Forbidden walls

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In this paper the presence of periodic twist-bend walls in the upper vicinity of the Frèedericksz critical point of a nematic calamitic sample is reported. According to the existing theory, the formation of these structures could not happen in this region of the magnetic field. The length of the periodicity of these walls as well as the time spent with their formation have been measured, and it was demonstrated that the coherent motion of the nematic material cannot drive their construction. We assume that the mechanism responsible for their appearance results from the magnification of some selected random fluctuations occurring at the neighborhood of the Frèedericksz critical point. [S1063-651X(99)02909-8]

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It is largely accepted that the coherent motion of matter can drive pattern formation in nonequilibrium hydrodynamic systems [1]. Therefore, when this coherent motion of matter goes to zero one expects that the formation of these patterns would become feeble and, in the limit, would disappear. In this paper an example of a system will be given where the pattern formation persists even when the coherent motion of matter can be neglected. For this purpose the periodic twist-bend walls [2,3] usually found in nematic liquid crystals (NLC) will be used.

Theoretical and experimental procedures have been used to investigate the formation of these structures [4–14]. They arise when an external magnetic field, perpendicular to the initial director direction and greater than the Frèedericksz threshold [15], is applied to a homogeneous aligned nematic sample subjected to strong anchoring boundary conditions. In this condition there is a competition between the magnetic susceptibility that tends to align the director along the magnetic-field direction, and the elastic energies, which tend to produce a director orientation consistent with its orientation at the surface of the sample. Due to this competition, and the π symmetry of the director, the nematic phase begins an unstable dynamical phase that produces the characteristic oscillatory patterns of the twist-bend walls.

It is well established that for high magnetic fields the internal motion of the nematic material, which starts at the moment at which the external magnetic field is turned on, has a decisive role in their construction This internal motion is responsible for their outstanding, one-dimensional, periodic character [12]. In fact, with the use of the anisotropic properties of NLC, Guyon *et al.* [4] and Lonberg *et al.* [6] have shown that the construction of the walls is possible because the coherent motion of the nematic material driving them has an effective viscosity, which is lower than the one resulting from the matter movement forming any other kind of pattern. In this paper the behavior of this selection mechanism will be investigated when the coherent motion of the matter goes to zero.

According to the existing theory [6], when the Frèedericksz threshold is approached—from the upper side—the coherent internal motion of the nematic material becomes smaller and smaller and there is a point h_w—greater than the Frèedericksz critical point h_f—below which it disappears. Between this point and the Frèedericksz threshold [8,9] the walls would be absent and the director would have a homogeneous alignment. Nevertheless, under experimental conditions, we have found that when the critical point is approached the emergence of these structures continues. Furthermore, the time spent in its construction is so long that the established picture must be abandoned. We conjecture that in the neighborhood of the Frèedericksz threshold there is a mechanism responsible for the selection of these patterns. It could result from the magnification of some random fluctuations occurring at the vicinity of the critical point. The detailed process by which this selection and magnification occurs deserves further investigation, but our theoretical and experimental results are sufficient to establish its existence.

Let us consider a NLC sample inside a microslide glass with the dimensions (a,b,d) such that $a \gg b \gg d$. An initial homogeneous orientation of the director along the x direction is obtained, and a fixed-strength-controlled magnetic field H (accuracy of about 2%) is applied along the y direction. It is assumed that the director evolves in the planar geometry given by $n_x = \cos\theta(x,y,z,t), n_y = \sin\theta(x,y,z,t)$, and $n_z = 0$, where $\theta(x,y,z,t)$ gives the instantaneous angle between the director $\vec{n}$ and the x direction. Strong boundary conditions are assumed at the borders of the sample [2]. The time evolution of this system is given by the so-called Eriksen-Leslie-Parodi (ELP) approach [16–18] in which the NLC motion is given by a set of differential equations composed by an anisotropic version of the Navier-Stokes equation [19,20], the balance of torques equation [2,21], and the equation of continuity. Furthermore, as our experiment is restricted to the neighborhood of the Fréedericksz critical point, the linear approximation of the ELP approach can be used. When high

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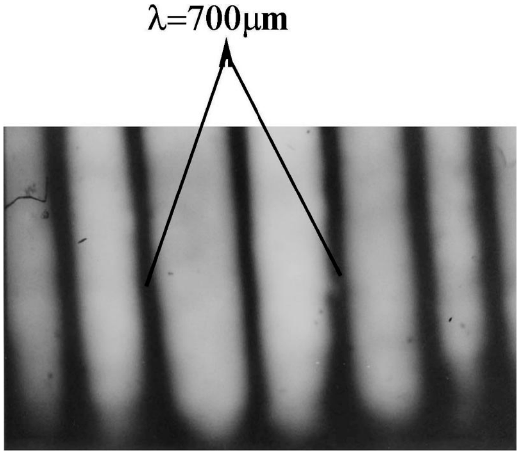

FIG. 1. Lyotropic nematic phase in 0.2-mm thick microslides between crossed polarizers. Magnetic field (2 kG) along the y axis at 0° of the light polarizing direction. The measured length of the periodicity of this wall was $\lambda = 700$ μm and only became perceptible after 36 hours of constant exposition to the magnetic field.

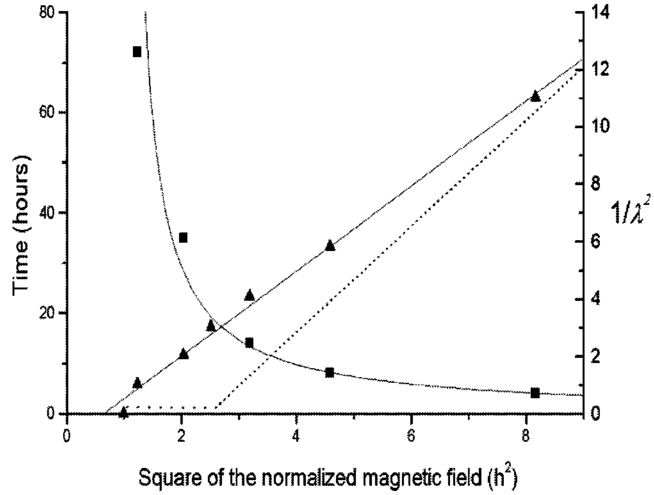

FIG. 2. Measured points of τ and $(2d/\lambda)^2$ versus h^2. The squares give the measured points for τ that are read at the left side. The triangles give $(1/\lambda)^2$ and are read at the right. The continuous line accompanying the squares and the triangles is only for guiding the eyes. The dotted line gives the curve along which are supposed be the experimental points of $(2d/\lambda)^2$ versus h^2. This curve would arrive at zero at $h^2 \approx 2.5$ and remain at zero until the point $h^2 = 1$. No such behavior was found in the experiment. The measured point goes directly to the point $h^2 = 1$, and no region with $(1/\lambda)^2 = 0$ was found. Observe that the first point only appears in the curve of k^2 vs h^2. For this point we do not find the formation of any kind of structure in the sample even after a *week* of continuous exposure to the magnetic field. As was demonstrated in our paper, when point h_w^2 is approached the bending of the director should become homogeneous, the walls would disappear, and the time spent with the construction of this homogeneous bending *must be finite*. We never saw a homogeneous bending of the director and, furthermore, the time spent for the walls' construction became infinite as the critical point was approximated.

magnetic fields are used this approach must be reconsidered and nonlinear modes must be taken into account [7].

Under these conditions the director acquires an oscillatory pattern along the x direction. Let λ be the wavelength of a component, this one-dimensional oscillatory pattern, and let $k = 2\pi/\lambda$ be the corresponding modulus of the wave vector. From the ELP approach [6], results of the time spent with the construction of this mode is given by

$$\tau = \frac{\gamma_1}{\chi_a H_c^2} \frac{1 - [k^2(\xi_1 - \xi_3)/(\pi/d)^2 \eta + k^2(\eta + \xi_1)]}{h^2 - 1 - \tilde{K}_{33} k^2}. \quad (1)$$

In these equations $h^2 = H^2/H_c^2$, $H_c^2 = (K_{11}(\pi/b)^2 + K_{22}(\pi/d)^2)$, $\tilde{K}_{33} = K_{33}/(\chi_a H_c^2)$; K_{11}, K_{22}, and K_{33} are the elastic constants, γ_1, ξ_1, ξ_3, and η are viscosity coefficients as defined by Vertogen and de Jeu [20], and χ_a is the magnetic susceptibility. Moreover, at $h^2 = h_f^2 = 1$ we have the Frèedericksz critical point. Observe that τ is strongly dependent on k^2, i.e., for each k^2 there will be a different time interval wasted with the building of the corresponding profile. The hypothesis of Lonberg *et al.* [6] is that the dominant pattern is the one that can be constructed in the shortest time (fastest mode). This results in a selection mechanism for the parameter k^2 that, through the minimization of τ, is given by the roots of the equation

$$\tilde{K}_{33}(\eta + \xi_1)(\eta + \xi_3)(k^2)^2 + 2\tilde{K}_{33}(\eta + \xi_1)\left(\frac{\pi}{d}\right)^2 \eta k^2 - (\xi_1 - \xi_3)\left(\frac{\pi}{d}\right)^2 \eta (h^2 - 1) + \tilde{K}_{33}\left(\frac{\pi}{d}\right)^4 \eta^2 = 0. \quad (2)$$

This selection mechanism has given good results for the regions far above the Fredericks threshold, but as can be easily verified [8,9] this equation predicts the existence of region $h_f^2 \leqslant h^2 \leqslant h_w^2$,

$$h_w^2 = 1 + \frac{\tilde{K}_{33}(\pi/d)^2 \eta}{\xi_1 - \xi_3}, \quad (3)$$

where the selected mode would be characterized by $k^2 = 0$, that is, the fastest mode would be the homogeneous bending of the director. Furthermore, it can also be shown that for this mode the coherent velocity of the nematic material would be zero.

In the experiment we looked for the outputs of the parameters k^2 and τ in the neighborhood of the Frèedericksz transition. It was used a nematic lyotropic mixture of potassium laurate (KL), potassium chloride (KCl), and water, in the nematic calamitic phase, with the respective concentrations in weight percentage: 34.5, 3.0, and 62.5. Nematic samples were encapsulated in a flat glass microslide (length $a = 20$ mm, width $b = 2.5$ mm, and thickness $d = 0.2$ mm) from Vitrodynamics. Figure 1 shows a periodic distortion of $\vec{n}$ with walls formed in the direction of $\vec{H}$ in a polarizing microscope. During the experiment the temperature was controlled at 25 ± 1 °C. According to the theory presented above we would expect from Eq. (2) that in the neighborhood of $h^2 \simeq h_w^2$,

$$k^2=\frac{\xi_1-\xi_3}{2\tilde{K}_{33}(\eta+\xi_1)}(h^2-h_w^2), \tag{4}$$

where h_w^2 was given in Eq. (3). At the same time as $k^2\to 0$ the time τ would converge to the value,

$$\tau_w=\frac{\gamma_1}{\chi_a H_c^2}\frac{1}{h_w^2-1}=\frac{\gamma_1}{\chi_a H_c^2}\frac{\xi_1-\xi_3}{\tilde{K}_{33}(\pi/d)^2\eta}=\frac{\gamma_1}{K_{33}}\left(\frac{d}{\pi}\right)^2\frac{\xi_1-\xi_3}{\eta}. \tag{5}$$

Therefore, according the above results, as $k^2\to 0$ it would be expected that a graph of k^2 vs h^2 would be a straight line converging to the point h_w^2. Below this point a homogeneous director bending would be found. In Fig. 2 the dotted line gives a picture of this supposed result. The results of our measurements are also shown. The surprising result is that the walls always exist and a region with homogeneous alignment was never found [22].

Furthermore, a graph of τ vs h^2 would converge as $k^2\to 0$, to the point τ_w calculated above, which is clearly a finite time interval. But, according to our experimental results the time spent with the formation of these structures diverges as the point at which $k^2\to 0$ is approached. We have obtained data so close to this critical point that the corresponding walls only appeared after *two* or *three days* of continuum exposition to the magnetic field. Observe that the first point only appears in the curve of k^2 vs h^2. At this point we did not find the formation of any kind of structure in the sample even after a *week* of continuous exposure to the magnetic field. Consequently, we were not approaching the point $h^2=h_w^2$. Furthermore, no signal of any kind of homogeneous alignment was found at this point, or below it.

Therefore, it is experimentally evident that the Fréedericksz threshold must be above this point and we are forced to conclude that the two curves are not approaching the point $h^2=h_w^2$, but the Frèedericksz critical point $h^2=1$. That is, the formation of walls in the upper neighborhood of the Frèedericksz transition has been detected in a region where, according to the existing theory, these structures would not exist. We have never seen a homogeneous bending of the director and, furthermore, the time taken with the walls' construction became infinite as the critical point was approximated. Furthermore, the final wave number presented by the observed patterns is not the one obtained by the fastest mode of Eq. (15). Indeed, the fastest mode is not even an approximation of the observed result, simply because it predicts the absence of patterns. Our measurement of the wavelength and the time spent with the formation of these objects indicate that these walls are built by the system as soon as the Frèedericksz threshold is exceeded. Moreover, the Lonberg model cannot explain the long time demanded by their construction. Indeed, it is impossible to conceive that there is a coherent motion of matter lasting three days in a viscous medium where the only external force is due to a constant magnetic field. In the neighborhood of the Frèedericksz threshold the time spent with the constructions of the walls becomes so long that the viscosity of the system would damp any coherent motion of mater. For this reason, we assume that the big random fluctuations existing in the neighborhood of the critical point [23] are driving the birth and selection of the observed structures.

If this mechanism of the construction of walls via the random fluctuations happening at the neighborhood of the Fréedericksz critical point in fact occurs, an observable consequence would be immediate. It is known that the usual walls—the ones that are built far from the Fréedericksz threshold—are unstable structures. That is, once created they decay and lose their periodicity and one dimensionality. It has been demonstrated that this happens because as soon as the fluid motion that gives rise to them stops, they become a local maximum of the static free energy [14]. However, the random fluctuations giving rise to the structures here described never stop. Therefore, these walls would be stable structures and would never decay. We have observed that these structures remain practically unaltered even after a week of its birth. This is strong evidence that they are really stable. But, due to the long time usually spent by the physics of these structures, new measurements must be done and analyzed.

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