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Ferrofluid pipe flow in an oscillating magnetic field

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Ferrofluid pipe flow in an oscillating magnetic field along the pipe axis is studied theoretically in a wide range of the flow rate. The field-dependent part of viscosity (it can be positive or negative) reveals significant dependence on the flow vorticity, i.e., ferrofluids exhibit non-Newtonian behavior. This is manifested in an alteration of the velocity profile—it ceases to be parabolic—and deviation of the flow rate from the value prescribed by Poiseuille's formula. The presented model based on the conventional ferrohydrodynamic equations and an assumption of the ferrofluid structure fits well experimental data recently obtained by Schumacher, Sellien, Konke, Cader, and Finlayson ["Experiment and simulation of laminar and turbulent ferrofluid pipe flow in an oscillating magnetic field," Phys. Rev. E **67**, 026308 (2003)].
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I. INTRODUCTION

An increase of ferrofluid viscosity with the application of a steady magnetic field was first revealed by McTague[1] for Poiseuille flow. During the next 35 years the *magnetoviscosity* was an object of intensive theoretical and experimental studies.[2–9] The mechanism of magnetoviscosity has been explained in Refs. 2 and 3. Magnetic field tends to fix particle's own magnetic moments in the field direction. Thereby it impedes free particle rotation in a vortex flow. Indeed, if the field is off, each particle in, e.g., plane Couette flow freely *rolls* along a corresponding shear plane, while in a sufficiently strong magnetic field the particle *slips* along a shear plane without rotation. In the latter case the liquid is forced to flow round the particles. This leads to an extra dissipation of kinetic energy of the fluid what is manifested in an additional—so-called *rotational*—viscosity η_r.

Interestingly, η_r becomes *negative*[10–12] if the field oscillates in time with a high enough frequency ω satisfying the inequality $\omega\tau_B > 1$; here $\tau_B = 3\eta V/(k_B T)$ is the Brownian relaxation time, V is the particle volume, and η is the fluid viscosity. The *negative-viscosity effect* provides some increase of the flow rate compared to the case when the field is off. It is worth noticing that existence of such a negative component of viscosity of course does not contradict thermodynamic laws. Actually, $\eta_r < 0$ simply means that some part of energy of the oscillating magnetic field transforms into kinetic energy of the fluid: high oscillating field *spins* the particles *up* and then accelerates the ferrofluid flow. Meanwhile in a steady magnetic field, η_r is always positive. Indeed, such a field may be created—even though in principle—by means of permanent magnets whose magnetostatic energy certainly cannot be expended on a maintenance of ferrofluid motion.

Predicted ten years ago by Shliomis and Morozov,[10] the negative viscosity effect was soon after observed and investigated by Bacri *et al.*[11] in Poiseuille flow with an ac solenoid wrapped around the pipe. Further, in a similar experiment, Zeuner, Richter, and Rehberg[13] extended the study[11] to a much larger range of magnetic field amplitude and frequency. Both these experiments were limited to slow, laminar flows satisfying the condition $\Omega\tau_B \ll 1$ where Ω is the azimuthal component of the flow vorticity $\mathbf{\Omega} = (\nabla \times \mathbf{v})/2$.

The magnetic torque $\mathbf{m} \times \mathbf{H}, (mH)$, trying to align the particle magnetic moment $\mathbf{m}$ along the field $\mathbf{H}$ is hindered by the random torque, $(k_B T)$, and the regular viscous one, $(6\eta V\Omega)$. Above values in brackets have the torque dimensionality and represent scales of the mentioned torques. Let us introduce nondimensional magnitudes of the magnetic and viscous torques:

$$\xi = mH/(k_B T), \quad 2\Omega\tau_B = 6\eta V\Omega/(k_B T). \tag{1}$$

In the limiting case of low shear rates, $\Omega\tau_B \ll 1$, only thermal agitation impedes orientation of particle magnetic moments along the field. Hence the rotational viscosity $\eta_r(\xi)$ is a function of the Langevin parameter ξ but does not depend on the fluid vorticity Ω. In other words, the ferrofluid behaves in this limit as a Newtonian fluid. Just such a behavior was observed in experiments.[1,11,13]

For a particle of typical size $d \sim 10$ nm settled in a fluid with viscosity $\eta \sim 10^{-2}$ P the Brownian relaxation time proves to be very short: $\tau_B \sim 10^{-7} - 10^{-6}$ s. Therefore, for

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single particles the condition of Newtonian behavior, $\Omega\tau_B \ll 1$, is always satisfied in practice. Very often, however, ferrofluids turn out to be structured, i.e., consisted of aggregates composed out of magnetic grains. The aggregation is provided by magnetic dipole interparticle interaction between the grains. Owing to strong anisotropy of the interaction, its excess over the energy of thermal fluctuations leads to formation of *chains* out of head-to-tail aligned magnetic dipoles. This surprising result of Monte Carlo simulations[14–17] has been recently clarified by Morozov and Shliomis.[18,19] Developing the association theory, they arrived at a natural extension of basic concepts of polymer physics to the case of magnetic dipolar chains and came to the conclusion that a ferrofluid represents an ensemble of flexible chains. Each chain takes conformation of statistical coil whose Brownian relaxation time can reach $\tau_B \sim 10^{-4}–10^{-2}$ s depending on the chain length. For such large magnitudes of τ_B the value of $\Omega\tau_B$ ceases to be small even for quite moderate flow vorticity. Then η_r becomes dependent on the flow rate which just signifies non-Newtonian properties of ferrofluid. The first attempt to explain these properties by the formation of long rodlike clusters (straight chains) in a strong steady magnetic field was performed by Kamiyama and Satoh.[5,6,20]

The dependence of η_r on $\Omega\tau_B$ in an oscillating magnetic field was found for the first time by Gazeau *et al.*[21] in two kinds of experiments: a ferrofluid filled either the inner moving cylinder of the Taylor machine and was rotated *as a whole* with an angular velocity Ω or it filled the gap between the outer and inner cylinders and was subjected to Couette flow with a shear rate Ω. In both these experiments the rotational viscosity turned out to be dependent on Ω. However, in the case of rigid rotation the spectrum of relaxation times remained invariable for any angular velocities, while experimental data for Couette flow indicated that longest relaxation times associated with the most long chains did not exist anymore in the spectrum. This change of dynamical behavior has been interpreted in Ref. 21 as a consequence of shear induced *fracture* of the chains.

Note, that Gazeau *et al.*[21] dealt with a *uniform* field of vorticity: in rigid rotation the vorticity Ω—and hence the rotational viscosity η_r, too—are independent on spatial coordinates. The same situation takes place in such a canonical shear flow as plane Couette flow is. As the velocity profile is linear from one constrained wall to the other, there are $\Omega=\text{const}$ and then $\eta_r=\text{const}$, too. Therefore, with the application of a magnetic field, the ferrofluid viscosity η is simply replaced by its effective value $\eta_{\text{eff}}=\eta+\eta_r$. This leads to an alteration of the *flow rate* (the latter is risen or reduced depending on the sign of η_r) but does not change the velocity profile: it remains to be linear as it is in the absence of magnetic field.

Meanwhile pipe flow, also called Hagen–Poiseuille flow, and similar to it plane Poiseuille flow do not possess such a property. In pipe flow $\Omega \propto r$: the vorticity reaches its largest magnitude Ω_0 on the pipe wall $r=R$ and vanishes at the pipe axis $r=0$. As the result, Ω-dependent η_r turns out to be dependent on r, too. So, if $\Omega_0\tau_B$ is sufficiently large, $\eta_r(0)$ differs significantly from $\eta_r(R)$ and then the flow pattern ceases to be parabolic.

Experimental results recently obtained by Schumacher *et al.*[22] give us a good possibility to test our predictions concerning the shear dependence of the ferrofluid viscosity and the pipe flow rate in a linearly polarized magnetic field oscillating along the axis of the pipe. Schumacher *et al.*[22] conducted an experiment similar to Bacri *et al.*[11] and Zeuner, Richter, and Rehberg.[13] An important distinction: the experiments[11,13] were limited to slow laminar flows, $\Omega_0\tau_B \ll 1$, while in Ref. 22 it was studied a wide range of laminar and turbulent flow rates. On the border between these regimes, the value $\Omega_0\tau_B$ reached unity and so no wonder that the reduced rotational viscosity $\eta_r(\xi,\Omega_0\tau_B)/\eta$ (referred to in Ref. 22 as the fractional pressure drop) displayed a strong dependence on the flow rate. To describe the dependence theoretically and numerically, authors of Ref. 22 made some questionable simplifications (see Sec. IV for details) and employed the magnetization relaxation time as a fitting parameter choosing its value for every flow rate and every frequency of magnetic field oscillations.

The paper is organized as follows. In Sec. II we introduce a complete set of ferrohydrodynamic equations for incompressible ferrofluids. Two formulations of magnetization equation are employed and compared below: one of the two was derived microscopically from the Fokker–Planck equation, while another one represents a generalization of the Debye relaxation equation. In Sec. III we study effects of no small flow vorticities upon the ferrofluid pipe flow taking as an example the material parameters of a ferrofluid used earlier in the experiments of Bacri *et al.*[11] To describe well experimental results obtained by Schumacher *et al.*,[22] we perform in Sec. IV numerical simulations of the full set of ferrohydrodynamic equations (without any simplifications) under an assumption of existence of chainlike aggregates. Finally, in Sec. V, we draw our conclusions.

II. BASIC EQUATIONS

We use here a conventional set of ferrohydrodynamic equations for incompressible ferrofluids. It consists of the equation of fluid motion (2) derived by Shliomis,[2,3] the magnetization equation (3) derived by Martsenyuk, Raikher, and Shliomis[23] from the Fokker–Planck equation, and the Maxwell magnetostatic equations (4):

$$\rho\frac{d\mathbf{v}}{dt} = -\nabla p + \eta\nabla^2\mathbf{v} + (\mathbf{M}\cdot\nabla)\mathbf{H} + \frac{1}{2}\nabla\times(\mathbf{M}\times\mathbf{H}), \tag{2}$$

$$\frac{d\mathbf{M}}{dt} = \boldsymbol{\Omega}\times\mathbf{M} - \frac{1}{\tau_B}\left[\mathbf{M} - \frac{3L(\zeta)}{\zeta}\chi\mathbf{H}\right] - \frac{3\chi}{2\tau_B M^2}\times\left[1-\frac{3L(\zeta)}{\zeta}\right]\mathbf{M}\times(\mathbf{M}\times\mathbf{H}), \tag{3}$$

$$\nabla\cdot\mathbf{B}=0, \quad \nabla\times\mathbf{H}=0 \quad (\mathbf{B}=\mathbf{H}+4\pi\mathbf{M}). \tag{4}$$

These equations were discussed in detail in Refs. 4 and 24–27. In the equations $d/dt=\partial/\partial t+\mathbf{v}\cdot\nabla$ is the total time derivative, ρ is the ferrofluid density, p is the pressure, η is the shear viscosity, χ is the initial magnetic susceptibility, and $\mathbf{M}$

is the ferrofluid magnetization. In a true equilibrium (possible only in a quiescent fluid in a steady magnetic field $\mathbf{H}$) the magnetization is described well by the Langevin formula

$$\mathbf{M}_{\mathrm{eq}} = M_s L(\xi)\frac{\boldsymbol{\xi}}{\xi}, \quad \boldsymbol{\xi} = \frac{3\chi\mathbf{H}}{M_s}, \quad L(\xi) = \coth \xi - \frac{1}{\xi}, \tag{5}$$

where M_s is the saturation magnetization. For an ideal gas of identical paramagnetic molecules of the number density n there are $M_s=nm$ and $\chi=nm^2/(3k_BT)$, thus the Langevin parameter takes its well-known form $\xi=mH/(k_BT)$. In a nonequilibrium state $\mathbf{M}$ and $\mathbf{H}$ are independent from each other: a magnetization may exist (though not for very long) even in the absence of $\mathbf{H}$. Nevertheless, one can consider $\mathbf{M}$ as an equilibrium at any moment in a certain (so-called *effective*) specially prepared magnetic field $\boldsymbol{\zeta}$. The instant nonequilibrium magnetization is expressed through the dimensionless effective field $\boldsymbol{\zeta}$ by the equilibrium formula (5):

$$\mathbf{M} = M_s L(\zeta)\frac{\boldsymbol{\zeta}}{\zeta}. \tag{6}$$

In Eq. (3) we used also the notation $M=M_sL(\zeta)$. In a true equilibrium $\boldsymbol{\zeta}$ coincides sure with $\boldsymbol{\xi}$ whereupon Eq. (6) is transformed into Eq. (5). Equations (3) and (6) determine the dependence $\mathbf{M}(t,\mathbf{r};\mathbf{H},\boldsymbol{\Omega})$ in an implicit form, where the effective field ζ is the parameter. Apart from Eq. (3) derived microscopically,[23] there are three phenomenological magnetization equations proposed by Shliomis[2,8] and Felderhof and Kroh.[28] Each of these three [cited below Eq. (18) is one of them] is much simpler than Eq. (3). However, their application is limited to small amplitudes of the oscillating magnetic field and weak flow vorticities, whereas Eq. (3) delivers a quite accurate description in a wide range of flow rates, magnetic field strengths and oscillation frequencies. Actually, this equation provides a very good approximation to the results of exact solution of the Fokker–Planck equation[29] and the computer simulation of the Brownian dynamics of magnetic grains.[30,31]

Let us consider Poiseuille flow of a ferrofluid in an oscillating magnetic field applied along the pipe—the situation studied in experiments of Bacri *et al.*,[11] Zeuner *et al.*,[13] and Schumacher *et al.*[22] In cylindrical coordinates with the z axis along the axis of the pipe, this one-dimensional flow,

$$\mathbf{v} = \{0,0,v(r,t)\}, \quad \boldsymbol{\Omega} = \{0,\Omega(r,t),0\}, \quad \Omega = -\frac{1}{2}\,\partial v/\partial r,$$

is caused by an imposed pressure gradient $\partial p/\partial z=-\Delta p/l$ where Δp is the pressure drop over the tube length l. In contrast to Felderhof[32] we do not assume the pressure gradient to be small.

The term $\boldsymbol{\Omega}\times\mathbf{M}$ in Eq. (3) provides an influence of ferrofluid flow on its magnetization. In fact, interaction of the azimuthal component of vorticity Ω with the axial component of magnetization M_z (the latter is formed directly by the applied field $H_z=H_0\cos \omega t$) gives rise to an off-axis (radial) component of the magnetization $M_r(r,t)$. But the presence of M_r dependent on r entails immediately—due to the Maxwell equation $\boldsymbol{\nabla}\cdot(\mathbf{H}+4\pi\mathbf{M})=0$—an appearance of the counteracting magnetic field component $H_r=-4\pi M_r$. Felderhof[32] truly noticed that the *demagnetizing field* was omitted in Refs. 10 and 11 and on the spot he omitted in his turn the radial component of the *magnetic force* density, $(\mathbf{M}\cdot\boldsymbol{\nabla})H_r =-2\pi(\partial M_r^2/\partial r)$, entering into the equation of fluid motion. In an incompressible ferrofluid this force is automatically equilibrated by the radial component of the pressure gradient, $\partial p/\partial r=(\mathbf{M}\cdot\boldsymbol{\nabla})H_r$, and then it does not play any role. But Felderhof[32] considered a *compressible* fluid. In this case the oscillating magnetic force should excite radially divergent *sound waves* with the fluid velocity $v_r(r,t)$. One ought to study this effect although it is probably weak and can hardly be detected.

Thus, vectors $\mathbf{M}$, $\mathbf{H}$, and $\boldsymbol{\nabla}p$ have the structure

$$\mathbf{M} = \{M_r(r,t),0,M_z(r,t)\},$$

$$\mathbf{H} = \{-4\pi M_r(r,t),0,H_0\cos \omega t\},$$

$$\boldsymbol{\nabla}p = \{-2\pi(\partial M_r^2/\partial r),0,-\Delta p/l\}. \tag{7}$$

For small flow vorticities, $|\Omega\tau_B|\ll 1$, the magnetization equation (3) can be solved in a perturbative way:[11,32]

$$M_r = \Omega\tau_B M_1(t) + (\Omega\tau_B)^3 M_3(r,t) + \cdots,$$

$$M_z = M_0(t) + (\Omega\tau_B)^2 M_2(r,t) + \cdots. \tag{8}$$

In the first-order approximation in $\Omega\tau_B$ the flow pattern remains parabolic and the flow rate

$$Q = 2\pi\int_0^R \langle v\rangle r dr \tag{9}$$

(here $\langle . \rangle$ means the time averaging over the period of magnetic field oscillations) obeys the Poiseuille formula

$$Q = \frac{\pi R^4}{8\eta_{\mathrm{eff}}}\left(\frac{\Delta p}{l}\right)$$

with some effective viscosity η_{eff} dependent on the dimensionless amplitude $\xi_0=3\chi H_0/M_s$ and frequency $\omega\tau_B$ of the external magnetic field. Consider the origin of η_{eff} a little more detailed. Choosing for the units of length R, time τ_B, and velocity $(\Delta p/l)R^2/(4\eta)$, we obtained from Eqs. (2) and (3) three dimensionless equations coupling the axial velocity $v(r,t)$ with the radial ζ_r and axial ζ_z components of the *effective field* $\boldsymbol{\zeta}$:

$$\alpha^{-1}\frac{\partial v}{\partial t} = 4 + \frac{1}{r}\frac{\partial}{\partial r}\left[r\frac{\partial v}{\partial r} - r\beta(\xi_0\cos \gamma t + 12\pi\chi L^*\zeta_z)L^*\zeta_r\right], \tag{10}$$

$$\frac{\partial}{\partial t}(L^*\zeta_r) = -\frac{\sigma}{2}\frac{\partial v}{\partial r}L^*\zeta_z - (1+12\pi\chi L^*)L^*\zeta_r - (1-3L^*)$$
$$\times(\xi_0\cos \gamma t + 12\pi\chi L^*\zeta_z)\frac{\zeta_r\zeta_z}{2\zeta^2}, \tag{11}$$

$$\frac{\partial}{\partial t}(L^*\zeta_z) = \frac{\sigma}{2}\frac{\partial v}{\partial r}L^*\zeta_r - L^*(\zeta_z - \xi_0\cos\gamma t) + (1-3L^*)$$
$$\times(\xi_0\cos\gamma t + 12\pi\chi L^*\zeta_z)\frac{\zeta_r^2}{2\zeta^2}. \qquad (12)$$

Here $L^* \equiv L(\zeta)/\zeta$ and the magnetization components M_r and M_z are expressed through ζ_r and ζ_z by the relations

$$M_r = M_s L(\zeta)\frac{\zeta_r}{\zeta}, \quad M_z = M_s L(\zeta)\frac{\zeta_z}{\zeta}, \quad \zeta = \sqrt{\zeta_r^2 + \zeta_z^2}.$$

Apart from the dimensionless strength of magnetic field ξ_0 and the initial magnetic susceptibility χ, Eqs. (10)–(12) contain four more nondimensional parameters:

$$\alpha = \frac{\eta\tau_B}{\rho R^2}, \quad \beta = \frac{2M_s^2}{3\chi R}\left(\frac{l}{\Delta p}\right),$$

$$\gamma = \omega\tau_B, \quad \sigma = \Omega_0\tau_B. \qquad (13)$$

The first represents the ratio of the magnetization time scale τ_B to the hydrodynamic one $\tau_h = \rho R^2/\eta$. The second defines the influence of a magnetic field on the ferrofluid flow. At $\beta = 0$ the steady solution of Eq. (10) satisfying the no slip boundary condition at the pipe radius, $v(1,t) = 0$, yields the common velocity of Poiseuille flow which in our notation reads $v(r) = 1 - r^2$. Finally, the last parameter, σ (sometimes it is called the *Péclet number*), is defined through the flow vorticity at the pipe wall $r = R$ in the absence of magnetic field,

$$\Omega_0 = \frac{R}{4\eta}\left(\frac{\Delta p}{l}\right), \qquad (14)$$

and measures the relative importance of viscous and Brownian torques on the particle.

For a tube of radius $R \sim 1$ mm the characteristic hydrodynamic time τ_h is not smaller than 10^{-1} s so the inequality $\alpha = \tau_B/\tau_h \ll 1$ is usually satisfied. It means that the velocity alters much slowly than the magnetization, which allows Eq. (10) to be uncoupled from Eqs. (11) and (12). Indeed, since $v(r,t)$ slowly varies for the time of the order of τ_B, the derivative $\partial v/\partial r$ in Eqs. (11) and (12) can be considered as independent on time (frozen). Solutions of these equations, $\zeta_r(r,t)$ and $\zeta_z(r,t)$, one should substitute into Eq. (10) and afterwards average this equation over the fast (magnetization) time.

If $\alpha \ll 1$ and besides σ is also small, one can restrict oneself to the *linear approximation* in σ. Then Eqs. (11) and (12) yield [cf. Eq. (8)]

$$\zeta_r \sim \sigma(\partial v/\partial r)\xi_0, \quad \zeta_z \sim \xi_0. \qquad (15)$$

Substituting the components into Eq. (10) and averaging over the fast time the equation of fluid motion takes the form

$$\frac{1 + \beta\sigma f(\xi_0,\gamma)}{r}\frac{\partial}{\partial r}\left(r\frac{\partial v}{\partial r}\right) = -4. \qquad (16)$$

Thus, in this approximation all interplay between hydrodynamic and magnetic phenomena reduces to a renormalization of the ferrofluid viscosity; according to Eq. (16) its dimensional value is

$$\eta_{\mathrm{eff}} = \eta[1 + \beta\sigma f(\xi_0,\gamma)]. \qquad (17)$$

As seen from definitions (13) and (14), the product $\beta\sigma$ does not contain the pressure drop Δp and hence is independent on the flow rate but represents the material constant: $\beta\sigma = M_s^2\tau_B/(6\eta\chi)$. For an ideal case of noninteracting magnetic grains this constant reduces to $3\phi/2$ where $\phi = nV$ stands for the volume fraction of suspended magnetic grains. Thus Eq. (17) for η_{eff} turns out to be similar to the Einstein formula for viscosity of suspensions. The function $f(\xi_0,\gamma)$ in this equation—it is positive or negative depending on values of its arguments—has been calculated in Refs. 11, 13, and 32.

Having in mind to study the ferrofluid flow at finite (i.e., not necessarily small) vorticities, we solved the set of Eqs. (10)–(12) numerically. For comparison we also used the phenomenological magnetization equation obtained by Shliomis[2] as a generalization of the Debye relaxation equation for the case of spinning magnetic grains:

$$\frac{d\mathbf{M}}{dt} = \mathbf{\Omega} \times \mathbf{M} - \frac{\mathbf{M} - \mathbf{M}_{\mathrm{eq}}}{\tau_B} - \frac{3\chi\mathbf{M} \times (\mathbf{M} \times \mathbf{H})}{2\tau_B M_s^2}, \qquad (18)$$

where $\mathbf{M}_{\mathrm{eq}}$ is defined in Eq. (5).

In numerical calculations the spatial derivatives were represented by the central finite differences with uniform mesh $\delta r = 1/200$ (the accuracy of the resulting solution was verified taking $\delta r = 1/400$). The system of first-order ordinary differential equations was integrated in time using the NAG Fortran Library routine with backward differentiation formulas code. This approach allowed us to keep the relative error of the solution less than 10^{-4} for a reasonable computation time. Starting with some initial conditions, the integration in time was performed until the solution converges to the periodic one. Then the flow rate (9) was calculated.

III. RESULTS AND DISCUSSION

Our calculations correspond to the experimental setup and conditions taking place at the first observation of the negative-viscosity effect by Bacri *et al.*[11] The ferrofluid of the density $\rho = 2.2$ g/cm^3 and the shear viscosity $\eta = 0.77$ P represented a water-based colloidal dispersion of cobalt ferrite with the volume fraction of magnetic grains $\phi = 0.2$, the relaxation time $\tau_B = 1.6$ ms, and the initial magnetic susceptibility $\chi = 0.22$ (i.e., the magnetic permeability was $\mu = 1 + 4\pi\chi \simeq 3.8$). The Poiseuille-like flow in a horizontal capillary tube ($R = 0.5$ mm) placed inside a solenoid was maintained by the gradient of pressure originated from the difference of the fluid levels between the inlet and outlet of the tube. The flow rate Q was measured as a function of the pressure drop Δp and the amplitude ξ_0 and frequency $\omega\tau_B$ of the magnetic field.

Results of our calculations are presented in Figs. 1–3. In Fig. 1 the contour plots for the relative change of the flow rate $\Delta Q/Q_0$ is shown in the $(\omega\tau_B, \xi_0)$ plane for different values of $\Omega_0\tau_B$. Here $\Delta Q = Q_H - Q_0$ is the difference between the flow rates in the presence (Q_H) and the absence (Q_0) of magnetic field while $\Omega_0\tau_B$ may be considered as the dimen-

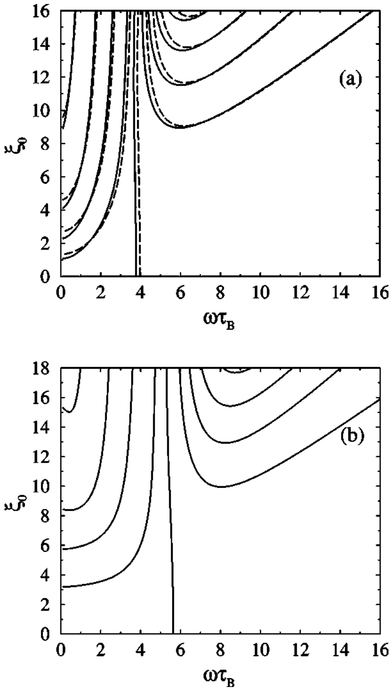

FIG. 1. Contour plot of the relative change of the flow rate $\Delta Q/Q_0$ in the $(\omega\tau_B, \xi_0)$ plane for values, successively from left to right, −0.168, −0.12, −0.072, −0.024, 0, +0.024, +0.04, +0.056, +0.072. The flow vorticity $\Omega_0\tau_B=0.01$ (solid lines), 1.5 (dashed lines) (a) and $\Omega_0\tau_B=5$ (b).

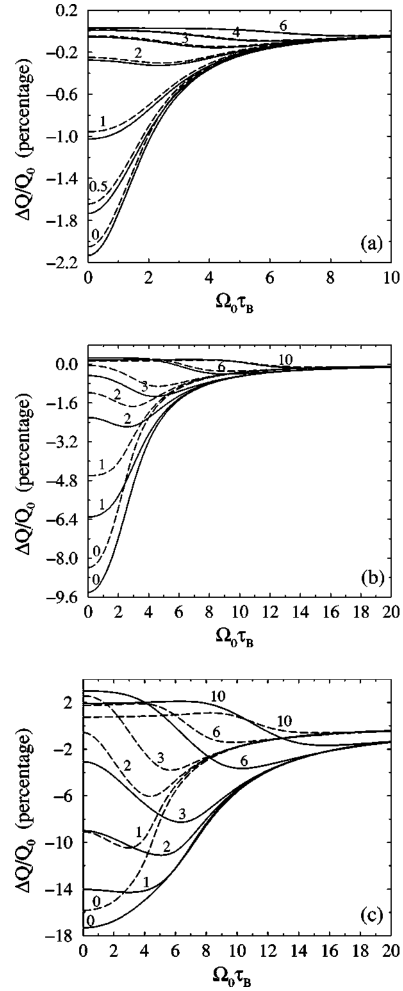

FIG. 2. The relative change of the flow rate $\Delta Q/Q_0$ vs flow vorticity $\Omega_0\tau_B$: (a) $\xi_0=1$, (b) $\xi_0=3$, and (c) $\xi_0=10$. Solid lines—magnetization equation (3); dashed lines—magnetization equation (18). The values of the magnetic field frequency $\omega\tau_B$ are given on top of each curve.

sionless pressure drop [see definitions (13) and (14)]. The main calculations were performed with the magnetization equation (3), or, precisely, with its nondimensional components (11) and (12). Positive values of ΔQ are caused by the negative-viscosity effect, while $\Delta Q<0$ corresponds to $\eta_r>0$. For small flow vorticity ($\Omega_0\tau_B=0.01$) the contour lines $\Delta Q/Q_0=\text{const}$ obtained from expansion (8) in the linear accuracy in $\Omega_0\tau_B$ practically coincide with the results of full numerical simulations presented in Fig. 1(a). As seen from the figure, the pattern of isolines weakly depends on the pressure drop until $\Omega_0\tau_B\leqslant 1.5$ but its alteration is quite visible at $\Omega_0\tau_B=5$—compare Figs. 1(a) and 1(b).

The dependence of the reduced flow rate $\Delta Q/Q_0$ on the dimensionless pressure drop $\Omega_0\tau_B$ calculated for different values of the magnetic field strength ξ_0 and the field frequency $\omega\tau_B$ is shown in Fig. 2. The calculations were performed with the magnetization equations (3) derived microscopically. For comparison, the results obtained by the use of the phenomenological magnetization equations (18) are also presented. One can see that Eq. (18) gives adequate description in weak magnetic fields, $\xi_0<1$. As the field strength increases, the difference between the results yielding by Eqs. (3) and (18) increases too.

Figure 2 clearly demonstrates a nonlinear and nonmonotonic dependence of $\Delta Q/Q_0$ on $\Omega_0\tau_B$ where the vorticity Ω_0 is defined in (14). Presented dependence reflects non-Newtonian properties acquiring by ferrofluids in magnetic fields and may be explained in terms of the shear-dependent viscosity. To give the qualitative explanation we consider the limit of weak applied field, $\xi_0\ll 1$. In this case one can neglect the nonlinear relaxation terms [containing the double vector product $\mathbf{M}\times(\mathbf{M}\times\mathbf{H})$] in Eqs. (3) and (18) and then both the linearized magnetization equations yield the same relationship for the rotational viscosity averaged over the period of the field variation:

$$\eta_r = \eta_r^0 \frac{\mu^2 + (\Omega^2-\omega^2)\tau_B^2}{[\mu + (\Omega^2-\omega^2)\tau_B^2]^2 + (\mu+1)^2\omega^2\tau_B^2},$$

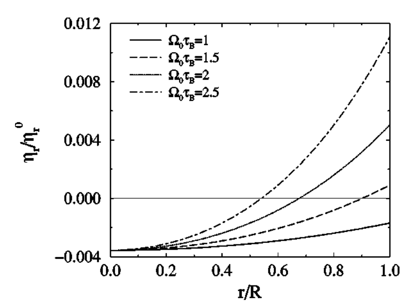

FIG. 3. The dependence η_r/η_r^0 on r for $\omega\tau_B=4$ and different values of $\Omega_0\tau_B$ from Eq. (19).

$$\eta_r^0 \equiv \frac{\chi\tau_B H_0^2}{8} = \frac{M_s^2\tau_B}{72\chi}\xi_0^2 \Rightarrow \frac{1}{8}\eta\phi\xi_0^2. \tag{19}$$

Here the last value, $\eta\phi\xi_0^2/8$, is coming in for noninteracting magnetic grains. The vorticity $\Omega(r)$ of an axial-symmetric flow equals zero at $r=0$ and reaches a maximum at $r=R$; for Poiseuille flow we have in our units $\Omega\tau_B=\sigma r$. In the Newtonian limit, $\sigma \ll 1$, one can omit shear-dependent terms $\Omega^2\tau_B^2$ in (19) whereupon this relationship reduces to the Felderhof's expression,[32]

$$\eta_r\big|_{\Omega\to 0} = \eta_r^0\frac{\mu^2-\omega^2\tau_B^2}{(\mu^2+\omega^2\tau_B^2)(1+\omega^2\tau_B^2)}. \tag{20}$$

So, in the case of weak vorticity, η_r is negative for a high frequency magnetic field, $\omega\tau_B>\mu$, and positive for a stationary or low frequency oscillating field, $\omega\tau_B<\mu$. The negative value of η_r provides some decrease of the total (effective) viscosity which leads to an increase of the flow rate, $\Delta Q>0$. Actually, Fig. 2 has been drawn for $\mu=3.8$ and hence initial (at $\Omega_0\tau_B\equiv\sigma\ll 1$) values of $\Delta Q/Q_0$ are positive for $\omega\tau_B \geqslant 4$ and negative for $\omega\tau_B\leqslant 3$.

In the case of finite values of σ one should employ Eq. (19). It limits the frequency range of existence of the negative viscosity by the condition $\omega^2\tau_B^2>\mu^2+\sigma^2 r^2$. Hence, if the field frequency satisfies inequalities $\mu<\omega\tau_B<\sqrt{\mu^2+\sigma^2}$, the rotational viscosity changes its sign at a certain $r_*=\sigma^{-1}\sqrt{\omega^2\tau_B^2-\mu^2}<1$. Figure 3 demonstrates the dependence $\eta_r(r)$ for some values of σ.

As seen, η_r is negative through the tube cross section for sufficiently small σ while for strong vorticities it is negative only near the tube axis. This r-dependence of viscosity results in a deviation of the velocity profile from parabolic one. The total velocity of the fluid may be written as $v=1-r^2+v_1$. The addition $v_1(r,t)$ oscillates with the double frequency of the field variation. This function has been found by numerical solution of Eqs. (10)–(12) and presented in Fig. 4 in different moments together with its mean value $\langle v_1\rangle$ averaged over the period of oscillations.

Depending on σ, the function $\langle v_1\rangle$ is positive [Fig. 4(a)], negative [Fig. 4(c)], or has a node [Fig. 4(b)]. Correspondingly, the field induced additional flux $\Delta Q/Q_0$ shown in Fig. 2 turns out to be positive or negative. This flux tends to zero in the limit $\Omega_0\tau_B \gg \xi_0$, i.e., when viscous torques acting upon magnetic particles predominate over magnetic torques, so the latter cannot create an orientational arrangement of the particle magnetic moments.

IV. THE CASE OF SHEAR DEPENDENT PARAMETERS

In recent work,[22] Schumacher *et al.* studied the ferrofluid pipe flow in an oscillating magnetic field at nonsmall values of $\Omega_0\tau_B$. The water based ferrofluid EMG-206 (Ferrotec) with the density $\rho=1.187$ g/cm^3 and viscosity $\eta=3.855\times 10^{-2}$ P was pumped through the capillary tube of radius $R=1.5$ mm. The reduced difference $(\Delta p_H-\Delta p_0)/\Delta p_0$ of the pressure drops with and without magnetic field was measured for the fixed flow rates Q at different values of magnetic field strength and frequency. For $Q\leqslant 800$ ml/min the ferrofluid flow was found to be laminar and the fractional pressure drop was decreasing with increasing of Q. For $Q>800$ ml/min the flow was turbulent and the fractional pressure drop remained nearly constant.

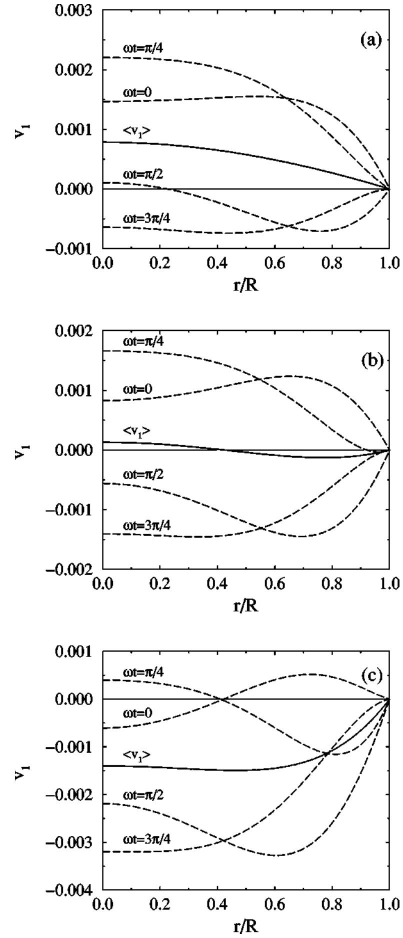

FIG. 4. Profiles of the velocity $v_1(r,t)$ at different moments of time and $\langle v_1(r)\rangle$ for $\xi_0=3$, $\omega\tau_B=4$, and $\Omega_0\tau_B=0.5$ (a), $\Omega_0\tau_B=1.8$ (b), and $\Omega_0\tau_B=3$ (c).

Schumacher *et al.*[22] performed numerical simulations of the ferrofluid flow on the base of a simplified version of the magnetization equation (18). Namely, instead of expression (5) for the instant equilibrium magnetization they used an approximation of the "effective susceptibility," $\mathbf{M}_{eq}=\chi_0\mathbf{H}$, where χ_0 is an unknown function of H. The authors of Ref. 22 assumed that χ_0 in the case with flow and an oscillating H is the same *constant* as in a nonflowing fluid and a *steady* magnetic field. Under the assumption, there is no trouble in doing time averaging of the magnetic torque $\mathbf{M}\times\mathbf{H}$ over the period of magnetic field oscillations and obtaining an analytic expression for the torque and steady equations for the fluid velocity and the magnetic particle's spin. This approach is valid, however, only in weak magnetic fields, i.e., when $\xi_0\ll 1$. Then the Langevin function of $\xi=\xi_0\cos\omega t$ reduces to $\xi/3$ and hence the effective susceptibility $\chi_0=3\chi L(\xi)/\xi$ reduces to the constant initial susceptibility χ. But this is not the case of Schumacher *et al.*:[22] they applied pretty strong magnetic fields—up to $\xi_0=4$—so that their χ_0 should certainly *depend on time*.

The Brownian relaxation time τ_B was treated in Ref. 22 as a fitting parameter whose magnitude decreases with increase of both the flow rate and the field frequency. Such a dependence of τ_B on Q looks quite reasonable. Actually, as we pointed out in Sec. I, only big enough aggregates—most probably chainlike ones—bring a noticeable contribution in non-Newtonian features of ferrofluids. Hence, if such aggregates really exist, the pipe flow with a high enough shear rate should induce destruction of the chains leading thereby to some decrease of their Brownian relaxation time.

Give a definition of the chain. Two neighboring magnetic grains are reputed to be bonded if their dipolar potential for the head-to-tail dipole configuration at the distance of closest approach exceeds the energy of thermal fluctuations, i.e., when $2m^2/d^3 \geqslant k_BT$. In other words, the dimensionless coupling parameter $\lambda = m^2/(d^3 k_B T)$ should be sufficiently large to form a long enough chain. The most informative quantity of chain conformation is the "end-to-end" vector $\hat{\mathbf{R}} = \Sigma_{i=1}^{N-1}\hat{\mathbf{r}}_i$ connecting centers of first $(i=1)$ and last $(i=N)$ particles in a chain. In the case $\lambda \gg 1$ there is $\langle \hat{R}^2 \rangle \simeq N\lambda d^2$—see Refs. 18 and 19. The magnetic moment of a chain $\hat{\mathbf{m}}$ is determined analogously to $\hat{\mathbf{R}}$, i.e., $\hat{\mathbf{m}} = \Sigma_{i=1}^{N}\mathbf{m}_i$ where $\mathbf{m}_i$ is magnetic moment of a grain of the number i. Hence $\langle \hat{m}^2 \rangle \simeq N\lambda m^2$ so that $\chi \propto \lambda$, $M_s \propto \lambda^{1/2}$, and $\tau_B \sim \hat{R}^3 \propto \lambda^{3/2}$.

Make allowance for the shear dependence of the fluid parameters in the frame of the following simple model. Let us take into account that the regular viscous torques induced by the shear flow lead—side by side with random (thermal) torques—to some reduction of the coupling constant λ. Therefore one should replace k_BT in the denominator of λ by the sum $k_BT + 6\eta V\Omega = k_BT(1+2\Omega\tau_B)$ [see Eq. (1)]. Thus, one ought to make the substitution

$$\lambda \Rightarrow \frac{\lambda}{1+2\Omega\tau_B}. \tag{21}$$

Then, in accordance with the pointed above scaling, the following dependence of the fluid parameters on the shear rate should be assumed:

$$\tau_B = \frac{\tau_B^0}{(1+b\ \Omega_0\tau_B^0)^{3/2}},$$

$$\chi = \frac{\chi^0}{1+b\ \Omega_0\tau_B^0},$$

$$M_s = \frac{M_s^0}{\sqrt{1+b\ \Omega_0\tau_B^0}}. \tag{22}$$

The superscript "0" means here the limit $\Omega_0 \to 0$ and the fitting parameter b is expected to be of the order of unity.

We have performed numerical calculations of the fractional pressure drop $\Delta p_H/\Delta p_0 - 1$ employing the microscopically derived magnetization equation (3). To solve it for given experimental values of flow rates, we have used the expressions (22) and undertaken iterations of the pressure drop by the secant method. Taking from Ref. 22 the whole set of experimental data corresponding to the laminar flow regime at the three frequencies of magnetic field, $f=\omega/2\pi = 60$, 400, and 1000 Hz (*32 points in total*), we have found a set of *four parameters* entering into relationships (22) and providing the best fit to *all* of the experimental data:

$$\tau_B^0 = 252\ \mu\text{s}, \quad \chi^0 = 0.0034, \quad M_s^0 = 0.924\ \text{G}, \quad b = 1.61.$$

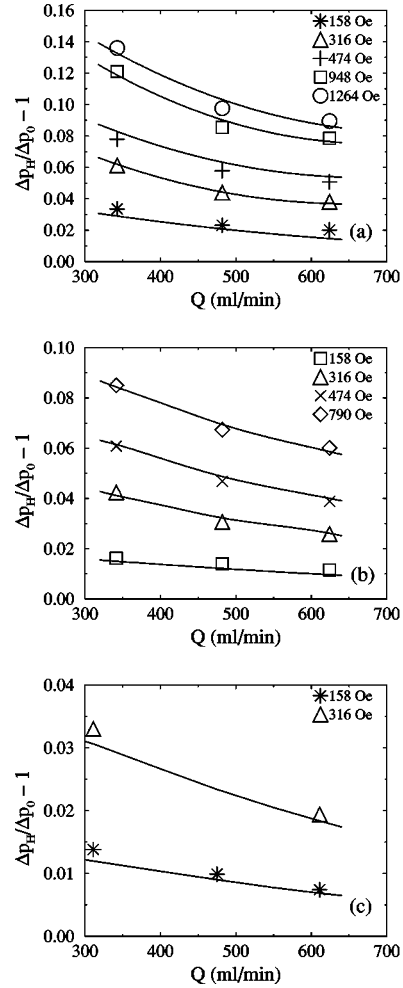

FIG. 5. The fractional pressure drop as a function of flow rate and magnetic field: (a) $f=60$ Hz; (b) $f=400$ Hz; (c) $f=1000$ Hz. Solid lines are the results of simulations, points are the experimental data from Ref. 22.

The total saturation magnetization of the used in Ref. 22 ferrofluid EMG-206 was $M_{s,FF} = 11.94$ G. Thus, only a small amount of magnetic grains, $M_s^0/M_{s,FF} \approx 8\%$, was combined with each other to form the chains. Those were the biggest grains: their Langevin parameter $\xi^0 \equiv 3\chi^0 H/M_s^0$ reached unity at already $H \approx 91$ Oe that corresponds to the diameter of magnetite particle $d=12$ nm whereas the mean diameter of particles in EMG-206 is about 9 nm. The results of our calculations together with experimental data of Schumacher *et al.*[22] are presented in Fig. 5. As seen, the fit is good enough for the field frequencies $f=60$ and 400 Hz. For $f=1000$ Hz and low flow rates it is a little bit worse since the values of fractional pressure drop obtained experimentally[22] at $f=1000$ Hz [the five points in Fig. 5(c)] are small and hence do not give an appreciable correction into the fitting procedure compared with the values at lower frequencies. Another reason could be due to a large uncertainty in the experimental determination of small values of fractional pressure drop especially at lower flow rates.

V. CONCLUSION

Effective viscosity of ferrofluids in an externally imposed magnetic field depends not only on the field amplitude and frequency but also on the flow vorticity, which causes non-Newtonian properties of the fluids. Their manifestations are especially interesting at the pipe flow since its vorticity $\Omega(r)$ is not a constant in the pipe's cross section but varies from point to point. As a result, the velocity profile ceases to be parabolic and the flow rate deviates from the Poiseuille formula.

The non-Newtonian behavior becomes detectable when the dimensionless characteristic vorticity $\Omega_0\tau_B$ exceeds unity which is possible in practice if "elementary units" in a magnetic suspension represent not single particles but their aggregates whose Brownian time of rotational diffusion τ_B is sufficiently large. The replacement of magnetic grains by their aggregates strongly complicates a theoretical description because the very parameters of these aggregates—such as the initial magnetic susceptibility and the Brownian relaxation time itself—turn out to be dependent on the flow rate. The dependence was established experimentally in a recent work[22] where the fluid parameter values were determined by the fit. We propose a complementary description based on an assumption of chainlike aggregates acted upon by the shear stress. Our simple model has allowed us to fit quite satisfactory experimental data of Schumacher *et al.*[22]

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