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DYNAMIC ELECTRORHEOLOGICAL EFFECTS OF ROTATING PARTICLES: A BRIEF REVIEW

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Particle rotation leads to a steady-state which is different from the equilibrium state in the absence of rotational motion. The change of the polarization of the particle due to the rotational motion is called the dynamic electrorheological effect (DER). There are three cases to be considered: rotating particles in a dc field, particle rotation due to a rotating field and spontaneous rotation of particle in dc field (Quincke rotation). In the DER of rotating particles, the particle rotational motion generally reduces the interparticle force between the particles. The effect becomes pronounced when the frequency is on the order of the relaxation rate of the surface charges. In the electrorotation of particles, the mutual interaction between approaching particles will change the electrorotation spectrum significantly. The electrorotation spectrum depends strongly on the medium conductivity as well as the conductivity contrast between the particle and the medium. In the collective behaviors of Quincke rotors, the mutual interactions between the individual rotors lead to the assembly of chain-like structures which make an angle with the applied field. This has an implication of a new class of material.

1 Introduction

The prediction of the strength of the electrorheological (ER) effect is the main concern in the theoretical investigation of ER fluids. [1,2] The ER effect leads to anisotropic forces between the particles. In deriving the induced forces between the particles, traditional theories assume that the particles are at rest. [3,4] In a realistic situation, the fluid flow exerts force and torque on the particles, setting the particles in both translational and rotational motions. [5] In this work, we shall consider the case of rotating particles, in an attempt to discuss the effect of the particle rotation on the interparticle force and the structures.

In electrorotation (EOR), interactions between a rotating electric field and suspended particles also lead to a rotational motion of the particles. EOR has received much attention in micromanipulation and separation of submicron size particles. [6,7] In the dilute limit, one can concentrate on the EOR response of individual particles by ignoring the mutual interactions between the particles. However, if the suspension is non-dilute, one may not ignore the interactions. Moreover, when the strength of the rotating electric field increases, the Brownian motion can be ignored and the polarized particles tend to aggregate along the rotating field even in the dilute limit. As an initial model, we shall consider a pair of interacting particles by invoking the method of multiple images, [8] in an attempt to investigate the effect of the mutual polarization interaction on the EOR spectrum and two Quincke rotors.

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2 Formalism

2.1 Rotating particles in a DC field

We consider two spherical particles, one of which is held fixed, and the other one rotates around the axis perpendicular to the line joining the particles centers, see Fig. 1. In the presence of an electric field, the induced dipole moment admits $\mathbf{p}_{a0} = p_{a0}\hat{z}$, where

$$p_{a0} = \epsilon_m E_0 \left(\frac{\epsilon_1 - \epsilon_m}{\epsilon_1 + 2\epsilon_m}\right) a^3. \tag{1}$$

In the case of a rotational motion of a particle, a displacement of the polarization

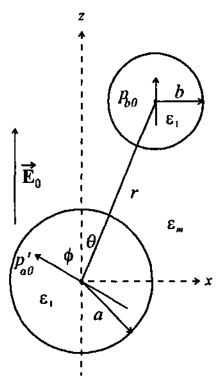

Figure 1. A rotating polarized dielectric sphere interacting with another non-rotating polarized dielectric sphere.

charge is expected to appear on the particle surface.

If the sphere undergos an anticlockwise rotation with angular velocity $\omega = -\omega\hat{y}$, when a steady state is reached, the steady induced dipole moment $\mathbf{p}$ is expressed as [9]

$$\mathbf{p} = \frac{\mathbf{p}_{a0} + \tau_r(\omega \times \mathbf{p}_{a0})}{1 + (\omega\tau_r)^2}, \tag{2}$$

where τ_r stands for the characteristic relaxation time. We are in a position to generalize the multiple image method [8] to calculate the force between the polarized spheres, when the induced dipole moments are not parallel. Thus, the total dipole moments can be separated into x and z composites, given by $\mathbf{p}_a = p_{ax}\hat{x} + p_{az}\hat{z}$, $\mathbf{p}_b = p_{bx}\hat{x} + p_{bz}\hat{z}$. The components can be found by using the multiple image method, see Ref. 8. Then, the force between the spheres is given by the following expression, based on the energy considerations [10]

$$\mathbf{F} = \frac{1}{2}\nabla[\mathbf{E}_0 \cdot (\mathbf{p}_a + \mathbf{p}_b)] = F_r\hat{r} + F_\theta\hat{\theta}. \tag{3}$$

We will calculate the results for an anticlockwise rotation ($\omega = -\omega\hat{\mathbf{y}}$) only. The results for a clockwise rotation can be found on the same footing.

2.2 Particle rotation due to a rotating field

We first consider an isolated spherical particle with diameter D(*i.e.*, $2a$), of complex dielectric constant $\tilde{\epsilon}_1$ dispersed in a suspension medium of $\tilde{\epsilon}_2$, where $\tilde{\epsilon} = \epsilon + \sigma/i2\pi f$. Under the application of a rotating electric field, the induced complex dipole moment inside the isolated particle is $\tilde{p} = \epsilon_2 D^3 b_0 E_0/8$, where the dipole factor $b_0 = (\tilde{\epsilon}_1 - \tilde{\epsilon}_2)/(\tilde{\epsilon}_1 + 2\tilde{\epsilon}_2)$.

To account for the effect of multipolar interaction on the EOR, we consider a pair of identical touching particles with center-to-center separation R suspended in a medium. We will calculate the effect of multipolar interaction on the dipole moment. For a rotating field, the total dipole moment of one particle in the pair is given by $\tilde{p}^* = (\tilde{p}_T + \tilde{p}_L)/2$, [11] where $\tilde{p}_T$ ($\tilde{p}_L$) is the transverse (longitudinal) dipole moment, being perpendicular (parallel) to the line joining the centers of the particles. The expressions for the transverse and longitudinal dipole moments can be found in Ref. 8. As a result, we obtain the dipole factor of a pair, [11]

$$b^* = \frac{1}{2} b_0 \left[\sum_{n=0}^{\infty} (-b_0)^n \left(\frac{\sinh \alpha}{\sinh(n+1)\alpha} \right)^3 + \sum_{n=0}^{\infty} (2b_0)^n \left(\frac{\sinh \alpha}{\sinh(n+1)\alpha} \right)^3 \right]. \tag{4}$$

Let us denote a complex material parameter $\tilde{s} = (1 - \tilde{\epsilon}_1/\tilde{\epsilon}_2)^{-1}$, and then the dipole factor of an isolated particle b_0 is given by spectral representation [12] $b_0 = F_1/(\tilde{s} - s_1)$, with $F_1 = -1/3$ and $s_1 = 1/3$. We further define two dimensionless parameters [13] $s = (1 - \epsilon_1/\epsilon_2)^{-1}$, $t = (1 - \sigma_1/\sigma_2)^{-1}$, where s and t are the real dielectric contrast and conductivity contrast, respectively. After simple manipulations, we have [11]

$$b_0 = \frac{F_1}{s - s_1} + \frac{\delta\epsilon_1}{1 + if/f_c}, \tag{5}$$

where f_c is the characteristic frequency, at which the maximum angular velocity of EOR occurs, and $\delta\epsilon_1$ is the dispersion strength. Similarly, the dipole factor of a pair of interacting particles b^* [Eq. (4)] can be exactly rewritten in the spectral representation as [11]

$$b^* = \sum_{m=1}^{\infty} \left(\frac{F_m{}^{(T)}}{\tilde{s} - s_m{}^{(T)}} + \frac{F_m{}^{(L)}}{\tilde{s} - s_m{}^{(L)}} \right). \tag{6}$$

For the detailed expressions for F_m and s_m, please refer to Ref. 11. We can also proceed to express each summand in Eq. (6) as the dispersion term which is similar to Eq. (5) [11].

2.3 Spontaneous rotation of particles in a DC field (Quincke rotation)

When a non-conducting particle immersed in a semi-insulating liquid is submitted to a sufficiently high amplitude DC or plane-polarized AC electric field, it begins to rotate spontaneously around itself with axis pointing in any direction perpendicular to the field. This symmetry break is known as Quincke rotation. [14]

Since relaxation process can be of different microscopic origin, then the retarding polarization results a dipole moment $\mathbf{p}$ which can be calculated by solving the relaxation for retarding part of the polarization [9,15]. The total polarization can be divided into two parts: the instantaneous polarization $\mathbf{p_0} = \chi\mathbf{E} = 4\pi\epsilon_2(\epsilon_1 - \epsilon_2)/(\epsilon_1 + 2\epsilon_2)a^3\mathbf{E}$ coming from the permittivity mismatch between the particle and the liquid, and the retarding polarization $\mathbf{p}$ related to the relaxation processes. The rotation is balanced by spin friction, *i.e.*, $\alpha\omega = 8\pi\eta a^3\omega$.

To consider two interacting Quincke rotors, we should introduce the mutual interaction effect of the rotors, especially when they are very close to each other. For simplicity, the two rotors are assumed to be identical, in steady state the two rotors form a head-to-tail chain and the dipole induced dipole moment is along the opposite direction of the original one. Thus we have

$$\frac{d\mathbf{p}}{dt} = \omega \times \mathbf{p} - \frac{1}{\tau_r}[\mathbf{p} - \chi\mathbf{E} + G(r)\mathbf{p}]. \tag{7}$$

$G(r)=\chi\rho(r)$ designates the multipolar interaction strength and is strongly dependent on the separation of the two rotors. [9] $G(r) = 0$ means an isolated rotor and Eq. (7) degenerates to the single particle one.[15] In estimation of Quincke rotation case, $G(r)$ is negative. The retarding polarization of a particle can be obtained:

$$p_x = \frac{\alpha\omega}{E} = \frac{\alpha}{\tau_r E}\sqrt{\frac{E^2}{E_c^2} - (1 - G(r))^2}, p_z = \frac{\alpha(1 - G(r))}{\tau_r E}. \tag{8}$$

It is easy to tell that the amplitude of the dipole moment is the same as that of the single one and keeps unchanged when the electric field increases [see Fig. 4(b)]. The multipolar interactions of the interacting rotors change the tilting angle of the chain, *i.e.*, $\tan\theta = \tau_r\omega/(1 - G(r))$ and the rotation velocity is reduced,

$$\omega = \pm\frac{1}{\tau_r}\sqrt{\frac{E^2}{E_c^2} - (1 - G(r))^2}\hat{y}. \tag{9}$$

3 Numerical results

For the sake of convenience, we let $a = b$ in Fig. 2, the radial force between the spheres is plotted against the separation parameter σ, defined by $\sigma = r/(a + b)$. In the transverse (longitudinal) field case, the line joining the spheres is perpendicular (parallel) to the applied field. We first examine the transverse field case, with $\omega\tau_r$ being chosen to be 0.1, 1, and 10, respectively. The dielectric contrast ($\tau = (\epsilon_1 - \epsilon_2)/(\epsilon_1 + \epsilon_2)$) is chosen to be $\tau = 1/3$ (low contrast) and $\tau = 9/11$ (high contrast). From Fig. 2, our results show that the rotational motion of the particle generally reduces the force between the particles. Because Fig. 2 plots the products $F_T\sigma^4$ and $F_L\sigma^4$ against σ, it can be seen that for a large separation ($\sigma > 3$), these quantities tend to be constant, indicating that the force varies as σ^{-4}. For a small separation ($\sigma < 1.5$), the magnitude of the transverse force increases rapidly. The longitudinal field case shows a similar behavior: the rotational motion generally reduces the magnitude of the interparticle force.

In Fig. 3, the EOR spectrum is given by the imaginary part of the corresponding dipole factors.[16] It is evident from the left panel that, for both cases, a small $|t|$ yields a high characteristic frequency, at which the peak occurs. Moreover, a

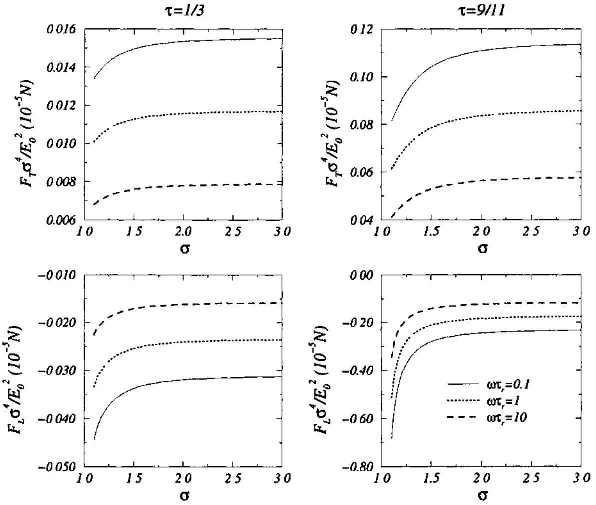

Figure 2. Interparticle force $F_T\sigma^4$ and $F_L\sigma^4$ plotted against σ. The subscript T denotes the transverse dipole $\theta = \pi/2$) and L the longitudinal dipole. The magnitude of the forces increases rapidly when the separation becomes small.

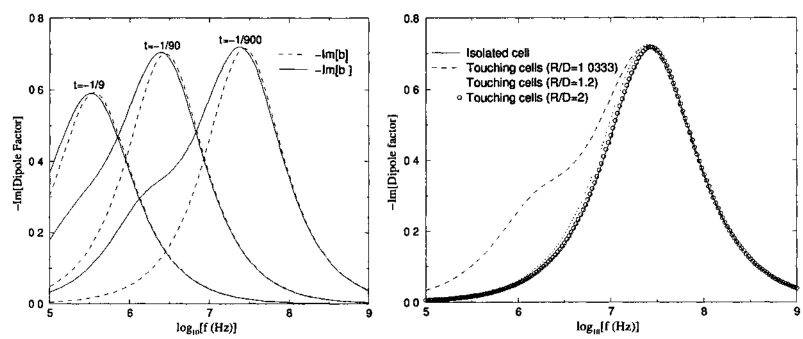

Figure 3. Left panel: Electrorotation spectrum for isolated particle (dashed curves) case and touching particle (solid curves) for three different t at $s = 1.1$, $\sigma_2 = 2.8\times10^{-4}$ S/m, and $\epsilon_2 = 80\epsilon_0$. Right panel: For three different R/D at $s = 1.1$, $t = -1/900$, $\sigma_2 = 2.8\times10^{-4}$ S/m, and $\epsilon_2 = 80\epsilon_0$

smaller $|t|$ gives a larger peak value. Given a constant t, the touching-particle case predicts a shift of the peak location to lower frequency than that predicted by the isolated-particle case. At the same time, the peak for the touching-particle case broadens. This behavior arises from the effect of the multipolar interactions. In addition, if t is given, the two cases also predict different behavior within the low-frequency region, but almost the same behavior within the high-frequency region. Especially for small $|t|$ (*e.g.*, $t = -1/900$), a second peak occurs at lower frequency. This is in accord with the prediction of the spectral representation. Effect of the variation of σ_2 can be found in Ref. 11. In Fig. 3 right panel, as $R/D \approx 1$ (*e.g.*, $R/D = 1.0333$), the deviation between the two cases is evident. In other words, the multipolar interaction does play an important role in the EOR spectrum and the effect cannot be neglected for touching particles. However, as the separation increases, say, $R/D = 2$, both cases predict the same EOR spectrum. From the results, we would say that the effect of multipolar interaction may be neglected as $R/D > 2$.

Figure 4 is plotted for $G = 0$ (*i.e.*, an isolated particle) and $G = -0.1$ (the estimated maximum correction of multiple image effect). Fig. 4(a) depicts the

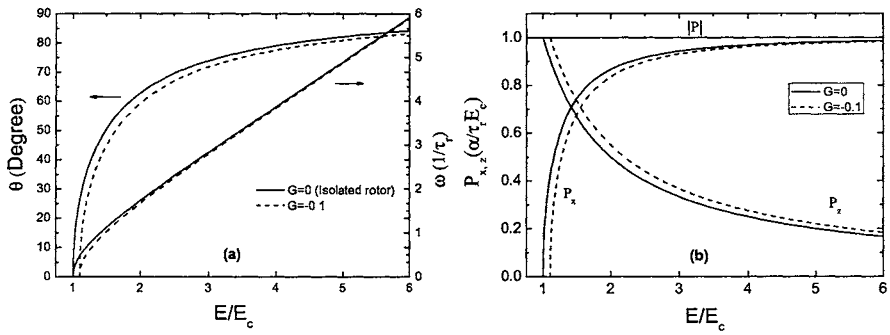

Figure 4. (a) The dependence of tilting angular θ and reduced angular velocity ω on the external electric field strength. (b)The dependence of induced dipole moment $\mathbf{p}$ and it's components p_x, p_z on the external electric field strength.

tilting angle θ and the rotational angular velocity ω dependence on the reduced electric field strength E/E_c ($E_c = \sqrt{-\alpha/(\tau_r \chi)}$). Both θ and ω increase with the field strength. And the upper boundary value of θ is 90 degree. The multipolar interaction introduce an increase of the critical electric field value for the starting of the particle rotation. And for the same electric field, it reduces θ and ω, When E/E_c becomes greater enough, the multipolar effect on the system becomes insignificant. Fig. 4(b) shows the variation of the dipole moment as the electric field increases. It is clear that p_x increases with the electric field strength, meanwhile p_z decreases concomitantly, but the dipole moment strength keeps to be $\alpha/(\tau_r E_c)$ for both cases. Note that the chain tilting angular is along the direction of $\mathbf{p}$, which is an assumption to keep it valid that in steady state no additional torque is added by the multiple image effect. Obviously, when they keep head-to-tail and approach closer, the mutual interaction of the two particles reduces p_x and the rotation, which means that the relaxation process tends to overwhelm the instantaneous polarization.

4 Discussion and conclusion

We have developed a model to calculate the interparticle force between a pair of dielectric spheres, in which one sphere is held fixed and the other is in rotational motion. By using the multiple image method, we found that the effect of rotation is large when the angular velocity of the sphere is large. In fact, our approach can treat the case of both rotating particle in the same way. In addition, we can also take into account the effect of oscillatory rotation on the interparticle force. [15] Also, we have attempted a theoretical study of the EOR of two approaching spherical particles in the presence of a rotating electric field. From the results, we find that the mutual polarization effects can change the characteristic frequency substantially.

We should remark that the multiple image method for two dielectric spheres is approximate, [17] but the approximation is quite good when compared with the integral equation approach. [8] More accurate calculations based on bispherical coordinates can be attempted. However, we believe that similar conclusions can be obtained.

Due to the spontaneous rotation of Quincke rotor, it shows different dynamic behavior and interaction, which is an implication of a totally new class of material that would be valuable for further investigation. Quincke rotor assembly is under simulation by molecular dynamic method, we found the chain could be further longer up to 7 particles.

Acknowledgments

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