Nonlinear Effects in Vibrating Smectic Films

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We observe nonlinear phenomena in vibrating smectic films. In particular, the shapes and positions of steps present in smectic films are affected by film vibrations. Above a certain threshold, we observe an instability in the meniscus consisting of the formation of a peninsula-shaped step. We point out that these effects are driven by a nonlinear inertial force which is tangent to the film surface and is proportional to the local density of the film and to the gradient of the film velocity squared.

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Unlike soap bubbles, the so-called free-standing smectic films are very robust and the number N of molecular layers (of the average thickness d) they contain can be controlled, one by one, from $N = 2$ to very large numbers [1,2]. For this reason, they have attracted much attention, and many structural studies have been devoted to them over the last two decades [3,4]. Smectic films have also been studied because of their remarkable physical properties. For example, they have been used for studies of nonequilibrium, nonlinear phenomena such as the pattern formation in a rotating electric field [5].

Another remarkable property of smectic films, important for the purpose of the present Letter, is that they cannot exist on their own but must always be suspended on a frame by the intermediate of a meniscus [6]. Because the meniscus plays the role of a reservoir of particles with respect to the film, smectic films of different thicknesses $h = Nd$ form a set of thermodynamically metastable states whose stability, one with respect to the other, is ruled by their tensions τ_N. These are defined as the derivative of the free energy f_N per molecule with respect to the surface area per molecule [6],

$$\tau_N = \partial f_N(a,T)/\partial a\,. \tag{1}$$

Since the tension of films plays such a crucial role, several methods have been developed for measuring it. One of these consists of "hearing the tension of films," that is to say, in measuring frequencies of the eigenmodes of transverse vibrations [6–8]. Indeed, smectic films composed of liquid Sm*A*- or Sm*C*-like layers can be seen as perfect membranes in the Rayleigh sense [9]: (1) their specific mass per unit area ρ_N is uniform, (2) they are subjected to the isotropic tension τ_N so that (3) their transverse vibrations are governed by the wave equation. Obviously, this is true only in the limit of small vibrations which later neither the film tension nor its density.

This is not always the case. Indeed, we will report here on the first observations of several *nonlinear phenomena* occuring in Sm*A*- or Sm*C*-like films when the amplitude of their transverse vibrations is large enough.

The experimental setup is depicted in Fig. 1. The smectic film is supported by a horizontal flat rectangular frame of dimensions $L_x = 0.53$ cm and $L_y \approx 0.53$ cm. Since one of the sides of the frame is mobile, it is easy to "pull" films of arbitrary thickness by controlling dynamically the size of the frame [6]. For convenience, room temperature Sm*A* and SmC^* mixtures (S2 and SCE4 from BDH) have been used. Vibrations of films $\zeta(x,y,t) = A\cos(\omega t)g(x,y)$ were excited acoustically by a loud speaker situated about 5 cm below the film, acting as a piston which modulates the air pressure below the film. The fundamental mode of approximate shape $g(x,y) = \sin(\pi x/L_x)\sin(\pi y/L_y)$ and of frequency $\sim$380 Hz is excited very efficiently by this method so its amplitude A can easily be made large enough for the observation of the nonlinear phenomena.

The most striking nonlinear phenomenon (and the one which was first observed) consists of the instability of the meniscus shown in the photographs of Fig. 2. We call this phenomenon the *peninsula instability.* The images are typical for a very thin (the number of layers N_0

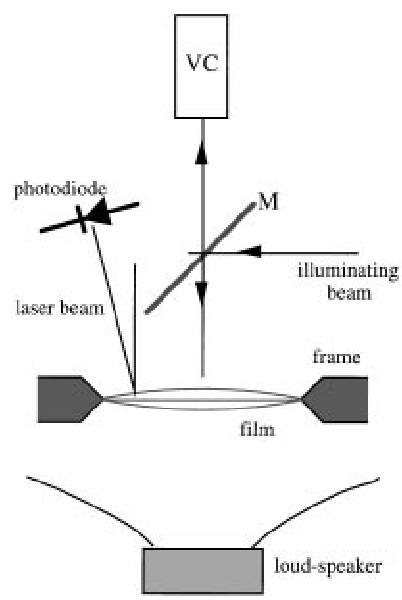

FIG. 1. Experimental setup. Vibrations of the smectic film held on the rectangular frame are driven acoustically by the loud speaker. The amplitude of vibrations is measured optically using a laser beam reflected from the edge of the film. The film is observed in reflecting illumination by the video camera (VC).

is less than 10) SmC^* film. At rest, the meniscus is very narrow and, consequently, its width (less than 0.1 mm in the x-y plane) is uniform everywhere except at the edges of the frame. In its thinnest portion, the meniscus can be considered as a collection of steps. At rest, the distribution of steps in the meniscus is stable and determined by the set of tensions τ_N and by the interactions between the steps. Above a threshold $A \approx 0.01$ cm, the meniscus becomes unstable; thick steps emanate from it and invade the film. The thickness ΔN of these steps depends on several factors such as the film thickness N_0, the film structure, and the amplitude of vibration. $\Delta N \approx 20$ layers is typical for SCE4 films of thickness $N_0 \approx 10$ at room temperature. Shapes $y = y(x)$ of these steps evolve as a function of time. First, they are bumplike [Fig. 2(a)], later, one (or several) of the bumps take the shape of a kind of peninsula connected to the meniscus on the side of the frame by a narrowing neck and, finally, when the neck breaks (this can take more than several minutes in SmC^* films) an island forms and floats freely in the film. The island goes to the center of the film and stays there [10]. The island shape $r(\theta)$ shows deviations $\delta r = r(\theta) - r_0$ from the circular one.

This evolution of the bump shape involves a progressive increase in the surface area $S_{N+\Delta N}$ of the bump (peninsula) and occurs on the time scale of several minutes. We think that this evolution is a quasistatic process in the sense that for a given surface area $S_{N+\Delta N}$ of the bump, its shape is in equilibrium with respect to the redistribution of the matter in it. Indeed, experiments show that when the amplitude of the film vibration is changed rapidly, the bump or the island change their shape in a characteristic time shorter than 1 sec. Theoretically, this relaxation time can be estimated in the same way as in the case of LB islands studied by Mann *et al.* [11]. For the step of tension $\gamma \approx 5 \times 10^{-6}$ dyn [6] and thickness $h \approx 6 \times 10^{-6}$ cm, floating in the film of viscosity $\eta \approx 1$ P one gets $T_r \approx \eta h/\gamma q \approx 10^{-2}$ sec when the wave vector of the shape deformation is $q \approx 10^2$ cm^{-1}.

In the presence of vibrations, the quasistatic shapes of bumps, peninsulas, or islands are determined by the balance of forces acting on the step. For a step of tension γ, one has

$$\gamma/R(x,y) = \Delta\tau(x,y) + f_{\mathrm{int}}(x,y) . \qquad (2)$$

The left-hand side of the equation represents the Laplace force proportional to the local curvature $1/R(x,y)$ of the step which has the tension γ. On the right-hand side, one finds the difference $\Delta\tau = \tau(x,y;N+\Delta N) - \tau(x,y;N)$ between the film tensions on the two sides of the step and the force of interaction f_{int} which exists between the steps when they are close enough.

Without vibrations, the mechanical equilibrium in fields of the uniform thicknesses N and $N + \Delta N$ requires that the tensions $\tau(x,y;N)$ and $\tau(x,y;N+\Delta N)$ must be uniform. Therefore, as expected, the curvature of the step must be constant if $f_{\mathrm{int}} = 0$.

As shown in the photographs of Fig. 2, in the presence of vibrations the curvature of the step varies along the bump and changes its sign between the summit and the base. In the absence of interactions with other steps, Eq. (2) indicates that the difference in the film tensions $\tau(x,y;N)$ and $\tau(x,y;N+\Delta N)$ must vary along the step. On the other hand, as there are no flows in the film, these spatial variations in the film tensions must result from some force $\mathbf{f}_{\mathrm{NL}}$ per unit area, which is generated by the film vibrations $\zeta(x,y,t) = A\cos(\omega t)g(x,y)$ and which depends on the film thickness N, such that

$$\vec{\nabla}\tau(x,y;N) = \vec{f}_{\mathrm{NL}}(x,y;N) . \qquad (3)$$

The same force causes the *motion of islands* floating in the film. By studying motions of a small island induced by the fundamental eigenmode of the rectangular film, one finds that all trajectories start from the meniscus and coverage at the center of the film. From the velocity of the island along such trajectories, one finds that the force $\mathbf{f}_{\mathrm{NL}}$ vanishes at the meniscus and in the center. Finally, balancing the gravitational force in tilted films, one finds that the force $\mathbf{f}_{\mathrm{NL}}$ is proportional to $(A\omega)^2$. The simplest

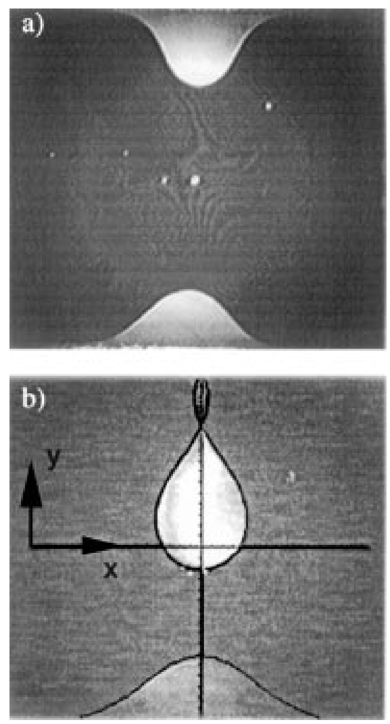

FIG. 2. Peninsula instability of the meniscus in a SmC^* film. The black background consists of a very thin ($N < 10$ layers) film. (a) The bright bumps are about $\Delta N = 20$ layers thicker. They come out from the meniscus above a certain excitation threshold, and their surface area $S_{N+\Delta N}$ increases as a function of time. The shape of the bump is a function of its surface area and becomes peninsulalike for large S. (b) The shape of steps calculated from Eq. (10) are superposed on photographs of real steps.

hypothesis, in agreement with all these indices, is that

$$\vec{f}_{\rm NL}(x,y;N) = \rho_N \vec{\nabla}(\dot{\zeta}^2)/2\,. \quad (4)$$

We argue that the *physical origin of this NL (nonlinear) force* is a follows: Let us consider a surface element of coordinates (x,y) belonging to the vibrating film of shape $\zeta = \zeta(x,y,t)$ and suppose that, as shown in Fig. 3, the trajectory $\mathbf{r}(t)$ of this element is at each time orthogonal to the surface of the film. The velocity of this element is then

$$\vec{v} = \frac{d\vec{r}}{dt} = \frac{\partial\zeta}{\partial t}\left(-\frac{\partial\zeta}{\partial x}, -\frac{\partial\zeta}{\partial y}, 1\right) \Big/ \left[\left(\frac{\partial\zeta}{\partial x}\right)^2 + \left(\frac{\partial\zeta}{\partial y}\right)^2 + 1\right]. \quad (5)$$

The acceleration of the surface element on its trajectory is then, up to the second order in ζ,

$$\vec{a} = \frac{\partial^2\zeta}{\partial t^2}\left(-\frac{\partial\zeta}{\partial x}, -\frac{\partial\zeta}{\partial y}, 1\right) + \frac{\partial\zeta}{\partial t}\left(-\frac{\partial^2\zeta}{\partial t\,\partial x}, -\frac{\partial^2\zeta}{\partial t\,\partial y}, 0\right). \quad (6)$$

The first term of this expression corresponds to the acceleration component orthogonal at each time to the film surface and, consequently, is balanced by the Laplace force due to the mean curvature of the film. The second term corresponds to the centripetal acceleration tangent to the film surface. Multiplied by $-\rho_N$, is gives the centrifugal force per unit area which is equal to the NL force suggested above in Eq. (4).

Let us suppose not that the shape of the film during its vibration is that of the eigenmode of frequency ω: $\zeta(x,y,t) = A\cos(\omega t)g(x,y)$. The time-averaged value of the NL force is then

$$\vec{f}_{\rm NL} = (\rho_N\omega^2/2)\vec{\nabla}\langle\zeta^2\rangle_t = [\rho_N(\omega A)^2/4]\vec{\nabla}[g(x,y)^2]\,. \quad (7)$$

From Eq. (3), we know that in the absence of flows in the film this NL force must be canceled by gradients in the film tension

$$\rho_N\omega^2\vec{\nabla}\langle\zeta^2\rangle/2 = -\vec{\nabla}\tau_N \quad (8)$$

so that the film tension defined in Eq. (1) becomes position dependent,

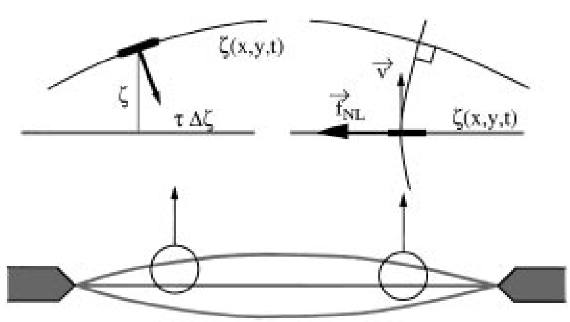

FIG. 3. Genesis of the nonlinear inertial force: trajectories of molecules in the vibrating smectic film are curved so that there is a nonzero time-averaged centrifugal force $\mathbf{f}_{\rm NL}$.

$$\tau(x,y;N) = (\rho_N\omega^2/2)\langle\zeta(x,y)^2\rangle + \tau_0(N)\,. \quad (9)$$

Therefore, one can say that the average effective tension τ of the films is lowered by its vibrations.

When the film of thickness N contains a step of height ΔN, the nonlinear forces are different on the two sides of the step because of the difference $\Delta\rho = \rho_{\rm cl}d\Delta N$ in the two-dimensional density ρ_N. Consequently, the discontinuity in the tension of the film

$$\Delta\tau(x,y) = -(\rho_{\rm cl}\Delta N d\omega^2/2)\langle\zeta(x,y)^2\rangle + \Delta\tau_0 \quad (10)$$

will build up along the step. Replacing the local step curvature $1/R(x,y)$ in Eq. (2) by its explicit expression, one gets for the eigenmode $\zeta(x,y,t) = A\cos(\omega t)g(x,y)$ the following differential equation:

$$\frac{d^2y}{dx^2} \Big/ \left[1 + \left(\frac{dy}{dx}\right)^2\right]^{3/2} = K_{\rm NL}g^2(x,y(x)) + K_0 + K_{\rm int}, \quad (11)$$

where $K_{\rm NL} = \Delta N\rho_{\rm cl}dA^2\omega^2/4\gamma_{\Delta N}$ is the step curvature due to the NL forces, $K_0 = \Delta\tau_{\Delta N}/\gamma_{\Delta N}$ is the rest curvature of a free step, and $K_{\rm int} = f_{\rm int}/\gamma_{\Delta N}$ results from interactions between steps when they become close enough.

In the experiments for which the photographs are shown in Fig. 2, the last two terms of Eq. (11) are important at the base of the peninsula where the curvature due to the NL forces vanishes. The spontaneous curvature K_0 is positive because the tension of the film increases with its thickness [6]. Its typical value is of the order of $10\,{\rm cm}^{-1}$. The interaction curvature $K_{\rm int}$ is negative when the interaction between steps is strictly repulsive and its modulus grows when steps get close together. Due to this, the sum $K_0 + K_{\rm int}$ varies between K_0 and zero when the step becomes tangent to the meniscus. When (starting from the base of the bump) one follows the step in the opposite direction, one observes that the local curvature decreases, changes sign, and reaches its maximum value at the bump summit. This behavior is due to the inertial curvature term $K_{\rm NL}$ growing as $g^2(x,y)$ when the bump $y(x)$ penetrates the vibrating area of the film. In order to get the inversion of the total curvature, the inertial term must be larger than the spontaneous one. The magnitude of the inertial curvature $K_{\rm NL}$ depends essentially on two factors. First of all it depends on $(\omega A)^2$, that is to say, on the square of the vibration velocity amplitude—this parameter is accurately controlled in our experiments through the loud-speaker excitation voltage. The second factor is the ratio between the 2D density discontinuity $\Delta\rho_{\Delta N} = \Delta N d\rho_{\rm cl}$ and the tension $\gamma_{\Delta N}$ of the step. If the step tension is approximated by its maximal value $\tau\Delta N d$, then this ratio is independent of the step thickness and equals $\rho_{\rm cl}/4\tau \approx 10^{-2}\,{\rm sec}^2/{\rm cm}^2$. In order to get $K_{\rm NL} > K_0$, one has to satisfy the condition $(\omega A)^2 > K_0/10^{-2} = 10^3$ cm/sec. For $\omega \approx 2\pi \times 380$ sec^{-1}, one gets $A > 0.01$ cm in agreement with experiments.

The validity of the differential equation (11) has been checked by comparing its numerical solutions, for $g(x, y) = \sin(x\pi/L_x)\sin(y\pi/L_y)$, with the shapes of the bump and of the peninsula on the Fig. 2(b). For the bump, the agreement is satisfactory but in the case of the peninsula, the calculation yields intersecting steps at the base of the neck. This nonphysical feature is due to the fact that the interaction curvature has been neglected in Eq. (11). The agreement between the experimental and calculated shapes of steps is surprising if one realizes that the shape $g(x, y)$ of the eigenmode could be perturbed by the discontinuity in the film density due to the presence of the bump. Fortunately, when experiments are performed in air and the film is thin enough, the inertia of the air participating in vibrations of the film is much larger than that of the film itself so that the density discontinuity in the film has little effect on $g(x, y)$.

This is no more the case when a small droplet of the smectic liquid crystal or a small tin sphere floats in the film. The mass of such an inclusion perturbs the film vibrations $\zeta(\mathbf{r}, t)$ in a way that depends on its position $\mathbf{R}_i$ in the film. Inversely, the inclusion is subjected to the nonlinear force similar to the one discussed above so that its position $\mathbf{R}_i$ is influenced by the vibrations. Because of this feedback loop, the behavior of the inclusion depends on the frequency and the amplitude of the excitation. In the simplest case, the position of the inclusion in the film is stable but depends on the frequency of the excitation. In particular, for a certain fundamental frequency ω_0, which depends on the mass of the inclusion, the stable position is situated at the center of the film. One can say that the vibrating film acts as a *mechanical trap* for particles. The behavior of heavy inclusions is much more complicated because they perturb the vibrations of the film so much that the shape $g(\mathbf{r}; \mathbf{R}_i)$ and the frequency $\omega(\mathbf{R}_i)$ of eigenmodes depend on the position $\mathbf{R}_i$ of the mass in the film.

In conclusion, we reported here for the first time on several nonlinear phenomena occuring in vibrating smectic films. The peninsula instability of the meniscus is one such example. Shapes and positions of steps, preexisting or created by this instability, are affected by the film vibrations. We have pointed out that this class of phenomena is due to the nonlinear force acting on each surface element of the vibrating film and this force is proportional to the gradient of the film velocity and to the local density of the film. Another class of nonlinear phenomena concerns the behavior of inclusions floating in smectic films. This depends significantly on the mass and the shape of the inclusion as well as on the thickness of the film. Since the motions of small inclusions are strongly damped by the film viscosity, they tend to take stable positions in the vibrating film which acts as a kind of mechanical trap. We think that this kind of trap could be used successfully for manipulation of large macromolecules or cells.

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