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

R. Zhao, T. Zhang [1], M. Diab, H. Gao, K.-S. Kim *

School of Engineering, Brown University, Providence RI, United States

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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.

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

* Correspondence to: Box D, School of Engineering, Brown University, Providence, RI 02912, United States. Tel.: +1 401 863 1456.

E-mail address: kyung-suk_kim@brown.edu (K.-S. Kim).

[1] Current address: Department of Mechanical Engineering, MIT, 77 Massachusetts Avenue, Room 1-025, Cambridge MA, 02139, United States.

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energy for various equilibrium phases [18], collection of monomorphic 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],

$$\bar{k} = 2\pi h/l = \left(3\mu_s/\mu_f\right)^{1/3}, \tag{1}$$

for $\mu_s/\mu_f \ll 1$, where l is the wavelength, and μ_s and μ_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 $\left(\varepsilon, \bar{k}\right)$, where ε represents the compressive strain applied on the PB; the display is the *PB ruga-phase 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 [24]; on the other hand, the soft substrate by itself has intrinsic characteristics of creasing at a large 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

$$u_2 = \zeta \cos(8\pi X_1/l), \quad \text{for } -\frac{L}{2} \le X_1 \le \frac{L}{2}, \tag{2}$$

where L and l denote the film span and the characteristic wavelength of the PB system, respectively; the perturbation amplitude ζ should be small enough to ensure the