Inhomogeneous Fréedericksz transition in nematic liquid crystals

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A theoretical and experimental analysis of a spatial instability developing in a homeotropically aligned nematic liquid crystal film is presented. The explanation for the existence of this instability is supplied through an amplitude equation. This model, which is valid in the vicinity of the Fréedericksz transition, assumes a strong difference between the nematic elastic constants. The first report of such an instability observed in the conditions accounted for by our model, was provided by Cladis and Torza [J. Appl. Phys. **6**, 584 (1975)]. We repeated these experiments in order to confirm the validity of the model. Although carried out far from the Fréedericksz transition, these latter show a good qualitative agreement with the theoretical predictions. The nonlinear analysis allows to understand the dynamical behavior of an interface separating domains of stripes through the occurrence of a zigzag instability.

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I. INTRODUCTION

Nonequilibrium processes often lead in nature to the formation of spatial periodic structures developed from a homogeneous state through the spontaneous breaking of symmetries present in the system [1]. In the course of recent decades, much effort has been devoted to the study of pattern formation (see review [2] and the references therein) arising in systems, such as chemical [3] or catalytic reaction systems [4], gas discharge systems [5], CO_2 lasers [6], and liquid crystals [7–14], or else emerging from hydrodynamic [15] or electroconvective instabilities (see review [16] and the references therein). A unified description for the dynamics of spatial periodic structures developed at the onset of bifurcation is achieved by means of amplitude equations for the critical modes. Such a description is valid in the case of weak nonlinearities and for a slow spatial and temporal modulation of the base pattern [2]. As an example, the Newell-Whitehead equation [17] describes the dynamics of a stripe pattern formed in a two-dimensional system. It is notable that most investigations on pattern formation consider two-dimensional extended isotropic systems. Meantime, few theoretical studies have been performed on anisotropic systems. Nonetheless, some phenomena are intrinsically related to anisotropy as, for instance, the zigzag instability of an interface connecting two symmetrical states [18]. Here, one cannot sweep away the anisotropy problem and achieve a proper description of the system just by rescaling the space coordinates. Liquid crystals are materials where anisotropy plays a fundamental role as it can, in particular, generate inhomogeneous spatial structures. Thus, a stripe pattern can be observed close to the nematic-smectic *A* transition [7–9], in polymer nematics with planar anchoring conditions [10,11] or in a nematic liquid crystal film submitted to an electromagnetic field [12].

The aim of this paper is to depict the appearance of such spatial periodic structures in a nematic liquid crystal sample with homeotropic alignment, under the application of an external electric field. An amplitude equation, based on the assumption of a *strong anisotropy*, gives the theoretical frame to the description of the instability in the vicinity of the Fréedericksz transition. Previous studies of the stationary stripe pattern observed in nematic films have already provided a linear stability analysis of the problem, thanks to variational calculations [8,10–14]. Through the use of trial functions [8] or the resolution of the Euler-Lagrange equations derived from the free energy of the system (Oseen-Frank energy) [10–14], one establishes a criterium for the existence of a stable periodic pattern. However, a nonlinear study is difficult to achieve. The limit that we shall use below makes accessible the study of nonlinear behaviors, such as the evolution of an interface separating two domains of stripes. This analysis is derived from the nonlinear elastic theory of nematic liquid crystals, which is valid here since backflow effects are negligeable close enough to the Fréedericksz transition. As a consequence of the strong difference between the elastic constants, nonlinear spatial terms appear in the amplitude equation even close to the Fréedericksz transition. These nonlinearities along with the anisotropy present in the system leads the inhomogeneous instability.

Experimental observations of this spatial instability have been carried out far from the Fréedericksz transition. These experiments, similar to those made by Cladis and Torza [7], are in good qualitative agreement with the theoretical description developed close to the Fréedericksz transition.

II. INHOMOGENEOUS FRÉEDERICKSZ TRANSITION IN LIQUID CRYSTALS

Most liquid crystal materials are formed of anisotropic-shaped organic molecules. This results in the anisotropy of all their physical properties, especially optical properties. In the nematic phase, the configuration of lowest energy is reached when all rodlike molecules are, on average, aligned

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along a single direction pointed out by a vector $\vec{n}$, which is called the *director* (any description must include the symmetry $\vec{n} \leftrightarrow -\vec{n}$) [19,20]. This direction can be experimentally specified either by applying an external field, e.g., an electric or magnetic field, or by imposing some particular boundary conditions (*anchoring* conditions) at the edges of the confined sample. When two of these constraints are competing, the long-range orientational order may be partially destroyed. Orientational deformations then appear in the system, which are theoretically described by a vector field $\vec{n}(\vec{r},t)$. This latter gives the average orientation of the molecules inside the fluid particle located in $\vec{r}$ at time t. The competition between two opposite constraints occurs, for example, when one tries to lead the reorientation of the molecules thanks to an external field in an anchored layer of nematic liquid crystal. For a sufficiently high magnitude of the field, the initial alignment due to the anchoring disappears in the bulk. This is the so-called Fréedericksz transition [19–21]. The threshold of this transition depends on the geometry of the setup, that is, on the orientation of the anchoring direction with respect to the external field. Besides, the chosen geometry determines the kind of uniform elastic distortion to appear in the medium at the onset of the transition. This distortion is one of the three basic elastic distortions, namely, splay, twist, and bend distortions whose energy cost is quantified by the intrinsic parameters K_1, K_2, and K_3 respectively [19,20]. Due to the nematic anisotropy, these *elastic constants* differ from each other, sometimes strongly enough to induce new phenomena, not achieved in isotropic situations. It has long been known that an inhomogeneous Fréedericksz transition (IFT) leading to the formation of stripes in a nematic sample can be observed due to a strong difference between the elastic constants [7,10]. This occurs in particular with some polymeric liquid crystals like PBG (polybenzyl glutamate) for which the ratio K_1/K_2 is of order 15 [10]. In the planar geometry, the nematic layer of this material undergoes an IFT in place of the usual homogeneous Fréedericksz transition (HFT). This new transition is a second-order transition, whereas the transition from stripes to uniform distortion, which takes place at a higher value of the applied external field, is a first-order transition.

Nevertheless, most thermotropic low-molecular-weight liquid crystals do not show a strong anisotropy in their nematic range [19]. Hence, similar phenomena will be visible only close to the nematic–smectic-*A* phase transition, where the divergence of the bend and twist elastic constants make possible to get high elastic ratios. The first report, to our knowledge, of a spatial instability in a homeotropically anchored layer was provided by Cladis and Torza [7], who studied a cyanobiphenyl (8CB) sample submitted to a magnetic field very close to the Nem/Sm-*A* transition. There, the IFT occurs after the HFT and the instability that generates periodic distortions in the medium from the initial uniform distortion is second order (supercritical bifurcation).

Previous theoretical works developed to explain those experimental observations have proved the possibility of an inhomogeneous instability, minimizing the free energy thanks to periodic trial functions [8] or expressing the condition on the control parameters that allow periodic functions to be solution of the Euler-Lagrange equations [10,11,13,14]. Besides, a quite comprehensive work on magnetically and electrically induced periodic deformations in nematics has been published by Kini [14]. A review of the different situations corresponding to different nematic materials and different external fields is provided and a linear analysis is performed for each situation.

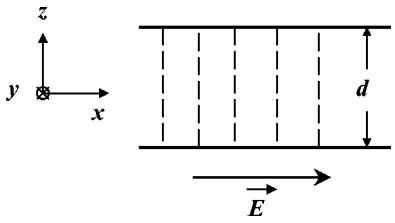

FIG. 1. Schematic representation of a nematic layer of liquid crystal with homeotropic alignment, submitted to a horizontal electric field.

III. THEORETICAL DESCRIPTION

We consider a thin film of nematic liquid crystal, which is confined between two glass plates and subjected to strong homeotropic anchoring conditions (molecules are, on average, perpendicular to the glass plates). The nematic sample is submitted to a horizontal electric field, as shown in Fig. 1. We assume that the dielectric anisotropy constant is positive ($\epsilon_a > 0$). Therefore, the interaction with the external electric field ($\vec{E} = E\hat{x}$, see Fig. 1) favors an alignment of the director parallel to this one. The dynamical equation for the director then reads [19,20]

$$\begin{aligned}\gamma_1 \partial_t \vec{n} = {} & K_3[\nabla^2 \vec{n} - \vec{n}(\vec{n}\cdot\nabla^2\vec{n})] + (K_3 - K_1) \\ & \times [\vec{n}(\vec{n}\cdot\vec{\nabla})(\vec{\nabla}\cdot\vec{n}) - \vec{\nabla}(\vec{n}\cdot\vec{\nabla})] + (K_2 - K_3) \\ & \times \{2(\vec{n}\cdot\vec{\nabla}\times\vec{n})[\vec{n}(\vec{n}\cdot\vec{\nabla}\times\vec{n}) - \vec{\nabla}\times\vec{n}] \\ & + \vec{n}\times\vec{\nabla}(\vec{n}\cdot\vec{\nabla}\times\vec{n})\} + \epsilon_a(\vec{n}\cdot\vec{E})[\vec{E} - \vec{n}(\vec{n}\cdot\vec{E})],\end{aligned} \tag{1}$$

with the conditions

$$\vec{n}\cdot\vec{n} = 1,$$

$$\vec{n}\left(x, y, z = \pm\frac{d}{2}\right) = \hat{z},$$

where K_1, K_2, K_3 are the elastic constants related to the distortions of splay, twist, and bend, respectively, γ_1 is the rotational viscosity, ϵ_a is the dielectric anisotropy constant, and d is the thickness of the cell.

For a critical magnitude of the electric field, the homeotropic state (all molecules vertical, $\vec{n} = \hat{z}$) undergoes a stationary instability, which is a second-order transition. This occurs when the electric field interaction is strong enough, compared with the elastic interaction, to induce a partial re-

alignment of the bulk molecules in the direction of the electric field so as to decrease the total energy of the system. This is the well-known homogeneous Fréedericksz transition [19–21]. The bifurcation parameter is

$$\varepsilon \equiv \epsilon_a E^2 - K_3(\pi/d)^2. \tag{2}$$

When it is positive, the homeotropic state is unstable. The linear study of Eq. (1) shows that the first Fourier mode of the x component of the director is unstable ($n_x = u\cos(\pi z/d)$, $z \in [-d/2, d/2]$). The amplitude u of this mode satisfies the following Landau equation:

$$\gamma_1 \partial_t u = \varepsilon u - b u^3 + K_1 \partial_{xx} u + K_2 \partial_{yy} u \tag{3}$$

where $b \equiv \frac{1}{2}(K_1 - \frac{3}{2}K_3)(\pi^2/d^2) + (3/4)\epsilon_a E^2$. Near the threshold, one has $b \approx K_1 \pi^2/2d^2$. In this configuration, the Fréedericksz transition is a second-order transition.

The preceding equation describes the appearance and dynamics of orientational domains that are formed beyond the Fréedericksz instability threshold, due to the twofold degeneracy of the bifurcated state [22]. Actually, owing to the reflection symmetry with respect to the plane (x,z), that is, to the plane perpendicular to the electric field, the molecules in the bulk can be tilted in two equivalent directions. The interfaces separating these orientational domains eliminate their spatial gradients so as to minimize the free energy of the system, and eventually become straight.

The last amplitude equation has been deduced from a solvability condition, by eliminating the dynamics of the stable modes and considering the asymptotic limit $u \sim \varepsilon^{1/2}$, $\partial_y^2 \sim \partial_x^2 \sim \varepsilon$, $K_1 \sim K_2 \sim K_3 \sim 1$. This limit means that the liquid crystal under consideration is *isotropic*, that is, the actual values of K_1 and K_2 can be renormalized to 1 ($K_1 = K_2 = 1$) by scaling the spatial variables x and y. Recently, in order to describe the zigzag instability of an Ising wall in a nematic liquid crystal sample, one has already considered the limit of *strong anisotropy* [18], where two elastic constants are larger than the third one ($K_1, K_3 \gg K_2$). It is noteworthy that this limit does not allow the simultaneous renormalization of the diffusion coefficients (K_1/γ_1, K_2/γ_1), since this would result in the divergence of higher-order terms in the amplitude equation. These higher-order terms have been asymptotically neglected in Eq. (3). Therefore, in the limit of strong anisotropy, one cannot renormalize the preceding equation and a proper description of the system requires an anisotropic amplitude equation.

When one studies the situation where the twist elastic constant is much smaller than the two other constants ($K_1, K_3 \gg K_2$), then, close to the Fréedericksz transition, the asymptotic amplitude equation reads

$$\gamma_1 \partial_t u = \varepsilon u - b u^3 + K_1 \partial_x^2 u. \tag{4}$$

Above the Fréedericksz threshold, the system contains domains that are separated by straight interfaces in the y direction (splay-bend walls [23]).

The previous equation does not describe the evolution of the system under a perturbation in the y direction, since it is marginal with respect to this sort of perturbations. In order to achieve such a description, one must first take into account higher-order terms in the amplitude equation and, then, consider that the twist elastic constant K_2 scales like the bifurcation parameter ($K_2 \sim \varepsilon$). For K_2 smaller than ε, the higher-order terms predict forthwith the appearance of a spatial structure. We shall later consider this limit to outline the mechanism of the instability. Besides, we shall see that the limit $K_2 \sim \varepsilon$ results in a more complex behavior than the case K_2 far larger than ε ($K_1 \gg K_2 \gg \varepsilon$), for which the perturbations in the y direction satisfy a simple diffusion equation. Henceforth the solvability equation reads

$$\partial_t u = \varepsilon u - b u^3 + K_1 \partial_{xx} u + K_2 \partial_{yy} u + \frac{K_1^2}{a} \partial_{xxyy} u + \frac{3}{4} K_3 \left[u(\partial_y u)^2 - \frac{u^2}{2} \partial_{yy} u \right] \tag{5}$$

with $a \equiv \epsilon_a E^2 + K_3 \pi^2/d^2$. In the previous equation, we have used the asymptotic limit $u \sim \varepsilon^{1/2}$, $\partial_t \sim \varepsilon$, $\partial_y^2 \sim \varepsilon$, $\partial_x^2 \sim \varepsilon$, $K_1 \sim K_3 \sim 1$, and $K_2 \sim \varepsilon$. Hence, the prevailing terms are of order $\varepsilon^{3/2}$, while the first corrections are of order $\varepsilon^{5/2}$. Equation (5) is variational. This follows from the variational aspect of the nematic elasticity model itself [see Eq. (1)].

The expression of the director, involving lower-order terms, can be written as

$$\vec{n} = \begin{pmatrix} n_x \\ n_y \\ n_z \end{pmatrix} = \begin{pmatrix} u\cos\left(\dfrac{\pi z}{d}\right) \\ \left(\dfrac{K_1}{a}\partial_{xy} u + \dfrac{K_1^2}{a^2}\partial_{xy^3} u\right)\cos\left(\dfrac{\pi z}{d}\right) \\ 1 - \dfrac{u^2}{2}\cos^2\left(\dfrac{\pi z}{d}\right) \end{pmatrix}. \tag{6}$$

If the order parameter u only depends on one spatial coordinate, then the director is bidimensional (splay-bend distortion) and lies everywhere in the xz plane. Conversely, when the order parameter depends on the other spatial coordinates, the director becomes tridimensional and all kinds of distortions are present in the nematic medium (splay-bend-twist).

For small positive ε, the system exhibits a second-order transition, as shown in Fig. 2. Near the threshold, the nonlinear antidiffusion term ($u^2 \partial_{yy} u$) is negligible. However, we shall see below that, far enough from the Fréedericksz transition, this nonlinear term can give rise to a spatial instability. In order to study the stability of the bifurcated solutions ($u = \pm\sqrt{\varepsilon/b}$), we shall consider the linear evolution of the system around these solutions. This evolution is described by

$$\partial_t \zeta = -2\frac{\varepsilon}{b}\zeta + K_1 \partial_{xx}\zeta + \frac{K_1^2}{a}\partial_{xxyy}\zeta + \left(K_2 - \frac{3}{8}\frac{\varepsilon}{b}K_3\right)\partial_{yy}\zeta, \tag{7}$$

where $u(x,y,t) \approx \pm\sqrt{\varepsilon/b} + \zeta(x,y,t)$. One can easily see that when $K_2 - \frac{3}{8}(\varepsilon/b)K_3 < 0$, the system presents a spatial instability in the y direction, that gives rise to the stripe pattern experimentally observed. The effective diffusion is actually

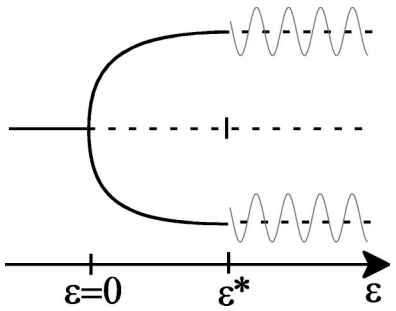

FIG. 2. Diagram of bifurcation. For $\varepsilon<0$ the homeotropic state is stable. $\varepsilon=0$ corresponds to the homogeneous Fréedericksz transition. For $\varepsilon>0$, the homeotropic state is unstable. $\varepsilon=\varepsilon^*$ is the inhomogeneous Fréedericksz transition. For $\varepsilon>\varepsilon^*$, the spatial periodic solution is stable.

negative (antidiffusion), that is, the system focalizes the gradients. Note that the negative part of this diffusion coefficient comes from the nonlinear term $[(u^2/2)\partial_{yy}u]$ of Eq. (5). In order to saturate this instability at short wavelengths, one must include, in the nonlinear elastic theory, higher-order terms that are quartic spatial terms, such as those included in Ref. [24] to study polymeric liquid crystals. These extra terms give rise to additional terms in the solvability equation (5), which now reads

$$\partial_t u=\varepsilon u-bu^3+K_1\partial_{xx}u+K_2\partial_{yy}u-J\partial_{yyyy}u+\left(\frac{K_1^2}{a}+J_1\right)\partial_{xxyy}u+\frac{3}{4}K_3\left[u(\partial_y u)^2-\frac{u^2}{2}\partial_{yy}u\right], \tag{8}$$

where J, J_1 are positive, of the same order of magnitude as the elastic constant K_1 $(J\sim J_1\sim K_1)$, and the term $-J\partial_{yyyy}u$ corresponds to a term of hyperdiffusion.

Considering these new terms, one can demonstrate that, close to the homogeneous Fréedericksz transition, the system undergoes a spatial instability whose bifurcation parameter is

$$\varepsilon^*\equiv\frac{8b}{9}\left(\frac{3}{2}K_3K_2+8J\right)\pm\frac{8b}{9}\sqrt{\left(\frac{3}{2}K_3K_2+8J\right)^2-\frac{9}{4}K_2^2}. \tag{9}$$

The diagram of bifurcation is outlined in Fig. 2: beyond the second-order HFT $(\varepsilon=0)$, the system undergoes a spatial instability or IFT, which is also of second-order type. In the strong anisotropy limit $(K_1,K_3\gg K_2)$, the germ of this nonlinear instability lies in the neighborhood of the HFT. Since the inhomogeneous instability originates from the nonlinearities developed beyond the HFT [see Eq. (5)], the stripe pattern can never appear before the uniform state. This result was already formulated by Allender *et al.* [8] from a linear stability analysis.

The strong anisotropy limit was chosen for the sake of clarity, because it allows a clear and natural explanation of the nonlinear origin of the IFT. The same limit has already been used elsewhere [18] to describe the zigzag instability of a splay-bend interface between two homogeneous distorted domains beyond the HFT. The reported interfacial dynamics will similarly affect the interface separating two domains of stripes, since the equation that describes this dynamics is actually identical to Eq. (5) except for the term $(u^2/2)\partial_{yy}u$, which is neglected, and for the spatial derivative $\partial_{x^2y^4}u$, which is added in order to saturate the instability at short wavelengths. Nevertheless, this limit of *strong anisotropy* does not account fully for the conditions of our experiments. Indeed, these have been performed close to the Nem/Sm-*A* transition characterized by the divergence of the bend and twist elastic constants. Figure 3 shows the evolution curves of the 8OCB elastic constants close to the Nem/Sm-*A* transition. These curves correspond to the parametrization used by Allender *et al.* [8] to compare their experimental results with their theoretical predictions (the authors have considered the experimental curves given in Ref. [25]). From them, one can see that the bend constant diverges much earlier than the twist constant when one approaches the Nem/Sm-*A* transition. Therefore, the use of the limit $K_2\ll K_3$, seems quite

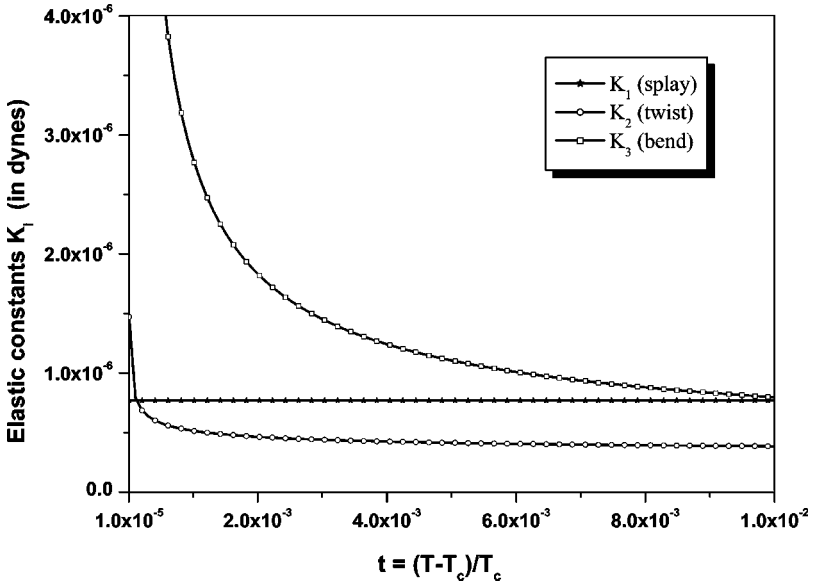

FIG. 3. Evolution curves of the 8OCB elastic constants in the vicinity of the Nem/Sm-*A* transition. These curves correspond to the parametrization, achieved by Allender *et al.* [8] and to the experimental results given by Madhusudana and Pratibha [25].

pertinent to describe the experimental situation. One can also justify the use of the limit $K_2 \ll K_1$. On the contrary, the limit $K_1 \sim K_3$ appears quite unrealistic. In order to outline the behavior of the system close to this divergence, one should rather consider that the bend and twist elastic constants scale like as a power function of ε ($K_1 \sim 1$, $K_2 \sim \varepsilon^{1-\alpha}$, $K_3 \sim \varepsilon^{-\alpha}$, $\alpha > 0$). The parameters α must be chosen between zero and 1 ($0 < \alpha < 1$), in order to satisfy the divergence of the bend elastic constant while keeping the twist elastic constant small. We have denominated extreme *anisotropy limit* the case of $K_3 \gg K_1 \gg K_2$. This limit is fulfilled by the conditions of the experiments (cf. Fig. 3). Considering this new limit, the amplitude equation now reads

$$\partial_t u = \varepsilon u - b u^3 + K_1 \partial_{xx} u + K_2 \partial_{yy} u + \frac{3}{4} K_3 \left[u (\partial_y u)^2 - \frac{u^2}{2} \partial_{yy} u \right] + \text{h.o.t.} \qquad (10)$$

The first three terms on the right-hand side are, once again, dominant. They are of order $\varepsilon^{3/2}$, while the corrections are of order $\varepsilon^{5/2-\alpha}$. Hence both limits, strong and extreme anisotropy limits, lead, close to the Fréedericksz transition, to similar equations. They both account for the appearance of an inhomogeneous spatial structure in the y direction and a zigzag instability of the interface between these states. Here it is important to emphasize that, although this extreme anisotropy limit can be fully justified by the behavior of the elastic constants near the nematic/smectic-A transition, the dependence of these constants with respect to the bifurcation parameter has no physical meaning. It is just a mathematical artifact that enables to correctly describe the dynamics close to the Fréedericksz transition.

One could eventually study the limit $K_3 \gg K_1 \sim K_2$. With this limit, one still finds the appearance of a spatial instability since its origin lies in the nonlinear diffusion term $(u^2/2)\partial_{yy} u$ [see Eq. (8)], which is governed by the bend elastic constant and is, therefore, still present in the amplitude equation. However, the spatial structure is now isotropic, that is, the system exhibits stripes whose orientation depends on the initial conditions. From this, one can conclude that the driving effect of the spatial instability is the bend deformation, but the reason for a privileged direction of the stripes is the anisotropy between the twist and splay deformations.

IV. EXPERIMENTS CLOSE TO THE NEMATIC/SM-*A* TRANSITION

The experiments have been carried out beyond the HFT, since in the vicinity of this transition any experimental imperfection is significant. Nevertheless a good qualitative agreement with the preceding theoretical description was noticed. Figure 5 shows the inhomogeneous solution observed experimentally.

In the experiments we used a cyanobiphenyl compound, namely, the octyloxycyanobiphenyl (8OCB) whose nematic range lies in the temperature interval 67 °C–80 °C [26] and ends at low temperatures by a nematic-smectic A phase transition. The cell that contains the nematic sample is made of two parallel glass plates separated by two mylar spacers, which determine the thickness of the cell ($d \sim 100\ \mu$m—see Fig. 4). The glass surfaces have been coated with lecithin in order to provide a homeotropic anchoring (molecules perpendicular to the plates). Besides, the lower glass plate is covered with a transparent conductive oxide, except for a thin channel (2 mm width) in the middle of the plate. This allows to apply a voltage difference to the opposite conductive areas and, hence to induce a horizontal electric field (see Fig. 1). The sample is thus subjected to a sinusoidal voltage drop $V_{PeakToPeak} = 200$ V with a high frequency ($f = 10$ kHz). All experimental observations were achieved thanks to a polarizing microscope. A three charge-coupled device camera placed on the top of the microscope allows to record images.

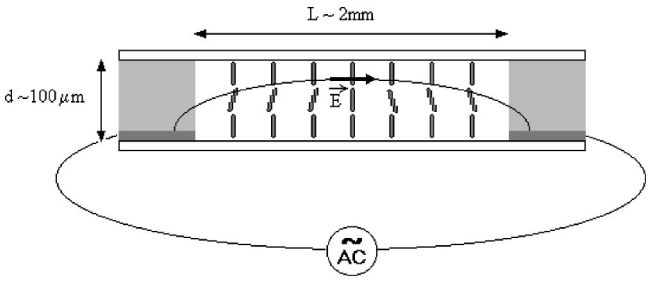

FIG. 4. Experimental setup.

The dielectric constant of the 8OCB is positive ($\epsilon_a = 8.7$ at $T = 69.8$ °C [27]), so that the electric interaction favors an alignment of the molecules in the field direction. In this geometry, usually denominated *bend geometry*, the orientational deformations that appear beyond the Fréedericksz instability are bend distortions along the vertical direction. Above this threshold, one expects a sample in a uniform distorted state that corresponds to one of the equivalent states $\pm \sqrt{\varepsilon/b}$. Actually, as shown in Fig. 5(a), the HFT leads here to the formation of an interface separating two domains of opposite orientational bend distortions. This is not a surprise since we know that such an interface is also the solution of Eq. (4) for the unstable mode amplitude. However, one must underline that, in our situation, such a solution is forced by the curvature of the electric flux lines (see Fig. 4). Even small, this one introduces a small vertical component in the electric field that lifts the degeneracy of the HFT by favoring one of the two homogeneous bifurcated states. Since this vertical component gets reversed across the sample, this results in two opposite domains whose interface is located at the position of null electric field gradient. This interface is called the splay-bend Ising wall [23], in view of the elastic distortions surrounding the interface.

In the main part of its nematic range, the 8OCB compound shows a weak elastic anisotropy: $K_1 \sim 6{,}7 \times 10^{-12}$ N, $K_2 \sim 3{,}4 \times 10^{-12}$ N, and $K_3 \sim 8 \times 10^{-12}$ N at $T = 69.8$ °C [25], so that $K_2/K_1 \approx 0.51$ and $K_3/K_1 \approx 1.19$. Thus, the IFT is not expected to occur, which is experimentally confirmed. However, the evolution curves of the 8OCB elastic constants provided by the literature (see, for example [25]) indicate that a strong increase in this anisotropy occurs at the Nem/Sm-A transition (see, also Fig.

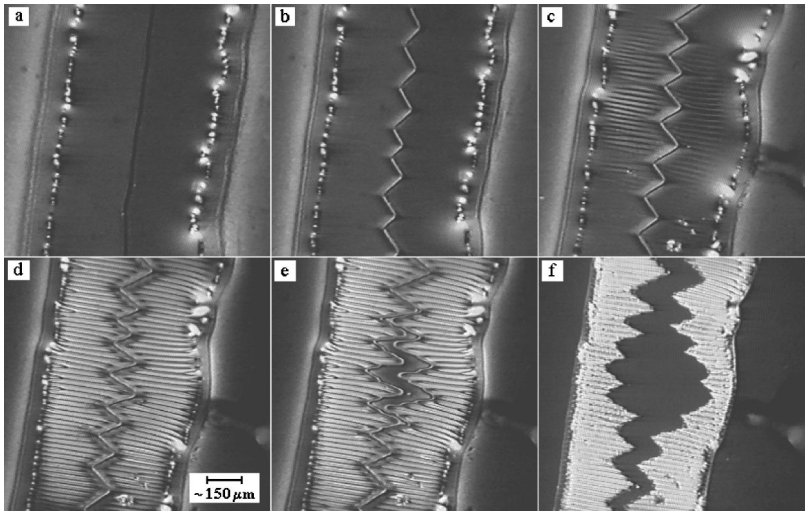

FIG. 5. Experimental pictures of the nematic sample between crossed polarizers. The temperature is gradually decreased from (a) to (f) in order to get closer and closer to the Nem/Sm-*A* transition.

3). Close enough to this transition, the bend and twist constants diverge whereas the splay one remains unchanged. This divergence, which is a pretransitional effect, expresses that bend and twist deformations are forbidden in smectic layers, since they are too costly in energy. Yet, the divergence of the bend elastic constant occurs quite before the twist one. For instance, $(K_3/K_1)_{t=10^{-5}} \approx 82(K_2/K_1)_{t=10^{-5}}$, where $t=(T-T_c)/T_c$ is the bifurcation parameter and T_c is the temperature of the Nem/Sm-*A* transition, while $(K_3/K_1)_{t=10^{-2}} \approx 3.8(K_2/K_1)_{t=10^{-2}}$. Since in the geometry of our experiment, the distortion that appears above the HFT is mainly a bend distortion, one must expect some big qualitative changes due to the proximity of the Nem/Sm-*A* transition.

In the experiments, the temperature was gradually decreased in order to get closer and closer to the Nem/Sm-*A* transition. Far enough from the Nem/Sm-*A* transition, one observes the HFT with one of its topological defect, that is the splay-bend Ising wall [see Fig. 5(a)]. Later on, this interface undergoes an instability and becomes a faceted line [see Fig. 5(b)]. As shown in Ref. [18], the equation (5) provides an explanation for the zigzag instability of an interface that connects the two homogeneous bifurcated states of the HFT $(0<\varepsilon<\varepsilon^*)$. One must underline that this interface instability expresses the appearance of an intrinsic length in the system, according to the terminology of Turing [28]. This instability always precedes the inhomogeneous instability, for large wavelengths, and occurs because the interface is marginal with respect to any spatial perturbation along the *x* axis, as long as the system is invariant by translation in that direction [30]. The interface between the two periodic states of the IFT $(\varepsilon>\varepsilon^*)$ follows the same zigzag dynamics, as actually observed experimentally [see Fig. 5(d)] and numerically (see Fig. 6). This dynamics is described by an order parameter that satisfies a one-dimensional Cahn-Hilliard equation. The domains or *facets* (zig and zag) exhibit a conservative coarsening dynamics characterized by a law of interaction between the domains, which is an exponential function of the distance [29]. In Ref. [18] the term $u^2\partial_{yy}u$ is neglected, since it does not play any role in the zigzag instability of an Ising wall, which connects the two homogeneous bifurcated states. Numerical simulations of Eq. (8) are in agreement with the experimental observations. In particular, one can observe the zigzag instability of an interface separating two inhomogeneous states.

If one further decreases the temperature, one can see domains of stripes develop on both sides of the interface [see Fig. 5(c)]. These domains first grow in the central part of the sample, because the temperature there is slightly lower than in the surrounding regions. Microscopy observations using crossed polarizers indicate that the stripes introduce a periodic distortion of splay in the *xy* plane [see Fig. 5(f)], as well as probable twist distortions in the vertical direction. The wavelength of the stripes is about 20 μm. For lower temperatures, the domains grow and eventually fill the sample [Fig. 5(d)]. Further cooling finally leads to the transition toward the smectic-*A* phase, which starts at the core of the splay-bend wall, where molecules are vertical. The wall opens up to let the smectic phase spread over the sample.

At an even lower temperature, one can see the *virgule texture* [7] [see Fig. 5(f)] and eventually the *honeycomb tex-*

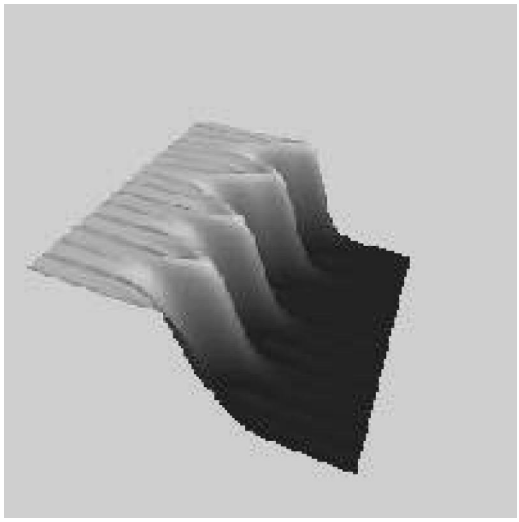

FIG. 6. Numerical simulation of Eq. (8) with $\varepsilon=0.80$, $a=b=1$, $K_1=2.00$, $K_2=0.17$, $K_3=3.5$, $J=0.11$, and $J_1=1$.

ture ([7], not shown). In Ref. [7] these textures are observed beyond the threshold of the Nem/Sm-*A* phase. They correspond to a metastable state that keeps the memory of the very distorted state of the nematic phase after the transition. In our experiments [see Fig. 5(f)] two fronts propagate in opposite directions in order to replace this corrugated structure by the smectic *A* phase.

V. CONCLUSION

We have theoretically described the growth of a spatial instability in a thin layer of nematic liquid crystal under homeotropic anchoring. Close to the Fréedericksz transition, the existence of this instability has been inferred from an amplitude equation, assuming a strong or extreme elastic anisotropy limit, that is, a large difference between the elastic constants. The strong anisotropy limit mimics a situation where the twist elastic constant is much smaller than the other ones ($K_1, K_3 \gg K_2$), whereas the extreme anisotropy limit stands for $K_3 \gg K_1 \gg K_2$. Although this instability has been experimentally observed far from the Fréedericksz transition, a good qualitative agreement between theory and experiments was noticed. The *strong* and *extreme anisotropy* limits allow a nonlinear analysis and, therefore, the comprehension of nonlinear behaviors, such as those presented by the interface between two inhomogeneous domains.

A spatial instability similar to the one studied here was observed in polymeric liquid crystal samples under planar anchoring [10]. The appearance of this instability was characterized experimentally as well as theoretically [10,11]. The theoretical predictions are based on variational calculations on the Frank-Oseen free energy, and require an expression of the director field that takes into account the spatial symmetry breaking. From our point of view, the appearance of this instability is once again related to a nonlinear instability. This can be emphasized by considering the first unstable Fourier mode in this geometry [$n_z = u \cos(\pi z/d)$], which, close to the Fréedericksz transition, satisfies the following equation:

$$\partial_t u = \mu u - \frac{3H^2}{4} u^3 + K_3 \nabla_\perp^2 u - \frac{3}{4}(K_3 - K_2) u^2 \partial_{yy} u + \frac{3}{4}(K_2 - K_1) u^2 \partial_{xx} u + \frac{3}{4}(2K_2 - K_3) u (\partial_x u)^2 + \frac{3}{4}(K_1 - K_2 + K_3) u (\partial_y u)^2 + \text{h.o.t.} \quad (11)$$

where $\mu = [\chi_a H^2 - K_1(\pi/d)^2]$, $\nabla_\perp^2 \equiv \partial_{xx} + \partial_{yy}$, and H is the amplitude of the vertical magnetic field.

The above equation is again asymptotically described by the isotropic Landau equation. The prevailing terms are actually the first three terms on the right-hand side. Then, one cannot predict the appearance of the inhomogeneous instability close to the Fréedericksz transition since the system is isotropic near this transition. Nevertheless, the nonlinear corrections give a clue to a possible nonlinear instability far enough from the HFT. Experimentally, the inhomogeneous transition occurs from the undistorted state and is a second-order transition [10], which cannot be justified by the above model. One way to provide an explanation would be to consider higher-order spatial linear terms like those used in [24] to describe the strong orientational correlation due to the interconnectivity of polymer chains.

For nematic samples with an oblique alignment and submitted to a magnetic field normal to the anchoring direction, the existence of a spatial instability has also been theoretically pointed out (see Ref. [13]). The elastic anisotropy was shown to be responsible for the instability.

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[30] Due to the curvature of the electric flux lines, the condition of a translational invariance along the *x* axis is not fulfilled in our experiments. Nevertheless, since the electric field gradient is small, this will not affect strongly the interfacial behavior and the zigzag instability will still be visible.