Spatial patterns mediated by subcritical and supercritical thermal fluctuations in the splay Fréedericksz transition

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The magnetically induced splay Fréedericksz transition is reexamined to look for pattern forming phenomena slightly above or below criticality. By using our traditional scheme of stochastic nematodynamic equations, situations are, respectively, found of transient and permanent predominance of transversal periodicities (wave numbers) along the direction perpendicular to the initial orientation within the sample. The relevance of these predictions in relation with recent observations in the electrically driven splay Fréedericksz transition, and in general with other pattern forming phenomena, is stressed.

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I. INTRODUCTION

The Fréedericksz transition in nematic liquid crystals constitutes a particularly well-investigated example of transient pattern forming instability [1–17]. The broad theoretical and experimental existing evidence supports the idea that the reorientational transition originated either electrically or magnetically induces, under appropriate conditions, hydrodynamic motions that finally result in the transient appearance of striped textures of characteristic periodicity in the plane of the sample. The usual magnetic experiments refer either to the splay or twist Fréedericksz geometries. In the first situation, perpendicular [1,2] as well as oblique [7] stripes, relative to the initial director alignment, were experimentally found. When referring to the twist instability, only textures of perpendicular stripes have been reported [6,11].

On the other hand, very recent experiments conducted by Buka *et al.* [12,13] on the electrically induced splay Fréedericksz instability reveal unexpected features of this pattern formation phenomenon. To our understanding two of their observations are singularly significative. The first one consists in the appearance of a set of fairly regular stripes, preferentially aligned parallel (neither perpendicular nor oblique) to the initial director orientation. Second, a certain degree of pattern structure was apparently detected even slightly below the threshold of the Fréedericksz instability. The authors refer themselves to this last texture as a fluctuating structure [13] and they clearly stressed the need for a proper consideration of thermal fluctuations to account for some of the observed experimental facts.

The convenient reformulation of the standard set of nematodynamic equations to incorporate thermal fluctuations, and the use of such an enlarged model to analyze the dynamics of the pattern formation process following the reorientational instability, has been our precise goal during these past years [14–17]. In particular, the splay geometry under a magnetic forcing was specifically investigated [15], but focusing on regimes well above criticality ($h^2 \equiv H^2/H_c^2 \geq 5$), for which conditions of perpendicular and oblique stripes were extensively analyzed. After the experiments by Buka *et al.*, we are singularly motivated to reexamine again the splay Fréedericksz instability slightly above or even below criticality to look for eventual patterned structures of parallel stripes relative to the initial orientation within the sample.

Our results, applying to the magnetic Fréederiecksz transition, indeed support those observations, by predicting both subcritical parallel periodicities. A direct quantitative comparison with the experiments mentioned above, however, is not pursued. Actually, this would probably be meaningless due to the well-known specificities of the electrically induced instability mainly arising in the inhomogeneities of the field inside the sample, its finite conductivity, or flexoelectric couplings [13]. Rather, we just want to show that such *a priori* anomalous behavior coherently fits into our scheme of stochastic nematodynamic equations, featuring in this way genuine and overlooked trends of pattern forming phenomena mediated by thermal fluctuations.

II. MODEL EQUATIONS

We proceed here, analogously as we did in Refs. [14–17], by formulating the set of stochastic nematodynamic equations including internal fluctuations through a generalized time-dependent Ginzburg-Landau model. For the splay geometry one considers as usual the director $\bar{n}$ aligned initially parallel to the limiting plates separated a distance d along the z direction: $\bar{n}^0=(1,0,0)$. The magnetic field is applied perpendicular to this initial orientation and to the plates: $\bar{H}=(0,0,H)$. At the limiting plates one prescribes standard free-free boundary conditions: $\partial_z v_x = \partial_z v_y = v_z = n_z = 0$ at $z = \pm d/2$.

The relevant dynamical variables we want to retain are the components n_y, n_z of the director and the three components of the velocity. Such a description allows for general three-dimensional reorientational processes taking place out of the plane dictated by the initial alignment and the applied magnetic field. Actually, by considering transversal reorientations along the y direction, the appearance of a striped texture parallel to the initial orientation ($q_y \neq 0$) is interpreted with quite a simple and intuitive argument. In this respect, note that modes of distortion with $n_y \neq 0$ introduce, relative to the initial orientation along the x-axis permanently fixed at the limiting plates, twist deformations that are elastically favored when compared with the standard splay distortions for $n_z \neq 0$ ($K_1 > K_2$). One could then imagine an alternative route to transfer distortion energy involving transversal distortions along the y direction and which would be especially favored under low-enough magnetic forcings. To support this explanation it is also apparent from the general nematodynamic equation [see Eq. (2) below] that the only available way to couple splay, n_z, and twist, n_y, modes of distortion requires the consideration of wave numbers $q_y \neq 0$. On the other hand, the usual bend modes associated to wave numbers $q_x \neq 0$ would only be excited at high-enough magnetic fields due tc their largest elastic energy ($K_3 > K_1 > K_2$).

Let us try now to transfer these qualitative ideas into the appropriate set of stochastic nematodynamic equations. When applied to the splay geometry they read [15]

$$d_{t'}\begin{bmatrix} n_y \\ n_z \\ v_x \\ v_y \\ v_z \end{bmatrix} = \underline{A}(\bar{n},\bar{v})\begin{bmatrix} \frac{\delta F}{\delta n_y} \\ \frac{\delta F}{\delta n_z} \\ \frac{\delta F}{\delta v_x} \\ \frac{\delta F}{\delta v_y} \\ \frac{\delta F}{\delta v_z} \end{bmatrix} - \frac{1}{\rho}\begin{bmatrix} 0 \\ 0 \\ \partial_x p \\ \partial_y p \\ \partial_z p \end{bmatrix} + \begin{bmatrix} \xi_y \\ \xi_z \\ \partial_\alpha \zeta_{\alpha x} \\ \partial_\alpha \zeta_{\alpha y} \\ \partial_\alpha \zeta_{\alpha z} \end{bmatrix},$$

$$\alpha = (x,y,z) \ . \qquad (1)$$

In Eq. (1), A is an operator containing both reversible and dissipative contributions that incorporates the appropriate set of reorientation and flow viscosity parameters [see Eq. (2.1) in Ref. [15]].

The functional derivatives of the free energy are

$$\frac{\delta F}{\delta n_y} - = -K_1(\partial_y^2 n_y + \partial_{yz}^2 n_z) - K_2(\partial_z^2 n_y - \partial_{yz}^2 n_z) - K_3 \partial_x^2 n_y \ ,$$

$$\frac{\delta F}{\delta n_z} - = -K_1(\partial_{yz}^2 n_y + \partial_z^2 n_z) - K_2(\partial_y^2 n_z - \partial_{yz}^2 n_y) - K_3 \partial_x^2 n_z - \chi_a H^2 n_z \ , \qquad (2)$$

$$\frac{\delta F}{\delta v_\alpha} = \rho v_\alpha \ , \quad \alpha = (x,y,z)$$

written in terms of the elastic constants K_i and the anisotropic part of the magnetic susceptibility χ_a. ρ and p are, respectively, the mass density and the pressure. Finally the last term in Eq. (1) introduces Gaussian random forces that represent sources of thermal noise. Accordingly they are prescribed with zero mean value and satisfying appropriate fluctuation-dissipation relations

$$\langle \xi_\alpha(\bar{r}_1,t'_1)\xi_\beta(\bar{r}_2,t'_2)\rangle = 2\delta_{\alpha\beta}\frac{K_B T}{\gamma_1}\delta(\bar{r}_1-\bar{r}_2)\delta(t'_1-t'_2) \ ,$$

$$\langle \zeta_{\alpha\beta}(\bar{r}_1,t'_1)\zeta_{\delta\gamma}(\bar{r}_2,t'_2)\rangle = 2K_B T \mathsf{M}_{\alpha\beta\delta\gamma}\delta(\bar{r}_1-\bar{r}_2)\delta(t'_1-t'_2) \ . \qquad (3)$$

In Eq. (3), γ_1 stands for the pure reorientational viscosity whereas M is a tensor that contains all the flow viscosity parameters [15].

Succesive standard manipulations [14–17], i.e., incompressibility requirement, minimal coupling approximation, and negligible inertia, transform Eq. (1) into a more appropriate dimensionless version ($t' = \tau_0 t; \tau_0 = \gamma_1/(K_1\pi^2/d^2)$) for the Fourier amplitudes ($n_{y;\bar{Q}}(t), n_{z;\bar{Q}}(t); \bar{Q} = \bar{q}/(\pi/d)$) of the director components. In a linear approximation we have

$$\underline{\gamma}\partial_t \bar{n} = \underline{K}\bar{n} + \bar{\psi}, \quad \bar{n} \equiv \bar{n}_{\bar{Q}}(t) = \begin{bmatrix} n_{y;\bar{Q}}(t) \\ n_{z;\bar{Q}}(t) \end{bmatrix} . \qquad (4)$$

$\bar{Q}$ is the dimensionless wave number for the periodicities in the plane of the sample. $\underline{\gamma}$ and $\underline{K}$ are self-adjoint matrices associated to the viscosity and elastic contributions, respectively, whereas $\bar{\psi}$ collects the entire effect of thermal fluctuations (for explicit expressions of all these quantitities, see Appendix B in Ref. [15]). Finally, Eq. (4) is converted into a dynamical equation for the components of the structure factor $S_{\alpha\beta}(\bar{Q};t) \equiv \langle n_{\alpha;\bar{Q}}(t) n_{\beta;-\bar{Q}}(t)\rangle$.

III. RESULTS

The emergence of periodic structures is monitored by studying the time-dependent evolution of the structure factor following a sudden change at $t=0$ from an initial situation $H_i = 0$ to a final one H either below, $H < H_c$, or slightly above criticality, $H > H_c$ ($H_c^2 = K_1\pi^2/\chi_a d^2$). Here we will restrict the range of considered values to $0.9H_c < H < 1.5H_c$. The position in $\bar{Q}$ space of the maximum of $S_{\alpha\beta}(\bar{Q};t)$ denoted $\bar{Q}_{\max}(t)$ is associated with the characteristic wave number of the striped texture and its temporal evolution form $\bar{Q}=0$ with the pattern development.

As anticipated in the preceding section, we will restrict our analysis to the temporal evolution of $S_{yy}(\bar{Q};t)$, i.e., the transversal component of the structure factor. In order to describe any possible pattern formation phenomenon, we compute the pair of wave numbers (Q_x, Q_y) corresponding to the maximum of the structure factor at different times. We always find that $Q_x = 0$ but Q_y evolves from zero as it corresponds to the development of stripes parallel to the initial orientation within the sample. In Figs. 1 and 2 we show definite evidence of this behavior applying, respectively, to slightly subcritical and supercritical reorientational evolutions from an initially undistorted sample.

It is very interesting to note, however, the different behavior shown by these two figures. In the first case, for $H < H_c$, the preponderance of modes $Q_y \neq 0$ appears to be

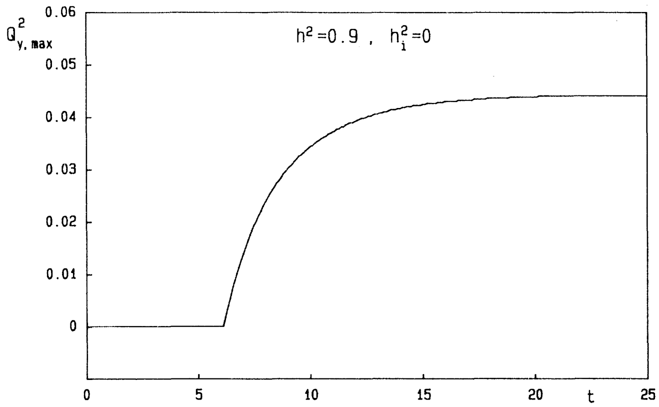

FIG. 1. Time evolution of the maximum of $S_{yy}(\overline{Q},t)$ corresponding to a transition from $H_i=0$ to $H=\sqrt{0.9}H_c$. The values of the material parameters are those of MBBA: $\eta_a=41.6$ cP, $\eta_b=23.8$ cP, $\eta_c=103.5$ cP, $\nu_1=50.8$ cP, $\gamma_1=76.3$ cP, $K_2/K_1=\frac{3}{5}$, $K_3/K_1=\frac{8}{5}$. Times are taken in units of $\tau_0=\gamma_1/(K_1\pi^2/d^2)\simeq 10$ sec for typical samples with $d=10^{-2}$ cm.

a persistent feature, after an initial transient has eliminated the predominance of the homogeneous mode $\overline{Q}=0$. It is also worth noting that for subcritical conditions, such as those corresponding to Fig. 1, nonlinear effects are not expected to be relevant enough to modify the predictions of our linear approach. Actually, the order of magnitude of the structure factor for $H<H_c$ is given by the intensity of the thermal fluctuations estimated here to be 10^{-10} scaled in units of the typical volume sample ($V\simeq 10^{-2}$ cm^3) and relaxation time ($\tau_0\simeq 10$ sec).

On the other hand, for $H>H_c$, Fig, 2, the preponderance of inhomogeneous modes of parallel stripes appears only as a singular transient effect, within the limits of validity of our linear theory [18], finally disappearing to give rise to homogeneous distortions in the plane of the sample.

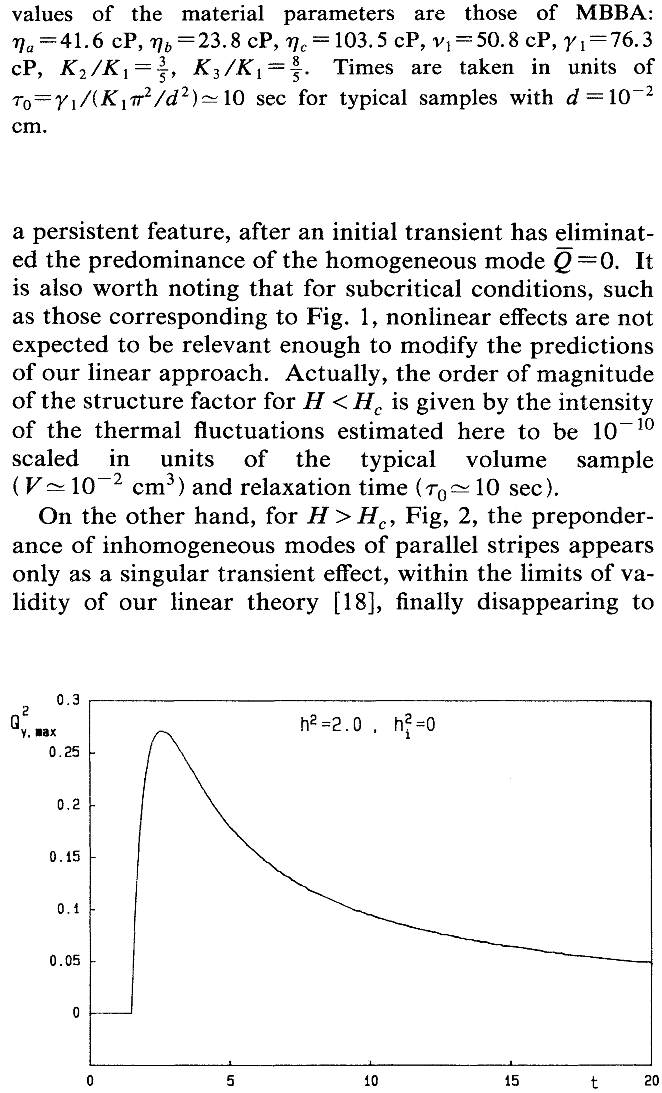

FIG. 2. Time evolution of the maximum of $S_{yy}(\overline{Q},t)$ corresponding to a transition from $H_i=0$ to $H=\sqrt{2}H_c$. Values of the material parameters are those of Fig. 1.

IV. CONCLUSIONS AND FURTHER PERSPECTIVES

The magnetically induced splay Fréedericksz transition has been reexamined to look for pattern forming phenomena slightly above or below criticality. According to the obtained results shown in Figs. 1 and 2, one is then tempted to interpret the spatial periodicity predicted subcritically as a true effect of fluctuations. One could even speculate in view of Fig. 2 that such an effect might be transitorily relevant also for supercritical reorientations until the mode of homogeneous distortions $\overline{Q}=0$, predicted to asymptotically dominate for such small enough values of the magnetic forcing [15], would be definitively predominant. Both below and above the instability threshold, twist fluctuations of the director may develop periodic modes of distortion along the transversal direction, to favorably relieve, in energetic terms, the otherwise more expensive splay distortions. Below threshold, such director fluctuations remain small, scaling essentially with intensity of thermal noise, whereas above it they become amplified as a result of their instability with respect to the symmetry-breaking transition caused by the magnetic field.

With respect to the electrically driven experiments of Buka *et al.*, we did not attempt, as mentioned in the Introduction, a quantitative comparison of our results with their observations. Certainly, the basic equations appropriate to each situation are different enough to permit a reasonable comparison. However, at a qualitative level some striking similarities are worth mentioning. First of all and most important, subcritical parallel periodicities are predicted as detected in the experiments. Second, it is also interesting to point out that the patterns observed supercritically for the electrically driven Fréedericksz transition are indeed transient [12,13] but their disappearance apparently follows a different mechanism with respect to the one based on wall recombination that was common to the earlier observations for the magnetically driven instability [1,2,6,7,11]. In this respect the predicted transient nature shown in Fig. 2 is also peculiar because in all the cases we have analyzed previously [14,17], corresponding to situations for which transient patterns have been experimentally predicted, the asymptotic evaluation of $Q_{\max}(t)$ has always led to non-zero values. Actually, when we were interested in the description of their late stage disappearance we necessarily had to adopt a completely different approach based on domain-wall dynamics [19]. Regarding subcritical periodicities it is worth emphasizing that what Buka *et al.* really observed in this situation was a fluctuating two-dimensional structure persisting as long as the field is applied. The persistent nature of the pattern agrees with our predictions. We also add that according to our results the structure function does not show below threshold a sharp maximum around $Q_{\max}(t)$, as happens above it, but strictly it displays a certain multimodal nature, this fact being interpreted as a signature of the somewhat arbitrary variations in time and space of the pattern observed experimentally.

From a much more general perspective, these results seem to suggest the possibility of detecting either permanent or transient spatial patterns directly mediated by the slightly subcritical or supercritical thermal fluctuations always intrinsic to any realistic physical system experiencing a symmetry-breaking instability. The generality of this phenonenon extending to other situations different from the Fréedericksz transition in nematic liquid crystals, as, for example, electroconvection in anisotropic liquid crystals or thermal convection in simple or binary fluids, could be an interesting subject of further theoretical and experimental research for specialists in these fields.

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