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Undulated blistering during thin film delamination

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We study the delamination of a compressed thin film from a solid substrate with continuum elasticity theory. Our model enables one to describe the advance of an undulated blister. It is shown that an elastic tension between the inner fold and the boundary of a blister is at the origin of the undulations. In addition, the essential experimental observations and recent simulations are reproduced very well: (i) Above a strain threshold, straight blister growth is unstable and starts to undulate. (ii) It is found that the period of the undulations scales with $\epsilon^{*-1/2}$, where ϵ^* is the isotropic compressive strain of the film. (iii) Similar periodic corrections to this power law scaling are recovered and are found to be associated with a growth instability.

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Compressed thin films and coating can delaminate and buckle away from the substrate on which they are deposited to form blisters [1]. The compressive strain ϵ^*, responsible for this kind of damaging phenomenon, can have different origins [2]. It is generally due to the difference in thermal expansion between the layer and the substrate. In that case, the strain can reach the typical value $\epsilon^* \approx 0.01$ with an associated stress $\sigma^* \approx 1$ GPa [3]. In many cases, the delaminated part of the film adopts a sinusoidal shape behind a propagating tip. For 40 years, ''telephone-cord'' blisters have been met in many different kinds of coated structures (for a review, see [2]). It has been experimentally established that undulated blisters propagate in films under isotropic stress [4]. Very recently this was confirmed numerically by Crosby and Bradley [5], who performed simulations on a lattice model with microscopic debonding.

For large size blisters (in comparison to the film thickness h), the delaminated region, bounded by a wavy boundary, exhibits folding patterns [6]. An argument which is often used [7], but never really proven, is that during delamination, the boundary undulations accommodate folding thus creating an ordered structure like a telephone-cord blister. In this Rapid Communication, we show that the physical origin of this accommodation is the elastic tension (neglected so far and considered here as a perturbation) between the fold and the boundary of a blister. We use a model of continuum elasticity that gives a simple interpretation of the undulations: they help the tensile strain to relax. This is also coherent with the heuristic explanation given by Ref. [5]. A very recent paper of Audoly [8] argues that, through a stability analysis of a semi-infinite straight blister, an in-plane displacement field has to be taken into account. However, sharp folding of the film is absent in Audoly's considerations. Furthermore, an important aspect of our simpler model is its capability to describe the advance of an undulated blister, which is otherwise very difficult technically, by attempting to solve the nonlinear Föppl and von Kármán equations [8]. Our model also recovers the experimental phenomenology [2] as well as the simulations results obtained by Crosby and Bradley [5]: (i) Straight shaped blisters are unstable when the isotropic compressive strain ϵ^* is bigger than a threshold ϵ^*_c. (ii) It is found that the period of the oscillations scales with $\epsilon^{*-1/2}$. (iii) Similar periodic corrections to this power law scaling are recovered and are found to be associated with a growth instability.

We briefly recall the results of Ortiz and Gioia [2,7], which play a role of reference in this work. They show that while neglecting the in-plane displacement field, the height of the delaminated film $\zeta(\mathbf{r})$ obeys the Eikonal equation known in geometrical optics [9], $|\nabla\zeta| = [2(1+\nu)\epsilon^*]^{1/2}$, where ν is the Poisson's ratio and $\mathbf{r}=(x,y)$ represents the in-plane position. The delaminated film buckles away from the substrate with a constant slope $k=[2(1+\nu)\epsilon^*]^{1/2}$. Considering that $\epsilon^* \approx 0.01$ and $\nu \approx 0.3$, the slope is rather small, $k \approx 0.16$. $\zeta(\mathbf{r})$ is calculated via the Nadai's sand heap construction [10] which minimizes the bending energy (i.e., the number of folds):

$$\zeta(\mathbf{r}) = k d(\mathbf{r}), \tag{1}$$

where $d(\mathbf{r})$ is the smallest distance from the point $\mathbf{r}$, within the delaminated region, to the given boundary. The results fit the observations remarkably well [2,7].

Consider the blister drawn in Fig. 1. The boundary is formed by a straight part of length L with a semicircular end of radius ρ. By using Eq. (1), the delaminated region is entirely described by $\zeta(\mathbf{r})$: a straight fold is obtained along the x axis. The in-plane displacement field $\mathbf{u}(\mathbf{r})$ is totally neglected. With the latter assumption, this construction has the remarkable property of relaxing the blister elastic energy density (energy per surface unit) by reducing it to a constant $\mathcal{W}_b = \frac{1}{2} E h \epsilon^{*2}$ [7], E being the Young's modulus. The compressed laminated film has an energy density $\mathcal{W}_f = E h \epsilon^{*2}/(1-\nu)$. Thus, the energy released by delamination is

$$E_{delam} = S(\mathcal{W}_b - \mathcal{W}_f + \mathcal{G}_c) + (\ell_B + 2\ell_F)\mathcal{T}_0, \tag{2}$$

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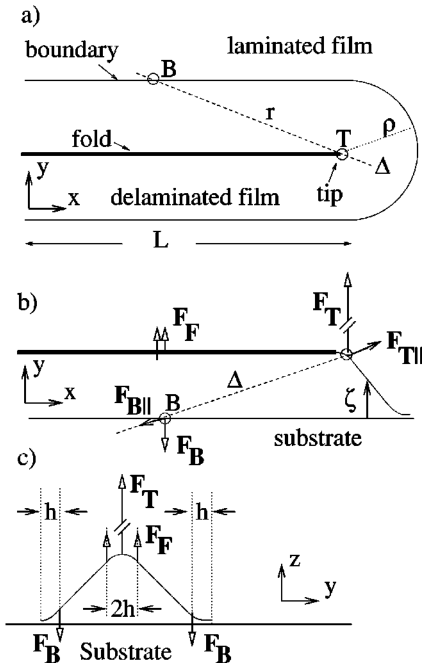

FIG. 1. Seed blister. (a) above view, (b) side view, and (c) front view. The bold line represents the fold obtained by Eq. (1). The slope k of the delaminated film and the ratio h/ρ are exaggerated for clarity.

where S is the surface of the blister, and ℓ_B and ℓ_F represent the length of the boundary and of the fold respectively. $\mathcal{G}_c$ is the fracture release energy due to delamination [7]. A pure mode I delamination (opening) is considered, and mode mixity is discarded [3,5,7]. $\mathcal{T}_0$ and $2\mathcal{T}_0$ are the respective boundary and fold line energy. For $\rho/h \gg 1$, we have [2,7]

$$\mathcal{T}_0 = \frac{Eh^2}{2(1-\nu^2)}\left[\tfrac{2}{3}(1+\nu)\epsilon^*\right]^{3/2}. \tag{3}$$

The growth is considered here as a purely dissipative phenomena. Thus, a straight growth along the x axis is allowed when $dE_{delam}/dL<0$, i.e., when $\rho>\rho_c$, for our seed blister we get $\rho_c = 2\mathcal{T}_0/(\mathcal{W}_f - \mathcal{W}_b - \mathcal{G}_c)$. Note that $\rho_c>0$ for $\mathcal{G}_c < \mathcal{W}_f - \mathcal{W}_b$.

By only considering the above energy E_{delam}, the blister does not undulate during its growth because of the cost in line energy. An additional energy of in-plane tensile strain origin, E_{tens}, must be added to E_{delam} in order for the blister to grow with undulations (which help in-plain tensile strain to relax). This elastic tension is due to the vertical forces (parallel to the z axis) $\mathbf{P}(\mathbf{r})$ located on the fold $\mathcal{F}$ and on the boundary $\mathcal{B}$, which tense the delaminated film. These forces can be calculated by using the Euler equilibrium equations for thin plates [11],

$$D\Delta^2\zeta - h\partial_\beta(\sigma_{\alpha\beta}\partial_\alpha\zeta) = P(\mathbf{r}). \tag{4}$$

The other equilibrium equation $\partial_\beta\sigma_{\alpha\beta}=0$ is always fulfilled thanks to Eq. (1). α and β represent the in-plane coordinates (x,y), $\partial_\alpha = \partial/\partial\alpha$. D is the bending stiffness, $D = Eh^3/[12(1-\nu^2)]$. The membrane stress is given by [7]

$$\sigma_{\alpha\beta} = \frac{Eh}{(1-\nu^2)}[(1-\nu)\epsilon_{\alpha\beta} + \nu\epsilon_{\gamma\gamma}\delta_{\alpha\beta}], \tag{5}$$

where $\epsilon_{\alpha\beta} = \frac{1}{2}\partial_\alpha\zeta\partial_\beta\zeta - \epsilon^*\delta_{\alpha\beta}$ is the membrane strain. Note that here, $\sigma_{\alpha\beta}$ only depends on the displacement along the z axis $\zeta(\mathbf{r})$. $\sigma_{\alpha\beta}$ should also depend on the in-plane displacement $u_\alpha(\mathbf{r})$; the latter dependence is neglected so far, consistent with Ref. [7]. Thanks to this approximation, $\mathbf{P}(\mathbf{r})$ is the force per surface unit created by the only out-of-plane displacement $\zeta(\mathbf{r})$ constructed with Eq. (1). $\mathbf{P}(\mathbf{r}) = \mathbf{0}$ everywhere exept on the fold and on the boundary, where the Eikonal equation is not satisfied. In order to have a differentiable field $\zeta(\mathbf{r})$, it is assumed [2] that near $\mathcal{F}$ or $\mathcal{B}$, $\zeta = \frac{1}{2}k\eta^2 + c$ for $\eta<h$, where η is the local normal coordinate to $\mathcal{B}$ or $\mathcal{F}$, c is a constant insuring $\zeta(\eta)$ continuity. As a consequence, the forces are mainly located within a distance h along $\mathcal{B}$ and $\mathcal{F}$.

The (x,y) plane is meshed on a 150×150 square lattice with a typical mesh size $\delta\ell = h$. We start with a seed blister (Fig. 1) with $\rho = 1.1\rho_c$ and $L = 5\delta\ell$. Below, we use $\mathbf{F}(\mathbf{r}) = \mathbf{P}(\mathbf{r})(\delta\ell)^2$, which is the force applied to the surface $(\delta\ell)^2$ of a mesh. Inside the blister boundary, the height $\zeta(x_i,y_i)$ of the mesh (x_i,y_i) is obtained via Eq. (1). This value is injected in the discrete forms of Eqs. (4) and (5) to calculate the applied force $\mathbf{F}(x_i,y_i)$. Along the fold a double set of forces $\mathbf{F_F}$ is obtained on each side of the fold. Each up-force $\mathbf{F_F}$ is counterbalanced by a down-force $\mathbf{F_B}$ at the boundary, $\mathbf{F_F} = -\mathbf{F_B}$ [Fig. 1(c)], resulting in a mechanical equilibrium. The up-forces $\mathbf{f_T}$, located on a semicircle of radius h around the tip of the fold, compensate the set of down-forces $\mathbf{F_B}$ located at the circular end of radius ρ of the boundary, hence $|f_T/F_B| = |f_T/F_F| \approx \rho/h$. As we consider thin layer $(\rho/h \gg 1)$, we assume that the main force applied on the fold is the one located at its tip $\mathbf{F_T} = \Sigma\mathbf{f_T}$ [12].

As mentioned above, in-plane displacement field u_α has been neglected so far. Now, u_α is free to relax under the action of $\mathbf{F}(\mathbf{r})$, resulting in a tensile strain between the fold and the boundary. Consider the line Δ joining the tip T and a point B located at the boundary [Figs. 1(a) and 1(b)], the distance between B and T is r. In our model $\zeta(\mathbf{r})$ creates $\mathbf{F}(\mathbf{r})$ via Eq. (4), hence ζ cannot in turn be affected by $\mathbf{F}$. As ζ is frozen, only u_α can be affected by the in-plane component $\mathbf{F}_\parallel$ ($\mathbf{F}$ projected onto Δ). The displacement field created in a very thin layer by an in-plane force $\mathbf{F}_\parallel$ at a distance $\mathbf{r}$ is known [13],

$$\mathbf{u} = \frac{(1+\nu)^2}{2\pi Eh}\left(\frac{\nu-3}{1+\nu}\mathbf{F}_\parallel \ln(r/h) + \frac{\mathbf{r}(\mathbf{r}\cdot\mathbf{F}_\parallel)}{r^2}\right). \tag{6}$$

The two opposite in-plane forces $\mathbf{F_{B\parallel}}$ and $\mathbf{F_{T\parallel}}$ applied in B and T tense the blister film, and the associated tensile energy [13] between the tip and a portion $\delta\ell$ of the boundary is

$$\mathcal{E}_{tens}\delta\ell = -\tfrac{1}{2}[\mathbf{F_{T\parallel}}\cdot(\mathbf{u_T^T} + \mathbf{u_T^B}) + \mathbf{F_{B\parallel}}\cdot(\mathbf{u_B^B} + \mathbf{u_B^T})]. \tag{7}$$

The subscript of $\mathbf{u}$ refers to the point where $\mathbf{u}$ is estimated, and the superscript denotes which force creates $\mathbf{u}$. $\mathcal{E}_{tens}\delta\ell$ can be divided into two components: a self-energy $\mathcal{E}_{self}\delta\ell = -\frac{1}{2}(\mathbf{F_{T\parallel}}\cdot\mathbf{u_T^T} + \mathbf{F_{B\parallel}}\cdot\mathbf{u_B^B})$ as if the two forces were alone, and an interaction energy $\mathcal{E}_{int}\delta\ell = -\frac{1}{2}(\mathbf{F_{T\parallel}}\cdot\mathbf{u_T^B} + \mathbf{F_{B\parallel}}\cdot\mathbf{u_B^T})$. $\mathcal{E}_{self}$

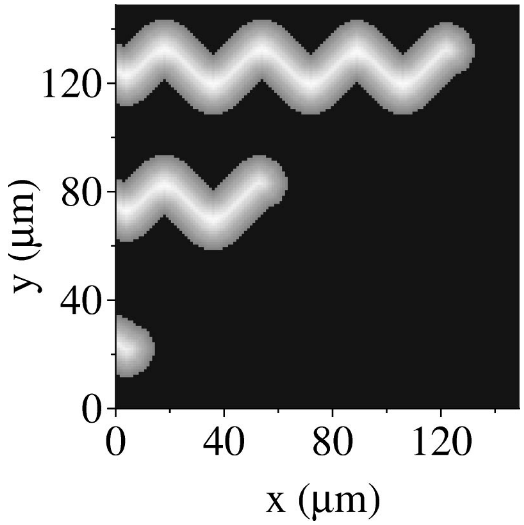

FIG. 2. A blister is represented at three different stages of its growth, the strain is $\epsilon^*=0.0085$. On the gray scale, the sharp fold (the highest values of ζ) is represented with white color. The laminated part of the film is black colored. The selected directions of growth are selected by the square meshing.

is a constant which shifts $\mathcal{E}_{tens}$, resulting in an additional line tension. In order to avoid divergence, $\mathbf{u_T^T}$ and $\mathbf{u_B^B}$ are arbitrarily estimated at $r=h$. Actually, this cutoff is not very important, it has been found that the typical wavelength of the undulations is not affected while $\mathcal{E}_{self} \ll \mathcal{T}_0$. By integrating along the boundary, we obtain the tensile energy between the fold-tip and the whole boundary,

$$E_{tens}=\int_{\ell_B}\delta\ell\,\mathcal{E}_{tens}=\int_{\ell_B}\delta\ell\,(\mathcal{E}_{int}+\mathcal{E}_{self}). \tag{8}$$

Thus, the total elastic energy E_{el} released by blistering is $E_{el}=E_{delam}+E_{tens}$, where the new term E_{tens} represents the driving force of the telephone-cord tailoring.

Let us now sketch our numerical procedure. The tip of the fold, located in the mesh (x_t,y_t) at step t, visits the four meshes $(x_t\pm\delta\ell, y_t\pm\delta\ell)$ at step $t+1$, leading to at most four different possible longer folds $\mathcal{F}_n$. Note that any fold overlap is forbidden, hence $n\leqslant 4$. For each possible fold, we reconstruct the blister around it by inverting Eq. (1) [14]. We calculate the energy $E_{el}^{(n)}(t+1)$ associated with the nth possible fold. The selected fold is the one for which $E_{el}^{(n)}(t+1)$ is minimum. We continue the same procedure at step $t+2$ and so on. If for a given step t, $E_{el}^{(n)}(t+1)>E_{el}^{(n)}(t)$ the blister growth is stopped. At each step t, the discrete force field $\mathbf{F}(x_i,y_i)$ is calculated as mentioned above and the mechanical equilibrium state is reached. Experimentally, this equilibrium is achieved through elastic relaxation, on a time scale associated with that of the sound speed ($\sim$1 ms), much smaller than the time associated with the blister propagation [$\sim$hour(s)] [2]. It allows us to consider that the blister is, at each step of its growth, in a mechanical equilibrium. Then, each force is projected as schematically illustrated in Fig. 1. $E_{el}(t)=E_{delam}(t)+E_{tens}(t)$ is straightforwardly calculated via Eq. (2) for E_{delam} and Eqs. (6)–(8) for E_{tens}.

It is found that above a strain threshold ϵ_c^*, a straight shape is no longer stable and the blister begins to undulate

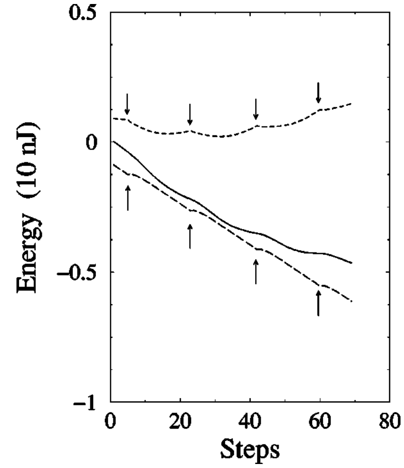

FIG. 3. Energy variation during the growth. Dotted line, E_{tens}; dashed line, E_{delam}; solid line, E_{el}. Arrows indicate changes of direction. The strain is $\epsilon^*=0.0085$.

(Fig. 2) with a regular wavelength. It comes out from our simulations that $\epsilon_c^*\approx 0.006$ for the chosen parameters: $E=10^{11}\,\mathrm{J/m^3}$; $\nu=0.3$; $G_c=0.1\,\mathrm{J/m^2}$ [15] and $h=1$ μm. Figure 3 shows how energy variation is affected during the blister growth. At each change of direction a cusp is obtained. λ is the length between two consecutive cusps. Each cusp corresponds to a sudden increase and decrease of the variations of E_{delam} and E_{tens} respectively: tension is partially relaxed by direction changes. If the variation of E_{tens} is bigger than the variation of E_{delam} (i.e., if $\epsilon^*>\epsilon_c^*$), it leads to a lowering of the total energy E_{el}, contributing to the creation of undulations.

λ is plotted in Fig. 4 in function of $\epsilon^*-\epsilon_c^*$ for $\epsilon^*>\epsilon_c^*$. A scaling power law with a $-\frac{1}{2}$ exponent is recovered (Fig. 4). This result is compatible with Refs. [2] and [5]. In addition, obvious periodic corrections to the power law are observed consistently with Ref. [5]. A careful inspection of the tip of the fold shows that at each direction change its propagation becomes unstable: the tip hesitates between two directions leading to a narrow zigzagging ($\sim$1 mesh thick) of the

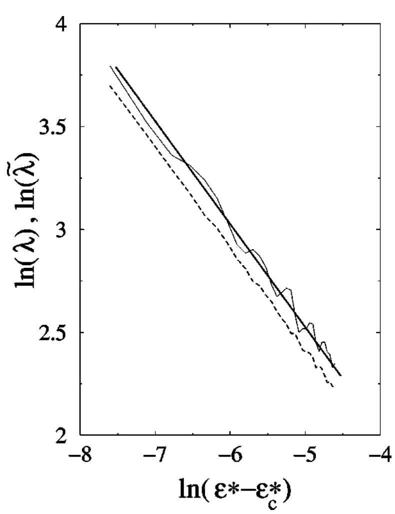

FIG. 4. Strain dependence of λ (solid line) and $\tilde{\lambda}$ (dashed line). The straight line indicates $\sim -\frac{1}{2}$ slope. $\epsilon_c^*\approx 0.006$. λ and $\tilde{\lambda}$ are in μm, and they are averaged on ten periods of blister undulations.

fold. The zigzag is not visible on Fig. 2, because it is averaged by the blister reconstruction [14]; that is why it has also no effect on energy curves (Fig. 3). This instability occurs on a length Λ of the order of a few meshes. It has been checked, by refining the meshing, that Λ is not due to a numerical instability. It is found that Λ depends on $\epsilon^* - \epsilon_c^*$. In Fig. 4, it is clear that $\tilde{\lambda} = \lambda - \Lambda$ obeys a power law with a $-\frac{1}{2}$ exponent without oscillation. It means that oscillations are due to the term Λ, i.e., to the growth instability. However, the origin of this instability remains an open question; it could be related to the one observed in Ref. [8].

In this paper we have demonstrated that the tensile strain between the tip of the fold and the boundary of a blister is the driving force of the undulated delamination growth. The results are coherent with experimental and numerical published works. Our model gives a deeper insight into periodic corrections: in the delamination case it appears that they are associated with a growth instability. However, a more specific study should be carried out to give a general explanation for all fracture problems [5,16], where these periodic corrections occur. We also believe that the calculations presented here could be extended to bifurcation growth and therefore to blisters with several folds.

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[1] J.W. Hutchinson and Z. Suo, Adv. Appl. Mech. **2**, 215 (1991).
[2] G. Gioia and M. Ortiz, Adv. Appl. Mech. **33**, 119 (1997).
[3] J.W. Hutchinson, M.D. Thouless, and E.G. Liniger, Acta Metall. Mater. **40**, 295 (1995).
[4] K. Ogawa *et al.*, Jpn. J. Appl. Phys. **25**, 695 (1986).
[5] K. Crosby and R. Bradley, Phys. Rev. E **59**, R2542 (1999).
[6] A.S. Argon *et al.*, J. Mater. Sci., Polym. Ed. **24**, 1207 (1989); Mater. Sci. Eng., A **107**, 41 (1989).
[7] M. Ortiz and G. Gioia, J. Mech. Phys. Solids **42**, 531 (1994).
[8] B. Audoly, Phys. Rev. Lett. **83**, 4124 (1999).
[9] *Principles of Optics*, edited by M. Born and E. Wolf (Pergamon Press, New York, 1993).
[10] A. Nadai, *The Theory of Flow and Fracture of Solids* (McGraw-Hill, New York, 1950).
[11] L.D. Landau and E. Lifchitz, *Theory of Elasticity* (Mir, Moscow, 1991).
[12] We have checked that the influence of the elastic interaction between the forces $\mathbf{F_F}$ along the fold and $\mathbf{F_B}$ along the boundary is indeed negligible while the ratio $\rho/h \geqslant 5$. However, below this value Eq. (1) is no longer valid and the sharp inner fold is absent. In that case, an analysis like [8] is preferable.
[13] P. Peyla, A. Vallat, C. Misbah, and H. Müller-Krumbhaar, Phys. Rev. Lett. **82**, 787 (1999).
[14] We construct disk of radius $\zeta(x_{fi}, y_{fi})/k$ centered on each mesh (x_{fi}, y_{fi}) of the fold. The envelop of all the disks is the boundary of the blister.
[15] G. Gioia and M. Ortiz, Acta Mater. **46**, 169 (1998).
[16] M. Sahimi and S. Arbabi, Phys. Rev. Lett. **77**, 3689 (1996).