The bulk viscosity of a ferromagnetic suspension

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The bulk viscosity of a ferromagnetic suspension is calculated as a function of the magnetic field strength and the sound frequency. It is assumed that the orientational Brownian motion of the suspended particles is significant but that the inertial effects due to the flow around the particles are small. It is shown that field-strength dependence of the bulk viscosity appears only in the presence of stereoisomerism of the shape of the particles (the symmetry group must admit of a pseudovector). The viscosity increases as the square of the field strength in weak fields and reaches saturation in strong fields. The viscosity dispersion is due to orientational relaxation of the particles (characteristic relaxation times, $\sim 10^{-6}$ sec). The viscosity of a suspension of particles, each of which consists of two slightly deformed spheres joined together, is calculated as an example.

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1. INTRODUCTION

The hydrodynamic flow of a suspension of solid particles in a liquid is accompanied by processes involving viscous friction owing to the flow of the liquid around the particles, and these processes lead to an increase in the effective viscosity. When the number density of the suspended particles is small the additional viscosity is proportional to the volume concentration φ of the particles, and the proportionality constant depends only on the shape of the particles. The viscosity of an incompressible liquid is described by a single coefficient, the shear viscosity η. As is well known, the correction to the shear viscosity for a suspension of spherical particles was calculated by Einstein (cf. Ref. 1, §22) and is equal to $2.5\eta_0\varphi$ (here and below a subscript zero will be used to designate quantities pertaining to the suspending liquid). In the general case of a compressible isotropic medium, the viscosity is characterized by still another coefficient, the bulk viscosity ζ, which is also changed by the presence of suspended particles in the liquid. Brenner[2] has calculated the bulk viscosity of a suspension of spherical particles:

$$\zeta=\zeta_0+\varphi(\zeta_0+{}^4/_3\eta_0). \tag{1.1}$$

According to the theory of Mandel'shtam and Leontovich (see, e.g., Ref. 1, §78), the appearance of a second viscosity is always due to relaxation processes going on in the system. Here the viscous flow around the particle plays the part of the relaxation process. However, the viscosity dispersion associated with this process in a ferromagnetic suspension may be neglected because the inertial effects are small (see Sec. 2 below). Nevertheless there is another relaxation mechanism in a ferromagnetic suspension that does lead to viscosity dispersion.

If the particles of a suspension are magnetized, an external magnetic field H will tend to orient the particles in the stream, and this gives rise to additional energy dissipation due to the flow of liquid around the particles and leads to a magnetic-field dependence of the viscosity (cf. Refs. 3 and 4). It is clear that a magnetic field will affect the bulk viscosity of a suspension only if a uniform (hydrostatic) compression of the liquid causes the particles to rotate, and this is possible only for particles having symmetry of a definite type (the symmetry group of the particle must admit of a pseudovector). The rotational motion of the particles is accompanied by thermal fluctuations with a characteristic relaxation time τ_0 of the order of 10^{-6} sec, so bulk-viscosity dispersion due to this relaxation process should be observed at frequencies ω of the order of $1/\tau_0$. We emphasize that orientational relaxation would not affect the bulk viscosity at all in the absence of a field: without the field to hinder their motion, the particles would rotate freely in the liquid stream. Thus, when the shape of the particles is given, the problem reduces to the calculation of the bulk viscosity of the suspension as a function of three variables: the frequency of the liquids's motions, the magnetic field strength, and the temperature.

Actually, the viscosity dispersion of a suspension can manifest itself only in the propagation of sound. In a sound wave, however, each particle not only suffers hydrostatic compression, but there is also a symmetric stream of liquid flowing around it which also tends to orient it (for example, rod-shaped particles will tend to be oriented along the propagation direction of a plane wave). In other words, not only the bulk viscosity, but also other viscosity coefficients of a ferromagnetic suspension,[4] which also are frequency dependent because of the relaxation of rotational degrees of freedom, may also play a part in the absorption of sound in a magnetic field. Unlike the effects of the bulk viscosity, however, the effects now being discussed depend essentially on the orientation of the magnetic field with respect to the propagation direction of the wave, and this permits us to refrain from discussing them in what follows.

2. A PARTICLE IN A COMPRESSIBLE FLOW

As an auxiliary problem let us further consider the effect on a flow having a constant velocity gradient of a single particle immersed in the liquid. The calculation of the bulk viscosity of a suspension differs from the calculation of the shear viscosity (Ref. 1, §22) in that

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one must assume that the flow has a nonvanishing velocity divergence at large distances from the particle:

$$v=\tfrac{1}{3}\Omega r, \quad r\to\infty, \tag{2.1}$$

where $\mathbf{r}$ is the distance from the center of the particle and $\Omega=\operatorname{div}\mathbf{v}$ is a constant.

Of course if $\operatorname{div}\mathbf{v}\neq 0$ one cannot assume that ρ is constant, since then it would not be possible to satisfy the equation of continuity:

$$\partial\rho/\partial t+\operatorname{div}\rho\mathbf{v}=0. \tag{2.2}$$

In that case one must also take the equation of motion of the liquid in the form

$$\rho d\mathbf{v}/dt=-\nabla p+\eta_0\Delta\mathbf{v}+(\zeta_0+\tfrac{1}{3}\eta_0)\nabla(\nabla\mathbf{v}) \tag{2.3}$$

with the boundary condition

$$\mathbf{v}=\mathbf{u}+[\boldsymbol{\omega}\times\mathbf{r}]; \tag{2.4}$$

at the surface s of the particle, where $\mathbf{u}$ and $\boldsymbol{\omega}$ are the velocity and angular velocity of the particle.

It is easily seen, however, that for all reasonable hydrodynamic processes the inertial terms in expression (2.3) for $d\mathbf{v}/dt$ are small as compared with the viscous terms, which are of the order of v/τ', where $\tau'=a^2\rho/\eta_0$, a being a linear dimension of the particle. In fact, if we take $a\sim 10^{-6}$ cm and $\eta_0/\rho\sim 10^{-2}$ cm^2/sec, we find $\tau'\sim 10^{-10}$ sec. As was noted in the Introduction, the fact that τ' is small also means here that the viscous friction associated with flow around the particle (velocity relaxation) does not lead to viscosity dispersion at any acoustic frequencies.

Since the velocity can now be regarded as a function of the coordinates alone, the space and time variables can be separated in the equation of continuity (2.2), and if at the initial time there was a uniform distribution of liquid throughout a volume containing the particle, that distribution will remain uniform at all subsequent times. In other words, the density ρ is to be regarded as a function of time, but not of the coordinates. Actually, this assumption imposes a limitation on the velocity (2.1) of the liquid at large distances from the particles: this velocity must be small as compared with the velocity c of sound, so that a local variation of the density will be dissipated in a much shorter time than the time required for a significant change in the density: $\Omega R/c\ll 1$, where R is the radius of a large sphere centered at the particle. Taking $R\approx 10a$ (cf. Ref. 2), $a\sim 10^{-6}$ cm, and $c\sim 10^5$ cm/sec, we find that the velocity v of the liquid at large distances from the particle must be small enough for the condition $\Omega\ll 10^{10}$ sec^{-1} to be satisfied, which is virtually always the case.

When ρ is a function of t alone, i.e., is a constant as far as the coordinates are concerned, we find from Eq. (2.2) that divv is also a constant in that same sense. Having evaluated the latter constant from the boundary conditions (2.1), we can write the equations of motion of the liquid in the form

$$\nabla p=\eta_0\Delta\mathbf{v}, \quad \operatorname{div}\mathbf{v}=\Omega. \tag{2.5}$$

Knowing the solution of Eqs. (2.5) under the boundary conditions (2.1) and (2.4), we can calculate the force F_i and the couple L_i acting on the particle as a result of its presence in the flowing liquid, as well as the quantity S_{ik}/V, i.e., the stress tensor averaged over the volume V of the particle:

$$F_i=\oint\sigma_{ik}\,ds_k, \quad S_{ik}=\oint x_i\sigma_{kl}\,ds_l, \quad L_i=e_{ikl}S_{kl}, \tag{2.6}$$

$$\sigma_{ik}=-p\delta_{ik}+\eta_0\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}-\frac{2}{3}\frac{\partial v_l}{\partial x_l}\delta_{ik}\right)+\zeta_0\frac{\partial v_l}{\partial x_l}\delta_{ik}, \tag{2.7}$$

where the vector element ds_k of the surface of the particle is directed along the outward drawn normal. In writing Eqs. (2.6), the volume integral S_{ik} has been transformed to a surface integral, using the equation of motion (2.5) and the equality of action and reaction at the surface of the particle (this makes it unnecessary to consider the internal stresses in the particle). Because the equations of motion are linear, F_i and S_{ik} will be proportional to the boundary values of Ω, u_i, and ω_i, and the form of the proportionality constants (which of course are tensors—we may call their components the generalized coefficients of friction) will depend only on the shape of the surface of the particle (cf. Ref. 4).

In the presence of hydrodynamic forces, the translational and rotational motions of the particle will be determined by the equations

$$m\dot{u}_i=F_i, \quad J_{ik}\dot{\omega}_k=L_i+K_i, \tag{2.8}$$

where m and J_{ik} are the mass and the tensor of inertia of the particle, and K_i is the couple exerted on the particle by the external forces. The inertial terms on the left in Eqs. (2.8) are significant only at small times of the order of the viscous time τ' introduced above; hence we may neglect them, as we did in the equations of motion (2.3) for the liquid. Equating the right-hand sides of Eqs. (2.8) to zero and bearing in mind that F_i and L_i are linear in u_i, ω_i, and Ω, we can write the expressions for the velocity and angular velocity of the particle in which their proportionality to Ω and K_i are explicitly exhibited:

$$\omega_i=g_i\Omega+g_{ik}K_k/\eta_0V \tag{2.9}$$

(for brevity we shall not write down the analogous expression for u_i). Then eliminating u_i and ω_i from the stress tensor S_{ik}, we find the following expression for the quantity $S=(1/3)S_{kk}$ of interest to us:

$$S=\eta_0Vg\Omega+g_iK_i. \tag{2.10}$$

The scalar, pseudovector, and tensor coefficients g, g_i, and g_{ik} occurring in Eqs. (2.9) and (2.10) may be expressed in terms of the generalized coefficients of friction; moreover, it is precisely because of the symmetry of the latter (see Ref. 4) that K_i occurs in S with the same coefficients g_i as Ω occurs in ω_i, and that the tensor g_{ik} is symmetric: $g_{ik}=g_{ki}$.

It is important that the coefficients g, g_i, and g_{ik} are invariant under the same symmetry transformations as are the particles themselves. As will be evident later on, the bulk viscosity of the suspension can depend on

the magnetic field strength only if the symmetry group of the particles allows the existence of the pseudovector g_i. The point groups C_n, S_{2n}, C_{nh}, C_{2v}, and D_2 have the property. No ellipsoid has this kind of symmetry, nor has a slighly deformed sphere (in the first order in the asphericity parameter). A fairly simple and physically interesting case is that of a surface consisting of two slightly deformed spheres fastened together to form a dumbbell; such a surface may serve as a model for two particles of a ferromagnetic suspension that are adhering to one another. This example will be treated in more detail below.

We note that in addition to Eq. (2.10), we should write down analogous relations for the symmetric tracefree part of the tensor S_{ik}; in the final result, however, this part leads to viscosity coefficients of the suspension that have already been investigated,[4] so we shall not consider it here.

3. THE EFFECTIVE VISCOSITY OF A SUSPENSION

Now let us turn to the calculation of the viscosity of a suspension treated as a homogeneous continuous medium. To do this we must obviously average the microscopic stress tensor over a volume that contains many particles (for more details see Refs. 1, 2, and 5). The microscopic stresses in the suspension are made up of the stresses (2.7) in the pure liquid and the stresses (2.6) in the particles due to the motion of the liquid. When the particle concentration is low, the contributions from the individual particles turn out to be additive, and as a result the averaged stress tensor has the form

$$\bar{\sigma}_{ik}=-\bar{p}\delta_{ik}+\eta_0\left(\overline{\frac{\partial v_i}{\partial x_k}}+\overline{\frac{\partial v_k}{\partial x_i}}\right)+\left(\zeta_0-\frac{2}{3}\eta_0\right)\overline{\frac{\partial v_l}{\partial x_l}}\delta_{ik}+N\langle S_{ik}\rangle, \quad (3.1)$$

where N is the number density of the particles and the angle brackets indicate averaging over the orientations of the particles. Since the tensor S_{ik} represents the internal stresses in a particle averaged over the particle's volume, the additional averaging over the angles ensures complete averaging over the ensemble of particles.

Limiting ourselves here to the calculation of the bulk viscosity of the suspension, we make use of Eq. (2.10) to rewrite the scalar part $\bar{\sigma}=(1/3)\bar{\sigma}_{kk}$ of the stress tensor (3.1) in the form

$$\bar{\sigma}=-\bar{p}+(\zeta_0+\varphi\eta_0 g)\Omega+N\langle g_i K_i\rangle, \quad (3.2)$$

where we have used the fact that $\varphi=NV$, as well as the notation

$$\Omega=\overline{\frac{\partial v_i}{\partial x_i}}=\frac{\partial\bar{v}_i}{\partial x_i}.$$

The effective bulk viscosity of the suspension should be defined as the proportionality constant between the viscous-stress tensor $\bar{\sigma}+\bar{p}$ and the quantity Ω (the velocity divergence of the suspension). As will be shown below, the last term in Eq. (3.2) is proportional to Ω; hence the bulk viscosity of the suspension can be written in the form

$$\zeta=\zeta_0+\Delta\zeta, \quad \Delta\zeta=\varphi(\eta_0 g+\langle g_i K_i\rangle/\Omega V). \quad (3.3)$$

As was already noted in Sec. 2, when $\mathrm{div}\,v\neq 0$ the density of the liquid cannot be regarded as constant in time, and it follows that none of the other thermodynamic quantities can be so regarded either. In particular, uniform compression results in heating of the liquid and therefore increases the effective stresses in the suspension. Because the thermal expansion coefficient of the liquid is very small, however, one can neglect effects of this sort. In fact, from the equation of continuity (2.2) and the relation (2.5) we find that

$$\partial\rho/\partial t=-\rho\Omega.$$

In the linear approximation in Ω the motion of the liquid may be regarded as adiabatic, i.e., we may assume that $\partial s'/\partial t=0$, where s' is the entropy per unit mass of the liquid. Treating the temperature as a function of the thermodynamic variables ρ and s' and forming its differential in these variables, we easily find that

$$\frac{\partial T}{\partial t}=-\left(\frac{\partial T}{\partial\rho}\right)_{s'}\frac{\partial\rho}{\partial t}=-\frac{c^2\beta T}{c_p}\Omega, \quad (3.4)$$

where β and c_p are the thermal expansion coefficient and the heat capacity per unit mass of the liquid, and c is the velocity of sound. If the volume occupied by a particle of the suspension were filled with liquid, then the rate of expansion of this volume due to heating of the liquid would be given by

$$\partial V/\partial t=\beta V\,\partial T/\partial t. \quad (3.5)$$

Neglecting the changes in the temperature and density of the particle for simplicity, we can effectively take the disturbance due to the particle into account by assuming that the volume of the particle changes in such a manner as to compensate the thermal expansion (3.5) of the liquid. For this it is necessary, according to Eq. (3.5), that the velocity of a surface element of the particle be of the order of $\beta a\,\partial T/\partial t$. At the same time, according to Eq. (2.1) the velocity of the undisturbed liquid flow at the surface of the particle is a Ω. Equating these two velocities and eliminating $\partial T/\partial t$ with Eq. (3.4), we obtain the condition for the effect under consideration to be negligible: $c^2\beta^2 T/c_p\ll 1$. For typical values of the parameters, say $c\sim 10^5$ cm/sec, $\beta\sim 10^{-4}$ deg^{-1}, $T\sim 300$ °K, and $c_p\sim 1$ cal/g·deg $\approx 4\times 10^7$ erg/g·deg, we find that $c^2\beta^2 T/c_p\sim 10^{-3}$, showing that the condition is well satisfied.

Thus, as is evident from formula (3.3), to calculate the viscosity it is necessary, first, to solve the hydrodynamic problem of the fluid flow around particles of the given shape (Sec. 2) and thereby to evaluate the coefficients g and g_i, and second, to know the distribution $W(\theta,\varphi,\psi)$ of the particles with respect to orientation (θ, φ, and ψ are Euler's angles) in order to perform the averaging necessary to evaluate $\langle g_i K_i\rangle$.

We shall assume that the magnetic moment μ of an individual particle is rigidly fixed to the particle (the frozen-in-dipole model[6]): $\mu=\mu\mathbf{e}$, where $\mathbf{e}$ is a unit vector which is constant in a coordinate system rigidly

fixed to the particle. We note that in atomic systems, on the other hand, the magnetic moments are not fixed to the particles themselves, but to their angular momenta (the relation $\mu = \mu e$ is replaced by the gyromagnetic relation). In the presence of a magnetic field $\mathbf{H}$, there is a couple $\mu \times \mathbf{H}$ acting on each particle. In addition, the small particles of a ferromagnetic suspension are subject thermal fluctuations, so the external forces on the particle must also include the couple $-kTi\hat{\mathbf{R}}\ln W$ due to the stochastic forces (here $i\hat{\mathbf{R}}$ is the operator for an infinitesimal rotation; the imaginary unit has been introduced to make $\hat{\mathbf{R}}$ Hermitian). Thus, the total couple acting on the particle is

$$\mathbf{K} = kT\{[\mathbf{e}\xi] - i\hat{\mathbf{R}}\ln W\}; \quad \xi = \mu H/kT. \tag{3.6}$$

The distribution function W satisfies the rotational-diffusion equation

$$\partial W/\partial t + i\hat{\mathbf{R}}\omega W = 0, \tag{3.7}$$

where the angular velocity ω of the particle is given by Eq. (2.9) with the external couple (3.6); thus, the kinetic equation (3.7) becomes

$$\tau_0 \partial W/\partial t + i\tau_0 \Omega g_i \hat{R}_i W + i\hat{R}_i g_{ik}([\mathbf{e}\xi]_k - i\hat{R}_k) W = 0, \quad \tau_0 = \eta_0 V/kT. \tag{3.8}$$

The characteristic time for changes in W is of the order of τ_0, which amounts to $\sim 10^{-6}$ sec at room temperature if $\eta_0 \sim 10^{-2}$ g/cm·sec and $V \sim 10^{-18}$ cm^3. Virtually all hydrodynamic flows may be assumed to satisfy the condition $\Omega\tau_0 \ll 1$, so we shall neglect higher order terms in $\Omega\tau_0$ and seek the solution to Eq. (3.8) in the form

$$W = W_0(1 + \tau_0 \Omega \chi), \tag{3.9}$$

where W_0 and χ are unknown functions of the angles and time.

Relaxation of the distribution function may lead to bulk-viscosity dispersion at frequencies $\omega \sim 1/\tau_0$. To examine this effect we shall assume that the velocity of the liquid surrounding the particle varies periodically according to the law $\Omega \sim \exp(i\omega t)$ (the use of the same letter to denote both the frequency of the sound wave and the angular-velocity vector of the particle cannot lead to confusion). Now we substitute the distribution function (3.9) into the kinetic equation (3.8). It is easily seen that during the short time τ_0 the solution to the equation will reach the regime of steady-state oscillations in which we shall be interested. In the absence of a hydrodynamic flow ($\Omega = 0$) the steady-state regime takes the form of an ordinary Boltzmann distribution:

$$W_0 = \frac{\xi}{8\pi^2 \operatorname{sh} \xi} e^{\mathbf{e}\xi}. \tag{3.10}$$

In the next approximation in $\Omega\tau_0$ one must, generally speaking, take into account the fact that distribution (3.10) is only locally an equilibrium distribution, since the temperature of the liquid, which occurs in it, varies with time in accordance with Eq. (3.4). Since the temperature occurs in W_0 only through Langevin argument $\xi = \mu H/kT$, however, its variation is equivalent to a weak ($\sim \Omega\tau_0$) longitudinal modulation of the magnetic field, which, in the final analysis, may contribute terms of higher order in $\Omega\tau_0$ to the stress tensor of the suspension.

The above discussion permits us to neglect time variation of the temperature of the medium, and after substituting (3.9) and (3.10) into (3.8) we obtain the following inhomogeneous equation for the nonequilibrium correction χ to the distribution function in the steady-state regime:

$$I\chi = P, \quad I = i\omega\tau_0 - W_0^{-1}\hat{R}_i W_0 g_{ik}\hat{R}_k, \quad P = \xi[\mathbf{ge}]. \tag{3.11}$$

Noting that the external couple (3.6) can be expressed in the form $K_i = -kT\Omega\tau_0 i\hat{R}_i\chi$ with the aid of Eqs. (3.9) and (3.10) and making use of the hermiticity of $\hat{R}_i$, we transform Eq. (3.3) for the bulk viscosity of the suspension to the form

$$\zeta = \zeta_0 + \eta_0\varphi(g + \langle P\chi\rangle_0), \tag{3.12}$$

where the subscript zero on the angle brackets indicates that the equilibrium distribution function (3.10) is to be used in averaging over the angles.

4. BULK-VISCOSITY DISPERSION

Formula (3.12), derived above, expresses the kinetic coefficient ζ in terms of the average value of the product $P\chi$ of the solution χ to the inhomogeneous kinetic equation (3.11) by the inhomogeneous term P of that equation. This makes it possible here, as in the kinetic theory of gases,[7] to formulate a variational principle and to use it to obtain an approximate solution to Eq. (3.11). We shall seek the unknown function χ as a linear combination of generalized spherical functions $D^{(l)}_{mm'}(\theta, \varphi, \psi)$, retaining only the functions with $l = 1$ in the simplest approximation (cf. Ref. 4). In this case, however, it is more convenient to express the $D^{(1)}_{mm'}$ in terms of the unit vectors n_i ($n = 1, 2, 3$) along the axes of a coordinate system fixed rigidly to the particle (the vectors n_i are the columns of a matrix that transforms a Cartesian vector under rotation of the coordinate system):

$$\chi = \sum_n a_n n_i h_i, \quad h_i = H_i/H, \tag{4.1}$$

where the coefficients a_n are to be found by the usual variational procedure: substituting (4.1) into (3.11), multiplying the equation by n_i', and averaging over the angles, we obtain the following set of linear algebraic equations for the a_n:

$$\sum_n a_n \langle n_k' \hat{I} n_i\rangle_0 h_i = \langle n_k' P\rangle_0. \tag{4.2}$$

The matrix element of the operator $\hat{I}$ can be calculated with the aid of averaging formulas given in Ref. 4:

$$h_i\langle n_k' I n_i\rangle_0 = I_{n'n}h_k,$$
$$I_{nm} = \frac{L_1}{\xi}(g_{nm} - g_{pp}\delta_{nm}) - L_2[\mathbf{ege}]_{nm} + i\omega\tau_0\left(\frac{L_1}{\xi}\delta_{nm} + L_2 e_n e_m\right). \tag{4.3}$$
$$[\mathbf{ege}]_{nm} = e_{npq}e_{mst}g_{ps}e_q e_t, \quad L_1 = \operatorname{cth}\xi - \frac{1}{\xi}, \quad L_2 = 1 - 3\frac{L_1}{\xi}.$$

The right-hand side of Eq. (4.2) can be calculated in a similar manner:

$$\langle n_k P\rangle_0 = P_n h_k, \quad P_n = L_1 n[ge]. \tag{4.4}$$

After substituting Eqs. (4.3) and (4.4) into (4.2), the equations for the a_n reduce to

$$I_{nm} a_m = P_n. \tag{4.5}$$

For an arbitrary orientation of the magnetic moment of the particle (specified by the vector e_i) with respect to the principal axes of the rotational-diffusion tensor g_{ik} the solution to Eq. (4.5) has a complicated form, so we shall consider only the simplest case and assume that e_i has the direction of one of the principal axes of the tensor g_{ik} and that this axis coincides with the z axis of a coordinate system rigidly fixed to the particle. Under this condition, the matrix I_{nm} is diagonalized in the system whose axes are the principal axes of the tensor g_{ik}, and the projections of the vector a_n onto these axes have the form

$$\begin{aligned} a_1 &= -\xi g_2 \Big/ \left[\left(1+\frac{\xi L_2}{L_1}\right) g^{(2)} + g^{(3)} - i\omega\tau_0\right], \\ a_2 &= \xi g_1 \Big/ \left[\left(1+\frac{\xi L_2}{L_1}\right) g^{(1)} + g^{(3)} - i\omega\tau_0\right], \quad a_3 = 0. \end{aligned} \tag{4.6}$$

where the $g^{(n)}$ are the principal values of the diffusion tensor, while the g_n are the components of the vector g in the chosen coordinate system.

By substituting the function χ in the form (4.2) into formula (3.12), we can express the viscosity of the suspension in terms of the coefficients a_n:

$$\Delta\zeta = \eta_0\varphi\{g + L_1 e[ga]\}. \tag{4.7}$$

Now using formula (4.6) and assuming for simplicity that the principal values of the tensor g_{ik} along the x and y axes are equal to one another, i.e., that $g^{(1)} = g^{(2)}$, we finally obtain the formula

$$\Delta\zeta = g\varphi\eta_0 + \tau\rho c^2\delta/(1-i\omega\tau), \tag{4.8}$$

which expresses the correction to the bulk viscosity in the same form as it appear in the Mandel'shtam-Leontovich theory (cf. Ref. 1, §78). Here the relaxation time τ and the relative change δ in the velocity of sound depend on the field strength:

$$\tau = \tau_0 \Big/ \left[\left(1+\xi\frac{L_2}{L_1}\right) g^{(1)} + g^{(3)}\right], \quad \delta = \eta_0\varphi\xi L_1 [ge]^2/\rho c^2\tau_0. \tag{4.9}$$

In view of the asymptotic behavior of the functions L_1 and L_2,

$$L_n = \frac{1}{(2n+1)!!}\xi^n \quad (\xi \ll 1), \quad L_n = 1 + 0\left(\frac{1}{\xi}\right) \quad (\xi \gg 1), \tag{4.10}$$

it is easily seen that the bulk viscosity $\Delta\zeta$ increases in weak fields ($\xi \ll 1$) as ξ^2. As was noted in the introduction, the relaxation of rotational degrees of freedom does not affect the viscosity at all in the absence of a field. In strong fields ($\xi \gg 1$) the bulk viscosity, like the other viscosity coefficients of a ferromagnetic suspension,[4] ceases to depend on the field strength (the saturation effect) and reaches the maximal value

$$\Delta\zeta_\infty = \eta_0\varphi\{g + [ge]^2/g^{(1)}\}. \tag{4.11}$$

It is interesting that in this limit the viscosity is also independent of the frequency, since the relaxation time τ decreases as $1/\xi$ with increasing field strength.

Turning now to the problem of sound absorption in a ferromagnetic suspension, we note that the correction to the bulk viscosity is of the same order as (or of lower order than) the ordinary viscosity of the liquid. Hence in calculating the absorption coefficients we may immediately make use of the general expression $\gamma = \omega^2\Delta\zeta/2\rho c^3$ (see Ref. 1, §77). Substituting the complex bulk viscosity (4.8) into this expression and separating the real and imaginary parts, we obtain the following formula for the absorption coefficient:

$$\gamma = \frac{\delta}{2\lambda}\frac{\omega\tau}{1+\omega^2\tau^2}, \tag{4.12}$$

in which $\lambda = c/\omega$ is the wavelength of the sound. The absorption coefficient, like the bulk viscosity itself, increases monotonically with increasing field strength and reaches saturation in strong fields. In order of magnitude we have $\gamma \approx \gamma_0\varphi$, where γ_0 is the absorption coefficient of the pure liquid. The volume concentration φ of the particles may ordinarily amount to some 20–30%, and when a magnetic field is applied to the ferromagnetic suspension the sound-absorption coefficient should obviously increase by that same amount.

The imaginary part of the absorption coefficient determines the change of the velocity of sound in the suspension. This effect may be neglected here, however, since estimates show that the quantity δ, which is a measure of the relative change in the velocity of sound, is small.

5. DUMBBELL-SHAPED PARTICLES

The formulas derived in the preceding section enable one to calculate the bulk viscosity of a ferromagnetic suspension in terms of the coefficients g. To determine these coefficients themselves, however, one must solve the auxiliary hydrodynamic problem of flow around particles of given shape, as was discussed in Sec. 2. This problem is comparatively easy to solve for spherical, or nearly spherical, particles. Below we shall consider an example in which the suspended particles have the shape of dumbbells whose balls are slightly deformed spheres (see Fig. 1). This example demonstrates the change in the bulk viscosity that takes place when the particles stick together. The bulk viscosity increases in the ab-

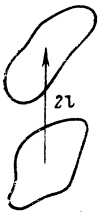

FIG. 1.

sence of a field even when spherical particles stick together, and when aspherical particles stick together the bulk viscosity becomes field dependent.

The equation of the surface of a slightly aspherical particle can be taken in the form

$$r=a[1+\varepsilon f(\theta, \varphi)], \tag{5.1}$$

where a is the radius of the approximating sphere, $f(\theta, \varphi)$ is an arbitrary function of the spherical-coordinate angles, and ε is a constant that may be called the asphericity parameter. The origin of coordinates and the mean radius of the particle can always be so chosen that the equations

$$\int f(\theta, \varphi)\left\{\begin{matrix}1\\ \mathbf{r}\end{matrix}\right\}\sin\theta\, d\theta\, d\varphi=0 \tag{5.2}$$

will be satisfied. Limiting ourselves to the first order in the asphericity parameter, we can solve Eqs. (2.5) and calculate the force, the couple,[6] and the scalar part S of the stresses[2] acting on the particle in the flowing liquid:

$$\begin{aligned} F_i&=-6\pi\eta_0 a(\delta_{ik}-\varepsilon f_{ik})u_k,\\ L_i&=-8\pi\eta_0 a^3(\delta_{ik}-3\varepsilon f_{ik})\omega_k,\ S=^{16}/_9\pi(\eta_0+^3/_i\xi_0)a^3\Omega, \end{aligned} \tag{5.3}$$

where

$$f_{ik}=\frac{3}{8\pi r^2}\int\left(x_ix_k-\frac{1}{3}r^2\delta_{ik}\right)f(\theta, \varphi)\sin\theta\, d\theta\, d\varphi. \tag{5.4}$$

Since we intend to calculate the coefficients of friction for dumbbell-shaped particles, let us assume that the particle is displaced from the center of the unperturbed liquid flow, and let us denote the displacement vector by $\mathbf{l}$. Since the velocity of the liquid no longer vanishes at the particles, the force $\mathbf{F}$ must be assumed to be proportional to the difference between the velocity of the particle and the velocity of the flow (2.1):

$$F_i\propto u_k-^1/_3\Omega l_k. \tag{5.5}$$

Now let us displace the origin of a coordinate system rigidly fixed to the particle by the quantity $-\mathbf{l}$, thus returning it to the center of the flow (2.1). It is known (see Ref. 9, §§31 and 34) that in this case the force $\mathbf{F}$ and the angular velocity $\boldsymbol{\omega}$ do not change, while the quantities $\mathbf{u}$, $\mathbf{L}$, and S transform as follows (cf. the definitions (2.6) of $\mathbf{L}$ and S):

$$\mathbf{u}=\mathbf{u}'+[\boldsymbol{\omega}\mathbf{l}], \tag{5.6}$$

$$\mathbf{L}'=\mathbf{L}+[\mathbf{l}\mathbf{F}],\quad S'=S+^1/_3\mathbf{l}\mathbf{F}. \tag{5.7}$$

Here the primes mark quantities referred to the new origin of coordinates.

Now let us consider two particles each of which lies at a distance l from their common center of mass (we assume that the particles have equal masses). If we neglect the hydrodynamic interaction between the particles, we can seek the force, the couple, and the stresses S_{ik} acting on the dumbbell-shaped particle as the sum of the contributions from the two component particles. To estimate the errors made in such a calculation, let us consider, for example, a particle of radius a rotating in the medium with the angular velocity ω. Then there will be a couple $L_1\sim\eta_0 a^3\omega$ acting on the particle as a result of the frictional forces due to the surrounding liquid. If there is a second particle rotating with the same angular velocity ω at a distance l from the first one, then there will be an additional couple $L_{12}\sim\eta_0 a^6\omega/l^3$ acting on the first particle (cf. Ref. 1, §20). The condition $L_1\gg L_{12}$ reduces to the condition $(a/l)^3\ll 1$. Thus, the interaction between the particles will be an order of magnitude weaker than the interaction of each particle with the principal flow, even if the particles touch one another, i.e., if $l=2a$. Now substituting (5.6) into (5.5) and then substituting (5.5) and (5.3) into (5.7) and noting that $\mathbf{l}$ must be replaced by $-\mathbf{l}$ for one of the particles, we find without difficulty that the condition $F=0$ yields the result $u_i=O(\varepsilon)$ (here u_i is the velocity of the center of inertia of the composite particle). Now by equating the total couple L_i+K_i to zero, we can express the angular velocity of the particle and the quantity S in the form of Eqs. (2.9) and (2.10) with

$$\begin{aligned} g&={}^6/_5+2\xi_0/\eta_0+q(1-\varepsilon e_ie_kf_{ik})+O(\varepsilon^2),\\ \mathbf{g}&=\varepsilon\frac{q}{4+3q}[(f\mathbf{e})\mathbf{e}]+O(\varepsilon^2),\quad g_{ik}=\frac{4\delta_{ik}+3qe_ie_k}{12(4+3q)}+O(\varepsilon), \end{aligned} \tag{5.8}$$

where we have used the notation $q=l^2/a^2$ and $(f\mathbf{e})_i=f_{ik}e_k$, and the quantity f_{ik} is to be understood here as the arithmetic average of the two tensors (5.4) for the two component particles. We have also assumed that the particles are magnetized in the direction of the line joining their centers (i.e., that $\mathbf{l}$ is parallel to $\mathbf{e}$).

Let us use this result to calculate the bulk viscosity of the suspension. First, by substituting the scalar g from (5.8) into formula (4.8) and comparing the result with (1.1), we can easily see that even in the absence of a field the correction to the viscosity increases when the dumbbell-shaped particles are formed by more than a factor of two (more precisely, by a factor of $2+3q/4$ if we assume that $\xi_0=0$; here we have used the fact that the volume concentration φ of the particles does not change when the particles stick together in pairs). But of course the magnetic-field dependence of the viscosity of the suspension is of greater interest since the composite particle, unlike its constituent particles, admits of a pseudovector $\mathbf{g}$ even in the first order in the asphericity parameter ε. According to Eqs. (4.11) and (5.8), the correction to the viscosity due to the field in the limit $\xi\gg 1$ has the form

$$\Delta\xi_\infty'=\eta_0\varphi\frac{3\varepsilon^2q^2}{3q+4}[(f\mathbf{e})\mathbf{e}]^2, \tag{5.9}$$

i.e., it is quadratic in the asphericity parameter.

As a specific example of slightly aspherical particles let us consider spheroidal particles whose surfaces are described by the equation

$$(x^2+y^2)/a^2+z^2/b^2=1.$$

When the difference between the principal diameters of the spheroid is small this equation can be written in the form of Eq. (5.1) with

$$\varepsilon=\frac{1}{10}\left(1-\frac{a^2}{b^2}\right), \quad f(\theta,\varphi)=5\left(\cos^2\theta-\frac{1}{3}\right)$$

(the angle θ is measured from the direction of the axis of the spheroid). Then calculating the tensor f_{ik} with Eq. (5.4) we find

$$f_{ik}=k_ik_k-{}^1/_3\delta_{ik}, \tag{5.10}$$

where k_i is a unit vector in the direction of the axis of the spheroid. Now by substituting (5.10) into (5.9) (and assuming for simplicity that the axes of the two spheroids are parallel) we can easily see that the viscosity coefficient is proportional to $(\mathbf{k}\cdot\mathbf{e})^2((\mathbf{k}\times\mathbf{e})\times\mathbf{e})^2$ and may vanish if the spheroid is magnetized either parallel or perpendicular to the axis.

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Magneto-optical investigations of the exciton band in CdTe:Mn^{2+}

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A strong enhancement of magneto-optical effects (Faraday rotation of the plane of polarization of light, magnetic circular dichroism of the reflection) was observed when CdTe crystals were doped with Mn^{2+} ions. It was established that the effect reduces to splitting, of the band of the 1*s* exciton into five components in an external magnetic field. The splitting is proportional to the concentration and to the spin polarization of the system of the Mn^{2+} impurity ions. At the employed manganese concentrations ($\sim 8\times 10^{18}$ cm^{-3}) the effective field in which the splitting of the exciton band takes place reaches several hundred kilo-oersteds in an external field of 30 kOe. The change of the spin polarization of the impurity system upon saturation of the microwave EPR transitions of the Mn^{2+} ions decreases the splitting. The magneto-optical effects observed in the exciton band of the CdTe:Mn^{2+} crystals, their dependence on the field H and on the microwave pump, and the polarization of the split components are all attributed to exchange interaction of the localized magnetic Mn^{2+} ions with the hole and electron contained in the free exciton.

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INTRODUCTION

Investigations of the exciton spectra of semiconducting CdTe crystals doped with certain ions (Fe, Mn) of the iron group[1] have shown that when these ions are introduced into the crystal a short-wave shift of the exciton line takes place. The magnitude of the shift depends strongly on the impurity concentration, but the exciton line itself does not disappear. Investigations of the influence of the impurity on the character of the Zeeman splitting of the exciton band can yield important information on the singularities of the excitons in crystals doped in this manner. Taking into account the difficulty of observing the Zeeman effect of a sufficiently broad exciton band in the CdTe : Mn^{2+} crystals, we have decided to measure the Faraday rotation (FR) and the magnetic circular dichroism (MCD) of samples with different degrees of doping by Mn^{2+} ions.

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