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Determination of the constitutive equations for a magnetic fluid

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The principal aim of this paper is to derive constitutive equations describing a magnetic fluid. The fluid is modelled as a dilute suspension of small spheroidal magnetic particles in a non-magnetic solute. The conditions for stability of fluid (against coagulation) are discussed and upper and lower bounds for particle sizes are determined. For a stationary fluid, the bulk magnetization is calculated with allowance for particle–particle interactions. The full stress tensor is determined for a flowing fluid that experiences an externally applied magnetic field. Both the flow and field may have arbitrary spatial and time dependences provided only that the lengthscale of spatial variations is large compared with particle dimensions, and that the timescale is long compared with the particle relaxation time due to Brownian motion. These results are applied to shear and pipe flows, where comparison with experiment is made, and to flow induced by rotating magnetic fields. Finally the damping of ultrasound having a characteristic period of the same order as the particle relaxation time is considered.

1. Introduction

In this paper, we calculate the stress tensor for a dilute monodisperse suspension of spheroidal magnetic particles, for which the Reynolds number based on particle size a is small. A variety of particle shapes is permitted, ranging from disks and spheres to rods. It may be shown that, even for a suspension of arbitrary-shaped particles, the tensorial character of the particle stress is the same as that predicted here, and so it should be representative of a real magnetic fluid. The result is then applied to some simple and practically important flow problems including some in which the appearance of a non-symmetric stress tensor has caused much confusion in the past.

Magnetic fluids have received some theoretical attention previously; Brenner & Weissman (1972) calculated the stress tensor for a suspension of *spherical* magnetic particles in the limit of weak magnetic field (and our calculation agrees in this case). This stress tensor is deficient, however, because for spheres there is no coupling between the ambient straining motion and particle rotation.

Schliomis (1968, 1972), by means of a phenomenological approach, calculated the stress tensor for a general time-dependent flow and magnetic field, by including the magnetic effects in the Navier–Stokes equation as a modified pressure and viscosity, with, in addition, two terms representing the bulk force and couple on the fluid due to the magnetic field. This formulation gives rise to a very special form of the stress tensor (not unlike that for spherical particles). But even in this case there are inconsistencies; for example, gradients of the magnetic field are included in the bulk

force, but the contribution to the stress from the time-dependence of the magnetic field as seen by moving particles is neglected.

Martsenyuk (1973) used a semiquantitative approach to calculate the stress tensor for a magnetic fluid of spheroidal particles under the influence of a homogeneous magnetic field, but made several *ad hoc* assumptions about the form of the particle orientational probability function, and in addition neglected the crucially important diffusion stress (this same approximate technique has also been used by Martsenyuk, Raikher & Schliomis 1973). The results of Martsenyuk (1973), when applied to spherical particles, are within 20 % of the exact solution. However, for non-spherical particles, where the coupling between rotary and straining motions is important, the results are incorrect even for zero magnetic field.

We consider a suspension of identical spheroidal magnetic particles, in a fluid that is assumed to be Newtonian, non-conducting and non-magnetic, concerning ourselves only with particles that are magnetic dipoles (i.e. the model presented neglects quadrupole and higher magnetic moments). Hydrodynamic interactions between particles will be neglected throughout, which gives an adequate approximation, since the volume concentration c of particles is in practice of the order of 10 %. Magnetic interactions are also neglected, except in §3 where they are investigated for a static magnetic fluid. By an order-of-magnitude argument, Rallison (1978) showed that rotational effects on the suspended particles are very much greater ($O(L/a)$, where L is a macroscopic lengthscale) than translational ones, so only orientational statistics will be considered.

This work begins (§2) with a discussion of the physical mechanisms in a magnetic fluid and the introduction of a 'standard magnetic suspension'. We then (§3) calculate the bulk magnetization of a magnetic fluid in the limit of small volume concentration correct to $O(c^2)$, and in §4 determine the full particle stress tensor, including the effects of time-dependent and inhomogeneous magnetic fields. Two applications of the theory are given in §§5 and 6, in which the stress tensor is applied to the industrially important problem of shear and pipe flows, and to flows induced by rotating magnetic fields. Finally, in §7 we investigate the theory and applications of the propagation of ultrasound through a magnetic fluid where the characteristic period of the signal is of the same order as the Brownian relaxation time.

2. Physical mechanisms in a magnetic fluid

2.1. *Stability*

For a magnetic fluid to be practically useful, it must be stable; that is, it must not separate out by the formation of aggregates during the experiment, or the working life of the sample. This imposes strong restrictions on the acceptable values of interparticle forces, and so on the particle size distribution.

Particles interact directly by three mechanisms: steric repulsion, van der Waals attraction and magnetic forces. The steric repulsion is an 'osmotic'-like barrier, due to a thin coat of surfactant. This is applied to prevent the van der Waals forces from generating aggregates; both these forces are short range (of the order of a few nanometres), the range of the van der Waals forces necessarily being the shorter for a stable magnetic fluid.

An estimate for the maximum allowable particle size can be made by insisting that the magnetic interactions should be too weak to cause aggregation. The particles of a typical magnetic fluid have $O(1)$ aspect ratios (the ratio of maximum to minimum dimensions) and so the interaction energy of two particles is dominated by the

dipole–dipole interaction. So, for the purposes of estimating the magnetic interaction energy $\mathscr{E}$ of two particles, we may take them to be uniformly magnetized spheres, giving

$$\mathscr{E} = \frac{1}{4\pi\mu_0 r^3}(\boldsymbol{m}_1 \cdot \boldsymbol{m}_2 - 3(\boldsymbol{m}_1 \cdot \hat{\boldsymbol{r}})(\boldsymbol{m}_2 \cdot \hat{\boldsymbol{r}})),$$

where $\boldsymbol{m}_1$, $\boldsymbol{m}_2$ are the magnetic dipole moments of the two particles, having separation r in direction $\hat{\boldsymbol{r}}$, and μ_0 is the magnetic permittivity of a vacuum. Let $\mathscr{E}_0$ be the energy required to separate two cold particles (i.e. neglecting Brownian-motion effects) from their minimum energy configuration (i.e. touching nose to tail) to infinite separation. For simplicity we assume that each particle has volume V_0 and internal magnetization density I; then

$$\mathscr{E}_0 = \frac{I^2 V_0}{12\mu_0}.$$

So, if Brownian agitation is to be sufficiently strong to keep the particles apart, it is required that $\mathscr{E}_0$ be less than or of the order of kT (where k is Boltzmann's constant, and T the absolute temperature).

For a magnetic fluid to be considered as a continuum, it is necessary that one phase does not move significantly relative to the other while it is in use. However, a 'typical' magnetic field $O(kt/IV_0)$, which in a practical sense for magnetic fluids is one that induces a significant magnetization (to be defined in §2.2), varying on a lengthscale L, will eventually give rise to a particle distribution varying on the same lengthscale. Making a simple estimate of the drift velocity v_{drift} by consideration of a spherical magnetic particle, we obtain

$$v_{\text{drift}} = O\left(\frac{kT}{6\pi\mu a}\frac{1}{L}\right),$$

where μ is the viscosity of the solvent. The time for the number density of particles to vary appreciably in an $O(L)$ distance is thus $O(6\pi\mu a L^2/kT)$, and for a 'standard' magnetic fluid (as will be defined in §2.2) $kT/6\pi\mu a$ is of the order of 5×10^{-11} m^2 s^{-1}, so, with L of the order of 1 cm, this time is of the order of one month, showing that, for all practical or laboratory timescales, the slippage may be neglected and a magnetic fluid that is initially homogeneous will remain so.

2.2. *Orders of magnitude*

It is helpful at this stage to define a 'standard magnetic suspension', which is typical of those used in experiments. Then, in the rest of this paper it is understood that any reference to dimensional quantities will be for our standard suspension, unless explicitly stated otherwise. A common magnetic solute used is magnetite, having magnetization 0.5 T, and most other solutes used are of comparable magnetization, so we shall take this as the particle magnetization (excluding the surfactant layer). We take the ambient fluid to be water, as it often is, and assume the system is at 'room temperature' (the exact value not being very important). If the surfactant layer is assumed to be 2 nm thick, the stability condition $W_0 \equiv \mathscr{E}_0/kT \lesssim 1$ is satisfied for particles of typical dimension 10 nm, which we shall take as the standard size. Another important parameter is the volume fraction c of particles (including surfactant layer) which is in the range $0.05 < c < 0.25$ for most magnetic fluids.

From a mathematical point of view, it is convenient to assume that all the particles are the same shape and size, and, although in practice there is a size distribution of particles, effects due to this polydispersivity have relatively minor physical importance. Further, we shall assume that the particles (including the surfactant layer) are spheroidal with axis ratio r, and with magnetization fixed in the direction of the axis

of symmetry. Some justification is possible for the latter from considerations of the crystal structure, and, dynamically, particle asymmetries are most significant in this case. However, it does imply a lower bound on particle size, typically of the order 1 nm, above which the thermal fluctuations of the direction of the magnetic dipole moment relative to the particle may be ignored.

In §2.1, we discussed the balance of the magnetic interaction between individual particles and Brownian motion; we now consider the balance between an externally applied magnetic field $\boldsymbol{H}$ and Brownian motion, so far as it affects particle orientation. The energy of interaction between a particle and an external magnetic field is $-\boldsymbol{m}\cdot\boldsymbol{H}$, where $\boldsymbol{m}$ is the magnetic dipole moment of a particle with magnitude $V_0 I$. Thus, for the magnetic field to have a significant orientating effect, this interaction energy must be of the order kT. This suggests that we should non-dimensionalize $\boldsymbol{H}$ with respect to kT/m, where $m = |\boldsymbol{m}|$, and take the non-dimensionalized dipole moment to be a unit vector in the direction of the internal magnetization. When $|\boldsymbol{H}| = 1$, this gives a magnetic field of 2×10^4 A/m (250 gauss) for our standard suspension. This is, in practice, a strong magnetic field, but much stronger fields are experimentally feasible.

We shall see in §4 that the tensorial quantity $\boldsymbol{A}$ describing the effect of viscous forces on particle orientation is not simply $\nabla\boldsymbol{u}$ (where $\boldsymbol{u}$ is the fluid velocity), but rather $\boldsymbol{\Omega} + [(r^2-1)/(r^2+1)]\boldsymbol{E}$ (where $\boldsymbol{\Omega}$ and $\boldsymbol{E}$ are respectively the symmetric and antisymmetric parts of $(\nabla\boldsymbol{u})^{\mathrm{T}}$). The relative importance of viscous and Brownian forces on particle orientations is described by the Péclet number $\mathbb{P}$. We thus define the Péclet number by

$$\mathbb{P} = \frac{\|\nabla\boldsymbol{u}\|}{D},$$

where $\|\nabla\boldsymbol{u}\|$ is the magnitude of a typical element of $\nabla\boldsymbol{u}$, and D is the rotational diffusivity of particles due to Brownian motion. We also non-dimensionalize $\boldsymbol{\Omega}$ and $\boldsymbol{E}$ with respect to $\|\nabla\boldsymbol{u}\|$, and all stresses with respect to $\mu\|\nabla\boldsymbol{u}\|$. As D is of the order 10^6 s^{-1}, the Péclet number is small for most flows of practical interest, suggesting that we non-dimensionalize time with respect to D.

3. The bulk magnetization

Each particle experiences a couple $\boldsymbol{m} \times \boldsymbol{H}$ due to the local magnetic field, which tends to align the particles with it, and a disorienting Brownian couple. The balance between the two is a function of $|\boldsymbol{H}|$, and the degree of overall alignment gives rise to the bulk magnetization, $\boldsymbol{M}$.

There is a difficulty in calculating $\boldsymbol{M}$, in that the magnetic field as seen by a particle is not simply the ambient magnetic field, but also includes a contribution due to particle–particle interactions. Assuming a homogeneous suspension $\boldsymbol{M} = n\langle\boldsymbol{m}\rangle$, where n is the particle number density and $\langle\ \rangle$ denotes an ensemble average, the precise meaning of which will be discussed in §3.1. However, $\boldsymbol{M}$ can be calculated in the limit of small concentration, where the orientational probability necessary for evaluating ensemble averages has a comparatively simple form. In §3.1 we consider the leading-order approximation to $\boldsymbol{M}$, i.e. neglecting particle–particle interactions; and in §3.2 we consider the first correction due to interactions.

3.1. *Bulk magnetization to leading order in concentration*

At this order, each particle can be considered separately, and interacts only with the ambient field $\boldsymbol{H}$, which is assumed uniform on the particle lengthscale. In consequence, this result is valid independently of the particle shapes, since this affects only particle–particle interactions.

If $\mathscr{E}$ is the Hamiltonian of a system in equilibrium, the probability P of the system being in an infinitesimal region $\mathrm{d}\boldsymbol{Q}$ of phase space is given by the Maxwell–Boltzmann distribution

$$P(\boldsymbol{Q}) = \alpha \mathrm{e}^{-\mathscr{E}(\boldsymbol{Q})/kT}, \tag{3.1}$$

where α is a normalization constant, chosen to make the total probability equal to unity, and $\boldsymbol{Q}$ is the state of the system.

Statistical equilibrium of fluid suspensions is achieved when none of the macroscopic state variables of interest changes in time. A magnetic fluid with a constant ambient magnetic field and no bulk fluid motion satisfies this condition, even though it can be described from the non-equilibrium point of view of translational and rotational diffusions (which imply a local entropy gradient) competing with the orientating effect of the ambient field. The Maxwell–Boltzmann distribution is therefore applicable if and only if there is an energy function for the system.

We are now in a position to calculate the leading-order approximation to $\boldsymbol{M}$ explicitly. First we will consider the case of no ambient flow, and secondly we will show how we can also obtain an expression for the bulk magnetization for a magnetic fluid in a uniform straining motion.

We define angular ensemble averages as averages over all orientations, weighted by the orientational probability distribution $P(\boldsymbol{m})$, wihich is obtained by applying (3.1) in this case, and is given by

$$P(\boldsymbol{m}) = \frac{1}{4\pi} \frac{H}{\sinh H} \mathrm{e}^{\boldsymbol{m}\cdot\boldsymbol{H}};$$

this gives

$$\begin{aligned} \boldsymbol{M} &= n \int_{\text{all orientations}} \boldsymbol{m} P(\boldsymbol{m}) \, \mathrm{d}^2\boldsymbol{m} \\ &= nm\mathscr{L}(H)\, \hat{\boldsymbol{H}}, \end{aligned} \tag{3.2}$$

where $\mathscr{L}(H) \equiv \coth H - 1/H$ is the Langevin function (Langevin 1905), and $\hat{\boldsymbol{H}}$ is a unit vector in the direction of $\boldsymbol{H}$.

It is possible to apply the Maxwell–Boltzmann distribution to a system containing a single ellipsoidal particle in an ambient straining motion $\boldsymbol{E}$ and ambient magnetic field $\boldsymbol{H}$, since there exists an energy function for both the magnetic and hydrodynamic forces. In non-dimensional units, the energy is given by

$$\mathscr{E} = -\boldsymbol{H}\cdot\boldsymbol{m} - \frac{\mathbb{P}}{2}\left(\frac{r^2-1}{r^2+1}\right)\boldsymbol{m}\cdot\boldsymbol{E}\cdot\boldsymbol{m}.$$

This can again be substituted in the Maxwell–Boltzmann distribution, so enabling us to calculate the bulk magnetization $\boldsymbol{M}$, although in general the integral appearing in the equation corresponding to (3.2) cannot be evaluated simply. However for $O(1)$ magnetic field the effect of the straining motion is small, $O(\mathbb{P})$, in which case perturbation methods may be used.

One might ask whether it is possible to achieve a similar result for a general linear flow (i.e. including a rotational part $\boldsymbol{\Omega}$). Unfortunately this is not possible as the steady state for such a system is not one of zero flux in orientation space, and so no single-valued energy function exists. We shall, however, tackle the problem of a magnetic fluid in a general linear flow in the limit of small Péclet number in §4.

3.2. *Magnetization including effect of particle interactions at* $O(c^2)$

The inclusion of interparticle interactions is not at all as simple as one might imagine. It would at first sight seem reasonable to include only the effect of interactions of close particles (those within a few diameters of each other). The fraction of particles

in close pairs is $O(c^2)$, in close triplets it is $O(c^3)$, and so on. This implies we only have to consider pair interactions of particles. To calculate the magnetization to $O(c^2)$, we need to find the orientational probability distribution for a test particle and use this probability to calculate angular ensemble averages. It might be thought that this could be done by simply defining an approximate energy of the system, including the direct interactions of particles with the ambient magnetic field and pair interactions of particles, then substituting this in the Maxwell–Boltzmann equation and integrating out all but the orientation of the test particle. Unfortunately, this does not work; this is because the pair interaction energy for particles of separation r is $O(1/r^3)$, and this cannot be integrated over all non-overlapping positions (it diverges logarithmically as r tends to infinity). This implies that the total effect of distant particles is always felt, no matter how distant they are. In short, for these purposes, we cannot talk about an infinite expanse of magnetic fluid without in some sense considering its 'shape', a difficulty encountered frequently in suspension-mechanical problems. We use the technique of O'Brien (1979) to resolve the difficulty.

Consider a test particle in a region of magnetic fluid with an ambient field $\boldsymbol{H}$, uniform over the interparticle lengthscale. As seen by the test particle, distant regions of the magnetic fluid are almost indistinguishable from a continuum, whereas, closer to the test particle, fine detail (e.g. the orientations and positions of particular particles) have an important effect. Thus it is possible to take an arbitrary surface S (containing volume V, with outward unit normal $\boldsymbol{n}$) around the test particle such that the magnetic fluid outside S is sufficiently far away that it can be replaced by a continuum and only the magnetic fluid inside has to be dealt with discretely (see figure 1).

Outside V we can use the following form of Maxwell's equations:

$$\nabla\cdot\boldsymbol{B}=0, \quad \nabla\wedge\boldsymbol{H}=0, \quad \boldsymbol{B}=\boldsymbol{H}+\boldsymbol{M},$$

where all the equations above have their usual meaning, and μ_0 has been set equal to 1. Defining magnetic charge ρ by

$$\rho=\nabla\cdot\boldsymbol{H},$$

we have

$$0=\nabla\cdot\boldsymbol{B}=\nabla\cdot\boldsymbol{H}+\nabla\cdot\boldsymbol{M}=\rho+\nabla\cdot\boldsymbol{M}. \tag{3.3}$$

As regards the region interior to V, the effect of the magnetization outside V may be replaced by a magnetic charge concentrated on the surface S, denoted by ρ_S. Then from (3.3) we have that ρ_S is equal to $\boldsymbol{M}\cdot\boldsymbol{n}$ and defining the magnetic field in V due to ρ_S to be $\boldsymbol{H}_1(\boldsymbol{r})$, we have

$$\boldsymbol{H}_1(\boldsymbol{r})=\nabla\int_S\frac{-\boldsymbol{M}\cdot\boldsymbol{n}}{4\pi R}\,\mathrm{d}S.$$

So the magnetic field in V due to the ambient field $\boldsymbol{H}$ and the magnetic fluid outside V is $\boldsymbol{H}+\boldsymbol{H}_1(\boldsymbol{r})$. The above surface integral can be replaced by a volume integral if we analytically continue $\boldsymbol{M}$ from outside V to the inside; or equivalently consider $\boldsymbol{M}$ to represent the averaged magnetization over all space. This can be done by constructing a spherical surface S_0 around the test particle and letting V^* be the volume between S and S_0; then we find, by use of the divergence theorem, that

$$\boldsymbol{H}_1(\boldsymbol{r})=\int_{V^*}\frac{\boldsymbol{M}-3\boldsymbol{M}\cdot\hat{\boldsymbol{R}}\hat{\boldsymbol{R}}}{4\pi R^3}\,\mathrm{d}V+\nabla\int_{S_0}\frac{-\boldsymbol{M}\cdot\boldsymbol{n}}{4\pi R}\,\mathrm{d}S.$$

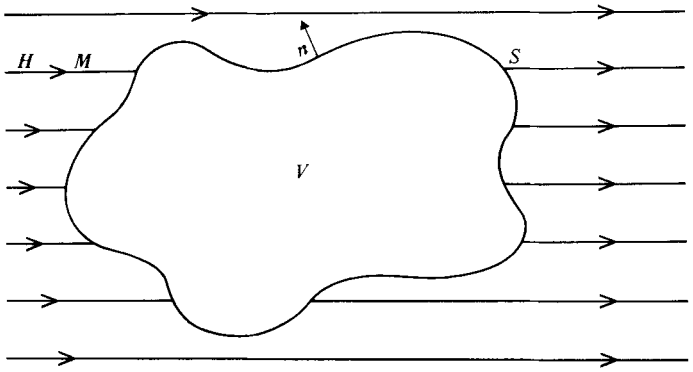

FIGURE 1. Diagram defining volume around the test particle.

Then, evaluating the integral over S_0, we have

$$\boldsymbol{H}_1(\boldsymbol{r}) = \int_{V^*} \frac{\boldsymbol{M} - 3\boldsymbol{M}\cdot\hat{\boldsymbol{R}}\hat{\boldsymbol{R}}}{4\pi R^3}\,\mathrm{d}V + \tfrac{1}{3}\boldsymbol{M}.$$

We must now deal with the particles in V discretely; this is possible by consideration of the Maxwell–Boltzmann equation for the system inside V. Consider $N+1$ particles in V with magnetic dipole moments $\boldsymbol{m}_0, \ldots, \boldsymbol{m}_N$ and positions $\boldsymbol{r}_0, \ldots, \boldsymbol{r}_N$; then the probability of this configuration is

$$P(\boldsymbol{m}_0, \ldots, \boldsymbol{m}_N; \boldsymbol{r}_0, \ldots, \boldsymbol{r}_N) \propto \exp\left\{\boldsymbol{m}_0\cdot(\boldsymbol{H}+\boldsymbol{H}_1(\boldsymbol{r}_0)) + \sum_1^N \boldsymbol{m}_i\cdot(\boldsymbol{H}+\boldsymbol{H}_1(\boldsymbol{r}_i)) - U\right\},$$

where U is the energy associated with particle interactions in V.

To find the $O(c^2)$ correction to the Langevin function, we only need consider pairwise interaction terms in U, giving

$$U = \frac{1}{2}\sum_{i\neq j} \mathscr{E}_{ij},$$

where

$$\mathscr{E}_{ij} = \frac{1}{4\pi\mu_0 r^3}(\boldsymbol{m}_i\cdot\boldsymbol{m}_j - 3(\boldsymbol{m}_i\cdot\hat{\boldsymbol{r}})(\boldsymbol{m}_j\cdot\hat{\boldsymbol{r}})).$$

It is convenient to define $\overline{\boldsymbol{H}} = \boldsymbol{H} + \frac{1}{3}\boldsymbol{M}$, which is simply the average magnetic field over all points outside of particles, and $\boldsymbol{H}_i' = \boldsymbol{H}_1(\boldsymbol{r}_i) - \frac{1}{3}\boldsymbol{M}$; then the Maxwell–Boltzmann distribution becomes

$$P(\boldsymbol{m}_0, \ldots, \boldsymbol{m}_N; \boldsymbol{r}_0, \ldots, \boldsymbol{r}_N) \propto \exp\{\boldsymbol{m}_0\cdot\overline{\boldsymbol{H}}\}\exp\left\{\sum_1^N \boldsymbol{m}_i\cdot\overline{\boldsymbol{H}}\right\}\exp\{-\tfrac{1}{2}\sum_{i,j}\tilde{\mathscr{E}}_{ij}\},$$

where $\tilde{\mathscr{E}}_{ij} = \mathscr{E}_{ij} - (1/2N)\,\boldsymbol{m}_i\cdot\boldsymbol{H}_i'$ and has been constructed so that $\langle\tilde{\mathscr{E}}_{ij}\rangle$, the average value of $\tilde{\mathscr{E}}_{ij}$ over V, vanishes to leading order in c.

Since the exponential function is absolutely convergent, we may expand the $\exp\{-\frac{1}{2}\Sigma_{i,j}\tilde{\mathscr{E}}_{ij}\}$ factor, and integrate term by term over all variables except $\boldsymbol{m}_0$. This gives the orientational probability function $P(\boldsymbol{m}_0)$ of a single particle accurate to $O(c)$ as being

$$P(\boldsymbol{m}_0) \propto P_0(\boldsymbol{m}_0)\left(1 + \frac{c}{V_0}\int P(\boldsymbol{m}_0)\,S(\mathscr{E}_{01})\,\mathrm{d}^2\hat{\boldsymbol{m}}_1\,\mathrm{d}^3\boldsymbol{r}_1 + O(c^2)\right), \tag{3.4}$$

where $P_0(\boldsymbol{m}_0) = \alpha_0\,\mathrm{e}^{\boldsymbol{m}_0\cdot\overline{\boldsymbol{H}}}$ (which is the simple Langevin probability function with $\boldsymbol{H}$ replaced by $\overline{\boldsymbol{H}}$) and $S(x) = \mathrm{e}^x - x - 1$. The term that produced the divergence in the

naive calculation vanishes (to leading order) by construction, and so $P(\boldsymbol{m}_0)$ is independent of the shape of V.

The cross-terms in the expansion of the exponential are $O(c^2)$ after integration, as they localize two or more of the $\boldsymbol{r}_i$ integrals; also for similar reasons we are able to replace $\tilde{\mathscr{E}}_{01}$ by $\mathscr{E}_{01}$, giving an $O(c^2)$ error in $P(\boldsymbol{m}_0)$.

The magnetization to $O(c^2)$ is thus given by

$$\boldsymbol{M} = \boldsymbol{M}_\infty \int \hat{\boldsymbol{m}}_0 P(\boldsymbol{m}_0)\, \mathrm{d}^2\hat{\boldsymbol{m}}_0,$$

where $\boldsymbol{M}_\infty = nm\hat{\boldsymbol{H}}$. The evaluation of this integral is difficult analytically, but we are able to evaluate it asymptotically. We assume that the suspension is stable; this implies that the interaction energy $\mathscr{E}_{01}$ is small, so not many particles are in close proximity to one another. We shall now replace $S(\mathscr{E}_{01})$ by $\frac{1}{2}\mathscr{E}_{01}^2$ in equation (3.4) for $P(\boldsymbol{m}_0)$; and after some algebra we obtain an approximate expression for the magnetization, *viz*

$$\frac{M}{M_\infty} = \mathscr{L}_1(H+\tfrac{1}{3}M)\{1+cW_0^2 f(H)+O(cW_0^3, c^2)\}. \tag{3.5}$$

Here $W_0 = \frac{1}{12}I^2V_0/\mu_0 kT$ is the stability parameter $\mathscr{E}_0/kT$ introduced in §2.1, and

$$f(H) = \frac{2}{5}\left\{\frac{\mathscr{L}_1(H)\,\mathscr{L}_2(H)}{H} - \frac{\mathscr{L}_2^2(H)}{H\mathscr{L}_1(H)}\right\},$$

where the functions $\mathscr{L}_n$ are defined recursively by

$$\mathscr{L}_n(x) = \mathscr{L}_{n-2}(x) - \frac{2n-1}{x}\mathscr{L}_{n-1}(x), \quad \forall n \in \mathbb{N},$$

with

$$\mathscr{L}_0(x) \equiv 1, \quad \mathscr{L}_1(x) = \mathscr{L}(x).$$

The function $f(x)$ has the asymptotic limits

$$f(x) \sim \tfrac{4}{1125}\, x^2 \quad \text{as} \quad x \to 0,$$

$$f(x) \sim \frac{2}{5x^2} \quad \text{as} \quad x \to \infty,$$

and has maximum value 0.008 at $x \approx 3$. Since for any real magnetic fluid $c \lesssim 0.5$ and $W_0 \lesssim 1$, the factor containing $f(H)$ in equation (3.5) for the magnetization will always be close to unity; this implies that the main contribution to the magnetization from particle interactions is due to the increase in magnetic field, as seen by a particle, caused by the continuum-like behaviour of distant particles.

In this calculation of the magnetization, we only had to make the assumption of small concentration to deal with the detailed structure of the magnetic fluid close to the test particle, but the effect of this is seen to be much smaller than the contribution from distant particles; so we would expect, for a stable suspension ($W_0 \lesssim 1$), the magnetization *for all values of* c to be given approximately by

$$M = M_\infty \mathscr{L}_1\left(\frac{H+\dfrac{1}{3\mu_0}M}{kT}\right). \tag{3.6}$$

This expression is interesting in that it gives rise to the possibility of a non-zero magnetization even if the ambient field H is zero; i.e. the magnetic fluid as a whole is ferromagnetic below some 'Curie temperature', T_c. It should be emphasized that

we are referring here to a property of the magnetic fluid and not a property intrinsic to the particles. Equations of the form of (3.6) were used by Weiss & Fox (1926) to explain ferromagnetism in metals, but did not give agreement with experiments. In fact the direct dipole interactions were of the order of 1000 times too small to explain the experimental results; this was because the main interactors were quantum-mechanical in nature. But, for a magnetic fluid governed by classical mechanics, the Weiss formula is seen to be applicable.

We define the Curie temperature T_c for an isolated volume of magnetic fluid as the temperature below which there exists a steady bulk magnetization, even when there is no externally applied magnetic field. This definition of the Curie temperature is not only a function of the magnetic fluid itself but also of the shape of the bounding volume through the ambient magnetic field. To demonstrate this we shall consider two geometries, namely rod-shaped and spherical volumes.

Rod-shaped volume

Consider a uniformly magnetized rod with magnetization in the direction of the axis of symmetry. In this case, zero externally applied magnetic field implies that $\boldsymbol{H} = 0$, and (3.6) has two solutions for the magnetization if $T < \frac{1}{9}c(I^2 V_0/\mu_0 k)$. However, this does not necessarily imply that this critical temperature is a Curie temperature, unless we are able to show that the solution with $M = 0$ is unstable. To investigate this we must consider the time evolution equation for M which may be derived simply from the time-evolution equation for P, which will be considered in detail in §4. Making the same assumptions as for the derivation of (3.6) and linearizing the equation for P about the isotropic probability distribution we find, in dimensional form,

$$\dot{\boldsymbol{M}} = 2D\left(\frac{c}{9}\left(\frac{I^2 V_0}{\mu_0 kT}\right) - 1\right)\boldsymbol{M},$$

from which we deduce that the $M = 0$ solution is always unstable when another solution exists, and so the Curie temperature for the system, $T_c = \frac{1}{9}c(I^2 V_0/\mu_0 k)$.

Spherical volume

The magnetic field $\boldsymbol{H}$ inside a uniformly magnetized sphere with no externally applied field is given by

$$\boldsymbol{H} = -\frac{1}{3\mu_0}\boldsymbol{M},$$

and substituting this into (3.6) we find that the only solution for the magnetization for all T is $M = 0$. This does not, however, exclude the possibility of non-zero magnetization in small domains, such as those found in iron, for example.

If we examine the stability condition derived in §2.1 for a system at its Curie temperature, we find that we require $c = O(1)$, in which case we would expect most magnetic fluids to be prone to aggregation. So the analysis given above is only valid either qualitatively or for short times.

4. Stress tensor for a dilute magnetic fluid

In this section the particle stress tensor for a magnetic fluid is calculated in the limit of small Péclet number to $O(c)$, so each particle can be treated as if alone in the ambient flow and magnetic field. Although we shall only consider the case of a monodisperse suspension of spheroidal particles, the results of this section can be easily generalized to a polydisperse suspension by a simple averaging process.

4.1. *Probability distribution for a non-equilibrium system*

As mentioned in §3.1, there are many situations where the Maxwell–Boltzmann distribution for particle orientations is inapplicable. In this subsection we derive a diffusion equation satisfied by the orientational probability density function P. It is permissible to write this function as $P(\boldsymbol{m}, t)$, as the orientation of an axisymmetric particle is specified by the direction of its principal axis, and so the orientation space is the surface of a sphere.

Since the particles of the magnetic fluid are much larger than the molecules of the ambient fluid, individual collisions of fluid molecules with a suspended particle give rise to a very small change in the orientation of the suspended particle. So the trajectories of individual particles in orientation space are well approximated by smooth curves and thus the probability density can be described in continuum terms.

$P(\boldsymbol{m}, t)$ is normalized so that

$$\int P(\boldsymbol{m}, t)\, \mathrm{d}^2\boldsymbol{m} = 1$$

for all t; the continuity of $P(\boldsymbol{m}, t)$ in orientation space gives

$$\frac{\partial P(\boldsymbol{m}, t)}{\partial t} + \boldsymbol{\nabla}\cdot\boldsymbol{F} = 0, \tag{4.1}$$

where $\boldsymbol{F}$ is the probability flux and $\boldsymbol{\nabla}$ is the two-dimensional gradient operator in orientation space. Using an extension of Einstein's (1905) argument, it can be shown that the effect of Brownian motion on the evolution of $P(\boldsymbol{m}, t)$ is equivalent to applying a (dimensional) couple $\boldsymbol{L}^{\mathrm{B}} = -kT\boldsymbol{\nabla}\log(P(\hat{\boldsymbol{m}}, t))$ to each particle (Brenner 1967). The tendency for the applied magnetic field to align particles with it is represented by a (dimensional) couple $\boldsymbol{L}^{\mathrm{M}} = m\boldsymbol{H}_{\perp}$, where $\perp$ represents taking the perpendicular part with respect to $\boldsymbol{m}$ (i.e. contracting once with $(\boldsymbol{I} - \hat{\boldsymbol{m}}\hat{\boldsymbol{m}})$), and the contribution to the (dimensional) angular velocity from hydrodynamic interactions between particles and the ambient flow is

$$\boldsymbol{\Omega}\cdot\hat{\boldsymbol{m}} + \frac{r^2-1}{r^2+1}(\boldsymbol{E}\cdot\hat{\boldsymbol{m}})_{\perp}$$

(Jeffrey 1922). Summing the contributions of these effects after translating couples into angular velocities via the orientational mobility $b(r)$, the full (dimensional) probability flux becomes

$$\boldsymbol{F} = P(\hat{\boldsymbol{m}}, t)\left\{\left(\boldsymbol{\Omega} + \frac{r^2-1}{r^2+1}\boldsymbol{E}\right)\cdot\hat{\boldsymbol{m}} + bm\boldsymbol{H} - kT\boldsymbol{\nabla}\log P(\hat{\boldsymbol{m}}, t)\right\}_{\perp}.$$

In terms of the above, the orientational diffusivity D is given by

$$D = bkT.$$

Substituting $\boldsymbol{F}$ into (4.1) and using the non-dimensionalizations of §2.2, we find

$$\frac{\partial P(\boldsymbol{m}, t)}{\partial t} + \boldsymbol{\nabla}\cdot\left\{\mathbb{P}\left(\boldsymbol{\Omega} + \frac{r^2-1}{r^2+1}\boldsymbol{E}\right)\cdot\boldsymbol{m}P(\boldsymbol{m}, t) + \boldsymbol{H}P(\boldsymbol{m}, t) - \boldsymbol{\nabla}P(\boldsymbol{m}, t)\right\} = 0, \tag{4.2}$$

where $\boldsymbol{\nabla}$ takes the perpendicular part of its operand.

4.2. *Particle stress tensor*

In the rest of this section we shall make the simplifying assumption of small Péclet number, neglecting all terms $O(\mathbb{P}^2)$. This is reasonable for any macroscopic system having timescales very much greater than the Brownian timescale $O(1/D)$ (i.e. of order 10^{-6} s).

It is convenient to split the stress tensor $\boldsymbol{\sigma}$ into symmetric and antisymmetric parts. The symmetric part is calculated via the particle stresslet, whereas the antisymmetric part can be calculated directly from consideration of the bulk magnetization and applied field.

Antisymmetric part of the stress $\boldsymbol{\sigma}^{\mathrm{a}}$

We consider the couples on a small fluid element V having surface S; the couple due to the fluid and particle stress must be exactly balanced by the couple due to the applied magnetic field. The couple due to contact stress is given by

$$\int_S \boldsymbol{r} \wedge \boldsymbol{\sigma}\cdot\boldsymbol{n}\,\mathrm{d}S, \tag{4.3}$$

where $\boldsymbol{n}$ is the outward normal of S. The contribution from the symmetric part of the stress is zero as it is contracted with the antisymmetric alternating tensor $\boldsymbol{\epsilon}$. Using the divergence theorem, (4.3) becomes $-\int_V \boldsymbol{\epsilon}:\boldsymbol{\sigma}^{\mathrm{a}}\,\mathrm{d}V$, which must equal the total magnetic couple on V, namely $\int_V n\langle\boldsymbol{m}\rangle \wedge \boldsymbol{H}\,\mathrm{d}V$, where n is the number density. Taking the limit as V tends to zero, we find

$$\boldsymbol{\sigma}^{\mathrm{a}} = \tfrac{1}{2}n\boldsymbol{\epsilon}\cdot(\langle\boldsymbol{m}\rangle \wedge \boldsymbol{H}), \tag{4.4}$$

where $n\langle\boldsymbol{m}\rangle$ can be replaced by the bulk magnetization $\boldsymbol{M}$. Such situations of suspensions under applied couples were first considered by Batchelor (1970).

Symmetric part of the particle stress

There are three contributions for the symmetric part of the particle stress, from straightforward hydrodynamic forces, from direct Brownian couples and from magnetic couples. The stresslet exerted by one particle on the fluid can be written, in dimensional form, as

$$\boldsymbol{S} = \mu V_0 \boldsymbol{C}:\boldsymbol{E} - \frac{3\mu V_0 D_0}{kT}(\boldsymbol{L}^{\mathrm{B}} + \boldsymbol{L}^{\mathrm{M}})\cdot\boldsymbol{B}, \tag{4.5}$$

where D_0 is the rotational diffusivity for a sphere of volume V_0, $\boldsymbol{C}$ is a particle tensor introduced by Batchelor (1970), and $\boldsymbol{B}$ is a symmetric traceless tensor first used by Bretherton (1962).

We expand $P(\boldsymbol{m}, t)$ as a power series in $\mathbb{P}$:

$$P(\boldsymbol{m}, t) = P_0(\boldsymbol{m}, t) + P_1(\boldsymbol{m}, t) + O(\mathbb{P}^2),$$

where $P_0(\boldsymbol{m}, t)$ is simply the probability distribution in the case of zero flow, i.e.

$$P_0(\boldsymbol{m}, t) = \frac{H}{4\pi \sinh H}\mathrm{e}^{-\boldsymbol{H}\cdot\boldsymbol{m}},$$

and $P_1(\boldsymbol{m}, t)$ is $O(\mathbb{P})$. Then (4.5) can be written, dimensionally, as

$$\boldsymbol{S} = \mu V_0 \boldsymbol{C}:\boldsymbol{E} + 3\mu V_0 D_0\left(\frac{\nabla P_1}{P_0} - \boldsymbol{H}_\perp\frac{P_1}{P_0}\right)\cdot\boldsymbol{B}.$$

In computing the ensemble average of this form for the stresslet, the ∇-operator in the Brownian term can be turned around by integrating by parts; this gives the symmetric part of the (dimensional) particle stress as

$$\langle n\boldsymbol{S}\rangle = \mu c\langle \boldsymbol{C}\rangle_0 : \boldsymbol{E} - 3\mu D_0 c(\langle \nabla\cdot\boldsymbol{B}\rangle_1 + \langle \boldsymbol{H}_\perp\cdot\boldsymbol{B}\rangle_1)$$

(Rallison 1978), where $\langle\phi\rangle_i \equiv \int \mathrm{d}^2\boldsymbol{m}\, P_i(\boldsymbol{m},t)\,\phi$ for any function ϕ of orientation space. In this expression

$$\langle \boldsymbol{C}\rangle_0 : \boldsymbol{E} = 4\{A_0\langle \hat{\boldsymbol{m}}\hat{\boldsymbol{m}}\hat{\boldsymbol{m}}\hat{\boldsymbol{m}}\rangle_0 : \boldsymbol{E} + B_0(\langle \hat{\boldsymbol{m}}\hat{\boldsymbol{m}}\rangle_0\cdot\boldsymbol{E} + \boldsymbol{E}\cdot\langle \hat{\boldsymbol{m}}\hat{\boldsymbol{m}}\rangle_0) + C_0\boldsymbol{E}\},$$

A_0, B_0 and C_0 being functions of axis ratio alone as defined by Batchelor (1970),

$$\nabla\cdot\boldsymbol{B} = -6\frac{r^2-1}{r^2+1}\hat{\boldsymbol{m}}\hat{\boldsymbol{m}} + \text{isotropic term},$$

and

$$\boldsymbol{H}_\perp\cdot\boldsymbol{B} = \frac{r^2-1}{r^2+1}(\boldsymbol{H}_\perp\hat{\boldsymbol{m}} + \hat{\boldsymbol{m}}\boldsymbol{H}_\perp).$$

Notice that, although the contribution to the stresslet from the Brownian motion would vanish if the perturbation probability were zero, the fact that it is small, $O(\mathbb{P})$, does not mean that its contribution will be small. The contribution of the Brownian motion is in fact as large as that of the hydrodynamic term because the Brownian term contains a factor D_0 which is large, $O(\mathbb{P}^{-1})$ times the size of the rate-of-strain tensor; this fact was unfortunately overlooked by Martsenyuk (1973).

4.3. *Particle stress tensor for the case of steady and homogeneous magnetic field*

In this subsection we calculate the full stress tensor for a magnetic fluid with an applied magnetic field constant in the Lagrangian sense (i.e. constant in the particle frame of reference) in the small-Péclet-number limit. It is not necessary, however, to assume that the flow is also constant in the Lagrangian sense, provided that its timescale of variation is no larger than the convective timescale. In this case, the perturbation to the probability density function is $O(\mathbb{P}^2)$, and so the appropriate form of (4.2) is

$$\nabla\cdot\left\{\mathbb{P}\left(\boldsymbol{\Omega} + \frac{r^2-1}{r^2+1}\boldsymbol{E}\right)\cdot\boldsymbol{m}P(\boldsymbol{m},t) + \boldsymbol{H}P(\boldsymbol{m},t) - \nabla P(\boldsymbol{m},t)\right\} = 0.$$

Again expanding $P(\boldsymbol{m},t)$ in terms of the Péclet number, we find that the equation for $P_1(\boldsymbol{m},t)$ is

$$\nabla\cdot(P_1\boldsymbol{H} - \nabla P_1) = -\mathbb{P}\nabla\cdot\left(P_0\left(\boldsymbol{\Omega} + \frac{r^2-1}{r^2+1}\boldsymbol{E}\right)\right),$$

where P_0 is exactly the probability density function in the case of no flow. Writing $\phi = P_1/P_0$, we have

$$\nabla\cdot(P_0\nabla\phi) = \mathbb{P}\nabla\cdot\left(P_0\left(\boldsymbol{\Omega} + \frac{r^2-1}{r^2+1}\boldsymbol{E}\right)\right). \tag{4.6}$$

Since ϕ depends linearly on $\boldsymbol{\Omega} + [(r^2-1)/(r^2+1)]\boldsymbol{E}$, it must be of the form

$$\phi = \mathbb{P}\boldsymbol{T} : \left(\boldsymbol{\Omega} + \frac{r^2-1}{r^2+1}\boldsymbol{E}\right),$$

where $\boldsymbol{T}$ is only defined up to an isotropic term, and, on substitution into equation (4.6), we find

$$\nabla^2\boldsymbol{T} + \boldsymbol{H}\cdot\nabla\boldsymbol{T} = \boldsymbol{m}\boldsymbol{H}_\perp - 3\boldsymbol{m}\boldsymbol{m}. \tag{4.7}$$

It is helpful to split $\boldsymbol{T}$ into symmetric and antisymmetric parts $\boldsymbol{T}^{\mathrm{s}}$ and $\boldsymbol{T}^{\mathrm{a}}$ respectively, and we find that the equation for the symmetric part can be solved exactly, giving

$$\boldsymbol{T}^{\mathrm{s}} = \tfrac{1}{2}(\boldsymbol{mm} - \langle \boldsymbol{mm} \rangle_0).$$

We have used the fact that $\langle \boldsymbol{T} \rangle_0 = 0$ because our particular choice of P_0 implies $\int P_1(\boldsymbol{m}, t)\, \mathrm{d}^2\boldsymbol{m} = 0$. The equation for the antisymmetric part is

$$\nabla^2 \boldsymbol{T}^{\mathrm{a}} + \boldsymbol{H}\cdot\nabla \boldsymbol{T}^{\mathrm{a}} = \tfrac{1}{2}(\boldsymbol{mH} - \boldsymbol{Hm}),$$

which implies that $\boldsymbol{T}^{\mathrm{a}}$ must have the form

$$\boldsymbol{T}^{\mathrm{a}} = A(\boldsymbol{m}\cdot\boldsymbol{H})\,(\boldsymbol{mH} - \boldsymbol{Hm}),$$

and after some algebra we arrive at the following equation for the scalar function $A(\boldsymbol{m}\cdot\boldsymbol{H})$:

$$A'' H_\perp^2 + A'(H_\perp^2 - 4H_\parallel) - A(2 + H_\parallel) = \tfrac{1}{2}.$$

Changing variables from $\boldsymbol{m}\cdot\boldsymbol{H}$ to θ, where $\cos\theta = \boldsymbol{m}\cdot\boldsymbol{H}/|\boldsymbol{H}|$, we find that $A(\theta; H)$ is given by

$$A'' - A'(H\sin\theta - 5\cot\theta) - A(2 + H\cos\theta) = \tfrac{1}{2}, \tag{4.8}$$

where a prime now denotes $\mathrm{d}/\mathrm{d}\theta$. The boundary conditions are regularity of A at $\theta = 0$ and π. The function A, which was assumed to be independent of θ in the theory of Martsenyuk (1973), can be calculated numerically for all values of $\boldsymbol{H}$, or asymptotically in the limit as H tends to zero, giving

$$A(\theta; H) \sim -\tfrac{1}{4} + \tfrac{1}{32}(\cos\theta)\, H - (\tfrac{5}{256} + \tfrac{1}{256}\cos^2\theta)\, H^2 + (\tfrac{3}{1664}\cos\theta + \tfrac{3}{6656}\cos^3\theta)\, H^3 + O(H^4).$$

The limit as H tends to infinity will be considered later.

We are now in a position to evaluate the various contributions to the particle stress discussed in §4.2; we find, in dimensional form, that the full particle stress tensor is given by

$$\begin{aligned}
\frac{\boldsymbol{\sigma}^{\mathrm{p}}}{\mu c} = {} & F_0(\boldsymbol{E}:\hat{\boldsymbol{H}}\hat{\boldsymbol{H}})\,\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} \\
& + F_1(\boldsymbol{E}\cdot\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} + \hat{\boldsymbol{H}}\boldsymbol{E}\cdot\hat{\boldsymbol{H}}) \\
& + F_2 \boldsymbol{E} \\
& + F_3(\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} + \hat{\boldsymbol{H}}\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}) \\
& + F_4(\boldsymbol{E}\cdot\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} - \hat{\boldsymbol{H}}\boldsymbol{E}\cdot\hat{\boldsymbol{H}}) \\
& + F_5(\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} - \hat{\boldsymbol{H}}\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}),
\end{aligned} \tag{4.9}$$

where the dimensionless functions $F_0, \ldots, F_5$, described in terms of non-dimensional variables, are

$$F_0 = \left\{4A_0 - 6\frac{D_0}{D}\left(\frac{r^2-1}{r^2+1}\right)^2\right\}\mathscr{L}_4(H),$$

$$F_1 = 4\left(\frac{2A_0\,\mathscr{L}_3(H)}{H} + B_0\,\mathscr{L}_2(H)\right) + 3\frac{D_0}{D}\left(\frac{r^2-1}{r^2+1}\right)^2\left(\mathscr{L}_2(H) - \frac{4\mathscr{L}_3(H)}{H}\right),$$

$$F_2 = 4\left(\frac{2A_0\,\mathscr{L}_2(H)}{H^2} + \frac{2B_0\,\mathscr{L}_1(H)}{H} + C_0\right) + 3\frac{D_0}{D}\left(\frac{r^2-1}{r^2+1}\right)^2\left(\frac{6\mathscr{L}_2(H)}{H^2} + \frac{2\mathscr{L}_3(H)}{H}\right),$$

$$F_3 = \frac{r^2-1}{r^2+1}\tau_\Omega^{\mathrm{B}}(H)\frac{D_0}{D},$$

$$F_4 = \frac{r^2-1}{r^2+1} 3\mathscr{L}_2(H) \frac{D_0}{D},$$

$$F_5 = \tau_{\Omega}^{a}(H) \frac{D_0}{D}.$$

The functions $\tau_{\Omega}^{B}(H)$ and $\tau_{\Omega}^{a}(H)$ depend on the probability perturbation due to the $\boldsymbol{\Omega}$ part of the ambient flow field, and can be expressed in terms of the function $A(\theta; H)$ of (4.8) as follows:

$$\tau_{\Omega}^{B}(H) = -\langle A(\theta; H)\,[3H^2(3\cos^2\theta - 1 - 2\cos^4\theta) + 18H(\cos\theta - \cos^3\theta)]\rangle_0,$$

$$\tau_{\Omega}^{a}(H) = 3H^2\langle A(\theta; H)\,(\cos^2\theta - 1)\rangle_0.$$

Using the asymptotic form of $A(\theta; H)$ as $H \to 0$, we find

$$\tau_{\Omega}^{B}(H) \sim \tfrac{9}{40}H^2 + O(H^4),$$

$$\tau_{\Omega}^{a}(H) \sim \tfrac{1}{2}H^2 - \tfrac{109}{1920}H^4 + O(H^6)$$

as $H \to 0$. Also $\tau_{\Omega}^{B}(H)$ and $\tau_{\Omega}^{a}(H)$ both tend to 3 as $H \to \infty$.

Many authors have used the results of Martsenyuk (1973), who derived an expression for the particle stress tensor similar to (4.9), but one consequence of his *ad hoc* assumptions is that his function corresponding to $\tau_{\Omega}^{a}(H)$, on which the stress tensor depends for all axis ratios, differs from that given here in the second term.

4.4. *The effect of time-dependent magnetic field*

If the magnetic field varies on an $O(1)$ (Lagrangian) timescale as seen by a particle, there is an additional perturbation at $O(\mathbb{P})$ to the probability, which is thus as important as the ambient flow. Since to first order in Péclet number the equation for $P_1(\boldsymbol{m}, t)$ is linear, the additional probability perturbation due to a time-varying magnetic field is additive. Writing the probability perturbation due to the time-varying magnetic field as $P_0\psi$, and using (4.2), we obtain the following evolution equation for ψ:

$$\nabla^2\psi + \boldsymbol{H}\cdot\nabla\psi = \dot{\boldsymbol{H}}\cdot\boldsymbol{m} + \frac{\mathrm{D}}{\mathrm{D}t}\left(\log\frac{H}{4\pi\sinh H}\right). \tag{4.10}$$

It is convenient to solve (4.10) by means of a decomposition into two cases: $\dot{H} = 0$, i.e. the magnetic field is changing its direction but not its strength, and $\dot{\boldsymbol{H}}$ parallel to $\boldsymbol{H}$, i.e. the magnetic field is changing its strength but not its direction. Since ψ is linear in $\dot{\boldsymbol{H}}$, these two special cases can be superposed to give the general solution.

Case (a), $\dot{H} = 0$

This implies that it is possible to move into a rotating frame in which $\dot{\boldsymbol{H}} = 0$. Since we can neglect time derivatives of $\nabla\boldsymbol{u}$, and the Stokes-flow equations neglect all inertial effects, this transformation does not invalidate any of the equations of §4.3. In effect, we have replaced the rotating magnetic field by an additional rotational part of the ambient flow, and the particle stress tensor in this case is thus given by (4.9) with every occurrence of $\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}$ replaced by $\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}} - \hat{\boldsymbol{H}}$.

Case (b), $\dot{\boldsymbol{H}}$ parallel to $\boldsymbol{H}$

In this case it is possible to solve (4.10) analytically, giving

$$\psi = \left(\frac{1}{H}\coth H \log(\operatorname{cosec}\theta - \cot\theta) - \frac{1}{H}\log\sin\theta - \frac{1}{H\sinh H}\int\frac{\mathrm{e}^{-H\sin\theta}}{\sin\theta}\,\mathrm{d}\theta + \text{const}\right)\dot{H}, \tag{4.11}$$

where θ is again defined by $\cos\theta = \hat{\boldsymbol{H}}\cdot\boldsymbol{m}$ and the constant is determined by $\langle\psi\rangle_0 = 0$ (i.e. the perturbation probability has zero mean).

We can now calculate the corresponding contributions to the particle stress tensor, as in §4.3, to give the following general result for the particle stress tensor in a homogeneous magnetic field:

$$\begin{aligned}\frac{\boldsymbol{\sigma}^{\mathrm{p}}}{c} = {} & (F_0(\boldsymbol{E}:\hat{\boldsymbol{H}}\hat{\boldsymbol{H}}) - R_0\dot{H})\,\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} \\ & + F_1(\boldsymbol{E}\cdot\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} + \hat{\boldsymbol{H}}\boldsymbol{E}\cdot\hat{\boldsymbol{H}}) \\ & + F_2\boldsymbol{E} \\ & + F_3((\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}} - \dot{\hat{\boldsymbol{H}}})\,\hat{\boldsymbol{H}} + \hat{\boldsymbol{H}}(\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}} - \dot{\hat{\boldsymbol{H}}})) \\ & + F_4(\boldsymbol{E}\cdot\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} - \hat{\boldsymbol{H}}\boldsymbol{E}\cdot\hat{\boldsymbol{H}}) \\ & + F_5((\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}} - \dot{\hat{\boldsymbol{H}}})\,\hat{\boldsymbol{H}} - \hat{\boldsymbol{H}}(\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}} - \dot{\hat{\boldsymbol{H}}})). \end{aligned} \tag{4.12}$$

Hence the inclusion of time dependence introduces only one new function, namely

$$R_0(H;r) = R_1(H)\frac{D_0}{D}\frac{r^2-1}{r^2+1},$$

where $$R_1(H) = -\langle\psi_1(27\cos^2\theta + 9H(\cos^3\theta - \cos\theta))\rangle_0$$

(ψ_1 representing ψ evaluated at $\dot{H} = 1$), which has the asymptotic form in the limit as H tends to zero

$$R_1(H) = \tfrac{2}{5}H + O(H^3).$$

On examining (4.12) we see that the tensorial dependence of the particle stress tensor on $\boldsymbol{E}$, $\boldsymbol{\Omega}$, $\hat{\boldsymbol{H}}$, $\dot{\hat{\boldsymbol{H}}}$ appears to be deficient only in a term proportional to $\boldsymbol{\Omega}$. However, from equation (4.4), we know that the antisymmetric part of the stress tensor is proportional to $\boldsymbol{\epsilon}\cdot(\langle\boldsymbol{m}\rangle \wedge \boldsymbol{H})$. Since $\langle\boldsymbol{m}\rangle$ can only depend tensorially on $\boldsymbol{\Omega}$ in the combination $\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}$, the only antisymmetric term in the particle stress tensor containing $\boldsymbol{\Omega}$ is proportional to $\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} - \hat{\boldsymbol{H}}\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}$. Thus the tensorial dependence of the particle stress tensor for spheroidal particles is complete.

4.5. *Effect of space-varying magnetic field*

Static magnetic fluids

If the magnetic field is a function of position, $\boldsymbol{x}$, there is an additional body force density $\mathscr{F}$ acting on the fluid

$$\mathscr{F} = \boldsymbol{M}(\boldsymbol{H})\cdot\nabla\boldsymbol{H}. \tag{4.13}$$

For a static magnetic fluid, we can calculate this force to $O(c^2)$ using the expression for the bulk magnetization calculated in §3.2. In this case, $\boldsymbol{M}(\boldsymbol{H})$ is parallel to $\boldsymbol{H}$, and if we also assume that $\nabla \wedge \boldsymbol{H} = 0$ (i.e. no free electric currents), we find

$$\mathscr{F} = M(H)\nabla H.$$

This can be integrated to give

$$\mathscr{F} = -\nabla p_{\mathrm{m}},$$

where p_{m} is a 'magnetic pressure' given by

$$p_{\mathrm{m}} = -\int_0^H M(h)\,\mathrm{d}h.$$

Using the expression for the bulk magnetization given in (3.5), and neglecting the $O(c^2W_0^2)$ term (as its maximum contribution is about 0.2 % for c as large as 0.25), we

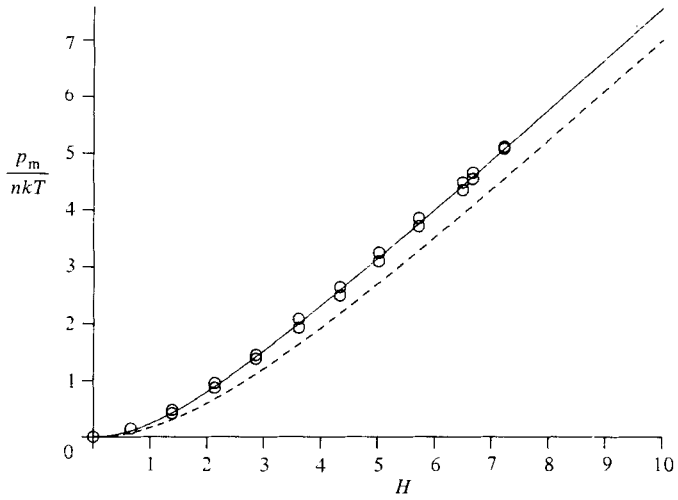

FIGURE 2. Comparison of theory with the experimental data (shown as ○) of Kamiyama *et al.* (1979), for a weight concentration of 25 %. The solid line is obtained from (4.14), and the dotted line is the $O(c)$ result.

find a particularly simple correction to the standard $O(c)$ result, given dimensionally as

$$p_m = nkT\left\{\log\left(\frac{\sinh H'}{H'}\right)+2cW_0\mathscr{L}^2(H')+O(c^2, cW_0^2)\right\}. \tag{4.14}$$

A possibly more accurate result could be obtained by integrating (3.6) directly, although this integral cannot be performed analytically.

Comparing (4.14) with the experimental data of Kamiyama *et al.* (1979) in figure 2, where it was necessary to fix the scale of H as the exact particle sizes were not known, we find remarkable agreement.

When solving problems containing free surfaces, it is not possible to include magnetic pressure in the hydrodynamic pressure, as they do not satisfy the same boundary conditions at the free surface. One such problem concerns the instability of a flat free surface with a uniform normal magnetic field (Cowley & Rosensweig 1967).

It is possible, at $O(c^2)$, for there to exist additional inhomogeneities in the magnetic field $\boldsymbol{H}$ due to variations in temperature or number density. In this case we can write the magnetic field as

$$\boldsymbol{H} = \boldsymbol{H}_0 + c\boldsymbol{H}_1,$$

where $\boldsymbol{H}_0$ is a homogeneous part of the magnetic field and $c\boldsymbol{H}_1$ is an $O(c)$ inhomogeneous perturbation. Similarly, the magnetization can be expanded as

$$\boldsymbol{M} = c\boldsymbol{M}_1 + c^2\boldsymbol{M}_2,$$

in which case the force density in the magnetic field is given by

$$\mathscr{F} = c\boldsymbol{M}_1\cdot\nabla\boldsymbol{H}_0 + c^2\boldsymbol{M}_1\cdot\nabla\boldsymbol{H}_1 + c^2\boldsymbol{M}_2\cdot\nabla\boldsymbol{H}_0 + O(c^3). \tag{4.15}$$

The second term in this expression for $\mathscr{F}$ models the effect of the interaction of the basic magnetization with the perturbed magnetic field, which is an effect not present at all at leading order in c. In the problem of the instability of a free surface in the presence of a non-uniform magnetic field, both the $O(c^2)$ terms in (4.15) are non-zero because the free surface can be considered as a surface on which the number density makes a discontinuous jump from a finite value to zero.

H	τ_Ω^a	τ_Ω^B	R_1	S
0.0	0.0000	0.0000	0.0000	1.0000
0.1	0.0050	0.0022	0.0399	0.9969
0.2	0.0199	0.0090	0.0794	0.9877
0.3	0.0445	0.0201	0.1180	0.9725
0.4	0.0783	0.0355	0.1552	0.9518
0.5	0.1209	0.0549	0.1908	0.9261
0.6	0.1716	0.0783	0.2244	0.8959
0.7	0.2296	0.1053	0.2557	0.8620
0.8	0.2943	0.1356	0.2844	0.8249
0.9	0.3645	0.1690	0.3105	0.7854
1.0	0.4396	0.2052	0.3339	0.7442
1.2	0.6005	0.2845	0.3721	0.6595
1.4	0.7700	0.3711	0.3994	0.5755
1.6	0.9420	0.4624	0.4166	0.4956
1.8	1.1113	0.5563	0.4250	0.4224
2.0	1.2742	0.6510	0.4261	0.3571
2.5	1.6371	0.8820	0.4059	0.2299
3.0	1.9266	1.0940	0.3682	0.1470
3.5	2.1471	1.2814	0.3256	0.0953
4.0	2.3118	1.4438	0.2846	0.0634
4.5	2.4344	1.5834	0.2480	0.0436
5.0	2.5264	1.7032	0.2164	0.0310
6.0	2.6507	1.8956	0.1667	0.0171
7.0	2.7278	2.0411	0.1312	0.0104
8.0	2.7789	2.1540	0.1055	0.0068
9.0	2.8148	2.2439	0.0864	0.0047
10.0	2.8412	2.3167	0.0720	0.0034

TABLE 1. Table of special functions of H

Dynamic magnetic fluids

For a flowing magnetic fluid in a non-uniform magnetic field, there is an additional contribution to the body force acting on the fluid, due to the flow, through the magnetization. Although this is of $O(\mathbb{P})$ smaller than the main contribution from the 'magnetic pressure', it can be important as it cannot be absorbed in the fluid pressure.

In the parameter regime under consideration, calculation of the bulk body force density, correct to $O(\mathbb{P})$, amounts to calculating the various contributions to the magnetization using the probability distributions discussed in §§4.1–4.4, and substituting them in (4.13). The result, in dimensional form, thus obtained is

$$\frac{\mathscr{F}}{\mu c} = \left\{3\frac{r^2-1}{r^2+1}(\mathscr{L}_3 - \mathscr{L}_2\mathscr{L}_1)(\boldsymbol{E}:\hat{\boldsymbol{H}}\hat{\boldsymbol{H}}) - S(H')\dot{H}'\right\}\frac{D_0}{D}\nabla H'$$
$$+\frac{D_0}{D}\frac{1}{H'}\tau_\Omega^a(\boldsymbol{\Omega}\cdot\hat{\boldsymbol{H}}-\dot{\hat{\boldsymbol{H}}})\cdot\nabla\boldsymbol{H}' + 6\frac{r^2-1}{r^2+1}\frac{\mathscr{L}_2}{H'}\frac{D_0}{D}(\boldsymbol{E}:\hat{\boldsymbol{H}}\nabla)\boldsymbol{H}' - \nabla p_m, \quad (4.16)$$

where $\boldsymbol{H}'$ represents the non-dimensional magnetic field, the functions $\mathscr{L}_1, \ldots, \mathscr{L}_3$ and τ_Ω^a are evaluated at $|\boldsymbol{H}'|\,(=H')$, and $S(H')$ is given by

$$S(H') = -6\langle(\cos\theta)\psi_1(\theta;H')\rangle_0,$$

where ψ_1 is as defined in §4.4.

We conclude this section with table 1, which gives all the functions of H that have been represented in terms of ensemble averages. This table enables the stress tensor to be easily calculated for any value of H in the range from 0 to 10.

5. Shear and pipe flows

Shear and pipe flows are of great practical significance, especially as magnetic fluids are often used as lubricants or to confine lubricants. Many experiments have been performed in order to determine the 'effective viscosity' for a magnetic fluid, although this notion is of limited value, because, even for simple shear flows, the 'effective viscosity' is dependent on the direction of $\boldsymbol{H}$. Several authors (McTague 1969; Mozgovoy, Blums & Cebers 1973, for example, whose results have been discussed by Schliomis & Raikher 1980), have measured the effective viscosity of a magnetic fluid by Poiseuille's method (i.e. in a pipe) for magnetic-field directions both perpendicular and parallel to the axis of the pipe; but we shall see later in this section that these directions are very special and give comparatively little information about the structure of the stress tensor.

5.1. *Simple shear flows with constant magnetic field*

We consider the shear flow with constant shear rate γ,

$$u_x = \gamma y, \quad u_y = u_z = 0,$$

so that

$$\boldsymbol{E} = \tfrac{1}{2}\gamma \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

$$\boldsymbol{\Omega} = \tfrac{1}{2}\gamma \begin{pmatrix} 0 & 1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

Applying (4.9), we find that the three components of the particle stress tensor that represent components of force on the bounding surface $y = 0$ are given by

$$\left.\begin{aligned} \frac{\sigma^{\mathrm{p}}_{12}}{\gamma c} &= F_0 h_1^2 h_2^2 + \tfrac{1}{2}(F_1 + F_5)(h_1^2 + h_2^2) + \tfrac{1}{2}(F_3 + F_4)(h_2^2 - h_1^2) + \tfrac{1}{2}F_2, \\ \frac{\sigma^{\mathrm{p}}_{22}}{\gamma c} &= F_0 h_1 h_2^3 + F_1 h_1 h_2 - F_3 h_1 h_2, \\ \frac{\sigma^{\mathrm{p}}_{32}}{\gamma c} &= F_0 h_1 h_2^2 h_3 + \tfrac{1}{2}(F_1 - F_3 - F_4 + F_5) h_1 h_3, \end{aligned}\right\} \tag{5.1}$$

where we have written $\hat{\boldsymbol{H}} = (h_1, h_2, h_3)$.

It is now evident that, if shear-flow experiments are performed with magnetic field directions coincident with one of the Cartesian axes, any term including products of different direction cosines h_i will vanish, in which case such an experiment can extract no information about the coefficients of these vanishing terms. In order to determine all the functions $F_0, \ldots, F_5$ experimentally, the three components of traction on a solid boundary for three independent magnetic field directions, none of which is coincident with any of the coordinate axes, must be measured for each value of H. An experiment of this kind is essential for a complete assessment of current theories.

The additional shear viscosity due to the magnetic interaction between suspended particles and the ambient magnetic field is denoted by most authors by the term 'magnetoviscosity' $\Delta\mu$. In our notation this is given by

$$\Delta\mu(\boldsymbol{H}) = \frac{1}{\gamma}(\sigma_{12}(\boldsymbol{H}) - \sigma_{12}(0)).$$

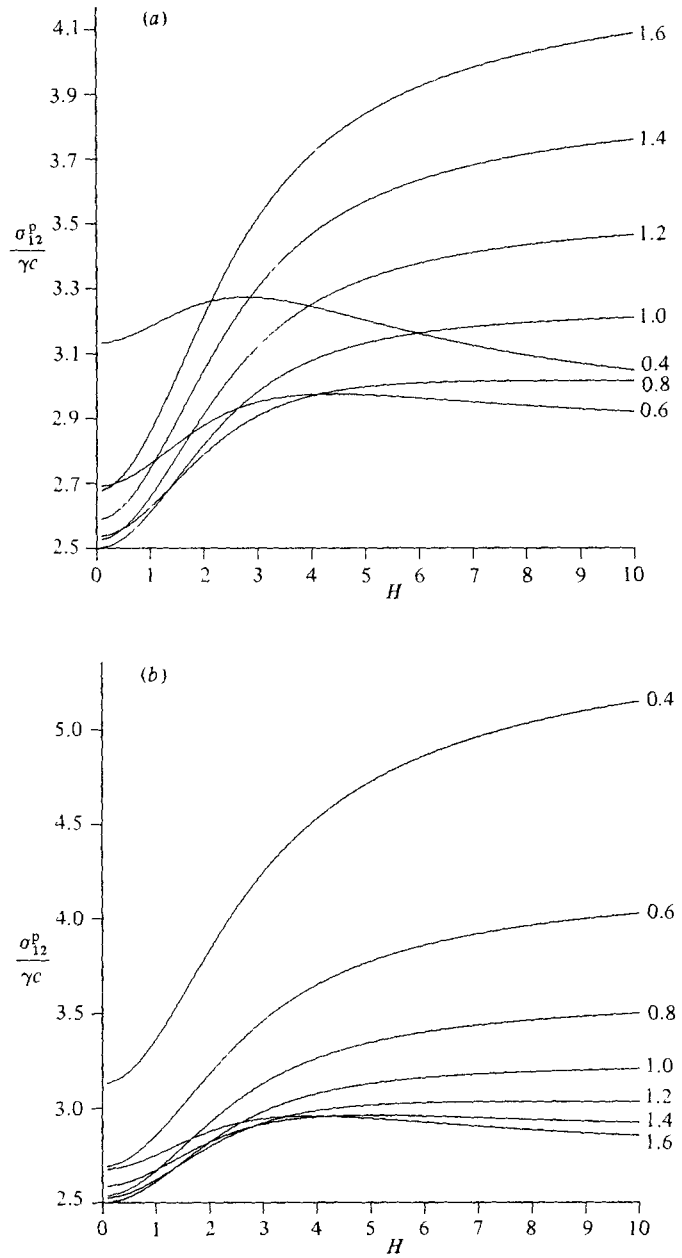

FIGURE 3. Graphs of contribution to 'effective viscosity' from the particle stress versus H in the cases (*a*) $\boldsymbol{H}$ parallel to $(0,1,1)$, and (*b*) $\boldsymbol{H}$ parallel to $(1,1,0)$; the number on the right-hand side of each curve denotes the corresponding axis ratio of the particles.

As mentioned previously, and is clear from (5.1), a magnetic fluid is non-Newtonian, and so is not completely described by $\mu+\Delta\mu$, even at $O(c)$; in particular, it exhibits a non-zero first normal stress difference $\sigma_{22}-\sigma_{11}$. From (4.9) we find

$$\frac{\sigma_{22}-\sigma_{11}}{\gamma c} = F_0 h_1 h_2 (h_2^2 - h_1^2) - 2F_3 h_1 h_2,$$

This phenomenon again would not be observed in shear or pipe flows with a magnetic field parallel to a coordinate axis.

We now present graphs (figure 3) of effective viscosity due to the particle stress for two magnetic-field directions. As there is no experimental data with which to compare these results, we leave discussion to the case of pipe flows, where experimental data is available.

5.2. *Pipe flows*

Since the flow in a pipe is unidirectional to leading order in c, it may be considered locally as a shear flow. Defining coordinates as shown in figure 4, the three components of the particle stress considered in §5.1 become

$$\left.\begin{aligned}
\frac{\sigma^{\mathrm{p}}_{12}}{\gamma c} &= F_0 h_\parallel^2 h_\perp^2 \cos^2\phi + \tfrac{1}{2}(F_1+F_5)(h_\parallel^2 + h_\perp^2\cos^2\phi) + \tfrac{1}{2}(F_3+F_4)(h_\perp^2\cos^2\phi - h_\parallel^2) + \tfrac{1}{2}F_2 \\
\frac{\sigma^{\mathrm{p}}_{22}-\sigma^{\mathrm{p}}_{11}}{\gamma c} &= F_0 h_\parallel h_\perp \cos\phi (h_\perp^2\cos^2\phi - h_\parallel^2) - 2F_3 h_\parallel h_\perp \cos\phi, \\
\frac{\sigma^{\mathrm{p}}_{32}}{\gamma c} &= F_0 h_\parallel h_\perp^3 \cos^2\phi \sin\phi + \tfrac{1}{2}(F_1 - F_3 - F_4 + F_5) h_\parallel h_\perp \sin\phi,
\end{aligned}\right\} \tag{5.2}$$

where $h_\parallel$ and $h_\perp$ are the components of $\hat{\boldsymbol{H}}$ parallel and perpendicular to the axis of the pipe, and ϕ is the azimuthal angle with origin in the direction of the perpendicular part of $\hat{\boldsymbol{H}}$. From (5.2) it can be seen that, in general, the particle stress drives a small secondary flow $O(c)$, and so it is not clear at first sight that the magnetoviscosity depends only on σ^{p}_{12}. However, for a circular pipe the secondary motion does not contribute at $O(c)$, although it would have to be included for an elliptical pipe, for example. For most non-Newtonian fluids, unidirectional flows are possible only in circular pipes (Bird, Armstrong & Hassager 1977), but even that is not the case here, except when the magnetic field is parallel to the axis of the cylinder.

Thus the magnetoviscosity, as determined by the Poiseuille method, is given by

$$\Delta\mu = \frac{\overline{\sigma^{\mathrm{p}}_{12}}(\boldsymbol{H}) - \overline{\sigma^{\mathrm{p}}_{12}}(0)}{\gamma},$$

where an overbar denotes an average with respect to ϕ. From (5.2) we find that $\overline{\sigma^{\mathrm{p}}_{12}}$ is given by

$$\frac{\overline{\sigma^{\mathrm{p}}_{12}}}{\gamma c} = \tfrac{1}{2}F_0 h_\parallel^2 h_\perp^2 + \tfrac{1}{2}(F_1+F_5)(h_\parallel^2 + \tfrac{1}{2}h_\perp^2) + \tfrac{1}{2}(F_3+F_4)(\tfrac{1}{2}h_\perp^2 - h_\parallel^2) + \tfrac{1}{2}F_2.$$

We compare this result with the experimental data of McTague (1969) in figure 5 for magnetic fields both perpendicular and parallel to the axis of the pipe.

It was necessary to determine the scale of H from the data, as the precise particle sizes were not known. As the limiting value of $\overline{\sigma^{\mathrm{p}}_{12}}$ for large magnetic fields is a function of axis ratio, we have restricted comparison of the data to the case $r = 1$ (i.e. spherical particles).

The greater part of the difference between theory and experiment is almost certainly due to polydispersivity in particle size rather than shape. For example, the experimentally measured magnetoviscosity is greater than the theory for small H and smaller than that for large H, which can be explained by observing that the larger particles are orientated more easily by the magnetic field, and so initially give a

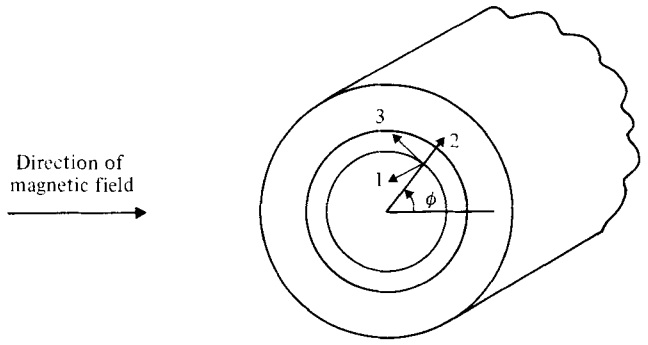

FIGURE 4. Diagram defining local Cartesian axes for pipe flow.

greater effect than the smaller particles, which are only significantly affected by much stronger magnetic fields. However, if all the particles were spherical, the magnetoviscosity at $O(c)$ for $\boldsymbol{H}$ parallel to the axis of the pipe would be simply twice that for $\boldsymbol{H}$ perpendicular to the axis, which is clearly not the case for the data given here. The dominant particle shape would seem to be one with axis ratio slightly less than unity (i.e. slightly disk-shaped), although the discrepancy between theory and experiment might equally well be attributed to particle–particle interactions (i.e. an $O(c^2)$ effect).

It is clear that the magnetoviscosity determined for a pipe with magnetic field parallel to its axis is identical to that for a shear flow with $\boldsymbol{H} = (1, 0, 0)$; however, it is interesting to note that the case of a pipe with perpendicular magnetic field also corresponds to a shear flow. At $O(c)$, $\overline{\sigma^{\mathrm{p}}_{12}}$ for perpendicular field is exactly equal to σ^{p}_{12} for a shear flow with $\boldsymbol{H}$ parallel to $(0, 1, 1)$, a result that is true for a polydisperse system at the same order. This observation could be used as a useful experimental tool for determining the importance of the $O(c^2)$ contribution to σ^{p}_{12}, by finding the difference in viscosity as calculated from the shear and pipe flows, since in this particular case the result would be $O(c^2)$, as $O(1)$ and $O(c)$ terms cancel identically.

6. Flow induced by rotating magnetic fields

The purpose of this section is to demonstrate some surprising and initially counterintuitive phenomena that magnetic fluids exhibit. We discuss three simple experiments, which, although of no practical importance in themselves, illustrate the role of the *antisymmetric* part of the stress tensor in the dynamics of magnetic fluids. These are intended primarily as thought experiments, although qualitative verification should not prove too difficult.

Each thought experiment consists of an infinitely long circular cylindrical tank, with a rigid boundary, filled with fluid (not necessarily just magnetic fluid), with a uniform magnetic field, of constant magnitude rotating with angular velocity $\boldsymbol{\Omega}_{\mathrm{m}}$ in the plane perpendicular to the axis of the cylinder, taken as the z-axis, which is assumed to be vertical. We shall assume that the radius R of the cylinder is small enough for any macroscopic flow induced by the magnetic field to be of sufficiently low Reynolds number for all inertia forces to be neglected, and so the Stokes-flow approximation is valid everywhere.

Since the flow field is induced solely by the rotating magnetic field, $\boldsymbol{E}$ and $\boldsymbol{\Omega}$ are $O(c)$ (i.e. proportional to the number density of magnetic particles), and since the particle stress tensor derived in §4.4 is correct to $O(c)$, any of its terms containing

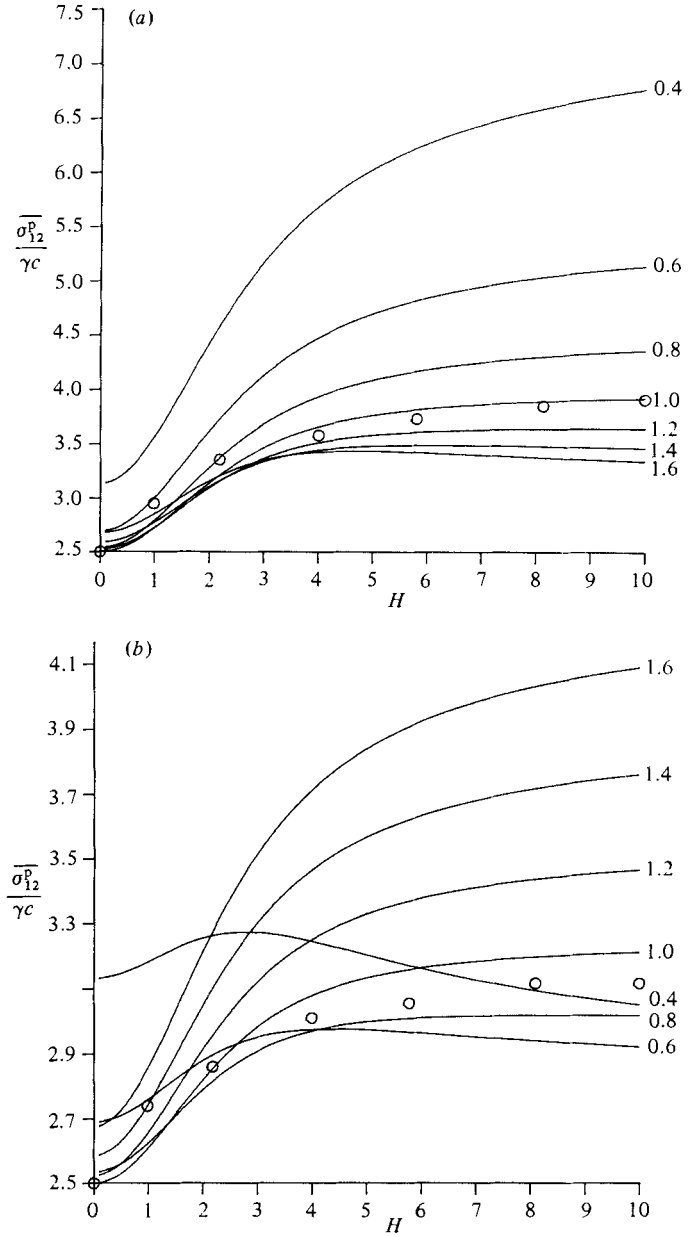

FIGURE 5. Graphs of contribution to 'effective viscosity' from the particle stress versus H in the cases (*a*) $(h_{\parallel}, h_{\perp}) = (1, 0)$; (*b*) $(h_{\parallel}, h_{\perp}) = (0, 1)$; the number on the right-hand side of each curve denotes the corresponding axis ratio of the particles. In each case the circles represent the experimental results of McTague (1969), scaled as if the particles were monodisperse with axis ratio of unity.

$\boldsymbol{E}$ or $\boldsymbol{\Omega}$ must be dropped for consistency, as they are $O(c^2)$. In this case, we find the appropriate form of the particle stress tensor is given by

$$\boldsymbol{\sigma}^{\mathrm{p}} = -c\{F_3(\dot{\hat{\boldsymbol{H}}}\hat{\boldsymbol{H}} + \hat{\boldsymbol{H}}\dot{\hat{\boldsymbol{H}}}) + F_5(\dot{\hat{\boldsymbol{H}}}\hat{\boldsymbol{H}} - \hat{\boldsymbol{H}}\dot{\hat{\boldsymbol{H}}})\},$$

so the full stress tensor, correct to $O(c)$, is

$$\boldsymbol{\sigma} = -p\boldsymbol{I} + 2\mu\boldsymbol{E} + \tfrac{1}{2}\boldsymbol{\epsilon}\cdot\boldsymbol{L} - \mu c F_3\{\dot{\hat{\boldsymbol{H}}}\hat{\boldsymbol{H}} + \hat{\boldsymbol{H}}\dot{\hat{\boldsymbol{H}}}\}, \tag{6.1}$$

where p is the pressure and

$$\boldsymbol{L} = 2\mu c F_5 \Omega_m \hat{\boldsymbol{z}},$$

which is the couple density due to the applied magnetic field.

Thought experiment 1: homogeneous magnetic fluid

This is an interesting thought experiment in that it often seems to be implicitly assumed that the induced rotation of magnetic particles due to the rotating magnetic field must manifest itself in some bulk motion of the fluid as a whole (Berkovsky 1978). There is also some confusion as to what boundary conditions should be applied at the rigid wall of the tank. The equation of fluid motion in this situation is simply $\nabla \cdot \boldsymbol{\sigma} = 0$, and since we have assumed that the magnetic fluid is homogeneous, so too will be the stress due to the rotating magnetic field (see (6.1)), which implies that

$$-\nabla p + \mu \nabla^2 \boldsymbol{u} = 0, \tag{6.2}$$

since the gradient of a homogeneous stress is identically zero. The boundary condition at the rigid wall will be simply the standard no-slip condition $\boldsymbol{u} = 0$, as appropriate for all hydrodynamic problems where the radius of curvature of the boundary is large compared with the range over which van der Waals forces act. Thus the only solution to (6.2), which is simply the standard Stokes-flow equation, is $\boldsymbol{u} \equiv 0$, which may be proved from the uniqueness theorem for viscous flows. However, as in all suspension-mechanical problems, there exists a depleted layer near the wall (i.e. a layer where the volume fraction of particles is less than that in the bulk medium just away from the wall) of thickness $O(a)$, as it is impossible for a particle to overlap the wall. So in this region there will be gradients in the ensemble-average stress, which will in general induce a small flow, although this flow will be of microscopic proportions, giving rise to ensemble-averaged velocities $O(\Omega_m a)$, and so is negligible in any macroscopic description.

The result obtained above may be surprising at first sight, since the rotating magnetic field will certainly induce a rotation of the individual magnetic particles, and so a stress on the fluid. It should be pointed out, however, that fluid motions are not induced by stresses, but rather by gradients of stresses, and in the experiment under consideration the region of space occupied by the magnetic fluid is one of constant stress. The natural question then arises as to what happens when the magnetic fluid is inhomogeneous.

Thought experiment 2: pure solvent core

For the following thought experiment we take a tank with non-magnetic fluid in the region $0 < r < R_1(\theta)$ and magnetic fluid in the region $R_1(\theta) < r < R$, where (r, θ) are polar coordinates in the plane perpendicular to the axis of the cylinder. In the case of a real experiment with gravity present, it would be necessary to ensure that the two fluids are of the same density to avoid buoyancy-driven convection. In the subsequent analysis, we shall assume that the surface tension between the two fluids is zero. This could be arranged if both fluids were suspensions, one a suspension of magnetic particles, and the other of non-magnetic particles, with the same solvent used in both cases, although in the rest of this section we shall take the non-magnetic fluid to be pure solvent for simplicity. We shall also assume that the experiment is of sufficiently short duration that translational Brownian motion does not have time to blur the interface significantly, and so we may consider the two fluids to be effectively immiscible.

Suppose that $R_1 = R_0 + f(\theta, t)$, where R_0 is a constant and $f = 0$ at $t = 0$. Then at

least for sufficiently small times, i.e. for the first few rotations of the magnetic field, $R_1 = R_0(1+O(c))$, in which case we may consider the effect of the two forcing terms in (6.1) separately; more precisely, we may solve

$$\nabla\cdot(-p\boldsymbol{I}+2\mu\boldsymbol{E}+\tfrac{1}{2}\boldsymbol{\epsilon}\cdot\boldsymbol{L}) = 0, \tag{6.3}$$

$$\nabla\cdot(-p\boldsymbol{I}+2\mu\boldsymbol{E}-\mu cF_3\{\dot{\hat{\boldsymbol{H}}}\hat{\boldsymbol{H}}+\hat{\boldsymbol{H}}\dot{\hat{\boldsymbol{H}}}\}) = 0 \tag{6.4}$$

separately, and then superpose the two solutions.

Solving (6.3), we notice that the expression for $\boldsymbol{\sigma}$ is radially symmetric; so in this case, if $R_1 = R_0$ initially, it will remain so. We may now solve for the flow by balancing stresses on coaxial cylindrical shells, from which we find that the region $r < R_0$ undergoes solid-body rotation, and in the region $R_0 \leqslant r \leqslant R$

$$u = -\frac{R_0^2 L}{4\mu}\left(\frac{1}{r}-\frac{r}{R^2}\right),$$

where u is defined by $\boldsymbol{u} = (0, u, 0)$ in cylindrical polar coordinates. So the angular velocity of the rigidly rotating core is

$$-\Omega_{\mathrm{m}}\frac{c\tau_\Omega^{\mathrm{a}} D_0}{2D}\left(1-\frac{R_0^2}{R^2}\right)+O(c^2),$$

giving the surprising result that the fluid rotates in the opposite direction to the magnetic field. This can be easily understood, however, by recalling experiment 1, and imagining a passive boundary at $r = R_0$. The rotating magnetic field applies a couple to the central region tending to rotate it with the field and this couple is exactly balanced by a hydrodynamic stress due to the magnetic fluid in the outer region. In the case of experiment 2, the outer region is essentially the same, whereas the inner region is pure solvent and hence has no couple, and so the central core region experiences a net couple tending to rotate in the opposite direction to the magnetic field.

We will now show that the so-far neglected term of the stress does not give any contribution to $O(c)$ to the time-averaged bulk flow, when averages are taken over several cycles of the magnetic field, and is in fact identically zero for spherical particles. Again the velocity field satisfies $\nabla\cdot\boldsymbol{\sigma} = 0$, which in each region reduces to the standard Stokes-flow equation, so the term $-\mu cF_3(\dot{\hat{\boldsymbol{H}}}\hat{\boldsymbol{H}}+\hat{\boldsymbol{H}}\dot{\hat{\boldsymbol{H}}})$ is only important in the stress jump boundary condition, which at $O(c)$ can be applied at $r = R_0$.

We shall now solve (6.4), which is best done by working in terms of a stream function ψ, noting that it must have the same angular dependence as the magnetic-forcing term. In this case

$$\psi = c\Omega_{\mathrm{m}} F_3 R_0^2 \cos 2(\theta-\Omega_{\mathrm{m}} t)\left(a_i\left(\frac{r}{R_0}\right)^4+b_i\left(\frac{r}{R_0}\right)^2+c_i+d_i\left(\frac{R_0}{r}\right)^2\right),$$

where $i = 1$ if $r < R_1$, $i = 2$ if $r > R_1$, and we have taken the magnetic field to be in the direction $\theta = \Omega_{\mathrm{m}} t$. The coefficients $a_1, \ldots, d_1, a_2, \ldots, d_2$ are determined by applying the boundary conditions at $r = R_0, R$, and insisting that the solution is regular at $r = 0$. We find

$$\begin{pmatrix} a_1 \\ b_1 \\ c_1 \\ d_1 \\ a_2 \\ b_2 \\ c_2 \\ d_2 \end{pmatrix} = \begin{pmatrix} (\lambda^3-1)/12\lambda^3 \\ -(\lambda^2-1)/8\lambda^2 \\ 0 \\ 0 \\ 1/12\lambda^3 \\ 1/8\lambda^2 \\ 0 \\ -\frac{1}{24} \end{pmatrix},$$

where $\lambda = (R/R_0)^2$.

The most interesting aspect of this flow field is that the boundary between magnetic fluid and pure solvent does not in general remain circular. The time evolution of this boundary (i.e. the time evolution of f) may be calculated to $O(c)$ by

$$\frac{\partial f}{\partial t} = \boldsymbol{u}\cdot\hat{\boldsymbol{r}}|_{R_0},$$

giving

$$f(\theta, t) = \tfrac{1}{24}cF_3 R_0\left(1-\frac{3}{\lambda^2}+\frac{2}{\lambda^3}\right)(\cos 2(\theta-\Omega_{\mathrm{m}} t)-\cos 2\theta).$$

We see that the boundary between the two regions has a wave-like disturbance of period π, which propagates at the same speed as the magnetic field. The sign of this disturbance is determined by F_3, which is positive for prolate spheroids, zero for spheres and negative for oblate spheroids, so this would be an amusing experiment for a direct determination of the dominant particle shape in a magnetic fluid, although, as can readily be seen, this is a small effect, the axis ratio having to be of order 10 for $\frac{1}{24}F_3$ to be approximately unity.

Thought experiment 3: magnetic-fluid core

In this thought experiment, which is included merely for the sake of completeness, the magnetic fluid and the pure solvent of experiment 2 are interchanged. The governing equations are exactly the same as for experiment 2, except for the stress jump condition at $r = R_1$, which is reversed. Thus the instantaneous velocity field to $O(c)$ is exactly equal and opposite to that for thought experiment 2, in which case, the fluid rotates *with* the magnetic field.

7. The effect of ultrasound on a magnetic fluid

In previous sections we have considered flows having small Péclet number, which allowed the stress tensor to be calculated from a quasi-steady analysis, since the macroscopic timescale is much longer than the fluid memory time $O(D)$. It is clear that a study of flows with timescales of the same order as the Brownian relaxation would reveal much more about particle sizes, and hence give better estimates of the degree of aggregation, because, the larger the aggregates or particles, the longer the relaxation time. In effect, the timescale of the flow could be tuned to the Brownian relaxation time of particles of a particular shape and size, so that measured bulk properties are most strongly influenced by these particles. It is then possible, at least in principle, to invert this information to give a distribution of particle sizes, including aggregates.

For practical reasons, the most obvious choice of flow is that induced by the propagation of an ultrasonic wave with characteristic frequency $\omega = O(D)$. If we further restrict ourselves to small-amplitude waves, it is again permissible to assume that the perturbation probability distribution P_1 is small, allowing us to linearize the diffusion equation for P. In the rest of this section, we shall also restrict ourselves, without loss of generality, to the case of monochromatic plane waves, since it is possible to recover the result for a general linear wave by superposition via an inverse Fourier transform. This is potentially very useful as some experiments use high-frequency pulses rather than continuous waves (Isler & Chung 1978).

The most often measured properties of an ultrasonic signal are the velocity and attenuation as functions of direction. The same information is obtained from each about the structure of the magnetic fluid, although, as will be mentioned later, in a stable suspension the direction-dependent part of the sound speed is relatively small

in magnitude, so for the rest of this section we shall only consider the problem of ultrasonic attenuation, as this is directly related to viscous dissipation and so is the easiest of the two phenomena to understand from a purely hydrodynamical point of view.

As the wavelength for an ultrasonic signal is very much larger than a particle lengthscale, being of the order of 1 mm for a water-based magnetic fluid, we may again use a linear approximation for the ambient flow. Taking the direction of propagation of the ultrasonic wave coincident with the x-axis, we have

$$(\nabla \boldsymbol{u})^{\mathrm{T}} = \Gamma \hat{\boldsymbol{x}}\hat{\boldsymbol{x}},$$

where Γ is a measure of the local rate of compression and is purely a function of time. However, if we assume the particles of the magnetic fluid are totally incompressible, it is more convenient to decompose $(\nabla \boldsymbol{u})^{\mathrm{T}}$ into an isotropic compression and a pure straining motion, giving

$$(\nabla \boldsymbol{u})^{\mathrm{T}} = \tfrac{1}{3}\Gamma \boldsymbol{I} + \boldsymbol{E},$$

where

$$\boldsymbol{E} = \tfrac{1}{3}\Gamma \begin{pmatrix} 2 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}.$$

The isotropic term has no effect on the orientational probability distribution, and contributes nothing to the angular dependence of the ultrasonic attenuation, and so we shall leave discussion of this term until later in the section.

Defining $\epsilon = \Gamma_0/D$, where Γ_0 is a typical value of Γ, and non-dimensionalizing $\nabla \boldsymbol{u}$ with respect to Γ_0 and other quantities as described in §2, the appropriate form of the evolution equation (4.2) for P is

$$\frac{\partial P(\boldsymbol{m},t)}{\partial t} + \nabla \cdot \left\{ \epsilon \frac{r^2-1}{r^2+1} \boldsymbol{E} \cdot \boldsymbol{m} P(\boldsymbol{m},t) + \boldsymbol{H} P(\boldsymbol{m},t) - \nabla P(\boldsymbol{m},t) \right\} = 0. \tag{7.1}$$

This implies

$$(\nabla^2 + \boldsymbol{H}\cdot\nabla - \mathrm{i}\omega)\,\phi = \epsilon \frac{r^2-1}{r^2+1} \boldsymbol{E} : \{\boldsymbol{m}\boldsymbol{H}_\perp - 3\boldsymbol{m}\boldsymbol{m}\}, \tag{7.2}$$

where $\phi = P_1/P_0$ and we have assumed that all time-dependent quantities are proportional to $\mathrm{e}^{\mathrm{i}\omega t}$. Since ϕ is proportional to $\epsilon[(r^2-1)/(r^2+1)]\boldsymbol{E}$, it may be written in the form

$$\phi = \epsilon \frac{r^2-1}{r^2+1} \boldsymbol{E} : \boldsymbol{T}$$

for some function $\boldsymbol{T}$ satisfying

$$(\nabla^2 + \boldsymbol{H}\cdot\nabla - \mathrm{i}\omega)\,\boldsymbol{T} = \boldsymbol{I} + \tfrac{1}{2}(\boldsymbol{m}\boldsymbol{H} + \boldsymbol{H}\boldsymbol{m}) - (H\cos\theta + 3)\,\boldsymbol{m}\boldsymbol{m}. \tag{7.3}$$

It follows from (7.3) that $\boldsymbol{T}$ may be written as

$$\boldsymbol{T} = \alpha(\theta; H, \omega)\,\hat{\boldsymbol{H}}\hat{\boldsymbol{H}} + \beta(\theta; H, \omega)\,(\hat{\boldsymbol{H}}\boldsymbol{m} + \boldsymbol{m}\hat{\boldsymbol{H}}) + \gamma(\theta; H, \omega)\,\boldsymbol{m}\boldsymbol{m} + \text{isotropic term},$$

where the isotropic term is unimportant as $\boldsymbol{E}$ is traceless, and $\cos\theta = \boldsymbol{m}\cdot\hat{\boldsymbol{H}}$. Substituting this expression for $\boldsymbol{T}$ into (7.3), we find

$$\alpha'' + (3\cot\theta - H\sin\theta)\,\alpha' - \mathrm{i}\omega\alpha - (4\operatorname{cosec}\theta)\,\beta' + 2H\beta = 0,$$

$$\beta'' + (5\cot\theta - H\sin\theta)\,\beta' - (2 + H\cos\theta + \mathrm{i}\omega)\,\beta - (2\operatorname{cosec}\theta)\,\gamma' + H\gamma = \tfrac{1}{2}H,$$

$$\gamma'' + (7\cot\theta + H\sin\theta)\,\gamma' - (6 + 2H\cos\theta + \mathrm{i}\omega)\,\gamma = -(H\cos\theta + 3),$$

where a prime denotes differentiation with respect to θ, and the boundary conditions for α, β and γ are simply those of regularity at $\theta = 0, \pi$.

We are now in a position to calculate the various components of the stress tensor as in §4. It is much more convenient, however, to calculate the dissipation due to the particle stress directly, as many of its terms simplify significantly on contraction with the symmetric traceless tensor $\boldsymbol{E}$. This allows much additional grouping of terms which would not have been possible otherwise, and so considerably simplifies the calculation. In this case, after some heavy algebra, the dissipation $\mathscr{D}_{\mathrm{B}}$ due to Brownian stresses, in dimensional form, is given by

$$\mathscr{D}_{\mathrm{B}} = \left[3c\mu\frac{D_0}{D}\left(\frac{r^2-1}{r^2+1}\right)^2\right]\{\langle A_3 f_1 + A_2 f_2 + A_4 f_0 + A_2 f_3 + A_1 f_4\rangle_0 (\boldsymbol{E}:\hat{\boldsymbol{H}}\hat{\boldsymbol{H}})^2$$
$$+ \langle 2B_3 f_1 + 4B_4 f_0 + B_2 f_3\rangle_0 (\boldsymbol{E}\cdot\hat{\boldsymbol{H}})^2 + \langle 2C_4 f_0\rangle_0 \boldsymbol{E}:\boldsymbol{E}\}.$$

Here

$$f_0 = 6\gamma + 2H\gamma\cos\theta,$$
$$f_1 = 12\beta - 2H\gamma + 4H\beta\cos\theta,$$
$$f_2 = 6\alpha + 2H\gamma\cos\theta,$$
$$f_3 = -4H\beta, \quad f_4 = -2H\alpha,$$

and

$$A_i = P_i(\cos\theta),$$
$$B_i = -\frac{1}{2i-1}(P_i(\cos\theta) - P_{i-2}(\cos\theta) - \tfrac{1}{4}P_{i-4}(\cos\theta)),$$
$$C_4 = \tfrac{1}{8}(\cos^4\theta - 2\cos^2\theta + 1),$$

where $P_i(\cos\theta)$ are Legendre polynomials of degree i with $P_i(\cos\theta) \equiv 0$ for i negative. The dissipation $\mathscr{D}_{\mathrm{H}}$ due to hydrodynamic particle stresses is the same as before, and is given by

$$\mathscr{D}_{\mathrm{H}} = 4\mu c\left\{(A_0(r)\,\mathscr{L}_4)(\boldsymbol{E}:\hat{\boldsymbol{H}}\hat{\boldsymbol{H}})^2 + 2\left(2A_0(r)\frac{\mathscr{L}_3}{H} + B_0(r)\,\mathscr{L}_2\right)(\boldsymbol{E}\cdot\hat{\boldsymbol{H}})^2\right.$$
$$\left. + \left(2A_0(r)\frac{\mathscr{L}_2}{H^2} + 2B_0(r)\frac{\mathscr{L}_1}{H} + C_0(r)\right)\boldsymbol{E}:\boldsymbol{E}\right\}.$$

In addition to the contributions to the dissipation already considered, we must include a contribution due to the straining motion in the ambient flow, namely $2\mu\boldsymbol{E}:\boldsymbol{E}$, and one due to the isotropic compressional motion, accurate to $O(c)$, although these terms do not contribute to the angular dependence.

Attenuation coefficient

The time-averaged internal energy density $\bar{U}$ due to the acoustic wave is given by

$$\bar{U} = K\overline{\Gamma^2}.$$

The overbar denotes a time average over several cycles and $K = K_0(1-c) + K_1$, where K_0 is a material constant, which is the value of K for a pure solvent and K_1 is the contribution to K from the entropic energy. Assuming that the change in $\bar{U}$ over one wavelength is small compared with $\bar{U}$, $\bar{U}$ satisfies

$$\boldsymbol{c}_{\mathrm{s}}\cdot\nabla\bar{U} = -\bar{\mathscr{D}}_{\mathrm{T}},$$

where $\boldsymbol{c}_{\mathrm{s}}$ is the velocity of the signal, and $\mathscr{D}_{\mathrm{T}}$ is the total dissipation, giving the attenuation coefficient

$$\alpha_0 = \frac{\bar{\mathscr{D}}_{\mathrm{T}}}{c_{\mathrm{s}}K\overline{\Gamma^2}},$$

which is therefore independent of Γ. Although, at $O(c)$, c_s and K depend on the magnetic field, this dependence is only via entropic or thermodynamic energies, which, in practice, are always small compared with the compressional energy in the solvent, since they contain a factor V_{molecule}/V_0 (where V_{molecule} is the space occupied by a molecule).

Shear waves

On ultrasonic timescales, a magnetic fluid behaves very much as a viscoelastic material, which gives rise to the possibility of ultrasonic shear (or transverse) waves. These may be analysed in essentially the same way as the compressional waves already considered.

For the propagation of a wave, it is necessary for the energy to oscillate between two forms commonly called 'kinetic' and 'potential' energy. In this case, the 'kinetic' energy is simply the kinetic energy of the bulk flow, and the 'potential' energy is the entropic energy, or the work done by the bulk motion in rotating the suspended particles away from their equilibrium orientations. If the frequency ω of the shear wave is much less than the Brownian diffusivity, the system is heavily damped because almost all the entropic energy will diffuse away in one cycle, in which case the fluid behaves very inelastically. However, even if ω is greater than or the order of D, there is substantial dissipation due to hydrodynamic forces, since the entropic driving force is $O(c)$ and the viscous dissipation is $O(1)$. Thus the amplitude of the shear waves decays by a factor $O(c)$ in one wavelength. It can be shown, by a simple order-of-magnitude calculation, that the shear waves propagate with velocity $O(nmH/\rho)$, where ρ is the density of the magnetic fluid; this velocity is approximately equal to 1 m s^{-1} for a standard suspension. However, these waves are of little practical interest because since they are unable to propagate much further than the order of 1 μm, energy transport due to shear waves can be neglected.

Discussion of experimental results

The direction-dependent part of α_0 is thus given by

$$\alpha_0^{\mathrm{A}} = (a_1\overline{(\boldsymbol{E}:\hat{\boldsymbol{H}}\hat{\boldsymbol{H}})^2} + a_2\overline{(\boldsymbol{E}\cdot\hat{\boldsymbol{H}})^2})/\overline{\Gamma^2},$$

where a_1, a_2 may be calculated from the dissipation due to the particle stress. Unfortunately no result of this general form gives a satisfactory fit to the data of Isler & Chung (1978), but it is clear that, correct to $O(c)$, the direction-dependent part of the attenuation of a suspension must be of this form, assuming that particles are small compared with the wavelength of the signal. So we must conclude that in the experiment of Isler & Chung (1978) particle interactions are important and dominate over the $O(c)$ result. This is then a strong indication of aggregation, and other anomalous effects which were mentioned by Isler & Chung could also indicate aggregation. In a paper by Chung & Isler (1978), response times of several seconds are measured, indicating the presence of very large aggregates. It is possible to have large response times in a stable magnetic fluid if it is near its Curie temperature (defined in §3), where large numbers of particles will behave in a similar manner to the domains in a ferromagnet, although this will only occur at temperatures much lower than those in the two papers mentioned above. However, in a theoretical paper by Gotoh, Isler & Chung (1980), using a theory with adjustable parameters, they obtain a remarkable fit with the data of Isler & Chung (1978), although they seem to have overlooked the fact that their theoretical curve of attenuation versus direction is not analytic everywhere, contrary to what would be possible from any suspension-mechanically based theory.

8. Conclusions

In this paper we have derived equations to describe the rheology of magnetic fluids in most situations of practical interest. In particular, the main results of this paper are summarized below.

1. The bulk magnetization is determined for a fluid at rest as a function of the applied magnetic field, with inclusion of the effect of the magnetic interactions between particles (§3). The first correction to the Langevin equation for magnetization thus derived significantly improves the agreement between predicted and measured magnetic pressures (see figure 2).

2. The bulk stress tensor is calculated, in the limit of small Péclet number, including the effects of the time and space dependences of the magnetic field (§4). (The time dependence of the bulk flow can be neglected at the order of the calculation.) For calculational purposes it is assumed that the particles are spheroidal; the calculation for general particle shapes has yet to be performed. It is noteworthy, however, that the tensorial structure of the stress derived here (4.12) is the most general available (which is not the case for spherical particles), so that even for more complex particle shapes only the multiplying coefficients will be altered. These results are then applied to shear and pipe flows (§5), as flows of this type appear in many applications where magnetic fluids are used as lubricants or magnetic seals. When comparing theory and experiment, one should note that real magnetic fluids are polydisperse, though for all the experiments performed to date the particle size and shape distributions were not measured. Simply choosing the distribution that gives 'best fit' could well give misleadingly good results, since this distribution might be far from the actual one. For this reason comparison between theory and experiment is restricted in this paper to monodisperse suspensions for which the agreement is found to be fair.

3. A magnetic suspension affords a rare example of a material for which the stress tensor is non-symmetric. Some of the surprising flow consequences are explored in §6 by means of simple thought experiments involving rotating magnetic fields.

4. Finally, in §7 we analyse the attenuation of ultrasound with characteristic frequency of the same order as the microscopic relaxation time (due to Brownian motion). We also explain how the results obtained could be used as a sensitive means of determining the degree of aggregation.

The next step in this field must almost certainly be experimental; in particular, new shear and pipe flow experiments, with magnetic field direction not aligned with any of the principal axes of flow (as described in §5) would be of interest, especially for fluids with known particle size and shape distributions. For a thorough test of the results in this paper, they should be used to determine the macroscopic behaviour of systems with space- and time-varying magnetic fields. As mentioned, ultrasound experiments are potentially useful in determining the degree of aggregation; however, for a direct test of the theory, careful experiments with no aggregation need to be performed.

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