"Negative-Viscosity" Effect in a Magnetic Fluid

J.-C. Bacri and R. Perzynski
Laboratoire d'Acoustique et Optique de la Matière Condensée, Université Pierre et Marie Curie, Tour 13, Boîte 78, 4 place Jussieu, 75252 Paris Cedex 05, France

M. I. Shliomis
Department of Physics of Complex Systems, the Weizmann Institute of Science, Rehovot 76100, Israel

G. I. Burde
Institute for Desert Research, Sede Boker Campus, 84993, Israel
(Received 19 July 1994)

The first experimental evidence of a "negative-viscosity" effect in a magnetic fluid is presented here. In a Poiseuille flow, a constant magnetic field balances vorticity and impedes the rotation of individual magnetic particles. Conversely, an alternating magnetic field helps vorticity and favors this rotation: The magnetic energy is partially transformed into the angular momentum of the particles, which in turn is converted into a hydrodynamic motion of the fluid. It manifests itself in a decrease of the total viscosity: Its rotational part becomes negative. The theory developed here corroborates the experimental results.

PACS numbers: 47.15.–x, 75.50.Mm, 83.85.Jn

Magnetic fluids (MF) [1] are colloidal solutions of magnetic nanoparticles in a fluid carrier. The ability of the suspended solid particles to rotate with respect to the surrounding liquid leads to the spectacular behavior of MF in alternating, circularly or linearly polarized magnetic fields [2–5]. A mean angular velocity of the particles, $\boldsymbol{\omega}_p$, is determined by the condition of magnetic and viscous torques balance [1,6]:

$$6\eta\varphi(\boldsymbol{\omega}_p - \boldsymbol{\Omega}) = \mathbf{M} \times \mathbf{H}. \tag{1}$$

So the magnetic torque supports some difference between $\boldsymbol{\omega}_p$ and the angular fluid velocity $\boldsymbol{\Omega} = \frac{1}{2}\operatorname{curl}\mathbf{v}$. In Eq. (1), $\mathbf{M}$ and η are the MF magnetization and viscosity and φ is the volume fraction of particles. Thus, the magnetic field $\mathbf{H}$ "switches on" an additional viscous friction, which manifests itself as an additional (rotational) viscosity $\Delta\eta$. One can show [4] (see below) that the rotational viscosity may be presented in the form

$$\Delta\eta = \frac{3}{2}\,\eta\varphi\,\frac{\Omega - \omega_p}{\Omega}. \tag{2}$$

A constant field always impedes free particle rotation (with the angular velocity Ω), so $\omega_p < \Omega$ and $\Delta\eta > 0$. Inverse inequalities are impossible on thermodynamic considerations because a constant field (which can be created by a permanent magnet) cannot produce any work.

Another situation takes place in an alternating linearly polarized magnetic field. In a quiescent MF ($\Omega = 0$), such a field induces rotational swings of the particles but does not single out any preferable direction of their rotation. Therefore an averaging over a physically small element of volume results in $\omega_p = 0$. Any vortex flow ($\Omega \neq 0$) is sufficient, however, to break down the degeneracy of the rotation direction of the particles and leads to a nonzero macroscopic ω_p. A slow oscillating magnetic field (just as a constant one) impedes free particle rotation, so $\Delta\eta > 0$. Conversely, a fast enough oscillating field forces the particles to rotate faster than the fluid: $\omega_p > \Omega$. As a result the particles *spin up* the flow, so the vortex flow rises up at the expense of the particle spinning, which in turn originates from the alternating magnetic field. This transformation of a part of the alternating field energy into kinetic energy of the fluid just manifests itself in a certain reduction of the total viscosity. Below, this viscosity reduction is named the *negative-viscosity effect:* $\Delta\eta < 0$. In this paper we describe some experiments devoted to showing the different regimes of the rotational viscosity ($\Delta\eta > 0$ or <0) as a function of the amplitude H_0 and the frequency ω of the field.

The magnetic fluid used here is chemically synthesized through Massart's method; the magnetic material of the colloidal particles is Co-ferrite with a magnetic anisotropy constant $K = 2 \times 10^6$ ergs/cm^3, a grain magnetization $M_s = 400$ G, and the typical size of particles of 10 nm. This large anisotropy constant induces a rigid dipole behavior of the particles, which means that the magnetic moment is frozen into the particles up to magnetic fields of 10^4 Oe. In order to maximize the experimental effects, we use a concentrated ferrofluid in water with a volume fraction of 20%. A Poiseuille flow in a horizontal capillary tube ($d = 1$ mm for the diameter) is used for viscosity measurements. The gradient of pressure is given by the difference of the MF level h between the inlet and outlet of the tube. The massic flow rate Q is measured by a numerical weighter at the outlet of the tube. This tube is put inside a solenoid which provides a magnetic field parallel to the flow direction up to $H = 1$ kOe at

 0031-9007/95/75(11)/2128(4)$06.00

a frequency $f = \omega/2\pi$ varying from 0 to 1 kHz. The massic flow rate Q is measured as a function of the pressure drop given by h. Figure 1 presents the variations of Q as a function of h for static fields $H = 0$ and 2000 Oe. The two straight lines establish that the MF has a Newtonian behavior with and without magnetic field and the slopes give the viscosity.

For the orders of magnitude, the viscosity of the solvent is 1 cP (water), with a volume fraction of particles of 20% this viscosity grows up to 77 cP in zero field and up to 220 cP under a static field of 2000 Oe. Despite its large viscosity, our MF is Newtonian.

In Fig. 2, we plot for different frequencies the value of the reduced viscosity

$$\eta_r(H,f) = \Delta\eta/\eta(0,0) = [\eta(H,f) - \eta(0,0)]/\eta(0,0)$$

as a function of the applied magnetic field. For a static magnetic field, the reduced viscosity $\eta_r(H,0)$ increases monotonously towards an asymptotic value $\eta_r(\infty,0)$ in high fields; this fact has been first pointed out experimentally by McTague [7] and theoretically explained by Shliomis [6]. Here $\eta_r(\infty,0) \simeq 2.5$.

At 52 Hz, the curve has more or less the same shape as for $f = 0$ with a maximum in high fields. At 130 Hz the shape of the curve becomes different; η_r first decreases, becomes negative, goes through a minimum, and only increases up to a supposed asymptotic value. For higher frequencies, the behavior is the same; the higher the frequency the more negative the minimum. At 1480 Hz, our apparatus does not permit us to obtain field values higher than 800 Oe. For $H \sim 2000$ Oe, the viscosity decreases from 220 to 100, 77, and 58 cP, respectively, for 50, 250, and 700 Hz.

In conclusion, with this Newtonian fluid we observe a decrease of the rotational viscosity as a function of the frequency from $\Delta\eta = 143$ cP at $f = 0$ down to $\Delta\eta = -19$ cP at $f \geq 700$ Hz.

The first prediction of the negative-viscosity effect [5] was founded on a combined solution of the hydrodynamic and magnetodynamic equations [6]

$$\rho \frac{d\boldsymbol{v}}{dt} = -\nabla\rho + (\mathbf{M}\nabla)\mathbf{H} + \eta\Delta\boldsymbol{v} + \frac{1}{2}\,\mathrm{curl}(\mathbf{M}\times\mathbf{H}), \tag{3}$$

$$\frac{d\mathbf{M}}{dt} = \boldsymbol{\Omega}\times\mathbf{M} - \frac{1}{\tau_B}(\mathbf{M} - \mathbf{M}_0) - \frac{1}{6\eta\varphi}\,\mathbf{M}\times(\mathbf{M}\times\mathbf{H}), \tag{4}$$

where $\tau_B = 3\eta V/kT$ is the Brownian relaxation time for the particles of volume V (kT is the thermal energy) and $\mathbf{M}_0(t) = \varphi M_s L(\xi)\mathbf{h}$ is the "instantaneous equilibrium" magnetization that would exist in given $\mathbf{H}(t)$ at $\tau_B = 0$, i.e., if the magnetization followed the field without retardation; in the Langevin formula $L(\xi) = \coth\xi - \xi^{-1}, \xi = mH(t)/kT, m = M_s V$ is the magnetic moment of the particle, and $\mathbf{h}$ is the unit vector along the applied field.

Our experimental data agree with the theory [5] on the whole, but there are some substantial discrepancies in details. So, the theoretical dependence of $\Delta\eta(\xi)$ has a maximum at $\omega\tau_B < 1$ and has no minimum at $\omega\tau_B > 1$ (see Fig. 4 in [5]), whereas in experiment we observe an inverse situation (see Fig. 2 in this paper). The matter is that the phenomenological magnetization equation (4) is working well for arbitrary intensity of a stationary magnetic field (see, e.g., a good agreement between McTague's experiment [7] and Shliomis's theory [6]) or for small amplitude and frequency of an alternating field. However, for finite values ξ and $\omega\tau_B$ one must use a more rigorous magnetization equation. Such a macroscopic equation was derived [8] from the Fokker-Planck

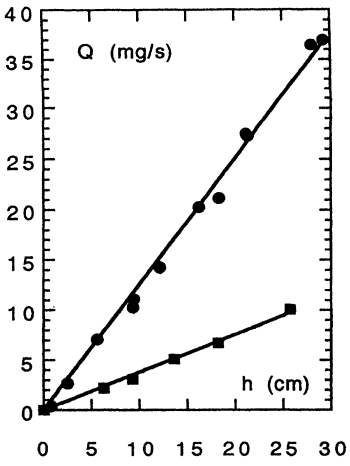

FIG. 1. Experimental massic flow Q as a function of h, the difference of MF level between inlet and outlet: ■: $H = 0, \eta = 77$ cP; ●: $H = 2200$ Oe (static field), $\eta = 220$ cP.

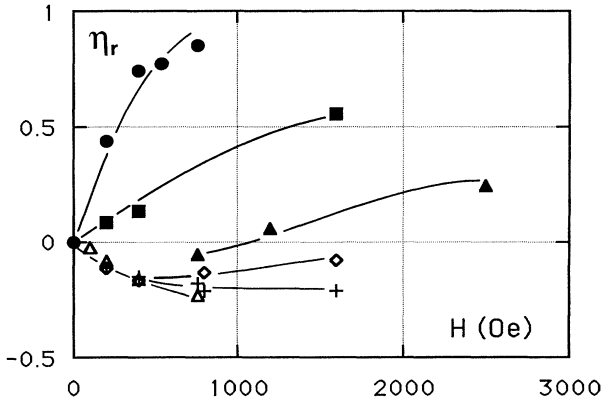

FIG. 2. Experimental reduced viscosity $\eta_r(H,f) = [\eta(H,f) - \eta(0,0)]/\eta(0,0)$ versus magnetic field H for different frequencies f : ●: $f = 0$; ■: $f = 52$ Hz; ▲: $f = 150$ Hz; ◇: $f = 345$ Hz; +: $f = 645$ Hz; and △: $f = 1480$ Hz. Full lines are guides for the eye.

equation for rotary diffusion of colloidal particles with assistance from the *effective field method* [9]. According to this method, the nonequilibrium actual magnetization is considered to be the equilibrium one:

$$\mathbf{M} = \varphi M_S L(\xi_e)\boldsymbol{\xi}_e/\xi \tag{5}$$

in the "effective field" $\boldsymbol{\xi}_e$. The latter is determined as a function of true field $\boldsymbol{\xi} = m\mathbf{H}/kT$ and flow vorticity $\boldsymbol{\Omega}$ by [8]

$$\frac{d}{dt}\left(L_e\frac{\boldsymbol{\xi}_e}{\xi_e}\right) = \boldsymbol{\Omega}\times\left(L_e\frac{\boldsymbol{\xi}_e}{\xi_e}\right) - \frac{1}{\tau_B}\frac{L_e}{\xi_e}(\boldsymbol{\xi}_e - \boldsymbol{\xi}) - \frac{1}{2\tau_B\xi_e^2}\left(1 - \frac{3L_3}{\xi_e}\right)\boldsymbol{\xi}_e\times(\boldsymbol{\xi}_e\times\boldsymbol{\xi}), \tag{6}$$

where $L_e = L(\xi_e)$. Equations (5) and (6) determine $\mathbf{M}$ in a parametric form, with the effective field ξ_e being the parameter. Solutions of Eqs. (4) and (6) are close to each other only for a constant or a slow oscillating magnetic field.

Our problem contains a small dimensionless parameter $\Omega\tau_B$. Indeed, a transient birefringence experiment [10] gives for the present MF $\tau_B \sim 1.6$ ms, so the inequality $\Omega\tau_B \ll 1$ is satisfied for a vorticity Ω smaller than 60 s^{-1}, which is always verified in our experiment. For small $\Omega\tau_B$ a solution of Eqs. (3) and (6) may be found by the theory of perturbation. The effective field ξ_e is determined by the equation of zero approximation:

$$\tau_B\frac{d\xi_e}{dt} = -\left(\frac{d\ln L_e}{d\xi_e}\right)^{-1}\left(1 - \frac{\xi_0}{\xi_e}\cos\omega t\right), \tag{7}$$

where $\xi_0 = mH_0/kT$ is defined over the amplitude of the field. In the linear approximation in $\Omega\tau_B$, Eq. (6) yields $\mathbf{M}^{(1)} = M^{(0)}F(\xi_e)\tau_B\boldsymbol{\Omega}\times h$, where $M^{(0)} = \varphi M_S L(\xi_e)$ and the function F is determined by

$$\tau_B\frac{dF}{dt} = 1 - \frac{1}{2}\left(\frac{1}{L_e} - \frac{1}{\xi_e}\right)\xi_0 F\cos\omega t\,. \tag{8}$$

Because of the appearance of $\mathbf{M}^{(1)}$, the magnetic torque $\mathbf{M}\times\mathbf{H}$ is no longer equal to zero. This torque can be averaged over the period of field variation $2\pi/\omega$, because in our experiments this period is always much smaller than the characteristic hydrodynamic time $\rho d^2/\eta$. For the mean magnetic torque one gets

$$\overline{\mathbf{M}\times\mathbf{H}} = -6\eta\varphi g\boldsymbol{\Omega}, \quad g = \frac{1}{2}\xi_0\,\overline{\cos\omega t L(\xi_e)F(\xi_e)}\,. \tag{9}$$

Substiting (9) into (3) it is easy to see that the last term in the fluid motion relationship may be grouped with the viscous one:

$$\eta\Delta\boldsymbol{v} - 3\eta\varphi g\,\mathrm{curl}\boldsymbol{\Omega} = -2\eta\left(1 + \frac{3}{2}\varphi g\right)\mathrm{curl}\boldsymbol{\Omega}\,. \tag{10}$$

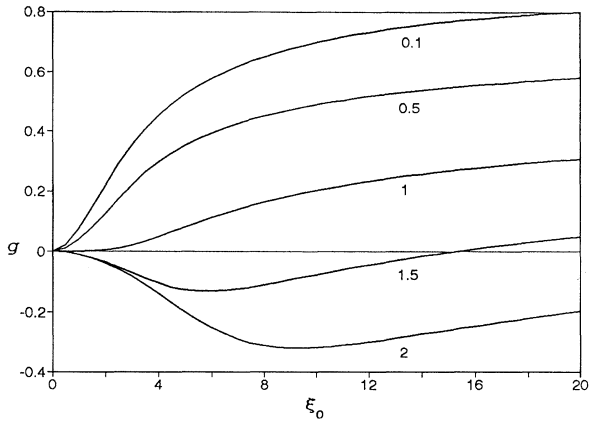

FIG. 3. Theoretical variations of $g(\xi_0, \omega\tau_B) = \eta_r(\xi_0, \omega\tau_B)/\eta_r(\infty, 0)$ for different values of $\omega\tau_B$ between 0.1 and 2.

Thus the rotational viscosity is $\Delta\eta = \frac{3}{2}\eta\varphi g$. According to Eqs. (1) and (9), the angular velocity of the particles may be written as $\boldsymbol{\omega}_p = (1 - g)\boldsymbol{\Omega}$. So $\Delta\eta$ can be presented in the form (2).

For arbitrary values of the field amplitude and frequency the problem is solved numerically. The procedure includes a numerical solution of the system of two differential equations (7) and (8) and a numerical computation of the integral over period in (9). For prescribed ξ_0 and $\omega\tau_B$ those equations are solved by the fourth-order Runge-Kutta method under the initial conditions $\xi_e = \xi_0$ and $F \sim 0$ at $t \sim 0$. The results of the computations are displayed in Fig. 3, which presents $g(\xi_0, \omega\tau_B) = \eta_r(\xi_0, \omega\tau_B)/\eta_r(\infty, 0)$ as a function of ξ_0 for different values of $\omega\tau_B$. These theoretical curves are very similar to experimental ones of Fig. 2, with, for the largest $\omega\tau_B$, negative values of g and its nonmonotonous variation as field increases. A more convenient representation is a plot of isolines g = const in the plane $(\xi_0, \omega\tau_B)$ as presented in Fig. 4. The isoline $g = 0$

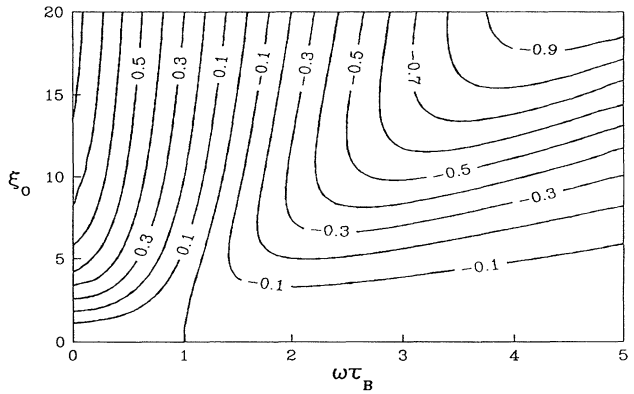

FIG. 4. Isolines of $g(\xi_0, \omega\tau_B)$ in the plane $(\xi_0, \omega\tau_B)$ for different values of g between +0.8 and −0.9.

parts the plane into two regions: $g > 0$ for $\omega\tau_B \leq 1$ and $g < 0$ for $\omega\tau_B \geq 1$. For a quantitative comparison, the experimental data are plotted in a similar representation in Fig. 5 using $\eta_r(\infty, 0) = 2.5, \xi_0 = M_S VH/kT = 5.25 \times 10^{-3} H$ (Oe) and $\omega\tau_B = 2\pi f \tau_B = 10^{-2} f$ (Hz). We observe a strong similarity between the experimental plots of Fig. 5 and those of Fig. 4, obtained with a single particle theory; in particular, in both cases g changes its sign for $\omega\tau_B \sim 1$.

Thus for $\omega\tau_B \geq 1$, the MF viscosity under an alternating magnetic field can become smaller than the MF viscosity in zero field. A decrease of 25% is observed experimentally. An interesting limit would be to reach an absolute zero in the MF viscosity; instabilities and flow recirculations would then develop spontaneously without any external flow. The marginal curve, theoretically predicted in [5], corresponds to a critical field $\xi_c = 126$ and a critical frequency $\omega_c\tau_B = 20$. Such values could be reached only with larger particles, for which both the colloidal stability and the Newtonian behavior would be difficult to preserve. Anyway, one of the basic results obtained with the present Newtonian MF is that its viscosity can be tuned between 220 and 50 cP by a field of 2000 Oe monitoring only the field frequency between 0 and 700 Hz. This attractive property could lead to a new generation of adaptative dampers.

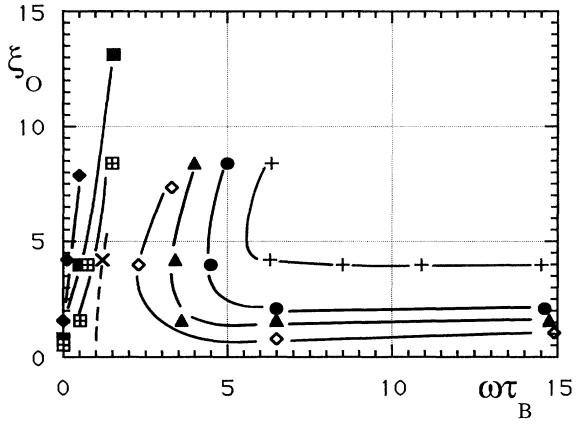

FIG. 5. Experimental isolines of g $(\xi_0, \omega\tau_B)$ in the plane $(\xi_0, \omega\tau_B)$. ◆: $g = 0.216$; ■: $g = 0.1$; ⊕: $g = 0.044$; ×: $g = 0$; ◇: $g = -0.036$; ▲: $g = -0.052$; ●: $g = -0.068$; and +: $g = -0.092$. Full and dotted lines are guides for the eye.

We are greatly indebted to Dr. S. Neveu for providing us with the MF sample and to J. Servais for his technical assistance. J.-C. B. is affiliated with the Université Paris 7. The Laboratoire d'Acoustique et Optique de la Matière Condensée is associated with the Centre National de la Recherche Scientifique.

[1] R. E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, New York, 1985).
[2] M. I. Shliomis, T. P. Lyubimova, and D. V. Luybimov, Chem. Eng. Commun. **67**, 275 (1988).
[3] R. E. Rosensweig, J. Popplewell, and R. J. Johnston, J. Magn. Magn. Mater. **85**, 171 (1990).
[4] J.-C. Bacri, A. Cebers, and R. Perzynski, Phys. Rev. Lett. **72**, 2705 (1994).
[5] M. I. Shliomis and K. I. Morozov, Phys. Fluids **6**, 2855 (1994).
[6] M. I. Shliomis, Sov. Phys. JETP **34**, 1291 (1972).
[7] J. P. McTague, J. Chem. Phys. **51**, 133 (1969).
[8] M. A. Martsenyuk, Yu. L. Raikher, and M. I. Shliomis, Sov. Phys. JETP **38**, 413 (1974).
[9] Yu. L. Raikher and M. I. Shliomis, Adv. Chem. Phys. Series **87**, 595 (1994).
[10] S. Neveu-Prin, F. A. Tourinho, J.-C. Bacri, and R. Perzynski, Colloids Surf. A **80**, 1 (1993).