PHYSICAL REVIEW E VOLUME 59, NUMBER 3 MARCH 1999

Pattern formation during delamination and buckling of thin films

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(Received 23 October 1998)

Using a simple lattice model, we study the failure of compressively strained thin films on solid substrates. Our simulations reproduce much of the observed phenomenology of thin film blistering. In particular, we observe a peculiar form of stress relief in which a wrinkle of delaminated film propagates along a sinusoidal path. At higher intrinsic strains, the sinusoidal wrinkles bifurcate, forming branches. Finally, we identify a high-strain regime in which the film delaminates by forming a buckling front with many irregular lobes. [S1063-651X(99)50603-X]

PACS number(s): 68.55.−a, 46.32.+x, 47.54.+r

In a classic experiment, Yuse and Sano [1] demonstrated that a crack propagating along a thermal stress gradient undergoes a sequence of instabilities. In their work, a heated glass plate was lowered at speed v into a cold bath. A crack grew along the moving stress gradient at the same speed. At small values of v and for small thermal stress gradients, the cracks were straight. A transition from straight to regular, wavy cracks was observed at a critical speed v_c. At still higher values of v, period-doubling branching of the sinusoidal cracks occurred. A scaling analysis suggested that these morphological transitions are Hopf bifurcations. Sinusoidal cracking has now been studied extensively, and a variety of analytic calculations and numerical simulations have been brought to bear on this interesting example of pattern formation [2–5].

Sinusoidal cracking has an intriguing analog in compressive stress relief in thin films on solid substrates. As they are deposited, thin solid films often have considerable intrinsic internal stresses. Depending on the deposition conditions, the internal stresses can be either compressive or tensile [6,7]. They can also be large in magnitude, for example, compressive stresses in excess of 10^9 Pa have been observed in sputtered molybdenum films [8]. Frequently, the compressive stress is too large to be supported by the adhesive forces binding the film and substrate together, and part of the film lifts off the substrate. Buckling occurs in the delaminated portion of the film. The process of compressive stress relief is very rich and complex. In many instances, the delaminated regions have a sinusoidal shape behind a propagating tip. Because their width is usually on the order of 10 μm, these ''sinusoidal wrinkles'' are readily observed under a light microscope. They have been observed in many different kinds of thin films since they were discovered in the 1960's [9].

A variety of mechanisms have been considered to explain the propagation of sinusoidal wrinkles. Anisotropy in the stress was at one time thought to be essential to the phenomenon [10,11]. However, it has since been established that sinusoidal wrinkles propagate in films under isotropic stress [12]. Mixing of the three fracture modes at the edge of the delaminated region [13], gradients in the film thickness, inertial effects, and viscoelastic effects [14] could all conceivably be essential to an explanation of the phenomenon. Unfortunately, virtually the only experimental data currently available to guide the construction of a theory are micrographs of sinusoidal wrinkles. Under these circumstances, we believe that computer simulations of compressive stress relief could be particularly useful.

Recently, Gioia and Ortiz have made some progress toward a theory of sinusoidal wrinkles [15]. However, while their model provides a means of determining the interfacial fracture energy in a thin film which has partially delaminated, it does not explain the origin of sinusoidal wrinkles or account for many of the peculiar structural properties of sinusoidal wrinkles. A convincing explanation for the origin of sinusoidal wrinkles is therefore still lacking.

In this Rapid Communication, we perform simulations of delamination and buckling in a thin film initially under isotropic compressive stress applied by a rigid, planar substrate. We find that sinusoidal wrinkles occur spontaneously for intrinsic strains above a threshold value. They occur in the absence of inertial or viscoelastic effects, and with uniform film thickness and isotropic intrinsic stress, demonstrating that sinusoidal wrinkle propagation is a generic stress relief mode for thin solid films under compressive stress applied by the substrate. In our model, delamination occurs only through opening (mode I) fracture at the interface, and not through sliding or tearing. Thus, mode mixing is also not essential to the phenomenon. Finally, we find an empirical relation between the wrinkle wavelength and the intrinsic strain, and give a heuristic explanation for the origin of sinusoidal wrinkles.

Our simulations produce three distinct types of pattern formation. In addition to the growth of regular sinusoidal wrinkles, at higher strains the wrinkles bifurcate, forming branches and sub-branches reminiscent of the bifurcation of propagating sinusoidal cracks [1]. At still higher strains, the seed blister evolves into a complex, lobed buckling front. These types of pattern formation have been observed experimentally [12,16,17], although the intrinsic stresses in the experiments were unknown, and so it was not understood that the mode of stress relief selected changes as the strain is increased.

Our aim is to demonstrate the occurrence of sinusoidal wrinkles in the simplest possible model of an elastic film on a rigid substrate. In keeping with this goal, we model the film as two coupled triangular lattices of masses joined by linear elastic springs. In its unstrained state, the film's geometry is identical to two adjacent (111) planes of an fcc lattice. Accordingly, each mass has nine nearest neighbors, six of which are in the same (111) plane as the mass. This is the simplest arrangement of springs and masses that has nonzero bulk and shear moduli for in-plane motions, and that has a nonzero flexural rigidity.

Adhesion to the substrate is through contact forces that anchor each mass in the lower plane of the film to an impenetrable, rigid substrate with a planar surface. A mass in the lower plane of the film does not move until the normal force f applied to it by the remainder of the film exceeds a critical value f_c, and then the contact force is irreversibly set to zero. In this way, the film delaminates only by opening (mode I) fracture and not by sliding or tearing. There is therefore no mode mixing at the interface between adherent and delaminated portions of the film. In the absence of the top film layer, our model resembles a model proposed by Meakin to study fracture in thin films [18].

We take the sites in the film to be massless so that the lattice dynamics are purely dissipative. A film site i in the top layer of the film obeys the equation of motion

$$\eta\dot{\vec{x}}_i=\sum_{j\neq i}\vec{F}(\vec{x}_i-\vec{x}_j), \tag{1}$$

where η is a phenomenological damping coefficient and the sum on j is over nine nearest neighbors. The displacement from equilibrium for the site i is $\vec{x}_i$. The force term in Eq. (1) represents the Hookean interaction with "spring constant" k_{ij}, i.e., $\vec{F}(\vec{x}_i-\vec{x}_j)=-k_{ij}(\vec{x}_i-\vec{x}_j)$. Sites in the bottom layer of the film that have lost adhesion to the substrate also obey Eq. (1). The units in the simulations are chosen so that η, f_c, and the lattice spacing all have unit magnitude. The interplane bonds have a common spring constant k_0 and the intraplane bonds have spring constant k_1. For the simulations described here, $k_0/k_1=16/15$ [19].

Initially, the film is in equilibrium and all bonds have their common natural length l_0. A seed blister is generated at an edge of the film by breaking a small patch of adhesive bonds. The natural length of the bonds in the lattice is then incrementally increased by 0.05% of the initial value at each time step until a target value l_T is reached. In this way, compressive stress develops within the film. The intrinsic strain ϵ will be defined to be $(l_T-l_0)/l_0$. At some point, the internal stresses in the film cause the seed blister to extend as the normal forces on the film sites in the bottom layer adjacent to the blister exceed f_c. In all cases studied, this occurs after the target value of l_0 has been reached.

Seed blisters do not grow for strains below a critical strain $\epsilon_1=0.185$. For $\epsilon\geqslant\epsilon_1$, a variety of remarkable stress-relief forms emerge from the simulations. The most striking of these is the sinusoidal wrinkle, an example of which is shown in Fig. 1(a). For $\epsilon_1\leqslant\epsilon<\epsilon_2=0.195$, each seed blister gives rise to a single sinusoidal wrinkle which propagates through the lattice at a constant speed v. v is a linearly increasing function of ϵ for $\epsilon_1\leqslant\epsilon\leqslant\epsilon_2$.

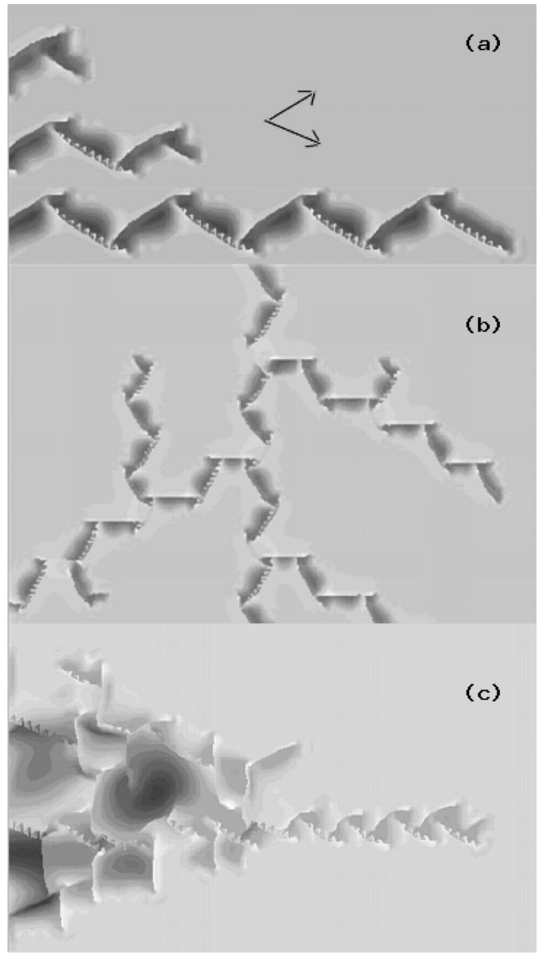

FIG. 1. (a) Single sinusoidal wrinkle at three stages in its growth from a simulation with $\epsilon=0.192$. The grey scale shows the film height, ranging from highest (dark) to lowest (light). The arrows indicate the principal lattice directions. The three snapshots have been drastically cropped. (b) A simulation with $\epsilon=0.196$ produces a wrinkle with multiple side-branches. The seed blister was at the base of the figure in this case. (c) A wrinkle that has spread laterally into a buckling front under a strain $\epsilon=0.230$.

The propagating tip leaves behind a quiescent, sinuous wrinkle with roughly constant lateral width. The wavelength of the wrinkle in Fig. 1(a) is constant along its length. It is important to note that the wrinkle wavelengths in the simulations are much larger than the lattice spacing, indicating that the wrinkles are not merely artifacts of the lattice geometry. The wrinkle shown in Fig. 1(a) has a wavelength of 25 lattice units, for example.

As observed in experiments, the wrinkle develops a sharply folded ridge [15]. The formation of a fold is not unexpected; indeed, much recent theoretical work has centered on singular folds in crumpled elastic sheets [20].

Through delamination and buckling, complete release of compressional strain energy along the ridge is achieved. In fact, the bonds along the ridge of a fully developed wrinkle carry a small tensile strain. In our simulations, the ridge does not lie midway between the edges of the wrinkle, but oscillates about the midline, just as observed experimentally [9].

The strain $\epsilon=\epsilon_2=0.195$ marks the onset of a distinct stress relief pattern in which the propagating sinusoidal wrinkle bifurcates, forming a side-branch that issues from a

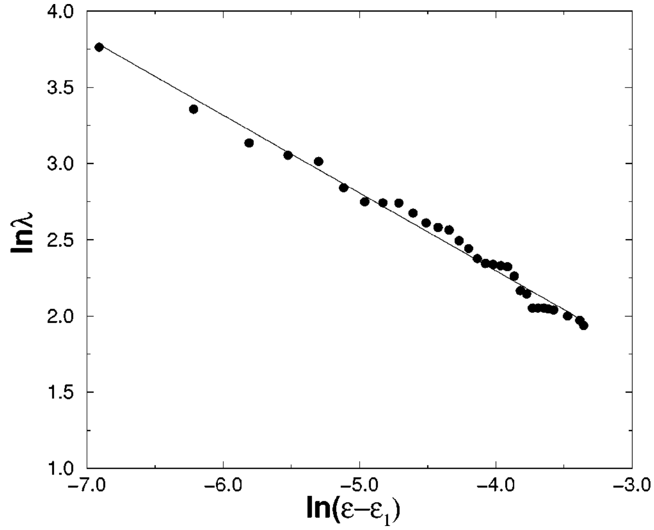

FIG. 2. λ vs ϵ is shown for $\epsilon_1 \leqslant \epsilon < \epsilon_3$. The solid line is a least-squares fit and has slope -0.51.

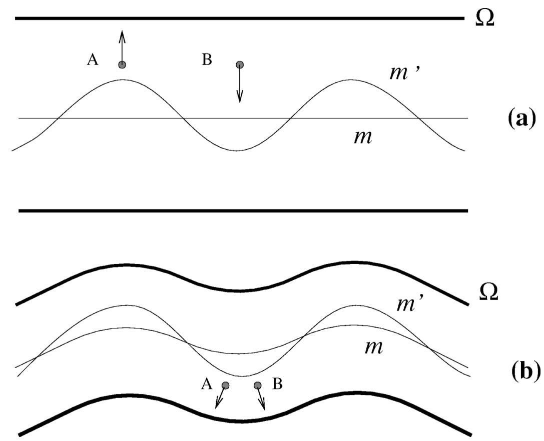

FIG. 3. (a) Straight-sided blister. The bold lines bound the region Ω which will lose adhesion with the substrate. An oscillating incipient ridge m' yields partial release of the strain in the direction parallel to the direction of propagation. The arrows show how the material points A and B will move upon delamination; note that the distance between them increases. (b) Improved strain energy release in the direction of propagation results if the edges of the delaminated region oscillate in phase with the incipient ridge m'.

turning point on the parent wrinkle. These side-branches form well behind the tip of the parent wrinkle and propagate away from it. For strains in excess of ϵ_2, additional branches appear and may themselves give rise to side-branches [Fig. 1(b)]. The side-branches have the same dimensions and propagate with the same speed as the parent branch. These side-branches are reminiscent of the branching of sinusoidal cracks seen in the experiments of Yuse and Sano [1].

For $\epsilon \geqslant \epsilon_3 = 0.230$, the internal stresses are too large to support propagation of sinusoidal wrinkles. Instead, the seed blister evolves into a complex, lobed buckling front that grows irregularly in all directions [Fig. 1(c)]. As has been observed in experiments [16], the buckling front often spawns one or more sinusoidal wrinkles [Fig. 1(c)]. At still higher values of ϵ ($\epsilon \geqslant 0.250$), large portions of the film delaminate.

It has been suggested that sinusoidal wrinkles may be a useful indicator of the intrinsic strain in the film prior to delamination [21]. Gioia and Ortiz have given heuristic arguments for a relation between wrinkle wavelength and intrinsic strain, $\lambda \propto \epsilon^{-1/2}$ [15]. Our simulations show that there is indeed a strong dependence of wrinkle wavelength on strain, similar to the form proposed by Gioia and Ortiz. Wrinkle wavelengths λ were obtained from Fourier transforms of the ridge line. Figure 2 shows a log-log plot of λ vs $\epsilon - \epsilon_1$. The overall trend of the data for strains ϵ ranging from ϵ_1 to ϵ_3 is consistent with a relationship of the form $\lambda \propto (\epsilon - \epsilon_1)^{-\beta}$ with $\beta = 0.51 \pm 0.04$. There is, however, additional structure in the data of Fig. 2. The periodic variations in slope are suggestive of the log-periodic corrections to power-law scaling proposed by Sornette and Sammis [22] in the context of critical phenomena, and by Sahimi and Arbabi [23] in the context of fracture.

There is a relatively simple heuristic explanation for sinusoidal wrinkle propagation. In particular, we will argue that the propagation of a sinusoidal wrinkle results in a greater release of elastic energy than a straight-sided wrinkle would. Consider a region of film Ω, which will lose adhesion with the substrate (Fig. 3). Let m denote the midline of the blister, so that the edges of Ω are equidistant from m. The ''incipient ridge'' m' is the curve that will become the ridge of the blister upon delamination. To begin, we shall assume that m is a straight line. The material points A and B lie on a line which, before delamination, is oriented along the direction of propagation. Clearly, a straight-sided wrinkle with a straight ridge would not relieve compressive strain along the direction of propagation; the distance between A and B would remain unchanged. Instead, the ridge of the wrinkle oscillates laterally as the wrinkle extends. In this way, the distance between A and B increases and, more generally, some of the compressive strain parallel to the direction of propagation is released [Fig. 3(a)]. There is a bending energy cost for the oscillation of the ridge, and this cost grows as the ridge nears an edge of the delaminated region. This keeps the amplitude of the oscillation bounded.

We will now dispense with the requirement that the edges of Ω be straight. As the ridge moves laterally, an additional release of compressional energy is made possible if the midline m also oscillates [Fig. 3(b)]. In this way, the oscillation of the ridge is echoed in the oscillation of the midline m and the boundary of Ω. The net result is an additional release of compressive strain in the direction parallel to the direction of propagation, a release that would not occur if the wrinkle's edges were straight.

While subject to strong lattice effects, the simulations are in excellent agreement with our heuristic explanation. In particular, a plot of m, m' and the wrinkle edges reproduce the phase relationships of Fig. 3 [24].

Our simulations provide several important clues which should aid in the development of a theory of sinusoidal wrinkle propagation. First, they strongly suggest that the formation of sinusoidal wrinkles is a *generic* stress relief mode for thin solid films under compressive stress applied by the substrate. This is consistent with the fact that sinusoidal wrinkles have been observed in many different kinds of thin films on a variety of substrates [15]. Mode mixity, viscoelastic effects, stress anisotropy, and film thickness gradients are not essential to the phenomenon and so may be omitted from a first theory. Since our simulations were carried out in the overdamped limit, sinusoidal wrinkles must form because they are the most efficient means of releasing the stored elastic energy in the compressed film. This observation is the underpinning of our heuristic explanation of sinusoidal wrinkle propagation, and we believe that it is the key to the development of a theory. A full theory could be tested by comparing with our empirical relation between the wrinkle wavelength and the strain, and will provide an explanation for wrinkle branching and buckling front propagation as well.

Although our work provides fresh insight into the propagation of sinusoidal wrinkles, the phenomenon remains incompletely understood more than four decades after its discovery. We hope that our simulations will stimulate new experimental and theoretical work on the fascinating types of pattern formation that occur during thin film delamination.

We would like to thank F. d'Heurle, M. P. Gelfand, J. M. E. Harper, D. Link, M. Mahadevan, and R. S. Robinson for helpful discussions.

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