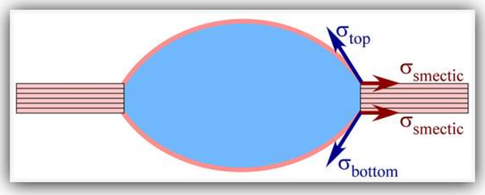
Project description

Wetting, elastic and capillary forces of thin freely suspended smectic films

(New proposal within SPP 2171)



General information

Applicants

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Topic

FREELY SUSPENDED FILM INTERFACES

Keywords

experimental physics, wetting, surface/interface tensions, polarizing microscopy, high-speed imaging

Summary

In this project, the dynamics of fluid interfaces during wetting and capillary interactions will be studied experimentally. We will take advantage of a distinctive property of smectic liquid crystals: their ability to form stable freely suspended films. These films represent quasi-2D liquids, with unique features. Their layered structure guarantees uniform film thicknesses in equilibrium, on a molecular scale. It also renders them extremely robust and inhibits drainage. At the same time, these films are extremely flexible and they exhibit complex shape dynamics, providing unique model systems for studies of thin fluid membranes interacting with impacting or embedded objects, or exposed to other static or dynamic stresses. Such films allow the investigation of wetting and dewetting in combination with related elastic deflections of the film surfaces.
The proposal is based on preliminary studies by the applicants of droplet impact on smectic films, interface tension dynamics and optical manipulation of interfaces, substantially expanding these studies and broadening the scope of the investigations. The research program is divided into three interrelated subtasks: impact of liquid droplets on thin free-standing films, embedding and motion of objects in the films, and studies of the dynamic interface tension including optical manipulation of wetting behaviour:

(i) Droplet impact on free-standing films: We propose to study the impact of droplets of immiscible liquids on thin freely suspended smectic films. At low kinetic energy, the droplets become trapped in the film plane. When they are sufficiently fast, they can penetrate the films. Thereby, they become encapsulated by a thin smectic coating, forming smectic 'shells'.

(ii) Droplets on smectic films: Impacting droplets that are trapped by the film first settle on its surface. Their subsequent shape transformations reflect wetting processes at the droplet top and bottom surfaces, within microseconds. We will employ high-speed imaging to analyze this wetting process for a representative selection of liquids and surfactants.

(iii) Dynamical interface tension: When a fresh smectic interface is created in a low-concentration surfactant solution (below the critical micelle concentration), it is initially covered only partially with surfactant molecules. The dynamics of the surface coverage results in a time dependent interface tension. In addition, photosensitive surfactants can be used to locally or globally manipulate the interface tension. This allows to control shapes, positions and stability of LC structures immersed in aqueous solutions by irradiation with UV and visible light, continuously and reversibly.

1 State of the art and preliminary work of the applicants

State of the art

Thin fluid membranes are of fundamental importance in many respects. Biological membranes are prerequisites of the evolution of complex living structures. Thin films forming foams are extremely important not only in biological systems as well as in a widespread variety of technical applications, from cleaning, food processing and construction industry to insulation and firefighting. Because of the huge surface to volume ratios of such systems, capillary forces play an important role in structural characteristics and dynamic processes.
Smectic liquid crystal phases have a distinctive property: Like soap solutions, they form stable freely suspended films (the term free-standing smectic films will be used here synonymously). Such films can be prepared with remarkably high aspect ratios (above one million to one) and they can serve as straightforward models of two-dimensional (2D) fluids. Depending on the symmetry of the respective smectic phase, they can be isotropic or anisotropic in the film plane. With photosensitive surfactants, one can optically switch transitions between those states. It has been demonstrated that subtle changes of interfacial tensions of such films can have substantial macroscopic consequences (cf. Fig. **??**). Capillary interactions can lead to the trapping of objects that are orders of magnitude thicker than the films. Shapes of embedded droplets are a direct measure of film tensions. Capillary interactions as well as elastic forces between trapped objects can be mediated by the smectic films.

Freely suspended smectic films: Freely suspended smectic films were first investigated about 40 years ago [1], and have been in the focus of liquid crystal (LC) research since then. Experimental and theoretical studies were devoted to the understanding of in-plane flow processes, texture dynamics, electro-optic switching, spontaneous pattern formation, phase transitions in 2D and many other topics. A comprehensive description of research on such films is found in the textbook by Oswald and Pieranski [2]. Apart from the structure and dynamics of the films themselves, an interesting aspect is the mobility of inclusions in such films [3–5], which is qualitatively different from mobilities in 3D fluids. Inclusions trapped in such films can move in the film plane and interact with other inclusions, with gradients of the film thickness or boundaries (menisci). In anisotropic films (e. g. smectic C), they can even interact with the local c-director orientation. A review of interactions and pattern formation of inclusions in freely suspended films can be found in Ref. [6].
In the context of the present proposal, such films with their huge surface to volume ratios will serve as soft surfaces, where kinetic energies compete with capillary forces and elastic deformations. An example of elasto-capillary interactions in nematics was reported by Liu et al. [7], and similar phenomena have been described for freely suspended smectic films [8]. The applicants of this proposal have studied structures and dynamics of free-standing smectic films for more than two decades (see below).

Droplet impact, bouncing on fluid surfaces: Droplet impact and splashing are phenomena that have been studied for more than a century, starting with pioneering work of Worthington [9] in 1895. Since then, droplet impact on solid surfaces, solid surfaces covered with thin fluid films and bulk liquids was studied intensely, and remains an exciting topic for scientific research [10]. A particularly interesting phenomenon is bouncing of droplets on vibrated liquid surfaces [11, 12]. Droplets bounce off a surface even of the same fluid when the rebound is faster than the time necessary to remove the thin air layer between droplet and fluid surface [13]. Impact scenarios are essentially governed by capillary forces, kinetic energies and oscillation dynamics of the droplets, and the dynamics of the thin air film entrapped between droplet and surface during impact. Previous investigations with thin films as targets were performed on soap films and oily ink films (see [14–16] and Refs. in [17]). If the droplets have sufficient kinetic energy, they can tunnel through the films which at the same time remain intact. Studies of droplet impact on freely suspended smectic films performed by the PIs are described in the Preliminary Work subsection.

Dynamics of the smectic interface tension: Smectic and nematic interfaces in surfactant solutions play an important role, e. g., for the stability of microshells [18], foams or similar structures. Smectic shells are stable under homeotropic anchoring conditions. They usually become unstable when the anchoring is planar. Homeotropic anchoring can be induced, e. g., by surfactants like sodium dodecyl sulfonate (SDS),

or sodium dodecyl benzene sulfonate (SDBS). Anchoring transitions triggered by surfactants are useful in chemical or biological sensors [19].
While surface tensions of LC phases with respect to air have been studied extensively, reports of interface tensions of smectics in aqueous environments are rather scarce. Kim et al. [20] measured the interface tension of the nematic 5CB using the pendant droplet method and obtained values of 1.5 mN/m or 7 mN/m, depending on which data for the LC density were used. Gharbi et al. [21] measured the interface tension of nematic 5CB to a mixture of water, $CaCl_2$ and polyvinyl alcohol. They found a value of 5.6 mN/m for the planar anchoring conditions in this environment.
The problem becomes even more complex when the surfactant concentration is below the critical micelle concentration (cmc). The adsorption of surfactants to fluid-fluid interfaces of isotropic fluids has been a permanent focus of research for nearly 80 years, first addressed in the 1940's [22]. The tension of an interface is determined by its coverage with surfactant molecules. When the solution is not saturated and not in equilibrium with the interface, the surface coverage is incomplete. Below the cmc, the surface coverage remains incomplete even in equilibrium, the interface tension increases with decreasing surfactant concentration. Interfacial tension and area coverage are interrelated by adsorption isotherms (Langmuir and Frumkin isotherms). Direct measurements of the area coverage have been achieved, e.g. by ellipsometry, infrared spectroscopy and neutron reflectrometry. Agreement of the heuristic models with the few available data is often not satisfactory (e. g. [23]). In previous research, we have developed novel interface tension measurement methods specifically for smectic materials, and we have reported for the first time quantitative data for the dynamic surface tension of smectics in aqueous surfactant solutions (see next subsection).
The idea to control surface characteristics of fluids by irradiation with UV or VIS light is nowadays widespread. For example, azobenzene chromophores can be incorporated into polymers to reversibly change their physical or chemical surface properties [24], molecular based devices can be conceived [25], or particle ensembles can be altered reversibly or irreversibly [26]. Among other properties, the wettability can be controlled [27]. Particularly suitable systems for photoswitching are LCs, where the director structure can promote long-range effects of surface switching. This is possible not only at solid substrates, but also at free surfaces [28]. Substantial progress in the field of LC dynamic interface tensions has been achieved by the PIs (see Preliminary Work section).

Interactions of director distortions and surface energy: The surface energies of anisotropic mesophases can be of comparable magnitude as elastic distortion energies. Then, the shapes of fluid inclusions may be determined by the competition of capillary forces and bulk elastic deformations of the director field. So-called tactoids reflect this competition [29]. Similarly, topological defects of the director field in the vicinity of the surface of nematics or smectics can generate dimples or other deformations of the interface. Surface undulations related to director defect structures in menisci of freely suspended smectic films have been detected [30,31] by a combination of AFM and polarizing microscopy. It should be emphasized that menisci of smectic films at the border of the support frames or embedded objects have structures that are much more complex that those of ordinary liquids [2].

Preliminary work of the applicants

The applicants have acquired expertise with smectic freely suspended films for more than two decades. Surface phenomena, dynamics and polar properties of such films were extensively investigated by both PIs over those years and published in various international journals. In the following, we focus only on preliminary work that is directly relevant to the intended research plan.

Shooting of picoliter droplets onto freely suspended films [17,32,33]: We performed the first experiments where freely suspended smectic films were targeted with liquid droplets in order to study the droplet impact. Droplets with diameters of about 30 μm to 50 μm were prepared using a commercial dispenser system. The drop impact was recorded with a high speed camera. Three scenarios could be identified, depending upon the impact velocity. When this velocity is low, the drops of immiscible liquids are integrated into the smectic film (Fig. 1). We observed the establishment of a stationary drop shape within a few hundred microseconds. From contact angles, we can conclude on the wetting of the drop with smectic material, which takes place within about 300 μs. Thereafter, the formation of a meniscus of smectic film material over a period of several seconds is observed.

a) b)

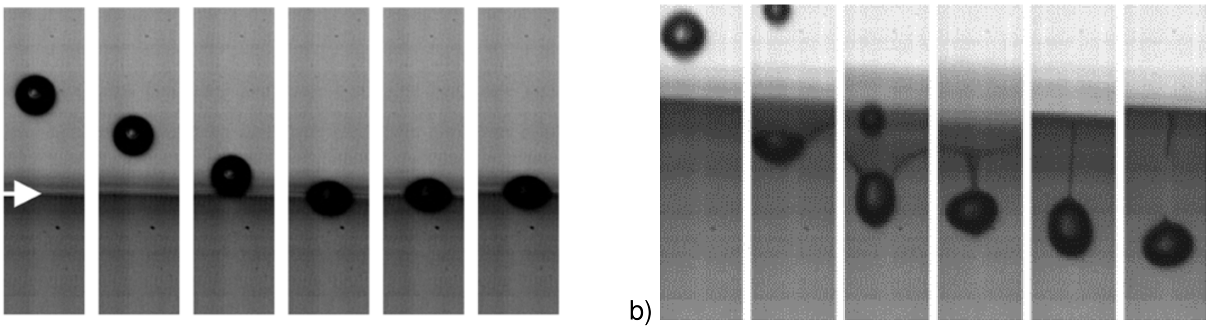

Figure 1: a) Side view of a droplet impact (water with 5% ethylene glycol) on a smectic freely suspended film [17]. The initial film position is indicated by a white arrow: impact velocity 0.8 m/s, frame rate 18 000 fps, image sizes are 346 μm $\times 93$ μm, droplet diameter ≈ 42 μm. b) droplet tunneling the film, impact velocity 5.3 m/s, frame rate 60 000 fps.

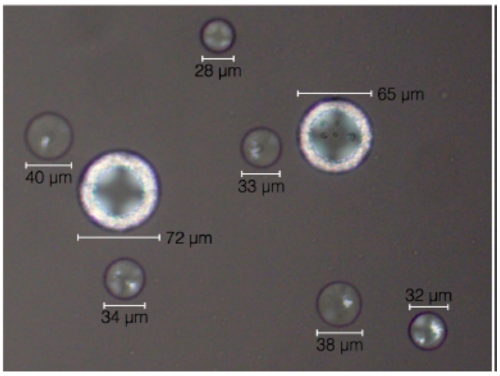

Figure 2: Smectic shells caught in a water bath [17]. The picture shows the shells under crossed polarizers. They consist of a thin ($< 1\mu$m) spherical smectic film and are filled with an ethylene glycol / water mixture. Two of the shells settled at the bottom plate of the water tank, they have the shapes of flat lenses. The others are intact and spherical.

Drops that hit the films at intermediate velocities can bounce back by a trampoline effect, in which the vibration of the impinging drop plays an essential role. At high energies, the droplets penetrate the smectic film without destroying it. They are thereby surrounded by a smectic shell. This provides a unique opportunity to produce smectic shells with diameters less than 50 μm. With a conventional microfluidic apparatus, minimum shell sizes that can be achieved are around 200 μm [34].

Motion of solid and liquid inclusions on smectic films [4, 33, 35, 36]: Using a micromanipulator, we can place small particles on freely suspended smectic films to study microrheology of the films. When the film plane is intentionally inclined to the horizontal, the particle motion under the influence of a variable effective gravitation can be studied. At the same time, we determine the mobility of the particles from the observation of their diffusion in horizontal films. There are four possible geometrical combinations:
(1) The motion of isotropic solid spherical particles in films of the smectic A phase (2D-isotropic liquid) has been described in Ref. [4]. The rheology of liquid microdroplets is much more complicated, such inclusions often have neither constant form nor constant volume during the experiments. Nevertheless, a few such experiments were successfully performed [33].
(2) The motion of droplets in the smectic C phase (a 2D-anisotropic liquid) was studied in tilted films under the influence of an effective gravity. The main focus was on the description of the mutual influences of the director field and the motion of inclusions. It was found that the orientation of the director has little influence on the effective viscosity [33].
(3) Motion of anisotropic particles, glass rods with dimensions in the micrometer range in films of the in-plane isotropic smectic A phase was described in a Master thesis [36] and in Ref. [35]. By investigating the diffusion of such rods, earlier theoretical predictions by Levine et al. could be verified. The mobility of the

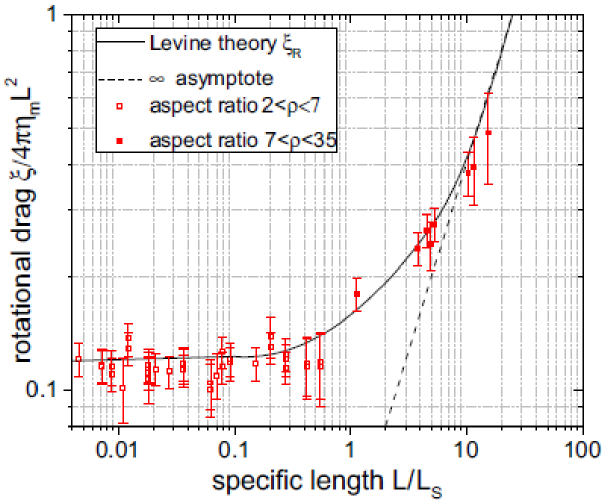

Figure 3: Dependence of diffusion properties on the ratio of rod length L and Saffman length L_s, the Levine theory (*Phys. Rev. Lett.*, **93** 038102 (2004)) is represented by a solid line.

rods is, to a good approximation, isotropic when the Saffman length (the product of the film thickness with the ratio of viscosities of the smectic film and ambient air) is greater than the rod length. If the rod length exceeds the Saffman length (in sufficiently thin films or with rods of about 1 mm length), the mobility, and accordingly the diffusion, becomes anisotropic (Fig. 3).

(4) The combination of anisotropic inclusions with 2D-anisotropic films (smectic C) has been explored in preliminary experiments, and further efforts are necessary to elucidate this complex issue.

Surface and interface tensions of smectic LCs [37–42]: The measurement of the surface tension, and in particular its temperature trend, is of paramount importance for the understanding of capillary interactions, Marangoni flow and geometrical structures as droplets or tactoids. We have developed several methods addressing the measurement of this quantity that are highly competitive and often superior to alternative approaches in LCs [37–41]. Of particular interest is the analysis of the shapes of film-embedded droplets [39], which allows to extract details of the smectic surface tension in the vicinity of the clearing point and in presence of surface-wetting smectic layers on an isotropic bulk phase of the same material.

Another challenge is the measurement of the interface tension of smectics respective to surrounding surfactant solutions. The classical pendant droplet shape methods fail because of the very small density differences of smectics and water. The technique developed by us [41] allows to determine, with unmatched precision, this interface tension as a function of the surfactant concentration not only in the asymptotic equilibrium state (Fig. 4), but also dynamically during the establishment of this equilibrium.

Menisci of smectic films [31,43–46]: Menisci of smectic freely suspended films differ substantially from such of ordinary, unstructured liquids. The equilibrium profile often develops only in time scales of minutes [2]. The menisci can be decorated with complex director patterns, both conservative (energy minima) [30, 31, 43–45], and dynamic patterns [46]. In previous work [31, 43, 45], we demonstrated that classical models developed for the description of the so-called 'splay domains' require fundamental revision. We have shown that the symmetry of the mesophase is correlated with the appearance of meniscus patterns, while a nematic phase is not required for their appearance (as it was presumed in Loudet's model [30]). Additionally, we were able to measure the profiles of smectic C menisci by means of AFM [31]. We have shown that defects of the c-director are correlated with periodic modulations of the film surface. These structures obviously represent compromises of elastic and surface energy minimization.

Shape dynamics of smectic films [47–50]: Smectic freely suspended films show a particularly complex shape dynamics. Owing to the layered internal structure, they possess elastic properties that are essentially different from, e. g. soap films. At short time scales, they rather behave like vesicles [48] We

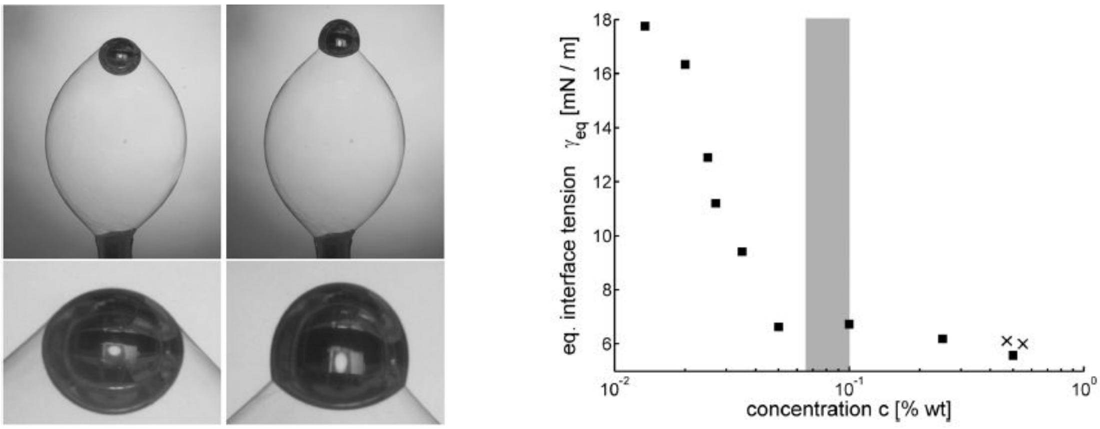

Figure 4: Left: Smectic bubble with a trapped air bubble in a surfactant solution below the critical micelle concentration (*cmc*). Buoyancy pushes the entrapped air up, the smectic film holds it fixed at the contact line. Initially, this line is above the equator of the air bubble (stable configuration). With lowering dynamic interface tensions it shifts downwards at the air bubble periphery. The position of the entrapped air bubble becomes unstable. Image size 13 mm $\times$ 15,2 mm (top), the images below show enlarged details. Right: Asymptotically established interface tension as a function of the surfactant concentration. The gray range marks the approximate cmc range. [40]

analyzed the shape transformations of distorted bubbles on their way towards the spherical equilibrium shape [47, 49], and identified a unique wrinkling effect of smectic films under lateral compression [50].

Reversible switching of surface anchoring between isotropic and anisotropic [51, 52]: Recently we demonstrated that a photoresponsive dendrimer dissolved in a nematic LC can spontaneously adsorb at its interface to form a commanding surface. Under visual light (VIS) illumination, the dendrimer supports homeotropic director anchoring, and thus an in-plane isotropic interface. UV illumination switches the alignment back to planar (anisotropic surface) When solid or liquid inclusions are embedded in an LC matrix that is doped with specific photoreactive dendrimers, the anchoring of the director can be switched reversibly. This transition can be used to trigger reorientations or relocations of solid inclusions [51], and shape transformations of liquid inclusions. It has been demonstrated that this transition can be continuously controlled by the ratio of UV and VIS irradiation intensities [52].

1.1 List of project-related publications

Articles published in peer-reviewed international journals

1. C. Klopp, R. Stannarius, A. Eremin. Role of shape anisotropy in diffusion dynamics in quasi-2D fluids. *Phys. Rev. Fluids*, **2** 124202 (2017).

2. H. Nádasi, et al., Photomanipulation of the anchoring strength using spontaneously adsorbed layer of azo dendrimers, *Phys. Chem. Chem. Phys.* **19** 7597 (2017).

3. R. Stannarius, K. Harth. Inclusions in freely suspended smectic films. In S. Lagerwall and G. Scalia, editors, *Liquid Crystals with Nano and Microparticles*, volume 1. World Scientific, (2016).

4. A. Eremin et al., Optically driven translational and rotational motions of micro-rod particles in a nematic liquid crystal. *PNAS* **112** 1716 (2015).

5. S. Dölle, R. Stannarius, Microdroplets impinging on freely suspended smectic films: three impact regimes, *Langmuir* **31** 6479 (2015).

6. S. Dölle et al., Impact and embedding of picoliter droplets into freely suspended smectic films, *Langmuir* **30** 12712 (2014).

7. K. Harth et al., Dynamic interface tension of a smectic liquid crystal in anionic surfactant solutions, *Phys. Chem. Chem. Phys.* **17** 26198 (2015).

8. K. Harth, R. Stannarius, Measurement of the interface tension of smectic membranes in water, *Phys. Chem. Chem. Phys.* **15** 7204 (2013).

9. A. Eremin et al., Two-dimensional microrheology of freely suspended liquid crystal films. *Phys. Rev. Lett.*, **107** 268301 (2011).

10. K. Harth,B. Schulz, C. Bahr, and R. Stannarius, Atomic force microscopy of menisci of free-standing smectic films, *Soft Matter* **7** 7103 (2011).

2 Objectives and work programme

2.1 Anticipated total duration of the project

The project's intended duration is 6 years, funding is needed for the complete period. The application period is 36 months.

2.2 Objectives

Wetting of LC surfaces by other liquids as well as wetting of solid surfaces by LCs are particularly complex for several reasons. The anisotropy of the mesophase can evoke an anisotropy of the interface, and even the geometry of the surface can be influenced by the orientation in the bulk. On the other hand, surface properties can be employed to control the bulk material over large distances via the anchoring of the director field. Near phase transitions, the surface of a nematic bulk phase can be wetted by a thin smectic layer, and isotropic bulk material can be wetted by LC layers (see e. g. [53, 54]).
Apart from this, particularly interesting geometrical structures are freely suspended smectic films. Those films, with nanometer or micrometer thicknesses, represent quasi-2D liquids with huge surface to volume ratios, and with unique dynamical and elastic properties. Smectic films are flexible and can be excited to vibration by external electric fields or mechanical stimuli. They can be shaped as bubbles, catenoids, or shells, and also easily transferred onto solid or liquid substrates. The molecular layer structure does not only render them extremely robust, and inert to drainage processes, it also provides uniform film thicknesses in equilibrium, even on a molecular scale.
We plan to investigate the dynamic aspects of the interactions between the flexible interfaces of smectic free-standing films, with thicknesses on a molecular scale, and immiscible liquids. This includes in particular the study of impacts of liquid droplets or solid particles, where wetting is strongly related to film deflections and droplet shape transformations. Wetting also affects the mobility of *droplets* embedded in the film plane, which is much more complex that the motion of solid microparticles [33]. An important aspect will be the opportunity to control relative time scales of elastic deformations and wetting processes.
The present proposal is based upon experience gained during previous studies on droplet impact, inclusion dynamics and film shape dynamics. The work in the application period is divided into three mutually related subtasks: impact of (liquid) objects on thin free-standing films, embedding and motion of such objects in the films, and experiments related to the dynamics of the interface tension of films immersed in aqueous solutions:

Work package (I): Droplet impact on thin smectic films
The aim of this task is the study of the impact of droplets in a micrometer to millimeter size range on freely suspended films. Droplets with low kinetic energy are known to be trapped by the film, while droplets with sufficiently high energy can penetrate the films. As a result, they become encapsulated in a thin smectic coating. We plan to systematically investigate the tunneling of droplets in a wide range of sizes and materials through smectic films, considering also the option to tunnel multiple films to generate multishell objects. We will both work with liquids that wet the smectic films and such that do not wet the smectic film material.
The encapsulation technique can be of some importance both from an academic point of view and for practical applications, since smectic shells with diameters between $\approx 20\ \mu$m and few millimeters may be prepared. This opens up opportunities to produce very small shells that cannot be prepared with contemporary microfluidic devices. They allow the study of smectic structures with large layer curvatures, e. g. interactions and self-organization of topological defects on curved surfaces. On the other hand, large droplets with smectic coating can be produced as analogues of giant vesicles. The study will substantially expand the range of droplet materials from the low concentration ethylene glycol solution in water used in preliminary experiments to aqueous and non-aqueous droplets with and without surfactants.
In previous work, we investigated picoliter droplets produced by piezoelectric dispensers, as commonly used in technical applications such as inkjet printers. With droplets up to millimeter sizes, we can control the relative time scales of wetting processes (which are practically independent of droplet sizes) and of elastic deformations, which crucially depend on droplet sizes. Thus, it is possible to study scenarios where wetting is slow compared to droplet and film oscillations, and such where shape transformations adiabatically couple to wetting processes.

Work package (II): Droplets on smectic free-standing films
Droplets of low kinetic energy are trapped by the film plane. They first settle on the film surface. An interesting detail is the question whether a thin air layer persists between film and droplet after impact, and how this layer disintegrates afterwards. A technique to explore this is total internal reflection (see below).
In previous experiments, it was shown that picoliter-size pure water droplets that are not wetted by the smectic material evaporate within milliseconds, while addition of ethylene glycol in low concentration inhibited evaporation and produced droplets that were wetted by smectic layers. Thereby, the droplets adopted new equilibrium shapes:
The spherical droplets change their shape to double sphere caps in response to capillary forces. Initially, the bottom droplet surface is coated by the smectic film, the upper surface is still bare, and the droplet is asymmetrically inserted in the film. It is flattened by lateral capillary drag of the film. After some delay, several hundred microseconds in picoliter droplets, the top surface is also covered by a smectic layer, and the droplet immediately adopts a new, symmetric shape. We aim to understand details of these wetting processes. The basic steps have been studied by us so far only for one specific combination of smectic material, droplet material and droplet size [32]. Work by Stebe and collaborators has shown that with other combinations, different geometries can be realized [7, 8], symmetrically and asymmetrically embedded objects, which may have attractive or repulsive elasto-capillary interactions in the film plane. We plan to analyze these phenomena quantitatively for different droplet materials, including pure water and water with different surfactants favouring planar or homeotropic anchoring, and for droplets in a broad size range. This allows to vary the relative strengths of gravitational, kinetic, elastic and surface energies during the impact (Ohnesorg, Weber, Bond numbers). While the wetting dynamics is relatively uninfluenced by droplet sizes, we can scale the elastic deformation dynamics of droplets and films by orders of magnitude, varying droplet sizes. This allows a comprehensive comparison of both processes individually and in mutual competition.
The droplets can move in oblique or vertical films under the action of gravity. Such studies were performed earlier with picoliter droplets and with small solid inclusions [4, 33, 35]. These objects were embedded in the films symmetrically to the central plane, and the films remained flat because of the low weights of these inclusions. Using much larger droplets, the motion of inclusions will evoke elastic deformations of the film depending on the embedding symmetry, the weight of the droplets and the wetting character. We expect to observe different advancing and receding contact angles and menisci.
Droplets in our previous experiments were wetted/encapsuled by the smectic material. We expect qualitatively different scenarios when using droplets are not wetted by the smectic material.
We have further the option to move droplets with optical tweezers available in our lab. This has the advantage that the drag can be reversed and one can perform multiple experiments with an individual droplet. The disadvantage is that the drag forces are not as reliably controlled as effective gravitation in tilted films.

Work package (III): Dynamical surface tension
When a fresh smectic surface is created in an aqueous surfactant solution with low surfactant concentration (below the critical micelle concentration, cmc), it is only partially covered with surfactant molecules. The dynamics of the coverage of the surface determines the time-dependence of the interface tension. We have studied this phenomenon [40] using a buoyancy method specifically developed for smectic interface tensions [41]. For the determination of the smectic interface tension in aqueous solutions, our method is much more reliable and more accurate than droplet shape measurements, primarily because the latter suffer from the almost identical mass densities of the smectic materials and water.
A novel experiment proposed here is to create a smectic film in the sub-cmc surfactant solution and to equilibrate this film, so that the low asymptotic value of the interface tension is reached, then to create more surface of the same film by expanding the support frame. The consequence of this will be Marangoni flow in the system, and shape distortions of non-planar films (spherical or Delauney shapes). The interface tensions of freshly formed and equilibrated films can differ by factors of up to two, so the effect will certainly be non-negligible. One can conceive situations where this Marangoni effect may lead to a dynamical stabilization of smectic film configurations, and others where this effect destabilizes certain configurations. Figure 4 evidences such macroscopic shape changes after minor variations of interface tension ratios.
Using specific photoresponsive dendrimers which spontaneously adsorb at LC-water or LC-glass interfaces, we can prepare commanding surfaces. Combining UV and visible light, one can not only switch between states with different interface properties, one can also continuously control anchoring strengths [52]. This technique can be combined with any of the three subtopics to such tasks as
(1) study of nematic pre-wetting transitions depending on anchoring strengths, and contact line dynamics,

(2) measure contact angles and alignment using Confocal Laser Scanning Microscopy (CLSM),
(3) control the shapes of droplets in smectic films and their dynamics,
(4) optically trigger the detachment of droplets from freely suspended films, and
(5) optically trigger Marangoni effects in thin films using patterned illumination.

2.3 Work programme including proposed research methods

Work package (I): Droplet impact on thin smectic films
The existing experimental setup for studying the droplet impact will be modified to accommodate for experiments with droplets of a broad size range, up to about one millimeter. It requires a set of dispensers to produce droplets in this size range, including a selection of suitable commercial dispensers and the development of custom-made devices. Droplets may be prepared using piezo-techniques or with vibrating nozzles.
The chamber for preparing the smectic films will be supplied with a temperature controller and an interferometric unit to record the film thickness. All films will first be characterized with routine techniques to determine surface tensions and 2D viscosities. The following experiments are planned:

- WP (Ia) Varying the droplet parameters, such as size and velocity, we will investigate the impact dynamics on films with variable thicknesses and in different phases (smectic A, smectic C). The conditions for re-bouncing, tunneling and trapping regimes will be determined. Of particular interest is the separation of the time scales of wetting and geometrical changes of droplets and films.
- WP (Ib) Tunneling through multiple films such as double (e. g. smectic/water), if possible even triple (e. g. smectic/water/smectic) membranes will be attempted and recorded by high-speed imaging. This may provide an original approach to create multi-shell structures. If this approach is successful, it should be continued in a separate project, outside the SPP 2171.
- WP (Ic) The shells are subsequently collected in a liquid bath, where we will record and evaluate the impact of shells and multi-shells on liquid surfaces. The aim is to understand how such encapsuled droplets spread after bursting on the bath surface, or how they are deformed when they enter the bath. This will depend crucially upon the composition of the involved fluids.
- WP (Id) The interfacial dynamics between the droplet and the film are essential for the rebound regime. Waveguiding/total reflection will be employed to determine if and when a direct contact between a droplet and the film occurs: Laser light is fed laterally into the film, and decoupling of the light will be detected by high-speed imaging. In that experiment, we will dope the materials with fluorescent dyes. This experiment shall be performed in collaboration with Kirsten Harth, who's project within the SPP is specialized to the detection of contact lines and entrapped air layers.

WPs (Ib,c) will be restricted to the demonstration of the feasibility of the techniques. A systematic investigation of multi-shell structures must be left for a potential continuation of this project, or be performed in a separate project.

The work in WP (I) will be started in the first year of the proposal. We estimate a time period of 1-2 months for the PhD student performing these research tasks to become acquainted with the topic and the state of the art, including a literature survey, and another 2 months to built up the setup, test the dispensers, and learn to prepare smectic free standing films. For the experiments, we expect a time period of 8 months, and 2-3 months for data evaluation and preparation of the results for publication, together with the PIs.
Within this subtopic, we plan collaboration with Gregory Lecrivain (Rossendorf), who will simulate impact on thin films by numerical methods. Existing numerical programs of impact of particles on elastic surfaces will be modified appropriately. We also plan to collaborate with Günter Auernhammer (Dresden), who has valuable experiences with droplet impacts on soap films [15]. Kirsten Harth (Twente) plans to investigate droplet impact with highspeed camera techniques, focusing on soft surfaces. We plan to utilize her equipment and experience in the analysis of contact lines that is part of her project in the SPP.

Work package (II): Droplets on free-standing films
This work package will be performed partially in a time line parallel with WP (I), since the same materials and in part the same observation techniques will be employed. The processes from deposition of a droplet

on a smectic substrate to its full embedding involve multiple interactions on different time scales. This requires a combination of various measuring techniques. The essential features for the investigations are

- WP (IIa) Analysis of details of shape changes of droplets during impact (employing long-range microscopy): The capillary forces of the films suffice in normal gravity to hold droplets of radii of the order of a millimeter. We will employ high-speed imaging in order to record the transformation of individual contact angles at the droplet top and bottom surfaces. These differ initially when the upper droplet surface is bare, but the lower one is wetted by the smectic film. The evolution of the contact angles and cap shapes will allow us to retrieve the wetting dynamics.
- WP (IIb) Dynamics of the contact line after the impact (studied using an inverted microscope in the confocal mode): Using Confocal Fluorescent Polarizing Microscopy, we can investigate the contact region between the isotropic fluid and the smectic substrate. The coupling between the flow, mechanical film deformation and liquid crystal director will affect the birefringence and the dichroism in fluorescence-labeled LCs. These features enable us to visualize the surface-layer dynamics.
- WP (IIc) Droplets sliding on films under the action of an effective gravitational drag: Wetting also influences the motion of droplets on inclined smectic films. We record the sliding motion of droplets in oblique films and measure advancing and receding contact angles. Interface tension and the viscosity of the film can be tuned by a selection of the smectic material. Sliding in anisotropic films, such as oriented smectic-C, will lead to coupling of the contact line dynamics and the director. A novel aspect is the study of large droplets which are not embedded symmetrically but deform the films locally. If the droplet material does not wet the films, a rolling motion of large droplets may take place instead of sliding.

The experimental techniques have been described in preliminary studies of the applicants, the essential novel aspect is the systematic variation of inclusion sizes and materials. For this task, the existing setup has to be modified. About 1-2 months will be needed for the modifications, including the installation of the film (and microscope) tilting mechanics, and the modification of the observation technique (top and side view cameras). This task will be essentially performed by the project technician under supervision of the PhD student and a PI. For the measurements, 6-7 months will be necessary. Then, 2-3 months are required for data evaluation and publication of the results.

For the modeling of the dynamics of the wetting behavior of settling and sliding droplets, we will collaborate with Martin Oettel (Tübingen, DFT) and Svetlana Gurevich (Münster).

Work package (III): Dynamical surface tension

The dynamics of the surface coverage of surfactants determines the time-dependent interface tension of smectic structures in aqueous solutions. Controlling the viscosity of the embedding fluid even allows us to tune this dynamics to the respective time scales of the available observation techniques. The proposed experiments aim to determine the time dependence of the dynamical surface tension and to investigate instabilities accompanying the interface tension gradients and the surface equilibration process. Smectic bubbles will be prepared in an aqueous environment with a surfactant well below the CMC. The dynamics of the surface tension will be determined using an improved air-bubble buoyancy method [40, 41].

A rapid extension or reduction of the bubble surface from the equilibrium state results in a non-equilibrium coverage of the film by surfactant molecules. The exchange dynamics of these molecules with the solution is reflected in the relaxation of the dynamic surface tension to equilibrium. A visualization of surfactant distributions may be achieved by using a fluorescence-labeled surfactant.

- WP (IIIa) So far, the air-buoyancy method was used only to determine interface tensions of the smectic film and the surfactant solution, but it also allows to determine the so far disregarded other interface tensions (air-surfactant solution, smectic-air) from the contact line geometry. The surface coverage of the air interface of the solvent is accessible, and a direct comparison of the dynamics with that of the smectic interface coverage is possible. No new experiments are needed, experimental data are available from earlier measurements.
- WP (IIIb) An initially *inhomogeneous* distribution of the surfactant on the film surface gives rise to a gradient of the surface tension which can drive Marangoni flow in the film. This flow can be visualized

by tracer particles or by mask bleaching of fluorescence-labeled smectic films. As described above, we prepare smectic films in sub-cmc aqueous solutions, then expand the film area and observe Marangoni effects arising from the locally inhomogeneous surfactant load on the film. It equilibrates only after several minutes, during this time, we expect flow and shape distortions of the smectic material.

- WP (IIIc): The anchoring energies can be optically manipulated using azo surfactants compatible with the LC mesogens. In previous studies, we investigated effects of anchoring transitions on the self-organisation of LC-dispersed microparticles. Here, we intend to modify the interface tension of the smectic films. Local or global modifications will trigger macroscopic shape changes of the immersed smectic structures, including contact line dynamics and topological changes.

  A novel aspect is light-driven Marangoni flow. The idea is to prepare smectic films doped with azo-dyes serving as surfactants and decorating the smectic-water interface. Local UV illumination results in a local modification of the interface tension and may trigger flow. The flow field can be visualized using either tracer particles or smectic islands (surplus smectic layers). Since similar experiments with bulk liquids are planned within the SPP 2171, we can provide expertise obtained in previous studies of photoswitchable nematic-solid interfaces to the other groups in the SPP.

WP (III) will start in the second half of the application period. For WP (IIIb,c), we will basically develop the techniques and demonstrate the phenomena in experiments. A systematic study of these effects will not be realistic within this period, it is left for continuation in the following application period.
For the installation and test of the setup, we estimate a necessary workload of 2-3 months. This work will be performed by the technician under supervision of a PI. The dynamic tension measurements require about 4-5 months. Further 5-6 months will be needed for the study of Marangoni effects. Finally, 3 months will be required for preparation of publications by the PhD student and the PIs.

Collaboration is planned with Kirsten Harth (Twente), who was involved in the development of the buoyancy method for the measurement of interface tensions. Further, we plan to collaborate with Günter Auernhammer (Dresden) on the subject of dynamic surface tensions. Suitable photoswitchable surfactants will be synthesized in collaboration with Bert Jan Ravoo (Münster). Our experimental data will provide a solid quantitative experimental data basis for numerical and analytical modeling. For modeling of the time dependence of the surfactant layer formation, we will collaborate with Martin Oettel (Tübingen, DFT) and Svetlana Gurevich (Münster). Topic (III) is related to research by Anita Roth-Nebelsick and collaborators (Dresden) on microbubbles in water transport conduits of plants, where we plan to provide observation techniques and expertise in microrheology, data analysis and modeling. Holger Stark (Berlin) deals with modeling of photo-switchable substrates. In the course of the ongoing work in the SPP, we will explore to apply his numerical simulations with boundary element methods (BEM) to our specific systems. For the project of Svetlana Santer (Potsdam), we will provide theoretical support in the field of dynamically switchable surfaces particularly for nematic films.

Data processing, theory and modeling
The data evaluation and analysis will be performed using the available imaging and computational software. Our group developed a solid background for data processing of the proposed experiments. Theoretical modeling will be performed throughout the complete project. Critical analysis of the experiments and the models will provide the material for scientific publications. Numerical simulations of the flows and droplet deformations will be made using Finite Elements Method implemented in the available *Comsol* software package. Collaboration with the theoreticians within the *Schwerpunktprogramm 2171* (Gregory Lecrivain, Svetlana Gurevich, Martin Oettel, Holger Stark, Uwe Thiele) is vital.

The two applicants of this project, R. S. and A. E., will advice the PhD student, supervise the design of the experimental setups and the experimental work. Both project leaders will be involved in each of the three subtasks, where R. S. will particularly take responsibility for the experiments and data evaluation of work packages (I) and (III), A. E. will particularly take responsibility for package (II).

2.4 Data handling

Experimental data will be stored in a repository for inspection and usage in further projects for an appropriate period.

2.5 Other information

not applicable

2.6 Explanations on the proposed investigations

Experiments involving humans, human materials or animals: not applicable

2.7 Information on scientific and financial involvement of international cooperation partners

not applicable

2.8 Information on scientific cooperation within SPP 2171

- Gregory Lecrivain (Rossendorf): numerical simulation of the membrane and droplet dynamics during droplet impact, and comparison to experiments in subtopic (I)
- Günter Auernhammer (Dresden), experiments with droplet impact on freely suspended films (I) and studies of dynamic surface tensions (III)
- Kirsten Harth (Twente): experiments to determine dynamic surface tensions and study of Marangoni flow in smectic films submerged in sub-cmc surfactant solutions (III)
- Svetlana Gurevich (Münster): theoretical modeling of the dynamics of surfactant coverage on smectic films in aqueous solutions (III)
- Martin Oettel (Tübingen): microscopic modeling of wetting dynamics for droplets placed on freely suspended films dynamics of surfactant layers on smectic films in aqueous solutions with Dynamic Functional Theories (II,III)
- Holger Stark (TU Berlin): modeling of the dynamics of surfaces, in particular photo-switchable surfaces, by Boundary Element Methods (III)
- Bert Jan Ravoo (Münster): synthesis of photoswitchable surfactants (II,III)
- Anita Roth-Nebelsick (Dresden): microfluidic analysis of the dynamics of air bubbles in biological channels (III).

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4 Requested modules/funds

4.1 Basis module

4.1.1 Funding for Staff

A 75 % TV L 13 position for 36 months is requested for a PhD student performing the experimental research. The PhD student will also be involved in the supervision of Bachelor and Master theses related to the project. For research assistants (students) carrying out routine measurements of smectic material parameters needed for the evaluation of experimental results, we request 400 Euros per year.

4.1.2 Direct Project Costs

4.1.2.1 Equipment up to 10.000 €, Software and Consumables

The commercial dispenser is needed for the preparation of microdroplets. The control unit plus three orifice sizes are requested.
A Laser is required to particle detection, shape measurements and contact detection in the impact measurements. Two temperature controllers are needed for the assembly of custom made heat stages for the confocal microscope and for the film holders in the droplet impact and rheology experiments.

Offer (2): Dispenser incl. controller	Horizon Instr., 27.09.2018	9,822 € (incl. VAT)
Offer (3: Solid state Laser	Laser2000, 21.09.2018	7,693 € (incl. VAT)
Offer (4): Temperature controller	Thorlabs , 21.09.2018	per unit 634 € (incl. VAT)

On the basis of these offers, a total of 18,800 Euros is requested for equipment.

The following consumables will be necessary to maintain the routine experiments and operation of the scientific equipment.

Item	**Costs**
Chemicals (per year)	500 €
Microscope bulbs (per year)	300 €
Electronic parts (sensors, controllers, detectors, per year)	1,500 €
Optical parts (lenses, prisms, mirrors)	7,000 €
Motorized positioning elements, breadboard, optical holders	9,000 €

A total of **41,700 Euros** is required for equipment (< 10 k Euros) and consumables.

4.1.2.2 Travel Expenses

We request funding for the participation in national and international conferences such as the International Soft Matter Conference, Droplets 2019, Jülich Soft Matter Days, European Nonlinear Dynamics Days, for presentation of the results. Travel expenses are requested for one international conference per year. We estimate a demand of 2,500 Euros for one non-European conference and 2,000 Euros each for two European conferences for one attendee, respectively.
Funding for one participant at one national meeting per year (e. g. DPG spring meeting) is requested (3 $\times$ 400 Euros). For planned SPP events, we request funding for three people (two PIs and one PhD student) attending two workshops in the first and second year (6 $\times$ 500 Euros), one person (PhD student) attending an advanced school and a PhD candidate workshop (2 $\times$ 600 Euros). For 3 researchers attending an international conference organized by the SPP, we request 1,800 Euros.
500 Euros per year are requested for visits to cooperating partners within the SPP 2171 (2-3 visits for 1-2 days each per year).
We apply for funding of total travel expenses of **15,200 Euros**.

4.1.2.3 - 4.1.2.5
not applicable

4.1.2.6 Publication costs

For contributions towards publication costs for submissions to *Open access*-Journals (most likely *New Journal of Physics*), we request **750 Euros** per year.

4.1.3 Instrumentation

4.1.3.1 Equipment exceeding 10,000 €

A high power light source is requested for light-demanding high-speed imaging acquisitions with the available fast cameras. The main requirements for the light source is high intensity in a narrow spectral range, and the ability to switch between different spectral bands which is needed for real-time interferometric film-thickness measurements.

High-Power Light Source for High-Speed Imaging
offer (1) by Laser2000 of 30.11.2017, amount (incl. VAT) **13,229 €**

5 Project requirements

5.1 Employment status information

The applicants are employed on permanent positions at OvGU Magdeburg. The engineer I.-U. Grodrian, who will be involved in the project, has a permanent position in the department of the project leaders.

5.2 First-time proposal data

no

5.3 Composition of the project group

Project leaders:	Prof. Dr. Ralf Stannarius, apl. Prof. Dr. Alexey Eremin
Project scientist	N.N.
Technician	Dipl.-Ing. Ines-Ute Grodrian
Bachelor-/Master student	N. N.

5.4 Cooperation with other researchers

5.4.1 Researchers with whom you have agreed to cooperate on this project

Günter Auernhammer (Dresden), Svetlana Gurevich (Münster), Kirsten Harth (Twente), Gregory Lecrivain (Rossendorf), Martin Oettel (Tübingen), Bert Jan Ravoo (Münster), Anita Roth-Nebelsick (Dresden), Holger Stark (TU Berlin), in alphabetic order. The details of these collaborations are specified in Section 2.8-

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

Previous collaborations existed with numerous research groups during the past three years, in the form of common projects, exchange of students and researchers, and common publications. Among those groups are synthetic chemists in Halle (Carsten Tschierske, Wolfgang Weissflog) and Mainz (Rudolf Zentel), providing mesogenic materials, physicists of HP Labs and the University of Bristol (Susanne Klein, R. M. Richardson) preparing and characterizing pigment suspensions, physicists of the University of Colorado (Noel Clark, Joseph Maclennan), the LCI in Kent (Antal Jakli) and Tokyo Institute of Technology (Hideo Takezoe, Fumito Araoka) collaborate in the physical characterisation of mesophases. We work with mathematicians (Lutz Tobiska) and engineers (Georg Rose) at OvGU. Tamas Börzsönyi and Janos Török (Budapest) are collaborators in granular physics. Within SPP 1681, we cooperate with several groups, e. g. Sabine Klapp (TU Berlin), Silke Behrens (KIT) and Stefan Odenbach (TU Dresden).

5.5 Scientific equipment

Otto von Guericke University Magdeburg provides laboratory and office space and the required basic facilities. The university finances administrative support for the project. The necessary experimental equipment is available in the Department of Nonlinear Phenomena (ANP) of the Institute of Physics. The experimental facilities include:
- Laboratory rooms for optical studies.
- AxioImager Pol polarising microscope fully equipped with Imaging hardware/software with a set of compensators for quantitative measurements of retardation and CRI Polscope module for spacially-resolved measurements of birefringence.
- Long-range microscopes.
- Temperature controlled chamber for the preparation of freely suspended films.
- High-speed CCD Camera (Photron Fastcam-Ultima APX) up to 100.000 frames per second.
- EMCCD Camera (Andor, iXON 888) up to 100.000 frames per second.
- Shamrock i500 imaging Spectrometer (Andor)
- Confocal Laser Scanning Microscope with an option for NLO microscopy (Leica TCS SP8)
- Computational facilities (Software: Comsol, Matlab and Mathematica).

5.6 Project-relevant interests in commercial enterprises

no

6 Additional information

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Magdeburg, October 14, 2018 Prof. Dr. Ralf Stannarius apl. Prof. Dr. Alexey Eremin