Magnetoviscosity in suspensions of grains with finite magnetic anisotropy

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(Received 2 February 2006; published 29 June 2006)

Coupling between magnetic and mechanical rotational degrees of freedom of fine ferromagnetic grains is provided by the energy of their magnetic anisotropy. In the limiting case of strong anisotropy, an applied stationary magnetic field induces the greatest obstacles to the "rigid dipole" spin in a vortex ferrofluid flow, while in the opposite ideal case, the "soft dipoles" twist freely with the liquid. As a result, the field-dependent part of the ferrofluids viscosity depends not only on the external magnetic field strength but also on the particle magnetic anisotropy. An explicit expression coming from simple physical arguments and describing both these dependencies of magnetoviscosity is derived and discussed.

DOI: 10.1103/PhysRevE.73.066312 PACS number(s): 47.65.-d

I. INTRODUCTION

The magnetic moment $\mathbf{m}$ of a small ferromagnetic grain is coupled with the particle body due to the energy of magnetic anisotropy KV, where K is the energy density and V is the particle volume. This coupling depends on the dimensionless ratio $\sigma=KV/k_BT$. At a high value of σ, the vector $\mathbf{m}=m\mathbf{e}$ is aligned strictly along the axis $\mathbf{n}$ of easy magnetization ($\mathbf{e}$ and $\mathbf{n}$ are unit vectors). In the limit $\sigma\gg 1$, the particle represents a rigid magnetic dipole ($\mathbf{e}=\mathbf{n}$): Any change of its orientation is possible only by Brownian rotation of the particle itself.

For a finite σ, the vector $\mathbf{m}$ is only partly frozen into the particle, thus it can turn within the particle body. For $\sigma\sim 1$, the magnetic moment effectively fluctuates around the particle easy axis—these fluctuations were predicted and described for the first time by Néel [1].

An externally imposed magnetic field, $\mathbf{H}$, tends to align the particle magnetic moment $\mathbf{m}$ along the vector $\mathbf{H}$. As the moment is coupled—to the extent of σ—with the particle body, the field $\mathbf{H}$ impedes free particle rotation with the flow vorticity $\mathbf{\Omega}=(\nabla\times\mathbf{v})/2$. Thus, there appears to be some difference between $\mathbf{\Omega}$ and the mean angular velocity of particle rotation $\boldsymbol{\omega}$. This lag induces viscous forces to act upon magnetic grains which leads to the additional dissipation of the ferrofluid kinetic energy, and is manifested in an additional *rotational* viscosity η_R. The latter is a growing function of σ and the dimensionless magnetic field strength $\xi=mH/k_BT$. For the limiting case of rigid magnetic dipoles ($\sigma=\infty$), the function $\eta_R(\xi,\sigma)$ was first obtained phenomenologically [2]:

$$\eta_R(\xi,\infty)=\frac{3}{2}\eta\phi\frac{\xi L(\xi)}{2+\xi L(\xi)}=\frac{3}{2}\eta\phi\frac{\xi-\tanh\xi}{\xi+\tanh\xi}. \tag{1}$$

Here, η is the ferrofluid viscosity in the absence of an imposed magnetic field, $\phi=nV$ is the volume fraction of suspended magnetic grains, and $L(\xi)$ is the Langevin function. Expression (1) is written under the assumption of orthogonality between $\mathbf{H}$ and $\mathbf{\Omega}$ (e.g., for planar Couette flow in the field directed across the confining solid surfaces or along the flow, and for Poiseuille pipe flow in an axial magnetic field). For arbitrary orientation of magnetic field, the right-hand side of Eq. (1) should be multiplied by $\sin^2\alpha$, where α is the angle between vectors $\mathbf{H}$ and $\mathbf{\Omega}$.

Soon after [2], Martsenyuk *et al.* [3] found for $\eta_R(\xi,\infty)$ another relation, derived microscopically from the Fokker-Planck kinetic equation by the *effective-field method*:

$$\eta_R(\xi,\infty)=\frac{3}{2}\eta\phi\frac{\xi L^2(\xi)}{\xi-L(\xi)}. \tag{2}$$

Though functions (1) and (2) do not appear alike, they agree fairly well in the entire range of the Langevin's parameter ξ – see two upper curves in Fig. 1. Indeed, both expressions yield the same magnitudes of η_R in low fields as well as at saturation:

$$\eta_R=\eta\phi\xi^2/4 \quad \text{at } \xi\ll 1, \tag{3}$$

$$\eta_R^{\text{sat}}\equiv\eta_R(\infty,\infty)=(3/2)\eta\phi \quad \text{at } \xi\to\infty. \tag{4}$$

The latter *doubly limiting* value is never reached for a finite σ. The first calculation, taking into account the finite value of magnetic anisotropy, was performed by Raikher and Shliomis [4] for the limiting case of strong magnetic field. Later authors used the Fokker-Planck kinetic rotary diffusion equation, also called the Smoluchowski equation, to find the orientational distribution function (i.e., the probability density function) $W(t)$ that determines statistical properties of an assembly of magnetic grains.

In the general case of the grains with a finite magnetic anisotropy, the function $W(\mathbf{e},\mathbf{n},t)$ is defined in the configuration space representing the direct product of two vector spaces, $\mathbf{e}\otimes\mathbf{n}$. A number of formalisms was developed to ob-

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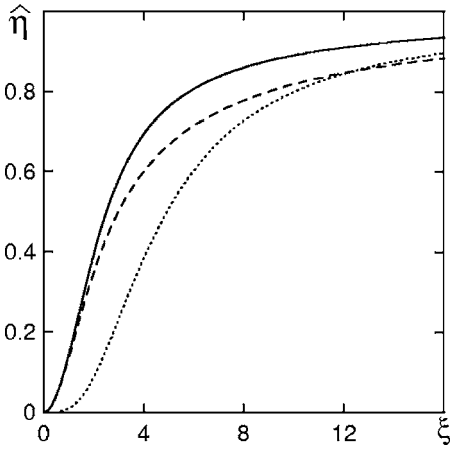

FIG. 1. Field dependence of the reduced viscosity $\hat{\eta}=\eta_R/\eta_R^{\mathrm{sat}}$ for a suspension of magnetically hard grains ($\sigma=\infty$) computed by: The effective-field method, Eq. (2),—solid line, the phenomenological expression (1)—dashed line, and Eq. (9) following from the Cebers-Stepanov expression (5)—dotted line.

tain the kinetic equations, and different techniques were used to solve them in various limiting cases, especially in the rigid-dipole limit [3], as well as in more general conditions [5–10]. Cebers [8] derived a very cumbrous expression giving the dependence of the rotational viscosity on ξ and σ in the range $\xi>\sigma$. Stepanov [9] reobtained recently the Cebers' result. He presented it in the following elegant form:

$$\eta_R(\xi,\sigma)=\eta_R^{\mathrm{sat}}\frac{35L_2^2(\xi)F_2^2(\sigma)}{14+5L_2(\xi)F_2(\sigma)+16L_4(\xi)F_4(\sigma)}, \quad (5)$$

and determined more precisely the region of its applicability: $\xi>2\sigma$. In Eq. (5), the functions of ξ are defined by the recurrent relationships

$$L_0=1, \quad L_1\equiv L(\xi)=\coth\xi-1/\xi,$$

$$L_{n-1}-L_{n+1}=(2n+1)L_n/\xi, \quad (6)$$

and the functions of σ are

$$F_2=\frac{3}{2}\left(\frac{R'}{R}-\frac{1}{3}\right), \quad F_4=\frac{1}{8}\left(3-30\frac{R'}{R}+35\frac{R''}{R}\right), \quad (7)$$

where

$$R(\sigma)=\int_0^1 e^{\sigma x^2}dx, \quad R'\equiv\frac{dR}{d\sigma}. \quad (8)$$

Note that the configurational integral $R(\sigma)$ is closely related to the error function $\mathrm{erf}(x)$ [11]:

$$R(\sigma)=\frac{\sqrt{\pi}}{2}\frac{\mathrm{erf}(i\sqrt{\sigma})}{i\sqrt{\sigma}}, \quad \mathrm{erf}(x)=\frac{2}{\sqrt{\pi}}\int_0^x e^{-t^2}\mathrm{d}t,$$

and the functions $F_n(\sigma)$ are none other than the Legendre polynomials $P_n(x)$ averaged with the distribution function $W(x)=\exp(\sigma x^2)/R(\sigma)$, i.e., $F_n(\sigma)=\langle P_n(x)\rangle$.

Both kinds of functions entering symmetrically into Eq. (5)—$L_n(\xi)$ and $F_n(\sigma)$—tend to unity when their argument tends to infinity, so that η_R reaches in this double limit its true saturation value (4). However, the approximation of a local equilibrium used by Cebers [8] and Stepanov [9] is justified only in a sufficiently strong magnetic field, $\xi>2\sigma$, thus Eq. (5) fails in the limiting case of rigid dipoles, $\sigma=\infty$, when it reduces to

$$\eta_R(\xi,\infty)=\eta_R^{\mathrm{sat}}\frac{35L_2^2(\xi)}{14+5L_2(\xi)+16L_4(\xi)}. \quad (9)$$

Indeed, this dependence $\eta_R(\xi)$, shown in Fig. 1 by the dotted line, differs significantly from that given by Eqs. (1) or (2). The discrepancy is especially large in the region of low magnetic field strength—just as expected. Note, that expression (5) is given (in the Cebers' notation) in the book [10] without any mention of the limited range of its applicability—see Eq. (5.10) on p. 245. On the same page in Ref. [10], Fig. 5.1 demonstrates the ξ-dependence of the rotational viscosity in the interval $0\leqslant\xi\leqslant15$ for $\sigma=4$, 9, and 25. This figure misleads the readers because both of the last σ values do not satisfy the condition $\xi>2\sigma$ within the entire interval of ξ presented in the figure.

To conclude this Introduction, an expression which describes the rotational viscosity of ferrofluids, over wide regions of applied magnetic and anisotropy fields, has not yet been found. Multiple attempts to derive such a formula *microscopically* have not yet been successful. A *phenomenological* expression for the viscosity has also not been found. There is even an opinion that, for the case of moderate values of σ, one cannot obtain the expression *macroscopically* in a reasonable way. This present work has the objective to fill this gap to obtain an appropriate formula for $\eta_R(\xi,\sigma)$ proceeding from simple physical arguments.

II. ROTATIONAL VISCOSITY

In an external field $\mathbf{H}=H\mathbf{h}$, $\mathbf{h}^2=1$, the energy of a magnetically uniaxial particle is [12]

$$U(\mathbf{e},\mathbf{n})=-mH(\mathbf{e}\cdot\mathbf{h})-KV(\mathbf{e}\cdot\mathbf{n})^2. \quad (10)$$

The derivatives of the energy with respect to $\mathbf{m}=m\mathbf{e}$ and $\mathbf{n}$ determine the net (total) magnetic field $\mathbf{H}_\Sigma$ acting upon the particle magnetic moment, and the magnetic torque $\boldsymbol{\mu}$ acting directly upon the particle body:

$$\mathbf{H}_\Sigma=-\frac{1}{m}\frac{\partial U}{\partial\mathbf{e}}=H\mathbf{h}+\frac{2KV}{m}(\mathbf{e}\cdot\mathbf{n})\mathbf{n}, \quad (11)$$

$$\boldsymbol{\mu}=-\mathbf{n}\times\partial U/\partial\mathbf{n}=-2KV(\mathbf{e}\cdot\mathbf{n})(\mathbf{e}\times\mathbf{n}). \quad (12)$$

At equilibrium in a constant field, the magnetic moment is parallel to $\mathbf{H}_\Sigma$, i.e., $\mathbf{m}\times\mathbf{H}_\Sigma=\mathbf{0}$, and the external torque is absent, $\boldsymbol{\mu}=\mathbf{0}$. For our subsequent calculations, it is important that the relaxation times of the magnetic and rotary mechanical degrees of freedom differ greatly. In fact, the solid-body relaxation time of the particle magnetic moment $\mathbf{m}$ to align with the direction of the net magnetic field $\mathbf{H}_\Sigma$ is $\tau_s=M_b/(2\alpha\gamma K)$ [13], where M_b is the bulk magnetization of the dispersed ferromagnet, α is a dimensionless attenuation

parameter entering into the Landau-Lifshitz magnetodynamic equation, and γ is the gyromagnetic ratio. For typical values of $M_b=480$ G (magnetite), $\alpha=4\times10^{-2}$, $\gamma=2\times10^{7}$ s^{-1} Oe^{-1}, and $K=3\times10^{5}$ erg/cm^3, one obtains $\tau_s=10^{-9}$ s. This is three to five orders of magnitude smaller than the Brownian rotary diffusion time $\tau_B=3\eta V/k_BT$—which characterizes the rate of the particle reorientation.

Thus, the equilibrium occurs in two steps: First, the magnetic moment rapidly—during the time of the order τ_s—settles along $\mathbf{H}_\Sigma$; second, the particle axis $\mathbf{n}$ rotates slowly (for the time $\sim\tau_B$) to the direction of $\mathbf{H}$. As seen from Eq. (11), a change of $\mathbf{n}$ causes some change of $\mathbf{H}_\Sigma$, which, in turn, deviates the vector $\mathbf{e}$ from its equilibrium. However, owing to the adiabatic condition $\tau_s\ll\tau_B$, perturbations of $\mathbf{e}$ decay rapidly, so in the process of orientational relaxation this vector goes through a sequence of quasi-equilibrium states determined by the condition $\mathbf{m}\times\mathbf{H}_\Sigma=\mathbf{0}$, i.e.,

$$mH(\mathbf{e}\times\mathbf{h})+2KV(\mathbf{e}\cdot\mathbf{n})(\mathbf{e}\times\mathbf{n})=\mathbf{0}. \tag{13}$$

Microscopic relationships (11)–(13) still fail to give a satisfactory description of the particle magnetic moments and easy axis' orientations, since they are ignoring orientational fluctuations of the unit vectors $\mathbf{e}$ and $\mathbf{n}$. In order to pass to macroscopic values, one needs to average Eqs. (11)–(13) with an appropriate orientational distribution function W dependent on $\mathbf{e}$, $\mathbf{n}$, ξ, σ and the flow vorticity $\mathbf{\Omega}=(\nabla\times\mathbf{v})/2$. In equilibrium, $\mathbf{\Omega}=\mathbf{0}$, the function W reduces to the Gibbs distribution $W_0=Z_0^{-1}\exp\{-U/k_BT\}$ with U from Eq. (10):

$$W_0=\frac{\exp\{\xi(\mathbf{e}\cdot\mathbf{h})+\sigma(\mathbf{e}\cdot\mathbf{n})^2\}}{16\pi^2R(\sigma)(\sinh\xi)/\xi}. \tag{14}$$

The equilibrium partition integral Z_0 in the denominator of this expression is calculated in the Appendix. Averages with W and W_0 are denoted below as $\langle\cdots\rangle$ and $\langle\cdots\rangle_0$, respectively.

Averaging Eqs. (12) and (13) gives

$$\langle\boldsymbol{\mu}\rangle=-2KV\langle(\mathbf{e}\cdot\mathbf{n})(\mathbf{e}\times\mathbf{n})\rangle, \tag{15}$$

$$mH(\langle\mathbf{e}\rangle\times\mathbf{h})+2KV\langle(\mathbf{e}\cdot\mathbf{n})(\mathbf{e}\times\mathbf{n})\rangle=\mathbf{0}. \tag{16}$$

Combining these equations yields an alternative expression for the magnetic torque: $\langle\boldsymbol{\mu}\rangle=mH(\langle\mathbf{e}\rangle\times\mathbf{h})$, thus the *torque-balance equation* takes its ordinary form [13,14]:

$$6\eta V(\langle\boldsymbol{\omega}\rangle-\mathbf{\Omega})=mH(\langle\mathbf{e}\rangle\times\mathbf{h}). \tag{17}$$

The torque $\langle\boldsymbol{\mu}\rangle$ impedes free rotation of magnetic grains by maintaining some difference between their angular velocity $\langle\boldsymbol{\omega}\rangle$ and the fluid vorticity $\mathbf{\Omega}$. In turn, the viscous torque acting upon the grain—see the left-hand side in Eq. (17)—induces some lag of $\mathbf{e}$ behind $\mathbf{h}$, thereby providing the very existence of the magnetic torque. On the other hand, the viscous torque causes an additional energy dissipation which is just manifested in the rotational viscosity η_R; the latter may be presented in the form [14]

$$\eta_R=\frac{3}{2}\eta\phi\frac{\Omega-\langle\omega\rangle}{\Omega}, \tag{18}$$

so the problem comes to the determination of the relation between the averaged frequency of the particle rotation and the local vorticity of the carrier liquid.

The averaging in Eq. (16) should take into account that the orientation of the particle magnetic moment is characterized by the unit vector $\mathbf{e}$, while orientation of the particle easy axis is described by the symmetric traceless tensor

$$s_{ik}=\tfrac{3}{2}\left(n_in_k-\tfrac{1}{3}\delta_{ik}\right).$$

The difference in parity reflects the double directedness of the axis of magnetic anisotropy: Neither tensor s_{ik} nor any other expression having a physical meaning change upon replacement of $\mathbf{n}$ by $-\mathbf{n}$. [This is just the reason why the contraction $\mathbf{e}\cdot\mathbf{n}$ in Eq. (10) is squared.]

Let us introduce a vector, $\mathbf{b}$, representing the contraction of the vector $\mathbf{e}$ with the tensor s_{ik}:

$$b_i=s_{ik}e_k,\quad \mathbf{b}=\tfrac{3}{2}(\mathbf{e}\cdot\mathbf{n})\mathbf{n}-\tfrac{1}{2}\mathbf{e}. \tag{19}$$

This vector determines the direction of the field of anisotropy which is given by the last term in Eq. (11):

$$\mathbf{H}_A=\frac{2KV}{m}(\mathbf{e}\cdot\mathbf{n})\mathbf{n}.$$

With the aid of definition (19), one can rewrite $\mathbf{H}_A$ as

$$\mathbf{H}_A=\frac{4KV}{3m}\mathbf{b}.$$

These last two expressions differ from each other by the $\mathbf{e}$-dependent term $(2KV/3m)\mathbf{e}$. Therefore, the transition from one of the two to another one means the insertion of an isotropic—and then unimportant [12]—addition $KV/3$ into the energy U of magnetic grains (10).

Dividing Eq. (16) by k_BT and using the definition (19) gives

$$\xi\langle\mathbf{e}\rangle\times\mathbf{h}=\tfrac{4}{3}\sigma\langle\mathbf{b}\times\mathbf{e}\rangle. \tag{20}$$

This equation relates the averages of the components e_i of vector $\mathbf{e}$ and the tensor e_ie_k composed of the components of this vector. To decouple the second moment, let us apply the following well known approximation [15] typical of mean field theory.

The identical transformation of $\mathbf{b}$ and $\mathbf{e}$ vectors,

$$\mathbf{b}=\langle\mathbf{b}\rangle+(\mathbf{b}-\langle\mathbf{b}\rangle),\quad \mathbf{e}=\langle\mathbf{e}\rangle+(\mathbf{e}-\langle\mathbf{e}\rangle),$$

leads to

$$\mathbf{b}\times\mathbf{e}=\langle\mathbf{b}\rangle\times\langle\mathbf{e}\rangle+\langle\mathbf{b}\rangle\times\Delta\mathbf{e}-\langle\mathbf{e}\rangle\times\Delta\mathbf{b}+\Delta\mathbf{b}\times\Delta\mathbf{e},$$

where $\Delta\mathbf{b}=\mathbf{b}-\langle\mathbf{b}\rangle$ and $\Delta\mathbf{e}=\mathbf{e}-\langle\mathbf{e}\rangle$ are fluctuations whose averages identically equal zero. Thus the average of the last expression is

$$\langle\mathbf{b}\times\mathbf{e}\rangle=\langle\mathbf{b}\rangle\times\langle\mathbf{e}\rangle+\langle\Delta\mathbf{b}\times\Delta\mathbf{e}\rangle. \tag{21}$$

Neglecting here the square of fluctuations, we pass from Eq. (20) to

$$3\xi\langle\mathbf{e}\rangle\times\mathbf{h}=-4\sigma\langle\mathbf{e}\rangle\times\langle\mathbf{b}\rangle. \tag{22}$$

In equilibrium, when magnetic grains are on the average at rest ($\langle\omega\rangle=0$) in a quiescent ferrofluid ($\mathbf{\Omega}=0$), the particle orientational distribution function reduces to (14) and we find (see the Appendix)

$$\langle\mathbf{e}\rangle_0=L(\xi)\mathbf{h},\quad \langle\mathbf{b}\rangle_0=F_2(\sigma)L(\xi)\mathbf{h}, \tag{23}$$

so that Eqs. (17) and (22) are identically satisfied. It is clear that in a moving ferrofluid the values $\langle\mathbf{e}\rangle$ and $\langle\mathbf{b}\rangle$ deviate from $\langle\mathbf{e}\rangle_0$ and $\langle\mathbf{b}\rangle_0$ to the extent of $\langle\omega\rangle$. To find these disturbed values, one must add to Eqs. (17) and (22) one more equation which would link $\langle\mathbf{b}\rangle$ and $\langle\omega\rangle$ with each other.

Such an equation can be obtained in the following way. Consider an element of ferrofluid volume from two frames of reference: a local one Σ' rotating with the angular velocity $\langle\omega\rangle$ and a fixed one Σ. It is reasonable to assume that in the system Σ' where the mean angular velocity of magnetic grains equals zero, small deviations of $\langle\mathbf{b}\rangle$ from its equilibrium value $\langle\mathbf{b}\rangle_0$ decay by the simple relaxation law

$$\frac{d'\langle\mathbf{b}\rangle}{dt}=-\frac{\langle\mathbf{b}\rangle-\langle\mathbf{b}\rangle_0}{\tau_B}, \tag{24}$$

where d'/dt means the derivative in the rotating system Σ' and $\tau_B=3\eta V/k_BT$ is the previously defined Brownian time of rotary particle diffusion. Using now Eq. (24) and the well-known kinematic relation

$$\frac{d\langle\mathbf{b}\rangle}{dt}=\langle\boldsymbol{\omega}\rangle\times\langle\mathbf{b}\rangle+\frac{d'\langle\mathbf{b}\rangle}{dt},$$

linking the rates of change of a vector in the frames of reference Σ and Σ', we obtain the equation

$$\frac{d\langle\mathbf{b}\rangle}{dt}=\langle\boldsymbol{\omega}\rangle\times\langle\mathbf{b}\rangle-\frac{1}{\tau_B}(\langle\mathbf{b}\rangle-\langle\mathbf{b}\rangle_0). \tag{25}$$

For a single grain of the diameter $d=10$ nm, the Brownian diffusion time $\tau_B=\pi\eta d^3/2k_BT$ is less than 10^{-6} s in water ($\eta=10^{-2}$ Ps) and does not exceed 10^{-3} s in high-viscous carrier liquids, such as glycerin ($\eta=10$ Ps). So, the condition $\Omega\tau_B\ll1$—and all the more $\langle\boldsymbol{\omega}\rangle\tau_B\ll1$—is usually satisfied. Then, with the linear accuracy in $\langle\boldsymbol{\omega}\rangle\tau_B$, the stationary solution of Eq. (25) reads

$$\langle\mathbf{b}\rangle=\langle\mathbf{b}\rangle_0+\tau_B\langle\boldsymbol{\omega}\rangle\times\langle\mathbf{b}\rangle_0. \tag{26}$$

On substitution of $\langle\mathbf{b}\rangle$ from Eq. (26) and $\langle\mathbf{b}\rangle_0$ from Eq. (23) into Eq. (22), one obtains with the same accuracy in $\langle\boldsymbol{\omega}\rangle\tau_B$

$$\langle\mathbf{e}\rangle\times\mathbf{h}=-\frac{4\sigma\tau_BF_2(\sigma)L^2(\xi)}{3\xi+4\sigma F_2(\sigma)L(\xi)}\langle\boldsymbol{\omega}\rangle. \tag{27}$$

Eliminating this vector product from the torque-balance equation (17) gives

$$\langle\boldsymbol{\omega}\rangle=\frac{3\xi+4\sigma F_2(\sigma)L(\xi)}{3\xi+4\sigma F_2(\sigma)L(\xi)+2\xi\sigma F_2(\sigma)L^2(\xi)}\mathbf{\Omega}. \tag{28}$$

As seen, the grains rotate freely ($\boldsymbol{\omega}=\mathbf{\Omega}$) if $\sigma=0$, i.e., if the particle magnetic moment is not coupled to the particle mass. [An analogy: The orientation of the magnetic needle of a compass from South to North does not impede a rotation of the compass frame around the vertical axis.] In the absence of magnetic field ($\xi=0$), the grains rotate freely as well. Vice versa, the stronger an applied magnetic field and the field of anisotropy, the slower the rotation of magnetic grains with respect to the surrounding liquid and hence the larger the rotational viscosity. Substituting $\langle\boldsymbol{\omega}\rangle$ from Eq. (28) into Eq. (18) yields the sought relationship for the viscosity coefficient:

$$\eta_R(\xi,\sigma)=\eta_R^{\mathrm{sat}}\frac{2\xi\sigma F_2(\sigma)L^2(\xi)}{2\sigma F_2(\sigma)L(\xi)[2+\xi L(\xi)]+3\xi}. \tag{29}$$

This is our main result.

III. DISCUSSION

Neglecting the square of fluctuations in Eq. (21), herewith we have supposed a smallness of deviations of $\langle\mathbf{e}\rangle$ and $\langle\mathbf{b}\rangle$ vectors from their equilibrium values (23). Therefore, the area of applicability of our approximation is limited to the case $\Omega\tau_B\ll1$. But this is not an additional limitation since the phenomenological approach itself is valid only for small $\Omega\tau_B$. If this product is of order one, the rotational viscosity coefficient becomes a function of Ω, so the ferrofluid acquires *non-Newtonian properties* [16,17]. However, phenomenological theories do not describe well the dependence $\eta_R(\Omega)$. For suspensions of rigid magnetic dipoles it was demonstrated in Refs. [10,18,19]: Only the effective-field approximation [3] guarantees the correct description of rotational viscosity even for $\Omega\tau_B\sim10$.

Setting about a discussion of our results, note first of all that Eq. (29) correctly describes the rotational viscosity in all conceivable limiting cases of strong and weak magnetic fields and fields of anisotropy. Let us consider them.

The formula (29) can be written

$$\eta_R=\eta_R^{\mathrm{sat}}\frac{\xi L(\xi)}{2+\xi L(\xi)+3(\xi/\sigma)[2F_2(\sigma)L(\xi)]^{-1}}. \tag{30}$$

This differs from Eq. (1) on only point: Eq. (30) contains an additional (last) term in the denominator. In the limit of magnetohard grains, $\sigma\to\infty$, this term disappears as $1/\sigma$ and Eq. (30) reduces to Eq. (1), i.e., we revert to the well-studied case of rigid dipoles. In Fig. 2, the reduced viscosity coefficient $\hat{\eta}$—calculated from Eqs. (5) and (29)—is plotted as a function of ξ for some fixed values of σ.

In the limiting case of high magnetic fields, Eq. (29) reduces to

$$\eta_R=\eta_R^{\mathrm{sat}}\frac{2\sigma F_2(\sigma)}{3+2\sigma F_2(\sigma)},\quad(\xi=\infty). \tag{31}$$

According to this, the ratio $\eta_R/\eta_R^{\mathrm{sat}}=4\sigma^2/45$ at small values of σ and tends to saturation as $1-3/(2\sigma)$ for $\sigma\gg1$. Figure 3 displays the reduced viscosity $\hat{\eta}(\infty,\sigma)$ given by our Eq. (31) and by the expression following from the Cebers-Stepanov's Eq. (5) at $\xi\to\infty$:

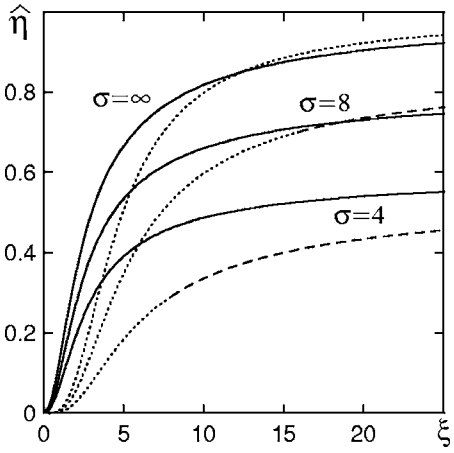

FIG. 2. The reduced viscosity $\hat{\eta}$ versus the dimensionless magnetic field ξ for suspensions of grains with the different magnetic anisotropy σ as calculated from our Eq. (31)—solid lines, and from the Cebers' expression (32)—dashed lines in the area of applicability of the expression, $\xi > 2\sigma$, and dotted lines out of the area.

$$\eta_R(\infty,\sigma) = \eta_R^{\mathrm{sat}} \frac{35F_2^2(\sigma)}{14 + 5F_2(\sigma) + 16F_4(\sigma)}.$$

Substituting here for F_2 and F_4 from Eq. (7) leads to the relationship [8]

$$\eta_R(\infty,\sigma) = \eta_R^{\mathrm{sat}} \frac{9(R'/R - 1/3)^2}{2(1 - 3R'/R + 4R''/R)}. \tag{32}$$

As seen in Fig. 3, the results of both approaches—phenomenological and microscopic—are similar in the case of large ξ but differ for small and moderate values of ξ as shown in Fig. 2.

At last, in the limit of low magnetic field, Eq. (29) gives [with allowance for the definition (4)]

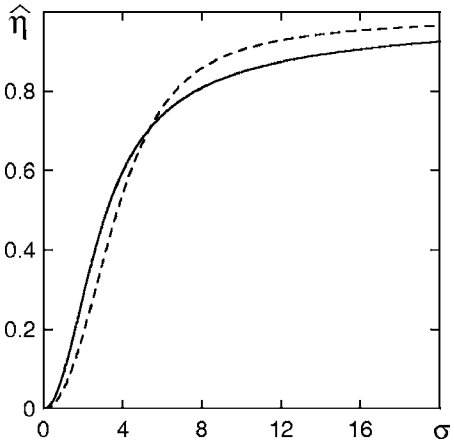

FIG. 3. The reduced saturation viscosity (in the infinitely strong magnetic field, $\xi=\infty$) versus the dimensionless energy of magnetic anisotropy σ as calculated from Eq. (31)—solid line, and from Eq. (31)—dashed line.

$$\eta_R = \eta\phi\xi^2 \frac{\sigma F_2(\sigma)}{9 + 4\sigma F_2(\sigma)}, \quad \xi \ll 1. \tag{33}$$

The Cebers-Stepanov resulting expression (5) is not valid in this limiting case. For the commonly used magnetite ferrofluids, the parameter σ lies within the interval 5…8, thus the condition of applicability of their microscopic theory, $\xi > 2\sigma$, is satisfied only at saturation, $\xi \gg 1$. This limitation greatly reduces the value of Eq. (5) since it cannot be applied to the most interesting and widely used region of the Langevin parameter $\xi < 10$ that corresponds to the field strength $H < 1500$ Oe.

Thus, we have demonstrated that the magnetoviscous effect in ferrofluids with the finite value of magnetic anisotropy of suspended grains does yield to phenomenological description. On the way, we obtained the sufficiently simple and compact expression (29) giving the reasonable dependencies of the rotational ferrofluid viscosity on both the applied magnetic field strength and the energy of magnetic anisotropy. Our formula is free from the above mentioned shortcomings and restrictions of previous microscopic theories. Interestingly, as it is seen from the dependencies presented in Figs. 2 and 3, ferrofluid viscometry—the typically hydrodynamic measurements—is capable of delivering a valuable information of such solid-body matter as the magnetic anisotropy of dispersed ferromagnets.

ACKNOWLEDGMENTS

The authors are grateful to Victor Stepanov for some explanations of his recent articles. The work of K.M. and M.S. was supported by INTAS (Grant No. 03-51-6064). M.S. and M.Z. acknowledge the support of the Binational (U.S.-Israel) Science Foundation (Grant No. 2004081).

APPENDIX

We indicate here the calculation of some quantities typical of the problem. Let us determine first the equilibrium partition function

$$Z_0 = \int \exp\{\xi(\mathbf{e}\cdot\mathbf{h}) + \sigma(\mathbf{e}\cdot\mathbf{n})^2\} d\mathbf{e}\, d\mathbf{n}. \tag{A1}$$

The integral is factorable. Indeed, integrating first over the orientation of anisotropy axis $\mathbf{n}$ and then over the orientation of magnetic moment $\mathbf{e}$, we find

$$Z_0 = \int e^{\sigma(\mathbf{e}\cdot\mathbf{n})^2} d\mathbf{n} \int e^{\xi(\mathbf{e}\cdot\mathbf{h})} d\mathbf{e} = 16\pi^2 R(\sigma) \frac{\sinh\xi}{\xi}. \tag{A2}$$

We now calculate the average value $\langle\mathbf{e}\rangle_0$. Since at equilibrium an external magnetic field sets a sole preferable direction in the problem, $\langle\mathbf{e}\rangle_0 = \langle(\mathbf{e}\cdot\mathbf{h})\mathbf{h}\rangle_0$. Thus,

$$\langle\mathbf{e}\rangle_0 = \mathbf{h}\frac{d\ln Z_0}{d\xi} = L(\xi)\mathbf{h}. \tag{A3}$$

To find the quantity $\langle\mathbf{b}\rangle_0 = \left\langle \frac{3}{2}(\mathbf{e}\cdot\mathbf{n})\mathbf{n} - \frac{1}{2}\mathbf{e}\right\rangle_0$, consider the auxiliary integral

$$N_{ik} = \int n_i n_k \exp\{\sigma(\mathbf{e}\cdot\mathbf{n})^2\}\mathrm{d}\mathbf{n}.$$

Generally, it can be written in the form

$$N_{ik} = A\,\delta_{ik} + B\left(\tfrac{3}{2}e_i e_k - \tfrac{1}{2}\delta_{ik}\right), \tag{A4}$$

where A and B are constants. Taking the trace of both sides of the last expression, we have $A = 4\pi R(\sigma)/3$. The constant B is determined by contraction of N_{ik} from Eq. (A4) with $e_i e_k$:

$$N_{ik}e_i e_k = A + B = \int (\mathbf{e}\cdot\mathbf{n})^2 e^{\sigma(\mathbf{e}\cdot\mathbf{n})^2}\mathrm{d}\mathbf{n} = 4\pi R'(\sigma). \tag{A5}$$

Therefore, $B = 4\pi(R'(\sigma) - R(\sigma)/3) = 8\pi R(\sigma)F_2(\sigma)/3$. Now one can write down $\langle b_i\rangle_0$ in the form

$$\langle b_i\rangle_0 = Z_0^{-1}\int \left(\tfrac{3}{2}N_{ik}e_k - \tfrac{1}{2}e_i\right)\exp\{\xi(\mathbf{e}\cdot\mathbf{h})\}\mathrm{d}\mathbf{e}. \tag{A6}$$

Substituting here N_{ik} from Eq. (A4), Z_0 from Eq. (A2), and the above constants A and B, finally gives

$$\langle \mathbf{b}\rangle_0 = F_2(\sigma)L(\xi)\mathbf{h}. \tag{A7}$$

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