ becomes larger than the critical value λ_0 , the second factor becomes the major one, and the nonlinear response exhibits a slight decrease together with the decrease in the local field despite the fact that the relative weight of the nonlinear core increases.

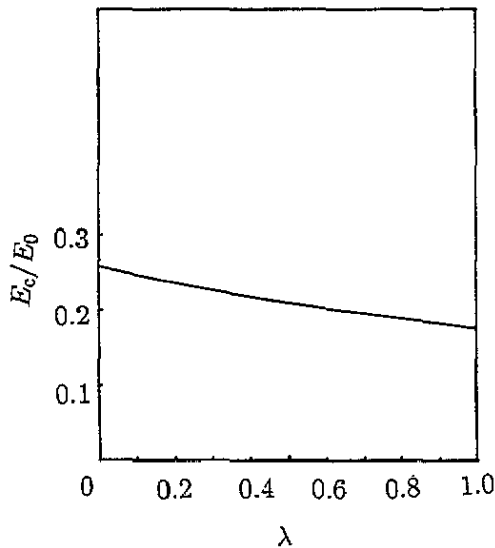


Figure 1. The third-order nonlinear susceptibility $4\pi\bar{\chi}^{(3)}/\alpha$ as a function of the structural parameter λ . The other parameters chosen in the calculation are as follows: $\epsilon_0 = 1.0$, $\epsilon_s = 5.0$, $\epsilon_1 = 15.0$ and $F = 0.05$.

4. Conclusion

So far, we have extended the T -matrix approach to the composite medium consisting of nonlinear coated spherical grains embedded in a linear host. Our results include all the higher-order nonlinear susceptibilities and they are exact in the case of a small volume fraction f . In our calculation, we have introduced a structural parameter λ which represents the thickness of the coating interfacial shell. In terms of the parameter λ , we have chosen one special case as shown in figure 1 and figure 2; we can find the effects of the interfacial shell

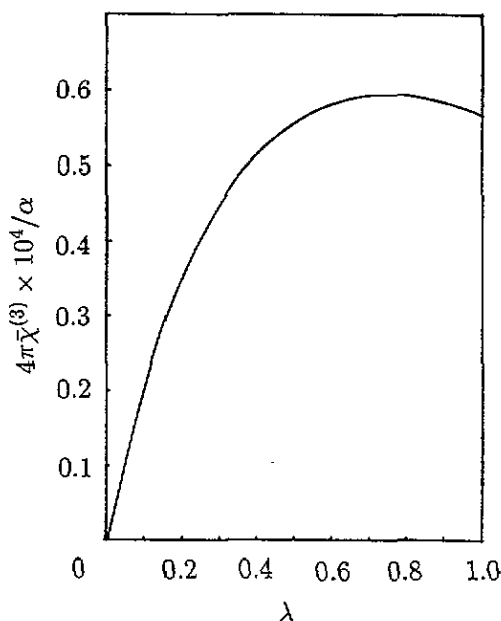


Figure 2. The local field E_c/E_0 within the nonlinear core as a function of the structural parameter λ . The parameters chosen in this calculation are as follows: $\epsilon_0 = 1.0$, $\epsilon_1 = 5.0$, $\epsilon_s = 15.0$ and $F = 0.05$.

around the nonlinear grain on the effective nonlinear response of the composite medium. It is reasonable from the discussions in this paper that we can find a optimum combination of the electrical and geometrical parameters to obtain a strong nonlinear response of a composite medium.

Acknowledgment

This work was supported by the National Natural Science Foundation of China.

References

- [1] Garland J C and Tanner D B (ed) 1978 *Electrical Transport and Optical Properties of Inhomogeneous Media* (AIP Conf. Proc. 40) (Ohio State University, 1977) (New York: American Institute of Physics)
- [2] Landauer R 1952 *J. Appl. Phys.* **23** 779
- [3] Maxwell-Garnett J 1904 *Phil. Trans. R. Soc. A* **203** 384
- [4] Shanker B and Lakhtakia A 1993 *J. Phys. D: Appl. Phys.* **26** 1746; 1993 *J. Compos. Mater.* **27** 1203
- [5] Agarwal G S and Inguva R 1984 *Phys. Rev. B* **30** 6108
- [6] Stroud D and Hui P M 1988 *Phys. Rev. B* **37** 8719
- [7] Stroud D and Wood V E 1989 *J. Opt. Soc. Am. B* **6** 778
- [8] Hui P M 1990 *J. Appl. Phys.* **68** 3009
- [9] Gu G Q and Yu K W 1992 *Phys. Rev. B* **46** 4502
- [10] Yu K W and Hui P M 1993 *Phys. Rev. B* **47** 14 150
- [11] Hui P M 1993 *J. Appl. Phys.* **73** 4072
- [12] Levy O and Bergman D J 1992 *Phys. Rev. B* **46** 7189
- [13] Middy T R, Basu A N and Sengupta S 1986 *J. Math. Phys.* **27** 2807

- [14] Agarwal G S and Dutta Gupta S 1988 *Phys. Rev. A* **38** 5678
- [15] Kothari N C 1990 *Phys. Rev. A* **41** 4486
- [16] Yoshida K 1982 *J. Phys. C: Solid State Phys.* **15** L87