



Interfacial dynamics of a bi-layer system with a small viscosity ratio: a simple mathematical model

Ensela Mema¹ · Lou Kondic² · Linda J. Cummings²

Received: 20 October 2025 / Accepted: 2 June 2026
© The Author(s) 2026

Abstract

Stability properties of thin liquid films on solid substrates are relevant in industrial applications such as coating and drying processes. The study of such systems helps researchers develop methods to stabilize thin films or to break them into droplets in a controlled manner. While the dynamics of film rupture of single-fluid systems have been thoroughly investigated, the stability properties of systems comprising two immiscible fluid layers have received less attention, being more complex due to the coupled interactions between the liquid–liquid and liquid–gas interfaces. This work focuses on developing a simplified, asymptotically-consistent mathematical model that describes the evolution of two immiscible liquid layers, of very different viscosities, on a solid substrate. We use the long-wave approximation scalings to obtain the evolution equations for the liquid–liquid and liquid–gas interface in the case where the viscosity ratio of the two liquid layers is asymptotically small. Analytical and numerical methods are used to gain insight into the conditions under which dewetting/rupture of the film occurs. Our model results are also compared to those arising from existing models in the literature.

Keywords Linear stability analysis · Long-wave approximation · Thin film stability and rupture · Two-layer thin films

1 Introduction

The study of two-layered immiscible thin films, where two Newtonian films of small aspect ratio are placed on a substrate as separate layers, has attracted significant inter-

✉ Ensela Mema
emema@kean.edu

Lou Kondic
kondic@njit.edu

Linda J. Cummings
linda.cummings@njit.edu

¹ Department of Mathematical Sciences, Kean University, Union, NJ, USA

² Department of Mathematical Sciences, New Jersey Institute of Technology, Newark, NJ, USA

est due to wide-ranging applications. These include coating and drying processes [1], biological scenarios such as the liquid lining of lung airways [2], and geophysical settings like mud and lava flow, ice sheet and glacier flow models, and snow avalanche formation [3, 4] to name just a few. A major focus in bi-layer film research is understanding the dynamics leading to instability, dewetting and/or rupture of one or both films. While the dynamics of film rupture in single-layer systems have been extensively studied (see the comprehensive review by Craster and Matar [5] and references therein), the stability properties of two-layer systems are more complex due to the possibility of van der Waals' interactions of the liquids with each other and with the underlying substrate, and hydrodynamic coupling at the interface between the two liquids [6, 7].

Multiple variations on this theme have been studied using long-wave theory, including the effects of gravity induced by an inclined substrate. Early work by Kao [8] performed stability analysis of a two-layer viscous system with different densities flowing down an inclined substrate, showing that the two-layer flow is more stable or unstable than the homogeneous case of equal depth, depending on whether the upper fluid is lighter or heavier than the lower one. Kao further investigated the effect of the viscosity ratio, demonstrating that it can either stabilize or destabilize long-wavelength surface modes [9]. Later work by Tilley et al. [10] investigated the stability of two immiscible fluids in an inclined channel, showing how parameters such as wave number, channel thickness and interfacial height influence stability; and more recently, Bhat and Samanta [11] examined two-layer thin films flowing down a slippery inclined plane with insoluble surfactants at both the interface and free surface, identifying four distinct instability modes. This latter work demonstrates how viscosity, density or thickness ratios, and wall slip, among other factors, influence the stability of the system. For a broader overview of bi-layer flows on inclined planes, see the reviews by Chattopadhyay et al. [12] and Craster and Matar [5].

On smaller lengthscales gravity may be negligible and other forces, particularly surface tension and van der Waals-type effects, may dominate behavior. Key studies that are particularly relevant to our investigation include work by Pototsky et al. [13], Fisher and Golovin [14], and Bandyopadhyay et al. [15], that take account of interactions (modeled by disjoining pressures) between one or both liquids and the underlying substrate. These works all use long-wave theory to derive coupled nonlinear fourth-order evolution equations for the free liquid–liquid and liquid–gas interfaces. Pototsky et al. [13] and Bandyopadhyay et al. [15] independently derived identical coupled nonlinear evolution equations for the profiles of the liquid–liquid and liquid–gas interfaces in two-layer films, incorporating destabilizing long-range van der Waals forces. Interactions of the liquids with the substrate (and possibly with each other) lead to exotic instabilities: in the studied systems two distinct modes of surface instability were identified: zigzag and varicose. A “zigzag” or “bending” mode describes in-phase deformations occurring simultaneously at both the upper and lower free interfaces, whereas a “varicose” or “squeezing” mode refers to out-of-phase deformations between these interfaces. Each may lead to breakup of one (or both) of the films into drops that are separated by precursor layers, an autophobic behavior commonly referred to as “dewetting”. The types of models considered in [13–16] and the

mathematical model considered in the present work typically lead to such behavior in the unstable regime, with the details depending on the disjoining pressure model used.

Pototsky et al. [13] demonstrated that both instability modes zigzag and varicose can be exhibited in a two-layer system on a horizontal substrate, potentially leading to rupture either at the substrate or at the liquid–liquid interface. They further observed that, when the two layers differ greatly in thickness, the faster-evolving layer can drive and significantly accelerate the growth of surface modulations in the slower layer. Bandyopadhyay et al. [15] classified the two-layer systems into several cases, using linear stability analysis to show that the type of instability (zigzag or varicose) depends on the relative magnitudes of the surface tensions of the liquid layers and the substrate. In a subsequent study, Pototsky et al. [16] investigated a similar immiscible bi-layer system, with particular attention paid to the roles of both long- and short-range intermolecular interactions. The linear stability analysis again reveals that both zigzag and varicose modes can manifest, depending on the ratios of layer thicknesses, viscosities, and surface tensions. In particular, when short-range interactions (typical of polar liquids) are included, it becomes possible to influence stability by adjusting the layer thicknesses, and the mode type can change during the instability evolution.

Independently, Fisher and Golovin [14] show, through nonlinear stability analysis, that a two-layer liquid film can exhibit complex dewetting behavior and interesting film morphologies, even when only long-range van der Waals interactions are present. They identify a range of parameters where the two-layer film exhibits two distinct types of autophobic behavior, breaking up into drops that coarsen over time: one type of behavior leads to drops of the lower liquid separated by a thin wetting layer and covered by the layer of the upper liquid; while the other consists of drops of the upper liquid resting on a slightly perturbed lower liquid. Each type of structure coarsens over time, leading to a final steady state that consists of a single drop in the dewetted layer.

Also relevant to our work are studies of bi-layer thin films where the bottom layer is a highly viscous polymer melt while the top layer consists of a lower viscosity polymer, a scenario of great importance in industrial and polymer-processing technologies such as multilayer coextrusion [17–19], spin coating of polymer solutions [7, 20], slot-die coating, etc. [21].

In this context, mathematical models investigating bi-layer systems where both layers are viscoelastic have been developed and analyzed; for example, by Jachalski *et al.* [22, 23]. The first of these works [22] focuses on establishing conditions for an asymptotically-consistent reduction of the free boundary problem in a two-layer liquid system to a thin-film model incorporating nonlinear viscoelastic rheology. In the companion work [23], the authors develop and compare two thin-film models for viscoelastic liquid–liquid systems, considering varying levels of interfacial slip at the solid–liquid and liquid–liquid interfaces. The findings highlight that, along with the thickness and rheology of the liquid layers, interfacial slip significantly influences the stability characteristics of these liquid–liquid systems. Bilayer systems where just one layer is viscoelastic have also been used to model fluid dynamics on soft substrates, with the lower viscoelastic layer representing the soft substrate, see, e.g., the works by van Gorcum et al. [24] and Henkel et al. [25]. In this context, recent works by Style et al. [26] and by Tamim Bostwick [27] that studied viscous drops spreading on viscoelastic fluid layers are also of interest. Among other works modeling such

soft substrate systems, we also note the study by Bommer et al. [28], who consider dewetting in a coupled bi-layer system comprising a dewetting layer and a deformable (liquid) substrate. The authors develop a theoretical model, which they simulate and compare with experiments (particularly with regard to interface morphologies) on polymer-polymer systems, and use to study how the dewetting rate depends on system parameters.

Inspired by the type of polymer-processing applications described above, where a polymer melt forms a highly viscous substrate, it is natural to consider configurations in which a much more viscous bottom layer is placed under a less viscous upper layer. In the present work, we restrict attention to the case of two immiscible Newtonian liquids as the simplest setting that can systematically probe the effects of such bi-layer flows with strong viscosity contrast. Extending the analysis to include a viscoelastic lower layer will be the focus of future work. Here, we present a simplified, asymptotically-consistent, mathematical model for the evolution of two immiscible Newtonian liquid films, in a particular limit where the lower film is significantly more viscous than the upper one. This viscosity contrast causes the lower layer to flow much more slowly, allowing the system to be considered as another possible prototype model for flow on a soft substrate (represented here by the highly viscous lower layer). Our model differs from those developed by Pototsky et al. [13], Bandyopadhyay et al. [15] and Fisher and Golovin [14], primarily due to differences in the disjoining pressure and the scaling assumptions used in the derivation. Specifically: (i) by assuming an asymptotically small viscosity ratio between the layers, our approach leads to a simpler, decoupled system of equations describing the evolving fluid thicknesses, in contrast to the fully coupled PDE systems in [13–15]; and (ii) our model adopts the intermolecular potential described by Jachalski et al. [23], which allows only interactions between the upper and lower fluid layers, rather than one of the forms used in the earlier works [13–16] (which assume that both layers also interact with the underlying supporting substrate). As the viscosity ratio decreases, in the appropriate limit these existing models [13–16] do ultimately converge to our simplified framework (modulo the noted differences in disjoining pressure); but for the scenarios considered here those models are not strictly asymptotically consistent, since they retain the terms in the asymptotically small viscosity ratio. We note that, while the fully-coupled models are computationally-tractable in the case of two-dimensional flow, our simplified model will offer significant computational savings when extended to three-dimensional bilayers on large domains.

We also carry out a linear stability analysis of the flat-interfaces case, which reveals that the system's stability is sensitive to the initial relative layer thicknesses and the disjoining pressure. Numerical simulations validate these findings, showing that a thicker upper layer stabilizes the system. Additionally, when applying the same disjoining pressure to the earlier models [13–15], we observe that the chosen intermolecular potential induces varicose evolution between the layers, leading ultimately to dewetting in the upper layer only. This contrasts with the earlier findings, where the multiple layer-layer and layer-substrate interactions can lead to dewetting in both upper and lower layers.

2 Mathematical model

We develop a mathematical model that describes the evolution of two immiscible Newtonian liquid layers: the lower layer (distinguished by subscript 1) rests on a solid substrate at $z = 0$, while the upper layer (subscript 2) occupies the region between the lower film and the gas phase. Figure 1 depicts the scenario described (our study in this manuscript is restricted to two-dimensional films, though the models are readily extended to three dimensions). The equation $z = h_1(x, t)$ represents the location of the liquid–liquid interface that separates the two layers, while $z = h_2(x, t)$ represents the location of the liquid–gas interface. We assume both gravity and inertia are negligible, so that the dynamics of the two layers can be described by the incompressible Stokes equations:

$$\partial_x u_i + \partial_z w_i = 0 \quad i = 1, 2, \quad (1)$$

$$\mu_i (\partial_{xx} u_i + \partial_{zz} u_i) = \partial_x p_i \quad i = 1, 2, \quad (2)$$

$$\mu_i (\partial_{xx} w_i + \partial_{zz} w_i) = \partial_z p_i \quad i = 1, 2, \quad (3)$$

where subscripts $i = 1, 2$ denote that the equations are to be solved on the lower and upper layers, respectively. Here, u_i and w_i represent the fluid velocity components in the horizontal (x) and vertical (z) directions, and p_i denotes the pressure, in each fluid layer. We consider films of large lateral extent, which we model by imposing periodic lateral boundary conditions, to be specified later in Sect. 4.1. No-slip and no-penetration boundary conditions are imposed at the solid/liquid interface, leading to:

$$u_1 = 0, \quad w_1 = 0, \quad \text{on } z = 0. \quad (4)$$

The same no-slip and no-penetration conditions, together with the kinematic boundary condition and a tangential and normal stress balance, lead to the following equations holding at the liquid–liquid interface $z = h_1$:

$$u_1 = u_2, \quad (5)$$

$$w_1 = w_2, \quad (6)$$

$$\partial_t h_1 = w_1 - u_1 \partial_x h_1, \quad (7)$$

$$(1 - \partial_x^2 h_1)(\tau_{2,12} - \tau_{1,12}) + \partial_x h_1(\tau_{2,22} - \tau_{2,11} - \tau_{1,22} + \tau_{1,11}) = 0, \quad (8)$$

$$\begin{aligned} p_2 - p_1 - \phi'(h_2 - h_1) + \frac{\partial_x^2 h_1(\tau_{1,11} - \tau_{2,11})}{(1 + \partial_x^2 h_1)} \\ - \frac{2\partial_x h_1(\tau_{1,12} - \tau_{2,12})}{(1 + \partial_x^2 h_1)} + \frac{(\tau_{1,22} - \tau_{2,22})}{(1 + \partial_x^2 h_1)} \\ = \sigma_1 \frac{\partial_{xx} h_1}{[1 + \partial_x^2 h_1]^{3/2}}. \end{aligned} \quad (9)$$

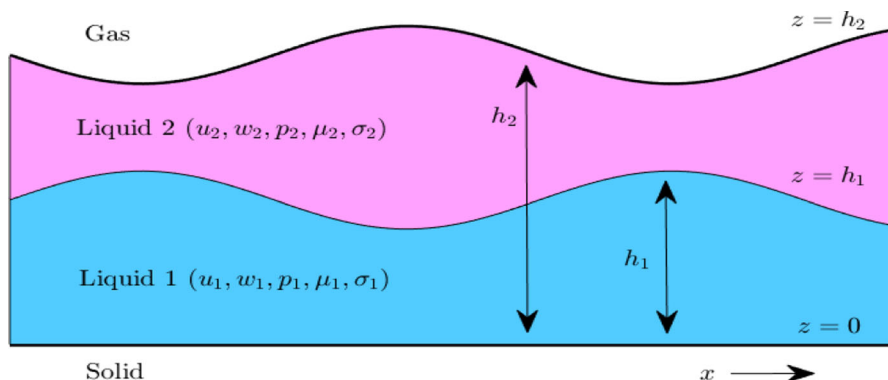


Fig. 1 Schematic of two immiscible Newtonian liquid layers where the lower layer is resting on a solid substrate and the upper layer is placed on top of the lower layer

Here, $\tau_{i,kj}$ denotes the (k, j) -component of the viscous (or deviatoric) part of the stress tensor for layer i , and ϕ is an interaction potential or disjoining pressure. In this study ϕ is assumed to represent interaction between the upper and lower fluids only, hence it is a function only of the upper film thickness $h_2 - h_1$ (though the model can be easily extended to include a potential that includes interactions between each fluid and the underlying rigid substrate as well). The parameter σ_1 is the surface tension between the two liquids. Since both liquid layers are assumed Newtonian, the deviatoric stress tensors are proportional to the strain rate and take the following form:

$$\tau_i = \mu_i \begin{pmatrix} 2\partial_x u_i & \partial_z u_i + \partial_x w_i \\ \partial_z u_i + \partial_x w_i & 2\partial_z w_i \end{pmatrix}. \quad (10)$$

The kinematic and stress-balance conditions at the liquid–gas interface lead to the following boundary conditions at $z = h_2$:

$$\partial_t h_2 = w_2 - u_2 \partial_x h_2, \quad (11)$$

$$(1 - \partial_x^2 h_2) \tau_{2,12} - \partial_x h_2 (\tau_{2,22} - \tau_{2,11}) = 0, \quad (12)$$

$$-p_2 + \phi'(h_2 - h_1) + \frac{\partial_x^2 h_2 \tau_{2,11} - 2\partial_x h_2 \tau_{2,12} + \tau_{2,22}}{(\partial_x^2 h_2 + 1)} = \sigma_2 \frac{\partial_{xx} h_2}{(\partial_x^2 h_2 + 1)^{3/2}}, \quad (13)$$

where σ_2 is the surface tension at the liquid 2–gas interface.

As noted above, we focus on the scenario where the surface forces arising from interactions of either fluid layer with the underlying solid substrate ($z \leq 0$) are negligible, while the interactions of the top layer ($h_1 \leq z \leq h_2$) with the bottom layer ($0 \leq z \leq h_1$) are relevant and can lead to dewetting, with the upper layer breaking up into droplets separated by much thinner regions of thickness h_* , sometimes referred to as precursor films. This is consistent with our view of the setup as a prototype for a film (the upper layer) flowing on a soft deformable substrate (the lower layer), where the rigid interface at $z = 0$ serves only to support the system. The interaction potential

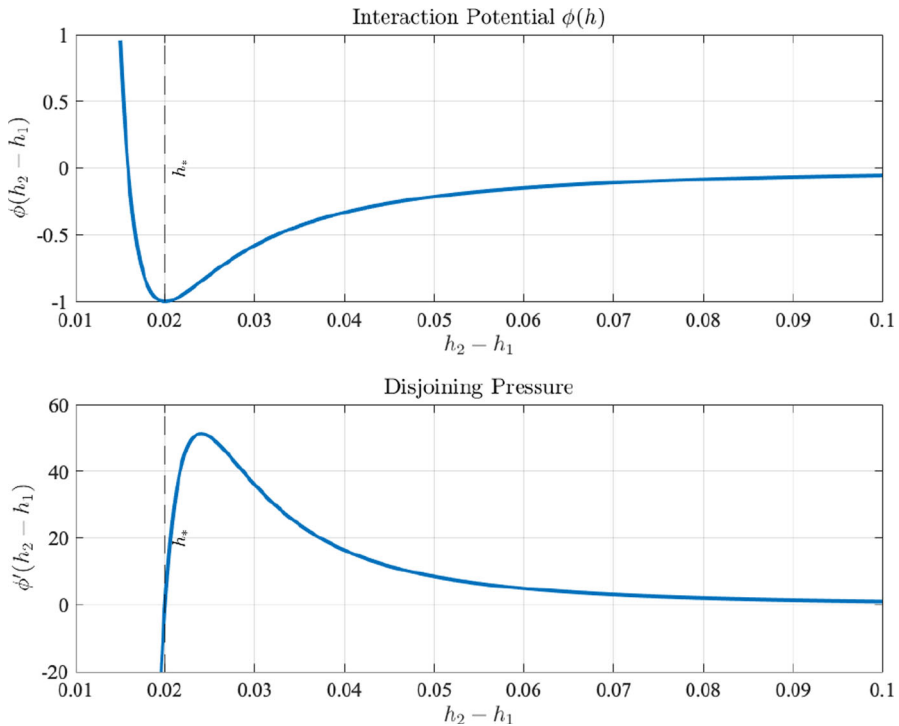


Fig. 2 Interaction potential $\phi(h_2 - h_1)$ (top) and corresponding disjoining pressure $\Pi(h_2 - h_1) = -\phi'(h_2 - h_1)$ (bottom) for $h^* = 0.02$ and $\phi^* = 1$

then depends only on the thickness of the upper layer, $h_2 - h_1$; following Jachalski et al. [23], we impose the following form:

$$\phi(h_2 - h_1) = \frac{8}{3}\phi_* \left[\frac{1}{8} \left(\frac{h_*}{h_2 - h_1} \right)^8 - \frac{1}{2} \left(\frac{h_*}{h_2 - h_1} \right)^2 \right], \quad (14)$$

where $\phi = -\phi_*$ when $h_2 - h_1 = h_*$. Equation (14) represents the interaction energy between the upper and lower layers. It captures a balance between the destabilizing long-range van der Waals forces, modeled by the term $(h_2 - h_1)^{-2}$, and stabilizing short-range repulsive forces, represented by $(h_2 - h_1)^{-8}$. The parameter h^* denotes the equilibrium film thickness between the attractive and repulsive forces, while ϕ^* is the minimum value of the interaction energy attained at that thickness. Figure 2 displays both the interaction potential ϕ and the associated disjoining pressure $\Pi(h_2 - h_1) = -\phi'(h_2 - h_1)$, where the prime denotes differentiation with respect to the single argument of ϕ . As seen in the upper graph, the potential exhibits a minimum at $h_2 - h_1 = h^*$, reflecting vanishing disjoining pressure, and the effective minimum upper layer thickness that can be obtained. With increasing layer thickness the potential and disjoining pressure both approach zero, indicating that the interaction weakens as the layers move farther apart.

We consider scenarios (e.g., sufficiently thin top layer and moderately thin bottom layer) where the interaction forces lead to dewetting of the system. Integrating the continuity equation (1) with respect to z across each layer and applying the boundary conditions (4) and (7) for the lower layer and (5), (6) and (11) for the upper layer, leads to the following dimensional flux conservation equations:

$$\partial_t h_1 + \partial_x \left(\int_0^{h_1} u_1 \, dz \right) = 0, \quad (15)$$

$$\partial_t h_2 + \partial_x \left(\int_0^{h_1} u_1 \, dz \right) + \partial_x \left(\int_{h_1}^{h_2} u_2 \, dz \right) = 0. \quad (16)$$

2.1 Nondimensional governing equations

We focus on the case where the viscosity ratio between liquid 2 and liquid 1 is small (so that the lower layer flows much more slowly than the upper layer), while the surface tension ratio is $O(1)$. We nondimensionalize the governing equations using the lubrication approximation scalings based on the upper (less viscous) liquid, valid if the typical film thicknesses H are much smaller than the characteristic lateral lengthscale L of the layer, so that the aspect ratio $\varepsilon = H/L \ll 1$:

$$\begin{aligned} x &= L\tilde{x}, \quad (z, h_i) = H(\tilde{z}, \tilde{h}_i), \quad t = \frac{L}{U}\tilde{t}, \\ (p_i, \phi') &= P(\tilde{p}_i, \tilde{\phi}'), \quad (u_1, u_2) = U(\varepsilon^2 \tilde{u}_1, \tilde{u}_2), \\ (w_1, w_2) &= U(\varepsilon^3 \tilde{w}_1, \varepsilon \tilde{w}_2). \end{aligned} \quad (17)$$

Here, U denotes the characteristic lateral velocity of fluid in the upper layer, and we assume that the typical thickness H (of both layers) is known. The different velocity scalings in the two layers reflect our inherent assumption on the viscosity ratio. More specifically, we define the viscosity and surface tension ratios of the two films by:

$$\frac{\mu_2}{\mu_1} = \varepsilon^2 M, \quad \sigma = \frac{\sigma_2}{\sigma_1}, \quad (18)$$

where M and σ are both assumed to be $O(1)$ quantities. Since the intermolecular potential ϕ defined in Eq. (14) is the dominant mechanism driving the bi-layer, we base the pressure scale on the characteristic value ϕ_* as follows:

$$P = \frac{8}{3} \frac{\phi_*}{H}. \quad (19)$$

Balancing the terms in the stress condition (13) and using the momentum equation (2), we obtain the further relations for the pressure and velocity scales P and U respectively:

$$P = \frac{\sigma_2 H}{L^2}, \quad U = \frac{\varepsilon^3 \sigma_2}{\mu_2}; \quad (20)$$

combining (19) and (20) then gives the appropriate lateral lengthscale as $L = H(3\sigma_2/(8\phi_*))^{1/2}$. Choosing the lengthscale to be determined by the physics of the interactions in this way reflects our assumption that we consider lateral domains sufficiently large that the simulation outcomes are determined by these physics, rather than any geometric constraints. With these scalings the nondimensional disjoining pressure term takes the form:

$$\tilde{\phi}' = \frac{1}{b} \left[- \left(\frac{b}{\tilde{h}_2 - \tilde{h}_1} \right)^9 + \left(\frac{b}{\tilde{h}_2 - \tilde{h}_1} \right)^3 \right], \quad (21)$$

where $'$ denotes the derivative of ϕ with respect to its argument. The parameter $b = h_*/H \ll 1$ denotes the dimensionless precursor layer thickness (to be chosen when carrying out numerical simulations).

By substituting the above scalings in (1)–(3) and retaining only the leading-order terms in ε , we obtain the following dimensionless continuity and momentum equations for the lower and upper layers, respectively. At this stage, we drop the tilde notation, since everything will be dimensionless henceforth. For the lower layer ($0 < z < h_1$):

$$\begin{cases} \partial_x u_1 + \partial_z w_1 = 0, & (22) \end{cases}$$

$$\begin{cases} \partial_x p_1 = \frac{1}{M} \partial_{zz} u_1, & (23) \end{cases}$$

$$\begin{cases} \partial_z p_1 = 0. & (24) \end{cases}$$

For the upper layer ($h_1 < z < h_2$):

$$\begin{cases} \partial_x u_2 + \partial_z w_2 = 0, & (25) \end{cases}$$

$$\begin{cases} \partial_x p_2 = \partial_{zz} u_2, & (26) \end{cases}$$

$$\begin{cases} \partial_z p_2 = 0. & (27) \end{cases}$$

The no-slip and no-penetration boundary conditions (shown in (4)) at the solid–liquid interface $z = 0$ are:

$$z = 0 : \quad u_1 = 0 = w_1. \quad (28)$$

At the liquid–liquid ($z = h_1$) and liquid–gas ($z = h_2$) interfaces, the no-slip and no-penetration conditions, together with the dynamic boundary conditions (Eqs. (5)–(9) for $z = h_1$ and (11)–(13) for $z = h_2$) become, at leading order:

$$z = h_1 : \quad u_2 = 0 = w_2, \quad (29)$$

$$\frac{1}{M} \partial_z u_1 - \partial_z u_2 = 0, \quad (30)$$

$$p_2 - p_1 - \phi' = \partial_{xx} h_1, \quad (31)$$

$$z = h_2 : \quad \partial_z u_2 = 0, \quad (32)$$

$$-p_2 + \phi' = \sigma \partial_{xx} h_2. \quad (33)$$

The dimensionless counterparts of (15)–(16), which incorporate the kinematic boundary conditions and represent conservation of flux, are given by:

$$\partial_t h_1 + \varepsilon^2 \partial_x \left(\int_0^{h_1} u_1 \, dz \right) = 0, \quad (34)$$

$$\partial_t h_2 + \partial_x \left(\int_{h_1}^{h_2} u_2 \, dz \right) = 0, \quad (35)$$

where we retain the $O(\varepsilon^2)$ term in (34) since that describes the leading-order motion of the lower layer that takes place on the chosen timescale (we do not retain it in (35) since that moves by an $O(1)$ amount regardless). Equation (34) confirms our expectation that, to leading order (on the timescale of motion of the upper layer), the highly viscous lower layer is stationary ($\partial_t h_1 = 0$). To describe the motion that arises at order ε^2 , we introduce an asymptotic expansion:

$$h_1(x, t) = f(x) + \varepsilon^2 h_{11}(x, t) + O(\varepsilon^4), \quad (36)$$

where $f(x)$ represents the leading-order thickness of the lower layer, and $h_{11}(x, t)$ represents the height of the lower layer at the next order. Substituting the configuration specified in (36) into (34) and retaining the $O(\varepsilon^2)$ terms, we obtain:

$$\partial_t h_{11} + \partial_x \left(\int_0^{h_1} u_1 \, dz \right) = 0. \quad (37)$$

To evaluate the integral in (37), we first solve the momentum equation (23) for u_1 , subject to (30). This provides an expression for $(\partial_z u_1)$, which is then integrated in z . Imposing the no-slip condition at $z = 0$ (28) yields the final expression for u_1 . Substituting this result back into (37) and integrating with respect to z gives the governing equation for $h_{11}(x, t)$:

$$\partial_t h_{11} = M \partial_x \left\{ \frac{1}{3} f(x)^3 \partial_x p_1 + \left(\frac{f(x)^2}{2} (h_2 - f(x)) \right) \partial_x p_2 \right\}. \quad (38)$$

Similarly, the evolution equation for $h_2(x, t)$, the height of the upper layer, is obtained by first solving (26), subject to (32). An expression for $(\partial_z u_2)$ is obtained, which is then integrated in z . Imposing the boundary condition (29) gives the final expression for the horizontal velocity u_2 ; substituting it in equation (35) leads to:

$$\partial_t h_2 = \partial_x \left\{ \frac{1}{3} (h_2 - f(x))^3 \partial_x p_2 \right\}. \quad (39)$$

The pressure terms p_1 and p_2 are determined by solving the leading-order pressure equations (24) and (27), subject to the pressure balance and jump conditions at $z = h_1$

and $z = h_2$ given by (31) and (33) respectively, leading to:

$$p_1(x, t) = -\partial_{xx}h_2 - \frac{1}{\sigma}f''(x), \quad (40)$$

$$p_2(x, t) = -\partial_{xx}h_2 + \phi'(h_2 - f). \quad (41)$$

Equations (38)–(41) constitute the governing equations for two thin immiscible films, where the ratio of the upper layer viscosity to that of the lower layer is small, $O(\varepsilon^2)$. Since the lower layer flows much more slowly than the upper layer it is “frozen” to leading order in its initial prescribed configuration $f(x)$ (assumed known); (39) thus decouples and can be solved independently for $h_2(x, t)$. This solution can then be used to solve (38) for $h_{11}(x, t)$, the time-dependent $O(\varepsilon^2)$ perturbation to the lower layer, which allows us to determine $h_1(x, t)$ correct to this order. This procedure is valid until $\varepsilon^2 h_{11}$ approaches $O(1)$.

2.2 Existing models

A similar setup in which two immiscible Newtonian layers, each assumed to have *comparable* viscosities ($O(1)$ viscosity ratio), has been explored by Pototsky et al. [13], Fisher and Golovin [14] and Bandyopadhyay [15]. Each of these papers presents a set of coupled PDEs for the heights of the two fluid layers, and explores the properties of solutions for various choices of disjoining pressure and with scaling assumptions that differ from those we use. We focus on comparing predictions of our model with those of Pototsky et al. [13] and Fisher and Golovin [14] (which differ from each other only in their choice of disjoining pressure), and refer to these mathematical models as the PBMT and FG models respectively, in the remainder of the manuscript.

To make a meaningful comparison with our model, we modify the PBMT [13] and FG [14] models by using our choice of disjoining pressure, (21), and our scalings. PBMT and FG then reduce to the same system of nondimensional equations:

$$\partial_t h_1 = \varepsilon^2 M \partial_x \left\{ \frac{h_1^3}{3} \partial_x p_1 + \left(\frac{1}{2} h_1^2 (h_2 - h_1) \right) \partial_x p_2 \right\}, \quad (42)$$

$$\begin{aligned} \partial_t h_2 = \varepsilon^2 M \partial_x \left\{ \frac{1}{2} h_1^2 \left(h_2 - \frac{1}{3} h_1 \right) \partial_x p_1 \right\} + \partial_x \left\{ \left(\frac{1}{3} - \frac{\varepsilon^2 M}{2} \right) h_1^2 (h_2 - h_1) \partial_x p_2 \right\} \\ + \partial_x \left\{ \left(\varepsilon^2 M - \frac{2}{3} \right) h_1 h_2 (h_2 - h_1) \partial_x p_2 \right\} + \partial_x \left\{ \frac{1}{3} h_2^2 (h_2 - h_1) \partial_x p_2 \right\}, \end{aligned} \quad (43)$$

with the pressure terms p_1 and p_2 given by:

$$p_1 = -\frac{1}{\sigma} \partial_{xx} h_1 - \partial_{xx} h_2, \quad p_2 = -\partial_{xx} h_2 + \phi'(h_2 - h_1), \quad (44)$$

and the viscosity and surface tension ratios M, σ as defined in (18). Note the explicit presence of the small parameter $\varepsilon^2 \ll 1$ in the equations, which appears here as a consequence of the scaling assumption made on the small viscosity ratio of upper and lower layers. When we set $\varepsilon^2 = 0$ in (43) we recover our model at leading order. However, under the stated assumptions the explicit presence of these terms in the governing equation is asymptotically inconsistent, since comparable terms were discarded in the derivation. In the following we compare and contrast our simplified model with this modification of these earlier models, first using linear stability analysis and then via full numerical solutions.

3 Linear stability analysis

3.1 LSA for simplified model

We first perform linear stability analysis (LSA) on our simplified mathematical model given by Eqs. (38)–(41), assuming that both interfaces are nearly flat initially. We proceed as follows: the total film thickness $h_2(x) = h_2^0$ is perturbed with an oscillatory Fourier mode of amplitude $\delta\eta$ where $\delta \ll 1$ and the wave number k varies. The leading-order thickness of the lower layer is constant, $f(x) = f^0$, and we perturb h_{11} (the ε^2 term) with an oscillatory Fourier mode of amplitude $\delta\xi$. Substituting $h_{11}(x, t) = \delta\xi e^{ikx + \lambda t}$ and $h_2(x, t) = h_2^0 + \delta\eta e^{ikx + \lambda t}$ into (38)–(39), we obtain the following system of equations for η and ξ :

$$\lambda\xi + \eta M \left[\frac{1}{2}(f^0)^2 h_2^0 k^2 (k^2 + \phi'') - \frac{1}{2}(f^0)^3 k^2 \left(\frac{1}{3}k^2 + \phi'' \right) \right] = 0, \quad (45)$$

$$\eta \left[\lambda + \frac{1}{3}k^2 (h_2^0 - f^0)^3 (k^2 + \phi'') \right] = 0, \quad (46)$$

where ϕ'' is evaluated at $h_2^0 - f^0$, the leading-order value of the upper layer thickness. Solving the above system leads to the following dispersion relations for the growth rate λ as a function of wavenumber k :

$$\lambda_1^S(k) = 0, \quad \lambda_2^S(k) = -\frac{1}{3}k^2 (h_2^0 - f^0)^3 \left[k^2 + \phi''(h_2^0 - f^0) \right], \quad (47)$$

where the superscript S indicates that the dispersion relation equations correspond to the simplified model (38)–(41). Note that although the normal mode ansatz permits a complex growth rate in principle, the linearized system obtained by retaining terms up to $O(\delta)$ admits only real eigenvalues, and therefore the growth rates λ are real. The default parameter values used to illustrate the LSA results for both simplified and PBMT/FG models (discussed in the next section) are listed in Table 1.

From (47), we observe that the nontrivial dispersion relation depends on the initial thickness of the top layer, $h_2^0 - f^0$, and the disjoining pressure, but curiously, is independent of the dimensionless viscosity parameter M . To better understand the system's

Table 1 Default parameters used in the linear stability analysis of simplified model (38)–(41) and modified PBMT/FG model (42)–(44)

Default parameters used in Linear Stability Analysis (all models)		
M	1.0	Dimensionless viscosity parameter
σ	1.0	Dimensionless surface tension ratio
b	0.02	Dimensionless precursor layer thickness
ε	0.01	Film aspect ratio
r	1.05, 1.10, 1.15	Initial ratio of total bi-layer thickness to upper layer film thickness

behavior, we plot the dispersion relation curves fixing the initial total thickness of the two-layer system at $h_2^0 = 1$, for different cases of the individual layer thicknesses. Specifically, we introduce a new dimensionless parameter $r = h_2^0/f^0 > 1$ to represent the ratio between the total bi-layer thickness, h_2^0 , and the lower layer thickness, f^0 ; the closer r is to 1, the thinner the upper (less viscous) layer is relative to the lower layer.

Figure 3 illustrates the dispersion relation curves for three values of r : $r = 1.05$ (black), $r = 1.10$ (blue dashed) and $r = 1.15$ (red dash-dotted). As r increases the film rapidly becomes more stable. This is expected; a higher value of r indicates an increased initial thickness of the upper layer, thereby reducing the strength of the destabilizing interaction between the upper and lower layers. Additionally, we observe that both the wave number of maximum growth, $k_{\max}^S$, and its corresponding growth rate, $\lambda^S(k_{\max}^S)$, decrease strongly as r increases. Similarly, the critical wave number, k_{cr}^S , that marks the transition from unstable to stable behavior, also decreases as r increases, further emphasizing the system's increased stability when the upper film is thicker. We discuss these results further below when comparing with the stability results obtained from the earlier (more complicated) models. Before closing this section we comment briefly regarding the influence of the precursor film thickness, b . Consistent with earlier works including similar power-law forms of disjoining pressure (see, e.g., [29, 30]), larger b values lead to increased growth rates, as well as larger wavenumber of maximum growth, and the critical wavenumber. In practical situations, b (though always small) is a function of the particular liquid–liquid system studied, and varies from one system to another. A detailed study of its influence is beyond the scope of the present work.

3.2 LSA for the modified PBMT/FG model

We next perform linear stability analysis on the dimensionless model given by (42)–(44), which results from using our scalings and disjoining potential in the PBMT/FG models. Similarly to the analysis above, we perturb both films $h_1(x, t)$ and $h_2(x, t)$ with oscillatory Fourier modes, substituting $h_1(x, t) = f^0 + \delta\varepsilon^2\xi e^{ikx+\lambda t}$ and $h_2(x, t) = h_2^0 + \delta\eta e^{ikx+\lambda t}$ in (42)–(44) to obtain the following linear system for ξ, η :

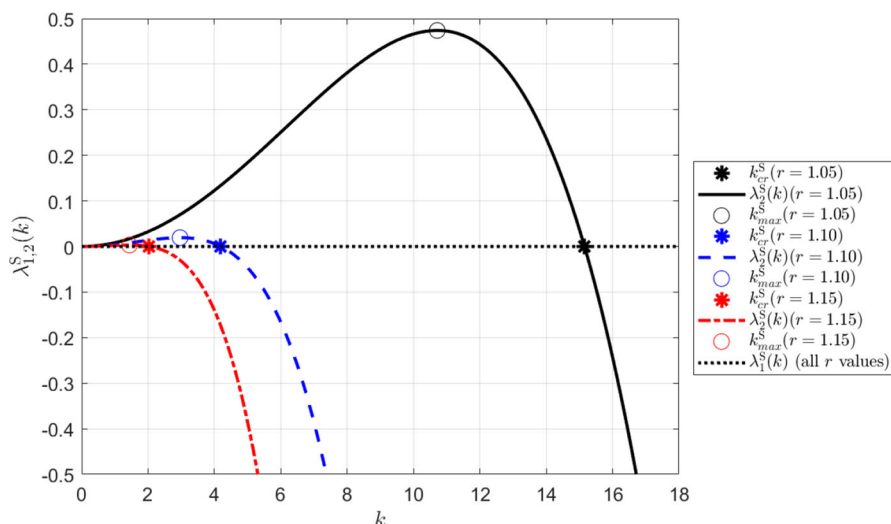


Fig. 3 Dispersion relation curves $\lambda_1^S(k) = 0$ (dotted black curve) and $\lambda_2^S(k) = -\frac{1}{3}k^2 \left(h_2^0 - f^0\right)^3 (k^2 + \phi'')$, for a two-layer film evolving according to the simplified model (38)–(41) of total thickness $h_2^0 = 1$ and a precursor layer thickness $b = 0.02$. The solid black curve represents the dispersion relation $\lambda_2^S(k)$ for $r = 1.05$; the dashed blue and dotted red curves represent the dispersion relation $\lambda_2^S(k)$ for $r = 1.10$ and 1.15 respectively. The stars represent the critical wave numbers denoted by k_{cr}^S , and the circles represent the wave numbers of maximum growth denoted by k_{max}^S , for each value of r considered here

$$\left[\varepsilon^2 \lambda + \frac{(f^0)^2 k^2 \varepsilon^4 M (2f^0 k^2 + 3(f^0 - h_2^0) \sigma \phi'')}{6\sigma} \right] \xi + \left[\frac{1}{6} (f^0)^2 k^2 \varepsilon^2 M [3h_2^0 (k^2 + \phi'') - f^0 (k^2 + 3\phi'')] \right] \eta = 0, \quad (48)$$

$$\left[\frac{1}{6\sigma} \left[(f^0)^2 [3h_2^0 - f^0] k^4 \varepsilon^4 M + (f^0 - h_2^0) k^2 \varepsilon^2 \left(2(f^0 - h_2^0)^2 - 3f^0 (f^0 - 2h_2^0) \varepsilon^2 M \right) \sigma \phi'' \right] \right] \xi + \quad (49)$$

$$\left[\frac{1}{3} k^4 \left(f^0 ((f^0)^2 - 3f^0 h_2^0 + 3h_2^0)^2 \varepsilon^2 M - (f^0 - h_2^0)^3 \right) + \lambda + \frac{1}{6} (f^0 - h_2^0) k^2 \left(3f^0 (f^0 - 2h_2^0) \varepsilon^2 M - 2(f^0 - h_2^0)^2 \right) \phi'' \right] \eta = 0.$$

The dispersion relation curves are obtained by solving the eigenvalue problem in (48)–(49). The corresponding (in)stability modes of the system are determined by the eigensolutions. One eigenvalue, $\lambda_1(k)$, is always negative, corresponding to a stable mode; therefore we focus primarily on the positive eigenvalue, $\lambda_2(k)$, as a function of wave number k . As we did for our simplified mathematical model, we plot the dispersion relation curves for scenarios in which the total initial unperturbed thickness of the bi-layer is $h_2^0 = 1$ while the unperturbed thickness of each individual layer (again represented by $r = h_2^0/f^0$) varies. Additionally, since (48)–(49) depend

explicitly on the dimensionless parameter $\varepsilon \ll 1$, we investigate how the stability regions depend on this parameter, in particular regarding convergence as $\varepsilon \rightarrow 0$.

Figure 4 depicts the dispersion relation curves corresponding to both positive and negative eigenvalues of the linearized system (48)–(49), for three different values of the layer thickness parameter r : as in Sect. 3.1 we choose $r = 1.05$ (Fig. 4a), $r = 1.10$ (Fig. 4b) and $r = 1.15$ (Fig. 4c). In each case we present results for different values of ε . As observed earlier, setting $\varepsilon = 0$ in the modified PBMT/FG models restores our simplified mathematical model, and thus the dispersion relation curves for $\varepsilon = 0$ are equivalent to those displayed in Fig. 3. Curiously, however, convergence is very slow: we have to take $\varepsilon = 10^{-4}$ before results are very close to those of the $\varepsilon = 0$ case. Another feature we observe is the nature of the convergence: the neutrally-stable eigenvalue ($\lambda_1^S(k) = 0$) of the simplified model analyzed in Sect. 3.1 emerges here as the union of two distinct eigenvalues: in the limit $\varepsilon \rightarrow 0$ we have $\lambda_1^S(k) \rightarrow \lambda_1(k)$ in $k < k_{\text{cr}}^S$, and $\lambda_1^S(k) \rightarrow \lambda_2(k)$ in $k > k_{\text{cr}}^S$. Similarly, for the unstable eigenvalue $\lambda_2^S(k)$; as $\varepsilon \rightarrow 0$ we have $\lambda_2^S(k) \rightarrow \lambda_2(k)$ in $k < k_{\text{cr}}^S$ and $\lambda_2^S(k) \rightarrow \lambda_1(k)$ in $k > k_{\text{cr}}^S$.

Regarding the system stability characteristics with respect to the thickness ratio parameter r , we observe that as r increases (the top, less viscous, layer becoming thicker), the rate of maximum growth, $\lambda(k_{\text{max}})$, decreases across all ε values. Secondly, we see that an increase in r leads to a decrease in the critical wave number k_{cr} for all ε values, indicating a greater stability of the two-layer system. Finally, we note another curiosity of the system: with r fixed, the critical wave number k_{cr} remains fixed for positive values of ε , attaining the critical wave number k_{cr}^S observed in the simplified mathematical model only at $\varepsilon = 0$ as observed in Fig. 4.

4 Numerical results

4.1 Numerical method and discretization scheme

The LSA discussed in the previous section provides insight into the behavior of the two mathematical models represented by (38)–(41) and (42)–(44) respectively, within the linear regime. To explore the effects of the nonlinear terms in each model's evolution, we solve the governing PDEs numerically using the method of lines, a numerical technique that is widely-used to solve nonlinear PDEs. Here, the spatial dimension is discretized using the central difference approximation on a uniform grid, resulting in a system of ordinary differential equations. These equations are then solved using MATLAB's built-in variable-step variable-order solver `ode15s` [31]. To simulate a two-layered film of infinite lateral extent, we impose periodic boundary conditions of the form:

$$\begin{aligned} h_i(0, t) &= h_i(L, t), & \partial_x h_i(0, t) &= \partial_x h_i(L, t), \\ \partial_{xx} h_i(0, t) &= \partial_{xx} h_i(L, t), & \partial_{xxx} h_i(0, t) &= \partial_{xxx} h_i(L, t), \end{aligned}$$

where $i = 1, 2$ in the respective fluid layers. In the following, we present results for the two models: the simplified model with simulations shown in Sect. 4.2; and the modified PBMT/FG model with numerical simulations discussed in Sect. 4.3. We

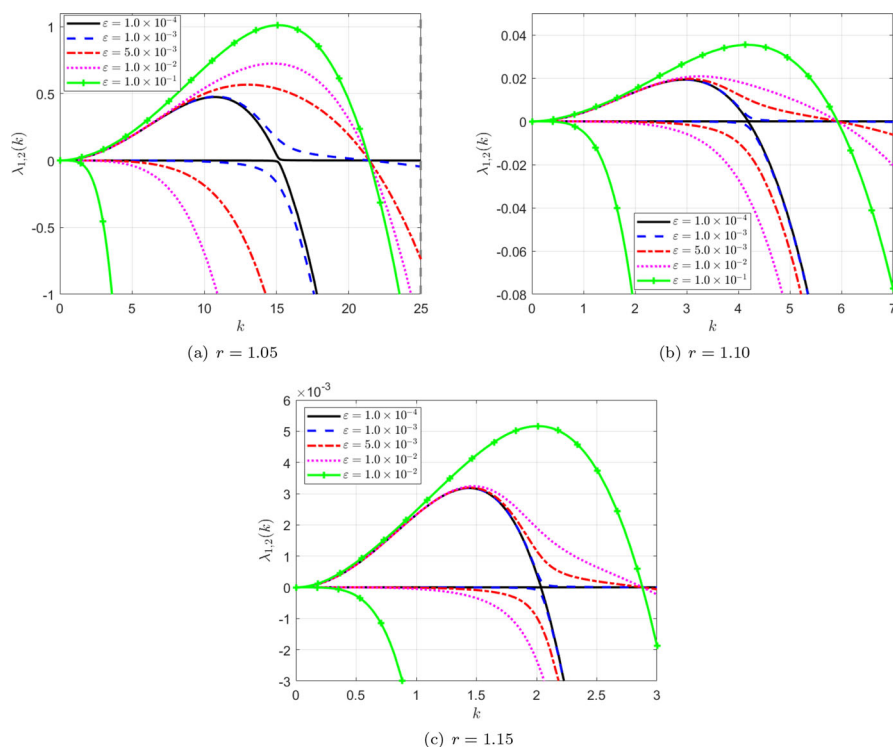


Fig. 4 Dispersion relation curves obtained by solving the eigenvalue system of (48)–(49) for different values of ε (specified in the legends) and **a** $r = 1.05$, **b** $r = 1.10$ and **c** $r = 1.15$

consider four different initial conditions: the first configuration consists of a flat lower layer of thickness $f(x) = 1/r$ with r taking values $r = 1.05$, 1.10 and $r = 1.15$ and an upper layer initialized as $h_2(x, 0) = 1 + a \cos(k_{\max}^S x)$ with $a = 0.001$ and $k_{\max}^S$ the wavelength of maximum growth determined by the LSA for the simplified model (when the value of $k_{\max}^S$ is not an integer, as in Figs. 3 and 4 for instance, the initial condition is constructed using the integer wavenumber closest to $k_{\max}^S$ so as to respect the periodic no-flux boundary conditions). The second and third cases explore in-phase and out-of-phase initial conditions, where $f(x) = 1/r \pm a \cos(k_{\max}^S x)$ and $h_2(x, 0) = 1 + a \cos(k_{\max}^S x)$. The fourth scenario examines initial conditions with different wavenumbers given by $f(x) = 1/r + a \cos(k_1 x)$ and $h_2(x, 0) = 1 + a \cos(k_2 x)$ where the wavenumbers k_1 and k_2 each take the values of $k_1 = 3$ and $k_2 = 6$ and vice-versa. The parameters used to simulate the four scenarios described above are displayed in Table 2.

Equations (38)–(39) for the simplified model form a decoupled system: at each time step, we solve (39) numerically, which determines the evolution of the top layer; then solve (38) to determine the lower layer's evolution. Conversely, (42)–(43) for the PBMT/FG model are fully coupled and therefore more challenging to solve. The same numerical approach can be applied, converting them into a system of ODEs that are

solved simultaneously using MATLAB's `ode15s` variable solver to determine the upper and lower layers' evolution at each time step.

4.2 Simplified model simulations

In this section, we explore the behavior of the system (38)–(41) in the nonlinear regime, via numerical simulations for the four scenarios described in Sect. 4.1. We start with the first scenario, where the bottom layer is initially flat, $f(x) = 1/r$, while the top layer is perturbed via $h_2(x, 0) = 1 + a \cos(k_{\max}^S x)$ where $a = 0.001$ and $k_{\max}^S$ is the wave number of maximum growth rate predicted by the LSA in Sect. 3.1. Analogous to that discussion of the LSA, we study the system behavior as the film thickness ratio parameter r is varied, choosing the same values, $r = 1.05$, $r = 1.10$ and $r = 1.15$. We display the evolution of each interface on a horizontal domain of length Λ , which is set to be double the wavelength of maximum growth, $\Lambda = 4\pi/k_{\max}^S$.

Figure 5 shows the evolution of each interface over time. As predicted by the LSA, systems with thicker upper films (larger values of r in our simulations) take longer to dewet. The dewetting time in our simulations is characterized by the dimensionless parameter $t_{\max}$, defined as the time at which the upper layer thickness, $h_2(x, t) - h_1(x, t)$, decreases to within 10% of the precursor layer thickness b for some x -value. As shown in Fig. 5 this threshold is achieved at $t_{\max} = 4.50$ when $r = 1.05$, $t_{\max} = 163.65$ when $r = 1.10$ and $t_{\max} = 1444.64$ when $r = 1.15$. While the simplified asymptotic model has the lower layer frozen to leading order in ε , our numerical simulations, with inclusion of the $O(\varepsilon^2)$ correction to the h_1 governing equation (see (36) and (38)), can describe the slow evolution of the lower layer to help dewet or “pinch off” the upper layer. When $r = 1.05$ (the thinnest upper layer considered, just 5% of the lower layer thickness, evolution displayed in Figure 5a), we observe that both layers evolve out-of-phase with one another (varicose instability mode) throughout the domain, with the less viscous upper layer deforming at a faster rate than the lower one and beginning to “pinch” into droplets. As time progresses this pinching evolution becomes more pronounced and we predict that dewetting would ultimately occur in the upper layer. As the initial thickness of the upper layer increases (see Fig. 5b for the simulation when $r = 1.10$), we observe a similar evolution: the upper layer evolves out-of-phase with the lower layer everywhere in the domain. These varicose deformations grow in size over the course of our simulation causing the film to dewet in the upper layer at $x \approx \pm 1$ (close to the prediction $\pm \pi/k_{\max}^S$ that one would expect based on LSA). We observe a similar behavior when $r = 1.15$, shown in Fig. 5c: the upper layer evolves out-of-phase with the lower layer throughout the domain, leading to dewetting and pinch-off in the upper layer at $x \approx \pm 3$ (see Fig. 5c; again this is close to the value expected from LSA). See `Animation_Fig_5a - Fig_5c.avi` in supplementary materials for animations of each evolution.

We now discuss the second scenario where both layers are initially perturbed with an in-phase perturbation of the form $f(x) = 1/r + a \cos(k_{\max}^S x)$ and $h_2(x, 0) = 1 + a \cos(k_{\max}^S x)$, again with $a = 0.001$ and $k_{\max}^S$ corresponding to the wave number of maximum growth as predicted by LSA in Sect. 3.1. As in the previous simulation, we vary the thickness of the upper layer by setting $r = 1.05$, $r = 1.10$ and

Table 2 Default parameters used in the numerical results of simplified model (38)–(41) and PBMT/FG model (42)–(44)

Default parameters used in numerical simulations			
	Simplified model	PBMT/FG model	Description
M	1.0	1.0	Dimensionless viscosity parameter
σ	1.0	1.0	Dimensionless surface tension ratio
b	0.02	0.02	Dimensionless precursor layer thickness
ε	0.01	0.01	Film aspect ratio
r	1.05, 1.10, 1.15	1.05, 1.10, 1.15	Ratio of initial configurations of upper and lower layers in both models
a	0.001	0.001	Amplitude of initial perturbation of lower and upper layer
$k_{\max}^S$	1.0, 3.0, 11.0	1.0, 3.0, 11.0	Wave number of maximum growth as established by LSA of simplified model
$h_2(x, 0)$	$1 + a \cos(k_{\max}x)$	$1 + a \cos(k_{\max}x)$	Perturbed configuration of upper layer in both models
$f(x)$	$1/r$	N/A	Flat configuration of lower layer in simplified model
	$1/r + a \cos(k_{\max}x)$	N/A	In-phase configuration of lower layer in simplified model
	$1/r - a \cos(k_{\max}x)$	N/A	Out-of-phase configuration of lower layer in simplified model
$h_1(x, 0)$	N/A	$1/r$	Flat configuration of lower layer in PBMT/FG model

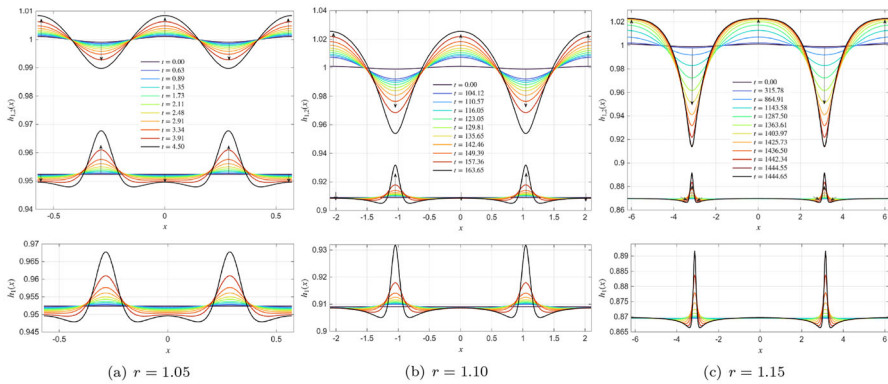


Fig. 5 Simplified Model: Evolution of a two layer film with a flat bottom layer of height $f(x) = 1/r$, a perturbed top layer of total height $h_2(x, 0) = 1 + 0.001 \cos(k_{\max}^S x)$, with $\varepsilon = 0.01$ for **a** $r = 1.05$, $k_{\max}^S = 11$ and $t_{\max} = 4.50$, **b** $r = 1.10$, $k_{\max}^S = 3$ and $t_{\max} = 163.65$ and **c** $r = 1.15$, $k_{\max}^S = 1$ and $t_{\max} = 1444.64$. Arrows indicate the evolution of each film over time. The second row figures illustrate a closeup of the bottom layer evolution

$r = 1.15$. When $r = 1.05$ (shown in Fig. 6a), both layers, initially in-phase, evolve rapidly to be out-of-phase with one another, with the upper layer inverting its initial configuration since it deforms faster than the lower layer. Over time, the lower layer develops stronger varicose deformations at $x = 0$ and $x \approx 0.55$, which continue to grow until the end of the simulation, aiding the dewetting. Figures 6b ($r = 1.10$) and 6c ($r = 1.15$) show a similar evolution in which an initially in-phase two layer system begins to evolve out-of-phase, with the upper layer inverting its initial perturbations while deforming faster than the lower one. As time progresses, the lower layer develops pronounced peaks around $x = 0, \pm 2$ when $r = 1.10$ and $x = 0, \pm 6$ when $r = 1.15$. These varicose deformations continue to grow (and presumably would ultimately lead to pinch-off of the upper layer, though such evolution would take us beyond the limits of our model assumptions) until our simulations are stopped. See Animation_Fig_6a - Fig_6c.avi in Supplementary materials for animations of each evolution.

The simulations of Figs. 5 and 6 suggest that the varicose state is strongly preferred under the considered conditions. Consequently, when the system is initialized in a varicose configuration, the evolution closely mirrors that of Figs. 5 and 6, but evolves at a faster rate. See supplementary materials for simulations of a system initially in a varicose configuration.

We now turn our attention to a scenario where both interfaces are perturbed with different wave numbers. We consider a system with moderate upper layer thickness (i.e., $r = 1.10$). Figure 7a shows the evolution of the two-layer system when the top layer is initially perturbed with a wave number $k = 3$ while the bottom layer is perturbed with a wave number $k = 6$. We observe that the upper layer evolves to form peaks around $x = 0, \pm 2$ and valleys around $x = \pm 1$. The lower layer starts to evolve so as to flatten the peaks initially present around $x = 0, \pm 2$ while forming pronounced peaks around $x = \pm 1$. Thus, over time, the layers again evolve to a

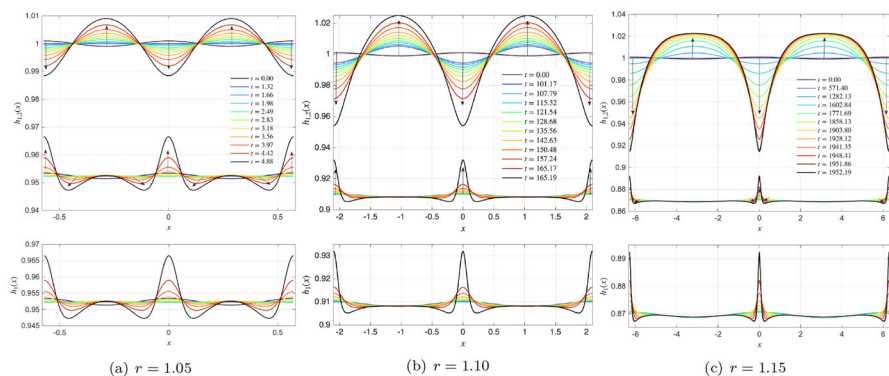


Fig. 6 Simplified Model: Evolution of a two layer film with an in-phase perturbed bottom interface of the form $f(x) = 1/r + 0.001 \cos(k_{\max}^S x)$, a perturbed top interface of similar form: $h_2(x, 0) = 1 + 0.001 \cos(k_{\max}^S x)$, with $\varepsilon = 0.01$ for **a** $r = 1.05$, $k_{\max}^S = 11$ and $t_{\max} = 4.88$, **b** $r = 1.10$, $k_{\max}^S = 3$ and $t_{\max} = 165.19$ and **c** $r = 1.15$, $k_{\max}^S = 1$ and $t_{\max} = 1952.19$. Arrows indicate the evolution of each film over time. The second row figures illustrate a closeup of the bottom layer evolution

varicose instability, with evolution dominated by the wavelength of maximum growth predicted by LSA (the perturbation wavelength that was applied to the top interface), leading to near-dewetting of the upper layer via pinch-off near $x = \pm 1$. As in the previous simulations, we stop the evolution at $t = t_{\max}$ when the thickness of the upper layer reaches 10% of the precursor thickness.

Figure 7b 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 this scenario, we again observe the system to evolve to a varicose instability, determined by the predicted wavelength of maximum growth, $k = 3$: the peaks initially present in the upper layer around $x = 0, \pm 2$ rapidly flatten, then become minima, continuing to evolve so as to decrease the thickness of the upper layer at those points. Meanwhile, the lower layer remains largely stationary initially. As time progresses, again dewetting/pinch-off is approached in the upper layer (but more slowly than in the prior simulation), around $x = 0, \pm 2$.

4.3 PBMT/FG model simulations

We now present the numerical results obtained by solving the modified PBMT/FG model described by (42)–(43) for a two-layer system in which the bottom layer is initially flat, and the top layer is defined as $h_2(x, 0) = 1 + a \cos(k_{\max}^S x)$, with $a = 0.001$ and $k_{\max}^S$ the wavenumber of maximum growth as predicted by LSA of the simplified model. We discuss the similarities and differences in the evolution of this system as compared with the analogous numerical solution of the simplified model (36)–(41) shown in Fig. 5. Results are again presented for the same three values of the initial layer height ratio parameter, r .

Figure 8a illustrates the evolution of the two-layer film for $r = 1.05$, the thinnest upper layer. Consistent with the predictions of our simplified mathematical model, the

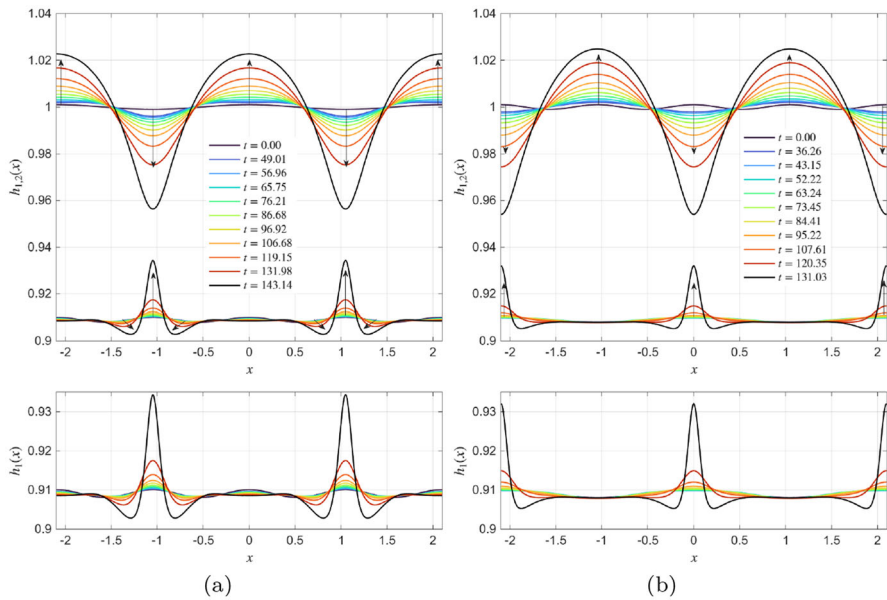


Fig. 7 Simplified Model: **a** Evolution of a two layer film with a perturbed bottom layer of the form $f(x) = 1/r + 0.001 \cos(6x)$, $r = 1.10$ and a perturbed top layer $h_2(x, 0) = 1 + 0.001 \cos(3x)$. **b** Evolution of a two layer film with a perturbed bottom layer of the form $f(x) = 1/r + 0.001 \cos(3x)$, $\varepsilon = 0.01$, $r = 1.10$ and a perturbed top layer $h_2(x, 0) = 1 + 0.001 \cos(6x)$. Arrows indicate the evolution of each film over time. The second row figures illustrate a closeup of the bottom layer evolution

layers evolve out-of-phase, leading to a thinning upper layer over time. This evolution occurs on a comparable timescale in both models: $t_{\max} = 4.30$ dimensionless time units for the modified PBMT/FG model and $t_{\max} = 4.50$ time units for our mathematical model (as shown in Fig. 5a). A notable quantitative difference between the models lies in the extent of the movement of the two interfaces during the evolution: our model predicts somewhat greater movement of the lower interface than does the PBMT/FG model, while the PBMT/FG model exhibits more movement of the upper interface than does the simplified model. Additionally, the peaks in the lower layer near $x = \pm 0.3$ are less prominent in the PBMT/FG model compared to our mathematical model, suggesting that our model perhaps slightly overpredicts the dynamics of the lower interface in this particular scenario.

The corresponding evolution of the modified PBMT/FG model when $r = 1.10$ and $r = 1.15$ is shown in Fig. 8b and c respectively. The evolution is very similar to that predicted by our simplified mathematical model (shown in Figs. 5b, c): both layers evolve out-of-phase with one another (varicose mode), with the top film approaching pinch-off close to $x = \pm 1$ ($r = 1.10$) and $x = \pm 3$ ($r = 1.15$). The simulation is halted at $t = t_{\max}$ when the thickness of the top layer reaches within 10% of the precursor thickness $b = 0.02$, occurring on a timescale similar to that predicted by the simplified model. Specifically, $t_{\max} = 151.3$ when $r = 1.10$ and $t_{\max} = 1410$ when $r = 1.15$, which are comparable to the values $t_{\max} = 164$ and $t_{\max} = 1445$ from the simulations in Fig. 5b, c. Similar to the qualitative observation for $r = 1.05$, our

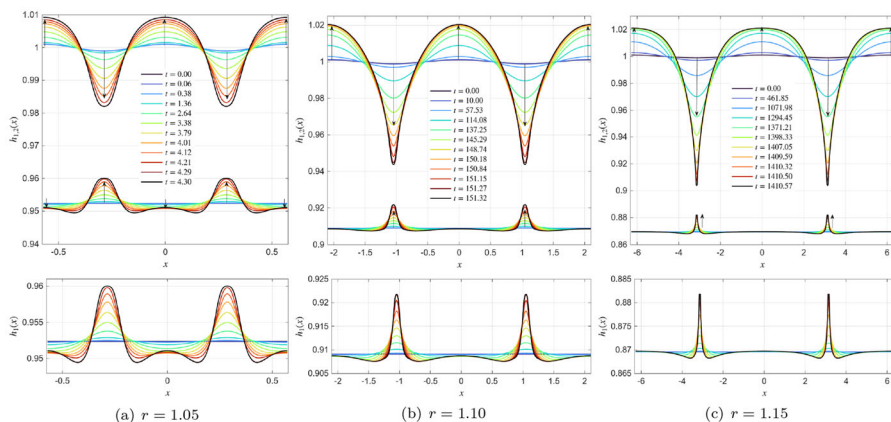


Fig. 8 PBMT/FG Model: Evolution of a two layer film with a flat bottom layer of height $h_1(x, 0) = 1/r$, a perturbed top layer of $h_2(x, 0) = 1 + 0.001 \cos(k_{\max}^S x)$ with $\varepsilon = 0.01$ for **a** $r = 1.05$, $k_{\max}^S = 11$ and $t_{\max} = 4.30$, **b** $r = 1.10$, $k_{\max}^S = 3$ and $t_{\max} = 151.32$ and **c** $r = 1.15$, $k_{\max}^S = 1$ and $t_{\max} = 1410.57$. Arrows indicate the evolution of each film over time. The second row figures illustrate a closeup evolution of the bottom layer

simplified model predicts slightly greater movement in the lower interface and slightly less movement in the upper one, compared with the PBMT/FG model. In making these comparisons, however, note that we use the same domain length and initial condition as we do for our simplified model, rather than using domain length and initial condition based on predictions of the PBMT/FG LSA. Since the domain length is fixed and wavenumbers are quantized by the imposed boundary conditions, the film may be forced into an instability that is not exactly aligned with the LSA predictions. Hence, the location of the lower/upper interface minima/maxima may not correspond exactly to the LSA estimate of $x \approx \pi/k_{\max}$. We note that comparisons with the PBMT/FG model were presented only for the flat case, since the in-phase (Fig. 6), out-of-phase (Supplementary Fig. S1), and varying wave number simulations (Fig. 7) exhibited the same qualitative and quantitative trends. In all cases, both models predict out-of-phase evolution of the interfaces and thinning of the upper layer on comparable timescales. The main difference is that the present model predicts slightly larger motion of the lower interface, whereas the PBMT/FG model exhibits greater displacement of the upper interface, with less pronounced lower layer peaks.

5 Conclusions

In this work, we present a simplified mathematical model that describes the evolution of a two-layer film with comparable surface tensions but small viscosity contrast. The lower film is assumed to be much more viscous than the upper film, causing the lower film to flow at a much slower rate. Such a setup provides a possible paradigm for a fluid flowing on a soft deformable substrate. Our model consists of a decoupled system of two partial differential equations (PDEs), describing the evolution of the upper and

lower layers respectively. The decoupling offers computational advantages compared to existing models in the literature [13–15], as well as being asymptotically-consistent for the specific viscosity ratio considered. We focus on scenarios where the surface forces arising from fluid 1-substrate and fluid 2-substrate interactions are negligible, and only surface forces arising from interactions between the upper and lower layers can lead to dewetting.

We use both analytical and numerical techniques to study the evolution of the two-layered system, focusing on conditions that lead to dewetting. Linear stability analysis (LSA) is performed by perturbing both interfaces with small oscillatory Fourier modes of the same wavelength. Our analysis shows that the initial thickness of the upper layer and the disjoining pressure influence the system's stability: a thicker upper layer increases the stability of the system. A similar LSA investigation is conducted on the modified version of the PBMT/FG model, with the same disjoining pressure as our model. The results are very similar to those for our model, confirming that systems with thicker upper layers dewet much more slowly when compared to systems with thinner upper layers. We note that the dispersion relation for the modified PBMT/FG model depends on the thickness of the upper layer, disjoining pressure term, and the viscosity ratio, while the dispersion relation of our model depends only on the initial thickness of the upper layer and the disjoining pressure.

Numerical simulations were used to investigate the nonlinear behavior of both our simplified mathematical model and the modified PBMT/FG model. For our model, we analyze systems with an initially flat bottom layer and a perturbed top layer, as well as scenarios with initially in-phase perturbed layer interfaces. Our numerical results confirm that a system with an initially thicker upper layer takes longer to dewet. In all cases investigated, even when the initial perturbation is in-phase, dewetting occurs through the growth of out-of-phase deformations (varicose instability mode), leading to thinning of the upper layer in specific regions of the domain. The modified PBMT/FG model exhibits very similar dynamics to our model. Both models predict out-of-phase (varicose) evolution of the upper layer, leading to local thinning and dewetting/pinch-off of the upper layer on comparable timescales. However, the dynamics differ quantitatively: our model shows greater movement in the lower layer, whereas the PBMT/FG model exhibits more activity in the upper layer.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s10665-026-10536-0>.

Acknowledgements E.M. gratefully acknowledges the support of Kean University through its Released Time for Research (RTR) Program, which provided the necessary time and support for the completion of this study. We acknowledge discussions with Jacco Snoeijer and Uwe Thiele, which motivated initial stages of this project.

Author contributions EM, LK, and LJC contributed equally to the conceptualization, methodology, formal analysis, implementation of numerical methods, and the writing and revision of the manuscript. EM implemented the numerical methods.

Data availability The datasets generated during this study are available upon request.

Declarations

Competing interests The authors declare no competing interests.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

- Weinstein SJ, Ruschak KJ (2004) Coating flows. *Annu Rev Fluid Mech* 36:29–53
- Grothberg JB (1994) Pulmonary flow and transport phenomena. *Annu Rev Fluid Mech* 26:529–571
- Ancey C (2007) Plasticity and geophysical flows: a review. *J Non-Newton Fluid Mech* 142(1):4–35
- Schoof C (2011) Marine ice sheet dynamics. Part 2. A Stokes flow contact problem. *J Fluid Mech* 679:122, 155
- Craster RV, Matar OK (2009) Dynamics and stability of thin liquid films. *Rev Mod Phys* 81:1131–1198
- Stewart PS, Feng J, Kimpton LS, Griffiths IM, Stone A (2015) Howard, Stability of a bi-layer free film: simultaneous or individual rupture events? *J Fluid Mech* 777(27):49
- Sahoo S, Arora A, Doshi P (2016) Two-layer spin coating flow of Newtonian liquids: a computational study. *Comput Fluids* 131:180–189
- Kao TW (1964) Stability of two-layer viscous stratified flow down an inclined plane. *Phys Fluids* 7:1658–1668
- Kao TW (1968) Role of viscosity stratification in the stability of two-layer flow down an incline. *J Fluid Mech* 33:453–465
- Tilley BS, Davis SH, Bankoff SG (1994) Linear stability theory of two-layer fluid flow in an inclined channel. *IMA J Appl Math* 54:245–265
- Bhat A, Samanta R (2020) Linear stability of surfactant-laden two-layer film flows down a slippery inclined plane. *Chem Eng Sci* 222:115646
- Chattopadhyay S, Mukhopadhyay A, Barua A (2019) A review on hydrodynamical stability of thin film flowing along an inclined plane. *J Math Sci Model* 2(2):133–142
- Pototsky A, Bestehorn M, Merkt D, Thiele U (2004) Alternative pathways of dewetting for a thin liquid two-layer film. *Phys Rev E* 70:025201
- Fisher L, Golovin A (2005) Nonlinear stability analysis of a two-layer thin liquid film: Dewetting and autophobic behavior. *J Colloid Interface Sci* 291(2):515–528
- Bandyopadhyay D, Gulabani R, Sharma A (2005) Instability and dynamics of thin liquid bilayers. *Ind Eng Chem Res* 44(5):1259–1272
- Pototsky A, Bestehorn M, Merkt D, Thiele U (2005) Morphology changes in the evolution of liquid two-layer films. *J Chem Phys* 122(22):224711
- Sollogoub C et al (2016) Kinetics of thin polymer film rupture: Model experiments for a better understanding of layer breakups in the multilayer coextrusion process. *Polymer* 96:152–160
- Miquelard-Garnier G et al (2023) Fabrication of architected multilayers with mismatched rheological behaviors. *Ind Eng Chem Res* 62(12):4970–4980
- Dmochowska A, Peixinho J, Sollogoub C, Miquelard-Garnier G (2023) Extensional viscosity of immiscible polymer multi-nanolayer films. *Macromolecules* 56(10):4015–4025
- Norrman K, Ghanbari-Siahkali A, Larsen NB (2005) Studies of spin-coated polymer films. *Annu Rep Sect C Phys Chem* 101:174–201
- Amruth C, Colella M, Griffin J, Kingsley J, Scarratt N, Luszczynska B, Ulanski J (2019) Slot?die coating of double polymer layers for the fabrication of organic light emitting diodes. *Micromachines* 10(1):53

22. Jachalski S, Münch AM, Wagner B (2015) Thin-film models for viscoelastic liquid bi-layers. WIAS-Preprint 2187
23. Jachalski S, Peschka D, Münch A, Wagner B (2014) Impact of interfacial slip on the stability of liquid two-layer polymer films. *J Eng Math* 86:9–29
24. van Gorcum M, Karpitschka S, Andreotti B, Snoeijer JH (2020) Spreading on viscoelastic solids: are contact angles selected by Neumann's law? *Soft Matter* 16:1306–1322
25. Henkel C, Snoeijer JH, Thiele U (2021) Gradient-dynamics model for liquid drops on elastic substrates. *Soft Matter* 17:10359–10375
26. Style RW, Jagota A, Hui C-Y, Dufresne ER (2017) Elastocapillarity: Surface tension and the mechanics of soft solids. *Annu Rev Condens Matter Phys* 8(8):99–118
27. Tamim S, Bostwick JB (2023) Spreading of a thin droplet on a soft substrate. *J Fluid Mech* 971:A32
28. Bommer S, Seemann R, Jachalski S, Peschka D, Wagner B (2016) Liquid–liquid dewetting: morphologies and rates, Preprint 2346. Weierstra-Institut für Angewandte Analysis und Stochastik, Berlin
29. Diez JA, Kondic L (2007) On the breakup of fluid films of finite and infinite extent. *Phys Fluids* 19:072107
30. González A, Diez JA, Wu Y, Fowlkes JD, Rack PD, Kondic L (2013) Instability of liquid Cu films on a SiO₂ substrate. *Langmuir* 29:9378
31. Shampine LF, Reichelt MW (1997) The Matlab ODE suite. *SIAM J Sci Comput* 18(1):1–22

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.