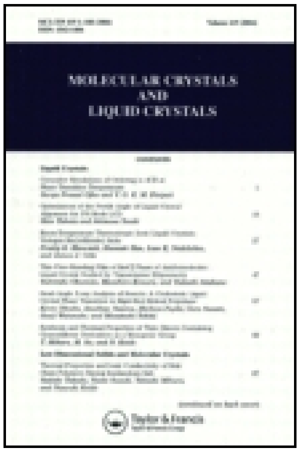

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PERIODIC DOMAIN STRUCTURES IN THIN HYBRID NEMATIC LAYERS

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Abstract Director distribution in the plane of a thin equilibrium nematic layer with different and degenerate orientations of molecules on upper and lower surfaces is found to be periodic in the absence of external fields (Fig.1).

To provide the above mentioned hybrid boundary conditions, we deposited the nematic (K-440, MBBA, or 5CB) on the surface of glycerin or the APGT polymer; the upper surface was free. The easy axis of nematic molecules was tangent to the lower surface and nealy normal to the free surface. Temperature gradients were absent.

A polarizing microscope was employed to study nematic films of thickness h~ 1μm. The observation revealed domain structure manifested through alternating dark and light bands (Fig.1). The light intensity modulation in the horizontal plane is caused by the director deformation in this plane rather than by variations of the molecule tilting at the layer surface. The latter assertion is confirmed by the fact that dark and light bands interchange under the rotation of the sample. The domain width decreases almost linearly with decreasing layer width (Fig.2).

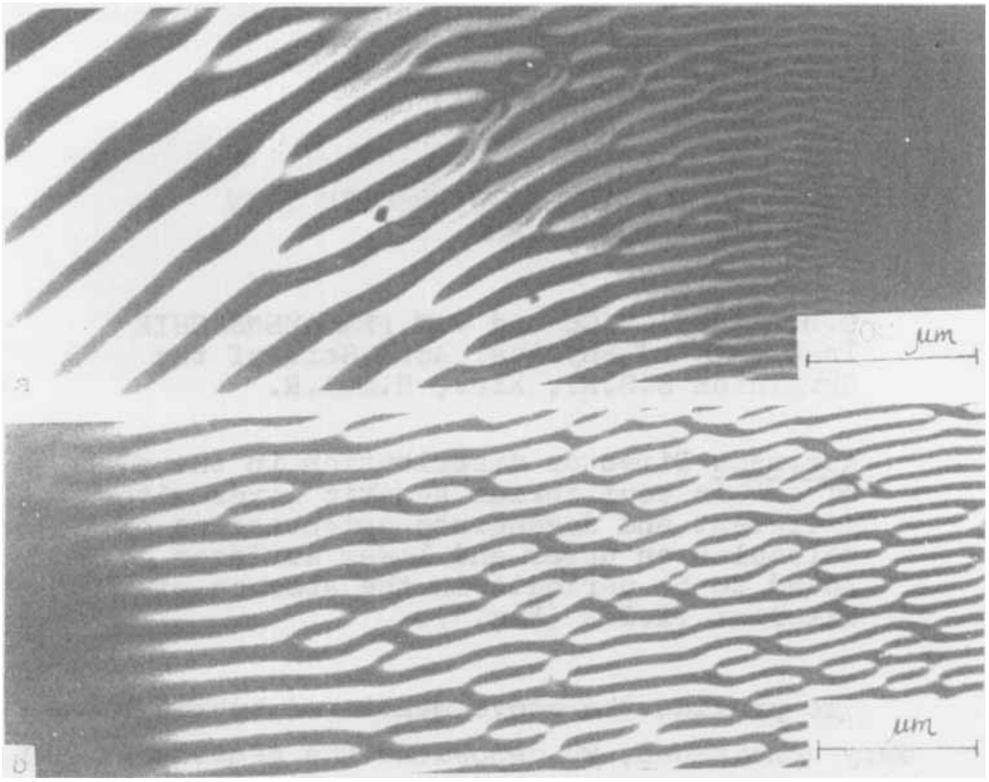

FIGURE 1. Periodic structures in HNL: K-440 on the APGT surface (a); 5CB on the glycerin surface (b).

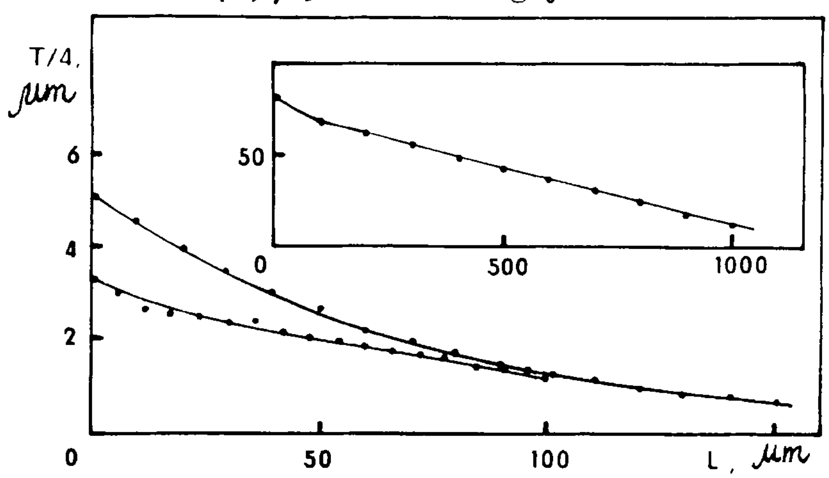

FIGURE 2. Typical dependences of the domain period T on the distance L taken from the domain boundary towards the film edge for two films of 5CB on glycerin. Insertion - K-440 on APGT.

Being able to form periodic spatial structures either due to specific intermolecular interactions (cholesterics, smectics, blue phases) or under the influence of external directive forces (temperature gradients, electric and magnetic fields) is one of the most important properties of liquid crystals[1-3]. In this paper, the periodic structure is discovered in the translationally symmetric phase with no external fields. It is formed because the boundary conditions on the layer surfaces are different and degenerated. In the earlier studies considering similar conditions in equilibrium, the director had been assumed to be deformed only in the plane normal to the layer[4-7]. However, this state turns out to be unstable with respect to deformations in the layer plane if the deformations in the normal plane are sufficiently strong. Since the boundary conditions are degenerated, the director rotates on the layer surfaces without energy losses.

The sufficient condition of the uniform state (US) instability in the plane of a hybrid nematic layer (HNL) is the existence of weak deformations $\delta \underline{n}$ of the director $\underline{n}_o$ for which the second variation $\delta^2 F\{\underline{n}_o, \underline{n}\}$ of the free energy $F\{\underline{n}_o\}$ is negative. Critical parameters are determined by the condition $\min \delta^2 F = 0$.

Suppose $K_{11}=K_{33}=K$, K_{22} and K_{24} are arbitrary, $K_{13}=0$. The first assumption is natural, and the last one is made for the sake of simplicity. Let z-axis be normal to the layer of thickness h, x- and y-axes be parallel and perpendicular to the domain respectively.

For US, we have $\underline{n}_0=(\sin\theta\cos\Phi, \sin\theta\sin\Phi, \cos\theta)$, $\Phi=0$ and $\theta(z)=(\theta_2-\theta_1)z/h+\theta_1$, where $\theta_1=\theta(0)$ and $\theta_2=\theta(h)$ are the angles between $\underline{n}_0$ and the normals to the layer surfaces (subscripts "1" and "2" henceforth label the lower and the upper surfaces respectively) which may be found in the standard manner[6] from the equations

$$2(\theta_2-\theta_1)+\frac{W_2 h}{K}\sin 2(\theta_2-\bar{\theta}_2)=2(\theta_1-\theta_2)+\frac{W_1 h}{K}\sin 2(\theta_1-\bar{\theta}_1)=0\,, \quad (1)$$

where W_i are the anchoring constants, $\bar{\theta}_i$ are the angles between the easy directions and the normals. For $K_{11}=K_{33}=K$, the free energy F (per unit length along x) is given by[8,9]

$$F=\frac{K}{2}\int dV\left\{(\underline{\nabla}\underline{n})^2+(\underline{\nabla}\times\underline{n})^2-(1-t)(\underline{n}\underline{\nabla}\times\underline{n})^2-\right.$$
$$\left.-p\underline{\nabla}\left(\underline{n}(\underline{\nabla}\underline{n})-(\underline{n}\underline{\nabla})\underline{n}\right)\right\}+\frac{1}{2}\sum_{i=1,2}\int dS_i W_i \sin^2(\theta_i-\bar{\theta}_i), \quad (2)$$

where $t=K_{22}/K$, $p=(K_{22}+K_{24})/K$. Within the context of the identity

$$(\underline{\nabla}\underline{n})^2+(\underline{\nabla}\times\underline{n})^2\equiv(\partial_j n_j)^2+\underline{\nabla}\left(\underline{n}(\underline{\nabla}\underline{n})-(\underline{n}\underline{\nabla})\underline{n}\right)$$

and the assumptions $\partial/\partial x=0$, $\delta\theta=\psi$, $\delta\Phi=\varphi$, $A_{x_i}=\partial A/\partial x_i$, $\Delta=1-t$, we find the second variation of F, $\delta^2 F=\delta^2 F_V+\delta^2 F_S$, in the vicinity of the director $\underline{n}_0$:

$$\delta^2 F_V=\frac{K}{2}\int_V dz\,dy\left\{\psi_z^2+t\psi_y^2+\sin^2\theta\cdot\varphi_y^2+\sin^2\theta(1-\Delta\sin^2\theta)\varphi_z^2\right.$$
$$\left.-2\Delta\sin^2\theta\,\varphi_z\psi_y\right\}, \quad (3)$$

$$\delta^2 F_S=\frac{K}{2}\int_{V_y} dy\left\{4(1-p)(\sin^2\theta\cdot\psi_y\cdot\varphi)\right|_1^2+\sum_{i=1,2} l_i\psi^2(z_i),$$

where $l_i=W_i\cos 2(\theta_i-\bar{\theta}_i)/K$, $(\ldots)_1^2=(\ldots)_{z=h}-(\ldots)_{z=0}$.

We are interested in the state that is periodic in y and so take it in the Fourier series form. We retain only the first harmonic of the series: $\psi = f(z)\sin qy$, $\varphi = g(z)\sin(qy+\lambda)$. Minimizing $\delta^2 F$ with respect to λ yields $\lambda = \pi/2$. Then, after one introduces $\varkappa = qh$, $\eta = (4h/KV_y)$(V_y is the dimensions of the system along the y-axis) and carries out integration over y, one obtains from (3)

$$\eta\delta^2 F = \int_0^1 dz\left\{f_z^2 + t\varkappa^2 f^2 + \varkappa^2 \sin^2\theta \cdot g^2 + \sin^2\theta(1-\Delta\sin^2\theta)g_z^2\right.$$
$$\left. - 2\varkappa\Delta\sin^2\theta\cdot g_z\cdot f\right\} + 4\varkappa(1-p)(\sin^2\theta\cdot f\cdot g)\Big|_1^2 + \qquad (4)$$
$$+ h\sum_{i=1,2} \ell_i f^2(z_i),$$

where the notation z implies now z/h, so that $0 \le z \le 1$. The Euler-Lagrange equations for the bulk of (4) are

$$f_{zz} - t\varkappa^2 f - \varkappa\Delta\sin^2\theta\cdot g_z = 0, \qquad (5)$$
$$\frac{\partial}{\partial z}\left[(1-\Delta\sin^2\theta)g_z + \varkappa\Delta\sin^2\theta\cdot f\right] - \varkappa^2 \sin^{\sim}\theta ...$$

The analysis of the complete expression (4) shows that $\delta^2 F$ is minimum for $\varkappa \ll 1$ and $g(z) = g_0 + \tilde{g}(z)$, where $g_0 = \text{const} \gg g(z)$(since the term $\sim (1-p)$ is dominant in (4)). In this case, the solution of Eqs. (5) is to be presented as power series of small quantities $\varkappa$, ξ, ζ, g_0, and to within the squares of these quantities, is of the form

$$f = \xi - \zeta z, \qquad (6)$$
$$\tilde{g} = \int_0^z dz \, \frac{\varkappa^2 g_0[1-\sin 2(\theta-\theta_1)/2(\theta-\theta_1)] - 2\varkappa\Delta\sin^2\theta\cdot f}{2(1-\Delta\sin^2\theta)}, \qquad (7)$$
$$g(z) = g_0 + \tilde{g}(z).$$

The quantity g_0 implies the angle $g(z=0)$. Substituting (6) and (7) in (4) and assuming $f \sim \varkappa g_0$, one obtains $\delta^2 F=\Delta_2+o(f^2)$, where

$$\Delta_2=as^2+4s(1-p)\left[\xi\sin^2\theta_1-(\xi-\zeta)\sin^2\theta_2\right]+ + h\left[l_1\xi^2+l_2(\xi-\zeta)^2\right]+\zeta^2 . \quad (8)$$

The latter is a quadratic form of the quantities $s=\varkappa g_0$, ξ and ζ . The domain state is energetically preferable for $\min\Delta_2<0$, the opposite condition is favorable for US; the critical point for which the US becomes unstable is determined by requirement $\min\Delta_2=0$. The necessery and sufficient condition for the latter to be satisfied is D=0, where D is the determinant of the coefficient matrix of the form Δ_2.The critical condition D=0 may be written as

$$h=\left(-B+\sqrt{B^2+16b^2A^2}\right)(2A)^{-1} , \quad (9)$$

where $A=al_1l_2$, $a=1/2-(1/4)\sin 2(\theta_1-\theta_2)/(\theta_1-\theta_2)$,

$$B=ac-4(1-p)^2(l_2\sin^4\theta_1-l_1\sin^4\theta_2), \quad (10)$$

$$c=l_1+l_2, \quad b=(1-p)(\sin^2\theta_1-\sin^2\theta_2).$$

We remind the reader that the quantity $\theta_i=\theta_i(h, W_1/K, W_2/K)$ in (10) is the solution of (1), therefore equation (9) determines the critical height h_c.

In order to find the wave number $\varkappa$ and the amplitudes g_0, ξ and ζ , one has to take into account in the expension of F higher orders than in Δ_2. We give here only their dependences on the determinant D, $0<-D\ll 1$, in the vicinity of the phase transition point for which D=0:

$$g_0, \varkappa \sim (-D)^{1/2} , \quad (11)$$
$$\xi, \zeta \sim -D .$$

Thus we see that if (9) possesses a solution, then there occurs a phase transition from the US into the domain state. There appear deformations of all the three types, including two twists: around the z-axis $\sim \tilde{g}(z)$ and around y-axis $\sim$ sinqy. However, the dominant contribution in the US instability in (4) is given by the surface term proportional to (1-p) which corresponds the interaction between the twist about the y-axis and splay $g_o$cosqy. So it is natural that $g_o \sim (-D)^{1/2}$ grows under transition much more rapidly than $\tilde{g}(z) \sim (-D)^{3/2}$ (cf. (11) and (7)). The structure period is $T_y = 2\pi h/\chi \gg h$.

In order to compare the theory and experiment in detail, one has to carry out detailed measurements (the wedge profile, Θ_i, etc.) and develop the theory with regard for the terms containing $K_{13} \neq 0$. This will be made in our next paper. But even the theory proposed here enables one to suggest that the stripes with $T_y \gg h \sim \mu m$ can be given rise to by no terms other than the surface one $\sim$ (1-p): with the latter being disregarded (i.e., for $K-K_{22}-K_{24}=0$), the US becomes unstable only for very small K_{22}, $\chi \sim 1$, $h_c \ll 1$ μm.

Equations (1) and (9) were solved numerically for $W_1=10^{-5}Jm^{-2}$, $W_2=4.5 \cdot 10^{-6}Jm^{-2}$, $K=10^{-11}N$. In standard case $p=K_{22}+K_{24}=0$, there exist two roots $h_{c1}=10\mu m(\Theta_1=83^\circ,\ \Theta_2=16^\circ)$ and $h_{c2}=3\mu m(\Theta_1=79^\circ,\ \Theta_2=39^\circ)$. The domain state occurs for the thickness $h_{c2}<h<h_{c1}$. The disappearance of stripe domains for sufficiently small h is confirmed experimentally. For $p>p_c \sim 0.4$, the roots coincide, $h_{c1}=h_{c2} \sim 4.6\mu m$ and the domain state does not occur.

As follows from the qualitative consideration, the US has to be unstable for sufficiently developed deformations $\sim(\theta_1-\theta_2)$ in the vertical plane. The numerical analysis confirms this statement and makes it more accurate:it shows that Eq.(9) has a solution for considerable $b\sim(\sin^2\theta_1-\sin^2\theta_2)$.

In conclusion we note that in the experiments with wedge-like HNL,the boundary of the US is associated with finite-period domains rather than infinite ones predicted by the theory in the critical point. The reason is, the domain period T_y varies very rapidly near h_c. Such variations of the stripe widths would require enormous numbers of defects. That is why in reality the stripes appears at the wedge beginning from $h<h_c$, when T_y varies slowly from the boundary towards the wedge margin and the defects are few.They are seen in Fig.1.

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