Push/pull actuation using opto-electrowetting

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Received 17 March 2007; received in revised form 25 July 2007; accepted 14 August 2007
Available online 19 August 2007

Abstract

It is shown theoretically and experimentally that opto-electrowetting may be used for both pulling and pushing liquid droplets. A theoretical analysis based on the Lippmann-equation and an electronic equivalent circuit model allows definition of a voltage and frequency range for which pushing may be achieved, a novelty in electrowetting-based actuation. Experimental confirmation of the effect demonstrates that enhanced flexibility in micro-fluidic actuation may be obtained under appropriate bias conditions.
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Keywords: Opto-electrowetting; Electrowetting; Electrowetting-on-dielectrics; Liquid handling; Micro-fluidic device; Lab-on-a-chip

1. Introduction

Electrowetting allows modification of the contact angle of a liquid droplet on a surface through application of an electric field. Typically, the contact angle of a conductive or polar liquid droplet resting on a dielectric substrate is decreased by applying a bias voltage between the droplet and an electrode underneath the dielectric layer. Although long known [1], the effect has in recent years seen a renaissance of interest since it has proven of great utility for applications in optical Microsystems or droplet-based micro-fluidic systems.

In micro-optics, electrowetting has been successfully applied as a tuning mechanism for adaptive liquid lenses without any mechanically moving parts [2–4]. In these systems, the change in contact angle and curvature of a liquid droplet caused by the applied voltage is exploited to incite a change in focal length of the droplet acting as a liquid lens. This approach is sufficiently well advanced that commercial products based on it are available.

Electrowetting is also a promising actuation mechanism in droplet-based micro-fluidic systems [5] because of its ability to precisely handle small amounts of liquid. In typical systems, an array of electrodes is configured in such a manner that voltage may be applied over only a part of the droplet; the contact angle is thus reduced only in that area. The induced net force pulls the droplet towards the region over the biased electrode. This mechanism allows movement of droplets along a path defined by a pattern of individually addressable electrodes, or over arbitrary paths over an array of electrodes. Furthermore, splitting, merging and creating of droplets has been demonstrated [6], important for analytical applications.

The limitations imposed by the large number of electrodes required for droplet manipulation can be overcome by opto-electrowetting (OEW), an effect which was first presented by Chiou et al. [7,8]. By means of a photoconductive layer deposited underneath the dielectric layer, the voltage drop over the dielectric layer can be controlled by light. High electric fields over parts of a droplet may therefore be defined by light irradiation of the substrate. By using moving light patterns, liquid droplets may be moved continuously over a substrate, and even cell sorting is accessible using very simple devices [9]. All electrowetting-based systems demonstrated so far rely on a reversible decrease of the contact angle of the conductive liquid under applied voltage, resulting in a pulling force on a droplet towards the higher electric field.

We present here a theoretical analysis and experimental confirmation for the existence of optical pushing behavior in opto-electrowetting. Using an electronic equivalent circuit model, we derive the necessary experimental conditions to achieve this effect and show how the results may be used to obtain greater flexibility in opto-electrowetting-based micro-fluidic actuation.
2. Theory

2.1. Electrowetting

The influence of electric fields on interfacial energies of liquid droplets was first described by Lippmann in 1875 [1] who investigated the interfacial energy of a mercury–electrolyte interface as a function of an applied bias voltage. He explained the observed effects by the reversible charging of the capacitance formed by the Helmholtz electric double layer formed at the interface between the liquids.

More recently, Berge [10] revived electrowetting through his idea of using an artificially generated insulating layer between a solid electrode and a conductive liquid droplet. Because of the thickness of this insulating layer, which exceeds the thickness of the Helmholtz double layer (typically a few nanometers) by several orders of magnitude, the actuation voltages in the configuration reported by Berge are significantly higher than in the work of Lippmann. This configuration is now commonly known as electrowetting-on-dielectrics (EWOD) and has become the one most frequently used.

The contact angle, \( \theta \), of a droplet sitting on a planar dielectric substrate in the EWOD configuration can be described using the so-called Lippmann-equation [10]

\[
\cos(\theta) = \cos(\theta_0) + \frac{C}{2\gamma_{LG}} V^2 = \cos(\theta_0) + \frac{\epsilon \epsilon_0}{2\gamma_{LG}} V^2
\]

where \( \theta_0 \) represents the initial contact angle, \( \epsilon \) the permittivity of the dielectric layer, \( \gamma_{LG} \) the interfacial energy between the droplet and the ambient phase, \( t \) the thickness of the dielectric layer, \( C \) the capacitance of the system, \( A \) the area covered by the droplet, and \( V \) is the voltage applied over the total capacitance.

Eq. (1) is derived using the Young-equation with an additional term representing the energy stored in the capacitance formed by the droplet and the electrode under the dielectric layer. Consequently, the voltage drop over the total capacitance is that which leads to the spreading of the droplet.

Eq. (1) allows us to draw a number of conclusions concerning electrowetting and its limitations: first, due to the quadratic voltage dependence, the polarity of the bias voltage is not relevant, such that dc or ac biases may be used and secondly, only a contact angle reduction can be achieved in EWOD. In addition, we see that the driving voltage amplitude may be reduced by a low thickness and a high permittivity of the dielectric layer.

2.2. Opto-electrowetting

Recall that it is the voltage drop across the total capacitance that is responsible for the reduction of the contact angle. Thus, by introducing a variable resistance into the system and using an ac-voltage, the contact angle may be varied by changing this resistance. This effect is exploited in opto-electrowetting, in which a the variable resistance is provided by a photoconductive layer. Since photoconductive materials reduce their resistivity by about three orders of magnitudes upon illumination, a contact angle difference of \( 10^\circ \) or more may be achieved between illuminated and dark states.

In a typical opto-electrowetting configuration, the photoconductive layer is placed underneath the dielectric layer as shown schematically in Fig. 1. The system thus consists of an electrode, the photoconductive layer, a dielectric coating, and the liquid droplet. The fabrication of these substrates is easy: only layer deposition, and no photolithography, is required.

The change of contact angle can then be used to move droplets from one position to an other. By non-uniform or partial illumination of the photoconductive layer beneath the droplet, the contact angle of the droplet changes (reduces) only on the light side of the droplet. The corresponding gain in interfacial energy leads to movement of the droplet towards those areas of the substrate in which the droplet has a smaller contact angle, i.e., generally in direction of the illumination. We will show below that, under certain experimental conditions, the smaller contact angle can be generated on the dark side of the droplet.

3. Simulation

3.1. Equivalent circuit model

The electrical and mechanical behavior of an OEW-system can be modeled by representing the structure in an equivalent circuit model, as shown in Fig. 1 and adjusting the capacitance formed between the droplet and the electrode. The dielectric layer is represented by a simple capacitance \( C_{de} \) with a complex impedance \( Z_{de} = 1/j\omega C_{de} \), in which \( \omega \) is the angular frequency of the ac bias voltage. The photoconductive layer is modeled as a parallel combination of a capacitance \( C_{pc} \) and a resistance \( R_{pc} \), where the latter is decreased by 3–4 orders of magnitude upon illumination due to carrier generation.

In Eq. (1) an additional term to the Young-equation was implemented representing the energy stored in the capacitor formed between the droplet and the electrode. In OEW the capacitance of the photoconductive layer has to be taken in care, too. Thus, the Lippmann-equation has to be adjusted by the total capacitance of the dielectric and the photoconductive layer. This
capacitance $C_{tot}$ is given by

$$C_{tot} = \frac{C_{de} \cdot (1 + \omega^2 R_{ph}^2 \epsilon_{ph}^2)}{1 + \omega^2 R_{ph}^2 \epsilon_{ph} (C_{de} + C_{ph})}$$  \hfill (2)

More, for calculating the voltage which drops over the total capacitance formed by the dielectric and the photoconductive layer, a resistance $R_p$ including the resistances of the liquid droplet and of the electrodes and all other parasitic resistances in the system has to be considered. The voltage between the droplet and the electrode beneath the dielectric and the photoconductive layer may then be calculated by

$$V_C = \frac{Z_{de} + Z_{pc}}{Z_{pc} + R_p + Z_{de}}$$

$$V = \frac{(1/j\omega C_{de}) + (R_{pc}/(j\omega C_{pc} R_{pc} + 1))}{(1/j\omega C_{de}) + (R_{pc}/(j\omega C_{pc} R_{pc} + 1)) + R_p}$$  \hfill (3)

in which $C_{de}$ is the capacitance of the dielectric layer, $V$ the applied voltage and $V_C$ is the voltage dropped over both the dielectric and the photoconductive layer.

Irradiation of the photoconductive layer with light leads to a change in its resistance $R_{pc}$, thereby changing the voltage drop over the dielectric layer. This, in turn, changes the contact angle in the illuminated areas of the substrate. It can be seen from Eq. (3) that OEW (in contrast to EWOD) only works with ac-voltages, since only in that case coupling between the capacitances and thus an induced voltage drop over the photoconductive layer occurs. For the same reason, the OEW-effect strongly depends on the frequency of the applied voltage.

Combining the influence of illumination of the photoconductive layer as modeled by the transfer function, Eq. (3), and the capacitance of the complete system, Eq. (2), with the contact angle change as a function of applied voltage, as described by Eq. (1), we obtain the following relationship:

$$\cos(\theta) = \cos(\theta_0) + \frac{C_{tot}}{2\gamma \sigma A} \times \left| \frac{(1/j\omega C_{de}) + (R_{pc}/(j\omega C_{pc} R_{pc} + 1))}{(1/j\omega C_{de}) + (R_{pc}/(j\omega C_{pc} R_{pc} + 1)) + R_p} \right|^2.$$  \hfill (4)

Here, the capacitance from Eq. (2) has to be taken into account as a capacitance per area, thus it is divided by the droplet base area on the substrate, $A$.

Eq. (4) finally describes the contact angle obtained in an OEW setup as a function of the resistance $R_{pc}$ and the frequency $\omega$ of the applied voltage.

3.2. Modeling results

To demonstrate the utility of this model for the OEW-system, we apply it to a configuration corresponding to the experimental setup used later in this paper. The system considered here consists of an n-doped silicon wafer as the substrate, a photoconductive layer of hydrogenated amorphous silicon (a-Si:H), and a silicon dioxide dielectric layer (SiO$_2$); for the liquid droplet, 1 mol NaCl solution is assumed. Hydrogenated amorphous silicon was chosen because of its high dark conductivity and ease of deposition. The values of the material parameters used in the calculations are listed in Table 1. They are used unless otherwise specified.

In Fig. 2, the calculated contact angle as a function of the frequency of the applied voltage, $f$, is compared for the illuminated and dark states. A voltage amplitude of 30 V was assumed in all calculations.

The influence of the parasitic resistance, $R_p$, may be seen from the relationship plotted in the figure: independently of the existence of a non-vanishing parasitic resistance $R_p$, the contact angle on the illuminated substrate is smaller than that on the substrate in the dark state in the frequency range from 100 Hz to 5000 Hz, leading to a pulling of the droplet by light, such as would be seen if illuminated by a focused laser beam. This frequency range is thus well suited for pulling motion of the liquid by light, and pulling motion may be observed in systems with zero parasitic resistance as well as with non-vanishing parasitic resistance. For the frequency tending to infinity, the
contact angles approach each other, such that optically-induced movement can no longer be accomplished.

In contrast to systems with zero parasitic resistance, systems with a finite parasitic resistance exhibit a frequency range in which the contact angle in the illuminated state exceeds the one in the non-illuminated state. Hence in this frequency range, *pushing* motion of the droplet is achieved. Compared to the contact angle difference for pulling motion, the contact angle difference in pushing motion is much smaller, as will be discussed below in more detail. It should be stressed, however, that the contact angles under bias voltage are still smaller than the Young’s contact angle, i.e., the initial contact angle at zero applied voltage.

We consider the contact angle difference between illuminated and dark states expected for systems with a non-vanishing parasitic resistance in more detail. In Fig. 3, the contact angle difference between illuminated and dark states for an OEW-device is plotted as a function of frequency. At a parasitic resistance of 500 kΩ, this contact angle difference is positive at frequencies higher than 900 Hz and the maximal difference is 2.74° at a frequency of 1601 Hz. For smaller parasitic resistances the frequency in which pushing can be achieved is shifted to higher values.

It can also be seen in Fig. 3 that the frequency range for the pulling regime decreases with increasing parasitic resistance. Conversely, *pushing* the droplet is facilitated at moderate resistances.

We thus expect that both pulling and pushing motion may be accomplished on the same substrate by an appropriate choice of the frequency and amplitude of the driving voltage, thereby allowing additional flexibility in the motion control of liquids. Furthermore, since pushing motion is achieved only in the case of a non-vanishing parasitic resistance, a resistor may be added to the system in the case of highly conductive materials of the droplet and the substrate. Therefore, for all types of OEW substrates, both types of motion may be accomplished.

### 3.3. Forces

For an estimation of the forces which can be achieved by OEW, we adopt the electro-mechanical model presented by Jones [11]. In this approach, the electrical force on the three-phase contact line of the liquid is calculated by the gradient of the electrical energy stored in the capacitor formed by the liquid and the conductive part of the substrate, assuming a wedge-shaped geometry. The result for the net force per unit length on the droplet in the z-direction is then given by

\[ f_e = \frac{C_{tot}}{2A} V^2, \]  

in which \( C_{tot}/A \) is again the capacitance per area.

Using Eq. (5) and combining it with the transfer function of Eq. (3), the net force per unit length as a function of frequency and parasitic resistance was calculated. Fig. 4 shows the results of these calculations. The maximum net force per length unit is
Fig. 6. Side view of a droplet with a volume of 0.28 μl on an (a) non-illuminated and (b) illuminated OEW substrate, both at a bias voltage of 59.5 V at 700 Hz.

44.02 mN/m at a frequency of 71 Hz and a parasitic resistance of 50 kΩ, i.e., for pulling actuation.

For pushing actuation at the same voltage level, the maximum force is smaller by three orders of magnitude, as can be seen in Fig. 5. The maximum pushing force is 0.403 mN/m at a frequency of 7000 Hz and at a parasitic resistance of 100 kΩ. An increase of the pushing force may be achieved by increasing the voltage amplitude. In the frequency range enabling pushing motion, the voltage drop over the dielectric layer is in the range of one third of the applied voltage, which may be seen from the high contact angles shown in Fig. 2. Consequently, the voltage amplitude may be increased in this frequency range without the danger of dielectric breakdown.

Note that all forces discussed here are forces per unit length. For achieving directional motion of a droplet, the contact angle must be modified only in parts of the three-phase contact line. Consequently, the total force acting on a droplet has to be calculated by integration over the force contributions of all parts of the three-phase contact line.

4. Experimental results

For experimental verification of the theoretical results presented above, OEW substrates were fabricated using a standard n-doped silicon wafer (resistivity 1–5 Ω cm). The wafer was covered by a photoconductive layer of hydrogenized amorphous...
silicon (a-Si:H) with a thickness of 800 nm, and a magnesium fluoride dielectric layer (MgF2) with a thickness of 400 nm. To obtain a high initial contact angle, a hydrophobic layer of C4F8 polymer (thickness 20 nm) was deposited. Finally, the wafer was diced into 20 mm × 20 mm chips.

The OEW-effect was demonstrated in a setup using the OEW substrate and a sessile droplet on top of it which was contacted directly by a probe needle. The profile of the droplet was imaged by a horizontally positioned microscope equipped with a video camera and pictures of illuminated and non-illuminated droplets at different frequencies and voltages were taken. The volume of the droplet was 0.28 μl. Applying a bias voltage of 59.5 V amplitude and a frequency of 700 Hz, the contact angle of the droplet decreases upon illumination, as seen in Fig. 6. At a bias voltage of 104.5 V and at a frequency of 8.5 kHz, the contact angle of the same droplet is increased under illumination, as seen in Fig. 7.

In a following experiment, a sandwich setup using an OEW substrate was used to demonstrate droplet motion. In the sandwich setup, electrical contact to the droplet was achieved by an indium-tin-oxide (ITO) coated glass slide mounted at a distance of 500 μm above the OEW substrate, as seen in Fig. 8. The ITO glass slide was also covered by a thin hydrophobic layer. A droplet with a volume of about 0.4 μl was placed between the substrates, and the residual volume between the flat substrates was filled with silicone oil (Dow Corning 200) for reducing pinning and to render smooth movement of the droplet. As a light source, a Helium–Neon laser with a power of 1 mW and at a wavelength of 633 nm was used. Other laser sources with a higher power were also tested but a significant change of the wavelength of 633 nm was used. Other laser sources with a higher power were also tested but a significant change of the wavelength of 633 nm was used.

Using this sandwich setup it was possible to experimentally demonstrate pushing motion of a liquid conductive droplet by optoelectrowetting for the first time. Fig. shows stills taken from a video in which pushing motion was observed at a frequency of 1.5 kHz and a voltage of 75 V. The obtained velocity of the pushing motion was in the range of a few millimeters per second.

5. Conclusion

We have studied the optoelectrowetting effect based on the Lippmann-equation together with a lumped parameter electric model. It was shown for the first time that by optoelectrowetting both pulling and pushing motion of liquid droplets can be achieved, depending on the electric parameters of the bias voltage and of the system itself. Experiments were shown which corroborate the theoretical predictions.

A key parameter for defining pushing and pulling is the series parasitic resistance resulting from, for example, the finite conductivity of the droplet or a resistor artificially added to the system. Taking this resistance into account, it is possible to switch between both pushing and pulling by merely changing the frequency and amplitude of the bias voltage. Applications such as droplet-based micro-fluidics, lab-on-a-chip systems and optical cell sorting or manipulating systems may benefit from these results.

6. Acknowledgements

We gratefully acknowledge the help of Holger Krause, Maximilian Marhöfer and Regina Ko in preparing the figures.

References

Biographies

Florian Krogmann (Non-member) was born in Freiburg, Germany. He studied microsystems engineering at the University of Freiburg, where he graduated in 2002 (Dipl-Ing) with a diploma thesis about a silicon optical cavity for optical sensors. Since 2003, he has been working as a PhD student in the Laboratory for Micro-optics in the field of tunable micro-optical devices based on electrowetting.

Hong Qu made her Bachelor for applied electronics in 1996 in China. In 2006 she graduated her studies in microsystems engineering at the University of Freiburg with a diploma thesis about moving droplets using opto-electrowetting. Since 2007 she is working at the Institut für Solarenergieforschung Hameln, where she makes researches within the field of local chemistry of solar cells.

Dr. Wolfgang Münch studied physics and graduated from the University of Konstanz in 1996. In 1999, he obtained his doctorate in 1999 with a thesis on the dynamics of liquids on structured substrates. Currently he is working as a senior scientist at the Laboratory for Micro-optics in Department of Microsystems Engineering of the University of Freiburg, Germany, with Prof. Hans Zappe. His scientific interests are focused to innovative applications of effects and materials from the field of soft matter science to microoptics, including electrowetting-based systems, tunable photonic crystals, and biophotonics.

Prof. Hans Zappe Born in Paris and raised in New York, Hans Zappe studied electrical engineering at the Massachusetts Institute of Technology (BSc & MSc, 1983) and at the University of California, Berkeley (PhD, 1989). He has worked at the IBM General Technology Division (Burlington, VT, USA) on silicon VLSI, at the Fraunhofer Institute for Applied Solid State Physics (Freiburg, Germany) on GaAs electronics and high-speed lasers and at the Centre Suisse d’Electronique et de Microtechnique (CSEM, Zurich, Switzerland) on integrated optical microsystems and surface-emitting lasers. Since 2000, Prof. Zappe has been professor of micro-optics in the Department of Microsystems Technology at the University of Freiburg, Germany, where he is also Dean of Studies. His current research specialties are in the areas of optical microsystems for medicine, tunable micro-optics and the use of novel optical materials.