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The director field distribution with the strongly pinned alignment in nematic structures at the polymer surface

Alexander M. Parshin[a,b]*, Victor Y. Zyryanov[a] and Vasily F. Shabanov[a]

[a]*Kirensky Institute of Physics, Krasnoyarsk Scientific Center, Siberian Branch of Russian Academy of Sciences, Krasnoyarsk, Russia;* [b]*Department of Energy, Siberian Federal University, Krasnoyarsk, Russia*

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We investigate orientational nematic liquid crystal structures that form at the polymer surface due to adsorption, their stability against the schlieren texture and features of point and linear singularities in them. We demonstrate that surface disclination lines, which also occur due to molecular adsorption, can lead to azimuthal anchoring of a bulk layer and make the singularities with the charge $m = \pm 1$ stable. We consider the director field distribution in a nematic layer as a result of the combination of two aligning factors, which are the linear disclination that tend to impose uniform planar ordering and point defects that impart the radial configuration to the director. With the use of the calculation, we analyse the energies of elastic distortions of the director field for the radial–planar, radial–homeotropic and radial–radial configurations, depending on the nematic layer thickness.

Keywords: nematic; polymer surface; adsorption; disclination; orientational order; director configuration

1. Introduction

Liquid crystals (LCs), due to the tendency of their molecules to align parallel to one another by their long axes, create point or linear singularities at the surface with degenerate boundary conditions if there are no easy orientation axes at this surface.[1] At present, much attention is focused on the topological study of singularities as reference objects of the field theory and physics of condensed matter.[2] A classical nematic LC structure is the schlieren texture.[3] Observations in crossed polarisers show the presence of point defects with the charge $m = \pm 1/2$ or ± 1, which corresponds to the number of extinction bands, in this texture.[4] The use of variational principles together with the topological methods makes it possible to investigate not only singularities but also their stability and the distribution of a LC director field around disclinations, which is important for comparative analysis of the investigated objects. In particular, in [5], minimizing the free energy of nematics at the degenerate surfaces, the equations were derived that yield the main director field configurations and their stability against m. Using this approach, it was established that all the LC structures related to linear singularities with the charge $m = \pm 1$ are unstable and near them the director 'escapes along the third dimension'.[6] Therefore, in the schlieren texture centres with four extinction bands are spread and represent not linear but point singularities. At the same time, structures with the singularities $m = \pm 1/2$ are stable and represent linear disclinations observed end-on in cross polarisers of a microscope.[2]

As it has been shown in [7], the conditions specified at the surface that bounds the LC layer can significantly affect structure and type of the singularities. Depending on the extrapolation length,[1] which is unambiguously related to the polar energy of surface nematic LC anchoring, one can obtain a Neel wall that extends throughout the bulk LC layer or a surface disclination line (SDL), which represents a similar wall closed at the surface. If we cover the schlieren texture with a glass of a specified uniform planar orientation, then inverse walls,[8] which are also similar to the Neel walls,[9] can be extended into the bulk of the LC.

On the other hand, at the smooth surfaces that bound the LC the azimuthal anchoring of the director can also take place.[10] This effect is pronounced at polymer surfaces where one or several easy orientation directions, which depend on structure and symmetry of polymer chains, can occur due to the intermolecular interactions.[11] Moreover, as we showed previously,[12] the use of special physicochemical treatment of polymer surfaces allows obtaining LC structures with the strong azimuthal anchoring, degenerate boundary conditions and an infinite set of easy orientation axes. In [12], we considered the technology for preparing nematic domain structures at the polycarbonate (PC) surface, described characteristic textures of these structures, and reported the results of temperature investigations.

*Corresponding author. Email: parshin@iph.krasn.ru

In this study, we investigate nematic orientational radial-planar (R^+P), radial-homeotropic (RH), and radial-radial (RR) structures formed at the PC surface, estimate their topology, study the singularities, and discuss the stability of disclinations and the distribution of the LC director vector field with the use of a minimization technique.

2. Experimental

Samples were prepared using the technique described in detail in [12]. A 5CB or MBBA nematic layer was deposited onto the PC film surface with a residual solvent. After a laps of time $t \sim 10$ min, structures with singularities formed on the polymer film. The upper boundary of the sample was either free or covered with a glass. The glass substrate surface was treated with lecithin to impose the homeotropic orientation or rubbed to impose the planar one. The wedge-shaped LC layer was formed by placing a teflon spacer on one side of a cell. Nematic orientational structures were investigated with the use of a polarizing optical microscope.

3. Results

3.1 Textures

Observations in cross polarisers show that a thin ($d < 20$ μm) LC layer has the classical schlieren texture with domain formations against its background (see Figure 1). In the schlieren texture, boundaries, which can be pronounced and broad, are observed between domains of the director field. The domain texture is formed during the growth from a nucleus to the size bound by neighbouring domains ($R \approx 85$ μm) and gradually replaces the entire schlieren texture.

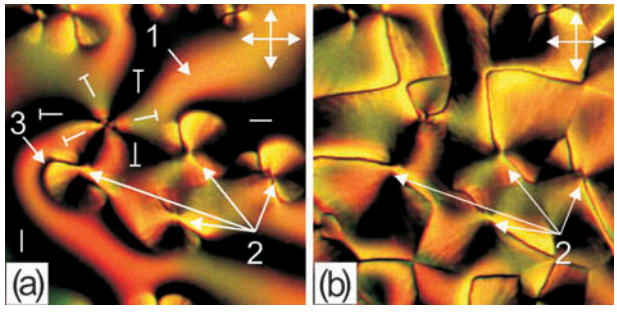

Figure 1. (colour online) Microphotograph of the formation of domain textures 2 with pronounced boundaries 3 against the background of schlieren texture 1 in the nematic layer: (a) during the formation and (b) after its finish. The upper boundary of the sample is free. Orientation of LC molecules is shown by short lines (planar alignment) and 'nails' (oblique orientation). The arrows show the direction of light oscillation between crossed polarisers in all the figures.

The domain texture observed in crossed polarisers strongly depends on nematic layer thickness d. At $d > 10$ μm, an individual domain looks like a disk with two conic extinction bands (see Figure 2a). A disclination line passing through a domain is clearly seen against the background of light bands and gradually vanishes depending on how close it is to the central parts of dark extinction bands, as shown in Figures 2b and c. For $d < 10$ μm, a dark sector with the beginning at the domain periphery arises near the disclination line (see Figure 2d). The sector acquires a sharp contour when the disclination line makes the angle $30° < \alpha < 60°$ with the polariser directions, as shown in Figure 2e. In addition, the sector becomes pronounced as the disclination line approaches the direction of one of the polarisers and the LC layer thickness decreases down to 5 μm (see Figure 2g), as well as at the disclination line orientation $30° < \alpha < 60°$, as shown in Figures 2h and i. In the latter case, the domain looks like a disk with four extinction bands. Sometimes, domains with a point disclination with interference rings around it can be observed at the center (see Figure 2f). In this case, the disclination line is hardly seen. It should be noted that domains with such a point disclination are met quite rarely and arise independent of thickness d.

In the wedge-like cell, domains are coloured in accordance with interference colours of the LC layer observed in white light in crossed polarisers (Table 1). It can be seen in the table that as the nematic layer thickness changes from 10 to 1 μm, the interference spectra lower from the third to first order. For $d > 10$ μm, the colours stop changing and for $d < 1$ μm the wedge remains black.

3.2 Disclinations

The disclination lines are always sharp. Their real width is $2b \approx 2$ μm. With decreasing nematic layer thickness in the wedge cell with the cover glass rubbed to impose the planar orientation, the disclination lines become increasingly ordered. The mutual orientation of the lines in a domain ensemble becomes noticeable starting with $d \approx 10$ μm (see Figure 3a); in thinner layers, the singularities are aligned almost parallel to one another and perpendicular to the rubbing direction as shown in Figure 3b.

4. Discussion

4.1 Structures

The schlieren texture core in Figure 1 has the topological charge m = +1, which was determined from the

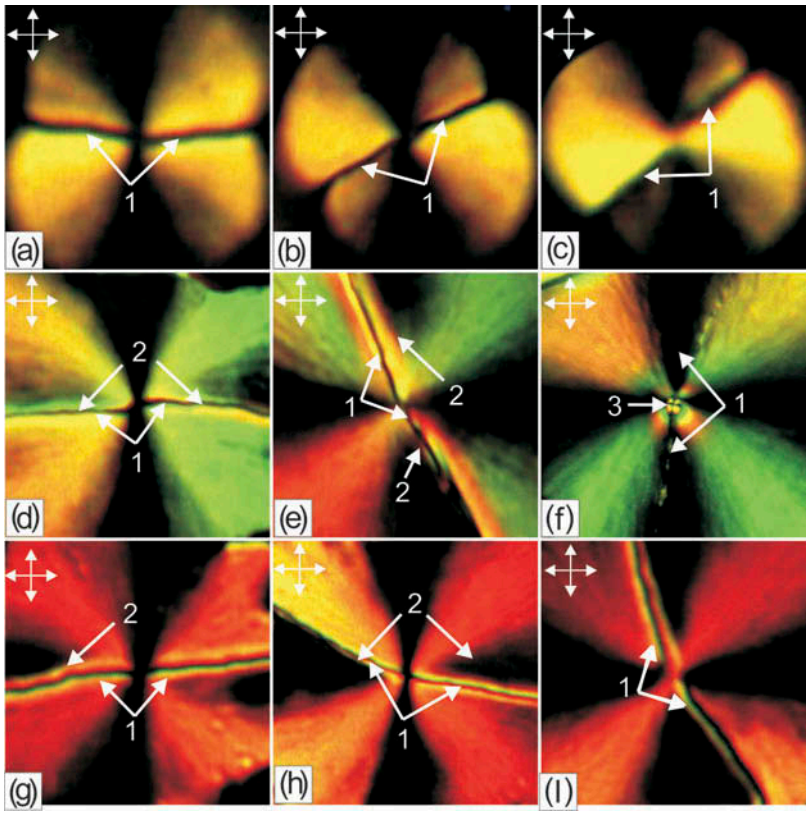

Figure 2. (colour online) Microphotographs of nematic domains formed on the PC films with the residual solvent in a wedge cell with rubbed cover glass at LC layer thickness of (a–c) 20, (d–f) 6, and (g–i) 4 μm. 1 is disclination lines, 2 is a dark sector and 3 is the core.

Table 1. Interference colours of the 5CB layer in the wedge-shaped cell with rubbed cover glass at LC layer.

Wedge thickness, μm	Wedge colour	Spectral order
10	Yellow	
9	Green	
8	Blue	Third
7.5	Purple	
7	Red	
6.5	Orange	
5.5	Green	Second
5	Blue	
4	Purple-red	
3.5	Orange	
3	Yellow	
2	Green	First
1	Black	

relation m = *N*/4, where *N* is the number of dark bands in the texture and the sign is determined by the fact that upon rotation of the texture in cross polarisers the bands rotate in the opposite direction. The director field distribution around the singularity corresponds to the radial (R) structure. As we previously established,[12] the domain texture core also has the charge m = +1. The topological charges located near each other should repulse and the director field lines should smoothly bend between the charges, which can be observed in nematic colloids.[13] The presence of sharp boundaries (walls) between the textures shown in Figure 1 is related to the features of the formation of the radial LC structure at the PC surface upon evaporation of the residual solvent from the polymer film. In this process, LC molecules form the radial structure, gradually aligning around the nucleating centre outgoing from polymer chains,[12] as it happens at their interaction with grains in colloidal solutions. Adsorption of nematic molecules at the polymer surface leads to the strongly pinned alignment, i.e., the Cheng–Boyd effect,[10] and the wall formation. For simplicity, hereinafter we call the domain structures presented in Figure 1 the *adsorption structures*. In contrast to the schlieren texture that arises at the degenerate surface, the adsorption radial structure without easy orientation axes should be considered as a structure formed at the surface with the continuous set of easy orientation axes [1] that outgo from the domain centre.

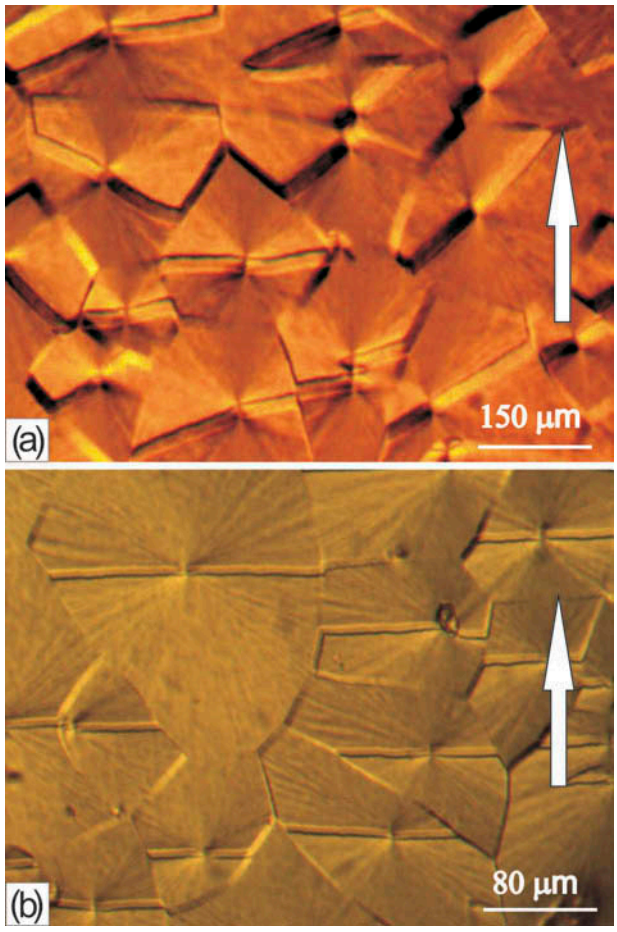

Figure 3. (colour online) Orientation of disclination lines in a wedge cell with a rubbed cover glass in dependence on the nematic layer thickness: (a) $d \approx 10$ μm and (b) $d \approx 2$ μm. The rubbing directions are shown by arrows.

4.2 *Disclinations*

Thorough observations of the domain growth showed that the R and SDL structures form with the simultaneous pinning of two competing alignments (Figure 4a), the first being related to the director field distribution relative to the surface disclination points and the second, to the SDLs.[12] This process results in the formation of the strongly pinned structure at the surface. In this case, the SDL screens the aligning effect of a defect core of the radial structure in the polar direction, thus preventing the director 'escapes along the third dimension', which usually occurs in the radial structure without SDLs [14] or in the structure with the low-quality SDLs (see Figure 2f). At the boundaries of the transition between the uniform SDL part and the radial configuration, the direction of director $\boldsymbol{n}$ sharply changes. Therefore, the SDLs can be seen in Figures 2 and 3 as broad bands with sharp boundary lines. Schematic of

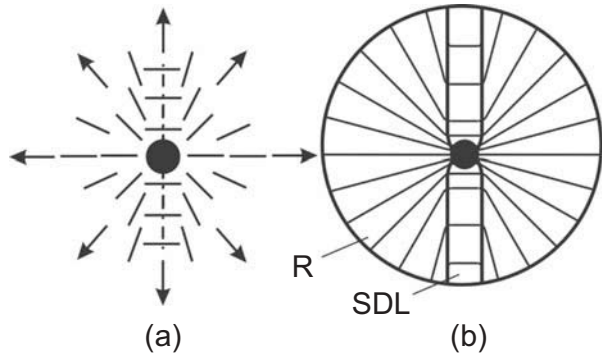

Figure 4. Schematic of nematic director field distribution at the PC surface: (a) during domain growth (is shown by arrows) and (b) after formation of the R and SDL structures.

director field distribution at the surface of the nematic domain after formation of the R and SDL structures at the PC film is shown in Figure 4b. The SDL tends to form a wall that extends into the bulk of the LC layer. However, in the investigated structures, the formed wall does not extend into the bulk to an infinite distance. As the distance from the surface is increased, adsorption forces weaker hold the strongly pinned alignment of the surface configuration and the director field distribution becomes dependent on the equilibrium state between the uniform orientation from the SDLs and the radial structure.

We may assume that at certain distance ξ from the surface, the radial structure smoothly transforms to the uniform planar structure, since the latter should have a lower free energy due to the absence of strains. Such a hypothetic representation of the structure in which the radial configuration with the SDLs is concentrated at the surface and there is a transition layer to the uniform orientation was experimentally proved. In Figure 2, the textures observed in thick LC layers have singularities with the charge $m = +1/2$ and transform to the textures with the charge $m = +1$ in thin layers. As was demonstrated in [15], such a transformation is forbidden by topological laws. The concept of the equilibrium existence of non-uniform and uniform structures in an LC layer readily resolves the contradiction. In thick LC layers, the planar orientation pinned at the SDL, being superimposed on the radial configuration, transforms the optical pattern such as in the texture with the charge $m = +1$ two extinction bands are observed. At the same time, in a thick sample with rubbed substrate influence the outer surface is not sufficient to orient SDL, which is observed as a broad dark line for all positions of crossed polarisers (Figure 2) due to the 'waveguide effect' of Mauguin[1]. The transition to

the texture with four extinction bands in thin layers occurs due to the change in the LC director orientation in a uniform structure, which leads to the transformation of the optical pattern. In this case, as shown in Figure 2, the interference effects appear in form of alternating colour bands arising in strongly deformed surface layer of nematic. Since the transformation observed as a dark sector and developing from the texture periphery has a sharp boundary with the SDLs, we may assume that the structure strain is caused by the break of the equilibrium between the wide radial field of the director and the uniform orientation caused by the narrow SDL. The effect of the SDL ordering by the rubbed outer surface via molecules of the bulk LC layer and the dependence of the degree of ordering of singularities on the nematic layer thickness, as shown in Figure 3, confirm our assumptions.

4.3 Director field distribution in the structures

For any configuration of director $\boldsymbol{n}$, the free energy of the LC in volume V in the approximation of equal elastic constants K is [1]

$$F = \frac{1}{2}K\int_V \left[(\mathrm{div}\,\boldsymbol{n})^2 + (\mathrm{rot}\,\boldsymbol{n})^2\right]dV \qquad (1)$$

For the R^+P configuration that corresponds to the transformation of the radial structure with the SDLs (R^+) to the uniform planar structure P (Figure 5a), we write in the cylindrical coordinates

$$n_\rho = -\cos\psi \quad n_\varphi = \cos\psi \quad n_z = 0 \qquad (2)$$

Angle ψ of the deviation of molecules from polar radius ρ is zero ($z = 0$, $\psi = 0$) at the surface and

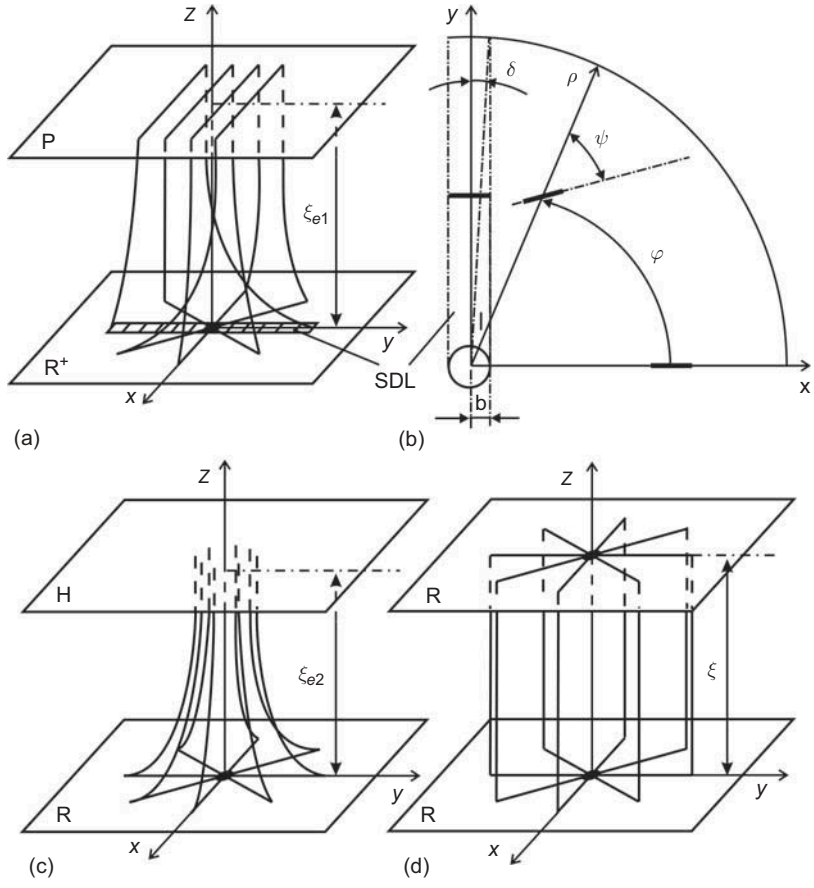

Figure 5. Distribution of nematic director $\boldsymbol{n}$ at the transformation of (a) the radial structure with SDLs R^+ to the uniform planar P structure at distance ξ_{e1} from the polymer film surface, (b) parameters for the calculation in cylindrical coordinates in the xOy plane, (c) the radial R structure to the uniform homeotropic H structure at distance ξ_{e2} from the polymer film surface, and (d) the radial–radial RR configuration under the condition of its stability over LC at distance ξ.

$\psi = \varphi$ at the equilibrium distance $z = \xi_{e1}$. We put $\psi = (1–\exp\ z/\xi)\varphi$, under the assumption that the locally distorted orientation decay approximately exponentially with distance from the surface.[16] Substituting director field distribution (2) in expression (1) and integrating over $l \le \rho \le R$, $0 \le \varphi \le 2\pi - 4\delta$, $0 \le z \le \infty$, we obtain

$$F_1 = \frac{1}{2}K\left[\pi \ln\left(\frac{R}{b}\right)\xi + \left(\frac{\pi^3}{12} - \frac{\pi^2}{4} + 1\right)\frac{R^2}{2}\frac{1}{\xi} + \left(\frac{\pi^2}{4} - \pi\right)R\right] + F_l, \tag{3}$$

where R is the domain radius, b is the half of SDL width and F_l is the SDL energy that contains the term calculated taking into account the δ characterising the angular size SDL and the geometric part related to the size $l = b/\cos\varphi$. Because the width of the SDL in the radial structure during the domain growth at the PC surface is greater than or equal to the radius core r [12] we used b instead of r in Equation (3). Parameters for the calculation in cylindrical coordinates in the xOy plane are shown in Figure 5b.

The equilibrium value of ξ is obtained by minimization of (3):

$$\xi_{e1} = \left(\frac{\left(\frac{\pi^3}{12} - \frac{\pi^2}{4} + 1\right)\frac{R^2}{2}}{\pi \ln\left(\frac{R}{b}\right)}\right)^{1/2}. \tag{4}$$

Following the above, ξ_{e1} is the distance at which the SDL ceases to feel the external influence. This is the length in which the radial configuration becomes a homogeneous planar structure.

To answer the question why the configuration R^+P remains even in the case when the outer LC surface is free or covered with a glass of a specified homeotropic orientation H, we consider the RH configuration (Figure 5c), which would be implemented without SDLs. For this configuration, we write

$$n_\rho = -\sin\theta \quad n_\varphi = 0 \quad n_z = \cos\theta, \tag{5}$$

where θ is the angle between director $\boldsymbol{n}$ and axis z. For the hybrid-oriented film with the degenerate boundary conditions,[14] $\theta = 0$ at $z = 0$, $\theta = \pi/2$ at the distance ξ_{e2}. For simplicity, we put $\theta = (1 - z/\xi)\ \pi/2$ at an arbitrary value of z. Substituting Equation (5) into Equation (1) and integrating over $r \le \rho \le R$, $0 \le \theta \le 2\pi$, $0 \le z \le \xi$, we obtain

$$F_2 = \frac{1}{2}K\left[\pi \ln\left(\frac{R}{r}\right)\xi + \frac{\pi^3}{4}R^2\frac{1}{\xi} - \pi^2 R\right] + F_c, \tag{6}$$

where r is the core radius and F_c is the core energy, which contains the geometric part related to r.

The first terms in Equations (3) and (6) are related to the R configuration in the surface layer of a nematic; the second ones, to the strains in the transition layer from R to the homogeneous planar or homeotropic ordering in volume; and the third ones, to splay cancelling.[14] Minimizing Equation (6), we obtain the equilibrium value of ξ

$$\xi_{e2} = \left(\frac{\frac{\pi^3}{4}R^2}{\pi \ln\left(\frac{R}{r}\right)}\right)^{1/2}. \tag{7}$$

If the R structure were stable, i.e., the director does not 'escapes along the third dimension' near the SDL, then the distribution of the director field would correspond to the configuration shown in Figure 5d. After substitution

$$n_\rho = -1 \quad n_\varphi = 0 \quad n_z = 0 \tag{8}$$

into expression (1) and integration over $r \le \rho \le R$, $0 \le \varphi \le 2\pi$, $0 \le z \le \xi$ one obtains

$$F_3 = \frac{1}{2}K\left[2\pi \ln\left(\frac{R}{r}\right)\xi\right] + F_c. \tag{9}$$

According to the experimental data, which yield $R \approx 85$ μm and $b \approx 2$ μm, we obtain $\xi_{e1} \approx 17$ μm and $\xi_{e2} \approx 63.3$ μm for the R^+P and RH configurations, respectively. Figure 6 shows the dependences of free energies from the length ξ for the R^+P, RH, and RR configurations. The values of F_1, F_2 and F_3 were plotted using expressions (3), (6), and (9) at $K = 6\ 10^{-12}$ N for 5CB [17] with regard to the values of F_1 and F_c. The calculated and plotted value of ξ_{e1} corresponds to the minimum free energy of the R^+P configuration, which was pronounced the best in the LC layers with $d \sim 30$ μm used in the experiment. The RH configuration could be effectively observed in the nematic layers with $d \sim 60 - 70$ μm near ξ_{e2}. However, since F_1 is smaller than F_2 over the entire range of realizable thicknesses ξ, we always observed the R^+P configuration instead of RH. Energy F_3 linearly depends on the nematic layer thickness and is minimum at $\xi \to 0$. The dependences F_3 (ξ) and F_1 (ξ) have a common point at $\xi_c \approx 10$ μm. As was demonstrated experimentally, this is the starting point of gradual variations in the R^+P structure, i.e., the increasing manifestation of the R structure at $\xi \to 0$. At small LC layer thicknesses ($\xi \sim b$) the transformation of R^+P to RR can be accompanied by the variation in the equilibrium director field

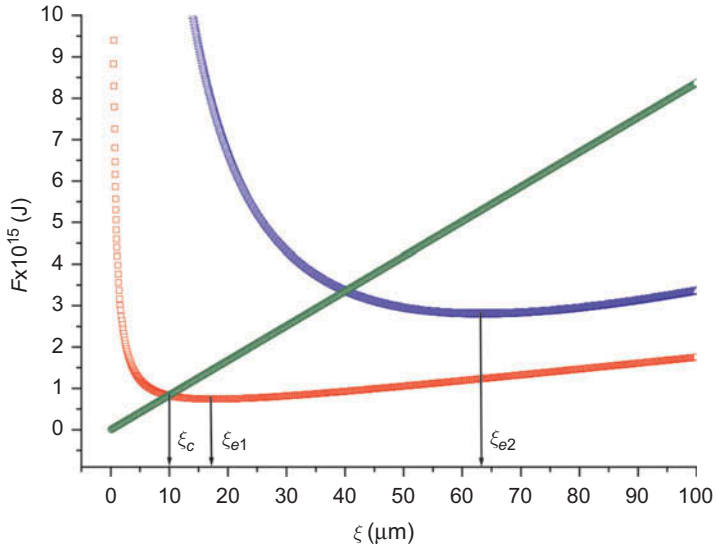

Figure 6. (colour online) Dependences of free energies F_1, F_2 and F_3 for the R^+P, RH and RR configurations; ξ_{e1} and ξ_{e2} are the distances from the PC surface that correspond to the minimum free energies of the R^+P and RH configurations and ξ_c is the critical distance from the surface that corresponds to the beginning of the structural transformation $R^+P \rightarrow RR$ (RH).

distribution in a wall between the SDL and R structure. This may lead to weakening of the orienting effect and 'escaping of the nematic director in the third dimension' near the core in the RR structure and transition to the RH structure. Regardless of whether the transformation $R^+P \rightarrow RR$ or $R^+P \rightarrow RR \rightarrow RH$ occurs, the optical observations visualise the transition of the singularity with the charge $m = 1/2$ to the singularity with the charge $m = 1$, as shown in Figure 2.

One can assume that the inequality of the splay, torsion and bend constants K_{11}, K_{22} and K_{33} could affect the stability of the structures under study, thus preventing the director 'escapes along the third dimension'. However, according to [18], the flat disclination with $m = 1$ is stable in the structure with director lines arranged in concentric circles only when $K_{22} > 2\ K_{33}$, $K_{11} > K_{33}$ what is not feasible for most classic LC. At this, the radial configuration is characterised by the disclination unstable under all values of the elastic modulus. The studies of the disclination stability with different m-values [6,14] prove the relevant application of the one-constant elastic approximation.

5. Conclusions

We studied the director field distribution in the nematic LC structures formed at the surface of PC films with a residual solvent. We considered the stability of radial formations obtained due to molecular adsorption of nematic molecules at the polymer surface, which leads to the strongly pinned alignment, i.e., the Cheng–Boyd effect. Using optical methods, we determined the structures of SDLs that form simultaneously with the radial nematic configurations. The SDLs are not projections onto the plane of inverse or Neel walls known to date but form by the polymer surface itself. In addition, adsorbed LC molecules favour pinning of the SDLs on a polymer film and serve as orienting elements for bulk nematic layers. We established that during the formation of a structure at the polymer surface, the strongly pinned alignment occurs; as the distance from the surface grows, the adsorption forces weaker hold the surface configuration, so the director field distribution becomes increasingly uniform. Using the calculation approach, we compared the energies of elastic distortions of the director field for the radial–planar, radial–homeotropic and radial–radial configurations, depending on the nematic layer thickness. We demonstrated that the radial–planar orientation with the uniform alignment element is energetically more favourable than the other configurations; therefore, it can be readily implemented and be more stable as compared with the other structures. We determined the critical distance for the experimentally observed radial–planar configuration. The presented adsorption structures can be considered as point singularities with the topological charge $m = +1$ and the strongly pinned force lines.

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