

Pattern Formation in a Thin Solid Film with Interactions

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We investigate a new type of surface instability of a thin elastic film subjected to surface interactions such as van der Waals and electrostatic forces from another solid surface in its vicinity. It is found that a sufficiently soft (shear modulus < 10 MPa) and nearly incompressible film deforms to form an undulating pattern without any mass transport. A novel feature is that the characteristic length scale of the pattern is nearly independent of the nature and magnitude of the external force, but varies linearly with the film thickness. These results explain some recent experiments and are applicable to problems such as adhesion and friction at soft solid interfaces, peeling of adhesives, patterning of solid surfaces, etc.

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Surface instability of viscous thin liquid films engendered by intermolecular interactions or external fields has attracted much recent attention [1,2], where the length scale and morphology of the instability depends very strongly on the precise nature and magnitude of the interactions. However, very recent experiments [3–5] employing a variety of elastic thin solid films in contact with different types of nearly rigid surfaces consistently show a periodic elastic deformation of the film surface, the wavelength of which is independent of the nature of the interaction between the film and the contacting surface. The wavelength is found to be always of the same order as the film thickness and scales linearly with it. There is as yet no precise theoretical understanding of either the underlying physics of the instability or the factors governing its characteristics. What happens when a surface approaches a soft elastic film is of fundamental importance in understanding morphological instabilities that arise in such systems without any concurrent mass flow. The interfacial instability in turn may have a profound influence on the interfacial properties such as friction and adhesion. Technological applications include peeling of adhesives [6], stability of polymer brushes [7], pattern transfer technology [2], etc.

Here we explore the stability of an elastic thin solid film (initially stress-free) bonded to a rigid substrate whose surface experiences effective forces which, in general, depend on the proximity of the contacting surface and also on the local film thickness. These forces may arise from any of the various causes such as the long-range van der Waals interaction with the contacting surface nearby and/or with the substrate, an external electric field, etc. The analysis presented in this Letter shows that morphological instabilities of a thin solid film are independent of the nature and strength of the interactions as long as it exceeds a threshold value. Physically, this instability occurs due to a competition between the combination of elastic and surface energies which acts as the stabilizing factor and the interaction energy with the contactor which is the destabilizing influence in the present problem. This may be contrasted to

instabilities in prestressed solid films studied hitherto [8] where the elastic energy is the destabilizing factor competing with the surface energy which inhibits surface roughening. Pattern formation in prestressed solid films can also occur due to nonlinear material behavior such as large deformation plasticity [9]. The key difference between our system and those cited above is that those instabilities are irreversible (due to diffusion, plasticity, etc.), while the present instability in interreacting thin films is an elastic one. In addition, the present instability occurs in soft films while those studied previously occur in much stiffer semiconductor/metal films.

The system considered here is shown in Fig. 1—a film of thickness h bonded rigidly to a rigid substrate described by coordinates (x_1, x_2) such that the surface of the film S interacting with external agency has $x_2 = 0$. We restrict attention to plane strain deformations of the film for the sake of mathematical simplicity and to understand the essential physics. The total potential energy of this system is

$$\int_V W(\boldsymbol{\epsilon}) dV + \int_S [\gamma \sqrt{1 + (u_{2,1})^2} - U(\mathbf{u} \cdot \mathbf{n})] dS, \quad (1)$$

where $\boldsymbol{\epsilon}$ is the strain tensor, $W(\boldsymbol{\epsilon})$ is the elastic strain energy density, γ is the surface energy, $U(\mathbf{u} \cdot \mathbf{n})$ is the interaction potential between the surface of the film and the external agency such as a contactor or an electric field, \mathbf{u} is

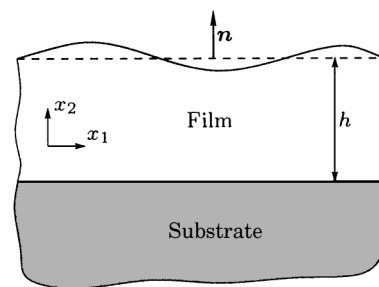


FIG. 1. Film bonded rigidly to a rigid substrate. The surface of the film experiences external forces.

the displacement vector, and \mathbf{n} is the outward normal to the surface. Linearized analysis is performed by expanding the interaction term $U(\mathbf{u} \cdot \mathbf{n})$ in a power series about $\mathbf{u} = \mathbf{0}$ and retaining all terms up to quadratic order in \mathbf{u} . The resulting approximate energy functional is

$$\int_V W(\boldsymbol{\epsilon}) dV + \int_S \gamma [1 + \frac{1}{2}(u_{2,1})^2] dS - \int_S [U_0 + F_0 \mathbf{u} \cdot \mathbf{n} + \frac{1}{2} Y (\mathbf{u} \cdot \mathbf{n})^2] dS, \quad (2)$$

where $U_0 = U(0)$, $F_0 = U'(0)$, and $Y = U''(0)$.

The equilibrium stress field $\boldsymbol{\sigma}$ in the film [which minimizes the potential energy (2) over an appropriate length of the film] satisfies the equilibrium equation $\nabla \cdot \boldsymbol{\sigma} = \mathbf{0}$ in V and the boundary condition

$$\boldsymbol{\sigma} \cdot \mathbf{n} = \gamma u_{2,1} \mathbf{n} + F_0 \mathbf{n} + Y (\mathbf{u} \cdot \mathbf{n}) \mathbf{n} \quad (3)$$

on S . Taking the film to be an isotropic linear elastic solid with shear modulus μ and Poisson's ratio ν gives a standard expression for the strain energy density [10]

$$W(\boldsymbol{\epsilon}) = \mu [\boldsymbol{\epsilon}_{ij} \boldsymbol{\epsilon}_{ij} + \nu (\boldsymbol{\epsilon}_{kk})^2 / (1 - 2\nu)] \quad (4)$$

with a resulting expression for the stress tensor expressed in terms of the gradient of displacement. Thus the problem can be cast into a boundary value problem for the unknown displacement field with the boundary condition of vanishing displacements at $x_2 = -h$ at the film substrate interface in addition to (3).

The homogeneous solution.—A solution to the above boundary value problem exists such that the stresses in the film are equal everywhere. This homogeneous solution (\mathbf{u}^h) is $u_1^h = 0$ everywhere, and u_2^h has a linear variation with x_2 starting from 0 at $x_2 = -h$, i.e.,

$$u_2^h(x_1, 0) = F_0 / (Y_m - Y), \\ Y_m = 2(1 - \nu)\mu / (1 - 2\nu)h. \quad (5)$$

For the case when $\nu = 0.5$, i.e., the incompressible limit, the homogeneous solution is such that the displacement vanishes everywhere in the film, and a pressure field p develops such that $p(x_1, x_2) = F_0$. So long as $Y < Y_m$, the homogeneous solution is meaningful in that $u_2^h(x_1, 0)$ has the same sign as F_0 . This condition on Y is most easily met when ν is close to 0.5 (Y_m tends to ∞ as ν tends to 0.5), i.e., when the material is nearly incompressible.

Bifurcations.—If an inhomogeneous bifurcation solution exists, it can be taken to be of the form $\mathbf{u}^h + \mathbf{u}$, where the symbol \mathbf{u} now stands for a ‘‘bifurcation’’ displacement field. This field must also satisfy the equilibrium equations in the bulk and the rigid boundary condition at the film substrate interface. On the surface of the film, the bifurcation field satisfies (here $\boldsymbol{\sigma}$ is the *additional* stress due to \mathbf{u}),

$$\boldsymbol{\sigma} \cdot \mathbf{n} = \gamma u_{2,1} \mathbf{n} + Y (\mathbf{u} \cdot \mathbf{n}) \mathbf{n}, \quad (6)$$

instead of (3). The bifurcation fields are assumed to have the form

$$u_j(x_1, x_2) = e^{ikx_1} u_j(x_2), \quad (7)$$

where k is a real positive wave number. The problem can now be cast into the problem of finding those values of k such that the functions $u_j(x_2)$ are nontrivial. It can be shown that (a detailed account will be published elsewhere) nontrivial bifurcation fields of the form (7) exist for those values of k that satisfy the equation

$$(k\{4e^{2hk}hk^2[h\mu - (1 - \nu)\gamma] + (e^{4hk} - 1)k\gamma(3 - 7\nu + 4\nu^2) + \mu[(3 - 4\nu)(1 + e^{4hk}) + 2e^{2hk}(5 - 12\nu + 8\nu^2)]\}) / \{(1 - \nu)[(3 - 4\nu)(e^{4hk} - 1) - 4hke^{2hk}]\} = Y. \quad (8)$$

This relation is valid for the incompressible case as well (i.e., when $\nu = 0.5$). Real roots of (8) are sought when $Y < Y_m$.

We first focus attention on the case when γ vanishes. Figure 2(a) graphically depicts the solution to (8); i.e., for a given value of ν , the values of k that solve (8) are plotted as a function of hY/μ . The important results may be noted as follows: (i) There are no bifurcation modes for any value of ν when $hY/\mu < 2$. (ii) For all values of ν , $k = 0$ is a bifurcation mode when $Y = Y_m$. (iii) When $\nu \leq 0.25$, there are no bifurcation modes for $Y < Y_m$. (iv) When $\nu > 0.25$, there are two modes starting from a critical value Y_c [such as the point C shown in Fig. 2(a)] that depends on the value of ν until Y reaches Y_m . When the film is incompressible, $hY_c/\mu = 6.22$ and the corresponding bifurcation mode has $hk_c = 2.12$. For this case bifurcations are possible for all values of Y greater than $6.22\mu/h$, with two possible values of k as shown in Fig. 2(a).

Next, we consider the case when $\gamma \neq 0$. Figure 2(b) shows a plot of the possible wave numbers of bifurcation modes for various values of γ with $\nu = 0.4$. The key effects of the surface energy on the bifurcation modes are noted as follows: (i) Surface energy inhibits bifurcation, in that a larger value of Y_c is affected with a nonzero value of γ . The critical mode k_c decreases with increasing γ . Both of these results are as expected since a larger value of k implies a larger energy penalty in terms of surface energy. (ii) As γ gets larger Y_c approaches Y_m . In fact, it can be shown that Y_c equals Y_m when $\gamma = \gamma_m$ where $\gamma_m/\mu h = 2\nu(4\nu - 1)/3(1 - 2\nu)^2$, a result which is pertinent when $\nu > 0.25$. The curve for $\gamma/\mu h = 4.0$ for the case of $\nu = 0.4$, shown in Fig. 2, graphically illustrates this point. If $\gamma > \gamma_m$, then there are no bifurcations in the physically meaningful range $Y < Y_m$.

An asymptotic analysis of (8) under conditions most relevant for elastomers ($\nu \rightarrow 0.5$ and $\gamma/\mu h \ll 1$) gives

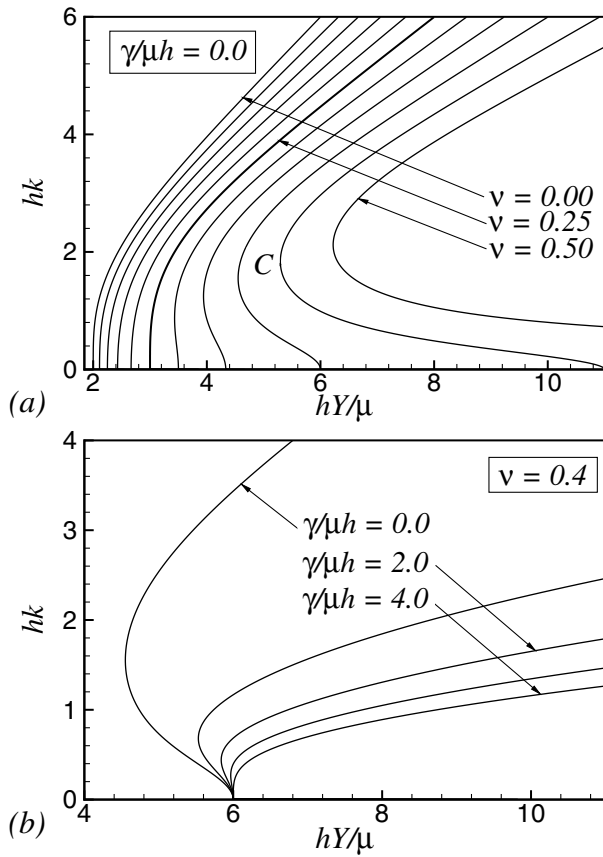


FIG. 2. (a) Bifurcation modes (hk) as a function of hY/μ for various values of ν with $\gamma/\mu h = 0$. (b) Bifurcation modes (hk) as a function of hY/μ for various values of $\gamma/\mu h$ with $\nu = 0.4$.

the following simple scalings for the critical force parameter and the wave number:

$$\begin{aligned} \frac{h}{\mu} Y_c &= 6.22 - 10.46(1 - 2\nu) + 4.49 \frac{\gamma}{\mu h}, \\ hk_c &= 2.12 - 2.86(1 - 2\nu) - 2.42 \frac{\gamma}{\mu h}. \end{aligned} \quad (9)$$

Clearly, the length scale of the instability is independent of the nature and magnitude of the interaction and elastic modulus of the film. For most elastomeric films of moderate thickness ($\nu \approx 0.5$, $\mu \approx 1$ MPa, $h > 1$ μm , and $\gamma = 0.1$ J/m²) the wavelength of the instability is of the same order as the film thickness and scales linearly with it, i.e., $\lambda_c = 2.96h$.

We now briefly consider modifications when the film is considered to be a viscoelastic solid with viscous stresses $\boldsymbol{\sigma}^v$ (in addition to elastic stresses) given by $\boldsymbol{\sigma}^v = 2\eta[\frac{1}{2}(\nabla\dot{\mathbf{u}} + \nabla\dot{\mathbf{u}}^T) - \frac{1}{3}\nabla \cdot \dot{\mathbf{u}}\mathbf{I}]$, where $(\dot{\cdot})$ stands for the time derivative, η is a viscosity parameter, and \mathbf{I} is the unit tensor. In the consideration of the time evolution of the system, inertial effects are neglected. The perturbations \mathbf{u} of the homogeneous solution are assumed to grow in the form $u_j(x_1, x_2, t) = e^{ikx_1} u_j(x_2) e^{\omega t}$ and satisfy equilibrium equations and boundary conditions. The solution for ω

indicates that for $Y_c < Y < Y_m$, all perturbation modes with wave numbers between the two bifurcation modes given by the elastic analysis are unstable; i.e., ω for these modes are positive. Indeed, there is a mode with wave number (k_m) between wave numbers of the two elastic bifurcation modes such that the rate of growth (ω) is a maximum. Just as in (9), an analytical result can be derived for k_m for small values of $\gamma/\mu h$, $\nu \rightarrow 0.5$, and $h(Y - Y_c)/\mu \ll 1$:

$$hk_m = hk_c + [0.39 \frac{\gamma}{\mu h} - 0.46(1 - 2\nu)] \frac{h}{\mu} (Y - Y_c). \quad (10)$$

The key result is that for an incompressible material, with $\gamma/\mu h \ll 1$, the critical viscoelastic mode is identical with the critical elastic mode obtained in Eq. (9).

The whole picture of stability and bifurcation in this system and its dependence on the nondimensional parameters (hY/μ , $\gamma/\mu h$, ν) is depicted in Fig. 3. Region I in Fig. 3 is where the homogeneous solution is unique and stable while the region marked III in the figure corresponds to the case when the homogeneous solution is ‘‘catastrophic,’’ in that the film jumps to make a uniform contact with the contacting surface. Physically, this is the preferred mode for highly compressible films subjected to strong attractive forces. Region II is the most interesting—this corresponds to nearly incompressible material behavior. In this region the homogeneous solution is unstable, with two possible elastic bifurcation modes.

Many material systems of scientific and technological importance in the area of friction and adhesion where the film is made of an elastomeric material (shear modulus between 0.1–10 MPa) in close proximity or ‘‘contact’’ (<25 nm) with a comparatively rigid contactor such as glass, diamond, or steel have properties which fall in the region II of Fig. 3. Particular examples of such a

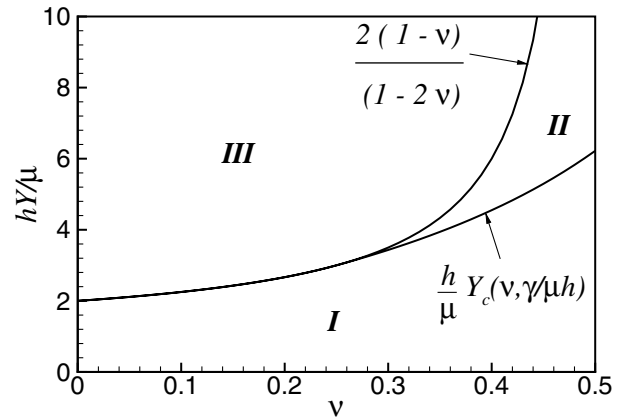


FIG. 3. The stable and unstable regions in the parameter space. Region I: Homogeneous solution stable. Region II: Homogeneous solution unstable with two elastic bifurcation modes and a fastest growing viscoelastic perturbation. Region III: Homogeneous solution is ‘‘catastrophic’’.

material system are those used in very recent experiments [4,5] which have observed and quantified the instabilities discussed in this Letter. The experiments in [4] used silicone elastomer (shear modulus 0.6–1 MPa and surface energy of 10 mJ/m²) for films and glass slides as the rigid contactor, some of which were silanized. The elastomeric films used in [5] were made of cross-linked polydimethylsiloxane (shear modulus 0.07–2.0 MPa, surface energy <0.1 J/m²) with glass as the contactor. The above works used films of thickness varying from 10–500 μm and found that the wavelength of the instability depends only on the thickness of the film and varies linearly with it.

We now turn to a quantitative discussion of the results of the above experiments [4,5] in comparison with our predictions. We consider a rigid contactor (glass) interacting with the film (elastomer) via van der Waals forces. Assuming that the contactor is at a distance d above the undeformed surface of the film, the van der Waals interaction U can be taken to be $U(\mathbf{u} \cdot \mathbf{n}) = A/12\pi(\mathbf{u} \cdot \mathbf{n} - d)^2$ with $F_0 = A/6\pi d^3$, $Y = A/2\pi d^4$. Taking the film to be made of an elastomer ($\mu = 1$ MPa, $\nu = 0.5$, $\gamma = 0.1$ J/m²) and $h = 10$ μm with $A \approx 10^{-19}$ J, the film becomes unstable for intersurface distance $d < 12.5$ nm since hY/μ exceeds its critical value 6.27. The wavelength of the instability in this case is about 30 μm. Experiments in [4] indicate that the relationship between the wave number of the instability and the thickness of the film is $hk_c = 2.3 \pm 0.3$ which is in excellent quantitative agreement with our prediction of $hk_c = 2.14$. Moreover, this relationship is not sensitive to type of surface (silanized versus unsilanized) used in the experiments—which corroborates with our prediction that the wavelength of the instability is independent of the nature of interaction as long as it exceeds a threshold value. Experiments in [5] with films contacting with a curved glass plate have $hk_c = 1.57$; this relationship is also found to be independent of the details of the interactions. The fact that the wave number depends inversely with the thickness is in agreement with our predictions, the proportionality constant is different owing to the fact that our analysis is based on a plane strain model while the experiments are three dimensional.

These instabilities can also be triggered in systems where a film interacts with an external electric field. Experiments of this type have been conducted recently [2] with liquid films. We present the discussion below in the hope that it will motivate experiments of a similar kind using solid films. These will not only serve to validate the theory but also take important steps towards the development of microscale pattern transfer technology discussed in [2]. The system considered consists of two plates separated by a distance d ; the bottom plate is coated

with a nearly incompressible elastomeric film of thickness h . A potential difference of V is applied between the two plates. The quantity of interest is the value of the gap thickness $d - h$ at which instability occurs in the film. Taking the interaction potential to be an electrostatic type [2] and the mechanical properties of the elastomer to be the same as in the previous case with a dielectric constant of 3, we have calculated that the critical gap thickness $d - h$ of 0.05 μm for a film of thickness 0.1 μm with the applied voltage of 100 V. A gap thickness smaller than 0.05 μm will cause the film to buckle.

The key finding in this Letter is that an instability in interacting soft films occurs which gives rise to a deformation pattern whose wavelength depends only linearly on the thickness of the film and not on the details of the interaction. Our results agree quantitatively with available experiments. These results have important implications in problems of adhesion, friction, and cavitation at soft interfaces.

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- [1] See, for example, S. Herminghaus *et al.*, *Science* **282**, 916 (1998); A. Sharma and R. Khanna, *Phys. Rev. Lett.* **81**, 3463 (1998); G. Reiter, R. Khanna, and A. Sharma, *Phys. Rev. Lett.* **85**, 1432 (2000).
- [2] E. Schäffer, T. Thurn-Albrecht, T. P. Russel, and U. Steiner, *Nature (London)* **403**, 874 (2000).
- [3] K. R. Shull, C. M. Flanigan, and A. J. Crosby, *Phys. Rev. Lett.* **84**, 3057 (2000).
- [4] W. Mönch and S. Herminghaus (to be published).
- [5] A. Ghatak, M. K. Chaudhury, V. Shenoy, and A. Sharma, *Phys. Rev. Lett.* **85**, 4329 (2000).
- [6] B. Z. Newby, M. K. Chaudhury, and H. R. Brown, *Science* **269**, 1407 (1995); R. J. Fields and M. F. Ashby, *Philos. Mag.* **33**, 33 (1976).
- [7] G. H. Fredrickson *et al.*, *Macromolecules* **25**, 2882 (1992).
- [8] R. J. Asaro and W. A. Tiller, *Metall. Trans.* **3**, 1789 (1972); M. A. Grinfeld, *Dokl. Akad. Nauk. SSSR* **283**, 1139 (1985); D. J. Srolovitz, *Acta Metall.* **37**, 621 (1989); B. J. Spencer, P. W. Voorhees, and S. H. Davis, *J. Appl. Phys.* **73**, 4955 (1993); L. B. Freund, *Acta Mech. Sin.* **10**, 16 (1994).
- [9] J.-C. Ramirez, *Int. J. Solids Struct.* **25**, 579 (1989), and references therein.
- [10] L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon Books Ltd., Oxford, England, 1989).