Nonlinear dynamics of phase separation in ultra-thin films

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We present a long-wavelength approximation to the Navier–Stokes Cahn–Hilliard equations to describe phase separation in thin films. The equations we derive underscore the coupled behaviour of free-surface variations and phase separation. Since we are interested in the long-time behaviour of the phase-separating fluid, we restrict our attention to films that do not rupture. To do this, we introduce a regularising Van der Waals potential. We analyse the resulting fourth-order equations by constructing a solution as the limit of a Galerkin approximation, and obtain existence and regularity results. In our analysis, we find a nonzero lower bound for the height of the film, which precludes the possibility of rupture. The lower bound depends on the parameters of the problem, and we compare this dependence with numerical simulations. We find that while the theoretical lower bound is crucial to the construction of a smooth, unique solution to the PDEs, it is not sufficiently sharp to represent accurately the parametric dependence of the observed dips in free-surface height.

I. INTRODUCTION

Below a certain critical temperature, a well-mixed binary fluid spontaneously separates into its component parts, forming domains of pure liquid. This process can be characterised by the Cahn–Hilliard equation, and numerous studies describe the physics and mathematics of phase separation. In this paper we study phase separation in a thin layer, in which the varying free surface and concentration fields are coupled through a pair of nonlinear evolution equations.

Cahn and Hilliard introduced their eponymous equation in [8] to model phase separation in a binary alloy. Since then, the model has been used in diverse appli-

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cations: to describe polymeric fluids [1], fluids with interfacial tension [21], and self-segregating populations in biology [10]. Analysis of the Cahn–Hilliard (CH) equation was given by Elliott and Zheng in [14], where they prove the existence, uniqueness, and regularity of solutions: given sufficiently smooth initial data, solutions to the CH equation are bounded for almost all time in the Sobolev space $W^{4,2}$. Several authors have developed generalisations of the CH equation: a variable-mobility model was introduced by Elliott and Garcke [13], while nonlocal effects were considered by Gajweski and Zacharias [15]. These additional features do not qualitatively change the phase separation, and we therefore turn to one mechanism that does: the coupling of a flow field to the Cahn–Hilliard equation [7]. In this case, the Cahn–Hilliard concentration equation is modified by an advection term, and the flow field is either prescribed or evolves according to some fluid equation. Ding and co-workers [11] provide a derivation of coupled Navier–Stokes Cahn–Hilliard (NSCH) equations in which the velocity advects the phase-separating concentration field, while concentration gradients modify the velocity through an additional stress term in the momentum equation. Such models have formed the basis of numerical studies of binary fluids [4], while other studies without this feedback term highlight different regimes of phase separation under flow [3, 19, 24]. In this paper, the NSCH equations form the starting point for our asymptotic analysis.

As in other applications involving the Navier–Stokes equations, the complexity of the problem is reduced when the fluid is spread thinly on a substrate, and the upper vertical boundary forms a free surface [26]. Then, provided vertical gradients are small compared to lateral gradients, a long-wavelength approximation is possible, in which the full equations with a moving boundary at the free surface are reduced to a single equation for the free-surface height. In the present case, the reduction yields two equations: one for the free surface, and one for the Cahn–Hilliard concentration. The resulting thin-film Stokes Cahn–Hilliard equations have already been introduced by the authors in [25], although the focus there was on control of phase separation and numerical simulations in three dimensions. Here we confine ourselves to the two-dimensional case: we derive the thin-film equations from first principles, present analysis of the resulting equations, and highlight the impossibility of film rupture.

Along with the simplification of the problem that thin-film theory provides, there are many practical reasons for studying phase separation in thin layers. Thin polymer films are used in the fabrication of semiconductor devices, for which detailed knowledge of film morphology is required [17]. Other industrial applications of polymer films include paints and coatings, which are typically mixtures of polymers. One potential application of the thin-film Cahn–Hilliard theory is in self-assembly [18, 22, 33]. Here molecules (usually residing in a thin layer) respond to an energy-minimisation requirement by spontaneously forming large-scale structures. Equations of Cahn–Hilliard type have been proposed to explain the qualita-
tive features of self-assembly [16, 22], and knowledge of variations in the film height could enhance these models. Indeed in [25] the authors use the present thin-film Cahn–Hilliard model in three dimensions to control phase separation, a useful tool in applications where it is necessary for the molecules in the film to form a given structure.

The analysis of thin-film equations was given great impetus by Bernis and Friedman in [2]. They focus on the basic thin-film equation,

\[
\frac{\partial h}{\partial t} = -\frac{\partial}{\partial x} \left( h^n \frac{\partial^3 h}{\partial x^3} \right),
\]

with no-flux boundary conditions on a line segment, and smooth nonnegative initial conditions. For \( n = 1 \) this equation describes a thin bridge between two masses of fluid in a Hele–Shaw cell, for \( n < 3 \) it is used in slip models as \( h \to 0 \) [23], while for \( n = 3 \) it gives the evolution of the free surface of a thin film experiencing capillary forces [26]. Using a decaying free-energy functional, they prove the existence of nonnegative solutions to Eq. (1) for \( n \geq 1 \), while for \( n \geq 4 \), and for strictly positive initial conditions, the solution is unique, strictly positive, and is almost always bounded in the \( W^{3,2} \) norm. This paper has inspired other work on the subject [5, 6, 20], in which the effect of a Van der Waals term on Eq. (1) is investigated. These works provide results concerning regularity, long-time behaviour, and film rupture in the presence of an attractive Van der Waals force. More relevant to the present work is the paper by Wieland and Garcke [32], in which a pair of partial differential equations describes the coupled evolution of free-surface variations and surfactant concentration. The authors derive the relevant equations using the long-wavelength theory, obtain a decaying energy functional, and prove results concerning the existence and nonnegativity of solutions. We shall take a similar approach in this paper.

When the binary fluid forms a thin film on a substrate, we shall show in Sec. II that a long-wave approximation simplifies the Navier–Stokes Cahn–Hilliard equations, which reduce to a pair of coupled evolution equations for the free surface and concentration. If \( h(x, t) \) is the scaled free-surface height, and \( c(x, t) \) is the binary fluid concentration, then the dimensionless equations take the form

\[
\frac{\partial h}{\partial t} + \frac{\partial J}{\partial x} = 0, \quad \frac{\partial}{\partial t} (hc) + \frac{\partial}{\partial x} (Jc) = \frac{\partial}{\partial x} \left( h \frac{\partial \mu}{\partial x} \right),
\]

where

\[
J = \frac{1}{2} h^2 \frac{\partial \sigma}{\partial x} - \frac{1}{3} h^3 \left\{ \frac{\partial}{\partial x} \left( -\frac{1}{C} \frac{\partial^2 h}{\partial x^2} + \phi \right) + \frac{r}{h} \frac{\partial}{\partial x} \left[ h \left( \frac{\partial c}{\partial x} \right)^2 \right] \right\},
\]

\[
\mu = c^3 - c - \frac{c^2}{h} \frac{\partial}{\partial x} \left( h \frac{\partial c}{\partial x} \right).
\]
Here $C$ is the capillary number, $r$ measures the strength of coupling between the concentration and free-surface variations (backreaction), and $C_n$ is the scaled interfacial thickness. Additionally, $\sigma$ is the dimensionless, spatially-varying surface tension, and $\phi$ is the body-force potential acting on the film. In this paper we take $\phi = -|A|h^{-3}$, the repulsive Van der Waals potential [27]. This choice stabilises the film and prevents rupture. Although rupture is in itself an important feature in thin-film equations [5, 6, 26], in this paper we are interested in late-time phase separation and it is therefore undesirable.

We present the asymptotic analysis that converts the NSCH equations into Eq. (2) and prove that the model equations possess smooth solutions that are bounded for almost all time in the $W^{1,2}$ norm. The principal tool in this analysis is the construction of a free-energy functional for Eq. (2) that is a decaying function of time. We prove that $h(x,t) > 0$ for all time, which is the no-rupture condition.

The paper is organised as follows. In Sec. II we discuss the Navier–Stokes Cahn–Hilliard equation and the scaling laws that facilitate the passage to the long-wavelength equations, and we derive Eq. (2). In Sec. III we analyse these equations by constructing a decaying free-energy functional. We prove the existence of solutions to Eq. (2) and provide regularity results. We obtain a condition on the minimum value of the free-surface height, and show that this is never zero. Using numerical studies, we discuss the dependence of the minimum free-surface height on the problem parameters in Sec. IV and compare with the analytic results. Finally, in Sec. V we present our conclusions.

II. THE MODEL EQUATIONS

In this section we introduce the two-dimensional Navier–Stokes Cahn–Hilliard (NSCH) equation set. We discuss the assumptions underlying the long-wavelength approximation. We enumerate the scaling rules necessary to obtain the simplified equations. Finally, we arrive at a set of equations that describe phase separation in a thin film subject to arbitrary body forces.

The full NSCH equations describe the coupled effects of phase separation and flow in a binary fluid. If $\mathbf{v}$ is the fluid velocity and $c$ is the concentration of the mixture, where $c = \pm 1$ indicates total segregation, then these fields evolve as

$$\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} = \nabla \cdot T - \frac{1}{\rho} \nabla \phi,$$  \hspace{1cm} (3a)

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = D \nabla^2 \left( c^3 - c - \gamma \nabla^2 c \right),$$  \hspace{1cm} (3b)

$$\nabla \cdot \mathbf{v} = 0,$$  \hspace{1cm} (3c)
\[ T_{ij} = -\frac{p}{\rho} \delta_{ij} + \nu \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right) - \beta \gamma \frac{\partial c}{\partial x_i} \frac{\partial c}{\partial x_j} \]  

(4)

is the stress tensor, \( p \) is the fluid pressure, \( \phi \) is the body potential and \( \rho \) is the constant density. The constant \( \nu \) is the kinematic viscosity, \( \nu = \eta / \rho \), where \( \eta \) is the dynamic viscosity. Additionally, \( \beta \) is a constant with units of \([\text{Energy}][\text{Mass}]^{-1}\), \( \sqrt{\gamma} \) is a constant that gives the typical width of interdomain transitions, and \( D \) a diffusion coefficient with dimensions \([\text{Length}]^2[\text{Time}]^{-1}\).

If the system has a free surface in the vertical or \( z \)-direction and has infinite or periodic boundary conditions (BCs) in the lateral or \( x \)-direction, then the vertical BCs we impose are

\[ u = w = c_z = c_{zzz} \text{ on } z = 0, \]  

(5a)

while on the free surface \( z = h(x,t) \) they are

\[ \hat{n}_i \hat{n}_j T_{ij} = -\sigma \kappa, \quad \hat{n}_i \hat{n}_j T_{ij} = -\frac{\partial \sigma}{\partial s}, \]  

(5b)

\[ w = \frac{\partial h}{\partial t} + u \frac{\partial h}{\partial x}, \]  

(5c)

\[ \hat{n}_i \partial_i c = 0, \quad \hat{n}_i \partial_i \nabla^2 c = 0, \]  

(5d)

where \( \hat{n} = (-\partial_x h, 1)/[1 + (\partial_x h)^2]^{1/2} \) is the unit normal to the surface, \( \hat{t} \), is the unit vector tangent to the surface, \( s \) is the surface coordinate, \( \sigma \) is the surface tension, and \( \kappa \) is the mean curvature,

\[ \kappa = \nabla \cdot \hat{n} = \frac{\partial_{xx} h}{\left[1 + (\partial_x h)^2\right]^{3/2}}. \]

This choice of BCs guarantees the conservation of the total mass and volume,

\[ \text{Mass} = \int_{\text{Dom}(t)} c(x,t) d^2x, \quad \text{Volume} = \int_{\text{Dom}(t)} d^2x. \]  

(6)

Here \( \text{Dom}(t) \) represents the time-dependent domain of integration, owing to the variability of the free surface height. Note that in view of the concentration BC (5d), the stress BC (5b) and does not contain \( c(x,t) \) or its derivatives.

These equations simplify considerably if the fluid forms a thin layer of mean thickness \( h_0 \), for then the scale of lateral variations \( \lambda \) is large compared with the scale of vertical variations \( h_0 \). Specifically, the parameter \( \delta = h_0 / \lambda \) is small, and after nondimensionalisation of Eq. (3) we expand its solution in terms of this parameter,
keeping only the lowest-order terms. For a review of this method and its applications, see [26]. For simplicity, we shall work in two dimensions, but the generalisation to three dimensions is easily effected [25].

In terms of the small parameter $\delta$, the equations nondimensionalise as follows. The diffusion time scale is $t_0 = \lambda^2/D = h_0^2/(\delta^2 D)$ and we choose this to be the unit of time. Then the unit of horizontal velocity is $u_0 = \lambda/t_0 = \delta D/h_0$ so that $u = (\delta D/h_0) U$, where variables in upper case denote dimensionless quantities. Similarly the vertical velocity is $w = (\delta D/h_0) W$. For the equations of motion to be half-Poiseuille at $O(1)$ (in the absence of the backreaction) we choose $p = (\eta D/h_0^2) P$ and $\phi = (\eta D/h_0^2) \Phi$. Using these scaling rules, the dimensionless momentum equations are

$$
\delta Re \left( \frac{\partial U}{\partial T} + U \frac{\partial U}{\partial X} + W \frac{\partial U}{\partial Z} \right) = -\frac{\partial}{\partial X} (P + \Phi) + \delta^2 \frac{\partial^2 U}{\partial X^2} + \frac{\partial^2 U}{\partial Z^2}
$$

$$
- \frac{1}{2} \frac{\beta\gamma}{\nu D} \frac{\partial}{\partial X} \left[ \delta^2 \left( \frac{\partial c}{\partial X} \right)^2 + \left( \frac{\partial c}{\partial Z} \right)^2 \right] - \frac{\beta\gamma}{\nu D} \frac{\partial c}{\partial X} \left[ \delta^2 \frac{\partial^2 c}{\partial X^2} + \frac{\partial^2 c}{\partial Z^2} \right],
$$

(7)

$$
\delta^3 Re \left( \frac{\partial W}{\partial T} + U \frac{\partial W}{\partial X} + W \frac{\partial W}{\partial Z} \right) = -\frac{\partial}{\partial Z} (P + \Phi) + \delta^4 \frac{\partial^2 W}{\partial X^2} + \delta^2 \frac{\partial^2 W}{\partial Z^2}
$$

$$
- \frac{1}{2} \frac{\beta\gamma}{\nu D} \frac{\partial}{\partial Z} \left[ \delta^2 \left( \frac{\partial c}{\partial X} \right)^2 + \left( \frac{\partial c}{\partial Z} \right)^2 \right] - \frac{\beta\gamma}{\nu D} \frac{\partial c}{\partial Z} \left[ \delta^2 \frac{\partial^2 c}{\partial X^2} + \frac{\partial^2 c}{\partial Z^2} \right],
$$

(8)

where

$$
Re = \frac{\delta D}{\nu} = \frac{(\delta D/h_0) h_0}{\nu} = O(1).
$$

(9)

The choice of ordering for the Reynolds number $Re$ allows us to recover half-Poiseuille flow at $O(1)$. We delay choosing the ordering of the dimensionless group $\beta\gamma/D\nu$ until we have examined the concentration equation, which in nondimensional form is

$$
\delta^2 \left( \frac{\partial c}{\partial T} + U \frac{\partial c}{\partial X} + W \frac{\partial c}{\partial Z} \right)
$$

$$
= \delta^2 \frac{\partial^2}{\partial X^2} (c^3 - c) + \frac{\partial^2}{\partial Z^2} (c^3 - c) - \delta^4 C_n^2 \frac{\partial^4 c}{\partial X^4} - C_n^2 \frac{\partial^4 c}{\partial Z^4} - 2\delta^2 C_n^2 \frac{\partial^2}{\partial X^2} \frac{\partial c}{\partial Z^2},
$$

(10)

where $C_n^2 = \gamma/h_0^2$. By switching off the backreaction in the momentum equations (corresponding to $\beta\gamma/D\nu \to \infty$), we find the trivial solution to the momentum equations, $U = W = \partial_X (P + \Phi) = \partial_Z (P + \Phi) = 0$, $H = 1$. The concentration
boundary conditions are then \( c_Z = c_{ZZZ} = 0 \) on \( Z = 0, 1 \) which forces \( c_Z \equiv 0 \) so that the Cahn–Hilliard equation is simply
\[
\frac{\partial c}{\partial T} = \frac{\partial^2}{\partial X^2} (c^3 - c) - \delta^2 c_n^2 \frac{\partial^4 c}{\partial X^4}.
\]
To make the lubrication approximation consistent, we take
\[
\delta C_n = \tilde{C}_n = \delta \sqrt{\gamma / h_0} = O(1).
\]
We now carry out a long-wavelength approximation to Eq. (10), writing
\[
U = U_0 + O(\delta), \quad W = W_0 + O(\delta), \quad c = c_0 + \delta c_1 + \delta^2 c_2 + \ldots.
\]
We examine the boundary conditions on \( c(x, t) \) first. They are \( \hat{n} \cdot \nabla c = \hat{n} \cdot \nabla c = 0 \) on \( Z = 0, H \); on \( Z = 0 \) these conditions are simply \( \partial_Z c = \partial_{ZZZ} c = 0 \), while on \( Z = H \) the surface derivatives are determined by the relations
\[
\hat{n} \cdot \nabla \propto -\delta^2 H_X \partial_X + \partial_Z,
\]
\[
\hat{n} \cdot \nabla^2 \propto -\delta^4 H_X \partial_{XXX} - \delta^2 H_X \partial_X \partial_{ZZ} + \delta^2 \partial_X \partial_Z + \partial_{ZZZ}.
\]
Thus, the BCs on \( c_0 \) are simply \( \partial_Z c_0 = \partial_{ZZZ} c_0 = 0 \) on \( Z = 0, H \), which forces \( c_0 = c_0(X, T) \). Similarly, we find \( c_1 = c_1(X, T) \), and
\[
\frac{\partial c_2}{\partial Z} = Z \frac{H_X}{H} \frac{\partial c_0}{\partial X}, \quad \frac{\partial^2 c_2}{\partial Z^2} = \frac{H_X}{H} \frac{\partial c_0}{\partial X}, \quad \text{for any } Z \in [0, H].
\]
In the same manner, we derive the results \( \partial_{ZZZZ} c_2 = \partial_{ZZZZ} c_3 = 0 \). Using these facts, Eq. (10) becomes
\[
\frac{\partial c_0}{\partial T} + U_0 \frac{\partial c_0}{\partial X} = \frac{\partial^2}{\partial X^2} (c_0^3 - c_0) - \tilde{C}_n \frac{\partial^4 c}{\partial X^4} + (3c_0^2 - 1) \frac{H_X}{H} \frac{\partial c_0}{\partial X} - 2 \tilde{C}_n \frac{\partial^2}{\partial X^2} \frac{H_X}{H} \frac{\partial c_0}{\partial X} - \tilde{C}_n \frac{\partial^4 c_4}{\partial Z^4}.
\]
We now integrate this equation from \( Z = 0 \) to \( H \) and use the boundary conditions
\[
\frac{\partial^3 c_4}{\partial Z^3} = 0 \quad \text{on } Z = 0,
\]
\[
\frac{\partial^3 c_4}{\partial Z^3} = H_X \frac{\partial^3 c_0}{\partial X^3} + H_X \frac{\partial}{\partial X} \left( \frac{H_X}{H} \frac{\partial c_0}{\partial X} \right) - H \frac{\partial^2}{\partial X^2} \left( \frac{H_X}{H} \frac{\partial c_0}{\partial X} \right) \quad \text{on } Z = H.
\]
After rearrangement, the concentration equation becomes

\[
H \frac{\partial c_0}{\partial T} + H \langle U_0 \rangle \frac{\partial c_0}{\partial X} = H \frac{\partial^2}{\partial X^2} \left[ c_0^3 - c_0 - \tilde{C}_n^2 \frac{\partial^2 c_0}{\partial X^2} - \frac{\tilde{C}_n}{H} \frac{\partial c_0}{\partial X} \right] + \frac{\partial H}{\partial X} \frac{\partial}{\partial X} \left[ c_0^3 - c_0 - \tilde{C}_n^2 \frac{\partial^2 c_0}{\partial X^2} - \frac{\tilde{C}_n}{H} \frac{\partial c_0}{\partial X} \right],
\]

where

\[
\langle U_0 \rangle = \frac{1}{H} \int_0^H U_0(X, Z, T) \, dZ
\]
is the vertically-averaged velocity. Introducing

\[
\mu = c_0^3 - c_0 - \tilde{C}_n^2 \frac{\partial}{\partial X} \left( \frac{H}{H} \frac{\partial c_0}{\partial X} \right),
\]
the thin-film Cahn–Hilliard equation becomes

\[
\frac{\partial c_0}{\partial T} + \langle U_0 \rangle \frac{\partial c_0}{\partial X} = \frac{1}{H} \frac{\partial}{\partial X} \left( \frac{H}{H} \frac{\partial \mu}{\partial X} \right).
\]

(12)

We are now able to perform the long-wavelength approximation to Eqs. (7) and (8). At lowest order, Eq. (8) is

\[
\frac{\partial Z}{\partial T} (P + \Phi) = 0,
\]
since \( c_0 = c_0(X, T) \), and hence

\[
P + \Phi = P_{\text{surf}} + \Phi_{\text{surf}} \equiv P(X, h(x, t), T) + \Phi(X, h(x, t), T).
\]

By introducing the backreaction strength

\[
r = \frac{\delta^2 \beta \gamma}{D \nu} = O(1),
\]
(13)
equation (7) becomes

\[
\frac{\partial^2 U_0}{\partial Z^2} = \frac{\partial}{\partial X} \left( P_{\text{surf}} + \Phi_{\text{surf}} \right) + r \frac{\partial}{\partial X} \left( \frac{\partial c_0}{\partial X} \right)^2 + r \frac{\partial c_0}{\partial X} \frac{\partial^2 c_2}{\partial Z^2}.
\]

Using \( \partial ZZc_2 = (H_X/H) (\partial c_0/\partial X) \) this becomes

\[
\frac{\partial^2 U_0}{\partial Z^2} = \frac{\partial}{\partial X} \left( P_{\text{surf}} + \Phi_{\text{surf}} \right) + r \frac{\partial}{H \partial X} \left[ H \left( \frac{\partial c_0}{\partial X} \right)^2 \right].
\]

(14)

At lowest order, the BC (5b) reduces to

\[
\frac{\partial U_0}{\partial Z} = \frac{\partial \Sigma}{\partial X} \quad \text{on} \quad Z = H,
\]
which combined with Eq. (14) yields the relation

$$\frac{\partial U_0}{\partial Z} = \frac{\partial \Sigma}{\partial X} + (Z - H) \left\{ \frac{\partial}{\partial X} (P_{surf} + \Phi_{surf}) + \frac{r}{H} \frac{\partial}{\partial X} \left[ H \left( \frac{\partial c_0}{\partial X} \right)^2 \right] \right\}. $$

Here $\Sigma$ is the dimensionless, spatially-varying surface tension. Making use of the BC $U_0 = 0$ on $Z = 0$ and integrating again, we obtain the result

$$U_0 (X, Z, T) = Z \frac{\partial \Sigma}{\partial X} + \left( \frac{1}{2} Z^2 - HZ \right) \left\{ \frac{\partial}{\partial X} (P_{surf} + \Phi_{surf}) + \frac{r}{H} \frac{\partial}{\partial X} \left[ H \left( \frac{\partial c_0}{\partial X} \right)^2 \right] \right\}. $$

The vertically-averaged velocity is therefore

$$\langle U_0 \rangle = \frac{1}{2} H \frac{\partial \Sigma}{\partial X} - \frac{1}{3} H^2 \left\{ \frac{\partial}{\partial X} \left( - \frac{1}{C} \frac{\partial H}{\partial X}^2 + \Phi_{surf} \right) + \frac{r}{H} \frac{\partial}{\partial X} \left[ H \left( \frac{\partial c_0}{\partial X} \right)^2 \right] \right\}, $$

where we used the standard Laplace–Young free-surface boundary condition to eliminate the pressure, and

$$C = \frac{\nu \rho D}{h_0 \sigma_0 \delta^2} = O (1). $$

Finally, by integrating the continuity equation in the $Z$-direction, we obtain, in a standard manner, an equation for free-surface variations,

$$\frac{\partial H}{\partial X} + \frac{\partial}{\partial X} (H \langle U_0 \rangle) = 0. $$

Let us assemble our results, restoring the lower-case fonts and omitting ornamentation over the constants. The height equation (17) becomes

$$\frac{\partial h}{\partial t} + \frac{\partial J}{\partial x} = 0, $$

while the concentration equation (12) becomes

$$\frac{\partial}{\partial t} (ch) + \frac{\partial}{\partial x} (Jc) = \frac{\partial}{\partial x} \left( h \frac{\partial \mu}{\partial x} \right), $$

where

$$J = \frac{1}{2} h^2 \frac{\partial \sigma}{\partial x} - \frac{1}{3} h^3 \left\{ \frac{\partial}{\partial x} \left( - \frac{1}{C} \frac{\partial^2 h}{\partial X^2} + \phi \right) + \frac{r}{h} \frac{\partial}{\partial x} \left[ h \left( \frac{\partial c}{\partial x} \right)^2 \right] \right\}. $$
and
\[ \mu = c^3 - c - C_n^2 \frac{1}{n} \frac{\partial}{\partial x} \left( h \frac{\partial c}{\partial x} \right), \]  \hspace{2cm} (18d)
and where we have the nondimensional constants
\[ r = \frac{\delta^2 \beta \gamma}{D \nu}, \quad C_n = \frac{\delta \sqrt{\gamma}}{h_0}, \quad C = \frac{\nu \rho D}{h_0 \sigma_0 \delta^2}. \]  \hspace{2cm} (19)

These are the thin-film NSCH equations. The integral quantities defined in Eq. (6) are manifestly conserved, while the free surface and concentration are coupled.

We note that the relation \( C_n = \delta \sqrt{\gamma}/h_0 = O(1) \) is the condition that the mean thickness of the film be much smaller than the transition layer thickness. In experiments involving the smallest film thicknesses attainable \( (10^{-8} \text{ m}) \) \cite{28}, this condition is automatically satisfied. The condition is also realised in ordinary thin films when external effects such as the air-fluid and fluid-substrate interactions do not prefer one binary fluid component or another. In this case, the vertical extent of the domains becomes comparable to the film thickness at late times, the thin film behaves in a quasi two-dimensional way, and the model equations are applicable.

The choice of potential \( \phi \) determines the behaviour of solutions. If interactions between the fluid and the substrate and air interfaces are important, the potential should take account of the Van der Waals forces present. A simple model potential is thus
\[ \phi = Ah^{-n}, \]
where \( A \) is the dimensionless Hamakar constant and typically \( n = 3 \) \cite{26}. Here \( A \) can be positive or negative, with positivity indicating a net attraction between the fluid and the substrate and negativity indicating a net repulsion. This choice of potential can also have a regularising effect, preventing a singularity or rupture from occurring in Eq. (18).

For \( \phi = -|A|/h^{3} \) (repulsive Van der Waals interactions), the system of equations (18) has a Lyapunov functional \( F = F_1 + F_2 \), where
\[ F_1 = \int dx \left[ \frac{1}{2C} \left( \frac{\partial h}{\partial x} \right)^2 + \frac{|A|}{2h^2} \right], \quad F_2 = r \int dx h \left[ \frac{1}{4} \left( c^2 - 1 \right)^2 + \frac{C_n^2}{2} \left( \frac{\partial c}{\partial x} \right)^2 \right]. \]

By differentiating these expressions with respect to time, we obtain the relation
\[ \dot{F}_1 + \dot{F}_2 \]
\[ = -\frac{1}{3} \int dx h^3 \left\{ \frac{\partial}{\partial x} \left( \frac{1}{C} \frac{\partial^2 h}{\partial x^2} + \frac{|A|}{h^3} \right) - \frac{r}{h} \frac{\partial}{\partial x} \left[ h \left( \frac{\partial c}{\partial x} \right)^2 \right] \right\}^2 - \int dx h \left( \frac{\partial \mu}{\partial x} \right)^2, \]  \hspace{2cm} (20)
which is nonpositive for nonnegative \( h \). This fact is the key to the analytic results of the next section.
III. EXISTENCE OF SOLUTIONS TO THE MODEL EQUATIONS

In this section we prove that solutions to the model equations do indeed exist. We set $C = \frac{1}{3}, r = |A| = 1$ in Eqs. (18) and focus on the resulting equation set

$$\frac{\partial h}{\partial t} = -\frac{\partial}{\partial x} \left[ f(h) \frac{\partial^3 h}{\partial x^3} + \frac{1}{g(h)} \frac{\partial h}{\partial x} + \frac{f(h) \partial}{g(h) \partial x} \left[ g(h) \left( \frac{\partial c}{\partial x} \right)^2 \right] \right], \quad (21a)$$

$$\frac{\partial}{\partial t} (cg(h)) = -\frac{\partial}{\partial x} \left[ cf(h) \frac{\partial^3 h}{\partial x^3} + \frac{c}{g(h)} \frac{\partial h}{\partial x} + \frac{f(h) \partial}{g(h) \partial x} \left[ g(h) \left( \frac{\partial c}{\partial x} \right)^2 \right] \right]$$

$$+ \frac{\partial}{\partial x} \left\{ g(h) \frac{\partial}{\partial x} \left[ c^3 - c - \frac{1}{g(h)} \frac{\partial}{\partial x} \left( g(h) \frac{\partial c}{\partial x} \right) \right] \right\}, \quad (21b)$$

where

$$f(h) = h^3, \quad g(h) = h.$$ 

The equations are defined on a periodic domain $\Omega = [0, L]$, while the initial conditions are

$$h(x, 0) = h_0(x) > 0, \quad c(x, 0) = c_0(x), \quad h_0(x), c_0(x) \in W^{2,2}(\Omega), \quad (22)$$

We shall prove that the solutions to this equation pair exist in the strong sense; however, we shall need the definition of weak solutions:

A pair $(h, c)$ is a weak solution of Eq. (21) if the following integral relations hold:

$$\int_0^{T_0} dt \int_{\Omega} d\varphi_x h =$$

$$\int_0^{T_0} dt \int_{\Omega} d\varphi_x \left\{ - f(h) \frac{\partial^3 h}{\partial x^3} + \frac{1}{g(h)} \frac{\partial h}{\partial x} + \frac{f(h) \partial}{g(h) \partial x} \left[ g(h) \frac{\partial}{\partial x} \left( \frac{\partial c}{\partial x} \right)^2 \right] \right\},$$

and

$$\int_0^{T_0} dt \int_{\Omega} d\psi_x cg(h) =$$

$$\int_0^{T_0} dt \int_{\Omega} d\psi_x \left\{ - cf(h) \frac{\partial^3 h}{\partial x^3} + \frac{c}{g(h)} \frac{\partial h}{\partial x} + \frac{f(h) \partial}{g(h) \partial x} \left[ g(h) \frac{\partial}{\partial x} \left( \frac{\partial c}{\partial x} \right)^2 \right] \right\}$$

$$+ \int_0^{T_0} dt \int_{\Omega} d\psi_x \left\{ g(h) \frac{\partial}{\partial x} \left[ c^3 - c - \frac{1}{g(h)} \frac{\partial}{\partial x} \left( g(h) \frac{\partial c}{\partial x} \right) \right] \right\}, \quad (23)$$
where \( T_0 > 0 \) is any time, and \( \varphi(x,t) \) and \( \psi(x,t) \) are arbitrary differentiable test functions that are periodic on \( \Omega \) and vanish at \( t = 0 \) and \( t = T_0 \).

In a series of steps in Secs. III A–III G, we prove this result:

Given the initial data in Eq. (22), Eqs. (21) possess a strong solution endowed with the following regularity properties:

\[
(h, c) \in L^\infty(0, T_0; W^{2,2}(\Omega)) \cap L^2(0, T_0; W^{4,2}(\Omega)) \cap C^{3,1}(\Omega \times [0, T_0]).
\]

The outline of the proof is as follows: In Sec. III A we introduce a regularised version of Eqs. (21), whose solution we approximate by a Galerkin sum in Sec. III B. In Sec. III C, we obtain a priori bounds on various norms of the approximate solution. Crucially, we show that the free-surface height is always positive. This enables us to continue the approximate solution in the time interval \([0, T_0]\). In Secs. III D and III E we show that the Galerkin sum converges to a solution of the unapproximated equations, in the appropriate limit. Finally, in Secs. III F and III G we discuss the regularity and uniqueness properties of the solution.

### A. Regularisation of the problem

We introduce regularised functions \( f_\varepsilon(s) \) and \( g_\varepsilon(s) \) such that \( \lim_{\varepsilon \to 0} f_\varepsilon(s) = f(s) \), and \( \lim_{\varepsilon \to 0, s \geq 0} g_\varepsilon(s) = g(s) \). For now we do not specify \( f_\varepsilon(s) \), although we mention that a suitable choice of \( f_\varepsilon(s) \) will cure the degeneracy of the fourth-order term in the height equation. On the other hand, we require that \( g_\varepsilon(s) \) have the properties: (i) \( g_\varepsilon(s) = s + \varepsilon \), for \( s \geq 0 \); (ii) \( g_\varepsilon(s) > 0 \), for \( s < 0 \); (iii) \( \lim_{s \to -\infty} g_\varepsilon(s) = \frac{1}{2} \varepsilon \); and (iv) \( g_\varepsilon(s) \) is at least \( C^3 \).

From Eqs. (21), the regularised PDEs we study are

\[
\begin{align*}
  h_t &= -J_{\varepsilon,x}, \quad \text{(24a)} \\
  (c g_\varepsilon(h))_t &= - (c J_{\varepsilon})_x - (g_\varepsilon(h) \mu_{\varepsilon,x})_x, \quad \text{(24b)}
\end{align*}
\]

where

\[
\mu_\varepsilon = c^3 - c - \frac{1}{g_\varepsilon(h)} (g_\varepsilon(h)_x)_x
\]

and

\[
J_\varepsilon = f_\varepsilon(h)_x x - \frac{1}{g_\varepsilon(h)} h_x - \frac{f_\varepsilon(h)}{g_\varepsilon(h)} (g_\varepsilon(h)c_x)_x.
\]
Equation (24b) can also be written as
\[
c_t = -\frac{1}{g_\varepsilon(h)} J_\varepsilon c_x - \frac{1}{g_\varepsilon(h)} (g_\varepsilon(h)\mu_{\varepsilon,x})_x - \frac{c}{g_\varepsilon(h)} [J_{\varepsilon,x} + g_\varepsilon'(h)h_t];
\] (25)
this form of the concentration equation will be useful in Sec. III G.

B. The Galerkin approximation

We choose a complete orthonormal basis on the interval \(\Omega\), with periodic boundary conditions. Let us denote the basis by \(\{\phi_i(x)\} \in \mathbb{N}_0\). We consider the finite vector space \(\text{Span}\{\phi_0, \ldots, \phi_n\}\). For convenience, let us take the \(\phi_i(x)\)'s to be the eigenfunctions of the Laplacian on \([0, L]\) with periodic boundary conditions, and corresponding eigenvalues \(-\lambda_i^2\). Let \(\phi_0\) be the constant eigenfunction. We construct approximate solutions to the PDEs (24) as finite sums,
\[
h_n(x, t) = \sum_{i=0}^{n} \eta_{n,i}(t)\phi_{i}(x), \quad c_n(x, t) = \sum_{i=0}^{n} \gamma_{n,i}(t)\phi_{i}(x).
\]
If the (smooth) initial data are given as
\[
h(x, 0) = h_0(x) = \sum_{i=0}^{\infty} \eta_i^0 \phi_i(x) > 0, \quad c(x, 0) = c_0(x) = \sum_{i=0}^{\infty} \gamma_i^0 \phi_i(x),
\]
then the initial data for the Galerkin approximation are
\[
h_n(x, 0) = h_0^0(x) = \sum_{i=0}^{n} \eta_i^0 \phi_i(x), \quad c_n(x, 0) = c_0^0(x) = \sum_{i=0}^{n} \gamma_i^0 \phi_i(x),
\]
and the initial data of the Galerkin approximation converge strongly in the \(L^2(\Omega)\) norm to the initial data of the unapproximated problem. Thus, there is a \(n_0 \in \mathbb{N}\) such that \(h_0^0(x) > 0\), everywhere in \(\Omega\), for all \(n > n_0\). Henceforth we work with Galerkin approximations with \(n > n_0\).
Substitution of \(h_n = \sum_{i=0}^{n} \eta_{n,i} \phi_i\) into a weak form of the \(h\)-equation yields
\[
\frac{d}{dt} (\langle h_n, \phi_j \rangle) = \langle (J_{\varepsilon,n}, \phi_{j,x}) \rangle, \tag{26}
\]
where
\[
J_{\varepsilon}(h_n, c_n) = f_\varepsilon(h_n)h_{n,xxx} - \frac{1}{g_\varepsilon(h_n)} h_{n,x} - \frac{f_\varepsilon(h_n)}{g_\varepsilon(h_n)} (g_\varepsilon(h_n) c_{n,x}^2)_x.
\]
is the flux for the regularised $h$-equation, and $(\varphi(x), \psi(x))$ is the pairing $\int_\Omega \varphi \psi \, dx$. We recast Eq. (26) as

$$\frac{d\eta_{n,j}}{dt} = (J_{\varepsilon,n}, \phi_{j,x}) = \Phi_{n,j}(\eta_n, \gamma_n),$$

where the function $\Phi_n(\eta_n, \gamma_n)$ depends on $\eta_n = (\eta_{n,0}, \ldots, \eta_{n,n})$ and $\gamma_n = (\gamma_{n,0}, \ldots, \gamma_{n,n})$, and is locally Lipschitz in its variables. This Lipschitz property arises from the fact that the regularised flux, evaluated at the Galerkin approximation, is a composition of Lipschitz continuous functions, and therefore, is itself Lipschitz continuous.

Similarly, substitution of $c_n = \sum_{i=0}^n \gamma_{n,i} \phi_i$ into the weak form of the $c$-equation (24b) yields

$$\frac{d}{dt} (g(h_n)c_n, \phi_j) = (K_{\varepsilon,n}, \phi_{j,x}),$$

where

$$K_{\varepsilon}(h_n, c_n) = c_n f_\varepsilon(h_n)h_{n,xxx} - \frac{c_n}{g_\varepsilon(h_n)} h_x - c_n \frac{f_\varepsilon(h_n)}{g_\varepsilon(h_n)} (g_\varepsilon(h_n)c_{n,x}^2) - g(h_n)\mu_{\varepsilon,n,x},$$

$$= c_n J_{\varepsilon,n} - g_\varepsilon(h_n)\mu_{\varepsilon,n,x},$$

is the flux for the regularised $c$-equation (24b). Rearranging gives

$$(g(h_n)c_{n,t}, \phi_j) = (K_{\varepsilon,n}, \phi_{j,x}) - (g'(h_n)c_n h_{n,t}, \phi_j),$$

and the left-hand side can be recast in matrix form as $\sum_{i=0}^n M_{ij} \dot{\gamma}_{n,i}$, where

$$M_{ij} = \int_\Omega \frac{d}{dx} g_\varepsilon \left( \sum_{\ell} \eta_{n,\ell} \phi_\ell \right) \phi_i \phi_j,$$

which is manifestly symmetric. It is positive definite because given a vector $(\xi_0, \ldots, \xi_n)$, we have the relation

$$\sum_{i,j} \xi_i M_{ij} \xi_j = \int_\Omega \frac{d}{dx} g_\varepsilon \left( \sum_{\ell} \eta_{n,\ell} \phi_\ell \right) \sum_{i,j} (\phi_i \xi_i) (\phi_j \xi_j) > 0, \quad \text{for} \ (\xi_0, \ldots, \xi_n) \neq 0,$$

which follows from the positivity of the regularised function $g_\varepsilon(s)$. We therefore have the following equation for $\gamma_{n,j}(t)$,

$$\frac{d\gamma_{n,j}}{dt} = \sum_{i=0}^n M_{ij}^{-1} \left[ (K_{\varepsilon,n}, \phi_{i,x}) - (g'(h_n)c_n h_{n,t}, \phi_i) \right].$$

(28)
Inspecting the expression for $M_{ij}$, $g(h_n)$, and $K_{\varepsilon,n}$, we see that $\eta_n$ and $\gamma_n$ appear in a Lipschitz-continuous way in the expression $\sum_{i=0}^{n} M_{ij}^{-1} (K_{\varepsilon,n}, \phi_i, x)$, while owing to the imposed smoothness of $g_\varepsilon(s)$, the variables $\eta_n$, $\gamma_n$ and $\dot{\eta}_n = (\dot{\eta}_{n,0}, \ldots, \dot{\eta}_{n,n})$ appear in a Lipschitz-continuous way in the quantity $\sum_{i=0}^{n} M_{ij}^{-1} ((g'_\varepsilon(h_n)c_n)h_{n,t}, \phi_i)$. The vector $\dot{\eta}_n$ can be replaced by the function $\Phi_n(\eta_n, \gamma_n)$ and thus we obtain a relation
\[
\frac{d\gamma_{n,j}}{dt} = \Psi_{n,j}(\eta_n, \gamma_n),
\]
in place of Eq. (28), where $\Psi_{n,j}(\eta_n, \gamma_n)$ is Lipschitz. We therefore have a system of Lipschitz-continuous equations
\[
\frac{d\eta_{n,j}}{dt} = \Phi_{n,j}(\eta_n, \gamma_n), \quad \frac{d\gamma_{n,j}}{dt} = \Psi_{n,j}(\eta_n, \gamma_n),
\]
and thus local existence theory [12] guarantees a solution for the $\eta_{n,i}$’s and $\gamma_{n,i}$’s for all times $t$ in a finite interval $0 < t < \sigma$. This solution is, moreover, unique and continuous. To continue this approximate solution to the PDE problem in Eq. (21) up to an an arbitrary time $T_0 > 0$, it is necessary to find a priori bounds on the approximate local-in-time solution.

C. A priori bounds on the Galerkin approximation

We identify the free energy
\[
F = \int_{\Omega} dx \left[ \frac{3}{2} h_x^2 + G_\varepsilon(h) \right] + \int_{\Omega} dx g_\varepsilon(h) \left[ \frac{1}{4} (c^2 - 1)^2 + \frac{1}{2} c_x^2 \right],
\]
where $G''_\varepsilon(s) = 1/ [f_\varepsilon(s)g_\varepsilon(s)]$. Since the Galerkin approximation satisfies the weak form of the PDEs given in Eqs. (26) and (27), it is possible to obtain the free-energy decay law
\[
\frac{dF}{dt} (t) = -\int_{\Omega-\Omega_-} dx f_\varepsilon(h_n) \left[ -h_{n,xxx} + \frac{h_{n,x}}{g_\varepsilon(h_n) h_\varepsilon(h_n)} + \frac{1}{g_\varepsilon(h_n)} (g_\varepsilon(h_n) c_n^2 x) \right]^2 dx
\]
\[
- \int_{\Omega} dx g_\varepsilon(h_n) \mu_{\varepsilon,n,x}^2 + \int_{\Omega_-} dx \ldots, \quad 0 \leq t < \sigma, \quad (29)
\]
where $\Omega_- (t) = \{ x \in \Omega | h_n(x, t) < 0 \}$. Now given the time-continuity of $h_n(x, t)$ in $(0, \sigma)$, and the initial condition $h_n^0(x) > 0$ (since $n > n_0$), there is a time $\sigma_1 > 0$ such that $h_n(x, t) > 0$ for all $x \in \Omega$ and all $t \in (0, \sigma_1)$. Therefore, $\Omega_- (t) = \emptyset$ for $t \in (0, \sigma_1)$, the last integral in (29) vanishes, and hence
\[
F [c_n(x, t), h_n(x, t)] \leq F [c_n(x, 0), h_n(x, 0)] \leq \sup_{\varepsilon,n} F [c_n(x, 0), h_n(x, 0)] < \infty,
\]
for $0 < t < \sigma_1$. Consequently, we obtain the bound $\|h_{n,x}\|_2 \leq k_1$, where $0 < t < \sigma_1$, and where $k_1$ depends only on the initial conditions. We have Poincaré's inequality for $h_{n,x}$,

$$\|h_n\|_2^2 \leq \left[ \int_\Omega dx h_n(x) \right]^2 + \left( \frac{L}{2\pi} \right)^2 \|h_{n,x}\|_2^2.$$ 

Now $\int_\Omega dx h_n(x, t) = L\eta_{n,0}(t)$. Inspection of Eq. (26) shows that $\eta_{n,0}(t) = \eta_{n,0}(0) = \eta_0^\varepsilon$. Thus,

$$\|h_n\|_2^2 \leq L^2 |\eta_0^\varepsilon|^2 + \left( \frac{L}{2\pi} \right)^2 k_1 \equiv k_2 < \infty.$$ 

Using result [A1] from Appendix A, we obtain the bound

$$\|h_n\|_\infty \leq \frac{1}{\sqrt{L}} \|h_n\|_2 + \sqrt{L} \|h_{n,x}\|_2 \equiv k_3.$$ 

Additionally, the following properties hold:

- The function $h_n$ is Hölder continuous in space, with exponent $\frac{1}{2}$;
- $\int_\Omega dx G_\varepsilon(h_n) \leq k_4$.

These results hold in $0 < t < \sigma_1$, and the constants $k_1$, $k_2$, $k_3$, and $k_4$ are independent of $\varepsilon$, $n$, $\sigma$, and $\sigma_1$, and in fact depend only on the functions $h_0(x)$ and $c_0(x)$.

To continue the estimates to the whole interval $(0, \sigma)$, we need to prove that $h_n(\cdot, \sigma_1) > 0$ almost everywhere (a.e.). If this is true, there is a new interval $(\sigma_1, \sigma_2)$, $\sigma_1 < \sigma_2 \leq \sigma$, on which $h_n(\cdot, t) > 0$ a.e., and we can then provide a priori bounds on $h_n(\cdot, t)$ and $c_n(\cdot, t)$ on the interval $[\sigma_1, \sigma_2]$. It is then possible to show that $h_n(\cdot, \sigma_2) > 0$ a.e. and thus, by iteration, we extend the proof to the whole interval $(0, \sigma)$, and find that $h_n(\cdot, t) > 0$ a.e. on $(0, \sigma)$.

We have the bound

$$\int_\Omega dx G_\varepsilon(h_n(\cdot, t)) \leq k_4,$$

where $k_4$ depends only on the initial conditions, and where $0 < t < \sigma_1$. We now specify $G_\varepsilon(s)$ in more detail. This function satisfies the condition

$$G_\varepsilon''(s) = \frac{1}{f_\varepsilon(s) g_\varepsilon(s)}.$$ 

We take $g_\varepsilon(s)$ to be as defined previously, and we choose a simple regularisation for $f(s)$:

$$f_\varepsilon(s) = g_\varepsilon(s)^3.$$
which is Lipschitz continuous. By defining
\[ \tilde{G}_\epsilon(s) = -\int_s^\infty \frac{dr}{f_\epsilon(r) g_\epsilon(r)}, \quad G_\epsilon(s) = -\int_s^\infty dr \tilde{G}_\epsilon(r), \]
we obtain a function \( G_\epsilon(s) \) that is positive for all \( s \in (-\infty, \infty) \), and
\[ G_\epsilon(s) = \frac{1}{6} \frac{1}{(s + \epsilon)^2}, \quad s \geq 0. \]

Using the boundedness of \( G_\epsilon(s) \), and the time-continuity of \( h_n(\cdot, t) \), we apply the Dominated Convergence Theorem,
\[ \lim_{t \to \sigma_1} \int_\Omega dx G_\epsilon(h_n(\cdot, t)) = \int_\Omega dx \lim_{t \to \sigma_1} G_\epsilon(h_n(\cdot, t)) = \int_\Omega dx G_\epsilon(h_n(\cdot, \sigma_1)) \leq k_4. \]

Similarly, since the constant \( k_1 \) in the inequality \( \| h_n, x \|_2 \leq k_1, 0 \leq t < \sigma_1 \) depends only on the initial data, we extend this last inequality to \( t = \sigma_1 \), and thus \( h_n(x, \sigma_1) \) is Hölder continuous in space.

In the worst-case scenario, the time \( \sigma_1 \) is the first time at which \( h_n(x, t) \) touches down to zero, and and we therefore assume for contradiction that \( h_n(x_0, \sigma_1) = 0 \), and that \( h_n(x, \sigma_1) \geq 0 \) elsewhere. Then, by Hölder continuity, for any \( x \in \Omega \) we have the bound \( 0 \leq h_n(x, \sigma_1) \leq k_1 |x - x_0|^\frac{1}{2} \), and thus
\[ \int_\Omega dx G_\epsilon(h_n(\cdot, \sigma_1)) \geq \frac{k_1}{6} \int_0^L \frac{dx}{|x - x_0| + \epsilon(2\sqrt{L} + \epsilon)}. \]

Hence,
\[ \frac{6}{k_1} \int_\Omega dx G_\epsilon(h_n(\cdot, \sigma_1)) \]
\[ \geq -2 \log \left[ \epsilon \left( 2\sqrt{L} + \epsilon \right) \right] + \log \left\{ \left[ L - x_0 + \left( 2\sqrt{L} + \epsilon \right) \epsilon \right] \left[ x_0 + \left( 2\sqrt{L} + \epsilon \right) \epsilon \right] \right\}. \]

Thus, the integral \( \int_\Omega G_\epsilon(h_n(x, \sigma_1))dx \) can be made arbitrarily large, which contradicts the \( \epsilon \)-independent bound for this quantity, obtained in Eq. (30). We therefore have the strong condition that the set on which \( h_n(\cdot, \sigma_1) \leq 0 \) is empty. Iterating the argument, we have the important result

The set on which \( h_n(\cdot, t) \leq 0 \) is empty, for \( 0 < t < \sigma \).
Using the same argument, we have an estimate on the minimum value of $h_n(x,t)$,

$$h_{\text{min}} = \min_{x \in \Omega, t \in (0, \sigma]} h_n(x,t),$$

namely,

$$h_{\text{min}} + \varepsilon \geq -k_1 \sqrt{L} + \sqrt{k_1^2 L + \frac{k_1^2 L}{e^{k_4 k_1^2} - 1}},$$

for all small positive $\varepsilon$. Thus,

$$h_{\text{min}} \geq M := -k_1 \sqrt{L} + \sqrt{k_1^2 L + \frac{k_1^2 L}{e^{k_4 k_1^2} - 1}}, \quad (31)$$

a lower bound that depends only on the initial data $c_0(x)$ and $h_0(x)$. Note that this result depends on the intimate relationship between the Hölder continuity of a function and its boundedness in the $W^2$ norm, a relationship that is true only in one dimension. Thus, generalisation of this lower bound, and by extension, long-time existence and uniqueness of solutions, does not necessarily hold in higher dimensions.

Now, using Eq. (31) and the boundedness result

$$\int_\Omega dx g_\varepsilon(h_n) \left[ \frac{1}{4} \left( c_n^2 - 1 \right)^2 + \frac{1}{2} c_{n,x}^2 \right] \leq k_5,$$

where $k_5$ depends only on the initial data, we obtain an *a priori* bound on $\|c_{n,x}\|_2^2$,

$$\int_\Omega dx c_{n,x}^2 \leq \frac{2k_5}{M}.$$

It is also possible to derive a bound on $\|c_n\|_2^2$. We have the relation

$$\int_\Omega dx \left( c_n^2 - 1 \right)^2 \leq \frac{k_5}{M},$$

which gives the inequality $\|c_n\|_4^4 \leq 2\|c\|_2^2 + (4k_5/M)$. Using the Hölder relation $\|c\|_2 \leq |\Omega|^{1/4} \|c\|_4$, we obtain a quadratic inequality in $\|c\|_2^2$,

$$\|c\|_2^4 \leq 2|\Omega|\|c\|_2^2 + \frac{4|\Omega|k_5}{M},$$

with solution

$$\|c\|_2^2 \leq \frac{4|\Omega|k_5}{M},$$

as required. From the boundedness of $\|c_{n,x}\|_2$ and $\|c_n\|_2$ follows the relation $\|c_n\|_\infty \leq k_6 < \infty$, a result that depends only on the initial conditions. Let us recapitulate these results:
• \( \| h_{n,x} \|_2 \) is uniformly bounded;
• \( \| h_n \|_\infty \) is uniformly bounded;
• The function \( h_n \) is Hölder continuous in space, with exponent \( \frac{1}{2} \);
• The function \( h_n \) is nonzero everywhere and never decreases below a certain value \( M > 0 \), independent of \( n, \varepsilon, \) and \( \sigma \).

• \( \| c_{n,x} \|_2 \) is uniformly bounded;
• \( \| c_n \|_\infty \) is uniformly bounded;
• The function \( c_n \) is Hölder continuous in space, with exponent \( \frac{1}{2} \).

These results are independent of \( n, \varepsilon \) and \( \sigma \), and hold for \( 0 < t < \sigma \).

D. Equicontinuity and convergence of the Galerkin approximation

Using Eq. (29), we obtain the bound

\[
\int_0^t dt' \int_\Omega dx \left\{ f_\varepsilon(h_n) \left[ -h_{n,xxx} + \frac{h_{n,x}}{g_\varepsilon(h_n)f_\varepsilon(h_n)} + \frac{1}{g_\varepsilon(h_n)} \left( g_\varepsilon(h_n)c_{n,x}^2 \right)_x \right]^2 + g_\varepsilon(h_n)\mu_{\varepsilon,n,x}^2 \right\} \leq F(0), \quad 0 < t < \sigma,
\]

a bound that is independent of \( n, \sigma, \) and \( \varepsilon \). Since the quantity \( \| f_\varepsilon(h_n) \|_\infty = (\| h_n \|_\infty + \varepsilon)^3 \) is bounded above by a constant \( A_1 \) independent of \( n, \varepsilon, \) and \( \sigma \), we have

\[
\int_0^t dt' \int_\Omega dx J_{\varepsilon,n}^2 \leq A_1 \int_0^t dt' \int_\Omega dx \left\{ f_\varepsilon(h_n) \left[ -h_{n,xxx} + \frac{h_{n,x}}{g_\varepsilon(h_n)f_\varepsilon(h_n)} + \frac{1}{g_\varepsilon(h_n)} \left( g_\varepsilon(h_n)c_{n,x}^2 \right)_x \right]^2 \right\} \leq A_1 F(0) \equiv A_2,
\]

and thus

\[
\int_0^t dt' \| J_{\varepsilon,n} \|_2^2 \leq A_2, \quad 0 < t < \sigma,
\]

where \( A_2 \) is independent of \( n, \varepsilon, \) and \( \sigma \). Similarly,

\[
\int_0^t dt' \| g_\varepsilon(h_n)\mu_{\varepsilon,n,x} \|_2^2 \leq A_3, \quad 0 < t < \sigma,
\]
and
\[ \int_0^t dt' \| c_n J_{\varepsilon,n} \|_2^2 \leq A_2 \| c_n \|_\infty^2 \leq A_2 k_0^2, \quad 0 < t < \sigma, \]
where \( A_2 \) and \( A_3 \) are independent of \( n, \varepsilon, \) and \( \sigma \). By rewriting the evolution equations as
\[ h_{n,t} = -J_{\varepsilon,n,x}, \quad (g_\varepsilon(h))_t = -K_{\varepsilon,n,x}, \]
where \( K_{\varepsilon,n} = c_n J_{\varepsilon,n} - g_\varepsilon(h_n)\mu_{\varepsilon,n,x} \), we see that there are uniform bounds for
\[ \int_0^t dt' \| J_{\varepsilon,n} \|_2^2 \quad \text{and} \quad \int_0^t dt' \| K_{\varepsilon,n} \|_2^2, \]
which depend only on the initial data \( c_0(x) \) and \( h_0(x) \).

Bernis and Friedman [2] proved the following claim:

Let \( \varphi_i(x,t) \) be a sequence of functions, each of which weakly satisfies the equation
\[ \varphi_{i,t} = -J_{i,x}, \quad J_i = J(\varphi_i). \]
If \( \varphi_i(x,\cdot) \) is Hölder continuous (exponent \( \frac{1}{2} \)), and if the fluxes \( J_i \) satisfy
\[ \int_0^t dt' \| J_i \|_2^2 \leq B_1, \quad 0 < t < \sigma, \]
where \( B_1 \) is a number independent of the index \( i \) and the time \( \sigma \), then there is a constant \( B_2 \), independent of \( i \) and \( \sigma \), such that
\[ |\varphi_i(\cdot,t_2) - \varphi_i(\cdot,t_1)| \leq B_2 |t_2 - t_1|^\frac{1}{8}, \]
for all \( t_1 \) and \( t_2 \) in \( (0,\sigma) \).

We now observe that the fluxes \( J_{\varepsilon,n} \) and \( K_{\varepsilon,n} \) satisfy the conditions of this theorem, and thus

The functions \( h_n(\cdot, t) \) and \( c_n(\cdot, t) \) are Hölder continuous (exponent \( \frac{1}{8} \)), for \( 0 < t < \sigma \).

We therefore have a uniformly bounded and equicontinuous family of functions \( \{(h_n, c_n)\}_{n=n_0+1}^\infty \). We also have a recipe for constructing a uniformly bounded and equicontinuous approximate solution \( (h_n(x,t), c_n(x,t)) \), in a small interval \( (0,\sigma) \). The recipe can be iterated step-by-step, and we obtain a uniformly bounded and equicontinuous family of approximate solutions \( \{(h_n, c_n)\}_{n=n_0+1}^\infty \), on \( (0,T_0) \times \Omega \), for an arbitrary time \( T_0 \). Then, using the Arzelà–Ascoli theorem, we obtain the convergence result:

There is a subsequence of the family \( \{(h_n, c_n)\}_{n=n_0+1}^\infty \) that converges uniformly to a limit \( (h, c) \), in \([0,T_0] \times \Omega \).
We prove several facts about the pair \((h, c)\).

Let \((h, c)\) be the limit of the family of functions \(\{(h_n, c_n)\}_{n=n_0+1}^\infty\) constructed in Secs. IIIA–IIID. Then the following properties hold for this limit:

1. The functions \(h(x,t)\) and \(c(x,t)\) are uniformly Hölder continuous in space (exponent \(1/2\)), and uniformly Hölder continuous in time (exponent \(1/8\));
2. The initial condition \((h, c)(x, 0) = (h_0, c_0)(x)\) holds;
3. \((h, c)\) satisfy the boundary conditions of the original problem (periodic boundary conditions);
4. The derivatives \((h, c)_t, (h, c)_x, (h, c)_{xx}, (h, c)_{xxx}\) and \((h, c)_{xxxx}\) are continuous in space and time;
5. The function pair \((h, c)\) satisfy the weak form of the PDEs,

\[
\int \int_{Q_{T_0}} dt dx h \varphi_t + \int \int_{Q_{T_0}} dt dx J_{\epsilon} \varphi_x = 0, \\
J_{\epsilon} = f_{\epsilon}(h)h_{xxx} - \frac{1}{g_{\epsilon}(h)} h_x - f_{\epsilon}(h) \left(\frac{g_{\epsilon}(h)c_{x}^2}{x}\right), \\
\int \int_{Q_{T_0}} dt dx g_{\epsilon}(h) c \psi_t + \int \int_{Q_{T_0}} dt dx K_{\epsilon} \psi_x = 0, \\
K_{\epsilon} = cJ_{\epsilon} - g_{\epsilon}(h) \left[c^3 - c - \frac{1}{g_{\epsilon}(h)} \left(g_{\epsilon}(h)c_x\right)_x\right],
\]

where \(\varphi(x,t)\) and \(\psi(x,t)\) are suitable test functions.

The statements 1, 2, and 3 are obvious. Now, any pair \((h_n(x,t), c_n(x,t))\) satisfies the equation set

\[
\int \int_{Q_{T_0}} dt dx h_n \phi_t + \int \int_{Q_{T_0}} dt dx J_{\epsilon,n} \phi_x = 0, \\
J_{\epsilon,n} = f_{\epsilon}(h_n)h_{n,xxx} - \frac{1}{g_{\epsilon}(h_n)} h_x - f_{\epsilon}(h_n) \left(\frac{g_{\epsilon}(h_n)c_{n,x}^2}{x}\right),
\]
\[ \int \int_{Q_{T_0}} dt dx \, g_\varepsilon(h_n) c_n \psi_t + \int \int_{Q_{T_0}} dt dx \, K_{\varepsilon,n} \psi_x = 0, \]

\[ K_{\varepsilon,n} = c_n J_{\varepsilon,n} - g_\varepsilon(h_n) \left[ c_n^3 - c_n - \frac{1}{g_\varepsilon(h_n)} \left( g_\varepsilon(h_n) c_{n,x} \right)_x \right], \]

and from the boundedness of the fluxes \( J_{\varepsilon,n} \) and \( K_{\varepsilon,n} \) in \( L^2(0,T_0;L^2(\Omega)) \), it follows that

\[ (J_{\varepsilon,n}, K_{\varepsilon,n}) \rightharpoonup (J_\varepsilon, K_\varepsilon), \quad \text{weakly in } L^2(0,T_0;L^2(\Omega)), \]

for a subsequence. Using the regularity theory for uniformly parabolic equations and the uniform Hölder continuity of the \((h_n,c_n)\)'s, it follows that

The derivatives \((h_n,c_n)_t, (h_n,c_n)_x, (h_n,c_n)_{xx}, (h_n,c_n)_{xxx}\), and \((h_n,c_n)_{xxxx}\) are uniformly convergent in any compact subset of \((0,T_0] \times \Omega\).

Thus,

\[ J_\varepsilon = f_\varepsilon(h) h_{xxx} - \frac{1}{g_\varepsilon(h)} h_x - \frac{f_\varepsilon(h)}{g_\varepsilon(h)} \left( g_\varepsilon(h) c_x^2 \right)_x, \]

\[ K_\varepsilon = c J_\varepsilon - g_\varepsilon(h) \left[ c^3 - c - \frac{1}{g_\varepsilon(h)} \left( g_\varepsilon(h) c_x \right)_x \right], \]

on \((0,T_0] \times \Omega\), and therefore, claims 4 and 5 on p. 21 follow.

**E. Convergence of regularised problem, as \( \varepsilon \to 0 \)**

The result in Sec. III D produced a solution \((h_\varepsilon,c_\varepsilon)\) to the regularised problem. Due to the result

\[ h_\varepsilon(x,t) \geq h_{\text{min}} \geq M = -k_1 \sqrt{L} + \sqrt{k_1^2 L + \frac{k_2^2 L}{e^{k_1 k_2^2} - 1}} > 0, \quad (32) \]

independent of \( \varepsilon \), the argument of Sec. III D can be recycled to produce a solution \((h,c)\) to the unregularised problem. This solution is constructed as the limit of a convergent subsequence, formally written as \((h,c) = \lim_{\varepsilon \to 0} (h_\varepsilon, c_\varepsilon)\), and the results of the theorem in Sec. III D apply again to \((h,c)\). The result (32) applies to \( h \) constructed as \( h = \lim_{\varepsilon \to 0} h_\varepsilon \), and thus all the derivatives \((h,c)_t, (h,c)_x, (h,c)_{xx}, (h,c)_{xxx}, \) and \((h,c)_{xxxx}\) are continuous on the whole space \((0,T_0] \times \Omega\) and therefore, the weak solution \((h,c)\) is in fact a strong one.
F. Regularity properties of the solution \((h, c)\)

Using a bootstrap argument, we show that the solution \((h, c)\) belongs to the regularity classes \(L^\infty (0, T_0; W^{2,2} \Omega)\) and \(L^2 (0, T_0; W^{4,2} \Omega)\). From Sec. III C it follows immediately that

\[
\|h_x\|_2, \|c_x\|_2 < \infty,
\]

with time-independent bounds. Thus, using Poincaré’s inequality, it follows that \(h, c \in W^{1,2} \Omega\) and, moreover,

\[
\sup_{[0, T_0]} \|h_x\|_2, \sup_{[0, T_0]} \|c_x\|_2 < \infty.
\]

From Sec. III D, it follows that \(J\) and \(\mu_x\) belong to the regularity class \(L^2 (0, T_0; L^2 \Omega)\), and hence \(J, \mu_x \in L^2 (0, T_0; L^1 \Omega)\). The functions \(J, \mu,\) and \(\mu_x\) take the form

\[
J = h^3 h_{xxx} - h_x h^{-1} - h^2 \left( h_x c_x^2 + 2 h c_x c_{xx} \right),
\]

\[
\mu = c^3 - c - h^{-1} (h_x c_x + h c_{xx}),
\]

and

\[
\mu_x = (3c^2 - 1) c_x + h^{-2} h_x^2 c_x - h^{-1} h_x c_{xx} - h^{-1} h_x c_{xx} - c_{xxx},
\]

respectively. We make use of the following observations:

- The function \(h(x, t)\) is bounded from above and below,

\[
0 < h_{\text{min}} \leq h(x, t) \leq h_{\text{max}} < \infty;
\]

we also have the boundedness of \(c(x, t), \|c\|_{\infty} < \infty\).

- Since \(\mu_x \in L^2 (0, T_0; L^2 \Omega)\), it follows that \(\mu \in L^2 (0, T_0; L^1 \Omega)\), by Poincaré’s inequality.

- From this it follows that \(c_x h_x + h c_{xx}\) is in the class \(L^2 (0, T_0; L^2 \Omega)\).

- Given the inequality

\[
\|h \mu c_x\|_2^2 \leq h_{\text{max}} \|\mu\|_\infty^2 \|c_x\|_2^2 \leq h_{\text{max}} \|c_x\|_2^2 \left[ \frac{1}{\sqrt{L}} \|\mu\|_2 + \sqrt{L} \|\mu_x\|_2 \right]^2,
\]

we have the result \(h_x c_x^2 + h c_x c_{xx} \in L^2 (0, T_0; L^2 \Omega)\).

- Similarly, since \(\int_0^{T_0} \|h \mu h_x\|_2^2 dt < \infty\), we have the bound \(h_x^2 c_x + h h_x c_{xx} \in L^2 (0, T_0; L^2 \Omega)\).
Using these facts, and relegating the details to Appendix B, it is possible to show that $c_{xx}$ is in $L^2(0, T_0; L^1(\Omega))$, from which follows the result $h_{xxx}, c_{xxx} \in L^1(0, T_0; L^1(\Omega))$. These results give rise to further bounds, namely $c_{xx} \in L^2(0, T_0; L^2(\Omega))$, and $\int_0^{T_0} dt \|h_x^2c_x\|_2 < \infty$, whence $h_{xx} \in L^2(0, T_0; L^2(\Omega))$. Using this collection of bounds, we obtain

$$ h, c \in L^1(0, T_0; W^{3,1}(\Omega)), $$

and hence finally,

$$ h, c \in L^2(0, T_0; W^{2,2}(\Omega)). $$

Thus, the solution $(h, c)$ belongs to the following regularity class:

$$ (h, c) \in L^\infty(0, T_0; W^{1,2}(\Omega)) \cap L^2(0, T_0; W^{3,2}(\Omega)) \cap C^{\frac{1}{2}, \frac{1}{2}} (\Omega \times [0, T_0]). \quad (33) $$

Extra regularity is obtained by writing the equation pair as

$$ \frac{\partial h}{\partial t} + h^3 h_{xxxx} = -3h^2 h_x h_{xxx} + \varphi_1 + \varphi_2 \equiv \varphi (x, t), $$

$$ \frac{\partial c}{\partial t} + c_{xxxx} = -2\frac{h}{h_x} c_{xxx} + \psi_1 + \psi_2 \equiv \psi (x, t), $$

where

$$ \int_0^\tau dt \|\varphi_1\|_2 \leq \sup_{[0, \tau]} \|c_{xx}\|_2 \int_0^\tau dt |\nu_1|, \quad \nu_1 \in L^2([0, \tau]), $$

$$ \int_0^\tau dt \|\psi_1\|_2 \leq \sup_{[0, \tau]} \|c_{xx}\|_2 \int_0^\tau dt |\nu_2|, \quad \nu_2 \in L^2([0, \tau]), $$

for any $\tau \in (0, T_0]$, and where $\varphi_2$ and $\psi_2$ belong to the class $L^2(0, T_0; L^2(\Omega))$. Here we have used the form of the concentration equation given by Eq. (25). By multiplying the height and concentration equations by $h_{xxxx}$ and $c_{xxxx}$ respectively, and by integrating over space and time, it is readily shown that

$$ (h, c) \in L^\infty(0, T_0; W^{2,2}(\Omega)), $$

and hence

$$ (\varphi, \psi) \in L^2(0, T_0; L^2(\Omega)), $$

from which follows the regularity result

$$ (h, c) \in L^\infty(0, T_0; W^{2,2}(\Omega)) \cap L^2(0, T_0; W^{4,2}(\Omega)) \cap C^{\frac{3}{2}, \frac{1}{2}} (\Omega \times [0, T_0]). \quad (34) $$
G. Uniqueness of solutions

Let us consider two solution pairs \((h, c)\) and \((h', c')\) and form the difference \((\delta h, \delta c) = (h - h', c - c')\). Given the initial conditions \((\delta c(x, 0), \delta h(x, 0)) = (0, 0)\), we show that \((\delta h, \delta c) = (0, 0)\) for all time, that is, that the solution we have constructed is unique. We observe that that the equation for the difference \(\delta c\) can be written in the form

\[
\frac{\partial}{\partial t} \delta c + \frac{\partial^4}{\partial x^4} \delta c = \delta \varphi(x, t),
\]

where \(\delta \varphi(x, t) \in L^2(0, T_0; L^2(\Omega))\), and where \(\delta \varphi(\delta c = 0) = 0\). Using semigroup theory \([29]\), we find that Eq. (35) has a unique solution. Since \(\delta c = 0\) satisfies the equation for \(\delta c\) can be written in the form

\[
\frac{\partial}{\partial t} \delta c + \frac{\partial^4}{\partial x^4} \delta c = \delta \varphi(x, t),
\]

where \(\delta \varphi(x, t) \in L^2(0, T_0; L^2(\Omega))\), and where \(\delta \varphi(\delta c = 0) = 0\). Using semigroup theory \([29]\), we find that Eq. (35) has a unique solution. Since \(\delta c = 0\) satisfies \(\delta \varphi(\delta c = 0) = 0\), and since \(\delta c = 0\), it follows that \(\delta c = 0\) for all times \(t \in [0, T_0]\).

It is now possible to formulate an equation for the difference \(\delta h\) by subtracting the evolution equations of \(h\) and \(h'\) from one another, mindful that \(\delta c = 0\). We multiply the resulting equation by \(\delta h_{xx}\), integrate over space, and obtain after using inequalities (see Appendix C)

\[
2 \kappa \sup_{\tau \in [0, T]} \|\delta h_x\|_2^2(\tau) \leq \sup_{\tau \in [0, T]} \|\delta h_x\|_2^2(\tau) \int_0^T dt \left( h^{-1} + h'^2 c_x^2 \right)^2 + \kappa_P \sup_{\tau \in [0, T]} \|\delta h_x\|_2^2(\tau) \int_0^T dt \left( \left\| h''_{xxx} + 4 c_x c_{xx} \right\|_2^2 + \left\| h_x' + h_x c_x^2 \right\|_2^2 \right),
\]

where \(\kappa\) and \(\kappa_P\) are numerical constants. Using the results of Sec. III F, it is readily shown that \(h^{-1} + h'^2 c_x^2 \in L^2(0, T; L^\infty(\Omega))\), and that the functions \(h''_{xxx} + 4 c_x c_{xx}\) and \(h_x' + h_x c_x^2\) belong to the class \(L^2(0, T; L^2(\Omega))\). By choosing \(T\) sufficiently small, it is possible to impose the inequality

\[
\frac{1}{2\kappa} \left[ \int_0^T dt \left( h^{-1} + h'^2 c_x^2 \right)^2 + \kappa_P \int_0^T dt \left( \left\| h''_{xxx} + 4 c_x c_{xx} \right\|_2^2 + \left\| h_x' + h_x c_x^2 \right\|_2^2 \right) \right] < 1,
\]

which in turn forces \(\sup_{\tau \in [0, T]} \|\delta h_x\|_2^2 = 0\), and hence the solution is unique.

IV. PARAMETRIC DEPENDENCE OF THE HEIGHT DIP

In this section we perform numerical simulations of the equations \([18]\) and focus on one feature of the equations: the drop in the free-surface height at the boundary between binary fluid domains. Our results in Sec. III gave a rigorous upper bound for the magnitude of this height dip, as a function of the problem parameters, and we compare this estimate with numerical solutions.
We perform numerical simulations of the full equations (18), with initial data comprising a perturbation away from the unstable steady state \((h, c) = (1, 0)\). The free surface and concentration evolve to an equilibrium state where the salient feature is the formation of domains (intervals where \(c \approx \pm 1\)) that are separated by smooth transition regions, across which the free surface dips below its average value. Since we are interested in this characteristic asymptotic feature of phase separation, we shift our focus instead to the steady version from Eq. (18) obtained by setting \(\partial_t = J = \mu_x = 0\). For this reason also, we consider only the repulsive Van der Waals force, for which the interaction potential is given by \(\phi = -|A|h^{-3}\). We then solve the boundary-value problem

\[
\frac{1}{C} \frac{\partial^2 h}{\partial x^2} = |A|C_n^2 \left(1 - \frac{1}{h^3}\right) + r \left[ \frac{1}{4} \left(c^2 - 1\right)^2 + \frac{1}{2} \left(\frac{\partial c}{\partial x}\right)^2 \right], \tag{36a}
\]

\[
\frac{\partial^2 c}{\partial x^2} = c^3 - c - \frac{1}{h} \frac{\partial h}{\partial x} \frac{\partial c}{\partial x}, \tag{36b}
\]

where now the domain is infinite and the boundary conditions are \(h(\pm\infty) = 1\), and \(\mu(\pm\infty) = 0\). We have rescaled lengths by \(C_n\). In Fig. 1 we present numerical solutions exhibiting the dependence of the solutions on the parameters in Eq. (36).

FIG. 1: Equilibrium solutions of Eq. (36) for \(C = C_n^2|A| = 1\) and \(r = 0.1, 1, 10, 50\). In (a) the valley deepens with increasing \(r\) although the film never ruptures, while in (b) the front steepens with increasing \(r\). (From Ó Náraigh and Thiffeault [25].)

As before, the free-surface height possesses peaks and valleys, where the valleys occur in the concentration field’s transition zone. These profiles are qualitatively similar to
the results obtained in experiments on thin binary films \[9, 30, 31\]. While the valley increases in depth for large \(r\), rupture never takes place, in agreement with the theory.

\[
\begin{align*}
F_{\text{cap}} &= -rh^{-1}(hc_x^2)_x, \\
F_{\text{VdW}} &= |A|(h^{-3})_x.
\end{align*}
\]

These forces have opposite sign: the Van der Waals force inhibits film rupture, while the backreaction promotes film thinning. The thinning of binary films due to capillary or backreaction effects has been documented in experiments \[30\].

We are interested in the magnitude of the dip in the free-surface height, as a function of the problem parameters, and we plot the dependence of \(h_{\text{min}}\) as a function of \(|A|\) and \(r\) in Fig. 3. We compare these results with the estimate for \(h_{\text{min}}\) found in Sec. \[III\]. In terms of the physical parameters of the system, this estimate is

\[
h_{\text{min}} \geq M \equiv \sqrt{2CL(F_0 + F_1|A|)} \left( \frac{e^{4C|A|^{-1}(F_0 + F_1|A|)^2}}{e^{4C|A|^{-1}(F_0 + F_1|A|)^2 - 1}} - 1 \right),
\]

where \(F_1 = \frac{1}{2} \int_\Omega dx \ [h(x, 0)]^{-2} \neq 0\), and \(F_0 = F(0) - F_1\). The function \(M(|A|, C)\) has no explicit \(r\)-dependence: although \(F_0\) depends on \(r\), it is possible to find initial data to remove this dependence. We show a representative plot of \(M(|A|, C)\) in Fig. 4.

Although a comparison between Fig. 3 and Fig. 4 is not exact, since the boundary conditions and domains are different in both cases, we see that the shape of the
FIG. 3: Dependence of dip magnitude $h_{\text{min}}$ on (a) the parameter $|A|$; (b) the parameter $r$. We have set $C = C_n = 1$. In (a) the dip decreases in magnitude with increasing $|A|$, demonstrating the tendency of the Van der Waals force to flatten the free surface, while in (b) the dip increases with increasing $r$, which shows how the backreaction thins the film across transition zones.

bound in Fig. 3 is different from that in Fig. 4. Since the bound in Fig. 3 is obtained from numerical simulations, and is intuitively correct, we conclude that it has the correct shape and that the bound of Fig. 4, while mathematically indispensable, is not sharp enough to be useful in determining the parametric dependence of the dip in free-surface height.

V. CONCLUSIONS

Starting from the Navier–Stokes Cahn–Hilliard equations, we have derived a pair of nonlinear parabolic PDEs that describe the coupled effects of phase separation and free-surface variations in a thin film of binary liquid. Since we are interested in the long-time outcome of the phase separation, we focused on liquids that experience a repulsive Van der Waals force, which tends to inhibit film rupture. Using physical intuition, we identified a decaying energy functional that facilitated analysis of the equations. We have shown that given sufficiently smooth initial data, solutions to the model equations (18) exist in a strong sense, and have certain regularity properties. Our proof follows the method developed in [2]. Central to the analysis is the decaying free energy, and the derivation of a no-rupture condition, which prevents the film from touching down to zero. The no-rupture condition is valid in one spatial dimension.
FIG. 4: A typical plot of $M(|A|, C)$ for $F_0 = F_1 = \frac{1}{2}$ and $C = 1$. This theoretical lower bound has a different shape from those in Fig. 3, which suggests that while $M(|A|, C)$ plays an important role in the analysis of the model equations, it does not capture the physics of film thinning.

only, and thus existence and uniqueness results will not necessarily be obtainable in higher dimensions.

We carried out one-dimensional numerical simulations of the full equations and found that the free-surface height and concentration tend to an equilibrium state. The concentration forms domains; that is, extended regions where $c \approx \pm 1$. The domains are separated by narrow zones where the concentration smoothly transitions between the limiting values $\pm 1$. At the transition zones, the free surface dips below its mean value, a feature of binary thin-film behaviour that is observed in experiments. To study the magnitude of this dip as a function of the problem parameters, we focused on solving the equilibrium version of Eq. (18) as a boundary-value problem. This simplification is carried out without loss of generality, since we have shown that the system tends asymptotically to such a state. We have shown that the magnitude of the dip decreases by increasing the strength of the repulsive Van der Waals force, while the dip depth actually increases by increasing the strength of the backreaction. Thus, in the absence of the Van der Waals force, the film would rupture, preventing the occurrence of the phase separation so characteristic of long-time Cahn–Hilliard dynamics. The film-thinning tendency of the backreaction has been observed experimentally [9, 30, 31]. Simulations involving two lateral directions have elsewhere been carried out by the authors [25], and the qualitative features are similar to those obtained here.

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APPENDIX A

In this appendix we give a list of nonstandard inequalities used in the paper and provide proofs.

1. Let $\phi : \Omega \subset \mathbb{R} \rightarrow \mathbb{R}$ belong to the class $W^{1,2}(\Omega)$. Then the following string of inequalities holds,

$$\sup_{\Omega} |\phi| \leq \frac{1}{L} \|\phi\|_1 + \|\phi_x\|_1 \leq \frac{1}{\sqrt{L}} \|\phi\|_2 + \sqrt{L} \|\phi_x\|_2,$$

\text{(A1)}

\textbf{Proof:} Using the Fundamental Theorem of Calculus, we have

$$\phi(x) = \phi(a) + \int_a^x ds \frac{\partial \phi}{\partial s},$$

for any distinct points $x$ and $a$ in $\Omega$. Since the function $|\phi(x)|$ is continuous on $\Omega$, it has a maximum value in $\Omega$, attained at the point $x_{\text{max}}$. Thus,

$$|\phi(x_{\text{max}})| \equiv \sup_{\Omega} |\phi| \leq |\phi(a)| + \int_a^{x_{\text{max}}} ds \left| \frac{\partial \phi}{\partial s} \right| \leq |\phi(a)| + \int_{\Omega} ds \left| \frac{\partial \phi}{\partial s} \right|.$$ 

Since this is true for any $a \in \Omega$, by integrating over $a$, we obtain the inequality

$$\sup_{\Omega} |\phi| \leq \frac{1}{L} \|\phi\|_1 + \|\phi_x\|_1 \leq \frac{1}{\sqrt{L}} \|\phi\|_2 + \sqrt{L} \|\phi_x\|_2,$$

where the last inequality follows from the monotonicity of norms.

2. Let $\phi : \Omega \subset \mathbb{R} \rightarrow \mathbb{R}$ belong to the class $W^{2,1}(\Omega)$. Then

$$\|\phi_x\|_2^2 \leq L \|\phi_{xx}\|_1^2 + \frac{4}{L} \|u\|_1 \|\phi_{xx}\|_1.$$ \text{(A2)}

\textbf{Proof:} We have the identity $\int \phi_x^2 dx = - \int \phi \phi_{xx}$, true for any function $\phi$ with periodic boundary conditions. Using Hölder’s inequality, this becomes

$$\|\phi_x\|_2^2 \leq \|\phi\|_\infty \|\phi_{xx}\|_1.$$

Using the relation (A1), this becomes
\[
\|\phi_x\|_2^2 \leq \left( \frac{1}{L} \|\phi\|_1 + \|\phi_x\|_1 \right) \|\phi_{xx}\|_1,
\]
which is a quadratic inequality in \(\|\phi_x\|_2^2\), with solution
\[
\|\phi_x\|_2 \leq \frac{1}{2} \left( \sqrt{L} \|\phi_{xx}\|_1 + \sqrt{L} \|\phi_{xx}\|_1^2 + 4L^{-1}\|\phi\|_1 \|\phi_{xx}\|_1 \right).
\]
By sacrificing the sharpness of the bound, we obtain a simpler one,
\[
\|\phi_x\|_2^2 \leq L \|\phi_{xx}\|_1^2 + \frac{4}{L} \|\phi\|_1 \|\phi_{xx}\|_1,
\]
as required.

**APPENDIX B**

In this appendix, we fill in the details missing from the discussion of the regularity of solutions in Sec. III F and prove the result
\[
h, c \in L^2 (0, T_0; W^{3,2}(\Omega)).
\]
To begin, we notice that Sec. III C gives rise to the result
\[
\|h_x\|_2, \|c_x\|_2 < \infty,
\]
with time-independent bounds. Thus, using Poincaré’s inequality, it follows that
\[
h, c \in W^{1,2}(\Omega) \text{ and, moreover,}
\]
\[
\sup_{[0,T_0]} \|h_x\|_2, \sup_{[0,T_0]} \|c_x\|_2 < \infty.
\]
From Sec. III D, it follows that \(J\) and \(\mu_x\) belong to the regularity class \(L^2 (0, T_0; L^2(\Omega))\), and hence \(J, \mu_x \in L^2 (0, T_0; L^1(\Omega))\). The functions \(J, \mu,\) and \(\mu_x\) take the form
\[
J = h^3 h_{xxx} - h_x h^{-1} - h^2 \left( h_x c_x^2 + 2hc_x c_{xx} \right),
\]
\[
\mu = c^3 - c - h^{-1} \left( h_x c_x + hc_{xx} \right),
\]
and
\[
\mu_x = (3c^2 - 1) c_x + h^{-2} h_x^2 c_x - h^{-1} h_{xx} c_x - h^{-1} h_x c_{xx} - c_{xxx}, \quad (B1)
\]
respectively. We make use of the following observations,
The function $h(x,t)$ is bounded from above and below,

$$0 < h_{\min} \leq h(x,t) \leq h_{\max} < \infty,$$

and the boundedness of $c(x,t)$, $\|c\|_\infty < \infty$.

Since $\mu_x \in L^2(0,T_0;L^2(\Omega))$, it follows that $\mu \in L^2(0,T_0;L^2(\Omega))$, by Poincaré’s inequality.

From this it follows that $c_x h_x + hc_{xx}$ is in the class $L^2(0,T_0;L^2(\Omega))$.

Given the inequality

$$\|h \mu c_x\|_2^2 \leq h_{\max} \|\mu\|_\infty^2 \|c_x\|_2^2 \leq h_{\max} \|c_x\|_2^2 \left[ \frac{1}{\sqrt{L}} \|\mu\|_2 + \sqrt{L} \|\mu_x\|_2 \right]^2,$$

we have the result $h_x c^2_x + h c_x c_{xx} \in L^2(0,T_0;L^2(\Omega))$.

Similarly, since $\int_0^{T_0} \|h \mu h_x\|_2^2 dt < \infty$, we have the bound $h^2_x c_x + hh_x c_{xx} \in L^2(0,T_0;L^2(\Omega))$.

Now inspection of $\mu$ shows that $c_{xx}$ is in $L^2(0,T_0;L^1(\Omega))$, from which follows the result $h_{xxx}, c_{xxx} \in L^1(0,T_0;L^1(\Omega))$. By repeating the same argument, we find that $c_{xx} \in L^2(0,T_0;L^2(\Omega))$. We also have the result that $\|h^2_x c_x\|_2$ is almost always bounded. To show that $h_{xx} \in L^2(0,T_0;L^2(\Omega))$, we take the evolution equation for $h(x,t)$, multiply it by $h$, and integrate, obtaining

$$-\frac{1}{2} \frac{d}{dt} \int_\Omega dx h^2 - 3 \int_\Omega dx h^2 h_x^2 h_{xx} - \int_\Omega dx h_x J_0 = \int_\Omega dx h^3 h_{xx}^2,$$

where $J_0 = h_x h^{-1} + 2h^3 c_x c_{xx} + h^2 h_x c_{xx}^2$. The time-integral of the first term on the left-hand side of this equation is obviously bounded in time. Let us examine the time-integral of the second term,

$$\int_0^{T_0} dt \int_\Omega dx h^2 h_x^2 h_{xx} \leq h_{\max}^2 \sup_{[0,T_0]} \|h_x\|_2^2 \int_0^{T_0} dt \|h_{xx}\|_\infty$$

$$\leq h_{\max}^2 \sup_{[0,T_0]} \|h_x\|_2^2 \int_0^{T_0} dt \left( L^{-1} \|h_{xx}\|_1 + \|h_{xxx}\|_1 \right) < \infty.$$

The third term on the left-hand side is dispatched in a similar way, so that $\int_0^{T_0} dt \|h_{xx}\|_2^2 < \infty$. We have now shown that $h,c \in L^2(0,T_0;W^{2,2}(\Omega))$. Using
this result, together with the previous facts gathered together in this appendix, it is readily shown that \( h, c \in L^1 (0, T_0; W^{3,1}(\Omega)) \), and it follows that

\[
h, c \in L^2 (0, T_0; W^{3,2}(\Omega)).
\]

To see this more clearly, we show that \( c_{xxx} \) is in the above class. For example, consider a typical term in \( \mu_x \),

\[
\int_0^{T_0} dt \int_{\Omega} dx h_x^2 c_{xx}^2 \leq \sup_{[0,T_0]} \| h_x \|_2^2 \int_0^{T_0} dt \left( L^{-1} \| c_{xx} \|_1 + \| c_{xxx} \|_1 \right)^2 < \infty,
\]

which implies \( h_x c_{xx} \in L^2 (0, T_0; L^2(\Omega)) \). This bound, together with \( h \mu h_x \in L^2 (0, T_0; L^2(\Omega)) \), implies \( h_x c_x \in L^2 (0, T_0; L^2(\Omega)) \). Gathering all these results, we have

\[
\mu_x, h_x c_{xx}, h_x^2 c_x, h_{xxx} c_x \in L^2 (0, T_0; L^2(\Omega))
\]

From (B1), it follows that \( c_{xxx} \) is in this class as well. A similar argument holds for \( h_{xxx} \). Thus, the solution \( (h, c) \) belongs to the regularity class

\[
(h, c) \in L^\infty (0, T_0; W^{1,2}(\Omega)) \cap L^2 (0, T_0; W^{3,2}(\Omega)) \cap C^{1,\frac{1}{2}} (\Omega \times [0, T_0]).
\]

### APPENDIX C

In this appendix, we describe in full the proof of the uniqueness of solutions sketched in Sec. III G. We consider two solution pairs \( (h, c) \) and \( (h', c') \) and form the difference \( (\delta h, \delta c) = (h - h', c - c') \). Given the initial conditions \( (\delta c(x, 0), \delta h(x, 0)) = (0, 0) \), we show that \( (\delta h, \delta c) = (0, 0) \) for all time, that is, that the solution we have constructed is unique. We observe that that the equation for the difference \( \delta c \) can be written in the form

\[
\frac{\partial}{\partial t} \delta c + \frac{\partial^4}{\partial x^4} \delta c = \delta \varphi (x, t),
\]

where \( \delta \varphi (x, t) \in L^2 (0, T_0; L^2(\Omega)) \), and where \( \delta \varphi (\delta c = 0) = 0 \). As discussed previously, this equation has a unique solution, given smooth initial data. Since \( \delta c = 0 \) satisfies Eq. (C1), and since \( \delta c (x, 0) = 0 \), it follows that \( \delta c = 0 \) for all times \( t \in [0, T_0] \).

It is now possible to formulate an equation for the difference \( \delta h \) by subtracting the evolution equations of \( h \) and \( h' \) from one another, mindful that \( \delta c = 0 \). We multiply
the resulting equation by \( \delta h_{xx} \) and integrate over space to obtain

\[
\frac{1}{2} \frac{d}{dt} \int_\Omega dx \, \delta h_x^2 + \int_\Omega dx \, h^3 \delta h_{xxx} = \int_\Omega dx \, \delta h_{xxx} \, \delta h_x (h^{-1} + h'^2 c_x^2) \\
- \int_\Omega dx \, (h^3 - h'^3) (h'_{xxx} + 4c_x c_{xx}) \, \delta h_{xxx} \\
+ \int_\Omega dx \, \delta h_{xxx} \, \left[ h_x' (h^{-1} - h'^{-1}) + h_x c_x^2 (h^2 - h'^2) \right].
\]

Using the lower bound on \( h(x,t) \geq h_{\text{min}} > 0 \) and Young’s first inequality, this equation is transformed into an inequality,

\[
\frac{1}{2} \frac{d}{dt} \int_\Omega dx \, \delta h_x^2 + h_{\text{min}}^3 \int_\Omega dx \, \delta h_{xxx} \leq \kappa_1 \int_\Omega dx \, \delta h_{xxx} + \frac{1}{4\kappa_1} \int_\Omega dx \, \delta h_x^2 \, (h^{-1} + h'^2 c_x^2)^2 \\
+ \kappa_2 \int_\Omega dx \, \delta h_{xxx} + \frac{1}{4\kappa_2} \int_\Omega dx \, (h^3 - h'^3)^2 \, (h'_{xxx} + 4c_x c_{xx})^2 \\
+ \kappa_3 \int_\Omega dx \, \delta h_{xxx} + \frac{1}{4\kappa_3} \int_\Omega dx \, [h_x' (h^{-1} - h'^{-1}) + h_x c_x^2 (h^2 - h'^2)]^2,
\]

where \( \kappa_1, \kappa_2, \) and \( \kappa_3 \) are arbitrary positive constants. By choosing \( \kappa_1 + \kappa_2 + \kappa_3 = h_{\text{min}}^3 \), the inequality simplifies to

\[
2\kappa \frac{d}{dt} \int_\Omega dx \, \delta h_x^2 \leq \int_\Omega dx \, \delta h_x^2 \, (h^{-1} + h'^2 c_x^2)^2 \\
+ \int_\Omega dx \, \delta h_x^2 \left[ (h'_{xxx} + 4c_x c_{xx})^2 + (h_x' + h_x c_x^2)^2 \right],
\]

where \( \kappa \) is another positive constant. We integrate over the time interval \([0, T]\), and use the fact that \( ||\delta h_x||_2(0) = 0 \) to obtain

\[
2\kappa \sup_{\tau \in [0,T]} ||\delta h_x||_2^2 (\tau) \leq \sup_{\tau \in [0,T]} ||\delta h_x||_2^2 (\tau) \int_0^T dt \, ||h^{-1} + h'^2 c_x^2||_\infty^2 \\
+ \sup_{\tau \in [0,T]} ||\delta h||_\infty^2 (\tau) \int_0^T dt \int_\Omega dx \, [h'_{xxx} + 4c_x c_{xx}]^2 + (h_x' + h_x c_x^2)^2].
\]

The Poincaré inequality can be combined with the one-dimensional differential inequalities discussed in Appendix A to yield the relation \( ||f||_\infty \leq \kappa_P ||f_x||_2 \), where \( f \) is some mean-zero function and \( \kappa_P \) is an \( f \)-independent constant. We therefore arrive
at
\[
2\kappa \sup_{\tau \in [0,T]} \| \delta h_x \|_2^2 (\tau) \leq \sup_{\tau \in [0,T]} \| \delta h_x \|_2^2 (\tau) \int_0^T dt \| h^{-1} + h'^2 c^2_x \|_{\infty}^2 \\
+ \kappa_p^2 \sup_{\tau \in [0,T]} \| \delta h_x \|_2^2 (\tau) \int_0^T dt \left( \| h'_{xxx} + 4c_x c_{xx} \|_2^2 + \| h'_x + h_x c^2_x \|_2^2 \right).
\]

Using the results of Sec. III F, it is readily shown that \( h^{-1} + h'^2 c^2_x \in L^2 (0, T; L^\infty (\Omega)) \), and that the functions \( h'_{xxx} + 4c_x c_{xx} \) and \( h'_x + h_x c^2_x \) belong to the class \( L^2 (0, T; L^2 (\Omega)) \). By choosing \( T \) sufficiently small, it is possible to impose
\[
\frac{1}{2\kappa} \left[ \int_0^T dt \| h^{-1} + h'^2 c^2_x \|_{\infty}^2 + \kappa_p^2 \int_0^T dt \left( \| h'_{xxx} + 4c_x c_{xx} \|_2^2 + \| h'_x + h_x c^2_x \|_2^2 \right) \right] < 1,
\]
which in turn forces \( \sup_{\tau \in [0,T]} \| \delta h_x \|_2^2 = 0 \), and hence the solution is unique.

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