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STATIONARY SOLUTIONS OF LIQUID TWO-LAYER THIN-FILM MODELS*

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Abstract. We investigate stationary solutions of a thin-film model for liquid two-layer flows in an energetic formulation that is motivated by its gradient flow structure. The goal is to achieve a rigorous understanding of the contact-angle conditions for such two-layer systems. We pursue this by investigating a corresponding energy that favors the upper liquid to dewet from the lower liquid substrate, leaving behind a layer of thickness h_*. After proving existence of stationary solutions for the resulting system of thin-film equations, we focus on the limit $h_* \to 0$ via matched asymptotic analysis. This yields a corresponding sharp-interface model and a matched asymptotic solution that includes logarithmic switch-back terms. We compare this with results obtained using Γ-convergence, where we establish existence and uniqueness of energetic minimizers in that limit.

Key words. thin films, gamma-convergence, matched asymptotics, free boundaries

AMS subject classifications. 76Dxx, 76Txx, 35B40, 35C20, 49Jxx

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1. Introduction.

Understanding stability and dewetting behavior of thin liquid films coating a solid or a liquid substrate is important in many technological applications and natural phenomena on the micro- to nano scale. These range from tear films of the human eye to organic photovoltaics to numerous applications in the polymer-based semiconductor industry.

Typical film thicknesses for these applications may range from tens to hundreds of nanometers and, depending on the material composition, may be susceptible to rupture and formation of holes due to intermolecular forces. Such rupture processes typically initiate complex dewetting scenarios, where holes grow further and their trailing rims merge into polygonal networks which eventually decay into patterns of droplets that evolve on a slow time scale towards a global minimal energy state.

The present study focusses on liquid substrates, which energetically favor an interface with the underlying solid. In this case the stages of the dewetting process for the upper liquid proceed to some extent in parallel to those exhibited during dewetting of a liquid film from a solid substrate. The latter system has been investigated much more intensely in recent decades, both experimentally and theoretically. Examples of complex pattern formation can be found in Sharma and Reiter [29] or Seemann, Herminghaus, and Jacobs [27], and further experimental and theoretical investigations in

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numerous references in the recent reviews by Craster and Matar [6] and Herminghaus, Brinkmann, and Seemann [10].

For liquid-liquid dewetting, experimental studies depicting some of these dewetting stages have been conducted by various groups such as by Segalman and Green [28], Lambooy et al. [17], Slep et al. [30], or Wang, Krausch, and Geoghegan [31] for the standard system of liquid polystyrene (PS) on a liquid polymethylmethacrylate (PMMA) substrate. They include investigations of rupture and hole growth, dewetting dynamics, and equilibrium contact angles that the liquid droplets make with the underlying liquid layer, where now the contact line is fixed by two angle conditions instead of one; i.e., Young's law is replaced by the Neumann triangle construction [22]. Following the pioneering study by Brochard-Wyart, Martin, and Redon [4], where various dewetting regimes were derived and analyzed, stability of liquid-liquid systems was investigated by Danov et al. [8], Pototsky et al. [25], and Fisher and Golovin [9]. Stationary states and the dynamics towards stationary states were studied by Pototsky et al. [26], by Craster and Matar [5], and by Bandyopadhyay and Sharma [1], and for the case of gravity-driven liquid droplets on an inclined liquid substrate by Kriegsmann and Miksis [15].

Interestingly, direct quantitative comparisons of theoretical with experimental results, in particular on the micro- and nanoscales, regarding, for example, the morphology of the interfaces, as performed in Kostourou et al. [14], or equilibrium values of the Neumann triangle, as discussed in [30], still leave many issues needing to be explained, such as the dependency of the morphology of the interfaces on the rheology of the liquids, their layer thicknesses, or material parameters. On the other hand, even for the simplest mathematical models of Newtonian two-layer liquid systems, mathematical theory is still largely open, and this is the main focus of the present study.

Here, we are guided by the many similarities to dewetting from a solid substrate, and we expect that some of the mathematical analysis developed for liquid film dewetting from a solid substrate can be carried over to liquid substrates. In particular, we aim to extend the existing theory for liquid droplets on a solid substrate to the situation on a liquid substrate.

As a starting point we recall the work by Bertozzi, Grün, and Witelski [2] regarding stationary states and coarsening of droplets on solid substrates, where they showed existence of smooth global solutions for positive data with bounded energy for the no-slip lubrication equation. In addition, they proved existence of global minimizers and determined a family of positive periodic solutions for admissible intermolecular potentials consisting of long-range attractive and short-range repulsive contributions and investigated their linear stability. Further extensions were given in Laugesen and Pugh [18], where linear stability of stationary solutions for the thin-film equation with Neumann boundary conditions or periodic boundary conditions was investigated. Extensions of the existence theory to thin-film equations accounting for slip at the liquid-solid interface were given in Otto, Rump, and Slepčev [24] and Kitavtsev, Recke, and Wagner [13]. Convergence to stationary solutions of the one-dimensional thin-film equation and the number of stationary states was recently investigated by Zhang [32].

The extension of the existence theory to two-layer liquid systems is given in section 3, after the formulation of the problem. With the appropriate energy functional for the two-layer system of coupled thin-film equations for the interfacial heights h_1 and h_2 (see sketch in Figure 1.1), together with Neumann boundary conditions, we show existence of smooth stationary solutions as well as existence of a global minimizer

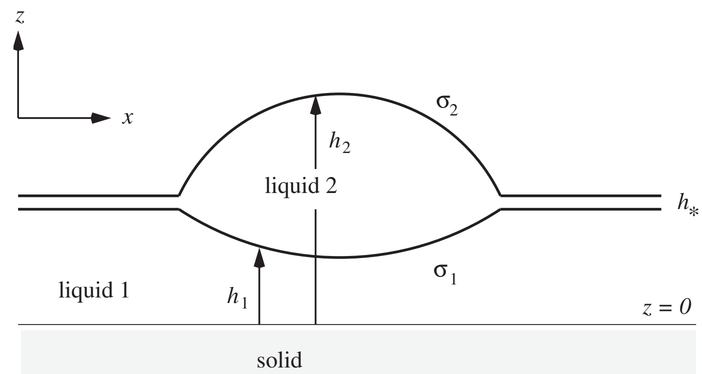

FIG. 1.1. *Sketch of liquid droplet with surface tension σ_2 between air and liquid$_2$, on top of a liquid layer with interfacial tension σ_1 between liquid$_2$ and liquid$_1$.*

for the steady-state problem.

Admissible intermolecular potentials for the liquid-liquid system are 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_*. We note that such potentials are widely used in the literature; see, e.g., the review [23]. A choice that is related to the standard Lennard–Jones potential and typically used in experiments is $(n, \ell) = (2, 8)$; see, e.g., [27]. This particular choice will be used in section 3 for the existence, and in section 4 for the asymptotics.

The Neumann triangle construction for contact angles at a triple junction is thereby replaced by properties of approximate contact angles resulting from the particular structure of the surface free energy ϕ. Starting from this energetic formulation, we seek to understand in sections 4 and 5 how the Neumann triangle construction is attained as a limit $h^* \to 0$. With this in mind, we first derive in section 4 a sharp-interface model in the limit of $h^* \to 0$ using matched asymptotic analysis. This yields the appropriate expression for the equilibrium contact angles of these droplet solutions and, as a result, the corresponding Neumann triangle. Note that we use the term *droplet solution* or *liquid lens* for solutions which are localized in the sense that $h_2 - h_1$ has compact support in the limit $h_* \to 0$.

We find that, while the equilibrium contact angle is easy to obtain, as expected, the complete asymptotic argument needs to include logarithmic switch-back terms. We note, in retrospect, that since equilibrium droplet solutions for solid substrates can be considered as limiting cases of liquid lenses, similar terms should also appear in the matched asymptotic derivation for that problem.

Finally, in section 5 we rigorously show existence and uniqueness of the limit $h_* \to 0$ within the framework of Γ-convergence. We obtain a sharp-interface problem for which we compute the Euler–Lagrange equations, from which one can immediately read off the contact angles. Existence and uniqueness of minimizers is shown here using a rearrangement inequality.

2. Formulation. We consider a layered system of two immiscible Newtonian liquids with negative spreading coefficient ϕ_*. We assume that the layered system lives in the two phases Ω_1 and Ω_2 defined by

$$\Omega_1(t) = \{(\mathbf{x}, z) \in \mathbb{R}^{d+1} : 0 \le z < h_1(t, \mathbf{x}), \mathbf{x} \in \mathbb{R}^d\}, \tag{2.1a}$$

$$\Omega_2(t) = \{(\mathbf{x}, z) \in \mathbb{R}^{d+1} : h_1(t, \mathbf{x}) \le z < h_2(t, \mathbf{x}), \mathbf{x} \in \mathbb{R}^d\} \tag{2.1b}$$

for all $t > 0$, as sketched (in two dimensions) in Figure 1.1. The dimensions which the functions h_i live on are $d = 1, 2$. Typical applications use liquids such as PMMA for the liquid substrate Ω_1, and PS for the upper liquid Ω_2, both on a scale where gravity can be neglected and, for this study, unentangled and density matched. These simplifying assumptions allow us to describe the flow of the viscous and incompressible liquids in each phase Ω_i $(i = 1, 2)$ by the Stokes equation and the continuity equation,

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

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

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

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

Here, $\mathbf{n}_i$ denotes the outer normal.

At the solid substrate a no-slip and an impermeability condition are imposed. At the liquid-liquid interface the velocity is continuous, i.e., $\mathbf{u}_2 = \mathbf{u}_1$; the tangential stress is continuous; and the normal stress makes a jump proportional to the mean curvature $[[-p_i\mathbb{I} + \mu_i(\nabla \mathbf{u}_i + \nabla \mathbf{u}_i^\top)]]_{1,2}\mathbf{n}_1 = 2\sigma_{1,2}\kappa\mathbf{n}_1$. 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 that other contributions to the intermolecular potential are negligible; i.e., $\mathbf{f}_1 = 0$.

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

Detailed derivations of thin-film models for liquid-liquid systems are given, for example, in [15, 25] or recently, by accounting for interfacial slip, in [11]. For the convenience of the reader we note here the choice of nondimensional variable and parameters, where the nondimensional horizontal and vertical coordinates are given by $\tilde{x} = x/L$, $\tilde{y} = y/L$, and $\tilde{z} = z/h_{\max}$, and $\tilde{h} = h/h_{\max}$. The nondimensional pressures $\tilde{p}_i = p_i/P$ and the derivative of the nondimensional intermolecular potential $\phi'_\varepsilon = \phi'/P$ are scaled such that $P = \phi_* n\ell/((\ell - n)h_{\max})$, 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 nondimensional thickness of the ultrathin film. For the dynamic problem, the velocities are scaled with the characteristic horizontal velocity of the dewetting upper layer such that for the nondimensional horizontal and vertical velocities we have $\tilde{u} = u/U$, $\tilde{v} = v/U$, and $\tilde{w} = w/\varepsilon_\ell U$, with $U = \varepsilon_\ell^3\sigma_2/\mu_2$, and the nondimensional 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 $\sim$'s. 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_2 and p_1,

$$p_1 = -\sigma\Delta h_1 - \phi_\varepsilon'(h_2 - h_1), \qquad p_2 = -\Delta h_2 + \phi_\varepsilon'(h_2 - h_1), \tag{2.5}$$

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 the 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

$$Q = \frac{1}{\mu}\begin{bmatrix} \dfrac{h_1^3}{3} & \dfrac{h_1^3}{3} + \dfrac{h_1^2(h_2-h_1)}{2} \\ \dfrac{h_1^3}{3} + \dfrac{h_1^2(h_2-h_1)}{2} & \dfrac{\mu}{3}(h_2-h_1)^3 + h_1h_2(h_2-h_1) + \dfrac{h_1^3}{3} \end{bmatrix}. \tag{2.7}$$

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

$$E_\varepsilon(h_1, h_2) = \int_\Omega \left[\frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + \phi_\varepsilon(h_2 - h_1)\right] dx, \tag{2.8}$$

where the potential function ϕ_ε is given as in (2.4).

We proceed to first show existence of a 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

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

The relation to the thin-film equations is $p_i = \delta E_\varepsilon / \delta h_i$. From (2.5)–(2.7) and (3.1) conservation of mass follows:

$$\int_\Omega h_1(t,x)\, dx = m_1, \tag{3.2a}$$

$$\int_\Omega (h_2(t,x) - h_1(t,x))\, dx = m_2 \quad \text{for all } t > 0, \tag{3.2b}$$

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

$$0 = Q \cdot \partial_x \mathbf{p} \tag{3.3}$$

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

$$\sigma \partial_{xx} h_1 = -\phi'_\epsilon(h_2 - h_1) - \lambda_2 + \lambda_1, \tag{3.4a}$$

$$\partial_{xx} h_2 = \phi'_\epsilon(h_2 - h_1) - \lambda_1, \tag{3.4b}$$

where constants λ_2 and λ_1 are Lagrange multipliers associated with conservation of mass (3.2a) and (3.2b), respectively. To solve (3.4) let us consider the equation for the difference

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

which reads as follows:

$$\partial_{xx} h = \frac{\sigma + 1}{\sigma} \phi'_\epsilon(h) + \frac{1}{\sigma}\lambda_2 - \frac{\sigma + 1}{\sigma}\lambda_1. \tag{3.5}$$

For brevity we set

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

According to [2], for positive P there exists a so-called *droplet* solution $\bar{h}$ to (3.5) 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 (3.4) two times with respect to x. We then obtain a solution to (3.4) with (3.1) in the form

$$\begin{aligned} h_1 &= -\frac{1}{\sigma + 1}\bar{h} - \frac{1}{2}\frac{\lambda_2}{\sigma + 1}x^2 + Cx + C_1, \\ h_2 &= \frac{\sigma}{\sigma + 1}\bar{h} - \frac{1}{2}\frac{\lambda_2}{\sigma + 1}x^2 + Cx + C_1. \end{aligned} \tag{3.6}$$

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

$$\begin{aligned} h_1 &= -\frac{1}{\sigma + 1}\bar{h} + C_1, \\ h_2 &= \frac{\sigma}{\sigma + 1}\bar{h} + C_1. \end{aligned} \tag{3.7}$$

The additive constant C_1 and the remaining Lagrange multiplier are determined from the conservation of mass (3.2a) and (3.2b), respectively. We conclude that the solution (3.7) is given by the combination of two symmetric droplets with constant outer layer. The next theorem establishes existence of a global minimizer to the energy functional (2.8) and shows that it satisfies (3.4) 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* (2.8) *exists in the class*

$$X_{\mathbf{m}} := \left\{ (h_1, h_2) \in H^1(\Omega)^2 : m_1 = \int_\Omega h_1,\ m_2 = \int_\Omega (h_2 - h_1),\ h_2 \geq h_1 \right\}. \tag{3.8}$$

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 (3.4) *with* (3.1) *and*

$$\lambda_1 = \frac{1}{L}\int_\Omega \phi'_\epsilon(h_2 - h_1)\, dx, \quad \lambda_2 = 0. \tag{3.9}$$

Proof. Even though the proof proceeds very analogously to the one of Theorem 2-3 in [2], using direct methods of the calculus of variations, for the convenience of the reader we provide the details for our system.

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

$$\int_\Omega |\nabla h_1^k|^2 + |\nabla h_2^k|^2 \, dx \le C_2 \quad \text{for all } k \in \mathrm{N}. \tag{3.10}$$

Rellich's compactness theorem implies that a subsequence (again denoted by $(h_1^k, h_2^k)_{k\in\mathrm{N}}$) exists 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

$$E_\epsilon(h_1, h_2) \le \liminf_{k\to\infty} E_\epsilon(h_1^k, h_2^k) = \inf_{(\bar h_1, \bar h_2)\in X_{\mathbf{m}}} E_\epsilon(\bar h_1, \bar h_2).$$

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.

Now let $d = 1$ and $\Omega = (0, L)$. The estimate (3.10) implies

$$\int_\Omega |\partial_x(h_2^k - h_1^k)|^2 \, dx \le C_3.$$

Next, the boundedness of $\int \phi_\epsilon(h_2^k - h_1^k)\, dx$ 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

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

where C_4 is constant. Therefore, the function $(h_2^k - h_1^k)^{-3}$ is uniform in k, bounded in $W^{1,1}(0, L)$. Owing to the continuous embedding of $W^{1,1}(0, L)$ into $L^\infty(0, L)$, 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

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

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

$$\int_0^L \varphi_1 \, dx = \int_0^L (\varphi_2 - \varphi_1)\, dx = 0.$$

Without this constraint, using Lagrangian multipliers and testing with general $(\psi_1, \psi_2) \in H^1(0, L)^2$, this yields

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

Standard elliptic regularity theory then implies that (h_1, h_2) are smooth solutions to (3.4) together with (3.1) and (3.9). ∎

Remark 3.2. Note that for Dirichlet boundary conditions we can proceed as follows: Let us impose on system (3.4) the Dirichlet boundary conditions

$$h_1(0) = h_1(L) = A, \qquad h_2(0) = h_2(L) = B \tag{3.11}$$

such that

$$B - A = \min_{x\in(0,L)} \bar{h}(x), \tag{3.12}$$

where $\bar{h}$ is the Neumann solution to (3.5) defined above. In this case it follows again that $h_2 - h_1 = \bar{h}$. Consequently, h_1 and h_2 are given by (3.6) with constants $\lambda_1, \lambda_2, C, C_1$ determined uniquely by (3.11) and conservation of mass (3.2a)–(3.2b). Using (3.12) and the asymptotics for $\bar{h}$ (see the next section for details), one obtains that the leading order of the solution (3.6) as $\varepsilon \to 0$ now has the form

$$\begin{aligned} h_1(x) &= \frac{1}{2}\frac{\lambda_1 - \lambda_2}{\sigma}\left(\left(x - \frac{L}{2}\right)^2 - s^2\right) + C_1, && x \in \omega,\\ h_2(x) &= -\frac{1}{2}\lambda_1\left(\left(x - \frac{L}{2}\right)^2 - s^2\right) + C_1, && x \in \omega,\\ h_1(x) = h_2(x) &= -\frac{1}{2}\frac{\lambda_2}{\sigma+1}\left(\left(x - \frac{L}{2}\right)^2 - s^2\right) + C_1, && x \in (0,L)\setminus\omega, \end{aligned} \tag{3.13}$$

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 the solution of (3.13) with Neumann conditions, solutions with Dirichlet boundary conditions are not constant but quadratic in the ultrathin 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, (3.3), is equivalent to the 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 [11] for a more detailed derivation. 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. We define

$$\phi_\varepsilon(h) = \Phi\left(\frac{h}{\varepsilon}\right) \quad \Longrightarrow \quad \phi_\varepsilon'(h) = \frac{1}{\varepsilon}\Phi'\left(\frac{h}{\varepsilon}\right). \tag{4.2}$$

Notice that in this section we will use $(n, \ell) = (2, 8)$ as exponents in ϕ.

4.1. Stationary solution for *h*. As we will later show rigorously, we can assume that the droplet is (axi)symmetric and that 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 to account for the logarithmic switch-back terms that come into play in this problem; see [16] for a discussion of these terms.

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

$$h(x;\varepsilon) = f_0(x) + \varepsilon f_1(x) + \varepsilon^2 f_2(x) + O(\varepsilon^3). \tag{4.9}$$

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

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

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

Stationary solution for h_1 and h_2. To complete the solution, we determine the solution to the liquid-liquid interface h_1 simply by adding (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 $h_1\to b$ as $x\to\pm\infty$ to fix the constants. This results in h_1 being

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

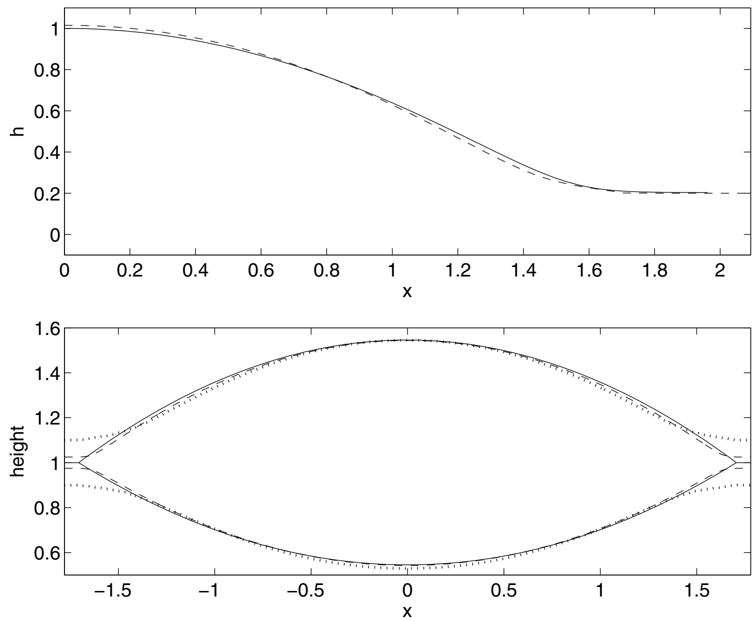

FIG. 4.1. *Top: Comparison of the composite solution h_c (dashed curve) with the numerical solution of* (4.3) *(solid curve) for $\varepsilon = 0.2$ and $\sigma = 1.2$. Bottom: Solutions for h_1 and h_2 reconstructed from the composite solution $\bar{h}$ for $\sigma = 1.2$ and $\varepsilon = 0.05$ (dashed curve) and $\varepsilon = 0.2$ (dotted curve) and the solution to the sharp interface model* (4.25) *(solid curve).*

and equivalently h_2 is

$$h_2 = \frac{\sigma}{\sigma+1}(h - h_\infty) + b + h_\infty. \tag{4.24}$$

Here b denotes the height h_1 as $x \to \pm\infty$, and we assume b to be large enough so that h_1 never becomes negative. Also note that for other boundary conditions, such as 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 ultrathin film connecting a droplet to the boundaries or to other droplets arise.

In Figure 4.1 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. 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

$$0 = \partial_{xxx} f_0, \tag{4.25a}$$

with boundary conditions

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

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

$$f_0(s) = 0, \quad \partial_x f_0(s) = -\sqrt{-2\frac{\sigma+1}{\sigma}\Phi(1)}, \quad \text{and} \quad \partial_x f_0(0) = 0. \tag{4.26}$$

In the following section we show how the sharp-interface model is obtained via Γ-convergence and also prove existence and uniqueness of its solutions.

5. Sharp-interface limit via Γ-convergence. In this section we investigate properties of stationary solutions of the sharp-interface two-layer model. Such a model can be obtained by considering the limiting problem $\varepsilon \to 0$ in the framework of Γ-convergence. For one-layer systems the corresponding minimizers are known as mesoscopic droplets [24]. 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 of the minimization problem $\min_{(h_1,h_2)} E_\infty$ exist and are unique up to translations. The main technique used here is the symmetric rearrangement; see, e.g., [19, 20].

For the section to come we consider energies such as in (2.8). For later convenience we define $W(h) = (\Phi(h) - \Phi(1))/|\Phi(1)|$ and $W_\varepsilon(h) = W(h/\varepsilon)$. Notice that $W(h)$ is independent of ε. The shift by $|\Phi(1)|$ has the advantage of working with a nonnegative energy without changing the Euler–Lagrange equations.

With these definitions consider the following family of minimization problems. For $\Omega \subset \mathbb{R}^d$ bounded with Lipschitz boundary, $m_1, m_2 > 0$ 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 $\Omega \subset \mathbb{R}^d$ bounded with Lipschitz boundary define the space of admissible interfaces as before in (3.8) 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 abstract convergence result. 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_∞, which is

$$E_\infty(h_1,h_2) = \int_\Omega \frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\,\chi\{h_2 > h_1\}. \tag{5.3}$$

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 [3, 7].

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 the following:*

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

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

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

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

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 for computing 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

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

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 $\delta_n/\varepsilon_n \to \infty$ as $n \to \infty$, which immediately gives

$$\liminf_n \int_\Omega W_{\varepsilon_n}(h_n) \ge \liminf_n \int_{\{h_n > \delta_n\}} W_{\varepsilon_n}(h_n) = \liminf_n \int_\Omega \chi\{h_n > \delta_n\}. \tag{5.4}$$

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

$$\liminf_n \int_\Omega \chi\{h_n > \delta_n\} < \int \chi\{h > 0\}. \tag{5.5}$$

Then employing the right-continuity of $s \mapsto \int_\Omega \chi\{h > s\}$ (see [19, Proposition 6.1]), there must exist some $\delta, \bar{\delta} > 0$ such that

$$0 < \limsup_n \int_\Omega \chi\{h > 0\} - \chi\{h_n > \delta_n\} - \delta \le \limsup_n \int_\Omega \chi\{h > \bar{\delta}\} - \chi\{h_n > \delta_n\}$$
$$\le \limsup_n \int_\Omega \chi\{h > \bar{\delta} \;\&\; h_n < \delta_n\} \le \left(2/\bar{\delta}\right)^2 \limsup_n \int_\Omega |h - h_n|^2 = 0,$$

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

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

(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:

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

where we used that $W(s) \le 1$ 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 in* $X_{\mathbf{m}}$ *is given by* (5.3),

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

Proof. The gradient terms in E_ε are weakly lower semicontinuous with respect to weak convergence in $H^1(\Omega)^2$. Taken 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 neither 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

$$\omega = \{x \in \Omega : h_2 > h_1\}, \tag{5.6}$$

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

$$E_\infty(h_1, h_2, h, \omega) = \int_\omega \left[\frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\right] + \int_{\Omega\setminus\omega} \frac{\sigma'}{2}|\nabla h|^2, \tag{5.7}$$

where we define $\sigma' := 1 + \sigma$. A perturbation of $\tau \mapsto (h_1(\tau), h_2(\tau), h(\tau), \omega(\tau))$ can be parametrized using a diffeomorphism $\psi(\circ, \tau) : \Omega \to \Omega$ with the property that $\omega(\tau) = \{\psi(x, \tau) : x \in \omega(0)\}$. 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 for $\bar{h}_2$ 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

$$\dot{h}_1 + \dot{\psi} \cdot \nabla h_1 = \dot{h}_2 + \dot{\psi} \cdot \nabla h_2 = \dot{h} + \dot{\psi} \cdot \nabla h =: \dot{\xi}. \tag{5.8}$$

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 the Reynolds transport theorem, we get

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

Applying integration by parts and boundary conditions (5.8) yields in one and two spatial dimensions, i.e., $\omega \subset \mathbb{R}$ and $\omega \subset \mathbb{R}^2$, the directional derivative

$$\begin{aligned}0 &= \frac{\mathrm{d}}{\mathrm{d}\tau}\left(E_\infty + \lambda_2 \int h_1 + \lambda_1 \int (h_2 - h_1)\right) \\ &= -\int_\omega \big[\sigma \Delta h_1 + \lambda_2 - \lambda_1\big] \dot{h}_1 + \big[\Delta h_2 + \lambda_1\big] \dot{h}_2 \ - \int_{\Omega\setminus\omega} \big[\sigma' \Delta h + \lambda_2\big] \dot{h} \\ &\quad + \int_{\partial\omega} \left[-\frac{\sigma}{2}(\nabla h_1)^2 - \frac{1}{2}(\nabla h_2)^2 + \frac{\sigma'}{2}(\nabla h)^2 + |\Phi(1)| + \eta^2\big(1 + \sigma - \sigma'\big)\right] (n \cdot \dot{\psi}) \\ &\quad + \int_{\partial\omega} \left[\sigma(n \cdot \nabla h_1) + (n \cdot \nabla h_2) - \sigma'(n \cdot \nabla h)\right] \big(\dot{\xi} + (t \cdot \dot{\psi})\big),\end{aligned} \tag{5.9}$$

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 chosen 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 idea of the proof is to show that for a minimizer the support of

$$h := h_2 - h_1 \tag{5.10}$$

is a ball contained in Ω, on which the solutions can be computed explicitly. The minimization itself is performed with masses $\mathbf{m} = (m_1, m_2)$ held fixed. Further extensions of our proof and properties of the solutions are discussed at the end of this section.

DEFINITION 5.5. *Let $A \subset \mathbb{R}^d$ be a Borel set of finite Lebesgue measure; then the symmetric rearrangement of the set A is defined by $A^* = \mathcal{B}_s(0)$ with s such that $\mu(A) = \mu(A^*)$. The symmetric decreasing rearrangement of the characteristic function is $(\chi(A))^* = \chi(A^*)$. Now let $f : \mathbb{R}^d \to \mathbb{R}$ be a Borel measureable function vanishing at infinity, then define the symmetric decreasing rearrangement of f by*

$$f^*(x) = \int_0^\infty \chi^*\{f > s\}(x)\ \mathrm{d}s.$$

THEOREM 5.6 (minimizer of sharp interface energy). *Let $\Omega = \mathcal{B}_R(0)$, $X = \{(h_1, h_2) \in X_{\mathbf{m}}(\Omega) : (h_1 - h_2)|_{\partial\Omega} = 0\}$, and energy*

$$E(h_1, h_2) := \int_\Omega \frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\,\chi\{h_2 > h_1\}\ \mathrm{d}x. \tag{5.11}$$

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

$$h_2 = \frac{\sigma}{\sigma+1}\zeta(x - x_0) + h, \qquad h_1(x) = h_2 - \zeta(x - x_0), \tag{5.12}$$

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):

$$\sigma(\nabla h_1)^2 + (\nabla h_2)^2 = 2|\Phi(1)| \qquad \text{at } |x| = r. \tag{5.13}$$

For large masses m_2, (5.13) is not required, and we get $r = R$, $x_0 = 0$ in (5.12).

Proof. Using ideas similar to those in [24], we proceed as follows. Symmetry: For given $(h_1, h_2) \in X$ let $h = (h_2 - h_1) \in H_0^1(\Omega)$ as in (5.10), nonnegative let

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

Using the 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, by virtue of the Pólya–Szegő inequality,

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

We have the freedom to translate h^* as long its support is contained in Ω. Equality holds only if h is already symmetric decreasing [20]. 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, by definition, $\{h^* > 0\} = \mathcal{B}_s(0) =: \omega^*$ with s such that $\mu(\omega^*) = \mu(h > 0)$. To check that $\zeta(x) = \alpha(s^2 - |x|^2)^+$ is now analogous to [24], one has to solve the Euler–Lagrange equation for the first variation of E given in (5.9) using standard methods. ∎

COROLLARY 5.7. *Let X be as before and the sharp interface energy be*

$$E_\infty(h_1, h_2, h, \omega) := \int_\omega \left(\frac{\sigma}{2}|\nabla h_1|^2 + \frac{1}{2}|\nabla h_2|^2 + |\Phi(1)|\right) \mathrm{d}x + \int_{\Omega\setminus\omega} \frac{\sigma'}{2}|\nabla h|^2 \, \mathrm{d}x \tag{5.14}$$

as in (5.7) *for $\sigma' > 0$ arbitrary. Then the minimizers of* (5.11) *and* (5.14) *in X are identical.*

Proof. Since we have $h = 0$ on $\partial\omega$, 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 5.8. Using the abbreviation $c = |\Phi(1)|(\sigma+1)/\sigma$, we can easily compute the parameters s 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 dimensions, respectively. The contact angles are then

$$\mathbf{n}\cdot\nabla h_1|_{s_-} = \pm\frac{\sqrt{2c}}{1+\sigma}, \qquad \mathbf{n}\cdot\nabla h_2|_{s_-} = \mp\sqrt{2c}\frac{\sigma}{1+\sigma}, \tag{5.15}$$

which actually hold in any spatial dimension. We also have

$$\mathbf{n}\cdot\nabla(h_2 - h_1) = \sqrt{2c} = \sqrt{-\frac{2(\sigma+1)}{\sigma}\Phi(1)}, \tag{5.16}$$

which can be compared with the appropriate boundary condition in (4.25) from the matched asymptotic expansion.

6. Discussion and outlook.

We have considered stationary solutions of systems of coupled thin-film equations for two-layer liquid films. After proving existence of stationary droplet solutions, we used matched asymptotic analysis to derive a corresponding sharp-interface model in the limit when the thickness of the ultrathin film in the dewetted region $\varepsilon \to 0$, which then yields the equilibrium Neumann angles. We point out that our asymptotic analysis requires the inclusion of logarithmic switch-back terms for the asymptotic droplet solution, which should in principle also be needed for the limiting case of droplet solutions on solid substrates.

We then proved the existence and uniqueness of the sharp-interface model using the variational structure of the equations, allowing us to formulate the problem as a minimization problem, for which we can study the limit $\varepsilon \to 0$ via 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 asymptotic analysis in (4.25) is the same as the one from the Γ-convergence in (5.16). Since the recovery of h_1, h_2 from h in both cases works via (4.23), (4.24), or (5.12), the second contact angle agrees as well. We note that for 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, could be proved.

We expect that, as for thin films on a solid substrate, the techniques of matched asymptotic analysis can be extended to the dynamic time-dependent problem. In particular, the derivation and study of the time-dependent sharp-interface model will also support the understanding of the energetic structure of the system of the coupled thin-film model and should still be valid in the time-dependent problem, e.g., in the gradient flow structure of a sharp-interface model. This will be important in the study of dewetting regimes, dewetting rates, and the stability properties of the evolving interfaces, as was seen previously for the dewetting liquid films from solid substrates; see, e.g., [21, 12].

As pointed out in the beginning of our study, mathematical theory for two-layer liquid flows still leaves many open questions and problems to be addressed. The present work can only be considered as a first step. Moreover, even considering only stationary solutions, we note that the general picture is much richer compared to the situation on a solid substrate, with energy structures leading to phase-inverted or more complicated patterns, and is the subject of our ongoing research.

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