

Viscoelastic properties of ferrofluidsD. N. Chirikov,¹ S. P. Fedotov,² L. Yu. Iskakova,¹ and A. Yu. Zubarev¹¹*Urals State University, Lenina Avenue, 620083 Ekaterinburg, Russia*²*Manchester University, Manchester, United Kingdom*

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The paper deals with theoretical study of non linear viscoelastic phenomena in ferrofluids placed in magnetic field. Our attention is focused on the study of nonstationary flow and Maxwell-like relaxation of the macroscopical viscous stress after alternation of the shear rate. We propose that these phenomena can be explained by finite rate of evolution of chainlike aggregates, consisting of the ferrofluid particles. Statistical model of the chains growth-disintegration is suggested. In this model the chain-single particle mechanism of the chains evolution is considered, the effects of the chain-chain interaction are ignored. The proposed model allows us to estimate the time-dependent function of distribution over number of particles in the chain. Having determined this function and using methods of hydromechanics of ferrofluids with chainlike aggregates, we have studied evolution of the ferrofluid viscosity after stepwise alternation of the fluid shear rate. The estimated time of relaxation is in a reasonable agreement with experimental results. Thus, our analysis shows that the observed macroscopical viscoelastic phenomena in ferrofluids can be provided by evolution of the chain ensemble.

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I. INTRODUCTION

Magnetic fluids (ferrofluids) present colloidal suspensions of single-domain ferromagnetic particles in a carrier liquid. The typical diameter of the particles is about 10–15 nm. In order to prevent the irreversible coagulation of the particles under the action of the colloidal dispersion forces, they are covered by special layers which screen these forces. For many modern ferrofluids the surface shells consist of surfactant molecules; the typical thickness of these layers is about 2–2.5 nm.

Ferrofluids attract considerable interest of investigators and engineers due to rich set of their unique physical properties, valuable for many modern technologies. Discussion of methods of ferrofluids synthesis as well as backgrounds of physics of these systems can be found in the book [1]. Modern state of physics of ferrofluids and various fields of their practical applications are discussed in [2].

One of the interesting and important features of ferrofluids is their ability to change rheological properties under the action of external magnetic field. The first theories of the magnetorheological effects in ferrofluids [3,4] deal with very dilute systems, where any interactions between the particles can be ignored.

The maximal magnetoviscous effect predicted by these models does not exceed several per cent. However, experiments, carried out with various commercial ferrofluids in recent decades, demonstrate increase of their viscosity under the field by one-two orders of magnitude [5,6]. Especially strong magnetoviscous effects are observed when magnetic field is aligned along gradient of the fluid shear flow.

Analysis shows that the strong magnetoviscous effects can take place due to appearance of heterogeneous aggregates consisting of the ferrofluid particles [6–9]. Two types of the structures in ferrofluids are well-known—the linear chains and the dense bulk “drops” as well. Theoretical and experimental study of these structures can be found in Refs. [6–14]. Both of them can induce strong magnetorheological

effects in ferrofluids [8]. Unfortunately, there is no universally recognized theory which would be able to predict either chains or drops dominate in ferrofluids under given conditions.

It should be noted that experiments [15,16] demonstrate various rheological effects which, in principle, cannot be explained in the framework of the classical single-particle model. However they can be explained, at least qualitatively, under assumption of appearance of internal heterogeneous structures.

Theories of magnetorheological properties of ferrofluids are mainly devoted to steady flow of these systems. However, from scientific as well as practical viewpoints, study of nonstationary flow of ferrofluids presents significant interest. Experiments [16] with typical commercial ferrofluids have demonstrated pronounced viscoelastic (relaxation) effects with the time of rheological relaxation about ten seconds. These magnitudes of the relaxation time are about four to five decimal orders of magnitude greater than the classical theories [3,4] predict.

Some attempts of theoretical description of the viscoelastic properties of ferrofluids with chainlike aggregates have been done in Refs. [17–19]. In these models, the viscoelastic effects have been explained by the finite time of the chain reorientation after alternation of the fluid shear rate. The time of viscoelastic relaxation obtained from the models [17–19] is significantly closer to the experimental results of [16] than that of the models [3,4]; nevertheless, the calculated relaxation time is one to two orders of magnitude less than that detected in experiments [16]. Thus one can conclude that microscopical physical cause of the strong viscoelastic phenomena in ferrofluids is still not understood even qualitatively.

It should be noted that phenomenological models of the nonstationary viscoelastic flow of ferrofluids with chainlike aggregates has been suggested in Refs. [20,21]. As usual, the phenomenological models propose macroscopical equations of dynamical behavior of ferrofluids, but do not allow estimating characteristic parameters (effective viscosity, time of

relaxation, etc.), which figurate in these equations, as well as dependences of these parameters on applied field, shear rate, etc. Moreover, the phenomenological approach can lead to constructive results only when mathematical forms of equations for the stress and ferrofluid magnetization are known. However, for ferrofluids the nonlinear functional forms of relation between these magnitudes *a priori* are unknown. From our point of view only microscopical analysis is able to uncover internal, intimate mechanisms of formation of macroscopical nonlinear rheological behavior of ferrofluids and gives possibility to derive the macroscopical governing equations.

One can suppose that experimentally detected relaxation viscoelastic phenomena in ferrofluids appear due to finite rate of evolution of ensembles of heterogeneous aggregates after alternation of macroscopical shear rate of the fluid. Characteristic time of evolution of these ensembles can determine the time of macroscopical viscoelasticity of the fluid.

In this paper, we propose a simple microscopical model of viscoelastic properties of a ferrofluid with the chainlike aggregates under assumption that these properties are due to evolution of the chain ensemble. The proposed model is based on statistical analysis of kinetics of the chains growth/disintegration in the shear flowing ferrofluid. Our attention is focused on the effects of the Maxwell-like relaxation of the macroscopical viscous stress after alternation of the imposed shear rate. That is why we suppose that length of the chains is much less than size of the ferrofluid sample. Elastic phenomena, which appear when the chains (or other heterogeneous structures) overlap the channel with the ferrofluid, are not considered here.

The structure of this paper is the following. In part II, we consider evolution of the chains in a ferrofluid after alternation of shear rate and/or applied magnetic field. In part III, by using microscopical approach, we determine the macroscopical stress in the ferrofluid. Evolution of the fluid viscosity after stepwise alternation of the shear rate is considered in part IV.

In presented model we use the same approximations as in the Refs. [7,8,16–19,22,23]. In spite of the fact that these approximations definitely oversimplify the physical situation which takes place in real ferrofluids, they have allowed to get quite reasonable agreement with experiments for the stationary magnetoviscous effects [5–7,9,16,22] and rate of the chain growth [23].

The main simplifications of the approach are the following. First, we model the ferrofluid as suspension of identical ferromagnetic spheres in a carrier liquid. One needs to note that the modern ferrofluids are, as a rule, polydisperse systems, very often with wide distribution over particle sizes. The typical diagrams of the particle size distribution can be found in Ref. [5]. Unfortunately account of many fractions of the particles with different sizes leads to very complicated and cumbersome mathematics. However it is known (see, for example [5]) that the macroscopical magnetorheological effects in ferrofluids are provided by the fractions of relatively big particles. As a rule, volume concentration of these particles in ferrofluids is quite small—about 1–2 %. That is why, for maximal simplification of calculations, we will consider only monodisperse ensemble of the big particles, as-

suming that their volume concentration is between 1% and 2%.

Second, it is well known [4,5] that magnetic moment m of the big ferrofluid particle is frozen in its body, i.e., rotates together with it. We will assume that this condition is fulfilled.

Third, we will ignore any interactions between the chains. This approximation is justified by the small total concentration of the big ferroparticles in the system.

Next, we will neglect the thermal flexibility of the chains and consider them as rigid rodlike aggregates. Magnetic moments of all particles in the chain are supposed to be aligned along the chain axis. In part, it means that the magnetic interaction between the nearest particles in the chain is assumed much greater than the thermal energy kT . The last condition is necessary for formation of any heterogeneous aggregates in ferrofluid.

II. KINETICS OF EVOLUTION OF THE CHAIN ENSEMBLE

Stationary distribution over number of particles in the chains. The chainlike aggregates can be considered as specific heterogeneous fluctuations of density. That is why the number of particles in a chain is stochastic quantity which is determined by competition between magnetic attraction of the particles, their thermal motion and hydrodynamical destruction. Theoretically, in macroscopically motionless system a chain can include infinite number of particles. In the shear moving ferrofluid too long chains must be destroyed by the viscous hydrodynamical forces. Competition of the hydrodynamical and magnetic forces leads to existence of a finite maximal number n_c of the particles in the chain. In the case when applied magnetic field is aligned along gradient of the fluid flow, this number has been estimated in [5,7] as

$$n_c \approx \sqrt{\varepsilon \frac{D_r}{\dot{\gamma}}} \quad \varepsilon = \frac{\mu_0 m^2}{2\pi d^3 kT}, \quad D_r = \frac{kT}{\pi \eta_0 d^3}. \quad (1)$$

Here d is hydrodynamical diameter of the particle, defined with account of the stabilizing shells, η_0 is viscosity of the carrier liquid, μ_0 is the vacuum permeability. Parameter ε presents dimensionless, with respect to the thermal energy kT , energy of the magnetodipole interaction between the nearest particles, D_r is the coefficient of rotational diffusion of the single particle, $\dot{\gamma}$ is the shear rate. Below, for simplification, we will use estimate Eq. (1) for n_c without any multiplier.

Let us denote the number of n -particle chains per unit volume of the system as g_n . If the convective motion of the particles near the chain is much weaker than their diffusion motion (the Pecklet number is small), then the scenario of the particles integration/disintegration from the chains is quite similar to that in the motionless media. That is why the stationary distribution function g_n can be found with the same way as the equilibrium function, taking into account the restriction [Eq. (1)] on the number of particles in the chain.

Obviously, the equilibrium distribution function must correspond to the principle of free energy minimum. Since con-

centration of the aggregated particles in the ferrofluid is supposed small, we may neglect any interactions between the chains. In this approximation the free energy of a unit volume of the system can be presented as (see, also [7,17–19]),

$$F = kT \sum_{n=1}^{n_c} \left(g_n \ln \frac{g_n v}{e} + g_n f_n \right), \quad v = \frac{\pi d^3}{6}. \quad (2)$$

The first term in the brackets [Eq. (2)] corresponds to the entropy of ideal gas of the chains, f_n is the dimensionless free energy of the chain. This magnitude is determined by interaction between particles in the chain as well as by their interaction with applied magnetic field. By using the well known approximation of the nearest neighbors, in the framework of approximation of the straight chains, this free energy can be estimated as

$$f_n = - \left[\varepsilon(n-1) + \ln \frac{\sinh(\kappa n)}{\kappa n} \right],$$

$$\kappa = \mu_0 \frac{mH}{kT}. \quad (3)$$

Here H is the magnetic field, dimensionless parameter κ is the ratio of energy of interaction between the particles with the field to the thermal energy kT . The first term in the brackets [Eq. (3)] corresponds to the energy of magnetic interaction between particles in the chain, estimated in the nearest-neighbors approximation. The second term is the Langevine free energy of the chain with the total magnetic moment (mn) in the field H .

Substituting Eq. (3) into Eq. (2), we get

$$F = kT \sum_{n=1}^{n_c} g_n \left\{ \ln \frac{g_n v}{e} - \left[\varepsilon(n-1) + \ln \frac{\sinh(\kappa n)}{\kappa n} \right] \right\}. \quad (4)$$

We determine the stationary distribution function from the condition of minimum of the free energy [Eq. (4)] with respect to g_n under the obvious normalization condition

$$\sum_{n=1}^{n_c} n g_n = C = \frac{\varphi}{v}. \quad (5)$$

Here C is the total number of the particles in the unit volume, φ is their hydrodynamical volume concentration defined with account of the shells on the particles.

Minimizing Eq. (4) under condition (5), after standard transformations we get

$$g_n = \frac{1}{v} \frac{\sinh(\kappa n)}{\kappa n} X^n \exp(-\varepsilon), \quad (6)$$

where X is undefined Lagrange multiplier. One can show, combining Eqs. (4)–(6), that $kT \ln X$ is chemical potential of the ferrofluid particles.

In order to determine X one needs to substitute Eq. (6) into the Eq. (5). As a result, we come to a transcendental equation with respect to X , which can be solved numerically.

Flux of free particles toward the chain. Let us pass now to analysis of kinetics of the chains growth. This kinetics is determined by the competition between flux of the particles

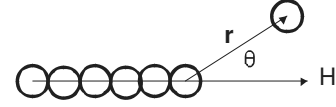


FIG. 1. Sketch of the chain and free particle.

toward the chain and evaporation of the particles from this chain. Here, in order to determine the flux of the particles to the chain, we will use the main ideas of the model [23] which have been successfully used at the analysis of evolution of chains of polarized particles in electric field.

In the framework of this model we suppose that the chain growth takes place due to the “chain-particle” aggregation and ignore the “chain-chain” way of aggregation. Indeed, first, for the realistic situations concentration g_1 of the single particles is significantly more than concentration g_n of any chains in the system [17–19]. Second, the hydrodynamical mobility of the single particle is more than mobility of a chain. Therefore, the flux of the single free particles to the chain must be significantly more than the flux of any other chains.

Additionally, we will assume that the particle “adsorb” only at extremities of the chain. Thus we neglect probability of the lateral aggregation. This assumption is justified by the fact that in the applied field average magnetic moments of the particles and chains are parallel. That is why for the lateral aggregation, the particle must overcome a potential barrier with the height which is significantly greater than kT . The probability to overcome this barrier due to diffusion effects is much less than unity. Thus the probability of particle attachment to the chain pole is significantly more than probability of attachment to the chain lateral surface.

It should be noted that too long chains in ferrofluid are thermodynamically unstable—they must transform to a dense drop or globule [24]. We assume that probability of appearance of these drops is negligible.

Our first aim now is to determine the flux of free particles toward the chain, i.e., number of the particles which attach to the chain extremities per unit of time. The flux of the particles evaporating from the chain we will determine later.

Let us consider a chain consisting of n particles surrounded by free particles (Fig. 1).

By c_∞ we will denote the numerical concentration of the free particles, $c(\mathbf{r})$ —their concentration in the point \mathbf{r} . In order to be in the framework of the approximation of the nearest neighbors, which is used for calculations of the stationary distribution function g_n , we will take into account interaction of the free particle only with the particle at the nearest extremity of the chain. It is shown in Ref. [23] that in this approximation the magnetic interaction of the free particle with the chain is quite similar to its interaction with a single particle.

Let us consider the particle, which models the chain and introduce a coordinate system with the origin in the center of the particle. It is convenient to use spherical coordinate system with the polar axis aligned along the applied field \mathbf{H} . This coordinate system is shown in Fig. 1.

Because concentration of the particles in ferrofluid is small, we can neglect interaction of the free particle with any

chains except the considered one. Then the equation of diffusion of the free particles near the particle in the origin of the coordinate system (i.e., near the particle which models the chain) can be written down as

$$\frac{\partial c}{\partial t} = \nabla[D(\mathbf{r}) \nabla c] + \nabla[D(\mathbf{r})c \nabla u_{dd}(\mathbf{r})]. \quad (7)$$

Here u_{dd} is the dimensionless, with respect to kT , energy of the dipole-dipole interaction between the fixed and free particles, $D(r) = D_0\psi(r)$, D_0 is the coefficient of translational diffusion of a single particle, $\psi(r) = 1 - 3d/4r$ is the multiplier which takes into account hydrodynamical interaction between the particles [23,25].

The boundary conditions to this problem are

$$\begin{aligned} c &\rightarrow c_\infty, & r &\rightarrow \infty, \\ c &\rightarrow 0, & r &\rightarrow 0. \end{aligned} \quad (8)$$

The first condition is obvious; the second one means that when the free particle touches the fixed one (i.e., it touches the chain), the free particle disappears and transforms into a particle which belongs to this chain.

Equation (7) does not have exact analytical solution. In order to find an approximate estimate, like in Ref. [23], we will use the following considerations. First, we will take into account that the time, which is necessary to establish stationary concentration profile around the fixed particle, is definitely less than the time needed to change the mean concentration c_∞ . This allows us to consider Eq. (7) in quasistationary approximation, neglecting the time derivative in this equation.

The potential u_{dd} of the dipole-dipole interaction depends not only on the distance r between the fixed and free particles, but also on the angle θ between the radius-vector \mathbf{r} and the field \mathbf{H} . Taking it into consideration, one can search solution of the stationary problem [Eqs. (7) and (8)] in the form of series in Legendre polynomials in $\cos \theta$. However this series contains infinite number of terms and converges slowly. Thus, this series is inconvenient for the practical calculations. In order to get reasonable estimates of the particle flux *toward* the chain, we will average the potential u_{dd} , like in [23], over angle θ in the region corresponding to the cone of attraction between the particles. The averaged magnitude of the potential is

$$\bar{u}_{dd} = \frac{\int_{\cos^2 \theta > 1/3} u_{dd}(r, \theta) d \cos \theta}{\int_{-1}^1 d \cos \theta}. \quad (9)$$

Instead of the exact potential $u_{dd}(\mathbf{r})$, in the Eq. (7) we will use this average magnitude of the potential, which depends only on the distance r . This simplification allowed in [23] to get quite accurate description of experiments on kinetics of aggregation of polarized particles in electric field.

The potential of dipole-dipole interaction between two particles with magnetic moments \mathbf{m}_1 and \mathbf{m}_2 can be written as

$$u_{dd} = -\frac{\mu_0}{4\pi kT} \left[3 \frac{(\mathbf{m}_1 \cdot \mathbf{r})(\mathbf{m}_2 \cdot \mathbf{r})}{r^5} - \frac{(\mathbf{m}_1 \cdot \mathbf{m}_2)}{r^3} \right].$$

Since ferromagnetic particles are small, they are involved into intensive Brownian motion, both translational and rotational. That is why, strictly speaking, their magnetic moments fluctuate. The simultaneous account of the translational and rotational Brownian motion in the Eq. (7) leads to very difficult mathematical problem. Here, for simplicity, we will suppose that the moments of both free and fixed particles are aligned along the field \mathbf{H} . This approximation overestimates effect of the magnetic field on the kinetics of the chain growth. However, for the case of strong enough magnetic fields ($\kappa > 1$) necessary for pronounced magnetorheological effects, deviations of the particle moments from the field are weak. That is why the error of this approximation cannot be significant.

Assuming that magnetic moments of the particles are parallel to the field \mathbf{H} , we get

$$u_{dd} = -\frac{\mu_0 m^2}{4\pi kT} \frac{3 \cos^2 \theta - 1}{r^3}. \quad (10)$$

Combining Eqs. (9) and (10) gives

$$\bar{u}_{dd} = -\frac{\varepsilon}{3\sqrt{3}x^3}. \quad (11)$$

Here $x = r/d$ is the dimensionless distance between the particle centers.

Substituting Eq. (11) into Eq. (7), we come to the following quasistationary equation

$$\frac{\partial}{\partial r} \left[D_0 \psi(r) r^2 \left(\frac{\partial}{\partial r} c + c \frac{\partial}{\partial r} \bar{u}_{dd}(r) \right) \right] = 0. \quad (12)$$

Equation (12) with the boundary condition (8) leads to the following expression for the flux J of the free particles onto the chain,

$$J = A c_\infty, \quad A = \frac{4\pi D_0 d}{W}, \quad W = \int_1^\infty \frac{\exp(\bar{u}_{dd})}{\psi(x)x^2} dx. \quad (13)$$

Evolution of the chain ensemble. Let us pass now from analysis of growth of a single chain to evolution of the chains ensemble. The distribution function g_n changes with time due to “adsorption” of the single particles at the extremities of the chains and desorption of the particles from the chains because of their Brownian motion. As the energy of interaction of the particles at the extremities of the chain is smaller approximately by a factor of 2 than the energy of “internal” particles in the chain, it seems reasonable to consider “evaporation” only of the particles located at the extremities of the chain and to neglect other kinds of the chain rupture.

Since the “adsorption” of a free particle by the n -particle chain transforms the chain into $(n+1)$ -particle chain and the “evaporation” of the terminal particle transforms n -particle chain into $(n-1)$ -particle chain, we come to the following kinetic equation:

$$\frac{\partial g_n}{\partial t} = -A g_1 (g_n - g_{n-1}) + B_1 \left(\frac{B_n}{B_{n+1}} g_{n+1} - \frac{B_{n-1}}{B_n} g_n \right), \quad n > 1. \quad (14)$$

Here A is the adsorption coefficient determined in Eq. (13), B_n are coefficients which determine intensity of desorption of the particles from the chains. They will be determined below.

The first term in the Eq. (14) determines the rate of change of concentration of the n -particle chains due to attachment of single particles to these chains and to $(n-1)$ -particle chains; the second one—due to desorption of the particles from the chains with $(n+1)$ and n particles. The fraction form of the coefficient multiplying g_{n+1} and g_n in the desorption term in Eq. (14) is chosen for the convenience.

Equation for g_1 has the following form:

$$\frac{\partial g_1}{\partial t} = -2A g_1^2 - A g_1 \sum_{n=2}^{n_c-1} g_n + 2 \frac{B_1}{B_2} g_2 + B_1 \sum_{n=3}^{n_c} \frac{B_{n-1}}{B_n} g_n. \quad (15)$$

Here we take into account that, first, when two particles form a doublet, their relative hydrodynamical mobility is approximately twice more than the relative mobility of individual particle and motionless chain. Second, after disappearing of the doublet, two new single particles appear. The equations similar to Eqs. (14) and (15) have been used in Ref. [23] at the analysis of evolution of ensemble of chains of polarized particles.

Equation for the chain with maximal number n_c of the particles has the following form:

$$\frac{\partial g_{n_c}}{\partial t} = A g_{n_c-1} g_1 - B_1 \frac{B_{n_c-1}}{B_{n_c}} g_{n_c}. \quad (16)$$

The system of Eq. (14)–(16) is closed and automatically satisfies to the condition

$$\sum_{n=1}^{n_c} n g_n = \text{const}. \quad (17)$$

It follows from the physical considerations that $\text{const} = C = \varphi / v$.

Now we are in a position to determine the coefficients B_n . To this end we will use the same considerations as in Ref. [23].

Obviously, the stationary solution g_n^0 of the system [Eqs. (14)–(16)] corresponds to the equilibrium state of the system. Thus it must coincide with the quasiequilibrium distribution function determined from the condition of the free energy minimum. One can easily show that the stationary solution of Eqs. (14)–(16) has the following form:

$$g_n^0 = \frac{B_n}{A} Y^n, \quad (18)$$

where Y is an undetermined multiplier.

Equating g_n in Eqs. (6) and (18), we come to the following results:

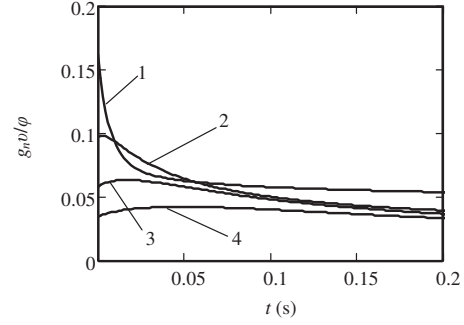


FIG. 2. Concentration g_n vs time t after stepwise increase, at $t=0$, of the applied magnetic field in magnetite ferrofluid. The dimensionless field κ changes from 0 to 3. Parameters of the system: hydrodynamical diameter of the particles is 16 nm, the volume concentration of particles $\varphi=0.015$; $\varepsilon=5.5$, viscosity of the carrier liquid $\eta_0=0.13$ Pa s, the shear rate $\dot{\gamma}=16$ s $^{-1}$. Figures near curves—number n of particles in the chain.

$$Y = X, \quad B_n = \frac{A \sinh(\kappa n)}{v \kappa n} \exp(-\varepsilon). \quad (19)$$

The way of calculation of the parameter X is discussed right after Eq. (6).

Note that similar method of determination of the desorption parameter through the adsorption coefficient and equilibrium distribution function have been used in Ref. [26] at the analysis of kinetics of polymer chains growth.

The system of Eqs. (14)–(16) can be solved numerically. Figures 2 and 3 illustrate the time dependences of the distribution function g_n after stepwise alternation of applied field and the shear rate, respectively. Parameters of the fluid used in our calculations correspond to the ferrofluid TTR of the University of Timisoara (Romania). This fluid has been used in experiments [16] on the viscoelastic phenomena in ferrofluids.

III. MACROSCOPICAL STRESS TENSOR

A model of viscoelastic phenomena in ferrofluids with the chainlike aggregates has been suggested in Refs. [17–19]. Kinetics of the chain growth/disintegration has not been considered in [17–19]—these processes have been assumed to

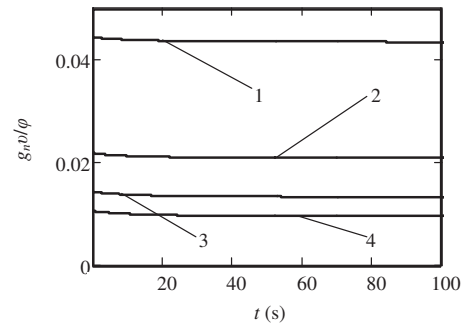


FIG. 3. The same as for Fig. 2 after stepwise decrease of the shear rate; at $t=0$ the shear rate $\dot{\gamma}$ changes from 16 s $^{-1}$ to 1.6 s $^{-1}$; the dimensionless field $\kappa=3$.

be momentary. Only kinetics of the chains reorientation after alternation of the fluid shear rate has been taken into account. Analysis shows that for the ferrofluids TTR, used in the experiments [16], this mechanism gives the time of rheological relaxation of about 10^{-1} – 10^{-2} s, whereas in experiments [16] this time varied in region from 1 to 10 s.

In this part of the work, we suggest a model of nonstationary rheological properties of ferrofluids with the chain-like aggregates. This model is based on the idea that the viscoelastic phenomena in ferrofluids can be explained by kinetics of formation/disintegration of the chains. Since time of the chain reorientation, according to the results [18,19], is much less than the time [16] of the fluid macroscopical rheological relaxation, we will neglect the time of reorientation and will consider this process as instant.

Similar to [7,17–19], we model the n -particle chain by ellipsoid of revolution with the minor and major axes equal to d and nd , respectively. It is of principal importance that the volume of this ellipsoid equals to the total volume $\pi nd^3/6$ of all particles in the chain. Thus the volume concentration of ellipsoids is the same as concentration of the particles in the ferrofluid.

By using results of statistical hydromechanics of suspensions of nonspherical particles [27], one can present the macroscopical viscous stress tensor σ as follows:

$$\sigma_{ik} = \sigma_{ik}^s + \sigma_{ik}^a,$$

$$\begin{aligned} \sigma_{ik}^s &= 2\eta_0\gamma_{ik} + \eta_0 \left\langle \left\langle (2\alpha_n\gamma_{ik} - \rho_n\langle e_j e_s \rangle_n \delta_{ik}\gamma_{js}) + (\zeta_n + \beta_n\lambda_n) \right. \right. \\ &\quad \times (\langle e_i e_j \rangle_n \gamma_{jk} + \langle e_k e_j \rangle_n \gamma_{ji}) + \beta_n(\omega_{ij}\langle e_j e_k \rangle_n + \omega_{kj}\langle e_j e_i \rangle_n) \\ &\quad \left. \left. + (\chi_n - 2\lambda_n\beta_n)\langle e_i e_k e_j e_s \rangle_n \gamma_{js} - \beta_n \frac{d}{dt} \langle e_i e_k \rangle_n \right\rangle \right\rangle, \\ \sigma_{ik}^a &= \frac{\kappa kT}{2\nu} \langle \langle \langle e_i \rangle h_k - \langle e_k \rangle h_i \rangle \rangle, \quad h_i = \frac{H_i}{H}, \\ \gamma_{ik} &= \frac{1}{2} \left(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} \right), \quad \omega_{ik} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_k} - \frac{\partial u_k}{\partial x_i} \right), \\ &\quad i, j, k, s = x, y, z. \end{aligned} \quad (20)$$

Here and below

$$\begin{aligned} \langle \langle \dots \rangle \rangle &= \sum_n \dots n \nu g_n, \quad \langle \dots \rangle = \int \dots \mathbf{e} \phi_n(\mathbf{e}) d\mathbf{e}, \\ \langle \dots \rangle^0 &= \int \dots \mathbf{e} \phi_n^0(\mathbf{e}) d\mathbf{e}, \quad i, \dots, k = x, y, z, \end{aligned}$$

\mathbf{u} is macroscopical velocity of the fluid flow. The unit vector \mathbf{e} is aligned along the chain axis, ϕ_n is normalized to unity function of distribution over orientations of \mathbf{e} , ϕ_n^0 is the equilibrium distribution function. Parameters α_n, \dots, ζ_n are given in the Appendix. The Einstein convention for summations over indexes is used here.

Under the conditions $\varepsilon \gg 1$ and $\varepsilon \ll \kappa$ one can propose that magnetic moments of all particles in a chain are aligned along the chain axis. Thus, the equilibrium orientational distribution function $\phi_n^0(\mathbf{e})$ coincides with that for the rigid magnetic rods with the moment mn .

By using well-known results of the equilibrium theory of ferrofluids (see, for example [1,4]), one can write down

$$\phi_n^0(\mathbf{e}) = \frac{\kappa n}{4\pi \sinh(\kappa n)} \exp[\kappa n(\mathbf{e} \cdot \mathbf{h})]. \quad (21)$$

In order to determine the nonequilibrium orientation function φ_n , one needs to find a solution of corresponding Fokker-Planck equation. By using the model of ellipsoids for the chains and well-known form of this equation for the ellipsoids of revolution (see, for example [27]), we can write the Fokker-Planck equation as

$$\begin{aligned} \frac{\partial \phi_n}{\partial t} + \lambda_n(e_s \gamma_{sl} - e_m e_s e_l \gamma_{ms}) \frac{\partial \phi_n}{\partial e_l} + \omega_{ls} e_s \frac{\partial \phi_n}{\partial e_l} - 3\lambda_n e_l e_s \gamma_{ls} \varphi_n \\ - D_n \kappa n \left[(e_j e_m h_m - h_j) \frac{\partial \phi_n}{\partial e_j} + 2e_j h_j \phi_n \right] \\ = D_n \left(\frac{\partial^2 \phi_n}{\partial e_j^2} - 2e_s \frac{\partial \phi_n}{\partial e_s} - e_j e_s \frac{\partial^2 \phi_n}{\partial e_j \partial e_s} \right), \end{aligned} \quad (22)$$

where

$$D_n = \frac{kT}{\pi \eta_0 d^3 n \delta_n}, \quad \delta_n = \frac{2(n^2 + 1)}{3n(n^2 \alpha_0 + \beta_0)}.$$

The shape parameters α_0, β_0 are given in the Appendix.

Equation (22) is the well known Fokker-Planck equation for ellipsoidal ferromagnetic Brownian particle, written down in a Cartesian coordinate system. Exact solution of this equation for arbitrary κn is unknown. In order to find its approximate solution, we will use the effective-field method developed in [4] for ferrofluids with single particles, and applied to the systems with chains in [7,8,17–19]. In the framework of this approach, which is a variant of the trial function method, we will look for the nonequilibrium distribution function in the form

$$\phi_n = \phi_n^0 [1 + a_i(e_i - \langle e_i \rangle_n^0) + b_{ik}(e_i e_k - \langle e_i e_k \rangle_n^0)]. \quad (23)$$

Here a_i and b_{ik} are Cartesian components of unknown vector and tensor which are to be determined.

To this end, we multiply both parts of Eq. (22) by components of vector \mathbf{e} and tensor $e_i e_m - \delta_{im}/3$, then integrate these expressions over all orientations of \mathbf{e} . As a result, in the linear approximation with respect to the components of gradient of the flow velocity, we get the following equations (see [27]):

$$\begin{aligned} \frac{d\langle e_k \rangle_n}{dt} &= -\frac{1}{\tau_{1n}} \langle e_k \rangle_n + \lambda_n (\langle e_i \rangle_n^0 \gamma_{ik} - \langle e_k e_j e_s \rangle_n^0 \gamma_{js}) + \omega_{kj} \langle e_j \rangle_n^0 \\ &\quad + D_n \kappa n (h_k - \langle e_k e_j \rangle_n h_j) \end{aligned} \quad (24)$$

and

$$\begin{aligned} \frac{d\langle e_i e_k \rangle_n}{dt} = & -\frac{1}{\tau_{2n}} \left(\langle e_i e_k \rangle_n - \frac{1}{3} \delta_{ik} \right) + \lambda_n (\langle e_i e_s \rangle_n^0 \gamma_{sk} + \langle e_k e_s \rangle_n^0 \gamma_{si}) \\ & + \omega_{ij} \langle e_j e_k \rangle_n^0 + \omega_{kj} \langle e_j e_i \rangle_n^0 - 2\lambda_n \langle e_i e_k e_s e_j \rangle_n^0 \gamma_{sj} \\ & + D_n \kappa n (\langle e_k \rangle_n h_i - 2\langle e_j e_i e_k \rangle_n h_j + \langle e_i \rangle_n h_k), \end{aligned} \quad (25)$$

where

$$\tau_{1n} = \frac{1}{2D_n}, \quad \tau_{2n} = \frac{1}{6D_n}.$$

The upper index 0 marks the equilibrium statistical moments determined by using the equilibrium distribution function ϕ_n^0 .

Approximation linear with respect to the components γ_{ik} and ω_{ik} is valid when the following strong inequality $\{\gamma_{ik}, \omega_{ik}\}/D_n \ll 1$ is held for all $n < n_c$. Simple estimates show that for majority of realistic situations the last inequality is fulfilled.

By using the trial function (23) for calculation of the non-equilibrium moments in Eqs. (24) and (25), we come to a system of ordinary differential equations with respect to $a_i(t)$ and $b_{ik}(t)$ for every given n . Having solved this system, we can use the function (23) to determine the moments in the expression (20) for the macroscopical stress tensor. It should be noted that similar approach has led to quite good agreement of the analytical calculations with the results of laboratory experiments [5,7] and computer simulation [28].

IV. RELAXATION OF VISCOSITY AFTER STEPWISE ALTERNATION OF THE SHEAR RATE

Let us introduce a Cartesian coordinate system x, y, z with the axis Oz parallel to the applied magnetic field. In this system $H_z = H = \text{const}$, $H_{x,y} = 0$. In general case the problem of calculation of a_i and b_{ij} is not difficult, however, quite cumbersome. Here we consider a case when the ferrofluid velocity \mathbf{u} is aligned along the axis Ox and its gradient—along the axis Oz (i.e., along the applied field \mathbf{H}). Let us denote by $\dot{\gamma}$ the components of the velocity gradient $\dot{\gamma} = \gamma_{xz} = \gamma_{zx} = \omega_{xz} = -\omega_{zx}$.

Estimates based on the model [18,19] show that the typical time τ_{1n} of reorientation of even long chains, consisting of several tens of particles, for the ferrofluid of type TTR is short, about 10^{-2} – 10^{-1} s. That is significantly less than the time of rheological relaxation of the fluid, detected in experiments [16] (from 1 to 10 s). That is why, for maximal simplification of calculations, we will neglect the time of the chains reorientation, considering this process as momentary. It allows restricting ourselves by the stationary approximation of the problem [Eqs. (24) and (25)].

In the case of flow under consideration, in the linear approximation with respect to $\dot{\gamma}$ Eq. (20) has the following form:

$$\sigma_{xz} = \sigma_{xz}^s + \sigma_{xz}^a,$$

$$\begin{aligned} \sigma_{xz}^s = & 2\eta_0 \dot{\gamma} + \eta_0 \left\langle \left[2\alpha_n + (\zeta_n + \beta_n \lambda_n) (\langle e_x^2 \rangle_n^0 + \langle e_z^2 \rangle_n^0) + \beta_n (\langle e_z^2 \rangle_n^0 \right. \right. \\ & \left. \left. - \langle e_x^2 \rangle_n^0) + 2(\chi_n - 2\lambda_n \beta_n) \langle e_x^2 e_z^2 \rangle_n^0 \right] \dot{\gamma} - \beta_n \frac{d}{dt} \langle e_x e_z \rangle_n \right\rangle, \end{aligned}$$

$$\sigma_{xz}^a = \frac{\kappa k T}{2\nu} \langle \langle e_x \rangle_n \rangle \quad (26)$$

Equations (23)–(25) now can be written respectively as follows:

$$\phi_n = \phi_n^0 [1 + a_x e_x + b_{xz} e_x e_z], \quad (27)$$

$$\begin{aligned} \frac{d\langle e_x \rangle_n}{dt} = & -\frac{1}{\tau_{1n}} \langle e_x \rangle_n + [\lambda_n (\langle e_z^2 \rangle_n^0 - 2\langle e_x^2 e_z^2 \rangle_n^0) + \langle e_z \rangle_n^0] \dot{\gamma} \\ & - D_n \kappa n \langle e_x e_z \rangle, \end{aligned} \quad (28)$$

and

$$\begin{aligned} \frac{d\langle e_x e_z \rangle_n}{dt} = & -\frac{1}{\tau_{2n}} \langle e_x e_z \rangle_n + [\lambda_n (\langle e_x^2 \rangle_n^0 + \langle e_z^2 \rangle_n^0 - 4\langle e_x^2 e_z^2 \rangle_n^0) \\ & + \langle e_z^2 \rangle_n^0 - \langle e_x^2 \rangle_n^0] \dot{\gamma} + D_n \kappa n (\langle e_x \rangle_n - 2\langle e_x e_z^2 \rangle_n) \end{aligned} \quad (29)$$

Direct calculations show that for the considered flow all components of a_i and b_{ij} , except a_x and b_{xz} , equal to zero.

Combining Eqs. (27) and Eqs. (28) and (29), after simple, however, cumbersome calculations, we come to the following system of equations with respect to a_x and b_{xz} :

$$\begin{aligned} & \left[\left(\frac{1}{\tau_{1n}} \right) \langle e_x^2 \rangle_n^0 + D_n \kappa n \langle e_x^2 e_z^2 \rangle_n^0 \right] a_x \\ & + \left[\left(\frac{1}{\tau_{1n}} \right) \langle e_x^2 e_z^2 \rangle_n^0 + D_n \kappa n \langle e_x^2 e_z^2 \rangle_n^0 \right] 2b_{xz} \\ & = [\lambda_n (\langle e_z \rangle_n^0 - 2\langle e_x^2 e_z^2 \rangle_n^0) + \langle e_z \rangle_n^0] \dot{\gamma}, \end{aligned} \quad (30)$$

and

$$\begin{aligned} & \left[\left(\frac{1}{\tau_{2n}} \right) \langle e_x^2 e_z^2 \rangle_n^0 + D_n \kappa n (2\langle e_x^2 e_z^2 \rangle_n^0 - \langle e_x^2 \rangle_n^0) \right] a_x \\ & + \left[\left(\frac{1}{\tau_{2n}} \right) \langle e_x^2 e_z^2 \rangle_n^0 + D_n \kappa n (2\langle e_x^2 e_z^2 \rangle_n^0 - \langle e_x^2 e_z^2 \rangle_n^0) \right] 2b_{xz} \\ & = [\lambda_n (\langle e_x \rangle_n^0 - 4\langle e_x^2 e_z^2 \rangle_n^0 + \langle e_z \rangle_n^0) + \langle e_z^2 \rangle_n^0 - \langle e_x^2 \rangle_n^0] \dot{\gamma}. \end{aligned} \quad (31)$$

Substituting Eq. (27) into Eq. (26), we get

$$\sigma_{xz} = 2\eta \dot{\gamma} \quad (32)$$

Here η is the ferrofluid effective viscosity which is equal to

$$\begin{aligned} \eta = & \eta_0 \left[1 + \left\langle \left\langle \alpha_n + \frac{1}{2} [(\zeta_n + \beta_n \lambda_n) (\langle e_x^2 \rangle_n^0 + \langle e_z^2 \rangle_n^0) + \beta_n (\langle e_z^2 \rangle_n^0 \right. \right. \right. \\ & \left. \left. - \langle e_x^2 \rangle_n^0) + 2(\chi_n - 2\lambda_n \beta_n) \langle e_x^2 e_z^2 \rangle_n^0] \right. \right. \\ & \left. \left. + \frac{1}{2\nu} \frac{\kappa k T}{\eta_0} (A_1 \langle e_x^2 \rangle_n^0 + B_1 \langle e_x^2 e_z^2 \rangle_n^0) \right\rangle \right], \end{aligned} \quad (33)$$

$$A_1 = \frac{a_x}{2\dot{\gamma}}, \quad B_1 = \frac{b_{xz}}{\dot{\gamma}}.$$

Having determined the function g_n from Eqs. (15) and (17), a_x and b_{xz} from the system [Eqs. (30) and (31)], combining

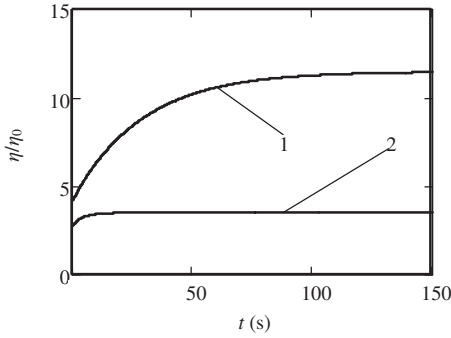


FIG. 4. The time dependence of the effective viscosity η after stepwise *decrease*, at $t=0$, of the shear rate $\dot{\gamma}$ from 16 s^{-1} to 1.6 s^{-1} . Parameters of the system are the same as in Fig. 2. Figures near curves: 1— $\kappa=3$; 2— $\kappa=1$.

them with Eq. (33), we can estimate the time-dependent effective viscosity $\eta(t)$.

Figure 4 illustrates evolution of the viscosity after stepwise *decrease* of the shear rate. Physical parameters of the fluid correspond to those of the fluid TTR which have been used in experiments [16].

Evolution of the viscosity after stepwise *increase* of the shear rate is illustrated in Fig. 5.

The difference between situations, illustrated in Figs. 4 and 5, is only in direction of the shear rate alternation. These results demonstrate that characteristic time of the viscosity evolution after increase of shear rate $\dot{\gamma}$ is shorter than after decrease of $\dot{\gamma}$.

Analysis shows that evolution of the viscosity after increase of the shear rate qualitatively depends on the magnitude of the shear rate step. Calculations of the viscosity behavior for different magnitudes of the step are illustrated in Fig. 6.

Evolution of the viscosity is determined by evolution of the distribution function g_n after the shear rate step. Shortly after this step chains with $n > n_c(\dot{\gamma}_2)$ disintegrate. The intimate details of this disintegration have not been studied yet. However, analysis shows that the hydrodynamical destruction forces have maximum in the middle of the chain. Thus one can expect that under the action of these forces a chain separates in half. This assumption has been used in our calculations.

Analysis shows that the further evolution of g_n is determined by difference between the initial $\dot{\gamma}_1$ and final $\dot{\gamma}_2$ mag-

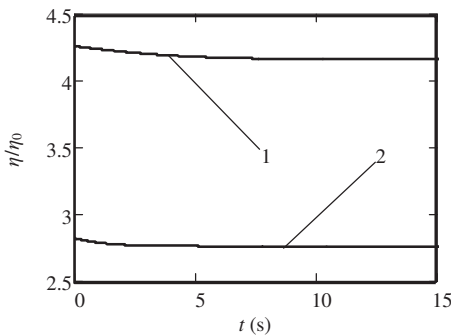


FIG. 5. The same as in Fig. 4 after stepwise *increase* of the shear rate from 1.6 s^{-1} to 16 s^{-1} .

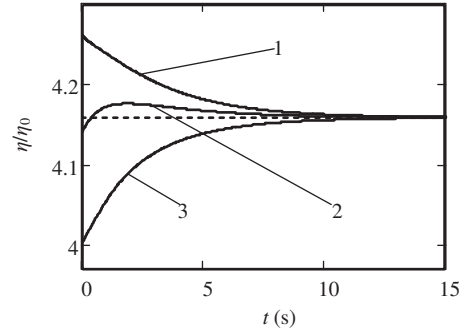


FIG. 6. Dimensionless effective viscosity η/η_0 vs time t . Parameters of ferrofluid are the same as for Figs. 2–5, dimensionless magnetic field $\kappa=3$. At $t=0$ the shear rate *increases* from $\dot{\gamma}_1$ to $\dot{\gamma}_2$. Figures near curves: 1—initial shear rate $\dot{\gamma}_1=1.6 \text{ s}^{-1}$, 2— $\dot{\gamma}_1=2 \text{ s}^{-1}$, 3— $\dot{\gamma}_1=7 \text{ s}^{-1}$. In all three cases the final shear rate $\dot{\gamma}_2=1.6 \text{ s}^{-1}$.

nitudes of the shear rate. In the case of relatively large difference, the function g_n evolves toward the short chains. It leads to decrease of the effective viscosity η with time (curve 1 in Fig. 6). In the case of relatively small difference between $\dot{\gamma}_1$ and $\dot{\gamma}_2$ the chains evolve toward $n=n_c(\dot{\gamma}_2)$, their characteristic length increases, therefore the effective viscosity η increases (curve 3). Curve 2 with the nonmonotonic behavior of η corresponds to some intermediate case and competition between the tendencies to the chain growth and disintegration.

Let us discuss now the characteristic time of the viscosity evolution after a stepwise change of the shear rate. It is natural to define the relaxation time τ as the time t for which the value $|\eta(t) - \eta(\infty)|$ is $e=2.72\dots$ times less than the initial difference $|\eta(0) - \eta(\infty)|$.

Some results of calculations of the time τ after stepwise alternations of the shear rate are shown in Fig. 7. Dots demonstrate results [16] for this time taken from the experiments with oscillating shear rate and measurements of the real and imaginary parts of the complex viscosity. The relaxation time in [16] has been determined as $\tau=1/\omega_{\max}$, where ω_{\max} is frequency of the imaginary viscosity maximum.

The theoretical and experimental results have the same order of magnitude. It should be noted that the relaxation

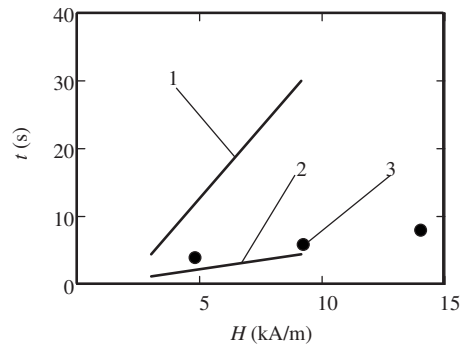


FIG. 7. Relaxation time τ vs applied magnetic field H . 1 and 2—theoretical calculations for the system with the same parameters as for Figs. 2–6; the shear rate changes stepwisely from 16 s^{-1} to 1.6 s^{-1} (line 1) and back (line 2); dots 3—experiments [16] with the oscillating shear flow.

times, calculated by using the previous models [4,18,19], are several orders of magnitude less than the experimental results [16]. What is more, the model [4] predicts decrease of the relaxation time when applied magnetic field H increases. This conclusion is in qualitative contradiction with experiments [16], whereas our model predicts the correct dependence of τ on H .

The fact that our results fit the correct order of magnitude of the relaxation time τ as well as the dependence of this time on the field shows that the model correctly reflects the main physical features of the microscopical cause of the viscoelastic phenomena in ferrofluids.

The quantitative difference between the theory and experiments can take place due to many reasons, for example—polydispersity of the real ferrofluid, flexibility of chains in this system, etc. Unfortunately, account of polydispersity of a ferrofluid as well as fluctuations of the chain shape leads to significant mathematical difficulties, which hardly can be overcome in an analytical model.

V. CONCLUSIONS

We propose a simple statistical model of kinetics of evolution of the chainlike aggregates in ferrofluids and effect of this process on the macroscopical nonstationary viscoelastic (relaxation) properties of ferrofluids. Our analysis shows that the viscoelastic phenomena can be determined by evolution of the heterogeneous aggregates in ferrofluids. In spite of the conscious oversimplification of the model, it leads to reasonable agreement with known experimental results. It should be noted that any modification of the model in order to take into account various factors which take place in the real ferrofluids but have been ignored here (polydispersity of these systems; flexibility and interaction of the chains, etc.), will lead to significant mathematical problems and very cumbersome calculations.

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APPENDIX

(1) The equilibrium statistical moments of orientation of n -particles chain have the following form [27]:

$$\langle e_i \rangle_n^0 = h_i L_1, \quad \langle e_i e_k \rangle_n^0 = \frac{1}{2}(1 - L_2) \delta_{ik} + \frac{1}{2}(3L_2 - 1) h_i h_k,$$

$$\begin{aligned} \langle e_i e_k e_j \rangle_n^0 &= \frac{1}{2}(L_1 - L_3)(\delta_{ik} h_j + \delta_{ij} h_k + \delta_{kj} h_i) \\ &+ \frac{1}{2}(5L_3 - 3L_1) h_i h_j h_k, \end{aligned}$$

$$\begin{aligned} \langle e_i e_k e_l e_m \rangle_n^0 &= \frac{1}{8}(1 - 2L_2 + L_4)(\delta_{ik} \delta_{lm} + \delta_{im} \delta_{kl} + \delta_{il} \delta_{km}) \\ &+ \frac{1}{8}(6L_2 - 5L_4 - 1)(h_i h_k \delta_{lm} + h_i h_m \delta_{kl} + h_i h_l \delta_{km}) \\ &+ h_i h_m \delta_{ik} + h_l h_k \delta_{im} + h_m h_k \delta_{li} \\ &+ \frac{1}{8}(3 - 30L_2 + 35L_4) h_i h_k h_l h_m, \\ \langle e_z \rangle_n^0 &= L_1, \quad \langle e_z^2 \rangle_n^0 = L_2, \quad \langle e_z^3 \rangle_n^0 = L_3, \quad \langle e_z^4 \rangle_n^0 = L_4, \\ \langle e_z^5 \rangle_n^0 &= L_5, \quad \langle e_x^2 \rangle_n^0 = \frac{1}{2}(1 - L_2), \\ \langle e_x^2 e_z \rangle_n^0 &= \frac{1}{2}(L_1 - L_3), \quad \langle e_x^2 e_z^2 \rangle_n^0 = \frac{1}{2}(L_2 - L_4), \\ \langle e_x^2 e_z^3 \rangle_n^0 &= \frac{1}{2}(L_3 - L_5), \end{aligned}$$

Here L_J are the Langevine functions of the order J and

$$L_J = L_J(\kappa n), \quad J = 1, 2, 3, 4, 5,$$

$$L_1(x) = \coth(x) - \frac{1}{x}, \quad L_2(x) = 1 - \frac{2}{x} L_1(x),$$

$$L_3(x) = \frac{1}{x} + L_1(x) - \frac{3}{x} L_2(x), \quad L_4(x) = 1 - \frac{4}{x} L_3(x),$$

$$L_5(x) = \frac{1}{x} + L_1(x) - \frac{5}{x} L_4(x).$$

(2) Shape coefficients $\alpha_n \dots \zeta_n$ are determined in Ref. [27] as

$$\begin{aligned} \alpha_n &= \frac{1}{n\alpha'_0}, \quad \beta_n = \frac{2(n^2 - 1)}{n(n^2\alpha_0 + \beta_0)}, \quad \zeta_n = \frac{4}{n\beta'_0(n^2 + 1)} \\ &- \frac{2}{n\alpha'_0}, \quad \chi_n = \frac{2\alpha''_0}{n\alpha_0\beta'_0} - \frac{8}{n\beta'_0(n^2 + 1)} + \frac{2}{n\alpha'_0}, \end{aligned}$$

$$\rho_n = \frac{1}{3n\alpha'_0\beta''_0} [2(\alpha''_0 - \beta''_0) + 3n(\alpha_0\alpha''_0 - \beta_0\beta''_0)], \quad \lambda_n = \frac{n^2 - 1}{n^2 + 1}.$$

where

$$\alpha_0 = \int_0^\infty \frac{ds}{(n^2 + s)Q}, \quad \beta_0 = \int_0^\infty \frac{ds}{(1 + s)Q},$$

$$\alpha'_0 = \int_0^\infty \frac{ds}{(1 + s)^2 Q}, \quad \beta'_0 = \int_0^\infty \frac{ds}{(1 + s)(n^2 + s)Q},$$

$$\alpha''_0 = \int_0^\infty \frac{s ds}{(1 + s)^2 Q}, \quad \beta''_0 = \int_0^\infty \frac{s ds}{(1 + s)(n^2 + s)Q},$$

$$Q = (1 + s)\sqrt{n^2 + s}$$

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