Interface Behavior between Two Fluids Vertically Oscillated in a Circular Cylinder under Nonlinear Contact Line Condition
(1st Report, Measurement and Modeling of the Contact Line Behavior)*

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Abstract
The development of waves on a fluid-fluid interface, excited by a vertical, relative motion of a solid wall enclosing the fluids, is affected significantly by the mobility of the interface on the wall. The effective, cycle-averaged mobility depends on the amplitudes of excitation and waves, because of a non-linear dependence of the velocity of the fluid-fluid-wall contact line on the angle of the interface hitting the wall. At higher amplitudes of excitation and waves, moreover, the interface motion is tied to the low mobility of the contact line only for a limited fraction of each cycle; for the rest of the cycle the interface is virtually untied from the wall, being connected to the contact line only through the surface of a thin, flat liquid film left on the wall. An analytical model is developed on the wall surface boundary conditions for the interface profile, in terms of non-linear relationship between the interface velocity along the wall and the near-wall inclination angle of interface. The model is based on optical data taken in a quasi-static experiment where measurements of near-wall interface configuration were essentially unaffected by wave generation on the interface. Numerical simulation with this model reproduces successfully the changes in the near-wall configuration of the interface in the experiment, including development and depletion of a liquid film on the wall associated with relative motion between the contact line and the interface elevation away from the wall.

Key words: Two-Layer Fluid, Numerical Analysis, Wave, Contact Line, Contact Angle, Hysteresis, Liquid Film

1. Introduction
The natural frequency and the damping rate of waves on a fluid-fluid interface, bounded by a solid wall, are affected by the mobility of the fluid-fluid-wall contact line, when the lateral size of the interface is comparable to, or less than, the capillary length \( l_c = \sqrt{\frac{\sigma}{\Delta \rho g}} \), where \( \sigma \) is the surface tension and \( \Delta \rho \) is the density difference between the fluids. The mobility of the contact line can be measured by the sensitivity of the contact angle (the slope angle of the interface at the contact line) \( \theta \) to the speed of the contact line.
relative to the wall, \( V_{CL} \). The two parameters provide the wall-surface boundary conditions for the wave profile \( \varsigma \), in terms of \( -\cot(\theta - 90^\circ) = (\partial \varsigma / \partial n)_{\text{wall}} \) and \( V_{CL} = (\partial \varsigma / \partial t)_{\text{wall}} \), where \( n \) is the distance from the wall. Hocking\(^{(1)}\) and Miles\(^{(2)}\) analyzed interface waves for cases where the two parameters above are linearly dependent on each other, such that \( [\cot(\theta - 90^\circ) - \cot(\theta_0 - 90^\circ) ] \times V_{CL} \), where \( \theta_0 \) is the static contact angle that is determined by the surface tensions of fluids and wall material. However, experimental studies have shown nonlinear dependence of \( \theta \) (or \( \cot \theta \)) on \( V_{CL} \), for contact lines in both steady\(^{(3)}\) and nonsteady motions.

For contact lines at the boundary of oscillating interfaces, where \( \theta \) is subjected to oscillatory changes, the contact angle hysteresis can cause significant nonlinearity in the relationship between \( \theta \) and \( V_{CL} \). The contact angle hysteresis means the ability of \( \theta \) to deviate from \( \theta_0 \), while keeping the contact line at rest. It is observed for ordinary solid surfaces that contain microscopic nonuniformity due to imperfections or contamination by foreign materials. The maximum and minimum values that \( \theta \) can take, without causing the motion of the contact line, are termed the advancing contact angle and the receding contact angle, \( \theta_{adv} \) and \( \theta_{rec} \), respectively. Consequently, the relationship between \( \theta \) and \( V_{CL} \) has strong nonlinearities at \( \theta_{adv} \) and \( \theta_{rec} \). Ting\(^{(4)}\), Cocciaro\(^{(5)}\) and Jiang\(^{(6)}\) studied experimentally the behavior of interfaces exited by a vertically moving wall\(^{(4)}\) or horizontal oscillation of a basin containing liquid\(^{(5,6)}\). Their studies showed that the amplitude of \( \theta \) depended on the amplitude and the frequency of excitation. The contact line remained stationary at a certain elevation on the wall when the amplitude of \( \theta \) was small, but larger amplitudes of \( \theta \) resulted in intermittent motion of the contact line in each cycle.

The behavior of a moving contact line becomes complicated when either or both of the fluids wet(s) the wall. When the contact line is moving in such direction that the more-wetting fluid than the other is receding, i.e., decreasing its contact area with the wall, the replacement of the fluid by the other will take time. Consequently, when the fluids are forced to move in that direction, more quickly than the replacement of the fluids on the wall can occur, a thin film of the more-wetting fluid is left on the wall, behind the fluid motion. The interface, outside the film, moves with the fluids, and the slope angle of the interface near the wall is little affected by the velocity of the interface relative to the wall. When the film is invisibly thin, therefore, it looks as if the contact line slips freely and the receding fluid is replaced instantaneously by the advancing fluid. The intersect of the visible interface profile and the wall is called the apparent contact line, and the slope angle of the visible profile near the wall is called the apparent contact angle. These differ from the actual contact line located at the edge of the fluid film, and the actual contact angle there. In considering the fluid-dynamic interactions between the interface waves and the wall, in the presence of a fluid film on the wall, the speed of the apparent contact line and the apparent contact angle are more important than those of the actual ones. In the previous study\(^{(7)}\), we reported on the effects of film formation on the motion and deformation of a fluid-fluid interface moving along a vertical wall. The motions of the actual and apparent contact lines, and the variations in the contact angle were observed.

Summarizing, the boundary conditions imposed by the contact line to the interface waves of finite magnitudes include nonlinearities due to the nonlinear relationship between \( \theta \) and \( V_{CL} \), including the contact angle hysteresis, and asymmetry with respect to the direction of the contact line motion if at least one of the fluids wets the wall. The delayed motion of the actual contact line, in the presence of fluid film on the wall, can introduce a time scale dependent on the amplitudes of the contact lines. This means that the boundary conditions at the contact line change with phase angle in each cycle in a manner that may depend on the wave amplitude and frequency. Such changes and dependence will affect the wave excitation or damping due to the wave-wall interactions.

The objective of the present paper is (a) to investigate the behavior of the contact line
and the contact angle under such conditions where fluid film is formed in response to the changes in the contact angle, and (b) to develop a model on such behavior that will be used in the second paper to study the effects of nonlinear, time-dependent boundary condition at the contact line on wave excitation due to relative motion between the fluids and the wall. We present the results of detailed measurements on the motion of the actual and apparent contact lines, including the relationship between $\theta$ and $V_{CL}$ at the actual and apparent contact lines. Although it would be preferable to make measurements in steady states to avoid transient effects on the interface profile, fluid film can form on a vertical wall only in transients unless the two fluids have the same density. The measurements were therefore made under slow, cyclic transients. In the present paper, the experimental data are incorporated into a contact line model, and the model is tested on a two-dimensional code for the capability to reproduce the transient changes in the experiment.

2. Experimental method

The experimental setup, shown in Fig. 1, is the same as the one used in the previous study\(^7\). The test section is a 56 mm i.d. vertical cylinder made of acrylic resin, where an interface is formed between kerosene (790 kg/m\(^3\) in density) and water (996 kg/m\(^3\)) layers. The capillary length on the interface is estimated to be 4.5 mm, as large as 15% of the cylinder radius because of the small density difference between the two fluids. This implies strong wall effects on the interface waves. The kerosene wets the surface of acrylic resin better than the water does. This results in the static meniscus to be convex upward, with a static contact angle measured in water greater than 90\(^\circ\).

The fluids were excited sinusoidally in the vertical direction, through the test section, at a frequency of 0.49 Hz and an amplitude of 5.5 mm. Only weak waves were exited on the interface under this condition, because the excitation frequency was more than four times lower than the fundamental natural frequency of the interface (2.2 Hz).

Two optical methods were used for the measurement of the interface and contact line behavior as shown in Figs. 2 and 3. In both methods the interface was visualized by the luminescence of Rhodamine-B fluorescent dye in water, excited by an Ar laser sheet illuminating a vertical plane in the test section. The image distortion due to the light refraction across the cylinder wall was suppressed by enclosing the cylinder with a transparent, rectangular water tank. The measurements were made with an interval of 1/30 s.

The whole image of the interface profile was obtained with the method A shown in Fig. 2. Video pictures (640×480 pixels) were taken from well below the interface to avoid the influence of light refraction across the curved interface. The spatial resolution was 0.3 mm and 0.18 mm in the horizontal and vertical directions, respectively. This was not enough to resolve the liquid film on the wall, or to measure the location of the actual contact line, but was enough to measure the location of the apparent contact line and the apparent contact angle. In the following sections, the contact angle means that measured through the water
The motion of the actual contact line, i.e., the lower edge of the kerosene film, was measured with the method B shown in Fig. 3. The laser light incident on the lower end of the kerosene film is refracted at a smaller angle than that going directly into water, or through the flat region of the kerosene film, due to the difference in refraction index between water and kerosene. As a result, the actual contact line makes a wedge-shaped shadow zone in water. The location of the actual contact line is then found by extrapolating the boundaries of the wedge to the wall. Although we thought of applying this method to the measurement of the actual contact angle, the continuous change in the interface slope angle at the lower end of the film and the small non-parallelity in the laser light were found to cause difficulties.

3. Experimental results

Figure 4 shows the temporal variations of measured quantities in a single cycle of excitation, where a kerosene film was present on the wall between $t=1.0$ s and 2.1 s from the initiation of the cycle. The excitation (cross-sectionally averaged fluid displacement), the contact angle, together with the vertical position of the actual and apparent contact lines are shown. The position of the apparent contact line was defined by extrapolating the
interface profile, shown Fig. 5, to the wall surface by means of second- or third-order polynomial curve fitting, since the measurement of interface location near the wall was affected by light diffusion at the wall surface. In the time period with the presence of a kerosene film, the contact angle indicated in the figure is the apparent contact angle.

The (actual) contact line is stationary until about $t=0.2$ s, i.e., until the contact angle decreases from $150^\circ$ to $120^\circ$, and then starts descending, following the average displacement of the fluids. Water recedes and kerosene advances, as the contact line descends the wall surface. The descending speed of the contact line increases with time, reaches a maximum at $t=0.3$ s, and then decreases. This is accompanied by a decrease in the contact angle, taking a minimum of $90^\circ$ at $t=0.4$ s, and by an increase afterward. After $t=0.7$ s, the actual contact line remains stationary for a while, despite the contact angle continues increasing.

The interface profile is nearly flat, except in the vicinity of the wall, between 0.2 and 0.7 s as shown in Fig. 5. The fluid motion is piston like in this time period, while the interface near the contact line is being deformed with the change in the actual contact angle that forced the actual contact line to descend. After 0.7 s, the interface profile starts becoming convex upward, since the actual contact line does not follow the forced ascending motion of the fluids. The actual contact angle increases continuously, reaching $150^\circ$ at $t=1.0$ s.

An apparent contact line emerges, segregating itself from the actual contact line, at $t=1.0$ s, and starts ascending with an apparent contact angle of $150^\circ$. A kerosene film forms between the apparent and actual contact lines.

The actual contact line starts ascending only after $t=1.1$ s. It keeps a constant ascending speed of 3.6 mm/s, apparently unaffected by the forced fluid motions or the changes in the film length with time. Now, kerosene recedes and water advances, as the actual contact line ascends the wall.

The forced upward motion of the fluids reaches its top dead point at 1.7 s. The kerosene film has grown to be 4.9 mm long at that time. Now, the interface starts descending keeping its profile, including the apparent contact angle, until the film disappears and the apparent contact line is captured by the actual contact line at 2.04 s (the end of the cycle). The actual contact line has climbed the wall for a distance of 4 mm after it had been left behind the apparent contact line.

In the present experiment, a fluid film is present on the wall for almost exactly a half of the cycle; however, this time fraction depends on the frequency and the amplitude of the excitation, because the ascending speed of the actual contact line appears to be independent of these excitation parameters, but determined by the combination of the materials.

The thickness of the film was not measured in the experiment but an approximate estimation can be made with established theories. For a steady film with uniform thickness, the mass conservation requires the thickness $\eta$ to be

$$\eta = \frac{Q_{\text{net}}}{V_0},$$

where $Q_{\text{net}}$ is the net flow rate in the film and $V_0$ is the speed of the lower edge of the film, i.e., the actual contact line.

The flow rate $Q_{\text{net}}$ is determined by the balance between the gravity and viscous forces in the film. If we neglect the influence of the shear stress on the surface of the film, as will be justified, we obtain,

$$Q_{\text{net}} = \frac{\Delta \rho g \eta^3}{3\mu}.$$  

Substituting Eq. (2) into (1) and using the measured value of $V_0=3.6$ mm/s, we find $\eta$ to be approximately 0.1 mm, and the shear rate in the film to be 49 s$^{-1}$ in the present experiment. The thickness of the oscillatory boundary layer on the wall, i.e., the Stokes layer given by $\sqrt{2\nu/\omega}$, is calculate to be about 0.8 mm at the angular frequency $\omega$ in the present experiment. With the peak velocity at the outer edge of the boundary layer of 17
mm/s in the experiment, the shear rate near the wall is calculated to be 21 s⁻¹. It is thus justifiable to assume the film surface to be shear free in evaluating the film thickness.

The interface geometry is convex upward throughout the period where a kerosene film is present. It looks as if the apparent contact line slips freely on the wall, since the interface profile remains nearly identical to that in the static state, and the apparent contact angle is kept constant value of about 150°.

4. Modeling of contact line behavior

We have developed a model on the contact line behavior including the relation between θ and VCL, based on the experimental results described in the previous section.

4.1 Modeling of the actual contact line unaccompanied by a kerosene film

Figure 6 shows the contact angle θ plotted against the speed of contact line, VCL, during the descending motion of the actual contact line. Although the contact angle at a moving contact line is regarded commonly to be a single valued function of the instantaneous value of VCL, the figure shows hysteretic changes in θ. That is, when VCL is decreasing with time, θ takes larger value than in the period where VCL is increasing. The reasons for such hysteretic behavior may include the non-steadiness of the interface configuration in the near-wall region. In the present experiment, it is likely that the interface profile near the wall is affected by the Stokes layer whose thickness has been estimated to be 0.8 mm, where the velocity gradient normal to the wall changes with time. Dussan et al. have shown that the viscous effect on the interface configuration become appreciable at least 10 μm away from the wall for a capillary number Ca=10⁻⁴, where Ca=\(\mu U/\sigma\), with \(\mu\) being the viscosity of the fluid and \(U\) the characteristic velocity. Since Ca=3×10⁻⁴ for the present experiment, the viscous deformation of the interface may occur even at smaller distances from the wall.

We use the lower curve in Fig. 6 as the relation between θ and VCL in our model, because we are interested in the prediction of the time of film formation that occurs when VCL is increasing. As shown in the figure, the curve can be fitted by

\[
\theta = \pi + \tan^{-1}\left[C_1/(C_2 + C_3)\right]
\]

where \(C_1\), \(C_2\) and \(C_3\) are the fitting parameters. The values of the parameters are -3.9 × 10⁻⁴ s/m, -5.3 × 10⁻⁴ and -0.1553, respectively.

The difference at VCL=0 between the two curves in Fig. 6 shows the contact angle hysteresis for the present water-kerosene-acrylic resin system. The receding contact angle \(\theta_{rec}\) is defined to be 120° in accordance with Eq. (3). The advancing contact angle \(\theta_{adv}\) is defined to be the measured value (150°) of the apparent contact angle at the time when the apparent contact line starts ascending.

4.2 Modeling of the apparent and actual contact lines accompanied by a kerosene film

The model does not consider the hydrodynamics in the kerosene film, or at the apparent
contact line that connects the bulk fluid motion to the flow in the film. The model only tracks the ascending motion of the lower edge of the film, i.e., the actual contact line, to be able to predict the time when it captures the apparent contact line.

The actual contact line starts ascending 0.1 s after the apparent contact line emerges from the actual contact line. By that time, the apparent contact line has become 1.3 mm higher than the actual contact line. The model uses this difference in elevation to trigger the ascending motion of the actual contact line.

5. Analytical method

5.1 Single-layer modeling of two-layer flow field

The flow fields in the upper and lower fluid layers are coupled through the mechanical interactions at the interface, since the two fluids have similar densities. However, when the flow fields are approximated by linear, inviscid and irrotational motions, the velocity potential distributions in the upper and lower fluids are symmetrical with respect to the interface. Based on this fact, we analyzed the coupled two-layer flow fields in the experiment using a single-layer model.

If we approximate the flow in each layer with an inviscid, irrotational flow with an infinite depth, the velocity potentials \( \Phi \) in the two layers satisfy the kinematic conditions at the interface,

\[
\frac{\partial \Phi_a}{\partial z} = \frac{\partial \Phi_b}{\partial z} = \frac{\partial \zeta}{\partial t} \quad (z = 0),
\]

and the boundary condition,

\[
\frac{\partial \Phi_a}{\partial z} = 0, \quad \frac{\partial \Phi_b}{\partial z} = 0 \quad (z = \pm \infty),
\]

where the subscripts \( a \) and \( b \) denote the upper and lower fluids, respectively.

This leads to,

\[
\Phi_a(r, z, t) = -\Phi_b(r, z, t),
\]

i.e., symmetric velocity distribution with respect to the interface.

The pressure balance equation at the fluid-fluid interface is,

\[
\frac{\partial}{\partial t} (\rho_a \Phi_a - \rho_b \Phi_b) = -(\rho_b - \rho_a) g \zeta - C_{\mu a} \Phi_a + C_{\mu b} \Phi_b + \sigma \kappa
\]

where \( \kappa \) is the curvature of the interface, and \( C_{\mu} \) is the viscous damping coefficient introduced by Rayleigh(10).

Substituting Eq. (6) into Eq. (7) we obtain,

\[
\frac{\partial \Phi_a}{\partial t} = \left( \frac{\rho_b - \rho_a}{\rho_a + \rho_b} \right) g \zeta - \left( \frac{C_{\mu a} + C_{\mu b}}{\rho_a + \rho_b} \right) \Phi_a - \frac{\sigma}{\rho_a + \rho_b} \kappa
\]

By comparing Eq. (8) with the pressure balance equation at the free surface of a single-layer flow field,

\[
\frac{\partial \Phi}{\partial t} = g \zeta - C_{\mu} \frac{\Phi}{\rho} - \frac{\sigma}{\rho} \kappa
\]

we can derive the following quantities, denoted by primed symbols, to be used in the single-layer model to represent the two-layer interactions,

\[
g' = \left( \frac{\rho_b - \rho_a}{\rho_a + \rho_b} \right) g, \quad \left( \frac{C_{\mu a} + C_{\mu b}}{\rho_a + \rho_b} \right) \frac{\Phi}{\rho} = \frac{\sigma}{\rho_a + \rho_b}
\]

Furthermore, if we assume \( C_{\mu} \) to be proportional to the viscosity \( \mu \), then the second equation in Eq. (10) can be rewritten into an expression for the equivalent kinetic viscosity for the single-layer model, \( \nu' = \left( \mu_a + \mu_b \right) / \left( \rho_a + \rho_b \right) \).

5.2 Numerical method

In the present study, we focus on the interface response at its fundamental axisymmetric mode, with such amplitudes as keeping the interface displacement single valued at any lateral location. We therefore used a cylindrical boundary-fitted coordinate (BFC) system...
for analysis. Non-uniform meshes, with finer resolution at the interface and the wall, were used.

In the cylindrical BFC system, the physical coordinate \((r, z)\) is transformed into the nondimensional coordinate \((\xi, \eta)\), as

\[
r = Z_1(\xi)R, \quad z = Z_2(\eta)h(r, t)
\]

where \(R\) and \(h\) are the radius of the cylinder and the height of the interface, respectively, and \(0 < \xi < 1, 0 < \eta < 1\). The functions \(Z_1\) and \(Z_2\) are transformation functions satisfying

\[
\begin{align*}
Z_1(0) &= 0, Z_1(1) = 1, \\
Z_2(0) &= 0, Z_2(1) = 1.
\end{align*}
\]

The function of \(Z_n\) is chosen as

\[
Z_n(\xi) = \frac{\exp(\alpha_n \xi) - 1}{\exp(\alpha_n) + 1}
\]

where \(\alpha_n\) is the positive constant for controlling the distribution of the mesh interval. The values of \(\alpha_n\) used in the analysis were 3.2 and 4.8 for \(n=1\) and 2, respectively.

The analysis was conducted for a two dimensional region covering the full radius of the test section (28 mm) and a water depth of 75 mm as shown in Fig. 7. The mesh number is 42 in the radial and 75 in the vertical directions, with a typical minimum mesh size of 0.18 mm \(\times\) 0.08 mm. The mesh size is smaller than the thickness of oscillatory boundary layer of about 0.8 mm at the angular frequency \(\omega\) in the present experiment.

The mass and momentum equations are discretized for staggered meshes, using the third-order upwind scheme for the convection term and the centered difference scheme for the diffusion term. The Navier-Stokes equation is solved using Simplified Marker-and-cell (SMAC) method\(^{(11)}\) where velocity distribution is calculated from the known pressure and then pressure is modified until the velocity distribution satisfies the mass conservation.

No-slip boundary condition was applied on the wall surface and on the interface. The latter is a crude approximation, but would not be too much in error since the tangential velocities for linear waves are symmetric with respect to the interface, as has been seen in Eq. (6), and the ratio of the viscosities of the two fluids is about 2. The fluid excitation in the experiment was represented by the vertical motion of the wall, and periodic change in the equivalent gravitational acceleration \(g'\) defined in Eq. (10). The fluid velocity at the lower boundary of the model was assumed to be zero.

The present numerical model, with a minimum mesh size of 0.18 mm \(\times\) 0.08 mm, does not consider explicitly or precisely the local flow field near the contact line, including the flow in the fluid film. The contact line is treated as a geometrical boundary condition, in terms of \(\theta\) vs. \(V_{CL}\), as described in Section 4, for the interface profile. The boundary condition constrains the velocity of the contact line, i.e., the edge of the interface, only in the time period where the fluid film is judged not to be present. In the time period with the presence of the film, the contact line is allowed to move without such a constraint on \(V_{CL}\), while the interface profile is given a boundary condition that holds \(\theta\) at a constant value.
6. Simulation results and discussion

Figure 8 shows the analytical results obtained with the model described the preceding section, in comparison with experimental data. The analytical results agree well with experimental results, reproducing the formation of a kerosene film at 1.0 s. The interface elevation at the centerline of the test section show a higher frequency component superposed on the sinusoidal excitation at 0.49 Hz, as in the experiment. The higher frequency component has a larger amplitude than in the experiment. Figure 9 shows the deviation of the interface elevation at the centerline from the cross-sectionally averaged interface elevation. This appears to reflect the changes in the interface profile in the respective time periods, shown in Fig. 5. The interface is convex upward at 0.0 s but becomes flat and slightly concave by 0.3 s, as the interface is forced to descend by the forced fluid motion, resulting in a negative deviation in the centerline elevation of the interface from the average. The interface configuration starts returning to be convex at about 0.7 s, as the forced fluid motion turns to be upward, resulting in positive deviations. The higher frequency component has a predominant period of 0.39 s, which is almost five times greater than the excitation period of 2.0 s, and is close to the period of the fundamental mode of the interface wave, 0.44 s. It is hence likely that that the fundamental mode was excited due to the nonlinear response of the system to the forced excitation. The nonlinearity includes that in the relation between $\theta$ and $V_{cz}$ shown in Fig. 6.

The higher frequency component is exited at about 0 s in both the experiment and analysis, and decays slowly. The higher frequency component dies out, quickly, at about 1 s in the experiment; however, in the analysis it is excited again at some time around 1 s, and damps out only slowly thereafter. The above times correspond to the time periods where the contact angle, and the interface profile, are forced to change quickly. If the higher frequency component is excited primarily in these two time periods, the phasing between the components excited in the two time periods will affect the change in the amplitude. Figure 9 shows the higher frequency component in the analysis is about 0.1 s in delay relative to that in the experiment, until 0.8 s. To see the influence of this delay in the analysis, we made an additional calculation where the phase angle of excitation was shifted artificially after 0.6 s. The result, dotted line in Fig. 6, shows a quicker decay of the high frequency component after 1.0 s, than that in the base analysis.
7. Conclusion

The behavior of the fluid-fluid-wall contact line moving on a vertical wall has been investigated experimentally, with a particular interest in the nonlinearity in the relation between the contact angle and the speed of contact line. The experiment was conducted by oscillating the interface between kerosene and water confined in an acrylic cylinder with considerably lower frequency than that of the interface wave of the fundamental mode. The position of the contact line and the contact angle were measured using optical methods.

A model has been developed for prediction of the motion and deformation of the fluid-fluid interface in response to relative motion between the fluids and the surrounding solid wall, taking account of the nonlinear boundary condition at the contact line. The following results were obtained:

- The nonlinearity in the relation between the contact angle and the speed of contact line stems from, in addition to the well known contact angle hysteresis, the dependence of the contact line behavior on the direction of contact line motion, which occurs when the two fluids have different wetting properties. If one of the fluids wets the wall better than the other, it tends to form a film ahead of its receding contact line, and this allows fluids away from the wall to move without constraint imposed by the contact line. For an oscillatory interface, the film can form intermittently, in a particular portion of the cycle. The emergence and extinction of the film can cause strongly nonlinear changes in the boundary condition for the interface motion.

- The numerical model developed in the present study well reproduced the experimental results on the interface motion and deformation in response to relative motion between the fluids and the surrounding solid wall.

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Appendix - Estimation of the apparent contact angle

The experimental definition of the apparent contact angle depends on the spatial resolution of the measurement and the curvature of the interface. When the measurement of the interface profile is made at two discrete distances from the wall, the apparent contact angle \( \theta_{\text{app}} \) is given by,

\[
\theta_{\text{app}} = \pi / 2 + \sin^{-1} \left( 1 - (\delta x + \delta x_0) \kappa_{\text{app}} \right)
\]

(A.1)

where \( \kappa_{\text{app}} \) is the curvature of the interface at the apparent contact line, \( \delta x \) the horizontal resolution of the measurement, \( \delta x_0 \) the horizontal distance of the first measurement point from the surface of the liquid film. In the present study the We and Ca numbers are so small that the static curvature of the interface, derived by Landau\(^{(12)}\), will give a good approximation of \( \kappa_{\text{app}} \), such that

\[
\kappa_{\text{app}} = \frac{2N_{\text{ap}}g}{\sigma},
\]

(A.2)

which is 2.2 mm for the present combination of liquids. In this study \( \delta x \) was 0.3 mm and \( \delta x_0 \) varies from 0 to 0.3 mm. Using these numbers, \( \theta_{\text{app}} \) is estimated to be in the range of 136° < \( \theta_{\text{app}} \) < 150°. The value of \( \theta_{\text{app}} \) used in Sec. III (150°) is based on this estimation.
References