

Thermal-fluctuation approach to Fréedericksz transitions in nematic liquid crystals

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The boundary effects on the thermal fluctuations of the director in a planar nematic-liquid-crystal cell with strong anchoring conditions in the presence of an electric field, coupled to the dielectric anisotropy and to flexoelectric polarization, are theoretically studied. The conditions for second-order Fréedericksz-type transitions and the actual shape of the distortion above the threshold are analyzed by considering the critical eigenmodes of the fluctuations. A phase diagram for the different types of distortions, periodic or aperiodic, is obtained through a fully analytic calculation. These results allow for a better interpretation of the light-scattering experiments and give a unified picture of the instabilities, induced by the flexoelectricity and by the elastic constant anisotropy, which give static periodic structures.

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I. INTRODUCTION

A very important characteristic of nematic liquid crystals is their strong light scattering due to the thermal fluctuations of the director $\hat{\mathbf{n}}$, that gives the local average alignment of the molecules [1].

Light-scattering experiments are a powerful tool for the verification of any theory of nematic order and for the measurement of many physical parameters, such as, for example, the curvature elastic constants and the viscosities of thermotropic [2] and lyotropic [3] liquid crystals.

The theory developed in Ref. [1] refers to an infinite medium and therefore can be applied only to thick enough samples. In fact, generally, the different Fourier components of the fluctuations become coupled if the boundary conditions are taken into account. This fact has been pointed out by various authors, considering some particular cases [4,5]. Here we give a more general approach to this problem by making use of the formalism developed in Ref. [4]. In particular the effects due to an external electric field and to the flexoelectricity are taken into account.

The mean director orientation is assumed to be uniform: therefore our analysis cannot be applied for values of the external field corresponding to distorted structures. The distortion appears above a critical value of the field, and it corresponds to a breaking of some symmetry element of the structure. The thermal fluctuations are the physical mechanism responsible for this symmetry breaking. The analysis given here allows us therefore to tackle in a very natural way the important problem of the transition from an undistorted to a distorted configuration.

Three types of transitions will be considered: the classical Fréedericksz transition that gives rise to a distorted

aperiodic structure, and the transitions towards periodic static structures related to the flexoelectric effect [6–8] and to the elastic anisotropy [9], respectively.

A unitary treatment of these three types of transitions, based on the analysis of the thermal fluctuations, gives a better understanding of the problem and a deeper insight into the physics of these phenomena.

In Sec. II we introduce the general theory; in Sec. III we discuss some approximated solutions, while in Sec. IV the exact solutions are considered. In Sec. V we separately analyze the effects of the flexoelectric coupling and of the elastic anisotropy. In Sec. VI we analytically derive a phase diagram for the different kinds of Fréedericksz-type transitions. Finally in Sec. VII we summarize our results.

II. THEORY

We consider a planar nematic-liquid-crystal (NLC) cell occupying the region $-d/2 \leq z \leq d/2$, with strong anchoring conditions and with the undistorted director $\hat{\mathbf{n}}_0$ parallel to the x axis, in the presence of a uniform electric field E directed along z . In a linearized theory, the fluctuations give a distortion field $\delta\mathbf{n}$ such that $\delta\mathbf{n} \cdot \hat{\mathbf{n}}_0 = 0$; therefore

$$\delta\mathbf{n}(\mathbf{r}) = \Theta_1(\mathbf{r})\hat{\mathbf{z}} + \Theta_2(\mathbf{r})\hat{\mathbf{y}}, \quad (2.1)$$

where $\hat{\mathbf{y}}$ and $\hat{\mathbf{z}}$ are the unit vectors parallel to the y and z axis, respectively, and

$$\Theta_1(x, y, z = \pm d/2) = \Theta_2(x, y, z = \pm d/2) = 0. \quad (2.2)$$

Taking into account the dielectric and the flexoelectric coupling to the external electric field, the free energy of the distorted configuration can be written as

$$\mathcal{F} = \mathcal{F}_0 + \mathcal{F}_1, \quad (2.3a)$$

$$\mathcal{F}_0 = \frac{1}{2} \int \{ K_1(\Theta_{1,z} + \Theta_{2,y})^2 + K_2(\Theta_{1,y} - \Theta_{2,z})^2 + K_3(\Theta_{1,x}^2 + \Theta_{2,x}^2) - \epsilon_0 \epsilon_a E^2 \Theta_1^2 - 2E[e_1 \Theta_1(\Theta_{1,z} + \Theta_{2,y}) + e_3(\Theta_{1,x} + \Theta_2 \Theta_{1,y} + \Theta_1 \Theta_{1,z})] \} d\mathbf{r}, \quad (2.3b)$$

where K_1 , K_2 , K_3 are the splay, twist, and bend elastic constants, respectively; ϵ_0 is the free-space permittivity; ϵ_a is the dielectric anisotropy, equal to the difference $\epsilon_{\parallel} - \epsilon_{\perp}$ between the dielectric constant parallel and perpendicular to the director, respectively; e_1 and e_3 are the flexoelectric coefficients; $\Theta_{i,j}$ ($i = 1, 2$; $j = x, y, z$) indicates the partial derivative of Θ_i with respect to j ; E is an average electric field, directed along z ; finally \mathcal{F}_1 is the contribution to the free energy coming from the gradients of the electric field that are generated by the polarization charges induced by the distortion.

The quantity \mathcal{F}_1 , which is zero in the absence of dielectric anisotropy, is usually neglected in the literature when small distortions are considered, such as, for instance, when the threshold field for periodic Fréedericksz transitions is evaluated [7,9,10]. However, this term is generally of the same order of magnitude as the others, and therefore it can give relevant effects. In Appendix A it is shown, with the help of a perturbation expansion of the electric field in terms of the distortions, that for fluctuation modes that are homogeneous in the x direction, \mathcal{F}_1 is of order higher than two in the distortions, and thus it can set to zero in our linear analysis. In the following we will concentrate our attention on such x -independent fluctuations, since the critical modes giving rise to all the known Fréedericksz-like transitions are of this type. It is therefore convenient to first consider only the contribution \mathcal{F}_0 to the free energy. The given analysis and the conclusions drawn will be exact in the particular case of fluctuations homogeneous in the x direction.

The effect of \mathcal{F}_1 on the other fluctuations will be briefly discussed at the end of this section.

With the help of the strong anchoring conditions (2.2) and imposing periodic boundary conditions along the transverse directions for $x = \pm L/2$ and $y = \pm L/2$, where the limit $L \rightarrow \infty$ is to be taken at the end, an integration by parts of \mathcal{F}_0 yields

$$\mathcal{F}_0 = \frac{1}{2} \int \Theta^t \mathcal{L} \Theta \, d\mathbf{r}, \quad (2.4)$$

where

$$\Theta = \begin{pmatrix} \Theta_1 \\ \Theta_2 \end{pmatrix}, \quad (2.5)$$

Θ^t is the transpose of Θ , and \mathcal{L} is the 2×2 linear matrix operator whose elements \mathcal{L}_{ij} ($i, j = 1, 2$) are given by

$$\mathcal{L}_{11} = -K_3 \frac{\partial^2}{\partial x^2} - K_2 \frac{\partial^2}{\partial y^2} - K_1 \frac{\partial^2}{\partial z^2} - \epsilon_0 \epsilon_a E^2, \quad (2.6a)$$

$$\mathcal{L}_{12} = -(K_1 - K_2) \frac{\partial^2}{\partial y \partial z} - (e_1 - e_3) E \frac{\partial}{\partial y}, \quad (2.6b)$$

$$\mathcal{L}_{21} = -(K_1 - K_2) \frac{\partial^2}{\partial y \partial z} + (e_1 - e_3) E \frac{\partial}{\partial y}, \quad (2.6c)$$

$$\mathcal{L}_{22} = -K_3 \frac{\partial^2}{\partial x^2} - K_1 \frac{\partial^2}{\partial y^2} - K_2 \frac{\partial^2}{\partial z^2}. \quad (2.6d)$$

Thanks to the boundary conditions that we have imposed, it is easily shown that \mathcal{L} is self-adjoint: this means that it is possible to build an orthonormal complete set of eigenfunctions, or normal modes, Θ_{α} of the operator \mathcal{L} ,

$$\mathcal{L} \Theta_{\alpha} = \Lambda_{\alpha} \Theta_{\alpha}, \quad \int \Theta_{\alpha}^t \Theta_{\beta} \, d\mathbf{r} = \delta_{\alpha\beta}, \quad (2.7)$$

where Λ_{α} is a real number and $\delta_{\alpha\beta}$ is the Kronecker delta.

At any given time t , the fluctuation $\Theta(\mathbf{r}, t)$ can then be decomposed as a weighted sum of eigenfunctions

$$\Theta(\mathbf{r}, t) = \sum_{\alpha} c_{\alpha}(t) \Theta_{\alpha}(\mathbf{r}); \quad (2.8)$$

inserting this expression in (2.4) and using Eqs. (2.7), the corresponding free energy reads

$$\mathcal{F}_0 = \frac{1}{2} \sum_{\alpha} c_{\alpha}^2 \Lambda_{\alpha}. \quad (2.9)$$

From the equipartition theorem, the thermal average of the square of the amplitude of each normal mode Θ_{α} is then given by

$$\langle c_{\alpha}^2 \rangle = \frac{K_B T}{\Lambda_{\alpha}}, \quad (2.10)$$

where K_B is Boltzmann's constant and T is the absolute temperature.

Therefore the eigenvalue Λ_{α} determines the amplitude of the fluctuation Θ_{α} ; it also determines its decay constant γ_{α} , which is given by $\Lambda_{\alpha}/\eta_{\alpha}$, where η_{α} is a suitable viscosity coefficient. The Fréedericksz-type second-order transitions are obtained by considering the E dependence of Λ_{α} ; the first eigenfunction whose eigenvalue Λ becomes equal to zero is the critical mode: it is characterized by a huge increase of its amplitude and a critical slowing down.

The symmetries of the operator \mathcal{L} suggest looking for eigenfunctions of the type

$$\Theta_1 = \cos(Q_x x) \cos(Q_y y) \theta_1(z), \quad (2.11a)$$

$$\Theta_2 = \cos(Q_x x) \sin(Q_y y) \theta_2(z). \quad (2.11b)$$

Fully equivalent solutions with $\sin(Q_x x)$ instead of $\cos(Q_x x)$ and with $\cos(Q_y y)$ and $\sin(Q_y y)$ interchanged are also allowed. By inserting these solutions into the eigenvalue equation $\mathcal{L} \Theta = \Lambda \Theta$ one obtains

$$\begin{pmatrix} -\Lambda + K_3 Q_x^2 + K_2 Q_y^2 - K_1 \frac{d^2}{dz^2} - \epsilon_a \epsilon_0 E^2 & -(K_1 - K_2) Q_y \frac{d}{dz} - (e_1 - e_3) E Q_y \\ (K_1 - K_2) Q_y \frac{d}{dz} - (e_1 - e_3) E Q_y & -\Lambda + K_3 Q_x^2 + K_1 Q_y^2 - K_2 \frac{d^2}{dz^2} \end{pmatrix} \begin{pmatrix} \theta_1 \\ \theta_2 \end{pmatrix} = 0. \quad (2.12)$$

For $Q_y = 0$ the matrix operator in (2.12) is diagonal and its eigenfunctions are identical to the solutions valid for an unbounded medium, with the only difference that the Q_z component of the wave vector \mathbf{Q} is now quantized. Two independent sets of solutions are obtained that correspond to splay-bend and twist-bend distortions, respectively. These fluctuation modes can be separately detected in light-scattering experiments with polarized light: in fact, most experiments have been performed under this condition and are very easily interpreted since the different Fourier components of the fluctuations are uncoupled.

The terms containing the component Q_x appear in such a way that the solutions (2.11a) and (2.11b) corresponding to the set of parameters $Q_x = 0$, $\Lambda = \Lambda_0$ and $Q_x \neq 0$, $\Lambda = \Lambda_0 + K_3 Q_x^2$ are exactly the same. The most important implication of this fact is that the first zero of Λ , which gives rise to a second-order Fréedericksz-type transition, is expected to occur at $Q_x = 0$, since Λ monotonically increases by increasing Q_x . This property gives a theoretical explanation to the fact that the static periodic structures of this type that have been observed are characterized by $Q_x = 0$. However, we recall that the above analysis is based only on the contribution \mathcal{F}_0 to the total free energy and therefore is exact only for $\epsilon_a = 0$, since in this case $\mathcal{F}_1 = 0$ even for $Q_x \neq 0$. In the general case $\epsilon_a, Q_x \neq 0$, using an approach similar to that outlined in Appendix A, or alternatively taking the fluctuation in the electric potential as a third free parameter (and therefore increasing by one the dimensions of the problem), one can show that the electric field gradients give rise to an additional dielectric contribution to the right hand side of Eq. (2.6a) that is proportional to ϵ_a^2 and always increases the free energy, plus two nondiagonal flexoelectric terms in the right hand sides of Eqs. (2.6b) and (2.6c) proportional to $\epsilon_a(e_1 + e_3)$ that are nonzero for fluctuation modes that depend both on x and y . Therefore in the absence of these latter flexoelectric contributions, i.e., for $e_1 + e_3 = 0$, the Fréedericksz transition, if any, will still occur at $Q_x = 0$, while for $\epsilon_a(e_1 + e_3) \neq 0$ a thorough numerical analysis would be required in order to check whether an oblique pattern is allowed for a suitable choice of the parameters.

For the above reasons, in the following we only consider the Q_y dependence of Λ and $\theta_i(z)$ for $Q_x = 0$. The corresponding fluctuations can be detected in a light-scattering experiment with the scattering plane orthogo-

nal to the undistorted director. Such a scattering geometry has some advantages with respect to the previously quoted one [3].

III. SOLUTIONS OF THE EIGENVALUE EQUATION FOR $Q_x = 0$ AND SMALL Q_y

For $Q_x = 0$ the eigenvalue equations (2.12) are exact. Even in this simple case, such equations are rather cumbersome from both the mathematical and the physical point of view, owing to the presence of many energy terms and of the corresponding parameters. A very simple analysis of the role played by the various terms is, however, possible if we consider the limit of small Q_y . In fact the nondiagonal elements of the matrix operator appearing in Eq. (2.12) are identically zero for $Q_y = 0$. In this limiting case two independent sets Λ_{1n} and Λ_{2n} of eigenvalues with the corresponding eigenfunctions are very easily found, which constitute a good basis for a perturbation expansion of the solutions in a power series of the parameter Q_y . This gives approximate solutions, where the roles of the various terms are very simply displayed. A perturbation expansion up to second-order powers in Q_y of the eigenvalues is given in Appendix B.

In order to simplify the system of Eqs. (2.12) and point out the essential parameters of the theory, we set

$$\begin{aligned} Q_x &= 0 \quad , \quad Q_y = \frac{\pi}{d}q \quad , \quad Q_z = \frac{\pi}{d}p \quad , \\ r &= \frac{K_2}{K_1} \quad , \quad \lambda = \left(\frac{d}{\pi}\right)^2 \frac{\Lambda}{K_1} \quad , \\ v &= \frac{1}{\pi} \left(\frac{\epsilon_0}{K_1}\right)^{1/2} Ed \quad , \quad e = \frac{e_1 - e_3}{\sqrt{\epsilon_0 K_1}} \quad . \end{aligned} \quad (3.1)$$

The explicit form of Eq. (2.12) with the new symbols is given in Appendix B.

We note that only the sign of the dielectric anisotropy ϵ_a is important: in fact its absolute value can be absorbed by a suitable redefinition of the normalized applied voltage v and of the normalized flexoelectric constant e ; in the following we will concentrate our attention on the case $\epsilon_a > 0$.

In order to display the main features of the solutions, we give—up to a normalization constant—the first-order expansion in q of the first splay eigenmode

$$\begin{pmatrix} \theta_1 \\ \theta_2 \end{pmatrix} = \left(-\frac{\cos(\pi z/d)}{1 - \epsilon_a v^2 - r} q e v + \frac{(1-r)q}{\pi} \sum_{m=2,4,\dots} \frac{\cos(\pi z/d)}{1 - m^2} \frac{4m}{1 - \epsilon_a v^2 - r m^2} \frac{\sin(m\pi z/d)}{1 - m^2} \right) \quad . \quad (3.2)$$

Equation (3.2) shows that the flexoelectricity, which is represented by the term containing the parameter e , simply mixes the first splay mode with the first twist mode. The elastic anisotropy, corresponding to nonzero values of the quantity $(1-r)$, has more dramatic effects, since it mixes the first even splay mode with all the odd twist modes.

These results can be generalized as follows: the unperturbed solutions of both splay and twist type are alternatively even and odd functions of z . The flexoelectricity, in the absence of the elastic anisotropy, simply mixes the splay and twist modes with the same Fourier components and the final exact solutions are indeed very simple, as we will show in Sec. V. The elastic anisotropy, in the

absence of flexoelectricity, instead, mixes even θ_1 functions with odd θ_2 functions, and vice versa. In the final exact solutions the functions $\theta_1(z)$, $\theta_2(z)$ are therefore, respectively, even-odd or odd-even. If both the elastic anisotropy and the flexoelectricity are present, the exact solutions are very complex, without any definite symmetry, as shown in the following section. The perturbation expansions given in Appendix B will be reconsidered in Sec. VI and will be used to study the Fréedericksz-type transitions.

IV. EXACT SOLUTIONS FOR $Q_x = 0$

The solutions of the eigenvalue equations (2.12) with the boundary conditions (2.2) for any allowed eigenvalue λ are given by a linear combination of the four solutions found for an unbounded medium with the same λ value. These are

$$\begin{pmatrix} \theta_1 \\ \theta_2 \end{pmatrix} = \begin{pmatrix} \theta_{01} \\ \theta_{02} \end{pmatrix} \exp(i\pi pz/d), \quad (4.1)$$

where θ_{01} , θ_{02} , and p must satisfy the system of equations

$$\begin{pmatrix} -\lambda + rq^2 + p^2 - \epsilon_a v^2 & -i(1-r)qp - evq \\ i(1-r)qp - evq & -\lambda + q^2 + rp^2 \end{pmatrix} \times \begin{pmatrix} \theta_{01} \\ \theta_{02} \end{pmatrix} = 0. \quad (4.2)$$

Nontrivial solutions are found for

$$\det \begin{pmatrix} c_1 & c_2 & s_1 & s_2 \\ c_1 & c_2 & -s_1 & -s_2 \\ \alpha_1 c_1 - \beta_1 s_1 & \alpha_2 c_2 - \beta_2 s_2 & \alpha_1 s_1 + \beta_1 c_1 & \alpha_2 s_2 + \beta_2 c_2 \\ \alpha_1 c_1 + \beta_1 s_1 & \alpha_2 c_2 + \beta_2 s_2 & -\alpha_1 s_1 + \beta_1 c_1 & -\alpha_2 s_2 + \beta_2 c_2 \end{pmatrix} = 0, \quad (4.7)$$

or, equivalently,

$$\left(\beta_2 - \frac{\beta_1 t_1}{t_2} \right) \left(\beta_2 - \frac{\beta_1 t_2}{t_1} \right) + (\alpha_1 - \alpha_2)^2 = 0, \quad (4.8)$$

where

$$c_i = \cos\left(\frac{\pi p_i}{2}\right), \quad s_i = \sin\left(\frac{\pi p_i}{2}\right), \quad (4.9)$$

$$t_i = \tan\left(\frac{\pi p_i}{2}\right), \quad i = 1, 2.$$

Equation (4.8) must be considered as an implicit definition of the eigenvalues λ as a function of v , q and of the material parameters r , ϵ_a , e , through an equation of the form

$$g(\lambda, v, q; r, \epsilon_a, e) = 0. \quad (4.10)$$

In fact Eqs. (4.3)–(4.5d) and (4.9) define α_i , β_i , c_i , s_i , and t_i as functions of the above parameters.

Once the eigenvalue problem is solved, the relations among the unknowns a_i that define the eigenfunctions

$$(p^2 + q^2)^2 - b(p^2 + q^2) + c = 0, \quad (4.3)$$

$$\frac{\theta_{02}}{\theta_{01}} = \alpha + i\beta, \quad (4.4)$$

where

$$b = \lambda \frac{(1+r)}{r} + \epsilon_a v^2, \quad (4.5a)$$

$$c = \frac{\lambda^2 - [\epsilon_a(1-r)q^2 - \epsilon_a \lambda + e^2 q^2]v^2}{r}, \quad (4.5b)$$

$$\alpha = \frac{evq}{q^2 + rp^2 - \lambda}, \quad (4.5c)$$

$$\beta = -\frac{(1-r)qp}{q^2 + rp^2 - \lambda}. \quad (4.5d)$$

The four solutions correspond to $p = \pm p_1, \pm p_2$. A linear combination of these solutions gives the following eigenfunctions:

$$\begin{aligned} \theta_1 &= a_1 \cos \xi_1 + a_2 \cos \xi_2 + a_3 \sin \xi_1 + a_4 \sin \xi_2, \\ \theta_2 &= a_1(\alpha_1 \cos \xi_1 - \beta_1 \sin \xi_1) + a_2(\alpha_2 \cos \xi_2 - \beta_2 \sin \xi_2) \\ &\quad + a_3(\alpha_1 \sin \xi_1 + \beta_1 \cos \xi_1) \\ &\quad + a_4(\alpha_2 \sin \xi_2 + \beta_2 \cos \xi_2), \end{aligned} \quad (4.6)$$

where $\xi_1 = \pi p_1 z/d$, $\xi_2 = \pi p_2 z/d$, $\alpha_1 = \alpha(p = p_1)$, $\alpha_2 = \alpha(p = p_2)$, $\beta_1 = \beta(p = p_1)$, $\beta_2 = \beta(p = p_2)$. The four boundary conditions (2.2) give a system of four linear homogeneous equations in the unknowns a_i ($i = 1, \dots, 4$), which have nontrivial solutions only if

are easily found to be

$$\begin{aligned} \frac{a_2}{a_1} &= -\frac{c_1}{c_2}, \quad \frac{a_3}{a_1} = -\frac{1}{t_1} \frac{\alpha_1 - \alpha_2 - b_1 t_1 + b_2 t_2}{\alpha_1 - \alpha_2 + \frac{b_1}{t_1} - \frac{b_2}{t_2}}, \\ \frac{a_4}{a_3} &= -\frac{s_1}{s_2}. \end{aligned} \quad (4.11)$$

Equations (4.1)–(4.11) require some comments. Starting from the complex solutions (4.1), we have tried to make use of strictly real quantities. However, the parameters p_1 , β_1 , and a_3 can actually be purely imaginary for large intervals of the curves $\lambda_{in}(q)$ for low n values. In order to recover real expressions, it is enough to express in Eqs. (4.6) the trigonometric functions containing p_1 in terms of hyperbolic functions and to make use of the new parameters $\bar{\beta}_1 = i\beta_1$ and $\bar{a}_3 = ia_3$, which are real.

V. EIGENFUNCTIONS AND EIGENVALUES

In Secs. III and IV we have shown that, even with the simplifying condition $Q_x = 0$, the actual shape of the

eigenmodes is rather complex in a cell of finite thickness, owing to the coupling effects of the flexoelectricity and of the elastic constant anisotropy. It seems therefore interesting to consider separately the two sources of coupling.

We first consider the role of flexoelectricity, by assuming

$$e \neq 0 \quad , \quad K_1 = K_2 . \quad (5.1)$$

Equations (4.2)–(4.9) now become strictly analytic and very simple, giving

$$p_{1n} = p_{2n} = n , \quad (5.2a)$$

$$\lambda_{in} = n^2 + q^2 - \frac{1}{2}\epsilon_a v^2 \mp v \left(\frac{1}{4}\epsilon_a^2 v^2 + e^2 q^2 \right)^{1/2} , \quad (5.2b)$$

$$\begin{aligned} \alpha_{in} &= qev(n^2 + q^2 - \lambda_{in})^{-1} \\ &= qe \left[\frac{1}{2}\epsilon_a v \pm \left(\frac{1}{4}\epsilon_a^2 v^2 + e^2 q^2 \right)^{1/2} \right]^{-1} \\ &\equiv \alpha_i , \end{aligned} \quad (5.2c)$$

$$\beta_{in} = 0 , \quad (5.2d)$$

with $i = 1, 2$ [the upper and lower signs in Eqs. (5.2b) and (5.2c) correspond to $i = 1$ and $i = 2$, respectively] and $n = 1, 2, 3, \dots$

As already noticed, the flexoelectricity couples the modes with the same wave vector \mathbf{Q} but different polarizations: the eigenmodes are harmonic functions of all the coordinates. Their dependence on the coordinate y is such that by increasing y at fixed x and z the director describes an elliptical helix. The principal axes of the ellipse are along z and y , and their ratio is equal to α_{in} . The static periodic distortion that appears immediately above the threshold field for the Pikin transition [8] displays the same characteristics. Such a distortion is similar to the structure of an S_c^* liquid crystal, with the difference that here the helix is elliptical with generatrices orthogonal to the undistorted director, whereas in S_c^* it is circular with generatrices parallel to the undistorted director. Figures 1 and 2 show the dependence of the eigenvalues λ_{in} and of the ellipticity α_i on the transverse normalized wave vector q for three different values of the normalized voltage v . The minimum voltage for which the first eigenvalue $\lambda_{11}(q)$ becomes equal to zero defines the critical voltage v_c and the critical transverse wave vector q_c for a second-order periodic Fréedericksz-type transition: the periodicity of the deformed equilibrium structure at the transition threshold is given by q_c .

Let us now consider the effect of the elastic anisotropy, by assuming

$$e = 0 \quad , \quad K_1 \neq K_2 . \quad (5.3)$$

As already pointed out in Sec. III, the free-energy term depending on the anisotropy parameter $(1 - r) = (K_1 - K_2)/K_1$ gives a coupling between the odd (even) functions $\theta_1(z)$ and the even (odd) functions $\theta_2(z)$. The eigenfunctions display therefore an odd-even or even-odd symmetry for what concerns the functions $\theta_i(z)$ given by Eq. (4.6), where the parameters α_1 and α_2 are now identically equal to zero. The characteristic equation (4.8)

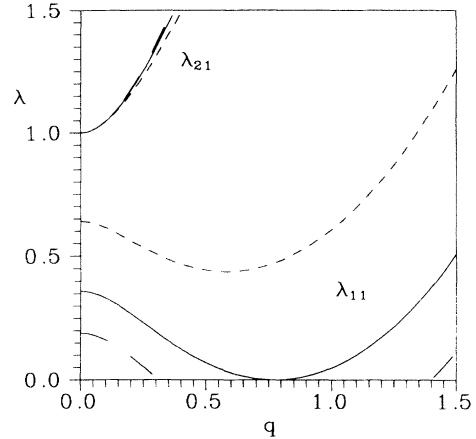


FIG. 1. Behavior of the first normalized eigenvalues λ [see Eq. (5.2b)] as a function of the normalized transverse wave vector q for $r = 1$ and $e^2/\epsilon_a = 4$. The eigenfunction associated to λ_{11} (λ_{21}) is a pure splay (twist) deformation for $q = 0$. The solid lines correspond to the critical voltage $\epsilon_a^{1/2}v = 0.8$, the short-dashed lines to $\epsilon_a^{1/2}v = 0.6$, the long-dashed lines to $\epsilon_a^{1/2}v = 0.9$.

splits in the two separate equations

$$t_1\beta_1 = t_2\beta_2 \quad \text{for even } \theta_1 \text{ and odd } \theta_2 , \quad (5.4a)$$

$$t_1\beta_2 = t_2\beta_1 \quad \text{for odd } \theta_1 \text{ and even } \theta_2 . \quad (5.4b)$$

The dependence of Θ_1 and Θ_2 on the other coordinates is such that the director describes an elliptical helix if we increase y at fixed x and z . However, the shape of the ellipse is here strongly dependent on the z coordinate.

For the most general case, corresponding to $e \neq 0$ and $K_1 \neq K_2$, except for the absence of the even-odd symmetry, no new features are found.

VI. FRÉDERICKSZ-TYPE TRANSITIONS

The free energy \mathcal{F} associated with any eigenmode and the corresponding eigenvalue are positive definite at zero

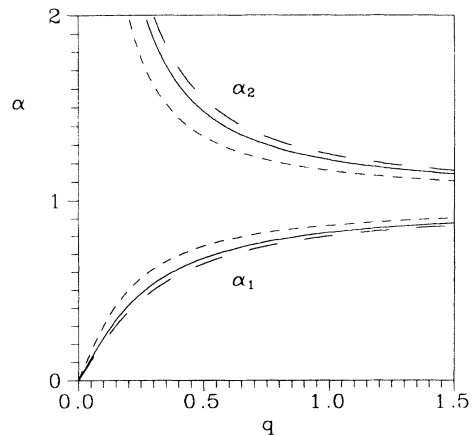


FIG. 2. Behavior of the ellipticity α_i [see Eq. (5.2c)] as a function of the normalized transverse wave vector q for the same parameters as in Fig. 1.

field. By increasing the field, all the eigenvalues decrease and the first one which becomes equal to zero defines the critical field and the critical wave vector q_c for a second-order Fréedericksz-type transition. As already pointed out, this occurs at $Q_x = 0$ if the term \mathcal{F}_1 is not taken into account.

The search for v_c and q_c requires in general a numerical computation, owing to the great complexity of Eq. (4.7) or (4.8) which defines the eigenvalues λ_{in} . It is convenient to consider the $\lambda_{in}(q)$ plots for different values of v and for fixed values of the material parameters. Figures 1 and 3 show the behavior of these curves. In Fig. 3 the critical point lies on the curve $\lambda_{22}(q)$ corresponding to $\epsilon_a^{1/2}v = 0.7536$. As is evident, this point is defined by the conditions $\lambda = 0$, $d\lambda/dq = 0$ (with $d^2\lambda/dq^2 > 0$), or, equivalently,

$$g(0, v_c, q_c; \epsilon_a, r, e) = 0, \quad (6.1a)$$

$$\frac{\partial g}{\partial q}(0, v_c, q_c; \epsilon_a, r, e) = 0, \quad (6.1b)$$

which constitute a system of two equations in the two unknowns v_c , q_c . We note that the eigenvalues have been labeled in such a way that, for $q = 0$, λ_{1n} (λ_{2n}) corresponds to pure splay (twist) deformations; this convention, and the strong mixing between splay and twist deformations with different parity for high elastic anisotropy, is the source of the interchange of the roles of λ_{11} and λ_{22} in Fig. 3 above the threshold voltage.

The dependence of v_c and q_c on r in the absence of flexoelectricity ($e = 0$) has been intensively studied recently by taking into account the influence of a further field and for different boundary conditions. The opposite case, where the flexoelectricity plays a dominant role, has been studied by Pikin and co-workers [7,8]. Here we want to give a more general picture of the dependence of q_c and v_c on the parameters ϵ_a , e , r , with the boundary conditions (2.2).

In Ref. [9] it has been shown that in the absence of flexoelectricity ($e = 0$) a critical value r_0 for r exists, such

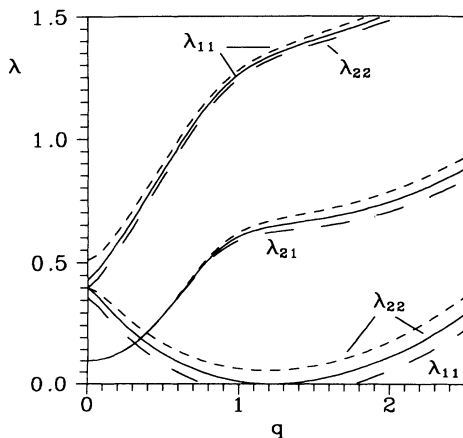


FIG. 3. Same as Fig. 1 but for $e = 0$, $r = 0.1$, and $\epsilon_a^{1/2}v = 0.7$ (short-dashed lines), $\epsilon_a^{1/2}v = 0.7536$ (solid lines), and $\epsilon_a^{1/2}v = 0.8$ (long-dashed lines). The λ_{1n} (λ_{2n}) eigenvalues correspond to pure splay (twist) deformations for $q = 0$.

that the Fréedericksz transition gives rise to an aperiodic distortion for $r \geq r_0$ and to a periodic distortion for $r < r_0$. A numerical analysis shows that for $r \lesssim r_0$ the dependence of q_c and v_c on r is of the type

$$q_c \propto (r_0 - r)^{1/2}, \quad (v_{0c} - v_c) \propto (r_0 - r)^2, \quad (6.2)$$

where v_{0c} is the critical field for $r = r_0$, $q_c = 0$. If we consider the critical wave vector q_c as a function of r , we have here a second-order transition, with r and q_c playing the roles of control parameter and order parameter, respectively. The functions $q_c(r)$ and $v_c(r)$ for $e = 0$ are plotted in Fig. 4 (curves a). The same figure gives the curves corresponding to different values of the flexoelectric coefficient e . As is evident, all these curves display a similar behavior for the asymptotic properties given by Eq. (6.2), where now the quantity r_0 depends on e . This dependence is shown in Fig. 5, which is a phase diagram for the two possible types of distortions that appear immediately above the critical field. The stable configuration is aperiodic if the material parameters are such that the representative point in the plane $(r, e^2/\epsilon_a)$ lies in the lower part of the diagram and periodic in the opposite case. Therefore Figs. 4 and 5 give a unified picture for the two types of instabilities. The striking feature that emerges from Fig. 4 is that for $r < 0.1$ the critical

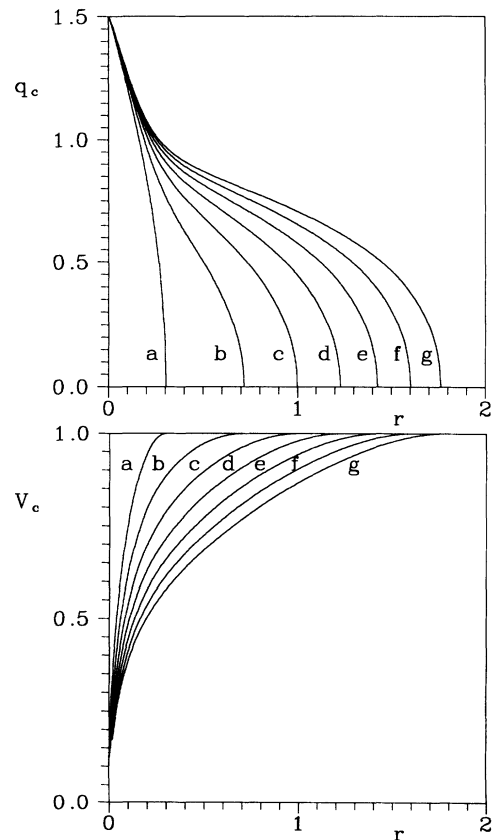


FIG. 4. Normalized critical transverse wave vector q_c and normalized critical voltage $V_c = \epsilon_a^{1/2}v$ as a function of the elastic anisotropy r for $e^2/\epsilon_a = 0$ (curves a), 0.5 (curves b), 1 (curves c), 1.5 (curves d), 2 (curves e), 2.5 (curves f), 3 (curves g).

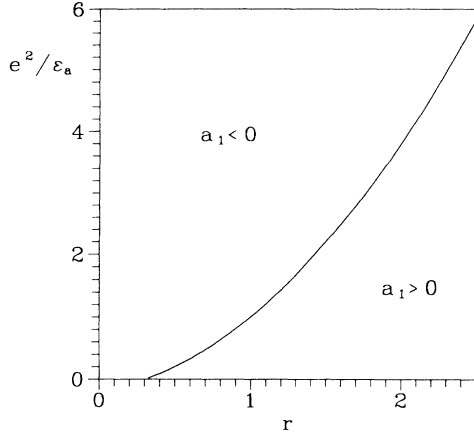


FIG. 5. Phase diagram for the various kinds of second-order transitions: the region $a_1 > 0$ ($a_1 < 0$) corresponds to transversally homogeneous (periodic) deformations.

wave vector q_c becomes practically independent of the flexoelectric coefficient e .

The functions $q_c(r)$ and $v_c(r)$ have been found by a numerical analysis. However, a fully analytical treatment of the transition in the limit of small q is possible and it has indeed been done in Ref. [11] for the particular case $e = 0$. Here we give a similar and even simpler analysis for any value of e , based on the assumption that the function $g(\lambda, q, v, e, \epsilon_a, r)$ and its first derivatives are continuous.

Let us start from the fact that the critical field for an aperiodic transition ($q = 0$) is given by

$$v^2 = \frac{1}{\epsilon_a}. \quad (6.3)$$

The λ value that vanishes is $\lambda_{11}(q = 0)$, whereas all the other values $\lambda_{in}(q = 0)$ are positive. Equations (B6), (B8a), and (B8b) of Appendix B give, for $v^2 = 1/\epsilon_a$ and small q ,

$$\lambda_{11} = a_1 q^2, \quad (6.4)$$

$$a_1 = \frac{8}{\pi^2 r} \left[r^2 + (2r - 1) \left(\frac{\pi^2}{8} - 1 \right) - \frac{\pi^2 e^2}{8 \epsilon_a} \right]. \quad (6.5)$$

It is very easy to show that the two domains of the phase diagram correspond to the two possible signs of the quantity a_1 .

If $a_1 > 0$, the critical value q_c of q is equal to zero. This means that the field defined by Eq. (6.3) is the critical field for the well-known Fréedericksz transition, giving rise to an aperiodic distortion. Strictly speaking, other zeros of λ could be present in addition to the one considered here, corresponding to $q \neq 0$ and $v^2 < 1/\epsilon_a$. In this case, they would give rise to a first-order transition in the sense specified above, namely, with a discontinuity of q_c (in the usual sense, a first-order Fréedericksz transition displays a discontinuity in the distortion amplitude; in order to study such types of transitions, higher-order terms in the free energy must be taken into account). Such a possibility has been tested numerically for differ-

ent values of the material parameters and it has not been found.

If $a_1 < 0$, the exact curve $\lambda(q)$ intercepts the q axis in at least another point, since $\lambda \rightarrow +\infty$ for $q \rightarrow \infty$. This means that the value of v given by Eq. (6.3) is above the critical value v_c for a Fréedericksz transition with $q_c \neq 0$. In fact, for $v^2 < 1/\epsilon_a$ and $q = 0$ all the eigenvalues are positive.

Now it is straightforward to verify that the upper and lower part of the diagram correspond to negative and positive values of a_1 , respectively. In fact, the quantity a_1 monotonically increases with r for fixed e^2/ϵ_a and monotonically decreases with e^2/ϵ_a for fixed r .

In conclusion, we have shown that the main characteristics of the Fréedericksz transition only depend on the material parameters r and e^2/ϵ_a . If one of these parameters is changed, in such a way that the representative point in the plane $(r, e^2/\epsilon_a)$ crosses the line $a_1 = 0$ from below, the quantity q_c changes continuously from the value $q_c = 0$ to a value $q_c \neq 0$.

We finally notice that the asymptotic relations (6.2) can be obtained under the same hypothesis of continuity of the function $g(\lambda, q, v, e, \epsilon_a, r)$ and its derivatives.

VII. DISCUSSION

Let us summarize the results obtained. The theory of the thermal fluctuations of the director in NLC has been originally developed for an infinite medium, where the fluctuations can be considered as a sum of independent overdamped plane waves. In a finite sample the actual shape of the fluctuation eigenmodes is more complex. Here we have considered the case of a planar NLC cell with strong anchoring conditions. In homeotropic cells no coupling of the different Fourier components occurs [12].

The main features of the eigenmodes in the absence of the electric field are the same as that expected for any wave with two possible polarization states in an anisotropic slab, such as, for example, an electromagnetic wave. In fact, the reflection at each boundary plane generates waves with two different polarizations and wave vectors. The eigenmodes are therefore, in general, a superposition of four plane waves with z components of the wave vector \mathbf{Q} equal to $\pm Q_{z1}, \pm Q_{z2}$. The fact that the waves associated to the director fluctuations are overdamped does not change this general feature.

In the presence of an electric field, reasonably simple and exact expressions for the eigenmodes and the eigenvalues are available only if $Q_x = 0$. The main part of this paper is devoted to such a case, and more precisely to the study of the static periodic structures that appear above the threshold field for a Fréedericksz-type second-order transition. In the framework of the theory developed here, these transitions are found by considering the $\Lambda(Q_y)$ curves and looking at the points where $\Lambda = 0$ and $\partial\Lambda/\partial Q_y = 0$. This approach to the Fréedericksz transition is physically more satisfactory and mathematically simpler than that given in Refs. [6–9,11], and allows us to recover all the already known results; in Ref. [12] it

has been applied to homeotropic cells. The case of planar cells considered here is much more complex, since the Fréedericksz transition can give rise to periodic or aperiodic distortions depending on the anisotropy parameter $r = (K_1 - K_2)/K_1$ and on the parameter e^2/ϵ_a . A phase diagram for the two types of distortions has been obtained numerically, and confirmed by a fully analytic calculation based on the perturbation expansion presented in Appendix B: it gives a quantitative basis to the experimentally well-known result that the flexoelectricity plays an essential role in driving the periodic distortions only for small dielectric anisotropy (typically for $|\epsilon_a| \lesssim 0.2$). This approach allows for a unified treatment of the classical Fréedericksz transition and of the Pikin and Lonberg-Meyer instabilities.

The thermal fluctuations that we have theoretically considered here are generally studied experimentally by light-scattering techniques. From this point of view, we have shown that the interpretation of any light-scattering experiment involving eigenmodes with low Q_z and $Q_y \neq 0$ requires a careful and complex analysis.

An even more complicated analysis, not given here, is required for the light-scattering experiments where the undistorted director lies in the scattering plane and the sample is acted upon by an electric field.

In conclusion, this paper gives a contribution to the understanding of the thermal fluctuations and to the interpretation of the light-scattering experiments in real NLC samples. The main characteristics of these topics have been known for many years, but an exact computation had never been done before and it requires rather involved mathematics, as shown here. To this purpose it must be noticed that the complexity of the problem is even greater if the dynamics of the fluctuation is considered. In fact, the boundary effects on the flow associated to the director fluctuations must be taken into account. A fully hydrodynamic treatment is done in Ref. [5] for the particular case $Q_y = 0$ and zero field. An extension of this treatment to the case $Q_y \neq 0$ and $E \neq 0$ is essential for the interpretation of the light-scattering experiments as long as the relaxation times are concerned, but it is far beyond the scope of this paper.

$$\Delta_a V^{(1)} = \frac{\partial}{\partial x} \left[\epsilon_a E^{(0)} \Theta_1 + \frac{e_1 + e_3}{\epsilon_0} (\Theta_{2,y} + \Theta_{1,z}) \right], \quad (\text{A7})$$

$$\begin{aligned} \Delta_a V^{(2)} &= \epsilon_a \left[\frac{\partial}{\partial x} (\Theta_2 E_y^{(1)} + \Theta_1 E_z^{(1)}) + \frac{\partial}{\partial y} (\Theta_2 E_x^{(1)} + E^{(0)} \Theta_1 \Theta_2) + \frac{\partial}{\partial z} (\Theta_1 E_x^{(1)} + E^{(0)} \Theta_1^2) \right] \\ &+ \frac{e_1 + e_3}{\epsilon_0} \left[-\frac{1}{2} \frac{\partial^2}{\partial x^2} (\Theta_1^2 + \Theta_2^2) + \Theta_{2,y}^2 + \Theta_{1,z}^2 + \Theta_1 \Theta_{2,zy} + \Theta_2 \Theta_{1,zy} + \Theta_1 \Theta_{1,zz} + \Theta_2 \Theta_{2,yy} \right] \\ &+ \frac{2e_1}{\epsilon_0} \Theta_{2,y} \Theta_{1,z} + \frac{2e_3}{\epsilon_0} \Theta_{1,y} \Theta_{2,z}, \end{aligned} \quad (\text{A8})$$

where

$$\Delta_a = \epsilon_{\parallel} \frac{\partial^2}{\partial x^2} + \epsilon_{\perp} \frac{\partial^2}{\partial y^2} + \epsilon_{\perp} \frac{\partial^2}{\partial z^2}. \quad (\text{A9})$$

The free-energy densities for fixed voltage and for fixed charge at the electrodes are given, respectively, by [13,14]

APPENDIX A

In order to evaluate the electrostatic energy we must solve the Maxwell equations for the distorted configuration. Only the terms up to the second order in the distortion parameter $\Theta = (\Theta_1, \Theta_2)$ are considered. We therefore write

$$\mathbf{E} = E^{(0)} \hat{\mathbf{z}} + \mathbf{E}^{(1)} + \mathbf{E}^{(2)} + O(\Theta^3). \quad (\text{A1})$$

The dielectric tensor is $\overset{\leftrightarrow}{\epsilon} = \epsilon_{\perp} \overset{\leftrightarrow}{\mathbf{1}} + \epsilon_a \hat{\mathbf{n}} \hat{\mathbf{n}}$, where $\hat{\mathbf{n}} = (1 - \Theta_1^2/2 - \Theta_2^2/2) \hat{\mathbf{x}} + \Theta_2 \hat{\mathbf{y}} + \Theta_1 \hat{\mathbf{z}}$. The flexoelectric polarization is given by

$$\mathbf{P}_f = e_1 \hat{\mathbf{n}} \nabla \cdot \hat{\mathbf{n}} - e_3 \hat{\mathbf{n}} \times \nabla \times \hat{\mathbf{n}}. \quad (\text{A2})$$

This gives for the vector $\mathbf{D} = \epsilon_0 \overset{\leftrightarrow}{\epsilon} \cdot \mathbf{E} + \mathbf{P}_f$ the expression

$$\mathbf{D} = \epsilon_0 \epsilon_{\perp} E^{(0)} \hat{\mathbf{z}} + \mathbf{D}^{(1)} + \mathbf{D}^{(2)} + O(\Theta^3), \quad (\text{A3})$$

where

$$\begin{aligned} \mathbf{D}^{(1)} &= \epsilon_0 \begin{pmatrix} \epsilon_{\parallel} E_x^{(1)} + \epsilon_a E^{(0)} \Theta_1 \\ \epsilon_{\perp} E_y^{(1)} \\ \epsilon_{\perp} E_z^{(1)} \end{pmatrix} + e_1 \begin{pmatrix} \Theta_{2,y} + \Theta_{1,z} \\ 0 \\ 0 \end{pmatrix} \\ &+ e_3 \begin{pmatrix} 0 \\ \Theta_{2,x} \\ \Theta_{1,x} \end{pmatrix}, \quad (\text{A4}) \\ \mathbf{D}^{(2)} &= \epsilon_0 \begin{pmatrix} \epsilon_{\parallel} E_x^{(2)} \\ \epsilon_{\perp} E_y^{(2)} \\ \epsilon_{\perp} E_z^{(2)} \end{pmatrix} + \epsilon_0 \epsilon_a \begin{pmatrix} \Theta_2 E_y^{(1)} + \Theta_1 E_z^{(1)} \\ \Theta_2 E_x^{(1)} + E^{(0)} \Theta_1 \Theta_2 \\ \Theta_1 E_x^{(1)} + E^{(0)} \Theta_1^2 \end{pmatrix} \\ &+ e_1 \begin{pmatrix} -\Theta_1 \Theta_{1,x} - \Theta_2 \Theta_{2,x} \\ \Theta_2 \Theta_{2,y} + \Theta_2 \Theta_{1,z} \\ \Theta_1 \Theta_{2,y} + \Theta_1 \Theta_{1,z} \end{pmatrix} \\ &+ e_3 \begin{pmatrix} -\Theta_2 \Theta_{2,x} - \Theta_1 \Theta_{1,x} \\ \Theta_2 \Theta_{2,y} + \Theta_1 \Theta_{2,z} \\ \Theta_1 \Theta_{1,z} + \Theta_1 \Theta_{2,y} \end{pmatrix}. \end{aligned} \quad (\text{A5})$$

The field equation $\nabla \times \mathbf{E} = \mathbf{0}$ can be satisfied by assuming

$$\mathbf{E} = E^{(0)} \hat{\mathbf{z}} - \nabla V^{(1)} - \nabla V^{(2)}. \quad (\text{A6})$$

The equation $\nabla \cdot \mathbf{D} = \mathbf{0}$ straightforwardly gives

$$\tilde{f} = f_e - \frac{1}{2} \epsilon_0 \mathbf{E} \cdot \overset{\leftrightarrow}{\epsilon} \cdot \mathbf{E} - \mathbf{P}_f \cdot \mathbf{E}, \quad (\text{A10})$$

$$f = \tilde{f} + \mathbf{D} \cdot \mathbf{E} = f_e + \frac{1}{2} \epsilon_0 \mathbf{E} \cdot \overset{\leftrightarrow}{\epsilon} \cdot \mathbf{E}, \quad (\text{A11})$$

where f_e is the elastic free-energy density.

Let us now discuss the equations obtained. Equations

tion (A7) clearly shows that a very important role is played by the x dependence of the distortion; in fact the first order-corrections for \mathbf{E} are identically zero if the distortion angles Θ_i are independent of x , namely, if $Q_x = 0$. In this case our equations greatly simplify, giving, up to second-order terms,

$$\mathbf{E} = E_y^{(2)}\hat{\mathbf{y}} + (E^{(0)} + E_z^{(2)})\hat{\mathbf{z}}, \quad (\text{A12})$$

$$\begin{aligned} \vec{\epsilon} \cdot \mathbf{E} = & \epsilon_\alpha \Theta_1 E^{(0)} \hat{\mathbf{x}} + (\epsilon_\perp E_y^{(2)} + \epsilon_\alpha E^{(0)} \Theta_1 \Theta_2) \hat{\mathbf{y}} \\ & + (\epsilon_\perp E^{(0)} + \epsilon_\perp E_z^{(2)} + \epsilon_\alpha E^{(0)} \Theta_1^2) \hat{\mathbf{z}}, \end{aligned} \quad (\text{A13})$$

$$f = f_e + \frac{1}{2} \epsilon_0 \epsilon_\perp E^{(0)2} + \frac{1}{2} \epsilon_0 E^{(0)} (2\epsilon_\perp E_z^{(2)} + \epsilon_\alpha E^{(0)} \Theta_1^2). \quad (\text{A14})$$

The evaluation of the field component $E_z^{(2)}$, appearing in Eq. (A14), requires the integration of Eq. (A8). This is not an easy task. Therefore we only show that, for $Q_x = 0$, the assumption of uniform electric field gives the exact value for the total free energy up to second-order terms, namely, that the term \mathcal{F}_1 in Eq. (2.3a) is equal to zero. More precisely we show that the theory developed in Sec. II under the assumption $\mathcal{F}_1 = 0$ is self-consistent if $\hat{\mathbf{n}} = \hat{\mathbf{n}}(y, z)$ and if the average electric field \mathbf{E} appearing in Eq. (2.3b) is suitably chosen. Equations (2.11a) and (2.11b) give for $Q_x = 0$

$$\Theta_1 = \cos(Q_y y) \theta_1(z), \quad (\text{A15a})$$

$$\Theta_2 = \sin(Q_y y) \theta_2(z). \quad (\text{A15b})$$

We now solve Eq. (A8) by neglecting the flexoelectricity (which will be considered later),

$$\begin{aligned} \left(\frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) V^{(2)} = & \frac{\epsilon_\alpha}{\epsilon_\perp} E^{(0)} \cos(2Q_y y) \\ & \times \left(Q_y \theta_1(z) \theta_2(z) + \frac{1}{2} \frac{d}{dz} \theta_1^2(z) \right) \\ & + \frac{1}{2} \frac{\epsilon_\alpha}{\epsilon_\perp} E^{(0)} \frac{d}{dz} \theta_1^2(z). \end{aligned} \quad (\text{A16})$$

The solution of Eq. (A16) can be written in the form

$$V^{(2)} = V_1^{(2)}(z) \cos(2Q_y y) + V_2^{(2)}(z). \quad (\text{A17})$$

The first term of $V^{(2)}$ gives no contribution to the free energy. In fact, the corresponding term in the free-energy density, given by Eq. (A14), depends linearly on $E_z^{(2)}$ and its integral vanishes when the integration is performed over a full period along y . The function $V_1^{(2)}(z)$ is easily evaluated from Eq. (A16), and depends on two arbitrary constants which can be adjusted to have $V_1^{(2)}(\pm d/2) = 0$. This ensures the equipotentiality of the electrodes.

The term $V_2^{(2)}(z)$, coming from the y -independent source term in Eq. (A16), gives $E_z^{(2)} = -E^{(0)} \theta_1^2 \epsilon_\alpha / 2\epsilon_\perp$. The final expression of the free-energy density has an average value

$$\bar{f} = f_e + \frac{1}{2} \epsilon_0 \epsilon_\perp (E^{(0)})^2 - \frac{1}{4} \epsilon_0 \epsilon_\alpha (E^{(0)})^2 \overline{\theta_1^2}. \quad (\text{A18})$$

The first term gives the electrostatic energy of the undistorted configuration, the second term becomes equal to the average value of the corresponding term in Eq. (2.3b) if we assume $E = E^{(0)}$.

In conclusion, we have shown that the actual field \mathbf{E} can be written as the sum of a uniform field $E\hat{\mathbf{z}}$ and of a nonuniform field $\mathbf{E}^{(2)}(y, z)$ which gives no contribution to the total dielectric free energy. It is now evident that this nonuniform part of the field, which is second order, gives a contribution to the flexoelectric part of the free energy which is of order higher than two.

We have considered here the free energy for fixed charge at the electrodes. It is immediately verified that such a charge, which is given by the integral of D_z over y at $z = \pm d/2$, does not depend on Θ . It is, however, to be noticed that the local value of D_z at $z = \pm d/2$ is not constant. This means that the fluctuations induce surface currents at the electrodes, a fact which gives a new dissipation term, in addition to the well-known terms related to the viscosity.

The use of the free energy for fixed voltage requires very little change in our computation, since the potential at each electrode is in any case constant, and gives the same result. In order to obtain a Θ -independent voltage the function $V_2^{(2)}(z)$ in Eq. (A17) must be chosen in such a way as to give

$$E_z^{(2)} = -\frac{1}{2} \frac{\epsilon_\alpha}{\epsilon_\perp} E^{(0)} \theta_1^2 + E_0^{(2)}, \quad (\text{A19})$$

with

$$E_0^{(2)} = \frac{1}{2} \frac{\epsilon_\alpha}{\epsilon_\perp} E^{(0)} \overline{\theta_1^2}. \quad (\text{A20})$$

Let us finally briefly discuss the general case where $Q_x \neq 0$. As is evident from Eq. (A7), the distortion gives field gradients which are of the same order of Θ , and which cannot be neglected in any treatment of the thermal fluctuations in the presence of an electric field. However, the integration of Eqs. (A7) and (A8) is not an easy task. It will not be done here, and we only give in this paper the exact solutions for the case $Q_x = 0$.

APPENDIX B

Let us first rewrite the eigenvalue equation (2.12) with the conditions and the symbols defined in Eq. (3.1),

$$\begin{pmatrix} -\lambda + rq^2 - \left(\frac{d}{\pi}\right)^2 \frac{d^2}{dz^2} - \epsilon_\alpha v^2 & -(1-r)q \frac{d}{\pi} \frac{d}{dz} - qev \\ (1-r)q \frac{d}{\pi} \frac{d}{dz} - qev & -\lambda + q^2 - r \left(\frac{d}{\pi}\right)^2 \frac{d^2}{dz^2} \end{pmatrix} \begin{pmatrix} \theta_1 \\ \theta_2 \end{pmatrix} = 0. \quad (\text{B1})$$

We consider the q -dependent terms as a perturbation. The unperturbed solutions correspond to pure splay ($\theta_1 \neq 0, \theta_2 = 0$) and pure twist ($\theta_1 = 0, \theta_2 \neq 0$) deformations, respectively. The nonzero functions $\theta_i(z)$ of each type are alternatively given by $\cos(\pi n z/d)$, with odd n , and $\sin(\pi n z/d)$, with even n . The corresponding λ values are

$$\lambda_{1n}^{(0)} = n^2 - \epsilon_a v^2 \quad (\text{splay}), \quad (\text{B2})$$

$$\lambda_{2n}^{(0)} = r n^2 \quad (\text{twist}). \quad (\text{B3})$$

There are no first-order corrections to λ_{in} and at second order one easily obtains

$$\lambda_{1n} = \lambda_{1n}^{(0)} + q^2 \left(r + \frac{e^2 v^2}{\lambda_{1n}^{(0)} - \lambda_{2n}^{(0)}} + \sum_{m=1}^{\infty} \frac{\Gamma_{nm}^2}{\lambda_{1n}^{(0)} - \lambda_{2m}^{(0)}} \right), \quad (\text{B4a})$$

$$\lambda_{2n} = \lambda_{2n}^{(0)} + q^2 \left(1 + \frac{e^2 v^2}{\lambda_{2n}^{(0)} - \lambda_{1n}^{(0)}} + \sum_{m=1}^{\infty} \frac{\Gamma_{nm}^2}{\lambda_{2n}^{(0)} - \lambda_{1m}^{(0)}} \right), \quad (\text{B4b})$$

where

$$\Gamma_{nm} = \begin{cases} 0 & \text{if } (n-m) \text{ is even} \\ \frac{1-r}{\pi} \frac{4nm}{n^2 - m^2} & \text{if } (n-m) \text{ is odd.} \end{cases} \quad (\text{B5})$$

The series can be summed exactly: we only give the results for $n = 1$, which are of interest for the discussion of the Fréedericksz-type transitions

$$\lambda_{11} = 1 - \epsilon_a v^2 + a_1 q^2, \quad (\text{B6})$$

$$\lambda_{21} = r + a_2 q^2, \quad (\text{B7})$$

where

$$a_1 = r + \frac{e^2 v^2}{1 - \epsilon_a v^2 - r} + \frac{(1-r)^2}{r} \left[\frac{1}{x_1^2 - 1} + \frac{4x_1}{\pi(x_1^2 - 1)^2} \cot\left(\frac{\pi x_1}{2}\right) \right], \quad (\text{B8a})$$

$$x_1^2 = \frac{(1 - \epsilon_a v^2)}{r}, \quad (\text{B8b})$$

$$a_2 = 1 - \frac{e^2 v^2}{1 - \epsilon_a v^2 - r} + (1-r)^2 \left[\frac{1}{x_2^2 - 1} + \frac{4x_2}{\pi(x_2^2 - 1)^2} \cot\left(\frac{\pi x_2}{2}\right) \right], \quad (\text{B8c})$$

$$x_2^2 = r + \epsilon_a v^2. \quad (\text{B8d})$$

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- [1] Group d'étude des Cristaux Liquide (Orsay), *J. Chem. Phys.* **51**, 816 (1969).
- [2] Orsay Liquid Crystal Group, *Phys. Rev. Lett.* **22**, 1361 (1969); *Mol. Cryst. Liq. Cryst.* **13**, 187 (1971).
- [3] V. G. Taratuta, A. J. Hurd, and R. B. Meyer, *Phys. Rev. Lett.* **55**, 246 (1985).
- [4] B. Ya. Zel'dovich and N. V. Tabiryan, *Zh. Eksp. Teor. Fiz.* **81**, 1738 (1981) [*Sov. Phys. JETP* **54**, 922 (1981)].
- [5] J. Papánek, *Mol. Cryst. Liq. Cryst.* **179**, 139 (1990).
- [6] R. B. Meyer, *Phys. Rev. Lett.* **22**, 918 (1969).
- [7] J. P. Bobilev and S. A. Pikin, *Zh. Eksp. Teor. Fiz.* **72**, 369 (1977) [*Sov. Phys. JETP* **45**, 195 (1977)]; Y. Bobilev, V. G. Chigrinov, and S. A. Pikin, *J. Phys. (Paris) Colloq.* **40**, C3-331 (1979).
- [8] S. A. Pikin, *Structural Transformations in Liquid Crystals* (Gordon and Breach, New York, 1991).
- [9] F. Lonberg and R. B. Meyer, *Phys. Rev. Lett.* **55**, 718 (1985).
- [10] P. Ribière, S. Pirkel, and P. Oswald, *Phys. Rev. A* **44**, 8189 (1985).
- [11] C. Oldano, *Phys. Rev. Lett.* **56**, 1098 (1986); E. Miraldi *et al.*, *Phys. Rev. A* **34**, 4348 (1986); W. Zimmermann and L. Kramer, *Phys. Rev. Lett.* **56**, 2655 (1986).
- [12] K. Eidner, M. Lewis, H. K. M. Vithana, and D. L. Johnson, *Phys. Rev. A* **40**, 6388 (1989).
- [13] L. Landau and E. Lifschitz, *Electrodynamique des milieux continus* (MR, Moscow, 1969), p. 70.
- [14] I. Dozov *et al.*, *Europhys. Lett.* **1**, 563 (1986).