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ANALYSIS OF PARTICLE-PARTICLE FORCES IN ELECTRORHEOLOGICAL FLUIDS*

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The Rayleigh identity, based on a multipole expansion theory, is extended to analyse the forces between particles in an electrorheological system. The shear modulus for chains of particles arrayed on a square lattice is calculated. It is found that the modulus increases linearly with the ratio of dielectric constants of the dispersed particles to that of the continuous phase; as the ratio becomes larger, contrary to the expectations from a simple dipole approximation, the modulus would saturate. In the case of conducting particles, the modulus varies with the frequency of the applied field. In a limiting case of perfectly conducting particles, the conductivity is also considered. It is found that the particle–particle forces are extremely sensitive to their separations from each other.

Keywords: electrorheological fluids, permittivity, conductivity, shear stress
PACC: 4750, 4660, 7740

I. INTRODUCTION

Electrorheological (ER) fluids have attracted considerable attention in recent years because of their potential applications as well as interest in unsolved scientific problems.[1−6] The conventional picture of particle–particle interactions is that an external electric field E_0 induces a dipole moment on the particle, which causes particles to form chains in the field direction. Both dispersed and continuous phases are assumed to be uncharged and have zero conductivity. This chain structure gives rise to the increase of viscosity and shear yield strength, which are generally proportional to E_0^2, as expected.[3] However, as pointed out by Davis *et al.*,[6,7] the simple dipole approximation must break down, and higher multipole and fields from other particles must be included. The dielectric polarization analysis is valid only when conductivity effects are absent. It was suggested that the conductivity dominates the field patterns even before the chains start to form. Hence, a conceptually more serious objection to the simple picture is that it omits, for dc and low-frequency excitation, the effects of conductivity.

In this paper, the Rayleigh identity based on a multipole expansion theory is extended to deal with the forces between particles in ER fluids within a Maxwell–Wagner approach. The evaluation of the effective dielectric constant is emphasized. A limiting case of infinite particle conductivity is considered, and the shear modulus for chains of particles arrayed on a square lattice is calculated for various ratios of particle to fluid dielectric constants and those of conductivities.

II. EXTENDED RAYLEIGH IDENTITY

In the Maxwell–Wagner model, the permittivities and conductivities of the individual phases are assumed to be independent of the applied field frequency. The complex dielectric constants of the dispersed and continuous phases are written as[2]

$$\epsilon_k^*(\omega_e) = \epsilon_k - \frac{j\sigma_k}{\epsilon_0 \omega_e}, \tag{1}$$

where $k = p, c$ represent the dispersed particles and continuous phase, respectively; $j = \sqrt{-1}$, and the asterisk represents complex quantities.

For simplicity, we assume that the particles have a uniform radius size a and are aligned along the applied ac field $\boldsymbol{E}_0 = \mathrm{Re}\{E_0 \mathrm{e}^{j\omega_e t}\}\boldsymbol{e}_z$ in chains which are arranged on a square lattice. Particles form a simple tetragonal lattice with spacing d in the z (vertical) direction and w in the x and y directions. Consider an isolated sphere in the uniform ac electric field. The

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complex potential will satisfy the Laplace equation in the bulk phase and can be expanded about a sphere centre O in the following ways,[8]

$$\psi^{*i}(r,\theta,\phi)=\sum_{l=0}^{\infty}\sum_{m=-l}^{l}C_{lm}r^{l}Y_{lm}(\theta,\phi), \tag{2}$$

$$\psi^{*o}(r,\theta,\phi)=\sum_{l=0}^{\infty}\sum_{m=-l}^{l}(A_{lm}r^{l}+B_{lm}r^{-l-1})Y_{lm}(\theta,\phi), \tag{3}$$

where $Y_{lm}(\theta,\phi)$ is the spherical harmonic of the order (l,m); ψ^{*i} and ψ^{*o} are the expansions inside and outside the central sphere, respectively. The coefficients A_{lm} and B_{lm} will be determined for the specific in question.

By using the boundary conditions at the surface $(r=a)$ of the sphere and the complete orthonormal nature of spherical harmonics, we can obtain the following relationships between the coefficients in the expansion of the potential outside the particle,

$$A_{lm}=\frac{B_{lm}}{T_l a^{2l+1}}, \tag{4}$$

where

$$T_l=\frac{1-\chi}{[\chi+(l+1)/l]}, \tag{5}$$

$$\chi=\epsilon_{\rm p}^{*}/\epsilon_{\rm c}^{*}. \tag{6}$$

We need one more relation between A_{lm} and B_{lm}, other than Eq.(4), to determine them. Consider the expansion (3) for $r>a$. It contains a series of terms which increase with r and thus correspond to the sources located outside the central sphere, since the sources in this sphere can only give a contribution decreasing with r. The other series represents the contribution to the potential from sources on the central sphere. Then we can write a crucial identity, the Rayleigh identity, which expresses the fact that the potential expansion not singular at (O) is due to terms originating at infinity and at the other lattice sites,[9]

$$\sum_{lm}A_{lm}r^{l}Y_{lm}(\theta,\phi)=E_0 z+\sum_{i\neq 0}\sum_{lm}\frac{B_{lm}}{\rho_i^{l+1}}Y_{lm}(\theta_i,\phi_i), \tag{7}$$

where the sum over i is understood to run over all spheres other than the central one. Coordinates (ρ_i,θ_i,ϕ_i) are measured relative to the centre of the ith sphere.

Equation (7) is valid for all points in the immediate environment of the sphere at the origin. Using the general form of the addition theorem of spherical harmonics and its completely orthonormal nature, we obtain the extended Rayleigh identity,

$$\sum_{l=1}^{\infty}\sum_{m=-l}^{l}\left[(\alpha_1 k_3+\alpha_2)+\frac{\alpha_1^2 k_4^2}{\alpha_1 k_3+\alpha_2}\right]B'_{lm}=E_0\delta_{l',1}\delta_{m',0}, \tag{8}$$

where

$$\alpha_1=\frac{\delta_{ll'}\delta_{mm'}(l+m)!}{a^{2l+1}(l-m)!}\frac{1}{k_3^2+k_4^2}, \tag{9}$$

$$\alpha_2=\frac{(-1)^{l'+m'+1}}{(l-m)!(l'-m')!}U_{l+l'}^{m-m'}, \tag{10}$$

$$k_1=\frac{\epsilon_{\rm p}\epsilon_{\rm c}+\sigma_{\rm p}\sigma_{\rm c}/(\epsilon_0\omega_{\rm e})^2}{\epsilon_{\rm c}^2+(\sigma_{\rm c}/\epsilon_0\omega_{\rm e})^2}, \tag{11}$$

$$k_2=\frac{(\epsilon_{\rm p}\sigma_{\rm c}-\epsilon_{\rm c}\sigma_{\rm p})/(\epsilon_0\omega_{\rm e})^2}{\epsilon_{\rm c}^2+(\sigma_{\rm c}/\epsilon_0\omega_{\rm e})^2}, \tag{12}$$

$$k_3=\frac{(1-k_1)[(l+1)/l+k_1]-k_2^2}{[k_1+(l+1)/l]^2+k_2^2}, \tag{13}$$

$$k_4=\frac{k_2[(l+1)/l]}{[k_1+(l+1)/l]^2+k_2^2}, \tag{14}$$

$$U_l^m=(l-m)!u_l^m, \tag{15}$$

$$u_l^m=\sum_{i=1}^{\infty}\rho_i^{-l-1}P_l^m(\cos\theta_i)\cos(m\phi_i), \tag{16}$$

and m and m' range from $-l$ to l and $-l'$ to l', respectively.

The frequency dependence of B'_{lm} which is the real part of B_{lm} gives rise to the frequency dependence of the yield stress and the shear modulus described below. The imaginary part of B_{lm} characterizes the electrical dissipation of the system. It should be emphasized that the present formulation is rigorous and automatically includes two important effects: local-field correction and all multipole terms.

Ⅲ. SHEAR STRESS AND SHEAR MODULUS

In the absence of nonelectrostatic force, the free electrostatic energy of an ER system is proportional to the real part of the complex effective dielectric constant ($\epsilon_{\rm eff}$), and thus the determination of the structure and properties of the ER fluid may be reduced to one of the effective dielectric constant optimization and perturbation calculations. Following the derivation of effective magnetic permeability, we have obtained the formula of the effective dielectric constant

of the two-component composite in which particles are arrayed in a certain periodic structure,[4]

$$\epsilon_{\text{eff}} = 1 - \frac{4N\pi B'_{1,0}}{VE_0}, \tag{17}$$

where N is the number of particles in a unit cell of volume V.

In order to calculate the shear stress, it is necessary to perturb the system away from its original structure. We assume the chain structure to be sheared by an angle θ and the shear rate to be infinitesimal. In the absence of nonelectrostatic force, the free energy of the system changes an amount of $\Delta(-\boldsymbol{E}\cdot\boldsymbol{D}/2)$, and thus the shear stress is

$$\tau = -\frac{1}{2}\epsilon_{\text{c}}\frac{\partial\epsilon_{\text{eff}}(\theta)}{\partial\theta}E_0^2. \tag{18}$$

When a small shear is applied to the system, the interparticle spacing d along the z direction increases, which results in the principal variation of ϵ_{eff}. In this case, $\partial d/\partial\theta = \theta d$ and $\tau = G\theta$, so we have

$$G = \frac{1}{2}\epsilon_{\text{c}}(-d\frac{\partial\epsilon_{\text{eff}}}{\partial d})E_0^2, \tag{19}$$

subject to the constraint of the constant particle volume fraction.

IV. NUMERICAL RESULTS

By solving identity (8), we can obtain the value of $B'_{1,0}$ which may be used in Eq.(17) to calculate the effective dielectric constant. Then, we evaluate the shear stress. The frequency of the applied field is assumed to be high enough and the conductivity effects are unimportant. The computed static yield stress τ is displayed in Fig.1 for a typical volume fraction, $f = 0.2$, and a typical electric field 2kV/mm. From Fig.1, we can see that τ almost increases linearly for a small shear. As θ increases, τ reaches a maximum which is defined as the static yield stress. The dashed curve indicates an unstable regime. This agrees with the results of Ma *et al.*[5]

A comparison for G between our result and the dipole approximation is made in Fig.2, in which the conductivity effects are also neglected. We can see that G becomes almost linear for $\alpha = \epsilon_{\text{p}}/\epsilon_{\text{c}}$ as large as 25, which differs entirely from the dipole approximation result in which it saturates at large α. This is because the dipole approximation is expected to break down, and multipole interactions become much more important when particles nearly touch or α becomes so large that $\beta = (\alpha-1)/(\alpha+2)$ is near unity. The present result agrees well with that of Davis.[7]

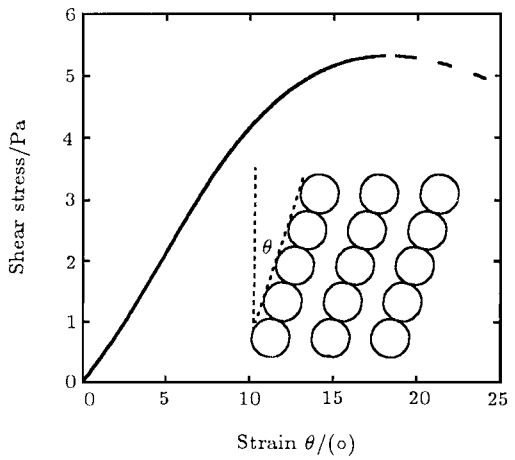


Fig.1. Calculated stress, in pascal units, plotted as a function of strain, i.e., the title angle θ as defined in the inset. The dashed curve indicates the unstable regime. The parameters used in the calculation are $\epsilon_{\text{p}}/\epsilon_{\text{c}} = 5.0$, E=2.0kV/mm and $f = 0.2$.

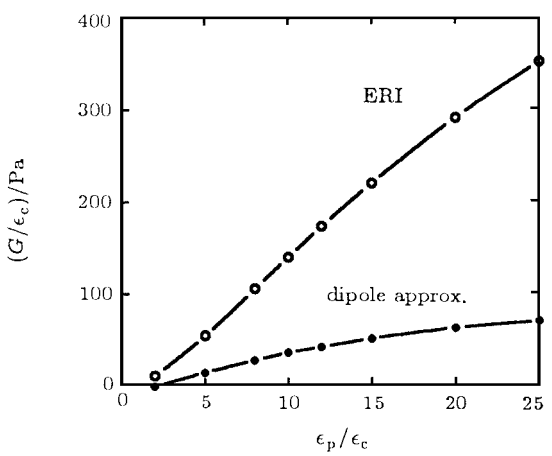


Fig.2. Shear modulus G divided by ϵ_{c} versus $\epsilon_{\text{p}}/\epsilon_{\text{c}}$. The volume fraction is $f = 0.2$ and the applied electric field is E=2.0kV/mm. The curve labelled "ERI" is from the extended Rayleigh identity and the curve labelled "dipole approx." is from point dipole approximation with local field corrections.

From the shear strain dependence of the effective dielectric constant, the dependence of shear modulus G on the frequency of the applied field is calculated. The computed shear modulus G/ϵ_{c} is shown in Figs.3(a) and 3(b), in which the volume fractions of particles are both 0.4. In order to investigate the conductivity effects of a real ER system, two cases are considered: the ratio of dielectric constants $(\epsilon_{\text{p}}/\epsilon_{\text{c}})$ is smaller or larger than that of conductivity $(\sigma_{\text{p}}/\sigma_{\text{c}})$. As can be seen in Fig.3(a), the shear modulus G is larger when the frequency of the applied electric field

is lower. This is because $\sigma_p/\sigma_c > \epsilon_p/\epsilon_c$ and the polarization of particles is dominated by conductivities. When the frequency becomes as large as 10Hz, G decreases rapidly as the polarization of particles is dominated by dielectric constants, unaffected by conductivities. The shear modulus in Fig.3(b) changes, in contrast to Fig.3(a), as $\sigma_p/\sigma_c < \epsilon_p/\epsilon_c$, i.e., the shear modulus under ac excitation is larger than that under dc excitation. The transition frequency is about 200Hz, which is larger than that in Fig.3(a), for the conductivity of fluids (10^{-8}S/m) is higher than that in Fig.3(a) (10^{-10}S/m). This result agrees with that of Davis *et al.*[2,10]

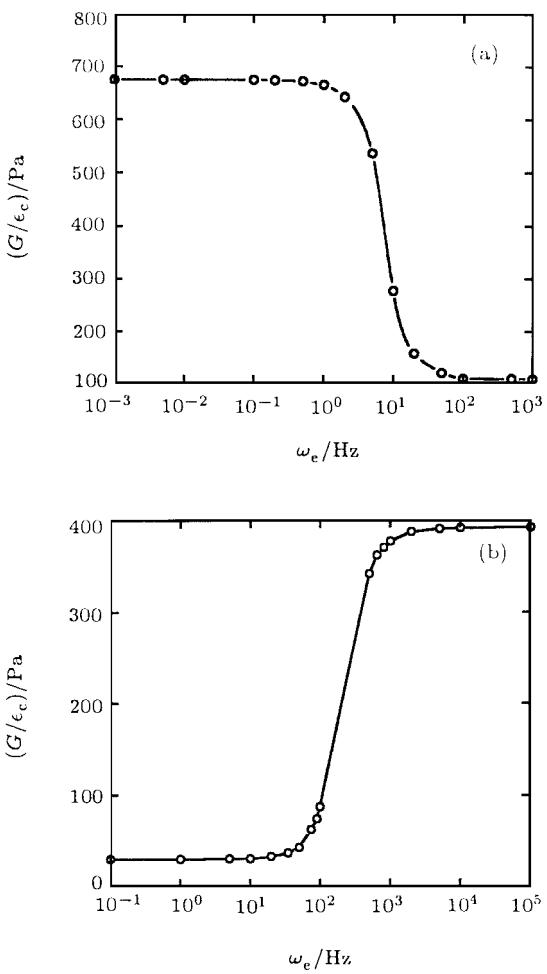


Fig.3. Shear modulus G divided by ϵ_c, as a function of the frequency of applied electric field ω_e: (a) $\epsilon_p/\epsilon_c = 10$ and $\sigma_p/\sigma_c = 100$; (b) $\epsilon_p/\epsilon_c = 32$ and $\sigma_p/\sigma_c = 5$. The particle volume fraction (f) is 0.4 and the electric field is $E_0 = 2.0$kV/mm.

The calculated results of a limiting case of infinite conductivity of particles are displayed in Fig.4. As d approaches $2a$ (particle touching), $\partial\epsilon_{eff}/\partial d$ appears to diverge, as has been noted before by Klingenberg *et al.*[11] This means that the particle–particle forces are extremely sensitive to their separation, but could be much larger than in the dielectric regime. If ϵ_p were much larger than ϵ_c, a similar effect would occur, but it is difficult to obtain ratios of ϵ_p to ϵ_c greater than about 80 : 2.5. However, it is likely that the value of σ_p/σ_c could be as large as 10^3.[7] This suggests that the common observation, that yield stress decreases in going from dc to ac excitation, may result from the passing of a conductivity regime (dc) into a dielectric regime at large ω_e. Furthermore, this picture would suggest that attempts to increase the yield strength by increasing ϵ_p will be fruitless unless ac excitation is employed.

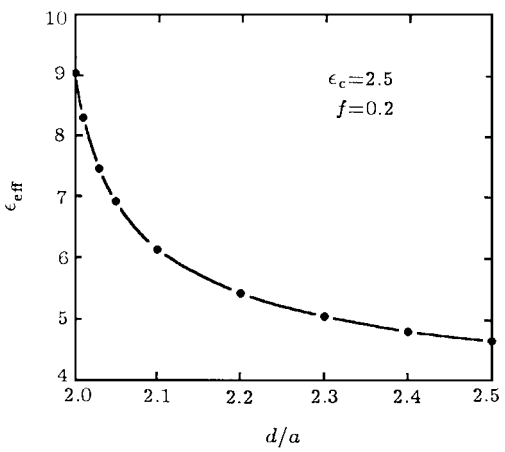


Fig.4. Effective dielectric constant ϵ_{eff} (in the direction of the electric field) of ER fluid with infinitely conductive spherical particles of interparticle spacing d. The slope of ϵ_{eff} versus d diverges when particles touch ($d = 2a$).

V. DISCUSSION AND CONCLUSIONS

The electrical interaction of particle–particle forces in an ER system is most important for the application of ER fluids. Dipole approximation is expected to break down when particles are nearly touching or α becomes large. However, realistic ER fluids often have large α, and thus multipolar interaction must be considered. In this paper, we have extended the Rayleigh identity based on a multipole expansion to deal with particle–particle forces and the effects of conductivity. One can see large differences between our results (Fig.2) and that of the dipole approximation. This is because the dipole approximation is expected to break down and multipole interactions become much more important when particles nearly touch or the α becomes so large that $\beta = (\alpha - 1)/(\alpha + 2)$ is near unity. We do not attempt to make a detailed comparison between our results and experiments because of the

lack of measurements reported on well characterized systems at frequencies where conductivity effects are included. However, the curve of $\epsilon_{\rm eff}$ of the limiting case of infinite conductivity of particles versus the interparticle spacing d and that of shear modulus $G/\epsilon_{\rm c}$ versus the frequency of the electric field show good agreement with the results of Ref.[9]. From our results, we may explain, qualitatively, why high dielectric constant materials, such as barium titanate, do not show a very large ER effect in dc fields, and why many systems exhibit a decrease in apparent viscosity with increasing electric field frequency.[12]

One of the implications of the present study for the design of stronger ER fluids is that a large ratio of particle-to-fluid conductivities, $\sigma_{\rm p}/\sigma_{\rm c}$, is necessary for dc or low-frequency applications. As noted previously, the frequently observed decrease in shear modulus or shear stress at high frequency is probably due to a change from a conductivity dominated regime to a dielectric regime (if $\sigma_{\rm p}/\sigma_{\rm c} > \epsilon_{\rm p}/\epsilon_{\rm c}$). By the same token, the benefits of a system with large $\epsilon_{\rm p}/\epsilon_{\rm c}$ can be obscured by conductivity effects in dc, and thus may have been overlooked previously if $\sigma_{\rm p}/\sigma_{\rm c} < \epsilon_{\rm p}/\epsilon_{\rm c}$. Equation (8) has a general form; it can be used to investigate other properties of ER fluids.

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