Layer dynamics of a freely standing smectic-A film

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Abstract

We study the hydrodynamics of a freely-standing smectic-A film in the isothermal, incompressible limit theoretically by analyzing the linearized hydrodynamic equations of motion with proper boundary conditions. The dynamic properties for the system can be obtained from the response functions for the free surfaces. Permeation is included and its importance near the free surfaces is discussed. The hydrodynamic mode structure for the dynamics of the system is compared with that of bulk systems. We show that to describe the dynamic correlation functions for the system, in general, it is necessary to consider the smectic layer displacement \(u\) and the velocity normal to the layers, \(v_z\), together. Finally, our analysis also provides a basis for the theoretical study of the off-equilibrium dynamics of freely-standing smectic-A films.

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I. INTRODUCTION

A three dimensional smectic-A phase is at its lower critical dimension: the smectic layer displacement fluctuations grow logarithmically with the size of the system, and the divergence is sufficiently weak that finite size effects stabilize laboratory samples [1].

Smectic liquid crystals also form freely standing films with a controlled size and high degree of uniformity. Due to the presence of two free surfaces and finite thickness, experimental and theoretical studies on the equilibrium [2–5], and off-equilibrium [6] properties of freely standing smectic-A films show drastically different properties from bulk systems. Hence freely standing smectic-A films are excellent materials to study the effect of reduced dimensionality and surface effects.

In addition to the static properties, the dynamics of a freely standing smectic-A film have been studied in the “low” frequency ($\omega \ll 50$kHz) and “long” wavelength ($\lambda \gg 0.1$ cm) regime, where it behaves like a simple elastic membrane [7]. The dynamic correlation functions have also been considered in the strongly overdamped regime, [5] both theoretically and experimentally. Notice, however, that these experiments were performed in a region where the effects of bulk elasticity are negligible; further experiments are needed to probe the dynamical crossover behavior of the system.

In this article we introduce a continuum theory for the dynamics of a smectic-A film in the isothermal, incompressible limit based on the linear hydrodynamic theory constructed by Martin et al [8]. Our study provides a finite-thickness counterpart to the hydrodynamic theory for bulk smectic-A systems. The following two questions are addressed and answered in our theory: (i) Permeation plays a role near the free surfaces [9], but how does it modify the smectic layer dynamics near the boundaries? (ii) The smectic layer displacement $u$, and the velocity of the liquid crystal normal to the layers, $v_z$, are coupled hydrodynamic variables, but how do the hydrodynamic modes for $u$ and $v_z$ for a film compare to the linear theory for a bulk smectic-A [1,8]? In the following sections we will show that the dynamic properties of the smectic layers in a freely standing film can be extracted from the response
functions of the free surfaces. Since the layer displacement for a smectic-A film has to satisfy certain boundary conditions on the free surfaces, the normal modes for the dynamics of the layers depend on both surface and bulk properties. We find that among all the normal modes, two are most strongly affected by the surface tension and film thickness. The decay rates for all other normal modes show the interplay of bulk-like behavior and effects from the surfaces. When the thickness of the film is small, the dominant thermal fluctuations of the layers come from these two modes, and the system can behave similarly to an ordinary fluid film. When the thickness becomes large, the other normal modes have sufficiently large contributions to the layer fluctuations, and the internal layer structure of the system can be detected experimentally. The present analysis reveals how the dynamics of a finite thickness smectic-A system depends on surface tension, viscosity, and also bulk elasticity. We also provide a theoretical picture for the crossover behavior of a freely standing smectic-A film from a thin film, which depends only on the surface properties, to a three-dimensional system with layer structure. Furthermore, this theory is also a starting point for theoretical calculations for the off-equilibrium dynamics of a freely standing smectic-A film.

The remainder of this paper is organized as follows. Section II contains a review of the linear elastic and hydrodynamic theory for a bulk smectic-A in the isothermal, incompressible limit. After a brief review of the linear response functions of the surfaces of a smectic-A film, we extract the smectic layer dynamics and compare the bulk and film hydrodynamics in Section III. The crossover of smectic dynamics from film-like behavior to bulk-like behavior is discussed in Section IV. In Section V we summarize results and add concluding remarks. Some additional details on permeation near the surfaces and the calculation of the dynamic correlation functions are provided in Appendices A and B, respectively.

II. ELASTICITY AND HYDRODYNAMICS OF BULK SMECTIC-A PHASE

The symmetry broken direction for a smectic-A system is chosen to be the $z$-direction. Assume that temperature and density fluctuations occur on time scales much faster than the
characteristic times under consideration. In this limit we have an isothermal, incompressible system, and the elastic free energy of the smectic layer distortion for a bulk smectic-A in thermal equilibrium is provided to leading order by

$$ F = \frac{1}{2} \int dV \left\{ B \left( \frac{\partial u}{\partial z} \right)^2 + K_1 \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right)^2 \right\} $$

where $u(r,t)$ is the layer displacement, $r = (x,y,z)$, and $B$ and $K_1$ are, respectively, the layer compression and undulation elastic moduli. The volume integral is performed over the bulk. In this article we consider exclusively a thermotropic smectic-A, for which typically $B \sim 2.5 \times 10^7$ dyn/cm$^2$, $K_1 \sim 10^{-6}$ dyn. The characteristic length $\lambda \equiv \sqrt{K_1/B}$ is typically on the order of the layer spacing ($\sim 10^{-7}$cm) [1]. It has the following physical interpretation: in bulk systems the dominant thermal fluctuations of the layers with momentum $q$ are those with $q_z \sim \lambda q_{\perp}^2$, where $q_{\perp}$ is the magnitude of the projection of $q$ on the $xy$-plane. When $q_{\perp} \ll \lambda^{-1}$, the dominant layer fluctuations come from $q_z \ll q_{\perp}$.

The dynamics of a bulk smectic-A have been studied thoroughly in, for example, Ref. [8], which we follow. In the absence of topological defects, under constant temperature and assuming incompressibility, to linear order the system satisfies the equations of motion

$$ \rho \frac{\partial v_i}{\partial t} = -\partial_i p + \partial_j \sigma'_{ij} + h \, \delta_{iz} $$

and

$$ \frac{\partial u}{\partial t} = v_z + \zeta_p \, h \, , $n

where $\rho$ is the density (typically $\rho \sim 1$ g/cm$^3$), $v_i$ is the $i$th component of the velocity field, and the pressure, $p$, is actually a Lagrange multiplier for the incompressibility condition. The viscous stress tensor is denoted by $\sigma'$ [8], $\zeta_p$ is the permeation constant, and the molecular field $h$ is defined by

$$ h \equiv \partial_i \left( \frac{\delta F}{\delta \partial_i u} \right) \, . $$

In Eqs. (2) and (3) we sum on repeated indices, and $\partial_j = \partial/\partial x_j$. For layer displacements of the form
\[ u(\mathbf{r}, t) = u(\mathbf{q}, \omega) e^{i(q_{\perp} \cdot \mathbf{r} + q_z z) - i\omega t} \]  

(5)

with \( q_z \ll q_{\perp} \), permeation may be neglected \([\text{II}]\), and the relation between \( \omega \) and \( \mathbf{q} \) is given by

\[
i\omega = \frac{\eta_3 q^2_{\perp}}{2 \rho} \times \left( 1 \pm \sqrt{1 - \frac{4 \rho}{\eta_3^2 q^4_{\perp}} (Bq^2_z + K_1 q^4_{\perp})} \right) \equiv i\omega_{\pm}(q_{\perp}, q_z),
\]

(6)

where \( \eta_3 \) (typically \( \eta_3 \sim 1p \)) is one of the five viscosity coefficients of a smectic-A \([\text{III}]\). The fact that \( u(\mathbf{r}, t) \) couples to \( v_z(\mathbf{r}, t) \) in the equations of motion means that, in general, a hydrodynamic normal mode described by Eq. (6) is a linear combination of these two variables. There are two other normal modes for a given \( \mathbf{q} \), but they correspond to the diffusive modes of \( v_x \) and \( v_y \), and they are decoupled from the motion of the smectic layers \([\text{IV}]\). We will not discuss these two other modes in the rest of this section. In the limit \( 4\rho(Bq^2_z + K_1 q^4_{\perp})/\eta_3^2 q^4_{\perp} \ll 1 \), the viscous loss of the system is large, and the motion of the layers is strongly overdamped. Then \( i\omega \) is real, and the two decay rates are given by

\[
\frac{1}{\tau_1} = \frac{\eta_3 q^2_{\perp}}{\rho}, \quad \frac{1}{\tau_2} = \frac{Bq^2_z + K_1 q^4_{\perp}}{\eta_3 q^2_{\perp}},
\]

(7)

where \( 1/\tau_1 \gg 1/\tau_2 \). We can identify \( 1/\tau_1 \) as the decay rate of the diffusive mode associated with \( v_z \) and \( 1/\tau_2 \) as the decay rate for the smectic layer undulations. Since we are interested in the low frequency behavior of the system, it is appropriate to neglect the effect of \( \tau_1 \). In the opposite limit, \( 4\rho(Bq^2_z + K_1 q^4_{\perp})/\eta_3^2 q^4_{\perp} \gg 1 \), the elastic restoring force is much stronger than the viscous loss, and the system performs underdamped oscillations, i.e., \( i\omega \) becomes complex. The decay rate is simply \( \eta_3 q^2_{\perp}/2\rho \), and the oscillation frequency is approximately \( \sqrt{(Bq^2_z + K_1 q^4_{\perp})/\rho} \), which is the frequency of “second sound” \([\text{I}]\). In this case, then, there are two propagating hydrodynamic modes associated with each \( \mathbf{q} \). Notice that when \( q_z \sim \lambda q^2_{\perp} \), the system is strongly damped as long as \( \rho K_1/\eta_3^2 \ll 1 \); for typical materials \( \rho K_1/\eta_3^2 \sim 10^{-6} \), so that the dominant thermal fluctuations of the layers are overdamped.
III. HYDRODYNAMIC NORMAL MODES OF A FREELY STANDING SMECTIC-A FILM

Let us now turn to the case of freely standing smectic-A films. In the absence of external fields, the smectic layers are always aligned and parallel to both free surfaces. We consider a film which extends from \( z = d/2 \) to \( z = -d/2 \) in the vertical direction and is of infinite extent in the \( x \) and \( y \)-directions. The geometry of the system is shown in Fig. 1, which shows \( \zeta^+(-) \) as the displacement of the upper (lower) surface from its equilibrium position. In this section we first briefly review previous results [11] on the derivation of the linear response function for the equilibrium surface fluctuations of a freely standing smectic-A film in the low frequency, long wavelength regime [12,13], then extend these results to the structure of hydrodynamic normal modes of the system.

Suppose the system were perturbed by external forces with fixed in-plane wave vector \( \mathbf{q}_\perp \) and frequency \( \omega \) on both upper and lower surfaces, i.e,

\[
P_{\text{ext}}^+(\mathbf{r}_\perp, t) = P_{\text{ext}}^+(\mathbf{q}_\perp, \omega) \, e^{i \mathbf{q}_\perp \cdot \mathbf{r}_\perp - i \omega t}
\]

\[
P_{\text{ext}}^-(\mathbf{r}_\perp, t) = P_{\text{ext}}^-(\mathbf{q}_\perp, \omega) \, e^{i \mathbf{q}_\perp \cdot \mathbf{r}_\perp - i \omega t}.
\]

(8)

We look for the linear response functions of the surfaces in the regime of weak external forces. On the free surfaces the system has to satisfy the following boundary conditions:

**bc 1** The velocities of the free surfaces are the same as the normal component of the liquid crystal velocity on the surfaces.

**bc 2** For free surfaces the normal component of the permeation force should vanish. [11]

**bc 3** The force acting on the system is continuous across the free surfaces.

The boundary conditions lead to the following equations for the system,

\[
\frac{\partial \zeta^+(-)}{\partial t} = [v_z]_{z=\pm d/2}.
\]

(9)
\[
\begin{bmatrix}
\frac{\partial u}{\partial z}
\end{bmatrix}
_{z=\pm d/2} = 0 . 
\] (10)

\[
\left[ \partial_i v_z + \partial_z v_i \right]_{z=\pm d/2} = 0 , \quad i = x, y ,
\] (11)

\[
\left[ \pm \left( -p + 2\eta_3 \frac{\partial v_z}{\partial z} + B \frac{\partial u}{\partial z} \right) - \alpha \nabla_\perp^2 \zeta^\pm - P_{\text{ext}}^\pm \right]_{z=\pm d/2} = 0 ,
\] (12)

where \( \alpha \) is the surface tension (typically \( \alpha \sim 30 \text{ dyn/cm} \)).

Solving the equations of motion, i.e., Eqs. (2), and (3), with these boundary conditions, one finds that, in the presence of the driving forces with given \( q_\perp \) and \( \omega \), there are four different \( q_z \)'s. One of them is associated with the diffusive motion for \( v_x \) and \( v_y \), and is decoupled from the motion of \( u \) and \( v_z \); hence it is irrelevant for the calculation of surface response functions (see bc 1). Another two values of \( q_z \) are associated with permeation, and these contributions to the layer displacement are small (see Appendix A). The last of these \( q_z \)-values dominates the contribution to the smectic layer displacement; hence \( u \) can be approximated by

\[
\begin{align*}
u(r,t) = u_S(q_\perp,\omega) \cos [q_z(q_\perp,\omega)z] \times e^{i q_\perp \cdot r_\perp - i \omega t} + u_A(q_\perp,\omega) \sin [q_z(q_\perp,\omega)z] \times e^{i q_\perp \cdot r_\perp - i \omega t} . 
\end{align*}
\] (13)

Using \( \omega_\pm(q_\perp,q_z) \) as defined in Eq.(3), \( q_z(q_\perp,\omega) \) is determined by the solution of the following with \( \text{Re} q_z > 0 \):

\[
i \omega = i \omega_\pm(q_\perp,q_z),
\] (14)

where \( |q_z| \ll q_\perp \), and, in general, \( q_z \) is complex under the conditions considered [12]. Recall that \( \omega \) and \( q_\perp \) describe the driving forces and \( q_z \) is not independent. This differs from the interpretation of the previous section in which the bulk hydrodynamics of a smectic-A was considered in the absence of external driving forces.

When \( P_{\text{ext}}^+ = P_{\text{ext}}^- \) (\( P_{\text{ext}}^+ = -P_{\text{ext}}^- \)), the dynamics of the system are symmetric (antisymmetric) under \( z \to -z \), and the layer displacement is described by \( u_S \) (\( u_A \)) alone. We define the response functions for both symmetric and anti-symmetric surface motion as
ζ^S(q_\perp, w) = X^S(q_\perp, w) \left[ P^+_{ext}(q_\perp, w) + P^-_{ext}(q_\perp, w) \right],
ζ^A(q_\perp, w) = X^A(q_\perp, w) \left[ P^+_{ext}(q_\perp, w) - P^-_{ext}(q_\perp, w) \right],
\tag{15}

where
\zeta^{S(A)} = \frac{1}{2} \left( \zeta^+ \pm \zeta^- \right). \tag{16}

A calculation from the continuum hydrodynamics leads to the following expressions for the response functions \cite{11},
\begin{align*}
X^S &= \frac{1}{2} \frac{1}{\alpha q_\perp^2 - B q_z(q_\perp, \omega) \tan(d q_z(q_\perp, \omega)/2)},
\tag{17}
X^A &= \frac{1}{2} \frac{1}{\alpha q_\perp^2 + B q_z(q_\perp, \omega) \cot(d q_z(q_\perp, \omega)/2)}. \tag{18}
\end{align*}

It appears that the permeation processes have no contribution to dynamics of the surfaces in the regime \cite{12} where our analysis is done. However, the solution for u(r, t) in Eq. (13) does not strictly satisfy bc 2. This means that the contribution from permeation enables the system to satisfy the boundary conditions \cite{9} but otherwise has little significance. The role of permeation in the dynamics of the system is discussed in further detail in Appendix A.

We have derived the response functions X^{S(A)} as functions of in-plane wave vector q_\perp and frequency \omega of the applied external forces. We can also define \omega on the complex plane; then the poles of the response functions provide the frequencies of the surface fluctuations in the absence of driving forces \cite{10}. In linear theory these poles also reveal the frequencies of layer displacement. Each pole in the complex \omega-plane corresponds to a hydrodynamic normal mode associated with the layer displacement. Since we are not interested in the dynamics of v_x, and v_y, which decouple from the dynamics of u and v_z, these poles provide the information of interest. Two classes of normal modes with opposite symmetry properties can be found. One class of normal modes is symmetric under z \rightarrow -z; the frequencies are the roots of,
\begin{equation}
\frac{B q_z(q_\perp, \omega)}{\alpha q_\perp^2} = \cot \left( \frac{d q_z(q_\perp, \omega)/2}{2} \right). \tag{19}
\end{equation}
The other class of normal modes is anti-symmetric under $z \rightarrow -z$, and their frequencies are determined by
\[
\frac{Bq_z(q_{\perp},\omega)}{\alpha q_{\perp}^2} = -\tan\left(\frac{q_z(q_{\perp},\omega)d}{2}\right).
\] (20)

Actually, we can go back and treat the above two equations as equations for $q_z$; we can find the $z$ dependence of the layer displacement for the normal modes from these equations. The form of the layer displacement in Eq. (13) suggests that, for given $q_{\perp}$, the layer displacement for an normal mode is an eigenfunction of the differential equation
\[
\frac{\partial^2 \psi(q_{\perp},z)}{\partial z^2} = -q_z^2 \psi(q_{\perp},z)
\] (21)

with boundary condition
\[
bc2' \quad \left[ B \frac{\partial \psi(q_{\perp},z)}{\partial z} \pm \alpha q_{\perp}^2 \psi(q_{\perp},z) \right]_{z=\pm d/2} = 0,
\] (22)

It is straightforward to show that these eigenfunctions form a complete basis for the interval $-d/2 < z < d/2$. The apparent contradiction between $bc\ 2'$ and $bc\ 2$ is discussed in Appendix A.

In general $q_z$ in Eqs. (19) and (20) cannot be determined analytically. However, the $q_z$’s are all real, and we can choose all of them to be positive. The graphical solutions for the $q_z$’s are shown in Fig. 2. We label the $q_z$’s for the normal modes as $q_z^{(n)}(q_{\perp})$, $n = 0, 1, 2, \ldots$, with $q_z^{(0)}(q_{\perp}) < q_z^{(1)}(q_{\perp}) < q_z^{(2)}(q_{\perp}) < \ldots$, and note there are two normal modes associated with each $q_z^{(n)}$. The layer displacement for normal modes is even (odd) under $z \rightarrow -z$ if $n$ is even (odd). The layer displacement for the normal modes with $n = 0$ is even under $z \rightarrow -z$. When both modes are underdamped, they form a pair of propagating waves, which in bulk systems corresponds to second sound \[1\]. When they are overdamped, they are two diffusive modes. In the strongly overdamped limit, the characteristic time scales are well separated; the fast mode corresponds to the diffusive motion of $v_z$, and the slow mode corresponds to the layer undulation mode in bulk systems.

To compare with the behavior of a bulk smectic-A, recall that translational invariance of the bulk system allows one to label modes by a $d$-dimensional wave vector. For a freely
standing smectic-A film, the boundary conditions and the equations of motion select a set of normal modes for the dynamics of the system. We can express the normalized time-independent part of the normal modes in the following way,

$$\psi(q_{\perp}, n; z) = N_S(q_{\perp}, n) \cos \left[ q_z^{(n)}(q_{\perp}) z \right],$$

(23)

for \( n = 0, 2, 4, \ldots \), and

$$\psi(q_{\perp}, n; z) = N_A(q_{\perp}, n) \sin \left[ q_z^{(n)}(q_{\perp}) z \right],$$

(24)

for \( n = 1, 3, 5, \ldots \), where

$$N_S(q_{\perp}, n) = \sqrt{\frac{2}{d}} \left( 1 + \frac{\sin(q_z^{(n)} d)}{q_z^{(n)} d} \right)^{-1/2},$$

$$N_A(q_{\perp}, n) = \sqrt{\frac{2}{d}} \left( 1 - \frac{\sin(q_z^{(n)} d)}{q_z^{(n)} d} \right)^{-1/2}.\quad (25)$$

These normal modes satisfy the orthonormality condition

$$\int_{-d/2}^{d/2} dz \, \psi(q_{\perp}, n; z) \psi(q_{\perp}, m; z) = \delta_{nm}.\quad (26)$$

Hence, these modes can be used as a basis to expand any \( u(q_{\perp}, z, t) \) as,

$$u(q_{\perp}, z, t) = \sum_n u(q_{\perp}, n, t) \times \psi(q_{\perp}, n; z).\quad (27)$$

From these relations one finds the dynamic correlation function for \( u \). As discussed in Appendix B, the general form for the dynamic correlation function is

$$c(q_{\perp}, n, t)$$

$$\equiv < u(q_{\perp}, n, t) \ u(-q_{\perp}, m, 0) >$$

$$= \delta_{nm} \frac{i \omega_+ e^{-i \omega_- t} - i \omega_- e^{-i \omega_+ t}}{i \omega_+ - i \omega_-} \times \frac{k_B T}{B q_z^{(n)} + K_1 q_{\perp}^4},$$

(28)

where \( \omega_\pm = \omega_\pm(q_{\perp}, q_z^{(n)}) \) is related to \( q_{\perp} \) and \( q_z^{(n)} \) through Eq. (6). In the underdamped case, \( \omega_\pm \) are complex and \( \omega_- = \omega_+^* \). The dynamic correlation function has a damping rate \( \eta_3 q_{\perp}^2/2 \rho \) and oscillates with frequency \( \sqrt{-\eta_3 q_{\perp}^2/2 \rho} + (B q_z^{(n)} + K_1 q_{\perp}^4)/\rho \). In the overdamped case,
two decay rates exist, and the fast mode can be neglected only when the time scales are well separated. In general, when an experiment is performed outside the strongly overdamped regime, two characteristic time scales should be detected. For weakly overdamped motion, these are the two decay rates, while for underdamped motion, these are the real and imaginary parts of $\omega_\pm$.

In summary, we find that in common with analysis of a liquid film, the normal modes are symmetric/anti-symmetric under $z \to -z$. They also are labeled by $(q_\perp, n)$, and the layer displacement can be treated as eigenfunctions of a differential equation. The frequencies for those normal modes can be obtained from the poles of the *surface response functions* in the complex $\omega$-plane. As will become clear in the next section, the difference between those $q_z^{(n)}$'s which correspond to the normal modes for a finite thickness smectic-A film, and the set of numbers $n\pi/d$, $n = 1, 2, 3, ...$ provides a measure of the degree to which finite thickness and surface tension change the dynamic properties of the layer displacement. Experimentally, these effects are typically obtained by measuring the frequencies and the magnitude of the autocorrelation function of the layer displacement. Layer dynamics will be discussed further in the following section.

**IV. CROSSOVER OF LAYER DYNAMICS FOR SMECTIC-A FILMS**

There are two special regimes where the $q_z^{(n)}$’s can be evaluated approximately, and the physics of the system is also of interest. We call these the *thin film* and *thick film* regimes, respectively. We will show that the pair of normal modes with $q_z^{(0)}$ is special since they strongly depend on the surface properties and show drastically different behavior from the thin film to thick film regime.

When the dimensionless measure of film thickness $\delta \equiv \alpha q_\perp^2 d/B \ll 1$, the solution for $q_z$ can be approximated by

$$q_z^{(0)} = \frac{\sqrt{2\delta}}{d} = \sqrt{\frac{2\alpha}{Bd}} q_\perp$$

and

$$q_z^{(n)} = \frac{\sqrt{2\delta}}{d} = \sqrt{\frac{2\alpha}{Bd}} q_\perp$$
\[ q_z^{(n)} = \frac{n\pi}{d} \left( 1 + \frac{2\delta}{(n\pi)^2} \right) = \left( \frac{n\pi}{d} + \frac{2\alpha q_z^2}{n\pi B} \right), \quad n = 1, 2, 3, \ldots \] (30)

For \( n > 0 \) the modes are spaced approximately by \( \pi/d \). This is the thin film regime (\( \delta \ll 1 \)).

The equilibrium thermal fluctuations of the smectic layers in this limit are dominated by surface effects. This can be illustrated by considering the pair correlation function for the layer displacement. From Appendix A, in thermal equilibrium, for a smectic-A film with given \( q_\perp \), the contribution to the pair correlation function from the modes with \( q_z^{(0)} \) is

\[ \langle |u(q_\perp, 0)|^2 \rangle = \frac{k_BT}{2\alpha q_\perp^2/d + K_1 q_\perp^4}. \] (31)

Furthermore, when \( K_1 q_\perp^2 d/\alpha \ll 1 \), the contribution from layer elasticity is negligible, and the thermal fluctuations of the smectic layers for these normal modes are independent of the bulk elastic coefficients \( [5] \). For a typical material, \( B \sim 2.5 \times 10^7 \text{ dyn/cm}^2 \), \( K_1 \sim 10^{-6} \text{ dyn} \), \( \alpha \sim 30 \text{ dyn/cm} \), and the thin film regime corresponds to \( q_\perp^2 d \ll B/\alpha \sim 10^6 \text{ cm}^{-1} \). The layer fluctuation is dominated by the surface tension term when \( q_\perp^2 d \ll \alpha/K_1 \sim 3 \times 10^7 \text{ cm}^{-1} \), i.e., in the thin film regime, layer fluctuations due to these two normal modes are typically dominated by the surface tension term. In the following we will simply drop the contribution from the \( K_1 \) elasticity term in the thin film regime.

The total contribution from the fluctuations of the other normal modes for fixed \( q_\perp \) is approximately

\[ \sum_{n=1}^{\infty} \frac{k_BT}{B (n\pi/d + 2\alpha q_z^2/n\pi B)^2} \sim \frac{k_BT d^2}{B}. \] (32)

This contribution is small compared to the contribution from the normal modes with \( q_z^{(0)} \) in thin film limit. As a result, we find that a freely standing smectic-A film can be treated as an ordinary fluid film with surface tension \( \alpha \) and thickness \( d \) in this limit. \( [5] \) The internal structure of the layers is not important. Comparing with bulk smectic-A systems where the dominant layer fluctuations for given \( q_\perp \) come from \( q_z \sim \lambda q_\perp^2 \), for a thin film the dominant layer fluctuations come from only two normal modes, and it is in general not true that \( q_z^{(0)} \sim \lambda q_\perp^2 \).
From Eq. (33) the normal mode frequencies $\omega_{\pm}(q_{\perp}, q_{z}^{(0)})$ are given by

$$
    i\omega_{\pm}(q_{\perp}, q_{z}^{(0)}) = \frac{\eta_{3}q_{\perp}^2}{2\rho} \times \left( 1 \pm \sqrt{1 - \left( \frac{q_{c}}{q_{\perp}} \right)^2} \right),
$$

where $q_{c} = \sqrt{8\rho\alpha/\eta_{3}d}$ is a characteristic wave number of the film. In the case of short wavelength fluctuations with $q_{\perp} \gg q_{c}$, viscous loss is strong compared to elastic effects, and the overdamped decay rates are

$$
    i\omega_{+}^{(0)}(q_{\perp}) = \frac{\eta_{3}q_{\perp}^2}{\rho},
$$
$$
    i\omega_{-}^{(0)}(q_{\perp}) = \frac{2\alpha}{\eta_{3}d}.
$$

Since we are interested in slow motion of the system, $i\omega_{-}^{(0)}(q_{\perp})$ provides the characteristic time for the system. Previous experimental studies of dynamic correlation functions were performed in this regime [4]. Notice that in this regime the decay rate is independent of the magnitude of the in-plane wave vector $q_{\perp}$ and bulk elastic constants. In general, experiments which probe the fluctuations of the film with smaller in-plane wave vector should be able to identify two decay rates. This is especially so when the motion of the system is close to being critically damped, as can be seen from Fig. 3. For long wavelength $q_{\perp} \ll q_{c}$ the film oscillates with frequency $\sqrt{2\alpha/\rho d} q_{\perp}$ and damping rate $\eta_{3}q_{\perp}^2/2\rho$. Again, both time scales in this range of $q_{\perp}$ are independent of $B$ and $K_{1}$. As $q_{\perp} \to 0$, the damping is small; this is the regime in which the long wavelength theory and experiments [7] have been performed.

We have shown that the physical properties for the system in the thin film regime are basically the same as a two dimensional object, i.e., a simple fluid film. The dynamics of the system show a competition between the elasticity due to surface tension and viscous loss characterized by the coefficient $\eta_{3}$. The underdamped motion for a smectic-A film should not be confused with “second sound” for smectic layers, because for the thin film the layer compression is negligible. In this regime, the underdamped motion is simply the vibration of a simple fluid film with surface tension $\alpha$. Also notice that although the magnitude of $q_{z}^{(0)}$ depends on the elastic constant $B$, this dependence does not show up in the dynamical
correlation function for the hydrodynamic normal modes, as can be seen in Eq. (31), which
is the same as that for the surface fluctuation.

To probe the crossover behavior of the film, experiments have to be performed outside the
thin film regime so that the effect of the bulk elasticity can be revealed. This can be done by
increasing the film thickness or adjusting the in-plane wave vector. When $\alpha q^2 \theta d/B$ increases
to order unity, the layer structure of the system starts to play a non-negligible role in the
system. First of all, this is reflected in the fact that the approximation for $q_{z}^{(0)}$ in Eq. (29)
is poor, and the higher order terms contribute. Then the $n = 0$ modes start to show the
influence of bulk elasticity. Second, the layer fluctuations of all other normal modes become
comparable to the layer fluctuations for the $n = 0$ modes. Hence, the dynamics of the
layers for fixed $q_{\perp}$ are no longer simply controlled by $\omega_+(q_{\perp}, q_{z}^{(0)})$ and $\omega_-(q_{\perp}, q_{z}^{(0)})$,
and this means that there are more relevant degrees of freedom in the system than the displacement
of the two surfaces. The first sign of this crossover behavior can be detected by exciting
the $n = 1$ modes. As pointed out in the study of the surface dynamics of freely standing
smectic-A films [11,14], this pair of modes is similar to the so-called *peristaltic* mode for a
soap film. However, the characteristic time scales of these modes strongly depend on the
elasticity of the smectic layers; hence, the dynamic properties of these modes actually reveal
the difference between a smectic film and a soap film. Possible future surface light scattering
experiments for probing these normal modes are also discussed in Ref. [11].

For $\delta \gg 1$, $q_{z}^{(0)}$ increases and $q_{z}^{(0)}d \sim \pi$; the $q_{z}$’s for all normal modes can be approximated
by a single expression,

$$q_{z}^{(n)} = \frac{(n + 1)\pi}{d} \left(1 - \frac{2}{\delta}\right) = \frac{(n + 1)\pi}{d} \left(1 - \frac{2B}{\alpha q_{\perp}^2 d}\right), \quad n = 0, 1, 2, 3, ... \quad (35)$$

From Eqs. (23) and (24) we find that the layer displacement for the normal modes on both
free surfaces becomes vanishingly small as $\delta^{-1} \to 0$. This means that the layer displacement
is not affected by the surface tension in this limit, and the behavior of the system is the
same as in a bulk system. Hence, the *thick film* regime is defined by $\delta = \alpha q_{\perp}^2 d/B \gg 1$

The dynamics of the layers in the thick film regime is similar to that of a bulk smectic-A.
For fixed $q_\perp$, when $q_\perp^{(n)}$ is large such that $4\rho (B q_\perp^{(n)2} + K_1 q_\perp^4)/\eta_3^2 q_\perp^4 > 1$, the smectic layers are underdamped. This is basically the second sound of the layers, but with a correction due to surface tension and finite film thickness. When $4\rho (B q_\perp^{(n)2} + K_1 q_\perp^4)/\eta_3^2 q_\perp^4 \ll 1$ the motion of the layers is strongly overdamped. The decay rates which are slow and correspond to layer undulation are

$$iw_-(q_\perp, q_\perp^{(n)}) = \frac{Bq_\perp^{(n)2} + K_1 q_\perp^4}{\eta_3 q_\perp^2},$$

(36)

while the fast decay modes now correspond to the diffusive mode for $v_z$.

To study the dynamics of the smectic-A film in the thick film regime for typical laboratory materials, ($B \sim 2.5 \times 10^7$ dyn/cm$^2$, $\alpha \sim 30$ dyn/cm) we need $q_\perp^2 d \gg B/\alpha \sim 10^6$ cm$^{-1}$ ($\delta \gg 1$). For $d \sim 10^{-3}$ cm, which can be achieved easily, the momentum transfer in the $xy$-plane should be at least of the order $10^5$ cm$^{-1}$, which can also be achieved.

The crossover of the dynamics for the system is illustrated in Fig. 4, where the normalized dynamic correlation functions $C(q_\perp, n, t) = c(q_\perp, n, t)/c(q_\perp, 0, 0)$ for normal modes with $n = 0, 1, 2, 3$, are shown for a smectic-A film with thickness $d = 10$ µm, and typical elastic coefficients and viscosities. The in-plane wave vector $q_\perp$ is chosen such that the dimensionless parameter $\delta^{-1} = B/\alpha q_\perp^2 d$ is 20000, 2000, 1, and 0.1, respectively. When $\delta^{-1} = 20000$, the normal modes with $n > 0$ are negligible and $n = 0$ normal modes are underdamped. For $\delta^{-1} = 2000$ the $n > 0$ normal modes are still negligible but the $n = 0$ normal modes are overdamped with a decay rate which is not a single exponential. This occurs when the two time scales are close to each other, as illustrated in Fig. 3. For $\delta = 1$ the normal modes with $n > 0$ can be observed; this is the crossover regime between the thin film and thick film limits. For $\delta^{-1} = 0.1$ the contribution from bulk elasticity is sufficiently large that the dynamic correlation functions for normal modes with $n > 0$ are easily seen in the figure. Similar to the undulation mode in bulk systems, all normal modes decay exponentially. This is the overdamped limit of the thick film regime. The actual $q_\perp$ values in the figure change over many orders of magnitude. Thus, experimentally probing this entire figure requires different scattering probes.
V. CONCLUDING REMARKS

The layer dynamics of a freely standing smectic-A film have been determined by analyzing the response functions for the surface displacement. By determining the positions of the poles of the response functions in the complex $\omega$-plane, we find not only the frequencies but also the spatial configurations for the hydrodynamic modes of the system. The fact that $u$ and $v_z$ are two non-separable hydrodynamic variables, in general, is reflected in the form of the dynamic correlation function for $u$.

When the dimensionless thickness $\delta = \alpha q^2 d/B \ll 1$, the internal structure of the layers is not important for the system. For fixed $q_\perp$ one pair of normal modes dominates the statics and dynamics of the system. Furthermore, the layer fluctuations and the characteristic frequencies for these important normal modes show basically no dependence on the elasticity of the smectic layers, i.e., only surface tension and viscosity are important. To find information on the internal structure of the system, i.e., the smectic layers, it is necessary to excite other normal modes. As discussed in reference \[11\], exciting the $n = 1$ modes provides information on the crossover behavior of the system from a quasi-two-dimensional system to a three-dimensional system. This can be done by adjusting the experimental parameters such that $\delta = \alpha q^2 d/B$ is of order unity. The mode structure for the system in the regime $\delta \gg 1$ is similar to that of bulk systems. However, the characteristic time scales for the hydrodynamic modes now acquire surface and finite thickness corrections as well.

To summarize, we have explored the dynamical properties of a freely-standing smectic-A film in the linear regime. It is shown that existing experiments were performed in the limit where only the two-dimensional character of the film can be detected. Future experiments on the dynamical correlations of the smectic layers can reveal the crossover behavior of the system to a regime in which layer elasticity begins to play a role. Our work also provides the basis for a future theoretical study on the dynamics of a freely-standing smectic-A film far from equilibrium, where the system has been studied experimentally and shows behavior which is drastically different from that of a bulk system \[6\]. The effect of finite thickness and
surface tension will have to play a central role in a theory which describes such dynamical behavior.

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APPENDIX A:

We discuss permeation processes in a free-standing smectic-A film in this Appendix. We look for solution of the equations of motion with boundary conditions and external forces given in Section [1]. In the long wavelength, low frequency limit [12] the solution is of the form

\[ u = u_{el} + u_p, \tag{A1} \]

where

\[ u_{el}(r, t) = u_S(q, \omega) \cos [q_z(q, \omega)z] \times e^{i\mathbf{q}_\perp \cdot \mathbf{r}_\perp - i\omega t} + u_A(q, \omega) \sin [q_z(q, \omega)z] \times e^{i\mathbf{q}_\perp \cdot \mathbf{r}_\perp - i\omega t} \tag{A2} \]

is the approximate solution used in the text. It is independent of permeation constant. The second contribution

\[ u_p(r, t) = [u_p^+(q, \omega) \times e^{(1+i)p(q, \omega)(z-d/2)} + c.c] \times e^{i\mathbf{q}_\perp \cdot \mathbf{r}_\perp - i\omega t} + [u_p^-(q, \omega) \times e^{-(1+i)p(q, \omega)(z+d/2)} + c.c] \times e^{i\mathbf{q}_\perp \cdot \mathbf{r}_\perp - i\omega t} \tag{A3} \]

contains an inverse length, \( p(q, \omega) = \sqrt{q^2/2(\zeta m \eta_3)}^{1/2} \), which characterizes the decay of permeation away from the free surfaces. In typical materials \( p(q) \gg q \gg q_z \) in the low
frequency, long wavelength regime \[12\]. The amplitudes $u^\pm_p$ contain the contributions from permeation and are related to $u_{el}$ through the full boundary conditions.

From Eqs. (12) and (10) we find that $|u^\pm_p| \ll |u_{el}|$, i.e., the contribution of permeation to the layer displacement is small \[11\]. Then Eq. (9) leads to

$$\zeta^\pm \approx [u_{el}]_{z=\pm d/2}. \quad (A4)$$

It is straightforward to show that the elastic free energy is

$$F = \frac{1}{2} \int dV \left\{ B \left( \frac{\partial u}{\partial z} \right)^2 + K_1 \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right)^2 \right\}$$

$$+ \sum_{i=+, -} \frac{1}{2} \int dS^i \alpha \left\{ \left( \frac{\partial \xi^i}{\partial x} \right)^2 + \left( \frac{\partial \xi^i}{\partial y} \right)^2 \right\}$$

$$\approx \frac{1}{2} \int dV \left\{ B \left( \frac{\partial u_p}{\partial z} \right)^2 + B \left( \frac{\partial u_{el}}{\partial z} \right)^2 + K_1 \left( \frac{\partial^2 u_{el}}{\partial x^2} + \frac{\partial^2 u_{el}}{\partial y^2} \right)^2 \right\}$$

$$+ \sum_{i=+, -} \frac{1}{2} \int dS^i \alpha \left\{ \left( \frac{\partial u_{el}}{\partial x} \right)^2 + \left( \frac{\partial u_{el}}{\partial y} \right)^2 \right\}_{z=\pm d/2}. \quad (A5)$$

In the linear theory the contribution from permeation can be separated from the contribution from $u_{el}$. Since $|u^\pm_p| \ll |u_{el}|$ in the regime where we perform this long wavelength analysis, we can neglect the $u_p$ part. However, it is $u_{el}$, not the full displacement $u$, that can be expanded as a linear combination of the normal modes discussed in the text and satisfies bc 2’ (Eq.\((22)\)). When $u_p$ is included, the permeation force $B \partial u/\partial z$ vanishes on the free surfaces due to the existence of $u_p$. This is noted in the literature \[15\] but has not been included in the previous study of the static \[3\] and dynamic \[4\] theory for free standing smectic-A films.

**APPENDIX B:**

In this Appendix we calculate the dynamic correlation functions for $u$ and $v_z$. We consider the $u_{el}$ part only; the $u_p$ part simply corresponds to the permeation mode discussed in the literature \[1,8\]. Our starting point is the equations of motion, i.e., Eqs. (4) and (3), and the
dynamics of the normal modes labeled by \((q_\perp, n)\) are considered. Eliminating the pressure via the incompressibility condition, we find that there are two diffusive modes associated with \(v_x\) and \(v_y\), but \(v_x\) and \(v_y\) decouple from \(v_z\) and \(u\) \[\text{III}\]. Neglecting permeation, \[\text{II}\] the equations for \(v_z\) and \(u\) can be expressed as

\[
\frac{\partial}{\partial t} \begin{pmatrix} u \\ v_z \end{pmatrix} = \begin{pmatrix} 0 & 1 \\ -R & -D \end{pmatrix} \begin{pmatrix} u \\ v_z \end{pmatrix} \tag{B1}
\]

where

\[
D = \frac{1}{\rho} \eta_3 q_\perp^2 \tag{B2}
\]

represents the dissipative part, and

\[
R = \frac{1}{\rho} \left(Bq_z^{(n)}_z + K_1 q_\perp^4\right) \tag{B3}
\]

represents the reactive part. We have used the fact that \(q_z^{(n)} \ll q_\perp\) for all \(n\) in the regime of our calculation to simplify the expressions. The eigenvectors of the above matrix equation are

\[
\Phi_\pm = \frac{1}{\sqrt{1 + (i\omega_\pm(q_\perp, q_z^{(n)}_z))^2}} \begin{pmatrix} 1 \\ -i\omega_\pm(q_\perp, q_z^{(n)}_z) \end{pmatrix}, \tag{B4}
\]

with eigenvalues \(-i\omega_\pm(q_\perp, q_z^{(n)}_z)\) given in Eq. \(\text{III}\). Straightforward algebra leads to the following relation

\[
\begin{pmatrix} u(t) \\ v_z(t) \end{pmatrix} = \frac{1}{i\omega_+ - i\omega_-} \begin{pmatrix} [i\omega_+ e^{-i\omega_- t} - i\omega_- e^{-i\omega_+ t}] u(0) + [e^{-i\omega_- t} - e^{-i\omega_+ t}] v_z(0) \\ -\omega_+\omega_- [e^{-i\omega_- t} - e^{-i\omega_+ t}] u(0) + [i\omega_+ e^{-i\omega_- t} - i\omega_- e^{-i\omega_+ t}] v_z(0) \end{pmatrix} \tag{B5}
\]

where we used simplified notation, \(\omega_\pm = \omega_\pm(q_\perp, q_z^{(n)}_z)\), \(u(t) = u(q_\perp, n, t)\), \(v_z(t) = v_z(q_\perp, n, t)\), etc. From this one concludes that the dynamic correlation function for \(u\) is

\[
<u(q_\perp, n, t) u(-q_\perp, m, 0)> = \delta_{nm} \frac{i\omega_+ e^{-i\omega_- t} - i\omega_- e^{-i\omega_+ t}}{i\omega_+ - i\omega_-} \times <u(q_\perp, n, 0)^2> \times \frac{k_B T}{Bq_z^{(n)}_z^2 + K_1 q_\perp^4}, \tag{B6}
\]

as given in Section \[\text{III}\]. The calculation for the correlation function for \(v_z\) is similar to that for \(u\).
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Figure Captions

Figure 1. Schematic of a freely standing smectic-A film of thickness d. The y-axis is pointing into the paper. The dotted lines are the equilibrium positions of the free surfaces.

Figure 2. Graphical solution for the $q_z$'s for dimensionless parameter $B/\alpha q_\perp^2 d = 0.5$. The abscissa in all cases is $q_z d/\pi$. The three functions plotted are: $Bq_z/\alpha q_\perp^2$ (dot-dashed line), $\cot(q_z d/2)$ (solid lines), and $-\tan(q_z d/2)$ (dashed lines). The crossings of the dot-dashed line and the solid lines give the values of $q_z^{(n)} d/\pi$ for normal modes with $n$ even; the crossings of the dot-dashed line and the dashed lines (except the origin) give the values of $q_z^{(n)} d/\pi$ for normal modes with $n$ odd.

Figure 3. Decay rates for normal modes with $n = 0$ in the thin film regime. The dimensionless wave vector is defined as $Q = q_\perp d$. The material parameters are chosen as $\eta_3 = 1.0$ p, $\rho = 1.0$ g/cm$^3$, $d = 10$ $\mu$m, and $\alpha = 30$ dyn/cm. When the decay rates merge, the system undergoes underdamped motion.

Figure 4. Dimensionless dynamic correlation function $C(q_\perp, n, t) \equiv c(q_\perp, n, t)/c(q_\perp, 0, 0)$, for $n = 0, 1, 2, 3$. The material parameters are chosen to be $d = 10$ $\mu$m, $B = 2.5 \times 10^7$ dyn/cm$^2$, $K_1 = 10^{-6}$ dyn, $\alpha = 30$ dyn/cm, $\rho = 1.0$ g/cm$^3$, $\eta_3 = 1.0$ poise, and $q_\perp$ is chosen such that $\delta^{-1} = B/\alpha q_\perp^2 d = 20000, 2000, 1, and 0.1$, respectively.
Figure 1
Figure 2

$cot(q_z d/2)$

$Bq_z/\alpha q_l$

$-\tan(q_z d/2)$
Figure 3

Re[\omega](kHz)

0.5 0.6 0.7

Q

Figure 3
Figure 4