When Boundaries Dominate: Dislocation Dynamics in Smectic Films

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We discuss the influence of dissipation at a system boundary (film-meniscus interface) on the dynamics of dislocation loops inside a smectic film. This dissipation induces a strong coupling between dislocations—effectively independent of their separation—leading to their nontrivial dynamics. Because of these dynamics, the effective "dynamical" radius of nucleation can be 10 times larger than the usual static critical radius.

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In the study of bulk properties of physical systems, one usually neglects the effect of boundary conditions; however, in exceptional cases where dissipation at the boundary dominates, this is not necessarily the case. One recently discussed example in solid state physics is electron conductance in quantum wires [1–3], where the finite resistance of a ballistic wire is a contact resistance that comes from processes that take place outside the wire. Here, we discuss a similar type of phenomenon, in soft-matter physics, where dissipation at the contact between a system and a reservoir leads to nontrivial, effectively infinite-range dynamics inside the system.

In many physical systems, transitions between metastable and stable states involve nucleation of a critical nucleus of the stable phase. The nucleus grows only when its size exceeds a critical size, which is set by the competition between the bulk and surface thermodynamic forces. We show in our example that dissipation at the boundaries of the system can change the size of the critical nucleus and its dynamics. The shift in the critical size induced by dissipation is analogous, but of different origin, to the one in Lifshitz-Slyozov theory [4].

A common problem in nonequilibrium statistical mechanics is the correct identification of dissipation function [5–8]. Here, we calculate explicitly the dissipation at a boundary and also show how to obtain the equations of motion either by minimizing the dissipation or by a local approach using the correctly identified thermodynamic forces.

In order to discuss the importance of dissipation at a system boundary, we take, as a paradigm, the dynamics of elementary dislocation loops in a freely suspended, smectic film [9,10]. This problem is directly connected to the dynamics of thinning transitions of smectic films [11], which occur when they are heated above their transition temperature towards the nematic (or isotropic) phases.

To perform the calculations, we assume that the film is circular, of thickness Nd (d is the layer thickness and N is their number) and radius r_m, and is coupled to a meniscus, which acts as a reservoir of particles. We consider n separated dislocation loops of radii r_k ($k = 1, \dots, n$), each characterized by the same line tension E and the same Burgers vector $b = d$ (Fig. 1). We suppose that they are far enough from each other to neglect their very short elastic interactions. We also neglect the hydrodynamic interactions coming from the flow inside the film. The film is stressed [10], due to the pressure difference $P_{\text{air}} - P_N$ across its free surface (P_{air} is the external pressure and P_N is the pressure inside the film), which is the driving thermodynamic force for dislocation growth.

Global approach to the dislocation dynamics: minimization of the dissipation.—To describe the dislocation dynamics we assume that the free energy gained per unit time, $W \equiv -\frac{dF}{dt}$ (with F = equilibrium free energy), in the whole system (film + meniscus) is entirely dissipated (no inertia). We find for W the following equation:

$$W = 2\pi \Delta P N d v_m r_m - 2\pi E \sum_{k=1}^{n} v_k , \qquad (1)$$

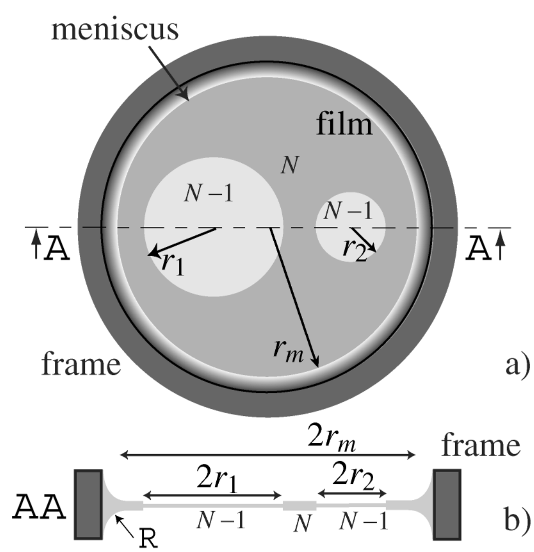

FIG. 1. Dislocation loops in a smectic film and its meniscus. (a) Top view; (b) cross section AA.

where v_k is the velocity of a kth dislocation (positive when the loop is growing and negative when it collapses) and

$$v_m = \frac{1}{N}\sum_{k=1}^{n} v_k r_k / r_m \qquad (2)$$

is the mean velocity of the fluid which is transported from the growing dislocation loops to the entrance of the meniscus (v_m is positive when the meniscus fills and negative when it drains). $\Delta P = P_{\mathrm{air}} - P_m = \gamma/\mathcal{R}$ [9,10] is the pressure drop across the free surface of the curved meniscus [P_m is the pressure in the bulk of the meniscus (in general different from the pressure in the film P_N) γ is the surface tension at the smectic-air interface and $\mathcal{R}$ is the radius of curvature of the meniscus profile]. The first term in Eq. (1) [also equal to $(\gamma/\mathcal{R})(dV_m/dt)$, where V_m is the volume of the meniscus] is the gain of the surface energy per unit time of the whole system (film + meniscus); the second term is due to the change of radii and, hence, of the line energy of the dislocations.

For the dissipation function we propose the following form:

$$\Phi = 2\pi d\mu \sum_{k=1}^{n} v_k^2 r_k + 2\pi C N d\mu v_m^2 r_m . \qquad (3)$$

The first term in Eq. (3) corresponds to the dissipation in the film due to the permeation flow around the core of the dislocations. The second term is associated with the flow of matter around the dislocations of the meniscus. It contains cross terms of type $v_i v_j$ with $i \neq j$, responsible for a dynamical coupling between the dislocations. Here $1/\mu$ corresponds to the usual dislocation mobility, assumed to be independent of the film thickness [10] ($1/\mu \approx \sqrt{\lambda_p/\eta}$ [12], where λ_p is the permeation coefficient and η is the shear viscosity parallel to the layers). The constant C characterizes the "strength" of the dissipation in the meniscus and will be calculated later.

In the absence of inertial effects, $W = \Phi$ at each instant of time. To obtain the equations of motion for the dislocations, we minimize Φ with respect to the velocities v_k, subject to the constraint $W = \Phi$. This is equivalent to assuming a minimum of the entropy production at each instant of time, which is valid only for stationary nonequilibrium states when phenomenological coefficients (η, λ_p, etc.) can be supposed to be constant [13]. This is obviously the case in our system, where velocities and corresponding Reynolds numbers are extremely small ($\mathrm{Re} = \rho v N d/\eta \approx 10^{-7} \ll 1$). We thus obtain a set of n equations ($k = 1, \dots, n$),

$$\frac{\partial \Phi}{\partial v_k} - 2\frac{\partial W}{\partial v_k} = 0, \qquad (4)$$

which gives, using Eqs. (1)–(3),

$$(\Delta P - C v_m \mu)d - d v_k \mu - E/r_k = 0. \qquad (5)$$

Local interpretation.—Equation (5) has a very simple interpretation in terms of *effective* forces acting on each dislocation: indeed, $-E/r_k$ corresponds to the inward line tension force, $-d v_k \mu$ corresponds to the frictional force opposed to the velocity, and $(\Delta P - C v_m \mu)d$ corresponds to the driving force ("Peach-Koehler" force [14]), also equal to $(P_{\mathrm{air}} - P_N)d$. Comparing the last two expressions yields

$$P_N - P_m = C v_m \mu . \qquad (6)$$

This equation shows that the pressure in the film differs from the pressure in the meniscus when matter enters or leaves the meniscus. The constant $C\mu$ thus characterizes the "permeability" of the meniscus.

Equation (5) ($k = 1, \dots, n$) may be rewritten in a form more suitable for numerical (or analytical) analysis:

$$V_k = 1 - 1/R_k - \frac{C\sum_{k=1}^{n} R_k(1 - 1/R_k)}{(R_m N + C\sum_{k=1}^{n} R_k)} . \qquad (7)$$

Here, we define the dimensionless velocity, $V_k = v_k/v_{\max}$, where $v_{\max} = \Delta P/\mu$ is the maximum velocity a dislocation can reach when $C = 0$. The dimensionless radius $R_k = r_k/r_c$, where $r_c = E/(\Delta P d)$ is the critical radius for a dislocation loop when $C = 0$.

Finally, we calculate the constant C. Two regions may be distinguished inside the meniscus (Fig. 2): In the first region, near the film, the dislocations are far enough from each other that the fluid velocity homogenizes over the sample thickness within the distance Λ_k that separates dislocations k and $k+1$ (we count the dislocations starting from the film-meniscus connection inward). This holds as long as the thickness of the permeation boundary layer that forms downstream from the kth dislocation at the distance Λ_k (of the order of $2\sqrt{l_p \Lambda_k}$, where l_p is the permeation length $l_p \sim \sqrt{\eta\lambda_p}$ [12]), is larger than the meniscus thickness $(N + k)d$ at this point. For a meniscus of circular profile and radius of curvature $\mathcal{R}$, $\Lambda_k = (d/2)\sqrt{\mathcal{R}/kd}$, so that this condition is fulfilled as long as $k < k^*$, with k^* the solution of the equation $2\sqrt{l_p \Lambda_{k}^{*}} = (N + k^*)d$, or, more explicitly,

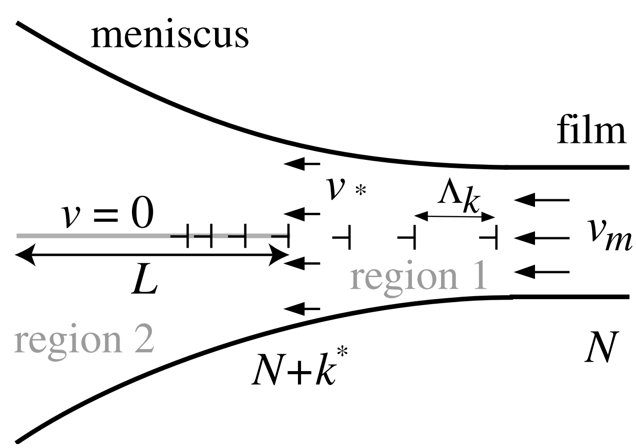

FIG. 2. Two zones of the meniscus showing the idea of the computation of the dissipation at the boundary.

$$k^*(k^* + N)^4 = \frac{4 l_p^2 \mathcal{R}}{d^3}. \tag{8}$$

As a consequence, the kth dislocation in region "1" ($k < k^*$) is immersed in a flow of constant velocity:

$$v_k = v_m \frac{N}{N + k - 1} \tag{9}$$

and dissipates energy $\phi_k = \mu d v_k^2$. Summing all these contributions gives the dissipation in region 1:

$$\phi^{(1)} = \sum_{k=1}^{k^*} \phi_k \approx \mu \frac{k^*}{k^* + N} N d v_m^2. \tag{10}$$

In region "2" ($k > k^*$), the dislocations are so close to each other that they form an obstacle similar to a ribbon of zero thickness and width L_m parallel to the layers, along which the velocity must vanish. This ribbon is immersed in a flow of velocity v_{k^*} given by Eqs. (8) and [9]. De Gennes has calculated the dissipation in this case [12] as

$$\phi^{(2)} = 8\mu \sqrt{\frac{2}{\pi}} \sqrt{L_m l_p}\, v_{k^*}^2. \tag{11}$$

Finally, the total dissipation per unit length of the meniscus reads

$$\phi_m = \left[\frac{k^* N d}{k^* + N} + 8\sqrt{\frac{2}{\pi}} \sqrt{L_m l_p} \left(\frac{N}{N + k^* - 1} \right)^2 \right] \mu v_m^2. \tag{12}$$

Comparing this to Eq. (3) gives the constant C:

$$C = \frac{k^*}{k^* + N} + 8\sqrt{\frac{2}{\pi}} \frac{\sqrt{L_m l_p}}{N d} \left(\frac{N}{N + k^* - 1} \right)^2. \tag{13}$$

The constant C is plotted as a function of N in Fig. 3, assuming $\mathcal{R} = 1$ mm, $d = 3$ nm, $l_p = 10$ nm, and $L_m = 300$ μm (this is the typical width for a circular meniscus). Figure 3 shows that C is typically between 20 and 110, in agreement with experiments [15]. We note that k^* given by Eq. (8) decreases almost linearly from 18 for $N = 3$ to 0 for $N \approx 40$.

Results.—We now discuss the case of a single dislocation loop ($n = 1$). Integrating Eq. (7) gives

$$t = \left(1 + \frac{C}{R_m N}\right)\left[R - R^i + \ln\left(\frac{R - 1}{R^i - 1}\right)\right] + C \frac{R^2 - (R^i)^2}{2 R_m N}, \tag{14}$$

where the time t is given in units of $r_c / v_{\max}$ and $R^i = R(t = 0)$ is the initial radius of the dislocation. Note that we assume in our calculation that $\mathcal{R}$ is constant: this is an excellent approximation because the film volume is negligible in comparison to the meniscus volume. In the absence of dissipation at the boundary ($C = 0$), one finds a linear growth ($R = t$ at long times when $R \gg 1$). The dissipation inside the meniscus slows down the growth of the dislocation: this effect is negligible in very thick films (more than 100 layers) but becomes very important in thin ones (typically less than 20 layers), where one finds a diffusive growth $R \sim \sqrt{t}$. This behavior has been observed experimentally and will be described in detail in a forthcoming article. Note that, for a single dislocation loop, the critical radius is not changed by the dissipation at the boundary.

When two or more loops are present, the critical radius of one dislocation depends on all the other radii, as may be inferred from Eq. (7). In particular, for two dislocations we find that $V_2 = 0$ when

$$R_2 = R_{2c} = \frac{N R_m + C R_1}{N R_m + C}. \tag{15}$$

This equation defines the dynamic critical radius of the loop "2" in the presence of the loop "1" of radius $r_1 = R_1 r_c$. This formula shows that the critical radius of the loop 2 increases when $r_1 > r_c$ ($R_1 > 1$) (i.e., when loop 1 is growing), whereas it decreases when $r_1 < r_c$ ($R_1 < 1$) (i.e., when loop 1 is collapsing). This effect may be easily understood in term of the pressure P_N inside the film. Indeed, R_{2c} may also be written in the equivalent form $R_{2c} = (P_{\text{air}} - P_m)/(P_{\text{air}} - P_N)$, where P_N is given by Eq. (6). When loop 1 is growing, P_N must be larger than P_m because the matter that is expelled from the loop must enter the meniscus. As a consequence, $r_{2c} > r_c$ ($R_{2c} > 1$). By contrast, P_N must be less than P_m when the loop 1 is collapsing so that $r_{2c} < r_c$ ($R_{2c} < 1$). The change of the critical radius can be large, especially in thin films (N small), when the radius of loop 1 becomes comparable to the radius of the meniscus. For example, $r_{2c} = 4.5 r_c$ when $N = 10$, $C = 77$, $R_m = 100$, and $r_1 / r_m = 0.5$. This value of r_c explains why it is so difficult to nucleate a new loop in thin films (for instance, by using the heating-wire technique described in Ref. [9]) while another loop of large radius (with respect to r_c) is growing.

To better illustrate the concept of the dynamic critical radius and show that the coupling via the meniscus may lead to nontrivial dynamics for two loops, we plot

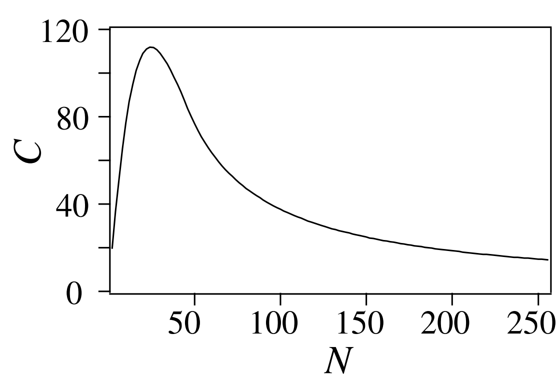

FIG. 3. The constant C as a function of the number of layers N in the smectic film.

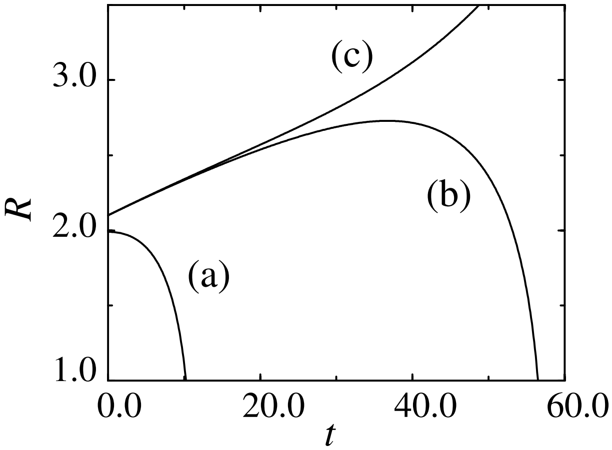

FIG. 4. The growth of a small dislocation in the presence of a big one for the following parameters: $N = 20$, $R_m = 100$, $C = 110$, $R_1^i = 20$, and $R_2^i = 1.99$ [curve (a)], $R_2^i = 2.1$ [curve (b)] $R_2^i = 2.101$ [curve (c)]. The parameters were chosen for the typical experimental case [9,10]. The radius is scaled by $r_c = E/\Delta pd$, and the time is scaled by $r_c/v_m ax$.

in Fig. 4 the radius $R_2(t)$ of loop 2 as a function of time t for different initial radii R_2^i while another (loop 1), of much larger radius, is growing. Our parameters are the following: $N = 20$, $R_m = 100$, $C = 110$ (see Fig. 3), and $R_1^i = 20$. Each value of R_1^i is associated with an initial critical radius for loop 2 given by Eq. (15). With our parameters, $R_{2c}^i = 1.99$. Figure 4 shows that three situations occur according to the value of R_2^i. If $R_2^i < R_{2c}^i$, loop 2 always collapses [curve (a)] as expected. If $R_2^i > R_{2c}^i$, loop 2 starts to grow, but its later behavior can change. More precisely, the loop grows continuously if $R_2^i > 2.1R_{2c}^i$ [curve (c)], whereas it starts to grow and then collapses when $R_{2c}^i < R_2^i < 2.1R_{2c}^i$ [curve (b)]: this nontrivial behavior shows that Eq. (15) can be satisfied at later times during the evolution of the two loops. The analogy with the Lifshitz-Slyozov model growth [4] is clear: as the first dislocation grows, the critical radius changes continuously and if the growth of the second dislocation is too slow its radius can eventually fall below the critical radius set by Eq. (15).

In conclusion, we have shown that in smectic films the separated dislocations do not grow independently because they are coupled via the dissipation in the meniscus. This dynamical coupling, which does not depend on the distance between the dislocations, may lead to nontrivial behavior, e.g., a change in critical radius during the growth process of many dislocations and to the diffusive growth of a single dislocation. A natural extension to this work would be the study of "step foams" (or "arch textures") that develop immediately after a film has been stretched. Finally, our example could serve as a paradigm for the physical phenomena in which dissipation at the system boundary constitutes an essential ingredient of the dynamics.

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[1] D. Maslov and M. Stone, Phys. Rev. B **52**, R5539 (1995).
[2] V. V. Ponomarenko, Phys. Rev. B **52**, R8666 (1995).
[3] I. Safi and H. J. Schultz, Phys. Rev. B **52**, R17040 (1995).
[4] I. M. Lifshitz and V. V. Slyozov, J. Phys. Chem. Solids **19**, 35 (1961).
[5] D. J. Evans and A. Baranyai, Phys. Rev. Lett. **67**, 2597 (1991).
[6] P. Gaspard and G. Nicoli, Phys. Rev. Lett. **65**, 1693 (1990).
[7] W. Breymann, T. Tel, and J. Vollmer, Phys. Rev. Lett. **77**, 2946 (1996).
[8] J. Vollmer, T. Tel, and W. Breymann, Phys. Rev. Lett. **79**, 2759 (1997).
[9] J. C. Géminard, R. Hołyst, and P. Oswald, Phys. Rev. Lett. **78**, 1924 (1997).
[10] F. Picano, R. Hołyst, and P. Oswald, Phys. Rev. E **62**, 3747 (2000).
[11] S. Pankratz, P. M. Johnson, R. Holyst, and C. C. Huang, Phys. Rev. E **60**, R2456 (1999); S. Pankratz, P. M. Johnson, A. Paulson, and C. C. Huang, Phys. Rev. E **61**, 6689 (2000); F. Picano, P. Oswald, and E. Kats, Phys. Rev. E **63**, 21 705 (2001).
[12] P. G. de Gennes, Phys. Fluids **17**, 1645 (1974).
[13] S. R. de Groot and P. Mazur, *Non-Equilibrium Thermodynamics* (Dover, New York, 1984), p. 45.
[14] J. Friedel, *Dislocations,* (Pergamon, London, 1964).
[15] The constant C can be estimated experimentally by measuring the equilibration time between two menisci of different radii of curvature. This method has given $C \approx 30$ for $N = 330$, in good agreement with our model.