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Buckling Structures, A Relevant Signature of the Mechanical Properties of Film/Substrate Systems

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Abstract

Thin films and coatings are used in a wide range of technological applications, such as microelectronics, packaging or optics. They often develop high residual stresses during the deposition process, sometimes about few GPa in compression. Such large compressive stresses may cause the nucleation and growth of buckling structures that generally result in the loss of functional properties that were initially conferred to such film/substrate composites. The aim of our studies is consequently to have a better understanding of the buckling phenomenon, by identifying the relevant parameters to prevent, to limit, or to control its occurrence.

Introduction

The buckling phenomenon has been widely investigated by the past, both analytically in the framework of the Foppl-Von Karman theory of thin plates and numerically by finite elements simulations. The influence of usual mechanical parameters, such as thickness, (internal) stresses or film Young's modulus has been now clearly identified (see (Hutchinson & Suo, 1991) for a general review on buckling-induced delamination of plates). From an experimental point of view, the fine investigation of the morphology of the buckling structures is consequently of great interest in order to qualitatively, or even quantitatively, extract some physical/mechanical parameters of the films. A review on what can be concluded on some experimental observations of specific buckling structures is presented in the following.

Relevant experimental signatures

Elementary morphologies as markers of the iso/anisotropy of stress

The most common buckling structures are the circular blisters, the straight-sided buckles and the telephone cords, as observed in Fig. 1 on various film/substrate systems. A mapping of morphology established by finite elements simulations allows understanding the appearance of these elementary buckling structures (Parry et al., 2006) (Fig. 2). The mapping is given as a function of non-dimensional parameters, depending in particular of the anisotropy of stresses and/or of the film elastic properties. It is shown that the straight-sided buckles (resp. lines of blisters) are energetically favorable for high transversal stresses (resp. longitudinal stresses), while the stability domain for the telephone cords is mainly located close to the isotropy stress state. It clearly explains why the telephone cords are the most observed buckling structures, sometimes just after the deposition process for instance, since they are related to an isotropic stress.

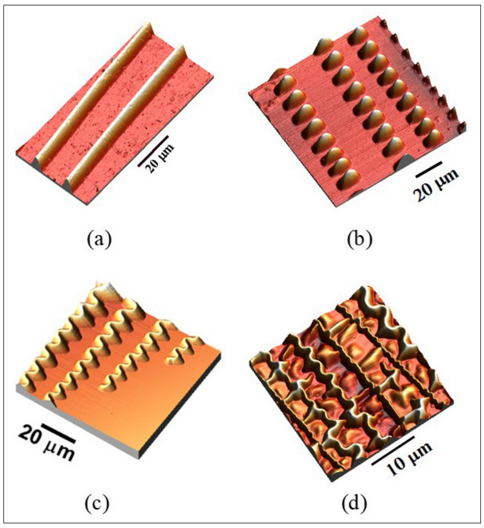


Figure 1: Various buckling structures observed by AFM (a) Straight-sided wrinkles in Ni 320 nm thick films on polycarbonate (b) Network of circular blisters in Au 150 nm thick films on Si (c) Telephone cords in Y_2O_3 50 nm thick films on GaAs (d) Branching structures in BN 160 nm thick films on NaCl.

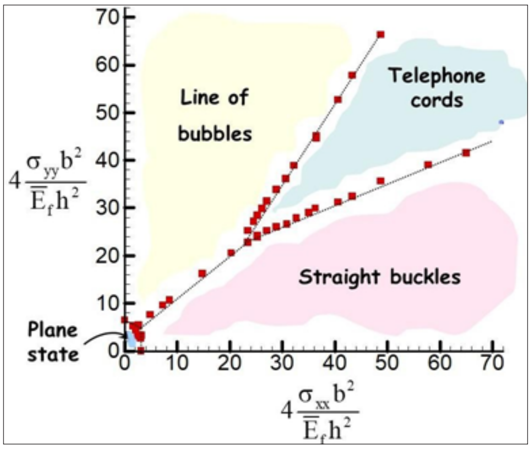


Figure 2: Mapping of elementary buckled morphologies obtained by finite elements simulations. h is the thickness of the film, 2b the width of the buckle, E_f the reduced Young's modulus of the film, σ_{xx} and σ_{yy} the transversal and longitudinal stresses, respectively.

Nanometer-scale depression as markers of the elastic film/ substrate contrast.

The effect of the elastic properties of the associated substrate has been also examined, particularly for stretchable device applications for which the Young's modulus contrast between the metallic film and the polymeric substrate can be very high. It has been shown both experimentally and numerically (Parry et al., 2005) that the critical stress for buckling to occur decreases with the increase of the soft character of the substrate. This mechanical response is associated with an enhanced maximum deflection (compared to the case of a rigid substrate) and the appearance of a nanometer scale depression on both sides of the buckle, just in front of the crack delamination (Fig. 3) (Parry et al., 2005). The equilibrium shape of the straight buckle can be no more described by the common sinusoidal function expected from the Foppl-Von Karman equations (case of a hard/stiff substrate with respect to the film). Finite elements simulations have shown (Boijoux et al., 2017) that the depression δ_2 over maximum deflection δ_1 ratio is related to the Dundurs' coefficient $\alpha = \frac{\overline{E}_f - \overline{E}_s}{\overline{E}_f + \overline{E}_s}$ that describes the elastic properties mismatch between the film and its substrate. α =-1 (resp. α =+1) corresponds to the case of a soft film on hard substrate (*resp*. of a hard film on a soft substrate). The depression over deflection ratio δ_2/δ_1 is found to be strongly increased with the increase of α. Taking advantage to numerical simulations (Boijoux et al., 2018), it is shown that α can be simply determined from a set of four parameters, namely δ_1, δ_2, h and b. The fine characterization of a buckle morphology can be consequently an interesting alternative method to the nanoindentation technique in the specific case of hard film on soft substrate for which the elastic properties of the film may be quite difficult to estimate.

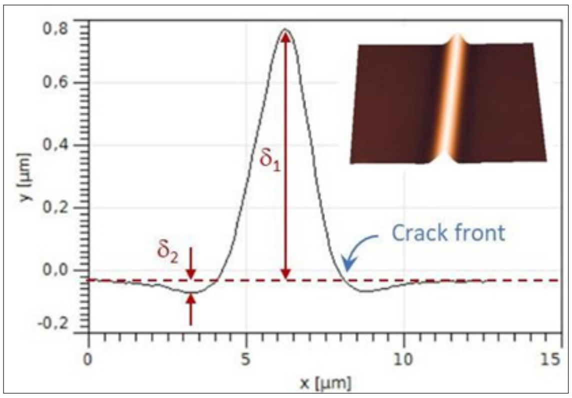


Figure 3: Experimental profile of a straight-sided buckle observed by atomic force microscopy on a Ni 100 nm thick film deposited on a polycarbonate substrate for which the elastic contrast is high (α > 0.9). h is the thickness of the film, 2b the width of the buckle, δ_1 and δ_2 are the maximum deflection and depression of the buckle, respectively.

Complex circular buckling structures as markers of int/out pressure mismatch.

Circular blisters are sometimes experimentally observed, but often at small scales. It is now established that the isotropic growth of a circular buckle is no more energetically favorable at large scales; they then develop instabilities at their circumference leading to flower-like structures that may then evolve to branching and finally full de- lamination of the films.

The possible pressure mismatch between the external and internal parts of the buck- led film was overlooked for many years, particularly because the effect was probably minimal and thus negligeable. It is noting that the idea of low pressure or vacuum (compared to the outside atmospheric one) below the buckling structures has been experimentally proved. It has thus been shown that the maximum deflection of straight- sided buckles slightly increases of only few tens of nanometers, after cutting/opening one side of the buckle by a focused ion beam (FIB) in order equalize the internal pressure with the external atmospheric one (Coupeau et al., 2010). Beyond a critical diameter of the circular buckle, the pressure mismatch may act to push some parts of the film downwards leading to a large variety of buckle morphology (Yu et al., 2021). The morphological mapping expected for a pressure mismatch Δp=1 atm (i.e. p_{ext}= 1 atm and p_{int}=0) is presented in Fig. 4 as a function of the normalized diameter $\overline{D} = D/h$ and normalized stress $\overline{\sigma} = \sigma/E$. As mentioned previously, the flower-like structures are energetically favorable for low diameter; beyond a critical diameter, the central part of the buckle is pushed down to the substrate leading to a polygonal shape those number of sides increases with the diameter. Note that more complex structures are observed for low values of stress, in pretty good agreement with experimental observations, such as the one presented in Fig. 5 (Goudeau et al., 2004).

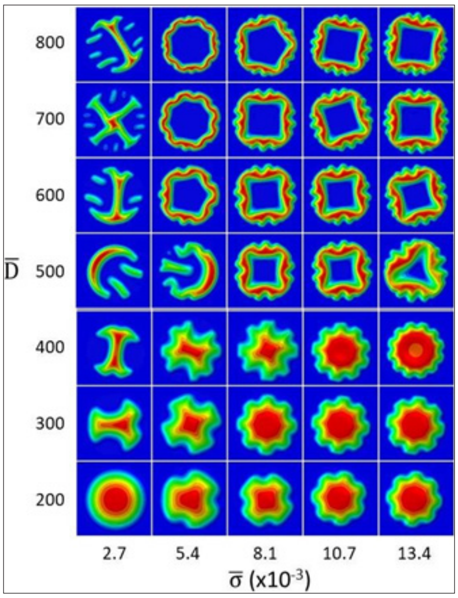


Figure 4: Morphological mapping determined by finite elements simulations of a circular buckle when a out/in pressure mismatch Δp=1 atm is considered, as a function of normalized diameter and stress.

Figure 5: Circular buckles induced by Ar irradiation on a Mo 200nm thick film de- posited by PVD on a Si wafer.

Donut- and croissant-like structure as markers of both pressure mismatch and plasticity

It is now established that plastic events inside the film and at their circumference allow stabilizing this circular morphology. The observation of circular buckles at large scales is thus a relevant marker of the occurrence of plastic events in the films. Such plastic events have been evidenced by molecular dynamic simulations (Durinck et al., 2008; Ruffini et al., 2013) and are characterized by a modified maximum deflection of buckles (as compared to a pure elastic response), in good agreement with experimental investigations on ductile films (Colin et al., 2009; Colin et al., 2007). Moreover, the combined effect of plastic events with a pressure mismatch mentioned previously (between the external and internal parts of the buckles) may now explain the formation of donut- and croissant-like structures (Hamade et al., 2015), experimentally observed on gold films in Fig. 6. As shown in Fig. 7, finite elements simulations have confirmed that a plastic folding all around a circular buckle allows first the enlargement of its stability domain, but also the donut-like and croissant-like buckles to be energetically stable. These two specific structures are however only expected if a pressure mismatch Δp between the outside and inside parts of the buckles is considered.

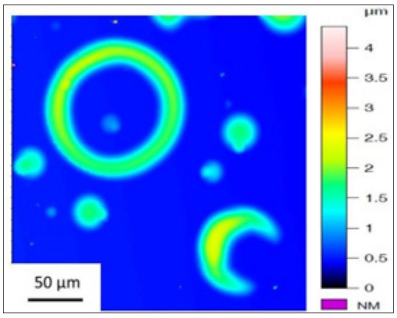
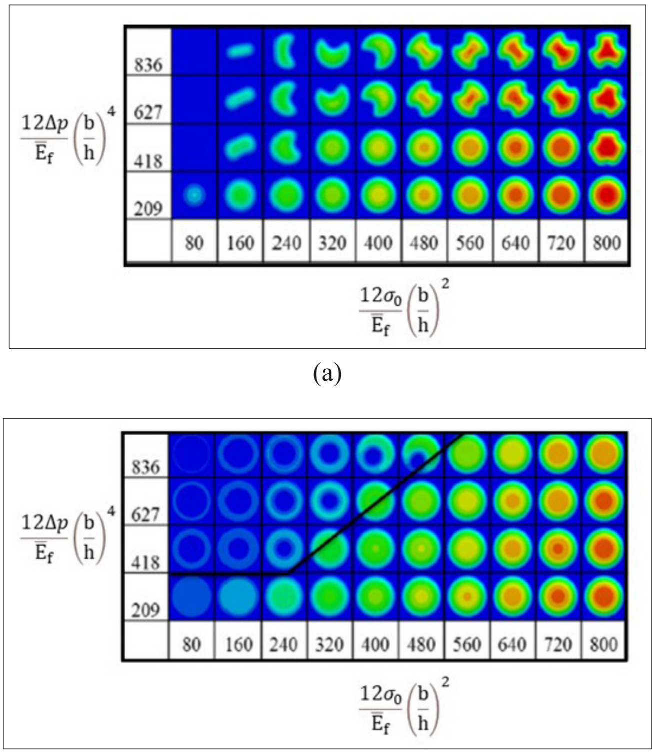


Figure 6: Circular, donut-like and croissant-like buckles experimentally observed by interferometric optical microscopy on Au 630 nm thick films on Si wafers.



(a)

(b)

Figure 7: Finite elements simulations mapping of circular blisters (a) without and (b) with a plastic folding around, as a function of normalized pressure and stresses.

Conclusion

The buckling of thin films and coatings is a damaging mechanism that we want to avoid, since it generally leads to the loss of functional properties initially conferred to the film/substrate system. It is however shown that the resulting structures can be exploited in ingenious ways to estimate some physical/mechanical properties of the related film/substrate systems. For instance, this applied to the internal stress in the film, the elastic contrast between the film and the substrate, the pressure mismatch between the outside and inside parts pf the buckle (i.e. the airtight nature of the film), the plastic events taking place in the film.

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