Splay-bend periodic deformation in nematic liquid crystal slabs

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We predict a two-dimensional splay-bend periodic deformation in a nematic slab with homeotropic boundary conditions. The nematic director modulation is induced by surface contributions in the elastic energy, which are linear in the deformation tensor. The instability appears only if the surface anchoring energy strength is small enough, and if the two surfaces are different. The wave vector of the stripe modulation is proportional to the thickness of the nematic slab. The order of magnitude of the surface elastic constants relevant to the linear elastic terms in the deformation tensor, and the critical value of the anchoring energy, $\lesssim 10^{-6}$ J/m^2, to observe the instability are discussed.

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I. INTRODUCTION

Stripe physics is an example of a nontrivial ordering phenomenon in condensed-matter physics. In particular, stripe patterns in achiral centrosymmetric materials have been of intense interest both for the underlying physics and for the possible connection to applications of these materials. In the case of samples in the form of thin films and of nanostructures, surfaces suppress the inversion symmetry of the bulk and modulated structures may appear.

Liquid crystals are anisotropic fluids formed by anisometric molecules. In the achiral nematic phase, molecules have random positions but align themselves along an average direction called nematic director $\mathbf{n}$ ($\mathbf{n}^2=1$), thus nematics behave as uniaxial crystals with positional isotropy [1]. In nematic media, modulated structures have been predicted and observed either under the action of an external bulk field or in nematic slabs with antagonistic alignment conditions (mechanical field). The possibility to observe periodic structures induced by an external magnetic field has been discussed long ago by Londberg and Meyer [2]. In a different context, e.g., instability induced by a mechanical field, the same problem has been discussed by several authors [3–9]. Recently, it has been analyzed under what conditions periodic structures can appear spontaneously in nematic liquid crystals (NLC), and it has been shown that the saddle-splay elastic constant could be responsible for a ground state presenting periodic modulation [10]. The latter periodic deformation involves splay-bend and twist deformations, i.e., it is a three-dimensional (3D) deformation [11,12].

Lately, an important effort in the LC technology concerns the realization of weak anchoring conditions in such a manner to have a low working voltage for display applications [13]. However, as we will show in this paper, at low enough anchoring energy, new effects can appear: the presence of a surface field inducing Lifshitz-invariants-like terms, can destroy the homogeneous alignment. In the absence of external fields, these terms are identically zero in the bulk. However, close to a limiting surface, where the bulk nematic symmetry is broken, these can exist. In particular, we predict a different mechanism of periodic spontaneous deformation, in a NLC slab confined by homeotropic surfaces, that acts in absence of any external field or competing anchoring, and involves only a splay-bend deformation: $\mathbf{n}$ is always parallel to a plane (2D deformation, described by a tilt angle). The periodic structure can be induced by elastic terms linear in the deformation tensor, but only when a rather weak anchoring energy is available. The characteristics of the periodic splay-bend deformation as well as the conditions under which it can be observed are discussed. A similar structure has recently been considered by another method, in the case of thin ferromagnetic films [15].

II. ELASTIC PROBLEM

In our analysis, we consider a nematic slab limited by two substrates that induce homeotropic alignment. Cartesian reference frame is chosen with z axis, of unit vector $\mathbf{k}$, perpendicular to the limiting surfaces at $z=\pm d/2$. The total elastic free-energy density of the nematic sample is given by

$$f=f_s+f_l+f_q. \tag{1}$$

The surface tension term f_s accounts for the nematic-substrate interactions, and as usually is given in the form [16]

$$f_s=f_{s0}-\frac{1}{2}V(z)(\mathbf{n}\cdot\mathbf{k})^2, \tag{2}$$

where f_{s0} is the isotropic part of the surface tension and the $V(z)$ term describes the nematic orientation dependent part of the surface tension [17]. In the following, we will assume that $V(z)=V_0U(z)$, where V_0 is the maximum value of $V(z)$ and $U(z)$ is a positive function different from zero only in the surface layers at $z\sim\pm d/2$. For homeotropic alignment, the anchoring energy strength V_0 is positive.

Terms f_l and f_q depend on the deformation tensor with elements $n_{i,j}=\partial n_i/\partial x_j$, where

$$f_l=-R_1(z)(\mathbf{n}\cdot\mathbf{k})(\boldsymbol{\nabla}\cdot\mathbf{n})-R_3(z)\mathbf{k}\cdot[\mathbf{n}\times(\boldsymbol{\nabla}\times\mathbf{n})] \tag{3}$$

is linear in the deformation tensor. The surface elastic constants $R_1(z)$ and $R_3(z)$ are different from zero only in the surface layers where the nematic symmetry of the bulk is broken [18]. The functional dependence of f_l on $n_{i,j}$ is identical to the flexoelectric free energy term, which appears when a NLC is submitted to a dc electric field [1]. Therefore, the presence of a limiting surface is equivalent to a kind of surface field. In the case of a solid isotropic surface, this surface field is of the form $\mathbf{E}(\zeta)=E(\zeta)\mathbf{k}$ where ζ is the distance from the solid surface. The coupling of $\mathbf{E}(\zeta)$ with the polar properties induced by a mechanical deformation gives rise to f_l. Note that if the nematic sample is limited by two identical substrates, the relevant surface field is an odd function of z.

f_q is the usual quadratic elastic free-energy density of NLCs, which can be written as the sum of the Frank free-energy density [19]

$$f_F=\frac{1}{2}[K_{11}(\nabla\cdot\mathbf{n})^2+K_{22}[\mathbf{n}\cdot(\nabla\times\mathbf{n})]^2+K_{33}[\mathbf{n}\times(\nabla\times\mathbf{n})]^2] \tag{4}$$

and of a surfacelike term

$$f_{SL}=-(K_{22}+K_{24})\nabla\cdot[\mathbf{n}\nabla\cdot\mathbf{n}+\mathbf{n}\times(\nabla\times\mathbf{n})]. \tag{5}$$

K_{11}, K_{22}, and K_{33} are the splay, twist, and bend bulk elastic constants, respectively, which favor homogeneous states, and K_{24} is the saddle-splay elastic constant. The term associated with K_{24}, by means of Gauss theorem, gives rise only to a surface contribution [20], but it has nothing to do with the surface properties of the material.

Now we assume that the nematic director is everywhere parallel to the x-z plane: $\mathbf{n}=\mathbf{i}\sin\theta+\mathbf{k}\cos\theta$, where $\mathbf{i}$ is the unit vector parallel to the x axis and θ is the tilt angle that $\mathbf{n}$ forms with z axis. Since $\theta=\theta(x,z)$ then $\mathbf{n}\nabla\cdot\mathbf{n}+\mathbf{n}\times(\nabla\times\mathbf{n})=-\mathbf{i}\theta_{,z}+\mathbf{k}\theta_{,x}$, where $\theta_{,x}=\partial\theta/\partial x$ and $\theta_{,z}=\partial\theta/\partial z$. Consequently, the surfacelike contribution vanishes identically: $f_{SL}=0$. The present analysis is restricted in the one elastic constant approximation in the bulk ($K_{ii}=K$), and in the interfacial layers: $R_1(z)=R_3(z)=R(z)$. The general anisotropic approach will be given elsewhere. In this framework, the total free-energy density, beside a constant term, is given by

$$f=-\frac{1}{2}V(z)\cos^2\theta-R(z)\theta_{,x}+\frac{1}{2}K(\theta_{,x}^2+\theta_{,z}^2). \tag{6}$$

Since we look for a periodic deformation along the x axis, the nematic tilt angle θ has to be of the type $\theta(x+\lambda,z)=\theta(x,z)+\pi$ because of the apolar character of the nematic director, where λ is the period of the modulated structure. Finally, the total energy per period of the nematic slab is

$$F=\int_{-d/2}^{d/2}\int_0^{\lambda} f\,dx\,dz. \tag{7}$$

By introducing the reduced coordinates $\xi=x/d$ and $\eta=z/d$, and the nondimensional constant $\varepsilon=V_0d^2/K$, F can be rewritten in the form

$$G=F/K=\int_{-1/2}^{1/2}\int_0^{\Lambda} g\,d\xi\,d\eta, \tag{8}$$

where $\Lambda=\lambda/d$ is the nondimensional wavelength and

$$g=fd^2/K=-\frac{1}{2}\varepsilon U(\eta)\cos^2\theta-\frac{R(\eta)d}{K}\theta_{,\xi}+\frac{1}{2}(\theta_{,\xi}^2+\theta_{,\eta}^2). \tag{9}$$

According to the variational calculus, for the function $\theta(\xi,\eta)$ to minimize G, the relevant differential equation is given by the condition $\delta G=\delta G_b+\delta G_\eta+\delta G_\xi=0$ [21], where

$$\delta G_b=\int_{-1/2}^{1/2}\int_0^{\Lambda}\left\{\frac{\partial g}{\partial\theta}-\left(\frac{\partial}{\partial\xi}\frac{\partial g}{\partial\theta_{,\xi}}+\frac{\partial}{\partial\eta}\frac{\partial g}{\partial\theta_{,\eta}}\right)\right\}\times\delta\theta(\xi,\eta)\,d\xi\,d\eta,$$

$$\delta G_\xi=\int_0^{\Lambda}\left\{\left(\frac{\partial g}{\partial\theta_{,\eta}}\right)_{\eta=1/2}\delta\theta(\xi,1/2)-\left(\frac{\partial g}{\partial\theta_{,\eta}}\right)_{\eta=-1/2}\times\delta\theta(\xi,-1/2)\right\}d\xi,$$

$$\delta G_\eta=\int_{-1/2}^{1/2}\left\{\left(\frac{\partial g}{\partial\theta_{,\xi}}\right)_{\xi=\Lambda}\delta\theta(\Lambda,\eta)-\left(\frac{\partial g}{\partial\theta_{,\xi}}\right)_{\xi=0}\delta\theta(0,\eta)\right\}d\eta. \tag{10}$$

The condition $\theta(\xi+\Lambda,\eta)=\theta(\xi,\eta)+\pi$ implies that the derivative $\theta_{,\xi}$ and the variation $\delta\theta(\xi,\eta)$ have the same periodicity, i.e., $\theta_{,\xi}(\xi+\Lambda,\eta)=\theta_{,\xi}(\xi,\eta)$ and $\delta\theta(\xi+\Lambda,\eta)=\delta\theta(\xi,\eta)$. By taking into account these conditions, δG_η is rewritten as

$$\delta G_\eta=\int_{-1/2}^{1/2}\left\{\left(\frac{\partial g}{\partial\theta_{,\xi}}\right)_{\xi=\Lambda}-\left(\frac{\partial g}{\partial\theta_{,\xi}}\right)_{\xi=0}\right\}\delta\theta(0,\eta)\,d\eta. \tag{11}$$

Substituting g from Eq. (9) into Eqs. (10) and (11), we obtain for δG_b, δG_ξ, and δG_η the expressions

$$\delta G_b=\int_{-1/2}^{1/2}\int_0^{\Lambda}\left\{\frac{\varepsilon}{2}U(\eta)\sin(2\theta)-(\theta_{,\xi\xi}+\theta_{,\eta\eta})\right\}\times\delta\theta(\xi,\eta)\,d\xi\,d\eta,$$

$$\delta G_\xi=\int_0^{\Lambda}\{\theta_{,\eta}(\xi,1/2)\,\delta\theta(\xi,1/2)-\theta_{,\eta}(\xi,-1/2)\times\delta\theta(\xi,-1/2)\}d\xi,$$

$$\delta G_\eta=\int_{-1/2}^{1/2}\{\theta_{,\xi}(\Lambda,\eta)-\theta_{,\xi}(0,\eta)\}\delta\theta(0,\eta)\,d\eta. \tag{12}$$

The periodicity $\theta_{,\xi}(\xi+\Lambda,\eta)=\theta_{,\xi}(\xi,\eta)$ yields $\delta G_\eta=0$. The condition $\delta G=0$, for all $\delta\theta(\xi,\eta)$ belonging to the C_1 class, such that $\delta\theta(\xi+\Lambda,\eta)=\delta\theta(\xi,\eta)$, gives the differential equation

$$\theta_{,\xi\xi}+\theta_{,\eta\eta}-\frac{\varepsilon}{2}U(\eta)\sin(2\theta)=0 \tag{13}$$

with the boundary conditions $\theta_{,\eta}(\xi,\pm 1/2)=0$.

III. PERTURBATION ANALYSIS

In what follows, we assume that $\varepsilon=V_0d^2/K\ll 1$. We expand θ as a power series in the small parameter ε: $\theta(\xi,\eta)=\theta_0(\xi,\eta)+\varepsilon\theta_1(\xi,\eta)+\varepsilon^2\theta_2(\xi,\eta)+\cdots$. Then we substitute it in Eq. (13) and in the relevant boundary conditions, and equate corresponding powers of ε. At the zeroth order in ε, Eq. (13) becomes

$$\theta_{0,\xi\xi}(\xi,\eta)+\theta_{0,\eta\eta}(\xi,\eta)=0, \tag{14}$$

and $\theta_{0,\eta}(\xi,\pm 1/2)=0$. While the first-order terms in ε give

$$\theta_{1,\xi\xi}+\theta_{1,\eta\eta}=\frac{1}{2}U(\eta)\sin(2\theta_0), \tag{15}$$

and $\theta_{1,\eta}(\xi,\pm 1/2)=0$.

Further, since the function $\theta(\xi,\eta)$ belongs to the class $\theta(\xi+\Lambda,\eta)=\theta(\xi,\eta)+\pi$, the perturbation expansion of θ yields $\theta_0(\xi+\Lambda,\eta)=\theta_0(\xi,\eta)+\pi$ and $\theta_i(\xi+\Lambda,\eta)=\theta_i(\xi,\eta)$, for $i\geq 1$. The general solution of Eq. (14) is then

$$\theta_0(\xi,\eta)=q\xi+p\eta+\sum_{n=1}^{\infty}\left\{A_n\cosh\left(n\frac{2\pi}{\Lambda}\eta\right)+B_n\sinh\left(n\frac{2\pi}{\Lambda}\eta\right)\right\}\sin\left(n\frac{2\pi}{\Lambda}\xi\right). \tag{16}$$

The relevant boundary conditions yield $p=0$ and $B_n\cosh(n\pi/\Lambda)\pm A_n\sinh(n\pi/\Lambda)=0$, for $n\geq 1$. The latter equation implies $A_n=B_n=0$. Thus, the solution, at the zeroth order in ε, is $\theta_0(\xi,\eta)=q\xi$. The wave vector q is then determined by imposing that the total free energy has a minimum. Substituting the perturbation expansion of θ into Eq. (9) and using the solution $\theta_0(\xi,\eta)=q\xi$, we obtain, to the first order in ε, $g=g_0+\varepsilon g_1$, where

$$g_0=-\frac{R(\eta)d}{K}q+\frac{1}{2}q^2,$$

$$g_1=-\frac{1}{2}U(\eta)\cos^2(q\xi)-\frac{R(\eta)d}{K}\theta_{1,\xi}+q\theta_{1,\xi}. \tag{17}$$

Since $\theta_1(\xi+\Lambda,\eta)=\theta_1(\xi,\eta)$, the total energy G per period is

$$G=G_0+\varepsilon G_1=\left(-\frac{rd}{K}q+\frac{1}{2}q^2\right)\Lambda-\frac{1}{4}u\Lambda\varepsilon, \tag{18}$$

where the parameters r and u are given by

$$r=\int_{-1/2}^{1/2}R(\eta)d\eta \quad \text{and} \quad u=\int_{-1/2}^{1/2}U(\eta)d\eta. \tag{19}$$

At the zeroth order in ε, the wave vector minimizing $\phi=G/\Lambda=G_0/\Lambda$ [22] is $q=rd/K$. The corresponding value of G_0 is $G_0(\min)=-(1/2)(rd/K)^2\Lambda$. At the first order in ε, we have

$$G=G_0+\varepsilon G_1=-\left\{\frac{1}{2}\left(\frac{rd}{K}\right)^2+\frac{1}{4}\varepsilon u\right\}\Lambda. \tag{20}$$

The modulated structure is stable if the relevant energy per period, G, is smaller than the energy G_H for homogeneous homeotropic alignment ($\theta=0$). From Eqs. (8) and (9), and (19), we obtain $G_H=-(\varepsilon/2)u\Lambda$. Then, at the zeroth order in ε, the modulated structure is always stable with respect to the homeotropic one. To the first order in ε, it is stable only if $G_0+\varepsilon G_1\leq G_H$, from which we derive $u\varepsilon\leq 2(rd/K)^2$. This inequality gives the upper limit of u to observe the periodic deformation. Instead of the quantities r and u, it is more convenient to introduce the parameters

$$b=\int_{-d/2}^{d/2}R(z)dz=rd \quad \text{and} \quad w=\int_{-d/2}^{d/2}V_0U(z)dz=u\varepsilon\frac{K}{d}. \tag{21}$$

In terms of b and w, the condition on the anchoring energy is written $w\leq 2b^2/Kd$, and the wavelength of the modulated structure is

$$\lambda=\Lambda d=(\pi/q)d=\pi(K/b)d, \tag{22}$$

i.e., it is proportional to the thickness of the sample. Using the latter expression for λ, the critical condition on the anchoring energy can be rewritten as

$$w\leq w_{cr}=2b^2/Kd=2\pi^2Kd/\lambda^2. \tag{23}$$

To the lowest approximation in ε, the above described modulated structure is z independent.

IV. DISCUSSION

As discussed above, if the nematic sample is limited by two identical substrates, the surface field is an odd function of the z coordinate: $R(z)=-R(-z)$, and therefore $b=0$, which leads to $\lambda\to\infty$, i.e., no-periodic deformation. In this situation, the linear terms in the deformation tensor could induce a deformation along the z coordinate, but cannot induce a periodic deformation along the x coordinate. Experimentally, the splay-bend instability could be obtained, e.g., with a nematic slab confined between two plates that induce homeotropic alignment, but treated with different kinds of surfactants. In practice, if not special care is taken, the two surfaces of a nematic cell are not identical even if the surfactant treatment is the same. Therefore, the latter condition, in order to observe the instability, can be easily fulfilled.

In the case that the linear elastic constants $R_1(z)$ and $R_3(z)$ have a flexoelectric origin, these are of the form $R_i(z)=e_{ii}E(z)$, where $E(z)$ is a surface field originating, e.g., from selective ionic adsorption [23]. $E(z)$ decreases in an exponential manner with the distance from the substrate, over a typical length of the order of the Debye screening length L_D. The amplitude of the surface field is $E_S=\sigma/\epsilon$, where σ is the surface density of adsorbed charges and ϵ is the average dielectric constant of the nematic medium. In this framework, for a compensated nematic liquid crystal with $e_{ii}\sim 10^{-11}$ C/m [24], $\sigma\sim 10^{-5}$ C/m^2, $\epsilon\sim 5\epsilon_0$, and $L_D \sim 0.6$ μm [25], $b\sim e_{ii}L_D\sigma/\epsilon$ is of the order of 10^{-11} N. For a nematic cell with thickness $d\sim 10$ μm, and taking $K \sim 10^{-11}$ N, we calculate the critical anchoring energy: $w_{cr} \sim 10^{-6}$ J/m^2. This value of anchoring energy is rather low and therefore the condition $w\leqslant w_{cr}$ is not satisfied by conventional homeotropic anchoring treatments. In homeotropic nematic cells, anchoring energy of the order of 10^{-5} J/m^2 has been reported [26]. A further reduction of the anchoring energy with the temperature is obtained especially in the vicinity of the clearing point [27], but in this case the growth of an isotropic wetting film [28] should prevent the observation of the above predicted texture instability. Nevertheless, surface treatments giving rise to very weak anchoring begin to be reported [13,14] and the predicted instability could be soon tested.

As discussed above, at the lowest order in ε, the periodic instability is characterized by a tilt angle which is z independent. Of course, this property does not exist any longer if θ is expanded to the first order in ε. In this case, the solution of Eq. (15) with boundary conditions $\theta_{1,\eta}(\xi\pm 1/2)=0$, and such as $\theta_1(\xi+\Lambda,\eta)=\theta_1(\xi,\eta)$, is

$$\theta_1(\xi,\eta)=[a\sinh(2q\eta)+b\cosh(2q\eta)+H(\eta)]\sin(2q\xi), \tag{24}$$

where $H(\eta)$ is the particular solution of the ordinary differential equation

$$H_{,\eta\eta}(\eta)-(2q)^2H(\eta)=(1/2)U(\eta), \tag{25}$$

and the constants a and b are given by

$$a=-\frac{H_{,\eta}(1/2)+H_{,\eta}(-1/2)}{4q\cosh q},$$

$$b=-\frac{H_{,\eta}(1/2)-H_{,\eta}(-1/2)}{4q\sinh q}. \tag{26}$$

According to theory of the potential (Poisson's equation) [29], the function $H(\eta)$ is given by

$$H(\eta)=\frac{1}{4\pi}\int_{-1/2}^{1/2}\int_0^{\infty}\cos(2q\zeta)U(\eta')\times\ln[\zeta^2+(\eta'-\eta)^2]d\zeta d\eta', \tag{27}$$

which can be easily determined once $U(\eta)$ is defined.

At the first order in ε, the tilt angle $\theta(\xi,\eta)=\theta_0(\xi)+\varepsilon\theta_1(\xi,\eta)$ is now η dependent. By operating as before, it is possible to determine the new critical anchoring energy to observe the modulated structure.

V. CONCLUSION

We have shown that the presence of surface fields can destroy the homogeneous nematic alignment when the anchoring energy is weak enough. The instability should cause a periodic 2D splay-bend deformation in a nematic slab with homeotropic boundary conditions. The instability is activated only if (a) the anchoring energy becomes lower than a critical anchoring energy evaluated of the order of 10^{-6} J/m^2, and (b) the two substrates are not identical. Note that the anchoring critical value is rather low suggesting special surface treatment in order to observe such an instability. The instability mechanism acts, in absence of any external bulk field (magnetic or electric) or mechanical field, and gives rise to a stripe modulation. We obtained the nematic director tilt angle profile by means of a perturbation method, in which the expansion parameter is proportional to the anchoring energy strength. At the lowest order, the tilt angle is z independent. The wavelength of the modulated structure is proportional to the thickness of the nematic slab.

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[22] If we have a sample of width L, and the period of the structure is Λ, for $L \gg \Lambda$, we can put $L = m\Lambda$, where m is an integer. In this case, the total energy G_T of the nematic sample under consideration is $G_T = Gm = (G/\Lambda)L = \phi L$. It follows that the profile minimizing the total energy of the sample coincides with the one minimizing ϕ.

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