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INTERPARTICLE FORCE IN NONLINEAR ELECTRORHEOLOGICAL FLUIDS

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The applied electric field used in most electrorheological (ER) experiments is usually quite high, and nonlinear ER effects have been measured recently. In this work, a self-consistent formalism has been employed to compute the interparticle force for a nonlinear ER fluid in an attempt to investigate the effect of a nonlinear characteristics on the particle interactions.

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1. Introduction

The prediction of the strength of the electrorheological (ER) effect is the main concern in a theoretical investigation of ER fluids. The ER effect originates from the induced interaction between the polarized particles in an ER fluid. As the mismatch in material parameters (either conductivities or dielectric constants) is responsible for the ER effects, previous theoretical studies have taken the point-dipole approximation,[1,2] which is now believed to be over-simplified. Since the many-body and multipole interactions have been ignored in these studies, the predicted yield stress has been off by an order in magnitude. The gap between theory and experiment further widens rapidly because the technological applications of ER fluids have stimulated many experiments which measure directly the interactions between particles of various materials under different experimental conditions. It is now known that for crystalline particles, not only force but also torque are exerted on the particles due to crystalline anisotropy. It is also known that particles coated with different materials have a significant impact on the ER response.[3,4]

Because of the inadequacy of the point-dipole approximation, substantial effort has been made to sort out more accurate models. Klingenberg and coworkers proposed an empirical force expression for the interaction between isolated pairs of equal spheres from the numerical solution of Laplace's equation.[5] Davis used the finite-element method, which is proved to be effective.[6] Clercx and Bossis constructed a fully multipolar treatment to account for the polarizability of spheres up to 1,000 multipolar orders.[7] Recently, Yu and coworkers developed an integral equation method which avoids the match of complicated boundary conditions on each interface of the particles and is thus applicable to nonspherical particles and multimedia.[8] Substantial improvements have been achieved in reducing the difference between theory and experiment.

On the other hand, the applied electric field used in most ER experiments is usually quite high, and important data on nonlinear ER effects induced by a strong electric field were recently unveiled experimentally by Klingenberg and coworkers.[9] However, the effect of a nonlinear characteristics on the particle interactions remains unknown. A theoretical explanation of these data is urgently required, which is a key aspect in this investigation. In this work, the effect of a nonlinear characteristics on the particle interactions is investigated via a self-consistent formalism,[10] in which the recently established (linear) multiple image results[11,12] will be converted to nonlinear ones to compute the interparticle force for a nonlinear ER fluid.

2. Multiple Image Dipole for a Pair of Dielectric Spheres

We first consider a standard textbook problem[13] in which a point dipole p is placed at a distance r from the center of a perfectly conducting sphere of radius a. The orientation of the dipole is perpendicular to the line joining the dipole and the center of the sphere. The electric field vanishes inside the conductor while the electric potential outside the sphere can be found by using the method of image. We put an image dipole p' inside the sphere at a distance r' from the center; the image dipole is given by $p' = -p(a/r)^3$, and $r' = a^2/r$. If the orientation of the point dipole is parallel to the axis, then $p' = 2p(a/r)^3$.

We next consider a pair of perfectly conducting spheres, of equal radius a, separated by a distance r. The spheres are placed in a host medium of dielectric constant ϵ_2. Assume that the two conductors are electrically neutral, and a constant electric field $\mathbf{E}_0 = E_0\hat{z}$ is applied to the spheres. Induced surface charge will contribute to each conductor a dipole moment given by $p_0 = \epsilon_2 E_0 a^3$. The dipole moment $p_0^{(1)}$ induces an image dipole $p_1^{(1)}$ in sphere 2, while $p_1^{(1)}$ induces yet another image dipole in sphere 1. As a result, multiple images are formed. Similarly, $p_0^{(2)}$ induces an image $p_1^{(2)}$ inside sphere 1, and hence another infinite series of image dipoles are formed. The multiple images obey a set of difference equations, which can be solved exactly.[11]

We are now in a position to generalize the above results to a pair of dielectric spheres of dielectric constant ϵ_1. Upon the application of $\mathbf{E}_0$, the induced dipole moment inside the spheres is given by:

$$p_0 = \epsilon_m E_0 b a^3 \,, \tag{1}$$

where b is the dipolar factor and is given by:

$$b = \frac{\epsilon_1 - \epsilon_m}{\epsilon_1 + 2\epsilon_m} \,. \tag{2}$$

For a point dipole placed in front of a dielectric sphere, the generalization reads $p' = -\tau p(a/r)^3$ and $p' = 2\tau p(a/r)^3$ for transverse and longitudinal fields respectively. The factor τ is known as dielectric contrast and is given by:

$$\tau = \frac{\epsilon_1 - \epsilon_m}{\epsilon_1 + \epsilon_m} . \tag{3}$$

By using the idea of multiple images, we can deduce the total dipole moment of each sphere (normalized to p_0), for a transverse field:

$$\frac{p_\mathrm{T}}{p_0} = (a\sinh\alpha)^3 \sum_{n=1}^{\infty} \left[\frac{(-\tau)^{2n-2}}{(a\sinh n\alpha + a\sinh(n-1)\alpha)^3} + \frac{(-\tau)^{2n-1}}{(r\sinh n\alpha)^3} \right] . \tag{4}$$

The subscript T denotes that the applied electric field is a transverse field. The parameter α is given by

$$\cosh\alpha = \frac{r^2}{2a^2} - 1 . \tag{5}$$

Similarly, for a longitudinal field:

$$\frac{p_\mathrm{L}}{p_0} = (a\sinh\alpha)^3 \sum_{n=1}^{\infty} \left[\frac{(2\tau)^{2n-2}}{(a\sinh n\alpha + a\sinh(n-1)\alpha)^3} + \frac{(2\tau)^{2n-1}}{(r\sinh n\alpha)^3} \right] . \tag{6}$$

We should remark that the present generalization is only approximate because there is a more complicated image method for a dielectric sphere.[12] However, in the limit $\tau \to 1$, the above expressions reduce to the perfectly conducting sphere results. We expect that this approximation to be good at high contrast, i.e. $\tau \to 1$. We have checked the validity by comparing these analytic expressions with the numerical solution of the integral equation method.[8] The transverse force F_T between the spheres is given by[14]

$$F_\mathrm{T} = E_0 \frac{\partial p_\mathrm{T}}{\partial r} . \tag{7}$$

Similarly, for a longitudinal field:

$$F_\mathrm{L} = E_0 \frac{\partial p_\mathrm{L}}{\partial r} . \tag{8}$$

3. Self-Consistent Formalism for Nonlinear ER Effect

We are in a position to examine the impact of a nonlinear characteristics on the ER effect. We concentrate on the case that only the suspending spheres have a nonlinear dielectric constant, with the host medium being linear. Naively, the nonlinear characteristics gives rise to a field-dependent dielectric coefficient $\widetilde{\epsilon}_1 = \epsilon_1 + \chi_1 \langle E_1^2 \rangle$, which depends on the average electric field inside the spheres,[4] where χ_1 is the nonlinear coefficient of the spheres. In other words, the electric displacement-electric field relation inside the spheres is given by

$$\mathbf{D}_1 = \epsilon_1 \mathbf{E}_1 + \chi_1 \langle E_1^2 \rangle \mathbf{E}_1 = \widetilde{\epsilon}_1 \mathbf{E}_1 . \tag{9}$$

This constitutes an approximation: the local field inside the spheres is assumed to be uniform and the assumption is called the decoupling approximation.[10] It has been shown that such an approximation yields a lower bound for the accurate result.[10] It is clear from the above consideration, the effect of a nonlinear characteristics enters the force expression in two places: from the dipolar factor b and from the dielectric contrast τ appearing in the infinite series [Eqs. (4) and (6)]. The latter effect gives rise to a deviation from the nonlinear point-dipole results, as we shall see below.

To calculate the force between a pair of separated spheres, we replace ϵ_1 by $\widetilde{\epsilon}_1$ in Eqs. (7) and (8), which means that we have to calculate the local field self-consistently inside the spheres. The electric field inside the spheres can be conveniently calculated by considering the effective dielectric constant of a composite in which the spheres are embedded in a host medium of much larger volume V. For a two-component composite, the effective dielectric constant ϵ_e is given by:

$$\epsilon_e = \frac{1}{E_0 V}\int_V \epsilon(\mathbf{r})|\mathbf{E}(\mathbf{r})|^2 dV = \frac{f\epsilon_1}{E_0^2}\langle E_1^2\rangle + \frac{(1-f)\epsilon_2}{E_0^2}\langle E_2^2\rangle\,, \tag{10}$$

where f is the (infinitesimal) volume fraction of component 1. For a pair of spheres inside a transverse field, the effective dielectric constant can be expressed as:

$$\epsilon_e = \epsilon_m + 3f\epsilon_m\left(\frac{2bp_{\rm T}}{p_0}\right). \tag{11}$$

The electric field inside the spheres can be calculated by using Eq. (11):

$$\langle E_1^2\rangle = \frac{1}{f}E_0^2\frac{\partial\epsilon_e}{\partial\epsilon_1} = 6E_0^2\epsilon_m\left[b\frac{\partial}{\partial\epsilon_1}\left(\frac{p_{\rm T}}{p_0}\right) + \left(\frac{p_{\rm T}}{p_0}\right)\frac{\partial b}{\partial\epsilon_1}\right], \tag{12}$$

where $p_{\rm T}/p_0$ is given by Eq. (4). For a nonlinear characteristics Eq. (9), we replace ϵ_1 by $\widetilde{\epsilon}_1$ and then solve Eq. (12) self-consistently.[10] The local field inside the spheres and the force between the spheres can be determined. The force for the longitudinal field case can be calculated in essentially the same way.

4. Results and Discussion

If we ignore the mutual polarization effect between the spheres, the force between point dipoles can be derived by noting that the nonlinear dipole moment varies as $\tilde{p} = \beta E_0 + \gamma E_0^3 + \cdots$, where β and γ are constants. Since the interparticle force F varies as $\tilde{p}^2$, we find that $F = (\beta E_0)^2(1 + cE_0^2 + \cdots)$, where c is a constant. A plot of F/E_0^2 against E_0^2 would yield a straight line.

In order to demonstrate the effect of the nonlinearity on the force between the spheres, we plot $F_{\rm T}/E_0^2$ versus E_0^2 (Fig. 1) and $F_{\rm L}/E_0^2$ versus E_0^2 (Fig. 2). In Fig. 1, the force for a transverse field is shown, the nonlinear coefficients are given by $\chi_1 = 0.01$ and 0.1, and the separation parameters are given by $\sigma = r/2a = 1.1$ and 1.5. In both case, the dielectric constants of the sphere and host medium are chosen to be $\epsilon_1 = 2$ and $\epsilon_m = 1$ respectively. The dashed lines represent the point dipole (PD) case and the solid line indicate the full multipole calculation or the multiple

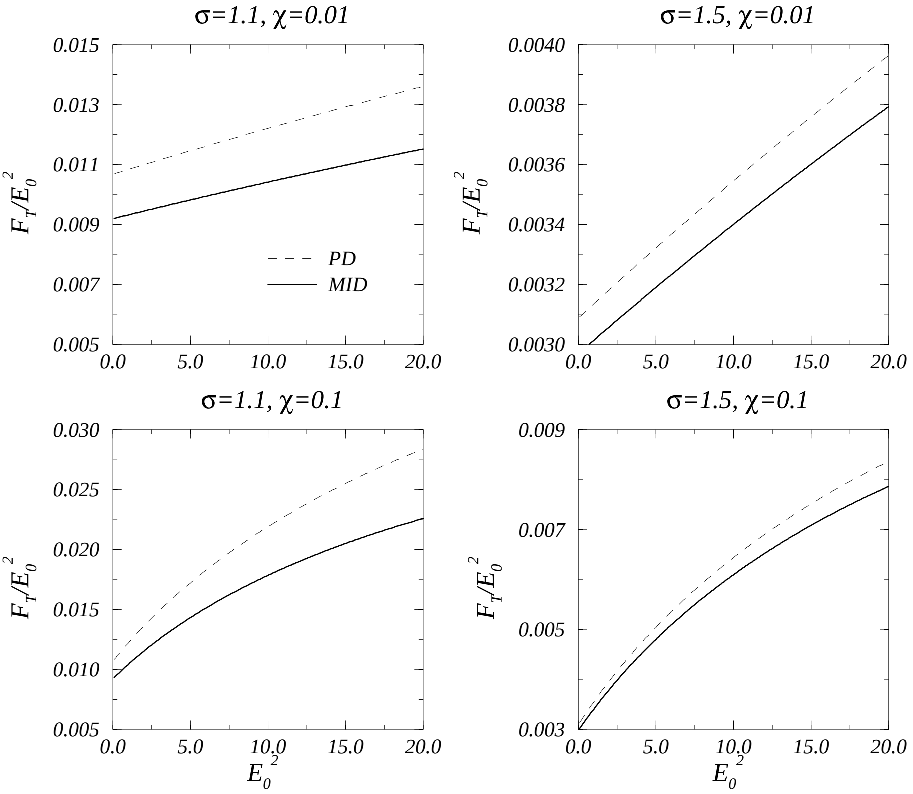

Fig. 1. Normalized force F_{T}/E_0^2 plotted against E_0^2 for a transverse field for two reduced separations $\sigma = 1.1$ and 1.5. The magnitude of the MID force is generally smaller than that of the PD force due to multipole interactions. For a weak nonlinearity ($\chi_1 = 0.01$), F_{T}/E_0^2 varies with E_0^2 almost linearly for both the PD and MID case. For a larger nonlinear coefficient ($\chi_1 = 0.1$), a significant deviation from a linear relationship occurs.

induced dipole (MID) case. For the transverse (longitudinal) case, the magnitude of the MID force is generally smaller (greater) than that of the PD force due to multipole interactions.

We will concentrate on the case for a transverse field; the analysis for the longitudinal case is similar. Let us first consider the case for a weak nonlinearity ($\chi_1 = 0.01$). In Fig. 1, we found that, for both the PD and MID case, F_{T}/E_0^2 varies with E_0^2 almost linearly, in accord with the nonlinear PD results. For the MID case, although the total dipole moment [as expressed in Eq. (4)] differs from the bare point dipole moment p_0, we can still expand F_{T}/E_0^2 to the first order of $\chi_1\langle E_1^2\rangle$. This explains the linear behavior of F_{T}/E_0^2 versus E_0^2. The linear behavior prevails even when the separation is small ($\sigma = 1.1$) and we recover the linear results when $E_0^2 \to 0$.

Next we extend our discussion to the case of a larger nonlinear coefficient ($\chi_1 = 0.1$). It is evident that a significant deviation from a linear relationship occurs.

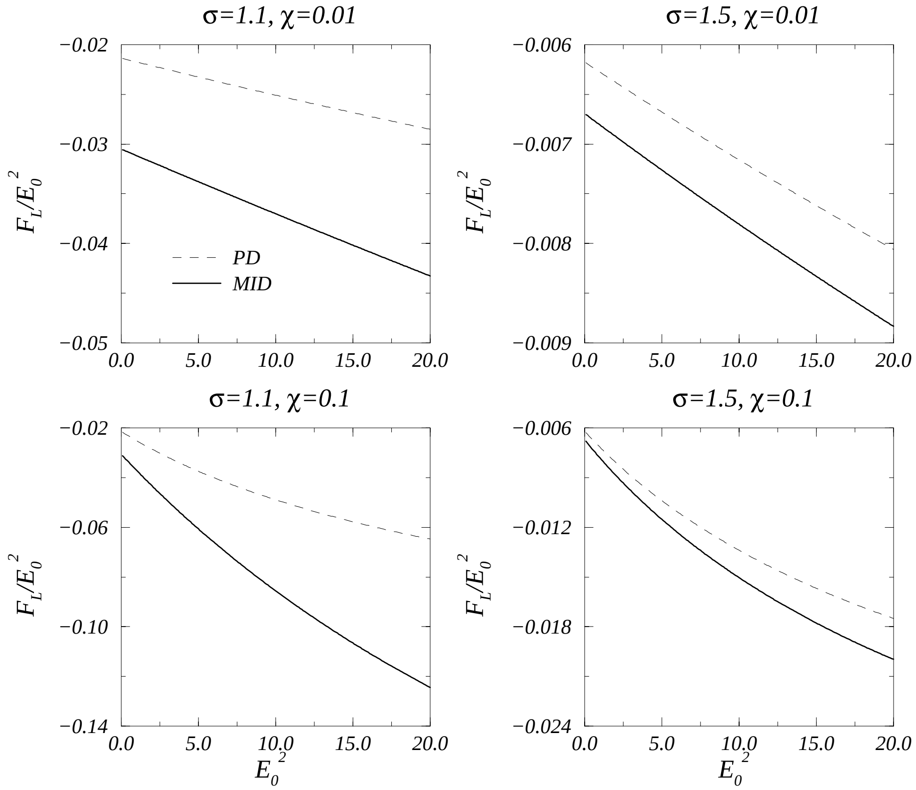

Fig. 2. Similar to Fig. 1 but for a longitudinal field. Here the magnitude of the MID force is generally greater than that of the PD force due to multipole interactions.

The slopes decrease as E_0^2 increases, which indicates a weaker dependence on E_0^2. The deviation from a linear relationship is attributed to when χ_1 gets larger, $F_{\rm T}/E_0^2$ will also depend on the higher orders of $\chi_1\langle E_1^2\rangle$. Here a few comments on our results are in order. As pointed out by Felici and coworkers,[15] when the applied field is sufficiently strong, The attractive force between two touching spheres will have a $F \sim E_0$ dependence while our results (Figs. 1 and 2) show a similar tendency. The major difference here is that while the previous results[15] were based on a conduction model for two touching spheres, we have extended the considerations to two spheres separated by an arbitrary distance. As we have included all multipole interactions in our self-consistent calculations, the results will be useful in computer simulation of ER fluids at an intense applied field. To this end, the implications of our results on the shear modulus deserves further exploration.

5. Conclusion

In summary, the effect of a nonlinear characteristics on the particle interactions is investigated via a self-consistent formalism, in which the previously established multiple image results have been converted to nonlinear ones to compute the interparticle force for a nonlinear ER fluid.

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