Wrinkling instabilities in soft bi-layered systems

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Wrinkling phenomena control the surface morphology of many technical and biological systems. While primary wrinkling has been extensively studied—experimentally, analytically, and computationally—higher order instabilities remain insufficiently understood, especially in systems with stiffness contrasts well below one hundred. Here, we use the model system of an elastomeric bilayer to experimentally characterize primary and secondary wrinkling at moderate stiffness contrasts. We systematically vary the film thickness and substrate prestretch to explore which parameters modulate the emergence of secondary instabilities including period-doubling, period-tripling, and wrinkle-to-fold transitions. Our experiments suggest that period-doubling is the favorable secondary instability mode and that period-tripling can emerge under disturbed boundary conditions. High substrate prestretch can suppress period-doubling and primary wrinkles immediately transform into folds. We combine analytical models with computational simulations to predict the onset of primary wrinkling, the post-buckling behavior, secondary bifurcations, and the wrinkle-to-fold transition. Understanding the mechanisms of pattern selection and identifying the critical control parameters of wrinkling will allow us to fabricate smart surfaces with tunable properties and to control undesired surface patterns like in the asthmatic airway.
1. Motivation

Mechanical instabilities are an important mechanism to control the formation and evolution of surface patterns in nature [27]. Mechanically stimulated changes in morphology can be crucial for proper functioning such as wrinkles in the mammalian brain [8,9,26] or the intestine [3], but also lead to pathological conditions, for instance in the human airway [15]. A similar mechanism underlies the emergence of wrinkles during the dehydration of fruit as illustrated in Figure 1. In technical systems, wrinkling instabilities allow to manufacture microscopic surface structures [31,32] or to measure the Young’s moduli of thin polymeric films and coatings.

![Figure 1. Wrinkling instabilities caused by dehydration in an apple (left), a date (middle), and an apricot (right).](image)

Various parameters control pattern selection in nonuniformly compressed film-substrate-systems. Depending on the stiffness ratio between film and substrate, their mismatch stretch, and adhesion energy, creases or wrinkles emerge, which transform into folds, period doubles, or ridges for higher compression, or delaminate [20,30,34]. Bifurcation analyses can predict the critical conditions for the onset of primary wrinkling. Early studies restricted to linear elastic materials with high stiffness contrasts [4,5] have recently been extended to account for nonlinear elastic material behavior [14], substrate prestretch [13,22], and stiffness contrasts lower than one [21]. Beyond the onset of wrinkling, weakly nonlinear analyses can capture the initial post-buckling evolution [6,16,22]. However, those provide only limited insight into the wrinkle-to-fold transition in highly non-linear confined systems. Another practical tool to explore multiple bifurcations is the finite element method [10,23,24]. Primary wrinkling has been extensively studied [11,12,17,28] and recent experimental [2,6,34], theoretical [6,18,36], and computational [25,36,37] studies have significantly advanced our understanding of the period-doubling mode. However, higher order secondary wrinkling modes such as period tripling or period quadrupling remain unsatisfactorily understood [34]. Those are highly associated with substrate nonlinearity [6,7,37] and substrate prestretch [13,22,35]. Most studies have focused on stiffness ratios of layer to substrate greater than 100. In soft materials such as living tissue, elastomers and gels, however, the stiffness contrast between different layers is only moderate.

Here, we use the model system of a differentially compressed elastomeric bilayer with a stiffness contrast of $\mu_f/\mu_s = 20$ to experimentally study wrinkling instabilities. We consider the case of a sufficiently strong adhesion between the layers to avoid delamination. We focus on the formation of primary wrinkles as well as the transition from wrinkles to folds or secondary instability modes such as period doubling and period tripling by varying film thickness and substrate prestretch. We combine previously derived analytical models [6,13] with computational simulations to predict the experimental behavior at the onset of wrinkling, in the initial post-bifurcation range, and during the wrinkle-to-fold transition. Understanding the origin of surface morphologies in technical and biological systems and identifying the control parameters of
pattern selection will help to assist diagnosis and curing of certain diseases such as asthma [27], and to facilitate the fabrication of smart surfaces [31].

2. Experimental model

To experimentally study geometric primary and secondary instabilities, we used bilayers of two RTV-2 silicone rubbers with different mechanical properties. The experimental procedure included three steps as illustrated in Figure 2. First, we cast the soft rectangular substrate composed of ECOFLEX 00-50 (Smooth-On) using a custom-built two-part casting mold. We mixed the binary system (A/B) in the recommended ratio of 1:1 and subsequently homogenized it in a SpeedmixerTM (DAC 150 SP, Hauschild) at 1000 rpm for three minutes at room conditions (25°C at environmental humidity and pressure). To visibly distinguish between film and substrate, we added a small amount of white coloring pigment (1% of total weight, SILC-PIG white, Smooth-On). Next, we deaerated the mixture using a desiccator equipped with a vacuum pump prior to curing at 123°C for 15 h.

![Figure 2. Wrinkling instabilities in bilayered systems. a) Elastomeric substrate at its initial length \( L_0 \). b) Elastomeric film molded onto the prestretched substrate with length \( l \). c) Wrinkled film attached to relaxed substrate at length \( L \) with the wavelength \( \lambda \) and the wrinkle amplitude \( A \). \( \lambda_0 \) denotes the wavelength at the onset of wrinkling.](image)

Second, we uniaxially prestretched the substrate from its initial length \( L_0 \) to a new length \( l \) using a second custom-built combined stretching and casting device. This allowed us to cast the rigid film composed of BLUESIL ESA 7250 A/B (BlueStar Silicones) on top. We mixed the film binary system in the ratio of 10:1, added a combination of white and blue coloring pigments (1% of total weight, SILC-PIG white, SILC-PIG blue), homogenized, deaerated, and cross-linked it similar to the soft substrate. To induce buckling in the film, we gradually released the prestretch in the substrate. We measured the wavelength \( \lambda \), the amplitude \( A \), and the film thickness \( t \) of the resulting wrinkling pattern as indicated in Figure 2 via optical methods. When gradually relaxing the substrate, the wavelength \( \lambda \) decreases, the amplitude \( A \) increases, and the film length \( l \) remains constant. We back-calculate the critical wavelength \( \lambda_{\text{crit}} \) at the onset of wrinkling by approximating the arc length of the final wrinkling pattern \( \lambda_0 \) as illustrated in Figure 2

\[
\lambda_{\text{crit}} = \lambda_0 = \int_0^\lambda \sqrt{1 + \left(\frac{2\pi A}{\lambda} \cos \left(\frac{2\pi x}{\lambda}\right)\right)^2} \, dx = \text{const.} \quad (2.1)
\]

Substrate-prestretch instabilities with \( \lambda_{\text{crit}} = \lambda_0 = \text{const.} \) and decreasing \( \lambda \) are therefore conceptually different from film-growth instabilities with \( \lambda_{\text{crit}} = \lambda = \text{const.} \) and increasing \( \lambda_0 \) [21]. We obtained the Young’s moduli of both RTV-2 silicone rubbers by uniaxial tension tests (custom-built tensile testing machine, Hegewald & Peschke) conducted with dog bone samples according to DIN 53504 (Type S3) at room temperature and environmental conditions. Table 1 summarizes the Young’s moduli and failure stretches averaged over \( n=7 \) samples. In the following experiments, we systematically varied two major control parameters, the film thickness \( t \), from 0.1 to 1.2 mm, and the pre-stretch \( l/L_0 \), from 1.3 to 2.8.
3. Analytical model

Analytically, to obtain closed-form estimates for the critical conditions of primary wrinkling in the elastomeric bilayer, we used an extension of the classical Allen solution [1], which accounts for substrate prestretch [13]. We assumed uniaxial homogeneous tension in the substrate prior to film attachment with the principal stretches $\lambda_{1s}^0 = l/L_0$ and $\lambda_{3s}^0 = \lambda_{3s}^0 = 1/\sqrt{l/L_0}$. Then, the critical strain $\epsilon_{\text{crit}} = 1 - L_{\text{crit}}/l$ and the critical wavelength $\lambda_{\text{crit}}$ at the onset of wrinkling are

$$\epsilon_{\text{crit}} = 1 - \sqrt[3]{\frac{3}{\alpha}} \left( \frac{\lambda_{1s}^0}{\mu t} \right)^{2/3} \frac{1}{\left[ 1 + 2 \sqrt[3]{\lambda_{1s}^0} \right] \lambda_{1s}^0}$$

and

$$\lambda_{\text{crit}} = \frac{2}{\pi} \sqrt{\frac{\mu}{\lambda_{1s}^0}} \left[ 1 + \frac{2}{\sqrt[3]{\lambda_{1s}^0}} \right]^{3/2}$$

(3.1)

In addition, we can compute the critical film stretch as $\vartheta_{\text{crit}} = L_{\text{crit}}/l = 1 - \epsilon_{\text{crit}}$.

4. Computational model

Computationally, we simulated the relaxation of prestretch in the substrate by gradually shrinking the substrate in the horizontal direction from its prestretched length $l$ to the relaxed length $L$ [33]. We modeled shrinkage using the nonlinear field theories of mechanics supplemented by the theory of finite growth. We introduced the deformation gradient, which we decomposed into an elastic and a shrinkage part [19],

$$F = \nabla X \varphi = F^e \cdot F^s \quad \text{and} \quad J = \text{det} F = J^e J^s.$$  (4.1)

A similar multiplicative decomposition holds for the Jacobian $J$. Constitutively, we modeled both film and substrate as neo-Hookean materials with the strain energy function parameterized exclusively in terms of the elastic tensor $F^e$ and its Jacobian $J^e$,

$$\psi(F^e) = \frac{\lambda}{2} \ln^2 (J^e) + \frac{\mu}{2} \left[ F^e : F^e - 3 - 2 \ln (J^e) \right],$$  (4.2)

where $\lambda$ and $\mu$ are the Lamé constants. In the linear limit, we can calculate the Lamé constants from Young’s moduli in Table 1 assuming a Poisson’s ratio of $\nu = 0.45$ in both substrate and film. Following standard arguments of thermodynamics, we can introduce the Piola stress $P$ as energetically conjugate to the deformation gradient $F$,

$$P = \frac{\partial \psi(F^e)}{\partial F} = \frac{\partial \psi(F^e)}{\partial F^e} \cdot \frac{\partial F^e}{\partial F} = P^e \cdot (F^e)^{-t}.$$  (4.3)

The Piola stress enters the standard balance of linear momentum, the equation of mechanical equilibrium, which in the absence of volume forces reduces to

$$\text{Div}(P) = 0.$$  (4.4)

We imposed shrinkage as

$$F^s = \vartheta I + \left[ 1 - \vartheta \right] n \otimes n \quad \text{with} \quad (F^s)^{-t} = \frac{1}{\vartheta} I + \left[ 1 - \frac{1}{\vartheta} \right] n \otimes n,$$  (4.5)

where $\vartheta$ denotes the ratio between the current and the initial length of the substrate $L/l$, $I$ is the second order unit tensor, and $n$ is the normal in the current, shrunk configuration. We solved the

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<th>Table 1. Material parameters of film and substrate</th>
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<td>Young’s modulus</td>
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nonlinear set of equations using the finite element method in a custom-designed MATLAB-based environment [8]. We chose geometry and boundary conditions according to the experimental setup described in Section 2. The initial thickness of the substrate was 1cm, while we varied the initial film thickness between 0.05 and 0.7mm. As the substrate was clamped at both sides in the custom-built stretching device at state \( b \) in Figure 2 when the film was molded on top, its total length was \( l + 2L_{cl} \), where \( L_{cl} \) is the clamping length not included in the stretching process. We modeled the film to cover 7/8 of the total substrate length to resemble the experiment. We discretized both film and substrate with one tri-linear Q1 element per 0.25mm in \( x \)-direction, the film with one element per 0.125mm, and the substrate with one element per 0.25mm in the \( y \)-direction. In systematic mesh refinement studies, we had observed that this discretization was sufficient to provide accurate results. We assumed a plane strain state and further fixed the nodes at the left lower edge of the shrinking substrate in \( x \)- and \( y \)-direction and the nodes at the right lower edge in \( y \)-direction. We simulated the gradual release of substrate prestretch by incrementally reducing \( \vartheta \) and imply that \( \vartheta \) is equivalent to the film stretch. To trigger the instability, we pre-shrink a small band of two elements in the center of the film by one percent in volume.

5. Results

Figure 3 shows the wrinkling evolution in the elastomeric bilayer when substrate prestretch is gradually released from state \( b \) to \( c \) in Figure 2. When the compression in the film exceeds the critical value \( \vartheta^{\text{crit}} \), it bifurcates into wrinkles with the critical wavelength \( \lambda = \lambda^{\text{crit}} \). With further compression, wrinkle amplitudes grow and the wavelength \( \lambda \) decreases while the number of undulations remains constant. Eventually, the wrinkles transform into folds, which are defined as sharp inward folds with self-contact [34]. A movie of the relaxation process is provided in supplementary materials.

5.1. Primary bifurcation for varying film thickness

Figure 4 illustrates the influence of film thickness \( t \) on the wrinkling pattern at a constant prestretch of \( l/L_0 = 2.25 \). The critical wavelength of primary wrinkles \( \lambda^{\text{crit}} \) increases with increasing film thickness \( t \) from left to right. When the film thickness exceed a certain threshold, the failure stretch of the substrate material is not high enough to induce buckling.

Figure 5 compares the experimentally determined critical wavelength \( \lambda^{\text{crit}} \) as a function of film thickness \( t \) with analytical and computational predictions. Our results confirm that the critical wavelength linearly depends on the film thickness. Both analytical and computational models agree well with the experiment. The linear perturbation analysis accounting for prestretch in the
Figure 4. Primary wrinkling for varying film thicknesses of \( t = 0.2\)mm, \( t = 0.4\)mm, \( t = 1.0\)mm, and \( t = 1.2\)mm (from left to right).

Figure 5. Primary bifurcation. Wavelength vs. film thickness. Experimental measurement, analytical solution [14], and computational simulation.

substrate [13] slightly underestimates the critical wavelength. The computational model predicts the correct relation between wavelength and thickness, but we observe a slight offset on the axis of the ordinate. This can be attributed to the fact that we are limited to finite size of elements. Thus, the computational approach presented here can not capture the behavior towards the zero thickness limit [29].

In addition to the experimental results in the model problem of an elastomeric bilayer, we plot the wavelength versus the thickness of all dehydrated fruits shown in Figure 1. The data suggest that the stiffness contrast in dates and apricots is higher than in the elastomeric bilayers, while it is lower or almost identical in apples.

(b) Primary bifurcation for varying prestretch

Figure 6 illustrates the influence of varying prestretch \( l/L_0 = 1.30, 1.65, 2.00, \) and \( 2.50 \) on the wrinkling pattern for a constant film thickness \( t = 0.35\)mm. The corresponding stretches in the compressed film were \( \vartheta = 0.85, 0.75, 0.68, \) and \( 0.65 \). For a low prestretch \( l/L_0 = 1.30 \), the compression in the film hardly exceeds the critical value for primary wrinkling. For \( l/L_0 = 1.65 \), the confinement in the film has yet passed a secondary treshold and we observe period-doubles. Further increasing the prestretch to \( l/L_0 = 2.00 \) evokes a wrinkle-to-fold transition: neighboring period-doubles form self-contact. Interestingly, we also observe period-triples. For an even higher prestretch of \( l/L_0 = 2.50 \), the surface morphology reveals primary wrinkles with imminent transition into folds. This can be attributed to the fact that the critical film strain for period-doubling increases linearly with substrate pre-stretch [37]. Thus, for high prestretches, we again observe periodic wrinkling rather than period-doubling. Although varying prestretch highly controls secondary wrinkling patterns, we will first focus on the influence of prestretch...
on the evolution of primary wrinkling and exclusively analyze corresponding data. An elaborate discussion on secondary modes is subject of Section (c).

Figure 6. Wrinkling pattern for varying prestretch $l/L_0=1.30, 1.65, 2.00, \text{ and } 2.50$ with film stretches $\vartheta=0.85, 0.75, 0.68, \text{ and } 0.65$ (from left to right) and a film thickness of $t=0.35\text{mm}$.

Figure 7 plots the normalized amplitude $A/\lambda$ of wrinkles versus $\vartheta = L/l$. As primary wrinkles exhibit an approximately sinusoidal morphology, the evolution of the surface pattern with $\vartheta$ is fully characterized by the evolution of the ratio $A/\lambda$. We gradually increased the prestretch in the substrate $l/L_0$, which gradually increased the compression in the film when the substrate is relaxed (see c in Figure 2). The corresponding film stretches were $\vartheta=0.85, \vartheta=0.75, \vartheta=0.68, \vartheta=0.67, \vartheta=0.65, \text{ and } \vartheta=0.64$. With increasing compression, the amplitude of the wrinkles increases, while their arc length remains constant. Thus, the wavelength $\lambda$ decreases and the ratio $A/\lambda$ increases.

For a periodic profile, Brau et. al [6] provided an analytical expression for the $A/\lambda$ evolution after the onset of wrinkling from geometrical considerations

$$\frac{A}{\lambda} = \frac{1}{\pi} \sqrt{\frac{1}{1 - \delta}}, \tag{5.1}$$

where $\delta = 1 - 1/[l/L - [l/L_{\text{crit}} - 1]]$. This analytical approach slightly underestimates the ratio $A/\lambda$, while experimental and computational data agree well. This can be attributed to the assumption of perfectly sinusoidal wrinkles underlying Equation 5.1, which does not necessarily hold for the relatively low stiffness ratio of $\mu_t/\mu_s = 20$ investigated here.
(c) Secondary bifurcation for varying prestretch

Figure 8 highlights the occurrence of different secondary wrinkling modes, folds, period-doubles and period-triples, in elastomeric bilayers. Primary wrinkles emerge for moderate film compression or high substrate prestretches. The secondary period-doubling mode evolves for high compression in the film, which we achieved by increasing the prestretch $l/L_0$ or decreasing the film thickness $t$. However, the critical film strain for period-doubling increases linearly with increasing substrate prestretch \cite{37}. Thus, increasing substrate prestretch above a certain threshold, evokes wrinkles, which transform into folds before the onset of period-doubling as shown in Figure 4, left and Figure 6, right. When the film is too thick, the release of substrate prestretch alone is not sufficient to induce period-doubling. Period-triples are the unfavorable mode \cite{10} and, thus, only occur when the film is inadvertently perturbed. In Fig. 6, center-right, for instance, a fissure in the film evoked the period-tripling pattern. Wrinkles but also period-doubles and period-triples transition into fold and form self-contact when the compression is further increased. A computational approach to simulate period-doubling and period-tripling within a finite element framework has been presented in \cite{10}. Choosing a domain size of twice the characteristic wavelength $\lambda_{\text{crit}}$ triggers period-doubling, while a domain of three times $\lambda_{\text{crit}}$ suppresses the mode of period-doubling and triggers period-tripling. Here we adopt this framework and choose material parameters and dimensions according to the elastomeric bilayer.

To account for the stiffening effect due to substrate pre-tension \cite{14,37}, we correct the stiffness contrast according to Equation (3.1).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig8.pdf}
\caption{Secondary bifurcations. Experiment and simulation of instabilities beyond the first bifurcation point. Folding (top), period-doubling (middle), and period-tripling (bottom).}
\end{figure}

Figure 8 demonstrates the excellent agreement between experiment and simulation. The computational model is capable of predicting localization phenomena including folding, which can not be captured by perturbation approaches that use the sinusoidal bifurcation mode as the leading term \cite{6,22}. Although the mode of period-doubling is the energetically favorable mode \cite{10}, disturbed boundary conditions—a fissure in the film in the experiment and a careful selection of the domain size in the model—can suppress period-doubling and evoke period-tripling.
Figure 9 shows the evolution of neighboring wrinkling amplitudes with increasing film compression $\vartheta$ for simulations of period-doubling and period-tripling. Beyond the critical compression $\vartheta^{\text{crit}}$, the amplitudes of primary wrinkles first grow uniformly across the film. In the case of period-doubling, beyond a second threshold for the confinement, we observe a change in surface morphology - only every second amplitude continues to grow on the expense of its neighbors. In the case of period-tripling, at an even higher threshold only every third amplitude continues to grow. Figure 9 indicates that period-tripling requires higher compression in the film than period-doubling. This agrees well with the experimental findings in Fig. 6, where we observed period-doubles for a lower compressive strain than period-triples. As such, period-tripling only emerges if period-doubling is suppressed, numerically by constraining boundary conditions or experimentally by structural imperfections.

### 6. Conclusion

We have performed elastomeric bilayer experiments to study primary and secondary wrinkling instabilities for a moderate stiffness contrast between film and substrate of $\mu_f/\mu_s = 20$. Both analytical and computational models capture the behavior of the system at the onset of wrinkling and in the initial post-buckling regime. In addition, computational modeling allows us to predict the actual surface morphology of secondary wrinkling modes and the transition into folds in a highly compressed film. Our experiments confirm that the wavelength of primary wrinkles increases linearly with film thickness. Depending on substrate prestretch and boundary conditions, wrinkles transform into period-doubles, period-triples, and folds. Although period-doubling is the favorable secondary wrinkling mode, it can be suppressed by disturbed boundary conditions and we observe a period-tripling bifurcation instead. In the experiments, a fissure in the film evoked period-tripling. In the simulations, we selected the domain size to three times the wavelength to suppress period-doubling and evoke period-tripling [10]. The numerical prediction that period-tripling requires higher film compression than period-doubling agrees well with the observation that period-tripling occurs for higher substrate prestretch and thus higher film compression in the experiments. With further compression beyond the secondary bifurcation point, period-doubles or period-triples transform into folds before period-quadrupling or period-octupling can emerge. This observation demonstrates that the finite thickness of the film geometrically limits the emergence of higher order wrinkling modes: the pattern remains constant.
as soon as self-contact occurs and folds appear [7], for instance in Fig. 4, left. Notably, the folding mode can arise after period-doubling or period-tripling, which had been previously indicated computationally [10,37]. In general, folding occurs when the system favors inward displacement, while period-doubling occurs when the system favors outward displacements [6,22,35]. At significantly higher stiffness contrasts than $\mu_f/\mu_s = 20$ used in the present study, wrinkles would transform into ridges instead of period-doubles [13,34].

Our experiments confirm that the critical film compression for period-doubling increases with substrate prestretch, which agrees with recent numerical studies [37]. For high prestretches, periodic wrinkles transition into folds before period-doubling can emerge as shown in Fig. 6, right. Higher wrinkling modes such as period-doubling and period-tripling have been observed at low stiffness contrasts in mammalian brains [10], whereas period-doubling and period-quadrupling have been recreated at high stiffness contrasts in polymer experiments [6] and in numerical studies [25]. Nevertheless, experimental evidence towards higher wrinkling modes remains sparse. Elaborate studies in model systems like the one presented in this study should further elucidate those higher order instabilities in the future. The computational model presented in Section 4 captures the nonlinearities in terms of material behavior, geometry, and boundary conditions, however, substrate shrinkage is not necessarily equivalent to substrate prestretch. While the influence of substrate prestretch, film growth, and whole domain compression on the critical conditions at the onset of wrinkling has been assessed analytically [21], the differences concerning the highly nonlinear post-buckling regime remain unclear and require further investigations.

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References