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Undulations in a confined lamellar system with surface anchoring

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We visualize undulations in layered systems using a cholesteric stripe phase with a macroscopic supramicron periodicity. The wave vector of stripe pattern is in the cell's plane. The undulation is induced by an in-plane magnetic field normal to the stripes. The observed displacement of layers is much larger than the value predicted by the Helfrich-Hurault classic theory. We propose a model of undulations that explains the data by finite surface anchoring of layers.

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I. INTRODUCTION

A variety of condensed phases possess reduced one-dimensional (1D) (smectic) or 2D (columnar phases) translational order that allows long-range curvature deformations (splay in smectic, bend in columnar phases) [1]. Curvature deformations are capable of relaxing dilation or field-induced stress. In many systems (smectic *A* [2,3], cholesteric [4] and columnar [5,6] liquid crystals, diblock copolymers [7,8], periodic patterns in ferrofluids [9], and ferrimagnets [10,11], etc.), the dilation-curvature coupling shows up as the undulation instability, often also called buckling or Helfrich-Hurault effect [12]. The mechanism of the phenomenon is as follows. The lamellar phase is confined between two flat plates; the layers are parallel to these plates. The magnetic field is applied normally to the plates and tends to reorient the layers. If there were no bounding plates, the layers would uniformly tilt and realign along the field. In reality, the surface anchoring at the plates does not allow the adjacent layers to rotate freely. As a result, the layers undulate with the tilt angle changing sign periodically in the plane of the cell. Undulations can be caused by other means, e.g., by mechanical tension [1]. The classic Helfrich-Hurault theory of the phenomenon [1,12] and all subsequent modifications [13–15] assume that the undulations vanish at the cell boundaries; i.e., the layers are clamped by an infinitely strong surface anchoring.

To determine the actual pattern of layers displacements and to verify the predictions of the theory we design a ''uniform fingerprint'' cholesteric texture as the model of undulating lamellar structure. The cholesteric pitch is large enough ($P \approx 15$ μm) to visualize the layers under a polarizing microscope, but small as compared to the characteristic radius of distortions. The last feature allows us to treat the cholesteric phase as a 1D periodic lamellar phase within the Lubensky–de Gennes coarse-grained model [1]. Experiments reveal that the layers displacements are much larger than one would expect from the classic Helfrich-Hurault theory and do not vanish at the bounding surfaces. We refine the theory by adding a finite surface anchoring term to the free energy functional; soft anchoring explains why the displacements are larger than in the Helfrich-Hurault model. Fitting of the experimental data allows us to determine the strength of the surface anchoring.

II. EXPERIMENT

The model system with an undulating stripe pattern is created in two steps: (i) obtaining a uniform cholesteric fingerprint texture [16]; (ii) generation of undulations by a magnetic field in the plane of the cell.

(i) The cell is assembled from a pair of glass plates coated with transparent (ITO) electrodes and an alignment material JALS 214 (Japan Synthetic Rubber) that sets homeotropic boundary conditions. Two mylar strips are placed between the glass plates parallel to each other, separated by a distance $a = 1.7$ mm in the plane of the cell. The mylar films fix the distance $l = (15.7-16)$ μm ($\approx P$) between the glass plates (along the *y* axis in Fig. 1) and serve as ''walls'' for the uniform fingerprint texture of the cholesteric liquid crystal filling the gap between the glass plates and the mylar strips. We used the cholesteric mixture of 4-*n*-pentyl -4′cyanobiphenyl (5CB) and 4-(2-methylbutyl) −1−cyanobiphenyl (CB15) in weight proportion 99.07:0.93. Uniform orientation of the cholesteric stripes parallel to the mylar strips (the *x* axis in Fig. 1) is achieved in two steps. First, an electric field is applied to the indium tin oxide (ITO) electrodes to unwind the cholesteric helix, and

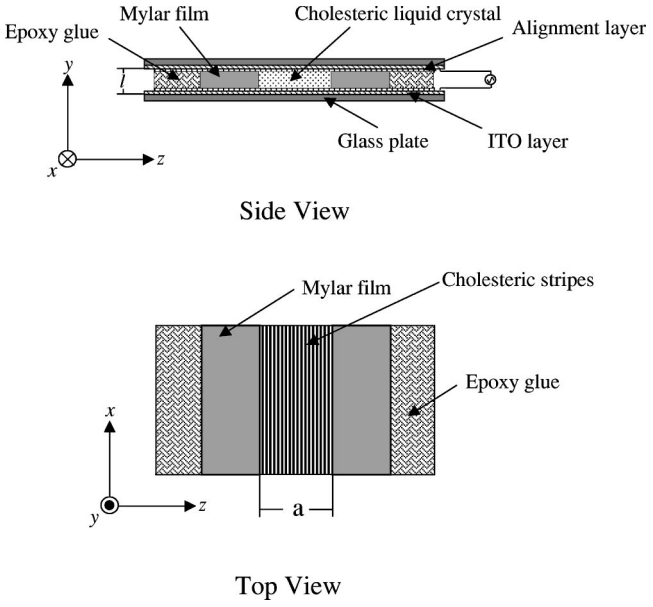

FIG. 1. Geometry of a sample.

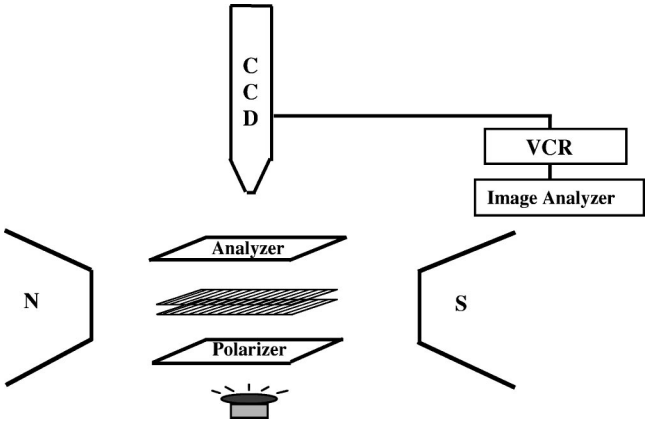

FIG. 2. Experimental setup.

then it is switched off to allow the cholesteric fingers to grow parallel to the magnetic field acting along the x axis. The stipe periodicity $w=16.5$ μm is close to P (and not to $P/2$; see [17]).

(ii) Once the stripes are grown, the magnetic field is applied in the direction z *perpendicular* to the cholesteric stripes to cause undulations along the x axis; see Fig. 2. The number of layers remains constant. The field was raised with the increment of 0.05 T and kept constant until the system shows no signs of evolution ($\geqslant 1$ hr).

III. RESULTS AND DISCUSSION

Figure 3 shows the field dependence of the displacement amplitude u_0 (along the z axis) of the layer initially in the middle of the cell, $z=0$. According to the classic theory, just above the threshold field H_c [18],

$$u_0=\frac{8\lambda}{3}\left(\frac{H^2}{H_c^2}-1\right)^{1/2}, \tag{1}$$

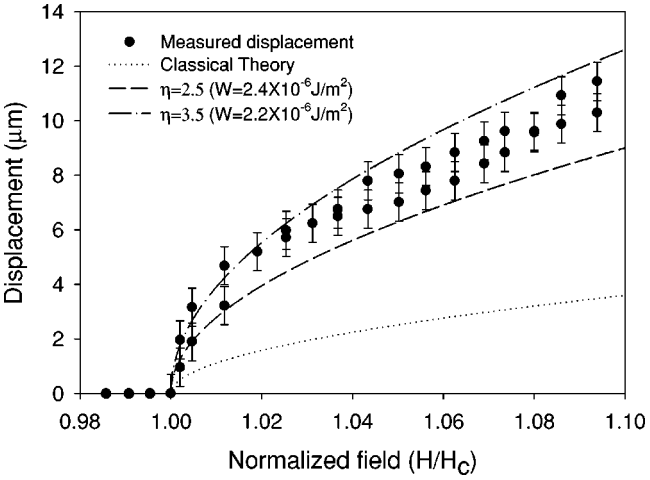

FIG. 3. Comparison of the measured displacement amplitude with the theory. Dotted line shows u_0 predicted by the classical theory, Eq. (1). The measured displacement falls between the two lines, Eq. (9) with $\eta=2.5$ and 3.5. With the value $B=0.44$ J/m^3 estimated from the coarse-grained theory, the upper and the lower curves correspond to $W=2.2$ and 2.4×10^{-6} J/m^2, respectively.

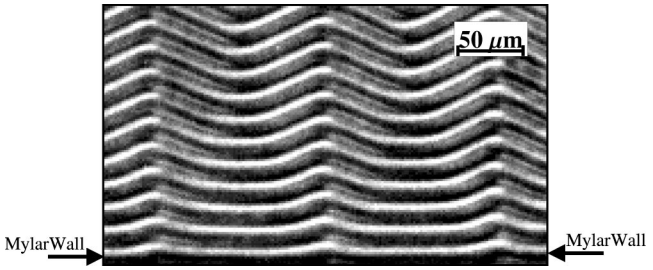

FIG. 4. Undulation pattern near the mylar wall ($H=1.05H_c$).

i.e., the function $u_0(H/H_c)$ depends only on one material parameter, the elastic length $\lambda=\sqrt{K/B}$ defined by the ratio of the curvature constant K to the compression modulus B of the stripe phase; the threshold field

$$H_c=\frac{2\pi K}{a\lambda|\chi_a|}$$

depends also on the diamagnetic anisotropy χ_a of the material. The experimental data $u_0(H/H_c)$ can be approximated by Eq. (1) only when $\lambda=(8.5\pm1.7)$ μm. On the other hand, according to the Lubensky–de Gennes theory, $K\approx3K_3/8$ and $B\approx(2\pi/w)^2K_2$, so that with the known bend $K_3\approx10^{-11}N$ and twist $K_2\approx0.3\times10^{-11}N$ elastic constants of 5CB [19], one expects much smaller length $\lambda\approx2.9$ μm. Intrigued by the discrepancy, we measured λ independently, by fitting the profile of an elementary dislocation, as described in [16], and found that for our system $\lambda=(2.9\pm0.1)$ μm, i.e., λ is indeed too small to allow Eq. (1) to describe the data in Fig. 3. Thus, the most plausible source of discrepancies between the experiment and the theory is the form of Eq. (1) itself.

Equation (1) was derived in the approximation that the layers displacement is strictly zero at the boundaries [1,12]. Closer examination of the undulations reveals that the displacement is actually nonzero, Fig. 4. Below we refine the theory by taking into account the finite surface anchoring at the walls.

The free energy of the system, assumed periodic along the x-direction, $u(x)\sim\sin q_x x$, writes (per one period $2\pi/q_x$) as

$$F=\int_0^{2\pi/q_x}dx\int_{-a/2}^{a/2}dz\left\{\frac{B}{2}\left[\partial_z u-\frac{1}{2}(\partial_x u)^2\right]^2+\frac{K}{2}(\partial_{xx}u)^2+\frac{\chi_a H^2}{2}(\partial_x u)^2\right\}+\frac{W}{2}\int_0^{2\pi/q_x}dx[(\partial_x u)^2_{z=-a/2}+(\partial_x u)^2_{z=a/2}], \tag{2}$$

where the surface term with the anchoring coefficient W is taken proportional to $(\partial_x u)^2\equiv(\partial u/\partial x)^2$ [1]. It is a legitimate assumption since the tilt $\partial_x u$ of layers is small and changes sign periodically along the x axis. A coherent tilt with $\partial_x u=$const would require a lattice of dislocation and a surface term $\sim|\partial_x u|$.

We first derive H_c and the undulation wavelength $2\pi/q_{xc}$ at H_c; for these calculations, the fourth order term in u in Eq. (2) can be disregarded [12]. We relax the condition $u(z=\pm a/2)=0$ and solve the Euler-Lagrange equation with boundary conditions following from Eq. (2). This yields the standard solution

$$u(x,z)=u_0\cos q_z z \sin q_x x, \tag{3}$$

with constraints on the wave vectors q_x and q_z,

$$q_z=q_x\sqrt{\kappa-\lambda^2 q_x^2}, \tag{4}$$

$$\frac{B}{W}=\frac{q_x\cot(aq_x\sqrt{\kappa-\lambda^2q_x^2}/2)}{\sqrt{\kappa-\lambda^2q_x^2}}\equiv g(q_x), \tag{5}$$

where $\kappa=|\chi_a|H^2/B$. The function $g(q_x)$ is even in q_x with two minima. When the abscissa of the two minima is B/W, the coordinates of the minima are $\pm q_{xc}$. Minimization of $g(q_x)$ gives the condition $\kappa_c=(\lambda^2 a/\alpha)q_{xc}^2$, which allows us to find the critical field

$$H_c^2=\frac{\lambda^2 aB}{\alpha|\chi_a|}q_{xc}^2, \tag{6}$$

and the relationship between q_{xc} and q_{zc} from Eq. (4),

$$q_{xc}^2=\frac{q_{zc}}{\lambda}\sqrt{\frac{\beta}{\alpha}}, \tag{7}$$

where

$$\alpha=a\left(1-\frac{\sin q_{zc}a}{q_{zc}a}\right)\Big/2 \quad \beta=a\left(1+\frac{\sin q_{zc}a}{q_{xc}a}\right)\Big/2$$

.

For $W\to\infty$, Eqs. (5)–(7) recover the results of the classic theory [1,12]: $H_c=2\pi K/a\lambda|\chi_a|$, $q_{xc}^2=\pi/a\lambda$, and $q_z=\pi/a$.

In order to calculate the displacement immediately above H_c, we retain the fourth order term in Eq. (2). With Eq. (3), the energy density per one period of undulation reads

$$\tilde{f}=F\frac{q_x}{2\pi a}=\frac{q_z|\chi_a|}{4\lambda a}\sqrt{\alpha\beta}\{H_c^2-H^2\}u_0^2+\frac{3Kq_z\alpha\rho}{1024\lambda^4\beta a}u_0^4, \tag{8}$$

where $\rho=6aq_{zc}+8\sin q_{zc}a+\sin 2q_{zc}a$. Minimization of Eq. (8) yields the dependence $u_0(H)$ above H_c Eq. (6),

$$u_0=\frac{8\lambda\eta}{3}\left(\frac{H^2}{H_c^2}-1\right)^{1/2}; \quad \eta=q_{xc}\left(\frac{6\lambda a}{\rho}\right)^{1/2}\left(\frac{\beta}{\alpha}\right)^{3/4}. \tag{9}$$

The last expression reduces to the Helfrich-Hurault result, Eq. (1), $\eta=1$, when $W\to\infty$ (as easy to see by calculating ρ, α, and β with $q_{zc}=\pi/a$).

The coefficient η in Eq. (9) depends on λ, a, and W/B [through the dependencies of ρ, α, and β on q_{zc}, which are the functions of W/B, see Eqs.(5), (7)]. A good fit of the data in Fig. 3 is obtained for $\eta=(3.0\pm0.5)$, and measured independently $\lambda=2.9$ μm and $a=1.7$ mm in Eq. (9).

The fitted values of η correspond to $W/B=(5.2\pm0.3)$ μm, which is of the order of the characteristic length of the cholesteric phase. The correspondence is established in the following way. First, we chose some value of W/B, and obtain the corresponding numerical values of q_{xc}, q_{zc}, α, β, and ρ from Eqs. (5)–(7) and then calculate η from Eq. (9) and compare it to the fitted value $\eta=(3.0\pm0.5)$. Using the coarse-grained value of the modulus $B\approx K_2(2\pi/w)^2\approx0.44$ J/m^3 and the result $W/B=(5.2\pm0.3)$ μm, we determine the anchoring strength $W=(2.3\pm0.1)\times10^{-6}$ J/m^2. The value of W agrees in the order of magnitude with a dimensional estimate $W\approx K_2(2\pi/w)\simeq10^{-6}$ J/m^2 that treats the surface anchoring as the ''intrinsic'' anchoring of a lamellar system [20,21] caused by a violation of layers equidistance near the surface. The same estimate $W\approx K_2(2\pi/w)$ follows from the studies of cholesteric oily streaks [22]. Note also that the finite W calculated above reduces the threshold field H_c [see Eq. (6)] by a factor of ≈0.8 as compared to its classical value at $W\to\infty$.

In summary, we have determined the pattern of displacements in an undulating 1D periodic system, both in the bulk and at the bounding surfaces. The displacement amplitude is few times larger than the one predicted by the classic theory [1,12]; it does not vanish at the boundaries. The pattern is explained by taking into account that the anchoring energy penalty for layers tilt is finite. Finite surface anchoring also decreases the threshold of undulations. The model can be applied to other undulating system to determine, for example, the strength of surface anchoring for materials with various elastic lengths λ. Furthermore, the present study should be extended from the immediate vicinity of the undulation threshold to the region of high fields (stresses), where the weakness of the surface anchoring becomes even more important. For example, our preliminary studies indicate that at $H\geqslant1.6H_c$ the tilt of the layers at the boundaries becomes practically equal to the tilt in the bulk, i.e., the approximation of the infinite anchoring fails completely. Analytical treatment of the high-stress region should take into account higher harmonics of undulations, since the layers acquire a saw-tooth profile.

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