

Modelling of thin liquid films - asymptotic approach vs. gradient dynamics

Prof. Uwe Thiele (Institut für Theoretische Physik & Center for Nonlinear Science,
Westfälische Wilhelms-Universität Münster, Germany)

Prof. Neil Balmforth (Department of Mathematics, University of British Columbia,
Vancouver, Canada)

Prof. Andrew Hazel (School of Mathematics & Manchester Centre for Nonlinear Dynamics,
University of Manchester, UK)

Prof. Chun Liu (Department of Applied Mathematics,
Illinois Institute of Technology, Chicago, USA)

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1 Overview of the Field

Thin films and shallow droplets of simple liquids on solid substrates have for many years been described quite successfully by thin-film evolution equations (often called lubrication or long-wave equations) derived from the basic transport equations of physico-chemical hydrodynamics employing a long-wave approximation [13, 2]. In the more recent years the approach has been extended to complex liquids. The wide variety of systems that are considered include relatively simple nonvolatile molecular liquids, volatile liquids, liquids with insoluble and soluble surfactants, mixtures of simple liquids, particle suspensions, liquid crystals and a number of non-Newtonian liquids. Film and drop dynamics is described for a number of distinct physical systems where the system either approaches equilibrium states or may remain in out-of-equilibrium states that are driven by persistent fluxes. Examples include the dewetting dynamics of thin films on horizontal or structured solid substrates, the behaviour of drops and films inside (rimming flow) and outside (coating flow) of a rotating cylinder, the spreading of surfactant solutions, the transfer of liquid from a bath onto a moving plate, particle-laden liquid films flowing down an incline, morphological transitions of individual sliding drops and the behaviour of their ensembles, and osmotically spreading biofilms. Additional influences may be included, e.g., thermal effects resulting in thermal Marangoni flows, chemical or topographical substrate heterogeneities, slip of the liquid at the solid substrate, phase transitions as evaporation/condensation, dissolution or deposition, and the dynamics of the surrounding phases.

2 Recent Developments and Open Problems

In most cases, thin-film models are derived by employing a consistent asymptotic procedure, i.e., a small parameter ε is introduced that corresponds to the ratio of typical length scales orthogonal and parallel to the substrate (e.g., equilibrium contact angle, plate inclination, ratio of film thickness and cylinder radius) and governing equations and boundary conditions are expanded in ε to derive evolution equations to different

order for the local film height, h , and other adequate order parameter fields (e.g., concentration fields). These are normally 4th order partial differential equations. In the case of a simple nonvolatile dewetting liquid on a smooth solid substrate under the sole influence of capillarity and wettability the equation reads

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

where η and γ stand for the dynamic viscosity and surface tension of the liquid and $\Pi(h) = -df(h)/dh$ is the Derjaguin (or disjoining) pressure that encodes the wettability of the substrate and is related to the wetting potential $f(h)$ [18]. It was noted early on by Oron & Rosenau as well as by Mitlin [14, 11, 19] that such equations can often be written as gradient dynamics for the conserved order parameter field h , namely, as

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

where $Q(h) = h^3/3\eta$ is a mobility function and $F[h]$ is the free energy functional (at the same time a Lyapunov functional)

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

that consists of contributions of surface energy (in long-wave or small-gradient approximation) and wetting energy (in Mitlin's case [11, 19]). The latter formulation brings the thin-film equation into the wide class of gradient dynamics models (for conserved fields) that also includes the Cahn-Hilliard equation, phase-field crystal equations, models for epitaxial growth [6, 19] and models for membrane dynamics — implying that addressing important general questions for one of these models will affect our knowledge regarding the others. These models can often be directly derived based on deeper thermodynamic principles such as Onsager's variational principle [4, 5].

However, although over the years quite a number of thin-film models have been derived employing asymptotic methods for various liquids in relaxational settings (i.e., without flow of energy or mass across the system boundaries) [13, 2, 12] gradient dynamics forms have been discussed for a few models only and these studies are typically more recent. Most involve several coupled fields; examples include dewetting two-layer films [15, 9], films of mixtures [1, 21, 24] and surfactant covered films [20]. In the course of discussions at previous international workshops and between colleagues it became clear that beyond the simple case given above where asymptotic and gradient dynamics approaches are identical, frequently asymptotically derived models differ in important details from the gradient dynamics form. For instance, there exist correctly derived asymptotic models that can not be brought into a gradient dynamics form. As an example, appendix A of [20] reflects on such a discussion that came up during the Programme *Mathematical Modelling and Analysis of Complex Fluids and Active Media in Evolving Domains* at the Isaac Newton Institute for Mathematical Sciences (Cambridge, May–Aug 2013). Further, there exist many *ad-hoc* amendments to thin-film models (in this way incorporating additional physical effects) that can influence the gradient dynamics structure.

One such improvement to the lubrication approximation proposed in [7] and later by [17] consists of using the full curvature (as opposed to its linearised form) of the free surface (i.e., Laplace pressure term) which at first view one might discard as being in conflict with the standard lubrication approach. However, in a gradient dynamics context one may interpret this approach as employing a better approximation for the surface energy while not touching the approximation of the dynamics. This provides an interpretation of why it is a successful approach. At present, comparisons of full Stokes flow calculations with lubrication models of different order on the one hand and a gradient dynamics model on the other hand are underway for the dynamics of a film outside a rotating cylinder. They show that the latter more faithfully captures the correct dynamics and even qualitative behaviour over a wider range of control parameters [23]. This development in approximated hydrodynamic models for free surface flows is in itself quite intriguing, however, at the same time it parallels a tendency in general Soft Matter Science to consider dynamical models that can be derived with Onsager's variational principle [16, 4]. One aim of our workshop has been to discuss these approaches between the different communities.

A second aim has been related to the question concerning the origin of mesoscale and macroscale quantities employed in asymptotic and gradient dynamics models in the case of simple and complex liquids. For instance, the gradient dynamics approach places a large weight on the underlying energy functional $F[h]$.

However, in many cases the energies themselves are approximations that are only valid for part of the thickness range in which they are employed in practical calculations. For instance, most employed Derjaguin (or disjoining) pressures diverge for vanishing film height implying that a microscopic precursor film exists everywhere outside a macroscopic drop of partially wetting liquid on a solid substrate. Early on, it was discussed that a cut-off should be introduced [3], but this is infeasible in most thin-film models. Another more modern approach is to obtain the necessary parameters and functional dependencies directly from appropriate microscopic approaches as Molecular Dynamics simulations [10, 22] or classical Density Functional Theories [8]. This is, however, not yet realised beyond the case of simple liquids. The workshop has aimed at highlighting the importance of a seamless connection of microscale, mesoscale and macroscale models that should be reached in the future and at discussing pathways towards this aim.

3 Realisation of the workshop

During the workshop we have considered the two related questions that one has to resolve when establishing new thin film models: (i) Which modelling strategy should one follow, i.e., is it more important that a model is asymptotically correct or that it corresponds (in the appropriate limits) to a gradient dynamics on a physically reasonable energy functional? (ii) The second question is how one obtains the energetic and dynamic ingredients of a thin-film model in a consistent manner?

This aim we have pursued on the one hand with a classical workshop programme consisting of scheduled talks of the individual scientists that spoke about particular projects in areas closely related to thin films and drops on substrates; see the final list of participants in the appendix. Each speaker had been asked to relate their particular considered questions, methodology and results to the theme and to the main questions of the workshop. Each 20min talk was accompanied by 5min general discussion. This has worked out quite well as most participants related their work to the general question and the discussions picked up on these relations. We report on the scheduled talks in section 3.1 below.

On the other hand our programme contained ample time for discussion: informal ones during coffee and lunch breaks, dinner time and during the free afternoon, and more formal ones in the scheduled well-attended discussion sessions on Thursday afternoon and Friday morning. We reflect on these discussions in section 3.2 below.

3.1 Presentation Highlights

Each morning started with the talk contribution of one of the organisers. In particular, the Monday was opened by an overview talk by U. Thiele on gradient dynamics approaches for thin liquid films; on Tuesday C. Liu spoke about general diffusion and dynamic boundary conditions; N. Balmforth presented on Wednesday, describing thin-film models for viscoplastic fluid; on Thursday, A. Hazel gave a direct comparison of asymptotic and gradient dynamics approaches to coating and rimming flows discussing their advantages and disadvantages. In this way he directly compared the two main approaches for improvements that are discussed in the literature, namely going to asymptotic models of higher order or establishing gradient dynamics models with a focus on an improved description of the energetic aspects.

Overall, about half of the talks discussed specific thin-film models for relevant hydrodynamic systems that are employed for the analysis of different physical systems related to modern experiments and applications. Examples include the talks by C. Falcon, Y. Stokes, T.-S. Lin, M. Chugunova, V. Ajaev, S. Kumar, S. Wilson, I. Hewitt, R. Cimpeanu, T. Witelski and J.J. Feng. In the course of these talks it was discussed how to improve and apply these models, e.g., when either stretching the parameter region where they are applied or when adapting them to novel experimental situations or new materials. For instance it was pointed out that one may introduce thin-film models for suspensions first in the dilute limit and then to expand them to higher solute concentrations by improving the underlying energy functional of the gradient dynamics formulation of the hydrodynamic model. This was further elaborated in the discussion sessions. It was pointed out that individual *ad-hoc* amendments of diffusion constants, Marangoni forces, and Derjaguin pressures often result in inconsistent models. Examples of simple liquids in more complex situations were presented by M. Sellier (optimal pancake control), A.G. Gonzalez (breakup of liquid grids into regular drop patterns) and M. Fontelos

López (discrete self-similarity in thin-film rupture). The general mathematical structure of thin-film model was discussed by R. Krechetnikov.

In a number of presentations state-of-the-art models were presented for rather complex fluids and (biological) soft matter. A. Rey discussed the dynamics of soft anisotropic media as liquid crystals and hierarchically structured biomolecules, while K. John presented biofilm models that consists of a gradient dynamics for biomass in the film, biosurfactants and water with nutrients that is supplemented by bioactive terms. Y.-N. Young spoke about long-wave dynamics of a lubrication layer under an inextensible elastic membrane and S. Gurevich reported on spatio-temporal patterns in dynamic self-assembly systems based on surfactant, and the control of such systems. Control was also the subject of the contribution of A. Thompson on falling films. Deposition patterns of colloidal particles were reported by O. Manor — relating also to the talk of X. Man and U. Thiele. The talks of R. Cuerno and O. Pierre-Louis showed that also the evolution of solid surfaces (irradiated by an energetic ion beam or dissolving into a solution) is governed by equations of the same class and should therefore be seen in the same context of thin-film equations. There the existence and form of gradient dynamics formulations is not yet clear. M. Shearer and S. Li discussed aspects of flow and instabilities in Hele-Shaw cell.

The different approaches were also brought into the context of the wider recent development in Soft Matter Science to consider models for complex media that can be derived with Onsager's variational principle, i.e., based on variations of the Rayleighian, which consists of the rate of change of a free energy functional and a dissipation functional, with respect to rates/fluxes. X. Man discussed how this approach is used to study the drying of liquid droplets based on a few macroscopic degrees of freedom and D. Peschka presented a description of drop and contact line dynamics via generalised gradient flows.

Thin-film models are often mesoscopic or macroscopic models that rely on input from the microscale. While the procedure is well established for quantities as surface tension, only a few approaches are pursued for other quantities such as wetting potential and Derjaguin pressure that encode wettability or indeed transport coefficients. Approaches for complex liquids are still rather scarce. During the workshop, M. Müller described how to connect particle-based simulations to continuum models employing examples from simple and multicomponent polymer liquids, S. Hendy discussed MD simulations of droplets on tilted superhydrophobic and SLIPS surfaces and irradiation by an energetic ion beam. A. Archer reported on hybrid thin-film kinetic Monte Carlo modelling of droplets evaporating, coalescing and sliding on surfaces.

3.2 Discussion Highlights

During the talks, a number of relevant and fundamental questions were raised that required perspectives broader than those of a single speaker to answer. Among those, the participants selected the three main themes below for discussion by all participants in separate sessions. While these discussions rarely provide immediate answers, they are important to identify themes and future research directions for the research community.

3.2.1 Discussion on fundamental assumptions and choice of dissipation in gradient flows

Short contributions on the board by: D. Peschka (abstract gradient flows); U. Thiele (mesoscopic models); X. Man (Onsager's variational principle and Rayleighian).

Introduction: Energetic variational principles are a cornerstone of modern modelling approaches in complex physical systems and are related to many important concepts from fluid dynamics, chemistry, thermodynamics, and soft condensed matter systems. While being a general abstract framework, the importance of this concept justifies itself through the many particular examples presented during the workshop. While in many systems equilibrium theory makes the proper choice of the driving thermodynamic potential easy to understand, the concept of dissipation appears sometimes more elusive. Therefore, this discussion was aimed at providing different viewpoints for this concept and to provide a few examples.

Discussion: The first part of the discussion focussed on the general gradient flow formalism. Different points of view for the general construction were presented by different participants of the workshop. Ranging from more abstract mathematical approaches to specific finite-dimensional examples and despite slight

technical differences, the experts from different fields unanimously agree on the general structure. In this part, the discussion mainly aimed at understanding the restrictions of gradient based models, understanding the restrictions that one has when adding terms to the dissipation, and discussing different aspects of the benefit one has when recasting a known partial differential equation in the form of a gradient dynamics model. While it seemed rather difficult to address all the different aspects that were interesting to the audience, the importance of different dissipation mechanisms became clear for everyone. Aspects of modelling correction terms for dissipative effects were also briefly discussed. Specific examples that had been mentioned during the workshop include diffusion & convection, conserved and non-conserved order parameters (Cahn-Hilliard and Allen-Cahn equations), thin liquid films, Stokes flow with free boundaries, reactions/evaporation/condensation/drying/solidifications, dynamic contact angles, flows with heat, biological systems (tear film and cellular systems), interface energies, pattern formation and deposition, higher order energies (full curvature), nematics, non-Newtonian rheology, porous medium and Hele-Shaw flows.

Questions that have been addressed in particular are

1. What is the theoretical foundation (Onsager, Rayleigh, Helmholtz, ...)?
2. What are possible extensions, e.g., to inertial effects, higher order terms, complex fluids, non-Newtonian rheology, hysteresis and memory, ...?
3. What are limitations / what is the validity range of such structures?
4. What is the role of boundary conditions or kinetic boundary equations?
5. How does one deal with degeneracy in the dissipation?

3.2.2 Discussion of linking MD simulations and continuum mechanics with and without inclusion of fluctuations

Short contributions by: Lou Kondic (jumping droplets), Andrew Archer (kinetic Monte-Carlo and continuum equations), Marcus Müller (model hierarchy, sequence of approximations)

Introduction: While molecular dynamics (MD) is widely considered a first-principles approach, but it is computationally very expensive even for mesoscopic length and time scales. On diffusive and hydrodynamic time scales, it is prohibitively so. Hybrid approaches combining mesoscopic continuum models and microscopic MD or kinetic Monte-Carlo (KMC) models offer a viable alternative to speed-up simulations. But continuum models also benefit from the input of molecular dynamics simulations, which can provide much needed material-specific parameters and behaviours (e.g. constitutive laws) as input data.

Discussion: To motivate the discussion, a set of MD simulation videos showing droplets bouncing from a substrate and corresponding continuum simulations where shown and short contributions summarised some main aspects. Then possible ways of how the different modelling approaches can benefit from each other were discussed. Additionally, approaches that include fluctuations in continuum models were of interest to the participants, but were only briefly discussed.

After the discussion all participants could appreciate the way in which microscopic models can benefit meso- and macroscopic modelling, either by parameter passing or by hybridisation, e.g., by “dragging a MD simulation along via a mesoscale simulation” or via microlevel timesteps with continuum time integration. However, it also became clear that the coarse-grained MD and KMC simulations also contain a number of assumptions and tricky details that themselves need connection to and derivation from more precise models on smaller scales as, e.g., force fields obtained in atomistic MD simulations or electronic (quantum) Density Functional Theory (DFT); e.g., there is no simple toolbox where one can just input the material(s) (e.g., some alcane, water, polystyrene, DNA) whose dynamics one wants to study, and run MD simulations to obtain precise quantitative predictions.

Other mentioned issues relate to the use of thermostats and assumption of certain thermodynamic ensembles in the MD simulations. Some participants mentioned Phase-Field-Crystal methods, but this was not further elucidated.

Questions that have been addressed in particular are

1. What can continuum models learn from microscopic MD/DFT models?
2. Which macroscopic parameters are needed to set the parameters of MD/DFT models?
3. What are the relevant timescales?
4. How can the ability to quantitatively predict system behaviour be improved?

3.2.3 Discussion of disjoining pressures, particularly in “complex” situations

Short contributions at the board by: U. Thiele (overview); L. Kondic (spinodal dewetting); M. Sellier (reconstruction of disjoining pressure from traveling waves).

Introduction: There is a general agreement on some general properties of the disjoining pressure. For example, the energetic minimum defines the contact angle, which is a macroscopic measurable quantity. The thickness corresponding to the location of the minimum can be interpreted as a precursor or adsorption layer liquid film thickness and has been observed in some experiments on the nanometer scale.

The general form of long-range interaction form that is often used is suggested by the classical theory of Derjaguin & Lifshitz for van der Waals forces. However, in particular the values and the form for concrete complex physical systems (mixtures, particle suspensions, multilayer, polymers) are often chosen in a seemingly heuristic manner. In this discussion different approaches to choose and measure disjoining pressures and the importance of these different choices were discussed.

Discussion: In a short presentation, the standard form of the disjoining pressure for thin-films was presented and different variants, i.e., dependence on concentration, dependence on position on the substrate, oscillatory behaviour (multiple minima of the wetting energy), higher order terms, and the algebraic form of the potential were discussed. It was stressed that choices of wetting energies, in particular, for complex liquids have implications for the resulting gradient flow dynamics (consistency). For instance, employing a concentration- and film height-dependent wetting energy will result in a concentration- and film height-dependent disjoining pressure and other contributions that are vaguely similar to Marangoni fluxes (Korteweg fluxes).

Besides ample evidence for long-ranged interactions, the discussion again showed that there is still some deficit in the use of the calculated values for theoretical predictions. This is related to the above discussion of the relation of microscopic and mesoscopic models.

Another line of argument concerned the form of the wetting energy/disjoining pressure as a function of film height, as this implicitly assumes there is a closed film of constant density and well defined free surface. However, instead of as “film height”, alternatively, the independent variable can be interpreted as “adsorption”, i.e., the excess number of molecules per area where the gas density at coexistence provides the reference value. This is a more general concept as it does not imply a constant liquid density throughout the layer. For thick liquid films the two measures are proportional to each other, but for very thin films adsorption is more general as it also captures sub-monolayers of diffusing molecules or densities close to a hydrophobic solid that are lower than the gas density (negative adsorption).

Questions that have been addressed in particular are

1. What are the generally accepted theoretical foundations?
2. Are there any particular systems for which fine details of disjoining pressure plays a role?
3. How does one measure the disjoining pressure in a convincing way across all film heights and for all wetting properties?
4. How does one implement the disjoining pressure into a gradient flow dynamics consistently?
5. What are realistic disjoining pressures: rough surfaces, oscillations in height and lateral direction, line tension, impact of ions/electrical fields?
6. Does the slope of the interface have to be taken into account?

4 Outcome of the Meeting

All participants agreed that the workshop was very instructive and resulted in many interesting discussions between participants of various scientific backgrounds that will influence the future direction of their research. The combination of individual presentations and discussion sessions allowed us to work out the problems many people are concerned with. In this way it became clear that there is a number of important questions, like the coupling of microscopic and mesoscopic models where competing approaches develop and many questions still remain open.

These questions are being right now increasingly discussed in a number of contexts and the participants believe that the ongoing “miniaturisation” of soft matter and fluidic systems considered by the scientific community will result in an ever increasing importance to solve the discussed problems. The workshop has already helped the scientists from the different communities to further develop a common understanding of the challenges in the development of meso- and macroscale thin-film models for complex liquids and of their self-consistency and consistency with neighbouring microscopic approaches.

We believe that during the workshop the participating scientists have established the basis for new stable working collaborations.

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