Project proposal SPP 2171

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Experimental and numerical study of wetting on liquid-infused surfaces: the role of surfactants

State of the art and preliminary work

State of the art

Liquid-infused surfaces (LISs), consisting of a micro texture impregnated with liquid (in the following referred to as "oil" or "lubricant"), were first suggested by David Quéré in 2005 [C37]. After the group of J. Aizenberg demonstrated a number of promising applications of LISs in 2011 [C52], they have become increasingly popular, and various unique properties of such surfaces have been confirmed in the past few years. For example, it was shown that droplets are highly mobile on LISs [C43], which relates to their self-cleaning properties [C52]. Furthermore, LISs promote condensation [C2] and prevent the formation of ice layers [C26]. LISs are prototypes of flexible surfaces. Since ideally a droplet on the surface is only in contact with the oil and not with the texture, the oil film gets deformed by surface tension forces and forms a rim around the foot of the droplet. LISs are also candidates for adaptive surfaces, since a component dissolved in the droplet can be extracted to the oil film, thereby modifying its properties. This effect appears to have been largely unexplored. Last but not least, LISs qualify as switchable or active surfaces, which has been demonstrated using ferrofluids as impregnating liquids [C25]. Recently, the application scope of LISs was expanded by establishing textured surfaces displaying regions impregnated with different oils [C36]. That way it becomes possible to form liquid-infused regions of almost arbitrary geometric shape on a background impregnated with a different oil. Specific structures in that context are oil-in-oil channels through which liquid can be pumped [C48], rendering such surfaces switchable and active. The primary aim of the proposed project is to study the dynamics of droplets on LISs, both experimentally and by numerical simulations. While the area of numerical simulations of wetting on LISs is still at its infancy, a number of experiments on droplet transport and actuation on LIS have already been performed. For example, electrowetting-on-dielectric was demonstrated [C4], droplets could be transported using thermocapillary stresses [A1], [C7] or surface-acoustic waves [C31]. One of the most elementary aspects of dynamic wetting is the friction coefficient of a droplet translating along a LIS. Surprisingly, this has remained an open question for quite a long time. In a recent paper it was shown that the rim around the droplet makes the most significant contribution to the friction coefficient for the case that the oil viscosity is much higher than the droplet viscosity [C24]. Correspondingly, the droplet velocity scales as the driving force to the power of 3/2.

In modeling and numerical simulation of this class of flow problems one has to deal with multiple phases (oil, water, air), moving contact lines (MCL) and potentially surfactants. A further level of complexity is added by the fact that droplet transport on LISs is inherently three-dimensional. In modeling such multiphase flow problems one usually distinguishes between diffuse interface and sharp interface models. This project is restricted to a class of sharp interface models. Related to numerical simulation, specific difficulties in this class of sharp interface multiphase flow models are the following. Usually the sharp interface Γ is only implicitly known (level set technique) and an accurate numerical approximation of quantities that depend on Γ, such as surface tension or surfactants, is a difficult task. The surface stress tensor $\boldsymbol{\sigma}_\Gamma$ is localized on the interface and depends on the curvature of Γ and a (surfactant dependent) surface tension coefficient. Due to these surface stresses the pressure is discontinuous across the (moving) interface Γ. A notorious difficulty in this field is an accurate discretization of the flow problem with sufficiently small parasitic velocities generated by the pressure jump across the interface. The MCL problem we are facing here differs from the usual MCL problem, since we have a liquid-liquid-gas contact line, so there is no contact-line singularity. For modeling the MCL one can use curvature based restoring contact line forces (cf. work package H2 in section 2.3.1 below), which then require the discretization of source terms localized on the moving contact line. An accurate numerical approximation of these interface and line forces, which is of major importance for the accuracy of the fluid dynamics simulation, is difficult to realize. For the numerical simulation of the surfactant transport one has to solve a convection-diffusion equation on the evolving interface.

Already the two-phase flow problem, e.g. an oil droplet in water, is from a numerical simulation point of view a very difficult one. An overview of recently developed numerical methods in this field is given in [A4], [C50]. Validated three-dimensional numerical simulation results for such flow problems are very rare. For a slightly more difficult two-phase flow problem, namely a Taylor flow (air bubble in water), such validated simulations are presented in [A3]. In many papers on MCLs, one uses numerical simulations for relatively simple model problems to validate the modeling approach or to illustrate certain physical phenomena, e.g., [C10, C27, C38, C41, C54, C56]. Other papers consider one particular numerical method, often for a restricted small class of specific models [C1, C14, C20, C46, C47]. There are a few papers which treat finite element discretization techniques specifically designed for MCL problems, e.g., [C18, C19, C32, C51], [A7].

For numerical simulation of (insoluble) surfactants in multiphase flow one has to solve a convection-diffusion equation on an evolving surface. In the past two decades the study of numerical methods for PDEs on surfaces has been a rapidly growing research area. An overview on the now widely used so-called surface-FEM is given in [C15]. Another approach has been introduced in [C12] and builds on the ideas of [C6]. The method in that paper applies to cases in which the surface is given implicitly by some level set function and the key idea is to solve the partial differential equation on a narrow band around the surface. Unfitted finite element spaces on this narrow band are used for discretization. Another surface finite element method based on an outer (bulk) mesh has been introduced in [C35]. The main idea of this method is to use finite element spaces that are induced by triangulations of an outer domain to discretize the partial differential equation on the surface.

We are not aware of any paper in which a numerical method for solving a multiphase flow problem with MCL coupled with (insoluble) surfactant transport is systematically studied.

Preliminary work

PI Hardt started his activities in the field of LISs in 2013. In the resulting journal publication [A1] an especially simple method of fabricating LISs is described. This method allows keeping the oil at the surface even without textures. The resulting LIS shows properties very similar to those of textured liquid-infused surfaces. Especially, the surface displays self-healing behavior, and droplets show a very low level of contact-angle hysteresis. It was also demonstrated that droplets can be transported using temperature gradients, which was later examined in more detail [C7]. One of the advantages of corresponding LISs is that they may be transparent, such that imaging through the substrate from below becomes possible. Since then, corresponding silicone-oil infused surfaces have been used quite frequently in experiments performed in the group of PI Hardt. One main research activity in that context was the study of superposed liquid films, where one of the films is a LIS (see, e.g. [A5]). Parallel to the experimental activities, there were theoretical activities focusing on computing the hydrodynamic drag coefficient of LISs (see, e.g. [A10]).

In 2018, two new research activities addressing LISs were started in the group of PI Hardt. In the first one, it was shown that droplets can be translated along LISs using electric fields in a no-contact mode. The corresponding experimental setup is shown in Fig. 1(a). An aqueous droplet (typical volume 10 μL) is sitting on a LIS that is electrically contacted on its backside. The counter electrode is a metal pin a distance away from the droplet. If the electric field is sufficiently strong and the pin electrode is displaced, the droplet follows the electrode. This stands in contrast to droplet actuation by electrowetting-on-dielectric that requires a contact between the droplet and the counter electrode. Fig. 1(b) shows a comparison between the experimental data for the force on a droplet as a function of the horizontal displacement between the droplet and the electrode and the corresponding simulation data. A journal publication describing the results on droplet transport using electric fields is currently in preparation.

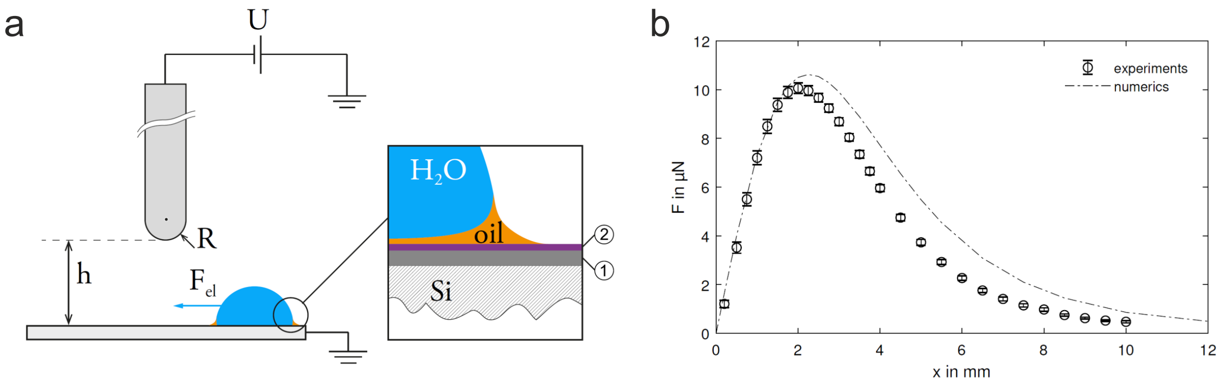


Figure 1: (a): Experimental setup for studying the electric-field induced displacement of droplets on LISs. In most of the experiments, the substrate moves driven by a linear translation stage. (b): Comparison between the experimental results and the simulation data for the force on a droplet as a function of horizontal displacement between the droplet and the pin electrode.

The second current research activity addressing LISs is a cooperation with the group of Pavel Levkin from the Karlsruhe Institute of Technology (KIT). The group of P. Levkin has recently succeeded in fabricating textured surfaces displaying regions impregnated with different lubricants [C36]. These lubricants may or may not show cloaking behavior, which means that they may or may not cover a water droplet sitting on the surface with a thin oil film. In terms of this cooperation, experiments were performed in which the transition of a water droplet between two regions impregnated with different lubricants was studied (see Fig. 2(a)). One of the lubricants was Krytox, a fluorinated oil. Krytox is more hydrophobic than the other lubricant, therefore a water droplet needs to overcome an energy barrier when penetrating into the Krytox-infused region. The experimental data were compared with the results of a simple theoretical model. Fig. 2(b) shows a comparison between the experimental and the theoretical data for the case that a non-cloaking lubricant (mineral oil) is used besides Krytox. The force available for overcoming the energy barrier between the two regions can be varied by varying the tilt angle α. The results are plotted in form of the maximum droplet volume that can be held back by the energy barrier (critical volume) as a function of α. The reasonably good agreement between the experimental and the theoretical data shown in the figure can only be achieved when assuming in the model that the droplet is cloaked by a thin Krytox film. This indicates that as soon the droplet comes in contact with the Krytox-infused region, cloaking happens. A journal publication describing the results of the cooperation with the group of P. Levkin is currently in preparation.

An experimental technique that will be of key importance for the work program described in section 2, although not immediately related to liquid-infused surfaces, is fluorescence imaging. PI Hardt has extensively used fluorescence imaging techniques in the past decade, for example for single molecule imaging (see, e.g. [A9]).

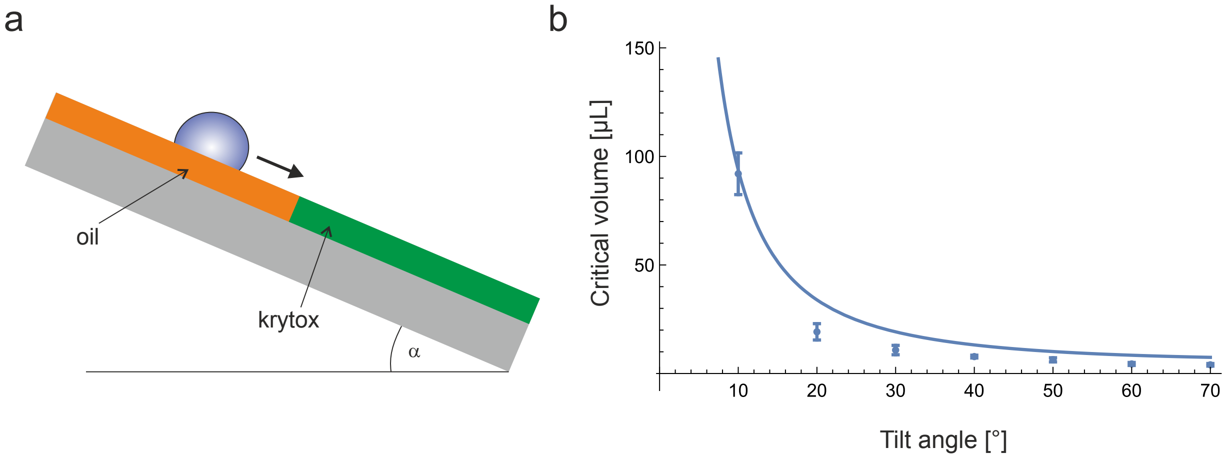


Figure 2: (a): Setup for studying the energy barrier between two liquid-infused regions. (b): Critical droplet volume as a function of tilt angle. In part b, the experimental data appear as symbols, the data from the theoretical model as the solid line.

Preliminary work on modeling and numerical simulation of multiphase flows and of flows with MCLs has been published by both the group of PI Hardt and of PI Reusken. In the past few years, the work of PI Hardt on formulation of mathematical models for various microscale flow and transport phenomena especially concentrated on free-surface flows, wetting, electrokinetics, electrohydrodynamics as well as the kinetic theory of gases. Examples of special relevance for the proposed research are thin-film flows and wetting phenomena. For example, in context with thin-film flows a model was formulated allowing to couple the solution of the thin-film equations with the Navier-Stokes equation for the bulk fluid [A5]. Referring to wetting phenomena, it was studied how the electric-double layer on a solid surface influences the static contact angle of a liquid wetting the surface and especially to which degree it contributes to the line tension [A2]. In recent years in the group of PI Reusken a finite element based code (DROPS, [C13]) for the simulation of two-phase incompressible flow problems has been developed [A4]. In the past decade results on the development, analysis and application of (new) methods for two-phase incompressible flows are presented in [A4] and a series of papers, e.g., [A6–A8], [C5, C16, C21, C22, C34]. We outline some main features of the numerical methods that are used in DROPS. A sharp interface approach is applied in the sense that there is no smoothing or regularization of discontinuities or delta functions at the interface. A level set method for interface capturing is used. For reparametrization of the level set function an improved fast marching technique is applied. A multilevel hierarchy of tetrahedral triangulations is used. Local refinement and coarsening routines are available. For spatial discretization finite element methods are applied. Special Cut-finite element spaces (CutFEM, cf. [C9]) suitable for functions that are discontinuous across the interface have been developed. A special Laplace-Beltrami method for the discretization of surface tension forces has been developed and analyzed [C22]. Fully implicit time integration methods are used, in which flow variables, surface tension forces and the level set function are strongly coupled. The Henry interface condition in a two-phase flow mass transport equation is treated by Nitsche's method. The spatial discretization of the surfactant transport equation on the interface is done by a CutFEM technique [A6]. For the solution of the resulting large sparse linear systems preconditioned Krylov subspace methods are applied. Several efficient preconditioners have been developed and implemented. For example, inexact Uzawa type solvers for saddle point problems and multigrid solvers are available. A message passing interface (MPI) based parallel version of DROPS has been developed in collaboration with the Chair of Scientific Computing, RWTH Aachen. In several papers and in joint projects within the SPP 1506 (cf. [C8]) it has been shown that the DROPS package is an efficient finite element and level set based solver for two-phase incompressible flow problems. All DROPS simulations are spatially three-dimensional. DROPS has been applied for the simulation of two-phase fluid dynamics, coupled with mass transport (e.g., [C28, C30]), with Marangoni effects and surfactant transport (e.g., [C55], [A8]) and with moving contact lines [A7]. Validation results in which numerical simulations are compared with experiments are presented in [A3], [C5, C33]. DROPS simulation results from [C28, C55] are shown in Figures 3, 4.

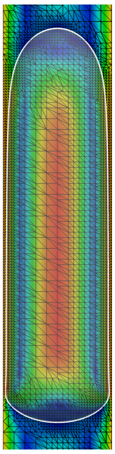


Figure 3: Droplet cross section from a 3D Taylor flow simulation.

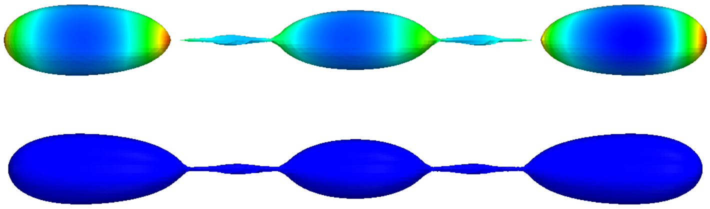


Figure 4: Droplet breakup with/without surfactants.

Project-related publications

Articles published by outlets with scientific quality assurance, book publications, and works accepted for publication but not yet published

[A1] A. A. Eifert, D. Paulssen, S. Varanakkottu, T. Baier, and S. Hardt. Simple fabrication of robust water-repellent surfaces with low contact-angle hysteresis based on impregnation. *Advanced Materials Interfaces*, 1:1300138, 2014.

[A2] A. Dörr and S. Hardt. Line tension and reduction of apparent contact angle associated with electric double layers. *Physics of Fluids*, 26:082105, 2014.

[A3] C. J. Falconi, C. Lehrenfeld, H. Marschall, C. Meyer, R. Abiev, D. Bothe, A. Reusken, M. Schlüter, and M. Wörner. Numerical and experimental analysis of local flow phenomena in laminar Taylor flow in a square mini-channel. *Physics of Fluids*, 28:012109, 2016.

[A4] S. Gross and A. Reusken. *Numerical Methods for Two-phase Incompressible Flows*. Springer, Berlin, 2011.

[A5] I. Nejati, M. Dietzel, and S. Hardt. Conjugated liquid layers driven by the short-wavelength Bénard–Marangoni instability: experiment and numerical simulation. *J. of Fluid Mechanics*, 783:46–71, 2015.

[A6] M. A. Olshanskii, A. Reusken, and X. Xu. An Eulerian space-time finite element method for diffusion problems on evolving surfaces. *SIAM J. Numer. Anal.*, 52:1354–1377, 2014.

[A7] A. Reusken, X. Xu, and L. Zhang. Finite element methods for a class of continuum models for immiscible flows with moving contact lines. *Int. J. Numer. Meth. Fluids*, pages 268–291, 2016.

[A8] A. Reusken and Y. Zhang. Numerical simulation of incompressible two-phase flows with a Boussinesq-Scriven surface stress tensor. *Numerical Methods in Fluids*, 73:1042–1058, 2013.

[A9] T. Roy, K. Szuttor, J. Smiatek, C. Holm, and S. Hardt. Stretching of surface-tethered polymers in pressure-driven flow under confinement. *Soft Matter*, 13:6189–6196, 2017.

[A10] C. Schönecker, T. Baier, and S. Hardt. Influence of the enclosed fluid on the flow over a microstructured surface in the Cassie state. *J. of Fluid Mechanics*, 740:168–195, 2014.

Objectives and work programme

Anticipated total duration of the project

Three years. Starting date 1.10.2019

Objectives

A main objective of the proposed research will be to achieve a comprehensive understanding of dynamic wetting on LISs. A special focus will be on the role of surfactants. In that context, true surfactants will be considered, but also the surfactant-like nature of lubricants with a positive spreading coefficient on water will be studied. The key question is how the liquid-like nature of the wetted surface gives rise to contact-line configurations, contact-angle dynamics and friction mechanisms different from those characteristic for a solid surface. The proposed project combines experiments with numerical simulations in a close cooperation between the research groups of PI Hardt and PI Reusken. A further main objective of this project will be the development and validation of appropriate mathematical models for this class of flow problems and of finite element based tailor-made simulation methods. Understanding the wetting scenarios will be a prerequisite for enabling adaptive and switchable wetting based on LISs.

Work programme incl. proposed research methods

Work packages PI Hardt

Apart from work package H2, the work packages of PI Hardt mostly address experimental tasks. The key objective is the detailed study of dynamic wetting on LISs with a special focus on surfactants. On the one hand, true (insoluble) surfactants will be considered. On the other hand, the surfactant-like nature of lubricants with a positive spreading coefficient on water will be studied. Work packages H2 to H4 form the basis of the cooperation with PI Reusken.

H1: *Establishing the experimental setup.*
The experimental setup for the proposed research is based on the existing setup shown in Fig. 1(a). The experiments on electric-field induced droplet transport on LISs are conducted in such a way that the pin electrode remains fixed and the substrate performs a translational motion. That way it is possible to keep the droplet at a fixed position in the lab frame (up to velocities of some centimeters per second) while it is moving relative to the substrate. This opens up the opportunity to study dynamic wetting using comparatively slow imaging techniques such as fluorescence imaging. The goal will be to image the distribution of fluorescent surfactants at the different fluid interfaces and/or the distribution of lubricants stained with a fluorescent dye. The imaging technique employed in that context will be light-sheet imaging. A thin light sheet cutting through the droplet excites the fluorescence of the surfactants or the fluorescent dye in the oil. The fluorescence signal from the light sheet is recorded using a long-distance microscope under a viewing angle of 90° relative to the light sheet. That way it will be possible to study the surfactants and/or the lubricant in a dynamic

wetting scenario slice by slice. The corresponding setup is shown in Fig. 5(a). All components are already available from the existing setup, with the exception of the optics creating the light sheet and the imaging components. For designing the light-sheet optics, the web platform available from the open access OpenSPIM project (openspim.org) will be utilized. OpenSPIM gives detailed instructions on establishing specific light-sheet imaging setups based on off-the-shelf components. A 3D CAD drawing of a setup suitable for the imaging of droplets on LISs is shown in Fig. 5(b). The light source is a laser diode with an emission wavelength compatible with the fluorescent dye. The light sheet is created by passing the laser light through a slit and finally focusing it with an objective. Imaging is performed using a long-distance microscope and a sensitive CCD camera, equipped with a bandpass filter. The typical size of a droplet will be a few millimeters. Correspondingly, the near-planar area of the light sheet needs to have a similar extension, which translates into a minimum light sheet thickness of 10 μm.

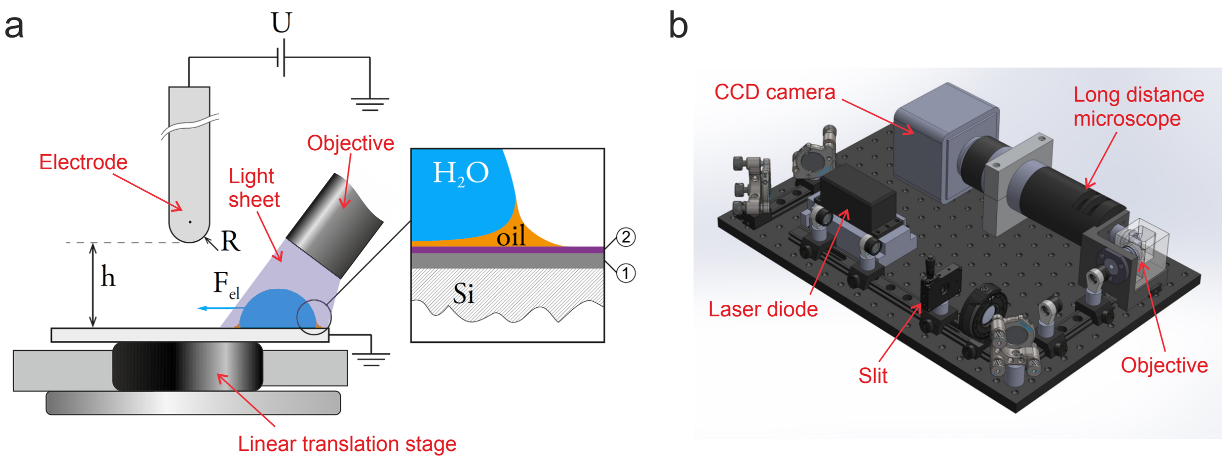


Figure 5: (a): Key components of the setup for studying dynamic wetting on LISs. (b): Optical setup enabling light-sheet imaging.

It needs to be emphasized here that establishing suitable imaging conditions and optimizing these conditions is a non-trivial task and part of the research work. It has already been demonstrated that the fluorescence imaging of lubricants on LISs [C43] and that of surfactant monolayers [C17] is feasible. While this shows that essentially the experiments outlined here are realistic, it remains a challenge to identify suitable fluorescent dyes and surfactants for a lubricant under consideration. A major criterion is a high enough quantum yield of the fluorescence markers to ensure a sufficient intensity. Furthermore, it is important that the surfactants are only found at the fluid interfaces and that the fluorescent dye for the lubricant does not diffuse into the droplet. For the corresponding optimization of the imaging conditions, separate experiments will be performed.

H2: *Mathematical models for liquid and surfactant transport.*

A key challenge lies in formulating a mathematical model for the MCL. Different from the classical MCL problem, in the case of wetting on a LIS, three fluid phases meet at the contact line. This eliminates the problem of the contact line singularity. Before considering moving contact lines on a LIS, it is instructive to study the case of an equilibrium contact line. According to Neumann's equation, the contact line can be regarded as a material object at which three interfacial tension forces need to balance, i.e., $\boldsymbol{\sigma}_{\rm wo} + \boldsymbol{\sigma}_{\rm wg} + \boldsymbol{\sigma}_{\rm og} = 0$, where the interfacial tensions are vector quantities in the two-dimensional space normal to the contact line. This it is depicted in Fig. 6, which also shows linear extensions of the three interfaces beyond the contact line as dashed lines.

For a MCL, however, Neumann's law does not hold, since the fluid interfaces get deformed by hydrodynamic forces. The level of interface deformation is determined by a balance of hydrodynamic forces and the restoring forces due to the interfacial curvature, i.e., the Laplace pressure. This is depicted in the inset of Fig. 6 for the example of the water-oil interface. The dashed line indicates the equilibrium interface, the vector indicates the interface orientation for an MCL. The curvature of the interface, only assuming nonzero values in the region marked by a red circle, can be numerically evaluated using techniques developed in the group of PI Reusken, cf. work package R3. The restoring forces at a MCL can be computed in an analogous manner (for the case of one of the phases being a solid) in [C49].

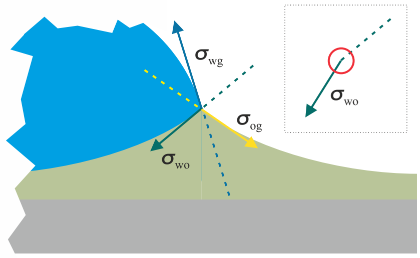


Figure 6: Force balance at the three-phase contact line according to Neumann's law. The inset shows the orientation of the water-oil interface for a MCL.

Apart from that, when considering surfactant transport, a constitutive relation is required connecting the surfactant concentration with surface tension. Corresponding equations of state for insoluble surfactants were analyzed and evaluated in a recent publication [C42], where it was shown that semi-empirical equations of state have significant advantages over often-used ad-hoc equations of state. It is planned to use the model framework reported in [C42], where the model parameters will be determined by experiments with pendant-drop or Wilhelmy plate tensiometers. Furthermore, a model is needed for the transition of surfactant molecules between different fluid interfaces. The basic structure of the problem is analogous to what is encountered when considering the spreading of a surfactant-covered droplet on a solid surface. In [C23] the transition of surfactants between a liquid-air and a solid-liquid interface was modeled using a first-order kinetic model in the surfactant concentrations. It is planned to use a similar model for the transition of surfactants between the water-gas and the water-oil interfaces. However, as briefly outlined in section 2.5, in a cooperation with Marcello Sega, Forschungszentrum Jülich, the transition of surfactants between the interfaces will also be studied using molecular dynamics simulations, which will assist in identifying and checking suitable kinetic models.

Another aspect that requires suitable models is the flow at the boundary between the lubricant and the "solid", shown as the grey area in Fig. 6. In many cases the "solid" is actually a lubricant-filled porous medium. The corresponding boundary condition can be written as a Navier-slip boundary condition, where the slip-length depends on the permeability of the medium, among others (see [C11] and references therein). The models described under this work package have to be considered as starting points for the implementation in the DROPS code, cf. work packages R3-R5. Constructing numerically consistent models will probably require further iterations in the model formulation. Naturally, this work package will require an especially close cooperation with PI Reusken.

H3: *Wetting on surfaces infused with a single liquid.*
This work package largely addresses the standard scenario of aqueous droplets translating along LISs, as for example considered in [C24]. Based on the setup described under work package H1, experiments will be performed on homogeneous surfaces impregnated with a single lubricant. Lubricants that do (e.g. silicone oil) as well as lubricants that do not show cloaking (e.g. mineral oil) will be considered, but at that stage without adding any true surfactants. Ref. [C40] gives an overview of the cloaking properties of different lubricants. To limit the experiments to a scenario that is dominated by the dynamic deformation of the surface, the focus will be on lubricants with a viscosity significantly higher than that of the droplet. As far as the fabrication of LISs for this and the following work packages is concerned, there are some possibilities in the group of PI Hardt, as described in section 1. However, for

this purpose it is also planned to continue the already existing cooperation with Pavel Levkin (KIT), who offers much more extensive options for fabricating different LISs. The key technique allowing to image the oil distribution around a droplet will be fluorescence excitation using a light sheet, as described under work package H1. While it has already been demonstrated that the lubricant on the surface and in the rim around a droplet can be studied using fluorescence imaging [C43], it is so far unclear if fluorescence imaging of the film cloaking the droplet will be feasible. The main problem lies in the fact that the cloaking film may be so thin that the fluorescence intensity becomes too low to reliably identify the film. Data obtained with cryogenic focused ion beam-scanning electron microscopy [C3] indicate that a cloaking silicon oil film is 65 nm thin. Therefore some optimization is required to tailor the experimental method towards accurate and reliable imaging of the fluorescence within the oil phase. The optimization will include the choice of the laser power and the exposure time as well as the choice of suitable fluorescent dyes. Special attention will be put to measuring the shape of the rim at the front end and the rear end of the droplet. While there appears to be consensus that in the considered situation the rim is responsible for most of the friction forces experienced by a droplet translating along a LIS, the views on the form of the friction law deviate (see [C43] and [C24]). Experiments with varying droplet speeds will be performed, allowing to study the dynamic configuration of the rim as a function of the capillary number. This will allow checking the scaling arguments given in [C24] that are based on a hydrodynamic picture of the rim dynamics referring to Tanner's law. A key parameter to be measured is the friction force on a droplet, evaluated using the known electric forces, as described in work package H1. Also, to study the importance of the three-phase contact line for droplet friction, the results obtained with cloaking lubricants will be compared with those for non-cloaking lubricants. Only for the case of non-cloaking lubricants, an actual three-phase contact line is formed. For this case, the experimental results will be compared to simulation results of PI Reusken, cf. work package R3. As a complementary method, interferometric imaging of the lubricant film profile will be employed in the case of transparent substrates. The corresponding method is not available in the group of PI Hardt, but Detlef Lohse (University of Twente) has agreed to cooperate on that topic.

It is planned to extend the experiments in one direction to study the adaptivity of LISs. For this purpose, mineral oil-infused surfaces will be employed. When a two component droplet sits on such a surface, one component can be extracted to the oil layer, thereby changing its properties. This could be observed with droplets consisting of water and alcohol, the latter being miscible with oil. Dynamic wetting experiments with corresponding droplets will be performed. It is expected that by extraction of alcohol to the oil layer a large asymmetry between the front and the rear end of the droplet will be induced. This phenomenon will also be studied in numerical simulations, cf. work package R4.

The experiments described in this work package aim at answering the following questions, among others:

- How does the oil rim at the front end of the droplet and at the rear end of the droplet deform dynamically?
- Does the rim attain a static configuration for all droplet speeds, or will there be instabilities/oscillations?
- Does cloaking of droplets still happen in the same way when the droplets are moving?
- How does a component extracted from the droplet to the oil layer influence wetting?

In combination with the numerical tools developed by PI Reusken, the experiments should enable un unparalleled understanding of wetting on LISs.

H4: *Dynamic wetting in the presence of surfactant molecules.*
The aim of this work package is to study the wetting of LISs in the presence of true surfactant molecules. Surfactants can have a strong influence on the wetting behavior. For example, they

can strongly enhance the spreading of liquids on surfaces, in effect termed "superspreading" [C23]. Usually surfactant molecules are not very mobile on a solid surface. However, at a fluid interface, fast transport of surfactants may occur by advection and diffusion. This provides the motivation for studying surfactants in context with LIS, presuming that the high mobility of surfactants on the surface wetted by a droplet will give access to novel dynamic regimes of wetting. To limit the complexity of these studies, only insoluble surfactants will be considered, i.e., surfactants that only occupy the fluid interfaces, but are not found in the bulk liquids. Insoluble surfactants usually form monolayers on a liquid surface. Therefore, imaging the surfactant distribution on the surface may appear even more challenging than the imaging of the cloaking oil film. However, the imaging of surfactants spreading on a liquid surface has already been performed successfully (see, e.g. [C17]). Nevertheless, similar to work package H3, the experiments require some optimization of the imaging setup.

The different scenarios considered in this work package are depicted in Fig. 7. Scenarios (a) and (b) refer to situations in which no cloaking is observed. Specific surfactants may also spread on the surface of the lubricant, which is shown in part b. Part c describes a situation in which cloaking is observed. As before, the main focus will be on dynamic wetting. Experiments will be performed in which the speed of the droplet and the viscosity of the lubricant is varied. Using fluorescence imaging the distribution of surfactants along the fluid interfaces will be recorded. Of special interest (in the case of non-cloaking lubricants) are the surfactant distributions close to the three-phase contact line. To obtain information about the coupling between the surfactant concentration and the fluid flow inside the droplet via Marangoni stresses, for each of the surfactants used the relationship between the surfactant concentration and the interfacial tension will be determined using a tensiometer.

Among others, the goal is to answer the following questions:

- How does the presence of surfactants change the friction coefficient of a droplet on a LIS?
- How large is the energy barrier for the cross-over of surfactants between different fluid interfaces, and how does this barrier influence the distribution of surfactants close to the three-phase contact line?
- How fast does a droplet surface become depleted of surfactants if these also spread on the surface of the lubricant?

The combination of the experiments and the simulation results by PI Reusken is expected to allow deep insights into how surfactants influence the dynamic wetting on LISs.

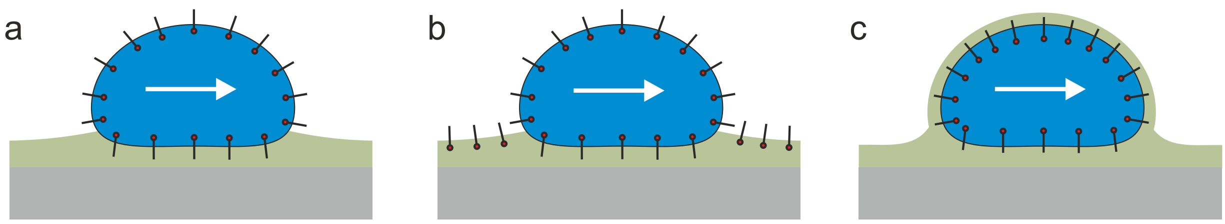


Figure 7: Different dynamic wetting scenarios in the presence of surfactants.

H5: *Wetting on surfaces infused with two different liquids.*
In this work package, the wetting on surfaces exposing two regions impregnated with different lubricants will be studied. A special focus will be put to the effect of cloaking. A basic scenario is depicted in Fig. 8. A combination of two lubricants will be considered in which at least one of the lubricants shows the cloaking effect. Either the setup described under work package H1 will be used to pull a droplet from the more hydrophilic to the less hydrophilic region, or a droplet will be deposited at the boundary between two regions such that it translates to the more hydrophilic region.

A version of this scenario is currently being studied in cooperation with P. Levkin from KIT (see Fig. 2), where it was already shown that cloaking plays a major role for the height of the energy barrier between the two regions. In these preliminary studies it appears as if for the combination of a cloaking and a non-cloaking lubricant, the energy barrier can be described by a simple model where instead of the water surface tension the surface tension of the cloaking liquid is used. However, the situation seems to be more complex at the boundary between two cloaking lubricants. It is planned to experimentally study the transition of droplets between two regions impregnated with different lubricants, varying the driving force on the droplet and the viscosity of the lubricants.

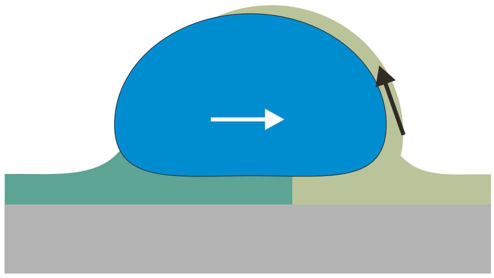


Figure 8: Schematic representation of a droplet crossing the boundary between a non-cloaking and a cloaking liquid.

Among others, the goal is to answer the following questions:

- Are there hysteresis effects when reverting the transition of a droplet from one region to the other?
- Does a droplet transiting from a region with a cloaking lubricant to a non-cloaking lubricant inherit its cloak? If yes, how does the structure of the rim around the droplet where the cloaking liquid, the droplet liquid and the lubricant on the surface come in contact look like?
- Do the transition dynamics and the corresponding energy barrier depend on the speed at which a droplet crosses the boundary?

Work packages PI Reusken

R1: *Mathematical models for fluid dynamics, mass and surfactant transport.*

The numerical simulation of the LIS flow systems is based on several model components. In collaboration with PI Hardt these components have to be adapted to the specific flow system that is considered. The key model components are listed below. We use the following notation. The computational domain is a fixed three-dimensional rectangular box containing the three phases oil (lubricant), water (droplet) and air. The three phases have different material properties ρ_i (density) and μ_i (dynamic viscosity), $i = 1, 2, 3$. These densities and viscosities are assumed to be constant in each phase. The domain Ω is partitioned into three subdomains $\Omega_i(t)$ (cf. Fig. 9), each of them containing one of the phases. These phases are separated from each other by the interfaces $\Gamma_{1,2}(t)$ (water-air), $\Gamma_{1,3}(t)$ (water-oil), $\Gamma_{2,3}(t)$ (air-oil). For convenience, below we drop the t-dependence in the notation. The boundary of the droplet is denoted by $\Gamma_d = \Gamma_{1,2} \cup \Gamma_{1,3}$ and the union off all three interfaces by $\Gamma = \Gamma_{1,2} \cup \Gamma_{1,3} \cup \Gamma_{2,3}$. $\mathbf{n}_\Gamma$ denotes the unit normal on Γ pointing from Ω_1 into $\Omega_2 \cup \Omega_3$ and from Ω_3 into Ω_2; often we write $\mathbf{n}$ instead of $\mathbf{n}_\Gamma$. We introduce the normal velocity $V_\Gamma = V_\Gamma(x,t) \in \mathbb{R}$ which denotes the magnitude of the velocity of the interface Γ at $x \in \Gamma(t)$ in normal direction. For $x \in \Gamma$ we define the projection $\mathbf{P}(x) = I - \mathbf{n}(x)\mathbf{n}(x)^T$. The operator $\nabla_\Gamma = \mathbf{P}\nabla$ is the tangential gradient.

Fluid dynamics: incompressible Navier-Stokes and coupling conditions.

Based on basic conservation laws of mass and momentum the following standard model (in strong formulation) for the fluid dynamics of a three-phase incompressible flow can be derived:

$$\begin{cases} \rho_i(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u}\cdot\nabla)\mathbf{u}) & = \operatorname{div}\boldsymbol{\sigma}_i + \rho_i \mathbf{g} \\ \operatorname{div}\mathbf{u} & = 0 \end{cases} \quad \text{in } \Omega_i, \quad i = 1,2,3,$$

$$\begin{aligned} [\boldsymbol{\sigma}\mathbf{n}_\Gamma]_\Gamma &= \operatorname{div}_\Gamma \boldsymbol{\sigma}_\Gamma \quad \text{on } \Gamma, \\ [\mathbf{u}]_\Gamma &= 0 \quad \text{on } \Gamma, \\ V_\Gamma &= \mathbf{u}\cdot\mathbf{n}_\Gamma \quad \text{on } \Gamma. \end{aligned} \tag{1}$$

with the stress tensor $\boldsymbol{\sigma}_i = -p\mathbf{I} + \mu_i(\nabla\mathbf{u} + (\nabla\mathbf{u})^T)$, i.e. we consider Newtonian bulk fluids and an interface stress tensor $\boldsymbol{\sigma}_\Gamma$, which models surface tension forces. A standard ansatz for this interface stress tensor is $\boldsymbol{\sigma}_\Gamma = \tau\mathbf{P}$, with a surface tension coefficient τ. The right-hand side in the interface force balance in (1) then takes the form

$$\operatorname{div}_\Gamma \boldsymbol{\sigma}_\Gamma = \operatorname{div}_\Gamma(\tau\mathbf{P}) = \tau \operatorname{div}_\Gamma \mathbf{P} + \nabla_\Gamma \tau = -\tau\kappa\mathbf{n} + \nabla_\Gamma\tau, \tag{2}$$

with κ the mean curvature of Γ. The vector $\mathbf{g}$ denotes an external (gravity) force. The operator $\operatorname{div}_\Gamma$ denotes the tangential divergence. For modeling the liquid-liquid-gas MCL we use an approach based on (curvature dependent) restoring forces, cf. work package H2 above. Finally we complement the model with suitable boundary conditions. On the lower boundary (between lubricant and "solid") we use a Navier boundary condition (cf. work package H2).

In the literature on the numerical simulation of such sharp interface multiphase models, these Navier-Stokes equations and coupling conditions are reformulated in a *one-fluid model* [A4], [C50], cf. also work package R2 below. Such an approach, which will also be applied in this project, is based on a (computable) interface representation, which is used to reformulate the model outlined above as *one* Navier-Stokes system with discontinuous density and viscosity coefficients and with localized (surface tension) forces at the interface Γ and a localized effective restoring force at the moving contact line. Thus both in the modeling and numerical simulation we use a *sharp interface* approach.

Mass transport: convection-diffusion equation and coupling condition.
We consider the case that the droplet contains a dissolved species that is transported to the oil film due to convection and molecular diffusion and does not adhere to the interface. The concentration of this species is denoted by $c(x,t)$. This flow problem can be described by the above Navier-Stokes model coupled with a convection-diffusion equation for the concentration c:

$$\begin{aligned} &\frac{\partial c}{\partial t} + \mathbf{u}\cdot\nabla c = \operatorname{div}(D_i\nabla c) \quad \text{in } \Omega_i, \quad i = 1,3, \\ &[D_i\nabla c\cdot\mathbf{n}]_\Gamma = 0 \quad \text{on } \Gamma_{1,3}, \\ &\qquad c_1 = C_H c_3 \quad \text{on } \Gamma_{1,3}. \end{aligned} \tag{3}$$

The interface condition $c_1 = C_H c_3$ (with $c_i = c_{|\Omega_i}$) is the so-called Henry condition, which causes a discontiuity in the concentration at the interface. Clearly the convection is determined by the velocity field $\mathbf{u}$. Often there is also a coupling back, i.e., a dependence of the Navier-Stokes equations on the concentration c, for example through Marangoni forces induced by $\tau = \tau(c)$. For this reverse coupling we will study different scenarios, with increasing level of numerical complexity, cf. work package R4 below.

Surfactant transport: surface convection-diffusion equation and coupling condition
We consider an insoluble surfactant which adheres to the interface Γ and is transported at this interface due to convection and diffusion, cf. scenarios (a)-(b) in Fig. 7. The concentration of this surfactant

is denoted by $S(x,t)$, $x \in \Gamma(t)$. From mass conservation and the constitutive law $q = -D_\Gamma \nabla_\Gamma S$ for the diffusive flux q (Fick's law), one obtains the following model for transport of surfactants on the droplet boundary and on the lubricant-air interface:

$$\frac{\partial S}{\partial t} + \mathbf{u} \cdot \nabla S + S \,\mathrm{div}_\Gamma \mathbf{u} = D_\Gamma \Delta_\Gamma S \quad \text{on } \Gamma_{i,j},\ (i,j) \in \{(1,2),(1,3),(2,3)\}. \tag{4}$$

In scenario (a), Fig. 7, we consider surfactants only on the closed droplet surface, modeled by the convection diffusion equation (4) on Γ_d. Clearly, for scenario (b) one needs a coupling of the three surfactant transport equations on $\Gamma_{i,j}$. For this we consider kinetic models as briefly addressed in work package H2 above. Such kinetic models lead to two coupled convection-diffusion equations of the form as in (4) (on Γ_d and on $\Gamma_{2,3}$) with an additional source term that couples the two equations via a flux balance. For larger surfactant concentrations significant Marangoni forces are expected, which means that one has to model the dependence of the surface tension coefficient on S, i.e., $\tau = \tau(S)$. In this project we address different scenarios with increasing level of numerical complexity, cf. work package R5 below.

These principal model components, which have already been used in the group of PI Reusken for the numerical simulation of a rising butanol droplet with mass- and surfactant transport, have to be adapted to the specific LIS flow problem considered. In particular the modeling of the contact line forces and the coupling conditions needed in mass and surfactant transport have to be developed in collaboration with the group of PI Hardt, cf. work package H2 above.

R2: *Hybrid interface representation method for LISs.*

In this three-phase system we need a numerically feasible representation of the interface Γ. In the DROPS code a level set technique is implemented and applied to several *two*-phase flow systems. This approach has to be extended to be able to deal with evolving interfaces between *three* phases. One possibility is to introduce an additional level set function [C44, C53]. In this project we develop another approach which makes use of the special property that the free surface of the oil film Γ_l is only moderately deformed and can be represented as a height function on the underlying fixed wall. The rectangular solid wall domain is denoted by $W \subset \mathbb{R}^2$. For the interface representation we propose the following approach (I1)-(I3):

(I1) The droplet boundary Γ_d is *implicitly* captured by a level set function $\phi(x,t)$, $x \in \Omega$, i.e., Γ_d is the zero level line of ϕ. For ϕ we have the usual transport equation $\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = 0$.

(I2) The lubricant surface Γ_l is *explicitly* represented by a height function $H : W \to (0,\infty)$. The interface condition $V_\Gamma = \mathbf{u} \cdot \mathbf{n}_\Gamma$ determines the evolution of $H(x,t)$. Different transport equations for H, resulting in the same geometry evolution, are possible, e.g., the nonlinear transport equation $\frac{\partial H}{\partial t} - \frac{1}{1+\|\nabla H\|^2} \mathbf{u} \cdot \begin{pmatrix} -\nabla H \\ 1 \end{pmatrix} = 0$ (in this case there is no tangential movement of points on Γ_l).

(I3) After steps (I1), (I2) one has two approximations (one implicit and one explicit) of the water-oil interface $\Gamma_{1,3}$, which in general do not coincide. Based on the available approximations a unique reconstruction of $\Gamma_{1,3}$ is determined.

The above procedure is (strongly) coupled with the Navier-Stokes equations for the evolution of the velocity field $\mathbf{u}$. As far as we know, such a treatment of interfaces in a three-phase flow problem has not been studied in the literature. The numerical components for the realization of (I1), comprising a level set reinitialization procedure and interface reconstruction techniques, are already available in DROPS. In this project we will develop methods for (I2), (I3), based on finite element techniques.

Clearly, there is a difficulty concerning the resolution of the very thin lubricant film. If we scale such that the droplet has diameter 1, the film thickness (cf. Fig. 9) is expected to be of size $\varepsilon \sim 10^{-3}$ or smaller (based on preliminary experiments). As is common in level set based finite element discretization methods for multiphase flows, we use *un*fitted grids, i.e., the grids do not align to the (evolving) interface. For the resolution of the very thin oil film we choose an a-priori fixed subdomain $W \times [0, \varepsilon]$ and construct a strongly anisotropic grid, which has aspect ratios of order ε^{-1} (Fig. 9). In the remaining part of the computational domain we use a shape regular tetrahedral grid, which is locally refined close to the droplet surface Γ_d. In DROPS there are methods implemented for the efficient construction of a hierarchy of (locally refined) grids. On such a grid we use quadratic finite elements for the spatial discretization of the level set function (already available in DROPS).

It is expected that the strong anisotropy does not cause a deterioration of the approximation quality of the finite element discretization because the triangulation used still satisfies the maximum angle condition. The strong anisotropy may cause difficulties for the iterative methods used for solving the resulting discrete system. In the research group of PI Reusken similar issues related to anisotropy were addressed in [C39, C45].

For the spatial discretization of the height function H we can use standard finite element spaces on a shape regular triangulation of the wall domain W.

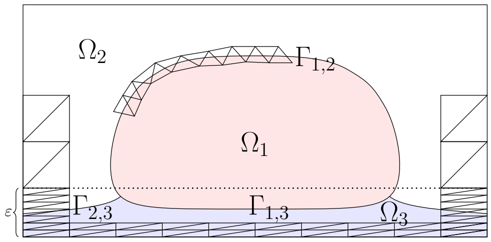


Figure 9: Interfaces and triangulations. $\Gamma_d := \Gamma_{1,2} \cup \Gamma_{1,3}$ (droplet surface); $\Gamma_l := \Gamma_{1,3} \cup \Gamma_{2,3}$ (lubricant surface).

R3: *Fluid dynamics simulation of droplets on LISs.*

We consider a flow system as in Fig. 7(a), but *without* surfactants, modeled by the Navier-Stokes equations (1) and the MCL treatment as outlined above. This flow system is also studied in work package H3. The aim is to develop an efficient and reliable solver for this three-phase flow problem. For the handling of the interface we use the method discussed in work package R2. For the discretization of the one-fluid Navier-Stokes equations major issues are the following. We have to discretize the (curvature dependent) surface tension forces on Γ, the discontinuities in density and viscosity must be treated appropriately and we need suitable finite element spaces for velocity and pressure approximation. All these issues also occur in two-phase flow simulations ([A4], [C50]) and we expect that the extension of the already available (two-phase) solver components in DROPS to the three-phase case is fairly straightforward. For the treatment of the MCL we have to discretize (curvature dependent) force terms localized at the moving contact line. For this we will adapt the techniques already developed in the two-phase MCL flow problems treated in [A7].

R4: *Fluid dynamics simulation of droplets on LIS with mass transport.*

A component dissolved in the droplet can be extracted to the oil film, thereby modifying its properties, cf. work package H3. Such a system is modeled by coupling the Navier-Stokes equations (1) with the mass transport equations (3). In the first funding period we only consider a passive component that does not change the properties of the water and lubricant phases (i.e., a one-way coupling only). This assumption can be weakened in the following funding period, cf. section 2.3.5. In addition to the fluid dynamics simulation in work package R3 we have to discretize the convection-diffusion equation (3) and treat the Henry interface condition. For the latter we use the well-established Nitsche method studied in e.g., [C28, C29]. Depending on the material parameters and the flow dynamics, sharp concentration boundary layers at the interfaces may occur. In a first step, with PI Hardt we will agree on a system with not too sharp boundary layers at the water-oil interface. We expect that based on the already available (two-phase) mass transport components in DROPS [C28], the extension of the fluid dynamics solver developed in work package R3 to this case with mass transport across the water-oil interface does not cause major difficulties. Once a reliable solver for this flow problem has been developed, it can be extended to more difficult cases, e.g. with (very) thin boundary layers, or a back coupling of the mass transport to the fluid dynamics (e.g., Marangoni forces). The study of these more difficult scenarios is planned for the second funding period, cf. section 2.3.5.

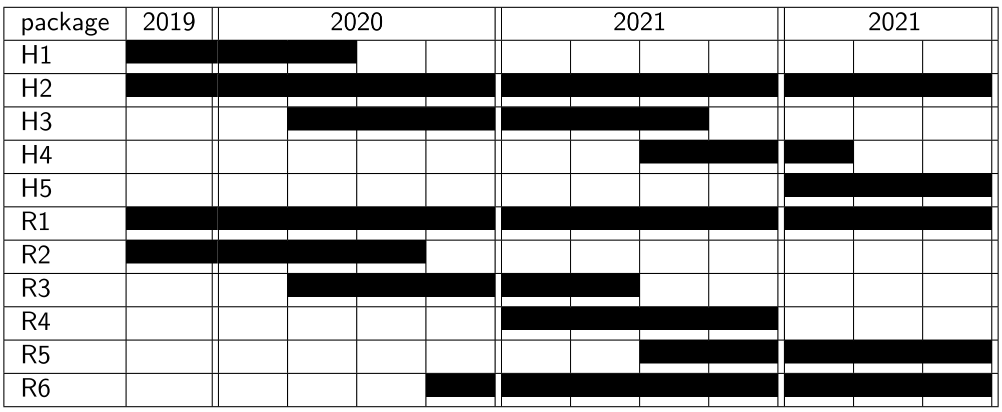
R5: *Fluid dynamics simulation of droplets on LISs with surfactant transport.*
We aim at the numerical simulation of scenario (a) in Fig. 7. For this we have to couple the fluid dynamics based on model (1) with the surfactant transport (4) on the droplet surface Γ_d. Methods for treating the convection-diffusion equation on Γ_d are already available in DROPS [C55], [A6]. In the first funding period we will extend these methods to the specific flow problem considered here and we will also treat a coupling back to the Navier-Stokes equation through Marangoni effects. For the latter we need a model $\tau = \tau(S)$, which will be developed in collaboration with PI Hardt, cf. work package H2. Based on this work, more complex scenarios such as the one in Fig. 7(b) will be addressed in the second funding period.

R6: *Creation of a tutorial on DROPS simulation of LISs* .
Based on the already available DROPS user manual a document will be created in which the application of DROPS for the simulation of specific flow problems as described in work packages R3–R5 is explained. This tutorial should facilitate the use of the DROPS solver by others, for example SPP 2171 members interested in numerical simulations of such flow problems.

Collaboration between PI Hardt and PI Reusken

In the first funding period, the cooperation will address various aspects of dynamic wetting on LIS. The modeling and simulation of the spreading of sub-100 nm lubricant films on droplets (cloaking) poses special challenges and will be addressed in the second funding period. The formulation of suitable mathematical models for the wetting on LIS is a main cooperation topic, cf. work packages H2 and R1. The PIs will jointly formulate numerically consistent models for the phenomena described above. The main modeling components (1), (3), (4) have to be complemented with models for the MCL and for Marangoni stresses $\tau = \tau(S)$. Furthermore, PI Hardt will provide characteristic material parameters, initial conditions and boundary conditions for the simulations. Specific experiments will be designed and performed such that the results can be used to validate the modeling and simulation framework, cf. work packages H3 and H4. Last but not least, after the simulations have reached a certain level of maturity, they will assist in the physical understanding of wetting experiments on LISs, in particular related to the questions formulated in the work packages H3 and H4, and in the planning of new experiments.

Schedule



Long term objectives

Concerning the experimental part (PI Hardt), in the second funding period the studies of droplets wetting surfaces infused with two different lubricants will be extended. In that context, two interesting limiting cases are droplets significantly larger or smaller than the typical domain size (impregnated with a specific lubricant) of the surface. Furthermore, a focus will be on adaptive surfaces that change their properties when a component dissolved in a droplet is extracted into the lubricant. It will be studied how this process influences dynamic wetting and also the interaction between different droplets. Last but not least, the photoswitching of surfactants will be studied as a tool for droplet manipulation. Concerning the numerical simulation, the solver components described in work packages R4, R5 will be extended such that Marangoni stresses in the mass transfer, a dependence of density and viscosity on the solute concentration, and surfactant transfer between droplet surface and lubrication surface can be treated. This again requires strong collaboration between PI Hardt and PI Reusken concerning model development. One further modeling and simulation aspect that we plan to address in the second funding period concerns the cloaking phenomenon. Due to the very thin cloaking film there arises a scale separation problem. We plan to develop an approach based on coupling a thin-film equation for the spreading lubricant film with the Navier-Stokes equation in the bulk. A similar strategy of coupling the thin-film equation to the Navier-Stokes equation has already been employed in [A5]. Furthermore the modeling will be extended to different lubricants, as shown in Fig. 8. The model descriptions for specific phenomena will be refined, especially that of the cross-over of surfactants between different interfaces. Overall the level of maturity of the simulation framework will be increased, such that the simulations qualify as a reliable research tool, allowing to better understand the physics of wetting. The devopment of tailor-made numerical methods will advance the state of the art in the field of numerical simulation of multiphase liquid-liquid-gas flows with MCLs and surfactants.

Data handling

Those data underlying the articles to be published will be archived according to the specifications given by the DFG. Otherwise, no special measures for data handling and management appear to be necessary.

Information on scientific cooperation within SPP 2171

Jeanette Hussong, Evgeny Gurevich (U. Bochum): PIV measurements of the flow inside liquid-infused filament arrays using confocal fluorescence microscopy by PI Hardt and Astigmatism Particle Tracking Velocimetry (A-PTV) measurements to characterize the internal drop circulation of droplets on LISs by PIs Hussong/Gurevich.

Gregory Lecrivain (Helmholtz-Zentrum Dresden-Rossendorf): Measurement of the local deformation of the thin oil film near the three-phase contact line by PI Hardt for validation of PI Lecrivain's numerical simulations.

Pavel Levkin (KIT): Fabrication of special liquid-infused surfaces by PI Levkin and sharing experimental data on wetting on liquid-infused surfaces by PI Hardt.

Marcello Sega (Forschungszentrum Jülich): MD simulations of surfactants crossing over between different fluid interfaces by PI Sega as input for the models developed by PIs Hardt/Reusken.

Uwe Thiele (U. Münster): Simulation of the surfactant distribution on droplets on LISs and the corresponding droplet shape, based on the long-wavelength approximation, by PI Thiele and corresponding measurements by PI Hardt.

Axel Voigt (TU Dresden) and Sebastian Aland (HTW Dresden): Definition and numerical study of a (soft) wetting benchmark problem with different substrate rheologies. Simulation is performed with different numerical techniques: ALE (Aland), phase-field method (Voigt) and a level-set method (Reusken).

Florian Kummer (TU Darmstadt): Regular exchange on numerical issues of extended Galerkin formulations, e.g., the variational formulation of contact line models, preconditioning of linear solvers and general stabilization techniques.
Holger Stark (TU Berlin): Methodological exchange on a level set based finite-element method (Reusken) and a boundary element technique (Stark), both applied to a Stokes free surface model with dynamic wetting.
Dirk Peschka (WIAS Berlin): Comparison of numerical schemes for (Navier)-Stokes flow with free boundary and moving contact lines. In particular an ALE approach (Peschka) will be compared to a level set based unfitted finite element method (Reusken).

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Requested modules/funds

Basic module

Funding for staff

Research staff

PI Hardt: One postdoctoral researcher. The task of the postdoctoral researcher will be to cover the work packages H1 to H5. This involves experimental as well as theoretical work, which poses a significant challenge concerning the background of suitable candidates. For this reason, a postdoctoral researcher is preferred over a PhD student. Suitable candidates for the remaining work packages have a background in mechanical engineering, chemical engineering or physics and extensive knowledge in fluid mechanics and wetting. A salary scale of 100 % E13 TV–L is applied for, and funding is needed for the complete duration of the project.

PI Reusken: One 75 % PhD position (E13 TV–L). This researcher will work on the topics described in packages R1 to R6. The candidate should have a background in (numerical) mathematics, computational science and engineering or a related field.

Miscellaneous staff

PI Hardt: One student assistant.

It would be beneficial to have a student assistant for the first two years of the funding period to help with the experimental setup and to do preperative experiments such as the identification of suitable fluorescent dyes for the lubricants and of fluorescent surfactants. Therefore, funding for a student assistant (35 hours per month, 11.75 EUR/hour) is applied for over a period of two years. Total: 9,870 EUR.

PI Reusken: One student assistant.

It would be beneficial to have a student assistant (for the three year funding period) who supports the PhD student with C++ code development and code testing. Furthermore the student assistant helps with the creation of the SPP 2171 specific DROPS tutorial (work package R6). Funding for a student assistant (20 hours per month, 11.75 EUR/hour) is applied for over a period of three years. Total: 8,460 EUR.

Direct project costs

Equipment up to Euro 10,000, Software and Consumables

PI Hardt:

Equipment up to 10,000 EUR and small parts

The following table lists the components necessary to set up the imaging system. Only those components are listed that are either not yet available in the group of PI Hardt or that are too frequently used to make them available for the proposed project.

Component	Purpose	Price[EUR]
	Light sheet creation	
Objective (Mitutoyo 10x, NA 0.28, 33.5 mm WD)	Projection of light sheet onto droplet	$1,050$
4-axis stepper motor (Picard Industries)	Positioning of objective relative to droplet	$5,400$
Different laser diodes (each approx. 200 EUR)	Fluorescence excitation, specific types of diodes only available after experimental assessment	$1,000$
Lenses (Thorlabs)		$1,000$
Optomechanical adapters & lens mounts (Thorlabs)		$1,000$
Large-beam fiber collimator (Thorlabs)	Mechanical decoupling of light-sheet objective from other components	$1,300$
Fiber port and fiber patch cable (Thorlabs)		800
Small parts		$1,000$
	Imaging	
Long-distance microscope (Navitar 12x UltraZoom)	Imaging of fluorescence intensity distribution	$3,900$
Band-pass filters	Selective imaging of specific dyes, need to be compatible with emission wavelength	600
		Σ $17,050$

Consumables

1. Fluorescent dyes and fluorescent surfactants.

As already described in section 2.3.1, it is a priori not clear which fluorescent dyes and surfactants are most suitable for the light-sheet imaging experiments. Therefore, optimization studies need to be performed. Fluorescent dyes and fluorescent surfactants are quite expensive chemicals for which at the current stage only a lump sum can be specified. Σ 3,000 EUR.

2. Lubricants & polymers

Different lubricants such as silicone oil or Krytox as well as polymers (for the porous matrix) are needed for fabricating the LISs. Again, a lump sum is specified. Σ 2,000 EUR.

Total PI Hardt: Σ 22,050 EUR. **Total PI Reusken**: 0

Travel Expenses

PI Hardt:

Travel funds are applied for mutual visits between the groups of the PIs, to support conference visits and for attending SPP events. In that context, one visit of PI Hardt and the researcher funded by the project to the group of PI Reusken for a period of two days scheduled. The estimated costs of this visit are 600 EUR. Concerning the conference visits, one visit per year is scheduled. Suitable conferences for presenting the project results would be the "American Physical Society, Division of Fluid Mechanics" conference series and the "flow" conference series (flow19, flow21 etc.). For one conference visit a cost of 2,500 EUR (incl. the registration fee) is estimated. For allowing the researcher and the PI to attend the SPP events during the whole funding period of three years, costs of 3,800 EUR are allocated.

Visit to the group of PI Reusken Σ 600 EUR

Conference visits Σ 7,500 EUR

SPP events: Σ 3,800 EUR

Total PI Hardt Σ 11,900 EUR

PI Reusken:

Travel funds are applied for mutual visits between the groups of the PIs, to support conference visits and for attending SPP events. Two visits of PI Reusken and the researcher funded by the project

to the group of PI Hardt for a period of two days scheduled. The estimated costs of one visit are 600 EUR. One conference visit for the researcher funded by the project per year is scheduled. For one conference visit a cost of 1,200 EUR (incl. the registration fee) is estimated. For allowing the researcher and the PI to attend the SPP events during the whole funding period of three years, costs of 3,800 EUR are allocated.
Visits to the group of PI Hardt Σ 1,200 EUR
Conference visits Σ 3,600 EUR
SPP events: Σ 3,800 EUR
Total PI Reusken Σ 8,600 EUR

Project requirements

Employment status information

Hardt, Steffen, Full Professor at TU Darmstadt; unlimited contract.
Reusken, Arnold, Full Professor at RWTH Aachen University; unlimited contract.

Composition of the project group

PI Hardt. Apart from the researcher and PI Hardt, Dr. Tobias Baier and Mr. Jörg Bültemann will be involved in the project. Dr. Baier works as a Research Associate (unlimited contract) and is paid by funds provided by the university (Landesmittel). His area of specialization is theoretical microfluidics, and he will assist in the model development described under work package H2. Mr. Bültemann works as a technician (unlimited contract) and is also paid by funds provided by the university (Landesmittel). He will support the project by services including fabrication of parts and assembly of components of the experimental setup.
PI Reusken. Apart from the researcher and PI Reusken, Dr. Sven Gross and Mr. Hauke Sass will be involved in the project. Dr. Gross is the main coordinator of the in house developed DROPS code and has been involved in most of the applications of this code. He works as a Research Associate (unlimited contract) and is paid by funds provided by the university (Landesmittel). Mr. Sass is on a PhD position financed by the university (Landesmittel) since 2017 and is expected to finish his PhD research project in 2021. His research focuses on the development and analysis of space-time finite element discretization methods for surface PDEs. He will support the project by making available DROPS components for the simulation of surfactant transport (package R5).

Cooperation with other researchers

Detlef Lohse (University of Twente): Interferometric imaging of the lubricant height profile for the case of transparent substrates in the group of D. Lohse.
Maxim Olshanskii (University of Houston): involved in the development and analysis of numerical methods for surface PDEs.

Researchers with whom you have collaborated scientifically within the past three years

PI Hardt:
Günter Auernhammer (Leibniz Institute of Polymer Research, Dresden), Aditya Bandopadhyay (IIT Kharagpur, India), Dominik Barz (Queen's University, Canada), Moran Bercovici (Technion-Israel Institute of Technology), Hans-Jürgen Butt (Max-Planck Institute for Polymer Research, Mainz), Suman Chakraborty (IIT Kharagpur, India), Ulrich Göringer (TU Darmstadt), Christian Holm (University of Stuttgart), Axel Klar (TU Kaiserslautern), Heinz Koeppl (TU Darmstadt), Pavel Levkin (Karlsruhe Institute of Technology), Cunjing Lu (Tsinghua University, China), Holger Marschall (TU Darmstadt), Ulrike Nuber (TU Darmstadt), David Quéré (ESPCI, France), Jens Smiatek (University of Stuttgart), Howard Stone (Princeton University, USA), Subramanyan Namboodiri Varanakkottu (NIT Calicut,

India).

PI Reusken:
Dieter Bothe (TU Darmstadt), Ralf Krause (University of Lugano), Christoph Lehrenfeld (Uni Göttingen), Holger Marschall (TU Darmstadt), Wolfgang Marquardt (FZ Jülich), Adel Mhamdi (RWTH), Maxim Olshanskii (University of Houston, US), Annalisa Quaini (University of Houston, US), Michael Schlüter (TU Hamburg-Harburg), Martin Wörner (KIT), Vladimir Yushutin (University of Houston, US).

Scientific equipment

As mentioned in section 1, many of the components needed for characterizing droplets on LISs by light-sheet imaging are already available. The fluorescence signals will be collected by an ultra-sensitive CCD camera. One of the three Andor Technology cameras available in the group of PI Hardt will be used for that purpose. Apart from that, the most important scientific infrastructure for the proposed project is available in the context of the Collaborative Research Center 1194: "Interaction between transport and wetting processes". PI Hardt is involved in two subprojects of the CRC 1194, among others a project in which a generic experimental configuration for the characterization of drops on surfaces has been developed that is available to all PIs of the CRC. This generic configuration allows for an extensive characterization of static and dynamic wetting. It permits high-speed imaging in brightfield mode both from the side and through the substrate from below (for transparent substrates) as well as fast confocal fluorescence imaging from below in a humidified chamber. For the latter, a Nikon-Eclipse Ti microscope equipped with a Yokogawa CSU-X1 Spinning Disk Unit and a Andor Technology iXon Ultra DU-897U-BV CCD camera is available. Last but not least, the infrastructure of the CRC offers extensive options for measuring surface tensions, interfacial tensions and contact angles with different instruments.