Towards interface toughness measurement in nanometric films

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Abstract Thin films deposited on substrates are usually submitted to large residual compression stresses, causing delamination and buckling of the film into various patterns. This phenomenon has been widely investigated in the past few decades, both experimentally and with nonlinear plates models. Nevertheless, the formation of the most commonly observed pattern, the "telephone cord blister", has only be understood recently, with FEM models coupling post-buckling studies and mixed-mode adhesion. Here, relying on these models, we show remarkable properties of these wavy blisters, in particular we show that the wavelength of the telephone cord patterns scales linearly with a parameter depending on the stress level in the film, the thickness and the adhesion energy. This result has an important practical application. As a matter of fact, since it is experimentally possible to control the stress level and the film thickness during elaboration, the measurement of this wavelength can indirectly lead to a measurement of the interface toughness, even in very small films.

Keywords Buckling, Adhesion, Mode mixity, Patterning,

1. Introduction

Compressively stressed thin films with low adhesion frequently buckle and delaminate simultaneously into various patterns, amongst which the so called "telephone cords" pattern is very often observed [1,2]. Although these buckles have been studied for decades, no complete understanding of their propagation had been presented until recently. As a matter of fact, the post-buckling equilibrium shapes of the blisters require the use of non-linear plates theory (e.g. see [3]). Furthermore adhesion is a complex process involving mode mixity at the crack tip. A nonlinear plate deformation model has been coupled with a cohesive zone model to simulate the kinematics of a propagating telephone cord buckle in very close agreement with experimental observations. Proper inclusion of the dependence of an adhesion upon the mode mixity proved to be central to the success of the approach. The clarification of the mechanism allows for a better understanding of buckle morphologies and highlights remarkable properties of these wavy blisters. In particular it is shown that the wavelength of the telephone cord patterns scales linearly with a parameter depending on the stress level in the film, the thickness and the adhesion energy. This result has an important practical application: since it is experimentally possible to control the stress level and the film thickness during elaboration, the measurement of this wavelength can indirectly lead to a measurement of the interface energy, even in very small films.

2. Mechanical model for blisters propagation

2.1. Model

A nonlinear plate model is used to capture the buckling equilibria. As a matter of fact, the blisters morphologies are characterized by very large out-of-plan displacements, meaning that the equilibria are located very far in the post-critical buckling regime compared to the flat (i.e. unbuckled) equilibrium. The thin film is modeled by a plate of thickness h, made of linear elastic material characterized by a Young's modulus E and Poisson's ratio ν. The surface of the plate is defined as the *(O,x,y)* plane and the out-of-plane displacement is denoted *w(x,y)*. In order to take into account the presence of the substrate, the unilateral contact condition $w(x,y) \geq 0$ is introduced. The calculations are made for large displacements *w*, using the Green Lagrange strain tensor. As only *w* is large, the strain tensor reduces to

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

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

The second key point is the description of adhesion rupture between the film and the substrate. A cohesive zone model (CZM) is introduced for this purpose between the elastic film and the rigid substrate. The tractions developed in the cohesive zone at the edges of the buckle provide boundary conditions for the buckled plate. The blister extends its boundary by forcing the separation along the delaminated edge as it buckles under large in-plane compression. Details of the cohesive zone model are given in [4]. A bilinear softening traction versus separation law is used, with a damage variable d increasing monotonically with the separation. The most important feature of this cohesive zone model is the mode mixity dependence of the interfacial toughness G^c. The relationship proposed by Hutchinson in [5] and based on experimental data[6] is used :

$$G^c(\psi) = G_I^c\left(1 + \tan^2 \eta\psi\right) \tag{2}$$

where ψ is the mode mixity angle defining the ratio between shear traction and normal traction at the interface ($\tan\psi = \dfrac{T_t}{T_n}$, with T_t the shear traction and T_n the normal traction. Hence, pure normal traction is obtained for ψ=0° whereas pure shear traction is obtained for ψ=90°). The definition given in Eq. (2) is meant to take into account that it is all the more difficult to break the interface that the proportion of shear is higher at the interface. Fig.1 describes both shapes of the traction versus separation law and of the toughness. Note that the quantity G_0, which is the energy per unit area stored in the film in its plane fully adherent state, has been introduced. G_0 is defined by:

$$G_0 = \frac{1-\nu}{E} h {\sigma_0}^2 \tag{3}$$

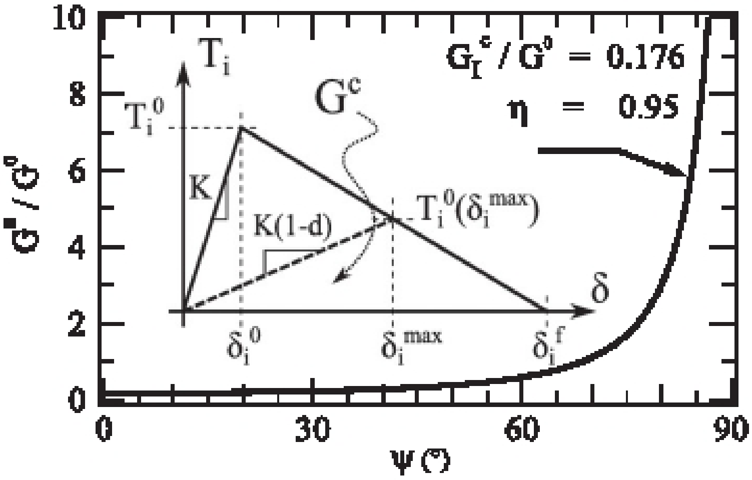

Figure 1 : From [4], Mixed-mode dependence of the work of adhesion at the film-substrate interface and bilinear traction vs separation law (Inset: i = n for mode I and t for mode II).

Finally, the thin film loading consists of an eigenstrain $\varepsilon_0 > 0$ uniformly applied to the plate ($\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_0$, $\varepsilon_{xy} = 0$). Since the film is relying on a rigid substrate, this is generating an equibiaxial stress state in the film ($\sigma_{xx} = \sigma_{yy} = -\frac{E}{1-\upsilon}\varepsilon_0 = \sigma_0, \sigma_{xy} = 0$). This loading is applied in the initial state. An explicit scheme is used for the calculation.
In order to trigger delamination, a small defect is created. This defect consists of an adhesion-free area. The blister is nucleated in this area.

2.2. Results

This model has been implemented in the finite element code ABAQUS [7]. As it has been described in [4], this model has been able to demonstrate the mechanism of propagation of the telephone cord blister. The specificity of this propagation comes from the pinning of the buckle front in mode II. As a matter of fact, as crack front is expanding in a circular shape with an increasing radius, a configurational instability occurs on the outer edge of the buckle front followed by the development of a strong mode II area. As G_c is much higher in mode II, the propagation is stopped in this area, which forms the pinning point responsible for the inversion of the curvature of the buckle and finally the wavy shape since the front has to go around this point for further propagation of the blister.
By varying the parameters of the model in the simulation, it is possible to obtain a wide variety of telephone cords buckles. Besides, the buckles are also obtained experimentally, with a setup allowing the control of the different parameters (film thickness h, residual stress σ_0, interface toughness G_I^c). The samples are molybdenum coatings deposited by magnetron sputtering on 30 µm thick silicon wafers. The deposition of a thin silver layer onto the substrate allows varying the adhesion (depending on the silver layer thickness). Immediately following, a 120 nm thick layer of

partially oxidized molybdenum, obtained by adding 2 sccm of O_2, is deposited. Controlling the oxygen content in the film leads to a fine tuning of the residual stress.

For the simulations, the elasticity parameters for the films are E=329 GPa and υ=0.31. The mode I interfacial toughness G_I^c is a variable, whereas $\eta = 0.95$. The calculations are carried out for different values of the film thickness h and the residual stress σ_0. Comparison between calculations results and experimental observations are reported in Fig. 2. Depending on the parameters, different aspect ratios (i.e. ratios between the wavelength and the width of the telephone cord) can be observed in the simulations. These ratios are usually observed to be ranging between 0.9 and 1.4, both in the simulations and in the experiments. Two cases are reported in Fig. 2: one large aspect ratio and one short aspect ratio.

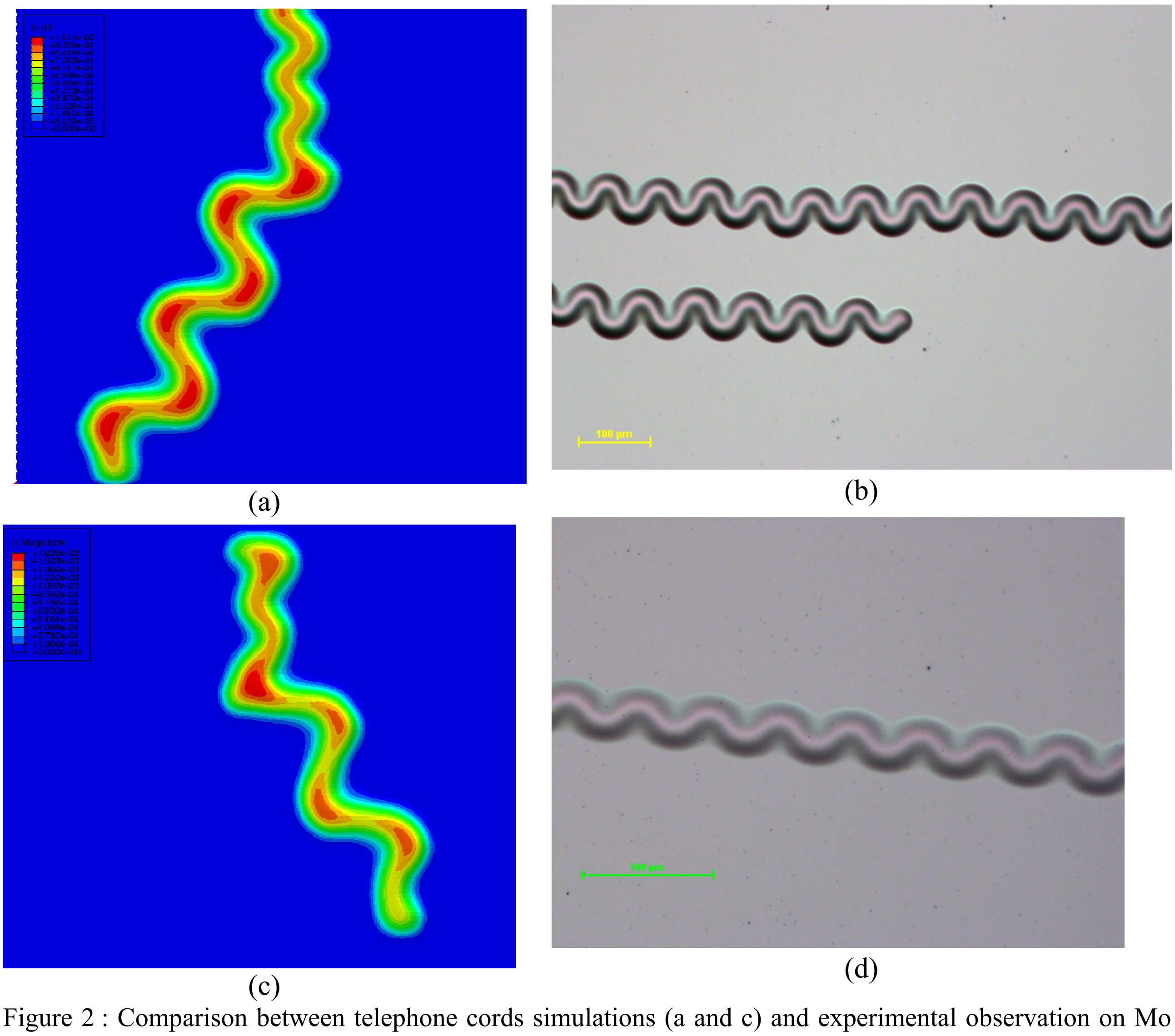

Figure 2 : Comparison between telephone cords simulations (a and c) and experimental observation on Mo thin films deposited on silicon wafers (b and d). The two upper cords are showing short aspect ratios (i.e. wavelength over width), whereas the two bottom cords have large aspect ratio.

(a): $h = 225nm$, $G_I^c = 1.35 J/m^2$, $\sigma_0 = 2.9GPa$, aspect ratio 1. (b): 600nm thick MoOx, potential -75V, aspect ratio 1. (c): $h = 300nm$, $G_I^c = 2.83 J/m^2$, $\sigma_0 = 3GPa$, aspect ratio 1.36. (d) 600nm thick MoOx floating potential, aspect ratio 1.2.

3. Scaling of the wavelength

A relationship between the wavelength λ of the telephone cords and the other parameters have been found. In order to express this relationship, it is convenient to introduce a limit mixity angle, denoted $\psi_{\lim}$. It corresponds to the mode mixity above which the interface cannot be fully separated. This is the case when the interface toughness is exceeding the stored energy ($G^c \geq G_0$), so the limit case is $G^c = G_0$. In this case, using Eq. (2) yields:

$$\psi_{\lim} = \frac{1}{\eta}\tan^{-1}\sqrt{\frac{G_0}{G_I^c} - 1} \tag{4}$$

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 $\psi_{\lim}$.

By similar arguments that can be found in [8], it is possible to show that the following non dimensional relationship is verified among the telephone cords equilibria.

$$\frac{\sigma_0}{\overline{E}}\left(\frac{\lambda}{h}\right)^2 = f(\psi_{\lim}) \tag{5}$$

So whatever the shape of the telephone cord, the wavelength can be expressed as $\lambda = h\sqrt{\frac{\overline{E}}{\sigma_0}} f(\psi_{\lim})$. The function f is determined numerically, giving the remarkable result exposed Fig. 3. It appears that λ is proportional to the quantity L defined as:

$$L = h\sqrt{\frac{\overline{E}}{\sigma_0}}\frac{1}{\sqrt{\tan\psi_{\lim}}} \tag{6}$$

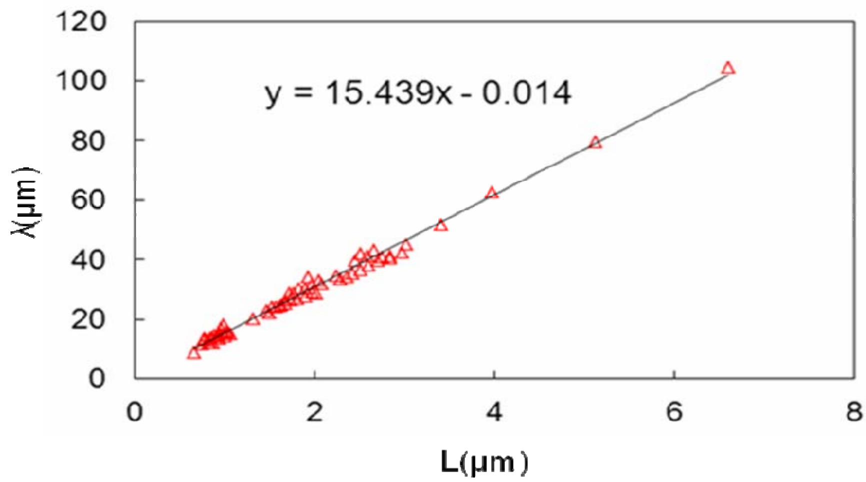

Figure 3 : Linear relationship between the telephone cord wavelength λ and the non dimensional quantity $L = h\sqrt{\frac{\overline{E}}{\sigma_0}}\frac{1}{\sqrt{\tan\psi_{\lim}}}$.

Once the relationship between λ and L is determined numerically, it is possible to find G_I^c from the experiments by measuring λ, assuming that σ_0, $\overline{E}$ and h are known, which is the case in our experiments.

4. Conclusions

We have a numerical model capable of describing the mechanisms of buckling driven delamination. A lot of patterns can be modeled, all observed in experiments. We have here emphasized the case of the telephone cords buckles, which is very often observed in experiments. The analysis of the telephone cords formation using the model has allowed for uncovering a very interesting relationship between the telephone cords wavelength, the stress level in the film, the thickness and the interface toughness. This result has an important practical application. When controlling the stress level and the film thickness during elaboration, the measurement of this wavelength can indirectly lead to a measurement of the interface toughness, even in very small films.

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