# Ostwald ripening kinetics in a magnetic fluid made metastable by a strengthening of an external magnetic field

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Ostwald ripening kinetics in a metastable magnetic fluid has been studied theoretically under the condition when the highly elongated ellipsoidal shape of the new phase elements stretched along the external magnetic field direction is taken into account. The self-similar solutions have been obtained for the time evolution of a supersaturation [$\sim t^{-1/3}(\ln t)^{5/18}$], of a critical and a mean droplike aggregate volume [$\sim t^{7/6}(\ln t)^{-2/3}$], of an aggregate concentration [$(\ln t)^{2/3}t^{-7/6}$]. An aggregate distribution density has been found as a function of the aggregate size and of the critical aggregate size. [S1063-651X(98)11311-9]

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## I. INTRODUCTION

Stable colloidal suspensions of the one-domain particles of ferromagnetic and ferrimagnetic materials are known as magnetic fluids (ferrofluids, ferrocolloids). The small sizes of dispersed ferroparticles ($\sim 10$ nm) provide the particles with an inherent magnetic moment of the constant value. The stabilization of suspension is usually obtained by coating the magnetic grains with a surfactant layer, which allows us to neglect the influence of the van der Waals forces. As a result, the ferroparticles interact with each other through the steric repulsion of surfactant coats and the dipole-dipole interaction of particle magnetic moments. The distinctive feature of the latter interaction is that it depends not only on the distance between the two ferroparticle centers, but also on the mutual orientation of the particle magnetic moments. The great number of experiments [1–4] show that the magneto-dipole interaction is responsible for the phase separation of magnetic fluids, accompanied by the existence of droplike aggregates. These aggregates can be considered as fluids [2,4] with an interfacial tension surface, representing, essentially, a highly concentrated ferrocolloidal phase suspended in a dilute matrix in the form of droplets [1–4]. Typical dimensions of droplike aggregates are of the order of, approximately, 1–5 $\mu$m, i.e., the number of ferroparticles comprising the aggregate is approximately $10^5-10^7$.

From the viewpoint of statistical mechanics the existence of droplike aggregates may be considered as a result of the violation of thermodynamic stability in a system of dispersed ferroparticles that leads to their condensation. Therefore, phase separation in magnetic fluids is treated as a first order phase transition of the ''ferrocolloidal gas–ferrocolloidal liquid'' type. In principle, existing statistical thermodynamic models of magnetic fluids [5–10] have demonstrated that, in a system of particles interacting through a noncentral dipole-dipole potential, the condensation may occur in the absence of a magnetic field at a temperature below a certain critical one, the latter is dependent on the value of the ferroparticle magnetic moments. This is associated with the fact that the noncentral dipole-dipole interaction of ferroparticle magnetic moments displays itself on the whole as an effective interparticle attraction. In a uniform external field this effective attraction strengthens [7,8,10], thus a magnetic field stimulates the process of phase separation in magnetic fluids. In this case, at isothermal and isobaric conditions, a magnetic field increase is equivalent to an effective lowering of temperature. Such a phenomenon looks like a nontrivial phase transition of the condensation type, induced by an external magnetic field. During such a phase transition the occurring droplike aggregates are not spherical, the demagnetization effects lead to an approximately ellipsoidal shape of the droplets stretched along the external field direction [3,11,12]. Common methods of equilibrium thermodynamics suffice to understand under what conditions the phase separation is bound to start [5–8,10]. However, they are certainly inapplicable to elucidate what happens next and so must be replaced by suitable kinetic methods.

The break of the thermodynamic stability of a magnetic fluid is followed by the origination of critical nuclei of a new phase, by their transformation into macroscopic droplike aggregates, and by the ensuing growth of those aggregates in a metastable environment. In theoretical research it is reasonable to distinguish three basic stages of the evolution of a particulate system that follows the preliminary stage of the development of a metastable state. The first one corresponds to the initiation of critical new phase nuclei, which further form either macroscopic new phase elements in a metastable molecular system or droplike aggregates in a colloid. A fact of great consequence for a theoretical treatment is that, during this initial stage, the state of the colloid is practically not affected by the emerging nuclei so that each of them can correctly be regarded as evolving under the constant metastability condition. The second, intermediate stage covers a combined process of the growth of existing aggregates and of the initiation of additional nuclei in the circumstance of permanently reducing metastability of the parent colloid. An analysis of this stage is greatly complicated by the presence of negative feedback between the process of aggregate formation and growth dependent on a transient degree of metastability (e.g., a value of the supersaturation) and that of the gradual reducing of metastability by the growing aggregates. At last, the third stage of final coalescence corresponds to the Ostwald ripening process when the mentioned dependence is of primary importance but the origination of new nuclei almost ceases and may be safely overlooked.

 

As far as the first and second stages are concerned, a relevant theory has been carried out in Ref. [13] under the conditions when the highly elongated ellipsoidal shape of the ferrofluid droplike aggregates is taken into account. The main conclusion of Ref. [13] is that the rate of nucleation and the kinetics of the intermediate stage of phase transition go on more rapidly in a magnetic fluid made metastable by an external field. This is due to the fact that a small strengthening of a magnetic field implies a significant increase of the ferrofluid initial supersaturation. The latter exerts primary control over the kinetics of the phase separation process.

In the present research we should focus our attention on the problem of the theoretical description of the Ostwald ripening kinetics in a magnetic fluid during the phase separation induced by a uniform external magnetic field. A mathematical model is formulated in Sec. II and includes the kinetic equation for the aggregate distribution density, the mass balance equation, and the aggregate growth rate; the latter is dependent both on the aggregate volume and on the volume of the critical aggregate. An analysis, presented in Sec. II, shows that the evolution of a system of droplike aggregates must be accompanied by the self-similar time behavior of the critical aggregate volume. On the basis of this result, the solution for the aggregate distribution density is obtained in Sec. IV and the self-similar time dependences of the aggregate concentration, the supersaturation, and the mean aggregate volume are discussed.

We shall consider a sterically stabilized magnetic fluid containing identical spherical ferroparticles, suspended in a neutral liquid carrier. The ferrocolloid is supposed to be thermodynamically stable in the absence of a magnetic field. But if a weak uniform external magnetic field $H$ is present, a macroscopically homogeneous state of the magnetic fluid becomes unstable and, as a consequence, the magnetic fluid is bound to be separated into two homogeneous phases characterized by the equilibrium values $\varphi_{\mathrm{I}}$ and $\varphi_{\mathrm{II}}$ of the concentration ($\varphi_{\mathrm{I}}<\varphi_{\mathrm{II}}$). At the thermodynamic equilibrium state the coexisting phases are separated by a plane interfacial surface, which is parallel to an external field. In what follows, we are going to study the kinetics of phase separation under the conditions when the supersaturation vanishes and the growth of the critical aggregate is of crucial importance.

We consider the phase separation kinetics of an initially weakly concentrated ferrofluid ($\varphi_0 \ll 1$). In this case, the number of occurring droplike aggregates will be small, and the average distance between the aggregates will be rather large. Hence, we may neglect the effects of the droplet collapse in view of the fact of very low hydrodynamic mobility of the large droplike aggregates.

## II. BASIC EQUATIONS

Let us study the evolution of a system of droplike aggregates suspended in a macroscopically homogeneous metastable magnetic fluid at the last stage of phase transition under the conditions when both the reduction in metastability (the decrease in the parent ferrocolloid supersaturation) and the corresponding growth of a critical aggregate volume are taken into account. The degree of metastability is supposed to be rather small so that it is possible to consider the critical aggregate nuclei as the macroscopic objects. The evolving aggregates are distributed over volume $V$ and it is convenient to describe this distribution with the help of the variable $R=V^{1/3}$. The distribution density $f(t,R)$ is governed by a kinetic equation:

$$\frac{\partial f}{\partial t}+\frac{\partial}{\partial R}\left(\frac{dR}{dt}f\right)=0, \quad t>0, \quad R>0, \tag{1}$$

under complete neglect of fluctuations of the diffusionally controlled growth rate of a single aggregate. We presume the function $f(t,R)$ to be normalized to the number concentration $N(t)$ of the aggregates.

A requirement of conservation of the overall number of ferroparticles in the system under study leads to the mass balance equation:

$$\Delta(t)=\Delta_0-(\varphi_{\mathrm{II}}-\varphi_{\mathrm{I}})\int_0^\infty R^3 f(t,R)\,dR, \quad \Delta(0)=\Delta_0, \tag{2}$$

where $\varphi_{\mathrm{I}}$ and $\varphi_{\mathrm{II}}$ stand for the equilibrium ferroparticle concentrations of the coexisting ferrocolloidal phases I and II with a plain interface, $\varphi_{\mathrm{I}} \ll \varphi_{\mathrm{II}}$; $\Delta(t)=\varphi-\varphi_{\mathrm{I}}$ has the meaning of the absolute ferrofluid supersaturation, and $\Delta_0$ is an inital value of the supersaturation.

In order to close the set of equations (1) and (2) it is necessary to define the growth rate $dR/dt$ as a function of parameters specific to an assemblage of evolving aggregates. The determination of the quantity $dR/dt$ as a function of aggregate volume $V=R^3$ has been the objective of Ref. [13]. As shown in Ref. [13], this function is greatly influenced by an interrelation between the volume and the shape of the aggregates. In the presence of a weak magnetic field a shape of small droplike aggregates is close to a sphere. But in the case of a rather large volume, the aggregates become highly elongated and stretched along the magnetic field direction. Accordingly to the phase transitions in molecular isotropic systems, the volume of a critical ferrofluid nucleus increases while the supersaturation of metastable phase decreases. Thus, all the aggregates (critical and supercritical) may be regarded as highly elongated. Some experimental studies [3,11,12] have demonstrated that the shape of the ferrofluid droplets under the presence of a magnetic field is successfully approximated by the simple ellipsoid of revolution. By using the results of Refs. [3,11,12] we consider the droplike aggregate as an ellipsoid stretched along the external field direction.

During the last stage of the phase separation process, the volumes of the aggregates are rather large. Hence, the strong inequality between the ellipsoid semiaxes $a$ and $b$ holds true: $b \ll a$. A quasiequilibrium value of the semiaxis ratio $c=b/a$ is dependent on the relation between the capillary forces and the effects of demagnetization field. The results of [3,11–13] lead to the following estimation for the highly elongated aggregate, $c \ll 1$:

$$V=R^3 \approx \frac{B}{c^7|\ln c|^3}, \quad B \sim \frac{\sigma^3}{H^6}. \tag{3}$$

Here $\sigma$ is the interfacial tension coefficient on the aggregate surface and $H$ stands for the external field strength. The

total expression for the positive parameter $B$ is presented in Ref. [13] [Eq. (3)] as a function of the interfacial tension coefficient, of the magnetic field strength, and of the magnetic permeabilities of ferrocolloid phases I and II. The value of the interfacial tension coefficient $\sigma$ may be calculated on the basis of well-known methods (see, for example, Ref. [14]). Some expressions are presented in Refs. [15,16]. The point is that the interfacial tension is dependent only on the concentrations of dispersed ferroparticles in coexisting phases and on the interparticle interaction energy. The theories [15,16] give the value $\sigma \sim (5-10)\times 10^{-7}$ N/m, which correlates well with the experimental results: $\sigma \sim (3-7)\times 10^{-7}$ N/m [3,11,12].

A problem of the aggregate growth rate has been solved in Ref. [13] on the basis of diffusion-limited conception. The ferroparticle concentration profiles outside the aggregate have been determined with the help of ellipsoidal coordinates. As a result, the expression for the single aggregate growth rate has been obtained in the form

$$\varphi_{\mathrm{II}}\frac{dV}{dt}=3\left(\frac{4\pi}{3}\right)^{2/3}D\frac{V^{1/3}-V_*^{1/3}}{c^{2/3}|\ln c|}\Delta(t),$$

$$V_*^{1/3}=\frac{2}{3}\frac{v}{kT}\frac{\varphi_{\mathrm{I}}}{\varphi_{\mathrm{II}}}\frac{\sigma\kappa(c_*)}{\Delta}, \tag{4}$$

$$\kappa(c_*)=\frac{S}{V^{2/3}}\approx\left(\frac{9\pi^4}{16c_*}\right)^{1/3},\quad c_*,c\ll 1,$$

which is valid for the highly elongated droplets. Here $v$ and $D$ are the ferroparticle volume and diffusion coefficient, respectively, $kT$ is a thermal energy, $S$ stands for an ellipsoid surface. Parameters $V_*$ and $c_*$ are the critical aggregate volume and the corresponding value of the semiaxis ratio: in the case $V>V_*$ the aggregate grows, otherwise for $V<V_*$ it dissolves.

The evolving ellipsoidal aggregates are described by the two following parameters: aggregate volume $V$ and the semiaxis ratio $c$. In order to simplify the analysis, we will use the quantities $R=V^{1/3}$ and $R_*=V_*^{1/3}$ and call them ''the aggregate size'' and ''the critical aggregate size.'' The corresponding semiaxis ratios $c$ and $c_*$ are the functions of these sizes within the expression (3).

Let us introduce the dimensionless characteristic sizes of the aggregate:

$$r=\left(\frac{V}{B}\right)^{1/3}=\frac{R}{B^{1/3}},\quad r_*=\left(\frac{V_*}{B}\right)^{1/3}. \tag{5}$$

In order to get the explicit form of the aggregate growth rate (4) as a function of $r$ and $r_*$ it is convenient to use the asymptotic expansion

$$z-\ln z=y,\quad y\to\infty,\quad z(y)\approx y\left(1+\frac{\ln y}{y}+\cdots\right). \tag{6}$$

After evident calculations we obtain the following from Eq. (3) with the terms of logarithmic accuracy:

$$c\approx\left(\frac{3}{7r\ln r}\right)^{3/7},\quad c^{2/3}|\ln c|\approx\left(\frac{3}{7}\right)^{5/7}\left[\frac{(\ln r)^{5/7}}{r^{2/7}}\right],\quad \ln r\gg 1. \tag{7}$$

By using the expressions (3)–(7), we get finally

$$\frac{dr}{dt}=\frac{b}{B}\frac{r-r_*}{r_*}\frac{r_*^{1/7}(\ln r_*)^{1/7}}{r^{12/7}(\ln r)^{5/7}}, \tag{8}$$

$$b=\left(\frac{4\pi}{3}\right)^{2/3}\frac{D}{\varphi_{\mathrm{II}}}\frac{2}{3}\frac{v\sigma}{kT}\frac{\varphi_{\mathrm{I}}}{\varphi_{\mathrm{II}}}\left(\frac{9\pi^4}{16}\right)^{1/3}\left(\frac{7}{3}\right)^{4/7}.$$

The set of equations (1), (2), and (8) describes the evolution of a system of droplike aggregates in a metastable magnetic fluid during the Ostwald ripening stage of the phase transition process. Further, we are going to obtain the self-similar solution for the distribution function according to the classical method originally developed by Lifshitz and Slyozov [17]. It should be noted that there exist some other methods of attacking the problem (see, for example, Refs. [18–22]). Nevertheless, the Lifshitz-Slyozov approach seems to be most applicable for our purpose.

## III. CRITICAL AGGREGATE SIZE

Let us introduce new self-similar variables:

$$u=\frac{r(t)}{r_*(t)},\quad x(t)=\frac{r_*(t)}{r_{*0}},\quad r_{*0}=\left(\frac{V_{*0}}{B}\right)^{1/3}, \tag{9}$$

where $u$ has the meaning of the aggregate size measured in units of the critical aggregate size, $x$ stands for a dimensionless critical size, and $V_{*0}$ is an initial value of the critical aggregate volume $V_*$. With the help of the dimensionless time variable $t'=tbB^{-1}r_{*0}^{-18/7}$ we get instead of Eq. (8),

$$\frac{d(ux)}{dt'}=\frac{(u-1)x^{1/7}[\ln(xr_{*0})]^{1/7}}{(ux)^{12/7}[\ln(uxr_{*0})]^{5/7}}. \tag{10}$$

After evident calculations we may write Eq. (10) in the form

$$\frac{du^{19/7}}{d\tau}=\gamma'\frac{(u-1)(\ln x+\ln r_{*0})^{1/7}}{(\ln x+\ln u+\ln r_{*0})^{5/7}}-u^{19/7}, \tag{11}$$

$$\gamma'=\frac{1}{x^{11/7}(dx/dt')},\quad \tau=\frac{19}{7}\ln x.$$

In view of the fact that the dimensionless critical size tends toward infinity while the supersaturation approaches zero, we may neglect the terms $\ln r_{*0}$ and $\ln u$ in comparison with $\ln x$. Hence,

$$\frac{du^{19/7}}{d\tau}=\gamma(u-1)-u^{19/7},$$

$$\gamma=\frac{\gamma'}{(\ln x)^{4/7}}=\frac{1}{x^{11/7}(\ln x)^{4/7}(dx/dt')}. \tag{12}$$

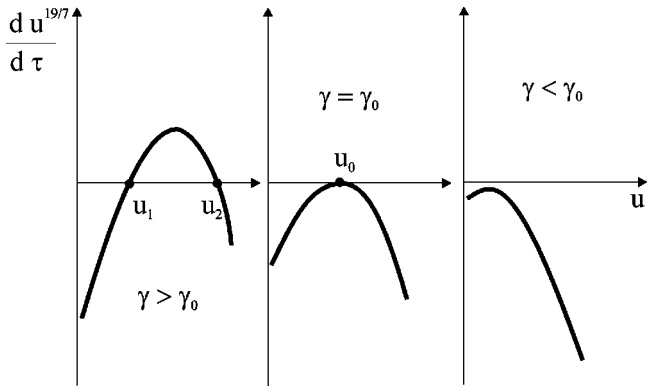


FIG. 1. Possible dependencies of the growth rate (12) on the self-similar aggregate size.

The right side of Eq. (12) may depend on $u$ by one of the three kinds illustrated in Fig. 1. This function is characterized by one maximum at the point $u=u_0$ and touches the $u$ axis only for the value $\gamma=\gamma_0$. Every point on the $u$ axis represents the state of the aggregate with respect to its growth. In the case of $\gamma>\gamma_0$ all the points to the left of $u_1$ move towards the left and disappear at the coordinate center. All the points $u_1<u<u_2$ move towards the right and all the points to the right of $u_2$ move towards the left up to the steady state at $u_2$. This means that all the aggregates will acquire the size $r=u_2 r_*(t)$ and will tend to infinity with the growth of the critical size. Thus, the mass balance equation (20) will not be satisfied. In the case $\gamma<\gamma_0$ all the points on the $u$ axis move towards the left and disappear, and the mass balance equation will be incorrect again. Consequently, according to the method of Ref. [14] we are able to state that the function $\gamma(\tau)$ must approach the limit $\gamma_0$ from below:

$$\gamma(\tau)=\gamma_0[1-\varepsilon(\tau)^2], \quad \varepsilon(\tau\to\infty)\to 0. \tag{13}$$

The value $\gamma_0$ is determined from the set of algebraic equations

$$\gamma_0(u_0-1)-u_0^{19/7}=0, \tag{14}$$

$$7\gamma_0-19u_0^{12/7}=0.$$

Thus, for the coordinates of the lockup point we get

$$u_0=\frac{19}{12}, \quad \gamma_0=\frac{19}{7}\left(\frac{19}{12}\right)^{12/7}. \tag{15}$$

Substituting the expression (13) to the growth rate (12), we come to the equation for the function $z(\tau)=[u(\tau)-u_0]/\varepsilon(\tau)$:

$$\frac{1}{\varepsilon}\frac{dz}{d\tau}=-\frac{72}{133}z^2+\eta z-\frac{7}{12}, \quad \eta=\frac{d}{d\tau}\left(\frac{1}{\varepsilon}\right). \tag{16}$$

Following the previous line of attack, we get for the lockup point of Eq. (16) and for the function $\varepsilon(\tau)$:

$$\eta_0=\frac{12}{\sqrt{114}}, \quad z_0=\frac{7}{12}\sqrt{\frac{19}{6}}, \quad \varepsilon(\tau)=\frac{\sqrt{114}}{12}\frac{1}{\tau}. \tag{17}$$

Therefore, the function $\gamma(\tau)$ tends to the limit $\gamma_0$ according to power $\tau^{-2}$. For the case $\tau\to\infty$ we may neglect the difference of $\gamma(\tau)$ from the lockup value $\gamma_0$. By using the definition (12) we get the differential equation:

$$x^{11/7}(\ln x)^{4/7}\frac{dx}{dt'}=\frac{1}{\gamma_0}, \tag{18}$$

which may be easily integrated

$$\frac{t'}{\gamma_0}=\int_1^x s^{11/7}(\ln s)^{4/7}ds$$

$$=\int_0^{\ln x}\exp\left(\frac{18}{7}\zeta\right)\zeta^{4/7}d\zeta$$

$$=\frac{7}{18}\zeta^{4/7}\exp\left(\frac{18}{7}\zeta\right)\Bigg|_0^{\ln x}-\frac{2}{9}\int_0^{\ln x}\zeta^{2/7}\exp\left(\frac{18}{7}\zeta\right)d\zeta$$

$$\approx(7/18)x^{18/7}(\ln x)^{4/7}, \quad x\to\infty, \quad \ln x\gg 1. \tag{19}$$

With the help of asymptotical expansion (6) we obtain the self-similar evolution law of the critical aggregate size:

$$x(t')=r_*(t')/r_{*0}\sim\frac{(t')^{7/18}}{(\ln t')^{2/9}}, \quad t'\to\infty. \tag{20}$$

The expression (20) represents the asymptotic time behavior of the critical ellipsoidal aggregate growth. We would like to recall that according to the Lifshitz-Slyozov theory [17] the similar law for spherical aggregates has the following scaling form: $x\sim t^{1/3}$. The comparison shows that at large times the highly elongated ellipsoidal critical aggregate grows faster than the spherical one.

Concerning the time evolution of the supersaturation, we have to take into account the expressions (4) and (7):

$$\Delta\sim\frac{c_*^{-1/3}}{x}\sim x^{-6/7}(\ln x)^{-1/7}\sim(t')^{-1/3}(\ln t')^{5/18}. \tag{21}$$

The asymptotic behavior of the supersaturation appears to be very close to the classical result $\Delta\sim t^{-1/3}$. However, the supersaturation in a magnetic fluid made metastable by a strengthening of an external field decreases slower than the same one for the system with the spherical nuclei of a new phase. The point is that the rate of decreasing of the supersaturation is larger in the case under study as compared with the system of spherical drops.

## IV. AGGREGATE DISTRIBUTION FUNCTION

Let us now determine the aggregate distribution function by size. It is convenient to turn from the function $f(t,R)$ to a new distribution density by the dimensionless aggregate sizes $\varphi(\tau,u)=f(t,R)R_*$: $\varphi(\tau,u)du=f(t,R)dR$. The latter is governed by a kinetic equation:

$$\frac{\partial\varphi}{\partial\tau}+\frac{\partial}{\partial u}\left(\frac{du}{d\tau}\varphi\right)=0. \tag{22}$$

According to the results of the previous section, the aggregate growth rate may be written as follows:

$$\frac{du}{d\tau}=\frac{7}{19u^{12/7}}[\gamma_0(u-1)-u^{19/7}]. \tag{23}$$

The solution of the kinetic equation (22) may be obtained with the help of the method of characteristics:

$$\varphi(\tau,u)=-\frac{\chi[\tau-\tau(u)]}{(du/d\tau)},$$
$$\tau(u)=\frac{19}{7}\int_0^u\frac{u^{12/7}du}{\gamma_0(u-1)-u^{19/7}}. \tag{24}$$

where $\chi(z)$ represents the unknown function of the characteristics. The only way to determine this function is to use the mass balance equation (2). By taking into account the condition $\Delta(t'\to\infty)\to 0$, this equation becomes

$$\frac{\varphi_{\mathrm{II}}}{\Delta_0}\int_0^\infty R^3 f(t,R)dR=\beta x^3\int_0^{u_0}u^3\varphi du=1,$$
$$\beta=\frac{\varphi_{\mathrm{II}}}{\Delta_0}R_{*0}^3, \tag{25}$$

where the upper integration limit turns to $u_0$ according to the asymptotic behavior of the aggregate growth rate (Fig. 1): in the realized case $\gamma=\gamma_0$ all the aggregates with $u>u_0$ must dissolve. Thus, at $t'\to\infty$ the distribution density $\varphi$ approaches zero fast for all $u>u_0$.

With substitution of expression (24) the mass balance equation (25) takes the form

$$\beta\exp\left(\frac{21}{19}\tau\right)\int_0^{u_0}u^3\varphi(\tau,u)du=1. \tag{26}$$

The left side of this equation becomes independent on $\tau$ only in the case when $\chi$ is the exponential function on $\tau$:

$$\chi[\tau-\tau(u)]=C\exp\left\{\frac{21}{19}[\tau(u)-\tau]\right\},\quad C=\text{const}. \tag{27}$$

Hence, the aggregate distribution density in self-similar variables is determined as follows:

$$\varphi(\tau,u)=C\exp\left[-\frac{21}{19}\tau\right]P(u), \tag{28}$$
$$P(u)=-\frac{21}{19}\exp\left[\frac{21}{19}\tau(u)\right]\left(\frac{du}{d\tau}\right)^{-1}H(u_0-u),$$

where $H(z)$ is the Heaviside step function and the factor $C$ has to be calculated from the equation (26):

$$C^{-1}=\beta\int_0^{u_0}u^3P(u)du. \tag{29}$$

The dependence of the distribution function $P(u)$ on $u$ is shown on the Fig. 2. This function appears to be normalized to unity:

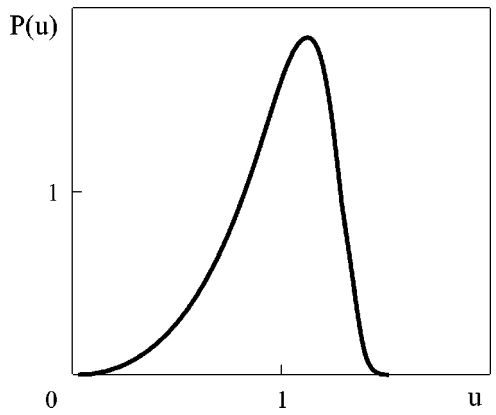


FIG. 2. Self-similar aggregate distribution density (28) as a function of the dimensionless aggregate size.

$$\int_0^{u_0}P(u)du=-\frac{21}{19}\int_0^{u_0}\exp\left[\frac{21}{19}\tau(u)\right]\left(\frac{du}{d\tau}\right)^{-1}du$$
$$=-\frac{21}{19}\int_0^{\infty}\exp\left[\frac{21}{19}\tau(u)\right]d\tau(u)=1. \tag{30}$$

The expressions (28) and (29) totally describe the aggregate distribution over self-similar size variable $u$ and the integral characteristics of the system of aggregates during the Ostwald ripening stage of the ferrocolloid phase separation process.

For example, the numerical concentration of the droplike aggregates asymptotically varies in time according to

$$N(t)=\int_0^\infty f(t,R)dR=\int_0^{u_0}\varphi(\tau,u)du$$
$$=C\exp\left[-\frac{21}{19}\tau\right]\sim\frac{(\ln t')^{2/3}}{(t')^{7/6}}. \tag{31}$$

The classical theory [17] predicts for the spherical new phase elements the asymptotics $N\sim t^{-1}$. The comparison of this and expression (31) shows that the number of ellipsoidal droplets decreases with time faster than for the spherical ones.

The mean volume of the aggregate is

$$\langle V(t)\rangle=\frac{R_*^3}{N(t)}\int_0^{u_0}u^3\varphi(\tau,u)du$$
$$=R_*^3\int_0^{u_0}u^3P(u)du\sim\frac{(t')^{7/6}}{(\ln t')^{2/3}} \tag{32}$$

and grows with time faster than for the spherical case with $\langle V\rangle\sim t$.

## V. DISCUSSION

To sum up we are able to describe by an analytical self-similar method the evolution of the system of droplike ellipsoidal aggregates in a magnetic fluid under the presence of an external magnetic field during the Ostwald ripening stage of the phase separation process. Essentially the same approach could be applied to numerous processes of a new

phase formation in molecular and colloid systems. As compared with the latter, the magnetic field induced phase separation in ferrofluids is essentially controlled by the mutual relation between the volume of an aggregate and its shape. An elongation of the aggregate during its growth is accompanied by an increase of both the interfacial surface and the concentration gradient in the vicinity of the side surface. The latter is caused by the relative decrease (as compared with that for a sphere) of the transverse size of the ellipsoid. Consequently, the aggregate elongation results in a higher value of growth rate in comparison with that of spherical droplets.

Besides that, an ellipsoidal shape of the aggregates leads to the nonclassical relation for the critical aggregate volume that is dependent not only on the ferrocolloid supersaturation, but on the critical aggregate semiaxis ratio as well. Taken together these special features result in the self-similar time evolution laws that differ from the classical theories. The general conclusion is that the system of highly elongated ellipsoidal aggregates evolves faster in comparison with the system of spherical drops.

It should be noted that the problem of the Ostwald ripening kinetics in magnetic fluids has been examined earlier in Ref. [23], where diffusion-limited conditions for the droplike aggregate growth rate have been considered. While determining the ferroparticle concentration profiles, the author of Ref. [23] has used the quasicylindrical solution of the stationary diffusion equation in the vicinity of a drop and the self-similar solution of this equation in spherical geometry far from a drop. The asymptotic joining of the two nonconsistent expressions has brought the author to a physically meaningless result: the drop growth rate obviously depends on time [$\sim(\ln t)^{-1}$] and decreases up to zero even at a constant metastability. During the analysis of the Ostwald ripening kinetics, the author of Ref. [23] has neglected this logarithmic dependence and has obtained the self-similar solutions as the power functions. The point is that the exponents for the time evolution of the drop concentration ($\sim t^{-7/6}$) and of the mean drop volume ($\sim t^{-7/6}$) coincide with the expressions (31) and (32) with logarithmic accuracy.

Unlike the method of Ref. [23], our results are based on the expression for the aggregate growth rate (4) obtained in Ref. [13]. In this paper we have considered the diffusion-limited growth rate of a highly elongated ellipsoidal ferrofluid drop under the presence of a weak magnetic field. In order to determine the ferroparticle concentration profiles we have used the ellipsoidal coordinates while solving the diffusion equation. As a result, expression (4) does not hold any discrepancies of physical nature. On the one hand, good agreement of the results of the present research and of Ref. [23] seems to be questionable, on the other hand, this agreement may be regarded as a verification of the theoretical predictions.

The evolution of the system of droplike aggregates at the final stage of phase transition also may be influenced by the process of drop coagulation. This phenomenon will be important under the high drop concentration and under the rather large magnetic interaction energy. This situation is realized, for example, in magnetorheological and electrorheological suspensions. A theoretical analysis of the evolution of the system of elongated droplike aggregates in the presence of an external magnetic field due to the drop coagulation mechanism represents the point of the separate study.

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