Nonlinear ac responses of electro-magnetorheological fluids

J. P. Huang
Department of Physics, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong and Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany

K. W. Yu
Department of Physics, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong

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We apply a Langevin model to investigate the nonlinear ac responses of electro-magnetorheological (ERMR) fluids under the application of two crossed dc magnetic (z axis) and electric (x axis) fields and a probing ac sinusoidal magnetic field. We focus on the influence of the magnetic fields which can yield nonlinear behaviors inside the system due to the particles with a permanent magnetic dipole moment. Based on a perturbation approach, we extract the harmonics of the magnetic field and orientational magnetization analytically. To this end, we find that the harmonics are sensitive to the degree of anisotropy of the structure as well as the field frequency. Thus, it is possible to real-time-monitor the structure transformation of ERMR fluids by detecting the nonlinear ac responses. [DOI: 10.1063/1.1798912]

I. INTRODUCTION

Electro-magnetorheological (ERMR) fluids[1,2] behave as both electrorheological (ER) fluids[3–6] and magnetorheological (MR) fluids[7–9] and are, in general, particle suspensions where the particles have large dielectric constants and permanent magnetic moments. In fact, there exist a variety of particles which can be polarized by both electric field and magnetic field.[10] For instance, there is one candidate for this system, namely, a suspension of titanium-coated iron particles in a silicon oil.[11] As the external electric field or magnetic field exceeds a threshold, ER or MR fluids turn into a semisolid, the ground state of which is a body-centered-tetragonal (bct) lattice. For an ERMR solid, one[1] proposed that a structure transformation of the ERMR solid from the bct lattice to some other lattices can appear when a magnetic field is simultaneously applied perpendicular to the electric field. To one's interest, such a structure transformation from bct to face-centered-cubic (fcc) lattice was experimentally observed as the ratio between the magnetic and the electric fields exceeded a minimum value.[2] Recently, one of the present authors showed that an alternative structure transformation from the bct structure to the fcc can appear under the application of electric fields only.[12]

Electric or magnetic fields of high strength applied to the ERMR solid produce a nonlinearity in the dependence of the polarization or magnetization on the field strength. Consequently, in the presence of an ac electric or magnetic field, the electrical or magnetic response will in general consist of ac fields at frequency of the higher-order harmonics.[13–16] A convenient method of probing the nonlinear characteristics of the composite is to measure the harmonics of the nonlinear polarization (or magnetization) under the application of a sinusoidal ac field.[17] In this case, the strength of the nonlinear polarization or magnetization should be reflected in the magnitude of the harmonics. For extracting such harmonics, the perturbation approach[18,19] and self-consistent method[19,20] can be used.

For the present system under consideration, the nonlinearity can be caused to appear by two effects, namely, normal saturation and anomalous saturation. In detail, the normal saturation arises from the higher terms of the Langevin function at large field intensities.[21] In contrast, the anomalous saturation results from the equilibrium between entities with higher and lower dipole moments which is shifted under the influence of the field.[21] Our formalism will hold for the coupling between the normal saturation and anomalous saturation.

In the present paper, to investigate the structural effect on the nonlinear ac responses of ERMR solids, we shall apply a Langevin model to derive the orientational magnetization as well as the magnetic field inside the ERMR solid. In this connection, the perturbation approach will be used to extract the harmonics of the magnetic field and orientational magnetization.

This paper is organized as follows. In Sec. II, we apply the Langevin model to derive the orientational magnetization as well as the magnetic field inside the ERMR solid and calculate the effective permeability of the ERMR solid by using a generalized Clausius-Mossotti equation. Also, based on the perturbation approach, we extract the harmonics of the magnetic field and orientational magnetization analytically. In Sec. III, we numerically investigate these harmonics as a function of the degree of anisotropy of the structure, as well as the frequency of the ac magnetic field. This is followed by a discussion and conclusion in Sec. IV.

II. FORMALISM

A. Nonlinear characteristics

Under the application of a strong magnetic field along z axis, a nonlinear characteristic can appear in the ERMR fluid, due to the particles with a permanent magnetic dipole moment p_0. Accordingly, the dependence of the magnetic induction $\mathbf{B}$ on the field $\mathbf{H}_0$ will be nonlinear,[21]

$$\mathbf{B}=\mu_e\mathbf{H}_0+4\pi\chi H_0^2\mathbf{H}_0, \tag{1}$$

where χ and μ_e stand for the nonlinear susceptibility and effective permeability for the longitudinal field case, respectively. In this case, the effective permeability μ_e is determined by the generalized Clausius-Mossotti equation[12]

$$\frac{g_L(\mu_e-\mu_2)}{\mu_2+g_L(\mu_e-\mu_2)}=\frac{4\pi}{3}N_1\left(\alpha_1+\frac{p_0^2}{3k_BT}\frac{1}{1+i2\pi f\tau_1}\right), \tag{2}$$

where μ_2 represents the permeability of the host fluid, N_1 the number density of the particles, k_B the Boltzmann constant, T the absolute temperature, f the frequency of the applied magnetic field, τ_1 the relaxation time of the particles, and α_1 the magnetizability of the particles. In Eq. (2), the longitudinal demagnetizing factor g_L deserves a thorough consideration. For an isotropic array of magnetic dipoles, the demagnetizing factor will be diagonal with the diagonal element $g_L=1/3$. However, in an anisotropic array such as ERMR solids, the demagnetizing factor can still be diagonal, but it deviates from 1/3. In fact, the degree of anisotropy of the system is just measured by how g_L is deviated from 1/3. It is worth noting that $g_L\leqslant 1/3$ in the present longitudinal field case. Furthermore, there is a sum rule for the factors, $g_L+2g_T=1$,[19,22] where g_T denotes the transverse demagnetizing factor. Such factors were measured by means of computer simulations.[23,24] Thus, to investigate the anisotropic structural information of the array, we have to modify the Clausius-Mossotti equation accordingly by including the demagnetizing factor. The substitution of $g_L(=g_T)=1/3$ into Eq. (2) yields the usual (isotropic) Clausius-Mossotti equation, which does not include the particle-particle interaction. In fact, when we studied the field-induced structure transformation in ER solids, we developed the generalized Clausius-Mossotti equation[12] by introducing a local-field factor β' which reflects the particle-particle interaction between the particles in a lattice. In detail, the generalized Clausius-Mossotti approach [Eq. (2)] is a self-consistent determination of the local field due to a lattice of dipole moments. That is, Eq. (2) should be expected to include the particle-particle interaction, and the degree of the particle-particle interaction depends on how much g_L deviates from 1/3 (note that $g_L=\beta'/3$). Indeed our numerical results will also show that as g_L deviates from 1/3 more (namely, more and more particle chains are formed, and the particle-particle interaction becomes more and more strong, too), the obtained harmonics become more large accordingly; see Figs. 1 and 2. These harmonics just reflect the magnitude of the nonlinear susceptibility, as expected.

In Eq. (2), the term $p_0^2/(3k_BT)$ results from the average contribution of the permanent magnetic dipole moment to the average value of the work required to bring a particle into the field H_0. More precisely, the mean value of the component of the permanent dipole moment in the direction of the field is given by

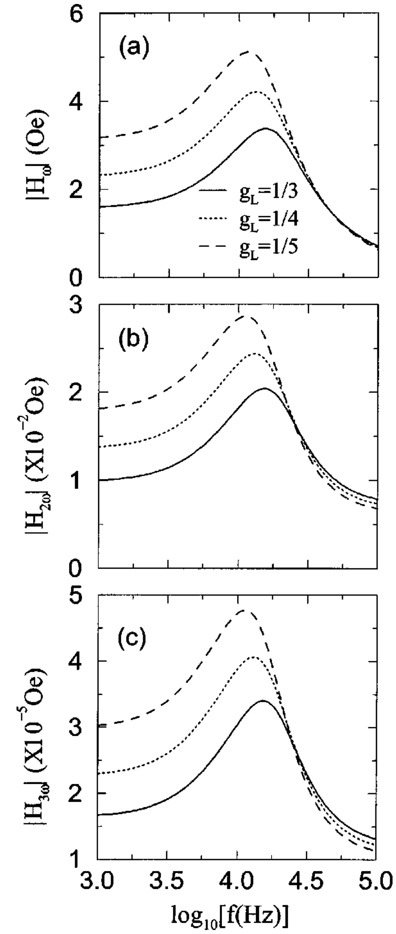

FIG. 1. Fundamental, second-order, and third-order harmonics of Fröhlich field as a function of the field frequency for various g_L. Parameter: $T=298$ K.

$$p_0L(\gamma)=\frac{p_0^2}{3k_BT}H_0, \tag{3}$$

with $\gamma=p_0H_0/(k_BT)$. That is, we set the Langevin function

$$L(\gamma)=\gamma/3. \tag{4}$$

Regarding this linear relation in use, we should make some remarks. In the present work, we shall adopt the perturbation approach,[25] which is suitable for weak nonlinearity. In this perturbation approach, it is well established that the effective third-order nonlinear susceptibility can be calculated from the linear field,[26] while the effective higher-order nonlinearity must depend on the nonlinear field.[25] Alternatively, we could adopt the self-consistent method,[19] but the perturbation approach appears to be more convenient for analytic expressions.[19] Thus, for focusing on (weak) third-order nonlinearity, it suffices to use the Clausius-Mossotti equation [Eq. (2)] by taking into account of the linear relation [Eq. (4)] only. Due to the same reason, the contribution from the nonlinear field will be omitted throughout the paper.

In view of Eq. (1), the desired field-dependent incremental permeability μ_H is given by[21]

$$\mu_H=\frac{\partial B}{\partial H_0}=\mu_e+12\pi\chi H_0^2. \tag{5}$$

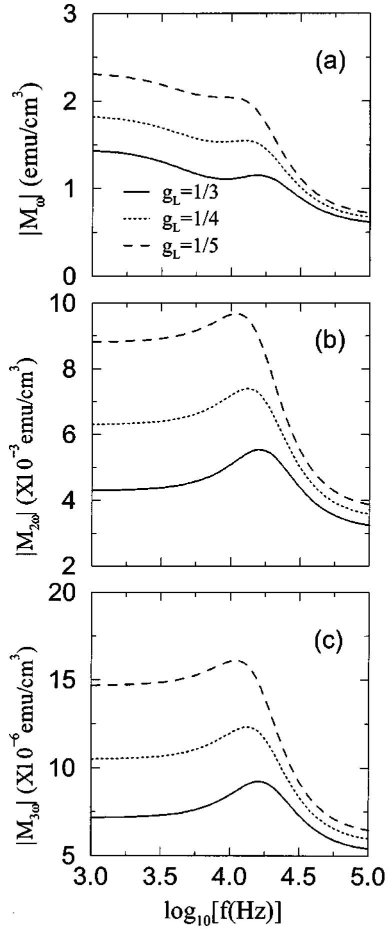

FIG. 2. Fundamental, second-order, and third-order harmonics of the orientational magnetization as a function of the field frequency for various g_L. Parameter: $T=298$ K.

Then, the nonlinear magnetic effect is characterized by $\Delta\mu/H_0^2$,[21]

$$\frac{\Delta\mu}{H_0^2}=\frac{\mu_H-\mu_e}{H_0^2}=12\pi\chi. \tag{6}$$

In the present work, the magnetization can be split up into two parts, i.e., the induced magnetization due to the magnetizability of the particles and the orientation magnetization M_{or} due to the alignment of permanent magnetic dipole moments inside the particles. Let us now consider the orientation magnetization M_{or} of a sphere of volume V, containing n_1 particles with a permanent magnetic dipole moment $\mathbf{p}_d$ [see Eq. (8)] embedded in a continuum with permeability μ_∞ [see Eq. (9)]. The sphere is surrounded by an infinite medium with the same macroscopic properties as the sphere.

The average component in the direction of the field of the magnetic dipole moment due the dipoles in the sphere $\langle \mathbf{M}_d\cdot\mathbf{e}\rangle=VM_{\mathrm{or}}$ is given by

$$\langle \mathbf{M}_d\cdot\mathbf{e}\rangle=VM_{\mathrm{or}}=\frac{\int dX^{n_1}\mathbf{M}_d\cdot\mathbf{e}e^{-u/k_BT}}{\int dX^{n_1}e^{-u/k_BT}}, \tag{7}$$

where $\mathbf{e}$ denotes the unit vector in the direction of the external field and X stands for the set of position and orientation variables of all particles. Here u is the energy related to the dipoles in the sphere, and it consists of three parts: the energy of the dipoles in the external field u_{de}, the magnetostatic interaction energy of the dipoles u_{mi}, the nonmagnetostatic interaction energy between the dipoles u_{nmi} which is responsible for the short-range correlation between orientations and positions of the dipoles. Regarding the particle-particle interaction energy, Eq. (2) should be expected to contain both u_{mi} and u_{nmi} (at least to some extent) as g_L is not equal to 1/3. Nevertheless, for the numerical calculations in the following section, Eqs. (39) and (40) will be used which mean that u_{nmi} is omitted due to the predominant external magnetic field. In Eq. (7), $\mathbf{M}_d$ is given by

$$\mathbf{M}_d=\sum_{i=1}^{n_1}(\mathbf{p}_d)_i, \tag{8}$$

with $\mathbf{p}_d=\mathbf{p}_0(\mu_\infty+2\mu_2)/3\mu_2$, where μ_∞ represents the permeability at frequencies at which the permanent dipoles cannot follow the changes of the field but where the atomic and the electronic magnetizations are still the same as in the static field. Therefore, μ_∞ is the permeability characteristic for the induced magnetization. In practice, μ_∞ can be expressed in the expression containing an intrinsic dispersion,

$$\mu_\infty=\mu_\infty(0)+\frac{\Delta\mu}{1+if/f_c}, \tag{9}$$

where $\mu_\infty(0)$ is the high-frequency limit permeability and $\Delta\mu$ stands for the magnetic dispersion strength with a characteristic frequency f_c.

The external field in this model is equal to the field within a spherical cavity filled with a continuum of permeability μ_∞, while the cavity is situated in a medium with permeability μ_e. This field is called Fröhlich field H_F, as given by the Fröhlich model,[21,27]

$$H_F=\frac{3\mu_e}{2\mu_e+\mu_\infty}H_0+\frac{12\pi\chi\mu_\infty}{(2\mu_e+\mu_\infty)^2}H_0^3. \tag{10}$$

In this equation, higher-order terms have been omitted. Regarding the derivation of the nonlinear Fröhlich field [Eq. (10)], here we would like to add some comments. For the spherical cavity under consideration, the linear cavity field (i.e., linear Fröhlich field) H_F^{lin} can be easily obtained by solving the usual electrostatic equation. That is, it should be the first term of the right-hand side of Eq. (10). Next, to obtain the present nonlinear Frölich field H_F, following Ref. 21 we took one step forward to develop H_F^{lin} in a Taylor series around μ_e neglecting terms in H_0^4 and higher powers of H_0. It is worth noting that the higher powers of H_0 can also contribute to the resulting harmonics. However, this contribution is small enough to be neglected.

Now we use

$$\frac{\partial u}{\partial H_F}=-\mathbf{M}_d\cdot\mathbf{e}. \tag{11}$$

Then, we obtain

$$\langle \mathbf{M}_d\cdot\mathbf{e}\rangle|_{H_F=0}=0, \tag{12}$$

$$\frac{\partial}{\partial_{H_F}}\langle \mathbf{M}_d\cdot\mathbf{e}\rangle=\frac{1}{k_BT}[\langle(\mathbf{M}_d\cdot\mathbf{e})^2\rangle-\langle\mathbf{M}_d\cdot\mathbf{e}\rangle^2], \tag{13}$$

$$\frac{\partial}{\partial_{H_F}}\langle \mathbf{M}_d\cdot\mathbf{e}\rangle|_{H_F=0}=\frac{1}{k_BT}\langle M_d^2\rangle_0, \tag{14}$$

$$\begin{aligned}\frac{\partial^3}{\partial H_F^3}\langle \mathbf{M}_d\cdot\mathbf{e}\rangle=\frac{1}{(k_BT)^3}[&\langle(\mathbf{M}_d\cdot\mathbf{e})^4\rangle-3\langle(\mathbf{M}_d\cdot\mathbf{e})^3\rangle\langle\mathbf{M}_d\cdot\mathbf{e}\rangle\\&+6\langle(\mathbf{M}_d\cdot\mathbf{e})^2\rangle\langle\mathbf{M}_d\cdot\mathbf{e}\rangle^2-3\langle(\mathbf{M}_d\cdot\mathbf{e})^2\rangle^2\\&-6\langle\mathbf{M}_d\cdot\mathbf{e}\rangle^4+6\langle\mathbf{M}_d\cdot\mathbf{e}\rangle^2\langle(\mathbf{M}_d\cdot\mathbf{e})^2\rangle\\&-\langle\mathbf{M}_d\cdot\mathbf{e}\rangle\langle(\mathbf{M}_d\cdot\mathbf{e})^3\rangle],\end{aligned} \tag{15}$$

$$\frac{\partial^3}{\partial H_F^3}\langle \mathbf{M}_d\cdot\mathbf{e}\rangle|_{H_F=0}=\frac{1}{15(k_BT)^3}[3\langle M_d^4\rangle_0-5\langle M_d^2\rangle_0^2]. \tag{16}$$

Note the subscript 0 indicates the absence of the field.

To express the macroscopic saturation behavior in terms of microscopic quantities, one should take into account the higher derivatives of the average moment such that

$$\begin{aligned}\langle \mathbf{M}_d\cdot\mathbf{e}\rangle&=\frac{\partial\langle\mathbf{M}_d\cdot\mathbf{e}\rangle}{\partial H_F}\bigg|_{H_F=0}H_F+\frac{1}{6}\frac{\partial^3\langle\mathbf{M}_d\cdot\mathbf{e}\rangle}{\partial H_F^3}\bigg|_{H_F=0}H_F^3\\&=\frac{\mu_e}{2\mu_e+\mu_\infty}\frac{\langle M_d^2\rangle_0}{k_BT}H_0+\frac{4\pi\chi\mu_\infty}{(2\mu_e+\mu_\infty)^2}\frac{\langle M_d^2\rangle_0}{k_BT}H_0^3\\&\quad+\frac{27\mu_e^3}{(2\mu_e+\mu_\infty)^3}\frac{3\langle M_d^4\rangle_0-5\langle M_d^2\rangle_0^2}{90(k_BT)^3}H_0^3,\end{aligned} \tag{17}$$

where higher-order terms than third-order have been neglected.

In addition, we have a general relation

$$\langle \mathbf{M}_d\cdot\mathbf{e}\rangle=VM_{\mathrm{or}}=\frac{\mu_e-\mu_\infty}{4\pi}VH_0+\chi VH_0^3. \tag{18}$$

In view of Eq. (6) and the terms in H_0^3 and H_0, we obtain

$$\begin{aligned}\frac{\Delta\mu}{H_0^2}=&\frac{18\pi}{5(k_BT)^3}\frac{\mu_e^4}{(2\mu_e+\mu_\infty)^2(2\mu_e^2+\mu_\infty^2)}\\&\times[3(\langle M_d^4\rangle_0/V)-5(\langle M_d^2\rangle_0^2/V)].\end{aligned} \tag{19}$$

On the other hand, we may write

$$\frac{\langle M_d^2\rangle_0}{V}=\left(\frac{\mu_\infty+2\mu_2}{3\mu_2}\right)^2\left[\frac{n_1}{V}p_0^2\sum_{j=1}^{n_1}\langle\cos\theta_{ij}\rangle\right], \tag{20}$$

$$\begin{aligned}\frac{\langle M_d^4\rangle_0}{V}=&\left(\frac{\mu_\infty+2\mu_2}{3\mu_2}\right)^4\\&\times\left[\frac{n_1}{V}p_0^4\sum_{j=1}^{n_1}\left\langle\cos\theta_{ij}\sum_{r=1}^{n_1}\sum_{s=1}^{n_1}\cos\theta_{rs}\right\rangle\right].\end{aligned} \tag{21}$$

In the right-hand sides of both Eqs. (20) and (21), the $\langle\cdots\rangle$'s denote

$$\langle\cdots\rangle=\int\frac{\int dX^{n_1-i}\cdots e^{-u/k_BT}}{\int dX^{n_1}e^{-u/k_BT}}dX^i. \tag{22}$$

Hence the nonlinear magnetic increment $\Delta\mu/H_0^2$ [Eq. (19)] can be explicitly expressed.

B. Nonlinear magnetization and high-order harmonics

1. Longitudinal field

For the longitudinal field case, there is $H_0(t)=H_{\mathrm{dc}}+H_{\mathrm{ac}}(t)=H_{\mathrm{dc}}+H_{\mathrm{ac}}\sin\omega t$ along z axis, with $\omega=2\pi f$. Here H_{dc} denotes the dc field which induces the anisotropic structure in the ERMR solid and $H_{\mathrm{ac}}(t)$ stands for a sinusoidal ac field. Then, the Fröhlich field is a superposition of odd- and even-order harmonics such that

$$H_F=H_F^{(\mathrm{dc})}+H_\omega\sin\omega t+H_{2\omega}\cos 2\omega t+H_{3\omega}\sin 3\omega t+\cdots. \tag{23}$$

Accordingly, the orientational magnetization contains harmonics as

$$M_{\mathrm{or}}=M_{\mathrm{or}}^{(\mathrm{dc})}+M_\omega\sin\omega t+M_{2\omega}\cos 2\omega t+M_{3\omega}\sin 3\omega t+\cdots, \tag{24}$$

where the harmonics (H_ω, $H_{2\omega}$, $H_{3\omega}$, M_ω, $M_{2\omega}$, and $M_{3\omega}$) and the dc components ($H_F^{(\mathrm{dc})}$ and $M_{\mathrm{or}}^{(\mathrm{dc})}$) can be expressed as

$$H_F^{(\mathrm{dc})}=H_{\mathrm{dc}}I_1+\tfrac{3}{2}H_{\mathrm{ac}}^2H_{\mathrm{dc}}I_3+H_{\mathrm{dc}}^3I_3, \tag{25}$$

$$H_\omega=H_{\mathrm{ac}}I_1+\tfrac{3}{4}H_{\mathrm{ac}}^3I_3+3H_{\mathrm{ac}}H_{\mathrm{dc}}^2I_3, \tag{26}$$

$$H_{2\omega}=-\tfrac{3}{2}H_{\mathrm{ac}}^2H_{\mathrm{dc}}I_3, \tag{27}$$

$$H_{3\omega}=-\tfrac{1}{4}H_{\mathrm{ac}}^3I_3, \tag{28}$$

$$M_{\mathrm{or}}^{(\mathrm{dc})}=H_{\mathrm{dc}}J_1+\tfrac{3}{2}H_{\mathrm{ac}}^2H_{\mathrm{dc}}\chi+H_{\mathrm{dc}}^3\chi, \tag{29}$$

$$M_\omega=H_{\mathrm{ac}}J_1+\tfrac{3}{4}H_{\mathrm{ac}}^3\chi+3H_{\mathrm{ac}}H_{\mathrm{dc}}^2\chi, \tag{30}$$

$$M_{2\omega}=-\tfrac{3}{2}H_{\mathrm{ac}}^2H_{\mathrm{dc}}\chi, \tag{31}$$

$$M_{3\omega}=-\tfrac{1}{4}H_{\mathrm{ac}}^3\chi, \tag{32}$$

with $I_1=3\mu_e/(2\mu_e+\mu_\infty)$, $I_3=12\pi\chi\mu_\infty/(2\mu_e+\mu_\infty)^2$, and $J_1=(1/4\pi)(\mu_e-\mu_\infty)$. In the above derivation, we used two identities, i.e., $\sin^2\omega t=(1-\cos 2\omega t)/2$ and $\sin^3\omega t=(3/4)\sin\omega t-(1/4)\sin 3\omega t$.

2. Transverse field

For the transverse field case, only a sinusoidal electric field $H_0(t)=H_{\mathrm{ac}}\sin\omega t$ is applied along x axis. In this connection, the Fröhlich field is a superposition of odd-order harmonics,

$$H_F=H_\omega\sin\omega t+H_{3\omega}\sin 3\omega t+\cdots. \tag{33}$$

Accordingly, the orientational magnetization contains harmonics as

$$M_{\mathrm{or}}=M_\omega\sin\omega t+M_{3\omega}\sin 3\omega t+\cdots. \tag{34}$$

In Eqs. (33) and (34), the harmonics H_ω, $H_{3\omega}$, M_ω, and $M_{3\omega}$ can be expressed as

$$H_\omega=H_{\mathrm{ac}}I_1'+\tfrac{3}{4}H_{\mathrm{ac}}^3I_3', \tag{35}$$

$$H_{3\omega} = -\tfrac{1}{4}H_{ac}^3 I_3', \tag{36}$$

$$M_{\omega} = H_{ac} J_1' + \tfrac{3}{4}H_{ac}^3 \chi', \tag{37}$$

$$M_{3\omega} = -\tfrac{1}{4}H_{ac}^3 \chi' \tag{38}$$

with $I_1' = 3\mu_e'/(2\mu_e' + \mu_\infty)$, $I_3' = 12\pi\chi'\mu_\infty/(2\mu_e' + \mu_\infty)^2$, and $J_1' = (1/4\pi)(\mu_e' - \mu_\infty)$, where μ_e' and χ' denote the effective permeability and nonlinear susceptibility in the transverse field case. For the transverse field case, χ' is induced to appear by the moderate ac magnetic field only. Thus, in view of Eqs. (7) and (8), we find $\chi' \ll \chi$ (χ is caused to appear by both the moderate ac and high dc magnetic fields in the longitudinal field case, as mentioned above). As a result, the third-order harmonics of the Fröhlich field and orientational magnetization are much smaller in the transverse field case than in the longitudinal field case. In this regard, for the following numerical calculations, we shall focus on the longitudinal field case only.

III. NUMERICAL RESULTS

Now we are in a position to do some numerical calculations. For the numerical calculation, we use

$$\sum_{j=1}^{n_2} \langle \cos \theta_{ij} \rangle = 1, \tag{39}$$

$$\sum_{j=1}^{n_2} \left\langle \cos \theta_{ij} \sum_{r=1}^{n_2} \sum_{s=1}^{n_2} \cos \theta_{rs} \right\rangle = \frac{1}{3}(5n_1 - 2). \tag{40}$$

Equations (39) and (40) imply that the correlation between the dipole moments of the particles and in turn the anomalous saturation are neglected in the sense that the current anomalous saturation is much more weak. This is reasonable because, in the presence of a magnetic field, the permanent magnetic moments of the particles are easily directed along the field. Therefore, the equilibrium between the higher dipole moment (of particle chains) and the lower dipole moment (of particle chains) might not be able to predict significant nonlinearity, when compared to the normal saturation.

Next, we take one step forward to obtain the nonlinear magnetic increment

$$\frac{\Delta\mu}{H_0^2} = \frac{-36\pi N_1 p_0^4}{5(k_B T)^3} \frac{\mu_e^4}{(2\mu_e + \mu_\infty)^2 (2\mu_e^2 + \mu_\infty^2)} \left(\frac{\mu_\infty + 2\mu_2}{3\mu_2} \right)^4. \tag{41}$$

For numerical calculations, we take the following parameters: $H_{dc} = 100$ Oe, $H_{ac} = 1$ Oe, $\mu_\infty(0) = 1.1$, $\Delta\mu = 8$, $f_c = 4.8 \times 10^3$ Hz, $\mu_2 = 1$, $p_0 = 10^{-13}$ emu, $\tau_1 = 4.8 \times 10^{-7}$ s, and $\alpha_1 = 10^{-10}\,\text{cm}^{-3}$. In addition, the radius of particle is taken to be 5 μm, and the volume fraction of the particles is 0.2.

Figure 1 shows the fundamental, second-order, and third-order harmonics of the Fröhlich field as a function of the field frequency for various g_L in the longitudinal field case. In this figure, a peak is observed always due to the existence of an intrinsic dispersion [Eq. (9)]. In particular, as the system changes from isotropic case ($g_L = 1/3$) to anisotropic ($g_L \neq 1/3$) because of the appearance of the particle chains, the harmonics of the field can be changed accordingly. In detail, stronger anisotropy (namely, decreasing the longitudinal demagnizing factor g_L) leads to larger harmonics, especially in the low-frequency region. Similar effect can be shown in Fig. 2 where the harmonics of the orientational magnetization in the longitudinal field case are investigated for various g_L as well. However, the fundamental harmonic of the orientation magnetization [see Fig. 2(a)] behaves in a different way from that of the Fröhlich field [see Fig. 1(a)]. In detail, as the frequency increases, this fundamental harmonic decreases first, then increases, and after reaching a peak they decreases again.

For the transverse field case, it could be concluded that as the demagnetizing factor g_L decreases, both the fundamental and third-order harmonics of the Fröhlich field and orientation magnetization are caused to decrease accordingly, which is just opposite to those obtained from the longitudinal field case (no figures shown here). The reason is that there is a sum rule between g_L and g_T, $g_L + 2g_T = 1$.

In other words, for the longitudinal field case, besides the odd-order harmonics, the even-order harmonics are also induced to appear due to the coupling between the applied dc and ac magnetic fields along z axis [see Figs. 1 and 2, or Eqs. (23)–(32)] even though only the cubic nonlinearity [Eq. (1)] is considered due to the virtue of symmetry of the system. On the other hand, for the transverse field case (i.e., there is only a single ac magnetic field applied along x axis), in view of the cubic nonlinearity [Eq. (1)] of interest, only the odd-order harmonics are induced, as already predicted by Eqs. (33)–(38).

In addition, even though there is no particle-particle interaction (i.e., $u_{mi} = u_{nmi} = 0$) as $g_L = 1/3$, the nonlinear behavior due to the normal saturation could still be induced to occur because of the presence of external fields, i.e., $u_{de} \neq 0$. This is the reason why the harmonics shown in Figs. 1 and 2 are nonzero at $g_L = 1/3$.

From Figs. 1 and 2, we find that the second-order harmonics of the Fröhlich field and orientational magnetization are of three orders of magnitude larger than the corresponding third-order harmonics. Thus, to monitor the structure transformation of ERMR solids, it is more attractive to detect the second-order harmonics than the third-order.

Finally, we display the temperature effect on the harmonics of the Fröhlich field and orientational magnetization in Fig. 3 and Fig. 4, respectively. It is shown that decreasing the temperature T causes all the harmonics to increase because of the change in the Fröhlich field.

IV. DISCUSSION AND CONCLUSION

Here some comments are in order. Based on a Langevin model, we have investigated the nonlinear ac responses of ERMR solids which are subjected to a structural transformation. In the present work, we have focused on the effect of the magnetic fields on the nonlinear ac responses. In fact, for the ERMR solid, the influence of electric fields can be investigated as well. However, the present Langevin model is invalid for this purpose because there is usually no permanent electric dipole moment inside the suspended particles, except for ferroelectric particles. To one's interest, when the particles own a nonlinear characteristic inside them, the electric-

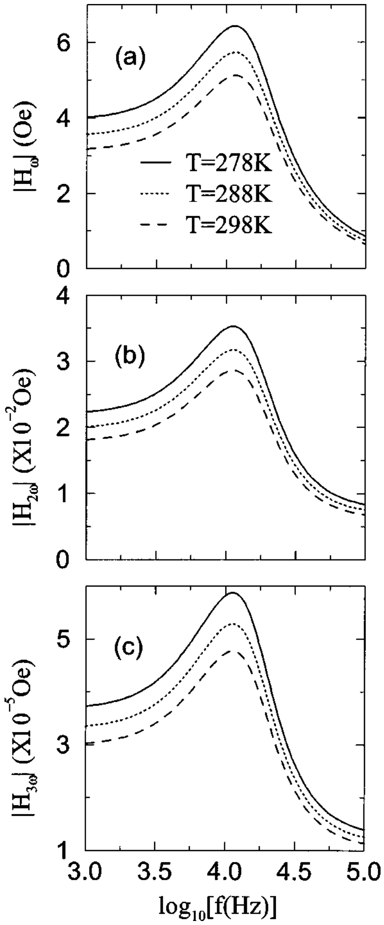

FIG. 3. Same as Fig. 1, but for different temperatures. Parameter: $g_L = 1/5$.

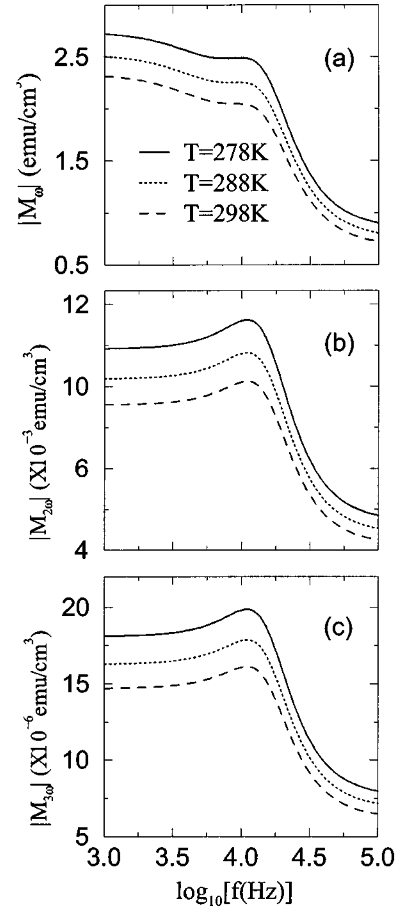

FIG. 4. Same as Fig. 2, but for different temperatures. Parameter: $g_L = 1/5$.

field effect on the nonlinear ac responses can still be discussed, based on the perturbation approach and self-consistent method[19] accompanying with an effective medium theory such as the Maxwell-Garnett approximation.

In this paper, the Fröhlich field is actually an effective field, which is similar to the Lorentz local field. The latter is only defined for induced dipole moments, while the former is introduced to deal with permanent dipole moments.

The present consideration can be extended to ferrofluids, which are a suspension[28] containing ferromagnetic particles embedded in a carrier liquid. However, the particles in a ferrofluid possess a much smaller size than in the ERMR solid. Also, the particles can form chains inside ferrofluids under the presence of a moderate magnetic field, without the need of a strong magnetic field. For ERMR solids, a strong magnetic field has to be used to induce the formation of particle chains.

Finally, since the particles in ERMR solids are located very close, it is instructive to take into account the effect of multipolar interactions between the particles[29–31] on the nonlinear ac responses.

To sum up, we have applied a Langevin model to investigate the nonlinear ac responses of ERMR solids. For the longitudinal field case, it has been shown that both even- and odd-order harmonics are induced to occur. In contrast, only the odd-order harmonics appear for the transverse field case. Moreover, these harmonics can be affected by the degree of anisotropy of the ERMR solid, as well as the field frequency. In particular, the second-order harmonics are of several orders of magnitude larger than the corresponding third-order. Thus, it is possible to real-time-monitor the structural transformation of ERMR solids by detecting the nonlinear ac reponses.

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