# Onset of Particle Rotation in a Ferrofluid Shear Flow

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A ferrofluid shear flow subjected to a magnetic field is investigated in the limit where viscous and magnetic forces prevail over thermal fluctuations. When the viscous torque slightly exceeds the magnetic hold torque the nanoparticles start to rotate anharmonically. By means of a weak field modulation, the rotation of the particles can be phase synchronized generating an overproportional ac contribution to the magnetization. The investigation is theoretical and based on the Fokker-Planck equation for a monodisperse ferrofluid of noninteracting spherical rigid dipoles. A resonance experiment is proposed by which the onset of this coherent particle rotation can be measured. [S0031-9007(99)09097-3]

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A ferrofluid is a colloidal suspension of ferromagnetic monodomain nanoparticles in a liquid carrier [1]. The fluid behaves like a paramagnetic gas of high permeability. The magnetic relaxation is dominated by two distinct mechanisms: The Néel relaxation describes the reorientation of the magnetic moment relative to the atomic lattice. This is in contrast to the Brownian process, where the magnetic moment is tightly fixed to the crystal orientation (rigid dipole), and relaxation takes place via rotation of the whole particle relative to the fluid. For sufficiently large particles (magnetic core diameter $\gtrsim 10$ nm for magnetite) the Brownian mechanism prevails. One of the most exciting properties of ferrofluids is related to the coupling of the microscopic particle rotation to the macroscopic vorticity of the flow. In a static magnetic field the magnetic torque prevents particles from rotating and thus causes an extra viscous dissipation in the carrier liquid, which leads to an enhanced effective viscosity [2,3]. This is the so-called rotational viscosity. On the other hand, an ac magnetic field with frequency $\omega$ close to the local vorticity $\Omega = \frac{1}{2}\nabla \times \mathbf{v}$ can resonate with the particle's rotation. Depending on whether $\omega < \Omega$ or $\omega > \Omega$ energy is transferred from the flow field into the magnetic field or vice versa. In the former case the particle rotation is slowed down and an extra ac magnetization component transverse to the applied field can be detected (magnetovortical resonance) [4,5]. In the opposite case $\omega > \Omega$ the ac field speeds up the particle's rotation, which leads to a negative viscosity increment [6–8] associated with the expression "negative viscosity."

The above mentioned phenomena are intimately related to the presence of suspended particles in the fluid. In weak magnetic fields the particle rotation follows the local vorticity of the flow, while in strong fields the dipole orientation is held fixed by the magnetic torque. It is thus a challenging problem to design an experimental arrangement, by which the onset of particle rotation can be detected. The idea is that a weak modulation superimposed to a strong static magnetic field gives rise to an overproportional (nonlinearly enhanced) resonance signal as soon as particle rotation has set in. In what follows this question will be addressed in detail. For simplicity a monodisperse ferrofluid of noninteracting rigid dipoles (spheres with point dipoles at their centers) is considered. Each particle has a hydrodynamic volume $V$ (magnetic core plus surfactant shell) and carries a magnetic moment $\mathbf{m} = m\mathbf{e}$, where $\mathbf{e}$ is a unit vector. Even though real ferrofluids are polydisperse, only the larger Brownian particles promote the desired effect; the smaller Néel particles do not contribute.

The system under consideration is a stationary plane Couette flow $\mathbf{v}(\mathbf{r})$ with spatially homogeneous vorticity $\boldsymbol{\Omega}\mathbf{e}_y = \frac{1}{2}(\nabla \times \mathbf{v})$ (see Fig. 1). The ferrofluid with dynamic viscosity $\eta$ is exposed to a homogeneous magnetic field $\mathbf{H}_0 = H_0\mathbf{e}_z$. Although thermal fluctuations are crucial for the particle dynamics in ferrofluids, let us first consider the deterministic motion as it is useful to outline

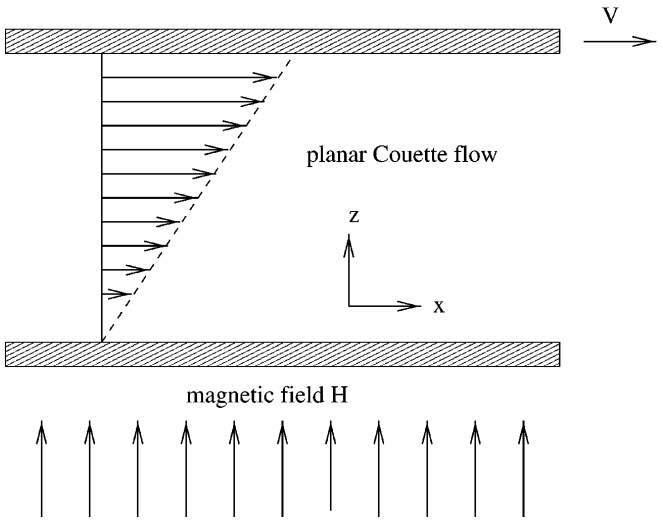


FIG. 1. A stationary plane Couette shear flow $\mathbf{v}(\mathbf{r})$ with uniform vorticity $\boldsymbol{\Omega}\mathbf{e}_y = \frac{1}{2}(\nabla \times \mathbf{v})$ is exposed to a homogeneous magnetic field $\mathbf{H} = H\mathbf{e}_z$.

  

the idea. Ignoring inertial effects, the dipole orientation $\mathbf{e}(t) = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ obeys an evolution equation given by Ref. [9]. To simplify the discussion we discuss here the two-dimensional rotation in the $x$-$z$ plane (i.e., $\phi = 0$) governed by

$$\frac{1}{\Omega}\frac{d\theta}{dt} = \frac{1}{\kappa}\sin\theta + 1\,. \tag{1}$$

Equation (1) reflects the angular momentum balance between the magnetic body couple $\mathbf{m} \times \mathbf{H}_0$ and the viscous friction $6\eta V\Omega\mathbf{e}_y$. The latter one is the classical hydrodynamic expression for the viscous torque of a macroscopic sphere floating in a carrier fluid [10]. Equation (1) (or its 3D generalization) can be integrated analytically [9]. The character of the solution depends on the nondimensional shear rate $\kappa = \frac{6\eta V\Omega}{mH_0}$. For $|\kappa| < 1$ the solution relaxes to a stationary state with $\sin\theta = \kappa$ and $\phi = 0$. On the other hand, for $|\kappa| > 1$ the azimuthal angle $\phi$ again relaxes to 0 but $\theta(t)$ becomes oscillatory indicating that particle rotation has set in. For $|\kappa|$ close above unity the rotation is extremely anharmonic but becomes increasingly uniform as $|\kappa|$ rises (Fig. 2). The particle's rotation frequency $\omega_P$ is independent of the initial orientation [9] and given by

$$\omega_P = \Omega\sqrt{1 - \frac{1}{\kappa^2}}\,. \tag{2}$$

Clearly, the outlined deterministic one-particle dynamics is not appropriate for the description of the many body system ferrofluid. To obtain a macroscopic magnetization

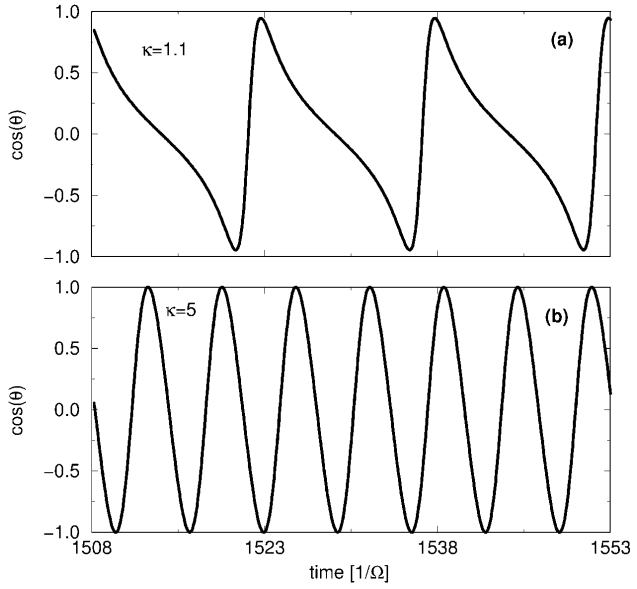


FIG. 2. The longitudinal component $\cos\theta$ of the orientational unit vector $\mathbf{e}(t)$ as a function of time. The curves are calculated by numerical integration of Eq. (1) with a dimensionless shear rate $\kappa = 1.1$ (a) and $\kappa = 5$ (b). Within the depicted time interval initial transients have died out and a steady state rotation has been established. For $\kappa \to 1^+$ the rotation becomes increasingly anharmonic, simultaneously the oscillation period $\omega_P$ diverges.

signal, two principal difficulties are to be overcome: (i) the rotation of all particles must be phase synchronized such that different dipole fields interfere constructively, and (ii) the Brownian motion, which destroys any kind of coherent particle motion, must be limited to a minimum amount.

Problem (i) can be solved by weakly modulating the applied magnetic field with a frequency $\omega$ close to the particle rotation frequency $\omega_P$. By replacing the static magnetic field $H_0$ by $H(t) = H_0(1 + \varepsilon\cos\omega t)$ with $\varepsilon \ll 1$, the weak ac modulation acts like a clock generator. Any particle that does not perform an in-phase rotation experiences a small restoring force until it follows the prescribed phase. This technique of phase locking has been checked by numerical integration of Eq. (1): Several runs with distinct initial dipole orientations lead always to the same long time phase behavior. Even if $\omega$ deviates from the optimum value $\omega_P$ by 20%, a modulation amplitude of $\varepsilon = 0.1$ suffices to make the phase synchronization work. Obviously the effect of this tiny modulation is enormous: Under stationary conditions ($\varepsilon = 0$) all particles rotate out of phase as they start from random initial conditions. Since their rotation is anharmonic, a finite but *stationary* magnetization remains. With the modulation switched on, the phase-synchronized dipole rotation generates an overproportional *ac contribution* to the magnetization, which can be macroscopically detected.

The preceding discussion is based on the deterministic evolution equation (1). To investigate whether the coherent dipole rotation persists under the influence of the thermal motion [problem (ii)] the investigation must be based on a stochastic approach. The probability distribution function (PDF) $P(\mathbf{e}, t)$ for the dipole orientation $\mathbf{e}$ is governed by the Fokker-Planck equation

$$2\tau_B\frac{dP}{dt} = \{-\alpha\mathbf{R}\cdot[P\mathbf{R}(\mathbf{e}\cdot\mathbf{e}_z)] - 2\tau_B\Omega\mathbf{e}_y\cdot(\mathbf{R}P)\} + \mathbf{R}^2P\,. \tag{3}$$

Equation (3) describes rotary diffusion with additional contributions from magnetic and viscous torques [11–13]. Here $\tau_B = 3\eta V/(k_BT)$ denotes the Brownian relaxation time, $k_B$ is the Boltzmann constant, and $T$ is the temperature. Furthermore $\mathbf{R} = \mathbf{e} \times \partial/\partial\mathbf{e}$ is the angular momentum operator and $\alpha(t) = mH(t)/(k_BT)$ is the Langevin parameter. Since the focus of this paper is on a deterministic effect, the magnetic energy must dominate over thermal fluctuations, i.e., $\alpha_0 = mH_0/(k_BT) \gg 1$. Furthermore, since the particle rotation starts at $|\kappa| = 1$, the condition $2\tau_B\Omega = 6\eta V\Omega/(k_BT) = \alpha_0\kappa \gg 1$ is necessary as well. Because of the smallness of the Brownian relaxation time the last requirement is not easy to fulfill. Indeed, most ferrofluid experiments are done in the limit $\tau_B\Omega \ll 1$. Nevertheless, by the use of high viscosity carrier fluids (e.g., pure glycerol as in Refs. [4,5]) Brownian relaxation times up to 10 ms can be achieved. The condition $\tau_B\Omega \gg 1$ prevents Eq. (3) from being solved by a Taylor expansion

in this quantity as previously done in Ref. [7]. Neither is a linearization around the equilibrium magnetization appropriate since deviations are large by virtue of the strong shear rate. Accordingly direct integration of Eq. (3) will be applied here. The decomposition of the PDF into spherical harmonics

$$P(\mathbf{e},t) = P(\theta,\phi,t) = \sum_{l=0}^{\infty}\sum_{m=-l}^{+l} a_{l,m}(t)Y_l^m(\theta,\phi) \quad (4)$$

transforms Eq. (3) to an infinite set of coupled ordinary differential equations, which can be numerically treated by an appropriate cutoff at $l \le l_{\max}$, say.

The physical observable of the system is the macroscopic magnetization $\mathbf{M}(t)$ related to the mean dipole orientation by [14]

$$\mu_0 \mathbf{M}(t) = mN\langle \mathbf{e} \rangle . \quad (5)$$

Here $\mu_0$ is the vacuum permeability, $N$ is the number of particles per volume, and $\langle \ldots \rangle$ denotes the statistical average evaluated with the time dependent PDF $P(\mathbf{e},t)$. In terms of the expansion coefficients $a_{l,m}$ one has for the longitudinal component $M_{\parallel}(t) = \mathbf{M}(t) \cdot \mathbf{e}_z$

$$\langle \cos\theta \rangle = \sqrt{\frac{4\pi}{3}}\, a_{1,0} . \quad (6)$$

In what follows results of a "numerical resonance experiment" with Eqs. (3) and (4) are presented. The data are obtained on the basis of the following protocol: A modulated magnetic field is imposed with $\alpha(t) = 10 \times (1 + 0.1\cos\omega t)$. The shear rate $\kappa$ is varied between 0 and 2.5 to pass over the onset of the particle rotation at $\kappa = 1$. On keeping $\kappa$ fixed the modulation frequency is scanned from $\omega = 0$ to $\omega = 2\Omega$ to detect the resonance maximum, which is expected at $\omega = \omega_P$. On starting from random initial conditions for the $a_{l,m}$ the evolution equations are solved by a Runge-Kutta procedure. For all runs the cutoff $l_{\max} = 10$ has been used. After the decay of initial transients the periodic time signal of $a_{1,0}(t)$ (longitudinal component of the magnetization) is decomposed into Fourier amplitudes $|A_n|$ related to the frequency channels $n\omega$, where $n = 0, 1, 2, \ldots$. The static contribution $|A_0|$ is rather insensitive to variations of $\omega$, while the fundamental mode $|A_1|$ exhibits a pronounced maximum at $\omega = \omega_{\max}(\kappa)$ (see Fig. 3). This resonance, which occurs at $\kappa \gtrsim 1$, clearly reflects the onset of particle rotation. As $\omega$ approaches $\omega_{\max}$ the phase locking mechanism becomes increasingly effective, a growing number of particles rotates coherently, generating a modulation ratio $|A_1(\omega_{\max})|/|A_0(\omega_{\max})|$ which can easily exceed 100%. Recall that the modulation ratio of the excitation signal $H(t)$ is only $\varepsilon = 10\%$. The square symbols in Fig. 4 demonstrate that the $\kappa$ dependence of the resonance frequency $\omega_{\max}$ agrees fairly well with the deterministic relation $\omega_P(\kappa)$ (solid line). Obviously, under the influence of the thermal fluctuations the onset of particle rotation is shifted towards smaller $\kappa$.

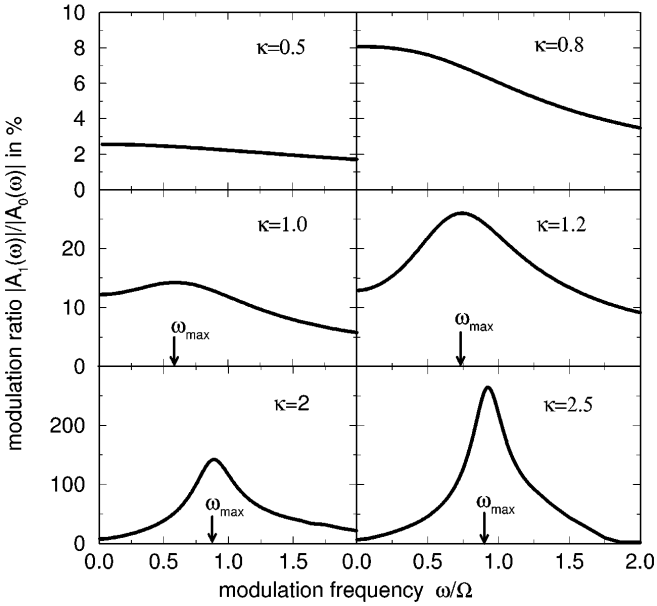


FIG. 3. The modulation ratio $|A_1(\omega)|/|A_0(\omega)|$ of the magnetization $M_{\parallel}(t)$ as a function of the excitation frequency $\omega$. Dimensionless shear rate $\kappa$ as indicated. Resonance maxima appear, provided $\kappa \gtrsim 1$ (onset of particle rotation).

It is also instructive to study the degree of anharmonicity of the rotation. To this end a quantitative measure can be established by $\Gamma = \sum_{n=2}^{5} |A_n| / \sum_{n=1}^{5} |A_n|$, which relates the power in the higher harmonics to the total oscillation intensity [15]. In the inset of Fig. 4 it is shown that the anharmonicity goes down with increasing shear rate as predicted from the deterministic particle dynamics shown in Fig. 2.

In what follows a possible experimental setup for the observation of the coherent particle rotation is proposed.

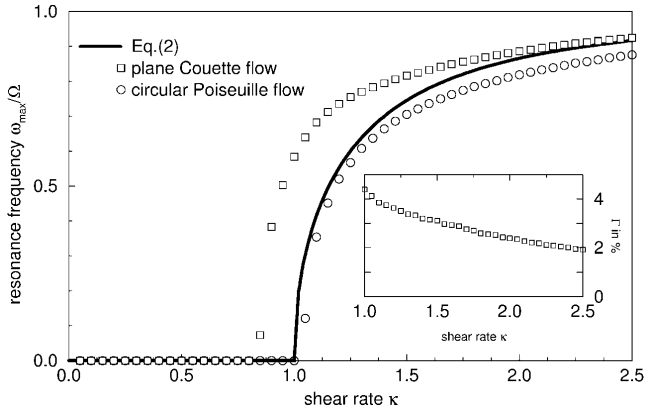


FIG. 4. The resonance frequency $\omega_{\max}$ as a function of the shear rate $\kappa$. Solid line: $\omega_P(\kappa)$ according to Eq. (2). Symbols represent simulation results with the Fokker-Planck equation (3) for the plane Couette flow (squares) and a circular Poiseuille flow (circles). In the latter case $\kappa$ denotes the shear rate at the inside of the capillary wall. Inset: By raising the shear rate $\kappa$ the anharmonicity $\Gamma$ goes down, indicating that the particle rotation becomes increasingly uniform.

Since a plane Couette flow is inconvenient for an experimental realization, a capillary tube flow will be treated here with an axis parallel magnetic field. Since the shear rate $\kappa$ within a Poiseuille tube flow is nonuniform ($\kappa = 0$ at the center and $\kappa_{\max}$ at the wall), cylindrical shells of different radii superimpose to the effective magnetization signal $M_{\|}(t)$. The resulting Fourier amplitude $|A_1^{\text{Poiseuille}}|$ is thus a weighted superposition of the form

$$|A_1^{\text{Poiseuille}}(\kappa_{\max}, \omega)| = \left| \frac{2}{\kappa_{\max}^2} \int_0^{\kappa_{\max}} A_1(\kappa, \omega) \kappa \, d\kappa \right|, \tag{7}$$

where $A_1(\kappa, \omega)$ is taken from the Couette flow as depicted in Fig. 3. Although this averaging procedure leads to a resonance broadening, the peaks at $\omega_{\max}$ are still easy to identify (see Fig. 5). The open circles in Fig. 4 show that the shear rate dependence of $\omega_{\max}$ for the tube flow is in favorable agreement with the deterministic result $\omega_P(\kappa)$.

In order to meet the condition $\tau_B \Omega \simeq 10$, a high viscosity ferrofluid (e.g., Ferrofluidics APG 933 with $\eta = 0.5$ Pa s) is recommended. With a capillary diameter of 2 mm and a length of 20 cm the necessary pressure drop is $\Delta p \simeq 4.8 \times 10^5$ Pa generating a through-flow rate of about 2 $\text{cm}^3/\text{s}$. The associated Reynolds number is $\simeq 3$, ensuring a laminar Poiseuille flow well below the turbulence threshold. The magnetic dc-field amplitude, $\alpha_0 = 10$, is reached at the moderate field amplitude $H_0 = 21$ kA/m. The modulation frequency $\omega/(2\pi)$ need not be higher than 200 Hz [16].

In summary, the present paper proposes an experiment to detect the onset of particle rotation in a ferrofluid shear flow. A small ac component of the applied magnetic field is greatly amplified (nonlinearly in the excitation) as soon as particle rotation has set in. This setup differs from the magnetovortical resonance experiments [4,5], which are operated at a pure ac magnetic drive. To prevent thermal fluctuations from masking the desired collective effect, the static magnetic field amplitude must be high ($\alpha_0 \simeq 10$). Likewise the condition $\tau_B \Omega \simeq 10$ is necessary for particle rotation to set in. This requires high viscosity ferrofluids with large Brownian relaxation times.

The analysis makes use of the idealized assumption of a monodisperse ferrofluid. Real ferrofluids, however, exhibit a finite size distribution, which implies a distribution of the individual rotation frequency $\omega_P$. This makes the phase synchronization more difficult, but the phase locking technique used here is probably robust enough to operate successfully with a polydisperse ferrofluid.

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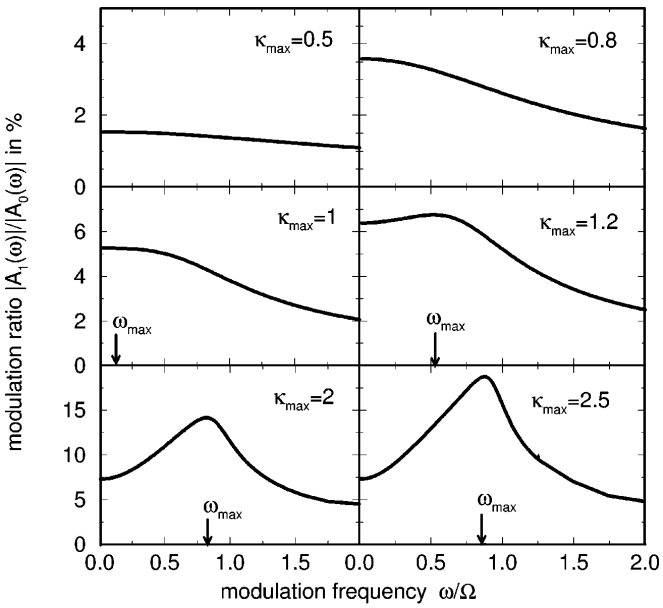


FIG. 5. The same as Fig. 3, but for the circular Poiseuille flow. The quantity $\kappa_{\max}$ is the maximum shear rate in the capillary, i.e., at the inside of the tube wall.

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[14] For consistency with the ferrofluid literature we follow the convention of Ref. [1] where the magnetic moment **m** has the unit of V s m.
[15] The sums have been cut off at $n = 5$ since higher modes do not appreciably contribute.
[16] The estimates for the particle's magnetic moment $m$ and the relaxation time $\tau_B$ rely on recent quantitative measurements of Zeuner *et al.* [8], abbreviated by the superscript $(Z)$. They obtained for a *low viscosity* ferrofluid (EMG 705, Ferrofluidics): $m^{(Z)} \simeq 2 \times 10^{-24}$ V s m, $\eta^{(Z)} = 6 \times 10^{-3}$ N s/m$^2$, and $\tau_B^{(Z)} \simeq 10^{-4}$ s. To transfer these data to the *high viscosity* ferrofluid APG 933 the following relations have been used: $m = m^{(Z)}$ and $\tau_B = (\eta/\eta^{(Z)})\tau_B^{(Z)}$.