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ON THE ROTATIONAL EFFECT IN NONUNIFORM MAGNETIC FLUIDS

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In the paper, two problems are considered in which the volume ponderomotive forces associated with internal rotations are shown to be mainly responsible for generation of circulation flows in magnetic fluids. One problem is devoted to an analytical investigation of the motion of the magnetic fluid in a vertical long cylinder. The motion is generated by the high-frequency rotating field orthogonal to the cylinder axis. The velocity profile is determined in the approximation of a weak field. Theoretical and experimental results are compared. The second problem deals with an experimental study of the flow of a magnetic fluid subjected to the joint action of rotating and bias fields. It has been found that, with the bias field on, the flow structure qualitatively changes and the flow direction may be reversed.

Introduction

Hydrodynamic flows arising in dielectric magnetic fluids under a rotating magnetic field have been investigated in some papers beginning with Moskowitz and Rosensweig [1]–[10]. Magnetization nonequilibrium is a necessary condition of generation of these flows. When the magnetic field $\mathbf{H}$ rotates, the magnetization $\mathbf{M}$ of the fluid also rotates at the same speed as the field but lags in phase by a certain angle due to the finite relaxation time. The individual colloidal particles also rotate and form microscopic vortices with the characteristic sizes of 10^{-2}–10^{-3} μm. However these vortices do not automatically induce a macroscopic hydrodynamic flow (rotational effect). Macroscopic flow is possible only under certain additional conditions. In recent years, several papers have been devoted to the mechanisms capable of generating macroscopic vortices.

1. *Instability of the state of rest.* Kashevskiy [11] and then Lyubimova and Lyubimov [12] showed that the mechanical equilibrium of the magnetic fluid becomes unstable in a uniform rotating field. As a result, the equilibrium is changed for the macroscopic motion when the field intensity and the rotation frequency exceed some critical values. However, these values increase with decreasing volume fraction ϕ of magnetic particles, and at $\phi \to 0$ the instability region extends to infinity. It is clear that the fluid flow observed experimentally is unrelated to this instability. The point is that the flow is not of a threshold character and exists in both concentrated and dilute ferrofluids.

2. *Nonuniformity of the internal moment of momentum.* Zaitsev and Shliomis [13] and Cebers [14] related the rotational effect to the nonuniformity of the internal moment of momentum $\mathbf{S}$. The distribution of $\mathbf{S}$ in any volume of fluid appears to be nonuniform since the rotation of the particles near the immovable walls is hampered. According to [13, 14], the macroscopic flow vorticity Ω inside the vertical cylinder of radius R is equal to $3\Omega_p\phi d/R$, where Ω_p and d are the rotational velocity and the diameter of a colloid particle, respectively. For the volume fraction $\phi \approx 0.1$ and the particle diameters $d = 10$ nm, from the last formula it follows that $\Omega/\omega \leq 10^{-7}$. This value of vorticity is three or four orders of magnitude less than that observed experimentally and implies that the studied effect is negligibly small.

3. *Asymmetric tangential stresses on the boundary of the magnetic fluid.* These stresses initiated by internal (microscopic) rotations have been determined by Cebers [15, 16] for the gap between two coaxial cylinders and by Vislovich [17, 18] for the plane layer. If one of the layer boundaries is free, then the tangential stresses are capable of generating cylindrical or plane Couette flows. The estimated values of velocity agree with the experimental

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data in the order of magnitude. Because of this, the proposed mechanism can be considered to be real, yet it fails to explain the occurrence of flows for immovable boundaries. The theory [15]–[18] predicts no movement in this case.

4. *Nonisothermity of the fluid related to the dissipation energy of the rotating field.* As has been shown in [19, 20, 21], an averaged hydrodynamic flow occurs in the fluid due to inhomogeneous heating caused by the rotating field. The potentiality of volume magnetic forces is violated in this case. The studies were performed using Shliomis' ferrohydrodynamic equations [22] which involve the relaxation equation for magnetization. In [20] these equations have been solved numerically for a long vertical cylinder and strong fields. In [21] the region of weak fields has been considered. The experimental data [9, 20] support the conclusion about the essential role of internal heat sources in the generation of hydrodynamic flows. On replacing the vertical cylinder with the magnetic fluid by the rotating magnetic field, there occur a radial temperature difference and, as the result, a circulation flow moving opposite to the field rotation.

Analysis of theoretical models and experimental data indicates that there are at least two independent reasons for the rotational effect. They are the tangential magnetic stresses on the free boundary of the fluid and the nonuniformity of magnetic permeability that results in volume ponderomotive forces. The nonuniformity of magnetic permeability, in turn, may be the result of the temperature gradient or the applied field nonuniformity. For weak fields and low frequencies, energy dissipation is low, the fluid is isothermal, and the surface effects play a leading role in generation of hydrodynamic flows. Volume forces manifest themselves in the case of strong fields or high frequencies. Moreover they remain the only reason for the rotational effect if the boundaries of the cavity with magnetic fluid are solid and fixed.

In this paper we investigate two problems for which volume ponderomotive forces are of crucial importance. The first allows us to study analytically the motion of a magnetic fluid under the influence of a high-frequency rotating field in a long vertical cylinder. The rotating field results in heating of the fluid and in the nonuniformity of magnetic permeability in the radial direction. The second problem involves the experimental investigation of the magnetic fluid motion in a plane cuvette under the action both of rotating and permanent bias fields.

1. Magnetic fluid in a long vertical cylinder

Let a dielectric magnetic fluid fill a vertical cylinder whose length is large compared to its diameter (the plane problem). The applied magnetic field $\mathbf{H}_0$ is uniform far from the cylinder and perpendicular to the cylinder axis. The field rotates in the horizontal plane, so the angular velocity vector $\boldsymbol{\omega}$ is directed upwards along the z axis. The problem consists in finding the amplitude and the profile of the hydrodynamic flow on the basis of ferrohydrodynamic equations [19, 22], taking the magnetization nonequilibrium into account.

$$\rho\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = -\nabla p + \eta\nabla^2\mathbf{v} + \mu_0\Big[(\mathbf{M}\cdot\nabla)\mathbf{H} + \frac{1}{2}\nabla\times(\mathbf{M}\times\mathbf{H})\Big], \tag{1}$$

$$\nabla\times\mathbf{H} = 0, \qquad \nabla\cdot\mathbf{B} = 0, \qquad \mathbf{B} = \mu_0(\mathbf{H}+\mathbf{M}), \tag{2}$$

$$\rho C\frac{\mathrm{d}T}{\mathrm{d}t} = \kappa\nabla^2 T + q. \tag{3}$$

Here, $\mathbf{v}$ is the flow velocity, T the temperature, q the density of internal heat sources, and $\mu_0 = 4\pi\times10^{-7}$ H/m. The physical parameters of the fluid (the density ρ, viscosity η, specific heat C, and heat conductivity κ) are considered to be constant, and the fluid itself is assumed to be incompressible ($\nabla\cdot\mathbf{v} = 0$). Gravitational convection is not taken into consideration.

The system of equations (1)–(3) is not closed. In order to close the system, it is necessary to write an additional relaxation equation for the magnetization, and the static magnetization curve linking the vectors $\mathbf{M}$,

$\mathbf{H}$, $\nabla \times \mathbf{v}$ and temperature. However, in this general formulation even a numerical solution would be extremely laborious and an analytical solution quite impossible. The problem can be considerably simplified within the framework of the weak vorticity approximation. According to this approximation $|\nabla \times \mathbf{v}|$ is assumed to be small as compared with the field rotation frequency ω. As shown in experiments [9, 20, 23], this condition is well satisfied in practice: The fluid rotation velocity is two to three orders smaller than ω. In the present case of a weak uniform rotating field the condition of $|\nabla \times \mathbf{v}| \ll \omega$ makes it possible to reduce the relaxation equation to a simple linear relation between $\mathbf{M}$ and $\mathbf{H}$ by introducing the dynamic susceptibility tensor:

$$M_i = \chi_{ij} H_j, \qquad \chi_{ij} = \begin{pmatrix} \chi_1 & \chi_2 \\ -\chi_2 & \chi_1 \end{pmatrix}, \qquad i, j = 1, 2. \tag{4}$$

Here, χ_1 and χ_2 are the real and imaginary parts of the dynamic susceptibility. Both of them can readily be measured experimentally, hence the temperature dependence of the susceptibility is assumed to be known. In this case, the internal structure of the magnetic fluid is of no importance. The fact that the frequency dependence of the susceptibility of the real magnetic fluids is logarithmically weak and the fluid vorticity is small means that the dependence of χ_1 and χ_2 on the flow velocity can be neglected. We cannot neglect the temperature dependence of the susceptibility, since it is precisely this dependence that determines the contribution of the magnetic forces to the right-hand side of Eq. (1). However, the susceptibility varies only slightly with temperature: its relative variation $\varepsilon \propto \Delta T/T$ (ΔT is the typical difference in temperatures) and does not exceed several percent in the experiment devoted to the rotational effect [9, 20]. This makes it possible to approximate the temperature dependence of the susceptibility with a linear law in calculating the magnetic forces and neglect that dependence in the expression for the internal heat sources in Eq. (3). Standard boundary conditions are imposed on the lateral surface of the cylinder. They are the no-slip condition for the velocity at the wall, continuity of the tangential components of the intensity $\mathbf{H}$, and continuity of the normal components of the induction $\mathbf{B}$. The temperature T_0 of the lateral surface is assumed to be constant.

The rotating field energy dissipation leads to the heating and nonisothermity of the fluid. The torque acting on a unit volume of the fluid by the field is equal to $\mathbf{N} = \mu_0(\mathbf{M} \times \mathbf{H})$, and the density of internal heat sources is equal to $q = \omega \mathbf{N} = \mu_0 \chi_2 H^2$. In general, the quantity q is nonuniform as a consequence of the temperature dependence of χ_2 and the field intensity of the fluid. However, this effect will be neglected because it leads only to the second-order corrections in terms of ϵ in Eq. (1). As a result of the axial symmetry of the problem, it is natural to seek the solution for which $v_r = v_z = 0$ and $v_\varphi = v(r)$ (here r, z and φ are cylindrical coordinates). In this case the convective term in heat equation (3) disappears and the fluid temperature is equal to

$$T(r) = T_0 + \frac{\mu_0 \chi_2 \omega H^2}{4\kappa}(R^2 - r^2). \tag{5}$$

We now find the contribution of the magnetic force $\mathbf{F}$ to the right side of the equation of motion (1). Initially, using the well-known vector identities and Maxwell equations (2), we write

$$\begin{aligned} \mathbf{F} &= \mu_0 \Big[(\mathbf{M} \cdot \nabla)\mathbf{H} + \frac{1}{2} \nabla \times (\mathbf{M} \times \mathbf{H}) \Big] \\ &= \frac{\mu_0}{2} \big[\nabla(\mathbf{M} \cdot \mathbf{H}) + (\mathbf{M} + \mathbf{H}) \nabla \cdot \mathbf{H} - \mathbf{H} \times (\nabla \times \mathbf{M}) \big]. \end{aligned} \tag{6}$$

The first term on the right-hand side of (6) can be omitted, since the forces associated with this term bring about only small radial variations of pressure. The other two terms are essential. Provided that χ is nonuniform, these terms make a time-independent contribution to the φ-component of the force $\mathbf{F}$. Using relation (4) and Maxwell equations (2), we obtain

$$\nabla \mathbf{M} = -\nabla \mathbf{H} = \frac{1}{1+\chi_1}\left[\mathbf{H}\cdot\nabla\chi_1 + \frac{\boldsymbol{\omega}}{\omega}(\nabla\chi_2 \times \mathbf{H})\right],$$

$$\nabla \times \mathbf{M} = (\nabla\chi_1 \times \mathbf{H}) - \frac{\boldsymbol{\omega}}{\omega}\nabla\cdot(\chi_2\cdot\mathbf{H}). \tag{7}$$

Substitution of (7) in (6), with allowance for the temperature dependence of χ_1 and χ_2 and expression (5), gives

$$\langle F_\varphi \rangle = (\mu_0\chi_2 H^2)^2\frac{\omega\sigma r}{4\kappa}, \quad \sigma = \frac{\partial}{\partial T}\ln\tan(\delta), \quad \tan(\delta) = \frac{\chi_2}{1+\chi_1}, \tag{8}$$

where σ is the temperature coefficient of loss tangent, and the angular brackets mean averaging over the angle φ (i.e., over time). In deriving (8) we have taken into account that $< H_r^2 >= H^2/2$, $< H_r H_\varphi >= 0$.

The field intensity H, entering into formulas (5) – (8), can differ substantially from the applied field intensity H_0, which alone can be controlled experimentally. In order to determine the relation between H and H_0, it is necessary, in general, to calculate the magnetic field both inside and outside the fluid using the Maxwell equations, the boundary conditions for $\mathbf{H}$ and $\mathbf{B}$, relation (4) and temperature profile (5). However, taking the nonisothermity of the fluid into account once again leads to second-order corrections. Therefore, it is simpler to use the solution of the analogous problem for an isothermal fluid. According to the data presented in [23] we obtain

$$H^2 = \frac{4H_0^2}{4+4\chi_1+\chi^2},$$

where χ is the modulus of the dynamic susceptibility. Finally, for the magnetic force we have

$$\langle F_\varphi \rangle = \left(\frac{\mu_0\chi_2 H_0^2}{4+4\chi_1+\chi^2}\right)^2\frac{\omega\sigma r}{4\kappa}. \tag{9}$$

With (9), the projection of Eq. (1) on the coordinate φ has the form

$$\eta\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial v}{\partial r}\right) - \frac{v}{r^2}\right] + \left(\frac{\mu_0\chi_2 H_0^2}{4+4\chi_1+\chi^2}\right)^2\frac{\omega\sigma r}{4\kappa} = 0.$$

This equation and the nonslip condition for the fluid at the wall ($V = 0$ when $r = R$) give the cubic velocity profile

$$V = \frac{\omega\sigma}{2\kappa\eta}\left(\frac{\mu_0\chi_2 H_0^2}{4+4\chi_1+\chi^2}\right)^2 r(R^2 - r^2), \tag{10}$$

$$V_m = \frac{\omega\sigma R^3}{3\sqrt{3}\kappa\eta}\left(\frac{\mu_0\chi_2 H_0^2}{4+4\chi_1+\chi^2}\right)^2. \tag{11}$$

Here, V_m is the amplitude of the velocity. At frequencies $\omega \leq 10^5\ \mathrm{s}^{-1}$ the loss tangent decreases with temperature [24], and the quantity s is negative. This means that the velocity is also negative: the fluid rotates in the direction opposite to that of field rotation. This effect was observed in the experiments. As an example, we will estimate the velocity amplitude for experimental conditions described in [9, 20]. Substitution of the physical parameters of the fluid ($\chi_1 = 5.0$, $\chi_2 = 0.22$, $\eta = 0.0085$ Pa·s, $\kappa = 0.18$ W/mK, $\sigma = -0.014\ \mathrm{K}^{-1}$) in (11) at $R = 16.5$ mm and $H_0 = 3.7$ kA/m gives $V_m = 0.1$ cm/s, which is in good agreement with the experimental results. Unfortunately, it is impossible to check the shape of profile (10), since the known experiment, in which such measurements were carried out, deals with the surface flow formed due to magnetic tangential stresses, and not due to the volume heat release.

2. Rotational effect in a bias field

In this section we consider the possibility of inducing the flow of magnetic fluid by changing its susceptibility in a manner different from heating. The magnetic susceptibility increases with the concentration of the magnetic phase. Therefore, tests on the radial gradient of concentration seem to be the simplest way of observing the rotational effect. However, it is impossible in this case to obtain the stationary and nonuniform concentration field. Gravitational convection and diffusion of colloidal particles lead to the fast smoothing of concentration disturbances even in the case of some concentration drop at the beginning of the experiment. For this reason we have chosen another way consisting in the application of the nonuniform constant (bias) field producing the spatial modulation of the magnetic susceptibility. It is clear that such modulation can be performed at the cost of the nonlinearity of the bias curve only. Because of this, the bias field must be rather strong. One can see from (6), (7) that in weak fields the volume magnetic force is potential (if the fluid is uniform in temperature) and cannot lead to the rotational effect no matter what the applied field configuration. The amplitude of the uniform rotating field can be arbitrary and small as compared with the bias intensity. It is hoped that the qualitative picture in this case may be the same as in the case of nonuniform heating of the fluid. The modulation of the differential susceptibility $\chi_d = \partial M/\partial H$ by the bias field will produce a ponderomotive force as well as temperature modulation of the initial susceptibility χ.

Figure 1 presents the geometry of the problem. The magnetic fluid *1* was poured into the circular cuvette *2* the height of which, 3 mm, is much smaller than its diameter, 47 mm. The horizontal magnetic field rotates around the vertical axis. The volume of fluid is taken so as to exclude the appearance of a meniscus near the cuvette wall. The point is that the sign of the magnetic term in the stress tensor of the magnetic fluid [19] depends on the shape of the meniscus. This fact was supported by experiments of [8, 9, 10] and is, in our opinion, quite obvious. So, the observed flow can be investigated quantitatively only on condition that the surface curvature is taken into account. That is the reason why the present work studies the problem dealing with plane surfaces, as in [10]. The cuvette edge forms an acute angle with the internal cylindrical surface. When the fluid is poured into the cuvette, its free surface clings to the acute angle, providing the joint coupling of the cuvette edge and the free surface. It was a simple matter to obtain the state with a horizontal surface by varying the volume of the fluid. The cuvette was manufactured from a nonwettable material (Teflon) to avoid the overflow of the fluid over the cuvette edge. The rotating magnetic field was generated by two couples of crossed Helmholtz coils. They were fed from alternating currents with a phase shear of $\pi/2$. The field amplitude was 3 kA/m at a rotation frequency 560 Hz. At this frequency and in the absence of the bias field, the real and imaginary parts of the dynamic susceptibility were $\chi_1 = 3.61$ and $\chi_2 = 0.27$, respectively. The bias field was generated by a cylindrical permanent magnet *3* of diameter 14 mm and thickness 5 mm. The magnet was magnetized along its own axis and placed under the cuvette bottom (Fig. 1). The field intensity near the magnet endwall was 50 kA/m. With the purpose of visualizing the flows, the magnetic fluid surface was covered by particles of aluminum powder. Having photographed the fluid surface and handled the pictures with an instrumental microscope, we obtained the flow velocity.

Figure 2 presents the profiles of the flow in the cuvette without (curve 1) and with (curve 2) the bias field. In the former case the velocity profile is of a shape similar to that obtained experimentally for a free surface [10]. The magnetic fluid rotates in the same direction as the field due to the magnetic leakage fields generated by the fluid itself and the tangential magnetic stresses on the fluid surface. The maximum of the flow velocity is near the cuvette edges, i.e., where the amplitude of leakage fields shows a maximum. The bias field changes qualitatively the flow structure: the internal part of the fluid rotates in the direction opposite to that of the field rotation. One can appreciate this result if one remembers that the bias field decreases the differential susceptibility. In the cuvette

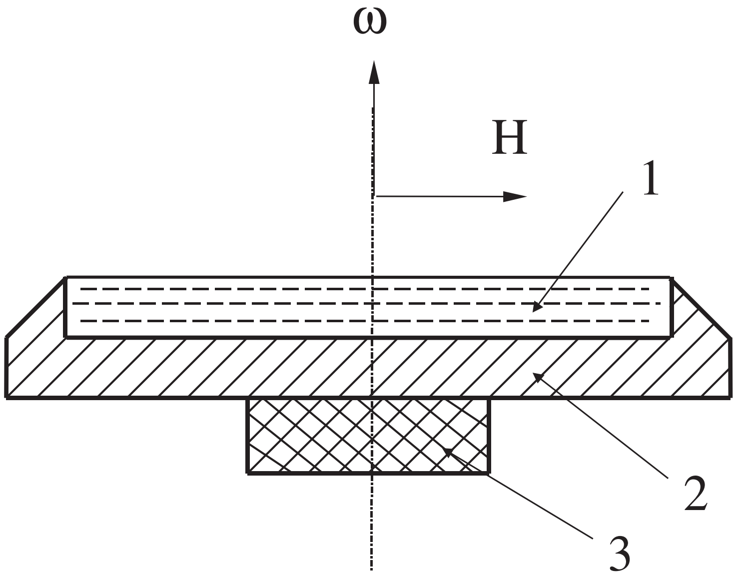

Fig. 1. Cross-section of the cuvette with the magnetic fluid: *1* – magnetic fluid; *2* – cuvette body; *3* – permanent magnet.

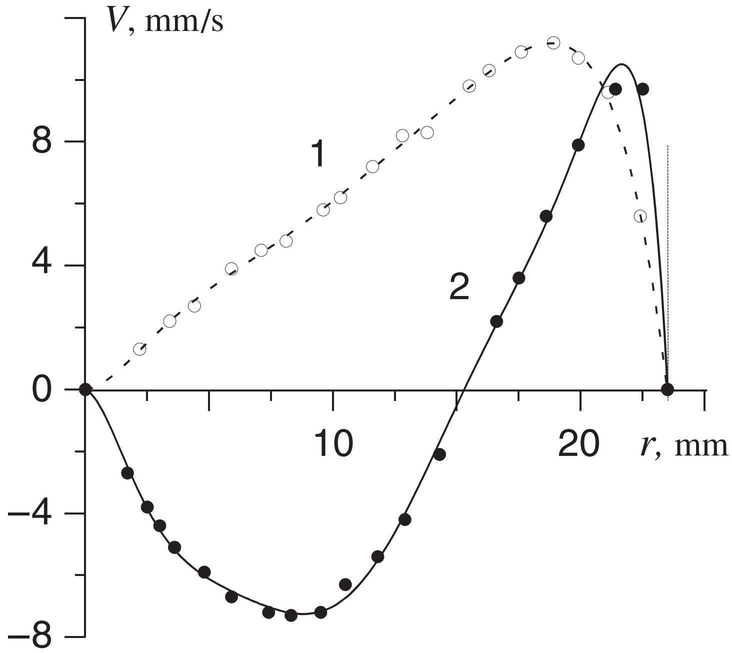

Fig. 2. Velocity profiles for the circular cuvette in the absence of the bias field (curve *1*) and with it (curve *2*).

center the bias field shows a maximum and generates a region with reduced magnetic susceptibility. This situation is similar to the case when the central part of the cuvette was heated. Clearly the motion of the central part of the fluid must be realized in the same way as in the case with internal heat sources, i.e., in the direction opposite to the field. This phenomenon is observed in the present study. The field intensity, on removing from the permanent magnet, increases inversely to the distance cubed, and at a short distance from the cuvette edges its influence becomes weaker as compared with that of the leakage fields. The fluid rotates around the field with about the same velocity as in the first case.

Conclusions

The obtained theoretical and experimental results fully support the supposition that the spatial nonuniformity of the magnetic susceptibility is a sufficient condition for observing the rotational effect. The reason why the magnetic properties are nonuniform (heating or bias fields) is of no importance in this case. Moreover the appearance of magnetic tangential stresses in the fluid boundary can be considered as a result of the spatial nonuniformity of magnetic properties. The only difference is that on the magnetic fluid boundary these properties change abruptly and in the volume continuously. The flow of the magnetic fluid generated by the internal heat sources is of low intensity (typical velocities of order 0.1 cm/s) and can be observed when the surface effects are inessential. The velocity of the magnetic fluid in the horizontal cuvette generated by the rotating and bias fields approaches 1 cm/s. In the latter case the effects associated with the volume and surface forces are equal in order of magnitude. They compete with each other and initiate the appearance of rather complicated profiles of the flow.

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