Dynamic effects on nonlinear alternating current responses in electrorheological fluids

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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 direct current electric field. We show that the dynamic effect due to a shear flow, which exerts a torque on ER particles and thus leads to the rotation of the particles about their centers, 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.

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I. 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 flow exerts force and torque on the suspended particles, setting the particles in both translational and rotational motion. For instance, the shear flow in an ER fluid exerts a torque on the particles, which leads to the rotation of particles about their centers [3–10]. Experiments [7] showed that the induced interparticle forces between rotating ER particles are quite different from the values predicted by the existing theories that have not included the motion of particles. Wan *et al.* [8,9] theoretically investigated this 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 experiments [11].

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 [12–21]. We shall show that, if the suspended particles in the suspension rotates with angular frequency ω_1, the desired frequencies of the higher-order harmonics will become more abundant due to the coupling between ω and ω_1. We shall also show that, once a direct current (dc) electric field is applied to such suspensions containing rotating particles with ω_1, harmonic signals can also be detected at various frequencies like ω_1 and $3\omega_1$.

In this work, we shall develop a perturbation approach [13,22] to investigate the dynamic effect due to a shear flow on nonlinear ac responses in ER fluids under an ac or dc electric field. This work is developed on the single-particle scale in the dilute limit. It is found that this kind of dynamic effect plays a significant role in the responses. According to our results, it seems possible to detect some physical parameters (e.g., relaxation times and rotation velocities) of ER particles, by measuring the nonlinear ac responses.

This paper is organized as follows. In Sec. II, based on a perturbation approach, we present the formalism for the nonlinear ac responses of a rotating ER particle without or with dispersion. This is followed by Sec. III where numerical results are presented under different conditions. The paper ends with a discussion and conclusion in Sec. IV.

II. FORMALISM

A. Without dispersion

We first consider a particle in an ER fluid under an applied external ac electric field $\mathbf{E}_{f0}=E_0\cos(\omega t)\hat{\mathbf{Z}}\equiv\mathbf{E}_0\cos(\omega t)$. The shear flow in the ER suspension will give the particle a torque, which leads it to get a rotational motion about its center (see Fig. 1). We assume that the angular velocity caused by shear flow is $\omega_1\hat{\mathbf{Y}}$. Then if we rotate with the

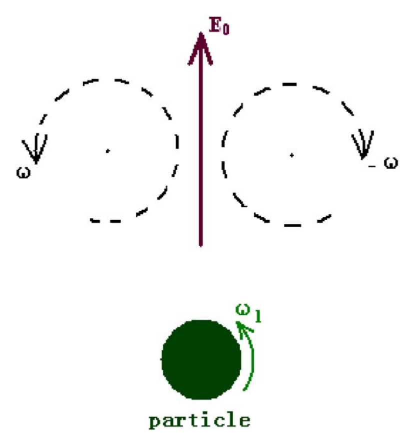

FIG. 1. (Color online) Schematic graph showing the rotation coupling [Eq. (1)] between an external ac electric field with angular frequency ω and a rotating ER particle with ω_1.

*Electronic address: jphuang@fudan.edu.cn particle, the particle will seem to be at rest, and it experiences an electric field written as (see Fig. 1)

$$\mathbf{E}_f = \frac{1}{2} e^{-it\omega_1}(e^{-it\omega} + e^{it\omega})\mathbf{E}_0. \tag{1}$$

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

$$\mathbf{D}_1 = \epsilon_1 \mathbf{E}_1 + \chi |\mathbf{E}_1|^2 \mathbf{E}_1 \equiv \tilde{\epsilon}_1 \mathbf{E}_1, \tag{2}$$

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

$$\mathbf{E}_1 = \frac{3\epsilon_2}{\epsilon_1 + \chi E_1^2 + 2\epsilon_2} \mathbf{E}_f. \tag{3}$$

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 meantime, we shall ignore the smaller terms. Thus, we get

$$\mathbf{E}_1 = \frac{3e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})\epsilon_2}{2(\epsilon_1 + 2\epsilon_2)} \mathbf{E}_0 - \frac{3e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})\epsilon_2 \chi E_1^2}{2(\epsilon_1 + 2\epsilon_2)^2} \mathbf{E}_0. \tag{4}$$

And the first term of the right part of Eq. (4) is the linear part. Because this first term is much larger than the second one (i.e., nonlinear part), we can take it as E_1 back to Eq. (4). Then E_1 takes the form

$$E_1 = \frac{3e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})\epsilon_2 E_0}{2(\epsilon_1 + 2\epsilon_2)} - \frac{27e^{-3it(\omega+\omega_1)}(1 + e^{2it\omega})^3 \epsilon_2^3 \chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4}. \tag{5}$$

By expanding this form of $E_1(\omega \neq 0)$, we obtain the harmonic terms of various frequencies:

$$E_1 = E_{\omega+\omega_1} e^{-it(\omega+\omega_1)} + E_{\omega-\omega_1} e^{it(\omega-\omega_1)} + E_{3\omega+3\omega_1} e^{-3it(\omega+\omega_1)} + E_{3\omega-3\omega_1} e^{3it(\omega-\omega_1)} + E_{\omega+3\omega_1} e^{-it(\omega+3\omega_1)} + E_{\omega-3\omega_1} e^{it(\omega-3\omega_1)}, \tag{6}$$

where

$$E_{\omega\pm\omega_1} = \frac{3\epsilon_2 E_0}{2(\epsilon_1 + 2\epsilon_2)},$$

$$E_{3\omega\pm3\omega_1} = -\frac{27\epsilon_2^3 \chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4},$$

$$E_{\omega\pm3\omega_1} = -\frac{81\epsilon_2^3 \chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4}. \tag{7}$$

On the other hand, the induced dipole moment

$$\mathbf{P} = \epsilon_2 \frac{\epsilon_1 + \chi E_1^2 - \epsilon_2}{\epsilon_1 + \chi E_1^2 + 2\epsilon_2} a^3 \mathbf{E}_f \tag{8}$$

can be treated in the same way. In Eq. (8) a denotes the radius of the particle. Then we expand $P(\omega \neq 0)$ by taking χE_1^2 as a perturbation and ignore the smaller terms. As a result, we obtain

$$\mathbf{P} = \frac{a^3 e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})(\epsilon_1 - \epsilon_2)\epsilon_2}{2(\epsilon_1 + 2\epsilon_2)} \mathbf{E}_0 + \frac{3a^3 e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})\epsilon_2^2 \chi E_1^2}{2(\epsilon_1 + 2\epsilon_2)^2} \mathbf{E}_0. \tag{9}$$

Now the linear term (namely, the first term) of E_1 in the right-hand side of Eq. (4) is introduced into Eq. (9). Then we obtain

$$P = \frac{a^3 e^{-it(\omega+\omega_1)}(1 + e^{2it\omega})(\epsilon_1 - \epsilon_2)\epsilon_2 E_0}{2(\epsilon_1 + 2\epsilon_2)} + \frac{27a^3 e^{-3it(\omega+\omega_1)}(1 + e^{2it\omega})^3 \epsilon_2^4 \chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4}. \tag{10}$$

This can be rewritten as

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

where the harmonic coefficients of various frequencies are given by

$$P_{\omega\pm\omega_1} = \frac{a^3(\epsilon_1 - \epsilon_2)\epsilon_2 E_0}{2(\epsilon_1 + 2\epsilon_2)},$$

$$P_{3\omega\pm3\omega_1} = \frac{27a^3 \epsilon_2^4 \chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4},$$

$$P_{\omega\pm3\omega_1} = \frac{81a^3 \epsilon_2^4 \chi E_0^3}{8(\epsilon_1 + 2\epsilon_2)^4}. \tag{12}$$

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

$$E_1^{(\omega=0)} = E_{\omega_1} e^{-it\omega_1} + E_{3\omega_1} e^{-3it\omega_1}, \tag{13}$$

where

$$E_{\omega_1} = \frac{3\epsilon_2 E_0}{\epsilon_1 + 2\epsilon_2},$$

$$E_{3\omega_1} = -\frac{27\epsilon_2^3 \chi E_0^3}{(\epsilon_1 + 2\epsilon_2)^4}. \tag{14}$$

And $P^{(\omega=0)}$is given by

$$P^{(\omega=0)} = P_{\omega_1} e^{-it\omega_1} + P_{3\omega_1} e^{-3it\omega_1}, \tag{15}$$

where

$$P_{\omega_1} = \frac{a^3(\epsilon_1 - \epsilon_2)\epsilon_2 E_0}{\epsilon_1 + 2\epsilon_2},$$

$$P_{3\omega_1} = \frac{27a^3\epsilon_2^4\chi E_0^3}{(\epsilon_1 + 2\epsilon_2)^4}. \tag{16}$$

Comparing Eq. (6) to Eq. (13), and Eq. (11) to Eq. (15), 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. (12) as long as the external field frequency ω is known.

B. With dispersion

Until now, the dielectric constants ϵ_1 and ϵ_2 that 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 [23,24], and they can be described as

$$\epsilon_1 = \bar{\epsilon}_1 - i\bar{\sigma}_1/\omega_0,$$

$$\epsilon_2 = \bar{\epsilon}_2 - i\bar{\sigma}_2/\omega_0, \tag{17}$$

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. (17), ω_0 denotes the angular frequency whose details can be found below. In this case, the induced dipole moment should be [25]

$$\mathbf{P} = \bar{\epsilon}_2 \frac{\epsilon_1 + \chi E_1^2 - \epsilon_2}{\epsilon_1 + \chi E_1^2 + 2\epsilon_2} a^3 \mathbf{E}_f. \tag{18}$$

Because $\bar{\sigma}_2 \ll \bar{\sigma}_1$ in real situations, we are allowed to set $\bar{\sigma}_2 = 0$ (i.e., $\epsilon_2 = \bar{\epsilon}_2$) in the following derivation as well as the numerical calculations presented in Sec. III B. And here, in the third-order harmonic terms, according to the mixing theory [26], the high-order harmonic output is combined with one input frequency and two mixing frequencies, by using basic angular frequencies $(\omega+\omega_1)$ and $(\omega-\omega_1)$. Specifically, we can set $3\omega\pm 3\omega_1 = 3(\omega\pm\omega_1)$ 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. (17) is replaced by harmonic angular velocities in the same way. So the harmonic terms of $P(\omega\neq 0)$ are given by

$$P_{\omega\pm\omega_1} = \frac{a^3\bar{\epsilon}_2[\bar{\epsilon}_1 - i\bar{\sigma}_1/(\omega\pm\omega_1) - \bar{\epsilon}_2]E_0}{2[\bar{\epsilon}_1 - i\bar{\sigma}_1/(\omega\pm\omega_1) + 2\bar{\epsilon}_2]},$$

$$P_{3\omega\pm 3\omega_1} = \frac{27a^3\bar{\epsilon}_2^4\chi E_0^3}{8[\bar{\epsilon}_1 - i\bar{\sigma}_1/(\omega\pm\omega_1) + 2\bar{\epsilon}_2]^3[\bar{\epsilon}_1 - i\bar{\sigma}_1/3(\omega\pm\omega_1) + 2\bar{\epsilon}_2]},$$

$$P_{\omega\pm 3\omega_1} = \frac{81a^3\bar{\epsilon}_2^4\chi E_0^3}{8[\bar{\epsilon}_1 - i\bar{\sigma}_1/(\omega\pm\omega_1) + 2\bar{\epsilon}_2]^2[\bar{\epsilon}_1 + i\bar{\sigma}_1/(\omega\mp\omega_1) + 2\bar{\epsilon}_2][\bar{\epsilon}_1 - i\bar{\sigma}_1/(\omega\pm 3\omega_1) + 2\bar{\epsilon}_2]}. \tag{19}$$

If $\omega=0$, based on Eq. (16) we obtain

$$P_{\omega_1} = \frac{a^3(\bar{\epsilon}_1 - \bar{\epsilon}_2 - i\bar{\sigma}_1/\omega_1)\bar{\epsilon}_2 E_0}{\bar{\epsilon}_1 - i\bar{\sigma}_1/\omega_1 + 2\bar{\epsilon}_2}, \tag{20}$$

$$P_{3\omega_1} = \frac{27a^3\bar{\epsilon}_2^4\chi E_0^3}{(\bar{\epsilon}_1 - i\bar{\sigma}_1/\omega_1 + 2\bar{\epsilon}_2)^3(\bar{\epsilon}_1 - i\bar{\sigma}_1/3\omega_1 + 2\bar{\epsilon}_2)}. \tag{21}$$

It is worth noting that in actual use, only the real parts of the harmonic terms of the induced dipole moment [Eqs. (19)–(21)] can be detected in experiments. Thus, we shall compute these real parts only in the following numerical calculations.

III. NUMERICAL RESULTS

A. Without dispersion

We investigate the cases without dispersion in Figs. 2–5. In Fig. 2, 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 (a), (d) $E_{\omega\pm\omega_1}$, (b), (e) $E_{3\omega\pm 3\omega_1}$, and (c), (f) $E_{\omega\pm 3\omega_1}$. If dielectric the constants ϵ_1 and ϵ_2 are given, higher nonlinear characteristics yield stronger harmonic responses. In other words, the magnitude of the harmonic responses reflects the strength of the nonlinear characteristics, as already experimentally reported by Klingenberg [12]. In the meantime, we find that increasing ϵ_1 leads to decreasing third-harmonic responses [see Figs. 2(a)–2(c)]. In contrast, increasing ϵ_2 causes the harmonic response to increase. Similar behavior can be found in Fig. 3 where a dc external electric field ($\omega=0$) is applied. These can be understood well from the local field

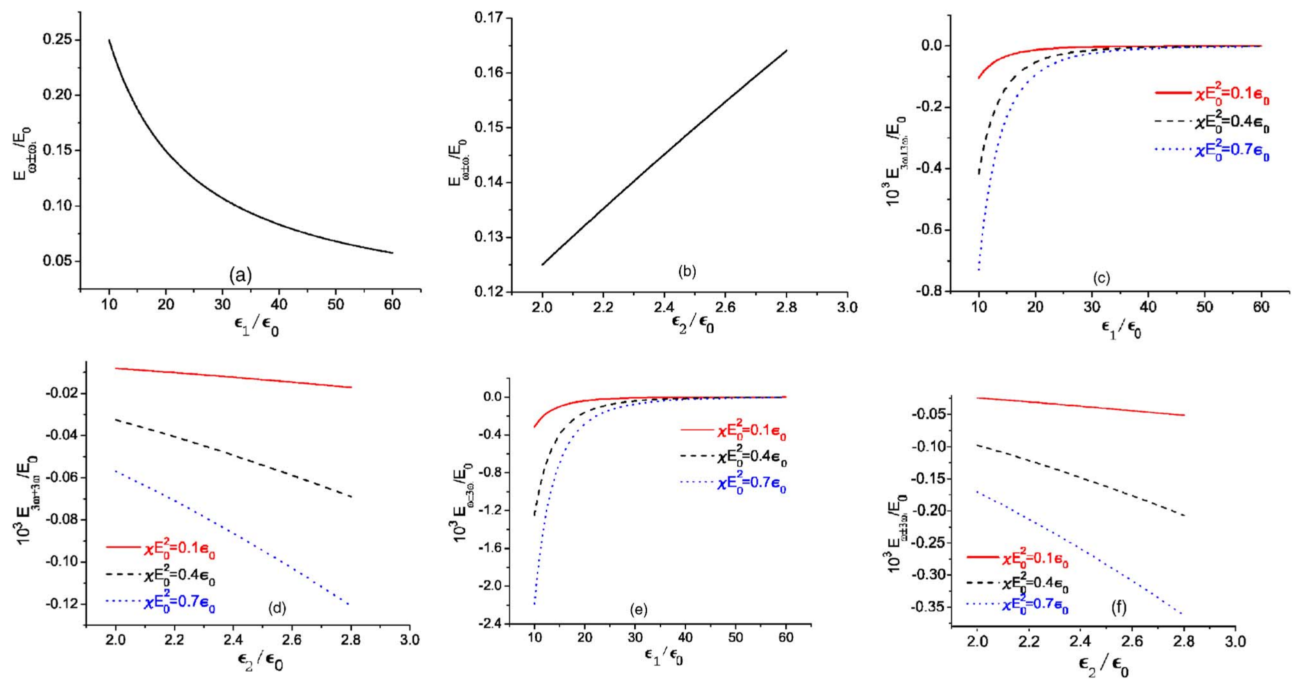

FIG. 2. (Color online) Without dispersion. For an ac electric field ($\omega \neq 0$), the harmonic responses of the local electric field E_1 versus the dielectric constants (a)–(c) ϵ_1 and (d)–(f) ϵ_2. Here ϵ_0 denotes the dielectric constant in vacuum. Parameters: (a)–(c) $\epsilon_2=2.5\epsilon_0$; (d)–(f) $\epsilon_1=20\epsilon_0$.

effect [Eq. (3)]. In detail, based on Eq. (3), it is apparent that increasing ϵ_1 leads to decreasing local electric field E_1. However, increasing ϵ_2 causes E_1 to increase. In Figs. 4 and 5, except for their first panels [i.e., Figs. 4(a) and 5(a)], all other panels show framework similar to the corresponding panels in Figs. 2 and 3. In Figs. 4(a) and 5(a), we find that increasing ϵ_1 yields increasing basic harmonics.

B. With dispersion

Figure 6 displays 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. For $P_{\omega+\omega_1}$, $P_{3\omega+3\omega_1}$, and $P_{\omega+3\omega_1}$(left panels), 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 $P_{\omega-3\omega_1}$ (right panels), 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 ω_1 on the harmonic responses (see Fig. 7). Figure 7(a) predicts the same result as Fig. 6(a).

By analyzing Eq. (21), we find that, when

$$\omega_1 = 1.124\frac{\bar{\sigma}_1}{\bar{\epsilon}_1 + 2\bar{\epsilon}_2}, \tag{22}$$

a maximum value should appear in $\mathrm{Re}(P_{3\omega_1})$, as displayed in Figs. 7(b) and 8 indeed. Here $\mathrm{Re}(\cdots)$ means taking the real part of $(\cdots)$. In fact, this value could appear at the frequency $\omega_1=\omega_1^*$ that satisfies the following known relation [27]:

$$\tau\omega_1^* = 1. \tag{23}$$

According to Eq. (22), we obtain

$$\tau = \frac{\bar{\epsilon}_1 + 2\bar{\epsilon}_2}{1.124\bar{\sigma}_1} = \frac{\tau_{MW}}{1.124}. \tag{24}$$

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 originates from the finite conductivity of the particle and host medium [27]. In Table I the τ was calculated according to Eq. (24), and ω_1^*, which was extracted from the curves of Fig. 8, is the frequency at which $\mathrm{Re}(P_{3\omega_1})$ reaches a maximum.

In Figs. 7(b) and 8, we show that the harmonic response can pass through zero (i.e., from negative to positive number as ω_1 increases). To understand this, we invoked the dielectric dispersion spectral representation (DDSR) [28,29] for the harmonics. The DDSR enables us to express the harmonics in terms of a series of subdispersions, each of which has analytic expressions for the dispersion strengths and their corresponding characteristic frequencies expressed in terms of the various parameters of the model. Let us define two parameters s and t,

$$s = \left(1 - \frac{\bar{\epsilon}_1}{\bar{\epsilon}_2}\right)^{-1}, \quad t = \left(1 - \frac{\bar{\sigma}_1}{\bar{\sigma}_2}\right)^{-1}.$$

In case of the parameters adopted in the present work, $\bar{\epsilon}_1=20\epsilon_0$, $\bar{\epsilon}_2=2.5\epsilon_0$, and $\bar{\sigma}_2=0$, the numerical values of s and t are, respectively, $-0.142\,857$ and 0. Thus, for the fundamental response, the real part will just be a monotonically decreasing function, as displayed in Fig. 7(a) indeed. For the third-harmonic response, it can be expressed as five terms in the DDSR, namely, constant [or $(s-1/3)^0$], $(s-1/3)^{-1}$, $(s$

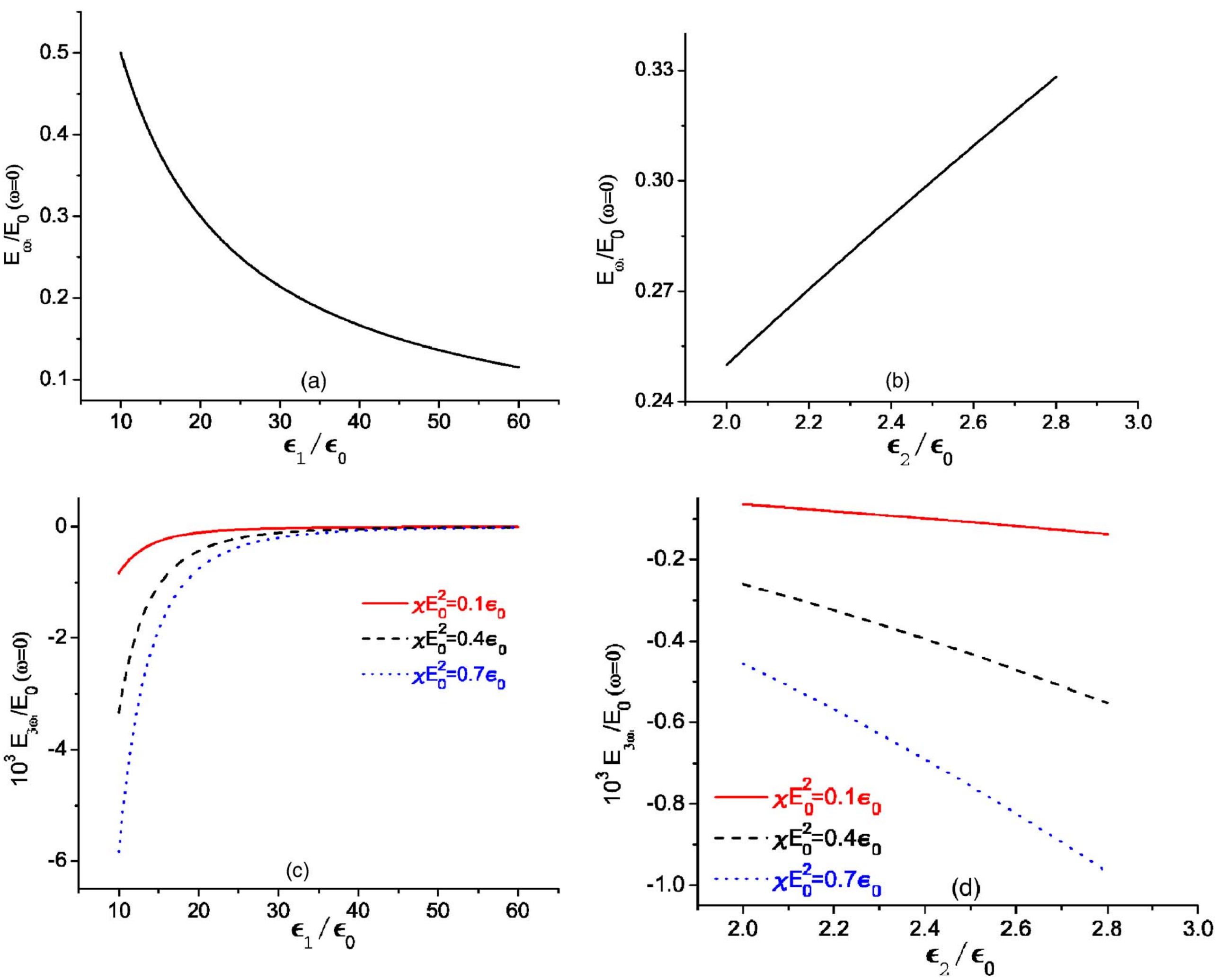

FIG. 3. (Color online) Without dispersion. Same as Fig. 2, but for a dc electric field ($\omega=0$).

$-1/3)^{-2}$, $(s-1/3)^{-3}$, and $(s-1/3)^{-4}$. Thus, the real part of the third-harmonic response should possess a peak at some peak frequency. At the same time it can cross zero and becomes positive at higher frequencies because $s<0$ and $t=0$. Also, the apparent shift of the peakfrequency ω_1^* from $1/\tau_{MW}$ to $1/\tau$ in Eq. (23) is due to the unconventional terms [29] in the DDSR.

Figure 7(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. (23) already. Thus, we investigate the effect of $\bar{\sigma}_1$, $\bar{\epsilon}_1$, and $\bar{\epsilon}_2$ on the peak, see Fig. 8. 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^* redshifted (namely, located at lower frequencies). Consequently, in experiments, based on the observed peak frequencyω_1^*, we can conveniently determine the relaxation time τ $(=1/\omega_1^*)$. 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_1 of particles caused by shear flow is obtained directly.

Klingenberg experimentally studied the nonlinear ac responses (harmonics) of electrorheological fluids [12]. 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 [see Figs. 2(b), 2(c), 2(e), 2(f), 3(b), 3(d), 4(b), 4(c), 4(e), 4(f), 5(b), 5(d), and 7(b)]. In addition, by using this kind of perturbation approach, Wei and coauthors also investigated nonlinear ac reponses of composite materials [20,21].

It turns out generally difficult to give a full account of all the numerical results in simple physical terms. However, we already explained the behaviors of the harmonic responses (at least partly) by the local field effects, aided by the spectral representation (see Ref. [17]). This is due to the fact that the spectral representation can reveal the dominant contribution through the self-consistent approach.

IV. DISCUSSION AND CONCLUSION

We have performed a perturbation approach to investigate nonlinear ac responses in ER fluids which are subjected to an ac or dc electric field. Our focus is on the dynamic effect due to a shear flow, which exerts a torque on ER particles and

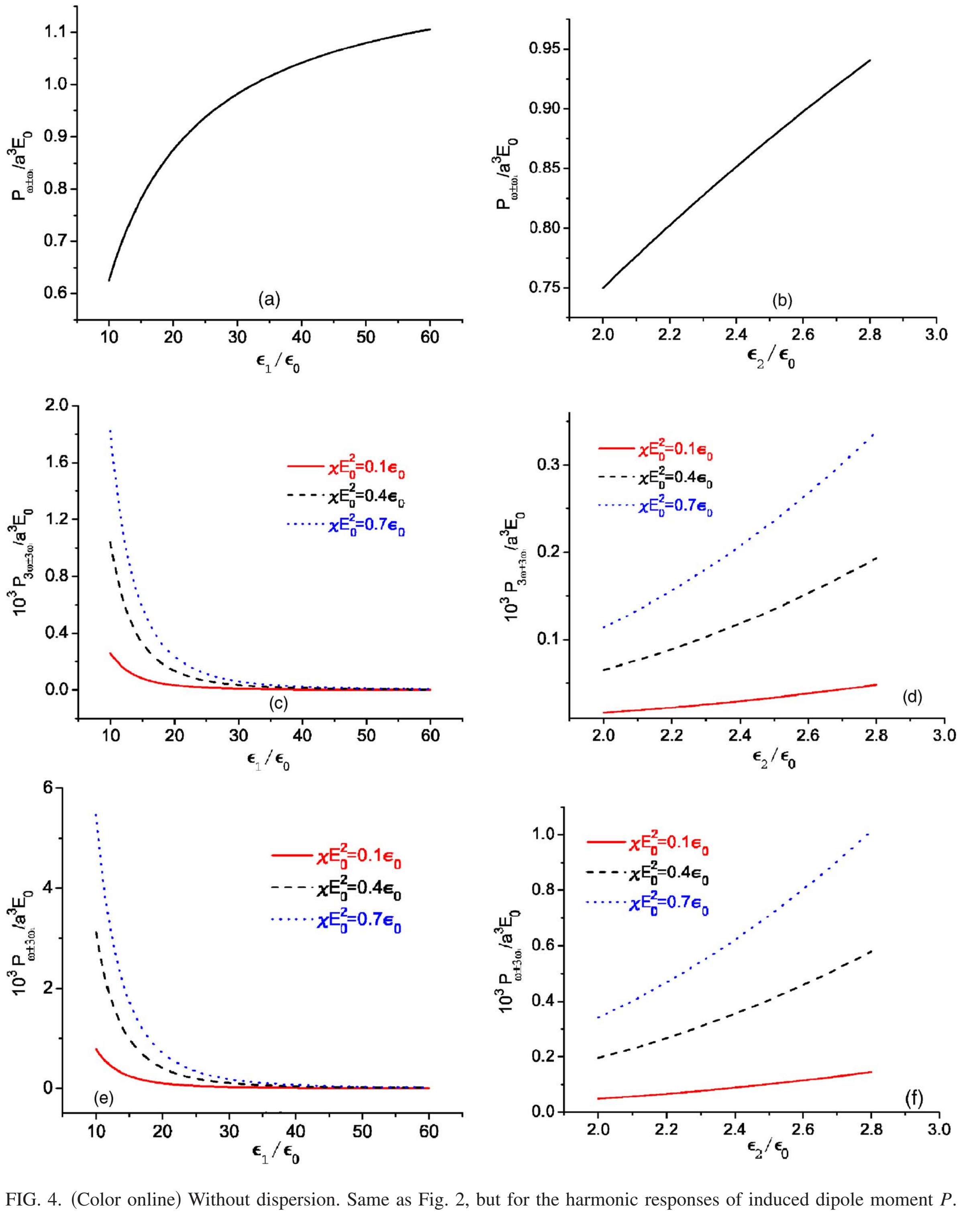

FIG. 4. (Color online) Without dispersion. Same as Fig. 2, but for the harmonic responses of induced dipole moment P.

thus leads to the rotation of the particles about their centers.

In real ER fluids, there is always $\epsilon_1 > \epsilon_2$ [2]. In this work, we used $\epsilon_1 > 10\epsilon_0$ because ϵ_2 is often $(2-3\epsilon_0)$. It is known that ER fluids are more stable in low frequencies than in high frequencies. In general, the real frequency in use can often be smaller than 10^4 Hz (or $2\pi \times 10^4$ rad/s) [30]. Thus, in our calculations, we take the range for frequencies $\omega = 10^{2.8} - 10^{3.2}$ rad/s and $\omega_1 > 40$ rad/s, in which the feature of a frequency-dependent ER activity has been included [30]. If we adopt smaller ϵ_1 or ω, the qualitative results we have achieved remain unchanged.

Strictly speaking, the condition $\omega = 0$ does not refer to the dc case, because the particle is actually rotating and ω_1 (angular velocity caused by shear flow) is nonzero. Thus in this case we still need to study the ac response.

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. (4) and (9).

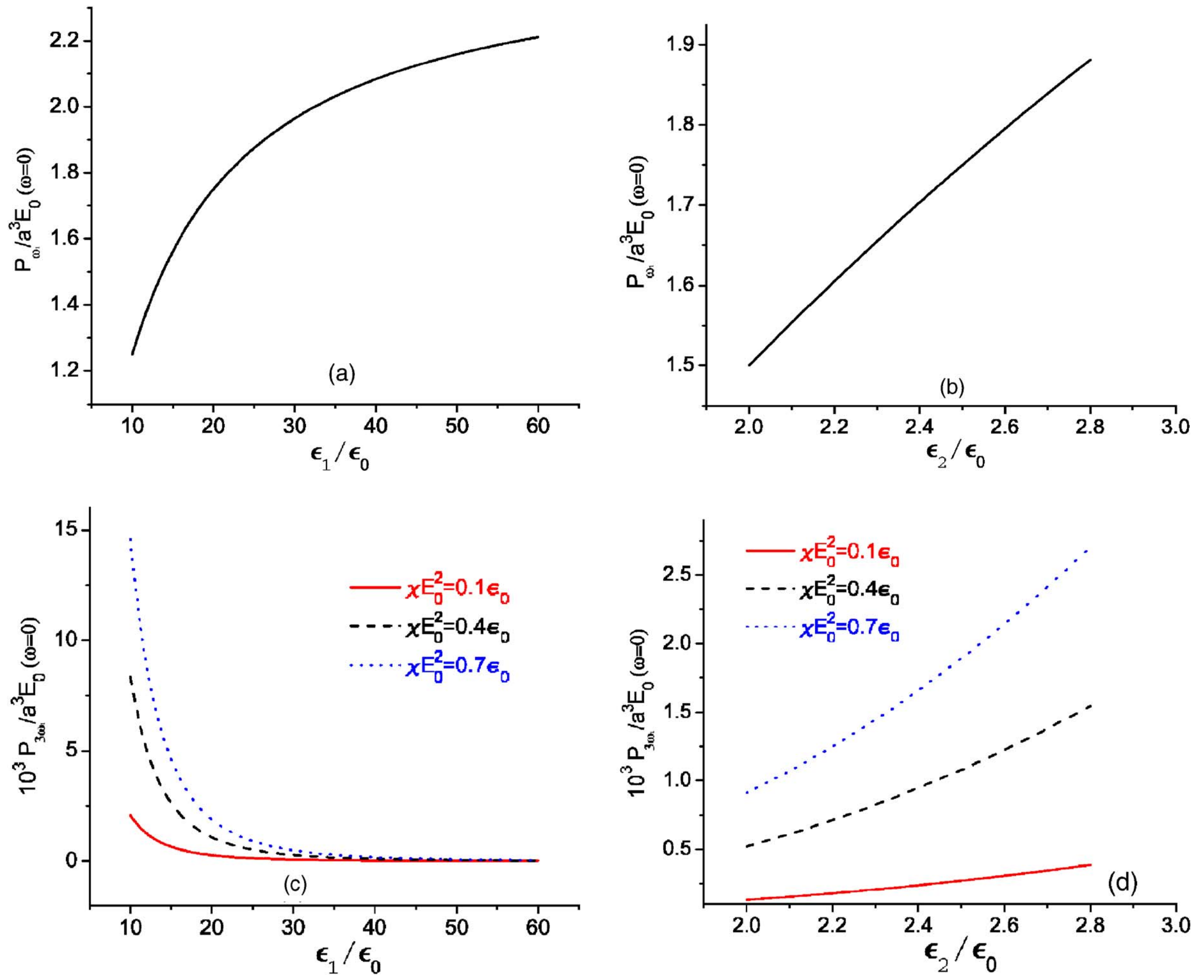

FIG. 5. (Color online) Without dispersion. Same as Fig. 4, but for a dc electric field (ω=0).

However, the strength of the higher-order harmonics (e.g., fifth order) is often several orders of magnitude smaller than of the lower-order terms. To some extent, it is more attractive to detect the lower-order harmonics. As a matter of fact, higher-order harmonics can arise from different origins. Let us take the fifth harmonic as an example. They can be induced to appear by third-order nonlinearity (see Appendix A), that is, lower-order nonlinearity can induce higher-order nonlinear responses. On the other hand, fifth harmonics can be induced to appear by fifth-order nonlinearity (see Appendix B). In the two appendixes, we derived the expressions for harmonic coefficients for particles without dispersion. Following Sec. II B, this can be directly extended to deal with particles with dispersion.

It would be interesting to see what happens if one takes into account a pair of rotating particles suspended in ER fluids. In this case, multipolar interaction should be included. For this purpose, a multipole expansion method [31] can be used. If there are many particles suspended in the system, the many-body (local field) effect should also be considered. In so doing, we can resort to an effective medium theory like the Maxwell-Garnett approximation [32]. In addition, our theory can also be extended to deal with other suspended objects in shear flow, e.g., erythrocytes [33], vesicles [34], etc.

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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APPENDIX A: FIFTH HARMONICS INDUCED BY THIRD-ORDER NONLINEARITY

Fifth harmonics can appear due to third-order nonlinearity. Let us start from Eq. (4). We expand the E_1 again and

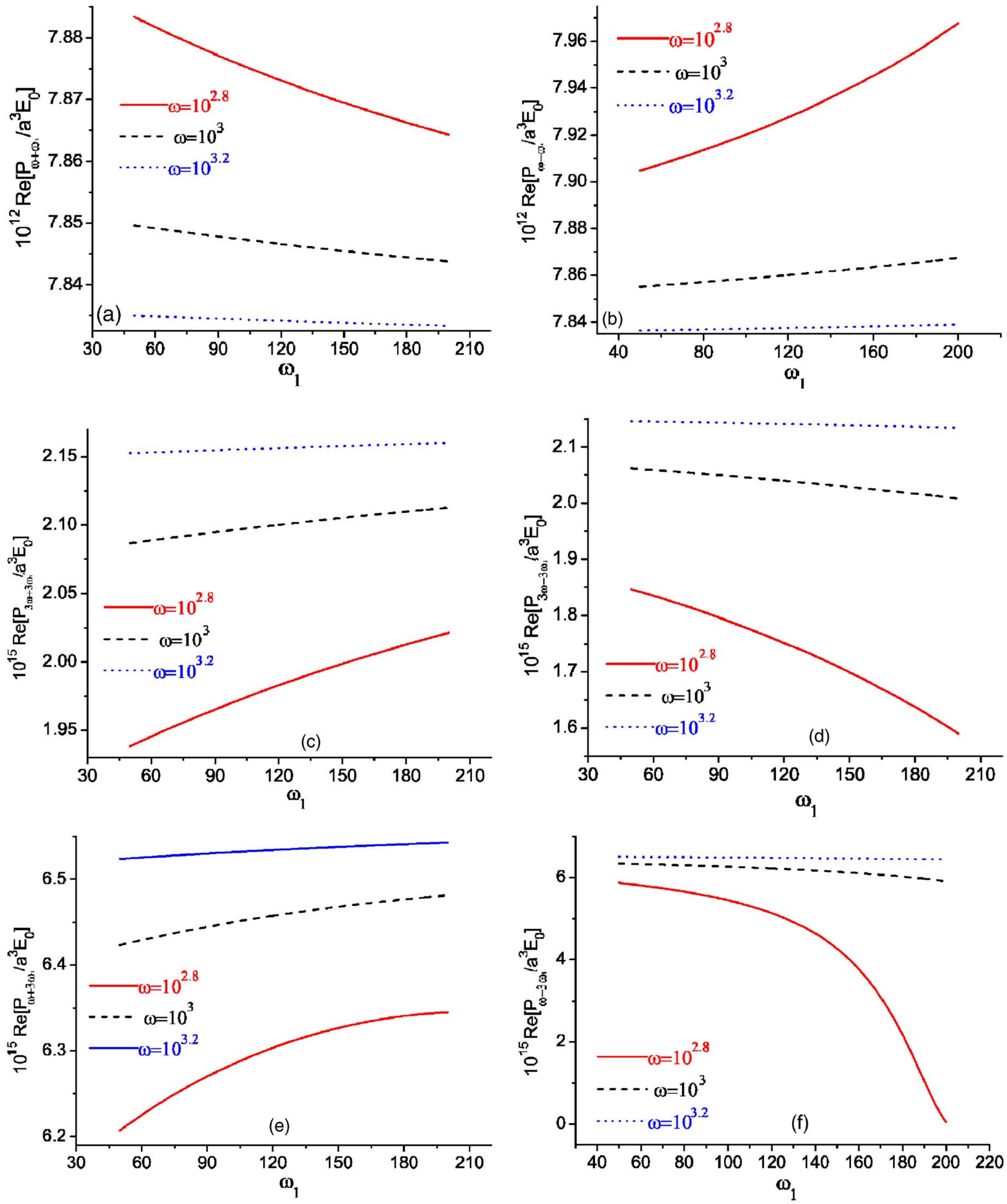

FIG. 6. (Color online) With dispersion. For an ac electric field with different angular frequencies ω, the real part of the harmonic terms of induced dipole moment P as a function of the angular velocity ω_1 of suspended rotating particles. Parameters: $\epsilon_1=20\epsilon_0$, $\epsilon_2=2.54\epsilon_0$, $\sigma_1=2\times10^{-8}$ S/m, $\sigma_2=1.0\times10^{-13}$ S/m, and $\chi E_0^2=0.7\epsilon_0$.

hold one more term, namely, the third one. So E_1 is given by

$$E_1=\frac{3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2}{2(\epsilon_1+2\epsilon_2)}E_0-\frac{3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2\chi E_1^2}{2(\epsilon_1+2\epsilon_2)^2}E_0+\frac{3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2\chi^2E_1^4}{2(\epsilon_1+2\epsilon_2)^3}E_0. \quad (A1)$$

Then we put the linear part of E_1 into the right-hand side of Eq. (A1). In view of Eqs. (1) and (3), we get its harmonic form

$$E_1=E_{\omega+\omega_1}e^{-it(\omega+\omega_1)}+E_{\omega-\omega_1}e^{it(\omega-\omega_1)}+E_{3\omega+3\omega_1}e^{-3it(\omega+\omega_1)}+E_{3\omega-3\omega_1}e^{3it(\omega-\omega_1)}+E_{\omega+3\omega_1}e^{-it(\omega+3\omega_1)}+E_{\omega-3\omega_1}e^{it(\omega-3\omega_1)}+E_{5\omega+5\omega_1}e^{-5it(\omega+\omega_1)}+E_{5\omega-5\omega_1}e^{5it(\omega-\omega_1)}$$

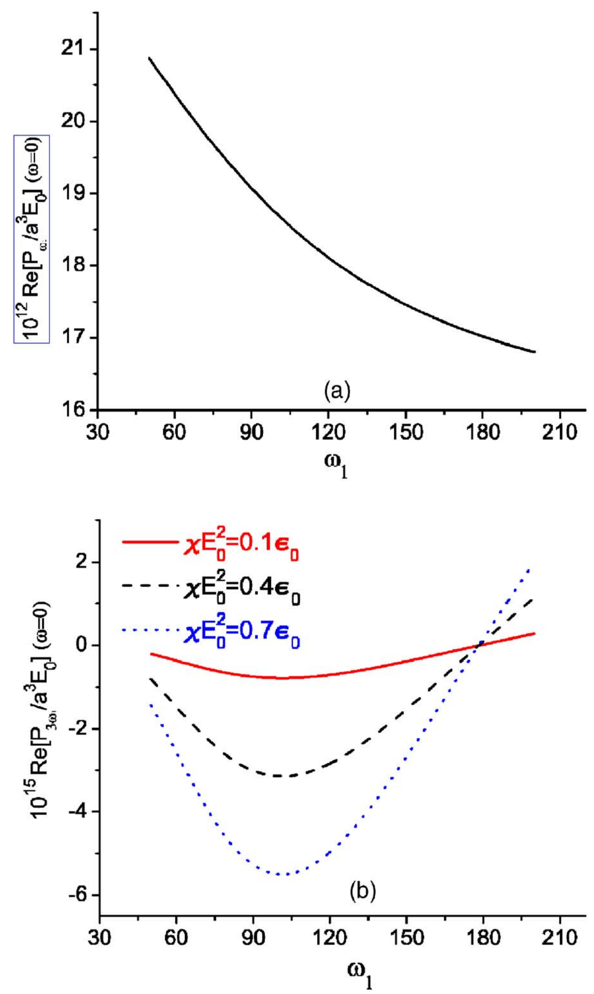

FIG. 7. (Color online) With dispersion. Same as Fig. 6, but for a dc electric field (ω=0), and (b) for different nonlinear characteristics χE_0^2.

$$+E_{3\omega+5\omega_1}e^{-it(3\omega+5\omega_1)}+E_{3\omega-5\omega_1}e^{it(3\omega-5\omega_1)}$$
$$+E_{\omega+5\omega_1}e^{-it(\omega+5\omega_1)}+E_{\omega-5\omega_1}e^{it(\omega-5\omega_1)}, \quad \text{(A2)}$$

where the harmonic coefficients are respectively given by

$$E_{\omega\pm\omega_1}=\frac{3\epsilon_2E_0}{2(\epsilon_1+2\epsilon_2)},$$

$$E_{3\omega\pm3\omega_1}=-\frac{27\epsilon_2^3\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$E_{\omega\pm3\omega_1}=-\frac{81\epsilon_2^3\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$E_{5\omega\pm5\omega_1}=\frac{243\epsilon_2^5\chi^2E_0^5}{32(\epsilon_1+2\epsilon_2)^7},$$

$$E_{3\omega\pm5\omega_1}=\frac{1215\epsilon_2^5\chi^2E_0^5}{32(\epsilon_1+2\epsilon_2)^7},$$

$$E_{\omega\pm5\omega_1}=\frac{1215\epsilon_2^5\chi^2E_0^5}{16(\epsilon_1+2\epsilon_2)^7}. \quad \text{(A3)}$$

And we expand the induced dipole moment P [Eq. (9)] by χE_1^2 to the term of χ^2. So P is given by

$$P=\frac{a^3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})(\epsilon_1-\epsilon_2)\epsilon_2}{2(\epsilon_1+2\epsilon_2)}E_0-\frac{3a^3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2^2\chi E_1^2}{2(\epsilon_1+2\epsilon_2)^2}E_0-\frac{3a^3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2^2\chi^2E_1^4}{2(\epsilon_1+2\epsilon_2)^3}E_0. \quad \text{(A4)}$$

Then we put the linear part of E_1 into the right part of the equation, and expand P to get its harmonic form. So we have

$$P=P_{\omega+\omega_1}e^{-it(\omega+\omega_1)}+P_{\omega-\omega_1}e^{it(\omega-\omega_1)}+P_{3\omega+3\omega_1}e^{-3it(\omega+\omega_1)}+P_{3\omega-3\omega_1}e^{3it(\omega-\omega_1)}+P_{\omega+3\omega_1}e^{-it(\omega+3\omega_1)}+P_{\omega-3\omega_1}e^{it(\omega-3\omega_1)}+P_{5\omega+5\omega_1}e^{-5it(\omega+\omega_1)}+P_{5\omega-5\omega_1}e^{5it(\omega-\omega_1)}+P_{3\omega+5\omega_1}e^{-it(3\omega+5\omega_1)}+P_{3\omega-5\omega_1}e^{it(3\omega-5\omega_1)}+P_{\omega+5\omega_1}e^{-it(\omega+5\omega_1)}+P_{\omega-5\omega_1}e^{it(\omega-5\omega_1)}, \quad \text{(A5)}$$

where the harmonic coefficients up to fifth order are given by

$$P_{\omega\pm\omega_1}=\frac{a^3(\epsilon_1-\epsilon_2)\epsilon_2E_0}{2(\epsilon_1+2\epsilon_2)},$$

$$P_{3\omega\pm3\omega_1}=\frac{27a^3\epsilon_2^4\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$P_{\omega\pm3\omega_1}=\frac{81a^3\epsilon_2^4\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$P_{5\omega\pm5\omega_1}=-\frac{243a^3\epsilon_2^6\chi^2E_0^5}{32(\epsilon_1+2\epsilon_2)^7},$$

$$P_{3\omega\pm5\omega_1}=-\frac{1215a^3\epsilon_2^6\chi^2E_0^5}{32(\epsilon_1+2\epsilon_2)^7},$$

$$P_{\omega\pm5\omega_1}=-\frac{1215a^3\epsilon_2^6\chi^2E_0^5}{16(\epsilon_1+2\epsilon_2)^7}. \quad \text{(A6)}$$

APPENDIX B: FIFTH HARMONICS INDUCED BY FIFTH-ORDER NONLINEARITY

The fifth-order harmonic terms discussed in Appendix A are induced by third-order nonlinearity. In this section, we shall study the effect of fifth-order nonlinearity on the fifth harmonics. In this case, the constructive relation between the local electric field $\mathbf{E}_1$ and the electric displacement $\mathbf{D}_1$ can be defined as

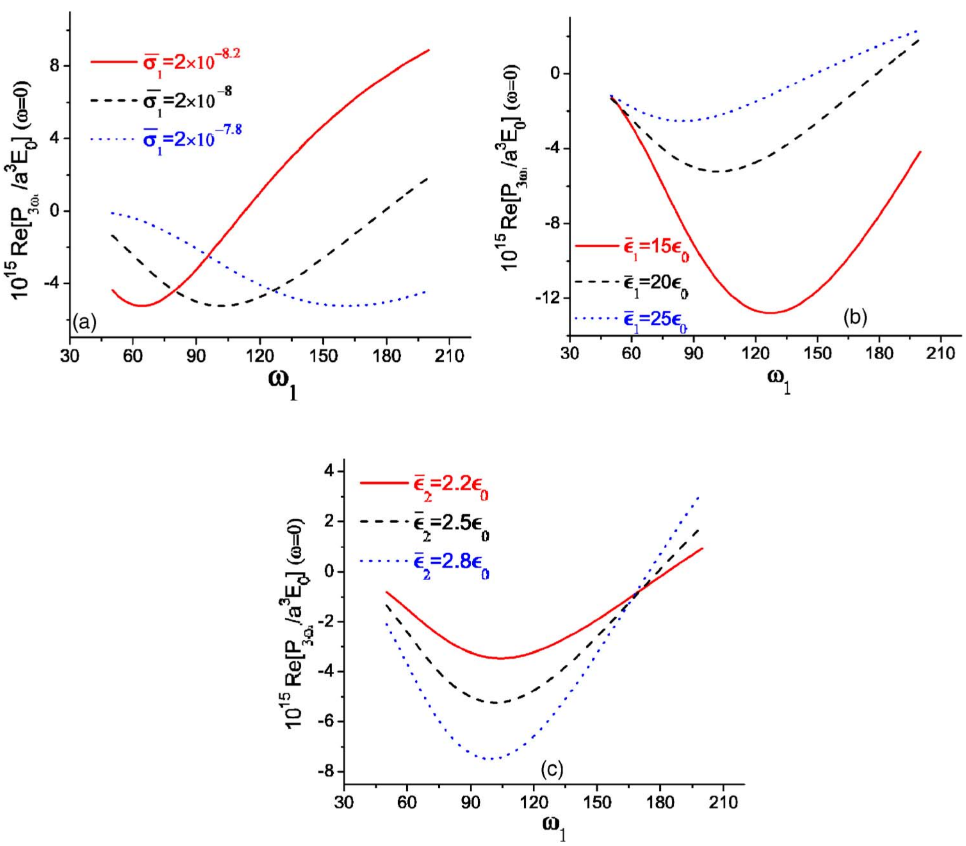

FIG. 8. (Color online) With dispersion. Same as Fig. 6, but for a dc electric field (ω=0), and for different (a) $\bar{\sigma}_1$, (b) $\bar{\epsilon}_1$, and (c) $\bar{\epsilon}_2$. Parameters: $\epsilon_1=20\epsilon_0$, $\epsilon_2=2.5\epsilon_0$, $\sigma_1=2\times10^{-8}$ S/m, $\sigma_2=1.0\times10^{-13}$ S/m, and $\chi E_0^2=0.7\epsilon_0$, if they are not the variables.

$$\mathbf{D}_1 = \epsilon_1\mathbf{E}_1 + \chi|\mathbf{E}_1|^2\mathbf{E}_1 + \eta|\mathbf{E}_1|^4\mathbf{E}_1 \equiv \tilde{\epsilon}_1\mathbf{E}_1, \qquad \text{(B1)}$$

where η denotes the fifth-order nonlinear coefficient. Similar to the definition $|\mathbf{E}_1|^2=\mathbf{E}_1\cdot\mathbf{E}_1$, we adopt the definition $|\mathbf{E}_1|^4=(\mathbf{E}_1\cdot\mathbf{E}_1)(\mathbf{E}_1\cdot\mathbf{E}_1)$.

Comparing with Eq. (2), it is not difficult to derive the contribution of η to each harmonic terms. In this case, the local electric field inside a particle is given by

$$E_1 = \frac{3\epsilon_2}{\epsilon_1 + \chi E_1^2 + \eta E_1^4 + 2\epsilon_2}E_f. \qquad \text{(B2)}$$

And we expand E_1 by χE_1^2 and ηE_1^4. So we have

TABLE I. List of τ and ω_1^*. Here τ was calculated according to Eq. (24) and ω_1^* was extracted from the curves of Fig. 8. The two parameters satisfy $\tau\omega_1^*=1$ [Eq. (23)].

	τ (s)	ω_1^* (rad/s)
$\bar{\sigma}_1=2\times10^{-8.2}$ S/m	0.0156	64.1
2×10^{-8} S/m	0.0099	101.7
$2\times10^{-7.8}$ S/m	0.0062	161.1
$\bar{\epsilon}_1=15\epsilon_0$	0.0078	127.1
$20\epsilon_0$	0.0099	101.7
$25\epsilon_0$	0.0118	84.7
$\bar{\epsilon}_2=2.2\epsilon_0$	0.0096	104.2
$2.5\epsilon_0$	0.0099	101.7
$2.8\epsilon_0$	0.0101	99.3

$$E_1 = \frac{3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2}{2(\epsilon_1+2\epsilon_2)}E_0 - \frac{3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2\chi E_1^2}{2(\epsilon_1+2\epsilon_2)^2}E_0 - \frac{3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2\eta E_1^4}{2(\epsilon_1+2\epsilon_2)^2}E_0. \qquad \text{(B3)}$$

Then the substitution of the linear part of E_1 into Eq. (B3) yields

$$E_1 = \frac{3e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2 E_0}{2(\epsilon_1+2\epsilon_2)} - \frac{27e^{-3it(\omega+\omega_1)}(1+e^{2it\omega})^3\epsilon_2^3\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4} - \frac{243e^{-5it(\omega+\omega_1)}(1+e^{2it\omega})^5\epsilon_2^5\eta E_0^5}{32(\epsilon_1+2\epsilon_2)^6}. \qquad \text{(B4)}$$

By expanding this equation, we get E_1 in the form of harmonics:

$$\begin{aligned} E_1 = {} & E_{\omega+\omega_1}e^{-it(\omega+\omega_1)} + E_{\omega-\omega_1}e^{it(\omega-\omega_1)} + E_{3\omega+3\omega_1}e^{-3it(\omega+\omega_1)} \\ & + E_{3\omega-3\omega_1}e^{3it(\omega-\omega_1)} + E_{\omega+3\omega_1}e^{-it(\omega+3\omega_1)} + E_{\omega-3\omega_1}e^{it(\omega-3\omega_1)} \\ & + E_{5\omega+5\omega_1}e^{-5it(\omega+\omega_1)} + E_{5\omega-5\omega_1}e^{5it(\omega-\omega_1)} \\ & + E_{3\omega+5\omega_1}e^{-it(3\omega+5\omega_1)} + E_{3\omega-5\omega_1}e^{it(3\omega-5\omega_1)} \\ & + E_{\omega+5\omega_1}e^{-it(\omega+5\omega_1)} + E_{\omega-5\omega_1}e^{it(\omega-5\omega_1)}, \end{aligned} \qquad \text{(B5)}$$

where the harmonic coefficients are given by

$$E_{\omega\pm\omega_1} = \frac{3\epsilon_2 E_0}{2(\epsilon_1+2\epsilon_2)},$$

$$E_{3\omega\pm3\omega_1} = -\frac{27\epsilon_2^3\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$E_{\omega\pm3\omega_1} = -\frac{81\epsilon_2^3\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$E_{5\omega\pm5\omega_1} = -\frac{243\epsilon_2^5\eta E_0^5}{32(\epsilon_1+2\epsilon_2)^6},$$

$$E_{3\omega\pm5\omega_1} = -\frac{1215\epsilon_2^5\eta E_0^5}{32(\epsilon_1+2\epsilon_2)^6},$$

$$E_{\omega\pm5\omega_1} = -\frac{1215\epsilon_2^5\eta E_0^5}{16(\epsilon_1+2\epsilon_2)^6}. \quad \text{(B6)}$$

With the dielectric constant $\tilde{\epsilon}_1$ given by Eq. (B1), the induced dipole moment is written as

$$P = \epsilon_2 \frac{\epsilon_1 + \chi E_1^2 + \eta E_1^4 - \epsilon_2}{\epsilon_1 + \chi E_1^2 + \eta E_1^4 + 2\epsilon_2} a^3 E_f. \quad \text{(B7)}$$

Then we expand P by χE_1^2 and ηE_1^2, so we get

$$P = \frac{a^3 e^{-it(\omega+\omega_1)}(1+e^{2it\omega})(\epsilon_1-\epsilon_2)\epsilon_2}{2(\epsilon_1+2\epsilon_2)}E_0 + \frac{3a^3 e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2^2\chi E_1^2}{2(\epsilon_1+2\epsilon_2)^2}E_0 + \frac{3a^3 e^{-it(\omega+\omega_1)}(1+e^{2it\omega})\epsilon_2^2\eta E_1^4}{2(\epsilon_1+2\epsilon_2)^2}E_0. \quad \text{(B8)}$$

Now the linear term of E_1 is introduced into the equation above. So P is given by

$$P = \frac{a^3 e^{-it(\omega+\omega_1)}(1+e^{2it\omega})(\epsilon_1-\epsilon_2)\epsilon_2 E_0}{2(\epsilon_1+2\epsilon_2)} + \frac{27a^3 e^{-3it(\omega+\omega_1)}(1+e^{2it\omega})^3\epsilon_2^4\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4} + \frac{243a^3 e^{-5it(\omega+\omega_1)}(1+e^{2it\omega})^5\epsilon_2^6\eta E_0^5}{32(\epsilon_1+2\epsilon_2)^6}. \quad \text{(B9)}$$

We expand Eq. (B9) to get P in the terms of harmonics:

$$P = P_{\omega+\omega_1}e^{-it(\omega+\omega_1)} + P_{\omega-\omega_1}e^{it(\omega-\omega_1)} + P_{3\omega+3\omega_1}e^{-3it(\omega+\omega_1)} + P_{3\omega-3\omega_1}e^{3it(\omega-\omega_1)} + P_{\omega+3\omega_1}e^{-it(\omega+3\omega_1)} + P_{\omega-3\omega_1}e^{it(\omega-3\omega_1)} + P_{5\omega+5\omega_1}e^{-5it(\omega+\omega_1)} + P_{5\omega-5\omega_1}e^{5it(\omega-\omega_1)} + P_{3\omega+5\omega_1}e^{-it(3\omega+5\omega_1)} + P_{3\omega-5\omega_1}e^{it(3\omega-5\omega_1)} + P_{\omega+5\omega_1}e^{-it(\omega+5\omega_1)} + P_{\omega-5\omega_1}e^{it(\omega-5\omega_1)}, \quad \text{(B10)}$$

where the harmonic coefficients are

$$P_{\omega\pm\omega_1} = \frac{a^3(\epsilon_1-\epsilon_2)\epsilon_2 E_0}{2(\epsilon_1+2\epsilon_2)},$$

$$P_{3\omega\pm3\omega_1} = \frac{27a^3\epsilon_2^4\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$P_{\omega\pm3\omega_1} = \frac{81a^3\epsilon_2^4\chi E_0^3}{8(\epsilon_1+2\epsilon_2)^4},$$

$$P_{5\omega\pm5\omega_1} = \frac{243a^3\epsilon_2^6\eta E_0^5}{32(\epsilon_1+2\epsilon_2)^6},$$

$$P_{3\omega\pm5\omega_1} = \frac{1215a^3\epsilon_2^6\eta E_0^5}{32(\epsilon_1+2\epsilon_2)^6},$$

$$P_{\omega\pm5\omega_1} = \frac{1215a^3\epsilon_2^6\eta E_0^5}{16(\epsilon_1+2\epsilon_2)^6}. \quad \text{(B11)}$$

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