Undulations of lamellar liquid crystals in cells with finite surface anchoring near and well above the threshold

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We study the undulations instability, also known as the Helfrich-Hurault or layers buckling effect, in a cholesteric liquid crystal confined between two parallel plates and caused by an electric field applied along the normal to layers. The cholesteric pitch is much smaller than the cell thickness but sufficiently large for optical

field increase rate is much lower than the rate of undulations growth.

We demonstrate that the anisotropic forces of surface anchoring responsible for layers alignment play a crucial role at the onset and development of undulations. A finite surface anchoring strength leads to the lower threshold of undulation instability, larger layers' displacements, and tilts in the bulk. When the field increases well above the threshold, the sinusoidal undulations first evolve into a chevron or zigzag pattern with an increased period [similarly to the two-dimensional (2D) case [13–16] and in accord with the recent theory by Singer for 3D [17]]. This transformation from a single Fourier mode into a zigzag pattern is accompanied by a weakened dependence of the layers shape on the vertical -coordinate; the finite surface anchoring facilitates the transformation as the layers become strongly tilted not only in the bulk but also at the surfaces. Well above the threshold the pattern transforms again, but not into the anticipated pattern of parabolic focal conic domains: rather unexpectedly, the 2D square pattern of zigzag undulations transforms into a 1D periodic pattern formed by a system of parabolic walls (PWs), which has never been described before. The PWs balance the dielectrically induced layers reorientation in the bulk and surface anchoring at the boundaries, managing to avoid unfavorable tilted orientation of layers near the surfaces, but allowing it in the bulk.

The outline of this paper is as follows. In Sec. II, after a short review of the field, we present the basic theory of undulations in a 3D lamellar system with finite surface anchoring. Section III describes materials and the experimental techniques. We discuss our main results and draw conclusions in Secs. IV and V, respectively.

II. LAYERS UNDULATIONS IN CHOLESTERIC LAMELLAE

A. Undulations instability in layered systems

Since pioneering works of Helfrich [4,5], layers undulations under mechanical, temperature, electric, and magnetic field action as well as shear [18,19] have been studied for smectic *A* [6,7,13,14,17,20–24], smectic *C* [25], and cholesteric [15,16,26–34] LCs, aqueous DNA solutions [35], lyotropic lamellar LCs [19,36], lamellae-forming block copolymers [18,37,38], LC elastomers [39,40], ferrofluids [41], ferrimagnets [42,43], and multiwall carbon nanotubes [44]. Wrinkling of thin elastic sheets [45] and undulations of columnar LC phases [46] might also be added to the list.

The original model assumed that the undulations develop only along one direction in the plane of the sample, Fig. 1(c). Such a model would describe a periodic buckling in 2D systems such as magnetic stripe phases [43], ferrofluids [41], or cholesteric "fingerprint" textures [15,16]. At the onset of instability, the layers profile in the bulk is well-described by a sinusoidal line [13]. As the field increases, the sinusoidal profile (a) evolves into the sawtooth (called also zigzag, chevron, or kink) structure and (b) increases its periodicity [13]. These trends have been observed in experiments with (effectively 2D) ferrimagnetic films [43] and 2D LC samples, both in part (a) [14–16] and (b) [16].

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above the threshold [47]. The layers change their topology and fold into a square lattice of parabolic focal conic domains (PFCDs). The layers are in the form of Dupin cyclides for which the focal surfaces degenerate into 1D singularities, in this case a pair of parabolae that pass through each other's focus [3]. The PFCDs are capable of relaxing the dilative strain because the layers within the PFCD are tilted and also because layers within a certain portion of the PFCD are multiply connected (

reaches some threshold value $\square_c^{\square}$, the ordinate of the two minima of $g(\square_\perp)$ decreases to zero, Fig. 2(a), so that Eq. (6) with $W_{\square 0} \to \square$ is satisfied for some $\square_\perp = \pm \square_{\perp c}$. The latter value is the critical wave number of undulations that become energetically preferable above the threshold electric field, $\square^{\square} \,\square\, \square_c^{\square}$.

$$\square^2 \frac{\partial^2 \square}{\partial\ ^2} + \square' \square - \frac{1}{2}\square^3 = 0 \qquad (17)$$

with $\square' = \frac{\square_0 |\square_a| E^2}{2B}$ is of the same form as in Refs. [13,14,16]. Therefore the spatial dependence of the tilt angle is

$$\square(\ ) = \square_{\ ax}\, \mathrm{sn}\left(\frac{\sqrt{2\square - \square^2_{\ ax}}}{2\square} \ \middle|\ \right), \qquad (18)$$

where $\mathrm{sn}(g|\ )$ is the Jacobi elliptic function [54], $^2 = \frac{\square^2_{\ ax}}{2\square - \square^2_{\ ax}} \square 1$ is the field-dependent parameter, and $\square_{\ ax}$ is the maximum tilt angle of the layers [16]. The displacement $.(\ )$ is obtained by integrating Eq. (18)

$$.(\ ) = \frac{\square_{\ ax}}{A\sqrt{\ }} \log[\mathrm{dn}(A|\ ) - \sqrt{\ }\,\mathrm{cn}(A|\ )] + \mathrm{const}, \qquad (19)$$

where $A = \frac{\sqrt{2\square - \square^2_{\ ax}}}{2\square}$, $\mathrm{cn}(g|\ )$ and $\mathrm{dn}(g|\ )$ are the Jacobi elliptic functions [54]. In the limit $\ \to 0$, the undulation profile $.(\ )$ is sinusoidal, while for $\ \to 1$ it is of a zigzag character with a longer period. As we shall see in Sec. IV, the experimental data on the 3D system are in a good agreement with these predictions and with the data for 2D systems [13,14,16].

III. MATERIALS AND EXPERIMENTAL TECHNIQUES

A. Materials

The LC cells were assembled from glass plates coated with transparent indium tin oxide (ITO) electrodes. To study the role of surface anchoring we used different alignment materials. Thin films of polyimide PI2555 (HD MicroSystem) and poly(vinyl alcohol) (PVA; Aldrich Chemical Company, Inc.) aqueous (

B. Imaging techniques

Polarizing microscopy observations were performed using a Nikon Eclipse E600 microscope equipped with a Hitachi HV-C20 CCD camera. The FCPM studies were performed using the modified BX-50 Olympus microscope [9]. By using the nematic host with low birefringence, □ =0.078, we mitigated the problem of beam defocusing and the Mauguin effect in the FCPM imaging [57,58]. The Ar laser (□ =488 nm) was used for excitation of the BTBP and the fluorescent light was detected in the spectral range 510–550 nm. It is important to note that in the FCPM images the registered fluorescence signal from the bottom of the cell can be weaker than from the top because of some light absorption, light scattering caused by director fluctuation, depolarization, and defocusing [9]. These effects are especially noticeable in

Figs. 4, 5, and 7

C. High-field unidirectional buckling with parabolic walls

As the applied electric field slowly ($\sim$50 mV/min) increases to $E \approx 1.8$ to $1.9E_c$, the undulations pattern changes one more time, as the 2D square lattice is replaced by a system of 1D stripes bounded by parabolic walls (PWs). The stripes with parabolic walls appear through a nucleation process that starts with the appearance of axially symmetric domains (ASDs), usually at the sites of surface imperfections, Fig. 10. When the ASD radius becomes comparable to d, it does not grow radially anymore, but transforms into a tip of growing stripe, Fig. 11(a). At a fixed voltage, the stripes slowly (□50 □m/s at $E=1.9E_c$) propagate replacing the original 2D square lattice of undulations, Fig. 11(b). Experimentally, the period L of the 1D stripe pattern is about two times larger than the period L_x of the square lattice and comparable to the cell thickness, $L/d \approx 1$. Even in the cells with rubbed substrates there was no preferred direction for the growth of 1D stripes within the 2D square lattice, Fig. 11(a). As would become clear from the discussion below, this is a natural consequence of the peculiar structure of stripes in which the layers near the boundaries are not tilted much and thus avoid the influence of in-plane surface anchoring.

The configuration of layers in stripes with PWs reconstructed from the 3D FCPM observations, Fig. 12(a), reveals that these structures are uniquely suited to balance the dielectric and surface anchoring forces, by combining titled layers in the bulk with po5y7

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The geometrical model in Fig. 12(b) captures the essential large-scale features of the stripe domains; however, at the scale of the pitch, the details are different. For example, the layer closest to the parabola focus is not cylindrical but forms a pair of disclinations with nonsingular cores, □$^{+1/2}$ and □$^{-1/2}$, Figs. 11(c) and 12(a). The vertex of the parabola O is located at a finite distance $/2$ from the bounding plate, to accommodate for the nonsingular core of the □$^{-1/2}$ disclination, while the core of the □$^{+1/2}$ is at the parabola's focus, at the distance $h \approx \ /2$distanthe

bounding walls, thus avoiding a large energy associated with surface anchoring.

The present work deals with a quasistatic regime of undulation development. This scenario is expected to be different when the field is applied abruptly [22]. The studies of undulation dynamics are in progress.

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