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Force between two spherical inclusions in a nonlinear host medium

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In an attempt to investigate the effect of nonlinear characteristics on particle interactions, a self-consistent mean-field theory in combination with a multiple image method has been employed to compute the interparticle force. Taking the nonlinearity of the host medium into account, the interparticle force exhibits a nonmonotonic behavior as the applied electric field is increased. We show that the interparticle force increases initially at low fields, goes through a maximum at an optimal electric field, then decreases with increasing field, and vanishes at a critical electric field at which the effective dielectric contrast between the host and the inclusions becomes zero. The influence of a larger volume fraction of inclusions on the interparticle force is also investigated.

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Electrorheological (ER) fluids consist of highly polarizable particles in a nearly insulating fluid. Upon the application of electric fields, the apparent viscosity of ER fluids can be changed by several orders of magnitude, due to the formation of chains of particles across the electrodes in the direction of the applied field. The ER effect originates from the mutual interaction between the polarized particles. As the mismatch in material parameters is responsible for the ER effects, previous theoretical studies have taken the point-dipole approximation [1,2], which is now believed to be oversimplified. Since many-body and multipole interactions have been ignored in these studies, the predicted ER effect was off by an order in magnitude. Moreover, the technological applications of ER fluids have stimulated many experiments which directly measure the interactions between particles of various materials under different experimental conditions.

Because of the inadequacy of the point-dipole approximation, substantial theoretical effort has been made to sort out more accurate models. Klingenberg *et al.* proposed an empirical force expression for the interaction between isolated pairs of spheres from the numerical solution of Laplace's equation [3]. Davis used the finite-element method, which proved to be effective [4]. Clercx and Bossis constructed a fully multipolar treatment to account for the polarizability of spheres up to 1000 multipolar orders [5]. Recently, Yu *et al.* 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 [6].

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 reported by Klingenberg [7]. However, the effect of a nonlinear characteristics on the particle interactions remains less well known. Felici *et al.* [8] pointed out that when the applied field is sufficiently strong, the attractive force between two touching spheres will have a quasilinear dependence. However, as the previous results [8] were based on a conduction model for two touching spheres, we must extend the considerations to two spheres separated by an arbitrary distance in order that the results can be useful in computer simulation of ER fluids at an intense applied field.

In this work, the effect of a nonlinear characteristics on the particle interactions is investigated via a self-consistent formalism [9], in which the recently established (linear) multiple image results [10,11] will be converted to nonlinear ones to compute the interparticle force for a nonlinear ER fluid.

We first consider a standard textbook problem [12], 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_m. Assume that the two conductors are electrically neutral, and a constant electric field $\mathbf{E}_0 = E_0\hat{\mathbf{z}}$ is applied to the spheres. Induced surface charge will contribute to each conductor a dipole moment given by $p_0 = \epsilon_m E_0 a^3$. 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 [10].

We are now in a position to generalize the above results to a pair of dielectric spheres of dielectric constant ϵ_p. 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

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

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

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

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

$$\frac{p_L}{p_0}=(a\sinh\alpha)^3\sum_{n=0}^{\infty}\left[\frac{(2\tau)^{2n}}{(a\sinh(n+1)\alpha+a\sinh n\alpha)^3}+\frac{(2\tau)^{2n+1}}{(r\sinh(n+1)\alpha)^3}\right]. \tag{4}$$

The subscript L denotes that the applied electric field is a longitudinal field. The parameter α satisfies

$$\cosh\alpha=2\sigma^2-1, \tag{5}$$

where $\sigma\equiv r/2a$ is the reduced separation parameter.

Similarly, for a transverse field,

$$\frac{p_T}{p_0}=(a\sinh\alpha)^3\sum_{n=0}^{\infty}\left[\frac{(\tau)^{2n}}{(a\sinh(n+1)\alpha+a\sinh n\alpha)^3}-\frac{\tau^{2n+1}}{(r\sinh(n+1)\alpha)^3}\right]. \tag{6}$$

We should remark that the present generalization is only approximate because there is a more complicated image method for a dielectric sphere [11]. 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; reasonable agreement is found [6]. The longitudinal (transverse) force $F_{L(T)}$ between the spheres is given by [13]

$$F_{L(T)}=E_0\frac{\partial p_{L(T)}}{\partial r}. \tag{7}$$

In order to describe the nonlinear effect in ER fluids, we concentrate on a system consisting of spherical inclusions of the volume fraction f and dielectric constant ϵ_p suspended in a volume fraction $1-f$ of the host medium. Here we assume that the host medium has a third-order nonlinear electric displacement ($\mathbf{D}$)–electric field ($\mathbf{E}$) relation of the form $\mathbf{D_m}=\epsilon_m\mathbf{E}_m+\chi_m|\mathbf{E}_m|^2\mathbf{E}_m$, where ϵ_m and χ_m are the linear dielectric constant and the third-order nonlinear coefficient, respectively. This kind of nonlinearity is the lowest-order nonlinearity appearing in a material with inversion symmetry [14].

Generally speaking, the electric field in the host medium varies in space and cannot be solved exactly. So we resort to the mean field approximation. The mean field theory amounts to approximating the nonlinear host medium as a component with the property [9]

$$\tilde{\epsilon}_m=\epsilon_m+\chi_m\langle|\mathbf{E}_m|^2\rangle, \tag{8}$$

where $\langle\cdots\rangle$ denotes the average of the local field taken over the volume occupied by the host medium. From the above consideration, it is evident that the nonlinearity influences the interparticle force through two factors, namely, the dipolar factor b and the dielectric contrast τ appearing in the infinite series.

For a pair of spherical particles with a small volume fraction f inside a longitudinal (transverse) field, the effective dielectric constant ϵ_e can be calculated using the dilute limit approximation for a three-dimensional composite,

$$\epsilon_e=\tilde{\epsilon}_m+3f\tilde{\epsilon}_m\left(\frac{2bp_{L(T)}}{p_0}\right), \tag{9}$$

where b and $p_{L(T)}/p_0$ are given by Eqs. (2), (4), and (6), with ϵ_m being replaced by $\tilde{\epsilon}_m$.

Then we can determine the average local field square $\langle|\mathbf{E}_m|^2\rangle$ self-consistently as follows:

$$\langle|\mathbf{E}_m|^2\rangle=\frac{1}{1-f}E_0^2\frac{\partial\epsilon_e}{\partial\tilde{\epsilon}_m}. \tag{10}$$

The equality in Eq. (10) results from an established formula in a random composite giving ϵ_e in terms of the local field,

$$\epsilon_e=\frac{1}{E_0^2V}\int_V\epsilon(\mathbf{r})|\mathbf{E}(\mathbf{r})|^2dV=\frac{f\epsilon_p}{E_0^2}\langle|\mathbf{E}_p|^2\rangle+\frac{(1-f)\tilde{\epsilon}_m}{E_0^2}\langle|\mathbf{E}_m|^2\rangle, \tag{11}$$

where $\epsilon(\mathbf{r})$ takes on the values ϵ_p (or $\tilde{\epsilon}_m$) for $\mathbf{r}$ in inclusions (or host medium), while V is the volume of the whole composite system.

Thus we have formulated the self-consistent mean field theory with Eqs. (9) and (10), and the multiple image method with Eqs. (4)–(7), to obtain the interparticle force in nonlinear ER fluids for the longitudinal and transverse field cases. In order to demonstrate the influence of the nonlinearity on the interparticle force, we plot the normalized force for a longitudinal field F_L/E_0^2 ($\mathrm{N\,m^2\,V^{-2}}$) as a function of E_0^2 ($\mathrm{V^2\,m^{-2}}$) in Fig. 1 for $\epsilon_p=2$ and in Fig. 2 for $\epsilon_p=10$. Without loss of generality, the linear dielectric constant of the host medium is taken to be $\epsilon_m=1$, because one can always define a relative permittivity ϵ_p/ϵ_m. Numerical results have also been calculated for the transverse field case, and will be reported elsewhere.

First of all, we compare our results of the fully multipole case with those of the point-dipole (PD) model, for which only the leading term is retained in the infinite series. It is known that the PD approximation is expected to be very good for large reduced separation such as $\sigma>1.5$. For our chosen values $\sigma=1.1$ and 1.5, the results agree with each other in the vicinity of a critical field E_c still, which is characterized by $\epsilon_p=\epsilon_m+\chi_m\langle|\mathbf{E}_m|^2\rangle$, but exhibit a large devia-

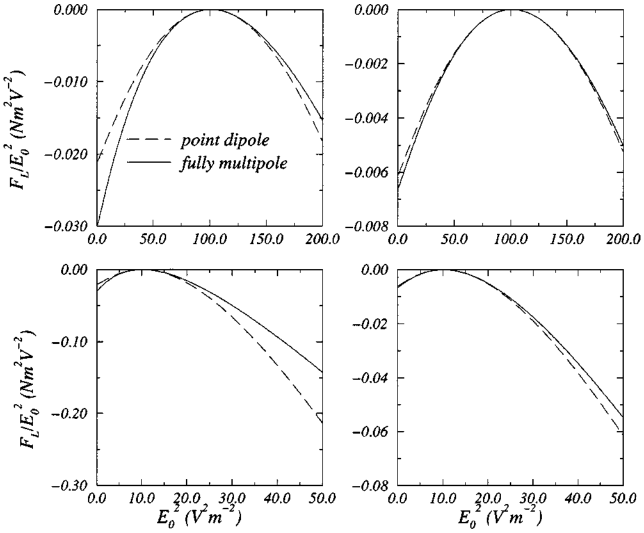

FIG. 1. Normalized force F_L/E_0^2 $(\mathrm{N\,m^2\,V^{-2}})$ plotted against E_0^2 $(\mathrm{V^2\,m^{-2}})$ for $\epsilon_p=2$. Other physical parameters are chosen as $\sigma=1.1$ and $\chi_m=0.01$ $\mathrm{m^2\,V^{-2}}$ for the upper left panel, $\sigma=1.5$ and $\chi_m=0.01$ $\mathrm{m^2\,V^{-2}}$ for the upper right panel, $\sigma=1.1$ and $\chi_m=0.1$ $\mathrm{m^2\,V^{-2}}$ for the lower left panel, and $\sigma=1.5$, and $\chi_m=0.1$ $\mathrm{m^2\,V^{-2}}$ for the lower right panel.

tion at applied fields much smaller or larger than E_c. Such a deviation becomes even more significant when the separation parameter $\sigma\equiv r/2a$ is decreased from $\sigma=1.5$ to 1.1. This is reasonable, because the PD approximation is expected to break down, and multipole interactions will become more important, when the separation is small or when the dielectric contrast is large even in the dilute limit. This demonstrates that the PD approximation is only valid for a small dielectric contrast, and multipole interaction must be considered for a large contrast. This is in agreement with the conclusion of Davis [4].

Next we examine the behavior of normalized force with E_0^2. For a small $\epsilon_p=2$ (Fig. 1), the magnitude of the interaction decreases to zero and then increases as the field is increased. However, for a large $\epsilon_p=10$ (Fig. 2), a more complex behavior is observed, i.e., the magnitude of the force

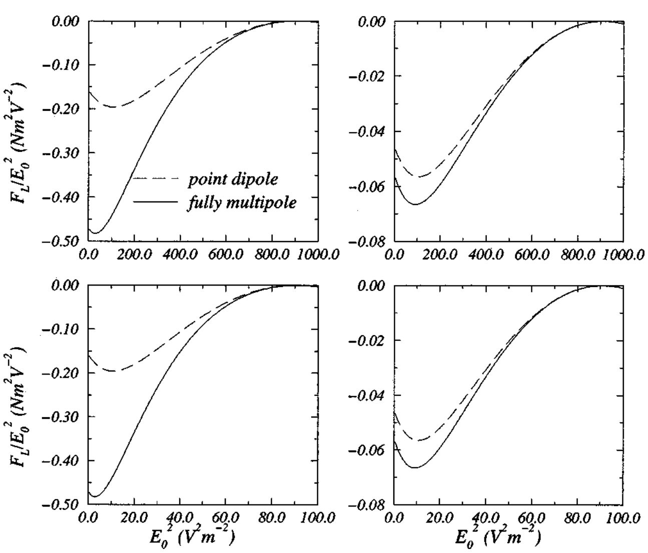

FIG. 2. Same as Fig. 1, but with $\epsilon_p=10$.

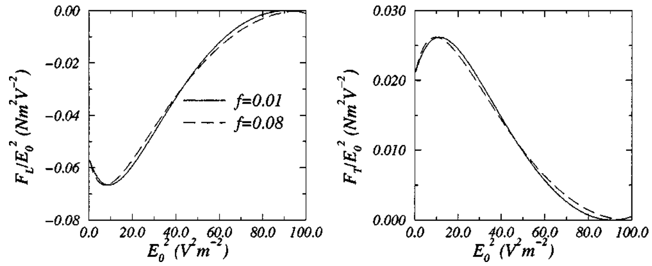

FIG. 3. Normalized force for both the longitudinal and transverse field cases plotted against E_0^2 for two volume fractions $f=0.01$ (solid line) and 0.08 (long dashed line). Other physical parameters are chosen to be $\epsilon_p=10$, $\sigma=1.5$, and $\chi_m=0.1$ $\mathrm{m^2\,V^{-2}}$.

increases at first, reaches a maximum at an optimal field E_{op}, and then decreases to zero at the critical field E_c. Beyond this point, a further increase in E_0 results in an overall increase of the interparticle force (it is believed this region is unstable for ER fluids). This nonmonotonic behavior admits a qualitative explanation. As we know, the interparticle force is directly proportional to $\tilde{\epsilon}_m$ and to the square of the dielectric contrast τ. As the electric field is turned on, $\tilde{\epsilon}_m$ increases, and so does F_L/E_0^2; however, this increase is then offset by the decrease in τ^2. For a small ϵ_p, the fast decrease of τ^2 leads to the overall decrease of F_L/E_0^2. Such a trend continues until the critical field is reached. A subsequent increase in the field will result in a large $\tilde{\epsilon}_m$ and τ^2, and F_L/E_0^2 will increase. For a large ϵ_p, the competition between τ^2 and $\tilde{\epsilon}_m$ yields a maximum at E_{op}. A similar behavior was reported in Ref. [15], but for linear ER fluids. We also find that the larger the nonlinear coefficient we choose, the smaller the critical field and the optimal field become. Most of the theoretical and experimental works predicted that F_L/E_0^2 is a monotonically increasing function of E_0^2, as these work only concentrated on the range that $\epsilon_p/\tilde{\epsilon}_m\gg 1$, although the applied field is relatively high [8]. For realistic experimental systems, we should choose a large nonlinear coefficient for the dielectric host medium, so as to operate at a small electric field far away from the breakdown field. On the other hand, a large ϵ_m is needed to obtain a high particle–liquid dielectric contrast, and hence a large ER effect.

Moreover, we investigate the influence of volume fraction f on the interparticle force, as shown in Fig. 3. For a large dielectric contrast, on which previous works mainly concentrated, it is observed that $F_{L(T)}/E_0^2$ increases slightly with f. Such a conclusion is in qualitative agreement with experimental observations [16,17]. However, in the range of large field (corresponding to small dielectric contrast), $F_{L(T)}/E_0^2$ can decrease with f. We also find that the optimal field becomes small and the critical field large for large volume fractions. To our knowledge, these behaviors have not been reported previously, as previous work concentrated only on the range of large contrasts. We explain that these phenomena result from a strong dependence of $\langle|\mathbf{E}_m|^2\rangle$ on the volume fraction f.

We would like to add a few comments here. We have considered a nonlinear host medium in this work. In fact, one can consider ER fluids of nonlinear inclusions of, say, ferroelectric materials. In this case, because of a positive nonlinear coefficient, F/E_0^2 will be a monotonic increasing function of E_0^2. Certainly, one can also consider the case when both the inclusions and the host medium are nonlinear and more complex behaviors can occur.

We considered particles of identical size in this work. Real ER fluids must be polydisperse in nature: the suspending particle can have various sizes or different permittivities. Recent work reported that a size distribution can have a nontrivial impact on the ER effects [18]. We have recently shown that the point-dipole approximation becomes even worse when the particle sizes differ too much [13]. It is instructive to extend the consideration to two spheres of unequal sizes.

So far we have considered ER fluids of isotropic materials with scalar dielectric constants. For crystalline particles with tensorial dielectric properties in an external electric field, not only force but also torque will be exerted on the particles due to crystalline anisotropy [19]. The extension of the multiple image method to anisotropic media seems very difficult. We may adopt a first-principles approach, e.g., the integral equation approach [6]. In this regard, in the case of a tensorial nonlinear characteristics, the nonlinear polarization of the particle may induce a force transverse to the applied electric field. The transverse force can increase with increasing applied electric fields. This situation is quite reminiscent of the crossed magnetic-electric field case in a magnetorheological and electrorheological fluid [20], in which a structural phase transition from the body-centered-tetragonal (bct) to face-centered-cubic (fcc) lattice was predicted when the magnetic force and electric force are perpendicular and of comparable strength. Such a bct-fcc structural transition was confirmed recently in experiments using coated particles with magnetorheological and electrorheological responses [21]. Here we suggest that a nonlinearity-induced structural transition can occur at a sufficiently strong field by using crystalline particles with an appropriate crystal-lattice symmetry.

We have considered spherical inclusions in the dilute limit. In this case, the PD model in conjunction with the dilute limit expression can still be used as a first step to present qualitative results. However, for larger volume fractions, the PD approximation will become worse; a fully multipole model and a Maxwell-Garnett type formula other than the dilute limit expression should be adopted. We believe that the conclusion remains essentially the same. As we have included the multiple interactions in our self-consistent calculations, the results will be useful in a computer simulation of nonlinear ER fluids.

In conclusion, we have presented a study of the interparticle force of nonlinear ER fluid by means of self-consistent mean-field theory and the previously established multipole method. Taking the nonlinear characteristic into account, we predict that the normalized interparticle force is a nonmonotonic function of the applied electric field. The normalized force can achieve a maximum value at an optimal electric field E_{op}. When $E_0 < E_{op}$, the normalized force increases with E_0, while, when $E_{op} < E_0 < E_c$, it decreases with E_0. It is interesting to perform numerical simulation or experiment to verify our theoretical predictions shown above.

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