Self-patterning through thin film buckling

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Abstract Thin films may spontaneously form buckles if the residual stresses are compressive and large and if adhesion is weak. Buckling driven delamination leads to periodic patterns, such as the telephone cord morphology which has been studied for decades. Until recently the role of adhesion in the formation of buckling patterns was rather poorly understood. The difficulties are: 1. buckling is a non-linear phenomenon 2. Adhesion is a complex process involving mode mixity at the crack tip. Here coupling buckling and mixed-mode adhesion in a FEM model, we investigate the influence of film thickness, adhesion and residual stresses on the final buckle morphology. In particular, we show that it is possible to control the delamination front to create branching morphologies. Such morphologies remain periodic and can cover large areas. Experimentally, we show how the relevant conditions can be achieved for multilayer stacks of thin films deposited by magnetron sputtering. In particular we demonstrate self-organized hexagonal networks of buckles at the 10 micron scale. We anticipate that these patterns can be scaled from the micron-scale to the nano-scale.

Keywords Buckling, Adhesion, Mode mixity, Patterning,

1. Introduction

Thin films may spontaneously form buckles[1,2] if the residual stresses are compressive and large and if adhesion is weak. Buckling driven delamination leads to periodic patterns, such as the telephone cord morphology which has been studied for decades. Until recently the role of adhesion in the formation of buckling patterns was rather poorly understood. The difficulties are: 1. buckling is a non-linear phenomenon 2. Adhesion is a complex process involving mode mixity at the crack tip. Here coupling buckling and mixed-mode adhesion in a FEM model, we investigate the influence of film thickness, adhesion and residual stresses on the final buckle morphology. We then use the results as a guideline for experiments

2. Numerical simulation

2.1. Model

In order to analyze the growth of the blister, we use a numerical model coupling finite elements with cohesive zone modeling. It is essential to use a nonlinear plate model to capture the buckling equilibrium. The surface of the plate is defined as the (O,x,y) plane and the out-of-plane displacement is $w(x,y)$. In order to take into account the presence of a purely rigid substrate, the unilateral contact condition $w(x,y)>0$ is introduced. The calculations are made for large displacements w, using the Green Lagrange strain tensor. When w is large, the strain tensor reduces to

$$\begin{cases} e_{xx} = \dfrac{\partial u}{\partial x} + \dfrac{1}{2}\left(\dfrac{\partial w}{\partial x}\right)^2 \\ e_{yy} = \dfrac{\partial u}{\partial y} + \dfrac{1}{2}\left(\dfrac{\partial w}{\partial y}\right)^2 \\ e_{xy} = \dfrac{1}{2}\left(\dfrac{\partial u}{\partial y} + \dfrac{\partial v}{\partial x}\right) + \dfrac{1}{2}\dfrac{\partial w}{\partial x}\dfrac{\partial w}{\partial y} \end{cases} \quad (1)$$

In Eq. (1) the terms which are non-linear in w are responsible for a third order term in the thin plate equilibrium equations and it is this non-linearity which is essential to capture the post-buckling evolution of the blisters (e.g. see [3]).

The second key point is the description of adhesion rupture for which a cohesive zone model (CZM) [29–32] is used. Detailed description of the model can be found in [4]. Cohesive elements are inserted at the interface. A bilinear softening behavior is used in order to take into account for the mode mixity of the interfacial toughness. The interfacial toughness Γ^c is defined as a function of the mode mixity angle ψ, see Fig.1, which is the relative proportion of shear and normal traction applied at the interface:

$$\tan\psi = \frac{K_{II}}{K_I} \quad (2)$$

We use for the definition of $\Gamma^c(\psi)$ the one proposed by Hutchinson in [5] and based on experimental data[6].

$$\Gamma^c(\psi) = \Gamma_I^c\left(1 + \tan^2 \eta\psi\right) \quad (3)$$

Where Γ_I^c is the mode I interfacial toughness, and η a dimension-less parameter to adjust the level of the mode II interfacial toughness Γ_{II}^c.

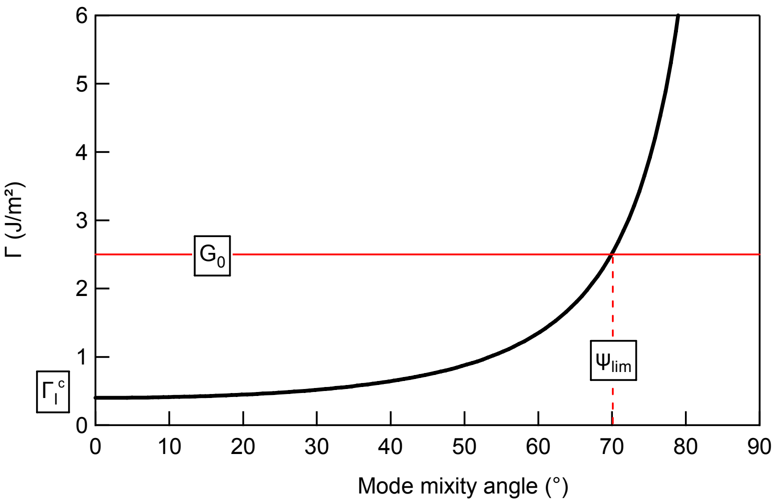

Figure 1 : Dependence of the interfacial toughness as a function of the mode mixity angle.

Thin film is modeled by a plate of thickness h, elastic modulus E and Poisson ratio ν. In-plane stress σ_0 is introduced by a thermal expansion.

2.2. Results

We fix the materials parameters of the film to *E*=329 GPa and υ=0.31. The interfacial toughness is kept constant choosing $\Gamma_I^c = 0.4 J.m^{-2}$ and $\eta = 0.95$. The morphology of the delamination is then studied as a function of loading, which is given by the couple $(h;\sigma_0)$. It is convenient to define the total energy per unit area stored in the film due to the in-plane stress.

$$G_0 = \frac{1-\nu}{E} h \sigma_0^{\ 2} \tag{4}$$

We also define the limit for the mode mixity above which the interface cannot be fully separated. In case where the energy release rate is close to G_0 then

$$\psi_{\lim} = \frac{1}{\eta} \tan^{-1} \sqrt{\frac{G_0}{\Gamma_I^c} - 1} \tag{5}$$

It should be noted that $\psi_{\lim}$ is attained at the side of the blister as the film sags[4] and the mode mixity reaches pure mode II, $\psi = \pi/2$. However the interface is not fully separated at all angles above $\psi_{\lim}$.

Changing level of loading with $\psi_{\lim}$, we observe different morphologies, from no delamination to complete delamination of the film and draw a phase diagram $\psi_{\lim}$ is a relevant parameter for the instability of the blister and is equivalent to the loading ratio σ_0/σ_c of the blister. In case of the straight sided blister, the critical buckling stress σ_c corresponds to the stress needed to enter in the post-buckling regime.

$$\sigma_c = \frac{\pi^2}{12} \frac{E}{1-\upsilon^2} \left(\frac{h}{b}\right)^2 \tag{6}$$

Secondary buckling instability computation on the sides of a straight sided blister by Jensen [7], and on the collapse of a circular blister by Hutchinson [8] show that such destabilization is solely a function of G_0 and therefore $\psi_{\lim}$.

3. Experiments

We aim to study morphologies of the delamination experimentally. We use the superlayer method to make blisters [9]. A compressive stressed layer of molybdenum is deposited on top of a weak interface to provide elastic energy for buckling. The residual stress in the Mo layer before buckling is around 3GPa. The stress can be tuned changing the deposition conditions of the Mo layer during sputtering [10]. The thickness is varied from 100nm to 200nm. We here use a silver layer to create the weak interface with the substrate.

Both control over the residual stress and the thickness enable scanning different loading conditions and therefore variation of $\psi_{\lim}$. Experimentally we observe several morphologies, from the telephone cord like blister to complete delamination, see Fig.2. Those morphologies can be compared to the numerical results and placed in a phase diagram.

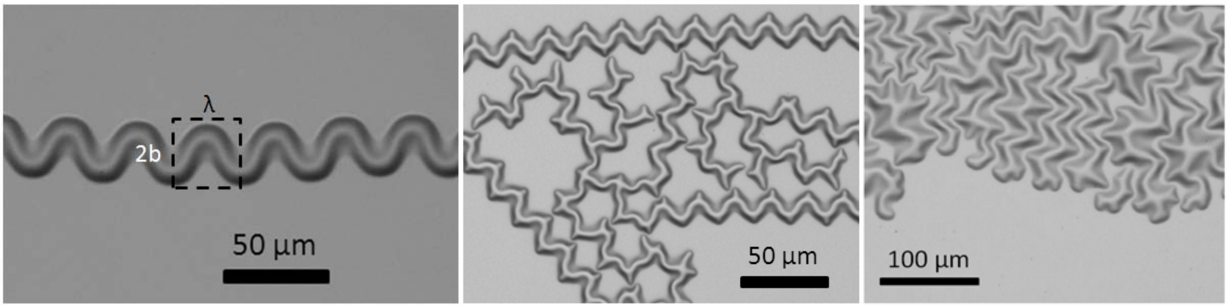

Figure 2: Evolution of the delamination morphology for Si/Ag interface for different layer of molybdenum.

In particular we use numerical simulations to demonstrate that a self-organized network of buckles can be generated and that the conditions to generate such a network experimentally can be achieved with the superlayer method.

4. Conclusions

We present a numerical model to explore the morphology of the buckling driven delamination pattern as a function of interfacial properties and loading conditions. Confronting the experimental data with the simulation, we find relevant parameters for patterning of large surfaces.

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