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An electromechanical interpretation of electrowetting

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Abstract
Electrowetting on dielectric-coated electrodes involves two independently observable phenomena: (i) the well-known voltage dependence of the apparent contact angle and (ii) a central electromechanical force that can be exploited to move and manipulate small liquid volumes on a substrate. The electromechanical force does not depend upon field-mediated changes of the contact angle; it is operative even if the liquid surface is constrained. Forces on liquid volumes may be determined using capacitance or the Maxwell stress tensor with no reference made to liquid surface profiles. According to this interpretation, a nonlinear mechanism manifesting a voltage threshold is responsible for both contact angle saturation and the observed clamping of the electromechanical force.

1. Introduction

When a droplet of conductive liquid is placed on a horizontal, dielectric-coated electrode and a voltage difference then applied between them, the sessile droplet flattens and spreads[1]. This well-known phenomenon, commonly referred to as *electrowetting on dielectric* (EWOD), is the basis of a number of potentially important microfluidic technologies, including laboratory-on-a-chip schemes [1], liquid dispensers [2, 3], voltage-controlled fluidic switches [4], electrostatically focused liquid lenses [5, 6], optoelectronic couplers [7] and electronic paper displays [8]. There are actually two exploitable manifestations of a conductive liquid's response to the electric field: (i) an observed change of the apparent contact angle θ_c made by the liquid with the solid surface and (ii) a net electrostatic force producing displacements of the center of mass (CM) of small liquid volumes. Authors introducing and describing applications for EWOD often ascribe the second of these (displacements and motions) to the first (contact angle effects).

The contention of this paper is that such an attribution is erroneous. Instead, it is argued that the force acting on the center of mass does not depend on contact angle at all and is more accurately described as electromechanical in nature. A simple thought experiment is invoked to dispel the link usually assumed to exist between contact angle and displacement-causing electromechanical forces. The alternate physical interpretation is offered that changes in the contact angle and net CM displacements are distinct observables resulting from the influence of the electric field on the liquid. The utility of these distinctions is less readily apparent in the hydrostatic equilibria, but emerges clearly in the practical case of EWOD-induced, transient, microhydrodynamic flows. The electrical force that causes CM displacements in dynamic EWOD systems can be derived from a general, electromechanical model based on capacitance or, alternately, the Maxwell stress tensor.

2. Contact angle model

For the electrowetting demonstration of figure 1, the applied voltage V_{drop} alters the contact angle according to the following relationship [9]:

$$\cos\theta_E = \cos\theta_0 + \kappa_d\varepsilon_0 V_{drop}^2 / 2\gamma d \qquad (1)$$

where θ_E and θ_0 are the static contact angle with and without voltage, κ_d and d are the dielectric constant and thickness of the dielectric layer, all respectively, $\varepsilon_0 = 8.854 \times 10^{-12}$ F m^{-1} is the permittivity of free space and γ is the liquid/air surface tension. Equation (1), derived from energy considerations,

[1] Electrowetting is also observed for aqueous solutions in contact with bare, metallic electrodes, but attention here is restricted to the situation of electrodes coated with a thin dielectric layer that blocks electrolysis and facilitates strong electromechanical interactions essential in microfluidic applications.

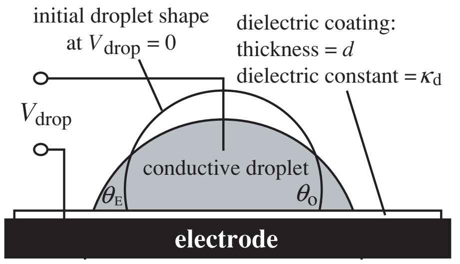

Figure 1. Electrowetting behavior of a sessile droplet of conductive liquid on a dielectric-coated electrode. An applied voltage between the droplet and the electrode caused the droplet to spread on the substrate.

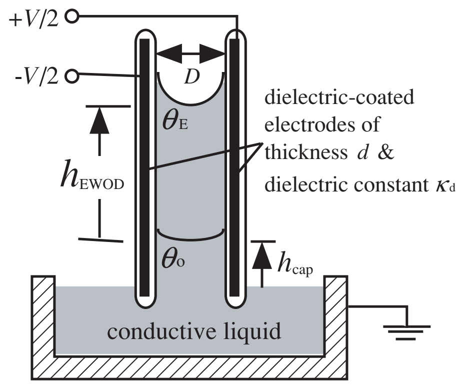

Figure 2. Height-of-rise experiment with coated electrodes and conductive liquid. Application of voltage lifts the liquid column upward between the parallel electrodes and reduces the apparent contact angle.

predicts with acceptable accuracy the observed dependence of the static contact angle upon voltage up to a threshold where it is observed that the contact angle stops changing [10]. This important limit, referred to as contact angle saturation, is discussed in a separate section below. Note that the saturation threshold is not defined by the fully wetted condition: $\cos(\theta_E) = 1$. Rather, saturation typically occurs in the range of 30°–60°.

The *apparent* connection of the contact angle effect to the net force acting on the center of mass of a liquid volume—the point disputed in this paper—is exemplified by considering the problem shown in figure 2. Two vertical, parallel, dielectric-coated electrodes are partially immersed into a pool of a conductive liquid such as water. We assume that the spacing D is much greater than the thickness of the dielectric, that is, $D \gg d$. Also, the length and width of the electrodes are large compared to D, so that fringing field effects may be ignored. Then, if $V = 0$, the static, capillary height-of-rise of the liquid column between the electrodes is

$$h_{\mathrm{cap}} = 2\gamma \cos\theta_0/\rho g D. \quad (2)$$

In equation (2), ρ is the liquid mass density and $g = 9.81\ \mathrm{m\ s^{-2}}$ is the acceleration due to gravity. Applying voltage V to the electrodes, the liquid column rises an additional amount h_{EWOD} and the contact angle decreases in accordance with equation (1) with $V = 2V_{\mathrm{drop}}$. In the experiment depicted, bipolar voltage is used and the liquid is grounded through a conducting container. One way to determine h_{EWOD} is to use a force diagram that balances the net surface tension 'force' per unit length at the contact line against gravity. According to this notion, the voltage-induced decrease in the contact angle pulls the liquid column up by an additional amount proportional to the difference in the cosines of θ_E and θ_0. Combining equation (1) and (2), we have

$$h_{\mathrm{EWOD}} = 2\gamma(\cos\theta_E - \cos\theta_0)/\rho g D = \kappa_d \varepsilon_0 V^2/4\rho g d D. \quad (3)$$

While equation (3) gives an answer that correlates with experiment [11], the attribution of a ponderable force per unit length to the surface tension is subject to the criticism of Rouse, who stated [12]

> ... there are so many physical inconsistencies in the surface tension concept that the continued designation of the quantity [γ] as the coefficient of surface tension is, to say the least, misleading[2].

Buehle and Mugele provide additional evidence calling into doubt the validity of using the surface tension concept to determine h_{EWOD} [13]. They computed the liquid profile near the contact line by enforcing electromechanical equilibrium at all points along the interface while simultaneously seeking the minimum energy condition. Their result is that, within distance $\sim d$ of the dielectric surface, the liquid contact angle asymptotically approaches θ_0 from the apparent value of θ_E at larger distances. If the angle made by the liquid at its actual contact with the dielectric-coated surface is θ_0, then the force diagram argument commonly invoked to explain the liquid rise as a consequence of θ_E becomes less persuasive. As shown below, the change in the angle from θ_0 to θ_E is not in fact the driving force of the liquid rise, but an independent consequence of the strong electric field near the three-phase contact that influences the shape of the meniscus.

3. Electromechanical model

The method of lumped parameter electromechanics, based on a virtual work principle applied to conservative systems, offers an alternate way to derive h_{EWOD} by direct determination of the force of electrical origin. As long as h_{EWOD} is much greater than the capillary meniscus height, this approach is insensitive to the profile of the meniscus. The system capacitance of the structure in figure 2 can be expressed as a function of the height of the liquid column h about the capillary equilibrium at $z = h_{\mathrm{cap}}$.

$$C(h) \cong \kappa_d \varepsilon_0 W h/2d + \text{constant} \quad (4)$$

$W \gg D$ is the width of the electrodes. The assumption $d \ll D$ guarantees that the capacitance is dominated by contributions from the liquid-covered areas between the coated electrodes.

We use the coenergy function $W'_e = C(h)V^2/2$ to evaluate the upward-directed force of electrical origin [14].

$$f^e = \left.\frac{\partial W'_e}{\partial h}\right|_V \cong \frac{\kappa_d \varepsilon_0 W}{4d} V^2. \quad (5)$$

The Maxwell stress tensor, an alternate way to calculate f^e, gives the same result, again without reference to the shape

[2] Rouse goes on to state in the same paragraph of [12] that 'the quantitative evaluation of surface phenomena by means of the surface tension concept yields perfectly accurate results, despite the erroneous physical picture upon which it is based'. But in the presence of electrical fields, even this reprieve is no longer true, as shown by the gedanken experiment presented in this paper.

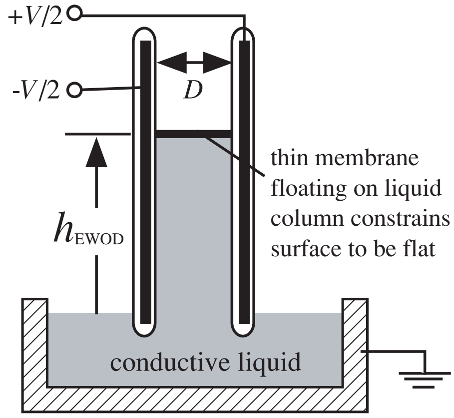

Figure 3. Thought experiment with the top surface of the liquid column between parallel electrodes constrained by a very thin, rigid, electrically insulating membrane that slides up and down without friction. The meniscus remains flat as the column rises in response to increasing the voltage.

of the meniscus [15][3]. Equating f^e to the gravitational force exerted on the elevated column of liquid in figure 2 yields the same expression for h_{EWOD} given by equation (3). Lumped parameter electromechanics avoids a problem inherent in the surface tension approach evident in equation (1) if the voltage is increased to the point where $\cos\theta_{\mathrm{E}} > 1$. This non-physical result is a clear demarcation of the limits of the contact angle model's predictive ability (equation (1)). On the other hand, the electromechanical model presented here does not have this problem.

Classical electromechanics ignores the shape of the meniscus yet gives the same result for the height-of-rise as obtained from the force diagram model using the surface tension concept. But this agreement does not constitute proof that electric-field-induced contact angle changes and CM displacements are separate and distinct observables. Instead, proof of this contention is provided by a rather simple gedanken experiment. Imagine the same apparatus of figure 2, but now with a very thin, rigid membrane that floats atop the surface of the liquid column and prevents formation of the usual curved meniscus, see figure 3. Let the membrane be just slightly less dense than the liquid, electrically insulating and non-polarizable, and assume that it slides without friction up and down between the electrodes. In form, the capacitance of this new system is identical to equation (4). Therefore, the force of electrical origin f^e on the column and the height-of-rise h_{EWOD} must be the same. So, despite the fact that the liquid surface is constrained and that the contact angle is fixed, the liquid column still must rise. One concludes that the central force responsible for pulling the liquid upward against gravity does not depend at all on electric-field-induced changes of the contact angle. This conclusion is fully consistent with Hendriksson and Eriksson, who argue from thermodynamic considerations that the capillary height-of-rise and the change in the curvature of the meniscus are distinct consequences of energetically favorable wetting of the solid surface by the liquid, in other words, that it is not the curvature of the meniscus that draws the liquid upward [16].

The above argument has an interesting analogy in the case of the force exerted on dielectric media situated between uncoated, parallel electrodes. Pellat's original 1895 experiment [17] used dielectric liquid; the usual analysis of this problem ignores any distortion of the meniscus. The net upward-directed electrical force can be shown to be identical to that exerted on a solid slab that is free to slide up and down between the electrodes. Many classic texts present this electromechanical problem as an example of the ponderomotive force on dielectrics [18].

The origin of the net electromechanical force pulling the liquid upward is the strong, non-uniform electric field near the contact lines on the electrodes. This field, normal to the top surface of the conductive liquid column, induces free electric surface charge and pulls it upward. The situation is identical to the case of a solid, conductive slab between two coated electrodes. It is easy to explain why the *net* value of this force does not depend on the details of the liquid profile near the contact. Imagine a small, virtual displacement of the column δh changing the capacitance by the amount $\delta C \cong (\kappa_{\mathrm{d}}\varepsilon_0 W/2d)\delta h$. This capacitive increment, proportional to the coenergy increment for the system at constant voltage, is not sensitive to the shape of the meniscus.

4. Saturation

Welters and Fokkink discovered that the liquid height-of-rise measured in an apparatus similar to figure 2 exhibits a voltage threshold, above which the column ceases to rise further [11]. It was found that this threshold correlates convincingly to the contact angle saturation exhibited by sessile droplets on identical dielectric coatings. In light of the argument presented above, the most reasonable interpretation of the correlation is that a nonlinear mechanism, or possibly several mechanisms, is responsible for *both* contact angle saturation and clamping of the CM force.

Several candidates have been proposed in explanation of contact angle saturation, including disruption of the liquid interface at the contact line leading to droplet ejection and/or corona discharge [9, 19, 20] and charge injection into the dielectric [21, 22]. Any or all of these mechanisms could simultaneously influence the contact angle and the electromechanical force. Recently reported transient height-of-rise and dynamic contact angle measurements, made with the parallel-electrode apparatus of figure 2, have revealed no clear link between the upward motion and contact angle changes [23]. Instead, force clamping manifests itself abruptly as the rising liquid column starts to slow down. The essential point is that there is no justification for assigning contact angle saturation as the causative agent in clamping the CM force.

5. Conclusion

Simple considerations have shown that the force of electrical origin resulting in displacement of the center of mass of a liquid volume does not depend on changes of the liquid contact angle. Instead, CM motions and contact angle changes are best regarded as independent observables. If the liquid interface

[3] The Korteweg–Helmholtz force density formulation used by Stratton and its associated stress tensor are very convenient for the case of incompressible liquids.

is unconstrained, then the *apparent* contact angle (far from the dielectric layer) assumes a value consistent with a balance of forces predicated on the surface tension concept. In this case, the dependence of contact angle on voltage is merely a manifestation of hydrostatic equilibrium. But if the surface is mechanically constrained, this consistency is neither required nor observed, and the central force is unchanged.

Despite the fact that electric-field-induced changes in the contact angle and center of mass displacements of liquid volumes by a non-uniform electric field are distinct observable phenomena, both have come to be encompassed by the term *electrowetting on dielectric* (EWOD) through common usage. For this reason, it becomes all the more important to distinguish between the two phenomena. Most microfluidic applications of electrowetting exploit the net force on the liquid masses to transport small volumes from point to point. Examples are laboratory-on-a-chip schemes, droplet dispensers, fiber-optic components and fluidic devices. In virtually all these cases, liquid displacements can be modeled in terms of a capacitance change; the appropriate derivative of capacitance gives the electrical driving force, which then can be used in an equation of motion. This electromechanical modeling approach is favored for its simplicity and generality. In particular, the Maxwell stress tensor successfully accounts for the strong frequency dependence of the height-of-rise in the parallel-plate experiment of figure 2 [24, 25].

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