Meniscus Instability in a Thin Elastic Film

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A new kind of meniscus instability leading to the formation of stationary fingers with a well-defined spacing has been observed in experiments with elastomeric films confined between a plane rigid glass and a thin curved glass plate. The wavelength of the instability increases linearly with the thickness of the confined film, but it is remarkably insensitive to the compliance and the energetics of the system. However, lateral amplitude (length) of the fingers depends on the compliance of the system and on the radius of curvature of the glass plate. A simple linear stability analysis is used to explain the underlying physics and the key observed features of the instability.

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While the instabilities of thin confined viscous films [1] have attracted considerable attention in the context of spontaneous surface deformation/dewetting and fingering phenomenon, there is as yet neither a systematic study of spontaneous surface roughening in confined elastic films nor an analysis of the factors responsible for it. The general question of what happens when an elastic surface approaches a confined elastic film is important for a fundamental understanding of purely elastic instabilities that may ensue without any concurrent mass flow, and in turn affect the interfacial properties such as friction and adhesion. This issue is also of technological importance in a variety of settings such as the peeling of a pressure sensitive adhesive from a solid surface [2–4], the crazing behavior of glassy polymers [5], interfacial bonding in composite materials, morphological instability of a biaxially stressed interface in crystal growth situation [6], elastic surfaces becoming unstable under the action of compressive [7,8] and tensile stresses [9], and stability of polymer brushes used for improved compatibility [10].

The purpose of this Letter is to describe and systematically characterize the properties of a new kind of elastic meniscus instability that develops merely by contacting a glass plate with a thin elastomeric film bonded to a rigid support, i.e., the situation of a crack closing in a confined elastic film. The elastomeric film was prepared by platinum catalyzed hydrosilation reaction of vinyl end-capped dimethylsiloxane oligomers to a methylhydrogensiloxane cross-linker. Films of uniform and controlled thickness (40–700 μm) were prepared by cross-linking the polymer within two glass slides of adjustable spacing. One of the glass slides was pretreated with a low energy silane monolayer, which ensured its easy removal from the elastomeric film after it was fully cross-linked. The elastomeric film, however, remained adhered to the other glass slide. The shear modulus (0.07–2.0 MPa) of the elastomer was controlled by the molecular weight of the dimethyl siloxane oligomer that varied from 2000 to 50 000.

In a typical experiment, a glass cover slip was brought into contact with the elastomeric film in the configuration of a cantilever beam or a curved elastica loaded by means of spacers inserted in the opening of the cracks (Fig. 1). The loading on the crack can be either external, in which the crack opens under external force, or internal, in which the crack heals by moving toward the stationary spacer by the forces of adhesion. For the most part of our studies the configuration depicted in Fig. 1(a) was used. Crack closures in both cases are governed by the thermodynamic work of adhesion (44 mJ/m^2) [11–13]. Figure 2 shows the instabilities at the crack front, in the form of wavy undulations that develop during the closure of the crack but remain stable even after the crack comes to a complete

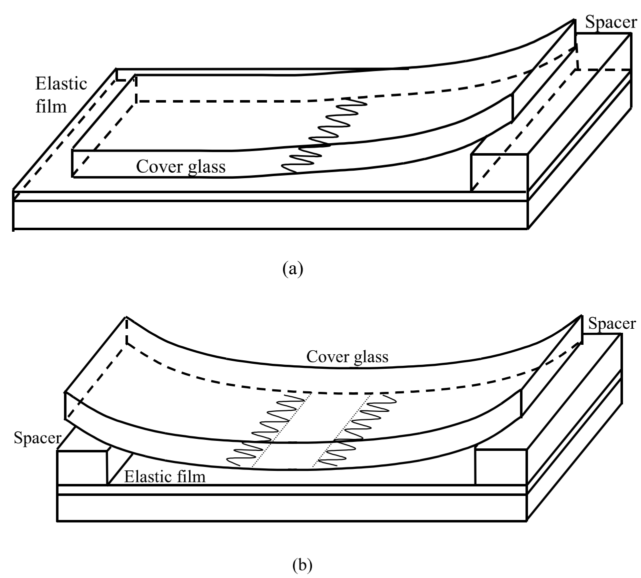

FIG. 1. Schematics of the experiment, in which a cover glass plate is brought in contact with the elastomeric film in the configuration of either a cantilever beam (a) or a curved elastica (b). In experiment (b), the radius of curvature of the elastica is controlled by adjusting the distance between the spacers.

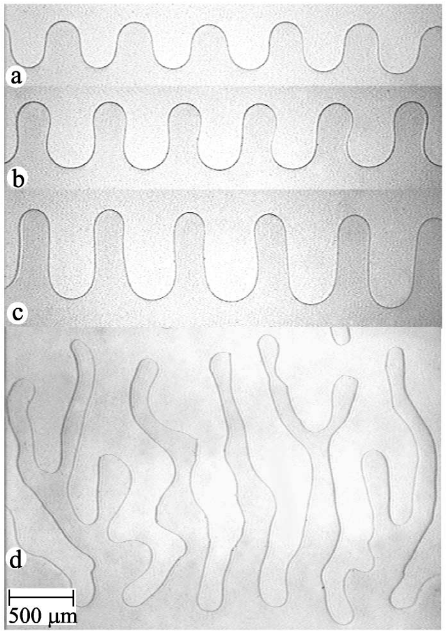

FIG. 2. Video micrographs of meniscus instabilities caused by the contact of glass beams [see Fig. 1(a)] of various flexural rigidities against an elastomeric film of thickness 150 μm and shear modulus 1.0 MPa. (a), (b), and (c) correspond to cover glasses of flexural rigidities 0.02, 0.09, and 0.2 N m, respectively. The arrow indicates the direction in which the crack closes. Note that the amplitude, but not the wavelength, depends on the rigidity of the backing. A highly irregular growth of fingers is observed (d) for a glass beam of flexural rigidity 1.0 N m.

rest. The wavelength (λ) and amplitude (a) of these instabilities are the two most important geometrical features that describe the morphology of the waves. Experiments with films of different thickness and modulus show that the wavelength increases linearly with thickness (h) of the film, but it is relatively independent of either the elastic modulus of the film or the flexural rigidity [14] (D) of the glass cover plate. The wavelengths obtained from numerous experiments can be plotted on a single master curve (Fig. 3) with the result $\lambda = (3.94 \pm 0.06)$ h. Interestingly, the wavelength is also independent of the surface energies of the film and the substrate. The later prediction was confirmed in experiments, where the elastic film was plasma oxidized in order to enhance its adhesion to glass. Although the increase of adhesion was evident from the asymmetric cantilever beam experiment, no discernible change of λ could be observed.

Unlike the well-known Saffman-Taylor instabilities, no material flow is involved in the formation of elastic fingers. Here the instability is triggered by such stresses as the van der Waals and/or electrostatic interactions near the contact zone of the cover beam, which also manifest in the energy of adhesion, normal stresses and deformation near the contact line. A simple model of Shenoy and Sharma [15] that reproduces the key features of the instability considers a nearly incompressible (Poisson's ratio, $\nu \to 0.5$) elastic film with a shear modulus μ, and a plane

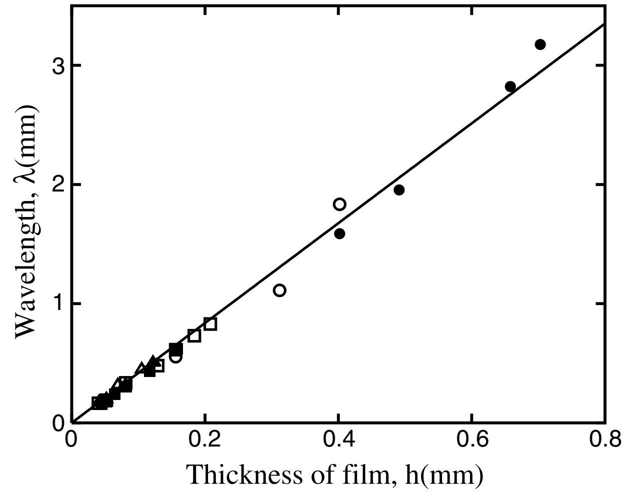

FIG. 3. The wavelength (λ) of the meniscus instability increases linearly with the thickness (h) of the confined elastomeric film. Although the wavelength increases linearly with thickness, it is insensitive to either the elastic modulus of the film or the stiffness of the cover glass. Symbols △, ▲, and ■ correspond to a cover glass plate of flexural rigidity $D = 0.02$ N m against elastomeric films of moduli (μ) 2.0, 1.0, and 0.25 MPa, respectively; □ corresponds to $D = 0.09$ N m and $\mu = 1.0$ MPa; ○ and ● correspond to $D = 1.0$ N m and $\mu = 0.2$ and 0.07 MPa, respectively. Data obtained from all these cases fall on a single straight line.

strain elastic model of the film for mathematical simplicity (Fig. 4).

The total energy budget of the system consists of the elastic energy in the film (stabilizing), energy of interaction with the cover beam (manifested in normal stresses causing deformation), and the surface energy of the film (stabilizing, but usually negligible). In the linearized approximation, these are, respectively,

$$\begin{aligned} E = & \int_V W(\epsilon)\, dV \\ & - \int_S \left(U_0 + F_0 \mathbf{u} \cdot \mathbf{n} + \frac{1}{2} Y (\mathbf{u} \cdot \mathbf{n})^2 \right) dS \\ & + \int_S \gamma \sqrt{1 + (u_{2,1})^2}\, dS \,, \end{aligned} \tag{1}$$

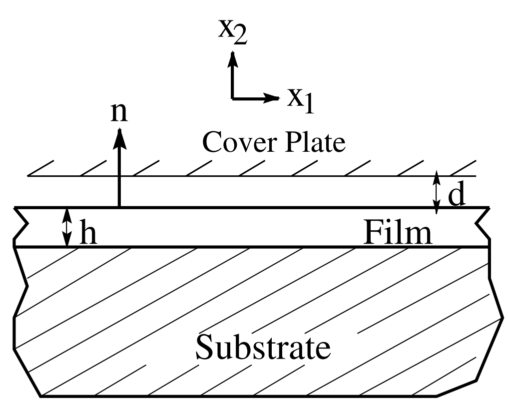

FIG. 4. Schematics of the theoretical model.

where ϵ is the strain tensor, γ is the surface energy of the film, $\mathbf{u}$ is the displacement vector in the film, and U is the interaction energy per unit area that depends on the gap thickness (d). Linearization produces $U_0 = U(0)$, $F_0 = U'(0)$, and $Y = U''(0)$, where (0) denotes the undeformed film configuration. A full solution of the problem, considering the equilibrium stress field, $\nabla \cdot \sigma = 0$ (which minimizes the total energy) with appropriate boundary conditions [$\mathbf{u} = 0$ at the rigid plate-film interface; $\sigma \cdot \mathbf{n} = \gamma u_{2,11}\mathbf{n} + F_0\mathbf{n} + Y(\mathbf{u} \cdot \mathbf{n})\mathbf{n}$ on the film surface], leads to the following interesting conclusions [15]. There exists a homogeneous solution such that stresses in the film are constant everywhere. A bifurcation analysis reveals that this homogeneous solution is unstable to periodic deformations of the form $u \sim \exp(ikx_1)$. Regardless of the nature of the force causing the instability, a periodic deformation of the film surface ensues whenever Y exceeds a critical value (i.e., when the gap thickness d is sufficiently small),

$$Y > 6.22\ \mu/h, \quad \text{and,} \quad \text{since } Y \sim d^{-n},\ d < dc\,. \quad (2)$$

For example, $n = 4$ and 5 for the long range nonretarded and retarded van der Waals forces, respectively. The wave number ($k = 2\pi/\lambda$) of the instability is independent of μ, and even more surprisingly, independent of both the precise nature of the force and its magnitude, and thus independent of the energy of adhesion. This is because in the first order approximation, the interaction energy does not depend on the wavelength λ. For nearly incompressible materials (Poisson's ratio $\nu \rightarrow 0.5$), and for small values of surface energy ($\gamma/\mu h \ll 1$), it is shown that [15]

$$kh = 2.12 - 2.86(1 - 2\nu) - 2.42(\gamma/\mu h)\,. \quad (3)$$

In our experiments ($\nu \rightarrow 0.5$, $\gamma/\mu h \ll 1$), this produces a linear variation $\lambda = 2.96$ h. A somewhat lower proportionality factor of the theory is due to the use of a plain strain model (plane rigid cover beam). This is confirmed by recent experiments using rigid flat covers where the numerical coefficient is precisely the same as the theoretical prediction [16].

Lateral amplitude (A) of the fingers, on the other hand, decreases only slowly with the increase of the modulus and the thickness of the elastomeric coating, but it increases substantially with the flexural rigidity (D) of the glass cover beam. Very long fingers with split tips and side branches are observed for cover plates with high rigidity. One interesting observation is that for each compliant cover plate (for a given D), a critical thickness is reached beyond which no instability is observed. The condition, Eq. (2), provides a gap distance beyond which no periodic deformation of the elastomeric film is possible. Typically, this critical distance is of the order of 10 nm for van der Waals interaction, which prevails very close to the crack tip region, but can be substantially higher for electrostatic interactions. However, once the critical wavelength is selected, the detailed morphology of the undulation is determined by the overall energetics. A simple picture emerges if we consider that the amplitude of the instability is related to the lateral width (b) of the stressed zone within the elastomer. For a thin elastomeric film, this length (b) is related to the radius of curvature (ρ) of the cantilever and the displacement (u_0) of the elastomer in the crack tip region as follows, $b \sim (u_0\rho)^{1/2}$. Using local energy balance, it can be shown that the deformation (u_0) of the elastomer scales as [12] $u_0 \sim (Wh/\mu)^{1/2}$, whereas the global energy balance for an asymmetric cantilever yields the well-known result [17] $\rho \sim (D/W)^{1/2}$. Using the above definitions of u_0 and ρ, one obtains a scaling relation for the lateral width of the stressed zone as

$$b \sim (Dh/\mu)^{1/4}. \quad (4)$$

Exactly the same scaling relation for b can also be obtained from the displacement equation of the cantilever [18], viz., $Dd^4u/dx^4 + (3\mu u/h) = 0$, where u is the displacement of the cantilever measured from the undisturbed surface of the film of thickness h and x is the distance measured from the crack tip (at $x = 0$, $u = u_0$; at $x = \infty$, $u = 0$).

Because of incompressibility, the amplitude, as a first approximation, can be taken to be proportional to b. Therefore, amplitude should increase with the flexural rigidity of the cover plate and decrease with the modulus of the elastomer. Both of these predictions are in qualitative agreement with the experimental observations. However, once the amplitude becomes larger than λ the fingers have the tendency to split and engender a two-dimensional "ripplelike" pattern with an increasingly isotropic wave number witnessed in the limiting case of a rigid plane cover plate [16]. This discussion is however incomplete as it does not predict the slow decrease of the amplitude with the thickness of the elastomer film and, in particular, the vanishing of amplitudes at critical values of h. This is a nonlinear phenomenon, a comprehensive treatment of which must account for the effects of the finite dilation (i.e., $\nu < 0.5$) of films on the release of confinement. Qualitatively, however, it is expected that the thicker films are less constrained to deform laterally than the thinner films and thus would inhibit instability.

For an elastica of fixed D but variable curvature, Eq. (4) predicts that the width of the stress zone (b) increases with ρ, i.e., $b \sim (W\rho^2h/\mu)^{1/4}$. The sensitivity of the amplitude on the width of the deformation zone is studied using the arrangement shown in Fig. 1(b), in which the radius of curvature of the cover glass plate is manipulated by adjusting the distance between the spacers. An increase in ρ increases the width of the deformation-induced contact zone (b) and also increases the amplitude of the instability (Fig. 5).

To summarize, a new type of elastic instability leading to the formation of stationary fingers arising from the contact between flexible glass plates and thin elastomeric films has been reported. The origin of the instability is fundamentally very different from the well-known problem of Saffman and Taylor [1] and that recently reported by Shull *et al.* [9]. These previously reported instabilities are

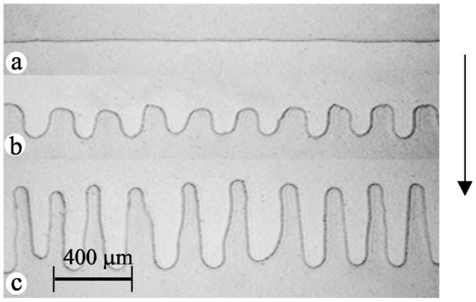

FIG. 5. Video micrographs of meniscus instabilities caused by the contact of a glass beam of flexural rigidity of 0.02 N m [in the configuration of Fig. 1(b)] with an elastomeric film of thickness 52 μm and shear modulus 1.0 N m. (a), (b), and (c) correspond to ρ equal to 25, 37, and 60 cm, respectively.

triggered by the gradient of hydrostatic stress and are inhibited by the surface tension. In our case, the wavelength of the instability is neither dependent on the surface tension nor on the modulus of the elastomer. The fact that these undulations evolve into a 2D rippling pattern for flat and infinitely rigid plates suggests that no hydrostatic pressure gradient is necessary for this type of instability. Furthermore, for cantilevers with finite rigidity, the hydrostatic tension decreases from the edge, which in fact tends to inhibit the meniscus instability in a pure elastic situation. We believe that these results are of importance in understanding the adhesion of thin films in general and friction in particular. The 2D rippling patterns, which are invisible to the eye, provide potential sites for cavitation when a normal or shear stress is applied to the junction. These cavitationlike appearances provide pathways for stress relaxation in the fracture and shearing of junctions. A detailed account of these studies will be published elsewhere.

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