Transport Coefficients and Orientational Distributions of Rodlike Particles with Magnetic Moment normal to the Particle Axis under Circumstances of a Simple Shear Flow and an External Magnetic Field

AKIRA SATOH

Professor, Akita Prefectural University, Japan

MASATAKA OZAKI[*1] and **TEPPEI ISHIKAWA**[*2]

[*1] *Professor and* [*2]*Graduate Student*, *Yokohama City University, Japan*

TAMOTSU MAJIMA

Professor, Chiba University, Japan

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ABSTARCT

We have investigated the influences of the magnetic field strength, shear rate, and random forces on transport coefficients such as viscosity and diffusion coefficient, and also on the orientational distributions of rodlike particles of a dilute colloidal dispersion. This dispersion is composed of ferromagnetic spheroidal particles with a magnetic moment normal to the particle axis. In the present analysis, these spheroidal particles are assumed to conduct the rotational Brownian motion in a simple shear flow as well as an external magnetic field. The basic equation of the orientational distribution function has been derived from the balance of the torques and solved numerically. The results obtained here are summarized as follows. For a very strong magnetic field, the rodlike particle is significantly restricted in the field direction, so that the particle points to a direction normal to the flow direction (and also to the magnetic field direction). However, the present particle does not exhibit a strong directional characteristic, which is one of the typical properties for the previous particle with a magnetic moment parallel to the particle axis. That is, the particle can rotate around the axis of the magnetic moment, although the magnetic moment nearly points to the field direction. The viscosity significantly increases with the field strength, as in the previous particle model. The particle of a larger aspect ratio leads to the larger increase in the viscosity, since such elongated particles induce larger resistance in a flow field. The diffusion coefficient under circumstances of an applied magnetic field is in reasonable agreement between theoretical and experimental results.

1. INTRODUCTION

Magnetic fluids (or ferrofluids) [1], magnetorheological (MR) suspensions [2], and electrorheological (ER) fluids [2] are functional fluids, which are made artificially to exhibit their functional properties under certain circumstances. These functional fluids are generated by dispersing functional particles, which respond to an external magnetic or electric fluid, in a base liquid. Spherical particles have generally been used as such functional particles, but magnetorheological or electrorheological effects of such dispersions cannot be obtained significantly. Large magnetorheological or electrorheological effects may be inevitable from a fluids engineering application point of view. In ER fluids, liquid crystals are attempted to be used in order to improve electrorheological effects [3]; such ER fluids with liquid crystals may exhibit sufficient electrorheological effects for applications in fluids engineering fields. Similarly, magnetorheological fluids with ferromagnetic rodlike particles may be expected to exhibit significant magnetorheological effects in an external magnetic field

From these backgrounds, our research group has been conducting a series of systematic studies concerning the behaviors of ferromagnetic particles in a dispersion under circumstances of an applied magnetic field as well as a flow field. First, we have studied the particle orientational distribution and rheological properties in a dilute dispersion in which particle-particle interactions are negligible [4,5]. Then, we have expanded such a study to a non-dilute dispersion, in which the interactions of the particle of interest with other particles in its same cluster are taken into account by means of the mean field approximation [6,7]. Furthermore, we have applied the mean field approximation to a dense dispersion in order to take into account the magnetic interactions with particles belonging to the neighboring clusters [8]. In these studies, we have used the model particle, for a rodlike ferromagnetic one, which has a magnetic moment in the particle axis direction.

For a dispersion, which is composed of rodlike particles with a magnetic moment normal to the particle axis, such as hematite particles [9-16], these particles are expected to incline in the direction normal to the magnetic field and exhibit different behaviors in a magnetic and flow fields. Hence, if we can control the behaviors of such particles in a flow field by an applied magnetic field, a functional fluid composed of such particles may be very attractive from an application point of view. However, studies concerning the behaviors of rodlike particles with a magnetic moment normal to the particle axis in a flow and magnetic fields, have not sufficiently been conducted theoretically, nor experimentally.

The present study, therefore, considers a dilute dispersion which is composed of ferromagnetic rodlike particles with a magnetic moment normal to the particle axis such as hematite particles, and investigates theoretically rheological properties and the orientational distribution of such dispersions under circumstances of an external magnetic field as well as a simple shear flow. Additionally, we attempt to clarify the dependence of the diffusion coefficient on the magnetic field strength, which helps us to understand the sedimentation phenomenon of these rodlike particles in the gravity field. Furthermore, the present numerical results are partially compared with experimental data obtained by another different study.

2. PARTICLE MODEL

As shown in Fig.1 [16], hematite particles observed experimentally have a nearly spheroidal shape, so that we adopt a spheroidal particle model shown in Fig. 2 as a ferromagnetic rodlike particle. We use the notation **e** for the unit vector denoting the particle direction, **m** (=$m\mathbf{e}_m$) for the magnetic moment which is normal to the particle axis, and $\mathbf{e}_m$ for the unit vector denoting the magnetic moment direction. For this particle model, the interaction energy U between such a particle and a uniform applied magnetic field **H**, and the torque acting on the particle, $\mathbf{T}^m$, are written as

$$U = -\mu_0 \boldsymbol{m}\cdot\boldsymbol{H} \ , \quad \boldsymbol{T}^m = \mu_0 \boldsymbol{m}\times\boldsymbol{H} \ , \tag{1}$$

in which μ_0 is the permeability of free space.

For the present particle model with a magnetic moment normal to the particle axis, one of the significant features is that the direction of the magnetic moment is never uniquely determined for a given particle direction, which is in contrast to the previous particle model with a magnetic moment along the particle axis. That is, the magnetic moment has the freedom of rotating around the particle axis to change its direction for a given particle direction. Hence we have to adopt a certain modeling concerning the direction of the magnetic moment for advancing the present analysis. Since we here consider the spheroidal particle with a large aspect ratio sufficiently apart from a spherical shape, it may be assumed that the characteristic time of the rotational motion around the particle axis is sufficiently shorter than that of the rotational motion around a line normal to the particle axis. The present study, therefore, uses such a particle model that the magnetic moment inclines in a direction which gives rise to the minimum of the interaction energy between the magnetic moment and an applied magnetic field for a given particle direction. Now we show the expression for the unit vector $\mathbf{e}_m$ denoting the direction of the magnetic moment.

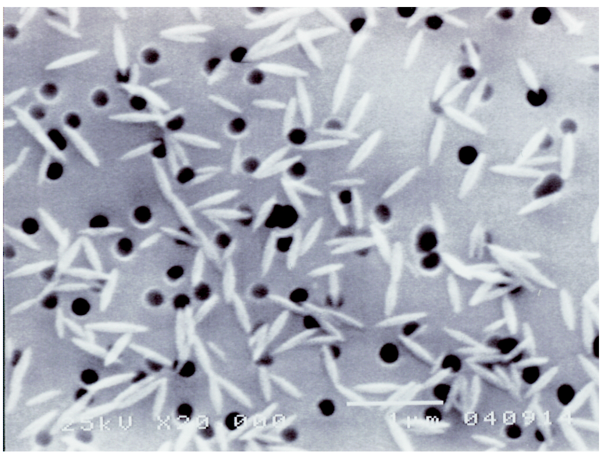

FIG.1. Electron microscopy image of a hematite particle dispersion ((l, d)=(0.45±0.05, 0.09±0.01) μm).

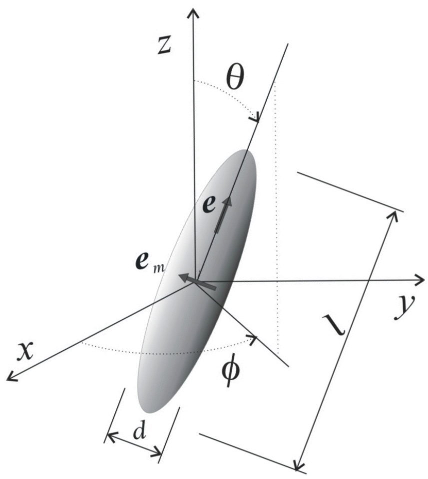

FIG.2. Particle model and system of coordinates.

With the definition of the coordinate system as shown in Fig. 2, the unit vector denoting the particle direction, **e**, can be expressed as

$$\boldsymbol{e} = (Sc, Ss, C) = Sc\boldsymbol{\delta}_x + Ss\boldsymbol{\delta}_y + c\boldsymbol{\delta}_z, \tag{2}$$

in which ($\boldsymbol{\delta}_x$,$\boldsymbol{\delta}_y$,$\boldsymbol{\delta}_z$) are the fundamental vectors of the orthogonal coordinate, and the following simplified notations have been used: S=sinθ,C=cosθ,s=sinφ, and c=cosφ. The body-fixed *XYZ*- coordinate of the particle can be obtained after the original *xyz*-coordinate system is rotated around the *z*-axis by the angle φ, and then rotated around the *y*-axis of the transformed coordinate by θ. In the new coordinate system, the *Z* axis coincides with the particle axis. The two unit vectors normal to each other and also to the particle axis, $\boldsymbol{e}_{\perp 1}$ and $\boldsymbol{e}_{\perp 2}$, can be obtained by setting (X, Y, Z)=(1,0,0) and (0,1,0) in the above-mentioned transformation:

$$\boldsymbol{e}_{\perp 1} = Cc\boldsymbol{\delta}_x + Cs\boldsymbol{\delta}_y - S\boldsymbol{\delta}_z, \quad \boldsymbol{e}_{\perp 2} = -s\boldsymbol{\delta}_x + c\boldsymbol{\delta}_y. \tag{3}$$

Using these unit vectors, the direction of the magnetic moment, $\mathbf{e}_m$, which gives rise to the minimum of the interaction energy with a magnetic field, can finally be expressed as

$$\boldsymbol{e}_m = \frac{Cc}{\sqrt{C^2c^2+s^2}}\boldsymbol{e}_{\perp 1} - \frac{s}{\sqrt{C^2c^2+s^2}}\boldsymbol{e}_{\perp 2} = \frac{1}{\sqrt{C^2c^2+s^2}}\left\{(C^2c^2+s^2)\boldsymbol{\delta}_x - S^2sc\boldsymbol{\delta}_y - SCc\boldsymbol{\delta}_z\right\}. \tag{4}$$

It is noted that the present study assumes a magnetic field to be applied along the *x*-axis, i.e., $\boldsymbol{H} = H\boldsymbol{\delta}_x$.

3. THE BASIC EQUATION FOR THE ORIENTATIONAL DISTRIBUTION FUNCTION

There are three kinds of torques acting on the particle: the torque due to the magnetic force, $\mathbf{T}^m$, the torque due to the rotational Brownian motion, $\mathbf{T}^{Br}$, and the torque due to the shear flow, $\mathbf{T}^{fl}$. Since the inertia term is negligible for usual colloidal dispersions, the governing equation for the rotational motion of particle can be derived from the balance of torques as

$$\boldsymbol{T}^{fl} + \boldsymbol{T}^{Br} + \boldsymbol{T}^{m} = 0. \tag{5}$$

The torque due to the rotational Brownian motion and the torque due to the shear flow are written, respectively, as

$$\boldsymbol{T}^{Br} = -kT\boldsymbol{e}\times\frac{\partial}{\partial \boldsymbol{e}}(\ln\Psi), \tag{6}$$

$$\boldsymbol{T}^{fl} = \eta_s\left\{X^C\boldsymbol{ee} + Y^C(\boldsymbol{I}-\boldsymbol{ee})\right\}\cdot(\boldsymbol{\Omega}-\boldsymbol{\omega}) - \eta_s Y^H(\boldsymbol{\varepsilon}\cdot\boldsymbol{ee}):\boldsymbol{E}, \tag{7}$$

in which Ψ is the orientational distribution function and will be defined in Sec.4, *k* is

Boltzmann's constant, T is the absolute temperature of fluid, $\boldsymbol{\Omega}$ and $\mathbf{E}$ are the rotational velocity vector and the rate-of-strain tensor of a simple shear flow, respectively, η_s is the viscosity of the base liquid, $\boldsymbol{\varepsilon}$ is called Eddington's epsilon (the three-rank tensor), $\mathbf{I}$ is the unit tensor, and X^C,Y^C and Y^H are resistance functions that are dependent only on the particle shape. Also, the expression for $\mathbf{T}^m$ has already been shown in Eq. (1).

If Eqs. (1),(6), and (7) are substituted into Eq.(5), we obtain the following equation:

$$\eta_s\left\{X^C\boldsymbol{ee}+Y^C(\boldsymbol{I}-\boldsymbol{ee})\right\}\cdot(\boldsymbol{\Omega}-\boldsymbol{\omega})-\eta_s Y^H(\boldsymbol{\varepsilon}\cdot\boldsymbol{ee}):\boldsymbol{E}-kT\boldsymbol{e}\times\frac{\partial}{\partial\boldsymbol{e}}(\ln\Psi)+\mu_0\boldsymbol{m}\times\boldsymbol{H}=0. \qquad (8)$$

If we multiply the both sides of Eq.(8) by (×**e**) and solve it for ($\boldsymbol{\omega}\times\mathbf{e}$), the equation giving the change in the particle direction $\dot{\mathbf{e}}$ is obtained as

$$\begin{aligned}\dot{\boldsymbol{e}}=\boldsymbol{\omega}\times\boldsymbol{e}=\boldsymbol{\Omega}\times\boldsymbol{e}+\frac{Y^H}{Y^C}\left\{\boldsymbol{E}\cdot\boldsymbol{e}-(\boldsymbol{E}:\boldsymbol{ee})\boldsymbol{e}\right\}-\frac{kT}{\eta_s Y^C}\frac{\partial}{\partial\boldsymbol{e}}(\ln\Psi)\\ -\frac{\mu_0 mH}{\eta_s Y^C}(\boldsymbol{e}\cdot\boldsymbol{h})\boldsymbol{e}_m.\end{aligned} \qquad (9)$$

In the derivation of this equation, the following relations have been used:

$$\left.\begin{aligned}&\boldsymbol{e}\times\left\{(\boldsymbol{ee})\cdot\boldsymbol{\Omega}\right\}=\boldsymbol{e}\times\left\{(\boldsymbol{ee})\cdot\boldsymbol{\omega}\right\}=0,\\ &\boldsymbol{e}\times\left\{(\boldsymbol{\varepsilon}\cdot\boldsymbol{ee}):\boldsymbol{E}\right\}=\boldsymbol{E}\cdot\boldsymbol{e}-(\boldsymbol{E}:\boldsymbol{ee})\boldsymbol{e},\\ &\boldsymbol{e}\times\left[\boldsymbol{e}\times\frac{\partial}{\partial\boldsymbol{e}}\left\{\ln(\Psi)\right\}\right]=-\frac{\partial}{\partial\boldsymbol{e}}\left\{\ln(\Psi)\right\},\\ &\boldsymbol{e}\times(\boldsymbol{m}\times\boldsymbol{H})=mH(\boldsymbol{e}\cdot\boldsymbol{h})\boldsymbol{e}_m,\end{aligned}\right\} \qquad (10)$$

in which $m=|\mathbf{m}|$, $H=|\mathbf{H}|$, and $\mathbf{h}=\mathbf{H}/H$ (=$\boldsymbol{\delta}_x$ in the present study), as defined previously.

4. EQUATION OF ORIENTATIONAL DISTRIBUTION FUNCTION

As shown in Fig.2, if the direction of the particle is described by the zenithal angle θ and the azimuthal angel φ, the orientational distribution function Ψ is defined such that the probability of the particle being found in a range (θ,φ) to (θ+dθ,φ+dφ) is written as $\Psi(\theta,\varphi,t)\sin\theta\, d\theta\, d\varphi$. Using this definition of the orientational distribution function, the average value of an arbitrary quantity G, which is dependent on the particle direction, is expressed as

$$\langle G\rangle=\int_0^{2\pi}\int_0^{\pi}G(\theta,\varphi)\Psi(\theta,\varphi,t)\sin\theta\, d\theta\, d\varphi. \qquad (11)$$

The average viscosity and diffusion coefficient will be evaluated later with this expression.

The basic equation for the orientational distribution function can be derived using the equation of continuity [17] and the equation for $\dot{\mathbf{e}}$ in Eq. (9). It is finally expressed as

$$\frac{\partial\Psi}{\partial t}=3\frac{Y^H}{Y^C}(\boldsymbol{E}:\boldsymbol{ee})\Psi-\left\{\boldsymbol{\Omega}\times\boldsymbol{e}+\frac{Y^H}{Y^C}(\boldsymbol{E}\cdot\boldsymbol{e})\right\}\cdot\frac{\partial\Psi}{\partial\boldsymbol{e}}$$
$$+\frac{kT}{\eta_s Y^C}\left(\frac{\partial}{\partial\boldsymbol{e}}\cdot\frac{\partial}{\partial\boldsymbol{e}}\right)\Psi+\frac{\mu_0 mH}{\eta_s Y^C}\left\{\Psi\frac{\partial}{\partial\boldsymbol{e}}\cdot((\boldsymbol{e}\cdot\boldsymbol{h})\boldsymbol{e}_m)+((\boldsymbol{e}\cdot\boldsymbol{h})\boldsymbol{e}_m)\cdot\frac{\partial\Psi}{\partial\boldsymbol{e}}\right\}. \tag{12}$$

Several relations, shown in Ref. [4], have been used in deriving Eq. (12). It is noted that $D_r=kT/\eta_s Y^C$ is called the rotational diffusion coefficient.

In the present study, we consider a problem in which the particles move in a simple shear flow in the *x*-axis direction under circumstances of a magnetic field applied in the *x*-axis direction. In this case, the flow velocity **U,** the angular velocity of the fluid **Ω**, and the rate-of-strain tensor **E** without particles are written as

$$\boldsymbol{U}=\dot{\gamma}y\boldsymbol{\delta}_x\ ,\quad \boldsymbol{\Omega}=-\frac{\dot{\gamma}}{2}\boldsymbol{\delta}_z\ ,\quad \boldsymbol{E}=\frac{\dot{\gamma}}{2}\begin{bmatrix}0&1&0\\1&0&0\\0&0&0\end{bmatrix}, \tag{13}$$

in which $\dot{\gamma}$ is the shear rate of the simple shear flow. If we take into account that Y^H/Y^C is nearly equal to unity for r_p (=*l*/*d*) »1 in the case of axisymmetric particles, Eq. (12) can finally be expressed as

$$\Lambda(\Psi)-Pe\Omega_s(\Psi)+\xi\Omega_{mx}(\Psi)=0, \tag{14}$$

in which the expressions for the operators, Λ and Ω_s, have already been shown in Ref. [17], and the operator Ω_{mx} is defined as follows:

$$\Omega_{mx}(\Psi)=\frac{1}{\sqrt{C^2c^2+s^2}}\left\{(1-2S^2c^2)\Psi+Sc\left(Cc\frac{\partial}{\partial\theta}-s\frac{\partial}{S\partial\varphi}\right)\Psi\right\}. \tag{15}$$

The non-dimensional numbers appearing in Eq. (14), *Pe* and ξ, are expressed as $Pe=\dot{\gamma}/D_r$ and $\xi=\mu_0 mH/kT$, respectively. *Pe* is called the Péclet number and means the ratio of the influence of the representative hydrodynamic shear force to that of the representative Brownian force. Similarly, ξ means the ratio of the representative magnetic particle-particle interaction energy to the thermal energy. It is noted that the following relations have been used in deriving Eq. (14).

$$\left.\begin{aligned}&\frac{\partial}{\partial\boldsymbol{e}}(\boldsymbol{e}\cdot\boldsymbol{h})\cdot\boldsymbol{e}_m=\frac{1}{\sqrt{C^2c^2+s^2}}(1-S^2c^2),\\&\frac{\partial}{\partial\boldsymbol{e}}\cdot\boldsymbol{e}_m=-\frac{Sc}{\sqrt{C^2c^2+s^2}}.\end{aligned}\right\} \tag{16}$$

5. SOLUTION OF THE BASIC EQUATION OF THE ORIENTATIONAL DISTRIBUTION FUNCTION

In the previous papers [4-8], the orientational distribution function was expanded in terms of spherical harmonics, and the basic equation for this distribution function was solved by means of Galerkin's method to get approximate solutions. In the present case, however, the basic equation in Eq. (14) has the irrational numbers of sine and cosine functions and, therefore, is very complicated to be solved by Galerkin's method. Hence, we adopt the numerical analysis approach based on the finite difference method in the present study. If we use the subscripts for specifying the position (θ, φ) on each grid point, for example, $\Psi_{i,j}$ for Ψ(θ,φ) and $\Psi_{i+1,,j+1}$ for Ψ(θ+Δθ, φ+Δφ), then Eq. (14) can be finite-differentiated as

$$\begin{aligned} & a_{i,j}^{(1)}\frac{\Psi_{i+1,j}-\Psi_{i-1,j}}{2\Delta\theta}+\frac{\Psi_{i+1,j}-2\Psi_{i,j}+\Psi_{i-1,j}}{(\Delta\theta)^2}+a_{i,j}^{(2)}\frac{\Psi_{i,j+1}-2\Psi_{i,j}+\Psi_{i,j-1}}{(\Delta\varphi)^2} \\ & -Pe\left[b_{i,j}^{(1)}\Psi_{i,j}+b_{i,j}^{(2)}\frac{\Psi_{i+1,j}-\Psi_{i-1,j}}{2\Delta\theta}+b_{i,j}^{(3)}\frac{\Psi_{i,j+1}-\Psi_{i,j-1}}{2\Delta\varphi}\right] \\ & \quad +\xi\left[c_{i,j}^{(1)}\Psi_{i,j}+c_{i,j}^{(2)}\frac{\Psi_{i+1,j}-\Psi_{i-1,j}}{2\Delta\theta}+c_{i,j}^{(3)}\frac{\Psi_{i,j+1}-\Psi_{i,j-1}}{2\Delta\varphi}\right] = 0, \end{aligned} \tag{17}$$

in which

$$\left.\begin{aligned} & a_{i,j}^{(1)}=\frac{C_i}{S_i}\ ,\quad a_{i,j}^{(2)}=\frac{1}{S_i^2}\ ,\quad b_{i,j}^{(1)}=s_jc_j(2C_i^2-S_i^2-2)\ , \\ & b_{i,j}^{(2)}=s_jc_jS_iC_i\ ,\quad b_{i,j}^{(3)}=-s_j^2\ ,\quad c_{i,j}^{(1)}=(C_i^2c_j^2+s_j^2)^{-1/2}(1-2S_i^2c_j^2)\ , \\ & c_{i,j}^{(2)}=(C_i^2c_j^2+s_j^2)^{-1/2}S_iC_ic_j^2\ ,\quad c_{i,j}^{(3)}=-(C_i^2c_j^2+s_j^2)^{-1/2}s_jc_j . \end{aligned}\right\} \tag{18}$$

Equation (17) is numerically solved by the method of successive approximation with taking the uniform distribution, Ψ=1/4π, as an initial condition. The following convergence criterion in the successive approximation procedure has been used:

$$\frac{\eta_{yx}^M(M+1)-\eta_{yx}^M(M)}{\eta_{yx}^M(M)} < 1\times10^{-8}, \tag{19}$$

in which η_{yx}^M is the viscosity and defined in Sec. 6.2, and $\eta_{yx}^M(M)$ is the value of η_{yx}^M which is evaluated using *M*-th approximation $\Psi^{(M)}$.

6. RESULTS

6.1. Particle Orientational Distribution

Figures 3 to 5 show the particle orientational distribution function for three different cases of the Péclet number: Fig.3 is for *Pe*=1, Fig.4 for *Pe*=5, and Fig.5 for *Pe*=20. In each figure, three different distributions are shown for three cases of the magnetic field strength, (a) ξ=1, (b) ξ=5, and (c) ξ=20. It is noted that a sharper peak at θ=90° and φ=90° means a significant inclination of the particle in the direction normal to the shear flow direction; an additional schematic figure concerning the state of the particle inclination for *H*=∞ is added to Figs. 3(c), 4(c), and 5(c) to make the reader understand the results more straightforwardly. Also, unless specifically noted, all results which will be shown bellow were obtained for r_p=5.

For the case of *Pe*=1, shown in Fig. 3, the rotational Brownian motion has the same order of the influence as the shear flow on the particle orientational distribution. Hence, if the influence of the magnetic force is also of the same order, the particle does not have a specific favorite direction, which is clearly shown in Fig.3(a). As the strength of the magnetic field increases from ξ=5 to 20, as shown in Figs.3(b) and (c), the orientational distribution function comes to exhibit a sharper peak at a position nearer to φ=90°. For a very strong magnetic field such as ξ=20, shown in Fig. 3(c), the magnetic field significantly dominates the other two mechanisms, and, therefore, the magnetic moment almost points to the magnetic field direction (or the flow field direction). This leads to a sharp peak at a position nearer to φ=90°. It should be noted that the direction of the rodlike particle is never restricted to θ=90°. In the previous studies [4-8], in which the rodlike particle has a magnetic moment along the particle axis, the orientational distribution function has a strong directional characteristic and has a sharp peak at θ=90° and φ=90° for a strong magnetic field, which is quite contrast to the result in Fig. 3(c). Hence, the above-mentioned characteristic concerning the orientational distribution function may be one of the significant features for the present particle model. That is, for a strong magnetic field, the magnetic moment is significantly restricted to the magnetic field direction (*x*-direction). Under these circum-

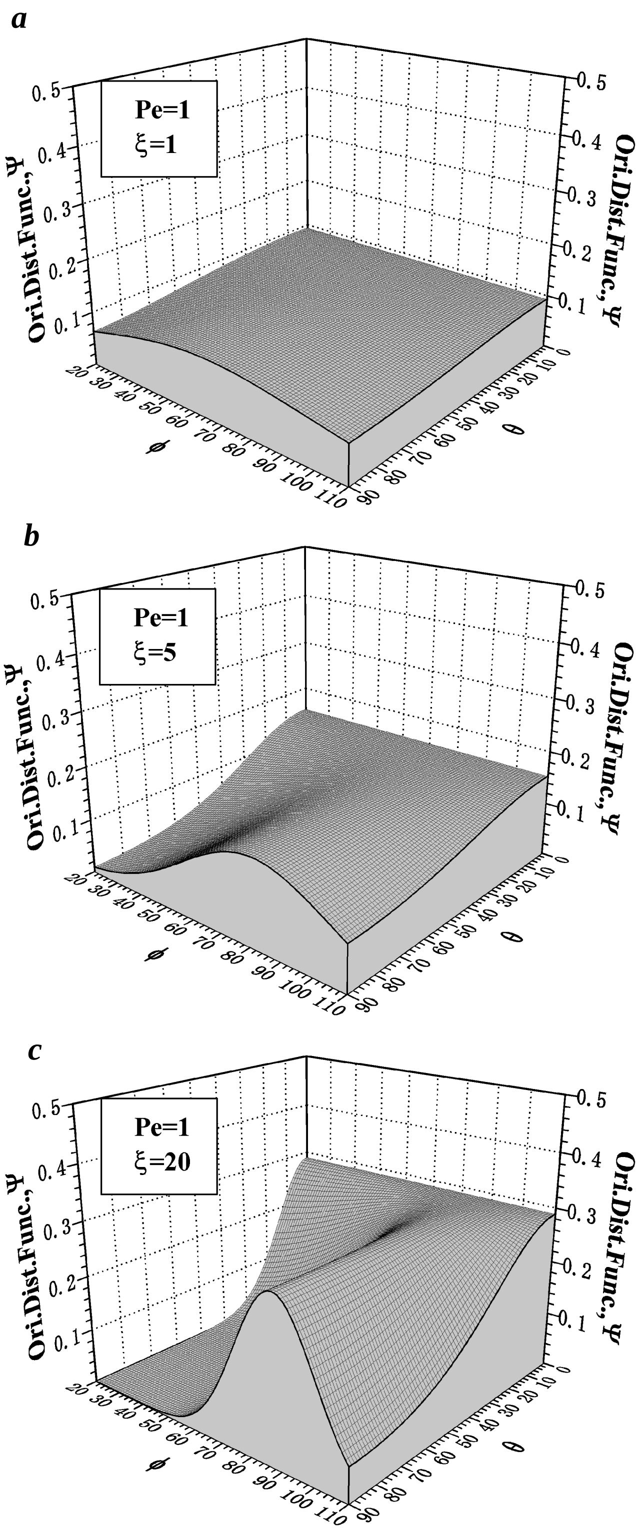

FIG.3. Orientational distribution function for *Pe*=1: (a) ξ=1 ; (b) ξ=5 ; (c) ξ =20.

stances, however, the particle can rotate around the *x*-axis freely to a certain degree unless the influence of the shear flow dominates the phenomenon. This induces a wide-spreaded sharp peak at a position of φ=90° and θ=0～180°, shown in Fig. 3(c), without a significant directional characteristic of a one-point sharp peak.

For the case of a larger shear rate *Pe*=5, shown in Fig.4, the influence of the shear flow comes to appear and gives rise to a peak around at φ=35° and θ=90° when the

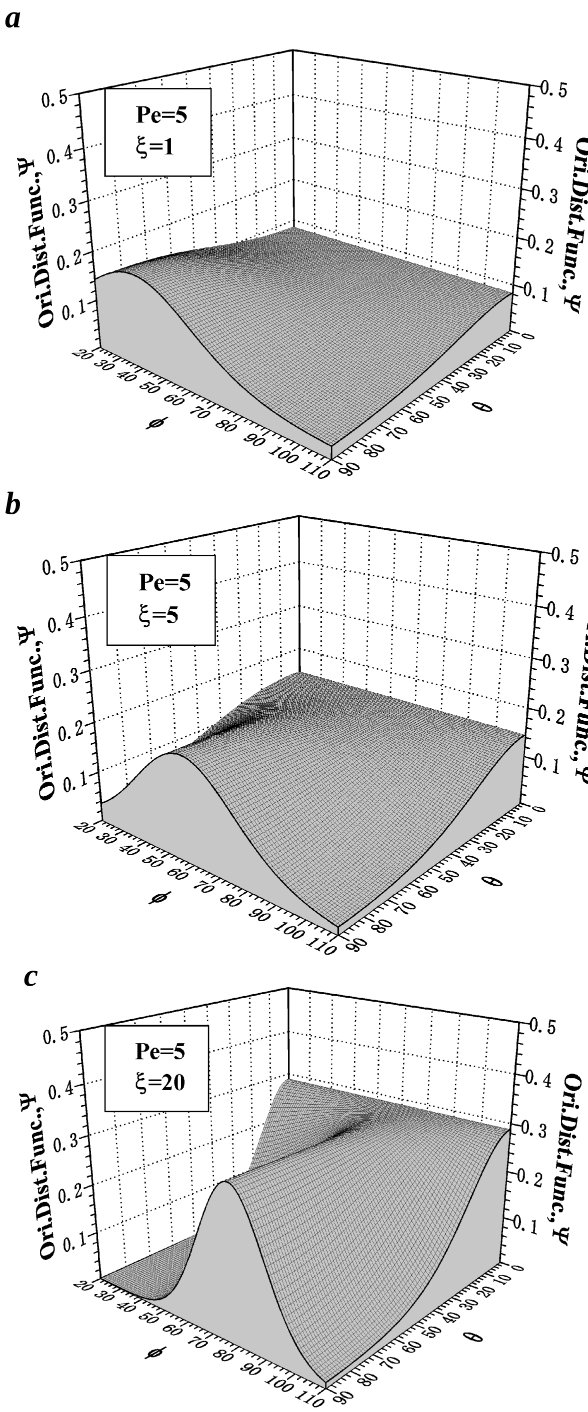

FIG.4. Orientational distribution function for *Pe*=5: (a) ξ=1 ; (b) ξ=5 ; (c) ξ=20.

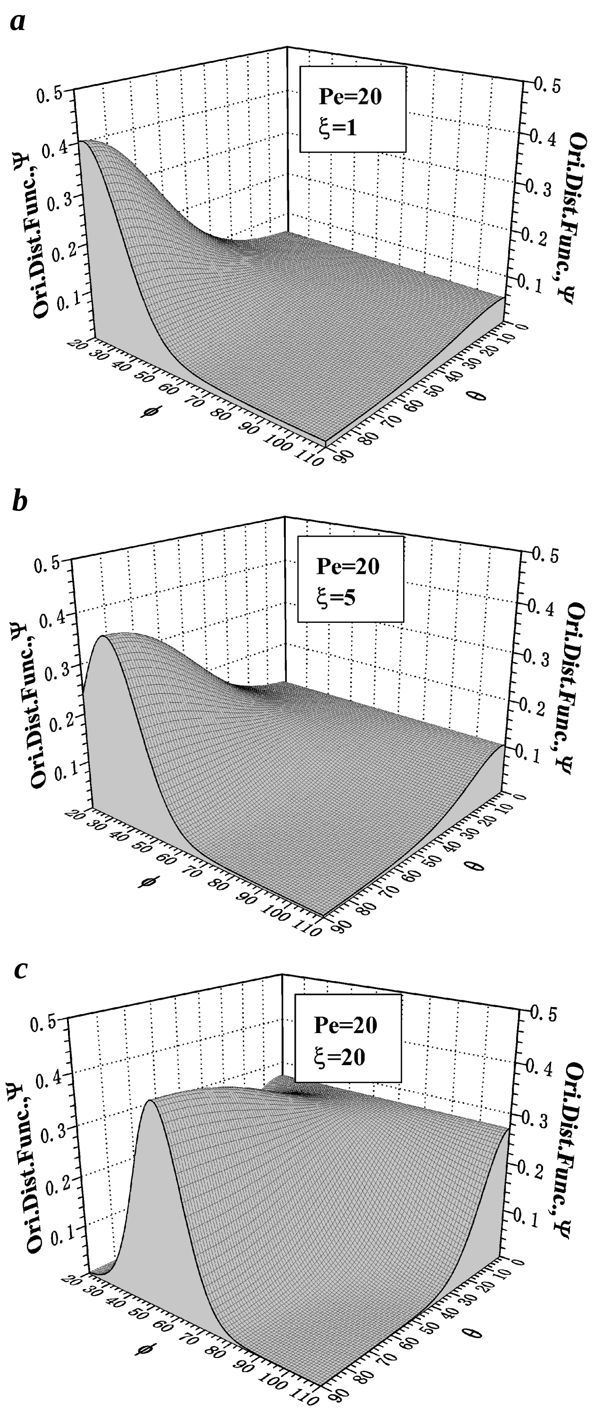

FIG.5. Orientational distribution function for *Pe*=20: (a) ξ=1 ; (b) ξ=5 ; (c) ξ=20.

magnetic field is weak such as ξ=1. As the magnetic field increases to ξ=5 and 20, the influence of the magnetic field comes to be dominant, so that the shape of the orientational distribution almost agrees with that shown in Fig. 3.

Figure 5 is for the case of a further larger shear rate of *Pe*=20, so that the influence of the simple shear flow comes to appear significantly. Since the shear flow significantly dominates the other two mechanisms for the result shown in Fig. 5(a), a sharp peak arises at a position nearer to the shear flow direction. This characteristic is in qualitative agreement with the results, shown in the previous papers [4-8], for the rodlike particle with a magnetic moment in the particle direction. With increasing the magnetic field from ξ=1, this peak arises at a position nearer to θ=90° without a significant increase in the height of the peak. As already pointed out, the magnetic moment is more significantly restricted to the magnetic field direction under circumstances of a stronger magnetic field, and the rodlike particle, however, has a sufficient freedom to rotate around the axis of the magnetic moment together with unchanging the direction of the magnetic moment. Since the influence of the shear flow is of the same order of the magnetic field for *Pe*=20, shown in Fig. 5(c), the height of the peak gradually decreases with changing values of θ from 90° to 0 for a constant value of φ which gives a maximum of the orientational distribution. This is in significant contrast to the result in Fig. 3(c). Figures 3(c), 4(c), and 5(c) clearly show that the peak comes to arise at a position nearer to φ=0 with increasing values of the Péclet number for the same value of ξ. This is because the influence of the shear flow becomes significant with values of *Pe*, compared with that of the magnetic force; it is noted that, in the limiting case of *Pe*=∞, the particle points to the flow direction of φ=0.

6.2. Rheological Properties

We show the equation of the viscosity before we proceed to the discussion of the results. In the present study, we concentrate our attention on the influence of the magnetic properties of the particle on the viscosity. The stress tensor $\boldsymbol{\tau}^m$ due to the magnetic properties of the particle is expressed as [17]

$$\boldsymbol{\tau}^{m} = -n\left\langle \boldsymbol{r}_i \boldsymbol{F}_i^{\,m} \right\rangle - \frac{n}{2}\left\langle \boldsymbol{\varepsilon}\cdot\boldsymbol{T}_i^{\,m} \right\rangle = -\frac{n}{2}\left\langle \boldsymbol{\varepsilon}\cdot\boldsymbol{T}_i^{\,m} \right\rangle = -\frac{n}{2}\mu_0 mH \left\langle \boldsymbol{\varepsilon}\cdot(\boldsymbol{e}_m \times \boldsymbol{h}) \right\rangle, \tag{20}$$

in which $\mathbf{F}_i^m$ is the magnetic force of the other particles acting on particle *i* and zero in the present dilute assumption, $\mathbf{T}_i^m$ is the torque acting on particle *i* due to the applied magnetic field, and *n* is the number density of particles. For the simple shear flow given in Eq.(13), the shear stress $\tau_{yx}{}^m$ can be expressed as

$$\tau_{yx}^{m} = \frac{n}{2}\mu_0 mH \left\langle \frac{S^2 sc}{\sqrt{C^2c^2+s^2}} \right\rangle . \tag{21}$$

From this equation, the shear viscosity $\eta_{yx}{}^m$ is written as

$$\eta_{yx}^{m}(\dot{\gamma}) = \frac{n}{2}\mu_0 mH \left\langle \frac{S^2 sc}{\sqrt{C^2c^2+s^2}} \right\rangle \Big/ \dot{\gamma} . \tag{22}$$

In the present paper, we discuss the viscosity $\eta_{yx}{}^M$ which is obtained by non-dimensionalizing $\eta_{yx}{}^m$ by the viscosity of the base liquid η_s, and further dividing this quantity by nV_p. That is,

$$\eta_{yx}^{M} = \eta_{yx}^{m*}/nV_p = \frac{\xi}{2Pe}\cdot\frac{Y^C}{V_p}\left\langle \frac{S^2 sc}{\sqrt{C^2c^2+s^2}} \right\rangle, \tag{23}$$

in which V_p is the volume of the spheroidal particle. The value of Y^C/V_p is dependent on the aspect ratio r_p and we here assume a sufficiently large aspect ratio, i.e., r_p »l. In this situation, the value of Y^C/V_p is given for a spheroidal particle as follows [17]:

$$\frac{Y^C}{V_p} = \frac{8s^3(2-s^2)r_p^2}{-2s+(1+s^2)L}, \tag{24}$$

in which

$$s=\sqrt{1-1/r_p^2}\ ,\quad L=\ln\left(\frac{1+s}{1-s}\right). \tag{25}$$

Figure 6 shows the influence of the magnetic field strength on the viscosity for three cases of the Péclet number, Pe=1, 5, and 20. Each curve shows that the viscosity exhibits a significant increase as the magnetic field becomes strong, and also that this significant increase starts at $\xi\simeq 1$ where the influence of the magnetic field comes to dominate that of the rotational Brownian motion. According to the previous results concerning the orientational distribution, the rodlike particle is more strongly restricted to a direction normal to the flow direction. This inclination of the particles induces more significant resistance in a flow field to yield a large increase in the viscosity. In the case of the same value of ξ/Pe, a larger increase in the viscosity is obtained for a larger value of Pe. This is because, under these circumstances, the influence of magnetic forces is more dominant than the influences of the shear flow and the rotational Brownian motion.

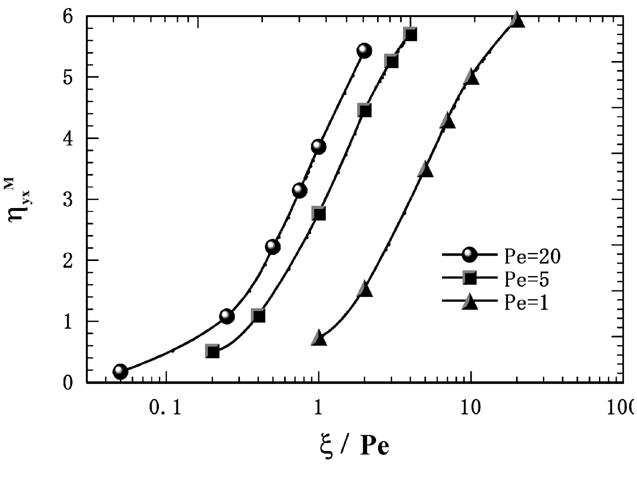

FIG.6. Viscosity as a function of ξ/Pe.

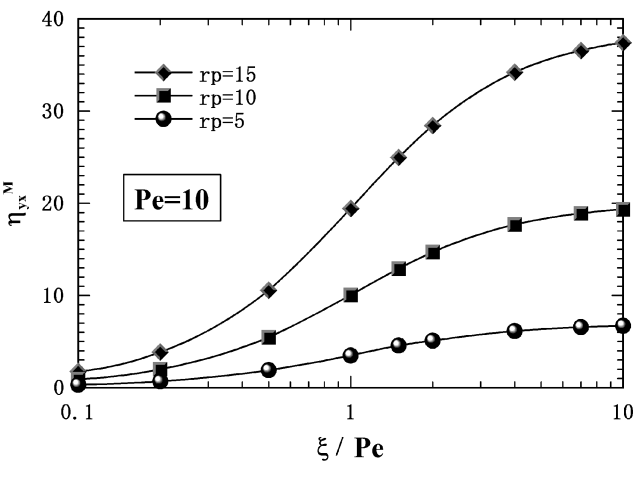

FIG.7. Influences of particle aspect ratio on viscosity for Pe=10.

The results in Fig. 7 were obtained for three cases of the particle aspect ratio, r_p=5, 10, and 15 for Pe=l0. As in the previous case of the particle with a magnetic moment along the particle axis [4], the rodlike particle with a larger aspect ratio exhibits a more significant increase in the viscosity as the magnetic field becomes strong.

The increase in the viscosity due to the magnetic properties relative to that of the base liquid, η_{yx}^{m*}, is obtained by multiplying η_{yx}^{M} by the volumetric fraction φ_V (=nV_p). We may, therefore, conclude from these results concerning the viscosity that, in order to generate a new functional fluid using ferromagnetic rodlike particles, particles with a large aspect ratio are desirable to be dispersed or suspended in a dense situation. Such suspensions or dispersions can be expected to exhibit a high magnetorheological effect under circumstances of an applied magnetic field.

6.3. Diffusion coefficients

Besides the application of ferromagnetic rodlike particles to fluids engineering fields, they are expected to be applicable to the surface change technology of the material surface, in which the orientations of rodlike particles are needed to be controlled under circumstances of an applied magnetic field, and then needed to be made to attach the material surface effectively in order to exhibit functional properties. In this case, we may need to control the orientation of particles in the sedimentation process under circumstances of gravity. In the present study, therefore, we concentrate our attention on the diffusion coefficient, which is the important parameter for determining the properties of the particle movement in an applied magnetic field and the gravity field.

If we use the notations $D_{\parallel}^{T}$ and $D_{\perp}^{T}$ for the diffusion coefficients of the parallel and normal movements to the particle axis, respectively, the expressions of these coefficients are expressed as [17]

$$D_{\parallel}^{T}=\frac{kT}{\eta_s}(X^A)^{-1}\ ,\quad D_{\perp}=\frac{kT}{\eta_s}(Y^A)^{-1}. \tag{26}$$

If the diffusion coefficients are non-dimensionalized by $kT/(6\pi a\eta_s$, $a=l/2)$, and the expressions of X^A and Y^A [17] are substituted into Eqs. (26), then the expressions of $D_{\parallel}^{T*}$ and $D_{\perp}^{T*}$ are written as

$$D_{\parallel}^{T*}=\frac{3}{8}\cdot\frac{-2s+(1+s^2)L}{s^3}\ ,\quad D_{\perp}^{T*}=\frac{3}{16}\cdot\frac{2s+(3s^2-1)L}{s^3}. \tag{27}$$

We now consider a phenomenon that the rodlike particle settles in the x-direction with the rotational motion under circumstances of an applied magnetic field in the x-direction. If the magnetic field is sufficiently strong, the particle should incline in a direction normal to the direction of the motion (or the x-direction) and settles with such an inclination. On the other hand, if the magnetic field is very weak, the particle should move with a random inclination due to the rotational Brownian motion. For a general case, the particle moves in the x-direction with a certain orientational distribution. If the diffusion coefficient for this general case is denoted by D_{ave}^{T*}, the expression can be written as [17]

$$D_{ave}^{T*} = (D_{\parallel}^{T*} - D_{\perp}^{T*})\left\langle (\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2 \right\rangle + D_{\perp}^{T*}, \tag{28}$$

in which $\langle\cdots\rangle$ means the average evaluated using the orientational distribution function which is obtained for a given magnetic field strength. If there is no external magnetic field and the system is in equilibrium, the orientational distribution is expressed as Ψ=1/4π, and, therefore, Eq. (28) reduces to $D_{ave}^{T*} = (D_{\parallel}^{T*} + 2D_{\perp}^{T*})/3$ in this situation; we use the notation D_{eq}^{T*} for this equilibrium diffusion coefficient. The value of D_{ave}^{T*} for an arbitrary magnetic field strength can be obtained by solving Eq. (17) for the limiting case of $Pe\rightarrow 0$ and evaluating the average in Eq. (28) with this numerical solution of the orientational distribution function. It is noted that this average procedure is justified under the situation that the characteristic time of the rotational Brownian motion is sufficiently shorter than that of the particle sedimentation under the gravity field.

Figure 8 shows the result of the diffusion coefficient D_{ave}^{T*}, which was obtained from Eq. (28). The orientational distribution approaches the equilibrium distribution as the magnetic field becomes zero, so that D_{ave}^{T*} converges to D_{eq}^{T*} for $\xi\rightarrow 0$. Since the particle orientation comes to be restricted to a direction normal to the x-axis, D_{ave}^{T*} converges to $D_{\perp}^{T*}$ with increasing values of ξ. The results seem to be independent of the particle aspect ratio, and this is straightforwardly shown in the following. From Eq. (28) and the expression for D_{eq}^{T*}, we can obtain the expression for the quantity of the ordinate in Fig. 8 as

$$\frac{D_{ave}^{T*} - D_{\perp}^{T*}}{D_{eq}^{T*} - D_{\perp}^{T*}} = \frac{(D_{\parallel}^{T*} - D_{\perp}^{T*})\left\langle (\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2 \right\rangle}{\frac{1}{3}(D_{\parallel}^{T*} - D_{\perp}^{T*})} = \frac{1}{3}\left\langle (\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2 \right\rangle. \tag{29}$$

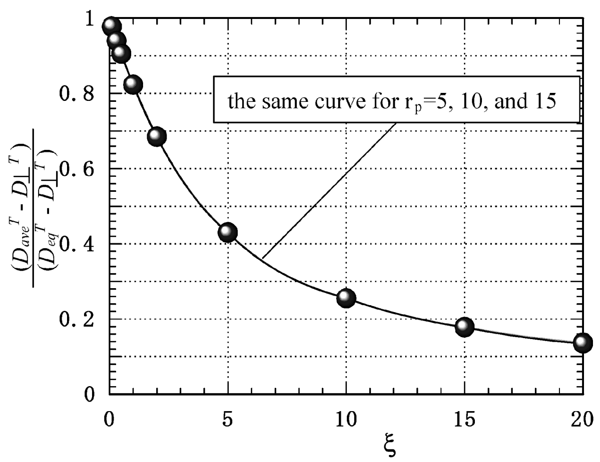

FIG.8. Influences of magnetic field on diffusion coefficients.

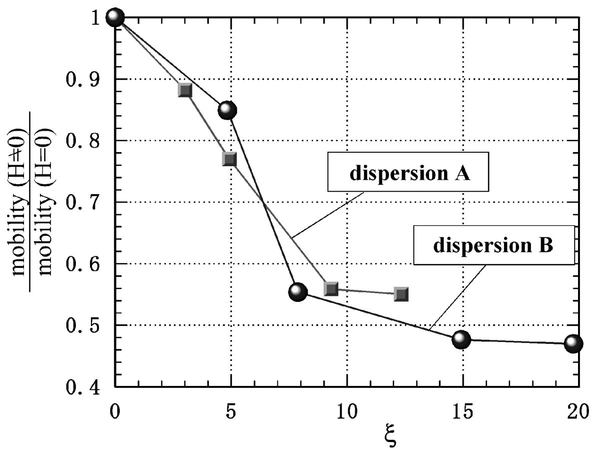

FIG.9. Experimental results concerning the strength dependence of the mobility of particles on the magnetic field strength (dispersion A and B for particles with (l, d)=(0.45±0.05, 0.09±0.01) μm and (l, d)=(0.48±0.08, 0.11±0.02) μm, respectively)

In the present limiting case of $\dot{\gamma}\to 0$ (or $Pe\to 0$), the average on the right-hand side in Eq. (29) depends only on ξ, but not on the particle aspect ratio, which is clear from Eq. (14). Hence, it is quite reasonable that the results in Fig.8 is independent of the aspect ratio.

Figure 9 shows the relationship between the mobility velocity of the rodlike particles and the magnetic field strength. These results were experimentally obtained by evaluating the velocity of the interface of a dispersion, which is composed of hematite particles shown in Fig. 1, by means of the electrical mobility equipment facility [16]; the dispersions A and B are significantly dilute and the volumetric fraction of particles is about 0.002 % for both cases. How the rodlike particle orients under circumstances of no external magnetic field is not clear at the moment, which is a subject to be clarified in future. If large particles and hydrodynamic interactions between them are more dominant than the rotational Brownian motion, it may be possible to use the diffusion coefficient $D_{\parallel}^{T*}$ for the case of ξ=0. On the other hand, if hydrodynamic interactions between particles are not significantly important and the rotational Brownian motion is dominant, we may use D_{eq}^{T*} for the case of ξ=0. According to this consideration, Fig. 10 is obtained by replotting the result shown in Fig. 8 for a straightforward comparison with the experimental result shown in Fig. 9; Fig. 10(a) is for the former case and Fig. 10(b) is for the latter case. The quantity of the ordinate in each figure is written as

$$\frac{D_{ave}^{T*}}{D_{\parallel}^{T*}}=\frac{(D_{\parallel}^{T*}-D_{\perp}^{T*})\left\langle(\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2\right\rangle+D_{\perp}^{T*}}{D_{\parallel}^{T*}}=\left\langle(\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2\right\rangle+\left\{1-\left\langle(\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2\right\rangle\right\}\frac{D_{\perp}^{T*}}{D_{\parallel}^{T*}}, \tag{30}$$

$$\frac{D_{ave}^{T*}}{D_{eq}^{T*}}=\frac{(D_{\parallel}^{T*}-D_{\perp}^{T*})\left\langle(\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2\right\rangle+D_{\perp}^{T*}}{\frac{1}{3}\left(D_{\parallel}^{T*}+2D_{\perp}^{T*}\right)}=\left\{\left\langle(\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2\right\rangle+\left\{1-\left\langle(\boldsymbol{e}\cdot\boldsymbol{\delta}_x)^2\right\rangle\right\}\frac{D_{\perp}^{T*}}{D_{\parallel}^{T*}}\right\}\Big/\frac{1}{3}\left(1+2D_{\perp}^{T*}/D_{\parallel}^{T*}\right). \tag{31}$$

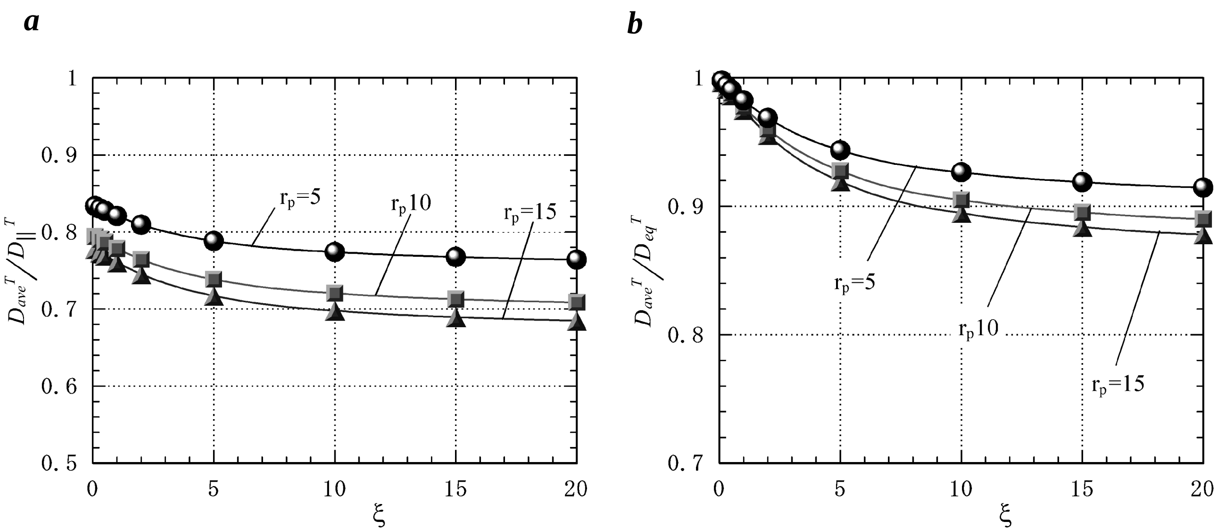

FIG.10. Calculation results of diffusion coefficients for comparison to the results in Fig. 9: (a) $D_{ave}^{T*}/D_{\parallel}^{T*}$ and (b) D_{ave}^{T*}/D_{eq}^{T*} as a function of ξ.

The result in Fig. 9 is in quantitatively reasonable agreement with the theoretical result in Fig. 10(a) except the small value range of ξ. In contrast, a good agreement between the experimental and numerical results cannot be obtained in Fig. 10(b). As already pointed out, from Figs. 10(a) and 10(b), it seems to be reasonable that the movement parallel to the particle axis is regarded as the motion for ξ=0; otherwise, such a significant change in the diffusion coefficient in the experimental results cannot be obtained. Other mechanisms concerning the change in D_{ave}^{T*} may be hydrodynamic interactions between particles and the distribution of the particle size. Finally, we may conclude that it is possible to control the sedimentation speed by means of an applied magnetic field. This characteristic may be very important if we consider the development of surface-changing techniques using ferromagnetic rodlike particles.

7. CONCLUSIONS

We have investigated the influences of the magnetic field strength, shear rate, and random forces on transport coefficients such as viscosity and diffusion coefficient, and also on the orientational distributions of rodlike particles of a dilute colloidal dispersion. This dispersion is composed of ferromagnetic spheroidal particles with a magnetic moment normal to the particle axis. In the present analysis, these spheroidal particles are assumed to conduct the rotational Brownian motion in a simple shear flow as well as an external magnetic field. The basic equation of the orientational distribution function has been derived from the balance of the torques and solved numerically. The results obtained here are summarized as follows. For a very strong magnetic field, the rodlike particle is significantly restricted in the field direction, so that the particle points to a direction normal to the flow direction (and also to the magnetic field direction). However, the present particle does not exhibit a strong directional characteristic, which is one of the typical properties for the previous particle with a magnetic moment parallel to the particle axis. That is, the particle can rotate around the axis of the magnetic moment, although the magnetic moment nearly points to the field direction. The viscosity significantly increases with the field strength, as in the previous particle model. The particle of a larger aspect ratio leads to the larger increase in the viscosity, since such elongated particles induce larger resistance in a flow field. The diffusion coefficient under circumstances of an applied magnetic field is in reasonable agreement between theoretical and experimental results.

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