Nonlinear Alternating Current Responses of Electrorheological Solids

J. P. Huang[†]

Department of Physics, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong, and Max Planck Institute for Polymer Research, Ackermannweg 10, 55128, Mainz, Germany

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When a composite containing nonlinear dielectric particles suspended in a host medium is subjected to a sinusoidal alternating current (ac) electric field, the dielectric response of the composite will generally consist of ac fields at frequencies of higher order harmonics. For an electrorheological (ER) solid under structure transformations due to external fields, we apply the Ewald−Kornfeld formulation to derive the local electric fields and induced dipole moments explicitly, and then we perform the perturbation expansion method to extract their fundamental and third-order harmonics analytically. It is shown that the degree of anisotropy of the ER solid can affect these harmonics significantly. Our results are well understood in the spectral representation theory. Thus, it seems possible to perform a real-time monitoring of the structure transformation by measuring the nonlinear ac responses of ER solids.

I. Introduction

Electrorheological (ER) fluids[1−4] have received much attention due to the potential application of the rapid field-induced aggregation[2] and the large anisotropy,[5] and they are actually a suspension in which the induced dipole moment can order the suspended polarizable particles into columns under the application of a strong electric field. Tao and Jiang examined the temporal evolution of three-dimensional structure in ER fluids by using a computer simulation.[6] For a wide range of the ratio of the Brownian force to the dipolar force, the ER fluid was found[6] to have a rapid chain formation followed by aggregation of chains into thick columns which have a body-centered tetragonal (bct) lattice structure, and the Peierls−Landau instability of single chains helps in the formation of thick columns.

An ER fluid can turn into an ER solid as the external field exceeds a critical field. For the ER solid, it is known that its ground state is a bct lattice. Tao and Jiang[7] proposed that a structure transformation of the ER solid from the bct lattice to some other lattices can appear when a magnetic field is simultaneously applied perpendicular to the electric field and the particles have magnetic dipole moments. Then, this proposal was verified experimentally and a structure transformation from the bct lattice to the face-centered cubic (fcc) lattice was observed indeed.[8] Recently, Lo and Yu have also shown that an alternative structure transformation from the bct structure to the fcc can appear under the application of electric fields only.[9]

Finite-frequency responses of nonlinear dielectric composite materials have also attracted much attention both in research and in industrial applications during the last two decades.[10] In particular, when a composite containing nonlinear dielectric particles embedded in a linear (or nonlinear) dielectric host medium is subjected to a sinusoidal alternating current (ac) electric field, the dielectric response of the composite will, in general, consist of ac fields at frequencies of higher order harmonics.[5,11−16] In experiments, 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.[2] In this case, the strength of the nonlinear polarization should be reflected in the magnitude of the harmonics. From the theoretical point of view, the perturbation expansion[5,14,16] and self-consistent methods[5,17] can be used for extracting such kinds of harmonics.

In this work, to investigate the lattice effect on the nonlinear ac responses, we shall use the Ewald−Kornfeld formulation[9,18−20] to derive the local electric fields and induced dipole moments in the ER solid, which is subject to a structure transformation due to external fields, and then perform the perturbation expansion method to obtain their fundamental and third-order harmonics. To this end, it is shown that the fundamental and third-order harmonics are sensitive to the degree of anisotropy of ER solids. Thus, by measuring the nonlinear ac responses, it seems possible to perform a real-time monitoring of the structure transformation of ER solids.

This paper is organized as follows. In Section II, we apply the Ewald−Kornfeld formulation[9,18,19] to derive the local electric field and induced dipole moment in ER solids, and then perform the perturbation expansion method to extract their fundamental and third-order harmonics. In Section III, we numerically investigate these harmonics under different conditions, which is followed by a discussion and conclusion in Section IV.

II. Formalism

A. Local Electric Field and Induced Dipole Moment: Ewald−Kornfeld Formulation. Let us start by considering the ground state of an ER solid, namely a bct (body-centered tetragonal) lattice, which can be regarded as a tetragonal lattice, plus a basis of two particles each of which is fixed with a point dipole at its center. One of the two particles is located at a corner and the other one at the body center of the tetragonal unit cell. Its lattice constants are denoted by $a(=b) = \sqrt{q}^{-1/2}$ and $c = q\sqrt{}$ along the $x(y)$ and z axes, respectively. In this case, the uniaxial anisotropic axis is directed along the z axis. As q varies, the volume of the unit cell remains the same, $V_c = \beta$. Consequently, the degree of anisotropy of the tetragonal lattice is measured by how q deviates from unity.

In case of an x-directed external electric field $\vec{E}_0$, the dipole moments $\vec{p} = p\hat{x}$ are perpendicular to the uniaxial anisotropic

[†] E-mail: jphuang@alumni.cuhk.net.

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axis. Then, the local field $\vec{E}$ (e.g., $\vec{E} = E_x\hat{x}$, $E_z = 0$) at the lattice point $\vec{R} = \vec{0}$ has the following form[9,18−20]

$$E_x = p\sum_{j=1}^{2}\sum_{R\neq 0}[-\gamma_1(R_j) + x_j^2 q^2 \gamma_2(R_j)] - \frac{4\pi p}{V_c}\sum_{G\neq 0}\Pi(\vec{G})\frac{G_x^2}{G^2}\exp\left(\frac{-G^2}{4\eta^2}\right) + \frac{4p\eta^3}{3\sqrt{\pi}} \quad (1)$$

In this equation, γ_1 and γ_2 are two coefficients, given by

$$\gamma_1(r) = \frac{\mathrm{erfc}(\eta r)}{r^3} + \frac{2\eta}{\sqrt{\pi}r^2}\exp(-\eta^2 r^2)$$

$$\gamma_2(r) = \frac{3\mathrm{erfc}(\eta r)}{r^5} + \left(\frac{4\eta^3}{\sqrt{\pi}r^2} + \frac{6\eta}{\sqrt{\pi}r^4}\right)\exp(-\eta^2 r^2)$$

where $\mathrm{erfc}(\eta r)$ is the complementary error function, and η an adjustable parameter making the summation converge rapidly. In eq 1, R and G denote the lattice vector and the reciprocal lattice vector, respectively,

$$\vec{R} = \ell(q^{-1/2}l\hat{x} + q^{-1/2}m\hat{y} + qn\hat{z})$$

$$\vec{G} = \frac{2\pi}{\ell}(q^{1/2}u\hat{x} + q^{1/2}v\hat{y} + q^{-1}w\hat{z})$$

where l, m, n, u, v, and w are integers. In addition, x_j and R_j of eq 1 are given by respectively

$$x_j = l - \frac{j-1}{2} \text{ and } R_j = |\vec{R} - \frac{j-1}{2}(a\hat{x} + a\hat{y} + c\hat{z})|$$

and the structure factor $\Pi(\vec{G}) = 1 + \exp[i(u + v + w)/\pi]$.

So far, based on a self-consistent method, we apply the result of the local field to evaluate the effective polarizability α_{eff} of the dipole lattice,

$$\alpha_{\mathrm{eff}} = \frac{\alpha}{1 - \alpha\gamma_x/V_c} \quad (2)$$

where α stands for the polarizability of an isolated dipole, and $\gamma_x = E_x V_c/p$, the local field factor that was measured in the computer simulations by Martin et al.[21,22] Let us use γ_z and γ_x ($\equiv\gamma_y$) to denote the local field factors parallel and perpendicular to the uniaxial anisotropic axis, respectively. Accordingly, we have $\gamma_z = \gamma_x = 4\pi/3$ for the bcc lattice ($q = 1$). In what follows, we set $\gamma' = 3\gamma/4\pi$. It is worth noting that γ' is a function of a single variable, namely degree of anisotropy q. Also, γ'_z and γ'_x satisfy the sum rule $\gamma'_z + 2\gamma'_x = 3$,[23] and $\gamma'_z = \gamma'_x = 1$ [at $q = 1$ (bcc)] just represents the isotropic limit.

Based on eq 2, it is straightforward to derive the dipole factor (also called the Clausius−Mossotti factor) for a specific particle b'_x and b'_z along the x (or y) and z axes, respectively

$$b'_{x(z)} = \frac{b}{1 - b\rho\gamma'_{x(z)}} \quad (3)$$

where ρ stands for the volume fraction of the particles, and b the known dipole factor for an isolated particle, $b = (\epsilon_1 - \epsilon_2)/(\epsilon_1 + 2\epsilon_2)$. Here ϵ_1 and ϵ_2 denote the dielectric constants of the particle and host fluid, respectively. The two parameters, b'_x and b'_z, have been represented as $b'_{x(z)}$ by using the joint subscript $x(z)$. A similar notation is also used for $\gamma'_{x(z)}$. In what follows, for convenience, $\mathcal{X}^{(T)[(L)]}$ will be used to stand for $\mathcal{X}^{(T)}$ and $\mathcal{X}^{(L)}$, which represent the transverse [i.e., the external field is parallel to the x (or y) axis] and longitudinal [namely, the external field is parallel to the z axis] field cases of the quantity $\mathcal{X}$, respectively.

So far, the local field effect arising from all the other particles in the lattice has been explicitly included in eq 3 by using the Ewald−Kornfeld formulation. Next, let us see the particle as the one embedded in an effective medium with an effective dielectric constant $\epsilon_e^{(T)[(L)]}$ which can be determined by

$$b'_{x(z)} = \frac{\epsilon_1 - \epsilon_e^{(T)[(L)]}}{\epsilon_1 + 2\epsilon_e^{(T)[(L)]}} \quad (4)$$

It is worth remarking that this $\epsilon_e^{(T)[(L)]}$ has included the detailed structural information of the lattice, as expected.

As the external field $\vec{E}_0$ is along the x or z axis, the volume average of the local electric field is given by

$$\langle\vec{E}_1^{(T)[(L)]}\rangle = \frac{3\epsilon_e^{(T)[(L)]}}{\epsilon_1 + 2\epsilon_e^{(T)[(L)]}}\vec{E}_0 \quad (5)$$

where $\langle\ \rangle$ stands for the volume average. In view of eqs 3 and 4, $\epsilon_e^{(T)[(L)]}$ of eq 5 is given by

$$\epsilon_e^{(T)[(L)]} = \frac{b\rho\gamma'_{x(z)} + b - 1}{b\rho\gamma'_{x(z)} - 2b - 1}\epsilon_1 \quad (6)$$

Then, it takes one step forward to express the corresponding induced dipole moment inside the particle

$$\vec{p}_1^{(T)[(L)]} = \epsilon_e^{(T)[(L)]} r_a^{\ 3} b'_{x(z)}\vec{E}_0 \quad (7)$$

So far, we have derived the local electric fields [eq 5] and induced dipole moments [eq 7], by taking into account the lattice effect in a rigorous manner. In what follows, we shall use p_1 (or $\tilde{p}_1$), γ', E_1 (or $\tilde{E}_1$), and ϵ_e to denote both the transverse and longitudinal field cases, if there are no special instructions.

B. Nonlinear Polarization and Its Higher Order Harmonics. For studying the nonlinear ac responses, let us consider the particle with a cubic nonlinearity like

$$\tilde{\epsilon}_1 = \epsilon_1 + \chi_1 E_1^{\ 2} \approx \epsilon_1 + \chi_1\langle E_1^{\ 2}\rangle \quad (8)$$

where χ_1 represents the nonlinear susceptibility of the particles embedded in a linear host fluid. Throughout the paper, we focus on the weak nonlinearity only, namely $\chi_1\langle E_1^{\ 2}\rangle \ll \epsilon_1$, and the low concentration limit, and we use "∼" to indicate the quantities having a nonlinear characteristic.

If a sinusoidal electric field

$$E_0(t) = E_0\sin(\omega t) \quad (9)$$

is applied, the local electric field $\langle\tilde{E}_1\rangle$ and the induced dipole moments $\tilde{p}_1$ will depend on time sinusoidally, too.

Owing to the inversion symmetry of the system, the local electric field is a superposition of odd-order harmonics such that[5,14]

$$\langle\tilde{E}_1\rangle = E_\omega\sin(\omega t) + E_{3\omega}\sin(3\omega t) + \cdots \quad (10)$$

Accordingly, the induced dipole moment contains harmonics as[5,14]

$$\tilde{p}_1 = p_\omega\sin(\omega t) + p_{3\omega}\sin(3\omega t) + \cdots \quad (11)$$

For the purpose of normalization, we shall use $p_0 = \epsilon_2 r_a^3 b E_0$, which is independent of the nonlinear characteristic as well as the degree of anisotropy γ'.

C. Analytic Solutions. In what follows, we apply the perturbation expansion method to extract the harmonics of the local electric fields (E_ω and $E_{3\omega}$) and the induced dipole moments (p_ω and $p_{3\omega}$).

Let us denote the volume average of the linear local electric field (namely, the local electric field at which the nonlinear characteristics of the particles disappears) [eq 5] as

$$\langle E_1 \rangle \equiv F(\epsilon_1, \epsilon_2, \rho, \gamma', E_0) \tag{12}$$

In view of the existence of nonlinearity, we have

$$\langle \tilde{E}_1 \rangle \equiv F(\tilde{\epsilon}_1, \epsilon_2, \rho, \gamma', E_0) \tag{13}$$

Next, expand the local field $\langle \tilde{E}_1 \rangle$ into a Taylor expansion, taking $\chi_1 \langle E_1^2 \rangle$ as the perturbative quantity. As a result, we obtain[5]

$$\langle \tilde{E}_1 \rangle = F(\epsilon_1, \epsilon_2, \rho, \gamma', E_0) + \frac{\partial}{\partial \tilde{\epsilon}_1} F(\tilde{\epsilon}_1, \epsilon_2, \rho, \gamma', E_0)|_{\tilde{\epsilon}_1 = \epsilon_1} \chi_1 \langle E_1^2 \rangle + \cdots \tag{14}$$

Keeping the lowest orders of $\chi_1 E_0(t)^2$ and $\chi_1 \langle E_1^2 \rangle$, because of the time dependence of the external electric field [eq 9], we obtain the local electric fields and induced dipole moments in terms of the harmonics, E_ω, $E_{3\omega}$, p_ω, and $p_{3\omega}$ [note the higher order harmonics (namely, fifth-order, seventh-order, etc.) have been neglected]. Here the harmonics of the local electric fields and induced dipole moments are respectively given by

$$\chi_1^{1/2} E_\omega = \frac{W_1}{W_2} (\chi_1^{1/2} E_0) - \frac{9\epsilon_2 W_1^2}{4 W_2^4} (\chi_1^{1/2} E_0^3)^3 \tag{15}$$

$$\chi_1^{1/2} E_{3\omega} = \frac{3\epsilon_2 W_1^2}{4 W_2^4} (\chi_1^{1/2} E_0)^3 \tag{16}$$

$$p_\omega / p_0 = -\frac{W_1(\epsilon_1 + 2\epsilon_2)(\Omega_1 + \Omega_2)}{4 W_2^2 \epsilon_2 (\epsilon_1 - \epsilon_2)} \tag{17}$$

$$p_{3\omega}/p_0 = \frac{W_1^2(\epsilon_1 + 2\epsilon_2)}{4\epsilon_2 W_2^4 (\epsilon_1 - \epsilon_2)[\gamma'\epsilon_2\rho + \epsilon_1(3 - \gamma'\rho)]^2} \sum_{N=1}^{5} Q_N (\chi_1^{1/2} E_0)^2 \tag{18}$$

where

$$W_1 = -\gamma'\epsilon_1\rho + \epsilon_2(3 + \gamma'\rho)$$

$$W_2 = \epsilon_1 - \gamma'\epsilon_1\rho + \epsilon_2(2 + \gamma'\rho)$$

$$\Omega_1 = -\frac{4\epsilon_1 W_2(\epsilon_1 - \epsilon_2)}{\gamma'\epsilon_2\rho + \epsilon_1(3 - \gamma'\rho)}$$

$$\Omega_2 = \frac{3W_1}{[\gamma'\epsilon_2^2\rho(2 + \gamma'\rho) + U_1 + U_2]^2} \sum_{N=1}^{5} Q_N (\chi_1^{1/2} E_0)^2$$

with

$$Q_1 = 4\gamma'\epsilon_1^3\epsilon_2\rho(3 + \gamma'\rho - \gamma'^2\rho^2)$$

$$Q_2 = \gamma'\epsilon_1^4\rho(3 - 4\gamma'\rho + \gamma'^2\rho^2)$$

$$Q_3 = \gamma'\epsilon_2^4\rho(6 + 5\gamma'\rho + \gamma'^2\rho^2)$$

$$Q_4 = -2\gamma'\epsilon_1\epsilon_2^3\rho(6 + 7\gamma'\rho + 2\gamma'^2\rho^2)$$

$$Q_5 = 3\epsilon_1^2\epsilon_2^2(-9 - 3\gamma'\rho + 3\gamma'^2\rho^2 + 2\gamma'^3\rho^3)$$

$$U_1 = Q_1/(2\gamma'\epsilon_1^2\rho),\ U_2 = Q_2/(\gamma'\epsilon_1^2\rho)$$

III. Numerical Results

We are now in a position to do some numerical calculations to discuss the effect of the degree of anisotropy q on the harmonics of the local electric fields and induced dipole moments. Without loss of generality, take $\epsilon_1 = 30\epsilon_0$, $\epsilon_2 = 2.25\epsilon_0$ (dielectric constant of silicone oil), and $\rho = 0.1$ for numerical calculations, where ϵ_0 denotes the dielectric constant of free space. In fact, here $\rho = 0.1$ corresponds to the dilute limit. Nevertheless, it would be realizable as the particles can have a (solid) hard core and a relatively soft coating (by long polymer chains, etc.) to avoid aggregation. Thus the model of a soft sphere with an embedded point dipole can be used. In this case, the many-body (local field) effects are important, while the multipolar effects can be neglected. This is exactly part of the emphasis in the present work. On the other hand, for a higher ρ, similar results can be achieved indeed, as expected.

In Figure 1, we investigate the dependence of the local field factor[4] on the degree of anisotropy q. Increasing q causes γ_x' (or γ_z') to increase (or decrease). Also, a plateau is shown at $\gamma_x' = \gamma_z' = 1$, which actually includes the transformations ranging from the bcc ($q = 1$) lattice to the fcc ($q = 2^{1/3}$). Accordingly, a similar plateau occurs at all other figures (see Figures 2 and 3).

Figure 2 displays the fundamental and third-order harmonics of the local electric field and induced dipole moment, as a function of the degree of anisotropy q for different $\chi_1^{1/2} E_0$ for the transverse field case. It is found that increasing q causes both the fundamental and third-order harmonics of the local electric field to decrease. Accordingly, the fundamental harmonics of the induced dipole moment is caused to decrease as well. However, the third-order harmonics of the induced dipole moment is caused to increase as q increases. Note the three curves in Figure 1c are overlapped.

Similarly, we also investigate the harmonics of the local electric field and induced dipole moment in Figure 3, but for the longitudinal field case. In contrast to the transverse field case (Figure 2), the exactly opposite effects have been shown for the longitudinal field case (Figure 3), due to the opposite dependence of q on γ_x' and γ_z', see Figure 1. Again, the three curves in Figure 1c are overlapped.

Both Figures 2 and 3 show that the harmonics are strongly dependent on the nonlinear response of the suspended particles. Moreover, increasing the nonlinear response $\chi_1^{1/2} E_0$ leads to an increase in the harmonics. This is in agreement with the results of our recent work.[5,24] In addition, we also studied the effect of volume fraction ρ, and found that increasing the volume fraction can enhance the harmonics too (no figures shown here).

To understand the results, we resort to the spectral representation approach.[25] Let us start by denoting $s = (1 - \epsilon_1/\epsilon_2)^{-1}$, and

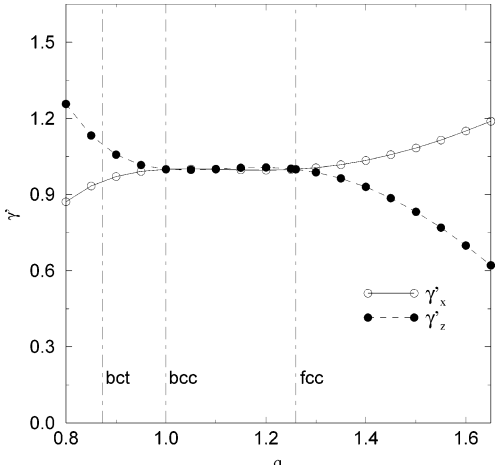

Figure 1. Local field factor γ' versus the degree of anisotropy q. The bct (ground state), bcc, and fcc lattices which are respectively related to $q = 0.87358$, 1.0, and $2^{1/3}$ are also shown (dot−dashed lines).

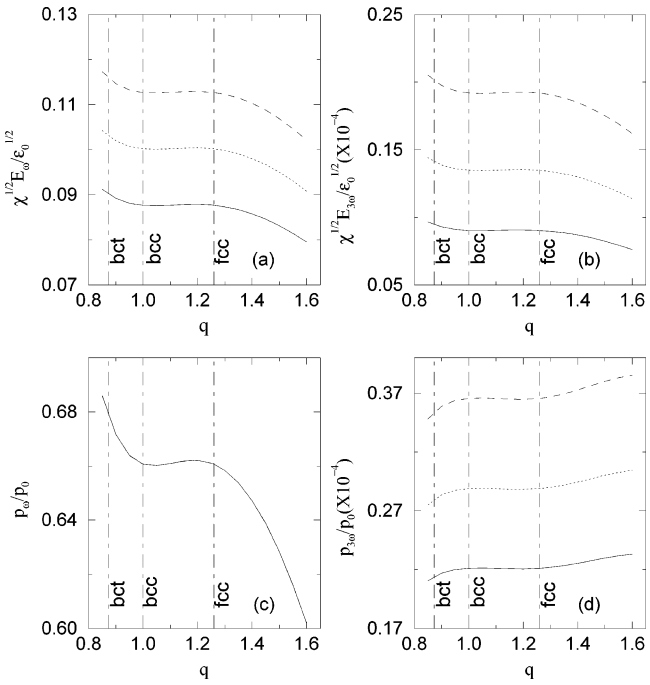

Figure 2. The fundamental and third-order harmonics of the local electric field and induced dipole moment, as a function of q for $\chi_1^{1/2}E_0 = 0.7\epsilon_0^{1/2}$ (solid lines), $\chi_1^{1/2}E_0 = 0.8\epsilon_0^{1/2}$ (dotted lines), and $\chi_1^{1/2}E_0 = 0.9\epsilon_0^{1/2}$ (dashed lines), for the transverse field case. Note the three curves in part c are overlapped. Parameters: $\epsilon_1 = 30\epsilon_0$, $\epsilon_2 = 2.25\epsilon_0$, and $\rho = 0.1$. The bct, bcc, and fcc lattices are shown as well (dot−dashed lines).

then the local electric field given by eq 5 admits

$$\langle E_1 \rangle = E_0 + \frac{F_1}{s - s_1} E_0 \tag{19}$$

where the residue $F_1 = {}^1/_3$ and the pole $s_1 = (1 - \rho\gamma')/3$. As $\rho \to 0$, s_1 tends to ${}^1/_3$, which produces the known value for an isolated particle. The substitution of the present model parameters yields $s = -0.081$, and hence we have

$$\langle E_1 \rangle = E_0 + \frac{1/3}{-0.081 - (1 - 0.1\gamma')/3} E_0 \tag{20}$$

Based on this equation, it is apparent to see that increasing γ_x' or γ_z' causes the electric field to decrease. In addition, we have shown that increasing q causes γ_x' (or γ_z') to increase (or decrease), see Figure 1. Also, a plateau was shown at $\gamma_x' = \gamma_z' = 1$, which actually includes the transformations ranging from the bcc ($q = 1$) lattice to the fcc ($q = 2^{1/3}$). Because of the dependence of the harmonics on γ', similar plateaus have already been shown in Figures 2 and 3, too. As a matter of fact, the effective linear dielectric constant mainly contributes to the magnitude of the fundamental harmonics of the induced dipole moment or local electric field. In the meantime, because of the existence of weak nonlinearity, the nonlinear polarization just has a perturbation effect, which can be neglected when compared to the linear part. Since the spectral representation approach is a different way of formulating the linear local electric field, to some extent, it can be used to explain the fundamental harmonics of the local electric field and hence those of the induced dipole moment which are displayed in Figure 2a,c and Figure 3a,c. Based on eq 20 (namely, an expression obtained from the spectral representation approach), it is convenient for us to understand that a decrease (or increase) of q should lead to an increase of the local electric field for transverse (or longitudinal) field cases. In this connection, for transverse (or longitudinal) field cases, the fundamental harmonics of the local electric field and hence corresponding induced dipole moment should increase as q decreases (or increases). This has also been displayed in Figure 2a,c (or Figure 3a,c). The explanation on the third-order harmonics of the local electric field and induced dipole moment seems to be more complicated. Nevertheless, since the strength of the nonlinear polarization of nonlinear materials should be reflected in the magnitude of the high-order harmonics of local electric fields and induced dipole moments, the effective nonlinear part in the effective nonlinear dielectric constant should be expected to determine the magnitude of the third-order harmonics. It appears that the third-order harmonics of the local electric field show a behavior similar to its fundamental harmonics. However, inverse behavior occurs to the third-order harmonics of the induced dipole moment. This difference would possibly be due to the nonlinearity fluctuation that might arise from the long-range interaction. This interaction existing in the system is explicitly evaluated in the current paper, by using the Ewald technique.

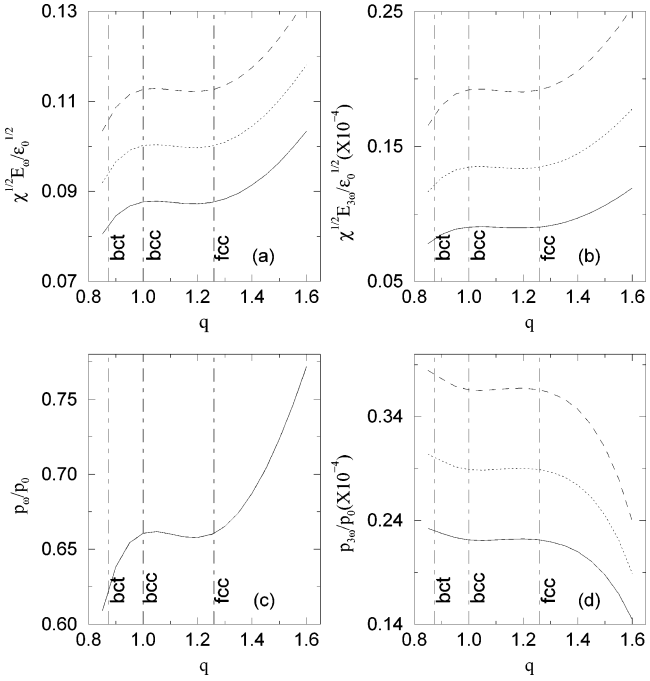

Figure 3. Same as Figure 2, but for the longitudinal field case.

IV. Discussion and Conclusion

Here some comments are in order. In this work, we have used the Ewald−Kornfeld formulation to derive the local electric fields and induced dipole moments in a rigorous manner, in an attempt to take into account the lattice (local field) effect on the nonlinear ac responses of ER solids under structure transformations.

In fact, we presented an (approximate) effective medium theory (EMT) for considering the local field effect on the electrorotation and dielectric dispersion spectra of colloidal particles or biological cells in the previous paper.[26] However, we were unable to study the detailed structural information via the EMT. In the present paper, we have considered the local field effect in a rigorous manner, based on the Ewald−Kornfeld formulation, which is quite different from the EMT.

We have considered the fundamental and third-order harmonics. As a matter of fact, it should be straightforward to investigate the higher order (e.g., fifth-order, seventh-order, etc.) harmonics by keeping more terms in eq 14. Due to the virtue of symmetry, only the odd-order harmonics appear. In fact, the even-order harmonics can also be induced to occur due to the coupling between the ac and dc external fields.[16] Since the second-order harmonics can often be of several orders of magnitude larger than the third-order, it is of value to discuss the lattice effect on the nonlinear ac responses by applying an ac field coupling with a dc field, to investigate the second-order harmonics. Finally, this work can be extended to polydisperse ER solids[27] in which the permittivities of particles can possess a distribution, or to a system[28] where anisotropies of material and geometry are caused to exist due to fabrication methods or field effects. On the other hand, the nonlinear response of composites to ac fields will become more complicated when the ac field contains different basic frequencies or different magnitudes of external fields at different basic frequencies.[29]

Our former work[5] covers a wider range of the degree of anisotropy q. However, the explicit form of the local field−q relation was lacking. By invoking a lattice structure (which exists in ER solids), this can be achieved, as is evident in this work. In detail, in ref 5 we used an effective medium theory to derive effective (anisotropic) dielectric constants, based on the Maxwell−Garnett approximation. In contrast, in the current paper, the Ewald technique is used to derive the effective dielectric constants, by including the long-range interaction explicitly (which was, however, lacking in ref 5). In this sense, for treating ER solids with lattice structures, the present derivation of the effective dielectric constants should be expected to be more accurate. Interestingly, some result of the current paper was also predicted in ref 5. For instance, as q decreases, for longitudinal (or transverse) field cases γ_z' (or γ_x') is caused to increase (or decrease), thus yielding an increasing (or decreasing) third-order harmonics of the induced dipole moment, see Figure 3d (or Figure 2d). Here γ_z' (or γ_x') corresponds to β_L (or β_T) of ref 5. It is worth mentioning that a different normalization factor (i.e., p_0) was used in ref 5.

To sum up, based on the Ewald−Kornfeld formulation, we have investigated the nonlinear responses of an ER solid with nonlinear spherical particles embedded in a linear host fluid and found that the fundamental and third-order harmonic ac responses are sensitive to the degree of anisotropy within the ER solid. Our results have been well understood through the spectral representation theory. Thus, by measuring the nonlinear ac responses of ER solids, it seems possible to perform a real-time monitoring of structure transformations.

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