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AGGREGATION OF NANOPARTICLES IN A NEMATIC LIQUID CRYSTAL

We present a short review of our general result in [1], which concerns the behavior of a collection of nanoparticles in a nematic liquid crystal. Such liquid crystal colloids are studied for a long time, have highly peculiar characteristics, and have attracted a lot of the interest in different practical applications. A simple model of the aggregation of nanoparticles in a liquid crystal is proposed. With regard for the collective effect of the interaction between the small particles through a change in the scalar order parameter in a nematic liquid crystal, the inhomogeneous distribution of particles is predicted as distinct from possible structures in the system of macroscopic particles in a liquid crystal. The inhomogeneity length and the size of a cluster of nanoparticles are determined, and the possible peculiarities in the behavior of liquid crystals with introduced nanoparticles are described.

Keywords: nanoparticles, nematic liquid crystal, collective effect, inhomogeneity.

The intensive consideration of the suspensions in liquids has been recently started in science and technology. The colloid suspensions of solid particles coated with a surfactant and the dispersions of liquid droplets are media, which has attracted a lot of crucial interest in different practical applications, as well as in medicine [2]. Small particles suspended in a nematic liquid crystal make a new composite material with unique physical properties. Mechanical and optical properties of this matter are defined, first of all, by the collective behavior of this system [3]. Depending on the size and anchoring energies, the particles form chains [4–6], anisotropic clusters [7], cellular structures [8, 9], and periodic structures [10–17]. The interaction between particles in such systems resulting in the formation of structures crucially depends on the sizes of particles [18–20] and the thickness of a liquid crystal cell [21].

The origin of the structure formation comes from the overlap of distortions of the director field that

are caused by single particles. These distortions interfere and result in a fascinating anisotropic interaction between the particles. The director deformations greatly depend on the size of particles and the anchoring energy. For the normal and planar anchorings, the director prefers to be, respectively, normal and parallel to the surface of the particles. In the case of a strong anchoring, the boundary conditions on \mathbf{n} are fixed and impose topological constraints on the director field around a particle. In this case, topological defects, which cannot be removed from the particle, arise. It was shown, however, that the interesting properties and new structures in suspensions of asymmetric particles can appear [22–24]. For the first time, Brochard and de Gennes in 1970 showed that the “doping” of a nematic liquid crystal with ferromagnetic cylindrical grains leads to the macroscopic collective behavior [23, 25]. This behavior is manifested as a distortion of the uniform molecular orientation of the entire matrix upon the application of an external magnetic field. In other words, magnetic grains govern the orientation of the whole nematic liquid

crystal matrix. This was confirmed experimentally by Chen and Amer [26, 27]. They found that the doped nematic exhibits a “cellular” texture with the “cells” of order of tens of micrometers. The doped nematic liquid crystal system in a magnetic field was examined theoretically by Burylov and Raikher [28, 29], but inasmuch as the elastic interaction between the grains was not taken into account.

Another class of liquid crystal colloids is presented by the media with small particles of the nanometer size. They are of great importance in medicine to be used as drugs and in food materials as components, which are necessary to save the needed peculiarities. From experimental investigations, it is well known that the particles with nanometer size do not form any structures, but are characterized by the aggregation process, which terminates by the formation of clusters of different sizes. Any external field cannot stop this aggregation process. As a result, the regions with poor and rich concentrations of small particles are observed. The sizes of these regions depend on the properties of particles, their concentration, and the temperature of a liquid crystal [3]. The importance of this effect for liquid crystals was emphasized in [1], where possible cellular structures in different condensed substances were described.

A cellular structure in a system of interacting particles in a liquid crystal dramatically changes the properties of a condensed matter [30, 1], in particular, changes the liquid state of the material to a soft solid [8, 9]. The liquid crystal exists in two phases: the isotropic phase, where we observe a non-orientation order along a long axis of molecules of a liquid crystal, and the ordering phase, where there exists an orientation order along a long axis of molecules. The particles introduced into such isotropic phase of a liquid crystal can cause the orientation ordering at the expense of the formation of a solvent area. The existence of such deformed areas produces the effective interaction [13, 31–33]. Such interaction appears at short distances. In the nematic phase, the orientation ordering occurs at long distances. The deformation elastic field causes the long-range interaction between particles, which are included in a nematic liquid crystal [20, 22, 23, 34]. But, actually, different interactions were revealed at different distances. At short distances, the interactions are induced by the change of the scalar parameter; while, at long distances, the interactions are induced by a deformation in the elas-

tic director field. In the case of low concentration, the interactions existing between two spherical particles was considered in [31–33]. For small nano-sized particles immersed in the liquid crystal phase, we can assume that every particle changes the order parameter. If the number of particles increases in a local area, the variation of the order parameter increases as well. For this process, we cannot take the change of the distribution of the director field into account. The same sort of the induction of the parameter on the surface of a separately introduced particle can occur also in ordinary liquids. For spherical particles, the energy of interaction by changing the scalar order parameter in the isotropic phase was first calculated in [31]. Analytical results of this problem were presented in works [32, 33]. For a case of high concentration of particles introduced into the matter, a more consecutive account of their collective effect is presented in [13, 23]. The interaction can cause a new ordering in a system of introduced particles. The effect of ordering is important for the formation of a new phase in substances.

In what follows, we consider the system of small particles immersed in a liquid crystal. Here, we will explain peculiarities of the behavior of the system of small particles in a liquid crystal and obtain the size characteristic of the formation of a cluster or cellular structure in dependence on the concentration and temperature of media. The distortions of the order parameter can lead to an effective interaction between particle as the screened Coulomb attraction, which can lead to nontrivial consequences. By considering such interaction potential, we can explain the formation of the “cellular” or cluster texture in the system of small particles immersed in a liquid crystal. We start from the elastic bulk free energy density in the well-known form [25],[35]:

$$f_b = \frac{1}{2} A Q_{ij}(\mathbf{r}) Q_{ij}(\mathbf{R}) + \frac{1}{2} L Q_{ij,k}(\mathbf{r}) Q_{ij,k}(\mathbf{r}), \quad (1)$$

where the comma indicates the derivative, and the summation over repeated indices is assumed. Here, A is a positive constant, because we describe the liquid crystal state after a phase transition, and $L > 0$ quantifies the cost of creating a distortion in the nematic phase. For simplicity, we take the one-constant approximation. Every particle locally changes the order parameter, and this fact can be taken into account

through the free energy density [24] in the form

$$f_c = \frac{1}{2} W_{ij} f(\mathbf{r}) Q_{ij}(\mathbf{r}), \quad (2)$$

where W_{ij} is, in general case, the tensorial coefficient, which determines the coupling between particles and the order parameter, and $f(\mathbf{r})$ presents the distribution function of particles in a liquid crystal. The order parameters in the bulk can be written in the well-known form:

$$Q_{ij}(\mathbf{r}) = S(\mathbf{r}) \left\{ n_i \mathbf{n}_j - \frac{1}{3} \delta_{ij} \right\}, \quad (3)$$

where \mathbf{n} is the director, and $S(\mathbf{r})$ describes the order parameter in the liquid crystal bulk. We can take the free energy of many-particle system in the following form:

$$F_b = \frac{L}{2} \int d\mathbf{r} \left\{ Q_{n_j, k}(\mathbf{r}) Q_{ij, k}(\mathbf{r}) + \frac{1}{\xi^2} Q_{ij}(\mathbf{r}) Q_{ij}(\mathbf{r}) + w_{ij} f(\mathbf{r}) Q_{ij}(\mathbf{r}) \right\}, \quad (4)$$

where we introduce the correlation length $\xi^2 = \frac{L}{|A|}$, and $w_{ij} = \frac{W_{ij}}{L}$ represents a normalized coupling constant. This free energy describes the behavior of the liquid crystal with particles. The changes, which separate particles brought in the medium, are developed and create the average field of deformations of the order parameter. This allows us to correctly account for the collective effect of all particles and to find the self-consistent interaction in the case of high concentration. The formal problem is split into two separate problems: to find the distribution of the order parameter and to find the interaction energy with regard to physical and geometrical properties of particles, which are included in the liquid crystal phase. The minimum of the free energy $\frac{\delta}{\delta Q_{ij}(\mathbf{r})} F = 0$ gives the Euler–Lagrange equation:

$$\Delta Q_{ij}(\mathbf{r}) - \frac{1}{\xi^2} Q_{ij}(\mathbf{r}) = w_{ij} f(\mathbf{r}). \quad (5)$$

The solution of this equation can be presented in the form

$$Q_{ij}(\mathbf{r}) = w_{ij} \int d\mathbf{r}' f(\mathbf{r}') G(\mathbf{r}, \mathbf{r}'), \quad (6)$$

where $G(\mathbf{r}, \mathbf{r}')$ is Green's function of the previous equation and takes the well-known form

$$G(\mathbf{r}, \mathbf{r}') = \frac{1}{|\mathbf{r} - \mathbf{r}'|} \exp\left(-\frac{|\mathbf{r} - \mathbf{r}'|}{\xi}\right). \quad (7)$$

This result presents the distribution of the order parameter produced by particles with the distribution function $f(\mathbf{r})$, which are included in the liquid crystal phase. Then we substitute this expression for $Q_{ij}(\mathbf{r})$ again in the formula for the free energy and, thus, obtain the free energy in the form

$$F_b = \frac{L w_{ij}^2}{2 \xi^2} \int d\mathbf{r} \int d\mathbf{r}' f(\mathbf{r}) f(\mathbf{r}') G(\mathbf{r}, \mathbf{r}') + \dots \quad (8)$$

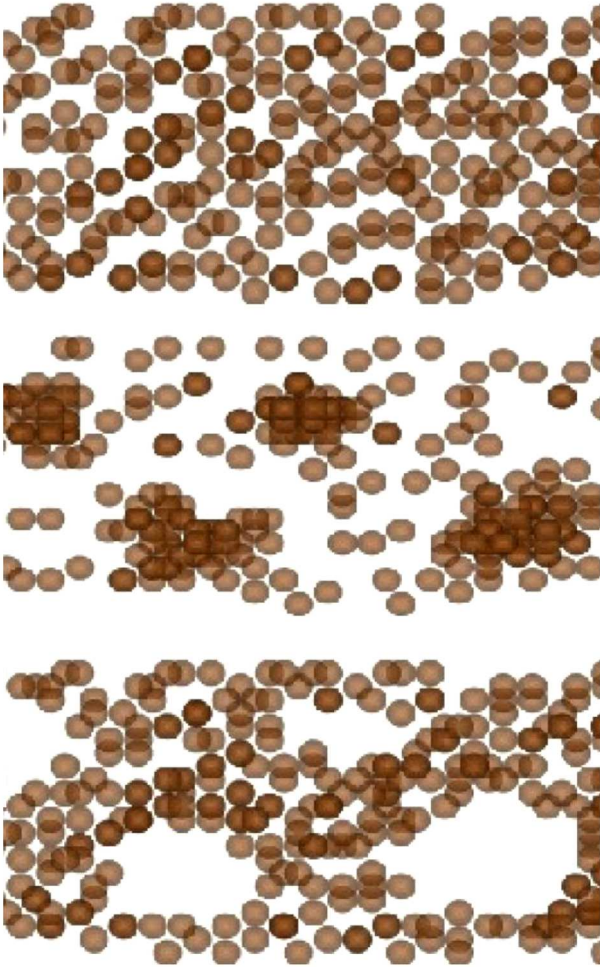
This free energy presents the energy of the interaction between areas with the concentrations of particles $f(\mathbf{r})$ in different spatial points. The immersed particles change the order parameter in dependence on the concentration, and this change of the order parameter produces the effective interaction between them.

Having found the elastic free energy with regard for particles, we can study the thermodynamic behavior of an aggregate of nanoparticles and describe the conditions for the formation of a new structure. The criterion of instability of the homogeneous distribution of particles can be interpreted as a condition for the formation of a spatially non-uniform distribution at a given temperature, which depends on the concentration of particles and on the characteristic length of the new structure. In order to determine this condition, we should add the entropy part to the elastic free energy. The former can be written in the standard form:

$$F_s = kT \int \{ f(\mathbf{r}) \ln f(\mathbf{r}) + [1 - f(\mathbf{r})] \ln [1 - f(\mathbf{r})] \} d\mathbf{r}. \quad (9)$$

The entropy part of the free energy is the reason for the classical particles not to occupy the same spatial positions.

The minimum of the two-part free energy $F_b + F_s$ corresponds to the self-consistent field solution for distribution function $f(\mathbf{r})$. This function corresponds to a solution that describes some thermodynamic state phase of the particle arrangement. If their distribution is inhomogeneous, then the solution serves us to find the stable phase associated with the character of interaction and the temperature. If the solution of particles is disordered, then, by definition, the mean value $f(\mathbf{r}) = c$, where c is the relative particle concentration. The concentration inhomogeneity gives rise to the additional term $f(\mathbf{r}) = c \pm \varphi(\mathbf{r})$, where $\varphi(\mathbf{r})$ is a change of the distribution function of particles. If



Schematic presentation the condition of formation of a cluster or cellular structure in the system of interacting particles [1]. The formation of different structures depends on the concentration and the relation between the interaction energy and the temperature

the concentration inhomogeneities are smooth, and if their scale is much longer than the inter-particle distance, the quantity may be interpreted as a change of the particle composition. We can write the free energy increment associated with the inhomogeneous particle distribution in terms of the power series expansion, by using the long-wavelength expansion of the concentration, i.e., $\varphi(\mathbf{r}') = \varphi(\mathbf{r}) + \rho_i \partial_i \varphi(\mathbf{r}) + \frac{1}{2} \rho_i \rho_j \partial_j \partial_i \varphi(\mathbf{r}) + \dots$, where $\rho = \mathbf{r} - \mathbf{r}'$ is the distance between two different space positions. In this case, we can write the free energy, which is dependent on the change of the distribution function of particles in the

standard form:

$$\Delta F(\varphi) = \frac{1}{2} \int d\mathbf{r} \left\{ l^2 (\nabla \varphi)^2 + \mu^2 \varphi^2 + \frac{1}{4} \lambda \varphi^4 + \epsilon \varphi \right\}, \quad (10)$$

where $\mu^2 \equiv (kT - c(1-c)V)$, $V = \frac{Lw_{ij}^2}{2\xi^2} \int \rho^2 G(\rho) d\rho$, $l^2 = \frac{Lw_{ij}^2}{2\xi^2} \int \rho^4 G(\rho) d\rho$ and $\epsilon = cV$. In our case we can obtain $\mu^2 \approx kT - 2c(1-c)\pi Lw_{ij}^2$ and $l^2 \approx +12\pi Lw_{ij}^2 \xi^2$. For the lowest temperature such as $kT_c = 2c(1-c)\pi Lw_{ij}^2$, this system is always unstable, and the length of the first instability is

$$\lambda = \sqrt{l^2/\mu^2} \sim 2\xi \quad (11)$$

for $\frac{kT}{c(1-c)} < 2\pi Lw_{ij}^2$, and ξ is the correlation length. In the reverse condition, we can observe the smallest length of inhomogeneous distribution of particles. After the formation of areas with different concentrations of particles, the Coulomb-like attraction appears between particles on the separate distance of few nanometers, which produces the strongly interacting cluster of particles [23, 13, 31–33]. In the liquid crystal phase, the correlation length is $\xi \sim 1 \mu\text{m}$, and it is the average size of a cluster of segregated nanoparticles.

To determine the size of a possible cluster in the present case, we can use the approach in [1]. As is easy to see, the expression for the free energy is a well-known functional, which describes the first-order phase transition with the accompanied formation of a cluster in the system of interacting particles. Thus, the minimum of the functional realizes a spatially inhomogeneous distribution of nanoparticles only provided the sign and the values of coefficients determined by the interparticle interaction satisfy some relations. The most important contribution to the concentration is associated with the concentration configuration, for which the value of the free energy is minimum, i.e.: $\Delta \varphi - \frac{dV}{d\varphi} = 0$, where $\Phi = -\frac{1}{2} \mu^2 \varphi^2 + \frac{1}{4} \lambda \varphi^4 - \epsilon \varphi$ is the potential energy. The substitution of this relation in the expression for the free energy yields its variation due to the formation of the new phase. In the case where the difference of minimum effective potential values is greater than the barrier height, the free energy in our case of one cluster is described by the expression $\Delta F = -\frac{4\pi}{3} r^3 \epsilon + 4\pi r^2 \sigma$, where σ is the surface energy of the cluster boundary, i.e. $\sigma = \int_0^\infty d\varphi \sqrt{2\Phi(\varphi)}$. The radius of the new-phase cluster is determined by the free energy

minimum. It is given by $\tilde{R}_0 = \frac{2\sigma}{\epsilon} = \frac{\mu^2 l}{3\lambda^{1/2}\epsilon}$, and the effective value of the free energy variation due to the cluster formation is given by $\Delta F = \frac{8\pi\sigma R^2}{3}$. The probability of the formation of one cluster can be written in the form: $P(\tilde{R}) = \exp(-\frac{\Delta F}{kT}) = \exp(-\frac{8\pi\sigma R^2}{3kT})$. The criterion of instability given by this relation can be interpreted as a condition for the formation of a spatially non-uniform distribution at a given temperature, which depends on the concentration of particles and on the characteristic length of the new structure. It is possible to describe the formation of voids in the system of interacting particles.

In our system, the condition for the formation of the cluster with minimal size is realized. If we have some fixed concentration of particles in the system, the process of aggregation finishes, when all particles are assembled in a few clusters. The size of the cluster will grow until all particles will be located only in the cluster. The cluster is formed by the interaction energy, and the distribution of particles in one cluster can consider homogeneous. If the concentration increases, the cluster volume grows too, and the moment will come, when the particles form one cluster which occupies some position in the volume of all particles. Another part of the volume will be formed by voids.

If the concentration is small, we will observe only a cluster with equilibrium size. But if the concentration increases to $c > \frac{1}{2}$, we will obtain one cluster and few voids. In the present approach, we can estimate the size of voids as the characteristic length of instability of the nonuniform distribution of particles. The size of a cell depends on the concentration of particles, as the above-presented formula describes. In summary, we have independently estimated the spontaneous formation of loosely bound ordered aggregates of colloidal particles and the possible formation of cellular structures, as a result of the interactions of different nature. The phase separation in the colloidal fluids is, thus, directly connected with the percolation transition of the wetting solvent phase. The solvent-induced phase separation is driven energetically. The cluster formation in a system of nanoparticles is a natural result of the elastic interaction of the area with different concentrations of particles. The particles produce a deformation of the elastic substance, which leads to the aggregation of small particles. The equilibrium distribution of particles corresponds to their spatially non-uniform distribution. This spatial

non-uniform distribution of the introduced particles causes the occurrence of areas that are free of particles [1]. The mutual effect of particles and the medium results in a non-uniform distribution of cooperating particles. Thus, some kind of a new soft body, whose properties differ from properties of the medium, is formed. It is possible to create a wide variety of cellular structures, if the forces of interaction possess some anisotropy. Another important property of such systems is that they are highly sensitive to weak external effects. This enables us to exert a specific effect under conditions of the formation of the structures and their transmutations.

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АГРЕГАЦІЯ НАНОЧАСТИНОК
У РІДКОМУ КРИСТАЛІ

Резюме

Стаття являє собою короткий огляд загального результату, отриманого в роботі [1], що стосується поведінки сукупності наночастинок у нематичному рідкому кристалі. Такі рідкокристалічні колоїди досліджуються протягом довгого часу і становлять непересічний інтерес для широкого практичного застосування. Приймаючи до уваги колективний ефект взаємодії між малими частинками через зміну скалярного параметра порядку в нематичному рідкому кристалі, передбачено неоднорідний розподіл частинок на відміну від можливих структур у системі макроскопічних частинок у рідкому кристалі. Це – основний природний розподіл для системи наночастинок. Визначено довжину неоднорідності та розміри кластера з наночастинок, а також описано можливі особливості в поведінці рідких кристалів з внесеними наночастинками.