

Transitions in Lyotropic Liquid Crystals Caused by Concentration Change and γ -Radiation

D. A. Badalyan, M. A. Stepanyan, and H. G. Badalyan*

Yerevan State University, Yerevan, Armenia

**hbadal@ysu.am*

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Abstract—A molecular-statistical theory of phase transitions in lyotropic liquid crystals, which describes the phase transitions between the isotropic (micellar), nematic and lamellar phases was developed. The equations describing the dependence of parameters of orientation and translational long-range order on the concentration were obtained. It was shown that depending on the values of the model microscopic constants, the nematic phase–lamellar phase transition can be both of the first and the second order. The influence of intensive and low intense γ -radiation on the phase transitions mentioned herein was considered. It was shown that the irradiation changes the model constants responsible for the phase transitions. On this basis, it can be assumed that the γ -radiation influences the course of the dependence of the long-range order parameters on concentration as well as it changes the values of the critical concentrations of the phase transitions and even the phase transition order.

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1. INTRODUCTION

As is known, the lyotropic liquid crystals (LLC) are obtained by the dissolving of amphiphile substances in polar solvents. Most often, these are the aqueous solutions of substances which have a rigid structure of molecules (the DNA, some synthetic polypeptides, etc.). The LLC can be formed also by the cylindrical micelles – the aggregates of amphiphilic molecules dissolved in water [1–3]. Depending on the solute concentration, the orientational and/or translational order appears in these systems, which is believed to be mainly a result of the interaction of the rod-like particles with each other, while their interaction with the solvent plays a secondary role [4].

The mechanisms of phase transitions and the thermodynamic characteristics of the LLC, in contrast to the conventional thermotropic liquid crystal (TLC), are studied very poorly. Therefore, the study of the thermodynamic properties of the LLC based on the molecular-statistical models is important. The aim of this work is the microscopic study of phase transitions in the LLC, formed from the rod-like particles (hereinafter – rods). The corresponding theory is an expansion of the previously developed method to describe the orientational and long-range translational order in the TLC and in organic solid solutions [5, 6].

2. FREE ENERGY OF THE LLC

For further development of the theory it is necessary to know the free energy F , which depends on the orientational and translational parameters of the long-range order. We obtain the expression for F , using a generalized model of a lattice gas. Consider an idealized system of rigid rods in a structureless inert

solvent. The centers of gravity of the rods and solvent molecules are in the lattice sites of some spatial lattice with the small size of the period. The rods in each site can have an arbitrary orientation described by the unit vectors

$$\mathbf{m} = (\sin \theta_m \cos \varphi_m, \sin \theta_m \sin \varphi_m, \cos \theta_m),$$

where θ and φ are the spherical coordinates. Pairwise energies of interaction of rods located in the lattice sites \mathbf{r} and \mathbf{r}' , and having the orientations \mathbf{m} and \mathbf{m}' are determined by the potentials $V_{\mathbf{m},\mathbf{m}'}(\mathbf{r}-\mathbf{r}')$. It is assumed that the orientations of \mathbf{m} and \mathbf{m}' are equivalent.

Assume that the orientation of particles is discrete and their number is equal to ν . The $\nu+1$ different states can arise in an arbitrary lattice site \mathbf{r} , since the lattice site may be occupied by the molecule of solvent, as well as by the rod with one of the orientations. We introduce the random functions $c_\alpha(\mathbf{r})$ ($\alpha=0,1,\dots,\nu$) equal to one, if \mathbf{r} lattice site is in a state α and is zero otherwise (the index $\alpha=0$ characterizes such a state, when the lattice site is occupied by the molecule of solvent). Functions $c_\alpha(\mathbf{r})$ are subject to the normalization conditions

$$\sum_{\alpha=1}^{\nu} c_\alpha(\mathbf{r}) = 1, \quad (2.1)$$

implying that an arbitrary lattice site \mathbf{r} is occupied either by the rod or by the molecule of solvent.

Additionally, we have

$$\sum_{\mathbf{r}} c_0(\mathbf{r}) = N_{\text{solv}} = N(1-c), \quad (2.2)$$

where N_{solv} is the number of molecules of solvent, N is the total number of lattice sites, and c is the concentration of rods. It is necessary to pay attention to the LLC model of a multicomponent solid solution: if in the LLC the each lattice site is occupied as by a solvent molecule, as by the rod with various orientations ν , in the $(\nu+1)$ -component solid solution of substitution, in each lattice site can also arise $\nu+1$ various states which depend on the type of an atom which falls into the lattice site. Therefore, the general results obtained in [6], can be applied to the model with the discrete orientations of particles. If we consider that the orientations in the LLC change continuously, the summation over the discrete orientations should be replaced by integration. Then the configurational free energy of the LLC in the self-consistent field approximation takes the form

$$F = \frac{1}{2} \sum_{\mathbf{r},\mathbf{r}'} \int V_{\mathbf{m},\mathbf{m}'}(\mathbf{r}-\mathbf{r}') \rho_{\mathbf{m}}(\mathbf{r}) \rho_{\mathbf{m}'}(\mathbf{r}') d\Omega_{\mathbf{m}} d\Omega_{\mathbf{m}'}, \quad (2.3)$$

$$+ kT \sum_{\mathbf{r}} (1-n(\mathbf{r})) \ln(1-n(\mathbf{r})) + kT \sum_{\mathbf{r}} \int \rho_{\mathbf{m}}(\mathbf{r}) \ln \rho_{\mathbf{m}}(\mathbf{r}) d\Omega_{\mathbf{m}},$$

where $n(\mathbf{r}) = 1 - \langle c_0(\mathbf{r}) \rangle$ is the probability to find the rod in the lattice site \mathbf{r} , $\rho_{\mathbf{m}}(\mathbf{r}) d\Omega_{\mathbf{m}} = \langle c(\mathbf{r}) \rangle d\Omega_{\mathbf{m}}$ is the simultaneous probability to find the rod in the lattice site \mathbf{r} in a small solid angle $d\Omega_{\mathbf{m}} = \sin \theta_m d\theta_m d\varphi_m$ around the orientation \mathbf{m} , T is the absolute temperature, k is the Boltzmann constant, $\langle \dots \rangle$ means the averaging over the ensemble of non-interacting particles with additional conditions (compare (2.1) with (2.2)) and

$$\int \rho_{\mathbf{m}}(\mathbf{r}) d\Omega_{\mathbf{m}} = n(\mathbf{r}); \quad \sum_{\mathbf{r}} n(\mathbf{r}) = c. \quad (2.4)$$

In the future, we will neglect the weak correlation between the orientations and the positions of the centers of gravity of rods. This assumption leads to the approximate equality

$$\rho_{\mathbf{m}}(\mathbf{r}) = n(\mathbf{r})\rho_{\mathbf{m}}^0, \quad (2.5)$$

where $\rho_{\mathbf{m}}^0$ is the probability of orientation of the rod in a small solid angle $d\Omega_{\mathbf{m}}$ around the orientation \mathbf{m} provided that the lattice site \mathbf{r} is occupied by the rod.

For further calculation, assume that the potential $V_{\mathbf{m}\mathbf{m}'}(\mathbf{r} - \mathbf{r}')$ is described by the function

$$V_{\mathbf{m}\mathbf{m}'}(\mathbf{r} - \mathbf{r}') = \tilde{U}(\mathbf{r} - \mathbf{r}') - \tilde{\Phi}(\mathbf{r} - \mathbf{r}')P_2(\cos\gamma_{\mathbf{m}\mathbf{m}'}), \quad (2.6)$$

where the first term describes the central forces of electrostatic repulsion, and the second the orientational (anisotropic and dispersion) forces of attraction, $P_2(\cos\gamma_{\mathbf{m}\mathbf{m}'})$ is the Legendre polynomial of the second order and $\gamma_{\mathbf{m}\mathbf{m}'}$ is the angle between \mathbf{m} and \mathbf{m}' orientations.

Substituting (2.5) and (2.6) in view of (2.4) to (2.3), we obtain the functional of free energy which contains two independent functions of distribution $\rho_{\mathbf{m}}^0$ and $n(\mathbf{r})$. The function $\rho_{\mathbf{m}}^0$ for the uniaxial LLC can always be represented as an expansion in terms of the Legendre polynomials

$$\rho_{\mathbf{m}}^0 = \frac{1}{4\pi} + \sum_{k=1}^{\infty} \frac{4k+1}{4\pi} P_{2k}(\cos\theta_{\mathbf{m}}) S_{2k}, \quad (2.7)$$

where S_{2k} are the expansion coefficients, which are playing the role of the orientational parameter of the long-range order.

For the layer structures the function $n(\mathbf{r})$ has the form

$$n(\mathbf{r}) = c + \frac{\eta}{2} e^{i\mathbf{k}_0 \mathbf{r}}, \quad (2.8)$$

where η is a parameter of translational long-range order and $2\mathbf{k}_0 = 2\pi G$ (G is the reciprocal lattice vector of the lattice gas).

Thus, from the formulas (2.3)–(2.8) we obtain

$$F = N \left\{ \frac{1}{8} U(\mathbf{k}_0) \eta^2 - \frac{1}{2} \Phi(0) S_2^2 - \frac{1}{8} \Phi(\mathbf{k}_0) \eta^2 S_2^2 + \frac{kT}{2} \left[\left(c + \frac{\eta}{2} \right) \ln \left(c + \frac{\eta}{2} \right) + \left(c - \frac{\eta}{2} \right) \ln \left(c - \frac{\eta}{2} \right) \right. \right. \\ \left. \left. + \left(1 - c + \frac{\eta}{2} \right) \ln \left(1 - c + \frac{\eta}{2} \right) + \left(1 - c - \frac{\eta}{2} \right) \ln \left(1 - c - \frac{\eta}{2} \right) \right] + kTc \int \rho_{\mathbf{m}}^0 \ln \rho_{\mathbf{m}}^0 d\Omega_{\mathbf{m}} \right\}, \quad (2.9)$$

where

$$\Phi(0) = \sum_{\mathbf{r}} \tilde{\Phi}(\mathbf{r}), \quad \Phi(\mathbf{k}_0) = \sum_{\mathbf{r}} \tilde{\Phi}(\mathbf{r}) e^{i\mathbf{k}_0 \mathbf{r}}, \quad U(\mathbf{k}_0) = \sum_{\mathbf{r}} \tilde{U}(\mathbf{r}) e^{i\mathbf{k}_0 \mathbf{r}}.$$

3. EQUATIONS FOR PARAMETERS OF LONG-RANGE ORIENTATIONAL AND TRANSLATIONAL ORDER

Let us find the equations for parameters η and S_{2k} from the conditions of minimum of the free energy

$$\frac{\partial F}{\partial \eta} = 0, \quad \frac{\partial F}{\partial S_{2k}} = 0 \quad (k = 1, 2, 3, \dots). \quad (3.1)$$

Substituting (2.9) to (3.1), we obtain a closed system of equations for parameters η and $S_{2k} \equiv S$:

$$\ln \frac{\left(1-c+\frac{\eta}{2}\right)\left(c+\frac{\eta}{2}\right)}{\left(1-c-\frac{\eta}{2}\right)\left(c-\frac{\eta}{2}\right)} = \frac{gS^2-b}{\tau} \eta, \quad (3.2)$$

$$S = \frac{3}{4m^2} \left(\frac{e^{m^2}}{D(m)} - 1 \right) - \frac{1}{2}, \quad (3.3)$$

where $g = \frac{\Phi(\mathbf{k}_0)}{\Phi(0)}$, $b = \frac{U(\mathbf{k}_0)}{\Phi(0)}$ and $\tau = \frac{kT}{\Phi(0)}$ is the dimensionless, $D(m)$ is the Dawson integral [7]:

$D(m) = \int_0^m e^{-x^2} dx$, $m^2 = \frac{3S}{2\tau} \left(1 + \frac{g}{4c} \eta^2 \right)$. Other parameters S_{2k} ($k < 1$) are expressed in terms of the parameter S .

For given τ , b and g , the system of equations (3.2) and (3.3) gives the dependence of the parameters of ordering S and η on the concentration C . For dilute solutions ($c \ll 1$), this system has the trivial solution describing the isotropic (micellar) solution $S = \eta = 0$. With the increasing concentration of the solid substance, the nontrivial solutions $\eta = 0$, $S \neq 0$ and $\eta \neq 0$, $S = 0$ can appear, which describe a lyotropic nematic and lamellar phase.

Concentrations that lead to the loss of stability of the nematic and isotropic phases are determined by the linearized equations (3.2) and (3.3), as a result of which one obtains

$$\frac{1}{c_L^*(1-c_L^*)} = \frac{gS_L^{*2}-b}{\tau}, \quad c_N^* = 5\tau, \quad (3.4)$$

where c_L^* , c_N^* are the concentrations of losses of stabilities of nematic and isotropic phases, S_L^* is the value of parameter S at the point c_L^* that coincides with the critical concentration c^0 of the phase transition of the second-order, if it takes place, and is slightly larger than c^0 if there is the phase transition of the first order.

In general, one can simplify the equations (3.2) and (3.3); one needs to take into account that for the lamellar phase the orientational long-range order is heavily saturated ($s \geq 0.8$), therefore $m^2 \gg 1$. Then, using the expansion of the function $D(m)$ at large m , $D(m) \approx \frac{e^{m^2}}{2m^2} \left(1 + \frac{1}{2m^2} \right)$, we obtain an approximate expression for the parameter of order S :

$$S = \frac{1}{2} \left(1 + \sqrt{1 - \frac{4\tau}{a}} \right), \quad (3.5)$$

where $a = c + \frac{g}{4c} \eta^2$. Excluding the parameter S from the formula (3.2), and with the use of (3.5) we obtain the equation for the parameter η . If we know η , we find the value of S from the formula (3.5).

In the lamellar phase and limiting ourselves by the region of state with the small values of parameter η , let us expand the left and right sides of the equation (3.2) by the powers of η . In the expansion, retaining only the first few terms, including the terms $\sim \eta^4$, we obtain the equation

$$Z_1 + Z_2 \eta^2 + Z_3 \eta^4 = 0, \quad (3.6)$$

$$\begin{aligned}
Z_1 &= \frac{1}{c(1-c)} - g \left(\frac{1+R}{2\tau} - \frac{1}{c} \right) + \frac{b}{\tau}, \\
Z_2 &= \frac{1}{12} \left(\frac{1}{c^3} + \frac{1}{(1-c)^3} \right) - \frac{g^2}{4c^3} \frac{1+R}{R}, \\
Z_3 &= \frac{1}{80} \left(\frac{1}{c^5} + \frac{1}{(1-c)^5} \right) + \frac{g^3}{16c^5} \frac{1+R}{R}, \\
R &= \sqrt{1 - \frac{4\tau}{c}}.
\end{aligned} \tag{3.7}$$

If the transition from the nematic phase to the lamellar phase may occur by the mechanism of transition of the second order, then at the point of the loss of stability of initial phase we have $\eta = 0$. The concentration of the loss of stability c_L^* , according to the formula (3.6), is determined by the condition

$$Z_1(g, b, \tau, c_L^*) = 0. \tag{3.8}$$

At the first-order phase transition the parameters η and S are changing very rapidly. The concentration of the transition c_L^0 , as well as the values of jumps of the order parameters are determined by the condition of equality of free energies $F(0, S_L^0)$ at $\eta = 0$ and $S = S_L^0$ in the nematic phase and $F(\eta_L^0, \tilde{S}_L^0)$, corresponding to the non-vanishing value of parameter η_L^0 in the lamellar phase. Also, the equations are used for η (3.6) and for S which follow from the formula (3.5) in view of the smallness of the parameter η in the studied region. The last equation has the form

$$S = \frac{1}{2} \left\{ 1 + R \left[1 + \frac{g\tau\eta^2}{2R^2c^3} \left(1 - \frac{g^2\eta^2}{4c^2} \right) \right] \right\}. \tag{3.9}$$

For the nematic phase, the formula (3.9) takes the simple form

$$S = (1 + R)/2. \tag{3.10}$$

At the critical concentration $c_N^0 = 4\tau$ this formula leads to the phase transition of the first order: the isotropic liquid–the lyotropic nematic. The jump of parameter S at this point is equal to $S_N^0 = 0.5$. A more accurate consideration, based on the formula (3.3) results in $c_N^0 = 4.54\tau$; the value $S_N^0 = 0.43$ is known from the TLC [4]. The comparison shows that even at the point of transition into the nematic phase, the approximation (3.9) leads to an error not exceeding 12–16%.

4. DEPENDENCE OF PARAMETERS η AND S ON THE CONCENTRATION

Let us investigate the dependence of long-range order parameters η and S on the concentration at different values g , b and τ . The roots of the biquadratic equation (3.6) can be obtained from the formula

$$\sigma_{\pm} = \frac{2C^2(-\tilde{Z}_2 \pm \sqrt{D})}{\tilde{Z}_3}. \tag{4.1}$$

Here $\sigma = \eta^2$, $D = \tilde{Z}_2^2 - 4\tilde{Z}_1\tilde{Z}_3$, \tilde{Z}_i are the coefficients, which are related with the previous coefficients (3.7) by the following relations:

$$\tilde{Z}_1 = cZ_1, \quad \tilde{Z}_2 = 4c^3Z_2, \quad \tilde{Z}_3 = 16c^5Z_3. \tag{4.2}$$

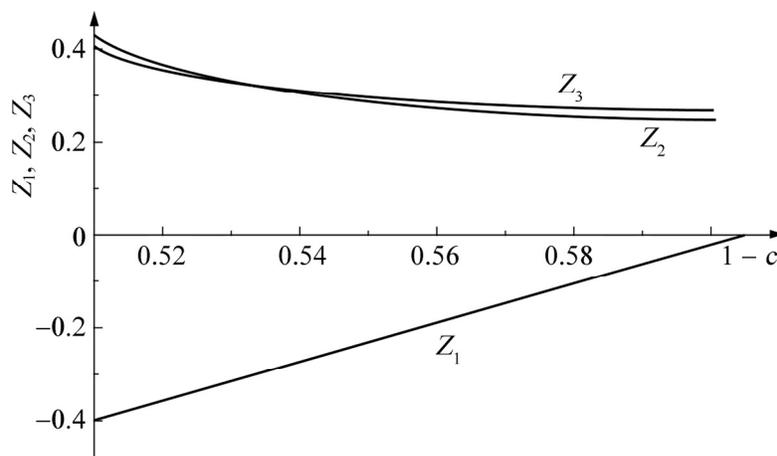


Fig. 1. Dependences of coefficients Z_1 , Z_2 and Z_3 on the concentration of solvent ($1 - c$) and ($b = 0.18$, $g = 0.3$ and $\tau = 0.02$).

Due to the requirements $\sigma > 0$, the calculation of the roots of equation (3.6) depends strongly on the sign of the coefficients \tilde{Z}_i . In this regard, it should be taken $g > 0$ in the formulas (3.7) (in the opposite case, the occurrence of the lamellar phase is too difficult). This assumption implies that $\tilde{Z}_3 > 0$ and the coefficients \tilde{Z}_1 and \tilde{Z}_2 can be both positive and negative. In general case, the following variants are possible

$$1 \begin{cases} \tilde{Z}_1 \geq 0 \\ \tilde{Z}_2 \geq 0 \end{cases}, \quad 2 \begin{cases} \tilde{Z}_1 \leq 0 \\ \tilde{Z}_2 \geq 0 \end{cases}, \quad 3 \begin{cases} \tilde{Z}_1 \geq 0 \\ \tilde{Z}_2 \leq 0 \end{cases}, \quad 4 \begin{cases} \tilde{Z}_1 \leq 0 \\ \tilde{Z}_2 \leq 0 \end{cases}. \quad (4.3)$$

The analysis shows that due to the requirements $\sigma > 0$ and $D \geq 0$ the variant 1 is excluded, as in this case there are the negative roots. In variants 2 and 4, $\sqrt{D} > |\tilde{Z}_2|$ and there is one positive root σ_+ . In the case of variant 3, $\sqrt{D} < |\tilde{Z}_2|$ and both roots (σ_+, σ_-) of equality (4.1) are positive.

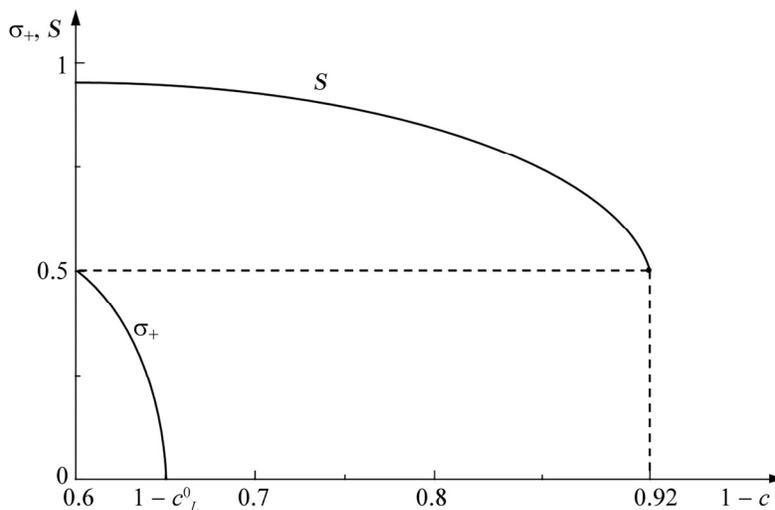


Fig. 2. Dependences of parameters σ_+ and S on the concentration of solvent ($1 - c$) and ($b = 0.18$, $g = 0.3$ and $\tau = 0.02$).

So, before the obtaining the curves of dependence of long-range order parameters on the concentration, it is necessary to investigate the concentration dependence of the coefficients \tilde{Z}_l . Fig. 1 shows the dependences \tilde{Z}_l at the constants $g = 0.30$, $b = 0.18$ and $\tau = 0.02$. It follows from this figure that this case corresponds to the second variant of (4.3), that is, the equation (4.1) has one positive root. Due to the intersection of dependence \tilde{Z}_1 with the axis $1 - c$ one can conclude that in this case the transition to the lamellar phase is possible, with the point of stability loss of the nematic phase $c_L^* = 0.365$.

Figure 2 shows the concentration dependences of the parameters S and σ_+ corresponding to formulas (3.9) and (4.1) with the same constants. It follows from this figure that the parameter σ_+ is decreasing continuously with the increase of $1 - c$ and vanishes at the concentration c_L^* . Consequently, there is a phase transition of the second order.

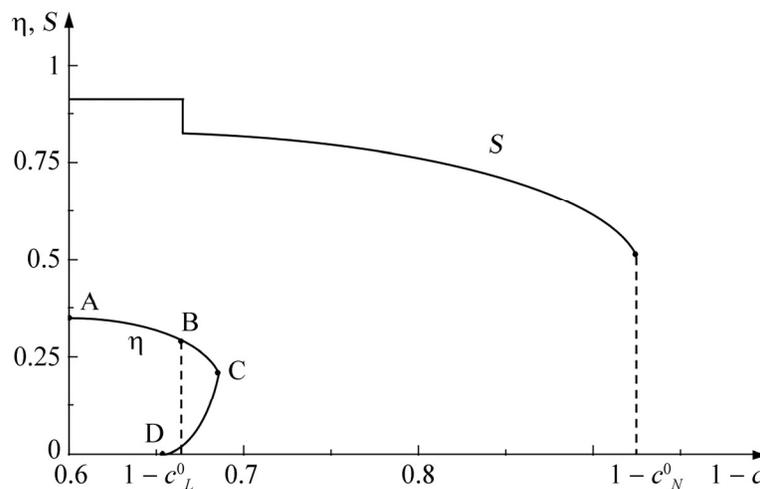


Fig. 3. Dependences of parameters η and S on the concentration of solvent ($1 - c$) and ($b = 0.304$, $g = 0.45$ and $\tau = 0.02$).

Consider another case: for constant τ let us increase the interaction constants g and b ($g = 0.45$, $b = 0.304$ and $\tau = 0.02$). The analysis of the curves \tilde{Z}_l shows that here is the variant 3 which takes place (the equation (4.3)), that is, both roots of (4.1) are positive. The dependences of functions η and S on $(1 - c)$ are shown in Fig. 3. This figure shows that in the interval between the concentrations $1 - c_L^*$ and $1 - \tilde{c}$ ($1 - \tilde{c}$ is the inflection point of the curve η) two values of parameter η correspond to the each point $1 - c$. As is well known, this situation is typical for the phase transitions of first order in the ordered alloys [8]. To find the solution corresponding to the equilibrium value of the long-range order parameter at the given concentration of the LLC is necessary to study the dependence of the free energy as on the long-range order parameter, and at the temperature phase transition of first order. Such a research shows that the plot AB corresponds to the minimum of free energy. Plot BC corresponds to metastable states. The values of η in the range CD correspond to the maximum of F , and these states are not realizable. As already mentioned, the concentration of the phase transition of the first-order is determined by the equality of free energies at $\eta = 0$ and $\eta = \eta_L^0$. The computations result in: $c_L^0 \approx 0.333$ and $\eta_L^0 \approx 0.257$.

5. EFFECT OF γ -RADIATION ON THE PHASE TRANSITIONS IN THE LLC

Let us discuss here the influence of an external γ -radiation on the phase transitions observed in the LLC, as well as let us consider the application of the LLC model developed here to the given problem. This problem is very important since the body fluids (cytoplasm, blood plasma, lymph etc.) are of the LLC type that at the 37°C are close to the phase transition point, so, they can respond even to the weak external signals, such as the laser radiation of low intensity used in medicine [9]. It is considered that one of the mechanisms of action of the weak laser radiation is the change of the physical and chemical characteristics of the solvent as a part of the LLC. It is to be considered that due to the low intensity of light (a few mW/cm²) thermal effects are negligible. Given that the electrostatic repulsion of the ions in the electrolyte is described by the Debye–Hückel potential, which depends strongly on the dielectric constant of the solvent; in the first place it can be assumed that the weak radiation changes the dielectric constant. From the viewpoint of the theory developed here, this variation affects the constant b .

At the high concentration of the amphiphilic substance, the influence of γ -radiation on the LLC by the γ -quants leads to two types of the phase transitions: the transition from lamellar phase to the crystalline (the α -phase) and the transition from the phase with the ordered tails (the crystalline state) to the phase with the disordered tails, which is called the ‘melting’ of tails [10–13]. At low concentrations of the amphiphilic substance there is the phase transition the nematic phase–lamellar phase. As a result of the radiation the shifting of the transition point takes place towards the small values of concentration of the amphiphilic substance.

The results of influence of γ -radiation on the LLC depends on the external parameters and the state of the LLC. As is known, the ionizing nature of γ -rays results in the change of a charge both lamellas and micelles, as well as of the solvent. In the first case there is a direct influence of γ -rays, while in the second case the influence is mediated. In the direct influence the increase of the ion charge leads to the increased electrostatic repulsion forces (constant b). Simultaneously, the peroxidation of tails of the molecules takes place, which reduces the portion of the van der Waals attractive forces (the constant g). As a result of an imbalance of these forces, the repulsion forces exceed the attractive forces, leading to the swelling of the lamellas. This, in its turn, leads to a loosening of the lamellas. As a result of irradiation, there is also the release of the thermal energy, which leads to an increase in temperature of the system (the constant τ) and to the ‘melting’ of the heating region

6. CONCLUSION

It follows from the foregoing that the theory of phase transitions in the LLC is interpreted by transition from the isotropic (micellar) into the lyotropic nematic phase, as well as by the transition between the nematic and lamellar phases. The equations are obtained, which describe the dependences of the parameters of orientational and translational long-range order on the concentration. It is shown that depending on the model constants, the phase transition from the nematic phase to the lamellar can be both of the first and the second order.

The influence of γ -radiation on the LLC leads to the change of the all constants of the model responsible for the equilibrium thermodynamic properties of the crystal. This means that the radiation changes the dependences of long-range order parameters on the concentration, influencing the critical concentrations of phase transitions, as well as the order of phase transition.

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