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Williams Domains and Dielectric Alignment in a Nematic Liquid Crystal†

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We investigate here the threshold behaviour of Williams domains and of the dielectric alignment in homeotropic and parallel oriented nematic films for different signs and magnitudes of the dielectric anisotropy. The theoretical predictions of the Helfrich theory are in agreement with the experiment.

INTRODUCTION

A nematic liquid crystal in an electric field shows at least four instabilities: the Williams domains[1] and the Freedericksz transition[2] or the diclectric alignment, the Chevrons[3,4] and the dc instability.[4] In this paper we will discuss only the Williams domains and the dielectric alignment. The Williams domains are theoretically discussed here for dc excitation since the frequency dependence is discussed by Dubois-Violette *et al.*[5]

Dielectric alignment

Figure 1(a) shows the usual geometry – a sandwich cell. The local optical axis is described by the director $L(|L| = 1)$. The applied electric field E produces a

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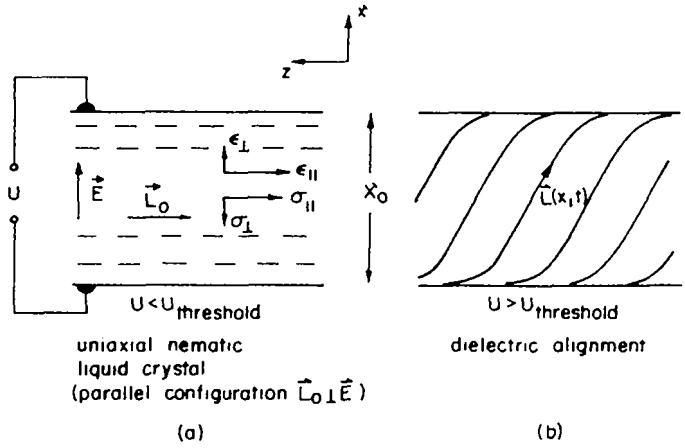

FIGURE 1 Illustration of the geometry of (a) a sandwich cell and (b) the dielectric alignment. The full lines in (b) indicate the curvature of the optical axis.

torque density, which tends to align the optical axis parallel to the electric field because the dielectric constant parallel to the optical axis $\epsilon_\parallel$ is in this case larger than the dielectric constant perpendicular to the optical axis $\epsilon_\perp$. However, the elastic torque density tends to bring the deformed sample into the undeformed configuration. In steady state these torques are equal and define the orientation of the local optical axis $L(x)$ (Figure 1(b). The threshold value is defined by $E_{0\epsilon} = \lim_{L \to L_0} E$.

This dielectric alignment occurs in an electric conducting as well as in an insulating nematic liquid crystal. For a deformation as is shown in Figure 1(b), no periodic space can be built up in steady state; therefore, the threshold value is independent of the electric conductivity of the nematic liquid crystal. This holds as long the dielectric constants as well as the elastic constants are independent of the electric conductivity.

Williams domains

The deformation of the local optical axis for the Williams domains is shown in Figure 2a. This type of deformation is periodic, and, therefore it is possible to build up electric space charges, which arise from the anisotropy of the electric conductivity and the dielectric constant. This electric space charge in the electric field produces a fluid flow which contributes to the torque density. In steady state the sum of the electrostatic, the elastic, and the viscous torque density is

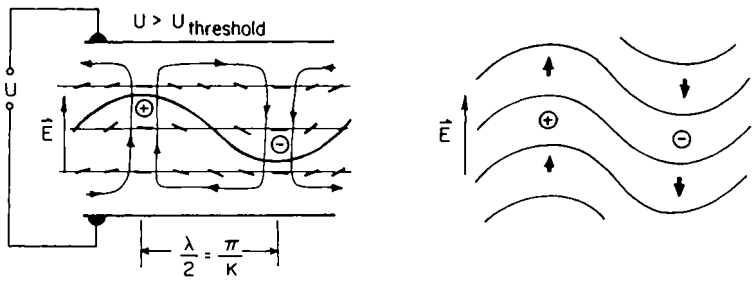

FIGURE 2 Illustration of the Williams Domains in (a) a sandwich cell and (b) without boundaries–Helfrich's model. The double arrows indicate the fluid flow.

zero. From this condition one can derive the orientation of the local optical axis L(x,z) The threshold value is again defined by $E_{0\sigma} = \underset{L \to L_0}{\text{Limit to}} E$.

WILLIAMS DOMAINS

The Williams domains have a one-dimensional periodicity as we have mentioned in the introduction. The stripes of the Williams domains are usually perpendicular to the undisturbed optical axis. This means that the director L is always in the plane which is defined by the director of the undisturbed optical axis and the applied electric field E[1,3,6,8]. (Near the smectic C phase the director can be out of this plane, since Rondelez[23] observed that the stripes of the Williams domains have an angle ($\neq \pi/2$) with the undisturbed optical axis.)

The differential equations which describe the director L and the fluid flow as a function of position and time, is given by several others.[5,6,7] The solution for the Williams domains has now the following form

$$L = (\sin \phi, 0, \cos \phi); \phi(x,z,t)$$

$$v = (v_x, 0, v_z); v_x(x,z,t), v_z(x,z,t)$$

We simplify the problem and use Helfrich's model[9] which neglects all boundaries (see Figure 2b). By doing this step we loose information about the periodicity of the deformation. The spatial dependence of the deformation angle and the fluid flow is now one dimensional:

$$\phi(z,t), \; v_x(z,t) \text{ and } v_z(z,t)$$

The first order treatment of the differential equations are satisfied by

$$\phi = \phi e^{-t/\tau + i(Kz + wt)} \tag{1a}$$

$$v_x = v_{x_0} e^{-t/\tau + i(Kz + wt)} \tag{1b}$$

$$v_z \propto \phi^2$$

This type of solution describes how a given deformation changes with time and space. The time constant τ describes the increase ($\tau < 0$) or the decrease ($\tau > 0$) of a given deformation. The space dependence of the deformation is given by the wave vector K. A transverse movement of the pattern is described by the frequency, w. ϕ_0 and v_{x0} are the maximum deformation angle and the maximum fluid flow. Since ϕ_0 and v_{x0} are complex, it is possible to have a phase difference between de deformation and the fluid flow.

If we put the expressions of Eq. (1a) and (1b) into the linearized differential equation (see, for example, Ref. 5), then we get the time constant τ as a function of the applied field from the real part of the solution. From Helfrich's model (Figure 2b) we get no information about the wave vector K, because no boundaries are considered. Penz and Ford[6] considered the Williams domains with electrodes (see Fifure 2a) and were able to get information about the wave vector K.

It is impossible to get a saturation value of ϕ_0 and v_{x0} from the linearized differential equations (first order treatment), but the dynamic behaviour of the Williams domains for small deformation angle and for small fluid flow can be considered.

We get for the case of electrostatic equilibrium, $|\tau| < \frac{\epsilon_\parallel \epsilon_0}{\sigma_\parallel}$, for a parallel oriented nematic liquid crystal film (see Fig. 1(a), $E \perp L_0$), the following dispersion relation:

$$\left(\frac{1}{\tau}\right)^2 (\alpha_3 - \alpha_2)\,\rho + \left(\frac{1}{\tau}\right)\left\{\rho K^2 \cdot k_{33} - \left(\frac{\sigma_\perp}{\sigma_\parallel}\right)\rho \cdot \Delta\epsilon \cdot E^2 - \frac{1}{2}K^2(\alpha_4 + \alpha_5 - \alpha_2)\right.$$

$$\left.(\alpha_3 - \alpha_2) + K^2 \cdot \alpha_2 \rho\right\} + \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2) \cdot K^2 \left\{\Delta\epsilon\left(\frac{\sigma_\perp}{\sigma_\parallel}\right)E^2 - K^2 k_{33}\right.$$

$$\left. - \left(\frac{2\alpha_2}{\alpha_4 + \alpha_5 - \alpha_2}\right)\left(\Delta\epsilon - \epsilon_\parallel\left(\frac{\Delta\sigma}{\sigma_\parallel}\right)\right)E^2\right\} = 0. \tag{2a}$$

Values for the viscosities α_i $(i = 1,6)$, the mass density ρ, the electrical conductivities σ, the dielectric constants ϵ and the elastic bend constant k_{33} are given in Ref. 5,6,7 and 9.

This equation simplifies for small electric field strength to

$$\left(\frac{1}{\tau}\right) = K^2 k_{33} \left(\frac{2\alpha_2^2}{\alpha_4 + \alpha_5 - \alpha_3}\right) + (\alpha_2 - \alpha_3)^{-1} \left(\left(\frac{E}{E_{0\sigma}}\right)^2 - 1\right) \tag{2b}$$

The threshold value $E_{0\sigma}$ is defined by $\left(\frac{1}{\tau}\right) \to 0$, because for field strength larger than $E_{0\sigma}$ a deformation gets larger and for $E < E_{0\sigma}$ a deformation decreases. The threshold value $E_{0\sigma}$ was first calculated by Helfrich:[9]

$$\left(\frac{E_{0\sigma}}{K}\right) = \left\{ \frac{k_{33}}{\epsilon_0 \epsilon_\parallel \left[\frac{\Delta\epsilon}{\epsilon_\parallel} \frac{\sigma_\perp}{\sigma_\parallel} - \frac{2\alpha_2}{\alpha_4 + \alpha_5 - \alpha_2} \left(\frac{\epsilon_\perp}{\epsilon_\parallel} - \frac{\sigma_\perp}{\sigma_\parallel}\right)\right]} \right\}^{\frac{1}{2}} \tag{3}$$

Penz and Ford[6] found for the two dimensional model (Figure 2a) that K^{-1} is proportional to the sample thickness. This means that the threshold value $2\pi(E_{0\sigma}/K)$ is a threshold voltage $U_{0\sigma}$ and independent of sample thickness.

From the imaginary part of the solution, we get for the dc limit, $w = 0$. This means that in the linearized theory of dc excitation there is no transverse motion of the pattern.

Dubois-Violette *et al.*[5] treated the differential equations in a different manner and found two types of time constants: the dielectric relaxation time for charges and the relaxation time for a sinusoidal curvature. By considering these time constants they found two types of instabilities: the Williams domains and the Chevrons.

The relation between the maximum of the fluid flow and the maximum of the deformation is

$$\frac{v_{x^0}}{\phi_0} = i K \frac{k_{33}}{\alpha_2} \left\{ \frac{\Delta\epsilon}{k_{33}} \frac{\sigma_\perp}{\sigma_\parallel} \left(\frac{E}{K}\right)^2 - 1 + \frac{\left(\frac{E}{E_{0\sigma}}\right)^2 - 1}{\alpha - 1} \right\} - \tag{4}$$

$$\frac{w}{K\alpha_2} (\alpha_3 - \alpha_2)$$

with

$$\alpha = 2\alpha_2^2\ (\alpha_4 + \alpha_5 - \alpha_2)^{-1}\ (\alpha_3 - \alpha_2)^{-1}$$

For the case $w = 0$, the strength of the fluid flow is for a given deformation proportional to the wave vector K or inversely proportional to the sample thickness. The difference

$$\left(\frac{v_x^0}{\phi_0}\right) - \left(\frac{v_x^0}{\phi_0}\right)\ E = 0$$

is proportional to the square of the applied field.

For a certain electric field E_*, the fluid flow vanishes ($v_{x0} = 0$). From Eq. 4, we get

$$\left(\frac{U_*}{U_{0\sigma}}\right)^2 = \left(\frac{E_*}{E_{0\sigma}}\right)^2 = \frac{1 + \alpha^2}{\dfrac{\epsilon_0 \epsilon_\parallel}{k_{33}}\ \dfrac{\Delta\epsilon}{\epsilon_\parallel}\ \dfrac{\sigma_\perp}{\sigma_\parallel}\ (\alpha - 1)} \tag{5}$$

This means that the flow produced by the reorientation of the director L cancels the fluid flow produced by the space charge in the electric field. The deformation of the Williams domains decreases with time when $U < U_{0\sigma}$, but it is only the voltage $U = \left(\frac{E \cdot 2\pi}{K}\right)$ that the deformation vanishes without fluid flow.

For $w \neq 0$, the phase difference δ between the fluid flow and the deformation is given by

$$\cot \delta = \frac{w}{K^2}\ \frac{\alpha_3 - \alpha_2}{\Delta\epsilon\ \dfrac{\sigma_\perp}{\sigma_\parallel}\ \left(\dfrac{E}{K}\right)^2 - k_{33} - \dfrac{k_{33}\left(\left(\dfrac{E^2}{E_{0\sigma}}\right) - 1\right)}{\alpha - 1}} \tag{6}$$

This phase shift leads to a change of the position of fluid flow as well as of the deformation with time.

Since $K = \frac{2\pi}{\lambda}$ with λ comparable to the sample thickness, the phase shift is small in thin samples. This can be a reason why in thin samples the dynamic scattering mode is suppressed as Greubel and Wolf[10] and Vistin[11] have observed.

In the second order treatment of the differential equations, $v_z(z,t)$ can be non-zero, but the equation of conservation of mass

$$\rho + \rho\ \mathrm{div}\ v = 0$$

restricts the z-component of the fluid flow to a constant value $v_z(t)$. The periodic solutions in space for ϕ and v_x leads in the second order treatment to the following equation:

$$\rho\dot{v} + \phi_0^2 e^{-2t/\tau + i2(Kz + wt)} \cdot K \left\{ -w \frac{(2\alpha_1 + \alpha_5 + \alpha_6)(\alpha_3 - \alpha_2)}{\alpha_2} - \right.$$

$$2(\alpha_2 + \alpha_3) + i[E^2\epsilon_{\parallel} \frac{\Delta\sigma_{\perp}}{\sigma_{\parallel}} (\frac{\sigma_{\perp}}{\sigma_{\parallel}} - \frac{\epsilon_{\perp}}{\epsilon_{\parallel}}) + \frac{2\alpha_1 + \alpha_5 + \alpha_6}{\alpha_2}$$

$$\left. (\Delta\epsilon \frac{\sigma_{\perp}}{\sigma_{\parallel}} - k_{33} K^2 - \frac{\alpha_3 - \alpha_2}{\tau}) - 2 \frac{(\alpha_2 + \alpha_3)}{\tau}] \right\} = 0$$

Since v_z can not have a periodic solution, the real and the imaginary part of this equation have to be zero. This means, that $v_z = 0$. From the imaginary part of the equation, we get a new relation between the time constant τ and the applied electric field. The real part, however, is zero when $w = 0$.

The second order treatment of the differential equation does not lead to a further instability but the field dependence of the time constant is changed. The periodicity can also be changed, but in the Helfrich model we do not get information about it.

The third order treatment of the differential eqautions leads to a saturation value of the deformation as Carroll [12] has shown, and w also becomes non-zero. This means that the whole pattern of the Williams domains starts to move in one direction. This can be the beginning of the dynamic scattering mode.

Penz and Ford[6] calculated higher order modes besides the ground mode for the Williams domains. In this case there exist more than one vortex in the sample thickness. By applying a very high voltage as a step function, there is a certain probability that the deformation starts in a higher mode. But by applying a ramp voltage with a large time constant, the deformation starts in the lowest mode and it is unlikely that a change occurs to a higher mode after the initial deformation. Therefore, we consider it unlikely that the higher modes of the Williams domains can be an explanation for the occurrence of the dynamic scattering mode.

Dielectric alignment

The disturbed nematic film is shown for this type of instability in Fig. 1(b). We know from optical observation that the director L is always in the plane, defined by the undisturbed optical axis L_0 and the applied electric field E.[8] The defor-

mation depends only on one coordinate. The static case of this type of deformation was first solved by Saupe [13] for the magnetic analog. The static deformation for the electric case was solved by several authors.[14,15,16] The dynamic of this type of deformation is discussed by Pieranski *et al.* [17] for the magnetic analog.

The threshold voltage for a parallel oriented sample is

$$U_{0\epsilon} = \tau \sqrt{\frac{k_{33}}{\epsilon_0 \Delta\epsilon}}$$

Experimental results

Figure 3 shows the theoretically predicted threshold behaviour of the dielectric alignment and the Williams domains as a function of the dielectric anisotropy $\Delta\epsilon$ for a parallel (full lines) and a homeotropic (dashed lines) oriented nematic liquid crystal film. All the other material constants like elastic constants and viscosities are held constant. For this calculation, we used the values of MBBA.[18] Experimentally, one might accomplish this by making a mixture of

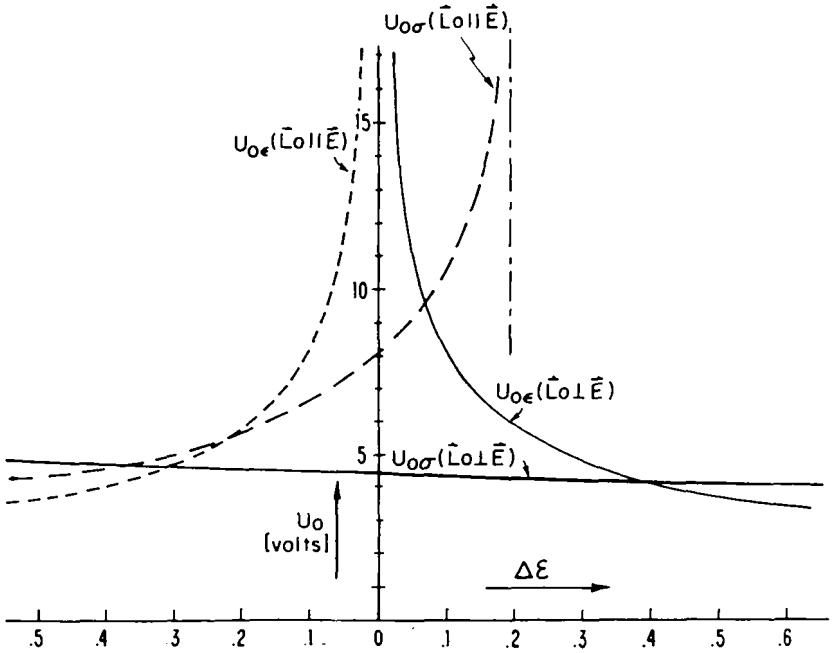

FIGURE 3 The calculated threshold voltage for the Williams Domains and for the dielectric alignment in the case of a parallel oriented sample (full lines) and in the case of a perpendicular oriented sample (dashed lines) as a function of the dielectric anisotropy $\Delta\epsilon$.

MBBA and a small amount of another compound with the proper dielectric anisotropy.

(I) $$\Delta\epsilon < 0 \textit{ and } L_0 \perp E$$

In the parallel oriented sample with a negative dielectric anisotropy only Williams domains were observed (Fig. 5, Ref. 8) as expected the dc instability and the Chevrons are still ignored.

For very large negative dielectric anisotropies the Williams domains can be surpressed since the dielectric torque is then for all electric field strength larger than the viscous torque which is driven by the electric space charges. This takes place for

$$\frac{\Delta\epsilon}{\epsilon_{\parallel}} \leq \frac{\Delta\sigma}{\sigma_{\parallel}} \quad \frac{1}{\frac{\sigma_{\perp}}{\sigma_{\parallel}} \frac{\alpha_4 + \alpha_5 - \alpha_2}{2\alpha_2} + 1}$$

For "MBBA" this negative dielectric anisotropy $-\Delta\epsilon/\epsilon_{\parallel}$ must be larger than -8.6. This is a pretty high anisotropy. In preliminary experiments, Wong[19] found that the Williams domains are surpressed by using a nematic liquid crystal where the molecules have a large dipole perpendicular to the molecular axis.

(II) $$\Delta\epsilon > L_0 \perp E$$

In the parallel oriented sample with a positive dielectric anisotropy, Williams domains as well as the dielectric alignment were observed as theoretically predicted. These two instabilities can be separated as is shown by Gruler and Meier[8] because the dielectric alignment shows below the dielectric relaxation frequency no frequency dependence whereas the Williams domains show a frequency dependence.[4,5]

The threshold voltage for the dielectric alignment $U_{0\epsilon}$ is for small dielectric anisotropy larger than the threshold voltage for the Williams domains (Fig. 7(a), Ref. 8) and for large dielectric anisotropy is $U_{0\epsilon} < U_{0\sigma}$ (Fig. 7(b), Ref. 8). The cross over between $U_{0\epsilon}$ and $U_{0\epsilon}$ is given by the following expression:

$$\frac{\Delta\sigma}{\sigma_{\parallel}} \left\{ 1 + \frac{\epsilon_{\parallel}}{\Delta\epsilon} \frac{2(-\alpha_2)}{\alpha_4 + \alpha_5 - \alpha_2} \right\} - 4 \frac{k_{33}}{k_{11}} + \frac{2(-\alpha_2)}{\alpha_4 + \alpha_5 - \alpha_2} \lessgtr 0$$

for

$$U_{0\sigma} \lessgtr E_{0\sigma} \cdot \lambda$$

The more rigorous calculation of Penz and Ford [20] shows that the threshold behaviour near the cross over is a smooth curve.

(III) $\Delta\epsilon > 0$ *and* $L_0 \parallel E$:

In a homeotropic oriented sample with a positive dielectric anisotropy and negative conductivity anisotropy, the theory predicts, as in case I, only the instability of the Williams domains. (The equations for the threshold voltages can still be used—only $\epsilon_{||}$, $\sigma_{||}$, k_{11} and $\frac{\alpha_2}{\alpha_4 + \alpha_5 - \alpha_2}$ are replaced by $\epsilon_\perp$, $\sigma_\perp$, k_{33} and $\frac{\alpha_3}{\alpha_3 + 2\alpha_3 + \alpha_4 + \alpha_5}$, respectively.) The instability of the Williams domains can be suppressed by a large positive dielectric anisotropy similar to case I. But in this case this effect takes place for relatively small dielectric anisotropies. The boundary value is, for "MBBA", $\Delta\epsilon/\epsilon_\perp = 0.036$. This small value can be the reason why Gruler and Meier[8] could not find a threshold value for a material, which has a dielectric anisotropy, $\Delta\epsilon$, of about 0.5 to 0.7. De Jeu, [21] however, found a threshold voltage, which was approximately 5 to 10 times larger than the threshold voltage in the parallel oriented sample (see Fig. 3).

(IV) $\Delta\epsilon < 0$ *and* $L \parallel E$:

In a hoeotropic oriented sample with a negative dielectric anisotropy similar to case II, two instabilities, $U_{0\epsilon}$ and $U_{0\sigma}$, were observed (Fig. 6(a) and 6(b), Ref. 8). The observation for large dielectric anisotropy is in agreement with the theory, but Gruler and Meier[8] observed a strange threshold behaviour for small dielectric anisotropy (see Fig. 6(a), Ref. 8). By increasing the voltage from zero to $U_{0\epsilon}$, nothing happened, but, for a voltage larger than $U_{0\epsilon}$, Williams domains appear. By decreasing the voltage, these Williams domains disappear at a voltage which is less than $U_{0\epsilon}$. This effect can be partly explained when we remember that the threshold voltage $U_{0\sigma}$ for the parallel oriented sample can be smaller than $U_{0\epsilon}$ (see arrow in Fig. 3). We know, furthermore, that for voltages a little above the threshold value, the maximum deformation angle in the middle of the sample is nearly $\pi/2$.[8, 14] Williams domains occur now in the middle of this deformed sample.[22] The threshold voltage $U_{0\sigma}$ for the parallel oriented sample can be used in order to estimate the threshold voltage of these Williams domains in a deformed homeotropic oriented sample.

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