

# HYDRODYNAMIC BEHAVIOR OF SUSPENSIONS OF POLAR PARTICLES

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## INTRODUCTION

Colloidal suspensions are one of a great variety of fluids that are called "complex fluids" (1–4). This name implies that the fluid has internal structure or microstructure and behaves very different from water or air, for example, when different forces are exerted on it. The difference has been shown experimentally in many conditions, in particular, for example, by means of a fundamental physical phenomenon like that of Rayleigh-Bénard instability (natural convection) due to buoyancy when the suspension is heated from below (5). It is also important to take that difference into account in industrial applications like, for example, when the suspended particles undergo phase transformations in solidification processes where natural convection controls the freezing and melting rate of the material (6). The "particles" in a colloidal suspension have a size of the order of 10 to 100 nm and may be polymers, emulsified drops, vesicles, or ink, paint, and smoke particles. They are so small that they may interact with other dissolved substances in the solvent. They are in constant motion due to thermal fluctuations, behavior called Brownian motion. The interactions are due to different forces like van der Waals and electrostatic. The macroscopic behavior of the suspension is then a consequence of a statistical average of all the fluctuations and interactions. An important consequence of the interaction of the particles among themselves and with other substances is the so-called self-organization (1–3, 7) and aggregation (flocculation) (8). It is the spontaneous ability to form regular structures when the concentration of particles is above a certain magnitude. The self-organization can be disrupted when some shear is applied to the suspension, giving rise to a relation between rheology (the science of flow) and structure. On the contrary, there are suspension systems able to form structures when shear is applied. By these means, it is possible to change the ratio between the applied stress and the shear rate, usually

called viscosity. The shear-thickening (that is, the increase of viscosity) of a suspension is a result of an internal structural change due to the application of a shear rate (9, 10). The flow of a colloidal suspension not only depends on the interaction forces but also on the size distribution of the particles and their geometry, among other things (10). Therefore, from the statistical mechanics point of view, the simplest model available is that which supposes the particles as hard spheres of the same size. Here, the interaction between particles is extremely repulsive and only occurs when they touch. The difficulty of the model may be increased considering soft particles of different kinds (1, 2), etc. The particle geometry has an important role like in fiber suspensions or polymeric solutions where the polymer chains are very rigid (10). They are modeled by rigid rods, which can be handled by statistical mechanics (1). It is interesting that these suspensions may show a natural orientational order, which can be modified (for applications) if the particles are susceptible to an external force field or to shear flow. When the particles deform under stationary or time-dependent shear flow, other phenomena are found like in red cells suspensions. Suspensions like these are simulated experimentally by means of artificial capsules (11).

The concentration of a colloidal suspension ranges from that of a dilute suspension to that of very high concentration as in the case of mud (suspensions of clay in water) (12, 13). For high concentrations, it could be necessary to apply a very high stress, to overcome the yield stress due to the close packing of the particles, before the suspension starts to flow.

The stability of a colloidal suspension is also an important issue in applications. If it is necessary to avoid sedimentation, then the suspension stability may be controlled by means of electrostatic interactions, for example (8). In particular, in dipolar electric and magnetic colloidal suspensions under strong fields, it is of importance to prevent sedimentation or coagulation to ensure stability (14).

Chemically and electrically stabilized colloids are widely used in the chemical industry and have been the subject of scientific research (15–17). When sedimentation is allowed or needed, like in the chemical and petroleum industries for particle separation, flocculation or aggregation plays an important role under gravity and centrifugation (18–21). To destabilize a colloidal suspension, it is not enough to apply an external force field. The force exerted on the particle has to exceed that of the hydrodynamic forces that depend on the viscosity of the carrying fluid, the velocity, and the size of the particle itself (22).

There are other ways to make a suspension inhomogeneous. One of them is the effect of shear flow on the migration of particles or polymers (23–25). However, there are methods developed to make a suspension or resuspend the particles. Leighton and Acrivos (26) investigated the

process of viscose resuspension by means of a shear flow. Chemical engineers have proposed different methods for making particle suspensions, like the new one by Taca and Paunescu (27) of spherical stirred vessels.

In the following sections, it will be supposed that the particles are hard spheres of the same size and that the suspension is homogeneous even though external force fields are applied to it. Therefore, a two-phase system (28, 29) is considered, which is composed of a Newtonian or a non-Newtonian fluid (30) (the carrying fluid) and a solid phase made of dispersed particles. The hard spheres are assumed to have rotation (intrinsic rotation or spin) and to be susceptible to polarization under the application of an external field. The suspensions whose particles have a spin interacting with the solvent hydrodynamic vorticity (rotation rate of a liquid elementary part) are called mi-

**Table 1** Suspension viscosity as a function of particles volume fraction and polar effect corrections

Colloidal dispersion	Hydrodynamic effective viscosity	Remarks
Diluted $0 < \phi < 0.01$	$\eta_E = \eta_0(1 + 2.5\phi)$	Einstein's formula $\eta_0$ : Steady shear viscosity of the solvent
Semidiluted $0.01 < \phi < 0.10$	$\eta = \eta_0(1 + 2.5\phi + 5.2\phi^2)$	Hydrodynamic contributions are considered in the second order term
Semidiluted and Brownian motion effect	$\eta = \eta_0(1 + 2.5\phi + (5.2 + 0.97)\phi^2)$	Second order corrections come from the Brownian motion contribution
Concentrated $\phi > 0.10$	$\eta = \eta_0 \frac{1 + 3A/2}{1 - A}$	$A = \phi[1 + S(\phi)]$ $S(\phi) = 0.87\phi - 1.15\phi^2$
Concentrated with interparticle interactions $0.10 < \phi < \phi_m$	$\eta = \eta_0 \left(1 - \frac{\phi}{\phi_m}\right)^{-2}$	Phase transition-type description for a particular $\phi_m$ $\phi_m$ : maximum packing fraction
Brenner's diluted dispersions	$\eta = \eta_E + \frac{3\phi}{2} \eta_0 [1 - (1 - \lambda^2)]^{\frac{1}{2}} F(\lambda)$	$F(\lambda) = \left(\frac{1}{\lambda^2} - 1\right) \operatorname{arctanh}(\lambda)$ $\lambda = \frac{\rho g d}{6\eta_0 \omega_0}$ , $\omega_0$ : vorticity before particles dispersion is mixed in the solvent
Diluted polar Electric particles	$\eta = \eta_E + \frac{\chi_0}{16\pi} \tau_D (\bar{E} \cdot \bar{k})^2$	Electroviscosity Effect
Diluted polar Magnetic particles	$\eta = \eta_E + \frac{1}{4} M_0 H \tau_M q^{-1}$	Magnetoviscosity effect Shliomis' Correction $q = \left(1 + \tau_M \frac{M_0 H}{I}\right)$

cropolar. The suspensions with polarizable particles (dielectric or magnetic particles or polymers, for example) are called polar. The micropolar suspension model is satisfied by a suspension if the spin viscosity is important enough. The spin viscosity, as will be seen, is a result of the friction between particles and the carrying fluid elements rotating at different velocity. Besides, the polar suspension model is satisfied by a suspension whose particles polarize under the application of an external force field. For example, when the particles or polymers show an electric charge displacement while an electric field is imposed on the suspension, they rotate, generating spin, in order to align in the direction of the external field. Due to fluid friction, the rotation is delayed in comparison to a particle in free space. With the action of the same field, a pressure gradient is created on the suspension producing flow (31). Therefore, spin and polarization have an intimate relation in the rheology of polar suspensions. The same effect appears in magnetic suspensions (32) and, in particular, in mass-dipolar suspensions under gravity where the particles have the center of mass displaced a distance from the geometric center (33) (see Brenner's diluted dispersion in Table 1). If the suspension particles are not susceptible to the applied particular external force field but the spin is still important, then the suspension satisfies the micropolar model under shear flow. Note that the external field does not necessarily have to produce a bulk motion on the polar suspension to present an interaction between polarization and spin. It is possible to produce particle spin in a polar fluid filling a closed vessel by the application of an oscillatory external field. This case is relevant because, in this way, it is feasible to characterize the response of the suspension to the applied field in the absence of bulk flow effects. However, bulk motion can still be produced in a closed vessel if the suspension is susceptible to an external magnetic or electric field that is under rotation or if the vessel rotates in a stationary external field. It is important to point out here that there is a confusion in the scientific literature where the terms micropolar and polar (dipolar) are used as synonyms. Note that they are different concepts and that their difference has already been made clear in the definitions given above.

Summing up, it is of interest to understand the equilibrium and dynamical properties of colloidal suspensions as well as to describe their transport and response coefficients that are necessary for their research and application. In particular, the study of the formation and behavior of colloidal suspensions under different conditions, such as shear flow, and the influence of external fields are of scientific and practical interest. In general, colloidal suspensions are subject to a variety of effects that act on

each individual particle such as hydrodynamic interactions, stress relaxation, Brownian fluctuations of particle velocity, internal rotation or spin, polarization diffusion, inertial and vorticity effects, etc. Different areas of physics contribute to the understanding of colloidal suspensions like continuum mechanics, electrostatics, nonequilibrium statistical mechanics, and irreversible thermodynamics. In what follows, the discussion of colloidal suspensions will develop in steps that increase the difficulty of the physical description of the model.

## HYDRODYNAMICS OF SUSPENSIONS OF NONPOLAR PARTICLES

The diffusion transport of hard sphere particles without long-range interactions is described by the following set of equations

$$\frac{\partial \phi}{\partial t} = -\nabla \cdot \bar{J} \quad (1)$$

$$\bar{J} = -D\nabla\phi \quad (2)$$

where  $\phi$ ,  $\bar{J}$ , and  $D$  are the volume fraction of suspended particles, the particles' flux, and the translational diffusion coefficient, respectively. The translational diffusion process is produced by the Brownian motion, which is independent of the particles' rotation. However, the translational effect of the flow modifies the particles flux by adding a convective term, that is  $\bar{J} = -D\nabla\phi + \phi\bar{u}$ . This is the base of the so-called constitutive convective-diffusion equation. Supposing that  $\bar{u} = 0$  and substituting Eq. 2 into Eq. 1, the Fick's second diffusion law is obtained, that is

$$\frac{\partial \phi}{\partial t} = D\nabla^2\phi \quad (3)$$

Here,  $D$  is assumed to be a constant in space. It is of interest to look for a solution of this equation in one dimension for a large number of noninteracting particles located initially at the origin. The solution is

$$\phi(x, t) = \frac{1}{2(\pi Dt)^{\frac{1}{2}}} \exp\left\{-\frac{x^2}{4Dt}\right\} \quad (4)$$

For the convective-diffusion case, the result given above still holds, replacing  $x$  with  $x - ut$ . After the diffusion process is over, the solid particles are considered isotropically dispersed with a final concentration that may be high, low, or intermediate. The dynamics of a New-

tonian fluid with suspended particles are described by the so-called two-phase flow with the Navier-Stokes equation

$$\rho \frac{d\bar{u}}{dt} = -\nabla p + \nabla \cdot Q^s \quad (5)$$

where

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \bar{u} \cdot \nabla$$

is the Lagrangian time derivative of hydrodynamics, and the total viscous stress tensor is given by  $Q^s = \tau^v + \Gamma = 2\eta_E(\nabla\bar{u})^s$ . Besides,  $\tau^v$  and  $\Gamma$  are the viscous stress tensor of the background fluid and the induced pressure tensor due to the presence of particles, respectively (34, 35). The constant  $\eta_E$  is the effective viscosity of the suspension. In particular, for low concentrations, the Einstein's formula is very useful (36, 37), along with other effective shear viscosities for semidiluted (38) and concentrated suspensions (39–41), which are given in Table 1.

The rheological behavior of concentrated suspensions of hard spherical particles has been the subject of extensive research. Several conclusions about their behavior have been reported. For example:

1. The distribution of particles is distorted by the presence of strain. The fluid and the solid phase give rise to separate mechanical stress contributions.
2. The Brownian motion modifies Einstein's formula for semidiluted and concentrated suspensions (see Table 1).
3. The linear relaxation of the viscous stress tensor has been confirmed experimentally for low deformation rate flow [for Maxwell's, Voigt's, or Oldroyd's model (42)].
4. A nonlinear relaxation modulus for a large deformation rate flow is reported for concentrated hard spheres. This effect is related to patterns of clusters' rupture or formation when increasing the shear stress (43–45). Order-disorder transition was reported in colloidal suspensions (46).

## INTERNAL PROCESSES IN MICROPOLAR FLUIDS AND SUSPENSIONS

A micropolar fluid is composed of molecules with intrinsic rotation. Similarly, a suspension is called micropolar when its colloidal particles have spin. Both cases will be treated under the same continuum formalism in which the linear momentum equation must be supplemented with a new

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variable, that is, the local average of internal rotations or the spin of the particles, respectively (47).

### Angular Velocity Distribution in Colloidal Suspensions

For spherical and nonspherical particles, it is possible to define a director vector along one of the symmetry axes. In this way, the rotation of the particles is described by means of the Euler angles in a stationary frame of reference in space. The angular velocity distribution for noninteracting particles is described by means of the Gibbs-Boltzmann statistics (48). Therefore, the distribution function for thermal equilibrium at the temperature  $T$  is given by

$$f(\bar{\omega}) = \left( \frac{I}{2\pi k_B T} \right)^{\frac{1}{2}} \exp \left\{ -\frac{I\bar{\omega}^2}{2k_B T} \right\} \quad (6)$$

where  $I$  is the moment of inertia of the dispersed particles,  $\bar{\omega}$  is the angular velocity defined around a perpendicular axis of the director vector of the particle. Eq. 6 is the analog of the Maxwellian distribution function of a particle-velocity component. This distribution is a Gaussian centered at  $\bar{\omega} = 0$ . Considering that the particles have two angular degrees of freedom and according to the equipartition principle, the average in the angular velocity is given by the equality

$$k_B T = \frac{1}{2} I \bar{\omega}_{av}^2.$$

Therefore, from the statistical mechanics point of view, the dispersed particles have rotational motion according to the temperature of the suspension. The average angular velocity is given by

$$\omega_{av} = \left( \frac{2k_B T}{I} \right)^{\frac{1}{2}} \quad (7)$$

Besides, the results of Eqs. 6 and 7 are fulfilled by particles suspended in a diluted gas under equilibrium conditions. For suspended particles in dense gases or liquids in motion, the friction between the particle and the fluid has to be taken into account. This friction effect produces a torque on the particle and thus an angular acceleration that modifies the thermal equilibrium distribution of angular velocities, as will be seen in Eq. 11.

### Rotational-Fick's Diffusion

The angular position  $\theta$  is the angle of the director vector in relation to a fixed axis and its temporal variation is

$$\omega = \frac{d\theta}{dt}.$$

In a nonequilibrium situation, the rotational-Fick's diffusion takes place, and the angular distribution function  $f(\theta)$  satisfies the following equation:

$$\frac{df(\theta, t)}{dt} + \nabla_{\theta} \cdot \bar{J}_f = 0 \quad (8)$$

where  $\bar{J}_f = -D_r \nabla_{\theta} f + f \bar{\omega}_{av}$ ,  $f \bar{\omega}_{av}$  is the rotational convective flow,  $D_r$  is the rotational diffusion coefficient, and  $\nabla_{\theta}$  is defined as

$$\nabla_{\theta} f = \hat{e}_{\theta} \frac{1}{r} \frac{\partial f}{\partial \theta}$$

Considering diffusion in one dimension, the solution of Eq. 8 has the form

$$f(\theta, t) = \frac{1}{2(\pi D_r t)^{1/2}} \exp\left\{-\frac{(\theta - \omega_{av} t)^2}{4D_r t}\right\} \quad (9)$$

The rotational-diffusion constant may be obtained by means of the angular root-mean-square displacement in a long time period  $\Delta t$

$$D_r = \frac{\langle \theta^2 \rangle}{2\Delta t} \quad (10)$$

### Rotational Friction on Colloidal Particles Due to Vorticity in the Flow Field

Spherical and nonspherical particles isotropically dispersed do not rotate freely according to the average angular velocity, as explained above. They rotate under an external couple caused by the friction between the particles and the fluid. In this case, the angular momentum balance equation and the linear momentum balance equation have to be taken into account. The former is

$$I \frac{d\bar{\omega}}{dt} = -2\zeta(\bar{\omega} - \bar{\Omega}) \quad (11)$$

where

$$\bar{\Omega} = \frac{1}{2} \nabla \times \bar{u}$$

is the vorticity of the surrounding fluid and  $\zeta$  is the vortex viscosity. It is clear that the friction between the particle and the liquid brings about the relaxation mechanism, which leads to the internal mechanical equilibrium of the suspension. The equilibrium is attained when the particle rotation rate follows the vorticity of the fluid flow, that is, when  $\bar{\omega} = \bar{\Omega}$ .

In case the flow velocity is very high, the effects of inertia produce a noninstantaneous coupling between par-

ticle internal rotation and local vorticity (49). Assuming that the relaxation of internal rotation or spin occurs in a time scale shorter than the hydrodynamic one, Eq. 5 decouples from Eq. 11, and the velocity field can be solved independently of the rotation dynamics but according to the boundary conditions. However, the spin still feels the effects of the flow field. Therefore, supposing  $\bar{\Omega}$  as constant in the rotation time scale and  $\omega(t=0) = \omega_{av}$ , the spin evolution can be given by the following expression

$$\bar{\omega}(t) = \bar{\omega}_{av} \exp\left(-\frac{t}{\tau_s}\right) + \left[1 - \exp\left(-\frac{t}{\tau_s}\right)\right] \bar{\Omega}(t) \quad (12)$$

where  $\tau_s$  is the spin-relaxation time given by

$$\tau_s = \frac{I}{2\zeta} = \frac{\rho_s d^2}{30\eta_E} \frac{v}{\phi} \quad (13)$$

The identity in Eq. 13 follows from the relations (50)

$$\zeta = \frac{3}{2} \eta_E \phi$$

and

$$I = \frac{2}{5} \rho_s v d^2,$$

where  $v$  is the particle volume,  $\rho_s$  is the density of the particle, and  $d$  is its characteristic length.  $\tau_s$  is of the order of 10 ps for particles of 0.01  $\mu\text{m}$ . If the particles are of 1  $\mu\text{m}$ ,  $\tau_s$  increases four orders of magnitude, giving the possibility of a coupling between the particle spin and the velocity field dynamics. In this case, the balance of momentum, Eq. 5, is modified by the inclusion of a new term arising from the antisymmetric contribution of the viscous stress tensor  $\mathbf{Q}^a$  (51), namely

$$\rho \frac{d\bar{u}}{dt} = -\nabla p + \nabla \cdot \mathbf{Q}^s + \nabla \cdot \mathbf{Q}^a \quad (14)$$

where

$$\mathbf{Q}^s = 2\eta_E (\nabla \bar{u})^s, \quad \mathbf{Q}^a = 2\zeta \underline{\underline{\varepsilon}} \cdot (\bar{\Omega} - \bar{\omega}) \quad (15)$$

where  $\underline{\underline{\varepsilon}}$  is the three subindexes alternating tensor. substituting Eq. 15 into Eq. 14, the modified linear momentum equation for an incompressible fluid ( $\nabla \cdot \bar{u} = 0$ ) is obtained,

$$\rho \frac{d\bar{u}}{dt} = -\nabla p + \eta_E \nabla^2 \bar{u} - 2\zeta \nabla \times (\bar{\Omega} - \bar{\omega}) \quad (16)$$

This result differs from the usual Navier-Stokes equation of hydrodynamics by the presence of the last term, which is related to the coupling of the spin with the flow via the antisymmetric stress tensor (52).

### Spin Velocity Diffusion

The evolution of the rotation angle of the particles was previously discussed as produced by the mechanism of Brownian motion. Now, the diffusion of the spin velocity is considered. This diffusion is produced by the hydrodynamic interaction via the vorticity induced by the particle rotation.

Until now, it was supposed that inertial effects of particle rotation are important in the description of the spin relaxation mechanism according to Eq. 11. Along the same line, inertial effects are a necessary condition to have diffusion of spin when spatial inhomogeneities are present in the system. In fact, when the spin variable is not uniform, a distribution of spin produces spatial variation in the symmetric part of the spin tensor given by the surface couple-stress tensor  $\mathbf{C} = 2\eta_{sp}(\nabla\bar{\omega})^S$ . This tensor is the analog to the viscous stress tensor (32) and modifies the balance of angular momentum Eq. 11, which changes according to

$$I \frac{d\bar{\omega}}{dt} = -2\zeta(\bar{\omega} - \bar{\Omega}) + \nabla \cdot \mathbf{C} \quad (17)$$

By substituting the surface couple-stress tensor in Eq. 17, it becomes a diffusion equation with a relaxation term, the first at the right hand side, which also works as a spin source by means of  $\bar{\Omega}$ . That is

$$\frac{d\bar{\omega}}{dt} = -\frac{1}{\tau_s}(\bar{\omega} - \bar{\Omega}) + D_{sp}\nabla^2\bar{\omega} \quad (18)$$

where it was supposed that  $\nabla \cdot \bar{\omega} = 0$ , and

$$D_{sp} = \frac{\eta_{sp}}{I}$$

The characteristic time for spin diffusion is

$$\tau_{sp} = \frac{\lambda^2}{4\pi^2 D_{sp}},$$

and the parameter  $\lambda$  is the characteristic length of the spatial inhomogeneities of the distribution of spin (53, 54). Eqs. 16 and 18 have also been derived from the microscopic point of view (55–57). It should be noted that the spin diffusion has mostly a short length scale (10–100 nm)

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(58). In this micro hydrodynamics scale, the spin diffusion results are more important than vorticity friction, and Eq. 18 becomes

$$\frac{d\bar{\omega}}{dt} = D_{sp}\nabla^2\bar{\omega} \quad (19)$$

where the characteristic diffusion time is very short for small particle radius ( $\tau_{sp} = 10^{-12}$  sec). This diffusion effect is very fast from the hydrodynamic point of view (59). However, spin diffusion is important to homogenize the magnitude of the local angular velocity  $\bar{\omega}$  in a volume element. In this way, it is shown that the terms of Eq. 19 have an important physical interpretation in the continuum formalism of micropolar fluids.

### EQUATIONS OF MOTION OF SUSPENSIONS OF POLAR PARTICLES

Several phenomena in electrorheology (ER) and magnetorheology (MR), like electroviscosity and magnetoviscosity, are produced by induced polarization (magnetization) when an external field (electric or magnetic) is applied. These effects are supported by the polarizability properties of the dispersed particles. It is necessary to point out the differences existing between an MR fluid and a magnetic suspension. When a magnetic field is applied to an MR fluid, the behavior changes from that of a fluid to a solid-like one for certain shear stresses. However, when the shear stress is increased above the magnitude of the yield stress, the rheological behavior follows a Bingham model in which case the solid-like behavior is disrupted and the viscosity changes suddenly from a high value to a lower one. On the other hand, in a magnetic suspension, it is supposed that the hydrodynamic character never changes. When a magnetic field is applied, the suspension viscosity increases depending on the square of the magnetic field strength (60) (see Table 1). In order to concentrate on the goals of this review, the ER and MR fluids will be excluded from the discussion, and the description will concentrate on the hydrodynamic behavior of electric dipolar and magnetic dipolar suspensions. The reader interested in ER and MR fluids may consult the following references (61–67). Here, a review of mass-dipolar suspensions will also be included. The dipolar moment property of suspended particles is a consequence of the separation of the geometric center from the center of mass of the particle. In the three cases of the electric, magnetic, and mass-dipolar suspensions, the application of an external (electric, magnetic, or gravitational) field produces a torque on the particle, which works to rotate its dipolar vector to an equilibrium position parallel to the direction of the exter-

nal field. This is a relaxation process whose mechanism is a combination of the rotational friction between the particle and the fluid and the rotational Brownian effect. This process was first described by Debye (68) for a set of electric dipolar molecules dispersed in a neutral fluid. His description was for an ideal system in which the interaction between particles is neglected. Similar assumptions have been made for magnetic (69) and mass-dipolar suspensions (33, 70).

A distinction should be made between a suspension of polar particles and a polar fluid (60). The former one is characterized by a second phase dispersed in the liquid, which is represented by the particle volume fraction  $\phi$ . A problem found in this system is that of agglomeration, which occurs due to the (dielectric or magnetic) interaction between the particles. Besides, a polar fluid is a one-phase system where the polarization vector is a volume average of the molecular dipolar moments. From the point of view of the continuum formalism, both kinds of fluids are described by the same constitutive equations because the parameter  $\phi$  for a suspension of polar particles is included in the transport coefficients.

### Polar Electric-Suspensions with Spin

In order to describe suspensions of polar particles, it is necessary to consider the linear and angular momentum balance equations

$$\rho \frac{D\bar{u}}{Dt} = -\nabla p_t + \nabla \cdot \mathbf{Q}_T^S + \nabla \cdot \mathbf{Q}_T^a \quad (20)$$

$$I \frac{D\bar{\omega}}{Dt} = \varepsilon : \mathbf{Q}_T^a \quad (21)$$

where  $\mathbf{Q}_T^S = \mathbf{Q}^S + \mathbf{T}^S$ ,  $\mathbf{Q}_T^a = \mathbf{Q}^a + \mathbf{T}^a$ ,  $p_t = p + p_r$ , and  $p_r$  is the radiation pressure defined by

$$p_r = \frac{1}{2\epsilon_0} \bar{D}^2.$$

This is also recognized as a dynamic pressure as that of magnetic colloidal dispersions, which takes the form

$$p_r = -\frac{1}{8\pi} \rho H_0^2 \left( \frac{\partial \mu}{\partial \rho} \right)_T$$

where  $H_0$  is the magnitude of the magnetic field and  $\mu$  is the magnetic permeability (71).  $\mathbf{T}^S$  and  $\mathbf{T}^a$  are the symmetric and antisymmetric parts of the electromagnetic-Maxwell stress tensor, respectively. They are defined as

$$\mathbf{T}^S = \frac{1}{2} \bar{E} \bar{D} = \mathbf{T}^0 + p_r \mathbf{I} \quad (22)$$

$$\varepsilon : \mathbf{T}^a = \bar{P} \times \bar{E} \quad (23)$$

where  $\bar{P}$ ,  $\bar{D}$ , and  $\bar{E}$  are the polarization vector, the displacement vector, and the local electric field, respectively.  $\mathbf{T}^S$  is the traceless electromagnetic stress tensor. The right-hand side of Eq. 23 is the torque produced by the electric field on the dipole of the fluid particle. It should be noted that the electromagnetic pressure ( $p_r$ ) does not contribute to the dissipative part of the polarization and magnetization (72), and this part will not appear in the constitutive equations. On the other hand, the explicit form of the electric or magnetic stress tensors depends on the properties of the material (73, 74).

Now, consider suspensions of small dipolar particles for which the rotational inertia is negligible in comparison with the viscous forces (75). In this case, the right-hand side of Eq. 21 is zero. Therefore, the antisymmetric tensors have no contribution to the stresses, and the last term of Eq. 20 is not present. This means that the factors favoring the internal rotation of the fluid do not affect the fluid velocity profile (76, 77).

On the other hand, the torque the electric field produces on the dipolar particles is counterbalanced by the frictional effect with the fluid. Therefore, the mechanisms controlling the evolution of the dipoles, which tend to align in an equilibrium position, are the thermal fluctuations (due to Brownian molecular motion). Thus, this is a relaxation process described by the Debye equation of the polarization vector, that is

$$\tau_D \frac{D\bar{P}}{Dt} = \chi(\bar{E}) \bar{E} - \bar{P} \quad (24)$$

where

$$\chi(\bar{E}) = \chi_0 \left( \coth \xi - \frac{1}{\xi} \right),$$

$$\xi = \frac{\mu_E |\bar{E}|}{kT},$$

and,  $\chi_0$  is the electric susceptibility at zero frequency of oscillation of the electric field. The operator  $D/Dt$  in Eq. 24 is the corrotational or the codeformational time derivative for a vector field as defined in Eqs. 3.2a and 3.2b of Ref. 78. The Debye relaxation time is given by

$$\tau_D = \frac{4\pi d^3 \eta_E}{k_B T} \quad (25)$$

Until now, the polarization relaxation does not contribute to the evolution of the flow field, but the velocity vector field does modify the polarization through

the corrotational time derivative. This is true for dilute dipolar suspensions, but for concentrated ones, the interaction effect becomes important for concentrations that have not reached the agglomeration point. In fact, for concentrated polar suspensions (electric or magnetic ones), inter-particle interactions also induce polarization, modify the distribution of particles, and produce the exchange of angular and linear momentum between particles and fluid. In this way, the effects of the polarization on the symmetric and antisymmetric stress tensors are understood.

In order to introduce these effects, it is supposed that the polarization vector plays a similar role as that of the velocity vector (31). Therefore, the constitutive equations of the viscous stress tensors are given by (78)

$$\mathbf{Q}_T^S + \lambda_2 \frac{D\mathbf{Q}_T^S}{Dt} = 2\eta_E(\nabla\bar{u})^S + 2\delta_3(\nabla\bar{P})^S \quad (26)$$

$$\mathbf{Q}_T^a + \lambda_5 \frac{D\mathbf{Q}_T^a}{Dt} = 2\zeta_\varepsilon : (\bar{\Omega} - \bar{\omega}) + 2\delta_7(\nabla\bar{P})^a \quad (27)$$

Under these assumptions, the  $\mathbf{Q}_T^S$  and  $\mathbf{Q}_T^a$  are the total stress tensors acting in a fluid particle, which also include the electric or magnetic contribution. The coefficients  $\delta_3$  and  $\delta_7$  are interpreted as shear-like viscosity and rotational-like viscosity for polarization, respectively. Besides, Eqs. 26 and 27 also contain the stress relaxation times  $\lambda_2$  and  $\lambda_5$ . In these equations, the retardation times for the delayed response of the coupling between the velocity and the polarization have been ignored for the sake of simplicity (79). Note that here the operator,  $D/Dt$ , is the corrotational or the codeformational (upper or lower convected) time derivative for tensors (80).

Moreover, Eq. 24 may be generalized, assuming that the interactions between dipoles produce a viscous-stress distribution that contributes to the evolution of the polarization vector. This is given by the introduction of coupling terms into the Debye equation

$$\tau_D \frac{D\bar{P}}{Dt} = \chi(\bar{E})\bar{E} - \bar{P} + \delta_2\tau_D \nabla \cdot \mathbf{Q}_T^S + \delta_6\tau_D \nabla \cdot \mathbf{Q}_T^a \quad (28)$$

Now, the situation is that the velocity field can modify the evolution of polarization by means of the velocity gradients that appear in the constitutive equations through the stress tensors (see Eqs. 26 and 27). Besides, the polarization in Eq. 28 modifies the velocity field in Eq. 20 by means of Eqs. 26 and 27. In fact, the same reciprocal process affects the spin in Eq. 21.

## Polar and Magnetic Suspensions with Spin

The evolution equation for the magnetization per unit volume is given by

$$\frac{D\bar{M}}{Dt} = -(\bar{M} - M_0\hat{e}_H) \quad (29)$$

where  $M_0$  is the equilibrium magnetization given by the magnetic equation of state

$$M_0 = \phi M_d \left( \coth \alpha - \frac{1}{\alpha} \right) \quad (30)$$

$$\alpha = \frac{\pi\mu_0 M_d H d^3}{k_B T} \quad (31)$$

where  $\mu_0$  is the permeability of free space,  $M_d$  is the saturation value of the magnetization, and  $\hat{e}_H = \bar{H}/H$  is a unitary vector in the magnetic field direction.

In the presence of spatial inhomogeneities of the magnetization, the term  $\eta_M \nabla^2 \bar{M}$  has to be added to the right-hand side of Eq. 29 (81).

In flow with vorticity ( $\bar{\Omega} \neq 0$ ), the viscous torque produces modifications in the resulting local equilibrium magnetization

$$\bar{M} \neq M_0\hat{e}_H \quad (32)$$

because of the change with respect to  $\hat{e}_H$  due to vorticity. Depending on the flow, a different magnetization vector can be obtained (75).

When magnetic suspensions are considered, another relaxation mechanism, different from the Debye-type relaxation, should be taken into account, that is, the so-called Néel effect, which contributes to the evolution of the magnetization. In this case, the relaxation time is given by

$$\frac{1}{\tau_M} = \frac{1}{\tau_D} + \frac{1}{\tau_N} \quad (33)$$

where  $\tau_N$  is Néel's relaxation time (82) defined as

$$\tau_N = \tau_{N0} \exp\left(\frac{Kv}{k_B T}\right) \quad (34)$$

where  $K$  is the anisotropy constant and  $v$  is the particle volume.

## DIFFUSIONAL PROCESSES IN SUSPENSIONS OF POLAR PARTICLES

### The Polarization Diffusion

Replacing Eqs. 26 and 27 with Eq. 28, an expression similar to that of Stiles and Hubbard (83) is obtained but



with two additional source terms appearing in the rhs of the following expression

$$\begin{aligned} \frac{D\bar{P}}{Dt} + \nabla \cdot \mathbf{J} - \frac{1}{\tau_D} (\chi(E)\bar{E} - \bar{P}) \\ = (\eta_E \delta_2 - \zeta \delta_6) \nabla^2 \bar{u} - 2\zeta \delta_6 \nabla \times (\bar{\omega} - \bar{\Omega}) \end{aligned} \quad (35)$$

The tensor  $\mathbf{J}$  is defined by

$$\mathbf{J} = D_2 (\nabla \bar{P})^s + D_3 (\nabla \bar{P})^a \quad (36)$$

By substituting Eq. 36 into Eq. 35, it is possible to identify  $D_2$  and  $D_3$  as diffusion coefficients related to translational and rotational polarization diffusion, respectively. In terms of the coupling parameters of Eqs. 26 and 27, the diffusion coefficients are defined as  $D_2 = 2\delta_2\delta_3$  and  $D_3 = -2\delta_6\delta_7$ . Moreover, the contribution of polarization diffusion coming from the symmetric and antisymmetric viscous stress tensors appears depending on the materials. In fact, according to the Stockmayer classification (84), the contribution of the antisymmetric viscous stress tensor is related to the rotational Brownian motion process, which appears in materials of low molecular weight or in polymers with molecular dipoles oriented perpendicularly to the backbone chains, for this case  $D_2 \ll D_3$ . On the other hand, the contribution of the symmetric viscous stress tensor is related to hydrodynamic fluctuations (Rouse's mode) of polymeric materials with molecular dipoles parallel to the backbone chains, for this case  $D_2 \gg D_3$ . A combination of these two relaxation modes is included in Eq. 35.

### Polarization Charge Diffusion

Now, consider the polarization charge density given by  $\rho_P = -\nabla \cdot \bar{P}$ , which represents the spatial variation of the polarization vector. The local variation of  $\rho_P$  only has contributions of the symmetric viscous stress tensor (79), as shown by taking the divergence of the linear Eq. 35 and substituting the linear Eqs. 26 and 27. The result is

$$\begin{aligned} \frac{\partial \rho_P}{\partial t} &= \left( \frac{\partial \rho_P}{\partial t} \right)_{sym} \\ &= -\frac{1}{\tau_D} \frac{\epsilon_0}{\epsilon_\infty} \rho_P + 2D_2 \nabla^2 \rho_P \end{aligned} \quad (37)$$

The subindex "sym" means that, in this equation, only the symmetric part of the stress tensor contributes to the variation of the polarization charge. This shows that the spatial distribution of polarization charge in a dipolar suspension evolves as a diffusion process, which influences the flow through the symmetric stress tensor.

### Diffusion of the Transverse Component of the Polarization Vector

Another diffusive phenomenon associated with suspensions of polar particles is the diffusion of the rotational of the polarization vector ( $\bar{P}_T = \nabla \times \bar{P}$ ). Here, the diffusion is that of a vector field (79) in contrast to that of a scalar field like in Eq. 37. In fact, applying the curl operator to the linear Eq. 28, the diffusion equation for  $\bar{P}_T$  is

$$\frac{\partial \bar{P}_T}{\partial t} + \frac{\bar{P}_T}{\tau_D} = D_T \nabla^2 \bar{P}_T \quad (38)$$

where the spin diffusion contribution was neglected and the spin was supposed to satisfy  $\nabla \cdot \bar{\omega} = 0$ . It should be noted that in the evolution of the transverse component of the polarization, both diffusion processes are present, that is, the hydrodynamic coupling with the symmetric stress tensor and that with the antisymmetric one, as can be seen in the total diffusion coefficient

$$D_T = D_2 + D_3 \quad (39)$$

### GENERALIZED HYDRODYNAMIC DESCRIPTION OF SUSPENSIONS OF POLAR PARTICLES

In the past few years, suspensions of Brownian particles (particles larger and heavier than the molecules of the solvent) have been investigated by a variety of experimental and theoretical methods. The neutron and light scattering (85), as well as ultrasonic absorption, are examples of experimental methods (86). Theoretically, suspensions have been investigated by means of nonequilibrium statistical mechanics, irreversible thermodynamics, and computer simulation. One of the aims of nonequilibrium statistical mechanics is the evaluation of the velocity autocorrelation function and the corresponding memory function in order to describe the dynamic response of the system to small perturbations of external or internal origin (fluctuations) (87). From this information and by means of the fluctuation-dissipation principle and the linear response theory, the transport coefficients or the response coefficients can be obtained. The theoretical description of the fast dynamics evolution in the local level (mesoscopic) is done in such a way that the transport and response coefficients may depend on  $\omega$  and  $\vec{k}$  (the frequency and the wavenumber). The slow dynamics (hydrodynamic limit) are obtained in the limit of zero  $\omega$  and  $\vec{k}$ . The reader interested in the details and the basis of these theories, should refer to Refs. 79 and 88.

### The Polarization-Autocorrelation Function

A particular insight of the evolution of the polarization vector of polar liquids and suspensions is obtained by means of the extended irreversible thermodynamics (89), which is useful to calculate a generalized Debye equation taking into account inertial terms, internal variables, and hydrodynamic-coupling effects (78). In this section, the first two effects are considered. The last one is discussed in the following sections.

Suppose that the polarization-autocorrelation function is

$$\phi_P(t) = \frac{\langle \delta \bar{P}(t) \cdot \delta \bar{P} \rangle}{\langle \delta \bar{P} \cdot \delta \bar{P} \rangle} \quad (40)$$

where  $\delta \bar{P}(t) = \bar{P}(t) - \langle \bar{P} \rangle$ .

The linear generalized Debye equation for  $\phi_P$  is expressed, in the absence of hydrodynamic couplings and  $\bar{u} = 0$ , (90), as

$$\begin{aligned} & \left( \frac{\lambda_3}{\omega_{av}^2} + \lambda_2^2 \tau_D \right) \frac{d^3 \phi_P}{dt^3} + \left( \frac{1}{\omega_{av}^2} + 2\lambda_2 \tau_D \right) \frac{d^2 \phi_P}{dt^2} \\ & + (\lambda_3 + \tau_D) \frac{d \phi_P}{dt} + \phi_P \\ & = \chi_0 \left( 1 + \lambda_3 \frac{d}{dt} \right) \phi_E \end{aligned} \quad (41)$$

where the correlation between the electric field and the initial polarization is given by

$$\phi_E(t) = \frac{\langle \delta \bar{E}(t) \cdot \delta \bar{P} \rangle}{\langle \delta \bar{P} \cdot \delta \bar{P} \rangle} \quad (42)$$

and  $\delta \bar{E}$  is the fluctuation of the local electric field.

The parameters  $\tau_D$  and  $\omega_{av}$  appearing in Eq. 41 have already been defined in Eq. 25 and Eq. 7, respectively. The parameter  $\lambda_3$  comes from the mean square torque produced by dipolar-dipolar interaction between molecules, while  $\lambda_2$  is the cross-correlation time between the dipolar relaxation and the libration of dipoles in the local electric field (91). The last parameter,  $\lambda_2$ , is considered as free, in the sense that it can be obtained by means of a comparison between the predictions of the above equation and the experimental data (90). When  $\lambda_2 = \lambda_3 = 0$ , a particular case of Eq. 41 is obtained, which is the first generalization of the Debye equation for  $\phi_P$ . That is

$$\frac{1}{\omega_{av}^2} \frac{d^2 \phi_P}{dt^2} + \tau_D \frac{d \phi_P}{dt} + \phi_P = \chi_0 \phi_E \quad (43)$$

This result describes a resonant absorption with the characteristic frequency  $\omega_{av}$ , which is a consequence of the inertial effect. A comparison with experimental data

for trichloroethane-hexane (92) solution is given in Ref. 90. Usually, this resonance peak is found at high frequencies, but for the frequency-domain of dielectric relaxation, this effect can be neglected. In this case, the decay relaxation equation is given by

$$\tau_D(t) \frac{d \phi_P}{dt} + \phi_P = \chi_0 \phi_E \quad (44)$$

Where  $\tau_D(t)$  is the Debye-relaxation time, which is a time-dependent parameter (93). According to the coupling model of Ngai (94), this parameter is given by

$$\tau_D(t) = \frac{\tau_0}{\alpha} \left( \frac{t}{\tau_0} \right)^{1-\alpha} \quad (45)$$

where  $\tau_0$  and  $\alpha$  are the average relaxation time and the disorder parameter, respectively. The corresponding correlation function is of the form of Kolhrausch-Williams-Watts (95) when there is no correlation between the electric field and the initial polarization and  $\phi_E = 0$  (the system has no ferroelectric properties):

$$\phi_P(t) = \exp \left\{ - \left( \frac{t}{\tau_0} \right)^\alpha \right\} \quad (46)$$

which indicates the presence of a distribution of characteristic times of a series of relaxation processes in parallel in such a way that the relaxation mechanism is due to multiple channels of polarization-relaxation with different rates (96).

On the other hand, a solution of Eq. 41 can be obtained supposing that the coefficients are constants and  $\phi_E = 0$ . The solution can be cast into a third-order continued fraction in the Laplace transform space. That is

$$\tilde{\phi}_P(S) = \frac{1}{s + \frac{M}{s + \frac{M + \gamma_2}{s + \frac{\gamma_3}{M}}}} \quad (47)$$

where

$$M = - \frac{\gamma_3}{\gamma_1} \quad (48)$$

and

$$\gamma_1 = \left( \frac{1}{\omega_{av}^2} + 2\lambda_2 \tau_D \right) \left( \frac{\lambda_3}{\omega_{av}^2} + \lambda_2^2 \tau_D \right)^{-1} \quad (49)$$

$$\gamma_2 = (\lambda_3 + \tau_D) \left( \frac{\lambda_3}{\omega_{av}^2} + \lambda_2^2 \tau_D \right)^{-1} \quad (50)$$

$$\gamma_3 = \left( \frac{\lambda_3}{\omega_{av}^2} + \lambda_2^2 \tau_D \right)^{-1} \quad (51)$$

Del Castillo et al. (90) have shown that the result given in Eq. 47 reduces to the three variable theory (97) and from comparison with the experimental data for chloroform (98), they also have shown that the introduction of the cross-correlation time is necessary. Now, considering the linear response theory, the complex electric susceptibility (88) is

$$\frac{\chi^*(\omega) - \chi_\infty}{\chi_0 - \chi_\infty} = \frac{\epsilon^*(\omega) - \epsilon_\infty}{\epsilon_0 - \epsilon_\infty} = L(-\dot{\phi}_P) \\ = 1 - i\omega \tilde{\phi}_P(\omega) \quad (52)$$

where  $\chi_0$  (or  $\epsilon_0$ ) and  $\chi_\infty$  (or  $\epsilon_\infty$ ) are the electric susceptibility (or dielectric constant) defined at zero and at high frequency, respectively.  $L(\cdot)$  denotes the Laplace transform.

### Suspensions with Dipolar Particles: Hydrodynamics Coupling and the Symmetric Stress Tensor

Now, a discussion is given about the problem of the complex electric and magnetic susceptibilities corresponding to the hydrodynamic-like description of the evolution of the polarization vector coupled with the symmetric and antisymmetric total stress tensors, and their experimental comparison. In particular, the effect of the polarization distribution on the absorption bands is discussed.

In Eq. 28, the evolution of the polarization vector is governed by the modified-Debye equation with a coupling term that represents the structural effect when the electric dipole-particles are coupled with the translational velocity, included formally in the total symmetric stress tensor (see Eq. 26).

Now, it is supposed that the polar suspension is in the equilibrium state at the onset. Then, an external oscillatory electric field of small strength is applied. Therefore, there is no saturation effect and  $\chi(E) = \chi_0$ . The polarization vector  $\delta\bar{P}$  and the stress tensor  $\delta\mathbf{Q}_T^S$  are induced and change with time at the same frequency as that of the electric field. Consequently, Eqs. 26 and 28 with  $\delta_6 = 0$  give (99) us

$$\delta\mathbf{Q}_T^S + \lambda_2 \frac{D\delta\mathbf{Q}_T^S}{Dt} = 2\eta_E (\nabla\delta\bar{u})^S + 2\delta_3 (\nabla\delta\bar{P})^S \quad (53)$$

$$\tau_D \frac{D\delta\bar{P}}{Dt} = \chi_0 \delta\bar{E} - \delta\bar{P} + \delta_2 \tau_D \nabla \cdot \delta\mathbf{Q}_T^S \quad (54)$$

In order to close the system of differential equations, the balance of the linear momentum equation for the fluctuations is required, and use is made of its form given in Eq. 20. Linearizing the system of equations, taking the Laplace transform, and supposing that the initial values of the variables are equal to zero, the following results are obtained for the longitudinal and transversal modes, respectively,

$$\frac{\chi_L(k, \omega)}{\chi_0} = [1 + i\omega\tau_D^L]^{-1} \quad (55)$$

$$\frac{\chi_T(k, \omega)}{\chi_0} = \left[ 1 + i\omega\tau_D^T + \frac{D_2 k^2 \tau_D^T}{(1 + i\omega\tau_2)} \right]^{-1} \quad (56)$$

The contribution of the velocity modifies the relaxation time  $\tau_2$  as

$$\tau_2 + \frac{v_0 k^2}{\omega^2}.$$

But if the electric field in the sample is supposed to be a plane wave ( $\omega = ck$  and  $c$  is the speed of light in the sample), the term that modifies  $\tau_2$  can be neglected for any  $v_0$ . Therefore, the effect of the translational velocity of the dipole particles on the absorption band can not be detected experimentally. However, the polarization diffusion effect is an important contribution that should be taken into account in the electric susceptibility, according to Eq. 56. A way to compare the result obtained in this equation with experimental data on dielectric relaxation is to consider the normal mode dielectric relaxation in which the polarization vector  $\bar{P}$  is the resultant sum of the molecular dipolar vectors found along the backbone of the polymeric chains. Therefore,  $D_2$  is the diffusion coefficient of the polarization process, which coincides with the same diffusion coefficient of the polymeric solution as described by the Rouse-Zimm theory in the dilute case and with the reptation theory in the case of concentrated solutions or polymeric melts (100).

### Suspensions with Dipolar Particles: Hydrodynamic Coupling and the Antisymmetric Stress Tensor

In a similar way as in the preceding section, Eq. 28 describes the evolution of the polarization vector governed by the modified-Debye equation with a coupling term, which represents the structural effect when the electric dipole-particles are coupled to the antisymmetric stress tensor (see Eq. 26).

Now, suppose the polar suspension is in the equilibrium state at the onset and then an external oscillatory

electric field of small strength is applied. Therefore, there is no saturation effect, and  $\chi(E) = \chi_0$ . The polarization vector  $\delta\bar{P}$  and the stress tensor  $\delta Q_T^a$  change slightly with the same frequency of the electric field. Consequently, Eqs. 27 and 28 with  $\delta_2 = 0$  give (101) us

$$\delta Q_T^a + \lambda_5 \frac{D\delta Q_T^a}{Dt} = 2\delta_7 (\nabla\delta\bar{P})^a \quad (57)$$

$$\tau_D \frac{D\delta\bar{P}}{Dt} = \chi_0 \delta\bar{E} - \delta\bar{P} + \delta_6 \tau_D \nabla \cdot \delta Q_T^a \quad (58)$$

Linearizing the system of equations, taking the Laplace transform, and assuming that the initial values of the variables are equal to zero, the following results for the longitudinal and transversal modes are obtained, respectively

$$\frac{\chi_L(k, \omega)}{\chi_0} = [1 + i\omega\tau_D^L]^{-1} \quad (59)$$

$$\frac{\chi_T(k, \omega)}{\chi_0} = \left[ 1 + i\omega\tau_D^T + \frac{D_3 k^2 \tau_D^T}{(1 + i\omega\tau_5)} \right]^{-1} \quad (60)$$

The polarization diffusion is present in the transverse mode, which is measured in the laboratory when the parallel plate capacitor arrangement is used. The diffusion mechanism for the polarization is produced by the Brownian-vortex motion, which controls the diffusion rate when the substance is in the undercooling regime at temperatures higher than that of glass transition. The comparison with experimental data is given considering the so-called  $\alpha$ -relaxation mode of dielectric relaxation. This mechanism is associated with the polymeric substances as well as with any glass-forming liquids (102).

### The Role of the Interactions in the Hydrodynamic Coupling Mechanism

Several authors have recognized (103, 104) that the effect of the long-range contributions produced by the dipole-dipole interaction can be introduced into the parameters involved in the Debye relaxation equation, that is, into the electric susceptibility,  $\tau_D$  and  $\omega_{av}$ . Therefore, this sort of interaction should be excluded from the interpretation of the hydrodynamic coupling diffusion.

The coupling between the polarization vector and the viscous stress tensors should be considered of long range and is of structural nature. Due to this, there exist local structural differences, and consequently the wavenumber shows dispersive effects, that is,  $k(\omega)$ . Therefore, the following dispersion formula is used to improve the compar-

### Hydrodynamic Behavior of Suspensions of Polar Particles

ison between experimental data and the results of Eqs. 56 and 60 (99)

$$k = k_0 \left( \frac{\omega}{\omega_0} \right)^n \quad (61)$$

where the parameter  $n$  can be determined by fitting the experimental data.

### General Expression for Dielectric $\alpha$ and Normal Relaxations

For polymeric materials that have dipolar components along the backbone of the polymeric chain as well as in the perpendicular directions, both mechanisms of diffusion are present. However, the frequency of the loss peak of the  $\alpha$ -relaxation is usually separated from the loss peak of the normal-mode relaxation by two or three frequency decades (105). Therefore, we can consider that there is no interaction between the respective relaxation mechanisms and that a superposition of the two relaxation modes can be established in the same equation as follows (99)

$$\frac{\chi_T(k, \omega)}{\chi_0} = \left[ 1 + i\omega\tau_e^T + \frac{D_3 k^2 \tau_e^T}{(1 + i\omega\tau_5)} + \frac{D_2 k^2 \tau_e^T}{(1 + i\omega\tau_2)} \right]^{-1} \quad (62)$$

where  $\tau_e^T$  is the composition of the relaxation times resulting from the expression

$$\frac{1}{\tau_e^T} = \left( \frac{1}{\tau_D^T} \right)_{sym} + \left( \frac{1}{\tau_D^T} \right)_{antisym} \quad (63)$$

Fig. 2 of Ref. 99 shows the presence of two peaks, one at the right associated with the dielectric  $\alpha$ -relaxation and another one at the left related to the dielectric normal mode relaxation for the cis-polyisoprene data from Adachi and Kotaka (106). In order to fit these curves with Eq. 62, use was made of the dispersion relation  $k(\omega)$  proposed above.

### MICROPOLAR SUSPENSIONS: APPLICATIONS AND EXPERIMENTS

Suspensions in which the spin of the particles is important are called micropolar. Different theories have been put forward to give the equations of motion of these fluids. A particularly popular one is that due to Cemal Eringen (107–115). Other theories have been developed, for example, by Shliomis (116), Kline (117), Pérez-Madrid et al. (118, 119), Dávalos-Orozco and del Castillo (120), and

Faria and Kramer (121). The model equations obtained by Dávalos-Orozco and del Castillo (120) reduce to those of a micropolar suspension, inside the framework of their theory, when the mass-polarization vector is zero and the viscoelastic time constants are zero. Apart from Ref. (120), a viscoelastic micropolar fluid theory has been proposed by Yeremeyev and Zubov (122). Some review works, for different periods, are those of Ariman et al. (123), Cowin (124), the book by Stokes (125), and the book by Lucaszewicz (115).

The equations of motion of a compressible micropolar fluid are (125)

$$\frac{d\rho}{dt} + \rho \nabla \cdot \bar{u} = 0 \quad (64)$$

$$\rho \frac{d\bar{u}}{dt} = -\nabla p + (\lambda + \mu - \kappa) \nabla(\nabla \cdot \bar{u}) + (\mu + \kappa) \nabla^2 \bar{u} + 2\kappa \nabla \times \bar{\omega} + \rho \bar{f} \quad (65)$$

$$\rho j \frac{d\bar{\omega}}{dt} = 4(\alpha + \beta) \nabla(\nabla \cdot \bar{\omega}) + 4\gamma \nabla^2 \bar{\omega} + 2\kappa \nabla \times \bar{u} - 4\kappa \bar{\omega} + \rho \bar{l} \quad (66)$$

Here,  $\rho$  is the suspension density,  $p$  is the pressure,  $\bar{u}$  is the fluid velocity,  $\bar{\omega}$  is the spin or internal rotation,  $j$  is the micro inertia (mean moment of inertia of the particles),  $\bar{f}$  is an external body force per unit mass, and  $\bar{l}$  is a body couple per unit mass. Besides,  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\kappa$ ,  $\lambda$ , and  $\mu$  are material constants (viscosities) that must satisfy the following inequalities derived from the Clausius-Duhem inequality:

$$\begin{aligned} 3\lambda + 2\mu &\geq 0 & \mu &\geq 0 & \kappa &\geq 0 & \gamma - \beta &\geq 0 \\ \gamma + \beta &\geq 0 & 3\alpha + \beta + \gamma &\geq 0 \end{aligned}$$

Next, some fundamental applications of the micropolar fluids will be discussed.

### Flow Solutions

The difficulty of the equations is based not only on the increase in the number of terms but also on the non-linearity. Therefore, one important goal is to obtain fundamental flow solutions. Saccomandi (126) obtained stationary solutions of plane flow by means of the Lie-group analysis. In the flow between two rotating disks, spin is generated, and the results obtained by Rao and Kasiviswanathan (127) are of interest to understand the flow behavior in an orthogonal rheometer, which has this geometry. Nonlinear equations were obtained by Chaturani and Narasimman (128) and analyzed numerically.

The problem of flow between coaxial walls in relative motion, which corresponds to that of lubrication in journal bearings, was investigated by Prakash and Sinha (129) who found that with micropolar fluids the effective viscosity increases. When the journal bearing is rough, Sinha and Singh (130) found that the effects of roughness were enhanced with a micropolar fluid. If the relative motion of the boundaries is that of two parallel plates, interesting applications to human joints have been found where the lubricating synovial fluid (made of long-chain hyaluronic acid molecules) is supposed to be a micropolar suspension. One model is that of two approaching solids, where one is porous, by Tandon and Jaggi (131). In the other model, Tandon and Rakesh (132) include the effect of cartilage roughness and found that load capacity increases with a decrease of gap width and with an increase of particle concentration. Sinha et al. (133) modeled the hip joint as a spherical bearing. They found that the effective viscosity increases and that, for a damaged joint, the load capacity decreases. Tandon and Gupta (134) introduced surface roughness by means of a cosine wave over the two plane surfaces and modeled the knee joint by rectangular plates and the hip joint by circular plates. Singh et al. (135) introduce a varying porosity model of the cartilage matrix in the human joints in order to avoid modeling it as a three-layer porous system. An interesting application of the micropolar fluid model was made by Sinha et al. (136) to the cervical mucus. Besides, the effect of a flexible cervical wall with muscular activity is introduced by Philip and Chandra (137) who found that the propulsive velocity of the spermatozoa increases when the fluid is more non-Newtonian. The blood has been modeled as micropolar fluid in small capillaries and large arteries taking into account the rotation and deformation of the red cells by Ariman et al. (138) who investigated the stationary and oscillatory blood flow and concluded that their results have certain agreement with the experimental results of Bugliarello and Sevilla (139). Chaturani and Upadhyaya (140) modeled the blood as made of a micropolar core region of red cells and a peripheral plasma layer and compare their results with other models. Chaturani and Mahajan (141) propose a method to calculate the boundary conditions of a suspension and compare their results with experiments of blood flow. Xiexian and Yuansheng (142) change the typical circular cross section of the blood vessel into an elliptical one to investigate the flow of micropolar blood. Stratification of the blood under flow is investigated by Akay and Kaye (143) considering small capillaries. Tandon et al. (144) introduced a small stenosis (constriction) in the tube to the flow of a two-fluid blood (the inner one being micropolar). The problem of stenosis was also investigated by Hogan and Henriksen (145).

More recently, Sanyal and Maiti (146) modeled the flow of blood through a bifurcation with the goal of understanding some cardiovascular problems at the bifurcation. The micropolar model has also been tested in the slow flow approximation. The paradox of the drag on a body of arbitrary shape was investigated by Ramkissoon (147). The resistance of a particle to translational and rotational motion was calculated independently by Brunn (148), Ramkissoon and O'Neill (149), and Avudainayagam and Ramakrishna (150). Calculations under the Oseen approximation for two-dimensional flow have been done by Olmstead and Majumdar (151). Small amplitude oscillations of a symmetric body stimulate vorticity, which generates spin motion through spin viscosity. In this case, Rao and Rao (152) showed that, for longitudinal and rotational oscillations of a sphere, the maximum drag or couple is larger than that in a Newtonian fluid. The simultaneous longitudinal and torsional oscillations of an infinite rod in an unbounded micropolar fluid were investigated by Ramkissoon et al. (153). Charya and Iyengar (154) found a general formula for the drag on an axisymmetric body performing longitudinal oscillatory motions. The boundary layer flow is important in many applications when the velocity is high and the viscosity is low. The boundary layer on an accelerating extruded sheet with the effects of heat transfer was investigated by Hassanien (155). Bhargava and Takhar (156) showed that in any stagnation point of flow, the temperature in the boundary layer is higher than that in a Newtonian fluid. An application to shear waves along the interface of a linear elastic medium and a micropolar fluid was investigated by Yerofeyev and Soldatov (157) who found that the shear surface wave propagates almost without dispersion, in contrast to the Newtonian fluid.

### Flow Instability

The stability of flow is important to understand the behavior of fluids with spin under a perturbation. Natural convection of a micropolar fluid layer heated from below was investigated by Bhattacharyya and Jena (158) who found that the spin does not affect the stability. Besides, Bhattacharyya and Abbas (159) concluded that the spin effects make a more unstable rotating convecting fluid layer. Franchi and Straughan (160) investigated, by the energy method, the nonlinear convective stability of a fluid with temperature-dependent viscosity and found that the effect of spin is very small. When the temperature gradient is nonuniform in convection, Siddheshwar and Pranesh (161) determined that the micropolar fluid is more unstable than the Newtonian one. Mrabti et al. (162) concluded that when the fluid is enclosed in a cylindrical con-

tainer heated from below, the heat transfer rate is smaller than that of a Newtonian fluid. When investigating natural convection with heat sources, Hsu et al. (163) found that a change of spin boundary conditions changes the heat transfer of the convective layer. For an electric conducting fluid under an external magnetic field, Rao (164) and Siddheshwar and Pranesh (165) found that the micropolar fluid is more stable than the Newtonian one under the same conditions. However, Rao (164) has shown that it is also possible to destabilize this system when the temperature gradient is not adverse (that is, heated from above).

The stability is important when the fluid has a main flow. For example, for the linear stability of a micropolar fluid flowing between two parallel walls, Kuemmerer (166) concluded that the spin has a destabilizing effect. The nonlinear stability of a fluid layer flowing down an inclined plane was investigated by Hung et al. (167). Their general conclusion is that the spin effects are stabilizing. Hung and Tsai (168) investigated the influence of the spin on the rupture of a micropolar fluid on a horizontal wall and found that the microstructure stabilizes by delaying the rupture of the film.

### Spin Boundary Conditions

In some papers reviewed above, the problem of the spin boundary conditions at a solid wall were discussed. This problem was taken as the main subject of research by some authors. Tözeren and Skalak (169) posed this question from the point of view of a suspension of spherical particles. An analysis of different boundary conditions was made by Migun (170) who also proposed a boundary condition that depends on experimental parameters. Because there is no experimental evidence of the correct boundary condition, in general, some authors feel safe using in their theoretical calculations a condition that connects the spin with the vorticity by means of a linear proportionality constant, as done by Hsu et al. (163) for natural convection and by Hogan and Henriksen (171) for numerical analysis of flow through a constricted tube. The problem is that this constant increases the number of parameters in calculations of flow of micropolar fluids, which was already large. Experimental research is needed to solve this problem.

### Fundamental and Applied Experiments

Some authors have developed experiments to obtain the magnitudes of the coefficients of micropolar fluids. For example, Ebert (172) measured three constants and showed that they can be determined by means of the geometry variation of the measuring apparatus. Fodemski and

Morawski (173) subjected water, ethyl alcohol, and nitrobenzen, to a rotating electric field to calculate the corresponding spin viscosity and the internal volume torque. Viscometric and thermal measurements were done by Kolphchikov et al. (174) to obtain two characteristic parameters of micropolar fluids. They also give formulas for the value of the linear proportionality constant of the boundary condition, which relates the spin with the vorticity at the solid boundary. Besides, the micropolar fluids theory is used for experimental capillary penetrant testing by Prokhorenko et al. (175). The micropolar theory has been checked experimentally for fluid behavior in micromachined channels by Papautsky et al. (176). They showed that the micropolar theory predicts experimental data better than the Newtonian one.

## MAGNETIC FLUIDS: APPLICATIONS AND EXPERIMENTS

The magnetic fluids have interesting applications, and, therefore, many experiments have been developed in order to characterize their response to magnetic fields under different conditions, isothermal or nonisothermal, with increase or decrease of particle concentration, etc. In what follows, some experiments on magnetic fluids are discussed, and later some applications are presented.

General reviews of magnetic fluids appear in the books by Rosensweig (32), Bashtovoy et al. (177), Berkovsky et al. (178), Berkovskii and Bashtovoy (179), and Blums et al. (180). Discussions ranging from the fundamental concepts, flow problems, and applications may be found in those references.

### Aggregation

One problem in magnetic fluids is that of the aggregation of particles, and it has been the subject of experiments (181, 182). Aggregation in pipe flow has been investigated by Kamiyama and Satoh (183). Zubko et al. (184) investigated magnetic suspensions under oscillating electric fields.

### Pattern Formation

Interfacial instabilities were found very early, were investigated by Cowley and Rosensweig (185), and were included in the more general area of pattern formation by Tsebers and Blums (186). Interesting patterns were found in a two-layer system made by a magnetic fluid over a nonmagnetic one by Petit et al. (187). Interfacial patterns were investigated by Okubo et al. (188) when an alternating magnetic field is applied. Boudovis et al. (189) inves-

tigated axisymmetric shapes of sessile drops under a magnetic field.

### Rotating Magnetic Field

The spin-up of a magnetic fluid in the presence of a rotating magnetic field has been the subject of different experiments made to confirm the theory and to find new mechanisms of flow (190–192). When small nonmagnetic spheres are confined in thin magnetic fluid layers and the magnetic field rotates, it has been shown by Helgesen and Skjeltop (193) that the spheres rotate in the opposite direction as the field but without translation. The bending of drops in the rotating magnetic field was investigated by Laci (194).

### Viscosity and Negative Viscosity

An interesting theoretical result has been corroborated experimentally, that is, the appearance of a negative viscosity when an alternating magnetic field is applied to a flow. Shliomis and Morozov (195) found this phenomenon theoretically, and Bacri et al. (196) obtained the experimental results (see also Ref. 197). It has been shown by Shliomis and Kamiyama (198) that a nonuniform magnetic field causes an inhomogeneous distribution of magnetic particles even in oscillatory flow. Besides, Larrondo and van de Ven (199) concluded that magnetic suspensions present viscoelastic properties under an oscillating shear and a magnetic field. The viscosity of magnetic suspensions was investigated by Choi et al. (200) as a function of concentration and shear rate.

### Flow Instability

The problem of stability of flow has been the subject of intensive research due to the difficulty of controlling the flow in potential applications. Therefore, experiments of fundamental character have been done in this area. Berkovsky and Bashtovoi (201) discuss the rupture of drops, layers, and cylinders. Besides, Rother and Richter (202) investigate the rupture of a liquid bridge. When a fluid layer is heated from below, natural convection patterns appear whose form depends on the field strength and boundary conditions as shown experimentally by Schwab et al. (203–205). In contrast with a Newtonian fluid, ferrofluids in natural convection have the magnetic Rayleigh number (apart from the usual one) whose influence alone was investigated in microgravity experiments by Odenbach (206). Another fundamental problem is that of Taylor-Couette flow investigated by Odenbach and Gilly (207) under an azimuthal magnetic field who used their results to obtain the rotational viscosity as a function of the

magnetic field strength. Recently, two methods for the measurement of the velocity profile have been proposed. One is the ultrasonic Doppler method by Kikura et al. (208), and the other one is small-angle neutron scattering by Odenbach et al. (209).

### Magnetic Susceptibility

An important investigation is that related with the experimental calculation of the magnetic susceptibility of a magnetic colloidal suspension. The experiments are made for particular kinds of suspended particles. Experiments have been done for manganese compounds (210, 211), ferrite particles (212), magnetite particles (213), cobalt particles (214, 215), and iron-oxide (211, 216). In some experiments, Néel relaxation has been detected (217–220). It is also of interest to check the dependence of susceptibility on temperature and concentration, like, for example, in the case of magnetite (221).

### Magnetic Fluids Applications

The increasing theoretical development of magnetic fluids is due to the potential they have for applications. Berkovsky et al. (222) have proposed the use of magnetic fluid coatings in different hydrodynamic systems to control flow separation, drag, and heat transfer. Applications to loudspeaker drivers, to inspection of internal passages, to instrument damping, and to accelerometers were explained by Bailey (223). Other uses are in seals, journal bearings, magnetogravimetric separators, and to transducers, as discussed by Anton et al. (224). Charles (225) reviews other uses, such as magnetic inks and to produce magneto-optic effects in the microwave region like birefringence and dichroism. As lubricants, they have other applications as supports and dampers (178). Applications to engineering reach areas like energy conversion systems (226), shock absorbers (227), inertial sensors (228), and brakes (229). In biomedicine, they have been applied to hyperthermia for the treatment of cancer by Jordan et al. (230) and to the treatment of other related problems by Hiergeist et al. (231). More information on applications is found in chapters 4 and 5 of Ref. 179 and in chapters 4, 5, and 6 of Ref. 178.

### FUTURE OF SUSPENSIONS

The attraction of suspensions has increased in recent years due to the important applications they have, not only in industry, but also in everyday life. The fascination of this subject has spread in such a way that today more scientists are working theoretically and experimentally in all the

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classic areas and in the new ones. Many scientists are investigating the so-called "intelligent" fluids of which new properties are found constantly. The frequency of conferences on electro- and magneto-rheological fluids has increased to almost one a year, and they are held in different geographical regions. This is proof that the fever to find new properties and possibilities for colloidal suspensions is increasing all over the world. The science of colloidal suspensions is far from being exhausted, and a large number of fundamental details are still waiting for an explanation and to be assigned a practical application, apart from the many more that are waiting to be discovered.

However, from the basic point of view, several problems are still open and are of particular interest. For example, they are the spin boundary conditions at a wall and at an interface, the nonhomogeneous stress distribution-induced diffusion, the nonlinear relation between the internal rotation of a polar fluid and hydrodynamics for high and low shear rate limits, and dispersion effects in compressible and concentrated colloidal dispersions.

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### REFERENCES

1. Gelbart, W.M.; Ben-Shaul, A. The "new" science of "complex fluids". *J. Phys. Chem.* **1996**, *100*, 13169.
2. Gast, A.P.; Russel, W.B. Simple ordering in complex fluids. *Phys. Today* **1998**, *51* (12), 24.
3. Larson, R.G. *The Structure and Rheology of Complex Fluids*; Oxford University Press: New York, 1999.
4. *Dynamics of Complex Fluids. Proceedings of the Royal Society-Unilever Indo-UK Forum in Materials Science and Engineering*; Adams, M.J., Mashelkar, R.A., Pearson, J.R.A., Rennie, A.R., Eds.; ICP and the Royal Society, 1998.
5. Girasole, T.; Darrigo, R.; Gouesbet, G.; Roze, C. Two-phases Rayleigh-Bénard instabilities. *Phys. Fluids* **1995**, *7*, 2659.
6. Mackie, C. Convective stability of a particle-laden fluid system in the presence of solidification. *Int. J. Heat Mass Transfer* **2000**, *43*, 1617.
7. Witten, T.A. Insights from soft condensed matter. *Rev. Mod. Phys.* **1999**, *71* (2), S367.
8. Goodwin, J.W.; Reynolds, P.A. The rheology of flocculated suspensions. *Curr. Opin. Colloid Interface Sci.* **1998**, *3*, 401.



9. van Egmond, J.W. Shear-thickening in suspensions, associating polymers, worm-like micelles, and poor solutions. *Curr. Opin. Colloid and Interface Sci.* **1998**, *3*, 385.
10. Metzner, A.B. Rheology of suspensions in polymeric liquids. *J. Rheol.* **1985**, *29*, 739.
11. Barthes-Biesel, D. Suspensions of Capsules. In *Flow of Particles in Suspensions*; Schaflinger, U., Ed.; CISM Courses and Lectures No. 370, Springer Wien: New York, 1996.
12. Coussot, P. *Mudflow: Rheology and Dynamics*; International Association for Hydraulic Research Monograph Series Balkema, A.A. Balkema, Lisse, The Netherlands: 1997.
13. Coussot, P.; Fiau, J.-M. The flow of mud. *La Recherche* **1993**, *258* (24), 1024 (in French).
14. Schramm, L.L. Suspensions: Basic Principles. In *Suspensions: Fundamentals and Applications in the Petroleum Industry*; Schramm, L.L., Ed.; American Chemical Society: Washington, DC, 1996.
15. Grüner, F.; Lehmann, W.P. Dynamics of colloidal crystals and colloidal liquids. *J. Phys. A* **1982**, *15*, 2847.
16. Overbeck, J.Th.G. Recent developments in the understanding of colloid stability. *J. Colloid Interface Sci.* **1977**, *58*, 408.
17. Mege, W.V.; Snook, I. Diffusion in concentrated monodisperse colloidal solutions. The hard sphere-thermodynamics or hydrodynamics? *Faraday Discuss. Chem. Soc.* **1983**, *76*, 151.
18. Ungarish, M. *Hydrodynamics of Suspensions: Fundamentals of Centrifugal and Gravity Separation*; Springer-Verlag: New York, 1993.
19. Ungarish, M. Recent Developments in the Analysis of Gravity and Centrifugal Separation of Non-Colloidal Suspensions and Unfolding Challenges in the Classic Mechanics of Fluids. In *Flow of Particles in Suspensions*; Schaflinger, U., Ed.; CISM Courses and Lectures No. 370, Springer Wien: New York, 1996; Vol. 370.
20. Davis, R.H.; Acrivos, A. Sedimentation of noncolloidal particles at low Reynolds numbers. *Annu. Rev. Fluid Mech.* **1985**, *17*, 91.
21. Blanc, R.; Guyon, E. The physics of sedimentation (in French). *La Recherche* **1991**, *234*, 866.
22. Happel, J.; Brenner, H. *Low Reynolds Number Hydrodynamics: with Special Applications to Particulate Media*; Kluwer Academic, 1991.
23. Chow, A.W.; Sinton, S.W.; Iwamiya, J.H.; Stephens, T.S. Shear-induced particle migration in Couette and parallel-plate viscometers: NMR imaging and stress measurements. *Phys. Fluids* **1994**, *6*, 2561.
24. Agarwal, U.S.; Dutta, A.; Mashelkar, R.A. Migration of macromolecules under flow: the physical origin and engineering implications. *Chem. Eng. Sci.* **1994**, *49*, 1693.
25. Del Castillo, L.F.; Criado-Sancho, M.; Jou, D. Nonequilibrium chemical potentials and shear-induced migration of polymer in dilute solutions. *Polymer* **2000**, *41*, 2633.
26. Leighton, D. Viscose resuspension. *Chem. Eng. Sci.* **1986**, *41*, 1377.
27. Taca, C.D.; Paunescu, M. Suspension of solid particles in spherical stirred vessels. *Chem. Eng. Sci.* **2000**, *55*, 2989.
28. Ross, S.; Morrison, I.D. *Colloidal Systems and Interfaces*; J. Wiley: New York, 1991.
29. Zuber, N. On the dispersed two-phase flow in the laminar flow regime. *Chem. Eng. Sci.* **1964**, *19*, 897.
30. Adams, M.J.; Briscoe, B.J.; Kamjab, M. The deformation and flow of highly concentrated dispersions. *Adv. Colloid Interface Sci.* **1993**, *44*, 143.
31. Dávalos-Orozco, L.A.; del Castillo, L.F. Dielectric behavior of viscous fluids. *J. Non-Equilib. Thermodyn.* **1990**, *15*, 11.
32. Rosensweig, R.E. *Ferrohydrodynamics*; Cambridge University Press: New York, 1985.
33. Brenner, H. Rheology of a dilute suspension of dipolar spherical particles in an external field. *J. Colloid Interface Sci.* **1970**, *32*, 141.
34. Alarcon Waess, O.; García-Colín, L.S. Hydrodynamic description of suspensions of Brownian particles. *J. Chem. Phys.* **1990**, *92*, 3086.
35. Ferrer, M.; Jou, D. On the nonequilibrium thermodynamics of diluted suspensions. *J. Chem. Phys.* **1988**, *89*, 1651.
36. Batchelor, G.K. The effect of Brownian motion on the bulk stress in suspension of spherical particles. *J. Fluid Mech.* **1977**, *83*, 97.
37. Batchelor, G.K.; Green, J.T. Determination of bulk stress in a suspension of spherical particles to order to  $C^2$ . *J. Fluid Mech.* **1972**, *56*, 401.
38. Wajnryb, E.; Dahler, J.S. The Newtonian Viscosity of a Moderately Dense Suspension. In *Advances in Chemical Physics*; Prigogine, I., Rice, S.A., Eds.; John Wiley and Sons: New York, 1997; Vol. 102.
39. Russel, W.B.; Gast, A.P. Nonequilibrium statistical mechanics of concentrated colloidal dispersion: hard spheres in weak flows. *J. Chem. Phys.* **1986**, *84*, 1815.
40. Bedeaux, D. The effective viscosity for a suspension of spheres. *J. Colloid Interface Sci.* **1987**, *118*, 80.
41. Brady, J.F.; Morris, J.F. Microstructure of strongly sheared suspensions and its impact on rheology and diffusion. *J. Fluid Mech.* **1997**, *348*, 103.
42. Ferry, J.D. *Viscoelastic Properties of Polymers*, 3rd Ed.; Wiley: New York, 1980.
43. Wessel, R.; Ball, R.C. Fractal aggregates and gels in shear flow. *Phys. Rev. A* **1992**, *46*, 3008.
44. Potonin, A.; Uriev, N.B. Microrheological models of aggregated suspensions in shear flow. *J. Colloid Interface Sci.* **1991**, *142*, 385.
45. Snabre, P.; Heider, L.; Baynard, M. Ultrasound and light scattering from a suspension of reversible fractal clusters in shear flow. *Eur. Phys. J. A* **2000**, *1*, 41.
46. Foss, D.R.; Brady, J.F. Brownian dynamics simulation of hard-sphere colloidal dispersions. *J. Rheol.* **2000**, *44*, 629.
47. Woods, L.C. *The Thermodynamics of Fluid Systems*; Clarendon Press: Oxford, 1975; Chap. 8.
48. Kestin, J.; Dorfman, J.R. *A Course in Statistical Thermodynamics*; Academic Press: New York, 1971.
49. De Groot, R.S.; Mazur, P. *Non-Equilibrium Thermodynamics*; Dover Pub: New York, 1984.

50. Shliomis, M.I. Effective viscosity of magnetic suspensions. *Soviet Phys. JETP* **1972**, *34*, 1291.
51. Brenner, H. Antisymmetric stresses induced by the rigid-body rotation of dipolar suspensions. *Int. J. Eng. Sci.* **1984**, *22*, 645.
52. Hynes, J.T.; Kapral, R.; Weinberg, M. Particle rotation and translation in a fluid with spin. *Physica* **1977**, *87A*, 427.
53. Grad, H. Statistical mechanics, thermodynamics, and fluid dynamics of systems with an arbitrary number of integrals. *Comm. Pure Appl. Math.* **1952**, *5*, 455.
54. Condiff, D.W.; Dahler, J.S. Fluid mechanical aspects of antisymmetric stress. *Phys. Fluids* **1964**, *7*, 842.
55. Dahler, J.S. *Research Frontiers. Fluid Dynamics*; Temple, G., Seeger, R., Eds.; Interscience Publishers, Inc.: New York, 1965; Chap. 15.
56. Ailawadi, N.K.; Berne, B.J.; Foster, D. Light scattering from shear waves: the role of angular momentum fluctuations in light scattering. *Phys. Rev. A* **1971**, *3*, 1492.
57. Gershon, N.D.; Oppenheim, I. Hydrodynamic equations for fluids of non spherical molecules. *Physica* **1972**, *62*, 198.
58. Hynes, J.T.; Kapral, R.; Weinberg, M. Molecular theory of translation diffusion: Microscopic generalization of normal velocity boundary condition. *J. Chem. Phys.* **1979**, *70*, 1457.
59. McCoy, B.J.; Sandler, S.I.; Dahler, J.S. Transport properties of polyatomic fluid IV: the kinetic theory of a dense gas of perfectly rough spheres. *J. Chem. Phys.* **1966**, *45*, 3485.
60. Kroh, H.J.; Felderhof, B.U. Electromagnetodynamics of polar liquids and suspensions. *Z. Phys.* **1987**, *B66*, 1.
61. Gast, A.P.; Zukoski, C.F. Electrorheological fluids as colloidal suspensions. *Adv. Colloid Interface Sci.* **1989**, *30*, 153.
62. Halsey, T.C. Electrorheological fluids. *Science* **1992**, *258*, 761.
63. Halsey, T.C.; Martin, J.E. Electrorheological fluids. *Sci. Am.* **1993**, *269*, 58.
64. Rankin, P.J.; Ginder, J.M.; Klingenberg, D.J. Electro- and magneto-rheology. *Curr. Opin. Colloid Interface Sci.* **1998**, *3*, 373.
65. Rodriguez, R.F.; Reyes, J.A.; Manero, O. Model for the electrorheological effect in flowing polymeric nematics. *J. Chem. Phys.* **1999**, *110*, 8197.
66. Rankin, P.J.; Horvath, A.T.; Klingenberg, D.J. Magnetorheology in viscoplastic media. *Rheol. Acta* **1999**, *38*, 471.
67. *Electro-Rheological Fluids, Magneto-Rheological Suspensions and their Applications*; Nakano, M., Koyama, K., Eds.; World Scientific: Singapore, 1998.
68. Frohlich, H. *Theory of Dielectrics*; Oxford U.P.: Oxford, England, 1949.
69. Shliomis, M.I. Magnetic fluids. *Sov. Phys. Usp.* **1974**, *17*, 153.
70. Sellers, H.S.; Brenner, H. Translational and rotational motions of a sphere in a dipolar suspension. *PhysicoChem. Hydrodyn.* **1989**, *11*, 455.
71. Hubbard, J.B.; Stiles, P.J. Hydrodynamics of magnetic and dielectric colloidal dispersions. *J. Chem. Phys.* **1986**, *84*, 6955.
72. Ido, Y.; Tanahashi, T. Nonequilibrium theory of viscoelastic magnetic fluids. *J. Phys. Soc. Jpn.* **1991**, *60*, 466.
73. Rosenweig, R.E. Fluid dynamics and science of magnetic liquids. *Adv. Electron. Electron Phys.* **1979**, *48*, 103.
74. Shizawa, K.; Tanahashi, T. A new complete set of basic equations for magnetic fluids with internal rotation. *Bull. JSME* **1985**, *28*, 1942.
75. Shliomis, M.I.; Lyubimova, T.P.; Lyubimov, D.V. Ferrohydrodynamics. An essay on the progress of ideas. *Chem. Eng. Commun.* **1988**, *67*, 275.
76. Brenner, H.; Condiff, D.W. Transport mechanism in systems of orientable particles III: arbitrary particles. *J. Colloid Interface Sci.* **1972**, *41*, 228.
77. Brenner, H.; Condiff, D.W. Transport mechanism in systems of orientable particles IV: convective transport. *J. Colloid Interface Sci.* **1974**, *47*, 199.
78. del Castillo, L.F.; Dávalos-Orozco, L.A. Dielectric relaxation in polar and viscoelastic fluids. *J. Chem. Phys.* **1990**, *93*, 5147.
79. Dávalos-Orozco, L.A.; del Castillo, L.F. Dielectric relaxation in polar and viscoelastic fluids with internal rotation. *J. Chem. Phys.* **1992**, *96*, 9102.
80. Bird, R.B.; Armstrong, R.C.; Hassager, O. *Dynamics of Polymeric Liquids*; Wiley: New York, 1976; Vol. 1.
81. Shliomis, M.I. Equations of motion of a fluid hydromagnetic properties. *Soviet Physics JETP* **1968**, *26*, 665.
82. Shliomis, M.I.; Raikher, Yu.L. Experimental investigations of magnetic fluids. *IEEE Trans. Magn.* **1980**, *16*, 237.
83. Stiles, P.J.; Hubbard, J.B. Polarization diffusion and dielectric friction in polar liquids. *Chem. Phys.* **1984**, *84*, 431.
84. Stockmayer, W.H. Dielectric dispersion in solutions of flexible polymers. *Pure Appl. Chem.* **1967**, *15*, 539.
85. Pusey, P.M. *Colloidal Suspensions. Liquids, Freezing and Glass Transition*; Levesque, D., Hansen, J.-P., Zinn-Justin, J., Eds.; Elsevier: Amsterdam, 1990.
86. Hansen, J.P.; McDonald, I.R. *Theory of Simple Liquids*; Academic Press: London, 1976.
87. Résibois, P.; De Leener, M. *Classical Kinetic Theory of Fluids*; J. Wiley and Sons: New York, 1977.
88. Kubo, R.; Toda, M.; Hashitsume, N. *Statistical Physics I, Nonequilibrium Statistical Mechanics*; Springer: Berlin, 1985.
89. Jou, D.; Casas-Vázquez, J.; Lebon, G. *Extended Irreversible Thermodynamics*, 2nd Ed.; Springer: Berlin, 1996.
90. del Castillo, L.F.; Dávalos-Orozco, L.A.; García-Colín, L.S. Ultrafast dielectric relaxation response of polar fluids. *J. Chem. Phys.* **1997**, *106*, 2348.
91. Fatuzzo, E.; Mason, P.R. A calculation of the complex dielectric constant of a polar liquid by librating molecule method. *Proc. Phys. Soc.* **1967**, *90*, 729.
92. Leroy, Y.; Constant, E.; Abbar, C.; Desplanques, P. Correlation, relaxation and ultrahertzian absorption in liquids. *Adv. Mol. Relax. Processes* **1967-1968**, *1*, 273.
93. del Castillo, L.F.; García-Colín, L.S. Dielectric relaxation

- thermodynamics: comparison with the experiment. *Phys. Rev. B* **1998**, *37*, 448.
94. Ngai, K.L. Evidences for Universal Behavior of Condensed Matter at Low Frequencies/Long Times. In *Non Debye Relaxation in Condensed Matter*; Ramakrishnan, T.V., Lakshmi, M.R., Eds.; World Scientific: Singapore, 1987.
  95. Williams, G.; Watts, D.C. Non-symmetric relaxation behavior arising from a simple empirical decay function. *Trans. Faraday Soc.* **1970**, *66*, 80.
  96. Palmer, R.G.; Stein, D.L.; Abraham, E.; Anderson, P.W. Models of hierarchically constrained dynamics for glass relaxation. *Phys. Rev. Lett.* **1984**, *53*, 958.
  97. Madden, P.; Kivelson, D. Dielectric friction and molecular reorientation. *J. Phys. Chem.* **1982**, *86*, 4244.
  98. Goulon, J.; Rivail, J.L.; Fleming, J.W.; Chamberlain, J.; Chantry, G.W. Dielectric relaxation and far infrared dispersion in pure liquid chloroform. *Chem. Phys. Lett.* **1973**, *18*, 211.
  99. del Castillo, L.F.; Hernández, S.I.; García-Zabala, A.; Díaz-Calleja, R. Effects of symmetric and antisymmetric stress tensors on N- and  $\alpha$ -dielectric relaxations. *J. Non-Cryst. Solids* **1998**, *235-237*, 667.
  100. Doi, M.; Edwards, S.F. *The Theory of Polymer Dynamics*; Clarendon: Oxford, 1988.
  101. del Castillo, L.F.; Díaz-Calleja, R. Effects of the gradient of polarization on dielectric relaxation. *Physica A* **1999**, *268*, 469.
  102. Baur, M.E.; Stockmayer, W.H. Dielectric relaxation in liquid polypropylene oxides. *J. Chem. Phys.* **1965**, *43*, 4319.
  103. Fulton, R.L. Contributions to the theory of dielectric relaxation in polar media. *J. Chem. Phys.* **1975**, *62*, 4355.
  104. Madden, P.; Kivelson, D. A consistent molecular treatment of dielectric phenomena. *Adv. Chem. Phys.* **1984**, *56*, 467.
  105. Shönhals, A. Relationship of Segmental and Chain Dynamic of Polymers Studied by Broadband Dielectric Spectroscopy. In *Keynote Lectures in Selected Topics of Polymer Science*; Riande CSIC: Madrid, 1996; 41, Chap. II.
  106. Adachi, K.; Kotaka, T. Dielectric normal mode relaxation. *Prog. Polym. Sci.* **1993**, *18*, 585.
  107. Eringen, A.C. Simple microfluids. *Int. J. Eng. Sci.* **1964**, *2*, 205.
  108. Eringen, A.C. Theory of micropolar fluids. *J. Math. Mech.* **1966**, *16*, 1.
  109. Eringen, A.C. Theory of thermomicrofluids. *J. Math. Anal. Appl.* **1972**, *38*, 480.
  110. Eringen, A.C. On nonlocal microfluid mechanics. *Int. J. Eng. Sci.* **1973**, *11*, 291.
  111. Eringen, A.C. The theory of thermo-microstretch fluids and bubbly liquids. *Int. J. Eng. Sci.* **1990**, *28*, 133.
  112. Eringen, A.C. Continuum theory of dense rigid suspensions. *Rheol. Acta* **1991**, *30*, 23.
  113. Eringen, A.C. Memory-dependent orientable nonlocal micropolar fluids. *Int. J. Eng. Sci.* **1991**, *29*, 1515.
  114. Eringen, A.C. Electrodynamics of microstretch and micropolar fluids. *ARI* **1998**, *50*, 169.
  115. Lukaszewicz, G. *Micropolar Fluids: Theory and Applications*; Birkhauser: New York, 1999.
  116. Shliomis, M.I. Hydrodynamics of a liquid with intrinsic rotation. *Soviet Physics JETP* **1967**, *24*, 173.
  117. Kilne, K.A. Structured fluid theory: kinematics of substructure redistribution. *Acta Mechanica* **1977**, *27*, 239.
  118. Pérez-Madrid, A.; Rubí, J.M.; Casas-Vázquez, J. On Brownian motion in fluids with spin. *Physica* **1983**, *A119*, 212.
  119. Pérez-Madrid, A.; Rubí, J.M. On Brownian motion in fluids with spin: II. Viscoelastic models. *Physica* **1984**, *A126*, 152.
  120. Dávalos-Orozco, L.A.; Del Castillo, L.F. Relaxation phenomena in viscoelastic colloidal suspensions with internal rotation. *J. Colloid Interface Sci.* **1996**, *178*, 69.
  121. Faria, S.H.; Kremer, G.M. Some aspects in the thermodynamic theory of polar fluids. *ARI* **1997**, *50*, 105.
  122. Yeremeyev, V.A.; Zubov, L.M. The theory of elastic and viscoelastic micropolar liquids. *J. Appl. Math. Mech.* **1999**, *63*, 755.
  123. Ariman, T.; Turk, M.A.; Sylverster, N.D. Microcontinuum fluid mechanics: a review. *Int. J. Eng. Sci.* **1973**, *11*, 905.
  124. Cowin, S.C. The Theory of Polar Fluids. In *Advances in Applied Mechanics*; Academic Press: New York, 1974; 14, 279.
  125. Stokes, V.K. *Theory of Fluids with Microstructure: An Introduction*; Springer-Verlag: New York, 1984.
  126. Saccomandi, G. Group properties and invariant solutions of plane micropolar flows. *Int. J. Eng. Sci.* **1991**, *29*, 645.
  127. Rao, A.R.; Kasiviswanathan, S.R. A class of exact solutions for the flow of a micropolar fluid. *Int. J. Eng. Sci.* **1987**, *25*, 443.
  128. Chaturani, P.; Narasimman, S. Numerical solution of a micropolar fluid flow between two rotating coaxial disks. *Acta Mechanica* **1991**, *89*, 133.
  129. Prakash, J.; Sinha, P. Lubrication theory for micropolar fluids and its application to a journal bearing. *Int. J. Eng. Sci.* **1975**, *13*, 217.
  130. Sinha, P.; Singh, C. Lubrication of rough surfaces—a microcontinuum analysis. *Int. J. Mech. Sci.* **1982**, *24*, 619.
  131. Tandon, P.N.; Jaggi, S. A polar model for synovial fluid with reference to human joints. *Int. J. Mech. Sci.* **1979**, *21*, 161.
  132. Tandon, P.N.; Rakesh, L. Effects of cartilage roughness on the lubrication of humane joints. *Wear* **1981**, *70*, 29.
  133. Sinha, P.; Singh, C.; Prasad, K.R. Lubrication of human joints—a microcontinuum approach. *Wear* **1982**, *80*, 159.
  134. Tandon, P.N.; Gupta, R.S. Microstructural and cartilage roughness effects on lubrication of human joints. *Indian J. Technol.* **1984**, *22*, 359.
  135. Singh, S.P.; Chadda, G.C.; Sinha, A.K. A model for the micropolar fluid film mechanism with reference to human joints. *Indian J. Pure Appl. Math.* **1988**, *19*, 384.
  136. Sinha, P.; Singh, C.; Prasad, K.R. A microcontinuum

- analysis of the self propulsion of the spermatozoa in the cervical canal. *Int. J. Eng. Sci.* **1982**, *20*, 1037.
137. Philip, D.; Chandra, P. Self-propulsion of spermatozoa in microcontinua: effect of transverse wave motion of channel walls. *Arch. Appl. Mech.* **1995**, *66*, 90.
  138. Ariman, T.; Turk, M.A.; Sylvester, N.D. On steady and pulsatile flow of blood. *J. Appl. Mech.* **1974**, *41*, 1.
  139. Bugliarello, G.; Sevilla, J. Velocity distribution and other characteristics of steady and pulsatile blood flow in fine glass tubes. *Biorheology* **1970**, *7*, 85.
  140. Chaturani, P.; Upadhyaya, V.S. On micropolar fluid model for blood flow through narrow tubes. *Biorheology* **1979**, *16*, 419.
  141. Chaturani, P.; Mahajan, S.P. Poiseuille flow of micropolar fluid with non-zero couple stress at boundary with applications to blood flow. *Biorheology* **1982**, *19*, 507.
  142. Xiexian, L.; Yuansheng, F. An analysis of blood flow in the elliptical venal tube. *Acta Mech. Sinica* **1984**, *16*, 521.
  143. Akay, G.; Kaye, A. Numerical solutions of time dependent stratified two-phase flow of micropolar fluids and its applications to flow of blood through fine capillaries. *Int. J. Eng. Sci.* **1985**, *23*, 265.
  144. Tandon, P.N.; Pal, T.S.; Siddiqui, S.U. Microstructural and peripheral layer viscosity effects on blood flow through a tube with small constriction. *J. Inst. Eng., Part I* **1985**, *65*, 60.
  145. Hogan, H.A.; Henriksen, M. An evaluation of a micropolar model for blood flow through an idealized stenosis. *J. Biomech. Eng.* **1989**, *22*, 211.
  146. Sanyal, D.C.; Maiti, A.K. Microscopic effect on blood flow in a branched artery. *Indian J. Theor. Phys.* **1998**, *46*, 139.
  147. Ramkissoon, H. A paradox in microcontinuum fluid mechanics. *Quart. of Appl. Math.* **1980**, *38*, 357.
  148. Brunn, P. The general solution to the equations of creeping motion of a micropolar fluid and its application. *Int. J. Eng. Sci.* **1982**, *20*, 575.
  149. Ramkissoon, H.; O'Neill, M.E. Stokes force in a Microcontinuum fluid mechanics. *Acta Mechanica* **1983**, *46*, 255.
  150. Avudainayagam, A.; Ramakrishna, J. Faxen's laws for a micropolar fluid. *J. Eng. Math.* **1984**, *18*, 23.
  151. Olmstead, W.E.; Majumdar, S.R. Fundamental Oseen solution for the 2-dimensional flow of a micropolar fluid. *Int. J. Eng. Sci.* **1984**, *21*, 423.
  152. Rao, S.K.L.; Rao, P.B. The oscillations of a sphere in a micropolar fluid. *Int. J. Eng. Sci.* **1971**, *9*, 651.
  153. Ramkissoon, H.; Easwaran, C.V.; Majumdar, S.R. Longitudinal and torsional oscillations of a rod in a polar fluid. *Int. J. Eng. Sci.* **1991**, *29*, 215.
  154. Charya, D.S.; Iyengar, T.K.V. Drag on an axisymmetric body performing rectilinear oscillations in a micropolar fluid. *Int. J. Eng. Sci.* **1997**, *35*, 987.
  155. Hassanien, I.A. Boundary layer flow and heat transfer on a continuous accelerated sheet extruded in an ambient micropolar fluid. *Int. Commun. Heat Mass Transfer* **1998**, *25*, 571.
  156. Bhargava, R.; Takhar, H.S. Numerical study of heat transfer characteristics of the micropolar boundary layer near a stagnation point on a moving wall. *Int. J. Eng. Sci.* **2000**, *38*, 383.
  157. Yerofeyev, V.I.; Soldatov, I.N. A shear surface wave at the interface of an elastic body and a micropolar liquid. *J. Appl. Math. Mech.* **1999**, *63*, 277.
  158. Bhattacharyya, S.P.; Jena, S.K. On the stability of a hot layer of micropolar fluid. *Int. J. Eng. Sci.* **1983**, *21*, 1019.
  159. Bhattacharyya, S.P.; Abbas, M. On the stability of a hot rotating layer of micropolar fluid. *Lett. Appl. Eng. Sci.* **1985**, *23*, 371.
  160. Franchi, F.; Straughan, B. Nonlinear stability for thermal convection in micropolar fluid with temperature dependent viscosity. *Int. J. Eng. Sci.* **1992**, *30*, 1349.
  161. Siddheshwar, P.G.; Pranesh, S. Effect of a non-uniform basic temperature gradient on Rayleigh-Bénard convection in a micropolar fluid. *Int. J. Eng. Sci.* **1998**, *36*, 1183.
  162. Mrabti, A.; Gueraoui, K.; Hihhi, A.; Terhmina, O. Effects of microstructure on natural convection flow of micropolar fluids in a vertical cylinder heated from below. *Int. J. Eng. Sci.* **2000**, *38*, 823.
  163. Hsu, T.-H.; Hsu, P.-T.; Tsai, S.-Y. Natural convection of micropolar fluids in an enclosure with heat sources. *Int. J. Heat Mass Transfer* **1997**, *40*, 4239.
  164. Rao, K.V.R. Thermal instability in a micropolar fluid layer subjected to a magnetic field. *Int. J. Eng. Sci.* **1980**, *18*, 741.
  165. Siddheshwar, P.G.; Pranesh, S. Magnetoconvection on a micropolar fluid. *Int. J. Eng. Sci.* **1998**, *36*, 1173.
  166. H. Stability of laminar flows of micropolar fluids between parallel walls. *Phys. Fluids* **1978**, *21*, 1688.
  167. Hung, C.-I.; Tsai, J.-S.; Chen, C.-K. Nonlinear stability of the thin micropolar liquid film flowing down on a vertical plate. *Trans. ASME J. Fluids Eng.* **1996**, *118*, 498.
  168. Hung, C.-I.; Tsai, J.-S. Rupture of thin micropolar liquid film. *Acta Mech.* **1997**, *122*, 217.
  169. Tözere, A.; Skalak, R. Micropolar fluids as models for suspensions of rigid spheres. *Int. J. Eng. Sci.* **1977**, *15*, 511.
  170. Migun, N.P. On hydrodynamic boundary conditions for microstructural fluids. *Rheol. Acta* **1984**, *23*, 575.
  171. Hogan, H.A.; Henriksen, M. A finite element formulation for laminar flow of a fluid with microstructure. *Int. J. Numer. Methods Fluids* **1991**, *13*, 1267.
  172. Ebert, F. On the measurement of the substance constants of a micropolar fluid (in German). *Rheol. Acta* **1975**, *14*, 258.
  173. Fodemski, T.; Morawski, T. Experimental ascertainment of micropolar effects in a liquid under influence of a rotating electric field (in Polish). *Archiwum Elektrotechniki* **1978**, *27*, 721.
  174. Kolpashchikov, V.L.; Migun, N.P.; Prokhorenko, P.P. Experimental determination of material micropolar fluid constants. *Int. J. Eng. Sci.* **1983**, *21*, 405.
  175. Prokhorenko, P.P.; Migun, N.P.; Grebenshchikov, S.V. Experimental studies of polar indicator liquids used in capillary penetrant testing. *Int. J. Eng. Sci.* **1987**, *25*, 769.

176. Papautsky, I.; Brazzle, J.; Ameal, T.; Frazier, A.B. Laminar fluid behavior in microchannels using micropolar fluid theory. *Sens. Actuators* **1999**, *73*, 101.
177. Bashtovoy, V.G.; Berkovsky, B.M.; Vislovich, A.N. *Introduction to Thermomechanics of Magnetic Fluids*; Hemisphere Publishing Corporation: New York, 1988.
178. Berkovsky, B.M.; Medvedev, V.F.; Krakov, M.S. *Magnetic Fluids: Engineering Applications*; Oxford University Press: Oxford, 1993.
179. *Magnetic Fluids and Applications Handbook*; Berkovskii, B.M., Bashtovoy, V., Eds.; Series of Learning Materials, UNESCO, Begell House Inc.: New York, 1996.
180. Blums, E.; Ceber, A.; Maiorov, M.M. *Magnetic Fluids*; Walter de Gruyter: New York, 1997.
181. Scholten, P.C.; Feliuss, J.A.P. Magnetical and rheological behavior of aggregating magnetic suspensions. *J. Magn. Magn. Mater.* **1990**, *85*, 107.
182. Krueger, D.A. Review of agglomeration in ferrofluids. *IEEE Trans. Magn.* **1980**, *Mag-16*, 251.
183. Kamiyama, S.; Satoh, A. Pipe-flow problems and aggregation phenomena of magnetic fluids. *J. Magn. Magn. Mater.* **1990**, *85*, 121.
184. Zubko, V.I.; Komjak, A.I.; Korobov, V.A.; Khrapovitsky, V.P. Electrical properties of magnetic fluids. *J. Magn. Magn. Mater.* **1990**, *85*, 151.
185. Cowley, M.D.; Rosensweig, R.E. The interfacial stability of a ferromagnetic fluid. *J. Fluid Mech.* **1967**, *30*, 671.
186. Tsebers, A.; Blums, E. Long-range magnetic forces in two-dimensional hydrodynamics of magnetic fluid pattern formation. *Chem. Eng. Commun.* **1988**, *67*, 69.
187. Petit, P.A.; De Albuquerque, M.P.; Cabuil, V.; Molho, P. Evolution of the domain-like pattern in a film of ferrofluid in normal fields. *J. Magn. Magn. Mater.* **1992**, *113*, 127.
188. Okubo, M.; Ishibashi, Y.; Oshima, S.; Katakura, H.; Yamane, R. Interfacial waves of the magnetic fluid in vertical alternating magnetic fields. *J. Magn. Magn. Mater.* **1990**, *85*, 163.
189. Boudovis, A.G.; Puchalla, J.L.; Scriven, L.E. Magneto-hydrostatic equilibria of ferrofluid drops in external magnetic fields. *Chem. Eng. Commun.* **1988**, *67*, 129.
190. Anton, I.; Vékás, L.; Potencz, I.; Suciú, E. Ferrofluid flow under the influence of rotating magnetic fields. *IEEE Trans. Magn.* **1980**, *Mag-16*, 283.
191. Rosensweig, R.E.; Popplewell, J.; Johnston, R.J. Magnetic fluid motion in rotating field. *J. Magn. Magn. Mater.* **1990**, *85*, 171.
192. Piso, M.I. Induced internal rotation in magnetic fluid composites. *Adv. Space Res.* **1998**, *22*, 1265.
193. Helgesen, G.; Skjeltorp, A.T. Experimental investigation of the dynamics of microspheres in ferrofluid. *J. Magn. Magn. Mater.* **1991**, *97*, 25.
194. Lacis, S. Bending of ferrofluid droplet in rotating magnetic field. *J. Magn. Magn. Mater.* **1999**, *201*, 335.
195. Shliomis, M.I.; Morozov, K.I. Negative viscosity of ferrofluid under alternating magnetic field. *Phys. Fluids* **1994**, *6*, 2855.
196. Bacri, J.-C.; Perzynski, R.; Shliomis, M.I.; Burde, G.I. "Negative-viscosity" effect in a magnetic fluid. *Phys. Rev. Lett.* **1995**, *75*, 2128.
197. Rosensweig, R.E. "Negative-viscosity" in a magnetic fluid. *Science* **1996**, *271*, 614.
198. Shliomis, M.I.; Kamiyama, S. Hydrostatics and oscillatory flows of magnetic fluid under a nonuniform magnetic field. *Phys. Fluids* **1995**, *7*, 2428.
199. Larrondo, L.E.; Van de Ven, T.G.M. Magnetoviscoelastic properties of chromium dioxide suspensions. *J. Rheol.* **1992**, *36*, 1275.
200. Choi, H.J.; Kim, C.A.; Kwon, T.M.; Jhon, M.S. Viscosity of magnetic particle suspensions. *J. Magn. Magn. Mater.* **2000**, *209*, 228.
201. Berkovsky, B.; Bashtovoi, V. Instabilities of magnetic fluids leading to a rupture of continuity. *IEEE Trans. Magn.* **1980**, *Mag-16*, 288.
202. Rothert, A.; Richter, R. Experiments on the breakup of a liquid bridge of magnetic fluid. *J. Magn. Magn. Mater.* **1999**, *201*, 324.
203. Schwab, L.; Hildebrandt, U.; Stierstadt, K. Magnetic Bénard convection. *J. Magn. Magn. Mater.* **1983**, *39*, 113.
204. Schwab, L.; Stierstadt, K. Field-induced wavevector-selection by magnetic Bénard convection. *J. Magn. Magn. Mater.* **1987**, *65*, 315.
205. Schwab, L. Thermal convection in ferrofluids under a free surface. *J. Magn. Magn. Mater.* **1990**, *85*, 199.
206. Odenbach, S. Magnetic Bénard convection for short-duration microgravity experiments. *Adv. Space Res.* **1993**, *13*, 105.
207. Odenbach, S.; Gilly, H. Taylor vortex flow of magnetic fluids under the influence of an azimuthal magnetic field. *J. Magn. Magn. Mater.* **1996**, *152*, 123.
208. Kikura, H.; Takeda, Y.; Sawada, T. Velocity profile measurement of the magnetic fluid flow using ultrasonic Doppler method. *J. Magn. Magn. Mater.* **1999**, *201*, 276.
209. Odenbach, S.; Gilly, H.; Lindner, P. The use of magnetic small angle neutron scattering for detection of flow profiles in magnetic fluids. *J. Magn. Magn. Mater.* **1999**, *201*, 353.
210. Fannin, P.C.; Vincent, D.; Massart, G.; Perov, P.; Neveu, S. A study of the frequency dependent susceptibility of a colloidal suspension of manganese ferrite nanoparticles. *Eur. Phys. J.: Appl. Phys.* **1999**, *8*, 247.
211. Hrianca, I.; Malaescu, I.; Claiici, F.; Marin, C.N. The influence of particle concentration in ferrofluids on broadening of the magnetic resonance line. *J. Magn. Magn. Mater.* **1999**, *201*, 126.
212. Fannin, P.C.; Vincent, D.; Noyel, G. On the measurement of the complex susceptibility and permittivity of magnetic fluids by means of two different measurement techniques. *J. Magn. Magn. Mater.* **1999**, *201*, 116.
213. Fannin, P.C.; Kinsella, L.; Charles, S.W. Wide-band complex susceptibility measurements of magnetic fluids as a function of temperature. *J. Magn. Magn. Mater.* **1999**, *201*, 91.
214. Fannin, P.C.; Charles, S.W. AC and DC magnetic meas-

- urements on colloidal suspensions of Co particles. *J. Magn. Magn. Mater.* **1999**, 196-197, 586.
215. Oka, H. Metrology of complex magnetic permeability of magnetic fluid in 1 MHz to 1.8 GHz band. *IEEE Trans. Magn.* **1995**, 31, 4172.
216. Johansson, C.; Hanson, M.; Pedersen, M.S.; Morup, S. Magnetic properties of magnetic liquids with iron-oxide particles-the influence of anisotropy and interactions. *J. Magn. Magn. Mater.* **1997**, 173, 5.
217. Fannin, P.C.; Kinsella, L.; Charles, S.W. The high-frequency complex susceptibility of ferrofluids deduced from fits to lower frequency measurements. *J. Phys. D: Appl. Phys.* **1997**, 30, 533.
218. Hanson, M.; Johansson, C. Interaction effects in the dynamic response of magnetic liquids. *J. Magn. Magn. Mater.* **1991**, 101, 45.
219. Malaescu, I.; Hrianca, I. Relaxation processes of magnetite-based ferrofluids in rf magnetic fields. *J. Magn. Magn. Mater.* **1996**, 157/158, 585.
220. Kötitz, R.; Weitschies, W.; Trahms, L.; Semmler, W. Investigation of Brownian and Néel relaxation in magnetic fluids. *J. Magn. Magn. Mater.* **1999**, 201, 102.
221. Pshenichnikov, A.F. Equilibrium magnetization of concentrated ferrocolloids. *J. Magn. Magn. Mater.* **1995**, 145, 319.
222. Berkovsky, B.M.; Bashtovoi, V.G.; Krakov, M.S. Flow and heat transfer under influence of magnetic fluid coatings. *J. Magn. Magn. Mater.* **1990**, 85, 190.
223. Bailey, R.L. Lesser known applications of ferrofluids. *J. Magn. Magn. Mater.* **1983**, 39, 178.
224. Anton, I.; De Sabata, I.; Vékás, L. Application orientated researches on magnetic fluids. *J. Magn. Magn. Mater.* **1990**, 85, 219.
225. Charles, S.W. Some applications of magnetic fluids-use as an ink and in microwave systems. *J. Magn. Magn. Mater.* **1987**, 65, 350.
226. Shimada, K.; Kamiyama, S.; Iwabuchi, M. Effect of a magnetic field on the performance of an energy conversion system using magnetic fluid. *J. Magn. Magn. Mater.* **1999**, 201, 357.
227. Krakov, M.S. Influence of rheological properties of magnetic fluid on damping ability of magnetic fluid shock-absorber. *J. Magn. Magn. Mater.* **1999**, 201, 368.
228. Piso, M.I. Applications of magnetic fluids for inertial sensors. *J. Magn. Magn. Mater.* **1999**, 201, 380.
229. Calarasu, D.; Cota, C.; Olaru, R. Magnetic fluid brake. *J. Magn. Magn. Mater.* **1999**, 201, 401.
230. Jordan, A.; Scholz, R.; Wust, P.; Föhling, H.; Felix, R. Magnetic fluid hyperthermia: cancer treatment with AC magnetic field induced excitation of biocompatible superparamagnetic nanoparticles. *J. Magn. Magn. Mater.* **1999**, 201, 413.
231. Hiergeist, R.; Andrä, W.; Buske, N.; Hergt, R.; Hilger, I.; Richter, U.; Kaiser, W. Application of magnetic ferrofluids for hyperthermia. *J. Magn. Magn. Mater.* **1999**, 201, 420.