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Measuring the rotational viscosity of ferrofluids without shear flow

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The shear free solid-body rotation of a ferrofluid in a magnetic field experiences a damping due to internal *rotational* friction. A device based on the principle of a torsional pendulum is presented. It allows us to perform direct and precise measurements of the field dependence of the genuine rotational viscosity without a disturbing shear flow. Experimental results for a moderately concentrated ferrofluid compare reasonably well with theoretical models for monodisperse ferrofluids of noninteracting rigid dipoles.

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One of the many fascinating properties of ferrofluids [1] is their tunable viscosity, which can be controlled by an external magnetic field. This so called magnetoviscous effect had first been observed by McTague [2]. Qualitatively it can be accounted for [3] by magnetic torques acting upon the suspended ferromagnetic particles: depending on the relative orientation between magnetic field and local vorticity the particle rotation is hindered. The friction at the coated particle surfaces generates an extra dissipation and thus leads to an enhanced effective viscosity. This additional rotational friction is described by a ''rotational viscosity'' η_R. Based on the theory of rotary Brownian motion of non-interacting rigid dipoles, Shliomis [4] derived an analytic expression for the magnetic field dependence of η_R.

We have measured this field induced magneto-viscous damping by means of a torsional pendulum of a high quality factor. While the setup is straightforward and relatively simple it does not seem to have been used before to measure η_R [5,6]. Yet it bypasses in an elegant way problems (c.f. our further discussion below) that arise in earlier experiments [2,7–13]. Therein the effects of shear and rotational friction become mixed up. Moreover, one has to invoke partly untested theoretical assumptions to extract the relatively small (only a few percent) rotational contribution. The main advantage of our setup is to ensure at every instance a solid-body rotation of the ferrofluid without dissipative shear. The rotational viscosity, i.e., the dissipation arising by internal viscous friction between magnetic particles and carrier fluid is then quantified by determining the damping rate of the pendulum.

In our experiment a cylindrical Plexiglas container (inner height 15.1 mm, diameter 16 mm) filled with the ferrofluid APG 933 (Ferrofluidics) [14] is mounted on the carrier platform of a torsional pendulum (Fig. 1). The latter is clamped at a 0.7 mm thick steel wire whose upper and lower ends are fixed in an aluminum frame. The moment of inertia Θ of the oscillating body was determined by measuring the resonance frequencies without payload and with a payload of known moment of inertia. Θ is dominated by the mass of the carrier platform. At zero magnetic field the resonance frequency of the pendulum with the ferrofluid is $\Omega_0/2\pi = 32.7$ Hz and the damping rate is $\Gamma_0 = 0.178$ Hz giving a quality factor of $\Omega_0/\Gamma_0 \simeq 1150$. Capacitive driving is realized at one arm of the carrier with a high voltage ac-source of tunable frequency $f = \omega/2\pi$. A second similar capacitor at the other carrier arm is connected to a lock-in amplifier and records the deflection angle $\varphi(t)$. Its amplitude was always below 1°. A static homogeneous external magnetic field $\mathbf{H}_0 = H_0\mathbf{e}_x$ of variable strength is applied transverse to the rotation axis $\mathbf{e}_z$ of the cylinder. H_0 is measured by a Hall detector.

In order to guarantee solid-body rotation of the fluid the container is partitioned by an array of densely packed plastic

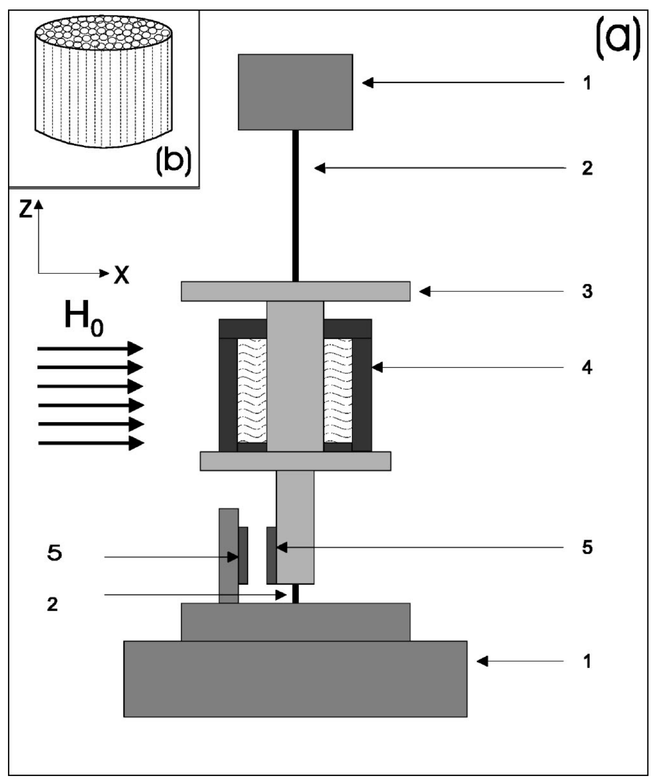

FIG. 1. (a) Sketch of the experimental setup. 1, outer aluminum frame; 2, torsion wire; 3, housing with carrier platform; 4, closed cylindrical Plexiglas container filled with ferrofluid; 5, capacitor for the electrostatic drive. A second capacitor (not visible here) records the deflection amplitude. The inset (b) shows the cylindrical container filled with plastic straws.

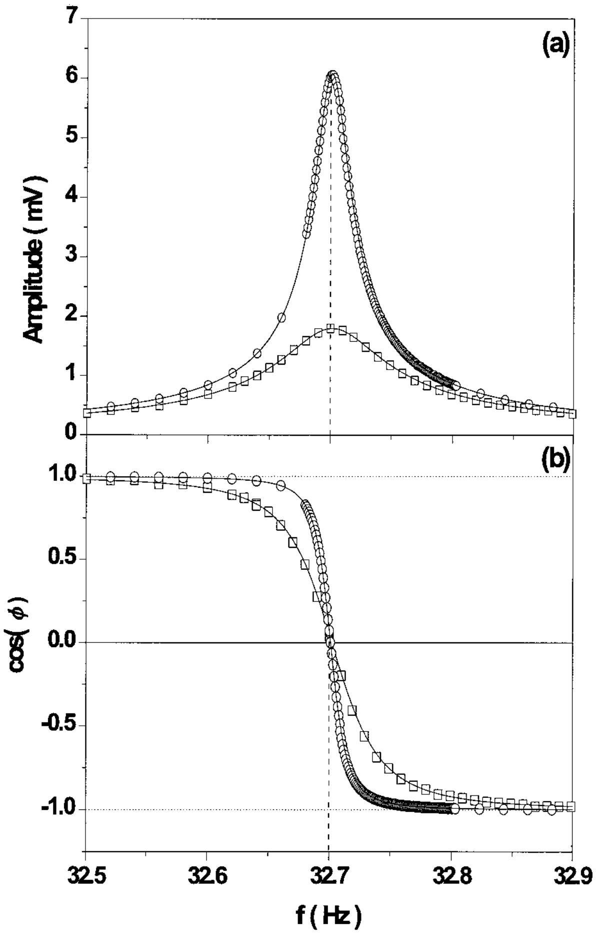

FIG. 2. Experimental raw data for the deflection amplitude (a) (measured in mV at the capacitor detector) and the phase lag (b) relative to the external drive signal. Circles (○) denote data taken for $H_0=0$, squares (□) are for $H_0=14.2\times10^4$ A/m. Solid lines are fits to the harmonically driven linear oscillator. For the damping coefficient we obtain $\Gamma=0.521$ Hz ($\Gamma_P=0.178$ Hz) in the presence (absence) of the magnetic field.

straws (inset of Fig. 1) of inner diameter of 2 mm, being slightly smaller than the viscous skin depth 2.2 mm. We checked the absence of shear flow by replacing the ferrofluid by a solid payload: For $H_0=0$ we observed with solid or fluid payload the same dissipation. However, without straws in the fluid the zero-field damping rises by a factor of 22 due to dissipative viscous shear flow relative to the container walls.

To measure frequency and damping of the pendulum we performed a computer controlled resonance experiment: the frequency band 32 Hz$<f<$35 Hz is scanned and the resulting deflection amplitude as well as the phase lag ϕ relative to the drive are recorded (Fig. 2). Each data point shown in Fig. 2 is the average over 50 single measurements, giving a standard deviation smaller than the point size. After every frequency change the data acquisition is suspended for 60 sec to allow for equilibration to the new drive. To determine damping Γ and eigenfrequency Ω we performed separate fits to amplitude as well as phase lag of the harmonically driven oscillator $\ddot{\varphi}+\Gamma\dot{\varphi}+\Omega^2\varphi=F\cos\omega t$. The agreement of the results from the two different fit-procedures was always better than 1%. Performing the above described frequency scans for fields between 0 and $H_0=2\times10^5$ A/m we obtain Γ,Ω as functions of H_0.

The solid-body rotation, $\dot{\varphi}\mathbf{e}_z$, of the ferrofluid creates a nonequilibrium situation where the magnetization is forced out of the direction of the magnetic field. This generates the torque [4]

$$N=\int_V \mu_0(\mathbf{M}\times\mathbf{H})_z dV \tag{1}$$

acting in the z direction on the ferrofluid volume V. Here $\mathbf{H}$ denotes the macroscopic magnetic field inside the ferrofluid probe. The magnetization $\mathbf{M}(\mathbf{r},t)$ is an average over a large number of ferromagnetic particles, each carrying a magnetic moment $\mathbf{m}$, say. Their statistics is largely governed by the Langevin parameter $\alpha_{local}=\mathbf{m}\cdot\mathbf{H}_{local}/(k_BT)$, where k_B is the Boltzmann constant and T the absolute temperature. The local field $\mathbf{H}_{local}=\mathbf{H}_0-\bar{N}\mathbf{M}+\frac{1}{3}\mathbf{M}$ seen by an individual dipole differs from the local macroscopic field $\mathbf{H}=\mathbf{H}_0-\bar{N}\mathbf{M}$ by the Lorentz field of the cavity which encloses the particle [15,8]. The demagnetizing field $\bar{N}\mathbf{M}$ is geometry dependent; for our cylindrical container of aspect ratio $\simeq1$ we get [16,17] $\bar{N}\simeq0.3485$ so that $\mathbf{H}_{local}\simeq\mathbf{H}_0$ and $\alpha_{local}\simeq\alpha_0$. Furthermore, the tiny oscillation amplitude of our apparatus guarantees that $\mathbf{M}$ deviates only slightly from the equilibrium magnetization $\mathbf{M}_{eq}=M_{eq}\mathbf{H}_0/H_0$ in the absence of rotation, $\dot{\varphi}=0$.

We model the dynamics of the nonequilibrium magnetization $\mathbf{M}-\mathbf{M}_{eq}$ by the relaxation approximation

$$\left[\partial_t+(\mathbf{v}\cdot\nabla)-\frac{\nabla\times\mathbf{v}}{2}\times\right]\mathbf{M}=-\frac{1}{\tau_\parallel}(\mathbf{M}_\parallel-\mathbf{M}_{eq})-\frac{1}{\tau_\perp}\mathbf{M}_\perp\,, \tag{2}$$

which can also be motivated by studying single-particle rotary Brownian motion [18]. Here $\mathbf{v}(\mathbf{r},t)=\dot{\varphi}(\mathbf{e}_z\times\mathbf{r})$ is the velocity field and $\parallel$ and $\perp$ denote the respective directions parallel and perpendicular to $\mathbf{H}_0$. The quantities $\tau_\parallel$, $\tau_\perp$ are the associated field dependent relaxation times. In the limit $H_0=0$ they reduce to the Brownian rotational diffusion time, τ_B. Typically τ_B lies between 10^{-3} and 10^{-6} s depending on the shape of the particle and the viscosity of the carrier liquid. With increasing field strength the relaxation times decrease according to complicated analytical expressions [18]. Ignoring spatial inhomogeneities and higher order contributions $\propto\dot{\varphi}(M_\parallel-M_{eq})$ in Eq. (2) the magnetization M_y in the relevant direction perpendicular to $\mathbf{H}_0$ and the rotation axis is given by

$$\partial_t M_y=\dot{\varphi}M_{eq}-\frac{1}{\tau_\perp}M_y\,. \tag{3}$$

Thus, in the low-frequency limit, $\tau_\perp\Omega_0\ll1$, which prevails in our experiment,

$$M_y(t)=\tau_\perp\dot{\varphi}(t)M_{eq} \tag{4}$$

follows the rotation frequency $\dot{\varphi}(t)$ without phase lag [19]. In this limit the torque (1) is purely dissipative

$$N\simeq N_d=-V\mu_0H_0M_{eq}\tau_\perp\dot{\varphi}, \tag{5}$$

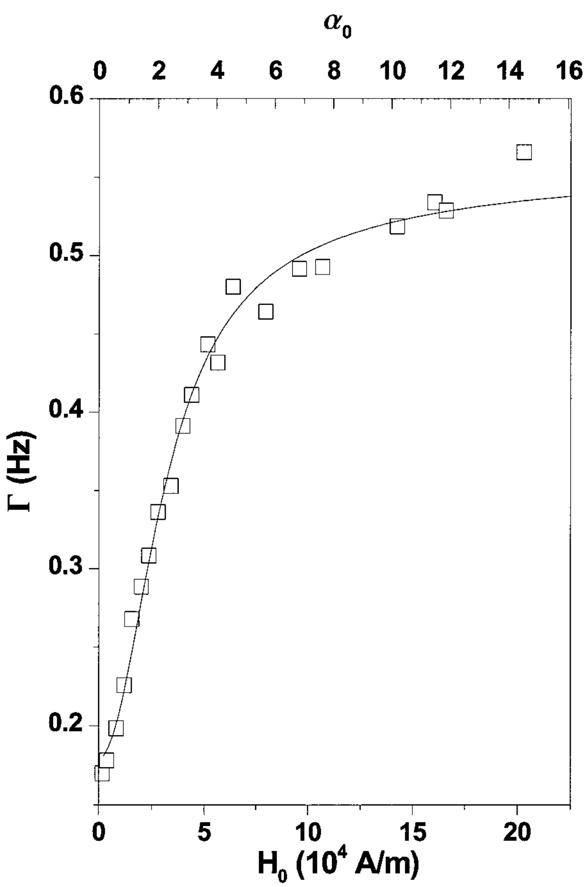

FIG. 3. Symbols denote the damping coefficient $\Gamma=\Gamma_{FF}+\Gamma_P$ measured as a function of the external magnetic field H_0. The solid line is a fit according to Eq. (7).

whereas in general the magneto-viscous torque resulting from Eq. (3) also acquires a reactive contribution. It gives rise to an $O(\Omega_0\tau_\perp\Gamma_{FF})$ increase in the eigenfrequency of the pendulum which, however, cannot be resolved with our present apparatus.

The overall damping rate $\Gamma(H_0)=\Gamma_P+\Gamma_{FF}(H_0)$ measured with our pendulum (see Fig. 3) is due to parasitic effects like air friction and mechanical losses with $\Gamma_P \simeq \Gamma(H_0=0)$ and to the magneto-viscous dissipation in the ferrofluid

$$\Gamma_{FF}=\frac{-N_d}{\dot{\varphi}\Theta}. \tag{6}$$

The latter is given by the total moment of inertia Θ and the oscillation frequency $\dot{\varphi}$ of the pendulum and the dissipative torque N_d of the ferrofluid. With the theoretical prediction (5) one has

$$\Gamma_{FF}=\frac{V\mu_0 H_0 M_{eq}\tau_\perp}{\Theta}=\frac{V\mu_0 M_s k_B T}{m\Theta}\alpha_0\mathcal{L}_0\tau_\perp . \tag{7}$$

The second equality holds for monodisperse particles with $M_{eq}=M_s\mathcal{L}_0$ where M_s denotes saturation and $\mathcal{L}_0=\coth\alpha_0 -1/\alpha_0$ is the Langevin function. Note that Γ_{FF} is expressed as a function of the *external* magnetic field strength α_0. The solid line in Fig. 3 is a fit according to Eq. (7) with $\tau_\perp(\alpha_0)$ taken from Ref. [18]. The magnetic moment m of the particles, the Brownian relaxation time τ_B, and the parasitic damping Γ_P are considered as fit parameters in Fig. 3 leading to values $m=2.91\times10^{-25}$ V s m, $\tau_B=4.68\times10^{-4}$ s, and $\Gamma_P=0.178$ Hz. Assuming spherical particles of hydrodynamic diameter d_h (core plus surfactant coating) with magnetite cores of diameter d_m we get $d_m\simeq9.9$ nm and $d_h \simeq 13.5$ nm. These values are in good agreement with the manufacturer's specification [14]. For the estimate of d_h and d_m we used the relations $m=(\pi/6)\mu_0 M_d d_m^3$ and $\tau_B =\pi\eta d_h^3/(2k_BT)$, where $M_d=4.5\times10^5$ A/m is the bulk magnetization of magnetite and η the shear viscosity of the ferrofluid.

The rotational viscosity $\eta_R(H_0)$ for our solid-body flow $\mathbf{v}=\dot{\varphi}\mathbf{e}_z\times\mathbf{r}$ can be identified by equating the magneto-viscous body force $(\mu_0/2)\nabla\times(\mathbf{M}\times\mathbf{H})$ to $\eta_R\nabla^2\mathbf{v}$ [4]. One thus finds that

$$\eta_R=\frac{1}{4}\frac{-N_d/V}{\dot{\varphi}}=\frac{\Theta}{4V}\Gamma_{FF} \tag{8}$$

is given by the ratio of dissipative torque density N_d/V and rotation rate $\dot{\varphi}$. Thus, the last equality in Eq. (8) provides a simple prescription for measuring $\eta_R(H_0)$ with our apparatus. It is the core result for using the torsional pendulum as a device to determine the field dependent rotational viscosity of a ferrofluid.

If one inserts Eq. (7) one gets (see also [20])

$$\eta_R=\frac{1}{4}\mu_0 H_0 M_{eq}\tau_\perp \simeq \frac{3}{2}\eta\Phi_h\frac{\alpha_0\mathcal{L}_0^2}{\alpha_0-\mathcal{L}_0}. \tag{9}$$

In the last expression we have replaced $\tau_\perp$ by the approximation [21,18] $\tau_\perp=2\tau_B\mathcal{L}_0/(\alpha_0-\mathcal{L}_0)$ that deviates by less than 4% from the exact $\tau_\perp$. Here $\Phi_h=\Phi_m(d_h/d_m)^3$ is the hydrodynamic volume fraction. The α_0 dependence of Eq. (9) deviates by 16% from the relation derived by Shliomis [4], who found $(\alpha_0-\tanh\alpha_0)/(\alpha_0+\tanh\alpha_0)$ instead. This difference comes from his $\tau_\perp^S$ [19] being different from $\tau_\perp$ [21,18].

Thus, the theory of Shliomis provides a reasonable prediction for η_R for our ferrofluid with a magnetic volume fraction of $\Phi_m=3.3\%$. This has to be seen on the background of partly conflicting previous results that we shortly discuss below. The rotational viscosity deduced by McTague from his through-flow experiment agreed *qualitatively* with the single-particle theory despite the fact that his Φ_m was so high that one would expect particle correlations to show up. Weser and Stierstadt [7,8] investigated a ferrofluid with $\Phi_m\simeq6.5\%$ by means of a viscobalance. On taking into account the polydisersity and also parasitic viscosity contributions from excess surfactant molecules they were able to confirm Shliomis' theory by fitting the effective depth of the steric particle coating to be 25% smaller than the length of the surfactant molecules. Subsequently η_R was determined from the onset of Taylor vortex flow in a Couette viscometer. Here, Holderied *et al.* [9] reported satisfactory agreement with the theory for a ferrofluid with $\Phi_m\simeq7.1\%$, while Ambacher *et al.* [10] and Odenbach *et al.* [11] observed for a less concentrated ferrofluid with $\Phi_m=6.7\%$ considerable discrepancies. They argued that the Shliomis approach applies only to very diluted ferrofluids with $\Phi_m < 1\%$ and that the detected deviations are due to particle interactions (e.g., chain formation) not covered by the theory. Gazeau *et al.* [12] perform measurements of η_R in a cylindrical volume rotating in an oscillating magnetic field. However, in their setup a nonvanishing shear flow destroys the solid body rotation. Rosensweig and Johnston [13] use a vessel containing a ferrofluid that is suspended by a fiber. The static deflection of the twisted fiber in a steadily rotating magnetic field is then related to the magnetic body couple. Here the assumption of vanishing fluid motion is questionable with regard to the flow entrainement seen by the authors in the same experimental setup.

For a *quantitative* comparison between theory and experiment the above listed experiments have to cope with (some of) the following problems: (i) How to ensure that the assumed flow profiles (Poiseuille tube flow, circular Couette flow, etc.) remain undeformed when the magnetic field is applied? (ii) The reported flows exhibit shear as well as rotational friction giving rise to a *total* effective viscosity, from which the small rotational contribution has to be separated. (iii) The important problem of identifying the proper magnetic field entering into the theoretical analysis of the experiments (c.f. our discussion of local versus macroscopic field) has largely been ignored; Refs. [7,8,12] are exceptions. (iv) For magnetizations $\mathbf{M}$ that deviate strongly from (local) equilibrium $\mathbf{M}_{eq}$ a reliable constitutive equation as required in the analysis is not available.

With the low-frequency, small-angle, rigid-body rotation of the fluid in our torsional viscometer we avoid or solve these problems and we obtain a direct and sensitive measurement of the rotational viscosity in a novel and straightforward way. By an appropriate choice of the probe geometry it was possible to evaluate the dissipative torque in terms of the (easily accessible) external magnetic field strength. For the moderately concentrated ferrofluid investigated here the results compare reasonably well with theoretical predictions based upon single-particle relaxation behavior and monodisperse size distribution. Measurements on higher concentrated ferrofluids are expected to show when particle correlations become relevant.

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