Nonlinear ER effects in an ac applied field

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Abstract

The electric field used in most electrorheological (ER) experiments is usually quite high, and nonlinear ER effects have been theoretically predicted and experimentally measured recently. A direct method of measuring the nonlinear ER effects is to examine the frequency dependence of the same effects. For a sinusoidal applied field, we calculate the ac response which generally includes higher harmonics. In this work, we develop a multiple image formula, and calculate the total dipole moments of a pair of dielectric spheres, embedded in a nonlinear host. The higher harmonics due to the nonlinearity are calculated systematically.

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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 and columns of particles across the electrodes in the direction of the applied field. The rapid transition between the fluid and solid phases renders this material the potential of important 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 coworkers [1]. Recently, the effect of a nonlinear characteristic on the interparticle force has been analyzed in an ER suspension of nonlinear particles [2] and further extended to a nonlinear host medium [3]. These work confirmed the previous theoretical results that the attractive force between two touching spheres can have a quasi-linear dependence [4].

A convenient method of probing the nonlinear characteristic is to measure the harmonics of the induced polarization under the application of a sinusoidal (ac) electric field [1]. 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 a nonlinear characteristic 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 work, 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.
II. NONLINEAR POLARIZATION AND ITS HIGHER HARMONICS

We first consider an isolated spherical particle of radius $a$ and dielectric constant $\epsilon_p$. The sphere is placed in a host medium of dielectric constant $\epsilon_m$. When a time-dependent electric field $\mathbf{E} = E(t)\hat{z}$ is applied. The particle will be polarized and its surface charge will contribute to it a dipole moment $p_0 = \epsilon_m a^3 \beta E(t)$, where $\beta$ is the dipolar factor given by:

$$\beta = \frac{(\epsilon_p - \epsilon_m)}{(\epsilon_p + 2\epsilon_m)}.$$ 

In ER fluids, when the particles get close to one another, they do not behave as point dipole and we have to consider the mutual polarization between the particles.

Next, we examine the effect of a nonlinear characteristic on the induced dipole moment. We concentrate on the case where the host medium have a nonlinear dielectric constant, while the suspended particles are linear. The nonlinear characteristic gives rise to a field-dependent dielectric coefficient. In other words, the electric displacement-electric field relation in the host medium is given by:

$$\mathbf{D}_m = \epsilon_m \mathbf{E}_m + \chi_m \langle E_m^2 \rangle \mathbf{E}_m = \bar{\epsilon}_m \mathbf{E}_m,$$ 

where $\chi_m$ is the nonlinear coefficient of the particles. We have assumed that the average field inside the host medium is uniform. This is called the decoupling approximation \[8\]. It has been shown that such an approximation yields a lower bound for the accurate result \[8\]. Furthermore, in ER fluids the frequency of the applied field is low; we can assume both $\epsilon$ and $\chi$ are independent of frequency. As a result, the induced dipole moment is field-dependent and is given by:

$$\bar{p}_0 = \bar{\epsilon}_m a^3 \bar{\beta} E(t),$$ 

where $\bar{\beta}$ is the field-dependent dipolar factor and is given by:

$$\bar{\beta} = \frac{\epsilon_p - \bar{\epsilon}_m}{\epsilon_p + 2\bar{\epsilon}_m} = \frac{\epsilon_p - (\epsilon_m + \chi_m \langle E_m^2 \rangle)}{\epsilon_p + 2(\epsilon_m + \chi_m \langle E_m^2 \rangle)}.$$ 

When the polarized spheres approach to 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 dielectric coefficient \( \epsilon_p \). By using the method of multiple images \([9,10]\), we deduce the total dipole moment of the spheres:

\[
\tilde{p}_T = \tilde{p}_0 \sum_{n=0}^{\infty} (-\tilde{\beta})^n \left( \frac{\sinh \alpha}{\sinh(n+1)\alpha} \right)^3.
\] (4)

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 \( \alpha \) is related to the separation between the particles: \( \cosh \alpha = r/2a = \sigma \), where \( \sigma \) is the reduced separation. For a longitudinal field, we change the factor from \( (-\tilde{\beta}) \) to \( (2\tilde{\beta}) \). The above formula can be generalized to account for a pair of dielectric spheres of the different sizes \([9]\). The multiple images method was widely adopted \([2,3,11]\) to calculate the total dipole moment as well as the interparticle force.

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 \([12,13]\), which gives the exact image dipole moment of a dielectric sphere that placed in front of a point dipole. However, the numerical results of the present version of formula agree with the numerical solution of an integral equation method \([14,15]\).

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:

\[
\tilde{p}_T = p_\omega \sin \omega t + p_{3\omega} \sin 3\omega t + p_{5\omega} \sin 5\omega t + \cdots
\] (5)

Due to the inversion symmetry of the dielectric media, only the harmonics of odd order survive. The coefficient \( p_{n\omega} \) of the \( n \)th harmonic is given by:

\[
p_{n\omega} = \frac{\omega}{\pi} \int_0^{2\pi/\omega} \tilde{p}_T \sin n\omega t dt.
\] (6)

In what follows, we report results for the transverse field case only. The longitudinal field case is similar. In the next section, 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 AVERAGE FIELD

According to Eqs. (4), the induced dipole moment of the dielectric spheres $\tilde{p}_T$ can be determined if we find the average field $\langle E_m^2 \rangle$. The electric field inside the host medium can be conveniently calculated by considering the effective nonlinear dielectric constant of a two-component composite. For a two-component composite, the effective nonlinear dielectric constant $\tilde{\varepsilon}_e$ is given by [7]:

$$\tilde{\varepsilon}_e = \frac{1}{E^2(t)V} \int_V \varepsilon(r)|E(r,t)|^2 dV = \frac{f \varepsilon_p}{E^2(t)} \langle E_p^2 \rangle + \frac{(1-f)\varepsilon_m}{E^2(t)} \langle E_m^2 \rangle,$$

(7)

where $f$ is the volume fraction of the particles. For a pair of spheres inside a transverse field, the effective nonlinear Dielectric constant can be expressed as [9]:

$$\tilde{\varepsilon}_e = \tilde{\varepsilon}_m + 3f\tilde{\varepsilon}_m \left( \frac{\tilde{\beta} \tilde{p}_T}{\tilde{p}_0} \right).$$

(8)

The average field of the host medium can be calculated by using Eq.(8):

$$\langle E_m^2 \rangle = \frac{1}{1-f} \frac{E^2(t)}{\partial \tilde{\varepsilon}_e}.$$

(9)

For a nonlinear characteristic [Eq.(1)], we solve Eq.(4) self-consistently [8]. The average field inside the host medium 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 first normalize the material parameters with the linear dielectric coefficient of the host $\varepsilon_m$:

$$\varepsilon_p' = \frac{\varepsilon_p}{\varepsilon_m}, \quad \chi_m' = \frac{\chi_m}{\varepsilon_m}, \quad \tilde{\varepsilon}_m' = \frac{\tilde{\varepsilon}_m}{\varepsilon_m} = 1 + \chi_m' \langle E_m^2 \rangle.$$

(10)

We then expand $\langle E_m^2 \rangle$ into a Taylor expansion:

$$\chi_m' \langle E_m^2 \rangle = \frac{\chi_m' E_m^2(t)}{1-f} + \frac{3f}{1-f} \chi_m' E_m^2(t) \sum_{s=0}^{\infty} c_s \left( \chi_m' \langle E_m^2 \rangle \right)^s,$$

(11)
where the expansion coefficient $c_s$ is given by:

$$
c_s = \frac{1}{s!} \frac{\partial^{s+1}}{\partial \tilde{e}_m^{s+1}} \left[ \tilde{e}_m' \tilde{\beta} \sum_{n=0}^{\infty} (-\tilde{\beta})^n \left( \frac{\sinh \alpha}{\sinh(n+1)\alpha} \right)^3 \right]_{\tilde{e}_m=1}. \quad (12)
$$

Hence the expansion coefficients do not depend on the applied field. Similarly, we expand $\tilde{p}_T$ into a Taylor expansion:

$$
\tilde{p}_T = \epsilon_m a^3 E(t) \sum_{s=0}^{\infty} a_s \left( \chi'_m \langle E^2_m \rangle \right)^s,
$$

where

$$
a_s = \frac{1}{s!} \frac{\partial^s}{\partial \tilde{e}_m^s} \left[ \tilde{e}_m' \tilde{\beta} \sum_{n=0}^{\infty} (-\tilde{\beta})^n \left( \frac{\sinh \alpha}{\sinh(n+1)\alpha} \right)^3 \right]_{\tilde{e}_m=1}. \quad (14)
$$

It is easy to show that: $c_s = (s+1)a_{s+1}$. In the case of a weak nonlinearity, i.e. $\chi'_m E^2(t) \ll 1$, we can rewrite Eqs.(11) and (13), keeping only the lowest orders of $\chi'_m E^2(t)$ and $\chi'_m \langle E^2_m \rangle$:

$$
\chi'_m \langle E^2_m \rangle = \frac{\chi'_m E^2(t)}{1-f} + \frac{3f}{1-f} \chi'_m E^2(t)(c_0 + c_1 \chi'_m \langle E^2_m \rangle + \cdots).
$$

The first order term of $\chi'_m E^2(t)$ gives the average field inside a linear dielectric host medium. Similarly, the induced dipole moment is given by:

$$
\tilde{p}_T = \epsilon_m a^3 a_0 E(t) + \epsilon_m a^3 a_1 \frac{\chi'_m E^3(t)}{1-f} + \frac{3f}{1-f} \epsilon_m a^3 a_1^2 \chi'_m E^3(t) + \cdots
= K_1 E(t) + K_3 E^3(t) + \cdots.
$$

It should be remarked that, from Eq.(13), $K_1 E(t)$ is the linear dipole moment $p_T$:

$$
p_T = \epsilon_m a^3 \beta E(t) \sum_{n=0}^{\infty} (-\beta)^n \left( \frac{\sinh \alpha}{\sinh(n+1)\alpha} \right)^3. \quad (16)
$$

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.(5) with Eq.(15), we find:

$$
p_\omega = K_1 E_0 + \frac{3}{4} K_3 E^3_0 \quad \text{and} \quad p_{3\omega} = -\frac{1}{4} K_3 E^3_0.
$$

The above results show that the induced dipole moment must include the higher harmonics $E^3_0$. Furthermore, the results show that $p_\omega$ also depends on $E^3_0$. 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 \( \tilde{p}_T \):

\[
\tilde{p}_T = K_1 E(t) + K_3 E^3(t) + K_5 E^5(t) + \cdots.
\]

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:

\[
\begin{align*}
p_{\omega} &= K_1 E_0 + \frac{3}{4} K_3 E_0^3 + \frac{10}{16} K_5 E_0^5, \\
p_{3\omega} &= -\frac{1}{4} K_3 E_0^3 - \frac{5}{16} K_5 E_0^5, \\
p_{5\omega} &= \frac{1}{16} K_5 E_0^5.
\end{align*}
\]

Consequently, in the case of nonlinear ac response, the convergence of the series expansion is questionable and a self-consistent formalism is needed. The self-consistent equation [Eq.(19)] can help to solve for the average field. From Eq.(11), (13) and (15), we find that:

\[
\frac{\tilde{p}_T}{p_0} = F(\chi'_m E_0^2) \quad \text{and} \quad \chi'_m \langle E_m^2 \rangle = G(\chi'_m E_0^2),
\]

where \( F \) and \( G \) are functions of a single variable, and \( p_0 = \epsilon_m a^3 \beta E_0 \); this is the linear dipole moment of an isolated dielectric particle under a time-independent applied field \( E_0 \). Similar conclusion can be drawn for \( p_{\omega} \) and \( p_{3\omega} \). This demonstrates that we can use \( \chi'_m E_0^2 \) as a natural variable in the nonlinear composite problem [16].

### IV. NUMERICAL RESULTS

In this section, we perform numerical calculations to investigate the effects of a nonlinear characteristic on the harmonics of the induced dipole moment and the average electric field. As shown in section III, the induced dipole moment of the dielectric spheres is affected by three factors: the strength of the applied field \( (E_0) \), the relative nonlinear dielectric coefficient of the host medium \( (\chi'_m) \) and the relative linear dielectric constants of the particles \( (\epsilon'_p) \).
In order to emphasize the effect of mutual polarization, we use a small reduced separation \( \sigma = 1.1 \). From Eq.(17), we can use \( \chi'_m E_0^2 \) as the variable to plot the numerical results.

Before showing the numerical results, we consider the simple case of an isolated sphere. In this case, the average field inside the host medium can be solved exactly \[3,16\]:

\[
\chi'_m \langle E_m^2 \rangle = \left( \frac{1}{1 - f} + \frac{3f \epsilon'_p - 2\epsilon_p \epsilon'_m - 2\epsilon_m^2}{1 - f} \right) \chi'_m E^2(t).
\] (18)

When the applied field is small, the effect of nonlinearity is negligible. The average field is dominated by the first part of the above equation. Since the volume fraction \( f \) have to be small \[7\], the average field can be well approximated by:

\[
\langle E_m^2 \rangle \approx E^2(t).
\] (19)

For a sinusoidal applied field \( E(t) = E_0 \sin \omega t \), we have:

\[
E_\omega \approx E_0,
\]

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

In Fig.1, we plot the first and the third harmonics of the average field, with \( \epsilon'_p = 2 \) \((\beta = 1/4)\) and \( \epsilon'_p = 10 \) \((\beta = 1/4)\), respectively. The volume fraction has a small influence on the nonlinear effects. The harmonics are plotted against \( \sqrt{\chi_m} E_0 \). The first harmonic \( \sqrt{\chi_m} E_\omega \) varies almost linearly with \( \sqrt{\chi_m} E_0 \). Moreover, the magnitude of the third harmonics is small compared with the first harmonics. These results show that the average field can be approximated by Eq.(19).

The numerical results of the average field suggest that the average field varies almost linearly with the applied field. However, as suggested by Eq.(15), the dipole moment does not vary linearly with the applied field. In Fig.2, we plot the first and the third harmonics of the induced dipole moment. We divide the harmonics by \( p_0 = \epsilon_m a^3 \beta E_0 \), which is the linear dipole moment of an isolated dielectric particle under a time-independent applied field \( E_0 \). In Fig.3, we find that the first harmonics changes from positive to negative. This is because
as the magnitude of the applied field increase, the field-dependent dipolar factor $\tilde{\beta}$ changes from positive to negative. Hence the first harmonics changes its sign.

Next we examine the third harmonics of the dipole moment. The ratio $p_{3\omega}/p_0$ increase with the applied field. The third harmonics is an evidence of nonlinear effects. The ratio increase significantly when the dielectric contrast between the particles and the host is small. In other words, a smaller dielectric contrast gives a larger nonlinear response.

**DISCUSSION AND CONCLUSION**

Here a few comments on our results are in order. In the present study, we have examined the case of linear particles suspending in a nonlinear host. The case of nonlinear particles suspending in a linear host has already been studied by similar formalism [17].

So far, we have not considered the frequency dependence of the dielectric constants of the host medium and the particles. In a realistic situation, the dielectric constants can decrease with the increase of the frequency. For simplicity, we may adopt the Debye relaxation expression.

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],

$$\langle F_T(t) \rangle = \frac{\partial \langle E(t)p_T(t) \rangle}{\partial r}.$$ 

We believe that the interparticle force in the ac case should differ significantly from that of the dc case of nonlinear ER fluids 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 [18].

In conclusion, we have considered the effects of a nonlinear characteristic 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 average electric field. We have also examined the conditions for obtaining large ac response in ER fluids.

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FIGURES

FIG. 1. The first and the third harmonics of the average field, with $\epsilon'_p = 2 \ (\beta = 1/4)$ and $\epsilon'_p = 10 \ (\beta = 1/4)$, respectively. The volume fraction has a small influence on the nonlinear effects.

FIG. 2. The first and the third harmonics of the induced dipole moment. The material parameters are chosen as the same as in Fig. 1. The nonlinear effects are significant when the dielectric contrast is small.
