Nonlinear ac response of an electrorheological fluid

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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. When a nonlinear ER fluid is subjected to a sinusoidal (ac) field, the electrical response will in general consist of ac fields at frequencies of the higher-order harmonics. In this paper, a self-consistent formalism has been employed to compute the induced dipole moment for ER fluids in which the suspended particles have nonlinear characteristics, in an attempt to investigate the ac response of a nonlinear ER fluid.

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I. INTRODUCTION

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 rapid transition between the fluid and solid phases renders this material potentially important for technological applications.

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 have been reported by Klingenberg and co-workers [1]. Recently, the effect of nonlinear characteristics on the interparticle force has been analyzed in an ER suspension of nonlinear particles [2] and further extended to a nonlinear host medium [3]. The nonlinear characteristics are due to the field dependence of the dielectric constants of the materials used in ER fluids, which have a constitutive relation $\mathbf{D}=\epsilon\mathbf{E}+\chi\mathbf{E}^3$. This paper confirmed the previous theoretical results that the attractive force between two touching spheres varies almost linearly with the applied field strength [4].

A convenient method of probing the nonlinear characteristics is to measure the harmonics of the induced polarization under the application of a sinusoidal (ac) electric field [1]. The intrinsic nonlinear dielectric response of the materials should be the origin of the higher harmonics, especially when the applied field strength is high. The nonlinearity caused by the motion of the suspended particles alone under the applied field should not be the reason for the higher harmonics. When a nonlinear composite with nonlinear dielectric particles embedded in a host medium, or with a nonlinear host medium is subjected to a sinusoidal field, the electrical response in the composite will in general be a superposition of many sinusoidal functions [5]. It is natural to investigate the effects of nonlinear characteristics on the interparticle force in an ER fluid, which can be regarded as a nonlinear composite medium [6]. The strength of the nonlinear polarization is reflected in the magnitude of the harmonics.

In this paper, we will develop a self-consistent theory to calculate the ac response of a nonlinear ER fluid. Our theory goes beyond the simple point-dipole approximation and accounts for the multipole interaction between the polarized particles. The paper is organized as follows. In Sec. II, we calculate the dipole moment of a pair of polarized spheres of a nonlinear characteristics and extract its harmonic response. In Sec. III, we perform a series expansion of the local field inside the spheres from the self-consistent solution, in an attempt to obtain analytic expressions of the higher harmonics of the dipole moment. Numerical results are performed in Sec. IV to validate the analytic results. Discussions on the results will be given.

II. NONLINEAR POLARIZATION AND ITS HIGHER HARMONICS

We first examine the effect of a nonlinear characteristics on the induced dipole moment. We concentrate on the case where the suspended particles have a nonlinear dielectric constant, while the host medium has a linear dielectric constant ϵ_m. The nonlinear characteristics gives rise to a field-dependent dielectric coefficient [7]. In which case, the electric displacement electric-field relation inside the spheres is given by

$$\mathbf{D}_p=\epsilon_p\mathbf{E}_p+\chi_p\langle E_p^2\rangle\mathbf{E}_p=\tilde{\epsilon}_p\mathbf{E}_p, \tag{1}$$

where ϵ_p and χ_p are the linear coefficient and the nonlinear coefficient of the suspended particles, respectively.

This constitutes an approximation: the local field inside the particles is assumed to be uniform and the assumption is called the decoupling approximation [8]. It has been shown that such an approximation yields a lower bound for the accurate result for the local field [8]. We further assumed that both ϵ and χ are independent of frequency, which is a valid assumption for low-frequency processes in ER fluids. As a result, the induced dipole moment under an applied field $\mathbf{E}=E(t)\hat{z}$ is given by

$$\tilde{p}_0=\epsilon_m a^3\tilde{b}E(t), \tag{2}$$

where $\tilde{b}$ is the field-dependent dipolar factor and is given by

$$\tilde{b}=\frac{\tilde{\epsilon}_p-\epsilon_m}{\tilde{\epsilon}_p+2\epsilon_m}=\frac{\epsilon_p+\chi_p\langle E_p^2\rangle-\epsilon_m}{\epsilon_p+\chi_p\langle E_p^2\rangle+2\epsilon_m}. \tag{3}$$

When the polarized spheres approach one another, they will be further polarized by the mutual polarization effect. As a result, the point-dipole approximation breaks down and we must consider the multipole moments.

Let us consider a pair of nonlinear dielectric spheres of the same radius a, separated by a distance r. Each of them has a field-dependent dielectric coefficient $\tilde{\epsilon}_p$. By using the method of multiple images [9], we can deduce the total dipole moment of the spheres:

$$\tilde{p}_{\mathrm{T}}=\tilde{p}_0\sum_{n=0}^{\infty}(-\tilde{\tau})^n\left(\frac{\sinh\beta}{\sinh(n+1)\beta}\right)^3. \tag{4}$$

The factor $\tilde{\tau}$ is the field-dependent dielectric contrast given by

$$\tilde{\tau}=(\tilde{\epsilon}_p-\epsilon_m)/(\tilde{\epsilon}_p+\epsilon_m). \tag{5}$$

The subscript T denotes that the applied electric field is perpendicular to the line joining the centers of the particles (i.e., a transverse field). The parameter β is related to the separation between the particles: $\cosh\beta=r/2a=\sigma$, where σ is the reduced separation. For a longitudinal field, we replace $(-\tilde{\tau})$ by $(2\tilde{\tau})$. While we have shown the results for a pair of dielectric spheres of the same size, the above formula can be modified to calculate the total dipole moment of two dielectric spheres of different sizes [9]. We should remark that the present multiple images method is an approximation only. In fact there is a more complicated images method for a dielectric sphere [10]. However, we have validated the above expressions by comparing the analytic expressions with the numerical solution of the integral equation method [11].

When we apply a sinusoidal electric field, i.e., $E(t)=E_0\sin\omega t$, the induced dipole moment will vary with time sinusoidally. Due to the nonlinearity of the particles, the induced dipole moment will be a superposition of harmonics. In other words, we have

$$\tilde{p}_{\mathrm{T}}=p_{\omega}\sin\omega t+p_{3\omega}\sin 3\omega t+p_{5\omega}\sin 5\omega t+\cdots. \tag{6}$$

It is clear that only the harmonics of odd order survive due to the inversion symmetry of the dielectric media. Similarly, the local electric field inside the particles will also contain the higher harmonics:

$$\sqrt{\langle E_P^2\rangle}=E_{\omega}\sin\omega t+E_{3\omega}\sin 3\omega t+E_{5\omega}\sin 5\omega t+\cdots. \tag{7}$$

In what follows, we report results for the transverse field case only. The longitudinal field case is similar. In Sec. III, we will use the series expansion to obtain analytic expressions for the harmonics of the induced dipole moment. We will obtain the coefficient $p_{n\omega}$ as a power series of the applied field E_0.

III. SELF-CONSISTENT EVALUATION OF THE LOCAL FIELD

According to Eq. (4), the induced dipole moment of the dielectric spheres $\tilde{p}_T$ can be determined if we find the average local field $\langle E_p^2\rangle$. The electric field inside the spheres can be conveniently calculated by considering the effective nonlinear dielectric constant of a nonlinear composite in which the spheres are embedded in a host medium of much larger volume V. For a two-component composite, the effective nonlinear dielectric constant ϵ_e is given by [7]

$$\begin{aligned}\tilde{\epsilon}_e&=\frac{1}{E^2(t)V}\int_V\epsilon(\mathbf{r})|\mathbf{E}(\mathbf{r},t)|^2dV\\&=\frac{f\tilde{\epsilon}_p}{E^2(t)}\langle E_p^2\rangle+\frac{(1-f)\epsilon_m}{E^2(t)}\langle E_m^2\rangle,\end{aligned} \tag{8}$$

where f is the (infinitesimal) volume fraction of the particles. For a pair of spheres inside a transverse field, the effective nonlinear dielectric constant can be expressed as [7]

$$\tilde{\epsilon}_e=\epsilon_m+3f\epsilon_m(\tilde{b}\tilde{p}_T/\tilde{p}_0). \tag{9}$$

The electric field inside the spheres can be calculated by using Eq. (9):

$$\langle E_p^2\rangle=\frac{1}{f}E^2(t)\frac{\partial\tilde{\epsilon}_e}{\partial\tilde{\epsilon}_p}. \tag{10}$$

The right-hand side of Eq. (10) depends on the local field itself. Hence for nonlinear characteristics [Eq. (1)], Eq. (10) must be solved self-consistently [8]. The local field inside the spheres as well as the dipole moment of the spheres [Eq. (4)] can be determined.

It remains to examine how the higher harmonics of $\tilde{p}_T$ depends, the nonlinearity. We expand $\tilde{p}_T$ and $\langle E_p^2\rangle$ into a Taylor expansion:

$$\tilde{p}_T=\epsilon_m a^3E(t)\sum_{s=0}^{\infty}a_s(\chi_p\langle E_p^2\rangle)^s, \tag{11}$$

$$\chi_p\langle E_p^2\rangle=3\epsilon_m\chi_pE^2(t)\sum_{s=0}^{\infty}c_s(\chi_p\langle E_p^2\rangle)^s, \tag{12}$$

where the expansion coefficient a_s is given by

$$a_s=\frac{1}{s!}\frac{\partial^s}{\partial\tilde{\epsilon}_p^s}\left[\tilde{b}\sum_{n=0}^{\infty}(-\tilde{\tau})^n\left(\frac{\sinh\beta}{\sinh(n+1)\beta}\right)^3\right]_{\tilde{\epsilon}_p=\epsilon_p}, \tag{13}$$

and $c_s=(s+1)a_{s+1}$. The expansion coefficients do not depend on the applied field. In the case of a weak nonlinearity, i.e., $\chi_pE^2(t)\ll 1$, we can rewrite Eqs. (12) and (11), keeping only the lowest orders of $\chi_pE^2(t)$ and $\chi_p\langle E_p^2\rangle$:

$$\chi_p\langle E_p^2\rangle=3\epsilon_m\chi_pE^2(t)(c_0+c_1\chi_p\langle E_p^2\rangle+\cdots).$$

The first term gives the local field inside a linear dielectric particle. Similarly, the induced dipole moment is given by

$$\begin{aligned}\tilde{p}_T&=\epsilon_m a^3a_0E(t)+3\epsilon_m^2a^3a_1^2\chi_pE^3(t)+\cdots\\&=K_1E(t)+K_3E^3(t)+\cdots.\end{aligned} \tag{14}$$

It should be remarked that, from Eq. (11), $K_1E(t)$ is the linear dipole moment p_T

$$p_T=\epsilon_m a^3 b E(t)\sum_{n=0}^{\infty}(-\tau)^n\left(\frac{\sinh\beta}{\sinh(n+1)\beta}\right)^3. \tag{15}$$

Let us consider a sinusoidal applied electric field $E(t)=E_0\sin\omega t$. By using the identity $4\sin^3\omega t=3\sin\omega t-\sin 3\omega t$, we can expand $E^3(t)$ in terms of the first and the third harmonics. By comparing Eq. (6) with Eq. (14), we find

$$p_\omega=K_1E_0+\tfrac{3}{4}K_3E_0^3 \quad \text{and} \quad p_{3\omega}=-\tfrac{1}{4}K_3E_0^3.$$

The above results show that the induced dipole moment must include the higher harmonics [6]. Furthermore, the results show that p_ω also depends on E_0^3. This is a nontrivial result as it implies that the first harmonic of the induced dipole moment depends on the strength of the nonlinearity. Concomitantly, the higher harmonics should become more significant as E_0 gets higher. In the case of a higher applied field, we must include even higher-order terms [i.e., higher powers of $E(t)$] in the expansion of $\tilde{p}_T$:

$$\tilde{p}_T=K_1E(t)+K_3E^3(t)+K_5E^5(t)+\cdots.$$

Again, by considering the identity $16\sin^5\omega t=10\sin\omega t-5\sin 3\omega t+\sin 5\omega t$, the harmonics are given by

$$p_\omega=K_1E_0+\tfrac{3}{4}K_3E_0^3+\tfrac{10}{16}K_5E_0^5,$$

$$p_{3\omega}=-\tfrac{1}{4}K_3E_0^3-\tfrac{5}{16}K_5E_0^5, \quad p_{5\omega}=\tfrac{1}{16}K_5E_0^5.$$

Consequently, in the case of nonlinear ac response, the convergence of the series expansion is questionable and a self-consistent formalism is needed. From Eqs. (12), (11), and (14), we find that

$$\tilde{p}_T/p_0=F(\chi_pE_0^2) \quad \text{and} \quad \chi_p\langle E_p^2\rangle=G(\chi_pE_0^2), \tag{16}$$

where F and G are functions of a single variable. A similar conclusion can be drawn for p_ω and $p_{3\omega}$. This demonstrates that we can use $\chi_p\langle E_0^2\rangle$ as a natural variable of the strength of nonlinearity in the nonlinear composite problem [12].

IV. NUMERICAL RESULTS

In this section, we perform numerical calculations to investigate the effects of nonlinear characteristics on the harmonics of the induced dipole moment and the local electric field. As shown in Sec. III, the induced dipole moment of the dielectric spheres is affected by three factors: the strength of the applied field, the nonlinear dielectric coefficient of the particles, as well as the linear dielectric constants of the particles and the host medium. Without loss of generality, we let $a=1$ and $\epsilon_m=1$. In order to emphasize the effect of mutual polarization, we use a small reduced separation $\sigma=1.1$ and compare the results of the multiple images dipole (MID) case with that of the point dipole (PD) case. From Eq. (16), we can use $\chi_pE_0^2$ as the variable to plot the numerical results.

Before showing the numerical results, we consider the simple case of an isolated sphere. For well-separated nonlinear dielectric spheres in the dilute limit where the mutual polarization effect can be neglected, the dipole moment is well approximated by that of a single sphere. In which case, the local field inside the particle can be solved exactly [12]:

$$\chi_p\langle E_p^2\rangle=3\epsilon_m\chi_pE^2(t)\frac{3\epsilon_m}{(\tilde{\epsilon}_p+2\epsilon_m)^2}=\frac{9\epsilon_m^2\chi_pE^2(t)}{(\epsilon_p+\chi_p\langle E_p^2\rangle+2\epsilon_m)^2}. \tag{17}$$

The local field can be readily solved and the solution obeys Eq. (16). The solution implies that the effect of the nonlinear characteristics is important only if the linear dielectric coefficient of the particle is small compared with that of the host. In the case of ER fluids, the opposite limit, $\epsilon_p>\epsilon_m$, holds, and we have

$$E_\omega=3\epsilon_mE_0/(\epsilon_p+2\epsilon_m)+\cdots,$$

which means that the first harmonic varies linearly with E_0. In other words, a linear relation between E_ω and E_0 is an indication of a weak nonlinearity.

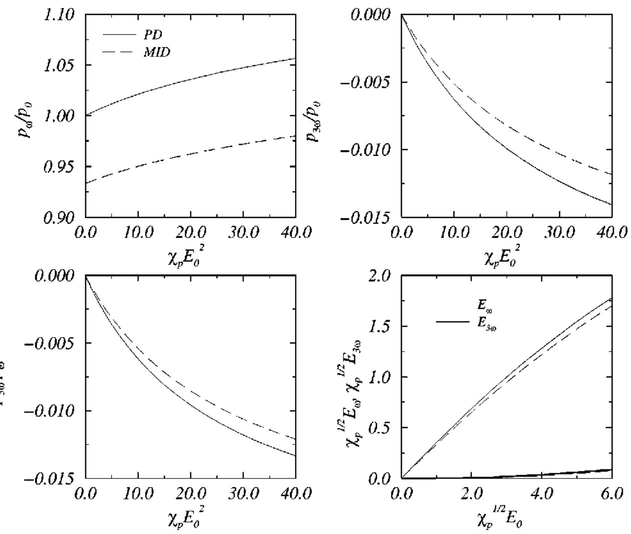

FIG. 1. The harmonics of the induced dipole moment and the local electric field inside a pair of nonlinear dielectric spheres, with $\epsilon_p=10$ and $\epsilon_m=1$. The nonlinear characteristics has a small effect on the nonlinear response as reflected in the graph of $\sqrt{\chi_p}E_\omega$ vs $\sqrt{\chi_p}E_0$.

In Fig. 1, we choose $\epsilon_p=10$ and plot the normalized harmonics p_ω/p_0 and $p_{3\omega}/p_0$ vs $\chi_pE_0^2$, where $p_0=\epsilon_m a^3 b E_0$. The ratio $p_{3\omega}/p_\omega$ is also plotted. Furthermore, E_ω and $E_{3\omega}$ are plotted with $\sqrt{\chi_p}E_0$. We find that the first harmonic of the dipole moment is close to the linear dipole moment, i.e., $p_\omega\approx p_0$, even when the applied field is high. On the other hand, the third harmonic is small compared to the first harmonic. The dielectric contrast between the particles and the host is relevant for determining the magnitude of the nonlinear response, namely, a smaller dielectric contrast gives a larger nonliner response in the case of a nonlinear particle, results that are in accord with Ref. [7]. Next, we examine the harmonics of the local field (E_ω and $E_{3\omega}$). The first har-

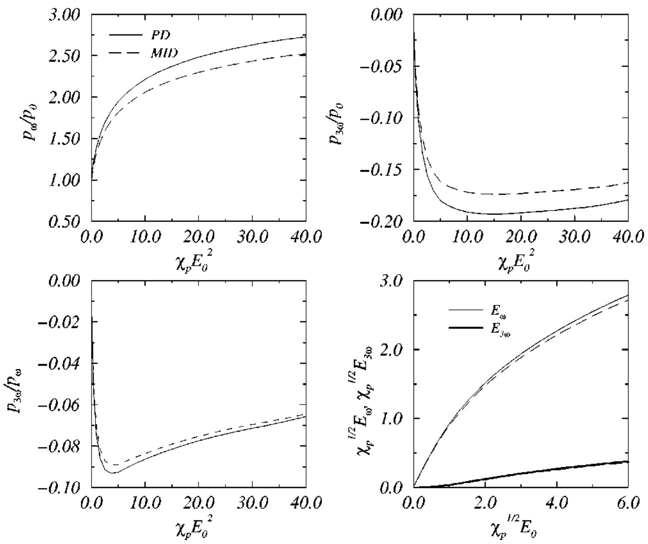

FIG. 2. The same quantities as plotted in Fig. 1, but with $\epsilon_p=2$ and $\epsilon_m=1$. The ratio $p_{3\omega}/p_\omega$ attains a maximum magnitude when $\chi_p E_0^2 \approx 4$. The nonlinearity has a strong effect on the third harmonic $p_{3\omega}$.

monic $\sqrt{\chi_p}E_\omega$ varies almost linearly with $\sqrt{\chi_p}E_0$. These results show that the nonlinearity is not significant for the present material parameters ($\epsilon_p=10, \epsilon_m=1$). A similar conclusion can be drawn for both the PD and the MID cases. The major difference between the PD and the MID cases is that the mutual polarization effect always reduces the dipole moment.

We repeat the same calculations with a smaller dielectric constant $\epsilon_p=2$ (Fig. 2). We found that the nonlinear characteristics have a more significant effect on the various quantities. Namely, the first harmonic of the local field E_ω shows a stronger nonlinear behavior. We observe an interesting result that the ratio $p_{3\omega}/p_\omega$ attains a maximum magnitude at $\chi_p E_0^2 \approx 4$. This behavior holds for both the PD and the MID cases.

V. DISCUSSION AND CONCLUSION

Here a few comments on our results are in order. In the present paper, we have examined the case of nonlinear particles suspending in a linear host. We may extend our considerations to a nonlinear host medium [3]. In this case, preliminary results show that qualitatively similar yet more complex behaviors in the ac response have been observed.

So far, we have not considered the frequency dependence of the particle dielectric constant. In a realistic situation, the dielectric constant of the particles can decrease with the increase of the frequency. For simplicity, we may adopt the Debye relaxation expression for ϵ_p. Preliminary results show that the ratio of the third to first harmonic decreases with frequency, results that are in accord with recent experimental data of Ref. [1].

As we have obtained the expression for the induced dipole moments, we may take a step forward to calculate the interparticle force via the energy approach [9]. We will find a time-dependent force $F(t)$ but only its time average could be measured during an experiment. From the energy approach [9],

$$\langle F_T(t)\rangle = \partial \langle E(t) p_T(t)\rangle / \partial r .$$

Since $E(t)$ contains the first harmonic only, the time average $\langle E(t)p_T(t)\rangle$ will be proportional to the first harmonic p_ω, to which all higher-order nonlinearities contribute. We believe that the interparticle force in the ac case should differ significantly from that of the dc case because, as we have shown, the nonlinearity enters into the composite problem in a nontrivial way. Results of the interparticle force will be published elsewhere [13].

In conclusion, we have considered the effects of a nonlinear characteristics on the ER fluid under the influence of a sinusoidal applied field. We have calculated the harmonic components of the induced dipole moment as well as the local electric field. We have also examined the conditions for obtaining large ac responses in ER fluids.

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