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Ferrohydrodynamics: Testing a third magnetization equation

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A new magnetization equation recently derived from irreversible thermodynamics is employed to the calculation of an increase of ferrofluid viscosity in a magnetic field. Results of the calculations are compared with those obtained on the basis of two well-known magnetization equations. One of the two was obtained phenomenologically, another one was derived microscopically from the Fokker-Planck equation. It is shown that the third magnetization equation yields a quite satisfactory description of magnetiviscosity in the entire region of magnetic-field strength and the flow vorticity. This equation turns out to be valid—like the microscopically derived equation but unlike the former phenomenological equation—even far from equilibrium, and so it should be recommended for further applications.

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The conventional set of ferrohydrodynamic equations [1–3] consists of the equation of ferrofluid motion, the Maxwell equations, and the magnetization equation. The latter was first derived thirty years ago by the author:

$$\frac{d\mathbf{M}}{dt}=\mathbf{\Omega}\times\mathbf{M}-\frac{1}{\tau}(\mathbf{M}-\mathbf{M}_0)-\frac{1}{6\eta\phi}\mathbf{M}\times(\mathbf{M}\times\mathbf{H}) \quad (1)$$

(Ref. [1], Sh '72) Here $\mathbf{M}$ stands for the ferrofluid magnetization under the magnetic field $\mathbf{H}$ and the flow vorticity $2\mathbf{\Omega}=\nabla\times\mathbf{v}$, η is the fluid viscosity, $\phi=nV$ the volume fraction of magnetic grains in the liquid, n is their number density, V the volume of a single particle, and $\tau=3\eta V/k_BT$ is the Brownian time of rotational particle diffusion. When the fluid is at rest in a stationary magnetic field, its *equilibrium* magnetization $\mathbf{M}_0$ is described by the Langevin formula

$$\mathbf{M}_0=nmL(\xi)\frac{\boldsymbol{\xi}}{\xi},\quad \boldsymbol{\xi}=\frac{m\mathbf{H}}{k_BT},\quad L(\xi)=\coth\xi-\xi^{-1}, \quad (2)$$

where m is the magnetic moment of a single particle. The phenomenological Eq. (1) generalizes the Debye relaxation equation in case of *spinning* magnetic grains. The spin originates from viscous and magnetic torques acting upon the particles—see Eq. (6).

Soon after [1], Martsenyuk, *et al.* [4] (MRSh) proposed another magnetization equation derived microscopically from the Fokker-Planck equation. They have employed for the purpose an original *effective-field method* (EFM) described in detail in [5]. According to the method, an arbitrary nonequilibrium magnetization $\mathbf{M}$ is considered at any moment as an equilibrium one in a certain—specially prepared—effective field $\mathbf{H}_e$, that is

$$\mathbf{M}=nmL(\zeta)\boldsymbol{\zeta}/\zeta,\quad \boldsymbol{\zeta}=m\mathbf{H}_e/k_BT. \quad (3)$$

The magnetization (3) relaxes to its equilibrium value (2) as the effective field $\mathbf{H}_e$ (or $\boldsymbol{\zeta}$) approaches the true field $\mathbf{H}$ (or $\boldsymbol{\xi}$). This relaxation process is governed by the equation [4,6]

$$\frac{d\mathbf{M}}{dt}=\mathbf{\Omega}\times\mathbf{M}-\frac{[\zeta^2-(\boldsymbol{\xi}\cdot\boldsymbol{\zeta})]}{\tau\zeta^2}\mathbf{M}-\frac{[\zeta-L(\zeta)]}{6\eta\phi\zeta L^2(\zeta)}\mathbf{M}\times(\mathbf{M}\times\mathbf{H})\quad (\text{MRSh}). \quad (4)$$

Equations (3) and (4) determine the dependence $\mathbf{M}(t;\mathbf{H},\mathbf{\Omega})$ in an implicit form, where dimensionless effective field $\boldsymbol{\zeta}$ is the parameter.

It is well established that Eq. (4) describes very fine real ferrofluids. Comparison with computer simulation of the Brownian motion in orientational space [7,8] indicated that the EFM yields quite accurate results for any values of ξ and $\Omega\tau$. The same conclusion has been made in [6] by comparing the solutions of Eq. (4) with the results of numerical integration of the Fokker-Planck equation. At that time, the calculations [6–8] have shown that the phenomenological equation (1) is valid for any field magnitudes but only sufficiently small vorticities, $\Omega\tau<1$. The applicability of Eq. (1) in the case $\Omega\tau\ll1$ was corroborated by numerical [9] and analytical [10] solutions of the Fokker-Planck equation as well. Therefore, under consideration of a weakly nonequilibrium situation, one should give preference to Eq. (1) as it is far more simpler for analysis than Eq. (4). The latter, however, guarantees the correct description of magnetization even if its deviation from equilibrium value (2) is large, $\Omega\tau\gg1$, that is when Eq. (1) leads to erroneous results.

A new phenomenological magnetization equation derived quite recently [11] from irreversible thermodynamics is free from the above-mentioned shortcoming. This equation,

$$\frac{d\mathbf{H}_e}{dt}=\mathbf{\Omega}\times\mathbf{H}_e-\frac{1}{\tau}(\mathbf{H}_e-\mathbf{H})-\frac{1}{6\eta\phi}\mathbf{H}_e\times(\mathbf{M}\times\mathbf{H}) \quad (5)$$

(Ref. [11], Sh '01), coincides with Eq. (1) in the limit of low field, $\xi\ll1$, when the true magnetization and its equilibrium value can be written as $\mathbf{M}=\chi\mathbf{H}_e$ and $\mathbf{M}_0=\chi\mathbf{H}$, respectively; here $\chi=nm^2/3k_BT$ stands for the initial magnetic susceptibility. However, due to nonlinearity of the magnetization law (2)–(3), Eqs. (1) and (5) predict very different magnitudes of magnetization for large magnitudes of ξ. As it will be shown, Eq. (5) is valid even far from equilibrium.

As a check on applicability of the old (1) and the new (5) phenomenological equations we have chosen their predictions about the *rotational* or *spin viscosity* η_r of ferrofluids. Below we shall compare η_r obtained from Eqs. (1) and (5) with its almost exact value resulting from the EFM Eq. (4).

A ferrofluid flow in a magnetic field is accompanied with an intertwinement of hydrodynamic and magnetic interactions. Being magnetized, the fluid is subject to magnetic force and torque with the volume densities $(\mathbf{M}\cdot\nabla)\mathbf{H}$ and $\mathbf{M}\times\mathbf{H}$, respectively. On the other hand, the flow vorticity causes a change of magnetization. As seen from Eqs. (1), (4), and (5), $\mathbf{\Omega}$ impedes alignment of $\mathbf{M}$ with the direction of the local field $\mathbf{H}$. The appearing magnetic torque is equilibrated by the viscous torque,

$$6\eta\phi(\boldsymbol{\omega}-\mathbf{\Omega})=\mathbf{M}\times\mathbf{H}, \tag{6}$$

where $\boldsymbol{\omega}$ is a macroscopic (i.e., averaged over physically small ferrofluid volume) angular spin rate of magnetic grains. But any deviation of $\boldsymbol{\omega}$ from $\mathbf{\Omega}$ leads to an additional dissipation which is just manifested in rotational viscosity. This dissipation contributes to the stress tensor [1,2]:

$$\sigma_{ik}=-p\delta_{ik}+\eta\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}\right)+3\eta\phi\epsilon_{ikl}(\omega_l-\Omega_l)+\frac{1}{4\pi}\left(H_iB_k-\frac{1}{2}H^2\delta_{ik}\right); \tag{7}$$

here ϵ_{ikl} stands for an antisymmetric unit tensor and the last term represents the Maxwell tensor of magnetic stresses in ferrofluids. Eliminating $\boldsymbol{\omega}-\mathbf{\Omega}$ from Eq. (7) with the aid of Eq. (6), we obtain [1,2]

$$\sigma_{ik}=-\left(p+\frac{H^2}{8\pi}\right)\delta_{ik}+\eta\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}\right)+\frac{H_iB_k}{4\pi}+\frac{1}{2}(M_iH_k-M_kH_i). \tag{8}$$

On recognizing that $B_k=H_k+4\pi M_k$, the stress tensor (8) takes an evidently *symmetric* form:

$$\sigma_{ik}=-\left(p+\frac{H^2}{8\pi}\right)\delta_{ik}+\eta\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}\right)+\frac{H_iH_k}{4\pi}+\frac{1}{2}(M_iH_k+M_kH_i),$$

as it must be in our approximation. Actually, the exact result reads as $\sigma_{ki}-\sigma_{ik}=I\epsilon_{ikl}(d\omega_l/dt)$ where $I=\rho_s d^2\phi/10$ is the volume density of the particle moment of inertia (ρ_s the particle density and d their diameter). However, taking into account an extreme smallness of I for the particles with $d\sim10$ nm, we have not inserted the inertia term $I(d\boldsymbol{\omega}/dt)$ in Eq. (6).

The boundary wall streamlined by the fluid is acted by the force $f_i=[\sigma_{ik}]n_k$ on unit area; here [ ] denotes difference evaluated across the fluid-solid interface and $\mathbf{n}$ is the normal to the interface. The friction (tangential) force exerted on the wall is $f_\tau=[\sigma_{\tau n}]$. By using the electrodynamic boundary conditions, $[H_\tau]=0$ and $[B_n]=0$, we get from Eq. (8)

$$f_\tau=\eta(\partial v_\tau/\partial x_n)+(M_\tau H_n-M_nH_\tau)/2. \tag{9}$$

For the Poiseuille or Couette flow, $\mathbf{v}=[0,v(x),0]$, in a transversal magnetic field, $\mathbf{H}=(H,0,0)$, magnetization has two components: $\mathbf{M}=(M_x,M_y,0)$. Then Eq. (9) can be written in the form $f_\tau=2(\eta+\eta_r)\Omega$ where $2\Omega=\partial v/\partial x$ and rotational viscosity is defined as

$$\eta_r=M_yH/4\Omega. \tag{10}$$

Thus, the additional viscosity is expressed through the off-axis component of magnetization, M_y. For small $\Omega\tau$ this component is also small; according to the all three of the magnetization equations cited above, $M_y\propto\Omega\tau$. So, η_r does not depend on the flow vorticity in the limit $\Omega\tau\ll1$. However, for finite values of $\Omega\tau$ the viscosity does depend on Ω. As a result, the function $\sigma_{\tau n}(\Omega)$ deviates from the linear one, i.e., a ferrofluid acquires non-Newtonian properties.

Proceeding to the calculation of rotational viscosity on the basis of Eq. (5), it is convenient to pass from the fields $\mathbf{H}$ and $\mathbf{H}_e$ to their nondimensional values $\boldsymbol{\xi}$ and $\boldsymbol{\zeta}$:

$$\frac{d\boldsymbol{\zeta}}{dt}=\mathbf{\Omega}\times\boldsymbol{\zeta}-\frac{1}{\tau}(\boldsymbol{\zeta}-\boldsymbol{\xi})-\frac{L(\zeta)}{2\tau\zeta}\boldsymbol{\zeta}\times(\boldsymbol{\zeta}\times\boldsymbol{\xi}). \tag{11}$$

At the stated above arrangement of the applied magnetic field with respect to the fluid flow, Eq. (11) admits a steady solution in which the effective field $\boldsymbol{\zeta}$ tracks the true field $\boldsymbol{\xi}$ with lag angle α, i.e., $\boldsymbol{\zeta}=(\zeta\cos\alpha,\zeta\sin\alpha,0)$. The dependence of ζ and α upon ξ and $\Omega\tau$ is given by

$$\sqrt{\xi^2-\zeta^2}=\frac{2\Omega\tau\zeta}{2+\zeta L(\zeta)},\quad \cos\alpha=\frac{\zeta}{\xi}. \tag{12}$$

Substituting $M_y=nmL(\zeta)\sin\alpha$ in (10) and using (12), we obtain

$$\eta_r=\frac{3}{2}\eta\phi\frac{\zeta L(\zeta)}{2+\zeta L(\zeta)}\quad(\text{Ref. [11], Sh}'01). \tag{13}$$

In the same way we find from Eq. (4)

$$\sqrt{\xi^2-\zeta^2}=\frac{2\Omega\tau\zeta L(\zeta)}{\zeta-L(\zeta)},\quad \cos\alpha=\frac{\zeta}{\xi}, \tag{14}$$

that results in

$$\eta_r=\frac{3}{2}\eta\phi\frac{\zeta L^2(\zeta)}{\zeta-L(\zeta)}\quad(\text{MRSh}). \tag{15}$$

The solution of Eq. (1) can be presented in a similar form. Let us introduce a new variable $\boldsymbol{\zeta}$ instead of $\mathbf{M}$ by the relation $\mathbf{M}=M_0(\boldsymbol{\zeta}/\xi)$ where $M_0=nmL(\xi)$. It is worth noting that ζ is no more an effective field unlike ζ in preceding

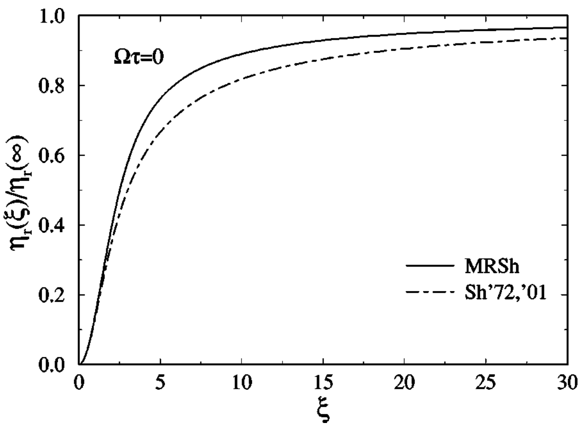

FIG. 1. Dependence of the reduced rotational viscosity on the dimensionless field strength given by Eq. (19) MRSh and by Eq. (18) (Refs. [1,11], Sh′72 and Sh′01).

relationships (12)–(15). By substituting the components $M_x = M_0(\zeta/\xi)\cos\alpha$ and $M_y = M_0(\zeta/\xi)\sin\alpha$ in Eq. (1), we get $\cos\alpha = M/M_0 = \zeta/\xi$ and

$$\sqrt{\xi^2-\zeta^2} = \frac{2\Omega\tau\xi\zeta}{2\xi+\zeta^2 L(\xi)}. \tag{16}$$

This and the definition (10) yield

$$\eta_r = \frac{3}{2}\eta\phi\frac{\zeta^2 L(\xi)}{2\xi+\zeta^2 L(\xi)}. \quad (\text{Ref. [1], Sh}'72) \tag{17}$$

Owing to the smallness of magnetic grains, the Brownian relaxation time τ does not exceed 10^{-4} s even in high-viscous ferrofluids. Hence the inequality $\Omega\tau \ll 1$ is usually satisfied. Then, on neglecting the value $\Omega\tau$ in Eqs. (12), (14), and (16), all three of these relationships are reduced to $\zeta = \xi$. Eliminating now ζ from Eqs. (13) and (17), we see that both the old (1) and the new (5) phenomenological equations predict *the same* dependence of rotational viscosity on the magnetic-field strength:

$$\eta_r(\xi) = \frac{3}{2}\eta\phi\frac{\xi L(\xi)}{2+\xi L(\xi)} = \frac{3}{2}\eta\phi\frac{\xi-\tanh\xi}{\xi+\tanh\xi}. \tag{18}$$

The EFM magnetization equation (4) yields a somewhat different result. Setting $\zeta = \xi$ in Eq. (15) gives

$$\eta_r(\xi) = \frac{3}{2}\eta\phi\frac{\xi L^2(\xi)}{\xi - L(\xi)} \quad (\text{MRSh}). \tag{19}$$

The viscosities (18) and (19) are compared in Fig. 1. Both of them approach the saturation value $\eta_r(\infty) = 3\eta\phi/2$ at $\xi \gg 1$. In the figure we plot the reduced rotational viscosity $\eta_r(\xi)/\eta_r(\infty)$ as a function of ξ. The upper curve calculated by the EFM [4] represents a very good approximation. Actually, as shown in [5,10], it hardly differs from the exact solution of the linearized Fokker-Plank equation. Both the phenomenological equations, (1) and (5), result in the lower curve in Fig. 1 that is described by the Shliomis' formula (18). This function agrees with Eq. (19) in the low- and high-field limits and deviates from it, at most, by 15% in the entire range of the argument ξ.

When the ferrofluid is subjected to viscous shear, the magnetic grains tend to be rotated out of alignment with the magnetic field. Thus the flow with a sufficiently large shear rate, $\Omega\tau \geq 1$, induces—along with the Brownian motion—a quotient *demagnetization*. Formally, this effect originates from decreasing the parameter ζ determined by Eqs. (12), (14), and (16). According to these equations, $\zeta = \xi$ when

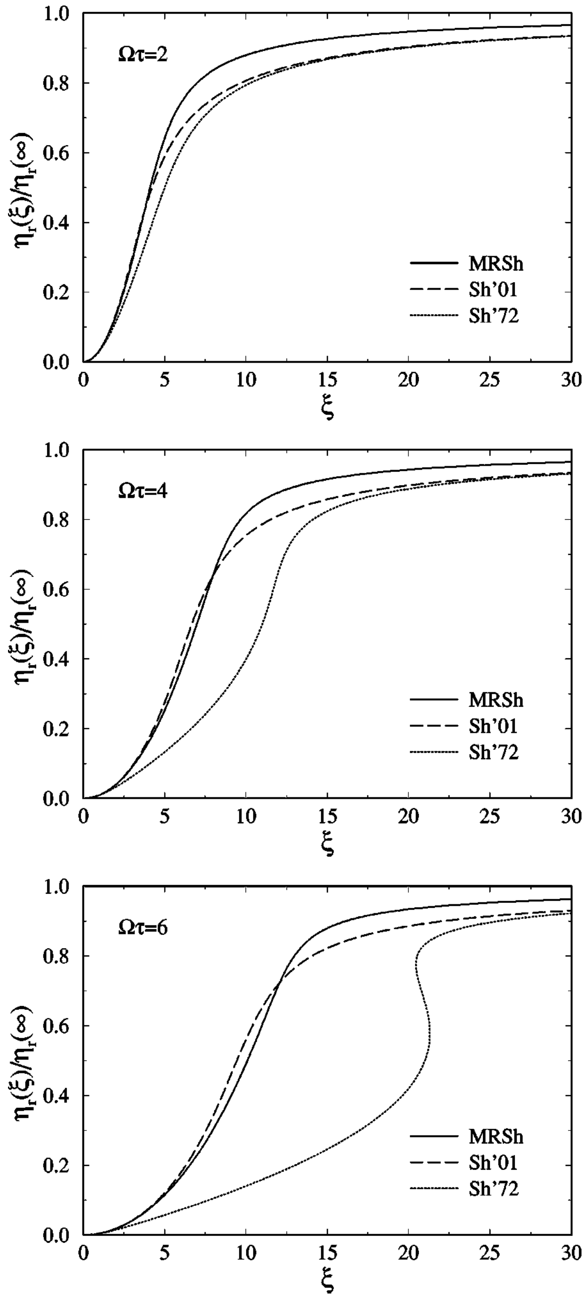

FIG. 2. Dependence of the rotational viscosity on the field for some values of the shear rate $\Omega\tau$, as calculated from the EFM [Eqs. (14)–(15), MRSh], and by the new [Eqs. (12)–(13), Ref. [11], Sh′01] and old [Eqs. (16)–(17), Ref. [1], Sh′72] phenomenological approaches.

$\Omega\tau = 0$ but the more there is of $\Omega\tau$ at constant ξ, the less there is of ζ. The reduction of the magnetization leads in turn to some decrease in the rotational viscosity. This decrease, imperceptible in practice up to $\Omega\tau \simeq 1$, then becomes very significant. Figure 2 illustrates the dependence of the viscosity increase on the magnetic-field strength for three values of the product $\Omega\tau$. Interestingly, under the finite shear rate the viscosities given by Eqs. (13) and (17) do not coincide with each other any more. As seen from the plot, the higher the shear the more discrepancy between viscosity values predicted by the new and the old phenomenological equations. At high shear in a high field, Eq. (1) predicts a *hysteresis* of viscosity, which however is corroborated neither by direct calculations of [6–8] or by the solution (14)–(15) of the EFM equation (4). Our new Eq. (5) also does not predict such a hysteresis but it provides us with a quite satisfactory viscosity description in a wide region of parameters ξ and $\Omega\tau$. Indeed, in this entire region the solutions of Eqs. (4) and (5) agree closely, as shown in Fig. 2. Thus, Eq. (5) can be recommended for an employment on the same level with Eq. (4). It is worth noting that all the above calculations, carried out for a shear flow, apply equally to a rigid rotation of a ferrofluid with an angular velocity $\mathbf{\Omega}$ in a constant transversal magnetic field, $\mathbf{H} \perp \mathbf{\Omega}$, and to a quiescent ferrofluid subjected to a uniform rotating field $\mathbf{H} = (H\cos\Omega t, H\sin\Omega t, 0)$ as well.

The difference between discussed magnetization equations is also manifested at the relaxation from an equilibrium magnetization in a quiescent ferrofluid after the field is suddenly switched off. Then the fluid remains at rest, $\mathbf{\Omega} = 0$, so $\mathbf{M}$ and $\mathbf{H}_e$ are always parallel to $\mathbf{H}$. Hence Eqs. (1), (4), and (5) are reduced to

$$dM/dt = -(M - M_0)/\tau, \tag{20a}$$

$$dM/dt = -(1 - H/H_e)M/\tau, \tag{20b}$$

$$dH_e/dt = -(H_e - H)/\tau, \tag{20c}$$

respectively. In Fig. 3 we plot the decay of reduced magnetization $M(t)/M_0$ according to Eqs. (20) with $M_0 = nmL(\xi)$ for some initial field magnitudes $\xi = mH/k_BT$. As the true field H is switched off at the moment $t = 0$, Eqs. (20a) and (20b) coincide with each other at $t > 0$, when their solution reads

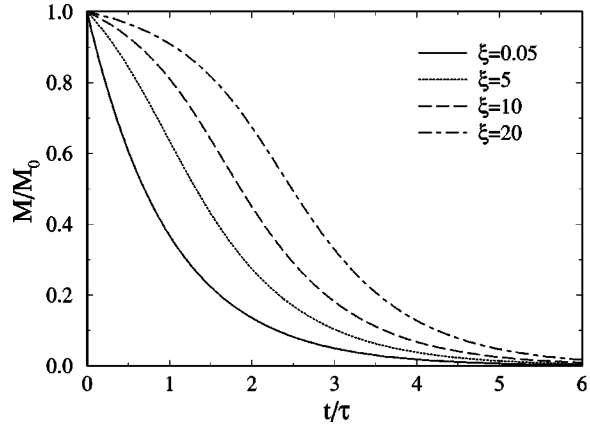

FIG. 3. Time dependence of the reduced magnetization $M(t)/M_0$ after the magnetic field ξ is switched off, as described by Eq. (22). The lowest curve also represents the solution (21) for any ξ.

$$M(t)/M_0 = \exp(-t/\tau), \tag{21}$$

i.e., it does not depend on ξ. Equation (20c) has the solution $H_e(t) = H\exp(-t/\tau)$, so that we obtain

$$M(t)/M_0 = L(\xi e^{-t/\tau})/L(\xi). \tag{22}$$

The last decay predicted by the new magnetization equation (5) is exponential only in the limit $\xi \ll 1$, while the EFM equation (4) together with the old phenomenological equation (1) predict the exponential decay of magnetization for any values of ξ. This difference in relaxation behavior side by side with the difference in the ferrofluid viscosity can be of relevance for testing the magnetization equations and the interpretation of corresponding experiments.

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