Relaxation of surface charge on rotating dielectric spheres: Implications on dynamic electrorheological effects

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We have examined the effect of an oscillatory rotation of a polarized dielectric particle. The rotational motion leads to a redistribution of the polarization charge on the surface of the particle. We show that the time-averaged steady-state dipole moment is along the field direction, but its magnitude is reduced by a factor that depends on the angular velocity of rotation. As a result, the rotational motion of the particle reduces the electrorheological effect. We further assume that the relaxation of the polarized charge is arised from a finite conductivity of the particle or host medium. We calculate the relaxation time based on the Maxwell-Wagner theory, suitably generalized to include the rotational motion. Analytic expressions for the reduction factor and the relaxation time are given and their dependence on the angular velocity of rotation will be discussed.

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I. INTRODUCTION

The prediction of the strength of the electrorheological (ER) effect is still a main concern in theoretical investigation of ER fluids [1–5]. An ER fluid is a suspension of highly polarized particles in an insulating host. The ER effect originates from the induced interaction between the polarized particles in an ER fluid. Upon the application of an intense electric field, the particles in ER fluid aggregate into chains and then aggregate into columns in a short response time [3,4]. The rapid field-induced transition between the fluid and solid phase makes this material important both for wide industrial applications and for experimental and theoretical investigation.

In deriving the induced interactions between particles, existing theories assume that the particles are at rest [6–10]. In a realistic situation, the fluid flow exerts force and torque on the particles, setting the particles in both translational and rotational motions. For instance, the shear flow in an ER suspension exerts a torque on the particles, which leads to a rotational motion of the particles about their centers [11]. Recent experiments gave evidence that the induced forces between the rotating particles can be different from the values predicted by existing theories [12].

To gain some insight into the phenomenon, we have recently formulated a theoretical model, which describes the relaxation of the polarized charge on the surface of a uniformly rotating particle [13]. We showed that the rotational motion of the particles reduces the induced forces between the particles. We called the reduction of interparticle forces due to the rotational motion of the particles the dynamic ER effects [13]. In this paper, we extend the consideration to an arbitrary rotational motion. In particular, we will obtain the steady-state dipole moment of a rotating sphere under a sinusoidal oscillatory shear motion. We further assume that the relaxation of the polarized charge is due to a finite conductivity of the particle or host medium. We will derive an analytic expression for the relaxation time. The dependence of the reduction factor and the relaxation time on the angular velocity of rotation will also be calculated.

II. STEADY-STATE DIPOLE MOMENT

Consider a dielectric sphere under the influence of an electric-field $\vec{E}_0 = E_0\hat{z}$; its induced dipole moment is given by: $\vec{p}_0 = p_0\hat{z}$. Assume that it is under a rotational motion of angular velocity $\vec{\omega} = -\omega\hat{y}$. For a rotating dielectric sphere in an electric field, the rotational motion leads to a displacement of its polarized charges on the surface of sphere. As a result, there is a change of the dipole moment, described by $\vec{\omega}\times\vec{p}$. The surface charges also suffer from relaxation of various kinds, and the rate of change of the dipole moment is described by $-(\vec{p}-\vec{p}_0)/\tau$, where τ is a relaxation time. The two effects have to be balanced against each other, resulting in a steady-state dipole moment $\vec{p}$, which deviates from the equilibrium dipole moment $\vec{p}_0$. Let the resultant dipole moment be $\vec{p} = p_x\hat{x} + p_y\hat{y} + p_z\hat{z}$. The rate of change of the dipole moment is given by

$$\frac{d\vec{p}}{dt} = \vec{\omega}\times\vec{p} - \frac{1}{\tau}(\vec{p}-\vec{p}_0), \tag{1}$$

where the first term on the right-hand side is due to the rotational motion and the second term is due to a relaxation process, in which the relaxation-time τ is determined by the details of the relaxation process. In component form, the differential equation reads

$$\dot{p}_x = -\frac{p_x}{\tau} - \omega p_z, \quad \dot{p}_y = -\frac{p_y}{\tau}, \quad \dot{p}_z = \omega p_x - \frac{(p_z - p_0)}{\tau}.$$

The equation for p_y may be readily integrated to yield $p_y = p_{y0}e^{-J(t)}$, where $J(t) = \int_0^t dt/\tau$. Since τ (can be time dependent) is real and positive, p_y vanishes as t goes to infinity. To solve the equations for p_x and p_z, we use the complex

notation: let $\tilde{p}=p_x+ip_z$ and $\tilde{p}_0=ip_0$, $\tilde{p}$ can be found by solving the following differential equation:

$$\frac{d\tilde{p}}{dt}=\left(i\omega-\frac{1}{\tau}\right)\tilde{p}+\frac{\tilde{p}_0}{\tau}. \tag{2}$$

With the initial condition $\tilde{p}=\tilde{p}_0$ when $t=0$, Eq. (2) admits a standard solution

$$\tilde{p}e^I-\tilde{p}_0=\tilde{p}_0\int_0^t\frac{e^I}{\tau}dt, \qquad I=\int_0^t\left(\frac{1}{\tau}-i\omega\right)dt, \tag{3}$$

where I is the integration factor. For a uniform rotational motion, $\omega=\omega_0$ is a constant, $I=t/\tau-i\omega_0 t$, Eq. (3) can be solved

$$\tilde{p}=\frac{\tilde{p}_0}{1-i\omega_0\tau}(1-i\omega_0\tau e^{-t(1-i\omega_0\tau)/\tau}).$$

As t goes to infinity, we obtain the steady-state solution for a uniform rotation

$$\tilde{p}=\frac{\tilde{p}_0}{1-i\omega_0\tau}. \tag{4}$$

In general, the analytic solution of Eq. (3) can be found only for a few simple cases and the integral must be evaluated numerically. We concentrate on the steady-state solution at a sufficiently long time and Eq. (3) may indeed be solved exactly. By using the L'Hôpital's rule, we find

$$\tilde{p}=\tilde{p}_0\lim_{t\to\infty}\frac{e^I}{\tau e^I\dot{I}}=\frac{\tilde{p}_0}{1-i\omega\tau}, \tag{5}$$

where $\dot{I}$ denotes the time derivative of I. We have assumed that τ is real and positive but ω may be an arbitrary function of time. Equation (5) is the general result for arbitrary rotational motion, being of the same form as Eq. (4). However, the transient solution has to be calculated numerically.

For a dielectric sphere undergoing a simple harmonic oscillation, $\theta(t)=\theta_0\sin(kt)$, the angular velocity is given by $\omega(t)=\dot{\theta}=\theta_0 k\cos(kt)$. From Eq. (5), the steady-state dipole moment is

$$\tilde{p}=\frac{\tilde{p}_0}{1-i\theta_0 k\tau\cos(kt)}=p_0\frac{i-\theta_0 k\tau\cos(kt)}{1+\theta_0^2k^2\tau^2\cos^2(kt)}. \tag{6}$$

The time dependence of the dipole moment is still periodic. Note that although the sphere is undergoing a simple harmonic oscillation, the dipole moment does not exhibit a simple harmonic motion. If τ is independent of time, we can calculate the time average of the dipole moment

$$\frac{\langle p_x\rangle}{p_0}=0 \quad \text{and} \quad \frac{\langle p_z\rangle}{p_0}=\frac{1}{\sqrt{1+\theta_0^2k^2\tau^2}}. \tag{7}$$

As a result, the motion of particles reduces the ER effect. We define the reduction factor R as

$$R=\frac{\langle p_z\rangle}{p_0}=\frac{1}{\sqrt{1+\theta_0^2k^2\tau^2}}. \tag{8}$$

The reduction is even more significant at high frequencies, $R\approx 1/\theta_0 k\tau$.

III. CALCULATION OF RELAXATION TIME

So far, our proposed relaxation time has no explicit expression. If the relaxation process is originated from a finite conductivity of the particle or host medium, then we can calculate the relaxation time based on the Maxwell-Wagner theory of leaky dielectrics [14]. For a (nonrotating) spherical inclusion embedded in a host medium, the expression is

$$\tau=\epsilon_0\left(\frac{\epsilon_1+2\epsilon_m}{\sigma_1+2\sigma_m}\right), \tag{9}$$

where ϵ_1,ϵ_m (σ_1,σ_m) denote the dielectric constant (conductivity) of the sphere and host medium, respectively, ϵ_0 is the permittivity of free space. For typical values of the permittivities and conductivities of common ER fluids, the relaxation time ranges from microseconds to milliseconds, and the dynamic ER effect can be observed in experiments.

In order to account for the impact of a rotational motion on the relaxation time, we first replace ϵ_1 in Eq. (9) by $\epsilon_1=1+\chi_1$, where χ_1 is the susceptibility of the sphere. We already showed that the dipole moment is reduced by a factor R. If we assume that the polarization is uniform throughout the sphere, which can be achieved when the oscillating frequency is high, we may write $\epsilon_1=1+R\chi_1$ in Eq. (9). Physically, it means that the effective polarization of the sphere is reduced as a result of the rotational motion, leading to a reduction of the effective dielectric constant of the sphere. After some simplifications, we obtain

$$\tau=\tau_\infty+\frac{\tau_0-\tau_\infty}{\sqrt{1+\theta_0^2k^2\tau^2}}, \tag{10}$$

where

$$\tau_\infty=\epsilon_0\left(\frac{1+2\epsilon_m}{\sigma_1+2\sigma_m}\right) \quad \text{and} \quad \tau_0=\epsilon_0\left(\frac{\epsilon_1+2\epsilon_m}{\sigma_1+2\sigma_m}\right).$$

It may be shown that $\tau=\tau_0$ for $k\theta_0=0$ and $\tau\to\tau_\infty$ for $k\theta_0\to\infty$. Equation (10) is a self-consistent equation for τ and we may calculate the relaxation time self-consistently.

IV. NUMERICAL RESULTS

To examine the dependence of the reduction factor R on the angular velocity $k\theta_0$, we plot R vs $k\theta_0$ in Fig. 1 for several different values of τ_0. Without loss of generality (i.e., in terms of some unit relaxation time), we choose $\tau_\infty=1$ and $\tau_0=2$, 4, and 8, respectively. The reduction factor decreases rapidly with the increase of $k\theta_0$, which means that the dipole

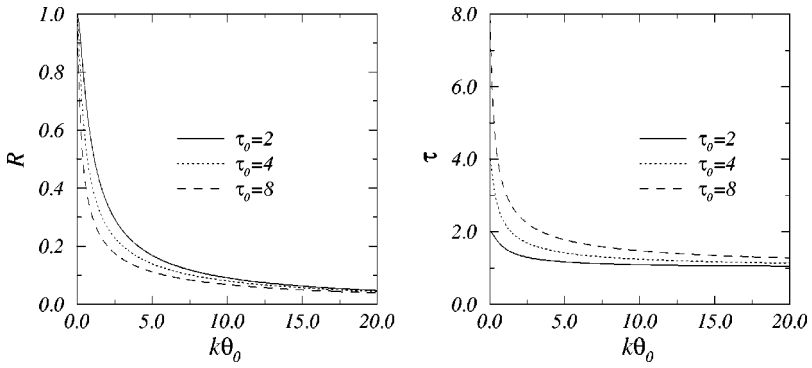

FIG. 1. The reduction factor R (left panel) and the relaxation-time (right panel) τ for $\tau_\infty=1$. The reduction factor decreases drastically for increasing $k\theta_0$. The relaxation time reaches its minimum value at large $k\theta_0$.

moment is greatly reduced when both the oscillation frequency k and the oscillation amplitude θ_0 become large.

Next, we see how the relaxation time depends on $k\theta_0$. From Eq. (10), τ is bounded between τ_0 and τ_∞. The lower bound τ_∞ is reached when $k\theta_0$ tends to infinity, which is achieved at large frequency. For a larger value of τ_0, the relaxation time decreases more rapidly with $k\theta_0$. The condition of high-oscillation frequency reads

$$k \gg \frac{1}{\tau_\infty}. \tag{11}$$

The time evolution of the dipole moment is worth studying. In Fig. 2, we plot the steady-state solution of the perpendicular component p_x/p_0 and parallel component p_z/p_0 against time. We set $\tau_0=2$ and $\tau_\infty=1$, τ is then calculated from Eq. (10). We first set $k=1$ and vary the oscillation amplitude θ_0. In the left panel of Fig. 2, we plot p_x/p_0 and p_z/p_0 against time for $\theta_0=\pi/4$, $\theta_0=\pi/2$, and $\theta_0=\pi$, respectively. For each value of θ_0, the magnitude of the perpendicular component (p_x/p_0) has a maximum value of 0.5 and it has a local minimum at $t=\pi/k$. When the oscillation amplitude increases, the local minimum value decreases, showing a large variation of p_x/p_0. Next, we concentrate on the parallel component (p_z/p_0). When the oscillation amplitude increases, p_z/p_0 reduces in general, although the maximum value is always equal to unity. These results are expected from Eq. (7),

$$\frac{\langle p_x\rangle}{p_0}=0 \quad \text{and} \quad \frac{\langle p_z\rangle}{p_0}=\frac{1}{\sqrt{1+\theta_0^2k^2\tau^2}}.$$

Hence, on the average, $\langle p_z\rangle/p_0$ must decrease when we increase the oscillation amplitude θ_0.

Now, we examine the case for a constant amplitude θ_0 but varying frequency k. This is a realistic case, since in experiment, we can hardly increase the amplitude but we may easily change the frequency. In the right panel of Fig. 1, we choose $\theta_0=\pi/4$ and $k=1$ and 3. The results for constant k and constant θ_0 show similar time dependence. However, p_x and p_z show a larger variation in their magnitudes, if we increase the value of k. It should be remarked that we have

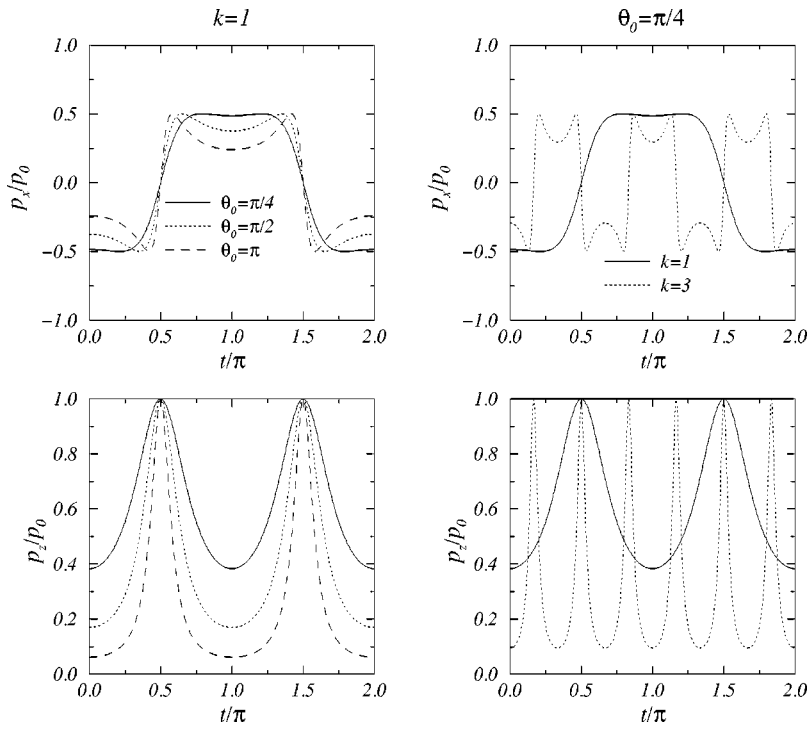

FIG. 2. The reduced dipole moment p_x/p_0 and p_z/p_0 plotted as a function of time for various frequency dependent τ ($\tau_0=2,\tau_\infty=1$).

assumed that the oscillation frequency is high so that the relaxation time is nearly constant during the motion.

V. DISCUSSION AND CONCLUSION

Here, a few comments on our results are in order. As our steady-state solution is general, one can extend the calculations to an arbitrary rotational motion. We have shown that the motion of particles reduces the strength of the dipole moment. It is natural to further calculate the interparticle force between two rotating spheres. We expect that the interparticle force will be reduced substantially because the force between parallel dipoles changes from attractive to repulsive when their orientation varies from the transverse to the longitudinal field case.

So far, our derivation of relaxation time is based on the mean-field theory. We may extend the Maxwell-Wagner theory to the polarization relaxation of oscillating particles. In this case, we should add a term $\rho_P \vec{v}$ to the polarization current density, where ρ_P is the polarized charge density and $\vec{v}=\vec{\omega}\times\vec{r}$ is the rotating velocity. However, it is not possible to convert the extra term into a dielectric constant and the generalization becomes more complicated due to the nonuniform polarized charge density inside the rotating spherical inclusions. We are currently examining the solution of the more complicated boundary-value problem.

The flow in ER fluid may be nonsteady in usual operation situations. But the prevailing situation in theory and simulation of ER fluids is to use formulas derived with respect to a steady flow. To remedy this drawback, we will endeavor to develop a calculation method for suspension hydrodynamics, and use it to study the interaction between particles and the oscillating fluid, and derive formulas of the force and torque exerted on particles for a suspension [15].

In this paper, hydrodynamic (HD) interaction effects have not been considered. However, electrorheological fluids are locally very concentrated suspensions and in considering dynamic effects, it seems that HD effects can be strong. This is a future problem.

In conclusion, we have investigated the problem of how the dipole moment of a dielectric sphere varies with time for an arbitrary rotational motion. We have developed a formalism for the rotational motion of the sphere and derived the relaxation time by using the mean-field theory. We have shown that the time-averaged steady-state dipole moment is along the field direction, but its magnitude is reduced by a factor that depends on the frequency of oscillation. As a result, the motion of particles reduces the ER effect. We further calculate the relaxation time based on the Maxwell-Wagner theory. The dependence of the reduction factor and the relaxation time on the angular velocity of rotation has also been discussed.

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