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## Saddle-splay and periodic instability in a hybrid aligned nematic layer subjected to a normal magnetic field

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**Abstract.** — A nematic layer with opposite boundary conditions (unidirectional planar and homeotropic) is considered, having strong anchoring at the planar wall. It is known that a periodic deformation of splay-type can intervene between the aperiodic hybrid alignment and undistorted planar state as one decreases the film thickness. This periodic state occurs due to the fact that the energetic cost for a mixed twist-splay can be lower than the surface energetic cost at the homeotropic wall in the case of undistorted planar alignment. Such a situation may also be achieved for nematics which have a bulk elastic isotropy. In this paper, the saddle-splay elastic constant  $K_{24}$  is shown to influence strongly the occurrence of the periodic pattern of splay-type, also in the presence of an external magnetic field normal to the cell plates, and the role of the geometrical anchoring in wedge-shaped cells is discussed.

### Introduction.

In a nematic liquid crystal cell weakly anchored with opposite boundary conditions, i.e. homeotropic (H) at one of the substrates, unidirectional planar (P) at the other, it is possible to achieve an aperiodic hybrid alignment (HAN) only if the cell thickness  $d$  exceeds a threshold  $d_a$  [1]. Generally speaking, for  $d < d_a$ , an undeformed alignment was expected, driven by the easy direction of the wall exhibiting the stronger tilt-anchoring [2], since in principle a two-dimensional distortion would imply too high an energetic cost. Nevertheless, in the absence of any distortion, an amount of potential energy is bounded at the surface presenting the weaker anchoring. Thus two deformation sources (elastic and interfacial) are in competition, and the existence of a periodic distortion (PHAN) may be preferred in a convenient range  $d_p < d < d_a$ , due to the balancing effect of the opposite alignment induced by the surfaces [3, 4] and mediated by the material elastic parameters. As a consequence, the homogeneous P-state can arise only below the lower threshold  $d_p$  of the periodic distortion.

As we discussed, in the case of hybrid nematic layers with a stronger tilt-anchoring at the P-wall, whose periodic texture exhibits essentially a twist-splay distortion, the PHAN structure is deeply favoured by the weakening of the twist-anchoring, especially at the H-wall [4, 5]. Moreover, such a periodic pattern may also occur for high values of the bulk elastic ratio  $K_{22}/K_{11}$ . This behaviour is quite different from that presented by an undeformed planar cell subjected to magnetic fields inducing periodic distortions [6-9].

In this paper we investigate the effect of an external magnetic field normal to the cell plates on the static periodic pattern [10] in a hybrid nematic layer with bulk elastic isotropy, i.e. with equal splay and twist elastic constants  $K_{11} = K_{22} = K$ . In order to study the influence of the H-boundary, both direct and mediated by the surface-like elasticity, the cell is supposed to have a strong planar and weak homeotropic anchoring for both the azimuth  $\Phi$  (in-plane twist angle) and the polar angle  $\theta$  (off-plane tilt angle). Hence the possible periodic deformation can only be of the splay type. Moreover, we consider liquid crystals which have different values of the saddle-splay elastic constant  $K_{24}$  [11-15], with the thermodynamical constraints  $|K_{24}| < K_{22}$  [16], in order to show the great influence of  $K_{24}$  on the PHAN, and to provide a possible method for measuring  $K_{24}$ .

### Theory.

**PHAN PATTERN.** — Let us consider a nematic layer confined between two substrates, placed at  $z = 0$  and  $z = d$ , where the easy directions are H- (along the  $z$ -axis) and P- (along the  $x$ -axis), respectively. The anchoring is supposed to be strong only at the planar wall. We are looking for the existence of the transverse periodicity, with the wave vector parallel to the  $y$ -axis: hence the tilt angle  $\theta$  and the twist angle  $\Phi$  turn out to be depending on  $y$  and  $z$ , i.e.,  $\theta = \theta(y, z)$  and  $\Phi = \Phi(y, z)$  (see Fig. 1). In order to simplify the notation of spatial derivatives which is necessary to describe the director distortion, hereafter the subscript  $y$  (or  $z$ ) means a partial derivative with respect to  $y$  (or  $z$ , respectively).

By assuming, as usual, the Rapini-Papoular form for the anchoring energy density [17-19], and by taking into account the surface contribution due to the surface-like saddle-splay elastic constant  $K_{24}$ , the linearized surface reduced free energy density (which is the free energy density divided by  $K/2$ ) is given by:

$$g_s = L_{\Phi_0}^{-1} \Phi_0^2 - L_{\theta_0}^{-1} \theta_0^2 + 2(1 + K_{24}/K) [\theta_0 \Phi_{y_0} - \Phi_0 \theta_{y_0}] \quad (1)$$

where  $L_{i_0} = K/W_{i_0}$  are de Gennes-Kléman extrapolation lengths ( $i = \Phi, \theta$ ) at the homeotropic wall ( $z = 0$ , index o) [20, 21],  $W_{i_0}$  being the relevant anchoring strengths. Also  $\Phi_{y_0}$  and  $\theta_{y_0}$  are calculated at the wall  $z = 0$  (index o).

When a magnetic field parallel to the  $z$ -axis is present, the bulk reduced free energy density is obtained as:

$$g_b = (\Phi_y + \theta_z)^2 + (\theta_y - \Phi_z)^2 - h^2 \theta^2 \quad (2)$$

where  $h = H (\chi_a/K)^{1/2}$  is the reduced field,  $\chi_a$  being the anisotropy of the magnetic susceptibility.

Hence the cell reduced free energy is written

$$G = \int_0^d \int_0^\lambda g_b dy dz + \int_0^\lambda g_s dy \quad (3)$$

where  $\lambda$  is the PHAN wavelength along the  $y$ -axis.

By applying the usual minimization procedure [22], the linearized Euler-Lagrange equations

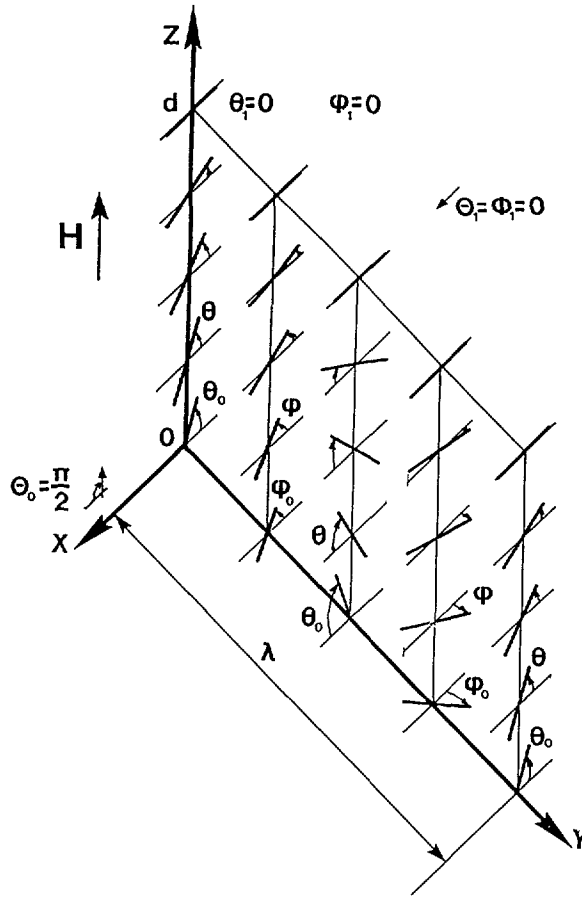


Fig. 1. — PHAN pattern of splay-type in a nematic cell with opposite boundary conditions. A magnetic field  $H$  is applied normally to the cell plates. The tilt- and twist-angles  $\theta(y, z)$  and  $\Phi(y, z)$  are periodic functions of the transverse in-plane coordinate  $y$ , with wavelength  $\lambda$ . Both tilt- and twist-anchorings are supposed to be weak at the H-wall ( $z = 0$ ) and strong at the P-wall ( $z = d$ ).

read :

$$\begin{aligned} \theta_{yy} + \theta_{zz} + h^2 \theta &= 0 \\ \Phi_{yy} + \Phi_{zz} &= 0 \end{aligned} \tag{4}$$

and their solutions must satisfy the linearized boundary conditions which are given by :

$$\begin{aligned} R\theta_{y0} + L_{\Phi_0}^{-1} \Phi_0 - \Phi_{z0} &= 0 \\ R\Phi_{y0} + L_{\theta_0}^{-1} \theta_0 + \theta_{z0} &= 0 \\ \Phi_1 &= 0 \\ \theta_1 &= 0 \end{aligned} \tag{5}$$

where  $R = 1 - 2(1 + K_{24}/K)$ .

Hence the tilt- and twist-angles for small periodic distortions are obtained as :

$$\begin{aligned} \theta &= (A \operatorname{sh} qz + B \operatorname{ch} qz) \cos \beta y \\ \Phi &= (a \operatorname{sh} \beta z + b \operatorname{ch} \beta z) \sin \beta y \end{aligned} \tag{6}$$

where  $\beta = 2\pi/\lambda$  is the in-plane wave vector of the periodic pattern along the  $y$ -axis, and  $q^2 = \beta^2 - h^2$ . Note that  $q = ik$  can also be imaginary: in this case, the first equation of system (6) reads:

$$\theta = (A \sin kz + B \cos kz) \cos \beta y. \quad (7)$$

By substituting (6) into the boundary conditions, a linear system in the integration constants is obtained. The coefficient determinant  $D$  of such a system is written

$$D = \begin{vmatrix} R\beta L_{\phi_0} & -1 & 0 & \beta L_{\phi_0} \\ 1 & R\beta L_{\theta_0} & qL_{\theta_0} & 0 \\ 0 & \text{ch } \beta d & 0 & \text{sh } \beta d \\ \text{ch } qd & 0 & \text{sh } qd & 0 \end{vmatrix}. \quad (8)$$

In the case of validity of (7),  $(q, \text{ch } qd, \text{sh } qd)$  shall be simply replaced, by  $(k, \cos kd, \sin kd)$ , respectively, in the determinant  $D$ . Obviously, in order to avoid a trivial solution,  $D$  must vanish. This means that the threshold thickness  $d_p$  has to be found by imposing  $D = 0$ , with fixed values of  $L_{\phi_0}$ ,  $L_{\theta_0}$ ,  $h$  and  $R^2$  (the square dependence on  $R$  is due to the structure of  $D$ ). Such an assumption gives  $d(\beta)$  biased by the material and field parameters. The minimum of the curve  $d(\beta)$  is  $d_p$ , provided that the reduced free energy  $G$  has also a minimum for  $d = d_p$ .

Note that the range of the allowed values of  $R$  was obtained by Ericksen through thermodynamical considerations, giving  $|K_{24}/K| < 1$ . Then  $R$  ranges within  $(-3, 1)$ .

In figure 2 the threshold  $d_p$  is shown as a function of  $|R|$  for different values of the reduced magnetic field ( $h = 0, 0.5, 1, 2$ ) and of the twist-extrapolation length  $L_{\phi_0}$ . More precisely, in figure 2 three families of the function  $d_p(|R|, h)$  are represented, where the values of  $L_{\phi_0}$  are chosen equal to  $0.5 \mu\text{m}$ ,  $1 \mu\text{m}$  and  $5 \mu\text{m}$ , respectively, whereas  $L_{\theta_0} = 0.5 \mu\text{m}$  is fixed.

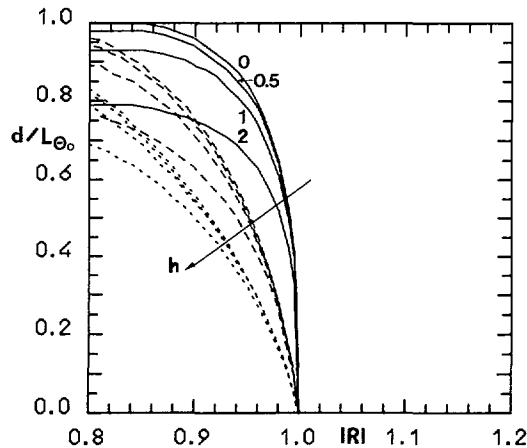


Fig. 2. — P-PHAN threshold reduced thickness  $d_p/L_{\theta_0}$  vs. the absolute renormalized saddle-splay  $|R|$ , where  $L_{\theta_0} = 0.5 \mu\text{m}$ . The three sets of curves correspond to different values of  $L_{\phi_0}$  ( $0.5 \mu\text{m}$  (—);  $1.0 \mu\text{m}$  (- -);  $5.0 \mu\text{m}$  (...)). The weakening of the torsional anchoring favours PHAN, so does the increasing of the reduced field  $h$ . Inside each set of curves,  $h$  takes the following values: 0, 0.5, 1 and 2. Note the critical value  $|R_c| = 1$  suppressing P-structure. We stress the fact that the P-HAN reduced threshold  $d_p/L_{\theta_0}$  coincides with the maximum of  $d_p/L_{\theta_0}$  and, being independent of  $|R|$  and  $L_{\phi_0}$ , is a decreasing function of  $h$ .

Furthermore, in figures 3 and 4 the corresponding results are reported when  $L_{\theta_0} = 1 \mu\text{m}$  and  $5 \mu\text{m}$ , respectively.

HAN PATTERN. — Coming back to the P-HAN threshold thickness  $d_a$ , its behaviour as a function of  $h$  has to be found. When  $d \rightarrow d_a$  from the P-side, obviously  $\theta(z)$  and  $\Phi = 0$ . Hence the only Euler-Lagrange equation is written

$$\theta_{zz} + h^2 \theta = 0 \tag{4'}$$

as the harmonic pendulum equation whereas the boundary conditions are given by :

$$\begin{aligned} \theta_{z_0} + L_{\theta_0} \theta_0 &= 0 \\ \theta_1 &= 0 \end{aligned} \tag{5'}$$

independently of  $K_{24}$ , as expected, since the distortion is not spatial [12], acting only in the planes parallel to  $[xz]$ .

By inserting the solution

$$\theta = A \sin hz + B \cos hz \tag{6'}$$

into (5'), and by looking for non-trivial results, a generalized Rapini-Papoular equation [17, 23] is obtained

$$d_a/L_{\theta_0} = \tan^{-1}(hL_{\theta_0})/(hL_{\theta_0}). \tag{7'}$$

By taking into account that  $d_a(h = 0) = L_{\theta_0}$ , as shown in reference [1], equation (7') demonstrates that  $d_a(h)$  is a decreasing function, for a given value of the tilt extrapolation length. More precisely,  $d_a/L_{\theta_0}$  goes from 1 to 0 when  $hL_{\theta_0} = H(\chi_a K)^{1/2}/W_{\theta_0}$  ranges from 0 to  $\infty$ . This was expected, since a normal magnetic field tries to suppress the undeformed P-alignment : the higher the bulk rigidity, the more pronounced this effect.

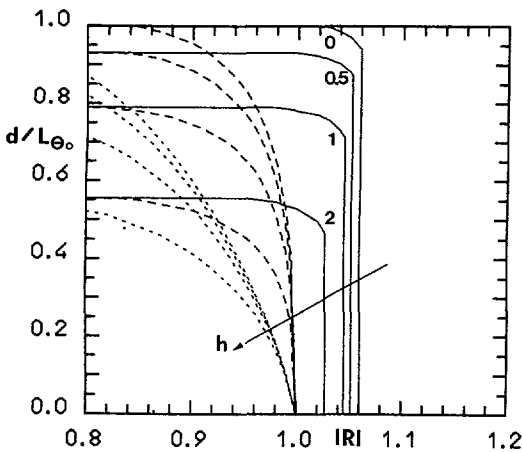


Fig. 3.

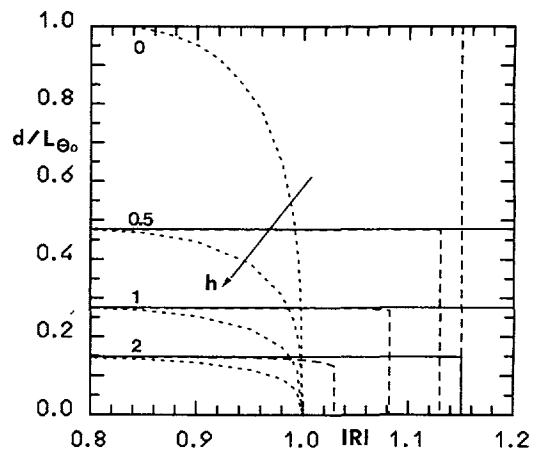


Fig. 4.

Fig. 3. — The same as in figure 2, but with  $L_{\theta_0} = 1 \mu\text{m}$ . Note that for a sufficiently strong torsional anchoring ( $L_{\phi_0} < L_{\theta_0}$ )  $|R_c|$  becomes dependent on  $h$  and larger than one ( $|R_c| \rightarrow 1$  when  $h \rightarrow \infty$ ).

Fig. 4. — The same as in figures 2, 3, but with  $L_{\theta_0} = 5 \mu\text{m}$ . The condition  $L_{\phi_0} < L_{\theta_0}$  always provides that  $|R_c|$  is larger than one and dependent on  $h$ . Note that, for  $L_{\phi_0} = 0.5 \mu\text{m}$ , PHAN is practically suppressed if  $|R| < |R_c|$ .

### Discussion.

By considering the obtained behaviour of the threshold thickness P-PHAN, depicted in the previous figures, one may easily realize that the PHAN acceptance area for each set of the triplet  $(L_{\phi_0}, L_{\theta_0}, h)$  is above the  $d_p$  threshold line in the phase plane  $[|R|, d]$ . Hence, first of all the PHAN distortion of the splay-type is always favoured by increasing values of  $|R|$  until a critical value  $|R_c|$  is reached — which means that the periodic pattern is favoured by decreasing values of  $|1 + K_{24}/K|$ . Above  $|R_c|$  the undeformed P-structure is suppressed for any value of the layer thickness, and  $d_p$  vanishes : only the PHAN and HAN structures are allowed. It turns out to be  $|R_c| = 1$  for sufficiently weak twist- with respect to tilt-anchoring  $L_{\phi_0} > L_{\theta_0}$ , independently of the values of  $h$ . Otherwise, for stronger twist- with respect to tilt-anchoring energies, the critical saddle splay value  $|R_c|$  increases with the reduced field  $h$  as well (see Figs. 2-4). Anyway, when  $|R| < |R_c|$ , PHAN is favoured with respect to the undistorted P-alignment by conveniently small twist-anchoring strength balanced by sufficiently high tilt-anchoring energy.

With respect to this conclusion, it is important to note that the first observations of the PHAN pattern were performed for the nematic film placed between two isotropic media (isotropic fluid and air) [3, 24]. The isotropic nature of the substrate usually implies the azimuthal degeneracy of the boundary conditions [25]. For example, the elastic energy per unit surface area of the aperiodic HAN solution for weak polar anchoring at both surfaces [1].

$$F_o = K(2d)^{-1}(\Delta\theta)^2 \quad (9)$$

$(\Delta\theta = \theta_o - \theta_1)$ , depends only on the mutual polar orientations at the boundaries and should not depend on the azimuthal angle  $\Phi$  when one has no special treatment of the nematic-isotropic medium interface. However, this azimuthal degeneracy can be removed just by simple inclination of the cell plates with respect to each other [26].

Thus, let the P-boundary be tilted with respect to the H-boundary by a small angle  $\gamma \ll \Delta\theta$ . Obviously, the difference  $\Delta\theta$  in the polar orientation at the walls will now be azimuthally dependent :

$$\Delta\theta = (\theta_o - \gamma \cos \Phi), \quad (10)$$

where  $\Phi$  is the azimuthal angle in the tilted plane measured from the normal to the rotation axis  $y$ . Equations (9) and (10) lead to an azimuthally dependent elastic energy of the aperiodic HAN solution with the minimum at  $\Phi = 0$  :

$$F = F_o + K\gamma(2d)^{-1}(\gamma \cos^2 \Phi - 2\Delta\theta \cos \Phi). \quad (11)$$

The second term in the last equation (11) may be considered as a special artificial « anchoring » induced by mutual inclination at the surfaces. The extrapolation length of this anchoring can be expressed as  $L_\gamma = d/\gamma$ . With  $d = (1 - 10 \mu\text{m})$  and  $\gamma \simeq (10^{-3} - 1)$ , one finds that  $L_\gamma$  can vary in the range  $\simeq (1 - 10^4) \mu\text{m}$ ; of course, much higher values of  $L_\gamma$  are easily reached. In the case of « physical » anchoring, due to a special surface treatment, one has usually  $0.1 \leq L_i \leq 10 \mu\text{m}$  for the extrapolation length  $L_i$  ( $i = \Phi, \theta$ ).

Thus,  $L_\gamma$  and  $L_i$  can be of the same order of magnitude, thus  $L_\gamma$  should be included in the solutions of the problems with a non-trivial shape of the liquid-crystalline volume.

In the experimental situation discussed in references [3, 24], where  $L_\phi = \infty$ , the domains appeared in the precursor part of the spreading nematic film, where  $d \leq 1 \mu\text{m}$  and  $\gamma \simeq 0.01 - 0.1$ . Thus  $L_\gamma = (10-100 \mu\text{m})$ , i.e. the corresponding anchoring is too small to prohibit the periodic pattern, as may be deduced from figures 2-4.

Instead, in the present case we consider  $L_{\phi_1} = L_{\theta_1} = 0$  at the P-wall : this fact enables us to compare the effect of only two anchoring parameters ( $L_{\phi_0}$ ,  $L_{\theta_0}$  at the H-wall) and of the saddle-splay elasticity  $|R|$  with the magnetic field effect. This means that three measurements of the threshold thickness  $d_p$  at different reduced fields  $h$  are enough to obtain an estimate of  $L_{\theta_0}$ ,  $L_{\phi_0}$  and  $|R|$ . Hence a method based on  $d_p$ -detection is unable to discriminate the sign of the Gaussian curvature [14] of the director profile at the sample surface, i.e. the sign of  $K_{24}$ .

From the point of view of the magnetic field-director coupling, it should be pointed out that increasing the intensity  $h$  of the reduced magnetic field always favours the occurrence of PHAN with respect to the P-undistorted structure, provided that the tilt-extrapolation length is not too large ( $L_{\theta_0} < 5 \mu\text{m}$ ) and the twist-extrapolation length is not too small ( $L_{\phi_0} > 0.5 \mu\text{m}$ ) at the same time. Note that the linearization method we used is a simple and powerful tool for obtaining the P-HAN and the P-PHAN threshold ( $d_a$  and  $d_p$ , respectively) : but it is unable to provide the HAN-PHAN threshold  $d_{ap}$ , which in principle could be greater than  $d_a$ . In fact,  $d_a$  represents just an asymptotical approximation of  $d_{ap}$  (when  $d_p \rightarrow d_a$ , then  $\beta \rightarrow 0$ ). In order to calculate  $d_{ap}$ , it is necessary either to consider the second variation of the cell free energy [3] or to solve the non-linear Euler-Lagrange equations with proper boundary conditions. The latter approach, based on a perturbation method, is under study, and will be published elsewhere.

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