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## SYMMETRY BREAKING, ELASTIC INTERACTION AND STRUCTURES IN NEMATIC COLLOIDS

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*We discuss the role of local director symmetry in the elastic interaction of micron size particles in nematic liquid crystals. Possibility of the Coulomb-like interaction as the origin of the 2D hexagonal structure of glycerin droplets in a nematic cell with hybrid boundary conditions [9] is proposed. Effect of gathering polymer drops near the regions with strong director deformations is explained.*

### 1. INTRODUCTION

Any interaction is connected with some symmetry breaking. Liquid crystals (LC) are materials, which break their continuous symmetry under the action of weak external influences. Orientational distortions in them are easily created by external fields and boundary conditions. Another way to break continuous symmetry in liquid crystals is to introduce a particle of distinct substance in the liquid crystal host. It distorts orientational order of the liquid crystal on the distances much larger than the size of the particle. When the regions of the broken continuous symmetry around each of the two particles are overlapped, it leads to interaction between them.

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A suspension of many particles efficiently breaks the continuous symmetry of the initial liquid crystal and thus gives rise to entirely different class composite material. Properties of such liquid crystal composites are defined by collective behavior of the immersed particles interacting via the deformations of the orientational ordering of the liquid crystal.

Colloid suspensions in nematic liquid crystals has attracted an intensive consideration during last few years [1–9]. Different elastic interactions between colloids induce nontrivial behavior with the formation of a variety of novel ordered or disordered structures. Among them are chains of the small water droplets in an aligned liquid crystal and in big nematic drops [2,3]; highly ordered arrays of silicon oil droplet chains in a nematic host [8]; 2D hexagonal lattice of glycerin droplets in a nematic cell with hybrid boundary conditions [9]. In every case colloid droplets distort the director field  $\mathbf{n}(\mathbf{r})$  around them and interact elastically because of overlapping of these distortions.

There are several theoretical approaches to the descriptions of colloid interactions in the nematic liquid crystal. One of them deals with spherical droplets which have strong anchoring strength on the surface [6,11]. This anchoring creates topological defects in the vicinity of the droplet which are necessary to satisfy topological global boundary conditions. A droplets with strong planar anchoring creates a pair of topological defects, known as boojums. A droplet with strong homeotropic boundary conditions could create an equatorial disclination ring or it could nucleate a hyperbolic hedgehog as a companion for the radial hedgehog on the surface of the droplet. Using variational techniques and electrostatic analogy Lubensky *et al.* [6] have obtained the director distribution near the droplet with homeotropic boundary conditions, as well as the long range pair interaction potential between such droplets. It has both the dipole-dipole and the quadrupole-quadrupole components. The dipole-dipole interaction explains the formation of the chains, which are aligned along the director in the nematic host [4]. In [12] Lopatnikov and Namiot have found the dipole-dipole interaction of cylindrical particles in nematic and smectic LC.

In the paper [7] authors proposed a general approach to the description of the long-ranged elastic interaction in the nematic colloids, which is based on the symmetry breaking of the director field. The type of the far-field interaction between immersed particles is shown to be determined by the way of the symmetry broken in the *near-field* region around the colloidal particle.

In this article we discuss physical reasons that lead to the specific elastic interactions and propose new possible mechanism for explanation of the 2D hexagonal structure of glycerin droplets in a nematic cell with hybrid boundary conditions [9]. As well we explain experimental fact of gathering polymer drops toward the regions with strong director deformation [17].

## 2. LOCAL SYMMETRY BREAKING, COAT AND ELASTIC INTERACTION IN NEMATIC COLLOIDS

In nematic liquid crystal long molecules have the same average orientation in point  $\mathbf{x}$ , which is specified by the unit vector  $\mathbf{n}(\mathbf{x})$  that is called the director. In the undistorted state the director has spatially uniform orientation  $\mathbf{n}_0$ , and we consider, that it is parallel to the axis  $z$  ( $\mathbf{n}_0 = (0,0,1)$ ). Immersed particles distort uniform orientation of the director in the bulk. The *source* of bulk director deformations is the surface of particles, on which nematics molecules prefer to lie either normally or tangentially. The phenomenological anchoring energy at the surface of particles can be written in the Rapini-Papoular form

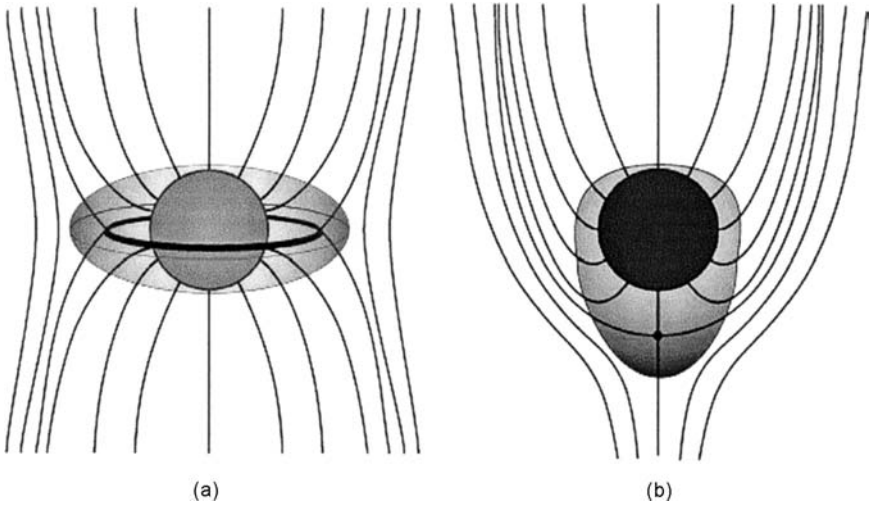
$$F_s = \sum_p W \oint ds (v(\mathbf{s}) \cdot \mathbf{n}(\mathbf{s}))^2, \quad (1)$$

where  $W$  is the anchoring coupling constant. For the homeotropic anchoring  $W < 0$  and for the planar one  $W > 0$ . Summation is over all particles in the liquid crystal. Bulk energy of the spatial distortions of the director field, that is called the Frank energy, is written in the form

$$F_b = \frac{1}{2} \int d^3r \{ K_{11} (\text{div} \mathbf{n})^2 + K_{22} (\mathbf{n} \text{rot} \mathbf{n})^2 + K_{33} (\mathbf{n} \times \text{rot} \mathbf{n})^2 \}, \quad (2)$$

The total free energy of the system is the sum of surface and bulk terms. In order to find possible director configurations one should solve Euler-Lagrange (EL) equations from the minimization of the Frank free energy with taking into account of the boundary conditions (BC), which are found from the minimization of both bulk and surface energies. But the situation can arise, when there exist several director distributions with different symmetry, which satisfy both EL equations in the bulk and boundary conditions at the surface.

The typical example is possible director configurations around the water droplet with strong homeotropic anchoring, shown in Figure 1. In both cases the director lies perpendicular to the sphere and it is equivalent to the radial hedgehog in the center. It has the topological charge, which is equal to unity. The uniform director distribution far from the droplet has zero topological charge and so there should be another topological defect near the droplet to compensate the hedgehog in the center. In the Figure 1(a) the droplet creates a  $-1/2$  disclination ring on the equator on the distance  $l = 1.08r$  from the center and in the Figure 1(b) it has a point hyperbolic hedgehog [6]. Obviously, the director configurations have different symmetry. In this case the type of the *local symmetry* of the director field is principal – it determines



**FIGURE 1** Coats that contain all topological defects inside. Anchoring  $W$  of the coat depends on the point of the surface. (a) Quadrupole coat around the Saturn-ring disclination. (b) Dipole coat with broken mirror symmetry contains radial and hyperbolic hedgehogs.

the symmetry of the far-field solution and thus the type of the far-field interaction. 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

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

Equilibrium equations are the Laplace equations for the transverse components  $n_\mu$  ( $\mu = x, y$ )

$$\Delta n_\mu = 0 \quad (4)$$

At large distances  $r$  it can be expanded in multipoles

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

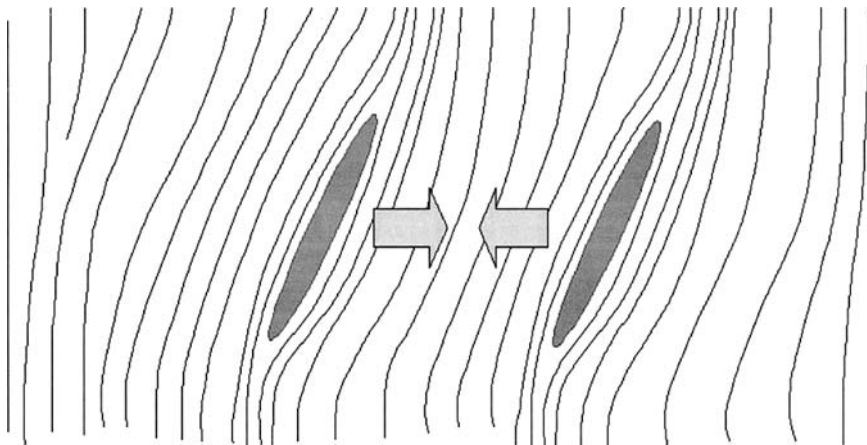
It is clearly seen, that transverse components can be treated as two components of the electric field potential and particles are multipolar sources,

similar to antennas. The first term is connected with the “charge”, the second with the dipole moment and the last term is connected with the quadrupole moment.

Below we shall follow paper [7] and show that three items in (5) represent different broken local symmetries of the director field around particles which are responsible for three different interaction law between particles. It is obvious from electrostatic analogy that the first tem should lead to the Coulomb interaction between particles. But what kind of – attraction or repulsion? If we imagine two elongated particles inclined in the same side (see Fig. 2) then the volume of the space occupied by deformations decreases when they draw together. So this means that attraction should take place. The second term should be responsible for dipole-dipole interaction and the last one for quadrupole-quadrupole interaction. This is within the scope of electrostatic analogy.

The multipole expansion is valid only in the region, where nonlinearities can be neglected. For particles with strong anchoring it is the far-region, because of strong director deformations in the near-region. But for particles with weak anchoring distortions are small elsewhere and the multiple expansion is applicable in the near-region too. In general the less anchoring strength, the less size of the region where the multiple expansion is inapplicable.

The amplitudes of this expansion in stong anchoring case can be found form the asymptotics either of exact solutions or of different variational *Ansatzs*, which correctly describe the director field in the near-region. This has been doen in [6] for spherical particles with homeotropic anchoring



**FIGURE 2** Inclined elongated particles attract each other according to the Coulomb law.

with help of electrostatic and dipole *Ansatzs*. In the weak anchoring case amplitudes in (5) are found directly from the boundary conditions for the linearized EL equations, as it has been done in [16].

In the paper [1] authors suggested the approach, which enables 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 bounds of the electrostatic analogy. In the weak anchoring case there are no topological defects and the director deformations  $\delta \mathbf{n}$  are small everywhere, so that the multipole expansion is valid at the particle surface too. That's why unknown amplitudes in (5) can be expressed through tensor characteristics of the particle's surface and orientation.

In the case of strong anchoring topological defects arise in the near-region, but outside the director deformations  $\delta \mathbf{n}$  are small. Therefore we can confine the particle and topological defects within the region that is called coat. This region contains all strong deformations of the director field. Outside the coat director deformations are small  $\delta \mathbf{n} \ll 1$ . The size of the coat has been estimated in [13] with help of the electrostatic analogy and it was shown to be few micrometers from the particle's surface. It is in a qualitative agreement with the result of [6]  $l = 0.26R$  for the distance between the droplet's surface and the hyperbolic hedgehog, when the radius  $R$  of the droplet is about ten micrometers. The symmetry of the coat is equivalent to the broken symmetry of the director in the vicinity. For example, a droplet with equatorial disclination ring (Saturn-ring configuration) could be put into the coat which has a horizontal symmetry plane (see Fig. 1(a)) and a water droplet with a companion hyperbolic hedgehog could be confined into the coat, which does not have horizontal symmetry plane (see Fig. 1(b)). The anchoring energy on the surface of the coat is determined as the interaction energy between the nematic molecules over the surface and the molecules under the surface of the coat. Phenomenologically it can be written in the Rapini-Papoular form, but it is natural, that the anchoring strength  $W_c$  becomes dependent on the point  $\mathbf{s}$  on the surface of the coat. The shape and the surface distribution  $W_c(\mathbf{s})$  determine the symmetry of the coat which is identical to the symmetry of the real director distribution in the near-region. Obviously, it is necessary to know the real director field in every point to find  $W_c(\mathbf{s})$  and shape of the coat exactly, that is hardly achieved. Instead of finding exact solutions of the EL equations, we show that the problem can be effectively solved in terms of some unknown tensors which characterize the surface of the coat. For this purpose we introduce the surface energy in the form

$$F_{cs} = \sum_p \oint dS W_c(\mathbf{s}) (v(\mathbf{s}) \cdot \mathbf{n}(\mathbf{s}))^2 \quad (6)$$



instead of (1). Such substitution allows us consider the director deformations  $\delta \mathbf{n}$  small everywhere in the space. All defects are hidden now inside the coat.

This makes it possible to find the interaction potential between ordinary the coats. In the one-constant approximation ( $K_{11} = K_{22} = K_{33} = K$ ) it has the form [7]:

$$U(\mathbf{R}) = -\frac{\alpha_{3m}\alpha_{3m}}{4\pi KR} + \beta_{3ms}\beta_{3ms'}(\mathbf{k}_s \cdot \nabla)(\mathbf{k}_{s'} \cdot \nabla) \frac{1}{4\pi KR} - \gamma_{3mst}\gamma_{3ms't'}(\mathbf{k}_s \cdot \nabla)(\mathbf{k}_{s'} \cdot \nabla)(\mathbf{k}_t \cdot \nabla)(\mathbf{k}_{t'} \cdot \nabla) \frac{1}{4\pi KR} \quad (7)$$

where the summation on the repeating indices is made,  $(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$  is the local basis rigidly bound with each particle and tensors  $\alpha, \beta, \gamma$  are defined as

$$\begin{aligned} \alpha_{kl} &= 2 \oint d\sigma W_c(\mathbf{s}) v_k(\mathbf{s}) v_l(\mathbf{s}), \\ \beta_{klm} &= 2 \oint d\sigma W_c(\mathbf{s}) v_k(\mathbf{s}) v_l(\mathbf{s}) \rho_m(\mathbf{s}), \\ \gamma_{klmn} &= \oint d\sigma W_c(\mathbf{s}) v_k(\mathbf{s}) v_l(\mathbf{s}) \rho_m(\mathbf{s}) \rho_n(\mathbf{s}). \end{aligned} \quad (8)$$

The integration is over the surface of the coat. The symmetry of these tensors contains all data about broken symmetry of the director field in the vicinity 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 asymptotics of *Ansatzes* solutions.

Expression (7) shows that the Coulomb attraction is present in general case. Let us consider the nature of this term. It is determined by the components  $\alpha_{13}$  and  $\alpha_{23}$ ,  $\alpha_{13} = 2 \oint d\sigma W_c(\mathbf{s}) v_1(\mathbf{s}) v_3(\mathbf{s})$ . It is obvious, that, if there is a horizontal symmetry plane, then the both components are zero. The presence of the symmetry plane  $XZ$  makes  $\alpha_{23} = 0$  and  $YZ$  makes  $\alpha_{13} = 0$ . So we come to the *significant conclusion*: the breaking of both horizontal and vertical mirror symmetry leads to the  $\alpha_{13} \neq 0$  or  $\alpha_{23} \neq 0$  and thus to the Coulomb attraction

$$U = -Q^2/R \quad (9)$$

where the role of charge plays geometrical factor  $Q = \sqrt{(\alpha_{13}^2 + \alpha_{23}^2)/4\pi K}$ . For the long cylinder which makes angle  $\vartheta$  with the director it is

$$Q = \frac{dL|W \sin 2\vartheta|}{4} \sqrt{\frac{\pi}{K}} \quad (10)$$

So Coulomb interaction appears 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 breaks mirror symmetry in a horizontal plane and in one vertical plane. In other terms, it exists, when there is a nonzero torque moment  $\Gamma$  acting to the particle by the nematic [19]. In the absence of the  $\Gamma$  it is absent.

Now we want to examine thoroughly the second term, which represents the dipole-dipole interaction. It takes the form

$$U_{dd} = \beta_{3ms}\beta_{3ms'} \frac{(3(\mathbf{k}_s \cdot \mathbf{R})(\mathbf{k}_{s'} \cdot \mathbf{R}) - \delta_{ss'})}{4^3} \quad (11)$$

Symmetry of the tensor  $\beta$  reflects symmetry of the director and defines dipole type of the interaction. Let us consider components  $\beta_{31s} = 2 \oint d\sigma W_c(\mathbf{s}) v_1 v_3 \rho_s$ . If the director distribution has horizontal symmetry plane, then for every point  $\mathbf{v}$  exists the mirror image, for which  $v_3$  changes the sign whereas  $v_1, \rho_1, \rho_2$  remain the same. Therefore  $\beta_{311} = \beta_{312} = 0$ . If *horizontal* symmetry plane is broken then  $\beta_{311}$  and  $\beta_{312}$  non equal zero and are measures of skewness. For particles with rotational symmetry like in the case of water droplets  $\beta_{311} = \beta_{322}$ , and dipole part takes the form:

$$U_{dd} = \frac{\beta_{311}^2}{4\pi K R^3} (1 - 3\cos^2(\theta)) \quad (12)$$

From comparison with similar expression in [6] we clearly see that  $\beta_{311}$  is proportional to the  $z$  component of the dipole moment  $p_z$ :

$$\beta_{311} = -4\pi K p_z$$

In general case such *conclusion* can be formulated: if the director distribution near the particle has three perpendicular planes (one of which is horizontal), then all  $\beta_{3ms}$  are zero and only quadrupole interaction remains, which dies off as  $R^{-5}$ .

For axial-symmetric director distribution around particles  $\gamma_{3131}$ , and  $\gamma_{3232}$  are equal and the quadrupole interaction takes the form

$$U_{qq} = \frac{3\gamma_{3131}^2}{2\pi K R^5} (3 - 30\cos^2\theta + 35\cos^4\theta)$$

where  $\cos\theta = \mathbf{nR}/R$ .

### 3. THE POSSIBLE ROLE OF COULOMB TERM IN 2D HEXAGONAL STRUCTURE

In [9] authors observed stable hexagonal structure formed by glycerol droplets in a nematic layer with hybrid boundary conditions. At the boundary

with air the LC molecules are oriented homeotropically, while at the boundary with glycerol their orientation is planar. At the top of the cell glycerol droplets are hanged because of Vandervaals interaction with interface LC-air. Drops introduce additional distortions in the director field that although unknown exactly but anyway produce elastic forces which should contain some combination of three possible terms in (7). Resulting elastic forces lead to the formation of 2D hexagonal structure in the distribution of glycerol droplets. This stable configuration should be formed both by attractive and repulsive forces. Moreover these forces should be isotropic in the plane of the structure. Good candidate for isotropic attractive force may be Coulomb-like term which arises when non-zero torque moment is applied to the droplet by nematic. Although this is unlikely for one particle (stable configuration corresponds to the zero torque moment) but may be profitable for many particles. Some energy is necessary to deflect a particle from its ground state. But this energy can be compensated by interaction energy with other particles which is negative and is proportional to the number of neighbors. So structure transition may occur under some critical concentration with spontaneous breaking of the local director symmetry and appearing of the elastic charge. The resulting Coulomb-like attraction can provide for 2D hexagonal structure because it is isotropic and stronger than any other elastic potential. Repulsive part of interaction is supplied with either dipole or quadrupole potential. This idea requires further confirmation or denial that can based mainly on the investigation of real distribution of the director field in the vicinity of particles.

#### 4. BEHAVIOR OF POLYMER DROPS NEAR STRONG DIRECTOR DEFORMATIONS [17]

In the paper [17] authors reported that polymer drops with planar anchoring in nematic liquid crystal assemble in the vicinity of strong deformations. We propose approach that enables to describe behavior of arbitrary particles in curved director field. We suppose that the director field is not homogenous because of the global boundary conditions, and that the director deformation length is much more, than the size of particles. So here we do not consider the deformations of the director, that are caused by particles itself. The anchoring strength on the surface of the coat is written

$$F_{surface} = \oint d\sigma W_c(\mathbf{s})(\mathbf{v}(\mathbf{s}) \cdot \mathbf{n}(\mathbf{s}))^2 \quad (13)$$

In as much as the global deformations scale is large, we can express the director on the surface  $\mathbf{n}(\mathbf{s})$  through the director in the center of mass of the coat

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

$\rho$  is the vector 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, if there is no coat in that place. Then the anchoring energy takes the form

$$F_{surface} = F_0 + F_1 + F_2$$

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

$$F_1 = 2 \oint d\sigma W_c(\mathbf{s}) (\mathbf{v} \cdot \mathbf{n}_0) (\rho \nabla) (\mathbf{v} \cdot \mathbf{n}_0)$$

$$F_2 = \oint d\sigma W_c(\mathbf{s}) [(\mathbf{v} \cdot \mathbf{n}_0) (\rho \nabla)^2 (\mathbf{v} \cdot \mathbf{n}_0) + ((\rho \nabla) (\mathbf{v} \cdot \mathbf{n}_0))^2]$$

The first item  $F_0$  is responsible for the orientation of the particle with respect to the director, whereas the second and the third items describes the behaviour of the whole particle in the curved director. All the scalars can be represented in the basis  $(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)$  like  $\mathbf{v} \cdot \mathbf{n}_0 = (\mathbf{v} \mathbf{k}_l) (\mathbf{n}_0 \mathbf{k}_l) = v_l n_l$ ,  $(\rho \nabla) = \rho_s (\mathbf{k}_s \nabla) = \rho_s \partial_s$ . Then the result is

$$F_1 = \beta_{l\mu s} n_l \partial_s n_\mu \quad (14)$$

$$F_2 = \gamma_{l\mu st} \partial_s (n_l \partial_t n_\mu) \quad (15)$$

These are the general expressions which describe movement of particles in the curved director field. The particle will move in the way to minimise the sum of  $F_1$  and  $F_2$ . The first term describes the behaviour of the coat with dipolar symmetry in the curved director and the second one describe the behaviour of the coat with quadrupolar symmetry in the curved director. Polymer drops with anchoring have two boojums on the surface so that no symmetry plane is broken and  $\beta_{l\mu s} = 0$ . As a result only  $F_2$  term remains. In as much as polymer drop with two boojums has azimuthal symmetry only  $\gamma_{1313} \neq 0$  remains and

$$F_2 = \gamma_{1313} (\mathbf{n} \nabla) \text{div} \mathbf{n} \quad (16)$$

For planar anchoring  $W > 0$  and  $\gamma_{1313} > 0$ . Let's consider particle in the radial hedgehog  $\mathbf{n} = \mathbf{R}/R$  on the distance  $R$  from the center. Then  $F_2 = -\gamma_{1313} 2/R^2 < 0$  and particle moves toward the center. So that parti-

cles with planar anchoring move toward places with high splay deformations that explains results of the [17].

## 5. CONCLUSION

In this paper we have examined theoretically elastic interactions in nematic colloids with help of general considerations concerning local symmetry of the director field in the vicinity of colloid particles. The one more distinctive feature of the nematic colloids in the Coulomb attraction between them, when the nonzero torque moment is applied to them by the nematic. We propose quantitative explanation of 2D hexagonal lattice of glycerol droplets in hybrid nematic cell in which this attraction plays crucial role. General expressions for the energy of arbitrary colloid particles in the curved director field are shown. Experimental result of the attraction of polymer droplets with planar anchoring toward strong director deformations is given.

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