

Fate of magnetic walls in nematic liquid crystals

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The origin of the instability that leads to the disappearance of the one-dimensional periodic walls formed above Fredericksz's threshold of some nematic liquid crystals is investigated. It is shown that these walls are built in a configuration that—even being an extreme of the elastic free energy—is not a local minimum, but a local maximum. The mechanism that gives rise to this instability is investigated, and it is shown how the director's fluctuations lead to the destruction of the walls. [S1063-651X(98)07308-5]

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I. INTRODUCTION

For a broad class of oriented nematic liquid crystals (NLC), with positive diamagnetic anisotropy, the action of an external magnetic field can lead to the formation of one-dimensional periodic structures (walls) above the Frèedericksz threshold [1,2]. This phenomenon has been the subject of several works dealing with its theoretical and experimental aspects [3,4]. Since the pioneering work of Lonberg *et al.*, there has been a more or less well-established framework to interpret the main mechanism of the *formation* of these structures. Recently, this subject has achieved a renewed interest because there are some open problems concerning the fate, i.e., the destination of these periodic structures. It is well known that for values of the applied magnetic field far above the Frèedericksz threshold, the walls tend to disappear after some time [5,6]. The basic mechanism for the destruction of these walls (henceforth denoted by MW) is the instability beginning at the moment in which the flux of matter [3] that gives rise to these structures stops. In this moment the extremely harmonic and periodic walls pattern begins an unstable phase in which all its one-dimensional and periodic regularity is lost.

Habitually unstable configurations are found in physical systems after the action of some transient force operating during a finite time interval [7,8]. While the transient action is working, the system goes to a configuration that, as soon as it vanishes, no longer has the least energy. In the case of the building of MW, the transient force is given by the flux of the nematic material inside the sample [3], which is also responsible, for example, for the system regularity and dimensionality [9]. In a remarkable work, Lonberg *et al.* [3] have shown how this mechanism works: the external magnetic field rotates the director, which, in turn, stimulates a fluid flow generating a nonuniform rotation pattern of the director. This rotation reinforces opposite rotations of neighboring regions of the sample. This fluid flow process has been confirmed by many experimental and theoretical studies [10]. Moreover, it has been shown that as soon as the director reaches its maximum bending, the velocity of the fluid

flow becomes negligible and disappears. Of course, the fact that the walls do not have the least energy is a necessary condition, but not sufficient, to promote its instability. Our aim in this work is to grasp the mechanisms by which these structures begin their decay and for this purpose some mathematical analysis must necessarily be done. This does not mean that the understanding of the fate of the walls cannot be put in physical terms. In fact, the fluid flow leads the system to a configuration that is an extremum of the energy but, as a simple elastic argument shows, it is not the configuration with the least energy. But even not being in the ground state, the walls could be in a local minimum at which the system might remain indefinitely. We will show that, as the experiments confirm, the MW configuration is indeed a local maximum. In order to prove it, the second functional derivative of the free energy around the walls' configuration must be studied. As in the differential calculus, the system will be in an unstable configuration if this second functional derivative is negative. One appropriate tool for deciding the sign of the second functional derivative is the Jacobi criterion [11], and through it we will show that around the walls the second functional derivative is really negative. An important aspect of this demonstration is that nothing more than the oscillatory and one-dimensional character of the MW must be assumed. Therefore, these walls' characteristics must be at the core of the reasons for their instability.

Another contribution of this work is the understanding of the mechanism of decay of the walls. It will be shown that the fluctuations responsible for the walls' decay are localized in the regions where the director, even in the presence of the external magnetic field, does not turn at all. Along these regions—which are the site of the walls' nodes—there is a critical balance between the action of the external magnetic field that tries to turn the director direction and the elastic energy that, due to the opposite configuration of the director in the neighbor regions, tries to maintain the director with a null bending. As we will demonstrate, the instability only appears due to the weakness of this balance. Around the points of null bending, the fluctuations will increase exponentially and destroy the walls' regularity.

This paper is organized as follows. Section II is dedicated to the presentation of the basic equations of the theoretical approach (the mathematical details are left to the Appendix). It is shown that, although it is at an extreme of the elastic free energy, the system is at an unstable configuration be-

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cause it is in a local maximum. In Sec. III, we present the main results of our calculations, with emphasis on the mechanism by which this instability begins the walls' decay. We also discuss how any small fluctuation around the equilibrium position of the MW gives rise to an irreversible exponential growing process of destruction of the parameters that characterizes the walls. In Sec. IV, some concluding remarks are drawn.

II. FUNDAMENTALS

The aim of this section is to show that the transient that takes the system to an unstable configuration is the coherent motion of the nematic material, and that the structures built in this way do not possess the smallest possible energy, being a local maximum. In order to prove this statement, it is assumed that the NLC can be described by the so-called Eriksen-Leslie-Parodi approach [12–16] and it is considered a slab with dimensions a along the x axis, b along the y axis, and d along the z axis such that $a \gg b \gg d$. The system is previously prepared to be uniformly aligned along the x direction. After that, an external controlled magnetic field H , greater than the Fredericks threshold, is applied along the y direction [17]. To describe the textures produced in the nematic material, the components of the director are expressed by the planar geometry:

$$n_x = \cos\theta(x, y, z), \quad n_y = \sin\theta(x, y, z), \quad n_z = 0, \quad (2.1)$$

where $\theta(x, y, z)$ is the angle between the director \vec{n} and the x direction. The expression of the total free energy in the two elastic constant approximation ($K_{11} = K_{33}$), by taking into account the magnetic field coupling, is [13,1,5]

$$F = \int_V \left\{ \frac{1}{2} K_{33} [(\partial_x \theta)^2 + (\partial_y \theta)^2] + \frac{1}{2} K_{22} (\partial_z \theta)^2 - \frac{1}{2} \chi_a H^2 n_y^2 \right\} dV, \quad (2.2)$$

where K_{11} , K_{22} , and K_{33} are the elastic constants of splay, twist, and bend, respectively, and V is the volume of the sample.

The motion of nematic fluid is described through the anisotropic version of the Navier-Stokes's equation

$$\rho \left(\frac{\partial \mathcal{V}_\alpha}{\partial t} + \mathcal{V}_\beta \frac{\partial \mathcal{V}_\alpha}{\partial x_\beta} \right) = \frac{\partial}{\partial x_\beta} (-p \delta_{\alpha\beta} + \sigma_{\beta\alpha}), \quad (2.3)$$

where ρ is the density of the system, \mathcal{V}_α is the α component of the velocity, p is the pressure, and $\sigma_{\beta\alpha}$ is the associated anisotropic stress tensor [1,18,19], which is dependent on the velocity of the fluid $\vec{\mathcal{V}}$, on the bending of the director θ , and on its time variation rate $\dot{\theta}$.

In the geometry fixed above, the equation of motion of the director (balance of torque equation) assumes the form [9]

$$\begin{aligned} \gamma_1 \partial_t \theta = & \gamma_1 W_{xy} - \gamma_2 [A_{xy}(n_x^2 - n_y^2) + (A_{yy} - A_{xx})n_x n_y] \\ & + K_{33} [\partial_x^2 \theta + \partial_y^2 \theta] + K_{22} \partial_z^2 \theta + \chi_a H^2 n_x n_y, \end{aligned} \quad (2.4)$$

where the inertial terms were not considered, γ_1 and γ_2 are the shear torque coefficients, $A_{\alpha\beta} = \frac{1}{2}(\partial_\alpha \mathcal{V}_\beta + \partial_\beta \mathcal{V}_\alpha)$, $W_{\alpha\beta} = \frac{1}{2}(\partial_\alpha \mathcal{V}_\beta - \partial_\beta \mathcal{V}_\alpha)$, and, as usual, the fluid is considered incompressible. Since the viscosity tensor σ_{ij} , appearing in Eq. (2.3), and the coefficients of Eq. (2.4) depend on θ [1,18,19] through combinations of n_x , n_y , and its powers, these two differential equations result in a set of coupled, and strongly nonlinear, equations. Of course, it seems impossible to find any analytical solution of this problem with the correct boundary conditions. Srajer *et al.* [10] studied a numerical solution of these equations, using the parameters of the poly- γ -benzil-glutamate (PBG). From the results of their paper, it is possible to see that after some minutes the bending of the director reaches its maximum value (i.e., $\partial_t \theta = 0$) and the matter flux flow becomes zero (i.e., $\vec{\mathcal{V}} = 0$). By substituting this condition in Eq. (2.3) and Eq. (2.4) one discovers that Eq. (2.3) becomes identically zero, and Eq. (2.4) results in

$$K_{33} [\partial_x^2 \theta + \partial_y^2 \theta] + K_{22} \partial_z^2 \theta + \chi_a H^2 n_x n_y = 0, \quad (2.5)$$

which is the equation governing the steady state of the walls.

Equation (2.5) describes the configuration assumed by the system after the coherent movement of the nematic fluid has ceased. It is also the equation that gives us the extrema of the Frank free energy, Eq. (2.2). Therefore, as we have pointed out, all that the dynamics of the flux flow permit us to conclude is that the flux brings the system to a configuration that corresponds to an extremum of the elastic energy of the problem. But the configuration built in this way is not static. After some time—in many cases, several hours—these one-dimensional walls disappear. A typical lyotropic sample consists of potassium laurate (KL, 34.5), potassium chloride (KCl, 3.0), and water (62.5) [17]. The concentrations are indicated in weight percent. The system is in the nematic calamitic phase (N_c) at room temperature. The method of generating the periodic distortion of the director consists in orienting an (N_c) sample in a planar geometry, with a magnetic field ($H \approx 10$ kG along the x axis). After a well-oriented sample is achieved, the field is applied along the y axis. In Fig. 1, three different moments of the walls' collapse in a NLC sample are shown. In the first photo, one sees the walls as they appear as soon as the coherent movement of the nematic fluid finishes. In the second photo, the collapse has started and the walls begin to lose their regularity. In the last photo, the one-dimensional and periodic walls no longer exist and all that remains are vestiges of the original structure.

This decay can be understood only if the elastic energy of MW is not the lower energy allowed in the system. This is indeed true [20] and a simple comparison between the two configurations (with walls and without them) can tell us that the presence of the walls puts the system out of its ground state. But it is not sufficient to show that this new state is an unstable one. However, if we could show that the walls are in a local maximum of the free energy, we could be sure of the instability, because any small fluctuation will lead the

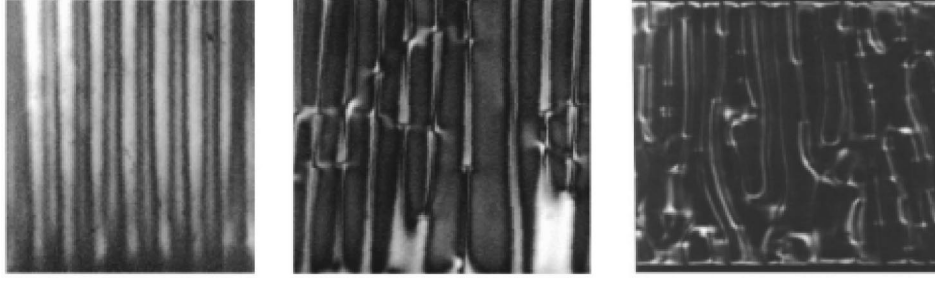


FIG. 1. Lyotropic calamitic nematic phase in microslide 200 μm thick between crossed polarizers in the presence of magnetic field $H \sim 3$ kG Walls parallel to \vec{H} : (a) typical periodic distortions; (b) the walls begin to lose their regularity, after a time of exposure to the magnetic field of 15 h; (c) the one-dimensional and periodic walls no longer exist and all that remains are vestiges of the original structures (after 40 h).

system to a new configuration. The main objective of this work is just to show that this is in fact the case.

Since the formal proof of this fact requires extensive mathematics, it was placed in the Appendix, where it is proved that the walls' configuration is indeed a local maximum of the free energy. Nevertheless, the idea behind the proof is quite simple. The second functional derivative of the free energy around the walls' configuration is computed and it is shown that such a derivative is negative. Therefore, this cannot correspond to a stable configuration because even the natural fluctuations of the system are enough to lead it to another configuration.

III. THE COLLAPSE OF THE WALLS

We have shown in the Appendix that the walls' configuration is unstable because there are fluctuations $\delta\eta$ that make the quadratic form $\delta^2 F$, given in Eq. (A6), negative. However, no indication about the dynamics of this collapse was made nor any trace of the kind of fluctuations that destroy the walls' regularity was found. The study of how we can find these destroyer fluctuations and extract from them their meaning is the aim of this section. The key of our search will be the use of the fact that Eq. (A6) is a quadratic form. Therefore we can diagonalize it and its negative eigenvalues will correspond to the normal modes of the fluctuation that erodes the walls.

Let us consider Eq. (A6), which, after an integration by parts, can be written as

$$\delta^2 F = \int_0^L dx \delta\eta \{ -\tilde{K}_{33} \partial_x^2 + [1 - 2h^2 u''(\eta)] \} \delta\eta. \quad (3.1)$$

By expanding $\delta\eta$ in the form $\delta\eta = \sum_{n=1}^{\infty} c_n \delta\eta^n$, where $\delta\eta^n$ belongs to a complete set of functions in which each $\delta\eta^n$ is a solution of the Sturm-Liouville problem [21],

$$-\tilde{K}_{33} \partial_x^2 (\delta\eta^n) + [1 - 2h^2 u''(\eta)] \delta\eta^n = \epsilon_n \delta\eta^n, \quad (3.2)$$

Eq. (3.1) becomes

$$\delta^2 F = \sum_{n=1}^{\infty} \epsilon_n (c_n)^2. \quad (3.3)$$

As it has been proved that there are fluctuations for which $\delta^2 F < 0$, there must be some ϵ_n , at least one, for which

$\epsilon_n < 0$ and these ones are the eigenvalues responsible for the walls' destruction. There is a simple way to understand the origin of these negative eigenvalues. Notice that by performing the association $\tilde{K}_{33} \rightarrow \hbar^2/2m$, Eq. (3.2) becomes a Schrödinger-like equation for a wave function represented by $\delta\eta^n$, in a potential given by

$$U(x) = 1 - 2h^2 u''(\eta(x)). \quad (3.4)$$

Using this association it becomes easy to understand that the negative eigenvalues of Eq. (3.2) can only exist around the regions where the potential $U(x)$ is negative. It will be shown [see Eq. (3.8) and subsequent discussion] that these points can occur only at the points where $\eta \approx 0$ and this will prove that the regions where the director does not bend at all are the ones responsible for the walls' decadence. Furthermore, as this null bend of the director only exists due to its oscillatory character, one concludes again that the director periodic profile is one of the reasons for the collapse of the walls.

To see the effect of these negative eigenvalues in the time development of the system, it is enough to take into account Eq. (2.4) and to observe that, as soon as the fluid flow stops, it becomes

$$\tilde{\gamma}_1 \partial_t \eta = \tilde{K}_{33} \partial_x^2 \eta - \eta + 2h^2 u'(\eta), \quad (3.5)$$

where $\tilde{\gamma}_1 = 4\gamma_1/bd\chi_a H$. Now, if we set $\eta = \eta_0 + \delta\eta$, where η_0 is a solution of $\delta F = 0$, one obtains for $\delta\eta$ the differential equation

$$\tilde{\gamma}_1 \partial_t \delta\eta = \tilde{K}_{33} \partial_x^2 \delta\eta - [1 - 2h^2 u''(\eta)] \delta\eta, \quad (3.6)$$

which governs the time behavior of the fluctuation. As is usual for the statistical fluctuations [22,23], this equation is analogous to the time-dependent Schrödinger equation. Obviously, this is only a formal analogy and it does not mean that the nematic structure has something to do, at least directly, with quantum mechanics. Using the expansion of $\delta\eta$ in its normal components, Eq. (3.6) becomes a set of equations with the form

$$\tilde{\gamma}_1 \partial_t c_n = -\epsilon_n c_n, \quad (3.7)$$

which, once ϵ_n are found, have a trivial solution. The important aspect of these equations is that all the fluctuations $\delta\eta^n$,

with positive eigenvalues ϵ_n , are exponentially suppressed, while the ones with negative eigenvalues will grow exponentially.

We use now the formal analogy between the time development of the fluctuations and the time-dependent Schrödinger equation to obtain a glimpse of how these negative modes start the MW destruction. In the expansion of the potential $U(x)$, Eq. (3.4), around the points where $\eta \approx 0$, one obtains

$$U(x) = 1 - h^2 + \frac{9}{8} h^2 [\eta'(0)]^2 x^2, \quad (3.8)$$

where it was used $\eta(x) \approx x \eta'(0)$. Equation (3.8) shows that in the regions around the null bending of the director, the potential $U(x)$ is really negative ($h^2 > 1$ and $x^2 \ll 1$) and can be approximated by a parabolic well. Putting this potential in Eq. (3.2), and remembering that this equation is a Schrödinger-like equation, we see that its most negative eigenvalue is nothing more than the ground state of a quantum oscillator. If one again identifies $\tilde{K}_{33} \rightarrow \hbar^2/2m$, and $9/8[\eta'(0)]^2 \rightarrow 1/2m\omega^2$, one obtains for the ground state of Eq. (3.2)

$$\epsilon_0 = 1 - h^2 + \frac{3}{4} h \sqrt{2\tilde{K}_{33}} \eta'(0), \quad (3.9)$$

which will be the leading term of the walls' destruction.

In this term it can be observed that $n'(0)$ is the parameter of the wall that determines the magnitude of the ground state ϵ_0 . The smaller $n'(0)$ is, the more negative ϵ_0 will be and the greater the fluctuations will be. Since by Eq. (3.7) these fluctuations must grow with time, one concludes that $n'(0)$ will become smaller and smaller with time. This fact can also be understood by observing that ϵ_0 is the energy of the fluctuations at the well given in Eq. (3.8) and, therefore, the least energy will have the smaller $n'(0)$. In other words, if one computes the generalized force associated with the variable $n'(0)$, $f = -\partial\epsilon_0/\partial n'(0)$, one obtains $f = -\frac{3}{4} h \sqrt{2\tilde{K}_{33}}$, which gives the same result: since the force is negative, the time evolution of the system will diminish $n'(0)$.

In order to fully appreciate the details of the walls' wasting process, we construct a wall profile in which its geometrical parameters are made clear and workable. Of course it is not the exact solution of Eq. (A3), but it results from a numerical study of the solution [17,24] that is useful only as long as it can help us to understand the fate of the walls. This wall profile is described by three parameters: the walls' amplitude φ_0 , the walls' length λ , and the walls' form factor δ , which are shown in Fig. 2. An analytical form for this wall is given by

$$\eta(x) = \varphi_0 \varphi(x), \quad (3.10)$$

where it is assumed that

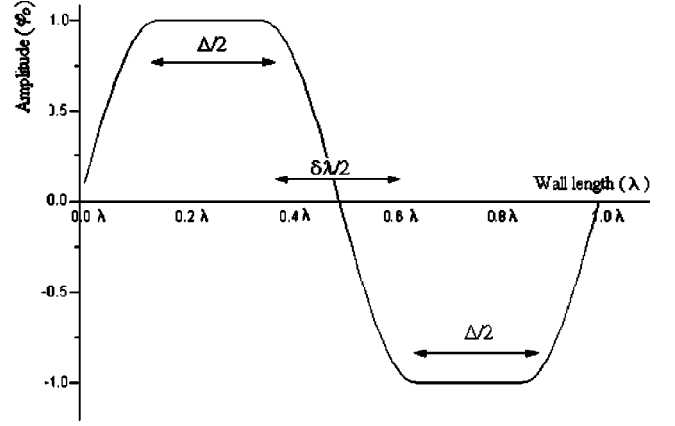


FIG. 2. Graphic representation of a typical wall. It is explicitly shown the saturated portion $\Delta/2$, the bending portion $\delta\lambda/2$, and the amplitude φ_0 . The irreversible exponential growing fluctuations act in such a way to reduce the amplitude φ_0 and the saturated portion Δ .

$$\varphi(x) = \begin{cases} \sin\left(\frac{2\pi}{w}x\right), & 0 < x < \frac{1}{4}w, \\ 1, & \frac{1}{4}w < x < \frac{\lambda}{2} - \frac{1}{4}w, \\ \sin\left(\frac{2\pi}{w}\left(\frac{\lambda}{2} - x\right)\right), & \frac{\lambda}{2} - \frac{1}{4}w < x < \frac{\lambda}{2}, \end{cases} \quad (3.11)$$

$$\varphi\left(\frac{\lambda}{2} + \epsilon\right) = -\varphi\left(\frac{\lambda}{2} - \epsilon\right), \quad 0 < \epsilon < \frac{\lambda}{2}, \quad w = \delta\lambda,$$

where δ , changing between 0 and 1, controls the wall form (Fig. 2). The MW described by $\varphi(x)$ has two distinct regions. In one of them (the w region) the director bends its orientation from one configuration to the symmetric one. The other region (the Δ region) describes a saturated portion of the director. The δ value gives the fraction of each portion. When $\delta \rightarrow 1$, the wall becomes a single sine function. On the other hand, when $\delta \rightarrow 0$ the saturated region assumes the entire wall. One observes from Fig. 2 that

$$\Delta = \lambda - w = \lambda(1 - \delta) \quad (3.12)$$

is a measure of the saturated portion of the wall. Furthermore, Eq. (A3) has a conserved quantity [17,24,25]

$$C = \frac{1}{2} \tilde{K}_{33} (\partial_x \eta)^2 - \frac{1}{2} \eta^2 + 2h^2 u(\eta), \quad (3.13)$$

which reflects the homogeneity of the system along the x direction and is a fixed number that exists only as long as the system remains one-dimensional. Therefore, its value at the point where $\eta = 0$ can be compared with its value at the region where $\partial_x \eta = 0$, giving

$$\frac{1}{2} \tilde{K}_{33} \varphi_0^2 \left(\frac{2\pi}{\lambda - \Delta}\right)^2 = -\frac{1}{2} \varphi_0^2 + 2h^2 u(\varphi_0), \quad (3.14)$$

which shows that as long as the system is one-dimensional the parameters λ , Δ , and φ_0 are not independent.

As we have already remarked, the cornerstone of the instability is the one-dimensional and oscillatory character of the walls. Therefore, the starting point of the walls' decay is strongly connected to the breakdown of this character. The system has to abandon the one-dimensionality. It has been demonstrated, in Eq. (3.13), that the one-dimensional character of the walls leads to the conservation of a quantity that, in Eq. (3.14), connects their geometrical parameters. The collapse of the walls begins with the destruction of this connection. This means that, if the system tries to be non-one-dimensional these parameters must evolve independently. Let us show now in what manner this happens.

By using the value of $n'(0)$, obtained from Eq. (3.11), and by putting it in Eq. (3.9), one obtains

$$\epsilon_0 = 1 - h^2 + \frac{3}{4} \varphi_0 \left(\frac{2\pi}{\lambda - \Delta} \right) h \sqrt{2\tilde{K}_{33}}. \quad (3.15)$$

As stressed above, when the collapse of the walls starts, the connection between their geometrical parameters, given by Eq. (3.14), is lost. The minimization of the energy of the fluctuations will lead to an independent development of λ , Δ , and φ_0 . For example, from Eq. (3.15) it is easy to obtain the force $f = -\partial\epsilon_0/\partial\lambda$ between two neighbor walls,

$$f = \frac{3}{2} \pi \varphi_0 \left(\frac{1}{\lambda - \Delta} \right)^2 h \sqrt{2\tilde{K}_{33}}, \quad (3.16)$$

which is repulsive and decays with the inverse of the square of the portion of the wall, $w = \lambda - \Delta$, where there is the director bending—see Fig. 2. That is, the shorter the bend portion is, the more repulsive is the force between the wall. This can also be seen in Eq. (3.15) because the shorter Δ is, the shorter will be the energy stored in the fluctuation. Finally, the minimization of the energy ϵ_0 led us to a similar conclusion for φ_0 : the shorter φ_0 is, the shorter will be ϵ_0 .

Summarizing, the fluctuations lead to the destruction of the walls' regular pattern by means of three mechanisms: (a) a repulsive interaction; (b) the reduction of the walls' saturated portion; (c) the reduction of the walls' amplitude.

Since the sample has a large number of equally spaced walls, the repulsive force between them may be counterbalanced and the net result may be an equilibrium situation. However, there is no way to get a counterbalanced effect in the reduction of the walls saturated portion Δ or in the reduction of the walls amplitude φ_0 . Therefore the reduction in Δ and φ_0 starts the destruction of the walls.

IV. CONCLUSION

We have used the Jacobi theorem about quadratic forms to demonstrate that, due to the matter flux, the one-dimensional periodic walls are built in a configuration that, even being an extremum of the Frank free energy, is not a local minimum but a local maximum of it. It was shown how the fluctuations, localized at the positions where the director even in the magnetic field presence does not bend at all, lead to the destruction of the walls' regularity. A simplified model was constructed for these walls and it was shown how their geometrical parameters start an independent evolution to the walls' end.

To arrive at these conclusions, the experimental fact that the walls are one-dimensional has been used. Nevertheless, the fluctuations need not be one dimensional. Indeed, a really quantitative treatment of this problem has to take into account explicitly the time evolution of the fluctuating parameters characterizing the walls' end as a three-dimensional problem. Of course this would result in a very difficult problem. However, in the approach presented above the fluctuations were treated as a one-dimensional problem and the result obtained makes sense on physical grounds. The essence of this result is that the fluctuations that destroy the walls are the ones localized at the places where the director does not bend at all.

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APPENDIX

We begin by remembering that it is an experimental and theoretically well established fact [3,9] that the walls arise as a one-dimensional and period structure. Consequently, we assume that the director profile $\theta(x,y,z)$ can be approximated by

$$\theta(x,y,z) = \eta(x) \sin\left(\frac{\pi y}{b}\right) \sin\left(\frac{\pi z}{d}\right), \quad (A1)$$

where $\eta(x)$ describes the bending of the director along the x direction. Along the y and z directions we suppose that the director assumes the simplest possible profile form, $\sin(\pi y/b)\sin(\pi z/d)$. This was done to make easy further calculations and, mainly, to clearly explore the one-dimensional character of the walls. Anyway, if other reasonable forms for the bending of the director along these directions are assumed, our results will not change because, as it will be seen, the core of our conclusions lies in the oscillatory character of $\eta(x)$.

By using Eq. (A1) and assuming that $\chi_a H_c^2 = K_{33}(\pi/b)^2 + K_{22}(\pi/d)^2$, and $h = H/H_c$, Eq. (2.2) can be set in the form

$$F = \frac{1}{4} b d \chi_a H_c^2 \int_0^a \mathcal{F} dx,$$

where $(A2)$

$$\mathcal{F} = \frac{1}{2} \tilde{K}_{33} (\partial_x \eta)^2 + \frac{1}{2} \eta^2 - 2h^2 u(\eta),$$

$u(\eta(x)) \equiv \int_0^1 \int_0^1 d\tilde{y} d\tilde{z} \sin^2(\eta(x) \sin(\pi\tilde{y}) \sin(\pi\tilde{z}))$, $\tilde{K}_{33} = K_{33}/(\chi_a H_c^2)$, and a is the sample length along the x direction.

Equation (A2) is the one-dimensional version of Eq. (2.2) and the corresponding Euler-Lagrange equation is given by

$$\tilde{K}_{33}\partial_x^2\eta_0 - \eta_0 + 2h^2u'(\eta_0) = 0, \quad (\text{A3})$$

which has to be solved for the strong anchoring conditions $\eta(0) = \eta(a) = 0$.

Let us now consider a free-energy fluctuation around the walls described by Eq. (A3) in the form

$$F = F_0 + \frac{1}{4}bd\chi_a H_c^2 \left\{ \delta F + \frac{1}{2}\delta^2 F \right\}, \quad (\text{A4})$$

where

$$\delta F = \int_0^L dx \{ [\eta - 2h^2u'(\eta)]\delta\eta + \tilde{K}_{33}(\partial_x\eta)\delta(\partial_x\eta) \} \quad (\text{A5})$$

and

$$\delta^2 F = \int_0^L dx \{ \tilde{K}_{33}(\partial_x\delta\eta)^2 + [1 - 2h^2u''(\eta)](\delta\eta)^2 \}, \quad (\text{A6})$$

with $u'(\eta) = du/d\eta$, $u''(\eta) = d^2u/d\eta^2$. The extremum F_0 is obtained by imposing $\delta F = 0$, which leads to Eq. (A3). Therefore, any small fluctuation around these configurations will lead to a change in the elastic energy of the form

$$F = F_0 + \frac{1}{8}bd\chi_a H_c^2 \delta^2 F. \quad (\text{A7})$$

It follows from this equation that a necessary and sufficient condition for the energy of the walls sequence F_0 to be a local minimum is that $\delta^2 F \geq 0$. It will be shown now that there are fluctuations $\delta\eta$ for which $\delta^2 F < 0$, thus indicating that the MW configuration is an unstable one.

To demonstrate that $\delta^2 F$ is indeed negative, we use the following (Jacobi) theorem [11]:

The quadratic functional

$$\int_{a_1}^{a_2} \{ P(x)(\partial_x\eta)^2 + U(x)\eta^2 \} dx, \quad (\text{A8})$$

where $P(x) > 0$, $a_1 < x < a_2$, is positive definite for all $\eta(x)$ such that $\eta(a_1) = \eta(a_2) = 0$ if and only if the solution of the differential equation

$$-\frac{d}{dx} \left(P(x) \frac{dj(x)}{dx} \right) + U(x)j(x) = 0, \quad (\text{A9})$$

with the boundary conditions $j(a_1) = 0$, $dJ(x)/dx|_{x=a_1} = 1$, does not vanish in the interval $a_1 < x < a_2$. The point c , $a_1 < c < a_2$, at which $j(c) = 0$, is said to be a conjugate point of the point a_1 , and $j(x)$ is known as the Jacobi field of the quadratic form given in Eq. (A8).

Therefore, if we intend to show that the quadratic form $\delta^2 F$ given in Eq. (A6) is negative, in the interval $0 < x < a$, all that we have to do is to show that its associated Jacobi field $j(x)$, which is the solution of the differential equation

$$-\tilde{K}_{33} \frac{d^2j(x)}{dx^2} + [1 - 2h^2u''(\eta)]j(x) = 0 \quad (\text{A10})$$

subjected to the boundary conditions stated above, has at least one conjugate point in the interval $0 < x < a$.

To demonstrate that there are indeed conjugate points in the interval $0 < x < a$, we observe that the function $\eta'_0(x) \equiv \partial_x[\eta_0(x)]$ also satisfies the differential equation given by Eq. (A10) [it is enough to differentiate Eq. (A3) with respect to x]. But $\eta'_0(x)$ is not a Jacobi field because it does not satisfy the appropriate boundary conditions. For example, $\eta'_0(0) \neq 0$. As $j(x)$ and $\eta'_0(x)$ satisfy the same differential equation, the Wronskian $W(x)$ of these two solutions is a constant, i.e.,

$$W(x) = j(x)\eta''_0(x) - \eta'_0(x)j'(x) = cte. \quad (\text{A11})$$

Using this equation and the fact that along one period there will be necessarily two consecutive points, x_1 and x_2 , where $\eta'_0(x)$ is zero, we find

$$\eta''_0(x_1)j(x_1) = \eta''_0(x_2)j(x_2) \quad (\text{A12})$$

but, as x_1 and x_2 are the extremum of $\eta_0(x)$, in a period the sign of $\eta''_0(x_1)$ is necessarily the opposite of $\eta''_0(x_2)$. Therefore, $j(x)$ changes sign between the two consecutive turning points x_1 and x_2 , which lead us to the conclusion that there must exist in this interval a point c for which $j(c) = 0$. Consequently, by the Jacobi theorem we have $\delta^2 F < 0$, and the configuration $\eta_0(x)$ is necessarily unstable.

[1] P. G. de Gennes, *The Physics of Liquid Crystals*, 2nd ed. (Clarendon Press, Oxford, 1993).
 [2] V. Fredericksz and V. Zolina, *Trans. Faraday Soc.* **29**, 919 (1933).
 [3] F. Lonberg, S. Fraden, A. J. Hurd, and R. B. Meyer, *Phys. Rev. Lett.* **52**, 1903 (1984).
 [4] E. Guyon, R. Meyer, and J. Salan, *Mol. Cryst. Liq. Cryst.* **54**, 261 (1979).
 [5] F. Brochard, *J. Phys. (Paris)* **33**, 607 (1972).
 [6] L. Léger, *Mol. Cryst. Liq. Cryst.* **24**, 33 (1973).
 [7] R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II, Nonequilibrium Statistical Mechanics* (Springer-Verlag, 1991).

[8] D. Chandler, *Introduction to Modern Statistical Mechanics* (Oxford University Press, New York, 1987).
 [9] M. Simões, *Phys. Rev. E* **56**, 3061 (1997).
 [10] C. Srajer, S. Fraden, and R. B. Meyer, *Phys. Rev. A* **39**, 9 (1989).
 [11] I. M. Gelfand and S. V. Fomin, *Calculus of Variations* (Prentice-Hall, New Jersey, 1963).
 [12] W. C. Oseen, *Trans. Faraday Soc.* **29**, 883 (1933).
 [13] F. C. Frank, *Discuss. Faraday Soc.* **25**, 19 (1958).
 [14] J. L. Ericksen, *Arch. Ration. Mech. Anal.* **4**, 231 (1960); **9**, 371 (1962).
 [15] F. M. Leslie, *Q. J. Mech. Appl. Math.* **19**, 357 (1966).
 [16] O. Parodi, *J. Phys. (Paris)* **31**, 581 (1970).

- [17] A. J. Palangana, M. Simões, L. R. Evangelista, and A. A. Arroteia, *Phys. Rev. E* **56**, 4282 (1997).
- [18] W. H. de Jeu, *Physical Properties of Liquid Crystalline Materials* (Gordon and Breach, New York, 1979).
- [19] G. Vertogen and W. H. de Jeu, *Thermotropic Liquid Crystals, Fundamentals* (Springer-Verlag, Berlin, 1988).
- [20] E. A. Oliveira, G. Barbero, and A. M. Figueiredo, *Phys. Rev. E* **54**, 5830 (1996).
- [21] F. W. Byron and R. W. Fuller, *Mathematics of Classical and Quantum Physics* (Dover Publications, Inc., New York, 1992).
- [22] R. F. Feynman and R. H. Hibbs, *Quantum Mechanics and Path Integrals* (McGraw-Hill, 1965).
- [23] R. F. Feynman, *Statistical Mechanics* (Springer-Verlag, Heidelberg, 1988).
- [24] M. Simões, A. J. Palangana, and L. R. Evangelista, *Phys. Rev. E* **54**, 3765 (1996).
- [25] M. Simões, A. J. Palangana, and L. R. Evangelista, *Phys. Rev. E* **56**, 7326 (1997).