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ASYMPTOTIC THEORY OF THE SPREADING OF PARTIALLY WETTING LIQUID

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A mathematical model of the spreading of liquid along a plane solid surface is constituted for a finite equilibrium angle of wetting.

There is presently no closed consistent method of describing the spreading of a partially wetting liquid along a dry surface by methods of continuum mechanics. The basic reason for this is the incompatibility of the equations of motion of the viscous liquid and the adhesion conditions at a solid surface close to the line of three-phase contact [1, 2].

In [3, 5], it was proposed to resolve this contradiction by specifying the slip of the spreading liquid relative to the solid surface. However, the reason for the appearance of the contact angle θ and its dependence on the velocity of motion of the line of three-phase contact remains unclear here.

In [6, 7], it was proposed to reject any consideration of the liquid-film motion at small thicknesses $h < h_m$, $h_m \sim 10^{-10}$ m close to the film boundary, because of the inapplicability of the hypotheses of continuum mechanics there. In this case, the corresponding boundary problem is unclosed, since the angle of slope of the free surface $\theta(h_m)$ and the velocity of motion of the film boundary are specified quantities.

It was noted in [8, 9] that the reason for the formation of a contact angle is the additional "splitting" pressure arising in thin liquid layers on account of the action of Van der Waals forces. Van der Waals forces are diffuse in character, and appear at distances of the order of $\ell \sim 10^{-6}$-10^{-7} m, $\ell >> h_m$. Therefore, the action of the splitting pressure may be included in the hydrodynamic description of the spreading of liquid films. Analysis of the correspondingly modified equations of liquid-film motion may be formally extended into the region $h < h_m$ up to the film boundary $h \to +0$ [6, 10].

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In isothermal conditions, for an incompressible liquid, the splitting pressure may be regarded as equal to the chemical potential of molecular interaction of the liquid F per unit volume [11]. In [12, 13], for a two-point molecular interaction potential, under the assumption of conservation of homogeneity of the molecular structure of the liquid film, an expression is obtained for the chemical potential at the film surface F_0 close to the contact line

$$F_0 = h^{-3} G(\alpha) \tag{1}$$

where α is the angle of inclination of the film to the plane solid surface. A fundamental feature of the function G(α) is that it vanishes when $\alpha = \alpha_0$: $G(\alpha_0) = 0$. In [12, 13], it was noted that it is natural to identify the angle α_0 with the wetting angle: $\alpha_0 = \theta$. Here it must be noted that a more complex dependence of the splitting pressure on the film thickness h is seen experimentally [14, 15]. Therefore, generally speaking, it must be assumed that G = G(α, h).

Taking account of the chemical potential F in the equations of motion allows the problem of the spreading of a partially wetting liquid to be formulated in closed consistent form. However, the formal extension of the hydromechanics equations to the region $h < h_m$ implies neglecting processes that are significantly kinetic in character. The role of these processes may be taken into account in formulating the boundary conditions for the hydrodynamic equations of a liquid film. In particular, the surface diffusion of liquid molecules may be taken into account by specifying the rate of slip of the liquid film U relative to the solid surface.

The asymptotic behavior of liquid-film flow close to the line of three-phase contact (h → 0) is considered below, taking account of the chemical potential F and the slip rate U.

1. Derivation of Asymptotic Equations of the Form of the Liquid Film

In the approximation of lubricant theory [16], the system of equations describing the one-dimensional spreding of a liquid film along a horizontal solid surface takes the form

$$\frac{\partial (P+F)}{\partial x} = \mu \frac{\partial^2 u}{\partial y^2}; \quad \frac{\partial (P+F)}{\partial y} = -\rho g; \quad \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0. \tag{2}$$

To the system in Eq. (2) must be added conditions at the free surface of the liquid, specified by the relation y = h(x, t), and at the solid surface y = 0

$$\frac{\partial h}{\partial t} + u \frac{\partial h}{\partial x} = v; \quad P - P_0 = -\sigma \frac{\partial^2 h}{\partial x^2}; \quad \frac{\partial u}{\partial y} = 0 \text{ when } y = h(x, t), \tag{3}$$
$$u = U; \quad v = 0 \text{ when } y = 0.$$

The slip rate U is determined by the properties of the solid-surface material and of the value of the surface diffusion coefficient. In the first approximation, it may be assumed that [13]

$$U = D \left. \frac{\partial (P+F)}{\partial x} \right|_{y=0}, \quad D = \text{const}. \tag{4}$$

A differential equation defining the surface of the liquid film follows from Eqs. (2)-(4). In dimensionless form

$$\frac{\partial \eta}{\partial t} + \frac{\partial}{\partial x} \left\{ (\eta^3 + S\eta) \left[\frac{\partial^3 \eta}{\partial x^3} - \frac{\partial \eta}{\partial x} - \frac{\partial \Phi_0}{\partial x} \right] \right\} = 0. \tag{5}$$

Here η(x, t) is the film thickness; x, t are the dimensionless coordinate and time; $\Phi_0(\eta, \partial\eta/\partial x)$ is the chemical potential at the free surface of the film; S = const is a dimensionless parameters proportional to the slip rate. In writing Eq. (5), the folowing characteristic quantities are taken: $L = \sqrt{(\sigma/\rho g)}$ as the length, and $T = 3\mu L/\sigma$ as the time.

The function G(α) in Eq. (1) is fairly complex in form [12, 13]. For qualitative analysis, the asymptote of the function G(α) as α → θ is written in the form $G(\alpha) \sim (\theta^2 - \alpha^2)$. Taking into account that $\alpha \approx \partial\eta/\partial x$; $\alpha^2 \ll 1$ in the approximation of Eq. (2), the following representation of $\Phi_0(\eta, \partial\eta/\partial x)$ is obtained: $\Phi_0 = R\eta^{-3}\left[\theta^2 - \left(\frac{\partial\eta}{\partial x}\right)^2\right]$, $R = \text{const}$.

If $x = x_f(t)$ is the position of the film boundary (the line of three-phase contact), then the following conditions must hold when $x = x_f(t)$

$$\eta = 0, \quad (\eta^3 + S\eta)\left[\frac{\partial^3\eta}{\partial x^3} - \frac{\partial\eta}{\partial x} - \frac{\partial\Phi_0}{\partial x}\right] = 0, \quad x = x_f(t), \tag{6}$$

these are the boundary conditions for Eq. (5). The second condition in Eq. (6) denotes the absence of liquid flow rate through the film boundary $x = x_f(t)$.

The conditions in Eq. (6) are inadequate for unique determination of the form of the liquid film from Eq. (5). On the basis of the hypothesis in [12, 13] (see also the introduction, above), the closing boundary condition adopted is the relation

$$\left(\frac{\partial\eta}{\partial x}\right)^2 = \theta^2 \text{ when } x = x_f(t), \tag{7}$$

which may be regarded as the Young's condition for a partially wetting liquid.

As is clear from further analysis [17], the solution of Eq. (5) satisfying Eq. (7) is the only one for which the derivatives $\partial\eta/\partial x$ and $\partial^2\eta/\partial x^2$ are finite when $x = x_f(t)$. This solution when $x = x_f(t)$ asymptotically coincides with the solution of the equation obtained from Eq. (5) by omitting the highest derivative. Physically, this corresponds to neglecting the action of surface tension as $\eta \to +0$. It is of fundamental importance that, in this analysis of the boundary condition in Eq. (7), there are no difficulties associated with the impossibility of introducing the concept of surface tension as $\eta \to +0$ [8, 9].

The form of Eq. (5) asymptotically valid as $x \to x_f(t)$ is now found, using the method of [18]. Differentiating the first condition in Eq. (6) with respect to t, it is found that

$$\frac{\partial\eta}{\partial t} + \dot{x}_f\frac{\partial\eta}{\partial x} = 0, \quad x = x_f(t), \quad \dot{x}_f \equiv \frac{dx_f}{dt}. \tag{8}$$

Assuming that Eq. (8) is asymptotically valid as $\eta \to +0$, $x \to x_f(t)$, it is found, after replacing the derivative $\partial\eta/\partial x$ by the expression in Eq. (5) and integration of the resulting expression, taking account of the second condition in Eq. (6), that

$$\frac{\partial^3\eta}{\partial x_*^3} - \frac{\partial\eta}{\partial x_*} + R\frac{\partial}{\partial x_*}\left\{\eta^{-3}\left[\left(\frac{\partial\eta}{\partial x_*}\right)^2 - \theta^2\right]\right\} + \frac{\dot{x}_f}{\eta^2 + S} = 0. \tag{9}$$

Here the coordinate $x_* = x_f - x$ is measured from the boundary of the film inside the liquid. Note that, in the case of a simple wave, $\dot{x}_f = \text{const}$, Eq. (9) is the accurate corollary of Eq. (5).

The two small parameters R and S in Eq. (9) may be estimated in terms of the Van der Waals interaction constant and the surface diffusion coefficient. For moderate values of the viscosity and surface tension, it may be found that $R, S \sim 10^{-12} \ll 1$. The region of variation in film thickness of interest below is that close to the line of three-phase contact $x_* \to +0$, $\eta \to +0$, in which the role of the splitting pressure and slip is significant. To determine the relative role of the terms in Eq. (8) in this region, the new variables $\delta = x_* R^{-1/2}$, $\xi = \eta R^{-1/2}$ are introduced. Then from Eq. (9), neglecting quantities of the order O(R), the following equaltion is obtained

$$\frac{\partial^3\xi}{\partial\delta^3} + \frac{\partial}{\partial\delta}\left\{\xi^{-3}\left[\left(\frac{\partial\xi}{\partial\delta}\right)^2 - \theta^2\right]\right\} + \frac{\dot{x}_f}{\xi^2 + \varkappa} = 0, \tag{10}$$

where $\kappa = S/R \sim 1$. Equation (10) is the desired asymptotic form of Eq. (5). The independent variable δ is an internal variable [19] for the given small region of the film. The solution of Eq. (10) when $\delta \gg 1$ must be mathced with the solution of Eq. (5). Assuming that the curvature of the film surface $\partial^2\eta/\partial x^2 = O(1)$ far from the film boundary $x_* \sim 1$, the derivative $\partial^2\xi/\partial\delta^2$ must be of order $O(R^{1/2}) \ll 1$ when $\delta \gg 1$. Therefore, following the general scheme for constructing asymptotic representations [19], the following condition must be added to Eq. (10) in the first approximation

$$\frac{\partial^2\xi}{\partial\delta^2} = 0 \text{ when } \delta = \infty. \tag{11}$$

Together with the boundary conditions in Eqs. (7) and (11) and the first condition in Eq. (6), Eq. (10) asymptotically completely determines the form of the liquid film as a function of the rate of spreading $\dot{x}_f$ and the parameter κ characterizing the slip rate of the liquid at the solid surface.

2. Construction and Analysis of Integral Curves

It is expedient, for purposes of analytical and numerical analysis, to reduce the order to Eq. (10) by introducing the new variable $z = (\partial\xi/\partial\delta)^2 - \theta^2$ and to regard the film thickness ξ as an independent varible. Then Eq. (10) is rewritten in the form

$$\frac{1}{2}\frac{\partial^2 z}{\partial\xi^2} + \frac{\partial}{\partial\xi}\left(\frac{z}{\xi^2}\right) + \frac{\dot{x}_f}{\theta}\left[(\xi^2+\varkappa)\sqrt{1+z/\theta^2}\right]^{-1} = 0. \tag{12}$$

Equation (12) must be considered together with the boundary conditions

$$z = 0 \text{ when } \xi = 0, \tag{13}$$

$$\frac{\partial^2 z}{\partial\delta^2} = 0 \text{ when } \delta = \infty, \tag{14}$$

which are a consequence of the conditions in Eqs. (6), (7), and (11).

As is evident, the point $\xi = 0$ is a singular point for Eq. (12). Integrating Eq. (12), it is found that

$$\frac{1}{2}\frac{\partial z}{\partial\xi} + \frac{z}{\xi^2} + \frac{\dot{x}_f}{\theta}\int_0^{\xi}\left[(\varepsilon^2+\varkappa)\sqrt{1+z/\theta^2}\right]^{-1} d\varepsilon = C. \tag{15}$$

Here C is a constant of integration. If it is now assumed that $z > -\theta^2$ at least when $\xi \to 0$, the integral in Eq. (15) may be neglected in comparison with the constant of integration C as $\xi \to 0$. Then, Eq. (15) yields a form of Eq. (12) that is asymptotically valid as $\xi \to 0$ and when $z > -\theta^2$:

$$\frac{1}{2}\frac{\partial z}{\partial\xi} + \frac{z}{\xi^3} = C.$$

This equation is linear. Its general solution is written in the form

$$z = 2C\exp(\xi^{-2})\left[\int_0^{\xi}\exp(-\varepsilon^2)\,d\varepsilon + C_1\right]. \tag{16}$$

Here C_1 is the constant of integration. It follows from Eq. (16) that, if the function $z(\xi)$ is assumed to be finite, there is only a single-parameter family of integral curves passing through the point $\xi = 0$ corresponding to $C_1 = 0$. The boundary condition in Eq. (13) is satisfied here, i.e., as noted above, it is equivalent to the requirement of finiteness of $z(\xi)$ as $\xi \to 0$. The constant C is chosen from the condition in Eq. (14). Integration by parts of Eq. (16) shows that $z \sim C\xi^3$ as $\xi \to 0$. This asymptotic representation may be improved directly on the basis of Eq. (12), assuming that

$$z \sim \xi^3 \sum_{i=0}^{N} a_i \xi^i, \quad \xi \to 0, \tag{17}$$

where N is some natural number. Substituting Eq. (17) into Eq. (12), it is found that $a_0 = C$, $a_1 = \dot{x}_f/\theta\varkappa$, $a_2 = -3C/2$, $a_3 = \dot{x}_f(1/3 + 2\varkappa)/\theta\varkappa^2$, $a_4 = C[\dot{x}_f/(8\varkappa\theta^3) + 15/4]$. Where necessary, the series in Eq. (17) may be extended, but it is practically impossible to use this series for the calculation of $z(\xi)$ when $\xi >> 1$. Therefore, to construct the integral curve of Eq. (12) satisfying the conditions in Eqs. (13) and (14), Eq. (12) is integrated numerically by the Runge–Kutta method using a scheme of fourth-order accuracy with a variable step. The initial value of $z(\xi)$ is chosen on the basis of Eq. (17) for $\xi = 0.1$.

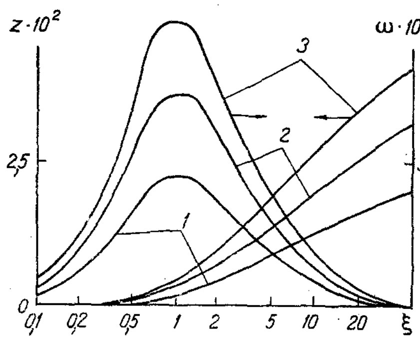

Fig. 1. Integral curves and derivatives for $\kappa = 1$ and $\dot{x}_f = 5 \cdot 10^{-4}$ (1), $9 \cdot 10^{-4}$ (2), and $13 \cdot 10^{-4}$ (3).

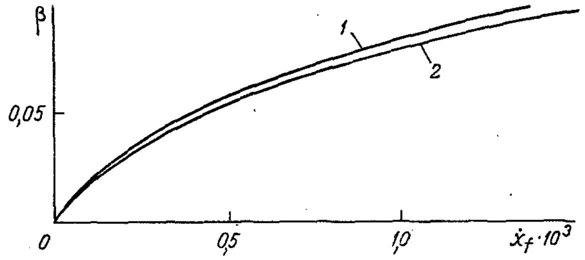

Fig. 2. Angle of slope β for $\kappa = 1/3$ (curve 1) and $\kappa = 3$ (curve 2).

Calculations show for the spreading of liquid on a solid surface $\dot{x}_f > 0$ that the condition $\delta \to \infty$ corresponds to $\xi \to \infty$; see also [6, 7]. The constant C is determined by the ranging method from the condition in Eq. (14), satisfaction of which is required when $\xi \approx 100 \gg 1$. In Fig. 1, the resulting integral curves are shown in semilogarithmic coordinates, together with the dependence of the derivative $\omega = dz/d\xi$. Analysis of the numerical results obtained shows that, close to the boundary of the spreading film $\xi = 0$ there is a thin transition layer, surface curvature of the film in which reaches considerable values. The maximum curvature corresponds to $\xi \approx 1$. Correspondingly, the angle of slope of the film surface within the limits of this narrow layer undergoes a sharp discontinuity, the magnitude of which depends on the spreading rate $\dot{x}_f$ and the parameter κ.

The dependence of the increment in the angle of slope $\beta = \partial\eta/\partial x - \theta$ when $\xi = \xi_0 = 20$ on the rate $\dot{x}_f$ is shown in Fig. 2.

Note here that the dimensionless thickness of the film $\eta = \xi\sqrt{R}$. Setting $R = 10^{-12}$ gives $\eta_0 = 2 \cdot 10^{-5} \ll 1$ for $\xi_0 = 20$. The corresponding dimensional thickness of the film $h_0 = (\sigma/\rho g)^{1/2}\eta_0$; for example, for water at normal temperature, $h_0 = 5.3 \cdot 10^{-8}$ m. As is evident, the angle β increases nonlinearly with increase in $\dot{x}_f$, in qualitative agreement with experimental data [1]. The characteristics of the variation in angle of slope of the film surface observed experimentally may be regarded as the existence of a dynamic angle of wetting depending on the rate of spreading [1, 8, 9].

At large values of ξ, the dependence $z(\xi)$ is near-logarithmic, in complete agreement with the conclusions of [6, 7] (Fig. 1). In our view, the theory developed in [6, 7] and elsewhere requires refinement in the determination of the minimum film thickness h_m at which the integration of the equations of motion of the spreading liquid begins. Evidently, h_m must be replaced here by h_m^*, defining the limit of action of the splitting pressure, and the angle of slope of the film surface at $h = h_m^*$ may be specified on the basis of asymptotic analysis analogous to that performed here.

As follows from Figs. 1 and 2 and the asymptotic representations here constructed, the angle of slope of the film of the film surface is practically the same as the wetting angle θ when $\dot{x}_f \to 0$, over the whole extent of the given range of film thickness; i.e., the solution for a moving film transforms continuously into the solution for a motionless film $z \approx 0$.

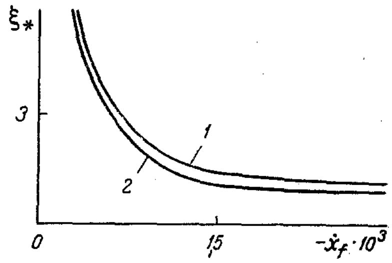

Fig. 3. Dimensionless film thickness ξ_* for $\kappa = 1$ (curve 1) and $\kappa = 0.1$ (curve 2).

For the case of liquid displacement $\dot{x}_f < 0$, calculations by Eq. (12) show that the thickness of the liquid film is finite when Eq. (14) holds. Passing to the limit as $\delta \to \infty$ in this case corresponds to $\xi \to \xi_* < \infty$. When $\xi = \xi_*$, the condition $z(\xi_*) = -\theta^2$ holds, i.e., with liquid displacement $\dot{x}_f < 0$, a thin liquid film of constant thickness $\eta = \eta_*$, $\eta_* = \xi_*\sqrt{R}$ is formed at the solid surface. The thickness of the film remaining on the solid surface ξ_* is uniquely related to the rate of motion of the film boundary $\dot{x}_f$. The dependence of ξ_* on $\dot{x}_f < 0$ calculated from Eq. (12) is shown in Fig. 3. Note that, as $\dot{x}_f \to -0$, the solution also transforms to the solution for a motionless film: $z \not\approx 0$.

Thus, the given analysis shows that the hypothesis of [12, 13] allows the spreading of a film of partially wetting liquid over a dry surface to be described in closed consistent form. A completely definite interpretation of the Young's condition and the dynamic contact angle of wetting is obtained here. The form of the liquid film close to the line of three-phase contact is asymptotically completely detrmined by the rate $\dot{x}_f$. Determining $\dot{x}_f$ itself entails considering an evolutionary problem for the initial form of the liquid film, which falls outside the scope of the asymptotic theory developed here.

NOTATION

P, pressure in liquid film; P_0, external pressure; u, v, horizontal and vertical components of liquid velocity; μ, ρ dynamic viscosity and density of liquid; y, transverse coordinate; g, accelration due to gravity; σ, surface tension.

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