ISSN 1063-7761, Journal of Experimental and Theoretical Physics, 2013, Vol. 116, No. 2, pp. 286–292. © Pleiades Publishing, Inc., 2013.
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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Structurization of Ferrofluids in the Absence of an External Magnetic Field

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Received July 10, 2012

Abstract—Structural transformations in a model ferrofluid in the absence of an external magnetic field have been theoretically studied. The results agree with well-known laboratory experiments and computer simulations in showing that, if the concentration of particles and their magnetic interaction energy are below certain critical values, most particles form separate linear chains. If these parameters exceed the critical values, most particles concentrate so as to form branched network structures. The passage from chains to network has a continuous character rather than represents a discontinuous first-order phase transition.

DOI: 10.1134/S1063776113020192

1. INTRODUCTION

Ferrofluids (magnetic fluids) represent colloidal suspensions of monodomain ferro- or ferrimagnetic particles in a carrying medium. The rich set of unique physical properties of these systems determines their wide use in advanced technologies and draws considerable attention in both basic and practical research. Modern ferrofluids consist of particles with characteristic dimensions about 10–20 nm. In order to prevent the coagulation of particles under the action of central dispersive forces, particles are covered with special surface layers consisting of surfactant molecules or having an ionic nature. These layers produce complete or partial screening of the dispersive forces.

Numerous experiments have shown that particles in ferrofluids are capable of forming various heterogeneous structures such as linear chains, closed rings, Y-like forks, and dense droplets [1–6]. These structures have a typical size of several microns, and they can be readily observed with common optical microscopes. Since the average particle size in ferrofluids is much smaller than the wavelength of visible light, separate particles, chains, and other clusters with linear structures cannot be revealed by optical techniques. However, these structures have been recently observed in electron microscopes [6, 7]. At the same time, linear chains, closed rings, Y-forks, and certain other branched structures have been obtained in numerous computer simulations (see, e.g., [8–12]).

The first models of droplet aggregates [13–16] treated their formation as a gas–liquid phase transition in a system of separate dipole particles, while the presence of chains and other clusters was ignored. However, at present, the appearance of linear chains at the pretransition stage in a macroscopically homogeneous ferrofluid consisting of well-screened particles is a reliably established fact. Recently, we proposed a model of the phase condensation of particles in a ferrofluid with allowance for the appearance of chains [17] and considered two limiting cases in which an external magnetic field is either absent or infinitely strong. Theoretical analysis [17] has confirmed that, as the energy of magnetic interaction between particles increases (or the temperature decreases), linear chains initially form and then exhibit condensation into dense volume phases. It should also be noted that, in the case of strong magnetic fields, dense phases of ferromagnetic particles representing ensembles of chains have been observed in both laboratory experiments [6] and computer simulations [10, 11].

However, in the absence of an external magnetic field, both experiments [6, 18, 19] and computer simulations [19–22] have shown that, when linear chains appear, an increase in the energy of magnetic interaction between particles and their concentration leads to the formation of branched structures and networks. The self-organization of dipole interacting particles into dense homogeneous globules without the preliminary formation of chains has also been observed in computer simulations [23], but this result was obtained for the so-called Stockmayer fluid, which consists of particles interacting like magnetic dipoles with a central Lennard-Jones component. The simulation [23] was performed for the case where the central attraction energy was about half of the energy of magnetic interaction between particles.

Thus, the results of recent experiments and simulations indicate that, in the absence of an external magnetic field, the condensation of ferromagnetic particles well screened from the central interaction follows a specific scenario with the initial formation of linear chains and their subsequent self-organization into

branched aggregates and network structures. Note that the ability of purely magnetic interactions between particles to induce their phase condensation of the gas–homogeneous fluid type in the absence of an external field is still under discussion and opposite viewpoints have been formulated (see, e.g. [20–22]).

It must be recognized that the basic features of the condensation of particles in ferrofluids, despite the long history of research in this field, have not been established so far. Investigations in this direction are important not only from the standpoint of the general theory of phase transitions, but also in view of the numerous practical applications of magnetic fluids and related compositions. For example, in recent years, a number of magnetic polymer compositions (ferrogels and ferroelastomers) consisting of magnetic particles embedded in polymeric matrices have been synthesized for various applications. The dispersion of particles takes place while the matrix occurs in a liquid state prior to polymerization. In this state, the composition represents a usual ferrofluid. The structures formed by magnetic particles in the matrix are fixed upon polymerization. The macroscopic properties and behavior of a polymerized composition depend on the type of structures formed by particles in the matrix [18].

Another field of research that investigates the structure of magnetic fluids is the biomedical technologies based on these systems [24]. In particular, the efficiency of magnetic hyperthermia procedures for the treatment of tumors significantly depends on the type of structures formed by magnetic nanoparticles in biological tissues.

The first model of a ferrofluid with particles combined in linear chains and Y-forks, which had been proposed by Tlusty and Safran [25], predicted a first-order gas–fluid phase transition, where the fluid represented a dense phase consisting of Y-forks. However, the distribution of particles between chains and forks was not considered, although these statistics play an important role in the formation of internal structures and their mutual transformations in ferrofluids [17], thus determining the macroscopic properties of these systems [26]. It should be noted that no conclusion on the first-order phase transition from a gas of chains to the fluid of branched structures [25] has been confirmed in numerical simulations [20, 22].

Thus, no statistical models of ferrofluids with allowance for the formation of branched and network structures have been formulated until now. Below we propose a simple statistical model that takes into account the possible appearance of linear chains, Y-forks, and related network structures and consider the behavior of this system in the absence of an external magnetic field. It is assumed that the central dispersive forces are completely screened by surface layers present on the particles, so that only the magnetic dipole interactions between particles are taken into account.

The results of our theoretical investigation show that, if the concentration of particles and their magnetic interaction energy are sufficiently low, most particles form separate linear chains. If these parameters exceed certain critical values, particles predominantly form branched network structures. The passage from chains to network has a continuous character rather than a discontinuous first-order phase transition. An analogous conclusion on the continuous passage from chains to networks was made earlier [20, 22] based on the results of Monte Carlo simulations.

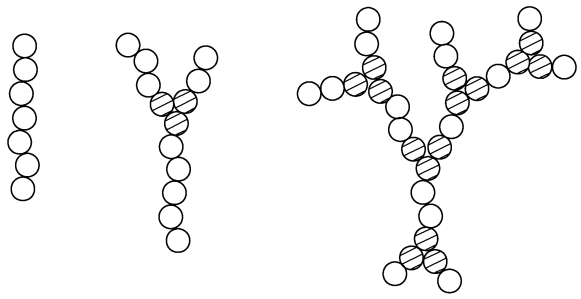

Fig. 1. Structures formed by particles in the ferrofluid model under consideration. Shaded particles form branching sites (nodes) in Y-forks and networks.

2. MAIN ASSUMPTIONS AND STATISTICAL MODEL

Consider a system of identical magnetic spheres possessing constant magnetic dipole moments m. The main assumption of the proposed model is the ability of particles to form linear chains, Y-like forks, and networks consisting of a large number of particles. It should be noted that 2D layers of ferromagnetic particles frequently contain closed ring-shaped aggregates (see, e.g., [6]). However, these rings were not found in 3D model computer simulations of ferrofluids. Moreover, analysis of the dependence of the initial magnetization of ferrofluids on the concentration of particles allows one to conclude that rings are absent in real 3D samples. Although the existence of rings in 3D ferrofluids is still under discussion in the literature, preliminary estimates show that the appearance of rings in the 3D case leads to considerable losses of system entropy, which makes the formation of rings unlikely. In 2D systems, where these losses are relatively small [27], the formation of rings is quite possible. For the simplest analysis, the presence of rings in the system under consideration is ignored.

Examples of possible structures are depicted in Fig. 1. In the framework of one model, it is impossible to stipulate all topological types of networks. Here, we will consider a network consisting of linked Y-forks, which is apparently the topologically simplest case. This network comprises three-particle nodes connected by linear segments (chains). It is assumed that the network represents an infinite structure that contains no triangles, loops, and other closed paths.

Let g_n^{c} be the number of n-particle chains per unit volume of the ferrofluid, g_{ijk}^{Y} is the number of Y-forks with i-, j-, and k-particle arms per unit volume, and g_{ijk}^{N} is the number of nodes connected by i-, j-, and k-particle segments per unit volume. In order to eliminate ambiguity, particles constituting the nodes are not included in segments—that is, not accounted for by the i, j, and k values.

In linear segments (chains), we will take into account only the magnetic interaction between nearest neighbors. This is the usual approximation in the theory of chain aggregates for systems of dipolar particles, and the related error in the free energy of a chain does not exceed ~20% [25]. Finally, we will ignore any interactions between chains, Y-forks, nodes, and segments of the network. This is also a quite well justified approximation for systems with not large concentrations of particles. The influence of interactions between chains on their characteristic dimensions was studied earlier [17] under the assumption that other aggregates and structures in a ferrofluid are absent.

Under the assumptions formulated above, the free energy per unit volume of the ferrofluid can be expressed as follows:

$$\begin{gathered} F = F^{\mathrm{c}} + F^{\mathrm{Y}} + F^{\mathrm{N}}; \\ F^{\mathrm{c}} = k_{\mathrm{B}}T\sum_{n=1}^{\infty} g_n^{\mathrm{c}}\left[\ln\frac{g_n^{\mathrm{c}}V}{e} - \varepsilon^{\mathrm{c}}(n-1)\right], \\ F^{\mathrm{Y}} = k_{\mathrm{B}}T\sum_{i\geq j\geq k=0}^{\infty} g_{ijk}^{\mathrm{Y}}\left[\ln\frac{g_{ijk}^{\mathrm{Y}}V}{e} - \varepsilon^{T} - \varepsilon^{\mathrm{c}}(i+j+k)\right], \\ F^{\mathrm{N}} = k_{\mathrm{B}}T\sum_{i\geq j\geq k=0} g_{ijk}^{\mathrm{N}}\left[\ln\frac{g_{ijk}^{\mathrm{N}}V}{e} - \varepsilon^{T} - \frac{\varepsilon^{\mathrm{c}}}{3}(i+j+k+3)\right]. \end{gathered} \tag{1}$$

Here, F^{c}, F^{Y}, and F^{N} are the free energies per unit volume for chains, Y-forks, and the network, respectively; $k_{\mathrm{B}}T$ is the absolute temperature expressed in energy units; ε^{c} is the average dimensionless (normalized to $k_{\mathrm{B}}T$) energy of magnetic interaction between nearest neighbors in linear chains (segments of branched structures); ε^{T} is the average dimensionless energy of interaction between three particles in the nodes; V is the hydrodynamic volume of a particle (including the surface screening layers). The values of dimensionless energies $\varepsilon^{c,\,T}$ will be estimated in what follows. The terms with logarithms in formulas (1) correspond to the entropy of the ideal gas of chains, Y-forks, and network nodes, respectively. The adopted order of summation over i, j, and k eliminates multiple counting of identical configurations for forks and the network [27]. In the last term of Eq. (1), the factor 1/3 takes into account the fact that the number of linear segments in the model network equals the number of three-particle nodes.

The distribution functions g^{c}, g^{Y}, and g^{N} in Eq. (1), which correspond to a thermodynamically equilibrium state of the ferrofluid, must ensure the minimum of free energy with allowance for the condition of particle balance in the system:

$$\begin{gathered} \varphi^{\mathrm{c}} + \varphi^{\mathrm{Y}} + \varphi^{\mathrm{N}} = \varphi, \\ \varphi^{\mathrm{c}} = \sum_{n=1}^{\infty} n g_n^{\mathrm{c}} V, \\ \varphi^{\mathrm{Y}} = \sum_{i\geq j\geq k=0}^{\infty} (i+j+k+3) g_{ijk}^{\mathrm{Y}} V, \\ \varphi^{\mathrm{N}} = \sum_{i\geq j\geq k=0}^{\infty} \left(\frac{i+j+k}{3}+3\right) g_{ijk}^{\mathrm{N}} V, \end{gathered} \tag{2}$$

where φ is the total volume concentration (number density) of particles in the system and φ^{c}, φ^{Y}, and φ^{N} are the volume fractions of particles entering in chains, Y-forks, and the network.

By minimizing the free energy (1) with allowance for condition (2) and accomplishing standard transformations, we obtain the following distribution functions:

$$\begin{gathered} g_n^{\mathrm{c}} = \frac{\exp(-\varepsilon^{\mathrm{c}})}{V} X^n, \\ g_{ijk}^{\mathrm{Y}} = \frac{\exp(\varepsilon^{T} - 3\varepsilon^{\mathrm{c}})}{V} X^{i+j+k+3}, \\ g_{ijk}^{\mathrm{N}} = \frac{\exp(\varepsilon^{T} - 2\varepsilon^{\mathrm{c}})}{V} X^{2+(i+j+k+3)/3}, \end{gathered} \tag{3}$$

where X is the Lagrange multiplier. In order to determine the latter quantity, expressions (3) have to be substituted into conditions (2). The first relation (3) readily yields

$$\varphi^{\mathrm{c}} = \exp(-\varepsilon^{\mathrm{c}})\frac{X}{(1-X)^2}. \tag{4}$$

Denoting the total number of particles in a fork by $n = i + j + k + 3$, we obtain the following expression for the volume fraction of forks:

$$\begin{aligned} \varphi^{Y} &= \exp(\varepsilon^{T} - 3\varepsilon^{\mathrm{c}})\sum_{n=3}^{\infty} nX^n\left(\sum_{j=1}^{n-2}\sum_{k=1}^{j} 1\right) \\ &= \exp(\varepsilon^{T} - 3\varepsilon^{\mathrm{c}})\frac{3X^3}{(1-X)^4}. \end{aligned} \tag{5}$$

Analogous considerations lead to the relation

$$\begin{aligned} \varphi^{\mathrm{N}} &= \exp(\varepsilon^{T} - 3\varepsilon^{\mathrm{c}}) \\ &\times X^2\sum_{n=3}^{\infty}\left(\frac{n}{3}+2\right)X^n\left(\sum_{j=1}^{n-2}\sum_{k=1}^{j} 1\right) \\ &= \exp(\varepsilon^{T} - 2\varepsilon^{\mathrm{c}})\frac{X^3(3-2X^{1/3})}{(1-X^{1/3})^4}. \end{aligned} \tag{6}$$

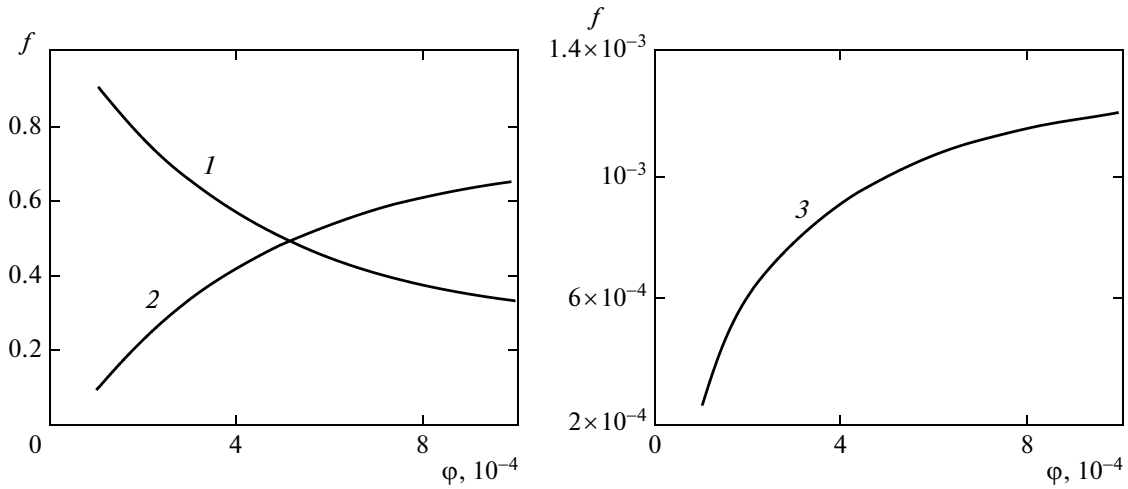

Fig. 2. Relative concentrations f of magnetic particles combined in (*1*) linear chains, (*2*) network, and (*3*) Y-forks as functions of the total volume concentration φ of particles, calculated in the fluctuation-free approximation (7) for $\varepsilon_0^c = 5$.

Substituting expressions (4)–(6) into condition (2), we obtain an equation with respect to X, which can be readily solved by numerical methods.

3. DISTRIBUTION OF PARTICLES OVER AGGREGATES

In order to determine the Lagrange multiplier X, it is necessary to evaluate dimensionless energies ε^c and ε^T. To a first approximation, this can be done by ignoring fluctuations in the arrangement and orientation of particles in linear segments and three-particle nodes, i.e., by considering the ground states of these clusters. The dimensionless energies of the ground states of linear segments and nodes were determined earlier [27] and, in the adopted notations, can be expressed as follows:

$$\varepsilon^c = \varepsilon_0^c = \frac{\mu_0}{2\pi}\frac{m^2}{d^3 k_B T}, \quad \varepsilon^T = \varepsilon_0^T = \frac{15}{8}\varepsilon_0^c, \qquad (7)$$

where μ_0 is the magnetic permeability of a vacuum, d is the hydrodynamic diameter of a particle (with allowance for the surface screening layer), and subscript "0" indicates the ground state of a cluster. By substituting expressions (7) into formulas (4)–(6) and then into the expression for φ^c in Eq. (2), one can determine X. Finally, formulas (4)–(6) yield expressions for φ^c, φ^Y, and φ^N.

Figure 2 presents some results of calculations of the relative concentrations of aggregates defined as $f^{c,Y,N} = \varphi^{c,Y,N}/\varphi$. Estimates show that, for typical magnetite-based ferrofluids with particles covered by approximately 2-nm-thick surfactant layers, the value of $\varepsilon_0^c = 5$ corresponds to a magnetic core diameter of about 15–16 nm. Particles with these dimensions are present in many modern ferrofluids. The results presented for this case in Fig. 2 show that, at small total volume concentrations φ of particles, most of them form linear chains. As φ increases, the fraction of particles entering into the network increases. When φ exceeds a certain critical value (corresponding to the intersection of curves *1* and *2*, the number of particles in the network becomes greater than that in separate chains. The number of particles occurring in forks is significantly smaller than their numbers in chains and the network.

The threshold value of concentration φ corresponding to the model ferrofluid (Fig. 1) is very small, on the order of several hundredths of a percent. In modern ferrofluids, the "magnetic" volume concentration of particles (i.e., that determined with allowance for only their magnetic cores, ignoring surface layers) frequently reaches 7–10% (see, e.g., [26]). Taking this into account, one might expect that, in the absence of an external magnetic field, all particles with magnetic core diameters of about 15 nm or more must be combined in network aggregates. However, this hypothesis has not been confirmed by experiments.

In order to obtain more adequate quantitative estimates of the concentration of particles in aggregates, it is necessary to take into account the fluctuations of particles in linear and triangular clusters. Allowance for these fluctuations leads to renormalization of the ε^c and ε^T values. Analysis shows that the results of this modification depend on the system dimensionality and are somewhat different for 2D layers and 3D volume samples. In the case of 2D layers, corrections to the energies of particle interactions in chains and three-particle nodes had been estimated earlier [27]. Calculations of the corrections to ε^T in 3D ferrofluids are very cumbersome and worthy of publication by themselves. Here, in order to obtain physically signif-

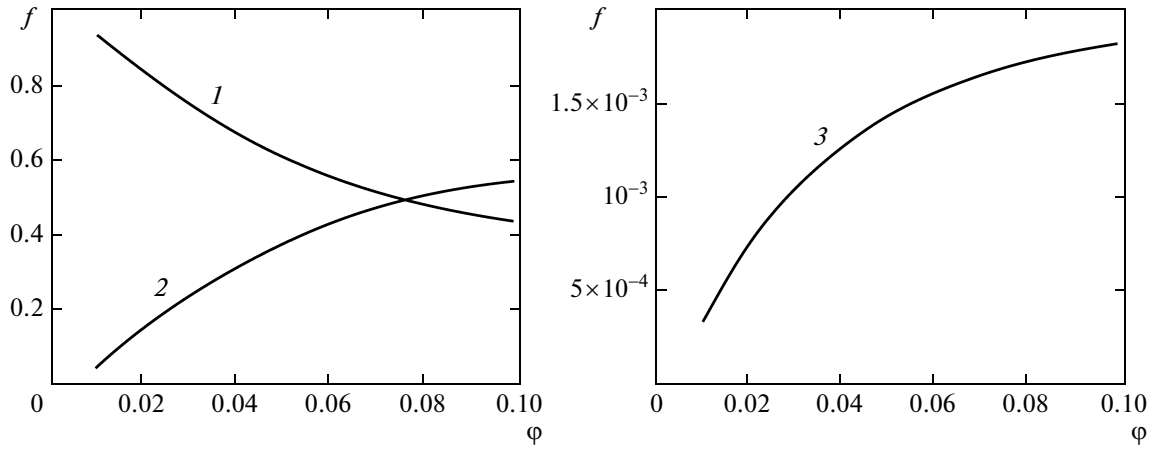

Fig. 3. Same as in Fig. 2, but calculated for $\varepsilon_0^c = 5$ using relations (8) instead of (7).

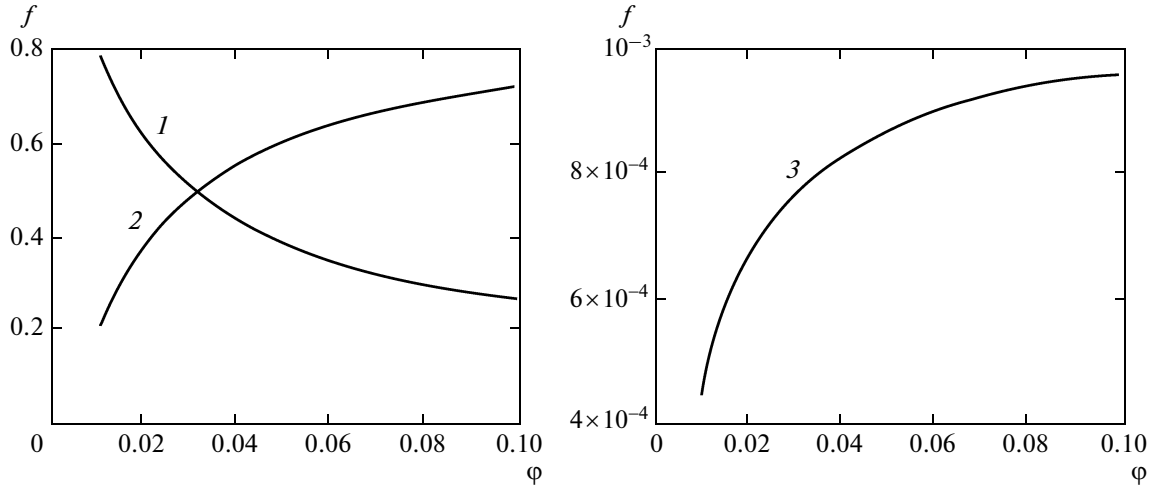

Fig. 4. Same as in Fig. 3, but calculated for $\varepsilon_0^c = 6$.

icant fundamental results, we will use estimates of the ε^c and ε^T values for 2D layers [27].

In the adopted notations, these are as follows:

$$\varepsilon^c \approx \varepsilon_0^c - \ln\left(\frac{3\sqrt{3}\varepsilon_0^c}{4}\right),$$
$$\varepsilon^T \approx \frac{15}{8}\varepsilon_0^c - \ln\left(\frac{2\pi^{1/2}}{0.034}\left(\frac{\varepsilon_0^c}{2}\right)^{9/2}\right). \quad (8)$$

Figure 3 shows the results of calculations of the relative concentrations $f^{c, Y, N}$ obtained using estimations (8). Qualitatively, the dependence of the relative concentrations f on the total volume concentration φ of particles in the ferrofluid is generally the same as in Fig. 2. However, the threshold value of φ for the transition from chains to network in Fig. 3 is about 8%, which seems more reasonable than the value obtained in the fluctuation-free approximation (7). In order of magnitude, the corrected value of the threshold concentration agrees with the results of computer simulations [20, 21], where the formation of networks was observed at concentrations on the order of several percent.

Note that the difference between the threshold concentrations according to Figs. 2 and 3 amounts to two orders of magnitude. Therefore, allowance for the fluctuations of particles in clusters is necessary for obtaining adequate results.

Figure 4 shows the results of calculations analogous to those presented in Fig. 3, but performed for a somewhat greater energy ε_0^c of interaction between particles in the chain. A comparison of Figs. 3 and 4 shows that the threshold concentration is very sensitive to this energy (i.e., to the size of interacting particles) and rapidly decreases with increasing ε_0^c.

Simple analysis shows that the dimensionless chemical potentials of particles in the ferrofluid can be expressed as

$$\chi = \frac{V}{k_B T}\frac{\partial F}{\partial \varphi} = nX - \varepsilon^c. \quad (9)$$

Figure 5 shows the results of calculating the dependence of χ on the particle concentration φ. The monotonic growth in $\chi(\varphi)$ in the entire interval, including the region of intersection of curves *1* and *2* in Fig. 4, shows that the passage from separate chains to a network is not a discontinuous first-order phase transition. An analogous conclusion that the onset of network formation is not a first-order phase transition was made [20, 22] based on the results of Monte Carlo simulations.

Our previous analysis [17] of the gas–liquid phase transition in a ferrofluid consisting of separate chains (without branched structures) showed that this transition could be expected for $\varepsilon_0^c > 5.6$–5.8 and $\varphi > 11$–12%. Therefore,

we can also be expect that the network formation in the system of magnetic particles takes place at lower ε_0^c and φ values than those necessary for the gas–liquid transition. This conclusion agrees with observations of network structures in experiments [6, 18, 19] and numerical simulations [19, 20, 22].

It should be noted that the formation of networks and other heterostructures in ferrofluids can lead to radical changes in the dynamic properties of these systems. In particular, these changes can be responsible for the fact that features of the magnetization reversal kinetics characteristic of spin glasses have been observed in ferrofluids [28]. The effect of internal structures on the dynamic properties of ferrofluids is worthy of special investigation.

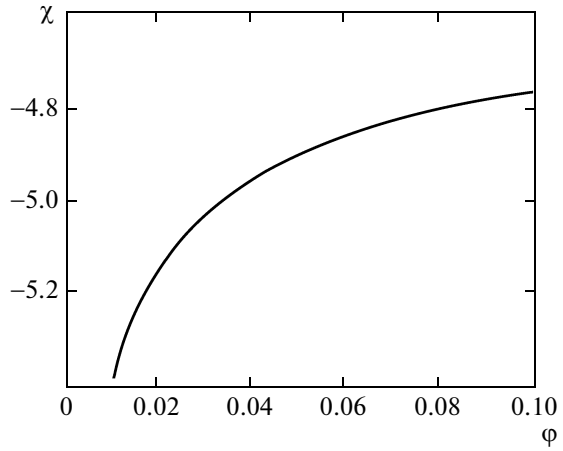

Fig. 5. Dimensionless chemical χ potential of particles as function of their total volume concentration φ, calculated for $\varepsilon_0^c = 6$.

4. CONCLUSIONS

We have theoretically studied a thermodynamically equilibrium state of the ferrofluid consisting of identical spherical monodomain particles in the absence of an external magnetic field. Based on the results of well-known laboratory experiments and computer simulations it was suggested that the magnetic particles can form linear chains, Y-forks, and connected network structures. It was assumed that the particles only interact as magnetic dipoles, while the central dispersive forces are completely screened by surface layers.

Our analysis showed that, if the volume concentration of particles and their magnetic interaction energy are small, most particles form separate linear chains. If these parameters exceed certain critical values, most particles concentrate so as to form branched network structures. The relative number of particles entering in Y-forks is small compared to the numbers of particles in the chains and network. In contrast to the model [25], which ignored the statistical character of the distribution of particles over linear chains and segments of branched structures, the proposed model analysis showed that the passage from a state with most particles being predominantly combined in chains to the state with a predominant network structure has a continuous character rather than a discontinuous first-order phase transition. This conclusion is confirmed by the results of numerical simulations [20, 22].

A comparison of our results to those obtained using a model [17] of the phase condensation in a ferrofluid consisting of chains leads to the conclusion that formation of the network is preceded, at least, by the condensation of particles in a homogeneous liquidlike phase. This agrees with the results of laboratory experiments and numerical simulations [6, 18–20, 22], in which network clusters of magnetic particles have been observed.

We have considered the topologically simplest structure with three-particle nodes connecting three linear segments, free of triangles and any other closed loops. Evidently, allowance for the appearance of many-particle nodes, loops, and other topological features would increase the probability of the formation of network clusters. Therefore, taking these features into account would not change the main conclusion of this study that network appear prior to the phase condensation of particles into a homogeneous dense liquidlike phase.

The proposed model ignores the interaction between particles belonging to different clusters. For this reason, the model analysis cannot describe the evolution of the network structure with an increasing number of particles and their interaction energy. In principle, there are two possible scenarios for this evolution. First, an increase in the interaction energy and particle concentration can lead to an increase in the

network density, the appearance of nodes shared by more than three linear segments, and an increase in the number of these nodes, while the characteristic network structure is retained. Second, the network can exhibit a collapse and transform into a homogeneous dense phase at certain values of the interaction energy and particle concentration. The former scenario was considered in [20, 22] and the latter was discussed in [21]. An analysis of the problem of network structure evolution and allowance for a more general network topology are subjects for separate investigations.

ACKNOWLEDGMENTS

This study was supported in part by the Russian Foundation for Basic Research (project nos. 10-01-96002-Ural, 10-02-96001-Ural, 10-02-00034, and 12-01-00132) and the Ministry of Education and Science of the Russian Federation (project no. 2.1267.2011).

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Translated by P. Pozdeev