Spontaneous ferromagnetic ordering in magnetic fluids

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This paper is devoted to the theoretical justification of spontaneous orientational order in magnetic fluids. We study the self-consistent solutions of the Bogoliubov-Born-Green-Kirkwood-Yvon equation connecting the one-particle distribution function with the pair correlation function. This self-consistent approach is used in the specific density functional method and proves to be equivalent to the mean field theory. On the basis of the second-order perturbation method over the intensity of dipole-dipole interparticle interaction the following effect is discovered: the self-consistent density functional approach leads to the spontaneous "ferrimagnetic" state of the magnetic fluid induced by the dipole-dipole interaction. This strange result seems to be physically meaningless and prejudices the validity of the density functional methods and mean field theories applied to orientational microstructure in ferrofluids.

Dipolar fluids are a widely studied model in statistical mechanics. These systems are important not only because dipolar interactions are omnipresent, but also because they play a prominent role in many artificial systems, such as ferrofluids, magnetorheological and electrorheological suspensions. Recent computer simulations [1–4] have shown that the microscopic structure of dipolar fluids is much more complex than previously expected. The system at high dipolar strength and low volume fraction has proved to associate in chain aggregates [2–4], while at higher volume fraction a spontaneous formation of a ferroelectric phase has been observed [1]. Presence of an additional attractive force has resulted in a usual condensation, as in the Stockmayer fluid [4].

Applying to ferrofluids, a lot of experimental studies, demonstrating not only the chainlike aggregate existence, but also their great influence upon rheological, diffusional and hydrodynamic properties of ferrofluids, are worth mentioning (see, for example, Ref. [5]). The condensation of ferroparticles, known as phase separation, has also been observed [6,7]. Concerning the spontaneous orientationally ordered state ("ferromagnetic order") in ferrofluids, it should be noted that direct experimental evidences have never been found. The only exception is the paper [7] reporting the dominance of ferromagnetic fluctuations for dense ferrofluids.

In this paper we focus our attention on the basic problem of spontaneous orientational order in magnetic fluids. The possibility of such state practical realization is not only of great interest from the principal physical point of view, but also is of, no doubt, importance for many applications. It was in the beginning of 1980s when the orientational order (ferromagnetic state) was predicted by using the Weiss mean field method for accounting the interparticle dipole-dipole interaction in ferrofluids [8]. In the 1990s, after computer modeling results on the properties of polar fluids were published [1], the interest in the problem arose again. In these and later papers [9] the following fact was pointed out: while resting spatially disordered, the polar fluid and ferrofluid undergo the phase transition resulting in the appearance of orientational order. The subsequent theories [10,11], developed for polar fluids and ferrofluids, proved the existence of "liquid paramagnetic–liquid ferromagnetic" phase transition, the conditions of which turned out to be dependent on the shape of the container with a ferrofluid. The highly elongated ellipsoid of revolution is the most suitable and, in this case, the direction of spontaneous magnetization coincides with the major semiaxis of the ellipsoid. It is worth noting that the mean spherical model and the thermodynamic perturbation theories [12,13], describing well the magnetic properties of practically used ferrofluids, do not predict the ferromagnetic order. The last mentioned result was not also obtained in computer calculations on ferrofluid and polar fluid microstructure [3,14].

Here we are not going to dwell on the peculiarities of computer simulations and physical reasons for the spontaneous orientational ordering in fluids with the interparticle magnetic dipole-dipole interaction. The properties of one-particle distribution function are considered below, and the density functional approach [10] is proved to be equivalent to the mean field theory. We study the self-consistent solutions of the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) equation connecting the one-particle distribution function with the pair correlation function. On the basis of the second-order perturbation method over the dipole-dipole interparticle interaction intensity, the following effect is discovered: the self-consistent density functional approach leads to the spontaneous "ferrimagnetic" state of the magnetic fluid induced by the dipole-dipole interaction. This strange result seems to be physically meaningless and prejudices the validity of the density functional methods and mean field theories applied to the orientational microstructure in ferrofluids. It should be pointed out that we do not cast doubt on the density functional theory in general, since a lot of physically successful applications of this method are known.

A modern theoretical approach to the problem of spontaneous orientational ordering in dipole fluids is based on the density functional algorithm [10]. Thermodynamic energy of the system is self-consistently expressed as a functional of the one-particle distribution function $g_1(\mathbf{\Omega})$. This function determines the probability for the randomly chosen magnetic particle 1 to be oriented along the $\mathbf{\Omega}_1(\omega_1;\zeta_1)$ direction.

Minimization of the energy functional leads to the integral equation for the one-particle distribution function, one of the solution of which is anisotropic in the orientational space $\mathbf{\Omega}$, even in the absence of an external field. The function $g_1(\mathbf{\Omega}_1)$ fulfills the normalization condition and determines the ferrofluid magnetization M:

$$\int g_1(\omega_1)d\mathbf{\Omega}_1 = 1,$$

$$M(H) = nm\frac{1}{2}\int_0^\pi \cos \omega_1 g_1(\omega_1)\sin \omega_1 \, d\omega_1, \qquad (1)$$

$$d\mathbf{\Omega}_i = (4\pi)^{-1}\sin \omega_i \, d\omega_i \, d\zeta_i,$$

where n is the ferroparticle concentration and m is the particle magnetic moment; $\mathbf{H}\|Oz$ stands for an external magnetic field being parallel to major axis Oz of the container with ferrofluid in a shape of infinitely elongated ellipsoid of revolution. Applying to ferrofluids the one-particle distribution function g_1 depends only on the angle ω_1 between the orientation of a magnetic moment of randomly chosen ferroparticle 1 and an external field direction. On the basis of BBGKY formalism the differentiation of function $g_1(\omega_1)$ with respect to ω_1 leads to the equation [13], connecting the one-particle distribution function with the pair correlation function $g_2(\mathbf{r}_1,\mathbf{r}_2,\mathbf{\Omega}_1,\mathbf{\Omega}_2) \equiv g_2(12)$,

$$\frac{dg_1(\omega_1)}{d\omega_1} = -\alpha \sin \omega_1 g_1(\omega_1) - \frac{n}{kT}\int d\mathbf{\Omega}_2$$

$$\times \int d\mathbf{r}_{12}\frac{dU_d(12)}{d\omega_1}g_2(12). \qquad (2)$$

$$d\mathbf{r}_{ij} = r_{ij}^2 \, dr_{ij}\sin \theta_{ij} \, d\theta_{ij} \, d\varphi_{ij}.$$

Here $U_d(12)$ is the energy of dipole-dipole interaction between the magnetic moments of ferroparticles 1 and 2; $\alpha = mH/kT$ has a meaning of the Langevin parameter; the vector $\mathbf{r}_{ij}(r_{ij},\theta_{ij},\varphi_{ij})$ connects the centers of spherical particles i and j; the integrations over $\mathbf{\Omega}_2$ and $d\mathbf{r}_{12}$ correspond to the averaging over all orientations and positions of the second ferroparticle; and we use the fact that the spatial two-particle probability depends only on the mutual interparticle distance $\mathbf{r}_{ij}$, that is, $g_2(\mathbf{r}_1,\mathbf{r}_2,\mathbf{\Omega}_1,\mathbf{\Omega}_2) = g_2(\mathbf{r}_{12},\mathbf{\Omega}_1,\mathbf{\Omega}_2)$. Equation (2) is exact and includes all the corrections to the one-particle distribution function influenced by multiparticle correlations. Moreover, Eq. (2) describes the equilibrium distribution and, thus, its solution ensures the minimum of the system free energy. In this case, this approach is equivalent to the variation method used in the density functional approach. Besides that, Eq. (2) is of principal nature. On this basis the cluster expansion methods may be developed to determine the ferrofluid magnetization (polar fluid polarization) in the arbitrary values of an external field. An important feature of the method is that the right part of Eq. (2) contains the first orders of particle concentration n and dipole-dipole interaction potential U_d. Thus, the determination of the pair correlation function $g_2(12)$ up to the order of $\sim n^k, U_d^k$ gives

the results in terms of order $\sim n^{k+1}, U_d^{k+1}$. Naturally, in the case of negligibly weak interparticle interaction, Eq. (2) gives the solution for an ideal paramagnetic gas:

$$g_1(\omega_1) \equiv g_0(\omega_1) = (\alpha/\sinh \alpha)\exp(\alpha \cos \omega_1), \qquad (3)$$

$$M = M_L(\alpha) = nmL(\alpha), \quad L(\alpha) = \coth \alpha - 1/\alpha,$$

where M_L stands for the Langevin magnetization.

From the definition of the pair correlation function, the following expression, equivalent to the virial expansion, holds true:

$$g_2(12) = g_0(\omega_1)g_0(\omega_2)\widetilde{g}_2(12), \qquad (4)$$

where the $\widetilde{g}_2(12)$ function takes into account the interparticle correlations in a ferroparticle system,

$$\widetilde{g}_2(12) = \frac{1}{ZV^{N-2}}\int \prod_{i=3}^N d\mathbf{r}_i \int \prod_{i=3}^N g_0(\omega_i)d\mathbf{\Omega}_i \exp\left(-\frac{\hat{H}}{kT}\right), \qquad (5)$$

$$Z = \frac{1}{V^N}\int \prod_{i=1}^N d\mathbf{r}_i \int \prod_{i=1}^N g_0(\omega_i)d\mathbf{\Omega}_i \exp\left(-\frac{\hat{H}}{kT}\right),$$

$$\hat{H} = \sum_{i<j=1}^N [U_s(ij) + U_d(ij)].$$

Here Z is the partition function and $\hat{H}$ is the interaction Hamiltonian of N ferroparticles freely moving and rotating inside volume V and interacting through the central energy U_s and the dipole-dipole energy U_d. It is worth mentioning that $\widetilde{g}_2$ should be considered as the integral-type functional (5) on g_0: $\widetilde{g}_2(12) = \widetilde{g}_2[12,g_0]$.

The solution of Eqs. (2) and (4) with the accuracy of terms $\sim U_d^2$ was considered in Ref. [13]. The results obtained for magnetic properties of dense ferrofluids and dielectric properties of polar fluids are in a good agreement with the experimental data [15] and the computer simulations [14] including the ferrofluids with maximum allowable magnetic phase concentration $\sim 18\%$, saturation magnetization ~ 90 kA/m, and the values of Langevin initial susceptibility $\chi_L = 4\pi nm^2/3kT \sim 6$. The ferrofluid magnetization [13] is expressed in a form of rather complicated series on the Langevin magnetization and equals to zero in the absence of an external field. The Eqs. (2) and (4) solution structure is typical in all orders of the thermodynamic perturbation theory over the dipole-dipole interaction energy. In other words, the exact expression (4) does not predict the existence of a "ferromagnetic" state in ferrofluids.

The density functional approach [10] uses the self-consistent expression of the pair correlation function $g_2(12)$ in terms of the one-particle distribution function g_1:

$$g_2(12) = g_1(\omega_1)g_1(\omega_2)\widetilde{g}_2[12,g_1]. \qquad (6)$$

This expression differs from the exact one, Eq. (4), in sub-

stitution of an ideal paramagnetic gas distribution function $g_0(\omega_i)$ by the unknown one $g_1(\omega_i)$. It should be pointed out that such replacement is only one of the approximations being used in statistical mechanics, which could not be treated

as an exact statistical result. With the help of Eqs. (2) and (6) one can easily get the self-consistent integral equation for the one-particle distribution function in an exponential form according to an ideal paramagnetic gas:

$$g_1(\omega_1) = \frac{\exp\left[\alpha\cos\omega_1 - \dfrac{n}{kT}\displaystyle\int d\omega_1 \int d\mathbf{\Omega}_2\, g_1(\omega_2) \int d\mathbf{r}_{12}\dfrac{dU_d(12)}{d\omega_1}\tilde{g}_2[12,g_1]\right]}{\displaystyle\int d\mathbf{\Omega}_1 \exp\left[\alpha\cos\omega_1 - \dfrac{n}{kT}\int d\omega_1 \int d\mathbf{\Omega}_2\, g_1(\omega_2) \int d\mathbf{r}_{12}\dfrac{dU_d(12)}{d\omega_1}\tilde{g}_2[12,g_1]\right]}. \tag{7}$$

The only difference between Eqs. (7) and (3) is that the exponential factor in Eq. (7) contains the sum of an external field and the function of interparticle dipole-dipole correlations. The last one should be expressed in terms of ferrofluid magnetization M. This sum should be considered as effective magnetic field acting on a single particle.

To study the properties of Eq. (7) let us consider the expansion of $\tilde{g}_2[12,g_1]$ with zero-order accuracy over the dipole-dipole potential U_d. As was mentioned earlier, this approximation was equal to the first-order perturbation method. In this case we assume that all interparticle correlations are controlled by the spherical part of the interparticle interaction energy $U_s(ij)$ (hard or soft sphere repulsion, van der Waals attraction, etc.), that is, $\tilde{g}_2[ij,g_1]=g_s(ij)$. Here $g_s(ij)$ stands for the pair distribution function of the reference system [system of particles, interacting through the central interparticle energy $U_s(ij)$ only]. Substituting the function $g_s(12)\equiv g_s(|\mathbf{r}_1-\mathbf{r}_2|)$ in Eq. (7) we solve the problem of dipole-dipole interaction U_d averaging over all positions and orientations of particle 2. This integration depends on the shape of the system (but does not depend on the volume and on the type of central interparticle interaction). It is important to stress that using the infinitely elongated ellipsoidal shape is of advantage because this is just the case when demagnetization factors are of no consequence and do not need to be accounted for. For this shape of a ferrofluid container we get

$$g_1(\omega_1)=(\alpha_e/\sinh\alpha_e)\exp(\alpha_e\cos\omega_1),$$

$$M=M_L(\alpha_e)=nmL(\alpha_e), \tag{8}$$

$$\alpha_e=mH_e/kT, \quad H_e=H+4\pi M(H)/3.$$

The expression for an effective magnetic field H_e coincides with those in the Weiss mean field theory. Thus, the self-consistent density functional approach (6) is equivalent to the mean field theory, and in the framework of the first-order perturbation method predicts the ferromagnetic phase transition in the absence of an external field, when $\chi_L\geqslant 3$. Apparently, this is caused by the fact that in exact virial expansion (4) of the pair correlation function all the influence of the dipole-dipole interaction is determined only by function $\tilde{g}_2[12,g_0]$. The approximate self-consistent expression (6)

imposes an additional influence by way of one-particle distribution functions $g_1(\omega_i)$. The latter also takes into account the dipole-dipole interaction, the influence of which turns out to be overestimated. (It is worth mentioning that for moderately concentrated ferrofluids with rather small particles and initial Langevin susceptibility $\chi_L\sim 3$, none of the experimental and computer studies demonstrate the existence of ferromagnetic order). Therefore, it is not a surprise that such excess account for the dipole-dipole interaction leads to the prediction of a ferromagnetic state in dipolar fluids.

The behavior appears to be much stranger when this effective field is calculated on the basis of the second-order perturbation method [13], in the framework of which the pair correlation function $\tilde{g}_2[12,g_1]$ is determined under the condition when all the corrections linear in dipole-dipole interaction energy U_d are taken into account. Using the self-similar replacement (6) from the definition of the pair correlation function, we get

$$\tilde{g}_2[12,g_1]=[1-\beta U_d(12)]g_s(12)+\exp[-\beta U_s(12)]n$$

$$\times\left\{\int d\mathbf{r}_3\int d\mathbf{\Omega}_3\, g_1(\omega_3)[-\beta U_d(13)\right.$$

$$-\beta U_d(23)]\exp[-\beta U_s(13)-\beta U_s(23)]$$

$$-2\int d\mathbf{r}_{34}\int d\mathbf{\Omega}_3\int d\mathbf{\Omega}_4\, g_1(\omega_3)g_1(\omega_4)$$

$$\left.\times[-\beta U_d(34)]\exp[-\beta U_s(34)]\right\},$$

$$\beta=1/kT. \tag{9}$$

The expression for the pair correlation function following from the averaging over the positions and orientation of the third and fourth particle in Eq. (9) could be found in Ref. [13]. The result for the one-particle distribution function represents the exponential factor in expression (7) in a form of expansion over the first- and second-order Lagrange polynomials $P_n(z)$, $n=1,2$:

$$g_1(\omega_1) = \frac{\exp[G_1(\alpha,\tilde{M},Q)P_1(\cos\omega_1) + G_2(\tilde{M})P_2(\cos\omega_1)]}{\int d\Omega_1 \exp[G_1(\alpha,\tilde{M},Q)P_1(\cos\omega_1) + G_2(\tilde{M})P_2(\cos\omega_1)]}, \tag{10}$$

$$G_1(\alpha,\tilde{M},Q) = \alpha + \chi_L\tilde{M} + (\chi_L^2/48)\tilde{M}(1+29Q-96\tilde{M}^2), \quad G_2(\tilde{M}) = 29\chi_L^2\tilde{M}^2/96,$$

$$\tilde{M} = \int d\Omega_1 g_1(\omega_1)P_1(\cos\omega_1), \quad Q = \int d\Omega_1 g_1(\omega_1)P_2(\cos\omega_1).$$

Here $\tilde{M} = M/nm$ is the relative magnetization, and Q has a meaning of quadrupole moment of the one-particle distribution function describing the degree of ferroparticle moment parallel alignment. Since we are interested in a one-particle distribution function zero field behavior, the coefficients G_1 and G_2 of expansion (10) should be determined numerically from the following set of equations:

$$\tilde{M} = \frac{\int d\Omega_1 \exp[G_1(0,\tilde{M},Q)P_1(\cos\omega_1) + G_2(\tilde{M})P_2(\cos\omega_1)]P_1(\cos\omega_1)}{\int d\Omega_1 \exp[G_1(0,\tilde{M},Q)P_1(\cos\omega_1) + G_2(\tilde{M})P_2(\cos\omega_1)]}, \tag{11}$$

$$Q = \frac{\int d\Omega_1 \exp[G_1(0,\tilde{M},Q)P_1(\cos\omega_1) + G_2(\tilde{M})P_2(\cos\omega_1)]P_2(\cos\omega_1)}{\int d\Omega_1 \exp[G_1(0,\tilde{M},Q)P_1(\cos\omega_1) + G_2(\tilde{M})P_2(\cos\omega_1)]}.$$

In the case where no external magnetic field is present ($\alpha=0$) two kinds of the solution exist. The first one is trivial, when the distribution moments $\tilde{M}$ and Q equal to zero. Consequently, we get $G_1=G_2=0$ and $g_1(\omega_1)=1$. It means that the orientational distribution of ferroparticle magnetic moments is homogeneous according to no magnetized liquid state of ferrofluid.

Besides that, when the interparticle dipole-dipole interaction is rather intensive and the particle concentration is not low, the bifurcation of a solution occurs, resulting in the appearance of the inhomogeneous orientational distribution. The unique parameter determining the bifurcation point is the initial Langevin susceptibility χ_L. From set (11) the critical bifurcation value χ_* may be easily obtained; $\chi_* = 12(\sqrt{5}-2) \approx 2.833$. The last bifurcation value is less than the critical one $\chi_L=3$ obtained from the Weiss mean field approach (8). In the region of $\chi_L > \chi_*$, the solution of the equation set (11) is demonstrated in Fig. 1. Both the quantities $\tilde{M}$, and Q, are the increasing functions of χ_L, and in the local vicinity of the critical point the following scaling dependencies hold true:

$$\tilde{M} \sim (\chi_L-\chi_*)^{1/2}, \quad Q \sim (\chi_L-\chi_*). \tag{12}$$

The analysis of Fig. 1 exposes the strange feature of this anisotropic solution. With the χ_L growth the quadrupole moment Q asymptotically reaches the maximum allowable value, that is, unity: $Q \to 1$, $\chi_L \gg \chi_*$. This means that all magnetic moments tend to align along the Oz axis of the container for strongly interacted and concentrated ferropar-

ticle system. However, the relative magnetization does not exceed the value $\tilde{M}_*$, $\tilde{M}_* \approx 0.6$. This leads to a very surprising conclusion that for $\chi_L \gg \chi_*$ about 80% of the particle magnetic moments are codirectional to the Oz axis, and nearly 20% of the ferroparticles are aligned antiparallel. This effect is demonstrated in Fig. 2, and the anisotropic orientational distribution $g_1(\omega_1)$ is characterized by the presence of two maximums describing the probabilities for the randomly chosen magnetic moment to be coparallel ($\omega_1=0$) or antiparallel ($\omega_1=\pi$) to a major ellipsoid Oz axis. This behavior of the one-particle distribution function looks like the liquid ferrimagnetic state instead of the liquid ferromagnetic state, predicted by the first-order perturbation method (8) (the Weiss mean field model [8]) and by the theoretical [10] and computer models [1,9]. A mathematical reason is that, unlike
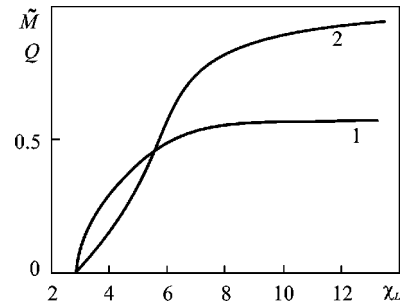

FIG. 1. Anisotropic solution of Eqs. (11): curve 1 is the dependence of the spontaneous relative magnetization $\tilde{M}$ on the initial susceptibility χ_L in the region $\chi_L > \chi_*$; curve 2 is the same dependence for quantity Q.
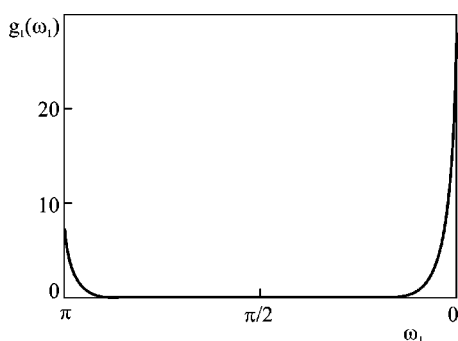
FIG. 2. Zero field inhomogeneous one-particle distribution function $g_1(\omega_1)$ (10): the character angle dependence for case $\chi_L = 8$.

the first-order perturbation method (7), the approximation of the second perturbation order over the dipole-dipole energy takes into account the interaction in all pairs and triplets of ferroparticles [13]: namely, the account of dipole-dipole interaction in ferroparticle triplets results in disordering of magnetic moments. Since we cannot find any physical reasons substantiating the ferrimagnetic ordering in ferrofluids

(and the ferromagnetic one as well), the only physically meaningful solution of Eqs. (11) is the homogeneous, orientationally undisordered liquid state of ferrofluid in the absence of an external field.

The point is that the second-order self-consistent perturbation model does not predict the spontaneous ferromagnetic orientational ordering. So, any principal physical conclusion concerning the possibility of a ferromagnetic state in ferrofluids, arising from the mean field theories and density functional models [10], has no experimental verification and is not to be treated as an exact theoretical result. This conclusion seems to be an artificial consequence from the self-consistent approximation (6), and the phenomenon of spontaneous orientational ordering in magnetic fluids induced by the dipole-dipole interaction has to be considered extremely questionable.

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