The primary bilayer ruga-phase diagram I: Localizations in ruga evolution

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ABSTRACT

It has been observed that coexistence of multiple ruga (wrinkle, crease, fold or ridge) phases hinders advancement of nano and soft materials technology to control and manufacture uniform ruga phases in bilayer material systems. In this paper, we construct the primary bilayer (PB) ruga-phase diagram which can guide manipulation of various ruga configurations in bilayer systems. The PB ruga-phase diagram is a generic phase diagram of a bilayer system composed of a thin film on a half-space substrate, both of which are represented as incompressible neo-Hookean solids. On the PB ruga-phase diagram, various phase boundaries represent bifurcation sites of ruga structures caused by lateral compression of the bilayer. We have identified eleven different ruga phases and five triple points of ruga phases on the PB ruga-phase diagram. All the ruga phases eventually evolve to a limit phase of either global crease or global fold localization, depending on the stiffness ratio of the bilayer, when compressed up to the Biot critical strain of 0.456. Another global localization – ridge localization which is principally caused by large substrate pre-stretch (or mismatch strain) – is treated in a sequel paper.

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1. Introduction

Over the past several decades, a series of analyses on solid-surface deformation under in-plane compression have revealed diverse characteristics of one dimensional (1D) ruga structures — various 1D corrugated patterns of solid surfaces, and their transitions [1–6]. In particular, a bilayer system generates relatively simple generic ruga phases with a single characteristic ruga wavenumber. The ruga phases include single- as well as multi-mode wrinkles, creases, folds and ridges. Morphological characteristics of the ruga phases are often found useful for various technological applications [7–11]. Moreover, study of ruga phase transitions helps understand formation processes of various biological and geological ruga structures [12,13]. In addition, fine control of self-organizing ruga phases will enable us to create advanced materials of unprecedented properties by folding 2D material structures (e.g. [14]). To this end, here, we investigate formation conditions of various 1D ruga phases, and criteria on their stabilities, localization, and co-existence of multiple phases by constructing a ruga-phase diagram of a soft-substrate bilayer system.

A bilayer system of an elastic thin film on a soft elastic substrate develops a variety of ruga phases when compressed laterally and/or experiences growth of mismatch strain between the film and the substrate. Collection of the ruga phases on a plane of the characteristic ruga wavenumber (or elastic stiffness ratio) versus the compressive strain of the film represents a ruga-phase diagram [15–17]. Similar to the phenomenological description of a conventional material phase diagram in terms of its minimum free
energy for various equilibrium phases [18], collection of monomorphogenic ruga configurations in equilibrium under well-defined constraints can constitute a ruga-phase diagram. While the material phase transitions can be rightly described by the Ginzburg–Landau theory [19], the ruga-phase transition is typically analyzed by bifurcation theories such as the Koiter theory [20,21].

In this paper, we only consider the most basic bilayer — an infinite-span film on a half-space substrate, both incompressible neo-Hookean, among diverse combinations of film/substrate geometries and elastic properties, as a primary bilayer (PB). Furthermore, we limit our ruga analyses to PB’s without mismatch strain (or substrate pre-stretch), for various stiffness ratios, by tracing ruga evolution caused by plane-strain compression up to complete ruga localization at the Biot strain of 0.456. When a PB is gradually compressed, the film first buckles to generate a periodic wrinkle or crease, depending on the relative film stiffness to the substrate; a stiff film wrinkles and a soft creases. Hutchinson [22] analyzed the role of nonlinear substrate elasticity on 1D PB wrinkling under general biaxial stretch of the substrate; he also got the critical stiffness ratio and strain for wrinkle instability, employing the Koiter method [20,21]. The PB-buckling period in undeformed reference configuration sets the characteristic ruga wavenumber.

In general, uniaxial ruga wavenumber of a deep-substrate bilayer depends on the stiffness ratio and the film span, when normalized by the film thickness. However, the normalized characteristic wavenumber of PB is governed only by the stiffness ratio. In particular, the dependence of the normalized characteristic wavenumber on the stiffness ratio is reduced to a simple formula [5,22,23],

\[
\tilde{k} = 2\pi h/l = (3\mu_s/\mu_f)^{1/3},
\]

for \( \mu_f/\mu_s \ll 1 \), where \( l \) is the wavelength, and \( \mu_s \) and \( \mu_f \) the shear stiffness of the substrate and the film respectively. Here, Eq. (1) represents not only the stiff-film limit wavenumber of a PB but also a convenient stiffness ratio index of the PB. Then, all possible ruga phases of PB’s can be displayed on a compact bound plane of \((\varepsilon, \tilde{k})\), where \( \varepsilon \) represents the compressive strain applied on the PB; the display is the PB ruga-phased diagram.

While a ruga-phase diagram can be constructed for absolute minimum energy configurations (e.g. [17]), here, we build the PB ruga-phase diagram by following ruga evolution under monotonic compression, employing extensive finite element analyses. Then, we can readily see ruga localizations, irreversible ruga transitions and substrate pre-stretch (or mismatch strain) effects on the PB ruga-phase diagram. We present studies of ruga localizations in this paper, while analyses of irreversible ruga transitions and substrate pre-stretch effects on ruga formations are reported in two sequel papers.

2. Ruga evolution leading to global ruga localization

A stiff film on a soft substrate has intrinsic characteristics of wrinkling at a small compressive strain \([2,3,25]\). Wrinkling typically disperses distribution of the film stress into the substrate, while creasing has tendency to localize surface deformation. Therefore, interaction between the film and the substrate deformation characteristics creates various ruga phases evolving from wrinkle initiation to global crease-like fold localization when compressed up to a large strain. The ruga phases of the PB system include not only single-, double- and quadruple-mode wrinkles, but also multiple-mode film creases, fold and ridge, leading to global crease, fold and ridge localizations.

Fig. 1(a) shows a schematic of a PB under lateral compression, which undergoes various ruga evolution pathways (Fig. 1(b)) to reach global ruga localizations (Fig. 1(c)). A soft film but stiffer than the substrate is typically observed in biological systems, and such a film often creases periodically, which eventually leads to a global crease localization under further compression [12]. In contrast, a very stiff film on a soft substrate is typically used in advanced engineering applications, and the film generally folds before it develops a global localization under large compression [5]. Compression with very large substrate pre-stretch causes ridge localization [6]. Now, we have the following questions. How stiff must a soft film be, relative to the substrate, not to crease upon lateral compression? What are the critical compressive strains for mode doubling and quadrupling of the characteristic wrinkle of a bilayer system? Can we have mode transitions of film-crease in a PB system, similar to the wrinkle folding process? What are the critical compressive strains for the onsets of global ruga localizations? How are the critical values influenced by a substrate pre-stretch (or mismatch) strain? These questions are answered by constructing the primary ruga-phase diagram in this and sequel papers.

3. Computational analysis of ruga localization

Finite element method (FEM) is used to implement the simulations of ruga phase evolution and associated plane-strain deformation in the PB system under lateral compression, employing the standard FEM package ABAQUS. The incompressible neo-Hookean constitutive model is adopted for both the film and substrate with different shear moduli. The length of the simulation sample is set to be \( L \) in the \( X_1 \) direction and \( H \) in the \( X_2 \) direction. The initial length of the sample is set to be at least four times of the critical wavelength of wrinkling in the film–substrate system to ensure that relevant phenomena (e.g., period quadrupling) can be observed in the simulation. To mimic a semi-infinite substrate, the aspect ratio (depth to length) of the sample is set to be 20 (i.e., \( H = 20L \)).

Prior to compression, a small sinusoidal perturbation in displacement in the \( X_2 \) direction is applied to the film surface to probe any instability in the system. The expression for this perturbation is written as

\[
u_2 = \zeta \cos(8\pi X_1/L), \quad \text{for} \quad -\frac{L}{2} \leq X_1 \leq \frac{L}{2},
\]

where \( L \) and \( l \) denote the film span and the characteristic wavelength of the PB system, respectively; the perturbation amplitude \( \zeta \) should be small enough to ensure the
accuracy of simulation, and is taken to be $\zeta/l = 0.0005$. The characteristic wavelength, $l$, for general PB is given by [16,22].

In the FEM model, hybrid elements CPE4RH are used to simulate the incompressible neo-Hookean material, and periodic boundary condition is applied at both lateral faces. The span of the film, $L$, is varied for $L = 4nl$, $n = 1, 2, \ldots, 5$ to test model-size sensitivity of various critical strains in ruga evolution. A self-contact interaction with the type of frictionless and “hard contact” is applied to the film surface to avoid penetration during creasing and folding deformations. The onset strains of ruga-phase transitions are identified by Fast Fourier Transform (FFT) with the criticality-identification method introduced in [15]. The stiffness-ratio index, $\bar{k}$, expressed in (1) ranges from 1.44 to zero, corresponding to the limiting case of a homogeneous solid ($\mu_s = \mu_f$) and an extremely stiff substrate ($\bar{k} = 0$). The stiffest film simulated is 10000 times stiffer than the substrate.

4. Results and discussions: the PB ruga-phase diagram

The extensive FEM simulations reveal that the PB system has four distinct ruga evolution pathways to global crease localization, one pathway to global fold localization, and another to global ridge localization. The ridge localization requires pre-stretch of the substrate, and the post ridge localization behavior is quite different from those of crease or fold localization. The critical strains of ruga-phase transitions identified by FEM are insensitive to the modeling span of the film for $L = 4nl$, $n = 1, 2, \ldots, 5$.

4.1. Global crease localization

Fig. 2(a) shows schematics of the four different ruga evolution pathways heading towards the limit phase of global crease localization (III). The first pathway is instantaneous film crease which directly leads to the global crease localization (III) from the flat phase (I), for a very low film-stiffness ratio (or high stiffness-ratio index; $1.2 \leq \bar{k} \leq 1.44$) of the bilayer. The second pathway is setback crease of the film, (I) $\rightarrow$ wrinkle (II) $\rightarrow$ wrinkle–crease (IIC), which leads to the global crease localization (IIC $\rightarrow$ III), for $0.8 \leq \bar{k} \leq 1.2$. The setback film crease has the wavenumber of $k_w/4$ where $k_w$ is the wrinkle wave number; in other words, the period of the film creasing is quadruple of the primary wrinkle period. The quadruple-period creasing is believed due to the reason that it is necessary to amplify local strains at wrinkle valleys to the level of the Biot critical strain of 0.456 and to have sufficient energy release for unstable crease growth at every fourth valleys. The third pathway is mode doubling of the wrinkle (IV) followed by setback crease (IVc) which leads to the global crease localization (III), for $0.6 \leq \bar{k} \leq 0.8$. Again, the period of the film creasing is quadruple of the primary wrinkle period. The fourth pathway is the setback crease (Vc) following wrinkle period doubling and quadrupling (V), which develops folds with crease tips (Vcf) before they proceed to the global crease localization (III), for $0.4 \leq \bar{k} \leq 0.6$. The fold period is also quadruple of the primary wrinkle period. While the initial quadruple crease and fold localizations are the most probable localization mode for a very large film span, it is also possible to have non-quadruple localizations for small film spans of $L = 4nl$ with non-integer $n$'s.

Figs. 2(b)–(e) show FEM generated ruga configurations of typical 1st ($\bar{k} = 1.44$; homogeneous half space), 2nd ($\bar{k} = 1$), 3rd ($\bar{k} = 0.87$) and 4th ($\bar{k} = 0.41$) pathways leading to a global crease localization, together with associated maximum main nominal strain, $\varepsilon_{p_{\text{max}}}$, distributions in the cross-section of the bilayer. Fig. 2(b) shows that the global crease localization (III) is made by relaxing periodic strain concentrations of initial perturbation (I), in the first evolution pathway. Similarly, Fig. 2(c) displays that the film wrinkles first (II), and every four other valleys of the wrinkle make setback creases in expense of relaxing three other wrinkle valleys (IIc). Then, the global crease localization (III) is achieved by releasing the periodic setback film-creases, in the second evolution pathway. Fig. 2(d) exhibits that the film forms a single-mode wrinkle (II), followed by transforming to a double-mode wrinkle (IV), and then creases in every fourth valleys of the primary wrinkle period (IVc) before it progresses to the globally localized crease (III), in the third pathway. Fig. 2(e) presents that the film forms a single-mode wrinkle (II), transforms to a double-mode wrinkle (IV), then to a quadruple-mode wrinkle (V) where all the deep valleys crease (Vc) before the creasing valleys fold (Vcf), in the fourth pathway. The crease fold eventually reaches the limit phase of global crease localization upon further compression.
Fig. 2. Pathways to global crease localization of soft-film bilayers: (a) schematics of four different pathways to global crease localization; FEM simulation plots of the maximum principal nominal strain, \( \varepsilon_{\text{max}} \), for (b) instantaneous crease \( \bar{k} = 1.44 \); (c) wrinkle–film crease–global crease localization \( \bar{k} = 1 \); (d) wrinkle–double-mode wrinkle–quadruple-mode crease–global crease localization \( \bar{k} = 0.87 \); (e) wrinkle–double-mode wrinkle–quadruple-mode wrinkle–quadruple-mode tip crease–fold–global crease localization \( \bar{k} = 0.41 \). Frames in the dotted box of (c) and (d) show final localization processes in a different scale. The color bar of \( \varepsilon_{\text{max}} \) ranges (b) 0–1.136, (c) 0–0.943, (d) 0–1.058 and (e) 0–0.770. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4.2. Fold and ridge localizations

The first row of Fig. 3(a) shows schematics of the ruga evolution pathway of global fold localization through wrinkling (II), mode doubling (IV), mode quadrupling (V), and folding (VI), to get to the limit phase of global fold localization (VII), as compressive strain increases for \( 0.0 < \bar{k} \leq 0.4 \). When the film is approximately 47 times or more stiffer than the substrate \( (0.0 < \bar{k} \leq 0.4) \), the film of the PB without substrate pre-stretch does not crease; instead, it folds in every four other wrinkle valleys after mode quadrupling of the wrinkle. The subsequent two rows of Fig. 3(a) show four FEM-generated ruga configurations in the process of global fold localization from the periodic folds (IV) for a stiff film \( \bar{k} = 0.17 \). The four frames of the localization process show that the localizing fold tip penetrates deep into the soft substrate in expense of opening other nearby folds.

If the substrate is pre-stretched beyond the stretch ratio of approximately 1.5, the film develops a global ridge localization instead of a global crease or fold localization, as shown in Fig. 3(b). The first row of Fig. 3(b) shows schematics of the ridge localization; the film wrinkles first, and then a ridge localizes in expense of unloading the wrinkles away from the localized ridge, as shown in the subsequent rows. However, unlike a global crease or a fold localization, growth of the ridge height has a limit upon further compression. Beyond the ridge-localization limit strain, multiple ridges begin to form and the ridge-period (i.e. inter-distance between nearby ridges) shortens when the lateral compression increases until it reaches the critical strain for the minimum period. Then, the period lengthens when it is further compressed until it begins to make ridge folds. The ridge localization is an intermediate process towards fold localization in a PB system with very large substrate pre-stretch. Details of the ridge behavior are presented in a sequel paper, part III.

4.3. The PB ruga-phase diagram

All the critical points of ruga evolution in the four pathways leading to the global crease localization and in the pathway leading to the global fold localization generate the PB ruga-phase diagram without pre-stretch, on the \((\varepsilon_c, \bar{k})\) plane, as shown in Fig. 4.

The black dots on the ruga-phase diagram represent critical points of wrinkle bifurcation identified by FEM analyses. The ruga-phase boundary \( T_{12} \) delineates the wrinkle phase (II) from the flat phase (I). The wrinkle-bifurcation boundary \( T_{12} \) indeed approaches the theoretical prediction \( \bar{k} = 2\sqrt{\varepsilon} \) for \( \varepsilon \ll 1 \), i.e. for a very stiff film. The solid curve for \( T_{12} \) & \( T_{13} \) in Fig. 4 is a plot of

\[ \bar{k} = 2 \left(1 + 0.15\varepsilon \right) \sqrt{\varepsilon}. \] 

(3)

The FEM results are closely represented by Eq. (3) for the entire range of \( 0 \leq \varepsilon \leq 0.456 \) up to the Biot critical strain. Here, the stiffness-ratio index, \( \bar{k} \), is not close to the actual wrinkle wavenumber \( k_w \), not exceeding 0.6, for compliant films \( (0.3 < \bar{k}) \). The formula for the actual wavenumber
Fig. 3. Pathways to global localization of stiff-film bilayers: (a) global fold localization with pathways of wrinkle–double-mode wrinkle–quadruple-mode wrinkle–fold–global fold localization (the FEM frame display is for $k = 0.17$); (b) global ridge localization caused by pre-stretch of the substrate (the FEM display is for a pre-stretch of 2.0). The color bar of $\varepsilon_{p}^{\max}$ ranges (a) 0–0.943 and (b) 0–1.925. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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\hat{k}_w$ of the PB can be found in Refs. [16,22]. Here, ‘B’ at $\varepsilon = 0.456$ on the line of homogeneous half space ($\hat{k} = 1.44$) denotes the Biot critical point [1], while ‘A’ signifies the subcritical limit of the homogeneous-half-space creasing at $\varepsilon = 0.35$ [3,25].

Along the wrinkling boundary $T_{12}$, we encounter a triple point $M^+$ at $\hat{k} = 1.2$ and $\varepsilon = 0.32$; above this point, i.e. $\hat{k} > 1.2$, the wrinkle instability turns into higher-order cascade instability, and the wrinkle instability line $T_{12}$ becomes an instantaneous crease line $T_{13}$ [16]. At the triple point $M^+$, the wrinkle instability line $T_{12}$, the instantaneous crease line $T_{13}$ and the setback crease line $T_{2c}$ merge together. Along the setback crease line $T_{2c}$, every four other wrinkle valleys crease in expense of relaxing the wrinkle, as discussed in Section 4.1 with Fig. 2(c). The setback crease line is extended from $M^+$ to another triple point $M^-$ along the trace of green square markers in Fig. 4. Along the setback crease trace, we also meet two other triple points $D^+$ and $Q^+$. At the triple point $D^+$ at $\hat{k} = 1.0$ and $\varepsilon = 0.243$, the setback crease line $T_{2c}$, the double-mode wrinkle instability line $T_{24}$ and another setback crease line $T_{4c}$
Fig. 4. The PB ruga-phase diagram: $\bar{k}$ and $\epsilon$ denote the stiffness-ratio index, and the compressive strain respectively; the solid curve $T_{13}$ & $T_{13}$ represents Eq. (3); A ($\epsilon_A = 0.35, \bar{k}_A = 1.44$) and B ($\epsilon_B = 0.456, \bar{k}_B = 1.44$) signify the lower bound of the subcritical state of creasing, and the Biot instability point; (I) flat phase; (II) single-mode wrinkle phase; (III) limit phase of global crease localization; (IV) double-mode wrinkle phase; (V) quadruple-mode wrinkle phase; (VI) fold phase; (VII) limit phase of global fold localization; (Ic), (Ivc) and (Vc) are three setback-crease phases; (Vcf) crease-fold phase; $M^+(\epsilon = 0.32, \bar{k} = 1.2), M^-(\epsilon = 0.27, \bar{k} = 0.4), D^+(\epsilon = 0.0.243, \bar{k} = 1.0), Q^+(\epsilon = 0.22, \bar{k} = 0.6),$ and $F^+(\epsilon = 0.24, \bar{k} = 0.6)$ are five ruga-phase triple points. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

merge together. The line of $\bar{k} = 1.0$ for $0.243 \leq \epsilon \leq 0.35$ sets the ruga-phase boundary $P_{24}$ which delineates the ruga-phase (Ivc) from (IVc). Along $T_{24}$ depicted by the trace of blue diamond markers in Fig. 4, the single-mode wrinkle (II) becomes unstable and transforms to a double-mode wrinkle (IV), as illustrated by the first two frames in each of (Ivc) and (Vc). Across the setback crease line $T_{4c}$, deep valleys of every fourth primary wrinkle period in the double-mode wrinkle (IV) cease to generate the phase (IVc) as illustrated in Fig. 2(d). At the triple point $Q^+$ at $\bar{k} = 0.6$ and $\epsilon = 0.22$, the quadruple-mode wrinkle instability line $T_{45}$ merges to the setback crease lines $T_{4c}$ and $T_{5c}$. The line of $\bar{k} = 0.6$ for $0.22 \leq \epsilon \leq 0.35$ sets the ruga-phase boundary $P_{45}$ which delineates the ruga-phase (IVc) from (Vc) and (Vcf). Along $T_{45}$ indicated by the trace of brown triangle markers, the double-mode wrinkle (IV) becomes unstable and transform to a quadruple-mode wrinkle (V). On the setback crease line $T_{5c}$, the deepest valleys of every fourth primary wrinkle period in the quadruple-mode wrinkle (V) cease to produce the phase (Vc) as illustrated in Fig. 2(e).

At the triple point $\bar{k} = 0.4$ and $\epsilon = 0.27$, the setback crease line joins the fold line composed of $T_{5f}$ and $T_{56}$, and extended from $F^-$ to $F^+$. The line of $\bar{k} = 0.4$ for $0.27 \leq \epsilon \leq 0.456$ sets the ruga-phase boundary $P_{56}$ which delineates the ruga-phase (Vc) from (VI). The fold line traces the purple diamond markers in Fig. 4. Across $T_{5f}$ the valleys of (Vc) fold with their tips creased to produce the phase (Vcf) as illustrated in Fig. 2(e). It is noted that the PB without mismatch strain cannot develop more than quadruple mode wrinkles, since the deep valleys of the quadruple mode wrinkle begin to fold before higher modes could be activated. As a consequence of folding after mode quadrupling, the PB without mismatch strain can only fold the wrinkles in every four other valleys. In our computer simulations it is noticed that if the bilayer is compressed with a large positive pre-stretch, it can fold every other valleys, but not every valley. In addition, it is found that stiff films of $k \leq 0.2$ have critical strains of double-mode and quadruple-mode wrinkle instabilities at $\epsilon_f = 0.18$ and $\epsilon_Q = 0.26$, independent of the stiffness ratio of the bilayer system. The folding strain $\epsilon_f = 0.29$ is also insensitive to variations of the stiffness ratio for such stiff-film bilayers. The last triple point that we see is $F^+$ at which three phases, (IVc), (Vc) and (Vcf) coexist.

When the wrinkles are compressed to go through setback creasing for $0.6 \leq \bar{k} \leq 1.2$ or folding for $0 \leq \bar{k} \leq 0.6$, the ruga begins to globally localize. The localization starts at a point on the trace of the red square markers in Fig. 4. The onset of global localization occurs consistently at $\epsilon = 0.35$ for $0.4 < \bar{k} \leq 1.2$, exhibiting imperfection insensitivity. In contrast, the compressive strain to trigger the global localization drops down to approximately 0.3 from 0.35 for $0.2 < \bar{k} \leq 0.4$, and the critical strain to start the global localization wildly fluctuate between 0.3 and 0.4 for $\bar{k} \leq 0.2$. The initiation points of global ruga localization likely indicate that the film crease triggers the substrate-creasing mode of the global localization at $\epsilon = 0.35$ [3] for $0.4 < \bar{k} \leq 1.2$, and that the folded wrinkle plays a role of imperfection to prompt the substrate-creasing mode in the range of $0.28 < \epsilon < 0.42$ [6] for $\bar{k} \leq 0.4$. Once the global ruga localization starts, advancements of the periodic localization fronts compete each other through the deformation field, similar to growth of parallel cracks. Eventually only one front will advance by unloading all other fronts. In our simulation, the localization nearly completes at the Biot critical strain $\epsilon = 0.456$, as indicated with a vertical dotted line in Fig. 4.

The PB ruga-phase diagram reveals that there exist five triple points, which can stimulate coexistence of multiple ruga phases caused by fluctuations in variables of ruga formation. The variables include the film thickness, the stiffness ratio and the size of the bilayer, and the
boundary conditions of compression. Coexistence of multiple ruga phases limits controllability in making uniform ruga patterns, and is caused by not only fluctuations in ruga-formation variables near triple points but also irreversibility of ruga-phase transitions. Various forms of irreversibility in ruga-phase transitions are discussed in the sequel part II paper. Since the PB ruga-phase diagram in Fig. 4 is for monotonic compression, much portion of the ruga-phase boundaries, other than $T_{02}$, will shift to the left on the diagram for unloading, due to irreversibilities of creating, some period multiplications and global localizations.

5. Conclusion

Extensive nonlinear FEM analyses have revealed various ruga evolution patterns leading to global ruga localization in PB’s subjected to large lateral compressive deformation. The ruga evolution is marked by a series of bifurcations which delineate various ruga phases. The PB wrinkles when compressed to $\varepsilon = \frac{k'}{2 + 0.3k}$; $A$ PB with a soft compliant film ($0.4 \leq k' \leq 1.44$) always encounters instantaneous film-crease for $1.2 \leq k' \leq 1.44$ or quadruple-mode setback film-crease for $0.4 \leq k' \leq 1.2$, which eventually develops into a global crease localization. In contrast, a PB with a stiff film ($k' \leq 0.6$) folds before progressing to a crease ($0.4 \leq k' \leq 0.6$) or fold ($k' \leq 0.4$) localization. All the folding processes of the PB without pre-stretch are preceded by double- and quadruple-mode wrinkling. Collection of the ruga phases on a $(\varepsilon, k)$ plane comprises the PB ruga phase diagram. Five triple points are observed on the PB ruga-phase diagram. Fluctuations in ruga-formation variables can provide coexistence of multiple ruga phases near the triple points. A uniform ruga phase can be effectively manufactured by avoiding the triple points. We expect that the PB ruga-phase diagram will play a significant role in controlling nano ruga structures of 2D atomic-layer materials to explore unprecedented functional properties.

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