

NONLINEAR ALTERNATING CURRENT RESPONSES IN ELECTRORHEOLOGICAL FLUIDS: DYNAMIC EFFECTS

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By using a perturbation approach, we investigate dynamic effects on nonlinear alternating current (ac) responses in electrorheological (ER) fluids under an ac or a direct current electric field. We show that the dynamic effect due to a shear flow plays a significant role in the responses. Our results can be well interpreted in the dielectric dispersion spectral representation, and they offer a convenient method to determine the relaxation time and rotation velocity of ER particles by measuring the nonlinear ac responses.

Keywords: Nonlinear; dynamic.

1. Introduction

An electrorheological (ER) fluid^{1,2} contains polarizable particles suspended in a liquid of low dielectric constant. The rapid field-induced aggregation and the large anisotropy of ER fluids render these materials potentially important for applications. In a realistic situation, the fluid shear flow will set the particles in both translational and rotational motions^{3,4,5}. Wan *et al.*^{5,6} theoretically investigated such kind of rotation and pointed out that the rotation-induced displacement of the polarization surface charge reduces the interaction forces between the ER particles. Most recently, this was demonstrated in experiments⁷.

When a suspension consisting of dielectric particles having nonlinear characteristics is subjected to an alternating current (ac) field with angular frequency ω , the electric response will generally consist of ac fields at frequencies of the higher-order harmonics^{8,9,10,11}.

In this work (see Ref. 12), we shall develop a perturbation approach^{9,13} to investigate the dynamic effect on nonlinear ac responses in ER fluids under electric field.

2. Formalism

2.1. Without dispersion

We first consider a particle in the ER fluids under an applied external ac electric field $\mathbf{E}_{f0} = E_0 \cos(\omega t) \hat{\mathbf{Z}} \equiv E_0 \cos(\omega t)$, its induced dipole moment is given by: $\vec{p}_0 = p_0 \hat{\mathbf{Z}}$. The shear flow in the ER suspension will give the particle a torque, which leads it to get a rotational motion about its center. We assume that the angular velocity caused by shear flow is $\omega_1 \hat{\mathbf{Y}}$. The change of the dipole moment and the relaxation of various kinds have to be balanced against each other, resulting in a steady-state dipole moment $\vec{p}_0$, which deviates from the equilibrium dipole moment $\vec{p}_0$. Wan *et al.* gave the rate of change of the dipole moment in their work^{5,6}:

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

where τ is a relaxation time. When a steady state is reached, we obtain

$$\vec{p} = \frac{\vec{p}_0 + \tau(\vec{\omega} \times \vec{p}_0)}{1 + (\omega\tau)^2}. \quad (2)$$

And it can be shown that the angle between $\vec{p}_0$ and $\vec{p}$ is equal to ϕ and the magnitude of $\vec{p}$ is given by $|\vec{p}| = p_0 \cos \phi$ where ϕ is given by $\phi = \omega\tau$.

Now, we try to solve the problem in another way. If we rotate with the particle, the particle will seem to be at rest, and it feels an electric field written as

$$\mathbf{E}_f = \frac{1}{2} e^{-i\omega_1 t} (e^{-i\omega t} + e^{i\omega t}) \mathbf{E}_0. \quad (3)$$

While the suspended particles have a nonlinear dielectric constant, the nonlinear constructive relation between the electric displacement $\mathbf{D}_1$ and the local electric field $\mathbf{E}_1$ is defined as

$$\mathbf{D}_1 = \epsilon_1 \mathbf{E}_1 + \chi |\mathbf{E}_1|^2 \mathbf{E}_1 = \hat{\epsilon}_1 \mathbf{E}_1, \quad (4)$$

where ϵ_1 denotes the linear dielectric constant of a suspended particle, and χ is the third-order nonlinear coefficient of the particle. It is worth noting that owing to the spherical symmetry under consideration, even-order nonlinearity disappears naturally. For convenience, we adopt the definition $|\mathbf{E}_1|^2 = \mathbf{E}_1 \cdot \mathbf{E}_1 \equiv E_1^2$. Throughout the work, only weak nonlinearity is considered. Next, the local electric field inside a particle is given by

$$\mathbf{E}_1 = \frac{3\epsilon_2}{\epsilon_1 + \chi E_1^2 + 2\epsilon_2} \mathbf{E}_f. \quad (5)$$

Here ϵ_2 represents the (linear) dielectric constant of the host medium (e.g., silicone oil). Owing to the small nonlinear coefficient χ , we can expand E_1 by taking χE_1^2 as a small perturbation. In the mean time, we shall ignore the smaller terms. Thus, we get

$$\mathbf{E}_1 = \frac{3e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})\epsilon_2}{2(\epsilon_1 + 2\epsilon_2)}\mathbf{E}_0 - \frac{3e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})\epsilon_2\chi E_1^2}{2(\epsilon_1 + 2\epsilon_2)^2}\mathbf{E}_0. \quad (6)$$

And the first term of the right part of Eq. (6) is the linear part.

On the other hand, the induced dipole moment

$$\mathbf{P} = \epsilon_2 \frac{\epsilon_1 + \chi E_1^2 - \epsilon_2}{\epsilon_1 + \chi E_1^2 + 2\epsilon_2} a^3 \mathbf{E}_f \quad (7)$$

can be treated in the same way. In Eq. (7) a denotes the radius of the particle. Then we expand $\mathbf{P}(\omega \neq 0)$ by taking χE_1^2 as a perturbation and ignore the smaller terms. As a result, we obtain

$$\mathbf{P} = \frac{a^3 e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})(\epsilon_1 - \epsilon_2)\epsilon_2}{2(\epsilon_1 + 2\epsilon_2)}\mathbf{E}_0 + \frac{3a^3 e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})\epsilon_2^2\chi E_1^2}{2(\epsilon_1 + 2\epsilon_2)^2}\mathbf{E}_0. \quad (8)$$

Because the linear term (namely, the first term) of E_1 in the right-hand side of Eq. (6) is much larger than the second one (i.e., nonlinear part), we can introduce it as E_1 into Eq. (8). Then we obtain

$$\begin{aligned} P = & P_{\omega+\omega_1} e^{-it(\omega+\omega_1)} + P_{\omega-\omega_1} e^{it(\omega-\omega_1)} + P_{3\omega+3\omega_1} e^{-3it(\omega+\omega_1)} \\ & + P_{3\omega-3\omega_1} e^{3it(\omega-\omega_1)} + P_{\omega+3\omega_1} e^{-it(\omega+3\omega_1)} + P_{\omega-3\omega_1} e^{it(\omega-3\omega_1)}, \end{aligned} \quad (9)$$

where the harmonic coefficients of various of frequencies are given by

$$\begin{aligned} P_{\omega\pm\omega_1} &= \frac{a^3(\epsilon_1 - \epsilon_2)\epsilon_2 E_0}{2(\epsilon_1 + 2\epsilon_2)}, \\ P_{3\omega\pm 3\omega_1} &= \frac{27a^3\epsilon_2^4\chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4}, \\ P_{\omega\pm 3\omega_1} &= \frac{81a^3\epsilon_2^4\chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4}. \end{aligned} \quad (10)$$

If $\omega = 0$, we can easily sum the terms of the same frequency up. So we achieve

$$P(\omega=0) = P_{\omega_1} e^{-it\omega_1} + P_{3\omega_1} e^{-3it\omega_1}, \quad (11)$$

where

$$\begin{aligned} P_{\omega_1} &= \frac{a^3(\epsilon_1 - \epsilon_2)\epsilon_2 E_0}{\epsilon_1 + 2\epsilon_2}, \\ P_{3\omega_1} &= \frac{27a^3\epsilon_2^4\chi E_0^3}{(\epsilon_1 + 2\epsilon_2)^4}. \end{aligned} \quad (12)$$

Comparing Eq. (9) to Eq. (11), it is evident that harmonic signals can be detected at more frequencies due to the coupling of the frequencies ω and ω_1 . Since we can detect each harmonic coefficient in experiment, it is easy to determine the angular velocity caused by shear flow with Eq. (10) as long as the external field frequency ω is known.

2.2. With dispersion

Until now, the dielectric constant ϵ_1 and ϵ_2 which have been used are both real numbers, that is, we have not taken into account the relaxation. To include the relaxation, one is allowed to see them as complex numbers^{14,15}, and they can be described as

$$\begin{aligned}\epsilon_1 &= \bar{\epsilon}_1 - i\bar{\sigma}_1/\omega_0, \\ \epsilon_2 &= \bar{\epsilon}_2 - i\bar{\sigma}_2/\omega_0,\end{aligned}\quad (13)$$

where both $\bar{\epsilon}$ and $\bar{\sigma}$ are real numbers, and they represent the real dielectric constant and the conductivity of the particle, respectively. In Eq. (13), ω_0 denotes the angular frequency whose details can be found below. In this case, the induced dipole moment should be¹⁶

$$\mathbf{P} = \bar{\epsilon}_2 \frac{\epsilon_1 + \chi E_1^2 - \epsilon_2}{\epsilon_1 + \chi E_1^2 + 2\epsilon_2} a^3 \mathbf{E}_f \quad (14)$$

Because of $\bar{\sigma}_2 \ll \bar{\sigma}_1$ in real situation, we are allowed to see $\bar{\sigma}_2 = 0$ (i.e., $\epsilon_2 = \bar{\epsilon}_2$) in the following derivation as well as the numerical calculations presented in Section 3.2. And here, in the third-order harmonic terms, according to the mixing theory¹⁷, the high-order harmonic output is combined with one input frequency and two mixing frequencies, by using of basic angular frequencies $(\omega + \omega_1)$ and $(\omega - \omega_1)$. Specifically, we can see $3\omega \pm 3\omega_1 = 3(\omega \pm \omega_1)$ and $\omega \pm 3\omega_1 = 2(\omega \pm \omega_1) - (\omega \mp \omega_1)$. As a result, the angular velocity ω_0 in Eq. (13) is replaced by harmonic angular velocities in the same way. So we can get the harmonic terms of $\mathbf{P}(\omega \neq 0)$. If $\omega = 0$, based on Eq. (12) we obtain

$$\begin{aligned}P_{\omega_1} &= \frac{a^3(\bar{\epsilon}_1 - \bar{\epsilon}_2 - \frac{i\bar{\sigma}_1}{\omega_1})\bar{\epsilon}_2 E_0}{\bar{\epsilon}_1 - \frac{i\bar{\sigma}_1}{\omega_1} + 2\bar{\epsilon}_2}, \\ P_{3\omega_1} &= \frac{27a^3\bar{\epsilon}_2^4\chi E_0^3}{[\bar{\epsilon}_1 - \frac{i\bar{\sigma}_1}{\omega_1} + 2\bar{\epsilon}_2]^3[\bar{\epsilon}_1 - \frac{i\bar{\sigma}_1}{3\omega_1} + 2\bar{\epsilon}_2]}.\end{aligned}\quad (15, 16)$$

It is worth noting that in the actual use, only the real parts of the harmonic terms of induced dipole moment can be detected in experiments. Thus, we shall compute these real parts only in the following numerical calculations.

3. Numerical Results

3.1. Without dispersion

We investigate the cases without dispersion. For an ac electric field with angular frequency ω and a rotating ER particle with ω_1 , we have numerically calculated the harmonic responses of induced electric fields. (Figures have not been displayed here.) If dielectric constants ϵ_1 and ϵ_2 are given, higher nonlinear characteristics yield stronger harmonic responses. In other words, the magnitude of harmonic responses reflects the strength of nonlinear characteristics, as already experimentally reported by Klingenberg⁸.

3.2. With dispersion

We calculate the harmonic responses of induced dipole moment versus the angular frequency ω_1 of the rotating particle, for the external electric field with different frequencies ω . (Figures have not been displayed here.) For $P_{\omega+\omega_1}$, $P_{3\omega+3\omega_1}$, and $P_{\omega+3\omega_1}$, increasing ω_1 yields decreasing $P_{\omega+\omega_1}$, but increasing $P_{3\omega+3\omega_1}$ and $P_{\omega+3\omega_1}$. Inverse results appear for $P_{\omega-\omega_1}$, $P_{3\omega-3\omega_1}$, and $P_{\omega-3\omega_1}$, due to the subtraction of the particle rotation frequency from the external field frequency. In case of a dc electric field, we can investigate the effect of τ on the harmonic responses, see Fig. 1.

By analyzing Eq. (16), we find that, when

$$\omega_1 = 1.124 \frac{\bar{\epsilon}_1}{\bar{\epsilon}_1 + 2\bar{\epsilon}_2}, \quad (17)$$

a maximum value should appear in $\text{Re}(P_{3\omega_1})$, as displayed in Figs. 1(b) and 2 indeed. Here $\text{Re}(\dots)$ means taking the real part of $(\dots)$. In fact, this value could appear at the frequency $\omega_1 = \omega_1^*$ that satisfies the following known relation¹⁸

$$\tau\omega_1^* = 1. \quad (18)$$

According to Eq. (17), we obtain

$$\tau = \frac{\bar{\epsilon}_1 + 2\bar{\epsilon}_2}{1.124\bar{\epsilon}_1} = \frac{\tau_{MW}}{1.124}. \quad (19)$$

Here the relaxation time τ is determined by the details of the relaxation process, and the Maxwell-Wagner relaxation time τ_{MW} is based on the Maxwell-Wagner theory of leaky dielectrics since the relaxation process is originated from a finite conductivity of the particle and host medium¹⁸. In Table 1 the τ was calculated according to Eq. (19), and ω_1^* , which was extracted from the curves of Fig. 2, is the frequency at which $\text{Re}(P_{3\omega_1})$ reaches a maximum.

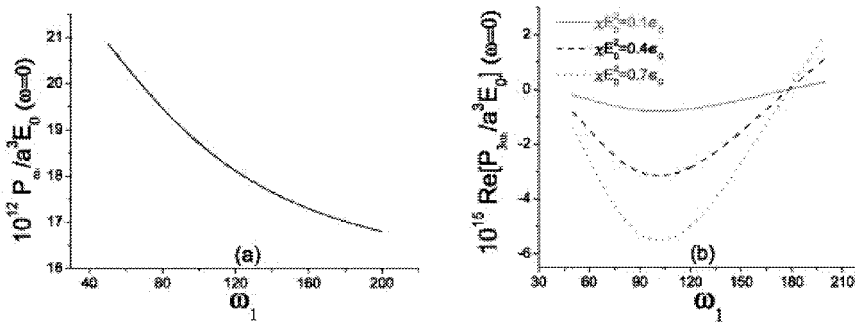


Fig. 1. (Cited from Ref. 12) *With dispersion*. For an dc electric field ($\omega = 0$), the real part of harmonic terms of induced dipole moment P as a function of the angular velocity ω_1 of suspended rotating particles. And (b) for different nonlinear characteristics χE_0^2 . Parameters: $\epsilon_1 = 20\epsilon_0$, $\epsilon_2 = 2.54\epsilon_0$, $\sigma_1 = 2 \times 10^{-8} \text{ S/m}$, $\sigma_2 = 1.0 \times 10^{-13} \text{ S/m}$, and $\chi E_0^2 = 0.7\epsilon_0$.

Figure 1(b) shows that the nonlinear characteristic does not affect ω_1^* . Actually, ω_1^* can be affected by the linear responses of the particles only, as shown in Eq. (18) already. Thus, we investigate the effect of $\bar{\sigma}_1$, $\bar{\epsilon}_1$, and $\bar{\epsilon}_2$ on the peak, see Fig. 2. It is shown that all the quantities have an effect on the ω_1^* . Especially, decreasing $\bar{\sigma}_1$ or increasing $\bar{\epsilon}_1$ or $\bar{\epsilon}_2$ makes the ω_1^* red-shifted (namely, located at lower frequencies). Consequently, in experiments, based on the observed peak frequency ω_1^* , we can conveniently determine the relaxation time $\tau (= 1/\omega_1^*)$. That is, our results offer an effective way to determine the relaxation time, which seems up to now a challenge. In addition, since we can detect each harmonic response at different ω_1 in experiment, the corresponding angular velocity ω of particles caused by shear flow is obtained directly.

Table 1. List of τ and ω_1^* . Here τ was calculated according to Eq. (19) and ω_1^* was extracted from the curves of Fig. 2. The two parameters satisfy $\tau\omega_1^* = 1$ [Eq. (18)].

	τ (s)	ω_1^* (rad/s)
$\bar{\sigma}_1 =$	$2 \times 10^{-8.2}$ S/m	0.0156
	2×10^{-8} S/m	0.0099
	$2 \times 10^{-7.8}$ S/m	0.0062
$\bar{\epsilon}_1 =$	$15\epsilon_0$	0.0078
	$20\epsilon_0$	0.0099
	$25\epsilon_0$	0.0118
$\bar{\epsilon}_2 =$	$2.2\epsilon_0$	0.0096
	$2.5\epsilon_0$	0.0099
	$2.8\epsilon_0$	0.0101

Klingenberg experimentally studied the nonlinear ac responses (harmonics) of electrorheological fluids⁸. Very interestingly, he experimentally showed that the harmonics of the electric current is caused to increase while the external electric field increases, which is qualitatively in agreement with our numerical results obtained from the local electric field and the induced dipole moment.

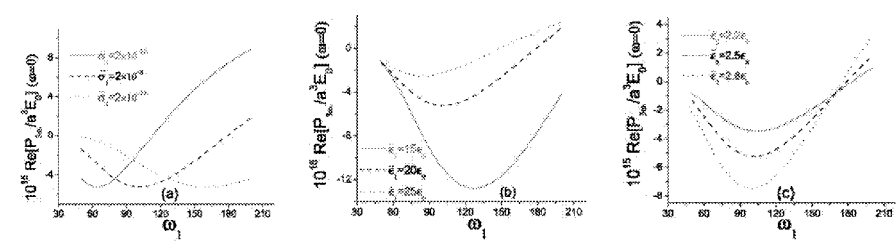


Fig. 2. (Cited from Ref. 12) *With dispersion*. Same as Fig. 1, but for different (a) $\bar{\sigma}_1$ (b) $\bar{\epsilon}_1$, and (c) $\bar{\epsilon}_2$. Parameters: $\epsilon_1 = 20\epsilon_0$, $\epsilon_2 = 2.54\epsilon_0$, $\sigma_1 = 2 \times 10^{-8}$ S/m, $\sigma_2 = 1.0 \times 10^{-13}$ S/m, and $\chi E_0^2 = 0.7\epsilon_0$, if they are not the variables.

4. Discussion and Conclusion

Throughout this work, only lower-order harmonics have been discussed. In fact, higher-order harmonics can also be studied as long as we keep more terms in Eqs. (6) and (8). However, the strength of the higher-order harmonics (e.g., fifth-order) is often several orders of magnitude smaller than the lower-order. To some extent, it is more attractive to detect the lower-order harmonics.

To sum up, we find that the dynamic effect due to a shear flow plays an important role in the nonlinear ac responses in ER fluids. Our results offer a convenient method to determine the relaxation time and the rotation velocity of ER particles, by measuring the nonlinear ac responses.

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