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Domain Structure in the Nematic Freedericksz Transition

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In the course of experiments using the Freedericksz transition on a planar nematic film in a magnetic field perpendicular to the limiting plates, we have observed the transient existence of a periodic roll structure very similar to published observations by Carr, and apparently similar to Williams domains. The present paper develops a model based on the effect of the decrease of the effective viscosity in the presence of the back flow, induced by the rotation of **n** and taking place as a convective roll pattern. Direct observations and order of magnitude estimates of the wavelength of the roll and of the time constant for the development of the rolls agree with the model. We also addressed ourselves with the problem of distortion in an initially well aligned bulk material placed in a field perpendicular to **n** and find that inertia effects can also lead to a roll structure in the problem.

I INTRODUCTION

The dynamics of the Freedericskz transition obtained when a uniformly aligned nematic liquid crystal (L.C.) film is subjected to a magnetic field, aligned perpendicular to the director **n**, has been extensively studied. In particular the "back flow" effect, which describes the velocity field induced by the rotation of the director ($d\mathbf{n}/dt$), has been found to lead to a reduction of the torque viscosity.[1] A similar problem arises in the case of the response to a high frequency electric field (this is the case if the dielectric anisotropy $\varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp} < 0$ is of the same sign as that of the magnetic susceptibility

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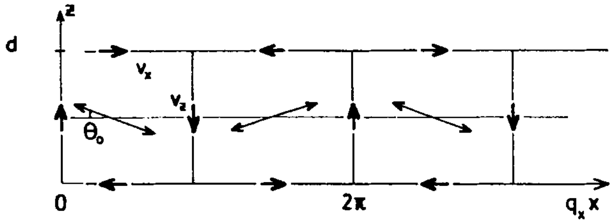

FIGURE 1 Defines the geometry. The variation of v_x, v_z, θ correspond to that given in (Eq. 5). In the unbounded medium problem ($d \to \infty$) one neglects v_x as well as the dependence on z. (Eq. 11).

$\chi_a = \chi_\parallel - \chi_\perp$; $\parallel$ and $\perp$ refer to directions with respect to **n**). The back flow causes, in the latter case, an oscillation of $\mathbf{n}^2$ when the field is suppressed which is responsible for a twinkling of dots in L.C. watches. In the analysis of (1) and (2) a uniform alignment in the plane of the film $\mathbf{n}(t, z)$ is assumed (the nematic film is bounded by parallel plates in planes $z = 0$ and d). In this case, the velocity field has components only parallel to the plates. In particular, if the initial (rest) state is *planar* ($\mathbf{n} \parallel x$), velocity gradients $\partial v_x/\partial z$ will develop during the transient state of reorientation in a field along z. This flow comes from the existence, in the Leslie stress tensor, of a term

$$\sigma_{zx} = (-\alpha_2 \sin^2 \theta + \alpha_3 \cos^2 \theta) \frac{\partial \theta}{\partial t}$$

where α_2 and α_3 ($\alpha_3 \sim -10^{-2}P$, $\alpha_2 \sim -0.5P$) are two viscosity coefficients and $\theta = \tan^{-1}(n_z/n_x)$. From the expression for the torque we see—in agreement with intuitive reasoning—that the back flow contribution will be largest when **n** is nearly perpendicular to the plates (to the flow): this is the case for a planar film when the distortion is large.[2]

There is however another possibility of back flow if we consider the growth of a distorted state periodically modulated along x, in a manner similar to that met in Williams domain instability,[3] far from saturation, of the form:

$$n_z(x, t) = n_{q_x} \cos q_x x \exp(s_{q_x} t)$$

such that σ_{xz} terms, and forces along z, $F_z = \partial_x \sigma_{xz}$, will develop. A roll circulation should develop as sketched in Figure 1.

We will see in fact that the reduction of viscosity due to the back flow is such that the fastest growth rate for the Freedericksz instability given by $ds_{q_x}/dq_x = 0$ is obtained for a finite value q_x^m, function of the applied field.

We will present in next paragraph a more detailed description of this result as well as experimental evidences for it. It is likely that the above mechanism explains the result of recent experiments by Carr[4] in a similar configuration both in electric and magnetic fields. Independently of our work, the probable connection between Carr's results and a transition controlled by back flow effects was mentioned in a review paper by Blinov.[5] Finally, we address ourselves to the possibility of observing similar structures in an unbounded sample. The role of inertia prevents the existence of back flow for very large wavelengths, and insures an optical finite wavelength of the growth of distortion.

II DISTORTION IN A FINITE SAMPLE

a Model

The geometry is sketched on Figure 1. By using the form of the Leslie stress tensor σ_{ij}[6] in the Navier Stokes equation

$$\rho \frac{\partial v_i}{\partial t} = -\frac{\partial p}{\partial x_i} + \frac{\partial \sigma_{ji}}{\partial x_j}$$

where p is a pressure term, the solution of the two dimensional problem ($n_y = v_y = 0, \partial/\partial y = 0$) is

$$\rho \frac{\partial v_x}{\partial t} = -\frac{\partial p}{\partial x} + \eta_1 \frac{\partial^2 v_x}{\partial z^2} + \eta_4 \frac{\partial^2 v_z}{\partial x\, \partial z} + \eta_5 \frac{\partial^2 v_x}{\partial x^2} + \alpha_3 \frac{\partial}{\partial z}\left(\frac{\partial \theta}{\partial t}\right) \tag{1}$$

$$\rho \frac{\partial v_z}{\partial t} = -\frac{\partial p}{\partial z} + \eta_2 \frac{\partial^2 v_z}{\partial x^2} + \eta_6 \frac{\partial^2 v_x}{\partial x\, \partial z} + \eta_7 \frac{\partial^2 v_z}{\partial z^2} + \alpha_2 \frac{\partial}{\partial x}\left(\frac{\partial \theta}{\partial t}\right) \tag{2}$$

$$\eta_1 = (\tfrac{1}{2})(\alpha_4 + \alpha_6 + \alpha_3), \qquad \eta_2 = (\tfrac{1}{2})(-\alpha_2 + \alpha_4 + \alpha_5)$$

as well as

$$\eta_4 = (\tfrac{1}{2})(\alpha_4 + \alpha_6 - \alpha_3), \qquad \eta_5 = (\alpha_1 + \alpha_4 + \alpha_5 + \alpha_6),$$
$$\eta_6 = (\tfrac{1}{2})(\alpha_2 + \alpha_4 + \alpha_5), \qquad \eta_7 = \alpha_4$$

The η_j are viscosity terms expressed in terms of the 6 Leslie coefficients α_i. The last terms in (1, 2) express the coupling leading to back flow.

On the other hand, the balance equation of the x component of the torque is

$$\gamma_1 \frac{\partial \theta}{\partial t} = \chi_a H^2 \theta + K_1 \frac{\partial^2 \theta}{\partial z^2} + K_3 \frac{\partial^2 \theta}{\partial x^2} - \alpha_2 \frac{\partial v_z}{\partial x} + \alpha_3 \frac{\partial v_x}{\partial z} \tag{3}$$

We consider only the limit of small distortion ($\sin\theta \sim \theta = n_x$) appearing immediately after the vertical magnetic field is applied. K_1 and K_3 are restoring Frank elastic constants. The last two terms are viscous torques which oppose the distortion. We must also write the equation of continuity for the incompressible medium.

$$\operatorname{div} \mathbf{v} = 0 \tag{4}$$

The solutions of the coupled Eqs. 1 to 4 lead to the back flow effect discussed in this and the next paragraph.

We assume the following boundary conditions

$$\frac{\partial v_x}{\partial z}(0) = \frac{\partial v_x}{\partial z}(\mathrm{d}) = 0, \quad \text{free boundaries} \tag{4a}$$

$$v_z(0) = v_z(\mathrm{d}) = 0 \tag{4b}$$

$$\theta(0) = \theta(\mathrm{d}) = 0, \quad \text{strong planar alignment} \tag{4c}$$

compatible with the simple trial functions whose variation is indicated on Figure 1:

$$v_x = -v_0 q_z \sin q_x x \cos q_z z \exp st \tag{5a}$$

$$v_z = v_0 q_x \cos q_x x \sin q_z z \exp st \tag{5b}$$

$$\theta = -\theta_0 \sin q_x x \sin q_z z \exp st \tag{5c}$$

with $q_z = \pi/\mathrm{d}$.

We can neglect the inertia terms in Eqs. (1) and (2): the dynamics governed by the viscoelasticity of the nematic is slow in comparison to that measured by viscous diffusivity η/ρ.

We can get rid of the pressure terms in 1, 2 by taking respectively partial derivatives relative to z and x. This leads to

$$v_0 \sin q_x x \sin q_z z\,(\eta_1 q_z^4 + \eta_2 q_x^4 + N q_x^2 q_z^2) = s\theta_0(-\alpha_2 q_x^2 + \alpha_3 q_z^2) \sin q_x x \sin q_z z \tag{6}$$

with $N = \eta_5 - \eta_6 + \eta_7 - \eta_4$ which is a linear relationship between the amplitudes v_0 and θ_0. The torque equation can be expressed as

$$0 = (\gamma_1 s - \chi_a H^2 + K_1 q_z^2 + K_3 q_x^2)\theta_0 \sin q_x x \sin q_z z + (\alpha_2 q_x^2 - \alpha_3 q_z^2) v_0 \sin q_x x \sin q_z z \tag{7}$$

The compatibility relation between the homogeneous Eqs. (6, 7) leads to

$$s = \frac{\chi_a H^2 - K_1 q_z^2 - K_3 q_x^2}{\gamma_1 - \dfrac{(\alpha_2 q_x^2 - \alpha_3 q_z^2)^2}{\eta_1 q_z^4 + \eta_2 q_x^4 + N q_x^2 q_z^2}} \tag{8}$$

If the distortion is homogeneous ($q_x = 0$), we recover the results of Ref. 1:

$$s = \frac{\chi_a H^2 - K_1 q_z^2}{\gamma_{1s}^*} = \frac{\chi_a (H^2 - H_{c_1}^2)}{\gamma_{1s}^*} \tag{9}$$

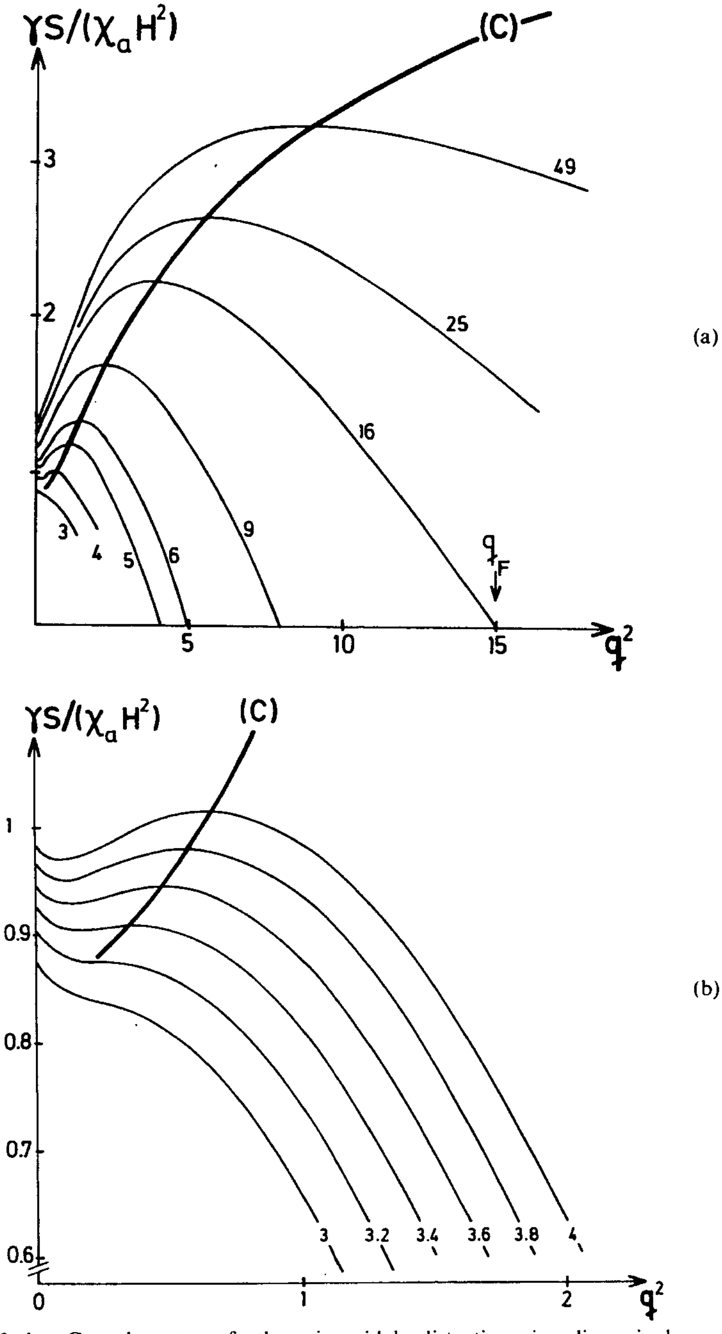

FIGURE 2a,b Growth rate of the sinusoidal distortion in dimensionless units $s\gamma_1/(\chi_a H^2)$. The values q_F such that $s = 0$ correspond to a Freedericksz threshold condition $[H_c(q_F)/H_c]^2 = 1 + q_F^2$. The curve for fastest growth rate (c) leads to a determination of the optimal wave vector q_c. Figure 2b is an expanded view around the origin.

The effective splay viscosity $\gamma_{1s}^* = \gamma_1 - \alpha_3^2/\eta_1$ is reduced typically by a few percent from γ_1 due to the back flow coupling.[1] The growth rate vanishes in a continuous manner when the Freedericksz critical field $H_{c_1} = (K_1 q_z^2/\chi_a)^{1/2}$ is approached from above. In the more general case considered here, the Eq. (8) can be written

$$s = \frac{\chi_a H_c^2[(H/H_c)^2 - 1 - q^2]}{\gamma_1 - \dfrac{(\alpha_2 q^2 - \alpha_3)^2}{\eta_1 + Nq^2 + \eta_2 q^4}} \tag{10}$$

where $q = q_x/q_z$. Note that the α_3 contribution is negligible except for very long wavelength ($q^2 < \alpha_3/\alpha_2$) solutions. We have assumed $K_1 = K_3$, as is well verified experimentally.

The solution of the equation for $s(q)$ is given in Figure 2a,b for various values of the ratio $(H/H_c)^2$ in units of $s\gamma_1/\chi_a H^2$ (q^2)†. For each values of the reduced field $H/H_c > 1$ there is an optimal wave vector $q_{xc} = q_c q_z = q_c(\pi/d)$ corresponding to the fastest growth of the distortion. If we assume that, at time $t = 0$, the fluctuations of all wave vectors q_x are of comparable importance, the growth of the distortion will take place with the wave vector q_{xc}.

An amusing feature can be observed on Figure 2b. For ratios $(H/H_c)^2 < 3.2$ the fastest growth rate is obtained for $q = 0$, i.e. for a homogeneous distortion. For larger ratios $3.2 < (H/H_c)^2 < 3.6$, the growth rate is still largest for $q = 0$ but there is a local maximum for a finite value q_c. The maximum becomes larger than that for $q = 0$, which eventually disappears for larger field (around $(H/H_c)^2 = 6$). This is equivalent to Landau diagrams with a hysteresis first-order transition. Note however that the numerical calculation was carried out assuming $\alpha_3 = 0$, a source of some error for very low $q^2(<(\alpha_3/\alpha_2) \sim 10^{-2})$.

b Experimental

The experimental set up is indicated in Figure 3. The liquid crystal (MBBA) is contained between parallel transparent sapphire plates separated by a 5 mm spacer. The plates have been treated by evaporating obliquely a SiO film to ensure planar alignment. We have also done experiments with a free upper surface (leaving a small gap to the upper plate) in order to get closer to the boundary conditions of the simple calculation above, which assumed free boundaries. (In MBBA the boundary condition at a free surface is nearly planar). Temperature gradients were minimized by circulating water at the same temperature at the outer surfaces of both plates. This is essential as

† The growth rate vanishes for $(H_c(q)^2/H_c) = 1 + q^2$ where $H_c(q)$ is the Freedericksz threshold in the presence of the q_x and q_z distortion. It goes to the limit (9) when $q \to 0$.

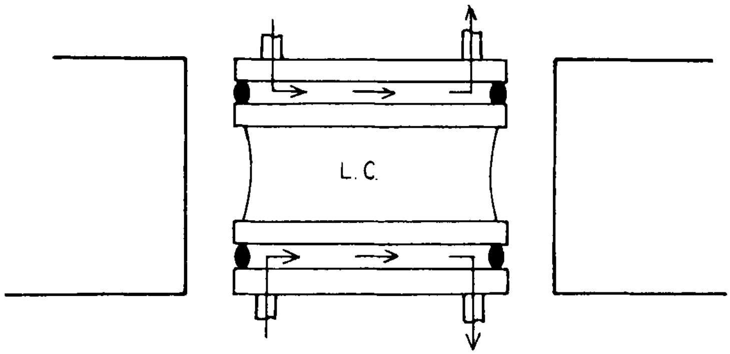

FIGURE 3 Geometry of the experiment.

thermal convective instabilities would take place with very low thresholds ($\Delta T < 0.1°$) for such thicknesses.[8]

To ensure uniform alignment across the cell, we first apply a 500 G horizontal magnetic field. At time $t = 0$, it is switched off and a vertical field $H(0 < H < 950$ G) is applied. The field is carefully aligned parallel and perpendicular to the plates as, otherwise, growth of one type of domains, say $+\theta$ (or $\langle\theta\rangle_z > 0$), would be favored. Transient periodic rolls are formed, very analogous to the structures observed by Carr[4] and called by him a "wall" structure. In agreement with Carr's findings we observe the following features: the distortion, which we follow qualitatively from the intensity of the scattered light due to the formation of the rolls, grows with a characteristic time t_c (connected with the maximum growth rate of Figure 2).

For $d = 2$ mm, $t_c \sim 10$ min for $H = 200$ G and $t_c \sim 1$ min for $H = 500$ G. The values agree qualitatively with the results of Figure 2 ($s^{-1} \sim 2$ min for $H/H_c = 4$). As the time progresses and the distortion saturates, the domain structure disappears. This takes place by the motion of walls separating $\pm\theta$ domains across the cell along the direction y, normal to the director. This can be a very slow process, especially when the sample is enclosed between two fixed boundaries such that the relaxation of defects is more difficult than with a free upper surface. It is clearly beyond the present continuum description, valid for small distortions. The wavelength of the very regular structure parallel to the y axis is measured before the distortion saturates. Figure 4 indicates some experimental results which show the decrease of the wavelength of the rolls as H increases, already noticed by Carr. We have also given the value of the optimal wave vector deduced from the results of Figure 2. The agreement is surprisingly good if one considers the crudeness of the model with respect to the actual boundary conditions. However it was found in Ref. 1, in an exact solution applicable to a sample with fixed boundaries, that

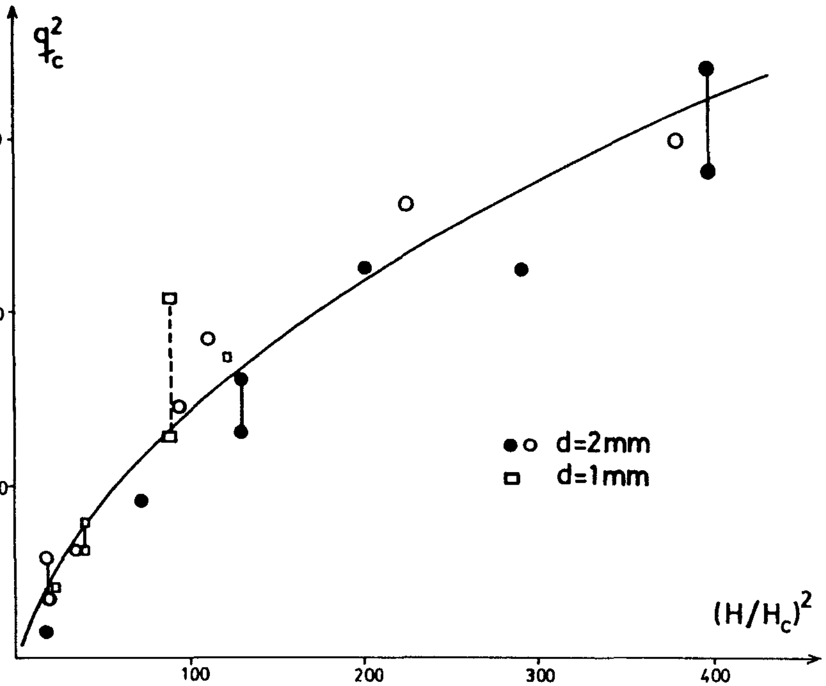

FIGURE 4 Variation of the optimum wave vector (maximums on Figure 2) versus field. Dots give experimental results obtained for small distortions.

the essential modification introduced by imposing rigid boundaries takes place in the inversion boundary layers of small thickness.

We have observed that a flow pattern compatible with the Figure 1 is indeed associated with the transient. The flow is most easily detected in the free surface configuration where the back flow reduction is largest (the displacement is a fraction of a millimeter for the values mentioned above).

We have also found, in accordance with the findings of Figure 2b that for small enough values of $(H/H_c)^2$—typically smaller than 3—the Freedericksz transition took place without any formation of domains. However, a uniform transient flow pattern was observed on the liquid crystal surface along the x direction.

III UNBOUNDED MEDIUM

We pursue the above analysis somewhat further by considering the transition of a large sample, initially uniformly aligned by the application of a magnetic field along x, when the field is suddenly rotated to the z direction. We investigate the possibility that, once again, the initial symmetry along x is broken by the formation of a regular array of $\pm\theta$ domains accompanied by a transient flow. More precisely, we look for solutions, describing the initial distortion, of the form

$$\begin{cases} v_z = v_0 \sin q_x x \exp st & \text{(11a)} \\ \theta = \theta_0 \cos q_x x \exp st & \text{(11b)} \end{cases}$$

We neglect the z dependence and, in particular, the end effects which provide the continuity of the flow pattern.

It is essential in this case to retain the inertia terms ($\rho(\partial/\partial t)$) in the Eqs. (1, 2) as it will be this contribution which will limit the possibility of obtaining uniform alignment and uniform shear ($\partial v_z/\partial x$) over too large distances x. The Eqs. (2) and (3) write

$$\rho \frac{\partial v_z}{\partial t} = +\alpha_2 \frac{\partial}{\partial x}\left(\frac{\partial \theta}{\partial t}\right) + \eta_2 \frac{\partial^2 v_z}{\partial x^2} \tag{12}$$

$$\gamma_1 \frac{\partial \theta}{\partial t} = \chi_a H^2 \theta + K_3 \frac{\partial^2 \theta}{\partial x^2} - \alpha_2 \frac{\partial v_z}{\partial x} \tag{13}$$

By substituting (11) in (12) we get

$$v_0 = -\left(\frac{\alpha_2 q_x s}{\rho s + \eta_2 q_x^2}\right)\theta_0 \tag{14}$$

(We can check that it is the presence of the inertia term ρs which prevents the divergence of v_0 for small wave vector q_x). By substituting (14) in (13), we finally have a relation between s and q_x

$$s\gamma_1\theta_0 = \chi_a H^2\theta_0 - K_3 q_x^2\theta_0 = \frac{\alpha_2^2 q_x^2 s}{\rho s + \eta_2 q_x^2}\theta_0 \tag{15}$$

whose solution is

$$s_\pm = \frac{\rho\chi_a H^2 + q_x^2 A \pm [(\rho\chi_a H^2 + q_x^2 A)^2 + 4\rho\gamma_1\eta_2 q_x^2(\chi_a H^2 - K_3 q_x^2)]^{1/2}}{2\rho\gamma_1} \tag{16}$$

with

$$A = \alpha_2^2 - \eta_2\gamma_1 - \rho K_3 \approx \eta_2\left(\frac{\alpha_2^2}{\eta_2} - \gamma_1\right) \approx -\eta_2\gamma_{1b}^*$$

where γ_{1b}^* is the bend effective torque viscosity reduced by the back flow effect.[7]

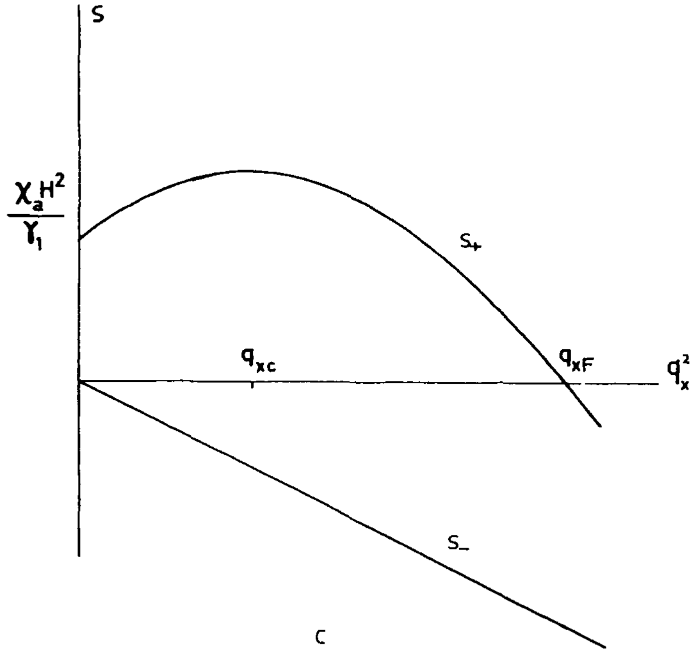

FIGURE 5 The two branches of the solutions of (Eq. 4) for any given value of field H. For the branch of positive growth rate s_+, a maximum is obtained for a value q_{xc} intermediate between the homogeneous solution ($q_x = 0$) and the upper Freedericksz limit q_{xF}.

The solutions of Eq. (16) are given in Figure 5. For small q_x, (16) approximates as

$$s_+(q_x) \sim \frac{\chi_a H^2}{\gamma_1} + \frac{(\alpha_2^2 + \gamma_1\eta_2)q_x^2}{\rho\gamma_1} > \frac{\chi_a H^2}{\gamma_1}$$

$$s_-(q_x) \sim -\frac{\eta_2 q_x^2}{\rho}$$

The lower branch (s_-) describes essentially a viscous decay of the flow field and does not concern us here.

The upper one describes the growth rate controlled by backflow. When $q_x \to 0$ it extrapolates to the classical result with the full viscosity γ_1 (no back flow). For an upper wave vector q_{xF} defined by a Freedericksz condition $(H_{c_3}(q_x) = K_3 q_{xF^2}/\chi_a$, $s_+(q_{xF}) = 0$ as the magnetic destabilizing energy becomes smaller than the elastic one for wavevector q_x larger than q_{xF} and would correspond to decaying solutions ($s_+(q_x) < 0$). There is a maximum optimal growth rate for an intermediate $q_{xc}(0 < q_{xc} < q_{xF})$: we should expect, as in a previous paragraph, that the Freedericksz transition will involve a periodic solution along x.

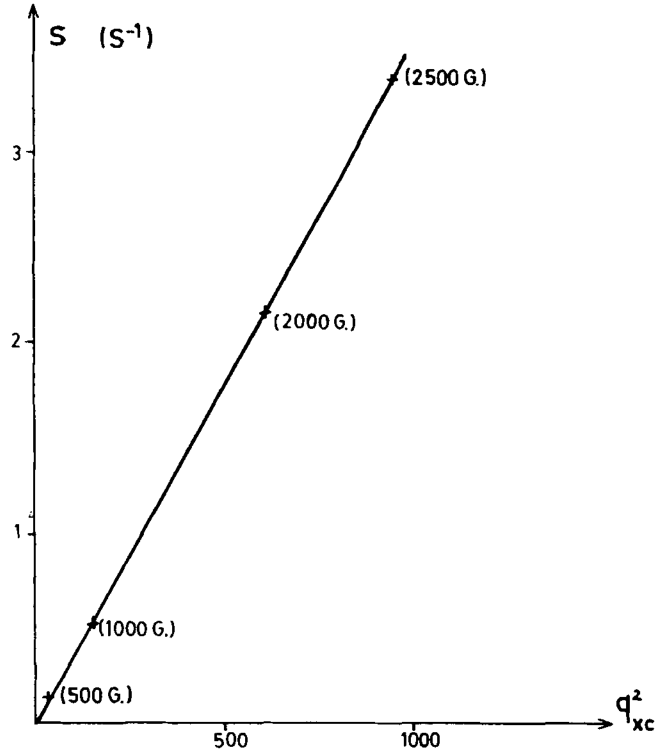

FIGURE 6 Growth rate for the optimal wave vector q_{xc}(cm^{-1}) for different values of field.

We can obtain an explicit expression for q_{xc} by looking for an extremum in Eq. (16) for s_+. We get

$$q_{xc}^2 = \chi_a H^2 \frac{-2\rho K_3(\gamma_1\eta_2 + A) + [4\rho K_3 A(\gamma_1\eta_2 + A)(A + 4\rho K_3)]^{1/2}}{K_3(A^2 - 4\rho\gamma_1\eta_2 K_3)} \tag{17}$$

We can easily get the order of magnitude of the above expression

$$q_{xc}^2 = \frac{2\chi_a H^2}{\gamma_1}\frac{\alpha_2}{\eta_2}\sqrt{\frac{\alpha_2}{K}\frac{\rho}{\eta_2}} \sim \frac{1}{D_e \tau}$$

The above expression simply states that over the time characteristic of the distortion $\tau[\sim\gamma_1/(\chi_a H^2)]$ the length explored q_{xc}^{-1} is controlled by a diffusive process taking into account both viscous and viscoelastic contributions: the diffusivity D_e is the geometric mean between the diffusivity of orientation $K/\alpha_2 \sim K/\gamma_1$ and the viscous one η_2/ρ.

For $H = 10^3$ G we get $2\pi/q_{xc} \sim 0.5$ cm. More exact results using the viscosity and elastic constants of MBBA are given in the Figure 6. Such an effect should be observable provided one uses a large enough liquid crystal cell.

IV CONCLUSION

In this work we have seen how the coupling between flow and distortion in the dynamics of the Freedericksz problem can lead to organized transient periodic structures. Another approach had been given previously by Léger and Brochard[9] who studied the development of walls separating $\pm\theta$ domains in similar conditions. The width of the walls is controlled by the magnetic correlation length $\xi = (K/\chi_a H^2)^{1/2}$, as can be verified dimensionally in Eq. 3.

This structure is obtained even if back flow is neglected. The geometry of the domains is determined by the local disorientations of the director in the initial state. The possibility of observing one or the other state should depend crucially on the quality of the initial alignment. However, as time proceeds, the solution with a sinusoidal distribution of orientation we have discussed here is not the most stable one and walls of a width ξ will form separating domains of uniform alignment (note that the dynamics of the wall formation itself is controlled by back flow!).

The present problem presents a superficial analogy with the spinodal growth:[10] starting from a solution AB cooled below a critical unmixing point, the phase separation will involve a periodic structure of the two phases A^* and B^*. The elastic energy which limits the wave vector of the distortion is replaced by the limitation by diffusion in the spinodal problem. However, the analogy is incomplete. There is no conservation law[11] for the species in the Freedericksz problem (a $+\theta$ distortion can become a $-\theta$ one) unlike the spinodal problem with conserved species A^* and B^*.

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