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Mode-dependent toughness and the delamination of compressed thin films

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Abstract

We consider the buckle-driven delamination of compressed thin films. For a wide class of patterns of delamination, it is shown that the loading on the delamination front progressively goes from mode I to mode II during growth of the blister. As a result, the mode dependence of the film/substrate interface excludes widespread delamination. This explains the observations of blisters of finite extent, which are otherwise difficult to interpret. We also study a model of interfacial fracture with friction. It reveals that a severe mode dependence can be induced by interfacial friction. This permits us to account for the mode dependence using only simple ingredients: friction and linear elasticity. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Coated materials have many applications in industry: coatings permit one to obtain wear-resistant metal-cutting tools, thermal barriers in the aircraft and automobile industry, insulating layers in microelectronics, etc. The reliability of coated materials has become a growing field of interest in the recent past. In this paper, we are concerned with a particular mode of failure of these materials, the buckle-driven delamination. Coated materials are often obtained by vapor deposition of a thin film on a substrate at high temperature. The film and the substrate are made of different

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materials, and because of the mismatch of thermal expansion coefficients between them, the film acquires a biaxial residual stress σ_0 upon cooling. When this residual stress is compressive ($\sigma_0<0$) and large enough, an elastic instability may take place: to release its compression, the film tends to lift off the substrate, fracturing the interface (Nir, 1984; Argon et al., 1989). This instability results from a competition between the elasticity of the film, and the cohesion of the interface. Complex patterns of delamination have been observed as telephone-cord blisters (Gille and Rau, 1984). The mechanical conditions that lead to rupture of the interface are not well understood yet, although some progress has been made towards the understanding of these patterns (Jensen, 1993; Audoly, 1999).

The ability of the interface to sustain a certain loading without fracturing is called its toughness. This quantity has been measured in experiments for a variety of interfaces. The interface toughness is systematically enhanced when the loading on the crack tip goes from mode I to mode II (Wang and Suo, 1990; Liechti and Chai, 1992): interfaces are more easily fractured when the faces are pulled apart than when sheared. This property is called the mode dependence of the toughness (we shall sometimes refer to it simply as the "mode dependence"). Previous theoretical studies by Whitcomb (1986), Hutchinson et al. (1992) and Hutchinson and Suo (1991) suggest that the mode dependence of the film/substrate interface is essential to account for the patterns of delamination. In Section 2 of the present paper, we explore this idea further by studying the patterns of delamination that are obtained when the toughness of the interface is assumed to be mode-dependent. In particular, we show that the existence of blisters of finite size can be accounted for. Indeed, as a blister gets bigger, the fracture becomes more and more a mode II crack. In Section 3, we show that the mode dependence in an interface crack can be explained using only simple ingredients: friction and linear elasticity. We derive an effective interface toughness when friction between the crack faces is modeled with a Coulomb law (constant proportionality factor between shear and normal contact stresses). A strong mode dependence obtained, due to interfacial friction.

For the sake of simplicity, we consider only *quasi-static* propagation of cracks. Moreover, we neglect the effect of transverse (mode III) loading on the cracks, and use two-dimensional (2D) elasticity. A justification for this approximation is given at the end of Section 2. The toughness of the interface, Γ, is defined as the energy that must be brought to the crack tip to fracture a unit area, and has the dimension of a surface tension. The mode-mixity parameter, ψ, measures the relative importance of mode I (opening) to mode II (shearing) at the crack tip: $\psi=\tan^{-1}(K_{II}/K_I)$, where K_I and K_{II} are the stress intensity factors (Rice, 1988). The mode dependence implies that Γ not only depends on the nature of the interface, but is also a function of the applied loading, through ψ. Note that this parameter is taken in the range $-180°<\psi<180°$, as this is the phase in the (K_I, K_{II}) plane. When $|\psi|\geq 90°$, the opening stress intensity factor, K_I, is negative and the crack tends to close. This situation, which we shall consider in Section 3, arises when the loading presses the crack faces against each other. The definitions of K_I, K_{II} and ψ are then somewhat arbitrary, because the standard crack analysis yields overlap of the crack faces.

2. Stability of delamination blisters

In this section, the mode-dependent toughness of the interface crack is assumed; we investigate what patterns of delamination can be stable in the presence of mode dependence. If the interface has been cracked over some region but the film has not buckled, then the film remains uniformly compressed and the crack front is unloaded: the loading on the interfacial crack tip arises from the buckling of the film. Moreover, the buckled configuration of the film depends on the geometry of the delamination front. This shows that buckle-driven delamination couples two problems: the elastic deformation of the film and the propagation of the interface crack. The buckling of the debonded portion of the film is governed by the Föppl–von Kármán equations for elastic plates. The propagation of the interface crack is approached using the concept of energy release rate. This quantity, G, is defined on the crack front using the Rice integral (Rice, 1968a,b) along a vanishingly small transverse contour. Like the toughness of the interface, Γ, the energy release rate, G, has the dimension of a surface tension. It measures the intensity of loading, as it is the energy available at the crack tip to break interfacial bonds per unit area of advance of the crack. A standard propagation criterion for the crack is $G \geq \Gamma(\psi)$ (becoming an equality for quasi-static propagation). The quantities G and ψ shall be determined by solving the equations of elasticity for the film and substrate. The dependence of Γ on ψ expresses the mode dependence of the interface toughness: Γ increases for large $|\psi|$. The geometry of a blister is presented in Fig. 1. We call E, h and ν the Young's modulus,

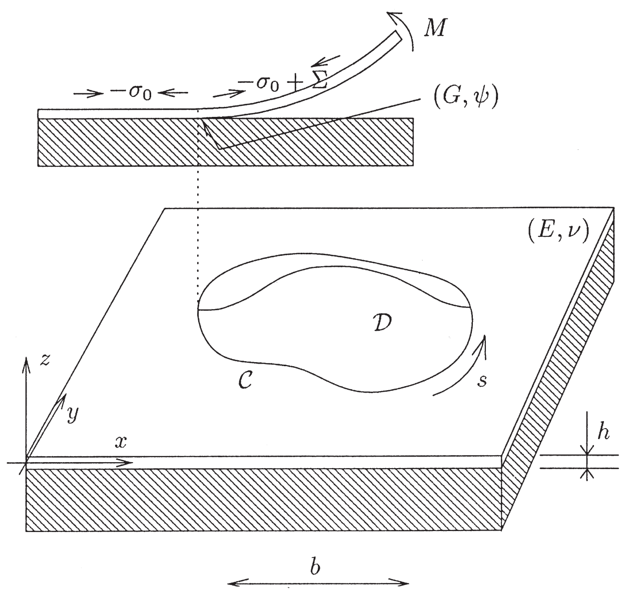

Fig. 1. Geometry of a generic 2D blister. The insert above shows a vertical section around the crack front. A discontinuity of normal stress in the film, Σ, is induced by the buckling.

thickness and Poisson's ratio of the film, respectively; b is the characteristic size of the blister.

2.1. Known results for one-dimensional (1D) blisters

One-dimensional patterns of delamination, i.e., circular or straight-sided blisters, have been studied previously. To prepare the study of more general patterns in Section 2.2, we first recall how 1D blisters are approached and how their stability is explained (see Evans and Hutchinson, 1984; Hutchinson et al., 1992 for the circular blister, and Chai et al., 1981; Gille and Rau, 1984 for the straight-sided one). The Föppl–van Kármán equations for the straight-sided blister (also called the Euler column) can be solved exactly. Those for the circular blister can be reduced to ordinary differential equations, and a weakly non-linear analysis can be carried out analytically. In both cases, the critical stress with clamped boundary conditions, σ_c, predicted by the classical buckling theory reads:

$$\sigma_c = \frac{k^g E}{1-\nu^2}\left(\frac{h}{b}\right)^2, \tag{1}$$

where b is half the width and $k^g = \pi^2/12$ for the straight-sided blister; or b is the radius and $k^g = 1.223$ for the circular blister (Timoshenko and Gere, 1961). All quantities that depend on the geometry of the blister are labeled by a superscript "g". Using σ_c, it is convenient to define a dimensionless buckling parameter, η, as:

$$\eta = \frac{-\sigma_0}{\sigma_c} - 1 = \frac{1-\nu^2}{k^g}\left(\frac{-\sigma_0}{E}\right)\left(\frac{b}{h}\right)^2 - 1. \tag{2}$$

The buckling instability takes place when the residual stress in the film is sufficiently compressive: $\eta > 0$, i.e. $\sigma_0 < -\sigma_c$.

Knowing the buckled configuration of the film, one can calculate two quantities of particular interest, M and Σ; these two quantities determine the loading on the crack tip and therefore govern the growth of the blister. M is the moment per unit length transmitted across the edge of the blister, and Σ is the normal stress *change* across this edge ($\Sigma = \sigma_{nn} - \sigma_0$, where σ_{nn} is the in-plane stress on the inner boundary of the blister, perpendicular to the boundary) — see Fig. 1. In 1D blisters, these quantities take the form:

$$M = c_1^g(\nu)\frac{Eh^4}{b^2}\eta^{1/2}, \quad \Sigma = c_2^g(\nu)\frac{Eh^2}{b^2}\eta, \tag{3}$$

where $c_{1,2}$ are positive *pure numbers* of order unity, which depend on the Poisson's coefficient of the film, ν, and on the geometry of blister, as indicated by the superscript "g".

The profile of the blister determines how the film is pulling on the interface crack. Suo and Hutchinson (1990) indeed give the crack-tip loading (the energy release rate, G, and the mode-mixity parameter, ψ) in terms of the so-called edge loads, Σ and M:

$$G=\frac{6(1-\nu^2)}{Eh^3}\left(M^2+\frac{1}{12}(h^2\Sigma)^2\right)=\frac{6(1-\nu^2)Eh^5}{b^4}\left([c_1^g(\nu)]^2\eta+\frac{1}{12}[c_2^g(\nu)]^2\eta^2\right) \tag{4a}$$

and

$$\begin{aligned}\psi &= -90^\circ+\omega(E/E_s,\nu,\nu_s)-\tan^{-1}\left(\frac{\Sigma h^2}{M\sqrt{12}}\right)\\ &= -90^\circ+\omega(E/E_s,\nu,\nu_s)-\tan^{-1}[c_3^g(\nu)\eta^{1/2}],\end{aligned} \tag{4b}$$

where ω is an angle in the range $50^\circ<\omega<65^\circ$ depending on the elastic constants of the film and substrate, E_s is the Young's modulus and ν_s the Poisson's ratio of the substrate, and $c_3^g=c_2^g/(c_1^g\sqrt{12})$. The angle ω is a constant of the interface, and does not depend on the shape or size of the blister.

The mode dependence is essential in interfacial fracture, as was seemingly first appreciated by Whitcomb (1986) in his study of compressive failure modes in composites. Indeed, let us consider the growth of a 1D blister of delamination at constant residual stress σ_0. We shall not discuss the initiation of the growth, which involves defects on the interface. As the blister grows, its size b increases, and so does η by Eq. (2). Eq. (4a) then shows that $G(b)$ increases with b (note that, by symmetry, G is uniform along the crack front in one dimension). Assume that the blister reaches an equilibrium size, b_{eq}; this size is such that the crack front is in equilibrium: $G(b)=\Gamma[\psi(b)]$. In the absence of a mode-dependent interface toughness ($\mathrm{d}\Gamma/\mathrm{d}\psi\equiv 0$), any fluctuation of b *above* b_{eq} would make G larger than Γ, so that the crack is unstable and b becomes even larger. This would indicate an instability of 1D blisters leading to full delamination of the film, contrary to experimental observations.

By the paragraph above, the mode dependence is necessary to explain the stability of 1D blisters. A simple argument shows that it is also sufficient. Indeed, the mode-mixity parameter, ψ, becomes more and more negative as the blister grows. This is because, in Eq. (4b), ψ depends on the size of the blister only through the term $-\tan^{-1}[c_3^g(\nu)\eta^{1/2}]$, and c_1, c_2 and so c_3 are positive. Typically, $\psi\approx -40^\circ$ at the buckling threshold ($\eta\ll 1$) reaches values of order $\psi\approx -90^\circ$ well above the buckling threshold $\eta\sim 1$ (Hutchinson et al., 1992). As mentioned above, the interface toughness, $\Gamma(\psi)$, is the minimum value of G that permits steady propagation of the crack; it is mode-dependent, and increases strongly for large absolute values of the mode-mixity parameter, $|\psi|$. This can explain the arrest of delamination of 1D blisters at a finite size: the blister pulls more and more strongly on the interface crack as it spreads (G increases) but, above all, less and less efficiently (Γ increases due to the mode dependence of the interface).

2.2. Extension of 2D blisters

Section 2.1 above suggests that the mode dependence is essential to explain the stability of blisters observed in experiments. However, a strong limitation of this argument is that only circular or straight-sided blisters have been considered. The question arises whether the proposed mechanism is specific to 1D geometries, or if it can stabilize blisters of any shape. Below, we show that a much broader class of patterns of delamination is in fact stabilized by the *same* mechanism. Indeed, we consider all blisters that have a single typical length scale. More accurately, if b is the typical extent of the blister, we assume that the curvature of its boundary is nowhere much larger than $1/b$. This class contains in particular circular and straight-sided blisters but also, more interestingly, a wide number of 2D, eventually non-convex, patterns. Telephone-cord blisters are however excluded, because their length is much larger than their width.

The study of 2D blisters (i.e., blisters that are neither circular nor straight-sided) is made difficult by the absence of analytical solutions to the non-linear Föppl–von Kármán equations when the boundary conditions are on an arbitrary curve $\mathcal{C}$. Even a numerical approach leads to overwhelming difficulties, owing to the presence of strong non-linearities in these equations (Patricio and Krauth, 1997). These difficulties are overcome, because we adopt a scaling law approach. To prove that the mode dependence excludes widespread delamination of 2D blisters, we proceed in several steps. First, we use energy arguments to show that a 2D blister always buckles by a supercritical bifurcation (i.e., the vertical deflection in the buckled configuration is vanishingly small just above threshold, for $\eta \ll 1$). This allows one to perform, in a second step, a weakly non-linear analysis of the buckling of a blister; the dependence of ψ on the blister size in Eq. (4b) is extended later to generic blisters. This permits one to establish that, for 2D as well as for 1D blisters, $|\psi|$ increases during growth of the blister; in consequence, the mode dependence induces a progressive toughening of the interface, which can prevent full delamination of the film.

2.2.1. *The Föppl–von Kármán equations*

Let $\mathcal{D}$ be the region where the film has debonded from the substrate, and $\mathcal{C}$ the boundary of $\mathcal{D}$, i.e., the edge of the blister. We consider the buckling of the film in $\mathcal{D}$. For a simple description, we take the unperturbed plane of the film horizontal (gravity has no effect). The profile of the blister is parametrized by the vertical displacement $\zeta(x, y)$ as a function of the horizontal coordinates (x, y). The tangential components of the stress derive from the Airy potential, $\chi(x, y)$: $\sigma_{xx}=\sigma_0+\chi_{,yy}$, $\sigma_{yy}=\sigma_0+\chi_{,xx}$ and $\sigma_{xy}=-\chi_{,xy}$, where the subscript comma indicates derivation. We recall that σ_0 is the initial compression of the film. The elastic energy functional for the thin film reads (Ben Amar and Pomeau, 1997):

$$E_{\mathrm{FvK}}\{\zeta, \chi\} = \iint_{\mathcal{D}} \mathrm{d}x\, \mathrm{d}y \left\{ \frac{D}{2}\{(\Delta\zeta)^2 - 2(1-\nu)[\zeta, \zeta]\} + \frac{h}{2E}(\Delta\chi + 2\sigma_0)^2 - \frac{h(1+\nu)}{E}([\chi, \chi] + \sigma_0\Delta\chi + \sigma_0^2) \right\}, \tag{5}$$

where $D=Eh^3/[12(1-\nu^2)]$, Δ is the 2D Laplacian operator, and $[f, f]=f_{,xx}f_{,yy}-f_{,xy}^2$. Minimization of this energy with respect to functions ζ and χ yields the equilibrium equations for the film, the Föppl–von Kármán equations (Landau and Lifschitz, 1986b):

$$\Delta^2\chi+E\left\{\frac{\partial^2\zeta}{\partial x^2}\frac{\partial^2\zeta}{\partial y^2}-\left(\frac{\partial^2\zeta}{\partial x\partial y}\right)^2\right\}=0 \tag{6a}$$

and

$$D\Delta^2\zeta-h\sigma_0\Delta\zeta-h\left(\frac{\partial^2\chi}{\partial y^2}\frac{\partial^2\zeta}{\partial x^2}+\frac{\partial^2\chi}{\partial x^2}\frac{\partial^2\zeta}{\partial y^2}-2\frac{\partial^2\chi}{\partial x\partial y}\frac{\partial^2\zeta}{\partial x\partial y}\right)=0. \tag{6b}$$

Because the substrate is infinitely thick, the film may be considered as clamped along its edge $\mathscr{C}$. In order to enforce the corresponding boundary conditions, the horizontal components of the displacement, (u_x, u_y), are needed. They have been eliminated in the Föppl–von Kármán equations, but can be recovered from the following relations (Landau and Lifschitz, 1986b), which are compatible by Eq. (6b):

$$\frac{\partial u_x}{\partial x}=\frac{1}{E}\left(\frac{\partial^2\chi}{\partial y^2}-\nu\frac{\partial^2\chi}{\partial x^2}\right)-\frac{1}{2}\left(\frac{\partial\zeta}{\partial x}\right)^2, \tag{7a}$$

$$\frac{\partial u_y}{\partial y}=\frac{1}{E}\left(\frac{\partial^2\chi}{\partial x^2}-\nu\frac{\partial^2\chi}{\partial y^2}\right)-\frac{1}{2}\left(\frac{\partial\zeta}{\partial y}\right)^2 \tag{7b}$$

and

$$\frac{\partial u_x}{\partial y}+\frac{\partial u_y}{\partial x}=\frac{-2(1+\nu)}{E}\frac{\partial^2\chi}{\partial x\partial y}-\frac{\partial\zeta}{\partial x}\frac{\partial\zeta}{\partial y}. \tag{7c}$$

2.2.2. The buckling is supercritical

We shall now prove the following intermediate result: if there exists a buckled configuration $(\zeta_{\rm b}, \chi_{\rm b})$ of the film, solution of the Föppl–von Kármán equations in the domain $\mathscr{D}$, then any *unbuckled* equilibrium configuration $(\zeta_{\rm u}\equiv 0, \chi_{\rm u})$ is unstable with respect to this buckled configuration. As a result, the situation depicted on the left of Fig. 2 is not possible, and the initial buckling of the film is necessarily a

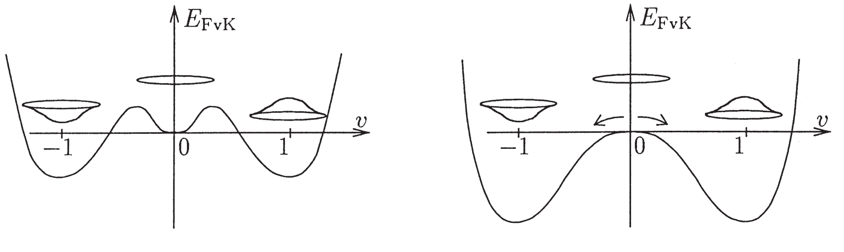

Fig. 2. The initial buckling of a 2D blister is always supercritical: metastability of the unbuckled configuration in the presence of a buckled equilibrium configuration (left) would be incompatible with the actual form of the Föppl–van Kármán energy (right).

supercritical bifurcation. This will permit scaling laws to be derived for the edges loads, M and Σ, later in Section 2.2.3.

To prove the intermediate result, we assume that a buckled equilibrium configuration (ζ_b, χ_b) exists; then, we introduce a one-parameter family of configurations for the film, (ζ_v, χ_v), indexed by a parameter v:

$$\zeta_v = v\zeta_b, \quad \chi_v = (1-v^2)\chi_u + v^2\chi_b. \tag{8}$$

One recovers the unbuckled configuration $(0, \chi_u)$ for $v=0$, the buckled configuration (ζ_b, χ_b) for $v=1$, and the mirrored one $(-\zeta_b, \chi_b)$ for $v=-1$. Because of the horizontal mirror symmetry $\mathcal{S}:(\zeta, \chi)\mapsto(-\zeta, \chi)$ in the Föppl–von Kármán equations, $v=-1$ is an equilibrium state of the film, as well as $v=0, 1$ by assumption. For other values of v, the film is not in equilibrium, and Eqs. (6a) and (6b) do *not* hold.

An important point is that *all* configurations indexed by v satisfy the clamped boundary conditions, provided the two original states $(0, \chi_u)$ and (ζ_b, χ_b) do. Indeed, these boundary conditions read, along the edge of the blister, $\mathcal{C}$: $u_x=u_y=0$ (no horizontal displacement) and $\zeta=\zeta_{,n}=0$, where the subscript "n" stands for a normal derivative. The horizontal components of the displacement are solutions of Eqs. (7) which, by definition (8), only depend on v through terms proportional to v^2. Therefore, they necessarily take the form: $u_x^v=u_x^u+v^2(u_x^b-u_x^u)$, and a similar formula for u_y^v. Since, by assumption, the clamped boundary conditions are satisfied for $v=0, 1$, one has $u_x^u=(u_x^b-u_x^u)=0$, hence $u_x^v=0$ along the edge $\mathcal{C}$, for all v. The same holds for u_y^v, and the first set of boundary conditions is satisfied for any configuration in the family indexed by v. Finally, by using similar arguments, $\zeta_v=(v\zeta_b)=0$ and $\zeta_{,n}^v=v(\zeta_b)_{,n}=0$ on the edge of the blister, and the second set of boundary conditions is also satisfied for any v.

All configurations (ζ_v, χ_v) therefore correspond to the *same* blister with a fixed boundary $\mathcal{C}$, and it makes sense to plot the elastic energy of the film as a function of v. The constraints on this plot are: (1) it must be symmetric under the mirror symmetry $\mathcal{S}:v\mapsto(-v)$; (2) the energy is stationary for $v=-1, 0, 1$; and (3) finally, combination of Eqs. (5) and (8) shows that this energy is a polynomial in v of degree four. In order to feature two symmetric stable equilibrium states at $v=\pm1$, plus a metastable one at $v=0$, the energy would need to be at polynomial of degree at least six in v (see left part of Fig. 2). This shows, by contradiction, that any unbuckled equilibrium state of the film, $v=0$, is unstable above the buckling threshold (Fig. 2, right), and not metastable. Therefore, we proved that the buckling of the film is always supercritical.

That the buckling bifurcation is supercritical implies that the critical compression σ_0 can he determined by studying the linear stability of the unbuckled configuration. Let $\zeta^{[1]}$, $\chi^{[1]}$, $u_x^{[1]}$ and $u_y^{[1]}$ represent an infinitesimal change in the configuration of the film near the unbuckled state. Linearization of Eqs. (6) yields the equation for a marginal mode:

$$\frac{Eh^3}{12(1-v^2)}\Delta^2\zeta^{[1]} - h\sigma_0\Delta\zeta^{[1]} = 0 \text{ on } \mathcal{D}, \tag{9}$$

with the boundary conditions $\zeta^{[1]}=\zeta_{,n}^{[1]}=0$ along $\mathcal{C}$. The Airy potential and the in-plane displacements vanish for a marginal mode, as the linearization of Eqs. (6) and (7) yields $\chi^{[1]}=0$ and $u_x^{[1]}=u_y^{[1]}=0$. That the marginal mode involves only vertical displacements will be explained below by symmetry arguments.

Dimensional analysis of Eq. (9) for the marginal mode again leads to Eq. (1) for the critical stress σ_c. Therefore, the critical stress for buckling in the region $\mathcal{D}$ is comparable to that for buckling in a circular region of comparable extent, b. Now, k^g is a coefficient depending only on the blister geometry [and not on b, which appears independently in Eq. (1)]. This geometrical coefficient k^g could be calculated for any particular blister geometry by solving Eq. (9), but this is unnecessary: since we consider blisters having a *single* typical length scale, k^g has to be of order unity. This is all we shall need to know about k^g for the rest of the analysis. Incidentally, this paragraph proves that, for the class of blisters studied here, the dimensionless buckling parameter, η, is still given by Eq. (2).

2.2.3. Weakly non-linear analysis above the buckling threshold

If the buckling were subcritical (as in Fig. 2, left), the only way to approach the buckled state would be by solving exactly the Föppl–von Kármán equations. Luckily, we have been able to prove that the buckling bifurcation is supercritical (Fig. 2, right), and a weakly non-linear analysis of the buckling can be performed. This approximate approach is considerably simpler than the exact solution of the Föppl–von Kármán equations; other plate buckling problems suggest that this method gives accurate results, including well above the threshold, although it is valid in full rigor for $\eta \ll 1$ only. In fact, it is not even necessary to carry out the weakly non-linear analysis in full details (which is anyway still intractable for an arbitrary blister): a symmetry argument straightforwardly yields the weakly non-linear behavior of a 2D blister. Indeed, as noted above. the Föppl–von Kármán equations are invariant under the horizontal mirror symmetry, $\mathcal{S}$. Under this symmetry, the moment, M, and the in-plane stress release caused by buckling, Σ, change according to $M \to -M$ and $\Sigma \to +\Sigma$. As usual for supercritical bifurcations (Landau and Lifschitz, 1986a), the invariance of the Föppl–von Kármán equations under the symmetry $\mathcal{S}$ imposes a scaling law for the quantities in the buckled state: $M \propto \eta^{1/2}$ and $\Sigma \propto \eta$ above the buckling threshold. The reversal of the sign of M upon $\mathcal{S}$ indeed corresponds to an indeterminacy in the sign of $\eta^{1/2}$ when the square root is extracted.

Given that η is the only dimensionless parameter of the problem that depends upon the size of the blister, b, the full expression for the edge loads follows from dimensional analysis:

$$M = c_1^g(\nu, s/b)\frac{Eh^4}{b^2}\eta^{1/2}, \quad \Sigma = c_2^g(\nu, s/b)\frac{Eh^2}{b^2}\eta. \tag{10}$$

Unknown numerical functions, $c_{1,2}^g$, had to be introduced; we shall see that their precise calculation, which goes beyond the present dimensional analysis, is not required. These functions may not depend on η, which has already been factored out. They depend on the blister shape, hence the superscript "g", and may vary along the edge of the blister: s is the curvilinear coordinate along the delamination front, and s/b is the dilatation-invariant coordinate. By definition of the typical size of the blister b, s/b is of order unity. As a result, the values taken by these functions $c^{g}_{1,2}$ have to be of order unity as well. As for 1D blisters, the loading at the crack tip, (G, ψ), can be derived from the edge loads in Eq. (10) (Suo and Hutchinson, 1990):

$$G(r)=\frac{6(1-\nu^2)Eh^5}{b^4}([c^{g}_{1}(s/b,\nu)]^2\eta+\tfrac{1}{12}[c^{g}_{2}(s/b,\nu)]^2\eta^2) \tag{11a}$$

and

$$\psi(r)=-90°+\omega(E/E_s,\nu,\nu_s)-\tan^{-1}[c^{g}_{3}(s/b,\nu)\eta^{1/2}], \tag{11b}$$

where, as before, $c^{g}_{3}=c^{g}_{2}/(c^{g}_{1}\sqrt{12})$ and $s/b=O(1)$. All functions c_i take positive values: in Eq. (10), the buckling is upwards, hence $c^{g}_{2}>0$. Since buckling permits a *release* of the initial compressive stress, $\sigma_0<0$, one must have $c^{g}_{1}>0$, as in the 1D case.

2.2.4. Stabilization of 2D patterns

Eqs. (11) show that the dependence of the crack tip loading on η in Eqs. (4) could be extended to the 2D case. The only difference is in the coefficients c_i, which now vary along the edge of the blister. Although the functions c_i are unknown, it must be pointed out that the dependence on the blister size, b, and on the initial compression, σ_0, is fully captured in the two equations above, through η. This remark makes determination of the c_i values unimportant to our problem. Indeed, during self-similar growth of a blister at constant residual stress, the dimensionless buckling parameter η increases by Eq. (2), while other quantities remain constant. In consequence, the energy release rate, G, increases uniformly along the delamination front. In the absence of mode-dependent toughness, this would make all blisters unstable against full delamination of the film, as in one dimension. However, the mode-mixity parameter, ψ, takes more and more negative values as η becomes larger, because $c_3>0$ in Eq. (11b). Therefore, the interface toughness increases uniformly along the edge of the blister as it spreads. This results in an effective toughening of the interface, which can prevent widespread delamination of the film: on the basis of the similarity of Eqs. (4b) and (11b), the mechanism proposed for 1D blisters can be extended word by word to 2D patterns.

Finally, we shortly discuss a few points. First, we have only considered blisters having a single typical length scale, b. We also have considered self-similar growth, and not growth in only one direction at constant width. For these reasons, elongated blisters, such as telephone cords, are excluded from the present analysis. The need to exclude elongated blisters may not be simply a weakness of the present theory. It may indicate that the mode dependence of the interface *cannot* prevent large-scale delamination in just one direction. As a matter of fact, telephone-cord blisters have been observed in a variety experimental conditions (Gioia and Ortiz, 1997). Second, we have neglected mode III (transverse) loading on the crack tip, which is in general present for non-axisymmetric 2D blisters. It is possible to extend the present analysis to determine the weakly non-linear mode III loads on the crack. In the absence of experimental toughness data with transversal loading, however, this would be useless. Presumably, the enhancement of the interface toughness, which is reckoned to be a consequence of progressive crack closure (see next section), should not be too sensitive to transverse loading. Third, we must emphasize that the mode dependence of the interface toughness does not systematically *prevent* complete delamination of the film (in particular, if the initial compression of the film is too high) but *discourages* it, allowing blisters of finite size to be stable under certain mechanical conditions.

3. Friction-induced mode dependence in brittle materials

In this section, we study a mechanism responsible for the mode dependence of the interface toughness. Several dissipative processes (Evans et al., 1990), like interfacial friction (Evans and Hutchinson, 1989; Stringfellow and Freund, 1993), plasticity (Swadener and Liechti, 1998; Shih and Asaro, 1991) and viscoelastic dissipation (Chai, 1990), are potential sources of mode dependence. We focus on the role of friction; to remove the other sources of mode dependence, we consider a brittle interface made of materials following linear elasticity, except of course in a *very small* non-linear region near the tip where the stress formally diverges. When the loading on the interface crack is such that the crack faces contact ($K_{\mathrm{I}} \leq 0$, i.e., $|\psi| > 90°$), the interaction of the crack lips causes dissipation. This certainly results in an increased toughness of the interface. This effect is, by nature, highly sensitive to the degree of closure of the crack, and is therefore mode-dependent. Two models have been proposed to assess the effect of interfacial friction on the propagation of the crack. Evans and Hutchinson (1989) have studied the screening of the crack tip by asperities on the crack surface. Following Stringfellow and Freund (1993), we instead study a model of interface fracture in which the crack faces remain planar, as the fracture follows a planar interface (see Fig. 3). The loading is such that $K_{\mathrm{I}} < 0$, and the crack faces fully contact. In the contact region, the interaction between the faces is modeled by a Coulomb law of friction.

Coulomb friction in interface cracks leads to anomalous divergence of the stress, as was found more than two decades ago by Comninou and Dundurs (1979): in Eq. (12), the stress does not follow the usual $1/\sqrt{r}$ behavior, where r is the distance to the tip. This unusual divergence has widely been overlooked, perhaps because it was believed to introduce inconsistencies in the theory of fracture (in the form of infinite

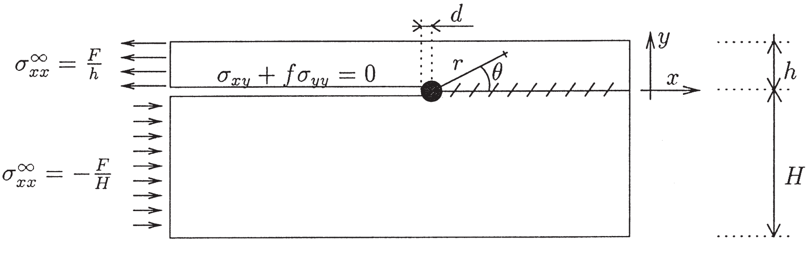

Fig. 3. An interface crack model allowing assessment of the effect of interfacial friction on the interface toughness, as introduced by Stringfellow and Freund (1993). A small non-linear region of size d centered on the tip is represented as a dark area.

energy release rates); for the same reason, observable effects following from it do not seem to have been investigated. Only recently, we were able to prove that the anomalous law (12) does not introduce inconsistencies in the theory (Audoly, 2000): it is perfectly safe to introduce the Coulomb friction in closed interface cracks. In consequence, the asymptotic law (12) for the stress should *not* be disregarded, and the discussion of frictional effects in interface cracks has to rely on it. This is the aim of the present section.

We present the model of interface crack introduced by Stringfellow and Freund (1993). For the sake of convenience, we shall continue to call "film" and "substrate" the two materials merging at the interface, although our arguments are not specific to delamination of thin films. The geometry is as in Fig. 3: a force F is pulling an elastic layer of width h partially bound to an infinitely thick substrate ($H \gg h$); f is the Coulomb coefficient of friction at the interface; E_f, ν_f, E_s, ν_s, are the Young's modulus and Poisson's ratio for the film and substrate, respectively. The mode mixity at the crack tip is known for this particular loading geometry: from Suo and Hutchinson (1990) one obtains $\psi_{SF} \approx -124°$. This is of course only an *apparent* value, as the definition of ψ is somewhat arbitrary when the crack lips contact.

When the film is stiffer than the substrate, the formation of a bubble near the tip has been observed numerically, a fact that has recently been explained (Audoly, 2000). To keep the arguments as simple as possible, we shall not be concerned with this possibility: we consider only the case when the substrate is weaker than the film. Then, there is no bubble. Because of dissipation in the contact region. the Rice integral (Rice, 1968a,b) is *not* path-independent, and the energy release rate at the tip, G_t, is less than the Rice integral calculated along the outer boundary of the sample, J_{ld} ($J_{ld} \propto F^2$ measures the intensity of external loading, hence the subscript "ld"). This effect is referred to as "frictional screening". We shall call T the transmission coefficient $T = G_t/J_{ld}$, which depends on the coefficient of interfacial friction and on the mismatch of elastic properties between film and substrate. Linear elasticity is used, so that T does not depend on the magnitude of F, as long as F remains positive (when $F < 0$, the crack opens. and the problem has different boundary conditions on the interface). Stringfellow and Freund have calculated T for different typical values of the parameters; to do this, they used a numerical scheme based on finite elements, and they evaluated the Rice integral along a small contour around the crack tip. A typical value of transmission coefficient (hence a typical frictional screening) for realistic values of the parameters is $T_{SF} = 0.68$ for $f = 0.5$ and $E_f/E_s = 0.25$ (Stringfellow and Freund, 1993).

We note that the value of T can be used to estimate the contribution of interfacial friction to the mode dependence of the crack. Retaining only friction among the various contributions, we shall indeed assume that the crack has an intrinsic toughness Γ_i: propagation of the crack occurs when the crack-tip loading reaches the critical value $G_t = \Gamma_i$. For a loading such that the crack is well open ($\psi = 0°$), the crack faces do not contact, and the Rice integral is contour-independent, $T = 1$; the critical *external* loading at the onset of crack propagation is simply $J_{ld} = G_t = \Gamma_i$. This quantity is also called the *effective* toughness: $\Gamma_{eff}^{open} = \Gamma_{eff}(\psi = 0°) = \Gamma_i$. In contrast, the effects of interfacial friction are maximized for the loading in Fig. 3, because $K_I < 0$; then, the onset of crack propagation is still at $G_t=\Gamma_i$, but in terms of the external loading this yields $TJ_{ld}=\Gamma_i$, hence the effective toughness is $\Gamma_{eff}^{closed}=\Gamma_i/T$ when $\psi=\psi_{SF}$. The relative increase in the effective toughness for large $|\psi|$ is therefore of order $\gamma=(\Gamma_{eff}^{closed}-\Gamma_{eff}^{open})/\Gamma_{eff}^{open}=T^{-1}-1$. Using the value of T obtained by Stringfellow and Freund (1993), given above, a relative increase of the interface toughness by a factor $\gamma_{SF}=47\%$ is obtained when the crack closes.

An asymptotic analysis of the interfacial crack with friction has been performed by Comninou and Dundurs (1980), and extended by Deng (1994). We have recently, complemented it by showing that the energy release rate at the tip, G_t, in fact *vanishes* in the model in Fig. 3 (Audoly, 2000). Indeed, when the faces of an interface crack contact near the tip, the components of the stress diverge near the tip according to:

$$\sigma_{ab}(r,\theta)\propto r^{(1/2)-(\delta/2)} \quad \text{where} \quad \delta=\frac{2}{\pi}\tan^{-1}(\beta f), \quad \beta=\frac{\mu_s(\kappa_f-1)-\mu_f(\kappa_s-1)}{\mu_s(\kappa_f+1)+\mu_f(\kappa_s+1)}. \tag{12}$$

Here $\mu=E/[2(1+\nu)]$ is the shear modulus of either material, $\kappa=(3-4\nu)$ (plane strain is imposed by the substrate of thickness $H\gg h$), and $a, b=x$ or y. Polar coordinates (r, θ) centered on the tip are used. The azimuthal dependence of σ_{ab} is not needed here, but can be found in Comninou (1977). We shall assume some elastic mismatch, $\beta\neq0$, between film and substrate, as these materials differ; then, the exponent δ is non-vanishing. This coefficient is moreover positive, as was generally proved by Audoly (2000): $\delta<0$ in the above formula would correspond to $\beta<0$, i.e., film stiffer than substrate, but the model is then inappropriate, because a bubble is formed near the tip and Eq. (12) does not hold. The positivity of δ means that the divergence of the stress is always *abnormally weak* near the crack tip, in comparison with standard crack theory. Dimensional analysis using Eq. (12) shows that the energy release rate calculated along a circle of vanishingly small radius r, centered on the tip, vanishes like $G_r\propto r^{\delta}$ for $r\rightarrow0$ ($\delta>0$). In the presence of friction *and* elastic mismatch, the energy release rate $G_t=G_{r=0}$ is therefore vanishing. The predictions of the asymptotic analysis are therefore: $T=0$, and $\gamma_{AA}=+\infty$. The effective toughness is infinite when the interface crack is closed. This shows that the energy available from external loading is completely dissipated at the interface and no finite energy flux enters the crack tip. In the absence of energy for breaking interfacial bonds, complete frictional locking of the crack is predicted by this analysis (for loadings such that the crack is closed near the tip).

Analytical and numerical results therefore seem to contradict each other. The consistency is restored, using the following remark (Deng, 1994). The mismatch parameter is of order $|\beta|\sim0.1$ for most interfaces (Rice, 1988); by Eq. (12), with a coefficient of friction of order unity, the anomalous exponent is of order $\delta\sim0.05$. As a result, the energy release rate, $G_r\propto r^{\delta}$, decreases very slowly to zero as the tip of the crack is approached ($r\rightarrow0$). The value of T calculated numerically depends on the size of the mesh elements near the tip, although this size is small compared with all other lengths in the problem. This probably accounts for the discrepancy between numerical and analytical approaches: in the limit of vanishingly small mesh elements, the analytical result would have been recovered numerically, i.e., $T=0$.

Neither the numerical nor the analytical predictions represent reality in an absolute sense. By the paragraph above, the numerical results depend on a hidden parameter, the mesh resolution. Using linear elasticity of continuous media, the analytical approach predicts a complete dissipation of the external loading; however, part of this dissipation occurs at very small scales, in a region near the tip where both the approximations of linear and continuous medium cease to be valid. This serves as a warning that any small (eventually microscopic) scale near the tip shall strongly influence the value of T, and, therefore, the importance of frictional effects. In order to model interfacial friction consistently, a small length scale near the tip, d, must therefore be introduced explicitly in the model. This length scale is the size of the region where the materials do not behave like a linear elastic and continuous medium, due to plasticity or cohesion for example.

To focus on interfacial frictional among other sources of mode dependence, we chose to consider ideally brittle materials. Barenblatt has proposed an estimate of the length d in such materials; it is the size of the region over which the stress overcomes the atomic cohesion: $K_{\mathrm{Ic}}/\sqrt{d}=\sigma_{\mathrm{m}}$ where σ_{m} is the maximal intensity of forces of cohesion and K_{Ic} is the critical stress intensity factor for crack propagation. For a perfect crystal with interatomic spacing a, $\sigma_{\mathrm{m}}\sim\mu/10$, where μ is a typical modulus of the material (Barenblatt, 1963). Moreover, a useful estimate for the critical stress is: $K_{\mathrm{Ic}}\sim\mu\sqrt{a}$. Combining these results, the width of the Barenblatt region is estimated as: $d_{\mathrm{br}}\sim(100a)\sim10^{-8}$ m in the ideally brittle case; in materials that are not perfectly brittle, this length can be larger by several orders of magnitude.

We arrive at a situation very similar to the one studied by Barenblatt (1963) in his famous regularization procedure. In order to remove the stress singularities from the theory of cracks, he introduced a non-linear zone near the tip, which can be seen as a black box. Making a balance of the energy flowing into this non-linear zone, Barenblatt derived a simple criterion for quasi-static advance of the crack: the energy release rate calculated around this black box of size d, G_d, must equal a fracture energy, Γ_{i}. This fracture energy is related to the cohesive and plastic properties of the crack. In standard crack theory, G_r converges to $G_{\mathrm{t}}\neq0$ for $r\longrightarrow0$ and the length d is much smaller than any other length, so that Barenblatt's approach is sometimes seen as a refined manner to establish the Griffith's criterion for crack propagation: $G_{\mathrm{t}}\geq\Gamma_{\mathrm{i}}$. Being very small, the length scale d disappears from the standard theory of cracks. However, in the framework of interfacial cracks with friction, Barenblatt's criterion does *not* reduce to Griffith's one: because of the very slow vanishing of G_r at the tip (δ is small), G_d is very different from $G_{\mathrm{t}}=0$, and the length scale d must be kept. The Barenblatt criterion for crack advance, $G_d\geq\Gamma_{\mathrm{i}}$, can be rewritten $J_{\mathrm{Id}}\geq\Gamma_{\mathrm{eff}}$, when Γ_{eff} is defined as $\Gamma_{\mathrm{eff}}=\Gamma_{\mathrm{i}}/T$, with $T=G_d/J_{\mathrm{Id}}$. This shows that a consistent definition of the frictional screening T should involve G_d, not G_{t}. The very small scale that we need to introduce in the theory is Barenblatt's length, d.

The effect of the interfacial friction on the interface toughness of a brittle interface can now be estimated. We shall rely on the numerical simulations performed by Stringfellow and Freund (1993), which have already solved the “macroscopic” part of the problem in Fig. 3; we simply need to extend their approach so as to correctly account for frictional effects below their numerical resolution. The same (typical) values for the parameters are used as theirs: f=0.5, E_f/E_s=0.25, $\nu_f=\nu_s$=0.3. We define the numerical resolution, q, as the ratio of the radius of the smallest numerical contour around the tip, r_{SF}, to the thickness of the film, h: $q=r_{SF}/h$. We remind that the calculated value T_{SF} in fact depends on q, as it would vanish for $q\to 0$ (perfect mesh). For the particular mesh of finite elements used by Stringfellow and Freund (1993), $q\approx 0.06$. Using definition (12), the anomaly in the stress divergence is δ=0.054. Assuming that the asymptotic law $G_r \propto r^{\delta}$ is satisfied in the small region of size r_{SF}/h, the energy release rate is damped by a factor $(d_{br}/r_{SF})^{\delta}$ when passing from the *numerical* resolution, r_{SF}, to the *physically* relevant Barenblatt scale, d_{br}. Therefore, the energy release rate entering in the Barenblatt criterion for crack advance is: $G_d=(d/r_{SF})^{\delta} G_{SF}$, where G_{SF} is the energy release rate obtained numerically in Stringfellow and Freund (1993). The relative increase of interface toughness for brittle materials therefore reads:

$$\gamma_{br}=\frac{1}{T_{br}}-1 \quad \text{where } T_{br}=T_{SF}\left(\frac{d_{br}}{r_{SF}}\right)^{\delta}=T_{SF}\left(\frac{d_{br}}{qh}\right)^{\delta}. \tag{13}$$

Assuming that the above layer has a thickness h=300 μm and using the Barenblatt estimate for d_{br}, we obtain γ_{br}=121%. As noted above, γ measures the contribution of interfacial friction in the effective toughness of the crack when $K_I<0$: when one arbitrary unit of energy is brought to the tip for breaking interfacial bonds, 1.21 units are at the same time dissipated by friction; as a result, most of the toughness is due to friction. The value of γ also permits one to characterize the mode dependence of the interface: due to friction, its toughness is multiplied by a factor of about $\Gamma_{eff}^{closed}/\Gamma_{eff}^{open}$=2.21 upon crack closure.

For comparison, the value γ_{SF}=47% has been obtained when dissipation below the numerical resolution (6% of the film thickness) is neglected (Stringfellow and Freund, 1993). It is much less than γ_{br}=121%. This demonstrates that dissipation at small scales near the tip can be quite important in the fracture of interfaces; in numerical simulations, a significant contribution to the interface toughness can be missed if dissipation below the numerical resolution is neglected. This result is after all quite intuitive: near the tip, the contact pressure diverges, which makes frictional screening especially efficient.

All of the present analysis is based on the anomaly in the scaling law (12) for the stress in a closed interface crack ($\delta\neq 0$): in the presence of friction and elastic mismatch, frictional screening does not simply result in a decrease of the stress intensity factor at the tip; more dramatically, it changes the nature of the divergence of the stress, making it smoother. In this section, we argued that the weakness of the stress singularity underlies observable effects: a strong frictional dissipation at small scales near the tip has been pointed out, which strongly enhances the effective toughness of the interface. As a result, our estimate of the mode dependence induced by friction is higher than when the anomalous law (12) is overlooked, and seems more compatible with experimental plots of $\Gamma(\psi)$. Finally, we note that, by Eq. (13), the effects of interfacial friction are stronger when the Barenblatt length is smaller, i.e., for more brittle materials [in plastic materials, the length d must be taken as the size of the plastic region, which is often larger than d_{br} by several orders of magnitude (Barenblatt, 1963)]. The effects of interfacial friction on the mode dependence are stronger when that of plasticity are weaker: plasticity and interfacial friction act as *complementary* sources of mode dependence. Which one dominates in a particular interface depends on the brittleness of the materials.

4. Conclusion

We have first considered the effect on the mode dependence on the patterns of delamination. In the absence of exact analytical solutions to the non-linear Föppl–von Kármán equations, an approach based on dimensional analysis turned out to be effective: the evolution of the mode-mixity parameter along the crack front during growth of a blister could be derived for quite generic geometries. We have found that the delamination front is more and more a mode II crack as the blister spreads in all directions. As a result, the mode dependence of the film/substrate interface can prevent widespread delamination. Experimental observation of finite-sized blisters, and of elongated ones, could be understood. All of these arguments generalize results obtained previously for circular or straight-sided blisters.

In a second step, we have shown that interfacial friction can explain mode dependence. Coulomb friction leads to an anomalous divergence of the stress near the tip of a closed interface crack. As a result, the effective toughness increases severely upon crack closure (typically by 120%; i.e., the interface becomes more than twice tougher). A physical interpretation was given: the frictional screening of the external load is very effective in the contact region near the tip, where the contact pressure is high. It was emphasized that these frictional effects are sensitive to a very small scale, d, which depends on the brittleness of the materials. Our treatment of Eq. (12) may be applicable to other mechanical problems involving anomalous asymptotic laws, which often appear in elasticity when non-standard boundary conditions are considered.

Collecting the results presented in the present paper, we are led to the following picture: when a blister spreads, the delamination front is more and more a mode II crack. Above a critical blister size, the lips of the crack contact (K_I=0), and strong frictional effects can prevent further expansion of the blister. This picture is of course simplified, as other sources of mode dependence arising in real materials are ignored. This scenario is nevertheless perfectly valid in the limit of an ideally brittle interface. Quite remarkably, it provides a picture of a complex process that is both simple (its ingredients are linear elasticity and Coulomb friction) and consistent with experiments (the existence of blisters of finite extent is explained).

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