Adaptive Droplets Transport Through Coupled Flow Paths

Joint proposal for a research project in the framework of the DFG priority programme SPP 2171

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Project Description

1 State of the art and preliminary work

Soft solids and their ability to create dynamic deformations on the microscale are relevant to a wide variety of exciting new applications such as in tissue engineering, adaptable optics, flexible electronics and soft robotics.

Elasto-capillary

It is generally accepted that surface tension can dominate the behavior of liquids on a small scale. Although solids have surface tensions of similar magnitude, these effects are usually overlooked and not considered. However, liquid surface tension of e.g. fluid drop can cause significant deformations in compliant elastic solids and play an important role in their mechanics. The comment ground of these effects is the ratio of the surface tension to the elastic modulus [1]. This ratio defines a length scale, the elasto-capillary length, γ/E, where γ is the surface tension and *E* is the Young's modulus. At length scales smaller than the elasto-capillary length, the deformation of a solid owing to surface tension can have strong deviations from the "classical behavior". Having surface tensions of simple liquids typically in the range of 10 - 75 mN/m, elasto-capillarity can be very important in elastomers, such as poly(dimethylsiloxane) (PDMS) with *E* in the order of magnitude of MPa. However, the effect of the elasto-capillarity can be dramatically amplified by the "right" choice of the geometry [2], such as in the project proposed microfluidic setup with thin PDMS membranes having an elasto-capillary length of many µm. Nadermann *et al.* could show that a drop of liquid (diameter of a few hundred µm), placed under an elastic thin membrane (□10 µm), causes the membrane to bulge by tens of µm [2]. The induced deformation is governed by the equilibrium of tensions exerted by the various interfaces and the solid membrane by a form of Neumann's triangle.

In experiments characterizing two moving adjacent droplets on a layer of a soft substrate (PDMS) driven by gravity, long-range interactions between the droplets could be observed [3]. The observed long-range elastic deformations lead to purely attractive interactions between the two droplets with a final droplet coalescence on thick PDMS layers, whereas on a thinner layer short-range repulsion occur preventing a direct contact and merging of the droplets. Although first and exciting experiments on the dynamics are carried out in the emerging field of elasto-capillarity [3,4], not many has been done on hydrodynamically dragged droplets on elastically deforming films or membranes. Moreover, the effect of deformations of thin soft membranes on the “backside environment”, such as fluids, droplets, bubbles and segmented flows in channels [5,A2], and their feedback has not yet been investigated in detail and will one goal of this proposed project.

Elastic elements in microfluidics

Elastic elements have also providing new opportunities in microfluidics, where a collection of design considerations to control fluid flows exists. Quake *et al.* were able to show that PDMS can be used to design pneumatically activated valves, which are structural elements for versatile platforms of fluidic control [6,7]. Here, the valves can block the flow in a primary channel by inflating control channels. Along this line, lab on a chip pumping and mixing devices could be developed.

However, already pressure-driven flows in narrow, low aspect ratio channels of PDMS induce elastic deformations. The deformations of the microfluidic channel geometry drastically affect the effective pressure drop and therefore significantly the flow rates as well [8,9]. Based on this phenomenon, a passive microfluidic device was developed that stabilized pulsating incoming flows and generated steady flows at the outlet. The device consists of series of fluid chambers along the flow direction with a thin PDMS membrane serving as the compliant boundary. The deformation of the membrane allows the accumulation of liquid during an overflow and the release of liquid during an underflow to stabilize the flow [10]. Embedding an elastic arch in a millifluidic channel device can be used for passive control of viscous flows. Depending on the flow rate, the arch can “snap” between two states, constricted and unconstructed channel, which have a difference in hydraulic resistance of up to an order of magnitude [11]. Using a “reverse” setup, elastic deformations via mechanical actuation could be used to control and direct fluid flow [12]. Here, a flexible arch, which was prepared by buckling a thin elastic membrane, was embedded in a flexible microfluidic device. The fluid flow could be controlled by coupling the elastic deformation of the arch to a small gap in the microchannel. The deflection of the arch, which is mechanical actuated, can be described by the theory of Euler buckling [12].

Most of the described elastic elements in microfluidics are used to control and manipulate flows in individual channels, however, not much has been done in back coupling channel flows by adjacent flows which are contacted via elastic components. This elastic coupling of fluid flows, which is the aim of the proposed project, may lead to a next generation of fluidic devices with adaption and self-regulation capabilities or for logic element in microfluidic circuits in e.g. soft robotics.

The use of electric fields in microfluidics

The use of electric fields in microfluidic devices is about as old as the discipline itself. The basic idea is attractive, since electric fields provide a contactless way of controlling the flow of liquids in channels [13] and they can be used to manipulate the droplets within the channels, one at a time. Electro-kinetic driving forces have been used since the early 1990s establishing electro-kinetic control of flow using electro-osmosis and demonstrating the separation of different chemical species. Particularly the separation of charged species (e.g. DNA) attracted considerable attention and coined the “Lab-on-a-chip” concept [14]. The promise of an integration of reagent handling allowing multiple operations such as dilution, mixing, incubation, separation, and detection has however been more challenging than initially anticipated [15]. Electric field have also been used to interact with droplets that are transported within the channels, allowing to form, direct, manipulate and sort individual drops [16].

One of the main challenges are the high potentials that are required to drive all of these effects. Given that channel cross sections are typically in the order of 10^{-3} mm^2 and the flow velocities are millimeters to centimeters per second, voltages up to a few kilovolts over 2–10 cm channel distances are required [13]. For droplet manipulation, similarly high potentials are typically used [16]. These high applied potentials not only incur a substantial risk of dielectric break-down in the typically aqueous systems, have additional drawbacks, including, for example, electrophoretic demixing of heterogeneous solutions during electro-kinetic pumping [15]. While Faradaic and Ohmic currents produce an electric field in the electrolyte, electrolytic reactions can strongly influence the acid–base equilibria (including pH) in the microfluidic device, which in turn determine the electro-kinetic transport [17]. Reactions occurring at the electrode surfaces are therefore intimately coupled to local chemistry and electro-kinetic phenomena. This complicates electro-kinetic control in microfluidics and makes it very strongly dependent on the liquid content of the microfluidic devices, which may change with time [17].

These problems may arise because the electrodes are often in direct contact with the fluids. Also, high applied potentials and the prolonged exposure of the microfluidic liquid to electric DC fields can give rise to the above-mentioned problems. This has prompted part of the microfluidic community to focus on pressure-controlled flow control [15].

In a different approach, a buckling instability of an elastic composite membrane has been harnessed to pump liquids through channels [18]. A voltage applied between two electrodes that are directly laminated onto the membrane causes a reduction in membrane thickness, and because of the incompressibility of rubbers, a lateral extension. If the membrane is clamped at its edges, the application of a voltage causes it to buckle. While this is related to our approach described below, potentials of more than 7 kV were required to cause an 80 µm thick membrane to buckle.

In the proposal below, we suggest an alternative mode in which electric fields can be employed to circumvent some of these disadvantages. By applying electric fields across, rather than along the channels, much lower potentials can used to achieve similarly high electric fields as in the examples above (several kV/m). Secondly, by using local electric fields to trigger membrane instabilities, which are enhancing flow instabilities, the temporal exposure of the microfluidic cargoes to the electric field is minimized. Finally, electrodes do not need to be in direct contact with the liquids, minimizing electrochemical reactions and AC fields can be used to minimize electrophoretic effects.

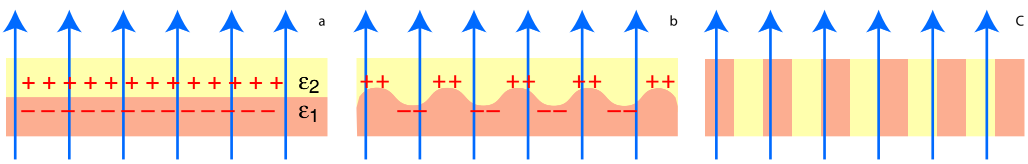

Figure 1: *Schematic representation of an electro-hydrodynamic (EHD) instability. (a) An electric field across an interface between to different dielectric liquids causes the build-up of interfacial displacement charges. These couple to the capillary wave spectrum of the interface, amplifying a single characteristic mode (b). The resulting interfacial torque causes the alignment of the interfaces parallel to the electric field lines (c). Note that the bending of the field lines is not shown in (b) (adapted from [A6]).*

The physical principle that we propose makes use of so-called electro-hydrodynamic (EHD) instabilities, which have been studied for more than 80 years [19]. These instabilities arise at the interface between two fluids, across which an electric field is applied (Fig. 1). A difference in dielectric constants of the two fluids causes the build-up of displacement charges at this interface. The free energy associated with this effect can be lowered by aligning the fluid-fluid interface along the electric field lines, causing a torque on interface segments as long as a perpendicular field component is present [A7]. Triggered by capillary fluctuations, this torque causes the build-up of a sinusoidal surface instability, the wavelength of which is controlled by the ration of the electric field to the interfacial tension. While the amplitude of this instability diverges, a similar effect acting on the interface of an elastic medium produces surface waves with controllable wavelength and amplitude [A6,A7].

The interesting aspect of EHD is that it is tunable over extremely wide length and time scales, controlled by the electrode geometry and applied potentials. When applied over micrometer distances, voltages as low as 10 V are sufficient to trigger instabilities, and spatial instability wavelength as low as 100 nm are feasible [A6]. Importantly for microfluidic applications, the instability can be driven by high-frequency AC fields, preventing many of the problems described above [A7].

This proposal aims to employ EHD instabilities to control the flow of liquids in elastomeric channels, in a minimally invasive fashion. Electric fields can be used to trigger instabilities in thin membranes that separate two channels, thereby controlling the flow of liquids in both channels. They can alternatively be used to interact with flow induced instabilities, and since EHD instabilities are affected by the dielectric content of the channels, they can be used in a feed-back mechanism that is sensitive to the drops and bubbles that are transported within the channels, enabling smart sorting and mixing protocols.

Preliminary work

Freiburg:

The research of applicant Thomas Pfohl is focused on experimental studies of the dynamics of soft matter, self-assembly, and self-organization processes in confinement and microflow. He has a strong expertise as a principal investigator (PI) in developing microfluidic tools and combining these with state-of-the art X-ray techniques, e.g. scanning small angle X-ray scattering (SAXS), X-ray photon correlation spectroscopy (XPCS), coherent diffractive imaging (CDI, ptychography) and microscopy methods, e.g. fluorescence microscopy, polarization microscopy, confocal Raman-microscopy, interferometric scattering microscopy (iScat), optical tweezers, in order to investigate soft materials in flow.

Moreover, the applicant has a deep knowledge in the physics as well as the application of droplet-based (oil-water emulsions-based) microfluidics including the generation, merging, storage and manipulation of these picoliter-sized reaction containers [A2]. His research group was able to form and encapsulate robust three-dimensional fibrin networks, which are essential in blood clotting [A3]. Here, microfluidics devices were tailored to strategically generate, image and study mechanical properties of these biological networks. Recently, nanofluidic systems for reliable and contact-free, geometry-induced electrostatic (GIE) trapping as well as manipulation of charged nano-objects in polydimethylsiloxane (PDMS) was developed [A1]. In these combined micro- and nanofluidic devices, pneumatic systems are implemented, which are capable of controlling the height of nanofluidic channels by the controlled deformation of thin PDMS membranes [A1].

In this proposed project, the Freiburg group is going to develop a microfluidic platform for interfacing fluid transport channels by flexible membranes. This setup is going to use to study, describe, analyze and introduce innovative concepts for adaptive droplets and fluid transport through coupled flow paths. Here, the Freiburg group does not only rely on their expertise in developing microfluidic devices for the proposed project, but also on their vast experience in controlling, imaging, and analyzing complex flows, such as single polymers [A4,A5], droplets [A3] or unicellular parasites. Owing to the collaboration with Prof. Steiner, additionally electrical fields can be used to interact with, stabilize and manipulate flow induced instabilities, which takes this project to a higher level.

Fribourg:

Prof. Steiner has a 30-year track record in soft matter science, focusing particularly on structure formation on sub-micrometer length scales. This includes the study of demixing, wetting and dewetting phenomena and interfacial instabilities. His current research interests include the study and manufacture of energy materials for perovskite photovoltaics and Li-ion batteries, the role of the order-disorder interplay in biological photonics, the use of polymer self-assembly for the creation of (optical) metamaterials, and a range of bioinspired project ranging from artificial nacre to the interaction of insects with plant surfaces.

In the context of the proposed project, he pioneered the use of EHD instabilities for controlled sub-micrometer pattern formation [A6], formulating also the underlying physical principles [A7]. EHD instabilities were also used to interrogate the rheological properties of liquids [A10]. The Steiner group is therefore very well placed to carry out the part of the proposed project that involve electric fields. This work on EHD instabilities is part of a wider interest of the Steiner group in interfacial structure and instabilities. In this topic, there has been a long-standing (28 year) collaboration with Prof. Günter Reiter, with many joint publications over the years. The proposed project aims to extend this successful collaboration with the University of Freiburg, through a microfluidics project, which is the expertise of Dr. Pfohl.

1.1 Project-related publications

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

[A1] M. A. Gerspach, N. Mojarad, D. Sharma, Y. Ekinci, **T. Pfohl**, Pneumatically controlled nanofluidic devices for contact-free trapping and manipulation of nanoparticles, *Part. Part. Syst. Charact.*, 1800161 (2018).
[A2] R. Seemann, M. Brinkmann, T. Pfohl, S. Herminghaus, Droplet based microfluidics, *Reports on Progress in Physics* **75**, 016601 (2012).
[A3] H. M. Evans, E. Surenjav, C. Priest, S. Herminghaus, R. Seemann, **T. Pfohl**, In situ formation, manipulation, and imaging of droplet-encapsulated fibrin networks, *Lab on a Chip* **9**, 1933-1941 (2009).
[A4] N. Strelnikova, M. Göllner, **T. Pfohl**, Direct observation of alternating stretch-coil and coil-stretch transitions of semiflexible polymers in microstructured flow, *Macromol. Chem. Phys.* **218**, 1600474 (2017).
[A5] D. Steinhauser, S. Köster, **T. Pfohl,** Mobility gradient induces cross-streamline migration of semiflexible polymers, *ACS Macro Letters* **1**, 541-545 (2012).
[A6] E. Schäffer, T. Thurn-Albrecht, T. P. Russell, **U. Steiner**, Electrically induced structure formation and pattern transfer, *Nature* **403**, 874-877 (2000).
[A7] E. Schaffer, T. Thurn-Albrecht, T.P. Russell, **U. Steiner**, Electrohydrodynamic instabilities in polymer films, *Europhysics Letters* **53**, 518-524 (2001).
[A8] Z. Lin, T. Kerle, T.P. Russell, E. Schaffer, **U. Steiner**, Structure formation at the interface of liquid/liquid bilayer in electric field*, Macromolecules* **35**, 3971-3976 (2002).
[A9] M. D. Morariu, N. E. Voicu, E. Schäffer, Z. Lin, T. P. Russell, **U. Steiner**, Hierarchical structure formation and pattern replication induced by an electric field, *Nature Materials* **2**, 48-52 (2003).
[A10] D. R. Barbero, **U. Steiner**, Nonequilibrium Polymer Rheology in Spin-Cast Films, *Physical Review Letters* **102**, 248303 (2009).

1.1.2 Other publications

1.1.3 Patents

1.1.3.1 Pending

1.1.3.2 Issued

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2 Objectives and work programme

2.1 Anticipated total duration of the project

36 month, anticipated start 01.10.2019

2.2 Objectives

The aims of the project are to study and analyze the transport of fluids and droplets in channels interfaced by flexible membranes in a microfluidic environment. The proposed microfluidic setup for multiphase flow experiments, consisting of two transport channels separated by a thin and flexible membrane, allows for a defined laminar flow-control within transport channels, an exact positioning of droplets and the opportunity to analyze droplet motion and transport within the channels and cross-correlation between the channels by means of optical microscopy (Fig. 2). Owing to the moving droplets and applied flow acting on both sides of the membrane, the thin elastic membrane will specifically respond and locally deform on the acting moving contact line, flow-induced pressure differences and shear-induced effects, having a strong impact on the transport properties in both channels. Moreover, using electro-hydrodynamics (EHD), which are going to be applied to interact with, stabilize and modify flow induced instabilities of the membranes and the feedback in both transport channels, the coupling and cross communication of the flowing materials and information transport will be amplified and specifically shaped. This membrane-initiated crosstalk will be used to move, adapt, govern, shift and stop droplet motion and specific flows within fluid transport routes and moreover to introduce flow patterns with adaption and self-regulation capabilities as well as on the long term logical links and operations within fluid transport networks.

The Pfohl and Steiner groups combine their areas of expertise, a combination of which is well suited to the aims of this project. To drive the project forward, close teamwork and continuous knowledge transfer, including a regular exchange of PhD students between the two groups, are envisaged.

In more detail, the following objectives will be addressed:

- Development of a strategy and methodology for a microfluidic membrane platform.
- Impact of a moving droplet on a thin elastic membrane.
- Analyzing continuous flow-induced instabilities of thin elastic membranes.

- Coupling of droplets, bubbles and continuous media transport flow by thin elastic membranes.
- Controlling membrane instabilities by electric fields.
- Control of segmented flow by electric fields.

2.3 Work programme incl. proposed research methods

Work-package 1: Impact of a moving droplet on a thin elastic membrane: Developing a microfluidic platform and experiments

Placing a fluid droplet on a thin elastic membrane covering a channel leads to a strong deformation of the membrane in the vicinity of the three-phase contact line. In order to study this phenomenon and later the effect of moving droplets on thin membranes, we are going to develop a microfluidic platform, as it is sketched in Fig. 2. Fluid droplets can be placed on an elastic membrane, which is separating two transport channels. The induced deformation of the membrane in the vicinity of contact line of the droplets does not only change the local cross-sectional areas in transport channel 1, but also has a direct impact on the local cross sectional areas in the underlying transport channel 2. The shape and strength of the deformation can be controlled by the interplay of the used membrane systems and multiphase fluid systems. The crucial parameters of the membrane system are the membrane thickness d, Young's modulus E, Poisson ratio ν, wettability or surface tension γ, surface roughness r, membrane permeability k, swelling behavior as well as the surface structure, which all will directly affect the deformability of the membrane.

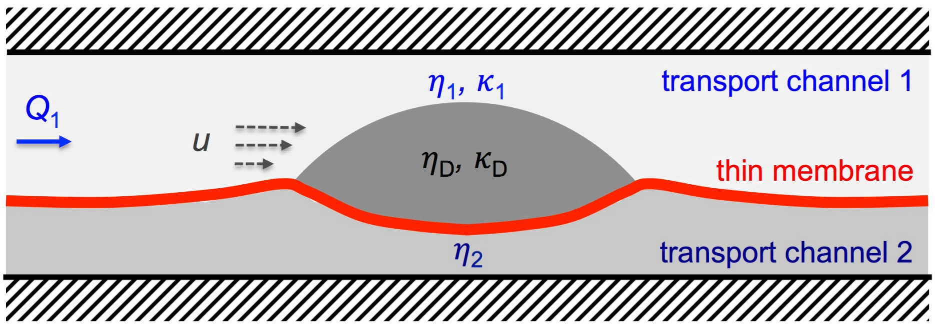

Figure 2: *Schematic representation of the impact of drag on the motion of a fluid droplet in contact with a thin elastic membrane separating two transport channels.*

The microfluidics setups will be fabricated by means of soft lithography. Two different device designs are planned to be realized. For the first design, the channel walls as well as the thin elastic membrane will be produced of polydimethylsiloxane (PDMS), whereas one ceiling will consist of PDMS and the second ceiling of glass (coated with a thin PDMS film) for optical access. The width w and the height h of the narrow transport channels ($w > h$) are much wider than the thickness of the membrane d ($d \ll h, w$), where d of only a few µm can be achieved. The Young's modulus of the membrane can be straightforwardly controlled by the cross-linker amount in the PDMS in the range of $E\sim$ 0.5 – 3.6 MPa [20]. The experimental setup can be imaged and analyzed by microscopy in a side view configuration, as it is sketched in Fig. 2. The second device design is based on a sandwich structure. The two transport channels are each built in PDMS molds and the separating thin membrane is contacted and sealed by these two molds from both sides. Using this design principle, more or less any membrane system of various materials can be incorporated in the microfluidic device. This setup allows for a much higher variability of the experiments, because membranes with a broad range of Young's moduli, porosity, wettability and surface decorations can be used for the sandwich structures. The standard observation direction for (interference) microscopy will be a top view configuration, however, side view microscopy should be realizable as well.

For both experimental designs, the formation and positioning of the droplets and the fluid flow will be controlled by syringe pumps. The droplets will be formed in immiscible oil/water systems with fluorocarbon and mineral oils as the “oils” of first choice due to the low swelling behavior of PDMS. However, organic solvents, such as cyclohexanone, toluene or chloroform, which show stronger swelling effects on PDMS membranes, will have an additional impact on the local deformation of the membrane, which is in direct contact with the droplet. The viscosity η of the aqueous phase can be increased by adding glycerol or dextran. Moreover, silicon oils of different viscosities can be used for characterizing viscosity effects. However, marker particles might be added to the silicon oil in order to increase the contrast, which is typically weak between PDMS and silicon oils. Alternatively, air bubbles/water systems will be investigated as well. In comparison to the droplet systems, the compressibility κ of bubbles has to be taken into account. Therefore, a different motion behavior between dragged droplets and bubbles and the respective occurring membrane deformations are expected. These different behaviors may be exploited for coupling in the transport flows of droplets and bubbles which are separated by thin membranes (see WP3).

The experiments will be imaged and characterized by optical (interference) microscopy and alternatively in case of weak optical contrast by fluorescence microscopy. For fluorescence microscopy studies, fluorescence markers will be added to the fluid phase of the droplets and/or in the membrane. The local flow velocity and local acting flow fields will be determined by particle imaging velocimetry (PIV), using (fluorescent) particle tracking.

Besides of the thickness and Young’s modulus of the thin membrane, further membrane properties, such as the wettability, which can be *in situ* chemically modified on each side of the membrane independently, have a direct impact on the motion of the droplets/bubbles as well as on the deformation of the membrane. Moreover, the motion of the three-phase contact line can be also manipulated by structured surfaces. Pinning of the three-phase contact line to a structured surface will not only show hysteretic effects on the apparent advancing and receding contact angles but will also have a strong impact on local deformations of the membranes and the flow within the channel as well as on the adjacent channels. The wettability and thickness of the membranes as well as the pinning can be controlled by a (switchable) surface nature and structure using *e.g.* surface decoration by switchable polymer brushes. Typically triggers of switching the brush thickness are e.g. the swelling behavior of the droplet fluid, temperature and light. Microfluidic devices (microfluidic design 2) with sandwiched polymer brush decorated flexible membranes will be developed within the framework of this SPP 2171 together with the group of *Rainer Jordan*, Macromolecular Chemistry, TU Dresden. Furthermore, sandwiched microfluidic devices of laterally structured and switchable elasticity may be also introduced. These sandwiched devices with photo-switchable elastic membranes will be developed in the framework of SPP 2171 together with *Ahmed Mourran*, DWI, Leibniz Institute for Interactive Materials, Aachen.

Starting from an almost static setup (after placing the droplet on one side of the membrane), in a next step, we will apply hydrodynamic flow in transport channel 1 in order to study the effects of drag on the motion of the droplets (Fig. 3). The drag-induced motion of the droplets and the initiated membrane deformations in the vicinity of the moving contact line will be studied. Owing to the difference in advancing and receding contact angles, different strengths of deformations are expected in the vicinity of the contact line at the front and rear part of the droplets. The deformation of the membrane has not only an impact in the motion of the droplets and in reverse on the flow conditions in transport channel 1 but also on the flow conditions in transport channel 2. Our microfluidic setup allows us not only to image the moving droplet and the membrane deformations but also to analyze the induced flow fields in channel 2 at once. Within the framework of the SPP 2171, the group of *Andreas Heuer*, Westfälische Wilhelms-Universität Münster, will support our experimental analysis and especially the development of theoretical descriptions.

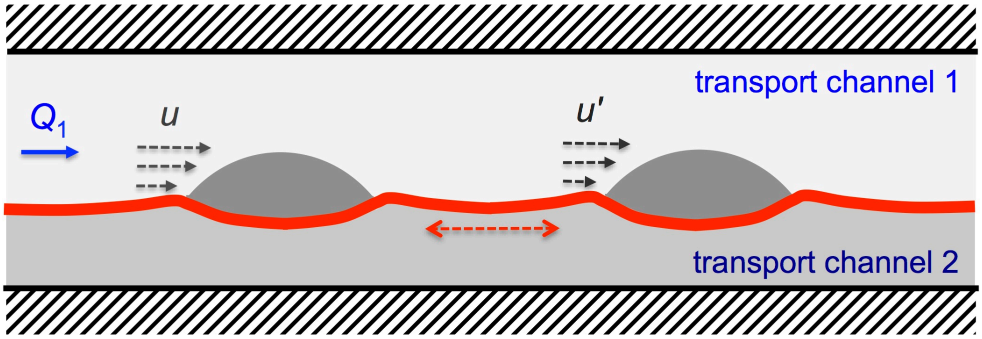

Figure 3: *Schematic of the motion of dragged droplets and their elastic and hydrodynamic interactions.*

In case of two dragged droplets moving in closer vicinity at a distance at which one droplet experiences the membrane deformation induced by the other droplet, the droplets will interact via the elastic membrane. This interaction will not only have a direct influence on the moving contact lines of the droplets but also on the local flow fields within transport channel 1 and the induced transport in channel 2. Moreover, this change of the droplet distance will lead to different conditions of the hydrodynamic drag on the two droplets ($u \neq u'$ in Fig. 3) as long as the distance is in the order of or smaller as the droplet radii, which culminates in a correlated motion of the droplets and correlated fluid transport.

<u>Work-package 2:</u> Continuous flow-induced instabilities of thin elastic membranes

Within work package 2, the general behavior of continuously flowing fluids separated by a thin elastic membrane will be investigated using the microfluidics platform developed in WP-1. Two fluids flowing with different velocities on either side of the thin elastic membrane are initiate local deformations owing to a net pressure difference at the membrane. These deformations change locally the two cross sections of the transport channels having a direct feedback on the local flow velocities. This temporal and spatial interplay between the flow velocities and the induced membrane deformations may evolve in an instability ("Kelvin-Helmholtz-like instability") as sketched in Fig. 4. However, the elasticity of the membrane will serve as a stabilizing component. Using our microfluidic setup, we are able to directly image spatial and temporal deformations of the membrane as well as local flow velocities by means of microscopy and PIV. We may expect the appearance of an instability by crossing a typical threshold, which can be controlled by the specific flow conditions for a given membrane system. In order to adjust the flow velocities in the two transport channels, we are using two different flow situations. In the first case, we are fixing the volumetric flow rates Q_1 and Q_2 (using syringe pumps) and in the second case the initial pressures p_1 and p_2 (using a pressure control setup) during the experiments. We expect differences in the onset of the deformations/instabilities as well as in the impact of the local deformations on the flow velocities holding either the volumetric flow rates or the local pressures constant.
In addition, we are able to analyze the effect of shear on the membrane from both sides. We define a membrane shear rate as $\tau_{M_{1,2}} = \eta_{1,2}(\partial u_{1,2}/\partial y)_{y=S_{1,2}}$, with $\eta_{1,2}$ the viscosity, $u_{1,2}$ the local velocity parallel to the membrane, y the distance from the membrane and $S_{1,2}$ the position

of membrane surface. As it can be seen, τ_M is dependent on the local velocity u (such as the local pressure p) and on the viscosity η of the fluids in the two transport channels, which can be straightforwardly characterized by using fluids of different η in our experiments. Starting our set of experiments with a PDMS membrane system, the impact of membrane properties, such as (switchable) wettability, surface structures, lateral structured elasticity (WP1) on the flow-induced deformations/instabilities and therefore on the transport properties of our microfluidic device will be analyzed. Moreover, these effects might be substantially amplified and modifies by applying electric field, which will be more deeply investigated in WP4.

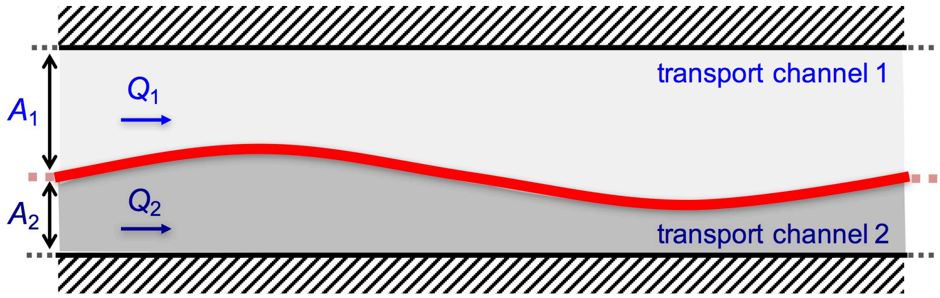

Figure 4: *Schematic representation of flow-induced deformation/instabilities of separating membranes within our microfluidic setup.*

<u>Work-package 3:</u> Coupling of droplets, bubbles and continuous media transport by thin elastic membranes

In this work package, the studies of moving droplets on a thin membrane (WP1) are combined with the investigations on flow-induced instabilities of separating membranes (WP2), in order to manipulate the crosstalk of the two transport channels via a flexible membrane to control the transport properties and routes within the whole microfluidic device. For example, employing droplets or bubbles flow in one of the transport channels, the moving deformations of the elastic membrane, due to the impact of the droplets or bubbles, will generate a fluid flow in the underlying transport channel (Fig. 5a). Moreover, the opposite scenario, continuous single-phase flow in one channel will generate a movement and flow in a droplets or bubbles channel, will be explored in detail as well.

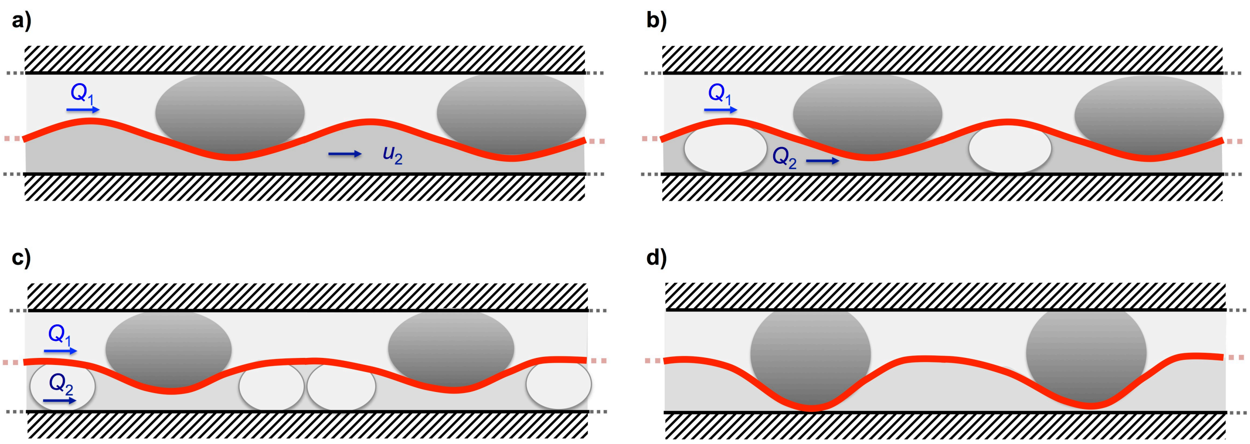

Figure 5: *Schematics of coupled flows in our microfluidics systems. a) Droplet flow in upper channel induces pumping of fluid adjacent channel. b) Anti-correlated droplet flow in adjacent channels. c) Flow pattern shaping in adjacent channels. d) Flexible obstacles block flow in adjacent channel.*

In all aforementioned setups, only one channel has a continuous inflow. Employing continuous (or periodic) inflows – either controlling the volumetric flow rates or the local pressures – in both transport channels introduce additional cross communications between adjacent channels. This crosstalk may lead to synchronized, correlated and anti-correlated flows (Fig. 5b) as well as more specific flow patterns, such as sketched in Fig. 5c, open the possibility to combine, merge or split droplets/bubbles in a highly dynamic and flexible way. Static and spatial periodic obstacles can be introduced by “parking” droplets at specific positions (Fig. 5d), due to e.g. strong pinning of the droplets at structured surfaces of the membrane and/or the channels walls. These “flexible” obstacles can be used to squeeze and possibly split droplets/bubbles or to block the flow in the adjacent channel and can be switched off by employing higher drag flow to overcome the pinning force of the droplets/bubbles. These effects might be dramatically amplified and modified by additionally using electric fields (see WP5). In WP3 and WP5, we will discover and develop models to describe the full beauty of the observed complex cross communications between transport channels via a thin membrane in order to generate to local pumping and pruning, correlation and synchronized flows and to shape flow and transport routes and patterns.

With WP1-WP3 providing the fundamental physics of how flow- and wetting-induced instabilities can be used to control liquid transport in segmented channels, exerting *local* control over these processes may be desirable when it comes to their applicability in microfluidic devices. WP4 and WP5 propose to use electric fields to interact with and control the processes outlined in WP1-WP3.

<u>Work-package 4:</u> Controlling membrane instabilities by electric fields

The principle to control membrane instabilities with electric fields is demonstrated in Fig. 6.

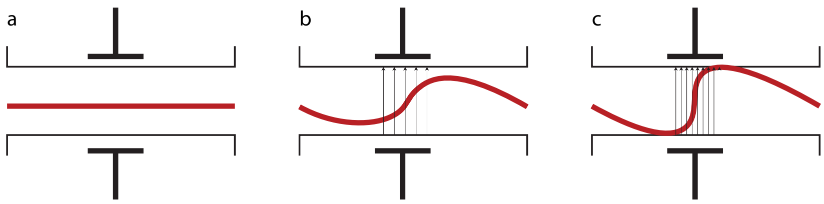

Figure 6: *Driven by the EHD instability in Fig. 1, the polarization of the planar membrane in (a) by an electric field in causes the elastic membrane to buckle (b). Both the instability onset and amplitude are controlled by the ratio of the applied electric field and the elastic modulus of the membrane (weak and strong fields in (b) and (c), respectively.*

The effect is based on the EHD instability described in Fig. 1. Translated to the membrane setup in WP1-WP3, the local application of an electric field across the membrane-separated channels will lead to the buckling of the membrane (Fig. 6). While the EHD instability of an interface between two fluids does not have a lower threshold electric field [7], an elastic medium introduces such a threshold, which is governed by the balance between electrostatic and elastic forces [21]. The presence of a threshold is useful because it enables a level of control that is absent in viscous fluids. In particular, beyond an on-off switch (Fig. 6 a vs. c), this force balance also controls the maximum amplitude of the instability (Fig. 6b). In a 3D system, the details of this instability will depend also on the channel geometry and sizes. It is evident from Fig. 6c that this effect can be used as channel valve, opening and closing either both channels, or, by introducing asymmetric channel width only one of them. Used at a channel junction, this can be used to sort particles into the different channels. The coupling strength of the electric field to the membrane can be adjusted though its dielectric contrast with the surrounding liquids, which can for example be increased by the incorporation of nanoparticles into the mebrane.

The first part of this WP will consist of the experimental characterization of electric field-triggered membrane instability in a static conformation (without flow), and the development of a theoretical model describing this instability.

Work in thus WP will then continue to examine the coupling of EHD and the Kelvin–Helmholtz-like instability of Fig. 4. In the absence of an electric field, this instability is triggered by the flow velocity difference in the two channels, the threshold of which is given by the Richardson number. The application of a weak electric field will modify this threshold, as illustrated in Fig. 7, where both the difference in flow velocities and the applied electric field are too weak to cause an instability, but their combination triggers and controls an instability.

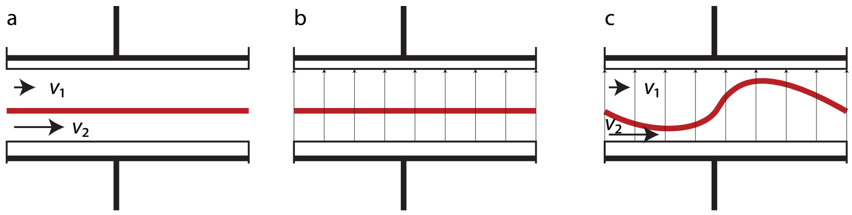

Figure 7: *A membrane, which is stable both with respect to a velocity difference (a) and in a weak electric field (b) exhibits and instability when both are simultaneously applied. This allows the spatial and temporal control of the instability by the electric field.*

This is interesting for several reasons. (1) In terms of liquid transport, the use of an electric field extends the parameter rage of this instability (i.e. it will be possible to "switch it on" for given flow properties, (2) an independent control over the oscillation amplitude is obtained, and (3) local control of the instability is attained, allowing to trigger the instability in only part of the channel network.

The main task in this work package consists of the experimental characterization of EHD and flow instabilities and their theoretical description. The same experimental techniques as in WP1 - WP3 will be employed.

<u>Work-package 5:</u> Control of segmented flow by electric fields

With the fundamental understanding of WP3 and WP4 in place we will now combine these two by adding liquid drops and bubbles into the channels. This renders the system more complex, since the drops alter not only the flow properties by also the spatial and temporal distribution of dielectric constants in the system, which modifies the EHD instabilities in a marked fashion. This work-package therefore splits into two different tasks. In the first task, we will investigate in which way the application of an electric field (along the entirety of the channels) will modify the segmented flow in individual and coupled channels, and in how far electric fields can be used to exert control over these transport phenomena.

In a second task, we will employ local fields with the aim to either locally trigger instabilities and investigate the transport of drops in these. Three examples are shown in Fig. 8. In Fig. 8a, the droplet induced instability is shown, where, because of the dielectric mismatch, a local instability is triggered only by the presence of a drop. In Fig. 8b, the local triggering of an instability acts as a "peristatic pump" for droplet transport. In Fig. 8c, the drops themselves are manipulated by the instability, by segmenting a large drop into two. The combination of these two should enable to trigger and control the mixing of two liquids within the channels.

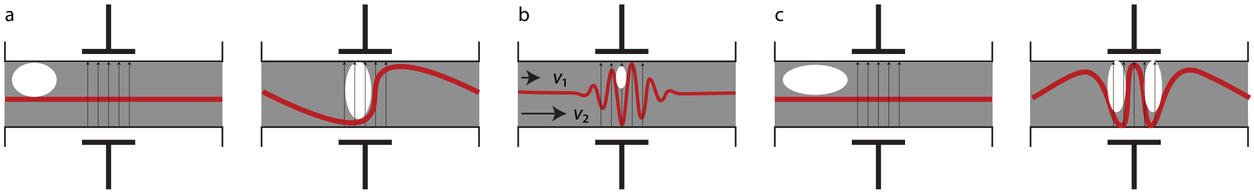

Figure 8: *Control of segmented flow by a locally applied electric field. (a) The flow of a dielectric drop into the electric field triggers the instability, arresting the transport. The local triggering of a membrane instability (see Fig. 7) can act as a peristaltic pump. (c) Applying an electric field across a laterally extended dielectric drop splits the drop into two.*

Where do we want go?

Based on these proposed studies, we should be able to develop more elaborated fluidic networks for e.g. innovative applications in soft robotics. These fluid networks have an incredible potential for material transport as well as information transport with information processing capabilities owing to the inherent correlation, synchronization and feedback mechanisms.

2.4 Data handling

We plan to publish all results of this project in peer-reviewed journals in order to make them available to the scientific community. All experimental and simulation data will be archived in accordance to DFG regulations.

2.5 Other information

Not applicable.

2.6 Descriptions of proposed investigations involving experiments on humans, human materials or animals as well as dual use research of concern

Not applicable.

2.7 Information on scientific and financial involvement of international cooperation partners

The project work by the Steiner group will be carried out at the University of Fribourg and AMI in Fribourg, Switzerland. The project will be conducted as part of the agreement between the DFG and the SNF. Prof. Steiner has applied for funding covering the staff (one PhD student), the consumables, project-related publication expenses as well as the travelling expenses (see 4).

2.8 Information on scientific cooperation within SPP 2171

Within the framework of the SPP 2171, we plan to collaborate with the groups of Rainer Jordan, Macromolecular Chemistry, TU Dresden, on “polymer brushes decorated flexible membranes and walls” and with Ahmed Mourran, DWI, Leibniz Institute for Interactive Materials, Aachen on “membranes of switchable elasticity”. The planned collaborative work is outlined in WP1. Moreover, we plan to cooperate with Andreas Heuer, Westfälische Wilhelms-Universität Münster, and Uwe Thiele, Westfälische Wilhelms-Universität Münster, on the theory of elasto-capillarity and electro-hydrodynamics of moving droplets on elastic membranes.

Beyond these planned collaborations, we would be glad if our studies on membrane-mediated adaptive droplets transport through coupled flow paths would inspire further researchers within the SPP and further fruitful collaborations will arise.

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4 Requested modules/funds

4.1 Basic Module

4.1.1 Funding for Staff

<u>Staff requested by Pfohl, Thomas:</u>
1 PhD student for three years - 75% position E13

<u>Staff requested by Steiner, Ullrich:</u>
1 PhD student for three years. SNF salary

Total 4.1.1	Group of Pfohl, Thomas	One PhD for three years	€ 145’125
	Group of Steiner, Ullrich	One PhD for three years	CHF 173’288

We apply for one PhD student (Pfohl group) carrying out mainly the investigations of WP1-3 and one PhD student (Steiner group) working mainly on the investigations of WP4-5. However, the PhD students will work in close collaboration and will under constant exchange.

4.1.2 Direct Project Costs

4.1.2.1 Equipment up to Euro 10,000, Software and Consumables

<u>Pfohl, Thomas</u>

2 x microfluidic pumps neMESYS von Cetoni 2 x € 4’000 € 8’000

Consumable per annum, based on prior experience, including lithography masks, microfluidic masters, chemicals, clean room chemicals, silicon wafers, photoresists, sylgard, tubing, colloid particles, fluorescence dyes:
per annum: € 15’000
for the application period (36 month): **€ 45’000**

<u>Steiner, Ullrich</u>

Consumable per annum, based on prior experience, including chemicals, gases, substrates, lithographic masks, cleanroom user fees, user fees for electron microscopy and FIB/SEM:

per annum: CHF 15’000
for the application period (36 month): **CHF 45’000**

Total	Group of Pfohl, Thomas	€ 53’000
	Group of Steiner, Ullrich	CHF 45’000

4.1.2.2 Travel Expenses

In order to facilitate the close cooperation between the two working groups, a constant exchange of the PhD students (2 ½ h train distance) from days to several weeks depending on the project-specific matters and continuous knowledge transfer between the groups are of paramount importance for the success of this project.
Moreover, for a successful cooperation within the SPP 2171 and the success of the SPP 2171, workshops (once a year), advanced school (1st year) and an international conference (3rd year) for the employed researchers and PIs are planned. In addition, a workshop of the PhD-candidates is planned in the 2nd year.

Pfohl, Thomas
per year: € 5’000
for the application period (36 month): € 15’000

Steiner, Ullrich
per year: CHF 5’000
for the application period (36 month): CHF 15’000

4.1.2.3 Visiting Researchers (excluding Mercator Fellows)

Not applicable

4.1.2.4 Expenses for Laboratory Animals

Not applicable

4.1.2.5 Other Costs

None

4.1.2.6 Project-related publication expenses

Pfohl, Thomas
Costs related to open access publishing € 2’250

Steiner, Ullrich
Cost related to data management and data archiving (open data) CHF 8’000

4.1.3 Instrumentation

4.1.3.1 Equipment exceeding Euro 10,000

4.1.3.2 Major Instrumentation exceeding Euro 50,000

4.2 Module Temporary Position for Principle Investigator

4.3 Module Replacement Funding

4.4 Module Temporary Clinician Substitute

Not applicable.

4.5 Module Mercator Fellows

Not applicable.

4.6 Module Workshop Funding

Not applicable.

4.7 Module Public Relations Funding

Not applicable.

5 Project requirements

5.1 Employment status information

Pfohl, Thomas	academic researcher and group leader, permanent position
Steiner, Ullrich	Professor at the Adolphe Merkle Institute (University of Fribourg)

5.2 First-time proposal data

5.3 Composition of the project group

Freiburg:
Dr. Thomas Pfohl has joined the Experimental Polymer Physics Lab (chair. Prof. Dr. Günter Reiter) as an academic researcher and group leader in 2017. Prof. Reiter fully supports this proposed project. Moreover, owing to his excellent expertise in soft matter, wetting and dewetting and polymer physics, Prof. Reiter may also participate in scientific discussions concerning this proposed project. The secretary of the Experimental Polymer Physics Lab, Susanne Brendl, will help with administrative procedures and technical support will be in parts provided by technical personnel of the Lab.

Thomas Pfohl
N.N., Master student

Fribourg:
Prof. Ullrich Steiner heads the Soft Matter Physics group at the Adolphe Merkle Institute (AMI) and serves as its vice-director. The Soft Matter Physics group is currently subdivided into five research thrusts, encompassing perovskite photovoltaics, Li-ion battery research, order-disorder interplay in photonic materials, self-assembled metamaterials, and bioinspired materials. Prof.

Steiner has a longstanding interest in interfacial phenomena in soft matter, which is the topic of this SPP. The infrastructure and personnel of the AMI will be partially available for this project, including secretarial and technical support, as well as help from AMI's chemistry and biology groups, whenever needed. In a collaboration with the University of Cambridge, one of Prof. Steiner's PhD student's, Johannes Bergmann, has recently set-up microfluidic experiments for a bioinspired project, within the Horizon 2020 ITN "PlaMatSu". Johannes' expertise and support will be available for this project.

5.4 Cooperation with other researchers

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

Within the framework of the DFG priority programme SPP 2171 the following researchers have agreed to cooperate:

Andreas Heuer, Westfälische Wilhelms-Universität Münster: Theory impact of moving three phase contact line on elastic membranes.

Rainer Jordan, Macromolecular Chemistry, TU Dresden: Polymer brushes decorated flexible membranes and walls.

Ahmed Mourran, DWI, Leibniz Institute for Interactive Materials, Aachen: Membranes of switchable elasticity.

Prof. Uwe Thiele, University of Münster

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

T. Pfohl:

U. Jenal, W. Meier, B. Müller, C. Palivan (University of Basel, CH), V. Zaburdaev (University of Erlangen, D), N. Bruns (AMI Fribourg, CH), A. Zumbuehl (University of Fribourg, CH), Y. Ekinci (PSI Villigen, CH), I. Tolic (RBI Zagreb, CRO),

U. Steiner:

U. Wiesner (Cornell), J. J. Baumberg, R. H. Friend, N. C. Greenham, S. Vignolini, B. Glover, C. J. Howe, C. Ducati, S. Hofmann, N. A. Fleck (Cambridge, UK), H. Snaith (Oxford), P. Vukusic (Exeter), O. Hess (Imperial College London), S. Guldin (UCL London), D. S. Wiersma (LENS, Italy), M. Kolle (MIT), H. Miguez (Seville), B. Ehrler (AMOF, NL), Y. Yanzov (Heidelberg), M. Grätzel, A. Hagfeld (EPFL), A. Abate (Helmholtz-Zentrum Berlin), G. E. Schröder-Turk (Murdoch, Australia), C. Neto (Sydney), A. Sepe (Shanghai), D. G. Stavenga (Groningen), M. Stefik (Columbia, USA), S. Hüttner (Bayreuth), B. Grobéty, K. Fromm (Fribourg), G. Reiter (Freiburg)

5.5 Scientific equipment

List larger instruments that will be available to you for the project. These may include large computer facilities if computing capacity will be needed.

Freiburg:

- Several optical microscopes (Olympus, Nikon, Zeiss) for polarization microscopy, DIC-microscopy, fluorescence microscopy, microscopically photoluminescence measurements equipped with color cameras, a high sensitive fluorescence camera, and spectrometer.
- custom-built microsyringe pumps for microfluidic experiments
- AFM and STM

- Chemical wet lab, access to clean room facilities

Fribourg:

- Various optical microscopy techniques, including DIC, polarization contrast, fluorescence confocal microscopy, SERS, etc.
- Scanning and cryo-transmission electron microscopy, FIB/SEM dual beam, AFM
- E-beam lithography, various evaporation and sputtering techniques
- Dedicated clean room for soft-lithography techniques, clean-room access at EPFL for methods that are not locally available
- Small and wide-angle x-ray scattering, including grazing incidence
- 3-4 beam times per year at synchrotrons for structural analysis
- 3-D printing techniques
- Custom built microfluidic setup
- Dry and wet-labs for sample preparation
- Glove-box setups for device preparation

5.6 Project-relevant cooperation with commercial enterprises

Not applicable.

5.7 Project-relevant participation in commercial enterprises

Not applicable.

6 Additional information

If applicable, please list proposals requesting major instrumentation and/or those previously submitted to a third party here.