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Stationary solutions for two-layer lubrication equations

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Abstract

We investigate stationary solutions of flows of thin liquid bilayers in an energetic formulation which is motivated by the gradient flow structure of its lubrication approximation. The corresponding energy favors the liquid substrate to be only partially covered by the upper liquid. This is expressed by a negative spreading coefficient which arises from an intermolecular potential combining attractive and repulsive forces and leads to an ultra-thin layer of thickness ε. For the corresponding lubrication models existence of stationary solutions is proven. In the limit $\varepsilon \to 0$ matched asymptotic analysis is applied to derive sharp-interface models and the corresponding contact angles, i.e. the Neumann triangle. In addition we use Γ-convergence and derive the equivalent sharp-interface models rigorously in this limit. For the resulting model existence and uniqueness of energetic minimizers are proven. The minimizers agree with solutions obtained by matched asymptotics.

1 Introduction

Understanding stability and dewetting behaviour of thin liquid (polymer) films coating a solid or liquid substrate is very important for many applications in technology and nature, ranging from tear films to organic solar cell production and numerous applications in the semiconductor industry. The films we consider have typical thicknesses of a few hundred nanometers and are unstable due to intermolecular forces. These forces consist of a combination of destabilising van-der-Waals and stabilising Born-repulsion potentials. The corresponding flow leads to formation of holes, where the liquid layer has receded from the substrate up-to an ultra-thin layer. The thickness of that ultra-thin layer is given by the minimum of the interfacial potential. After an initial rupture singularity we encounter a complex dewetting scenario, where holes grow further and their trailing rims merge into a polygonal network which finally decays into droplets. These droplets evolve slowly towards a global minimal energy state. Reviews on existing theory and experiments can be found in [37, 10].

Experimental studies on dewetting from solid substrates exhibiting the scenario described above and can be found for example in [47, 45] and more references in the reviews [13, 20, 17]. For liquid-liquid dewetting a number of similar experimental investigations on have been conducted by the groups [46, 28, 48, 49]. These works include studies of break-up and hole growth in a liquid-liquid system, dewetting dynamics and equilibrium contact angles of droplets. The standard system consists of liquid polystyrene (PS) on a polymethylmethacrylate (PMMA) substrate.

Compared to dewetting on a solid substrate, the mathematical description of liquid/liquid flows has a number of new challenges. For instance, the substrate interface itself is deformed under the action of the liquid flow. Furthermore the contact line is fixed by two angle conditions instead of one, i.e. Youngs law is replaced by the Neumann triangle construction [36]. Still one might expect that some of the mathematical analysis for solid substrates can be carried over to liquid substrates.

Concerning dewetting for liquid-liquid systems one of the pioneering theoretical studies is the paper by Brochard-Wyart et al. [7], where various dewetting regimes are derived and analysed. Besides this early work, there have been studies considering stability of liquid-liquid systems by Danov et al. [12], Pototsky et al. [41], Golovin & Fisher [16]. Stationary states and the dynamics towards stationary states were investigated by Pototsky et al. in [42], by Craster & Matar in [9] and by Bandyopadhyay & Sharma in [2] and Bandyopadhyay et al. [1]. Kriegsmann & Miksis [27] computed quasi-stationary states of gravity-driven liquid droplets on a inclined liquid substrate. Interestingly, direct comparisons of theoretical results with experiments regarding for example the morphology of the interfaces, as performed in [26], or equilibrium contact angles of the Neumann triangle, as in [48], are incomplete.

Apart from the numerical analysis, mathematical theory in this field is still largely open. In that paper we aim to extend the existing theory for liquid droplets on a solid substrate to the situation on a liquid substrate. Note that regarding the stationary states and coarsening of the droplets on solid substrates, Bertozzi et al. [4] showed existence of smooth global solutions for positive data with bounded energy. In addition they prove existence of global minimizers and determine a family of positive periodic solutions for admissible intermolecular potentials and investigate their linear stability. Similarly, the linear stability of the stationary solutions for the thin film equation with Neumann boundary conditions or periodic boundary conditions was investigated by Laugesen & Pugh [30] and Kitavtsev et al. [24]. In a recent paper by Zhang [51], convergence to stationary solutions of the one-dimensional degenerate parabolic PDE and the number of stationary states is investigated. Rump et al. [38] also prove existence and uniqueness (up to translation) of mesoscopic droplet solutions.

The liquid-liquid intermolecular potential is of the form

$$\phi(h_2 - h_1) = \frac{\phi_*}{\ell - n}\left[\ell\left(\frac{h_*}{h_2 - h_1}\right)^n - n\left(\frac{h_*}{h_2 - h_1}\right)^\ell\right]. \tag{1.1}$$

where h_1 is the height of the liquid-liquid interface, h_2 the height of the free surface and its minimal value $\phi_* < 0$ is attained at h_*. Such potentials are widely used in the literature, e.g. [37]. A choice that is related to the standard Lennard-Jones potential and typically used in experiments is $(n, \ell) = (2, 8)$, see e.g. [45] for a liquid PS film dewetting from a SiO/Si substrate.

The Neumann triangle construction for contact angles at triple junction is now replaced by properties of approximate contact angles resulting from the particular structure of the free surface energy ϕ. One of the main tasks of this study is to derive a sharp-interface model, where the thickness of the ultra-thin film h_* deceases to zero and thereby obtain appropriate contact angles for the corresponding Neumann triangle.

The content of the paper is as follows. First we introduce the problem formulation and model. Then, in section 3 we begin our analysis with a straightforward generalisation of the existence proof for stationary solutions by Bertozzi et al. [4]. With the appropriate energy functional for the two-layer system and Neumann boundary conditions, we show existence of smooth stationary solutions as well as existence of a global minimizer for the steady state problem. Here it turns out to be advantegous to work with the difference $h = h_2 - h_1$.

In section 4 we derive expressions for the equilibrium contact angles of these droplet solutions. We pursue this with a similar approach as for example in [34], using matched asymptotic expansions in the limit as the minimum thickness h_* approaches zero. As a result, we obtain the equilibrium contact angles of the corresponding Neumann triangle and a corresponding sharp-interface model. Interestingly, we observe that, while the equilibrium contact angle is easy to obtain, in order to complete the asymptotic argument, logarithmic switch-back terms enter the derivation. In retrospect, these terms should also appear in the matched asymptotic derivation for the limiting problem of a droplet on a solid substrate.

In section 5 we study the limit $h_* \to 0$ within the framework of Γ-convergence and obtain a sharp-interface problem. For that problem we compute the Euler-Lagrange equations, from which one can immediately read-off the contact angles. Moreover, we rigorously show existence and uniqueness of minimizers using the rearrangement inequality, a concept which was used by Otto et al. [38] in the context of mesoscopic droplets on a solid substrate.

In the Conclusion and Outlook section we compare our results using the approach of matched asymptotics and Γ-convergence and discuss how we can extend our analysis of liquid-liquid systems to the dynamic regimes of dewetting processes, in particular concerning hole growth and rim dynamics but also film rupture and coarsening dynamics of quasistationary droplets.

2 Formulation

We consider a layered system of two viscous, immiscible, Newtonian liquids with negative spreading coefficient ϕ_*. Let the layered system live in the two phases Ω_1 and Ω_2 defined by

$$\Omega_1(t) = \{(x,y,z) \in \mathbb{R}^3 : 0 \le z < h_1(t,x,y)\}, \tag{2.1a}$$
$$\Omega_2(t) = \{(x,y,z) \in \mathbb{R}^3 : h_1(t,x,y) \le z < h_2(t,x,y)\}, \tag{2.1b}$$

for all $t > 0$ as sketched below in figure 1.

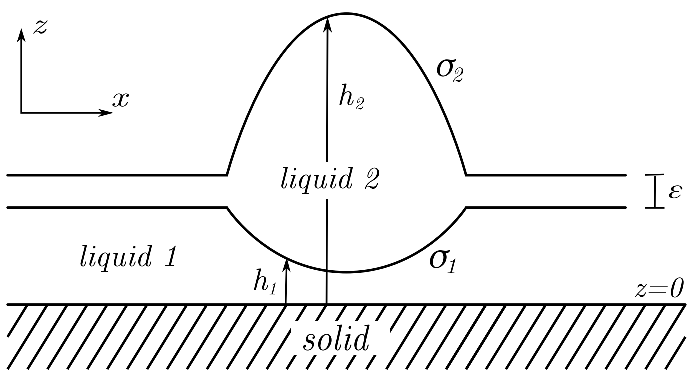

Figure 1: Sketch of stationary droplet on a liquid layer

A typical choice of liquids is PS for the liquid substrate Ω_1 and PMMA as the upper liquid Ω_2. Since the liquids are very viscous the flow in each phase Ω_i in (2.1) is governed by the Stokes equation and the continuity equation

$$-\nabla\cdot\big(-p_i\mathbb{I}+\mu_i(\nabla\mathbf{u}_i+\nabla\mathbf{u}_i^\top)\big)=\mathbf{f}_i, \tag{2.2a}$$

$$\nabla\cdot\mathbf{u}_i=0, \tag{2.2b}$$

together with a kinematic condition at each free boundaries $z=h_i$, i.e.

$$(\mathbf{e}_z\partial_t h_i-\mathbf{u}_i)\cdot\mathbf{n}_i=0, \tag{2.3}$$

with outer normal $\mathbf{n}_i$. At the solid substrate a no-slip and an impermeability condition are imposed and at the liquid-liquid interface the velocity is continuous, i.e. $\mathbf{u}_2=\mathbf{u}_1$. The dewetting process is driven by the intermolecular potential of the upper liquid layer, i.e. $\mathbf{f}_2=-\nabla\phi$. We assume that the thickness of the lower layer is sufficiently thick, so other contributions to the intermolecular potential are negligible, i.e. $\mathbf{f}_1=0$.

In addition we assume that the the ratio $\varepsilon_\ell=H/L$ of the vertical to horizontal length scales is always small and the two-layer system can be approximated by a lubrication model. We denote by L the length scale of the typical horizontal width and $H=h_{\max}$ the maximum of the difference h_2-h_1, see the sketch in figure 1.

Detailed derivations of lubrication models for liquid-liquid systems are given for example in [27], [41] or in [21]. For the convenience of the reader we note here the choice of non-dimensional variable and parameters, where the non-dimensional horizontal and vertical coordinates are given by $\tilde{x}=x/L$, $\tilde{y}=y/L$ and $\tilde{z}=z/h_{\max}$, respectively, and $\tilde{h}=h/h_{\max}$. The non-dimensional pressures $\tilde{p}_i=p_i/P$ and the derivative of the non-dimensional intermolecular potential $\phi_\varepsilon'=\phi'/P$ are scaled such that $P=\phi_* n\ell/\big((\ell-n)h_{\max}\big)$ and hence

$$\phi_\varepsilon{}'(\tilde{h})=\frac{1}{\varepsilon}\left[\left(\frac{\varepsilon}{\tilde{h}}\right)^{n+1}-\left(\frac{\varepsilon}{\tilde{h}}\right)^{\ell+1}\right] \tag{2.4}$$

attains the minimal value $\min\phi_\varepsilon=-1$ at $\tilde{h}=\varepsilon$, where $\varepsilon=h_*/h_{\max}$ is the non-dimensional thickness of the ultra-thin film. For the dynamic problem, the velocities are scaled with the characteristic horizontal velocity of the dewetting upper layer, such that for the non-dimensional horizontal and vertical velocities we have $\tilde{u}=u/U$, $\tilde{v}=v/U$ and $\tilde{w}=w/\varepsilon_\ell U$, respectively, with $U=\varepsilon_\ell^3\sigma_2/\mu_2$, and the non-dimensional time $\tilde{t}=(U/L)t$.

For the remainder of this paper it is convenient to introduce the ratios $\sigma=\sigma_1/\sigma_2$ and $\mu=\mu_1/\mu_2$ of surface tensions and viscosities, respectively, and drop all the "$\sim$". Within this approximation the normal and tangential stress conditions at the free surface h_2 and at the free liquid-liquid interface h_1 yield the expressions for the pressures p_1 and p_2

$$p_1=-\sigma\Delta h_1-\phi_\varepsilon'(h_2-h_1), \tag{2.5a}$$

$$p_2=-\Delta h_2+\phi_\varepsilon'(h_2-h_1), \tag{2.5b}$$

respectively. Under these assumptions the coupled system of nonlinear fourth order partial differential equations for the profiles of the free surfaces h_1 and h_2 takes the form

$$\partial_t\mathbf{h}=\nabla\cdot(Q\cdot\nabla\mathbf{p}), \tag{2.6}$$

where $\mathbf{h} = (h_1, h_2)^\top$ is the vector of liquid-liquid interface profile and liquid-air surface profile. The components of the vector $\mathbf{p} = (p_1, p_2)^\top$ are the interfacial pressures given in (2.5). The gradient of the pressure vector is multiplied by the mobility matrix Q which is given by

$$Q = \frac{1}{\mu}\begin{bmatrix} \dfrac{h_1^3}{3} & \dfrac{h_1^3}{3} + \dfrac{h_1^2(h_2 - h_1)}{2} \\ \dfrac{h_1^3}{3} + \dfrac{h_1^2(h_2 - h_1)}{2} & \dfrac{\mu}{3}(h_2 - h_1)^3 + h_1 h_2 (h_2 - h_1) + \dfrac{h_1^3}{3} \end{bmatrix}. \tag{2.7}$$

We proceed to first show existence of stationary solution to the system (2.4)-(2.7) with Neumann and Dirichlet boundary conditions on a finite domain $\Omega = (0, L) \subset \mathbb{R}$.

3 Energy functionals and existence of stationary solutions

Consider the two-layer thin film equations (2.5)-(2.7) defined on $\Omega = (0, L) \subset \mathbb{R}$ with Neumann boundary conditions:

$$\partial_x h_1 = \partial_x h_2 = \partial_{xxx} h_1 = \partial_{xxx} h_2 = 0 \ \text{ for } \ x \in \partial\Omega. \tag{3.1}$$

The energy functional associated to the gradient flow of the lubrication equation is given by

$$E_\varepsilon(h_1, h_2) = \int_0^L \left[\frac{\sigma}{2}\left|\partial_x h_1\right|^2 + \frac{1}{2}\left|\partial_x h_2\right|^2 + \phi_\varepsilon(h_2 - h_1)\right] \mathrm{d}x \tag{3.2}$$

where the potential function ϕ_ε is given as in (2.4) with $(n, \ell) = (2, 8)$. The relation to the lubrication equations is $p_i = \delta E_\varepsilon / \delta h_i$. From (2.5)-(2.7) and (3.1) conservation of mass follows

$$\int_\Omega h_1(t, x)\,\mathrm{d}x = m_1, \tag{3.3a}$$

$$\int_\Omega \left(h_2(t, x) - h_1(t, x)\right)\,\mathrm{d}x = m_2 \ \text{ for all } \ t > 0, \tag{3.3b}$$

where m_1 and m_2 are positive constants. Any stationary solution of (2.5)-(2.7) considered with (3.1) satisfies

$$0 = Q \cdot \partial_x \mathbf{p} \tag{3.4}$$

in Ω, where the mobility matrix Q is not singular i.e. $\det Q \neq 0$ for all $h_1,\ h_2 - h_1 > 0$. Therefore, one obtains that any positive stationary solution of (2.5)-(2.7) satisfies $\partial_x p_1 = \partial_x p_2 = 0$ in Ω. This in turn is equivalent to

$$\sigma \partial_{xx} h_1 = -\phi_\epsilon'(h_2 - h_1) - \lambda_2 + \lambda_1, \tag{3.5a}$$

$$\partial_{xx} h_2 = \phi_\epsilon'(h_2 - h_1) - \lambda_1, \tag{3.5b}$$

where constants λ_2 and λ_1 are Lagrange multipliers associated with conservation of mass (3.3a) and (3.3b), respectively. To solve (3.5) let us consider the equation for the difference

$$h(t, x) = h_2(t, x) - h_1(t, x)$$

which reads as follows

$$\partial_{xx}h = \frac{\sigma+1}{\sigma}\phi'_\epsilon(h) + \frac{1}{\sigma}\lambda_2 - \frac{\sigma+1}{\sigma}\lambda_1. \tag{3.6}$$

For brevity we set

$$P := -\frac{1}{\sigma}\lambda_2 + \frac{\sigma+1}{\sigma}\lambda_1.$$

According to [4] for positive P, there exists a so called *droplet* solution $\bar{h}$ to (3.6) satisfying boundary conditions (3.1), such that $\bar{h}(y + L/2)$ is an even function and monotone decreasing for $y \in (0, L/2)$.

For this solution the asymptotics and main properties are derived in the next section, here we consider $\bar{h}$ as a known analytical function and integrate (3.5) two times w.r.t. x. We then obtain a solution to (3.5) with (3.1) in the form

$$h_1 = -\frac{1}{\sigma+1}\bar{h} - \frac{1}{2}\frac{\lambda_2}{\sigma+1}x^2 + Cx + C_1, \tag{3.7a}$$

$$h_2 = \frac{\sigma}{\sigma+1}\bar{h} - \frac{1}{2}\frac{\lambda_2}{\sigma+1}x^2 + Cx + C_1. \tag{3.7b}$$

Using now again (3.1) one obtains that $\lambda_2 = 0,\ C = 0$ and

$$h_1 = -\frac{1}{\sigma+1}\bar{h} + C_1, \tag{3.8a}$$

$$h_2 = \frac{\sigma}{\sigma+1}\bar{h} + C_1. \tag{3.8b}$$

The additive constant C_1 and the remaining Lagrange multiplier are determined from the conservation of masses (3.3a) and (3.3b), respectively. We conclude that the solution (3.8) is given by combination of two symmetric droplets with constant outer layer. The next theorem establishes existence of a global minimizer to the energy functional (3.2) and shows that it satisfies (3.5) with (3.1).

Theorem 3.1. *Let Ω be a bounded domain of class $C^{0,1}$ in $\mathbb{R}^d,\ d \geq 1$ and let $\mathbf{m} = (m_1, m_2)$ with $m_1, m_2 > 0$. Then a global minimizer of $E_\epsilon(\cdot,\cdot)$ defined in (3.2) exists in the class*

$$X_{\mathbf{m}} := \left\{(h_1, h_2) \in H^1(\Omega)^2 : m_1 = \int_\Omega h_1,\ m_2 = \int_\Omega (h_2 - h_1),\ h_2 \geq h_1\right\}, \tag{3.9}$$

For $d = 1$ and $\Omega = (0, L)$ the function $h_2 - h_1$ is strictly positive and (h_1, h_2) are smooth solutions to the ODE system (3.5) with (3.1) and

$$\lambda_1 = \frac{1}{L}\int_\Omega \phi'_\epsilon(h_2 - h_1)\,\mathrm{d}x, \qquad \lambda_2 = 0. \tag{3.10}$$

Proof. The proof proceeds very analogously to one of Theorem 2-3 in [4] using direct methods of the calculus of variations. However, for the convenience of the reader we state it here. Let

$(h_1^k, h_2^k)_{k\in\mathrm{N}}$ be a minimizing sequence which exists since $E_\epsilon(m_1, m_2 + m_1) < \infty$. Observe that $\phi_\epsilon(\cdot)$ is bounded from below by a constant. Hence, a constant C_2 exists such that

$$\int_\Omega |\partial_x h_1^k|^2 + |\partial_x h_2^k|^2 \,\mathrm{d}x \le C_2 \text{ for all } k \in \mathrm{N}. \tag{3.11}$$

Rellich's compactness theorem implies that there is a subsequence (denoted by $(h_1^k, h_2^k)_{k\in\mathrm{N}}$) which converges strongly in $L^2(\Omega)^2$ and pointwise almost everywhere to $(h_1, h_2) \in H^1(\Omega)^2$. By Fatou's lemma, we deduce that also $\phi_\epsilon(h_2 - h_1)$ lies in $L^1(\Omega)$. Using the weak lower semicontinuity of the norm, we obtain

$$E_\epsilon(h_1, h_2) \le \lim\inf_{k\to\infty} E_\epsilon(h_1^k, h_2^k) = \inf_{(\bar h_1, \bar h_2)\in V} E_\epsilon(\bar h_1, \bar h_2).$$

Consequently, (h_1, h_2) is a minimizer and the integrability of $\phi_\epsilon(h_2 - h_1)$ implies that $h_2 - h_1 > 0$ almost everywhere in Ω.

Let now $d = 1$ and $\Omega = (0,\ L)$. The estimate (3.11) implies

$$\int_\Omega |\partial_x(h_2^k - h_1^k)|^2 \,\mathrm{d}x \le C_3.$$

Next, the boundedness of $\int \phi_\epsilon(h_2^k - h_1^k)\,\mathrm{d}x$ and definition of ϕ_ϵ imply a uniform in k bound on $||(h_2^k - h_1^k)^{-4}||_2$. Using this, one can estimate

$$\begin{aligned}\int_0^L |\partial_x((h_2^k - h_1^k)^{-3})|\,\mathrm{d}x &= 3\int_0^L \frac{|\partial_x(h_2^k - h_1^k)|}{(h_2^k - h_1^k)^4}\,\mathrm{d}x \\ &\le 3||\partial_x(h_2^k - h_1^k)||_2\, ||(h_2^k - h_1^k)^{-4}||_2 \le C_4,\end{aligned}$$

where C_4 is constant. Owing to the continuous embedding of $W^{1,3}(0,1)$ into $L^\infty(0,1)$ the strong positivity of $h_2 - h_1$ follows. This in turn implies the differentiability of the function $F_\epsilon(s) := E_\epsilon(h_1 + s\varphi_1, h_2 + s\varphi_2)$ considered with fixed $(\varphi_1, \varphi_2) \in H^1(0,L)^2$ for sufficiently small s. Since (h_1, h_2) is a minimizer, by differentiation of $F_\epsilon(s)$ at $s = 0$, we obtain that

$$\int_0^L (-\sigma\partial_{xx}h_1 - \phi_\epsilon'(h_2 - h_1))\varphi_1 + (-\partial_{xx}h_2 + \phi_\epsilon'(h_2 - h_1))\varphi_2 \,\mathrm{d}x = 0,$$

for all $(\varphi_1, \varphi_2) \in H^1(0,L)^2$ such that

$$\int_0^L \varphi_1 \,\mathrm{d}x = \int_0^L (\varphi_2 - \varphi_1)\,\mathrm{d}x = 0.$$

Without this constraint and using the Lagrangian multipliers which we defined previously one can test with general $(\psi_1, \psi_2) \in H^1(0,L)^2$ and gets

$$\int_0^L \Big[(-\sigma\partial_{xx}h_1 - \partial_{xx}h_2)\psi_1 + (-\partial_{xx}h_2 + \phi_\epsilon')\psi_2\Big]\,\mathrm{d}x - \frac{1}{L}\int_0^L\int_0^L \phi_\epsilon'\,dy\,\psi_2\,\mathrm{d}x = 0.$$

Standard elliptic regularity theory then implies that (h_1, h_2) are smooth solutions to (3.5) considered with (3.1) and (3.10). □

Remark 3.2. *Note, that for Dirichlet boundary conditions we can proceed as follows: Let us impose on system (3.5) the Dirichlet boundary conditions*

$$h_1(0) = h_1(L) = A, \;\; h_2(0) = h_2(L) = B, \tag{3.12}$$

such that

$$B - A = \min_{x\in(0,L)} \bar{h}(x), \tag{3.13}$$

where $\bar{h}$ is the Neumann solution to (3.6) defined above. In this case it follows again that $h_2 - h_1 = \bar{h}$. Consequently, h_1 and h_2 are given by (3.7) with constants $\lambda_1, \lambda_2, C, C_1$ determined uniquely by (3.12) and the conservation of masses (3.3a)-(3.3b). Using (3.13) and asymptotics for $\bar{h}$ one obtains that the leading order of the solution (3.7) as $\varepsilon \to 0$ has now the form

$$h_1(x) = \frac{1}{2}\frac{\lambda_1 - \lambda_2}{\sigma}\left((x - \tfrac{L}{2})^2 - s^2\right) + C_1 + O(\varepsilon), \qquad x \in \omega \tag{3.14a}$$

$$h_2(x) = -\frac{1}{2}\lambda_1\left((x - \tfrac{L}{2})^2 - s^2\right) + C_1 + O(\varepsilon), \qquad x \in \omega \tag{3.14b}$$

$$h_1(x) = h_2(x) = -\frac{1}{2}\frac{\lambda_2}{\sigma + 1}\left((x - \tfrac{L}{2})^2 - s^2\right) + C_1 + O(\varepsilon), \qquad x \in (0, L)\setminus\omega, \tag{3.14c}$$

where $\omega = (L/2 - s, L/2 + s)$ and

$$s^2 = \frac{2\sigma(\sigma + 1)}{(\lambda_2 - (\sigma + 1)\lambda_1)^2}.$$

In contrast to solution of (3.14) *with Neumann conditions, solutions with Dirichlet boundary conditions are not constant but quadratic in the ultra-thin layer $(0, L) \setminus \omega$.*

4 Matched asymptotic solution and contact angles

Note first that the system of equations for h_1 and h_2 (3.4) is equivalent to following system for h_1 and h

$$0 = \partial_x\left(-\sigma\partial_{xx}h_1 - \phi'_\varepsilon(h)\right), \tag{4.1a}$$

$$0 = \partial_x\left(-\frac{\sigma}{\sigma + 1}\partial_{xx}h + \phi'_\varepsilon(h)\right). \tag{4.1b}$$

where we denote $\sigma = \sigma_1/\sigma_2$, see [21] for more details. For our asymptotic analysis, this is convenient, since now for the variable $h = h_2 - h_1$ we can distinguish the core droplet region, which we will call the “outer region” and the adjacent thin regions of thickness ε, which we call the “inner region”. We will derive a sharp-interface limit using matched asymptotic analysis in the limit as $\varepsilon \to 0$. For this we first write the equations in the form such that the intermolecular potential is small in the core region and becomes order one in the adjacent thin regions. Using $(n, \ell) = (2, 8)$ we define

$$\phi'_\varepsilon(h) = \frac{1}{\varepsilon}\Phi'\left(\frac{h}{\varepsilon}\right) \tag{4.2}$$

4.1 Stationary solution for h

As we will later show rigorously, we can assume that the droplet is (axi)symmetric and the profile a decreasing function, so that without loss of generality, the maximum of h is at the origin of our coordinate system. Now consider the problem h and $x \geq 0$

$$0 = \partial_x \left[\frac{\sigma}{\sigma+1} \partial_{xx} h - \frac{1}{\varepsilon} \Phi' \left(\frac{h}{\varepsilon} \right) \right], \tag{4.3a}$$

$$\lim_{x\to\infty} h = h_\infty, \quad \lim_{x\to\infty} \partial_x h = 0, \quad \lim_{x\to\infty} \partial_{xx} h = 0. \tag{4.3b}$$

We can integrate this twice and use the conditions as $x \to \infty$ to fix the integration constants to obtain

$$\partial_x h = \sqrt{2\frac{\sigma+1}{\sigma}} \sqrt{\Phi\left(\frac{h}{\varepsilon}\right) - \Phi\left(\frac{h_\infty}{\varepsilon}\right) - \frac{1}{\varepsilon}\Phi'\left(\frac{h_\infty}{\varepsilon}\right)(h - h_\infty)} \tag{4.4}$$

The solution to this problem shows a steep decline in height towards $O(\varepsilon)$ in an ε-strip around $x = s$, where we would like to determine the apparent contact-angle. This can be obtained by writing the problem in so-called outer and inner coordinates, valid in the core and the adjacent thin regions, and matching as $\varepsilon \to 0$. Interestingly, while it is easy to obtain the condition for the contact angle, it turns out that in order to carry out the complete matching consistently, we need to go up to second order in the matching, in order to account for the logarithmic switch-back terms, that come into play in this problem.

Note, that the coefficient $(\sigma+1)/\sigma$ can be removed by rescaling x appropriately, leading to the classical problem of a droplet of height h on a solid substrate. Interestingly, to our knowledge for this problem the above mentioned logarithmic switch-back terms have not been noticed before.

Outer problem

The symmetry of the problem leads us to the condition that at the symmetry axis $x = 0$ we have

$$\partial_x h = 0 \tag{4.5}$$

It is also convenient to normalize the height such that $h(0) = 1$. In this case we obtain from (4.4) an algebraic equation for h_∞ and ε that can be approximated as $\varepsilon \to 0$.

$$0 = \Phi\left(\frac{1}{\varepsilon}\right) - \Phi\left(\frac{h_\infty}{\varepsilon}\right) - \frac{1}{\varepsilon}\Phi'\left(\frac{h_\infty}{\varepsilon}\right)(1 - h_\infty) \tag{4.6}$$

Solving this by making the ansatz for the asymptotic expansion for h_∞

$$h_\infty = \varepsilon h_{\infty,0} + \varepsilon^2 h_{\infty,1} + \varepsilon^3 h_{\infty,2} + O(\varepsilon^4) \tag{4.7}$$

we obtain

$$h_{\infty,0} = 1, \quad h_{\infty,1} = \frac{1}{16}, \quad h_{\infty,2} = \frac{45}{512}, \tag{4.8}$$

Next, we assume that the asymptotic solution to the outer problem can be represented by the expansion

$$h(x;\varepsilon) = f_0(x) + \varepsilon f_1(x) + \varepsilon f_2(x) + O(\varepsilon^3). \tag{4.9}$$

The leading order outer problem then becomes

$$\partial_x f_0 = -\sqrt{\frac{3}{4}\frac{\sigma+1}{\sigma}}\,(1-f_0), \tag{4.10a}$$

$$f_0(0) = 1 \tag{4.10b}$$

which has the solution

$$f_0(x) = -\frac{3}{16}\frac{\sigma+1}{\sigma}x^2 + 1. \tag{4.11}$$

Hence, the leading order outer solution will vanish as x approaches the location

$$s = \frac{4}{\sqrt{3}}\sqrt{\frac{\sigma}{\sigma+1}} \tag{4.12}$$

However, the full solution does not vanish and will be obtained by matching to the solution of the "inner" problem near s. We will find that in order to complete the solution, we will need to solve the expansion up-to second order. We find for f_1 and f_2

$$\partial_x f_1 = \frac{f_1}{x} - \frac{3}{16}\frac{\sigma+1}{\sigma}\,x, \tag{4.13a}$$

$$f_1(0) = 0 \tag{4.13b}$$

and

$$\partial_x f_2 = 2\frac{f_2}{x} + \frac{1}{x}\left(\frac{8}{3}\frac{1}{f_0^2} + \frac{1}{16}(f_0-1) - \frac{2}{3}(f_0+3) + 2f_1\right) + \frac{3}{8}\frac{\sigma+1}{\sigma}\,x \tag{4.14a}$$

$$f_2(0) = 0 \tag{4.14b}$$

Inner problem

The solution of the inner problem lives in an ε neighborhood of $x = s$ and extends towards $x \to +\infty$. It will be matched to the outer problem in the other direction. Hence we introduce the inner variables $v(z)$ and independent variable z via

$$h(x) = \varepsilon\, v(z;\varepsilon) \quad \text{and} \quad x = s + \varepsilon z. \tag{4.15}$$

Rewriting the problem (4.4) in these coordinates and making the ansatz

$$v(z;\varepsilon) = v_0(z) + \varepsilon v_1(z) + O(\varepsilon^2) \tag{4.16}$$

we find to leading order the problem

$$\partial_z v_0 = -\sqrt{2\frac{\sigma+1}{\sigma}}\sqrt{-\frac{1}{2v_0^2} + \frac{1}{8v_0^8} + \frac{3}{4}} \tag{4.17}$$

and to $O(\varepsilon)$ the problem

$$\partial_z v_1 = -\sqrt{2\frac{\sigma+1}{\sigma}}\sqrt{-\frac{1}{2v_0^2}+\frac{1}{8v_0^8}+\frac{3}{4}}\;\frac{3v_0^9(v_0-1)+8v_1(1-v_0^6)}{2v_0(4v_0^6-3v_0^8-1)} \tag{4.18}$$

We solve and match in the inner coordinates and obtain by expanding v at $z=-\infty$

$$\begin{aligned} v_0+\varepsilon v_1 = &-\frac{\sqrt{3}}{2}\sqrt{\frac{\sigma+1}{\sigma}}\,z + a_1 - \frac{4}{9}\sqrt{3\frac{\sigma}{\sigma+1}}\,\frac{1}{z}+\cdots \\ &+\varepsilon\left(-\frac{3}{16}\frac{\sigma+1}{\sigma}\,z^2+\frac{\sqrt{3}}{4}\frac{\sigma+1}{\sigma}(a_1-1)\,z-\ln(-z)\right. \\ &\left.+C+\frac{1}{6}+\frac{2}{3}\sqrt{3\frac{\sigma}{\sigma+1}}(a_1+1)\frac{1}{z}+\cdots\right). \end{aligned} \tag{4.19}$$

For the corresponding outer expansion we have

$$\begin{aligned} \frac{f_0+\varepsilon f_1+\varepsilon^2 f_2}{\varepsilon} = &-\frac{\sqrt{3}}{2}\sqrt{\frac{\sigma+1}{\sigma}}\,z - 1 - \frac{4}{9}\sqrt{3\frac{\sigma}{\sigma+1}}\,\frac{1}{z}+\cdots \\ &+\varepsilon\left(-\frac{3}{16}\frac{\sigma+1}{\sigma}\,z^2-\frac{\sqrt{3}}{2}\frac{\sigma+1}{\sigma}\,z-\ln(-z)\right. \\ &\left.-\ln\left[\frac{\sqrt{3}}{8}\sqrt{\frac{\sigma+1}{\sigma}}\right]-\frac{19}{96}-\ln(\varepsilon)+\cdots\right). \end{aligned} \tag{4.20}$$

We note that all of the terms in the first row of (4.19) and (4.20) match provided $a_1=-1$. The terms in the second row match provided

$$C = -\ln(\varepsilon)-\frac{35}{96}-\ln\left(\frac{\sqrt{3}}{8}\sqrt{\frac{\sigma+1}{\sigma}}\right) \tag{4.21}$$

where we note the appearance of a so-called “logarithmic switch-back” term $\ln(\varepsilon)$.

Hence, the composite solution is

$$\begin{aligned} \bar{h} = \;&\varepsilon\left[v_0\left(\frac{x-s}{\varepsilon}\right)+\frac{\sqrt{3}}{2}\sqrt{\frac{\sigma+1}{\sigma}}\,\frac{x-s}{\varepsilon}\right] \\ &+\varepsilon^2\left[v_1\left(\frac{x-s}{\varepsilon}\right)+\frac{3}{16}\frac{\sigma}{\sigma+1}\left(\frac{x-s}{\varepsilon}\right)^2+\frac{\sqrt{3}}{2}\frac{\sigma+1}{\sigma}\,\frac{x-s}{\varepsilon}\right] \\ &+f_0(x)+\varepsilon\Big[f_1(x)+1\Big] \\ &+\varepsilon^2\left[f_2(x)+\frac{4}{9}\sqrt{\frac{3\sigma}{\sigma+1}}\,\frac{1}{x-s}+\ln(s-x)+\frac{19}{96}+\ln\left(\frac{\sqrt{3}}{8}\sqrt{\frac{\sigma+1}{\sigma}}\right)\right] \end{aligned} \tag{4.22}$$

with s given in equation (4.12) and for $x<s$. For $x\geq s$ only the inner expansion $\bar{h}=\varepsilon v_0+\varepsilon^2 v_1$ remains.

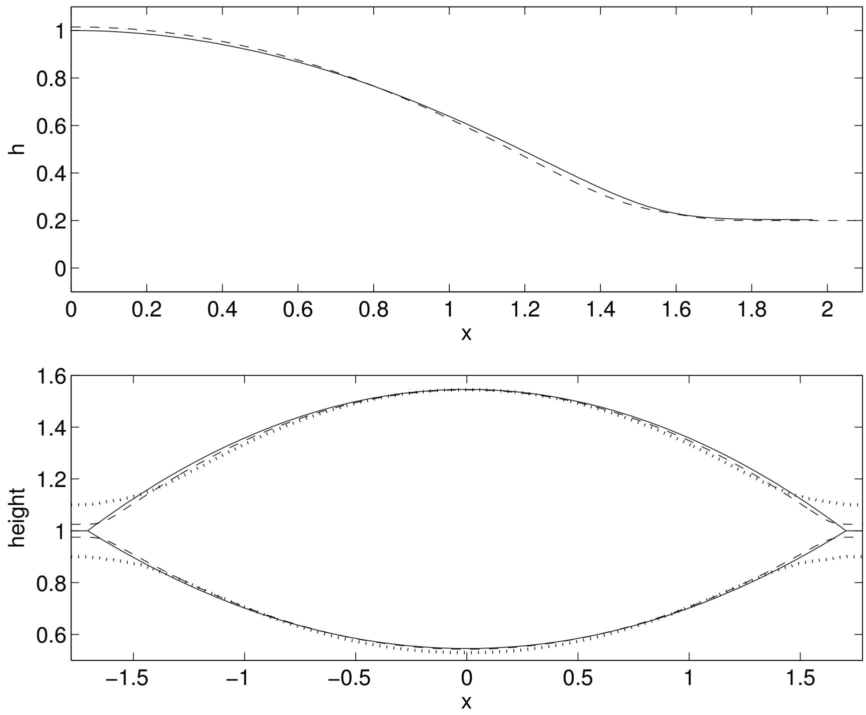

Figure 2: Top: Comparison of the composite solution h_c (dashed curve) with the numerical solution of (4.3) (solid curve), for $\varepsilon = 0.2$ and $\sigma = 1.2$. Bottom: Solutions for h_1 and h_2 reconstructed from the composite solution $\bar{h}$ for $\sigma = 1.2$ and $\varepsilon = 0.05$ (dashed curve) and $\varepsilon = 0.2$ (dotted curve) and the solution to the sharp interface model (4.24) (solid curve).

Stationary solution for h_1 and h_2

To the complete solution, we determine the solution to the liquid-liquid interface h_1 simply by adding equations (4.1a) and (4.1b). Then we integrate thrice and use the far-field conditions $\partial_{xx}h, \partial_x h, \partial_{xx}h_1, \partial_x h_1 \to 0$, $h \to h_\infty$ and $h_1 \to d$ as $x \to \pm\infty$ to fix the constants. This results in h_1 being

$$h_1 = -\frac{1}{\sigma+1}\,(h - h_\infty) + d, \tag{4.23a}$$

and equivalently h_2 is

$$h_2 = \frac{\sigma}{\sigma+1}\,(h - h_\infty) + d + h_\infty. \tag{4.23b}$$

Here d denotes the height h_1 as $x \to \pm\infty$ and we assume d to be large enough so that h_1 never becomes negative. Also note that for other boundary conditions, such as e.g. Dirichlet conditions mentioned in the previous section, further contributions will arise. Unlike the solutions for droplets on solid substrates, here new families of profiles for the ultra-thin film connecting a droplet to the boundaries or to other droplets arise.

In figure 2 we compare our asymptotic solution with the numerical solution. Observe that the $O(\varepsilon)$ solution already gives an excellent approximation of the exact solution for $\varepsilon = 0.2$, where the exact solution is approximated by the higher-order numerical solution of the boundary value problem. This suggests that a sharp-interface model should also be a good approximation of the full model. In the following section we show how the sharp-interface model is obtained via Γ-convergence and also proof existence and uniqueness of its solutions.

Sharp-interface model and contact angles

The sharp-interface model for h is simply the leading order outer problem for h, but now with boundary conditions, that result from the leading order matching. Hence, we obtain

$$0 = \partial_{xxx} f_0 \tag{4.24a}$$

with boundary conditions

$$f_0 = 0 \quad \text{and} \quad \partial_x f_0 = \partial_z v_0 = -\sqrt{-2\frac{\sigma+1}{\sigma}\Phi(1)} \quad \text{at} \quad x = \pm s \tag{4.24b}$$

where the contact angle has been determined via matching. Equivalently, for the half-droplet, by imposing symmetry we have the boundary conditions

$$f_0(s) = 0, \quad \partial_x f_0(s) = -\sqrt{-2\frac{\sigma+1}{\sigma}\Phi(1)} \quad \text{and} \quad \partial_x f_0(0) = 0 \tag{4.25}$$

5 Sharp-interface limit via Γ-convergence

In this section we investigate properties of stationary solutions of the sharp-interface two-layer model. Such model can be obtained by considering the limiting problem $\varepsilon \to 0$ in the framework of Γ-convergence. For one-layer systems corresponding minimizers are known as mesoscopic droplets [38]. In this approach equilibrium contact angles can be directly deduced from the Euler-Lagrange equation of the resulting Γ-limit energy. On the other hand showing an equi-coerciveness property we have that the sequence of minimizers of E_ε converges to a minimizer of the Γ-limit energy E_∞.

For boundary conditions $h_1 = h_2$ and certain domains we show that solutions exists and are unique up-to translations. The main technique used here is the symmetric-rearrangement, see e.g. [31, 32], which was previously applied by Otto et al. [38].

For the section to come we consider energies such as in (3.2). For later convenience we define $W_\varepsilon(h) = \big(\phi_\varepsilon(h) - \Phi(1)\big)/|\Phi(1)|$ where as before $W(h) = W_\varepsilon(h/\varepsilon)$ is independent of ε. The shift by $|\Phi(1)|$ has the advantage of working with a non-negative energy without changing the Euler-Lagrange equations.

With these definitions consider the following family of minimization problems. For $\Omega \subset \mathbb{R}^n$ bounded with Lipschitz boundary and $m_1, m_2 > 0$ given and $\varepsilon > 0$ we look for minimizers of $E_\varepsilon : X_{\mathbf{m}} \to \mathbb{R}^\infty$ defined as

$$E_\varepsilon(h_1, h_2) = \int_\Omega \frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\, W_\varepsilon(h_2 - h_1) \tag{5.1}$$

with $\sigma > 0$ and $W_\varepsilon(h) = W\big(h/\varepsilon\big)$ as $\varepsilon \to 0$. Note that nonnegativity $h_1 > 0$ is not enforced because otherwise extra terms in E_ε are required.

5.1 Γ-convergence

For a given domain $\Omega \subset \mathbb{R}^n$ bounded with Lipschitz boundary define the space of admissible interfaces as before in (3.9) by

$$X_{\mathbf{m}} := \left\{ (h_1, h_2) \in H^1(\Omega)^2 : m_1 = \int_\Omega h_1,\ m_2 = \int_\Omega (h_2 - h_1),\ h_2 \geq h_1 \right\}, \tag{5.2}$$

and in general let $W : \mathbb{R} \to \mathbb{R}$ be a function with the properties

(i) $W \geq 0$ and $W(h) = 0 \Leftrightarrow h = 1$.

(ii) $W(h) \xrightarrow{h \to +\infty} 1$ and $W(h) \leq 1$ for $h > 1$.

We want to investigate the family of minimization problems (5.1). First we note that the sequence E_ε is equi-coercive in the weak topology of $H^1(\Omega)^2$. This is a simple consequence of

$$E_\varepsilon(h_1, h_2) \geq c(\|\nabla h_1\|_{H^1}^2 + \|\nabla h_2\|_{H^1}^2),$$

which holds for all $(h_1, h_2) \in X_{\mathbf{m}}$. Together with the Γ-convergence the equi-coercivity implies the following. We know that any sequence $\{h_{1,n}, h_{2,n}\}$ of minimizers to the energies E_{ε_n} has a weakly converging subsequence $(h_{1,n}, h_{2,n}) \rightharpoonup (h_1^*, h_2^*)$. Furthermore the limit (h_1^*, h_2^*) is a minimizer of the Γ-limit energy E_∞. This relates minimizers of E_ε to minimizers of the Γ-limit E_∞.

Now we investigate the Γ-limit of (5.1) in the topology of weak convergence in the space $H^1(\Omega)^2$. We recall the definition of Γ-convergence , see also [6, 11].

Definition 5.1. *We say that a sequence $E_\varepsilon : X \to \mathbb{R}^\infty$ Γ-converges in X in the weak topology to $E_\infty : X \to \mathbb{R}^\infty$ if for all $x \in X$ we have*

(i) (lim-inf inequality) For every sequence $\{x_\varepsilon\} \subset X$ weakly converging to x there holds

$$E_\infty(x) \leq \liminf_\varepsilon E_\varepsilon(x_\varepsilon).$$

(ii) (lim-sup inequality) There exists a sequence $\{x_\varepsilon\} \subset X$ weakly converging to x such that

$$E_\infty(x) \geq \limsup_\varepsilon E_\varepsilon(x_\varepsilon).$$

The function E_∞ is called the Γ-limit of (E_ε), and we write $E_\infty = \Gamma\text{-}\lim_\varepsilon E_\varepsilon$.

The key proposition to compute the overall Γ-limit is to consider the Γ-limit of the potential separately. Here we use that weak convergence in H^1 implies strong convergence in L^2 and the right continuity of $q \mapsto \int \chi\{h > q\}$ for any given $h \in H^1$.

Proposition 5.2. *Consider the functional $F_\varepsilon : H^1(\Omega) \to \mathbb{R}^\infty$ defined as*

$$F_\varepsilon(h) = \begin{cases} \int_\Omega W_\varepsilon(h) & h \in X_m, \\ \infty & \textit{otherwise}, \end{cases}$$

where

$$X_m = \left\{ h \in H^1(\Omega) : h \geq 0, \int_\Omega h = m \right\}.$$

Then

$$\Gamma\text{-}\lim_\varepsilon F_\varepsilon(h) = F_\infty(h) = \begin{cases} \int_\Omega \chi\{h > 0\} & h \in X_m, \\ \infty & \textit{otherwise}, \end{cases}$$

with χ being the characteristic function.

Proof. Consider an arbitrary sequence $\varepsilon_n \to 0$ and $h \in X_m$.
(i) (*lim-inf condition*)
Let $\{h_n\} \subset X_m$ such that $h_n \rightharpoonup h$ weakly in $H^1(\Omega)$, then $h_n \to h$ strongly in $L^2(\Omega)$. Choose $\delta_n \to 0$ such that $\varepsilon_n/\delta_n \to 0$ as $n \to \infty$ which immediately gives

$$\liminf_n \int_\Omega W_{\varepsilon_n}(h_n) \geq \liminf_n \int_{\{h_n > \delta_n\}} W_{\varepsilon_n}(h_n) = \liminf_n \int_\Omega \chi\{h_n > \delta_n\}. \tag{5.3}$$

Next we want to use $\int \chi\{h > 0\} \leq \liminf \int_\Omega \chi\{h_n > \delta_n\}$. Conversely assume

$$\liminf_n \int_\Omega \chi\{h_n > \delta_n\} < \int \chi\{h > 0\}. \tag{5.4}$$

Then employing right-continuity of $s \mapsto \int_\Omega \chi\{h > s\}$ (see [31], Proposition 6.1) there must exist some $\delta, \bar{\delta} > 0$ such that

$$\begin{aligned} 0 < \liminf_n \int_\Omega \chi\{h > 0\} - \chi\{h_n > \delta_n\} - \delta &\leq \liminf_n \int_\Omega \chi\{h > \bar{\delta}\} - \chi\{h_n > \delta_n\}, \\ \leq \liminf_n \int_\Omega \chi\{h > \bar{\delta} \ \&\ h_n < \delta_n\} &\leq \left(\frac{2}{\bar{\delta}}\right)^2 \liminf_n \int_\Omega |h - h_n|^2 = 0, \end{aligned}$$

where we used Chebyshev's inequality. This is a contradiction and thus by the previous assertions

$$\liminf_n \int_\Omega W_{\varepsilon_n}(h_n) \geq \int_\Omega \chi\{h > 0\}.$$

(ii) (*lim-sup condition*)
Define a recovery sequence by $h_n = \alpha_n h + \varepsilon_n$ where $\alpha_n = m - \varepsilon_n |\Omega|/m$. Then $h_n \in X_m$ and $h_n \to h$ even strongly in $H^1(\Omega)$ and the following estimate holds

$$\begin{aligned} \limsup_n \int_\Omega W_{\varepsilon n}(h_n) &= \limsup_n \left(\int_{\{h>0\}} W\left(1 + \frac{\alpha_n}{\varepsilon_n} h\right) + \int_{\{h=0\}} W\left(1 + \frac{\alpha_n}{\varepsilon_n} h\right) \right), \\ &\leq \limsup_n \int_\Omega \chi\{h > 0\} + \int_{\{h=0\}} W(1) = \int_\Omega \chi\{h > 0\}, \end{aligned}$$

where we used that $W(s) \leq 1$ for $s > 1$ and $W(1) = 0$. □

To prove the Γ-convergence to the sharp-interface model we can exploit the property that the behavior of gradient terms can be easily controlled.

Theorem 5.3. *For the family of energies* (5.1) *the* Γ*-limit is*

$$E_\infty(h_1,h_2) = \int_\Omega \frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\,\chi\{h_2 > h_1\}$$

Proof. The gradient terms in E_ε are weakly lower semicontinuous with respect to weak convergence in $H^1(\Omega)^2$. Together with Proposition 5.2 this gives the *lim-inf inequality*. On the other hand the gradient terms are continuous with respect to strong convergence in $H^1(\Omega)^2$. Choosing the recovery sequence as in the proof of Proposition 5.2 one gets the desired *lim-sup inequality*. □

Now we want to deduce necessary conditions for minimizers of E_∞. We are especially interested in conditions at the points where the two phase domain meets the one-phase domain. One problem is that one cannot immediately compute the Euler-Lagrange equations (L^2-gradient) of the sharp-interface energy functional E_∞. This is due to $\int_\Omega \chi\{h_2 > h_1\}$ being only lower semicontinuous in the strong $H^1(\Omega)$ topology, but not continuous nor differentiable. In fact directional derivatives of this part of the energy will almost surely be zero or infinite. Therefore we compute the directional derivative of E_∞ in another topology as follows: For ease of notation introduce

$$\omega = \{x \in \Omega : h_2 > h_1\} \tag{5.5}$$

and restrictions of h_1 and h_2 to ω and $\Omega \setminus \omega$ are called

$$h_1 := h_1|_\omega, \quad h_2 := h_2|_\omega, \quad h := h_1|_{\Omega\setminus\omega} = h_2|_{\Omega\setminus\omega},$$

with boundary condition $h_1 = h_2 = h$ on $\partial\omega$. We will now vary h_1, h_2 and h but also ω. The formal calculation is restricted to smooth h_1, h_2, h and ω. Using these we can rewrite the energy using the restrictions as

$$E_\infty(h_1,h_2,h,\omega) = \int_\omega \left[\frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\right] + \int_{\Omega\setminus\omega} \frac{\sigma'}{2}|\nabla h|^2, \tag{5.6}$$

where we define $\sigma' := 1 + \sigma$. A perturbation of $\tau \mapsto \big(h_1(\tau), h_2(\tau), h(\tau), \omega(\tau)\big)$ can be parametrised using a deformation $\psi(\circ,\tau) : \Omega \to \Omega$ with $\psi(\omega(0),\tau) = \omega(\tau)$. In the same spirit define $\bar{h}_1(x_0,\tau) = h_1(\psi(x_0,\tau),\tau)$ as the pullback of h_1 by ψ and similarly $\bar{h}_2$ and $\bar{h}$. The boundary conditions $\partial_\tau \bar{h}_1 = \partial_\tau \bar{h}_2 = \partial_\tau \bar{h}$ on $\partial\omega(\tau)$ translate into

$$\dot{h}_1 + \dot{\psi}\cdot\nabla h_1 = \dot{h}_2 + \dot{\psi}\cdot\nabla h_2 = \dot{h} + \dot{\psi}\cdot\nabla h =: \dot{\xi}. \tag{5.7}$$

Here we use the notation $\dot{\psi} := \partial_\tau\psi$, $\dot{h}_1 := \partial_\tau h_1$, $\dot{h}_2 := \partial_\tau h_2$ and $\dot{h} := \partial_\tau h$. Then using Reynolds transport theorem we get

$$\begin{aligned}\frac{d}{d\tau}E_\infty(h_1,h_2,h,\omega) = &\int_\omega \left(\sigma\nabla h_1\nabla\dot{h}_1 + \nabla h_2\nabla\dot{h}_2\right) + \int_{\Omega\setminus\omega}\sigma'\nabla h\nabla\dot{h} \\ &+ \int_{\partial\omega}\left[\frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 - \frac{\sigma'}{2}|\nabla h|^2 + |\Phi(1)|\right](n\cdot\dot{\psi}).\end{aligned}$$

Applying integration-by-parts and boundary conditions (5.7) yields in one and two spatial dimensions, i.e. $\omega \subset \mathbb{R}$ and $\omega \subset \mathbb{R}^2$ the directional derivative

$$
\begin{aligned}
0 =& \frac{d}{d\tau}\Big(E_\infty + \lambda_2 \int h_1 + \lambda_1 \int (h_2 - h_1)\Big), \\
=& -\int_\omega \big[\sigma \Delta h_1 + \lambda_2 - \lambda_1\big]\dot{h}_1 + \big[\Delta h_2 + \lambda_1\big]\dot{h}_2 \; - \int_{\Omega\setminus\omega} \big[\sigma' \Delta h + \lambda_2\big]\dot{h} \\
&+ \int_{\partial\omega} \left[-\frac{\sigma}{2}(\nabla h_1)^2 - \frac{1}{2}(\nabla h_2)^2 + \frac{\sigma'}{2}(\nabla h)^2 + |\Phi(1)| + \eta^2\big(1+\sigma-\sigma'\big)\right](n\cdot\dot{\psi}) \\
&+ \int_{\partial\omega} \left[\sigma(n\cdot\nabla h_1) + (n\cdot\nabla h_2) - \sigma'(n\cdot\nabla h)\right]\big(\dot{\xi} + (t\cdot\dot{\psi})\big). \qquad (5.8)
\end{aligned}
$$

where $\eta^2 := (t\cdot\nabla h_1)^2 \equiv (t\cdot\nabla h_2)^2 \equiv (t\cdot\nabla h)^2$. The expressions inside square brackets have to vanish independently, since the perturbations $(\dot{h}_1, \dot{h}_2, \dot{h}, \dot{\xi}, \dot{\psi})$ are independent.

Remark 5.4. *Above we added Lagrange multipliers λ_1, λ_2 to take care of the mass conservation. In one dimension there is no tangential contribution and hence $\eta \equiv 0$, whereas in two dimensions the contribution with η^2 vanishes due to definition $\sigma' = \sigma + 1$. However, if σ' could be choosen independently of σ, there would be an extra contribution in that case.*

5.2 Existence and uniqueness of solutions

In this part we consider the sharp interface energy derived by Γ-convergence and study its minimizers. The main idea of the proof is to apply similar techniques as in Otto et al. [38] to the difference

$$h := h_2 - h_1. \qquad (5.9)$$

Minor additional assumptions are necessary for the proof, e.g. $\Omega \subset \mathbb{R}^n$ is a ball and the boundary condition $(h_2 - h_1)|_{\partial\Omega} = 0$ must be included into $X_{\mathbf{m}}$. The idea of the proof is still to show that for a minimizer the support of h is a ball, on which the solutions can be computed explicitly. The minimization itself is performed with masses $\mathbf{m} = (m_1, m_2)$ held fixed. Extensions of the proof and properties of the solutions are discussed in the end of this section.

Theorem 5.5. *(Minimizer of sharp interface energy)*

Let $\Omega = \mathcal{B}_R(0)$ and $X = \{(h_1, h_2) \in X_{\mathbf{m}}(\Omega) : (h_1 - h_2)|_{\partial\Omega} = 0\}$ and energy

$$E(h_1, h_2) := \int_\Omega \frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 \; + |\Phi(1)|\, \chi_{\{h_2 > h_1\}}\, \mathrm{d}x. \qquad (5.10)$$

Then using $\zeta(x) := \alpha(s^2 - |x|^2)^+$ minimizers of E with mass (m_1, m_2) are

$$h_2 = \frac{\sigma}{\sigma+1}\zeta(x - x_0) + h, \qquad h_1(x) = h_2 - \zeta(x - x_0), \qquad (5.11)$$

with constant $x_0 \in \Omega$ and $r, \alpha, h \in \mathbb{R}$. Prescribing the mass (m_1, m_2) fixes r and h, whereas α is fixed by the contact angle (Neumann triangle)

$$\sigma(\nabla h_1)^2 + (\nabla h_2)^2 = 2|\Phi(1)|, \qquad \text{at } |x| = r. \qquad (5.12)$$

For large masses m_2 (5.12) *is not required and we get $r = R$, $x_0 = 0$ in* (5.11).

Proof. Symmetry: For given $(h_1, h_2) \in X$ let $h = (h_2 - h_1) \in H_0^1(\Omega)$ as in (5.9), non-negative let

$$\lambda := \frac{\|\nabla h_2\|}{\|\nabla h\|} \geq 0.$$

Using Cauchy-Schwarz inequality in $L^2(\Omega)$ the following energy estimate holds

$$\begin{aligned} E(h_1, h_2) &\geq (\sigma + 1)\|\nabla h_2\|^2 + \sigma\|\nabla h\|^2 - 2\sigma\|\nabla h_2\|\,\|\nabla h\| + |\Phi(1)|\mu(\{h > 0\}), \\ &= \|\nabla h\|^2 \left(\lambda^2 + \sigma(1-\lambda)^2\right) + |\Phi(1)|\mu(\{h > 0\}). \end{aligned}$$

Minimizing with respect to λ gives the lower bound

$$E(h_1, h_2) \geq \frac{\sigma}{\sigma + 1}\|\nabla h\|^2 + |\Phi(1)|\mu(\{h > 0\})$$

which is attained only if $\lambda = \sigma/(\sigma + 1)$ and ∇h_2 is a multiple of ∇h. Now let h^* be the symmetric-decreasing rearrangement of h, then

$$\int_{\mathbb{R}^n} h^* \,\mathrm{d}x = \int_{\mathbb{R}^n} h \,\mathrm{d}x, \qquad \|\nabla h^*\|_{L^2(\mathbb{R}^n)} \leq \|\nabla h\|_{L^2(\mathbb{R}^n)},$$

and equality holds only if h is symmetric-decreasing [32]. Now assume that h is not symmetric decreasing, or $\nabla h \neq \sigma/(\sigma + 1)\nabla h_2$. Then we can reduce the energy by defining h_1^* and h_2^* by

$$h_2^*(x) := \frac{\sigma}{\sigma + 1}h^*(x - x_0) + h, \qquad h_1^*(x) := h_2^*(x) - h^*(x - x_0),$$

and have $\nabla h_2^* = \lambda\nabla h^*$ and $\mu\{h > 0\} = \mu\{h^* > 0\}$ so that

$$E(h_1, h_2) > \frac{\sigma}{\sigma + 1}\|\nabla h^*\|^2 + |\Phi(1)|\mu(\{h^* > 0\}) = E(h_1^*, h_2^*).$$

Note that $\{h^* > 0\} = \mathcal{B}_s(0) =: \omega^*$ with s such that $\mu(\omega^*) = \mu\{h > 0\}$. To check $\zeta(x) = \alpha(s^2 - |x|^2)^+$ is analogous to [38]. One has to solve the Euler-Lagrange equation for the first variation of E given in (5.8) using standard methods. □

Corollary 5.6. *Let X be as before and the sharp interface energy*

$$E_\infty(h_1, h_2, h, \omega) := \int_\omega \left(\frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\right)\mathrm{d}x + \int_{\Omega\setminus\omega} \frac{\sigma'}{2}|\nabla h|^2 \,\mathrm{d}x \quad (5.13)$$

as in (5.6) *for $\sigma' > 0$ arbitrary. Then the minimizers of* (5.10) *and* (5.13) *in X are identical.*

Proof. Since we have $h = 0$ on the complement of ω the estimates of the previous proof are valid if the domain of integration is restricted to ω. By construction we have $\|\nabla h\|^2_{L^2(\Omega\setminus\omega)} \geq \|\nabla h^*\|^2_{L^2(\Omega\setminus\omega^*)} = 0$. □

Remark 5.7. *Using the abbreviation $c = |\Phi(1)|(\sigma+1)/\sigma$ we can easily compute the parameters r and α from the previous theorem and get*

$$s_{1d} = \left(\frac{9m_2^2}{8c}\right)^{1/4}, \qquad \alpha_{1d} = \left(\frac{2c^3}{9m_2^2}\right)^{1/4},$$
$$s_{2d} = \left(\frac{8m_2^2}{\pi^2 c}\right)^{1/6}, \qquad \alpha_{2d} = \left(\frac{\pi c^2}{2m_2}\right)^{1/3}.$$

in one and two spatial dimension respectively. The contact angles are then

$$\mathbf{n}\cdot\nabla h_1\big|_{s_-} = \pm\frac{\sqrt{2c}}{1+\sigma} \qquad \mathbf{n}\cdot\nabla h_2\big|_{s_-} = \mp\sqrt{2c}\frac{\sigma}{1+\sigma}. \tag{5.14}$$

and which actually holds in any spatial dimension. We also have

$$\mathbf{n}\cdot\nabla(h_2-h_1) = \sqrt{2c} = \sqrt{-\frac{2(\sigma+1)}{\sigma}\Phi(1)} \tag{5.15}$$

which can be compared with the appropriate boundary condition in (4.24) *from the matched asymptotic expansion.*

6 Conclusions and Outlook

We considered stationary solutions of systems of coupled lubrication equations for liquid bilayers with an ultra-thin layer of thickness ε. Firstly, we studied the Euler-Langrange equations using matched asymptotics. The essential idea here is to rewrite the equations in terms of the difference $h_2 - h_1$ which then turned out to be the lubrication equation for a droplet on a solid substrate. For that equation we included a discussion of switch-back terms, which appear in the matching procedure. From the leading order solution of the matched solution we constructed a sharp-interface model.

Secondly, we derived a sharp interface model rigorously. Here we used the variational structure of the equations to formulate the problem as a minimization problem, for which we can study the limit $\varepsilon \to 0$ by Γ-convergence. In one spatial dimension on an interval both sharp-interface models are equivalent. In particular the contact angle of h from the matched asymptotics in (4.24) is the same as the one from the Γ-convergence in (5.15). Since the recovery of h_1, h_2 from h in both cases works via (4.23a,4.23b) or (5.11) the second contact angle agrees as well. In dimensions $d > 1$ one has to prove that the shape of the domain $\{h_2 - h_1 > 0\}$ is a ball of a certain radius. Using symmetric decreasing rearrangement this property, and thereby existence and uniqueness of minimizers, was proven rigorously.

We think that as for thin films on solid substrate, the technique of matched asymptotic analysis can be extended to the dynamic time-dependent problem, i.e. by computing traveling wave solutions. Furthermore we hope that the computation of the sharp-interface model also supports the understanding of the energetic structure of the system of PDEs, which should still be valid in the time-dependent problem, e.g. in the gradient flow structure of a sharp-interface model.

These insights can be applied to study dewetting regimes, dewetting rates and the stability properties of the evolving interfaces, as it was done previously for the dewetting liquid films from solid substrates, see e.g. [34, 23].

Another topic where mathematical theory could be extended concerns the early time rupture process of the film. For the single thin film on a solid substrate techniques by Bernis et al. [3] on degenerate fourth-order parabolic equations were further developed by Bertozzi et al. [5] and more recently by Chou and Kwong [8] to prove existence of weak solutions and finite time blow-up for the case of no-slip and van-der-Waals type intermolecular potentials. Experimental evidence and corresponding new lubrication models taking account of an effective interfacial slip at [15, 43, 35, 22, 33, 29, 34], led to an extension of this analysis and the one by [44] to the more complicated slip-models and can be found in Peschka et al. [40, 39]. Extensive discussions and further references on the self-similar approach to rupture can be found in Witelski and Bernoff [50] and the recent review by Eggers [14]. In a future paper we will explore also how these ideas and the approaches of this manuscript can be used to further develop the theory of rupture in liquid-liquid systems.

Finally, we found that the general structure of stationary solutions is richer compared to the situation on a solid substrate. This suggests that it is worthwhile to study the slow quasistationary coarsening dynamics of droplets on a liquid substrate. The techniques developed in the corresponding case for the single thin film can be found in [19, 18, 38, 25].

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