

Note on thin film equations for solutions and suspensions

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Abstract. This note discusses how one may further develop thin film evolution equations for solutions and suspensions. First, we review the time evolution equation of a film or drop of simple liquid under the sole influence of wettability and capillarity and its formulation as a gradient dynamics. Second, we introduce such a gradient dynamics for a film of suspension or solution and show that it is equivalent to thin film equations in the literature. Finally, the new formulation is used to discuss extensions towards solute molecules/particles with net attractive interactions, decomposing solute-solvent systems and a solute-dependent wettability.

1 Introduction

The effects of capillarity and wettability are important interfacial phenomena whose detailed understanding is relevant to many small scale fluidic systems. Paradigmatic physical processes where both effects are involved are, for instance, the dewetting of very thin films on solid substrates [1, 2], the spreading of droplets on solid or liquid substrates [3–6] and the meniscus dynamics at a plate that moves slowly in or out of a bath [7, 8]. For related reviews see [4, 9–11].

For simple liquids most of the observed effects may in the limit of small contact angles be described employing a thin film evolution equation in long-wave approximation (see reviews in [12, 13]). For dewetting or spreading on a two-dimensional horizontal smooth substrate under the sole influence of capillarity and wettability the equation that governs the time evolution of the film thickness profile $h(x, y, t)$ is

$$\partial_t h = -\nabla \cdot \left[\frac{h^3}{3\eta} \nabla (\gamma \Delta h + \Pi(h)) \right], \quad (1)$$

where γ is the liquid-gas surface tension and η is the dynamic viscosity of the liquid. Here, $\nabla = (\partial_x, \partial_y)$ and ∂_i denotes partial derivatives w.r.t. to the variable i . The term $\gamma \Delta h$ stands for the curvature pressure whereas the disjoining pressure $\Pi(h)$ models wettability. It is related to a local free energy $f(h)$ by $\Pi = -\partial_h f(h)$. For details

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and specific choices for the disjoining pressure see, e.g., Refs. [4, 12, 14–16]. Thin film equations like (1) are widely used to describe dewetting films [17–20] or spreading drops [11, 21, 22] of simple liquids.

It is important to observe that Eq. (1) can be brought into the form of a gradient dynamics for a single conserved field (the film thickness) [23]

$$\partial_t h = \nabla \cdot \left[Q(h) \nabla \frac{\delta F}{\delta h} \right] \quad (2)$$

where $\delta/\delta h$ denotes functional variation with respect to h . The resulting relaxation dynamics is governed by the energy functional

$$F[h] = \int \left[\frac{\gamma}{2} (\nabla h)^2 + f(h) \right] dA \quad (3)$$

and $Q(h) = h^3/3\eta$ is the mobility for the present case of Poiseuille flow in the film and no-slip boundary conditions at the substrate. Other typical examples of equations of the form (2) are the Cahn-Hilliard equation describing the demixing of a binary mixture [24–26], evolution equations for the density field in the framework of dynamical density functional theory [27, 28], and kinetic equations for phase field crystals [29–31]. More details for the case of the thin film equation and a discussion of the incorporation of slow evaporation as a non-conserved Allen-Cahn dynamics can be found in [32]. Multiplying Eq. (2) by $\delta F/\delta h$ and integrating one may show that $dF[h]/dt \leq 0$ confirming that $F[h]$ is a Lyapunov functional.

The thin film evolution equation (2) for simple liquids is quite versatile. Additional effects can be incorporated into $F[h]$ as, e.g., the influence of an electrical field for dielectric liquids in a capacitor [33, 34], hydrostatic effects [12], the influence of a temperature difference between the substrate and the ambient air [35–37], and substrate heterogeneities [38–40]. The mobility Q can be adapted to account for slip at the substrate [41] or a porous fluid-saturated substrate [42]. With incorporated lateral forces, Eq. (1) is also widely used in studies of film flow on inclines or for sliding drops [11, 12].

However, an increasing number of experiments is not performed with simple but with complex liquids. Examples include the dewetting of drops and films of (evaporating) solutions of polymers or of nanoparticle suspensions [43–47], dewetting and decomposing films of polymer mixtures [48], the spreading of drops covered by soluble or insoluble surfactants [49, 50], the spreading of nanoparticle suspensions [51] and nematic liquid crystals [52]. Although in these systems the interfacial effects of capillarity and wettability are still main driving forces, they may now interact with diffusive transport of solutes or surfactants, phase separation and other phase transitions, evaporation/condensation of solvent and complex rheologies. The theoretical description of many of the observed dynamical processes is still far from complete.

In the present note we discuss a possible way to develop thin film descriptions for solutions and suspensions. We first sketch a simple version of the corresponding thin film model employing its “hydrodynamic formulation”, i.e., a formulation similar to Eq. (1). Second, we re-formulate the model as a gradient dynamics, i.e., in a form similar to Eq. (2). One may call this the “thermodynamic formulation”. Then it is explained how the latter formulation may be employed to extend the use of the evolution equations towards solute molecules/particles with net attractive interactions, decomposing solute-solvent systems and a solute-dependent wettability. Finally we point out some resulting new questions, limitations of the approach, and discuss some literature models in the context of the present work.

2 Hydrodynamic formulation

Consider a thin film of an (evaporating) partially wetting suspension or solution in contact with its vapour on a flat solid substrate. If all surface slopes and lateral gradients are small one may employ a long-wave approximation [12] and derive two coupled evolution equations for the film thickness profile $h(x, y, t)$ and the vertically averaged solute concentration field $\phi(x, y, t)$:

$$\partial_t h = \nabla \cdot [Q(h, \phi) \nabla p(h)] - \frac{\beta}{\rho} (p(h) - \mu \rho), \quad (4)$$

$$\partial_t (\phi h) = \nabla \cdot [\phi Q(h, \phi) \nabla p(h)] + \nabla \cdot [D(\phi) h \nabla \phi]. \quad (5)$$

Here the mobility is $Q(h, \phi) = h^3/3\eta(\phi)$. The dynamic viscosity may be a constant $\eta(\phi) = \eta_0$ or exhibit a (possibly strongly non-linear) dependence on the local solute concentration as, e.g., given by the Krieger–Dougherty law [53, 54]. Note that ϕ is a dimensionless *per volume* concentration. The first term on the right hand side of Eq. (4) (conserved dynamics) corresponds to convective transport of the liquid whereas the second term (non-conserved dynamics) models evaporation. The convective flow is driven by the gradient of the pressure

$$p(h) = -\gamma \Delta h - \Pi(h), \quad (6)$$

discussed in Section 1. The second term in Eq. (4) assumes the system is close to equilibrium. In this limit evaporation with a rate β is driven by the difference of the scaled pressure p/ρ and the chemical potential of the ambient vapour μ [55, 56]. Latent heat effects may be neglected, and the density ρ is assumed to be equal for particles and solvent. The first and second terms on the right hand side of Eq. (5) model convective and diffusive transport of the solute, respectively. At high concentration the diffusion coefficient may depend on concentration. We employ the Einstein-Stokes relation $D(\phi) = k_B T / 6\pi r_0 \eta(\phi)$, where k_B is the Boltzmann constant, T the temperature, and r_0 the particle radius.

Alternatively, one may define convective \mathbf{J}_{conv} , evaporative J_{evap} and diffusive \mathbf{J}_{diff} fluxes and write Eqs. (4) and (5) as

$$\partial_t h = -\nabla \cdot \mathbf{J}_{\text{conv}} - J_{\text{evap}} \quad \text{and} \quad \partial_t (\phi h) = -\nabla \cdot (\phi \mathbf{J}_{\text{conv}} + \mathbf{J}_{\text{diff}}). \quad (7)$$

Models related to Eqs. (4) and (5) are used in studies of particle-laden film flow (without evaporation or wettability effects, but on an incline and with density contrast between particles and solvent) [57], dewetting of suspensions [58], particle convection in an evaporating colloidal droplet (no solute diffusion, radial geometry) [59] and in studies of the deposition of regular and irregular line patterns of nanoparticles or polymers from a receding contact line [60, 61]. In the limit $\phi \rightarrow 0$, the model reduces to that used in [55] to study the fingering instability of a receding front of a dewetting and evaporating pure liquid.

3 Formulation as gradient dynamics

As the system is relaxational, i.e., the boundary conditions do not sustain energy or mass fluxes, we expect the system to approach a static equilibrium. Without evaporation the approach to equilibrium can be described by a gradient dynamics for the *conserved fields* film thickness $h(x, y, t)$ and local amount of solute $\psi(x, y, t) = h(x, y, t) \phi(x, y, t)$. As ϕ is a dimensionless *per volume* concentration, ψ is in units of length and can be seen as an effective local solute layer thickness.

Note that the height averaged concentration $\phi(x, y, t)$ is not a conserved field as $\int \phi(x, y, t) dx dy$ is not constant in time. The evolution equations are

$$\partial_t h = \nabla \cdot \left[Q_{hh} \nabla \frac{\delta F}{\delta h} + Q_{h\psi} \nabla \frac{\delta F}{\delta \psi} \right] \quad (8)$$

$$\partial_t \psi = \nabla \cdot \left[Q_{\psi h} \nabla \frac{\delta F}{\delta h} + Q_{\psi\psi} \nabla \frac{\delta F}{\delta \psi} \right] \quad (9)$$

with the symmetric positive definite mobility matrix

$$\mathbf{Q} = \begin{pmatrix} Q_{hh} & Q_{h\psi} \\ Q_{\psi h} & Q_{\psi\psi} \end{pmatrix} = \frac{1}{3\eta} \begin{pmatrix} h^3 & h^2\psi \\ h^2\psi & h\psi^2 + 3\tilde{D}\psi \end{pmatrix} \quad (10)$$

where

$$F[h, \psi] = \int \left[\frac{\gamma}{2} (\nabla h)^2 + f(h) + h g \left(\frac{\psi}{h} \right) \right] dA. \quad (11)$$

with

$$g(\phi) = \frac{k_B T}{a^3} \phi \log(\phi) \quad (12)$$

where a is a molecular length scale. The resulting variations are

$$\begin{aligned} \frac{\delta F}{\delta h} &= -\gamma \Delta h + \partial_h f(h) + g \left(\frac{\psi}{h} \right) - \frac{\psi}{h} g' \left(\frac{\psi}{h} \right) \\ \frac{\delta F}{\delta \psi} &= g' \left(\frac{\psi}{h} \right) \end{aligned} \quad (13)$$

For the corresponding gradients one obtains

$$\begin{aligned} \nabla \frac{\delta F}{\delta h} &= -\gamma \nabla \Delta h + \partial_{hh} f(h) \nabla h - \frac{\psi}{h} \left(\frac{\nabla \psi}{h} - \frac{\psi \nabla h}{h^2} \right) g'' \left(\frac{\psi}{h} \right) \\ \nabla \frac{\delta F}{\delta \psi} &= \left(\frac{\nabla \psi}{h} - \frac{\psi \nabla h}{h^2} \right) g'' \left(\frac{\psi}{h} \right). \end{aligned} \quad (14)$$

This implies that with $Q_{\psi h} = (\psi/h) Q_{hh}$ and $\tilde{Q}_{\psi\psi} = (\psi/h) Q_{h\psi}$ where we have split $Q_{\psi\psi} = \tilde{Q}_{\psi\psi} + \tilde{Q}_{\psi\psi} = (h\psi^2/3\eta) + \tilde{D}\psi/\eta$, the convective contributions to Eqs. (4) and (5) follow directly from Eqs. (8) and (9). The diffusive flux in Eqs. (5) [cf. (7)] corresponds to

$$\mathbf{J}_{\text{diff}} = -\tilde{Q}_{\psi\psi} \nabla \frac{\delta F}{\delta \psi} = -\frac{\tilde{D}\psi}{\eta} g'' \left(\frac{\psi}{h} \right) \nabla \left(\frac{\psi}{h} \right). \quad (15)$$

With Eq. (12) and $D = \tilde{D}k_B T/a^3\eta$, we have $\mathbf{J}_{\text{diff}} = -Dh\nabla(\psi/h)$, i.e., the correct diffusive flux in Eq. (5). Note that the definition of D agrees with Einstein's relation if $\tilde{D} = a^2/6\pi$ and $a = r_0$.

Note, that the variational formulation of the non-conserved part of Eq. (4) is not obvious. A term $\sim \delta F/\delta h$ correctly gives the contributions due to laplace and disjoining pressure. However, Eq. (13) implies the appearance of the further terms $g - \phi g'$ that have no counterpart in Eq. (4). If one accepts that some ϕ -dependent terms are missing in (4) the question remains what is the correct ratio of mobilities for the $\delta F/\delta h$ and $\delta F/\delta \psi$ terms.

Note that the form of the dynamical equations (8) and (9) is known from other relaxational physical systems where two conserved fields evolve in a coupled way. Another thin film example is the evolution of two-layer liquid films [62,63]. This will allow for the use of general results for such systems as e.g. on the linear stability of homogeneous states.

4 Extensions

The established variational form for the conserved part of the dynamics allows us to discuss extensions of the formalism in the case without evaporation. We base such extensions on changes in the energy functional $F[h, \psi]$ thereby implicitly assuming that the mobilities do not change. Here we briefly discuss two possibilities: (i) the incorporation of solute-solute and solute-solvent interactions within the fluid, and (ii) the incorporation of a solute-dependent wettability.

(i) With Eq. (12), $g(\phi)$ only accounts for entropic contributions to the local free energy, i.e., one assumes the solute molecules/particles do not show any net attractive interactions between them. They only have excluded volume interactions. One may introduce attractive interactions in the form of additional contributions to g . The additional contribution g_{int} would normally be nonlinear and therefore result in effectively ϕ -dependent diffusion “constants” $D_{\text{eff}} = D(1 + \phi g''_{\text{int}})$ in Eq. (5). For instance, for an attractive interaction $g_{\text{int}} = -\varepsilon\phi^2/2$ with $\varepsilon > 0$, $D_{\text{eff}} = D(1 - \varepsilon\phi)$ is reduced for increasing concentration and may become negative in the case of solvent-solute decomposition. To discuss such a decomposition one may furthermore include a gradient contribution for the concentration in F , i.e., a term proportional to $h(\nabla\phi)^2 = h(\nabla(\psi/h))^2$. Doing this and replacing g by a double-well potential ($\sim(\phi^2 - 1)^2$), one obtains from Eqs. (8) and (9) instead of Eqs. (4) and (5) the thin film limit of model-H (Korteweg-Navier-Stokes equation coupled to convective Cahn-Hilliard equation [64–66]). The thin film equations obtained in that way agree with the ones that were recently derived from model-H employing a long-wave expansion [67], if one takes into account that the double-well potential arises from a Taylor expansion about some concentration value. This also implies that $\tilde{Q}_{\psi\psi}$ is proportional to h (and not to ψ as above in Eq. (10) of the general formulation). This involves some subtle issues regarding the formulation of model-H itself that should be further discussed.

(ii) The contribution in F [Eq. (11)] that accounts for wettability is $f(h)$. It is related to the disjoining pressure by $\Pi(h) = -\partial_h f$. When employing $f(h)$ for a film or drop of a solution or suspension one assumes that the wettability of the film does not depend on the solute concentration. This might not be a good approximation at high solute concentrations, e.g., close to contact lines of evaporating solutions or for spreading nanofluids. With the presented variational formulation (8) there exists a way to account for the influence of the solute on wettability: One may amend f by including an explicit dependence on ϕ , i.e., one employs $f(h, \phi) = f(h, \psi/h)$ in (11). Then the convective flux becomes

$$\begin{aligned} J_{\text{conv}} &= -Q_{hh}\nabla\frac{\delta F}{\delta h} - Q_{h\psi}\nabla\frac{\delta F}{\delta\psi} \\ &= Q_{hh}[\nabla(\gamma\Delta h - \partial_h f(h, \phi)) + \frac{1}{h}(\partial_\phi f(h, \phi))\nabla\phi]. \end{aligned} \quad (16)$$

The diffusive flux becomes

$$J_{\text{diff}} = -\tilde{Q}_{\psi\psi}\nabla\frac{\delta F}{\delta\psi} = -\tilde{D}\psi\nabla\left[\left(\frac{1}{h}\partial_\phi f(h, \phi)\right) + g'(\phi)\right]. \quad (17)$$

Incorporating the amended fluxes into Eqs. (7) gives a coupled pair of evolution equation for a solution with a solute-dependent wettability.

5 Conclusions

In this note we have presented a gradient dynamic formulation of the coupled thin film evolution equations that are employed to describe thin films of solutions or

suspensions in the long-wave limit. We have further argued that based on the reformulation one may incorporate additional “energetic” effects directly into the thin film model. The discussed examples have been (i) solute-solute and solute-solvent interactions and (ii) a solute-dependent wettability. It is encouraging that the approach has for instance allowed us to obtain the thin film equations for coupled decomposition and dewetting of a film of a binary mixture that are in [67] derived from model-H employing a formal long-wave approximation.

Our approach allows us to discuss a number of approaches taken in the literature for similar systems:

(1) A dynamical model for the coupled decomposition and dewetting of a thin film of a binary polymer blend is proposed in [68] based on an energy functional developed in [69]. Their functional is similar to (11) together with extensions (i) and (ii). However, the dynamical equations in [68] do not agree with our Eqs. (8–9) as they express the time evolution of h and $\psi = h\phi$ in terms of constrained functional derivatives with respect to the conserved h and the non-conserved ϕ resulting in integro-differential evolution equations. We are not able to see how these equations will in the limit of “non-interacting” molecules result in the hydrodynamic equations (4) (without evaporation) and (5).

(2) Another set of works [70,71] employs particle-concentration dependent structural disjoining pressures (cf. [4,72]) to model the spreading of nanofluids [51], i.e., of suspensions of nanoparticles. In Refs. [70,71], is done by replacing the disjoining pressure $\Pi(h)$ in Eqs. (4) and (5) by an expression $\Pi(h, \phi)$ depending on film thickness and local height-averaged particle concentration. As a result of our discussion in point (ii) above, it becomes clear that one needs to include additional terms into the evolution equations for film thickness and concentration to obtain a consistent description.

Note finally that although we think that the present variational formulation allows for the incorporation of several important physical effects into thin film models, it also poses questions that need further discussion. For instance, it is not obvious how one correctly models evaporation (close to equilibrium) by non-conserved Allen-Cahn-type terms in Eq. (8). Another question concerns the correct incorporation of a solute-dependent capillarity (solutal Marangoni effect) in a way consistent with the solute-dependent wettability. Here, we have separated the two effects and only discussed the latter.

To conclude, we believe that the presented gradient dynamics equations and their future extensions will help to improve our understanding of various dynamic phenomena in free surface films and drops of complex liquids. The present drive towards further miniaturisation of fluidic systems towards micro- [73] and eventually nanofluidic devices [74] implies that such understanding will in the future become even more important as one needs to gain control of the various interfacial effects.

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