

Stability in systems with moving contact lines

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An energy stability theory is formulated for systems having moving contact lines. The method derives from criteria obtained from the integral mechanical-energy balance manipulated to reflect general material and dynamical properties of moving-contact-line regions. The method yields conditions for both stability and instability and is applied to the two-dimensional Rayleigh–Taylor problem in a vertical slot.

1. Introduction

There are relatively few analyses in the literature that deal with systems containing immiscible fluids with contact lines moving along solid bounding surfaces. Analyses of the stability in such systems are rarer still. A principal reason for this lack is the existence (Dussan V. & Davis 1974) of a contact-line singularity when one makes the usual hydrodynamic assumptions, viz. Newtonian incompressible fluids, no-slip boundary conditions, and smooth, rigid, solid walls. Although the singularity can be ignored under special circumstances, it cannot be in general since the dynamics of the fluids in the immediate vicinity of moving contact lines can have substantial effects on the motions of the entire fluid. Specifically, when the characteristic lengthscale in the system is several centimeters or less, surface tension plays an important role in determining the shape of the fluid interface. If the contact angle θ , as shown in figure 1, is specified and surface tension is important, the interface shape in the far field is dependent on the local dynamics near the contact line. The present work addresses such systems.

Linear stability analysis enables one to determine sufficient conditions for instability, and the rates of growth of the disturbances. However, in order to perform such an analysis one must state explicitly the constitutive nature of the fluids as well as give a detailed description of the mechanism used to remove the singularity at the moving contact line. Further, the calculation of weakly nonlinear effects requires the consideration of interface shapes that are relatively close to those of the basic state. The present work follows a different path and has its roots in two previous analyses, those of Dussan V. (1975) and Davis (1980).

Dussan V. (1975) investigates the stability of static states in a system containing two immiscible fluids in a closed container. The stability analysis is based upon a derived form of the balance equation of mechanical energy. We make use of this equation in the present study and so present it here as applied to the system shown in figure 2:

$$\frac{d}{dt} \left\{ \int_V [\rho |\mathbf{u}|^2 + \rho \gamma] dV + \sigma A_I \right\} = - \int_V \text{tr } \mathbf{T} \cdot \mathbf{D} dV + \int_{\partial V} \mathbf{u} \cdot \mathbf{T} \cdot \mathbf{v} ds + \sigma \int_C \mathbf{U} \cdot \mathbf{m} dl. \quad (1.1)$$

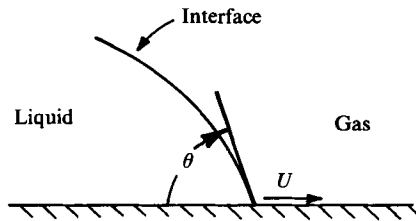


FIGURE 1. A sketch of the contact-line region where liquid displaces gas.

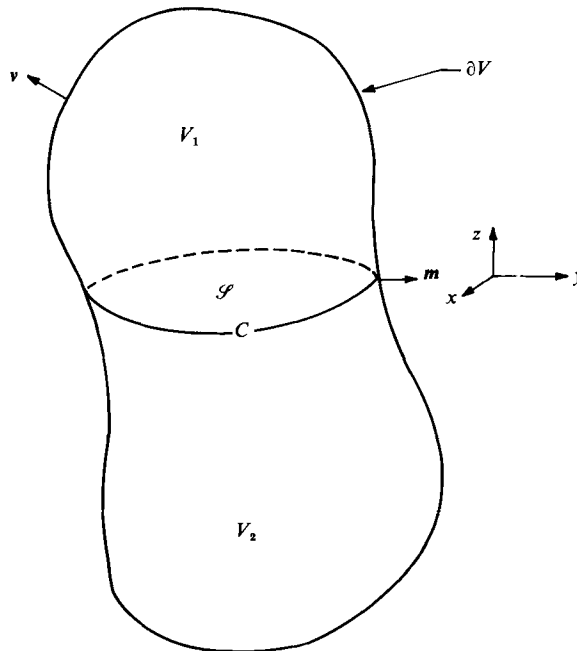


FIGURE 2. A sketch of the finite domain V separated by an interface \mathcal{S} with contact line C .

where γ is the potential per unit mass associated with the body force; σ is the constant surface tension associated with the fluid interface \mathcal{S} formed by the two immiscible fluids; \mathcal{S} has area A_1 ; ρ is the density, a different constant for each phase; \mathbf{u} is the velocity field; $V = V_1 + V_2$ is the entire fluid body with ∂V denoting the location of its surface, the inside surface of the solid container; C is the location of the contact line lying at the intersection of \mathcal{S} and ∂V ; \mathbf{U} is the velocity of the contact line; \mathbf{v} is the unit outward normal to ∂V ; \mathbf{m} is the unit normal to the contact line lying within the local tangent plane to \mathcal{S} and pointing outward from the fluid body; \mathbf{T} is the stress tensor and $\int_V \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV$ is the rate at which mechanical is converted into non-mechanical energy. The problem was further restricted by assuming a *fixed location* of the contact line, and that the fluids obey the no-slip boundary condition on ∂V . If we define

$$K = \int_V \rho |\mathbf{u}|^2 \, dV, \tag{1.2}$$

$$J = \int_V \rho \gamma \, dV + \sigma A_1, \tag{1.3}$$

then (1.1) takes the form

$$\frac{d}{dt}\{K+J\} = - \int_V \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV. \quad (1.4)$$

Sufficient conditions for both the stability and instability of *static states* are then deduced from (1.4) for systems containing fluids that can be described as dissipative, viz. $\int_V \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV > 0$. The key factors are that K is positive, and that J is a functional of only the shape of the fluid interface (its value does not depend explicitly on the velocity field of either fluid). No restrictions are made on the size of the disturbance of the fluid interface other than the exclusion of the formation of detached drops of one fluid in the other.

Davis (1980) examines the linearized stability of a static rivulet. He permits the contact lines to move and the dynamic contact angle to depend (linearly) on contact-line speed. The singularity at the moving contact line is removed by allowing the liquid to slip in a specified way at the solid boundary. He converts the partial differential equations describing the dynamics of the infinitesimal disturbances into an integral form, and shows that the system behaves like a damped linear-harmonic oscillator. He finds that the damping coefficient contains three terms, one accounting for the viscous dissipation of mechanical into non-mechanical energy for the Newtonian fluid, another reflecting the fact that mechanical energy is converted to non-mechanical energy at the solid surface due to the slip, and with the remaining term being proportional to $d\theta/dU$, adding to the dissipation of mechanical energy when $d\theta/dU$ is a positive constant.

The object of the present study is to present an approach for analysing the stability of a system that draws upon the least-limiting characteristics of the above two investigations. It will closely follow Dussan V. (1975) in that sufficient conditions will be deduced for both stability and instability of a system subjected to possibly large-amplitude disturbances through the use of the mechanical-energy balance for material systems which we shall identify as dissipative. As in Davis (1980), the contact line will be permitted to move; however, our approach will *not* depend on the details of how the singularity at the moving contact line is removed, nor on the assumption of a linear relationship between θ and U but will generalize the notion that $d\theta/dU > 0$ always contributes to the dissipation. In §2 an expression quite similar to (1.4) will be derived from (1.1) that is appropriate for systems containing *moving* contact lines. A precise definition of dissipative fluids will be presented in §3. In §4 an illustration will be given of the application of the stability criteria to systems with moving contact lines by analysing the two-dimensional Rayleigh–Taylor problem for a vertical parallel-sided slot.

2. Conservation of mechanical energy

The evaluation of the second and third terms on the right-hand side of (1.1) represents our point of departure from Dussan V. (1975). There is a great temptation to set the second term equal to zero, a consequence of the no-slip boundary condition evaluated along the inside surface of the container, and to set the third term equal to $\sigma \int_C U \cos \theta \, dl$, where θ is the contact angle defined through fluid 2. However, as already mentioned, when the usual hydrodynamic assumptions are made, a singularity (Dussan V. & Davis 1974) exists at the moving contact line. This singularity gives rise to an infinite rate of work performed by the solid on the fluid so that the second term on the right-hand side is undefined.

The primary approach taken over the past decade to remove this singularity has been to allow the fluid to slip along the solid surface. For example, one of the more popular assumptions (Huh & Mason 1977; Lowndes 1980; Hocking & Rivers 1982) has been to replace the no-slip boundary condition on the stationary walls with that introduced by Navier,

$$\mathbf{e} \cdot \mathbf{u} = -\beta \mathbf{e} \cdot \mathbf{T} \cdot \mathbf{v}. \quad (2.1)$$

Here, $\mathbf{e} = \mathbf{U}/|U|$, and β is a constant slip coefficient. When (2.1) is substituted into the second term on the right-hand side of (1.1), one gets

$$\int_{\partial V} \mathbf{u} \cdot \mathbf{T} \cdot \mathbf{v} ds = -\beta \int_{\partial V} \{\mathbf{e} \cdot \mathbf{T} \cdot \mathbf{v}\}^2 ds. \quad (2.2)$$

Another boundary condition (Huh & Mason 1977; Bach & Hassager 1985) allows the fluids instantaneously located within a small distance of the moving contact line to exert a zero shear stress on the solid, while the no-slip boundary condition is obeyed elsewhere. Under this assumption, the second term on the right-hand-side of (1.1) is identically zero.

To further complicate matters the precise value of the third term on the right-hand side of (1.1) is unknown. There is experimental evidence that leads one to question the utility of the dynamic-contact-angle data reported in the literature (Ngan & Dussan V. 1982). It seems that typical experimental techniques cannot detect the significant deformation in the fluid interface resulting from viscous forces in the immediate vicinity of the moving contact line. Thus, the rate of work done by the solid on the fluid at C , viz. the third term on the right-hand side of (1.1), cannot be evaluated accurately.

Despite all these difficulties, there does exist evidence which supports another procedure (Hansen & Toong 1971; Kafka & Dussan V. 1979; Ngan & Dussan V. 1986) for evaluating the last two terms on the right-hand side of (1.1). It relies on the view that the details of the flow, the shape of \mathcal{S} and the location of C cannot be obtained precisely by a viewer 'far' from the contact line. One excludes from consideration the fluids instantaneously located within a very small distance R of the contact line; refer to figure 3. (The Hansen & Toong procedure differs from that of Kafka & Dussan V. and Ngan & Dussan V., see Ngan 1985). This leads to a correct expression for the mechanical-energy balance as long as one can accurately account for the rate at which work is done by the fluids instantaneously occupying the excised region ΔV_R on the rest of the fluid body. Thus, one must know both the velocity field of the fluids at the 'outer boundary' S of ΔV_R , and the angle θ formed between the planes tangent to \mathcal{S} at $r = R$ and tangent to the solid surface. For the case when fluid 1 is a passive gas and fluid 2 is a Newtonian liquid, it is argued that to lowest order in R the velocity field near $r = R$ is given by

$$(u, v, w) = \left(-\frac{1}{r} \frac{\partial \psi}{\partial \phi}, \frac{\partial \psi}{\partial r}, 0 \right),$$

where

$$\begin{aligned} \left[\frac{\psi}{rU} - \sin \phi \right] [\Theta^2 - \sin^2 \Theta] &= -\phi \sin(\phi - \Theta) \sin \Theta + \phi \Theta \sin \phi - \Theta^2 \sin \phi \\ &+ \left[\frac{\sin \Theta - \Theta \sin \Theta}{\Theta - \sin \Theta \cos \Theta} \right] [\phi \Theta \sin(\phi - \Theta) + (\Theta - \phi) \sin \phi \sin \Theta] \quad (0 \leq \phi < \Phi(r, t)), \end{aligned}$$

and (r, ϕ, z) represents the local cylindrical coordinate system. The moving contact line is perceived to be located at $r = 0$, and the liquid occupies the region given by

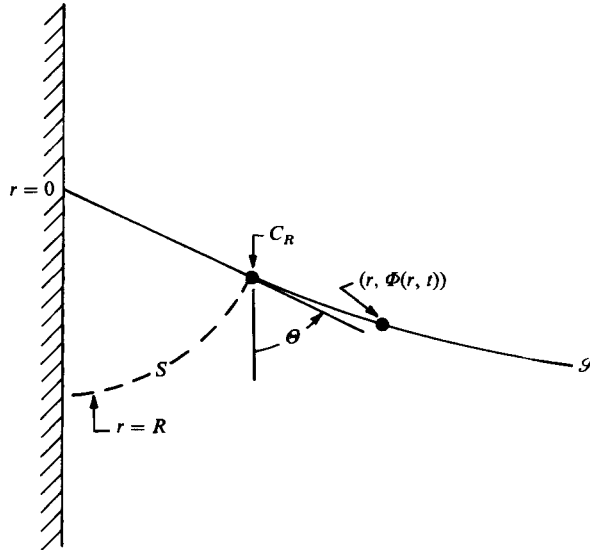


FIGURE 3. A sketch with the region ΔV_R excised. The polar coordinates of a point on \mathcal{S} are $(r, \Phi(r, t))$.

$\phi \in [0, \Phi(r, t)]$. The mechanical-energy balance for the one-fluid system takes on the approximate form

$$\frac{d}{dt} \{K + J\} = - \int_{V_R} \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV + 2\mu_2 \int_{C_R} \frac{U^2 \sin^2 \Theta \, dl}{\Theta - \sin \Theta \cos \Theta} + \sigma \int_{C_R} \cos \Theta \, dl, \quad (2.3)$$

where μ_2 is the viscosity of fluid 2. S has $r = R$ and $0 \leq \theta \leq \Theta$, $V_R = V - \Delta V_R$, and C_R denotes the intersection of S_R with \mathcal{S} . Note that when V_R is excluded, all the quantities V , \mathcal{S} and C are different by $O(R)$ from those quantities associated with the slip procedure. In what follows we shall not distinguish these slight differences. The angle Θ , equivalent to $\Phi(R, t)$, may depend on U . Thus Θ , which depends on R , and is the only parameter appearing on the right-hand side of (2.3), is experimentally measurable. The terms on the right-hand side of (1.1) are related approximately to the terms on the right-hand side of (2.3) by

$$0 = - \int_{\Delta V_R} \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV + \int_{\partial(\Delta V_R) \cap \partial V} \mathbf{u} \cdot \mathbf{T} \cdot \mathbf{v} \, ds - 2\mu_2 \int_{C_R} \frac{U^2 \sin^2 \Theta \, dl}{\Theta - \sin \Theta \cos \Theta} + \sigma \int_C U \cos \theta \, dl - \sigma \int_{C_R} U \cos \Theta \, dl = 0, \quad (2.4)$$

where it is understood that the region on the solid surface along which the fluid slips is embedded well within $\partial(\Delta V_R) \cap \partial V$, and θ is the contact angle†.

Thus far we have only addressed the problem of evaluating the right-hand side of (1.1) based upon current knowledge of the dynamics of fluids in the immediate neighbourhood of the moving contact line. The *key* to our approach for analysing the

† If the continuum theory employed omits long-range molecular forces, the contact-line region appears as shown in figure 1 and θ is the ‘microscopic’ or actual contact angle. If the continuum theory includes such forces, the interface undergoes large deformation near the contact line, even in the static state, so that figure 1 is not representative. In this case also θ is the actual contact angle.

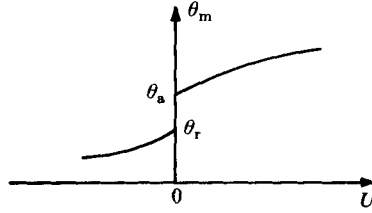


FIGURE 4. A sketch of typical measured contact angles θ_m versus contact-line speed U ; θ_a and θ_r the advancing and receding static angles respectively.

stability of the system lies in the decomposition of the third term on the right-hand side of either (1.1) or (2.3) into reversible and irreversible portions.

The general behaviour of the dynamic contact angle reported in the literature, denoted by θ_m , is illustrated in figure 4. As already mentioned, θ_m does not represent the dynamic behaviour of either θ or Θ due to the deformation of the fluid interface near the contact line by the viscous forces. However, one may anticipate that the behaviour of these three angles would coincide in the limit $U\mu_2/\sigma \rightarrow 0$, when the capillary forces dominate the viscous forces. For this reason, we assume that all three angles approach θ_a (or θ_r) as $U \rightarrow 0$, for $U > 0$ (or $U < 0$). In fact, we further restrict our attention to the case $\theta_a = \theta_r$, where contact-angle hysteresis is absent and the static contact angle is unique; we denote their common value by θ_s . If we let ϑ denote either θ or Θ and write $\cos \vartheta \equiv \cos \theta_s + (\cos \vartheta - \cos \theta_s)$, then the third term on the right-hand side in either (1.1) or (2.3) can be written as

$$\sigma \int U \cos \vartheta \, dl = \sigma \cos \theta_s \frac{dA_{2s}}{dt} + \sigma \int U \{\cos \vartheta - \cos \theta_s\} \, dl, \quad (2.5)$$

where A_{2s} denotes the wetted area of the solid/fluid 2 interface.

In summary, we can write the equation for conservation of mechanical energy in two alternative forms:

$$\frac{d}{dt} \{K + J - \sigma A_{2s} \cos \theta_s\} = - \int_V \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV + \int_{\partial V} \mathbf{u} \cdot \mathbf{T} \cdot \mathbf{v} \, ds + \sigma \int_C U \{\cos \theta - \cos \theta_s\} \, dl, \quad (2.6)$$

or,

$$\frac{d}{dt} \{K + J - \sigma A_{2s} \cos \theta_s\} = - \int_{V_R} \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV + 2\mu_2 \int_{C_R} \frac{U^2 \sin^2 \Theta \, dl}{\Theta - \sin \Theta \cos \Theta} + \sigma \int_{C_R} U \{\cos \Theta - \cos \theta_s\} \, dl. \quad (2.7)$$

These are the equations that will be used to analyse the stability of the system. Note that (2.7) applies when fluid 1 is a passive gas. It could, of course, be extended to the case of two immiscible fluids in which case the second term on the right-hand side of this equation would be augmented.

3. Dissipative materials

Our attention will be restricted to that class of materials that we shall define as *dissipative*. As indicated in Davis (1980), there are two mechanisms by which mechanical energy can be dissipated in systems with moving contact lines. It can

occur within the interior of the fluids, viz. when $\text{tr } \mathbf{T} \cdot \mathbf{D} > 0$. However, it can also occur at the moving contact line. By this we mean $I_S < 0$, and $I_R < 0$, where we define

$$I_S = \int_{\partial V} \mathbf{u} \cdot \mathbf{T} \cdot \mathbf{v} \, ds + \sigma \int_C U \{ \cos \theta - \cos \theta_s \} \, dl, \quad (3.1)$$

$$I_R = 2\mu_2 \int_{C_R} \frac{U^2 \sin^2 \Theta \, dl}{\Theta - \sin \Theta \cos \Theta} + \sigma \int_{C_R} U \{ \cos \Theta - \cos \theta_s \} \, dl. \quad (3.2)$$

Note that if $I_S < 0$, and $\text{tr } \mathbf{T} \cdot \mathbf{D} > 0$ throughout the fluids, then $I_R < 0$; see (2.4). Although one cannot yet say that all physically realistic descriptions of the dynamic behaviour of fluids near a moving contact line must necessarily be dissipative, the statement is consistent with all the information available, as discussed below.

The most thoroughly measured contributor to I_S and I_R is the variation of contact angle with U . To our knowledge there exist no dynamic-contact-angle data giving the measured contact angle θ_m as a decreasing function of U . Although this fact is consistent with

$$\int U \{ \cos \vartheta - \cos \theta_s \} \, dl \leq 0,$$

for ϑ equalling either θ or Θ , our discussion in §2 underlines the fact that θ_m is neither θ nor Θ . Thus such data in their present form are insufficient for determining either I_S or I_R directly.

As discussed in §2, there have been a number of analyses of the dynamics of systems with moving contact lines with the objective of determining the dynamic behaviour of the fluids in the immediate vicinity of the contact line by comparison with experimental data. The studies which assume the slip boundary condition of Navier (Huh & Mason, Lowndes, Hocking & Rivers), also assume that θ equals θ_s for all contact-line speeds. Each study finds, upon choosing a small constant positive value for β , varying degrees of agreement with experimental data. If $\theta > \theta_s$ and slip is given by (2.2), then (3.1) gives $I_S < 0$. However, the agreement of these analyses with experiment is not conclusive. At each speed of the contact line two parameters, θ and β , must be obtained from only one data point. Presumably, more than one family of values for θ and β could be found†.

As discussed in §2, Bach & Hassager (1985) use a slip boundary condition. They assume that the shear stress is zero within a small distance near the contact line, and that the no-slip boundary condition applies elsewhere. For specific values of θ , U , and of the size of the shear-free region, they are able to predict the experimentally measured values of θ_m in an experiment (Ngan & Dussan V. 1982) where silicone oil displaces air through slots of two different spacings. They find $\theta = 35^\circ$. Since $\theta_s = 0$ for silicone on glass in the presence of air, then $I_S < 0$.

Hansen & Toong (1971) and Ngan & Dussan V. (1986) surmise Θ from experimental measurements of the apex heights of menisci and an analysis of the dynamics of the fluids. Hansen & Toong examine Nujol displacing air in a glass capillary and find that $\Theta = 21^\circ$ for $0 \leq \mu_2 U/\sigma \leq 0.0025$; hence, $I_R > 0$, implying that mechanical energy is *generated* near C . However, more extensive measurements have been made by Ngan

† In general, a one-parameter family of values of (θ, β) will give rise to the same outer solution. Here we refer to the fact that *more than one* family of values of (θ, β) can be identified so that the analytical solution in the outer region is consistent with θ_m at each value of U . When analysing immiscible fluid displacement through a capillary, it is difficult to devise more than one experiment for a given material system for the contact line moving at a specified speed. One such set of experiments would be to perform them in a centrifuge rotating at such a high rate that the effective Bond number is much greater than unity.

Range of $\mu_2 U/\sigma$	Θ	$\frac{2\mu_2 U^2 \sin^2 \Theta}{\Theta - \sin \Theta \cos \Theta}$	$\sigma U \cos \Theta$	I_R (per unit length)
0.013–0.014	35°	0.034	–0.0977	–0.0637
0.014–0.015	38°	0.0358	–0.123	–0.0872
0.015–0.016	42°	0.0365	–0.159	–0.123

TABLE 1. Estimates of I_R from Ngan & Dussan V. (1986)

& Dussan V. (1986) of silicone oil displacing air in the narrow slot formed by two parallel microscope slides. For $0.013 < U\mu_2/\sigma < 0.016$ and for seven different gap widths d , $0.01 \text{ cm} \leq d \leq 0.12 \text{ cm}$, they find that $I_R < 0$, as shown in table 1. There is *dissipation* of mechanical energy near C . Clearly, more data is needed.

4. Sufficient conditions for stability and instability

Define

$$J = \int_V \rho\gamma \, dV + \sigma A_1 - \sigma A_{2s} \cos \theta_s, \quad (4.1)$$

and
$$\mathcal{D} = \int \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV + I_S \quad (4.2a)$$

or
$$\mathcal{D} = \int \text{tr } \mathbf{T} \cdot \mathbf{D} \, dV + I_R. \quad (4.2b)$$

If a slip model is used, (4.2a) applies, while if an exclusion method is used, (4.2b) applies. Thus, the energy balance (1.1) has the general form†

$$\frac{d}{dt}(K+J) = -\mathcal{D} < 0. \quad (4.3)$$

If the materials are dissipative, $\mathcal{D} > 0$, the theorems presented in Dussan V. (1975) apply to the present study. Here J is a functional of the shape of the inside surface of the container, as well as the fluid interface. However, the admissible class of interface shapes is now modified. Here they need only be continuously differentiable and be consistent with the fact that fluids 1 and 2 are incompressible. No restriction need be specified concerning the location of the contact line or the local values of the contact angle. As in the previous study, we restrict our attention to systems consisting of dissipative materials. Hence, the inequality in (4.3) holds.

It is instructive to illustrate the implications of the theorems cited above by analysing the two-dimensional Rayleigh–Taylor problem for a vertical slot as shown in figure 5. A formal statement of these theorems appears in Appendix A.

Let us begin by analysing the special case $\theta_s = \frac{1}{2}\pi$. For this particular problem the static basic state is given by $h = 0$, and

$$J = \int_{-\frac{1}{2}}^{\frac{1}{2}} \left[1 + \left(\frac{dh}{dx} \right)^2 \right]^{\frac{1}{2}} dx - \frac{1}{2}G \int_{-\frac{1}{2}}^{\frac{1}{2}} h^2 dx, \quad (4.4)$$

where $z = h(x)$ describes any element in the set of admissible interface shapes, dimensionless variables x and z are scaled with the gap width L , $G = (\rho_1 - \rho_2)gL^2/\sigma$

† Form (4.2b) holds only for the case where fluid 1 is a passive gas. However, we expect that the two-fluid case would be similar; in what follows we shall presume this.

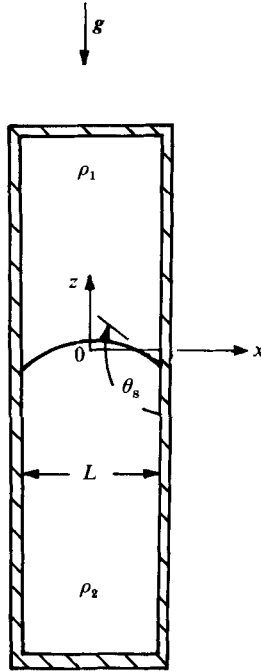


FIGURE 5. The basic state corresponding to the Rayleigh–Taylor problem in two dimensions. A heavier fluid is assumed to be above lighter immiscible fluid, $\rho_1 > \rho_2$, with L denoting the width of the slot.

is the Bond number, and J is scaled with σL . Note that (4.4) is identical with the equation for J in Dussan V. (1975) (refer to the first equation on p. 371). We integrate (4.3) to get

$$J(t) \leq K(t) + J(t) < K(0) + J(0). \tag{4.5}$$

The first inequality follows from the fact that K denotes the kinetic energy of the system, see (1.2), and is hence non-negative. Thus, we have shown that the instantaneous value of J has an upper bound $K(0) + J(0)$.

We next parameterize the elements in the set of admissible interface shapes by their L^2 -norms ϵ , where $\epsilon^2 = \int_{-\frac{1}{2}}^{\frac{1}{2}} h^2 dx$. It is convenient to define a normalized form μ of each interface shape by $h = \epsilon\mu$, and $\int_{-\frac{1}{2}}^{\frac{1}{2}} \mu^2 dx = 1$.

In order to determine sufficient conditions for stability and instability, the solution must be obtained to the variational problem that follows.

Variational problem

For each fixed value of ϵ^2 determine the function $\mu_m(x; \epsilon^2)$ that minimizes the value of J :

$$J = \int_{-\frac{1}{2}}^{\frac{1}{2}} \left[1 + \epsilon^2 \left(\frac{d\mu}{dx} \right)^2 \right]^{\frac{1}{2}} dx - \frac{1}{2} G \epsilon^2 \int_{-\frac{1}{2}}^{\frac{1}{2}} \mu^2 dx, \tag{4.6a}$$

subject to the constraints

$$\int_{-\frac{1}{2}}^{\frac{1}{2}} \mu dx = 0, \quad \int_{-\frac{1}{2}}^{\frac{1}{2}} \mu^2 dx = 1. \tag{4.6b}$$

It is straightforward to use *a priori* estimates (Joseph 1976, p. 255 ff) to find that

$$\epsilon - \frac{1}{2} G \epsilon^2 < J_m < [1 + \pi^2 \epsilon^2]^{\frac{1}{2}} - \frac{1}{2} G \epsilon^2. \tag{4.7}$$

This implies a form of J_m illustrated in figure 6. The Lagrange–Euler equation and boundary conditions associated with the variational problem stated above are as follows:

$$\frac{d}{dx} \left\{ \left[1 + \epsilon^2 \left(\frac{d\mu_m}{dx} \right)^2 \right]^{-\frac{1}{2}} \frac{d\mu_m}{dx} \right\} + \lambda \mu_m + \alpha = 0 \quad \text{for } -\frac{1}{2} \leq x \leq \frac{1}{2}, \quad (4.8a)$$

$$\frac{d\mu_m}{dx} = 0 \quad \text{at } x = \pm \frac{1}{2}, \quad (4.8b)$$

$$\int_{-\frac{1}{2}}^{\frac{1}{2}} \mu_m dx = 0, \quad \int_{-\frac{1}{2}}^{\frac{1}{2}} \mu_m^2 dx = 1, \quad (4.8c)$$

where λ and α represent Lagrange multipliers. The solution to this boundary-value problem can be obtained numerically or in approximate form as a perturbation series in powers of ϵ .

The sufficient conditions for stability and instability follow directly from the dependence of J_m on ϵ^2 and G . Since the static basic state consists of a flat interface, it is characterized by $J = 1$ and $\epsilon^2 = 0$, for all values of G . Let us define $J_m^*(G)$ as the maximum value of J_m for $0 \leq \epsilon^2 < \infty$, and $\epsilon_*^2(G)$ as the value of ϵ^2 satisfying $J_m^*(G) = J_m(\epsilon_*^2, G)$. From figure 6 we see that

$$\left. \begin{aligned} J_m^* \rightarrow \infty, \epsilon_*^2 \rightarrow \infty & \quad \text{for } G < 0, \\ 1 < J_m^* < \infty, \quad 0 < \epsilon_*^2 < \infty & \quad \text{for } 0 \leq G < \pi^2, \\ J_m^* = 1, \quad \epsilon_*^2 = 0 & \quad \text{for } G > \pi^2. \end{aligned} \right\} \quad (4.9)$$

If the system is disturbed in such a way that $J(0) < J_m^* - K(0)$ and $\epsilon^2(0) < \epsilon_*^2$, then the system will return to its basic state, the configuration available with the lowest potential energy. This follows from the fact that the instantaneous value of J associated with the system as it dynamically responds to the initial disturbance must obey inequality (4.5); hence $J(t) < J_m^*$. On the other hand, from the definition of J_m we have that $J_m < J(t)$. Thus, we can draw several conclusions:

- (i) The system is stable to any initial disturbance when $G < 0$.
- (ii) The system is stable to disturbances of only limited size when $0 < G < \pi^2$. These include disturbances such that $J(0) < J_m^* - K(0)$ and $\epsilon^2(0) < \epsilon_*^2$.
- (iii) No initial disturbances fulfill the above stated criterion for stability when $G > \pi^2$.

If the system is disturbed in such a way that $J(0) < J_m^* - K(0)$ and $\epsilon^2(0) > \epsilon_*^2$, then it is *impossible* for the system to return to its basic state. No such initial disturbances exist when $G < 0$, but they do exist when $G > 0$.

Let us return to the more general case when $\theta_s \neq \frac{1}{2}\pi$. The static basic state no longer corresponds to a flat surface. The function J has the generalized form

$$J = \int_{-\frac{1}{2}}^{\frac{1}{2}} \left[1 + \epsilon^2 \left(\frac{d\mu}{dx} \right)^2 \right]^{\frac{1}{2}} dx - \frac{1}{2} G \epsilon^2 - \epsilon \left[\mu \left(-\frac{1}{2} \right) + \mu \left(\frac{1}{2} \right) \right] \cos \theta_s \quad (4.10a)$$

and μ satisfies the constraints

$$\int_{-\frac{1}{2}}^{\frac{1}{2}} \mu dx = 0, \quad \int_{-\frac{1}{2}}^{\frac{1}{2}} \mu^2 dx = 1. \quad (4.10b)$$

The variational problem is the same as that in (4.6) but now has the modified

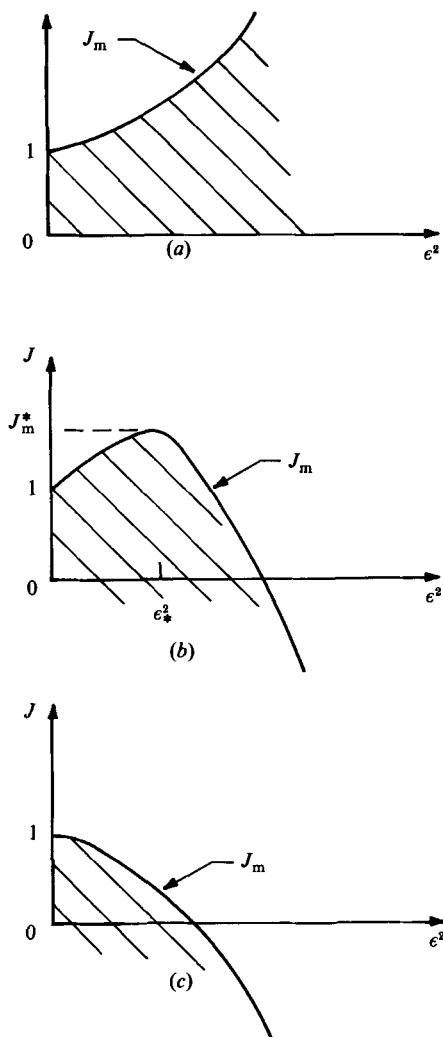


FIGURE 6. A sketch of J_m for the case $\theta_s = \frac{1}{2}\pi$ for the three ranges of G : (a) $G > 0$; (b) $0 < G < \pi^2$; (c) $\pi^2 < G$. The shaded regions represent values of J for which no admissible configuration exists.

definition (4.10a). The corresponding Lagrange–Euler equation and boundary conditions are as follows:

$$\frac{d}{dx} \left\{ \left[1 + e^2 \left(\frac{d\mu_m}{dx} \right)^2 \right]^{-\frac{1}{2}} \frac{d\mu_m}{dx} \right\} + \lambda \mu_m + \alpha = 0 \quad \text{for } -\frac{1}{2} < x < \frac{1}{2}, \quad (4.11a)$$

$$e \frac{d\mu_m}{dx} = \pm \tan(\theta_s - \frac{1}{2}\pi) \quad \text{at } x = \mp \frac{1}{2}, \quad (4.11b)$$

$$\int_{-\frac{1}{2}}^{\frac{1}{2}} \mu_m dx = 0, \quad \int_{-\frac{1}{2}}^{\frac{1}{2}} \mu_m^2 dx = 1, \quad (4.11c)$$

where again, λ and α are Lagrange multipliers. The general solution for μ_m can be expressed in terms of elliptic functions; however, the unknown constants must be evaluated numerically. Since the purpose of this discussion is demonstrative, we write

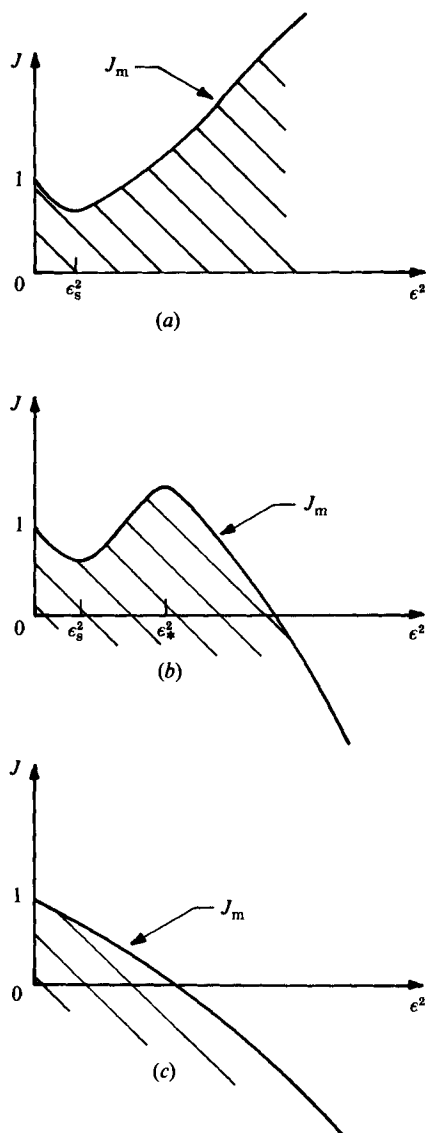


FIGURE 7. A sketch of the anticipated form of J_m when $\theta_s \neq \frac{1}{2}\pi$ for the three ranges of G : (a) $G < 0$; (b) $0 < G < G_c$; (c) $G_c < G$. The shaded regions represent values of J for which no admissible configuration exists. The quantities ϵ_s^2 and ϵ_*^2 denote the values of ϵ^2 corresponding to the basic state and the local maximum value of $J_m > 1$.

the first two terms of an asymptotic expansion valid in the limit as $\tan(\theta_s - \frac{1}{2}\pi) \rightarrow 0$ as derived in Appendix B. Though, the expansion is not uniform in ϵ^2 for large ϵ , we can sketch in figure 7 the anticipated global behaviour of J_m based upon the known global characteristics of J_m when $\theta_s = \frac{1}{2}\pi$, and the asymptotic solution valid for small values of $|\theta_s - \frac{1}{2}\pi|$. Define G_c to be the critical value of G obtained from linear instability theory of the basic state with $\theta_s \neq \frac{1}{2}\pi$.

The results shown in figures 6 and 7 can be interpreted using bifurcation theory.

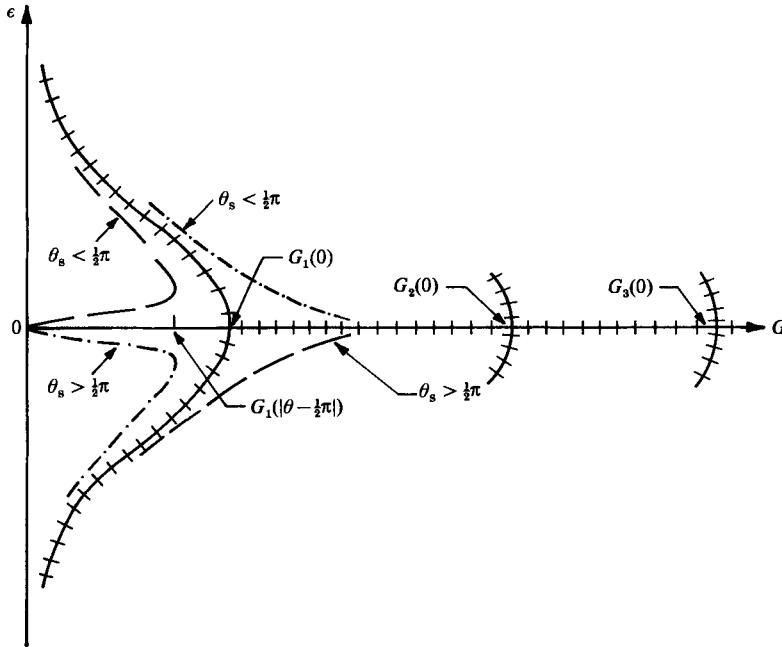


FIGURE 8. A sketch of the anticipated bifurcation picture for two-dimensional Rayleigh–Taylor instability. When $\theta_s = \frac{1}{2}\pi$, there are bifurcation points at $G_n(0)$ from which downward bifurcation emerge. Hatches denote unstable branches. When $\theta_s > \frac{1}{2}\pi$, the branch splits into two parts (———). When $\theta_s < \frac{1}{2}\pi$, the branch splits into two parts (— · — · —).

For $\theta_s = \frac{1}{2}\pi$, the inviscid linearized theory gives bifurcation points at $G = G_n(0) = (2n-1)^2\pi^2$ for $n = 1, 2, 3, \dots$, as shown in figure 8. If these bifurcations are subcritical as shown, then the global branch emanating from $G_1(0)$ should be as shown in figure 8. It begins at $\epsilon = 0$, $G = G_1(0)$ and approaches $\epsilon \rightarrow \infty$ as $G \rightarrow 0$, consistent with (4.8) above. The planar-interface basic state is unstable to infinitesimal disturbances for $G > G_1(0)$, but the whole subcritical branch for $0 < G < G_1(0)$ is also unstable and serves as the threshold amplitude above which the basic state is also unstable. This threshold amplitude corresponds to $\epsilon = \epsilon_*$ above.

For $\theta_s \neq \frac{1}{2}\pi$ the basic state has $\epsilon \neq 0$ as shown in figure 8. The dashed curves show the cases for the two signs of $\theta_s - \frac{1}{2}\pi$. As G is increased from zero, a dashed curve is followed but cannot proceed further than the limit point at $G = G_1(|\theta_s - \frac{1}{2}\pi|)$ after which the basic state no longer exists. When we examine the inviscid linearized stability of this solution we find that stability is lost at $G \sim \pi^2[1 - \frac{3}{8}(\theta_s - \frac{1}{2}\pi)^2]$ for $\theta_s \rightarrow \frac{1}{2}\pi$, showing that, consistent with figure 8, both a slight bulging up ($\theta_s > \frac{1}{2}\pi$) and down ($\theta_s < \frac{1}{2}\pi$) of the interface in the basic state give less stable states than the case $\theta_s = \frac{1}{2}\pi$. The bulging makes the bifurcation imperfect.

Recall that if $\mathcal{D} > 0$, then the stability and instability conditions from the energy theory do not depend on the viscosities of the fluid (or even on their constitutive behaviour). Given this independence, it is natural and certainly simpler to compare the results with those of linear inviscid theory; we have done this. Viscosity should affect the dissipation and modify the growth rates but not the threshold conditions, as in the case of the capillary breakup of a Rayleigh jet.

5. Discussion

In this paper we present a stability theory for static-equilibrium basic states tailored to systems containing more than one immiscible fluid† and having *moving contact lines*. We have generalized the variational technique of Dussan V. (1975), which is restricted to fixed contact lines, and Davis (1980), who performs a linear stability analysis of static rivulets with moving contact lines. The key to the approach is the decomposition of the rate of work associated with the contact line into reversible and irreversible portions. We restrict our attention to systems consisting of dissipative materials and extend the definition of dissipation to include the familiar rate of loss of mechanical energy in the interior of the fluid, as well as at moving contact lines, the latter being a consequence of contact angle being an increasing function of contact-line speed.

The theory identifies minima J_m of a potential-energy functional J as a means for determining *sufficient conditions* for both the *stability* and *instability* of the system. The functional includes effects of a conservative body force, the surface tension of the fluid–fluid interface, and the unique static contact angle. These minima J_m are taken over sets of admissible functions and are parameterized by the L^2 -norm ϵ of the interface shape for fixed values of the Bond number G . An examination of J_m as a function of ϵ^2 determines the nonlinear stability behaviour of the system.

Thus, for static basic states, one is able to by-pass the solution of a boundary-value problem for the velocity, pressure and interface shape for a viscous fluid in favour of the variational approach. The energy method posed here stands as an alternative to linear and bifurcation theories and is capable of dealing with strongly nonlinear systems. The nonlinearity enters through the size of the disturbance and the fact that $\lim_{U \rightarrow 0^+} (d\vartheta/dU)$ need not equal $\lim_{U \rightarrow 0^-} (d\vartheta/dU)$, where ϑ denotes either the actual contact angle θ or the experimentally measurable angle Θ . This method has broad applications beyond the Rayleigh–Taylor problem.

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Appendix A

The two theorems appearing in Dussan V. (1975) apply to the present study. They are as follows:

THEOREM 1. *If, over the set of admissible interfacial configurations, the functional J is bounded from below, and if there exists a positive number q^2 such that $\mathcal{D} > q^2 K$, then $\lim_{t \rightarrow \infty} \int_t^{t+T} K(\tau) d\tau = 0$ for any $T > 0$.*

THEOREM 2. *Let $M = \inf_{h \in H} \sup_{s \in [0, s_0]} J$. For a particular disturbance if for any $\epsilon > 0$, $K(0) + J(0) < M - \epsilon$, then the system cannot return to its basic state.*

Here H denotes the set of all admissible one-parameter families of elements in the set containing all admissible fluid-interface configurations. The variable s is a continuous real variable contained within the interval $[0, s_0]$ such that the interface corresponding to $s = 0$ represents its initial configuration, and the fluid interface corresponding to s_0 represents its configuration in its basic state. The value s_0 is not a fixed number, and may be infinite. In the Rayleigh–Taylor problem $J_m^* = M$.

† In part of the analysis for purposes of simplicity we took one fluid to be a passive gas. We expect the results to apply to the two-fluid case.

Appendix B

We seek a solution to (4.10a) in terms of an asymptotic expansion in $\tan(\theta_s - \frac{1}{2}\pi)$ as $\theta_s \rightarrow \frac{1}{2}\pi$, of the form

$$\begin{aligned}\mu_m &= \mu_0(x) + \tan^2(\theta_s - \frac{1}{2}\pi)\mu_2(x) + \dots, \\ \lambda &= \lambda_0 + \tan^2(\theta_s - \frac{1}{2}\pi)\lambda_2 + \dots, \\ \alpha &= \alpha_0 + \tan^2(\theta_s - \frac{1}{2}\pi)\alpha_2 + \dots, \\ J_m &= J_{m0} + \tan^2(\theta_s - \frac{1}{2}\pi)J_{m2} + \tan^4(\theta_s - \frac{1}{2}\pi)J_{m4} + \dots,\end{aligned}$$

with δ held constant, where $\delta \equiv \epsilon/\tan(\theta_s - \frac{1}{2}\pi)$. Two classes of principal eigenfunctions emerge: class 1 for which

$$\mu_0 = \frac{\cos \Gamma x}{\Gamma \sin \frac{1}{2}\Gamma} - \frac{2}{\Gamma^2}, \quad (\text{B } 1a)$$

with

$$\delta^2 = \frac{\Gamma + \sin \Gamma}{2\Gamma^3 \sin^2 \frac{1}{2}\Gamma} - \frac{4}{\Gamma^4}, \quad (\text{B } 1b)$$

and class 2 for which

$$\mu_0 = \frac{1}{\pi^2} (2\delta^2 \pi^4 - \pi^2 + 8)^{\frac{1}{2}} \sin \pi x - \frac{2}{\pi^2}, \quad (\text{B } 2a)$$

with

$$\Gamma = \pi. \quad (\text{B } 2b)$$

In the above we have written $\Gamma = \lambda_0^{\frac{1}{2}}$.

When $0 < \delta < 1/\sqrt{180}$, case 1 applies and λ_0 is real, ranging from $-\infty$ to 0. Here

$$\begin{aligned}J_{m0} &= 1, \\ J_{m2} &= \frac{4}{|\Gamma|^2} - \frac{|\Gamma| + 3 \sinh |\Gamma|}{4|\Gamma| \sinh^2 \frac{1}{2}|\Gamma|} - \frac{1}{2}G\delta^2, \\ J_{m4} &= \frac{\cosh \frac{1}{2}|\Gamma|}{|\Gamma| \sinh \frac{1}{2}|\Gamma|} - \frac{2}{|\Gamma|^2} + \frac{8 \sinh |\Gamma| - 6|\Gamma| - \sinh 2|\Gamma|}{128|\Gamma| \sin^4 \frac{1}{2}|\Gamma|},\end{aligned}$$

where $|\Gamma| = (-\lambda_0)^{\frac{1}{2}}$.

When $1/\sqrt{180} < \delta \lesssim 0.9796$, case 1 applies and λ_0 is real, ranging from 0 to π^2 . Here

$$\begin{aligned}J_{m0} &= 1, \\ J_{m2} &= \frac{3 \sin \Gamma + \Gamma}{4\Gamma \sin^2 \frac{1}{2}\Gamma} - \frac{4}{\Gamma^2} - \frac{1}{2}G\delta^2, \\ J_{m4} &= \frac{2}{\Gamma^2} - \frac{\cos \frac{1}{2}\Gamma}{\Gamma \sin \frac{1}{2}\Gamma} + \frac{8 \sin \Gamma - 6\Gamma - \sin 2\Gamma}{128\Gamma \sin^4 \frac{1}{2}\Gamma}.\end{aligned}$$

When $0.0976 \lesssim \delta < \infty$, case 2 applies. Here

$$\begin{aligned}J_{m0} &= 1, \\ J_{m2} &= \frac{2}{\pi^2} + \frac{1}{2}\delta^2(\pi^2 - G), \\ J_{m4} &= \frac{2}{\pi^2} - \frac{3}{\pi^4} \left(1 + \frac{\pi^4 \delta^2}{4}\right)^2.\end{aligned}$$

REFERENCES

- BACH, P. & HASSAGER, O. 1985 An algorithm for the use of the Lagrangian specification in Newtonian fluid mechanics and applications to free-surface flows. *J. Fluid Mech.* **152**, 173–190.
- DAVIS, S. H. 1980 Moving contact lines and rivulet instabilities. Part 1. The static rivulet. *J. Fluid Mech.* **98**, 225–240.
- DUSSAN V., E. B. 1975 Hydrodynamic stability and instability of fluid systems with interfaces. *Arch. Rat. Mech. Anal.* **57**, 363–379.
- DUSSAN V., E. B. & DAVIS, S. H. 1974 On the motion of a fluid–fluid interface along a solid surface. *J. Fluid Mech.* **65**, 71–95.
- HANSEN, R. J. & TOONG, T. Y. 1971 Dynamic contact angle and its relationship to forces of hydrodynamic origin. *J. Colloid Interface Sci.* **37**, 196–207.
- HOCKING, L. M. & RIVERS, A. D. 1982 The spreading of a drop by capillary action. *J. Fluid Mech.* **121**, 425–442.
- HUH, C. & MASON, S. G. 1977 Effects of surface roughness on wetting (theoretical). *J. Colloid Interface Sci.* **60**, 11–38.
- JOSEPH, D. D. 1976 *Stability of Fluid Motions II*, p. 255ff. Springer.
- KAFKA, F. Y. & DUSSAN V., E. B. 1979 On the interpretation of dynamic contact angles in capillaries. *J. Fluid Mech.* **95**, 539–565.
- LOWNDES, J. 1980 The numerical simulation of the steady movement of a fluid meniscus in a capillary tube. *J. Fluid Mech.* **101**, 631–645.
- NGAN, C. G. 1985 An assessment of the proper modeling assumptions for the spreading of liquids on solid surfaces. Ph.D. thesis, University of Pennsylvania, Department of Chemical Engineering.
- NGAN, C. G. & DUSSAN V., E. B. 1982 On the nature of the dynamic contact angle: an experimental study. *J. Fluid Mech.* **118**, 27–40.
- NGAN, C. G. & DUSSAN V., E. B. 1986 The moving contact line. (pending publication).