Available online at www.sciencedirect.com
SCIENCE DIRECT®
Physica D 209 (2005) 105–116
PHYSICA D
www.elsevier.com/locate/physd

Transient growth in driven contact lines

Roman O. Grigoriev*

School of Physics, Georgia Institute of Technology, 837 State St., Atlanta, GA 30332-0430, USA

Available online 11 July 2005

Abstract

The problem of transient growth in the distortion of driven contact lines has recently sparked a controversy as to whether this mechanism can provide an alternative route to pattern formation in the absence of linear instability. To resolve the disagreement between previous studies we conduct a generalized linear stability analysis of different lubrication models of gravity-driven spreading and compare our results with those based on direct numerical simulations. We find that linear and non-linear theory are in reasonable qualitative agreement and show that the quantitative discrepancies in the predicted transient growth are caused by the differences in: (1) the choice of initial disturbances and (2) the definition of the maximal transient amplification used in different studies. We further show by comparing the predictions of the precursor and the slip model that the latter substantially underestimates transient growth by neglecting the disturbances in the slip parameter.

Keywords: Contact line dynamics; Free-surface flows; Stability; Transient growth

1. Introduction

The dynamics of gravitationally driven contact lines have received a lot of attention over the past two decades [1–5]. More recently attention has shifted to re-examination of the validity of the linear stability analysis which underlies most theoretical studies, prompted by a paper due to Bertozzi and Brenner [6] who pointed out that existing experimental data of de Bruyn [4] appear to suggest that the liquid films spreading on solid surfaces at low angles of inclination could undergo a contact line instability where the conventional hydrodynamic linear stability analysis predicted stable evolution. A similar inconsistency has been pointed out by Münch and Wagner [7] who analyzed the data of Jarrett and de Bruyn [5].

Strong transient growth of spontaneous disturbances was proposed [6] as the mechanism for instability in that regime. A similar mechanism is believed to be responsible for destabilization of laminar high-Reynolds number shear flows [8,9]. Since low-Reynolds number coating flows also have strong shear, it is natural to expect somewhat similar dynamics (in fact, transient growth was also found

* Corresponding author. Tel.: +1 404 385 1130;
fax: +1 404 385 2506.
E-mail address: roman.grigoriev@physics.gatech.edu.

0167-2789/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.physd.2005.06.015

in thermocapillary-driven films [10,11]). Shear of the base flow is an essential ingredient leading to transient growth of disturbances, as it is partially responsible for making the evolution operator of the linearized system strongly non-normal, with many nearly degenerate eigenfunctions (non-uniformity in the thickness of the film is the other essential ingredient [10]). The connection between non-normality and transient growth is very easy to understand by recalling the fact that the general solution of a linear differential equation with a degenerate characteristic polynomial is given in terms of a linear superposition of products of powers and exponentials, so that even in a linearly stable system asymptotic exponential decay can be preceded by transient algebraic growth. As a result, linear stability becomes a poor predictor of short-time dynamics, prompting the need for a *generalized* linear stability analysis [12] based on detailed study of transients.

In order for the transient growth mechanism to destabilize a linearly stable (transversely uniform) base state and produce an observable distortion of the contact line, two conditions have to be met. First, transient growth has to occur for disturbances with *finite* transverse wave numbers (zero wave number disturbances do not lead to distortion of the contact line and hence will not be observed experimentally). Second, transient growth has to be strong enough to amplify *typical* spontaneous disturbances sufficiently for non-linear terms to become important (otherwise, the disturbances will eventually decay as predicted by the linear theory). However, so far no experimental investigations of this phenomenon have been reported, while the existing theoretical studies [6,13–15] produce rather contradicting results: predicted transient amplification at finite wave numbers goes as low as a factor of five [15] and as high as a factor of 800 [6], a difference of more than two orders of magnitude.

The paper is organized as follows: we first review the lubrication models of thin film dynamics and the results of linear stability analysis in Section 2. Section 3 contrasts different ways of describing and quantifying transient dynamics. The results of generalized linear stability analysis of the problem are presented in Sections 4 and 5 which treat, respectively, the transient dynamics of disturbances initially localized behind and ahead of the contact line. We conclude in Section 6 by discussing the origin of the differences in predictions of various theoretical analyses and suggest directions for further experimental studies of transient growth.

2. Lubrication models

In the following we will consider thin liquid films spreading on an inclined solid plane surface under the action of gravity. We will orient our coordinate system such that the x axis points in the direction up the plane (so that the film flows in the negative x direction), the y direction is in the plane transverse to the flow and z is perpendicular to the plane. In the lubrication approximation the evolution of the film in the *precursor model* [6,13,14] is described in terms of the fourth order non-linear partial differential equation (PDE) for the non-dimensional film thickness $h(x, y, t)$:

$$h_t + (h^3)_x + \nabla[h^3(\nabla\nabla^2 h - D\nabla h)] = 0, \tag{1}$$

where $D(\theta) = (3\mathrm{Ca})^{1/3}\cot(\theta)$, $\mathrm{Ca} = \mu v_c/\sigma$ is the capillary number and θ is the inclination angle (from horizontal) of the plane. Furthermore, $\mu, \rho, \sigma, v_c = \rho g h_c^2 \sin\theta/3\mu$ and h_c are, respectively, the viscosity, density, surface tension, the characteristic speed and the characteristic thickness of the spreading film. The corresponding length scales are h_c in the z direction, $l_c = (3\mathrm{Ca})^{-1/3}h_c$ in the x and y directions, and the time scale is l_c/v_c. In the constant flux case, which is easiest to analyze, (1) has to be solved subject to boundary conditions $h = b, h_x = h_{xxx} = 0$ for $x \to -\infty$ (flat precursor film ahead) and $h = 1, h_x = h_{xxx} = 0$ for $x \to \infty$ (flat tail behind the contact line). Respectively, the *slip model* [3,7,15] leads to a PDE

$$h_t + (h^3 + \alpha h)_x + \nabla[(h^3 + \alpha h)(\nabla\nabla^2 h - D\nabla h)] = 0, \tag{2}$$

where α is a phenomenological slip coefficient (sometimes defined as the square of the slip length $h_s \ll 1$). The corresponding constant flux boundary conditions are $|\nabla h| = c$ (constant slope at the contact line where $h = 0$) and $h = 1, h_x = h_{xxx} = 0$ for $x \to \infty$ (flat tail).

For the stated boundary conditions both models admit a traveling wave solution $h(x, y, t) = h_0(x + ut)$ describing a transversely uniform spreading film.

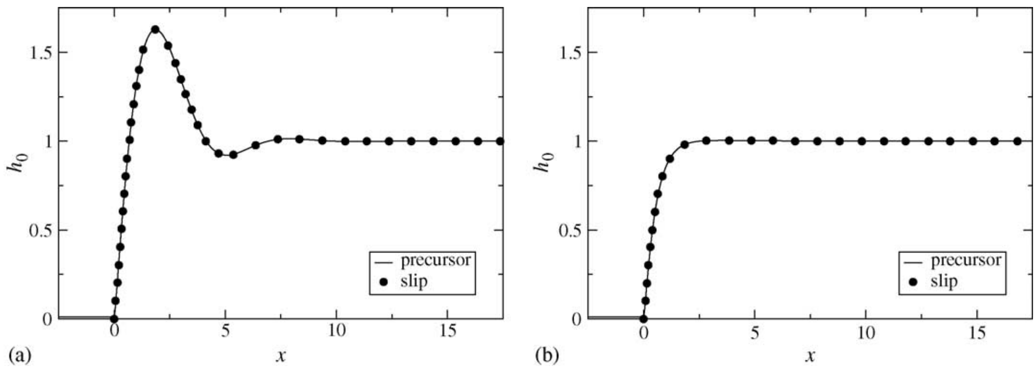

Fig. 1. Asymptotic film thickness profiles produced by the two models for $b = \alpha = 0.01$ and (a) $D = 0$ (90° inclination angle) or (b) $D = 5$ (10° inclination angle). The contact angle in the slip model is chosen to obtain the maximal thickness equal to that produced by the precursor model: (a) $c = 1.236$ and (b) $c = 1.303$.

Substituting h_0 into (1) and integrating once yields

$$h_0''' = \frac{(h_0 - 1)(h_0 - b)(h_0 + 1 + b)}{h_0^3} + Dh_0' \tag{3}$$

and the propagation speed $u = 1 + b + b^2$. Similarly, the slip model yields

$$h_0''' = \frac{h_0^2 - 1}{h_0^2 + \alpha} + Dh_0' \tag{4}$$

with $u = 1 + \alpha$. It is easy to see that these two equations coincide in the limit $\alpha, b \to 0$. Furthermore, for small enough b and α the numerically computed profiles are virtually indistinguishable with appropriate choice of parameters (see Fig. 1).

The stability of the asymptotic (traveling wave) state is determined by considering perturbed solutions $h(x, y, t) = h_0(x + ut) + g(x + ut, t)e^{iqy}$, which upon substitution into either (1) or (2) yield a linear evolution equation for the disturbance $g(x, t)$,

$$g_t = L(q)g, \tag{5}$$

with $L(q)$ a non-normal fourth order differential operator. (Explicit expressions for L along with the boundary conditions on g are given in [6] for the precursor and [15] for the slip model.) The leading eigenvalue β_0 of this operator for small q can be found analytically using long wave analysis. As shown in, e.g., Refs. [6,16] the corresponding left and right eigenfunctions of $L(0)$ are given by $f_0(x) = \text{const.}$ and $g_0(x) = h_0'(x)$. Using this fact the following expression has been derived [6] for the precursor model

$$\beta_0 = q^2 \int_{-\infty}^{\infty} \frac{(h_0-1)(h_0-b)(h_0+1+b)}{1-b} \mathrm{d}x + O(q^4). \tag{6}$$

Respectively, the slip model (2) yields

$$\beta_0 = q^2 \int_0^{\infty} h_0(h_0^2 - 1)\mathrm{d}x + O(q^4). \tag{7}$$

The last expression was originally derived in [7] with a different prefactor due to a different choice of non-dimensionalization. A quick comparison of (6) and (7) shows that good agreement between the growth rates predicted by the two models is expected for small b and α because, as we have already pointed out, in that limit the corresponding asymptotic state profiles are virtually identical. These expressions show that the asymptotic film profile becomes unstable to long wave length disturbances in the presence of a pronounced capillary ridge, e.g., for large inclination angles (as in Fig. 1a). Smaller inclination angles produce smoother film profiles (Fig. 1b) which are stable with respect to long wave length disturbances.

For larger values of q the leading eigenvalue, along with the rest of the spectrum, can be computed numerically. The procedure is rather standard [15,17] and hence will not be described here. However, for the following it will be important to note that the evolution operator is discretized on a finite domain of overall length l in the x direction. Taking the contact line to be at $x = 0$, this corresponds to $0 < x < l$ for the slip model or $-l_p < x < l - l_p$ for the precursor model, with

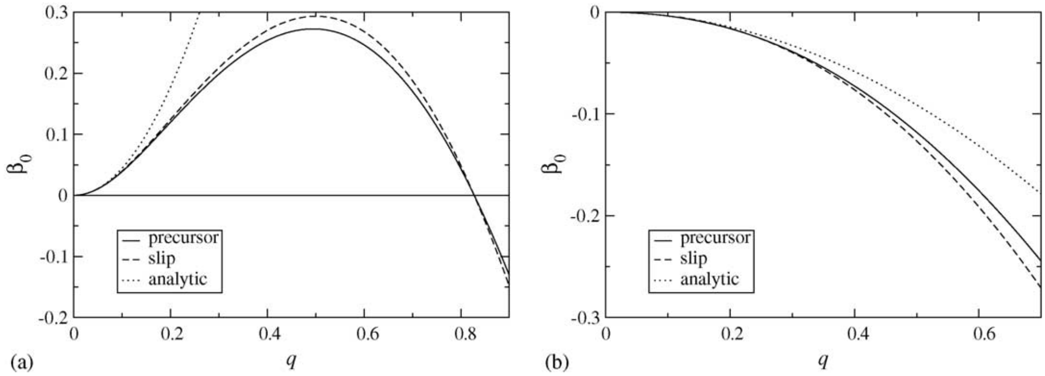

Fig. 2. The leading eigenvalue β_0 as a function of the transverse wave number q for (a) $D = 0$ and (b) $D = 5$. The other parameters are as in Fig. 1. The dotted curves correspond to the asymptotic result (7).

l_p the width of the numerical precursor film. Unless stated otherwise, we take $l = 20$ and $l_p = 0.16$ in the remainder of the paper. Fig. 2 compares the asymptotic result (7) with numerically computed values, showing excellent agreement for small q. Fig. 2b also confirms that at small inclination angles the asymptotic state h_0 is linearly stable for all wave numbers. Interestingly enough, the predictions of the two models regarding the wave number of the fastest growing disturbance in the unstable case (Fig. 2a) agree very well, while the corresponding growth rates differ by about 7%. In comparison, the difference at small q is below 1%.

3. Transient growth

For strongly non-normal evolution operators the leading eigenvalues (such as those shown in Fig. 2) provide a poor description of the short-term dynamics. We therefore turn to the generalized stability analysis, which takes into account the full spectrum of eigenmodes. As pointed out previously, even in a linearly stable system (such as a film spreading down a slope at small angle of inclination) disturbances can undergo transient growth before asymptotic decay eventually sets in. The strength of transient growth can be characterized by computing the norm of the amplified disturbance $\delta h(x, y, t)$ and comparing it with the norm of the initial disturbance, say $\delta h(x, y, 0) = g(x, 0)e^{iqy}$, giving the following measure:

$$\gamma_p(q, t) = \frac{\|\delta h(x, y, t)\|_p}{\|\delta h(x, y, 0)\|_p} = \frac{\|\delta h(x, y, t)\|_p}{\|g(x, 0)\|_p}, \tag{8}$$

where $\|\ \|_p$ denotes the conventional L_p-norm. Such a definition with $p = \infty$ was used in several studies based on the precursor model. For instance, Bertozzi and Brenner [6] used initial disturbances

$$g(x, 0) = s\left(1 - \frac{(x - x_c)^2}{w^2}\right)^2 \sin(kx),$$
$$x_c - w < x < x_c + w \tag{9}$$

with a fixed amplitude s, width $w = 20$ and modulation wave number $k = 1/3$ in the x direction [18]. Kondic and Bertozzi [13] chose a family of functions

$$g(x, 0) = -s\exp\left[-4\ln 2\frac{(x - x_c)^2}{w^2}\right] \tag{10}$$

with adjustable parameters s and w. Finally, Ye and Chang [14] used the essential eigenfunctions of $L(q)$ in place of the initial disturbances. Essential eigenfunctions are plane waves

$$g(x, 0) = se^{ikx} \tag{11}$$

ahead of the contact line and decay exponentially fast behind it. Although easy to compute, the definition (8) provides somewhat limited information, since the amount of transient amplification may (and in fact does) depend quite sensitively on the choice of initial disturbances $g(x, 0)$. One should therefore regard (8) as a lower bound on transient amplification. Yet all three studies find strong transient growth that can easily amplify initial disturbances by two orders of magnitude or more. Furthermore, in all of these studies transient amplification achieves its maximum at a nonzero value of the transverse wave number q, suggesting that transient amplification can, indeed, provide an alternative mechanism for the contact line instability.

A different approach is to define transient amplification as a maximum of (8) over all initial conditions. Although one clearly cannot sample all initial conditions, in the linear theory where $\delta h(x, y, t) = g(x, t)e^{iqy}$, the resulting quantity can be related to the matrix norm of the evolution operator [9]

$$\gamma_p(q, t) = \sup_{g(x,0)} \frac{\|g(x, t)\|_p}{\|g(x, 0)\|_p} = \left\|e^{L(q)t}\right\|_p, \tag{12}$$

which can be computed numerically using the standard technique of spectral decomposition. This latter definition, with $p = 2$, was used by Davis and Troian in the most recent study [15] based on the slip model. The authors found transient amplification to be weak (barely more than an order of magnitude) and peak at zero wave number, contrary to the results of previous studies, and therefore incapable of producing a significant distortion of the contact line.

In the unstable band transient growth is followed by exponential growth, rather than decay, so (12) produces a diverging result. Factoring out the exponential growth as in Refs. [6,10], we can generalize (12) to give an expression

$$\gamma_p(q, t) = \sup_{g(x,0)} \frac{\|g(x, t)\|_p}{\|g(x, 0)\|_p} e^{-\beta_0(q)t} = \|e^{[L(q)-\beta_0(q)]t}\|_p \tag{13}$$

describing the growth due to transient effects. Initial disturbances that are amplified the most at a given time t are called "optimal disturbances" or "optimal inputs" and can be found via the singular value decomposition of the evolution operator $e^{L(q)t}$ [12]. We will often use the maximal transient amplification for all times $\gamma_p(q) = \max_t \gamma_p(q, t)$ instead of (12) and (13).

4. Disturbances behind the contact line

We have computed transient amplification numerically from (12) and (13) for both the precursor and the slip model using different norms. With few exceptions the results are qualitatively similar for different p, so we will concentrate primarily on the ∞-norm considered in the majority of studies. Transient amplification as a function of the wave number is shown in Fig. 3 and provides information complementary to the dispersion relation $\beta_0(q)$ shown in Fig. 2. In the slip model we find that transient amplification increases with decreasing q, reaching a maximum of a few tens at $q = 0$, with a rather weak dependence on the parameter D describing the inclination angle of the plane.

Our results for the slip model are qualitatively and quantitatively similar to those of Davis and Troian [15] who used similar parameters but a different norm, $p = 2$. Direct comparison of magnitudes of transient amplification computed in different studies, however, should be made with care: the magnitude of transient amplification depends both on its definition (the choice

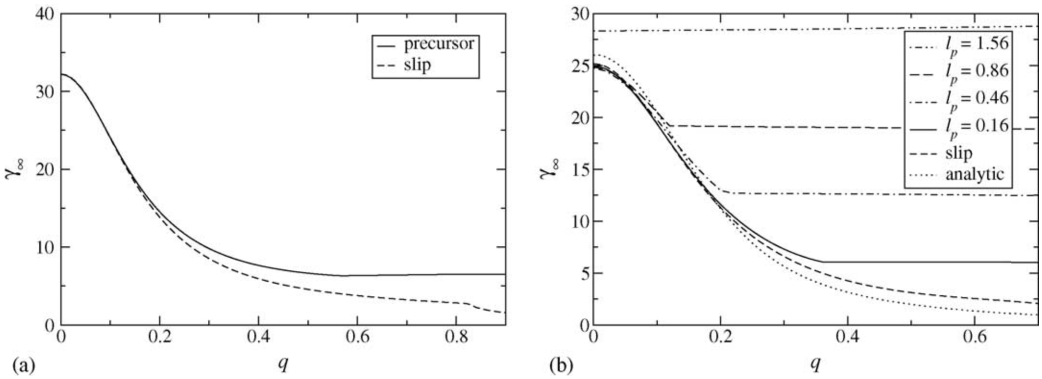

Fig. 3. Maximal transient amplification γ_∞ as a function of the transverse wave number q for (a) $D = 0$ and (b) $D = 5$. The precursor width in (a) is $l_p = 0.16$ and in (b) is as indicated. The other parameters are as in Fig. 1. The dotted curve in (b) shows the analytic result (18).

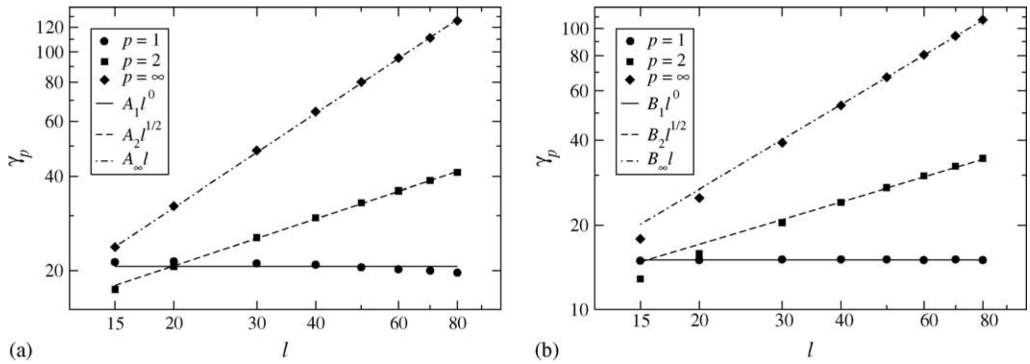

Fig. 4. Maximal transient amplification γ_p at $q = 0$ as a function of the system size l for (a) $D = 0$ and (b) $D = 5$. The other parameters are as in Fig. 1. A_i and B_i are constants determined from a least squares fit.

of the norm) and on the size l of the computational domain on which the evolution operator is discretized. For instance, our calculations for different l presented in Fig. 4 show that $\gamma_1 \approx$ const., while $\gamma_2 \propto l^{1/2}$ and $\gamma_\infty \propto l$, when computed at $q = 0$. This dependence can be understood by considering the structure of the leading eigenfunction g_0 and its adjoint f_0.

Numerical calculations show that γ_p, for any p and $q = 0$, achieves a maximum for $t \to \infty$ (this is a global maximum for the slip model, but may be a local maximum for the precursor model as we will see later). Using the spectral decomposition of the matrix exponential we obtain

$$\mathrm{e}^{L(0)t} = \sum_n g_n \mathrm{e}^{\beta_n(0)t} f_n^\dagger \to g_0 f_0^\dagger, \quad t \to \infty, \tag{14}$$

as $\beta_0(0) = 0$ is the only non-negative eigenvalue. Hence, the maximal transient amplification is achieved for the optimal initial disturbances equal to multiples of f_0. The evolution amplifies these disturbances and transforms them into multiples of the leading eigenfunction g_0 as $t \to \infty$. From (12) and (14) we have

$$\gamma_p(0) = \frac{\|g_0 f_0^\dagger f_0\|_p}{\|f_0\|_p} = \frac{\|g_0\|_p \|f_0\|_2^2}{\|f_0\|_p}. \tag{15}$$

Since $g_0(x) = h_0'(x)$ has an essentially finite support (it is exponentially small outside of the capillary shock region), we have $\|g_0\|_p =$ const. (with a different const. for different p) for l greater than the width of the capillary shock. Furthermore, $f_0(x) =$ const., so

$$\|f_0\|_p^p = \int |f_0(x)|^p \, \mathrm{d}x \propto l, \tag{16}$$

which immediately leads to $\gamma_p(0) \propto l^{1-1/p}$. Therefore, all γ_p with $p > 1$ diverge in the limit $l \to \infty$, so that, at least in principle, one can construct an initial disturbance which will produce arbitrarily large transient growth.

Restricting the size of the computational domain is equivalent to imposing an initial disturbance of finite spatial extent on an unbounded film. Initial disturbances with larger spatial extent are closer to the optimal disturbance $f_0 =$ const. and thus are amplified stronger. Once the identification of the computational domain size l with the width w of the initial disturbance is made, the blow-up of γ_p with l (or w) can be readily understood. For a film of infinite spatial extent the optimal initial disturbance $g(x, 0) = \delta h\, f_0$, with $\delta h =$ const. $\ll 1$, corresponds to a change in the thickness of the film's flat tail, $h_0 \to h_0 + \delta h$. From (1) and (2) we see that the contact lines of the undisturbed and the disturbed film will move with constant but different speeds, which for small enough b and α are given by $u \approx h_0^2(\infty) = 1$ and $u + \delta u \approx (h_0(\infty) + \delta h)^2 \approx 1 + 2\delta h$, respectively, leading to a linear growth in the separation $\delta x = \delta u\, t \approx 2\delta h\, t$. If such uniform initial disturbance extends only a finite (but large compared with the characteristic width of the capillary shock l_s) distance w from the contact line, the separation in the position of the two contact lines will grow linearly for a finite time t^*, while the disturbance is advected toward the contact line and then stop. This time t^* and the maximal separation δx^* can be computed using volume conservation: the initial volume difference $w\delta h$ has to be equal to the final volume difference $h_0(\infty)\delta x^* = \delta x^*$, yielding $\delta x^* = w\delta h$ and

$t^* = \delta x^*/\delta u \approx w/2$. We therefore obtain

$$\gamma_p(0) \approx \frac{\|g(x,t^*)\|_p}{\|g(x,0)\|_p} \approx \frac{\left(\int_0^\infty |g_0(x)\delta x^*|^p \, \mathrm{d}x\right)^{1/p}}{\left(\int_0^w |\delta h|^p \, \mathrm{d}x\right)^{1/p}} \propto w^{1-1/p}. \quad (17)$$

For initial disturbances with small (compared with l_s^{-1}) but non-zero wave numbers the distortion of the contact line will experience transient growth until the transverse modulation in the thickness of the film behind the contact line dies away due to the stabilizing action of gravity and surface tension. This modulation will decrease exponentially with the time scale $\tau(q) = (Dq^2 + q^4)^{-1}$, as can be seen from the evolution Eqs. (1) or (2) linearized about the flat profile $h(x, y) = 1$. Transient amplification factor can be estimated using the maximal distortion of the contact line. For instance, taking $p = \infty$ and assuming $\max_x |g_0(x)| \approx c$, the slope at the contact line, we obtain

$$\gamma_\infty(q) \approx 2c \int_0^{t^*} \mathrm{e}^{-t/\tau(q)} \, \mathrm{d}t = 2c\tau(q)(1 - \mathrm{e}^{-w/(2\tau(q))}) = cw - \frac{cDw^2}{4} q^2 + O(q^4). \quad (18)$$

Given the crudeness of this estimate, it is in reasonable agreement with our numerical result for $D = 5$, as Fig. 3b shows, even for relatively large q. For $q = 0$ transient amplification (18) grows linearly with w and can be made arbitrarily large by increasing the spatial extent of the disturbance. However, zero wave number disturbances do not lead to distortion of the contact line. For any non-zero q there is a finite upper bound independent of w,

$$\gamma_\infty(q) \lesssim 2c\tau(q) = 2c(Dq^2 + q^4)^{-1}, \quad (19)$$

which is a more practically important result.

5. Disturbances ahead of the contact line

The preceding analysis applies equally to the slip and the precursor model, assuming that the initial disturbances are localized behind the contact line. It should come as no surprise then that the precursor model produces transient amplification essentially identical to that of the slip model when the numerical precursor is very narrow (Fig. 3). For intermediate values of l_p we again find good agreement at small q, while at larger q there is a significant difference: $\gamma_\infty(q)$ levels off for the precursor model, with the height of the plateau which increases with l_p, while the slip model predicts a monotonic decay. This difference can be understood by looking at the time dependence of $\gamma_\infty(q, t)$. Fig. 5a shows that while the slip model produces a curve with a single maximum, in the precursor model transient amplification can peak twice. The first maximum is achieved rather quickly (at time $t_1 \approx l_p$ independent of q, as Figs. 5b and 6b show), while the second maximum is achieved much later (at about the same time t_2 when transient amplification peaks in the

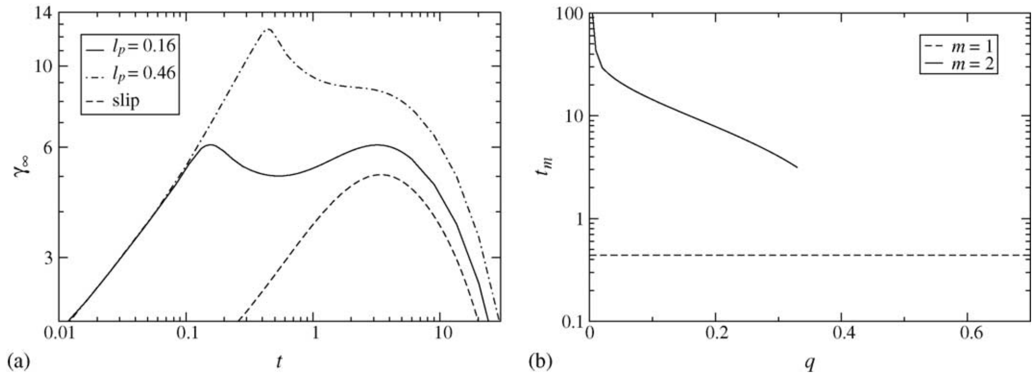

Fig. 5. (a) Transient amplification γ_∞ at $q = 0.36$ for the slip and precursor model as a function of time. The width of the numerical precursor film is as indicated. (b) The times at which the maxima in γ_∞ are achieved as a function of wave number in the precursor model with $l_p = 0.46$. The second maximum disappears at larger wave numbers. The other parameters are $D = 5$, $\alpha = b = 0.01$ and $c = 1.303$.

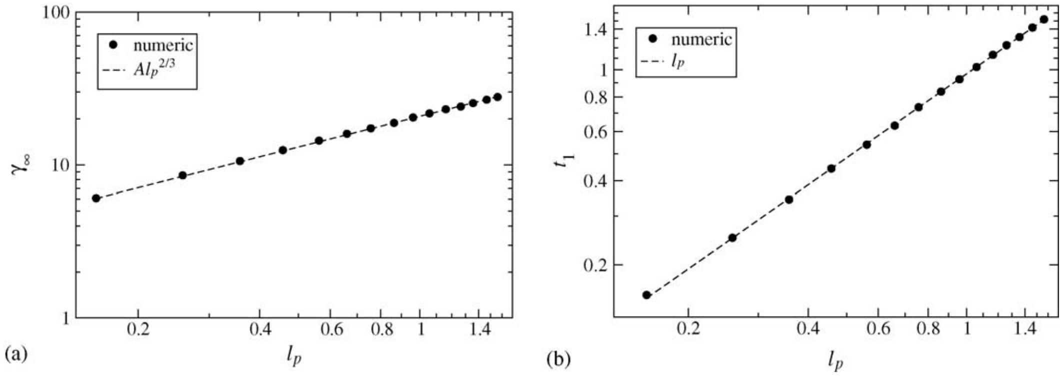

Fig. 6. (a) Transient amplification γ_∞ and (b) the time at which the maximum is achieved as a function of the precursor width l_p. We used a relatively large wave number $q = 0.6$ for which γ_∞ has a single maximum. The other parameters are $D = 5$ and $b = 0.01$. A is a constant determined from a least squares fit.

slip model). For relatively small precursor width l_p the second maximum dominates for small q and the first one for large q, explaining the crossover effect observed in the precursor model. At larger l_p the first maximum is dominant for all q, so one obtains transient amplification which is essentially independent of the transverse wave number of the initial disturbance (see Fig. 3b). Furthermore, Fig. 6a shows that $\gamma_\infty(q, t_1)$ scales as $l_p^{2/3}$ with the width of the numerical precursor film (for the relatively small values of l_p considered in this study).

These results can be understood qualitatively by identifying l_p with the spatial extent w_p of the initial disturbance in the physical precursor film extending to $x = -\infty$. Precursor thickness affects the shape of the capillary shock. For large inclination angles (Fig. 1a) the most obvious effect is the change in the thickness of the capillary ridge, as noted by Bertozzi and Brenner [6]. However, for small inclination angles (Fig. 1b) the capillary ridge disappears. In this regime it is more appropriate to consider the effect of the precursor thickness b on the slope of the capillary shock. This dependence (which follows from the non-linear theory) is shown in Fig. 7b. The data points are well fitted by a curve

$$\max_x h_0'(x) = A_2 + B_2(\ln b^{-1})^{1/3}, \tag{20}$$

which corresponds to the thickness profile $h_0 \sim x[\ln(x/b)]^{1/3}$ near the contact line computed by Dussan and Davis [19] and de Gennes [20]. The argument of Bertozzi and Brenner [6] for the scaling of transient

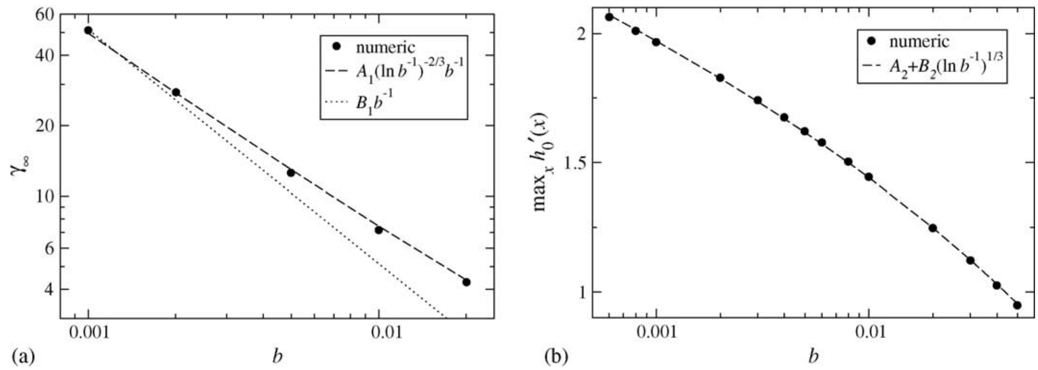

Fig. 7. (a) Transient amplification γ_∞ for $q = 0.6$ and (b) the maximal slope of the capillary shock as a function of the precursor thickness b. The other parameters are $D = 5$, $b = 0.01$ and $l_p = 0.2$. A_i and B_i are constants determined from a least squares fit.

amplification with b then gives

$$\gamma_\infty(0) \propto \max_x \frac{\partial h_0(x)}{\partial b} \propto \frac{\partial \max_x h_0'(x)}{\partial b} \propto (\ln b^{-1})^{-2/3} b^{-1}, \quad (21)$$

in good agreement with our numerical results obtained for fixed l_p (or w_p), as Fig. 7a shows. Previous studies mostly found power law scaling $\gamma_\infty(0) \propto b^\chi$ with various exponents χ. For instance, the study of Ye and Chang claimed the exponent to be -1, although the reported data (Fig. 6b of Ref. [14]) would, in fact, agree much better with a scaling relation such as (21). The limited data of Bertozzi and Brenner [6] roughly corresponds to the exponent -1. A later study by Kondic and Bertozzi [13] found the exponent to be closer to $-4/3$ for non-infinitesimal disturbances (their disturbances were comparable to the precursor thickness itself), which can be regarded as a non-linear effect. Fig. 7a also shows that the transient amplification in the precursor model deviates rather significantly from the b^{-1} scaling.

Next we turn to the description of time dependence starting, as in the previous Section, with the case $q = 0$. Once the contact line encounters a region of a locally thicker/thinner precursor (of width w_p) the capillary shock cannot adjust to the change in the precursor thickness instantaneously. Instead the deformation of the shock will continuously increase (while the contact line moves over the disturbed region of the precursor), reaching a maximum when the contact line completely crosses the disturbed region. This would happen at time $t_1 = w_p/u \approx l_p$ (recall, $u = 1 + b + b^2 \approx 1$ and $w_p = l_p$), in agreement with our numerically obtained scaling (Fig. 6b). According to this scenario, spatially uniform initial disturbances would produce the largest net amplification $\gamma_\infty(0)$. The optimal initial disturbance $g(x, 0)$ producing peak transient amplification at time t_1 can also be found numerically by computing the singular value decomposition of the evolution operator $e^{L(q)t}$. The result for $q = 0$ is shown in Fig. 8a. Indeed, we find $g(x, 0)$ to be nearly constant for $-l_p < x < 0$ (aside from a capillary shock at $x = -l_p$), which is consistent with our qualitative description. Most important, this optimal initial disturbance is quickly transformed by the evolution operator, such that at time t_1 its shape looks almost identical to the leading eigenfunction $g_0(x) = h_0'(x)$ near the contact line, as Fig. 8a shows. Therefore, the main effect of disturbances in the precursor thickness is to change the position of the contact line, not the thickness of the capillary ridge, as suggested by Bertozzi and Brenner [6].

As we increase l_p, wider and wider initial disturbances are allowed. Simultaneously t_1 increases, moving peak transient amplification toward later and later times, so that, according to the analysis of the previous section, the optimal initial disturbances approach multiples of the leading adjoint eigenfunction $f_0 =$ const. and are transformed by the evolution operator into the multiples of the leading eigenfunction $g_0(x) = h_0'(x)$, describing the translation of the contact line. The distortion corresponds to non-zero transverse wave numbers, for which we can invoke the same argument as before: the contact line will advance with a speed

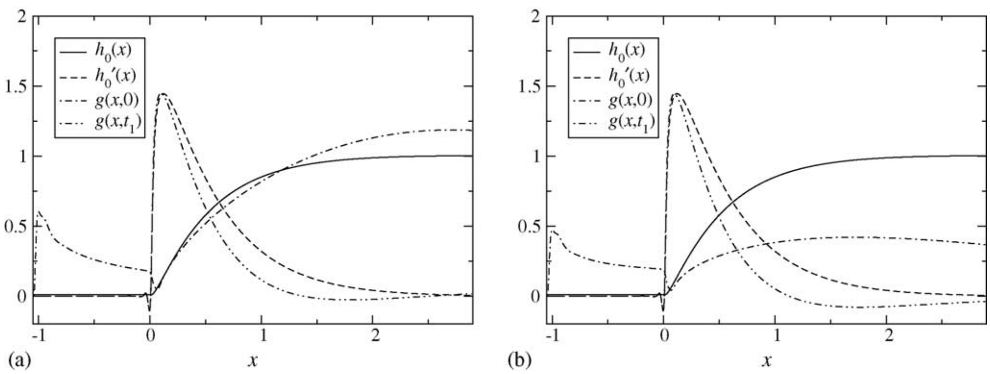

Fig. 8. Optimal initial disturbance $g(x, 0)$ that produces the largest disturbance $g(x, t)$ at the peak time t_1 for (a) $q = 0$ and (b) $q = 0.6$. The other parameters are $D = 5$, $b = 0.01$ and $l_p = 1.06$.

determined by the local thickness of the precursor. The transverse modulation in the thickness of the precursor will decrease exponentially with time, just as the modulation in the film thickness behind the contact line, but at a much slower rate $\tau(q) = b^{-3}(Dq^2 + q^4)^{-1}$. Effectively, for the range of wave lengths of interest and realistic values of b, the effect of this decay is completely negligible, explaining our earlier observation that $\gamma_\infty(q)$ becomes flat for the range of q where the first maximum in $\gamma_\infty(q, t)$ dominates (see Fig. 3). Indeed, a quick comparison of Fig. 8a and b shows that the shape of the optimal initial disturbance ahead of the contact line is essentially independent of q, while behind the contact line $g(x, 0)$ is sharply reduced for non-zero q — the modulation behind the contact line dies out much quicker and is therefore less capable of producing large transient growth than the modulation ahead of the contact line which persists for a long time.

This picture suggests the following scaling of the maximal transient amplification

$$\gamma_p(q) \propto (\ln b^{-1})^{-2/3} b^{-1} w_p^{1-1/p} \tag{22}$$

for w_p large (again compared with the capillary shock width l_s). At small $l_p(= w_p)$ our numerical computations also produce a power law scaling for $\gamma_\infty(q)$ with l_p (see Fig. 6a), but with exponent 2/3 rather than 1, as predicted by (22). This disagreement should not be too alarming — when the peak amplification occurs at small times $t_1 \approx w_p$ the decomposition (14), which forms the basis for the scaling, is not yet valid. However, we expect $\gamma_\infty(q)$ to approach linear scaling at larger w_p and eventually diverge for $w_p \to \infty$. This expectation seems to be supported by the data of Kondic and Bertozzi (Fig. 3c and especially Fig. 4 of Ref. [13]). Their scaling at small w_p is also consistent with our result. The data of Ye and Chang who found the amount of transient amplification to increase with decreasing longitudinal wave number k of initial plane wave disturbances (Fig. 6a of Ref. [14]) are also consistent with our predictions: initial disturbances with smaller k are closer to the optimal disturbance $f_0 =$ const. and thus are amplified stronger.

Perhaps the only qualitative difference between our results and those of other studies is the q-dependence of transient amplification in the precursor model. The data of Bertozzi and Brenner [6] and Ye and Chang [14] show the amount of transient amplification to depend rather strongly on the transverse wave number. For instance, Ref. [6] produces $\gamma_\infty(0.6) \approx 800$ compared with $\gamma_\infty(0) \approx 200$ (for $b = 0.001$ and $D = 5$) – a factor of four difference. It is possible that this difference is due to the specific choice of initial disturbances (recall, we compute $\gamma_p(q, t)$ as the maximum over *all* initial disturbances, while Refs. [6,14] considered only longitudinally modulated disturbances (9) and (11)). The calculation of Kondic and Bertozzi [13], on the other hand, shows very weak q-dependence of transient amplification for initial disturbances (10) with no longitudinal modulation. For instance, for a disturbance of width $w_p = 20$ they get $\gamma_\infty(0.5) \approx 75$ compared with $\gamma_\infty(0) \approx 60$ (for $b = 0.01$ and $D = 5$).

6. Discussion

Summing up, we find that much of the reported disagreement between the predictions of linear and non-linear theory regarding transient amplification of spontaneous disturbances is due to a rather arbitrary way in which the amplification itself is defined. For instance, direct comparison requires that the same norm be used (e.g., studies based on non-linear theory [6,13] used the ∞-norm, while those based on the linear theory [15] used the 2-norm). Furthermore, the predictions obtained for particular initial disturbances [6,13,14] cannot be directly compared with the predictions based on the optimal initial disturbances [15], as the former could be highly non-optimal. Finally, this dependence on the choice of initial disturbances implies that the size of the computational domain has to be at least as large as the spatial extent of the class of initial disturbances under consideration.

Nevertheless, we find reasonable quantitative agreement between the predictions of linear and non-linear theory based on the precursor model, provided that similar classes of initial disturbances are used. The non-linear effects appear to be relatively mild and do not qualitatively alter the predictions of the linear theory. The only qualitative difference between the predictions of different studies, the origin of which we have not been able to determine conclusively, is the detailed dependence of transient amplification on the transverse wave number of the initial disturbance. Further studies using both linear and non-linear theory are needed to

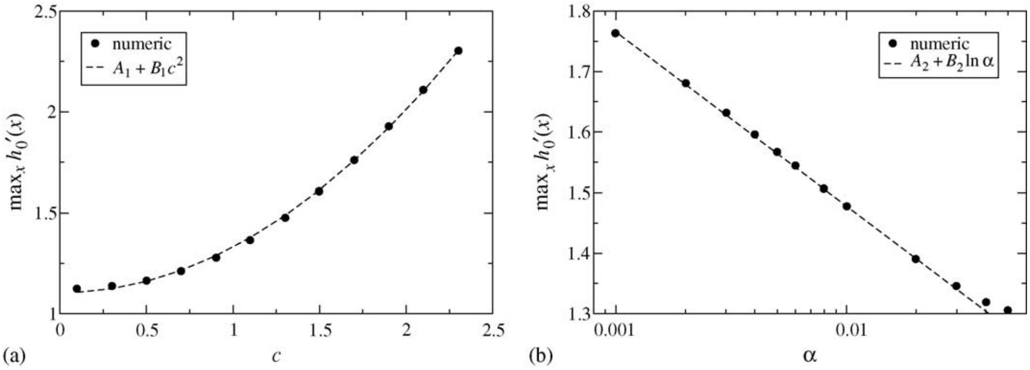

Fig. 9. The maximal slope of the capillary shock (a) as a function of the contact angle for $\alpha = 0.01$ and $D = 5$ and (b) as a function of the slip parameter for $c = 1.303$ and $D = 5$. A_i and B_i are constants determined from a least squares fit.

determine whether the maximum is indeed achieved at finite wave numbers and, if yes, why.

We find significant disagreement between the predictions of the precursor model and the slip model. In a nutshell, the largest transient amplification in the precursor model is produced by disturbances localized in the precursor film due to the extreme sensitivity of the capillary shock to the microscopic structure of the precursor. This sensitivity has been artificially eliminated in the slip model considered by Davis and Troian [15]. In order to incorporate the effect of microscopic structure of the liquid–solid interaction on the dynamics of the contact line in the slip model one needs to consider the response of the capillary shock to: (i) the disturbances in the contact angle and (ii) the disturbances in the slip coefficient α. Only (i) was considered in Ref. [15]. As is well known [3], the shape of the capillary ridge depends relatively weakly on the slope parameter c (also see Fig. 9a). As a result, one should only expect moderate transient amplification of disturbances in the contact angle:

$$\gamma_\infty \propto \frac{\partial \max_x h_0'(x)}{\partial c} \propto c, \tag{23}$$

in agreement with our analytic result (18) and consistent with the numerical results of Davis and Troian [2]. On the other hand, the dependence on the slip parameter α is quite sensitive, just like the dependence on the parameter b in the precursor model. We find this dependence to be logarithmic (see Fig. 9b), in agreement with the analytical predictions [7]. We can therefore expect disturbances in the slip parameter to be amplified much stronger:

$$\gamma_\infty \propto \frac{\partial \max_x h_0'(x)}{\partial \alpha} \propto \alpha^{-1}, \tag{24}$$

i.e., by many orders of magnitude for realistic values of α. Numerical evidence supporting this conclusion has been obtained by Hoffmann et al. [21]. Therefore, to match the predictions of the precursor model, the slip model has to be reformulated for *parametric* disturbances. As it stands, the slip model does not adequately describe transient dynamics.

As we have shown, the linear theory (and hence the non-linear theory, too) can produce arbitrarily large transient amplification. For films spreading on a dry plane strong transient amplification can be obtained by imposing initial disturbances which correspond to a very slow transverse modulation of the film thickness in a wide strip immediately behind the contact line. An even stronger transient amplification can be produced for spreading on a prewetted plane, if a similar thickness disturbance is imposed in the precursor film just ahead of the contact line. Practically achievable transient amplification is only limited by the width of the strip and the modulation period, which can be increased by increasing the system size. Of course, the disturbances that are realized in typical (rather than specially designed) experimental conditions will likely be highly non–optimal and as a result will be amplified much less than the optimal ones. On the other hand, transient amplification should also become very large in the limit of small precursor thickness (or small slip coefficient), in which case even non-optimal disturbances should be amplified very strongly leading to breakdown of the linear stability analysis. The key questions, therefore, are: (i) to what degree the naturally occurring disturbances are optimal; (ii) whether they can be considered small, that is whether the linear or non-linear theory should be used and (iii) whether the limit of very small precursor thickness (or slip coefficient) is physical.

A targeted experimental investigation would help answer these questions. As Garnier et al. have shown recently [22], desired thickness disturbances can be imposed dynamically on the spreading film using the thermocapillary effect. The idea is to use the dependence of surface tension on the local interfacial temperature to drive the fluid from warmer to cooler regions, thus changing the local thickness. The temperature field can be conveniently controlled optically by illuminating the film with intensity-modulated visible or infrared radiation. The amount of transient amplification for disturbances initially localized behind the contact line can be determined experimentally by modulating the film thickness in the transverse direction in a strip of certain width and studying the resulting distortion of the contact line. Disturbances ahead of the contact line can be imposed more conveniently via chemical patterning of the substrate [23] to change the local contact angle/slip constant for spreading on a dry plane or the precursor thickness for spreading on a prewetted plane. A similar effect can be achieved by etching the surface of the substrate, imposing microscopic surface roughness [24].

Finally, we should note that although the scope of our discussion was limited to gravity-driven spreading, the same basic conclusions should apply to films driven by centrifugal forces (as in spin coating) or temperature gradients.

References

[1] H. Huppert, Flow and instability of a viscous current down a slope, Nature 300 (1982) 427.

[2] S.M. Troian, E. Herbolzheimer, S.A. Safran, J.F. Joanny, Fingering instabilities of driven spreading films, Europhys. Lett. 10 (1989) 25.

[3] M.A. Spaid, G.M. Homsy, Stability of Newtonian and viscoelastic dynamic contact lines, Phys. Fluids 8 (1996) 460.

[4] J.R. de Bruyn, Growth of fingers at a driven three-phase contact line, Phys. Rev. A 46 (1992) R4500.

[5] J.M. Jarrett, J.R. de Bruyn, Fingering instability of a gravitationally driven contact line, Phys. Fluids A 4 (1992) 234.

[6] A.L. Bertozzi, M.P. Brenner, Linear stability and transient growth in driven contact lines, Phys. Fluids 9 (1997) 530.

[7] A. Münch, B.A. Wagner, Numerical and asymptotic results on the linear stability of a thin film spreading down a slope of small inclination, Eur. J. Appl. Math. 10 (1999) 297.

[8] L. Boberg, U. Brosa, Onset of turbulence in a pipe, Z. Naturforsch A 43 (1988) 697.

[9] L.N. Trefethen, A.E. Trefethen, S.C. Reddy, T.A. Driscoll, Hydrodynamic stability without eigenvalues, Science 261 (1993) 578.

[10] R.O. Grigoriev, Contact line instability and pattern selection in thermally driven liquid films, Phys. Fluids 15 (2003) 1363.

[11] J.M. Davis, S. Troian, Influence of attractive van der Waals interactions on the optimal excitations in thermocapillary-driven spreading, Phys. Rev. E 67 (2003) 016308.

[12] B.F. Farrell, P.J. Ioannou, Generalized stability theory part I: autonomous operators, J. Atmos. Sci. 53 (1996) 2025.

[13] L. Kondic, A.L. Bertozzi, Nonlinear dynamics and transient growth of driven contact lines, Phys. Fluids 11 (1999) 3560.

[14] Y. Ye, H.-C. Chang, A spectral theory for fingering on a prewetted plane, Phys. Fluids 11 (1999) 2494.

[15] J.M. Davis, S. Troian, On a generalized approach to the linear stability of spatially nonuniform thin film flows, Phys. Fluids 15 (2003) 1344.

[16] S. Kalliadasis, Nonlinear instability of a contact line driven by gravity, J. Fluid Mech. 413 (2000) 355.

[17] E.O. Tuck, L.W. Schwartz, A numerical and asymptotic study of some third-order ordinary differential equations relevant to draining and coating flows, SIAM Rev. 32 (1990) 453.

[18] A.L. Bertozzi, private communication.

[19] E.B. Dussan V and S. Davis, "On the motion of a fluid fluid interface along a solid surface", J. Fluid Mech. 65, 71 (1974).

[20] P.G. de Gennes, Wetting: statics and dynamics, Rev. Mod. Phys. 57 (1985) 827.

[21] K.-H. Hoffmann, B. Wagner and A. Münch, "On the generation and spreading of finger instabilities in film coating processes", Lect. Notes Comp. Sci. Eng. 8 (Springer-Verlag, 1999).

[22] N. Garnier, R.O. Grigoriev, M.F. Schatz, Optical manipulation of microscale fluid flow, Phys. Rev. Lett. 91 (2003) 054501.

[23] D.E. Kataoka, S.M. Troian, Patterning liquid flow on the microscopic scale, Nature 402 (1999) 794.

[24] L.E. Stillwagon, R.G. Larson, Leveling of thin films over uneven substrates during spin coating, Phys. Fluids A 2 (1990).