Spontaneous periodic distortions in nematic liquid crystals: Dependence on the tilt angle

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The possibility of spontaneous periodic distortions, depending on the tilt angle in a nematic liquid crystal sample, is investigated by means of a general formulation of the stability problem. It is shown that due to the presence of a surfacelike term in the free-energy density, the uniform pattern can be destabilized, giving rise to a periodic distortion of the director. Our analysis establishes, in general terms, the conditions for the formation of stable periodic structures in nematic samples. In particular, we determine the wavelengths for which the periodic distortion exists by investigating its dependence on the tilt angle, characterizing the uniform pattern, and on the saddle-splay elastic constant. The effect considered in our paper is a finite size effect, related to the slab geometry of the nematic sample under consideration.

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I. INTRODUCTION

Bulk elastic properties of nematic liquid crystals (NLC) are well described by means of the so-called Frank elasticity. In this framework, the elastic energy density of NLC is given by [1–3]

$$f_e=\frac{1}{2}\{K_{11}(\boldsymbol{\nabla}\cdot\mathbf{n})^2+K_{22}[\mathbf{n}\cdot(\boldsymbol{\nabla}\times\mathbf{n})]^2+K_{33}[\mathbf{n}\times(\boldsymbol{\nabla}\times\mathbf{n})]^2\} -(K_{22}+K_{24})\boldsymbol{\nabla}\cdot[\mathbf{n}\boldsymbol{\nabla}\cdot\mathbf{n}+\mathbf{n}\times(\boldsymbol{\nabla}\times\mathbf{n})]. \quad (1)$$

In Eq. (1), the director $\mathbf{n}$ is a unit vector representing the average molecular orientation of the nematic phase. The coefficients $K_{ii}>0$, with $i=1,2,3$ are, respectively, the well-known bulk elastic constants of splay, twist, and bend. Furthermore, K_{24} is the saddle-splay elastic constant, and the term associated to it plays the role of a surfacelike term. In fact, the last term, by means of the Gauss theorem, gives only a surface contribution [3]. If this term is neglected, the elastic energy density becomes a positive definite quadratic form in the distortions. In this case, in the absence of any other external influence, the ground state of an NLC is expected to be a uniform one [4].

The existence of periodic distortions in nematic liquid crystals, in the presence of external fields, has been investigated by several authors [1,5–12]. Recently, Pergamenshchick [13,14] considered the possibility of a spontaneous appearance of periodic distortions in planar samples, induced by surfacelike terms. He showed that if the elastic constant of saddle splay, K_{24}, is large enough, the ground state of a nematic sample characterized by planar easy axes on both surfaces could be periodically distorted. Very recently, the same problem was reconsidered by means of a more general stability analysis, considering the investigation of the positivity of the quadratic form representing the total elastic energy of the system in this context [15]. It was shown that the uniform planar profile can be unstable against fluctuations, giving rise to periodic structures in the medium.

In this paper, we establish the general conditions for the formation of periodic structures in NLC, in the absence of external fields. Our analysis is not limited to the planar case, and explicitly takes into account the role of the tilt angle in the formation (or not) of stable periodic structures in NLC. The effect considered in our paper is a finite size effect, related to the slab geometry of the nematic sample under consideration.

II. THEORETICAL BACKGROUND

We consider a sample in the shape of a slab, whose thickness is d, such that the x_3 axis of a Cartesian reference frame is normal to the bounding surfaces, placed at $x_3=0$ and $x_3=d$. The uniform state is assumed to be $\mathbf{n}_0=n_{01}\mathbf{e}_1+n_{03}\mathbf{e}_3$, where $\mathbf{e}_i$ are the usual unit vectors. The director of the distorted pattern is $\mathbf{n}=\mathbf{n}_0+\delta\mathbf{n}$, where $\delta\mathbf{n}=\mathbf{u}(x_2,x_3)$ represents the fluctuations around the nondeformed state $\mathbf{n}_0$. Since $|\mathbf{n}|=1$, at the first order in $\delta\mathbf{n}$, $\mathbf{n}_0\cdot\delta\mathbf{n}=0$, and hence $u_1=-(n_{03}/n_{01})u_3$. Consequently, the bulk elastic energy density, Eq. (1), can be written in terms of the spatial derivatives $u_{\alpha,\beta}=\partial u_\alpha/\partial x_\beta$ as

$$f=\frac{1}{2}K_{11}(u_{2,2}+u_{3,3})^2+\frac{1}{2}\frac{K_{22}}{n_{01}^2}(u_{3,2}^2+n_{01}^4u_{2,3}^2-2n_{01}^2u_{3,2}u_{2,3}) +\frac{1}{2}K_{33}\left(\frac{n_{03}}{n_{01}}\right)^2(n_{01}^2u_{2,3}^2+u_{3,3}^2)-2(K_{22}+K_{24}) \times(u_{2,2}u_{3,3}-u_{2,3}u_{3,2}). \quad (2)$$

The surface energy is assumed to be in the form

$$f_S(u_i)=\frac{1}{2}(w_1u_1^2+w_2u_2^2+w_3u_3^2) =\frac{1}{2}\left[\left(\frac{n_{03}^2w_1+n_{01}^2w_3}{n_{01}^2}\right)u_3^2+w_2u_2^2\right], \quad (3)$$

where w_i $(i=1,2,3)$ are the anchoring strengths, and the above relation connecting u_1 and u_3 has been used.

The average total energy, per unit length along the x_1 axis, is given, in general, by

$$F=\frac{1}{\lambda}\left\{\int_0^{\lambda}\int_0^{d} f(u_{i,j})dx_2dx_3+\int_0^{\lambda} f_S[u_2(0),u_3(0)]dx_2 + \int_0^{\lambda} f_S[u_2(d),u_3(d)]dx_2\right\}, \tag{4}$$

where $\alpha,\beta=2,3$, and $\lambda=2\pi/q$ is the wavelength of the periodic deformation whose stability is being analyzed. The actual director orientation is the one minimizing F given by Eq. (4). The usual minimization techniques yield

$$\frac{\partial f}{\partial u_{\alpha}}-\sum_{\beta}\ \partial_{\beta}\frac{\partial f}{\partial u_{\alpha,\beta}}=0 \tag{5}$$

for $0\leqslant x_3\leqslant d$ and $0\leqslant x_2\leqslant\lambda$, where $\alpha=2$ and 3, and $\beta=1,2,3$. The boundary conditions to be satisfied by the solutions of the bulk differential equations (5) are

$$\mp\frac{\partial f}{\partial u_{2,3}}+\frac{\partial f_S}{\partial u_2}=0 \quad\text{and}\quad \mp\frac{\partial f}{\partial u_{3,3}}+\frac{\partial f_S}{\partial u_3}=0 \tag{6}$$

at $x_3=0$ $(-)$ and $x_3=d$ $(+)$. The other boundary conditions at x_2 and $x_2+\lambda$ are automatically satisfied, since we are searching for periodic deformations along y, i.e., if $\mathbf{n}(x_2,x_3)=\mathbf{n}(x_2+\lambda,x_3)$, then $\delta\mathbf{n}(x_2,x_3)=\delta\mathbf{n}(x_2+\lambda,x_3)$. In the present case, the differential equations (5) and boundary conditions (6) are

$$K_{11}u_{2,22}+(K_{22}n_{01}^2+K_{33}n_{03}^2)u_{2,33}+(K_{11}-K_{22})u_{3,32}=0,$$

$$K_{22}u_{3,22}+(K_{11}n_{01}^2+K_{33}n_{03}^2)u_{3,33}+n_{01}^2(K_{11}-K_{22})u_{2,23}=0; \tag{7}$$

and

$$[(K_{22}n_{01}^2+K_{33}n_{03}^2)u_{2,3}+(K_{22}+2K_{24})u_{3,2}]\mp w_2u_2=0,$$

$$(K_{11}n_{01}^2+K_{33}n_{03}^2)u_{3,3}+n_{01}^2[K_{11}-2(K_{22}+K_{24})]\times u_{2,2}\mp(n_{03}^2w_1+n_{01}^2w_3)u_3=0, \tag{8}$$

respectively. The bulk differential equations (7) with the boundary conditions (8) have always the trivial solution $u_2=u_3=0$, which corresponds to the uniform alignment along $\mathbf{n}_0$. Our aim is to determine under what conditions this configuration does not correspond to a stable state, and to show that a periodic deformation, of a well-defined wave vector, can appear in the sample.

III. LINEAR ANALYSIS OF THE STABILITY

For a linear analysis around the nondeformed state, the periodic solutions of the bulk differential equations are chosen of the form $u_2(x_2,x_3)=g(x_3)\sin(qx_2)$ and $u_3(x_2,x_3)=f(x_3)\cos(qx_2)$. In this case, $g(x_3)$ and $f(x_3)$ are solutions of two coupled differential equations of second order, and $u_2(x_2,x_3)$ and $u_3(x_2,x_3)$ can be expressed in terms of four independent integration constants C_i. This means that, in the linearized case, the solutions of Eqs. (5) may be put in the form

$$u_2(x_2,x_3)=u_2(C_i;x_2,x_3),$$

$$u_3(x_2,x_3)=u_3(C_i;x_2,x_3) \tag{9}$$

for $i=1,2,3,4$. The four integration constants are determined by the boundary conditions (6). These conditions form a linear and homogeneous system. The total energy F, given by Eq. (4), is a quadratic form of these integration constants because, in this linearized analysis, $u_2(x_2,x_3)$ and $u_3(x_2,x_3)$ depend linearly on C_i. To know if the nondeformed state is stable, it is necessary to analyze the sign of the quadratic form representing F, which is symmetric. In other words, by substituting Eqs. (9) in Eq. (4) we obtain for F an expression of the kind $F=1/2\Sigma_{i,j}M_{ij}C_iC_j$, where $M_{ij}=M_{ji}$, because the asymmetric part of the matrix $\mathcal{M}$, of elements M_{ij}, does not contribute to F. The quantities C_i are obtained by minimizing F with respect to C_i, i.e., by imposing that $\partial F/\partial C_i=0$. This gives $\Sigma_jM_{ij}C_j=0$, which is equivalent to the boundary conditions (6). Notice, however, that the knowledge of the matrix $\mathcal{M}$ allows a simpler investigation of the stable state. The nondeformed state $C_i=0$, for $i=1,2,3$, and 4, corresponds to a minimum of F if all four determinants of the principal minors of the matrix $\mathcal{M}$, $m_1=M_{11}$, $m_2=M_{11}M_{22}-M_{12}^2$, and so on, are positive. On the contrary, the knowledge of the system obtained by Eqs. (6) does not allow to conclude anything about the stability of the nondeformed state.

The elements of the matrix $\mathcal{M}$ can be easily obtained by substituting solutions (9) in Eq. (4), which permits us to transform F in an ordinary function of the integration constants C_i, in the form $F=F(C_i)$. Once this task is accomplished, one obtains

$$\frac{\partial F}{\partial C_i}=\frac{1}{\lambda}\int_0^{\lambda}\left\{\sum_{j=2,3}\left[\left(-\frac{\partial f}{\partial u_{j,3}}+\frac{\partial f_S}{\partial u_j}\right)\frac{\partial u_j}{\partial C_i}\right]_{z=0} + \sum_{j=2,3}\left[\left(\frac{\partial f}{\partial u_{j,3}}+\frac{\partial f_S}{\partial u_j}\right)\frac{\partial u_j}{\partial C_i}\right]_{z=d}\right\}dx_2. \tag{10}$$

From Eq. (10) it follows that $\partial F/\partial C_i$ is obtained by means of a linear combination of the boundary conditions (6). In order to explicitly obtain the elements of matrix $\mathcal{M}$, we introduce the quantities X_i $(i=1,2,3,4)$ defined as

$$u_3(0)=X_1\cos(qx_2),\quad u_2(0)=X_2\sin(qx_2),$$

$$u_3(d)=X_3\cos(qx_2),\quad u_2(d)=X_4\sin(qx_2), \tag{11}$$

which are linear combinations of C_i in the form $X_k=\Sigma_kB_{kl}C_l$. It is also useful to introduce the quantities V_i $(i=1,2,3,4)$ in the form

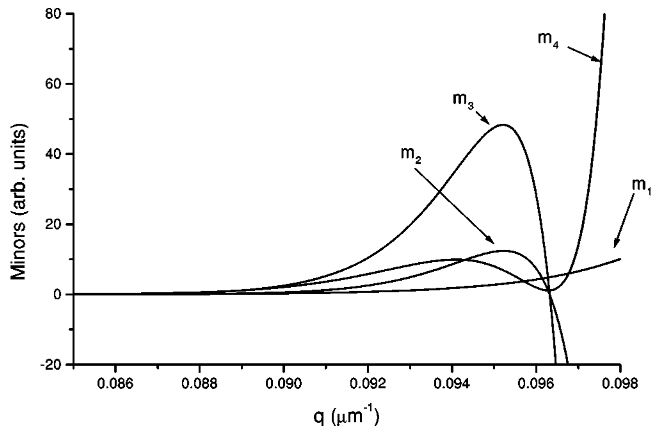

FIG. 1. Behavior of $m_i(q)$ for $i=1$–4, $\nu=0.5$, $\mu=3.0$ ($K_{24}=K_{22}/2$), $w_1=w_3\neq 0$, $w_2=0$, and $n_{03}=0.5$. The vertical axis is in arbitrary units.

$$\left(-\frac{\partial f}{\partial u_{3,3}}+\frac{\partial f_S}{\partial u_3}\right)_{x_3=0}=V_1\cos(qx_2),$$

$$\left(-\frac{\partial f}{\partial u_{2,3}}+\frac{\partial f_S}{\partial u_2}\right)_{x_3=0}=V_2\sin(qx_2),$$

$$\left(\frac{\partial f}{\partial u_{3,3}}+\frac{\partial f_S}{\partial u_3}\right)_{x_3=d}=V_3\cos(qx_2),$$

$$\left(\frac{\partial f}{\partial u_{2,3}}+\frac{\partial f_S}{\partial u_2}\right)_{x_3=d}=V_4\sin(qx_2), \qquad (12)$$

which are also linear combinations of C_i, given in the form $V_k=\Sigma_m A_{km}C_m$. Substitution of Eqs. (11) and (12) in Eq. (10) yields

$$\frac{\partial F}{\partial C_i}=\frac{1}{2}\sum_k V_k\frac{\partial X_k}{\partial C_i}=\frac{1}{2}\sum_{k,m} B_{ik}A_{km}C_m. \qquad (13)$$

By using the condition $\partial F/\partial C_i=0$, one deduces that $M_{ij}=(1/2)\Sigma_k B_{ik}A_{kj}$. This completes the formalism to fully analyze the stability of the nondeformed ground state in a nematic liquid crystal sample. The extension of the formalism for the case in which an external field is applied to the system presents no difficulty.

IV. RESULTS OF THE NUMERICAL ANALYSIS

To explore some of the immediate consequences of the results previously presented, we particularize our analysis to the case in which $w_1=w_3$ and $w_2=0$ (no azimuthal anchoring energy). We first remember that in the case of a planar uniform state ($n_{01}=1$ and $n_{03}=0$), there exists a critical thickness d_c, given by $\det\mathcal{M}=0$, such that for $d<d_c$ the homogeneous pattern is unstable. In this case, for $q\rightarrow 0$, $m_4(q)=\alpha(d-d_c)q^2$, where $\alpha>0$. The trend of $m_4(q)$ vs q depends on the sign of $d-d_c$. In particular, for $d\gg d_c$, m_4

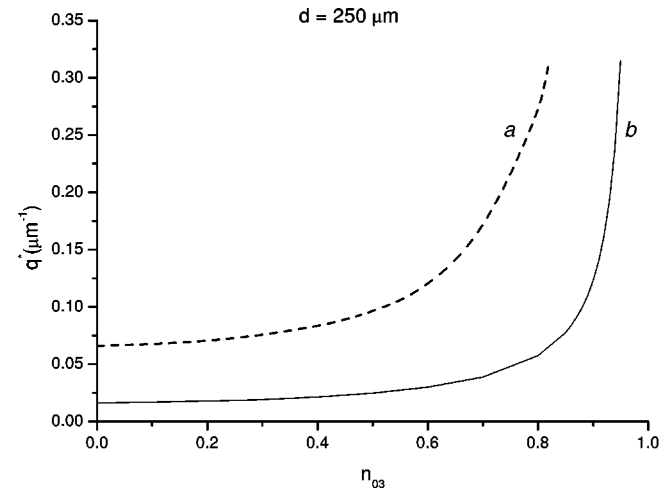

FIG. 2. Behavior of q^* as a function of the z component of the director (n_{03}) for $w_1=w_3$ with no azimuthal anchoring $w_2=0$. Curve *a* corresponds to $\nu=0.5$, $\mu=3.0$ ($K_{24}=K_{22}/2$); and curve *b* to $\nu=0.5$, $\mu=5.0$ ($K_{24}=3\,K_{22}/2$). Periodic instabilities are not favored for $n_{03}>0.82$ in curve *a* and for $n_{03}>0.95$ in curve *b*. q^* corresponds to the points for which $m_4(q)$, after presenting a positive maximum, is zero in correspondence to a vanishing value of $m_3(q)$, as is illustrated in Fig. 1.

vanishes for a well defined q^*. For $q<q^*$, all the determinants of the principal minors of the matrix $\mathcal{M}$ are positive. On the contrary, for $q>q^*$, $m_2<0$ as well as $m_3<0$. These results permit the determination of the values of the wave vector q^* for which the planar orientation is unstable [15]. The general analysis, in which $n_{03}=\sqrt{1-n_{01}^2}\neq 0$, is more complex. However, it is simple to deduce that for $q\rightarrow 0$, $m_4(q)\propto q^{-10}$. This indicates that there is no longer a critical thickness for which periodic instabilities appear in the system in the limit of $q\rightarrow 0$. However, periodic instabilities can, in principle, be present in the system depending strongly on the value of the ratios $\nu=K_{11}/K_{22}$ and $\mu=2(1$

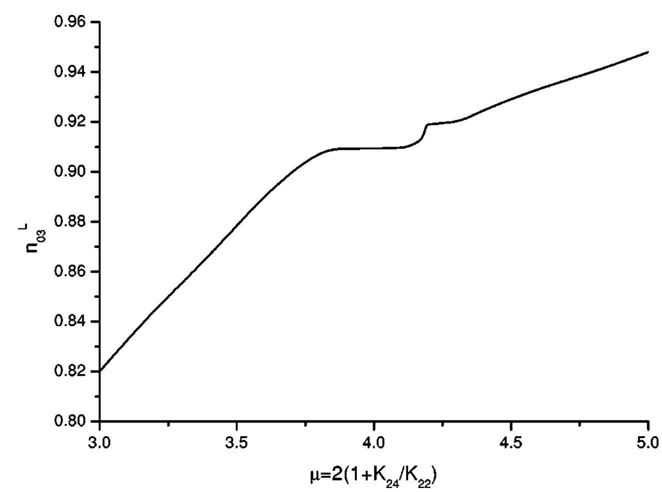

FIG. 3. n_{03}^L vs μ for $\nu=0.5$ and $d=250$ μm. n_{03}^L represents the value of the z component of the nematic director above which stable periodic distortions are not allowed in the system.

$+K_{24}/K_{22})$, and on the value of the tilt angle of the uniform state. This conclusion follows from the behavior of $m_4(q)$ that, for arbitrary values of d, presents a positive maximum for $q\neq 0$ and vanishes for $q=q^*$, according to the value of n_{03}, as it is illustrated in Fig. 1. This value corresponds to the wave vectors for which periodic instabilities appear in the system. Our linear analysis does not allow us to determine the profile of the favored instabilities; it indicates, however, their arising in the system. In Fig. 2, the behavior of q^* is shown as a function of n_{03} for curve a ($\nu=0.5$ and $\mu=3.0$) and curve b ($\nu=0.5$ and $\mu=5.0$). The dependence of q^* vs n_{03} is such that for $n_{03}\to n_{03}^L$, $q^*\to\infty$. For $n_{03}>n_{03}^L$, periodic deformations are forbidden. In particular, for $n_{03}\to 1$, i.e., for uniform state near the homeotropic configuration, periodic instabilities are forbidden. This is expected because for small fluctuations around the homeotropic configuration, the term connected with K_{24} is of third order, whereas the usual bulk terms are of second order in the variation of the director. In this case, it plays a minor role in destabilizing the uniform pattern. Notice, however, that for curve b which refers to higher value of K_{24}, periodic distortions may exist also for values of n_{03} near to 1. This means that the surfacelike term in the elastic energy density becomes very important and dictates the behavior of the system. Of course, for $n_{03}\to 1$ we again conclude that the homeotropic ground state is favored against periodic deformations of the director.

In Fig. 3, a plot of n_{03}^L vs μ is shown. According to our analysis, n_{03}^L is a decreasing function of μ, presenting a plateau in the vicinity of $K_{24}\simeq K_{22}$.

V. CONCLUSION

We have presented a general formalism to investigate the possibility of the formation of periodic instabilities in a nematic sample. Our analysis is not limited to the planar case, already considered, and is applied to the general case in which the nondeformed pattern can change continuously from the planar to the homeotropic one. It can be applied to analyze the behavior of the system with or without external field (the extension for this latter case is immediate). Therefore, it permits us to investigate, in a complete and conclusive way, the dependence on the tilt angle of the nondeformed state in the formation of periodic instabilities. Our analysis is based on the investigation of the positivity of the quadratic form representing the total elastic energy of the sample. It indicates that (1) the role of the elastic constant of saddle splay is dominant in destabilizing the uniform pattern; (2) different from the planar uniform case (i.e., $n_{01}=1$ and $n_{03}=0$), there is no critical thickness below which the periodic instabilities are favored in the system.

According to our calculations, periodic deformations, connected with the saddle-splay elastic constant, can be observed only in samples presenting an initial tilt. If the nematic sample is homeotropically oriented, to observe a periodic structure it is necessary to have a surface transition inducing a tilt.

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