

Dynamics of tilted hexatic phases in liquid-crystal films Jonathan Selinger

▶ To cite this version:

Jonathan Selinger. Dynamics of tilted hexatic phases in liquid-crystal films. Journal de Physique II, 1991, 1 (11), pp.1363-1373. 10.1051/jp2:1991145. jpa-00247597

HAL Id: jpa-00247597 https://hal.science/jpa-00247597

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers. L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Dynamics of tilted hexatic phases in liquid-crystal films

Jonathan V. Selinger (*)

(*) Department of Physics, Harvard University, Cambridge, MA 02138, U.S.A.

(Received 16 November 1990, revised 29 July 1991, accepted 1 August 1991)

Abstract. — We develop a theory for the dynamics of tilted hexatic phases in liquid-crystal films. A renormalization-group analysis gives exponents for the softening of the optical-mode relaxation rate near the hexatic-to-hexatic transitions. These exponents may be observable in experiments on thermotropic and lyotropic liquid crystals.

1. Introduction.

Liquid-crystal films often exhibit tilted hexatic phases, with at least quasi-long-range order in bond and tilt directions but only short-range crystalline order. Tilted hexatic phases can differ from each other in the relation between the local bond and tilt directions. Two recent experiments have studied transitions among different tilted hexatic phases. Dierker and Pindak [1] use light scattering to investigate thin tilted hexatic films of thermotropic liquid crystals. They observe a weak first-order transition from the hexatic-I phase, in which the local tilt (azimuthal) angle is locked along one of the six local bonds, to the hexatic-F phase, in which the local tilt angle is locked halfway between two local bonds, 30° from each. Smith *et al.* [2] use x-ray scattering to examine the $L_{\beta'}$ phases of lyotropic liquid crystals, which are probably hexatic but may have finite in-plane crystallites. They find three distinct $L_{\beta'}$ phases : the $L_{\beta I}$ and $L_{\beta F}$ phases (analogous to hexatic-I and hexatic-F) and a new intermediate $L_{\beta L}$ phase, in which the local tilt is locked at an angle between 0° and 30° away from a local bond. The $L_{\beta I} - L_{\beta L}$ and $L_{\beta L} - L_{\beta F}$ transitions are second-order.

In earlier papers [3, 4] we presented a Landau theory with fluctuation corrections for transitions among tilted hexatic phases in two-dimensional (2D) liquid-crystal films. Our theory is based on the interaction potential

$$V(\theta - \phi) = -h_6 \cos 6(\theta - \phi) - h_{12} \cos 12(\theta - \phi)$$
(1.1)

between the bond-angle field θ and the tilt-azimuthal-angle field ϕ . Using the renormalization group, we derive a phase diagram in the three parameters h_6 , h_{12} , and K_- , the stiffness constant for the $\theta_- \equiv \theta - \phi$ mode. This phase diagram exhibits the hexatic-I, -F, and -L

^(*) Current address: Department of Physics, University of California, Los Angeles, CA 90024, U.S.A.

JOURNAL DE PHYSIQUE II - T 1 N H, NOVEMBRE 1991

phases as well as an unlocked tilted hexatic phase, in which the bond and tilt angles fluctuate independently. Two cross sections of the phase diagram are shown in figure 1.

The purpose of this paper is to extend the Landau theory to describe the dynamics of tilted hexatic films near the hexatic-to-hexatic transitions. This paper complements the work of Zippelius *et al.* [5] and Ostlund *et al.* [6], who have studied the dynamics of 2D systems near the liquid-hexatic and hexatic-solid transitions, and Pleiner and Brand [7], who have studied the dynamics of 3D stacked hexatic liquid crystals. In section 2 we derive a kinetic equation for the $\theta_{-}(\mathbf{r}, t)$ mode using the potential (1.1). This kinetic equation can be solved easily if one approximates (1.1) by a harmonic potential. We show that the harmonic approximation is self-consistent if and only if the restoring force at the minimum of the potential (a simple function of h_6 and h_{12}) is sufficiently large. This condition is satisfied in most of the phase diagram, but it is not satisfied near the second-order transitions, and it may not be satisfied near the first-order I-F transition.

In section 3 we use a dynamic renormalization-group technique (similar to the dynamic renormalization group for roughening [8, 9]) to find the behavior when the harmonic approximation breaks down. By integrating out the short-wavelength components of the noise and rescaling distances and times, we transform all the parameters of the kinetic equation. We iterate until h_6 and h_{12} either become large, in which case we can make a self-consistent approximation, or else go to zero, in which case the problem is trivial. Using this



Fig. 1. — Two cross sections of the theoretical phase diagram, for (a) constant $h_{12} > 0$ and (b) constant $h_{12} < 0$, as a function of h_6 and the temperature-like variable K_{-}^{-1} . The double lines represent first-order and the single lines second-order transitions. The arrows labeled (1)-(4) indicate the transitions discussed in section 3.

technique, we derive the relaxation rate of the $\theta_{-}(\mathbf{r}, t)$ mode near each of the hexatic-tohexatic transitions in figure 1. Our results are consistent with dynamic measurements near the first-order I-F transition in thermotropics [1], and they could be tested by dynamic measurements near the second-order I-L and L-F transitions in lyotropics.

2. Model of dynamics.

We consider the Hamiltonian for a 2D tilted hexatic film [3, 4, 10]

$$\frac{H}{k_{\rm B}T} = \int d^2\mathbf{r} \left[\frac{1}{2} K_6 |\nabla\theta|^2 + \frac{1}{2} K_1 |\nabla\phi|^2 + g\nabla\theta \cdot \nabla\phi + V (\theta - \phi) \right].$$
(2.1)

In the first three terms, K_6 is a Frank constant for variations in the bond orientation $\theta(\mathbf{r}, t)$, K_1 is a stiffness constant for variations in the tilt azimuthal angle $\phi(\mathbf{r}, t)$, and g is a gradient cross-coupling. These 2D elastic constants are implicitly integrals of 3D elastic constants across the thickness of the film. We neglect elastic anisotropy, which is irrelevant at long length scales [3, 4, 11]. The function $V(\theta - \phi)$ is a general tilt-bond interaction potential, which can be expressed in general as the Fourier series

$$V(\theta - \phi) = -\sum_{n=1}^{\infty} h_{6n} \cos 6 n(\theta - \phi)$$
(2.2)

because of the local hexagonal symmetry. The first two terms of the series dominate near the hexatic-to-hexatic transitions, and a qualitatively correct phase diagram may be obtained by truncating the series to obtain the potential (1.1) [3, 4].

In this paper, we consider only tilted hexatic phases, in which disclinations in $\theta(\mathbf{r}, t)$ and vortices in $\phi(\mathbf{r}, t)$ are rare and are irrelevant to the statistical mechanics. Nelson and Halperin [10] have shown that these defects are irrelevant when the renormalized $K_6 > 72/\pi$ and the renormalized $K_1 > 2/\pi$. Under these circumstances, we can treat both $\theta(\mathbf{r}, t)$ and $\phi(\mathbf{r}, t)$ as single-valued functions. We can then simplify the Hamiltonian by defining the linear combinations [10]

$$\theta_{+}(\mathbf{r},t) = \alpha \theta(\mathbf{r},t) + \beta \phi(\mathbf{r},t), \qquad (2.3a)$$

$$\theta_{-}(\mathbf{r},t) = \theta(\mathbf{r},t) - \phi(\mathbf{r},t), \qquad (2.3b)$$

where $\alpha = 1 - \beta = (K_6 + g)/(K_6 + K_1 + 2g)$. In terms of $\theta_{\pm}(\mathbf{r}, t)$, the Hamiltonian becomes

$$\frac{H}{k_{\rm B}T} = \int d^2 \mathbf{r} \left[\frac{1}{2} K_+ |\nabla \theta_+|^2 + \frac{1}{2} K_- |\nabla \theta_-|^2 + V(\theta_-) \right], \qquad (2.4)$$

with $K_+ = K_6 + K_1 + 2g$ and $K_- = (K_1 K_6 - g^2)/K_+$. The average value of θ_- is 0° (mod 60°) in the I phase, 30° (mod 60°) in the F phase, and between 0° and \pm 30° (mod 60°) in the L phase. By analogy with the terminology for phonons, variations in θ_+ (r, t) and θ_- (r, t) can be called « acoustic » and « optical » modes, respectively, with in-phase and out-of-phase variations of bond and tilt angles [1].

In references [3, 4], the phase transitions of this model were studied by first using meanfield theory and then examining the effects of fluctuations using the renormalization group. We suppose that h_{12} is fixed and h_6 decreases from positive to negative values as a function of temperature or humidity. In mean-field theory, the nature of the phase transitions depends on the sign of h_{12} . In a material with $h_{12} > 0$, there is a first-order transition from I to F at $h_6 = 0$. In a material with $h_{12} < 0$, there is a second-order, Ising-type transition from I to L at $h_6 = 4 |h_{12}|$ and another second-order, Ising-type transition from L to F at $h_6 = -4 |h_{12}|$. When we analyze the effects of fluctuations using the renormalization group, we find that mean-field theory is valid for $K_- > K_{c,12}$, where h_6 and h_{12} are both relevant parameters, becoming large as the renormalization-group transformation is iterated. However, for $K_{c,6} < K_- < K_{c,12}$, the parameter h_{12} is irrelevant, and there is a second-order transition from I to F at $h_6 = 0$. For $K_- < K_{c,6}$, the parameters h_6 and h_{12} are both irrelevant, and there is an unlocked phase. The critical initial stiffnesses $K_{c,6}$ and $K_{c,12}$ depend on the initial values of h_6 and h_{12} ; they are approximately $K_{c,6} \approx 9/(2\pi)$ and $K_{c,12} \approx 18/\pi$. The corresponding renormalized stiffnesses are exactly $9/(2\pi)$ and $18/\pi$. Figure 1 shows the phase diagram derived using the renormalization group.

We now investigate the dynamic behavior of a tilted hexatic film. Following references [12, 8], we assume that the low-wave-vector, low-frequency dynamics can be described by the simplest kinetic equations that relax to the correct static limit. In this problem there are no conservation laws for θ_+ or θ_- However, there is a reversible coupling between the θ_+ mode and the transverse part of the momentum density, $g_T(\mathbf{r}, t)$. The origin of this coupling is that a local vorticity field causes θ_+ to precess, and conversely, an inhomogeneity in θ_+ causes the fluid to move [5]. There is no such coupling between θ_- and g_T because the two angles θ and ϕ precess together in response to a local vorticity field, and hence the difference $\theta_- = \theta - \phi$ does not change. As a result, the kinetic equations for θ_+ and g_T can be written as [5]

$$\frac{\partial \theta_{+}(\mathbf{r},t)}{\partial t} = \Gamma_{+} K_{+} \nabla^{2} \theta_{+}(\mathbf{r},t) + \frac{1}{2\rho_{0}} \hat{\mathbf{z}} \cdot \nabla \times \mathbf{g}_{\mathrm{T}}(\mathbf{r},t) + \Gamma_{+} f_{+}(\mathbf{r},t) + \zeta_{+}(\mathbf{r},t), \quad (2.5a)$$

$$\frac{\partial \mathbf{g}_{\mathrm{T}}(\mathbf{r},t)}{\partial t} = -\frac{1}{2} K_{+} k_{\mathrm{B}} T(\hat{\mathbf{z}} \times \nabla) \nabla^{2} \theta_{+}(\mathbf{r},t) + \nu \nabla^{2} \mathbf{g}_{\mathrm{T}}(\mathbf{r},t) + \nu \rho_{0} \nabla^{2} \mathbf{f}_{\mathrm{T}}(\mathbf{r},t) + \zeta_{\mathrm{T}}(\mathbf{r},t), \quad (2.5b)$$

and the kinetic equation for θ_{-} becomes

$$\frac{\partial \theta_{-}(\mathbf{r},t)}{\partial t} = \Gamma_{-} K_{-} \nabla^{2} \theta_{-}(\mathbf{r},t) - \Gamma_{-} V'(\theta_{-}(\mathbf{r},t)) + \Gamma_{-} f_{-}(\mathbf{r},t) + \zeta_{-}(\mathbf{r},t). \quad (2.6)$$

In these equations Γ_{\pm} are kinetic coefficients [13], ν is the kinematic viscosity of the fluid, ρ_0 is the equilibrium mass density, and $f_{\pm}(\mathbf{r}, t)$ and $\mathbf{f}_{\mathrm{T}}(\mathbf{r}, t)$ are external forces that couple to $\theta_{\pm}(\mathbf{r}, t)$ and $\mathbf{g}_{\mathrm{T}}(\mathbf{r}, t)$. The functions $\zeta_{\pm}(\mathbf{r}, t)$ and $\zeta_{\mathrm{T}}(\mathbf{r}, t)$ are independent Gaussian noise sources satisfying

$$\langle \zeta_{+}(\mathbf{q},t) \rangle = \langle \zeta_{-}(\mathbf{q},t) \rangle = \langle \zeta_{T}(\mathbf{q},t) \rangle = 0,$$
 (2.7a)

$$\langle \zeta_{+}(\mathbf{q},t) \zeta_{+}(\mathbf{q}',t') \rangle = 2 \Gamma_{+} (2 \pi)^{2} \delta(\mathbf{q}+\mathbf{q}') \delta(t-t'),$$
 (2.7b)

$$\left\langle \zeta_{-}(\mathbf{q},t) \zeta_{-}(\mathbf{q}',t') \right\rangle = \begin{cases} 2 \Gamma_{-}(2 \pi)^{2} \delta(\mathbf{q}+\mathbf{q}') \delta(t-t'), & \text{if } |\mathbf{q}| < \Lambda, \\ 0, & \text{otherwise}, \end{cases}$$
(2.7c)

$$\langle \zeta_{T_{i}}(\mathbf{q}, t) \zeta_{T_{j}}(\mathbf{q}', t') \rangle = 2 \nu \rho_{0} k_{B} T q^{2} \left(\delta_{ij} - \frac{q_{i} q_{j}}{q^{2}} \right) (2 \pi)^{2} \delta(\mathbf{q} + \mathbf{q}') \delta(t - t').$$
 (2.7d)

The parameter Λ is an ultraviolet cutoff (of order the inverse intermolecular spacing), which we impose on the noise ζ_{-} , as in reference [9]. We could impose a similar ultraviolet cutoff on ζ_{+} and ζ_{T} , but that is not necessary. Note that the kinetic equations are invariant under the

transformation $\theta_+ \rightarrow -\theta_+$ and $\mathbf{g}_T \rightarrow -\mathbf{g}_T$, and independently under the transformation $\theta_- \rightarrow -\theta_-$.

The kinetic equations for the acoustic mode θ_+ and the transverse momentum density g_T have been solved by Zippelius *et al.* [5]. They find that the linear response function has poles at the eigenfrequencies

$$\omega_{1,2} = -\frac{1}{2} i D_{1,2} q^2, \qquad (2.8)$$

where

$$D_{1,2} = \Gamma_{+} K_{+} + \nu \pm \sqrt{(\Gamma_{+} K_{+} - \nu)^{2} - \frac{K_{+} k_{\rm B} T}{\rho_{0}}}$$
(2.9)

If the quantity under the square root is positive, the two eigenmodes are purely diffusive, with relaxation rates given by $\gamma_{1,2} = \frac{1}{2} D_{1,2} q^2$. If the quantity under the square root is negative, the two eigenmodes propagate but are heavily damped, with a relaxation rate of $\gamma = \frac{1}{2} (\Gamma_+ K_+ + \nu) q^2$. The scaling of the relaxation rate with q^2 is consistent with the experiments of Dierker and Pindak [1]. These eigenmodes and eigenfrequencies do not change as one passes through any of the hexatic-to-hexatic transitions.

The kinetic equation for the optical mode θ_{-} is nonlinear and hence cannot be solved exactly. For an approximate, mean-field solution, we assume the thermal fluctuations are small enough that $\theta_{-}(\mathbf{r}, t)$ always remains close to a particular minimum of $V(\theta_{-})$; we will later determine when this assumption is self-consistent. When this assumption is self-consistent, we can approximate $V(\theta_{-})$ by a harmonic potential about the appropriate minimum. The minima of $V(\theta_{-})$ are located at

$$\theta_{-}^{\min} = \begin{cases} 0^{\circ} \pmod{60^{\circ}}, & \text{if } h_{6} > \max (0, -4 h_{12}); \\ 30^{\circ} \pmod{60^{\circ}}, & \text{if } h_{6} < \min (0, 4 h_{12}); \\ \pm \frac{1}{6} \cos^{-1} (h_{6}/4 | h_{12} |) (\mod 60^{\circ}), & \text{if } h_{12} < 0 \text{ and} \\ -4 | h_{12} | < h_{6} < 4 | h_{12} |. \end{cases}$$
(2.10)

We therefore approximate

$$V(\theta_{-}(\mathbf{r},t)) \approx \frac{1}{2} V''(\theta_{-}^{\min})(\theta_{-}(\mathbf{r},t) - \theta_{-}^{\min})^{2}, \qquad (2.11)$$

where

$$V''(\theta_{-}^{\min}) = \begin{cases} 36 h_{6} + 144 h_{12}, & \text{if } h_{6} > \max(0, -4 h_{12}); \\ -36 h_{6} + 144 h_{12}, & \text{if } h_{6} < \min(0, 4 h_{12}); \\ (144 h_{12}^{2} - 9 h_{6}^{2})/|h_{12}|, & \text{if } h_{12} < 0 \text{ and} \\ -4|h_{12}| < h_{6} < 4|h_{12}|. \end{cases}$$
(2.12)

The kinetic equation (2.6) now becomes linear and can be solved easily. For any particular realization of the noise ζ_{-} , we find that the Fourier transform of $\delta \theta_{-} \equiv \theta_{-} - \theta_{-}^{\min}$ is

$$\delta \theta_{-}(\mathbf{q}, \omega) = G_{-}^{\mathrm{MF}}(\mathbf{q}, \omega) [f_{-}(\mathbf{q}, \omega) + \Gamma_{-}^{-1} \zeta_{-}(\mathbf{q}, \omega)], \qquad (2.13)$$

with the linear response function

$$G_{-}^{\rm MF}(\mathbf{q},\,\omega\,) = \left[-\,i\,\omega\,\Gamma_{-}^{-1} + K_{-}\,q^{2} + \,V''(\theta_{-}^{\rm min})\right]^{-1} \tag{2.14}$$

The imaginary pole of this response function shows that the optical-mode relaxation rate is

$$\gamma_{-}^{\rm MF}(\mathbf{q}) = \Gamma_{-} \left(K_{-} q^{2} + V''(\theta_{-}^{\rm min}) \right)$$
(2.15)

in the mean-field approximation.

Equations (2.15) and (2.12) give the relaxation rate near the first-order I-F transition in a material with $h_{12} > 0$. Near this transition, we can write $h_6 \propto (T - T_{IF})$, and we have

$$\gamma_{-}^{\text{MF}}(\mathbf{q}=0) = \Gamma_{-} (36|h_{6}| + 144|h_{12}). \qquad (2.16)$$

As $T \to T_{\rm IF}$, the relaxation rate $\gamma_{-}^{\rm MF}(q=0)$ decreases linearly toward a nonzero limit.

We must now determine when the mean-field, harmonic approximation is self-consistent. From equation (2.13), if the external force $f_{-} = 0$, the thermal fluctuations in θ_{-} are given by

$$\left\langle \left(\delta\theta_{-}(\mathbf{r},t)\right)^{2}\right\rangle = \frac{1}{4\pi K_{-}}\ln\left[1+\frac{K_{-}\Lambda^{2}}{V''(\theta_{-}^{\min})}\right].$$
(2.17)

The mean-field, harmonic approximation is valid if and only if the fluctuations remain well inside one of the parabolic wells of $V(\theta_{-})$. As a rough criterion, we require $\langle (6 \,\delta \theta_{-}(\mathbf{r}, t))^2 \rangle < 1/4 \,\pi$, and hence the approximation is valid for

$$V''(\theta_{-}^{\min}) > \frac{K_{-}\Lambda^{2}}{e^{K_{-}/36} - 1}$$
(2.18)

For $K_{-} \leq 36$, this criterion simplifies to

$$V''(\theta_{-}^{\min}) \ge 36 \Lambda^2 \tag{2.19}$$

This criterion is satisfied in most of the phase diagram, whenever h_6 is large. It may be satisfied near the first-order I-F transition if h_{12} is of order Λ^2 . It is definitely *not* satisfied near the second-order I-L and L-F transitions, where $V''(\theta_-^{\min})$ vanishes. Furthermore, it is not satisfied near the second-order I-F transition, where h_{12} is irrelevant, or in the unlocked phase, where h_6 and h_{12} are both irrelevant. To understand the dynamics in all the regimes where the criterion (2.19) is not satisfied, we must use the dynamic renormalization group of section 3.

3. Renormalization-group analysis.

In the regimes where we cannot solve the kinetic equation (2.6) directly, we can use the renormalization group to transform it into an equation that can be solved. In the renormalization-group calculation, we exploit an analogy between the dynamics of tilted hexatic phases and the dynamics of roughening, which has been investigated by Chui and Weeks [8] and Nozières and Gallet [9]. Equation (2.6) for $\theta_{-}(\mathbf{r}, t)$ is almost equivalent to the kinetic equation for the height of an interface. The tilt-bond interaction potential $V(\theta_{-})$ corresponds to the pinning potential that tends to fix the height of the interface at an integral number of lattice spacings. The only difference is that $V(\theta_{-})$ is the sum of two cosine terms, but the potential studied in references [8] and [9] is a single cosine term. In this section,

we generalize the dynamic renormalization group for roughening to treat the potential $V(\theta_{-})$, and then we use the renormalization group to study the dynamics of $\theta_{-}(\mathbf{r}, t)$ near each of the hexatic-to-hexatic transitions.

To derive the recursion relations, we follow the procedure of Nozières and Gallet. We first transform the differential equation (2.6) into the integral equation

$$\theta_{-}(\mathbf{r},t) = \int d^{2}\mathbf{r}_{1} dt_{1} G_{-}^{(0)}(\mathbf{r}-\mathbf{r}_{1},t-t_{1}) [f_{-}(\mathbf{r}_{1},t_{1}) + \Gamma_{-}^{-1}\zeta_{-}(\mathbf{r}_{1},t_{1}) - V'(\theta_{-}(\mathbf{r}_{1},t_{1}))],$$
(3.1)

where

$$G_{-}^{(0)}(\mathbf{q},\,\omega) = \left[-\,i\,\omega\,\Gamma_{-}^{-1} + K_{-}\,q^{2}\right]^{-1},$$
(3.2a)

$$G_{-}^{(0)}(\mathbf{r}, t) = \begin{cases} (1/4 \ \pi K_{-} \ t) \ e^{-r^{2}/4 \ \Gamma_{-} \ K_{-} \ t}, & \text{if } t > 0; \\ 0, & \text{otherwise}. \end{cases}$$
(3.2b)

By iterating this integral equation, we solve for $\theta_{-}(\mathbf{r}, t)$ to second order in h_6 and h_{12} . We then average over the Fourier components of the noise $\zeta_{-}(\mathbf{q}, t)$ in the range $e^{-\ell} \Lambda < |\mathbf{q}| < \Lambda$, thereby obtaining a « partially averaged » solution for θ_{-} in terms of the external force f_{-} and the unaveraged components of $\zeta_{-}(\mathbf{q}, t)$, with $|\mathbf{q}| < e^{-\ell} \Lambda$. Note that the averaging procedure does not eliminate any components of f_{-} or θ_{-} The partially averaged solution $\theta_{-}(\mathbf{r}, t)$ satisfies an integral equation similar to (3.1), and hence a differential equation similar to (2.6), to second order in h_6 and h_{12} . Using some approximations described by Nozières and Gallet, we put the new differential equation into a form equivalent to (2.6), but with different parameters in place of h_6 , h_{12} , K_{-} , and Γ_{-} As a final step, we rescale distances by a factor of e^{ℓ} and times by a factor of $e^{2\ell}$. We obtain the following differential recursion relations, valid to second order in h_6 and h_{12} .

$$\frac{dh_6(\ell)}{d\ell} = \left(2 - \frac{9}{\pi K_-(\ell)}\right) h_6(\ell) + \frac{c_1 h_6(\ell) h_{12}(\ell)}{\Lambda^2}, \qquad (3.3a)$$

$$\frac{dh_{12}(\ell)}{d\ell} = \left(2 - \frac{36}{\pi K_{-}(\ell)}\right) h_{12}(\ell) + \frac{c_2 h_6(\ell)^2}{\Lambda^2}, \qquad (3.3b)$$

$$\frac{\mathrm{d}K_{-}(\ell)}{\mathrm{d}\ell} = \frac{c_{3}h_{6}(\ell)^{2} + c_{4}h_{12}(\ell)^{2}}{\Lambda^{4}}, \qquad (3.3c)$$

$$\frac{\mathrm{d}\Gamma_{-}(\ell)}{\mathrm{d}\ell} = \frac{[c_{5}h_{6}(\ell)^{2} + c_{6}h_{12}(\ell)^{2}]\Gamma_{-}(\ell)}{\Lambda^{4}}$$
(3.3d)

The c coefficients are dimensionless functions of K_{-} that can be calculated explicitly by the method of Nozières and Gallet. We do not need their exact expressions for the purposes of this paper.

In the renormalization-group transformation, we change the kinetic equation (2.6) into an equation with the same form but with different parameters. It is straightforward to relate the linear response function G_{-} for the original equation to the linear response function for the transformed equation. Because distances and times are rescaled but the field $\theta_{-}(\mathbf{r}, t)$ is not rescaled, we obtain the generalized homogeneity relation,

$$G_{-}(\mathbf{q},\omega;h_{6},h_{12},K_{-},\Gamma_{-}) = e^{2\ell}G_{-}(e^{\ell}\mathbf{q},e^{2\ell}\omega;h_{6}(\ell),h_{12}(\ell),K_{-}(\ell),\Gamma_{-}(\ell)). \quad (3.4)$$

The static susceptibility $\chi_{-}(\mathbf{q})$ defined by

$$\left\langle \theta_{-}(\mathbf{q},t) \; \theta_{-}^{\cdot}(\mathbf{q}',t) \right\rangle = \chi_{-}(\mathbf{q})(2 \; \pi)^{2} \, \delta\left(\mathbf{q}+\mathbf{q}'\right) \tag{3.5}$$

satisfies a similar relation,

$$\chi_{-}(\mathbf{q}; h_{6}, h_{12}, K_{-}) = e^{2\ell} \chi_{-}(e^{\ell} \mathbf{q}; h_{6}(\ell), h_{12}(\ell), K_{-}(\ell)).$$
(3.6)

The recursion relations (3.3) show that h_6 is relevant for $K_- > K_{c,6} \approx 9/(2\pi)$ and h_{12} is relevant for $K_- > K_{c,12} \approx 18/\pi$. If h_6 and h_{12} are both irrelevant then we can iterate the recursion relations for $\ell \to \infty$. In this case h_6 and h_{12} are both driven to 0, and the problem of determining G_- or χ_- becomes trivial. On the other hand, if either h_6 or h_{12} is relevant, then we must stop iterating when $\ell = \ell^*$ such that max $(|h_6(\ell^*)|, |h_{12}(\ell^*)|) = O(\Lambda^2)$ [14]. At that point the recursion relations (derived for small h_6 and h_{12}) cease to be valid, and we must match onto some approximation for G_- or χ_- that is valid for large h_6 or h_{12} . We consider the dynamic behavior in four regimes, which are indicated by the arrows in figure 1.

(1) If $K_{-} < K_{c,6}$, then h_6 and h_{12} are both irrelevant. The system is in the unlocked phase, with independent fluctuations of bond and tilt angles. The linear response function is given by equation (3.4); the right-hand side can be evaluated for $\ell \to \infty$ using equation (3.2a). We obtain

$$G_{-}(\mathbf{q},\omega) = [-i\omega\Gamma_{-}^{R-1} + K_{-}^{R}q^{2}]^{-1}, \qquad (3.7)$$

where the renormalized coefficients Γ_{-}^{R} and K_{-}^{R} are defined by

$$\Gamma_{-}^{\mathbf{R}} = \lim_{\ell \to \infty} \Gamma_{-}(\ell) , \qquad (3.8a)$$

$$K_{-}^{\mathsf{R}} = \lim_{\ell \to \infty} K_{-}(\ell) .$$
(3.8b)

The argument of references [8, 9], in the context of roughening, shows that these limits are finite. The optical-mode relaxation rate is therefore

$$\gamma_{-}(\mathbf{q}) = \Gamma_{-}^{\mathbf{R}} K_{-}^{\mathbf{R}} q^{2}$$
(3.9)

By a similar argument, the static susceptibility is

$$\chi_{-}(\mathbf{q}) = [K_{-}^{\mathbf{R}} q^{2}]^{-1}$$
(3.10)

At the critical stiffness $K_{c,6}$, the coupling h_6 becomes relevant, and there is a «lock-in» transition from the unlocked phase to the I phase (if $h_6 > 0$) or the F phase (if $h_6 < 0$). This transition is exactly analogous to the transition from the rough phase to the smooth phase in the theory of roughening, and the results of references [8] and [9] can be carried over directly. In particular, both $K_{\rm r}^{\rm R}$ and $\Gamma_{\rm r}^{\rm R}$ have square-root cusps at the transition.

(2) If $K_{c,6} < K_{-} < K_{c,12}$, then h_6 is relevant but h_{12} is irrelevant. The system has a secondorder I-F transition at $h_6 = 0$. Near the transition, we can write $h_6 \propto (T - T_{\rm IF})$. We use the recursion relations (3.3) to iterate up to the length scale ℓ^* where $|h_6(\ell^*)| = O(\Lambda^2)$. A characteristic value for $K_-(\ell)$ during the iteration is K_-^R , which is between $9/(2\pi)$ and $18/\pi$. The flow of $h_6(\ell)$ is approximately

$$h_6(\ell) = h_6 e^{(2 - 9/\pi K_-^{\kappa})\ell}, \qquad (3.11)$$

and l^* is therefore given by

$$e^{\ell^*} = \left(\frac{\Lambda^2}{|h_6|}\right)^{\pi K_-^{\mathbb{R}}/(2\pi K_-^{\mathbb{R}}-9)}$$
(3.12)

By integrating the recursion relations for $K_{-}(\ell)$ and $\Gamma_{-}(\ell)$ from $\ell = 0$ to ℓ^* , we obtain

$$K_{-}^{\mathrm{R}} - K_{-} = \frac{c_{3}}{2} \left(\frac{\pi K_{-}^{\mathrm{R}}}{2 \pi K_{-}^{\mathrm{R}} - 9} \right) \left(1 - \frac{h_{6}^{2}}{\Lambda^{4}} \right), \qquad (3.13a)$$

$$\ln \frac{\Gamma_{-}^{R}}{\Gamma_{-}} = \frac{c_{5}}{2} \left(\frac{\pi K_{-}^{R}}{2 \pi K_{-}^{R} - 9} \right) \left(1 - \frac{h_{6}^{2}}{\Lambda^{4}} \right).$$
(3.13b)

Note that K_{-}^{R} and Γ_{-}^{R} do not have singularities as $h_{6} \rightarrow 0$.

At the length scale l^* , the criterion (2.19) is satisfied, and hence we can use the mean-field theory of section 2 to evaluate the right-hand side of equation (3.4). The linear response function is therefore

$$G_{-}(\mathbf{q},\omega) = \left[-i\omega\Gamma_{-}^{R-1} + K_{-}^{R}q^{2} + 36\Lambda^{2}\left(\frac{|h_{6}|}{\Lambda^{2}}\right)^{2\pi K_{-}^{R}/(2\pi K_{-}^{R}-9)}\right]^{-1}, \quad (3.14)$$

and the optical-mode relaxation rate is

$$\gamma_{-}(\mathbf{q}) = \Gamma_{-}^{\mathbf{R}} \left[K_{-}^{\mathbf{R}} q^{2} + 36 \Lambda^{2} \left(\frac{|h_{6}|}{\Lambda^{2}} \right)^{2 \pi K_{-}^{\mathbf{R}}/(2 \pi K_{-}^{\mathbf{R}} - 9)} \right].$$
(3.15)

Similarly, the static susceptibility is

$$\chi_{-}(\mathbf{q}) = \left[K_{-}^{\mathbf{R}} q^{2} + 36 \Lambda^{2} \left(\frac{|h_{6}|}{\Lambda^{2}} \right)^{2 \pi K_{-}^{\mathbf{R}}/(2 \pi K_{-}^{\mathbf{R}} - 9)} \right]^{-1}, \qquad (3.16)$$

and the correlation length ξ diverges as

$$\xi \propto |h_6|^{-\pi K_-^R/(2\pi K_-^R - 9)}$$
(3.17)

These results can be summarized by the critical exponents

$$\gamma = 2 \nu = \frac{2 \pi K_{-}^{R}}{2 \pi K_{-}^{R} - 9}, \qquad (3.18a)$$

$$\eta = 0, \qquad (3.18b)$$

$$z = 2$$
. (3.18c)

The results could be tested by experiments on liquid-crystal films with the appropriate stiffness K_{-} . Note that $K_{c, 12}$ is about a factor of 5 less than the stiffness of the five-layer films studied by Dierker and Pindak [1].

(3) If $K_- > K_{c, 12}$ and $h_{12} > 0$, then h_6 and h_{12} are both relevant, and the system has a first-order I-F transition at $h_6 = 0$. Close to the transition, $h_6 \propto (T - T_{IF})$ is small. If the initial value of h_{12} is of order Λ^2 , we can use the mean-field theory of section 2 directly; otherwise,

we must use the recursion relations (3.3) to iterate up to the length scale ℓ^* where $h_{12}(\ell^*) = O(\Lambda^2)$. Near the tricritical point, as $K_- \to K_{c,12}^+$, we obtain

$$\ell^* \propto (K_- - K_{c,12})^{-1/2}$$
 (3.19)

by the method of Kosterlitz [15]. At the length scale l^* , we can use mean-field theory to evaluate the right-hand sides of equations (3.4) and (3.6), and hence we obtain

$$G_{-}(\mathbf{q},\omega) = \left[-i\omega\Gamma_{-}^{R-1} + K_{-}^{R}q^{2} + 36\left|h_{6}\right| e^{-c(K_{-}-K_{c,12})^{-1/2}} + 144\Lambda^{2}e^{-4c(K_{-}-K_{c,12})^{-1/2}}\right]^{-1}$$
(3.20)

and

$$\chi_{-}(\mathbf{q}) = \left[K_{-}^{R} q^{2} + 36 \left|h_{6}\right| e^{-c(K_{-} - K_{c,12})^{-1/2}} + 144 \Lambda^{2} e^{-4c(K_{-} - K_{c,12})^{-1/2}}\right]^{-1}$$
(3.21)

The optical-mode relaxation rate is therefore

$$\gamma_{-}(\mathbf{q}) = \Gamma_{-}^{R} [K_{-}^{R} q^{2} + 36 | h_{6} | e^{-c(K_{-} - K_{c,12})^{-1/2}} + 144 \Lambda^{2} e^{-4c(K_{-} - K_{c,12})^{-1/2}}]. \quad (3.22)$$

Note that $\gamma_{-}(\mathbf{q}=0)$ decreases linearly with $|h_6| \propto |T - T_{\rm IF}|$, and it has a nonzero minimum value at the transition. This minimum value vanishes with an essential singularity at the tricritical point, where $K_{-} \rightarrow K_{c,12}^{+}$. Note also that $\gamma_{-}(\mathbf{q})$ has the conventional quadratic dependence on \mathbf{q} (contrary to Eqs. (5.3) of Ref. [4], which are incorrect because h_6 and h_{12} are evaluated at an incorrect length scale). The behavior of $\gamma_{-}(\mathbf{q})$ as a function of $(T - T_{\rm IF})$ and as a function of \mathbf{q} is consistent with the experiments on thermotropic films by Dierker and Pindak [1].

(4) If $K_- > K_{c,12}$ and $h_{12} < 0$, then h_6 and h_{12} are both relevant, and the system has secondorder I-L and L-F transitions. We can use the recursion relations (3.3) to iterate up to a length scale ℓ^* where $h_6(\ell^*)$ and $h_{12}(\ell^*)$ are large. However, we cannot use mean-field theory close to the I-L and L-F transitions, because $V_{\ell^*}(\theta_-^{\min}) = 0$ there. Instead, we must use an analogy with the Ising model. The renormalized potential $V_{\ell^*}(\theta_-)$ consists of a series of valleys with Ising-type internal structure. If $h_6(\ell^*)$ and $h_{12}(\ell^*)$ are of order Λ^2 , then fluctuations in θ_- are confined to one valley, and our problem becomes equivalent to a 2D continuum Ising model with fluctuations. Because there are no conservation laws, the dynamic universality class is model A of Hohenberg and Halperin [12]. From dynamic scaling, we have

$$\boldsymbol{\gamma}_{-}(\mathbf{q}) = \boldsymbol{\xi}^{-z} \, \boldsymbol{\Omega}\left(\boldsymbol{\xi} \, | \, \mathbf{q} \, | \, \right) \tag{3.23}$$

near the second-order transitions. The relaxation rate $\gamma_{-}(\mathbf{q}=0)$ therefore vanishes as

$$\gamma_{-} (\mathbf{q} = 0) \propto \xi^{-} \propto \begin{cases} |h_6 - h_6^{\mathrm{IL}}|^{\nu_2}, & \text{near the I-L transition}; \\ |h_6 - h_6^{\mathrm{LF}}|^{\nu_2}, & \text{near the L-F transition}. \end{cases}$$
(3.24)

For the 2D Ising model, $\nu = 1$ exactly and $z \approx 2.18$ in the ε -expansion [12]. In the lyotropic films studied by Smith *et al.* [2], h_6 can be adjusted by varying either the chemical potential of water (μ) or the temperature. Near the I-L transition, $(h_6 - h_6^{\rm IL}) \propto (\mu - \mu_{\rm IL})$ or $(T - T_{\rm IL})$; a similar result holds near the L-F transition. The power law (3.24) could therefore be tested by dynamic measurements near the I-L and L-F transitions in lyotropic films.

Acknowledgments.

I am grateful to D. R. Nelson for many helpful discussions about this work, and to S. B. Dierker, R. Pindak, and E. B. Sirota for many helpful discussions about their experiments. This work was supported by the National Science Foundation through Grants DMR 88-17291 at Harvard and DMR 88-05443 at UCLA, and through the Harvard Materials Research Laboratory.

References

- [1] DIERKER S. B. and PINDAK R., Phys. Rev. Lett. 59 (1987) 1002.
- [2] SMITH G. S., SIROTA E. B., SAFINYA C. R. and CLARK N. A., Phys. Rev. Lett. 60 (1988) 813.
- [3] SELINGER J. V. and NELSON D. R., Phys. Rev. Lett. 61 (1988) 416.
- [4] SELINGER J. V. and NELSON D. R., Phys. Rev. A 39 (1989) 3135.
- [5] ZIPPELIUS A., HALPERIN B. I. and NELSON D. R., *Phys. Rev. B* 22 (1980) 2514;
 See also NELSON D. R., in Phase Transitions and Critical Phenomena, C. Domb and J. L. Lebowitz Eds. (Academic Press, London, 1983) p. 1.
- [6] OSTLUND S., TONER J. and ZIPPELIUS A., Ann. Phys. 144 (1982) 345.
- [7] PLEINER H. and BRAND H. R., Phys. Rev. A 29 (1984) 911.
- [8] CHUI S. T. and WEEKS J. D., Phys. Rev. Lett. 40 (1978) 733;
 See also WEEKS J. D., in Ordering in Strongly Fluctuating Condensed Matter Systems, T. Riste Ed. (Plenum, New York, 1980).
- [9] NOZIÈRES P. and GALLET F., J. Phys. France 48 (1987) 353.
- [10] NELSON D. R. and HALPERIN B. I., Phys. Rev. B 21 (1980) 5312.
- [11] NELSON D. R. and PELCOVITS R. A., Phys. Rev. B 16 (1977) 2191.
- [12] HOHENBERG P. C. and HALPERIN B. I., Rev. Mod. Phys. 49 (1977) 435.
- [13] This use of Γ_{\pm} is a slight change of notation from reference [4]. The bond-orientational viscosity ν of Dierker and Pindak [1] corresponds to $1/\Gamma_{\pm}$ in our notation.
- [14] This choice of l^* differs slightly from the choice in references [3, 4]. That difference does not affect any of the results.
- [15] KOSTERLITZ J. M., J. Phys. C7 (1974) 1046; see also reference [4].