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New mechanism for harmonic generation in magnetorheological fluids

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Abstract

We present a new mechanism for harmonic generation in magnetorheological (MR) fluids, which differs from the usual ones due to magnetic anisotropy existing in amorphous ferromagnetic materials. Based on thermodynamics, we derive the incremental magnetic susceptibility of the MR fluid which is partially situated in a nonuniform magnetic field, and further extract the desired harmonics analytically. By measuring such harmonics, it seems possible to monitor the structure of MR fluids.

1. Introduction

During the last decades, the structure of colloidal suspensions has received much attention in scientific research [1–6]. Magnetorheological (MR) fluids [5, 6] contain usually ferromagnetic particles (iron) in carrier liquids (e.g., oil), and have gained much attention due to their potential applications, ranging from shock-absorbers for cars to cancer therapy [7]. All of these applications are due to the novel property of MR fluids that switch from a liquid state to a semi-solid, the ground state of which is a body-centred tetragonal structure [6]. It is known that the existence of magnetic anisotropy in amorphous ferromagnetic materials can induce harmonics in the magnetization [8]. Also, the determination of the anisotropy field distribution and in turn the magnetic anisotropy distribution is made by the second harmonic of the magnetization [8] measured perpendicularly to the easy magnetization direction, in the range from saturation to remanence, for a number of magnetic recording systems, provided that the easy axes are oriented at right angles to the applied magnetic field. This kind of magnetic anisotropy is intrinsic, and serves as a physical basis for magnetic recording systems. In contrast, we shall present a quite different mechanism for harmonic generation in MR fluids which are partially subjected to a nonuniform dc magnetic field coupling with an ac field. The current mechanism we exploit is not intrinsic, but results from the circumstance of the system, and the resulting harmonic generations are expected to be an efficient way to monitor the structure of MR fluids.

It is known that an inhomogeneous magnetic field **H** can exert on a ferromagnetic particle with a permanent magnetic dipole moment a translational force. In this regard, if the permanent magnetic moment points in the direction of **H**, the particles will be displaced towards regions of higher field strength. In a macroscopic sample the average moment is in the direction of the field, namely the particles favour orientations with their permanent magnetic moments in the direction of the field. Thus, an inhomogeneous field acting on a macroscopic sample causes a concentration gradient with high concentrations at high field strengths. If a sample is situated partially in a field at a constant pressure, the density of the matter in the field will increase, leading to an increase in the permeability.

The paper is organized as follows. In section 2, we derive the incremental magnetic susceptibility and then extract the harmonics in the magnetization analytically. Also, we do some numerical simulations under different conditions. This paper ends with a discussion and conclusion in section 3.

2. Formalism and numerical results

2.1. Incremental magnetic susceptibilities

If a model MR fluid is situated partially in an inhomogeneous magnetic field at a constant pressure, the density of the ferromagnetic particles will increase due to the interaction between the particles and field, thus yielding an increase in the effective permeability. More precisely, there is a field-affected (FA) area with volume V_c, in which the magnetic field and the magnetic induction are denoted by H_c and B_c, respectively. They both satisfy the magnetostatic equations [9], $\nabla \cdot \mathbf{B}_c = 0$ and $\nabla \times \mathbf{H}_c = 0$, the latter of which implies that the magnetic field $\mathbf{H}_c$ can be expressed as the gradient of a magnetic scalar potential Φ such that $\mathbf{H}_c = -\nabla\Phi$. Under an appropriate boundary condition, the inhomogeneous MR fluid inside the FA area can be represented as a region of volume V_c, surrounded by surface S'. Such a kind of boundary condition is $\Phi = -\mathbf{H} \cdot \mathbf{X}$ on S', which, if the MR fluid within V_c were uniform, would give rise to a magnetic field which is identical to **H** (external field) everywhere within V_c. In fact, even in an inhomogeneous MR fluid under this boundary condition, the volume average of the magnetic field $\langle \mathbf{H}_c \rangle$ within V_c is still equal to that of the external field $\langle \mathbf{H} \rangle$, namely, $\langle \mathbf{H}_c \rangle \equiv (1/V_c) \int \mathbf{H}_c(\mathbf{X})\, \mathrm{d}^3x = \langle \mathbf{H} \rangle$. The considered MR fluid with volume V is situated both inside and outside the FA area at a constant pressure p. It is worth noting that in this case there is no external field outside the FA area (or the external field outside the FA area is weak enough to be neglected).

In the presence of an inhomogeneous magnetic field **H** along z axis, the usual linear relation between the magnetization and magnetic field $\mathbf{M} = \chi\langle \mathbf{H} \rangle$ should be changed to a nonlinear form

$$\mathbf{M} = [\chi + \Delta\chi(\langle \mathbf{H} \rangle)]\langle \mathbf{H} \rangle, \tag{1}$$

where χ denotes the (effective) linear magnetic susceptibility, and $\Delta\chi(\langle \mathbf{H} \rangle)$, a function of $\langle \mathbf{H} \rangle$, stands for the incremental magnetic susceptibility (nonlinear term). That is, for the MR fluid inside the FA area its effective permeability $\tilde{\mu}_\mathrm{e}$ including the incremental part can be expressed as

$$\tilde{\mu}_\mathrm{e} = \mu_\mathrm{e} + 12\pi\Delta\chi(\langle \mathbf{H} \rangle), \tag{2}$$

where μ_e is the effective linear permeability.

On the other hand, based on thermodynamics the permeability $\tilde{\mu}_e$ can be defined as

$$\tilde{\mu}_e = \left(\frac{\partial\langle\mathbf{B}\rangle}{\partial\langle\mathbf{H}\rangle}\right)_{T,p} = \left(\frac{\partial\langle\mathbf{B}\rangle}{\partial\langle\mathbf{H}\rangle}\right)_{T,\rho} + \left(\frac{\partial\langle\mathbf{B}\rangle}{\partial\rho}\right)_{T,\langle\mathbf{H}\rangle}\left(\frac{\partial\rho}{\partial\langle\mathbf{H}\rangle}\right)_{T,p}, \tag{3}$$

where ρ stands for the density of the particles inside the FA area. In equation (3), $(\frac{\partial\langle\mathbf{B}\rangle}{\partial\langle\mathbf{H}\rangle})_{T,\rho} \equiv \mu_e$ is given by the anisotropic Kirkwood–Fröhlich equation

$$\frac{(\mu_e - \mu_{e0})[\mu_e + (\mu_{e0} - \mu_e)\beta^{(L)}]}{\mu_e} = \frac{4\pi N}{3k_B T} g m'^2, \tag{4}$$

with $m' = m_0(\mu_{e0} + 2\mu_2)/(3\mu_2)$, where μ_2 represents the permeability of the carrier fluid, g the Kirkwood correlation factor [10], m_0 the permanent magnetic dipole moment of the particles, N the number density of the particles, k_B the Boltzmann constant, and T the temperature. In equation (4), μ_{e0} stands for the (linear) permeability characteristic for the induced magnetization. In the presence of the magnetic field particle chains can be formed along the z axis in MR fluids, thus yielding structural anisotropy inside the system. In this case, the degree of anisotropy of the system is measured by how $\beta^{(L)}$ deviates from $1/3$ (note that there is $0 < \beta^{(L)} \leqslant 1/3$), where $\beta^{(L)}$ represents the demagnetizing factor in longitudinal field cases. This kind of parameter was measured for electrorheological fluids by using computer simulations [11] and theoretical analysis [12]. In particular, $\beta^{(L)} = 1/3$ corresponds to the isotropic case (i.e., the particles are randomly distributed), which yields the well known (isotropic) Kirkwood–Fröhlich equation [10, 13, 14]. It is worth remarking that there is a sum rule $\beta^{(L)} + 2\beta^{(T)} = 1$ [15, 16], where $\beta^{(T)}$ is the demagnetizing factor in transverse field cases. In this work, we shall focus on longitudinal field cases only. For convenience, $\beta^{(L)}$ will be denoted by β in the following.

In equation (4), the term $m'^2/(3k_B T)$ 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 $\langle\mathbf{H}\rangle$. More precisely, the mean value of the component of the dipole moment in the direction of the field is given by $m'L(\eta) = m'^2\langle H\rangle/(3k_B T)$, with Langevin parameter $\eta = m'\langle H\rangle/(k_B T)$. As a matter of fact, for the weak nonlinearity of interest, it suffices to set the linear Langevin function $L(\eta) = \eta/3$. It is true that the Langevin function can be nonlinear, too, by keeping more terms. However, a perturbation approach [17] can be adopted for weak-nonlinearity cases. In the perturbation approach, it is well established that the effective third-order nonlinearity (namely, the incremental magnetic susceptibility, see equation (8) below) can be calculated from the linear field [18], while the effective higher-order nonlinearity must depend on the nonlinear field [17]. Nevertheless, this higher-order nonlinearity is always much weaker than the third-order, and thus its effect on the harmonic generations can be neglected in the present paper.

Regarding the incremental susceptibility (i.e., last term of equation (3)), it is equivalent to $12\pi\Delta\chi(\langle\mathbf{H}\rangle)$. That is, we have

$$\Delta\chi(\langle\mathbf{H}\rangle) = \frac{\langle\mathbf{H}\rangle}{12\pi}\left(\frac{\partial\mu_e}{\partial\rho}\right)_{T,\langle\mathbf{H}\rangle}\left(\frac{\partial\rho}{\partial\langle\mathbf{H}\rangle}\right)_{T,p}. \tag{5}$$

The differential increase of the density inside the FA area $d\rho$ corresponds to the increase in mass equal to $V_c\,d\rho$. This increase in mass is equal to a decrease in mass outside the FA area given by $-\rho\,d(V - V_c) = -\rho\,dV$, so that $d\rho = -[\rho/V_c]\,dV$. Accordingly, equation (5) is rewritten as

$$\Delta\chi(\langle\mathbf{H}\rangle) = -\frac{\rho\langle\mathbf{H}\rangle}{12\pi V_c}\left(\frac{\partial\mu_e}{\partial\rho}\right)_{T,\langle\mathbf{H}\rangle}\left(\frac{\partial V}{\partial\langle\mathbf{H}\rangle}\right)_{T,p}. \tag{6}$$

Then we can obtain $(\partial V/\partial\langle\mathbf{H}\rangle)_{T,p}$ based on the differential of the free energy $\mathrm{d}F = -S\,\mathrm{d}T - p\,\mathrm{d}V + V_c/(4\pi)\langle\mathbf{H}\rangle\,\mathrm{d}\langle\mathbf{B}\rangle$, where S denotes the entropy. Using the transformed free enthalpy $\tilde{\Omega} = F + pV - V_c/(4\pi)\langle\mathbf{H}\rangle\langle\mathbf{B}\rangle$, its differential can be given by $\mathrm{d}\tilde{\Omega} = -S\,\mathrm{d}T + V\,\mathrm{d}p - V_c/(4\pi)\langle\mathbf{B}\rangle\,\mathrm{d}\langle\mathbf{H}\rangle$. From this equation, we find $(\partial V/\partial\langle\mathbf{H}\rangle)_{T,p} = -V_c\langle\mathbf{H}\rangle/(4\pi)(\partial\mu_\mathrm{e}/\partial p)_{T,\langle\mathbf{H}\rangle}$. Then, the substitution of this into equation (6) leads to

$$\Delta\chi(\langle\mathbf{H}\rangle) = \frac{\rho\langle\mathbf{H}\rangle^2}{48\pi^2}\left(\frac{\partial\mu_\mathrm{e}}{\partial\rho}\right)_{T,\langle\mathbf{H}\rangle}\left(\frac{\partial\mu_\mathrm{e}}{\partial p}\right)_{T,\langle\mathbf{H}\rangle}. \tag{7}$$

Next, we use $(\partial\mu_\mathrm{e}/\partial p)_{T,\langle\mathbf{H}\rangle} = \alpha\rho(\partial\mu_\mathrm{e}/\partial\rho)_T$, where $\alpha = -1/V(\partial V/\partial p)_T$ is the compressibility in the absence of the field. In this equation, terms depending on $\langle\mathbf{H}\rangle$ have been neglected since they lead to terms in powers of $\langle\mathbf{H}\rangle$ higher than the second (nonlinear field) in equation (7). So far equation (7) can be rewritten as

$$\Delta\chi(\langle\mathbf{H}\rangle) = \frac{\alpha\rho^2\langle\mathbf{H}\rangle^2}{48\pi^2}\left(\frac{\partial\mu_\mathrm{e}}{\partial\rho}\right)_T^2. \tag{8}$$

2.2. Harmonic generation

Now let us investigate the harmonic generation owing to the nonlinear term (see equations (1) and (8)). In view of equation (1), the orientational magnetization along z axis has the following general form:

$$\mathbf{M} = \frac{\mu_\mathrm{e} - \mu_2}{4\pi}\langle\mathbf{H}\rangle + \xi\langle\mathbf{H}\rangle^3, \tag{9}$$

where the third-order nonlinear coefficient ξ is defined as $\xi \equiv \Delta\chi(\langle\mathbf{H}\rangle)/\langle\mathbf{H}\rangle^2$. We use an inhomogeneous dc field like $\tilde{\mathbf{H}}_\mathrm{dc} = (l_z(z)/L_z)H_\mathrm{dc}\hat{\mathbf{z}}$ where $0 < l_z(z) \leqslant L_z$ with L_z being the length of the FA area (which is here assumed to be cubic) along the z axis. Without loss of generality, L_z is set to unity. Meanwhile, a sinusoidal ac magnetic field $\mathbf{H}_\mathrm{ac} = H_\mathrm{ac}(t)\hat{\mathbf{z}} = H_\mathrm{ac}\hat{\mathbf{z}}\sin(\omega t)$ is applied. Therefore, the magnetization M can be further expressed in terms of odd- and even-order harmonics such that

$$M = M_\mathrm{dc} + M_\omega\sin(\omega t) + M_{2\omega}\cos(2\omega t) + M_{3\omega}\sin(3\omega t), \tag{10}$$

where the dc component (M_dc), fundamental harmonics (M_ω), and second- and third-order harmonics ($M_{2\omega}$ and $M_{3\omega}$) are respectively given by

$$M_\mathrm{dc} = \frac{\mu_\mathrm{e} - \mu_2}{8\pi}H_\mathrm{dc} + \frac{3\xi}{4}H_\mathrm{dc}H_\mathrm{ac}^2 + \frac{\xi}{8}H_\mathrm{dc}^3, \tag{11}$$

$$M_\omega = \frac{\mu_\mathrm{e} - \mu_2}{4\pi}H_\mathrm{ac} + \frac{3\xi}{4}H_\mathrm{ac}H_\mathrm{dc}^2 + \frac{3\xi}{4}H_\mathrm{ac}^3, \tag{12}$$

$$M_{2\omega} = -\frac{3\xi}{4}H_\mathrm{dc}H_\mathrm{ac}^2, \tag{13}$$

$$M_{3\omega} = -\frac{\xi}{4}H_\mathrm{ac}^3. \tag{14}$$

In the above derivation, we have used two identities, $\sin^3(\omega t) = (3/4)\sin(\omega t) - (1/4)\sin(3\omega t)$ and $\sin^2(\omega t) = [1 - \cos(2\omega t)]/2$.

2.3. Numerical results

For a quantitative understanding, let us do some numerical simulations. Using the Onsager consideration, we set the Kirkwood correlation factor $g = 1$, which means that there are no more correlations between the particle orientations than can be accounted for with the aid of the

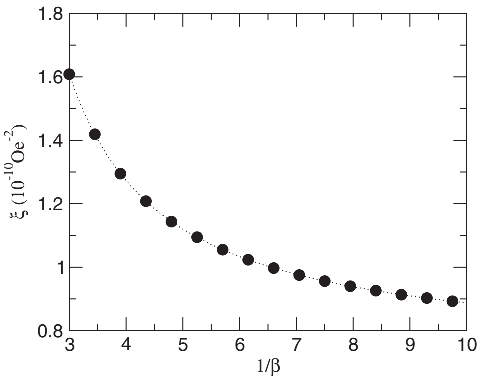

Figure 1. Nonlinear coefficient ξ against the degree of anisotropy $1/\beta$ (dimensionless). Parameters: volume fraction of the particles 0.2, particle radius 95 μm, $m = 10^{-9}$ emu, $T = 298$ K, $\alpha = 2.1 \times 10^{-10}$ Pa^{-1}, $\mu_2 = 1$, and $\mu_{e0} = 6$.

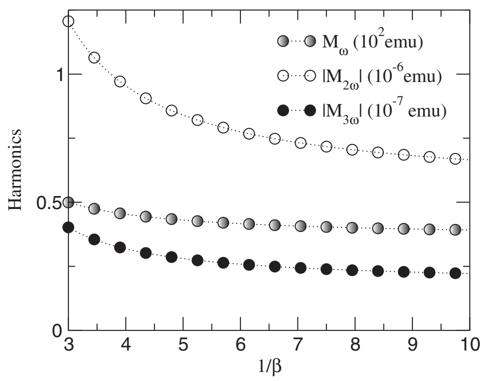

Figure 2. Fundamental and second- and third-order harmonics of the magnetization, M_ω, $M_{2\omega}$, and $M_{3\omega}$, against the degree of anisotropy $1/\beta$. Parameters are the same as those in figure 1. Other parameters: $H_{dc} = 100$ Oe and $H_{ac} = 10$ Oe.

continuum method. In figure 1 we show the nonlinear coefficient ξ as a function of the degree of anisotropy $1/\beta$. Figure 2 displays the fundamental and second- and third-order harmonics of the magnetization as a function of $1/\beta$. Here $1/\beta = 3$ represents an isotropic limit. As more particle chains are formed, the corresponding $1/\beta$ increases. This causes the nonlinear coefficient to decrease (see figure 1), thus yielding decreasing harmonics of the magnetization (see figure 2). In this sense, it becomes possible to monitor the structure of MR fluids by detecting such harmonics.

3. Discussion and conclusion

Here some comments are in order. In this work, we have presented a new mechanism for harmonic generation in MR fluids, which differs from the usual ones due to magnetic anisotropy existing in amorphous ferromagnetic materials. Based on thermodynamics, we derive the incremental magnetic susceptibility of the MR fluid which is partially situated in a nonuniform magnetic field, and further extract the harmonics analytically.

For the present system under consideration, the nonlinearity could be caused to appear by three effects [14], namely, normal saturation, anomalous saturation, and magnetostriction. In detail, the normal saturation arises from the higher terms of the Langevin function at large field intensities. As the strength of the field is large enough, this kind of normal saturation should be taken into account. In the present paper, since the field strength may be moderate, the normal saturation might be expected to be small enough to be neglected. Regarding the anomalous saturation, it results from the equilibrium between entities (i.e., particle chains in this work) with higher and lower dipole moments which is shifted under the influence of the field. In this work, the correlation between the dipole moments of the particle chains and in turn the anomalous saturation are neglected in the sense that the current anomalous saturation is much more weak. This is 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 higher dipole moment (of particle chains) and lower dipole moment (of particle chains) would not be able to predict significant nonlinearity. So, the focus of this work is on the effect of magnetostriction which arises from an inhomogeneous magnetic field.

From equation (8), we find that the obtained third-order nonlinear coefficient ξ is proportional to the compressibility in the absence of the field. To some extent, it can be concluded that ξ is actually related to the pressure in the absence of the field, which is a well defined quantity. However, the pressure is not a well defined quantity in the presence of fields. To study the nonlinearity higher than the third order, the pressure in the presence of the field must be further taken into account, based on the present formalism. In this work, we have neglected the higher-order nonlinearity since it is always much weaker than the third order. If one does need to investigate the higher-order nonlinearity, one had better work with the chemical potential instead in the sense that it remains a well defined quantity.

It is also of interest to extend the present theory to other colloidal suspensions like ferrofluids [1], electrorheological fluids [2], or charged colloids [3]. In addition, one can also discuss a system containing graded particles, in an attempt to take into account the gradation effect [19].

To sum up, based on thermodynamics we have theoretically exploited a new mechanism for harmonic generation in MR fluids. Such harmonics are expected to play a role in monitoring the structure of MR fluids.

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