

# On the motion of a small viscous droplet that wets a surface

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A model for the movement of a small viscous droplet on a surface is constructed that is based on the lubrication equations and uses the dynamic contact angle to describe the forces acting on the fluid at the contact line. The problems analysed are: the spreading or retraction of a circular droplet; the advance of a thin two-dimensional layer; the creeping of a droplet or cell on a coated surface to a region of greater adhesion; the distortion of droplet shape owing to surface contamination. Relevant biological problems concerning cell movement and adhesion are described.

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## 1. Introduction

Although we shall examine a purely fluid problem of droplet motion, the motivation for this work derives from an investigation of the analogous biological phenomenon of cell spreading. Since the theory is shaped to this end some knowledge of cell processes is necessary.

The life cycle of a normal cell cultured *in vitro* is marked by extreme changes in shape that seem to be intrinsically related to basic metabolic functions (Folkman & Greenspan 1976). A cell, which is globular and almost spherical during mitosis, is otherwise very flat and spread extensively on the substratum. Both configurations are required: the normal cell must be flat for a substantial time in order to survive (in contrast to the abnormal or cancerous cell), but 'round-up' is essential for division and reproduction. For example, an eighteen-hour cell cycle might consist typically of a long, sixteen-hour period in the flat, spread state during which time materials are accumulated and synthesized, followed by round-up to a sphere that lasts half an hour, division into two daughter cells (spheres) in, say, one hour and a spread once again to the pancake geometry in another half hour.

Interfacial tensions between membrane, substratum and medium are known to be involved in cell motility (Carter 1967) and tissue organization (Steinberg 1972) and they must also be a part of the physical mechanism of spreading and round-up. Although the cell is far too complex a structure for any exact analogy with the wetting of a surface by an oil droplet, neither can it be immune to the action of interfacial forces. Indeed, the descriptions and photographs of the early stages in the spreading of a cell given by Bragina, Vasiliev & Gelfand (1976), and Pegrum & Maroudas (1975) and the dependence of cell attachment and the degree of its flattening on chemical factors and on the composition of the solid substratum (noted by the latter authors) are forceful arguments for such an analogy. Carter (1967) reported that a cell on a palladium-coated cellulose acetate surface moves in the direction of greater adherence (i.e. greater palladium concentration) and this too seems comparable to the motion of an oil globule caused by variable surface tension.

A fluid-dynamical model of the cell is required to describe round-up, division (see Greenspan 1977) and spread and to comprehend the observed correlations of form and function during these phases. In addition, a model of this kind can be used to examine cell motility on the substratum as well as to initiate a more ambitious attack on tissue fluid dynamics in general. A very important theoretical objective is the ability to differentiate, at some level of understanding, between cell phenomena that depend mainly on inanimate fluid mechanisms, instabilities for example, and those extraordinary cell processes that characterize life. The chemical system of the cell is miraculous and can be invoked (like a Maxwell demon) to explain almost any activity or observation. If, however, an inanimate and quite ordinary fluid mechanism accounts as well for particular events then there is much less reason for a complex life process to the same end. Fluid-dynamical studies of cell biology are certain to provide insight into and understanding of the motion of protoplasm (where too often, at present, movement is attributed solely to the contraction of polymerized fibres).

Hydrodynamics on the microscale is a largely unexplored area (see Batchelor 1976) and the paucity of data on the rheology of protoplasm compounds the difficulties that confront the theoretician. In these circumstances, it is propitious to examine the features of cellular motion and behaviour that can be simulated by liquid droplets with approximately the same gross physical properties, as expressed by the appropriate dimensionless numbers. To be specific, we consider the adhesion, spreading and movement on a plane surface of a very small, very viscous droplet whose physical parameters are as follows: kinematic viscosity,  $\nu > 0.1 \text{ cm}^2/\text{s}$  (Hiramoto 1967); density,  $\rho \simeq 1 \text{ g/cm}^3$ ; interfacial tension,  $\sigma \leq 0.1 \text{ dyne/cm}$  (Harvey 1954); diameter,  $2a \simeq 2 \times 10^{-3} \text{ cm}$ . Furthermore, the photographs of Bragina *et al.* (1976) indicate that the static contact angle of a fibroblast cell fully spread on glass is in the range  $10\text{--}20^\circ$ . The velocity of the advancing contact line is approximately  $U = 10^{-6} \text{ cm/s}$ , a value calculated from a  $20 \mu\text{m}$  increase in the diameter of a cell in, say, half an hour. In essence then the fluid problem analogous to the spread of a cell concerns a deformable droplet and low Reynolds number motion that is produced by surface-tension forces at the contact line. Effects of the ambient, less viscous medium are neglected.

## 2. Formulation

Any attempt to analyse the simply described problem of a self-spreading (or moving) droplet on a plane surface encounters formidable theoretical difficulties. Since motion is controlled by forces *at* the fluid/solid contact line, there is a serious question as to how these local molecular processes are to be represented within a macroscopic continuum theory. In static situations, the contact angle at the interface is the classical means of describing the balance of surface-tension forces there. The dynamic contact angle, which is the instantaneous slope angle at a moving edge, is the most natural extension of this concept.

Experiments on advancing contact lines and dynamic contact angles (Ghiradella & Radigan 1975; Schwartz & Tejada 1972) indicate that the process by which fluid molecules displace those of the surrounding medium in adhering to the surface involves an unseen protruding lip of fluid only Ångströms thick. Blake & Haynes (1969), in a study of dynamic contact angles, described the fluid motion within a narrow zone about the contact line as a sliding of molecules along the solid surface. Their

analysis, which is based on the theory of absolute reaction rates and is applicable to slow flows, gave the velocity of the contact line as a function of  $\cos \theta_s - \cos \theta$ , where  $\theta_s$  and  $\theta$  are the static and (observed) dynamic contact angles. The validity of this relationship at low velocities is confirmed by the experiments of Schwartz & Tejada (1972) and Hoffman (1975), who by means of a shift factor reduced the test data from various fluids to a single curve of velocity *vs.* contact angle.

Hydrodynamical studies by Huh & Scriven (1971), Dussan V. & Davis (1974), Dussan V. (1976) and Hocking (1976) show that when a fluid is allowed to slip on the surface in the immediate neighbourhood of the contact line previously unacceptable results regarding the magnitude of the shear force at the edge can be corrected. In other words, the usual no-slip condition of continuum theory must be modified near the site of adherence. Indeed, slippage in this region yields finite shear stresses there and only a negligibly small correction to the flow field elsewhere, a result confirmed again shortly.

The dynamic contact angle calculated from a macroscopic theory can be a very sensitive mathematical abstraction that is unrelated to the angle actually observed and measured in experiments. However, this is not the case for the particular class of problems considered here. The conditions assumed imply that the theoretical dynamic contact angle is only slightly dependent on the slippage near the liquid/solid boundary and as such the concept seems well founded and applicable. In anticipation of this result and the restrictions that the dynamic and static contact angles  $\theta$  and  $\theta_s$  are small with  $|\theta - \theta_s| \ll \theta_s$ , the velocity of a fluid particle at the contact line is assumed to be

$$\mathbf{q}_e = \kappa(\theta - \theta_s) \hat{\mathbf{n}},$$

where  $\hat{\mathbf{n}}$  is the outward unit normal to the boundary curve  $\Gamma$  in the plane of the surface. Since

$$\cos \theta_s - \cos \theta \simeq (\sin \theta_s) (\theta - \theta_s)$$

this is consistent with the formula derived by Blake & Haynes (1969).

Let

$$z = h(x, y, t) \quad (2.1)$$

describe the free surface of the droplet so that  $\Gamma$  is defined by

$$h(x_e, y_e, t) = 0$$

and

$$\hat{\mathbf{n}} = -\nabla h / |\nabla h| \quad \text{at} \quad h = 0.$$

For small contact angles

$$\theta_s \simeq \tan \theta_s, \quad \theta \simeq \tan \theta = |\nabla h|$$

and we can then write

$$\mathbf{q}_e = -\kappa(\nabla h) (1 - \theta_s / |\nabla h|) |_{h=0}. \quad (2.2)$$

Since

$$\mathbf{q}_e = \frac{dx_e}{dt} \hat{\mathbf{i}} + \frac{dy_e}{dt} \hat{\mathbf{j}} = \dot{x}_e \hat{\mathbf{i}} + \dot{y}_e \hat{\mathbf{j}}, \quad (2.3)$$

(2.2) provides two differential equations for the Lagrangian co-ordinates of a point on the boundary whose integration gives the locus of the particle and hence the position of the contact line at a later time  $t$ .

The intrinsic nonlinearity of fluid flow within a deforming geometry makes for an interesting but almost intractable analytical problem which is also very difficult to

solve numerically. That motion is produced by forces at the contact line, where there is zero mass in a continuum theory, only further complicates the problem. Fortunately, the specific conditions of cell motility described earlier, and of analogous fluid problems which concern the slow motion of a small, flat, viscous droplet, are exactly those for which lubrication theory is known to be an adequate approximation. Basically the physical situation is one for which viscous stresses and surface-tension forces are predominant, inertial effects are unimportant and the fluid is always in a state of near equilibrium. However, the evolution of the fluid domain with time means that even the simplified dynamical problem is still highly nonlinear.

Lubrication theory consists of a depth-averaged equation of mass conservation and a simplified form of the Navier–Stokes equations that is appropriate for a thin layer of very viscous fluid. In the approximate momentum law, the pressure gradient is horizontal and balanced entirely by the vertical shear of the horizontal velocity  $\mathbf{q}_H = \mathbf{q} - (\mathbf{q} \cdot \hat{\mathbf{k}}) \hat{\mathbf{k}}$ , so that

$$p = p(x, y, t), \quad \nabla_H p = \mu \partial^2 \mathbf{q}_H / \partial z^2. \quad (2.4)$$

The conservation-of-mass equation when averaged vertically across the thickness  $h$  of the drop or layer is

$$\partial h / \partial t + \nabla_H \cdot (h \mathbf{Q}) = 0, \quad (2.5)$$

where

$$\mathbf{Q} = \frac{1}{h} \int_0^h \mathbf{q}_H dz. \quad (2.6)$$

The pressure at  $z = h(x, y, t)$  (and hence for  $0 \leq z \leq h$ ) is equal to the capillary pressure, which is the product of the interfacial tension  $\sigma$  and the mean curvature of the free surface. For a fairly flat geometry, the balance is given by

$$p = p(x, y, t) = -\sigma \nabla^2 h. \quad (2.7)$$

On the free surface, the horizontal shear stress is assumed to be zero,

$$\partial \mathbf{q}_H / \partial z = 0 \quad (2.8)$$

(the effects of a surfactant or variable surface tension could be introduced at this stage), whereas on the solid boundary  $z = 0$

$$\kappa(h) \partial \mathbf{q}_H / \partial z = \mathbf{q}_H. \quad (2.9)$$

The slip coefficient function  $\kappa(h)$  is essentially zero except in the vicinity of a contact line when the thickness  $h$  of the layer is less than say  $O(1000 \text{ \AA})$ . (Alternatively, a nearby contact line is implied whenever  $h$  is so small.) The explicit form used is

$$\kappa(h) = \alpha / 3h, \quad (2.10)$$

where  $\alpha$  is a small number,  $O(10^{-10} \text{ cm}^2)$  or less is a likely range. [Other, more general formulae for the slip coefficient within a lubrication theory are given by Huh & Scriven (1971). Neogi & Miller (1976) introduce an effective wall velocity for the spreading fluid that is related to the porosity and roughness of the substratum, but formally this turns out to be equivalent to (2.10).] Finally, the mass or volume of the droplet remains a constant  $V_i$  as its shape changes and this implies

$$\iint_{\mathcal{A}} h dx dy = V_i, \quad (2.11)$$

where  $\mathcal{A}$  is the area within the contact line.

Vertical integration of (2.4) incorporating conditions (2.8) and (2.9) yields

$$\mathbf{q}_H = \mu^{-1} (\nabla_H p) \left( \frac{1}{2} z^2 - hz - \kappa(h) h \right),$$

from which it follows that

$$\mathbf{Q} = -\mu^{-1} (\nabla_H p) \left( \frac{1}{3} h^2 + \kappa(h) h \right),$$

or by (2.7) and (2.10)

$$\mathbf{Q} = (\sigma/3\mu) (h^2 + \alpha) \nabla(\nabla^2 h). \quad (2.12)$$

The substitution of this formula for the depth-averaged velocity in (2.5) leads to a single equation for the thickness of the droplet:

$$\partial h / \partial t + (\sigma/3\mu) \nabla \cdot [h(h^2 + \alpha) \nabla(\nabla^2 h)] = 0. \quad (2.13)$$

This must be solved, subject to initial conditions, within the time-varying domain bounded by the contour  $\Gamma = h(x_e(t), y_e(t), t) = 0$ , which moves in accordance with

$$\mathbf{q}_e = \mathbf{Q}|_{h=0} = \frac{\alpha\sigma}{3\mu} \nabla(\nabla^2 h)|_{h=0} = -\kappa(\nabla h) \left( 1 - \frac{\theta_s}{|\nabla h|} \right) \Big|_{h=0}. \quad (2.14)$$

The shape of the fully spread, static droplet provides the characteristic values of the length, contact angle and thickness which are used to make the problem dimensionless. If  $a_s$  and  $\theta_s$  are the final radius of the drop and the equilibrium contact angle and if  $U = \kappa\theta_s$  characterizes the velocity of spread, dimensionless variables can be introduced by the following transformations:

$$h \rightarrow a_s \theta_s h; \quad x, y \rightarrow a_s x, a_s y; \quad t \rightarrow (a_s / \kappa \theta_s) t; \quad \mathbf{q} \rightarrow U \mathbf{q}.$$

With dimensionless parameters defined by

$$\epsilon = 3\mu\kappa/\theta_s^2\sigma, \quad \beta^2 = \alpha/\theta_s^2 a_s^2, \quad (2.15)$$

the governing equation in dimensionless variables is

$$\epsilon \partial h / \partial t + \nabla \cdot [h(h^2 + \beta^2) \nabla(\nabla^2 h)] = 0, \quad (2.16)$$

which must be solved in the area  $\mathcal{A}$  contained by the contact line

$$\Gamma = h(x_e(t), y_e(t), t) = 0.$$

At the edge

$$\mathbf{q}_e = \dot{x}_e(t) \hat{\mathbf{i}} + \dot{y}_e(t) \hat{\mathbf{j}} = -(\nabla h + \hat{\mathbf{n}})|_{h=0} = -(\nabla h) (1 - 1/|\nabla h|)|_{h=0} \quad (2.17)$$

and since

$$\mathbf{Q} = \epsilon^{-1} (h^2 + \beta^2) \nabla(\nabla^2 h) \quad (2.18)$$

it follows that

$$\mathbf{q}_e = \lim_{h \rightarrow 0} \mathbf{Q} = (\beta^2/\epsilon) \nabla(\nabla^2 h)|_{h=0}. \quad (2.19)$$

The initial condition is

$$h(x, y, 0) = h_i(x, y) \quad (2.20)$$

and the constancy of droplet volume is expressed by

$$\iint_{\mathcal{A}} h dS = V_s / \theta_s a_s^3. \quad (2.21)$$

### 3. Procedure

In circumstances for which  $\sigma = 0.1$  dyne/cm,  $\rho = 1$  g/cm<sup>3</sup>,  $\mu/\rho = 0.1$  cm<sup>2</sup>/s,  $\theta_s \simeq 0.2$  rad ( $10^\circ$ ),  $a_s = 10^{-3}$  cm and  $\kappa = 10^{-5}$  cm/s, it follows that

$$\epsilon \simeq 10^{-3}. \quad (3.1)$$

The parameter  $\alpha$  in (2.10) is chosen to make  $\beta^2$  moderately small, say  $\beta^2 < 1$  since this seems consistent with the approximation that fluid slippage on the substratum is to be significant only when the droplet thickness is  $O(100 \text{ \AA})$  or less. For example,  $\alpha < 10^{-10}$  cm<sup>2</sup> implies  $\beta^2 \leq 10^{-2}$ , but the only restriction really required by subsequent analysis is

$$|\epsilon \log \beta| \ll 1. \quad (3.2)$$

This is certainly satisfied as  $\beta$  varies over a very wide range including the extreme values  $\beta^2 = O(\alpha)$  which might be encountered for a larger droplet (with  $\epsilon$  small but  $a_s = 1$  and  $\theta_s = 1$ ).

The basic procedure is to express  $h$ , and all other dependent variables, as power series in  $\epsilon$  (but to regard  $\beta$  as an independent parameter). Given the approximate nature of the model, only the lowest-order results for  $h$  and  $\mathbf{Q}$  are of real interest or relevance. However, it is important to check, in at least one example, that the slope or contact angle at the boundary is a well-defined and useful concept in that it too can be described by a regular perturbation series in  $\epsilon$ .

Let

$$h = h_0 + \epsilon h_1 + \dots; \quad (3.3)$$

the substitution of this series in (2.16) implies that

$$\nabla \cdot [h_0(h_0^2 + \beta^2) \nabla(\nabla^2 h_0)] = 0. \quad (3.4)$$

Likewise, the replacement of  $\mathbf{Q}$  by

$$\mathbf{Q} = \mathbf{Q}_0 + \epsilon \mathbf{Q}_1 + \dots \quad (3.5)$$

in (2.18) shows that

$$\mathbf{Q}_0 + \epsilon \mathbf{Q}_1 + \dots = \epsilon^{-1}(h_0^2 + \beta^2) \nabla(\nabla^2 h_0) + [(h_0^2 + \beta^2) \nabla \nabla^2 h_1 + 2h_1 h_0 \nabla(\nabla^2 h_0)] + \dots \quad (3.6)$$

In order that all physical variables of the problem be regular functions of  $\epsilon$ , the solution of (3.4) must also annihilate the  $O(1/\epsilon)$  term in the last equation for the velocity. It follows that the only acceptable solution satisfies

$$\nabla(\nabla^2 h_0) = 0,$$

or

$$\nabla^2 h_0 = -A(t), \quad (3.7)$$

where  $A(t)$  is an arbitrary function of time. The interpretation is clear: the mean curvature of the droplet surface depends only on time and not on position. In other words, the surface curvature is spatially uniform during the slow motion of the droplet, and a state of quasi-equilibrium is established by the balance of internal and capillary pressures, both of which are functions only of time.

The depth-averaged velocity is determined at the next order of calculation from

$$\mathbf{Q}_0 = (h_0^2 + \beta^2) \nabla(\nabla^2 h_1), \quad (3.8)$$

where

$$\partial h_0 / \partial t + \nabla \cdot [h_0(h_0^2 + \beta^2) \nabla \nabla^2 h_1] = 0. \quad (3.9)$$

The boundary conditions must also be expressed as perturbation series in order to complete the boundary-value problem for  $h_1$ . Although the theory is developed to this extent for a few problems considered in the next section, the main interest at present concerns droplet shape and movement rather than internal fluid velocity and this information is already obtainable from  $h_0(x, y, t)$ . (For certain idealized geometries,  $Q_0$  can be calculated directly from (3.9) but in general  $h_1$  must be determined first and this is a difficult task.) In view of these objectives, it is convenient to eliminate the subscript notation (so that  $h_0$  is written simply as  $h$ ), in which case the basic,  $O(1)$ , boundary-value problem to be solved is

$$\nabla^2 h = -A(t), \quad (3.10)$$

with

$$h(x_e(t), y_e(t), t) = 0 \quad (3.11)$$

on the moving boundary  $\Gamma$ , for which

$$\mathbf{q}_e = \dot{x}_e \mathbf{i} + \dot{y}_e \mathbf{j} = -(\nabla h)(1 - 1/|\nabla h|)|_{h=0}. \quad (3.12)$$

The initial condition is

$$h(x, y, 0) = h_i(x, y) \quad (3.13)$$

and the constancy of drop volume requires

$$\iint_{\mathcal{A}} h \, dx \, dy = V_s/\theta_s a_s^3 \quad (\text{a constant}), \quad (3.14)$$

where  $\mathcal{A}$  is the area enclosed by  $\Gamma$  at time  $t$ . Therefore, a solution of Poisson's equation is sought within a moving boundary whose specification is part of the problem.

#### 4. Spreading and retraction of droplets

##### *The circular drop*

We consider first the radial spread or retraction of a circular droplet. Since this motion is axisymmetric, the independent variables are the radius  $r$  and time  $t$ . The moving contact line is

$$r = a(t), \quad (4.1)$$

for which

$$h(a(t), t) \equiv 0, \quad (4.2)$$

and  $a = 1$  is the final equilibrium radius. The solution of (3.10) which satisfies (4.2) and is regular at the origin is

$$h = \frac{1}{4} A a^2 [1 - (r/a)^2].$$

The arbitrary function  $A$  is determined from (3.14) and the condition that in the final equilibrium state  $|h_r| = 1$  at  $r = a = 1$ . The result is  $A = 2/a^4$ , and the droplet shape at time  $t$  is then

$$h = (2a^2)^{-1} [1 - (r/a)^2]. \quad (4.3)$$

The velocity at the contact line is radial and in this case (3.12) becomes

$$\dot{a}(t) = -(1 + h_r)|_{r=a},$$

or by (4.3)

$$\dot{a}(t) = (1/a^3 - 1). \quad (4.4)$$

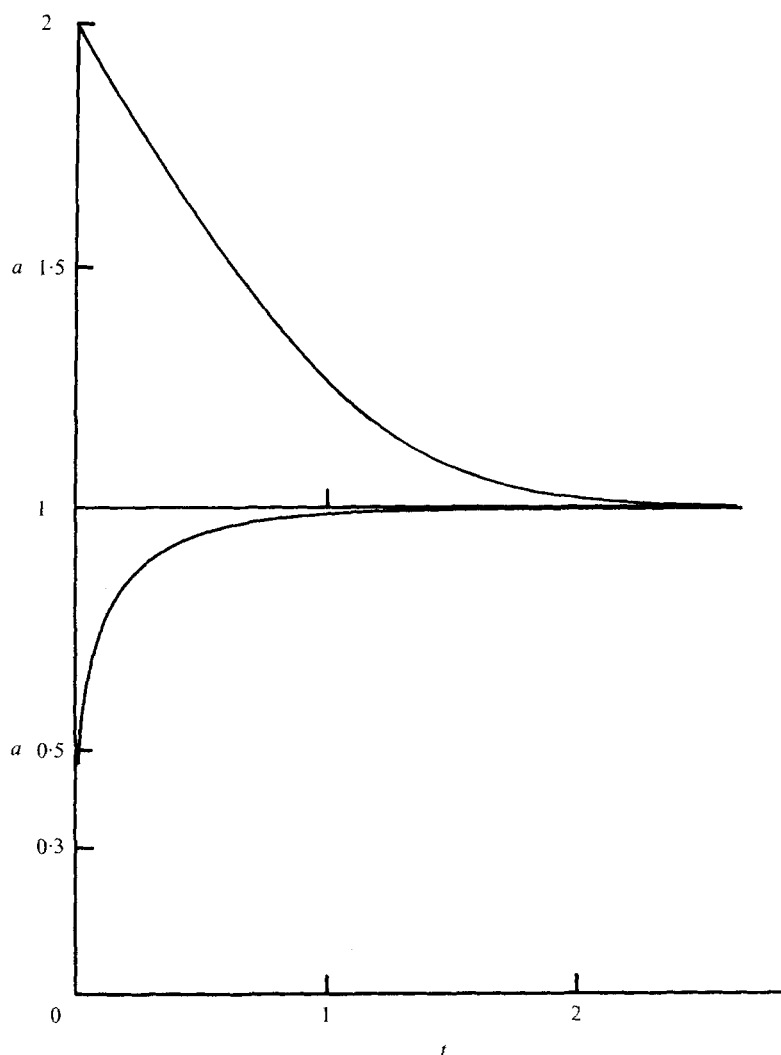


FIGURE 1. Droplet radius *vs.* time for circular spreading and retraction with initial values of 0.3 and 1.5.

The position of the contact line is determined by integrating this differential equation to obtain

$$a + \frac{1}{6} \log \left[ \frac{(a-1)^2}{a^2 + a + 1} \right] - \frac{1}{3^{\frac{1}{2}}} \tan^{-1} \left( \frac{2a+1}{3^{\frac{1}{2}}} \right) = -(t+c). \quad (4.5)$$

The arbitrary constant, which can be evaluated from the initial conditions, affects only the origin of the time axis and an appropriate translation converts a particular solution into a universal function  $a = a(\tau)$ , where  $\tau = t+c$ . Figure 1 shows  $a(t)$  for spreading and retraction for initial values  $a_i = 0.3$  and 2.

The dimensionless scheme adopted is not especially appropriate for studying the spreading of a droplet whose equilibrium contact angle is *zero*. However, this particular limiting case corresponds to the solution of (4.4) when the radius is small, which is

$$a \simeq [4(t+c)]^{\frac{1}{2}}.$$



The spreading rate  $\dot{a}$  is then proportional at large times to  $t^{-\frac{1}{2}}$ . Neogi & Miller (1976), in a similar analysis of spreading on a rough surface that employs porosity instead of the dynamic contact angle, found a spreading rate proportional to  $t^{-\frac{1}{2}}$ , in agreement with experiments on polymer melts and glycerol. The difference in rates does not seem significant.

Since  $\mathbf{Q} = Q\hat{\mathbf{r}}$ , equation (3.9) can be written as

$$\frac{\partial}{\partial t}(rh) + \frac{\partial}{\partial r}(rhQ) = 0.$$

However, it follows from (4.3) that

$$\frac{\partial}{\partial t}(rh) = -\frac{\dot{a}}{a} \frac{\partial}{\partial r}(r^2h),$$

which allows the integration of the velocity equation and leads to the formula

$$Q = (\dot{a}/a)r. \quad (4.6)$$

Of course (4.3), (4.4) and (4.6) are the lowest-order terms  $h_0$ ,  $a_0$  and  $Q_0$  of a consistent perturbation expansion, details of which are given in the appendix in order to resolve certain mathematical issues. The principal conclusion of that analysis is that

$$h = h_0 + O(\epsilon \log \beta)$$

and

$$\partial h / \partial r = \partial h_0 / \partial r + O(\epsilon \log \beta),$$

which implies that the correction to the contact angle is small when  $\epsilon$  is small for all physically realistic values of  $\beta$ . In other words, the contact angle is a well-defined quantity as long as the restrictions imposed are satisfied.

### Frontal spreading

It is instructive to examine the self-spreading of a thin layer  $h = h(x, t)$  with a straight, infinitely long contact line. The motion of this two-dimensional ‘droplet’ can be completely determined to  $O(\epsilon^2)$  in the perturbation formalism.

The solution of the basic boundary-value problem for this geometry is

$$h = (2a)^{-1} [1 - (x/a)^2], \quad (4.7)$$

where  $x = a(t)$ , the position of the contact line, satisfies

$$\dot{a} = 1/a^2 - 1. \quad (4.8)$$

It follows that

$$-a + \frac{1}{2} \log \left| \frac{1+a}{1-a} \right| = t + c. \quad (4.9)$$

This implies a spreading rate for small  $a$  and large  $t$  (values corresponding to the spread of a fluid whose equilibrium contact angle is zero) that is proportional to  $t^{-\frac{1}{2}}$ . Examination of the terms of next order again shows only  $O(\epsilon \log \beta)$  corrections to both  $h$  and  $h_x$ .

### 5. Creeping motion of a droplet on a coated surface

A surface is coated with a material that affects the equilibrium contact angle of a droplet placed upon it. If the contaminant layer is deposited uniformly, the droplet will spread to a static configuration dictated by the new contact angle. However, if the coating is not uniform then the droplet will move in a direction that increases the adherence, or equivalently, that lowers the contact angle. The contact line advances or recedes where the dynamical contact angle is larger or less than the local equilibrium value. Carter's experiment with cells that migrate towards a region of greater concentration of palladium coating a surface of cellulose acetate is the biological analogue of such droplet phenomena.

To study this effect, it is assumed that the equilibrium contact angle is a function of position on the plane, i.e.  $\theta = \theta(x, y)$ . (The equilibrium contact angle at  $(x, y)$  is defined as the contact angle of a droplet on an equivalent surface whose coating has everywhere the properties that the real coating has at  $(x, y)$ .) In the simplest case, we assume a linear variation expressed dimensionally by

$$\theta(x) = \theta_s(1 - \lambda x). \quad (5.1)$$

The equilibrium contact angle then decreases as  $x$  increases but the gradient is assumed to be very gradual,  $\lambda \ll 1$ . (Obviously, the weak restriction,  $x < 1/\lambda$ , also holds.)

The only modification required in the formulation is to retain the equilibrium contact angle as a function of position. The net effect of this in the dimensionless boundary-value problem is to replace (3.12) by

$$\mathbf{q}_e = -(\nabla h)(1 - \theta(x)/|\nabla h|). \quad (5.2)$$

The problem then is to determine the motion and shape of an initially circular droplet placed on a surface for which the local equilibrium contact angle varies linearly with distance. Surprisingly, this complicated problem admits a very simple solution: the droplet remains circular but expands slightly as it moves with a velocity that is essentially proportional to  $\lambda$  (i.e.  $-\theta'(x)$ ).

The hypotheses that the droplet is always circular and moves with variable speed in the  $+x$  direction are proved correct by actually constructing the solution on this basis. To this end, let  $a(t)$  be the radius of the circular droplet and  $V(t)$  its velocity in the  $x$  direction. It is then advantageous to view the movement from the  $(\xi, y)$  co-ordinate system centred in the droplet (see figure 2) with

$$\xi = x - \int_0^t V(t) dt; \quad (5.3)$$

the cylindrical co-ordinates  $(r, \phi)$  appropriate for this geometry are defined by

$$\xi = r \cos \phi, \quad y = r \sin \phi.$$

The transformation to the moving system leaves (3.10) unchanged, so that the solution of this equation for a circular droplet shape is still

$$h = (2a^2)^{-1} [1 - (r/a)^2]. \quad (5.4)$$

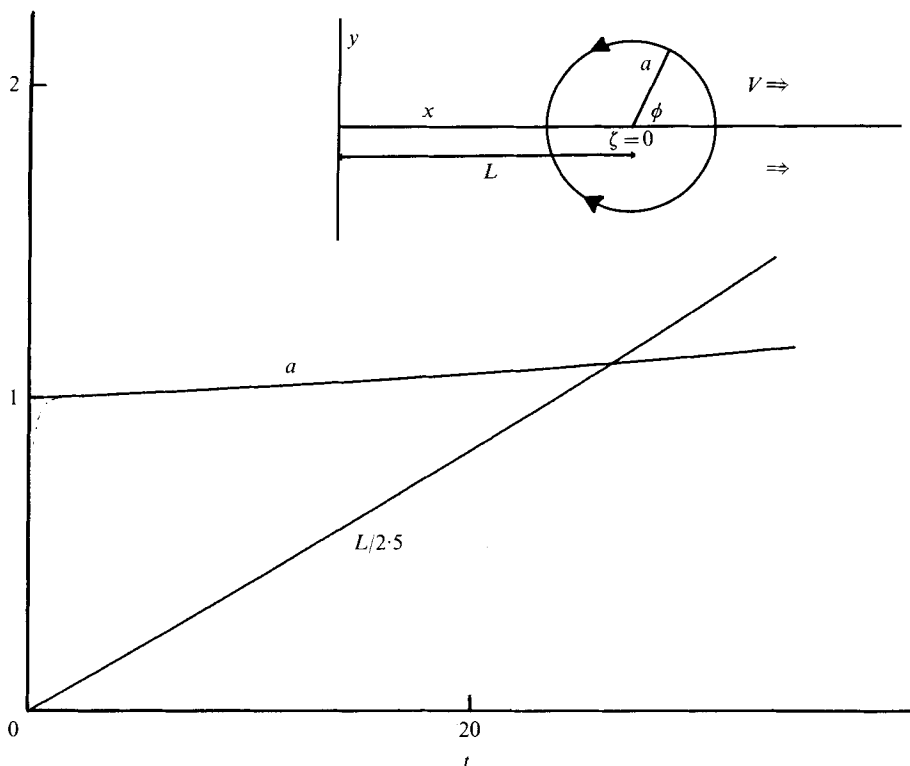


FIGURE 2. Radius  $a(t)$  and distance travelled  $L(t)$  for a droplet moving on a surface with a linear gradient of the equilibrium contact angle;  $\lambda = 0.1$ . The light line for  $a$  near  $t = 0$  corresponds to the movement of a droplet whose initial radius is 0.5. Insert defines variables and shows the motion of particles on the contact line as the droplet creeps forward.

The true velocity of a particle located at polar co-ordinates  $(a(t), \phi(t))$  on the contact line is the sum of the translational velocity of the centre of the droplet and the rotational velocity about the origin:

$$\mathbf{q}_e = V\hat{\mathbf{i}} + \dot{a}\hat{\mathbf{r}} + a\dot{\phi}\hat{\boldsymbol{\phi}}, \quad (5.5)$$

which is equivalent to

$$\mathbf{q}_e = (V \cos \phi + \dot{a})\hat{\mathbf{r}} + (a\dot{\phi} - V \sin \phi)\hat{\boldsymbol{\phi}}. \quad (5.6)$$

This velocity is related to the contact angle at  $(a, \phi)$  by (5.2) and since  $-\nabla h = \hat{\mathbf{r}}$  it follows that

$$\dot{a}(t) + V \cos \phi(t) = 1/a^3 - \theta(x)|_{r=a}, \quad (5.7)$$

and

$$a\dot{\phi}(t) - V \sin \phi(t) = 0. \quad (5.8)$$

However,

$$\theta = 1 - \lambda x = 1 - \lambda \left( r \cos \phi + \int_0^t V dt \right), \quad (5.9)$$

and the result of substituting this formula in (5.7) is

$$\dot{a}(t) + V \cos \phi(t) = \frac{1}{a^3} - 1 + \lambda \int_0^t V dt + \lambda a \cos \phi(t). \quad (5.10)$$

According to (5.8), particles at  $\phi = 0, \pi$  do not change their positions on the circumference as the droplet moves. Therefore, by evaluating this equation at these two locations, both  $a(t)$  and  $V(t)$  may be determined. That is, at  $\phi = 0$

$$\dot{a} + V = \frac{1}{a^3} - 1 + \lambda \int_0^t V dt + \lambda a$$

and at  $\phi = \pi$

$$\dot{a} - V = \frac{1}{a^3} - 1 + \lambda \int_0^t V dt - \lambda a,$$

from which it follows by addition and subtraction that

$$\dot{a} = \frac{1}{a^3} - 1 + \lambda \int_0^t V dt, \quad (5.11)$$

$$V = \lambda a. \quad (5.12)$$

A single equation for the radius  $a$  is

$$\ddot{a} + (3/a^4) \dot{a} - \lambda^2 a = 0; \quad (5.13)$$

the initial conditions at  $t = 0$  are

$$a = a_i, \quad \dot{a} = 1/a_i^3 - 1. \quad (5.14)$$

The distance moved by the droplet is given by the position of its centre:

$$L(t) = \int_0^t V(t) dt = \lambda \int_0^t a(t) dt. \quad (5.15)$$

Equations (5.4), (5.11), (5.12) and (5.15) completely describe the shape, position and velocity of the droplet at any time. In addition, integration of (5.8) completes the determination of the Lagrangian co-ordinates  $(a(t), \phi(t))$  of a particle on the contact line.

Substitution for  $V$  in (5.8) gives

$$\dot{\phi} = \lambda \sin \phi,$$

the integration of which yields

$$\phi(t) = 2 \tan^{-1} (e^{-\lambda t} \tan \tfrac{1}{2} \phi_i), \quad (5.16)$$

where  $\phi_i$  is the initial polar angle of the point being tracked. As time increases so does  $|\phi(t)|$  (unless  $\phi_i = 0$ ), and this means that a particle on the contact line moves around the circumference towards the rear of the droplet as the droplet advances, see figure 3.

For  $\lambda$  small, the motion of a droplet whose initial radius is  $a_i$  occurs in two stages. The initial phase, lasting only a few time units, is one in which the droplet quickly adjusts to its immediate surroundings by spreading or retracting to unit size. This is followed by a much longer period (whose time scale is  $O(1/\lambda)$ ) during which the droplet slowly moves in the direction of greater adherence to the substratum, i.e. along the  $+x$  axis.

Since the initial stage has already been examined in §4, we shall concentrate here on the motion over the long time scale  $\lambda t$ , in which case the initial radius of the droplet appears essentially to be  $a_i = 1$ . An accurate approximate solution of (5.13) and (5.14) is then readily achieved by the substitution

$$a(t) = 1 + \lambda^2 \zeta(t) \quad (5.17)$$

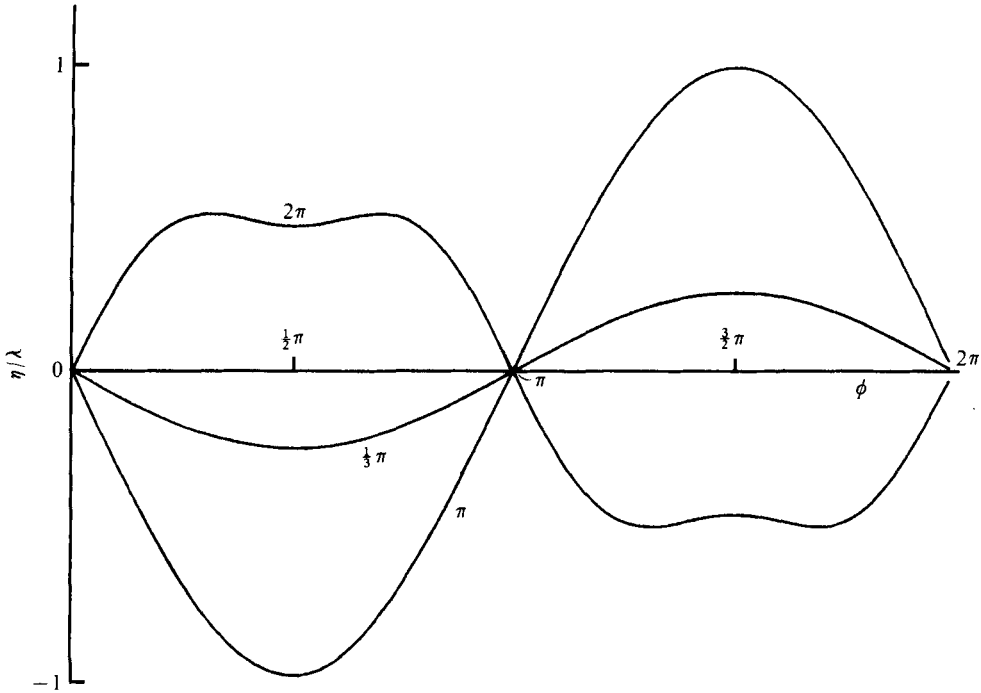


FIGURE 3. Contact-line distortion *vs.* polar angle for a droplet on a contaminated surface for  $K = \frac{1}{3}\pi$ ,  $\pi$  and  $2\pi$ .

so that

$$\ddot{\zeta} + 3(1 + \lambda^2 \zeta)^{-4} \dot{\zeta} - (1 + \lambda^2 \zeta) = 0 \quad (5.18)$$

with

$$\zeta = 0 = \dot{\zeta} \quad \text{at} \quad t = 0.$$

It is sufficient to set  $\lambda = 0$  in this equation (instead of developing  $\zeta$  as a perturbation expansion) and to this degree of approximation we find that

$$\zeta = \frac{1}{3}t - \frac{1}{9}(1 - e^{-3t}),$$

or

$$a(t) = 1 + \frac{1}{3}\lambda^2[t - \frac{1}{3}(1 - e^{-3t})]. \quad (5.19)$$

It follows that

$$L(t) = \lambda[t + \frac{1}{6}\lambda^2(t^2 - \frac{2}{3}t + \frac{2}{9}(1 - e^{-3t}))]. \quad (5.20)$$

In terms of the long time scale variable  $\tau = \lambda t$ , these formulae can be expressed approximately as

$$\left. \begin{aligned} a &\simeq 1 + \frac{1}{3}\lambda\tau + \dots, \\ L &\simeq \tau + \frac{1}{6}\lambda\tau^2 + \dots \end{aligned} \right\} \quad (5.21)$$

The radius evidently changes very little as the droplet moves a substantial distance; for  $\lambda = 0.01$  the radius of the droplet expands by 3% in moving a distance of 5 diameters ( $\tau = 10$ ). The numerical evaluations of (5.13) for  $a(t)$  and  $L(t)$  are shown in figure 3 and confirm the accuracy of these conclusions.

If the gradient  $\theta'(x)$  of the equilibrium contact angle on the coated surface is small, so that there is very little variation about the perimeter of the droplet at any time,

then the velocity of the droplet (or cell) will be opposite but proportional to the gradient. To see this, the contact angle is written as a Taylor series about the centre of the droplet:

$$\theta(x) = \theta(L + a \cos \phi) \cong \theta(L) + \theta'(L) a \cos \phi. \quad (5.22)$$

Two terms of the series suffice because  $\theta'(L) = -\lambda$  is assumed small. Since this expression is entirely analogous to (5.9) in its  $\phi$  dependence, the analysis proceeds as before to the stated conclusions:

$$\dot{a} = 1/a^3 - \theta(L) \quad (5.23)$$

and

$$V = -\theta'(L) a = \lambda a. \quad (5.24)$$

Many variants of this problem are possible, including the response of a droplet to a contamination field that produces a wavelike variation of the contact angle

$$\theta = \theta(x - Ut),$$

but these will not be presented here. We also omit discussion of the corresponding one-dimensional problems, which, though easily solved, provide little additional information.

## 6. Droplet distortion due to surface contamination

Surface coating, contamination or roughness distorts the shape of a droplet by modifying the local equilibrium contact angle. The droplet is drawn towards regions of greater adherence (i.e. smaller contact angle) and likewise retracts from regions of weaker attachment. It is of interest to examine distortion of the droplet as a function of the structure and scale of the contamination field. The spread of cells and of comparably sized droplets of Dow Corning 550 fluid do show asymmetries which may be attributable to the variation of surface properties.

The minor anomalies of the solid surface, whatever their source, will be described by an equilibrium contact angle that is a function of position given in dimensionless notation by

$$\theta(x, y) = 1 + \lambda \Theta(x, y), \quad (6.1)$$

where  $\lambda$  is a small parameter. The effect of this slight variation on the spread and shape of an initially circular droplet can be determined by a conventional perturbation analysis of the fundamental boundary-value problem, (3.10) *et seq.*, in which (3.12) is replaced by

$$\mathbf{q}_e = -(\nabla h) (1 - \theta(x, y)/|\nabla h|)|_{h=0}. \quad (6.2)$$

The general solution of

$$\nabla^2 h = -A$$

in cylindrical co-ordinates  $(r, \phi)$  is

$$h = -\frac{1}{4}A(t)r^2 + B(t) + \lambda \sum_{n=1}^{\infty} (b_n(t) \cos n\phi + c_n(t) \sin n\phi) r^n; \quad (6.3)$$

the coefficient functions must satisfy the requisite boundary conditions (6.2), which in component form are

$$\left. \begin{aligned} \dot{r} &= -h_r(1 - \theta/|\nabla h|)|_{h=0}, \\ r\dot{\phi} &= -r^{-1}h_\phi(1 - \theta/|\nabla h|)|_{h=0}. \end{aligned} \right\} \quad (6.4)$$

The conservation of droplet volume is expressed by

$$\iint_{\mathcal{A}} r h dr d\phi = \frac{\pi}{4}, \quad (6.5)$$

where  $\mathcal{A}$  is the surface area contained by the curve  $h = 0$ . It is the extreme complexity of the Lagrangian boundary conditions that necessitates the use of perturbation theory. However, the procedure is carried out for only one step, to  $O(\lambda)$  terms, and the formalism is simplified accordingly. Therefore, let the contact line be described by

$$r = a(t) + \lambda \eta(\phi, t) \quad (6.6)$$

so that

$$h(a + \lambda \eta, t) = 0 \quad (6.7)$$

and write

$$A = A_0 + \lambda A_1, \quad B = B_0 + \lambda B_1. \quad (6.8)$$

The coefficients  $b_n$  and  $c_n$  need not be expanded to this order of approximation, but the Lagrangian co-ordinates of a point on the contact line, which are involved in (6.4), must be expressed in powers of  $\lambda$ . The relationship between the radial and azimuthal velocity components of a particle on the edge is provided by differentiating (6.6) to obtain

$$\dot{r}(t) = \dot{a}(t) + \lambda(\eta_t + \eta_\phi \dot{\phi}). \quad (6.9)$$

This shows that only the first term of the expansion

$$\phi = \phi_0 + O(\lambda)$$

appears in the calculation for either  $\dot{r}$  or  $\dot{a}$  to the degree of accuracy required and the subscript zero is dropped.

The substitution of these perturbation series in (6.4), (6.5), (6.7) and (6.9) and the expenditure of the usual amount of effort required by this method (the details of which are omitted in the interest of brevity) lead to the following results. From (6.5) and (6.7), we obtain

$$A_0 = 2/a^4, \quad B_0 = 1/2a^2, \quad B_1 = \frac{1}{8}a^2A_1, \quad (6.10)$$

while (6.4) and (6.9) yield (4.4) once again and

$$\dot{\phi} = 0, \quad (6.11)$$

$$\eta(\phi, t) = a^3 \left[ -\frac{1}{8}A_1 a^2 + \sum_{n=1}^{\infty} (b_n \cos n\phi + c_n \sin n\phi) a^n \right], \quad (6.12)$$

$$\eta_t = -\Theta(a \cos \phi, a \sin \phi) + \frac{\eta}{a^4} + \frac{1}{2}aA_1 - \sum_{n=1}^{\infty} n(b_n \cos n\phi + c_n \sin n\phi) a^{n-1}. \quad (6.13)$$

Thus (6.11) implies that the azimuthal co-ordinate  $\phi$  of a point on the contact line is a constant to  $O(\lambda)$  i.e. the particle path is a ray from the centre. The last two equations can then be treated as ordinary Fourier series expansions. The elimination of  $\eta$  between (6.12) and (6.13) allows equations for  $A_1$ ,  $b_n$  and  $c_n$  to be determined from the corresponding Fourier coefficients of  $\Theta(a \cos \phi, a \sin \phi)$  and the initial conditions. These ordinary differential equations are

$$\frac{d}{dt}(a^5 A_1) + 3aA_1 = \frac{4}{\pi} \int_0^{2\pi} \Theta(a \cos \phi, a \sin \phi) d\phi, \quad (6.14)$$

$$\frac{d}{dt}[a^{n+3}(b_n + ic_n)] + (n-1)a^{n-1}(b_n + ic_n) = -\frac{1}{\pi} \int_0^{2\pi} \Theta e^{in\phi} d\phi; \quad (6.15)$$

the initial conditions are  $A_1 = 0 = b_n = c_n$  and  $a = a_i$ . Once the coefficient functions are known, the contact line is determined by (6.6) and (6.12) and the droplet shape is obtained from (6.3) or

$$h = \frac{1}{2a^2} \left( 1 - \left( \frac{r}{a} \right)^2 \right) + \lambda \left[ \frac{1}{8} A_1 \left( 1 - 2 \left( \frac{r}{a} \right)^2 \right) + \sum_{n=1}^{\infty} r^n (b_n \cos n\phi + c_n \sin n\phi) \right]. \quad (6.16)$$

As a specific example, we calculate the final shape of a unit droplet ( $a = 1$ ) on a surface for which

$$\Theta(x, y) = \sin kx \sin ky. \quad (6.17)$$

This function satisfies the necessary condition for the droplet to remain motionless,

$$\int_0^{2\pi} \Theta(\cos \phi, \sin \phi) e^{i\phi} d\phi = 0,$$

and describes a periodic distortion of surface contamination whose length scale is  $\pi/k$ . The distortion of the droplet depends on the magnitude of this length compared with unity, the radius of the droplet, but for the theory to be valid  $\lambda k/\pi$  must also be small.

It follows from the substitution of (6.17) in (6.14) and (6.15) that

$$A_1 \equiv 0, \quad b_n \equiv 0, \quad c_{2n+1} \equiv 0$$

and

$$c_{2n} = -\frac{2}{2n-1} \sin \frac{n\pi}{2} J_{2n}(2^{\frac{1}{2}}k), \quad (6.18)$$

where  $J$  is the Bessel function of the first kind. Therefore

$$\eta(\phi, t) = 2 \sum_{n=0}^{\infty} \frac{(-1)^{n+1}}{4n+1} J_{4n+2}(K) \sin(4n+2)\phi, \quad (6.19)$$

where for convenience we define  $K = 2^{\frac{1}{2}}k$ . This is a rapidly converging series and unless  $K$  is large the deviation of the contact line is almost entirely given by the first term

$$-2J_2(K) \sin 2\phi.$$

Figure 3 illustrates the distortion of the contact line in three cases:  $K = \frac{1}{3}\pi$ ,  $\pi$  and  $2\pi$ . It is evident that the higher harmonics are not significant for  $K < \pi$ ; moreover, the effect of the contamination on the droplet is very nearly maximal at  $K = \pi$ .

The spatial distribution given by (6.17) is one for which the perturbation of the contact angle is alternately positive and negative on a chequered grid of identical squares of diagonal  $2\pi/K$ . For the values chosen the respective diagonals are 6, 2 and 1 and the grids for the last two cases are shown superposed on the unit droplet in figure 4. When  $K = \pi$  and a maximal response is induced, each square is approximately the size of the droplet. The droplet distorts by moving into regions where the contact angle is decreased (marked with a minus) and retracting from regions of higher contact angle (marked plus). The situation is similar but much less pronounced for the largest grid; the smallest grid ( $K = 2\pi$ ) offers many possibilities for local spreading and retraction and hence for the generation of higher harmonics in the Fourier series. We conclude that surface variations on the length scale of the droplet are most important in producing distortions.

The response of a round, spreading or retracting droplet on an uncontaminated surface to a perturbation of shape can be examined by setting  $\Theta = 0$  in (6.14) and (6.15). For  $a = 1$ , the coefficients  $A_1$ ,  $b_n$  and  $c_n$  ( $n \neq 1$ ) tend to zero exponentially as



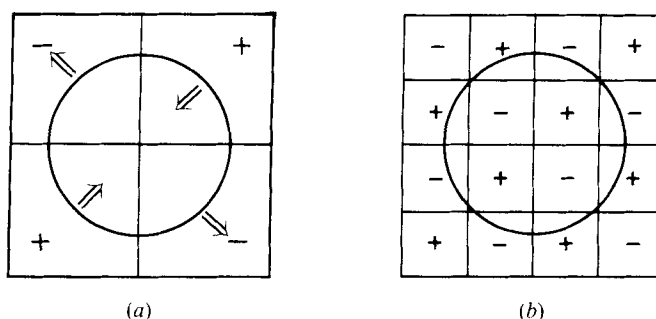


FIGURE 4. Droplet on a surface contaminated in a chequered pattern corresponding to the values (a)  $K = \pi$  and (b)  $K = 2\pi$  of figure 3. The equilibrium contact angle decreases or increases in regions marked minus or plus. The droplet moves in the directions of the arrows.

$t$  increases. All initial perturbations must decay at the same rate, which shows that the circular geometry is a stable configuration. Perturbations can also be shown to decay in the cases of a spreading or retracting droplet whose radius is a function of time  $a(t)$  given by (4.5).

## 7. Summary

Despite rather obvious limitations, lubrication theory is an effective means of studying the motion of thin viscous layers on a surface. (Lacking adequate numerical models, it is perhaps the only feasible method at present.) The analysis here was directed specifically at the movement of small fluid droplets and analogous biological problems of cell motion and we have described in some detail contact-line behaviour and the conditions of droplet motion and distortion.

The salient features and principal conclusions can be summarized as follows: (i) the model is based on the lubrication equations and the dynamic contact angle expresses the forces exerted on the fluid at the moving contact line; (ii) the dynamic contact angle is shown to be a well-founded and meaningful concept within the imposed restrictions; (iii) the droplet maintains a spatially uniform, but time-dependent mean curvature in moving on the solid surface; (iv) the spreading and retraction of droplets are stable processes in that a circular contact line always evolves in time; (v) the velocity of a small droplet on a contaminated surface is opposite but proportional to the gradient of the equilibrium contact angle; (vi) a creeping droplet expands only slightly in moving several diameters from its original position; (vii) surface contamination on the scale of the droplet diameter is most effective in producing distortions of the droplet surface and its contact line. Agreement with certain experiments is good, and the theory suggests others for further comparison, but it is not surprising that many (if not most) adherence phenomena lie outside the scope of this work. However, some of these may be amenable to analysis along similar lines by including new physical processes. Among the more important modifications is the introduction of a surface-active chemical of either external or internal origin (in the cell) which would be incorporated by an appropriate condition on the free surface. This and other variations relevant to biological problems are being studied and will be reported in due course.

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## Appendix

In § 4, the solution for the spreading droplet was expressed as perturbation series

$$h = h_0 + \epsilon h_1 + \dots,$$

$$a = a_0 + \epsilon a_1 + \dots;$$

the functions  $h_0$  and  $a_0$  are given as  $h$  and  $a$  in (4.3) and (4.5). The complete boundary-value problem for  $h_1$  is

$$\frac{\partial h_0}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left( h_0 (h_0^2 + \beta^2) r \frac{\partial}{\partial r} \frac{1}{r} \frac{\partial}{\partial r} r \frac{\partial h_1}{\partial r} \right) = 0 \quad (\text{A } 1)$$

with

$$\partial h_1 / \partial r = 0 = \partial^3 h_1 / \partial r^3 \quad \text{on} \quad r = 0$$

and

$$\left. \begin{aligned} h_1 + a_1 \partial h_0 / \partial r &= 0 \\ \dot{a}_1 &= - \left( \frac{\partial h_1}{\partial r} + a_1 \frac{\partial^2 h_0}{\partial r^2} \right) \Big|_{r=a_0} \end{aligned} \right\} \quad \text{at} \quad r = a_0. \quad (\text{A } 2)$$

Constant droplet volume requires that

$$\int_0^{a_0} h_1 r dr = 0.$$

If  $\xi = r/a_0$ , then substitution for  $h_0$  in (A 1) and integration of that equation yields

$$\begin{aligned} \xi \frac{\partial h_1}{\partial \xi} &= \frac{a_0^5 \dot{a}_0}{2\beta} \left[ (1 - \xi^2) \tan^{-1} \left( \frac{1 - \xi^2}{\lambda} \right) - \tan^{-1} \left( \frac{1}{\lambda} \right) \right. \\ &\quad \left. - \frac{1}{2} \lambda \log \left( \frac{\lambda^2 + (1 - \xi^2)^2}{\lambda^2 + 1} \right) + \xi^2 \tan^{-1} \frac{1}{\lambda} \right] + \frac{c}{2} \xi^2, \end{aligned} \quad (\text{A } 3)$$

where

$$\lambda = 2a_0^2 \beta, \quad c = 8a_1/a_0^3 - 2a_0^7 \dot{a}_0 [1 - \lambda \tan^{-1} (1/\lambda)].$$

Therefore  $\partial h_1 / \partial \xi$  is a regular function in  $0 \leq \xi \leq 1$ , and at the edge  $\xi = 1$

$$\partial h_1 / \partial \xi = -\omega a_0^7 \dot{a}_0 + 4a_1/a_0^3, \quad (\text{A } 4)$$

where

$$\omega = \log [\lambda(1 + \lambda^2)^{-1/2}] + 1 - \lambda \tan^{-1} (1/\lambda).$$

Moreover it follows from (A 2) that

$$\dot{a}_1 + 3a_1/a_0^4 = \omega a_0^8 \dot{a}_0. \quad (\text{A } 5)$$

Analysis of these equations shows that, for all  $\xi$  in the unit interval

$$\partial h_1 / \partial \xi = O(\log \lambda)$$

when  $\lambda$  is small. The final integration of (A 3),

$$h_1 = \int_1^\xi \frac{\partial h_1}{\partial \xi} d\xi + \frac{a_1}{a_0^3},$$

is not reducible to simple functions but examination of the integrals involved leads to the conclusion that

$$h_1 = O(\log \lambda).$$

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