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A. Münch and A. L. Bertozzi

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Rarefaction–undercompressive fronts in driven films

A. Münch

Technical University, Munich, Arcisstraße 21, 80333 Munich, Germany

A. L. Bertozzi

Departments of Mathematics and Physics, Duke University, Durham, North Carolina 27708

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We consider experiments by Ludviksson and Lightfoot [AIChE J. **17**, 1166 (1971)] on thin liquid films driven up a vertical plate by a thermally induced surface tension gradient with a counteracting gravitational force, and revisit their theoretical analysis, which neglects the effects of curvature, for predicting the climbing rate of the front. We present a new theory for the lubrication model with curvature effects, and get rising rates that depend on the microscopic length scale at the contact line. The predictions are, in general, in better agreement with the experiment. © 1999 American Institute of Physics. [S1070-6631(99)01310-0]

In a series of experiments¹ in 1971, Ludviksson and Lightfoot (LL) studied thin liquid films driven up a plate by a thermal gradient, with a counteracting gravitational force. Using a simplified model that omitted the effects of curvature, they predicted the climbing rate of the rising front. Their analysis explains qualitative features of the experiments, but always overestimates the climbing rates, in one case by up to 40%. They suggest that in the extreme cases, surface effects might play a role.

A subsequent paper of Teletzke *et al.*² (see pp. 66–72) notes these discrepancies and attempts to understand the dynamics via an extended lubrication model that includes the additional pressure contribution from surface tension, as well as other effects. Via an analysis of traveling wave solutions, they conclude that the revised theory does not explain the experimental dynamics. A recent paper of Kataoka and Troian³ revisits the Teletzke approach and, with some corrections to the numerical method, finds good agreement between traveling wave theory and the shape of the front profiles in the original LL experiment. In their analysis they use the LL experimental climbing film thickness as a fitting parameter to the numerical computations of the theoretical profile.

Via a new analysis of the full lubrication model,⁴ we show that the actual dynamical behavior is quite different from that of the model in Ref. 1. Instead of a classical ‘‘rarefaction-shock’’ proposed by LL to describe the large-scale structure of the film, we show that curvature effects cause a ‘‘rarefaction-undercompressive shock’’ to form, with a lower climbing rate and larger thickness of the film front. The deviation from classical theory depends strongly on the microscopic length scale at the contact line. We make new predictions for rise rates in Marangoni-driven films.

LL introduces the model

$$h_t + [F(h)]_x = \frac{(h^3 p_x)_x}{3\mu}, \quad F(h) := \frac{\gamma h^2}{2\mu} - \frac{\rho g h^3}{3\mu}, \quad (1)$$

to describe the evolution of fluid profile $h(x, t)$ at a position x in the downstream direction (i.e., away from the bath). The

parameters ρ , μ , and γ denote the liquid density, viscosity, and the thermally induced surface tension gradient; g is the gravitational acceleration. The pressure p becomes important in regions of strong curvature. Neglecting curvature, they drop p from the above equations to get the *conservation law*,

$$h_t + [F(h)]_x = 0. \quad (2)$$

The initial value problem with $h(x, 0) = h_0(x)$ can be solved implicitly using the method of characteristics: $h(x, t) = h_0[x - F'(h)t]$, which, for jump initial data,

$$h_0(x) = \begin{cases} h_\infty, & \text{if } x < 0, \\ 0, & \text{if } x \geq 0, \end{cases}$$

results in an unphysical multivalued film height after a finite time $t > 0$. In a common procedure in other applications of conservation laws (e.g., gasdynamics), LL replaced this result with a classical (centered) rarefaction-shock solution (see the left column, p. 1168, in Ref. 1),

$$h(x, t) = \begin{cases} h_\infty, & \text{if } x \leq F'(h_\infty)t \\ (F')^{-1}(x/t), & \text{if } F'(h_\infty)t \leq x \leq F'(h_m)t, \\ 0, & \text{if } F'(h_m)t < x. \end{cases} \quad (3)$$

Note that (3) consists of an expanding rarefaction wave, and of a jump discontinuity, or shock, from h_m to 0 traveling at speed $s(h_m, 0; F)$ given by the Rankine–Hugoniot condition,

$$s(h_-, h_+; F) = \frac{F(h_+) - F(h_-)}{h_+ - h_-}. \quad (4)$$

The left and right edges of the rarefaction wave are of height h_∞ and h_m and travel with characteristic speed $F'(h_\infty)$ and $F'(h_m)$, respectively. Nonseparation of the rarefaction wave and the leading shock means $s(h_m, 0; F) = F'(h_m)$, equivalently, $h_m = 3\gamma/(4\rho g)$. A graph of (3) is shown for $t = 1000$ by the dashed line in Fig. 1, after rescaling (3) with the scalings of the next section. This is the solution that arises with second-order diffusion. For comparison we also

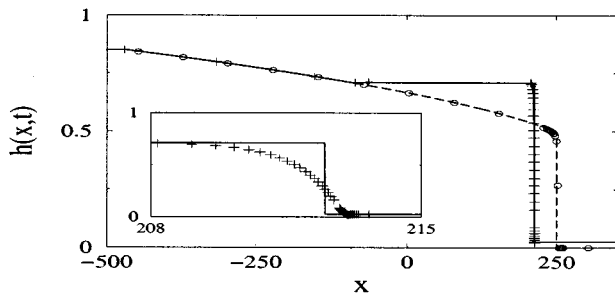


FIG. 1. Comparison of the classical rarefaction-shock solution (dashed line) and the new rarefaction-undercompressive shock [(8), solid line] with the solution (+) of the initial value problem for the full PDE (6) at $t=1000$. A close-up of the leading front is shown in the inset. Here $h_\infty=0.85$ and $b=0.025$. Circles correspond to the solution of $h_t+[f(h)]_x=\epsilon h_{xx}$, $\epsilon=0.05$, with no precursor.

include the solution of the PDE obtained by introducing a small amount of second-order diffusion; it is denoted by circles in the figure.

Though there is some qualitative agreement with experiments, in the sense that LL did observe a sharp leading edge and, for the initial stages, a smooth draining portion of the solution, some discrepancies are noticeable. The theoretical predictions always exceed the experimental rising rates, by up to 40%. The experimental front profiles shown in Fig. 10 in their article are nearly horizontal less than 2 mm away from the front, and appear to have a constant shape after some hours of spreading. This is inconsistent with nonseparating rarefaction-shock solutions. Below, we show that the inclusion of curvature leads to a different solution for the conservation law (2) than (3).

Including the effect of surface tension and hence curvature,^{2–5} we replace (2) with

$$h_t+[F(h)]_x=-\frac{\sigma}{3\mu}(h^3 h_{xxx})_x, \tag{5}$$

where σ denotes the surface tension coefficient. As in Refs. 2, 3, we assume a small precursor layer of microscopic thickness b ahead of the film. This makes sense for a completely wetting fluid,⁶ and LL suggest that a precursor layer of less than $0.1 \mu\text{m}$ indeed is present in their experiment.

Rescaling h , x , and t with $H=3\gamma/(2\rho g)$, $l=[3\sigma\gamma/(2\rho^2g^2)]^{1/3}$, and $T=2\mu/3(12\sigma\gamma\rho g)^{1/3}/\gamma^2$, we obtain

$$h_t+[f(h)]_x=-(h^3 h_{xxx})_x, \quad f(h):=h^2-h^3. \tag{6}$$

Note that here, b and h_∞ have been rescaled with respect to the aforementioned scalings. Using the values for the temperature gradient, etc. reported by LL to calculate the dimensionless precursor height for their experiment, we find a dimensionless $b<0.06$.

We integrated the PDE (6) numerically with a finite difference scheme,⁷ using smoothed jump initial data, $h(x,0)=[h_\infty+b-\tanh(x)(h_\infty-b)]/2$, where $h_\infty=0.85$, and $b=0.025$. The profile at $t=1000$ is marked by a “+” in Fig. 1; in addition to a rarefaction wave and a leading front, we

see the appearance of a very flat plateau of height $h_{uc}(b)$, separating the two. The corresponding discontinuous solution of the conservation law,

$$h_t+f(h)_x=0, \tag{7}$$

is a rarefaction-undercompressive shock, shown in Fig. 1 by a solid line, given by

$$h(x,t)=\begin{cases} h_\infty, & \text{if } x\leq f'(h_\infty)t, \\ (f')^{-1}(x/t), & \text{if } f'(h_\infty)t\leq x\leq f'(h_{uc})t, \\ h_{uc}, & \text{if } f'(h_{uc})t<x\leq s(h_{uc},b;f)t, \\ b, & \text{if } s(h_{uc},b;f)t<x, \end{cases} \tag{8}$$

with a plateau extending between $f'(h_{uc})t$ and $s(h_{uc},b;f)t$. The discontinuous solution and the smooth profile agree very well on a large scale, with the major deviations near the edges of the rarefaction wave and at the leading front. There, curvature smoothes out the sharp discontinuity over a length of $O(1)$; see the inset. This picture persists in time, hence the smoothed front travels at the same speed $s_{uc}(b)=s(h_{uc},b;f)$ as the discontinuous shock, given by the Rankine–Hugoniot condition (4). The separation indicates that the shock velocity exceeds the characteristic velocity for the left state of the shock, $s(h_{uc},b;f)-f'(h_{uc})>0$. Shocks for which characteristics emanate from the shock trajectory are called *undercompressive shocks*.

An extensive study in Ref. 4 found that undercompressive shocks are the typical leading front in long-time solutions of (6), as long as h_∞ is above a certain threshold $h_2(b)$. The undercompressive shock is followed by a rarefaction wave for $h_\infty>h_{uc}$, or a second, slower shock, if $h_2<h_\infty<h_{uc}$. For the latter case, the signature of such a double shock has been found in very recent experiments.⁸ For $h_\infty<h_2$, the solution to (6) evolves into a simple compressive traveling wave with farfield states h_∞ and b . In this case, one finds that surface tension induces a capillary ridge. Traveling waves with capillary ridges have been observed for films significantly thinner than those in the LL experiments.^{9,5} Characteristically, capillary ridges do not appear in the experimental profiles reported by LL. The unusual “undercompressive” shock dynamics is due to the fourth-order structure of the diffusive term on the right-hand side of (6).

The new solution (8) is slower than that of LL (13), which explains the deviations found by LL when they compared their predictions with the experimental observations. We computed the precursor heights for which $s_{uc}(b)$ matched the experimental rising rates, and found them to be consistent with the previously mentioned range for b . Also note that LL observe marked nonuniformities in the climbing rate, which is in accordance with the sensitivity of b to microscopic changes at the contact line.

Our computations show that the undercompressive front speed s_{uc} decreases with b . This raises a new question, since the speed of the front in turn affects the height of the precursor that can develop:⁶ the faster the front advances, the smaller b . It would be interesting to know whether equilibration of these two effects can play a role in selecting a specific s_{uc} .

TABLE I. Dimensionless heights and rising rates of the undercompressive profile, for different precursor thicknesses. The LL theory predicted a dimensionless rise rate of 0.25 for small b .

b	h_{uc}	$s(h_{uc}, b; f)$
0.0001	0.8946	0.0943
0.001	0.8542	0.1247
0.01	0.7724	0.1780
0.05	0.6528	0.2415
0.1	0.5679	0.2786

Another interesting observation by LL is that, after the film has spread for several hours, its shape remains constant and appears in their Fig. 10 to be very flat. In our numerical simulations we found, in fact, that for $b < 0.044$, $h_{uc}(b) > \frac{2}{3}$, where f has its maximum, implying a negative characteristic speed for the leading edge of the rarefaction wave. Hence the edge moves backward in the laboratory frame of reference, while the front continues to advance, consistent with the observation of LL. It would be interesting to observe this dynamical behavior in an experiment, possibly using a controlled prewet surface.

Experimentalists may therefore consider further studies in the LL regime of film thicknesses to allow observation of the full profile, including the rarefaction wave and its ongoing separation from the leading undercompressive front, while reducing uncertainties that complicate the comparison with theory. For example, they could choose liquids with a less temperature-dependent viscosity than squalane, and

maintain higher precision in the surface tension gradient. For future comparisons, we include Table I, which shows the theoretical predictions for h_{uc} and s for three orders of magnitude in b .

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