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Interface Deformation and Lippmann-Young equation in Electrowetting

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전기습윤현상에서의 계면 변형과 Lippmann-Young 식

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Abstract

For the system of a droplet on an insulator-coated electrode, the Lippmann-Young equation is derived by considering the deformation of interface near the three-phase contact line. The equation governing the deformation of interface, which describes the local balance of the Maxwell stress and the capillary pressure, is integrated along the interface. The integration leads to the Lippmann-Young equation which is shown to represent the macroscopic balance of horizontal force acting on entire meniscus. Young's angle is assumed to be not affected by the Maxwell stress. The meaning and validity of the assumption are discussed.

1. Introduction

The electrical control of wettability, which is called the electrowetting, is a versatile tool for handling of micro- and nano-liter droplets. The most commonly used configuration is the so called electrowetting-on-a-dielectric (EWOD) in which a thin insulating layer is inserted between the liquid and the counter electrode to prevent the current flow. The electrowetting can be used as a very fast and efficient means to handle nearly any kind of droplets with a relatively low electrical potential and power consumption. Potential applications of the electrowetting have been demonstrated for the optical switch, variable focal lens, micro-pump, micro-mixer, and electronic display (see ref. 1, and references therein).

The following Lippmann–Young equation (LY equation) has been successfully employed in correlating empirical results on the apparent contact angle (θ) with external voltages (V) [2,3]:

$$\cos\theta = \cos\theta_Y + \frac{\varepsilon V^2}{2\gamma d} \qquad (1)$$

Here, θ_Y denotes the contact angle when $V = 0$ (i.e., Young's contact angle), ε the electric permittivity of the insulating layer beneath the droplet, γ the interfacial tension between the droplet and the surrounding fluid, and d the thickness of insulating layer. Above a certain voltage, the LY equation becomes invalid due to the occurrence of contact-angle saturation. Although several mechanisms have been suggested [1], there is no commonly accepted theory on how the contact angle saturation occurs.

The LY equation was derived by way of the minimum energy principle of thermodynamics. [1] Later, Kang [3] has shown that the electrowetting is a direct consequence of the concentrated Maxwell stress, in which the stress is concentrated within a distance of O(d) from the three-phase contact line (TCL). Kang approximated the contact line as a straight line to obtain the analytical solution for the electric field. He recovered the LY equation by applying the force-balance condition at the TCL. Jones[4] and Kang et al.[5] have shown analytically that the horizontal component of the electrical force, which is the so called the electrical wetting tension, is independent of the interface shape. The Maxwell stress acting on an interface causes the deformation of interface in the distance of O(d) from the TCL, as manifested by a recent experiment of Bienia et al.[6] The interface deformation caused by the Maxwell stress has been analyzed numerically by Buehrle et al.[7] and Papathanasiou and Boudouvis,[8] and analytically by Bienia et al.[6] Buehrle et al.[7] analyzed the interface deformation by an iterative numerical method and suggested that the microscopic contact angle at the contact line approaches Young's angle, being independent of the Maxwell stress. Bienia et al.[6] deduced a similar conclusion based on the method of conformal mapping. It has been questioned whether the deformation of interface could have any effect on the validity of the LY equation.[7,8] Buehrle et al.[7] obtained the contact angle which gives a consistent result to that of the LY equation down to 5^o. Papathanasiou and Boudouvis[8] repeated the analysis of Buehrle et al., but their result showed a substantial deviation from that predicted by the LY equation. Their result is certainly contradictory to that of Buehrle et al. and the macroscopic result of Jones[4] and Kang et al.[5]

The LY equation has been derived in several approaches, which includes the thermodynamic approach4 and the approach based on the macroscopic force balance at the TCL. [2-4] As demonstrated previously, [5-7] the change of the macroscopic contact angle in electrowetting is associated with the deformation of interface. It is expected that the LY equation can be derived in an explicit form considering the deformation of interface, under some limiting condition.

In this work, the equation governing the deformation of interface, which describes the local balance of the Maxwell stress and the capillary pressure, is integrated along the liquid meniscus to recover the LY equation. It is explained how the present approach gives the same result to that relying on the macroscopic balance of the forces acting on entire meniscus. The microscopic interactions which could be involved in the deformation at very close to the TCL are neglected. The meaning and the validity of the assumption are discussed.

2. Analysis

2.1 System

Consider a two-dimensional droplet in stable equilibrium on a horizontal solid substrate, being immersed in another fluid (air or liquid) (see Fig. 1). The droplet is assumed to be an electrically perfect conductor. The cross-section of the droplet shown in Fig. 1 can be imagined to continue normal to the page. Considering the edge region as a part of a two-dimensional droplet is equivalent to disregarding any effect of circumferential curvature. This simplification may be valid when the size of the droplet is sufficiently large compared to the thickness of an insulating layer, which is readily satisfied for a conventional millimeter-sized droplet. The surrounding fluid region and the insulating-layer region are represented by Ω_f and Ω_s, respectively, and the droplet–fluid interface is denoted by Σ. A Cartesian (x, y) coordinate system is introduced as shown in Fig. 1. The x- and y-axes are parallel and normal to the substrate surface, respectively. The arc length *s* is measured from a point B which is on the substrate surface.

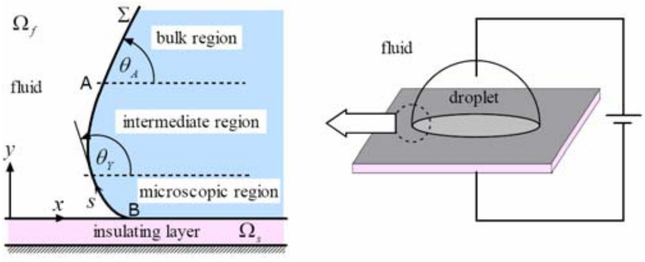

Figure 1. Division of meniscus around the three-phase contact line.

For the sake of convenience, the meniscus of the droplet around the TCL is divided into three regions: the microscopic, intermediate, and bulk regions (see Fig. 1). In the *microscopic region*, which is at very close to the substrate, the short-range interaction (e.g., steric interaction) and the long-range interaction (e.g., van der Waals interaction) are to be significant. The electrical double layer may normally be included in this region. The scale of the microscopic region is in the order of 100Å. [9] For the sake of convenience, we call here those interactions mentioned above the *microscopic interactions*. When there is no Maxwell stress, the tangential angle at the upper edge of the microscopic region is defined by Young's angle which is in fact a consequence of the microscopic interactions near the TCL. Note that here the Maxwell stress represents the electrical stress induced by the externally applied field, disregarding the contribution from the electrical double layer.

Above the microscopic region, there exists the *intermediate region* where the microscopic interactions become negligible. In this region, the Maxwell stress is dominant, and the deformation of interface is dominated by the Maxwell stress. As a matter of fact, the Maxwell stress is distributed over the microscopic region as well as the intermediate region. Accordingly, the Maxwell stress may contribute to the deformation of interface in the microscopic region in a certain degree. However, if the thickness of the microscopic region (l_m) becomes very thin, the net force acting on the microscopic region by the Maxwell stress becomes negligible compared to the net force acting on entire meniscus. This condition will be satisfied when *m l* << *d*, since the thickness of the intermediate region is proportional to *d*. Here, we assume that *m l* << *d*. Under this assumption, one can define Young's angle which will be hardly affected by the Maxwell stress. As will be discussed later, the conclusion of Buehrle et al. [7] that Young's angle is not affected by the Maxwell stress is deduced by neglecting the deformation of interface in the microscopic region, from the beginning.

At a sufficiently far distance from the substrate, say 10*d* (*d* is normally in the order of micrometer), there is the *bulk region* in which the Maxwell stress becomes negligible. In this region, the deformation of interface is governed by the hydrostatic pressure and the capillary pressure corresponding to the macroscopic curvature of the droplet, i.e., γ / R in which *R* represents the length scale of the droplet. In fact, the LY equation describes the change of contact angle at the lower edge of the bulk region. If there is no Maxwell stress acting on the interface, Young's angle corresponds to the macroscopic contact angle.

2.2 Derivation

We describe the surface profile by a function $x = f(y)$. Then, the macroscopic representation of the Helmholtz free energy per unit length (*F*) of a strictly two-dimensional system is written as follows:

$$F = (\gamma_{sl} - \gamma_{sf})x_0 + \int_A^B \gamma\sqrt{1+f'^2}\,dy - \frac{1}{2}\int_{\Omega_f}\varepsilon_f E^2 d\Omega - \frac{1}{2}\int_{\Omega_s}\varepsilon E^2 d\Omega \tag{2}$$

where γ_{sl} represents the interfacial tension at solid-liquid interface, γ_{sf} the interfacial tension at solid-fluid interface, x_0 the horizontal coordinate of the TCL, ε_f the electric permittivity of the surrounding fluid, and E = | **E** | the electric-field strength. The first and second terms represent the surface energy. The third and fourth terms denote the electrostatic energy in the surrounding fluid and the insulating layer, respectively. (The contribution of droplet vanishes due to no internal electric fields inside a conducting droplet.)

Applying the formal variational procedure with respect to *f*,

$$\gamma\kappa = -\frac{f''(y)}{\left[1+f'(y)^2\right]^{3/2}} = \frac{1}{2}\varepsilon_f E^2 \tag{3}$$

Here, k denotes the local curvature of the droplet surface. Using the geometric relation of the local curvature (κ) and the local tangential angle (θ) with the arc length (*s*), Eq. (3) can be written as follows:

$$\gamma\frac{d\theta}{ds} = -\frac{1}{2}\varepsilon_f E^2 \tag{4}$$

After multiplying sinq on both sides and integrating both sides with respect to s, from B to A, we obtain

$$\gamma(\cos\theta_A - \cos\theta_B) = \int_B^A \frac{1}{2}\varepsilon_f E^2 \sin\theta ds \tag{5}$$

On the other hand, for the perfect conducting droplet, the electrostatic force (per unit length) acting on the droplet surface is represented by [2,4]

$$\mathbf{F}_{el} = \int_B^A \mathbf{T}\cdot\mathbf{n}ds = \int_B^A \frac{1}{2}\varepsilon_f(\mathbf{E}\cdot\mathbf{n})\mathbf{E}ds = \int_B^A \frac{1}{2}\varepsilon_f E^2\mathbf{n}ds \tag{6}$$

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Here, **T** is the Maxwell stress and **n** is the outward unit vector normal to the droplet surface which can be related to the local tangential angle. (**n** = -**i** $\sin\theta$+ **j** $\cos\theta$, where **i** and **j** are the unit vector in x- and y-direction, respectively.) From the Eqs. (5) and (6), the horizontal force balance says

$$\cos\theta_A - \cos\theta_B = -\frac{F_{el}^x}{\gamma} . \qquad (7)$$

If we further assume that the microscopic interactions can be represented by Young's angle, $\theta_B = \theta_Y$. And the horizontal component of the electrical force (per unit length) is represented like [4]

$$F_{el}^x = -\frac{\varepsilon V^2}{2d} \qquad (8)$$

Finally, we can obtain the Lippmann-Young equation: ($\theta_A = \theta$)

$$\cos\theta = \cos\theta_Y + \frac{\varepsilon V^2}{2\gamma d} \qquad (8)$$

3. Discussion

In the present work, In the present work, the effect of microscopic interactions in the microscopic region is just represented by Young's angle. Let's consider what this simplification practically represents. The Maxwell stress is distributed within a distance of O(d) from the TCL. For a demonstration purpose, the electrostatic field around a hemispherical droplet is analyzed numerically with changing the thickness of insulating layer as d/R = 0.2, 0.1, and 0.05, where R is the radius of the droplet. The applied voltage is changed to keep V/d constant. Figure 2 shows the distribution of the Maxwell stress. As shown, as the thickness of the insulating layer gets smaller, the region where the Maxwell stress is effective becomes reduced. When d is in the order of micrometer, the thickness of the intermediate region is much greater than that of the microscopic region, and most of stress may act on the intermediate region. In this case, Young's angle may be hardly affected by the Maxwell stress.

In contrast, consider the case in which d is in the order of nanometer, which corresponds to the case of electrowetting on self-assembled monolayers.17-21 Then, the deformation of interface should be determined by considering the combined effect of the Maxwell stress and the microscopic interactions around the TCL. In this case, the LY equation may be inappropriate to describe the change of contact angle; even, it may be difficult to make a distinction between Young's angle and Lippmann's angle. Consequently, the present assumption of neglecting the microscopic region and microscopic interactions may have validity only for the case of a sufficiently large d. In electrowetting, a complete description for a droplet including the microscopic interactions, Maxwell stresses, and capillary pressure, is still lacking.[9]

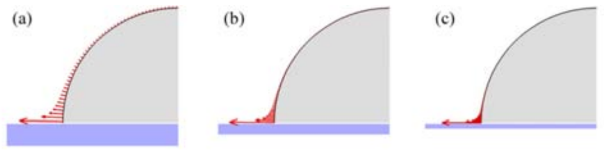

Figure 2. The Maxwell stress acting on a hemispherical droplet for different thicknesses of insulating layer: (a) d / R = 0.2; (b) d / R = 0.1; (c) d / R = 0.05. The numerical analysis is carried out by using the commercial software, COMSOL Multiphysics™.

Buehrle et al.[7] calculated the microscopic contact angle for a fixed macroscopic contact angle by analyzing the deformation of interface. They showed numerically that the microscopic contact angle approaches to Young's angle which is calculated by the LY equation. Based on this result, they concluded that Young's angle is not affected by the Maxwell stress. They did not, however, consider any influence of the microscopic interactions at the microscopic region. As shown here, Young's angle and Lippmann's angle should essentially be related by the LY equation as long as the microscopic interactions are neglected. The result of Buehrle et al.[7] for the limiting angle (their Young's angle) is no more than a numerical verification of the validity of the LY equation. Their conclusion, however, will be valid when d is sufficiently large compared to the thickness of the microscopic region.

The effect of hydrostatic pressure is neglected from the beginning. The change of cosθ for a region is related with the net horizontal force acting on the region. Thus, we can estimate the change of cosθ at the intermediate region caused by the hydrostatic pressure as $\Delta\rho gRd\ /\gamma$. Here, $\Delta\rho$ is the density difference between in and out of the droplet, and g the gravitational acceleration. The Maxwell stress is in the order of $\varepsilon(V\ /\ d)^2$, and the consequent change of cosθ will be in the order of $\varepsilon V2\ /\gamma d$. It is evident $\varepsilon\ V2\ /\gamma d$ is in the order of 1. When $\Delta\rho$ = 1000 kg/m3, g = 9.81 m/s^2, R = 1 mm, d = 10 μm, and γ = 0.02 N/m, the change of cosθ due to the hydrostatic pressure is as small as 5×10− 3. The capillary pressure which is associated with the curvature of the macroscopic droplet is in the order of γ / R. Therefore, the change of cosθ due
to the capillary pressure is in the order of d / R, which is usually less than 10^{-2} .

4. Conclusion

The Lippmann-Young equation was derived considering the deformation of interface at the TCL. From the integration of the local balance equation of the Maxwell stress and the capillary pressure, we recovered the LY equation. In the derivation, the microscopic interaction at the TCL was neglected and represented using Young's angle which is assumed to be independent on the Maxwell stress. These assumptions will be valid for the case of a sufficiently thick insulating layer in which the net electric force acting on the microscopic region constitutes only a minor portion relative to the total electrical force.

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