Nonequilibrium Brownian dynamics analysis of negative viscosity induced in a magnetic fluid subjected to both ac magnetic and shear flow fields

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We study the rheological and magnetic characteristics of a magnetic fluid. The system, which we investigate, is as follows. Ferromagnetic particles are dispersed in a solvent, which is subjected to both ac magnetic and shear flow fields. The translational and rotational motions of particles are calculated by the Brownian dynamics method based on Langevin equations and the rheological and magnetic characteristics of the magnetic fluid system are estimated. First, we investigate the rheological and magnetic characteristics of the system in a dc magnetic field and then we analyze the effect of an ac magnetic field on those characteristics. We find that the negative viscosity effect is induced at a certain frequency range of the ac magnetic field. We also find that there are two main mechanisms responsible for the occurrence of negative viscosity. (1) Resonance between the rotational motions of the dipoles of particles and the fluctuation of ac magnetic fields occurs when applied magnetic fields are weak compared to the shear rate, in which case particles can still rotate in magnetic fields. Beyond this resonance frequency, negative viscosity appears. (2) The magnetic dipole moments of particles are forced to stay in the direction of the magnetic field when strong magnetic fields are applied in relatively low shear flow fields. However, negative viscosity occurs when the frequency of external magnetic fields exceeds a critical value, in which case the dipoles rotate continuously in a shear flow without stopping. In both cases, the mean angular velocity of the particles becomes higher than that of the solvent.

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I. INTRODUCTION

Solutions, in which ferromagnetic colloidal particles are dispersed, are called magnetic fluids [1,2]. The diameter of each particle is of the order of 10 nm and each particle is composed of a single magnetic domain, the magnitude of the magnetic dipole moment of which is constant. The direction of the dipole moment changes with the rotational motion of the particle. Since the motion of each particle is affected by interactions with the molecules of the solvent and other ferromagnetic particles, macroscopic flow in the solvent, and external magnetic fields, magnetic fluids show various interesting rheological and magnetic characteristics under different conditions. The increase in the apparent viscosity of magnetic fluids in dc magnetic fields has been observed in several experiments [3,4], which is, in fact, caused by the reduction or suppression of the rotational motions of ferromagnetic particles by the fields. Shliomis [5] carried out theoretical analysis of this viscosity increase based on ferrohydrodynamic equations, which consist of the equations of magnetic fluid motions, the Maxwell equations, and the relaxation equation for the system magnetization. The viscosity increase is also analyzed on the basis of modified magnetization equations, which are derived from the Fokker-Planck equation [6] and irreversible thermodynamics [7]. Morimoto and Maekawa [8] investigated the rheological property from a microscopic point of view by a nonequilibrium Brownian dynamics method.

The effect of ac magnetic fields on the physical properties of magnetic fluids has also been studied recently [9]. Whereas dc magnetic fields reduce or suppress the rotational motions of ferromagnetic particles as we mentioned above, ac magnetic fields may encourage the rotational motions under certain conditions. The mean angular momentum of ferromagnetic particles in the solvent is zero without any macroscopic flow even in ac magnetic fields, but imposing a macroscopic velocity gradient can give it a biased value. Shliomis and Morozov [10] predicted that the apparent viscosity of magnetic fluids, which are subjected to both an ac magnetic field and velocity gradient, may become negative. Bacri *et al.* [11] and Zeuner, Richter, and Rehberg [12] actually observed the negative viscosity effect in the Poiseuille flow of a magnetic fluid. Gazeau *et al.* [13] investigated the magnetic characteristics of a magnetic fluid, which was rotating in an ac magnetic field, and discovered the so-called vorticomagnetic resonance in the magnetic susceptibility. Furthermore, Gazeau *et al.* [14] observed the vorticomagnetic resonance and the negative viscosity effect at the same time using the same experimental system. The negative viscosity effect and the vorticomagnetic resonance have been analyzed theoretically on the basis of the ferrohydrodynamic equations [10–16]. The above phenomena are induced by the motions of ferromagnetic particles in the solvent, which is subjected to both an external ac magnetic field and velocity gradient. However, in the negative viscosity and vorticomagnetic resonance regions, the particle motions in magnetic fluids have not yet been calculated directly from a microscopic point of view and the relation between the motions and the physical properties such as the apparent viscosity and magnetization have also not yet been studied.

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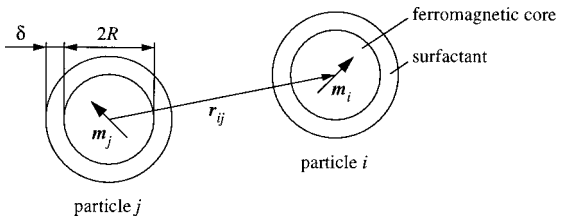

FIG. 1. Ferromagnetic particles. Each particle is composed of a single magnetic domain and covered with surfactant molecules.

In this paper, we investigate the rheological and magnetic characteristics of a magnetic fluid, which is subjected to both ac magnetic and shear flow fields, by a nonequilibrium Brownian dynamics method. We analyze the effect of the amplitude and frequency of an external magnetic field and the shear rate on the rheological and magnetic features. In the Sec. II, we show our numerical modeling and procedure and introduce the interparticle potentials, the equations of motions, the control parameters, and the calculation conditions. We investigate the rheological and magnetic features of the magnetic fluid in both dc magnetic and shear flow fields. In the Sec. III, we investigate the rheological and magnetic characteristics of the magnetic fluid, which is subjected to both ac magnetic and shear flow fields, and discuss the mechanism, which causes the negative viscosity effect, in detail. In Sec. IV, we summarize the result we obtained in this study.

II. NUMERICAL MODELING OF MAGNETIC FLUIDS AND RHEOLOGICAL AND MAGNETIC CHARACTERISTICS IN A dc MAGNETIC FIELD

Magnetic fluids consist of ferromagnetic particles dispersed in a solvent. We assume that each ferromagnetic particle is spherical, composed of a single magnetic domain and covered with surfactant molecules as shown in Fig. 1. The interparticle potentials are dipole potential u_m [17], the van der Waals attraction u_v, and the repulsion caused by surfactant-surfactant contact u_s [2].

$$u_m=\frac{1}{4\pi\mu_0}\left\{\frac{\boldsymbol{m}_i\cdot\boldsymbol{m}_j}{r_{ij}^3}-\frac{3}{r_{ij}^5}(\boldsymbol{m}_i\cdot\boldsymbol{r}_{ij})(\boldsymbol{m}_j\cdot\boldsymbol{r}_{ij})\right\}, \tag{1}$$

$$u_v=-\frac{A}{6}\left\{\frac{2R^2}{r_{ij}^2-4R^2}+\frac{2R^2}{r_{ij}^2}+\ln\left(\frac{r_{ij}^2-4R^2}{r_{ij}^2}\right)\right\}, \tag{2}$$

$$u_s=2\pi R^2NkT\left\{2-\frac{r_{ij}-2R}{\delta}-\frac{r_{ij}}{\delta}\ln\left(\frac{2R+2\delta}{r_{ij}}\right)\right\}, \tag{3}$$

where μ_0, $\boldsymbol{m}_i$, A, R, N, k, T, and δ are, respectively, the permeability, the magnetic dipole moment of particle i, the Hamaker constant, the radius of the particle, the surface density of the surfactant molecules, the Boltzmann constant, the temperature, and the thickness of the surfactant layer. The interactive energy between a particle and an external magnetic field is expressed as

$$u_H=-\boldsymbol{m}_i\cdot\boldsymbol{H}, \tag{4}$$

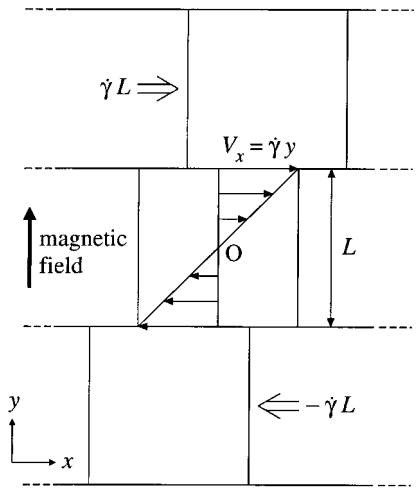

FIG. 2. Schematic representation of the physical system. Ferromagnetic particles are dispersed in the system and dc or ac magnetic fields are applied in the y direction. A shear flow field, in which a linear velocity distribution is established, is imposed in the x direction. Moving boundary conditions developed by Lees and Edwards are employed. The top and bottom cells, in which linear velocity distributions are also established, are moved at a speed of $\dot\gamma L$ and $-\dot\gamma L$, respectively.

where $\boldsymbol{H}$ is the external magnetic field.

The physical system is shown in Fig. 2. The system is two-dimensional and a uniform external magnetic field is applied in the y direction. Particles are subjected to a shear flow field, in which a linear velocity distribution is established. The translational and rotational motions of ferromagnetic particles are expressed by the following Langevin equations:

$$\frac{\partial\boldsymbol{u}}{\partial t}=-\frac{\zeta_t}{m}(\boldsymbol{u}-\boldsymbol{V})+\frac{\boldsymbol{F}}{m}+\boldsymbol{P}, \tag{5}$$

$$\frac{\partial\boldsymbol{\omega}}{\partial t}=-\frac{\zeta_r}{I}(\boldsymbol{\omega}-\boldsymbol{\Omega})+\frac{\boldsymbol{T}}{I}+\boldsymbol{\tau}, \tag{6}$$

where $\boldsymbol{u}$, $\boldsymbol{\omega}$, m, and I are, respectively, the velocity, the angular velocity, the mass, and the moment of inertia. $\boldsymbol{F}$ and $\boldsymbol{T}$ are the force and torque acting on a particle through interactions with the other particles and the external magnetic field. ζ_t and ζ_r are the friction coefficients. We assume that Stokes law applies to the coefficients. $\boldsymbol{P}$ and $\boldsymbol{\tau}$ are the random force and torque caused by collisions with solvent molecules. The mean values and autocorrelations of the random force and torque satisfy the following relations:

$$\langle\boldsymbol{P}(t)\rangle=\langle\boldsymbol{\tau}(t)\rangle=0, \tag{7}$$

$$\langle\boldsymbol{P}(t)\cdot\boldsymbol{P}(t+\Delta t)\rangle=\frac{6\zeta_tkT}{m^2}\delta(\Delta t), \tag{8}$$

TABLE I. Calculation conditions and physical properties.

Diameter of ferromagnetic core	10 (nm)
Thickness of surfactant	2 (nm)
Mass of particle	2.9×10^{-21} (kg)
Surface density of surfactant molecules	10^{18} (molecules/m^2)
Dynamic viscosity of solvent	10^{-3} (Pa s)
Hamaker constant	10^{-19} (J)
Temperature of solvent	300 (K)
Number density of particles	121 (particles)/0.352^2 (μm^2)
	900 (particles)/0.960^2 (μm^2)

$$\langle \boldsymbol{\tau}(t)\cdot\boldsymbol{\tau}(t+\Delta t)\rangle = \frac{6\zeta_r kT}{I^2}\delta(\Delta t), \tag{9}$$

where $\langle\cdots\rangle$ represents the time average and δ is the Dirac δ function. $\boldsymbol{V}$ and $\boldsymbol{\Omega}$ are the external velocity and angular velocity,

$$\boldsymbol{V}=\begin{pmatrix}\dot\gamma y\\0\\0\end{pmatrix},\quad \boldsymbol{\Omega}=\frac{1}{2}\mathrm{rot}\boldsymbol{V}=\begin{pmatrix}0\\0\\-\dot\gamma/2\end{pmatrix}, \tag{10}$$

where $\dot\gamma$ is the shear rate. Moving boundary conditions, which were developed by Lees and Edwards [18], are employed and the Langevin equations are integrated based on Ermak's procedure [19,20]. After the system has reached the steady state in a dc magnetic field, a shear flow field is imposed. When the system is subjected to an ac magnetic field, a shear flow field and a fluctuating magnetic field are applied after the system has reached the steady state in a dc magnetic field. The stress tensor, σ_{yx}, and the apparent viscosity, $\Delta\eta$, are expressed as follows:

$$\sigma_{yx}=-\frac{1}{V}\sum_i\left(y_iF_{x,i}-\frac{T_{z,i}}{2}\right), \tag{11}$$

$$\Delta\eta=\frac{\sigma_{yx}}{\dot\gamma}=\Delta\eta_t+\Delta\eta_r, \tag{12}$$

where V is the volume of the system; that is, the area of the system multiplied by the diameter of a particle including the surfactant layer; $2(R+\delta)$, and i represents the ith particle. The first and second terms on the right-hand side of Eq. (11) are symmetric and asymmetric components, respectively. $\Delta\eta_t$ and $\Delta\eta_r$, which we call the translational and rotational viscosities, are viscosities induced by the symmetric and asymmetric components of the stress tensor.

The control parameters are the ratio of the magnetic dipole moment energy to thermal energy, λ, the ratio of the interactive energy between the dipole moment and the external magnetic field to thermal energy, ξ, and the Péclet number, Pe, which is the nondimensional shear rate,

$$\lambda\equiv\frac{|\boldsymbol{m}|^2}{4\pi\mu_0 d^3kT}, \tag{13}$$

$$\xi\equiv\frac{|\boldsymbol{m}|H}{kT}, \tag{14}$$

$$\mathrm{Pe}\equiv\frac{3\pi\eta_0(d+2\delta)^3\dot\gamma}{8kT}, \tag{15}$$

where $\boldsymbol{m}$, d, H, and η_0 are, respectively, the magnetic dipole moment of a particle, the diameter of a particle, the intensity of the magnetic field, and the viscosity of the solvent.

We carried out Brownian dynamics simulations for investigating the rheological and magnetic characteristics of the magnetic fluid system, using 121 and 900 particles. The maximum difference in the apparent viscosities caused by the particle number difference was within 6%. Therefore, the result, which we show in the following, is based on the simulations, in which 121 particles are used. The calculation conditions and physical properties are summarized in Table I. The value of the control parameter, λ, is set at 1.5, which corresponds to a usual magnetic fluid case, in which for example, Fe_3O_4 particles of 10 nm are dispersed. The ratio of the area of the particles including the surfactant layers to the area of the system is 0.15. If the system is considered to be a rectangular parallelepiped, the depth of which is the diameter of the particle, the volume fraction is 0.10. Note that we calculated the stress tensor and the apparent viscosity based on the volume of the rectangular parallelepiped, as explained above.

It is known that ferromagnetic particles form clusters in the solvent under certain conditions and, in particular, straight chain clusters are formed along an external magnetic field [21]. The cluster formation process is investigated in detail by the Brownian dynamics method [20,22]. Before starting the rheological and magnetic analysis, we checked the cluster formations in the present system. A dc magnetic field was imposed in the y direction and ξ was changed from 0 to 30 in the absence of a shear flow. The result shows that the average number of particles included in a cluster is less than 2 even under $\xi=30$. What is more, the lifetime of each cluster is very short, less than 0.05 ms. Therefore, in the present case ($\lambda=1.5$), the chain cluster formation is not significant.

We also checked the rheological and magnetic characteristics of the magnetic fluid system in a dc magnetic field by imposing an external shear flow before analyzing those characteristics in an ac magnetic field. The dependence of the

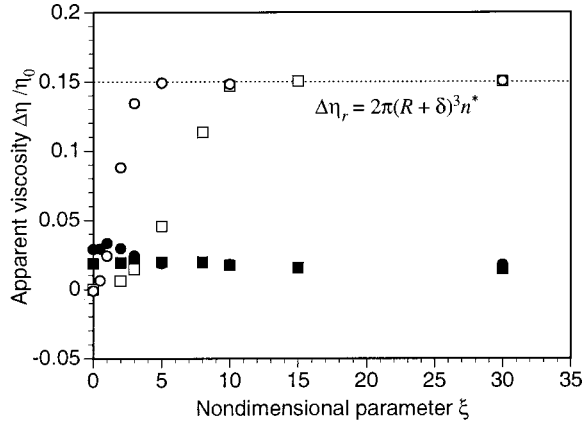

FIG. 3. Dependence of the apparent viscosities on control parameter ξ in a dc magnetic field, Pe=1 and 5. The rotational viscosity, $\Delta\eta_r$ (○, Pe=1; □, Pe=5), increases with the intensity of the magnetic field and reaches a constant value, $2\pi(R+\delta)^3 n^*$, where R is the radius of a particle, δ the thickness of the surfactant layer, and n^* the number density of particles. The translational viscosity, $\Delta\eta_t$ (●, Pe=1; ■, Pe=5), is low.

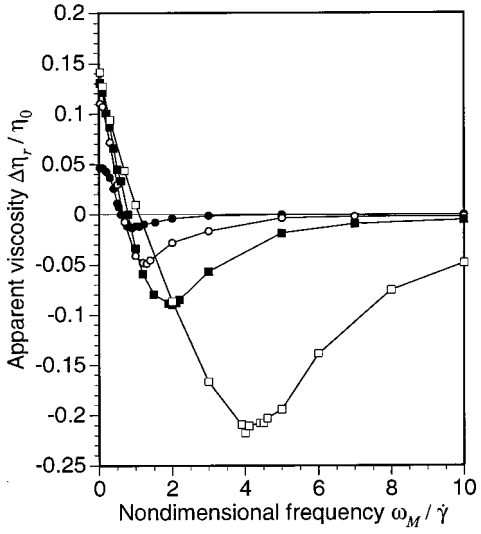

FIG. 5. Dependence of the apparent viscosity on the amplitude and frequency of an ac magnetic field, Pe=1. ●, $\xi=2$; ○, $\xi=5$; ■, $\xi=10$; and □, $\xi=30$.

apparent viscosities, $\Delta\eta_r$ and $\Delta\eta_t$, on the control parameter, ξ, is shown in Fig. 3 for Pe=1 and 5. The rotational viscosity, $\Delta\eta_r$, increases with the intensity of the dc magnetic field and finally reaches a constant value; $\Delta\eta_r=2\pi(R+\delta)^3 n^*$, where n^* is the number density of particles (see Ref. [8] for the derivation of the equation), since the rotational motions of particles are stopped by the magnetic field. The critical intensity of dc magnetic fields for the suppression of the rotational motions of particles increases with an increase in the Péclet number, that is, the shear rate. The dependence of the apparent viscosity on the external dc magnetic field in our magnetic fluid system coincides with the experimental result [3] and the theoretical predictions [5–7]. The magnetic characteristics of the magnetic fluid in the dc magnetic field are shown in Fig. 4 where the magnetization is normalized by the saturation magnetization of the system. The system is magnetized in the shear flow direction (x direction) as well as in the magnetic field direction (y direction) when it is subjected to a shear flow field. As is expected, the magnetization in the magnetic field direction, M_y, increases with the intensity of dc magnetic fields and finally becomes saturated. M_y is reduced in shear flow fields since particles rotate and the dipole moments have a component in the shear flow direction. The magnetization in the x direction, M_x, is zero in the absence of a shear flow and when the system is subjected to a shear flow field, M_x increases with the intensity of external dc magnetic fields as long as magnetic fields are not very strong, since the particles can rotate in such weak magnetic fields and the magnetic dipoles stay longer in the x direction than in the $-x$ direction. The magnetization in the shear flow direction becomes maximum near the point where the rotational motions of the particles are stopped by the magnetic field and beyond this point, it decreases monotonically since the dipoles tend to line up in the magnetic field direction.

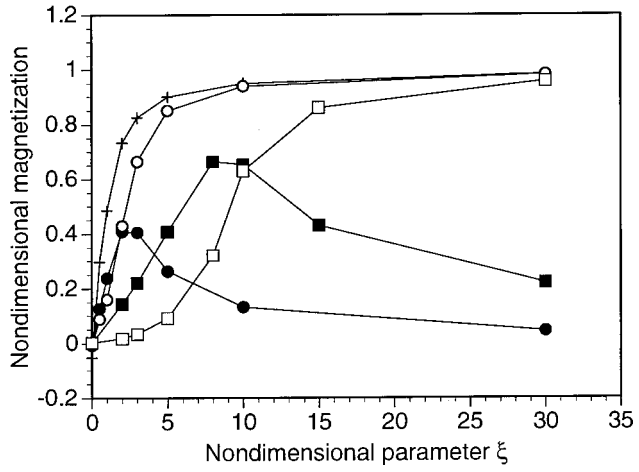

FIG. 4. Dependence of the magnetization of the system on control parameter ξ, Pe=0, 1 and 5. The system magnetization is normalized by the saturation magnetization. The magnetization in the magnetic field direction, M_y (+, Pe=0; ○, Pe=1; □, Pe=5), increases with the intensity of the magnetic field and saturates. The magnetization in the shear flow direction, M_x (●, Pe=1; ■, Pe=5), becomes maximum near the frequency at which the particles' rotational motions are stopped.

III. RHEOLOGICAL AND MAGNETIC CHARACTERISTICS IN AN ac MAGNETIC FIELD

We carried out Brownian dynamics simulations for investigating the rheological and magnetic characteristics of the magnetic fluid system in an ac magnetic field. The magnetic field is changed with time as follows: $H_y=H\cos(\omega_M t)$, where ω_M is the angular frequency of the external ac magnetic field. The order of the apparent viscosity caused by the translational motion, $\Delta\eta_t$, was the same as that in the dc magnetic field (see Fig. 3). Therefore, the rotational viscosity, $\Delta\eta_r$, mainly accounts for the rheological characteristics of magnetic fluids.

The effect of the amplitude and frequency of an ac mag-

netic field on the apparent rotational viscosity, $\Delta\eta_r$, is shown in Fig. 5 for Pe$=1$ where the viscosity, $\Delta\eta_r$, and the frequency of the magnetic field, ω_M, are normalized by the solvent viscosity, η_0, and the shear rate, $\dot{\gamma}$. The viscosity is high when the frequency of magnetic fields is low and it decreases as the frequency increases. The viscosity becomes negative at an intermediate frequency range and when the frequency is very high, there is no effect of the magnetic field on the apparent viscosity. The minimum value in negative viscosity decreases and the frequency corresponding to the minimum viscosity increases as the amplitude of the ac magnetic field, ξ, increases. The frequency range, under which negative viscosity appears, becomes wider with an increase in ξ.

Here, we compare the results, which have been obtained in the present analysis, with those obtained by the previous theoretical and experimental studies. According to the analysis of Shliomis and Morozov [10], the minimum value of the apparent viscosity decreases as the amplitude of the fluctuating magnetic field increases, which agrees with the present result, but the frequency of the magnetic field, at which the apparent viscosity becomes zero, decreases with an increase in the amplitude of the magnetic field, which disagrees with both the present result (see Fig. 5) and the experimental result obtained by Bacri *et al.* [11]. Analytical models, in which magnetization relaxation equations, which are derived from the Fokker-Planck equation, are used, have been introduced by Bacri *et al.* [11] and Feldherhof [15]. Müller and Liu [16] developed a different model, in which a generalized magnetization equation is used. The results obtained by the above models agree with the present result and the experimental result obtained by Bacri *et al.* [11]; that is, the minimum value of the apparent viscosity decreases and the frequency of the magnetic field corresponding to the zero viscosity increases with the amplitude of the magnetic field. Note that when the demagnetization effect is taken into account in the Felderhof's model, the frequency, which gives zero viscosity, slightly decreases with an increase in the amplitude of the magnetic field. Zeuner, Richter, and Rehberg [12] carried out an experimental investigation of the negative viscosity effect. Their experimental results, as a whole, coincide with the results obtained by the present analysis, those models and the experiment of Bacri *et al.*, except that the negative viscosity effect is not induced any longer when the intensity of the magnetic field exceeds a certain value. The discrepancy might have been caused by the differences in the interparticle forces, which, in fact, Zeuner, Richter, and Rehberg themselves also mentioned in their paper. The value of the control parameter λ, which is the ratio of the magnetic dipole moment energy to thermal energy, was approximately 1.1 in the case of the experiment of Bacri *et al.*, whereas it was 5.1 in the case of the experiment of Zeuner, Richter, and Rehberg, judging by the particle size distributions which they measured. Therefore, the interaction between particles is quite strong in the case of the experiment of Zeuner, Richter, and Rehberg. Since the interparticle forces were not taken into account in the models introduced by Bacri *et al.* [11] or Felderhof [15], the physical conditions in the models are quite similar to those in the experiment of Bacri *et al.*

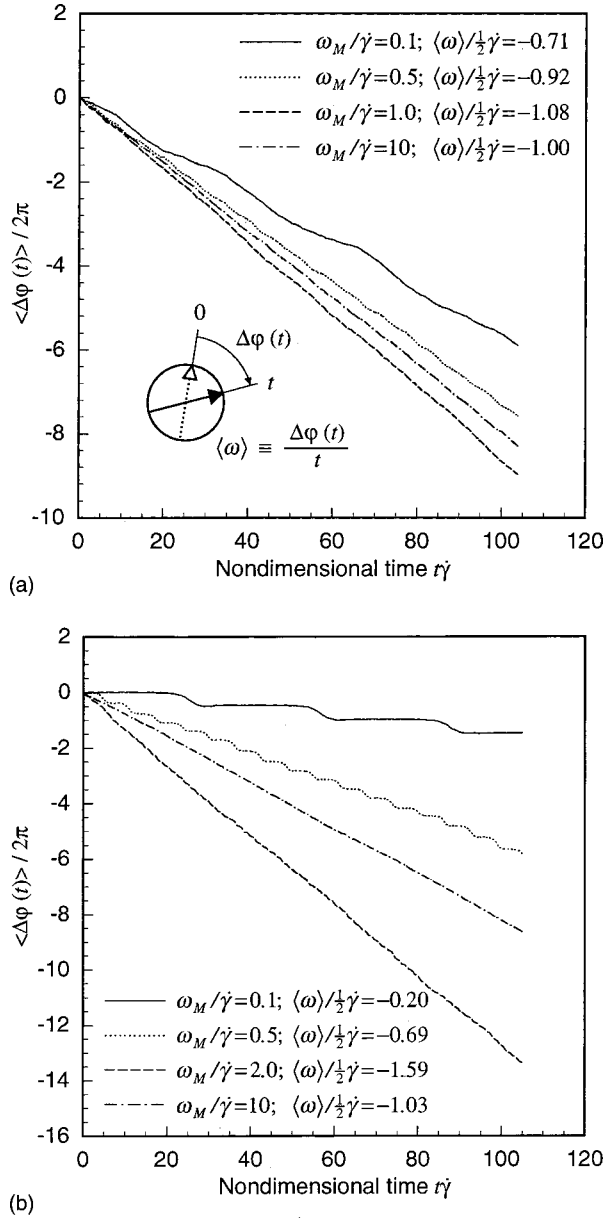

FIG. 6. Time variations of the mean angular displacement of ferromagnetic particles in both ac magnetic and shear flow fields, Pe$=1$. (a) $\xi=2$, (b) $\xi=10$. The slope gives the mean angular velocity. The angular velocity of particles is higher than that of the solvent in the negative viscosity regions.

The analytical conditions of Müller and Liu [16] are based on those of the experiment of Bacri *et al.* The value of λ in our analysis is 1.5, which is close to that in the experiment of Bacri *et al.* Therefore, the interparticle forces are quite weak in the cases of Refs. [11,15,16] and the present analysis, compared to the experiment of Zeuner, Richter, and Rehberg [12]. We need to investigate the effect of the control parameter λ on the rheological and magnetic characteristics in more detail.

In the negative viscosity regions, the mean angular velocity of the particles should be higher than the angular velocity of the solvent. Therefore, we checked the time variations of the mean angular displacement of the particles. The time

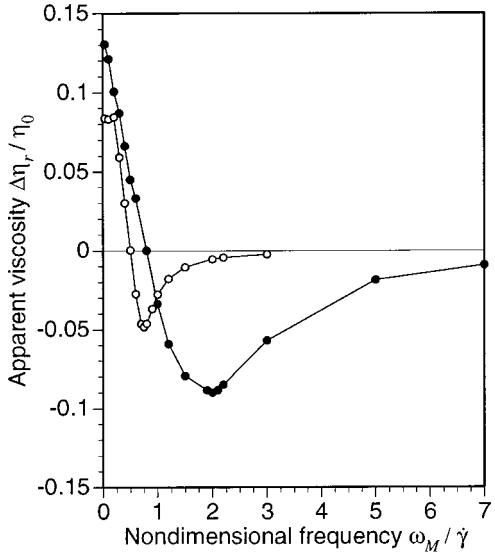

FIG. 7. Dependence of the apparent viscosity on the frequency of an ac magnetic field and the Péclet number. $\xi=10$: ●, Pe=1; ○, Pe=5.

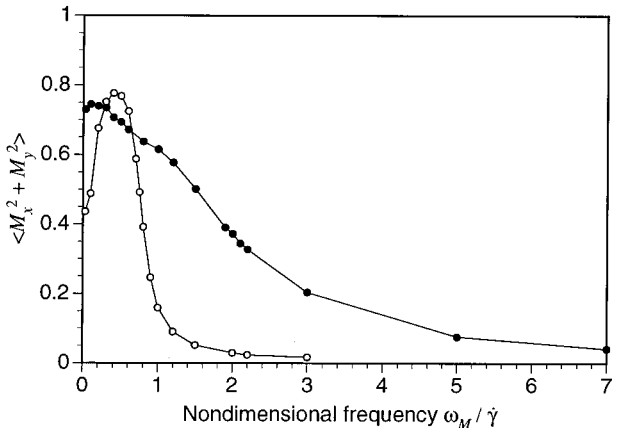

FIG. 8. Dependence of the magnetization of the system on the frequency of an ac magnetic field and the Péclet number. $\xi=10$: ●, Pe=1; ○, Pe=5.

variations of the mean angular displacement, $\langle \Delta\varphi(t) \rangle$, are shown in Fig. 6 for $\xi=2$ and 10, where the Péclet number is fixed at 1. The mean angular velocity, $\langle \omega \rangle$, is calculated by $\langle \Delta\varphi(t) \rangle / t$. Note that the slope in Fig. 6 gives the mean angular velocity. In the case of $\xi=2$ [Fig. 6(a)], the angular velocity of the particles increases as the magnetic field frequency, ω_M, increases and when $\omega_M / \dot{\gamma}=1.0$, the mean particle angular velocity is higher than the solvent angular velocity; $|\langle \omega \rangle| > \dot{\gamma}/2$. The mechanism, which causes this negative viscosity effect in rather weak magnetic fields, will be discussed later. When $\omega_M / \dot{\gamma}=10$, the particles' rotational motions do not follow the magnetic field fluctuation and therefore the angular velocity of the particles is almost the same as that of the solvent. When the amplitude of the magnetic field is as large as $\xi=10$ [Fig. 6(b)], on the other hand, the mode of the rotational motions of particles becomes different depending on the frequency of the magnetic field, ω_M. When ω_M is low ($\omega_M / \dot{\gamma}=0.1,0.5$), the particles' motions become cyclic; that is, the particles' rotational motions stop for some period and then rotate quickly. The external magnetic field is strong enough to stop the rotational motions of the particles even in the shear flow field (see Fig. 3). However, the external magnetic field is fluctuating from the y direction to $-y$ direction and vice versa, and therefore, once the intensity of the magnetic field in the y direction becomes low, the particles start rotating quickly due to the shear applied to the system, but as the intensity of the magnetic field in the $-y$ direction becomes higher in the meantime, the particles' rotational motions stop again. When $\omega_M / \dot{\gamma}=2.0$, the mode is changed. The rotational motions become smooth and the mean angular velocity becomes higher than that of the solvent. The mechanism responsible for the negative viscosity effect in strong magnetic fields will also be discussed later. When $\omega_M / \dot{\gamma}=10$, the particles' rotational motions cannot follow the magnetic field fluctuation and therefore the effect of the ac magnetic field on the apparent viscosity disappears.

As we have shown in the above, the rheological characteristics of magnetic fluids are influenced by the frequency, ω_M, as well as the amplitude, ξ, of ac magnetic fields. In this section, to understand the basic mechanism that causes the negative viscosity effect more clearly, we investigate the interactions between the rotational motions of the particles or dipoles and the magnetic field fluctuations, changing the Péclet number, 1 and 5 under a fixed magnetic field, $\xi=10$. Note that in the case of Pe=1 in a dc magnetic field, the rotational motions of the particles are stopped and the viscosity reaches the constant value for $5<\xi$, whereas in the case of Pe=5, the rotational motions are not stopped for $\xi<10$ (see Fig. 3). The dependence of the apparent viscosity on the frequency of the ac magnetic field and the Péclet number is shown in Fig. 7 where ξ is fixed at 10 and Pe=1 and 5. In the case of Pe=5, the frequency of the magnetic field corresponding to the minimum viscosity is lower and the frequency range, which gives the negative viscosity effect, is narrower, compared to the case of Pe=1. The relation between the amplitude of the magnetization fluctuation in the magnetic fluid system, $\langle M_x^2 + M_y^2 \rangle$, and the frequency of the ac magnetic field is shown in Fig. 8 for $\xi=10$, Pe=1 and 5, where the amplitude of the magnetization fluctua-

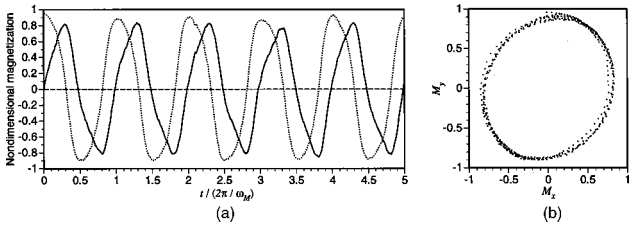

FIG. 9. Time variations of the magnetization components of the system and the phase diagram. $\xi=10$, Pe=5, and $\omega_M / \dot{\gamma}=0.5$. (a) Magnetization components (——, M_x; − − −, M_y). (b) Phase diagram. Resonance occurs between the magnetization and the external magnetic field fluctuation. Beyond this resonance frequency, the apparent viscosity becomes negative.

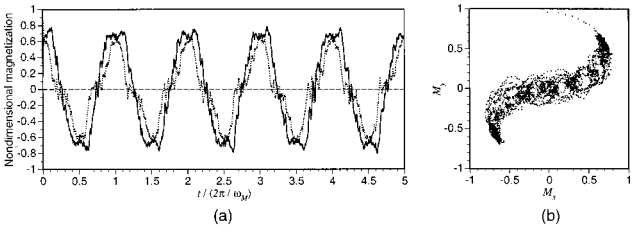

FIG. 10. Time variations of the magnetization components of the system and the phase diagram. $\xi=10$, Pe$=5$, and $\omega_M/\dot{\gamma}=0.03$. (a) Magnetization components (——, M_x; − − −, M_y). (b) Phase diagram. The angular frequency of the particles is higher than that of the external magnetic field, but lower than that of the solvent.

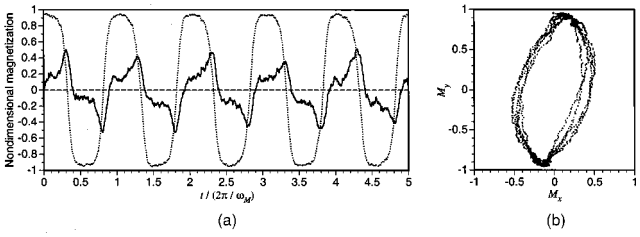

FIG. 11. Time variations of the magnetization components of the system and the phase diagram. $\xi=10$, Pe$=1$, and $\omega_M/\dot{\gamma}=0.5$. (a) Magnetization components (——, M_x; − − −, M_y). (b) Phase diagram. The dipoles are pointing in the magnetic field direction for some period due to the strong magnetic field and low frequency. As a result, the apparent viscosity increases.

tion is normalized by the saturation magnetization. In the case of Pe$=5$, the amplitude of the magnetization fluctuation becomes maximum at the magnetic field frequency, which corresponds to the angular velocity of the solvent; $\omega_M=\dot{\gamma}/2$, in which case the apparent viscosity is zero (see Fig. 7). This magnetization resonance is induced by the synchronization between the magnetic field fluctuation and the rotational motions of the dipoles of the particles. To demonstrate the synchronization, we show the time variations of the magnetization components at $\omega_M=\dot{\gamma}/2$ in Fig. 9(a) and the corresponding phase diagram in Fig. 9(b). In such a case as the shear rate is, relatively speaking, higher than the amplitude of magnetic fields, the negative viscosity effect is induced beyond the resonance frequency, at which the synchronization between the magnetic field fluctuation and the rotational motions of the dipoles of the particles occurs. As a result, the frequency range of magnetic fields, under which negative viscosity appears, is limited near the resonance frequency (Fig. 7) and therefore the peak in the amplitude of the magnetization becomes sharp (Fig. 8). Gazeau *et al.* observed the vorticomagnetic resonance in a system where magnetic fluid is confined in a rotating circular cylinder which is subjected to an ac magnetic field [13] and investigated the relation between the negative viscosity effect and the resonance [14]. They found that the resonance in the system magnetization occurs when the angular velocity of the fluid becomes the same as the frequency of the magnetic field and the apparent viscosity becomes negative when the frequency of the magnetic field exceeds the resonance frequency, which agrees very well with the present result obtained in the case of a rather weak magnetic field compared to the shear rate (see Figs. 7 and 8). The time variations of the magnetization components and the phase diagram when the frequency is as low as $\omega_M/\dot{\gamma}=0.03$ is shown in Figs. 10(a) and 10(b), in which case the apparent viscosity is positive. When the frequency of ac magnetic fields is lower than the resonance frequency in general, the angular velocity of particles does not coincide with the angular frequency of the magnetization fluctuation of the system. When $\omega_M/\dot{\gamma}=0.03$, the fundamental frequency of the magnetization fluctuation is the same as that of the external ac magnetic field, but the angular frequency of the rotating particles is higher than that of the magnetization fluctuation. Although the angular velocity of the particles is higher than that of the fluctuating magnetic field, it is still lower than the angular velocity of the solvent due to the magnetic field, which causes the increase in the apparent viscosity. In the case of Pe$=1$, on the other hand, the negative viscosity effect is more significant than that in the case of Pe$=5$; that is, the minimum value of the viscosity is lower and the frequency range, under which negative viscosity occurs, is wider (Fig. 7). The amplitude of the magnetization of the system decreases monotonically with an increase in the magnetic field frequency and the rate of decrease is low, in contrast to the magnetization feature for Pe$=5$, in which case the amplitude becomes maximum at the resonance frequency and decreases sharply beyond the resonance frequency. In such a case as when the magnetic fluid system is subjected to a strong magnetic field and relatively low shear rate ($\xi=10$,Pe$=1$), the dipoles are orientated in the direction of the external magnetic field (see Figs. 3 and 4). In the present case, as the magnetic field fluctuates with time, the orientation of the dipoles occurs while the intensity of the magnetic field is high. In order to check this dipoles' orientation in the system, we calculated the time variations of the magnetization components and the phase diagrams, which are shown in Fig. 11 for $\omega_M/\dot{\gamma}=0.5$ and in Fig. 12 for $\omega_M/\dot{\gamma}=2$. When the frequency of the external ac magnetic field is low ($\omega_M/\dot{\gamma}=0.5$), the magnetization in the magnetic field direction, M_y, becomes high and constant for some period (Fig. 11). During this moment, the dipoles are orientated in the magnetic field direction and therefore the rotational motions of the particles are stopped. As a result, the

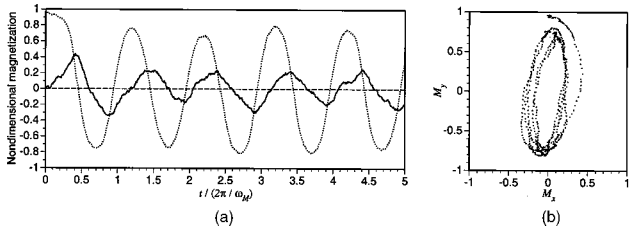

FIG. 12. Time variations of the magnetization components of the system and the phase diagram. $\xi=10$, Pe$=1$, and $\omega_M/\dot{\gamma}=2.0$. (a) Magnetization components (——, M_x; − − −, M_y). (b) Phase diagram. The direction of the dipoles changes smoothly following the magnetic field fluctuation due to the strong magnetic field and rather high frequency. The viscosity becomes negative because of the high frequency.

apparent viscosity becomes positive and high. When $\omega_M/\dot{\gamma}=2$, on the other hand, the directions of the dipoles are forced to change continuously without stopping by the external magnetic field (Fig. 12) and since the angular velocity of the dipoles or particles is higher than that of the solvent, the negative viscosity effect appears. Thanks to this mechanism, the particles can still rotate, following the magnetic field fluctuation, at higher frequencies compared to the resonance case. However, since the dipoles cannot follow the magnetic field fluctuation when the frequency of the magnetic field is very high, the system magnetization decreases gradually and the negative viscosity effect is reduced with an increase in the magnetic field frequency (see Figs. 7 and 8).

In summary, the negative viscosity effect is induced by two mechanisms:

(1) Resonance between the magnetic field fluctuation and the rotational motions of dipoles is induced in a magnetic field, which is, relatively speaking, weaker than a shear rate. Beyond the resonance frequency, negative viscosity appears.

(2) Forced fluctuations of dipoles are induced by strong ac magnetic fields under comparatively low shear rates. Generally speaking, the above two mechanisms are coupled depending on the combinations of the amplitude of ac magnetic fields, ξ, and the Péclet number, Pe.

IV. CONCLUSIONS

We investigated the rheological and magnetic characteristics of a magnetic fluid, which is subjected to both ac magnetic and shear flow fields, by a nonequilibrium Brownian dynamics method and obtained the following results.

(1) The viscosity is high when the frequency of magnetic fields is low and it becomes negative in an intermediate frequency range. The minimum value in the negative viscosity decreases and the frequency corresponding to the minimum viscosity increases as the amplitude of magnetic fields increases. The frequency range, under which the negative viscosity effect appears, becomes wider with an increase in the amplitude of ac magnetic fields.

(2) In the negative viscosity regions, the average angular velocity of particles is higher than the angular velocity of the solvent.

There are two mechanisms responsible for the occurrence of negative viscosity.

(a) When the amplitude of external ac magnetic fields is low, compared to the shear rate, resonance occurs between the rotational motions of the dipoles of particles and the fluctuation of the magnetic fields and beyond the resonance frequency, negative viscosity appears.

(b) When strong magnetic fields are applied in relatively low shear flow fields, the magnetic dipoles of particles are forced to stop for some period at low magnetic field frequency regions. However, negative viscosity appears in higher frequency regions where the dipoles rotate continuously following the magnetic field fluctuation.

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