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COULOMB-LIKE ELASTIC INTERACTION IN LIQUID CRYSTAL COLLOIDS

This article is dedicated to the memory of my teacher P.M. Tomchuk and contains a review of his important results in one of the many areas of theoretical physics in which new solutions and approaches have been proposed, namely, in the theory of liquid crystal colloids. A theoretical approach to the description of long-range elastic interaction between particles immersed in a liquid crystal is proposed. It is shown that the nature of the interaction between particles is dictated by the symmetry breaking in the distribution of the elastic director field around each particle. The symmetry breaking is caused by deformations of the director field by the surface of the particle introduced into the liquid crystal. In cases where the particles induce a deformation with a non-zero torque moment, the Coulomb-like interaction between them is predicted. In addition, it is determined that the Coulomb interaction occurs in the cases of interaction of a particle with a deformation region characterized by a specific distribution of the elastic field. The paper presents experimental data confirming the theoretical predictions of the Coulomb-like interaction of immersed macroscopic particles in a liquid crystal.

K e y w o r d s: liquid crystal colloids, Coulomb-like interaction, soft matter, elastic interaction.

1. Introduction

Liquid crystals are materials, which break their ground state symmetry in distribution elastic director field under the action of a weak external influences. Another way to break a continuous symmetry in liquid crystals is to introduce a particle of other substance in the liquid crystal. Such immersed particles distort the director field of the liquid crystal over distances much larger than the size of the particle. As shown below, any interaction between immersed particle in the liquid crystal is connected with some symmetry breaking in the begraund state. The elastic interactions between particles through the deformation elastic director field lead to nontrivial behaviors with the formation of different of novel ordered or disordered structures [1–8]. Was observed are linear chains of small water droplets in liquid crystal or in large nematic drops [9, 10]; highly ordered arrays of silicon oil droplet in nematic liquid crystal [11]; 2D hexagonal lattice of glycerin droplets in a nematic cell with hybrid boundary conditions [6, 7] and 3D structure in system hard macroparticles immersed in liquid crystals. Many articles have already been published in this area [7–24], in which we can find a lot of interesting things both from the point of view of fundamental and applied sciences.

The symmetry is broken for two reasons: the shape of the particle and anchoring strength of the bond between the liquid crystal molecules and the particle surface. In the case of weak anchoring, this is primarily determined by the shape of the particle. In the case of strong anchoring, both factors are important, since the directional distribution near the particle is determined by topological defects in its vicinity. In order to describe these two cases in a universal way, the concept of a deformation shell around a particle was introduced [2]. The deformation coat encompasses the associated topological defects and has the same symmetry as the resulting director field around the particle. The distribution of the director field outside the deformation shell undergoes only smooth changes and does not contain any topological defects. It can then be argued that

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the long-range interaction between particles is determined by the symmetry of the deformation shell and can be expressed through its physical characteristics, such as the shape and magnitude of the coupling with surface.

Another way of breaking the continuous symmetry of ground state of director distribution in liquid crystals is the spontaneous appearance of a deformation region. An external impact or boundary conditions on the cell surface can cause a deformation of uniform distribution of the director field. In this case, we must consider the superposition of the strain field around the particle and the global deformation of the equilibrium distribution of director field. The superposition of different deformation leads to an effective interaction of the particles immersed in liquid crystal. This interaction leads to the motion of the particle in the deformed directional field. In this case, a non-trivial behavior of particles in the deformed elastic director field can be realized and a different type of elastic interaction can be observed.

The interaction between particles immersed in a liquid crystal occurs due to a change in the free energy of the deformations of the director field by the immersed particles. The free energy of the liquid crystal with introduced particles under such conditions will depend on the distance between them. Minimizing this free energy will be determines the nature of the interaction between the particles introduced into the elastic medium. In this way, the behavior of a separate particle in a distorted director field can also be considered. Let us assume that the elastic field of the director is inhomogeneous due to global boundary conditions and that the length of the director deformation is much larger than the particle size. In this case, we only have the free energy of the particle in the deformed director field, and this energy varies from point to point in these deformation regions. The particle will be moved until it reaches the minimum of the resulting free energy. Anchoring conditions on the surface particles of particle and the global symmetry of the liquid crystal have an mpact on the value of the elastic interparticle interactions and on the collective behavior of the particles immersed in liquid crystals [1, 2].

Currently, there have been two approaches to describing the nature of the interaction between particles of different shapes introduced into a liquid crystal. The first one deals with particles which have a strong anchoring on the surface [12, 13]. The particle with strong planar anchoring produce the pair topological defects, known as boojums. The particles with strong homeotropic boundary conditions produce the disclination ring on equator or a hyperbolic hedgehog near of surface as a companion for the radial hedgehog in the center of the particle. Using the analogy with electrostatic, Lubensky *et al.* [12] obtained an approximate of director filed distribution near the droplet with homeotropic boundary conditions, as well as the long-range pair interaction potential between the particles. In this case, the interaction between particles has the dipole-dipole and the quadrupole-quadrupole character. The dipole-dipole interaction explains the formation of the chains, which are aligned along the director in the nematic liquid crystal. About such character of interaction and formation linear chains of small water droplets in nematic liquid crystal or in large nematic drops was described in the articles [9, 10].

The second approach was proposed jointly with P.M. Tomchuk in the articles [1, 2], where the case of weak anchoring for particles of different shape is considered. An analytically interaction potential was found, with regard for various Frank constants, and this potential was expressed through the tensor characteristics of the particle shape. When the distribution of director field in a vicinity of the particle has three planes of symmetry, the pair interaction potential are quadrupole and dependence as d^{-5}, where d is the distance between the particles. When one plane of symmetry is broken, a dipole moment arises, which leads to dipole-dipole interaction between the particles. If the deformation coat has only one plane of symmetry (the director $\mathbf{n}$ or when it has no planes of symmetry at all, then arises the Coulomb-like interaction between particles at a large distance [2]. The main achievement of these studies is that it has been shown that this interaction far exceeds the thermal energy and may be the main reason for the creation of structures in a system of macroscopic particles immersed in a liquid crystal. The nature and magnitude of the interaction between particles of different shapes have been verified experimentally with great accuracy and, in addition, the corresponding structures predicted theoretically have been observed.

2. Interaction between Particles in Liquid Crystal

Let us now turn to the main provisions of the proposed theoretical approach, which allows us to reveal the reasons for the interaction between colloidal particles through deformations of the elastic director field. A nematic liquid crystal is an anisotropic liquid in which the elongated molecules have the same average orientation, which is described by a unit vector $\mathbf{n}$ called the director. In the undistributed state, the nematic has a spatially uniform orientation of $\mathbf{n}_0$, and we assume here that it is parallel to the z axis ($\mathbf{n}_0 = (0, 0, 1)$). The immersed particles destroy the uniform orientation of the director in the bulk. These deformations are caused by boundary conditions on the surface of particles immersed in a liquid crystal. The bulk orientations of the director field imposed at the surface of particles in such a way that the nematic molecules lie either normally or tangentially to surface of particles. The phenomenological free anchoring energy at the surface of particles can be written in the well-known Rapini–Papoular form

$$F_s = \sum_p W \oint ds(\nu(\mathbf{s}) \cdot \mathbf{n}(\mathbf{s}))^2, \tag{1}$$

where W is the anchoring energy coefficient. For the homeotropic anchoring $W < 0$ and, for the planar one, $W > 0$. The summation of bulk deformation director fields should be taken over all particles in the liquid crystal for determination of the free energy.

The bulk energy of the spatial distortions of the director field, that is called the Frank energy, is written in the form

$$F_b = \frac{1}{2}\int d^3r \Big\{K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n})^2 + + K_{33}(\mathbf{n}\times\operatorname{rot}\mathbf{n})^2\Big\}. \tag{2}$$

In order to find possible director configurations one should solve Euler–Lagrange (EL) equations from the minimization of the Frank free energy with regard for the boundary conditions (BC), which are found from the minimization of bulk and surface energies. But the situation can arise, when there are several director field distributions with different symmetries, which satisfy both EL equations in the bulk and boundary conditions at the surface.

The breaking of the symmetry in the near-field region is achieved either by the anchoring strength or by the shape of particle's and leads to the different solutions in the far-field region. At the far distances from the particle, the director field $\mathbf{n}(\mathbf{r})$ tends to be uniform $\mathbf{n}_0 = (0, 0, 1)$ and can be written in the form $\mathbf{n} = (n_x, n_y, 1)$. In the one-constant approximation the Frank free energy is given by

$$F_b = \frac{1}{2}K \int d^3\{(\nabla n_x)^2 + (\nabla n_y)^2\}. \tag{3}$$

Equilibrium equations which leads from minim of the free energy are the Laplace equations for the transverse components n_μ ($\mu = x, y$)

$$\Delta n_\mu = 0. \tag{4}$$

The solution of this equation at large distances r can be expanded in multipole presentation

$$n_\mu = \frac{A_\mu}{r} + \frac{\mathbf{p}_\mu \mathbf{r}}{r^3} + \frac{c_\mu^{ij} r_i r_j}{r^5} + \dots. \tag{5}$$

It is clearly seen that transverse components can be treated as the elastic field potential by analogy with the electrostatic where particles are as sources of possible deformation. The first part is connected with the "charge", the second with the dipole moment, and the last term is connected with the quadrupolar moment produced by particles.

Three items represent different broken symmetries of the director field around particles and are responsible for three different interaction laws between particles, as will be show below. The first term exists, when the director distribution does not have any plane of the symmetry at all or it has only one vertical plane of the symmetry. It appears when the particle in its vicinity disappear mirror symmetry in a horizontal plane and in one vertical plane. In other terms, it exists, when there is a nonzero torque moment $\mathbf{\Gamma}$ acting to the particle by the nematic [15]. In the absence of the torsion moment $\mathbf{\Gamma}$ it is absent. The second term represents broken symmetry in one plane and the dipole moment $\mathbf{p}$ is the measure of skewness. The last term exists in any case, because it has the same quadrupolar symmetry as the distribution of uniform director field has.

The multipole expansion is valid only in the region, where nonlinearity can be neglected. For particles with strong anchoring, it is the far-region because of a strong director deformations in the near-region. But, for weakly anchored particles, the dis-

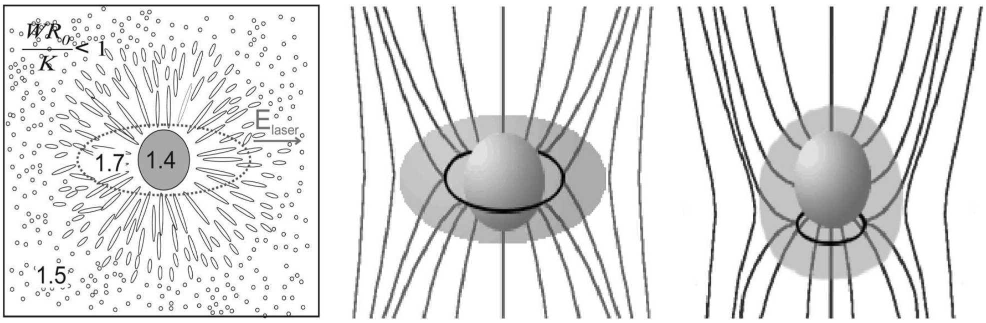

Fig. 1. Representation of the region of the director's elastic field deformation under weak and strong anchoring

tortions are small, and multiple expansion is applicable in the near region. In general, the smaller the anchoring strength, the smaller the size of the region, where multipole expansion is not applicable. These many ties are presented on the Fig. 1.

In the first paper for liquid crystal colloids theory [1] the authors suggested the approach, which enables one to find the interaction potential for particles of ordinary shape with weak anchoring at the surface. It is valid for the different Frank constants and so exceed the results of the electrostatic analogy. For the weak anchoring strength there are no exist topological defects and the director deformations $\delta\mathbf{n}$ are small everywhere, so that the multiple expansion is valid also at the particle surface as well. Unknown coefficients can be expressed through tensor characteristics of the particle's surface and orientation. This article appeared almost simultaneously with the paper [12] where the interaction potential for colloidal particles in the case of strong anchoring to the surface was phenomenological obtained.

The approximation of self-consistence director behavior was used, where the deformations of the director's elastic fields created by other particles are adjusted to the director's boundary conditions required by the single particle shape. This was a physical assumption under which the results are valid and allows us to find the interaction potential between the particles. Mathematically this can be right in the case of $\delta\mathbf{n} \ll \mathbf{1}$ far from the particle surface. According to the article [1,3] interaction potential between the two macroscopic particles separated by the distance $\mathbf{R}$ in general case may be written as:

$$U(\mathbf{R}) = -\frac{1}{8\pi} \sum_{m,m'=1,2,3} \widehat{A}^p_m \widehat{A}^{p'}_{m'} \sum_{\mu=1,2} \frac{1}{\sqrt{K_{\mu\mu}}} \times$$

$$\times \left\{ \frac{Q^+_{m,m'}}{\sqrt{K_{33}R_\perp^2 + K_{\mu\mu}R_\parallel^2}} + \right.$$

$$\left. +(-1)^\mu \frac{Q^-_{m,m'}}{R_\perp^2} \frac{\left(\sqrt{K_{33}R_\perp^2 + K_{\mu\mu}R_\parallel^2} - \sqrt{K_{\mu\mu}R_\parallel^2}\right)^2}{\sqrt{K_{33}R_\perp^2 + K_{\mu\mu}R_\parallel^2}} \right\}. \quad (6)$$

In this expression $R_\parallel$ and $R_\perp$ are parallel and perpendicular to the unformed director $\mathbf{n}_0$ components of the $\mathbf{R} = \mathbf{r}_p - \mathbf{r}_{p'}$:

$$\mathbf{r}_1 = \frac{\mathbf{R}_\perp \times \mathbf{n}_0}{R_\perp}, \quad \mathbf{r}_2 = \frac{\mathbf{R}_\perp}{R_\perp}, \quad \mathbf{r}_3 = \mathbf{n}_0, \qquad (7)$$
$$\mathbf{R}_\perp = \mathbf{n}_0 \times \mathbf{R},$$

$Q^{(\pm)}_{m,m'} = (\mathbf{r}_1 \cdot \mathbf{k}_m)(\mathbf{r}_1 \cdot \mathbf{k}_{m'}) \pm (\mathbf{r}_2 \cdot \mathbf{k}_m)(\mathbf{r}_2 \cdot \mathbf{k}_{m'})$, where $(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$ is the local basis rigidly bound with each particle. Operators $\widehat{A}_m$ are defined as:

$$\widehat{A}_m = (\mathbf{k}_l \mathbf{n}_0) \left[\alpha_{lm} + \beta_{lms}(\mathbf{k}_s \cdot \nabla) + \right.$$
$$\left. + \gamma_{lmst}(\mathbf{k}_s \cdot \nabla)(\mathbf{k}_t \cdot \nabla)\right]. \qquad (8)$$

Superscript p means that, in operator $\widehat{A}^p_m$, we need to substitute $\nabla = \frac{\partial}{\partial \mathbf{r}_p}$ that relate with point of p particle.

Here, $\alpha_{lm}, \beta_{lms}, \gamma_{lmst}$ are tensor characteristics of the deformation coat, which contain all information about symmetry of director field around a particle. If ρ is the vector pointing from the center of mass of the particle to the point $\mathbf{s}$ at the surface of the coat, and ν is the unit normal to the surface at this point, this

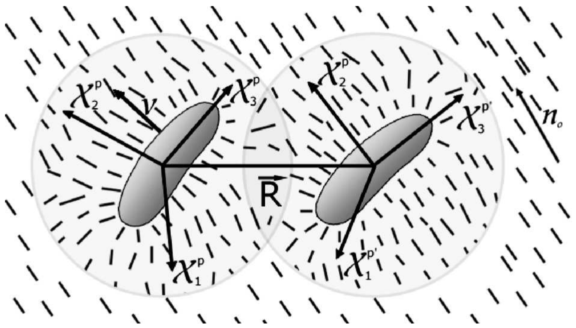

Fig. 2. A schematic representation of the arrangement of two macroparticles of different shapes in a liquid crystal. The figure shows the three main axes of the particles and the deformation of the director field around an individual particle

coefficients can be presented as

$$\alpha_{kl} = 2\oint d\sigma W_c(\mathbf{s})\nu_k(\mathbf{s})\nu_l(\mathbf{s}),$$
$$\beta_{klm} = 2\oint d\sigma W_c(\mathbf{s})\nu_k(\mathbf{s})\nu_l(\mathbf{s})\rho_m(\mathbf{s}), \quad (9)$$
$$\gamma_{klmn} = \oint d\sigma W_c(\mathbf{s})\nu_k(\mathbf{s})\nu_l(\mathbf{s})\rho_m(\mathbf{s})\rho_n(\mathbf{s}).$$

The integration is over the surface of the deformation coat which determine the area, where deformation is strong. The symmetry of these tensors contains all data on the broken symmetry of the director field in vicinities of particles and defines distinctive features of the interaction potential in the far-region. The magnitudes of it can be treated like variational parameters for the concordance with experimental data, or it can be evaluated from the comparison with long-range asymptotic of solutions.

For instance, let us consider the comparison with the potential which presented in article [12]. For this purpose, we consider one-constant approximation, and present our results in the form:

$$U(\mathbf{R}) = -\frac{1}{8\pi}\sum_{m,m'=1,2,3}\widehat{A}^p_m\widehat{A}^{p'}_{m'}\left(\frac{Q^+_{m,m'}}{R}\right). \quad (10)$$

It was shown that $Q^+_{m,m'} = 0$ for m or m' equal 3, and $Q^+_{m,m'} = \delta_{m,m'}$, for $m, m' = 1, 2$. Using of this allows us to write the expression in the form

$$U(\mathbf{R}) = -\frac{\alpha_{3m}\alpha_{3m}}{4\pi R} + \beta_{3ms}\beta_{3ms'}(\mathbf{k}_s\cdot\nabla)(\mathbf{k}_{s'}\cdot\nabla)\frac{1}{4\pi R} -$$
$$-\gamma_{3mst}\gamma_{3mst}(\mathbf{k}_s\cdot\nabla)(\mathbf{k}_{s'}\cdot\nabla)(\mathbf{k}_t\cdot\nabla)(\mathbf{k}_{t'}\cdot\nabla)\frac{1}{4\pi R}, \quad (11)$$

where the summation on the repeating indices is made. After that, it is clear that the results of both approaches coincide and make it possible to use the electrostatic analogy to explain many facts of the behavior of colloidal particles in a liquid crystal. This formula shows how the shape and size of the particles introduced into the liquid crystal affect the interaction and also makes it possible to follow the elements of broken symmetry in the director field distribution. If the shape of the particles, their size and the value of the energy of adhesion to the particle surface are known, it is possible to obtain the magnitude and nature of the interaction between them, when they are immersed into the liquid crystal.

3. Coulomb-Like Elastic Interaction

The bases of the theoretical description of these phenomena were outlined in [1, 2, 12, 23]. Their main idea is rooted in the fact that, far from the particle, the director deviations $\delta\mathbf{n}$ from its ground state $\mathbf{n}_0$ are small and satisfy the Euler–Lagrange equations $\Delta\delta\mathbf{n} = 0$. The deformations dierector field which are produced by single immersed particle arise as a break of the symmetry of the ground state. At the short distance from the particle, the deformation of the ground state is impossible to be obtained, because the liquid crystal is nonlinear with the natural condition $\mathbf{n}^2(\mathbf{r}) = 1$. The topology of distribution of the director field around the particle depends on the strength of the anchoring energy. The deviation from the symmetry of the ground state can be determine at the far distance. At the far distance, we can determine all possible small deformations of the director field which account for the breaking of the symmetry of the distribution director field on a short distance from the particle [1, 2]. The possibility of the appearance of monopole-monopole interaction was predicted in articles [1, 2] and was presented in experimental result to verify theoretical prediction in articles [4, 5].

The every immersed particle by the shape and the boundary condition on the surface produce the deformation of the ground state $\mathbf{n}_0$ which are small everywhere at the far distance: $\mathbf{n}(\mathbf{r}) = \mathbf{n}_0 + \delta\mathbf{n}(\mathbf{r})$, $|\delta\mathbf{n}| \ll 1$ and $\mathbf{n}_0 \cdot \delta\mathbf{n}(\mathbf{r}) = 0$. In the case of nematic liquid crystals, we have only $\delta\mathbf{n}(\mathbf{r}) = (\delta\mathbf{n}_\perp, 0)$, where zero determine ground state and, in the case of a twisted liquid crystal, we have all components.

From the Euler–Lagrange equation, we are allowed to expand $\delta\mathbf{n}(\mathbf{r})$ in multiple expansion

$$\delta\mathbf{n}(\mathbf{r}) = \frac{\mathbf{q}}{r} + \frac{\mathbf{p}^{\alpha}\mathbf{r}_{\alpha}}{r^3} + \frac{\mathbf{Q}^{\alpha\beta}\mathbf{r}_{\alpha}\mathbf{r}_{\beta}}{r^5} + ..., \tag{12}$$

where α and β take values x, y, z and summation over repeated Greek indices is assumed. Coefficients q, p^{α}, $Q^{\alpha\beta}$ are called elastic monopoles (charges) or torque moment, dipoles and quadrupole, respectively. As it follows from multiple presentation (12), director deviations have a long-range nature. This means that the deformations caused by different particles can overlap, even if the particles are located far from each other. Since this system cannot minimize its energy by minimizing all the deformations separately. In the practice of the overlapping manifests itself in the fact that a colloidal particle "feels" the presence of the other particles mediated by a nematic host, i.e., in the appearance of the effective long-range elastic interactions between colloidal particles.

These elastic long-range interactions in bulk nematic colloids are determined completely by the coefficients q_{μ}, p^{α}_{μ}, $Q^{\alpha\beta}_{\mu}$. In the case of a strong anchoring, they must be found from asymptotic behavior of the solutions of nonlinear equations describing $\mathbf{n}(\mathbf{r})$ in a vicinity of the particle. But when the anchoring is weak, $\delta\mathbf{n}$ is small and consequently expansion (12) is valid everywhere outside the particle. Under these circumstances coefficients are determined by the symmetry of the particle surface [2]. Dipoles appear as a result of the broken mirror symmetry [2]. Quadrupole moment have the vertical and horizontal elements of symmetry and saved the symmetry of the ground state. Deformation charge arises is results of the breaking of any symmetry of the ground state.

But, till now, it is commonly believed that a colloidal particle itself, despite its symmetry, never produces an elastic monopoles, because this violates the mechanical equilibrium condition. The only way to obtain the deformations of the director field falling off as r^{-1} is to exert an external torque $\mathbf{\Gamma}_{\text{ext}}$ on the colloid [26]. Using the electrostatic analogy helps us only to understanding the possible form of the interaction energy, but it is not valid in the general case [5]. Was demonstrate that this statement is not quite correct, and elastic monopoles, as well as dipoles and quadrupoles, can be induced without and presence the external influence only by the particle itself.

3.1. Interaction between ellipsoidal particles

In present approach interaction between two ellipsoidal particles in a homogeneously oriented nematic liquid crystal was studied as well. An exact analytic formula for the Coulomb-like long distance interaction potential between two ellipsoids with arbitrary nonzero eccentricity and different orientation of long ellipsoidal axis was obtained in the case of weak anchoring. Final expression for the Coulomb elastic interaction between two arbitrarily oriented ellipsoidall particles with weak anchoring can be presented as

$$U^{[CC]} = -\frac{W_1 W_2}{8\pi K R}\cos(\psi_1 - \psi_2)\sin 2\theta_1 \times$$

$$\times \sin 2\theta_2 F(b_1, \varepsilon_1) F(b_2, \varepsilon_2), \tag{13}$$

where

$$F(b,\varepsilon) = \pi b^2\sqrt{1-\varepsilon^2}\left\{\frac{\sqrt{1-\varepsilon^2}}{\varepsilon^2}(2\varepsilon^2 - 3) + \right.$$

$$\left. + \frac{\arcsin\varepsilon}{\varepsilon^3}(3 - 4\varepsilon^4)\right\} \tag{14}$$

for polar and azhimutal orientation angles in spherical coordinate system θ, ψ and eccentricity ε. This is an exact formula for the case of weak anchoring, when the director field has no defects near the ellipsoids. General estimate shows that topological defects do not appear for anchoring strengths $Wb/K \leq 1$, with b being the characteristic length of the particle (in our case, the long axis). Formula above shows that the Coulomb-like interaction exists, only when $\theta_1, \theta_2 \neq 0, \pi/2$, i.e., when particles are neither parallel nor perpendicular but tilted with respect to the director field. Namely, only in the tilted case the local symmetry is broken in two planes – one horizontal and one vertical, and a nonzero torque moment is also exerted on the particle from the nematic. Interaction potential $U^{[CC]}$ has the following set of basic properties:

1. Particles with the same sign of the anchoring and with the same inclined orientation ($|\psi_1 - \psi_2| < \pi/2$) attract each other following the Coulomb law.

2. Particles with different signs of the anchoring (one – planar, other – homeotropic) and with the same inclined orientation ($|\psi_1 - \psi_2| < \pi/2$) repel one another following the Coulomb law.

3. Particles with the same sign of the anchoring and opposite inclined orientation ($|\psi_1 - \psi_2| > \pi/2$) repel each other by the Coulomb law.

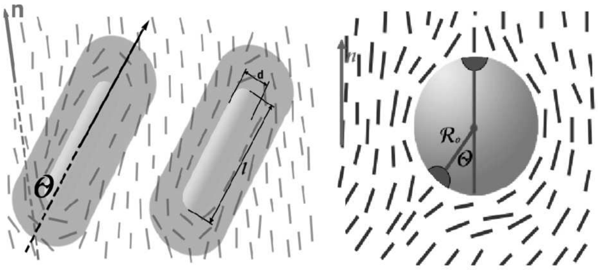

Fig. 3. The figure shows how the energies of two tilted cylinders in a liquid crystal change with a decrease in the distance between them. The deformation region of the director decreases, and hence the elastic energy, due to the convergence of the tilted cylinders. This causes an attraction and the magnitude of the attraction force depends on the angle of inclination

4. Coulomb interaction goes to zero as the fourth power of the eccentricity $U_{\varepsilon \Rightarrow 0}^{[CC]} \propto \varepsilon^4$.

The emergence of the deformation Coulomb interaction was tested on the example of elongated magnetic particles under a change in the external magnetic field, which changed the equilibrium orientation of these particles. Theoretically, the Coulomb-like interaction was predicted even for spherical particles with non-symmetrically placed regions with an another energy of adhesion of liquid crystal molecules to the surface of the inserted particle (see Fig. 3).

3.2. Experimental confirmation of the deformation coulomb interaction

In this part, we briefly describe the experimental situation concerning all directly observed Coulomb-like interaction between two monopoles which produced by special boundary condition on the surface of the sample [5]. We have examined theoretically elastic interactions between particle and different deformations director field which can also pose the external influence. We examined it with help of the general considerations concerning the breaking of different elements of symmetries of the director field in vicinities of particles. This is caused both by the shape of particles and the anchoring strength on their surfaces. In special case, we may obtain the Coulomb-like interaction which can observed by experiment.

It is well known that nematics, unlike isotropic liquids, transmit torques. As it was shown in [26], torque $\mathbf{\Gamma}$, acting on NLC, may be written in the form

$$\mathbf{\Gamma} = \left[\mathbf{n} \times \frac{\delta F}{\delta \mathbf{n}}\right], \tag{15}$$

where F is the nematic free energy. Since the director deformations have energy

$$F_{\mathrm{def}} = \frac{K}{2} \int dV \left[(\nabla \cdot \mathbf{n})^2 + (\nabla \times \mathbf{n})^2\right], \tag{16}$$

they are coupled with some torque $\mathbf{\Gamma}_{\mathrm{def}}$. But only monopoles make a nonzero contribution to $\mathbf{\Gamma}_{\mathrm{def}}$

$$\mathbf{\Gamma}_{\mathrm{def}} = \left[\mathbf{n} \times \frac{\delta F_{\mathrm{def}}}{\delta \mathbf{n}}\right] = 4\pi K \mathbf{q}^T, \tag{17}$$

where $\mathbf{q}^T = (q_y, q_x, 0)$ and $\Gamma_z^{\mathrm{def}} = 0$ since a rotation around $\mathbf{n}_0$ does not alter F_{def}. Deformations decreasing faster then r^{-1} are not related to any torque. In turn, $\mathbf{\Gamma}_{\mathrm{def}}$ can be treated as the torque we need to exert on a nematic to induce elastic monopoles q_x and q_y in there. Now, let us assume that we have a particle immersed in some bulk sample of NLC, and there are no external torques exerted on it, $\mathbf{\Gamma}_{\mathrm{ext}} = 0$. If there exist elastic monopoles the particle will "feel" torque $-\mathbf{\Gamma}_{\mathrm{def}}$ and, under these circumstances ($\mathbf{\Gamma}_{\mathrm{ext}} = 0$), will constantly rotate. Obviously this is not a physical situation. Therefore, we ought to state that the only source of elastic monopoles is the external torque exerted on the particle, $\mathbf{\Gamma}_{\mathrm{ext}} = -\mathbf{\Gamma}_{\mathrm{def}}$.

But the point is that the energy of the colloidal system is not exhausted just by bulk deformations. It contains the energy of the nematic – particle's surface interaction as well. This energy can be written in Rapini–Papoular form

$$F_{\mathrm{surface}} = \oint dS\ W(\mathbf{s}) \left[\boldsymbol{\nu}(\mathbf{s}) \cdot \mathbf{n}(\mathbf{s})\right]^2, \tag{18}$$

where $W(\mathbf{s})$ is the anchoring strength. As it was noted above, in the weak anchoring case $\mathbf{n} = \mathbf{n}_0 + \delta\mathbf{n}$, $\delta\mathbf{n} \ll 1$ everywhere, and the surface energy gives rise to torque $\mathbf{\Gamma}_{\mathrm{surface}}$

$$\mathbf{\Gamma}_{\mathrm{surface}} = \left[\mathbf{n} \times \frac{\delta F_{\mathrm{surface}}}{\delta \mathbf{n}}\right] \approx$$
$$\approx 2 \oint dS\, W\, (\boldsymbol{\nu} \cdot \mathbf{n}_0)\, [\mathbf{n}_0 \times \boldsymbol{\nu}]. \tag{19}$$

If the particle has been broken "horizontal" (i.e., perpendicular to the $\mathbf{n}_0$), and at least one of "vertical" symmetry planes integrals (19) can be nonvanishing. In the equilibrium the total torque acting on the system: particle + LC has to be zero

$$\mathbf{\Gamma}_{\mathrm{total}} = \mathbf{\Gamma}_{\mathrm{ext}} + \mathbf{\Gamma}_{\mathrm{def}} + \mathbf{\Gamma}_{\mathrm{surface}} = 0. \tag{20}$$

Hence, in the general case, q_μ are produced by both external torque and particle itself

$$\mathbf{q}^T = -\frac{\mathbf{\Gamma}_{\text{ext}} + \mathbf{\Gamma}_{\text{surface}}}{4\pi K}. \qquad (21)$$

We can also look at this issue from another viewpoint. In terms of mathematics, expression (17) is obtained from the divergence theorem. Indeed, volume integral $\frac{\delta F}{\delta \mathbf{n}}$ may be transformed into some integral over a closed surface Σ. This implies that torques acting on the nematic bulk must be balanced by surface torques [26]. When we deal with a bulk nematic Σ can be chosen at $r \to \infty$, and we come to (17), i.e., torques associated with monopoles can be balanced only by external agents. But, in a colloidal system, we have a slightly different situation. Besides Σ, there is the particle surface. This real surface cannot be ignored in the divergence theorem and leads to the expression (21). This fact is a simple illustration of the difference between electrostatics and nematostatics. If the electric charge (monopole) is a real physical point object, the elastic monopole is to a certain extent artificial object. The multipole expansion in nematostatics is just a way to describe deformations of the director field via point source. Although, in fact, they are produced by real particle surface.

Overall, particle–particle interactions consist of all contributions of the overlapping director field deformations that are caused by the surface anchoring of particles and substrate boundaries. In this study, was observed the motion of pairs of dipole interacting particles at the boundary of two different alignment regions. One region had a small twist structure due to the different orientation of the director on the cell walls. This region bordered another one, where there was pure homeotropic alignment. Colloidal particles with a dipole configuration first moved toward the interface between the two regions and then approached each other at different speeds, with the speed of mutual motion being greater at longer distances. Theoretically, this could be reconciled by accepting the hypothesis of the emergence of the Coulomb interaction due to the violation of symmetry in the distribution of the director arriving at different points on the surface. That is, external conditions for inducing a torque are created. The mechanical equilibrium is not disturbed. The experimental results reveal the first proof of the presence of Coulomb-like interactions on the far distance in elastic media between separate colloidal particles.

In article [27] was demonstrate theoretically and experimentally that deformation elastic charges (monopoles), as well as dipoles and quadrupoles deformation, can be induced through anisotropic boundary conditions. Was reported the first direct observation of Coulomb-like elastic interactions between colloidal particles in a nematic liquid crystal. The behaviour of two spherical colloidal particles with asymmetric anchoring conditions induced by asymmetric alignment is investigated experimentally; the interaction of two particles located at the boundary of twist and parallel aligned regions is observed. It was demonstrate that such particles produce deformation elastic charges and interact by Coulomb-like interactions.

3.3. Screening of the elastic interaction

Theoretical approach to the interaction of the macroparticles via deformations of the director field [3] found that distortion of the director field induced by many particles leads to the screening of pair elastic interaction potential. This screening strongly depends on the shape of the immersed particles: it exists for anisotropic particles and is absent for spherical ones. These results are valid both for the homeotropic and planar anchoring on the particle's surface and for the different Frank constants.

The effective "charge" in the screened Coulomb-like attraction greatly depends on the angle between the cylindrical grains and the director. This angle is zero in the equilibrium states, when the grains lie parallel or perpendicular to the director in the case of the planar or homeotropic anchoring. External magnetic field which is not parallel to the initial orientation of the magnetic grains bring them out of the equilibrium state and make inclined angle between the grains and the director so that effective "charge" arises. When the cylinders make an angle with the director, the screened Coulomb attraction of the Yukawa form arises between them which can lead to nontrivial consequences. It was shown that it is this potential that is responsible for the "cellular" texture in ferronematics which was observed by Chen and Amer [28]. It was also shown that the screening has not always exponential but it can be trigonometrical under some conditions. It can be only in the presence of the external field when the angle between the grains and the director exceeds the critical threshold.

In the one-constant approximation $K_{\mu\mu} = K_{33} = K$ this potential becomes dependent only on the scalar of the vector $\mathbf{R}$ of distance between gains:

$$U_{pp'} = -\frac{Q^{+}_{l,l'}}{4\pi K}\widehat{A}^{p}_{l}\widehat{A}^{p'}_{l'}\left[\frac{\exp(-\xi\,|\mathbf{r}_p-\mathbf{r}_{p'}|)}{|\mathbf{r}_p-\mathbf{r}_{p'}|}\right], \qquad (22)$$

$$\xi^{-1}(\theta) = \sqrt{K/ca(\theta)}. \qquad (23)$$

It is clearly seen that collective distortions of the director lead to the screening of the pair interaction potential with the screening length $\xi^{-1} \approx \sqrt{K/WcS}$ (we mean W is absolute value here not depending on the sign), S surface area of the particle. For spherical particle $S = 4\pi R^2$ with radius R. This screening takes place both for the homeotropic and for the planar anchoring. Concentration here is included in the inverse screening length ξ only, so that the limit $c \to 0$ makes $\xi = 0$ and gives us back to the unscreened result [1], which is equivalent to the result of [30] for the case of asymmetric cylinders. All this consideration is true only if $\xi^{-1} \gg \langle l\rangle = 1/\sqrt[3]{c}$ – the average distance between the particles.

4. Behavior of Colloidal Particle in Deformed Director Field

Next, consider the behavior of particles in the curved director field. We suppose that the director field is not homogeneous because of the global boundary conditions, and that the deformation length of director field is much more than the size of particles. In this case, we have only free energy of particle in curved director field and this energy can change from point to point for deformation area. The particle must moving to spatial points minima of the free energy.

Inasmuch as the global deformations scale is large, we can express the director on the surface $\mathbf{n}(\mathbf{s})$ through the director at the center of the masses of the deformation coat

$$\mathbf{n}(\mathbf{s}) = \mathbf{n}_0 + (\rho\nabla)\,\mathbf{n}_0 + \frac{1}{2}\,(\rho\nabla)^2\,\mathbf{n}_0$$

ρ is the vector of distance from the center of mass to the point $\mathbf{s}$. Here, $\mathbf{n}_0$ is the local director field, which would have been in the center. Then the anchoring energy can wright in the form

$$F_s = F_0 + F_d + F_q,$$

$$F_0 = \oint d\sigma W_c(\mathbf{s})(\nu\cdot\mathbf{n}_0)^2,$$

$$F_d = 2\oint d\sigma W_c(\mathbf{s})(\nu\cdot\mathbf{n}_0)\,(\rho\nabla)\,(\nu\cdot\mathbf{n}_0),$$

$$F_q = \oint d\sigma W_c(\mathbf{s})[(\nu\cdot\mathbf{n}_0)\,(\rho\nabla)^2\,(\nu\cdot\mathbf{n}_0) + ((\rho\nabla)\,(\nu\cdot\mathbf{n}_0))^2].$$

The first part F_0 is responsible for the orientation of the particle with respect to the director, whereas the second and the third parts describes the behavior of the whole particle induced by the inhomogeneous distribution of director field. All the scalar value can be represented in the basis $(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$ like $\nu \times \times\, \mathbf{n}_0 = (\nu\mathbf{k}_l)(\mathbf{n}_0\mathbf{k}_l) = \nu_l n_l$, $(\rho\nabla) = \rho_s\,(\mathbf{k}_s\nabla) = \rho_s\partial_s$. Then as result can present anchoring free energy as

$$F = \alpha_{ij}n_i n_j + \beta_{l\mu s}n_l\partial_s n_\mu + \gamma_{l\mu st}\partial_s(n_l\partial_t n_\mu). \qquad (24)$$

These are the general expressions which describe movement of the particles in the curved director field. Coefficients α_{ij}, β_{ijk} and γ_{ijklm} is characteristic of shape of the particle and boundary condition on the surface on this particle. The component n_i describe the distribution director field where located this particle. The particle will move in the way to minimize the anchoring energy. The first term describes the behavior of the particle with all breaking symmetry in the curved director field. The second one describe the behavior of the particle with dipole symmetry, and last term describe the behavior of particles with quadruple symmetry in the curved director field. If the particle has azimuthal symmetry, then nonzero components of the β are $\beta_{311} = \beta_{322}$ and free energy takes the form

$$F_d = \beta_{311}n_3(\partial_1 n_1 + \partial_2 n_2) = -4\pi K p_z n_z \,\mathrm{div}\,\mathbf{n}, \qquad (25)$$

which coincides with the result [12] and indicates, that dipoles assemble in the places with high splay deformations. This is confirmed experimentally: small water drops gather in the center of the big nematic droplet with homeotropic anchoring on the surface (near radial hedgehog) and assemble near the surface boojums, when the global conditions are planar [10].

The F_q term for the quadruple particles with azimuthal symmetry takes the form

$$F_q = \gamma_{1313}\,(\mathbf{n}\nabla)\,\mathrm{div}\,\mathbf{n}. \qquad (26)$$

We see that particles, which do not have dipole moment (for example, small spherical particles for which $WR/K \ll 1$) move differently in dependence on the sign of the anchoring strength. For planar anchoring $W > 0$ and $\gamma_{1313} > 0$, for homeotropic anchoring $\gamma_{1313} < 0$, so that particles with planar anchoring move toward the places with high splay and particles with homeotropic anchoring repel from the regions with high splay. We see that although the sign of the anchoring do not influence the quadrupole-quadrupole interaction of particles, it plays the crucial role in the behavior of particles in the curved director field. We will describe, in the next part, the possible behavior of particles with different shape in deformation director field which can be arise in different sample of the liquid crystal. Curved director field can arise as result of boundary condition as external influence. In present approach can describe the possible interaction between different particle and different deformation area of distribution elastic director field. Next can consider interaction of colloidal particle with possible and well know the director field deformation.

4.1. Particles in the droplet of liquid crystal

In this part, consider the motion own particle for the experiment of Poulin [9, 10] where exist the liquid crystal droplet and water droplets which found in this more droplet. In this case, we have configuration of the director field everywhere of with radial topological defect in the center. In the spherical coordinate system this distribution can right in the form $n_r = 1, n_\theta = 0, n_\varphi = 0$. In this case we have only splay deformation when $\operatorname{div} \mathbf{n} = \frac{2}{r}$ and free energy can be present by the form $F_d = -4\pi K p_r n_r \operatorname{div} \mathbf{n}$. Every particle with dipole moment p_r will interaction with radial distribution of director field, when the free energy takes the form $F_d = -4\pi K p_r \frac{2}{r}$. Need note, that interaction between particle and splay deformation dependent from sign dipole moment.

If we determined the sign dipole moment as direction between positive topological charge (hedgehog defect) an negative topological defect (radial) we can obtain that the particle with positive dipole moment will attractive to center droplet and, in opposite case, will repulsive from center. In the first case, we have configuration, when the hedgehog defect in the center must observed motion the particle to the center by the Coulomb-like law. In this case, the particle moving to the center of the liquid crystal droplet up to that time, when stop in the center. Negative topological defect in center destroy hedgehog defect and will be existence only radial defect. If the particle has no dipole moment, but only a qudrupole moment, we can obtain the free energy in the form $F_q = -\gamma_{rrrr} \frac{2}{r^2}$. For the particle with quadrupole configuration of director field attractive interaction must arise with radial distribution of director field in the liquid crystal droplet. In the case band deformation after rotating the director field at every point on $\frac{\pi}{3}$ about the vertical axis in the case radial hedgehog we can obtain a circular hedgehog. In this case exist the nonzero dipole component p_φ and distribution director field in the form $n_\theta = 1, n_r = n_\varphi = 0$. For this distribution director field, we have $\operatorname{rot}_\varphi \mathbf{n} = \frac{1}{r}$ and can obtain the energy of interaction between particle and circular hedgehog in the form $F_d = -4\pi K p_\varphi \frac{1}{r}$. In this case, we have Coulomb-like attractive interaction between particle and circular hedgehog too.

4.2. Interaction particle with disclination line

In this case, we have a particle in a cell, where there is a disclination line. Let this line be oriented along the z-axis. In this case, a cylindrical coordinate system is easy to use. In this coordinate system we can take the director field distribution in the form $n_\rho = 1$, $n_z = n_\varphi = 0$. For disclination lines there is only a splay deformation and can be obtained that $\operatorname{div} \mathbf{n} = \frac{1}{\rho}$. We obtain the free energy of particle in this deformation director field as $F_d = -4\pi K p_\rho \frac{1}{r}$.

It can be noted that the interaction between the particle and the deformation of the director field that creates the disclination lines has a Coulomb-like character in the plane perpendicular to this line. This Coulomb interaction between the particle and the disclination line is observed experimentally [29]. The nature of the interaction depends on the sign of the dipole moment. If the particle has no dipole moment but only a quadrupole one, we can obtain the free energy of interaction between particle and disclination line in the form $F_q = -\gamma_{\varrho\varrho\rho\rho} \frac{1}{\varrho^2}$. This interaction is depend on the sign of the anchoring strength [29].

4.3. Particle in laser beam

An experiment was conducted [25] where a single particle interacted with a region of deformed director field. The region of deformation was at the focus

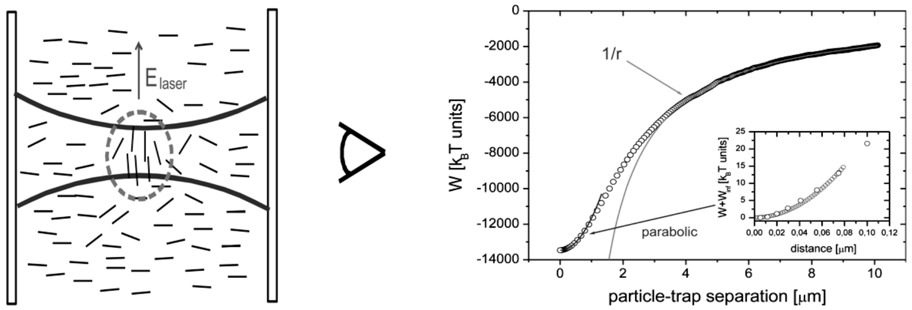

Fig. 4. Schematic representation of the experiment and the result of observing the Coulomb-like interaction of a single colloidal particle with the deformation region created by laser irradiation

of laser light incident on the liquid crystal (Fig. 4). This region was located at some distance from the particle. Having the recorded time-dependence of the distance between the particle and the laser focus, it is possible to restore the effective elastic pair potential acting on the particle. Using the Stokes force, $F = 6\pi R_{\rm eff}\eta \partial r/\partial t$, we can calculate the force between the "ghost" deformation area and the particle. Finally by integrating the force over the separation distance, we can get the interaction potential. Analysis of the interaction as a function of distance between laser focus and particle demonstrates that at the large distance the interaction is proportional to $1/r$ as in Coulomb-like case.

In the real experiment, we have three dimensional distribution of director field which can take in the cylindrical coordinate system in the form $n_z = \cos\theta$ and $n_\rho = \sin\theta, n_\varphi = 0$, where θ is angle between director and z axis. In cylindrical coordinate system in experimental case, we have

$$\text{div}\ \mathbf{n} = \frac{1}{\rho}\frac{\partial\left(\varrho n_\varrho\right)}{\partial\rho} + \frac{1}{\varrho}\frac{\partial n_\varphi}{\partial\varphi} + \frac{\partial n_z}{\partial z} = \\ = \frac{\sin\theta}{\varrho} + \cos\theta\frac{\partial\theta}{\partial\varrho} - \sin\theta\frac{\partial\theta}{\partial z},$$

and can, writ the free energy of particle, with only p_z component of dipole moment in the curved director field with splay deformation in the form

$$F_d^{\rm splay} = -2\pi K p_z \frac{1}{\rho}\sin 2\theta \qquad (27)$$

if θ = const. If θ is small the free energy has the simple form $F_d^{\rm splay} = -4\pi K p_z \frac{1}{\rho}\theta$ and we must observed Coulumb like attractive the particle to the deformation area. For the laser beam induced deformation described above, we have generally bend deformation. For a bend deformation, the free energy

$$F_d^{\rm band} = -4\pi K p_z [\mathbf{n}\times \text{rot}\,\mathbf{n}]_z = -4\pi K p_z \theta\frac{\partial\theta}{\partial z}.$$

The quadrupole part, the free energy in the case of the band deformation, can be obtained in the form $F_q^{\rm band} = -\gamma_{z\rho z\rho}\frac{\partial^2\theta}{\partial\rho\partial z}$ and this energy can take different sign from different side the deformation area. The quadrupole part of the free energy on the splay deformation is very small, because $F_q^{\rm splay} = -\gamma_{\rho\rho\rho\rho}\frac{\theta^2}{\rho^2}$. If we have non symmetric particle $\beta_{123} \neq 0$, the additional part exists in the free energy

$$F_d^{\rm twist} = -\beta_{123}\mathbf{n}\cdot \text{rot}\ \mathbf{n} = -\beta_{123}\frac{\partial\theta}{\partial\varphi},$$

which describe the interaction between non symmetric particle with twist deformation area which produce the laser beam. In all cases, the particle will move in the way to minimize the deformation energy. We still need guess, that at the short distance the director relaxation have the different type.

5. Conclusions

This paper describes a theoretical approach to determining the possible interaction of macroscopic particles of different shapes in nematic liquid crystals. It is shown that the breaking of the symmetry in the distribution of the elastic director field created by the surface of a single particle induces the corresponding type of interaction. When all the symmetry elements of the ground state of the director distribution are broken, a long-range Coulomb-like interaction arises. The effects of collective shielding, which are essential for real colloidal systems, are also taken into account. It is found that the shape of the particles has a significant influence on the screening effects for both homeotropic and planar anchoring. For spherical particles, the shielding effect is absent. Anisotropic particles (e.g., cylinders) with magnetic or electric moment, when exposed to an inclined external magnetic or electric field, induce deformation in the distribution of director field depending on the anchoring, concentration and magnitude of the external field. The collective effects in doped nematic liquid crystals are strongly dependent on the anchoring strength, particle shape, concentration, and the external field. This makes doped liquid crystals an excellent medium for further experimental and theoretical studies of various structures arising under deformations of an elastic director field.

The contribution of P.M. Tomchuk to the development of the described theory of liquid crystal colloids is very significant and important. After the results obtained, it became clear that the magnitude and nature of the interaction between colloidal particles due to the deformation of the elastic field of the director significantly exceeds the thermal energy and is crucial for the creation of structures in the system of colloidal particles. The described approach allowed us to identify and explain many non-trivial and interesting effects of the behavior of colloidal particles in such elastic media.

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КУЛОНОПОДІБНА ПРУЖНА
ВЗАЄМОДІЯ В КОЛОЇДАХ РІДИННИХ КРИСТАЛІВ

Ця стаття присвячена пам'яті мого вчителя П.М. Томчука та містить огляд його важливих результатів, отриманих в одній з багатьох галузей теоретичної фізики, де були запропоновані нові рішення та підходи, а саме, в теорії колоїдів рідинних кристалів. Запропоновано теоретичний підхід до опису далекосяжної пружної взаємодії між частинками, зануреними в рідинний кристал. Показано, що характер взаємодії між частинками визначається порушенням симетрії розподілу пружного поля директора навколо кожної частинки. Порушення симетрії спричинене деформаціями поля директора поверхнею частинки, введеної в рідинний кристал. У випадках, коли частинки викликають деформацію з ненульовим обертальним моментом, передбачається кулоноподібна взаємодія між ними. Крім того, визначено, що кулонівська взаємодія відбувається у випадках взаємодії частинки з областю деформації, що характеризується певним розподілом пружного поля. У статті представлено експериментальні дані, що підтверджують теоретичні передбачення кулонівської взаємодії макроскопічних частинок, занурених у рідинний кристал.

Ключові слова: рідиннокристалічні колоїди, кулонівська взаємодія, пружна взаємодія, м'яка речовина.