Shape Oscillation and Internal Mixing in Sessile Liquid Drops Using Electrowetting-on-Dielectric (EWOD)

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ABSTRACT

Internal mixing within a sessile liquid drop can be significantly enhanced by means of so-called electrowetting-on-dielectric (EWOD), using an alternating electric potential. This is done experimentally by monitoring the coalescence and mixing of dyed liquid drops that are brought together by electrowetting actuation. The process is monitored using high-speed imaging and the extent of mixing with AC forcing after coalescence is tracked. Here, EWOD refers to manipulation of contact angle of a liquid drop on a surface by application of an electrical potential between the drop and an electrode embedded in the surface, separated from the drop by a thin dielectric layer. To understand the internal mixing, which appears to be driven by shape oscillations of the sessile drop, we also conduct fundamental studies of the shape oscillations of drops that are attached to a planar solid surface. Through image processing, we show that depending on the frequency of the applied alternating field, various modes of oscillation, including non-axisymmetric ones, may be excited.

Keywords: Microfluidics, Electrowetting, Droplets, Mixing

1 INTRODUCTION

Liquid droplets have been the focus of study in many applications such as electrostatic spraying, ink-jet printing, materials processing of melts in space, and nuclear physics. With recent developments in microfluidics and small scale (micro/nano-liter) discrete liquid handling techniques, liquid droplets and their manipulation continue to provide the basis for development of novel technologies [1]–[4].

Active mixing within sessile liquid drops has applications to speedup of reactions or biomolecular interactions in biological assays that can be conducted in microliter drops, such as mixing of reagents as well as amplification, detection and hybridization of DNA. For biological reactions which are limited by diffusion and mass transport, reduction of the reaction time by active mixing will play an important role in advancing the use of so-called 'digital' or droplet microfluidics in high-throughput technologies.

One of the most widely investigated topics in droplet fluid dynamics, with great potential in active mixing, is the problem of shape mode oscillations. Early theoretical work in this area focused on identifying the linearized shape modes and characterizing their natural frequencies and weak viscous damping rates [5]–[7]. Both large and small amplitude oscillations of liquid drops have now been investigated theoretically and experimentally. In order to create such oscillatory motions, several approaches have been tested. One such approach is acoustic excitation used, for instance, by Trinh and Wang [8]. This usually involves a second medium (mostly water and silicone oil) used for transmitting the acoustic energy and optical imaging of the drop. Other methods tested for droplet oscillation include break-up of laminar jets [9], electromagnetic levitation of metal drops [10] and mechanical excitation [11]. Azuma and Yoshihari [12] excited 3-D large-amplitude oscillations of a mercury drop using electrical excitation in low gravity using a drop tower. In their experiment, a sessile mercury droplet submerged in an electrolyte solution and in contact with a wire was excited over a wide range of frequencies and relationships between oscillation patterns and frequencies of excitation were studied theoretically and experimentally.

In this work, we use electrowetting-on-dielectric to excite shape oscillations of sessile drops and to achieve active mixing within the drops. EWOD is a recent method for manipulation of contact angle (i.e., wettability) of a liquid drop on a surface by application of an electrical potential between the drop and an electrode embedded in the surface, when that electrode is separated from the drop by a thin dielectric layer [1]–[4]. The emergence of contact line manipulation of small scale droplets by EWOD motivates us to revisit the problem of shape oscillations of sessile drops induced by contact angle change. This type of oscillatory motion has the unique capability of inducing small and large amplitude oscillation of a sessile drop resting on a solid surface without any additional requirements, e.g., second fluid, vibration of the solid wall, etc.

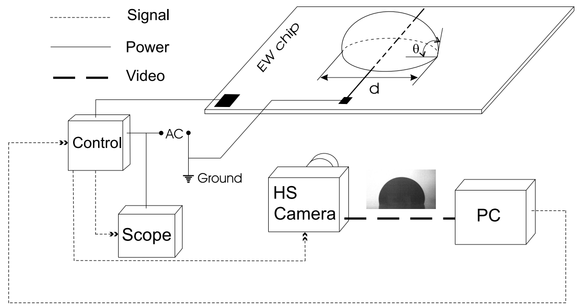

Figure 1: Schematic of the experimental apparatus with components and flow diagram of voltage, command signal and video signal. A snapshot of the drop is shown in the inset.

2 Experimental Setup and Methods

In order to study the shape oscillations of a sessile drop, the experimental setup shown in Fig. 1 was designed and assembled. The setup consists of an electrowetting chip, high speed imaging unit and electronic timing devices. Forcing in the from of AC signals was provided by a power-supply (California Instruments model 351-TL) with an output voltage of 60 V RMS and frequencies ranging from 1 to 4000 Hz. A 7-μL liquid droplet of 10% Potassium Chloride was positioned on the electrowetting chip using pipette dispensers.

Imaging was performed using a Photron Inc. FASTCAM-X 512 PCI 10-bit high-speed camera. Typical imaging speeds were 2000 and 4000 frames per second depending on frequency of oscillation. Imaging was performed within a 512×256 pixel window corresponding to a 3.8 by 1.9 mm field of view. The field of view was illuminated using two 150 watt fiber optic lamps illuminating a white screen behind the droplet resulting in high contrast between the droplet and the background screen. To avoid excessive heating of the droplet and light scattering from the surface of the drop, a tent-shaped shield made from regular paper was positioned on the droplet (not shown in Fig. 1).

To synchronize the excitation voltage, signal measurement and high-speed imaging during the oscillation of the droplet, a triggering software was developed using LabVIEW^{TM}. As a result of synchronized timing, values of excitation voltage and shape of the droplet (i.e., diameter, contact angle and maximum height) were obtained and correlated for the entire duration of the oscillations. Typical activation time of the electrodes was 0.1–0.2 ms corresponding to 400–500 acquired frames.

2.1 Electrowetting Platfrom

The solid substrate used for excitation of the droplet was made of a 2.5-cm long by 2.5-cm wide glass slide covered by 120 nanometers of Indium Tin Oxide (CG-51IN-3084 from Delta Technologies). A 1-μm layer of Paralyne C (Cookson Electronics) was deposited using chemical vapor deposition to create the dielectric layer. Chromium/gold electrical grounding lines were patterned with a photolithographic lift-off technique on the dielectric. The entire surface of the chip was coated with 1200Å Teflon-AF (601, 6%, Dupont) diluted in a perfluoro-compound (FC75, Acros) creating a hydrophobic surface. The entire surface of the ITO-coated glass was considered one single electrode on which droplets were positioned. Grounding was performed via 'grounding-from-below' as described in [13] through the ground line shown schematically in Fig. 1.

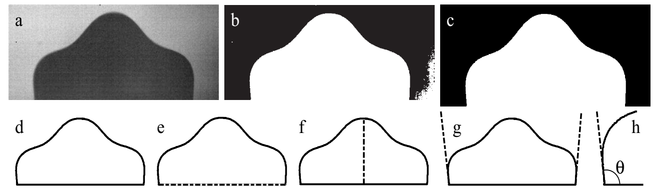

Figure 2: Boundary detection scheme applied to acquired images. Raw images (a) are converted to gray-scale (b) and threshold filtering to get rid of noise (c). Subsequent to detection of the droplet boundary (d), diameter, maximum height and contact angles are estimated (e) to (h).

2.2 High-Speed Imaging and Image Processing

The boundary of the droplet at any time (i.e., from an image frame) was determined through an automated image processing script. Fig. 2 demonstrates different stages of the image processing technique for individual frames. First, the frame was converted to a gray-scale image. Using a threshold value, the image was then converted to black-and-white based on a comparison of each pixel to the threshold. The border of the largest, contiguous, white region within this image was determined to be the boundary of the drop. To determine the width of the drop, a pixel sweep was performed for the left-most and right-most points on the boundary at the bottom of the drop. The difference in the resulting values represented the width of the drop. The height of the drop was determined to be equivalent to the value of upper-most point of the boundary which intersects the center of the width of the droplet. Contact angles were found by selecting all the left-most points and all the right-most points on the boundary of the drop between the bottom of the drop and 25 pixels ($\sim$ 0.2 mm) above the contact line. A linear fit was performed on both sets. The slope of each line represented the left and right side contact angles.

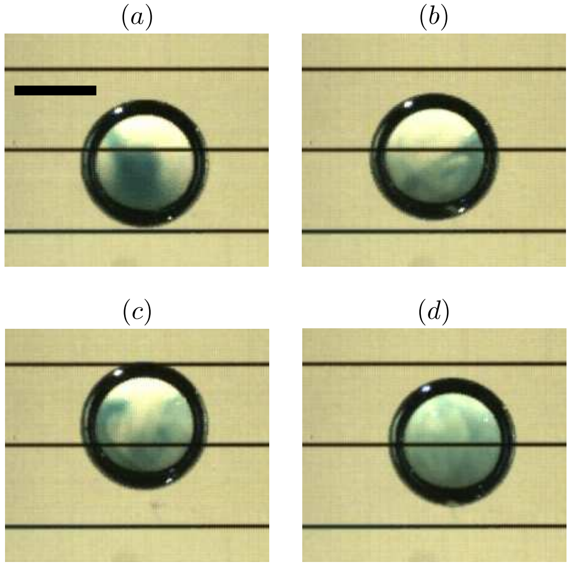

Figure 3: Bottom view of mixing of two 5-μL 10% KCl droplets using EWOD at (a) immediately after coalescence; (b) 2 seconds after coalescence; (c) 4 seconds after coalescence; and (d) 8 seconds after coalescence. One of the droplets was marked with food coloring. Scale bar corresponds to 1 mm.

3 RESULTS AND DISCUSSION

3.1 Internal Mixing

To investigate the effects of oscillatory motion on internal mixing, mixing inside two coalesced KCl drops was studied. Two 5-μL droplets were dispensed onto an array of electrodes and were made to coalesce following protocols explained in [13]. One of the drops was marked by green food coloring to better visualize the process. Subsequent to the coalescence of the droplets, a 50 V AC, 200 Hz voltage was applied to the four electrodes that the droplet was resting on. The four 1-cm by 1-cm electrodes formed a 2 by 2 square with the droplet located in the middle of the array. The voltage was maintained for 0.4 s on all four electrodes resulting in oscillatory motion of the drop. Snapshots in Fig. 3 show that oscillations of the droplet create strong internal flows resulting in an increase in the portion of the drop that appears to be infused with dye when viewed from the bottom. Another observation based on the snapshots in Fig. 3 is that the droplet did not maintain its original post-coalescence position (i.e., center of the 2 by 2 array of four electrodes) as electrodes were charged and discharged. The random translation of the drop proved to be an effective way for creating flow inside the drop in addition to that induced by oscillatory motion alone, consistent with [14].

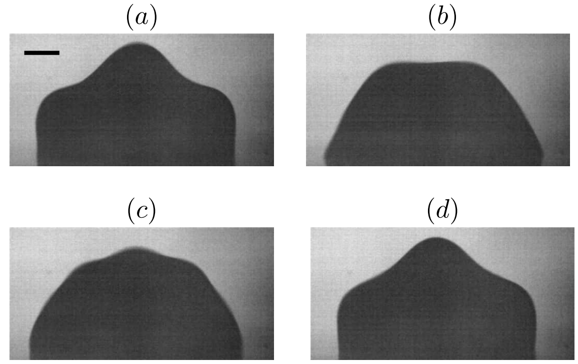

Figure 4: Snapshots of a 7-μL KCl droplet oscillating with a 60 V AC forcing at 180 Hz. Snapshots are 1 ms apart. Scale bar corresponds to 0.5 mm.

3.2 Shape Modes of Droplet Oscillation

Visualization of droplet oscillation via high-speed imaging over forcing frequencies ranging from 30 to 300 Hz resulted in observing both axisymmetric and non-axisymmetric motions. The non-axisymmetric oscillations were detected within the forcing frequency range of 90 to 120 Hz while the oscillations remained essentially axisymmetric for the rest of the frequencies tested. Fig. 4 shows snapshots of a 7-μL droplet excited with a 60 V AC voltage at 180 Hz forcing frequency. For this frequency, axisymmetry of the oscillations was verified by monitoring the motion of the drop from a bottom view (similar to Fig. 3.)

Applying the image processing techniques described in section 2.2 to each frame of the captured movies, time variations of droplet diameter d, contact angle α, and maximum height h, depicted in Fig. 5, were obtained. Spectral analysis of these signals results in graphs of the type presented in Fig. 6. As seen therein, under an AC forcing of frequency 180 Hz, the primary response of the drop, as characterized by $d(t)$, $h(t)$ and $\alpha(t)$, occurs at a frequency of 360 Hz. This is expected since electrowetting is known to be an effect which is proportional to the square of the voltage difference. Thus an AC voltage forcing of 180 Hz effectively drives the drop at a frequency of 360 Hz. In Fig. 6, one can also discern 'superharmonic' components in some of the signals at 720 Hz.

As the forcing frequency is swept over a range of values, response of the drop under each forcing condition can be monitored by carrying out an analysis of this kind. As expected, by sweeping through a range of frequencies, it is possible to excite various shape modes of the sessile drop, with higher mode numbers appearing at higher forcing frequencies. Under high-amplitude excitation, multiple mode numbers may be present in the

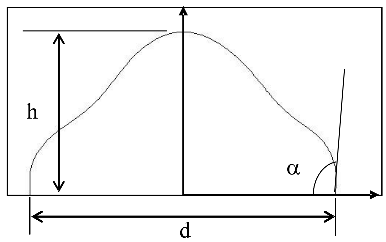

Figure 5: Basic geometric parameters of the droplet obtained from image processing of the individual frames.

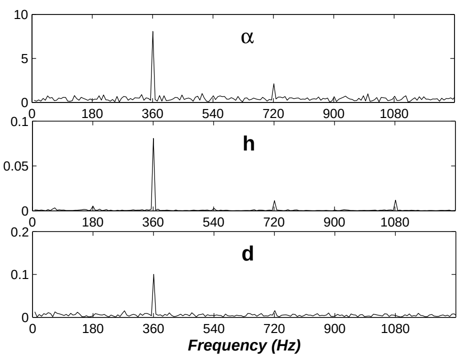

Figure 6: Power spectrum, obtained via Fast Fourier Transform, of the contact angle, droplet height and diameter time series.

drop shape response at one forcing frequency. This is evident by comparing some of the frames in Fig. 4. We have also observed nonlinear effects in such oscillations including superharmonic and subharmonic resonances in which the drop's primary response is at twice or half the forcing frequency provided by the square of the voltage. Videos of these oscillations, captured by the high-speed camera, can be found at our laboratory's website [15].

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