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Buckling of Stressed and Pressurized Thin Films on Substrates

The buckling solutions for a stressed thin film deposited on a semi-infinite rigid substrate have been determined in the framework of the Föppl–von Karman's theory of thin plates and the perturbed bifurcation theory when pressures are applied onto the lower and upper free surfaces of the buckled film. It is found that the equilibrium solutions of the film are modified compared with the classical case of the Euler column, as well as the critical stress above which the film buckles. [DOI: 10.1115/1.4000923]

1 Introduction

Thin films on substrates and multilayers are currently used for their functional properties in an increasing number of engineering fields such as micro-electronics or aeronautics, for example [1]. It is now well-admitted by the metallurgical and materials science communities that the reliability of such coatings is strongly dependent on their mechanical behavior and their stability [2]. Recently, the classical Föppl–von Karman (FvK) theory of thin plates initially developed to study macroscopic structures in engineering [3,4] has provided a natural theoretical framework to investigate the buckling of thin films on substrates at the mesoscale [5,6]. Indeed, in parallel with atomic force microscopy (AFM) observations of surfaces of coatings [7], a number of configurations have been characterized, such as circular blisters, straight-sided wrinkles, or telephone-cord structures [8–11], under the action of different sources of stress such as the residual stress coming from the deposition methods [12], the thermal coefficient mismatch, or external applied stresses [13–15]. The effects of substrate elasticity have been also investigated by means of finite elements simulations [16–19]. It is found that the shape of the buckling patterns and the critical stress for buckling are strongly modified when the deformation of the substrate is considered [20]. More recently, several effects of plasticity [21], such as the formation of low-tilted boundaries, have been integrated in the FvK theory of thin plates to explain the folding of buckling patterns observed on the surface of gold thin films deposited on silicon substrates [22].

In micro-electronics industry, the bulge test consisting in studying the deformation of clamped thin films under controlled pressure is widely used to characterize the film/substrate adherence of the interfaces such as oxide/metal interfaces, for example [23]. Likewise, hydrogen or helium implantation performed in semiconductors is well-known to induce pressurized gas bubbles under specific conditions of irradiation. These bubbles are used in the smart-cut process to delaminate the wafer, resulting in low-roughness surfaces [24,25]. Moreover, materials used in fusion reactors are commonly subjected to buckling and peeling under 14 MeV neutron irradiations [26]. From these experiments, one question arises that concerns the influence of pressure on the buckling process of the films. In this paper, we investigate in the framework of FvK theory of thin plates [6,27] and perturbed bifurcation theory [28], the buckling of stressed thin films on substrates, considering a pressure between the delaminated film and the substrate. In particular, one focuses on the effects of pressure on the critical stress above which the film buckles as well as on the deflection and the profiles of the buckles. The morphological evolution of the structures is finally discussed.

2 Modeling

A thin film of thickness h assumed to be small compared with the other dimensions of the problem is considered on a semi-infinite substrate (see Fig. 1 for axes). Following Hutchinson and Suo [1], an initial compression stress $\sigma_{xx}^0=-\sigma_0$ is considered with σ_0 a constant parameter. The thin film is initially delaminated on a distance $2b$. The pressure mismatch is defined as $\Delta p=p_{\text{int}}-p_{\text{ext}}$ with p_{int} the pressure between the substrate and the delaminated film, and p_{ext} the pressure acting on the upper part of the film. The variations of the displacement field of the buckled film from the reference state are labeled u, v, and w along the (Ox), (Oy), and (Oz) axes, respectively. Since a 2D buckled structure lying along the (Oy) axis is considered, the v component is zero and u, w only depend on the x variable. In this case, the mechanical equilibrium of the film assimilated to a thin plate reads [3,4]

$$\frac{\partial^4 w}{\partial x^4}-\frac{h}{D}\sigma_{xx}\frac{\partial^2 w}{\partial x^2}=\frac{\Delta p}{D} \tag{1}$$

where σ_{xx} is the stress in the buckled film found to be constant [3,4]. The bending stiffness is defined as $D=Eh^3/[12(1-\nu^2)]$, with ν as the Poisson ratio and E as the Young modulus of the film. The complete set of equations for the wrinkle requires to consider the compatibility equations given by

$$\begin{aligned}\frac{1-\nu^2}{E}(\sigma_{xx}+\sigma_0)&=\frac{\partial u}{\partial x}+\frac{1}{2}\left(\frac{\partial w}{\partial x}\right)^2\\ \sigma_{yy}&=\nu\sigma_{xx}\end{aligned} \tag{2}$$

and the following boundary condition:

$$u(\pm b)=w(\pm b)=0,\quad \frac{\partial w}{\partial x}(\pm b)=0 \tag{3}$$

In Secs. 3–5, the solutions of Eq. (1) have been determined in the case where the buckled film under pressure ($\Delta p\neq 0$) is unstressed

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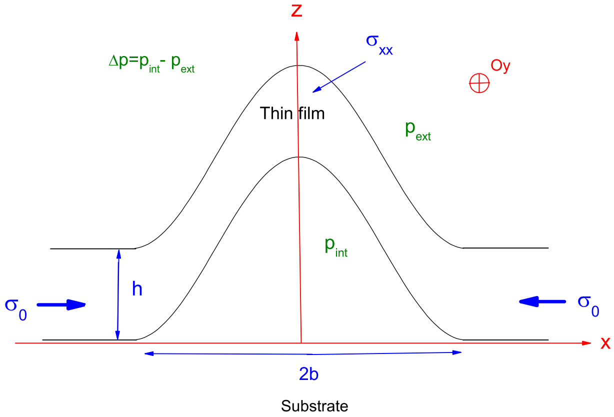

Fig. 1 A thin film of thickness h under compression stress σ_0 is considered on a stiff substrate. The film/substrate interface is initially delaminated on a distance $2b$ along the (Ox) axis. The pressure mismatch is defined as $\Delta p = p_{int} - p_{ext}$ with p_{int} the pressure between the substrate and the film, and p_{ext} the pressure acting on the upper part of the film. σ_{xx} corresponds to the stress in the buckled part.

($\sigma_{xx}=0$), in tension ($\sigma_{xx}>0$), or under compression ($\sigma_{xx}<0$).

3 A Fully Relaxed Buckled Film

For a critical value Δp_c, the film can be completely relaxed. This is obtained when σ_{xx} is cancelled in Eq. (1) for $\Delta p \equiv \Delta p_c$. It yields

$$\frac{\partial^4 w}{\partial x^4} = \frac{\Delta p_c}{D} \tag{4}$$

The general polynomial solution of the above Eq. (4) is given by

$$w(x) = \frac{\Delta p_c x^4}{24D} + c_0 + c_1 x + c_2 x^2 + c_3 x^3 \tag{5}$$

The profile of the wrinkle being symmetric with respect to (Oz) axis, one takes $c_1 = c_3 = 0$. The coefficients c_0 and c_2 have been determined using Eq. (3) and the profile of the wrinkle finally reads

$$w(x) = \frac{\Delta p_c}{24D}[(x^4 - b^4) - 2b^2(x^2 - b^2)] \tag{6}$$

with the following maximum deflection $\delta_c = w(x=0)$

$$\delta_c = \frac{\Delta p_c b^4}{24D} \tag{7}$$

The critical pressure variation Δp_c is determined by integrating Eq. (2) with respect to x between $-b$ and $+b$

$$\frac{1-\nu^2}{E}(\sigma_0)2b = \frac{1}{2}\int_{-b}^{+b}\left(\frac{\partial w}{\partial x}\right)^2 dx + u(b) - u(-b) \tag{8}$$

and using the boundary conditions defined in Eq. (3) and the expression of the $w(x)$ profile given in Eq. (6). One gets

$$\Delta p_c = \sqrt{\frac{105\sigma_0 E}{16(1-\nu^2)}}\left(\frac{h}{b}\right)^3 \tag{9}$$

It is emphasized that when the pressure mismatch Δp is smaller than this critical value Δp_c defined in Eq. (9), the buckled film is in compression. When $\Delta p > \Delta p_c$ the buckled film is in tension.

4 A Buckled Film Under Tension

Assuming now $\sigma_{xx}>0$ with $\alpha^2 = (h/D)\sigma_{xx}$, Eq. (1) becomes

$$\frac{\partial^4 w}{\partial x^4} - \alpha^2 \frac{\partial^2 w}{\partial x^2} = \frac{\Delta p}{D} \tag{10}$$

whose general solution is given by

$$w(x) = c_1 \cosh \alpha x + c_2 + c_3 \sinh \alpha x + c_4 x + \frac{\Delta p}{2\alpha^2 D}x^2 \tag{11}$$

For symmetry reason the c_3 and c_4 coefficients cancel, the other two c_i with $i=1, 2$ are determined with the help of the boundary conditions (3). Finally, the profile function w has been found to be

$$w(x) = \frac{\Delta p b^2}{2\beta^2 D}\left[\frac{2b^2}{\beta \sinh \beta}\left(\cosh\frac{\beta x}{b} - \cosh \beta\right) + (b^2 - x^2)\right] \tag{12}$$

with $\beta = \alpha b$. The maximum deflection δ is given by

$$\delta(\beta) = \frac{\Delta p b^2}{2\beta^2 D}\left[\frac{2b^2}{\beta \sinh \beta}(1 - \cosh \beta) + b^2\right] \tag{13}$$

The domain of variation in β is defined such that the w profile must be positive for $-b \le x \le +b$. It leads to

$$\Delta p\left(\frac{\beta}{\tanh \beta} - 1\right) \ge 0 \tag{14}$$

To determine the relation between β, Δp, and σ_0, the compatibility Eq. (2) must be integrated between $-b$ and $+b$ with respect to x. The calculation gives

$$\sigma_0(\beta) = -\frac{D\beta^2}{hb^2} + \frac{E}{1-\nu^2}\left(\frac{\Delta p b^3}{2\beta^2 D}\right)^2\left[\frac{2}{3} - \frac{1}{\sinh^2 \beta} + \frac{4}{\beta^2} - \frac{3 \coth \beta}{\beta}\right] \tag{15}$$

With the help of Eq. (15), the pressure variation is determined to be

$$\Delta p(\beta) = \frac{2D\beta^2}{b^3}\sqrt{\frac{\left(\sigma_0 + \frac{D\beta^2}{hb^2}\right)\left(\frac{1-\nu^2}{E}\right)}{\frac{2}{3} - \frac{1}{\sinh^2 \beta} + \frac{4}{\beta^2} - \frac{3 \coth \beta}{\beta}}} \tag{16}$$

It is worth noting that contrary to the case of the Euler column where α is quantified [1], β appears here as a free parameter. The case of a nickel 400 nm thick film ($E=200$ GPa and $\nu=0.312$) has been considered in the following. The half-width of the delamination zone is fixed arbitrary to $b=10h$. Using Eqs. (13) and (15), the maximum deflection $\delta(\beta)$ has been parametrically plotted as a function of $\sigma_0(\beta)$ in the case where the pressure mismatch Δp on the buckled film under tension is positive and equal to 0.02, 0.04, and 0.08 GPa (Fig. 2). As expected, the deflection of the wrinkle increases with increasing σ_0. The values of σ_0 corresponding to the maximum of relaxation is also an increasing function of Δp, taking 0.27, 1.1, and 4.4 GPa for $\Delta p=0.02$, 0.04, and 0.08 GPa, respectively, as expected by Eq. (9). The solutions for such a configuration ($\sigma_{xx}>0$ and $\Delta p>0$ in this section) can provide a natural framework to study relevant problems such as the mechanical thin film evolution during bulge test (using Eqs. (13), (15), and (16)) where an external pressure between the film and the substrate is applied. Irradiation of materials resulting in the formation of gas cavities below the free surfaces can be also addressed within this framework.

5 A Buckled Film Under Compression

In this last case, one has $\sigma_{xx}<0$ with $\alpha^2 = -(h/D)\sigma_{xx}$. The equilibrium Eq. (1) of the film becomes:

$$\frac{\partial^4 w}{\partial x^4} + \alpha^2 \frac{\partial^2 w}{\partial x^2} = \frac{\Delta p}{D} \tag{17}$$

and the general solution for the w profile is given by

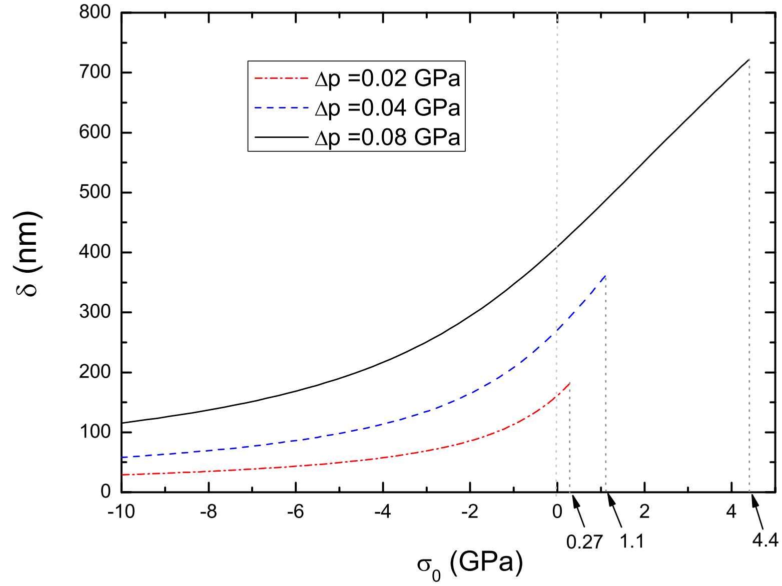

Fig. 2 Case of a nickel thin film under tension. δ versus σ_0 for a positive pressure mismatch Δp=0.02, 0.04, and 0.08 GPa. The stress values for the fully relaxed films are σ_0=0.27, 1.1, and 4.4 GPa, respectively. E=200 GPa, ν=0.312, and b=10h with h=400 nm.

$$w(x) = c_1 \cos \alpha x + c_2 + c_3 \sin \alpha x + c_4 x + \frac{\Delta p}{2\alpha^2 D} x^2 \tag{18}$$

The boundary conditions (3) allow to determine like in the previous sections, the profile w of the film, the maximum deflection δ, the stress σ_0, and the pressure mismatch Δp versus β. One finds

$$w(x) = \frac{\Delta p b^2}{2\beta^2 D}\left[\frac{2b^2}{\beta \sin \beta}\left(\cos\frac{\beta x}{b} - \cos \beta\right) + (x^2 - b^2)\right] \tag{19}$$

$$\delta(\beta) = \frac{\Delta p b^2}{2\beta^2 D}\left[\frac{2b^2}{\beta \sin \beta}(1 - \cos \beta) - b^2\right] \tag{20}$$

$$\sigma_0(\beta) = \frac{D\beta^2}{hb^2} + \frac{E}{1-\nu^2}\left(\frac{\Delta p b^3}{2\beta^2 D}\right)^2\left[\frac{2}{3} + \frac{1}{\sin^2 \beta} - \frac{4}{\beta^2} + \frac{3 \cot \beta}{\beta}\right] \tag{21}$$

$$\Delta p(\beta) = \frac{2D\beta^2}{b^3}\sqrt{\frac{\left(\sigma_0 - \frac{D\beta^2}{hb^2}\right)\left(\frac{1-\nu^2}{E}\right)}{\frac{2}{3} + \frac{1}{\sin^2 \beta} - \frac{4}{\beta^2} + \frac{3 \cot \beta}{\beta}}} \tag{22}$$

Assuming that $w>0$ for $-b<x<+b$, the domain of variation in β is given by

$$\Delta p\left(1 - \frac{\beta}{\tan \beta}\right) \geq 0 \tag{23}$$

It can be noticed that the results obtained in the case of a buckled film in compression (see Eqs. (20)–(22)) can be derived from the case in tension changing β by $i\beta$ in the Eqs. (13), (15), and (16).

5.1 Case of a Positive Pressure Mismatch. In the case of a nickel thin film considered in Sec. 5, straight-sided buckling structures are frequently observed [16,19]. In the following, a value of b=4.275 μm corresponding to experimental AFM observations is used [29]. In Fig. 3(a), the maximum deflection $\delta(\beta)$ has been parametrically plotted as a function of $\sigma_0(\beta)$ in the case where the pressure variation on the buckled film under compression is positive and zero. For a thin film on a stiff substrate only the upper part of the curve is physically acceptable (for $\delta>0$). The lower part of this curve ($\delta<0$) in the dashed areas corresponds to the case where a clamped free-standing film is allowed to buckle in the downward direction ($z<0$). In this subsection, Δp has been taken to be of the order of the atmospheric pressure (10^{-4} GPa), which corresponds for instance to the relevant experimental situation where one has residual gas below the buckle. It can be observed that the buckling phenomenon occurs for any positive value of the applied stress σ_0 since no critical stress (σ_c^*) can be defined and no bifurcation is evidenced.

5.2 Case of a Negative Pressure Mismatch. In the case of a negative pressure mismatch (Δp=-10^{-4} GPa), which corresponds to the relevant experimental situation where one has vacuum below the buckle, the physically acceptable solution for the buckled film on the substrate is given in Fig. 3(b), also for a nickel thin film. The dashed areas in this figure still correspond to nonphysical situations. Contrary to Sec. 5.1, the curve representing the negative pressure mismatch is now below the curve obtained for Δp=0. In the case where Δp=0, the buckling structure reduces to the classical Euler column for which the maximum deflection is given by [1]

$$\delta = h\sqrt{\frac{4}{3}\left(\frac{\sigma_0}{\sigma_c} - 1\right)} \tag{24}$$

with σ_c=$D\pi^2/hb^2$. The set of curves displayed in Fig. 3(b) is characteristic of a supercritical bifurcation whose normal form $y(y^2-(x-x_c))=0$ when perturbed by a constant term λ is given by [28]

$$y(y^2 - (x - x_c)) - \lambda = 0 \tag{25}$$

with x_c the critical point for the y=$f(x)$ curve. Indeed, this above canonical form can be directly derived expending σ_0 and δ (see Eqs. (20) and (21)) as a function of β near the bifurcation point β_{bc}=π. It yields

$$\delta\left(\delta^2 - \frac{4h^2}{3}\left(\frac{\sigma_0}{\sigma_c} - 1\right)\right) - k\Delta p = 0 \tag{26}$$

with k=$(16h^2b^4)/(3\pi^4 D)$. The form of Eq. (26) allows for determining the pressure effect on the shape of the buckled film near the new critical stress σ_c^* for buckling. Writing

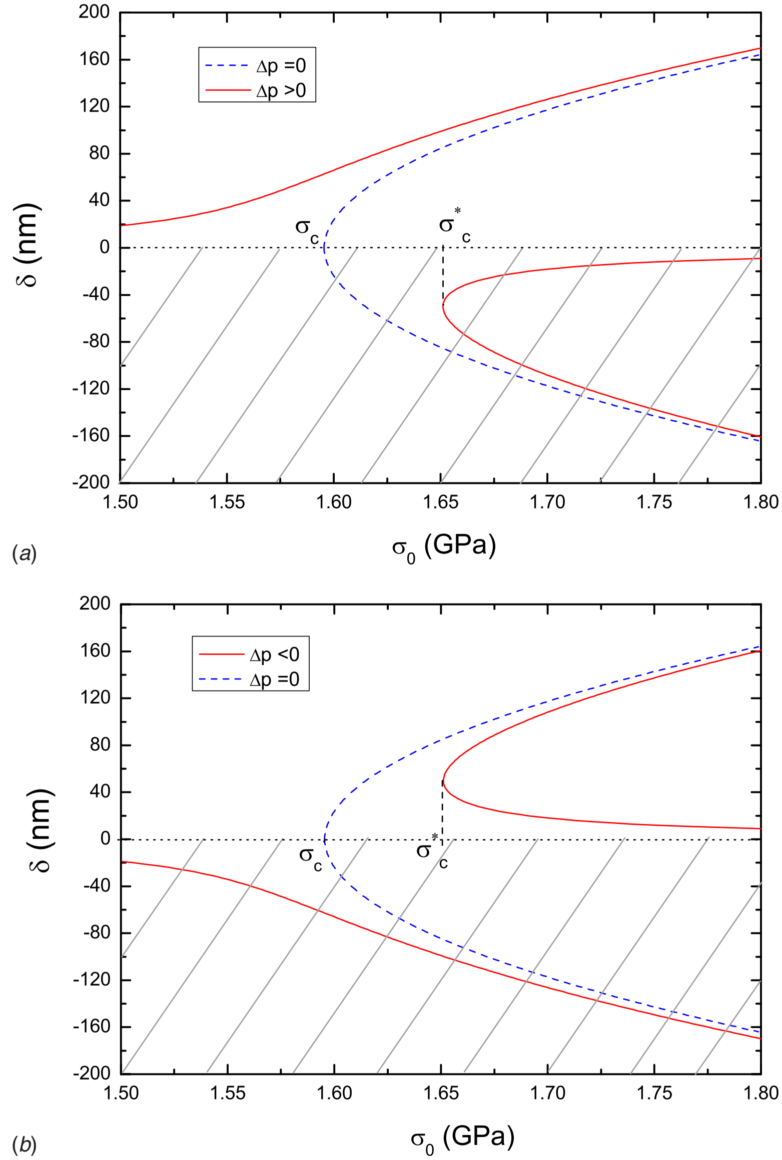

Fig. 3 Case of a nickel thin film under compression. E=200 GPa, ν=0.312, b=4275 nm, and h=400 nm. (*a*) δ as a function of σ_0 for a positive pressure mismatch $\Delta p=10^{-4}$ GPa and for $\Delta p=0$. The zero pressure curve corresponds to the classical Euler column. (*b*) δ versus σ_0 for a negative pressure mismatch $\Delta p=-10^{-4}$ GPa and for $\Delta p=0$.

$$\left.\frac{\partial \sigma_0}{\partial \delta}\right|_{\delta=\delta^*}=0 \tag{27}$$

the critical deflection δ^* at the onset for buckling is given by

$$\delta^*=\frac{1}{2^{1/3}}|\Delta p|^{1/3}k^{1/3} \tag{28}$$

Introducing then this expression of δ^* in Eq. (26), σ_c^* can be defined as follows:

$$\sigma_c^*=\sigma_c\left[1+\frac{3}{4}\left(\frac{1}{2^{2/3}}+2^{1/3}\right)\frac{k^{2/3}|\Delta p|^{2/3}}{h^2}\right] \tag{29}$$

The effect of the pressure mismatch on the critical stress σ_c^* and the deflection δ^* have been investigated. In Fig. 4, σ_c^*/σ_c has been plotted versus $|\Delta p|$. It can be observed in this figure that the critical stress σ_c^* increases with the absolute value of the pressure mismatch and is always greater than the value obtained in the case of the classical Euler column for a thin film under compression. The effect of a negative pressure mismatch is thus to enhance the mechanical stability of the film. Finally, using Eqs. (24), (28), and (29), the ratio δ^*/δ characterizing the jump at the onset for buckling is found to be

$$\left[\frac{\delta^*}{\delta}\right]_{\sigma_0=\sigma_c^*}=\frac{1}{\sqrt{3}} \tag{30}$$

It is emphasized that this ratio does not depend on various parameters of the problem (Δp, σ_c, σ_c^*, $h,\ldots$). Experiments are in progress to confirm this result.

6 Conclusion

In this paper, a set of new buckling solutions for a thin film deposited on a substrate and delaminated on a constant length has been provided in the framework of FvK theory of thin plates and perturbed bifurcation theory when a pressure mismatch is applied

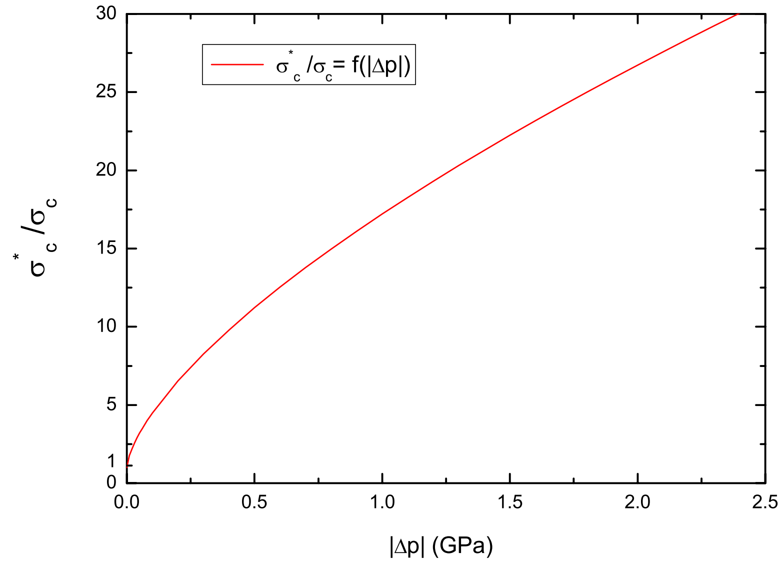

Fig. 4 Variation in the new critical stress σ_c^* with respect to the Euler critical stress σ_c versus the absolute value of the pressure mismatch $|\Delta p|$. $E=200$ GPa, $\nu=0.312$, $b=4275$ nm, and $h=400$ nm.

on its both free surfaces. In particular, the effect of pressure on the critical stress above which the film buckles and on the maximum amplitude of deflection has been characterized. It is believed that these pressure dependent solutions for the thin film profile may be useful to investigate a number of problems of technological interest among which are the buckling of a free-standing thin film during bulge test. The results obtained in this work may be also applied to the problem of the decohesion of coatings when gas cavities appear in the bulk under irradiation, which is encountered during the peeling in fusion reactor or smart-cut process in semiconductors.

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