Flow of a ferrofluid down a tube in an oscillating magnetic field

B. U. Felderhof

Institut für Theoretische Physik A, RWTH Aachen Templergraben 55, 52056 Aachen, Germany (Received 22 January 2001; published 19 July 2001)

The magnetoviscosity of a ferrofluid flowing down a circular tube in the presence of a magnetic field oscillating in the direction of the axis is studied on the basis of ferrohydrodynamics, Maxwell's equations of magnetostatics, and a relaxation equation for the magnetization. Three different relaxation equations, proposed in the literature, are considered. For large amplitude of the oscillating field the three equations lead to different values of the magnetoviscosity. For large magnetic permeability the self-consistent magnetic field generated by the magnetization has significant effect.

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I. INTRODUCTION

In the absence of an applied magnetic field the flow of a ferrofluid down a circular tube shows the familiar parabolic Poiseuille pattern [1]. If a steady magnetic field is applied along the tube, then for the same pressure gradient the flow pattern remains parabolic but the flow rate is strongly reduced. This is interpreted as an increase of viscosity, and is called magnetoviscosity [2-4]. Surprisingly, if the field oscillates in time then the flow rate increases provided the frequency of oscillation is sufficiently high [5,6]. This phenomenon has been called "negative viscosity" of a ferrofluid. The name is not quite appropriate, since the transport coefficient viscosity does not change noticeably. Rather, in addition to the pressure gradient there is an oscillating magnetic force density, with nonvanishing time average accelerating the fluid. At low frequency the net magnetic force density acts against the pressure gradient and the fluid is slowed down.

The so-called negative viscosity effect was predicted theoretically by Shliomis and Morozov [5], and was first demonstrated experimentally by Bacri *et al.* [7]. The original theory [5], based on Shliomis' relaxation equation for the magnetization [2], did not fit the experimental data well. A modified theory, based on a different relaxation equation [8,9], provided qualitative agreement with the data [7].

The theories [5,7] mentioned above omit the effect of the demagnetizing field. As we shall show, the omission can be justified only if the initial susceptibility of the ferrofluid is small. In the experiment of Bacri *et al.* [7] the susceptibility is large [10] and the demagnetizing field has a considerable effect. In the theory of Zahn and Greer [11] of flow in a planar duct the demagnetizing effect was taken into account correctly but the authors limit themselves to a linear magnetic equation of state and neglect the generation of higher harmonics. Experimentally one easily gets into the nonlinear regime of the equation of state, and the neglect of higher harmonics cannot be justified theoretically.

In the following we calculate the effective viscosity to first order in the applied pressure gradient. It is assumed that the radius of the tube is much larger than the Stokes length characterizing the boundary layer thickness. The Stokes length is given by $\sqrt{\eta/(\rho\omega)}$, where η is the shear viscosity, ρ the mass density, and ω the frequency.

The calculation is based on ferrohydrodynamics, Maxwell's equations of magnetostatics, and a relaxation equation for the magnetization. The latter is the least established. For dense ferrofluids a detailed kinetic theory allowing a microscopic calculation of the relaxation behavior is not available, and one is forced to take a phenomenological point of view. We investigate the consequences of three different relaxation equations, which have been proposed in the literature, namely, Shliomis' relaxation equation [2], the modified equation of Martsenyuk et al. [8], and the relaxation equation of Felderhof and Kroh [12]. The relaxation equation of Martsenyuk et al. [8] was derived for a dilute ferrofluid in an effective field approximation for the orientational distribution function. The relaxation equation of Felderhof and Kroh [12] was proposed on the basis of irreversible thermodynamics. For definiteness we make the additional assumption that the relaxation time appearing in the equation does not depend on magnetic field or magnetization.

Zeuner *et al.* [13] measured the negative viscosity effect for a dilute ferrofluid, and compared with the theory of Bacri *et al.* [7]. For a dilute ferrofluid the magnetic field generated by the magnetization is much smaller than the applied field and may be neglected. Zeuner *et al.* [14,15] discussed the theory in some detail.

II. BASIC EQUATIONS

In the approximation of fast rotational relaxation the mean rate of rotation $\boldsymbol{\omega}_p$ of the suspended particles of a ferrofluid is determined by the local fluid vorticity $\boldsymbol{\Omega} = \frac{1}{2} \nabla \times \boldsymbol{v}$ and the local magnetic torque density as

$$\boldsymbol{\omega}_{p} = \boldsymbol{\Omega} + \frac{1}{4\zeta} \boldsymbol{M} \times \boldsymbol{H}, \qquad (1)$$

where ζ is the vortex viscosity, M the magnetization, and H the local magnetic field. In this approximation the antisymmetric part of the total stress tensor vanishes [12,16], and the fluid equation of motion becomes

$$\rho \frac{D\boldsymbol{v}}{Dt} = \boldsymbol{\nabla} \cdot (\boldsymbol{\sigma}_{hyd}^{S} + \boldsymbol{\sigma}_{m}^{S}), \qquad (2)$$

where $D/Dt = \partial/\partial + \boldsymbol{v} \cdot \boldsymbol{\nabla}$ is the substantial derivative, and $\boldsymbol{\sigma}_{hyd}^{S}$ is the symmetric part of the hydrodynamic stress tensor, given by

$$\sigma_{hyd,\alpha\beta}^{S} = -p\,\delta_{\alpha\beta} + \eta \left[\partial_{\alpha}v_{\beta} + \partial_{\beta}v_{\alpha} - \frac{2}{3} (\boldsymbol{\nabla} \cdot \boldsymbol{v}) \,\delta_{\alpha\beta} \right] \\ + \zeta_{v} (\boldsymbol{\nabla} \cdot \boldsymbol{v}) \,\delta_{\alpha\beta}, \qquad (3)$$

where *p* is the pressure, η is the shear viscosity, and ζ_v is the volume viscosity. The symmetric part of the magnetic stress tensor is given by [4]

$$\boldsymbol{\sigma}_{m}^{S} = \frac{1}{8\pi} (\boldsymbol{B}\boldsymbol{H} + \boldsymbol{H}\boldsymbol{B}) + \frac{1}{8\pi} H^{2} \mathbf{1}$$
(4)

with magnetic induction

$$\boldsymbol{B} = \boldsymbol{H} + 4\,\boldsymbol{\pi}\boldsymbol{M}.\tag{5}$$

The magnetic induction B and magnetic field H satisfy Maxwell's equations of magnetostatics

$$\nabla \cdot \boldsymbol{B} = 0, \quad \nabla \times \boldsymbol{H} = 0. \tag{6}$$

Using these equations one can write

$$\nabla \cdot \boldsymbol{\sigma}_{m}^{S} = \boldsymbol{M} \cdot (\nabla \boldsymbol{H}) + \frac{1}{2} \nabla \times (\boldsymbol{M} \times \boldsymbol{H}).$$
(7)

The first term on the right is the Kelvin force density. Note that the second term can be expressed as the divergence of an antisymmetric tensor.

The above equations must be supplemented with a constitutive equation for the magnetization. We shall study the consequences of three different relaxation equations, which have been proposed in the literature. The first equation is due to Shliomis [2]. When extended to the case of a compressible fluid it reads

$$\frac{\partial \boldsymbol{M}}{\partial t} + \boldsymbol{\nabla} \cdot (\boldsymbol{v}\boldsymbol{M}) - \boldsymbol{\Omega} \times \boldsymbol{M}$$
$$= \frac{-1}{\tau_B} (\boldsymbol{M} - \boldsymbol{M}_e) - \frac{1}{4\zeta} \boldsymbol{M} \times (\boldsymbol{M} \times \boldsymbol{H}), \qquad (8)$$

where τ_B is the Brownian relaxation time, and the local equilibrium magnetization M_e is determined by the local magnetic field H according to the equilibrium equation of state

$$\boldsymbol{M}_{\rho} = \boldsymbol{H} \boldsymbol{A}(\boldsymbol{H}). \tag{9}$$

Shliomis used the expression

$$A(H) = \frac{M_S}{H} L\left(\frac{3\chi_0 H}{M_S}\right) \tag{10}$$

with the Langevin function $L(\xi) = \operatorname{coth} \xi - \xi^{-1}$ and with saturation magnetization M_s and initial susceptibility χ_0 . He considered a dilute ferrofluid for which $M_s = nm$ and $\chi_0 = nm^2/(3k_BT_0)$, where *n* is the number density of Brownian

particles, *m* is the size of the magnetic moment of a particle, and T_0 is the temperature. More generally we may use Eq. (10) as an approximate equation of state with parameters M_S and χ_0 . In our numerical work we shall assume that the vortex viscosity ζ is related to the relaxation time τ_B by [2,4]

$$\zeta_1 = \frac{M_S^2}{6\chi_0} \tau_B. \tag{11}$$

We use the subscript 1 to distinguish this case. For a dilute ferrofluid the relation is exact and becomes $\zeta_1 = \frac{1}{2}nk_BT_0\tau_B$. More generally τ_B and ζ are independent parameters.

A second relaxation equation was derived by Martsenyuk *et al.* from Brownian motion theory in an effective field approximation [8,9]. It takes the form

$$\frac{\partial \boldsymbol{M}}{\partial t} + \boldsymbol{\nabla} \cdot (\boldsymbol{v}\boldsymbol{M}) - \boldsymbol{\Omega} \times \boldsymbol{M}$$
$$= -\frac{1}{\tau_B} \left[\boldsymbol{M} - \frac{3\chi_0 L(\boldsymbol{\xi}_e)}{\boldsymbol{\xi}_e} \boldsymbol{H} + \frac{1}{2} \left(1 - \frac{3L(\boldsymbol{\xi}_e)}{\boldsymbol{\xi}_e} \right) \right]$$
$$\times \frac{3\chi_0}{M^2} \boldsymbol{M} \times (\boldsymbol{M} \times \boldsymbol{H}) , \qquad (12)$$

where $\xi_e(t)$ follows from the magnetization M(t) by the relation $M = M_S L(\xi_e)$.

A third relaxation equation was proposed by Felderhof and Kroh [12] on the basis of irreversible thermodynamics. The equation reads

$$\frac{\partial M}{\partial t} + \nabla \cdot (\boldsymbol{v}\boldsymbol{M}) - \boldsymbol{\Omega} \times \boldsymbol{M}$$
$$= \frac{\chi_0}{\tau_B} (\boldsymbol{H} - \boldsymbol{H}_e) - \frac{1}{4\zeta} \boldsymbol{M} \times (\boldsymbol{M} \times \boldsymbol{H}), \qquad (13)$$

where the local equilibrium field H_e is determined by the magnetization according to the equilibrium equation of state

$$\boldsymbol{H}_{e} = \boldsymbol{M}\boldsymbol{C}(\boldsymbol{M}), \tag{14}$$

with the function C(M) satisfying the identity C(M) = 1/A(MC(M)). We note that with $B_e = H_e + 4\pi M$ and $B = H + 4\pi M$ one can write alternatively $H - H_e = B - B_e$. We have chosen the relaxation time in Eq. (13) such that the equation reduces to Shliomis' relaxation equation Eq. (8) for small deviations from equilibrium at zero field. We shall show that the three relaxation equations Eqs. (8), (12), and (13) lead to drastically different predictions for the dependence of the magnetoviscosity on amplitude and frequency of the applied oscillating magnetic field.

III. FLOW IN OSCILLATING MAGNETIC FIELD

We consider a ferrofluid in a circular tube of radius a, perturbed by an oscillating magnetic field directed along the axis of the tube. We take the z axis along the axis of the tube.

The uniform applied magnetic filed is

$$\boldsymbol{H}_{0}(t) = \boldsymbol{H}_{0z} \boldsymbol{e}_{z} \cos \omega t. \tag{15}$$

Consider first the case of zero applied pressure gradient. Then the fluid remains at rest, and the magnetization oscillates according to

$$\boldsymbol{M}_0(t) = \boldsymbol{M}_S F(t) \boldsymbol{e}_z \tag{16}$$

with a periodic function F(t) that averages to zero over the period $T=2\pi/\omega$. The pressure will oscillate in time, but is spatially uniform. The function F(t) depends on amplitude H_{0z} and frequency ω . Its behavior for each of the three relaxation equations is different. For Shliomis' relaxation equation Eq. (8) the function F(t) satisfies

$$\tau_B \frac{dF}{dt} = -F + L(\xi \cos \omega t), \qquad (17)$$

if the equation of state Eq. (10) is used, with

$$\xi = \frac{3\chi_0}{M_S} H_{0z} \,. \tag{18}$$

For the relaxation equation Eq. (12) of Martsenyuk *et al.* [8] the function F(t) satisfies

$$\tau_B \frac{dF}{dt} = -F + \frac{F}{\xi_F} \xi \cos \omega t, \qquad (19)$$

with ξ_F defined by $F = L(\xi_F)$. For the relaxation equation Eq. (13) the function F(t) satisfies

$$\tau_B \frac{dF}{dt} = -\chi_0 F C(|M_S F|) + \frac{1}{3} \xi \cos \omega t.$$
 (20)

For small ξ the function F(t) remains small and the three equations Eqs. (17), (19), and (20) become identical, since $\xi_F \approx 3F$ and $C(0) = \chi_0^{-1}$. For larger values of ξ the solutions of the three equations can differ significantly.

An applied pressure gradient perturbs the situation described above. We shall calculate the resulting flow to first order in the applied pressure gradient, and indicate the corresponding additional fields v_1 , M_1 , H_1 . The pressure is

$$p = p_0 - kz, \tag{21}$$

where p_0 may be time dependent, but does not depend on spatial coordinates. We solve the basic equations to first order in the pressure gradient k by making the ansatz

$$\boldsymbol{v}_{1}(\boldsymbol{r},t) = f(\boldsymbol{r},t)\boldsymbol{e}_{z}, \quad \boldsymbol{r} = \sqrt{x^{2} + y^{2}},$$

$$\boldsymbol{M}_{1}(\boldsymbol{r},t) = \boldsymbol{M}_{r}(\boldsymbol{r},t)\boldsymbol{e}_{r}, \quad (22)$$

$$\boldsymbol{H}_{1}(\boldsymbol{r},t) = -4\pi\boldsymbol{M}_{r}(\boldsymbol{r},t)\boldsymbol{e}_{r},$$

for $r \le a$. Here e_r is the radial unit vector. In the vacuum outside the tube $B = H_0$. It follows from Eq. (22) that $B_1 = 0$ everywhere, so that the normal component of **B** is con-

tinuous across the tube boundary. Maxwell's equations Eq. (6) are clearly satisfied everywhere. The field H_1 is the demagnetizing field.

The flow \boldsymbol{v}_1 is incompressible, $\nabla \cdot \boldsymbol{v}_1 = 0$, and the vorticity is

$$\mathbf{\Omega}_1 = -\frac{1}{2} \frac{\partial f}{\partial r} \boldsymbol{e}_{\varphi} \tag{23}$$

with azimuthal unit vector e_{φ} . By substitution into Eqs. (8), (12), and (13) we find that to first order the radial magnetization component $M_r(r,t)$ satisfies the equation

$$\frac{\partial M_r}{\partial t} + \frac{1}{2} \frac{\partial f}{\partial r} M_{0z} = -\frac{1}{\tau_B} V(t) M_r \tag{24}$$

with different damping factor V(t) for each of the three relaxation equations. The three damping factors will be specified later. It suffices to note here that each of the factors is spatially uniform and periodic in time with period $\frac{1}{2}T$.

The z component of the equation of motion Eq. (2) becomes with the ansatz Eq. (22) to first order in k

$$\rho \frac{\partial f}{\partial t} = \eta \left(\frac{\partial^2 f}{\partial r^2} + \frac{1}{r} \frac{\partial f}{\partial r} \right) + k - \frac{1}{2} B_{0z} \left(\frac{\partial M_r}{\partial r} + \frac{1}{r} M_r \right).$$
(25)

We must satisfy the stick boundary condition at the tube radius

$$f(a,t) = 0. \tag{26}$$

It is instructive to consider first the solution of Eqs. (24), (25), and (26) for the static case $\omega = 0$. Then $M_{0z} = H_{0z}A(H_{0z})$ is the equilibrium magnetization M_{eq} in field H_{0z} and the damping factor is a constant V_{st} . The equations are solved by

$$f(r) = \frac{1}{4} \frac{k}{\eta + \Delta \eta_{\parallel}} (a^2 - r^2),$$

$$M_r = \frac{1}{4} \frac{k}{\eta + \Delta \eta_{\parallel}} \tau_B \frac{M_{eq}}{V_{st}} r$$
(27)

with viscosity change

$$\Delta \eta_{\parallel} = \frac{M_{eq} B_{eq}}{4 V_{st}} \tau_B \,. \tag{28}$$

At frequency $\omega \neq 0$ the functions f(r,t) and $M_r(r,t)$ will be periodic in time. In the following we assume that the tube radius is sufficiently large that boundary layer effects can be neglected. We restrict attention to the corresponding bulk solution.

IV. CALCULATION OF MAGNETOVISCOSITY

At frequency ω the viscous term in Eq. (25) leads to a Stokes length scale $\ell = \sqrt{\eta/(\rho\omega)}$. If the frequency is sufficiently high that this length scale is much smaller than the

radius *a* of the tube, then the solution can be divided into a bulk part and a boundary layer part. In the following we assume $\omega \ge \eta/(a^2\rho)$, and concentrate on the bulk part, neglecting the boundary layer. In the experiment of Bacri *et al.* [7] the zero field viscosity is $\eta = 77$ cP, the radius *a* is 0.05 cm, the mass density is $\rho \approx 1.3$ g/cm³, so that the inequality is satisfied for frequencies $(\omega/2\pi) \ge 40$ Hz.

The bulk part of the flow consists of the time-averaged flow $\overline{\mathbf{v}_1} = \overline{f}(r) \mathbf{e}_z$ and a spatially uniform flow oscillating in the *z* direction. For a calculation of the effective magnetoviscosity only the time-averaged flow and the oscillating radial component of the magnetization are relevant. For the bulk solution the radial component of the magnetization behaves as $M_r(r,t) \approx R_1(t)r$, with a periodic function $R_1(t)$. The time-averaged equation, Eq. (25), for the bulk flow reads

$$\eta \left(\frac{\partial^2 \overline{f}}{\partial r^2} + \frac{1}{r} \frac{\partial \overline{f}}{\partial r} \right) + k - \overline{B_{0z} R_1} = 0.$$
⁽²⁹⁾

The solution satisfying the boundary condition $\overline{f}(a)=0$ has the Poiseuille form

$$\overline{f}(r) = w(a^2 - r^2). \tag{30}$$

From Eq. (29) one finds for the corresponding viscosity change

$$\Delta \eta(\omega) = \frac{\overline{B_{0z}R_1}}{4w}.$$
(31)

We write $R_1(t) = M_S w \tau_B G(t)$ and find from Eq. (24) the ordinary differential equation

$$\tau_B \frac{dG}{dt} + V(t)G = F(t), \qquad (32)$$

where we have used Eq. (16) and the fact that the oscillating part of the bulk flow is spatially uniform. We need the solution of Eq. (32) which is periodic in time. This can be found by integration from $t = -\infty$. As a result

$$G(t) = \frac{1}{\tau_B} \int_0^\infty \exp[X(t-t') - X(t) - \bar{V}t'/\tau_B] F(t-t') dt',$$
(33)

where \overline{V} is the time average of V(t) and X(t) is the periodic solution of the equation

$$\tau_B \frac{dX}{dt} = V(t) - \bar{V}.$$
(34)

The solution X(t) is unique up to a constant, which cancels in Eq. (33). The period is $\frac{1}{2}T$. We define the dimensionless magnetic induction b(t) by $B_{0z}(t) = (M_S/3\chi_0)b(t)$. With this definition

$$b(t) = \xi \cos \omega t + 12\pi \chi_0 F(t) \tag{35}$$

and Eq. (31) becomes

$$\Delta \eta(\omega) = \frac{1}{2} \zeta_1 \overline{Gb}, \qquad (36)$$

where we have used Eq. (11). The calculation of the time average \overline{Gb} requires the solution F(t) of Eqs. (17), (19), or (20), the damping factor V(t) corresponding to one of the three relaxation equations, the solution X(t) of Eq. (34), and calculation of the integral in Eq. (33). Except in the limit of weak imposed field these calculations must be performed numerically.

At this point we give the explicit expressions for the damping factor V(t) corresponding to the three relaxation equations. For Shliomis' relaxation equation Eq. (8) the damping factor becomes

$$V(t) = 1 + 12\pi\chi_0 \frac{L(\xi\cos\omega t)}{\xi\cos\omega t} + \frac{\zeta_1}{2\zeta}F(t)b(t).$$
(37)

Shliomis [2] assumed moreover $\zeta = \zeta_1$. For the relaxation equation Eq. (12) of Martsenyuk *et al.* [8] the damping factor is

$$V(t) = 1 + 12\pi\chi_0 \frac{F(t)}{\xi_F(t)} + \frac{1}{2} \left(\frac{1}{F(t)} - \frac{3}{\xi_F(t)} \right) b(t).$$
(38)

For the relaxation equation, Eq. (13), of Felderhof and Kroh [12] the damping factor is

$$V(t) = \chi_0 C(|M_S F(t)|) + 4\pi\chi_0 + \frac{\zeta_1}{2\zeta}F(t)b(t).$$
(39)

In the limit of weak applied field the function F(t) becomes

$$F_{w}(t) = \frac{1}{3}\xi\cos\alpha\cos(\omega t - \alpha)$$
(40)

with lag angle $\alpha = \arctan \omega \tau_B$. The three relaxation equations become identical with damping factor

$$V_w = 1 + 4\,\pi\chi_0 = \mu,\tag{41}$$

where μ is the magnetic permeability. From Eq. (33) one finds correspondingly

$$G_{w}(t) = \frac{1}{3\mu} \xi \cos \alpha \cos \beta \cos(\omega t - \alpha - \beta) \qquad (42)$$

with $\beta = \arctan(\omega \tau_B/\mu)$. Hence one finds the time average

$$\overline{G_w b_w} = \frac{1}{6} \xi^2 \frac{\mu^2 - \omega^2 \tau_B^2}{(1 + \omega^2 \tau_B^2)(\mu^2 + \omega^2 \tau_B^2)}.$$
 (43)

This agrees with the result of Shliomis and Morozov [5] only in the limit $\chi_0=0$. These authors omitted the demagnetization effect. In the experiment of Bacri *et al.* [7,10] the magnetic permeability is $\mu = 5.4$, so that the frequency at which $\Delta \eta_w(\omega)$ vanishes is shifted to a significantly higher value.

It is of interest to consider the zero frequency limit $\omega \tau_B \ll 1$ for arbitrary values of the applied field. It follows from Eqs. (17) and (19) that in this limit $F(t) = L(\xi \cos \omega t)$

for both relaxation equations. The same expression follows from Eq. (20) if Eq. (10) is used. From Eq. (32) we find in the limit $\omega \tau_B \rightarrow 0$ the relation G(t) = F(t)/V(t). The damping factor V(t) follows from Eqs. (37), (38), or (39) with the substitution $F(t) = L(\xi \cos \omega t)$. The zero fequency limit of $\Delta \eta(\omega)$ then follows from the time average \overline{Gb} according to Eq. (36). The product \overline{Gb} must be averaged over the period. We cannot take the limit $\omega \rightarrow 0$ for each factor separately. This would yield the steady state value $\Delta \eta_{\parallel}$ given by Eq. (28), which differs from the zero frequency limit of $\Delta \eta(\omega)$.

For the steady state value V_{st} we find for Shliomis' relaxation equation

$$V_{st} = \left(\frac{1}{\xi} + \frac{\zeta_1}{2\zeta}L(\xi)\right) [\xi + 12\pi\chi_0 L(\xi)]. \tag{44}$$

The second factor is related to the equilibrium magnetic induction by $B_{eq} = (M_s/3\chi_0)b_{eq}$ with

$$b_{eq} = \xi + 12\pi\chi_0 L(\xi), \tag{45}$$

so that this factor cancels in Eq. (28). For the relaxation equation of Martsenyuk *et al.* we find the steady state value

$$V_{st} = \left(\frac{1}{2L(\xi)} - \frac{1}{2\xi}\right) b_{eq} \,. \tag{46}$$

For the relaxation equation of Felderhof and Kroh we find

$$V_{st} = \left(\frac{1}{3L(\xi)} + \frac{\zeta_1}{2\zeta}L(\xi)\right)b_{eq}.$$
(47)

In the remainder of this section we assume the equality $\zeta = \zeta_1$. Substituting Eqs. (44), (46), and (47) into Eq. (28) we find for the steady state magnetoviscosity expressions of the form

$$\Delta \eta_{\parallel} = \zeta_1 R(\xi), \tag{48}$$

with for Shliomis' relaxation equation, Eq. (8)

$$R(\xi) = \frac{\xi L(\xi)}{2 + \xi L(\xi)},\tag{49}$$

for the relaxation equation, Eq. (12), of Martsenyuk et al.

$$R(\xi) = \frac{\xi L^2(\xi)}{\xi - L(\xi)},$$
(50)

and for the relaxation equation Eq. (13) of Felderhof and Kroh

$$R(\xi) = \frac{3L^2(\xi)}{2+3L^2(\xi)}.$$
(51)

The corresponding equations for the zero frequency limit read

$$\Delta \eta(0) = \frac{\zeta_1}{2\pi} \int_0^{2\pi} R(\xi \cos x) dx.$$
 (52)



FIG. 1. Plot of the ratio $\Delta \eta(0)/\Delta \eta_{\parallel}$, i.e., the ratio of the zerofrequency limit of the magnetoviscosity to its steady-state value, as a function of ξ , the dimensionless applied magnetic field, for Shliomis' relaxation equation Eq. (8) (long dashes), for the relaxation equation Eq. (12) of Martsenyuk *et al.* (solid curve), and for the relaxation equation Eq. (13) of Felderhof and Kroh (short dashes).

Note that the above expressions are independent of the initial susceptibility χ_0 . In Fig. 1 we compare the ratio $\Delta \eta(0)/\Delta \eta_{\parallel}$ as a function of ξ for the three relaxation equations. In the weak field limit the ratio equals $\frac{1}{2}$ in all three cases. In Fig. 2 we plot the ratio $\Delta \eta(0)/\zeta_1$ as a function of ξ for the three relaxation equations.

As indicated below Eq. (36), for strong imposed oscillating field the calculation of the frequency-dependent viscosity $\Delta \eta(\omega)$ requires numerical integration. For frequencies different from zero the viscosity depends on the initial susceptibility χ_0 . We consider in particular the limit of small χ_0 and the value $\chi_0 = 0.35$ corresponding to the experiment of Bacri et al. [7,10]. In the limit of small χ_0 the self-consistent magnetic field generated by the induced magnetization is much smaller than the applied field. For $\chi_0 = 0.35$ the selfconsistent field cannot be neglected. In Fig. 3 we show a contour plot of $\Delta \eta(\omega)/\zeta_1$ in the $\omega \xi$ plane as calculated in the limit of small χ_0 for the relaxation equation of Martsenyuk et al. In Fig. 4 we show the analogous plot for χ_0 =0.35. In Fig. 5 we compare the ratio $\Delta \eta(\omega)/\zeta_1$ for χ_0 $\rightarrow 0$ and $\chi_0 = 0.35$ as a function of ω for the particular value $\xi = 10$, again for the relaxation equation of Martsenyuk *et al.* [8].



FIG. 2. Plot of the ratio $\Delta \eta(0)/\zeta_1$ as a function of ξ for the three relaxation equations. Equation (8) (long dashes), Eq. (12) (solid curve), Eq. (13) (short dashes).



FIG. 3. Contour plot of $\Delta \eta(\omega)/\zeta_1$ in the $\omega \xi$ plane as calculated from the relaxation equation of Martsenyuk *et al.* in the limit of small susceptibility χ_0 . We plot the contours for values, successively from left to right, 0.3, 0(bold), -0.1, -0.2, -0.3, -0.4, -0.5.

V. DISCUSSION

The calculations performed above on the effect of an oscillating magnetic field on the rate of flow of a ferrofluid through a circular tube provide useful insight into the magnetoviscosity phenomenon. As evident from Eq. (29), the rate of flow is determined by the time average of the product of the magnetic induction in the direction of the tube and the



FIG. 4. Contour plot of $\Delta \eta(\omega)/\zeta_1$ in the $\omega\xi$ plane as calculated from the relaxation equation of Martsenyuk *et al.* for susceptibility $\chi_0 = 0.35$. We plot the contours for values, successively from left to right, 0.4, 0.3, 0.2, 0.1, 0(bold), -0.01, -0.02, -0.03, -0.04.



FIG. 5. Plot of the ratio $\Delta \eta(\omega)/\zeta_1$ as a function of $\omega \tau_B$ in the limit $\chi_0 \rightarrow 0$ (solid curve) and for $\chi_0 = 0.35$ (short dashes) for $\xi = 10$ and for the relaxation equation Eq. (12) of Martsenyuk *et al.*

component of the magnetization transverse to the tube. The rate of flow can be enhanced or reduced depending on the magnitude of these two fields and on their phase relationship. As an example we show in Fig. 6 the dimensionless induction b(t), defined in Eq. (35), the reduced transverse magnetization G(t), as defined above Eq. (32), as well as the product G(t)b(t), for $\xi=10$, $\omega\tau_B=10$, $\chi_0=0.35$, and the relaxation equation of Martsenyuk *et al.* [8]. For this choice of parameters the time average of the product G(t)b(t) is negative. Correspondingly the magnetoviscosity, given by Eq. (36), is negative.

If the susceptibility of the ferrofluid is high, then it is essential to calculate the magnetic field self-consistently from the induced magnetization. As a consequence the geometry of the flow situation must be considered. Magnetoviscosity cannot be used as a local transport coefficient. Nonetheless, for a given flow situation it is a convenient quantity, expressing the net effect of the magnetic field on the flow.

The calculation has been based on macroscopic equations. For dense ferrofluids the equation describing the relaxation of magnetization is not well established. We have used three different relaxation equations to show the effect of each



FIG. 6. Plot of the dimensionless induction b(t) (solid curve), the reduced transverse magnetization G(t) (multiplied by 600, long dashes), and the product G(t)b(t) (multiplied by 300, short dashes) for $\xi=10$, $\omega\tau_B=10$, $\chi_0=0.35$, and the relaxation equation Eq. (12). The magnetoviscosity is given by the time average of the product G(t)b(t) according to Eq. (36).

choice on magnetoviscosity. For a dilute ferrofluid the relaxation equation of Martsenyuk *et al.* [8] provides a good approximation to the exact result [17] for the steady state magnetoviscosity. We have used this relaxation equation in the numerical work for Figs. 3–6. However, it should be kept in mind that the equation may not describe the relaxation behavior in a dense ferrofluid correctly. A more accurate theory of magnetoviscosity of a dense ferrofluid in an oscillating magnetic field would require an independent study of the relaxation of magnetization.

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