Plastic Folding of Buckling Structures

Jérôme Colin,* Christophe Coupeau, and Jean Grilhé

Laboratoire de Métallurgie Physique, UMR 6630 du CNRS, Université de Poitiers, BP 30179, 86962 Futuroscope Cedex, France

(Received 27 March 2007; published 25 July 2007)

Atomic force microscopy observations of the free surface of gold thin films deposited on silicon substrates have evidenced the buckling of the films and the formation of blister patterns undergoing plastic folding. The classical elastic buckling and plastic deformation of the films are analyzed in the framework of the Föppl–Von Kármán theory of thin plates introducing the notion of low-angle tilt boundaries and dislocation distributions to describe this folding effect. It is demonstrated that, in agreement with elementary plasticity of bent crystals, the presence of such tilt-boundaries results in the formation of buckling patterns of lower energy than "classical" elastic blisters.

DOI: 10.1103/PhysRevLett.99.046101 PACS numbers: 68.55.−a, 46.32.+x, 61.72.Lk

It is now well established that buckling of many macroscopic structures such as thin sheets can accurately be described in the framework of Föppl–Von Kármán's (FVK) theory of thin plates [1,2]. Recently, this theory has been applied at the mesoscopic scale to investigate the buckling patterns observed when thin films deposited on substrates or multilayers are submitted to compressive stresses, these structures being used for a number of engineering applications ranging from microelectronic devices to coatings in the fields of metallurgy and materials science [3–7]. A plethora of patterns has thus been already characterized, such as straight-sided wrinkles, telephone cords, or axi-symmetrical blisters taking advantage of atomic force microscopy (AFM) and have been investigated using continuum elasticity theory. Recently, the effect of substrate plasticity on the formation of straight-sided wrinkles has been investigated [8]. However, a number of problems among which is the study of the influence of elementary plasticity at the microscopic scale of crystalline thin films on their "pure" elastic buckling still remains mainly unexplored to the best knowledge of the authors. It is the purpose of this Letter to investigate how thin film elementary plasticity mechanisms can modify the classical elastic buckling patterns. This Letter is organized as follows. Experimental observations of the plastic folding of spontaneous buckling patterns generated in thin films deposited on substrates are first reported. The formation of these structures is then investigated in the framework of FVK's theory of thin plates introducing the notion of low-angle tilt boundaries characteristic of thin film plasticity at the microscopic scale.

AFM observations of a compressed polycrystalline gold thin film have been performed in contact mode using Si_3N_4 cantilevers. The 630 nm thick film has been deposited by a physical vapor deposition method on a (100) silicon substrate. Unusual shapes of buckling structures are evidenced on the whole specimen. These spontaneous buckling patterns are characterized by well-defined circular blisters, with a strong bending of the film localized at the circular boundary of the delaminated structures that is characteristic of an irreversible or plastic deformation [see Figs. 1(a) and 1(b)]. This plastic deformation is a commonly observed phenomenon for ductile metallic films [9] such as gold thin films [10]. The average stress in the planar part of the film has been estimated by curvature method to be $\approx$0.6 GPa; the small angle between the buckled and flat parts of the film is around 9 degrees.

In the following, the coupling effects between elastic buckling and plastic deformation have been investigated. For the sake of simplicity, one considers the simple case of one-dimensional wrinkle for a monocrystalline thin film. This approach can be generalized to axi-symmetrical blisters and polycrystalline thin films, assuming that the elementary plasticity mechanisms involved in the folding effect take place in the different grains of the blister circumference. Tilt boundary should be thus considered in each of these grains. The resultant "plastic" energy of the folding blister [equivalent to that derived in Eq. (10)] is then obtained by summing up the energy contribution of

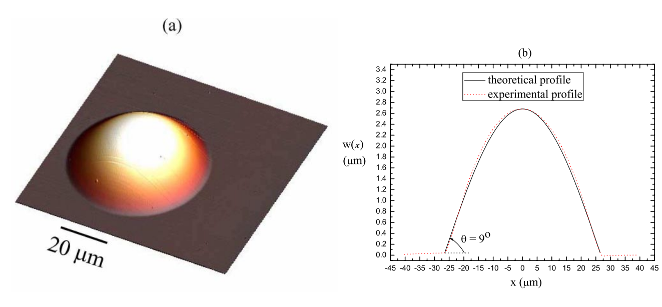

FIG. 1 (color online). (a) A circular blister exhibiting plastic folding is observed on the free surface of a gold thin film deposited on a silicon substrate. (b) Taking $\theta = 9$ degrees and $\sigma_0 = 0.61$ GPa, the theoretical profile of the wrinkle $[\mathrm{x} = x + u(x), w(x)]$ is superimposed to the cross-section of the buckled thin film.

each tilt-boundary of the considered grain distribution, the classical elastic energy (bending plus stretching) being also calculated for axi-symmetrical blister.

A straight-sided wrinkle lying along (0y) axis and undergoing plastic folding is thus considered (see Fig. 2 for axes). As usual, it is assumed that in its initial planar configuration, the film of thickness h is already delaminated on a distance $2b$. Since the ratio of Young's modulus of the film and silicon substrate, respectively, labeled $E_f = 78$ GPa and $E_s = 150$ GPa, is $E_f/E_s \approx 0.5$, the elasticity of the substrate is not considered [5,11,12]. The compressive reference stress in the initially flat thin film is defined as $\sigma_{xx}^0 = \sigma_{yy}^0 = -\sigma_0$, the total stress in the film $\sigma_{ij}^{\mathrm{tot}}$ is thus written as $\sigma_{ij}^{\mathrm{tot}} = \sigma_{ij}^0 + \sigma_{ij}$, with σ_{ij} the stress variation resulting from buckling [3]. The corresponding x, y, and z variations of the displacements from the reference state are labeled u, v, and w, respectively. Since the wrinkle is assumed to be infinite along (0y) axis, u, v, and w only depend on the x variable. The nonlinear FVK's theory of thin plates is used to investigate the wrinkle formation. Within this framework, the w component of displacement field satisfies [3]

$$w^{(4)} + \lambda^2 w^{(2)} = 0, \tag{1}$$

with $w^{(i)} = \partial^i w/\partial x^i$ and $\lambda = \sqrt{h(\sigma_0 - \sigma_{xx})/D}$. The bending stiffness D is defined as $D = E_f h^3/[12(1-\nu_f^2)]$ with ν_f the Poisson's ratio of the film. The boundary conditions write

$$u(\pm b) = v(\pm b) = w(\pm b) = 0, \quad \frac{\partial w}{\partial x}\Big|_{x=\pm b} = \mp\theta, \tag{2}$$

with θ the small positive angle at the edges between the buckled thin film and the flat substrate (see Fig. 2). The complete setting of the problem requires us to consider two supplementary compatibility equations leading to [2,3]

$$\sigma_{yy} = \nu_f \sigma_{xx}, \qquad \frac{\partial u}{\partial x} + \frac{1}{2}\left(\frac{\partial w}{\partial x}\right)^2 = \frac{1-\nu_f^2}{E_f}\sigma_{xx}. \tag{3}$$

Using Eqs. (1)–(3), one finds the following displacement field characterizing the shape of the buckled structure:

$$u(x) = \frac{1-\nu_f^2}{2E_f}\left(\sigma_0 - \frac{\lambda^2 b^2}{\pi^2}\sigma_c\right) b \frac{\sin 2\lambda x - \frac{x}{b}\sin 2\lambda b}{\lambda b - \sin\lambda b\cos\lambda b}, \tag{4}$$

$$w(x) = \frac{h}{2}\sqrt{\frac{4}{3}\frac{\sigma_0 - \frac{\lambda^2 b^2}{\pi^2}\sigma_c}{\sigma_c}\frac{\pi^2}{\lambda^2 b^2}\frac{\sin^2\lambda b}{1 - \frac{\sin\lambda b\cos\lambda b}{\lambda b}}}\frac{\cos\lambda x - \cos\lambda b}{\sin\lambda b}, \tag{5}$$

with $\sigma_c = \pi^2 h^2 E_f/[12(1-\nu_f^2)b^2]$ the classical critical stress above which the Euler column (with $\theta = 0$) appears [3]. The compatibility Eq. (3) finally writes

$$\theta = \sqrt{\frac{4(1-\nu_f^2)}{E_f}\left(\sigma_0 - \frac{\lambda^2 b^2}{\pi^2}\sigma_c\right)\frac{\sin^2\lambda b}{1 - \frac{\sin\lambda b\cos\lambda b}{\lambda b}}}. \tag{6}$$

Once θ is known, Eq. (6) allows for determining a discrete set of λ parameters, each of them being associated with a specific buckling profile. According to the experimentally observed morphology of buckling patterns shown in Fig. 1(a) for gold thin films, one focuses on the study of λ values such that $\lambda \leq \pi/b$, the Euler column with $\theta = 0$ being obtained for $\lambda = \pi/b$. The cases $\lambda > \pi/b$ leading to multioscillating profiles are not studied in this Letter. It can also be emphasized that when $\lambda \to 0$, a wrinkle with $\theta \neq 0$ is still obtained with a limit amplitude given by $\frac{h}{2} \times \frac{\pi}{\sqrt{2}}\sqrt{\frac{\sigma_0}{\sigma_c}}$. For each value of θ, Eq. (6) allows for determining a first restriction on σ_0 variations. One gets $\sigma_0 \geq \sigma_0^* = \frac{E_f}{6(1-\nu_f^2)}\theta^2$. It can be thus deduced that as θ decreases, σ_0^* may be smaller than the critical stress σ_c above which the Euler column appears. In this Letter, based on energetic consideration, the formation of the plastically deformed structure is compared with respect to planar configuration when $\sigma_c \geq \sigma_0 \geq \sigma_0^*$ and with respect to classical Euler column when $\sigma_0 \geq \sigma_0^* > \sigma_c$.

Following Landau and Lifchitz [13], the bending and stretching energy variations with respect to the flat configuration, defined per unit length along (0y) axis and, respectively, labeled ΔE_{bend} and ΔE_{stre}, can easily be determined in the case where u and w only depend on x using the relationships:

$$\Delta E_{\mathrm{bend}} = \frac{E_f h^3}{24(1-\nu_f^2)}\int_{-b}^{+b}\left(\frac{\partial^2 w}{\partial x^2}\right)^2 dx, \tag{7}$$

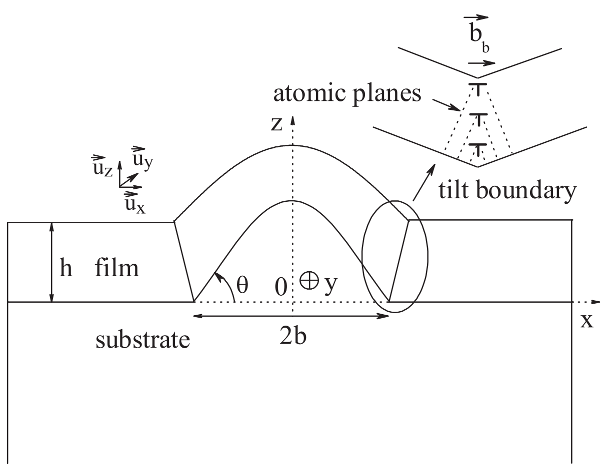

FIG. 2. The scale along (0z) has been enlarged. A straight-sided wrinkle undergoing plastic folding is considered for a thin film of thickness h deposited on a rigid substrate. Two low-angle tilt boundaries made of distributions of edge dislocations are considered leading to crystal folding.

$$\Delta E_{\mathrm{stre}} = \frac{h}{2}\int_{-b}^{+b}\left[-2\frac{\partial u}{\partial x} + \frac{E_f}{1-\nu_f^2}\left(\frac{\partial u}{\partial x}\right)^2 - \sigma_0\left(\frac{\partial w}{\partial x}\right)^2 + \frac{E_f}{1-\nu_f^2}\left(\frac{\partial w}{\partial x}\right)^2\frac{\partial u}{\partial x} + \frac{E_f}{4(1-\nu_f^2)}\left(\frac{\partial w}{\partial x}\right)^4\right]dx. \quad (8)$$

Introducing the expressions of u and w given by Eqs. (4) and (5) in Eqs. (7) and (8), one gets $E_b = \Delta E_{\mathrm{bend}} + \Delta E_{\mathrm{stre}}$, the total buckling energy variation with respect to the flat configuration per unit length along $(0y)$ axis:

$$\Delta E_b = \frac{h}{288b^2}\left[\frac{48bh^2\sigma_0(-\pi^2\sigma_0+\lambda^2b^2\sigma_c)}{\sigma_c} - \frac{4bE_fh^4\lambda^2(-\pi^2\sigma_0+\lambda^2b^2\sigma_c)(2\lambda b+\sin 2\lambda b)}{\sigma_c(1-\nu_f^2)(2\lambda b-\sin 2\lambda b)} + \frac{E_fh^4\lambda(-\pi^2\sigma_0+\lambda^2b^2\sigma_c)^2(12\lambda b-8\sin 2\lambda b+\sin 4\lambda b)}{\sigma_c^2(1-\nu_f^2)(2\lambda b-\sin 2\lambda b)^2} - \frac{24bh^2(-\pi^2\sigma_0+\lambda^2b^2\sigma_c)^2(-1+4\lambda^2b^2+\cos 4\lambda b+\lambda b\sin 4\lambda b)}{\pi^2\sigma_c(2\lambda b-\sin 2\lambda b)^2} + \frac{144b^3(-\pi^2\sigma_0+\lambda^2b^2\sigma_c)^2(1-\nu_f^2)(-1+4\lambda^2b^2+\cos 4\lambda b+\lambda b\sin 4\lambda b)}{\pi^4E_f(2\lambda b-\sin 2\lambda b)^2}\right]. \quad (9)$$

Taking $\lambda = \pi/b$ (or $\theta = 0$) in Eq. (9), ΔE_b reduces to $\Delta E_b^{\mathrm{Euler}}$ the energy variation associated with the formation of the pure elastic Euler column [3].

It is well admitted that in crystals, individual grains contain rotated subgrains whose sub-boundaries are made of distributions of dislocations (see [14–18] and refs. therein). Likewise, when a crystal is plastically bent of a small angle θ along one axis, the dislocations rearrange themselves in such a way that in the lower configuration of energy, sub-boundaries or tilt boundaries are formed which separate stress-free blocks of matter. In this Letter, such low-angle tilt boundaries are considered in the bent parts of the film located at the edges of the wrinkle where plastic deformation takes place (see Fig. 2). One tilt boundary is introduced at each edge, which is composed of a distribution along $(0z)$ axis of infinite straight edge dislocations lying along $(0y)$ axis of Burgers' vector $b_b\vec{u}_x$ and separated by a distance d such that $d = b_b/\theta$ [15]. The corresponding supplementary energy $E_{2\mathrm{tb}}$ resulting from the formation of the two considered low-angle tilt boundaries yields [15,18]

$$E_{2\mathrm{tb}}(\theta, h) = \frac{h}{4\pi}\frac{E_f}{1-\nu_f^2}b_b\theta\log\left(\frac{1}{2\pi}\frac{e}{\theta}\right), \quad (10)$$

with θ the angle given by Eq. (6). In order to determine in which conditions the plastic folding and buckling is favorable with respect to planar thin film or Euler column, the energy variations $\Delta E_1 = \Delta E_b + E_{2\mathrm{tb}}$ (when $\sigma_c \geq \sigma_0 \geq \sigma_0^*$) and $\Delta E_2 = \Delta E_b + E_{2\mathrm{tb}} - \Delta E_b^{\mathrm{Euler}}$ (when $\sigma_0 \geq \sigma_0^* > \sigma_c$) have been, respectively, studied as a function of σ_0 for different values of θ in Figs. 3(a) and 3(b). The experimental parameters of the considered gold thin film on silicon substrate are taken as follows: $\nu_f = 0.44$ $h = 630$ nm, $b = 26750$ nm, $\sigma_c = 44.1$ MPa, $b_b = 0.28$ nm. In Fig. 3(a), it is observed that for small values of θ angle (smaller than 3 degrees with the considered physical parameters), there exists a critical stress σ_0^c above which the energy variation ΔE_1 is negative. The plastic folding (+ buckling) become thus energetically favorable with

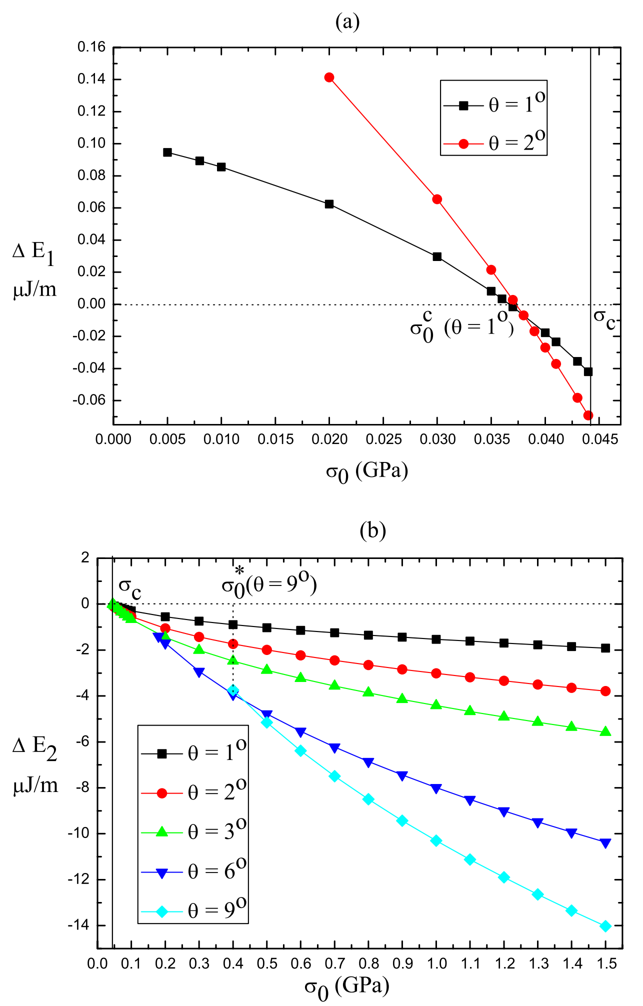

FIG. 3 (color online). (a) ΔE_1 versus σ_0 when $\sigma_0 \leq \sigma_c$. (b) ΔE_2 versus σ_0 for $\sigma_0 > \sigma_c$.

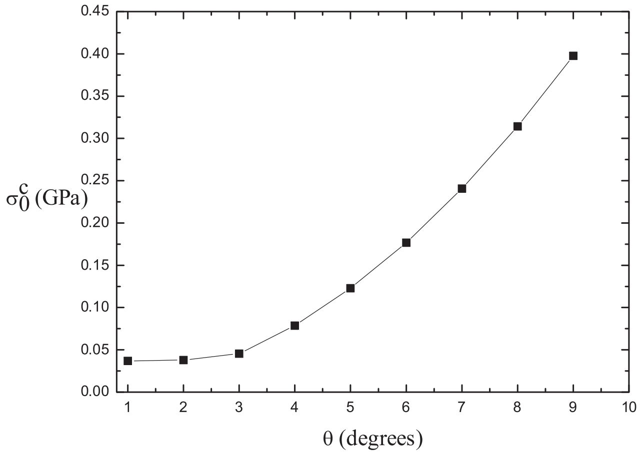

FIG. 4. Critical stress σ_0^c versus θ. When $\theta < 3°$, the reference state is the planar thin film. When $\theta \geq 3°$, the reference state is the Euler column.

respect to planar configuration in a range of stress σ_0 where the classical Euler column can not develop. In Fig. 3(b), it is found that for $\theta < 3°$, ΔE_2 is always negative. For $\theta = 6°$ and $9°$, ΔE_2 is negative for σ_0 greater than the critical value $\sigma_0^c = \sigma_0^*$. For $\theta = 3°$, one finds $\sigma_0^c \approx \sigma_0^* \approx 1.0016\sigma_c$. As a consequence, the formation of the plastically deformed structure becomes energetically favorable with respect to Euler column as soon as the stress is greater than its critical threshold. It can be thus first concluded that irrespective of σ_c value, one can define for each value of θ angle a range of stress σ_0 where the plastically fold structure is susceptible to develop. The variation of the critical stress σ_0^c versus θ has been finally plotted in Fig. 4. It can be observed that σ_0^c increases with θ, so that it becomes harder in terms of stress to develop plastically deformed structures as θ increases.

In the case of gold thin films on silicon substrates, taking for stress $\sigma_0 = 0.61$ GPa and angle $\theta = 9$ degrees, it is found from Fig. 3(b) that the energy variation ΔE_2 associated with the formation of a wrinkle is negative. As a consequence, our simplified 1D model clearly justifies the formation of the plastically folded wrinkle resulting from thin film buckling which has been experimentally observed [see Fig. 1(a)]. The model is also relevant to determine accurate values of the characteristic length scales of the buckling patterns. In particular, it can be observed in Fig. 1(b) that the theoretical profile $[x + u(x), w(x)]$ given by Eqs. (4) and (5) matches the experimental one.

As a conclusion, it is demonstrated in this Letter from both an experimental and theoretical point of view that folding of thin films deposited on substrates may strongly modify the buckling patterns when plasticity can occur depending on dislocation nucleation and mobility. It is found, in particular, that below the classical stress threshold of Euler column σ_c, a plastically folded wrinkle with small angle value may appear for thin film stress σ_0 greater than the critical value σ_0^c. Above the Euler column threshold, the plastically deformed wrinkle is found to be energetically favorable with respect to Euler column (when $\sigma_0 \geq \sigma_0^c$). It is finally believed that the coupling effects between elasticity and plasticity may also play an important role in the formation of other buckling patterns such as those resulting from the vertical superimposition of blisters.

The authors gratefully acknowledge discussions with F. Foucher.

*jerome.colin@univ-poitiers.fr

[1] M. Ben Amar and Y. Pomeau, Proc. R. Soc. Lond. A **453**, 729 (1997).
[2] B. Audoly, Phys. Rev. Lett. **83**, 4124 (1999).
[3] J. W. Hutchinson and Z. Suo, Adv. Appl. Mech. **29**, 63 (1992).
[4] M. W. Moon, H. M. Jensen, J. W. Hutchinson, K. H. Oh, and A. G. Evans, J. Mech. Phys. Solids **50**, 2355 (2002).
[5] M. W. Moon, K. R. Lee, K. H. Oh, and J. W. Hutchinson, Acta Mater. **52**, 3151 (2004).
[6] G. Parry, A. Cimetière, C. Coupeau, J. Colin, and J. Grilhé, Phys. Rev. E **74**, 066601 (2006).
[7] E. A. Jagla, Phys. Rev. B **75**, 085405 (2007).
[8] F. Foucher, C. Coupeau, J. Colin, A. Cimetière, and J. Grilhé, Phys. Rev. Lett. **97**, 096101 (2006).
[9] R. Spolenak *et al.*, Phys. Rev. Lett. **90**, 096102 (2003).
[10] R. W. Cheary, E. Dooryhee, P. Lynch, N. Armstrong, and S. Dligatch, J. Appl. Crystallogr. **33**, 1271 (2000).
[11] B. Cotterell and Z. Chen, Int. J. Fract. **104**, 169 (2000).
[12] H. H. Yu and J. W. Hutchinson, Int. J. Fract. **113**, 39 (2002).
[13] L. D. Landau and E. M. Lifchitz, *Theory of Elasticity* (Pergamon, New York, 1986), 3rd ed..
[14] F. R. N. Nabarro, *Theory of Crystal Dislocations* (Oxford University Press, Ely House, London W. 1, 1967).
[15] J. P. Hirth and J. Lothe, *Theory of Dislocations* (Wiley, New York, 1992), 2nd ed..
[16] J. Friedel, *Dislocations* (Pergamon Press, London W.1, 1964).
[17] S. Amelinckx, *Solid State Phys. Suppl. 6: 1* (Academic Press, New York, London, 1964).
[18] W. T. Read and W. Shockley, Phys. Rev. B **78**, 275 (1950).