

Supplementary Material: Interfacial dynamics of a bi-layer system with a small viscosity ratio: A simpler mathematical model

In this section we present some additional simulations demonstrating further features of the model, and animations of the simulations presented in the main text.

We first discuss a two layer film evolving according to the simplified model (38)-(41), where the top film is initially perturbed with an out-of-phase perturbation of the same wave number as the lower film, namely $f(x) = 1/r - a \cos(k_{max}^S x)$ and $h_2(x, 0) = 1 + a \cos(k_{max}^S x)$ with $a = 0.001$ and values of k_{max}^S (wavenumber of maximum growth as predicted by LSA of the simplified model) as specified in the caption to Fig. S1. As in the scenarios presented in the main text, different initial relative film thicknesses are considered, characterized by the parameter r (the ratio between total unperturbed bilayer thickness and lower film thickness). Figure 1(a) shows the evolution of the two layer system for $r = 1.05$. Here, we observe that an initially out-of-phase (varicose) configuration will continue to evolve out-of-phase: the upper layer forms peaks around $x = 0, \pm 0.6$ and valleys around $x = \pm 0.3$. As time progresses, the lower layer forms peaks around $x \approx \pm 0.3$, which continue to grow in the course of the simulation, indicating dewetting of the upper layer in those regions. A similar evolution is observed as r increases to $r = 1.10$ (shown in Fig. 1(b)) and $r = 1.15$ (shown in Fig. 1(c)): the top layer, initially out-of-phase with the lower layer, continues to evolve out-of-phase. We observe that the upper layer moves at a faster rate than the lower layer initially, forming peaks around $x \approx 0, \pm 2$ when $r = 1.10$ and $x = 0, \pm 6$ when $r = 1.15$. As time progresses, the lower layer starts moving at a faster rate (shown in animations `Animation_Fig_1a_supp - Fig_1c_supp.avi` provided as Supplementary materials), with peaks around $x \approx \pm 1$ when $r = 1.10$ and $x \approx \pm 3.55$ when $r = 1.15$ respectively. We predict pinch-off of the upper layer at these points. This behavior is qualitatively similar to that seen in Figs. 5 and 6, but occurs on a faster timescale due to the imposed initial condition being already varicose.

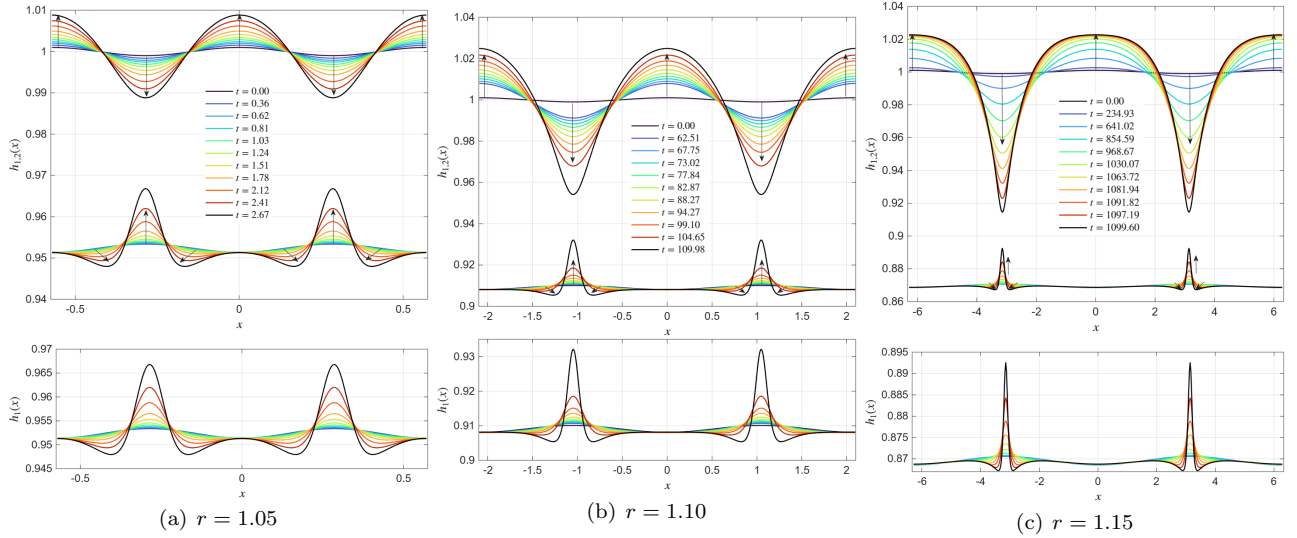


FIG. S1. Simplified Model: Evolution of a two layer film with an out-of-phase perturbed bottom layer of height $f(x) = 1/r - 0.001 \cos(k_{max}^S x)$, a perturbed top layer of total height $h_2(x, 0) = 1 + 0.001 \cos(k_{max}^S x)$, with $\varepsilon = 0.01$ for (a) $r = 1.05$, $k_{max} = 11$ and $t_{max} = 2.67$, (b) $r = 1.10$, $k_{max} = 3$ and $t_{max} = 109.98$ and (c) $r = 1.15$, $k_{max} = 1$ and $t_{max} = 1099.60$. Arrows indicate the evolution of each film over time. The figures on the second row illustrate a closeup of the evolution of the bottom layer.

Supplementary animations (`Animation_Fig_5a - Fig_5c.avi`) show the evolution of a two-layer film with a flat bottom, $f(x) = 1/r$, and a perturbed top layer $h_2(x, 0) = 1 + 0.001 \cos(k_{max}^S x)$ for $r = 1.05, 1.10$ and $r = 1.15$. These correspond to simulations presented in Fig. 5 of the main text. In all cases considered, the two layers evolve out-of-phase with one another (varicose mode), with the upper layer deforming at a faster rate than the lower one. This leads to dewetting in the upper layer, with the thicker upper films taking longer to dewet.

Supplementary animations (`Animation_Fig_6a - Fig_6c.avi`) show the evolution of a two layer film with an in-phase perturbed bottom, $f(x) = 1/r + 0.001 \cos(k_{max}^S x)$, and a similarly perturbed top, $h_2(x, 0) = 1 + 0.001 \cos(k_{max}^S x)$ for $r = 1.05, 1.10$ and $r = 1.15$. These animations correspond to simulations shown in Fig. 6 of the main text. Although the layers start in-phase, the system evolves toward an out-of-phase configuration, with the upper layer deforming

faster than the lower layer. Over time, the lower layer develops varicose deformations, which continue to grow over the course of the simulation. Thicker films ($r = 1.10$ and $r = 1.15$) show a similar evolution, where the initially in-phase system evolves out-of-phase, with the upper layer deforming at a faster rate than the lower one.

Supplementary animations **Animation_Fig_7a.avi** and **Animation_Fig_7b.avi** show the evolution of the two-layer system where both interfaces are perturbed with different wave numbers: **Animation_Fig_7a.avi** considers the scenario where the top layer is initially perturbed with a wave number $k = 3$ while the bottom layer is perturbed with a wave number $k = 6$ and **Animation_Fig_7b.avi** considers the flipped scenario in which a perturbation of wave number $k = 6$ is applied to the upper layer while the lower layer is perturbed with a wave number $k = 3$. In both cases, the system transitions to a varicose instability, ultimately leading to dewetting in the upper layer, albeit more slowly in the flipped scenario than in the first one.

Supplementary animations (**Animation_Fig_8a - Fig_8c.avi**) show the evolution of a two-layer film with a flat bottom, $h_1(x, 0) = 1/r$, and a perturbed top layer $h_2(x, 0) = 1 + 0.001 \cos(k_{max}^S x)$ for $r = 1.05, 1.10$ and $r = 1.15$ using the modified PBMT/FG model. These correspond to simulations presented in Fig. 8 which are compared with Fig. 5 of the main text. We observe that both models predict out-of-phase evolution with similar timescales, though they differ quantitatively: our simplified model predicts slightly greater movement in the lower interface and slightly less movement in the upper one, compared with the PBMT/FG model.