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INFLUENCE OF THE SADDLE-SPLAY CONSTANT ON THE DIRECTOR FIELD DISTRIBUTION IN STRONG MAGNETIC FIELDS

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The role of anchoring effects in thin nematic films confined between two parallel plates was theoretically examined. The bulk and surface free energy densities were expanded up to $\mathcal{O}(\varepsilon^2)$ and the perturbated contributions were calculated. It is shown that the minimum of the free energy corresponds to the solution of the Euler-Lagrange equations and satisfies the Ericksen inequalities. The identified bifurcation points can estimate the influence of the saddle-splay constant k_{24} towards periodic perturbations of a director in the presence of strong magnetic field.

Key words: nematic liquid crystals, thin films, saddle-splay elasticity, Fréedericksz transition, linear perturbation theory.

Introduction

Nematic liquid crystals (LCs) confined in restricted geometries are technologically important and have been the subject of extensive experimental and theoretical research for five decades. Fréedericksz transitions caused by an external electric or magnetic field in nematic LCs have attracted attention since their discovery in 1933.

When the magnetic field is applied perpendicular to the uniformly aligned nematic, confined between two parallel plates, above a certain threshold, the director is affected on by deformations and tends to align along the field. The critical threshold of the magnetic field in one constant approximation is inversly proportional to the thickness of the nematic sample $2d$, and is given by

$$H_c = \frac{\pi}{2d}\sqrt{\frac{k}{\chi_a}},$$

where χ_a is the anisotropy of the diamagnetic susceptibility, which is assumed a positive number, and k is the elastic constant [1].

A surface term k_{24}, which is often omitted represents elastic contribution to the surface free energy that, originally, has been indicated as a part of the elastic energy having the form of a divergence. This contribution – the so-called the saddle-splay

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term k_{24}, which is important only for particular situations, in which a distortion has a two- or three-dimensional structures [2, 3].

One of the recent theoretical results [4] in the one-constant approximation shows that k_{24} causes instabilities if the ratio $k_{24}/k \geqslant 0.707$. Another result from this study is the suggestion of how to measure k_{24}.

The results in ref. [4] are essentially limited to the particular case in which the layer of nematic LC is infinite, and no proof that the distorted contributions of the distribution of polar and azimuthal angles yield the minimum of free energy. Hence, the study of possible mechanisms of interactions of the saddle-splay energy and strong magnetic fields ($H \gg H_c$) requires a quantitatively accurate description of the instability that goes beyond these limitations.

In this work we report the theory of the interaction of the saddle-splay elastic constant under the onset of periodic perturbations of a director in the presence of strong magnetic field. Our theoretical model is based on the Oseen-Zocher-Frank continuum theory of LCs. In the next Section we define the geometry of the problem and introduce free energy. In Section 2. we obtain the bulk and surface free energies in two-constant approximation. Then we determine bifurcation points by minimizing free energy. In Section 3. we analyze the influence of the saddle-splay term on the stability of the unperturbated state. In Section 3. we briefly outline the results of our research and the perspectives on open problems.

1. Geometry of the problem and free energy

Nematic LCs are described by director $\mathbf{n}$, which is a unit vector, characterising the average orientation of molecules. The standart expression for the elastic free energy associated with $\mathbf{n}$ is given by

$$F_{el} = \frac{1}{2} \int_V dV \{k_{11}(\operatorname{div}\mathbf{n})^2 + k_{22}(\mathbf{n}\cdot\operatorname{curl}\mathbf{n})^2 + k_{33}[\mathbf{n}\times\operatorname{curl}\mathbf{n}]^2 \\ -(k_{22}+k_{24})\operatorname{div}[\mathbf{n}\times\operatorname{curl}\mathbf{n} + (\operatorname{div}\mathbf{n})\mathbf{n}]\}\,, \tag{1}$$

where k_{11}, k_{22}, k_{33} and k_{24} are the splay, twist, bend and saddle-splay moduli respectively. In view of Gauss' theorem, the divergence terms only contribute to the surface free energy density. To guarantee a stable configuration of nematic LC in the absence of external fields, the saddle-splay constant must fulfill the Ericksen inequalities [2]:

$$k_{ii} \geqslant 0\,, \quad |k_{24}| \leqslant k_{22}\,, \quad k_{22}+k_{24} \leqslant 2k_{11}\,, \quad |k_{11}-k_{22}-k_{24}| \leqslant k_{11}. \tag{2}$$

We assume that $k_{11} = k_{33} = k$ and $k_{24} \neq 0$, then equation (1) takes the form

$$F_{el} = \frac{1}{2} \int_V dV \{k(\operatorname{div}\mathbf{n})^2 + k_{22}(\mathbf{n}\cdot\operatorname{curl}\mathbf{n})^2 + k[\mathbf{n}\times\operatorname{curl}\mathbf{n}]^2 \\ -(k_{22}+k_{24})\operatorname{div}[\mathbf{n}\times\operatorname{curl}\mathbf{n} + (\operatorname{div}\mathbf{n})\mathbf{n}]\}\,. \tag{3}$$

Consider one of the basic configurations of the director and the magnetic field for the Fréedericksz transition (e.g. [3], p. 307). Let the distance in the bulk (see Fig. 1), which is needed to orient the director along the applied magnetic field $\mathbf{H} = H\mathbf{e}_x$ be expressed in the CGS system through the dimensionless coherence length

$$\varepsilon_\theta = \frac{1}{d}\sqrt{\frac{k}{\chi_a H^2}}\,,$$

where d is the half-thickness of the layer and $\varepsilon_\theta \ll 1$.

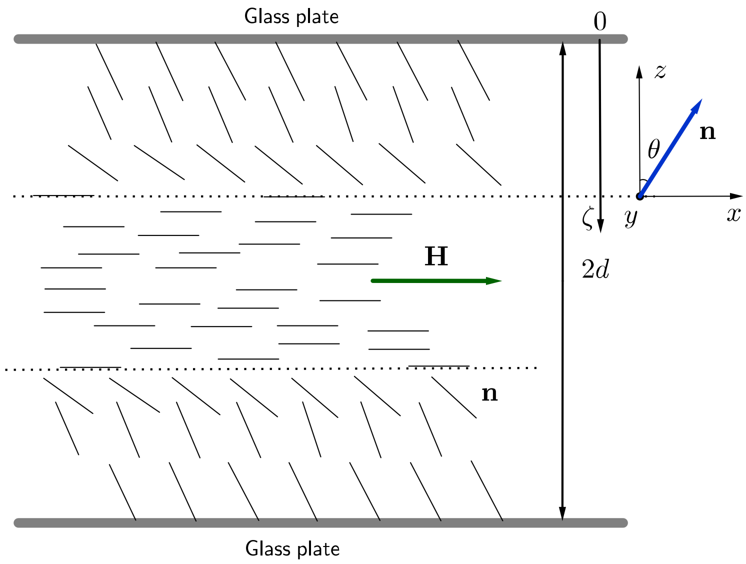

Fig. 1. Representation of the director **n** under the influence of the magnetic field H in a cell of the thickness $2d$

The magnetic field contribution to the elastic free energy is represented by the functional

$$F = F_{el} - \frac{\chi_a}{2} \int_V (\mathbf{n}, \mathbf{H})^2 \, dV \,,$$

which yields the Euler equation [1, 2]

$$\frac{d^2\theta}{d\zeta^2} + \sin\theta\cos\theta = 0\,, \tag{4}$$

where $\zeta : 2/\varepsilon_\theta \geqslant \zeta \geqslant 0$ is the scaled coordinate, which is related with the space coordinate $-d < z < d$ as $z = d - \varepsilon_\theta d\zeta$, θ is the angle between the director and the z-axis (see Fig. 1).

When $\zeta \to 1/\varepsilon_\theta$, we suppose $\theta = \pi/2$, $(d\theta/d\zeta)_{\zeta\to 1/\varepsilon_\theta} = 0$ and $\rho \cdot (d\theta/d\zeta)_{\zeta=0} = \sin\theta\cos\theta|_{\zeta=0}$, where $\rho \equiv \sqrt{kH^2\chi_a}/W_a$ is the dimensioless parameter characterising the relative strength of the magnetic field to the surface anchoring energy W_a. The solution of equation (4) yields the function

$$\theta(\zeta) = \arcsin\left(\frac{Ae^{2\zeta} - 1}{Ae^{2\zeta} + 1}\right)\,, \tag{5}$$

where $A(\rho) = \dfrac{1+\rho}{1-\rho}$.

It is widely known that the saddle-splay constant contributes to the equations for problems involving weak anchoring of the LC at the surface. Due to this reason, a mathematical condition $0 < \rho < 1$ must hold.

2. Stability analysis

Supposing that the state (5) is linearly unstable, consider small perturbations of the director $\mathbf{n}$ induced by the magnetic field under the assumption that the configuration of the perturbations of the director in the yz plane is defined by

$$\mathbf{n} = \sin(\theta+\psi)\cos\varphi\,\mathbf{e}_x + \sin(\theta+\psi)\sin\varphi\,\mathbf{e}_y + \cos(\theta+\psi)\,\mathbf{e}_z\,, \tag{6}$$

where $\varphi(y,z)$, $\psi(y,z)$ are small $\mathcal{O}(\varepsilon)$ and periodic functions, i.e.

$$\varphi(y,z) = g(z)\cos(qy), \quad \psi(y,z) = f(z)\sin(qy)\,, \tag{7}$$

where q is the wavenumber.

The Taylor series expansion of the elastic and magnetic free energy up to $\mathcal{O}(\varepsilon^2)$ gives

$$\begin{gathered} f_b^{(2)} = \tfrac{1}{2}\sin^2\theta\left(k\left(\tfrac{\partial\varphi}{\partial y}\right)^2 + k_{22}\left(\tfrac{\partial\varphi}{\partial z}\right)^2\right) + \\ \tfrac{1}{2}\left(k\sin^2\theta\left(\tfrac{\partial\psi}{\partial z}\right)^2 + k_{22}\cos^2\theta\left(\tfrac{\partial\psi}{\partial z}\right)^2 + k_{22}\left(\tfrac{\partial\psi}{\partial y}\right)^2\right) + \\ (k_{22}-k)\left[\sin^2\theta\tfrac{\partial\varphi}{\partial y}\tfrac{\partial\psi}{\partial z} + \tfrac{\psi^2}{2}\cos 2\theta\left(\tfrac{\partial\theta}{\partial z}\right)^2\right] + \\ \tfrac{1}{2}(k_{22}-k)\sin 2\theta\tfrac{\partial\theta}{\partial z}\left(\varphi\tfrac{\partial\psi}{\partial y} - 2\psi\tfrac{\partial\psi}{\partial z} - 2\psi\tfrac{\partial\varphi}{\partial y}\right) - \\ \tfrac{\chi_a H^2}{2}\left(\psi^2\cos 2\theta - \varphi^2\sin^2\theta\right)\,, \end{gathered} \tag{8}$$

and the surface energy obtained from the Rapini-Papoular functional [5] yields

$$f_s^{(2)} = \psi^2\frac{W_a}{2}\cos 2\theta - (k_{22}+k_{24})\,\varphi\sin^2\theta\frac{\partial\psi}{\partial y} + k_{24}\psi\frac{\partial\psi}{\partial y}\,. \tag{9}$$

Under the assumption that the field-induced distortions (7) depend only on ζ, the configuration of the director can be found by solving the Euler-Lagrange equations. The variational problem for f and g (7) written in terms of the scaled variable ζ leads to the following system of differential equations:

$$\begin{gathered} \gamma\left(\tfrac{d^2 g}{d\zeta^2} + \tfrac{8A\,e^{2\zeta}}{A^2 e^{4\zeta}-1}\tfrac{dg}{d\zeta}\right) - (\gamma-1)\sqrt{\omega_1^2-1}\left(\tfrac{df}{d\zeta} - \tfrac{8A\,e^{2\zeta}f}{A^2e^{4\zeta}-1}\right) \\ -g\omega_1^2 = 0\,, \end{gathered} \tag{10}$$

$$\begin{gathered} \left[\tfrac{1}{\gamma}\left(\tfrac{Ae^{2\zeta}-1}{Ae^{2\zeta}+1}\right)^2 + \tfrac{4Ae^{2\zeta}}{(Ae^{2\zeta}+1)^2}\right]\tfrac{d^2f}{d\zeta^2} - f\left(\omega_2^2 - \tfrac{8Ae^{2\zeta}}{(Ae^{2\zeta}+1)^2}\right) \\ +\tfrac{1}{(Ae^{2\zeta}+1)^2}\left[20Ae^{2\zeta}q\tfrac{Ae^{2\zeta}-1}{Ae^{2\zeta}+1}g + q(Ae^{2\zeta}-1)^2\tfrac{dg}{d\zeta}\right. \\ -12Ae^{2\zeta}\left(\tfrac{8A}{(Ae^{2\zeta}+1)^2}-1\right)f + 8Ae^{2\zeta}\left(\tfrac{Ae^{2\zeta}-1}{Ae^{2\zeta}+1}\right)^2 f \\ \left.+16Ae^{2\zeta}\tfrac{Ae^{2\zeta}-1}{Ae^{2\zeta}+1}\tfrac{df}{d\zeta}\right]\left(1-\tfrac{1}{\gamma}\right) = 0\,, \end{gathered} \tag{11}$$

where $\gamma = \dfrac{k_{22}}{k}$, $\omega_1^2 = 1 + q^2\varepsilon_\theta^2 d^2$, $\omega_2^2 = \dfrac{1}{\gamma} + q^2\varepsilon_\theta^2 d^2$. Equations (10,11) written in terms of the one constant approximation will look identical with the corresponding variational problem given in ref. [4].

The obtained equations do not have an analytical solution, and we will treat (10,11) for the isotropic approximation $k_{11} = k_{22} = k_{33}$, assuming lower and upper boundaries. Then $\omega_1 = \omega_2\big|_{\gamma=1}$, and consider a common value ω.

The boundary conditions for weak anchoring are due to the minimization of the bulk and surface free energy terms. Hence, the influence of the surface on the equilibrium director field of the LC is represented by the boundary conditions

$$-\frac{\partial f_b^{(2)}}{\partial \varphi_{,\zeta}} + \frac{\partial f_s^{(2)}}{\partial \varphi}\bigg|_{\zeta=0} = 0\,, \quad -\frac{\partial f_b^{(2)}}{\partial \psi_{,\zeta}} + \frac{\partial f_s^{(2)}}{\partial \psi}\bigg|_{\zeta=0} = 0 \tag{12}$$

or

$$\begin{aligned} g_\zeta(0) &= (1+\tau)\sqrt{\omega^2-1}f(0)\,; \\ f_\zeta(0) &= -\left(\tfrac{1}{\rho} - 2\rho\right) f(0) + \tau\sqrt{\omega^2-1}\,g(0)\,, \end{aligned} \tag{13}$$

where $\tau = k_{24}/k$ is the dimensionless saddle-splay modulus. To give a quantitative estimation, we carried out further calculations for $2d = 40 \cdot 10^{-4}$ cm, $\chi_a = 10^{-7}$, $H = 1.5 \cdot 10^4$ G, $k = 10^{-6}$ dyn and $W_a = 9 \cdot 10^{-3}$ erg/cm^2, then $\varepsilon_\theta \simeq 0.1$ and $\zeta \simeq 10$. Likewise, the second pair of the boundary conditions is given by

$$\frac{\partial f_b^{(2)}}{\partial \varphi_{,\zeta}} + \frac{\partial f_s^{(2)}}{\partial \varphi}\bigg|_{\zeta=10} = 0\,, \quad \frac{\partial f_b^{(2)}}{\partial \psi_{,\zeta}} + \frac{\partial f_s^{(2)}}{\partial \psi}\bigg|_{\zeta=10} = 0 \tag{14}$$

or

$$\begin{aligned} g_\zeta(10) &= -(1+\tau)\sqrt{\omega^2-1}f(10)\,; \\ f_\zeta(10) &= \tfrac{1}{\rho} f(10) + \tau\sqrt{\omega^2-1}\,g(10)\,. \end{aligned} \tag{15}$$

With the aid of a computer algebra system, we obtain the distribution of perturbations for the polar and azimuthal angles

$$\begin{aligned} f(\zeta) &= C_1 \tfrac{e^{\omega\zeta}(\omega+1+Ae^{2\zeta}(\omega-1))}{Ae^{2\zeta}+1} + C_2 \tfrac{e^{-\omega\zeta}(\omega-1+Ae^{2\zeta}(\omega+1))}{Ae^{2\zeta}+1}\,, \\ g(\zeta) &= C_3 \tfrac{e^{\omega\zeta}(\omega+1+Ae^{2\zeta}(\omega-1))}{Ae^{2\zeta}-1} + C_4 \tfrac{e^{-\omega\zeta}(\omega-1+Ae^{2\zeta}(\omega+1))}{Ae^{2\zeta}-1}\,, \end{aligned} \tag{16}$$

where the integration constants C_i must satisfy the boundary conditions (13) and (15). The resulting equations represent the system of linear homogeneous equations $\sum\limits_{i,j=1}^{4} \mathcal{M}_{i,j} C_j = 0$, which has a non-trivial solution if

$$\det\ \mathcal{M} = 0\,. \tag{17}$$

By computing the determinant of $\mathcal{M}$, we get an implicit equation with respect to ω, τ and ρ. It is clear from (2) that (17) is regarded to the case $\tau \leqslant 1$.

In order to test the model, it is necessary to clarify if solutions (16) of the Euler-Lagrange equations yield minimum energy with respect to the absence of perturbations (6). Therefore, if the sum of the bulk and surface free energy terms with the supposed perturbations (6) is less than the sum without perturbations, i.e. $\varphi(y,z) = \psi(y,z) = 0$ (stable state), then the supposed perturbations exist. Thus, the condition

$$\int\limits_0^\lambda \int\limits_0^d f_b^{(2)} dz dy + \int\limits_0^\lambda f_s^{(2)} dy\bigg|_{z=d} < 0 \tag{18}$$

must hold. If we suppose that $C_i/C_j \simeq 1$, where $i,j = 1 \ldots 4$, then the substitution of the solutions of the Euler-Lagrange equations (16) to (18) yields the expression $F_b^{(2)} + F_s^{(2)} = G(\tau, d, \lambda, W_a, H, \chi_a) < 0$, where λ is the wavelength of the periodic distortions of the director along the y axis. Using the parameters given above, we can plot the condition of the existence of perturbations (see Fig. 2).

For a boundary case when $H \gtrapprox H_c$, periodic distortions of the director are possible if $|\tau| \gg 1$, which violates the Ericksen inequalities. It is also easy to challenge the model on the stability of with respect to $C_i/C_j \simeq 0.1 \ldots 10$, and the corresponding results do fulfill conditions (2).

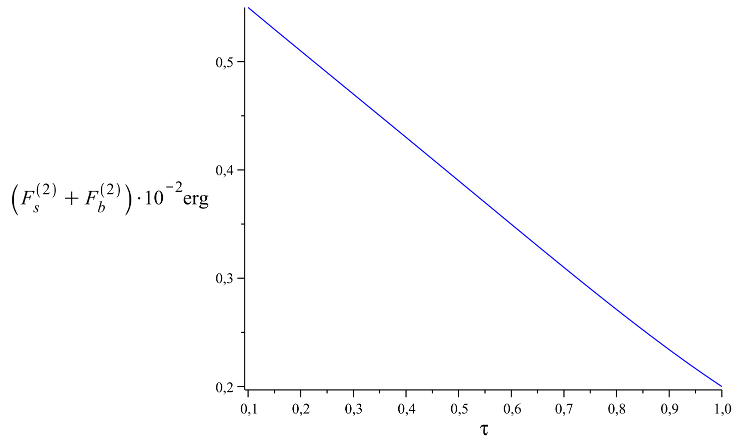

Fig. 2. Condition for the existence of perturbations in strong magnetic field. The area below the line corresponds to $F_b^{(2)} + F_s^{(2)} \ < \ 0$

3. Results and discussion

It is clear that for $\varphi(y,z) = \psi(y,z) = 0$, then $f_s^{(2)} = 0$. Therefore, the saddle-splay term does not contribute to the free energy for configurations in which the director is constant within a plane. The torsional strains considered here are two-dimensional, and so the contribution of k_{24} to $f_s^{(2)}$ is reasonable.

The state for $\omega < 1$ can not be achieved because it corresponds to complex wavenumbers q, i.e. $\omega^2 = 1 + q^2\varepsilon_\theta^2 d^2$.

Each curve in Figs. 3 divides the parametric space $\rho - \omega$ into two regions and the instability threshold is determined by the matrix $\mathcal{M}$. In view of Fig. 3a, the minimum of each curve represents the critical values of the dimensionless parameter ρ_c. The instability threshold between the stable state $\det \mathcal{M} > 0$ (below each curve) and periodically distorted state $\det \mathcal{M} \ < \ 0$ occurs in the vicinity of the glass surface for $0.53 \leqslant \rho_c \leqslant 0.578$.

The role of the saddle-splay term within the present problem statement can be seen by letting $k_{24} \to k$, $(\tau \to 1)$, then the stability diagram (17) grows faster, and means that wavelength of perturbations λ increases (Fig. 3b).

The obtained results show that for the determination of the saddle-splay constant one need to measure the distance $2d$ between two plates, elastic constants and by changing the magnetic field, observe periodic perturbations, and measure the wavelength at the plate surface. The determination of ω and τ can give a value of the saddle-splay constant with a certain error because the presented theoretical results are based on the one constant approximation.

Conclusions

In this paper we have studied the saddle-splay term, which is represented in the Frank free energy (1) and influences on the surface free energy. Periodic perturbations of the director yield deviation from the base state (5) when the saddle-splay energy

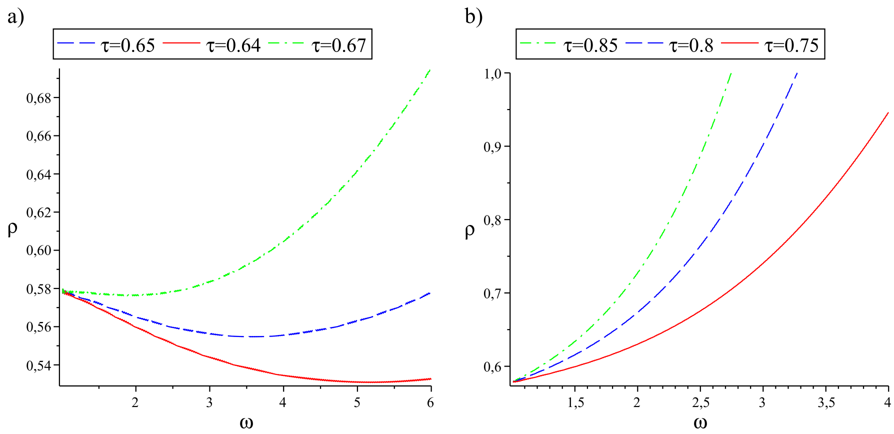

Fig. 3. Stability diagrams ($\det \mathcal{M} = 0$) for various values of the saddle-splay ratio $\tau = k_{24}/k$

becomes important. This issue becomes important only for high magnetic fields, i.e. $H \gtrsim 10^4$ G, which are difficult to achieve in many laboratories. The similarity of the effects induced by magnetic fields and electric fields shows that this effect can also be viewed in the electric field with ratio $\mathbf{E} = \sqrt{4\pi\chi_a/\varepsilon_a}\mathbf{H}$, where $\varepsilon_a = 0.1$ is the dielectric anisotropy. Thus, the behavior of LCs in magnetic field 10^4 G is equivalent to electric field with strength $E = 35.4$ V/cm.

Likewise in [4], this paper contributes to the theory of the surface terms on the periodic Fréedericksz transition. However, the analysis performed in the present paper has shown that there is no low threshold for τ when the bifurcation occurs in the layer of nematic LC with a finite thickness. Moreover, the critical values of parameter ρ_c satisfy the range criterion $0.53 \leqslant \rho_c \leqslant 0.578$.

The differential equations (10,11) written in terms of the two-constant approximation can give more accurate results at the expense of algorithmic complexity. In particular, the solution of the variational problem will cause two wave vectors and more complex boundary conditions.

Finally, we remark that the experimental description of the boundary orientational relaxation of the director caused by k_{24} modulus requires the study in optical transmittance of aligned nematic liquid crystals [6]. The authors will continue to treat the problem in this direction.

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