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On the Theory of Domain Walls in Planar Nematic Films

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Abstract. — The theory of simple domain walls in planar nematic films is reconsidered taking into account azimuthal director rotations additionally. A perturbation treatment is useful to derive analytical equations which are asymptotically valid in the limit of small distortion amplitudes. In comparison to previously known approaches a more complete description leads to a correction in the formula for the wall thickness. This correction also influences the shape of closed domains and causes an array of walls in materials with high elastic anisotropy.

1. Introduction

A sufficient strong magnetic or electric field applied across a planar nematic film can induce distortions which grow continuously above a certain threshold [1]. Since this transition corresponds to a pitchfork bifurcation, two equally stable configurations differing only in the sense of the director rotation are possible. In well aligned samples there were found domain walls separating regions with clockwise and anticlockwise director rotation towards the applied magnetic or electric field [2]. Two examples of simple walls are shown in Figure 1 differing in the angle κ between the normal to the wall and the director orientation at the film surfaces. According to Brochard [3], the director configuration in a wall is described by $\theta(X, Y) = b_0 \cos(X\pi/d) \tanh(Y/\xi_0)$, where the angle θ is enclosed by the director and the plane $X = \text{constant}$, b_0 is the maximal rotation angle, d the film thickness and ξ_0 defines the thickness of a domain wall. In the mid-plane of the film $X = 0$ ($-d/2 < X < +d/2$) the director rotates from $-b_0$ (for $Y \rightarrow -\infty$) to $+b_0$ ($Y \rightarrow +\infty$). In accordance with experimental observation, the elastic continuum theory predicts that the width ξ_0 of walls diverges when the strength of the applied field approaches the Freedericksz-threshold [1–3].

It was found experimentally that closed domains have an elliptic shape [1, 2]. The ellipticity remains constant even if the domain is not stable but shrinks gradually. The original theory [3] postulates the simple relation $a/c = \sqrt{K_{33}/K_{22}}$, where a and c are the principal axes of the ellipse, while the elastic constants K_{22} and K_{33} for twist and bend distortions are defined in the framework of the elastic continuum theory [1]. This relation is derived neglecting a possible azimuthal director rotation inside the wall region. Actually, it was recently observed that besides the polar angle also the azimuthal angle is altered inside a domain wall [4]. This

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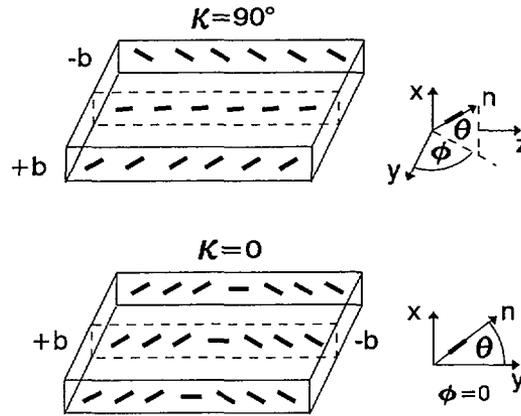


Fig. 1. — Two borderline cases for simple domain walls. For a twist-bend wall the y -axis is perpendicular to the director at the substrates ($\kappa \equiv \phi(X = \pm d/2) = \pi/2$), whereas in the case of the splay-bend wall shown below both directions are parallel ($\kappa = 0$). With the exception of the splay-bend wall, an azimuthal director rotation always occurs.

means that the ellipticity of the loops could be enlarged considerably as predicted by a rough estimation [5].

In this paper we reconsider the theoretical model for simple domain walls in planar nematic films taking into account all degrees of freedom of the director rotation. It turns out that there is generally an azimuthal director rotation inside a wall. This additional degree of freedom provides a correction to Brochard's model. For a high elastic anisotropy the correction becomes important for the appearance of a periodic array of walls in accordance with the theory of Lonberg and Meyer [6]. As long as the period is large compared to the film thickness and if the distortion amplitudes are sufficiently small an analytical mathematical model is applicable for simple and periodic domain walls.

2. Free Energy and Torque Balance

It is assumed that an a.c. voltage is applied at the bounding plates of the slab. The frequency of the alternating voltage should be sufficiently high for avoiding flexoelectric and hydrodynamic effects. Performing some minor modifications, the results obtained below for the electric field are also applicable to the case of a magnetic field. In the continuum theory of nematic liquid crystals, the director, denoted by a unit vector \mathbf{n} , defines the local optical axis of the uniaxial phase. The density of the free energy due to elastic distortions is represented as [1]

$$f_n = \frac{1}{2}K_{11}(\text{div } \mathbf{n})^2 + \frac{1}{2}K_{22}(\mathbf{n} * \text{rot } \mathbf{n})^2 + \frac{1}{2}K_{33}(\mathbf{n} \times \text{rot } \mathbf{n})^2 \quad (1)$$

where the coefficients K_{11} , K_{22} and K_{33} are the elastic constants related to splay, twist and bend deformations, respectively. If an electric field is applied, the corresponding free energy contribution is

$$f_e = -\frac{1}{2} \sum \varepsilon_{ij} E_i E_j \quad (2)$$

For the uniaxial nematic phase the dielectric tensor is defined by

$$\varepsilon_{ij} = \varepsilon_{\perp} \delta_{ij} + \Delta \varepsilon n_i n_j$$

where n_i are the director components in a Cartesian coordinate system, δ_{ij} is the unit matrix and $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$ defines the dielectric anisotropy, which is assumed to be positive in this paper. The dielectric constants ε_{\parallel} and ε_{\perp} are related to the field directions parallel and perpendicular to the director, respectively. Then the free energy is obtained by an integration over the volume

$$F = \int dV f \tag{3}$$

where $f = f_n + f_e$ is explicitly written down in the Appendix. The director orientation for an equilibrium state is deduced from the condition that the free energy (3) is a minimum. For a stability analysis, however, the application of the torque balance equation

$$n \times \left(\lambda \frac{\partial n}{\partial t} + \frac{\delta F}{\delta n} \right) = 0 \tag{4}$$

is often more useful. The coefficient λ denotes the rotational viscosity and t is the time. Equation (4) allows to describe director reorientations but neglects material flows. It is convenient to introduce dimensionless space coordinates by $x = \pi X/d$ and $y = \pi Y/d$ proposing that the film is confined between the planes $x = -\pi/2$ and $x = \pi/2$. The director attached to the bounding plate and the axis y , which is parallel to the domain wall normal (Fig. 1), enclose the angle $\kappa = \phi(x = \pm\pi/2)$. Expressing the director components in terms of θ and ϕ we obtain

$$n_x = \sin \theta, \quad n_y = \cos \theta \cos \phi \quad \text{and} \quad n_z = \cos \theta \sin \phi \tag{5}$$

Then equation (4) is replaced by

$$-\frac{\partial \theta}{\partial \tau} = \frac{\partial f}{\partial \theta} - \frac{d}{dx} \frac{\partial f}{\partial \theta_x} - \frac{d}{dy} \frac{\partial f}{\partial \theta_y} \tag{6}$$

$$-\cos^2 \theta \frac{\partial \phi}{\partial \tau} = \frac{\partial f}{\partial \phi} - \frac{d}{dx} \frac{\partial f}{\partial \phi_x} - \frac{d}{dy} \frac{\partial f}{\partial \phi_y} \tag{7}$$

where the indices x , y and τ symbolize the partial derivatives ($\theta_x = \frac{\partial \theta}{\partial x}$, $\theta_y = \frac{\partial \theta}{\partial y}$, $\theta_\tau = \frac{\partial \theta}{\partial \tau}$, etc.) and $\tau = \pi^2 K_{11} t / d^2 \lambda$ is the time in dimensionless units. The electric field is not homogeneous if distortions occur. Introducing a potential V for the electric field by

$$E_x = \frac{U}{d} \frac{\partial V}{\partial x} \quad \text{and} \quad E_y = \frac{U}{d} \frac{\partial V}{\partial y} \tag{8}$$

(U , effective value of the applied voltage) we arrive at the equation

$$\frac{d}{dx} \frac{\partial f}{\partial V_x} + \frac{d}{dy} \frac{\partial f}{\partial V_y} = 0 \tag{9}$$

assuming that the electric potential responds immediately when the director is slowly reorienting. At the substrates the director and the electric potential V satisfy the boundary conditions

$$\begin{aligned} \theta(x = -\pi/2) = 0, \quad \theta(x = \pi/2) = 0, \quad \phi(x = -\pi/2) = \kappa, \quad \phi(x = \pi/2) = \kappa, \tag{10} \\ V(x = -\pi/2) = -\frac{\pi}{2} \quad \text{and} \quad V(x = \pi/2) = \frac{\pi}{2} \end{aligned}$$

3. Solution for Simple Domain Walls

Since the distortion amplitude is small in the vicinity of the Freedericksz-threshold, a perturbation method is suitable to solve the set of nonlinear equations (6, 7, 9). Let us start with the perturbation expansions

$$\begin{aligned}
 \theta &= \varepsilon\theta^{(1)} + \varepsilon^2\theta^{(2)} + \varepsilon^3\theta^{(3)} + \dots \\
 \phi &= \phi^{(0)} + \varepsilon\phi^{(1)} + \varepsilon^2\phi^{(2)} + \varepsilon^3\phi^{(3)} + \dots \\
 V &= V^{(0)} + \varepsilon V^{(1)} + \varepsilon^2 V^{(2)} + \varepsilon^3 V^{(3)} + \dots \\
 \mu &= \mu^{(0)} + \varepsilon\mu^{(1)} + \varepsilon^2\mu^{(2)} + \dots
 \end{aligned} \tag{11}$$

where ε is a small parameter indicating the order of magnitude and the bifurcation parameter μ is defined in the Appendix. Furthermore, the time variable τ and the coordinate y can be expanded, because there is generally a critical slowing down for director motions and the thickness of the domain walls diverges if the Freedericksz-threshold is approached. In the spirit of the multiple scale method, which is well established in the theory of non-linear oscillators [7], we introduce slowly varying time variables $\tau_1 = \varepsilon\tau$, $\tau_2 = \varepsilon^2\tau$, . . . and space variables $y_1 = \varepsilon y$, $y_2 = \varepsilon^2 y$, . . . and the derivatives are replaced by

$$\partial/\partial y = \varepsilon\partial/\partial y_1 + \varepsilon^2\partial/\partial y_2 + \dots \quad \text{and} \quad \partial/\partial\tau = \varepsilon\partial/\partial\tau_1 + \varepsilon^2\partial/\partial\tau_2 + \dots \tag{12}$$

The series (11) and (12) are inserted in the equations (6, 7, 9) and the resulting expressions are expanded with respect to the small parameter ε . Collecting all terms with the same order of magnitude ε^n leads to a hierarchy of linear differential equations which can be solved stepwise. To lowest order of magnitude we obtain

$$\varepsilon^0 \cdot \quad \phi_{xx}^{(0)} = 0 \quad \text{and} \quad V_{xx}^{(0)} = 0$$

The solutions of these equations satisfying the boundary conditions (10) are

$$\phi^{(0)} = \kappa \quad \text{and} \quad V^{(0)} = x \tag{13}$$

Obviously, higher order terms satisfy the boundary conditions

$$\phi^{(n)}(x = \pm\pi/2) = 0, \quad V^{(n)}(x = \pm\pi/2) = 0 \quad \text{and} \quad \theta^{(n)}(x = \pm\pi/2) = 0 \tag{14}$$

for $n = 1, 2, 3, \dots$. Collecting all terms which are proportional to ε^1 leads to

$$\varepsilon^1 \cdot \quad \theta_{xx}^{(1)} + \mu^{(0)}\theta^{(1)} = 0, \quad \phi_{xx}^{(1)} = 0 \quad \text{and} \quad V_{xx}^{(1)} = 0$$

In further calculations the notation

$$\gamma = \frac{\Delta\varepsilon}{\varepsilon_{\perp}}, \quad k_2 = \frac{K_{22}}{K_{11}}, \quad k_3 = \frac{K_{33}}{K_{11}} \quad \text{and} \quad U_F^2 = \pi^2 K_{11}/\Delta\varepsilon$$

is used. The equations for $\phi^{(1)}$ and $V^{(1)}$ with the boundary conditions (14) have only the trivial solution

$$\phi^{(1)} = 0 \quad \text{and} \quad V^{(1)} = 0 \tag{15}$$

The equation for $\theta^{(1)}$, however, possesses nontrivial solutions with the eigenvalues $\mu^{(0)} = m^2$, where m is a natural number. Only the smallest eigenvalue $\mu^{(0)} = 1$ is of physical significance

and the corresponding voltage $U = U_F$ turns out to be the Freedericksz threshold. Thus $\theta^{(1)}$ is expressed as

$$\theta^{(1)} = b^{(1)}(\tau_1, \tau_2, \dots; y_1, y_2, \dots) \cos x$$

where the deformation amplitude $b^{(1)}$ depends on the slowly varying time variables τ_i and space variables y_i . Collecting all terms proportional to ϵ^2 in the equations (6, 7, 9) we obtain

$$\begin{aligned} \epsilon^2 \cdot \quad \theta_{xx}^{(2)} + \theta^{(2)} &= -\mu^{(1)}\theta^{(1)} + \theta_{\tau_1}^{(1)} \\ \phi_{xx}^{(2)} &= -\frac{(1 - k_2)}{k_2} \sin \kappa \sin x b_{y_1}^{(1)} \end{aligned} \tag{16}$$

and

$$V_{xx}^{(2)} = -b_{y_1}^{(1)}\gamma \cos \kappa \cos x + 2(b^{(1)})^2\gamma \sin x \cos x.$$

The equations for ϕ and V are solved by a straightforward integration. Considering the boundary conditions (14) the result is

$$\phi^{(2)} = F(x)b_{y_1}^{(1)} \quad \text{with} \quad F(x) = \frac{(1 - k_2)}{k_2} \sin \kappa \left(\sin x - \frac{2x}{\pi} \right)$$

and

$$V^{(2)} = b_{y_1}^{(1)}\gamma \cos \kappa \cos x - \frac{1}{2}(b^{(1)})^2\gamma \sin x \cos x$$

For functions θ and $\hat{\theta}$, which belong to a suitably defined function space and satisfy the boundary conditions (14), the linear operator L defined by $L\theta \equiv \theta_{xx} + \theta$ is self-adjoint when the scalar product $\langle \theta, \hat{\theta} \rangle = \int_{-\pi/2}^{+\pi/2} dx \theta(x)\hat{\theta}(x)$ is used. Because of $\langle L\theta, \hat{\theta} \rangle = \langle \theta, L\hat{\theta} \rangle$ and since the homogeneous equation $L\hat{\theta} = 0$ has the nontrivial solution $\hat{\theta} = \cos x$, the corresponding non-homogeneous equation $L\theta = h$ is solvable only in those cases where the condition $\langle h, \cos x \rangle = 0$ is satisfied (Fredholm's alternative). Applying this solvability condition to equation (16) yields

$$\int_{-\pi/2}^{+\pi/2} (-\mu^{(1)}\theta^{(1)} + \theta_{\tau_1}^{(1)}) \cos x dx = 0$$

and performing the integration we obtain $-\mu^{(1)}b^{(1)} + b_{\tau_1}^{(1)} = 0$. For $\mu^{(1)} \neq 0$ the amplitude $b^{(1)}$ grows without limits or tends to zero. Accordingly, for describing static domain walls, only the possibility $\mu^{(1)} = 0$ and $b_{\tau_1}^{(1)} = 0$ has to be considered. Thus we conclude that $b^{(1)}$ does not depend on the variable τ_1 .

Finally, let us collect all terms of equation (6) which are proportional to ϵ^3 . Taking into account the results for $\phi^{(2)}$ and $V^{(2)}$ we get

$$\epsilon^3 \cdot \quad \theta_{xx}^{(3)} + \theta^{(3)} = G^{(3)}$$

where

$$\begin{aligned} G^{(3)} &= (b_{\tau_2}^{(1)} + b_{\tau_1}^{(2)}) \cos x - \mu^{(2)}b^{(1)} \cos x + 3 \cos \kappa (k_3 + \gamma - 1) \sin x \cos x b^{(1)} b_{y_1}^{(1)} \\ &\quad - \left[(k_2 \sin^2 \kappa + (k_3 + \gamma) \cos^2 \kappa) \cos x - (1 - k_2) \sin \kappa \frac{dF(x)}{dx} \right] b_{y_1 y_1}^{(1)} \\ &\quad - \left[\gamma \cos x - \left(\frac{2}{3} + 2\gamma \right) \cos^3 x + (k_3 - 1)(\cos x \sin^2 x - \cos^3 x) \right] (b^{(1)})^3 \end{aligned}$$

The integrability condition

$$\varepsilon^3 \int_{-\pi/2}^{+\pi/2} G^{(3)} \cos x dx = 0$$

leads to an equation which can be written as

$$\varepsilon^3 (b_{\tau_2}^{(1)} + b_{\tau_1}^{(2)}) - \varepsilon^2 \mu^{(2)} \varepsilon b^{(1)} - \delta \varepsilon^3 b_{y_1 y_1}^{(1)} + B(\varepsilon b^{(1)})^3 = 0 \quad (17)$$

where

$$\delta = \left[k_2 - \left(1 - \frac{8}{\pi^2} \right) \frac{(1 - k_2)^2}{k_2} \right] \sin^2 \kappa + [k_3 + \gamma] \cos^2 \kappa \quad \text{and} \quad B = \frac{1}{2}(k_3 + \gamma).$$

Now we define an amplitude b and insert the definitions for the slowly varying time variables $\tau_n = \varepsilon^n \tau$ and space variables $y_n = \varepsilon^n y$ taking into account that $b^{(1)}$ does not depend on τ_1 :

$$b(\tau, y) = \varepsilon [b^{(1)}(\varepsilon^2 \tau, \dots; \varepsilon y, \dots) + \varepsilon b^{(2)}(\varepsilon \tau, \varepsilon^2 \tau, \dots; \varepsilon y, \dots)] \quad (18)$$

Omitting higher order terms (proportional to ε^n with $n \geq 4$) equation (17) is transformed into ($\varepsilon^2 \mu^{(2)} \Rightarrow \hat{\mu}$, $\varepsilon \partial_{y_1} \Rightarrow \partial_y$)

$$b_\tau - \hat{\mu} b - \delta b_{yy} + B b^3 = 0 \quad (19)$$

with $\hat{\mu} = \frac{U^2 - U_F^2}{U_F^2}$ The director angles and the electric potential are expressed as

$$\begin{aligned} \theta(\tau, x, y) &= b(\tau, y) \cos x + O(\varepsilon^3) \\ \phi(\tau, x, y) &= \kappa + F(x) b_y + O(\varepsilon^4) \end{aligned} \quad (20)$$

and

$$V(\tau, x, y) = x - \gamma \cos x \cos \kappa b_y + 2\gamma \sin x \cos x b^2 + O(\varepsilon^4).$$

where $b(\tau, y)$ is the solution of the amplitude equation (19).

4. Discussion

For $\hat{\mu} > 0$ equation (19) has some nontrivial static solutions for the distortion amplitude being valid just above the Freedericksz threshold U_F . There are two homogeneous states $b = \pm \sqrt{\hat{\mu}/B}$ and the third solution

$$b(y) = \sqrt{\hat{\mu}/B} \tanh(y/\xi) \quad (21)$$

with $\xi = \sqrt{2\delta/\hat{\mu}}$ describes both states separated by a domain wall. The thickness ξ of the wall depends on the angle κ between the wall normal and the orientation of the director at the film substrates. In comparison to previously obtained results the relations (20) reveal that the azimuthal director angle ϕ is influenced by the presence of a wall. This angle is only constant for $k_2 = 1$ and for the case of a splay-bend wall ($\kappa = 0$). Usually, the assumption $k_2 = 0.5$ provides a good estimation for the elastic anisotropy of many substances consisting of simple rod-like molecules. Polymeric material, however, could have a considerably larger elastic anisotropy. Then remarkable differences to the predictions of the simplified model [3] with $\phi(x, y) = \text{const.}$ are expected to occur. After switching the electric field on, domains with clockwise and anticlockwise director rotation appear at least in well oriented samples. Elliptic loops surrounding domains with unique director alignment possess a fixed ellipticity which depends on the elastic anisotropy. Following the calculation presented in [3], the wall

ellipticity a/c is expressed as the ratio ξ_1/ξ_2 of the wall thicknesses $\xi_1 = \sqrt{2\delta(\kappa = \pi/2)/\hat{\mu}}$ (twist-bend wall) and $\xi_2 = \sqrt{2\delta(\kappa = 0)/\hat{\mu}}$ (splay-bend wall). Thus we obtain the formula

$$\frac{a}{c} = \sqrt{\frac{k_2 - \left(1 - \frac{8}{\pi^2}\right) \frac{(1 - k_2)^2}{k_2}}{k_3 + \gamma}} \quad (22)$$

which replaces the result $a/c = \sqrt{k_2/k_3}$ [2, 3] obtained for a magnetic field ($\gamma \ll 1$) and with the simplification $\phi(x, y) = \text{const.}$ Equation (22) offers a proper method to determine experimentally the ratio k_2/k_3 of the elastic constants.

Consistency of the theory for simple domain walls requires that $\delta(\kappa) > 0$ for all possible angles κ ensuring that the expression $\xi = \sqrt{2\delta(\kappa)/\hat{\mu}}$ for the domain wall thickness yields a real value. Obviously, $\delta(\kappa = \pi/2)$ is negative if $k_2 < 0.303$. Then the equation (19) for simple domain walls is not longer applicable and must be replaced by the extended equation

$$b_\tau - \hat{\mu}b - \delta(\kappa = \pi/2)b_{yy} + Ab_{yyy} + Bb^3 = 0 \quad (23)$$

which can be derived by a slight alteration of the ansatz (11). The additional coefficient $A > 0$ is found to be [8]

$$A = \frac{2(1 - k_2)^2}{\pi^2} \left(3 - \frac{16 - \pi^2}{\pi^2 - 8} + \frac{5\pi^2 - 48}{6k_2^2} \right), \quad (24)$$

while the other coefficients $\delta(\kappa = \pi/2) = \delta_m$, $\hat{\mu}$ and B in equation (23) are defined in the same way as for equation (19). If $\delta_m < 0$ ($k_2 < 0.303$) equation (23) describes a periodic array of walls for a certain region of the bifurcation parameter $\hat{\mu}$. In the case $\delta_m > 0$ ($k_2 > 0.303$) the solutions of equations (21) and (23), which correspond to stable states, are $b^I = 0$ for $\hat{\mu} < 0$ and $b^{II} = \pm\sqrt{\hat{\mu}/B}$ for $\hat{\mu} > 0$. The transition between the states I and II is the ordinary Fredericksz transition.

In the other case, if $\delta_m < 0$ ($k_2 < 0.303$), a periodic distorted state (III) described by $b^{III}(y) = \sqrt{\frac{4\hat{\mu}A + \delta_m^2}{3AB}} \sin qy$ and the wavenumber $q = \sqrt{-\delta_m/(2A)}$ is stable in a region $\hat{\mu}_1 < \hat{\mu} < \hat{\mu}_2$, where the lower limit is defined by $\hat{\mu}_1 = -\delta_m^2/(4A)$. It should be noted, however, that equation (23) and its solution (III) are valid only in the vicinity of the Lifshitz-point defined by $\hat{\mu} = 0$ and $\delta_m = 0$, because in this region the period of the distortions is large compared to the film thickness. The determination of the upper limit $\hat{\mu}_2$ requires a more sophisticated calculation, as the transition between state (III) and state (II) is discontinuous [9]. Equation (23) is related to the Ljapunov-functional

$$L[b(y)] = \frac{1}{2} \int dy (Ab_{yy}^2 + \delta_m b_y^2 - \hat{\mu}b^2 + \frac{1}{2}Bb^4) \quad (25)$$

which always decreases ($dL/d\tau \leq 0$) until any dynamics ceases. For such a system Maxwell's rule [10]

$$L[b^{II}] = L[b^{III}(y)] \quad (26)$$

holds when an equilibrium between the stable film states b^{II} and $b^{III}(y)$ occurs. Otherwise, if $L[b^{II}] \neq L[b^{III}(y)]$, the absolutely stable state could grow at the cost of the metastable one by domain wall motion.

Inserting the expressions for b^{II} and $b^{III}(y)$ in the functional (25) and solving equation (26) yields

$$\hat{\mu}_2 = \frac{\delta_m^2}{4A} (2 + \sqrt{6}) \quad (27)$$

Outside the region $\hat{\mu}_1 < \hat{\mu} < \hat{\mu}_2$ the non-distorted state $b^I = 0$ is stable for $\hat{\mu} < \hat{\mu}_1$, whereas the Freedericksz-distorted state b^{II} minimizes the Ljapunov-functional for $\hat{\mu} > \hat{\mu}_2$.

In conclusion, we have extended the mathematical model for simple domain walls by taking into account additionally azimuthal director rotations. The correction to the equation for the distortion amplitude becomes more pronounced with increasing elastic anisotropy. In accordance with the theory of Lonberg and Meyer [6] the model predicts the occurrence of a periodic array of walls if the elastic anisotropy is sufficiently high.

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Appendix

Free energy density for a nematic film subjected to an electric field

$$\begin{aligned}
 f = & \frac{\pi^2 K_{11}}{2d^2} \{ \cos^2 \theta \theta_x^2 + \sin^2 \theta \cos^2 \phi \theta_y^2 + \cos^2 \theta \sin^2 \phi \phi_y^2 - 2 \sin \theta \cos \theta \cos \phi \theta_x \theta_y \\
 & - 2 \cos^2 \theta \sin \phi \theta_x \phi_y + 2 \sin \theta \cos \theta \sin \phi \cos \phi \theta_y \phi_y \\
 & + k_2 [\cos^4 \theta \phi_x^2 + \sin^2 \phi \theta_y^2 + \sin^2 \theta \cos^2 \theta \cos^2 \phi \phi_y^2 + 2 \cos^2 \theta \sin \phi \theta_y \phi_x \\
 & - 2 \sin \theta \cos^3 \theta \cos \phi \phi_x \phi_y - 2 \sin \theta \cos \theta \sin \phi \cos \phi \theta_y \phi_y] \\
 & + k_3 [\sin^2 \theta \theta_x^2 + \cos^2 \theta \cos^2 \phi \theta_y^2 + \sin^2 \theta \cos^2 \theta \phi_x^2 + \cos^4 \theta \cos^2 \phi \phi_y^2 \\
 & + 2 \sin \theta \cos \theta \cos \phi \theta_x \theta_y + 2 \sin \theta \cos^3 \theta \cos \phi \phi_x \phi_y] \\
 & - \frac{\mu}{\gamma} \{ (1 + \gamma \sin^2 \theta) V_x^2 + 2 \gamma \sin \theta \cos \theta \cos \phi V_x V_y + (1 + \gamma \cos^2 \theta \cos^2 \phi) V_y^2 \}
 \end{aligned}$$

where the notation $\mu = U^2/U_F^2$, $U_F^2 = \pi^2 K_{11}/\Delta\epsilon$, $\gamma = \Delta\epsilon/\epsilon_\perp$, $k_2 = K_{22}/K_{11}$ and $k_3 = K_{33}/K_{11}$ is used (V , potential of the electric field; U , applied voltage; d , film thickness).

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