PHYSICAL REVIEW E VOLUME 54, NUMBER 4 OCTOBER 1996

Periodic distortions in lyotropic nematic calamitic liquid crystals

M. Simões
Departamento de Física, Universidade Estadual de Londrina, Campus Universitário, 86051-970, Londrina, Parana, Brazil

A. J. Palangana and L. R. Evangelista
Departamento de Física, Universidade Estadual de Maringá, Avenida Colombo, 5790, 87020-900 Maringá, Parana, Brazil

(Received 18 December 1995)

A theoretical tool to determine the dependence of the wavelength of periodic structures on the applied magnetic field in lyotropic liquid crystals in the nematic calamitic phase is proposed. It is assumed that the periodic structure experimentally found can be represented by a sequence of walls. Our calculations indicate that in order to describe the measured behavior of the system above the Fredericks threshold the interaction between these walls must be taken into account. The strength of these interactions and the ratio between the bend and twist elastic constants are also determined. [S1063-651X(96)04509-6]

PACS number(s): 61.30.Gd, 61.30.Jf, 64.70.Md

I. INTRODUCTION

The distortion of the texture of nematic liquid crystals (NLC) under the action of a magnetic or electric field is a well known phenomena. It can be observed that under the action of the magnetic field, two symmetrical distorted textures can be created, separated by a wall [1]. These walls in thermotropic liquid crystals have been studied many years ago both from the theoretical and experimental point of view [2–7]. More recently, the situation in which a magnetic field is applied to a previously oriented sample of lyotropic nematic liquid crystals has been analyzed [8,9]. These NLC are formed by mixtures of amphiphilic compounds and water, at proper temperature and concentrations conditions [10]. In general, the periodic distortions of the textures can be observed in a polarizing microscope, with the polarizing direction parallel to the long axis of the micelle, the basic constituent of lyotropics.

In recent papers the wavelength of the periodic distortion (λ) was utilized to determine the bend elastic constant (K_3) and the anisotropy of diamagnetic susceptibility χ_a of two different lyotropic systems [11]. In the frame of the elastic theory a theoretical approach has been recently proposed in order to estimate the anchoring strength, in the Rapini-Papoular sense, for the situation of weak anchoring [12]. The behavior of the distortion angle as a function of the magnetic field has also been investigated, in the one constant approximation [13]. The periodic structures arising in these systems, under the action of magnetic fields, correspond to metastable states. In principle, the usual elastic approach, based on the one constant approximation, cannot provide a completely satisfactory description of the equilibrium state of this system. This implies that a more complicated nonlinear analysis is required in order to fully describe its behavior. However, as can be experimentally observed, the relaxation time for these periodic structures is very large. Only after a few hours the instabilities collapse into closed elliptical loops [4]. This fact can be responsible for the good agreement obtained between a simplified elastic approach and several experimental results.

In this work we investigate the dependence of the wavelength λ of the periodic structure on the applied magnetic field. Once a magnetic field is applied perpendicularly to the direction of the director of the NLC sample, one observes the arising of walls parallel to the magnetic field as soon as the Fredericks threshold is reached [14]. The nature and physical properties of the walls we are dealing with were studied by Brochard in Ref. [3], where the dependence on the magnetic field of the length of just one wall was measured. Here we use the fact that usually these walls appear as a periodic array in the nematic structure, and study the length of this periodicity when it appears along the x axis. The experimental data we are utilizing were obtained in regions of a magnetic field nearly equal and greater than the Fredericks threshold [12].

This paper is organized as follows. Section II is dedicated to the presentation of the basic equations of the theoretical approach. We also discuss the experimental procedure. In Sec. III we present the main results of our calculation. In Sec. IV some concluding remarks are drawn.

II. FUNDAMENTALS

The lyotropic sample consists of potassium laurate (KL), decanol (DeOH) and water, with the following concentrations in weight percent: 29.4, 6.6, and 64, respectively. The phase sequence, determined by optical and conoscopic observations is as follows: isotropic (15 °C)—calamitic nematic (50 °C)—isotropic. Calamitic nematic (N_c) samples were encapsuled at room temperature, in flat glass microslides from Vitro Dynamics with dimensions $a=20$ mm (parallel to the x axis), $b=2.5$ mm (parallel to the y axis), and thickness $d=0.2$ mm (parallel to the z axis). The method of generating the periodic distortion of the director [8] consists in orienting a N_c sample in a planar geometry, with a high magnetic field (10 kG along the x axis). After a well-oriented sample is achieved, the field is switched off and a controlled magnetic field is applied along the y axis. The resulting competition between the magnetic susceptibility (which tends to align the director along the field) and the elastic energies (which tend to retain a uniform orientation of the director consistent with the orientation at the surface of

1063-651X/96/54(4)/3765(6)/$10.00

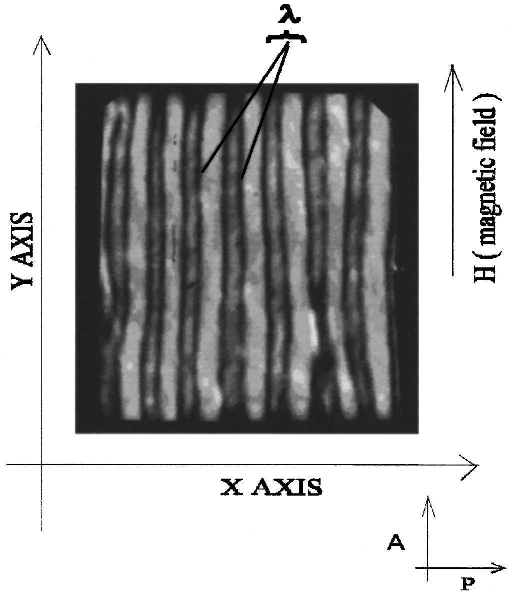

FIG. 1. Lyotropic nematic phase in microslides 200 mm thick between crossed polarizers. Magnetic field (3.0 kG) along the y axis at 0^0 of the light polarizing direction. P and A are the directions of the polarizer and the analyzer, respectively.

the sample) creates a distortion of the director at small magnetic fields. The director in the interior of the sample is subjected to a torque trying to rotate it. However, at the same time, it experiences an elastic restoring torque due to the anchored surface layers. For an applied field below a critical strength H_c, no distortion occurs. For fields larger than H_c, the distortion of the texture is observed [12], indicating that the magnetic coupling between $\vec{n}$ and H is bigger than the mechanical coupling between $\vec{n}$ and the boundaries of the microslide. To understand the observed periodicity we remember that as the director has the π symmetry ($\vec{n}$ and $-\vec{n}$ are equivalent), in the very moment of the Fredericks transition it has two choices: it bends clockwise or counterclockwise. Therefore two disconnected portions of the sample can bend in different directions, and this is the mechanism producing the periodic walls that we see in the sample [1]. A typical periodic distortion [8,13] is depicted in Fig. 1. The wavelength (λ) of the periodic distortion is then directly measured as a function of the applied magnetic field. With this geometry we suppose that the components of the director can be expressed by

$$n_x = \cos\theta(x,y,z), \quad n_y = \sin\theta(x,y,z), \quad n_z = 0, \tag{1}$$

where $\theta(x,y,z)$ is the angle between the director **n** and the x axis direction. The expression of the free energy density in the two elastic constant approximation ($K_1 = K_3$), taking into account the magnetic field coupling is [1,3]

$$F = \int_v \{ \tfrac{1}{2} K_3 [(\partial_x \theta)^2 + (\partial_y \theta)^2] + \tfrac{1}{2} K_2 (\partial_z \theta)^2 - \tfrac{1}{2} \chi_a H^2 \sin^2\theta \} dv, \tag{2}$$

where K_1, K_2, and K_3 are, respectively, the elastic constants of splay, twist, and bend. According to the experimental observations, we are dealing with period structures along the x axis, without structures along the y and z axes. In this way we assume, for future use in a variational approach, an explicit form for the function $\theta(x,y,z)$, that represents the spatial configuration of the director $\vec{n}$. For simplicity strong anchoring boundary conditions are assumed at the edges of the sample, that is,

$$\theta(0,y,z) = \theta(a,y,z) = \theta(x,0,z) = \theta(x,b,z) = \theta(x,y,0) = \theta(x,y,c) = 0. \tag{3}$$

Moreover, since the periodic structures are observed along the x direction, we suppose trivial configurations satisfying these boundary conditions along y and z directions. Thus

$$\theta(x,y,z) = \eta(x) \sin\left(\frac{\pi y}{b}\right) \sin\left(\frac{\pi z}{d}\right), \tag{4}$$

for $0 \leqslant x \leqslant a, 0 \leqslant y \leqslant b, 0 \leqslant z \leqslant d$, where $\eta(x)$ is the configuration of the nematic periodic structure along the x axis. For instance, observe that $\sin(\pi y/b)$, $0 \leqslant y \leqslant b$, and $\sin(\pi z/c)$, $0 \leqslant z \leqslant d$ have half wavelength which corresponds to the most simple structure along these directions. The periodicity of $\eta(x)$ implies that $\eta(x+\lambda) = \eta(x)$. For convenience of the following analysis the configuration $\eta(x)$ will be chosen in such a way that $\eta(x) = \varphi_0 g(x/\lambda)$, where φ_0 is a constant characterizing the amplitude of the periodic structure and $g(t)$, with $t = x/\lambda$, the profile of $\eta(x)$. The scaling $(t \to x/\lambda)$ implies that the periodicity of $g(x)$ will assume the form $g(t+1) = g(t)$. With these replacements, after a straightforward calculation, Eq. (2) can be rewritten as

$$F = \frac{1}{4} b d K_2 \left(\frac{\pi}{d}\right)^2 N\lambda \int_0^1 dt \left\{ \frac{1}{2} \frac{K_3}{K_2} \left(\frac{d}{\pi\lambda}\right)^2 \varphi_0^2 (\partial_t g)^2 + \frac{1}{2}(1-h^2)\varphi_0^2 g^2 + \frac{h^2}{4} \frac{\varphi_0^4}{\theta_0^2} g^4 \right\}, \tag{5}$$

where N ($= a/\lambda$) is the number of periods of $\eta(x)$ along the x direction and in order to achieve a compactness in notation we have introduced $\theta_0^2 = 8/3$, $h = H/H_c$, with H_c being the usual Fredericks threshold $[\chi_a H_c^2 = K_2(\pi/d)^2$, in the limit $a \gg b \gg d]$ for the strong anchoring case [12]. We also made $\sin^2\theta \approx \theta^2 - (2/3!)\theta^4$ in Eq. (2). Furthermore, the periodicity of $g(t)$ allows us to express the integration in Eq. (5) along just one period.

To obtain the periodic pattern configuration of the director, Eq. (5) can be extremized in terms of the function $g(t)$ and the parameters φ_0 and λ. From the Euler-Lagrange equation for g, which gives the analytical form of the periodic structure, we obtain

$$\left(\frac{K_3}{K_2}\right)\left(\frac{d}{\pi\lambda}\right)^2 \partial_t^2 g + (h^2 - 1) g - h^2 \left(\frac{\varphi_0}{\theta_0}\right)^2 g^3 = 0. \tag{6}$$

This equation corresponds to the field equation for the "$\lambda\varphi^4$" field theory. Then its solutions can be searched between the usual kink solutions of the nonlinear theories [15]. The analysis reported in Ref. [3] uses the kink solution of Eq. (6), with only one wall. However the observed periodicity along the x direction suggests that we consider this structure as composed by a family of these walls. Meanwhile it is important to observe that the exact solution of a family of kinks in the "$\lambda\varphi^4$" theory is unknown [15]. Thus any ap-

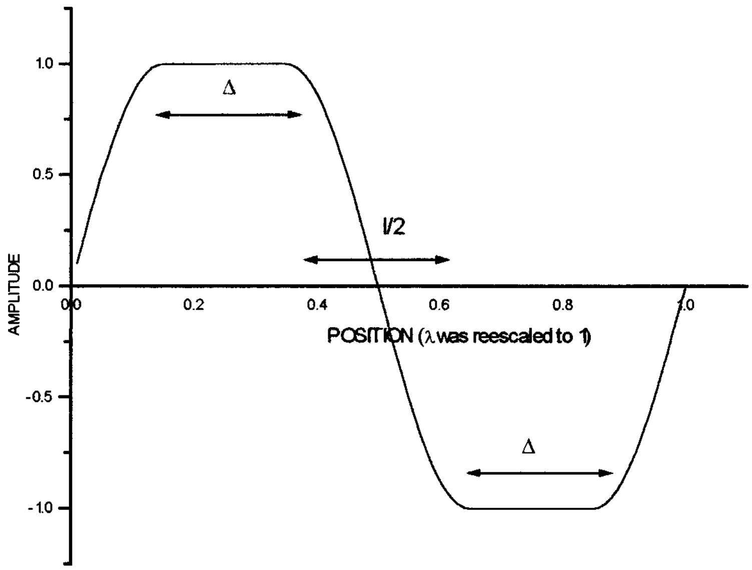

FIG. 2. Graphic representation of $g(t)$. The distance Δ is explicitly shown between two consecutive walls, and the portion l where there is bending between the two stable situations. We have $l+2\Delta=1$.

proach to this problem requires some kind of approximation. As an explicit and approximated form for the wall let us consider the following ansatz:

$$g(t)=\begin{cases} \sin\left(\frac{2\pi}{l}t\right), & 0<t<\frac{l}{4} \\ 1, & \frac{l}{4}<t<\frac{1}{2}-\frac{l}{4} \\ \sin\left[\frac{2\pi}{l}\left(\frac{1}{2}-t\right)\right], & \frac{1}{2}-\frac{l}{4}<t<\frac{1}{2} \end{cases} \tag{7}$$

$$g(\tfrac{1}{2}+\epsilon)=-g(\tfrac{1}{2}-\epsilon), \quad 0<\epsilon<\tfrac{1}{2}.$$

The profile of this structure is depicted in Fig. 2, from which it is not difficult to see that

$$l+2\Delta=1. \tag{8}$$

From this figure it is also easy to see that when the wall is absent the director points along the direction given by the angle φ_0. As the angle $-\varphi_0$ is also possible, the wall just links these two configurations, bending the director from one configuration, φ_0 for example, to the another $-\varphi_0$[13]. The portion of the interval (0,1) in which $g(t)$ is composed by walls is just given by l, while 2Δ is the portion of $g(t)$ in which the director remains pointing in the φ_0 (or $-\varphi_0$) direction, which allows us to say that Δ is a measure of the distance between the walls. As we have scaled the length λ of the periodicity to 1, the values of l and Δ are, respectively, the fraction of the period λ occupied by walls and the φ_0 (or $-\varphi_0$) is the inclination of the director.

It is also easy to see that as we considered this Δ portion of $g(t)$ as a straight line, the free energy density of the interaction between these disclinations could not be taken into account. In order to give a complete description of the phenomenon of periodic distortions, we need to add a term for the interaction energy that we have neglected when we made this approximation. Its explicit form is difficult to obtain, but it may be responsible for the number of walls, and the distance among them. It plays the role of a chemical potential.

In this sense we may assume that this term has the generic form

$$f_W=bdN\lambda\gamma, \tag{9}$$

and will be added to Eq. (5). Observe that b, d, N, and λ have been defined above, and γ is a function characterizing the interaction between the walls to be determined below.

In spite of the fact that Eq. (6) can not be integrated for a solution corresponding to a sequence of many walls, global properties of these solutions can be obtained from the conserved quantities that follows from Eq. (6). In this way we recall that

$$C=\frac{1}{2}\left(\frac{K_3}{K_2}\right)\left(\frac{d}{\pi\lambda}\right)^2(\partial_t g)^2+\frac{1}{2}(h^2-1)g^2-\frac{1}{4}h^2\left(\frac{\varphi_0}{\theta_0}\right)^2 g^4, \tag{10}$$

follows from Eq. (6) as a constant. This is an exact result that can be used with Eq. (7) in order to obtain a relationship characterizing the parameters of $\eta(t)$ [λ, l (or Δ) and φ_0]. From Eq. (7) it is possible to deduce that in the points for which $g(t)=0$ ($t=0$, for example) we have $\partial_t g=(2\pi/l)$. Thus, Eq. (10) is reduced to

$$C=\frac{1}{2}\left(\frac{K_3}{K_2}\right)\left(\frac{2d}{\lambda}\right)^2\frac{1}{l^2}. \tag{11}$$

Similarly, for the points where $g(t)=1$ we have $\partial_t g=0$. Therefore,

$$C=\frac{1}{2}(h^2-1)-\frac{1}{4}h^2\left(\frac{\varphi_0}{\theta_0}\right)^2. \tag{12}$$

From Eqs. (11) and (12) we obtain

$$\left(\frac{K_3}{K_2}\right)\left(\frac{2d}{\lambda}\right)^2 = l^2\left\{(h^2-1)-\frac{1}{2}h^2\left(\frac{\varphi_0}{\theta_0}\right)^2\right\}. \quad (13)$$

This equation indicates that $(1/\lambda)^2$ depends separately on h^2, φ_0, and also on l^2. This is the main motivation for the scaling that sets apart the length λ of the periodic structure and the factorization of Eq. (5) in amplitude φ_0 and the form $g(t)$.

Let us now consider the extremization of Eq. (5) in terms of φ_0. The variational procedure yields

$$\left(\frac{\varphi_0}{\theta_0}\right)^2 = \frac{8}{8-5l}\frac{1}{h^2}\left\{(h^2-1)\left(1-\frac{l}{2}\right)-\left(\frac{K_3}{K_2}\right)\left(\frac{2d}{\lambda}\right)^2\frac{1}{2l}\right\}. \quad (14)$$

From Eqs. (13) and (14) we obtain

$$\left(\frac{K_3}{K_2}\right)\left(\frac{2d}{\lambda}\right)^2 = l^2(h^2-1)\frac{4-3l}{8-7l}. \quad (15)$$

Note that if we have assumed, from the beginning, that $l=1$ which means that all the periodic structures would be composed by walls, without a finite distance Δ between them [see Eq. (8)], this last equation would predict that a plot of $(1/\lambda)^2$ vs h^2 would be just a straight line with (K_2/K_3) as an angular coefficient. This result does not account for the entire behavior of the experimental curve. In fact, this curve presents a nonlinear behavior for values of the magnetic field far from the Fredericks threshold. Therefore, we can conclude that we must have $l\neq 1$ (or $\Delta\neq 0$).

Similar procedure can be employed for the extremization of Eq. (5) in terms of λ, giving

$$\left(\frac{K_3}{K_2}\right)\left(\frac{2d}{\lambda}\right)^2 = l\left\{(1-h^2)(2-l)+h^2\left(\frac{\varphi_0}{\theta_0}\right)^2\left(1-\frac{5}{8}l\right)+\frac{2\tilde{\gamma}}{\varphi_0^2}\right\}, \quad (16)$$

where we have used the interaction term Eq. (9) and $\tilde{\gamma}=(4/K_2)(d/\pi)^2\gamma$.

By using Eqs. (15) and (16) we obtain

$$4\theta_0^2(h^2-1)^2(1-l)(l^2+5l-8)+\tilde{\gamma}h^2(8-7l)^2=0. \quad (17)$$

This equation gives l as a function of h^2. However, to solve the problem in a complete manner, we should find also $\tilde{\gamma}$, i.e., we have to determine the interaction among the walls. To obtain an approximated form for this interaction we remember that near the Fredericks threshold ($h^2\to 1$), the periodic structure tends to a sinusoidal behavior with very small amplitude φ_0. As the magnetic field h is raised, an increasing number of nematic monodomains tends to be aligned with the magnetic field. This is equivalent to a growing of the fraction of $g(t)$ having amplitude 1. Therefore, the fraction of $g(t)$ representing the distance between the walls [Δ in Eq. (8)] increases with the magnetic field. On the other hand, it is important to stress that the walls are just the objects whose presence in the nematic structure does not permit a homogeneous alignment, that is, the portion $l/2$ of the wall (see Fig. 2) tends to destroy this alignment. Thus there is a competition between these two ''forces'' leading the director to oscillate between φ_0 and -φ_0. In this way there is a competition between the walls that tend to disturb the director alignment and the aligned portion that tends to arrange the director in the same direction along the sample. This competition must reach a stable pattern for some value of Δ. From these considerations we may assume an elastic form for this force, whose ''potential energy'' is represented by

$$\tilde{\gamma}=\alpha\Delta^2. \quad (18)$$

Observe that we have assumed that the parameter α does not depend on the magnetic field. In fact, we could not say from beginning that it is true. Actually the real form of the interaction between the walls is yet unknown. We just assume a stable configuration governed by an elastic interaction given by Eq. (18). Therefore α is just an elastic parameter that we have determined from the experimental data. Our results indicate that it can be taken as a constant, as we discuss in the next section.

Substitution of Eqs. (18) into (17), after considering Eq. (8), yields

$$16\theta_0^2(h^2-1)^2(l^2+5l-8)+\alpha(1-l)h^2(8-7l)^2=0, \quad (19)$$

which may be used to determine l. When l is substituted in Eqs. (15) and (14) we obtain $(1/\lambda)^2$ and $(\varphi_0/\theta_0)^2$. The parameters α and (K_2/K_3) can be adjusted in order to fit the experimental points. The importance of these parameters is obvious: α characterizes the interaction between adjacent walls, and $(K_2/K3)$ is the ratio between the elastic constants.

III. RESULTS AND DISCUSSION

Let us now present the main results of our model and calculations. Our aim was to use Eq. (15) in order to obtain a good fit for the experimental points depicted in Fig. 3. To obtain this fit we have solved Eq. (19) at fixed α for each value of h. The result was substituted in Eq. (15) and a value for the ratio K_2/K_3 was chosen to get a curve for $(2d/\lambda)^2$. This procedure was repeated until the best fit for the points of Fig. 3 was found. Its corresponds to the values $\alpha=1600$, and $K_3/K_2\approx 2.2$. This value for the ratio between the bend and twist constants is in good agreement with a recent experimental determination [12]. Its is important to underline the advantage of a presentation of a graph of $(2d/\lambda)^2$ versus h^2. If the interaction between the walls is neglected, this graph would be a straight line [see Eq. (15) and the text below it]. Therefore the curvature that can be observed in Fig. 3 reflects the interaction between the walls.

As a consequence of our procedure we have found the solution for l in Eq. (19). Thus it is possible to determine the fraction of λ which is occupied by walls connecting the two stables configurations φ_0 and -φ_0. The result is shown in Fig. 4. We observe that when the magnetic field tends to the Fredericks threshold ($h^2\to 1$) all the periodic structure represented by $\eta(t)$ is occupied by walls, as expected. As the magnetic field increases, l decreases and, hence, the fraction Δ for which the director points in the φ_0 (or -φ_0) direction also increases with h.

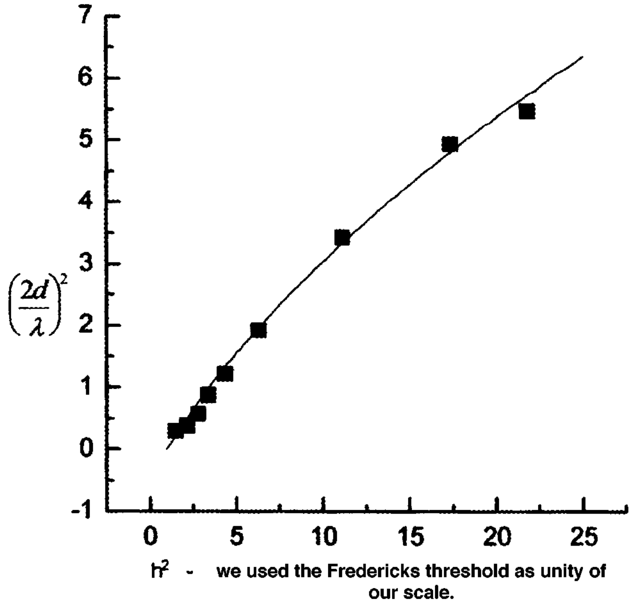

FIG. 3. The behavior of $(2d/\lambda)^2$ as a function of h^2. The points are the experimental data. The continuous line is the result of the present calculations. The best fit was obtained for $\alpha=1600$ and $(K_3/K_2)=2.2$.

Our procedure also gives enough information to solve Eq. (14). The result is shown in Fig. 5. Note that when the field tends to the Fredericks threshold ($h^2 \rightarrow 1$), $\varphi_0 \rightarrow 0$. On the contrary, φ_0 tends to θ_0 for higher magnetic fields.

IV. CONCLUDING REMARKS

In this paper the periodic structures formed by the action of a magnetic field in a lyotropic nematic have been investigated. To account for the experimental behavior of the system, a theoretical analysis in which this periodic structure is considered as a sequence of domain walls was proposed. The analysis is carried out by means of a variational procedure developed in the frame of the elastic continuum theory. The main purpose of the analysis was the investigation of the dependence of the wavelength on the applied magnetic field. We have shown that, in order to achieve a satisfactory description for the behavior of the real system, the interaction between the walls has to be taken into account in an explicit way. Once the best fits for the curves are established, it is easy to also obtain a good estimation for the ratio between the elastic constants of bend and twist. This estimation is in good agreement with independent measurements recently performed. Furthermore, in the theoretical context that we have analyzed the system, the strength of the interaction between the walls is also easily determined.

We stress again that we are dealing with a metastable system whose relaxation time is very large. Then a complete description of the system requires the development of a more sophisticated analysis of the entire elastic problem. In this analysis other relevant features of the system like viscosities and more appropriated boundary conditions have to be taken into account. Nevertheless, the phenomenological approach we are proposing can be extended to treat another lyotropic system. In this sense it can constitute a useful tool to investigate the elastic properties of these systems.

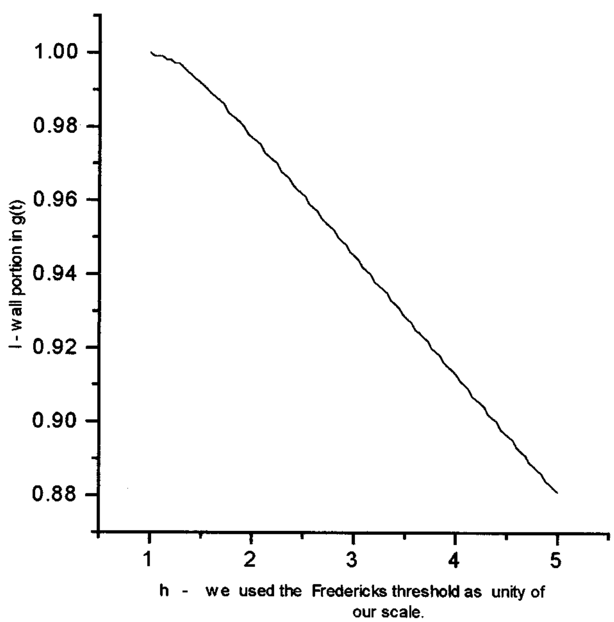

FIG. 4. The trend of l vs the reduced magnetic field h.

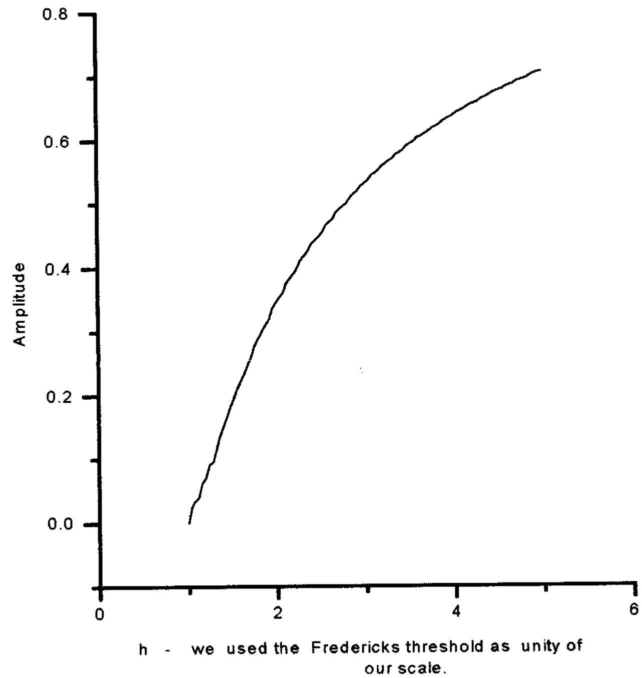

FIG. 5. Trend of the amplitude φ_0/θ_0. We have used $\alpha=1600$, and $K_3/K_2=2.2$.

ACKNOWLEDGMENTS

Financial support was received from the Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq).

[1] P. G. de Gennes, *The Physics of Liquid Crystals*, 2nd ed. (Clarendon, Oxford, 1993).
[2] W. Helfrich, Phys. Rev. Lett. **21**, 1518 (1968).
[3] F. Brochard, J. Phys. (Paris) **33**, 607 (1972).
[4] L. Léger, Mol. Cryst. Liq. Cryst. **24**, 33 (1973).
[5] F. Lonberg, S. Fraden, A. J. Hurd, and R. B. Meyer, Phys. Rev. Lett. **52**, 21 (1984).
[6] F. Lonberg and R. B. Meyer, Phys. Rev. Lett. **55**, 718 (1985).
[7] C. Srajer, S. Fraden, and R. B. Meyer, Phys. Rev. A **39**, 9 (1989).
[8] T. Kroin and A. M. Figueiredo Neto, Phys. Rev. A **36**, 2987 (1987).
[9] J. Charvolin and Y. Hendrix, J. Phys. (Paris) Lett. **41**, 597 (1980).
[10] *Introduction to Liquid Crystals*, edited by E. B. Priestley, P. J. Wojtowickz, and P. Sheng (Plenum, New York, 1979).
[11] T. Kroin, A. J. Palangana, and A. M. Figueiredo Neto, Phys. Rev. A **39**, 5373 (1989).
[12] A. M. Ribas, L. R. Evangelista, A. J. Palangana, and E. A. Oliveira, Phys. Rev. E **51**, 5204 (1995).
[13] A. J. Palangana, L. R. Evangelista, and A. K. Zvezdin, Phys. Lett. A **200**, 56 (1995).
[14] V. Frederisckz and V. Zolina, Trans. Faraday Soc. **29**, 919 (1933).
[15] R. Rajaraman, *Solitons and Instantons* (North-Holland, Amsterdam, 1984).