

# Magnetic viscosity of a suspension of ferromagnetic particles

M. A. Martsenyuk

Perm' State University

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An expression is obtained for the viscosity tensor of a dilute suspension of ferromagnetic particles placed in a uniform magnetic field. The independent viscosity coefficients are calculated for particles of arbitrary shape by taking into account their rotational Brownian motion. The relation between the viscosity coefficients and the particle symmetry is investigated. It is shown that in the general case the number of independent components of the viscosity tensor is seven (six of them are even in the field and one odd). The nature of the dependence of the coefficients on the value of the magnetic field is explained, and it is shown that at large fields all the viscosity coefficients saturate. For nearly spherical particles, the even coefficients are linear in the asphericity parameter, the odd one at least quadratic.

## 1. STATEMENT OF THE PROBLEM

A ferromagnetic suspension is a suspension of fine ( $\sim 100 \text{ \AA}$ ) particles, magnetized to saturation, in a neutral liquid. If the external magnetic field  $\mathbf{H}$  is less than the internal anisotropy field and if it is permissible to neglect relaxation processes in the solid phase, then the direction of the magnetic moment  $\boldsymbol{\mu}$  of an individual particle remains constant with respect to the particle (a rigid dipole)<sup>[1]</sup>.

The field exerts on each particle of the suspension a torque  $[\boldsymbol{\mu} \times \mathbf{H}]$  that tends to orient the particle. In a rotation of the suspension, this leads to a dissipation of energy. From this it may be concluded that in contrast to ordinary fluids<sup>[2]</sup>, the viscous-stress tensor  $\sigma_{ik}$  of a ferrosuspension depends not only on a symmetric combination of the velocity gradients but also on the antisymmetric  $\Omega^{(1)} = \frac{1}{2} \text{curl } \mathbf{v}$ . On separating  $\sigma_{ik}$  into its irreducible parts

$$\sigma_{ik} = \sigma^{(0)} \delta_{ik} + \frac{1}{2} \epsilon_{ikl} \sigma_l^{(1)} + \sigma_{ik}^{(2)} \quad (1.1)$$

and using the equation of motion of the suspension

$$\rho \frac{dv_i}{dt} = \frac{\partial}{\partial x_k} \sigma_{ki}, \quad \text{div } \mathbf{v} = 0, \quad (1.2)$$

one finds that the energy dissipated in the whole liquid in unit time has the form (see<sup>[2]</sup>, sect. 16)

$$E_{\text{kin}} = - \int (\sigma_{ik}^{(1)} \Omega_i^{(1)} + \sigma_{ik}^{(2)} \Omega_{ik}^{(2)}) dV, \quad \Omega_{ik}^{(2)} = \frac{1}{2} \left( \frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i} \right). \quad (1.3)$$

The appearance of the new thermodynamic force  $\Omega^{(1)}$  necessitates the introduction of additional kinetic coefficients of viscosity. The additional dissipation of energy is determined by the way in which the liquid stream flows around the particles of the suspension and thus depends on their shape. For given particle shape, the problem consists in the calculation of the additional coefficients of viscosity and of their dependence on the field. To find the viscosity tensor of a dilute suspension, one uses Einstein's method (see<sup>[2]</sup>, Sec. 22), generalized to the case of particles of arbitrary shape<sup>[3]</sup>. In addition, because of the smallness of the suspended particles it is necessary to take into account also their rotational (orientational) Brownian motion.

When we go over to spherical components of the tensors (see Sec. 2), we shall write the relation between the thermodynamic fluxes  $\sigma^{(l_1)}$  ( $l_1 = 0, 1, 2$ ) and forces  $\Omega^{(l_2)}$  ( $l_2 = 1, 2$ ) in the form

$$\sigma_{m_1}^{(l_1)} = -\rho \delta_{l_1 0} + \sum_{l_2 m_2} \eta_{m_1 m_2}^{(l_1 l_2)} \Omega_{m_2}^{(l_2)} \quad (-l_2 \leq m_2 \leq l_2), \quad (1.4)$$

where  $p$  is the pressure and where  $\eta_{m_1 m_2}^{(l_1 l_2)}$  is the viscosity tensor to be determined. On decomposing the spherical tensor of second rank  $\eta^{(l_1 l_2)}$  into irreducible parts and on taking into consideration that the tensor character of the viscosity is determined by the magnetic field alone, we have

$$\eta_{m_1 m_2}^{(l_1 l_2)} = \sum_i C_{l_1 m_1 l_2 m_2}^{lm} \eta(l_1 l_2) Y_{lm}(\mathbf{h}), \quad \mathbf{h} = \mathbf{H}/H, \quad (1.5)$$

$$|l_1 - l_2| \leq l \leq l_1 + l_2, \quad m = m_1 + m_2,$$

where  $C_{l_1 m_1 l_2 m_2}^{lm}$  is the Clebsch-Gordon coefficient and  $Y_{lm}(\mathbf{h})$  is a spherical harmonic. Formula (1.5) contains sixteen reduced coefficients of viscosity  $\eta(l_1 l_2)$  (the first index in parentheses shows the parity with respect to the field, the second and third the tensor character of the flux and force). As will be shown below (Sec. 3), the viscosity tensor of a ferrosuspension contains only seven independent coefficients  $\eta(l_1 l_2)$ ; six of these,  $\eta(020)$ ,  $\eta(011)$ ,  $\eta(212)$ , and  $\eta(222)$  ( $l = 0, 2, 4$ ), are even in the field, and one,  $\eta(112)$ , is odd. The reduction of the number of coefficients is due here to the Stokesian character of the flow around particles of arbitrary shape; this leads to symmetry of the generalized coefficients of friction (Sec. 2). If the particles are symmetric, then there is a further decrease in the number of independent coefficients; for example, for spherical particles (Cf. <sup>[4,5]</sup>) there remain only two coefficients: the ordinary (shear) viscosity  $\eta(022) = 4\sqrt{5}\pi \eta_0 (1 + \frac{5}{2}\varphi)$  ( $\eta_0$  is the initial viscosity of the liquid, and  $\varphi$  is the volume concentration of particles) and the additional rotational viscosity  $\eta(011)$ .

Because of the effect of thermal motion, all the viscosity coefficients depend on the field through the Langevin argument  $\xi = \mu H/kT$  and saturate with increase of field. Because of the large value of  $\mu$  ( $\sim 10^4$  to  $10^5$  Bohr magnetons), saturation occurs at room temperature in fields  $H \sim 10^3$  Oe.

## 2. GENERALIZED COEFFICIENTS OF FRICTION

To calculate the viscosity of a suspension, it is necessary first to consider the auxiliary hydrodynamic problem of the flow around a solid particle, of given shape, by a liquid stream which at infinity has a constant velocity  $v_i$  and constant velocity gradients  $\Omega_{ik}^{(1)}$ ,  $\Omega_{ik}^{(2)}$  (at the surface of the particle, the usual boundary conditions of "adherence" are satisfied). In the Stokes approximation, the force  $F_i$ , the torque  $L_i$ , and the

stress tensor  $S_{ik}/V$  averaged over the volume  $V$  of the particle, exerted on the particle by the stream, are linear in the differences  $v_i - u_i$  and  $\Omega_i^{(1)} - \omega_i$  and in  $\Omega_{ik}^{(2)}$  ( $u_i$  and  $\omega_i$  are the velocity and the angular velocity of the particle):

$$\begin{pmatrix} F_i \\ L_i \\ S_{ik}^{(2)} \end{pmatrix} = \begin{pmatrix} a_{ij}^{(11)} & c_{i,j}^{(11)} & c_{i,jl}^{(12)} \\ c_{i,j}^{(11)} & b_{i,j}^{(11)} & b_{i,jl}^{(12)} \\ c_{ik,j}^{(21)} & b_{k,j}^{(21)} & b_{ik,jl}^{(22)} \end{pmatrix} \begin{pmatrix} v_j - u_j \\ \Omega_j^{(1)} - \omega_j \\ \Omega_{jk}^{(2)} \end{pmatrix}, \quad (2.1)$$

$$F_i = \oint \sigma_{in} n_n ds, \quad S_{ik} = \oint x_i \sigma_{kn} n_j ds, \quad L_i = S_i^{(1)}$$

Here  $\sigma_{ik}$  is the stress tensor of the liquid that flows around the particle, the integration extends over the surface of the particle, and the decomposition of  $S_{ik}$  into irreducible parts is analogous to (1.1). The generalized coefficients of friction in (2.1) depend on the shape of the particle and on its orientation with respect to the flow. As can be shown by use of the general properties of solutions of the equations of hydrodynamics in the Stokes approximation [6] (see also [7], Sec. 123), the tensors  $a$ ,  $b$ , and  $c$  possess the following symmetry property:

$$a^{(i_1 i_2)} = a^{(i_2 i_1)}, \quad b^{(i_1 i_2)} = b^{(i_2 i_1)}, \quad c^{(i_1 i_2)} = c^{(i_2 i_1)} \quad (2.2)$$

(with appropriate transposition of the lower indices, separated by commas); and in addition, they remain unchanged under transformations of the group  $G$  that leave the shape of the surface of the particle unchanged.

Since inertial effects are being neglected, the conditions  $F_i = 0$  and  $L_i + K_i = 0$  ( $K_i$  is the torque due to external forces) enable us to express, from (2.1), the difference  $\Omega_i^{(1)} - \omega_i$  (and also  $S_{ik}$ ) in terms of  $K_i$  and  $\Omega_{ik}^{(2)}$ :

$$\omega_i = \Omega_i^{(1)} + \frac{1}{\eta_0 V} g_{i,k}^{(11)} K_k + g_{i,kl}^{(12)} \Omega_{kl}^{(2)}. \quad (2.3)$$

On turning now to the main problem, determination of the viscosity of the suspension, we note that the volume-average stress tensor  $\bar{\sigma}_{ik}$  can be expressed in terms of the values of  $S_{ik}$  and  $\Omega_{ik}^{(2)}$  [2,3]:

$$\bar{\sigma}_{ik} = -p \delta_{ik} + 2\eta_0 \bar{\Omega}_{ik}^{(2)} + N S_{ik}$$

( $N = \phi/V$  is the number density of the particles). Thus the stresses in the suspension are composed of the stresses in a Newtonian fluid and of internal stresses in the particles (the latter are caused by the motion of the liquid).

On further averaging  $\bar{\sigma}_{ik}$  over orientation of the particles and on using (2.1) and (2.3), we obtain the effective stress tensor of the suspension:

$$\begin{aligned} \sigma^{(0)} &= -p + N \langle S^{(0)} \rangle = -p - \frac{\Phi}{V} \langle g_i^{(11)} (01) K_i \rangle + \eta_0 \Phi \langle g_{ik}^{(21)} (02) \rangle \Omega_{ik}^{(2)}, \\ \sigma_i^{(1)} &= N \langle S_i^{(1)} \rangle = -\frac{\Phi}{V} \langle K_i \rangle, \end{aligned} \quad (2.4)$$

$$\sigma_{ik}^{(2)} = 2\eta_0 \langle \bar{\Omega}_{ik}^{(2)} \rangle + N \langle S_{ik}^{(2)} \rangle = -\frac{\Phi}{V} \langle g_{ik,i}^{(21)} K_i \rangle + \eta_0 \Phi \langle g_{ik,lm}^{(22)} \rangle \Omega_{lm}^{(2)}.$$

Here  $\sigma_{ik}$  and  $\Omega_{ik}^{(2)}$  now denote the mean values  $\langle \bar{\sigma}_{ik} \rangle$  and  $\langle \bar{\Omega}_{ik}^{(2)} \rangle$  (angular brackets denote the average over angles). For brevity, the values  $2\eta_0 \langle \bar{\Omega}_{ik}^{(2)} \rangle$  corresponding to the stresses in the liquid in the absence of the particles are included in  $g^{(22)} \Omega^{(2)}$ . The tensors  $g^{(l_1 l_2)}$  in (2.3) and (2.4) can be expressed in an obvious manner in terms of the generalized coefficients of friction  $a$ ,  $b$ , and  $c$ , and in consequence of their symmetry have the properties

$$g_{i,k}^{(11)} = g_{k,i}^{(11)}, \quad g_{i,k,l}^{(12)} = g_{k,l,i}^{(12)}, \quad g_{ik,lm}^{(22)} = g_{lm,ik}^{(22)}. \quad (2.5)$$

In a system of coordinates  $\bar{S}$  rigidly attached to the

particle, the tensors  $g^{(l_1 l_2)}$  have constant values  $\bar{g}^{(l_1 l_2)}$  independent of the orientation of the particle (hereafter a bar over the tensor denotes transformation to the system  $\bar{S}$ ). In order to establish the relation between  $g$  and  $\bar{g}$  it is convenient to go over to the spherical components of these tensors. We introduce the unitary transformation matrix  $t_{ik...r}^{(lm)}$  ( $l$  lower indices), which converts an irreducible Cartesian tensor of rank  $l$  to a spherical tensor of first rank and of weight  $l$  (cf. [8]):

$$g_m^{(l)} = t_{ik...r}^{(lm)} g_{ik...r}^{(l)}, \quad t_{ik...r}^{(lm)} = c_l \int n_i n_k \dots n_r Y_{lm}(n) dn, \quad (2.6)$$

where  $\mathbf{n}$  is a unit vector, and where the constant  $c_l$  is determined by the unitarity condition  $\bar{t} t^* = 1$ . Spherical tensors  $g_m^{(l)}$  and  $\bar{g}_m^{(l)}$  given in different coordinate systems (rotated with respect to each other) are related by the transformation [9]

$$g_m^{(l)} = \sum_{m'} D_m^{(l)}(\alpha) \bar{g}_{m'}^{(l)}, \quad (2.7)$$

where  $D_m^{(l)}(\alpha)$  is the finite-rotation matrix, dependent on the Euler angles  $\alpha = (\varphi, \theta, \psi)$ . The tensors  $g^{(l_1 l_2)}$  are irreducible only with respect to each of the sets of indices separated by a comma. Therefore on transformation to spherical components they are transformed to spherical tensors of the second rank, which in turn can be represented as sums of tensors of the first rank

$$g_{m_1 m_2}^{(l_1 l_2)} = t_{i_1 \dots i_{l_1}}^{(l_1 m_1)} g_{i_1 \dots i_{l_1} k_1 \dots k_{l_2}}^{(l_1 l_2)} t_{k_1 \dots k_{l_2}}^{(l_2 m_2)}, \quad g_{m_1 m_2}^{(l_1 l_2)} = \sum_i C_{i m_1 m_2}^{l_1 l_2} g_m^{(l_1 l_2)}. \quad (2.8)$$

Formulas (2.8) and (2.7) give the desired relation between  $g^{(l_1 l_2)}$  and  $\bar{g}^{(l_1 l_2)}$ .

Together with the generalized coefficients of friction, the tensors  $g^{(l)}(l_1 l_2)$  are invariant with respect to the group  $G$ . By the definition of these quantities, they are unchanged under spatial inversion ( $g^{(l)}(l_1 l_2)$  is a pseudo-tensor for odd  $l$ ); therefore all operators of the group  $G$  in their action on  $g^{(l)}$  reduce to rotations. Simply analysis enables us to separate the following possibilities:

**A. Particles of spherical type.** The group  $G$  contains at least two noncoincident axes of symmetry of  $n$ th order ( $n > 2$ ). In this case  $g^{(l)}(l_1 l_2) = 0$  ( $l \neq 0$ ).

**B. Particles of the solid-of-revolution type.** The group  $G$  contains a two-sided axis of  $n$ th order ( $n > 2$ ). In this case only the values of  $g^{(l)}(l_1 l_2)$  with odd  $l$  vanish.

**C. In all remaining cases** the tensors of even and of odd weight  $l$  are different from zero.

### 3. ROTATIONAL DIFFUSION

In order to calculate the averages over angles in (2.4), it is necessary to know the distribution function  $W(\alpha, t)$ . The product  $W d\alpha$  ( $d\alpha = \sin \theta d\theta d\varphi d\psi$ ) has the meaning: the probability that the direction of the  $z$  axis of the coordinate system  $\bar{S}$  (see above) lies in the element of solid angle  $\sin \theta d\theta d\varphi$  and the direction of the  $x$  axis in the element of angle  $d\psi$ .

As is well known [10], the function  $W$  satisfies the equation of rotational diffusion

$$\frac{\partial W}{\partial t} + i(\hat{R}\omega)W = 0, \quad (3.1)$$

where the Hermitian operator of infinitely small rotation  $R$  can be expressed in terms of derivatives with

respect to the Euler angles, while the angular velocity  $\omega$  of the particle is given by the expression (2.3), in which the torque  $\mathbf{K}$  due to external forces is determined by the sum of the moments of the magnetic and the random forces:

$$\mathbf{K} = kT \{ [e\hat{\xi}] - i\hat{\mathbf{R}} \ln W \}, \quad e = \mu/\mu, \quad (3.2)^*$$

On substituting (2.3) into (3.1) and using (3.2), we get

$$\frac{\partial W}{\partial t} + i\hat{\mathbf{R}}_i \left\{ \Omega_i^{(1)} + g_{i,k}^{(1)} \Omega_k^{(2)} + \frac{1}{\tau} g_{i,k}^{(1)} ([e\hat{\xi}]_k - i\hat{\mathbf{R}}_k) \right\} W = 0, \quad (3.3)$$

where the constant  $\tau = \eta_0 V / kT$  and coincides in order of magnitude with the Brownian time of rotational diffusion.

In a quiescent suspension, the normalized stationary solution of (3.3) is the usual Boltzmann distribution:

$$W_0 = (\xi/8\pi^2 \text{sh } \xi) \exp(e\hat{\xi}). \quad (3.4)$$

As is evident from (3.3), the relaxation time of  $W(t)$  to  $W_0$  agrees in order of magnitude with the inverse coefficient of rotational diffusion  $g_{i,k}^{(1)}/\tau \sim g/\tau$ , where  $g$  is a multiplier dependent on the shape of the particles. On taking  $g \sim 10^{-1}$ , the volume of a single particle  $V \sim 10^{-18} \text{ cm}^3$ ,  $\eta_0 \sim 10^{-2} \text{ g/cm sec}$ , and  $kT \sim 4 \times 10^{-14} \text{ erg}$ , we get as an estimate of the Brownian relaxation time  $\tau/g \sim 10^{-5} \text{ sec}$ . This time is small, first in comparison with the hydrodynamic times  $\rho l^2/\eta_0$  ( $l$  is the hydrodynamic scale of length), and second in comparison with the inverse gradient of the hydrodynamic velocity. The first fact permits us, in the calculation of the viscosity, to use the stationary solution of (3.3), while consideration of the second makes it possible to restrict ourselves to an approximation linear with respect to the velocity gradient.

We write the stationary distribution function in a moving suspension in the form

$$W = W_0(1 + \chi), \quad \chi = \tau \sum_{im} \chi_m^{(i)} \Omega_m^{(i)*}, \quad (3.5)$$

$$\langle \chi \rangle_0 = \int \chi W_0 d\alpha = 0.$$

By substituting (3.5) into (3.3) one can obtain inhomogeneous equations for the function  $\chi_{im}^{(l)}$ :

$$N_m^{(1)} + \hat{I} \chi_m^{(1)} = 0, \quad \hat{I} = W_0^{-1} \hat{\mathbf{R}}_i W_0 g_{i,k}^{(1)} \hat{\mathbf{R}}_k, \quad (3.6)$$

$$N^{(1)} = W_0^{-1} i\hat{\mathbf{R}} W_0 = [e\hat{\xi}], \quad N_{ik}^{(2)} = W_0^{-1} i\hat{\mathbf{R}}_i g_{i,k}^{(2)} W_0.$$

On taking into account the hermiticity of the operator  $\hat{\mathbf{R}}_i$ , we have for arbitrary functions  $\chi$  and  $\zeta$

$$\langle \chi \hat{\mathbf{R}} \zeta \rangle_0 = -\langle g_{i,k} (\hat{\mathbf{R}}_i \chi) (\hat{\mathbf{R}}_k \zeta) \rangle_0,$$

whence, on noting the symmetry of the dimensionless diffusion tensor  $g^{(1)}$  (see (2.5)), we find that the "collision operator"  $\hat{I}$  has the property  $\langle \hat{I} \chi \zeta \rangle_0 = \langle \zeta \hat{I} \chi \rangle_0$ . This enables us to prove the equality

$$\langle N_m^{(1)} \chi_m^{(1)} \rangle_0 = \langle \chi_m^{(1)} N_m^{(1)} \rangle_0. \quad (3.7)$$

In order to prove (3.7) it is necessary to multiply equation (3.6) for  $l = l_1$  and  $l = l_2$  by  $\chi^{(l_2)}$  and  $\chi^{(l_1)}$  respectively and to subtract one from the other, using the symmetry property of the operator  $\hat{I}$  indicated above.

On noting that  $\mathbf{K} = -kT i\hat{\mathbf{R}} \chi$  and allowing for (2.5) and (3.5), one can rewrite the relations (2.4) in the form (1.4), where the viscosity tensor is determined by the expression

$$\eta_{m_1 m_2}^{(l_1 l_2)} = \eta_0 \Phi \{ -\langle N_{m_1}^{(l_1)} \chi_{m_1}^{(l_1)} \rangle_0 + k_{l_1 l_2} \langle g_{m_1 m_2}^{(l_1 l_2)} \rangle_0 \}. \quad (3.8)$$

Here  $N^{(0)} = g_{i_1}^{(1)} (01) N_{i_1}^{(1)}$ ; the coefficient  $k_{l_1 l_2}$  is zero if

$l_1$  or  $l_2$  has an odd value, and otherwise  $k_{l_1 l_2} = 1$ .

From (2.5), (3.7), and (3.8) it is easily seen that the viscosity tensor is symmetric:  $\eta_{m_1 m_2}^{(l_1 l_2)} = \eta_{m_2 m_1}^{(l_2 l_1)}$ ; this automatically guarantees fulfillment of Onsager's principle. The latter fact requires explanation. As is well known, in the presence of a magnetic field Onsager's relations have the form

$$\eta^{(i_1 i_2)}(\mu, \mathbf{H}) = \eta^{(i_1 i_2)}(-\mu, -\mathbf{H}).$$

But because of the fact that the field dependence of the quantities  $N^{(l_1)}$  and  $\chi^{(l_2)}$  (and along with them of the viscosity tensor) occurs only through the function  $W_0$  (see (3.6)), which does not change with reversal of sign of the field and of the magnetization, Onsager's principle can be formulated here in simpler form (without the substitutions  $\mu \rightarrow -\mu$  and  $\mathbf{H} \rightarrow -\mathbf{H}$ ).

By using the symmetry of the viscosity tensor and the permutation properties of the Clebsch-Gordan coefficients, we have from (1.5)  $\eta(l_1 l_2) = (-1)^{l_1 + l_2} \eta(l_2 l_1)$ , so that for  $l_1 = l_2$  and for odd  $l$  the reduced viscosities vanish; that is, coefficients odd in the field can relate only fluxes and forces of different tensorial character. Furthermore, since  $N^{(1)} \perp \mathbf{h}$ , it can be seen that the corresponding solution  $\chi^{(1)}$  of equation (3.6) is also perpendicular to the field. This means that the role of thermodynamic force is actually played not by the angular velocity itself, but only by its projection  $\Omega_{\perp}^{(1)} = \Omega^{(1)} - \mathbf{h}(\Omega^{(1)} \cdot \mathbf{h})$  on a plane perpendicular to  $\mathbf{H}$ . The same applies also to the flux  $\sigma^{(1)}$ , as can be seen from (1.4) and (3.8). One can arrive at this conclusion also from physical considerations: for  $\mathbf{H} \parallel \Omega^{(1)}$  no additional dissipation mechanism operates as a result of the orienting effect of the field on the particle (see Sec. 1). Hence it follows that  $\eta(101) = 0$ , whereas the viscosities  $\eta(211)$  and  $\eta(312)$  are expressed linearly in terms of  $\eta(011)$  and  $\eta(112)$  respectively. Thus it turns out that in the general case the viscosity tensor possesses seven independent coefficients. Returning to Cartesian coordinates, we can write the stress tensor (1.4) in the form

$$\sigma^{(0)} = -p + 3\beta h_i h_k \Omega_{ik}^{(2)},$$

$$\sigma_{i_1}^{(1)} = 4\eta_{R\Omega_{\perp}^{(1)}} + 4[\gamma_1 e_{ik} h_k h_m + \gamma_2 (h_i h_m - \delta_{im}) h_l] \Omega_{lm}^{(2)},$$

$$\sigma_{ik}^{(2)} = 2[\gamma_1 (h_i e_{km} + h_k e_{im}) h_m - \gamma_2 (h_i \delta_{kl} + h_k \delta_{li})] \Omega_{li}^{(1)} + 2(2\eta_2 - \eta_1) \Omega_{ik}^{(2)} + 2[(\eta_2 - \eta_1) h_l h_m \delta_{ik} + (\eta_1 + \eta_2 - 2\eta_2) (h_i \delta_{mk} + h_k \delta_{im}) h_l + (\eta_1 + \eta_2 - 2\eta_2) h_i h_k h_l h_m] \Omega_{lm}^{(2)}. \quad (3.9)$$

The independent viscosity coefficients  $\beta$ ,  $\eta_R$ ,  $\gamma_1$ ,  $\gamma_2$ ,  $\eta_1$ ,  $\eta_2$ , and  $\eta_3$  introduced here are expressed linearly in terms of the reduced viscosities  $\eta(l_1 l_2)$ . The quantity  $\eta_R$  (proportional to  $\eta(011)$ ) relates the asymmetric parts of the stress tensor ( $\sigma^{(1)}$ ) and of the velocity-gradient tensor ( $\Omega^{(1)}$ ) and therefore can be called the rotational viscosity of the suspension. The coefficient  $\beta$  ( $\sim \eta(202)$ ) is a cross-coefficient between shear and volume effects of viscous friction,  $\gamma_1$  and  $\gamma_2$  (proportional to  $\eta(212)$  and to  $\eta(112)$  respectively) between rotational and shear. Here  $\gamma_1$  is even in the field,  $\gamma_2$  odd. The shear viscosities  $\eta_1$ ,  $\eta_2$ , and  $\eta_3$  coincide with those introduced in [11] and are linear combinations of the quantities  $\eta(l22)$  with even  $l$  ( $l = 0, 2, 4$ ).

#### 4. CALCULATION OF VISCOSITY COEFFICIENTS

In order to calculate the viscosity tensor (3.8), it is necessary to find the "nonequilibrium" corrections to

the distribution function  $W_0$ ; they must satisfy the kinetic equation (3.6). If the suspended particles possess symmetry of type B (cf. Sec. 2), then in the expansion (2.8) of the tensor  $g^{(12)}$ , which determines the inhomogeneity  $N^{(2)}$ ,  $g^{(1)}$  (12) and  $g^{(3)}$  (12) are absent, but  $g_m^{(2)}$  (12) =  $g(212)Y_{2m}(\mathbf{n})$  ( $\mathbf{n}$  is a unit vector directed along the axis of symmetry). In this case it is appropriate to seek a solution of Equation (3.6), with  $l = 2$ , in the form

$$\chi_m^{(2)} = \sum_{m'} \bar{a}_m^{(2)} \{D_{m'm}^{(2)}(\alpha) - \langle D_{m'm}^{(2)}(\alpha) \rangle_0\}. \quad (4.1)$$

We substitute this function in (3.8) and find that in this case the coefficient odd in the field,  $\eta(112)$ , vanishes. For this purpose we note that the cross part of the viscosity tensor  $\eta^{(12)}$  can be rewritten in the form

$$\frac{1}{\eta_0\Phi} \eta_{m'm}^{(12)} = - \langle N_{m'}^{(1)} \chi_m^{(2)} \rangle_0 = \langle i\hat{R}_{m'}^{(1)} \chi_m^{(2)} \rangle_0 \quad (4.2)$$

(here  $\hat{R}_m^{(1)}$  is a spherical component of the vector operator  $\hat{R}$ ), and the mean is

$$\langle D_{m'm}^{(2)}(\alpha) \rangle_0 = \frac{4\pi}{2l+1} (-1)^m L_l(\xi) \bar{Y}_{lm'}(\mathbf{e}) Y_{lm}(\mathbf{h}), \quad (4.3)$$

$$L_l(\xi) = \langle P_l(\mathbf{e}\mathbf{h}) \rangle_0 = I_{l+\frac{1}{2}}(\xi) / I_{\frac{1}{2}}(\xi), \quad (4.4)$$

$$L_l(\xi) = \frac{\xi^l}{(2l+1)!} \left(1 - \frac{l}{3(2l+3)} \xi^2 + \dots\right) \quad (\xi \ll 1),$$

$$L_l(\xi) = 1 + O(\xi^{-1}) \quad (\xi \gg 1),$$

where  $P_l(x)$  is a Legendre polynomial and  $I_{l+1/2}(x)$  is a modified Bessel function of half-integral index (the bar over the tensor  $Y_{lm}(\mathbf{e})$  means, as usual (see Sec. 2), that we are taking its constant value in the system  $\bar{S}$ ). As is well known, the operator  $\hat{R}$  when it acts on a function  $D_{m'm}^{(l)}(\alpha)$  interchanges only its components with different values of the index  $m$ , leaving unchanged its tensorial character  $l$ . According to this principle, on substitution of (4.1) in (4.2)  $\eta^{(12)}$  is expressed in terms of the mean of  $D_{m'm}^{(2)}$  and contains only the even function  $Y_{2m}(\mathbf{h})$ . Thus we arrive at the important conclusion that for particles with symmetry of type B, viscosity  $\gamma_2$  odd in the field is absent.

The vanishing of some of the reduced coefficients of viscosity can be understood also from the following considerations. As is evident from formulas (3.4) and (3.6),  $\mathbf{e}$  and  $\mathbf{h}$  appear simultaneously in all expressions. In the end this leads to the result that a kinetic coefficient  $\eta(l_1 l_2)$  with given  $l$  (see (1.5)) is proportional to  $\sum_{\bar{m}} \bar{p}_{\bar{m}}^{(l)} \bar{Y}_{l\bar{m}}(\mathbf{e})$ , where the tensor  $\bar{p}_{\bar{m}}^{(l)}$  is determined solely by the shape of the particle. It is clear that if the symmetry group of the particle does not allow the existence of such a tensor, having the corresponding rank and parity, then this kinetic coefficient vanishes.

Now let the suspended particles have symmetry of type A. In this case  $N^{(2)} = 0$ , which leads to  $\chi^{(2)} = 0$ , so that for such particles there remain only two independent coefficients:  $\eta(011)$  and  $\eta(022)$ , corresponding to rotational and shear viscosities; and the stress tensor, just as for spherical particles, takes the form

$$\sigma^{(0)} = -p, \quad \sigma_i^{(1)} = 4\eta_R \Omega_{\perp i}^{(1)}, \quad \sigma_{ik}^{(2)} = 2\eta \Omega_{ik}^{(2)}. \quad (4.5)$$

The exact function  $\chi^{(1)}$  (and in the general case  $\chi^{(2)}$ ) can be described only in the form of an infinite series in the generalized spherical functions  $D_{m'm}^{(l)}(\alpha)$ . For our purposes an approximate solution of equations (3.6) is sufficient. It is convenient to apply a variational method similar to that used in the kinetic theory of

gases<sup>[12]</sup>. We shall seek a solution of equations (3.6) in the form of finite truncations of series in the functions  $D_{m'm}^{(l)}$ . In the simplest approximation we have

$$\chi_m^{(1)} = \sum C_{l_1 m_1 m}^{l m} Y_{l_1 m_1}(\mathbf{h}) \bar{a}_m^{(1)}(l_1, \xi) \{D_{m_1 m}^{(1)} - \langle D_{m_1 m}^{(1)} \rangle_0\}. \quad (4.6)$$

The unknown coefficients  $\bar{a}_1^{(1)}(l_1, \xi)$  (hereafter we shall denote them by  $\bar{a}_1^{(l)}$ ), in accordance with the usual variational procedure<sup>[12]</sup>, are found by solution of the system of linear algebraic equations

$$I_{ik}^{(0)} a_k^{(l-1)} = N_i^{(l-1)}, \quad I_{ik}^{(2)} a_k^{(l)} - I_{ik}^{(4)} a_k^{(l+1)} = N_i^{(l)}, \quad (4.7)$$

$$I_{ik}^{(1)} a_k^{(l+1)} + I_{ik}^{(3)} a_k^{(l+1)} = N_i^{(l+1)},$$

where  $l = 1, 2$  and the following notation has been introduced:

$$I_{ik}^{(0)} = e_{ip} g_{i,m}^{(11)} \left( \frac{L_1}{\xi} \delta_{pq} + L_2 e_p e_q \right) e_{qmk}, \quad I_{ik}^{(1)} = \frac{L_1}{2} e_{ikl} g_{i,R}^{(11)} e_R,$$

$$I_{ik}^{(2)} = \frac{1}{2} e_{ipl} g_{i,m}^{(11)} \left[ \left(1 - \frac{L_1}{\xi}\right) \delta_{pq} - L_2 e_p e_q \right] e_{qmk};$$

$$N_i^{(10)} = N_i^{(12)} = 0, \quad N_i^{(11)} = e_i L_1,$$

$$N_i^{(21)} = \frac{1}{2} e_{iri} g_{i,pq}^{(12)} e_q \left( L_3 e_i e_q + \frac{L_2}{\xi} \delta_{rp} \right), \quad (4.8)$$

$$N_i^{(22)} = \frac{1}{2} L_2 (e_i g_{r,rp}^{(12)} - g_{p,ir}^{(12)} e_r) e_p,$$

$$N_i^{(23)} = \frac{1}{10} e_{iri} g_{i,pq}^{(12)} e_q [10L_3 e_i e_q + (2L_3 - 3L_1) \delta_{rp}].$$

In (4.7) and (4.8), for simplicity of writing, the bar indicating transition to the system  $\bar{S}$  has been omitted over the tensors; the functions  $L_n = L_n(\xi)$  are defined in (4.4).

On substituting (4.6) in (3.8) and using formulas (2.7), (2.8), and (4.3), we find

$$\beta = \eta_0 \Phi (L_1 e_{ik} g_i^{(1)}(0) e_i a_k^{(21)} + \frac{1}{2} L_2 g_{ik}^{(2)}(0) e_i e_k),$$

$$\eta_n = \frac{1}{2} \eta_0 \Phi L_1 a_i^{(11)} e_i, \quad (4.9)$$

$$\gamma_1 = -\frac{1}{2} \eta_0 \Phi L_1 a_i^{(22)} e_i, \quad \gamma_2 = -\frac{1}{2} \eta_0 \Phi L_1 a_i^{(22)} e_i,$$

in the coefficients  $\eta_n$  ( $n = 1, 2, 3$ ) it is convenient to separate the part that is independent of the field:

$$\eta_n = \eta_0 + \Delta \eta_0 + \Delta \eta_n, \quad \Delta \eta_0 = \eta_0 \Phi g^{(0)},$$

$$\Delta \eta_1 = 2\eta_0 \Phi (N_i^{(21)} a_i^{(21)} + L_2 g^{(2)} + \frac{1}{2} L_1 g^{(4)}), \quad (4.10)$$

$$\Delta \eta_2 = \eta_0 \Phi (N_i^{(21)} a_i^{(21)} + \frac{1}{2} L_1 g^{(4)}),$$

$$\Delta \eta_3 = \eta_0 \Phi (N_i^{(23)} a_i^{(23)} - \frac{1}{2} N_i^{(22)} a_i^{(22)} + L_2 g^{(2)} - L_1 g^{(4)}).$$

Here

$$g^{(0)} = \frac{1}{3} g_{ik,ik}^{(22)}, \quad g^{(2)} = \frac{1}{7} (3g_{ij,ij}^{(22)} e_i e_k - 5g^{(0)}),$$

$$g^{(4)} = g_{ik,lm}^{(22)} e_i e_k e_l e_m - 2(g^{(2)} + g^{(0)}).$$

The solution of the system (4.7) has a simple form if the suspended particles are magnetized along a direction that coincides with one of the principal axes of the diffusion tensor  $g^{(11)}$ . On denoting the principal values of the tensor  $g^{(11)}$  by  $g_1, g_2, g_3$  (the last corresponds to the direction of magnetization), we have, for example,

$$\eta_n = \frac{1}{2} \eta_0 \Phi \frac{L_1^2 \xi^2}{(\xi - L_1)(g_1 + g_2)}, \quad (4.11)$$

$$\gamma_1 = -\eta_0 \Phi \frac{L_1 \xi N_i^{(23)} e_i}{(\xi - L_1)(g_1 + g_2)}, \quad \gamma_2 = -\eta_0 \Phi \frac{L_1 \xi N_i^{(22)} e_i}{(\xi - L_1)(g_1 + g_2)}.$$

As can be seen from (4.7)–(4.11) and (4.4), all the viscosity coefficients saturate at large fields. From a physical point of view, this is explained by the fact that in a strong field the magnetic moments of all the particles are oriented along the field direction, and the flow

around the particles encounters maximum obstruction.

In the absence of a field, all the viscosity coefficients (except  $\eta(022)$ ) vanish, and the stress tensor takes the form (4.5) with  $\eta = \eta_0 + \Delta\eta_0$  and  $\eta_R = 0$ . It should be noted that the correction to the shear viscosity  $\Delta\eta_0$  can be calculated exactly if one takes into account that at  $H = 0$  the exact solution of equation (3.6) with  $l = 2$  has the form (4.1):

$$\begin{aligned} \Delta\eta_0 &= \eta_0 \varphi (g^{(0)-2} / g_{ik}^{(2)}(12) a_{ik}^{(2)}), \\ g_{ik}^{(2)}(12) &= e_{i,r} g_{r,ik}^{(2)} - 1/2 e_{ik} g_{r,rs}^{(2)}, \end{aligned} \quad (4.12)$$

where the irreducible tensor of second rank  $a^{(2)}$  is determined by solution of the equation

$$p_{ik}^{(2)}(12) = g_{ik}^{(2)}(12), \quad p_{i,ki}^{(2)} = g_{i,r}^{(2)} (e_{rsk} a_{rs}^{(2)} + e_{rsi} a_{rk}^{(2)}),$$

here  $p^{(2)}(12)$  is expressed in terms of  $p^{(12)}$  just as  $g^{(2)}(12)$  is expressed in terms of  $g^{(12)}$ .

## 5. DISCUSSION OF RESULTS

Formulas (4.7) enable us, for given generalized coefficients of friction, to find the independent viscosity coefficients of a suspension of ferromagnetic particles. A calculation of the viscosity for the case of particles in the form of ellipsoids of revolution was carried out in a paper of the author<sup>[13]</sup> (in the classification adopted here, such particles belong to type B, and for them the coefficients  $\gamma_2$  odd in the field vanish).

We turn now to the important case in which the suspended particles are nearly spherical. Let the form of the surface of the particles be given by the equation

$$r = r_0 [1 + \epsilon f(\theta, \varphi)], \quad f(\theta, \varphi) = \sum_{lm} f_m^{(l)} Y_{lm}(\theta, \varphi), \quad (5.1)$$

where  $\epsilon$  is a small asphericity parameter and  $f(\theta, \varphi)$  is an arbitrary function. In the approximation linear in  $\epsilon$ , the tensors  $g_m^{(l)}(l_1 l_2)$  ( $l \neq 0$ ) are proportional to the coefficients  $f_m^{(l)}$ . Since among these quantities there are no pseudotensors of odd rank, in this approximation the viscosity coefficients odd in the field vanish. In the next approximation, the tensors  $g_m^{(l)}$  are combinations of quantities  $\sum C_{l_1 m_1 l_2 m_2}^{lm} f_{m_1}^{(l_1)} f_{m_2}^{(l_2)}$  and an odd coefficient may turn out to be different from zero.

We pass on now to an explanation of the role of these viscosity coefficients in the hydrodynamic flow of the suspension. The equation of motion (1.2) with allowance for (3.9) can be written in the form

$$\rho \frac{dv_i}{dt} = - \frac{\partial p}{\partial x_i} + \hat{T}_{ik} v_k, \quad (5.2)$$

where the operator  $\hat{T}_{ik}$ , by virtue of the symmetry of the viscosity tensor (see Sec. 3), is symmetric in the indices  $i$  and  $k$ . The action of this operator (in its tensor part) reduces basically to similarity transformations and rotations through certain angles of the coordinates ( $r \rightarrow r'$ ) and the velocities ( $v \rightarrow v'$ ), different for each of these quantities. The parameters of

these transformations are determined by the field and the viscosity coefficients. For certain simple flows,  $\hat{T}_{ik}$  can be reduced to the usual form  $\eta' \delta_{ik} \Delta'$ <sup>[14]</sup>, with corresponding change of the boundary conditions.

It is well known that a dependence of the viscosity on the field is observed also in molecular gases (the Senteleben-Beenakker effect<sup>[8,12,15]</sup>). But in a paramagnetic gas the coefficients  $\eta_R$ ,  $\gamma_1$ , and  $\gamma_2$  are small (since they contain high powers of the nonlocality parameter (cf. <sup>[8]</sup>)) and are usually neglected. On the other hand, in a gas it is necessary to allow for a connection between the magnetization and rotation of the molecules (unimportant for the case of a suspension), and this leads to the occurrence of shear-viscosity coefficients  $\eta_4$  and  $\eta_5$  odd in the field<sup>[8,11,12]</sup>. Thus a ferromagnetic suspension in a magnetic field behaves similarly to a paramagnetic gas (the differences indicated above are unimportant and of purely quantitative nature).

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$$*[e\xi] \equiv e \times \xi.$$

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