Bulk Flow in Ferrofluids in a Uniform Rotating Magnetic Field

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Direct measurements of the bulk flow of a ferrofluid in a uniform rotating magnetic field were obtained using the ultrasonic velocity profile method. The fluid was observed to corotate with the field in a rigid-body-like fashion throughout the bulk of the container, except near the air-fluid interface, where it was observed to counterrotate. The results were found in qualitative agreement with the spin diffusion theory of Zaitsev and Shliomis [J. Appl. Mech. Tech. Phys. **10**, 696 (1969)].

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In 1967, Moskowitz and Rosensweig [1] reported an experiment where flow of a ferrofluid was obtained in a stationary cylindrical container when placed in a uniform rotating magnetic field. This phenomenon, referred to as spin-up flow, has since captured the attention of many scientists. Unfortunately, experimental measurements of the bulk flow had not been made, precluding direct comparison of the phenomenon to theories presented in the literature.

The first such theory is due to Zaitsev and Shliomis [2], who predicted corotation of fluid and field in the bulk, with a rigid-body-like azimuthal velocity profile throughout the core and a thin boundary layer near the fluid container interface. It is important to note that, according to Zaitsev and Shliomis, there would be no flow in a uniform rotating magnetic field unless the spin viscosity is nonzero, that is, unless the balance of internal angular momentum in the ferrofluid includes the effect of the couple stress [3,4]. This result was verified theoretically by Jenkins [5] and Glazov [6], who concluded that the observed flow must be due to field inhomogeneity.

Confusion arose when Brown and Horsnell [7] made observations similar to those of Moskowitz and Rosensweig [1] but noted that the fluid and field counterrotate with respect to each other. A partial explanation was provided by Rosensweig *et al.* [8], who made measurements on concave and convex free surfaces. They observed counterrotation of fluid and field for a concave surface and corotation for a convex surface. Based on these observations, they concluded that the spin-up phenomenon is produced by surface stresses rather than volumetric effects (i.e., body couples) and that bulk flow should be negligible compared to the surface flow. In 1987, Shliomis *et al.* [9] suggested that this flow is produced by nonuniformity of magnetic permeability due to radial temperature gradients produced by the microeddies that arise around the rotating particles, especially at high frequencies. Based on this assumption, Pshenichnikov *et al.* [10] obtained a theoretical expression for the velocity profile in the bulk of a nonisothermal fluid, predicting counterrotation between flow and field for frequencies lower than $10^5\ \mathrm{s}^{-1}$ and a velocity maximum at $r \quad R/3^{1/3}$, where R is the radius of the container, independent of the properties of the ferrofluid. For weak fields and low frequencies (isothermal fluid), they claimed the flow was driven by tangential magnetic stresses on the free surface of the fluid.

We note that the observations of Moskowitz and Rosensweig [1], Brown and Horsnell [7], and Rosensweig *et al.* [8], as well as most others reported, were all made at the ferrofluid top free surface. Because the fluid is opaque, no direct unobtrusive measurements of the bulk flow had been made. Because the spin diffusion theory of Zaitsev and Shliomis [2] and the temperature gradient theory of Pshenichnikov *et al.* [10] apply only in the bulk, the need remained for such direct bulk measurements.

Here we report direct measurements of bulk velocity profiles for water- and kerosene-based ferrofluids in a uniform rotating magnetic field. These were obtained using the ultrasonic velocity profile (UVP) method, which is based on pulsed ultrasound echography. The technique uses ultrasound pulses emitted from a transducer into the liquid. These pulses are reflected by small tracer particles in the fluid, and the echo is collected by the same or another transducer. A subsequent analysis of the echo signal yields the velocity component parallel to the direction of pulse propagation [11]. The principal advantages of the method are summarized by Takeda [12], the most relevant being the possibility of obtaining spatiotemporal information of the flow in opaque liquids.

The water- (EMG 705) and kerosene-based (EMG 900) ferrofluids used in these experiments were obtained from Ferrotec Corporation. Fluid characterization and all experiments described below were carried out within two weeks of receiving the ferrofluid to avoid particle agglomeration, which commonly occurs as commercial water-based ferrofluids age.

The values of mass density for EMG 705 and EMG 900 determined gravimetrically were 1220 and 1270 $\mathrm{kg/m^3}$, respectively, while shear viscosity values of 0.0025 and 0.0077 $\mathrm{N\,s/m^2}$ were measured using a STRESSTECH HR rheometer (ATS Rheosystems, Bordentown, New Jersey) with double-gap geometry in a shear rate range of 10–110 s^{1} in the absence of magnetic field. The ferrofluid showed Newtonian behavior under these conditions. The magnetic properties of the ferrofluids were measured using an MPMS-XL7 SQUID magnetometer (Quantum Design, San Diego, California) at 300 K. From analyzing the high-field asymptotes of the magnetization curves, we obtain saturation magnetizations (M_S) of 17.4 and 38:1 kA m^{1}, respectively. The magnetic volume fractions were estimated to be $_{EMG\ 705}$ 0:035 and $_{EMG\ 900}$ 0:085, using the relation M_S M_d and a value of M_d 446 kA=m for the domain magnetization of magnetite. From the low-field slope of the magnetization curve, we obtained the initial susceptibility $_{0;EMG\ 705}$ 1:4 and $_{0;EMG\ 900}$ 1:3.

From fitting the low-field and high-field asymptotes of the Langevin equation to the magnetization measurements, using a log-normal particle size distribution [13], we estimated the volume median particle diameter and geometric deviation, respectively, to be 12 nm and 0.39 for EMG 705 and 8.7 nm and 0.40 for EMG 900. Using the volume median particle diameter, the Brownian relaxation times ($_B$) were calculated to be approximately 3:9 10^{6} s for EMG 705 and 5:9 10^{6} s for EMG 900, assuming a surfactant coating thickness of 2 nm, whereas the Néel relaxation time ($_N$) was estimated to be approximately 2:5 10^{2} s for EMG 705 and 6:6 10^{7} s for EMG 900, using a value of 78 000 J=m^3 for the magnetocrystalline anisotropy constant of the magnetite nanoparticles [14] and $_0$ 10^{9} s.

The speed of sound ($_s$) in the ferrofluids were determined to be $_{s;EMG\ 700}$ 1450 m=s and $_{s;EMG\ 900}$ 1071 m=s by measuring the time required for a pulse to traverse a known distance.

A rotating magnetic field was obtained using balanced three-phase currents to excite a two-pole, three-phase stator winding. The amplitude, frequency, and direction of the field can be controlled using the signal generator or linear amplifier gain. The length and diameter of the stator winding were 77.7 and 63.6 mm, respectively. Measurements of the magnetic fields produced by the stator, in the absence of the container and ferrofluid, were made using a gauss meter (Sypris Test & Measurement, Orlando, Florida) with a three-axis probe (Model ZOA73). From these measurements, we determined that the stator produces a magnetic field of 4.15 mT rms per ampere rms of input current. The radial field inhomogeneity was determined to be 2.2% from axis to outer container radius at midheight, with a maximum of 6% at the top of the container, while axial field inhomogeneity was 4% from middle to 3=4 height and 21% to top, along the gap axis.

Bulk velocity profiles were obtained using a DOP2000 ultrasound velocimeter (Signal Processing, Lausanne, Switzerland). The equipment measures the component of the velocity vector parallel to the direction of beam propagation, with the spatial location being determined from the time delay between emitted burst and recorded echo and the magnitude of velocity being determined from the correlation of the random echo signals from a pulsed emission train. The velocity signal is positive when tracers are moving away from the probe. Assuming that flow is purely in the azimuthal direction, the azimuthal component V of the velocity can be related to the measured velocity V_k (parallel to the ultrasound pulse) by:

$$V \quad V_k \frac{\sqrt{R^2 \quad x_p^2 \quad 2R\, x_p \cos}}{R \sin}; \qquad (1)$$

where R is the radius of the container, x_p is position along the beam path, and is the angle between the probe and the diagonal. Because the magnetite particles suspended in a ferrofluid are too small to produce a UVP signal, we used polyamide powder as a tracer. These microparticles had a mean diameter of 15–20 m and density of 1:13 g cm^{3}. Minute amounts were used, such that the estimated weight fraction of tracer particles was less than 0.000 15. Using such low tracer fractions ensures that the measured profiles are representative of the ferrofluid flow, avoiding other phenomena such as are found in so-called inverse ferrofluids [15]. Our transducers had an emission frequency of 4 MHz, a diameter of 8 mm, and a length of 10 mm. We used a pulse repetition frequency of 200 Hz, taking 100 emissions per profile and averaging over 70 profiles to obtain our reported results. The DOP averages the tracer velocities in a sampling volume with cylindrical shape, whose diameter is 3 mm and length along the beam propagation direction is 1.45 mm. By using overlapping sampling volumes, the spatial resolution along the beam propagation direction can be made as small as 0.36 mm. These conditions ensured a spatial resolution of 0.36 mm and a maximum measurable velocity of 18:12 mm=s for the component parallel to the ultrasound beam.

Velocity profiles were measured for the ferrofluids in a cylindrical container of 49.4 mm inner diameter and 63.5 mm height. Our apparatus design, illustrated in Fig. 1, permitted simultaneous measurement of velocity profiles using ultrasonic beams propagating at three distinct angles (using three transducers) relative to the diagonal. The probes were adjusted in external slots around the container to avoid disturbing the flow field and to control the angle between probe and the diagonal. The container material was polycarbonate to avoid internal reflections in the wall, which would produce atypical saturation of the transducer. A specially designed cover was used to suppress the surface effect reported by Rosensweig *et al.* [8] and to measure the axial velocity component of the flow field.

The rotating magnetic field was generated using the three-phase, two-pole magnetic induction motor stator winding described above. Experiments were carried out for field frequencies of 50, 65, 75, 85, and 100 Hz and amplitudes of 6.2, 8.3, 10.3, 12.3, 14.3, and 16.5 mT rms.

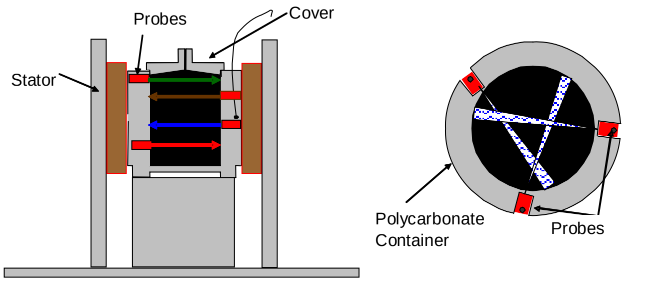

FIG. 1 (color online). Illustration of the experimental setup for measuring velocity profiles of ferrofluids in a cylindrical container and subjected to a uniform rotating magnetic field. Left: Container with ultrasonic transducers located inside a two-pole induction motor stator. Right: Top view showing transducers at different incident angles. The transducers are separated from the ferrofluid by a thin polycarbonate wall.

The ultrasonic transducers were placed at different heights of the container or at three different angles with respect to the diagonal. The experiments were carried out in intervals in order to avoid temperature increases of more than 2 C. The radial temperature profile was monitored, and the maximum observed radial temperature variation was 0.2 C. The limitations of the method are the inability of obtaining measurements within 5–8 mm of the ultrasonic transducer and inaccuracy in the velocity measurements at the far wall (at the boundary of the container). The first limitation is due to saturation of the transducer receiver because the same probe acts as emitter and receiver of the acoustic signal. The second limitation arises because the ultrasonic beam is reflected by the far wall, introducing errors in the measured velocity.

In the first series of experiments, transducers were placed at half the height of the container, and a cover was used to suppress surface driven flow. Three transducers, at angles of 7.1 , 11.7 , and 14.2 , were used, and a counterclockwise rotating field was applied such that a positive velocity measurement implied corotation of field and fluid. Figure 2 shows the velocity profiles for a frequency of 85 Hz and applied magnetic field amplitude of 10.2 and 14.3 mT rms for EMG 705 and 8.6 and 10.5 mT rms for EMG 900. These velocity profiles show a linear dependence of the velocity with radial position (i.e., rigid-body-like motion) over most of the fluid, with corotation of field and fluid in agreement with the predictions of Zaitsev and Shliomis [2]. We note that the maximum velocity occurs at different radial positions for the two ferrofluids. For EMG 705 the maximum occurs at an approximate radius of 17 mm, whereas for EMG 900 it occurs beyond 21 mm. The location of the maximum for the EMG 900 was not determined precisely due to limitations of the UVP method described previously. In addition, velocity profiles taken for different probes superimpose one another, confirming that the flow is purely azimuthal. The observation of corotation of field and fluid and the fact that the location of the velocity maximum depends on the nature of the fluid under similar geometric and field conditions demonstrate that the analysis of Pshenichnikov *et al.* [10] does not apply to our situation. This was to be expected, as the measured radial variation in temperature during the experiments was negligible.

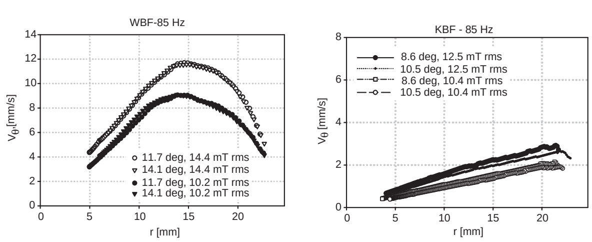

FIG. 2. Velocity profiles for the (left) water- and (right) kerosene-based ferrofluid in a cylindrical container with cover in a uniform rotating magnetic field, for three transducers at various angles with respect to the diagonal.

Another series of experiments studied the variation of velocity profile with axial position by using probes at H, $3=4H$, $1=2H$, and $1=4H$ (H 63:5 mm being the container height). Simultaneously, the axial velocities along the axis (r 0 mm) and at r 15 mm were measured. The results are shown in Fig. 3, where it is seen that the velocity field is uniform throughout most of the container and the axial velocity component is negligible compared to the azimuthal component, especially along the axis of the container. The deviation in the velocity profile measured at the top of the container is likely due to reflection of the pulse with the upper wall. The observation of uniform flow throughout the bulk and negligible axial velocity is indicative that, though the magnetic field generated in the stator varies by as much as 21% along the vertical axis (4% throughout most of the fluid), this variation does not induce appreciable flow in the axial direction. Furthermore, as noted before, the location of the velocity maximum is different for the two fluids, which is contrary to what would be expected if the flow were driven by a field inhomogeneity which is concentrated close to the container wall. Hence, it is unlikely that our observations are the result of field inhomogeneity.

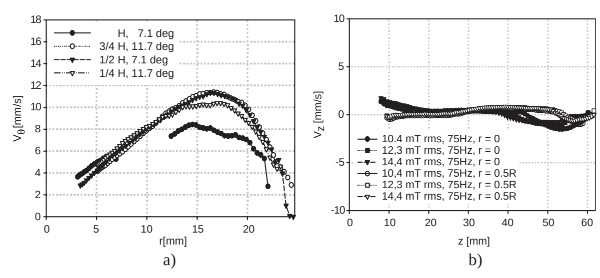

FIG. 3. (a) Velocity profiles for the water-based ferrofluid EMG 705 under an applied field of 75 Hz and 14.4 mT rms at different heights in a container with a cover, for fixed transducers with different angles with respect to the diagonal. (b) Axial velocity component measured using a probe placed on the fixed top cover (z measured from the top).

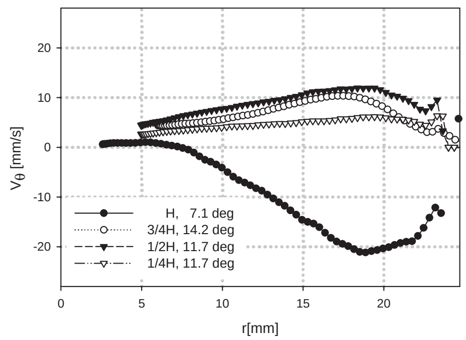

FIG. 4. Velocity profiles for the water-based ferrofluid EMG 705 under an applied field of 75 Hz and 10.3 mT rms at different heights in a container without a cover. The negative velocities indicate counterrotation of fluid and field.

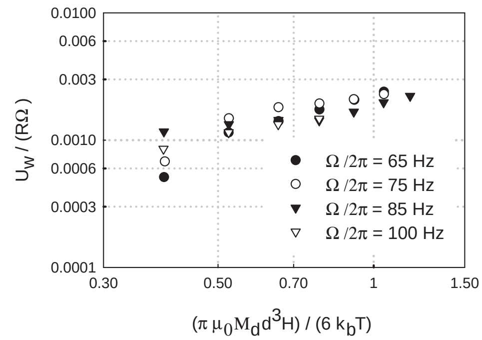

FIG. 5. Log-log plot of the dimensionless core fluid rotation rate versus the applied magnetic field amplitude, made dimensionless using the Langevin parameter. The slope of the curve is near unity (0.997), indicating a power-of-one dependence of the velocity with the applied field amplitude.

A third series of experiments was carried out under the same conditions but using an open container. In these experiments, the top probe was placed 4 mm below the free surface (measured from the probe center), to avoid reflections of the pulse. The velocity profiles in Fig. 4 show a transition from corotation of field and fluid in the bulk to counterrotation close to the free surface. These observations are in agreement with the measurements of Rosensweig *et al.* [8], but they demonstrate that the bulk flow is not negligible in comparison to the surface flow. Both the surface driven flow and the body-couple induced flow coexist under our experimental conditions.

In Fig. 5, we summarize the experimental values obtained for the dimensionless core fluid rotation rate for three different frequencies and six values of magnetic field strength for the EMG 705 ferrofluid. The slope of the log-log plot is close to unity (0.997), indicating a power-of-one dependence of the velocity profile on the field frequency and amplitude. The analysis of Zaitsev and Shliomis [2] predicts a power-of-one dependence of the velocity with field frequency. The dependence on field amplitude predicted by their analysis is determined by the equilibrium magnetization response of the ferrofluid, as described by the Langevin equation. For low values of the Langevin parameter $_0M_d d^3H=6k_BT$ 1, the equilibrium response is linear and the azimuthal velocity is expected to have a power-of-two dependence with field amplitude. As the field increases, the Langevin parameter increases and the fluid eventually becomes saturated. Under such conditions, the azimuthal velocity is expected to have a power-of-one dependence with field amplitude. As can be seen from Fig. 5, the Langevin parameter under our experimental conditions is of the order of unity; hence, nonlinear saturation effects become relevant.

In conclusion, our measurements show that body-couple induced flow indeed arises in the bulk of a ferrofluid subjected to a rotating uniform magnetic field and that the observed profiles are in qualitative agreement with the analysis of Zaitsev and Shliomis [2]. Rigid-body-like rotation of the fluid is observed except close to the container wall. The fluid corotates with the field, and the magnitude of the flow shows a linear dependence with field frequency and with field amplitude. Measurements at various heights confirmed that the observed flow is representative of the bulk and the flow has a negligible axial component. Measurements with a free surface demonstrated that, though the fluid counterrotates with the field at the surface, it corotates in the bulk.

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