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STRUCTURE OF A HYBRID ALIGNED CHOLESTERIC LIQUID CRYSTAL CELL

I. DOZOV and I. PENCHEV

*Institute of Solid State Physics, Bulgarian Academy of Sciences,
 Bvd. Lenin 72, Sofia, Bulgaria*

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Résumé.- La structure d'une cellule hybride d'un cristal liquide cholestérique est étudiée expérimentalement et théoriquement. Pour des petites épaisseurs la cellule présente une structure caractérisée par une déformation unidimensionnelle-structure uniforme (dans le plan xy). Cette structure est décrite théoriquement et des résultats numériques sont présentés et discutés. Pour des grandes épaisseurs de la cellule apparaît une déformation tridimensionnelle caractérisée par une modulation du champ du directeur dans le plan xy et par l'apparition de domaines. Une idée simple concernant cette déformation est présentée et discutée qualitativement.

Abstract.- The structure of a hybrid aligned cholesteric liquid crystal sample is investigated both experimentally and theoretically. At small thickness the cell presents a structure characterized by unidimensional deformation - uniform (in the xy plane) structure. This structure is described theoretically and numerical results are presented and discussed. At large thickness there appears a more complex three dimensional deformation characterized by a domain structure due to a modulation of the director field in the xy plane. A simple idea about the type of the deformation involved in this structure is proposed and qualitatively discussed.

In the recent years there has been an increased interest in the so called hybrid aligned nematic (HAN) structure [1,2]. This structure is characterized by a continuous splay-bend deformation due to the antagonistic orientation imposed on the two glass-liquid crystal interfaces - a homeotropic and a planar one respectively. In the present letter we consider a similar but more complex structure - a hybrid aligned cholesteric liquid crystal (HAC). The uniform HAC structure has been already discussed both theoretically [3] and from the point of view of a possible practical application [4], but up to now some interesting properties of this complex splay-bend-twist deformation are not discussed in the literature. Our theoretical discussion is based upon the experimental data for a HAC cell containing a long-pitch nematic-cholesteric mixture.

1. Experimental.

In order to observe the structure we use a 0.75 % (by weight) solution of cholesterin undecylat in methoxybenzylidene butylaniline (MBBA). The natural pitch of the mixture is $P = 30 \mu\text{m}$. A 10 mm long wedge-shaped cell with deposited indium-tin oxide electrodes on both glass plates provides a liquid crystal thickness gradient from 0 to $90 \mu\text{m}$ (Fig. 1). The lower plate is treated with

polyvinyl alcohol and rubbed to give a planar orientation along the x-axis. The upper plate is treated with a silane agent to induce a homeotropic alignment.

As in the HAN case a bend-splay deformation would arise in order to satisfy the boundary conditions imposed on our cell. Due to the intrinsic twist of the cholesteric and to the lack of any azimuthal torque on the homeotropic plate we expect that the equilibrium state would be twisted. The result is the splay-bend-twist deformation schematically represented in figure 1.

The sample is observed on the stage of a polarizing microscope. To measure the twist in the cell we use the wave guide property of the twisted structure. We send a light beam normal to the sample and polarized along the x-axis on the lower plate and we observe the polarization of the transmitted beam outside the sample. As we shall see in the next section the criterion for the wave guide regime is well satisfied in our sample except for a small region in the vicinity of the upper plate. Thus the rotation of the polarization through the sample is a good measure of the angle $\varphi_d = \varphi(d)$ near the homeotropic side.

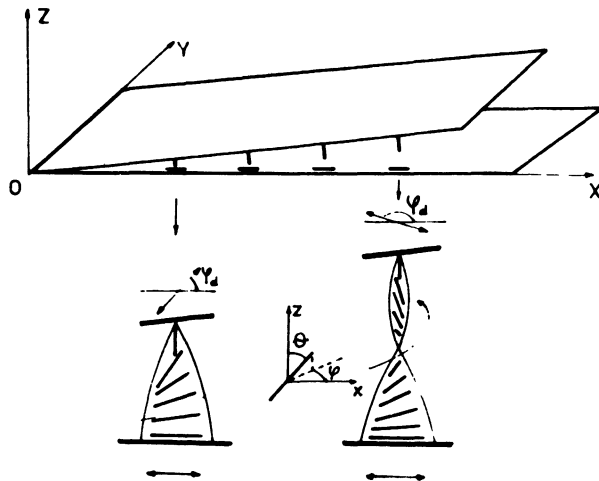


Fig. 1.- Geometry of the cell and splay-bend-twist deformation of the hybrid aligned cholesteric.

After filling the sample we observe domains delimited by disclination lines. We identify these domains as the expected splay-bend-twist twins similar to the splay-bend twin domains in the HAN cell [5]. Applying an oblique magnetic field and after waiting for an hour at temperature near the cholesteric-isotropic transition we obtain a monodomain sample suitable for observation of the HAC structure. A typical structure is shown in figure 2 : a) Perpendicular polarizers; the black bands correspond to x-polarization of the transmitted light - $\varphi_d = n\pi$; $n = 0, 1, \dots$ b) Parallel polarizers; the black bands correspond to $\varphi_d = (2n + 1) \frac{\pi}{2}$ c) Perpendicular polarizers and electric field applied along the z-direction ($U = 5V$, $f = 3000$ Hz). At small thickness (the left part of the photographs) one sees a structure with uniform rotation of the polarization (at fixed x). This region corresponds to a structure characterized by θ and φ which are functions only of z (at constant thickness). Hereafter we shall refer to this structure as uniform HAC (UHAC). At large thickness there appears gradually a continuous modulation of the φ angle along a direction approximately perpendicular to the local molecular director at the upper plate (modulated HAC structure - MHAC).

The modulation wavelength increases slowly with increasing sample thickness starting from about $100 \mu\text{m}$ at $d = 40 \mu\text{m}$ and reaching about $150 \mu\text{m}$ at $d = 90 \mu\text{m}$. The amplitude of the modulation goes from zero at small thickness to about 45 degrees at $d = 90 \mu\text{m}$. This modulation seems to be superposed on the UHAC structure.

2. Discussion.

2.1. UHAC structure.- Even in this relative

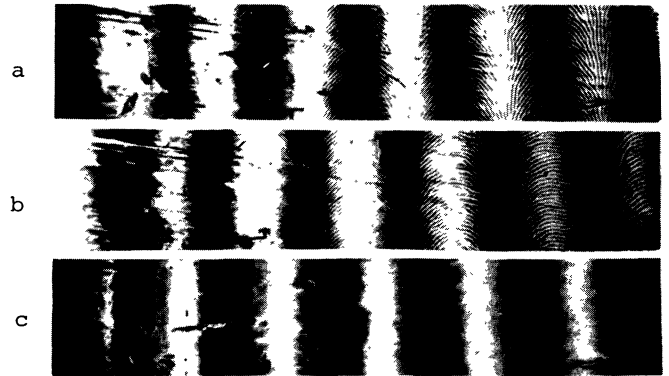


Fig. 2.- A set of micrographs of a hybrid aligned cholesteric cell : a) Crossed polarizers ; b) Parallel polarizers ; c) Crossed polarizers and electric field along the direction of observation. The thickness d increases from $d = 0$ (left) to $d = 90 \mu\text{m}$ (right). Close to the left edge of the micrograph c) is seen the border between the regions with and without electric field. It is due to the fact that we have removed the conductive layer on one of the plates in order to prevent short-circuit.

vely simple case when θ and φ are functions only of the z coordinate we make the following assumptions in order to make the analysis more simple and transparent :

A. We assume that the anchoring is strong, i.e. that the surface anchoring energy is much greater than the volume elastic energy. In this case the boundary conditions for θ and φ are extremely simple :

$$\theta = \frac{\pi}{2}, \quad \varphi = 0 \quad \text{at } z = 0$$

$$\theta = 0, \quad \partial F / \partial \frac{\partial \varphi}{\partial z} = 0 \quad \text{at } z = d \quad (1)$$

where F is the density of the volume elastic free energy.

B. We adopt a "two constant" approximation for the elastic properties of the cholesteric :

$$K_1 = K_3 \neq K_2 \quad (2)$$

This is the simplest possible approximation which conserves the main properties of the UHAC structure, e.g. the coupling between $\theta(z)$ and $\varphi(z)$.

Under these assumptions the equilibrium equations are [3] :

$$\frac{d\varphi}{du} = \frac{Q}{1 + K \cos^2 \phi} \quad (3)$$

$$\left(\frac{d\phi}{du}\right)^2 = \frac{Q^2 \cos^2 \phi}{1 + K \cos^2 \phi} + \text{Const.} \quad (4)$$

where $u = z/d$; $k = (K_1 - K_2)/K_2$; $Q = \alpha_0 d$.

The numerical results are presented in figure 3 for some values of the dimensionless parameter Q and for $k=1$ (the approximate value for our mixture). One sees that the large Q -values favour a planar alignment in the bulk of the sample.

A characteristic feature of the UHAC structure is the coupling between $\varphi(z)$ and $\theta(z)$ given by equation (3). As a result the experimentally obtained value for φ_d carries some information about $\theta(z)$. This coupling explains the displacement of the bands on figure 2c. In fact, as our mixture has negative dielectric anisotropy $\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp} < 0$, the electric field applied along z -axis favours the planar alignment of the molecules. The equation for θ is now :

$$\left(\frac{d\theta}{dz}\right)^2 = \frac{Q^2 \cos^2 \theta}{1 + K \cos^2 \theta} - \frac{\Delta\epsilon}{4\pi K_1} d^2 E^2 \cos^2 \theta + \text{Const.} \quad (5)$$

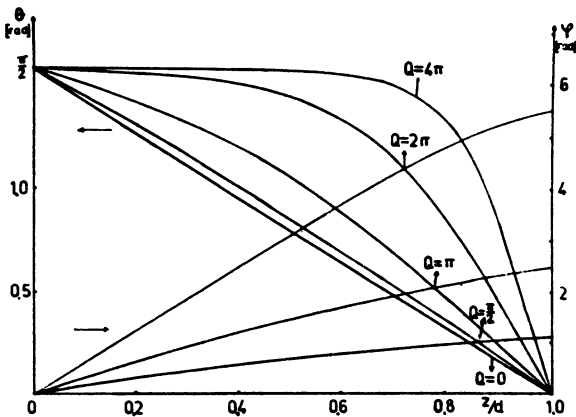


Fig. 3.- Numerical results for θ and φ as functions of z/d for $k = 1$.

The equation (3) is unchanged, but $\varphi(z)$ would also depend on E due to the coupling with $\theta(z)$. At large E values θ is different from $\pi/2$ only in a small region near the homeotropic side. The rotation of the polarization will be $\varphi_d \approx Q$, which results in displacement of the bands to the left side. This approach enables the direct determination of Q and therefore of the natural cholesteric pitch $2\pi/q_0$.

We can now briefly discuss the case when our assumptions are not satisfied. If $K_1 \neq K_3$ (as in most liquid crystals) one can obtain numerically φ_d as a function of Q and of the elastic anisotropies $k = (K_1 - K_2) / K_2$ and $k' = (K_1 - K_3) / K_3$ from the corresponding equilibrium equations. The comparison with the experimental data enables the determination of k and k' without application of an external field, i.e. in a more direct way than in most known methods for determination of the elastic constants.

Our second assumption is the strong anchoring. It can easily be satisfied using a sample with large d and small q_0 . In the case of weak anchoring φ_d depends also on the surface anchoring energy on both plates and therefore it is possible in this way to measure these energies. From figure 3 one can see that with increasing Q the torque $d\theta/dz$ decreases on the planar side and slightly increases on the homeotropic one. This property enables the independent determination of both surface anchoring energies from the dependence of φ_d on Q . This matter will be treated rigorously in a forthcoming paper. In conclusion to this subsection we shall demonstrate that φ_d is approximately equal to the polarization rotation. The criterion for wave guide regime being $[6] 2\pi\Delta n_0 > \lambda \frac{d\varphi}{dz}$ we have (close to the homeotropic plate) :

$$2\pi\Delta n_0 \theta^2 > \lambda q_0 / (1 + K)$$

where $\Delta n_0 \sim 0.2$ is the birefringence of the mixture and λ is the wavelength of the light beam. The thickness of the region in which the criterion fails is

$$d_f \sim \frac{1}{\frac{d\theta}{dz}} \sqrt{\frac{\lambda q_0}{2\pi\Delta n_0 (1 + K)}} \approx \sqrt{\frac{\lambda}{2\pi\Delta n_0 q_0}}$$

Here we take into account that at $\theta \cong 0$ (see equation (4)) :

$$\frac{d\theta}{dz} \geq \frac{q_0}{\sqrt{1 + K}}$$

The resulting error for φ_d is :

$$\Delta\varphi_d \sim \frac{q_0 d_f}{1 + K} \sim \frac{1}{1 + K} \sqrt{\frac{\lambda q_0}{2\pi\Delta n_0}}$$

In our case $\lambda \sim 0.5 \times 10^{-4}$ cm ; $2\pi/q_0 \sim 30 \times 10^{-4}$ cm ; $k \sim 1$ and we have $d_f \sim 1.5 \mu\text{m}$; $\Delta\varphi_d \sim 0.4$ rad. This error is reasonable for the present qualitative work, but should be accounted for if the UHAC structure is used to measure the surface energy or elastic anisotropy, especially in the case of weak anchoring on the planar side of the cell.

2.2 MHAC structure. - To obtain the structure of our sample in the MHAC region we need to minimize the elastic energy for the case when θ and φ are functions of all the coordinates. The equations of state in this case are very complicated and here we shall not try to obtain the general solution (a separate paper on this matter being in progress), but shall give only an idea of the type of deformation involved. Further for simplicity we assume an isotropic elastic constant : $K_1 = K_2 = K_3$.

Figure 2 suggests that θ and φ are superposition of the solutions for the UHAC structure - $\theta_0(z)$, $\varphi_0(z)$ and the small periodical functions $\theta_1(x,y,z)$ and $\varphi_1(x,y,z)$. The wavevectors of θ_1 and φ_1 depend only on the thickness, are parallel to one another and lie in the xy -plane. The angle α between the wavevectors and the x -axis is also a function only of the thickness. It is convenient to consider a sample with constant thickness and to rotate our coordinate system around the z -axis at angle α so that the wavevectors lie along the new x -axis. In the new coordinate system the polar angle is unchanged $\theta = \theta_0(z) + \theta_1(x,z)$ and the azimuth is $\varphi = \varphi_0(z) + \varphi_1(x,z) - \alpha$. Without great loss of generality we can take x -dependences of θ_1 and φ_1 as pure sine waves :

$$\begin{aligned}\theta_1 &= T(z) \sin(q_1 x + \delta_1(z)) \\ \varphi_1 &= F(z) \sin(q_2 x + \delta_2(z))\end{aligned}\quad (6)$$

The dephasing $\delta_1(z) - \delta_2(z)$ is in general a function of z and as we shall see later it plays an important role in the problem.

Under our assumptions the elastic free energy density is :

$$\begin{aligned}F &= F_0 + \frac{K_1}{2} \left\{ \theta_{1z}^2 + \theta_{1x}^2 + (\varphi_{1x}^2 - \varphi_{1z}^2) + \right. \\ &\quad \left. \sin^2 \theta_0 - q_0^2 \theta_1^2 \cos 2\theta_0 + \right. \\ &\quad \left. + 2q_0 \cos(\varphi_0 - \alpha) (\varphi_1 \theta_{1x} + \right. \\ &\quad \left. \theta_1 \varphi_{1x} \cos 2\theta_0) + \right. \\ &\quad \left. q_0 \varphi_1 \varphi_{1x} \sin(\varphi_0 - \alpha) \sin 2\theta_0 \right\}\end{aligned}\quad (7)$$

Here $F_0 = (\theta_{0z}^2 - q_0^2 \sin^2 \theta_0) K_1/2$ is the energy of the UHAC structure and the subscript x or z means a differentiation with respect to x or z . We neglect the terms of order higher than two as θ_1 and φ_1 are supposed to be small. The first order terms are also neglected because in the following averaging on x they vanish.

The boundary conditions for θ_1 and φ_1 are :

$$\begin{aligned}\theta_1 &= 0, \quad \varphi_1 = 0 \text{ at } z = 0 ; \\ \theta_1 &= 0, \quad \varphi_{1z} = 0 \text{ at } z = d\end{aligned}\quad (8)$$

A possible gain in the cholesteric-like twist energy of the modulated structure is related to the terms linear in q_0 . After averaging the free energy with respect to x these terms do not vanish only if $q_1 = q_2$. If we consider a slab of the sample at constant z , the free energy of this structure is minimized at :

$$\varphi_0(z) - \alpha = 0 ; \quad \delta_1(z) - \delta_2(z) = \frac{\pi}{2}\quad (9)$$

The resulting deformation in the slab is presented in figure 4. The director \vec{n} describing the MHAC structure moves on the surfa-

ce of a elliptical cone whose axis is the director \vec{n}_0 corresponding to the UHAC structure. The semiaxes of the basis of the cone are the amplitudes $T(z)$ and $F(z)$. In fact this structure is only the leading term in the general x -dependence of θ_1 and φ_1 . The other terms with $q_1 = mq_2$ or $q_2 = nq_1$ ($m, n = 2, 3, \dots$) can be obtained from the terms of order higher than two in the free energy, which have been neglected in equation (7).

Let us now briefly consider the z -dependences of the amplitudes $T(z)$ and $F(z)$ in equation (6) for the structure shown in figure 4. The boundary conditions (8) imply that the amplitudes can be expanded in the following Fourier series :

$$\begin{aligned}T(z) &= \sum_k t_k \sin(k\pi z/d) \\ F(z) &= \sum_k f_k \sin(k\pi z/2d)\end{aligned}\quad (10)$$

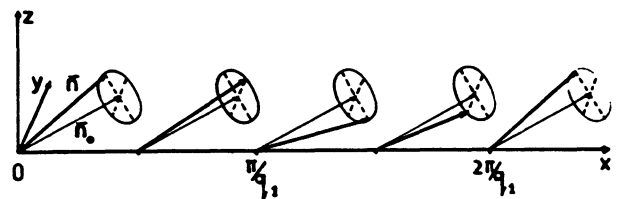


Fig. 4.- Deformation in a thin slab of hybrid aligned cholesteric in the case of modulated HAC structure.

Unfortunately in our case we cannot expect that the amplitudes are pure Fourier components. In fact, the conditions (9) cannot be satisfied for all z -values as α is a constant and φ_0 is z -dependent. So we have some gain in the twist energy in the slab at z_1 corresponding to $\cos(\varphi_0(z_1) - \alpha) > 0$ and some loss at z_2 corresponding to $\cos(\varphi_0(z_2) - \alpha) < 0$. In order to have a MHAC structure with lower energy than the UHAC one, we need a solution which ensures the gain in the z_1 -region to overcompensate the loss in the z_2 -region, for example an amplitude which is pure Fourier component multiplied by $\cos(\varphi_0(z) - \alpha)$ or a dephasing $\delta_1(z) - \delta_2(z) \cong \varphi_0(z) - \alpha + \frac{\pi}{2}$. We see from the above discussion that the minimization of the free energy of the MHAC structure and the demonstration of its stability is a difficult problem which is beyond the scope of this letter.

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