New Application (New proposal within SPP 2171)

Drop Impact on Soft (Adaptive) Surfaces

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1 State of the art and preliminary work of the applicant

A. State of the art

A tremendous amount of studies of droplets in fluid dynamics have been concerned with condensation or evaporation, drop spreading and drop impact mainly on three classes of substrates: either rigid solids, deep liquid pools or rigid solids covered by more or less shallow layers of liquid. Related problems range from the entrainment of surrounding gas to different modes of drop decomposition and splashing - and are yet often insufficiently understood. New directions include effects of phase transitions on the impacting drops when in contact with the hot or cold surfaces and the influence of chemical or topographic surface micro- or nano-patterning.
Only recently has the community started to discover the immense potential of soft, flexible or switchable surfaces - both to gain scientific understanding and for technology. Firstly, soft or flexible surfaces interacting with liquids widely occur in nature and daily life, e.g. for drop spreading / impact on skin or food, dew collection on plants, drop impact on wet paint or during printing processes. Second, exploitation of visco-elastic substrates for, e.g., improved rates of condensation [1], cell-culturing [2], measurement of cellular attachment forces and tractions [3] and splash prevention [4], was demonstrated. Third, the rigid-surface limit of contact line dynamics on soft surfaces may solve conceptual mathematical problems of flow singularities at moving liquid-solid contact lines [5] and allow a generalization of the wetting of solids to a broader class of materials. This led to a constantly rising interest in the field of contact line statics and dynamics on soft solids. However, particularly rapid dynamics are still insufficiently characterized and understood. Previous studies are almost completely limited to side view imaging on large scales or to low contact line velocities. Overviews of literature can be found in Refs. [6–8].

A.1. General concepts of rigid, liquid-coated and soft (visco-)elastic substrates

The equilibrium shape of a small droplet on rigid, smooth and chemically isotropic surfaces can be determined by the well-known Young-Laplace equation, balancing the components of the surface tensions tangential to the substrate, $\gamma_{\rm lv}\cos\theta_{\rm eq} = \gamma_{\rm sv} - \gamma_{\rm sl}$, where $\theta_{\rm eq}$ is the equilibrium contact angle and $\gamma_{\rm lv}$, $\gamma_{\rm sv}$ and $\gamma_{\rm sl}$ are the liquid - vapor, solid - vapor and solid - liquid interface tensions, respectively. Solid interface tensions are of the same order of magnitude as those of liquids to vapor. For heterogeneous substrates, the Wenzel [9] or Cassie-Baxter [10] equations are the most prominent extensions.
The normal component in the surface tension balance, pulling the three-phase contact line out of the substrate plane as well as the Laplace pressure acting on the contact line are disregarded. Exactly those forces lead to the formation of a "wetting ridge" in soft solids, see Fig. 1a. Its shape and size are determined by both the soft solid's and liquid's interface tensions and the shear modulus E, typical size scales are on the order of the elasto-capillary length, $\ell_{\rm ec} = \gamma_{\rm lv}/E$, tens of micrometers for solids of elastic moduli E <100 kPa. For rigid solids, E is on the order of 100 GPa. Thus, the normal force is so small that the corresponding deformation of the solid would be on the nanometer, thus molecular, scale and can be ignored. A dimple forms under the drop due to Laplace pressure in the drop [11–13].
Carre et al. determined the profile of a soft solid near a sessile drop optically [16]. The *entire* deformation profile of the substrate in vicinity of a contact line has been, e.g., determined using optical profilometry (after removal of the drop) [17–21] and confocal microscopy [22]. Those profiles are well described by the theory of Karpitschka et al. [23]. The theory of Long et al. [24] for rubbers was extended to thin layers by Zhao et al. [25], where a small dimple is formed next to the contact line, in contrast to the continuously decreasing slope on bulk gels [13, 26]. Here, nonlinear effects seem to become relevant [27]. Overall, thin layers of elastic material above rigid substrates typically display smaller wetting ridges [13]. However, the geometry in immediate vicinity of the three phase contact line is *only* determined by the liquid's properties, independent of drop size and substrate thickness [26, 28]. The shape of the ridge can be predicted by measuring both the

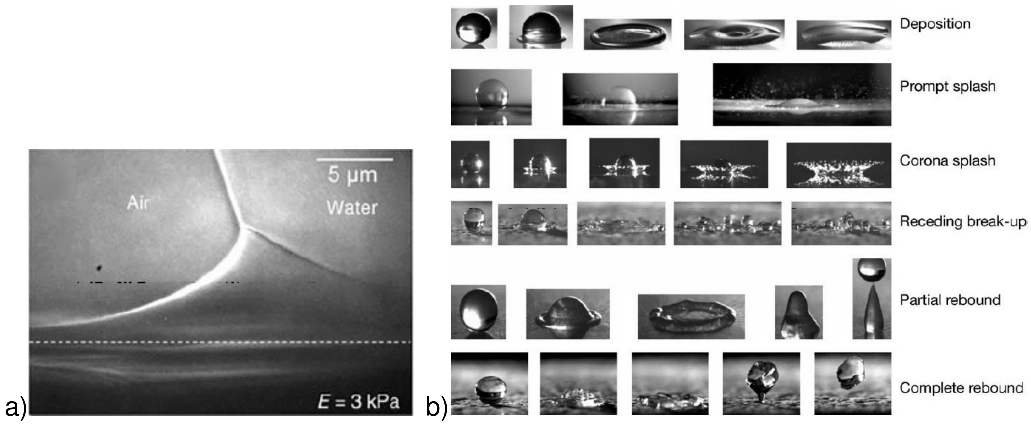

Figure 1: a) X-ray image of a wetting ridge of a water drop on PDMS gel [14], b) Possible scenarios of drop impact on a smooth solid substrate [15].

microscopic and the macroscopic contact angles [14, 29]. On the microscopic scale, the Neumann relation applies at the contact line, while Young's law holds on large scales. Phase separation of the gel matrix from its solvent occurs at the contact line [30].

The contact angle of sessile drops on soft substrates of comparable surface stress increases with decreasing substrate stiffness. Apart from the ratio of the elasto-capillary length to the drop radius, the length scales of molecular interactions were proposed to be relevant [31]. Advancing and receding contact angles are lower than on rigid substrates [6]. Contact angle hysteresis increases with decreasing stiffness [32–35], and the wetting ridges can adjust their profiles as the contact angle changes [19–21].

A.2. Wetting and de-wetting: spreading and impacting drops

Spontaneous drop spreading: The spontaneous spreading of low-viscosity drops on rigid solids occurs in 3 regimes: During the initial 0.1 ms, spreading is dominated by a balance of capillary forces and inertia, expressed in a scaling of the contact radius $R(t) \propto t^{1/2}$ irrespective of the contact angle. After this, the balance of capillarity and inertia prevails, but the contact angle to the substrate becomes relevant, causing lower scaling exponents [36, 37]. Third, after ≈10 ms, viscous dissipation becomes the main resistance to spreading, the wetting follows Tanner's law $R(t) \propto t^{0.1}$ [38].

On soft solids, one expects additional dissipation due to the formation and motion of wetting ridges as well as potential effects of the surface deformation on the spreading dynamics. However, the latter seems to be less relevant. Experimental results exhibit a cross-over from an initial regime independent of softness and contact angle [39, 40] to a regime dominated by substrate elasticity and inertia [41] to a late regime where the time scale of viscous relaxation becomes relevant [16, 42–45]. The crossover time from the inertial to the visco-elastic regime decreases with decreasing elastic shear modulus E of the substrate [40]. This visco-elastic regime is characterized by a stick-slip motion of the contact line. Due to the surplus dissipation in the substrate, spreading is slower than on rigid solids of identical contact angle, an effect termed "visco-elastic braking" [42]. The wetting ridge was found to spread at the base and tilt at the top during such stick-slip events using high-speed X-ray imaging [46]. The microscopic Neumann balance is maintained between slip events. The deformation and stick-slip dynamics of wetting ridges during spreading, related to a dynamic traction on the substrates, was recently modeled by Karpitschka et al. [23]. It is usually reported that substrate thickness does not affect the spreading dynamics in a large range, however recent experiments of Zhao et al. [25] showed sliding towards thicker (effectively softer) regions: Thus, this result should be re-visited.

Spreading of impacting drops: Drop impact on soft substrates intuitively bears a number of interesting aspects, as here not only deformations near the contact line, due to capillary forces, but also due to the impact pressure must be expected. Particularly at high impact velocities, substrate deformation may substantially influence the direction and dynamics of ejecta sheets and thus splashing. Drop impact is of large practical relevance, but it also offers a convenient method to force rapid contact line motion. Drop impact studies

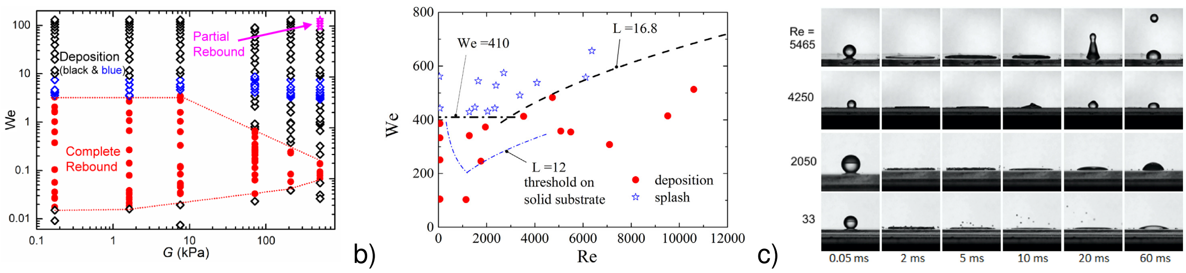

Figure 2: Impact regimes for millimetric water drops on soft visco-elastic substrates a) at low velocities [35]. The splash threshold increases compared to stiff substrates b) on $\approx 45\ \mu$m thick PDMS gel layers on a glass slide [52], c) impacting drops different viscosities on identical soft substrates at identical impact conditions [52]. The Weber number ranges from We=$300 \dots 490$.

on soft substrates are still scarce, and limited to side view imaging (except Ref. [47]) showing the overall phenomenology, but no detailed view of neither the contact line region nor the interaction of entrained gas, drop liquid and substrate at the base of the drop. Further experimental studies accompanied by a theoretical / numerical description are severely needed.

Impact on rigid solids is typically categorized using dimensionless numbers, most prominently the Reynolds and Weber numbers ($Re = \rho VR/\eta$, $We = \rho V^2 R/\gamma_{\mathrm{LV}}$, with V impact velocity, R drop radius, η dynamic viscosity, ρ liquid density). Besides properties of the liquid, the ambient gas, the surface roughness, patterning, porosity as well as its wetting properties are of essential relevance to the outcome of individual impact events [48, 49]. One typically distinguishes six impact scenarios for vertical impact [15] , shown in Fig. 1. General reviews are found in Refs. [48, 49].

Similar to the spontaneous spreading of drops on visco-elastic substrates discussed above, there is little influence of the substrate stiffness on the spreading phase and maximum spreading diameter of impacting drops [35,50,51]. Changing the substrate thickness from 0.3 mm to 3 mm did not influence the impact outcome in the experimental range of Ref. [35]. The extended occurrence of rebound, presumably due to entrained air layers, is remarkable (Fig. 2a). For one structure, it was shown that visco-elastic instead of rigid hydrophobic pillars lead to enhancement of drop sticking [51].
However, the soft layer's elastic modulus and thickness do not always leave the spreading dynamics unaltered: Howland et al. [4] found a substantial increase of the splashing threshold with decreasing elastic modulus for ethanol drops impacting on thick visco-elastic samples. They identified a decrease in the radial velocity of the levitated lamella with decreasing elastic modulus and explained the splash reduction by substrate deformations occurring during the early spreading stages. However, substrate deformations have never been quantified. In addition, the increase of splash threshold is smaller on thin elastic layers on a rigid substrate [4, 52]. On thin gel layers of 45μ m thickness on rigid substrates, the splash threshold is elasticity-independent [52]. Kittel et al. also varied the drop's viscosity to obtain a splashing map in terms of Re and We (Fig. 2b). Interestingly, splashing occurs preferably at low Re, i.e. it is enhanced by an increase in drop viscosity, see Fig 2c.

De-wetting: Much stronger manifestations of the substrate softness are found for receding contact lines, e.g. after drop impact [35, 53] or in dip coating investigations [54]. The receding of the contact line of water on PDMS gels and bitumen slows down immensely with decreasing elastic modulus, where more energy is dissipated near the contact line. In addition, droplets pause in their maximum spreading diameter, supposedly due to adaption of the wetting ridge, for a longer time on softer substrates [35]. In this case, faster retraction is accompanied by larger receding contact angles [35]. This allows for complete rebound in a large range (Fig. 2a). In case of partial rebound, more fragments are formed from the central liquid column on harder substrates [35]. Kajiya et al. [54] found four regimes of contact line receding, determined by the characteristic time scale of substrate relaxation compared to the receding velocity. At high as well as low velocities, the contact line moves continuously. Two modes of stick-slip occur when both time scales become similar.

A.3. Related Systems: Layer of Viscous Liquids and Lubricant Infused Slippery Surfaces (SLIPS)

Moving contact lines on viscous liquids, in particular thin oil layers, at first glance bear striking similarities with those on soft solids. Spreading of a liquid on a thin oil layer likewise leads to the formation of an oil ridge

near the contact line (Ref. [55] and own preliminary experiment). Again, this ridge grows over time and moves with the contact line, and also leads to pinning upon receding. Analogies of thin elastic and capillary surface layers were pointed out recently [5]. A unified description of aspects of drop impact of viscous, visco-elastic fluids and soft solids on hard rigid solids has been put forward [56]. But to which extent can such analogues be employed? This requires detailed studies of the substrate deformations coupled to the dynamic forcing of liquid contact lines.
Studies of drop spreading and impact on shallow liquid layers focused on identical liquids, impact on deep pools of different liquids was investigated recently (e.g. Ref. [57]). Different viscosities in both liquids or Marangoni stresses can lead to micro-and macroscopic pattern formation. Drop impact in layers of immiscible liquids focused on films of millimetre or slightly submillimeter depths [53, 58–60]. Few micrometer thin oil layers have been further exploited to eliminate impurity effects of rigid solids [61, 62], while the influence of film deformations was disregarded.
Lubricant infused patterned or porous substrates are of immense interest: self-cleaning, water-repellency and even ice-phobicity are predicted. Few aspects of drop impact on such surfaces were studied [63–65]. As on visco-elastic substrates, the influence of the substrate properties is most pronounced during the receding phase if the drop made contact with the oil [63, 64]. Effects are most pronounced for a large viscosity ratio of drop to substrate. Even here, little is known about the detailed dynamics of the oil layer in interaction with the contact line. For the slow dynamics in the contact line region of sessile [66, 67] and sliding [55], the oil ridge dynamics was measured using confocal microscopy. This demonstrates that also these flexible and adaptive substrates bear a vast number of open questions concerning their behavior upon spontaneous and forced, in particular rapid, contact line motion.

B. Preliminary work of the applicant

The applicant has mainly experimental and fundamental modeling expertise in the fields of liquid crystals, hydrodynamics, micro-gravity research and granular matter, including responsibilities and own funded research projects, see CV.

B.1 Experimental techniques

The applicant has substantial expertise in setting up, conducting and evaluating complex optical (high-speed imaging) investigations of soft matter, in particular thin films, bubbles and phenomena related to impacting drops. Classic shadowgraphy and other side-view imaging techniques have been employed to obtain basic information of drop or bubble dynamics. I have experience in (polarizing) microscopy and basic experience in confocal microscopy. I am experienced in high-speed phase contrast X-ray imaging. In addition, I prepared topographic micro-structures and patterns of variable wettability due to chemical surface modification in the Nanolab cleanroom in Twente. This emphasizes my ability to develop / employ the sophisticated optical imaging techniques in this proposal.
Experience in non-standard imaging techniques relevant to the proposal are described in detail in the following:

Monochromatic, bi-chromatic and white-light interferometry have been employed to measure thicknesses of membranes, air layers and thin liquid layers. During my PhD studies, I have employed interferometry to measure the thickness of thin free-standing liquid crystal films and free floating bubbles. Concerning the subject of this proposal, such interferometric measurements can be employed to smooth, flat but also to height patterned and to deformable substrates under assumption of small deformations. For larger substrate deformation, e.g., the thickness of entrained gas layers can still be deduced if the shape of one of the deformed surfaces is known. Depending on the choice of light source, relative or even quantitative height profiles in the range of tens of micrometers can be obtained [68]. One important disadvantage is the need of very high light intensity due to substantial loss by multiple partially reflective interfaces. This is particularly disadvantageous for fast processes occurring on length scales larger than 2-3 mm.
Frustrated total internal reflection imaging (FTIR) is an advanced technique for the determination of contact / non-contact of a liquid with a transparent substrates. The angle of incidence of a polarized, monochromatic incident beam is adjusted such that total internal reflection occurs at the substrate's surface in contact with vapor, whereas light is transmitted in wetted regions. Such wetted spots appear dark while dry regions appear bright, see Fig. 3. More strictly speaking, FTIR additionally provides distance information in a range of up to $\approx 250\ \mathrm{nm}$ between the substrate and the liquid through grey-scale information, due to a partial reflection of the

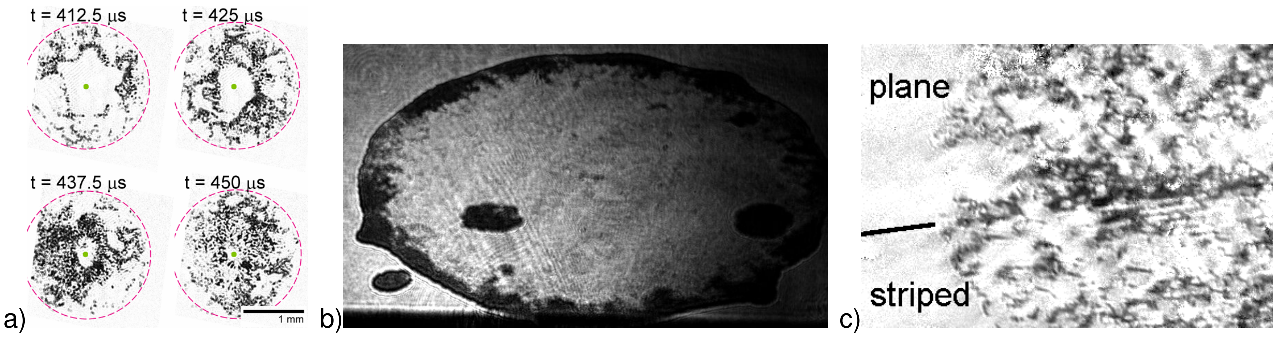

Figure 3: FTIR images obtained on different substrates: a) propagating wetting front under a methanol drop impacting on a hot sapphire plate (processed image, dashes: instantaneous spreading radius), b) solidifying hexadecane drop after impact onto a cold plate (raw image), c) wetting pattern during water drop impact on a hot sapphire plate of micro-patterned wettability (background corrected image, 470μs after impact, rec. at 100 000 fps, structure period 25 μm)

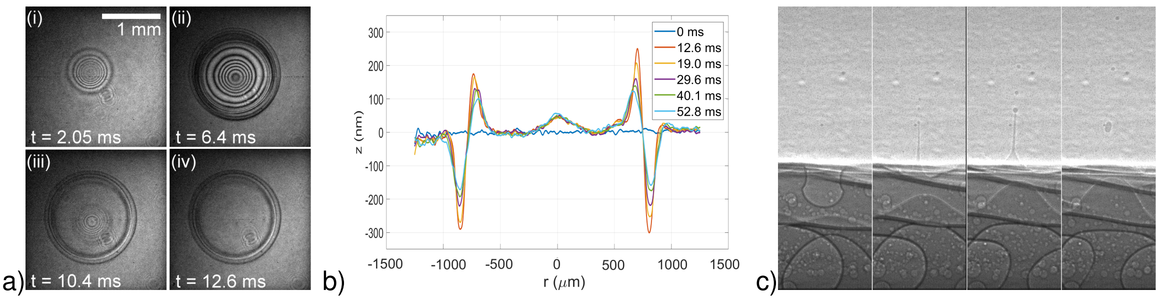

Figure 4: a) Reflection DHM intensity images of a drop impacting and rebounding on an 8.5 μm thick silicon oil layer, central ring patterns interference due to an entrained air film, outer profile in (iii) and (iv) is due to oil layer deformation; b) surface deformation profiles of the oil film during relaxation; c) high-speed phase contrast X-ray image sequence of jet and daughter drop ejection by vapor bubble bursting after impact of an ethanol-water mixture drop on a hot plate. Sessile and floating vapor bubbles exist in the drop.

evanescent wave [69]. A different adjustment of the angle of incidence, such that total internal reflection occurs at both the substrate-gas and substrate-liquid interfaces, allows the observation of solidification processes (Fig. 3) . Gas-liquid-substrate contact lines are visible due to the geometry and the local change of refractive index. TIR imaging can also be employed on non-flat surfaces: micro-structured surfaces as well as thin oil layers (in limit of small deformation) covering rigid substrates (see e.g. Ref. [61]). Both were proven in own test experiments. FTIR requires comparatively low light intensity, allowing for extremely short shutter times (on the order of 1/1 000 000 s) even for comparatively large magnifications and exposure of centimeter-sized spots, see e.g. Fig. 3c. A second advantage is the comparatively unambiguous detection of wetted regions.

An efficient technique for determining surface profiles of, e.g., oil films or visco-elastic surfaces of sufficient reflectivity, is **digital holographic microsopy (DHM, by Lyncee Tec)**. At the University of Twente, transmission and reflection DHM are available, reaching frame rates of 1 000 fps (full resolution). Height profiles are reconstructed from the phase and amplitude of a hologram taken at known angle by the commercial software Koala. While the spatial resolution is still diffraction limited, the height resolution is down to the nanometer scale. I have developed a drop spreading and impact setup for the reflection holographic microscope, we are currently studying drop impact on thin oil layers (Fig. 4). Current activities involve attempts to measure the thickness of an oil film *underneath* a spreading water drop, by adding absorbing dyes to resolve film profiles during air entrainment or by changing the refractive index ratio between surface layer and the drop.

High-speed phase contrast X-ray imaging [70] provides a different view on processes related to rapidly changing liquid interface geometries due to slight X-ray diffraction. However, all interfaces in a millimeter-sized bulk object are projected onto the same image due to the large coherence of the white beam X-rays from the synchrotron. Frames rates of up to 270 000 fps and recording times of approx. 50 ms can be reached in transmission imaging at a spatial resolution of max. 2 $\mu\mathrm{m}$ at the APS at Agronne Ntnl. Labs. The X-rays are slightly absorbed by the drop liquid, and they are slightly diffracted at any liquid-gas interface, see example image Fig. 4c. During the next years, an upgrade to the facility is scheduled, allowing for high-speed imaging at lower resolution. We recently investigated the dynamics of vapor bubbles and of the vapor layer under drops impacting on hot plates. X-ray tomography has been previously applied to visualize both liquid-liquid [71] as well as gel-gas and gel-liquid interfaces [14, 46], in high spatial but very low temporal resolution. In context of this proposal, X-ray imaging is advantageous for imaging, e.g., axisymmetric structures with few features (e.g. the evolution of the initially entrained gas bubble [72]) and for obtaining high-resolution of self-covering 3D structures as during splashing processes.

B.2 Spreading and Impacting Drops

During the last two years, the applicant has investigated drop spreading and impact experimentally in mainly three situations: on hot plates, on cooled substrates and on rigid substrates covered by thin oil layers. First results on all three topics are currently in preparation for publication.

Drop impact on hot plates: Drops impacting on a sufficiently hot substrate hover over a tens of micrometers thin layer of vapor while they slowly evaporate (Leidenfrost effect). The minimal temperature for floatation is the static Leidenfrost point T_{LS}. Under the action of impact pressure, vapor layers thin to nanometers or vanish completely. Thus, the dynamic Leidenfrost temperature for impacting drops depends on the impact velocity, $T_{\mathrm{LD}}(v) > T_{\mathrm{LS}}$. An intermediate regime of partial lamella hovering appears [73]. However, increase in temperature is also accompanied by drastic qualitative and quantitative changes in the wetting patterns. This includes a sudden change from patterns of large bubbles and channels with slow dynamics to small-scale structures exhibiting high-frequency propagating wetting patterns when exceeding $\approx T_{\mathrm{LS}}$, see Fig. 3a. This correlates with the formation of an immense vapor bubble near the impact point in X-ray data. Frequencies increase with increasing temperatures, while length and time scale of local contacts decrease. Detailed analysis of TIR data provides clear evidence that this oscillation must be attributed to pressure buildup and release beneath the drop and capillary wave propagation. First experiments indicate that the dynamic wetting patterns under such violently boiling drops can still be influenced by modifying the substrate's wettability, Fig. 3c. Oscillatory wetting is absent for drops impacting a topographically patterned substrate. In our recent X-ray studies, we also revealed that the actual T_{LD} is substantially higher than expected from the previous optical measurements in Ref. [73].

In a second study, we investigate drop decomposition on hot plates in reduced ambient pressure. The splashing threshold is tremendously reduced for $T > T_{\mathrm{LS}}(p)$, which can be understood within the model of Riboux and Gordillo [74] as the lack of lamella touch-down after its sufficiently fast ejection. Additionally, one would expect splash suppression below a critical pressure [75], with some modification of the threshold due to the temperature dependence of the liquid's properties. Astonishingly, we observe a counter-intuitive re-entrant no splash - splash -no splash - splash transition in a range of pressures and temperatures.

X-ray phase contrast imaging provides a new perspective on drops impacting on hot plates. Droplets hitting a highly thermally conductive substrate, whose initial impact occurs without contact, may still touch down at a later instant due to spontaneous local breakdown of the vapor layer. At later stages, capillary waves propagate radially inward on the retracting lamella, often focussing in a central jet towards the substrate. Interestingly, the spread out lamella of drops slightly below the temperature of complete floatation often ruptures near the impact point, forming a donut-shaped (toroidal) drop whose hole re-coalesces later. Statistically, temporary lamella rupture is a relevant criterion to determine the dynamic Leidenfrost temperature.

Drop solidification during spreading and impact on cold substrates: When a liquid drop of, e.g., tin or water, hits a supercooled surface, it will eventually freeze. We are interested in how and with which solidification structure freezing evolves spatio-temporally. We quantify the solidification of spreading and impacting hexadecane drops at varied impact velocity using TIR. Three regimes are distinguished: dendritic growth from seeds present on the substrate, nucleation in small-scale structures from local points, and solidification with uncountable number of seeds from the point of touchdown radially outward. Regime transitions coincide with predictions of nucleation theory. Current work in progress is the extension of the setup to operate in nitrogen atmosphere at temperatures below 5°C to investigate icing and extending our impact study.

Drop interaction with substrates covered by thin oil layers: In collaboration with J. Snoeijer in the ITN Lubiss, we study droplet interaction with lubricant infused surfaces. As even little is known about impact on thin viscous oil layers, we first focus on this part. While spreading is hardly affected by thickness and viscosity of the film, the relaxation is substantially slowed down with increasing oil viscosity. We conducted a side view / bottom interferometry study, revealing the global contact line dynamics, impact regimes and details on air layer entrainment and its breakdown which is accompanied by pattern formation. We study the long-term relaxation of the oil layers' distortion using DHM, Fig. 4 a,b. Currently, we work at an extension to early times and wetted situations.

B.3 Other Related Studies

Floatation and impact of hydrogel beads (collaboration with S. Waitukaitis, Leiden)[P1]: When hydrogel beads impact the hot surface of, e.g., a pan, they start bouncing spontaneously [76]. Energy is constantly supplied via evaporation-driven pressure oscillation cycles underneath the slightly deformed sphere. We found and investigated the Leidenfrost floatation of these soft solids in [P1]. In order to understand the mechanism of energy input, we currently analyze the dynamic wetting patterns under bouncing hydrogel beads using FTIR.

Measurement of interface tensions [P2,P3]: We developed and applied a novel method for the accurate measurement of interface tensions of liquid crystalline films to aqueous surfactant solutions. We exploit the buoyancy of an air bubble trapped under a closed liquid-crystalline film submerged in water.

Relaxation dynamics of soap and smectic Bubbles [P4,P5,P6]: This complex deals with the question how specific properties of nanometer thin membranes can affect the macroscopic relaxation dynamics of millimeter and centimeter-sized free-floating bubbles. First, we analyzed oscillations of soap bubbles experimentally and numerically [P4]. Next, we reproducibly produced bubbles of distorted initial shapes from smectic liquid crystalline films and characterized their relaxation dynamics (e.g. [P5,P6]). Layer reorganizations substantially hinder the relaxation, such that surface area oscillations are suppressed. With increasing film thickness and homogeneity, relaxation is immensely slowed and the smectic-air surface tension becomes practically irrelevant. Surface deformations and wrinkling, typical signatures of elastic systems, are observed[P6].

Drop impact and embedding in free-standing smectic liquid crystalline films [P7]: We investigate the evolution of contact angles and curvature during the embedding of picolitre drops in free-standing smectic films. An abrupt transition is observed, probably due to wetting by a smectic monolayer.

Menisci and patterns of free-standing liquid crystal films [P6,P8,P9]: Due to the layered structures of smectic liquid crystals, their meniscus profiles deviate from those of unstructured liquids. In addition, spontaneous patterns form in the smectic C phase. Interferometry and Atomic Force Microscopy are applied to determine shapes of smectic menisci, patterns are mainly characterized by polarizing microscopy. Patterns in the homogeneously thick regions of the film can arise as a consequence of meniscus patterns.

1.1 List of project-related publications

Articles published in peer-reviewed international journals:

[P1] S. Waitukaitis, **K. Harth**, M. van Hecke, From Bouncing to Floating: The Leidenfrost Effect with Hydrogel Spheres, *Phys. Rev. Lett.* **121** 048001(2018).

[P2] K. Harth, L. Shepherd, J. Honaker, R. Stannarius, Dynamic interface tension of a smectic liquid crystal in anionic surfactant solutions, *Phys. Chem. Chem. Phys.* **17**, 030201 (2015).

[P3] K. Harth, R. Stannarius, Measurement of the interface tension of smectic membranes in water, *Phys. Chem. Chem. Phys.* **15**, 7204 (2013).

[P4] U. Kornek, F. Müller, **K. Harth**, A. Hahn, S. Ganesan, L. Tobiska, R. Stannarius, Oscillations of soap bubbles, *New J. Phys.* **12**, 073031 (2010).

[P5] K. May, **K. Harth**, T. Trittel, R. Stannarius, Dynamics of freely floating smectic bubbles, *Europhys. Lett.* **100**, 16003 (2012).

[P6] K. Harth, Episodes of the Life and Death of Thin Fluid Membranes - Patterns and Dynamics at the Crossover from two to three Dimensions, *PhD Dissertation*, Otto von Guericke University Magdeburg (2016).

[P7] S. Dölle, **K. Harth**, T. John, R. Stannarius, Impact and embedding of politer droplets into freely suspended smectic films, *Langmuir* **30**, 12712 (2014).
[P8] K. Harth, R. Stannarius, Corona patterns around inclusions in freely suspended smectic films, *Eur. Phys. J. E* **28**, p. 265 (2009).
[P9] K. Harth, B. Schulz, C. Bahr, R. Stannarius, Atomic force microscopy of menisci of free-standing smectic films, *Soft Matter* **7**, p. 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

As the review of the literature shows, there is a substantial lack of understanding of the *rapid dynamic* interaction of the contact lines of liquid droplets with soft visco-elastic substrates. The vast majority of studies considers only the macroscopic outcome of the interaction of spreading or receding drops with the substrate, and a number of speculations are made upon the processes near the contact line. Despite its high fundamental relevance, detailed studies of rapid contact line motion on such soft solids are practically non-existent. I presume this is mainly due to the only recent immense development of the field and due to the lack of commercial availability of appropriate measurements methods with high spatial and temporal resolution. Drop impact represents an ideal test case.
In this project, I aim at shedding light on the connections of macroscopic to microscopic processes related to moving contact lines on soft (and adaptive / switchable) materials. Several (mainly) optical methods will be employed to quantify the deformation dynamics in a variety of substrates in connection with the macroscopic behavior.
The main objectives are:

- **(i) Methods:** Development and test of applicability of different optical measurement methods to obtain simultaneous time-resolved information on the macroscopic behavior coupled to the local substrates' deformation dynamics. The appropriate methods will then be applied in the different situations of the research objectives.

- **(ii) Impact on flat, visco-elastic layers:** The key objective is understanding the coupled liquid and soft-substrate dynamics near rapidly moving contact lines and in a global context. Drop impact is an ideal and highly relevant test scenario. Static drops and even slow drop spreading on visco-elastic layers have been in the focus of research on wetting on soft elastic substrates starting about ten years ago. Mostly, non-miscible systems were studied, predominantly water on PDMS gels. However, the case of impact has been addressed only recently, scarcely, and without substantial theoretical insight. The problem lies in the usually insufficient temporal / spatial resolution accessible within the previously applied techniques. Quantitative comparison between oil films and gel layers is lacking.

  I will apply sophisticated high-speed imaging techniques (WP(i)) in both bottom and side view to gain fundamental insights to the coupled liquid-solid deformations and contact line dynamics in response to fast contact line motions. The study will cover the influence of the gel's thickness and its rheological properties and the influence of the liquids' properties (viscosity, surface tension ratios). I will vary the impact velocity in a large range, such that phenomena from air-cushioning induced rebound to violent splashing are observed. The hosting Ohl Group (Magdeburg) owns a facility for ultra-high impact velocities of up to 400 m/s. Splashing on soft solids bears several peculiarities (increased splash threshold in general, reduction of splash threshold at increased drop viscosity), which are insufficiently characterized experimentally. However, one may expect additional surprises also during the very early stages of drop impact on a visco-elastic medium, also including instability and retraction characteristics of entrained air layers, which have never been imaged in detail. Microscopic data near the contact line as well as global features will be analyzed.

- **(iii) Substrates of spatially varying elasticity:** The literature so far only comprises studies of artificially produced visco-elastic surfaces of homogeneous rheological properties. On the other hand, oil infused slippery surfaces are handled as the solution for easy drop removal, and even prevention of icing. Not much is known about their stability and about the detailed droplet dynamics on such surfaces. This served as inspiration to study gel surfaces of spatially varying stiffness. First, we require a comparison between oil-infused and gel-infused substrates covered by identical hard matrix structures, the latter automatically solve the problem of lubricant depletion. Second, we will investigate contact line statics and dynamics on substrates of spatially varying elastic moduli. This is achieved by filling a structure of flexible PDMS pillars with PDMS of different stiffness. I will vary the visco-elastic properties as well as the topography, i.e. pillar height, thickness and spacing in the range accessible by preparation. As these surfaces have not been studied before, all time scales from statics to fast impact will be of interest. Last, we vary the pattern of the initial structures in a selective manner, e.g. to induce certain drop decomposition patterns.
- **(iv) Deformable pillars:** In nature, we often find textured surfaces where the topographic features are flexible, e.g. the setae on gecko feet or surfaces of plant leaves. Here, I study the interaction of moving droplets contact lines with artificially produced structures (flexible PDMS pillars). The objectives are a) understanding of the influence of pillar height, thickness and shape on the dynamics of spreading and receding drops, including the potential for recovery of the substrate after removal of the liquid, and b) studying the influence of such pillars' visco-elastic properties and positioning on the outcome of (high-speed) drop impacts, e.g. in the drop decomposition during retraction or splashing, and c) investigation of the wetting of the base substrate during drop impact in comparison to stiff textures, where wetting is related to a Cassie-Baxter to Wenzel transition known for superhydrophobic surfaces.
- **(v) Adaptive / switchable substrates:** The optical high-speed characterization facilities developed in (i) shall be made accessible to other participating groups in the SPP. The objective in this part is based on the specific needs of the collaborator, depending on their research questions. Current plans are listed in WP(v).

From the present perspective, the second funding period could include an in-depth continuation of objectives (iii)-(v) depending on the results of the first funding period. Aspects of interest beyond this would include a) spreading and impact on tilted soft surfaces, b) the interaction of the above soft substrates with particle-laden and suspension drops, c) the influence of substrate softness on boiling or solidification processes.

2.3 Work programme including proposed research methods

Millimetric droplets will be prepared by dripping from capillaries of different diameters at low flow rate controlled by a syringe pump. Impact velocities are controlled via the release height, up to few meters. For large heights, the path of the droplet will be encased in order to reduce the scatter in the impact point due to disturbances by air flow.

WP(i) Imaging methods: Besides standard side and bottom view imaging, a number of **sophisticated high-speed imaging techniques** will be applied to access the substrates' dynamics upon interaction with liquids. All bottom view data will be accompanied by synchronized side views. The optimal high-speed imaging method for different stages of the experiment will need to be determined. We aim to mainly use techniques A-E, due to their local availability, high-frame rate potential and broad applicability.

- (i.A) (Confocal) Fluorescence Height Mapping (FHM): The soft materials will be mixed with a fluorescent dye, which is excited by a laser in bottom view. The fluorescence signal is isolated with a filter and captured by a high-speed camera. For large deformations compared to the layer thickness, the thickness profile can be extracted directly from the overall fluorescence intensity, based on proper calibration [77]. An increased height resolution and imaging of thicker substrate layers can be achieved using a confocal setup (also applied for i.B) previously applied to measure deformations of $30\ \mu\mathrm{m}$ thick oil layers [62]. These methods provide access to substrate profiles under the drop and in non-wetted regions.
- (i.B) Local Strain and Deformation from Particle Tracking (APTV): (Fluorescent) colloidal particles will be suspended in the gel layers, either at pre-defined immersion heights (as in Ref. [26]) or randomly distributed in the bulk of the gel slab. Their (3D) displacements will be tracked in high-speed videos. For this, one may either focus on a defined immersion height and apply the confocal setup from WP(i.A) or obtain

global images with increased focal depth, depending on the particle size, positions and concentration. Astigmatic particle tracking velocimetry [78] is a method to track the 3D positions of particles dynamically, from a sequence of 2D images. This method is expected to be ideally applicable to track deformations and strains in my substrates at high temporal resolution. A second candidate is streak analysis developed in the host group of C.-D. Ohl [79]. The methods are applicable under the drop and in non-wetted regions.

- (i.C) Quantitative Schlieren imaging: High-resolution surface profiles of gel layers have been recently obtained using Schlieren imaging [25]. This technique is easily extendable to high-speed imaging. As it relies on top illumination, it is only applicable to late stages of drop spreading and in the receding phase or for sliding drops, in absence of shading.

- (i.D) Interferometry: Interferometry is a versatile method to extract thickness profiles, in particular of thin films. Using a dual wavelength setup (e.g. Refs. [62, 68]) allows extraction of slopes and quantitative thickness data. However, the method cannot be easily applied to obtain thickness profiles underneath spreading and impacting drops: Usually, the reflectivity of the soft layer / drop interface is very small due to small change in refractive index, and no information can be extracted. In addition, when larger areas need to be illuminated (typically 0.6-1 cm diameter) for the spreading stages of impacting drops, interferometry suffers from low image intensity, and thus requires comparatively long exposure times. Second, deduction of complex height profiles in particular in regions with large slopes can be challenging or impossible. Here, the direct methods WP(i.A) are advantageous. A second problem for height mapping of the surfacial layer arises when thin gas layers are entrained between the drop's underside and the substrate, e.g. upon low-velocity impact or lamella levitation during high-velocity impacts. Then, the interference pattern related to the gas layer's thickness dominates. We will exploit this for the quantitative thickness measurements of such entrained gas layers, and if needed couple it to a fluorescence method from WP(i.A).

- (i.E) (Frustrated) total internal reflection: FTIR is an optical method exploiting the total internal reflection at an interface and the related emergence of an evanescent wave to obtain information on local wetting and thickness profiles in the range up to ≈ 200 nm. It can be applied in case of soft and liquid surface layers given the local deformations are small enough such that the critical angle of total internal reflection at the substrate-gas interface is not exceeded - such regions will appear dark. Due to high intensity of the reflection images, it allows imaging at ultra high speeds $> 100\,000$ frames per second. Here, it will be applied to detect contact patterns and morphologies under impacting drops, in particular when entrained gas is present. In case of rigid surfaces, we will also be able to track height information [69]. In addition we aim at exploiting fluorescence generated by the evanescent waves for the early detection of phase separation in collaboration with S. Karpitschka (Göttingen).

- (i.F) Digital Holographic Microscopy: DHM extracts the topographic information of a reflective interface in a single step from the amplitude and phase of a hologram. The measurable slope is limited by the resolution of the microscope objective. Transmission and reflection DHM are available at the University of Twente through collaboration with J. Snoeijer. During my current work, we have constructed a setup allowing drop impact and side view imaging. At the moment, frame rates of approx. 1 000 fps can be reached, with potential plan of extension to higher frame rates. It is ideally applicable to deformations of the substrate where there is only one sufficiently reflective interface, e.g. oil layer profiles ahead of a moving contact line where the contact line is concave. We are currently working on methods to extend it to the regions underneath the drop in contact and non-contact cases. DHM may be used for characterization of comparatively slow processes, e.g. receding motion.

These custom-built high-speed imaging solutions will be validated using slow processes, e.g., sessile drops or slowly moving contact lines. Validation, substrate characterization and also the measurements related to textured substrates will include low frame rate imaging as well. Here, I will apply scanning **confocal microscopy** to characterize the substrates' (static or dynamic) deformation as in Refs. [55, 66, 67] or the position of fluorescent tracers (WP (i.B)). This includes collaboration with D. Vollmer (Mainz), who is an expert in the method. A confocal microscope is available in Magdeburg through collaboration with R. Stannarius and A. Eremin.

WP(ii) Impact on flat, visco-elastic layers: I will apply the methods from WP(i) to study the substrate's dynamics coupled to the droplet's dynamics in detail. This includes mapping of a) potential entrapped gas

layers (interferometry), b) the deformation of the substrate prior to contact with the liquid (FHM), c) the process and morphology of contact formation (TIR and interferometry), d) the substrate's surface deformations (FHM)) as well as e) the dynamic evolution of displacements and strains (via APTV) underneath the drop and in uncovered regions, (d and e) when applicable with high-speed Schlieren imaging WP(i.C). DHM may be employed to obtain substrate deformations, but will be less favorable to the previous techniques. For fast impacts, fast and thin sheets of liquid are ejected near the contact line as precondition for splashing, this ejection changes with substrate properties. Both substrate deformations as well as the liquid's ejection characteristics need to be characterized simultaneously. Controlled low-velocity contact line motion will be achieved by investigating a liquid brigde between two flat plates whose distance can be changed at controlled rate. This latter experiment is mainly a test case for the application of this setup to textured surfaces in WP(iii) and (iv). As the clarity of imaging is substantially increased, and some features are inaccessible optically, we will characterize a) early times of lamella ejection during high-speed impacts, b) dynamics of an entrained bubbles and c) possible propagating waves on the droplet surface coupled to the substrate's elastic response by high-speed X-ray phase contrast imaging at APS Argonne in collaboration with K. Fezzaa.
While the shape of the soft solid surface deforms during the impact, there are also elastic waves emitted from the impact site. In particular shear waves are interesting as they have a relatively low velocity of only a few meters per second and vary with the shear modulus. Due to the shortness of the impact duration the stressed region remains confined before it is emitted into the soft elastic solid. The droplet impact would thus act very similar to a "seismic hammer" used in geophysics to probe the mantle of the Earth. In medicine, a vibrational source is combined with elastography to probe tissues [80]. There, the speckle pattern obtained with an ultrasonic receiver array is analysed and the elastic properties of the tissue determined. A research grade ultrasound system from Versasonics (Vantage 64LE) and the previous expertise of Prof. Ohl is available at the OvGU through a research partnership between Stimulate (Prof. G. Rose) and Prof. Ohl's group (host). We will measure not only the surface deformation, but also the deformation within the soft elastic substrate using the ultrasound receiver.
To gain fundamental insights, I will vary the properties of the substrates (thickness of gel layer, rheological properties of the gel layer) but also the properties of the liquid drop, including surface tension and viscosity. Theoretical modelling and simulations using the boundary integral method will be in collaboration with J. Snoeijer (Twente) and U. Thiele (Münster). I experiments are accompanied by and serve as validation for numerical simulations in collaborations with G. Lecrivain and S. Aland (both Dresden, on drop impact on oil layers and visco-elastic surfaces) and F. Kummer (Darmstadt, primarily in liquid bridge geometry). I plan to collaborate with S. Karpitschka on the analysis and modeling of deformation and strain fields from APTV.

WP(iii) Substrates of spatially varying elasticity: As very first step in WP(iii), I will comparatively study the dynamics on classical oil infused slippery surfaces (in particular pillar structures filled with silicone oil) and hard micro-textures filled with visco-elastic PDMS gel, for identical surface topography and, e.g. similar viscous properties. Subsequently, I will consider substrates of flexible PDMS micro-structure (initially pillars of varying height, thickness and spacing) filled with a PDMS gel of different rheology or silicon oil. These substrates will then be used in drop spreading, possibly sliding and impact experiments. In addition, I plan to investigate the contact line motion in the geometry of a liquid bridge spanned between two flat substrates. Contact line motion is then induced by reducing / increasing the plate distance dynamically, allowing better control of the contact line velocity. This setup will be tested with homogeneous visco-elastic substrates.
FTIR (own test experiments) and interferometry [81] are applicable on hard micro-structured substrates. DHM is also not expected to provide problems if the magnification is large enough, we plan similar experiments with oil-infused surfaces in the beginning of next year. Regarding the fluorescence-based techniques, there are no obvious restrictions. Substrates are prepared in collaboration with M. Kappl and D. Vollmer (Mainz). Confocal characterization of static and slow-moving contact line situations are in collaboration with D. Vollmer (Mainz), additional measurements in collaboration with R. Stannarius / A Eremin (access to confocal microscope facility in Nonlinear Phenomena Group). Numerical modeling is planned with S. Aland (Dresden).

WP(iv) Deformable pillars: Here, we use the same substrates as in WP(iii), however in the bare, unfilled state. We will perform dynamic experiments including drop spreading, sliding and impact. We will characterize the influence of the flexible patterned surface on the contact line motion micro- and macroscopically using high-speed imaging and microscopy. The (expected) transient deformation patterns and local pinning processes will be analyzed. Topographic properties will be varied in the accessible range of substrate production. Collaborations are as in WP(iii).

WP(v) Adaptive / switchable substrates: The methodology used in this work package will be determined by the exact requirements of the substrates provided by collaborating SPP groups. I expect to use side / bottom view imaging and microscopy in the vicinity of the contact line to be successful for all samples. The applicability of the other techniques is decided in situ. My optical high-speed imaging facilities are meant to be made accessible to other groups in the SPP, mainly experimental and surface preparation, who seek to obtain according data in a collaboration. Currently intended substrates for investigation are: a) Adaptive substrates containing rotating Janus-particles at fusable waxy surfaces (A. Synytska, Dresden, theory: J. Harting, Jülich), b) we will attempt to resolve structures and wetting ridges related to the interaction of grafted polymer brushes with moving drops (F. Schmid and D. Vollmer, Mainz), c) samples covered by aligned elongated cellulose fibres and responsive surfaces (C. Zollfrank and O. Lieleg, München, and S. Gorb, Kiel), d) surfaces covered by photo-switchable AAP brushes in interaction with nano-particle laden liquid drops (B. J. Ravoo, Münster), e) light-induced switchable deformation of soft gel surfaces (A. Böker, S. Reinicke, Potsdam), 1) quantify and 2) study the effects of the altered topographic (and visco-elastic) properties of a gel substrate on the contact line dynamics during drop spreading and impact.

Sample preparation and characterization:
The samples used in WP(ii) consist mainly of layers of PDMS gels of different thickness and rheological parameters and viscous oil layers on top of rigid transparent substrates (glass microscope slides or cover slips, or sapphire when temperature control is implemented). Viscoelastic PDMS gels are prepared by mixing the precursor and crosslinker in different mixing ratios. For fluorescent imaging or APTV, a fluorescent dye / colloidal particles are added. The mixtures are then de-gassed and either spin-coated on rigid support or filled in a mold and left to cure at elevated temperature. The thickness of spin-coated layers up to $\approx 50\ \mu\mathrm{m}$ is determined using a spectrometer. For a given mixing ratio and batch of gel precursor, a rheological characterization is performed: A small amount of the uncured mixture is placed between the plates of a rheometer and left to cure. After this, a standard measurement of the visco-elastic properties is performed.
The samples in WP(iii) and WP(iv) are planned to be prepared / obtained in collaboration with M. Kappl and D. Vollmer (Mainz). Template structures are prepared from hard UV-curable PDMS by spin-coating a thin layer and subsequent selective curing by exposure under a mask (similar to the standard preparation procedure for SU-8 photoresist micro-structures). The template is subsequently filled with softer PDMS and cured, which is next extracted. For very soft gel structures, this process is repeated. The different parts of the structure are dyed flourescently, to distinguish them in the final sample. At first, we aim at studying pillar structures of different spacing, area filling fraction and heights. This can in a second step be extended to more complex patterns. In case of WP(iv), these structures are used as is or with surface treatment. For WP(iii), the structure is filled by capillary suction, either with gel precursor of different rheological properties or with silicon oil. The extreme case of hard structures is considered comparatively. The soft PDMS structures need to be characterized regarding their stiffness against local deformations. For this, experimental results can be obtained indirectly by changing the aspect ratios, keeping one dimension fixed. For pillars of known (e.g. circular or square) cross-section, one may estimate the parameters from finite element modeling or standard formulas. In addition, force-displacement measurements could be carried out in funding phase 2, e.g. application of local force by an AFM tip.
The samples in WP(v) are of diverse surface properties. They are obtained from other groups in the SPP, and the investigations and methods applied will need to be adjusted to the substrate.

Measurements of topography in static cases as well as of advancing and receding contact angles are conducted on my setup in side view, with evaluation in matlab.

Data evaluation:
The raw experimental data (mainly high-speed videos) will be processed using mainly custom-written, e.g., matlab scripts. The applicant has previous experience in the evaluation of FTIR, interferometry and digital holographic microscopy raw data. The evaluation for (confocal) fluorescence height mapping is based on careful calibration on the present sample (confocal, described in Ref. [62]) or calibration samples of the identical batch of dyed gel / oil of known thickness. General astigmatism particle tracking velocimetry algorithms and techniques are known in the literature [78]. Realization of experiment and evaluation of WP(i.A,i.B) is expected to require the highest effort of the proposed methods. Validation of the 3D position reconstruction will be done, e.g., against confocal measurements of the particle positions in static test samples. I intend intense knowledge and technology exchange with S. Karpitschka, who will implement APTV to track deformations near fluid-fluid-visco-elastic solid contact lines. After the relevant parameters are extracted, they are further processed

and then compared to a modeling ansatz (see below). For adaptive / switchable substrates in WP(v), the observation methods and evaluation needs will be adjusted to the specific requirements of the samples. This may include application of additional characterization methods such as high-resolution microscopy (available in the Group of R. Stannarius), AFM, etc. (in collaboration with the specific group).

Numerical and analytical modeling
First, we will attempt to describe the data with scaling arguments, as commonly done in the literature. However, we aim at a deeper understanding in particular of the dynamics of the substrate in interaction with the wetting front. For this, detailed analytical models of the will be derived in collaboration with J. Snoeijer (Twente) and U. Thiele (Münster) mainly for WP(i). In particular, we aim at a description in the framework of lubrication theory where possible, which can be extended to simplified numerical models using boundary integral methods. Full numerical simulations will be achieved in collaboration within the SPP whenever the complexity of the problem exceeds theoretical capabilities. In addition, some simple experimental cases will serve for validation of new codes. Numerical (Finite Element Method) modeling will be achieved in collaboration: For WP(i), with G. Lecrivain (Dresden, coupled droplet and substrate dynamics on viscous oil and visco-elastic soft layers) and with F. Kummer (Darmstadt) concerning tests in liquid bridge geometry and impact scenarios. For WP(ii) and (iii), and partly WP(i), Finite Element simulations will be contributed by S. Aland (Dresden).

Presentation / Publication of results:
The results will be published in quality-controlled scientific outlets (mainly peer-reviewed journals) and made available for open access through either open access publication or deposition in a repository (e.g., arxiv), and presented during meetings, seminars and conferences.

Time Schedule of the Project:
WP(i):During the **initial 6 months** of the project, I expect to have received most of the necessary equipment and have built up the main part of my experimental setup (impact and positioning, setup capacity for back-lighted imaging, TIR, interferometry, height mapping (without confocal option). The more sophisticated methods of Confocal Height Mapping and 3D particle tracking (WP(i.A) and (i.B)) first require detailed considerations for the best choice of objectives, dyes, etc. I expect to finish the experimental setup planning and a more detailed literature study in these months, possibly also already the constructions of preliminary test setups for these facilities. In parallel, I will start working on methods of substrate preparation (also for structured substrates) and the first characterization using confocal laser scanning microscopy. **During the first year**, I plan to have finished the setup for confocal height measurements, and for the particle tracking methods, including development of basic evaluation software. Improvements of the methods are expected due to experience during their application during the subsequent duration of the project.
WP(ii): Studies of impact on different soft substrates with the methods of WP(i) are the main objective of the project, with a wide range of phenomena to study. Including the analysis as publication we expect ongoing work spread out over the almost **complete project duration after month 3**, with breaks due to work on other WPs. The highest intensity workload on this WP is expected until month 24.
WP(iii) and WP(iv): Development and learning of substrate fabrication methods are expected in months 1-6 during, e.g. waiting times to obtain fundamental equipment. This will include basic characterization in confocal microscopy. I expect to perform first preliminary studies of the textured surfaces after successful preparation and (partial) characterization in approx. month 10. The first task will be the study of hard oil and gel-filled substrates. For experiments and evaluation of the raw data, we plan approx. **2 months in year 2** and the **majority of year 3**.
WP(v): Experiments with substrates provided in collaboration with other groups in the SPP and their analysis are expected for **short duration (1-2 weeks)** mainly in months 18-36.

2.4 Data handling

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

2.5 Other information

2.6 Explanations on the proposed investigations

Experiments involving humans or human materials: not applicable

Experiments with animals: not applicable

2.7 Information on scientific and financial involvement of international cooperation partners

A collaboration with the Physics of Fluids Group of Prof. D. Lohse at the University of Twente is planned. This allows experiments using digital holographic microscopy and, if needed, spreading and impact experiments in reduced ambient pressure. Both setups are currently available and used by the applicant. In addition, I plan a collaboration on theoretical modeling of spreading and impact experiments (within the SPP) via a joint project of Prof. J. Snoeijer (Twente) and Prof. U. Thiele (Münster).
We plan collaboration with K. Fezzaa (Argonne National Labs) on synchrotron high-speed X-ray imaging of high-velocity drop impact on visco-elastic substrates.

2.8 Information on scientific cooperation within SPP-2171.

My general high-speed imaging setup for imaging substrate and drop interaction dynamics will be available through (also new) collaboration within SPP-2171. Present planned collaborations are:

Modeling: Analytical models and boundary integral method based approaches for (rapid) moving contact lines on homogeneous substrates are developed with J. Snoeijer (Twente) and U. Thiele (Münster). Numerical models of my experimental research will be implemented using finite element codes in anticipated collaboration with S. Aland (Dresden, mainly structured visco-elastic substrates, also plain visco-elastic layers), G. Lecrivain (Dresden, thin liquid films and visco-elastic layers) and F. Kummer (Darmstadt, unstructured visco-elastic substrates, bridge geometry). Selected experimental data will serve for model calibration. The response of the Janus-Particle based surfaces in collaboration with A. Synytska will be modeled by J. Harting (Jülich).

Experimental methods: Confocal microscopy of static and slowly moving contact lines is carried out in collaboration with D. Vollmer (Mainz) and R. Stannarius / A. Eremin (Magdeburg). S. Karpitschka (Göttingen) and I plan close interaction on experimental implementation and data evaluation methods regarding astigmatic particle tracking velocimetry. We plan to apply total internal reflection fluorescence imaging to visualize early stages of phase separation on soft substrates. Digital holographic microscopy and a low-pressure impact facility are available through collaboration with J. Snoeijer and D. Lohse (Twente).

Substrate preparation and interaction with drops: For the preparation of textured substrates, I plan a collaboration with D. Vollmer and M. Kappl (Mainz, elasticity patterned substrates, flexible pillar structures), and with A. Synytska (Dresden, chemical surface modification). Both groups also offer facilities for rheological characterization of gel substrates, if needed. With A. Synytska, we plan experimental investigations of the reorientation dynamics of substrates of Janus-Particles embedded in a waxy matrix in interaction with moving contact lines. I plan collaboration with C. Zollfrank, O. Lieleg (München) and S. Gorb (Kiel) regarding droplet interactions with substrates covered by aligned cellulose fibres, including spreading and impact experiments in Magdeburg and part of the final characterization in München. I will attempt imaging of the response of grafted polymers with D. Vollmer, F. Schmid(Mainz). In a collaboration with A. Böker and S. Reinicke (Postdam), we will 1) quantify the light-induced switchable deformation of their soft gel surfaces and 2) study the effects of the altered topographic (and visco-elastic) properties of a gel substrate on the contact line dynamics during drop spreading and impact. I intend to collaborate with B. J. Ravoo (Münster) regarding the interaction of particle laden drops with photo-switchable AAP brushes.

3 Bibliography

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4 Requested modules/funds

4.1 Basis module

4.1.1 Staff

I apply for funding for an undergraduate research assistant (HIWI) to perform routine characterization and evaluation tasks, in total 2000 Euro for 3 years.

4.1.2 Direct Project Cost

4.1.2.1 Equipment up to Euro 10000, Software and Consumables

General Impact Setup:
A complete droplet impact setup as basis for all more essential optical experiments needs to be constructed. I will need an optical table for mounting the complete setup (Thor Labs: $\approx$ 4000 Euro). The setup requires basic construction materials as well as lab equipment for positioning of cameras and drop ejection (2 large base and large range lab jacks, 2 manual large range linear stages, 3 small-range translation stages for needle positioning), rails and connectors, air shielding tube, additional smaller parts, total $\approx$ 6500 Euro). In addition, I request a syringe pump (4000 Euros).

Camera / microscope lenses:
I further require a telecentric long-distance microscope (e.g. Navitar telecentric Zooom lens system, approx. 6 500 Euro) for flexible and fast magnification / refocussing in interferometry, bottom view, TIR or detailed

high-speed side view imaging. In addition, I request one standard zoom camera objective ($\approx$800 Euro) and appropriate microscope objectives for confocal imaging / fluorescent particle tracking (4000 Euro).

Specific optics for high-speed bottom view imaging methods, see WP(i):
Optical components for readjustment of the laser output to experimental needs are required, including rotation / positioning mounts for the fibre, beam expander, diffusor(s), polarizer.
In addition, the different optical methods will need to be implemented with their specific requirements. This includes optical components as lenses, mirrors, 2 microscope objectives of appropriate numerical aperture for confocal imaging at low and high magnification, pinhole, appropriate prisms for TIR, filters and beam splitter cubes for interferometry and confocal imaging, mounts, posts and connectors (total 12 000 Euro).
Another major component for the height and stress measurements is a high-accuracy (0.1 $\mu\mathrm{m}$) piezo positioning stage with digital control for the confocal objective and a small linear stage (2 000 Euro). In addition, I request a small, two-axis goniometer stage, 5 range, for substrate leveling (300 Euro).
To control the samples' temperature, I request a temperature controller and appropriate (home-made) substrate mount (1 000 Euro).
In addition, I will need standard electronic sensors, to measure e.g. temperatures, humidity, etc. (300 Euro).

Consumables:
The major cost of consumables are fluorescent dyes to dye the custom-prepared PDMS gel substrates for carrying out height measurements and fluorescent colloidal particles for the bulk deformation and strain tracking. These need to be suspended in the respective PDMS precursor mixture prior to spin coating / bulk solidification, or need to be sprinkled on the surfaces in layers. For studies of the textured surfaces, they are similarly needed also to distinguish regions of different stiffness. Typical costs are 900 Euro / 5 mg of fluorescent dyes (Sigma-Aldrich) and 500 Euro / 5 ml fluorescent particles (not bio-compatible, diameter 1 $\mu\mathrm{m}$, Sigma-Aldrich) in solution. Substrates will be re-used as long as possible, but the fabrication requires non-vanishing amounts of material. In addition, I will need the gel precursor components (e.g. from Dow Corning).I request (3000 Euro + 200 Euro)/ year.
For chemicals for cleaning of setup and substrates and decontamination of TIR optics from silicon-oil based residues (e.g. acetone, toluene, absolute ethanol), I request 1200 Euro / year.
For setup elements prone to aging and possible irreversible contamination (syringes, tubing, needle tips, connectors, etc.), I request 500 Euro / year.

In total, I request 56 100 Euro for the 3 years funding period.

4.1.2.2 Travel

For travel to **scientific conferences**, I plan one larger international meeting / year (e.g. APS DFD meeting, European Fluid Mechanics Conference, full conference registration fees), 2000 Euro / year, and 2 local or smaller meetings, e.g. DPG meeting (750 Euro / year, 5 days) and 1 specialized smaller conference (1100 Euro / year, 5 days, based on full registration fee) each year. Additional funding will be needed to attend the meetings / conferences organized by the SPP, in the first and second year 1300 Euro /year (1 x 5 days, 1 x 4 days), in the third year for the planned international conference 1200 Euro (based on full registration fee).
For collaboration: Necessary stays at the Max Planck Institute in Mainz in order to prepare substrates, perform part of the confocal microscopy measurements, I request funding for 3 travels of 5 days per year, 3x600 Euro / year. For measurements in collaboration with the Physics of Fluids group at the University of Twente, I request funding for two travels of 4 days / year, 2 x 520 Euro / year. These collaborations are independent of the participation of the PIs in SPP2171. For other short collaboration visits within the SPP (substrate preparation and characterization or discussion of numerics / theory) I request in total 1000 Euro for 2 travels of 2 days each in years 2 and 3.
In addition, I plan to perform high-speed X-ray imaging experiments at Argonne National Labs (USA) in collaboration with K. Fezzaa, one period of 4 measurement days, dependent on APS relatively short-term scheduling. For this, I request 1600 Euros.

For the total funding period, I request 22670 Euros.

4.1.2.3-4.1.2.5 not applicable

4.1.2.6 Project-related publication expenses

For contributions towards publication costs for submissions to high-level *Open access*-Journals (*Nature Communications, New Journal of Physics* or similar), I request 750 Euros / year.

4.1.3 Instrumentation

The proposed work critically depends on the permanent availability of a high-quality (high light sensitivity sensor, high resolution on the order of 700x200 pixel^2 even at frame rates of $\approx$ 70 000 - 80 000 fps) high-speed camera. The proposed high-speed fluorescence (confocal) height imaging during impact processes, the fluorescent defocusing particle tracking and interferometry all suffer from comparatively low light intensity while short shutter times of the camera are needed to resolve the high-velocity processes. The high-resolution is required to resolve, e.g. the early splashing and sheet ejection dynamics both in side or (varied versions of) bottom view. This requires the choice of high-quality, high-intensity light sources in combination with the proper high-speed camera with high-sensitivity sensor fulfilling the frame rate / resolution requirements. Such a camera is not available in the hosting group of Prof. Ohl and also not in the collaborating group of Prof. Stannarius at the Otto-von-Guericke University Magdeburg. Furthermore, execution of the experiment usually requires operation on two synchronized high-speed cameras for side and (the current implementation) of bottom view (see WP (i)), in case of dual color interferometry even 3 synchronized high-speed cameras. The available cameras Photron Mini AX200 is ideal for side view imaging. For the dual-wavelength interferometry, one high-sensitivity high time and spatial resolution camera is sufficient, if less frequent sampling at a second wavelength occurs concurrently. The high-speed camera consists a central part of all investigations and thus will be in almost daily experimental use.

The high spatial and temporal resolution bottom view imaging techniques in WP(i) in addition require:
FTIR: single-wavelength (low bandwidth) polarized light in case of height-resolved measurements
Interferometry: dual separated wavelength high intensity simultaneous output (for quantitative height measurements)
Fluorescent deformation tracking (particles and confocal thickness measurements): strong excitation of a dye at proper wavelength to maximize the fluorescence intensity, variable exposure spot size (usually (sub)-millimetric for tracking near the contact line, up to approx. 1.5 cm diameter for global thickness measurements). In addition, a line of exposure would be desirable to increase the intensity and thus recording speed in axially symmetric global measurements.
Switchable substrates of B. J. Ravoo and A Böker / S. Reinicke would require UV wavelenth $\approx$360 nm and vis. ≈ 520 nm for reversible switching. Distinguishing different parts of the patterned substrates also requires usage of different fluorescent dyes simultaneous to observation.
Thus, a 4-wavelength simultaneous output laser with optical fibre is required for execution for the proposed technique. Large coherence length is not required.

Based on the above experimental necessities, I selected obtained an offer for a laser (under 4.1.3.1) and the necessary high-speed camera (under 4.1.3.2), which will both be essential for the successful implementation of the proposed work.

4.1.3.1 Equipment exceeding Euro 10000

The laser offered by Laser2000 ([OXX-L4Cc-488/638/x/x-MM] 4-Kanal-Laser-Combiner) fulfills the above requirements and would be versatile and intense enough for use in my experimental setups. It is almost as permanently required as the cameras (except for side view imaging and Schlieren imaging). Another positive feature is that the output intensity at the different wavelengths can be individually tuned. The choice of the laser power is based on experience from my current work in Twente.
I thus request $\approx$ **17650 Euro** for an appropriate laser.

4.1.3.2 Major Instrumentation exceeding Euro 50000

I have reviewed the technical details of high-speed cameras of different companies with regard to the above criteria. In addition, the availability of numerous up-to-date cameras in the Physics of Fluid Group in Twente and their usage in my current research on drop impact on hot substrates, leads to the following decision: The suitable camera for the proposed work is the Photron Nova S12 (quote attached). A further requirement is a sufficient recording time for manual triggering and sufficient observation times for the receding process of the drops, thus the minimal storage version is insufficient. The decision is based on a) the resolution criterion

(Nova S12 allows, e.g., 640x208 pixel^2 or 1024 x 144 pixel^2 at 80 000 fps), and b) the sensitivity of the sensor of ISO 40000 (at least, as the camera will only be available during the next months). A further advantage are the comparatively small dimensions and the option to switch off the cooling fans (avoiding vibrations during the measurement). My current experiments in Twente showed that the lower ISO 25000 is inacceptable for the above techniques when an area of approx 1 cm diameter shall be imaged, as necessary in interferometry and height deformation measurements.
Alternatives: Other cameras with sufficient sensitivity are the Photron Mini AX200 (as available in the Group of Prof. Ohl) and the Photron SA-Z. The Photron Mini cameras do not fulfill the temporal / spatial resolution requirements. The Photron SA-Z is the high-end camera, and thus both substantially more expensive, it is heavier and larger (harder to handle in the setup). The Phantom cameras v2512, v2012, v1612 (quote attached)are larger and heavier and have a smaller sensitivity (ISO 32000). In terms of resolution, the Phantom camera v1612 would be preferable. The sufficient option would be v1612 with fast option. These cameras are more expensive than the Nova S12, ≈ 85000 Euro for the Phantom v1612.

The current preference is clearly for the Photron NOVA S12, due to the higher sensitivity. I request ≈ 65000 Euro.

4.2 Module Temporary Position for Principle Investigator

A full position for the applicant, Dr. Kirsten Harth, is requested.

5 Project requirements

5.1 Employment status information

I receive a DFG fellowship (University of Twente) until 30.6.2019. An application for reintegration funds is planned.

5.2 First-time proposal data

This is a first time proposal.

5.3 Composition of the project group

As I apply for my own position, I will mainly work on the project myself. I will be member of the Soft Matter Group of Prof. C.-D. Ohl in the Institute of Physics, Otto von Guericke University Magdeburg. Collaboration in terms of exchange of experimental expertise, knowledge on preparation and dynamics of soft gels and discussions of experimental results are planned. In addition, I plan supervise Bachelor or Master students.

5.4 Cooperation with other researchers

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

Experimental: Within the SPP, I have agreed to cooperate with Prof. D. Vollmer (Mainz) concerning initial measurements of slow contact line dynamics on deformable substrates using confocal microscopy. I will collaborate with M. Kappl and D. Vollmer (Mainz) in the preparation of topographically patterned PDMS substrates of variable softness (WP (iii) and (iv)). With A. Synytska (Dresden), I agreed on performing optical experiments regarding the substrate and contact line dynamics on switchable - surfaces (WP(v)). She further offered expertise and facilities for surface modification. Collaboration on structural changes related to and dynamics of drop spreading and impact on aligned cellulose nanofibre substrates are planned with C. Zollfrank, O. Lieleg and S. Gorb (München, Kiel) (WP (v)). With B. J. Ravoo (Münster), I plan studies of contact line dynamics of particle laden drops on switchable AAP substrates (WP (v)). I plan collaboration with J. Snoeijer and D. Lohse (both Twente) in terms of using the DHM and reduced pressure facilities which I both partially built up and used during my Postdoc period from 2016-2019 in the Physics of Fluids Group (Twente). I will collaborate with K. Fezzaa (Beamline Scientist High-Speed X-ray facility 32-ID at Argonne National Labs) regarding high-speed X-ray imaging of drops impacting soft solids. In a first phase, this can better resolve dynamics during early stages of splashing and gas entrapment. During the total period of SPP2171, the facility will be updated to higher spatial resolution ($< 1\ \mu$m/pixel), allowing for high spatial and temporal resolution imaging of the contact line dynamics.

Theory / Modeling: I agreed on collaboration with G. Lecrivain (Dresden) and F. Kummer (Darmstadt) on finite element modeling of drop spreading and impact on oil layers and spatially homogeneous visco-elastic substrates (WP(ii)). Experiments shall be carried out for validation of the numerical code, and likewise more detailed simulations shall aid understanding of the experiments. With S. Aland (Dresden), I agreed on collaboration regarding finite element modeling of impact on patterned surfaces (WP(iii,iv)). We attempt (semi-)analytical modeling of spreading and impact experiments in collaboration with U. Thiele (Münster) and J. Snoeijer (Twente) (WP(ii,iii)).

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

D. Lohse, J. Snoeijer, C. Sun, D. van der Meer and A. Prosperetti (Twente, The Netherlands) on drop impact; J. H. Je (Pohang University, Korea) and K. Fezzaa (Argonne Ntnl. Labs, USA) on drop impact on hot plates, X-Ray measurements; R. Stannarius, A. Eremin (Uni Magdeburg) on PhD topic liquid crystals and on granular matter; D. Vollmer (MPI Mainz) on drop impact on lubricant infused surfaces; Scott Waitukaitis and Martin van Hecke (Leiden, The Netherlands) on impact of hydrogel spheres on hot plates; Joseph Maclennan and Noel Clark (Boulder, USA) on liquid crystal dynamics; T. Börzsonyi (Budapest, Hungary) on granular matter

5.5 Scientific equipment

The Otto von Guericke University Magdeburg provides laboratory and office space and the required basic facilities. The project will be carried out in the Group of C.-D. Ohl. The university finances administrative support for the project. Part of the necessary equipment is available in the group of C.-D. Ohl, this includes:
vacuum oven, spin coater, heatable magnetic stirrer, plasma cleaner, light sources (Sugar cube by Edmund Optics, flash lamps), 3 high-speed cameras (Shimadzu HPV: 1 000 000 fps but only 120 frames total and 2x Photron Mini AX 200-900k), gas catapult for projectile acceleration to up to 400 m/s (usable for ultra high-speed impact), pulsed lasers (Nd:YAG Q-switched Laser green and IR), circular polariscope (for birefringence), acoustic levitator, inverted microscopes and objectives, research grade ultrasound system from Versasonics (Vantage 64LE), access to data evaluation software (Matlab, Mathematica etc.), access and support in mechanical constructions by the workshop of the institute.

A spectrometer and another high-speed camera can be borrowed from Nonlinear Dynamics Group of R. Stannarius for interferometric measurements. A confocal laser scanning microscope (LEICA TCS SP8) is accessible via collaboration with R. Stannarius for static / low-velocity 3D substrate characterization. A similar facility is accessible through collaboration with D. Vollmer (Mainz). A rheometer for rheological characterization of the gel mixtures is accessible through collaborations with D. Vollmer (Mainz), J. Snoeijer (Twente), A. Synytska (Dresden). Facilities and expertise for topographic and chemical surface patterning accessible through collaboration with D. Vollmer (Mainz), J. Snoeijer (Twente) and A. Synytska (Dresden). Finite Element Simulation software (not specifically needed for realization of this project) is available in the Nonlinear Dynamics Group (Stannarius, Magdeburg), or access to alternative codes and computational resources (Boundary Integral Method, Volume of Fluids within Gerris / Basilisk open source software) in collaboration with the Physics of Fluids Group (Snoeijer, Lohse, Twente).

Enschede, October 15, 2018 Kirsten Harth