Effective Viscosity of Magnetic Suspensions

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The effect of a homogeneous magnetic field on the viscosity of a suspension whose solid particles possess intrinsic magnetic moments is investigated. The orienting field impedes rotation of the particles in a vortical liquid flow, the result being an increase of the effective viscosity. Brownian motion and hydrodynamic forces exert a disorienting effect on the magnetic moments. The influence of the aforementioned factors on the rotation of suspended particles is taken into account macroscopically within the framework of the hydrodynamics of a homogeneous liquid with an internal angular momentum. The theoretical results are in good agreement with the experimental data on the dependence of the viscosity of magnetic suspensions on the field strength.

1. Einstein's formula for the viscosity of suspensions was obtained without taking into account the fact that the solid particles of the suspension can, in principle, rotate in ordered fashion relative to the liquid. When the particle rotation velocity $\boldsymbol{\omega}$ does not coincide with the local angular velocity of rotation of the liquid $\boldsymbol{\Omega} = (1/2)\,\mathrm{curl}\,\mathbf{v}$, friction forces arise, which should become manifest in an increase of the effective viscosity of the suspension.

Under ordinary conditions, when the suspended particles are acted upon by moments due only to the friction forces exerted by the surrounding liquid, the equalization of the angular velocities $\boldsymbol{\omega}$ and $\boldsymbol{\Omega}$ occurs within a very short time τ_S, on the order of a^2/ν, where a is the dimension of the particle and $\nu = \eta/\rho$ is the kinematic viscosity of the liquid. In stable suspensions $a \lesssim 10^{-5}$ cm, so that $\tau_S \lesssim 10^{-8}$ sec at $\nu \sim 10^{-2}$ cm^2/sec. It is clear therefore, that the "rotational" viscosity can become noticeable only if the difference $\boldsymbol{\Omega} - \boldsymbol{\omega}$ is maintained by moments from some extraneous forces acting directly on the particles. Concretely, we might deal here with the influence of a magnetic (electric) field on the motion of particles having their own magnetic (electric) moments.

A suspension of magnetic particles is a colloidal dispersion of a ferromagnet in an ordinary non-conducting liquid. At the present time such colloids are obtained either by grinding a ferromagnet in a suitable liquid (for example magnetic in kerosene[1]), or by precipitating the particles from a solution containing atoms of a ferromagnetic metal (for example, from a solution of $Co_2(CO)_8$ in toluene[2]). Particles with dimensions $a \lesssim 10^{-6}$ cm turn out to be single-domain; their magnetic moments μ are of the order of 10^{-16}–10^{-15} erg/G (10^4–10^5 Bohr magnetons). A suspension of such particles is very sensitive to a magnetic field and behaves in many respects like a homogeneous medium. During the last five—six years, the hydrodynamics of magnetic suspensions—"ferrohydrodynamics"—has been the subject of several dozen papers[1)].

So long as there is no external magnetic field and the particle concentration is not too high ($N \sim 10^{14}$–10^{16} cm^{-3}), the properties of a suspension are close to the properties of the main liquid, and the viscosity satisfies the Einstein formula. When the suspension is placed in a homogeneous field $\mathbf{H}$, the latter causes a partial orientation of the magnetic moments ("superparamagnetism"). In order to explain the dependence of the viscosity on the field, let us consider the motion of an individual spherical particle in a homogeneous shear stream ($\boldsymbol{\Omega}$ = const, planar couette flow). In the absence of the field, the particle "glides" along the corresponding shear plane with angular velocity $\boldsymbol{\omega}$ equal to $\boldsymbol{\Omega}$. In a magnetic field, the particle is acted upon by the moment of the force $\boldsymbol{\mu} \times \mathbf{H}$, which changes the state of its rotation. As a result, a friction-force torque is produced

$$8\pi a^3\eta(\Omega - \omega),$$

i.e., a rotational-viscosity mechanism "is turned on." The latter reaches a maximum value when the particle is carried by the stream without rotating. Equilibrium of the torques ($\omega = 0$) should occur at

$$\mu H \geqslant 8\pi a^3\eta\Omega. \tag{1}$$

Such a conclusion was arrived at by Hall and Busenberg[4]. For typical values of the parameters ($\mu \sim 10^{-16}$ erg/G, $\eta \sim 10^{-2}$ g/cm-sec, $a \sim 10^{-6}$ cm) and $\Omega \sim 10^2$ sec^{-1}, formula (1) gives a value $H \sim 1$ Oe. This result contradicts strongly the experimental data of McTague[5], according to whom saturation of the viscosity as a function of the field sets in in a suspension with the indicated parameters (cobalt particles in toluene) at $H \sim 10^3$ Oe.

McTague states correctly that the reason for the discrepancy between the calculated and the experimental data is the fact that the rotational Brownian motion of the particle is neglected in the derivation of formula (1). In fact, allowance for the thermal motion leads to the condition

$$\mu H \gg kT, \tag{2}$$

which turns out to be stronger than (1) at $a \lesssim 10^{-5}$ cm.

The theory presented below is valid for all values of the ratio $8\pi a^3\eta\Omega/kT \equiv 2\Omega\tau$ (τ is the characteristic time of rotation of a particles suspended in a viscous liquid). The results of the theory agree well with experiment[5].

[1)]A bibliography can be found in the review[3].

2. In the hydrodynamic description of a suspension as a homogeneous medium, we should consider as an independent variable (besides the velocity v, the density ρ, and the pressure p) also the internal angular momentum, the volume density of which will be denoted by **S**. This quantity characterizes the intensity of the rotation of the solid particles of the suspension: in the case of small concentrations of identical spherical particles we can write $\mathbf{S} = I\omega$, where I is the sum of moments of inertia of the spheres per unit volume and ω is the average velocity of their ordered rotation[2)].

For a liquid with internal rotation, the laws of momentum and angular-momentum conservation are expressed by the equations

$$\rho\frac{dv_i}{dt}=\frac{\partial\sigma_{ik}}{\partial x_k},\qquad \frac{dS_{ik}}{dt}=\sigma_{ki}-\sigma_{ik},$$
$$\frac{d}{dt}=\frac{\partial}{\partial t}+v_k\frac{\partial}{\partial x_k},\qquad S_{ik}=e_{ikl}S_l. \tag{3}$$

The stress tensor σ_{ik} was calculated by the author in[7]

$$\sigma_{ik}=-\left[p+\frac{\mathbf{S}}{I}(\mathbf{S}-I\Omega)\right]\delta_{ik}+\eta\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}\right)$$
$$+\frac{1}{2\tau_s}(S_{ik}-I\Omega_{ik})+\frac{1}{4\pi}\left[H_iB_k-\frac{1}{2}(\mathbf{HB})\delta_{ik}\right], \tag{4}$$
$$\Omega_{ik}=\frac{1}{2}\left(\frac{\partial v_k}{\partial x_i}-\frac{\partial v_i}{\partial x_k}\right)=e_{ikl}\Omega_l,\quad \mathbf{B}=\mathbf{H}+4\pi\mathbf{M}.$$

We have included in σ_{ik} the Maxwellian stress tensor, since we are dealing here with a magnetic suspension. Substituting the tensor σ_{ik} and its antisymmetrical part

$$\sigma_{ki}-\sigma_{ik}=(M_iH_k-M_kH_i)-(S_{ik}-I\Omega_{ik})/\tau_s$$

in (3), we obtain

$$\rho\frac{d\mathbf{v}}{dt}=-\nabla\left[p+\frac{1}{2}(\mathbf{MH})+\frac{\mathbf{S}}{I}(\mathbf{S}-I\Omega)\right]+\eta\Delta\mathbf{v}$$
$$+(\mathbf{M}\nabla)\mathbf{H}+\frac{1}{2\tau_s}\operatorname{rot}(\mathbf{S}-I\Omega), \tag{5}$$
$$\frac{d\mathbf{S}}{dt}=[\mathbf{MH}]-\frac{1}{\tau_s}(\mathbf{S}-I\Omega). \tag{6*}$$

In the calculation of the divergence of the stress tensor, we have used the equations

$$\operatorname{div}\mathbf{v}=0,\qquad \operatorname{rot}\mathbf{H}=0,\qquad \operatorname{div}\mathbf{B}=0, \tag{7}$$

i.e., the liquid is assumed to be incompressible and non-conducting.

In the hydrodynamics of liquids with gyromagnetic properties[8,6] (for example, for diatomic paramagnetic gases), the system of equations (5)—(7) turns out to be complete, since $\mathbf{M} = \gamma\mathbf{S}$ in that case. This relation transforms (6) into the equation of the dynamics of magnetization[8]. On the other hand, in the case of ferromagnetic suspensions, there is no direct connection between M and S. Thus, for example, the medium may be magnetized also in the absence of ordered rotation of the particles ($\mathbf{M} \parallel \mathbf{H}$, $\mathbf{S} = \Omega = 0$) or, to the contrary, the particles may rotate in similar fashion in the absence of any predominant orientation of the magnetic moments ($\mathbf{S} = I\Omega$, $\mathbf{M} = \mathbf{H} = 0$). Therefore, in order to obtain a closed system of equations, it is necessary to add the equation for dM/dt to the system (5)—(7).

Let us derive this missing equation. To this end, we introduce, for a fixed volume element of the medium, a local reference frame R′, in which the average velocity of the suspended particles is equal to zero. The magnetization in the R′ system is described by the relaxation equation

$$\frac{d'\mathbf{M}}{dt}=-\frac{1}{\tau}\left(\mathbf{M}-M_0\frac{\mathbf{H}}{H}\right). \tag{8}$$

The equilibrium magnetization M_0 of the considered system of "floating" magnetic moments μ (homogeneously magnetized single-domain particles) satisfies the Langevin formula

$$M_0=N\mu\mathscr{L}(\mu H/kT). \tag{9}$$

As indicated by Neel[9], a spontaneous change of the magnetization direction can occur in very minute single-domain particles under the influence of thermal fluctuations. The mobility of the vector μ is characterized by a relaxation time τ_N determined by the expression

$$1/\tau_N=f\exp(-KV/kT), \tag{10}$$

where K is the anisotropy constant, V the volume of the particle, and f a frequency factor of the order of $10^9\ \mathrm{sec}^{-1}$. Another mechanism of reorientation of the magnetic moment of a particle suspended in a liquid is connected with the rotation of the particle itself, and is characterized by the Brownian time of rotational diffusion

$$\tau_B=3V\eta/kT. \tag{11}$$

The relaxation time τ of the magnetization of the suspension is obviously determined by the shorter of the times τ_N and τ_B. At room temperatures $K \sim 10^6$ erg/cm^3 and $\eta \sim 10^{-2}$ g/cm-sec, and the equality $\tau_N = \tau_B$ is satisfied for particles having a radius $a_* = 20$ Å, but we get $\tau_N \approx 300\,\tau_B$ already for $a = 30$ Å. Thus, when $a > a_*$ we have $\tau_N \gg \tau_B$, i.e., such particles can be regarded as rigid magnetic dipoles. The relaxation time τ which enters in (8) coincides in this case with the Brownian time τ_B.

Equation (8) has been written out in a coordinate system R′ which rotates relative to the immobile system R with angular velocity $\omega = \mathbf{S}/I$. With the aid of the general formula

$$\frac{d\mathbf{M}}{dt}=\frac{d'\mathbf{M}}{dt}+[\omega\mathbf{M}],$$

which connects the velocities of the change of the vector **M** in the systems R and R′, we obtain

$$\frac{d\mathbf{M}}{dt}=\frac{1}{I}[\mathbf{SM}]-\frac{1}{\tau}\left(\mathbf{M}-M_0\frac{\mathbf{H}}{H}\right). \tag{12}$$

Equations (5)—(7) and (12) form a complete system of equations.

3. Let us consider one-dimensional stationary Couette flow: the liquid is contained between two parallel planes, of which one (d = 0) is immobile and the other (z = h) moves along the x axis with velocity

[2)]We have in mind averaging over volume elements that are small in comparison with hydrodynamic dimensions, but still contain a large number of suspended particles. For nonspherical particles, formulas connecting S with ω are given in[6].

*[MH] ≡ M × H.

$u \equiv 2\Omega h$. Equations (5) and (7) are satisfied identically at

$$\mathbf{v} = (2\Omega z,\, 0,\, 0), \quad \mathbf{\Omega} = (0,\, \Omega,\, 0),\ \mathbf{H} = (0,\, 0,\, H), \quad p,\ H,\ \Omega,\ \mathbf{M},\ \mathbf{S} = \mathrm{const}, \tag{13}$$

and Eqs. (6) and (12) take the form

$$\mathbf{S} - I\mathbf{\Omega} = \tau_s[\mathbf{MH}], \tag{14}$$

$$\frac{1}{I}[\mathbf{SM}] = \frac{1}{\tau}\left(\mathbf{M} - M_0\frac{\mathbf{H}}{H}\right). \tag{15}$$

Equation (14) makes it possible to eliminate S from (15) and (4):

$$[\mathbf{\Omega M}] = \frac{1}{\tau}\left(\mathbf{M} - M_0\frac{\mathbf{H}}{H}\right) + \frac{\tau_s}{I}[\mathbf{M}[\mathbf{MH}]], \tag{16}$$

$$\sigma_{ik} = (\ldots)\delta_{ik} + \eta\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right) + \frac{1}{2}(M_iH_k - M_kH_i) + \frac{H_iB_k}{4\pi}. \tag{17}$$

In all cases when $dS/dt = 0$, the stress tensor turns out to be symmetrical (3), as can be easily verified by adding the last two terms in (17).

The magnetization of the suspension is determined by Eq. (16). Let us indicate first an approximate solution of this equation, which is valid when

$$\Omega\tau \ll 1. \tag{18}$$

We note immediately that the condition (18) can be violated only in very viscous liquids at high shear velocities. For ordinary liquids ($\eta \sim 10^{-2}$ g/cm-sec) at temperatures $kT \approx 4 \times 10^{-14}$ erg and particle radii $a \sim 10^{-X}$ cm formula (11) yields a value $\tau \sim 10^{-5}$ sec, so that the condition (18) is satisfied at all these little values of Ω. In this case, assuming

$$\mathbf{M} = M_0\mathbf{H}/H + \mathbf{m}, \tag{19}$$

we can linearize Eq. (16) with respect to $\mathbf{m}$:

$$\tau\frac{M_0}{H}[\mathbf{\Omega H}] = \mathbf{m} + \frac{\tau_s\tau}{I}\frac{M_0}{H}[\mathbf{H}[\mathbf{mH}]]. \tag{20}$$

It follows therefore that

$$\mathbf{m} = \frac{M_0\tau}{H(1 + \tau_s\tau M_0H/I)}[\mathbf{\Omega H}], \tag{21}$$

and substituting the obtained value of $\mathbf{M}$ in (17) we get

$$\sigma_{ik} = (\ldots)\delta_{ik} + \eta\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right) + \frac{H_iB_k}{4\pi} - \frac{M_0\tau}{2H(1 + \tau_s\tau M_0H/I)}e_{ikl}[\Omega_lH^2 - H_l(\mathbf{\Omega H})]. \tag{22}$$

Let us calculate the friction force acting on a solid surface $z = 0$ in contact with the liquid. A unit surface area is acted upon by a force $f_i = (\sigma_{ik} - \sigma'_{ik})n_k$, where σ'_{ik} is the Maxwellian stress tensor in the solid and $\mathbf{n}$ is a unit vector in the direction of the outward normal to the surface of the liquid. Since the boundary conditions require continuity of H_x and B_z, we obtain

$$f_x = \sigma_{xz} - \sigma_{xz}' = \eta\left(\frac{\partial v_x}{\partial z} + \frac{\partial v_z}{\partial x}\right) + \frac{M_0\tau[\Omega_yH^2 - H_y(\mathbf{\Omega H})]}{2H(1 + \tau_s\tau M_0H/I)},$$

or, substituting the vector components from (13)

$$f_x = 2\Omega\{\eta + \tfrac{1}{4}M_0H\tau(1 + \tau_s\tau M_0H/I)^{-1}\}. \tag{23}$$

The quantity in this expression, summed with the usual viscosity, should be regarded as the rotational viscosity

$$\Delta\eta = \frac{M_0H\tau}{4(1 + \tau_s\tau M_0H/I)}. \tag{24}$$

Substituting in (24) the expression obtained in[7]

$$\tau_s = a^2\rho'/15\eta, \quad I = \tfrac{2}{5}a^2\rho'\varphi, \quad \varphi = \tfrac{4}{3}\pi a^3N$$

(ρ' and φ are the density and volume concentration of the solid phase) and the values indicated above

$$M_0 = N\mu\mathscr{L}(\xi), \quad \tau = 3\varphi\eta/NkT, \quad \xi = \mu H/kT, \quad \mathscr{L}(\xi) = \mathrm{cth}\,\xi - \xi^{-1},$$

we obtain ultimately

$$\Delta\eta = \frac{3}{2}\varphi\eta\frac{\xi - \mathrm{th}\,\xi}{\xi + \mathrm{th}\,\xi}. \tag{25}$$

In the absence of a field, when $\Delta\eta = 0$, the viscosity of the suspension is described by the Einstein formula

$$\eta = \eta_0(1 + \tfrac{5}{2}\varphi), \tag{26}$$

where η_0 is the viscosity of the main liquid. The sum of the last two expressions determines the effective viscosity of the magnetic suspension in the field. With first-order accuracy with respect to φ we have

$$\eta(\xi) = \eta_0\left(1 + \frac{4\xi + \mathrm{th}\,\xi}{\xi + \mathrm{th}\,\xi}\varphi\right). \tag{27}$$

Let us ascertain also the influence of the field on the rotation of the particles in this stream. From (14) and (21) we get

$$\mathbf{S} = I\mathbf{\Omega}(1 + A)^{-1}, \quad A = \tfrac{1}{2}\xi\mathscr{L}(\xi). \tag{28}$$

We see therefore that with increasing field intensity the particle rotational velocity ω decreases from its maximum value $\omega = \Omega$ at $\xi = 0$ to zero as $\xi \to \infty$. The slowing down of the particle rotation is accompanied by an increase of the effective viscosity (27)

$$\eta(\infty) - \eta(0) = \tfrac{3}{2}\varphi\eta_0.$$

At $\Omega\tau \gtrsim 1$, the rotational viscosity turns out to be dependent on Ω. The exact solution of (16) leads to the formula

$$\Delta\eta = \tfrac{3}{2}\varphi\eta F(\xi, \Omega\tau), \quad F = A\delta(1 + A\delta)^{-1}, \tag{29}$$

where the parameter δ should be determined for each pair of values of ξ and $\Omega\tau$ from the equation

$$(1 - \delta)(1 + A\delta)^2 = (\Omega\tau)^2\delta, \quad 0 \leqslant \delta \leqslant 1. \tag{30}$$

It is easy to verify that at $\Omega\tau = 0$ we get

$$F(\xi, 0) = (\xi - \mathrm{th}\,\xi)/(\xi + \mathrm{th}\,\xi) \tag{31}$$

and formula (29) goes over into (25). The function $F(\xi)$ for different values of $\Omega\tau$ is shown in Fig. 1.

The dependence of the viscosity on Ω means that at $\tau\Omega \gtrsim 1$ the stress tensor becomes a nonlinear function

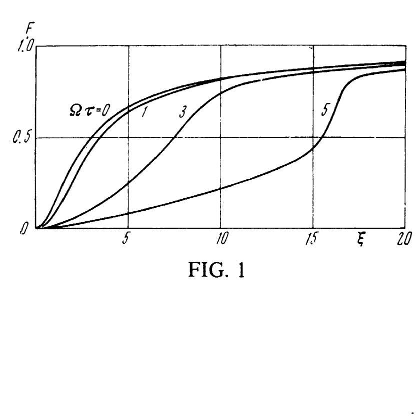

FIG. 1

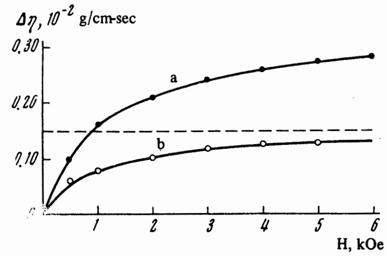

FIG. 2

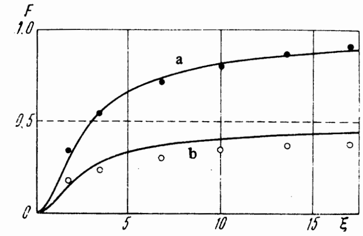

FIG. 3

of the velocity gradients, i.e., the suspension exhibits non-Newtonian properties. From (30) and from the expression for F, written in the form $F = A\sqrt{\delta(1-\delta)}/\Omega\tau$, it follows that at $\Omega\tau \gg 1$ the rotational viscosity reaches a maximum value in fields corresponding to $\xi \geq 4\Omega\tau$.

We have considered the case when the vector **H** is perpendicular to **Ω**. At an arbitrary orientation of these vectors, formulas (25) and (28) take the form

$$\Delta\eta = \frac{3}{2}\varphi\eta\frac{\xi - \operatorname{th}\xi}{\xi + \operatorname{th}\xi}\sin^2\alpha, \quad S = I\Omega\frac{1 + A\cos^2\alpha}{1 + A}, \qquad (32)$$

where α is the angle between **H** and **Ω**. As seen from (32), when **H** ∥ **Ω** the viscosity is independent of the field. This result is perfectly understandable, since the orientation of the magnetic moment of the particle along the field does not prevent it from rotating with an angular velocity **Ω** in the same direction.

4. The dependence of the viscosity of a magnetic suspension on the field was investigated experimentally in[5]. The investigation was made on a suspension of cobalt particles in toluene. The average radius a_0 of the metallic particles, measured with an electron microscope, was approximately 30 Å. Owing to the adsorption of polymer molecules introduced into the solution to stabilize the suspension, the effective hydrodynamic radius was larger by one order of magnitude ($a \approx 3 \times 10^{-6}$ cm), so that at a particle number concentration $N \approx 1.6 \times 10^{15}$ cm^{-3} the volume concentration of the suspension reached 0.2. The viscosity was determined from the time required for the suspension to flow through a round capillary placed in a homogeneous magnetic field. The experiments revealed a monotonic increase of the viscosity from an initial value $\eta = 1.15 \times 10^{-2}$ g/cm-sec at H = 0 to a maximum value $\eta = 1.45 \times 10^{-2}$ at H = 8 kOe. The results of one of McTague's experiments are shown in Fig. 2, which is taken from[5]. The two experimental curves on this diagram were obtained at different orientations of the magnetic field relative to the liquid flow direction: the upper curve (a) corresponds to **H** ∥ **v** and the lower (b) to **H** ⊥ **v**. In the Poiseuille flow with which we are dealing, the isolines of the velocity curl (Ω = const) are concentric circles in the capillary cross section plane. Thus, in the case a the vector **H** at each point of the stream is perpendicular to **Ω**, and therefore, in accord with (32), we should have

$$\Delta\eta_a = \frac{3}{2}\varphi\eta F(\xi).$$

In case b, the angle between **H** and **Ω** assumes all values from zero to 2π, so that

$$\Delta\eta_b = {}^3/_2\varphi\eta F(\xi)\overline{\sin^2\alpha} = {}^3/_4\varphi\eta F(\xi),$$

i.e., $\Delta\eta_a = 2\Delta\eta_b$. This result, as seen from the diagram, agrees with experiment.

Figure 3 shows theoretical plots of $F(\xi)$ and $F(\xi)/2$, calculated from formula (31)[3)] The figure shows also the experimental points taken from Fig. 2. In calculating from the H scale to the scale of $\xi = \mu H/kT$, we used the experimental temperature T = 24°C indicated in[5] and $\mu = 1.4 \times 10^{-16}$ erg/G. This value of the magnetic moment of the particle is in good agreement with an independent estimate, according to which $\mu = (4\pi/3)a_0^3 M_S$, where $M_S \approx 1400$ G is the magnetization of cobalt. For the value of μ chosen by us, this formula yields a_0 = 29 Å, which is close to the mean value a_0 = 30 Å measured in[5]. At the indicated μ and T, the value $\xi = 1$ corresponds to H = 300 Oe.

It should be noted in conclusion that the action of the magnetic field will far from always be reduced to a simple "renormalization" of the viscosity. Thus, when a suspension moves in an inhomogeneous field there arise forces (see Eq. (5)) which change the very character of the motion. It is also important that in the examples considered above (Couette and Poiseuille flows) the nonlinear terms $(\mathbf{v}\cdot\nabla)\mathbf{S}$ and $(\mathbf{v}\cdot\nabla)\mathbf{M}$ vanish identically from Eqs. (6) and (12). In the general case, the dependence of the viscosity on the field is only a fraction of the effect of the influence of the field on the motion of a suspension of magnetic particles.

3)The value used in the experiments of [5] was $\Omega \approx 4 \times 10^2$ sec^{-1}, so that condition (18) was well satisfied.

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