PRAMANA — journal of physics
© Indian Academy of Sciences
Vol. 61, No. 2
August 2003
pp. 385–393

Bidimensional distortion in ferroelectric liquid crystals with strong anchoring in bookshelf geometry

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Abstract. In the last decade, it has been experimentally found that in certain conditions a periodic domain pattern arises in ferroelectric liquid crystals (FLC) in bookshelf geometry. Such a periodic texture appears after switching-off an external electric field, even in strong anchoring conditions. It has a static character and is bidimensional, being dependent on directions normal to both the smectic planes and the cell plates. Here a new model explaining this phenomenon is proposed, valid in the case of FLC with strong anchoring. The model is based on the coupling between the spontaneous polarization field in the first semi-period of its modulation along the cell plates and the same field in the second semi-period. This coupling competes with the FLC medium elasticity and is trapped by the anchoring. According to our model, in the ferroelectric state the biperiodic texture is favored by increasing the values of spontaneous polarization. The critical value of the spontaneous polarization for which the undeformed state becomes unstable is found. It is shown to be proportional to the square root of the ratio between the FLC elastic constant and the cell thickness. Moreover, it is inversely proportional to the sinus of the pre-tilt angle with respect to the cell walls.

Keywords. Ferroelectric liquid crystals; spontaneous polarization; flexoelectricity; anchoring; elasticity.

PACS Nos 61.30.-v; 61.30.Gd

1. Introduction

Ferroelectric liquid crystals (FLC) are known to be an excellent medium for devices transducing electric signals into optical pattern suitable for electro-optical sensors, like optic valves, displays, and monitors with high contrast and large viewing angle. For this purpose, the linear electro-optic effect induced by the coupling of the spontaneous polarization $\mathbf{P}_\mathrm{S}$ with an external applied electric field $\mathbf{E}$ is of main importance [1]. The effect can be characterized by bistability and optical memory in the surface-stabilized bookshelf [2,3] or quasi-bookshelf [4,5] geometry. In this typical configuration, all smectic layers exhibit the same orientation of spontaneous polarization, and consequently the whole cell spontaneous polarization is simply the product of the contribution of each smectic layer and the layer numbers (e.g. $\mathscr{N}\mathbf{P}_\mathrm{S}$-up). In this ordered state the whole FLC cell polarization

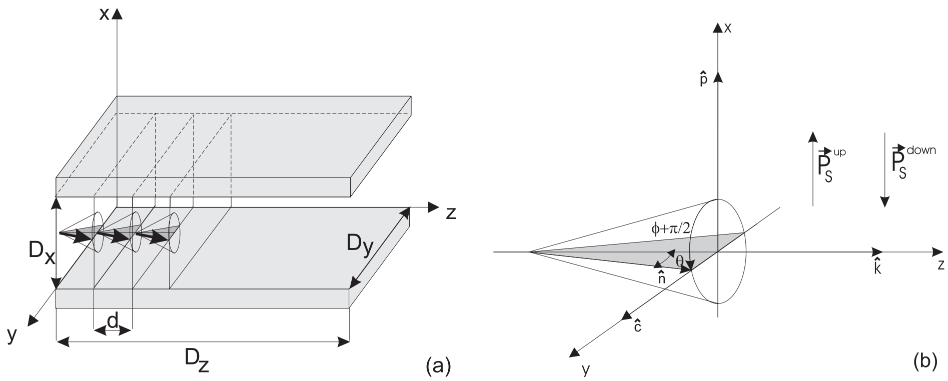

Figure 1. Smectic C* cell in bookshelf geometry (**a**). The cell volume is $D_x D_y D_z$, where D_x is the cell thickness. The smectic cone is considered with the axis parallel to z-axis (**b**). The $(\hat{\mathbf{n}}, \hat{\mathbf{c}})$-director azimuth is $\phi + \pi/2$, measured from x-axis normal to the cell plates. The $\hat{\mathbf{n}}$-director polar angle is ϑ. In principle, for pre-tilt angle equal to zero, the spontaneous polarization $\mathbf{P}_{\mathrm{S}}$ can be up or down, being parallel to $\hat{\mathbf{p}} = \hat{\mathbf{c}} \times \hat{\mathbf{k}}$.

under applied electric field can move towards another stable state (e.g. $\mathscr{N}\mathbf{P}_{\mathrm{S}}$-down) [3]. In the last decade, static modulated patterns were observed in such systems in ferroelectric phase [6–13] in certain conditions, for instance after switching off a DC field, applied to a uniform surface-stabilized cell. Regular periodic domains occur in this case as static stripes, oriented either parallel [8–12] or perpendicular [6,9,13] to the normal to the smectic layers, which lies in a plane parallel to the cell walls (see figure 1). Up to now several qualitative models describe the possible role played in the phenomenon by the occurrence of a periodic disclination array [14] and by flexoelectricity and charge conduction [15,16]. In the present paper a new model is provided, explaining the phenomenon in FLC state via another mechanism, which describes the biperiodic distortion as the result of the spontaneous polarization modulated by the distortion coupled with the local modulated electric field due to the polarization itself. This coupling competes with the FLC medium elasticity [16] and is driven by the surfaces characterized by strong anchoring. The flexoelectric contribution can renormalize the elastic constants [17,18]. In the present work, the threshold of the spontaneous polarization for which the undeformed state becomes unstable is obtained.

2. Theory

We consider a liquid crystalline material exhibiting ferroelectric phase organized in bookshelf geometry. In figure 1a the cell reference frame $[x,y,z]$ has the x-axis normal and the y-axis parallel to the cell plates, and the z-axis is normal to the smectic planes. $\hat{\mathbf{n}}$ is the molecular director and $\hat{\mathbf{c}}$ the tilt director. ϑ is the tilt angle characterizing the cone of the smectic phase, and $(\phi + \pi/2)$ is the azimuth of both $\hat{\mathbf{c}}$- and $\hat{\mathbf{n}}$-director orientations measured from x-axis: its variation $\delta\phi$ describes a soft distortion. The spontaneous polarization lies in the smectic layer parallel to the xy-plane, ϕ being its azimuth. It is given by

$$\mathbf{P}_{\mathrm{S}} = \mathscr{P}_{\mathrm{S}} \hat{\mathbf{p}}, \tag{1}$$

where $\hat{\mathbf{p}} = \hat{\mathbf{c}} \times \hat{\mathbf{k}}$ ($\hat{\mathbf{k}}$ is the unit vector parallel to the z-axis), and $\mathscr{P}_S$ is the modulus of the FLC spontaneous polarization.

In figure 1b is shown the particular case in which a unidirectional planar initial configuration of the FLC cell is considered, with $\hat{\mathbf{c}}$ parallel to y-axis and $\mathbf{P}_S$ parallel to x-axis ($\phi = \phi_0 = 0$). Generally, ϕ_0 can assume any value according to the possible presence of a pre-tilt angle with respect to the cell plates. By applying an external electric field $\mathbf{E}_{\text{ext}}$ along x, the linear coupling with the polarization $\mathbf{P}_S$ induces the azimuthal rotation $(\phi - \phi_0)$. After switching off the field, the $\hat{\mathbf{c}}$-distribution can relax to a configuration different from the previous one. The new configuration is periodic in two directions (2D periodicity), with the director deformation parallel to the xz-plane and invariant along the y-axis (see figures 2a and 2b). The bidimensional texture can be described by the azimuth angle of the spontaneous polarization

$$\phi = \phi_0 + \varphi(x,z), \tag{2}$$

where ϕ_0 is the initial azimuth angle and φ the wave amplitude assumed to be very small ($|\varphi| \ll 1$ rad). For small distortions the tilt director can be linearized as

$$\hat{\mathbf{c}} = -\sin\phi\ \hat{\imath} + \cos\phi\ \hat{\jmath} \approx -(\sin\phi_0 + \varphi\cos\phi_0)\ \hat{\imath} + (\cos\phi_0 - \varphi\sin\phi_0)\ \hat{\jmath}. \tag{3}$$

The unit vector $\hat{\mathbf{p}}$ is given by

$$\hat{\mathbf{p}} = \hat{\mathbf{c}} \times \hat{\mathbf{k}} \equiv \cos\phi\,\hat{\imath} + \sin\phi\,\hat{\jmath} \approx (\cos\phi_0 - \varphi\sin\phi_0)\,\hat{\imath} + (\sin\phi_0 + \varphi\cos\phi_0)\,\hat{\jmath}. \tag{4}$$

In what follows we will consider an FLC with isotropic dielectric tensor ε. In the frame of the mean field theory the dielectric displacement $\mathbf{D}$ is given by

$$\mathbf{D} = \varepsilon\mathbf{E} + 4\pi\mathbf{P}, \tag{5}$$

where $\mathbf{P}$ in the presence of mixed splay-bend $\hat{\mathbf{n}}$ – distortions has not only the spontaneous $\mathbf{P}_S$ – but also the flexoelectric contribution:

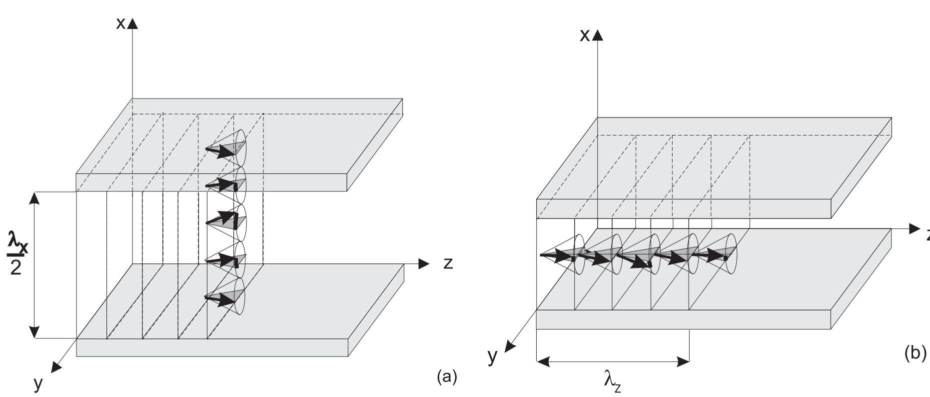

Figure 2. Biperiodic deformation with wave vectors $q_x = 2\pi/\lambda_x = \pi/D_x$ in the direction normal to the cell walls, when the anchoring is strong (**a**), and $q_z = 2\pi/\lambda_z$ in the direction normal to the smectic layers (**b**).

$$\mathbf{P} = \mathbf{P}_\mathrm{S} + \mathbf{P}_\mathrm{flexo}. \qquad (6)$$

The flexoelectric polarization is given by [15]:

$$\begin{aligned}\mathbf{P}_\mathrm{flexo} &= d_3(\nabla\cdot\hat{\mathbf{c}})\hat{\mathbf{c}} - d_4(\hat{\mathbf{c}}\cdot\nabla\times\hat{\mathbf{c}})\hat{\mathbf{p}} + d_6(\hat{\mathbf{k}}\cdot\nabla\times\hat{\mathbf{c}})\hat{\mathbf{p}} + d_9(\nabla\cdot\hat{\mathbf{c}})\hat{\mathbf{k}} \\ &= -d_3\cos\phi_0\varphi_x\hat{\mathbf{c}} + \left(d_4\varphi_z + d_6\sin\phi_0\varphi_x\right)\hat{\mathbf{p}} - d_9\cos\phi_0\varphi_x\hat{\mathbf{k}},\end{aligned} \qquad (7)$$

d_i being the flexoelectric moduli for smectic C* media. In eq. (7) φ_x and φ_z are the partial derivatives of $\varphi(x,z)$ with respect to x and z, respectively.

In the absence of free charges, $\nabla\cdot\mathbf{D} = 0$, which implies that $\mathbf{D} = \mathrm{const.}$ within the cell. In particular, it can be assumed that $\mathbf{D} = 0$. Hence, eq. (5) can be written as

$$0 = \varepsilon\mathbf{E} + 4\pi\left(\mathbf{P}_\mathrm{S} + \mathbf{P}_\mathrm{flexo}\right), \qquad (8)$$

from which the local electric field can be expressed in terms of the flexoelectric and of the spontaneous polarization as follows:

$$\mathbf{E} = -\frac{4\pi}{\varepsilon}\left(\mathbf{P}_\mathrm{S} + \mathbf{P}_\mathrm{flexo}\right). \qquad (9)$$

In the case of fixed voltage for the virtual deformations, the thermodynamical potential per unit volume f, whose minima define the stable states of the system, can be written as [19,20]

$$f = f_\mathrm{elast}(\varphi,\varphi_x,\varphi_z) + f_\mathrm{diel} + f_\mathrm{coupl}(\varphi_x,\varphi_z) + f_\mathrm{C}, \qquad (10)$$

where [3,17]

$$f_\mathrm{elast} = \frac{1}{2}\left(B_1\sin^2\phi_0 + B_2\cos^2\phi_0\right)\varphi_x^2 + \frac{1}{2}B_3\varphi_z^2 - B_{13}\sin\phi_0\varphi_x\varphi_z. \qquad (11)$$

Let us remind that the smectic elastic constants B_1, B_2, B_3 are relevant to the pure bend, splay, twist respectively of the tilt director $\hat{\mathbf{c}}$. In fact, pure distortion of $\hat{\mathbf{c}}$ can imply mixed distortions of the director $\hat{\mathbf{n}}$: for example both B_1, B_3 comprise mixed symmetrical twist–bend deformations of $\hat{\mathbf{n}}$, whereas B_2 implies only $\hat{\mathbf{n}}$-splay distortion. Moreover, the mixed B_{13} elastic constant refers to the coupling between in-plane (xz)-bend and twist across smectic layers, affecting the $\hat{\mathbf{n}}$-deformations [3]. The new elastic constants read:

$$\begin{aligned}B_1 &= \sin^2\vartheta[K_{22}\cos^2\vartheta + K_{33}\sin^2\vartheta], \\ B_2 &= K_{11}\sin^2\vartheta, \\ B_3 &= \sin^2\vartheta[K_{22}\sin^2\vartheta + K_{33}\cos^2\vartheta], \\ B_{13} &= \frac{1}{2}(K_{33} - K_{22})\sin^2\vartheta\sin 2\vartheta.\end{aligned} \qquad (12)$$

The other terms in eq. (10) read as:

$$f_\mathrm{diel} = -\frac{1}{8\pi}\varepsilon E^2 \equiv -\frac{2\pi}{\varepsilon}\left(\mathscr{P}_\mathrm{S}^2 + P_\mathrm{flexo}^2 + 2\mathbf{P}_\mathrm{S}\cdot\mathbf{P}_\mathrm{flexo}\right), \qquad (13)$$

$$f_\mathrm{coupl} = -\mathbf{E}\cdot\mathbf{P} = \frac{4\pi}{\varepsilon}\left(\mathscr{P}_\mathrm{S}^2 + P_\mathrm{flexo}^2 + 2\mathbf{P}_\mathrm{S}\cdot\mathbf{P}_\mathrm{flexo}\right). \qquad (14)$$

The Coulomb interaction free energy density f_C between two spontaneous polarization charges dQ, dQ' corresponding to two infinitesimal areas dA and dA' separated at the same surface is given by

$$f_C = \frac{1}{2}\int\int \frac{\mathrm{d}Q\mathrm{d}Q'}{|z'-z|D_xD_yD_z}, \tag{15}$$

where D_i is the cell size in the i-direction. In the presence of a periodic distortion, dQ and dQ' read as:

$$\begin{cases} \mathrm{d}Q = (\sigma_0 + \delta\sigma)\mathrm{d}A \\ \mathrm{d}Q' = (\sigma_0 - \delta\sigma)\mathrm{d}A' \end{cases} \tag{16}$$

with $\delta\sigma > 0$, being the small variation of the surface charge density due to the modulation of the spontaneous polarization. Equation (15) due to Gauss' theorem becomes

$$f_C = \frac{1}{2}\int_0^{D_z}\int_0^{D_z} \frac{(\nabla\cdot\delta\mathbf{P}_S)(\nabla\cdot\delta\mathbf{P}'_S)}{|z'-z|D_z} D_xD_y\,\mathrm{d}z\,\mathrm{d}z', \tag{17}$$

where

$$\begin{cases} \delta\mathbf{P}_S = \mathscr{P}_S\varphi(-\sin\phi_0\hat{\imath} + \cos\phi_0\hat{\jmath}) \\ \delta\mathbf{P}'_S = \mathscr{P}_S\varphi(\sin\phi_0\hat{\imath} - \cos\phi_0\hat{\jmath}) \end{cases} \tag{18}$$

since the contribution f_{C_0} due to $\mathbf{P}_{S_0} = \mathscr{P}_S(\cos\phi_0\hat{\imath} + \sin\phi_0\hat{\jmath})$, being a constant can be neglected. Approximating the Delta Dirac function by $1/|z'-z|$, the integration of eq. (17) over the whole cell gives

$$f_C = -\frac{1}{2}(\nabla\cdot\delta\mathbf{P}_S)^2 D_xD_y, \tag{19}$$

which yields, taking into account eq. (18):

$$f_C = -\frac{1}{2}\mathscr{P}_S^2\sin^2\phi_0 D_xD_y\varphi_x^2. \tag{20}$$

By substituting eqs (13), (14) and (20) into eq. (10), straightforward calculations give

$$f = f_{\mathrm{elast}}(\phi,\phi_x,\phi_z) + \frac{2\pi}{\varepsilon}\left[\mathscr{P}_S^2 + P_{\mathrm{flexo}}^2 + 2\mathscr{P}_S\cdot P_{\mathrm{flexo}}\right] - \frac{1}{2}\mathscr{P}_S^2\sin^2\phi_0 D_xD_y\varphi_x^2, \tag{21}$$

where $\mathscr{P}_S^2$ in square brackets can be neglected since it is a constant. By using definition (1) for P_S and the result obtained in (7) for P_{flexo}, the thermodynamical potential becomes

$$\begin{aligned} f = {} & f_{\mathrm{elast}}(\phi,\phi_x,\phi_z) + \frac{2\pi}{\varepsilon}\left[(d_3^2+d_9^2)\cos^2\phi_0 + d_6^2\sin^2\phi_0\right]\varphi_x^2 \\ & + \frac{2\pi}{\varepsilon}d_4^2\varphi_z^2 + \frac{4\pi}{\varepsilon}d_4d_6\sin\phi_0\varphi_x\varphi_z \\ & + \frac{4\pi}{\varepsilon}\mathscr{P}_S[d_4\varphi_z + d_6\sin\phi_0\varphi_x] - \frac{1}{2}\mathscr{P}_S^2\sin^2\phi_0 D_xD_y\varphi_x^2. \end{aligned} \tag{22}$$

By means of the standard variational calculus, we obtain the following equilibrium equation:

$$\left(\frac{4\pi}{\varepsilon}d_4^2+B_3\right)\varphi_{zz}+2\left(\frac{4\pi}{\varepsilon}d_4d_6-B_{13}\right)\sin\phi_0\varphi_{xz}$$
$$+\left(\frac{4\pi}{\varepsilon}(d_3^2+d_9^2)\cos^2\phi_0+\frac{4\pi}{\varepsilon}d_6^2\sin^2\phi_0-\mathscr{P}_\mathrm{S}^2\sin^2\phi_0D_xD_y\right.$$
$$\left.+B_1\sin^2\phi_0+B_2\cos^2\phi_0\right)\varphi_{xx}=0, \tag{23}$$

where φ_{ij} means second-order partial derivative in the variables i and $j=x,z$. It is possible to simplify such differential equation taking into account that the flexoelectric moduli are present only to the second power, and the products $d_i d_j$ are expected to be of the order of 10^{-10} dyne, whereas the values of the main elastic constants for tilt director deformations are $B_i\simeq 10^{-7}$ dyne. Then, if the expected accuracy is of the order of 0.1%, it is possible to neglect the flexoelectric contributions.

The boundary conditions are

$$\varphi(x=-D_x/2,z)=\varphi(x=D_x/2,z)\equiv 0. \tag{24}$$

This means that the solution $\varphi(x,z)$ along the x-axis must be even and periodic with a period $\lambda_x=2D_x$:

$$\begin{cases}\varphi(x,z)=\varphi(-x,z)\\ \varphi(x+\lambda_x,z)=\varphi(x,z)\end{cases} \tag{25}$$

and periodic along the z-axis too:

$$\varphi(x,z+\lambda_z)=\varphi(x,z), \tag{26}$$

where λ_x and λ_z are the wavelengths of the biperiodic deformation along x- and z-axes, respectively. For the sake of simplicity, one elastic constant approximation is assumed. Hence, $B_1=B_2=B_3\equiv B$ and $B_{13}=0$, and the Euler Lagrange equation (eq. (23)) becomes

$$B\varphi_{zz}+(B-\mathscr{P}_\mathrm{S}^2\sin^2\phi_0D_xD_y)\varphi_{xx}=0, \tag{27}$$

which admits a solution of the type

$$\varphi(x,z)=\psi(z)\cos(q_xx). \tag{28}$$

From the boundary conditions we derive that the wave number $q_x=\pi/D_x$. By substituting eq. (28) in (27) the following second-order differential equation is deduced:

$$\psi_{zz}+\left(\frac{\mathscr{P}_\mathrm{S}^2\sin^2\phi_0D_xD_y}{B}-1\right)q_x^2\psi=0, \tag{29}$$

which has the periodic solution:

$$\psi=c_1\cos(q_zz)+c_2\sin(q_zz), \tag{30}$$

where $q_z = 2\pi/\lambda_z$ only if

$$\mathscr{P}_{\rm S}^2 \sin^2\phi_0 D_x D_y/B > 1. \tag{31}$$

In other words, the appearance of the biperiodic distortion is a *threshold phenomenon*, where the critical parameter is the spontaneous polarization $\mathscr{P}_{\rm S}$:

$$\mathscr{P}_{\rm S\,th} = \frac{1}{\sin\phi_0}\sqrt{\frac{B}{D_x D_y}}. \tag{32}$$

Only for values of the spontaneous polarization $\mathscr{P}_{\rm S} > \mathscr{P}_{\rm S\,th}$, q_z is real, and the biperiodic ratio $r = \lambda_z/\lambda_x = q_x/q_z$ is real as well. The biperiodic ratio turns out to be

$$r = \sqrt{\frac{\mathscr{P}_{\rm S}^2}{\mathscr{P}_{\rm S\,th}^2} - 1} \tag{33}$$

and the equilibrium equation admits the periodic solution along both x- and z-axes

$$\varphi(x,z) = \cos(q_x x)\left[c_1\cos(q_z z) + c_2\sin(q_z z)\right]. \tag{34}$$

The distortion amplitudes c_1 and c_2 can be determined by imposing the boundary conditions along the z-axis. In order to be a stable solution, eq. (34) must correspond to the minimum energy of the system. This can be determined by comparing the total free energy per unit period $\mathscr{F}$ corresponding to the deformed state with the energy of the undistorted configuration $\mathscr{F}_0 = 0$. The deformed state described by eq. (34) represents the stable configuration if

$$\mathscr{F} \leq 0. \tag{35}$$

The total free energy $\mathscr{F}$ per each z-wavelength for the deformed state is given by

$$\mathscr{F} = \int_0^{\lambda_z}\int_0^{\lambda_x/2} f\,{\rm d}x\,{\rm d}z. \tag{36}$$

By using (22) without the flexoelectric term, according to the previous approximation, we get

$$\mathscr{F} = \frac{\pi^2}{4}\frac{B}{r}(c_1^2 + c_2^2)\left[1 - \left(\frac{\mathscr{P}_{\rm S}^2}{\mathscr{P}_{\rm S\,th}^2} - 1\right)r^2\right]. \tag{37}$$

Let us underline that the linear coupling between spontaneous and flexoelectric polarizations in a half- and a whole period is obtained as identically zero. From eq. (37) $\mathscr{F} = 0$ means that the undistorted solution becomes *metastable* with respect to the distorted one. This happens for a spontaneous polarization $\mathscr{P}_{\rm S}$ greater than the critical value $\mathscr{P}_{\rm S\,th}$ given by (32). This simple but very important relation states that the critical value of the spontaneous polarization for the appearance of the biperiodic deformation in FLC cells strongly anchored in bookshelf geometry is proportional to the square root of the ratio between the smectic C* elastic constant B and the cell thickness D_x. Moreover, it is inversely proportional to $\sin\phi_0$, and in the limit of $\phi_0 \ll 1$ rad it is inversely proportional to the pre-tilt angle with respect to the cell plates ϕ_0. Usual values of the FLC elastic parameters ($B \sim 10^{-7}$ dyne) [23], together with a pre-tilt angle on the cell plates $\phi_0 = 1^\circ$ and a cell thickness $D_x = 2$ μm, give $\mathscr{P}_{\mathrm{S\,th}} \simeq 1.3$ nC/cm^2. This means that the presence of a small pre-tilt angle, according to the present model, is essential for ensuring the occurrence of the biperiodic distortion.

Many FLC compounds (as for instance the commercial DOBAMBC and HOBACPC) have spontaneous polarizations greater than the estimated threshold of the spontaneous polarization [3,21,22,24], thus promoting the periodic instability described here. It is possible to conclude that the deformed state corresponding to a bidimensional modulated texture consists of a threshold phenomenon mainly induced by the ferroelectric polarization. It is similar to the Frederiks transition, the spontaneous polarization acting as a source of instability of the system in the undeformed state, instead of an external applied electric or magnetic field. The main difference between the Frederiks transition and the effect described here is related to the fact that in the former case the induced deformation occurs in a plane and is one-dimensional, whereas in the latter case it involves all the 3D volume of the cell and is bidimensional.

3. Conclusions

The arising of periodic instability in FLC cells, experimentally found in the last decade by many authors, has been analyzed and a new simple model in the frame of continuum theory has been established. According to it, a soft mixed distortion of splay-, twist- and bend-type can spontaneously occur in homogeneous surface-stabilized cells ordered in bookshelf geometry, in the case of strong anchoring, when the external field is switched off. Such distortion is biperiodical, the two wave vectors having the direction normal to both the smectic plane, and the cell walls. It can be explained without invoking layer deformation. Our model describes such a biperiodic distortion essentially as a result of the competition between the elastic energy and the spontaneous polarization field in the first semi-period of its modulation along the cell plates, coupled with the same field in the second semi-period. Such competition is driven by the strong anchoring. The flexoelectric contribution just gives small corrections (within 0.1%) in this picture, renormalizing the elastic constants. Remarkably, the effect analyzed exhibits a threshold in the spontaneous polarization which is inversely proportional to the sinus of the pre-tilt angle with respect to the cell walls. All common FLC media have spontaneous polarization values definitely greater than the threshold, and consequently can exhibit the biperiodic instability for usual pre-tilt angles.

Acknowledgements

This work was partially supported by European Community in the frame of INCO Copernicus Concerted Action ‘Photocom’, under Contract No. IC15-CT98-0806. The support of TMR Network ‘SILC’ under contract No. FMRX-CT98-0209 is also gratefully acknowledged. Thanks are due to S Lagerwall and W Haase for their questions during the Conference. We are also indebted to Giovanni Barbero for very useful discussions.

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