Mechanically induced Helfrich-Hurault effect in lamellar systems

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(Received 16 June 2008; revised manuscript received 6 July 2009; published 21 September 2009)

Layered phases are a common pattern of self-organization for several soft materials. These phases undergo buckling instability when subjected to dilatative strain: beyond a critical threshold, layers, initially flat, exhibit a periodical undulation. By using a continuum model, in a finite deformation framework, an expression for the critical threshold is provided, which differs from that predicted by the Helfrich-Hurault theory and yet it reverts to it in a thick specimen limit. With respect to the relevant literature, an analogous disagreement is found in the undulation amplitude expression as well. The obtained results appear particularly relevant when dealing with layered materials whose intrinsic coherence length is comparable to the cell thickness.

DOI: 10.1103/PhysRevE.80.031710 PACS number(s): 61.30.Dk, 64.70.Nd, 46.32.+x

I. INTRODUCTION

A certain number of condensed phases (cholesteric and smectic liquid crystals [1], elastomers in liquid crystalline phase [2,3], bilayer lipid membranes [4], diblock copolymers [5], ferrofluids [6], ferrimagnets [7], and skin [8]) can be regarded as two-dimensional layered systems with one-dimensional translational order in the direction normal to the layers. Such layered materials are often modeled as a stack of layers, characterized by a curvature energy, which resists to layer bending, and by a dilatative elastic energy across, that opposes to layer thickness variation. Such materials, usually confined between two flat plates, are commonly employed in the design of technological devices [9–11].

In their natural configurations, layers are flat, equispaced and arranged in two possible configurations: either parallel to the delimiting plates, in the so-called homeotropic alignment; or perpendicular to the sample cell boundary plates, in the so-called bookshelf structure. Different perturbation sources exist, such as mechanical forces, electric and magnetic fields, temperature changes, and boundary conditions, which attempt to alter such ordering arrangement.

Layers of samples in homeotropic alignment can undulate when subject to external electric or magnetic field [12–17]. In practice, the material is confined between two parallel plates to which the layers are initially parallel. A magnetic or electric field applied normally to the plates tends to reorient the layers. This effect competes against both the elastic behavior and the surface anchoring at bounding plates, which hinder the lamellar free rotation. There exists a critical applied field beyond which the layers undergo undulation instability. In cholesteric or smectic liquid crystals, this phase transition is called the Helfrich-Hurault effect [12,13].

Layers undulation in lamellar phases may also be triggered through a mechanically induced dilatation of the layers [18–21]. Beyond a critical displacement of the boundary plates, the lamellae undulate in order to compensate the increased sample thickness. A short review of smectic layer undulation induced by tensile strains on a semi-infinite cell, may be found in ([1], Sec. 7.1.7, p. 364). Control of this effect is relevant to the design of industrial appliances.

In this paper, buckling instability is triggered by imposing a dilatative strain on one bounding plate of the cell. It is shown that the critical threshold predicted by the classical theory is the limiting condition of the general expression as the imposed displacement becomes negligible with respect to the sample thickness d. Indeed, the critical displacement γ_0 predicted by the classical theory, measured in units of sample thickness d, is given by

$$\gamma_0 = 2\pi\frac{\lambda}{d}, \tag{1}$$

where λ is a characteristic length of the material, obtained in terms of the ratio between the curvature and the dilatative elastic constants. Conversely, a critical threshold is found

$$\gamma_{cr} = -\frac{1}{2} + \frac{1}{2}\sqrt{1 + 8\pi\frac{\lambda}{d}},$$

which reduces to the classical result inasmuch as λ/d is very small. We name the condition $\lambda \ll d$ the thick cell limit; likewise, samples where λ is of the order of d are addressed as thin cells.

This paper is organized as follows: in Sec. II, we give the expression of the free energy, which characterizes the material right beyond the critical threshold. In Sec III, the related free-energy Euler-Lagrange equations are derived, therefrom the critical threshold and layer buckled profile are assessed. A discussion is put forward in Sec. IV in order to justify the results and the discrepancy with the classical ones. An Appendix at the end of the paper presents an alternative derivation of the relevant geometrical quantities in continuum mechanics language.

II. POST-THRESHOLD FREE ENERGY

Layers of lamellar phases can be identified through the isosurfaces of a scalar function ω. It is then possible to introduce the field of the unit vector normal to the layers $\mathbf{n}=\nabla\omega/|\nabla\omega|$, where ∇ is the spatial gradient operator.

To better fix ideas within the realm of layered materials, the case of smectic-A liquid crystals is considered in the

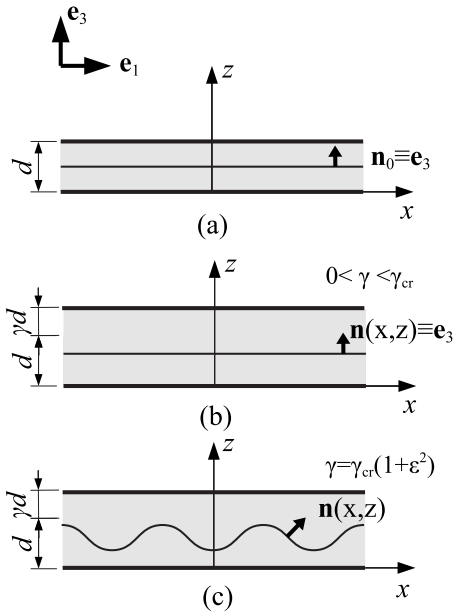

FIG. 1. Instability in layered materials: (a) natural state, (b) homogeneously deformed state, and (c) actual state right beyond the critical threshold. The actual state is obtained superposing an infinitesimal perturbation $\epsilon\hat{u}\mathbf{e}_3$ to the homogeneously deformed state at $\gamma_{cr}(1+\epsilon^2)$.

following. In their natural configuration, smectic layers are flat and the material can be described by a stack of parallel and equispaced planes. Let $\mathbf{n}_0$ be the homogeneous field of unit normal vectors for a cell of material in its natural configuration. Then, let $O(x,y,z)$ be a Cartesian coordinate system, with unit basis vectors $\{\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3\}$. It is assumed that the z axis is directed along the undistorted normal direction, $\mathbf{e}_3=\mathbf{n}_0$.

The elasticity of the material is characterized by the free energy per unit volume:

$$f_e = 2Kh^2 + \frac{B}{2}\zeta^2, \tag{2}$$

where $h=\nabla\cdot\mathbf{n}/2$ represents the layer mean curvature (or, in the liquid crystals language, the splay distortion), while ζ denotes the layers dilatation in the $\mathbf{n}$ direction. K and B are two positive constants named the curvature rigidity and the compression modulus, respectively. The quantity $\lambda=\sqrt{K/B}$ defines a characteristic length of the material.

In a plane deformation layout, it is assumed that the deformation takes place in the plane spanned by $\mathbf{e}_1$ and $\mathbf{e}_3$ while the material is taken to occupy the infinite region bounded by the planes $z=0$ and $z=d$ (see Fig. 1). Then, let a uniform finite displacement γd along the z axis be applied at the boundary $z=d$. As a result, a uniform displacement field arises in the material in the $\mathbf{e}_3$ direction. Beyond a critical displacement $\gamma_{cr}d$, layers inside the sample acquire an undulating pattern. In order to study the deformation field right beyond the critical displacement, a perturbative scheme is adopted and the boundary displacement (in units of d) is taken in the form ([22], Chap. 10)

$$\gamma = \gamma_{cr}(1+\epsilon^2), \tag{3}$$

where ϵ is a small dimensionless quantity which takes up the role of the perturbative parameter. It is worth noticing that ϵ can be kept arbitrarily small through controlling the imposed displacement, while it appears squared in (3) to yield a positive contribution to the critical threshold.

Successively, let us assume that the smectic layers are described through the isosurfaces σ_k (k labels the layer) defined by

$$\omega(\mathbf{r},k)=0,$$

where $\mathbf{r}$ denotes the position in space of a point on the surface σ_k. In particular, in the undeformed state such surfaces are equispaced planes represented by

$$\omega_0(\mathbf{r}_0,k)=\mathbf{n}_0\cdot\mathbf{r}_0-k\ell_0=0, \quad \mathbf{r}_0=x\mathbf{e}_1+z\mathbf{e}_3,$$

where ℓ_0 denotes the distance between two adjacent planes. Inasmuch as a finite homogeneous displacement γd below the critical threshold is induced, the smectic surfaces remain flat and equispaced according to the following description:

$$\bar{\omega}(\bar{\mathbf{r}}_0,k)=\mathbf{n}_0\cdot\bar{\mathbf{r}}-k\bar{\ell}=0, \quad \bar{\mathbf{r}}=\bar{x}\mathbf{e}_1+\bar{z}\mathbf{e}_3,$$

where $\bar{\ell}$ denotes the new distance between two adjacent planes. Then, the deformation is homogeneous, i.e., constant in space, and it is easy to check that

$$\bar{\ell}=\ell_0(1+\gamma), \quad \bar{x}=x, \quad \bar{z}=(1+\gamma)z. \tag{4}$$

In a perturbative approach, the critical displacement is reached and surpassed by a very small quantity, so that an infinitesimal displacement field $\epsilon\hat{u}(\bar{x},\bar{z})\mathbf{e}_3$ may be added to the homogeneously deformed state to arrive at the actual (inhomogeneously deformed) configuration. The Appendix at the end of the paper brings along a continuum mechanics argument, which yields the deformation field and the energy density expression. In the following, vector calculus is used instead and each of the two energy terms in Eq. (2) separately discussed. Such an approach appears simpler, yet it demands that the finite nature of the homogeneous deformation is explicitly taken care of. To this aim, a careful distinction is made between the natural and the homogeneously deformed configuration. Quantities pertaining to the latter are defined through an overline. Since the homogeneous deformation is finite, it is important to emphasize that the small perturbation $\epsilon\hat{u}\mathbf{e}_3$ is superposed onto the homogeneously deformed state as opposed to the natural state.

A. Curvature energy

A point in the actual configuration may be defined through the position vector

$$\mathbf{r}=x\mathbf{e}_1+[(1+\gamma)z+\epsilon u(x,z)]\mathbf{e}_3, \tag{5}$$

where the displacement field $\hat{u}(\bar{x},\bar{z})$ is here written, out of convenience, in terms of the undeformed coordinates $u(x,z)=\hat{u}(\bar{x},\bar{z})$ through Eq. (4). In the deformation plane $\{\mathbf{e}_1,\mathbf{e}_3\}$, each layer is described by a plane curve whose infinitesimal element has a length given by the expression

$$ds = \sqrt{1 + (\epsilon \partial_x u)^2} dx,$$

and no distinction is made between $\bar{x}$ and x in light of the second of Eqs. (4). Then, the tangent vector $\mathbf{t}$, the unit principal normal $\mathbf{n}$ and the principal curvature c at a point are given by [23]

$$\mathbf{t} = \frac{d\mathbf{r}}{ds}, \quad \frac{d\mathbf{t}}{ds} = c\mathbf{n}.$$

Accordingly, through the chain rule of differentiation and within second order terms in ϵ, the unit normal vector field and the principal curvature become, respectively,

$$\mathbf{n} = -\epsilon(\partial_x u)\mathbf{e}_1 + \left[1 - \frac{\epsilon^2}{2}(\partial_x u)^2\right]\mathbf{e}_3, \quad c = \epsilon \partial_{xx} u. \tag{6}$$

Now, the mean curvature of a layer h is the average between the two principal curvatures and, in a plane framework, it reduces to half the principal curvature c. It then follows that, within second-order terms in ϵ, the curvature energy density works out

$$2Kh^2 = K\frac{c^2}{2} = \epsilon^2 \frac{K}{2}(\partial_{xx} u)^2. \tag{7}$$

Within this approximation, the curvature elastic energy coincides with the classical splay energy [[1], Eq. (7.13), Sec. 7.1.2, p. 343]. It is further observed that the saddle-splay energy density term has been *a priori* omitted, because it vanishes in the case of plane deformation. More generally, the saddle-splay term is a null Lagrangian and thereby it does not contribute to the free energy when strong boundary conditions are imposed.

B. Dilatation energy

Let ℓ denote the actual distance between two adjacent undulated layers. Following de Gennes and Prost [[1], Eq. (7.50), Sec. 7.1.7, p. 365], up to the second order in ϵ, it is obtained

$$\frac{\ell}{\bar{\ell}} - 1 = \epsilon \partial_{\bar{z}} \hat{u} - \frac{\epsilon^2}{2}(\partial_{\bar{x}} \hat{u})^2, \tag{8}$$

wherein differentiation takes place with respect to the homogeneously deformed coordinates. In light of $\mathbf{r}_0$ being mapped onto $\bar{\mathbf{r}}$ through a finite transformation, $\bar{\boldsymbol{\chi}}(\mathbf{r}_0)$, such that $\bar{\mathbf{r}} = \bar{\boldsymbol{\chi}}(\mathbf{r}_0) = \mathbf{r}_0 + \gamma z \mathbf{e}_3$, a combination of the Eqs. (4) and (8) yields the layer dilatation with respect to the natural configuration

$$\zeta = \frac{\ell}{\ell_0} - 1 = \gamma + \epsilon \partial_z u - \frac{\epsilon^2}{2}(1 + \gamma)(\partial_x u)^2. \tag{9}$$

Accordingly, the layer dilatation energy density takes the form

$$\frac{B}{2}\zeta^2 = \frac{B}{2}\{\gamma^2 + 2\epsilon\gamma(\partial_z u) + \epsilon^2[-\gamma(\gamma+1)(\partial_x u)^2 + (\partial_z u)^2]\}, \tag{10}$$

within second order terms. It is interesting to note that a comparison between the free-energy density thus written [obtained through adding up Eqs. (7) and (10)] and the corresponding expression in [1,18,19] shows extra terms in the dilatation energy density contribution.

III. CRITICAL THRESHOLD

In order to assess the threshold displacement $\gamma_{cr} d$, which triggers instability, we assume that the perturbative displacement field may be written as a periodic function in the form $\epsilon u(x,z) = \epsilon a(z)\cos(q_x x)$, wherein $a(z)$ is an unknown amplitude. Homogeneous boundary conditions upon the perturbation are set

$$u(x,0) = u(x,d) = 0, \tag{11}$$

which enforce a vanishing displacement at the boundary $z=0$ and a finite imposed displacement $\gamma_{cr} d$ at the boundary $z=d$. According to the boundary conditions (11), the amplitude function $a(z)$ possesses vanishing boundary values

$$a(0) = a(d) = 0. \tag{12}$$

Then, integrating the free-energy density over a period $T_x = 2\pi/q_x$ and dividing by the period length, yields

$$F = F_0 + \frac{\epsilon^2}{4}\int_0^d \{B(a')^2 + q_x^2 a^2[Kq_x^2 - B\gamma_{cr}(1+\gamma_{cr})]\} dz, \tag{13}$$

where prime denotes differentiation with respect to z. F_0 gathers all the inessential constant terms, i.e., terms which depend neither on a nor on q_x. It should be remarked that the free energy (13) still resembles an energy density in that it pertains to a unit length.

The Euler-Lagrange equation related to the quadratic free-energy F is a linear ordinary differential equation of the second order in $a(z)$:

$$Ba'' - q_x^2[Kq_x^2 - B\gamma_{cr}(1+\gamma_{cr})]a = 0. \tag{14}$$

Therefore, the linear differential Eq. (14), coupled with the boundary conditions (12), gives an eigenvalue problem, whose non trivial solutions are in the form (with n an integer and A an undetermined parameter)

$$a_n(z) = A \sin\left(\pi n \frac{z}{d}\right), \tag{15}$$

provided that the displacement boundary value $\gamma_{cr}^{(n)} d$ complies with the second degree equation

$$B\frac{n^2\pi^2}{d^2} + q_x^2\{Kq_x^2 - B\gamma_{cr}^{(n)}[1+\gamma_{cr}^{(n)}]\} = 0. \tag{16}$$

Likewise, substituting Eq. (15) in (13), integrating and minimizing with respect to q_x it is found

$$\bar{q}_x^{(n)} = \pm\sqrt{\frac{B}{2K}\gamma_{cr}^{(n)}(1+\gamma_{cr}^{(n)})} = \pm\frac{1}{\lambda\sqrt{2}}\sqrt{\gamma_{cr}^{(n)}(\gamma_{cr}^{(n)}+1)},$$

which, plugged into Eq. (16), yields a fourth order algebraic equation in $\gamma_{cr}^{(n)}$

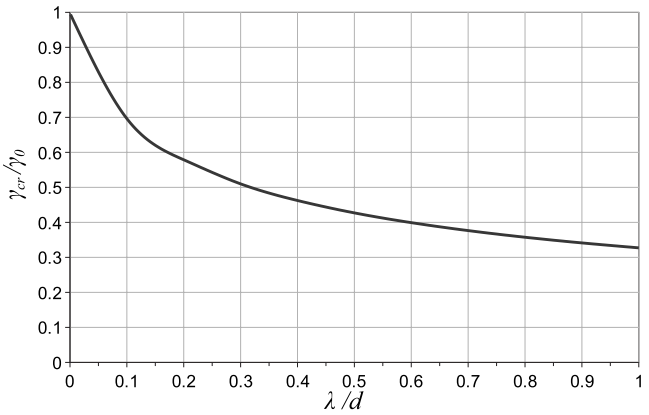

FIG. 2. The ratio γ_{cr}/γ_0 as function of λ/d. The critical threshold γ_{cr} reduces to γ_0 as λ/d tends to zero, while significant disagreement occurs for a specimen thickness comparable to λ.

$$[\gamma_{cr}^{(n)}(1+\gamma_{cr}^{(n)})]^2 = 4\lambda^2\frac{n^2\pi^2}{d^2}.$$

The sole physically meaningful solution (which does not lead to either interpenetration or complex values) works out to be $\gamma_{cr}^{(n)}=(-1+\sqrt{1+8n\pi\lambda/d})/2$. Obviously, the critical threshold γ_{cr} is the minimum value within the set $\{\gamma_{cr}^{(n)}\}$, which is indeed retrieved at $n=1$,

$$\gamma_{cr} = -\frac{1}{2}+\frac{1}{2}\sqrt{1+8\pi\frac{\lambda}{d}}, \tag{17}$$

for which it is

$$\bar{q}_x = \pm\sqrt{\frac{\pi}{\lambda d}}. \tag{18}$$

It is easy to check (see Fig. 2) that the critical threshold thus obtained is smaller than the one predicted by the classical theory, while the undulation wavelength $2\pi/\bar{q}_x$ remains unchanged.

Up to this point, the solution to the linear problem is incomplete, for its amplitude A rests undetermined. Indeed, the nonuniform displacement is written in the form

$$\epsilon u(x,z) = \epsilon A\cos\left(\sqrt{\frac{\pi}{\lambda d}}x\right)\sin\left(\frac{\pi}{d}z\right). \tag{19}$$

In order to assess the displacement amplitude ϵA, fourth order terms in the energy expansion are retained

$$\begin{aligned}F = F_0 + \frac{\epsilon^2}{4}\int_0^d \{B(a')^2 + \bar{q}_x^2[K\bar{q}_x^2 - B\gamma_{cr}(1+\gamma_{cr})]a^2\}dz \\ + \frac{1}{16}\epsilon^4\bar{q}_x^2\int_0^d\left\{-3a^2\bar{q}_x^4K + \left[\frac{3}{4}(4\gamma_{cr}+1)(1+\gamma_{cr})\bar{q}_x^2a^2 \right.\right. \\ \left.\left. - 4(2\gamma_{cr}+1)\gamma_{cr} - (a')^2\right]B\right\}a^2dz\end{aligned} \tag{20}$$

and Eqs. (15), (17), and (18) are employed. Having performed the integration, it remains to minimize the energy with respect to the amplitude A. Details on the general procedure may be found in [15,20,24]. It is thus found

$$A = \pm 4d\sqrt{\frac{2\eta\varpi(\varpi-1)}{\pi(1+9\varpi+8\varpi^2)}}, \tag{21}$$

having let $\eta=\lambda/d$ and $\varpi=\sqrt{1+8\pi\eta}$. In the limiting case $8\pi\eta \ll 1$, the layer's peak profile ϵA is found to be

$$\epsilon A = \pm\frac{8}{3}\lambda\sqrt{\frac{\gamma}{\gamma_{cr}}-1}\left[1-\frac{16\pi}{9}\frac{\lambda}{d}+O\left(\frac{\lambda^2}{d^2}\right)\right]. \tag{22}$$

As opposed to the classical model, wherein the amplitude is simply $\epsilon A_0 = \pm(8/3)\lambda\sqrt{\gamma/\gamma_0-1}$ ([20], Sec. 2.5.1), here, for a given ϵ, the amplitude depends on λ/d. Equation (22) shows that the amplitude reduces to the classical result in a thick cell limit.

IV. DISCUSSION

As shown, phase transition occurs in traction beyond the critical value γ_{cr} given by Eq. (17), whereas the well known classical model threshold value γ_0 [1,18,19] is given by Eq. (1). An attempt to explain the difference is put forward hereinafter. The quantity $\zeta=\frac{\ell}{\ell_0}-1$ measures the dilatation of a layer with respect to the undeformed configuration. Up to second order terms in ϵ, the layer dilation is given by Eq. (9)

$$\zeta = \gamma_{cr} + \epsilon\partial_z u - \frac{\epsilon^2}{2}(1+\gamma_{cr})(\partial_x u)^2$$

as opposed to the corresponding result presented in [1]

$$\zeta_{dG} = \gamma_0 + \epsilon\partial_z u - \frac{\epsilon^2}{2}(\partial_x u)^2,$$

wherein the second order term $-\gamma(\partial_x u)^2/2$ appears to be missing. Indeed, in [1,18,19] infinitesimal displacements are added to the homogeneous deformation starting from the natural configuration whereas, in view of the nonlinearity of the problem, a more conservative approach would require starting from the homogeneously deformed configuration. In fact, it may be seen that the free-energy density expression adopted in ([1], p. 365, Eq. 7.53) neglects the energy term $-\epsilon^2B\gamma^2(\partial_x u)^2/2$. As a consequence, the minimization procedure of Sec. 7.1.7 in [1] leads to a layer equilibrium equation which differs from Eq. (14).

It may be objected that this disagreement is never brought out by experimental results on liquid crystals. Indeed, there are two different ways to extract the smectic coherence length: either by measuring the critical threshold or by detecting the undulation wave vector. The reason may be that in typical experimental conditions [18,20], authors employ specimen samples whose thickness is in the order of several hundred micrometer, that is far greater than the smectic coherence length, which ranges about a few tens of angstrom. Inasmuch as $\lambda \ll d$, it is

$$\gamma_{cr} = \frac{2\pi\lambda}{d}+O\left(\frac{\lambda^2}{d^2}\right) = \gamma_0 + O\left(\frac{\lambda^2}{d^2}\right),$$

which shows that no appreciable difference exists between the two expressions. In other words, for specimen samples,

which present a critical displacement that is small with respect to d, the addition of the perturbed infinitesimal displacement to either the natural or the homogeneously deformed configuration leads to comparable outcomes, which amounts to saying that infinitesimal perturbation and homogeneous deformation commute in the limit of a small threshold compared to the specimen thickness.

Tangentially, it may be remarked that the thin cell limit appears to be particularly relevant for bilayer lipid membranes. Such materials are usually employed in the design of biosensors aimed at detecting biological agents [9] and their actual application seems to be hindered by their poor stability to environmental disturbances such as, for instance, mechanical stresses. In a recent paper [25], bilayer lipid membranes are modeled as smectic-A liquid crystals and the effect of hydrostatic pressure on their equilibrium configuration is investigated.

In order to put forward a numerical comparison between the expressions for the thick cell critical threshold and the classical one, we borrow the constitutive constants currently available in the experimental literature for 1-stearoyl-2-oleoyl-3-sn-phosphatidylcholine [26,27], $K=10^{-6}$ dyne, $B=5\times10^{-8}$ dyne Å^{-2}, and set the sample thickness $d=50$ Å [28]. A critical threshold $\gamma_{cr}=0.40$ is found as opposed to the classical value $\gamma_0=0.56$, which shows a 29% difference. Nonetheless, in this instance a comparison between the layer profiles appears inappropriate. Indeed, for an applied boundary displacement right beyond the classical threshold $\gamma_0 d$, the corresponding value of ϵ, as defined in (3), is quite large, as a result of the fact that the thick cell threshold γ_{cr} is significantly smaller. As a consequence, the actual deformation is quite a long way far from being in the neighborhood of instability and the linear theory does not lend reliable results. On the contrary, if we were to choose γ just beyond γ_{cr}, the instability would not take place in the classical framework. In any case, given that the perturbation amplitude is proportional to ϵ which, in turn, expresses the distance from the critical threshold, different models yield different ϵ and different amplitudes alike. In this light, a comparison between the two amplitude expressions becomes meaningful in the limit of $8\pi\lambda/d \ll 1$. In fact, within this limit $\gamma_0 \approx \gamma_{cr}$ and the amplitude ratio reduces to

$$\frac{A}{A_0} \approx 1 - \frac{16\pi}{9}\frac{\lambda}{d}.$$

In conclusion, we provide a threshold expression for lamellar systems subjected to tensile strains. Our result arises owing to a geometrical nonlinearity in the expression of the layers dilatation, which, in our opinion, the classical computation does not take into account. Figure 2 shows the ratio between the critical threshold expressions as a function of the dimensionless quantity λ/d. It may be observed that significant disagreement occurs for a specimen thickness comparable with λ, which suggests that a thin specimen experiment be carried out to effectively test the threshold critical value. The experimental appreciation of the correction to the linear theory is perhaps hard to obtain in the case of liquid crystals, standing the stress relaxation and strain rate dependence which accompany their deformation [19] and the small characteristic length λ that they possess. Nonetheless, different actual materials fall into the class of layered materials, some of which exhibit wider stability to the deformation and a higher-characteristic length. A nice review of the perspective that the mechanics of layered material instability lends in every day experience, ranging from human skin to lipid bilayers vesicles wrinkling, is provided in [29]. Likewise, in [8] wrinkling instability is cast within the broad framework of layered material as a compromise between curvature and stretching energy, and it is pointed out that observation confirms that thin sheets wrinkle easily in tension, well below the elastic limit, so that a linear theory is of little use and we must consider the geometrically nonlinear behavior of the wrinkles.

ACKNOWLEDGMENTS

This work has been carried out under the research program "Modellazione teorica e numerica di elastomeri smettici," supported by the *Young researchers project* of the *Gruppo Nazionale per la Fisica Matematica (GNFM).*

APPENDIX: CONTINUUM MECHANICS ARGUMENT

In this section the unit normal vector to the layers, their dilatation and the free-energy density are deduced through continuum mechanics arguments. Following [30]

$$|\nabla\omega|\ell = \ell_0,$$

where ℓ denotes the distance between two adjacent planes in the distorted state, and, as previously defined, ℓ_0 represents the same distance in the undeformed situation. Therefore, $|\nabla\omega|^{-1}$ measures the thickness of the layers in units of ℓ_0. In particular, if $|\nabla\omega|>1$ layer compression (with respect to the reference placement) is found, whereas $|\nabla\omega|<1$ amounts to layer dilatation. Hence, for finite deformations, the following expression for the layer dilatation is retrieved

$$\zeta = |\nabla\omega|^{-1} - 1. \tag{A1}$$

Recalling the assumption that the smectic layers are described by the material surfaces σ_k (k labels the layer), defined by

$$\omega(\mathbf{r},k) = 0, \tag{A2}$$

where $\mathbf{r}$ defines the position in space of a point on the surface σ_k, the same expression may be written in the undeformed state wherein the smectic layers are equidistant planes

$$\tilde{\omega}(\mathbf{r}_0,k) = \mathbf{n}_0\cdot\mathbf{r}_0 - k\ell_0 = 0, \quad \mathbf{r}_0 = x\mathbf{e}_1 + z\mathbf{e}_3. \tag{A3}$$

Let us consider the invertible transformation $\mathbf{r}=\boldsymbol{\chi}(\mathbf{r}_0)$, which maps the undeformed configuration in the distorted one. By using its inverse $\mathbf{r}_0=\boldsymbol{\chi}^{-1}(\mathbf{r})$, Eq. (A3) yields

$$\omega(\mathbf{r},k) = \mathbf{n}_0\cdot\boldsymbol{\chi}^{-1}(\mathbf{r}) - k\ell_0 = 0. \tag{A4}$$

Now, let the deformation gradient $\mathbf{F}$ be considered, i.e., the Jacobian of the transformation $\boldsymbol{\chi}$. Taking the spatial gradient of Eq. (A2), it is found

$$\nabla\omega = \frac{\partial\omega}{\partial\mathbf{r}} = \frac{\partial\tilde{\omega}}{\partial\mathbf{r}_0}\frac{\partial\mathbf{r}_0}{\partial\mathbf{r}} = \mathbf{F}^{-T}\frac{\partial\tilde{\omega}}{\partial\mathbf{r}_0},$$

where the superscript $-T$ denotes the inverse of the transpose. Hence, taking into account Eq. (A3),

$$\nabla\omega = \mathbf{F}^{-T}\mathbf{n}_0 = \mathbf{F}^{-T}\mathbf{e}_3. \tag{A5}$$

Letting a special form to the general expression of the deformation

$$\mathbf{r} = \boldsymbol{\chi}(\mathbf{r}_0) = \mathbf{r}_0 + U(x,z)\mathbf{e}_3,$$

it is found

$$\mathbf{F} = \begin{pmatrix} 1 & 0 \\ \partial_x U & 1+\partial_z U \end{pmatrix}, \quad \mathbf{F}^{-T} = (1+\partial_z U)^{-1}\begin{pmatrix} 1+\partial_z U & -\partial_x U \\ 0 & 1 \end{pmatrix},$$

respectively deformation gradient matrix and its inverse transposed in the basis set $\{\mathbf{e}_1,\mathbf{e}_3\}$. Then, in view of Eq. (A5), it is

$$|\nabla\omega| = (1+\partial_z U)^{-1}\sqrt{1+(\partial_x U)^2}, \tag{A6}$$

thereby, setting the displacement expression

$$U(x,z) = \gamma z + \epsilon u(x,z) \tag{A7}$$

into Eq. (A6), Eq. (A1) yields, up to second order terms in ϵ, the layer dilatation Eq. (9). The layer dilatation energy density, Eq. (10), follows immediately by the definition. Finally, the unit normal vector to the lamellae, in the deformed configuration, is given by

$$\mathbf{n} = \frac{\nabla\omega}{|\nabla\omega|}. \tag{A8}$$

Making use of Eq. (A5) and (A6), given the special form of the deformation expression (A7), the first of Eqs. (6) is retrieved. The layer mean curvature is obtained taking the spatial divergence of (A8)

$$h = \tfrac{1}{2}\mathrm{div}\ \mathbf{n} = \tfrac{1}{2}\mathbf{F}^{-T}\cdot\mathrm{Grad}\ \mathbf{n}, \tag{A9}$$

whence Eq. (7) is found.

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