Nonlinear alternating current responses of graded materials

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Abstract

When a composite of nonlinear particles suspended in a host medium is subjected to a sinusoidal electric field, the electrical response in the composite will generally consist of alternating current (AC) fields at frequencies of higher-order harmonics. The situation becomes more interesting when the suspended particles are graded, with a spatial variation in the dielectric properties. The local electric field inside the graded particles can be calculated by the differential effective dipole approximation, which agrees very well with a first-principles approach. In this work, a nonlinear differential effective dipole approximation and a perturbation expansion method have been employed to investigate the effect of gradation on the nonlinear AC responses of these composites. The results showed that the fundamental and third-harmonic AC responses are sensitive to the dielectric-constant and/or nonlinear-susceptibility gradation profiles within the particles. Thus, by measuring the AC responses of the graded composites, it is possible to perform a real-time monitoring of the fabrication process of the gradation profiles within the graded particles.

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I. INTRODUCTION

Graded materials with spatial gradients in their structure [1] are abundant in nature, which have received much attention as one of the advanced inhomogeneous composite materials in diverse engineering applications [2]. These materials can be made to realize quite different physical properties from the homogeneous materials, and thus, to some extent, more useful and interesting. For graded materials, the traditional theories [3] for homogeneous materials do not work any longer. Recently, we presented a first-principles approach [4,5] and a differential effective dipole approximation (DEDA) [6,7], to investigate the dielectric properties of the graded materials. To our interest, the two methods have been demonstrated in excellent agreement between each other [4]. In the case of graded materials, the problem will become more complicated by the presence of nonlinearity inside them. Fortunately, for deriving the equivalent nonlinear susceptibility of graded particles, we have succeeded in putting forth a nonlinear differential effective dipole approximation (NDEDA) [8]. As expected, this NDEDA has also been demonstrated in excellent agreement with a first-principles approach [8].

In addition, the finite-frequency response of nonlinear composite materials have attracted much attention both in research and industrial applications during the last two decades [9]. When a composite with linear/nonlinear particles embedded in a linear/nonlinear host medium is subjected to a sinusoidal electric field, the electrical response in the composite will generally consist of alternating current (AC) fields at frequencies of higher-order harmonics [10–14]. In fact, a convenient method of probing the nonlinear characteristics of the composite is to measure the harmonics of the nonlinear polarization under the application of a sinusoidal electric field [15]. The strength of the nonlinear polarization should be reflected in the magnitude of the harmonics. For the purpose of extracting such harmonics, the perturbation expansion [12–14] and self-consistent methods [13,16] can be used.

In this work, based on the NDEDA, we shall investigate the effect of gradation (inhomogeneity) inside the particles (inclusions) on the AC responses of the graded composite
by making use of a perturbation expansion method [17]. Here, the composite under consideration is composed of linear/nonlinear graded particles which are randomly embedded in a linear/nonlinear host medium in the dilute limit. To this end, it is shown that the fundamental and third-order harmonic AC responses are sensitive to the dielectric-constant (or nonlinear-susceptibility) gradation profile within the particle. Thus, by measuring the AC responses of the graded composites, it is possible to perform a real-time monitoring of the fabrication process of the gradation profiles within graded particles.

This paper is organized as follows. In the following section, we shall present the formalism, which is followed by the numerical results in Section III. In Section IV, the discussion and conclusion will be given.

II. FORMALISM

Let’s consider nonlinear graded spherical particles with radius \( a \) and dielectric gradation profile \( \tilde{\epsilon}_1(r) = \epsilon_1(r) + \chi_1(r)E_1^2 \) inside it, being embedded in a nonlinear host medium of dielectric constant \( \tilde{\epsilon}_2 = \epsilon_2 + \chi_2E_2^2 \), in the presence of a uniform external electric field \( E_0 \) along \( z \)-axis. Here \( \epsilon_1(r) \) or \( \epsilon_2 \) (\( \chi_1(r) \) or \( \chi_2 \)) denotes the corresponding linear dielectric constant (nonlinear susceptibility), \( E_1 \) and \( E_2 \) stands for the local electric field inside the particles and the host medium, respectively. Note both gradation profiles \( \epsilon_1(r) \) and \( \chi_1(r) \) are radial functions where \( r < a \). Throughout the paper, we shall consider the case of weak nonlinearity only, that is, \( \chi_1(r)E_1^2 \ll \epsilon_1(r) \) and \( \chi_2E_2^2 \ll \epsilon_2 \).

A. Comparison between a differential effective dipole approximation and a first-principles approach

Recently, we put forth a DEDA (differential effective dipole approximation) [6,7] for calculating the equivalent dielectric constant \( \tilde{\epsilon}_1(r) \) [8] of the spherical graded particle. This DEDA receives the form
\[
\frac{d\bar{\epsilon}_1(r)}{dr} = \left[ \epsilon_1(r) - \bar{\epsilon}_1(r) \right] \cdot \left[ \bar{\epsilon}_1(r) + 2\epsilon_1(r) \right].
\] (1)

Note Eq. (1) is just the Tartar formula, derived for assemblages of spheres with varying radial and tangential conductivity [1]. So far, the equivalent \( \bar{\epsilon}_1(r = a) \) for the whole graded particle can be calculated, at least numerically, by solving the differential equation [Eq. (1)], as long as \( \epsilon_1(r) \) (dielectric-constant gradation profile) is given. Once \( \bar{\epsilon}_1(r = a) \) is determined, we can readily take one step forward to obtain the volume average of the linear local electric field inside the particles as

\[
\langle E_1^{\text{lin}} \rangle = \frac{3\epsilon_2}{\bar{\epsilon}_1(r = a) + 2\epsilon_2} E_0.
\] (2)

Hence, the DEDA [Eq. (1)] offers a convenient way to obtain the local electric field [Eq. (2)]. It is worth remarking that the DEDA [Eq. (1)] is valid for arbitrary gradation profiles.

To show the correctness of Eq. (2), we shall alternatively present a first-principles approach for calculating the local electric field inside the particle. For this purpose, let’s take the power-law gradation profile \( (\epsilon(r) = A \cdot (r/a)^n) \) as a model. For this profile, the potential within the graded particle can be given by solving the electrostatic equation, \( \nabla \cdot (\epsilon_1(r) \nabla \Phi) = 0 \) [4],

\[
\Phi_1(r) = -\eta_1 E_0 r^n \cos \theta, \quad r < a,
\] (3)

where the coefficient \( \eta_1 \) is determined by performing appropriate boundary conditions, \( \eta_1 = \frac{3a^{1-s} \epsilon_2}{sA + 2\epsilon_2} \), and \( s = [\sqrt{9 + 2n + n^2} - (1 + n)]/2 \). Based on the relation between the linear local electric field and the potential \( \langle E_1^{\text{lin}}(r) = -\nabla \Phi_1(r) \rangle \), we have

\[
E_1^{\text{lin}}(r) = \eta_1 E_0 r^{n-1} \{ (s - 1) \cos \theta \sin \theta \cos \phi \hat{x} + (s - 1) \cos \theta \sin \theta \sin \phi \hat{y} \\
+ [(s - 1) \cos^2 \theta + 1] \hat{z} \},
\] (4)

where \( \hat{x}, \hat{y}, \) and \( \hat{z} \) are the unit vectors along \( x-\), \( y-\), and \( z-\)axes, respectively. So far, it is straightforward to obtain the volume average of the local electric field inside the particles,
\[
\langle \mathbf{E}_1^{(\text{lin})} \rangle = \frac{1}{V} \int_V \mathbf{E}_1^{(\text{lin})}(r) dV, \tag{5}
\]

where \( V \) is the volume of the spherical particles.

In Fig. 1, we shall numerically compare Eq. (2) (local field predicted by the DEDA) with Eq. (5) (local field obtained from the first-principles approach).

**B. Nonlinear polarization and its higher harmonics**

1. **Nonlinear differential effective dipole approximation**

In a recent work [8], we have established an NDEDA (nonlinear differential effective dipole approximation), by deriving a differential equation for the equivalent nonlinear susceptibility \( \tilde{\chi}_1(r) \), namely,

\[
\frac{d\tilde{\chi}_1(r)}{dr} = \tilde{\chi}_1(r) \left[ \frac{4d\tilde{\epsilon}_1(r)/dr}{2\epsilon_2 + \tilde{\epsilon}_1(r)} \right] + \tilde{\chi}_1(r) \cdot \frac{8y - 3}{r} + \frac{3\chi_1(r)}{5r} \cdot \left( \frac{\tilde{\epsilon}_1(r) + 2\epsilon_1(r)}{3\epsilon_1(r)} \right)^4 (5 + 36x^2 + 16x^3 + 24x^4). \tag{6}
\]

where

\[
x = \frac{\tilde{\epsilon}_1(r) - \epsilon_1(r)}{\tilde{\epsilon}_1(r) + 2\epsilon_1(r)} \quad \text{and} \quad y = \frac{[\epsilon_1(r) - \epsilon_2] \cdot [\tilde{\epsilon}_1(r) - \epsilon_1(r)]}{\epsilon_1(r)[\tilde{\epsilon}_1(r) + 2\epsilon_2]}. 
\]

Similarly, \( \tilde{\chi}_1(r = a) \) can be obtained, at least numerically, by solving Eq. (6), once the initial conditions, namely, \( \epsilon_1(r = 0) \) and \( \chi_1(r = 0) \) are given.

In what follows, we can investigate the nonlinear AC response of the graded spherical particle by seeing it as a homogeneous particle having the constitutive relation between the displacement and the local electric field, [18]

\[
\mathbf{D}_1 = \tilde{\epsilon}_1(r = a)\mathbf{E}_1 + \tilde{\chi}_1(r = a)E_1^2\mathbf{E}_1 \equiv \tilde{\epsilon}_1(r = a)\mathbf{E}_1,
\]

where \( \tilde{\epsilon}_1(r = a) \) and \( \tilde{\chi}_1(r = a) \) are determined by Eqs. (1) and (6), respectively. For the sake of convenience, we shall represent \( \tilde{\epsilon}_1(r = a) \) by \( \tilde{\epsilon}_1 \), \( \tilde{\chi}_1(r = a) \) by \( \tilde{\chi}_1 \) as well as \( \tilde{\chi}_1(r = a) \) by \( \tilde{\chi}_1 \), if no special instructions.
2. Nonlinear AC responses

If we apply a sinusoidal electric field like

$$E_0(t) = E_0 \sin(\omega t), \quad (7)$$

the local electric field $\sqrt{\langle E_1^2 \rangle}$ and the induced dipole moment

$$\tilde{p} = \tilde{\epsilon} a^3 \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1 + 2\tilde{\epsilon}_2} E_0 \quad (8)$$

will depend on time sinusoidally, too. Here the effective dielectric constant of the system ($\tilde{\epsilon}_e$) is given by the following dilute-limit expression,

$$\tilde{\epsilon}_e = \tilde{\epsilon}_2 + 3\tilde{\epsilon}_2 f \frac{\tilde{\epsilon}_1 - \tilde{\epsilon}_2}{\tilde{\epsilon}_1 + 2\tilde{\epsilon}_2}, \quad (9)$$

where $f$ is the volume fraction of the particles. By virtue of the inversion symmetry, the local electric field is a superposition of odd-order harmonics such that

$$\sqrt{\langle E_1^2 \rangle} = E_\omega \sin(\omega t) + E_{3\omega} \sin(3\omega t) + \cdots. \quad (10)$$

Similarly, the induced dipole moment contains harmonics as

$$\tilde{p} = p_\omega \sin(\omega t) + p_{3\omega} \sin(3\omega t) + \cdots. \quad (11)$$

These harmonics coefficients can be extracted from the time dependence of the solution of $\sqrt{\langle E_1^2 \rangle}$ and $\tilde{p}$.

3. Analytic solutions for the nonlinear AC responses

In what follows, we will perform a perturbation expansion method to extract the third harmonics of the local electric field and the induced dipole moment. It is known that the perturbation expansion method is applicable to weak nonlinearity only, limited by the convergence of the series expansion.

Let’s start from the dilute-limit expression for the effective linear dielectric constant ($\epsilon_e$) of the system of interest, namely, Eq. (9) where $\bar{\chi}_1 = \chi_2 = 0$. 
Next, we obtain the linear local electric fields $\langle E_1^2 \rangle$ and $\langle E_2^2 \rangle$, respectively,

$$\langle E_1^2 \rangle = \frac{E_0^2}{f} \frac{\partial \epsilon_1}{\partial \epsilon_1} \equiv F(\bar{\epsilon}_1, \epsilon_2, f, E_0),$$

$$\langle E_2^2 \rangle = \frac{E_0^2}{1 - f} \frac{\partial \epsilon_2}{\partial \epsilon_2} \equiv G(\bar{\epsilon}_1, \epsilon_2, f, E_0).$$

In view of the existence of nonlinearity inside the two components, we readily obtain the following local electric fields for the nonlinear particle and host, respectively,

$$\langle E_1^2 \rangle = F(\tilde{\bar{\epsilon}}_1, \tilde{\epsilon}_2, f, E_0),$$

$$\langle E_2^2 \rangle = G(\tilde{\bar{\epsilon}}_1, \tilde{\epsilon}_2, f, E_0).$$

For the below series expansions, we will take $\tilde{\bar{\epsilon}}_1 = \bar{\epsilon}_1 + \chi_1 E_1^2 \approx \bar{\epsilon}_1 + \bar{\chi}_1 \langle E_1^2 \rangle$ and $\tilde{\epsilon}_2 = \epsilon_2 + \chi_2 E_2^2 \approx \epsilon_2 + \chi_2 \langle E_2^2 \rangle$, in Eqs. (14) and (15), where $\langle \cdots \rangle$ denotes the volume average of $\cdots$. Let’s expand the local electric field $\langle E_1^2 \rangle$ and $\langle E_2^2 \rangle$ into a Taylor expansion, taking $\bar{\chi}_1 \langle E_1^2 \rangle$ and $\chi_2 \langle E_2^2 \rangle$ as the perturbative quantities,

$$\langle E_1^2 \rangle = F(\bar{\epsilon}_1, \epsilon_2, f, E_0) + \frac{\partial}{\partial \bar{\epsilon}_1} F(\bar{\epsilon}_1, \epsilon_2, f, E_0)|_{\bar{\epsilon}_1=\bar{\epsilon}_1} \bar{\chi}_1 \langle E_1^2 \rangle +$$

$$\frac{\partial}{\partial \epsilon_2} F(\bar{\epsilon}_1, \epsilon_2, f, E_0)|_{\epsilon_2=\epsilon_2} \chi_2 \langle E_2^2 \rangle + \cdots,$$

$$\langle E_2^2 \rangle = G(\bar{\epsilon}_1, \epsilon_2, f, E_0) + \frac{\partial}{\partial \bar{\epsilon}_1} G(\bar{\epsilon}_1, \epsilon_2, f, E_0)|_{\bar{\epsilon}_1=\bar{\epsilon}_1} \bar{\chi}_1 \langle E_1^2 \rangle +$$

$$\frac{\partial}{\partial \epsilon_2} G(\bar{\epsilon}_1, \epsilon_2, f, E_0)|_{\epsilon_2=\epsilon_2} \chi_2 \langle E_2^2 \rangle + \cdots.$$ 

Keeping the lowest orders of $\bar{\chi}_1 \langle E_1^2 \rangle$, we can rewrite Eq. (16) as,

$$\langle E_1^2 \rangle = h_1 E_0^2 + (h_2 + h_3) E_0^4,$$

where

$$h_1 = \frac{9\epsilon_2^2}{(\bar{\epsilon}_1 + 2\epsilon_2)^2}, \quad h_2 = \frac{-16\epsilon_2^4 \bar{\chi}_1}{(\bar{\epsilon}_1 + 2\epsilon_2)^5},$$

$$h_3 = \frac{18 \bar{\epsilon}_1 \epsilon_2 \chi_2 [(1 + 3f)\bar{\epsilon}_1^2 + (4 - 6f)\bar{\epsilon}_1 \epsilon_2 + (4 - 6f)\epsilon_2^2]}{(1 - p)(\bar{\epsilon}_1 + 2\epsilon_2)^5}.$$ 

Because of the time-dependence of the electric field [Eq. (7)], we can take one step forward to obtain the local electric field in terms of the harmonics ($E_\omega$ and $E_{3\omega}$),
\[ \sqrt{\langle E_1^2 \rangle} = E_\omega \sin(\omega t) + E_{3\omega} \sin(3\omega t), \]  

where

\[ E_\omega = \sqrt{h_1 E_0 + \frac{3}{8} h_2 + h_3 E_0^3}, \]

\[ E_{3\omega} = -\frac{1}{8} \frac{h_2 + h_3}{\sqrt{h_1}} E_0^3. \]

Similarly, based on Eq. (8), we obtain the induced dipole moment in terms of the harmonics \( p_\omega \) and \( p_{3\omega} \),

\[ \tilde{p}/a^3 = (p_\omega/a^3) \sin(\omega t) + (p_{3\omega}/a^3) \sin(3\omega t), \]

where

\[ p_\omega/a^3 = k_1 E_0 + \frac{3}{4} (k_2 + k_3) E_0^3, \]

\[ p_{3\omega}/a^3 = -\frac{1}{4} (k_2 + k_3) E_0^3, \]

with

\[ k_1 = \epsilon_e \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}, \quad k_2 = \frac{3 \epsilon_2^2 h_1 \chi_1 [\epsilon_1 + 6 f \epsilon_1 + (2 - 6 f) \epsilon_2]}{(\epsilon_1 + 2\epsilon_2)^3}, \]

\[ k_3 = j_1 \chi_2 [(1 + 3 f) \epsilon_1^3 - 18 f \epsilon_1^2 \epsilon_2 - 3 (2 - 3 f) \epsilon_1 \epsilon_2^2 - 2 (2 - 3 f) \epsilon_2^3] \]

\[(\epsilon_1 + 2\epsilon_2)^3.\]

In the above derivation, we have used an identity \( \sin^3(\omega t) = (3/4) \sin(\omega t) - (1/4) \sin(3\omega t) \).

### III. Numerical Results

For numerical calculations, we take \( \chi_1(r) = \chi_1(0) + D(r/a) \), and other parameters: volume fraction \( p = 0.09 \), external field strength \( E_0 = 1 \), linear part of host dielectric constant \( \epsilon_2 = 1 \).

The validation of using the DEDA is shown in Fig. 1. In this figure, we investigate the linear local electric field by using a power-law gradation profile inside the particles, in an attempt to compare the DEDA with the first-principles approach. As expected, the excellent
agreement is demonstrated between the DEDA [Eq. (2)] and the first-principles approach
[Eq. (5)]. In addition, it is worth noting that, for a linear gradation profile within the graded
particles, the first-principles approach holds as well [4], and the excellent agreement between
the two methods can also be found (figure not shown here).

Next, we discuss a power-law gradation profile \[\epsilon_1(r) = A \cdot (r/a)^n\], see Fig. 2. In this
figure, the harmonics of local electric field and the induced dipole moment are investigated as
a function of \(A\) for various \(n\). In this case, increasing \(A\) (or decreasing \(n\)) leads to increasing
\(\tilde{\epsilon}_1\) (namely, the equivalent dielectric constant of the graded particle under consideration) and
in turn yields a decreasing local electric field inside the particle. Thus, either an increase
in \(A\) or a decrease in \(n\) leads to the weakening third-order harmonics \((E_{3\omega} \text{ and } p_{3\omega})\), as
displayed in Fig. 2.

The \(x\)-axes of Figs. 3 and 4 represent the slope \((C)\) of a linear gradation profile. It is
because during the fabrication of graded spherical particles by using diffusion, the dielectric
constant at the center \(\epsilon_1(0)\) may vary slightly while that at the grain boundary can change
substantially.

For a linear gradation profile \[\epsilon_1(r) = \epsilon_1(0) + C \cdot (r/a)\], Fig. 3 shows the harmonics as
a function of \(C\) for various \(\epsilon_1(0)\). In this case, increasing \(C\) or \(\epsilon_1(0)\) yield increasing \(\tilde{\epsilon}_1\), and
hence one obtains the decreasing local electric field. As a result, larger \(C\) or \(\epsilon_1(0)\), weaker
the third-order harmonics \((E_{3\omega} \text{ and } p_{3\omega})\), see Fig. 3.

Fig. 4 displays the effect of \(\chi_2\) on the harmonics, for a linear gradation profile \[\epsilon_1(r) = \epsilon_1(0) + C \cdot (r/a)\]. Here, increasing \(\chi_2\) leads to increasing the local electric field inside the
graded particle of interest. Therefore, the third-order harmonics \((E_{3\omega} \text{ and } p_{3\omega})\) increase for
increasing \(\chi_2\).

As mentioned above, as \(A\) and \(C\) increases, the equivalent dielectric constant of the
particle should be increased accordingly, which in turn yields a decreasing local electric
field, and hence, in Figs. 2~4, \(E_\omega\) decreases for increasing \(A\) or \(C\). On the other hand, it
is found that, in Figs. 2~4, \(p_\omega\) increases for increasing \(A\) or \(C\) which is, in fact, due to the
increasing effective dielectric constant \(\tilde{\epsilon}_e\) [refer to Eq. (8)]. Similarly, this analysis works
fairly for understanding the effect of \( n \) and \( \epsilon_1(0) \) on \( E_\omega \) and \( p_\omega \), as displayed in Figs. 2 and 3. However, increasing \( \chi_2 \) can increase not only the local electric field inside the particles, but also the effective dielectric constant \( \tilde{\epsilon}_e \), and hence we observe increasing \( E_\omega \) and \( p_\omega \), as shown in Fig. 4.

In addition, we also discuss the effect of nonlinear-susceptibility gradation profiles (no figures shown here). For linear gradation profile \( \chi_1(r) = \chi_1(0) + D(r/a) \), as \( \chi_1(0) \) (or \( D \)) increases, the third harmonics of both the electric field and the induced dipole moment increases accordingly. On the other hand, for pow-law gradation profile \( \chi_1(r) = B(r/a)^m \), increasing \( B \) (or decreasing \( m \)) leads to increasing third harmonics. To understand such results, we can again resort to the above analysis on the effect of the relevant parameters on the local field as well as the effective dielectric constant.

**IV. DISCUSSION AND CONCLUSION**

Here some comments are in order. We investigate the nonlinear AC responses of the graded material where linear/nonlinear graded particles are randomly embedded in a linear/nonlinear host medium in the dilute limit. As a matter of fact, the NDDEA (nonlinear differential effective dipole approximation) is valid for arbitrary gradation profiles, besides the power-law and linear profiles of interest. In particular, based on the first-principles approach, the exact solution is obtainable, for not only power-law profiles (see Section II A), but also linear profiles (refer to Ref. [4]).

As an extension, it is of particular interest to see what happens to the nonlinear AC responses of graded particles in electrorheological fluids, in which a field-induced anisotropic structure often occurs. For discussing this anisotropy effect, we can make use of the Maxwell-Garnett approximation for anisotropic structures [13].

To sum up, based on our recently-established NDEDA, we have investigated the nonlinear AC responses of a composite with linear/nonlinear graded spherical particles embedded in a linear/nonlinear host medium, and found the fundamental and third harmonic AC responses
are sensitive to the dielectric-constant (or nonlinear-susceptibility) gradation profile within the particles. Again, for extracting the linear local electric field, the DEDA agrees very well with the first-principles approach. Thus, by measuring the AC responses of the graded composites, it is possible to perform a real-time monitoring of the fabrication process of the gradation profiles within graded particles.

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REFERENCES

[1] G. W. Milton, *The Theory of Composites*, Chapter 7 (Cambridge University Press, Cambridge, 2002).

[2] M. Yamanouchi, M. Koizumi, T. Hirai and I. Shioda, in *Proceedings of the First International Symposium on Functionally Graded Materials* (Sendi, Japan, 1990).

[3] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).

[4] L. Dong, G. Q. Gu, and K. W. Yu, Phys. Rev. B 67, 224205 (2003).

[5] G. Q. Gu and K. W. Yu, J. Appl. Phys., to be published on Sep. 15, 2003.

[6] K. W. Yu, G. Q. Gu and J. P. Huang, preprint: cond-mat/0211532.

[7] J. P. Huang, K. W. Yu, G. Q. Gu, and M. Karttunen, Phys. Rev. E 67, 051405 (2003).

[8] L. Gao, J. P. Huang, and K. W. Yu, preprint: cond-mat/0308032.

[9] D. J. Bergman and D. Stroud, Solid State Phys. 46, 147 (1992).

[10] O. Levy, D. J. Bergman, and D. Stroud, Phys. Rev. E 52, 3184 (1995).

[11] P. M. Hui, P. C. Cheung, and D. Stroud, J. Appl. Phys. 84, 3451 (1998).

[12] G. Q. Gu, P. M. Hui, and K. W. Yu, Physica B 279, 62 (2000).

[13] J. P. Huang, J. T. K. Wan, C. K. Lo, and K. W. Yu, Phys. Rev. E 64, 061505(R) (2001).

[14] J. P. Huang, L. Gao, and K. W. Yu, J. Appl. Phys. 93, 2871 (2003).

[15] D. J. Klingenberg, MRS Bull. 23, 30 (1998).

[16] J. T. K. Wan, G. Q. Gu, and K. W. Yu, Phys. Rev. E 63, 052501 (2001).

[17] G. Q. Gu and K. W. Yu, Phys. Rev. B 46, 4502 (1992).

[18] D. Stroud and P. M. Hui, Phys. Rev. B 37, 8719 (1988).
FIGURES

FIG. 1. For power-law gradation profile $\epsilon_1(r) = A \cdot (r/a)^n$, comparison between the approximation result [obtained from the DEDA, Eq. (2)] and the exact solution [predicted by a first-principles approach, Eq. (5)], for linear electric field $E_1^{(\text{lin})}$ as a function of $A$ for various $n$. Parameters: $\chi_1(0) = 0.1$, $D = 0.1$, $\chi_2 = 0$.

FIG. 2. For power-law gradation profile $\epsilon_1(r) = A \cdot (r/a)^n$, harmonics of the local electric field and induced dipole moment, as a function of $A$, for various $n$. Parameters: $\chi_1(0) = 0.1$, $D = 0.1$, $\chi_2 = 0$.

FIG. 3. For linear gradation profile $\epsilon_1(r) = \epsilon_1(0) + C \cdot (r/a)$, harmonics of the local electric field and induced dipole moment, as a function of $C$, for various $\epsilon_1(0)$. Parameters: $\chi_1(0) = 0.1$, $D = 0.1$, $\chi_2 = 0$.

FIG. 4. Same as Fig. 3, but for various $\chi_2$. Parameters: $\chi_1(0) = 0$, $D = 0$, $\epsilon_1(0) = 3$. 

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