INSTABILITY OF THE NEMATIC-LIKE PHASES FILLED WITH IMPURITIES AGAINST THE FORMATION OF MODULATED STRUCTURES

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We examine spatial distribution of impurity rigid-sphere-like macroparticles in the mesomorphic liquid crystal host. Using continuum statistical mechanical theories, we analyze the thermodynamic conditions necessary for a modulated lamellar-structure to appear. There is a long-range effective interaction between the impurity particles. This interaction is considered as being responsible for the formation of superstructures. In the general case, this interaction includes two components: a van der Waals-type direct interaction and an indirect interaction (through the director-field distortions). The last one depends on both the temperature of a sample and the concentration of particles. This effective interaction controls the structure and properties of the system. Analytical solutions for a director-field distortion, density inhomogeneity of the host medium, temperature of the formation of a modulated structure, and its spatial period are obtained. The proposed theoretical approach can be applied to other anisotropic and inhomogeneous systems.

1. Introduction

Nematic liquid crystal (LC) colloids have attracted a lot of attention in recent years [1–5]. These materials have all properties of the LC phase but a non-trivial behavior of the impurity subsystem is possible as well. For instance, the modulated structures, which would have great influence on the physical properties of entire system, can be formed. Macromolecules or other particles with relatively large molecular mass and diameter of about 10-1000 nm (that is much larger than these parameters for typical nematic molecules) can act as impurities. When these impurities are introduced into the nematic host crystal, a distortion of its structure occurs, because of the presence of 'tight' anchoring between LC molecules and the impurity surface. The director field distortions can occupy a much bigger volume than that per one impurity particle. If the areas of deviations induced by different impurities are overlapping, each of those particles will react upon the director deviation. In other words, this means that the effective interaction between impurity particles through the nematic host crystal will take place. The indirect interaction energy can be much larger than the energy of the direct van der Waals interaction. All of these effects result in the key relevant property of such a system, namely, in the formation of modulated structures with certain spatial period. Zumer with collaborators carried out the experiments and simulations of the formation of superstructures in LC (see, e.g., [5]). It has been known long ago that an interaction induced by a director deformation plays the crucial role in systems based on a filled liquid crystal [6–9]. Moreover, the main problem of a filled LC focuses certainly on the system stability which will be considered in the present paper.

There are plenty of applications of the systems based on filled liquid crystals. Such applications as diffraction gratings, different magnetooptical switches, as well as information processors and even medical devices [1, 10, 11] are only few of examples.

In this paper, a theoretical description of the instability of a homogeneous system associated with the formation of modulated structures is presented in the manner of Ref. [12].

2. Statement of the Problem

Let us consider a highly dispersed system that occupies a volume V and consists of two interacting subsystems – nematic host and impurity sphere-like particles.

The behavior of the system depends of the phase state of the liquid crystal host. Let us assume that the sample is isotropic above a certain temperature T_1. The corresponding global space-symmetry group is $S_1 \equiv R^3 \wedge O(3)$.

It may be presumed that, below T_1, the nematic phase of the host medium takes place and is characterized by the existence of the axis of a preferred orientation of molecules and a uniform impurity distribution. In contrast to isotropic liquids in the mesophase, a long-range correlation between orientations of elongated molecules can exist. The symmetry group $S_N \equiv R^3 \wedge D_{\infty h}$ responds to this state [13] ($D_{\infty h}$ – cylinder group with the axis of rotation C_∞ which is directed along the axis of a preferred orientation of liquid crystal molecules).

On the further cooling of the sample below a certain temperature T_{0c} ($T_1 \geq T_{0c} \geq T_M$, where T_M is the melting point temperature of the liquid crystal), the uniform distribution of interacting impurities can disappear, and the so-called focal conic texture can be formed due to the appearance of impurity layers of the same thickness which can quasifreely glide over one another. The host medium molecules have kept the direction perpendicular to the impurity layers, on the average. The fact that layers glide over one another indicates the absence of a long-range translation order in their plane. Such a phase is somewhat similar to the phase called smectic A. The smectic A phase differs from the nematic one by breaking the translation symmetry along the direction that is perpendicular to the layer plane. Thus, the space symmetry group corresponding to this state is $S_A \equiv \left(R^2 \times Z\right) \wedge D_{\infty h}$, where Z is the group of discrete translations along the axis of the preferential orientation of host medium molecules, the Oz direction for definiteness, and R^2 is a group of two-dimensional continuous translations. Thus, the phase transition that is accompanied by the appearance of (modulated) periodic structures is similar, in respect to symmetry, to the phase transition between nematic and smectic A.

We will characterize the bulk density (the number of molecules per unit volume) of the system with the function of spatial distribution $\rho(\mathbf{r})$ through "lattice sites" $\{\mathbf{r}\}$ of the quasilattice. Moreover, $\rho(\mathbf{r}) \ll 1/V_m$, where V_m is the natural volume of one host medium molecule. The mean position-independent density of the host medium can be determined with the expression $\rho_0 \equiv V^{-1} \int\limits_V \rho(\mathbf{r}) d\mathbf{r}$; $\rho_0 \ll 1/V_m$.

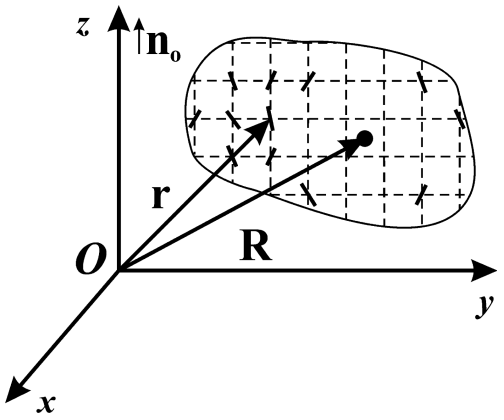


Fig. 1. Geometry of the problem

To describe a spatial distribution of impurities embedded into a host medium, the quasi-lattice-gas representation can be used. Impurity particle centers are located in "interstices" which are distributed randomly throughout the lattice $\{\mathbf{R}\}$ which is embedded in the host medium lattice $\{\mathbf{r}\}$. The fraction c_0 of occupied sites $\{\mathbf{R}\}$ is determined by the relative concentration of impurity particles: $c_0 \equiv N^{-1} \sum\limits_{\mathbf{R} \in V} C(\mathbf{R})$ $c_0 \ll 1$. Here, $C(\mathbf{R})$ is a stochastic function being equal to 1, if the "lattice site" with radius vector $\mathbf{R}$ is occupied by an impurity particle (from the system $\{\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, ...\}$) and is equal to 0 otherwise: $C(\mathbf{R}) = \left\{ {}^{1\,\mathbf{R}=(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, ...),}_{0\,\mathbf{R}\neq(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, ...);} \right.$ $c(\mathbf{R})$ is the one-particle impurity distribution function which is determined by the formula $c(\mathbf{R}) = \langle C(\mathbf{R}) \rangle$ [14], where the averaging is realized over the ensemble of impurity particles under additional constrained condition of the impurities number conservation; $N = V/V_m$ is the total number of "lattice sites", where liquid crystal molecules can be arranged. There is same number of "interstice" accessible for the penetration of an impurity, but only some of them will be occupied. To be exact, we have $\left(\pi\sqrt{2}/6\right) V/V_{\text{im}} \approx 0.7405\, V/V_{\text{im}} \ll N$, where V_{im} is the natural volume of one sphere-like impurity particle [14]. Therefore, it is supposed that the largest rest of "sites" of the embedded lattice remains vacant. In other words, $c(\mathbf{R}) \ll 1$.

The nematic host is described with a director vector field $\mathbf{n}(\mathbf{r})$ which is directed along the primary orientation of nematic molecules in a vicinity of the point $\{\mathbf{r}\}$. The initial director orientation $\mathbf{n}_0$ is chosen along the axis $\mathbf{n}_0 = (0, 0, 1)$. The geometry of the problem is illustrated in Fig. 1.

The elastic part of the free energy of a host-medium is determined by the well-known expression [15]:

$$\Delta F_{\rm el} = \frac{1}{2}\int\limits_V \left\{K_1(\operatorname{div}\mathbf{n})^2 + K_2\left(\mathbf{n}\cdot\operatorname{rot}\mathbf{n}\right)^2 + \right.$$

$$\left. + K_3\left[\mathbf{n}\times\operatorname{rot}\mathbf{n}\right]^2\right\} d\mathbf{r}. \quad (1)$$

Here, K_1, K_2, and K_3 are Frank–Oseen coefficients. In expression (1), we have neglected the saddle-splay elasticity characterized by the divergence coefficient, K_{24} [16, 17], since we do not consider the effect of external boundaries of the system at issue. Nevertheless, when the nematic cell is sufficiently small, but is considerably thicker than the anchoring extrapolation length [17], a modulated phase in the planar nematic may arise, as a result of saddle-splay distortions. The positive modules K_j (j = 1, 2, 3) are implicit functions of the absolute temperature T, through their dependence on the density distribution $\rho(\mathbf{r})$ and mainly on the scalar measure $Q\left(T,\rho(\mathbf{r}),c_0\right)$ that describes a long-range orientation ordering of the system. We suppose that K_j don't depend on $\mathbf{r}$ and will express them through the mean order parameter $S\left(T,\rho(\mathbf{r}),c_0\right) \equiv V^{-1}\int\limits_V Q\left(T,\rho(\mathbf{r}),c_0\right)\, d\mathbf{r}$:

$$K_j\left(\rho(\mathbf{r});\, Q\left(T,\rho(\mathbf{r}),c_0\right)\right) \cong K_j\left(\rho_0;\, S\left(T,\rho_0,c_0\right)\right) + \ldots.$$

It is quite natural to consider that the energy of a wetting-like bond between an every surface section of an individual impurity sphere-like macroparticle and the nematic host depends on the orientation of a director and the surrounding environment density. Then the total energy of such an interaction of all impurities with the medium can be written as

$$\Delta F_{\rm rel} = \sum_{\mathbf{R}\in V} c(\mathbf{R}) \int\limits_V g\left(\mathbf{r}-\mathbf{R},\mathbf{n},\rho;Q\left(T,\rho,c_0\right)\right)\, d\mathbf{r}\,, \quad (2)$$

where $g\left(\mathbf{r}-\mathbf{R},\mathbf{n},\rho;Q\left(T,\rho,c_0\right)\right)$ is the phenomenological density of such an interaction.

In the self-consistent field (correlationless) approximation, let us consider explicitly the director orientation independent term of the free energy [18]:

$$F_{\rm pos} = \Delta F_{\rm unfloc} + k_{\rm B}T\sum_{\mathbf{R}\in V} c(\mathbf{R})\ln c(\mathbf{R}) +$$

$$+k_{\rm B}T\sum_{\mathbf{R}\in V}\left(1-c(\mathbf{R})\right)\ln\left(1-c(\mathbf{R})\right) +$$

$$+\frac{1}{2}\iint\limits_V \rho(\mathbf{r})\rho(\mathbf{r}')E\left(\mathbf{r}-\mathbf{r}';\mathbf{S}\right) +$$

$$+k_{\rm B}T\int\limits_V \rho(\mathbf{r})\ln\left(\frac{\rho(\mathbf{r})}{\rho_{\mathbf{0}}}\right)\mathbf{dr}. \quad (3)$$

Here, $\Delta F_{\rm unfloc} = \frac{1}{2}\sum\limits_{\mathbf{R},\mathbf{R}'\in V} c(\mathbf{R})c(\mathbf{R}')U(\mathbf{R}-\mathbf{R}')$ is a contribution of the direct impurity–impurity pairwise interaction (van der Waals-type) with the potential energy $U(\mathbf{R}-\mathbf{R}')$. Two next terms describe a contribution of the impurity subsystem mixing entropy to the free energy. The quantity $E(\mathbf{r}-\mathbf{r}';S)$ characterizes the nonlocal interaction of a pair of physically small host volume elements. The last term is host medium subsystem configurational entropies in the self-consistent field approximation.

Thus, the full free energy of the system is

$$F = F_0 + \Delta F_{\rm el} + \Delta F_{\rm rel} + \Delta F_{\rm pos}, \quad (4)$$

where F_0 is independent of the impurity distribution configuration, and depends on three variables: $\mathbf{n}(\mathbf{r})$ is the director vector field, $\rho(\mathbf{r})$ is the distribution function of the nematic liquid crystal density, and $c(\mathbf{R})$ is the one-particle distribution function of impurities.

3. State Stability Loss

To obtain the statistical-thermodynamic state equations for the "equilibrium liquid crystal – impurities" system and to find the conditions of stability loss, we will consider small deviations from the initial uniform state: $c(\mathbf{R}) \equiv c_0 + \delta c(\mathbf{R})$, $\mathbf{n}(\mathbf{r}) \equiv \mathbf{n}_0 + \delta\mathbf{n}(\mathbf{r})$, $\rho(\mathbf{r}) \equiv \rho_0 + \delta\rho(\mathbf{r})$.

Let us expand the energy density of the interaction between nematic molecules and impurity particles in a series in the variables $\delta\rho(\mathbf{r})$, $\delta c(\mathbf{R})$, and $\delta\mathbf{n}(\mathbf{r})$ to within the second order (see details in Ref. [12]). Further, we will use the representation of this series in terms of the parameters $g_0 \equiv g(\mathbf{r}-\mathbf{R},\mathbf{n_0},\rho_0;Q_0)$ ($Q_0 \equiv Q(T,\rho_0,c_0)$) and the derivatives of $g(\mathbf{r}-\mathbf{R},\mathbf{n_0},\rho;Q)$, which are taken at values of their arguments for the initial pseudohomogeneous conditions, $\mathbf{n}=\mathbf{n_0}$ and $\rho=\rho_0$. For example, the derivatives $\frac{\partial^2 g(\mathbf{r}-\mathbf{R},\mathbf{n_0},\rho;Q)}{\partial\delta n_j\partial\rho}\,|_{n_0,\rho_0}$ can be written as $g''_{n_j\rho}(\mathbf{r}-\mathbf{R})$.

For the further consideration of stability-loss conditions, we will apply the static fluctuation-wave method

[19] following the rules

$$\tilde{f}(\mathbf{k}) = \frac{1}{v}\int_V f(\mathbf{r})e^{-i\mathbf{kr}}d\mathbf{r},$$

$$f(\mathbf{r}) = \frac{1}{N}\sum_{\mathbf{k}\in BZ}\tilde{f}(\mathbf{k})e^{i\mathbf{kr}} \cong \frac{v}{(2\pi)^3}\int_{\mathbf{k}\in BZ}\tilde{f}(\mathbf{k})e^{i\mathbf{kr}}d\mathbf{k}.$$

With the wave vector $\mathbf{k} = \mathbf{0}$ – the parameter of Fourier transform, the integration and the summation are carried out over all wave vectors of the first Brillouin zone (BZ), and $v = V/N$ is the volume of a primitive unit cell of the notionally chosen quasilattice. The reality of the function under transformation, $f(\mathbf{r})$, provides an additional constraint on the Fourier components: $\tilde{f}(\mathbf{k}) = \tilde{f}^*(-\mathbf{k})$. As mentioned above, the initial director direction is chosen as $\mathbf{n_0} = (0, 0, 1)$; thus, the director normalization leads to the relation

$$\sum_{\mathbf{k}}\left(\delta\tilde{\mathbf{n}}(\mathbf{k})\cdot\delta\tilde{\mathbf{n}^*}(\mathbf{k})\right) = -2\left(\mathbf{n_0}\cdot\delta\tilde{\mathbf{n}}(\mathbf{0})\right). \quad (5)$$

While minimizing functional (4), we take constraint (5) into account by means of the indefinite Lagrange multiplier, λ (see also Appendix A). The constraints, $\sum_{\mathbf{R}\in V}\delta c(\mathbf{R}) = 0$ and $\int_V \delta\rho(\mathbf{r})d\mathbf{r} = 0$, which correspond to the conservation of the numbers of impurity particles and nematic-LC-molecules, respectively, lead to the following properties of the 'zeroth' Fourier components: $\delta\tilde{\rho}(\mathbf{0}) = 0$ and $\delta\tilde{c}(\mathbf{0}) = 0$.

With the second-order accuracy in $\delta\tilde{c}(\mathbf{k})$, $\delta\tilde{\rho}(\mathbf{k})$, and $\delta\tilde{\mathbf{n}}(\mathbf{k})$, let us find functional F (4) by taking into account the latest constraints, as well as the director normalization (5).

In the case of $\mathbf{k} \neq 0$ after applying the Lagrange–Euler equations (for instance, in the form of $\frac{\partial F}{\partial(\mathrm{Re}\delta\tilde{\rho}(\mathbf{k}))} + i\frac{\partial F}{\partial(\mathrm{Im}\delta\tilde{\rho}(\mathbf{k}))} = 0$ and $\frac{\partial F}{\partial(\mathrm{Re}\delta\tilde{n}_j(\mathbf{k}))} + i\frac{\partial F}{\partial(\mathrm{Im}\delta\tilde{n}_j(\mathbf{k}))} = 0\,(j = x, y, z)$), one can obtain a system of linearized equations of state in a vicinity of the stability-loss point, $T_{\mathrm{bif}}(\rho_0, c_0)$ (see Ref. [12] and Appendix A).

To simplify the system of equations, we will specify the density of interaction energy between the host molecules and surfaces of the impurities. The following form of the interaction corresponds to the homeotropic boundary condition [15] on the impurity surface, which is realized in most experiments:

$$g(\mathbf{r}-\mathbf{R}, \mathbf{n_0}, \rho; Q) \cong$$

$$\cong -\frac{\left(\mathbf{n}(\mathbf{r})\cdot(\mathbf{r}-\mathbf{R})\right)^2}{|\mathbf{r}-\mathbf{R}|^2}\beta(\mathbf{r}-\mathbf{R}, \mathbf{n_0}, \rho; Q), \quad (6)$$

$$g'_{n_i}(\mathbf{r}-\mathbf{R}) = g'_{n_i}\,|_{\mathbf{n_0},\rho_0} \cong$$

$$\cong -2\frac{(\mathbf{r}-\mathbf{R})_i(\mathbf{r}-\mathbf{R})_z}{|\mathbf{r}-\mathbf{R}|^2}\beta(\mathbf{r}-\mathbf{R}, \mathbf{n_0}, \rho; Q), \quad (7)$$

for $|\mathbf{r}-\mathbf{R}| \geq d_{\mathrm{im}}$. Here, β is a function which characterizes the radial dependence of the density of interaction energy between the impurities and the liquid-crystal molecules. In the case of $|\mathbf{r}-\mathbf{R}| < d_{\mathrm{im}}$ (where $\rho(\mathbf{r}) \equiv 0$), we will suppose that $g \equiv 0$. The Fourier components of various derivatives of $g(\mathbf{r}-\mathbf{R}, \mathbf{n_0}, \rho; Q)$ (6) are presented in Appendix B.

To obtain a non-trivial solution of the linearized Lagrange–Euler equations (which is true near the point of uniform-state stability loss), we require that the determinant of the above-mentioned system of equations be equal to zero (see Appendix A). This condition allows us to find the solution bifurcation temperature $T_{\mathrm{bif}}(\rho_0, c_0)$, where a loss of the homogeneous-state stability against an inhomogeneous one can occur.

In such a way, we can obtain the director-distortion profile of a host nematic LC and the spatial density distribution of its molecules:

$$\delta\tilde{n}_j(\mathbf{k}) \approx -\frac{1}{K_1K_3k^4}\left\{\left[(K_3-K_1)(\mathbf{k}\cdot\tilde{\mathbf{g}}'_{\mathbf{n}}(\mathbf{k}))k_j + \right.\right.$$
$$\left. + K_1k^2\tilde{g}'_{n_j}(\mathbf{k})\right]\delta\tilde{c}(\mathbf{k}) + \left[c_0(K_3-K_1)k_z\tilde{g}''_{n_z\rho}(\mathbf{0})k_j + \right.$$
$$\left.\left. + c_0K_1k^2\tilde{g}''_{n_z\rho}(\mathbf{0})\delta_{jz}\right]\delta\tilde{\rho}(\mathbf{k})\right\}, \quad (8)$$

$$\delta\tilde{\rho}(\mathbf{k}) \approx$$
$$\approx \left\{-\tilde{g}'_\rho(\mathbf{k}) + \frac{c_0}{K_1K_3k^4}\left[(K_3-K_1)(\mathbf{k}\cdot\tilde{\mathbf{g}}'_{\mathbf{n}}(\mathbf{k}))k_z + \right.\right.$$
$$\left.\left. + K_1k^2\tilde{g}'_{n_z}(\mathbf{k})\right]\tilde{g}''_{n_z\rho}(\mathbf{0})\right\}\delta\tilde{c}(\mathbf{k}) \times$$
$$\times\left\{\frac{k_{\mathrm{B}}T_{\mathrm{bif}}}{\rho_0} + v\tilde{E}(\mathbf{k}) + c_0\tilde{g}''_{\rho\rho}(\mathbf{0}) - \right.$$
$$\left. - \frac{c_0^2}{K_1K_3k^4}\left[(K_3-K_1)k_z^2 + K_1k^2\right]\left[\tilde{g}''_{n_z\rho}(\mathbf{0})\right]^2\right\}^{-1}, \quad (9)$$

(δ_{jz} is the Kronecker symbol). The existence condition of nontrivial solutions of the system of respective linearized equations (see Appendix A), in the case at issue, (6) and (7), is as follows:

$$\frac{1}{v}\frac{k_bT_{\mathrm{bif}}}{c_0(1-c_0)} + \frac{1}{v}\tilde{U}(\mathbf{k}) -$$

$$-\frac{1}{K_1K_3k^4}\left[(K_3-K_1)|(\mathbf{k}\cdot\tilde{\mathbf{g}}'_{\mathbf{n}}(\mathbf{k}))|^2 + K_1k^2|\tilde{g}'_n(\mathbf{k})|^2\right] -$$

$$-\left|\tilde{g}'_\rho(\mathbf{k}) - \frac{c_0}{K_1K_3k^4}\left[(K_3-K_1)(\mathbf{k}\cdot\tilde{\mathbf{g}}'_{\mathbf{n}}(\mathbf{k}))k_z + \right.\right.$$

$$+K_1k^2\tilde{g}'_{n_z}(\mathbf{k})\big]\,\tilde{g}''_{n_z\rho}(\mathbf{0})\Big|^2\left\{\frac{k_{\rm B}T_{\rm bif}}{\rho_0}+v\tilde{E}(\mathbf{k})+c_0\tilde{g}''_{\rho\rho}(\mathbf{0})-\right.$$

$$\left.-\frac{c_0^2}{K_1K_3k^4}\left[(K_3-K_1)k_z^2+K_1k^2\right]\left[\tilde{g}''_{n_z\rho}(\mathbf{0})\right]^2\right\}^{-1}\cong 0. \quad (10)$$

As seen even from this particular relation and from the non-linearized impure-LC equilibrium equations based on a general expression for the free energy, one can define the Fourier components of the effective pair-interaction energy (both direct and indirect ones) between the sphere-like impurity particles (in particular, by means of a nematic-LC medium). This interaction, which acts as a collective mechanism of formation of modulated structures, can be presented in the following way:

$$\tilde{V}_{\rm eff}(\mathbf{k})\cong\tilde{U}(\mathbf{k})-$$

$$-\frac{v}{K_1K_3k^4}\left[(K_3-K_1)|(\mathbf{k}\cdot\tilde{\mathbf{g}}'_{\mathbf{n}}(\mathbf{k}))|^2+K_1k^2|\tilde{g}'_n(\mathbf{k})|^2\right]-$$

$$-v\left|\tilde{g}'_\rho(\mathbf{k})-\frac{c_0}{K_1K_3k^4}\left[(K_3-K_1)(\mathbf{k}\cdot\tilde{\mathbf{g}}'_{\mathbf{n}}(\mathbf{k}))k_z+\right.\right.$$

$$\left.+K_1k^2\tilde{g}'_{n_z}(\mathbf{k})\right]\tilde{g}''_{n_z\rho}(\mathbf{0})\Big|^2\left\{\frac{k_{\rm B}T_{\rm bif}}{\rho_0}+v\tilde{E}(\mathbf{k})+c_0\tilde{g}''_{\rho\rho}(\mathbf{0})-\right.$$

$$\left.-\frac{c_0^2}{K_1K_3k^4}\left[(K_3-K_1)k_z^2+K_1k^2\right]\left[\tilde{g}''_{n_z\rho}(\mathbf{0})\right]^2\right\}^{-1}, \quad (11)$$

where $\mathbf{k}\neq\mathbf{0}$. It is easy to understand that $\lim_{\mathbf{k}\to\mathbf{0}}\tilde{V}_{\rm eff}(\mathbf{k})\neq\tilde{V}_{\rm eff}(\mathbf{0})$. The *non-analytic* features of $\tilde{V}_{\rm eff}(\mathbf{k})$ in a vicinity of $\mathbf{k}=\mathbf{0}$, including $\tilde{V}_{\rm eff}(\mathbf{0})$, which are concerned with the self-interaction exclusion, will be published elsewhere. Nevertheless, we will consider this dependence on the distance from the point $\mathbf{k}=\mathbf{0}$ in a subsequent section of this work.

We note that the elastic constant, K_2, which is responsible for the resistance to a twist distortion of a nematic LC, does not enter into expressions (8)–(11) for sphere-like impurity particles. Thus, in the general case, the effective interaction energy between the impurities depends on the temperature, nematic-medium density, and concentration of impurities. This feature allows controlling the structure and properties of a system under study by means of changing the external thermodynamic conditions.

4. Numeric Calculations

Let us estimate some numerical parameters. With a goal of numerical calculations by formulas (10) and (11), we will neglect the energy parameters of the direct interaction, $\tilde{U}(\mathbf{k})$, for the pairwise van der Waals-type interaction between impurities in further discussions. It can be done because their values seem to be lower by several orders than the respective indirect-interaction parameters (related to distortions of the director-vector field) at typical interimpurity distances. In addition, as expected, $\tilde{E}(\mathbf{k})$ describing the 'interaction' between small host-volume elements may be compensated by $\frac{k_{\rm B}T}{\rho_0}$ (see the denominator of the third term on a right-hand side of (11)).

Let us specify the radial factor of the density of interaction energy between the host molecules and the impurity surface (6) as follows [3]:

$$\beta(\mathbf{r}-\mathbf{R},\mathbf{n_0},\rho;Q)\cong W\rho\exp\left(-\kappa(|\mathbf{r}-\mathbf{R}|-d_{\rm im})/\sigma_{\rm m}\right) \quad (12a)$$

or

$$\beta(\mathbf{r}-\mathbf{R},\mathbf{n_0},\rho;Q)\cong W\rho\delta_+\left(-\kappa(|\mathbf{r}-\mathbf{R}|-d_{\rm im})/\sigma_{\rm m}\right) \quad (12b)$$

for $|\mathbf{r}-\mathbf{R}|>d_{\rm im}$, where $\rho(\mathbf{r})>0$ is the host-LC volume density, and $\delta_+(\xi)$ is the asymmetric Dirac delta function $\delta_+(+0)=1$ [20]. W and κ are (fitting) parameters which depend on the surfactant kind and its concentration on the impurity surface and characterize the interaction energy between an impurity particle (at $\mathbf{R}$) and a nematic-LC element (at $\mathbf{r}$) with a characteristic linear size of $\sigma_{\rm m}\cong\sqrt[3]{V_{\rm m}}$. (Further, we use values of the κ parameter in (12a) to make the 'screened' impurity-nematic interaction range of the order of the 'length' of several nematic molecules.) The parametrization of expressions (12a) and (12b) results from both the explicit form of functional dependences (2) and (3) with regard for (4) and the fact that, without a nematic-LC medium, the contribution concerned with director distortions must also disappear in (4) and (11).

We considered different types of the nematic host medium (PAA, MBBA, 5CB, 8CB), but the results turned out to be similar. Therefore, we will illustrate them only for impurities in 8CB. Values of the elasticity parameters, K_1 and K_3, are considered to have dependence on the temperature according to experimental data [21]. Calculations were carried out for the impurity size $d_{\rm im}=5\times10^{-4}$ cm that is the most common experimental result.

In the case of typical nematics, our numerical estimations demonstrate that (11) can be reduced to

$$\tilde{V}_{\rm eff}(\mathbf{k}) \cong \tilde{U}(\mathbf{k}) - \frac{W^2\rho_0^2}{vK_1K_3k^4} \times$$

$$\times \left\{ \Big[(K_3-K_1)|(\mathbf{k}\cdot\tilde{\mathbf{G}}'_{\mathbf{n}}(\mathbf{k}))|^2 + K_1k^2|\tilde{G}'_n(\mathbf{k})|^2\Big] + \right.$$

$$\left. + \frac{\Big|\Big[(K_3-K_1)(\mathbf{k}\cdot\tilde{\mathbf{G}}'_{\mathbf{n}}(\mathbf{k}))k_z + K_1k^2\tilde{G}'_{n_z}(\mathbf{k})\Big]\Big|^2}{[(K_3-K_1)k_z^2 + K_1k^2]} \right\}, \quad (13)$$

$\tilde{g}'_n(\mathbf{k}) \equiv \frac{W\rho_0}{v}\tilde{G}'_n(\mathbf{k})$. We note that the third term on the right-hand side of (13) gives a considerable contribution to the total effective interaction energy. However, this contribution is neglected in the literature concerned with phenomenological theories, but it was often investigated numerically [22]. The origin of this term in (13) is related to a spatial inhomogeneity of the nematic-host density, $\delta\tilde{\rho}(\mathbf{k})$, even in spite of the explicit absence of derivatives with respect to the density. In addition, one can notice that the second and third terms in (13) have competitive character, which results in the formation of a local minimum of the effective interparticle interaction energy (13) corresponding to certain nonzero values of the modulation wave vector.

Let us examine the simplified formula (13) for Fourier components (with allowable wave vectors, $\mathbf{k} \neq \mathbf{0}$) of the renormalized impurity-impurity interaction energy, $\tilde{\Upsilon}(\mathbf{k}) = \frac{v\tilde{V}_{\rm eff}(\mathbf{k})}{W^2\rho_0^2}$, which does not depend on the intrinsic structure of the notionally chosen quasilattice describing the LC medium. For the chosen parametrization of the interaction energy between the LC medium and the impurity particles (6), (12a), and (12b), expression (13) turns out to be dependent on both the magnitude, $|\mathbf{k}|$, and the k_z component of the wave vector $\mathbf{k}$. Surfaces for $\tilde{\Upsilon}(\mathbf{k})$ (corresponding to (13) and interaction energies between the impurities with the radius $d_{\rm im} = 5 \times 10^{-4}$ cm) are plotted in Figs. 2 and 3.

Let us determine the wave vector $\mathbf{k_0}$ ($k_0 < 2\pi/(2d_{\rm im})$) which minimizes $\tilde{\Upsilon}(\mathbf{k})$ and generates a modulated structure, *i.e.* it corresponds to its period that cannot be smaller than the impurity radius. For impurity particles of the radius $d_{\rm im} = 5 \times 10^{-4}$ cm within the host LC with $K_1 \neq K_3(\neq K_2)$, $k_{0z} = 2853$ cm^{-1}, $k_{0x} = k_{0y} = 1554$ cm^{-1}, which corresponds to impurity-impurity periods $a_z = 4.4d_{\rm im}$ $a_x = a_y = 8.1d_{\rm im}$ for the case of (12a) and $k_{0z} = 4010$ cm^{-1}, $k_{0x} = k_{0y} = 1777$ cm^{-1}, which corresponds to impurity-impurity periods $a_z = 3.1d_{\rm im}$ $a_x = a_y = 7.1d_{\rm im}$ for case of (12b). For the impurity particles of the radius $d_{\rm im} = 5 \times 10^{-4}$ cm within the host LC with $K_1 = K_3(\neq K_2)$, $k_{0z} = 2354$ cm^{-1}, $k_{0x} = k_{0y} = 1636$ cm^{-1}, which corresponds to impurity-impurity periods $a_z = 5.3d_{\rm im}$ $a_x = a_y = 7.7d_{\rm im}$ for the case of (12a) and $k_{0z} = 5110$ cm^{-1}, $k_{0x} = k_{0y} = 2529$ cm^{-1}, which corresponds to impurity-impurity periods $a_z = 3.4d_{\rm im}$ $a_x = a_y = 5d_{\rm im}$ for case of (12b). These collections of periods weakly change along the entire temperature interval of the nematic phase of an LC (306-313 K for 8CB). One can see that there are the long-range and quasioscillatory characters of this interaction in its energy dependence on the interimpurity distance. Our calculations demonstrate that a larger screening parame-

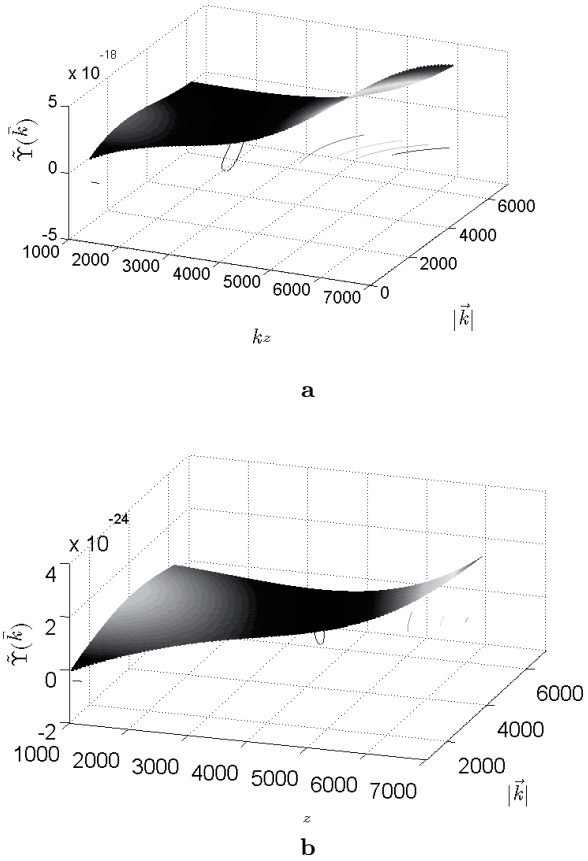
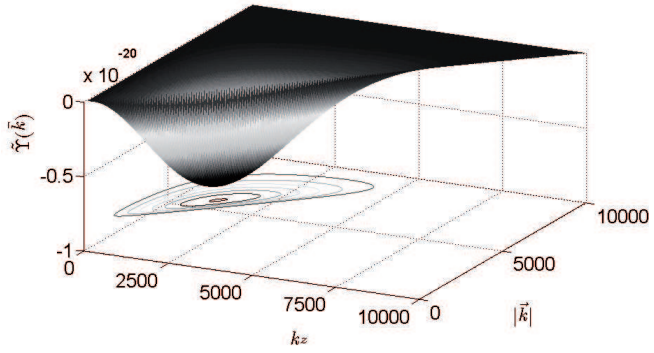
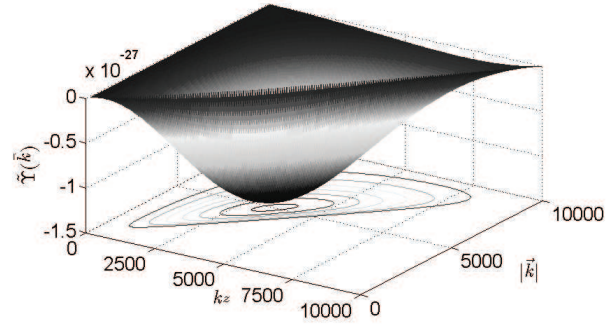


Fig. 2. Dependence of Fourier components of the effective pairwise interimpurity interaction on the wave vector for particles with the radius $d_{\rm im} = 5 \times 10^{-4}$ cm in an LC medium with $K_1 \neq K_3(\neq K_2)$ for (12a) (*a*); and (12b) (*b*)



a



b

Fig. 3. Dependence of Fourier components of the effective pairwise interimpurity interaction on the wave vector for particles with the radius $d_{\rm im} = 5\times10^{-4}$ cm in the LC medium with $K_1 = K_3 (\neq K_2)$ for (12a) (*a*); and (12b) (*b*)

ter corresponds to a smaller value of the wave vector $\mathbf{k_0}$.

5. Conclusions

We have considered the stability loss against the formation of modulated structures, basing on the phenomenological theory of filled liquid crystals.

We have obtained the system of linearized equations which describe a distortion of the director field and deviations of the nematic-density distribution and the concentration of impurity particles from their initial homogeneous-state values. These equations take into account the structure and the anisotropy of properties of the 'nematic LC + impurity' system under study.

We have obtained the condition of homogeneous-distribution stability loss against the formation of modulated structures. This condition allows one to calculate the temperature of stability loss and to estimate the period of a formed structure, for instance, along a direction of the undistorted director.

An expression for the Fourier components of the effective pairwise interaction energy between the impurity particles is derived. It takes into account the direct and indirect (by means of the nematic-LC medium) contributions to this interaction.

We find that, along with an indirect contribution to the effective impurity-impurity interaction by means of induced director-field distortions, a significant contribution to its energy is given by the interaction of impurities by means of spatial inhomogeneities of the nematic-LC density associated with nematic-director distortions and caused by impurities. Exactly such indirect contributions act as a part of a collective mechanism of formation of modulated structures.

There are the long-range and quasioscillatory characters of this interaction in its energy dependence on the interimpurity distance. In the realistic case of $K_1 \neq K_3 (\neq K_2)$, the $\mathbf{k}$-dependence of Fourier components for such an interaction is non-analytic in a vicinity of $\mathbf{k} = \mathbf{0}$.

Generally, the interaction energy depends on the temperature, density of a nematic-LC medium, and concentration of impurities. This property allows controlling the structure and properties of the studied system by changing the external thermodynamic conditions.

APPENDIX A

In the case of $\mathbf{k} \neq \mathbf{0}$, after applying the Lagrange–Euler equations, we obtain the following system of linearized equations of state in a vicinity of the stability-loss point, $T_{\rm bif}(\rho_0, c_0)$, for a uniform distribution of the system characteristics:

$$\Big[K_1k_x^2 + K_2k_y^2 + K_3k_z^2 + c_0(\tilde g''_{n_xn_x}(\mathbf{0}) - \tilde\lambda)\Big]\,\delta\tilde n_x(\mathbf{k}) +$$
$$+ \Big[(K_1 - K_2)k_xk_y + c_0\tilde g''_{n_xn_y}(\mathbf{0})\Big]\,\delta\tilde n_y(\mathbf{k}) +$$
$$+ \big[(K_1 - K_3)k_xk_z + c_0\tilde g''_{n_xn_z}(\mathbf{0})\big]\,\delta\tilde n_z(\mathbf{k}) +$$
$$+ \big[\tilde g'_{n_x}(\mathbf{k})\big]\,\delta\tilde c(\mathbf{k}) + \big[c_0\tilde g''_{n_x\rho}(\mathbf{0})\big]\,\delta\tilde\rho(\mathbf{k}) = 0, \qquad (14)$$

$$\Big[(K_1 - K_2)k_xk_y + c_0\tilde g''_{n_xn_y}(\mathbf{0})\Big]\,\delta\tilde n_x(\mathbf{k}) +$$
$$+ \Big[K_1k_y^2 + K_2k_x^2 + K_3k_z^2 + c_0(\tilde g''_{n_yn_y}(\mathbf{0}) - \tilde\lambda)\Big]\,\delta\tilde n_y(\mathbf{k}) +$$
$$+ \Big[(K_1 - K_3)k_yk_z + c_0\tilde g''_{n_yn_z}(\mathbf{0})\Big]\,\delta\tilde n_z(\mathbf{k}) +$$
$$+ \Big[\tilde g'_{n_y}(\mathbf{k})\Big]\,\delta\tilde c(\mathbf{k}) + \big[c_0\tilde g''_{n_x\rho}(\mathbf{0})\big]\,\delta\tilde\rho(\mathbf{k}) = 0, \qquad (15)$$

$$\left[(K_1-K_3)k_xk_z+c_0\tilde{g}''_{n_xn_z}(\mathbf{0})\right]\delta\tilde{n}_x(\mathbf{k})+$$
$$+\left[(K_1-K_3)k_yk_z+c_0\tilde{g}''_{n_yn_z}(\mathbf{0})\right]\delta\tilde{n}_y(\mathbf{k})+$$
$$+\left[K_1k_z^2+K_3(k_x^2+k_y^2)+c_0(\tilde{g}''_{n_zn_z}(\mathbf{0})-\tilde{\lambda})\right]\delta\tilde{n}_z(\mathbf{k})+$$
$$+\left[\tilde{g}'_{n_z}(\mathbf{k})\right]\delta\tilde{c}(\mathbf{k})+\left[c_0\tilde{g}''_{n_z\rho}(\mathbf{0})\right]\delta\tilde{\rho}(\mathbf{k})=0, \quad (16)$$

$$\left[c_0\tilde{g}''_{n_x\rho}(\mathbf{0})\right]\delta\tilde{n}_x(\mathbf{k})+\left[c_0\tilde{g}''_{n_y\rho}(\mathbf{0})\right]\delta\tilde{n}_y(\mathbf{k})+$$
$$+\left[c_0\tilde{g}''_{n_z\rho}(\mathbf{0})\right]\delta\tilde{n}_z(\mathbf{k})+\left[\tilde{g}'_{\rho}(\mathbf{k})\right]\delta\tilde{c}(\mathbf{k})+$$
$$+\left[c_0\tilde{g}''_{\rho\rho}(\mathbf{0})+v\tilde{E}(\mathbf{k})+(k_{\mathrm{B}}T)/\rho_0\right]\delta\tilde{\rho}(\mathbf{k})=0, \quad (17)$$

$$\left[\tilde{g}'^{*}_{n_x}(\mathbf{k})\right]\delta\tilde{n}_x(\mathbf{k})+\left[\tilde{g}'^{*}_{n_y}(\mathbf{k})\right]\delta\tilde{n}_y(\mathbf{k})+\left[\tilde{g}'^{*}_{n_z}(\mathbf{k})\right]\delta\tilde{n}_z(\mathbf{k})+$$
$$+\frac{1}{v}\left[\tilde{U}(\mathbf{k})+\frac{k_{\mathrm{B}}T}{c_0(1-c_0)}\right]\delta\tilde{c}(\mathbf{k})+\left[\tilde{g}'^{*}_{\rho}(\mathbf{k})\right]\delta\tilde{\rho}(\mathbf{k})=0, \quad (18)$$

where $\tilde{\lambda}=2\lambda/c_0$ is a renormalized Lagrange multiplier. The system of equations (14)-(18) is obtained in the general case, *i.e.* without any assumptions about the interaction between LC molecules and impurities, which allows us to use it under various anchoring conditions on the impurity surface.

For $\mathbf{k}=\mathbf{0}$ separately, applying the Lagrange–Euler equations, $\frac{\partial F}{\partial\delta\tilde{n}_x(\mathbf{0})}=\frac{\partial F}{\partial\delta\tilde{n}_y(\mathbf{0})}=\frac{\partial F}{\partial\delta\tilde{n}_z(\mathbf{0})}=0$, one can find a system of equations for the Lagrange-multiplier with the use of Eq.(5):

$$\left[\tilde{g}''_{n_xn_x}(\mathbf{0})-\tilde{\lambda}\right]\delta\tilde{n}_x(\mathbf{0})+\tilde{g}''_{n_xn_y}(\mathbf{0})\delta\tilde{n}_y(\mathbf{0})+$$
$$+\tilde{g}''_{n_xn_z}(\mathbf{0})\delta\tilde{n}_z(\mathbf{0})=-\tilde{g}'_{n_x}(\mathbf{0}), \quad (19)$$

$$\tilde{g}''_{n_xn_y}(\mathbf{0})\delta\tilde{n}_x(\mathbf{0})+\left[\tilde{g}''_{n_yn_y}(\mathbf{0})-\tilde{\lambda}\right]\delta\tilde{n}_y(\mathbf{0})+$$
$$+\tilde{g}''_{n_yn_z}(\mathbf{0})\delta\tilde{n}_z(\mathbf{0})=-\tilde{g}'_{n_y}(\mathbf{0}), \quad (20)$$

$$\tilde{g}''_{n_xn_z}(\mathbf{0})\delta\tilde{n}_x(\mathbf{0})+\tilde{g}''_{n_yn_z}(\mathbf{0})\delta\tilde{n}_y(\mathbf{0})+$$
$$+\left[\tilde{g}''_{n_zn_z}(\mathbf{0})-\tilde{\lambda}\right]\delta\tilde{n}_z(\mathbf{0})=\tilde{\lambda}-\tilde{g}'_{n_z}(\mathbf{0}). \quad (21)$$

APPENDIX B

Let us determine the Fourier components which are the parts of (10) and (11):

$$\tilde{g}'_{n_x}(\mathbf{k})=\frac{8\pi}{v}\frac{k_xk_z}{k^2}\times$$
$$\times\int\limits_{d_{\mathrm{im}}}^{\infty}ds\beta_0(s)s^2\left[\frac{3\sin(ks)}{(ks)^3}-\frac{3\cos(ks)}{(ks)^2}-\frac{\sin(ks)}{ks}\right], \quad (22)$$

$$\tilde{g}'_{n_y}(\mathbf{k})=\frac{8\pi}{v}\frac{k_yk_z}{k^2}\times$$
$$\times\int\limits_{d_{\mathrm{im}}}^{\infty}ds\beta_0(s)s^2\left[\frac{3\sin(ks)}{(ks)^3}-\frac{3\cos(ks)}{(ks)^2}-\frac{\sin(ks)}{ks}\right], \quad (23)$$

$$\tilde{g}'_{n_z}(\mathbf{k})=-\frac{8\pi}{3v}\int\limits_{d_{\mathrm{im}}}^{\infty}ds\beta_0(s)s^2\frac{\sin(ks)}{ks}+\frac{8\pi}{3v}\frac{3k_z^2-k^2}{k^2}\times$$
$$\times\int\limits_{d_{\mathrm{im}}}^{\infty}ds\beta_0(s)s^2\left[\frac{3\sin(ks)}{(ks)^3}-\frac{3\cos(ks)}{(ks)^2}-\frac{\sin(ks)}{ks}\right], \quad (24)$$

$$\tilde{g}'_{\rho}(\mathbf{k})=-\frac{4\pi}{3v}\int\limits_{d_{\mathrm{im}}}^{\infty}ds\beta'_{\rho0}(s)s^2\frac{\sin(ks)}{ks}+\frac{4\pi}{3v}\frac{3k_z^2-k^2}{k^2}\times$$
$$\times\int\limits_{d_{\mathrm{im}}}^{\infty}ds\beta'_{\rho0}(s)s^2\left[\frac{3\sin(ks)}{(ks)^3}-\frac{3\cos(ks)}{(ks)^2}-\frac{\sin(ks)}{ks}\right], \quad (25)$$

where d_{im} is the impurity particle radius, $\beta_0(s)=\beta(|\mathbf{r}-\mathbf{R}|,\rho_0)$, $\beta'_{\rho0}(s)=\beta'_{\rho}(|\mathbf{r}-\mathbf{R}|,\rho_0)|_{\rho_0}$, $s=|\mathbf{r}-\mathbf{R}|$, and, $k^2=|\mathbf{k}|=k_x^2+k_y^2+k_z^2$.

In the case of $\mathbf{k}=\mathbf{0}$, we obtain

$$\tilde{g}_0(\mathbf{0})=-\frac{4\pi}{3v}\int\limits_{d_{\mathrm{im}}}^{\infty}ds\beta_0(s)s^2, \quad (26)$$

$$\tilde{g}'_{n_x}(\mathbf{0})=\tilde{g}'_{n_y}(\mathbf{0})=0,\ \tilde{g}'_{n_z}(\mathbf{0})=2\tilde{g}_0(\mathbf{0}), \quad (27)$$

$$\tilde{g}'_{\rho}(\mathbf{0})=\tilde{g}'_{0_\rho}(\mathbf{0})=0,\ \tilde{g}''_{\rho\rho}(\mathbf{0})=\tilde{g}''_{0_{\rho\rho}}(\mathbf{0}),$$
$$\tilde{g}''_{n_z\rho}(\mathbf{0})=2\tilde{g}'_{0_\rho}(\mathbf{0}), \quad (28)$$

$$\tilde{g}''_{n_xn_x}(\mathbf{0})=\tilde{g}''_{n_yn_y}(\mathbf{0})=\tilde{g}''_{n_zn_z}(\mathbf{0})=2\tilde{g}'_0(\mathbf{0}), \quad (29)$$

$$\tilde{g}''_{n_xn_y}(\mathbf{0})=\tilde{g}''_{n_yn_z}(\mathbf{0})=\tilde{g}''_{n_yn_z}(\mathbf{0})=$$
$$=\tilde{g}''_{n_x\rho}(\mathbf{0})=\tilde{g}''_{n_y\rho}(\mathbf{0})=0. \quad (30)$$

Therefore, considering the case of parametrization (6) and Eqs. (19)–(21), one can obtain the Lagrange multiplayer: $\lambda=c_0\tilde{g}_0(\mathbf{0})$.

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НЕСТАБІЛЬНІСТЬ НЕМАТИЧНО-ПОДІБНИХ ФАЗ, НАПОВНЕНИХ ДОМІШКАМИ, ЩОДО ФОРМУВАННЯ МОДУЛЬОВАНИХ СТРУКТУР

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Р е з ю м е

Розглянуто просторовий розподіл домішкових жорстких сфероподібних макрочастинок у мезоморфному рідкокристалічному носії. Використовуючи континуальні статично-механічні теорії, проаналізовано термодинамічні умови, необхідні для появи модульованих шаруватих надструктур. Між домішковими частинками присутня ефективна далекодійна взаємодія. Вважається, що вона відповідає за формування структур. У загальному випадку така взаємодія містить два внески: пряму взаємодію типу Ван-дер-Ваальса та непряму (через поле викривлення директора). Остання залежить від температури зразка та концентрації частинок. Ця ефективна взаємодія контролює структуру та властивості системи. Було одержано аналітичні розв'язки для розподілу поля директора, неоднорідності густини носія, температури формування модульованих структур та їх просторового періоду. Запропонований теоретичний підхід може бути застосований до інших анізотропних та неоднорідних систем.