Electro–optic response in thin smectic C* film with chevron structures*

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The effects in electrostatic models of chevron surface-stabilized ferroelectric liquid crystals are investigated through numerical modeling. To study smectic C* director distribution within the cell, we consider two nonlinear approaches: the chevron interface does not interplay with the electric field; the electric field interplays with the chevron interface. The obtained results of the director field distribution are compared with the earlier linearized studies. We find that whether or not the electric field interplays with the chevron interface, the electro–optic response requires a generalized approach for its description. The threshold electric field, which is necessary for switching between two stable director states in the chevron cell is evaluated. This study suggests that, in many cases of practical interest, electro–optic response to the electric field and the threshold electric field can be precisely estimated. We argue that, beside being numerically efficient, our approach provides a convenient and a novel standpoint for looking at the electro–optic response problem.

Keywords: surface-stabilized ferroelectric liquid crystal, chevron structure, thin films, bistability, Euler's equation

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1. Introduction

Considerable efforts have been made since Meyer *et al's* pioneering work[1] to exploit the physics of ferroelectric liquid crystals (FLCs) in smectic C* (SmC*) phase. Correspondingly, a number of studies in SmC* liquid crystals confined between parallel boundaries and subjected to homogeneous boundary conditions is now available. For the so-called "bookshelf" geometry, the orientational order of the long SmC* molecular axis in thin films exhibits a one-dimensional positional order of the molecules arranged in a layered structure.

In the "bookshelf" geometry, smectic layers are normal to the cell surfaces, and often spontaneously deform into the chevron structures due to strong positional anchoring. In such structures, the layer edges at the boundary surfaces are not shifted, but the layers are tilted, and meet in the center of SmC* sample, which is called the chevron tip. Consequently, the understanding of the chevron structure is of considerable importance for the development of applications based on FLCs.

A series of theoretical models of chevron structures has been developed in recent decades. The pioneering theoretical description of the chevron structures in terms of a discontinuity in the layer tilt, while maintaining a continuous director structure at the chevron interface was given by Clark and Rieker.[2] A model of compressible layers was considered in studies.[3–5] Another approach assumed that the molecular tilt in FLCs is related to the layer tilt.[6] In Ref. [7], Limat extended the model,[4] where unequal cone and layer tilt angles are allowed. A similar phenomenological Landau–de Gennes approach was employed in a series of other studies (see e.g., Refs. [8]–[11]).

The early studies[2,12] consider models, where the layer tilt angle is fixed, i.e., $\pm\delta_0$ in each part of the chevron, but the azimuthal angle φ is varied between the plates, while the cone angle θ is the constant. Thus, the system represents two flat crossing planes at the angle $2\delta_0$. Continuous researchers' interest to FLCs generated another model with fair simplifications,[13] where Nakagawa's approach[3,4] was developed.

The chevron structure of smectic layers is a well-known characteristic of most surface-stabilized cells filled with FLC in the SmC* phase. These cells generally exhibit two stable director states. They can be switched between the stable states by an external voltage applied to the cell. Switching occurs only at fields stronger than the threshold field $E_{\rm th}$ because the director rotation is accompanied by a significant increase in the elastic energy around the chevron tip.[2] The bistability of such cells does not allow getting the desirable gray scale performance.[14,15]

Field-induced molecular reorientations in a monolayer of SmC* can be investigated by determining the spatial dependence between the azimuthal angle φ of the director and the applied electric field. Earlier, simplified models of biaxial

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modeling[16] and electro–optic response[13,17,18] were developed. The main simplifications in these studies regard the assumption that the layer tilt angle and the cone angle are small, which correspond to the temperature range close to the SmC*-SmA* (smectic A*) phase transition that has many dislocation defects.[19] Therefore, for lower temperatures[20] the electro–optic response of SmC* is expected to be different from the previous models with fair simplifications, but for a narrow temperature range.

In this study we aim to develop the generalized electrostatic model of the chevron FLC in the SmC* phase. On the basis of the continuum theory, we derive the functional, which represents the total free energy density of SmC*, and explains why the dielectric anisotropy term for weak electric fields should not be ignored. To calculate the spatial dependence of the azimuthal angle, we minimize the free energy density assuming that the smectic layers interplay with the electric field and do not. The threshold field $E_{\rm th}$ is calculated for the generalized model.

The plan of the article is as follows. The model and basic equations are presented in Section 2. In Section 3 we present numerical simulations under the assumption that the chevron configuration does not depend on the electric field. In Section 4 we turn our attention to the model when the chevron interface couples with the electric field. Details on computation of the threshold electric field are sketched in Section 5. Concluding remarks are given in Section 6.

2. Model and free energy

We examine a cell of thickness d filled with SmC* in the chevron configuration, and confined between two parallel substrates. Let the cell be maintained at a temperature far below the SmC*–SmA* phase transition.[19,20] It is known that molecules of SmC* form a layered structure and are tilted from the layer normal $\boldsymbol{a}$ by a fixed angle θ as shown in Fig. 1(a). The molecules are translationally anchored along the confining surfaces with the period of the smectic layer spacing h. The average direction of the molecular long axis is characterized by the director field $\boldsymbol{n}$, which also represents the optical axis. The unit orthogonal projection of $\boldsymbol{n}$ onto the tangential to the smectic plane is called the $\boldsymbol{c}$-director. With the given geometry, the director field is represented by

$$\begin{aligned}\boldsymbol{n} = {} & (-\sin\theta\cos\delta\sin\varphi + \sin\delta\cos\theta)\,\boldsymbol{e}_x + \sin\theta\cos\varphi\,\boldsymbol{e}_y \\ & + (\sin\delta\sin\theta\sin\varphi + \cos\delta\cos\theta)\,\boldsymbol{e}_z, \end{aligned} \tag{1}$$

where $\boldsymbol{e}_x$, $\boldsymbol{e}_y$, $\boldsymbol{e}_z$ are the bases of the Cartesian coordinate system, δ is the local tilt angle of the smectic layer, which is also the angle between the z axis and the smectic layer normal, i.e., $\boldsymbol{a} = (\sin\delta, 0, \cos\delta)$. We exclude edge dislocations that increase the number of layers along the y axis. However, we assume that the layer displacement field $u(x)$ between the chevron tip and the substrates admits a misalignment of the smectic layer normal (see Fig. 1(b)).

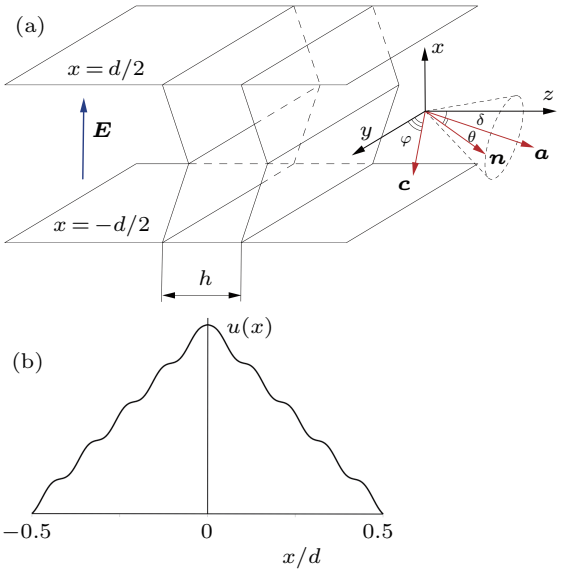


Fig. 1. (color online) (a) Spatial orientation of vectors $\boldsymbol{a}$, $\boldsymbol{c}$, and director $\boldsymbol{n}$ in the chevron surface-stabilized FLC cell. Electric field $\boldsymbol{E}$ is applied across the cell. Here the x axis is perpendicular to the substrate plates, the y axis is parallel to the chevron tip. (b) Model smectic layer interface; the displacement field is measured along the z axis.

The total free energy density f of the layer includes the elastic free energy density $f_{\rm elas}$, contribution to the electric energy density $f_{\rm elec}$, and deformation of smectic layers f_u, i.e., $f = f_{\rm elas} + f_{\rm elec} + f_u$. A commonly used term for $f_{\rm elas}$ in the one-constant approximation can be written as

$$f_{\rm elas} = \frac{K}{2}\left((\nabla\cdot\boldsymbol{n})^2 + (\nabla\times\boldsymbol{n})^2\right), \tag{2}$$

where K is the Frank elastic constant.[21]

The spontaneous polarization vector $\boldsymbol{P}$ gives a significant contribution to the orientation of $\boldsymbol{n}$, and it is defined[22] by the relation

$$\boldsymbol{P} = P_0\frac{\boldsymbol{a}\times\boldsymbol{n}}{|\boldsymbol{n}\times\boldsymbol{a}|}, \quad P_0 = |\boldsymbol{P}|.$$

The chevron tip significantly influences the orientational ordering of the director field and the spontaneous polarization. In particular, the spontaneous polarization allows switching of the director from up to down position, which are called U- and D-states with $n_y(0) < 0$ and $n_y(0) > 0$ correspondingly. A more detailed insight into these states is given in Refs. [2] and [23]. A commonly used electrostatic contribution to the free energy density has the form

$$f_{\rm elec} = -(\boldsymbol{P}\cdot\boldsymbol{E}) - \frac{1}{2}(\boldsymbol{D}\cdot\boldsymbol{E}). \tag{3}$$

The relation between the electric displacement $\boldsymbol{D}$ and the applied electric field can be expressed via the vectorial representation

$$\boldsymbol{D} = \varepsilon_0\varepsilon_\perp\boldsymbol{E} + \varepsilon_0\varepsilon_{\rm a}(\boldsymbol{n}\cdot\boldsymbol{E})\cdot\boldsymbol{n}, \tag{4}$$

where ε_0 is the electric constant, $\varepsilon_\perp$ is the unitless dielectric constant measured perpendicular to the FLC director, ε_a is the dielectric anisotropy, which can be positive or negative.[22,24]

In order to introduce the contribution to the free energy density, associated with mechanical dislocations of smectic layers, consider a molecular length a_0 and the smectic layer spacing h along the z axis. The layer spacing can be found by the identity

$$h = \frac{a_0 \cos\theta}{\cos\delta}.$$

The energy density related to the strain along the z axis and deformation of smectic layers can be evaluated by the functional

$$f_u = \frac{K_u}{2}\left(\frac{\mathrm{d}^2 u}{\mathrm{d}x^2}\right)^2 + \frac{B}{2}\left(1 \pm \frac{\cos\theta}{\cos\delta}\right)^2, \tag{5}$$

where B is the de Gennes smectic layer compressibility elastic constant, K_u is the Frank elastic constant associated with the splay deformation.[25] We note that in Eq. 5 identities $\sqrt{1+\tan^2\delta} = \pm 1/\cos\delta$ and $\cos(\arctan\delta) = 1/\sqrt{1+\delta^2}$ were applied.[26] For the electric field applied along the $+x$ axis,[27] we choose '+' in functional (5).

With the above assumptions, the total free energy density takes the form

$$\begin{aligned} f = {} & \frac{K}{2}\sin^2\theta\left(\frac{\mathrm{d}\varphi}{\mathrm{d}x}\right)^2 - \frac{K}{2}\sin 2\theta\cos\varphi\frac{\mathrm{d}\delta}{\mathrm{d}x}\frac{\mathrm{d}\varphi}{\mathrm{d}x} + \frac{K}{2}\left(\frac{\mathrm{d}\delta}{\mathrm{d}x}\right)^2\left(\sin^2\varphi + \cos^2\theta\cos^2\varphi\right) \\ & + P_0 E\cos\delta\cos\varphi - \frac{\varepsilon_0\varepsilon_a E^2}{2}\left(\cos\delta\sin\varphi\sin\theta - \cos\theta\sin\delta\right)^2 - \frac{\varepsilon_0\varepsilon_\perp E^2}{2} \\ & + \frac{B}{2}\left(1 + \frac{\cos\theta}{\cos\delta}\right)^2 + \frac{K_u}{2\cos^4\delta}\left(\frac{\mathrm{d}\delta}{\mathrm{d}x}\right)^2, \end{aligned} \tag{6}$$

where it is supposed that δ and φ depend on the coordinate along the thickness of the film. In general, the contribution due to dielectric anisotropy is ignored. Here, we do not ignore the dielectric term because it has a φ-dependence different from the P_0E term. If $\varphi = \pi/2$, then the dielectric term will be larger by absolute value than the vanishing P_0E term. In addition, the top and bottom substrates with a plurality of electrodes can apply electric field to the SmC* layer at different angles.[28] The corresponding oblique electric field changes the total free energy density in a sample of SmC*.[29]

3. Decoupled chevron layer with electric field

Supposing that the shape of the chevron layer displacement field $u(x)$ does not depend on the electric field, we minimize f with respect to φ. After routine trigonometric simplifications, the Euler–Lagrange equation takes the form

$$\begin{aligned} & K\left(\sin^2\theta\left[\frac{1}{2}\sin 2\varphi\left(\frac{\mathrm{d}\delta}{\mathrm{d}x}\right)^2 - \frac{\mathrm{d}^2\varphi}{\mathrm{d}x^2}\right] + \frac{1}{2}\sin 2\theta\cos\varphi\frac{\mathrm{d}^2\delta}{\mathrm{d}x^2}\right) \\ & \quad - \frac{\varepsilon_0\varepsilon_a E^2}{2}\left(\sin^2\theta\cos^2\delta\sin 2\varphi - \frac{1}{2}\sin 2\theta\sin 2\delta\cos\varphi\right) - P_0 E\sin\varphi\cos\delta = 0. \end{aligned} \tag{7}$$

We note that for the "bookshelf" geometry with the disregarded dielectric anisotropy term, equation (7) takes the well-known form[22]

$$\frac{\mathrm{d}^2\varphi}{\mathrm{d}x^2} + \frac{P_0 E}{K\sin^2\theta}\sin\varphi = 0.$$

Before investigating the solution of Eq. (7), we need to introduce the boundary conditions, and set the displacement field $u(x)$ independent of $\delta(x)$ and cell thickness.

The chevron interface in the cell center impacts on the orientational ordering of the director field $\boldsymbol{n}$, in particular, $n_x(0) = 0$. Following the basic requirement for bistability in surface-stabilized cells, i.e., $\delta < \theta$, cones intersect at the open circles. Here, we assume strong anchoring of the director in the plane of the boundary plates. Under this assumption, we use the following boundary conditions for the D state

$$\varphi(0)|_{x\to\pm 0} = \pm\varphi_0, \quad \varphi\left(\pm\frac{d}{2}\right) = \pm\frac{\pi}{2}, \tag{8}$$

where $\varphi_0 = \arcsin(\tan\delta/\tan\theta)$. In view of Eq. (6), the free energy density of the system remains constant when φ is replaced by $\pi - \varphi$, which means the presence of two equilibrium configurations, i.e., the bistability of the system.[2] Therefore, the boundary conditions for the U-state can be written as

$$\varphi(0)|_{x\to\pm 0} = \pm(\pi - \varphi_0), \quad \varphi\left(\pm\frac{d}{2}\right) = \pm\frac{\pi}{2}. \tag{9}$$

For a decoupled chevron layer with the electric field, we introduce the displacement piecewise function

$$u(x) = \begin{cases} -2\times10^{-8}\sin(1.2\times10^{7}x)+0.3x+7.698\times10^{-7}, & -d/2 \le x \le 0, \\ 2\times10^{-8}\sin(1.2\times10^{7}x)-0.3x+7.698\times10^{-7}, & 0 < x \le d/2, \end{cases} \tag{10}$$

which we define in accordance with the range of common layer tilt angles and angular undulations in smectic layers, e.g., Refs. [24], [30], and [31]. Once $u(x)$ is known, the azimuthal angle $\varphi(x)$ across the cell can be evaluated from Eq. (7) and the identity $\tan\delta = -\mathrm{d}u/\mathrm{d}x$ with the boundary conditions (8) and (9).

To substantiate the assessment of the φ-dependencies with common parameters $d = 5\,\mu\mathrm{m}$, $\theta = 22.5^\circ$, $K = 5\times10^{-12}$ N, $\varepsilon_a = 2.5$, $P_0 = 2.9\times10^{-4}\,\mathrm{C\cdot m^{-2}}$, we plot Figs. 2 and 3 for a range of electric fields.

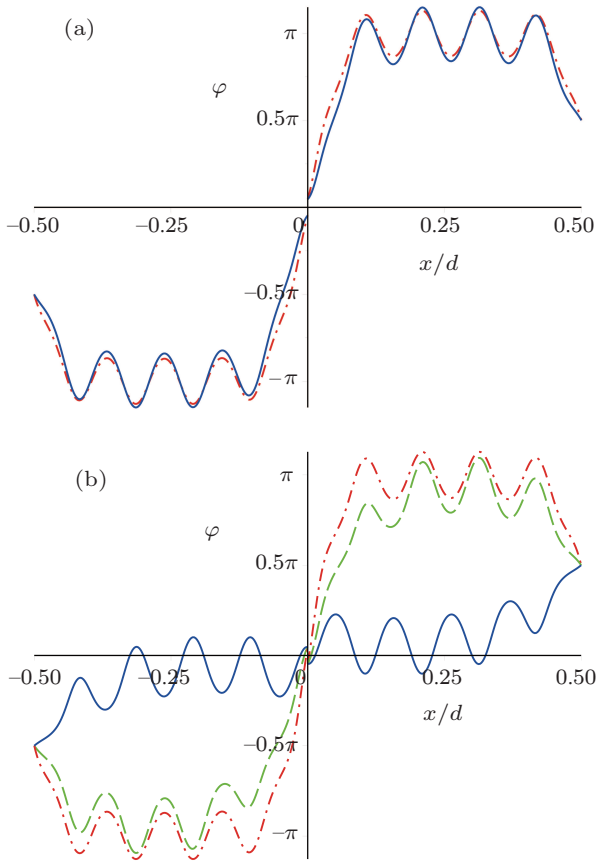


Fig. 2. (color online) Spatial orientation of the azimuthal angle φ in the D-state for $E = 10^2\,\mathrm{V\cdot m^{-1}}$ (blue solid line), $E = 10^4\,\mathrm{V\cdot m^{-1}}$ (green dashed line), $E = 10^5\,\mathrm{V\cdot m^{-1}}$ (red dashed–dotted line): (a) simplified model for small θ and δ; (b) model of common approach Eq. (7) with boundary conditions (8).

As we expected, consideration of common polar angle and space-dependent layer tilt angle yields several differences in spatial distribution of the c-director field from its profile based on the linearized models for small δ and θ. Our computations indicate that in the generalized approach the φ-dependence differs from that found from models.[13,18] The proposed model indicates that weak electric field is unable to reorient the c-director parallel to the plates due to the interplay between the P_0E term, strong boundary conditions, and non-coupling layer tilt with the electric field (Fig. 2(b). The φ-dependencies in Fig. 2(b) show that when the voltage between the plates increases, the electric energy density dominates over the elastic energy density, and thereby the c-director tends to align parallel to the plates. Another interesting detail of our solution is that the orientation of the c-director is governed by the chevron interface: the intervals, where $u(x)$ and $\varphi(x)$ are convex, do coincide, and *vice versa*.

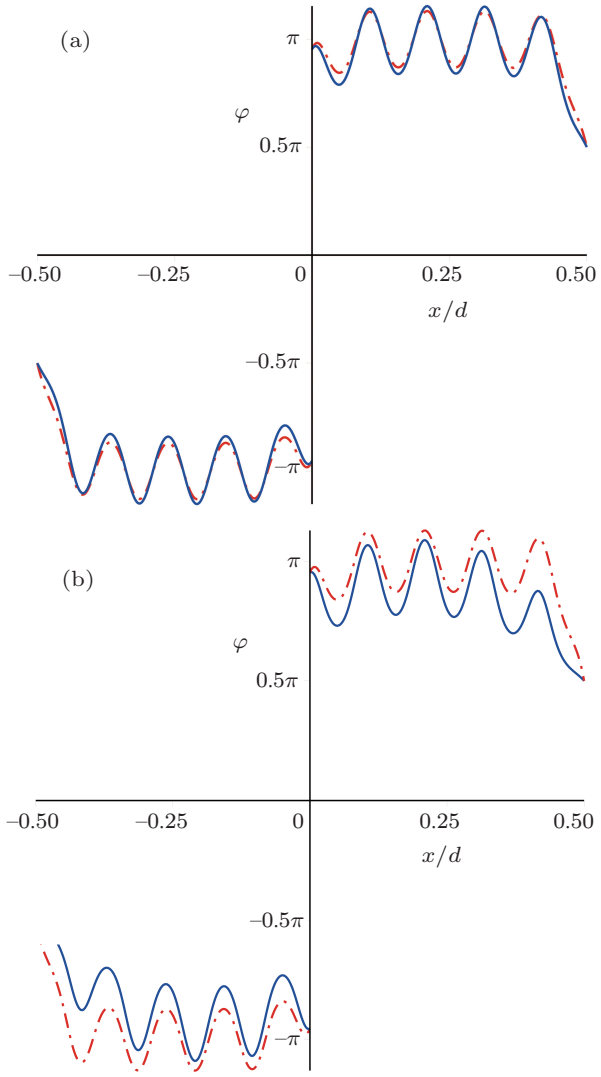


Fig. 3. (color online) Spatial distribution of the c-director field for the U-state with $E = 10^2\,\mathrm{V\cdot m^{-1}}$ (blue solid line), $E = 10^5\,\mathrm{V\cdot m^{-1}}$ (red dashed–dotted line): (a) existing model for small θ and δ; (b) model of common approach Eq. (7) with boundary conditions (9).

When the director is in the U-state, the deviations between the generalized model and the simplified model are less significant for the weak electric field and the strong electric field because the c-director has already oriented almost paral-

lel to the boundary plates (as shown in Fig. 3). As well as for the D-state, the undulated $\varphi-$dependence is due to the structure of the layer displacement field. The role of the spontaneous polarization becomes important if one needs to reduce the undulation amplitude in the φ-dependence. This can be implemented by increasing the spontaneous polarization. In accordance with the selected model parameters, we remark that the electrostatic contribution f_{elec} in the simplified model, plays a subdominant role in the alignment of the c-director. So, it is apparent that the electro–optic response of the SmC* requires a generalized approach for its description.

4. Coupled chevron layer and electric field

A number of factors can influence the layer configuration when SmC* molecules interact with the electric field. At this point it is worth examining the possible layer and director configurations within the cell. The system will show equilibrium structures of the c-director and layer configurations that reach minimum states of the total free energy density (6). In addition to the governing equation for azimuthal angle (7), minimization of Eq. (6) with respect to $\delta(x)$ gives the following equation

$$K\left(\sin^2\theta\left[\cos^2\varphi\frac{\mathrm{d}^2\delta}{\mathrm{d}x^2}-\sin 2\varphi\frac{\mathrm{d}\delta}{\mathrm{d}x}\frac{\mathrm{d}\varphi}{\mathrm{d}x}\right]+\frac{1}{2}\sin 2\theta\left[\cos\varphi\frac{\mathrm{d}^2\varphi}{\mathrm{d}x^2}-\sin\varphi\left(\frac{\mathrm{d}\varphi}{\mathrm{d}x}\right)^2\right]-\frac{\mathrm{d}^2\delta}{\mathrm{d}x^2}\right)$$
$$+\frac{\varepsilon_0\varepsilon_a E^2}{2}\left(\sin^2\theta\sin^2\varphi\sin 2\delta+\sin 2\theta\sin\varphi\cos 2\delta\right)-\frac{K_u}{\cos^4\delta}\frac{\mathrm{d}^2\delta}{\mathrm{d}x^2}$$
$$+\sin\delta\left[\cos^2\theta\left(\frac{B}{\cos^3\delta}-\varepsilon_0\varepsilon_a E^2\cos\delta\right)-\frac{B\cos\theta}{\cos^2\delta}-P_0E\cos\varphi-\frac{2K_u}{\cos^5\delta}\left(\frac{\mathrm{d}\delta}{\mathrm{d}x}\right)^2\right]=0. \tag{11}$$

The governing equations (7) and (11) were subsequently solved numerically with boundary conditions for strong anchoring

$$\varphi\left(\pm\frac{d}{2}\right)=\pm\frac{\pi}{2},\quad \delta\left(\pm\frac{d}{2}\right)=\pm\mu\theta, \tag{12}$$

where μ is the coefficient that shows relative difference between δ and θ.

In Fig. 4, we substantiate the difference between the nonlinear model and the linear model[13,17,18] of the SmC* layer tilt angle and the splay deformation of the c-director. According to the nonlinear model, we observed that for the electric field applied along the $+x$ axis, the c-director deforms between the chevron tip (at $x=0$) and the boundary surfaces (at $x=\pm d/2$).

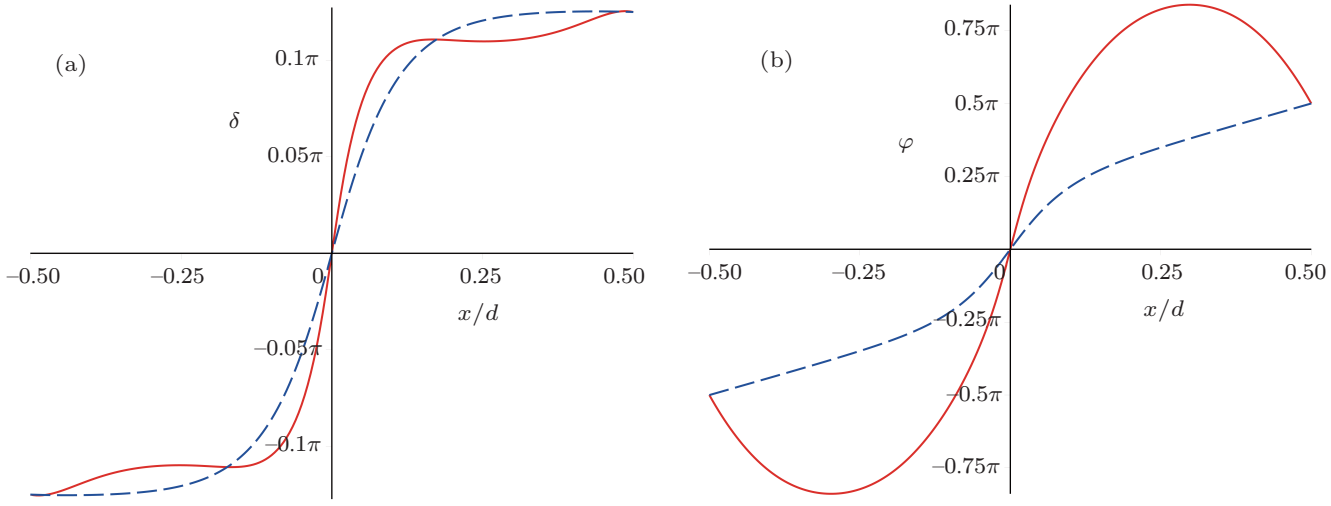


Fig. 4. (color online) Numerical solutions of equations (7) and (11) shown by red solid curves; model for small smectic cone angle θ and chevron tilt angle δ (blue dashed curves): (a) the dependence of the layer tilt δ upon the normalized x-coordinate; (b) the spatial dependence of the azimuthal angle within the monolayer sample of chevron SmC*. Parameters of the problem are $E=10^4\,\mathrm{V\cdot m^{-1}}$, $K_u=5\times10^{-12}$ N, $B=4\times10^6$ N, and $\mu=0.85$.

Layer bending, which is characterized by $\delta(x)$ is greatly dependent on the constant B.[13] As indicated in Fig. 4(a), layers bend very slightly that confirms previous experimental data.[30,32] Due to the layer bending induced by the electric field, the splay deformation of the c-director field reduces in comparison with the φ-dependence in the D-state (Fig. 2). In addition to the obtained results, numerical computations show that the solution of governing equations (7) and (11) does not

give any significant error [the absolute error for φ and δ is 10^{-4}]. At this point, we stress that here we have obtained the model for determination of the chevron interface.

5. Threshold electric field

To find the threshold field $E_{\rm th}$, we suppose that the director switches from the D- to the U-state, i.e., rotating by angle π. When the director switches to the U-state, molecules rotate along the allowed trajectory forming the structure, which is energetically unfavorable, and determines the potential barrier W [the smectic cone angle reduces to zero, i.e., the liquid crystal in the smectic A phase]. Therefore, the difference between free energy of the system in the state $F_{\pi/2}$ with $\varphi(0)=\pi/2$ and free energy of the D- or U-state F_0 represents the potential barrier, i.e., $W=F_{\pi/2}-F_0$.

Having determined profiles for δ and φ from the model represented in Sections 3, the electrostatic forces holding SmC* dipoles at their equilibrium state can be expressed by the integral

$$F_{\rm elec}=-S\int_{-d/2}^{d/2} f_{\rm elec}\,{\rm d}x, \tag{13}$$

where

$$f_{\rm elec}=P_0E\cos\delta\cos\varphi - \frac{\varepsilon_0E^2}{2}\left[\varepsilon_\perp+\varepsilon_a\Big(\cos\theta\sin\delta-\sin\theta\cos\delta\sin\varphi\Big)^2\right].$$

Here S is the area of the sample. To perform computations of this integral for a model of non-coupled electric field with smectic layers, we represent Eq. (13) as the sum of contributions for $-d/2\le x\le 0$ and $0<x\le d/2$. In order to get the threshold value of the applied electric field $E_{\rm th}$ in switching of SmC* molecules, which are described by means of non-linearized system of equations, we extend the idea given in Ref. [17]. The contribution of the electric energy density in our case includes the dielectric term as well. The threshold field $E_{\rm th}$ can be evaluated from the condition of the equality of the total energy of the interaction with the electric field Eq. (13) to the potential barrier W. The preceding investigation of the potential barrier leads us to expect that the barrier height represents a function of the cell thickness, i.e., $W=W(d)$. With the previously set parameters for undulated chevron structures, we obtain $E_{\rm th}\approx 2790\,{\rm V\cdot m^{-1}}$ with $\varepsilon_\perp=5$. Further computations confirm that the threshold electric field depends on the cell thickness: $E_{\rm th}$ monotonically decreases with increasing d.[17,27]

The results obtained in this study are more general because we studied non-linearized potential (6) and substituted δ- and φ-dependencies into functional (13). The computations show that the undulated chevron profiles require higher threshold electric field than undistorted chevron layers.

6. Conclusions

We have presented a theoretical study that determines the spacial dependence of the director field by taking into account a detailed layer structure and common parameters of SmC*. Many recognized electro–optical phenomena in SmC* with a chevron structure could be satisfactorily explained by this generalized description. Hereby, the presented study of electro–optic response in FLCs can be successful when we cannot ignore a number of terms in Eq. (6) and splay undulation of SmC* layers.

Two separate aspects of the influence of the electric field on the layer structure were studied. First, the model of non-coupled smectic layers with electric field. Second, the problem of coupled layers with the electric field. The latter problem represents the system of nonlinear differential equations of the second order, and it gives comprehensive results. Calculations of the azimuthal angle distribution in such layers were investigated by numerical methods within the framework of the continuum model. The obtained δ- and φ-dependencies were compared with the results derived from the model for small layer tilt and cone angle. The differences between the models of our focus imply that the issues of the study may be useful in optimization of mesomorphic properties of liquid crystal cell.

The threshold electric field, which is necessary for switching between two stable configurations was determined for non-coupled electric field with smectic layers. The generalized model is consistent with the previous studies, and confirms that $E_{\rm th}$ is inversely proportional to the cell thickness. To calculate $E_{\rm th}$ for the model of coupled electric field with smectic layers, a more accurate description of the interaction between the chevron structures and the electric field is required.

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