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Nano- and Pico-Dispensing of Fluids on Planar Substrates Using SAW

Christoph J. Strobl, Zeno von Guttenberg, and Achim Wixforth

***Abstract*—The increasing interest in miniaturization of biological and chemical experiments or assays demands precise metering of the smallest amounts of reagents, e.g., on a planar substrate. Very sophisticated spotting systems can nowadays produce arrays of many thousands of different substances on an area of a few square inches. Such micro arrays and the technology behind them have become an important tool in genomic expression assays, proteomic applications, and even in the field of combinatoric chemistry. We present a technique to dispense the smallest amounts of fluids in the form of either simple spots or more complicated microarrays, where we use surface acoustic waves in combination with a predetermined surface chemistry. In addition to a detailed description of the technique, several examples of applications are presented.**

I. Introduction

The idea of a lab-on-a-chip refers to a system that can perform all steps of a complex biological or chemical reaction or analysis in a miniaturized device. The efforts are directed toward inexpensive solutions that can be automatically controlled and are work reliable. The most obvious attempt is for the downscaling of common fluid handling systems. This can be achieved by etching channels deep into glass, with hot embossing or by casting polymer solutions. The drawback of the narrow channels produced by the latter or by other methods is the high pressure difference that has to be applied in order to pump liquids through them. Therefore the pumps for the actuation have to be very powerful, which is difficult to realize on the same small scale as the channels. More sophisticated methods for pumping liquids through small channels use electrophoretic and electroosmotic effects that induce a flow by the displacement of ions in the solution. This causes a strong dependence on the ion content of the fluid in the channel. Besides making other attempts for the transport in small channels there is another disadvantage of miniaturized flow systems. The Reynolds numbers for a narrow tube is normally smaller than unity, resulting in completely laminar flows. This makes mixing processes, necessary for nearly all biological or chemical reactions, very difficult to perform. To avoid the problems of small tubing systems we developed a microfluidic system on a planar chip that is operated by surface acoustic wave (SAW) actuation. Instead of continuous channels the liquid is transported in single droplets, kept in shape by the hydrophobicity of the chip surface. The SAW can be used for the movement of the droplets, for mixing, and for sensor applications. In a previous publication, we presented the transport and mixing of fluids by SAW [1]. These results will now be augmented by the study of an additional feature permitted by the interplay of SAW actuation and chemically structured surfaces. With hydrophilic spots in a hydrophobic area, small volumes of liquid can be dispensed out of a large droplet. We present two different methods for dispensing with the SAW technique, one that utilizes the influence of the actuation power on the SAW wavelength and a second one that depends more on the physics of wetting on structured surfaces.

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II. Fluid Actuation

The manipulation of small droplets with a SAW utilizes the effect of acoustic streaming [2]. This phenomenon occurs when strong sound fields are in contact with liquids. The liquid is then moved in the direction of the sound wave. The interaction between the SAW and the fluid at the surface of the SAW substrate can either induce acoustic streaming [3] or move a small quantity of fluid, like a droplet, as a whole. In both cases, energy from the SAW is absorbed by the fluid [4].

A SAW with a nonvanishing amplitude in the z-direction, i.e., normal to the surface of the substrate, is then strongly absorbed by the fluid leading to a decaying amplitude in the direction of SAW propagation. Moreover, it creates a small but finite pressure difference $2\Delta p$ in the fluid between the ridges and the wells of the wave, which transforms into a small but finite difference $2\Delta\rho$ in the liquid density. Both quantities then spatially and temporally oscillate around their equilibrium value p_0, and ρ_0, respectively. The pressure difference directly above the surface of the substrate leads to the excitation of a longitudinal sound wave into the liquid. As the sound velocities for the liquid and the solid substrate are in general not equal, this wave is launched under a diffraction angle Θ_R, given by

$$\Theta_R = \arcsin(v_s/v_f). \qquad (1)$$

Here, v_s and v_f denote the sound velocities of the substrate and the fluid, respectively. In addition, the SAW is

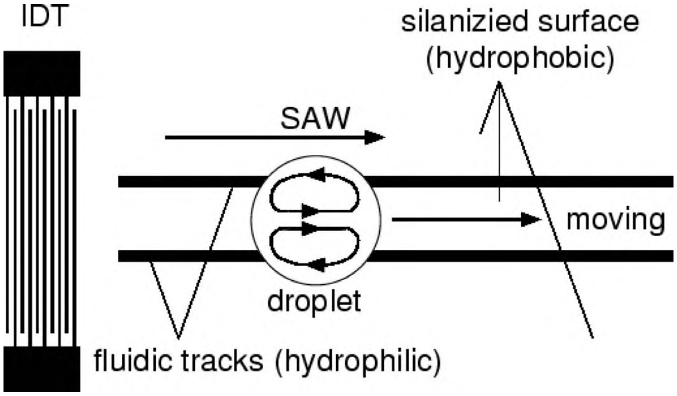

Fig. 1. Sketch of the acoustically driven actuation of a small droplet. A SAW is excited on a piezoelectric substrate by means of an interdigital transducer (IDT). Employing a lithographically defined modulation of the wetting properties of the surface, fluidic tracks are created to hold the droplet within the sound path and to predetermine its trajectory.

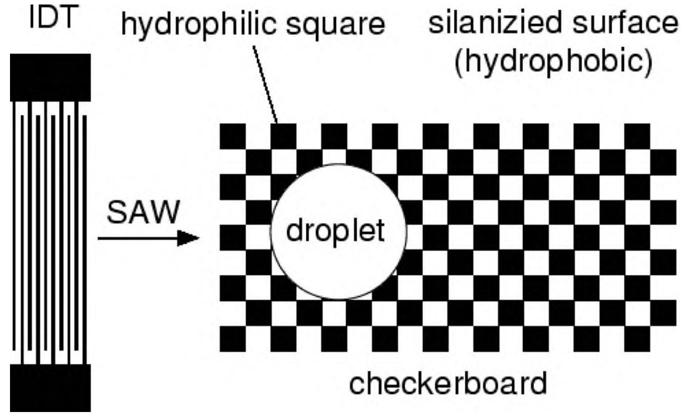

Fig. 2. The chip layout for an array dispenser. A modulation of the wetting properties (black: hydrophilic, white: hydrophobic) employing an OTS-based surface chemistry defines the position and the size of the dispensed droplets.

responsible for the buildup of an acoustic radiation pressure

$$P_S = \rho_0 v_S^2 \left(\Delta\rho/\rho_0\right)^2 \qquad (2)$$

in the direction of the sound propagation in the fluid. This leads to an internal streaming in a closed volume, such as a droplet, as the boundary of the droplet reflects the actuated fluid back to the source. For larger SAW power, the droplet becomes deformed from its equilibrium shape, which leads to different wetting angles in ‘luff’ and ‘lee’ of the SAW and is moved as a whole into the direction of SAW propagation.

The SAW in our experiments was generated on a piezoelectric 128°rot $LiNbO_3$ substrate with gold interdigital transducer (IDT) consisting of twelve pairs of split-4 fingers with 600 μm aperture. The resonant frequencies were $f = 114$, 340, 567, and 800 MHz. The corresponding wavelengths were $\lambda = 33$, 11, 6.8, and 4.8 μm. The IDT showed an insertion loss of about 6 dB each, if measured in a delay line configuration. To control the direction of droplet movement, the surface wetting properties of the lithium niobate were modulated. In a first step the substrate was coated with 200 nm quartz in a plasma-enhanced chemical vapor deposition process. Then the surface was silanized with octadecyltrichlorsilane (OTS) [5], [6]. In this process, a self-assembled monolayer is formed on the chip surface which changes the wetting properties from hydrophilic to hydrophobic. To define so-called fluidic tracks [1] or more complex modification geometries in the surface chemistry of the chip, the silanized surface is structured by photolithography in a series of subsequent standard processing steps. A pulse-modulated RF signal is fed to the IDT and a SAW propagates over the chip. When the SAW reaches the contact line of a droplet on the substrate surface situated in the acoustic path (600 μm wide), it moves along the fluidic tracks over the chip (Fig. 1).

A water droplet pushed over the surface keeps its circular shape and does not lose parts of the liquid. The experimental setup therefore leads to reproducible results. For larger droplets ($V > 50$ nL), the humidity conditions are not controlled because the water droplet does not evaporate during the course of the experiments. For smaller droplet sizes, evaporation can be prevented by controlling the humidity of the surrounding in a sealed sample holder or by cooling down the sample below the dew point. For high temperature processes, the droplets can even be covered by a mineral oil layer, as will be discussed elsewhere.

III. Array Dispenser

To dispense small amounts of liquid with an exact volume is still a matter of intensive research. Most ideas are based on the principle of an ink jet printer, where a small droplet is formed by piezoelectric compression of a small cavity. For lab-on-a-chip systems the dispensing needs to incorporate small amounts of reagent out of an exterior reservoir into the processing device. The combination of SAW actuation and wetting properties on chemically structured surfaces can very effectively be used for such an exact dispensing of small amounts of liquid on a planar chip surface. On a patterned surface containing hydrophobic and hydrophilic areas, water droplets will preferably stay on the latter to reduce the free energy. To investigate how this phenomenon can be utilized for dispensing purposes, a checkerboard pattern was processed into the hydrophobic OTS surface of a SAW device. The squares had side lengths of 65 μm and were situated in the acoustic path (Fig. 2).

After placing a large droplet ($V = 50$ nL) on the checkerboard pattern within the acoustic path, a RF signal with a frequency of $f = 114$ MHz pulse-modulated with a repetition rate of 10 ms and a pulse width of 1 ms and at least 20 dBm power was applied to the IDT. The droplet is pushed over the checkerboard by the propagating SAW. The checkerboard ensures the same droplet guidance effect as a fluidic track. During the movement of the droplet it looses its circular shape and the contact line is deformed by tonguelike tethers. They are ruptured when the contact line moves on, leaving a small amount of water in the hydrophilic squares. This scheme operates reproducibly and every square is filled with the same amount of liquid. The size distribution of the droplets is very narrow, as their volume is solely determined by the difference in surface energy and the geometry of the squares acting as hydrophilic anchors. A very interesting fact is that the small droplets are not affected by the SAW and the large droplet is not retarded, owing to the mass loading on the substrate between droplet and IDT.

The size of the dispensed droplets was measured with a microscope. The wetting angle of water on OTS of about 105° and the side length of the squares are the determining geometrical values. With these values the droplet volume can be estimated to be of the order of 50 pL. Direct measurement yielded a volume between 10 and 30 pL. The small size of the droplets in this case leads to a fast evaporation of the dispensed droplets in 0.4 sec at 18°C. Reducing the temperature close to the dewpoint increases the life time up to 10 s. Below the dewpoint the dispensed droplets can be conserved for a much longer time for further experiments.

Based on the split-4 geometry IDT, a SAW with a higher frequency could be generated on the same chip. To test the influence of the SAW wavelength on the dispensed droplets in the square array, the frequency was changed to $f = 340$ MHz with the RF power kept constant. At this higher frequency, the dispensed very small droplets could also be moved. With the SAW acting like a snow plow, the droplets are pushed to their next neighbors where they merge. This happens in a cascade fashion until the acoustic path is emptied. Moving a large droplet with an $f = 340$ MHz SAW over the checkerboard yields no dispensing effect, as expected from the preceding results.

IV. Selective Actuation

In the following experiment, the influence of the wavelength of the SAW is more closely inspected. As observed before, the dispensed droplets (base diameter $d = 50$ μm) are too small for a SAW at $f = 114$ MHz ($\lambda = 33$ μm) to be affected, but not for $f = 340$ MHz ($\lambda = 11$ μm). Therefore, the ratio of the droplet diameter and the wavelength of the SAW seems to be critical for the actuation of small droplets.

To measure this influence more directly, a sample comprising lithographically defined fluidic tracks was used (see Fig. 1). While keeping RF power, frequency, and modulation constant, the droplet velocity as a function of their volume was investigated. When the diameter of the wetted area is large compared to the SAW wavelength, no dependence of the droplet velocity on its volume can be recognized (Fig. 3). However, if the volume is reduced and the radius becomes comparable or smaller than the SAW wavelength, the velocity decreases dramatically. A

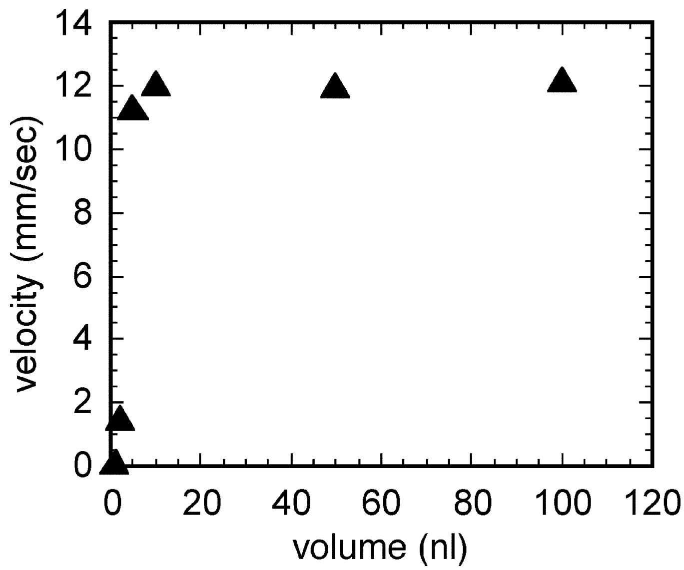

Fig. 3. Measurement of the droplet velocity as a function of the droplet volume for fixed SAW power and frequency. For droplet volumes corresponding to a base diameter of less than the SAW wavelength, a strong dependence of the velocity on the volume is observed; as soon as the base diameter of the droplet becomes larger than the SAW wavelength, saturation of the reachable velocity is observed.

$V = 1$ nL droplet is nearly immobile at $f = 114$ MHz. When the same droplet was moved with a SAW frequency of $f = 340$ MHz, the velocity increased.

V. Passive Dispenser

For an alternative, acoustically mediated dispensing method, a different chip design and surface chemistry was used. The chip had six transducers, allowing actuation of the droplets on two parallel and one perpendicular pathways. The surface in this case was covered with gold and modified successively by two different kinds of thiols. The chemical patterning of the surface was achieved with the help of photolithography.

A small droplet can also dispensed in a more passive way by the SAW. In the setup shown in Fig. 4, a fluid reservoir is formed by a large prolate hydrophilic spot surrounded by a hydrophobic area. Using a SAW to move the fluid bulge partly out of the spot leads to an increase of the free energy and therefore creates a backward force. If the SAW is switched off, the former equilibrium state of the bulge is restored. This can be used to dispense a small amount of liquid in a hydrophilic spot anchor close to the prolate area. Under the action of a SAW, this anchor is covered by the fluid, and after the RF power is switched off, the retracting bulge leaves the spot wetted. With a second perpendicularly orientated SAW transducer, the dispensed droplet can then be moved to other positions on the chips (Fig. 5). In this setup, transducers with the same RF frequency can be employed because the spot size was chosen according to the design rule (diameter large as compared to the SAW wavelength at 114 MHz). The spot diameters

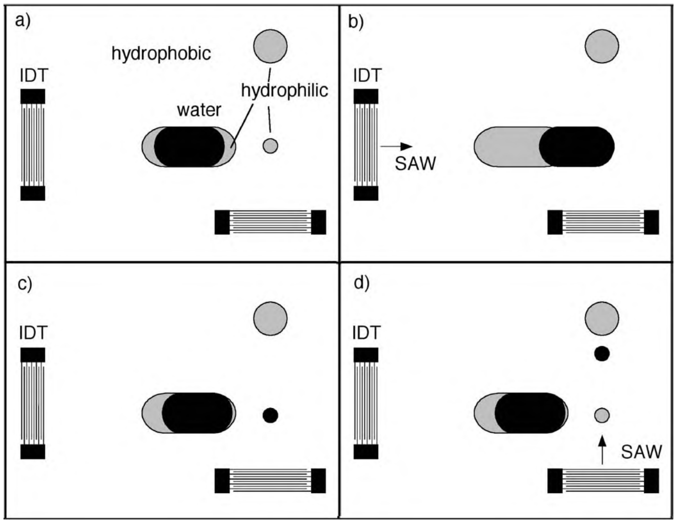

Fig. 4. Sketch of the SAW-driven passive dispenser and time sequence of the dispensing process as explained in the text. In (a), the IDT on the left is activated to push the reservoir droplet towards the hydrophilic anchor. In (b), the hydrophilic anchor is wetted by the reservoir. In (c), switching off the SAW retracts the reservoir droplet leaving a small droplet at the anchor site. In (d), a second SAW is used to push the dispensed droplet toward a container anchor (larger gray area to the upper right).

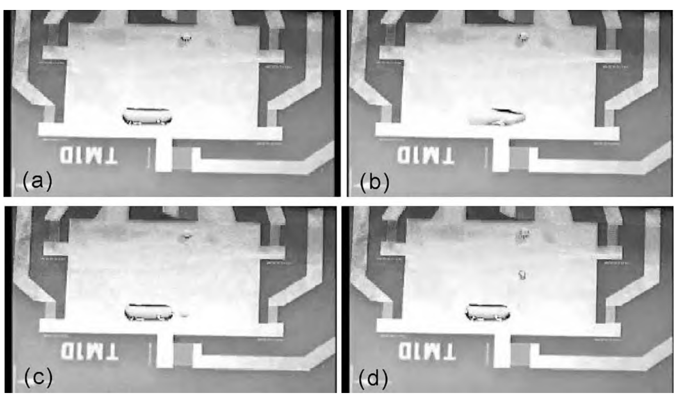

Fig. 5. Photograph of the dispenser chip as described in Fig. 4.

in the experiment were in the range of 300 μm, yielding a liquid volume of around 2 nL. This dispensing method works very reproducibly, allowing distribution of a large number of droplets on the chip or collection of them in a single spot to yield larger volumes.

VI. Conclusion

All of our fluidic chips are produced employing common methods of chip processing. The use of planar processes opens the possibility for a low-cost production. Furthermore, the application of a dispenser needs no additional step in the production because all the processing steps are integrable in the former steps. Also, the dispensers are integrable to a SAW-based lab-on-a-chip system without any technology break.

Two methods for dispensing with the SAW technique were presented. The array dispenser utilizes different SAW wavelengths to separate the actuation of large and small droplets, and the passive dispenser uses a hydrophilic area to remove a small amount of water from a predefined dispensing spot. An array dispenser is suitable to create an array of droplets or dried spots for applications like DNA-test. The array dispenser works reliably and fast and needs no control system. A passive dispenser is suitable to dispense any volume with an external control. In this case there is no expensive change in hardware setup needed and thus represents a cheap solution.

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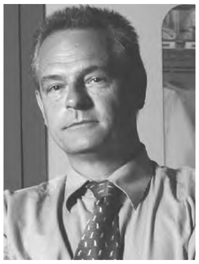
Christoph Strobl was born in 1975 in Munich, Germany. He received the Diploma in Physics degree from the University of Munich. He was one of the first students in Prof. Wixforth’s group to work on the acoustically driven fluid actuation on piezoelectric substrates. Presently, he is a Ph.D. student both in Augsburg and in Munich investigating the effect of the piezoelectric fields of a SAW in combination with planar fluidic biochips.

Zeno von Guttenberg was born in 1970 in Werneck, Frankonia, Germany, and studied physics in Freiburg and in Munich, Germany. He received his Diploma in biophysics from the Harvard Medical School in Boston and his Ph.D. degree from the Technical University in Munich (biophysics). Apart from a strong interest in biophysical phenomena, his research activities extend also into surface chemistry. Since 2001, Dr. von Guttenberg has been a member of the research staff at the Advalytix AG, Brunnthal, Germany.



Achim Wixforth was born in 1956 in Bielefeld, Germany. He graduated in physics at the University of Hamburg, where he received his Ph.D. degree in 1987. He spent a postdoctoral year at the University of California, Santa Barbara, where he cooperated with Profs. Gossard, Kroemer, and Petroff as an assistant research engineer. Back in Germany, he received his "Habilitation" from the University of Munich in 1994 where he was senior scientist and lecturer before accepting a chair for experimental physics in Augsburg, Germany. He is author and co-author of more than 160 scientific papers, and has a long list of invited talks. His research interests include collective phenomena, transport, mechanical, and optical properties of semiconductor nanostructures, the biophysics of membranes and cells, and surface acoustic waves in all aspects. Among other awards, he received in 1998 the "Walter-Schottky-Prize" for solid state research, and in 2002 the first German NanoBio award.

Prof. Wixforth is Chief Technology Officer and co-founder of Advalytix AG, Brunnthal, Germany, a young spin-off company of the Center for NanoScience (CeNS) of the University of Munich.