On the Generation and Spreading of 'Finger' Instabilities in Film Coating Processes

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1 Introduction

Spreading viscous films are of considerable interest in industrial applications, especially in coating processes, where, in general, a uniform quality of the final coating is preferred. Experiments show that the contact-line of, in this case, liquid films driven down an inclined plane through gravity, rapidly destabilizes, forming either sawtooth or finger patterns. For the latter situation, portions of the contact-line may stop to move at all, leaving uncovered regions on the plane. Theoretical investigations using linear stability analysis for the lubrication approximation are in good agreement with experimental results for large to moderate inclination angles [2,4,8,12]. However, for small inclination angles, they predict stability in contrast to experimental observations.

The present work studies this regime. We use a contact-line model and investigate variations in the slip parameter as a possible source of instability, in analogy to the non-constant precursor height used by Bertozzi and Brenner [2]. Using a linear analysis, we find a considerable impact on the fluid profile and contact-line. Furthermore, for the first time for this model, we attempt a step into the nonlinear regime by a weakly nonlinear analysis which includes two additional modes, and find that they reinforce the response shown by the linearized model.

2 Formulation

We introduce a coordinate system attached to the inclined plane, with the x, y, z axis pointing in the stream-, spanwise, and normal direction, respectively. $h(x, y, t)$ denotes the height of the fluid profile, and $\Gamma(y, t)$ is the position of the moving contact-line.

The bulk of the liquid is governed by the Navier-Stokes equation, with a contribution from gravity $\rho \boldsymbol{g} = (\rho g \sin\alpha,\ 0,\ -\rho g \cos\alpha)$, and by the equation of continuity. Here, ρ denotes the fluid density, g the gravitational constant, and α is the inclination angle. At the liquid/gas interface, we have a kinematic boundary condition, a pressure jump due to surface tension, and continuity of tangential stress. For thin films of sufficiently viscous fluids, lubrication theory allows these equations to be used in a simplified form (see, for example,

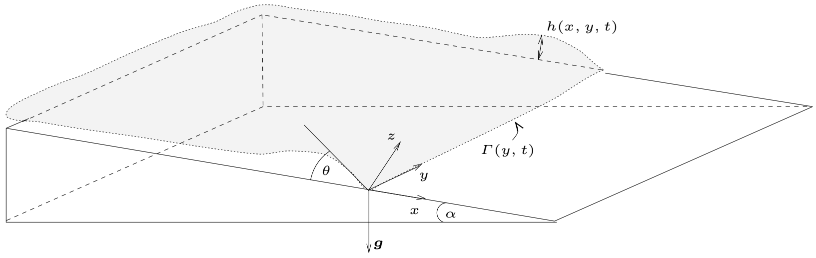

Fig. 1. A thin fluid sheet spreading down an inclined plane.

[5], [6]), namely

$$p_x = \rho g \sin\alpha + \mu u_{zz}, \quad p_y = \mu v_{zz}, \quad p_z = -\rho g \cos\alpha, \quad u_x + v_y + w_z = 0, \quad (1)$$
$$h_t + u h_x + v h_y = w, \quad p - p_a = \sigma(h_{xx} + h_{yy}), \quad u_z = v_z = 0 \ \text{ at } z = h, \quad (2)$$

where p, u, v, w are the pressure field and the three velocity components, respectively, μ is the viscosity, σ the surface tension, and p_a the external pressure.

At the contact-line, h vanishes, and we assume a linear relationship between the dynamic contact angle θ and the velocity of the contact-line, valid for small velocities [3],

$$\Gamma_t (1 + \Gamma_y^2)^{-1/2} = \kappa(\theta - \theta_S), \qquad \tan\theta(y,t) = -h_x|_{x=\Gamma(y,t)} (1 + \Gamma_y^2)^{1/2},$$

where κ is a material constant and θ_S the static contact angle. Far upstream, we assume a flat film with a constant fluid height h_∞ and a constant flow rate.

Since the plate is assumed to be at rest and impermeable, the normal velocity component vanishes for $z = 0$. For the tangential components u and v, in order to avoid the stress singularity at the moving contact-line, we allow the fluid to slip, according to

$$u = \lambda(x,y) h^{-1} \, u_z, \qquad v = \lambda(x,y) h^{-1} \, v_z, \qquad \text{at} \quad z = 0, \qquad (3)$$

with $\lambda(x,y) > 0$ small compared to the square of the typical fluid height away from the contact-line. This ensures that slip is negligible except very close to Γ, where h vanishes.

This contact-line model, but with constant slip parameter $\lambda(x,y) \equiv \lambda_0$, was used by Greenspan [5] to investigate the spreading of droplets on a horizontal surface and by Lopez et al. [8] for fluid spreading on an inclined plane.

Greenspan's model actually originates from a careful study by Neogi & Miller [10] of the spreading of fluids on porous surfaces, where the slip term coincides with (3) to leading order. So (3) may be considered to reflect the average effect of microscopic roughness on the velocity field near the contact-line. Therefore it appears reasonable to assume that variations in surface roughness affect the local value of λ. Instead of being constant, we therefore allow λ to vary about its average value λ_0, so that

$$\lambda(x, y) = \lambda_0(1 + \delta\varphi(x, y)). \tag{4}$$

In view of our later analysis, we assume the function φ, which describes the variations in the slip parameter, has order one and $\delta \ll 1$, but the model itself only requires $\lambda(x, y)$ to be positive.

We non-dimensionalize x, y, z, with h_∞ and t with $h_\infty \sin\alpha/V$, where the velocity scale is set by the upstream depth-averaged flow rate, i. e. $V = h_\infty^2 \rho g \sin\alpha(1 + 3\lambda_0 h_\infty^{-2})/\sigma$. We average (1) with respect to z and, by using the boundary conditions at the plane and the surface of the film, obtain

$$a^2(1 + d_0^2)h_t = -\nabla \cdot \Big[(h^3 + d_0^2(1 + \delta\varphi(x, y))h) \left(\nabla\triangle h - a^2\cos\alpha\nabla h + a^2\sin\alpha\begin{pmatrix}1\\0\end{pmatrix}\right)\Big], \tag{5}$$

$$D_1\Gamma_t = h_y\Gamma_y - h_x - \theta_S(1 + \Gamma_y^2)^{1/2}, \quad h = 0 \quad \text{at } x = \Gamma, \tag{6}$$

$$\lim_{x\to-\infty} h = 1, \qquad \lim_{x\to-\infty} \frac{\partial^\nu h}{\partial x^\nu} = 0, \qquad \nu \in \mathbb{N}. \tag{7}$$

The following dimensionless quantities appear in (5)–(7):

$$a^2 = \frac{\rho g h_\infty^2}{\sigma}, \qquad D_1 = \frac{\rho g h_\infty^2(1 + d_0^2)}{3\mu\kappa}, \qquad d_0^2 = \frac{3\lambda_0}{h_\infty^2}.$$

3 Linear Analysis

In experimental situations (see, for example, [4,7]) one observes that, after the fluid is released at the top of the plane, it initially forms a traveling wave with a straight contact-line. Indeed, in the absence of slip parameter variations ($\varphi \equiv 0$), inserting the ansatz

$$h(x, y, t) = h_0(x - t\sin\alpha), \qquad \Gamma(y, t) = t\sin\alpha,$$

into (5)–(7) and integrating once, one obtains

$$h_0''' - a^2\cos\alpha\, h_0' = a^2\sin\alpha\frac{1 - h_0^2}{h_0^2 + d_0^2},$$
$$h_0(0) = 0, \quad h_0'(0) = -D_1\sin\alpha - \theta_S,$$
$$h_0(-\infty) = 1, \quad h^{(\nu)}(-\infty) = 0, \qquad \nu \in \mathbb{N}.$$

A traveling wave has been computed numerically, [8,9] and derived asymptotically, for small inclination angles [12].

In experimental situations, the contact-line and the profile of the traveling wave quickly corrugate, forming a periodic structure, which rapidly evolves into a finger- or sawtooth-like pattern. Much of the work done so far has been devoted to investigating this instability by analyzing the growth/decay of initially infinitesimally small disturbances of the profile and the contact-line, but with a constant slip parameter ($\varphi \equiv 0$). This results in a linear eigenvalue problem for the growth/decay rate as a function of the wavenumber k. The predicted preferred wavelength, i.e. the one which maximizes the growth rate, was found to be in good agreement with experiments, for large to moderate inclination angles. For small α, however, some questions remain. The wavelength actually observed in experiments of, for example, Johnson [7] was consistently overestimated, [8,9], and in one case, the interval of measured wavenumbers was about the theoretical cut-off wavenumber k_c. Interestingly, in the second reference, Lopez et al. mention pinning through surface defects as a possible explanation for the decreased wavelength. Also, in the third author's PhD thesis [9] and in an upcoming paper [11], a long wave approximation shows that, for small θ_S/a, stabilization occurs below a certain critical inclination angle α^*. In experiments carried out by de Bruyn [4], however, instability has actually been observed at distinctively lower values. Furthermore, in an extension of the normal mode analysis into the weakly nonlinear regime, we found a supercritical bifurcation at the cut-off wavenumber, which hints to a further stabilizing nonlinear mechanism, see [9,11].

For this reason, we investigate a different mechanism as a cause of instability, where initial perturbations are neglected but a nontrivial shape for φ is assumed instead. More precisely, we attempt to assess the impact of slip parameter variations by analyzing the effect of a deviation from constant slip (4), localized in the streamwise direction, on h and Γ. We expect a response in the same order of $\delta \ll 1$, i.e, we decompose h and Γ as

$$h(x, y, t) = h_0(\xi) + \delta h^*(\xi, y, t), \tag{8}$$

$$\Gamma(y, t) = t \sin\alpha + \delta\gamma^*(y, t). \tag{9}$$

Here, we have simultaneously introduced new coordinates for the streamwise direction, $\xi = x - \Gamma(y, t)$, to map the spatial variables to a fixed domain, so that the contact-line is now represented by $\xi = 0$ independent of time t. This avoids having to expand the boundary conditions in terms of $\delta\gamma^*$, which is of some advantage especially for the weakly nonlinear extension of the analysis in the next section.

Inserting (8) and (9), and expanding in orders of δ, the zeroth order terms cancel out. Neglecting higher than first order terms yields a linear system of PDEs/ODEs for h^* and γ^*, with φ appearing as a forcing term. Since the coefficients are independent of y, we use the theory of Fourier transforms, and assume a special form for the slip variation φ and for the solution h^*,

γ^*, i.e.,

$$\varphi(x,y) = \varphi_1(x)e^{iky}, \tag{10}$$
$$h^*(\xi,y,t) = A(\xi,t)e^{iky}, \tag{11}$$
$$\gamma^*(y,t) = c(t)e^{iky}, \tag{12}$$

with a fixed wavenumber k. We thus obtain the following set of equations for A and c, where the hats have been dropped,

$$bA_t - b\dot{c}h_0' = \mathbf{P}(h_0,k^2;\partial_\xi^\mu)A + H(h_0,k^2;\xi)\,c + \frac{\partial}{\partial\xi}\left[\frac{\varphi_1(\xi + t\sin\alpha)d_0^2 h_0 b\sin\alpha}{h_0^2+d_0^2}\right], \tag{13}$$

$$A = 0, \quad D_1\dot{c} = -A_\xi \quad \text{at} \quad \xi = 0, \quad \lim_{\xi\to-\infty}\frac{\partial^\nu A}{\partial\xi^\nu} = 0, \quad \nu\in\mathbb{N}_0. \tag{14}$$

where the abbreviation $b = a^2(1+d_0^2)$ has been used. $\mathbf{P}(h_0,k^2;\partial_\xi^\mu)$ denotes the following differential operator,

$$\begin{aligned}\mathbf{P}(h_0,k^2;\partial_\xi^\mu) = & -(h_0^3+d_0^2h_0)\frac{\partial^4}{\partial\xi^4} - (3h_0^2+d_0^2)h_0'\frac{\partial^3}{\partial\xi^3}\\ & +(h_0^3+d_0^2h_0)(2k^2+a^2\cos\alpha)\frac{\partial^2}{\partial\xi^2}\\ & +\left[(3h_0^2+d_0^2)h_0'(k^2+a^2\cos\alpha) - \frac{2bh_0^2}{h_0^2+d_0^2}\sin\alpha\right]\frac{\partial}{\partial\xi}\\ & -\left[(h_0^3+d_0^2h_0)(k^4+k^2a^2\cos\alpha) + \frac{4bd_0^2h_0h_0'}{(h_0^2+d_0^2)^2}\sin\alpha\right]\end{aligned} \tag{15}$$

and $H(h_0,k^2;\xi)$ the following function,

$$\begin{aligned}H(h_0,k^2;\xi) = & -\left[(h_0^3+d_0^2h_0)h_0''\right]_\xi k^2 + k^4(h_0^3+d_0^2h_0)h_0'\\ & -k^2(a^2\sin\alpha(1-h_0^2)h_0).\end{aligned} \tag{16}$$

Note that here, due to the choice of coordinates, the traveling wave is stationary, but the support of φ is shifted upstream with a rate given by $\sin\alpha$.

For φ_1, we choose a smooth bump of length $x_1 - x_0 > 0$ located at a distance $x_0 > 0$ of the initial position of the contact-line,

$$\varphi_1(\cdot) = \begin{cases} \exp\left(-\dfrac{\tilde{x}^2}{1-\tilde{x}^2}\right) & \text{if} \quad x_0 \le \cdot \le x_1, \\ 0 & \text{otherwise,}\end{cases} \tag{17}$$

where $\tilde{x} = 2(\cdot - x_0)/(x_1 - x_0) - 1$.

Before the traveling wave has reached this position, we assume it to remain unperturbed,

$$A(\xi, 0) = 0 \qquad \text{for all} \quad \xi, \qquad c(0) = 0. \tag{18}$$

We remark that, even as the fluid passes over the slip parameter variation, the forcing term remains bounded by $\sim \sin\alpha$ and in fact is negligible (of order $\ll d_0^2$) everywhere except near the contact-line $\xi = 0$. Thus, its impact on the spreading diminishes as soon as the contact-line seizes to intersect with the support of $\varphi_1(\xi + t\sin\alpha)$.

We solve (13)–(18) numerically using a pseudo-spectral discretization in ξ and an implicit Euler in time, for a choice of parameters based on a typical experimental situation [7], i. e. $a^2 = 0.265$, $\theta_S = 0.178$ and $D_1 = 1$. For d_0^2 we chose a value for which we could obtain reliable numerical results, $d_0^2 = 0.003$. α was set to 0.0525, which is below the critical angle, and $|x_1 - x_0|$ was 21.44.

The impact of the slip parameter variation on the profile and the contact-line of the travelling wave as it passes over the localised slip parameter variation is shown in fig. 2, where $||A|| := \sup_\xi |A(\xi, t)|$. We see that the contact-line is modified by a perturbation on the order of $0.4 \ldots 0.6$. For values of δ within the range of validity of linear theory, $\delta \approx 0.5$, we obtain a corrugation of $0.2 \ldots 0.3$ lengthscales. The effect on the profile appears to be much smaller, but we remark that $||A||$ only measures the perturbation amplitude in the fixed domain coordinates (ξ, y, t). Transforming back to physical coordinates would yield an additional contribution related to the contact-line corrugation. Trials with different d_0^2 showed that the maximum value of $||A||$ and c depended only weakly on d_0^2.

Since we are in a regime of α where the linear system is stable in the sense that for all k unforced perturbations eventually die off, $||A||$ and c decay exponentially after the contact-line has left the region where the slip parameter varies. However, at its peak, the perturbation could be strong enough to incite possibly amplifying nonlinear effects. We shall address this question in the next section.

4 Weakly Nonlinear Analysis

The aforementioned mechanism cannot give rise to a persistent instability without the aid of nonlinear effects. This raises at least two questions, whether the induced perturbations of the profile/contact-line are sufficient to excite the nonlinearities and whether they amplify or stabilize the perturbations.

A definite answer could be obtained by solving (5) – (7) directly, with an unperturbed traveling wave as initial data. But doing so even numerically is a demanding task, since a strongly nonlinear, spatially two dimensional and time dependent problem has to be solved. The high order of the PDE and the presence of small length-scales – d_0^2 – near the contact-line, which have to be resolved accurately, further contribute to the difficulties. Until now, we believe, no two-dimensional solution of a problem like this has been performed.

As a first step to answering the two questions we instead generalize an approach used by Benney [1] for the investigation of surface waves on falling liquid films, and perform a weakly nonlinear analysis. Here, we study how the evolution of the profile/contact-line perturbation is modified if the interaction of the fundamental mode (of order δ) with itself and with the side modes it forces predominantly (at order δ^2) is taken into account. Thus in replacement of the single-mode ansatz (8), (9) and (11)–(12) we put

$$\begin{aligned} h(\xi,y,t) &= h_0(\xi) + \delta\left\{A_1(\xi,t)e^{iy} + \bar{A}_1(\xi,t)e^{-iy}\right\} \\ &\quad + \delta^2\left\{A_0(\xi,t) + A_2(\xi,t)e^{2iy} + \bar{A}_2(\xi,t)e^{-2iy}\right\}, \qquad (19) \\ \Gamma(y,t) &= t\sin\alpha + \delta\left\{c_1(t)e^{iy} + \bar{c}_1(t)e^{-iy}\right\} \\ &\quad + \delta^2\left\{c_0(t) + c_2(t)e^{2iy} + \bar{c}_2(t)e^{-2iy}\right\}, \qquad (20) \end{aligned}$$

where again $\delta \ll 1$ and the A_l, c_l are possibly complex valued functions (except for $l = 0$) and the bar denotes complex conjugation. For φ, rather than just taking twice the real part of (10), we assume a slightly more general form, where we include contributions to the side mode at the same order of magnitude at which they are forced by the fundamental mode, i.e., consistent with (19) and (20),

$$\begin{aligned} \varphi(x,y) &= \varphi_1(x)e^{iky} + \bar{\varphi}_1(x)e^{-iky} \\ &\quad + \delta\left\{\varphi_0(x) + \varphi_2(x)e^{2iky} + \bar{\varphi}_2(x)e^{-2iky}\right\}. \qquad (21) \end{aligned}$$

We insert (19)–(21) into (5)–(7), after having mapped the latter to a fixed domain using $\xi = x - \Gamma(y,t)$, and, since we intend to include only those interactions with the strongest impact on the fundamental mode, retain only terms up to third order in δ. Note that (21) too has to be expressed in terms of the fixed domain coordinates (ξ, y, t), so that, in these coordinates, the variation of the slip parameter explicitly depends on time. The zeroth order terms fulfill the traveling wave equation, so they drop out and we divide the equations by δ or δ^2, respectively. We obtain an initial boundary value problem for a nonlinearly coupled system of PDEs, which we solve using a pseudo-spectral and implicit Euler discretization, for, as in the linear case, vanishing initial data.

Examples of the result of the computations are shown in fig. 3. Here, we used the bump profile (17) for all three functions φ_0, φ_1, φ_2 but with $|x_1 - x_0|$ set to 42.88, and $a = 0.35$, $\theta_S = 0.25$, $\alpha = 0.0525$, for the parameters. D_1 and d_0^2 were as before, and the wavenumber was $k = 0.165$, to the right of k_c. For $\delta = 0$, we recover the linear behavior, where the corrugation eventually decays. For $\delta = 0.175$ and $\delta = 0.195$, the increase in amplitude is reinforced markedly, both for the profile and the contact-line perturbations. The impact increases for larger δ and, for the second of the two values shown here, we get a finite time blowup, interestingly for the amplitudes of the profile perturbation.

In both cases, care was taken that the fundamental mode clearly dominated the side modes, i. e.

$$||A_1|| \gg \delta ||A_2|| \qquad \text{and} \qquad |c_1| \gg \delta |c_l| \qquad l = 0, 2.$$

for all times, in order to be consistent with the ansatz (19)–(21).

In the absence of information about the neglected higher modes, though, we remark that the weakly nonlinear ansatz using (19)–(20) is strictly valid only in the limit of small amplitudes $\delta \searrow 0$. The influence of the higher modes in the exact solution may rapidly increase to equal order as the side modes retained in (19)–(20) and modify the solution's behavior. In fact, we expect such a mechanism to become important where the finite time blow-up of the three-mode approximation occurs in the case $\delta = 0.195$.

In spite of its limitations, the analysis performed here indicates that the profile/contact-line perturbation induced by slip parameter variations at a fraction of its value is enough to excite a strong nonlinear effect, one that is capable of promoting further destabilization.

5 Discussion and Conclusion

In this paper, we investigate the influence of variations of the slip parameter on the contact-line and profile of a traveling wave solution for a liquid film spreading down an inclined plane. We find a significant impact even for the linearized model. An extension using weakly nonlinear theory indicates that this effect is strong enough to incite a further destabilizing nonlinear mechanism, so that, the slip variations could play a role in the onset of a macroscopic, visible instability.

This compares favorably with a previous result by Bertozzi and Brenner [2] for the situation of completely wetting fluids. Instead of a slip law, they assume the presence of a very thin precursor layer in front of the contact-line, where the small precursor height b represents the average effect of the microscopic physics in the lubrication model, hence taking the role of the slip parameter d_0^2. They then place a small bump (of the same order b in height) on this precursor as a simple model for the microscopic fluctuations, and find that the solution of the linearized problem undergoes significant "transient growth" of order $0.3h_\infty$ and more as the bump traverses the front of the traveling wave, and argue that this is "enough" to incite nonlinear, possibly destabilizing effects which can lead to finger formation. Thus, both models give qualitatively similar answers, which indicates that our results reflect some essential effect of the physics at the contact-line and are not a mere artifact of a special way in which we introduce the (still incompletely understood) microscopic physics into the lubrication model.

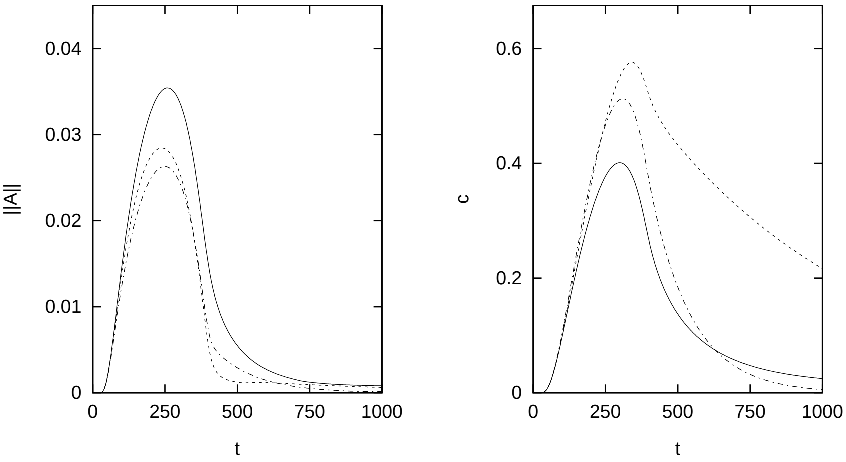

Fig. 2. Evolution of amplitude of the perturbation of the profile (**left**) and contact-line (**right**) in time, for wavenumbers $k = 0$ (—), $k = 0.125$ (- - -) and $k = 0.25$ (- · · -).

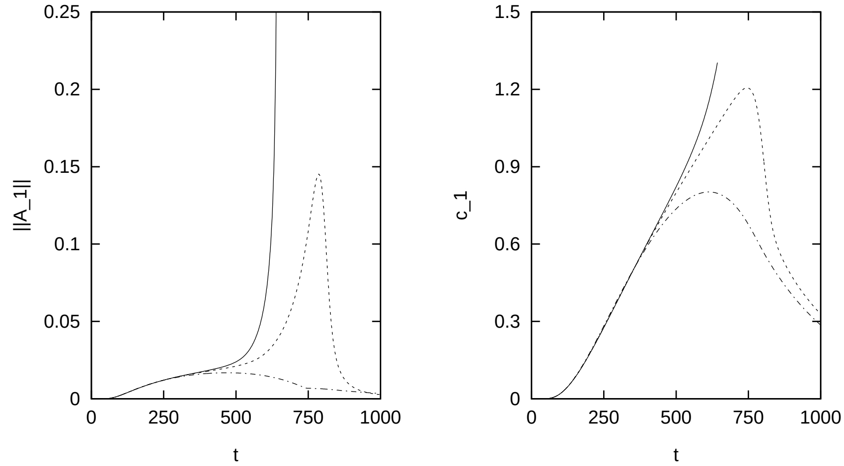

Fig. 3. Evolution of the amplitude of the perturbation of the profile (**left**), and contact-line (**right**), for the fundamental mode, with $\delta = 0.195$ (—), $\delta = 0.175$ (- - -) and $\delta = 0$ (- · · -).

An interesting aspect of our findings is that the slip variation needs to act for a sufficient length of time in order to have a macroscopic impact. Remember that in our case the support of φ_1 had a length of 21 or more dimensionless units, which, for typical $h_\infty \sim 1$ mm, corresponds to 2 cm or more. A bump of comparable length was also used by Bertozzi and Brenner in their numerical trials. This could originate from surface defects which pin the contact-line, surface contaminations or small but long deviations from a perfectly flat plane.

Systematic experimental studies for small inclination angles should be done to verify our predictions, for example, about the critical inclination angle. Perhaps one could also probe our and Bertozzi & Brenner's findings [2] by looking at how deliberately introduced variations of the plate's surface affect the spreading.

References

1. D. J. Benney. Long waves on liquid films. *J. Math. and Phys.*, 45:150–155, 1966.
2. A. L. Bertozzi and M. P. Brenner. Linear stability and transient growth in driven contact lines. *Phys. Fluids*, 9(8):530–539, 1997.
3. T. D. Blake and J. M. Haynes. Kinetics of liquid/liquid displacement. *J. Colloid and Interface Sci.*, 30:421–423, 1969.
4. J. R. de Bruyn. Growth of fingers at a driven three-phase contact line. *Phys. Rev. A*, 46(8):R4500–R4503, 1992.
5. H. P. Greenspan. On the motion of a small viscous droplet that wets a surface. *J. Fluid Mech.*, 84:125–143, 1978.
6. L. M. Hocking. Spreading and instability of a viscous fluid sheet. *J. Fluid Mech.*, 211:373–392, 1990.
7. M. F. G. Johnson, R. A. Schluter, M. J. Miksis, and S. G. Bankoff. Measurements of wavelength, dynamic contact angles, front velocity and shape of rivulets on an inclined plate by fluorescent imaging. *Preprint*, 1996.
8. P. G. López, M. J. Miksis, and S. G. Bankoff. Inertial effects on contact line instability in the coating of a dry inclined plate. *Phys. Fluids*, 9(8):2177–2183, 1997.
9. A. Münch. *Untersuchung der Fingerinstabilität einer sich auf einer schwach geneigten Ebene ausbreitenden Flüssigkeitsschicht.* PhD thesis, Zentrum Mathematik, Technische Universität München, 1998.
10. P. Neogi and C. A. Miller. Spreading kinetics of a drop on a rough solid surface. *J. Colloid and Interface Sci.*, 92(2):338–349, 1983.
11. B. A. Wagner and A. Münch. On the linear and weakly nonlinear stability of a thin film spreading down a slope. *In preparation.*
12. B. A. Wagner and A. Münch. Numerical and asymptotic results on the linear stability of a thin film spreading down a slope of small inclination. *To appear in EJAM*, 1998.