Modulated structures of flexoelectric origin in nematic liquid crystals

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A structural instability of flexoelectric origin is predicted in a homeotropic cell, of insulating nematic liquid crystal, by the action of an electric field applied in the direction of the initially nonperturbed nematic director. The instability gives rise to a two-dimensional periodic structure. The critical field to observe the predicted modulated structure as well as the wavelength at the threshold are evaluated. Both vary as the inverse square root of the cell thickness. The role of the dielectric anisotropy on the phenomenon is investigated. Our analysis is performed in the limit of weak anchoring energy strength, where the extrapolation length is large with respect to the thickness of the nematic sample.

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I. INTRODUCTION

Liquid crystals are roughly cigar shaped organic molecules with anisotropic properties. In the nematic phase, molecules are, on an average, parallel to a mean direction called the nematic director $\mathbf{n}$ ($\mathbf{n}^2=1$). Therefore, the nematic phase possesses long range orientational order and behaves as a uniaxial crystal with its optical axis parallel to $\mathbf{n}$ [1]. Uniformly oriented nematic textures possess inversion symmetry and consequently are not ferroelectric. As shown by Meyer [2], the inversion symmetry can break by imposing a splay or bend curvature distortion to the nematic director. Therefore, distorted nematic liquid crystals can present an electric polarization, termed flexoelectric polarization: the nematic equivalent of the piezoelectric polarization observed in strained solids.

The Cartesian components of the flexoelectric polarization are given by $P_i=f_{ijk}n_{j,k}$, where f_{ijk} are the components of the flexoelectric tensor and $n_{i,j}=\partial n_i/\partial x_j$ are the spatial derivatives of the nematic director. In the bulk, nematic liquid crystals are nonpolar media: the states $\mathbf{n}$ and $-\mathbf{n}$ are equivalent. This implies that f_{ijk} is odd in $\mathbf{n}$. By decomposing f_{ijk} in the usual manner [3], one obtains

$$f_{ijk}=f_1 n_i n_j n_k+f_2 n_i \delta_{jk}+f_3 n_j \delta_{ik}+f_4 n_k \delta_{ij}. \qquad (1)$$

Since $\mathbf{n}$ is such that $n_j n_j=1$, the parameters f_1 and f_3 do not contribute to P_i. A simple calculation gives $P_i=f_2 n_i n_{j,j}+f_4 n_k n_{i,k}$ [4]. By taking into account that $n_{j,j}=\boldsymbol{\nabla}\cdot\mathbf{n}$ and that $n_k n_{i,k}=-[\mathbf{n}\times(\boldsymbol{\nabla}\times\mathbf{n})]_i$, the flexoelectric polarization can be rewritten as $\mathbf{P}=f_2\mathbf{n}(\boldsymbol{\nabla}\cdot\mathbf{n})-f_4\mathbf{n}\times(\boldsymbol{\nabla}\times\mathbf{n})$. The phenomenological parameters f_2 and f_4 are usually indicated by e_{11} and e_{33}, respectively. Finally, the flexoelectric polarization is written as

$$\mathbf{P}=e_{11}\mathbf{n}(\boldsymbol{\nabla}\cdot\mathbf{n})-e_{33}\mathbf{n}\times(\boldsymbol{\nabla}\times\mathbf{n}). \qquad (2)$$

e_{11} and e_{33} are known as flexoelectric coefficients.

Long ago it has been shown that in *unbounded* nematic samples, the coupling of the flexoelectric polarization with an external electric field can give rise to a two-dimensional periodic deformation [2,5,6]. For *bounded* nematic samples, the instability appears above a threshold field when the dielectric anisotropy is lower than an upper limit [9]. Up to now, only the case of infinitely *strong anchoring* conditions on the limiting surfaces of the nematic sample has been considered [9]. In the advent of surface treatments giving rise to weak anchoring [10], it becomes appealing to extend the previous studies towards the weak anchoring limit. The aim of the present paper is to analyze, theoretically, the two-dimensional periodic deformations of flexoelectric origin, in bounded nematic samples, characterized by *weak anchoring* energy. In our analysis, the anisotropic part of the surface tension relevant to the interface, nematic liquid crystal–solid substrate, is considered very weak and treated as a perturbation. In this framework, we show that if $e_{11}+e_{33}=0$, planar periodic deformations should be observed in nematic samples, in the low voltage range. We further assume that the nematic liquid crystal is a perfect insulating material. In this case, no ionic charges are present in the medium and Debye's screening length is infinite [7]. In real nematic materials, the conductivity is finite and the ions play an important role in the effective electric field distribution inside the sample. Our model works well only if Debye's screening length is larger than the thickness of the sample. In the opposite case, in which Debye's screening length is comparable with the thickness of the sample, it is necessary to take into account that the electric field is no longer constant across the sample, and that the charge density of flexoelectric origin is partially screened by the ionic charge density [8].

In Sec. II, we first consider the case of a dielectrically compensated nematic liquid crystal. The threshold electric field to induce the instability, which drives the system from the homogeneous state to a modulated one, and the wavelength of the modulation at the threshold are deduced in the limit of weak anchoring. The influence of the dielectric anisotropy on the modulated structure is discussed in Sec. III. Section IV is devoted to final remarks and conclusions.

II. MODULATED STRUCTURES IN COMPENSATED NEMATIC LIQUID CRYSTALS

We consider a nematic liquid crystal cell in the shape of a slab of thickness d. The Cartesian reference frame used in

our analysis has the z axis, of unit vector $\mathbf{k}$, normal to the limiting surfaces, at $z=\pm d/2$. We limit the study to two-dimensional deformations, where the nematic director is fully characterized by the tilt angle θ formed by $\mathbf{n}$ with the z axis. The bulk free energy density of a nematic liquid crystal submitted to an external electric field $\mathbf{E}=E\mathbf{k}$ is [9]

$$f=\tfrac{1}{2}K(\theta_{,x}^2+\theta_{,z}^2)-eE\theta_{,x}, \tag{3}$$

where $\theta_{,x}=\partial\theta/\partial x$ and $\theta_{,z}=\partial\theta/\partial z$. Expression (3) has been written by assuming $K_{11}=K_{33}=K$, $\epsilon_a=\epsilon_\parallel-\epsilon_\perp=0$, where $\parallel$ and $\perp$ refer to $\mathbf{n}$, and $e_{11}=-e_{33}=e$. This means that we are working in the one-elastic constant approximation and the nematic liquid crystal is assumed to be dielectrically compensated. The influence of the dielectric anisotropy on the phenomenon will be considered in the following section. We assume that the limiting surfaces induce homeotropic alignment. In the Rapini-Papoular approximation [11], the relevant surface anchoring energy is written as

$$f_{si}=-\tfrac{1}{2}w_i\cos^2\theta_i, \tag{4}$$

where $i=u,l$ refer to the upper and lower surfaces, at $z=\pm d/2$, respectively. Expression (4) holds only if the surface polarization can be neglected, as we assume in the following. This assumption implies that the two extremities of the molecules forming the nematic phase have the same chemical affinity with the bounding surfaces [12,13].

We look for a tilt angle distribution $\theta(x,z)$ minimizing the total energy of the sample of the type

$$\theta(x+\lambda,z)=\theta(x,z)+2\pi, \tag{5}$$

where λ is the wavelength of the modulated structure. The total free energy of one period, per unit length along the y axis, is given by

$$F=\int_0^\lambda\int_{-d/2}^{d/2}f\,dx\,dz+\int_0^\lambda f_{sl}dx+\int_0^\lambda f_{su}\,dx. \tag{6}$$

We define $G=F/K$, introduce the reduced coordinates $\xi=x/d$ and $\eta=z/d$, and the reduced wavelength $\Lambda=\lambda/d$, and write Eq. (6) in the form

$$G=\int_0^\Lambda\int_{-1/2}^{1/2}g\,d\xi\,d\eta+\int_0^\Lambda g_{sl}\,d\xi+\int_0^\Lambda g_{su}\,d\xi, \tag{7}$$

where

$$g=\frac{fd^2}{K}=\frac{1}{2}(\theta_{,\xi}^2+\theta_{,\eta}^2)-\frac{eEd}{K}\theta_{,\xi} \tag{8}$$

and

$$g_{si}=\frac{f_{si}d}{K}=\frac{d}{2L_i}\cos^2\theta_i. \tag{9}$$

$L_i=K/w_i$ are the extrapolation lengths [14]. The function θ, we are looking for, is the one minimizing the average energy per period defined as $\phi=G/\Lambda$, where G is given by Eq. (7) [15]. Standard calculations [16] give the differential equation

$$\theta_{,\xi\xi}+\theta_{,\eta\eta}=0, \tag{10}$$

that has to be solved with the boundary conditions

$$\theta_{,\eta}-\frac{d}{2L_l}\sin(2\theta_l)=0,$$

$$\theta_{,\eta}+\frac{d}{2L_u}\sin(2\theta_u)=0. \tag{11}$$

In the following, we limit our investigation to the symmetric case, in which $w_l=w_u$. In this framework, $\theta(\xi,\eta)=\theta(\xi,-\eta)$ and the boundary conditions reduce to

$$\theta_{,\eta}+\frac{\varepsilon}{2}\sin(2\theta)=0, \tag{12}$$

where $\varepsilon=d/L=dw/K$, at $\eta=1/2$.

We assume $\varepsilon\ll1$ and expand θ in power series of ε as follows [16]:

$$\theta(\xi,\eta)=\theta_0(\xi,\eta)+\varepsilon\theta_1(\xi,\eta)+O(\varepsilon^2). \tag{13}$$

By substituting expansion (13) into Eqs. (10) and (12) we obtain at the zeroth order in ε

$$\theta_{0,\xi\xi}+\theta_{0,\eta\eta}=0,$$

$$\theta_{0,\eta}=0 \quad \text{for} \quad \eta=1/2, \tag{14}$$

and

$$\theta_{1,\xi\xi}+\theta_{1,\eta\eta}=0,$$

$$\theta_{1,\eta}+\tfrac{1}{2}\sin(2\theta_0)=0 \quad \text{for} \quad \eta=1/2, \tag{15}$$

at the first order in ε. We look for a solution of the problem of the type $\theta(\xi+\Lambda,\eta)=\theta(\xi,\eta)+2\pi$. Consequently, $\theta_0(\xi+\Lambda,\eta)=\theta_0(\xi,\eta)+2\pi$ and $\theta_i(\xi+\Lambda,\eta)=\theta_i(\xi,\eta)$, for $i\geqslant1$. The solutions of Eqs. (14) and (15) that satisfy these requirements are

$$\theta_0=m\xi \tag{16}$$

and

$$\theta_1=-\frac{1}{4m}\frac{\cosh(2m\eta)}{\sinh m}\sin(2m\xi), \tag{17}$$

where $m=2\pi/\Lambda$ is a constant to be determined by imposing that the total energy of the system has its minimum value. By substituting the θ expansion in Eq. (8), we obtain $g=g_0+\varepsilon g_1+O(\varepsilon^2)$, where

$$g_0=\frac{1}{2}(\theta_{0,\xi}^2+\theta_{0,\eta}^2)-\frac{eEd}{K}\theta_{0,\xi} \tag{18}$$

and

$$g_1=\theta_{0,\xi}\theta_{1,\xi}+\theta_{0,\eta}\theta_{1,\eta}-\frac{eEd}{K}\theta_{1,\xi}. \tag{19}$$

Since $\theta_0=\theta_0(\xi)=m\xi$, g_0 and g_1 take the forms

$$g_0=\frac{1}{2}m^2-\frac{eEd}{K}m \tag{20}$$

and

$$g_1=\left(m-\frac{eEd}{K}\right)\theta_{1,\xi}. \tag{21}$$

The function $\theta_1(\xi,\eta)$ is such that $\theta_1(\xi+\Lambda,\eta)=\theta_1(\xi,\eta)$. Consequently, g_1 does not contribute to the total energy G at the first order in ε. For what concerns the surface contributions we have, at the first order in ε,

$$g_{sl}=g_{su}=-\frac{\varepsilon}{2}\cos^2(m\xi). \tag{22}$$

The total energy G, at the first order in the expansion parameter, is then $G=G_0+\varepsilon G_1+O(\varepsilon^2)$, where

$$G_0=\left(\frac{1}{2}m^2-\frac{eEd}{K}m\right)\Lambda \tag{23}$$

and

$$G_1=-\int_0^\Lambda \cos^2(m\xi)d\xi=-\tfrac{1}{2}\Lambda. \tag{24}$$

Finally, the average energy per period $\phi=G/\Lambda$ is

$$\phi=\frac{1}{2}m^2-\frac{eEd}{K}m-\frac{1}{2}\varepsilon. \tag{25}$$

m is determined by imposing that, at the zeroth order in ε, ϕ is minimum. In this case the minimum of ϕ, at the zeroth order in ε, coincides with the one at the first order in the same parameter. A simple calculation gives

$$m=\frac{eEd}{K}. \tag{26}$$

For the modulated structure under consideration, the minimum value of G is then

$$G_{MS}=-\frac{1}{2}\left[\left(\frac{eEd}{K}\right)^2+\varepsilon\right]\Lambda. \tag{27}$$

The G energy of the corresponding homogeneous pattern ($\theta=0$ everywhere) is

$$G_H=-\varepsilon\Lambda. \tag{28}$$

The modulated structure is stable only if $G_{MS}<G_H$. This condition gives

$$\left(\frac{eEd}{K}\right)^2>\varepsilon. \tag{29}$$

The latter inequality defines a threshold field $E_{th}(0)$,

$$E>E_{th}(0)=\frac{K}{ed}\sqrt{\varepsilon}=\sqrt{\frac{Kw}{e^2d}}, \tag{30}$$

to observe the modulated structure in a compensated nematic liquid crystal. The relevant threshold voltage for the phenomenon under consideration is then

$$V_{th}(0)=E_{th}(0)d=\sqrt{\frac{Kwd}{e^2}}. \tag{31}$$

At the threshold, since $\varepsilon=d/L$, the wavelength of the modulated structure is

$$\lambda_{th}(0)=2\pi\frac{d}{\sqrt{\varepsilon}}=2\pi\sqrt{Ld}. \tag{32}$$

According to the above analysis, $V_{th}(0)$ and $\lambda_{th}(0)$ for a compensated nematic liquid crystal depend on the thickness of the sample as $\sqrt{d}$.

The expressions for $E_{th}(0)$ and $\lambda_{th}(0)$ are valid only if $\varepsilon\ll 1$, and they give the order of magnitude of the critical field and of the wavelength at the threshold of the instability towards the modulated structure. By assuming $K\sim 10^{-11}$ N [14], $e\sim 10^{-11}$C m [17], $d\sim 1\ \mu$m, $\varepsilon\sim 0.1$, which corresponds to $L\sim 10\ \mu$m, and hence $w\sim 10^{-6}$ J/m^2, we obtain $V_{th}(0)=E_{th}(0)d\sim 0.3$ V, a rather low value. Note that the corresponding threshold in the case of strong anchoring is two orders of magnitude higher. From this result, we can conclude that planar periodic deformations in nematic samples weakly anchored induced by a low dc electric voltage are a clear indication that $e_{11}+e_{33}=0$. Furthermore, any dependence of the threshold voltage on the sample thickness should be a signature of departure from strong anchoring conditions. Figure 1 shows the nematic director distribution in a x-z cross section of the cell at an electric field value of $1.5E_{th}(0)$. The initially homeotropic cell is distorted under the field action and gives rise to planar domains. The **n** dependence on z appears in the transition region between homeotropic and planar domains and it is merely concentrated close to the glass plates. Further, the **n** distribution shows that the transition starts at the middle of the cell.

III. INFLUENCE OF THE DIELECTRIC ANISOTROPY ON THE MODULATED STRUCTURE OF FLEXOELECTRIC ORIGIN

Let us consider now the case $\epsilon_a=\epsilon_\parallel-\epsilon_\perp\neq 0$. In this framework, the free energy density f is given by

$$f=\tfrac{1}{2}K(\theta_{,x}^2+\theta_{,z}^2)-eE\theta_{,x}-\tfrac{1}{2}\epsilon_aE^2\cos^2\theta. \tag{33}$$

We consider first the case of positive dielectric anisotropy ($\epsilon_a>0$). By operating step by step as in the case of $\epsilon_a=0$, we obtain now the free energy density g as

$$g=\frac{fd^2}{K}=\frac{1}{2}(\theta_{,\xi}^2+\theta_{,\eta}^2)-\frac{eEd}{K}\theta_{,\xi}-\frac{1}{2}\Omega^2\cos^2\theta, \tag{34}$$

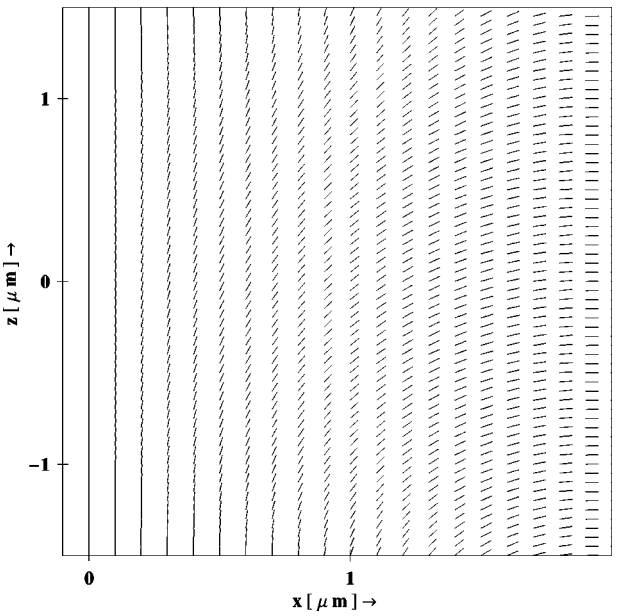

FIG. 1. The **n** distribution in a x-z cross section of the nematic cell at $1.5E_{th}(0)$. $d=3\ \mu$m, $\varepsilon=0.3$. In the absence of electric field, the nematic cell is homeotropic with **n** parallel to the z axis.

where

$$\Omega^2=\frac{\epsilon_a E^2 d^2}{K}=\left(\pi\frac{E}{E_c}\right)^2. \tag{35}$$

$E_c=(\pi/d)\sqrt{K/\epsilon_a}$ is the critical field for the Freedericksz transition [14]. The bulk differential equation obtained by minimizing the total energy G is

$$\theta_{,\xi\xi}+\theta_{,\eta\eta}=\frac{\Omega^2}{2}\sin(2\theta), \tag{36}$$

which has to be solved with the boundary condition (12), in the symmetric case under consideration, where $\theta(\xi,\eta)=\theta(\xi,-\eta)$. By substituting expansion (13) into Eq. (36), we obtain at the zeroth order in ε,

$$\theta_{0,\xi\xi}+\theta_{0,\eta\eta}=\frac{\Omega^2}{2}\sin(2\theta_0),$$

$$\theta_{0,\eta}=0 \quad \text{for} \quad \eta=1/2, \tag{37}$$

and

$$\theta_{1,\xi\xi}+\theta_{1,\eta\eta}=\Omega^2\theta_1\cos(2\theta_0),$$

$$\theta_{1,\eta}+\tfrac{1}{2}\sin(2\theta_0)=0 \quad \text{for} \quad \eta=1/2, \tag{38}$$

at the first order in ε.

The solution of Eq. (37) is η independent. It is given by the nonlinear ordinary differential equation

$$\frac{d^2\theta_0}{d\xi^2}-\frac{\Omega^2}{2}\sin(2\theta_0)=0. \tag{39}$$

From Eq. (39), beside the trivial solution $\theta_0=0$, we obtain

$$\frac{d\theta_0}{d\xi}=\sqrt{c^2-\Omega^2\cos^2\theta_0}, \tag{40}$$

where $c^2>\Omega^2$ is an integration constant to be determined by imposing that the total energy of the sample is minimum. The wavelength of the modulated structure we are looking for is

$$\Lambda=\int_0^{2\pi}\frac{d\psi}{\sqrt{c^2-\Omega^2\cos^2\psi}}. \tag{41}$$

Routine calculations give, for the G energy at the zeroth order in ε, the expression

$$G_0=\int_0^{2\pi}\sqrt{c^2-\Omega^2\cos^2\psi}\,d\psi-\frac{1}{2}c^2\Lambda-2\pi\frac{eEd}{K}, \tag{42}$$

from which the average G_0 energy per period $\phi_0=G_0/\Lambda$ is easily obtained. By minimizing ϕ_0 with respect to c^2, we get the equation

$$\int_0^{2\pi}\sqrt{c^2-\Omega^2\cos^2\psi}\,d\psi=2\pi\frac{eEd}{K}, \tag{43}$$

which defines c^2.

As it follows from Eq. (41), $\Lambda\to\infty$ for $c^2\to\Omega^2$. In the limit $c^2=\Omega^2$, from Eq. (43) we obtain $c=(\pi/2)(eEd/K)$. The condition $c^2>\Omega^2$ is then a condition on the dielectric anisotropy of the type [9]

$$\epsilon_a<\epsilon_a^*=\left(\frac{\pi}{2}\right)^2\frac{e^2}{K}. \tag{44}$$

Otherwise, the homogeneous nondistorted state is more favored. By means of the elliptical functions $E_1(k)$ and $E_2(k)$ defined by

$$E_1(k)=\int_0^{\pi/2}\frac{d\psi}{\sqrt{1-k^2\cos^2\psi}},$$

$$E_2(k)=\int_0^{\pi/2}\sqrt{1-k^2\cos^2\psi}\,d\psi, \tag{45}$$

it is possible to rewrite Eq. (41) and (43) as follows:

$$\Lambda=\frac{1}{\Omega}4kE_1(k) \tag{46}$$

and

$$\frac{1}{k}E_2(k)=\sqrt{\frac{\epsilon_a^*}{\epsilon_a}}, \tag{47}$$

respectively, where $k^2=\Omega^2/c^2$ is the modulus of the elliptical functions. Equation (47) determines $k=k(\epsilon_a/\epsilon_a^*)$, when the physical parameters of the nematic liquid crystal are

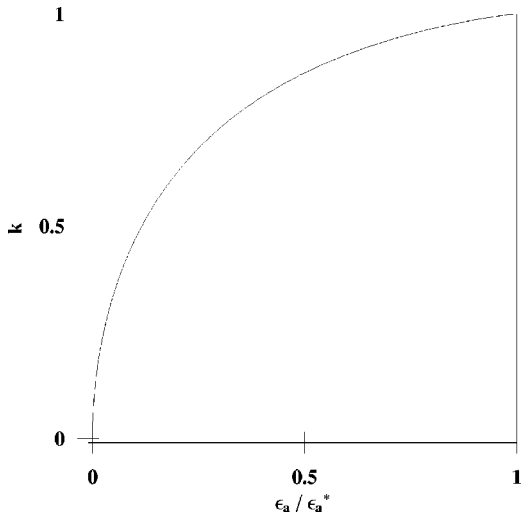

FIG. 2. Behavior of k vs ϵ_a/ϵ_a^*.

known. As it follows from Eqs. (45) and (47), in the limit $\epsilon_a \to 0$, $k \to (\pi/2)\sqrt{\epsilon_a/\epsilon_a^*}$. In Fig. 2, we show k vs ϵ_a/ϵ_a^*. Once k has been determined, Eq. (46) gives Λ vs $\Omega = \pi(E/E_c)$. In Fig. 3, the product $\Lambda\Omega$ vs ϵ_a/ϵ_a^* is reported.

The tilt angle profile, at the zeroth order in ε, is given by

$$k\int_0^{\theta_0(\xi)} \frac{d\psi}{\sqrt{1-k^2\cos^2\psi}} = \Omega\xi, \tag{48}$$

and it is plotted in Fig. 4, for $k=0.1$ and $k=0.95$. As it is evident from this figure, $\theta_0(\xi)$ is, practically, a linear function of the reduced coordinate ξ, for all values of k. At the zeroth order in ε, we have that the minimum value of the G energy, relevant to the modulated structure is, as it follows from Eqs. (42) and (43),

$$G_{MS0} = -\tfrac{1}{2}c^2\Lambda. \tag{49}$$

At the same approximation, the G energy of the homogeneous orientation, with $\theta=0$, is

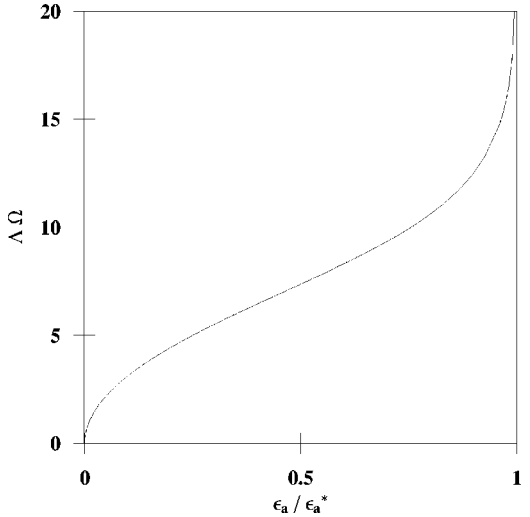

FIG. 3. $\Omega\Lambda$ vs ϵ_a/ϵ_a^*.

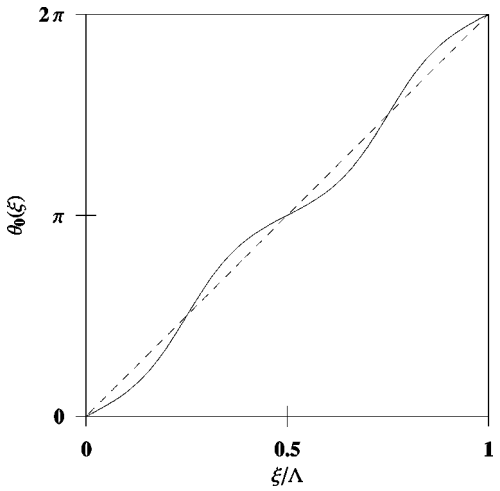

FIG. 4. Tilt angle $\theta_0(\xi)$, at the zeroth order in the expansion parameter, vs the reduced coordinate ξ/Λ, for $k=0.1$ (dashed line), and $k=0.95$ (solid line), which correspond to $\epsilon_a/\epsilon_a^*=0.004$ and $\epsilon_a/\epsilon_a^*=0.742$, respectively. Note that $\theta_0(\xi)$ is, practically, a linear function of ξ for all values of k.

$$G_{H0} = -\tfrac{1}{2}\Omega^2\Lambda. \tag{50}$$

Since $c \geqslant \Omega$, it follows that $G_{MS0} \leqslant G_{H0}$, i.e., at the zeroth order in the expansion parameter, the modulated structure is stable with respect to the homogeneous one.

Let us consider now the energy G at the first order in the expansion parameter. By substituting Eq. (13) into Eq. (34) and taking into account that $\theta_0 = \theta_0(\xi)$ we obtain $g = g_0 + \varepsilon g_1$, where

$$g_0 = \frac{1}{2}\left(\frac{d\theta_0}{d\xi}\right)^2 - \frac{eEd}{K}\frac{d\theta_0}{d\xi},$$

$$g_1 = \frac{d\theta_0}{d\xi}\frac{\partial\theta_1}{\partial\xi} + \frac{1}{2}\Omega^2\theta_1\sin(2\theta_0) - \frac{eEd}{K}\frac{\partial\theta_1}{\partial\xi}. \tag{51}$$

It follows that $G = G_0 + \varepsilon G_1$, where

$$G_0 = \int_0^\Lambda \int_{-1/2}^{1/2} g_0 d\xi d\eta,$$

$$G_1 = \int_0^\Lambda \int_{-1/2}^{1/2} g_1 d\xi d\eta - \int_0^\Lambda \cos^2\theta_0 \, d\xi. \tag{52}$$

A simple calculation allows one to show that

$$\int_0^\Lambda \int_{-1/2}^{1/2} g_1 d\xi d\eta = 0. \tag{53}$$

To this end, we have to just note that

$$\frac{d\theta_0}{d\xi}\frac{\partial\theta_1}{\partial\xi} = \frac{\partial}{\partial\xi}\left(\theta_1\frac{d\theta_0}{d\xi}\right) - \theta_1\frac{d^2\theta_0}{d\xi^2}. \tag{54}$$

By substituting the latter expression into g_1, Eq. (51) yields

$$g_1=\frac{\partial}{\partial\xi}\left(\theta_1\frac{d\theta_0}{d\xi}\right)-\theta_1\left(\frac{d^2\theta_0}{d\xi^2}-\frac{1}{2}\Omega^2\sin^2(2\theta_0)\right)-\frac{eEd}{K}\frac{\partial\theta_1}{\partial\xi}. \tag{55}$$

The second addendum of this expression is identically zero because $\theta_0(\xi)$ is a solution of Eq. (39). Consequently, g_1 can be written as

$$g_1=\frac{\partial}{\partial\xi}\left[\theta_1\left(\frac{d\theta_0}{d\xi}-\frac{eEd}{K}\right)\right]. \tag{56}$$

Since $\theta_1(\xi+\Lambda,\eta)=\theta_1(\xi,\eta)$ and $(d\theta_0/d\xi)(\xi+\Lambda,\eta)=(d\theta_0/d\xi)(\xi,\eta)$, the integral of g_1 over one spatial period vanishes identically. Therefore, G_1 reduces to the contribution connected with the anchoring energy. At the first order in ε and taking into account Eq. (49), the energy G of the modulated structure is

$$G_{MS}=-\frac{1}{2}c^2\Lambda-\varepsilon\int_0^\Lambda\cos^2\theta_0(\xi)d\xi, \tag{57}$$

while the G energy of the homogeneous pattern is

$$G_H=-\tfrac{1}{2}\Omega^2\Lambda-\varepsilon\Lambda. \tag{58}$$

The modulated structure is stable only if $G_{MS}<G_H$. This condition yields

$$c^2-\Omega^2>2\varepsilon\langle\sin^2\theta_0\rangle, \tag{59}$$

where

$$\langle\cdots\rangle=\frac{1}{\Lambda}\int_0^\Lambda\cdots d\xi. \tag{60}$$

Equation (59) defines the threshold field above which the modulated structure is stable, for a given anchoring energy $w=(K/d)\varepsilon$. As it has been underlined above, $\theta_0(\xi)$ is, practically, a linear function of ξ, for all k. Consequently $\langle\sin^2\theta_0\rangle\sim1/2$. In this framework, from the definition $k=\Omega/c$ and using Eq. (59), we obtain the value of Ω at the threshold:

$$\Omega_{th}=\sqrt{\frac{k^2}{1-k^2}\varepsilon}, \tag{61}$$

from which the threshold field is calculated,

$$E_{th}=\frac{2}{\pi}\sqrt{\frac{\epsilon_a^*}{\epsilon_a}\frac{k^2}{1-k^2}}E_{th}(0). \tag{62}$$

$E_{th}(0)$ has been defined in Eq. (30). Since, as discussed above, $k\to(\pi/2)\sqrt{\epsilon_a/\epsilon_a^*}$ when $\epsilon_a\to0$ then E_{th} reduces to $E_{th}(0)$: the threshold field for a compensated nematic liquid crystal given by Eq. (30).

From Eq. (46), it follows that the reduced wavelength at the threshold is

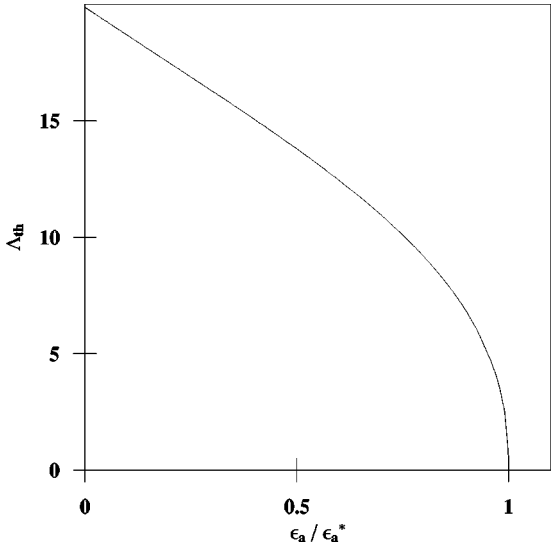

FIG. 5. Reduced wavelength at the instability threshold Λ_{th} vs ϵ_a/ϵ_a^*, for a nematic homeotropic cell with $\varepsilon=d/L=0.1$.

$$\Lambda_{th}=4\sqrt{\frac{1-k^2}{\varepsilon}}E_1(k)=\frac{2}{\pi}\sqrt{1-k^2}E_1(k)\Lambda_{th}(0). \tag{63}$$

In the limit $\epsilon_a\to0$, the latter equation gives $\Lambda_{th}=2\pi/\sqrt{\varepsilon}$, as obtained in the case of a compensated nematic. The dependence of Λ_{th} on the ratio ϵ_a/ϵ_a^* is shown in Fig. 5 for a nematic cell with $\varepsilon=d/L=0.1$. The transition between the homogenous state and the stripe pattern state is a second-order transition and close to $\epsilon_a=\epsilon_a^*$, Λ_{th} varies as $(1-\epsilon_a/\epsilon_a^*)^{1/2}$. Note that when $k\to1$, the elliptic integral $E_1(k)$ diverges logarithmically. Figure 6 shows $E_{th}/E_{th}(0)$ vs ϵ_a/ϵ_a^*, which could be useful to compare our theoretical predictions with experimental data. However, no experimental data are available in literature, for the $E_{th}=E_{th}(\epsilon_a/\epsilon_a^*)$ dependence, relevant to periodic instabilities of flexoelectric origin in nematic liquid crystals. The first attempt to relate

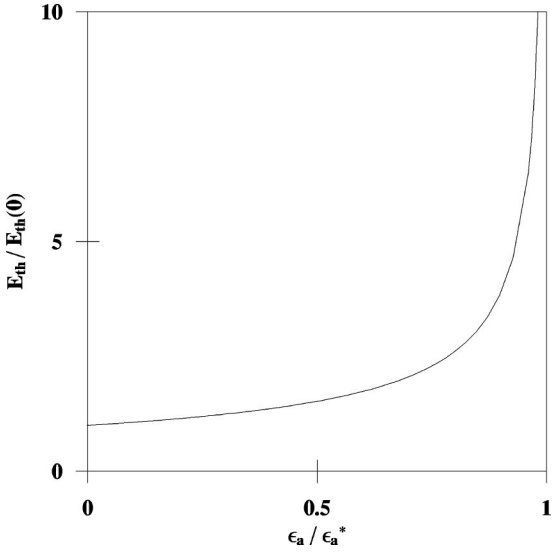

FIG. 6. Reduced threshold field $E_{th}/E_{th}(0)$ vs ϵ_a/ϵ_a^*.

the dielectric anisotropy with the threshold field for the flexoelectric instability has been performed by Barnik *et al.* long ago [18,19]. Unfortunately, for the compounds investigated in Refs. [18,19], the flexoelectric coefficients have not been precisely determined. Consequently, ϵ_a^* are unknown and we cannot fit the threshold voltage dependence on the dielectric anisotropy using our results. Moreover, the nematic cells used by Barnik *et al.* have strong planar anchoring, i.e., $d>L$, and therefore their results cannot be used to test our model. However, as it follows from Eq. (48), the modulus k depends only on ϵ_a/ϵ_a^*. Hence, from Eqs. (62) and (63) we derive that the threshold voltage and the wavelength of the deformation at the threshold depend on the thickness of the sample as $\sqrt{d}$, as in the case of a compensated nematic liquid crystal. These dependencies could allow to test, experimentally, our model.

In the analysis reported above we have assumed $\epsilon_a>0$. In the opposite case, of negative dielectric anisotropy, the dielectric contribution to the g energy density given by Eq. (34) is $+(1/2)\Omega^2\cos^2\theta$, where $\Omega^2=|\epsilon_a|E^2d^2/K=(\pi E/E_c)^2$ and $E_c=(\pi/d)\sqrt{K/|\epsilon_a|}$. In this case on the right-hand side of Eqs. (36)–(38) Ω^2 is changed to $-\Omega^2$, and Eq. (39) takes the form

$$\frac{d^2\theta_0}{d\xi^2}+\frac{\Omega^2}{2}\sin(2\theta_0)=0. \tag{64}$$

By operating as in the case of $\epsilon_a>0$, instead of Eq. (40) we obtain now

$$\frac{d\theta_0}{d\xi}=\sqrt{C^2-\Omega^2\sin^2\theta_0}, \tag{65}$$

from which we derive that $C^2>\Omega^2$. In the present case, condition (44) becomes

$$|\epsilon_a|<\epsilon_a^*=\left(\frac{\pi}{2}\right)^2\frac{e^2}{K}. \tag{66}$$

The main equations of the problem are identical to the ones reported above with $E_1(k)$ and $E_2(k)$ changed to

$$\mathcal{E}_1(\kappa)=\int_0^{\pi/2}\frac{d\psi}{\sqrt{1-\kappa^2\sin^2\psi}},$$

$$\mathcal{E}_2(\kappa)=\int_0^{\pi/2}\sqrt{1-\kappa^2\sin^2\psi}\,d\psi, \tag{67}$$

where $\kappa^2=\Omega^2/C^2$.

IV. FINAL REMARKS AND CONCLUSION

To observe experimentally the predicted instability, special cautions have to be taken. In fact, as underlined above, our analysis works well only if the nematic liquid crystal can be considered as a perfect insulating material. In the opposite case, when ions are present in the liquid crystal, it is necessary to take into account that the electric field is no longer constant across the sample, due to the double layer formation of ionic clouds close to the electrodes [20]. The presence of the ions is equivalent to a renormalization of the effective bulk electric field and of the effective anchoring energy [21–23]. To observe our periodic instability, it is necessary to avoid selective ions adsorption and to use a pulsed square electric field with a frequency such that the ions cannot participate in the phenomenon.

In conclusion, we have theoretically analyzed the possibility to observe modulated structures of flexoelectric origin in bounded nematic liquid crystal samples. Our investigation has been performed by assuming that $e_{11}+e_{33}=0$, and that the anchoring energy w of the solid substrate–nematic liquid crystal interface is very weak and can be treated as a perturbation. In this limit, the tilt angle profile, formed by the director with the normal to the limiting surfaces, can be expanded in power series of $\varepsilon=d/L$, where $L=K/w$ is the extrapolation length. We have determined the threshold field for the spatially modulated instability, and the wavelength at the threshold. The threshold field varies as $\sqrt{1/d}$, while the wavelength at the threshold goes as $\sqrt{d}$. The influence of the dielectric anisotropy on the phenomenon has also been analyzed. The instability can arise only if the dielectric anisotropy is lower than an upper limit defined from the material constants of the liquid crystal compound. In the case of an unbounded nematic liquid crystal sample, our results confirm the prediction published long ago [2,5,6].

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