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Magnetophoresis of particles and aggregates in concentrated magnetic fluids

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Experimental and theoretical studies were carried out to investigate the problem of magnetophoresis in a thin layer of concentrated magnetic fluids, concerning the aspect of particle aggregation. A heuristic theoretical model, describing diffusion fluxes of individual and aggregated particles, is suggested. The solution of related diffusion and magnetostatic problems are compared with the experimental data. The analysis of the data shows that the aggregates essentially change the concentration profile. Good agreement between experimental and theoretical curves is observed in the case when the aggregates contain, on average, more than ten particles.

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I. INTRODUCTION

It is known that, in the course of time, an initially homogeneous magnetic fluid filling an arbitrary cavity becomes spatially inhomogeneous with respect to the magnetic phase concentration due to gravitational sedimentation and magnetophoresis (the motion of particles under the action of nonuniform magnetic field). In the absence of convective motion, the only factor that prevents the concentration stratification of the fluid is the gradient diffusion of particles. The concentration profile in a cavity at some arbitrary time can be obtained from the solution of the boundary-value problem, including Maxwell's equations for the magnetic field and the dynamic mass transfer equation with consideration for terms responsible for magnetophoresis and sedimentation of particles [1–4]. In the case of dilute colloids, the interparticle interactions and demagnetizing fields are not very important (except for the effects of particle aggregation), and the mass transfer equation is linear in concentration of particles. A stationary solution of this equation including the case of particle aggregation can be expressed analytically [5,6].

In concentrated magnetic fluids, the steric, magnetodipole and hydrodynamic interparticle interactions are rather strong and therefore the mass transfer processes are generally described by the nonlinear equations, in which the term responsible for magnetophoresis depends on magnetization and magnetic field strength. In turn, the strength of the magnetic field in the fluid is specified by the spatial distribution of particles, which suggests that the magnetic and diffusion parts of the problem are interrelated and should be solved simultaneously. A distinguishing feature of this problem is that the characteristic damping time of concentration perturbations $\tau_D \approx L^2/(\pi^2 D)$ are at least six to seven orders of magnitude higher than the relaxation time of the magnetic moment $\tau_B \approx 3\eta V/(kT)$ (L is the characteristic dimension of cavity, D is the diffusion coefficient, η is the viscosity of the magnetic fluid, V is the volume of a colloidal particle). For this reason, magnetization of the colloid is considered to be equilibrium, and the concentration field is considered to be frozen when calculating the magnetic field.

Several versions of the mass transfer equation for magnetic fluids, differing by the extent to which magnetophoresis, sedimentation, gradient diffusion, interparticle interactions, and anisotropy of transfer coefficient are taken into account, have been previously considered in Refs. [2,3,5–14]. It would seem that one of the most complete mass transfer equations has been proposed in Ref. [12]. The equation is constructed as an expansion of the free energy of the interacting dipole system in terms of the particle concentration. This equation describes the temporal and spatial variations of the volume fraction φ of single-domain colloidal particles and, in the absence of convective flows, is written as

$$\frac{\partial\varphi}{\partial t} = -\nabla\cdot\left\{D_0K(\varphi)\left(\varphi L(\xi_e)\nabla\xi_e + \varphi G_\gamma\mathbf{e} - \left[1+\frac{2\varphi(4-\varphi)}{(1-\varphi)^4} - \varphi\frac{\partial^2(\varphi^2 G)}{\partial\varphi^2}\right]\nabla\varphi\right)\right\}. \quad (1)$$

Here, $K(\varphi) = b/b_0$, where b and b_0 are the particle mobility in the magnetic fluid and carrier fluid, respectively, $D_0 = b_0kT$ is Einstein's value of the diffusion coefficient for a Brownian particle in dilute solution, $\mu_0 = 4\pi \times 10^{-7}$ H/m, $L(\xi) = \coth(\xi) - 1/\xi$ is the Langevin function, and $\xi = \mu_0 mH/(kT)$ is the Langevin parameter, $\lambda = \mu_0 m^2/(4\pi d^3 kT)$ is the parameter of magnetodipole interactions (the ratio of the magnetodipole interaction energy to thermal energy), m and d are the magnetic moment and full diameter of the particle (including a protection shell), respectively, kT is the energy of the thermal motion, G_γ is the gravitational parameter, which is equal to the inverse height of the barometric distribution, $\mathbf{e}$ is the unit vector in the direction of the gravitational acceleration $\mathbf{g}$, and $G(\lambda,\varphi)$ is the contribution of magnetodipole interactions to the free energy density referred to the density of the thermal energy of the Brownian particle motion. The first term in Eq. (1) represents magnetophoresis, and the three terms in square brackets reflect the existence of three physical effects determining the intensity of diffusion processes in a magnetic fluid: gradient diffusion, steric interactions, and effective attraction of particles due to magnetodipole interactions, respectively. Steric interactions are taken into account in the frame of the Carnagan-Starling approximation for the system of hard spheres, similarly as has been done in Refs. [9,12]. Relative mobility of particles $K(\varphi)$ in a concentrated suspension can be calculated by using Russell's approximation [15,16], according to which

$$K(\varphi) = (1-\varphi)^{6.5}.$$

This approximation ignores the anisotropy of particle mobility in the magnetic field, although this circumstance does not lead to large errors. The influence of the mobility anisotropy

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on the transfer processes in magnetic fluids is an order of magnitude weaker than the influence of the anisotropy of thermodynamic forces [10,11] accounted for by the first term in Eq. (1).

The first term in Eq. (1) describes magnetophoresis and corresponds to the formula for the particle flux density, derived in the framework of the effective field approximation

$$\mathbf{j}_m = nD_0K(\varphi)L(\xi_e)\nabla\xi_e.$$

The effective field is used to describe both the rotational diffusion of particles (in the argument of the Langevin function) and their spatial drift (under the gradient sign). It is assumed that the influence of magnetic particles on a probe particle can be taken into account by replacing the intensity of the magnetic field H by its effective value $H_e = H_e(H,\varphi)$,which itself is dependent on the field intensity and local concentration of particles. Such approximation turns out to be very helpful, because it makes it possible to use formulas derived for dilute solutions (i.e., ignoring magnetodipole interactions) in the case of concentrated solutions. In particular, the expression for the time-averaged force $\mu_0 mL(\xi)\nabla H$, acting on the superparamagnetic particle in dilute liquid, is replaced in the case of concentrated liquids by the $\mu_0 mL(\xi_e)\nabla H_e$ expression, which leads to the formula for the magnetophoresis flux. The accuracy of this approximation depends mainly on the choice of the $H_e(H,\varphi)$ function. In the present work, the modified model of effective field (MMEF) is used. A comprehensive description of the model is offered in Refs. [17] and [18]. According to this model

$$H_e = H + \frac{M_L(H)}{3}\left[1 + \frac{1}{48}\frac{dM_L(H)}{dH}\right], \quad M_L = mnL(\xi), \tag{2}$$

$$\mathbf{M} = mnL(\xi_e)\frac{\mathbf{H}}{H}, \quad \xi_e = \frac{\mu_0 mH_e}{kT}, \tag{3}$$

where $n = 6\varphi/(\pi d^3)$ is the number concentration of particles. By the field strength H in Eqs. (2) and (3) is meant the strength of the field in the magnetic fluid (i.e. a superposition of demagnetizing fields and fields generated by external sources). Equations (2) and (3) have been subjected to repeated verifications by different methods. They fit well the experimental data on the initial susceptibility of magnetic fluids and are consistent with the results of magnetization calculations by the Monte Carlo and molecular dynamics methods in the range of small and moderate values of the parameter of magnetodipole interactions $\lambda \leqslant 2$, which is inherent in real magnetic fluids [18,19]. The effective field method is well suited to describing the translational Brownian diffusion, as was demonstrated in the works of Morozov [10,11] and J.-C. Bacri with coauthors [14]. In the right-hand side of Eq. (1), the coefficient in front of $\nabla\varphi$ enclosed in braces can be considered as an effective diffusion coefficient D of colloidal particles

$$D = D_0K(\varphi)\left[1 + \frac{2\varphi(4-\varphi)}{(1-\varphi)^4} - \varphi\frac{\partial^2(\varphi^2 G)}{\partial\varphi^2}\right]. \tag{4}$$

Its contribution related to the magnetodipole interparticle interactions (effective attraction) is equal to

$$\begin{aligned}\Delta D &= -D_0\varphi K(\varphi)\frac{\partial^2(\varphi^2 G)}{\partial\varphi^2}\\ &= -\frac{8}{3}\lambda^2\varphi D_0K(\varphi)\left(1 - \frac{5}{2}\lambda\varphi + \frac{\lambda^2}{25} + \cdots\right).\end{aligned} \tag{5}$$

Here we should note a significant difference between the two ways of describing mass transfer in Refs. [11,14] on the one hand, and in Ref. [12] on the other hand. In Refs. [11,14] the influence of interparticle interactions on the gradient diffusion of particles in the external magnetic field is taken into account by introducing the anisotropic diffusion coefficient, which depends on the mutual orientation of the magnetic field and the gradient of concentration. Unlike Refs. [11,14], the authors of Ref. [12] consider the strong anisotropy of thermodynamic forces by way of incorporation of the additional term in the mass transfer Eq. (1) responsible for magnetophoresis. This approach allows the researchers to ignore the anisotropy of the diffusion coefficients. The coefficient of gradient diffusion (4) remains a scalar function. Both these approaches give identical results when describing anisotropic effects caused by interparticle interactions (at least in the case of the flat layer and in the linear concentration approximation), but the approach of Ref. [12] is more universal because it can be used for arbitrary cavity shape and magnetic field configuration. The isotropic term (5) inserted into the diffusion coefficient obtained in Refs. [9] and [12] describes the additional effect, observed in the zero external field.

Expansion of the free energy for the system of interacting spherical dipoles in terms of particle concentration and parameter λ to an accuracy of terms quadratic in φ is given in Ref. [12]. Equations (1) and (4) provide a rather good description of the results of numerical experiments over a wide range of dimensionless parameters ($\lambda < 2$ and $\varphi < 0.4$), but do not allow modifications which would take into account the influence of aggregates because it is impossible to evaluate the free energy of a partially aggregated system.

In this paper, the isotropic correction for the diffusion coefficient is derived heuristically. The applied approach is less rigorous but allows us to write the system of equations, describing the interacting fluxes of individual particles and quasispherical aggregates in the framework of a two-fraction model. This system of equations is used to analyze the spatial distribution of particles in a thin layer of a concentrated magnetic fluid placed in the nonuniform magnetic field.

II. EFFECTIVE ATTRACTION BETWEEN PARTICLES

Averaging over the orientations of an anisotropic dipole-dipole interaction leads to an effective attraction between colloidal particles, which is one more reason for a drift of particles in a magnetic fluid in the case of inhomogeneous particle distribution. Since the corresponding contribution $\mathbf{j}_{at}$ in the particle flux is proportional to the concentration gradient, it can formally be taken into account by renormalization of the diffusion coefficient, as has been done in Ref. [9] and represented in formulas (4) and (5). As has been mentioned earlier, in contrast to the anisotropic effects described in

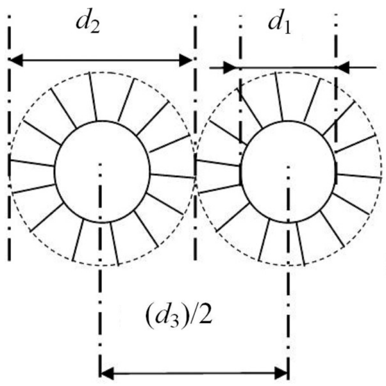

FIG. 1. Schematic view of two adjacent magnetic particles stabilized by oleic acid.

Refs. [11,14], this term does not vanish even in zero external field and, in this case of course, it is apparently isotropic.

An approximate structure of the formula allowing for terms nonlinear in concentration can be obtained with the help of qualitative reasoning. To this end, we will first find the magnetostatic energy of a trial colloidal particle placed in the magnetic fluid with magnetic susceptibility χ. This trial particle with diameter d_2 and diameter of the magnetic kernel d_1 resides in a spherical cavity with diameter $d_3 > d_2$, which is a space inaccessible to neighboring particles due to steric interactions (see Fig. 1). The kernel of the particle is magnetized spontaneously and generates in the surrounding space a nonuniform magnetic field, which orients the magnetic moment of the neighboring particles. We need to define the magnetic field strength inside the magnetic kernel $\mathbf{H}_1$ in the nonmagnetic gap between the kernel and the cavity wall $\mathbf{H}_2$, in the magnetic fluid $\mathbf{H}_3$ at a distance of $r > d_3/2$ and magnetostatic energy of the particle. The problem is solved by a standard method (see, for example, Ref. [20]) and therefore we will come directly to the expression for the trial particle energy:

$$U = -\mu_0 \mathbf{m}\mathbf{H}_1 = -\mu_0 \frac{mM_s}{3}\left[1 + \frac{2\chi}{3+2\chi}\left(\frac{d_1}{d_3}\right)^3\right], \quad (6)$$

where $M_s = 6m/(\pi d_1^3)$ is magnetization of the particle kernel. In the medium with inhomogeneous particle concentration the magnetic susceptibility is a function of coordinates suggesting that the particle is subject to the force

$$\mathbf{F} = -\nabla U = \frac{2\mu_0 m M_s}{(3+2\chi)^2}\left(\frac{d_1}{d_3}\right)^3 \nabla\chi, \quad (7)$$

which causes a particle flux density

$$\mathbf{j}_{at} = nb(\varphi)\mathbf{F} = nD_0K(\varphi)\frac{48\lambda}{(3+2\chi)^2}\left(\frac{d_2}{d_3}\right)^3 \nabla\chi$$
$$= nD_0K(\varphi)\frac{6\lambda}{(3+2\chi)^2}\nabla\chi. \quad (8)$$

Not counting the deformation of the protective shells, the radius of the sphere inaccessible to the centers of neighboring particles coincides with the full diameter of the particle and hence $d_2/d_3 = 0.5$.

The formula (8) was derived on the assumption that the probe particle is immersed into a continuous media with the static magnetic susceptibility χ. This susceptibility is defined as a response of a small macroscopic volume of liquid (averaged over time and over an ensemble of particles) on the permanent magnetic field. Magnetic particles in this volume are homogeneously oriented by the external magnetic field, which causes the appearance of the additional term in formula (2) that stands for the collective response (i.e., for the magnetodipole interparticle interactions). Actually, the magnetic field of the particle is not permanent due to the rotational fluctuations of its magnetic moment, and not locally homogeneous because its intensity depends on the polar angle and decreases by an order of magnitude at the distance of one particle diameter from its center. The real particle is surrounded by discrete media, and only a relatively small number of neighbor particles is affected by the alternative, strongly inhomogeneous magnetic field of the probe particle. So formula (8) can be treated as a coarse estimation of the density of the magnetophoresis flux. Nevertheless, several refinements can be made according to simple qualitative arguments.

Under the action of thermal fluctuations, the local magnetic field induced by the probe particle changes randomly and the moments of neighboring particles respond to its action with a delay. The influence of the fluctuations is rather strong because their characteristic frequency ω has an order of magnitude of the inverse relaxation time of the magnetic moment, $1/\tau$. This means that the substitution of the equilibrium value of susceptibility into Eq. (8) will lead to overvaluation of the particle flux density. To take into account thermal fluctuations of the local field we use the Debye formula for dynamic susceptibility

$$\dot{\chi} = \frac{\chi_L}{1+i\omega\tau}, \quad \chi_L = \frac{\mu_0 m^2 n}{3kT}, \quad (9)$$

where χ_L is the Langevin susceptibility. With the constraint $\omega\tau = 1$, χ in Eq. (8) should be replaced by the real part of dynamic susceptibility defined as $\chi_L/2 = 4\lambda\varphi$. Collective effects are not taken into account because the magnetic field of the probe particle is localized in its surrounding, it is nonuniform with respect to the polar angle, and rapidly decreases with the distance measured from the center of the probe particle. There is a small number of nearest-neighbor particles, and their magnetic moments are oriented in different directions. In this case

$$n\nabla\chi = 0.5\chi_L\nabla n = 4\lambda\varphi\nabla n.$$

Bearing in mind the above reasoning and assuming that the temperature of the magnetic fluid is uniform, we can rewrite Eq. (8) for a density of volume flux of particles:

$$\mathbf{j}_{at} = D_0K(\varphi)\frac{24\lambda^2\varphi}{(3+8\lambda\varphi)^2}\nabla\varphi. \quad (10)$$

The contribution to the diffusion coefficient related to an effective attraction between the magnetic dipoles is equal to

$$\Delta D = -D_0K(\varphi)\frac{24\lambda^2\varphi}{(3+8\lambda\varphi)^2}$$
$$= -\frac{8}{3}\lambda^2\varphi D_0K(\varphi)\left(1 - \frac{16}{3}\lambda\varphi + \cdots\right). \quad (11)$$

Formulas (5) and (11) describe one and the same physical quantity—the decrease of the particle diffusion coefficient associated with the effective attraction of the fluctuating magnetic dipoles which, however, has been obtained by different methods. Earlier the effect associated with magnetodipole interactions was theoretically predicted in Refs. [9,11,14] and experimentally confirmed in Refs. [13] and [14]. In the approximation linear with respect to particle concentration the right-hand side of Eq. (11) coincides with the accurate expansion (5) and thereby agrees well with the results of Refs. [9,12]. However, there is nearly a twofold overestimation of the coefficient in front of the term quadratic with respect to Langevin susceptibility $\chi_L = 8\lambda\varphi$. Most likely, this discrepancy is due to the fact that equation (6)–(8) have been derived without taking into account one more essential factors—the spatial inhomogeneity of the magnetic phase concentration in the vicinity of the trial particle caused by the local nonuniformity of the magnetic field. Generally speaking, this nonuniformity can be considered by solving a coupled (magnetic and diffusion) boundary value problem. The attempts to find analytical solution to this problem have failed. Therefore, to allow for inhomogeneity of susceptibility, the coefficient in front of $\lambda\varphi$ in the denominator of Eq. (10) has been corrected so that the right-hand sides of Eqs. (11) and (5) agree up to the terms quadratic in χ_L. In view of this correction the effective coefficient of particle diffusion in magnetic fluids is defined as

$$\Delta D = D_0 K(\varphi)\left[1 + \frac{2\varphi(4-\varphi)}{(1-\varphi)^4} - \frac{8\lambda^2\varphi}{3(1+1.25\lambda\varphi)^2}\right]. \qquad (12)$$

In the absence of the gravitational force and convective flows the density of volume flux of particles is written as

$$\mathbf{J} = D_0 K(\varphi)\Bigg\{\varphi L(\xi_e)\nabla(\xi_e) - \left[1 + \frac{2\varphi(4-\varphi)}{(1-\varphi)^4} - \frac{8\lambda^2\varphi}{3(1+1.25\lambda\varphi)^2}\right]\nabla\varphi\Bigg\}. \qquad (13)$$

We consider expression (12) to be the interpolation formula, which is valid in the range of moderate values of the parameter $\lambda \leqslant 1$. The main conclusion one can make out of this formula is that the isotropic term in the diffusion coefficient that describes the mutual attraction of magnetic dipoles in weak fields is negative. It always decreases the diffusion coefficient regardless of the values of λ and φ parameters. The formal expansion of Eq. (4) cannot clarify this issue, because the terms in the right-hand side of Eq. (4) are alternating and have slow convergence.

Strictly speaking, formula (12) can be used only in the case of weak fields, because formula (7) is incorrect for strong fields and can be used to estimate the order of magnitude only. Nevertheless, we have applied formula (7) in our analysis of the experimental data obtained for the case of relatively strong fields, because we are not aware of any other (more reliable) approximation. We do not expect a large error due to the heuristic approach to consideration of the effective attraction between the fluctuating magnetic dipoles, because for the majority of magnetic fluids the parameter of the magnetodipole interactions $\lambda \leqslant 1$. In this case, the last term in Eq. (12) introduces a rather small error for the diffusion coefficient. The situation may get worse only at $\lambda \geqslant 2$ (magnetic fluids with large particles); if so, this term will require a more thorough analysis and can potentially lead to spinodal decomposition of the system [12]. When $\lambda \leqslant 1$ the major input into the diffusion coefficient is made by the second term in Eq. (12) that stands for steric interactions.

III. CONSIDERATION OF AGGREGATES IN FRAMEWORK OF TWO-FRACTION MODEL

Equation (13) implies identity (monodispersity) of all colloidal particles inside the fluid. In real fluids, polidispersity of particles and their aggregation due to van der Waals and magnetodipole interparticle interactions require application of a more complicated theoretical model. As a first step towards the solution of this problem we can use a two-fraction model, in which the first fraction consists of individual particles and the second fraction consists of aggregates composed of few up to a few tens of particles. Previously this model has been successfully used to describe the results of tests of particle diffusion in magnetic fluids [21], spectra of dynamic susceptibility [22,23], and magnetophoresis in dilute magnetic fluids [5,6]. These studies have shown that experimental data can be adequately described only in the framework of the model which takes into account partial aggregation of particles. In this paper the two-fraction model is used to describe magnetophoresis in concentrated fluids. In what follows, we assume that aggregates are quasispherical seemingly formed as the result of defects of the protective shells. This is at least true for colloidal solutions of a magnetite stabilized by oleic acid [21–23]. Quasispherical aggregates are nearly insensitive to temperature variation (compared to the chains), behave as independent kinetic formations, and exert a dramatic effect on the physical properties of magnetic fluids. The characteristic size of quasispherical aggregates is several dozens of nanometers.

Magnetic fluids that undergo phase separation and stratify into dilute and concentrated phases under the influence of magnetic field are not studied in our work. The stratification of magnetic fluids (phase transition of "gas-liquid" type) includes formation of microdroplets of condensed phase (so-called drop-like aggregates) with characteristic dimensions 1–10 mkm [24]. Drop-like aggregates are macroscopic objects which are visible with an optical microscope and practically not involved in Brownian motion. The surface, which separates the drop-like aggregate from the surrounding colloidal solution, has tension that is responsible for the spherical shape of the aggregate in the absence of the external magnetic field. Under the influence of external magnetic field, the drop-like aggregate stretches along the field and in the case of strong field its shape resembles a needle. This transformation of drop-like aggregates leads to additional anisotropy of diffusion processes and rheological properties of magnetic fluids, which have been studied recently in experimental works [25,26]. On the other hand, quasispherical aggregates, which are being discussed in the present work, appear at the stage of preparation of magnetic fluids. Their characteristic dimensions are several dozen nm (i.e., they are about 2–3 orders of magnitude smaller than the drop-like aggregates), and application of the external magnetic field deforms these aggregates very slightly. Even in strong fields (∼100 kA/m), their relative deformation does not exceed 20%–30% and has no significant effect on the mobility of such aggregates [6].

In the framework of our model, polidispersity of particles in the first fraction and discrepancy of aggregates in sizes are neglected. This assumption should be clarified, because it restricts the applicability of the model to real polydisperse magnetic fluids. However, we believe that these restrictions are not critical to pose serious problems for the comparison of experimental and theoretical results. This assumption is justified by the fact that (in the approximation that is linear with respect to particle concentration) the increment in the effective viscosity of the solution and relative mobility of particles is defined by their total volume fraction irrespective of particle size distribution (inversely proportional), so that the results of calculations for average-size particles agree with the experimental data [21]. The possibility of applying the two-fraction model to the problems of particle magnetophoresis is less obvious, since the force exerted on a colloidal particle by the gradient of the magnetic field is proportional to its magnetic moment m (i.e., cubed diameter of the magnetic kernel [20,27]). It appears from this that the dependence is strong. However, there are two circumstances that clear up the situation. First, the largest particles making the greatest contribution to the fluid magnetization are combined into aggregates whose influence is treated separately. Second, small particles make inessential contributions to the system magnetization and their influence can be roughly taken into account. Size distribution of the remaining particles is rather narrow, so that they can be represented by one fraction. On the other hand, the difference in dimensions of individual particles and aggregates cannot be ignored. With reference to the problem under consideration it means that the fluxes of individual particles and aggregates must be described by separate equations.

An expression for the flux of individual particles is constructed on the basis that the rate of diffusion of aggregates is relatively small and their size is large compared to the size of a single particle. Under these conditions the influence of a separate aggregate is analogous to the influence of a stationary disk of the equivalent radius. In this case, the flux of individual particles is described by the expression analogous to Eq. (13), in which we introduce the correction factor allowing for a decrease in permeability of the medium. It can be shown that this correction factor (the ratio of the area of aggregate-free cross section to the total area) is given by

$$\sigma = 1 - \varphi_{\mathrm{agr}} = 1 - \frac{\varphi_2}{\gamma},$$

where φ_{agr} is the relative volume occupied by the aggregates, γ is the coefficient of particle packing in the aggregate. The equation describing density of the flux of individual particles takes the following form:

$$\mathbf{J}_1 = D_0 K(\varphi_1)\left(1 - \frac{\varphi_2}{\gamma}\right)\Big\{\varphi_1 L(\xi_e)\nabla(\xi_e) - \left[1 + \frac{2\varphi_1(4-\varphi_1)}{(1-\varphi_1)^4}\right]\nabla\varphi_1 + \frac{8\lambda^2\varphi_1}{3(1+1.25\lambda\varphi)^2}\nabla\varphi\Big\}, \tag{14}$$

where φ_1, φ_2 are the volume fractions of single and aggregated particles, respectively. The last term in Eq. (14) is written based on the knowledge that the particle flux caused by magnetodipole interactions is proportional to the concentration of individual particles and the gradient of magnetic susceptibility, viz. to the gradient of total concentration $\varphi = \varphi_1 + \varphi_2$. All other terms are derived from Eq. (13) by replacing the volume fraction φ by the volume fraction of individual particles φ_1.

In turn, the aggregates drift in a colloidal solution of small single particles, which can be viewed as a continuum with relative viscosity depending on the hydrodynamic concentration of these particles φ_1. The concentration dependence of viscosity η of the magnetic fluid in weak fields can be described perfectly well by the refined Chong-Christiansen approximation [28]

$$\eta_1(\varphi_1) = \eta_0\,[1 + 0.75\varphi_1/(\gamma_m - \varphi_1)]^2\,, \tag{15}$$

or by the Chow formula [29]

$$\frac{\eta_1(\varphi_1)}{\eta_0} = \exp\left(\frac{2.5\varphi_1}{1-\varphi_1}\right) + \frac{A\varphi_1^2}{1 - A\varphi_1^2\gamma_m}. \tag{16}$$

Here, η_0 is the viscosity of the carrier fluid, $A = 4.67$, and γ_m is the coefficient of dense packing of particles. Actually, the determination of γ_m poses a problem. Approximating the unit cell by the cubic centered lattice, Chow supposed that $\gamma_m = 0.68$, while the random close packing of the dry particles is equal to 0.64. As for us, we used $\gamma_m = 0.605$, which corresponds to random packing of particles in highly concentrated suspensions which are still capable of viscous flow [30,31].

Formally, the main disadvantage of approximations (15) and (16) is that they describe the suspension viscosity only in the case of zero magnetic field. The induced magnetic field leads to the appearance of the anisotropic increase of the effective viscosity of the suspension [32,33]. Here, this increase is not considered (neither is the anisotropic increase of particle mobility). We can set forth two additional arguments to validate this approach. First, the anisotropic increase of the effective fluid viscosity depends on the energy of the magnetic anisotropy of single-domain colloidal particles. For particles of magnetite (the most popular material for preparation of magnetic fluids) it proves to be small. The ratio of the magnetic anisotropy energy to the thermal energy is no more than one or two units for most of the magnetite particles. Therefore the Neel rather than Brown mechanisms of the magnetic moment relaxation is characteristic of such particles [22]. Under these conditions the anisotropic increase of viscosity (rotational viscosity coefficient) is an order of magnitude lower than the hard-dipole limit [34,35]. Second, the errors in the approximation expression (15), (16) or particle mobility $K(\varphi)$ affect only the dynamics of magnetophoresis and have no effect on the equilibrium (steady-state) concentration profiles. Since in this paper we focus on the equilibrium profiles, it seems reasonable to restrict ourselves to these approximations extending them, if necessary, to the case of arbitrary magnetic field. As for the Einstein diffusion coefficient for aggregates, with the supposition of their quasispherical shape it can be conveniently expressed in terms of diffusion coefficient for a single particle:

$$D_2 = D_1 \sqrt[3]{\frac{\gamma}{N}} \frac{K(\varphi_2/\gamma)}{\eta(\varphi_1)}, \tag{17}$$

where N is the average number of particles in the aggregate. At small or moderate values of the magnetodipole interaction parameter $\lambda \leqslant 1$ the correlation between the magnetic moment of aggregate particles is inessential, and thus the force acting on the aggregate in the external field is N times greater than the force acting on the single particle. In view of the above reasoning the density of the aggregated particle flux can be written as

$$\mathbf{J}_2 = D_1 \sqrt[3]{\frac{\gamma}{N}} \frac{K(\varphi_2/\gamma)}{\eta(\varphi_1)} \Bigg\{ N\varphi_2 L(\xi_e) \nabla(\xi_e) - \left[1 + \frac{2\varphi_2\gamma^2(4\gamma - \varphi_2)}{(\gamma - \varphi_2)^4}\right] \nabla\varphi_2 + \frac{8\lambda^2\varphi_2}{3(1+1.25\lambda\varphi)^2} \nabla\varphi \Bigg\}. \tag{18}$$

The dynamics of the magnetophoresis in the absence of convective motion is described by the system of two standard diffusion equations

$$\frac{\partial\varphi_i}{\partial t} = -\nabla \cdot \mathbf{J}_i, \tag{19}$$

with an obvious condition of impermeability of cell boundaries for colloidal particles

$$\mathbf{J}_{1n} = \mathbf{J}_{2n} = 0. \tag{20}$$

The system of equations (14), (18), (19) with boundary conditions (20) has been solved numerically using explicit calculation scheme by the finite volume method for a thin layer of magnetic fluid in the gradient magnetic field.

IV. NUMERICAL SIMULATION OF MAGNETOPHORESIS

Numerical simulation of the particle segregation in magnetic fluids was carried out to determine the role of the aggregates and to estimate sensitivity of the concentration profiles to parameters governing the aggregate properties: average volume fraction $\langle\varphi_2\rangle$ of the aggregated particles, average number N of particles in the aggregate, and packing index γ of particles in aggregates. These parameters cannot be obtained from independent measurements and in processing of magnetophoresis experiment data they were used as fitting parameters. The system of diffusion equations (19) with the boundary conditions (20) were solved by the method of finite volumes for a thin layer of the magnetic fluid which is oriented relative to the external gradient magnetic field, as shown in Fig. 2 (see Sec. V of the manuscript). The time step size was defined according to the standard condition of the stability of the explicit scheme. The computation was carried out until the system reached an equilibrium state. All parameters of the system, except for the aggregate parameters, were determined independently from auxiliary experiments (see the next section) or calculated. Here the basic challenge is the evaluation of the parameter of magnetodipole interactions λ for polydisperse system. According to Ref. [19] we calculated this parameter by the known particle concentration φ using the relations

$$\chi_L = 8\lambda\varphi, \quad \chi = \chi_L \left(1 + \frac{\chi_L}{3} + \frac{\chi_L^2}{144}\right), \tag{21}$$

where χ is the value of the initial magnetic susceptibility measured by the mutual inductance bridge.

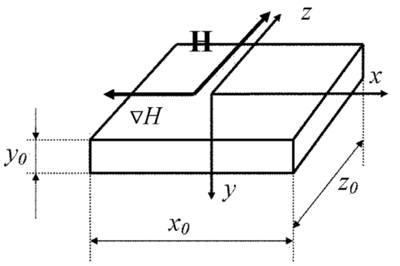

FIG. 2. Schematic representation of measuring cell with magnetic fluid.

Figure 3 shows the results of numerical simulation at a fixed value of the average particle concentration $\langle\varphi\rangle$. The profiles depicted in Fig. 3 correspond to different values of N. The simulation was done for a weakly aggregated fluid under the assumption that the aggregates contain only 5% of particles. As might be expected, the number of particles in the aggregate has a profound effect on the concentration profiles: a smooth distribution $\varphi = \varphi(x)$, taking place at $N < 10$, gives way to a stepwise distribution at $N \geqslant 15$. The reason of this phenomenon is quite clear—with increasing number of particles in the aggregate, magnetophoresis becomes stronger according to Eq. (18).

The packing index of aggregated particles γ is also an important parameter, although its influence on the concentration profile is not so strong as the influence of the number of aggregated particles. All other things being equal, the packing index affects the intensity of steric interactions: with a decrease in γ the aggregates tend to increase in size and

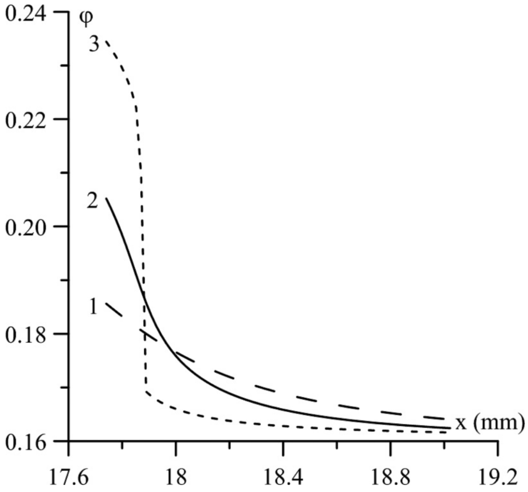

FIG. 3. Equilibrium distribution of particles along the cell at different numbers of particles in the aggregate. $\langle\varphi\rangle = 0.171$, $\langle\varphi_2\rangle = 8.5 \times 10^{-3}$, $\gamma = 0.6$. Curve 1 corresponds to $N = 7$, curve 2 corresponds to $N = 10$, and curve 3 corresponds to $N = 15$. The average strength of the magnetic field $\langle H_0\rangle = 95$ kA/m and the mean gradient of the field strength is 3×10^6 A/m^2.

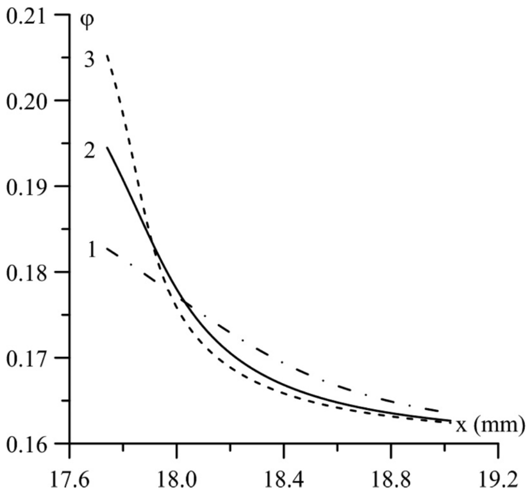

FIG. 4. Equilibrium distribution of particles along the cell at different packing index of aggregated particles. $\langle\varphi\rangle = 0.171$, $\langle\varphi_2\rangle = 8.5 \times 10^{-3}$, $N = 10$. Curve 1 corresponds to $\gamma = 0.3$, curve 2 corresponds to $\gamma = 0.5$, and curve 3 corresponds to $\gamma = 0.6$. The average strength of the magnetic field $\langle H_0\rangle = 95$ kA/m, mean gradient of the field strength 3×10^6 A/m^2.

the effects of excluded volume are enhanced. An increase of steric interaction should lead to a decrease of particle segregation [12], which is the case. The smaller is γ (the larger the excluded volume), the lower is the spatial inhomogeneity of concentration in the equilibrium (Fig. 4).

V. CELL GEOMETRY

The experimental measuring cell is represented schematically in Fig. 2. The cell represents a thin horizontal layer of magnetic fluid enclosed between two plane-parallel glass plates and oriented along the magnetic lines of force (along the z axis). The gradient of magnetic field intensity was directed along the x axis. The layer thickness y_0 (from 20 to 100 μm) was small compared to the horizontal dimensions of the cell ($x_0 = z_0 \approx 1.5$ mm) and therefore the distribution of concentration in the cell could be considered plane with a good degree of accuracy despite the action of the gravitational field ignored by equations (14) and (18). A small thickness of the layer allows applying the photometric technique for magnetite concentration measurements described in detail in Refs. [5,6]. The local concentration of magnetite was defined by the extent of absorption of a thin (0.02 mm) light beam penetrating through the layer of the fluid. As it was previously shown in Ref. [5], light absorption in magnetic fluids is described well by the Bouguer-Lambert-Beer law. The photometric method apart from high sensitivity and great resolving power has one more point in its favor: the useful signal is not sensitive to polydispersity of particles because it is independent of the ratio of coarse to fine particles in the fluid, provided that the sizes of the particles are small compared to the length of the light wave. The coefficient of light absorption is proportional to the local concentration of magnetite. Horizontal orientation of the plane layer was chosen to minimize the influence of the gravitational field. To ensure minimal reduction of demagnetizing fields, the external magnetic field was oriented along the fluid layer. Since the gravitational and demagnetizing fields can essentially affect the results of experiment and thereby make their interpretation difficult, we will examine their influence on the particle segregation more thoroughly.

The demagnetizing field is known as a gain of the field intensity inside the substance caused by refraction of force lines at boundaries of a body. The existence of the demagnetizing field is the main reason for a difference between the strength of the field inside the substance and that of the field generated by an external source. In the case of homogeneous magnetization the value of the demagnetizing field is defined by the shape of the body and for geometrically similar bodies is independent of their dimensions. If the external field is homogeneous and the body has the shape of a triaxial ellipsoid or its ultimate shapes (thin plate, sphere, and circular cylinder of infinite length), then the strength of the field inside the body is related to the strength of the external field H_0 by a simple equation

$$H = H_0 - \kappa M,$$

where $\kappa =$ const. is the demagnetization factor and depends only on the ratio of the ellipsoid axes and their orientation in the magnetic field. The demagnetization factor is maximal ($\kappa = 1$) for a plane plate magnetized in the transverse direction and is minimal ($\kappa \to 0$) for a long cylinder or plate extending along the force lines. In the latter case the demagnetizing fields can be ignored whereas in all other cases they must be taken into account because they produce effects which are similar in order of magnitude to the effects of magnetodipole interactions. The evaluation of magnetic fields in a magnetic fluid of inhomogeneous density is a rather complicated problem [36]. Therefore, a simpler way of solving the problem is to select the geometry of the measuring cell (and its orientation in the external field) so that it can provide a rather small value of demagnetizing fields. In our measuring cell (Fig. 2), considerable demagnetizing fields are concentrated near the end surfaces $z = \pm z_0/2$ normal to the magnetic field lines. The region with minimal demagnetizing fields locates in the middle part of the layer (in cross section $z = 0$). The concentration profile $\varphi = \varphi(x)$ in this cross section is the objective of our investigation.

The effect of demagnetizing fields on the concentration profiles can be estimated according to real experimental conditions: average strength of the magnetic field $\langle H_0\rangle = 95$ kA/m, mean gradient of the field strength 3×10^6 A/m^2, the saturation magnetization M_∞ of the colloids used in the experiment was about 50 kA/m, and the typical relative concentration drop along the measuring cell in the state of thermodynamic equilibrium $\Delta\varphi/\langle\varphi\rangle$ was about 10% (see, for example, Fig. 5). The magnetic field used in our experiment can be considered strong in the sense that the corresponding Langevin parameter $\xi \approx 6 \gg 1$ and magnetization changes weakly with the local strength of the field. Under these conditions the magnetization and concentration inhomogeneity have the same order of smallness: $\Delta M/\langle M\rangle \approx \Delta\varphi/\langle\varphi\rangle \approx 0.1$. For an approximate estimate of the demagnetizing field we can neglect this inhomogeneity and use the known result for the demagnetizing field ΔH generated by a rectangular parallelepiped with homogeneous magnetization $\langle M\rangle$ [37]. Taking into account the peculiarity of a thin layer ($y_0 \ll x_0$, $y_0 \ll z_0$), the wanted expression for the section $z = 0$ can

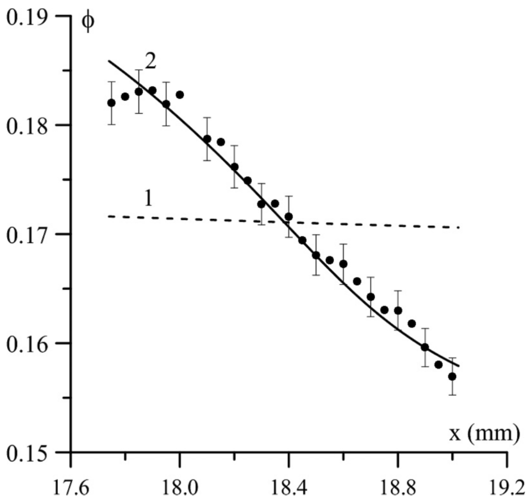

FIG. 5. Distribution of particles in a kerosene-based magnetic fluid with an average volume fraction of the magnetite $\langle\phi\rangle = 0.171$. Curve 1 is for $\langle\phi_2\rangle = 0$, curve 2 is for $\langle\phi_2\rangle = 0.1\langle\phi\rangle$, $N = 10$, $\gamma = 0.34$. Dots show the experimental results.

be written as

$$\Delta H = -\frac{\langle M\rangle}{\pi}\frac{y_0}{z_0}\left\{\frac{(2x+x_0)}{\sqrt{(2x+x_0)^2+z_0^2}} - \frac{(2x-x_0)}{\sqrt{(2x-x_0)^2+z_0^2}}\right\}. \tag{22}$$

According to (22) the maximum value of the demagnetization factor is as high as 3×10^{-2} (the ratio $\Delta H/H \leqslant 1.5\times10^{-2}$ for a layer 100 μm thick) and decreases in proportion to the layer thickness in other cases. In view of this fact we can consider the demagnetizing field as a small correction and in the following restrict ourselves to its approximate estimation using formula (22).

The influence of the gravitational field on the distribution of particles in the measuring cell can be estimated by making use of the known solution of the problem obtained within the approximation for dilute magnetic fluid, which is valid for a cell of arbitrary shape and the field of arbitrary configuration [5,12]. As applied to the section $z = 0$ the spatial distribution of particles in the magnetic fluid is described by the following simple expression:

$$\varphi(x,y) = A\frac{\sinh\xi(x,y)}{\xi(x,y)}\exp(-G_\gamma y). \tag{23}$$

Here, the Langevin parameter ξ is considered to be the known function of coordinates and the normalization constant A is expressed in terms of the average concentration of particles in the cell. In the limits of weak magnetic fields, formula (23) is transformed into the expression for barometric distribution. As it follows from (23), the isolines of equal concentration $y = y(x)$ are determined implicitly by

$$\frac{\sinh\xi}{\xi}\exp(-G_\gamma y) = \text{const}. \tag{24}$$

Under the conditions of our tests $\xi = \xi(x)$, and in the absence of the gravitational force these lines are arranged vertically. Under the action of the gravitational field the concentration isolines are inclined with respect to the vertical by a small angle β. By differentiating (24) we obtain

$$\tan(\beta) = \left(\frac{dy}{dx}\right)^{-1} = \frac{G_\gamma}{\xi L(\xi)}\left(\frac{1}{H}\frac{\partial H}{\partial x}\right)^{-1}. \tag{25}$$

The substitution of the relevant experimental parameters into (25) ($G_\gamma = 3$ m^{-1}, $\xi = 6$, $H = 96$ kA/m, $\partial H/\partial x = 3\times10^6$ A/m^2) gives $\beta \approx 1.6\times10^{-2} \ll 1$. The inclination of the concentration isolines can be interpreted as an additional uncertainty in the horizontal coordinates, not exceeding yet $y_0\beta/2 \approx 0.8$ μm. It is obvious that such a small uncertainty in the coordinates can be neglected with a good degree of accuracy. In the case of concentrated solutions one might expect a further decrease in the angle of isoline inclination, since the magnetodipole interactions increases the effective Langevin parameter and the steric interactions have equal effect on the numerator and denominator of the right-hand side of Eq. (25). For the same reason formula (25) can also be used in the case when the magnetic fluid contains both the individual drifting particles and drifting aggregates including in the average N particles. In this case, each of the particle fluxes associated with the magnetophoresis and particle sedimentation in the field of gravity increase by N times although their ratio remains unchanged.

VI. RESULTS OF EXPERIMENTS

For tests we used the concentrated magnetic fluids, which were prepared by the standard chemical precipitation method. The average volume fraction of the magnetite in the solution varied in the range from 0.1 to 0.2, and saturation magnetization varied from 40 to 60 kA/m. A magnet with pole pieces was used as a source of the constant gradient magnetic field. The average magnetic moment of particles and the average diameter of the magnetic kernel were determined from the magnetogranulometric analysis using the technique described in Ref. [38]. The photometric method for measuring concentration used in this paper allows us to determine the volume fraction ϕ of the magnetite whereas the hydrodynamic concentration φ, which is the volume fraction of particles including the protective shells, enters into Eqs. (14)–(19). A transition from one concentration to the other can readily be accomplished due to the fact that the thickness δ of the protective monolayer, consisting of the molecules of oleic acid, is the known constant value. In particular, in the case of using the oleic acid as a stabilizer, $\delta = 2.1$ nm. For a polydisperse fluid we obtain

$$\varphi = \frac{\langle(d_m+2\delta)^3\rangle}{\langle d_m^3\rangle}\phi, \tag{26}$$

where d_m is the diameter of the hard kernel of the colloidal particle, and angle brackets denote averaging over the particle ensemble. The coefficient in front of the right-hand side of Eq. (26) was calculated for each sample based on the results of magnetogranulometric analysis.

In our tests, the particles under the action of the nonuniform magnetic field drift slowly ($\tau_D \sim 10^5$ s) towards the left side

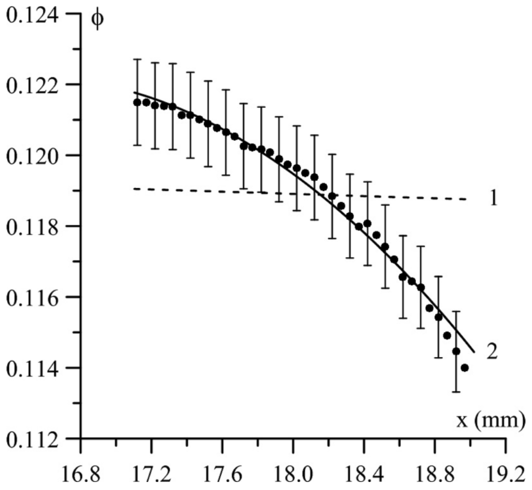

FIG. 6. Distribution of particles in a polyethylsiloxane-based magnetic fluid with an average volume fraction of the magnetite $\langle\phi\rangle = 0.1187$. Curve 1 is for $\langle\phi_2\rangle = 0$, curve 2 is for $\langle\phi_2\rangle = 0.4\langle\phi\rangle$, $N = 45$, $\gamma = 0.38$. Dots show experimental results.

wall of the cell, which in the course of time leads to the formation of the equilibrium concentration profile. Depending on the conditions of the experiment, the time of the equilibrium profile formation varied from 3 to 4 days up to two weeks. The equilibrium concentration profiles for three samples of the magnetic fluid prepared on the basis of kerosene are shown in Figs. 5 and 7 and those on the basis of polyethylsiloxane are shown in Fig. 6. The solid lines in the figures were obtained by solving numerically Eqs. (14), (15), (18), and (19). The average volume fraction $\langle\phi_2\rangle$ of the aggregated particles, the average number N of particles in the aggregate, and the packing index γ of particles in the aggregate were used as fitting parameters. They were determined from the condition of the minimum mean square deviation of the computed curve from the experimental points. Dashed lines correspond to the absence of aggregates and the difference between the dashed and solid lines demonstrates the contribution of aggregates to spatial inhomogeneity of the magnetic fluid. As it is evident from the figures, this contribution is crucial. Neglect of aggregation reduces the calculated concentration difference by two orders of magnitude against the experimental data. Thus, including aggregates into the simulation is a necessary condition for a correct estimation of the particle segregation both in diluted [5,6] and concentrated systems. In the general case, the effect of the aggregates on the spatial segregation of particles manifests itself as two competing mechanisms. On the one side, the appearance of aggregates increases the excluded volume (increases steric interactions), which stabilizes the system [12]. On the other hand, aggregation leads to a multiple strengthening of magnetophoresis and increase of particle segregation. Under the conditions of our experiments the second mechanism at the back of this effect prevails over the first one. Depending on the relation between the aggregate parameters, the concentration profile can be close to a linear profile (Fig. 5), can be convex (Fig. 6), or can be concave (Fig. 7).

For all three samples of the magnetic fluid the calculated profiles describe the experimental data with sufficient accuracy except for the periphery zone, in which the strength of the magnetic field is maximal and the volume fraction of the magnetite is about 0.2. The reason for this discrepancy is still unclear. In our opinion, the most probable reason is a rather high degree of inaccuracy of equations (14) and (18) (underestimation of the excluded volume and steric interactions) at high total concentration of particles. A considerable disagreement is observed for kerosene-based samples, in which the volume fraction of particles φ (excluded volume) is ≈0.4 in the case when aggregates are absent and ≈0.5 when about 10% of particles are involved in the formation of aggregates with the packing index $\gamma = 0.4$.

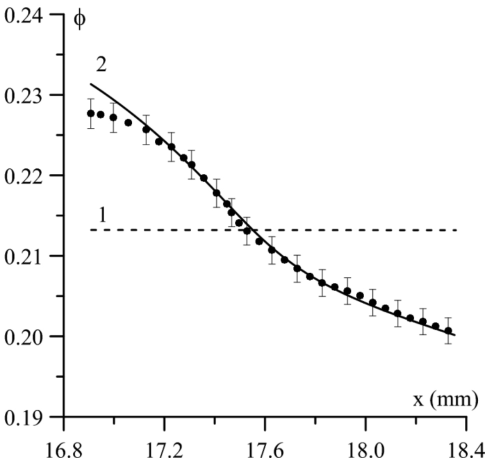

FIG. 7. Distribution of particles in a kerosene-based magnetic fluid with an average volume fraction of the magnetite $\langle\phi\rangle = 0.213$. Curve 1 is for $\langle\phi_2\rangle = 0$, curve 2 is for $\langle\phi_2\rangle = 0.1\langle\phi\rangle$, $N = 10$, $\gamma = 0.54$. Dots show experimental results.

Before concluding this section we should note one more important feature of the problem on magnetophoresis of colloidal particles in magnetic fluids: fitting parameters N, $\langle\phi_2\rangle$, and γ are not derived uniquely from the results of experiment. We may find at least several triplets of numbers which allow us to describe the experimental profile $\phi = \phi(x)$ with nearly the same accuracy. As an example, four such triplets, corresponding to the data of Fig. 6, are presented in Table I.

As seen from the Table I, the average number and percentage of the aggregated particles found by the fitting technique can differ essentially. This suggests that the inverse problem—determination of the aggregate parameters from the concentration profiles—is not correct, because it may have several solutions. To eliminate the parameter ambiguity we need to obtain additional data; for example, from tests on particle diffusion in a zero magnetic field [21], rheological measurements, or analysis of dispersion curves for dynamic

TABLE I. Sets of fitting parameters describing with nearly the same accuracy the experimental data for a magnetic fluid on the basis of polyethylsiloxane.

No.	N	γ	$\langle\phi_2\rangle/\langle\phi\rangle$
1	45	0.38	0.40
2	45	0.42	0.43
3	75	0.50	0.62
4	280	0.49	0.72

susceptibility [23]. Nevertheless, if we exclude from consideration the last row in Table I, the remaining parameters agree in order of magnitude with the results obtained by other methods [21–23].

VII. CONCLUSION

We have proposed the system of equations (14), (18), and (19) to describe the processes of magnetophoresis and diffusion of particles in a concentrated magnetic fluid containing aggregates. These equations take into account the steric, magnetodipole, and hydrodynamic interparticle interactions and agree quite well with the results of laboratory experiments of particle segregation in magnetite colloids in a gradient magnetic field. A rather wide range of particle size distribution observed in commercial magnetic fluids is not taken into account in these equations, which can lead to errors in calculations. At present we are unable to give an estimate of possible errors. However, for magnetic fluids used in our experiments the error is too small to cause considerable disagreement between the results of simulation and experimental data.

We also do not expect a large error due to the heuristic approach to consideration of the effective attraction between the fluctuating magnetic dipoles, because for the majority of magnetic fluids the parameter of the magnetodipole interactions $\lambda \leqslant 1$. In this case, the last term in relation (12) introduces a rather small error for the diffusion coefficient. The situation may get worse only at $\lambda \geqslant 2$ (magnetic fluids with large particles); if so, this term will require a more thorough analysis.

By analogy with Ref. [12], the allowance for steric interactions has been made in the Carnagan-Starling approximation for the system of hard spheres [39] and for magnetodipole interactions—in the framework of the modified model of the effective field [18,19]. Both approximations agree well with the results of laboratory and numerical experiments. Some problems may arise only in the range of high particle concentration ($\varphi \geqslant 0.4$) and high energies of dipole-dipole interactions ($\lambda \geqslant 2$).

The laboratory experiments were carried out in thin horizontal layers of the magnetic fluid providing the minimum level of demagnetizing fields, the minimum effect of the gravitational field, and the possibility to use the photometric technique for measuring local particle concentration. Three samples of the concentrated magnetic fluid have been examined and in all cases the measured concentration difference inside the cell was two orders of magnitude higher than the difference calculated without taking into account the aggregates. This result is very significant, implying that, without consideration of aggregates (if only they exist in a magnetic fluid), a correct computation of the concentration profiles is impossible.

ACKNOWLEDGMENTS

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