Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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Online publication date: 11 November 2010

To cite this Article Casquilho, J. P. and Figueirinhas, J. L.(2002) 'Linear analysis of transient pattern evolution in the non-Freedericksz twist geometry', Liquid Crystals, 29: 1, 127 — 132
To link to this Article: DOI: 10.1080/02678290110093750
URL: http://dx.doi.org/10.1080/02678290110093750

LIQUID CRYSTALS, 2002, VOL. 29, NO. 1, 127–132

Linear analysis of transient pattern evolution in the non-Fréedericksz twist geometry

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(*Received 17 March 2001; in final form 1 August 2000; accepted 2 August 2001*)

We present a study of the evolution of the transient periodic pattern in the nematic director field reorientation in the magnetic non-Fréedericksz twist geometry. The stability of the uniform director field reorientation with respect to periodic perturbations is studied as a function of the magnetic field $\mathbf{H}$, the angle α between $\mathbf{H}$ and the initial homogeneous nematic director $\mathbf{n}_0$ ($\mathbf{H}$ not normal to $\mathbf{n}_0$) and the nematic viscoelastic parameters. The results predict that for $\alpha < \pi/2$, the amplitude of the periodic modes becomes damped after a critical time and eventually fade away and consequently does not give way to periodic inversion walls as in the Fréedericksz geometry ($\alpha = \pi/2$). Also for $\alpha < \pi/2$, it is predicted that the selected periodic modes have progressively smaller wave vectors as the director reorients back to equilibrium. The amplitude becomes damped earlier and the wave vector of the periodic pattern decreases faster with time when the magnetic field acts away from the normal to the initial director.

1. Introduction

The spontaneous formation and evolution of well-organized structures such as periodic patterns in non-equilibrium liquid crystals is a very active area of research in the field of the physics of these materials (see surveys in [1, 2]). The results are interesting both from the technological and the scientific points of view. Experimental and theoretical studies on transient patterns have been carried out mainly for nematic samples in Fréedericksz geometries [3–13] and rotating magnetic fields [14–18]. Studies on non-Fréedericksz geometries are less common [19–23]. In this work we attempt to obtain some insight into the evolution of the transient periodic pattern in the non-Fréedericksz twist geometry (for which the magnetic field is not normal to the initial nematic director field). A sealed sample between two parallel plates with planar boundary conditions and rigid anchoring and having a positive anisotropy of the magnetic susceptibility χ_a is studied.

The possibility of forming periodic patterns in the non-Fréedericksz twist geometry has already been studied using linear stability analysis [23]. A perturbation theory based on the linearization of the dynamic equations around the uniform director reorientation, taken at the instant when a magnetic field $\mathbf{H}$ is applied at an angle α with respect to the initially homogeneous director $\mathbf{n}_0$, allowed us to explain the formation of spatial periodic director structures in the non-equilibrium nematic sample.

In this work we generalize the perturbation theory described in [23] and we linearize the dynamic equations around the uniform reorientation taken at a time $t > 0$ after the magnetic field is applied. This method allows us to study qualitatively the time evolution of the periodic pattern for early times, before non-linear effects dominate as discussed in [7]. Our results predict that for $\alpha < \pi/2$, the periodic modes selected in each instant have successively smaller wave vectors as the reorientation proceeds. Also for $\alpha < \pi/2$, the initially increasing amplitude of the periodic modes is damped after a critical time and eventually vanishes. Consequently, it does not give way to inversion walls as in the $\alpha = \pi/2$ case but instead a uniform reorientation regime eventually develops. The amplitude is damped earlier and the wave vector of the periodic pattern decreases faster with time when the magnetic field acts away from the normal to the initial director. Our linear analysis allows us to obtain analytical

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Liquid Crystals ISSN 0267-8292 print/ISSN 1366-5855 online © 2002 Taylor & Francis Ltd
http://www.tandf.co.uk/journals
DOI: 10.1080/02678290110093750

expressions for the time dependent wave vector of the periodic pattern and of the amplitude critical time.

2. Theory

2.1. *Linear stability analysis of the magnetic reorientation*

Consider a bulk nematic aligned monodomain. The magnetic field $\mathbf{H}$ is applied at an angle α with respect to the initial homogeneous director $\mathbf{n}_0$. We study the stability of the uniform reorientation in respect to a (spatially) periodic reorientation where the director remains in the sample plane such that the distortion angle θ depends both on the height y and the position z along the axis defined by the initially unperturbed director. This corresponds to a twist–bend deformation of the director and to a pattern of periodic stripes normal to $\mathbf{n}_0$. The possibility of a dependence on x (splay–bend deformation) is studied in §2.2. Finally, an out of plane component of the director is expected only at later stages of the reorientation process [8, 13].

To study the dynamics of the director field in the case of a twist–bend deformation, the following magnetic, velocity and director fields are considered:

$$\begin{aligned} &H_x = H\sin\alpha,\ H_y = 0,\ H_z = H\cos\alpha \\ &v_x(y, z\ t),\ v_y = v_z = 0 \\ &n_x = \sin\theta(y, z, t),\ n_y = 0,\ n_z = \cos\theta(y, z, t). \end{aligned} \tag{1}$$

We now follow the method described in [23]: we write the Ericksen–Leslie equations [24] for the fields (1) and take the following functions for the velocity and the director fields:

$$\begin{aligned} v_x(y, z, t) &= 0 + v_0(t)\cos(q_y y)\sin(q_z z) \\ \theta(y, z, t) &= u(t) + \theta_0(t)\cos(q_y y)\cos(q_z z). \end{aligned} \tag{2}$$

In the rhs of equations (2) the first terms correspond to the uniform reorientation and the second terms to the perturbations of the velocity and the director fields respectively, where q_y and q_z are the y and z components of the wave vector of the distortion, with $q_y = \pi/d$ where d is the sample thickness in the OY direction. This analysis is consistent with the planar boundary conditions at $y = \pm d/2$ used in the study of the twist Fréedericksz geometry [5, 7].

Following standard stability analysis, we obtain the variational equations up to first order in the perturbations. The terms that cancel out in the variational equations correspond to the uniform reorientation equation [23] (see the Appendix). Next, we obtain the linearized equations for the perturbations around the unperturbed state $u(t)$. Here, the variational equations are taken around the uniform reorientation state at $u(t \ll \tau_0)$, where $\tau_0 = \gamma_1/\chi_a H^2$ is the uniform reorientation time. The solution of the uniform reorientation equation (A1)

$$u(t) = \alpha - \tan^{-1}[\tan\alpha\exp(-t/\tau_0)] \tag{3}$$

can be written for $t \ll \tau_0$

$$u(t) \cong \frac{1}{2}\sin 2\alpha\frac{t}{\tau_0}. \tag{4}$$

The result (4) shows that $u(t) = 0$ is a solution of equation (A1) at $t = 0$ (as taken in the zero order theory in $u(t)$ [23]) and that $t \ll \tau_0 \Rightarrow u \ll 1$ which allows the variational equations to be linearized in $u(t)$ (in the first order theory in $u(t)$ presented here). As discussed in [23], linear theories in $u(t)$ are only valid for $\pi/4 < \alpha \leqslant \pi/2$ and should be a good approximation for angles α not too far from $\pi/2$. Neglecting the inertial term in the velocity equation, the variational equations read

$$0 = -(\eta_a q_y^2 + \eta_c q_z^2)v_0 - \alpha_2 q_z\frac{d\theta_0}{dt} \tag{5}$$

$$\gamma_1\frac{d\theta_0}{dt} = -\alpha_2 q_z v_0 - a\theta_0 \tag{6}$$

where

$$a = K_2 q_y^2 + K_3 q_z^2 + \Gamma \tag{7}$$

with the magnetic torque Γ given by

$$\Gamma = \chi_a H^2\cos 2\alpha + [(\chi_a H^2\sin 2\alpha)^2/\gamma_1]t. \tag{8}$$

The equations (5, 6) are formally analogous to the zero order theory equations, but now in the magnetic torque (8) a time-dependent extra term appears. The material parameters are the Leslie viscosity coefficients α_i, $i = 2, \ldots, 5$, the rotational viscosity $\gamma_1 = \alpha_3 - \alpha_2$, the Miesowicz viscosities $\eta_a = \alpha_4/2$ and $\eta_c = (\alpha_4 + \alpha_5 - \alpha_2)/2$, and the twist and bend Frank elastic constants K_2 and K_3, respectively [24].

The substitution of equation (5) in (6) yields an equation for the amplitude of each mode. Its solution can be written

$$\theta_0(\rho_q^2, t') = A\exp\left\{-\frac{\gamma_1}{\gamma_{\mathrm{eff}}}\left[\begin{aligned}&\left(1 + \frac{K_3}{K_2}\rho_q^2 + h^2\cos 2\alpha\right)t' \\ &+ \frac{1}{2}(h^2\sin 2\alpha)^2(t')^2\end{aligned}\right]\right\} \tag{9}$$

where A is the initial amplitude, $\rho_q = q_z/q_y$ is the reduced wave vector of the periodic mode, $h = H/H_c$ is the reduced field where $H_c = (K_2\pi^2/\chi_a d^2)^{1/2}$ is the critical field for the aperiodic twist Fréedericksz transition [24], t' is a reduced time given by

$$t' = \frac{t}{\gamma_1/\chi_a H_c^2} \tag{10}$$

and γ_{eff} is an effective (linear) viscosity given by

$$\gamma_{\text{eff}} = \gamma_1 - \frac{\rho_q^2 \alpha_2^2}{\rho_q^2 \eta_c + \eta_a}. \quad (11)$$

The amplitude of the periodic modes given by equation (9) is damped for $t' > t'_c$, where t'_c is a critical (reduced) time determined by computing the maximum of the amplitude in t':

$$t'_c = -\frac{1}{h^2}\left(1 + \frac{K_3}{K_2}\rho_q^2 + h^2 \cos 2\alpha\right) \operatorname{cosec}^2 2\alpha. \quad (12)$$

The plot of t'_c as a function of α for a given value of h, $\rho_q^2 > 0$ and material parameters listed in the table, shows that for $\alpha < \pi/2$ the amplitude of the periodic distortion should fade away faster the more α is set away from $\pi/2$ (see figure 1). This is in agreement with experimental observations [25]. The divergence of the critical time for $\alpha = \pi/2$ as observed in figure 1 is of course non-physical: it is a consequence of the two-dimensional director field used in this theory, which leads at $\alpha = \pi/2$ to a (metastable) pattern of frozen splay–bend inversion walls when $t \to \infty$ [8].

Maximizing the growth rate of equation (9) in respect to ρ_q^2, one obtains the (squared) reduced wave vector corresponding to the fastest growing mode at each instant

$$\rho_q^2 = \frac{-\eta_a \eta_c \gamma_1 K_3 + [\alpha_2^2 \eta_a \eta_c K_3 \{\eta_a \gamma_1 K_3 - \eta_c \eta_{\text{bend}} K_2 \times [1 + h^2 \cos 2\alpha + (h^2 \sin 2\alpha)^2 t'/2]\}]^{1/2}}{\eta_c^2 \eta_{\text{bend}} K_3} \quad (13)$$

where $\eta_{\text{bend}} = \gamma_1 - \alpha_2^2/\eta_c$ is the effective viscosity associated with a pure bend mode [24]. This shows that for $\alpha < \pi/2$ the modes selected at each instant have smaller wave vectors as the magnetic reorientation proceeds. The wave vector of the periodic pattern starts at $t = 0$ from the zero order theory value [23], and the slope of the curve increases with the reduced magnetic field h and the angle α (see figure 2).

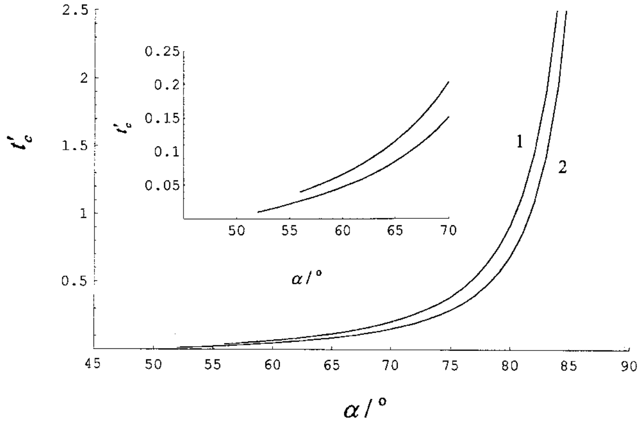

Figure 1. Plot of the critical reduced time t'_c given by equation (12) as a function of α with $h^2 = 5$ and $\rho_q^2 = \rho_q^2(t' = 0)$ given by equation (13) for the two material parameters sets listed in the table, for (1) the low molecular mass liquid crystal 5CB and (2) the polymer liquid crystal PBG. The critical angle below which the periodic pattern may not appear is given by $\cos 2\alpha_c = -1/h^2(1 + \eta_a \gamma_1 K_3/\alpha_2^2 K_2)$ [23], which gives for PBG, $\alpha_c = 52.2°$ and for 5CB, $\alpha_c = 55.6°$. The inset is a magnification of the main figure.

2.2. *On the possibility of oblique stripes*

To study the possibility of the formation of oblique stripes in the plane of the sample we will simplify our problem and consider $d \to \infty$, i.e. an infinite sample in the y direction. Thus we start with the following general

Table. Material parameters used in the numerical simulation.

Parameter	Material[a] 5CB [26]	PBG [7]
$\alpha_1/\text{g cm}^{-1}\,\text{s}^{-1}$	−0.066	−36.7
α_2	−0.77	−69.2
α_3	−0.042	0.20
α_4	0.634	3.48
α_5	0.624	66.1
$K_1/10^{-7}$ dyn	5.95	12.1
K_2	3.77	0.78
K_3	7.86	7.63

[a] 5CB = 4-n-pentyl-4′-cyanobiphenyl; PBG = poly(benzyl glutamate).

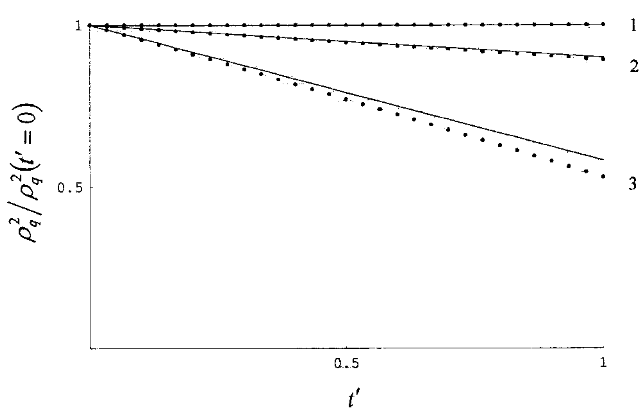

Figure 2. The reduced wavevector ρ_q^2 given by equation (13) is expanded as a Taylor series in the reduced time t' and the result is divided by the zero order term $\rho_q^2(t' = 0)$. This allows a direct comparison of its values for 5CB and PBG given in the table: $\alpha = 90°$ (1), 85° (2), 80° (3), with $h^2 = 5$. Lines PBG, dots 5CB.

director and velocity fields

$$v_x(x,z,t),\ v_y=0,\ v_z(x,z,t)$$
$$n_x=\sin\theta(x,z,t),\ n_y=0,\ n_z=\cos\theta(x,z,t) \tag{14}$$

corresponding to a reorientation of the director with a splay–bend deformation, and take the following perturbations of the uniform reorientation for the velocity and director fields:

$$\xi_{v_x}(x,z,t)=v_0(t)q_z\cos(q_xx+q_zz)$$
$$\xi_{v_z}(x,z,t)=-v_0(t)q_x\cos(q_xx+q_zz) \tag{15}$$
$$\xi_\theta(x,z,t)=\theta_0(t)\sin(q_xx+q_zz).$$

This description of the velocity obeys the incompressibility condition.

We now follow the general procedure described in [23] and obtain the following variational equations around $u(t=0)$ in canonical form, where the inertial term must be kept in the velocity equation because we are dealing with an infinite sample [23].

$$\rho(q_x^2+q_z^2)\frac{\mathrm{d}v_0}{\mathrm{d}t}=-\left(c-\frac{b^2}{\gamma_1}\right)v_0+\left(d+\frac{ab}{\gamma_1}\right)\theta_0 \tag{16}$$

$$\gamma_1\frac{\mathrm{d}\theta_0}{\mathrm{d}t}=-bv_0-a\theta_0 \tag{17}$$

where

$$a=K_1q_x^2+K_3q_z^2+\chi_aH^2\cos 2\alpha \tag{18}$$
$$b=\alpha_3q_x^2-\alpha_2q_z^2 \tag{19}$$
$$c=Nq_x^2q_z^2+\eta_bq_x^4+\eta_cq_z^4,\ N=\alpha_1+\eta_b+\eta_c \tag{20}$$
$$d=\frac{\gamma_2}{\gamma_1}q_xq_z\chi_aH^2\sin 2\alpha,\ \gamma_2=\alpha_3+a_2 \tag{21}$$

where in the term d in equation (21) we used $(\mathrm{d}u/\mathrm{d}t)_{t=0}$ given by equation (A7) with $f(y)=1$.

Proceeding as described in the Appendix of [23], we calculate the eigenvalues $\lambda_\pm$ of the stability matrix of the system, equations (16, 17). Numerical calculations with the material parameters listed in the table show that λ_- corresponds to a mode that decays with time for all values of the control parameters h and α. We now seek the selected wave vector maximizing the growth rate given by the eigenvalue λ_+ in respect to q_x and q_z. The results of numerical calculations for q_x and q_z with the values of the table show that $q_x, q_z\propto H^2$ and consequently the ratio q_x/q_z is field independent. From these results, as α departs from $\pi/2$ we get $q_x/q_z\ll 1$ for PBG and $q_x/q_z<1$ for 5CB (see figure 3). This suggests that the approximation of neglecting the x dependence should be much better for the polymeric LC than for the low molecular mass LC of the table.

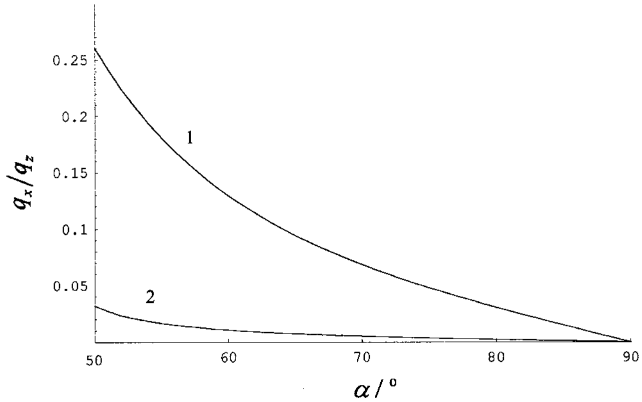

Figure 3. Ratio q_x/q_z as a function of the angle α for the parameters listed in the table: (1) 5CB, (2) PBG.

3. Conclusions

Our linear theory gives a physically correct qualitative picture of the magnetic reorientation mechanism: the more α is set away from $\pi/2$ or the more the director reorientation proceeds, the smaller the magnetic energy of the system; from which there results shorter living modes or modes selected in each instant that have smaller wave vectors. For a quantitative analysis one should take into account the non-linearities of the problem.

As the magnetic reorientation proceeds, the periodic modes selected at each instant have smaller wave vectors. The rate of decay of the wave vector of the periodic pattern increases the more α is set away from $\pi/2$. This decay is not predicted for $\alpha=\pi/2$, which is consistent with the fact that two-dimensional reorientation models like the one used in this work lead at $t\to\infty$ to frozen periodic inversion wall patterns. These are unstable to out of plane perturbations [8, 13], from which there results an evolution of the final homogeneous aligned state through a defect production mediated, roll pattern destruction [27]. The evolution towards equilibrium for $\alpha<\pi/2$ should involve a different mechanism, as predicted by our results: the amplitude of the periodic perturbation diminishes after a critical time and vanishes eventually, giving way to a final uniform reorientation regime. The more the magnetic field acts away from the normal to the initial director, the shorter the lifespan of the periodic pattern.

Appendix

On the effect of the boundary conditions on the non-periodic director reorientation

The uniform reorientation equation is [23]:

$$\gamma_1\frac{\mathrm{d}u(t)}{\mathrm{d}t}+\frac{1}{2}\chi_aH^2\sin 2[u(t)-\alpha]=0. \tag{A1}$$

In the description given in equation (2) the boundary conditions are taken into account for the periodic perturbations of the velocity and director fields. If the effects of the plates at $y = \pm d/2$ are to be studied also for the non-periodic reorientation, for the case studied here one obtains the following equation of motion

$$\gamma_1 \partial_t \theta + \frac{1}{2}\chi_a H^2 \sin 2(\theta - \alpha) - K_2 \partial^2_{yy}\theta = 0. \quad \text{(A2)}$$

We now show that the uniform solution $u(t)$ used in the linear theories presented here and in [23] is a good approximation for early times to the solution of equation (A2). We first linearize equation (A2); with the boundary conditions

$$\begin{aligned} \theta(0, y) &= 0 \\ \theta(t, \pm d/2) &= 0 \end{aligned} \quad \text{(A3)}$$

we can prove that the general solution of the resulting linear equation is

$$\begin{aligned} \theta'(t', y) = 1 &- \frac{\cosh(h'\pi y/d)}{\cosh(h'\pi/2)} \\ &+ \sum_{n=0}^{\infty} \frac{(-1)^n}{2n+1} \frac{-4(h')^2}{\pi[(2n+1)^2 + (h')^2]} \\ &\times \cos[(2n+1)\pi y/d] \exp\{-[(2n+1)^2 + (h')^2]t'\} \end{aligned} \quad \text{(A4)}$$

where t' is given by equation (10) and

$$\theta'(t, y) = \theta(t, y)/(1/2 \tan 2\alpha) \quad \text{(A5)}$$

$$h' = h(\cos 2\alpha)^{1/2} \quad \text{(A6)}$$

Plots of this solution for an angle $\alpha < \alpha_c$ (see figure 4) show that the uniform reorientation is a good approximation in the bulk up to $t' \sim 1$. The approximation is excellent for $t' \ll 1$ in both the cases $\alpha < \alpha_c$ and $\alpha > \alpha_c$; the latter case corresponds to the formation of the periodic structure. In the limit $t \to 0$ one obtains from equations (A4–6)

$$\frac{d\theta(0, y)}{dt} = \frac{1}{2} \sin 2\alpha \frac{\chi_a H^2}{\gamma_1} f(y);\ f(y) = \begin{cases} 0 & y = \pm d/2 \\ 1 & y \neq \pm d/2 \end{cases} \quad \text{(A7)}$$

in agreement with the derivative of equation (4).

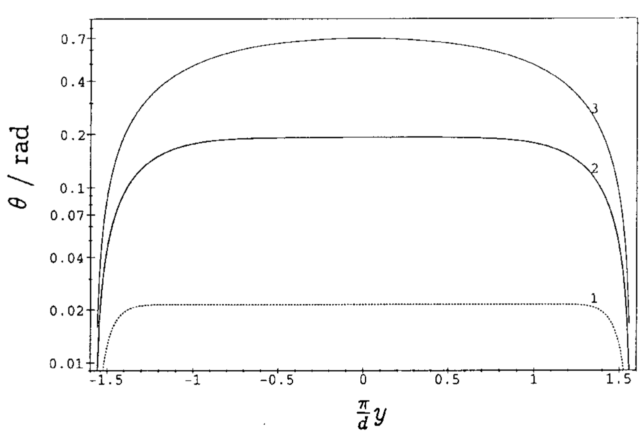

Figure 4. Plot of equation (A4). $t' = 0.01$ (1), $t' = 0.1$ (2), $t' = 1$ (3). Parameters of PBG are listed in the table, with $\alpha = 30°$.

This research was supported in part by the European Union under research contract FMRX-CT97-0121 and by PRAXIS XXI Program (Portugal) under contract No. 3/3.1/MMA/1769/95.

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