Collective motion in ferrofluids

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Abstract. Collective motion in a ferrofluid is discussed on the basis of ferrohydrodynamics. Simple examples elucidate the effect of a collection of spinning suspended particles on Stokes flow. Spin viscosity emerges as a crucial transport coefficient. In the case of a uniform rotating magnetic field applied to a ferrofluid in a cylinder or a spherical cavity the collective fluid motion is driven by the curl of the torque density, which differs from zero only in a thin boundary layer with thickness proportional to the square root of the spin viscosity.

1. Introduction
Ferrofluids find use in many technical applications[1]. Recent work shows that effective pumping of a ferrofluid by the application of a running magnetic wave is possible[2]. No doubt, this will lead to interesting novel applications. The corresponding theory is being developed, and gives rise to new questions and problems.

Ferrofluids have the important property that one can act on them with a magnetic field, since that couples to the magnetic dipole moments of suspended particles. The theoretical description is based on an extension of the Navier-Stokes equations, with account of the magnetic force and torque density acting on the fluid. Collective motions of the fluid can be generated by judicious application of a field which varies in space and time.

The equations of motion are complicated, since they couple the flow velocity and the rotational velocity of spinning particles to magnetic field and magnetization in a nonlinear way. Magnetic field and magnetization satisfy Maxwell’s equations of magnetostatics, and this gives rise to long range effects, so that confinement and geometry crucially affect fluid behavior. The relaxation of magnetization to its thermal equilibrium value is described by a phenomenological relaxation equation, which involves a nonlinear coupling to flow velocity and spin velocity.

The hydrodynamic stress tensor of a ferrofluid is not symmetric[1], and involves the transport coefficients vortex viscosity $\zeta$ and spin viscosity $\eta'$, besides the shear viscosity $\eta$. The vortex viscosity can be estimated fairly well for a dilute suspension as $\zeta = \frac{3}{2}\phi\eta$, where $\phi$ is the volume fraction of suspended particles. The spin viscosity is more problematic. We have suggested elsewhere[3], that it can possibly be determined in computer simulation from its effect in simple flow situations.

In the following we discuss first how a torque density can generate flow in a viscous incompressible fluid described by the Stokes equations. Subsequently we recall the equations of ferrohydrodynamics, and review recent work on the rotation of a ferrofluid, confined in a cylinder or a sphere, caused by the application of a uniform rotating magnetic field.
2. Torque density acting on a viscous incompressible fluid

In this section we consider a viscous incompressible fluid and study the flow caused by an applied torque density in some simple situations. The torque density may be caused by a rotating magnetic field acting on magnetic particles with permanent magnetic dipole moment. For the time being we neglect the interaction between dipole moments. We consider only phenomena on a slow timescale and a small length scale, so that we may use the equations of low Reynolds number hydrodynamics\[4\]. Thus we consider the steady-state Stokes equations

\[
\eta \nabla^2 v - \nabla p = -F, \quad \nabla \cdot v = 0,
\]

where \(v(r)\) is the flow velocity, \(p(r)\) is the pressure, and \(F(r)\) is the force density acting on the fluid via suspended particles. It is useful to express the force density, like the charge density in electrostatics, in terms of a multipole expansion about the particle centers\[5\]

\[
F(r) = F_1(r) - \nabla \cdot F_s^2(r) + \frac{1}{2} \nabla \times T(r) + \ldots,
\]

where \(F_1(r)\) is the monopole force density, \(F_s^2(r)\) is the symmetric force dipole density, and \(T(r)\) is the torque density, which may also be regarded as the antisymmetric force dipole density. In the absence of applied forces the monopole force density vanishes. The symmetric force dipole density is induced by local velocity gradients, and leads to an effective viscosity. Higher order force multipole densities will be neglected. Thus we consider instead of Eq. (2.1) the equations

\[
\eta \nabla^2 v - \nabla p = -\frac{1}{2} \nabla \times T, \quad \nabla \cdot v = 0,
\]

where now \(\eta\) is the viscosity of the suspension, rather than that of the pure fluid. Due to the presence of the curl operator, regions of uniform torque density cause no flow, and only the spatial variation of the torque density matters.

For a uniform torque density \(T(r) = Te_y\) in a planar layer \(-L < z < L\) in infinite fluid one finds a flow in the \(x\) direction given by

\[
v_x(z) = \begin{cases} 
U, & L < z < \infty, \\
Uz/L, & -L < z < L, \\
-U, & -\infty < z < -L, 
\end{cases}
\]

with \(U = LT/2\eta\). This shows that in the absence of a confining boundary a uniform but localized torque density acts like a pump on the fluid. One can visualize the flow as being generated by spinning particles in the layer. A more complete description including the spinning particles leads to a smooth flow profile for non-vanishing spin viscosity\[3\]. Two parallel layers of opposite and equal torque density generate a uniform plug flow between the two layers. The question is whether a collective motion of this type can be used for practical purposes. As shown in the next two examples, confinement tends to annihilate the flow.

Consider first a fluid confined to a spherical cavity of radius \(R\) with no-slip boundary condition at the wall of the cavity. We choose the origin at the center of the cavity and use spherical coordinates \((r, \theta, \varphi)\). A uniform torque density is applied in the spherical region \(0 < r < c\), with \(c < R\),

\[
T(r) = Te_z \Theta(c - r),
\]

where \(\Theta(x)\) is the Heaviside step-function. The torque density has an effect only at \(r = c\), since

\[
\nabla \times T(r) = Te_\varphi \delta(r - c).
\]
One solves the flow problem by putting
\[ \mathbf{v}(r) = f(r) \sin \theta \, \mathbf{e}_\varphi, \quad p(r) = p_0, \] (7)
with scalar function \( f(r) \). Substituting one finds
\[ f(r) = Wr, \quad 0 < r < c, \]
\[ = Ar + Br^{-2}, \quad c < r < R, \] (8)
with coefficients \( W, A, B \). Solving for the coefficients from the jump conditions at \( r = c \) and the boundary condition at \( r = R \) one finds in particular for the coefficient \( W \)
\[ W = \frac{R^3 - c^3 T}{3R^3} \frac{1}{\eta}. \] (9)
This shows that the rotational velocity of the inner sphere \( r < c \) tends to zero as \( c \to R \) due to friction with the wall. For small gap \( R - c \) one has approximately
\[ W \approx (R - c) \frac{T}{\eta R}, \] (10)
with angular velocity proportional to the width of the gap.

Similarly, for a cylinder of radius \( R \) with no-slip condition at the wall of the cylinder one introduces cylindrical coordinates \((r, \varphi, z)\). Assuming a torque density of the form Eq. (2.5) one solves the flow problem by putting
\[ \mathbf{v}(r) = f(r) \, \mathbf{e}_\varphi, \quad p(r) = p_0, \] (11)
with scalar function \( f(r) \) given by
\[ f(r) = Wr, \quad 0 < r < c, \]
\[ = Ar + Br^{-1}, \quad c < r < R. \] (12)
This leads to the coefficient \( W \)
\[ W = \frac{R^2 - c^2 T}{2R^2} \frac{1}{\eta}. \] (13)
For small gap this is again approximated by Eq. (2.10).

3. Ferrohydrodynamics
In practical applications a torque density can be generated by an electric field acting on a permanent electrical dipole moment, or by a magnetic field acting on a permanent magnetic moment. There must be an angle between field and dipole moment. This can be achieved for example by acting with a rotating field. Then due to dissipation there will be a lag between field and moment, and hence a non-vanishing torque density. We consider the magnetic case. Then one must use the equations of ferrohydrodynamics[1]. The long range interactions between dipole moments are described on the macroscopic level by Maxwell’s equations. The lag between magnetization and magnetic field must be found from a magnetic relaxation equation.

In ferrohydrodynamics Stokes’ equations are generalized to
\[ \nabla \cdot (\mathbf{\sigma}_{hyd} + \mathbf{\sigma}_m) = 0, \quad \nabla \cdot \mathbf{v} = 0, \quad \nabla \cdot \mathbf{\omega}_p = 0, \] (14)
where $\sigma_{\text{hyd}}$ is the hydrodynamic stress tensor, given in cartesian component form by

$$
\sigma_{\text{hyd},\alpha\beta} = -p \delta_{\alpha\beta} + \eta \left( \frac{\partial v_\alpha}{\partial x_\beta} + \frac{\partial v_\beta}{\partial x_\alpha} \right) + \zeta \varepsilon_{\alpha\beta\gamma}(\nabla \times v - 2\omega_p)_{\gamma},
$$

(15)

where $\varepsilon_{\alpha\beta\gamma}$ is the Levi-Civita tensor, $\omega_p$ is the spin velocity of suspended particles, and $\sigma_m$ is the Maxwell stress tensor. The latter is given by

$$
\sigma_m = BH - \frac{1}{2}\mu_0 H^2 I,
$$

(16)

in SI units, where $B$ is the magnetic induction, $\mu_0$ is the magnetic permeability of vacuum, $H$ is the magnetic field, and $I$ is the unit tensor. Induction and field are related by

$$
B = \mu_0 (H + M),
$$

(17)

where $M$ is the magnetization, and are assumed to satisfy Maxwell’s equations of magnetostatics,

$$
\nabla \cdot B = 0, \quad \nabla \times H = j.
$$

(18)

where $j$ is the externally applied current density. The divergence of the Maxwell stress tensor yields both a magnetic force density and a magnetic torque density, acting on the fluid. The first equation (3.1) is the equation of motion for momentum, with neglect of inertial terms. We must also consider the equation of motion for the spin velocity $\omega_p$, or equivalently for the spin per unit mass $S = I\omega_p$, where $I$ is a moment of inertia per unit mass, with dimension $a^2$, where $a$ is a radius of gyration. The equation of motion for spin reads

$$
\rho \frac{dS}{dt} = 2\zeta(\nabla \times v - 2\omega_p) + \mu_0 M \times H + \eta' \nabla^2 \omega_p,
$$

(19)

where $\rho$ is the mass density and $\eta'$ is the spin viscosity, which may be interpreted as a spin diffusion coefficient. The second term on the right is the magnetic torque density. On a slow time scale we may drop the inertial terms on the left, like in the momentum equation, and are then left with

$$
\eta' \nabla^2 \omega_p + 2\zeta(\nabla \times v - 2\omega_p) + \mu_0 M \times H = 0.
$$

(20)

Substituting this into Eq. (3.2) we find that the first equation (3.1) may be rewritten as

$$
\eta' \nabla^2 v - \nabla p = -F^s_m - \frac{1}{2}\eta' \nabla \times (\nabla^2 \omega_p),
$$

(21)

where $F^s_m$ is the magnetic force density, which follows from the symmetric part of the Maxwell stress tensor as

$$
F^s_m = \nabla \cdot \sigma^s_m = \mu_0 M \cdot (\nabla H) + \frac{1}{2}\mu_0 \nabla \times (M \times H).
$$

(22)

The first term on the right is the Kelvin force density. The second term may be combined with the last term in Eq. (3.8) to yield a torque density $\mu_0 M \times H + \eta' \nabla^2 \omega_p$, as in Eq. (2.2). Thus the equation of motion (3.8) for the flow velocity $v$ is quite similar to the equation we have studied in the preceding section.

The equations of motion (3.7) and (3.8), together with the condition of incompressibility $\nabla \cdot v = 0$, must be supplemented with a constitutive equation for the magnetization. In practice one uses a relaxation equation with a single relaxation time of the form

$$
\frac{\partial M}{\partial t} + v \cdot \nabla M - \omega_p \times M = -\gamma(M - M_{eq}(H)),
$$

(23)
where $\gamma$ is a relaxation rate, and $M_{eq}(H)$ is the equilibrium magnetization corresponding to the local magnetic field according to the equilibrium equation of state at the local temperature. Heat conduction is assumed to be fast, so that the temperature can be taken to be uniform. The relaxation rate $\gamma$ can be identified with $1/\tau$, where $\tau$ is the relaxation time for rotation of a particle. The suspension is usually polydisperse, and the longest relaxation time, corresponding to the largest particles will dominate. The actual relaxation process will be more complicated, but Eq. (3.10) captures the main features, and is suitable for exploratory calculations. The equation introduces a time-dependence into the problem. Moreover it is nonlinear, so that a perturbation method must be used.

4. Collective motion of a ferrofluid

In a ferrofluid one can generate collective motion by the action of a time-dependent applied field. It is convenient to take the field to be periodic in time, and to ask for a systematic collective motion of the fluid on time average. The early experiments and theory have been concerned with rotation of a ferrofluid in a cylindrical vessel under the influence of a rotating magnetic field. Later, pumping by a running wave magnetic field in planar or cylindrical geometry has been considered.

Rotation of a ferrofluid in a cylindrical vessel by means of a rotating magnetic field was achieved first by Rosensweig and Moskowitz[6]. A theoretical explanation of bulk rotational motion was provided by Zaitsev and Shliomis[7]. They found that the ferrofluid rotates with the field at a rate proportional to the magnetic field. Experimentally the situation is confused. Brown and Horsnell[8] repeated the experiment, and found rotation in the opposite direction. It was realized later that in experiments with an open cylinder, the coupling to a surface wave dominates, and that one can get both directions of rotation, depending on whether the surface is convex or concave[9],[10].

More recently, experiments with a closed cylinder have been performed[11],[12], in order to eliminate the surface wave effect. On the basis of these experiments Chaves et al. concluded to a value of the spin viscosity of about $\eta' \approx 10^{-12} - 10^{-8}$ Ns. This is much larger than Rosensweig’s earlier estimate $\eta' \approx \eta d^2$, where $d$ is the mean distance between suspended particles. The estimate leads to a value $\eta' \approx 10^{-18}$ Ns at volume fraction $\phi = 0.1$, much too small to be compatible with observed rotation rates.

As a test, the rotation experiment has been repeated with a closed sphere filled with ferrofluid[13]. Surprisingly, the fluid did not rotate noticeably in the rotating field. A theoretical estimate with the value of the spin viscosity found by Chaves et al. leads to a rotation rate which should have been observable. Khushrushahi and Zahn[13] conclude that the earlier analysis[12] led to an incorrect value due to neglect of a non-uniform demagnetization field. In the analogous situation of an electrically polar liquid Bonthuis et al.[14] estimated the spin viscosity as $\eta' \approx \eta a^2$, where $a$ is a molecular radius. This agrees with computer simulation results for nonpolar molecular liquids of Evans and Streett[15]. Hansen et al.[16] found, on the basis of molecular dynamics simulations and a theoretical expression of the spin viscosity as the integral of a time-correlation function, a value for $\eta'$ two orders of magnitude larger than this estimate. This suggests that the spin viscosity is affected significantly by long range dipolar interactions.

Further theoretical analysis of the rotation experiment has been based on the equations of the preceding section. As a starting point one performs perturbation theory in the amplitude of the applied field. It is assumed that the applied field and the resulting magnetization are much smaller than the saturation magnetization of the ferrofluid, so that the equilibrium of state in Eq. (3.10) may be linearized as $M_{eq}(H) \approx \chi_0 H$, where $\chi_0$ is the initial susceptibility. We consider a uniform rotating applied field with $x,y$ components

$$H_{1x}(t) = A \cos \omega t, \quad H_{1y} = A \sin \omega t,$$

(24)
where the subscript 1 indicates that the field is linear in the amplitude $A$. A perturbation analysis in powers of the amplitude $A$ then takes the form

\[
\begin{align*}
H &= H_1 + H_3 + ..., \\
M &= M_1 + M_3 + ..., \\
v &= v_2 + v_4 + ..., \\
p &= p_0 + p_2 + p_4 + ..., \\
\omega_p &= \omega_{p2} + \omega_{p4} + ...
\end{align*}
\]  

(25)

where the subscript denotes the power of $A$. To lowest order one evaluates $(M_1, H_1, v_2, \omega_{p2})$. The field $H_1 = (H_{1x}, H_{1y}, H_{1z})$ is given by Eq. (4.1). The magnetization $M_1$ is harmonic as well, but lags the field,

\[
M_{1x} = A \cos(\omega t - \alpha), \quad M_{1y} = A \sin(\omega t - \alpha).
\]  

(26)

The lag angle $\alpha$ is expressed in terms of the complex susceptibility $\chi(\omega) = \chi'(\omega) + i\chi''(\omega)$ as

\[
\alpha = \arctan \frac{\chi''(\omega)}{\chi'(\omega)}.
\]  

(27)

The susceptibility follows from Eq. (3.10) and

\[
M_{1\omega} = \chi(\omega)H_{1\omega}
\]

as

\[
\chi(\omega) = \frac{\gamma \chi_0}{\gamma - i\omega}.
\]  

(28)

We use complex notation with time-factor $\exp(-i\omega t)$. Hence the lag angle is

\[
\alpha = \arctan \frac{\omega}{\gamma}.
\]  

(29)

In the experiment one has $\omega << \gamma$, so that $\alpha \approx \omega/\gamma$. To second order the magnetic torque density is

\[
T_2 = \mu_0 M_1 \times H_1.
\]  

(30)

This is uniform and independent of time

\[
T_2 = T_{22} e_z, \quad T_{22} = \mu_0 \chi''(\omega) A^2,
\]  

(31)

with

\[
\chi''(\omega) = \chi_0 \frac{\omega \gamma}{\omega^2 + \gamma^2} \approx \chi_0 \frac{\omega}{\gamma}.
\]  

(32)

The torque density vanishes at zero frequency, since then the lag angle $\alpha$ vanishes.

To second order Eqs. (3.7) and (3.8) become

\[
\begin{align*}
\eta \nabla^2 v_2 - \nabla p_2 &= -\frac{1}{2} \eta' \nabla \times (\nabla^2 \omega_{p2}), \\
\eta' \nabla^2 \omega_{p2} + 2\zeta (\nabla \times v_2 - 2\omega_{p2}) &= -T_2.
\end{align*}
\]  

(33)

The force density $F_{m2}^s$ vanishes to this order, since $M_1$ and $H_1$ are uniform. The equations can be solved for spherical geometry, and lead to steady-state flow velocity $v_2$ and spin velocity $\omega_{p2}$. For a cylinder the solution is similar to that of Zaitsev and Shliomis [7], but differs because these authors used the unsatisfactory approximation $M_{eq}(H) \approx M_s H/H$, where $M_s$ is the saturation magnetization.

For a sphere the flow velocity $v_2(r)$ takes the form (2.7), and for a cylinder it takes the form (2.11). Correspondingly, for a sphere the spin velocity takes the form

\[
\omega_{p2}(r) = \nabla \times (g(r) \sin \theta e_\varphi),
\]  

(34)
with radial function \( g(r) \), whereas for a cylinder one has simply
\[
\omega_{p2}(r) = g(r)e_z. \tag{35}
\]

As an example, for a sphere the solution takes the form
\[
f(r) = Wr + XF(\kappa r), \quad g(r) = \frac{T_2}{8\zeta} + \frac{1}{2}Wr + \frac{\eta + \zeta}{2\zeta}XF(\kappa r), \tag{36}
\]
with the function
\[
F(x) = \frac{\cosh x}{x} - \frac{\sinh x}{x^2}, \tag{37}
\]
and inverse screening length
\[
\kappa = \sqrt{\frac{4\eta\zeta}{\eta(\eta + \zeta)}}. \tag{38}
\]
The coefficients \( W \) and \( X \) are determined from the boundary conditions \( v = 0 \) and \( \omega_{p2} = 0 \) at \( r = R \).

For a cylinder the solution of Eqs. (3.7), (3.8), and (3.10) can be carried out to all orders in the amplitude \( A \) in a numerical iterative scheme in a first harmonic approximation, keeping only steady-state terms and first harmonics, and with linearized equilibrium equation of state\[18\]. The second order perturbation theory result is used as a starting point in the iteration. Surprisingly, one finds that for a large amplitude the solution bifurcates into two solutions with rotation in opposite directions. Experimentally this corresponds to flow reversal at strong applied field, since the solution with rotation opposite to the field has the lower dissipation. Such flow reversals have been observed experimentally, but in some experiments the surface wave phenomenon may have dominated.

5. Discussion
For simplicity we have restricted attention to a rotating applied magnetic field of the form (4.1). This is the simplest example of collective motion generated by a varying applied magnetic field. In order to achieve pumping of a ferrofluid more complicated variations in space and time will be required. Solutions which have been considered involve plane magnetic waves running along a planar duct or a circular tube\[19\]-\[21\]. It will be important to study other situations, and find improved ways of technical interest.

The spin diffusion described by the transport coefficient spin viscosity leads to a smooth boundary layer. Theoretical estimates of the spin viscosity and the experiment of Khushrushahi and Zahn\[13\] suggest that the boundary layer is quite thin, but more work on the value of the transport coefficient is desirable.

The boundary condition used for the spin velocity is not well established. It is known that a single particle, confined in a spherical cavity filled with viscous incompressible fluid and subjected to a constant torque, shows circular orbital motion, with reversal of direction for orbits close to the surface\[22\]. It would be of interest to find the motion of a small number of particles confined to a spherical cavity or a circular tube by Stokesian dynamics simulation. The transition to collective motion and the behavior near the boundary may provide information on the role of spin viscosity and the proper boundary condition to be used in ferrohydrodynamics.

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