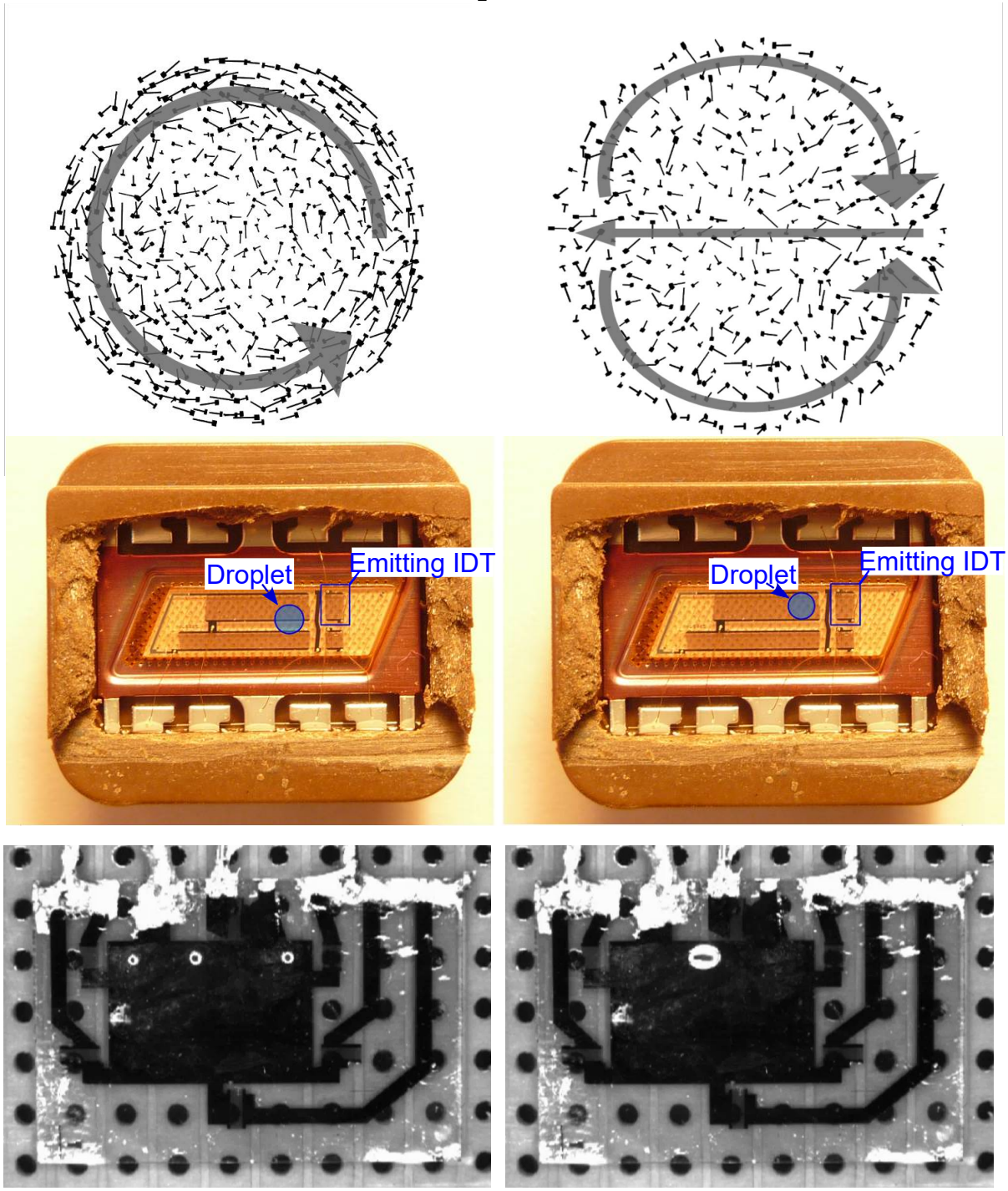
Acoustic Microfluidics with tiny Droplets



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1. Abstract

In our project, we develop and construct control units for microfluidics. Microfluidics, has become an important field of modern research and allows the construction of entire miniaturized laboratories, so-called Labs-on-a-Chip. Furthermore microfluidics promises a great reduction in costs and risk potential. Since common mechanical control units cannot be used at the micrometer scale, alternative control methods are being researched. Using computer simulations and experiments we developed an tested a control unit able to create flows inside droplets. Using our invention, we are able to mix multiple chemicals, concentrate solids in the middle the droplet and move them precisely over the piezos surface.

2. Introduction & Objectives

The benefits of Lab-on-a-Chip systems are mentioned in a lot of scientific papers. In addition to reducing the laboratory size, a Lab-on-a-Chip also lowers the volume of required materials and chemicals, reducing both risks and costs associated with their supply, use and disposal. The production of Labs-on-a-Chip is a major technical problem as the physical laws of our familiar surroundings can't be transferred directly onto the microcosm. Strong electrical forces and an increase in liquid's viscosity are results of the drastically rising surface to volume ratio. Therefore Labs-on-a-Chip are not yet ready for mass production. In particular, small and nevertheless powerful control units are needed. In our opinion, piezoelectrically generated surface waves provide a promising route towards control of droplets. Our project “Acoustic Microfluidics with tiny Droplets“ focuses on transport of droplets and creation of flows inside them. We are able to mix microliter scale volumes of liquids and to concentrate solids within the droplets.

3. Theory

To get a fundamental understanding of acoustic waves, it is important to distinguish between waves propagating inside a medium or on the surface of it. A wave inside a solid can either be a longitudinal wave or a transverse wave. Longitudinal- and transverse waves differ in their direction of oscillation relative to their propagation direction. While the oscillators of a longitudinal wave swing in the direction of wave propagation, the transverse wave oscillates perpendicularly to the direction of propagation. In contrast to longitudinal waves, transverse waves can have a polarization defined by their direction of oscillation. For example, a distinction is made between horizontally and vertically polarized transverse waves.
Surface waves consist of a combination of transverse oscillations perpendicular to the surface and longitudinal oscillations which is also called a “Rayleigh wave”. The transverse

wave only propagates in a thin surface layer of the medium. Due to the lower propagation speed of transverse waves in comparison to longitudinal waves, transverse waves are slower in penetrating the medium.

Particularly important for our investigations is the interaction between surface waves and their surroundings. Since every wave can be refracted, surface waves can also deliver energy to a body by being refracted into the body. If the delivered energy and momentum is high enough, the wave can accelerate the body in the direction of wave propagation and act as a pump e. g. for liquids. In case of a liquid droplet, a surface waves that does not have enough energy to move the droplet can create flows inside the droplet. Through controlled adjustment of the created flows, the droplet can be re-purposed for mixing chemicals.

Two counter-propagating surface waves can be used to create a standing wave. In contrast to normal (propagating) waves, it contains nodes and anti-nodes. While nodes have no moving oscillators, oscillators at anti-nodes have the most energy and therefore are moving the most. The nodes have a fixed position, i.e. they maintain their position over time, which also explains the term “standing wave”. Therefore a standing wave has different features in terms of interactions with other bodies. Bodies interacting with a standing wave experience a force, pulling them towards the nodes and holding them there, because of the energy laying inside the anti-nodes. Manipulation of the frequency or phase of a standing wave allows the position of the bodies to be modified by moving the nodes, the bodies are being held in. Therefore, controlled modification of the wave/nodes allows the movement of bodies.

For our experiments, we need said surface waves and therefore precise surface wave generation is the main experimental challenge of our project. Piezoelectrics can provide a solution. They are crystalline materials, which can act as an electromechanical transducer due to their special structure. Mechanical deformation of a piezo element, creates a dipole inside the element on which a voltage is measurable. Reversing this process, it is also possible to apply a voltage to the piezo element which will result in the element deforming mechanically.

To generate a surface wave, an interdigital structure, short IDT for “Inter Digital Transducer”, has to be applied to the piezo element. Said structures consist of two comb-like metal conductors interleaved without touching each other. If an AC voltage is applied to the IDTs, the structures create a surface wave. The ideal wavelength matches the distance between two equally polarized fingers of a structure. When a frequency corresponding to this wavelength is applied, every single finger will reinforce the generated wave. Due to the IDTs having a certain width, they are not acting as a point source and thus have special features. Right after the generation, the wave propagates in the so-called “Fresnel region” in a straight line until entering the so-called “Fraunhofer region”. Only after entering this area, the wave propagates as if it originated from a point source. Depending on the cut, piezo crystals can have different characteristics. The cut we need has to allow us to generate surface waves along two axes, to be able to create a two dimensional standing wave field. To achieve that, we use a 128° Y-cut of lithium niobate.

4. Simulations

4.1 Simulations of Wave-controlled Movements of Droplets and Solids

To test our theoretical knowledge, for our experiments and, most importantly, to get a physical explanation for the occurring phenomena, we simulated some of our experiments. Computer simulations offer huge advantages to us in comparison to experiments, since we can influence and analyze the behavior of single particles to arrive at conclusions the effect of various experimental parameters. There are many different variants of computer simulations which have their own advantages and disadvantages. The simulation variant we need has to be able to (i) simulate different kinds of liquids at the same time, (ii) allow us to apply a wave field, (iii) let us observe the occurring reactions and (iv) support the addition of solids where required by the experiment. For our requirements, a N-body-simulation is the best choice. One benefit of N-body-simulations is the ability to simulate interactions between a large number of particles, for example atoms and molecules, simultaneously. Further it is also possible to measure the position and velocity of a particle or change it's characteristics at any time. Using this simulation variant, we are able to explain the behavior of particles more accurately than using experiments, as long as the occurring and interacting forces are known. However, two major disadvantages are the high amount of computing power required and the long calculating times. Unlike most other simulations, we use the GPU, or graphics card, instead of the CPU, to work around the problem of high computing power requirements. Single processes may be clocked slower, but due to graphics cards having several thousands of processors, if used in the right way, it can speed up the calculation process dramatically. Running N-body-simulations on the GPU enables us to simulate complex physical systems without a supercomputer.the effect of various experimental parameters. There are many different variants of computer simulations which have their own advantages and disadvantages. The simulation variant we need has to be able to (i) simulate different kinds of liquids at the same time, (ii) allow us to apply a wave field, (iii) let us observe the occurring reactions and (iv) support the addition of solids where required by the experiment. For our requirements, a N-body-simulation is the best choice. One benefit of N-body-simulations is the ability to simulate interactions between a large number of particles, for example atoms and molecules, simultaneously. Further it is also possible to measure the position and velocity of a particle or change it's characteristics at any time. Using this simulation variant, we are able to explain the behavior of particles more accurately than using experiments, as long as the occurring and interacting forces are known. However, two major disadvantages are the high amount of computing power required and the long calculating times. Unlike most other simulations, we use the GPU, or graphics card, instead of the CPU, to work around the problem of high computing power requirements. Single processes may be clocked slower, but due to graphics cards having several thousands of processors, if used in the right way, it can speed up the calculation process dramatically. Running N-body-simulations on the GPU enables us to simulate complex physical systems without a supercomputer.

To be able to perform N-body-simulations with a normal computer, we need a computer program that can run such simulations on the graphics card. Since no simulation program we know is capable of doing this kind of work, the only choice we had was writing our own program. Controlling the graphics card is bound to be a lot of work. Using the programming language C++ in combination with CUDA, we were still able to write a program capable of controlling the GPU. This grants us the possibility to place many particles in a room and to set their characteristics as well as their interactions with other particles. Using a graphical output, it is possible to watch the simulation in real time.

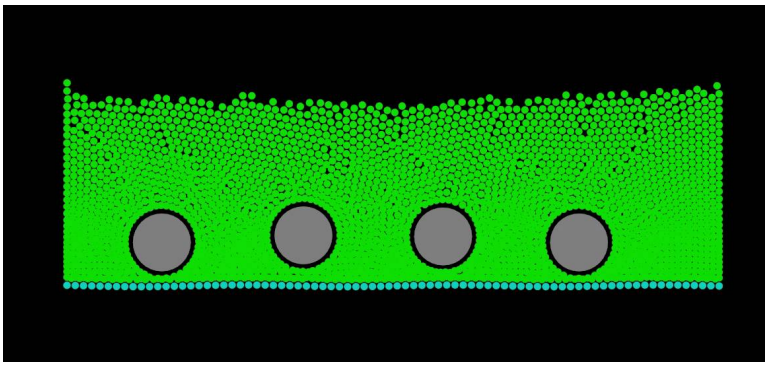

Fig. 4.1: Solids at the beginning of the simulation equally spread

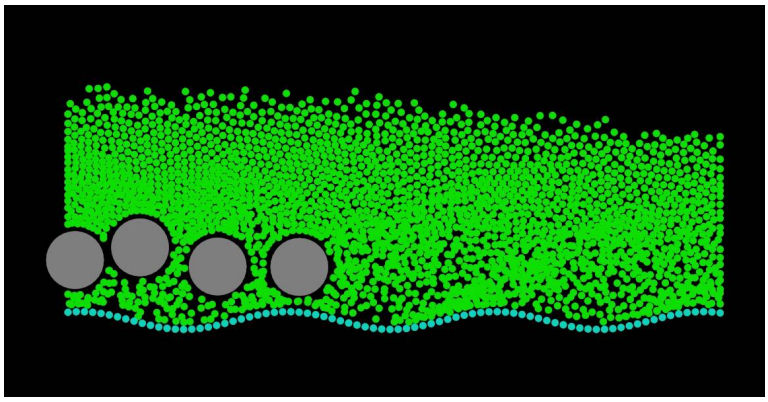

Fig. 4.2: After some time, the surface wave, coming from the right of the piezo, moves the solid bodies to the left.

In our first simulation, we wanted to see the behavior of bodies inside a liquid, while a wave is applied to the liquid. Following our theory, the wave should be able, to accelerate the bodies in it's direction of propagation, as long as the wave is powerful enough. If this works out, we can find out both why the bodies are moving and how energy is transmitted between waves and bodies. In this simu-lation, the wave is modeled as a layer of particles, which also represents the top layer of our piezo element. With the help of the simple wave equation $y_1 = y_0 \sin(k x + t \omega)$ we simulate a sine wave of amplitude y_0, wave number k and angular frequency ω propagating in the -x direction on the surface. To make interactions visible, we put a liquid medium on the surface. After putting multiple bodies into the liquid medium, the bodies should move in the -x direction Right after the start of this simulation, an interesting effect was visible. A part of the energy trans-ported by the wave was absorbed by the liquid medium and generated a longitudinal wave. However, this wave did not propagate along the surface but it propagated with a certain angle to the surface. Therefore the surface wave was partly refracted into

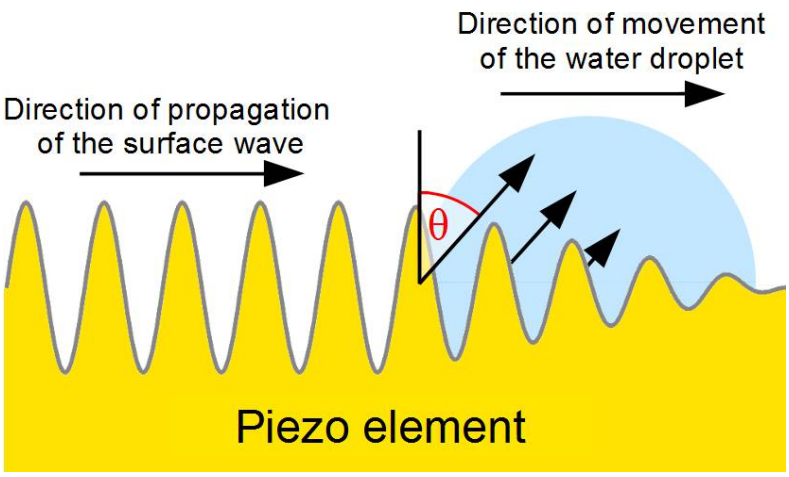

Fig. 4.3: Schematic drawing of the droplet movement

the liquid. A description of this phenomenon is made by the Snell–Descartes law $\theta = \arcsin(c_s / c_1)$, where c_1 and c_s are the propagation speeds inside the liquid and the solid. Something similar happens to the bodies as well, with the difference that the bodies can get energy from both the piezo surface and the longitudinal wave inside the liquid medium, supporting the force affecting the bodies and moving them along the wave. To verify the theory, we modified the simulation a little and repeated it. Now we removed the liquid medium and the solid bodies, switched the propagation direction of the surface wave and only put a water droplet on the piezo surface. As before we saw the droplet moving in the propagation direction of the surface wave supporting our theory discussed in the previous section.

In the next step, we wanted to simulate the interactions between solid bodies and a standing wave field. Therefore we use the wave equation $y_1 = y_0 2\sin(kx)\cos(t\omega)$ to describe the standing surface wave. Again we use a liquid medium in which solid bodies are placed. Following our theory, the bodies should move to the nodes and stick to them. After some time this was also visible in our simulation. And again it is interesting, how the behavior of the bodies can be explained. As before, the wave loses a part of it's energy to the liquid, creating longitudinal waves inside it, propagating circularly away from the source. Due to this, supersonic jet pressures are created at the anti-nodes. This phenomenon occurs in liquid and gaseous wave media permeated by a longitudinal wave. The

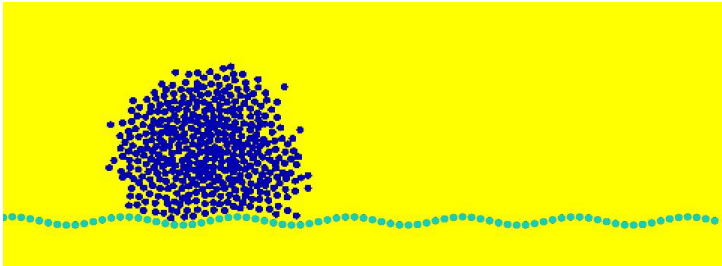

Fig. 4.4: Waterdroplet at the beginning of the simulation at the left end of the piezo element

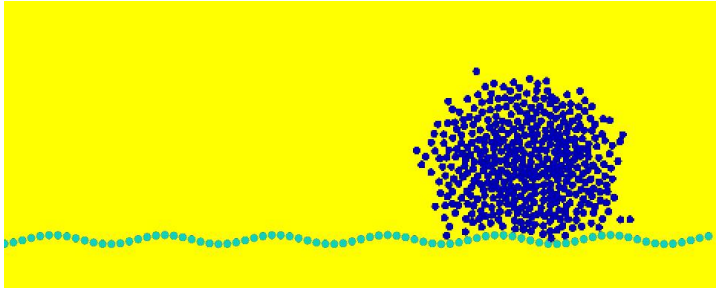

Fig. 4.5: Water droplet, after some time, at the right end of the piezo element

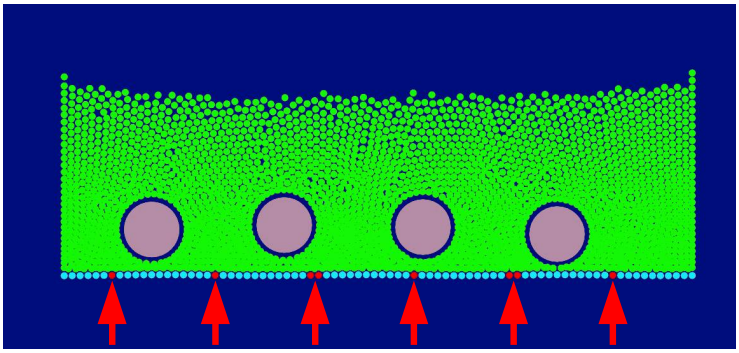

Fig. 4.6: Simulation of the standing wave field at the beginning, with solid bodies placed randomly. The red arrows mark the nodes.

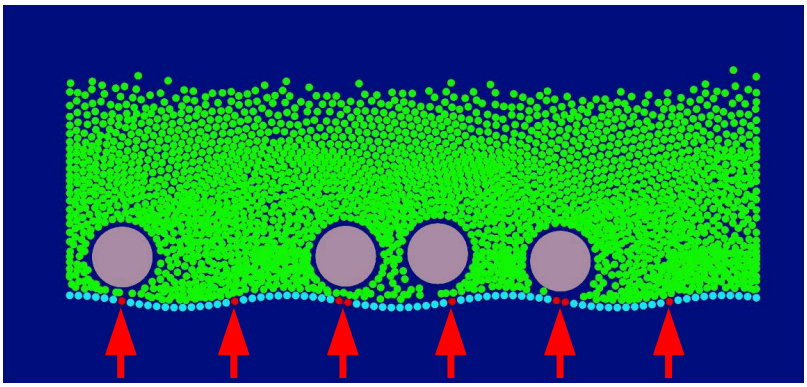

Abb. 4.7: Simulation of a standing wave field, after the solid bodies have moved to the nodes

longitudinal wave makes the particles of the medium oscillate forwards and backwards in the direction the longitudinal wave is propagating in. On high oscillation frequencies are the particles unable to get back to their prior position fast enough so other particles take those spots. Therefore the particles affected by the wave, are permanently displaced, creating a pressure pointing away from the source affecting the bodies. Since there is no supersonic jet pressure at the nodes, the bodies placed around them, are affected by a force, pulling them towards and holding them at the nodes. This force can be described by $F = \frac{2\alpha I}{c}$, where α equals the absorption coefficient, *I* the intensity of the wave, *c* the speed of sound inside the wave medium and *F* the resulting force. Indirectly the equation also shows, that the supersonic jet pressure decreases with increasing frequency. This is due to the absorption coefficient as well as the intensity decreasing with rising frequency or high frequency waves being absorbed faster and high intensities being difficult to generate at high frequencies. However, this effect has little impact when using typical frequencies used to control piezo elements.

4.2 Simulations of Flows inside a Droplet

So far, we only simulated the effect of surface waves on the movement of entire droplets. However, our theory says that waves not powerful enough to accelerate droplets, should be able to generate flows inside droplets. Therefore we wanted to simulate those flows as well to get a better understanding of their effect on the droplet and how we can use them effectively. Since we already knew the forces affecting a droplet when it interacts with a surface wave from the previous simulations, we were able to simply remove the simulation of the piezo surface without a loss in the simulations accuracy. In this simulation, we filled a room, shaped like a droplet on the surface of a piezo element, with a liquid to represent the droplet. The spatial force distribution associated with the interaction of a surface wave with particles within the droplet is calculated in advance. We performed this in simulation three dimensions to be able to simulate more complicated flows inside the droplet. We also payed attention to the relative position of the droplet to the IDTs, because we were expecting different results depending on it. At first, we simulated the case, where only one half of the droplet

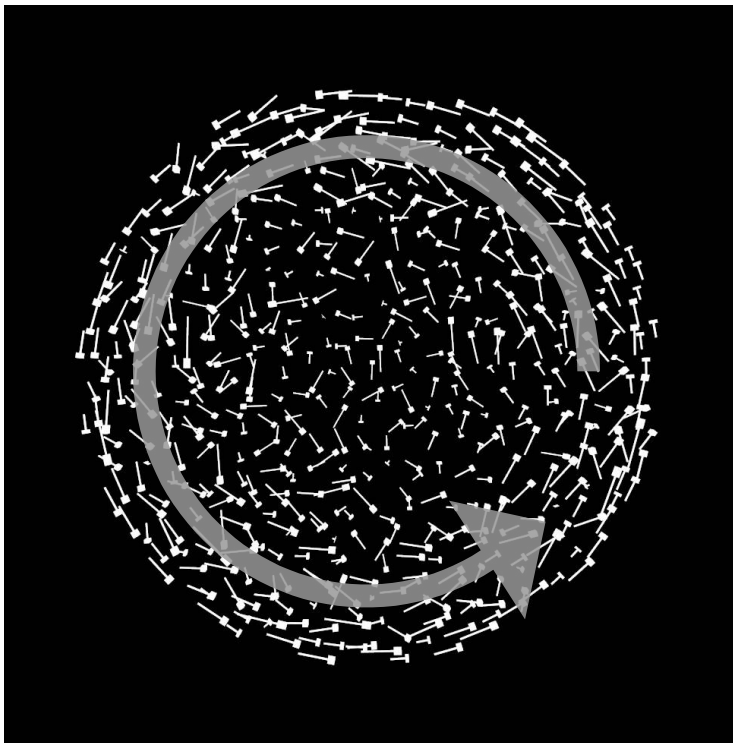

Fig. 4.8: Circular flows within a droplet viewed from the top. The movement direction of single particles is presented by small arows, while the thick end equals the tip of the arrow.

interacts with the surface wave. As described by our theory, we were expecting a circular flow inside the droplet. After some time, the simulation showed this exact case, confirming our theory. Further, the simulation showed an interesting effect which could be really useful in microfluidics. The simulation showed a pressure disparity. In this case, the pressure, i.e. the density of particles inside the droplet, was rising with increasing distance from the droplet's center. As well, the flow only occurred in a small ring at the edge of the droplet, while no clear flows were visible in its center. Therefore, denser objects like solids should gather in the droplet's center. This leads to a pressure gradient pointing away from the center of the droplet, which pushes denser objects towards the middle, as well as a difference in speed between the middle section and the edge section of the droplet, which generates shear forces affecting any solids. Therefore, this kind of flow is ideal for concentrating solid components.

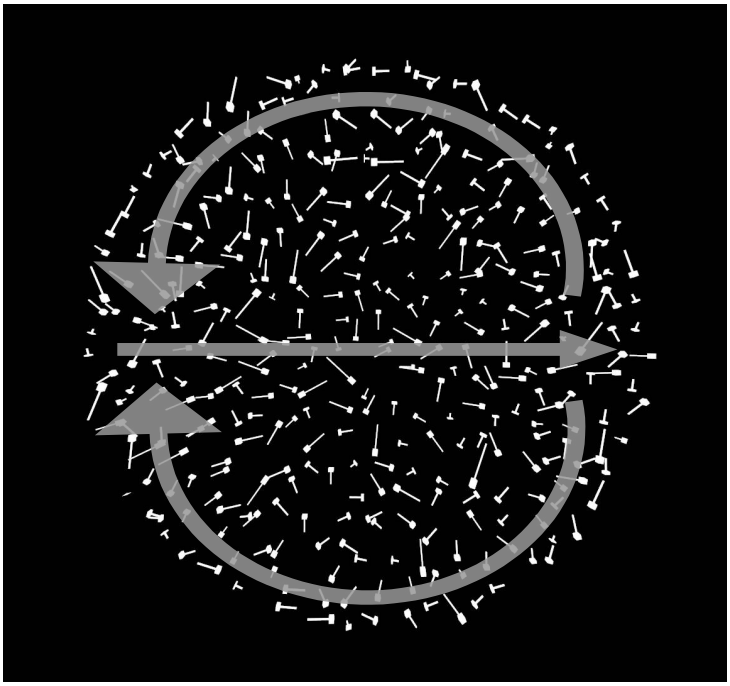

Fig. 4.9: A rightward facing central stream within a droplet

The second simulation deals with the situation of a droplet interacting with a surface wave on its entire width. We expect a more complicated flow, since two annular flows are colliding. After some time, the simulation showed a central stream in the middle of the droplet, splitting up into two streams at the edge of the droplet and flowing back along the droplet's surface to the start. Using two streams from different parts of the droplet, it is possible to use them for mixing chemicals.

5. Experiments with Homemade Devices

In our first experiment, we wanted to generate a standing wave field to precisely control and move droplets. However, this involves a lot of effort, since we have to apply a specific conductor pattern to the piezo elements, allowing us the generation of the wave field in the first place. This pattern would consist of two opposite positioned IDTs, which can be controlled independently from each other. As the company “Johnson Matthew” kindly provided us with said piezo elements, we could focus on applying the IDT pattern. The precise application of the patterns is extremely important, because the distance between the two IDT fields has to be an integer multiple of the wavelength corresponding to the applied frequency. As well, the distance between two fingers in one IDT field has to be consistent for this experiment to work. Deviations are hard to correct so precision is required. Since the piezo elements were already coated in gold, our first idea was to vaporize the gold layer in certain areas by applying a voltage. This only worked because

the layer of gold was very thin. We used a function plotter attached to an Arduino, which was programmed to move the plotter over the surface of the piezo following the pattern of our IDTs. We attached a needle to the plotter, using its tip for vaporizing the gold layer. Like this, we were able to transfer the IDT pattern onto the piezo element. Unfortunately, this process did not allow enough control leaving the piezo useless for our experiments.

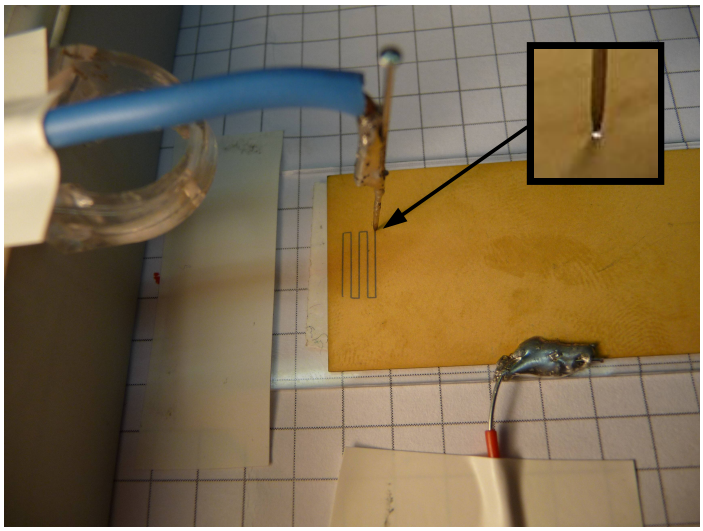

Fig. 5.1: Production of a piezo element using a function plotter

A swap in technology was necessary, so we orientated on the method of photolithography which allows a more precise creation of conductor tracks and is also used in semiconductor technology. First, a metal layer is applied to the piezo material, which is later used as conductor track. Then, a layer of positive or negative photoresist is applied on top of the metal. In the next step, the exposure, UV-light illuminates the photoresist through an exposure mask. The mask absorbs the light in specific places, preventing these places from being illuminated by the UV-light. When being exposed to UV-light, the photoresist changes its chemical structure. How its structure changes depends on the photoresist. While positive photoresist turns soft in illuminated spots, negative photoresist is soft until being illuminated. When developing solution is added, the soft photoresist is removed, transferring the mask onto the metal and revealing the metal on the soft areas. Using etchant, metal that is not protected by photoresist is removed. After cleaning the metal from photo resist remains, the mask is visible as conductor tracks on the metal.

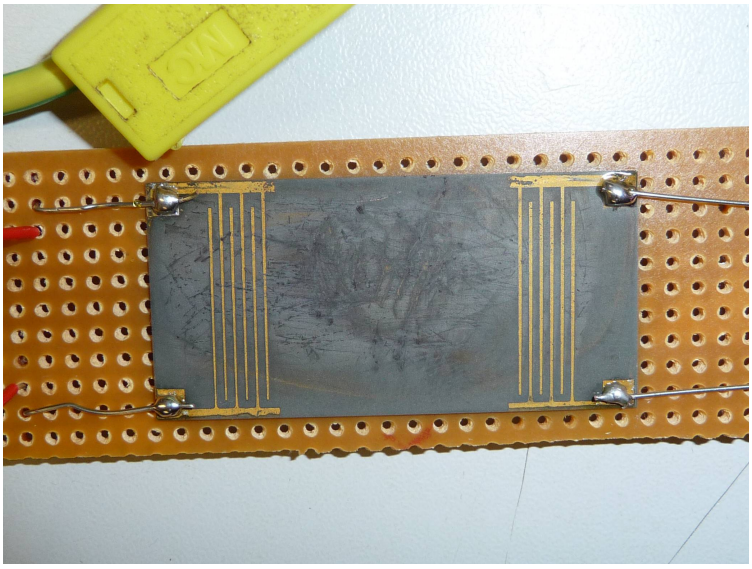

Fig. 5.2: Through photolithography produced layout

This process works fine for millimeter scale structures, but becomes more and more difficult without special equipment as the required feature size becomes smaller. This problem occurred to us since small conductor tracks of about 25 micrometer width are necessary for our experiments. Further, it was not possible for us to remove the gold layer from our piezo elements using an etchant. After multiple tests, we found out that the layer of gold could be removed by adding concentrated oxygen. With the help of electrolysis of water, we could develop pure oxygen straight at the gold layer. The areas covered by photoresist remained as they simply could not touch the oxygen. Since this method of

removing the gold layer was really aggressive, the manufacturing process was still difficult. After further tests, with changes in exposure time, we managed to manufacture a piezo element with two IDTs.

Through applying an AC voltage to the IDTs, we should be able to generate surface waves and create a standing wave field. To test the functionality of the IDTs beforehand, we applied an AC voltage with an audible frequency. Since both IDTs generated a roughly equally loud sound, we expected them to be able to generate a standing wave field. Further, we put a droplet of water placed in oil, to reduce the surface friction, on the piezo. If a standing wave field developed, the water droplet in oil should move to one of the nodes of the field. But after several experiments, this was not visible. Even running through all frequencies, that the droplet should react to, did not result in any noticeable movement. The applied control voltage of ±20 volts, should have been enough to at least create flows inside the droplet. Therefore, we assumed that the problem had another origin. The material our piezo element consists of is a piezoceramic. Those do have a coupling constant or a good efficiency, but as we found out, they are comparably bad at transmitting waves, due to their microscopical structure. Piezocrystals on the other hand do not have this problem.

After some research, we decided to buy a lithium niobate ($LiBbO_3$) wafer. A lithium niobate wafer with a 128° Y-cut would allow us to create a two-dimensional standing wave field using the right layout. They can be used with comparably high voltages to compensate their small coupling constant of only 0.055 or 5.5%. They are also affordable as they are common in industry.

Our first problem was how to coat the wafer. To be able to transfer our IDT layout onto the wafer using photo lithography, we need a layer of metal on it. Since we wanted to do as much work as we could ourselves, we tested several approaches. First we tried using conductive silver paste and copper spray to coat the wafer. This worked, but we were unable to transfer our layout onto the wafer using photolithography, because the silver was not etchable and the copper spray separated from the wafer while applying the photoresist. Therefore, we had the idea to use the metal indium, which has a low melting temperature of about 156°C (312.8°F) and sticks well to glacial surfaces like our wafer. From initial tests on slides, we knew that we could create very thin and easily etchable indium layers at a temperature of 300°C (572°F). When we tried coating one of our wafer with indium, we heated it up too quickly, causing strong static charges and therefore strong mechanical tensions were created inside the piezo due to the pyroelectrical effect, which lead to

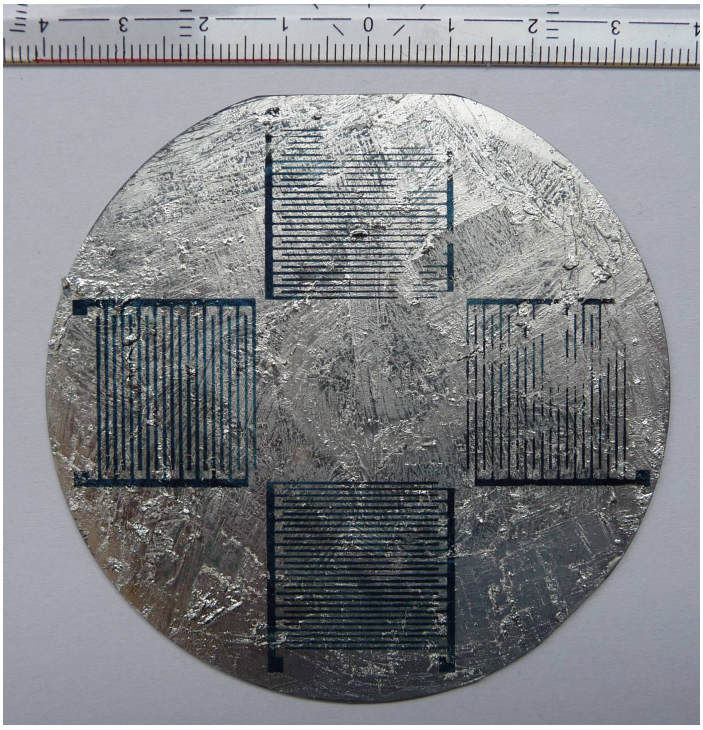

Fig. 5.3: An with indium coated and developed lithium niobate wafer

the piezo tearing apart. To prevent this from happening again, we increased the heat up time and reduced the target temperature to 200°C (392°F). As a result, the piezo didn't tear apart anymore, but the layer of indium was thicker and clumped together in some places. Due to the increased difficulty in exposure and etching, we were only able to transfer a single IDT completely onto the wafer, which still had a relatively long wavelength of 2 mm. Unfortunately, we were unable to move a droplet or generate flows within, using this IDT. The problem, only solvable through professional help, is that our IDT's wavelength is too long. As we discover in the following chapter, we need IDT structures with very short wavelengths, which we cannot create ourselves, since the production of such a small mask is not possible for us. Therefore we are trying to apply the IDT structures professionally, which should allow controlled movement of droplets.

6. Experimental Investigation of flows inside Droplets

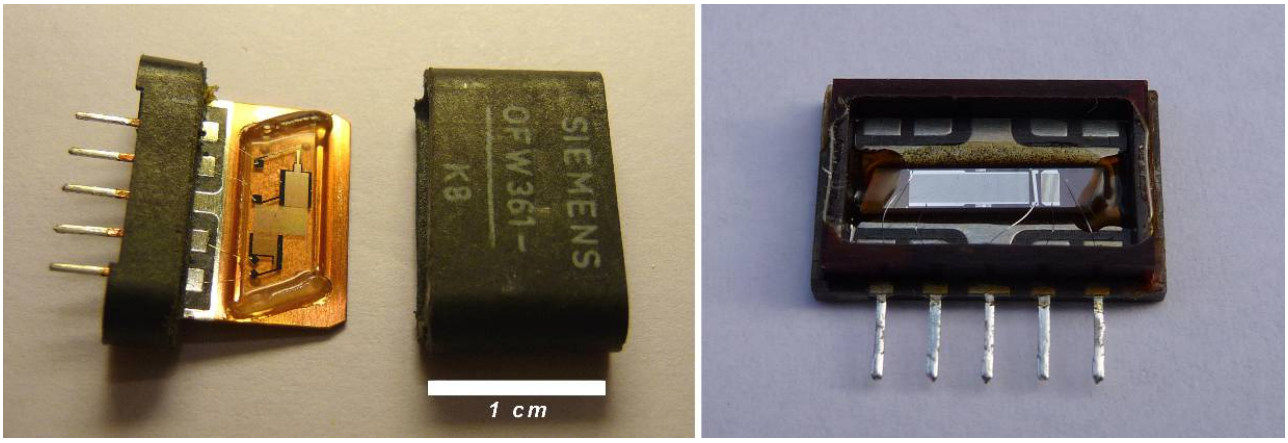

Fig 6.1: Two parted SAW-Filters with different layouts

Since our homemade SSAW units (SSAW for **s**tanding **s**urface **a**coustic **w**ave) were unfortunately not working as hoped, we were facing a great problem. Fortunately, during an internet research, we found cheap surface acoustic wave filters (short SAW filter), also based on the piezo electric effect, which are used to filter specific frequencies mechanically. They are used for example in circuits of mobile phones and TVs, where they are used to transmit data over W-LAN or mobile networks. As in our SAW units, IDT structures are placed on piezo crystals, generating surface waves through applied high frequent AC voltages.

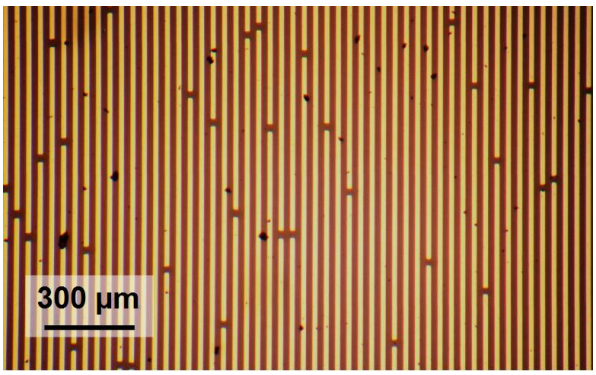

Fig. 6.2: Closeup of a SAW-Filter's layout

At the beginning, we tried experiments with oil droplets floating in deionized water. This did not work well, since the lighter oil floated on the top of the water as thin film after some time. Later on, we tried using a mixture of water and latex spheres with a diameter of about. 1 µm. We were hoping, that the latex spheres would allow a better analysis of the

movement of water inside the droplet. Unfortunately the spheres were not visible under our microscope. As well, the attempt to illuminate the droplet with a laser from the side, did not work. During later experiments, we exchanged the latex spheres with soot, which brought us a step further, since we were now able to get an insight on the flows inside the droplet.

Further, we added some glycerin to the water to slow down the movement and allow a better comparison with our previous simulations. In combination with some condensed milk, we were able to achieve even better results. On the pictures 6.4 and 6.5, the flows inside the droplet are shown. While picture 6.4 is showing a circular flow, 6.5 is showing a central flow, splitting up on

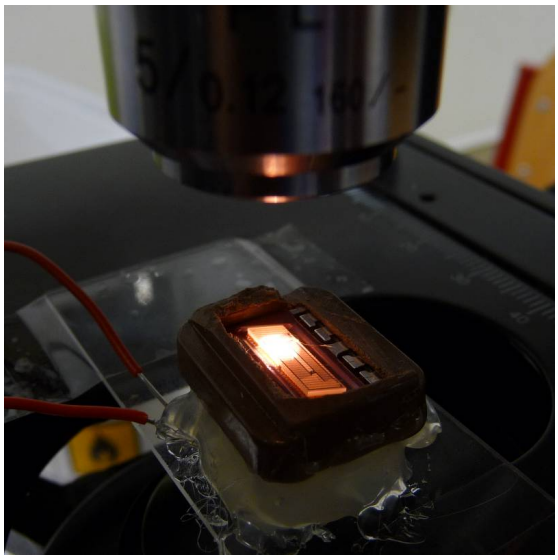

Fig. 6.3: A SAW-Filter under a reflected light microscope

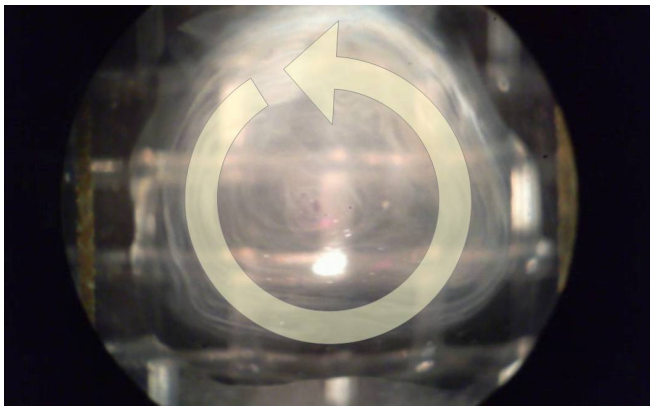

Fig. 6.4: Circular flow inside a droplet when being affected by a surface wafe on just one half.

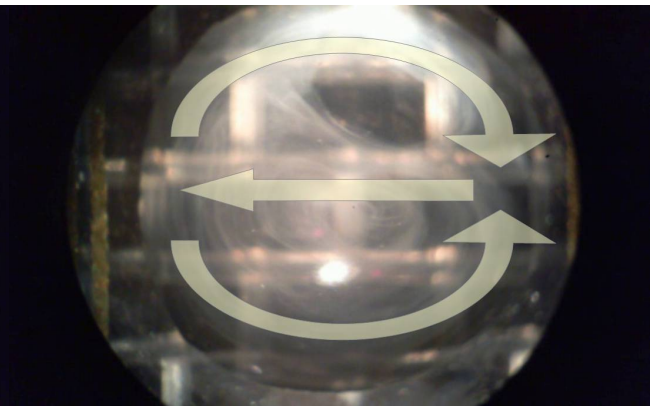

Fig. 6.5: Central flow within a droplet, when being affected by a surface wave on its entire width.

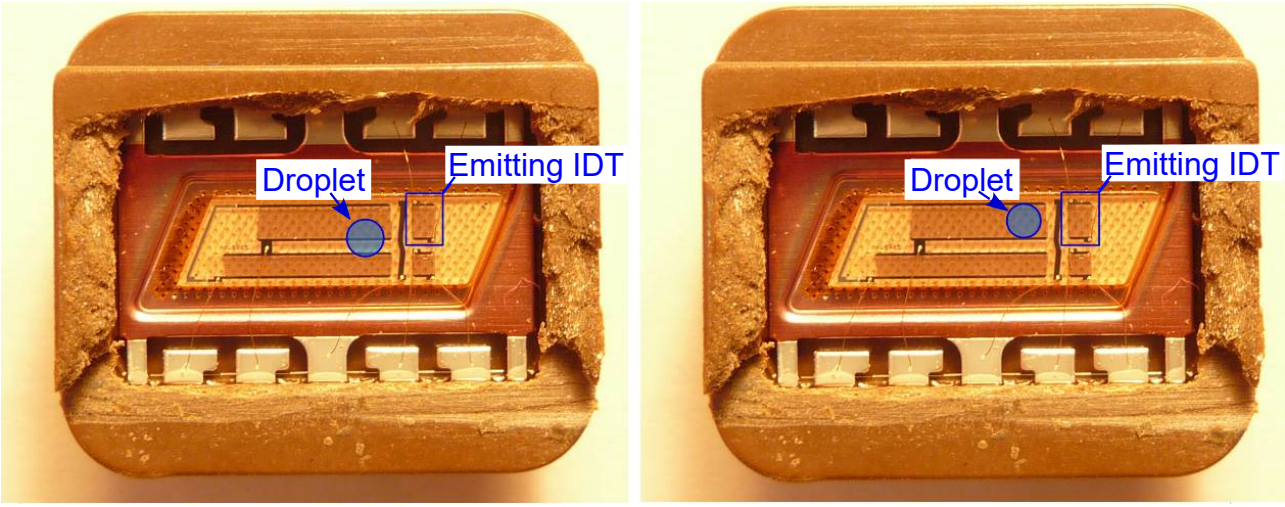

Fig. 6.6: Different positions for a droplet on a SAW-Filter. A circular flow developes in the left picture. A central flow developes in the right one.

the left edge of the droplet, flowing back and merging again on the right side. What we already noticed in our simulations was, that the kind of stream and it's direction is dependent on the position of the droplet, relative to the IDTs on the SAW filter and that we can control the speed through the applied frequency and voltage. The base frequency we were using during our experiments was 32 MHz. During this experiment, we changed the frequency, which resulted in a decrease in speed.

7. Experiments regarding controlled movement of Droplets

As we found out during our experiments, we were missing both the power needed to generate surface waves capable of moving a droplet and the right IDT structures to do so. Therefore we decided to get some professional help from Prof. Achim Wixforth, Dr. Andreas Hörner and Adrian Mainka from the University of Augsburg, Germany, who have lots of experience regarding microfluidics. After we designed a new wafer layout with extremely small IDT structures, we were finally able to produce a wafer that in combination

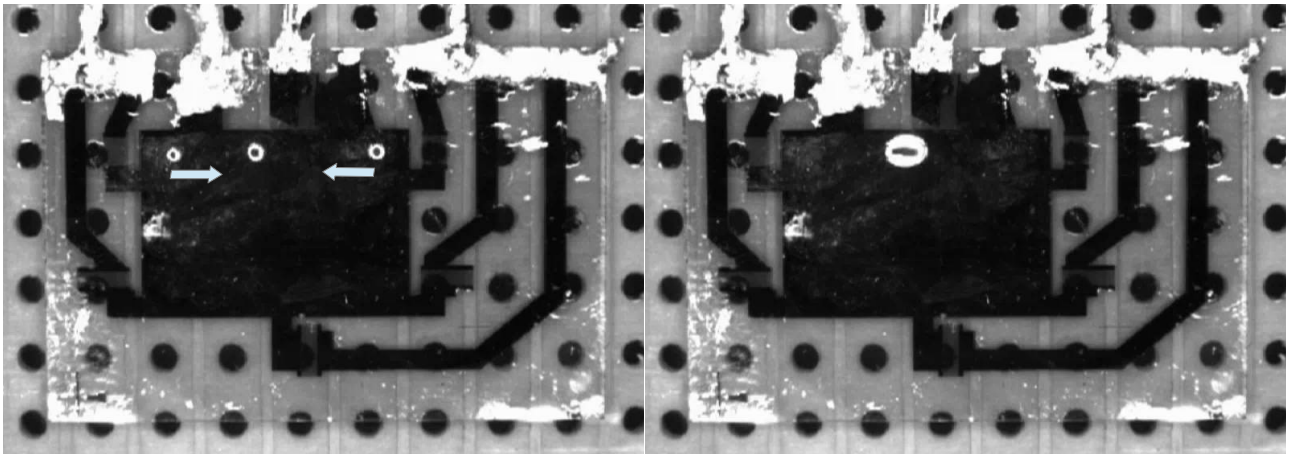

Fig. 7.1: 3 Droplets merging into one large droplet in the middle of the wafer

with one of their amplifiers is able to precisely move droplets across the surface and, as seen in picture 7.1, merge multiple droplets in one spot.

Briefly describing our setup, our new piezo elements (21x14mm in size) feature three pairs of IDT patterns, whereas each pair uses a different frequency (60.6 MHz, 76.75 MHz and 85 MHz; Fig. 7.2). This element is connected to the amplifier, which again is connected to our frequency generator. The camera above our experimental setup sends a live image of the experiment to the laptop, which gives us a better view of the experiment and allows us to record it at the same time (Fig. 7.3). While our frequency generator has a maximum output voltage of 5 Vpp at 60 MHz, the effective output voltage created by the amplifier lies between 300 and 400 Vpp.

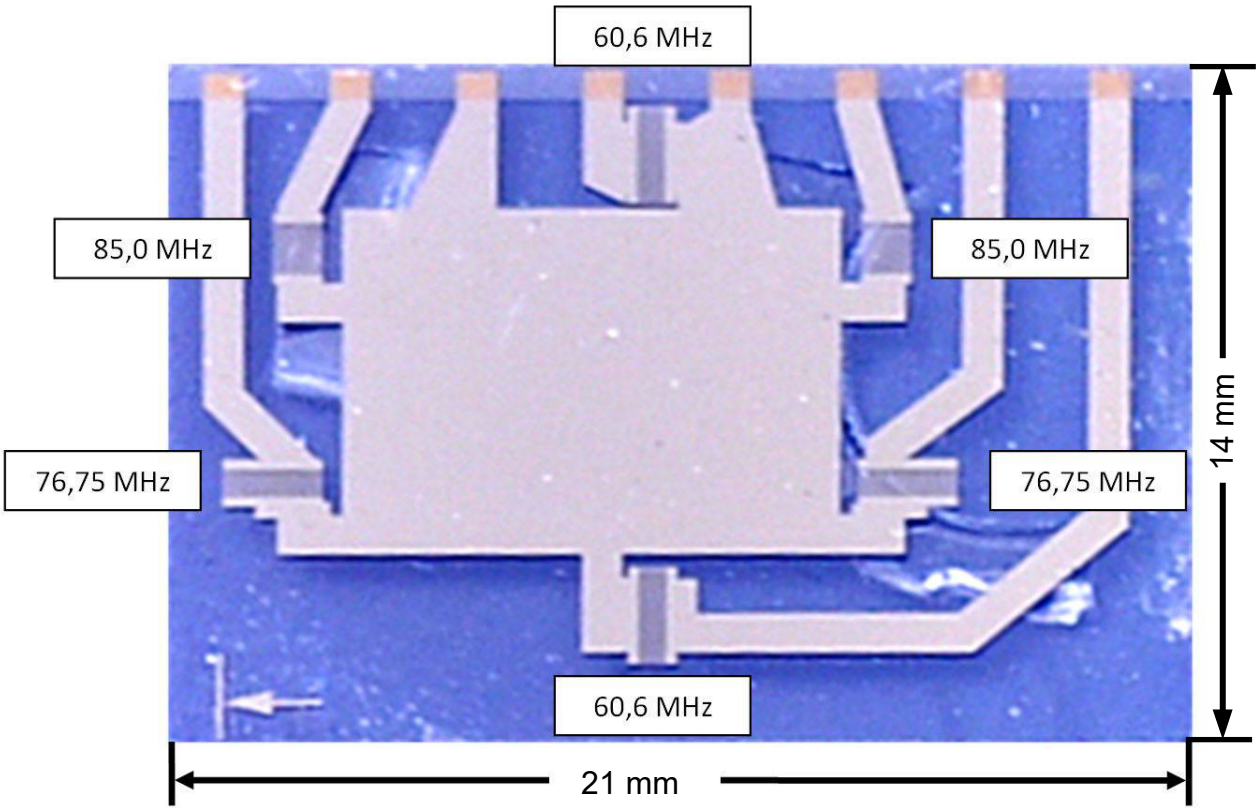

Fig. 7.2: Layout of the new piezo elements

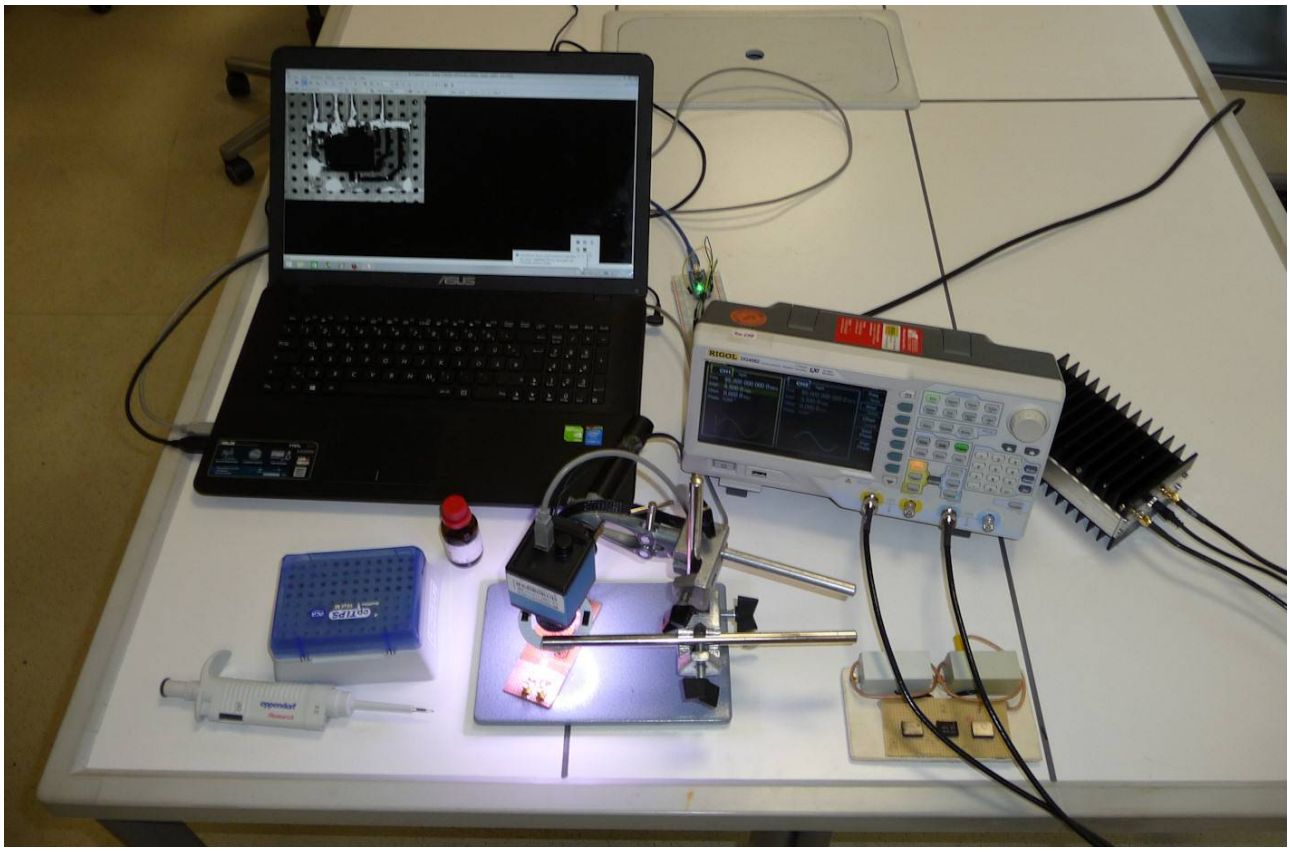

Fig. 7.3: Experimental setup for our experiments

8. Conclusion

During our project, we were able to develop an inexpensive method, for the creation of controlled flows inside of droplets, using N-body-simulations, which we then tested in multiple experiments. Our experiments reproduce our simulations to a high degree, which shows that our underlying theory is correct. Our work shows that it is possible to mix multiple chemicals, to concentrate solids in a droplet and to move these droplets precisely using surface acoustic waves. This is crucial for the construction of Lab-on-a-Chip systems as it enables controlled reactions between multiple chemicals.

9. Acknowledgments

Without the great support of various people, our experiments would not have been possible. We especially thank Prof. Achim Wixforth, Dr. Andreas Hörner and Adrian Mainka from the University of Augsburg, as well as Luca Banszerus and Stefan Hück, two former students from our school, Horst Günter Thum from the “Astropeiler” radiotelescope in Bad Münstereifel, and our supervisor Walter Stein.

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