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Channel flow of smectic films

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The hydrodynamics of smectic films at an air-water interface is discussed, with particular focus on the viscous response of the film under flow normal to the layers. The corrections to the response functions of the smectic phase, arising from the coupling between the flow and the smectic order parameter, are calculated. The results for the effective viscosity are illustrated by analyzing smectic film flow in a channel geometry. Two limiting cases of the flow, namely, motion dominated by dislocation-induced shear-softening and dislocation-free motion dominated by the permeation mode of mass transfer, are studied. The effect of drag from a finite depth liquid subphase is considered. The results are compared to those for hexatic and liquid films.

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I. INTRODUCTION

The rheological properties of liquid crystalline systems continue to be of considerable interest because of their rich and complex behavior. Thin films of anisotropic molecules, such as Langmuir monolayers at an air-water interface, are relevant to many industrial applications, and as such, have been subjected to detailed experimental studies. In particular, the viscous response of liquid crystalline films is often found to be non-Newtonian. Among the dominant causes is coupling of the flow to molecular alignment. In smectic films (crystalline in one dimension, but liquidlike in the other), the presence of unbound dislocations becomes a major factor affecting viscous response. When the film is riding over another phase, usually water, viscous drag from this subphase, if large, can modify the flow profile of the film quite significantly.

The coupling between molecular alignment and flow has been seen in some cases in experiments by Mingotaud *et al.* [1], Maruyama *et al.* [2], and Kurnaz and Schwartz [3]. The experiments involve Langmuir monolayers of rod-shaped molecules that are usually tilted with respect to the surface normal, forming a hexatic phase with anisotropic in-plane bond orientations. Typically the film consists of domains of a liquid crystalline phase coexisting with another liquid crystalline phase or with the liquid expanded phase (no orientational order). The domains can be distinguished through Brewster angle microscopy which is sensitive to molecular orientation, making it possible to follow the shape and movement of the domains along the flow. There is evidence of nonlinear shear response [3,4] emerging from such studies, as well as of the molecular orientation being influenced by flow [1,2].

Shear thinning has often been observed in experiments involving Langmuir monolayers [3,4]. A possible explanation of this phenomenon is provided by Bruinsma *et al.* [5] in terms of shear-induced defect proliferation. Dislocation defects in a solid, if unbound, can relax an applied strain by moving in response to the resulting stress. Since the force on such a defect depends on its ''charge,'' oppositely charged defects tend to separate under an external stress. Tightly bound pairs cannot contribute to the steady state viscous response, although they can modify the response at nonzero frequencies and wave vectors [6]. However, at finite temperatures, bound pairs can dissociate under this separating influence which effectively tilts the potential well confining the pair, liberating them once they escape beyond a potential barrier. Shear thinning might be expected once the stress-induced density of free dislocations becomes comparable to those present due to thermal activation. Being thermally activated, the free dislocation density, and hence the viscous response, should be very sensitive to temperature. Experiments conducted by Schwartz [7] on anisotropic hexatic and crystalline phases of Langmuir monolayers do indeed see a strong temperature dependence of the critical shear rate for onset of non-Newtonian behavior.

In this paper we study a simpler problem, the linear hydrodynamics of two-dimensional smectic films in a channel flow geometry. Dislocations still play an important role, and it is easier to analyze their effect on the smectic order embodied in a *single* set of Bragg planes. Although we do not study this here, channel flow of two-dimensional smectics would also be a promising context in which to explore a tractable model of shear thinning.

In the absence of external strains, free dislocations or disclinations don't occur in the most ordered two-dimensional phases; they are instead bound in pairs of opposite charges by a logarithmic potential. However, in two-dimensional layered materials such as smectics or cholesterics, there is exponential decay of translational order in both the layering direction, and the liquidlike direction along the layers [8]. As a result, isolated dislocations have a finite energy and exist in a finite concentration at any finite temperature. In these materials, shear response at long wavelengths is primarily due to the free dislocations; the viscosity diverges inversely as the dislocation density when it becomes small at low temperatures. This divergence is cut off at short length scales by the permeation mode of mass transfer in smectics, where a layer distortion induces molecules to jump from layer to layer without affecting the layering structure, allowing the distortion to relax over a finite distance.

In three dimensions, one finds diverging smectic response functions near the second-order smectic-to-nematic transition. The coupling of the nematic order parameter to fluctuations in the magnitude of the smectic order parameter causes, among other quantities, the permeation constant of the smectic and the viscosity denoted η_1 in the literature, to diverge [9]. However, in two dimensions, a dislocation-driven thermodynamic transition to the nematic state occurs at zero temperature [8]. At a finite temperature, the nematic melts into an isotropic liquid via a disclination unbinding transition. Below this temperature, local smectic order is disrupted by singularities in the phase of the order parameter (i.e., dislocations). However, the local smectic order parameter has a finite magnitude, and fluctuations in the magnitude are irrelevant in the renormalization sense [10]. Thus renormalization of the elastic coefficients and response functions as in the three-dimensional (3D) case does not occur.

Coupling of the film flow to a subphase (a fluid body supporting the film on its surface) can significantly alter its flow profile. Such experiments have been conducted by Schwartz and co-workers [11,3] using Langmuir monolayers on water. When the subphase drag dominates the flow, the flow profile becomes semielliptical. Stone [12] has performed calculations which confirm this profile and also yield the profiles interpolating between the elliptical and the parabolic, as the viscosity of the film relative to that of the subphase is increased. The depth of the subphase was also a parameter in the calculations, since decreasing the depth results in increased drag. As we show below, Stone's results can also be applied to hexatic films.

In the next section, we briefly review the equilibrium properties of two-dimensional hexatic [13] and smectic [8] films. Section III discusses the hydrodynamics of two-dimensional smectics, and the implications of the coupling between the smectic order parameter and the flow for the response functions of both quantities. Section IV looks at flow of a smectic film in a channel flow geometry, and examines the behavior in different regimes of the channel width. In Sec. V, we consider the effect of subphase drag on the smectic film flow as compared to previous results for an isotropic film. The results of both Secs. IV and V are consistent with the results of Sec. III for the effective viscosity. The last section summarizes the results of this paper.

II. REVIEW OF HEXATIC AND SMECTIC FILMS

It was recognized quite some time ago that two-dimensional films allow for an unusual phase diagram characterized by an intermediate hexatic phase, separating the solid and conventional liquid phase. The origin of this phenomenon lies in the pronounced role of dislocations, i.e., pointlike translational defects in a two-dimensional crystal. Dislocations interact via elastic deformations of the solid similar to charges in a two-dimensional Coulomb gas. Below the melting temperature dislocations of opposite Burgers vector are bound in pairs and the overall effect is a finite renormalization of the elastic moduli. There is quasi-long-range translational and long-range orientational order which manifests itself in a structure factor as measured by, e.g., x-ray scattering, which exhibits a regular array of quasi-Bragg peaks.

At the melting temperature the solid melts into a hexatic when these dislocations unbind and destroy the quasi-long-range translational order just as in a liquid. However, the hexatic retains a quasi-long-range sixfold symmetry. As a consequence the structure factor now exhibts concentric diffraction rings which have an angular modulation superimposed (this modulation vanishes as a small power of the system size with increasing illumination area). At a still higher temperature this modulation disappears and the hexatic melts into a conventional isotropic liquid with both short-ranged translational and orientational order.

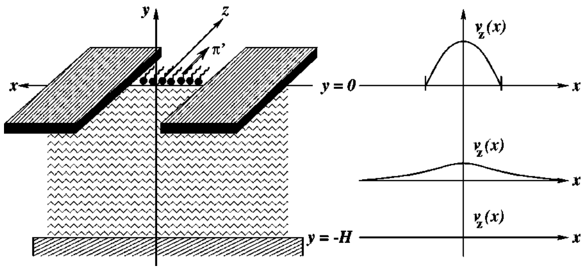

FIG. 1. Channel flow geometry with a subphase. The film and the barriers forming the channel are on the surface of a water table. A surface pressure gradient is applied to the film along the channel. On the right are the typical flow profiles at various depths.

The free energy of a hexatic involves an addtitional term

$$\delta\mathcal{F}=\frac{1}{2}K_A\int d^2r|\nabla\theta|^2, \tag{2.1}$$

which describes the long-wavelength distortions of the local bond angle θ. The temperature-dependent stiffness constant K_A is finite in the hexatic phase. A renormalization calculation shows that K_A jumps from an universal value to zero when the hexatic melts via a Kosterlitz-Thouless transition into an isotropic liquid.

The hydrodynamics of partially ordered hexatic films has been studied in detail by Zippelius *et al.* [13]. These authors find a correction to the effective viscosity under flow conditions where the hexatic bond orientation is pinned at the boundaries, as compared to the case where the bond orientation is free to rotate. The correction comes from coupling of the flow to the bond orientation order parameter under the constraint imposed by the boundaries (see Appendix A of Ref. [13]).

A similar coupling can be enforced by imposing a pressure gradient on the flow. In the experiments conducted by Kurnaz and Schwartz [3] on hexatic film flow, the domain structure of the hexatic mesophase can impose constraints on the bond orientation at domain boundaries, thus increasing the viscosity from its bare value. Annealing of the domains would then lead to a reduction in the effective viscosity. The experimental signature of this effect would be a time-dependent viscous response. Some transients have indeed been observed in these experiments, although other factors may be involved, such as domain boundary elasticity [7] and shear thinning.

To illustrate how to incorporate effects of a subphase into the hydrodynamics of a partially ordered film, we adapt the analysis of Ref. [12] to films with hexatic order. In the presence of a subphase, the hydrodynamic equations of motion for a hexatic are modified by adding a subphase drag term to the viscous force: denoting by x the co-ordinate across the channel, z along the channel, and y along the channel depth (see Fig. 1), one derives the equations of motion [13]

$$\frac{\partial g_z}{\partial t}=\pi'+\partial_x\left(\eta\partial_x v_z-\frac{K_A}{2}\partial_x^2\theta\right)-\eta_b\,\partial_y v_z|_{film\ surface}, \tag{2.2a}$$

$$\frac{\partial\theta}{\partial t}=\frac{\partial_x g_z}{2\rho}+\Gamma_6 K_A\partial_x^2\theta, \tag{2.2b}$$

where $\mathbf{g}=\rho\mathbf{v}$ is the surface momentum density, Γ_6 the kinetic coefficient corresponding to dissipative processes of the hexatic bond-angle order, η the surface shear viscosity, η_b the viscosity of the bulk subphase, and π' the surface pressure gradient driving the film down the channel.

Assuming $\partial_x\theta$ is time independent in the steady state, we have $\partial_t\partial_x\theta=0$. Therefore $\partial_t\theta$ must be constant across the channel. Since g_z is an even function of x in this flow situation, $\partial_t\theta$ is odd, and hence, must be zero. Equation (2.2b) then gives us the coupling between the flow and the bond orientation:

$$\partial_x^2\theta=-\frac{1}{2\rho\Gamma_6 K_A}\partial_x g_z\,. \tag{2.3}$$

Upon substituting this result into Eq. (2.2a), we find

$$\frac{\partial g_z}{\partial t}=0=\pi'+\left(\eta+\frac{1}{4\Gamma_6}\right)\partial_x^2 v_z-\eta_b\,\partial_y v_z|_{film\ surface}. \tag{2.4}$$

Thus g_z obeys an equation of motion identical to that for an isotropic film with a subphase, but with the modified viscosity $\eta+1/4\Gamma_6$, and we can take over the results of Ref. [12]. As we shall see (Sec. V), this simplification does not apply to smectic flow.

In contrast to hexatics, smectics are characterized by a crystal-like periodic modulation of the density along one direction, say, the z direction, and liquidlike correlations perpendicular to it. In two dimensions, we take this to be the x direction. The preferred orientation of the ‘‘layers’’ is also the average direction along which the directors $\hat{\mathbf{n}}$ of the nematic molecules are oriented. Although it represents a spontaneously broken rotational symmetry, the layer orientation can be forced by boundary conditions on the molecules, or even by flow. Smectic order is characterized by a wave vector $\mathbf{q}_0=\hat{\mathbf{z}}2\pi/d$, where d is the layer spacing, usually slightly larger than the molecular length.

The smectic density wave can be represented as [14]

$$\rho(\mathbf{r})=\rho_0[1+\psi(\mathbf{r})e^{i\mathbf{q}_0\cdot\mathbf{r}}]. \tag{2.5}$$

Here, $\psi(\mathbf{r})$ is the complex smectic order parameter: its amplitude represents the strength of the smectic ordering, whereas the phase $\phi(\mathbf{r})=q_0 u(\mathbf{r})$ describes the phonons associated with broken translational symmetry along the layering direction. Phonons in two dimensions are very effective in destroying the one-dimensional translational order: The correlation $\langle\psi(\mathbf{r})\psi^*(\mathbf{0})\rangle$ decays as the exponential of a power of the displacement. Since the square of the wave vector appears in the exponent [8], the contributions of higher harmonics of q_0 in the density modulation are less important.

The Landau-Ginzburg free energy describing the smectic takes the form [15]

$$\mathcal{F}=\int d^2r\left[\frac{a}{2}|\psi|^2+\frac{u}{4}|\psi|^4+\frac{c_\parallel}{2}|\partial_z\psi|^2+\frac{c_\perp}{2}|(\partial_x-iq_0\delta n)\psi|^2+\frac{K_1}{2}(\partial_x\delta n)^2+\frac{K_3}{2}(\partial_z\delta n)^2\right]. \tag{2.6}$$

Here, K_1 and K_3 are splay and bend elastic constants. The twist elastic constant K_2 is absent in two dimensions. The coupling between $\partial_x\psi$ and $\delta n\equiv(\hat{\mathbf{n}}-\hat{\mathbf{z}})\cdot\hat{\mathbf{x}}$ is required to satisfy the rotational invariance of $\mathcal{F}$. Terms of order higher than quadratic in the order parameter and its gradients have been neglected. Well below the mean-field smectic-nematic transition temperature, fluctuations in the amplitude of ψ can also be ignored, and in the absence of singularities in $\hat{\mathbf{n}}$ (disclinations), δn can be integrated out.The remaining long-wavelength fluctuations can be expressed completely in terms of the layer displacement $u(\mathbf{r})$ as [14,8]

$$\mathcal{F}=\int d^2r\frac{1}{2}B[(\partial_z u)^2+\lambda^2(\partial_x^2 u)^2], \tag{2.7}$$

with $B=\psi_0^2q_0^2c_\parallel$, and $\lambda^2=K_1/B$. Uniform gradients of u along the layer direction $(\partial_x u)$ don’t cost any energy, because they represent tilting of the layering direction. This important difference compared to two-dimensional solids, hexatics, etc., implies that the lowest energy defects in the system, dislocations, have a *finite* energy E_D and are not constrained to be bound in pairs at low temperatures [8].

Whereas a smectic with thermally excited phonons would behave like a nematic with only a splay degree of freedom, the presence of dislocations allows for bend in the average layer orientation over scales larger than the typical size ξ_D of a correlated ‘‘smectic blob’’ [8], given by $\xi_D^2\equiv n_D^{-1}\approx a_D^2e^{E_D/k_BT}$ (a_D is a dislocation core diameter, $a_D^2\sim d\sqrt{\lambda d}$). Therefore the long-wavelength behavior of the smectic is that of a nematic with free energy

$$\mathcal{F}=\int d^2r\frac{1}{2}[K_1(\partial_x\delta\hat{\mathbf{N}})^2+K_3(\partial_z\delta\hat{\mathbf{N}})^2], \tag{2.8}$$

where $\hat{\mathbf{N}}$ denotes the layer normal, and $K_3\propto\xi_D^2$. As discussed by Nelson and Pelcovits [16], nonlinearities in the nematic free energy modify the nematic Frank constants K_1 and K_3 such that at scales longer than $de^{\xi_D^2/a^2}$ the nematic can be described by a single Frank constant $\propto\xi_D^2$. In practice, this length scale can be very large compared to typical system sizes, so one usually sees a constant nematic described by two Frank constants. A study of the dynamics of smectic films, taking dislocations into account [8], yields nematic behavior corresponding to Eq. (2.8) at long length scales, with a nematic kinetic coefficient that vanishes like $n_D\propto e^{-E_D/k_BT}$ at low temperatures.

III. SMECTIC HYDRODYNAMICS

The hydrodynamic variables for a two-dimensional smectic are the layer displacement u, and the conserved momentum densities g_x, g_z. In this section we focus for simplicity on the dynamics of free-standing smectic films [17], where the momentum is conserved to a good approximation. The drag due to a liquid subphase is considered in Sec. V. We assume the smectic to be incompressible, and so neglect density fluctuations, setting the density $\rho=$const and consequently $\partial_x g_x+\partial_z g_z=0$. The viscous stress tensor of an uniaxial two-dimensional fluid is characterized by four independent viscosities (as opposed to five for the three-dimensional case). Incompressibility reduces this number to 2. The independent components of the symmetric (traceless) strain rate tensor can be written as $(\partial_z g_x+\partial_x g_z)/2$ and $(\partial_x g_x-\partial_z g_z)/2$, respectively, which have to be related to the viscous stress tensor σ'_{ij}. Symmetry implies $\sigma'_{xz}=\nu(\partial_z g_x+\partial_x g_z), \sigma'_{xx}-\sigma'_{zz}=(\nu-\nu')(\partial_x g_x-\partial_z g_z)$. The trace $\sigma'_{xx}+\sigma'_{zz}$ can be absorbed in the pressure, and in the following we assume that this has been done already. Upon denoting

$$h\equiv-\frac{\delta\mathcal{F}}{\delta u}=B(\partial_z^2-\lambda^2\partial_x^4)u, \tag{3.1}$$

the equations of motion can be written as [14]

$$\frac{\partial u}{\partial t}=\frac{g_z}{\rho}+\lambda_p h, \tag{3.2a}$$

$$\frac{\partial g_x}{\partial t}=-\partial_x p+\nu\partial_z^2 g_x+\nu'\,\partial_x\partial_z g_z, \tag{3.2b}$$

$$\frac{\partial g_z}{\partial t}=h-\partial_z p+\nu\partial_x^2 g_z+\nu'\,\partial_z\partial_x g_x, \tag{3.2c}$$

where p is the surface pressure and λ_p the permeation constant for the smectic. Permeation refers to the dissipative mode of mass transfer in smectics where the molecules jump from layer to layer in order to relax a layer distortion.

Dislocations in the smectic introduce cuts into the displacement field, but it is possible to define locally the gradient $\mathbf{s}=\nabla u$ as a single-valued quantity [8]. In the presence of dislocations, s_x and s_z are considered as independent variables. Since there are cuts in the displacement field, the line integral $\int_\Gamma \mathbf{s}\cdot d\mathbf{r}$ does not vanish for closed loops Γ if the loop encloses dislocations. This line integral counts the number of cuts in units of the layer spacing d. Stokes' theorem implies $\nabla\times\mathbf{s}=-\hat{\mathbf{y}}dm(\mathbf{r})$, where $m(\mathbf{r})$ is the dislocation density. Since the number of dislocations is conserved one has the continuity equation

$$\partial_t m+\nabla\cdot\mathbf{J}_D=0, \tag{3.3}$$

where $\mathbf{J}_D$ is the two-dimensional dislocation current. The time evolution of the "strain" $\mathbf{s}$ can be inferred from its irrotational and its solenoidal part

$$\frac{\partial\mathbf{s}}{\partial t}=\nabla\frac{\partial u}{\partial t}+d\hat{\mathbf{y}}\times\mathbf{J}_D. \tag{3.4}$$

A Fokker-Planck description for the diffusion of the dislocations in the strained smectic yields an expression for the dislocation current [8]

$$\mathbf{J}_D=n_D\underline{\Gamma}\cdot\mathbf{f}-T\underline{\Gamma}\cdot\nabla m. \tag{3.5}$$

Here we have introduced the unsigned dislocation density n_D; we have also set $k_B=1$ for convenience. The mobility tensor $\underline{\Gamma}$ is diagonal in the coordinate system aligned with the preferred axis of the smectic. Since the principal values Γ_z and Γ_x correspond to dislocation glide and climb, respectively, we expect $\Gamma_z\gg\Gamma_x$. A strain results in a net force on dislocations, viz., the 2D analog of the Peach-Koehler force $\mathbf{f}=d(Bs_z, B\lambda^2\partial_x^2 s_x)$. Thus strain can be released by a flow of dislocations. This process is similar to the charge separation due to an applied electric field in a semiconductor. Note that the Einstein relation connects the mobility embodied in the first term to the diffusion constant implicit in the second through the common matrix $\underline{\Gamma}$.

For later purposes we also include external stresses σ_{ij}^{ext} in the momentum balance. The equations of motion are then

$$\frac{\partial g_x}{\partial t}=-\partial_x p+\nu\partial_z^2 g_x+\nu'\,\partial_x\partial_z g_z-\partial_x\sigma_{xx}^{ext}-\partial_z\sigma_{xz}^{ext}, \tag{3.6a}$$

$$\frac{\partial g_z}{\partial t}=B(\partial_z s_z-\lambda^2\partial_x^3 s_x)-\partial_z p+\nu\partial_x^2 g_z+\nu'\,\partial_z\partial_x g_x-\partial_x\sigma_{zx}^{ext} -\partial_z\sigma_{zz}^{ext}, \tag{3.6b}$$

$$\frac{\partial s_x}{\partial t}=\partial_x\frac{g_z}{\rho}+\lambda_p B(\partial_x\partial_z s_z-\lambda^2\partial_x^4 s_x)+\Gamma_z[n_D d^2 B\lambda^2\partial_x^2 s_x -T\partial_z(\partial_x s_z-\partial_z s_x)], \tag{3.6c}$$

$$\frac{\partial s_z}{\partial t}=\partial_z\frac{g_z}{\rho}+\lambda_p B(\partial_z^2 s_z-\lambda^2\partial_x^3\partial_z s_x)-\Gamma_x[n_D d^2 B s_z -T\partial_x(\partial_x s_z-\partial_z s_x)]. \tag{3.6d}$$

Ignoring the external stresses one can calculate the eigenmodes of the system. If conservation of momentum is neglected [8], Eqs. (3.6) lead in the limit of long wavelengths and low frequencies to a relaxation frequency for s_z (which describes layer compression)

$$\omega_{s_z}=-i\Gamma_x n_D d^2 B, \tag{3.7}$$

and for s_x (i.e., layer undulations) a diffusive frequency

$$\omega_{s_x}(\mathbf{q})=-i(\Gamma_z n_D d^2 B\lambda^2 q_x^2+T\Gamma_z q_z^2). \tag{3.8}$$

Using the relation $\delta n=\partial_x u$ [14], this last result corresponds to a nematiclike behavior for the director $\hat{\mathbf{N}}=\hat{\mathbf{z}}+s_x\hat{\mathbf{x}}$. Including g_x, g_z in the hydrodynamic treatment introduces a pair of coupled g-s_x modes with both diffusive and propagating characteristics. The pressure is eliminated via the incompressiblity condition in Fourier space $q_x g_x+q_z g_z=0$. The use of the transversal momentum density $qg_\perp=q_z g_x-q_x g_z$ results to leading order in the wave vector in

$$-i\omega g_\perp = -iB\frac{q_x}{q}(q_z s_z + \lambda^2 q_x^3 s_x) - \nu q^2 g_\perp + (2\nu + 2\nu')\frac{q_x^2 q_z^2}{q^2} g_\perp , \tag{3.9a}$$

$$-i\omega s_x = -i\frac{q_x^2}{q\rho} g_\perp - (\Gamma_z n_D d^2 B \lambda^2 q_x^2 + T\Gamma_z q_z^2) s_x - (\lambda_p B - T\Gamma_z) q_x q_z s_z , \tag{3.9b}$$

$$-i\omega s_z = -i\frac{q_x q_z}{q\rho} g_\perp - \Gamma_x n_D d^2 B s_z + T\Gamma_x q_x q_z s_x . \tag{3.9c}$$

Again one finds the layer compression mode, Eq. (3.7), for s_z. Denoting $i\omega_{g_\perp}(\mathbf{q}) = \nu q^2 - [2\nu + 2\nu' - 1/(\rho \Gamma_x n_D d^2)] q_x^2 q_z^2 / q^2$, the two remaining modes have characteristic frequencies

$$\omega(\mathbf{q}) \approx \left(\frac{\omega_{g_\perp} + \omega_{s_x}}{2}\right) \pm \sqrt{\left(\frac{\omega_{g_\perp} - \omega_{s_x}}{2}\right)^2 + \frac{q_x^4}{q^2 \rho}\left(B\lambda^2 q_x^2 + \frac{T}{n_D d^2} q_z^2\right)} . \tag{3.10}$$

Propagation dominates for $\mathbf{q} \| \hat{\mathbf{x}}$ if the dissipation is small enough, which is possible at low temperatures, leading to

$$\omega(\mathbf{q}) = \pm \sqrt{\frac{B}{\rho}} \lambda q_x^2 . \tag{3.11}$$

As in the case of hexatics, coupling to the smectic displacement field modifies the viscosity of the film. In the absence of dislocations, it is not possible to shear the smectic film perpendicular to the layers without breaking it. The glide motion of dislocations facilitates shear deformation. Permeation can also support shear at short length scales. To calculate the effective viscosity, we apply a static external stress $\sigma_{ij}(\mathbf{q})$ to the system, and calculate the steady state response for $g_i(\mathbf{q})$. We consider the two special cases $\sigma_{zx}(\mathbf{q} \| \hat{\mathbf{x}})$ and $\sigma_{xz}(\mathbf{q} \| \hat{\mathbf{z}})$. In the former case one derives

$$\sigma_{zx}(q_x) = i q_x \left(\nu + \frac{1/\rho}{\Gamma_z n_D d^2 + \lambda_p q_x^2} \right) g_z(q_x), \tag{3.12}$$

which suggests to introduce an effective viscosity ($\eta = \rho\nu$)

$$\eta^{eff}(q_x) = \eta \left(1 + \frac{\Delta^{-1}}{1 + \delta^2 q_x^2} \right), \tag{3.13}$$

with the dimensionless dislocation density $\Delta = \eta \Gamma_z n_D d^2$ and the effective permeation length $\delta = \sqrt{\lambda_p / (\Gamma_z n_D d^2)}$. Note that for a three-dimensional smectic the permeation length is conventionally defined as $\sqrt{\eta \lambda_p}$, i.e., the length where permeation and viscous damping become equally important. Here the viscous process is replaced by dislocation glide. In the long-wavelength limit the form of the viscositiy is similar to the corresponding correction for hexatics: $\eta \rightarrow \eta(1 + 1/(4\eta\Gamma_6))$.

At low temperatures (or large dislocation energy E_D), n_D rapidly approaches 0 as $e^{-E_D/T}$, and the effective viscosity of the smectic begins to diverge as $e^{E_D/T}$. However, since the permeation mode relaxes shear over scales shorter than the permeation length δ, the divergence of the shear viscosity is cut off for $q_x \gg 1/\delta$.

Since these hydrodynamic equations are valid only for wavelengths longer than the dislocation correlation length in the x direction, i.e., $q_x \ll \xi_\perp^{-1}$ where $\xi_\perp = (\lambda \xi_D^2)^{1/3}$ [8], this rounding off of the viscosity will extend to the hydrodynamic range only if $\xi_\perp / \delta = (\lambda \xi_D^2)^{1/3} / \delta \ll 1$. We expect the bare viscosity η and $\lambda = \sqrt{K_1/B}$ to stay finite as $T \rightarrow 0$. However, ξ_D diverges as $e^{E_D/2T}$. We expect the permeation constant λ_p to behave like $e^{-E_p/T}$ where E_p is the energy barrier for molecules to jump from one layer to the next. The dislocation kinetic coefficient Γ_z would similarly correspond to the activation energy E_g for dislocation glide by breaking and reforming of bonds around the dislocation core. But this energy barrier should be small compared to that required for molecular hopping across the layers, and we shall ignore it in comparison. Then the above condition is satisfied provided $E_D/3 > E_p$, so that $\xi_D^{2/3} \rightarrow \infty$ faster than $\delta \rightarrow \infty$.

For an external stress $\sigma_{xz}(\mathbf{q} \| \hat{\mathbf{z}})$ the stress-strain relation is given simply by

$$\sigma_{xz}^{ext}(q_z) = -i\nu q_z g_x(q_z). \tag{3.14}$$

Thus the viscosity is not modified by the presence of dislocations. The reason is of course that the smectic layers already exhibit a liquidlike response to shear parallel to the layers and no layers have to be broken.

Although we have assumed the viscosity to be independent of shear rate, at high shear rates we must account for shear thinning brought about by the increase in unbound dislocations in the presence of the shear strain. The mechanism for dislocation proliferation under a shear stress is similar to that described by Bruinsma *et al.* [5] for a 2D crystal of point particles. The stress tilts the effective potential well binding the dislocation pair, allowing the pair to dissociate. The extra density of unbound dislocations facilitates further relaxation of the stress so that the effective viscosity decreases with increasing shear rate (the shear strain in the steady state depends on the shear rate imposed upon the flow). Note from Eq. (3.13) that the effective viscosities do indeed drop with increasing dislocation density n_D.

The same mechanism would also apply to shear flow in a hexatic film where disclination unbinding would occur in the presence of a strain in the bond-orientation angle. Since the orientational order parameter is coupled to the flow as in Eqs. (2.2), disclinations can mediate the shear thinning mechanism in the hexatic phase.

IV. CHANNEL FLOW OF SMECTIC FILMS

We are interested in flow under shear or a pressure gradient for a film oriented with the layering direction along the channel (see Fig. 2). From the previous discussion, we expect a nematiclike profile for s_x unless the dislocation den-

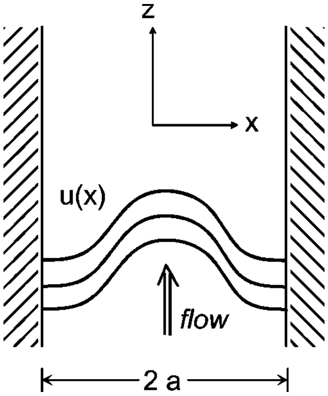

FIG. 2. Channel flow under the influence of a surface pressure gradient along the channel: the smectic layers are normal to the flow.

sity is small, in which case we are in the permeation regime and shear is only supported in a boundary layer. We assume the channel is much wider than the dislocation correlation length ξ_D, so that the hydrodynamic treatment is valid. Discussion of the effects of a subphase will be deferred to Sec. V.

In the steady state, we expect s_z to be constant. Translational invariance along the channel implies vanishing z derivatives except for the pressure head $\pi' = -\partial_z p = \text{const}$. Correspondingly, the equations of motion (3.6) reduce to

$$0=\pi' + \nu\partial_x^2 g_z - B\lambda^2\partial_x^3 s_x, \tag{4.1a}$$

$$0=\partial_x \frac{g_z}{\rho} - \lambda_p B\lambda^2\partial_x^4 s_x + \Gamma_z n_D d^2 B\lambda^2\partial_x^2 s_x. \tag{4.1b}$$

The solution shall be expressed in terms of the dimensionless dislocation density $\Delta = \eta\Gamma_z n_D d^2$, and the effective permeation length $\delta = \sqrt{\lambda_p/(\Gamma_z n_D d^2)}$. In the presence of dislocations, it is convenient to define the characteristic length $l = \sqrt{\Delta/(1+\Delta)}\,\delta = \sqrt{\eta\lambda_p/(1+\Delta)}$. The channel is bounded by walls at $x=\pm a$. Upon solving the equations above with the no-slip boundary condition at $g_z(x=\pm a)=0$ and the permeation current $\propto \partial_x^3 s_x(x=\pm a)=0$ [see Eqs. (3.1) and (3.2a)], we find

$$g_z(x) = \frac{\pi'/\nu}{1+\Delta}\left[\Delta\frac{(a^2-x^2)}{2} + l^2\left(1-\frac{\cosh(x/l)}{\cosh(a/l)}\right)\right]. \tag{4.2}$$

There are two regimes of interest here.

(i) Narrow channel: $a \ll l$:

$$g_z \to \frac{\pi'}{\nu}\frac{(a^2-x^2)}{2},$$

i.e., we recover the usual Poiseuille profile expected for a structureless fluid.

(ii) Wide channel: $a \gg l$: There are two distinct contributions to g_z (Fig. 3):

$$g_z \to \frac{\pi'/\nu}{1+\Delta}\left[\Delta\left(\frac{a^2-x^2}{2}\right) + l^2(1-e^{-(a-|x|)/l})\right] \quad \text{for} \quad l \ll |x| \leq a.$$

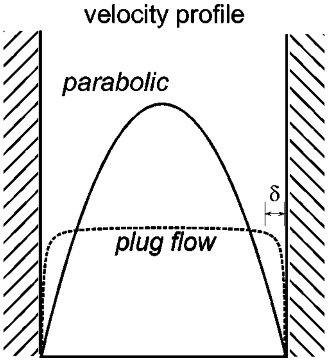

FIG. 3. In the wide channel/macroscopic limit, there are two contributions to flow: the solid line represents the parabolic profile due to dislocation-assisted shear, whereas the dotted line represents the plug-flow profile characteristic of flow in the permeation regime.

If $a^2\Delta \gg l^2$, then the second term can be neglected and dislocations restore a fluid like response, but with an effective viscosity

$$\eta^{eff} = \eta\left(1+\frac{1}{\Delta}\right), \tag{4.3}$$

confirming the result we found in the previous section. On the other hand, if the dislocation density is so small that $a^2\Delta \ll l^2$, then the second term dominates and one recovers the plug flow profile characteristic of permeation flow.

For the general case, we can estimate the effective viscosity from the flow rate: for Poiseuille flow, the momentum flux is given by $\int_{-a}^{a} g_z dx = 2\pi' a^3\rho/(3\eta)$. Using this as the *definition* of η^{eff}, we find

$$\frac{\eta}{\eta^{eff}} = \frac{1}{1+\Delta}\left[\Delta + 3\left(\frac{l}{a}\right)^2 - 3\left(\frac{l}{a}\right)^3 \tanh\frac{a}{l}\right]. \tag{4.4}$$

For $a \gg l$,

$$\eta^{eff} = \eta\left(\frac{1+\Delta}{\Delta + 3l^2/a^2}\right), \tag{4.5}$$

which reduces to Eq. (4.3) for $a^2\Delta \gg l^2$, whereas for $a^2\Delta \ll l^2$, and hence $\Delta \ll 1$,

$$\frac{\eta^{eff}}{\eta} = \frac{a^2}{3l^2} \quad \text{or} \quad \eta^{eff} = \frac{a^2}{3\lambda_p}, \tag{4.6}$$

reminiscent of the result $1/\lambda_p q_x^2$ we found for low dislocation densities in the previous section.

For $a \ll l$, we have

$$\frac{\eta}{\eta^{eff}} = 1 - \frac{2}{5}\frac{1}{1+\Delta}\frac{a^2}{l^2} \quad \text{or} \quad \eta^{eff} = \eta + \frac{2}{5}\frac{a^2}{\lambda_p}, \tag{4.7}$$

which is a small correction due to the permeation boundary layer.

V. CHANNEL FLOW WITH A NORMAL FLUID SUBPHASE

In practice [11], both the film and the barriers that restrict its flow to a channel geometry, float on a volume of fluid with viscosity η_b and finite depth H (Fig. 1). If the dimensionless parameter [12]

$$\Lambda = a\,\eta_b/\eta$$

is small, then, as for a normal film, the effect of this subphase can be neglected, and the analysis of the previous section is sufficient to describe the two-dimensional film flow. If the subphase drag cannot be neglected, the flow profile can be calculated through an analysis similar to Ref. [12]. We outline the steps here, and comment on the limiting cases.

We consider a subphase extending from $y=0$ (surface with film) to $y=-H$ (bottom). Let $\vec{v}(x,y)$ be the (z independent) velocity field describing the subphase, $\vec{v} \equiv (v_x, v_y, v)$. The velocity profile in the film itself is $v_0(x) \equiv v(x, y=0)$.

In steady state, the equation of motion for $\vec{v}$ in the bulk of the subphase is

$$(\partial_x^2 + \partial_y^2)\vec{v} = 0.$$

The boundary conditions on the subphase are:

$\vec{v} = \vec{0}$ for $x \to \pm\infty$ or $y=-H$ or $y=0$, $|x|>a$,

and

v_x and $v_y = 0$ for $y=0$, $|x|<a$

as well. We assume that the subphase is incompressible ($\vec{\nabla}\cdot\vec{v}=0$), which implies that v_x and v_y must be zero.

For the film, the equations of motion at the surface are modified by the subphase drag:

$$\pi' + \eta\partial_x^2 v_0 - B\lambda^2\partial_x^3 s_x - \eta_b \partial_y v|_{y=0} = 0, \tag{5.1a}$$

$$\partial_x(v_0 - \lambda_p B\lambda^2 \partial_x^3 s_x) + \Gamma_z n_D d^2 B\lambda^2 \partial_x^2 s_x = 0. \tag{5.1b}$$

It is convenient to scale variables such that

$$x \to ax, \quad y \to ay, \quad v \to \left(\frac{\pi' a^2}{\eta}\right) v,$$

and

$$\partial_x^3 s_x \equiv \left(\frac{\pi'}{B\lambda^2}\right) v_p, \tag{5.2}$$

as well as

$$\delta \to a\delta, \quad H \to aH. \tag{5.3}$$

All quantities are now dimensionless. Since v_p is proportional to the "permeation current," it obeys the same boundary conditions as v_0.

The equations of motion can now be written as

$$\partial_x^2 v + \partial_y^2 v = 0 \quad \text{for} \quad -H<y<0$$

$$\text{with} \quad v=0 \quad \text{at} \quad y=0,\ -H, \tag{5.4a}$$

$$1 + \partial_x^2 v_0 - v_p - \Lambda \partial_y v|_{y=0} = 0, \tag{5.4b}$$

$$\partial_x^2 v_0 = \Delta(\delta^2 \partial_x^2 - 1) v_p, \tag{5.4c}$$

where δ and Δ were defined in Sec. IV. Equation (5.4a) implies that v must have the form

$$v(x,y) = \int_0^\infty dk \;\frac{A(k)}{\cosh kH} \cos kx \sinh k(H+y). \tag{5.5}$$

In terms of the Fourier transform $A(k)$, the film velocity is then

$$v_0(x) = \int_0^\infty dk \; A(k) \tanh(kH) \cos kx. \tag{5.6}$$

The permeation current is obtained by solving Eq. (5.4c) with the boundary conditions given above

$$v_p(x) = \int_0^\infty dk A(k) \tanh(kH) \frac{k^2/\Delta}{1+\delta^2 k^2} \times \left[\cos(kx) - \frac{\cos k}{\cosh \delta^{-1}} \cosh(x/\delta)\right]. \tag{5.7}$$

Upon substituting these relations into Eq. (5.4b), we obtain a relation for the Fourier transform $A(k)$:

$$1 = \int_0^\infty A(k)\tanh(kH) \left\{ \left[\frac{\Lambda k}{\tanh(kH)} + k^2 + \frac{k^2/\Delta}{1+\delta^2 k^2} \right] \cos(kx) - \frac{k^2/\Delta}{1+\delta^2 k^2} \frac{\cos k}{\cosh \delta^{-1}} \cosh(x/\delta) \right\}. \tag{5.8}$$

The boundary condition $v_0(x)=0$ for $|x|>1$ imposes another constraint on the $A(k)$:

$$\int_0^\infty dk \; A(k)\tanh(kH)\cos kx = 0 \quad \text{for} \quad |x|>1. \tag{5.9}$$

This can be satisfied identically by expanding $A(k)$ in terms of Bessel functions

$$A(k)\tanh(kH) = k^{1/2-\beta} \sum_{m=0}^{\infty} a_m J_{2m-1/2+\beta}(k) \tag{5.10}$$

where β can be chosen for convenience of computation. If this form is substituted into Eq. (5.8), the x dependence can be integrated out by multiplying with $(1-x^2)^{\beta-1}\mathcal{P}_n(\beta-1/2, 1/2; x^2)$, where $\mathcal{P}_n$ are the Jacobi polynomials, to yield an infinite set of linear equations for the coefficients a_m:

$$\sum_{m=0}^{\infty} a_m G_{mn}^{\beta}(\Delta, \delta, \Lambda, H) = \frac{\delta_{n0}}{2^{\beta-1/2}\Gamma(\beta+1/2)}, \quad n=0,1,2,\ldots. \tag{5.11a}$$

Here, the coefficients are given as integrals in k space

$$G^{\beta}_{mn}(\Delta,\delta,\Lambda,H)=\int_0^{\infty}dk\ \ k^{1/2-\beta}J_{2m-1/2+\beta}(k)$$

$$\times[k^{1/2-\beta}G(k;\Delta,\delta,\Lambda,H)J_{2n-1/2+\beta}(k)$$

$$-(-1)^n\delta^{\beta-1/2}L(k;\Delta,\delta)I_{2n-1/2+\beta}(\delta^{-1})]. \tag{5.11b}$$

where I_{ν} is the modified Bessel function of order ν. The ‘‘kernels’’ for the smectic case,

$$G(k;\Delta,\delta,\Lambda,H)=k^2+\frac{k^2/\Delta}{\delta^2k^2+1}+\frac{\Lambda k}{\tanh(kH)}, \tag{5.11c}$$

$$L(k;\Delta,\delta)=\frac{k^2/\Delta}{1+\delta^2k^2}\,\frac{\cos k}{\cosh\delta^{-1}}, \tag{5.11d}$$

embody the properties of the smectic film as well as the importance of the fluid subphase and the depth of the subphase. Equations (5.11a) can serve as a starting point for numerical studies. For a structureless fluid film similar formulars have been derived [12]. The expressions differ from that for a structureless fluid by the absence of $L(k;\Delta,\delta)$ and the term $k^2\Delta^{-1}/(1+\delta^2k^2)$ in kernel $G(k;\Delta,\delta,\Lambda,H)$. This term reflects the correction to ηq_x^2 we found in Sec. III. As discussed there, the correction is small compared to the normal term k^2 for $\Delta\gg1$, but can grow at low temperatures. For wave numbers $k\ll\delta^{-1}$, this correction simply appears as an enhancement of the effective viscosity. However, for $k\gg\delta^{-1}$, the correction gives rise to a qualitative change in the velocity profile, characteristic of the permeation regime.

When the subphase drag on the film is large ($\Lambda\gg1$), the coefficients G^{β}_{mn} are in leading order proportional to Λ. This leads to equations identical to the ones for a normal fluid film on a sublayer. Further simplifications occur if one considers the deep channel limit, $H\rightarrow\infty$, which can be solved analytically by choosing $\beta=3/2$. One finds a semielliptical flow profile [12]

$$v_0(x)=\frac{1}{2\Lambda}\sqrt{1-x^2}. \tag{5.12}$$

On the other hand, for a shallow channel, $H\rightarrow0$, an analytical sultion can be obtained by the choice $\beta=1$ which results in plug flow [12]

$$v_0(x)=\frac{H}{\Lambda} \tag{5.13}$$

except for the boundary layer. Let us mention again that these two limiting cases also arise for a normal fluid film and merely reflect that the flow is dominated by the sublayer, i.e., the specific properties of the smectic film do not manifest themselves to leading order in Λ.

VI. SUMMARY

We have studied the hydrodynamics of two-dimensional smectics incorporating dislocations [8] in the context of shear flow across the layers. The behavior resembles that of a nematic for length scales beyond the dislocation correlation length, with an effective viscosity that represents the role of dislocation motion in making shear possible. At smaller length scales, the permeation mode of smectics determines the shear response. These different regimes can be observed in channel flow under a pressure head where the channel width sets the observation length scale, provided the drag due to the subphase can be neglected. At small dislocation densities, the permeation mode determines the flow profile, which evolves from a parabolic profile for channels narrower than the permeation length to a plug-flow shape as the channel becomes much wider. In the latter case (the permeation regime), shear is supported only in a boundary layer of thickness equal to the permeation length, hence the effective viscosity as determined by the net flow rate across the channel grows as the square of the channel width. On the other hand, for large dislocation densities, the flow profile is again parabolic, but with the viscosity modified by the dislocation density.

The dislocation density in turn depends on the shear rate through the shear strain supported by the layers in steady state. Under this strain, dislocation pairs in the smectic unbind at a lower energy cost, increasing their equilibrium density and helping to further relax the imposed strain, resulting in a shear thinning effect. This effect has been calculated for a 2D crystal of point particles by Bruinsma *et al.* [5], and could also be present in the hexatic phase, mediated by disclinations rather than dislocations. The flow results in steady state strains in the bond-orientation order parameter, which are relaxed by disclination motion. It would be interesting to explore this mechanism for shear thinning for the smectic films discussed here.

When the film flows on the surface of a fluid subphase, and drag from the subphase must be taken into account, the flow profile depends on the relative viscosities of the film and the subphase as well as on the channel width and the subphase depth. The analysis by Stone [12], which is supported by experiments, predicts the evolution of the parabolic profile into a semielliptical or plug-flow profile, depending on whether the drag is due to the subphase viscosity or a shallow subphase. In the Introduction, we showed that the same results apply to a hexatic film if described by an effective viscosity incorporating the coupling to the bond-orientation order parameter. In the situations described above where the subphase drag dominates the flow, these results are also applicable to smectic fillms, since the modification to the ‘‘flow kernel’’ of a smectic film with respect to an isotropic film is decoupled from the terms describing the influence of the subphase. The subphase drag manifests itself at long length scales where internal order in the film is unimportant.

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[1] C. Mingotaud, B. Agricole, and C. Jego, J. Phys. Chem. **99**, 17 068 (1995).

[2] T. Maruyama, G. Fuller, C. Frank, and C. Robertson, Science **274**, 233 (1996).

[3] M. L. Kurnaz and D. K. Schwartz, Phys. Rev. E **56**, 3378 (1997).

[4] R. S. Ghaskadvi, J. B. Ketterson, and P. Dutta, Langmuir **13**, 5137 (1997).

[5] R. Bruinsma, B. I. Halperin, and A. Zippelius, Phys. Rev. B **25**, 579 (1982).

[6] For analogous phenomena in superfluid helium films, see V. Ambegaokar, B. I. Halperin, D. R. Nelson, and E. D. Siggia, Phys. Rev. Lett. **40**, 783 (1978); Phys. Rev. B **21**, 1806 (1980).

[7] D. K. Schwartz (private communication).

[8] J. Toner and D. R. Nelson, Phys. Rev. B **23**, 316 (1981).

[9] F. Jähnig and F. Brochard, J. Phys. (France) **35**, 301 (1974); F. Brochard, J. Phys. (Paris), Colloq. **37**, C3-85 (1976).

[10] P. C. Hohenberg, B. I. Halperin, and D. R. Nelson, Phys. Rev. B **22**, 2373 (1980).

[11] D. K. Schwartz, C. M. Knobler, and R. Bruinsma, Phys. Rev. Lett. **73**, 2841 (1994).

[12] H. A. Stone, Phys. Fluids **7**, 2931 (1995).

[13] A. Zippelius, B. I. Halperin, and D. R. Nelson, Phys. Rev. B **22**, 2514 (1980).

[14] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, 2nd ed. (Oxford University Press, Oxford, 1993).

[15] P. G. de Gennes, Solid State Commun. **10**, 753 (1972).

[16] D. R. Nelson and R. A. Pelcovits, Phys. Rev. B **16**, 2191 (1977).

[17] H. T. Chiang, V. S. Chen-White, R. Pindak, and M. Seul, J. Phys. II **5**, 835 (1995).