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M. Simões<sup>a</sup> & A. J. Palangana<sup>a</sup>

<sup>a</sup> Departamento de Física Universidade Estadual de Londrina Campus Universitário, 86051-970, Londrina, (PR), Brazil

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## Periodic Patterns at the Neighborhoods of the Frèedericksz Threshold

M. SIMÕES and A.J. PALANGANA \*

*Departamento de Física Universidade Estadual de Londrina Campus  
Universitário, 86051-970, Londrina (PR), Brazil*

The theory that describes the arising twist-bend periodic walls at the neighborhoods of the Frèedericksz critical point affirms that the mode with the fastest initial grown will fix the observed properties of these patterns. From this principle it follows that just above the Frèedericksz threshold there is a region where this leading mode becomes null and, therefore, a homogeneous bending of the director would be detected. This prediction was not confirmed by the experiment and walls with very well defined wavelength were found. We will shown here that around the Frèedericksz threshold the fastest growing mode can not be defined and a new way to compute the observed structures will be proposed.

*Keywords:* patterns; patterns formation; Frèedericksz threshold

### INTRODUCTION

It is well known that, in nematic liquid crystals (NLC), the coupling between the bending of the director and the movement of the nematic fluid can produce high symmetric patterns. The so-called magnetic walls are examples of these well-studied textures. They are found when, under appropriated conditions, the NLC is submitted to an external magnetic field[1] greater than the Frèedericksz threshold[2]. In liquid crystals, the theory that firstly described the formation of these structures stated that the observed periodicity results from a selection mechanism that amplifies to macroscopic scale the modes having the fastest initial amplification[3, 4, 5]. Now on this mechanism will be called by leading mode principle. Nevertheless, there are some theoretical

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\* Permanent Address: Departamento de Física, Universidade Estadual de Maringá Avenida Colombo, 5790, 87020-900 Maringá(PR), Brazil.

and experimental evidences indicating that, even providing a good image of the beginning of these structures, this principle may not give a full explanation to the pattern observed on the magnetic walls. For example, it is known that for many physical process the study of pattern formation via the fastest growing mode gives incorrect results[6] and, therefore, one can be doubtful about its efficacy for the description of the magnetic walls of the liquid crystals. Furthermore, Amengual et al[7], in a study of the formation of the magnetic walls, have arrived at conclusions that do not corroborate the results of Lonberg et al[4]. They had studied a generic model for pattern transient formation and, in the non-linear regime, they did not find any signal of the selection mechanism proposed by Lonberg et al.

However, up to recently, an irrefutable evidence that the leading mode principle is not applicable to the formation of the magnetic walls in nematic liquid crystals did not exist[8]. Concretely, the disagreement between the calculations that follow from the leading mode principle and the experiment is given by a slight discrepancy[5]. In this paper it will be shown that at the neighborhoods of the Fréedericksz critical point the comparison between the predictions of the leading mode principle and experimental facts are in irreconcilable disagreement. That is, the leading mode principle predicts that when the Fréedericksz threshold is approached - from above - the coherent internal motion of the nematic material becomes smaller and smaller and there is a point -  $H_{lm}$  greater than the Fréedericksz critical point -  $H_f$  - below which there is no more induced motion of the nematic material. The region  $H_f < H < H_{lm}$  is known as the forbidden region, and at this region the torque of the external field on the nematic molecules would produce a uniform alignment, and the periodic walls would not be detected. Nevertheless, as it will be shown along this paper, when the Fréedericksz threshold is approached the emergence of these structures continues and, no matter how close the Fréedericksz critical point is approached, a homogeneous alignment of the director have never been found. Moreover, is also shown that the time spent in the construction of these periodic walls diverges as the critical point is approached. This is also in contradiction with the extensions of the Lonberg calculations to the neighborhoods of the critical point because, according to it, the formation of any magnetic wall would spent a finite time interval and only with the formation of a homogeneous bending - produced at the neighborhoods of the Fréedericksz critical point - a infinite time interval would be expected. Concluding, we will show that at the neighborhoods of the Fréedericksz threshold the predictions of the theory based on leading mode principle were not confirmed by the experiment.

## FUNDAMENTALS

In order to study the profile of the periodic walls, a NLC sample inside a microslide glass with dimensions  $(a, b, d)$  that satisfy the relation  $a \gg b \gg d$  will be considered. The director is initially uniformly aligned along the  $\vec{e}_x$  direction and an external controlled magnetic field  $H$  is applied along the  $\vec{e}_y$  direction. When  $H$  is greater than the Fredericks threshold  $H_F$  the magnetic susceptibility overcomes the elastic resistance of the medium, and the director begins to bend towards the direction of the external magnetic field trying to become parallel or anti-parallel to it. Due to this symmetry breaking, the bending of the director is not homogeneous in the sample[9, 10, 2, 1]. The presence of a set of unidimensional structures, magnetics walls, periodically distributed along the  $\vec{e}_x$  direction is observed.

As the external magnetic field is applied along the direction perpendicular to the initial orientation of the director, we can suppose that the director components will always remain in the plane defined by the direction of the magnetic field and the initial orientation of the director[4], that is  $n_x = \cos\theta(x, y, z)$ ,  $n_y = \sin\theta(x, y, z)$ ,  $n_z = 0$ . In order to study the NLC dynamics, the so-called Eriksen-Leslie-Parodi (ELP) approach[11, 12, 13] is used. The time evolution of the director direction and the motion of the nematic material is given by a set of differential equations composed by the anisotropic version of the Navier-Stokes equation, the balance of torques equation and the equation of continuity[14, 15]. The full form and the way by which these equations are handled on this problem can be found elsewhere[4, 16, 17]. As our aim is the obtainment of a non-linear formulation of this problem, some attention will be given to the approximations done on the equations of the ELP approach. The usual approximations assume that the fluid velocity does not have components along the  $\vec{e}_x$  direction, that the viscosity dominates the motion of the nematic material and the inertial component can be neglected, that the conditions prevailing at the borders of the sample have not any decisive influence in the shape of these structures, that the component of the velocity along the direction of the magnetic field,  $V_y$ , is the dominant one and only it needs to be considered[4], that save for the edges of the sample[16] the bending of director will be constant along the direction of the external magnetic field.

Using the above mentioned approximations we can show that the time evolution of  $\theta$  is given by

$$\partial_t \theta = \frac{1}{\tau_o} \left\{ \theta - \frac{\theta^3}{\theta_{\max}^2} \right\}, \quad (0.1)$$

where

$$\tau_o(\tilde{k}) = \frac{(1 - \frac{\gamma_1 \tilde{k}^2}{\eta_3 + \tilde{k}^2 \eta_1})}{(h^2 - 1 - \tilde{K} \tilde{k}^2)}, \quad \theta_{\max}^2(\tilde{k}) = \frac{(h^2 - 1 - \tilde{K} \tilde{k}^2)}{((h^2 - 1 - \tilde{K} \tilde{k}^2) \frac{R_0}{1-R_0} (1 + \frac{1}{\varphi_3^2}) + \frac{2}{3} h^2)}, \quad (0.2)$$

and  $\partial_x^2(\partial_x V_y) = -k^2 \partial_x V$ ,  $\partial_x^2(\partial_t \theta) = -k^2 \dot{\theta}$ ,  $R_0 = \gamma_1 \tilde{k}^2 / (\eta_3 + \tilde{k}^2 \eta_1)$ ,  $\varphi_o^2 = (\eta_3 + \tilde{k}^2 \eta_1) / ((\eta_2 + 2\eta_3) + \tilde{k}^2(2\eta_1 + \eta_2 + 2\eta_3))$ ,  $\tilde{k}^2 = (kd/\pi)^2$ ,  $\tau = (\chi_a H_c^2 / \gamma_1) t$ ,  $h^2 = H^2 / H_F^2$ ,  $\tilde{K} = K_{33} / K_{22}$ ,  $\chi_a H_F^2 = K_{22}(\pi/d)^2 + K_{33}(\pi/b)^2 \simeq K_{22}(\pi/d)^2$ .

The first order term of the Eq. (0.1) reproduces the know results about the initial moments of the time evolution of  $\partial_t \theta$ . The next order term of this equation predicts that for each mode  $\tilde{k}$  there is a different saturation angle,  $\theta_{\max}(\tilde{k})$ , for the bending of the director.

The leading mode principle states that the wave number,  $k_o$ , observed in the periodic walls is determined by choosing the mode that gives the highest velocity to the growing of the periodic walls,  $\partial_t \theta$ , at that the initial moments. That is, this mode must satisfies  $\partial_k(\partial_t \theta) = 0$ , at  $t = 0$ . Around the critical point ( $h^2 \simeq 1$  and  $k^2 \simeq 0$ ) an analytical computation shows that the non-null and real solution of this equation, and the corresponding time  $\tau_o$  are given by

$$k^2 = \frac{(h^2 - h_{lm}^2)}{P(\eta_1, \eta_2, \eta_3, \gamma_1, h^2)}, \quad \tau_o(\tilde{k} \approx 0) = \frac{\gamma_1 (3\eta_3 - \theta_o^2(3\eta_2 + 11\eta_3))}{\eta_3 (3\tilde{K}_{33}\eta_3 + 2\theta_o^2\gamma_1)}. \quad (0.3)$$

where  $P(\eta_1, \eta_2, \eta_3, \gamma_1, h^2)$  is a awkward and non null function that has no influence on the results presented below, and  $\tau_o(\tilde{k} \approx 0)$  is the time that must be spent to observe the arising of the walls with  $\tilde{k} \approx 0$ . From this equation it is easy to see that in the interval  $1 \leq h^2 \leq h_{lm}^2$ , where

$$h_{lm}^2 = (\frac{H_{lm}}{H_F^2})^2 = 1 + \frac{\eta_3 (3\tilde{K}_{33}\eta_3 + 2\theta_o^2\gamma_1)}{\gamma_1 (3\eta_3 - \theta_o^2(3\eta_2 + 11\eta_3))}, \quad (0.4)$$

and  $\theta_o$  stands for the initial distribution of the mode  $\theta(0, \tilde{k})$  that, as usual[1, 5], can be found using the equipartition theorem, the unique real solution of  $\partial_k(\partial_t \theta) = 0$  is given by  $\tilde{k}^2 = 0$ .

In the experiment the outputs of the parameters  $k^2$  and  $\tau_o$  were looked for in the neighborhoods of the Fréedericksz threshold. 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, 62.5. Nematic samples were encapsulated in flat glass microslide (length  $a = 20mm$ , width  $b = 2.5mm$ , and thickness  $d = 0.2mm$ ) from Vitrodynamics. During the experiment the temperature was controlled at  $25 \pm 1^\circ C$ .

According the above results, as  $k^2 \rightarrow 0$  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. 1 the dotted line gives a picture of this supposed result. It is also shown the results of our measurements. The surprising result is that the walls always exist and a region with homogeneous alignment was never found[18].

Furthermore, a graph of  $\tau$  vs.  $h^2$  would converge, as  $k^2 \rightarrow 0$ , to the point  $\tau_w$  calculated above, which is clearly a finite time interval. But, according our experimental results the time spent with the formation of these structures diverges as the point for which  $k^2 \rightarrow 0$  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. Therefore, it is experimentally evident that the two curves are not approaching the point  $h^2 = h_w^2$ , but the Fréedericksz critical point  $h^2 = 1$ .

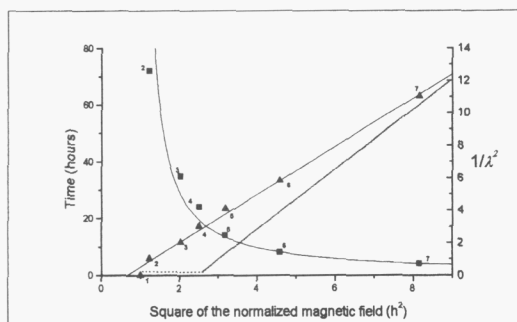


Figure 1-Measured points of  $\tau$  and  $(2d/\lambda)^2$  versus  $h^2$ . The squares give the measured points for  $\tau$  that are read in left side. The triangles give  $(1/\lambda)^2$  and are read at the right. The continuous line accompanying the squares and the triangles is only for eyes guiding. 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 remains zero until the point  $h^2 = 1$ . No such behavior was found in the experiment. The measured points 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 found the formation of any kind of structure in the sample even after a week of continuous exposure to the magnetic field. As was demonstrated along our paper when the point  $h_w^2$  is approached the bending of the director should becomes 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 lasted for the walls construction becomes infinite as the critical point was approximated.

## THE LEADING MODE

In order to understand the physical origin of this fail in the theory of the leading mode, the second derivative,  $\partial_k^2 (\partial_r \theta)$ , must be considered. It is straightforward to show that at the region  $\tilde{k}^2 \simeq 0$  one has

$$\partial_k^2 (\partial_r \theta) = 2\eta_3^2 (3\gamma_1(h^2 - 1)(\eta_3 - \theta_o^2\eta_2) - 3\tilde{K}_{33}\eta_3^2 + \gamma_1\eta_3\theta_o^2(9 - 11h^2)). \quad (0.4)$$

From this result it is easy to see that

$$\partial_k^2 (\partial_r \theta) = 0 \quad \text{at} \quad h = h_{lm}. \quad (0.5)$$

Likewise, a straightforward calculation shows that

$$\partial_k^3 (\partial_r \theta) = 0 \quad \text{at} \quad h = h_{lm}. \quad (0.6)$$

Finally it is found that

$$\partial_k^4 (\partial_r \theta) = -24\tilde{K}\frac{\eta_1}{\eta_3} + O(\theta_o^2) < 0 \quad \text{at} \quad h = h_{lm}, \quad (0.7)$$

where  $O(\theta_o^2)$  represents the terms of the order of  $\theta_o^2$ , or higher, that are insignificant.

So,  $h_{lm}$  is a special point of the leading mode principle; only the fourth derivative of the growing speed of the director with relation to  $k$  is non-null and negative. Due to the results of the above equations we see that at  $h = h_{lm}$  the width of the distribution of the modes contributing to the arising of the walls become so large that the contribution of an isolated mode can not be considered. These results can be seen in the fig. (2) where a numerical computation, using the known parameters of the MBBA, of the behavior of  $\partial_r \theta$ , as a function of  $\tilde{k}$ , as the point  $h_{lm}$  is approached is shown. In these figures we see that as  $h_{lm}$  is approached a isolated contribution of a leading mode no more exist. Clearly, this is origin of the fail in the finding of a homogeneous alignment in the forbidden region.



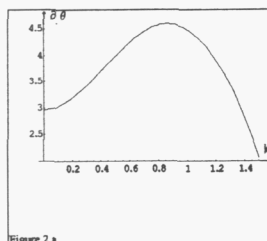


Figure 2.a

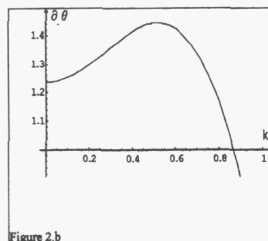


Figure 2.b

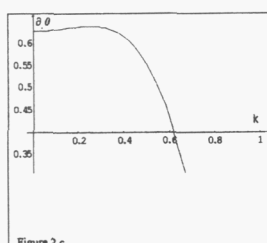


Figure 2.c

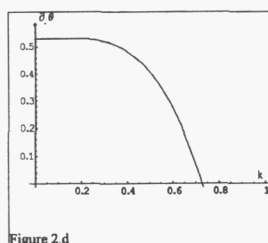


Figure 2.d

Figure 2 - In these figures it is shown the graphics of the function  $\partial_\tau \theta$  at  $\tau = 0$  when  $\tilde{k}$  is changed. Each figure was computed for a different value of  $h$ . These figures, obtained using the parameters of the MBBA, shown the evolution of the leading mode as the point  $h_{lm}$ , defined at Eq. (0.3), is approached. The figure 1(a) was computed at  $h = 2h_{lm}$ , the point  $\tilde{k}$  for which the function  $\partial_\tau \theta$  has the maximum growing is clearly observed, this is the fastest mode that will determine the final periodicity of the wall. The figure 1(b) was computed at  $h = 1.35h_{lm}$ , the leading mode exist but is less pronounced. The figure 1(c) was computed at  $h = 1.1h_{lm}$ , the leading is not clearly recognizable, but we can see that it exist. The figure 1(d) was computed at  $h = h_{lm}$ , the leading mode collapsed to the point  $\tilde{k} = 0$ . From these figures we have a clear evidence that as  $h_{lm}$  is approached the width of the modes around the leading mode has become so large that an isolated leading mode can not be recognized.

As, at the neighborhoods of the Fréedericksz threshold, the observed periodicity of the walls cannot be understood as a result of the contribution of a unique and isolated mode, we may suppose that the final periodicity of the walls will be determined by the long time collective growing of the modes neighboring the mode  $\tilde{k} = 0$ . In this case, the natural candidate to fix contribution of each of these modes to the final profile is just the maximum amplitude that each

of them can attain. So, it can be assumed that around  $\tilde{k} \simeq 0$  the final profile  $\theta(x)$  of the wall will be given by

$$\theta(x) = \int \theta_{\max}(\tilde{k}) \cos(\tilde{k}x) d\tilde{k}. \quad (0.9)$$

With the form of  $\theta_{\max}(k)$  given in Eq.(0.2) this integration is impossible. As the maximum of  $\theta_{\max}(k)$  occurs at  $\tilde{k} = 0$ , it can be expanded around this point and it is obtained  $\theta_{\max}(k) \simeq \sqrt{a - bk^2}$ , where  $a = \frac{3}{2h^2}(h^2 - 1)$  and  $b = \frac{3\tilde{K}}{2h^2}$ , and it have been assumed that  $(h^2 - 1)^2$  is small.

So,

$$\begin{aligned} \theta(x) &= \sqrt{b} \int_{-\sqrt{\frac{a}{b}}}^{\sqrt{\frac{a}{b}}} \sqrt{\frac{a}{b} - k^2} \cos(kx) dk = \\ &= \frac{\pi\sqrt{a}}{x} J_1\left(\sqrt{\frac{a}{b}}x\right) \simeq \frac{\pi}{2} \sqrt{\frac{3}{2\tilde{K}h^2}} (h^2 - 1) \cos \frac{1}{2} \sqrt{\frac{(h^2 - 1)}{\tilde{K}}} x \end{aligned} \quad (0.10)$$

where  $J_1(\sqrt{a/b}x)$  is a Bessel function of first class and order 1. Which has been approximated by  $J_1(x) \simeq (x/2) \cos(x/2)$ .

So, the wave vector of this periodic pattern is given by

$$\tilde{k}^2 = \frac{1}{4\tilde{K}}(h^2 - 1). \quad (0.11)$$

Which is in accord with the result experimentally found. Furthermore, according to the Eq. (0.2) the time spent in the formation of this structure is given by

$$\tau_o(h^2) = \frac{4}{3(h^2 - 1)} \frac{(h^2 - 1)(\eta_1 - \gamma_1) + 4\tilde{K}\eta_3}{(h^2 - 1)\eta_1 + 4\tilde{K}\eta_3} \quad (0.12)$$

That, as was experimentally found, diverges when  $h^2 \rightarrow 1$ .

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