Spreading of a micrometric fluid strip down a plane under controlled initial conditions

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We experimentally study the spreading of a small volume of silicon oil down a vertical plane with small Bond number. The initial condition is characterized by a horizontal long fluid strip with cross sectional area A and width w_0. We find that the experiments are characterized by a unique nondimensional parameter, $R \propto w_0^4/(a^2 A)$, where a is the capillary length. An empirical criterium to estimate the onset of the contact line instability is established. The later rivulet formation at the contact line leads to a pattern which is characterized by a dominant wavelength. We find that this wavelength is approximately proportional to $R^{-1/4}$.

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The coating of a solid surface by a thin liquid film is a process of basic research and technical interest. When a volume of fluid is released on an inclined plane, the gravity drives the spreading and the liquid uniformly coats the surface. Nevertheless, the process is affected by the growth of corrugations at the contact line. These corrugations form a set of channels or "fingers" in the downslope direction that guides the fluid, leaving dry patches at both sides until the fluid eventually covers the whole surface.

After the pioneer work of Huppert [1], other authors have studied the instability of the contact line under complete wetting conditions [2–6]. All fluids employed in those experiments (typically, silicon oils and glycerin) are very viscous, so that the Reynolds number is very small. Their liquid volumes V yield large values of the Bond number $B=(h_c/a)^2$, where h_c is the characteristic height of the fluid and $a=\sqrt{\gamma/\rho g}$ is the capillary length (γ is the surface tension, ρ the density, and g the gravity). If the initial condition is uniform along the transverse direction of the incline, then the flows can be characterized by the fluid cross section $A=V/L$, where L is the transverse extension.

In the experiments reported in the literature, A ranges from 1 to 10 cm^2. In this paper we shall present results from experiments with A between 10^{-4} and 10^{-2} cm^2, and whose characteristic height is of some tens of microns, thus ensuring small Bond numbers. As a consequence, the flows studied here are closer to the coating processes used in applications, and the outcome of the investigation is of particular interest to this field. Furthermore, previous works consider relatively small inclination angles α, spanning the values $1° < \alpha < 55°$. Here we concentrate on flows on a vertical plane ($\alpha=90°$).

Recently [7] we have studied these flows by focusing on (a) the comparison between experimental profiles and 1D numerical simulations and (b) the prediction of the modal growth rates by using a mixed analytical-numerical linear model. Instead, the main goal of the present paper is to explore in detail the influence of the initial configuration and discuss its importance on the stable spreading as well as on the emerging pattern formation. This is done by performing a parametric study on a much more complete series of experiments which covers a larger range of area A and width w_0.

Under the lubrication approximation, the height $h(x,y,t)$ obeys the equation

$$3\mu\partial_t h + \gamma\nabla(h^3\nabla\nabla^2 h) + \rho g(\sin\alpha)\partial_x h^3 - \rho g(\cos\alpha)\nabla(h^3\nabla h) = 0, \qquad (1)$$

where μ is the viscosity, x-axis is in the downslope direction and y is the cross-slope direction (horizontal). Huppert [1] considers the y-independent flow, and neglecting capillary effects and the normal gravity force, he obtains a self similar solution for the position of the front contact line (measured from the rear contact line) as

$$w_H = \left(\frac{9A^2 g\sin\alpha}{4\nu}\right)^{1/3} t^{1/3}, \qquad (2)$$

where $\nu=\mu/\rho$ is the kinematic viscosity. We will see below that this solution is appropriate only for cases with large Bond numbers.

Our experiment is carried out by placing a thin (few tenths of mm) silicon oil filament [PDMS, $\nu=20$ St (Stokes) and $\rho=0.96$ g/cm^3] on a glass plate. The goal is to achieve a uniform initial condition with straight and parallel contact lines. The dimensions of the glass plate is 20 cm in the downslope direction and 8.7 cm in the transverse direction. The filament is generated from the draining of PDMS out of a nozzle at the base of a container. Both the height of the fluid in the container and the diameter of the nozzle determine the filament cross section. A mechanical device allows the filament to be captured by the substrate, which is then conveniently rotated in such a way that the filament is left on horizontal position [7].

The flow evolution is monitored by means of two optical techniques. The first one is based on the use of an anamorphic lens [8] that shows on a screen a light curve displaying the first derivative, $\partial h/\partial x$, of the height profile, $h(x,t)$, which is then obtained by integration. The second one is a schlieren technique that gives a two-dimensional view of the spreading, such as the shape of both the rear and leading contact lines. More details can be found in Ref. [7].

Our experiments show that the initial shape of the cross section of the filament is very like a cylindrical cap (see also

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TABLE I. List of experiments for several A's and w_0's. The smallness of the initial aspect ratio $r=h_0/w_0$ ensures the validity of the lubrication approximation from the very beginning of the experiment. The dimensionless parameter R is defined in Eq. (8).

Expt.	A (10^{-4} cm^2)	w_0 (10^{-2} cm)	r (units of 10^{-2})	R
E_1	4.7	8.83	9.00	4.1
E_2	3.6	8.58	7.30	4.8
E_3	10.0	12.98	8.90	9.0
E_4	6.7	12.86	6.08	12.9
E_5	96.0	31.45	14.56	32.3
E_6	7.2	24.07	1.86	148.0

Ref. [7]). Thus, the apex thickness h_0 is given by

$$h_0 = \frac{A}{I w_0}, \tag{3}$$

where w_0 is the initial width of the filament and I is the *shape factor* (in this case, $I=2/3$). Since A is determined by the conditions at the outlet of the container, we use the following procedure to obtain different w_0's for a fixed A: the substrate with the fluid strip is left in horizontal position for a few minutes before it is set vertically. Measurements of the thickness profile with the above mentioned anamorphic lens show that the cross section remains as a cylindrical cap. The parameters of the initial conditions of six experiments are detailed in Table I. Each experiment is characterized by A and w_0.

We define the width of the fluid strip as $w=x_f-x_r$, where x_f and x_r are the positions of the frontal and rear contact lines, respectively (except for the very early stages, x_r does not vary appreciably during the spreading). The time evolution of $w(t)$, registered from early times till the onset of the contact line instability, is shown in Fig. 1 by employing the same scaling as used in Fig. 3 of Ref. [1]. In that work, Huppert shows that Eq. (2) (solid line in Fig. 1) is in very good agreement with his experiments. However, this law overestimates the asymptotic front position in our experiments and does not give the correct time dependence. Only at the very end of the stable spreading, the front seems to approach the $t^{1/3}$ law as shown by the dashed lines in Fig. 1. The agreement of Eq. (2) with Huppert's large area experiments is due to the fact that capillary effects are negligible. Here A is 10^{-3} to 10^{-4} times smaller, so that these effects are important ($B \ll 1$) and must be taken into account. It is well known that the contact line instability is related with the bump induced by surface tension at the front [2,9]. For large values of B, such as in Huppert's experiments, the front presents a rolling motion [10,11], and consequently this implies a different front dynamic. A feature of the experiments reported here is that the onset of the instability occurs before the spreading can reach an asymptotic self-similar regime (an analogous behavior was reported in spin coating experiments [12]). Thus the description of the previous stable stage corresponds to an initial value problem.

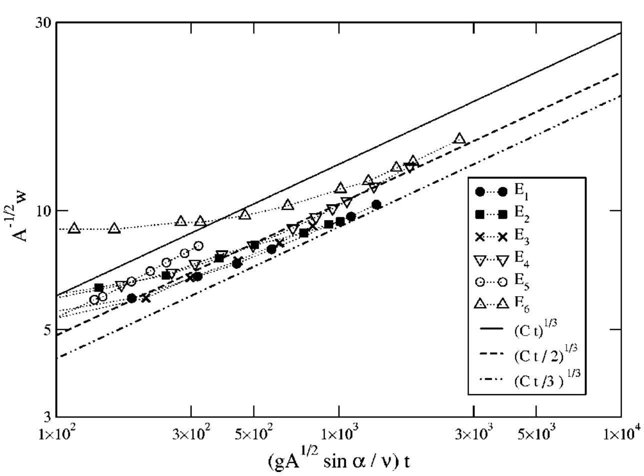

FIG. 1. Time evolution of the width of the fluid strip, w, scaled as suggested by Huppert [1]. The solid line is given by Eq. (2), and the dashed and dotted-dashed lines correspond to the same equation with different prefactors, with $C=9gA^{1/2}/(4\nu)$. The time range is the same as used in Fig. 3 of Ref. [1].

Let us consider now Eq. (1) in dimensionless form. If we take $x_c(=y_c)$, h_c and t_c as scales of the problem, Eq. (1) can be written as

$$\frac{3\mu}{\gamma}\frac{x_c^4}{h_c^3 t_c}\partial_{\tilde t}\tilde h + \tilde\nabla(\tilde h^3\tilde\nabla\tilde\nabla^2\tilde h) + \frac{\rho g \sin\alpha}{\gamma}\frac{x_c^3}{h_c}\partial_{\tilde x}\tilde h^3 - \frac{\rho g \cos\alpha x_c^2}{\gamma}\tilde\nabla(\tilde h^3\tilde\nabla\tilde h) = 0, \tag{4}$$

where the tilde indicates dimensionless quantities or operators. The normal gravity term ($\propto\cos\alpha$) can be neglected in comparison with the parallel gravity term ($\propto\sin\alpha$) provided the inclination angle satisfies

$$\tan\alpha \gg r \equiv h_c/x_c, \tag{5}$$

where r is the characteristic aspect ratio. Under this assumption and defining

$$x_c = w_0, \quad h_c = h_0, \quad t_c = \frac{3\mu}{\gamma}\frac{w_0^4}{h_0^3}, \tag{6}$$

Eq. (4) becomes

$$\partial_{\tilde t}\tilde h + \tilde\nabla(\tilde h^3\tilde\nabla\tilde\nabla^2\tilde h) + R\partial_{\tilde x}\tilde h^3 = 0, \tag{7}$$

where

$$R = (I w_0^4 \sin\alpha)/(a^2 A). \tag{8}$$

Thus the flow evolution is characterized by a single dimensionless parameter R, which gives the ratio between gravity and capillary forces. Note that Eq. (6) implies that the dimensionless initial values of width and height are always equal to unity.

The effects of the parameters A and w_0 on the flow evolution in the stable stage are shown in Fig. 2. It is usually

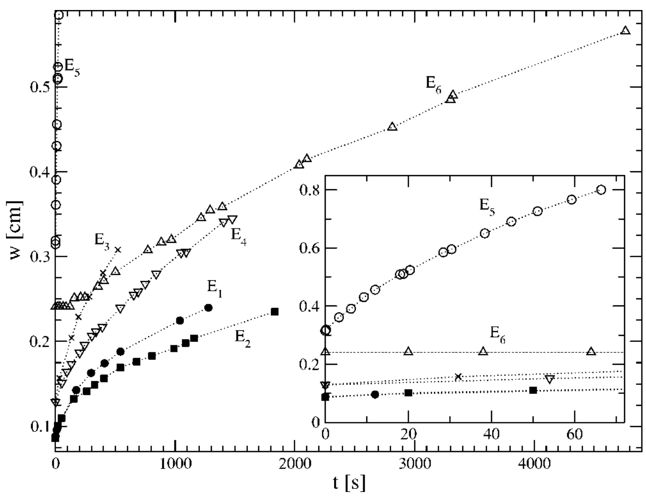

FIG. 2. Time evolution of the width of the spreading, w. The inset is a magnification for very early times, which allows us to see case E_5.

accepted that larger A's imply faster spreadings. This is precisely what happens in experiments E_1 and E_2 with areas $A_1 = 4.7\times 10^{-4}$ cm^2 and 3.6×10^{-4} cm^2, respectively, which have practically the same initial width w_0. Nevertheless, the experiment E_4, which has an area smaller than E_6, evolves faster than E_6. These results suggest that both the cross section A and the initial width w_0 determine the dynamic of the evolution. In fact, for the same area, greater values of w_0 imply lower dynamic contact angles, and according to Tanner's law, smaller advancing velocities.

In Fig. 3 we show the evolution of the spreadings by using the scaling defined in Eq. (6). Notice that R constitutes an ordering parameter since, in dimensionless units, the time evolution is faster for larger R's. This can be understood considering the increase of R as an effective increase of the downslope component of gravity.

Up to our knowledge, there is in the literature no analytical solution of this problem including capillary effects. An elaboration of our experimental data shows that the position

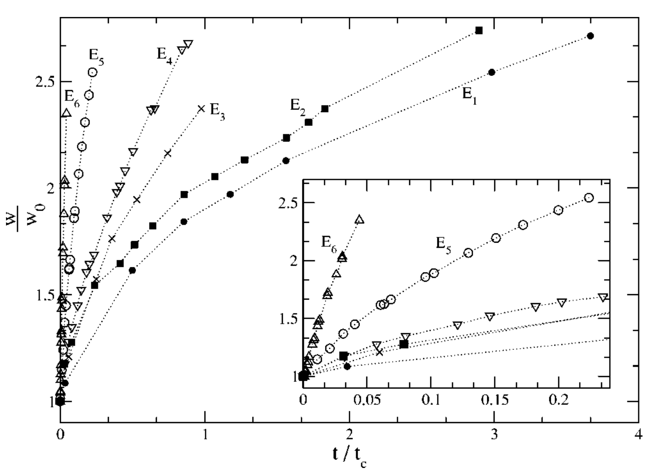

FIG. 3. Evolution of the width of the spreading w, using the scaling defined in Eq. (6). The spreading rate increases for increasing R (see Table I).

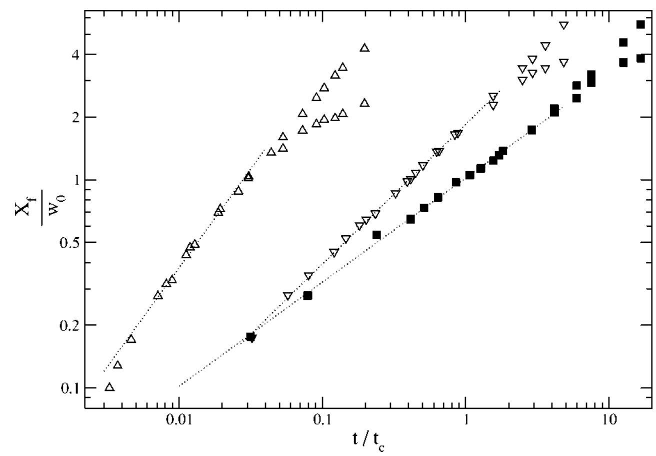

FIG. 4. Frontline evolution for three experiments: E_2 (squares), E_4 (triangle down), and E_6 (triangle up). The dotted lines are given by Eq. (9). The beginning of the instability is shown by the splitting of the data, which then correspond to the positions of a fingertip and a trough.

of the front (contact line) measured from its initial position can be described by

$$x_f/w_0 = \kappa \tilde{t}^{\theta}, \tag{9}$$

where the prefactor κ and the exponent θ are only functions of R. In Fig. 4 we present power law fittings for E_2, E_4, and E_6. The analysis of all the experiments shows that when R varies from 4 to 148, κ increase from 0.8 to 30, whereas the exponent θ growths slowly from 0.47 to 0.95. Finally, the stable stage ends when the dimensionless width w/w_0 is about 2.7±.6. This result shows another difference with Huppert's experiments, where the critical width $w_H^c \propto A^{1/2}$.

When the unstable stage is reached, the contact line develops a pattern that can be described by its spatial discrete Fourier transform [7]. The dependence of the dominant wavelength of the spectrum, λ, with the parameter R is shown in Fig. 5, where the straight line represents the power

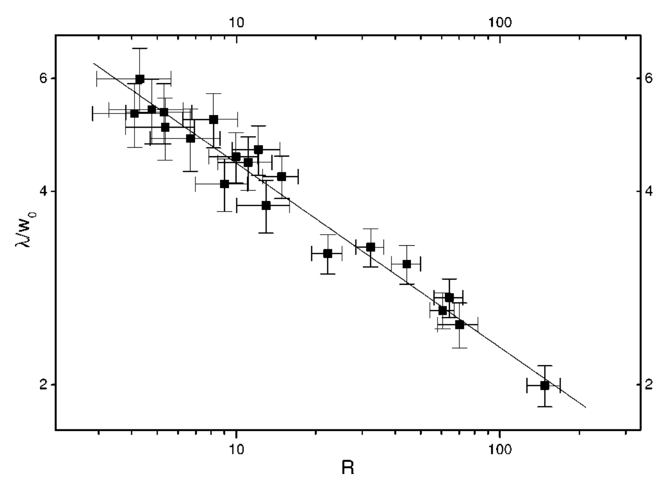

FIG. 5. Dominant wavelength in the Fourier spectrum of the unstable contact line λ in units of w_0. The solid line is the power law given by Eq. (10).

law relationship:

$$\lambda / w_0 = (8.34 \pm 0.35) R^{-0.27\pm0.022}. \quad (10)$$

Since $R \propto w_0^4$ [see Eq. (8)] and the exponent is close to $1/4$, the initial width w_0 cancels out. This implies that the dimensional value of λ is practically independent on w_0 and it is basically determined by the area A.

Huppert [1] characterizes the unstable contact line pattern in terms of the mean distance between fingers, namely $\langle\lambda\rangle$. It is expected that the mean wavelength $\langle\lambda\rangle$ be comparable with the dominant wavelength λ. He summarizes his experimental results with the expression

$$\langle\lambda\rangle = 7.5(A^{1/2} a^2 / \sin \alpha)^{1/3}. \quad (11)$$

Note that the dependence of $\langle\lambda\rangle$ on A has exponent $1/6$ instead of 0.27 as given by Eq. (10). This shows that the spreadings with small Bond number correspond to a different regime, possibly because the rolling motion is completely absent.

A key point in the analysis is that Eq. (7) establishes that the solution of the problem depends only on R [see Eq. (8)]. Thus, even though all the experiments reported here have been carried out on a vertical substrate ($\alpha = \pi/2$), it is possible to infer the general dependence of λ on the inclination angle α. As a consequence, under the assumption given by Eq. (5), λ must depend on $\sin \alpha$ with the same exponent as it depends on A. This points out another difference with the large cross section experiments, where the exponents on A and $\sin \alpha$ are different [see Eq. (11)]. Analogously, even if we employ here only cylindrical cap shapes for the cross section of the initial condition, a power law dependence on I with an exponent equal to minus the exponent on A is expected, i.e., ≈ -0.27.

In summary, we experimentally study microfluidic spreadings on vertical substrates in which, contrary to most of the previous experiments in the literature, capillary effects are significant from the very beginning of the process. We produce controllable and reproducible initial conditions, and study the evolution of the early stable stage. Interestingly, experiments with different areas and widths can be studied by employing a single dimensionless parameter R. Among other results, this analysis shows that the critical contact line position at the onset of the instability is proportional to the width w_0 and it is independent of the area A. In the unstable stage, the contact line pattern is spatially characterized by its dominant wavelength λ. We find that λ depends on the cross section area A with an exponent higher than previously reported in experiments performed with much larger areas. Also, we find that λ is practically independent on the initial width of the fluid, w_0.

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