

**SPP 2171 Summer School:  
Dynamic Wetting of Flexible,  
Adaptive, and Switchable  
Substrates**

**Reporte der Beiträge**

Beitrag ID: 1

Typ: **Talk**

## How Surface and Substrate Chemistry Affect Slide Electrification

When water droplets move over a hydrophobic surface, they and the surface become oppositely charged by what is known as slide electrification. This effect can be used to generate electricity, but the physical and especially the chemical processes that cause droplet charging are still poorly understood. The most likely process is that at the base of the droplet an electric double layer forms and the interfacial charge remains on the surface behind the three-phase contact line. Here we investigate the influence of the chemistry of surface (coating) and bulk (substrate) on slide electrification. We measured the charge of series of droplets sliding over hydrophobically coated (1-5 nm thickness) glass substrates. Within a series the charge of the droplet decreases with increasing droplet number and reaches a constant value after about 50 droplets (saturated state). We show that the charge of the first droplet depends on both, coating and substrate chemistry. For a fully fluorinated or fully hydrogenated monolayer on glass, the influence of the substrate on the charge of the first droplet is negligible. In the saturated state, the chemistry of the substrate dominates. Charge separation can be considered as an acid base reaction between the ions of the water and the surface. By exploiting the acidity (Pearson hardness) of elements such as aluminum, magnesium, or sodium a positive saturated charge can be obtained by the counter charge remaining on the surface. With this knowledge, the droplet charge can be manipulated by the chemistry of the substrate.

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Beitrag ID: 3

Typ: **Talk**

## Dancing drops on lubricated surfaces

Recently, there is much interest in droplet condensation on soft or liquid or liquidlike substrates. Droplets can deform soft and liquid interfaces resulting in a wealth of phenomena not observed on hard, solid surfaces (e.g., increased nucleation, interdroplet attraction). Here, we describe a unique collective motion of condensate water droplets that emerges spontaneously when a solid substrate is covered with a thin oil film. Droplets move first in a serpentine, self-avoiding fashion before transitioning to circular motions. We show that this self-propulsion (with speeds in the  $0.1\text{--}1\text{ mm s}^{-1}$  range) is fueled by the interfacial energy release upon merging with newly condensed but much smaller droplets. The resultant collective motion spans multiple length scales from sub-millimeter to several centimeters, with potentially important heat-transfer and water-harvesting applications.

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Beitrag ID: 4

Typ: **Poster**

## The Waltz of Condensate Droplets on Lubricated Surfaces

Infuse a nanostructured surface with a thin oil-film, and now you have prepared a dance floor for droplets to waltz on! We recently discovered when droplets condense on lubricated surfaces, they spontaneously dance in serpentine, self-avoiding fashions (left), before switching to circling motions, like whirling dervishes in a trance (right, bottom). The driving force behind this dance is the Cheerios effect, the conversion of interfacial into kinetic energy. As larger droplets gobble up their smaller neighbors, they leave fresh areas for re-condensation and rebirth of new drops which start their own dance routines in a captivating ballet of renewal. These dancing droplets can revolutionize the way we capture water from the atmosphere, with potentially important heat-transfer applications.

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Beitrag ID: 5

Typ: **Poster**

## Dynamic Wetting of Adaptive Polyelectrolyte Substrates: A multiscale approach

Studying wetting phenomena is of great importance across a wide range of scientific disciplines. As a fundamental research area, wettability dynamics of adaptive and responsive substrates gained significant attention. These substrates include polyelectrolyte multilayers (PEM), Poly-N-isopropylacrylamide (PNIPAM) microgels (MG), and Polydimethylsiloxane pseudo-brushes (PDMS brushes), among others. Notably, an interesting property of polyelectrolyte (PE) substrate is their propensity to swell in a liquid environment, yet the complexities like disparity between time and length scales, and surface deformation beyond the three-phase contact line (TPCL) makes experimental investigations to be quite challenging. Here, we prepare PE substrates by the layer-by-layer method invented by Decher and coworkers [1], with a focus on their wettability at the nanoscale. We use atomic force microscopy (AFM) as the main characterization technique at the nanoscale for investigation of layer properties, swelling kinetics, saturation, with an optical contact angle (CA) tensiometry method for measurements at macroscopic scale. The layer thickness is determined by ellipsometry. The results show that the water CA on silicon wafers coated with polystyrene sulfate (PSS) as outermost layer decreases in water-saturated atmosphere as previously described by Hänni-Ciunel et al. [2]. Furthermore, the PDMS brushes CA measurements are compared to these of PEM of different composition and layer thickness. Here fore, the dip protocol was adjusted to fabricate different PEMs consisting of PEI/PSS/PAH with different layer thickness, polyanion or polycation as outermost layer and addition of NaCl. [3]

[1] G. Decher, *Science* 1997, 1232 –1237.

[2] K. Hänni-Ciunel, G. H. Findenegg, R. v. Klitzing, *Soft Materials* 2007, 61 –73.

[3] The authors acknowledge funding from the DFG, Priority program 2171, KL1165/34, Grant Number 505842923.

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Beitrag ID: 6

Typ: **Talk**

## Contact spreading under drops impacting a substrate covered by a thin oil layer

A drop impacting on a substrate deforms before the wetting occurs and a thin film of air is formed between the liquid and surface. When a drop gets sufficiently close to the surface within a few micrometers, it will most likely contact the underlying surface in a time on the order of 0.02 s. For experiments done under the same conditions, the time that the wetting occurs varies. Here, we consider rigid substrates covered by thin oil layers of different viscosities and thicknesses. We are less interested in the moment of wetting itself, but rather in the dependence of the velocity of the propagating wetting front once contact has been established. It propagates at velocities of few meters per second. The process can also be seen from the perspective of the thin air layer: It ruptures, and the drop and the substrate coalesce. Important parameters are expected to be the entrained air layer thickness, the oil layer thickness and its viscosity, as well as the actual spreading / receding motion of the drop. The actual air layer profile depends on the impact velocity, ambient gas, drop liquid as well as the deformability of the substrate.

We study the wetting front velocity for water and oil droplets impacting on lubricated substrates using high-speed single color interferometry at oblique incidence. Figure 1 shows a time series of the impact of a droplet with velocity  $v = 0.44$  m/s in side view images and bottom view interference. We show how the wetting velocity depends on the oil parameters and the air thickness.

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Beitrag ID: 7

Typ: **Poster**

## Adaptive Wetting: Surface ordering-induced wetting transition on thermos-responsive oleophilic polymer brushes

Stimulus-responsive polymer brushes exhibit a remarkable ability to adapt and transform their properties in response to specific external triggers. Here, we investigate the wetting behaviour of oil on hydrophobic temperature-responsive poly(*n*-octadecyl methacrylate) (PODMA) brushes below and above the bulk melting temperature. Upon depositing a hexadecane droplet on top of PODMA brushes at ambient temperature, a distinctive high contact angle ( $31^\circ$ ) prevents the droplet from spreading on the surface. However, above the melting temperature, a first temperature range characterized by brush swelling without spreading is observed, followed by a second temperature range characterized by a transition from partial to complete wetting. AFM adhesion measurements initially conducted on dry (and later also on oil-wetted) brush layers also exhibit a two-stage transition. At low temperature, the layer behaves like a hard solid surface with minimal deformation as the tip is pulled off the surface. At high temperature, polymer chains are liquified and hence very mobile. Individual chains can stick to the AFM tip and stretch before detaching. At intermediate temperature, the behaviour is more variable and depends also on the details of the measurement procedure in a manner that suggests that the surface layer is still solid while the underlying bulk of the brush layer is already liquid. Comparison to the literature on bulk polymers led to the conclusion that the surface layer, which remains solid at intermediate temperature, consists of a densely packed layer of aligned alkyl side chains. It is indeed known that linear hydrocarbon chains ( $16 < n < 50$ ) exhibit surface freezing: an ordered crystalline layer is formed atop a liquid bulk [1]. Non-linear optical sum frequency generation (SFG) measurements confirmed the scenario of a highly ordered and CH<sub>3</sub>-terminated layer at low and intermediate temperatures and their melting at the upper transition temperature observed in the adhesion measurements. AFM, and the SFG measurements thus provide a consistent scenario of the wetting and swelling behaviour of PODMA, in which the swelling is controlled by the melting of the bulk of the brush layer, whereas the wetting of HD is controlled by the melting of the ordered surface layer of alkyl chains that remain frozen up to a few degrees above the bulk melting temperature. This offers the ability to induce a wetting and swelling transition that can be tuned by varying the length of the side chains. Furthermore, from our observations, PODMA brush layers emerge as a promising platform for finely controlling the entrapment and release of picolitres of fluid locally.

[1] B. M. Ocko, X. Z. Wu, E. B. Sirota, S. K. Sinha, O. Gang, M. Deutsch "Surface freezing in chain molecules: Normal alkanes." APS 1997, 55 3164-3182.

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Beitrag ID: 8

Typ: **Talk**

## Numerical simulation of wetting on biomembranes

After the discovery of biomolecular condensates in biological cells, research in this field has increased dramatically. Cells use those condensate droplets as well as membranes to structure their interior. The interaction between membrane and droplet can lead to topological changes in cells like fission or fusion of membranes, which are little understood.

In this talk, we will present a first numerical model to simulate the wetting of biomembranes via condensates, which can also include topological changes. The model uses a ternary phase-field approach and is thermodynamically consistent, coupling hydrodynamics, surface tensions as well as bending stiffness and inextensibility of the membrane

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Beitrag ID: 9

Typ: **Poster**

## Exploding drops on lubricated surfaces

Traditionally, investigations of Coulomb explosions have focused on charged microdrops levitated using quadrupole electric fields, i.e., a Paul trap. In a surprising twist, our work introduces a simple method to observe Coulomb explosions, with no drop levitation and no external electric field. Instead, we generate a charged water drop using a conventional micropipette, which we then deposit on a plastic petri dish lubricated with a thin oil film. As the droplet evaporates, its radius shrinks until it reaches the Rayleigh limit at which point we observe multiple, highly periodic Coulomb explosions ( $> 60$  events over 30 mins)—the first time Coulomb explosions have been reported for a sessile drop on a surface. The exploding drop produces a finely ejected liquid jet which disintegrate into microdroplets explosively within microseconds, i.e., an electrospray-in-a-drop. Intriguingly, the application of a small electric field  $\sim 10^3 \text{ V m}^{-1}$ , just larger than the atmospheric electric field, aligns the electrospray direction. Our Coulomb explosions span diverse length scales (from micron to millimetres) and time scales (from microseconds to minutes), with potentially wide-ranging applications from nanoscopic material fabrication to electrospray ionization.

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Beitrag ID: 10

Typ: **Poster**

## Strong stretching theory of polydisperse curved polymer brushes

We investigate the effect of polydispersity on the properties of curved linear brushes in good solvent and for molten brushes. To this end, we extend the strong stretching theory for polydisperse brushes to curved geometries and investigate the polymer chain end profiles, bending moduli and other properties for experimentally relevant polymer chain length distributions of the Schulz-Zimm type. We also investigate the properties of End Exclusion Zones (EEZ) that may appear in convex geometries under certain conditions, and show that their position in the brush can be engineered by careful selection of the polymer length distribution. Lastly, we propose a method to engineer chain end profiles by engineering the polymer length distribution.

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Beitrag ID: 11

Typ: **Poster**

## Understanding Microscopic Contact Angle on Super-Smooth Monolayer Silanized Surface Using Atomic Force Microscopy

The phenomenon of wetting holds considerable significance, particularly when examined at the microscale, where it underpins a plethora of technological and fundamental advancements. In this study, we employ Atomic Force Microscopy to delve into the size-dependency of microdroplets, focusing on a scale where surface forces and gravity effects are minimal. To achieve this, we employ a post-silanization technique called peel-off to fabricate smooth surfaces with reduced pinning sites. The roughness values show a dramatic decrease after peel-off down to approximately 1.5 Angstrom. Subsequently, we subject droplets of various non-volatile glycols to impingement from a heated liquid pool. Our findings reveal a non-linear relationship between the contact angle profile and droplet size, which stabilizes at a steady-state value beyond a critical size. This critical size coincides with a significant lengthscale in coalescence theory, where the interplay of capillary, pinning, and viscous energies reaches equilibrium, resulting in a nullification of net available energy. To support this observation, we develop a theoretical model based on energy balance within a merging droplet system, yielding a physically meaningful equation for predicting the size dependency of microscale wetting. Theoretical predictions align well with experimental data and offer insights applicable to diverse liquid-surface combinations.

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Beitrag ID: 12

Typ: **Poster**

## **Investigation of ATR-FTIR-based wetting characterization for nano-patterned surfaces**

The study of wetting mechanisms on patterned surfaces is critical for surface cleaning and functional surface design. We accurately characterize the complete wetting and Cassie-Baxter wetting states on nano-pillar patterned surfaces through an ATR-FTIR-based optical method. The dynamic evolution of the wetting state transitions is also monitored in situ. In addition, through experimental measurements and optical simulations, we verified the technique's sensitivity dependence on pattern geometry. This investigation provides a valuable reference for applying this ATR-FTIR-based technology to more complex nano-texted surfaces.

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Beitrag ID: 13

Typ: **Poster**

## Contact line dynamics on moving fibres measured by X-ray holography

The wetting properties of solid surfaces are extremely important for many daily and industrial processes, like for example inkjet printing, painting, and adhesion of bacteria or viruses on catheter and medical devices [1]. While significant progress has been made in describing the static properties of drops on surfaces, the wetting dynamic is less understood. In particular, the dependence of the characteristic receding and advancing contact angles on velocity cannot be quantitatively described so far. It is furthermore under debate whether the contact line is disturbed by pinning at nanoprotusions and whether this is accompanied by pinching of nanodroplets. At high velocities the interfacial tension might cause that the contact line slides over the topological defects. Most experimental studies hereby rely on optical or confocal microscopy and thus lack the combined spatial and temporal resolution to distinguish these scenarios [2,3].

We present data from X-ray phase contrast imaging experiments, imaging the three-phase contact line of an aqueous solution on a moving glass fibre in transmission geometry. We used the GINIX nanofocusing setup [4] at the P10 beamline at PETRA III (DESY, Hamburg). In our experiment, we achieved a spatial resolution of around 500 nm and a temporal resolution of 10 Hz. We measured the contact line using water and different water-glycerol mixtures as liquid. A glass fibre of about 17  $\mu\text{m}$  thickness (clean as well as PDMS coated) was pulled out of the liquid bath with a velocity in the range of 0.1 mm/s to 25 mm/s by a programmable motor. We observed clear changes in the contact line in relation to the velocity and the viscosity of the aqueous solutions with a high spatial resolution.

[1] J.H. Snoeijer, B. Andreotti, "Moving contact lines: Scales, regimes, and dynamical transitions", *Ann. Rev. Fluid Mech.* 45 (2013), 269-292.

[2] B.B. Straub, H.-J. Butt, A.K. Auernhammer, "Flow profiles near receding three-phase contact lines: Influence of surfactants", *Soft Matter*. 17 (2021), 10090.

[3] P. Papadopoulos, H.-J. Butt, D. Vollmer, "Wetting on the microscale: Shape of a liquid drop on a microstructured surface at different length scales", *Langmuir*. 28 (2012), 8392.

[4] T. Salditt, M. Osterhoff, M. Krenkel, et al. "Compound focusing mirror and X-ray waveguide optics for coherent imaging and nano-diffraction", *Journal of Synchrotron Rad.* 22 (2015), 867.

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Beitrag ID: 14

Typ: **Talk**

## Investigating equilibrium droplets on elastic substrates

The dewetting and equilibration of liquids on soft solids is studied and compared quantitatively with numerical simulations from our tandem project using the experimentally derived input parameters. The size of the explored liquid structures is well below the capillary length and in the range of the elastocapillary length. For these studies, we use a system consisting of a 120 nm thick liquid layer of Polystyrene (PS) with a molecular weight of 18 kg/mol, which is dewetting from polydimethylsiloxane (PDMS) substrates with elasticities varying between 3 kPa and 1.2 MPa. Combining optical microscopy, atomic force microscopy and a lift-off technique, we explore the influence of elasticity on the dynamics of the dewetting process and the 3d shape of the formed transient dewetting rims and the equilibrium droplets shapes.

The dewetting rims reveal a characteristic shape with a dewetting velocity that depends non-monotonously on substrate elasticity. The global shape of equilibrium drops is lenticular and can be described well with numerical models. At the three-phase contact line, however, there is a deviation from this simple geometric shape. The three phase contact line is pulled further upwards and inwards than predicted with an unexpected region of liquid PDMS extracted from the bulk. Besides the changes in drop shape, the demixed liquid also leads to cloaking of the PS droplet and a reduced interfacial tension. This demixing and the deviation from the lenticular shape is more pronounced for softer substrates and smaller droplets.

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Beitrag ID: 15

Typ: **Keynote**

## **Adaptive Wetting: Surface ordering-induced wetting transition and vapor-controlled drop transport on thermos-responsive oleophilic polymer brushes**

Oleophilic polymer brushes serve as liquid-infused slippery surfaces that facilitate e.g. the motion of water drops. Similarly, they can act as flexible surface-grafted transport layers for oil in advanced lubrication techniques. In this lecture, I discuss the wetting properties of poly n-alkyl methacrylate bottle brushes with variable length of the n-alkyl side chains. Brushes with short lauryl side chains (P12MA) are liquid-like at room temperature, swell easily and display near-complete wetting for pure alkanes with contact angles well below  $5^\circ$  [1]. In contrast, brushes with longer octadecyl side chains (P18MA) are solid at room temperature and remain collapsed upon exposure to alkanes with contact angles of approximately  $30^\circ$ .

I will first describe a mechanism of drop mobilization for volatile alkanes enabled by controlling the local vapor concentration above the drop on P12MA. For these samples, the contact angle decreases from  $2^\circ$  to  $1^\circ$  between low and saturated vapor concentration, which induces drop motion. High resolution interferometric measurements of local swelling profiles and contact angles reveal a peculiar non-monotonic distribution along the contact line of the moving drops, which results from a local balance of viscous dissipation and imbalanced Young forces.

Alkane drops on the longer chain P12MA brushes display a two-stage melting and wetting transition upon increasing the temperature: first the bulk of the brush layer melts, while a thin  $\text{CH}_3$ -terminated surface layer remains solid, as evidenced by non-linear optical sum frequency generation (SFG). This first transition leads to partial swelling of the brush layer while the contact angle hardly changes. A few  $^\circ\text{C}$  higher, the surface layer melts and the contact angle drops to near-zero values, as evidenced by the loss of order in the SFG signal concomitant with a strong increase in AFM adhesion.

Our experiments suggest a use of brush layers as a versatile platform for controlled drop motion as well as local entrapment and release of femtoliters of fluids.

[1] Ö. Kap, S. Hartmann, H. Hoek, S. de Beer, I. Siretanu, U. Thiele, F. Mugele, 'Nonequilibrium configurations of swelling polymer brush layers induced by spreading drops of weakly volatile oil', J. Chem. Phys. 158, 174903 (2023).

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Beitrag ID: 16

Typ: **Poster**

## Lateral adhesion of droplets measured with the scanning drop friction force instrument

The understanding of wetting phenomena plays a crucial role in many daily processes. For example, dirt repelling surfaces can be achieved by a hydrophobic coating. Typically, the wetting properties of such a coating are characterized by measurements of the advancing and receding contact angles by sessile drop goniometry [1]. This method provides only information on selected points and local imperfections may remain hidden.

Recently, Hinduja et al, reported on a fast and easy method to analyse friction forces of drops on surfaces [2]. The method was called scanning drop friction force instrument (sDoFFI) and allows to image wetting properties on cm<sup>2</sup> large areas within a few minutes. A drop adheres to an elastic glass capillary while the sample underneath is moved with a constant speed to imitate sliding of the drop. The deflection of the elastic glass capillary provides information about the friction force between the drop and the surface.

Often drops do not adhere strongly to the elastic glass capillary. Then the drop detaches from the capillary while scanning. In order to improve drop adhesion to the elastic glass capillary, I glued metal rings to it. Shaping the metal rings forces the drop to shape as well. In particular, shaping the drop into different width allows to verify the Furmidge equation [3]

$$F=k \cdot \gamma \cdot w \cdot (\cos(\theta_{\text{rec}}) - \cos(\theta_{\text{adv}})).$$

Where F is the friction force of the drop, k is a geometrical factor,  $\gamma$  is the liquid surface tension, w the width of the drop at the three-phase contact line and  $\theta_{\text{rec}}$  and  $\theta_{\text{adv}}$  are the receding and advancing contact angles, respectively. I will present initial measurements of the friction force of drops in dependence of drop shape. Furthermore, I will present sDoFFI measurements of real samples that we tested.

[1] Huhtamäki, Tommi, et al. "Surface-wetting characterization using contact-angle measurements." *Nature protocols* 13.7 (2018): 1521-1538.

[2] Hinduja, Chirag, et al. "Scanning drop friction force microscopy." *Langmuir* 38.48 (2022): 14635-14643.

[3] Furmidge, C. G. L. "Studies at phase interfaces. I. The sliding of liquid drops on solid surfaces and a theory for spray retention." *Journal of colloid science* 17.4 (1962): 309-324.

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Beitrag ID: 17

Typ: Talk

## Fabrication of functional materials with gradient wettability via 3D printing

Control over physical and chemical properties such as porosity and wettability during 3D printing allows for fabrication of functional materials with gradient properties for many applications such as controlled water imbibition and condensation. Common methods for fabrication of functional materials with gradient properties via Digital Light Processing (DLP) 3D printing such as VAT exchange<sup>1,2</sup> and gray scale light projection<sup>3</sup> have drawbacks like weak interfacial bonding between the layers, and no potential for chemical composition change. In this work, we show a facile method for 3D printing of functional materials with gradient wetting properties. We utilize porogen-induced phase separation (PIPS) for the 3D printing of porous materials<sup>4</sup>. By altering the resin mixture during the print through addition of an acrylic monomer component, a gradual change in properties and composition of the final structure is achieved. This way, by increasing the hydrophobic or hydrophilic acrylic components, we enhance the hydrophobicity or hydrophilicity in z-direction to achieve a 3D printed porous part with a wettability gradient. By increasing the porogen content during the printing of porous materials we also achieved an increase in pore size along the z-direction. For both cases with gradient chemical or physical properties, the water droplets could be patterned regarding the imbibition, contact angle or wetting state on different area of the 3D printed structure. The versatility of the method allows for alteration of any resin component during the print generating functional materials with gradient wettability upon demand.

References:

- [1] Balakrishnan et al., Lab Chip, 2024, 24, 505
- [2] Levkin et al. Nature Communications, 2021, 12, 247
- [3] Balakrishnan et al., Small Struct. 2023, 4, 2200314
- [4] Mayoussi et al., J. Mater. Chem. A, 2021, 9, 21379

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Beitrag ID: 18

Typ: **Poster**

## Spontaneous charge separation by drop motion across of a hydrophobic tube

Sliding water drops on hydrophobic insulating surfaces leads to charge separation at the three-phase contact line [1]. Although slide electrification has been investigated thoroughly, understanding the exact mechanism is still a challenge. Electron transfer, formation of the electrical double layer structure, and adsorption of ions have been associated to this phenomenon [2]. Slide electrification is mainly studied through the drops motion on a tilted surface [3]. In this study, we make use of the better control over parameters like drop velocity and drop contact time with the surface to investigate the influence of wetting kinetics on charge separation. Furthermore, the effect of PH and salt concentration of water on slide electrification was explored. Our findings not only provide further insight into the charge separation mechanisms but also can be useful for possible future integration of slide electrification into energy harvesting systems.

[1] B. Leibauer et al., "How Surface and Substrate Chemistry Affect Slide Electrification," Journal of the American Chemical Society, 2024.

[2] P. Bista, A. D. Ratschow, A. Z. Stetten, H.-J. Butt, and S. A. Weber, "Surface Charge Density and Induced Currents by Self-Charging Drops," Soft Matter, 2024.

[3] C. Hinduja, H.-J. Butt, and R. Berger, "Slide electrification of drops at low velocities," Soft Matter, 2024.

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Beitrag ID: 19

Typ: **Talk**

## Numerical simulations of liquid bridges between colloids

Colloids suspended in a primary fluid can be made to aggregate by adding a small amount of an immiscible secondary liquid: the three types of interfacial tension in this system conspire to form liquid bridges between the colloids that bind the colloids together, see as illustrated. The resulting mixtures vary from viscous fluids to elastic gels, with various promising applications, e.g. in the food industry, porous filter manufacturing and battery slurries production. We use Multibody Dissipative Particle Dynamics (mDPD) to simulate two equally sized colloids connected by a liquid bridge suspended in a bulk fluid. The force exerted on the solid particles by the liquid bridge is measured by constraining the distance between the colloids in the simulations. We compare the force versus elongation curves with fitted expressions in the literature and numerical solutions of a phenomenological macroscopic theory. We propose an improved fit function of the force valid over a much wider range of contact angles and droplet volumes

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Beitrag ID: 20

Typ: **Poster**

## Electro(de)wetting with Photoswitches: Control of wetting by electric fields and light

Electro-dewetting (EDeW) was introduced as a novel method to modify surface wetting properties. By using ionic surfactants like dodecyl tetrethyl ammonium bromide (DTAB), the wettability of hydrophilic silicon oxides on a conductive Si substrate can be controlled through an applied electric potential, resulting in an increased contact angle. This behavior is significantly influenced by the pH of the aqueous solution, surfactant concentration, and the applied potential driving EDeW [1]. However, the underlying mechanism remains unclear. For that reason additional experiments are necessary.

In this contribution, we report on EDeW experiments using classical DTAB surfactants which confirm previous results [1], while we also report on the application of new cationic photoswitchable surfactants namely azobenzene triethylammonium bromide (Azo-TB) [2], arylazopyrazole triethylammonium bromide (AAP-TB) [2] and butyl-arylazopyrazole triethylammonium bromide (butyl-AAP-TB) for EDeW. These photoswitches can undergo E/Z photoisomerization reactions that cause drastically changes in surface activity [2]. This offers to fine tune the contact angle changes by E/Z photoisomerization of the AAP-TB amphiphile and we find a further increase in contact angle by about 6° when the samples were irradiated by UV light that triggers the photoisomerization from the E to the less surface-active Z isomer. In previous work [1], it was suggested that surfactants are deposited on the silicon oxide surface through the EDeW process. In order to study the possible deposition of surfactants and the role of a possible prewetting layer outside of the drop and at some distance from the 3-phase contact line we have done SFG spectroscopy and find that surfactants are deposited even several mm adjacent to the drop and that the structure of this surfactant layer as well as the structure of the accompanying water layer changes drastically close to the 3-phase contact line.

[1] J. Li, Noel. S.Ha, T Leo Liu, R. Michael van Dam, CJ Kim; Nature , 572, 507–510 (2019).

[2] M.Schnurbus, R.A.Campbell, J.Droste, C.Honnigfort, D.Glikman, P.Gutfreund, M.R.Hansen, B.Braunschweig; J.Phys.Chem.B.124,6913–6923 (2020).

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Beitrag ID: 21

Typ: Talk

## Switchable topography of lamellar structure by changing temperature and capillary forces

Nowadays, there are many applications where knowledge of surface wettability is needed. For example, these are microfluidic devices, oil/water separation, drug delivery, etc. There are two approaches to control the wettability properties of materials: changing their topography or modifying them with different surface groups. Changing the surface topography, which can be achieved by applying external stimuli including temperature, magnetic fields, pH, light and others, can modify the wettability. One type of material that can reversibly change its topography when exposed to external stimuli are shape memory polymers (SMPs), which can be used to create smart surfaces. Changes in topography can also be associated with crystallization or melting processes. One popular material for this purpose is polyether urethane with poly(1,4-butylene adipate) (PEU-PBA) as a soft segment. These soft segments can crystallize at temperatures below 8 °C allowing switching of mechanical properties of topographically structured surfaces. This ability to change topography has been utilized in the field of wettability [1].

Here we present an advancement of our strategy to allow active switching of surface topography. For this, we use polyurethane with poly(1,10-decylene adipate) (PEU-PDA) as a soft segment [2], which undergoes expansion/contraction associated with melting/crystallization, respectively. A lamellar structure with a height of about 2.5 mm and an interlamellar distance of 2 mm was fabricated using meltelectrowriting technique. The polymer was heated to 200°C and a voltage of 3 kV was applied between the polymer and the substrate. These lamellae can buckle when heated to a temperature higher than the melting point (ca. 61°C) of the soft segments, and this buckling process is reversible. However, the buckling process doesn't have a definite direction. In this case, we can add an additional force that has a direction, such as capillary forces when water droplets are added to the interlamellar space. These forces cannot bend the lamellae at room temperature because the elastic modulus is high enough. However, at temperatures above the melting point, the elastic modulus decreases by more than 12.7 times, and the lamellae are easily bent by capillary forces. After evaporation of the water drop, the deflection of the lamellae remains, but we can restore or change the volume of the bent lamellae by changing the direction of the capillary forces. Controlling the elastic and capillary forces by changing the temperature or perimeter of the drop allows us to change the interlamellar volume. Such adjustments may open us to new possibilities for using such topographies in microfluidic devices or creating tubular structures.

[1] Constante, G.; Apsite, I.; Auerbach, P.; Aland, S.; Schönfeld, D.; Pretsch, T.; Milkin, P.; Ionov, L. Smart Mechanically Tunable Surfaces with Shape Memory Behavior and Wetting-Programmable Topography// *ACS Applied Materials & Interfaces* 2022, 14, 17, 20208–20219

[2] Schönfeld, D.; Chalissery, D.; Wenz, F.; Specht, M.; Eberl, C.; Pretsch, T. Actuating Shape Memory Polymer for Thermoresponsive Soft Robotic Gripper and Programmable Materials// *Molecules* 2021, 26, 522.

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Beitrag ID: 22

Typ: **Poster**

## Microscopic wetting behavior of droplets on swollen polymer brushes

Polymer brushes with lubricants show significant potential in reducing contact angle hysteresis, yet the microscopic wetting phenomena and underlying mechanisms remain poorly understood. Our study focuses on observing and analyzing the formation and deformation processes of microscopic wetting ridges to elucidate the macroscopic reduction of contact angle hysteresis and the dynamic wetting behavior of various liquid drops on Poly Lauryl Methacrylate (PLMA) brushes. Using the hot hexadecane vapor infusion method, we achieved controllable swelling of brush, e.g., from 160 nm to 640 nm within ten minutes. We developed a thin-film interference-based color-matching algorithm to measure the real-time brush thickness. Through this method we observed the oil extraction from brush layer, resulting in a local brush collapsing for tens of nanometers and the region of collapsing extend to around two hundreds of micrometers. However, visualizing the microscopic wetting ridge to establish local Neumann's triangle equilibrium remains challenging. Furthermore, recording and analyzing the dynamic wetting behavior is elusive. In our future studies, we aim to visualize static and dynamic microscopic wetting ridges using a high-resolution confocal microscope. This approach may help us unravel the competition between surface tension-driving solvent extraction and disjoin pressure-induced solvent retention.

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Beitrag ID: 23

Typ: **Poster**

## Modeling the growth of biofilms on soft substrates

We investigate the influence of substrate softness on biofilm growth amending the thin-film model developed by Trinschek et al for rigid solid substrates [1] by the inclusion of a simple description of an elastic substrate [2]. Wettability (described in the mesoscopic model by a wetting energy) is a key factor in the transition between arrested and continuous spreading on rigid substrates [1]. Our focus are changes in the spreading process with changing character of the substrate studied by time simulations of 2d drops/biofilms at fixed surface tension and initial drop volume. We find that with increasing softness from rigid via elastic to liquid-like substrate the spreading velocity of the biofilm decreases at fixed biofilm growth rate and wettability. Further, we discuss how these changes depend on wettability and growth rate. In particular, we show that the transition between arrested and continuous spreading is for increasing softness shifted towards larger wettability and larger growth rate.

[1] S. Trinschek, K. John, S. Lecuyer, and U. Thiele, Phys. Rev. Lett 119, 078003 (2017).

[2] C.Henkel, J.H. Snoeijer, and U.Thiele, Soft Matter 17, 10359 (2021).

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Beitrag ID: 24

Typ: **Talk**

## Dynamical wetting of droplets on flexible sheets

Droplets resting on soft flexible sheets deform into a lens-like shape, presenting exciting opportunities for developing tunable fluidic lenses. Building upon our previously established droplet model, which utilises the boundary element method (BEM), we continue to explore dynamic wetting on thin, flexible sheets. Our research focuses on the intricate interplay between the mechanical properties of the sheet and the behaviour of the droplet, with particular attention to the contact angle during wetting. Inspired by biological membranes, we have incorporated the Skalak model for elastic deformations and the Helfrich Hamiltonian for bending mechanics of the sheet. This integration allows us to examine how tension forces and sheet rigidity can influence droplet shape and wetting dynamics, thereby enabling both droplet deformation as well as controlled navigation of a droplet across the substrate. While primarily applicable to material science, our model also offers valuable insights into interfacial biology, shedding light on the complex interactions between soft flexible substrates and liquid interfaces.

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Beitrag ID: 25

Typ: **Poster**

## Dynamics of Droplet Motility on Hydrophobic Polymer Brush Surfaces Facilitated by Vapor Interaction

Polymer brushes swell completely in good solvents and partially in good vapours. The spreading of volatile liquids on brush layers is governed by subtle combination of hydrodynamic flow, vapour transport and swelling kinetics.

We studied the wetting dynamics of alkanes on oleophilic polymer brush layers of poly-lauryl methacrylate (PLMA), synthesised via surface initiated activators regenerated by electron transfer atom transfer radical polymerization (SI-ARGET-ATRP).

A rich phenomenology is observed including the formation of a wide halo, characterised by a gradient in the degree of swelling of the brush layer in front of the slowly advancing contact line [1]. Local evaporation and condensation conspire to stabilise the inhomogeneous non-equilibrium stationary swelling profiles.

Significantly, when a vapour concentration gradient is imposed above an alkane droplet, macroscopic motion can be induced without exerting external forces.

We obtain both spatial and time dependent swelling profiles in the halo region as well as droplet contact angles from interferometric measurements. We find that the advancing contact angle is lower than the receding contact angle, contrary to typical moving droplets.

We believe that the vapour concentration imposes in a gradient in local swelling near the contact line, corresponding to a gradient in the equilibrium contact angle. As a result, the droplet experiences a pulling force in the direction of the lower contact angle, causing it to move. Additionally, the contact angle appears to exhibit notable viscous drag along the contact line. Utilizing the acquired contact angles, the total horizontal component force is calculated and related to the droplet motion.

[1] Ö. Kap et al. 'Nonequilibrium configurations of swelling polymer brush layers induced by spreading drops of weakly volatile oil'. In: *The Journal of Chemical Physics* 158.17 (2023), p. 174903. issn: 0021-9606. doi: 10.1063/5.0146779.

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Beitrag ID: 26

Typ: **Poster**

## Volatile binary mixtures on polymer brushes

We present a mesoscopic thin-film model in gradient dynamics form for binary liquid mixtures on brush-covered substrates incorporating volatility in a narrow gap. Thereby, we expand models established in [1, 4–6] by incorporating two substances present in each of three bulk phases - liquid, brush and gas. We discuss the different contributions to the free energy, thereby employing Flory-Huggins theory of mixing for the condensed phases and assuming ideal gases for the vapor phase. Interface energies are modeled as linear interpolations of known limiting cases. The resulting six-field model is then analyzed with numerical time simulations showing results with a focus on lateral concentration gradients, notably at the contact line. Limitations and possible expansions are discussed and briefly outlined.

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[3] S. Schubotz, Q. A. Besford, S. Nazari, P. Uhlmann, E. Bittrich, J.-U. Sommer, and G. K. Auernhammer. *Langmuir*, 39, 2023. doi: 10.1021/acs.langmuir.2c03009.

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[5] U. Thiele and S. Hartmann. *The European Physical Journal Special Topics*, 229, 2020. doi: 10.1140/epjst/e2020-900231-2.

[6] Özlem Kap, S. Hartmann, H. Hoek, S. de Beer, I. Siretanu, U. Thiele, and F. Mugele. *The Journal of Chemical Physics*, 158, 2023. doi: 10.1063/5.0146779.

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