Interparticle force in electrorheological solids: Many-body dipole-induced dipole model

J. P. Huang[1,2,*] and K. W. Yu[1,†]
[1]*Department of Physics, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong*
[2]*Max Planck Institute for Polymer Research, Ackermannweg 10, 55128, Mainz, Germany*
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One often investigates electrorheological (ER) solids by using the point-dipole (PD) approximation, which, however, is known to err considerably for touching particles due to the existence of many-body (local-field) effects and multipolar interactions. Beyond the PD model, previous attempts have been restricted to either local-field effects only or multipolar interactions only, but not both. In the present work, we have developed a many-body dipole-induced dipole model for an ER solid the lattice structure of which can be changed due to the application of external fields, in an attempt to take into account both local-field effects and multipolar interactions. The results show that the multipolar interaction can indeed be dominant over the dipolar interaction, while the local-field effect may yield an important correction. Also, the results are well understood with the aid of spectral representation theory.

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I. INTRODUCTION

In electrorheological (ER) fluids [1], the suspended polarizable particles can form columns under the application of a strong electric field [2]. The rapid field-induced aggregation [3] and the large anisotropy [4] of ER fluids render these materials potentially important for applications. As the external field exceeds a critical field, the ER fluid turns into an ER solid, the ground state of which is a body-centered tetragonal (bct) lattice. It has been found that the structure of the ER solid can be changed from the bct lattice to other lattices by the application of external electric and/or magnetic fields [5–7].

To discuss the ER effect, the existing theory [8] and simulation [9] are often based on the simple point-dipole (PD) approximation. Since many-body (local-field) effects and multipolar interactions between particles have been neglected, the predicted strength of the ER effect is of an order lower than the experimental results. Hence, much work has been done to sort out more accurate models [9–13]. Recently, we put forth a dipole-induced dipole (DID) model [14] to improve the PD model. In that work, we considered the particle interaction between two polarized dielectric particles by means of the multiple image formula, but neglected the local-field effect because it was believed that the multipolar interactions can be dominant over the local-field effects. To some extent, we should question the quantitative accuracy of this claim since in ER solids chain and sheet structures are known to occur. In fact, this issue has already been quantified for particles with large dielectric contrast with the liquid phase by Martin and Anderson [15].

In view of the fact that the previous work treated either local-field effects only [16] or multipolar interactions only [12], but not both, it is necessary to develop a different model which can be used to study both the local-field effects and the multipolar interactions. Thus, in this work, we shall use the Ewald-Kornfeld formulation [7,17,18] to calculate the effective dielectric constant of ER solids, in order to derive an analytic formula for the many-particle DID model. Here the effective dielectric constant contains the desired detailed lattice structural information of ER solids. To one's interest, by using this model, we can assess the importance of the local-field effects against the multipolar interactions.

This paper is organized as follows. In Sec. II, we present the many-body DID model and derive the effective dielectric constant of the ER solid, based on the Ewald-Kornfeld formulation, by taking into account the lattice effect. In Sec. III, we investigate the local-field effect and multipolar interaction by calculating the interparticle force as well as the dipole factor (also called the Clausius-Mossotti factor). This is followed by a discussion and conclusion in Sec. IV.

II. FORMALISM

We concentrate on the case where highly polarized dielectric particles of diameter d and dielectric constant ϵ_1 are embedded in a host fluid of ϵ_2. In the dilute limit, the dipole factor for an isolated particle is known as

$$b = \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}. \tag{1}$$

For a pair of touching particles, one should take into account the effect of multiple images, and hence based on the multiple image method [14] the corresponding dipole factor has the following form:

$$b^* = b\sum_{n=0}^{\infty} (pb)^n \left[\frac{\sinh \alpha}{\sinh(n+1)\alpha}\right]^3, \tag{2}$$

where p is the polarization index with $p=-1$ and $p=2$ representing the transverse field case (T) and the longitudinal field (L), respectively. In Eq. (2), α satisfies the relation

*Email address: jphuang@mpip-mainz.mpg.de
†Email address: kwyu@phy.cuhk.edu.hk

cosh $\alpha = s/d$. Here s denotes the center-to-center separation between the two particles.

Next, consider the ground state of an ER solid, namely, a bct 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 $c = q\ell$ and $a(=b) = \ell q^{-1/2}$ along the z and $x(y)$ axes, respectively. In this case, the uniaxial anisotropic axis is directed along the z axis. As q varies, the volume of the unit cell stays the same, $V_c = \ell^3$. Thus, the degree of anisotropy of the tetragonal lattice is measured by how q deviates from unity.

When one applies an x-directed external electric field $\vec{E}_0$, the dipole moments $\vec{p} = p\hat{x}$, are perpendicular to the uniaxial anisotropic 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:

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

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. (3), 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, w are integers. In addition, x_j and R_j in Eq. (3) are given by, respectively,

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

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

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

$$\alpha_{\mathrm{eff}} = \frac{\alpha}{1 - \alpha\beta_x/V_c}, \quad (4)$$

where α stands for the polarizability of an isolated dipole, and $\beta_x = E_x V_c/p$ is the local field factor which was measured in computer simulations by Martin *et al.* [19,20]. Let us use β_z and β_x ($\equiv\beta_y$) to denote the local-field factors parallel and perpendicular to the uniaxial anisotropic axis, respectively. Accordingly, we have $\beta_z = \beta_x = 4\pi/3$ for the bcc lattice ($q = 1$). In what follows, we set $\beta' = 3\beta/4\pi$. It is worth noting that β' is a function of a single variable, namely, the degree of anisotropy q. Also, β_z' and β_x' satisfy the sum rule $\beta_z' + 2\beta_x' = 3$, and $\beta_z' = \beta_x' = 1$ [at $q = 1$ (bcc)] just represents the isotropic limit.

Based on Eq. (4), it is straightforward to derive the dipole factor for a particle (b') in the lattice under consideration, which admits

$$b' = \frac{b}{1 - bf\beta_x'}, \quad (5)$$

where f stands for the volume fraction of the particles. Thus, the local-field effect arising from all the other particles in the lattice has been included in Eq. (5). In this connection, the effective dielectric constant ϵ_e can be determined by

$$\frac{b}{1 - bf\beta_x'} = b' \equiv \frac{\epsilon_1 - \epsilon_e}{\epsilon_1 + 2\epsilon_e}. \quad (6)$$

That is, we see the particle as one that is embedded in an effective medium with (effective) dielectric constant ϵ_e. It is worth remarking that this ϵ_e includes the detailed structural information of the lattice, as expected.

Next, to put forth the many-body DID model, let us consider a pair of dielectric spherical particles which are both located in the x axis and placed in an effective medium with dielectric constant ϵ_e. Apply a constant electric field $\mathbf{E}_0 = E_0\hat{\mathbf{x}}$ to the particles, which contributes to each particle a dipole moment given by p_{10} and p_{20} ($\equiv p_{10} = \epsilon_e E_0 d^3 b'/8$). Then, the dipole moment p_{10} induces an image dipole p_{11} in particle 2, while p_{11} induces yet another image dipole in particle 1. As a result, multiple images are formed. The same description holds for particle 2 as well. After deriving the sum of the dipole moments inside each particle, we obtain the desired expressions for the corresponding dipole factor

$$b'^* = b'\sum_{n=0}^{\infty}(pb')^n\left[\frac{\sinh\alpha}{\sinh(n+1)\alpha}\right]^3 \quad (7)$$

for the transverse field case ($p = -1$) and the longitudinal field ($p = 2$), respectively. This equation is a nontrivial result indeed because it includes both the multipolar interaction and the local-field effect.

To discuss the interparticle force, we take one step forward to calculate the force between the two dielectric particles. Since we have already obtained the dipole factors b'^*

and b^*, the force can be readily calculated by an energy approach. In doing so, the dipole energy (E_g) of the two particles is determined by the dot product of the electric field and the corresponding dipole moment, and hence the force between them is given by the derivative of the dipole energy with respect to the separation, namely, $-dE_g/ds$. Thus, based on this relation, we obtain the expressions for the interparticle forces, respectively,

$$F^* = -(1/8)\epsilon_2 E_0^2 d^3 b \sum_{n=0}^{\infty} (pb)^n \Phi_n, \tag{8}$$

$$F'^* = -(1/8)\epsilon_{eT} E_0^2 d^3 b' \sum_{n=0}^{\infty} (pb')^n \Phi_n, \tag{9}$$

with

$$\Phi_n = \frac{\sinh\alpha\cosh\alpha\sinh[(n+1)\alpha] - (n+1)\sinh^2\alpha\cosh[(n+1)\alpha]}{(d/3)\sinh^4[(n+1)\alpha]},$$

where F^* indicates the interparticle force between the two particles in the pure host fluid for the transverse field case ($p=-1$) and the longitudinal field ($p=2$), and F'^* the interparticle force between the two particles in the effective medium of dielectric constant ϵ_e. It is shown that both the multipolar interaction and the local-field effect have been taken into account for F'^* [Eq. (9)]. In Eqs. (8) and (9), setting n to 1 yields the corresponding point-dipole force, namely, F and F'. The force expressions are reasonable, as the $n=0$ term of Eqs. (8) and (9) vanishes while the $n=1$ term gives the correct point-dipole force.

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 b'^*/b^*, which indicates the correction of the local-field effect on the multiple image effect, as well as on the ratio of b^* or b'^* to b. Finally, we shall study the q effect on the interparticle force which is normalized by the corresponding point-dipole force. Without loss of generality, we set $\epsilon_1 = 30\epsilon_0$, $\epsilon_2 = 2.8\epsilon_0$, and $s/d = 1.1$ for numerical calculations, where ϵ_0 denotes the dielectric constant of free space.

Figure 1 shows the dependence of the local-field factor on the degree of anisotropy q. It is evident that β_x' (or β_z') is caused to increase (or decrease) as q increases. A plateau is shown at $\beta_x' = \beta_z' = 1$, which actually includes the transformations ranging from the bcc ($q=1$) lattice to the fcc ($q=2^{1/3}$). Accordingly, a similar plateau occurs in all other figures (see Figs. 2–4).

In Fig. 2, we investigate the ratio of $b_T'^*/b_T^*$ and $b_L'^*/b_L^*$ as a function of q. It is found that the local-field effect is dominant over the multipolar interactions since all the ratios ($b_T'^*/b_T^*$ and $b_L'^*/b_L^*$) are larger than unity. More precisely, for both the transverse field case [Fig. 2(a)] and the longitudinal field [Fig. 2(b)], the local-field effect can be large, especially at high volume fractions and/or large q. In this case, the correction due to the local field cannot be neglected. Moreover, as the volume fraction f is given $b_L'^*/b_L^*$ (longitudinal field) is larger than $b_T'^*/b_T^*$ (transverse field).

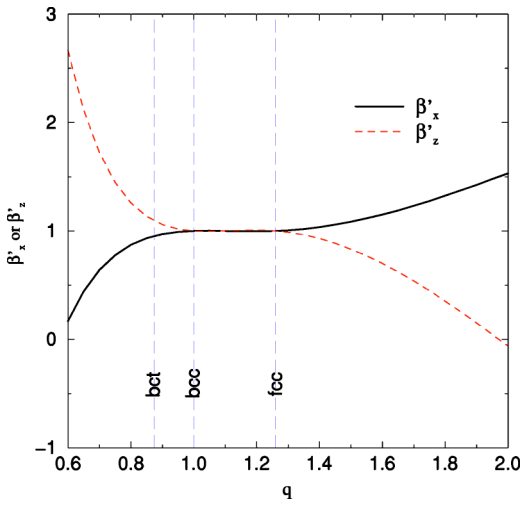

FIG. 1. (Color online) Dependence of the (dimensionless) local field factor β_x' and β_z' on the (dimensionless) degree of anisotropy q. The bct ($q=0.873\,58$, i.e., ground state), bcc ($q=1.0$), and fcc ($q=2^{1/3}$) lattices are also shown (long-dashed lines).

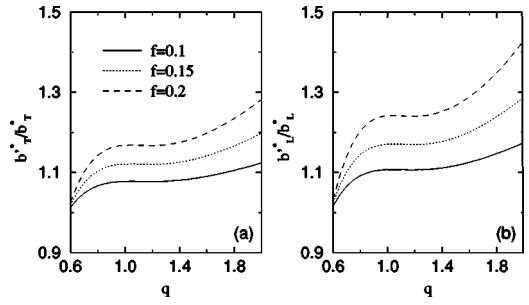

FIG. 2. Ratio of dipole factor b'^* to b^* versus q for different volume fractions $f=0.1$, 0.15, and 0.2. (a) Transverse field cases; (b) longitudinal field cases. Parameters $\epsilon_1 = 30\epsilon_0$, $\epsilon_2 = 2.8\epsilon_0$, and $s/d = 1.1$.

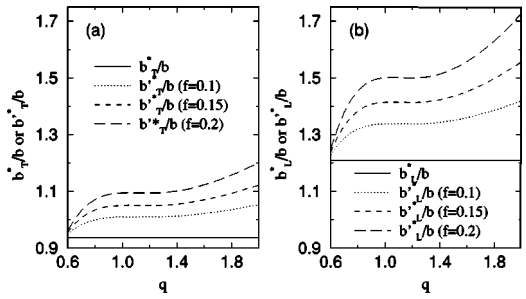

FIG. 3. Same as Fig. 2, but for the ratio of b^* or b'^* to b.

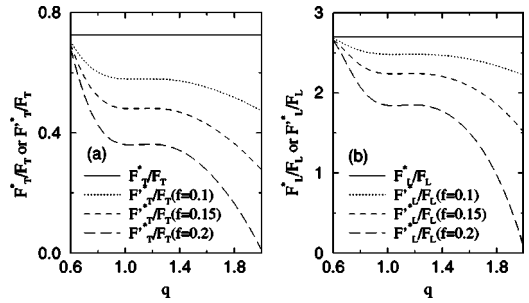

FIG. 4. Same as Fig. 2, but for the ratio of interparticle force F^* or F'^* to point-dipole force F.

We plot the ratio of b^*/b and b'^*/b in Fig. 3. As expected, both the local-field effect and the multipolar interaction can be dominant over the dipolar interaction, and hence they should be taken into account. Again, such effects are stronger for the longitudinal field case than for the transverse field.

Figure 4 displays the interparticle force which is normalized by the corresponding point-dipole force. From this figure, it is found that the multipolar interaction can indeed be dominant over the dipolar interaction, while the local-field effect may yield an important correction. Moreover, the correction due to the local-field effect can be very large, especially at large volume fractions and/or large q. Nevertheless, for small volume fractions, the correction due to the local field is very small. Furthermore, the reduction of the magnitude of the effective interaction between the two particles is shown for the transverse field case [Fig. 4]. In contrast, the increase in the effective interaction is found for the longitudinal field case.

As a matter of fact, the q dependence of the dipole factor [Eq. (5)] can qualitatively be understood with the aid of the spectral representation theory [21]. In doing so, let us denote $\varrho=(1-\epsilon_1/\epsilon_2)^{-1}$, and then Eq. (5) is reexpressed as

$$b' = \frac{\vartheta_1}{\varrho - \theta_1}, \tag{10}$$

where the residue $\vartheta_1=-1/3$ and the pole $\theta_1=(1-f\beta_x')/3$. Using the numerical values of the present model calculation, we find $b'=(-1/3)/[(-0.103)-(1-f\beta_x')/3]$. In this case, increasing q leads to increasing β_x (see Fig. 1), and hence b' is caused to increase, as shown in Figs. 2 and 3.

In the above calculations, we calculated the multiple image effect on the particle interaction by taking particles which are located along the x axis as an example. On the other hand, if we choose the particles located on the z axis, the opposite effect should be found accordingly (no figures shown here) due to the fact that β_z' is caused to decrease by increasing q (see Fig. 1) in view of the sum rule for β_z' and β_x', namely, $\beta_z'+2\beta_x'=3$.

IV. DISCUSSION AND CONCLUSION

Here a few comments are in order. We put forth a many-body DID model for the ER solid, in an attempt to investigate both local-field effects and multipolar interactions. The Ewald-Kornfeld method for dipolar systems has provided an accurate means of assessing the local-field effect. The claim that the multipolar interaction can be dominant over the dipolar interaction for touching particles has been confirmed in a rigorous manner.

The local-field effects on the interparticle force are always present in a many-particle system such as ER fluids. However, as the particles approach and finally touch, the multipolar interaction becomes more prominent. In fact, when the center-to-center separation between the two interacting particles is larger than $2d$, the effect of multipole interaction can be small enough to be neglected [22]. In this case, the local-field effect should play a role, as expected.

Klingenberg *et al.* have obtained the numerical results for interacting particles by solving the empirical force expression [10,23], in which there are three force functions being determined from the numerical solution of Laplace's equation. Fortunately, we have shown that the three force functions could relate to our multiple image moments [24]. Further, we [24] have compared the results of our multiple image theory with the numerical results provided by Klingenberg *et al.* [10,23]. Regarding the comparison, we would like to refer to Fig. 1 of Ref. [24]. Based on the comparison, we would say that reasonable agreements have been obtained, and hence our multiple image expressions should be expected to give reliable results.

In fact, we presented an effective medium theory (EMT) for considering the local-field effect on the electrorotation and dielectric dispersion spectra of colloidal particles or biological cells in a previous paper [25]. However, we were unable to study the detailed structural information via the EMT. In this connection, it is also of interest to use the many-body DID model to discuss the electrokinetics of colloidal particles or biological cells [22,26].

The present model is expected to be used in various studies of the behavior of ER solids, e.g., in a computer simulation [24]. By including the local-field effect, the DID model can be used with higher accuracy. In addition, it is also of value to extend the present work to polydisperse ER solids [27], in which the permittivities of particles can have a distribution.

To sum up, we have developed a many-body DID model for the ER solid, the lattice structure of which can be changed due to the application of external fields, in an attempt to take into account both local-field effects and multipolar interactions. The effective dielectric constant for the

ER solid has been derived, based on the Ewald-Kornfeld formulation. It has been shown that the multipolar interaction can indeed be dominant over the dipolar interaction, while the local-field effect may yield an important correction. Also, the results are well understood with the aid of the spectral representation theory.

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