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From period-doubling to folding in stiff film/soft substrate system: The role of substrate nonlinearity

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ABSTRACT

Uniaxial compressed stiff films on soft substrates can evolve into the period-doubling and folding instabilities, beyond the onset of sinusoidal wrinkling. The substrate is modeled as a neo-Hookean solid with a pre-stretch prior to film attachment, and its nonlinearity is obtained. Both the pre-stretch and the external nominal strain imposed on the film/substrate system can induce different substrate nonlinearity, and thus have different effects on the post-buckling mode evolution of the system. This study shows that the critical strain of period-doubling instability is linear to the pre-stretch. As the compressive nominal strain increases, the folding mode occurs beyond the onset of period-doubling in both the pre-tension and the pre-compression case, due to the softening/hardening effects for the inward/outward displacements generated by the positive substrate nonlinearity.

1. Introduction

Thin stiff films bound to thick compliant substrates can lose stability when the in-plane compressive stress exceeds a critical value, giving rise to micro and nano-scale wrinkles [1]. Such instability phenomenon is apparent in nature, such as human skins [2], tubular organs of animals [3] and hard exocarp/soft sarcocarp plants [4,5]. The buckling of the stiff film/compliant substrate system has been treated as a desirable means of generating various self-organized patterns with intrinsic wavelengths [6–8]. It finds a broad range of potential applications in, for instance, the thin-film metrology [9,10], tunable optical gratings [11,12], and stretchable electronics which can undergo a considerable deformation with the internal stress being less than the material strength [13–16].

The post-buckling behavior of film/substrate systems has received increasing attention due to the variety of instability modes. The primary instability modes include wrinkling [17], creasing [18,19], and buckle delamination [20,21], while the secondary bifurcation modes consist of period-doubling [22], period-tripling [23], localized ridges [24,25] and folds [26,27]. If a film is well-bonded to a substrate, and the film modulus is much higher than that of the substrate, the buckle delamination and creasing instability can be avoided [25,28]. Therefore, the initial flat state of the thin film will lose stability and switch into sinusoidal wrinkles when the compressive stress reaches the onset of wrinkling. The compression is commonly generated by the release of substrate pre-stretch imposed prior to film attachment [7,8,14]. Further compression of the system can induce period-doubling [22], period-tripling [23], folds [27] and localized ridges [25]. Some previous studies revealed that the substrate pre-stretch and the film/substrate modulus ratio determine the occurrence and evolution of the post-buckling modes [23–25,28]. Localized ridges appear when the pre-stretch is sufficiently large and folds form in systems that have low film/substrate modulus ratio [25,28]. The period-doubling configuration forms in a wide range of pre-stretch and modulus ratios [25,28]. The period-tripling morphology is identified in the low modulus ratio case, and can occur instead of period-doubling at a later secondary bifurcation point [23]. However, the quantitative connections between the onset of these post-buckling modes and the system parameters have not been established. In this study, we focus on the period-doubling instability, and an approximate relationship governing the onset of this instability mode is presented.

Some theoretical and numerical investigations have focused on the role of substrate nonlinearity in the occurrence of the various post-buckling modes [22,24,29–31]. An elastic strut supported by an elastic foundation experiences mode jumping due to the stiffening effect of the cubic nonlinearity [31,32]. Zhuo and Zhang [33] revealed that the substrate quadratic nonlinearity in the stiff film/soft substrate system causes mode coupling and then induces the period-doubling, which is regarded as a pitchfork bifurcation [34]. The experiment carried by Brau et al. [22] also showed that the period-doubling instability is triggered by the substrate quadratic nonlinearity, which induces asymmetric traction–displacement

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relation: the normal tension/compression on the substrate surface is no longer equivalent for the same given outward/inward displacement. In addition, the pre-tension and pre-compression imposed on the substrate prior to film attachment have different effect on the substrate nonlinearity, giving rise to the formation of localized ridges and folds, respectively [24,25]. Hutchinson [30] established the relation between the pre-stretch and the nonlinearity of a neo-Hookean substrate and identified that the pre-stretch is one of the parameters that control the instability of the film/substrate bilayer. In this study, the expression for the nonlinear effect is further given as a function of the pre-stretch and the overall nominal strain. Moreover, the mode transition from period-doubling to folding is possible as the compression increases considerably.

This study explores the role of substrate nonlinearity in the morphology evolution of a compressed stiff film on a compliant substrate. Based on the perturbation method, the nonlinear response of a neo-Hookean half-space under periodic displacement constraints is presented and the substrate nonlinearity is obtained. Finite element simulations of neo-Hookean film/substrate systems are also carried out, which show the morphology transitions from sinusoidal wrinkling to period-doubling and then the folding mode. The model also predicts the critical strain of period-doubling instability.

2. Nonlinear traction–displacement behavior of a neo-Hookean substrate

In this section, a perturbation analysis is presented to analyze the nonlinear response of a semi-infinite neo-Hookean substrate which undergoes plane-strain deformation. Fig. 1 illustrates three different states during the wrinkling of a stiff thin film/soft substrate bilayer. An elastomeric (e.g., poly(dimethylsiloxane)

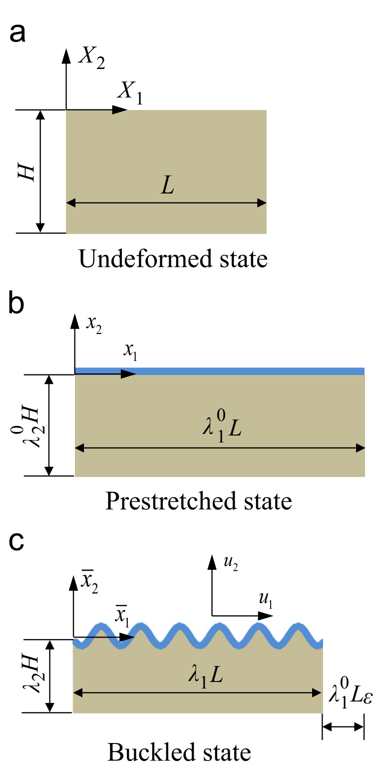

Fig. 1. Schematics of the wrinkling process of the film/substrate bilayer and the coordinate systems. (a) The initially undeformed substrate of length L and of height H. (b) The substrate undergoes a pre-stretch λ_1^0 before attaching the thin film. (c) The film is buckled and the substrate stretch is λ_1 compared to the undeformed state. The nominal compressive strain applied on the bilayer is $\varepsilon=(\lambda_1^0-\lambda_1)/\lambda_1^0$.

(PDMS)) substrate of length L and height H in the undeformed state (Fig. 1(a)) is uniformly pre-stretched prior to the attachment of a stiff thin film (Fig. 1(b)). Lagrangian coordinates, X_i and x_i $(i=1,2)$, specify the material points in the undeformed state and pre-stretched state, respectively. The pre-stretches in three principal axes X_1, X_2 and X_3 are denoted by λ_1^0, λ_2^0 and λ_3^0, respectively. To characterize the plane-strain deformation, the pre-stretch in X_3 direction is set zero, i.e., $\lambda_3^0=1$. The material is assumed to be incompressible, i.e., $\lambda_1^0\lambda_2^0=1$. The coordinates of the material points with respect to the pre-stretched state are related to that of the undeformed state by the following

$$x_i=\lambda_i^0 X_i \quad (i=1,2). \tag{1}$$

In the buckled state (Fig. 1(c)), the coordinates are denoted by

$$\begin{cases}\bar{x}_1=\lambda_1 X_1+u_1\\ \bar{x}_2=\lambda_2 X_2+u_2\end{cases}, \tag{2}$$

where u_1 and u_2 are the displacements parallel to the coordinate axes X_1 and X_2, respectively. λ_i $(i=1,\ 2)$ is the stretch of the substrate, which is defined as the length in the current state (the buckled state) divided by the length in the reference state (the undeformed state). It is related to the pre-stretch by

$$\begin{cases}\lambda_1=\lambda_1^0(1-\varepsilon)\\ \lambda_2=\lambda_2^0/(1-\varepsilon)\end{cases}, \tag{3}$$

where $\varepsilon=\left(\lambda_1^0-\lambda_1\right)/\lambda_1^0$ is the nominal strain (positive for compression), as shown in Fig. 1(c). The strain ε should be compressive to induce the buckling of the film/substrate system, which means that the stretch $\lambda_1<\lambda_1^0$ from Eq. (3).

The substrate is assumed to be an incompressible neo-Hookean material and hence its strain energy densities in the pre-stretched (W_{I}) state and the buckled state (W_{II}) are given as follows [35]

$$W_{\mathrm{I}}=\frac{\mu_s}{2}\left[\left(\lambda_1^0\right)^2+\left(\lambda_2^0\right)^2-2\right], \tag{4}$$

and

$$W_{\mathrm{II}}=\frac{\mu_s}{2}\left[(\lambda_1+u_{1,1})^2+u_{2,1}^2+(\lambda_2+u_{2,2})^2+u_{1,2}^2-2\right]. \tag{5}$$

Therefore, the increment of the strain energy density in passing from the pre-stretched state to the buckled state is thus given as the following

$$\Delta W=W_{\mathrm{II}}-W_{\mathrm{I}}=\mu_s\left[\frac{1}{2}\left(u_{1,1}^2+u_{1,2}^2+u_{2,1}^2+u_{2,2}^2\right)+\lambda_1 u_{1,1}+\lambda_2 u_{2,2}+f_\varepsilon\right],, \tag{6}$$

where $f_\varepsilon=\left(\lambda_1^0\right)^2\left[(1-\varepsilon)^2-1\right]+\left(\lambda_2^0\right)^2\left[1/(1-\varepsilon)^2-1\right]$; $\mu_s=E_s/[2(1+\nu_s)]$ is the shear modulus of the substrate; E_s and ν_s are the Young's modulus and Poisson's ratio, respectively.

The incompressibility condition of the substrate in the buckled state is written as [35]

$$J-\lambda_1\lambda_2=\lambda_2 u_{1,1}+\lambda_1 u_{2,2}+u_{1,1}u_{2,2}-u_{1,2}u_{2,1}=0, \tag{7}$$

where J is the determinant of the deformation gradient, namely, the volume change from the reference state to the current state.

Let $Q=Q(X_1,X_2)$ be the function to be determined which has the same period as the surface displacements [19,30]. Here a Lagrangian multiplier q is introduced to enforce the incompressibility condition in Eq. (7) [30]. Physically, the Lagrangian multiplier q indicates the hydrostatic pressure, namely, $q=q_0+\Delta q=\mu_s(r+Q)$, where $r=\lambda_2/\lambda_1=1/\lambda_1^2$. $q_0=\mu_s r$ is the pressure in the pre-stretched state and $\Delta q=\mu_s Q$ is the increment of the pressure from the pre-stretched state to the buckled state [26]. The change of the total potential energy in passing from the pre-stretched