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Domain structures in ferroelectric liquid crystals

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Domain structures called steric (or striped) domains are investigated in ferroelectric liquid crystals (FLCs). The domain lines are aligned perpendicular to the smectic layers with a period proportional to the FLC cell thickness *L*. Neither FLC polarization nor helix pitch affects the domain pattern. Striped domains disappear in thick cells ($L > 8$–$10\,\mu$m) and transform to a new type of surface domain without any preferred orientation in thin samples ($L < 1{\cdot}0\,\mu$m). The existence and structure of the steric domains define properties such as contrast and bistability of the FLC cell. We propose a qualitative explanation of this domain pattern and compare its characteristics with ferroelectric domains discovered earlier by Beresnev *et al.* in FLCs.

1. Introduction

Since the discovery of surface stabilized structures by Clark and Lagerwall [1] electrooptical effects in ferroelectric liquid crystals (FLCs) remain the subject of growing interest. The nature of different modulated domain patterns [2–8], which affect optical bistability [2] and the average contrast of the FLC cell [2, 7] remain unclarified in FLC electrooptics.

In this paper we pay main attention to the origin of the so-called steric domains, which appear as a regular striped texture perpendicular to the smectic layers [2–5]. According to us the appearance of the steric domains is defined only by the geometry of the FLC director and layer deformation by the electrooptical effect. This is different from the ferroelectric domains discovered by Beresnev *et al.* [6–8] with the striped orientation parallel to the FLC layers and the period h_f inversely proportional to the square of the FLC spontaneous polarization P_s

$$h_f \propto P_s^{-2}. \qquad (1)$$

In sufficiently thin FOL layers ($L < 1{\cdot}0\,\mu$m) the orientation of the domains is not fixed and the surface defects play an important role. This type of steric domain we call surface domains.

Here, we compare different domain patterns in FLCs. A possible explanation of steric domain appearance is proposed, based on our experimental investigations.

2. Experimental

The FLC used in the experiment is a mixture based on alkylalkoxypyrimidines with the following phase sequence

$$\mathrm{X} - 10^\circ\mathrm{C}\ \mathrm{S}_\mathrm{C}^*\ 60^\circ\mathrm{C}\ \mathrm{S}_\mathrm{A}\ 75^\circ\mathrm{C}\ \mathrm{N}\ 78^\circ\mathrm{C}\ \mathrm{I}.$$

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By varying the concentration of the two chiral dopants we changed the value of the spontaneous polarization P_s within the range of 20–80 nC cm^{-2} and the helix pitch from 0·5 μm to 10 μm. At $T = 22°C$ the equilibrium value of the helix pitch is $p_0 = 7$ μm.

The cells were made from two glass plates, coated with a layer of ITO. PVA films of 500 Å thickness were used to orient the two substrates, which were rubbed in opposite directions. The cell was filled with liquid crystal in the isotropic phase under the action of capillary forces and slowly cooled down to room temperature.

The electrooptical experiments were carried out with a polarizing microscope. To measure the characteristic angles of the structure, we rotated the sample, placed between crossed polarizers, in the polarizer's plane. The contrast was defined by measuring the intensities of the light signals in a photomultiplier. In the experiment we used square pulses of 10 V μm^{-1} electric field with a frequency of 10 Hz.

When we cooled the cell down into the FLC phase zig-zag defects and defect lines in the form of needles appeared, recognized by Shao *et al.* [5] as parallel zig-zag walls. When the electric field $E > 3$–4 V μm^{-1} is applied, the needles elongate, covering the whole surface area of the FLC cell. During this process each needle transforms into two stripes. The FLC director continues to reorient with increasing field as it could be observed by the change of the stripe colour under the polarizing microscope. Finally, the smectic layers orient themselves perpendicular to the substrates, and we observe a black and white domain pattern similar to those mentioned in [2–5] (see figure 1). The domain lines are aligned parallel to the rubbing direction and coincide approximately with the FLC layer normal.

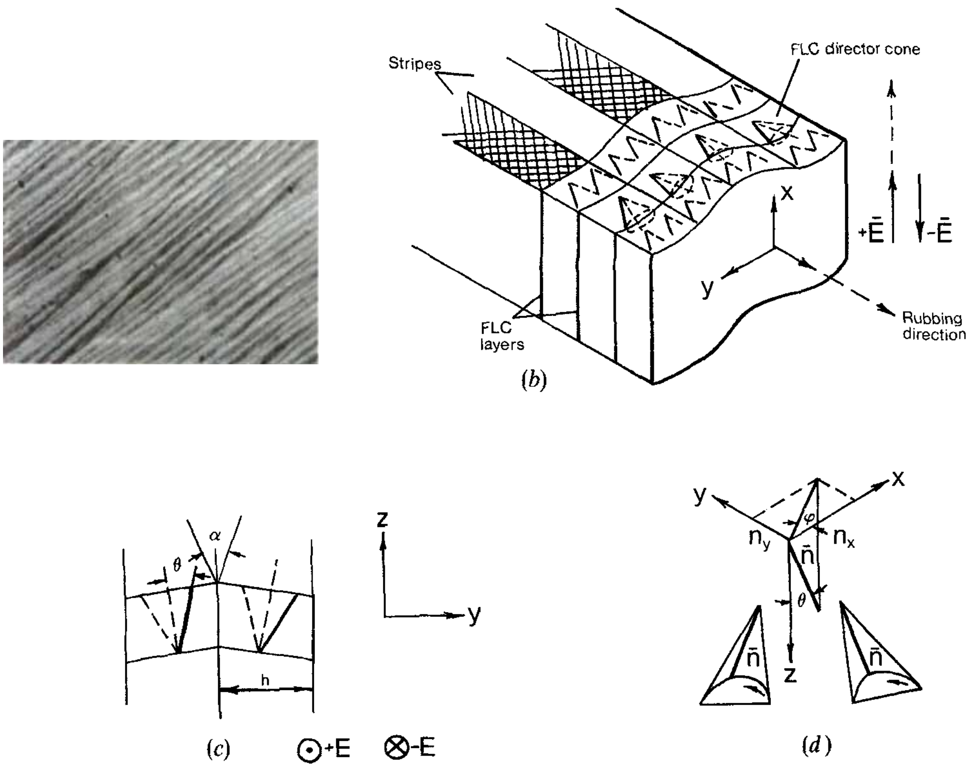

Figure 1. Steric domains in FLC layers.

An investigation of the dark and bright states in each stripe by placing the FLC cell between two crossed polarizers and by subsequent rotation of the cell in the polarizer's plane clearly shows, that the FLC layers undergo a periodic curvature deformation in the plane of the substrates (see figure 1). Thus the orientations of the director cones in the two neighbouring stripes (see figures 1 (*b*) and (*c*)) form a certain angle α with respect to each other.

When the electric field E changes its sign the director in each stripe undergoes half a cone rotation (see figures 1 (*b*) and (*c*)). As it follows from figure 1 the smaller α angles correspond to the more uniform FLC orientation, resulting in the higher contrast between $(+E)$ and $(-E)$ states of the FLC cell placed between crossed polarizers. This explains our earlier observed data, showing that the contrast improves by increasing the thickness of the FLC cell (see figure 2). Thus we have shown that the curvature deformation becomes weaker for thicker FLC layers and the stripes tend to disappear (see figure 2).

The period of the domains, h, for the thin cells grows approximately proportional to the layer thickness L and saturates in sufficiently thick cells ($L > 6$–$7\,\mu$m) (see figure 3). The value of h does not really depend either on the size of the FLC polarization P_s (see figure 3) or on the value of the equilibrium pitch (in our experiments we change the pitch from $16\,\mu$m to $5\,\mu$m by varying the concentration of the chiral dopants).

It should be noted that the results of our experiments do not really confirm the relations $h = L$ and $\alpha = 2\delta$ obtained in [5] (see figure 3). (δ is the angle of the smectic layer with respect to the normal to the substrates.)

As mentioned previously, for very thin cells ($L < 1{\cdot}5\,\mu$m) the striped lines do not follow any preferred orientation, resulting in surface domains (see figure 4). This domain pattern depends considerably on the surface treatment and in particular on surface defects. The nature of this domain pattern is not yet clear.

For thick FLC cells ($L > 8$–$10\,\mu$m) the stripes become unstable to external actions, such as heating up to the transition temperature to the smectic A phase or application of a high frequency AC electric field.

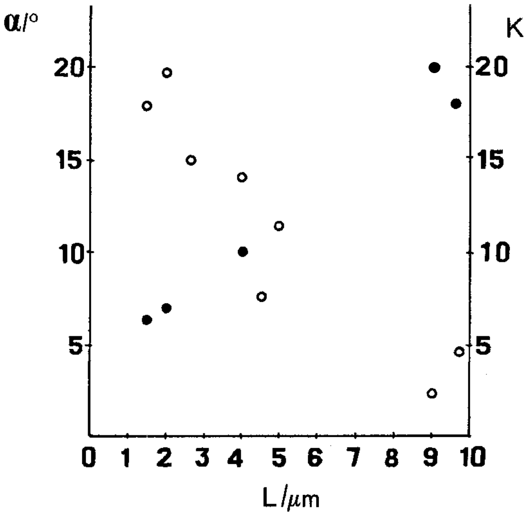

Figure 2. The angle α (○) between two layer normals in neighbouring stripes and the contrast ratio K (●) of $(+E)$ and $(-E)$ states of the FLC cell under crossed polarizers versus the layer thickness L.

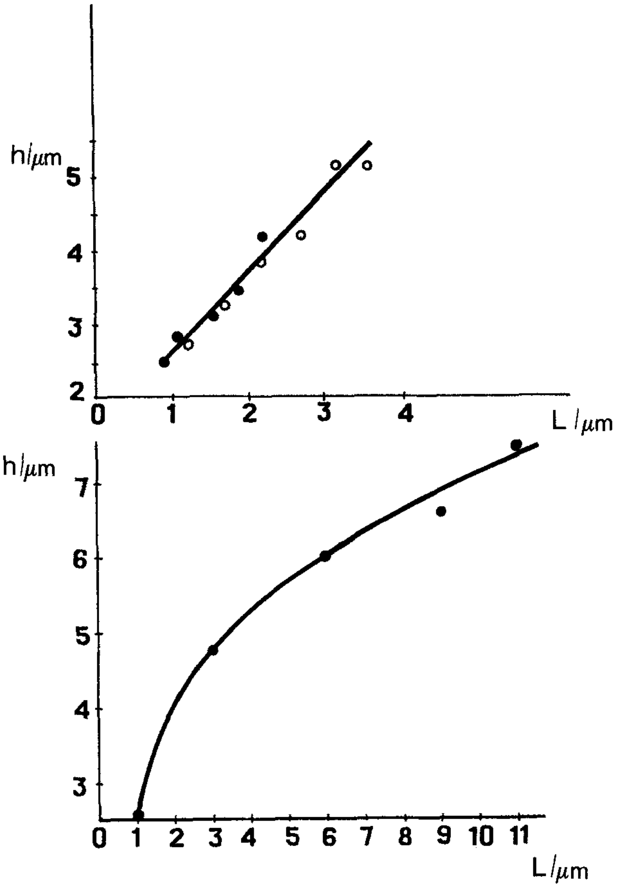

Figure 3. The stripe period h versus the FLC layer thickness. White circles correspond to a value of the spontaneous polarization $P_s = 37\,\mathrm{nC\,cm^{-2}}$ and black circles a value of $P_s = 58\,\mathrm{nC\,cm^{-2}}$. The other mixture parameters remain the same.

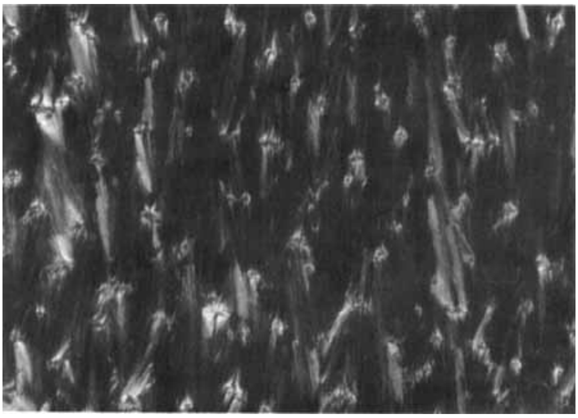

Figure 4. Surface domains in thin FLC cells.

3. Discussion

We propose the following qualitative explanation of the striped domain pattern (see figure 1). We should take into account two possible effects. The first takes place when, due to the interaction of the electric field E with the FLC spontaneous polarization P_s, the director n rotates up to a certain azimuthal angle φ along the cone surface, giving the director components

$$\mathbf{n}=(n_x, n_y, n_z)=(\sin\theta\cos\varphi, \sin\theta, \sin\varphi, \cos\theta), \quad \varphi=\varphi(x,y). \tag{2}$$

This linear electrooptical effect provides a linear uniform director component n_y along the y axis, and the dependence of n_y along the layer thickness (x axis) is defined according to the boundary condition on the substrates of the electrooptical cell. As the direction of the stripes along the z axis is perpendicular to the smectic layers, all the variables do not depend on z. The second effect takes place due to the so-called azimuthal electrohydrodynamic instability [9, 10], which becomes possible in a FLC for the previously mentioned geometry and when action of the electric field E is along the x axis. This instability results in a modulation of a small director disturbance,

$$n_y(x,y)\sim\sin\theta\varphi(x,y)\sim\sin\theta\exp(iqy)\varphi_0(x), \tag{3}$$

along the y axis as well as, in addition, the modulated disturbances of the electric potential U and the flow velocity V in the smectic plane (x, y). This quadratic electrooptical effect takes place mainly due to the existence of the charge carriers and apparent non-uniformity of the space electric potential.

If the smectic planes are not distorted the two effects oppose each other, resulting in the domination of the uniform director rotation along the cone surface. However the possibility of the second effect becomes evident, if we take into account one more degree of freedom, namely the curvature deformation of the smectic layers. In this case the smectic layer displacement along the z axis proves to be modulated both along the y axis and the layer thickness. The uniform azimuthal rotation of the FLC director is due to the linear electrooptical effect and there is, therefore, an added sign alternating director deviation

$$n'_y\sim\exp(iqy) \tag{4}$$

in the yz plane due to the periodical curvature deformation of the smectic layers.

The coupling of the variables u_z and n'_y is linear and takes place as a result of the existence of the following invariants

$$bn_z(n'_y\partial u_z/\partial y+n'_x\partial u_z/\partial x)+c/2((\partial u_z/\partial y)^2+(\partial u_z/\partial x)^2) \tag{5}$$

in the FLC free energy. This expression shows that the modulations n'_y and u_z are opposite in phase, i.e.

$$\left.\begin{aligned} n'_y\sim\sin qy, \quad u_z\sim\cos qy \\ \partial u_z/\partial y\sim-(b/c)n'_y. \end{aligned}\right\} \tag{6}$$

From the approximations of [9, 10] we can take into account the same equations for the variable of n'_y by a simple renormalization of one term

$$(\varepsilon_a E^2/4\pi)n'_y\Rightarrow((\varepsilon_a E^2/4\pi)-b^2/c)n'_y, \tag{7}$$

where ε_a is the FLC dielectric anisotropy, while the other equations remain the same. Therefore to describe the FLC electrohydrodynamic instability accompanied by the appearance of the smectic layer curvature mode we may qualitatively use the results of

[9, 10] and evaluate the effective threshold. In particular, the threshold voltage u_{th} and the period of the structure h must be dependent on the FLC director tilt with respect to the substrates $\theta_0 \sim \arcsin(n_y) \sim \theta \sin\varphi$, anisotropies of conductivity σ_a, viscosity, dielectric anisotropy ε_a, etc. The period of the structure h should be proportional to the layer thickness L.

It is known [9], that if the voltage exceeds the threshold, the formation of the curvature deformation in a pile of smectic planes results in the appearance of a number of dislocations due to local ruptures of the smectic layers and in an inevitable increase of smectic layer number. A defect smectic structure contains a series of dislocations along the domain boundaries parallel to the z axis and is stored by the sample, as in usual crystals. Consequently switching the external electric field E off does not lead to the disappearance of the given domain structure.

4. Conclusions

In conclusion let us compare the main specific features of various domain patterns exhibited by FLCs.

(i) Ferroelectric domains discovered by Beresnev *et al.* [6–8]. These domains appear in thick ($L > 10\,\mu$m) FLC cells with high spontaneous polarization. The domains occur as black and white stripes parallel to the smectic layers. The period of the domains h_f considerably decreases with rising spontaneous polarization of the FLC cell ($h_f \sim P_s^{-2}$) but does not depend on cell thickness. The physical model of these domains is proposed on the basis of the interaction of the disclination loops formed in FLCs with a space charge which exists mainly due to the high spontaneous polarization P_s [8].

(ii) Steric or striped domains [2–5]. According to our experiments neither spontaneous polarization nor helical pitch are important in this case. The domains appear perpendicular to the smectic layers with a period proportional (but not equal) to the layer thickness (see figure 1). Striped domains prove to be a periodic curvature deformation of smectic layers in the FLC structure. We consider steric domains to be a kind of electrohydrodynamic instability similar to those existing in nematics with oblique boundary tilt of the director [10]. In sufficiently thick cells ($L > 8$–$10\,\mu$m) steric domains become unstable and disappear.

(iii) Surface domains arise in thin ($L < 1{\cdot}0\,\mu$m) cells with no preferred orientation (see figure 4). We believe that the surface defects play an important role in this case. Further experiments are underway.

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