# Shear Instabilities of Freely Standing Thermotropic Smectic-*A* Films

Hsuan-Yi Chen and David Jasnow

*Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, Pennsylvania 15260*

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In this Letter we discuss theoretically the instabilities of thermotropic freely standing smectic-*A* films under shear flow [3]. We show that, in Couette geometry, the centrifugal force pushes the liquid crystal toward the outer boundary and induces smectic layer dilation close to the outer boundary. Under strong shear, this effect induces a layer buckling instability. The critical shear rate is proportional to $1/\sqrt{d}$, where $d$ is the thickness of the film.

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Consider shear flow experiments on freely standing fluid films in a Couette geometry (Fig. 1), in which the inner cylinder is rotating with speed $v_0$, and the outer cylinder is at rest. Both cylinders are perpendicular to the plane of the film. When a soap film is used in the experiment, the laminar flow between the cylinders is stable up to very large $v_0$, and the film gets thicker towards the outer cylinder due to centrifugal force [1]. Brightly colored patterns are exhibited because of this thickness variation. However, for smectic-*A* films with smectic layers parallel to the air-fluid interface, which were treated as two-dimensional systems in many analyses [2], there exists a critical speed $v_c$, such that, when $v_0$ is above $v_c$, defects are generated [3]. This clearly indicates that the layer structure becomes important, and the system can no longer be treated as two dimensional. On the other hand, experimental [4–7] and theoretical [8,9] studies on the rheological properties of bulk smectic-*A* systems show that, under shear flow, bulk systems are most stable when the layers are parallel to the shear plane. Thus the finite thickness and/or the free surfaces have to be essential to the observed defect generation in the experiments on freely standing smectic-*A* films.

In this Letter, we study the rheology of freely standing thermotropic smectic-*A* films under shear flow. In Couette geometry the centrifugal force pushes the liquid crystal away from the inner wall, which induces a layer dilation close to the outer wall. When this dilation exceeds a certain limit, the smectic layers are no longer stable against small undulational perturbations, hence, defects are generated. For typical materials and experimental setups, the strain induced by centrifugal force provides a critical velocity, $v_c$, comparable to the experimentally observed result [3]. Thus we conclude that this scenario is a good candidate for the mechanism which is responsible for the defect generation in Ref. [3]. Our analysis indicates that this instability is a result of the interplay between crossover and boundary effects (2D to 3D, and the existence of free surfaces), and the externally imposed flow. This instability is similar to the classical Helfrich-Hurault–type instabilities [10], but the film has free surfaces and is far from equilibrium. It is also similar but not identical to (Eckhaus) instabilities in convection roll patterns [11], in which both squeezing and dilating the system by controlling the boundaries leads to instabilities.

*Strain induced buckling instability for freely standing smectic-A films.*—We begin with the classical strain induced buckling instability for smectic-*A* in a new geometry, i.e., freely standing films. Let the smectic layers be parallel to the surfaces of the film, which are located at $z = \pm d/2$, and ignore the boundaries in the $x$ and $y$ directions. The total elastic free energy of the film is [12,13]

$$F = \alpha\left(\int dA_+ + \int dA_-\right) + \frac{1}{2}\int d^2r_\perp \int_{-d/2}^{d/2} dz\,[B(E[u])^2 + K_1(\partial_\perp^2 u)^2], \tag{1}$$

where $\int dA_\pm = \int d^2r_\perp \sqrt{1 + (\vec{\partial}_\perp [u]_{z=\pm d/2})^2}$ is the area of the surfaces, $u$ is the smectic layer displacement, $\alpha$ is the surface tension, $K_1$ and $B$ are layer undulation and compression elastic moduli, and $E[u] = \partial_z u - \frac{1}{2}(\vec{\partial}_\perp u)^2$ is the layer dilation. The continuity of the normal stress at the surfaces requires [14–16]

$$\pm[BE[u]]_{z=\pm d/2} = \alpha(\delta A_\pm/\delta u) + P_{\rm ext}^{\pm}, \tag{2}$$

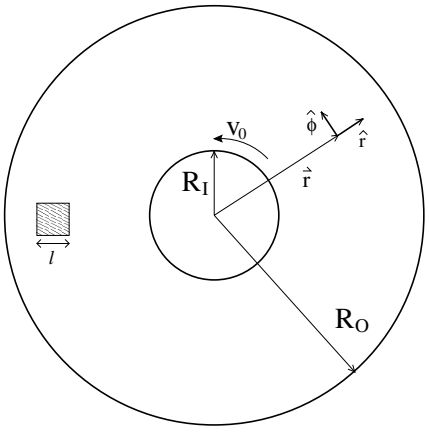


FIG. 1. Schematic of the experimental setup in Couette geometry; the $z$ axis points out of the paper.

  

where $\delta/\delta u$ stands for variational differentiation, and $P_{\rm ext}^{\pm}$ are the external normal stresses acting on the surface at $z = \pm d/2$.

Suppose the smectic layers are subject to a uniform but opposite external stress $P_{\rm ext}^{\pm} \equiv \pm P_{\rm ext}$ on the surfaces; for small $P_{\rm ext}$ the layers are uniformly dilated with $\partial_z u = P_{\rm ext}/B \equiv \epsilon$. We study the stability of small layer undulations of the form $u = u_0 + u_1$ with $u_0 = \epsilon z$ and $u_1 = UN(q_y)\sin(q_y y)\cos(q_z z)$, where, for given $q_y > 0$, $q_z > 0$ is chosen such that Eq. (2) is satisfied, and the normalization factor $N(q_y)$ is chosen such that $[N(q_y)]^2 \times \int_{-d/2}^{d/2} dz \cos^2(q_z z) = 1$.

Expanding $F$ to $O(U^2)$ and using the boundary condition for $u_1$ to the corresponding order, one finds

$$F \propto \frac{1}{4}[B(q_z^2 - \epsilon q_y^2) + K_1 q_y^4]U^2, \tag{3}$$

where, from Eq. (2), $q_z$ satisfies the equation

$$q_z d = (l_d q_y)^2 \cot\frac{q_z d}{2}, \tag{4}$$

in which $l_d = \sqrt{\alpha d/B}$ is a characteristic length. As discussed in Ref. [15], this length characterizes the crossover from behavior dominated by surface tension to behavior dominated by layer elasticity in freely standing smectic-$A$ films. Since we are interested in the onset of instability, we consider only the smallest $q_z$ which satisfies Eq. (4) throughout this Letter. From Eq. (3) it is clear that, when the induced strain $\epsilon$ is large enough, there is an instability, and one would expect growth of the amplitude $U$.

From Eq. (3), the critical strain for given $q_y$ is $\epsilon_c(q_y) = q_z^2/q_y^2 + \lambda^2 q_y^2$, where $\lambda = \sqrt{K_1/B}$ is a characteristic length for smectic $A$ [12]. Defining dimensionless parameters $X = q_z d, Q = q_y l_d$, then

$$\epsilon_c(Q) = g\frac{\lambda}{d}\left(\frac{X^2}{Q^2} + \frac{Q^2}{g^2}\right) \equiv g\frac{\lambda}{d}T(Q,g), \tag{5}$$

where $g = \alpha/\sqrt{K_1 B}$ is a dimensionless parameter; typically $g \sim O(1)$. The exact value of $X$ cannot be found analytically, but numerically one finds that $T(Q,g)$ has a minimum at $Q = Q^* \geq 0$. From the definition of $T(Q,g)$, $Q^*$ depends only on $g$. For given material parameters, one can find the minimum critical strain, $\epsilon^* = \epsilon_c(Q^*)$, numerically, which depends only on $g$ and $\lambda/d$. Figure 2 shows $Q^*$ and $6\epsilon^* d/\lambda g$ as functions of $g$. Notice that there is a $g_m$ such that, when $g < g_m$, the first unstable mode has infinite wavelength. We can estimate this parameter by expanding the equation for $X$ around $Q = 0$, and then substituting this approximate value of $X$ into Eq. (5). We find $X^2/Q^2 + Q^2/g^2 = 2 + (1/g^2 - 1/3)Q^2 + O(Q^4)$. For the first unstable mode to occur at $Q = 0$, the coefficient of the $Q^2$ term in the small $Q$ expansion of the critical strain has to be positive, yielding $g_m = \sqrt{3}$. On the other hand, when $1/g = \sqrt{K_1 B}/\alpha \to 0$, the surface tension is sufficiently large such that the instability essentially becomes the Helfrich-Hurault instability originally derived for hard boundaries, where the first unstable mode is $q_y^* = \sqrt{\pi/d\lambda}$ and the minimum critical strain is $2\pi\lambda/d$ [10].

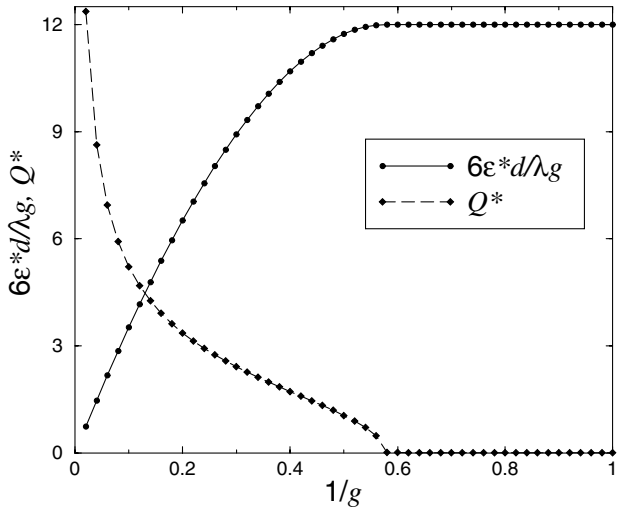


FIG. 2. The first unstable mode $Q^*$ and the critical strain $6\epsilon^* d/\lambda g$ for freely standing smectic-$A$ films under a uniform strain.

In the shear flow experiments conducted in Ref. [3], the freely standing smectic-$A$ films were made of 4-cyano-4′-octylbiphenyl (8CB), which has $\alpha \approx 25$ dyn/cm [17], $B \approx 5 \times 10^7$ dyn/cm$^2$ [18], $K_1 \approx 5 \times 10^{-7}$ dyn [19], i.e., $g \approx 5$. From Fig. 2 one finds that (in equilibrium), under a uniform layer dilation, the minimum critical strain $\epsilon^* \sim \lambda g/d = \alpha/Bd$, and the first unstable mode has wavelength on the order of $l_d$.

*Shear induced instability for freely standing smectic-A films in the Couette geometry.*—The experimental setup is shown in Fig. 1. We choose cylindrical polar coordinates to describe the system in which the $z$ axis is the axis of the cylinders. The radii of the inner and outer cylinders are $R_I$ and $R_O$, respectively. Let the film be suspended horizontally on the $r\phi$ plane between the inner and outer cylinders with thickness $d$ (from $z = -d/2$ to $z = d/2$) [20]. In principle, $d$ depends on position in the $r\phi$ plane. The smectic layers are parallel to the $r\phi$ plane away from the meniscus. The films are usually thicker near the meniscus due to wetting [21], and the liquid crystal near the meniscus acts similar to a reservoir of material in shear flow experiments [3]. Furthermore, the interactions between the liquid crystal and the walls also cause edge dislocations [22]. Since our focus is the effect of shear flow, to avoid additional complications, we assume both walls to be neutral to the liquid crystals, and neglect the effect of gravity and the surrounding air so that in equilibrium the smectic film is perfectly flat with a uniform thickness $d = d_0$. As a result, in our model there is no material reservoir near the meniscus, and the total volume of the film under shear flow is the same as in the film at rest, a simplification which does not occur in real experiments.

The equations of motion for this system in the isothermal, incompressible limit are [13]

$$\frac{\partial u}{\partial t} + \mathbf{v} \cdot \nabla u = v_z + \zeta_p h, \tag{6}$$

$$\rho\left(\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v}\right) = -\nabla p + h\hat{\mathbf{z}} + \eta \nabla^2 \mathbf{v}, \tag{7}$$

where $\mathbf{v}$ also satisfies the incompressibility condition $\nabla \cdot \mathbf{v} = 0$. Here, $\mathbf{v}$ is the velocity of the liquid crystal, $\zeta_p$ is the permeation constant, $p$ is the pressure, and $h = \vec{\partial} \cdot (\delta F/\delta \vec{\partial} u)$. In general, the viscosity $\eta$ should be a second rank tensor due to the anisotropy of the smectic-$A$ phase [12,13]. However, to simplify the expressions we choose $\eta$ to be a scalar. Because of the particular experimental geometry under consideration, the results of our analysis, i.e., the steady state configuration and the linear stability analysis, are not affected by this simplification. A similar calculation with full anisotropic viscosity yields the same results.

The stress should be continuous across the free surfaces, which leads to the following conditions [16]:

$$\left(\frac{\partial v_\phi}{\partial z} + \frac{1}{r}\frac{\partial v_z}{\partial \phi}\right)_{z=\pm d/2} = 0, \tag{8}$$

$$\left(\frac{\partial v_z}{\partial r} + \frac{\partial v_r}{\partial z}\right)_{z=\pm d/2} = 0, \tag{9}$$

$$\left(-p + BE[u] \mp \alpha \partial_\perp^2 u + 2\eta \frac{\partial v_z}{\partial z}\right)_{z=\pm d/2} = 0. \tag{10}$$

We take no-slip boundary conditions for the velocity on the inner and outer walls, i.e., $\mathbf{v} = v_0 \hat{\phi}$ at $r = R_I$, and $\mathbf{v} = \mathbf{0}$ at $r = R_O$. We first consider the steady state. Subsequently, we will study the linear stability of the steady state. We assume rotational invariance around the $z$ axis. As a result it is natural to set $v_r = 0$ everywhere. Then incompressibility and no-slip boundary conditions lead to $v_z = 0$. The momentum equation, Eq. (7), now becomes

$$\frac{v_\phi^2}{r} = \frac{1}{\rho}\frac{\partial p}{\partial r}, \tag{11}$$

$$0 = \nabla^2 v_\phi - v_\phi/r^2 = \frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial v_\phi}{\partial r}\right) + \frac{\partial^2 v_\phi}{\partial z^2} - \frac{v_\phi}{r^2}, \tag{12}$$

$$0 = -\frac{\partial p}{\partial z} + h. \tag{13}$$

The solution of Eq. (12) which satisfies the boundary conditions is $v_\phi = ar + \frac{b}{r}$, where $a = -R_I v_0/(R_O^2 - R_I^2)$, $b = R_I R_O^2 v_0/(R_O^2 - R_I^2)$.

With $v_z = 0$ and $v_\phi$ given above, the steady state layer displacement has to satisfy [from Eqs. (6) and (13)] $h = B\partial_z^2 u - K_1 \partial_\perp^4 u = 0$, and $\frac{\partial p}{\partial z} = 0$; i.e., the pressure depends only on $r$. Now one can solve Eq. (11) by direct integration, leading to

$$p(r) - p(R_I) = \rho\left(\frac{a^2}{2}(r^2 - R_I^2) + 2ab \ln\frac{r}{R_I} + \frac{b^2}{2}\left(\frac{1}{R_I^2} - \frac{1}{r^2}\right)\right), \tag{14}$$

where $p(R_I)$ will be determined later, by conservation of total volume. Since $p(r) - p(R_I)$ is positive definite for nonvanishing $v_\phi$, the pressure near the outer wall is larger than the pressure near the inner wall. The boundary condition for the normal stress, the condition $h = 0$, and Eq. (14) together with the conservation of total volume will completely determine the steady state configuration.

To understand the effect of this centrifugal force induced pressure, let us first consider the long wavelength behavior of the layer displacement to linear order in $u$. To this order the boundary condition for the normal stress is $B[\partial_z u]_{z=\pm d/2} = \pm\alpha[\partial_\perp^2 u]_{z=\pm d/2} + [p]_{z=\pm d/2}$. Simple dimensional analysis [replacing $\partial_z$ by $1/d$, $\partial_\perp$ by $1/(R_O - R_I)$, and typically $B/d \gg \alpha/(R_O - R_I)^2$ [20]] shows that, for nonvanishing $p$, the surface tension term is negligible compared to the layer compression for typical experiments, and the layer dilation on the surfaces is related to the pressure by

$$B[\partial_z u]_{z=\pm d/2} = [p]_{z=\pm d/2}. \tag{15}$$

The pressure "pushes" the smectic layers on the free surfaces, but with a very large characteristic in-plane length scale $(R_O - R_I)$. Since the walls are neutral to the liquid crystal, and $h = 0$ everywhere, the layer displacement is approximately $u = \frac{p(r)}{B} z = \epsilon z$. This layer displacement causes a change of the smectic film thickness from $d_0$ to $d_0[1 + p(r)/B]$. Since the total volume of the liquid crystal in the steady state is the same as that of the initial state, we determine the pressure at $r = R_I$ from the condition $\int_{R_I}^{R_O} dr\,(2\pi r d_0) = \int_{R_I}^{R_O} dr\,(2\pi r d_0[\frac{p(r)}{B} + 1])$. One finds the layers are compressed for small $r$ but are dilated for large $r$ due to the effect of centrifugal force and conservation of total volume.

Consider a small patch located near the outer cylinder having size $l \ll R_O - R_I$. The stability to a layer undulation with wave vector in the radial direction can be analyzed as follows. Choosing $\hat{\boldsymbol{x}} = -\hat{\phi}$, $\hat{\boldsymbol{y}} = \hat{\boldsymbol{r}}$, when $r$ is close to $R_O$ with $R_O - r \gg l$, we can work in Cartesian coordinates and ignore the effect of the walls. The small layer undulation can be expressed as $UN(q_y)\sin(q_y y) \times \cos(q_z z)$. Since the shear flow is in the $x$ direction, the small layer undulation with in-plane wave vector in the $y$ is not convected by the flow [i.e., in Eq. (6), $\vec{v} \cdot \vec{\nabla} u$ vanishes], and it evolves towards $h = \vec{\nabla} \cdot \delta F/\delta \vec{\nabla} u = 0$. Thus the equilibrium analysis can be used to show that, for large enough $v_0$, the induced layer dilation close to the outer cylinder can cause a layer undulation instability. The fact that the first unstable mode for 8CB has a wavelength on the order of $l_d$ justifies our use of $R_O - R_I \gg l$ [20].

We are now in a position to estimate $v_c$ for the experimental geometry used Ref. [3], i.e., $R_I = 1.2$ cm, $R_O = 1.5$ cm. Choosing $r \sim R_O$, and estimating $\epsilon^* \sim \alpha/Bd$ to estimate the layer dilation close to the outer cylinder and the critical strain, one finds, for a typical material, $\rho \approx 1$ g/cm$^3$, $\alpha \approx 25$ ergs/cm$^2$, $v_c^2 d \sim 1500$ cm$^3$/s$^2$. For $d = 3.5 \times 10^{-4}$ cm, one finds $v_c \approx 2.1 \times 10^3$ cm/s, and the critical shear rate is $\dot{\gamma}_c = v_c/(R_O - R_I) \sim 7 \times 10^3$ s$^{-1}$. The experimental critical shear rate is about $10^3$ s$^{-1}$. Considering the simplifications made in this model, the agreement indeed indicates that the effect of layer dilation induced by centrifugal force can cause the instability observed in the experiments. Notice that our model shows that the initial instability should occur close to the outer boundary of the Couette cell, and this is consistent with the experimental observation [23].

*Concluding remarks.*—We have shown that freely standing smectic-*A* films are in principle unstable against strong shear flow. The characteristic length $l_d$ introduced in Ref. [15] plays an important role. We considered specifically the Couette geometry and included the effects of centrifugal force. We showed that centrifugal force is capable of inducing a layer dilation close to the outer cylinder. When the shear rate is large enough, defects are generated due to a layer buckling instability similar to Helfrich-Hurault–type instability [10], and the calculated critical shear rate is on the same order as the experimental measurements.

However, notice that the linear stability analysis in this Letter is appropriate only for layer undulations with in-plane wavelength small compared to $R_O - R_I$. As we point out, when $g = \alpha/\sqrt{K_1 B} < \sqrt{3}$, the first unstable mode against a uniform layer dilation has very long wavelength. Hence, our analysis does not apply to certain materials with weak surface tension. For the material (8CB) used in Ref. [3], $g \approx 5$, and our analysis is appropriate.

Our calculations have assumed perfectly aligned smectic layers and ignored any interactions between the air or solid boundaries with the liquid crystal. Edge dislocations and the material reservoir commonly observed near the meniscus [22], which may not be negligible in the experiment, are also ignored in our approach. Because of these simplifications, our work should be compared only semiquantitatively with the experiments. We also note that, under strong shear flow [24], defects are generated in freely standing lyotropic smectic-*A* films in Couette geometry. Since the density of the solvent is an extra hydrodynamic variable in lyotropic systems [25,26], whether the mechanism for defect generation is the same as that for the thermotropic systems remains to be answered. However, our study has provided the basic ingredients of the experimental instability, i.e., the finite film thickness, surface tension, and the geometry of the experimental setup. It also provides the physical picture of the instability, i.e., the differences between a smectic-*A* film and a soap film in Couette geometry, and the differences between a bulk system and a freely standing film. Hence, we believe that we have achieved our goal, which was to elucidate the basic mechanism behind the interesting experiments reported in Ref. [3]. We also add to the growing catalog of fascinating properties of smectic-*A* films far from equilibrium.

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