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Electric field-induced oscillation of sessile droplets

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We report a novel oscillatory behavior of a sessile droplet on a hydrophobic surface. The droplet was placed on an electrode with a hydrophobic surface and close to, but not touching, a second needle-like electrode. The change in the contact angle was observed only when the droplet oscillated. In a traditional electro-wetting experiment, however, the contact angle decreased immediately when an alternating current electric field was applied. In addition, the non-contact mode gave rise to a true sessile condition of the droplet, whose oscillation amplitude was not linearly proportional to the driving voltage but reached a maximum value.

oscillation, sessile droplet, ac electric field, hydrophobic surface

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The recent surge of interest in micro- and nano-systems has focused increasing attention on small-scale liquid droplets [1–3] and their manipulation, e.g. droplet transport, separation and fusion. Droplet oscillation is an intriguing phenomenon, which has many potential applications, including the measurement of surface tension, viscosity [4] and contact angles [5], the establishment of internal mixing within droplets [6,7], improving reaction rates and biomolecular interactions in biotechnology and combinatorial chemistry [8], and changing the shape of droplets used in liquid lenses [9] and reflective displays [10]. The oscillations of droplets have been studied extensively since the nineteenth century. In 1879, a mathematical model for the small-amplitude oscillation of an isolated inviscid droplet was originally given by Rayleigh [11] and later extended by Lamb [12] to account for the external fluid surrounding the droplet. They reported that there are an infinite number of discrete oscillation modes and calculated the corresponding oscillation frequencies. In contrast to the oscillation of isolated droplets, the analytical solution for the oscillation of a droplet in partial contact with a substrate was proposed by Strani et al. [13]. It was found that an oscillation movement of the center of the droplet mass along the axis of symmetry occurred for a sessile droplet. This is not possible for an isolated droplet due to momentum considerations. Thus the oscillation of a sessile droplet is basically different from that of an isolated droplet. Recently, the investigation into the behavior of a hemispherical droplet on a solid plate was carried out by Lyubimov et al. [14] providing a profound understanding of droplet oscillation. In this work, the equation of the frequency of a sessile droplet is used and expressed as

$$f_n^2 = \sigma \Big/ \left(4\pi^2 \rho R^3 \lambda_n\right), \tag{1}$$

where R is the radius of the droplet, ρ is the density of the liquid, σ is the interfacial tension coefficient, and λ_n is the eigenvalue for mode n corresponding to the nth Legendre polynomial.

Many methods can be used to create droplet oscillation, e.g. electromagnetic force [15] and acoustic and mechanical excitation [13,16]. Electric fields were also used to actuate the oscillation of sessile droplets. Bormashenko et al. [17] and Takeda et al. [18] reported the behavior of sessile droplets exposed to a constant and uniform electric field. Shape variations in isolated droplets in both static and time-periodic electric fields have been investigated theoretically

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and experimentally [19,20]. More recently, the existence of sessile droplet oscillations actuated by an ac electric field was reported [21,22]. These oscillations were actuated by electro-wetting with a wire inserted into the droplet. Hydrodynamic flows have also been observed in ac-electro-wetting [7]. However, the wire used in the experiments limits the true sessile condition. In this work, we report on a novel method of actuating the oscillation of a sessile droplet, which is not related to the electro-wetting phenomenon of droplets on a solid surface. We found that droplet oscillations could be induced without piercing the droplet by applying a vertical ac electric field between a bottom electrode (substrate) and a suspended needle-like electrode (Figure 1). The relationship between the frequency and the droplet radius and between the amplitude of droplet oscillation and the applied voltage were studied experimentally.

1 Experimental setup and method

The schematic diagram for the experimental setup is shown in Figure 1(a). Liquid droplets of deionized water (conductivity 0.061 μs/cm) were positioned on the substrate. To reduce the influence of gravity on the droplet deformations, the droplet diameters in this experiment were adjusted to be smaller than the capillary length $\kappa^{-1}=(\sigma/\rho g)^{1/2}$ (g is gravitational acceleration). The capillary length for water is approximately 2.7 mm. The substrate was a copper electrode covered with a 10 μm thick hydrophobic film of Teflon. The static contact angle of the water droplet on the hydrophobic surface was about 130°. To avoid air bubbles between the copper and Teflon, we wiped the hydrophobic test surface clean with cotton dipped in alcohol, and then blew it dry. A hollow, steel needle electrode with an outer diameter and thickness of 0.56 mm and 0.13 mm respectively was used and suspended above the droplet. The distance from the needle tip to the lower surface was defined as H (Figure 2(a)). This is a typical device for the combination of needle and plate electrodes in electro-wetting experiments. The application of this device will lead inevitably to the inhomogeneity of the electric field. We designed this experimental device for the purpose of comparing our experiments with typical electro-wetting experiments. Unlike the oscillation induced by electro-wetting, in which the droplet was disturbed by a connecting conductive wire, the droplet in our case was in a truly sessile condition. An ac voltage (Figure 1(b)) was supplied by a function generator (model AFG3022B, Tektronix Inc., Beaverton, OR, USA) which generated a sinusoidal signal with frequency ranging from 1 μHz to 25 MHz. However, because the maximum output of 10 V did not meet the requirements of our experiment, the voltage signal between the needle electrode and the copper electrode was further amplified (model HA-800, Pintek Electronics Co. Ltd., Taiwan, China).

In this work, the voltage, V_{pp}, was defined as the peak-to-peak value of the ac voltage. Images of droplet oscillation were captured by a charge-coupled device (CCD) camera at 25 frames per second and transferred to a computer for subsequent analysis. We recorded the frequency of droplet oscillation exhibiting the greatest amplitude as the

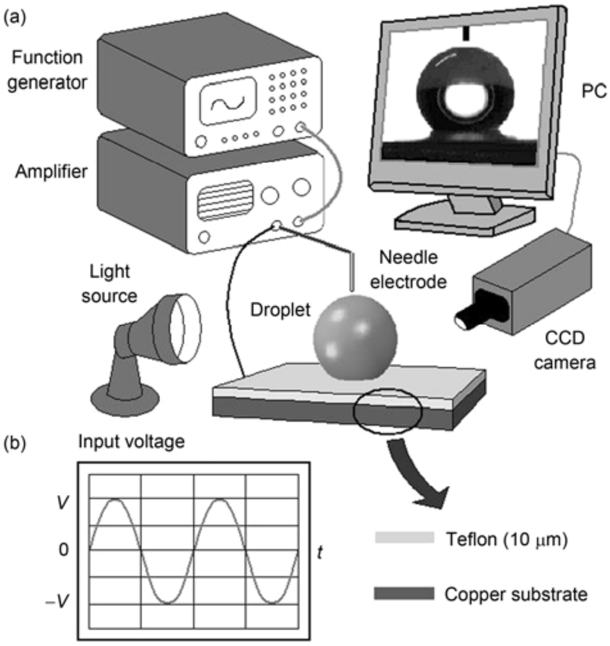

Figure 1 Schematic diagram of the experimental setup and voltage signal used in the experiment. (a) Experimental setup. A droplet was positioned between a copper substrate covered with a hydrophobic layer (Teflon) and a needle-like electrode (steel) suspended above it. (b) Input ac voltage signal generated by a function generator.

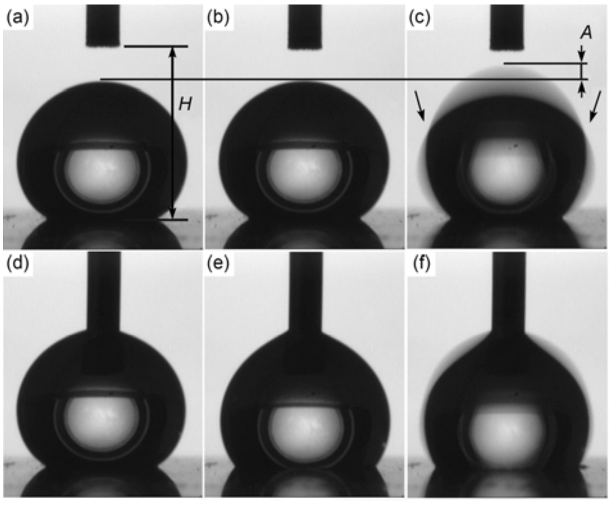

Figure 2 Comparison of a droplet oscillating in non-contact mode and in contact mode. (a)–(c) In non-contact mode ($H = 2.693$ mm), an obvious oscillation of the droplet was observed when the frequency of the ac voltage ($V_{pp} = 360$ V) reached 30 Hz, but no discernable change occurred in the static contact angle. (d)–(f) In contact mode, a lower voltage ($V_{pp} = 200$ V) was used to prevent breakdown of the Teflon surface and the oscillation was found at 49 Hz; there was a clear change in the static contact angle caused by electro-wetting (e).

resonance frequency. The value of the oscillation mode *n* was equal to the number of node points at the resonance frequency, which was indicated by the arrows in Figure 2(c). Because the frequency of droplet oscillation was higher than that of the CCD camera, there would be ghosting in the captured images. We defined the distance between the highest point of the ghosting image and the highest point of the original droplet as the oscillation amplitude *A* (Figure 2(c)). The oscillation frequency of the droplet equaled the driving voltage frequency controlled by the function generator, which was demonstrated by the oscillation behavior of a sessile droplet driven by 1 Hz ac electric field. Thus, with a similar conclusion to the work of Ko et al. [23], we believe that the droplet could oscillate with the same frequency as that of the driving voltage. During the whole experiment, temperature and relative humidity were controlled in the range (20±0.5)°C and (85±5)%, respectively.

2 Results and discussion

2.1 Comparison of a droplet oscillating in two modes

Initially, we compared oscillation shapes and static contact angles of sessile droplets in non-contact and contact modes. Several side view images of a droplet are shown in Figure 2. A 10 μL droplet was positioned between the needle electrode and the substrate (Figure 2(a)). There was no obvious change in the droplet shape when a low frequency (10 Hz) ac voltage of $V_{pp} = 360$ V was applied (Figure 2(b)). Because the frequency of the driving voltage was far from the frequency of the droplet resonance, the contact angle was time-independent and could be measured as in electro-wetting experiments [4]. Meanwhile, the measurement was also carried out under a higher voltage to eliminate the possibility that the change in contact angle was too small to be detected. The change, however, was not obviously observed even when an 800 V voltage was applied. The droplet was then swept with an applied ac electric field ranging in frequency from 1 Hz to 300 Hz. A significant oscillation of the droplet with $n=2$ was observed at 30 Hz (Figure 2(c)). We explained the effects using the polarizability of the liquid and inferred that the rapid and alternative deformation of the droplet surface caused by the electric field force led to the oscillation. Subsequently, we turned off the supply voltage and moved the needle down into the droplet (Figure 2(d)). To prevent the Teflon surface from being broken down, the voltage was reduced to $V_{pp}=200$ V in the contact mode. In contrast to the droplet in the non-contact mode, a 10° decrease in the static contact angle was associated with electro-wetting of the droplet and was clearly visible when a voltage with a low frequency (10 Hz) was applied (Figure 2(e)). In this case, the droplet oscillation was not observed until the frequency was increased to 49 Hz (Figure 2(f)). Furthermore, the oscillation shape at 49 Hz changed due to the additional constraints imposed by the needle electrode. It can be seen that the electric field force used in our experiment gave rise to a true sessile condition, and that the droplet oscillation had a better, more symmetrical, appearance than that in the contact mode. Meanwhile, the droplet was initially stretched in a vertical direction in the non-contact mode when the voltage was applied. In contact mode, however, the droplet was stretched in a horizontal direction due to the reduction of the contact angle. Therefore, the experiment in our case is essentially different from previous electro-wetting experiments.

2.2 Effects of droplet radius on the oscillation frequency

To better understand the nature of sessile droplet oscillation induced by an electric field force, the dependence of the oscillation frequency on the droplet radius was investigated. When an ac voltage ($V_{pp} = 800$ V) was applied to the droplet positioned on the Teflon surface, oscillations were observed over a range of frequencies with the same mode ($n = 2$). This phenomenon is similar to that reported in [24]. For one mode (e.g. $n = 2$), the frequency of the droplet oscillation exhibiting the greatest amplitude was recorded as the oscillation frequency. The relationship between oscillation frequency and droplet radius corresponding to $n = 2$ is shown in Figure 3 and compared with that of the theoretical prediction using eq. (1). It can be seen that the measured oscillation frequencies generally agree with the theoretical prediction. The discrepancy between the theoretical prediction and the experimental measurements may result from the ideal assumption in the theory that the droplet is axisymmetric and inviscid [13]. In fact, the droplet positioned on the substrate can neither be an inviscid fluid nor exactly axisymmetric in shape.

2.3 Effects of voltage on the oscillation amplitude

The correlation between oscillation amplitude of the droplet

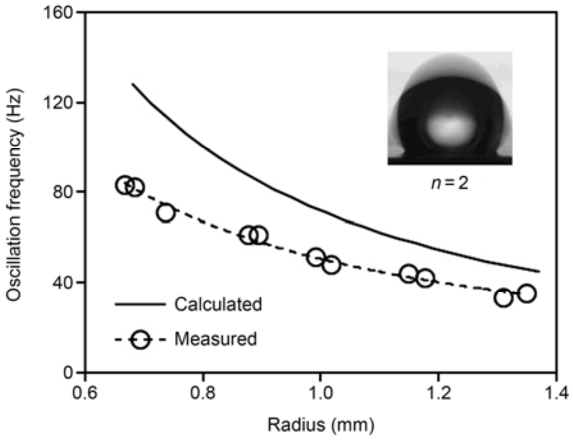

Figure 3 Measured droplet oscillation frequency versus droplet radius, compared with that of the theoretical prediction for mode $n = 2$. Input voltage $V_{pp} = 800$ V.

and the applied voltage was also studied. In one experiment, the oscillation amplitude was measured as a function of voltage for 6 μL, 7 μL and 9 μL of droplets with a fixed value of $H = 2.281$ mm (Figure 4(a)). Larger oscillation amplitudes were obtained with increasing droplet volumes. In another experiment, the influence of H (2.396 mm, 2.454 mm and 2.578 mm, respectively) on the oscillation amplitude is shown in Figure 4(b). For droplets with the same radius ($R = 1.2$ mm), a small value of H was needed to create a large oscillation amplitude. In addition, a unique phenomenon was observed in this group of experiments, as shown in Figure 4. The oscillation amplitude initially increased with voltage, before reaching a maximum value at $V_{pp} = \sim 400 - 500$ V. Finally, the amplitude decreased continuously with further increases in voltage. Although the exact reason for the final decrease of the oscillation amplitude is still unknown, we suggest that the non-uniform electric field could be responsible for this phenomenon. Clearly, a more detailed investigation is needed in the future.

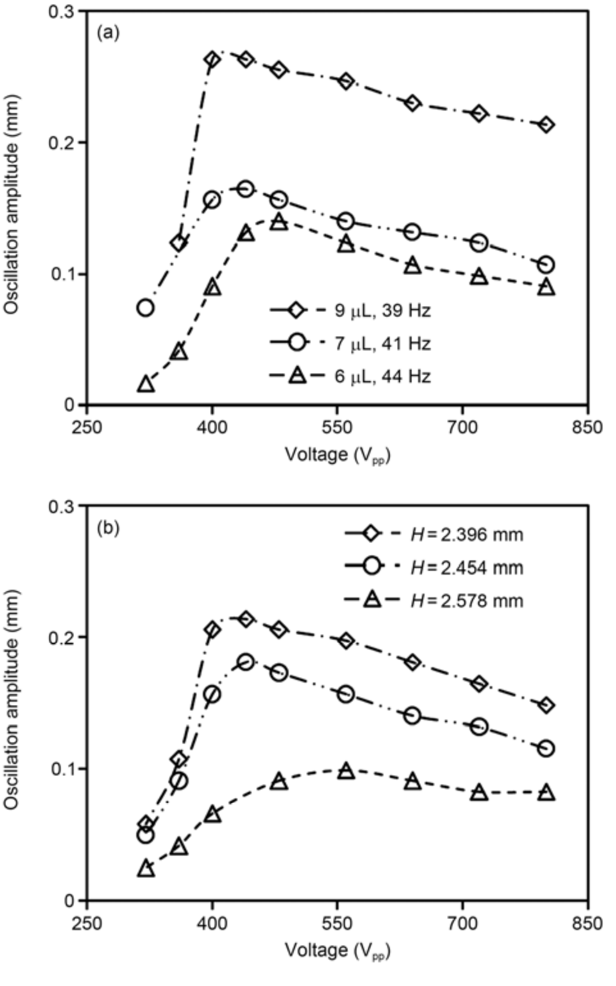

Figure 4 Oscillation amplitude of droplets versus the applied voltage in the mode $n = 2$. (a) For 6 μL, 7 μL and 9 μL of droplets with a fixed value of $H = 2.281$ mm. (b) For different values of H (2.396 mm, 2.454 mm, 2.578 mm, respectively), with the same radius $R = 1.2$ mm and frequency $f = 36$ Hz.

3 Conclusions

The behavior of a sessile droplet in an ac electric field was studied. To the best of our knowledge, there have been few previous publications concerning the oscillations of a sessile droplet actuated by ac electric field force. In our experiment, for the purpose of comparing our experiments with previously reported electro-wetting experiments, a device consisting of a needle and plate electrodes was used. However, a different droplet deformation was observed. The effects can be explained using the polarizability of the deionized water. An electric field exerted a force on the surface of the charged droplet. Rapid and alternative vertical deformation of the droplet surface controlled by ac electric fields then led to oscillation. The oscillation frequencies corresponding to the droplet radius were measured, and showed good agreement with the theoretical predictions. A novel observation was that the droplet oscillation amplitude was not linearly proportional to the driving voltage but exhibited a maximum value. Our findings may provide important inspiration for microfluidic applications. Droplet transport, mixing and integration may be achievable using a system consisting of a maneuverable needle electrode and a conductive substrate with a hydrophobic coating. This may, in turn, be instrumental in the design and fabrication of future digital microfluidic systems.

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