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THEORY OF ANISOTROPIC FERROMAGNETIC COLLOIDS

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Introduction. In present theoretical descriptions of the flow of a ferrofluid (a stabilized surface-active colloid consisting of ferromagnetic particles in a fluid carrier) various approaches and approximations have been used [1-12]. One of the first hydrodynamic models of a ferrofluid [1] assumed that the magnetization relaxation time τ was small in comparison to the characteristic macroscopic times of the problem; in this case one can put $\tau = 0$, and consider the magnetization of the medium as given by an equilibrium equation of state.

Unlike this approach, in [2-4], models were developed where the relaxation time τ is finite, so that the change in magnetization is given by a relaxation equation, which together with the other equations of the theory describe the magnetization dynamics of magnetically isotropic ferrofluids. From the microscopic point of view, magnetic isotropy of the ferrofluid implies that the magnetization is frozen in the particles or the generalized magnetic anisotropy constant K of the particles is infinite. This means that the orientational change of the magnetization in these models is determined entirely by the rotation of the particles (Brownian relaxation [5] with relaxation time τ_2).

In the case where the thermal energy kT is comparable to the (finite) magnetic anisotropy energy of a particle kV , where V is the volume of a particle and k is the Boltzmann constant, the latter energy is not large enough to keep the magnetic moment frozen inside the particle against the thermal fluctuations. The orientational change of the magnetization in the general case where the magnetic moments are partially frozen will be determined both by diffusion of the moment with respect to the particle (Néel relaxation with relaxation time τ_1 [5]), and by the Brownian rotation of the particles, which for hydrodynamical flow and for $\tau_1 \ll \tau_2$ will determine the macroscopic magnetic anisotropy of the ferrofluid.

The first treatment of the effect of partial freezing of the magnetic moments on the effective viscosity of the ferrosuspension was considered in [10]. However, this treatment, based on kinetic ideas, did not yield a definite formula for the effective viscosity, which would contain the previously known formula for the rigid magnetic dipole model, or a general macroscopic equation of motion.

A series of papers [8, 11, 12] are of interest, in which a general macroscopic equation was formulated taking into account the finiteness of the magnetic anisotropy energy of the particles by introducing into the theory a macroscopic vector parameter A defined as the internal magnetic anisotropy field. In the equilibrium state this parameter is taken to be proportional to the magnetization $A = \alpha m$, where the coefficient α is defined as a parameter giving the degree of freezing of the magnetic moments with respect to the particles.

Calculations of the dependence of the viscosity on the field strength ξ in a state of partial equilibrium using these equations [8] yields remarkable agreement between theory and experiment for a suspension of magnetite in kerosene. However, this agreement should not be taken as unequivocal support for the above interpretation of the parameter α (or for the assumption that the deviation of the experimental viscosity dependence from the rigid dipole

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model is due only to partial freezing of the magnetic moments) since the agreement between theory and the experiment in [8] occurs because the experimental dependence of the parameter α on ξ is substituted into the theory. Also one cannot exclude the effects of impurities in the experimental data, the influence of such factors as the presence of clusters, chainlike aggregates, polydispersion of the particles, and other factors not taken into account in the theory [8]. It is well known that these effects are important in real ferrofluids at high concentrations and significantly affect the hydrodynamics and other processes. For example, the features of the Debye relaxation spectra for ferrofluids such as a suspension of magnetite in kerosene were explained in [9] assuming the existence of two kinds of particles with different relaxation times τ_1 and τ_2 such that $\tau_1 \ll \tau_2$, i.e., the assumption of an isotropic, rigid dipole component in the ferrofluid. It is thus of interest to study the effect of particle-size dispersion on the effective viscosity of the ferrofluid.

In addition, the theory [8] uses a vector anisotropy field A to characterize the partial freezing of the magnetic moments. Although such a device is widely used in physics (such as polymers, where internal scalar, vector, and tensor variables are used to characterize relaxation processes), in a state of partial equilibrium, the relaxation equation used in [8] reduces to a single relaxation equation for the magnetization, which can be written as the sum of two parts: the instantaneous value m_s and the relaxation part m_p . This suggests that instead of introducing a vector anisotropy parameter in the theory, from the beginning we write the magnetization as a sum of two parts, each of which is described by its own relaxation equation.

The present paper discusses these aspects of the theory of an anisotropic ferrofluid. The theory is developed using the ideas of interpenetrating magnetizable continua worked out in [3]. We consider a physical model of the magnetization of the ferrofluid in which there are two relaxation mechanisms. The magnetization is written as a sum of two parts, m_1 and m_2 , with characteristic relaxation times given by τ_1 and τ_2 . In a real ferrosuspension this situation can occur in two cases: 1) the ferrofluid consists of two types of particles with significantly different magnetic properties (the two-particle model discussed in Secs. 1 and 2); 2) the ferrofluid consists of only a single kind of particle, but the magnetic moments are partially frozen (the single-particle model discussed in Secs. 3 and 4). In the first case, the relaxation times τ_1 and τ_2 will be determined by the usual Brownian relaxation times. In the second case τ_1 and τ_2 are determined by Brownian and Neel relaxation times.

The anisotropy in both cases arises due to the effect of the two relaxation mechanisms of the magnetization when $\tau_1 \ll \tau_2$. If we apply an external field over a period of time of order τ_1 , the component of the ferrofluid magnetization in the direction of the field will be given by m_1 and will increase to the value m_2 over a time of order τ_2 . Under certain dynamical conditions, it is possible to separate the effects of the magnetizations m_1 and m_2 on processes which also can arise from flow anisotropy. We illustrate the theory by an analysis of the effective viscosity of a ferrosuspension.

1. Basic Equations for the Two-Particle Model of an Anisotropic Ferrocolloid. The set of equations for a ferrosuspension mixture consisting of interpenetrating components capable of separating was formulated in [3]. We write out the equations in modified form for the following physical model. The ferrofluid will be considered as a suspension of ferromagnetic particles of two kinds, with different magnetic properties, in a fluid solvent.* The different properties of the particles may be due to differing particle sizes, or due to the formation of coagulated colloidal particles [7] which lead to different values of the particle magnetic moments. Other interpretations are also possible. We assume that the magnetic moments of one group of particles are completely frozen; in the other group the moments are assumed to be partially frozen.† The magnetization relaxation times for the two groups of particles will be different in all cases; thus, it is necessary to consider two relaxation equations for the magnetization. In addition, the force created by the rotating magnetic moments will be smaller for particles of smaller size (or for particles with partially frozen magnetic moments) than for the other group of particles. This is due to the differing

*This is an approximation because in most real ferrosuspensions there is dispersion in particle sizes which obeys a distribution law.

†Here we do not describe in detail the processes of partial freezing of magnetic moments; it is mentioned only to illustrate the differences in magnetic properties of the two groups of particles. Larmor precession of the magnetic moments is everywhere ignored.

rotational velocities of the particles, $\omega_1 \neq \omega_2$. We will assume that particles of both groups are frozen translationally in the fluid solvent so that their translational velocities will be equal to the velocity of the fluid. Hence we have $\mathbf{v}_1 = \mathbf{v}_2 = \mathbf{v}$.

Under the above assumptions, and in the absence of free charges, and vanishing conductivity and polarization of the medium, the set of equations developed in [3] can be written as follows:

The electrodynamic equations:

$$\begin{aligned} \nabla \times \mathbf{e} + \frac{\partial \mu_0 \mathbf{h}}{\partial t} &= - \frac{\partial \mu_0 \mathbf{m}}{\partial t} - \nabla \times (\mu_0 \mathbf{m} \times \mathbf{v}), \quad \nabla \times \mathbf{h} - \frac{\partial \varepsilon_0 \mathbf{e}}{\partial t} = 0, \\ \nabla \cdot \mu_0 (\mathbf{h} + \mathbf{m}) &= 0, \quad \nabla \cdot \varepsilon_0 \mathbf{e} = 0, \quad \mathbf{m} = \mathbf{m}_1 + \mathbf{m}_2. \end{aligned} \quad (1.1)$$

The equations of continuity, the conservation laws for momentum and angular momentum, and the entropy production inequality γ :

$$\begin{aligned} \rho \cdot + \rho \nabla \cdot \mathbf{v} &= 0, \quad \rho \mathbf{v} \cdot = \nabla \cdot \mathbf{t} + (\mu_0 \mathbf{m} \cdot \nabla) \mathbf{h} - \mathbf{v} \times (\mu_0 \mathbf{m} \cdot \nabla) \mathbf{e} - \rho \mu_0 \mathbf{m} \cdot \times \varepsilon_0 \mathbf{e}, \\ \rho_\alpha I_\alpha \omega_\alpha &= \nabla \cdot \mathbf{s}_\alpha + \mathbf{t}_\alpha \times \cdot \mathbf{I} + \mu_0 \mathbf{m}_\alpha \times \eta + U(\alpha) \mathbf{L}, \quad \eta = \mathbf{h} - \mathbf{v} \times \varepsilon_0 \mathbf{e}, \quad \mathbf{t} = \mathbf{t}_1 + \mathbf{t}_2, \\ \rho T \gamma &= - T^{-1} \mathbf{Q} \cdot \nabla T + (\omega_2 - \omega_1) \cdot \mathbf{L} + D \mathbf{t}^s \cdot \cdot (\nabla \mathbf{v})^s + \end{aligned} \quad (1.2)$$

$$+ \sum_{\alpha=1}^2 \left[\mathbf{s}_\alpha \cdot \cdot \nabla \omega_\alpha + \frac{1}{2} (\mathbf{t}_\alpha \times \cdot \mathbf{I}) \cdot (\nabla \times \mathbf{v} - 2\omega_\alpha) + (\rho_\alpha \mu_0 \dot{\mu}_\alpha - \omega_\alpha \times \mu_0 \mathbf{m}_\alpha) \cdot D \eta_\alpha + \omega_\alpha \mathbf{R}_\alpha \right] \geq 0, \quad \alpha = 1, 2,$$

where

$$\eta = - \frac{\partial \varphi}{\partial T}; \quad \pi = - \frac{\partial \varphi}{\partial \rho^{-1}}; \quad E \eta_\alpha = \frac{\rho}{\mu_0 \rho_\alpha} \frac{\partial \varphi}{\partial \mu_\alpha}; \quad \eta = E \eta_\alpha + D \eta_\alpha; \quad \mathbf{R}_\alpha = E \eta_\alpha \times \mu_0 \mathbf{m}_\alpha; \quad (1.3)$$

$$\begin{aligned} \mathbf{t} &= -\pi \mathbf{I} + D \mathbf{t}; \quad \mathbf{m}_\alpha = \rho_\alpha \mu_\alpha; \quad \rho = \rho_1 + \rho_2; \quad \rho \mu = \rho_1 \mu_1 + \rho_2 \mu_2; \quad \mathbf{I} = \{\delta^{ij}\}; \\ 2\mathbf{t}^s &= \{t^{ik} + t^{ki}\}; \quad U(\alpha) = (-1)^{\alpha-1}. \end{aligned}$$

Here \mathbf{e} and \mathbf{h} are the electric and magnetic field vectors, ε_0 , μ_0 are the electric and magnetic constants, \mathbf{t}_α , \mathbf{s}_α are the force and torque stress tensors, \mathbf{L} is the diffusive interaction torque, φ is the free energy, \mathbf{Q} is the heat flux vector, ∇ is the Hamiltonian operator; I_α are scalar constants with units of moment of inertia per unit mass. The operations \cdot and \times stand for the scalar and vector products, respectively. For the density ρ_α we take the ratio of the mass of particles of type α and the surrounding part of the fluid to the volume of the entire mixture. Relations between ρ_α , I_α , and the number of particles n_α and the microscopic characteristics are not specified. The dot superscript denotes a total time derivative.

The set of equations (1.1) and (1.2) must be supplemented by constitutive equations for the 19 irreducible thermodynamic fluxes; these equations relate the fluxes to the thermodynamic forces via tensor kinetic coefficients which will be functions of the magnetization vectors. Here we do not write out the explicit general form of these equations, but give a particular form of the constitutive equations used below for the case of an incompressible isothermal momentless ferrofluid

$$\begin{aligned} t^{ih} &= -p \delta^{ih} + \alpha_2 (v_{i,h} + v_{h,i}) + \varepsilon^{ihk} (b_1^s + b_2^s), \quad L^h = l_1 (\omega_2^h - \omega_1^h), \quad \mathbf{b}_\alpha = \\ &= \beta_1^{(\alpha)} (\nabla \times \mathbf{v} - 2\omega_\alpha), \\ \rho_1 \dot{\mu}_1 - \omega_1 \times \mathbf{m}_1 &= \frac{1}{\tau_1} (m_{10} \dot{\mathbf{h}} - \mathbf{m}_1) + \frac{1}{\tau} (m_{20} \dot{\mathbf{h}} - \mathbf{m}_2), \quad m_{10} = K_1 h, \quad \mathbf{h} = \mathbf{h}/h, \\ \rho_2 \dot{\mu}_2 - \omega_2 \times \mathbf{m}_2 &= \frac{1}{\tau_2} (m_{20} \dot{\mathbf{h}} - \mathbf{m}_2) + \frac{1}{\tau} \frac{K_1}{K_2} (m_{10} \dot{\mathbf{h}} - \mathbf{m}_1), \quad m_{20} = K_2 h. \end{aligned} \quad (1.4)$$

Here K_1 and K_2 are the equilibrium susceptibilities of the ferrosuspension from particles of the first and second kinds, and p is the hydrostatic pressure. The relaxation times τ_1 , τ_2 , τ are related to these constants [3] as follows: $\tau_1 = \mu_0 K_1 / \mu_1$ ⁽¹⁾, $\tau_2 = \mu_0 K_2 / \mu_1$ ⁽²⁾, $\tau = \mu_0 K_2 / \mu_2$ ⁽¹⁾, and they satisfy the thermodynamic restrictions

$$K_1, K_2, \tau_1, \tau_2 \geq 0, \quad \tau^2 K_1 - \tau_1 \tau_2 K_2 \geq 0. \quad (1.5)$$

Equations (1.1) to (1.5) are a closed set of equations for \mathbf{v} , ω_α , \mathbf{m}_α , and h for the case $e = 0$.

2. Effective Viscosity in the Two-Particle Model. 2.1. General Expression for the Effective Viscosity. We determine the effective viscosity of the ferrosuspension for flow into a plane aperture in a uniform transverse magnetic field and a pressure gradient $p_{,1} = \text{const}$. The x_1 axis of a Cartesian coordinate system $Ox_1x_2x_3$ is taken along the direction of flow, while the x_2 axis is along the external magnetic field. The x_3 axis is perpendicular to the plane of motion. The velocity and vector magnetization fields are written in the form

$$\begin{aligned} v &= v_1(x_2), \quad v_2 = v_3 = 0, \quad \omega_\alpha = \omega_\alpha^3(x_2), \quad \omega_\alpha^1 = \omega_\alpha^2 = 0, \\ \mathbf{m}_1 &= m_{10}\mathbf{h} + \mathbf{m}_{1\perp}, \quad \mathbf{m}_2 = m_{20}\mathbf{h} + \mathbf{m}_{2\perp}. \end{aligned} \quad (2.1)$$

From the assumed smallness of the relaxation times, it follows that the \mathbf{m}_α are of the same order and $\omega_2\tau_2 \ll 1$, $\omega_1\tau_1 \ll 1$. Then the magnetization equation (1.4) in the linear approximation gives

$$\begin{aligned} m_{1\perp} &= \frac{m_{20}}{\tau_2\Delta} \omega_2^3 - \frac{m_{10}}{\tau_1\Delta} \omega_1^3, \quad m_{2\perp} = \frac{m_{10}}{\tau_1\Delta} \frac{K_2}{K_1} \omega_1^3 - \frac{m_{20}}{\tau_2\Delta} \omega_2^3, \quad \Delta = \\ &= \frac{K_1\tau^2 - \tau_1\tau_2 K_2}{\tau_1\tau_2 K_1 \tau^2} > 0. \end{aligned} \quad (2.2)$$

The equations of motion (1.1), using (2.1) take the form

$$\begin{aligned} -p_{,1} + (\alpha_2 + \beta_1^{(1)} + \beta_1^{(2)})v_{,22} + 2\beta_1^{(1)}\omega_{1,2}^3 + 2\beta_1^{(2)}\omega_{2,2}^3 &= 0, \\ -2\beta_1^{(1)}(v_{,2} + 2\omega_1^3) + \mu_0 m_{1\perp} h + l_1(\omega_2^3 - \omega_1^3) &= 0, \\ -2\beta_1^{(2)}(v_{,2} + 2\omega_2^3) + \mu_0 m_{2\perp} h - l_1(\omega_2^3 - \omega_1^3) &= 0. \end{aligned} \quad (2.3)$$

With the help of (2.2) we can write

$$\begin{aligned} -p_{,1} + (\alpha_2 + \Delta\eta)v_{,22} &= 0, \\ \omega_1^3 &= \Delta_1^{-1} [2\beta_1^{(1)}B_2' + 2\beta_1^{(2)}B_3'] v_{,2}, \quad \omega_2^3 = \Delta_1^{-1} [2\beta_1^{(2)}B_1' + 2\beta_1^{(1)}B_3'] v_{,2}, \\ B_1 &= 4\beta_1^{(1)} + \mu_0 m_{10} h / \tau_2 \Delta, \quad B_2 = 4\beta_1^{(2)} + \mu_0 m_{20} h / \tau_1 \Delta, \quad B_3 = m_{20} h \mu_0 / \tau \Delta, \\ B_2' &= B_2 + l_1, \quad B_1' = B_1 + l_1, \quad B_3' = B_3 + l_1, \quad \Delta_1 = -B_1 B_2 - l_1(B_2 + B_1 - 2B_3) + B_3^2, \\ B_4 &= m_{10} \mu_0 h / \tau_2, \quad B_5 = m_{20} h \mu_0 / \tau_1; \end{aligned} \quad (2.4)$$

$$\begin{aligned} \Delta\eta &= [-\beta_1^{(1)}B_4B_2 - \beta_1^{(2)}B_5B_1 - (\beta_1^{(1)} + \beta_1^{(2)})l_1(B_5 + B_4 - 2B_3\Delta) + \\ &+ (\beta_1^{(1)} + \beta_1^{(2)})B_3^2\Delta + 8\beta_1^{(1)}\beta_1^{(2)}B_3\Delta] / \Delta\Delta_1. \end{aligned} \quad (2.5)$$

Equation (2.5) gives the correction $\Delta\eta$ to the usual shear viscosity. The correction is determined by the field strength h , the rotational viscosities $\beta_1^{(1)}$, $\beta_1^{(2)}$, l_1 , and the relaxation times τ_1 , τ_2 , τ .

2.2 Effect of Dispersion in the Values of the Magnetic Moments of the Particles on the Effective Viscosity. We consider the viscosity (2.5) in the limit $\tau \rightarrow \infty$. In this case we have $\Delta = (\tau_1\tau_2)^{-1}$, and the viscosity can be written out explicitly in the form

$$\begin{aligned} \Delta\eta &= \left[\frac{\beta_1^{(1)}m_{10}\mu_0 h \tau_1}{4\beta_1^{(1)} + \tau_1\mu_0 m_{10} h} + \frac{\beta_1^{(2)}m_{20}\mu_0 h \tau_2}{4\beta_1^{(2)} + \tau_2\mu_0 m_{20} h} + \right. \\ &+ (\beta_1^{(1)} + \beta_1^{(2)})l_1 \frac{\mu_0 h m_{20} \tau_2 + \mu_0 h m_{10} \tau_1}{[4\beta_1^{(1)} + \tau_1\mu_0 m_{10} h][4\beta_1^{(2)} + \tau_2\mu_0 m_{20} h]} \left. \right] A_1 \\ A_1 &= [1 + l_1 / (4\beta_1^{(1)} + \tau_1\mu_0 h m_{10}) + l_1 / (4\beta_1^{(2)} + \tau_2\mu_0 h m_{20})]^{-1}. \end{aligned} \quad (2.6)$$

We assume that the relations of the theory of polar dielectrics and the Langevin theory of paramagnetism are valid for both kinds of particles (with concentrations n_1 and n_2 and magnetic moments M_{o1} and M_{o2}):

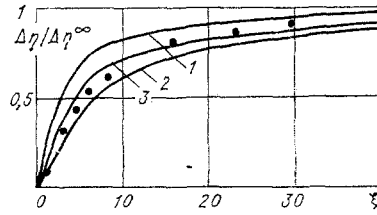


Fig. 1

$$\begin{aligned} \beta_1^{(\alpha)} &= \frac{3}{2} \varphi_\alpha \eta_0, \quad \tau_\alpha = \frac{3\varphi_\alpha \eta_0}{n_\alpha kT}, \quad \xi_1 = \frac{\mu_0 \mu_{01} h}{kT}, \quad \xi = \frac{\mu_0 M_{02} h}{kT}, \quad L(\xi) = \\ &= \text{cth } \xi - \frac{1}{\xi}, \quad m_{10} = n_1 M_{01} L(\xi_1), \quad m_{20} = n_2 M_{02} L(\xi), \quad \varphi = \varphi_1 + \varphi_2. \end{aligned} \quad (2.7)$$

When (2.7) is valid, the correction to the viscosity (2.6) takes the form

$$\begin{aligned} \frac{\Delta \eta}{3\varphi \eta_0 / 2} &= \left[\frac{\varphi_1}{\varphi} D(\xi_1) + \frac{\varphi_2}{\varphi} D(\xi) + \frac{l_1}{\eta_0} \frac{D(\xi)}{3\varphi_1 (2 + \xi_1 L(\xi_1))} + \right. \\ &+ \left. \frac{l_1 D(\xi_1)}{\eta_0 3\varphi_2 (2 + \xi L(\xi))} \right] A, \quad A = \left[1 + \frac{l_1}{\eta_0 3\varphi_1 (2 + \xi_1 L(\xi_1))} + \frac{l_1}{\eta_0 3\varphi_2 (2 + \xi L(\xi))} \right]^{-1}, \\ D(\xi) &= \frac{\xi L(\xi)}{2 + \xi L(\xi)}. \end{aligned} \quad (2.8)$$

The rotational velocities of the particles are given by

$$\begin{aligned} \omega_1 &= \Delta_1^{-1} [9\varphi_1 \varphi_2 \eta_0^2 (2 + \xi L(\xi)) + 3\varphi \eta_0 l_1] v_{,2}, \quad \omega_2 = \Delta_1^{-1} [9\varphi_1 \varphi_2 \eta_0^2 (2 + \\ &+ \xi_1 L(\xi_1)) + 3\varphi \eta_0 l_1] v_{,2}, \quad \Delta_1 = -9\varphi_1 \varphi_2 \eta_0^2 (2 + \xi_1 L(\xi_1)) (2 + \xi L(\xi)) - \\ &- l_1 3\eta_0 [\varphi_1 (2 + \xi_1 L(\xi_1)) + \varphi_2 (2 + \xi L(\xi))]. \end{aligned} \quad (2.9)$$

When the magnetic moments of the first and second groups of particles are the same, i.e., $\xi = \xi_1$, the viscosity correction (2.8) reduces to that in the single-particle rigid dipole model: $\Delta \eta = (3/2)\varphi \eta_0 D(\xi)$ for all values of l_1 . Hence, the rotational diffusion friction l_1 does not contribute to the viscosity correction in this case. This follows from the equality of the rotational velocities of the particles in this approximation, as seen directly from (2.9).

It is clear from (2.8) that if the parameter l_1 , which has dimensions of a viscosity, does not depend on ξ and ξ_1 , then in the limit $\xi_1, \xi \rightarrow \infty$ we obtain for the saturation viscosity the value in the rigid dipole model, $3\varphi \eta_0 / 2$. It follows from (2.9) that the rotational motion of the particles is completely frozen. For $\xi_1 = \xi = 0$ on the other hand, the particles rotate freely with the circulation velocity of the fluid solvent $\omega_\alpha^3 = -(1/2)v_{,2}$.

The dependence of $\Delta \eta / 3\varphi \eta_0 / 2$ on ξ calculated from (2.8) is shown in Fig. 1. Curves are shown for the values $\varphi = 0.19$, $\varphi_1 = 0.1235$, $\varphi_2 = 0.0665$, $\xi_1 = 0.3\xi$. Curve 1 refers to the rigid dipole model for a ferrosuspension with particles of a single kind. Curves 2 and 3 are constructed for $l_1/\eta_0 = 0$ and $l_1/\eta_0 = 1$, respectively.

Comparison of curves 1 and 2 shows that dispersion in particle sizes (or particle magnetic moments) leads to a decrease in the viscosity correction $\Delta \eta$ in comparison to the single-particle rigid dipole model. This is in qualitative agreement with the data [8], shown as points in Fig. 1. In [8], the viscosity correction was studied for angular Couette flow for a suspension of magnetite in kerosene (stabilized by oleic acid) in a transverse field. The hydrodynamic concentration of magnetite was 0.19. According to [9], in a ferrofluid of this kind the fraction of particles undergoing Néel relaxation is 0.65. In addition, the magnetic moment of the colloidal particles is 3.64 times larger than that for the separate ferromagnetic particles [7]. This suggests that in the calculation we assume that the fraction of particles with smaller diameter is 0.65 and $M_{01} \approx 0.3M_0$ and use the polydispersion of the ferrosuspension to interpret the data of [8].

The above analysis shows that the experimentally observed decrease of the viscosity of the ferrosuspension in comparison to the theoretical value for the single-particle model, along with the idea of a partial freezing of the magnetic moments in the particles [8] can

be partially explained by dispersion in the sizes of the particles and their magnetic moments. Also an increase in the parameter l_1 lessens the effect of lowering the viscosity (see curve 3 of Fig. 1), i.e., rotational diffusion friction leads to an additional increase in the flow viscosity.

2.3. Effect of Néel Superparamagnetism of Particles of the First Kind. We assume that the magnetic moments of particles of the first kind can diffuse relative to the particles with a relaxation time $\tau_1 \ll \tau_2$, where τ_2 is the Brownian relaxation time for particles of the second kind. Since we can then put $\tau_1 = 0$, this means a total freezing of the orientational degrees of freedom of the magnetic moments for particles of the first kind. The limit $\tau_1 \rightarrow 0$ also implies that we ignore dissipation of energy due to the relaxation of the magnetization m_1 . Then from (2.6), the assumption (2.7), and $\tau_1 = 0$, we find for the viscosity correction

$$\Delta\eta = \frac{3}{2}\varphi_2\eta_0 \frac{\xi L(\xi)}{2 + \xi L(\xi)} \left(1 + \frac{\varphi_1 l_1}{6\varphi_1\varphi_2\eta_0}\right) \left(1 + \frac{l_1}{6\varphi_1\eta_0} + \frac{l_1}{3\varphi_2\eta_0(2 + \xi L(\xi))}\right)^{-1}. \quad (2.10)$$

The rotational velocities of the particles can be written in the form

$$\omega_1^3 = \Delta_1^{-1} [9\varphi_1\varphi_2\eta_0^2(2 + \xi L(\xi)) + 3\varphi\eta_0 l_1] v_{,2}, \quad \omega_2^3 = \Delta_1^{-1} [18\varphi_1\varphi_2\eta_0^2 + 3\varphi\eta_0 l_1] v_{,2}, \quad \Delta_1 = -18\varphi_1\varphi_2\eta_0^2(2 + \xi L(\xi)) - 3\eta_0 l_1(2\varphi + \varphi_2 \xi L(\xi)). \quad (2.11)$$

Taking the limit $\xi \rightarrow \infty$ we obtain from (2.10) the limiting value of the saturation viscosity:

$$\Delta\eta^\infty/\eta_0 = \frac{3}{2}\varphi_2 \frac{6\varphi_1\varphi_2\eta_0 + \varphi_1 l_1}{6\varphi_1\varphi_2\eta_0 + \varphi_2 l_1}. \quad (2.12)$$

It follows from (2.12) that $\Delta\eta^\infty/\eta_0 < 3/2 \varphi$ where the right-hand side is the viscosity in the rigid dipole model. This is in qualitative agreement with the data of [7] where for a suspension of magnetite in kerosene with concentration $\varphi = 0.24$, the value $\Delta\eta^\infty/\eta_0 \approx 0.06$ is found for the saturation viscosity. This is significantly smaller than the theoretical value of $3\varphi/2 = 0.36$ for the rigid dipole model. According to estimates made in [9], for this kind of ferrosuspension the fraction of particles undergoing Néel relaxation is 0.65, which gives $\varphi_1 = 0.156$, $\varphi_2 = 0.084$. Calculation of the viscosity from (2.12) for the minimum value $l_1 = 0$ leads to the value $\Delta\eta^\infty/\eta_0 = 3\varphi_2/2 = 0.126$ which qualitatively corresponds to experiment, but quantitatively exceeds the experimental value of 0.06. This discrepancy can be explained by the fact that in a real ferrosuspension, particles of the second kind have only partially frozen magnetic moments, and this is not account for in the derivation of (2.12).

It is of interest to consider the effect of increasing field strength and parameter l_1 on the rotation of particles. From (2.11) with $\xi = 0$ it follows that the particles rotate with the circulation velocity of the solvent $\omega_2^3 = -(1/2)v_{,2}$, and the viscosity correction $\Delta\eta = 0$ vanishes. For $\xi \rightarrow \infty$ rotation of particles of the second kind is completely frozen ($\omega_2^3 = 0$) while particles of the first kind rotate with velocity

$$\omega_1^3 = \frac{-3\varphi_1\eta_0}{6\varphi_1\eta_0 + l_1} v_{,2},$$

which is less than the circulation velocity of the solvent when $l_1 \neq 0$. The rotational damping of particles of the first kind is due to diffusive interaction with particles of the second kind which leads to an increase in the flow viscosity. When $l_1/\eta_0 \rightarrow \infty$ particles of the first kind are also completely frozen and the saturation viscosity (2.12) is a maximum and equal to the rigid dipole value of $\Delta\eta^\infty/\eta_0 = 3\varphi/2$. Thus with a decrease in the diffusive rotational friction, agreement between theory and experiment is improved. The dependence of the viscosity ratio $\Delta\eta/\Delta\eta^\infty$ on ξ for $l_1/\eta_0 = 1$ is close to curve 3 in Fig. 1. Hence the effect of lowering the viscosity with respect to the rigid dipole model also occurs when (2.10) is used.

3. Basic Equations for the Single-Particle Model of an Anisotropic Ferrocolloid. We assume that the difference in the rotational velocities of the particles vanishes, i.e., $\omega = \omega_1 = \omega_2$. Then (1.2) for $e = 0$ takes the form

$$\begin{aligned}
\rho \cdot + \rho \nabla \cdot \mathbf{v} &= 0, \quad \rho \mathbf{v} \cdot = \nabla \cdot \mathbf{t} + (\mu_0 \mathbf{m} \cdot \nabla) \mathbf{h}, \quad \rho J \omega \cdot = \nabla \cdot \mathbf{s} + \mathbf{t} \times \cdot \mathbf{I} + \mu_0 \mathbf{m} \times \mathbf{h}, \\
\rho T \gamma &= -T^{-1} \mathbf{Q} \cdot \nabla T + D \mathbf{t}^s \cdot (\nabla \mathbf{v})^s + \mathbf{s} \cdot \nabla \omega + \frac{1}{2} (\mathbf{t} \times \cdot \mathbf{I}) \cdot (\nabla \times \mathbf{v} - 2\omega) + \\
&+ \omega \cdot \mathbf{R} + \rho \mu_0 (\mu_1 \dot{\cdot} - \omega \times \mu_1) \cdot D \eta_1 + \rho \mu_0 (\mu_2 \dot{\cdot} - \omega \times \mu_2) \cdot D \eta_2 \geq 0, \quad \mathbf{m} = \mathbf{m}_1 + \mathbf{m}_2, \\
\mathbf{s} &= \mathbf{s}_1 + \mathbf{s}_2, \quad \rho = \rho_1 + \rho_2, \quad \mathbf{R} = \varepsilon \eta_1 \times \mu_0 \mathbf{m}_1 + \varepsilon \eta_2 \times \mu_0 \mathbf{m}_2, \quad a \cdot \rho = \partial(a\rho)/\partial t + \nabla \cdot (\mathbf{v}\rho a), \quad \rho \mu_\alpha = \mathbf{m}_\alpha.
\end{aligned} \tag{3.1}$$

The restriction $\mathbf{R} \equiv 0$ resulting from the entropy production inequality (3.1), unlike the restriction $\mathbf{R}_\alpha = 0$ from (1.3), gives the free energy φ in the form

$$\varphi = \varphi_0(\rho^{-1}, T) + \frac{\mu_0 m_1^2}{2\rho K_1} + \frac{\mu_0 m_2^2}{2\rho K_2} + \frac{\mu_0 \mathbf{m}_1 \cdot \mathbf{m}_2}{\rho K_3}, \quad K_1, K_2 > 0, \quad \Delta_0 = K_3^2 - K_1 K_2 > 0, \tag{3.2}$$

where terms involving the constant K_3 can be interpreted as a dipole-dipole interaction. From (3.2), (3.1), and (1.3) we find the equation of state

$$\pi = -\frac{\partial \varphi_0}{\partial \rho^{-1}} + \rho(\varphi_0 - \varphi), \quad \varepsilon \eta_1 = \frac{\mathbf{m}_1}{K_1} + \frac{\mathbf{m}_2}{K_3}, \quad \varepsilon \eta_2 = \frac{\mathbf{m}_2}{K_2} + \frac{\mathbf{m}_1}{K_3}. \tag{3.3}$$

The dissipative parts of the constitutive equations, in the simplest case of an incompressible, isothermal fluid, are given by

$$\begin{aligned}
t^{ik} &= -p\delta^{ik} + \alpha_2(v_{i,k} + v_{k,i}) + \varepsilon^{ihsb^s}, \quad \mathbf{b} = \alpha_3(\nabla \times \mathbf{v} - 2\omega), \quad \mathbf{s} \equiv 0, \\
\rho \mu_1 \dot{\cdot} - \omega \times \mathbf{m}_1 &= \frac{1}{\tau_1} \left[m_{10} \dot{\mathbf{h}} - \mathbf{m}_1 - \frac{K_1}{K_3} \mathbf{m}_2 \right] + \frac{1}{\tau} \left[m_{20} \dot{\mathbf{h}} - \mathbf{m}_2 - \frac{K_2}{K_3} \mathbf{m}_1 \right], \\
\rho \mu_2 \dot{\cdot} - \omega \times \mathbf{m}_2 &= \frac{1}{\tau_2} \left[m_{20} \dot{\mathbf{h}} - \mathbf{m}_2 - \frac{K_2}{K_3} \mathbf{m}_1 \right] + \frac{1}{\tau} \frac{K_2}{K_1} \left[m_{10} \dot{\mathbf{h}} - \mathbf{m}_1 - \frac{K_1}{K_3} \mathbf{m}_2 \right].
\end{aligned} \tag{3.4}$$

The equations (3.1) to (3.4) along with (1.1) are a closed set of equations for \mathbf{v} , ω , \mathbf{m}_1 , \mathbf{m}_2 , \mathbf{h} .

4. Effective Viscosity for the Single-Particle Model of an Anisotropic Ferrocolloid.

Since Eqs. (3.1)-(3.4), unlike the model of [2], have two magnetization relaxation mechanisms, they can describe the effect of partial freezing of the magnetic moments with respect to the particles on the effective viscosity of the ferrosuspension. Following the analysis of Sec. 2 above, we find the value $\mathbf{m}_\alpha = \chi_\alpha \mathbf{h} + \mathbf{m}_{\alpha \perp}$, where

$$\begin{aligned}
\chi_\alpha &= K_\alpha K_3 (K_3 - K_{\alpha+(-1)^{\alpha-1}}) / \Delta_0; \quad \Delta_1 = \Delta_0 / \tau_1 \tau_2 K_3^2 - K_2 / \tau^2 K_1; \quad \mathbf{m}_\perp = \mathbf{m}_{1\perp} + \\
&+ \mathbf{m}_{2\perp}; \quad \mathbf{m}_\perp = -B_1 \omega^3 \mathbf{h}; \quad B_1 = \Delta_1^{-1} \{ [\tau(K_3 - K_1) - \tau_1(K_3 - K_2)] \chi_2 / K_3 \tau_1 + [\tau K_1(K_3 - K_2) - \tau_2 K_2(K_3 - K_1)] \chi_1 / K_1 K_3 \tau \tau_2 \},
\end{aligned}$$

which gives the viscosity correction

$$\begin{aligned}
\Delta \eta_j &= \frac{\alpha_3 B_j \mu_0 \dot{\mathbf{h}}^2}{4\alpha_3 + B_j \mu_0 \dot{\mathbf{h}}^2}, \quad B_2 = K_3 [\chi_2 \tau_2 (K_3 - K_1) + \chi_1 \tau_1 (K_3 - K_2)] / \Delta_0, \\
B_3 &= \tau_2 K_3^2 (K_3 - K_1) / \Delta_0^2, \quad B_4 = K_2 \tau_2
\end{aligned} \tag{4.1}$$

The viscosity correction $\Delta \eta_1$ depends on the relaxation times τ_1 , τ_2 , τ , the rotational viscosity α_3 and the parameters K_1 , K_2 , K_3 .

The values $j = 2$ and $j = 3$ correspond to $\tau \rightarrow \infty$ and $\tau \rightarrow \infty$, $\tau_1 = 0$. For $j = 4$ we take in addition $K_3 \rightarrow \infty$. Because $\mathbf{m}_0 = \chi \mathbf{h}$ and $K_2 = \chi - K_1$, we can transform $\Delta \eta_4$ from (4.1) to the form $\Delta \eta_4 = \alpha_3 M / (\alpha_3 4 + M)$, $M = \mu_0 m_0 \dot{\mathbf{h}} \tau_2 (\chi - K_1) / \chi$, which is the value obtained in [8] under the assumption of partial equilibrium, when the magnetization is directed along the total field $\mathbf{h} + \mathbf{A}$ where \mathbf{A} is an internal anisotropy field.

Our analysis shows that in the description of partial freezing of magnetic moments, it is in general not necessary to introduce a hypothetical internal anisotropy field as was done in [8]. The assumption of two magnetization relaxation mechanisms is sufficient, and

is consistent with the physics of the magnetization in a ferrosuspension. For an applied field the magnetization of the ferrosuspension in the direction of the field instantly (for $\tau_1 = 0$) takes the value m_{10} because of Néel relaxation. But over a time of order τ_2 the magnetization of the ferrosuspension increases to the value m_{20} because of Brownian relaxation. In the equations given here, terms involving the coefficient K_3 in the equation of state (3.3) correspond to the effect of internal fields on the magnetization m_1 and m_2 due to the existence of magnetizations m_1 and m_2 , respectively. This recalls the situation in antiferromagnets where there are also two effective fields resulting from the magnetizations m_1 and m_2 of the sublattices.

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EVOLUTION EQUATION FOR THE VORTEX DISTRIBUTION FUNCTION IN THE PLANAR CASE

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In [1-5] a system of linear vortex lines in an ideal fluid is used as a model of two-dimensional turbulence. There are examples which support the plausibility of this model; in particular, results have been obtained with the model which are interesting from the point of view of the statistical theory of turbulence [1], dynamical meteorology [4], and numerical modeling of the streamlining of bodies at large Reynolds numbers [5].

In the above papers the system of vortices was studied in a state of statistical equilibrium. But in real hydrodynamic turbulence, nonequilibrium states of the fluid are important as well, where the evolution is characterized by statistical irreversibility. It is therefore of interest to consider nonequilibrium evolution in model systems by the methods of kinetic theory [7, 8]. Some asymptotic solutions of the BBGKY hierarchy for a system of linear vortices have been considered in [1].

In the present paper, the nonequilibrium statistical properties of this model are studied using the Liouville equation for an ensemble of vortex lines. Analysis and summation of the formal time-dependent perturbation series are carried out with the help of the dia-

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