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Symbolic Computation as a Tool for High-Order Long-Wave Stability Analysis of Thin Film Flows with Coupled Transport Processes

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Many fundamental studies based on the evolution equations derived by long-wave approximation have contributed to the fact that the dynamics of a thin film flowing down an inclined plane is now one of the best-understood problems of hydrodynamic stability. In most engineering applications however, the stability behaviour of the film flow is modified by complex coupled transport processes, and because of the huge amount of algebra needed to derive the evolution equations in these cases, an investigation by numerical methods is often preferred by engineers. In this paper, we illustrate how computer algebra techniqes can be used to derive and analyse long-wave evolution equations even for very complex situations automatically, thus making the advantages of symbolic solutions available for such applications. Using these methods, higher-order approximations can also be obtained automatically. These are of interest since they can provide heuristic estimates for—and extensions of—the range of validity of the long-wave approximation.

Key Words: numerical analysis; automated algorithms; hydrodynamic stability; nonlinear effects.

1. INTRODUCTION

Since the experiments of the Kapitza [1] in the forties, the interfacial wave patterns developing on a thin liquid film flowing down an inclined plane have been investigated by many researchers as a fascinating example for complex nonlinear dynamics (see the reviews by Chang [2] and Lin and Wang [3]). In addition to this fundamental interest, the understanding of the stability behaviour of thin liquid films is of great importance for many industrial applications like coating and drying, heat exchangers, or chemical reactors. In

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most of these applications the stability behaviour is modified by the interaction of heat and mass transport processes with the flow.

For low Reynolds number flows, the long-wave approximation proposed by Benney [4] turned out to be a very successful model which captures much of the nature of transitional flow of thin films. It is based on the expectation that the wavelength of unstable interfacial disturbances of uniform film flow must be large compared to the film thickness because of the stabilizing effect of surface tension for short waves. Thus—by an asymptotic analysis in the limit $\alpha \to 0$, with α being the dimensionless wavenumber—a single nonlinear evolution equation for the shape of the free surface of the film can be derived from the Navier–Stokes equations (and any transport equations coupled with them). The technical effort of a qualitative analysis is reduced dramatically by considering this evolution equation rather than the original system, such that simple formulas describing the dependence of the linear stability of uniform film flow on the parameters can be found. Moreover, the primary bifurcation of unstable uniform film flow into permanent wave trains can be predicted readily by weakly nonlinear stability analysis (see, e.g., Benney [4], Lin [5], and Gjevik [6]). More complex dynamical phenomena such as solitary waves (see, e.g., Pumir *et al.* [7]) or the stability of two-dimensional permanent waves with respect to three-dimensional disturbances (see Joo *et al.* [8]) were also studied using the long-wave approximation.

Usually the asymptotic expansion is truncated at first order in α assuming a strong surface tension influence. Lin [5] investigated the second-order evolution equation which allows for consistent prediction of the dispersion of the linear and nonlinear waves. For the case of weak surface tension, Nakaya [9] and Chang [10] considered the third-order evolution equation.

The long-wave approximation was also applied successfully to investigate film flows coupled with complex transport processes such as evaporation and condensation (see Burelbach *et al.* [11] and Joo *et al.* [12]), which have important applications in engineering.

For a more complete outline of the fundamental stability results obtained by the long-wave approximation we refer to Chang [2] and Lin and Wang [3]. In the present paper, we are concerned with the application of this well-developed theory to engineering problems. From that point of view, the following two issues are addressed.

First, many practical problems involve heat and mass transfer, or even chemical reactions. Since the stability behaviour of such systems may depend on many parameters, the long-wave theory has the advantage that the dependence of the relevant stability results on the parameters can be obtained directly as formulas rather than by extensive parameter studies, which would be necessary using numerical methods. However, even the derivation of first-order evolution equations can already be a very tedious and error-prone task for problems involving transport processes, and it is practically impossible to derive higher orders of the long-wave approximations manually. Fortunately, all the algebraic manipulations necessary for the derivation of evolution equations can be formulated as a standardized algorithm and implemented in a computer algebra system, as, for example, MAPLE [13]. After specifying the general form of the governing equations and a model problem in Section 2 of this paper, we illustrate such an implementation in Section 3, which automatically computes higher-order evolution equations for film flow coupled with an arbitrary number and arbitrary types of transport processes.

This symbolic algorithm extends the basic ideas as given by Atherton and Homsy [14], who used an early version of the computer algebra system REDUCE in the mid-seventies to calculate the evolution equation for axially symmetric and three-dimensional isothermal

film flow. It should be pointed out, however, that the second-order approximation stated in their paper contains errors.

A second issue of practical importance is the fact that the range of validity of the first-order approximations may be very limited, as is indicated by comparison with experimental stability results and comparisons with numerical solutions of the Orr–Sommerfeld equations (see, e.g., Krantz and Owens [15], Liu *et al.* [16]). Theoretically, the long-wave approximation should be valid for small wavenumbers and small Reynolds numbers, but there is no quantitative a priori estimate for this range of validity. Such an estimate would be very useful in order to assess the suitability of the approximation for a specific application.

Since many of the methods of linear and weakly nonlinear analysis can also be fully automatized by computer algebra, it is not only possible to derive, but also to analyse the higher-order evolution equations. In Section 4, we perform a linear stability analysis for the fifth-order equation and illustrate how the comparison of the results of different order can be used as an indicator for the range of validity of the long-wave approximation. Using Padé-approximation, we show that the information obtained at fifth order is even sufficient to predict the published experimental data for shorter wavelengths and relatively high Reynolds numbers remarkably well.

Using Landau theory as an simple example for a weakly nonlinear analysis, we illustrate in Section 5 that computer algebra methods also provide automatic modeling of the primary bifurcation of the uniform flow into permanent waves, even if complex transport processes are involved. Similar to the question of the range of validity in α already discussed in connection with the linear analysis, it is useful to know how far away from the bifurcation point the weakly nonlinear analysis will still be valid. It is illustrated that the comparison of the results obtained using different orders of the long-wave approximation provides here a heuristic criterion, too.

2. GENERAL FORM OF THE GOVERNING EQUATIONS AND A MODEL PROBLEM

The governing equations for most problems involving a film flow with several (N) coupled transport processes can be written in the dimensionless form

$$\nabla \cdot \mathbf{u} = 0, \tag{1}$$

$$Re(\mathbf{u}_t + (\mathbf{u} \cdot \nabla)\mathbf{u}) = -\nabla p + \Delta \mathbf{u} + \mathbf{G}, \tag{2}$$

$$Re\, Sc_i((C_i)_t + (\mathbf{u} \cdot \nabla)C_i) = \Delta C_i + F_i \qquad (i = 1 \ldots N). \tag{3}$$

The velocity field $\mathbf{u} = (u, v, w)$ is scaled with a characteristic velocity U, which may depend on the specific problem under consideration. The spatial coordinates (x, y, z) are scaled with the unperturbed film thickness H, and the time coordinate t is scaled by H/U. The transport quantities C_i are also scaled by problem-dependent characteristic values Γ_i. Thus, $Re = \rho_0 U H/\mu$ denotes the Reynolds number, with ρ_0 being a characteristic value of the density and μ being the dynamic viscosity of the liquid, and the $Sc_i = \nu/D_i$ denote Schmidt (or Prandtl) numbers with D_i being the diffusivities of the transport processes.

The source term $\mathbf{G}$ in Eq. (2) models the gravitational force and is usually constant; however, it may also account for bouyancy effects caused by variations in the transport quantities. The source terms F_i in the transport equations (3) may model chemical reactions.

These source terms are scaled by

$$\mathbf{G} = \frac{H^2}{\mu U}\mathbf{G}^* \qquad \text{and} \qquad F_i = \frac{H^2}{D_i \Gamma_i} F_i^*. \tag{4}$$

Here the terms with the asterisk denote the dimensional quantities. The velocity field must satisfy the no-slip conditions ($\mathbf{u} = 0$) at the wall ($y = 0$), as well as the kinematic and stress boundary conditions at the free surface ($y = h(t, x, z)$):

$$\left.\begin{aligned} h_t + u h_x &= v \\ (\mathbf{n})^T \cdot T \cdot \mathbf{n} &= K \\ (\mathbf{t})^T \cdot T \cdot \mathbf{n} &= L \end{aligned}\right\} \qquad \text{for } y = h(t, x, z). \tag{5}$$

Here, $T = -p \cdot I + (\nabla \mathbf{u})^T + (\nabla \mathbf{u})$ denotes the dimensionless stress tensor and $\mathbf{t}(\mathbf{n})$ denotes the unit tangential (normal) vectors on the free surface. The right-hand sides of the normal and tangential stress conditions may represent models for different surface forces and are scaled by

$$K = \frac{H}{\mu U} K^* \qquad \text{and} \qquad L = \frac{H}{\mu U} L^*. \tag{6}$$

Most of the physically relevant boundary conditions for the transport equations can be modeled by operators of the form

$$\begin{aligned} B_w(C_i) &= 0 \qquad \text{for } y = 0, \\ B_s(C_i, \mathbf{u}) &= 0 \qquad \text{for } y = h(t, x, z). \end{aligned} \tag{7}$$

As an example for a transport process coupled with the film flow, we will assume throughout this paper that a species is absorbed from the ambient air into the film, where it is consumed by a first-order chemical reaction (see Fig. 1). While the density ρ is considered to be independent of the concentration C_1^* of the species, the dependence of the surface tension on C_1^* is modeled by

$$\sigma = \sigma_\infty - \sigma_c (C_1^* - C_\infty^*), \tag{8}$$

where C_∞^* is the concentration of the species in the ambient air. For this example, the dimensional source terms read $\mathbf{G}^* = (\rho g \sin\beta, -\rho g \cos\beta, 0)^T$ and $F_1^* = -k_r C_1^*$ with g denoting gravitational acceleration, β the inclination angle, and k_r the velocity of the first order reaction. Thus, if we take the concentration in the ambient air as the characteristic scale ($\Gamma_1 = C_\infty^*$) and choose the surface velocity ($U = \rho g \sin\beta H^2/2\mu$) of the film as the scale for the velocities, the dimensionless source terms in Eqs. (2) and (3) read for our example problem

$$\mathbf{G} = 2(1, -\cot\beta, 0)^T \qquad \text{and} \qquad F_1 = -\gamma^2 C_1 \tag{9}$$

with $\gamma^2 = H^2 k_r / D_1$ being a dimensionless measure of the reaction velocity. The right-hand sides of the stress conditions are given by

$$L = 2M \cdot (\mathbf{t} \cdot \nabla C_1) \qquad \text{and} \qquad K = -2(\nabla \cdot \mathbf{n}) \cdot (S - M(C_1 - 1)) \tag{10}$$

with $S = \sigma_\infty / (\rho g \sin\beta H^2)$ being the Weber number and $M = \sigma_c C_\infty^* / (\rho g \sin\beta H^2)$ being

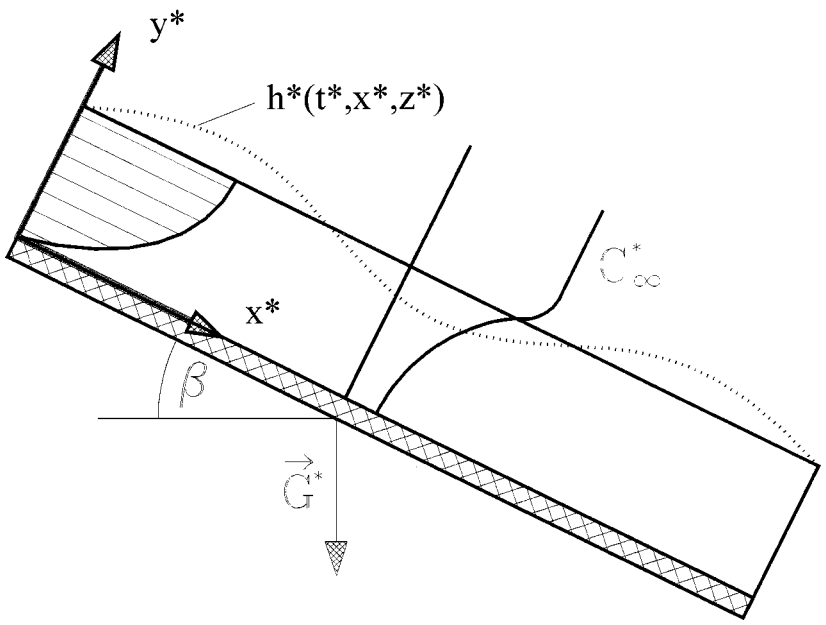

FIG. 1. Consumption by first-order chemical reaction of a species absorbed into a thin film flowing down an inclined plane.

the Marangoni number. Finally we assume that, while there is no mass transfer through the wall, the mass transfer through the surface can be described by a mass transfer coefficient h_c. Hence the boundary operators for the mass transport equation are given by

$$B_w(C_1) = \frac{\partial C_1}{\partial y} = 0 \qquad \text{and} \qquad B_s(C_1, \mathbf{u}) = \mathbf{n} \cdot \nabla C_1 - Sh(1 - C_1) = 0. \tag{11}$$

Here, $Sh = h_c H/D$ is the Sherwood number.

3. OUTLINE OF THE SYMBOLIC ALGORITHM

In the two-dimensional case ($\partial/\partial z = 0$), it is convenient to introduce a stream function ψ by $(u, v) = (\psi_y, -\psi_x)$. Equations (1)–(3) can then be reduced to

$$Re\Delta\psi_t + [\psi, \Delta\psi] = \Delta\psi + \mathrm{rot}_3\mathbf{G}, \tag{12}$$

$$Re\, Sc_i(C_i)_t + [\psi, C_i] = \Delta C_i + F_i \qquad (i = 1 \ldots N). \tag{13}$$

where we used the notations $[f, g] := f_y g_x - f_x g_y$ and $\mathrm{rot}_3 := (G_1)_y - (G_2)_x$.

The no-slip conditions at $y = 0$ yield in terms of the stream function

$$\psi = 0 \qquad \text{and} \qquad \psi_y = 0. \tag{14}$$

The kinematic condition at the free surface can be written in the following "conservation form" (using the notation $\psi(h) := \psi(t, x, h(t, x))$ and the chain rule $\psi_x(h) + \psi_y(h) \cdot h_x = [\psi(h)]_x$):

$$h_t + [\psi(h)]_x = 0 \qquad \text{for } y = h(t, x), \tag{15}$$

The following conditions can be derived from the stress conditions at $y = h(t, x)$:

$$-p - 2\frac{1+h_x^2}{1-h_x^2}\psi_{xy} - K + 2\frac{h_x}{1-h_x^2}L = 0, \tag{16}$$

$$\psi_{yy} - \psi_{xx} - \left(1-h_x^2\right)^{-1}\left(4h_x\psi_{xy} - \left(1+h_x^2\right)L\right) = 0. \tag{17}$$

The pressure in condition (16) can be eliminated by replacing that condition by its tangential derivative and then using the momentum equations

$$p_x = \psi_{xxy} + \psi_{yyy} - Re(\psi_{yt} + [\psi, \psi_y]) + G_1 \tag{18}$$

$$p_y = -\psi_{xxx} - \psi_{xyy} - Re(\psi_{xt} + [\psi, \psi_x]) + G_2 \tag{19}$$

to eliminate p_x and p_y in the new condition (see Atherton and Homsy [14]). Note that this means that the highest derivative with respect to y in the new condition will be ψ_{yyy}. Also note that all these manipulations of the equations can easily be done automatically by a computer algebra system.

The crucial step in the long-wave approximation is to take a large wavelength as a characteristic length scale for the x-direction rather than the film thickness H which is only an appropriate length scale for the y-direction. This suggests a rescaling of the independent variables,

$$t \to \alpha^{-1}\tau, \qquad x \to \alpha^{-1}\xi, \qquad y \to \eta \tag{20}$$

with $\alpha \ll 1$ being the dimensionless wavenumber. Moreover, we assume a "strong" surface tension, i.e., $S = \bar{S}\alpha^{-2}$ with $\bar{S} = O(1)$. Now we substitute ψ and C_i by asymptotic series in α

$$\psi = \psi_0 + \psi_1\alpha + \psi_2\alpha^2 + \cdots \tag{21}$$

$$C_i = C_{i,0} + C_{i,1}\alpha + C_{i,2}\alpha^2 + \cdots \tag{22}$$

and by equating like powers of α—a standard operation of computer algebra systems—we obtain a hierarchy of equations. For our example, this hierarchy can be written in the form

$$\psi_{n,\eta\eta\eta\eta} = A_n(\psi_0, \ldots, \psi_{n-1}, C_{1,0} \ldots C_{1,n-1}), \qquad n = 0, 1, 2 \ldots \tag{23}$$

$$C_{1,n,\eta\eta} - \gamma^2 C_{1,n} = B_n(\psi_0, \ldots, \psi_{n-1}, C_{1,0} \ldots C_{1,n-1}), \qquad n = 0, 1, 2 \ldots. \tag{24}$$

Because the boundary conditions (14) and (11) for ψ and C_1 are linear, all orders ψ_n and $C_{1,n}$ have to satisfy them. The (nonlinear) stress conditions (16), (17) yield the following conditions for ψ_n at the surface $\eta = h(\tau, \xi)$:

$$\psi_{n,\eta\eta\eta}(\tau, \xi, h(\tau, \xi)) = a_n(\psi_0, \ldots, \psi_{n-1}, C_{1,0} \ldots C_{1,n-1}), \qquad n = 0, 1, 2 \ldots \tag{25}$$

$$\psi_{n,\eta\eta}(\tau, \xi, h(\tau, \xi)) = b_n(\psi_0, \ldots, \psi_{n-1}, C_{1,0} \ldots C_{1,n-1}), \qquad n = 0, 1, 2 \ldots. \tag{26}$$

For $n = 0$, the right-hand sides of Eqs. (23), (24), and (26) are zero, while the right-hand side of Eq. (25) is given by $a_0 = -G_1 = -2$. Thus ψ_0 is a cubic polynomial in η and $C_{1,0}$

can be found by integrating the homogenous linear differential equation given by (24). This yields

$$\psi_0 = h\eta^2 - \frac{1}{3}\eta^3, \tag{27}$$

$$C_{1,0} = \frac{Sh\cosh(\gamma\eta)}{Sh\cosh(\gamma h) + \gamma\sinh(\gamma h)}. \tag{28}$$

For $h \equiv 1$, this corresponds to the basic state of the absorption flow.

If all lower orders $\psi_j, C_{1,j}$ $(j < n)$ are already determined for $n > 0$, ψ_n can be found by integrating the right-hand sides four times with respect to η, and then matching the boundary conditions by solving a simple linear equation system. By the method of variation of constants, the problem of solving the inhomogenous differential equation (24) is also reduced to an integration and solving a simple linear equation system. It can be shown by induction that the right-hand sides of (23) and (24) will be (formal) polynomials in the terms η, $\cosh(\gamma\eta)$, and $\sinh(\gamma\eta)$. Hence all necessary integrations can easily be performed automatically by the computer algebra system.

The series solution (21) obtained in this way is finally substituted in the kinematic condition (15), which has not been used up to now. This yields the evolution equation for the film thickness h:

$$h_\tau + [\psi_0(h)]_\xi + \alpha[\psi_1(h)]_\xi + \alpha^2[\psi_2(h)]_\xi + \cdots = 0. \tag{29}$$

For our example, this equation is given up to first order in α by

$$h_\tau + 2h^2h_\xi + \alpha\frac{\partial}{\partial\xi}\left[\frac{8}{15}Rh^6h_\xi - \frac{2}{3}\cot\beta h^3h_\xi + \frac{2}{3}\bar{S}h^3h_{\xi\xi\xi} - \frac{4M\,Sh\,\gamma^2h^2h_\xi}{(Sh\cosh(\gamma h) + \gamma\sinh(\gamma h))^2}\right] + \cdots = 0. \tag{30}$$

If the absorption has no influence of the film flow (i.e., $M = 0$), the second-order expansion obtained in this way is equal to the one given by Lin [5]. If the surface tension terms are rescaled according to $\bar{S} \to \alpha^2\bar{S}$, the third-order expansion is the same as the one given by Nakaya [9].

Some of the steps in the above outline of the symbolic algorithm were presented for the model problem only in order to avoid some necessary, but minor case distinctions for the general equation. For example, if a buoyancy effect is present, the right-hand side of the nth order equation (23) for ψ_n depends also on $C_{1,n}$, which means that the species equations must always be solved prior to the stream function equations. Some of these case distinctions were included in our MAPLE-implementation of the above algorithm, which is thus capable of deriving the evolution equations automatically for a wide range of different film flow problems, if the problem dependent terms $\mathbf{G}$, F_i, L, K, B_w, and B_s in the general equation system are given as input.

4. LINEAR STABILITY ANALYSIS

Using computer algebra for a linear stability analysis of the $(2n+1)$st order evolution equation is straightforward: We substitute a normal mode perturbation of the uniform flow

$h = 1 + \delta \exp(ik(\xi - c\tau))$ into Eq. (29), taylor-expand up to first order in δ, and then divide by $\delta \exp(ik(\xi - c\tau))$. The result is the $(2n+1)$st order dispersion relation $D(k, c) = 0$. In a spatial stability analysis, we assume that $k = 1 + ik_i$ and c real. A symmetry consideration shows that k_i must be an odd and c must be an even function of α. Hence a symbolic solution of the dispersion relation, which is consistent with its truncation order, can easily be found by assuming expansions

$$k_i = \sum_{j=0}^{n} \kappa_{2j+1} \alpha^{2j+1}, \qquad c = \sum_{j=0}^{n} \lambda_{2j} \alpha^{2j} \tag{31}$$

for k_i and c and then determining the coefficients κ_{2j+1} and λ_{2j} by equating like powers of α. Thus the following formula for the spatial growth rate—which is given by $-\alpha k_i$ in the original scaling—is found from the first-order equation for our model problem:

$$-\alpha k_i = \alpha^2 \left(\frac{1}{15}(4R - 5\cot\beta - 5\alpha^2 S) - \frac{1}{2} \frac{M\, Sh\, \gamma^2}{(Sh\cosh(\gamma) + \gamma \sinh(\gamma))^2} \right). \tag{32}$$

If the chemical reaction is not present or has no effect on the surface tension (i.e., $M = 0$, $Sh = 0$, or $\gamma = 0$), this reduces to the well-known result that film flow is unstable for small wavenumbers if $R > 5/4 \cot\beta$ and that increasing the capillary parameter S decreases both the critical wavenumber and the growth rates of the unstable modes (see, e.g., Lin and Wang [3]).

If neither M, Sh, nor γ are zero, Eq. (32) shows that the Marangoni effect caused by the absorption stabilizes the film flow: The critical Reynolds number R_c is increased to

$$R_c = \frac{5}{4}\cot\beta + \frac{15}{8} \frac{M\, Sh\, \gamma^2}{(Sh\cosh(\gamma) + \gamma \sinh(\gamma))^2}. \tag{33}$$

This stabilizing effect can be explained qualitatively by the fact that the gradient of the concentration profile in the base state is positive. Thus the surface tension will be decreased at crests and increased at troughs of a disturbance, which causes a leveling flow.

In order to resolve the dispersion of linear waves, higher-order evolution equations must be considered. Up to fourth-order corrections, the phase velocity is thus found to be given by

$$\begin{aligned} c = 2 + \alpha^2 \Bigg[& \left(\frac{40}{63} R(\alpha^2 S + \cot\beta) - \frac{32}{63} R^2 - 2 \right) + M R\, Sh \Bigg\{ \frac{19}{5} \frac{\gamma^2}{(Sh\cosh(\gamma) + \gamma\sinh(\gamma))^2} \\ & + Sc \left(\frac{(16\gamma^3 - 9\gamma)\cosh(\gamma) + 6(\gamma^2+1)\sinh(\gamma) + 3\gamma\cosh(3\gamma)}{6\gamma(Sh\cosh(\gamma) + \gamma\sinh(\gamma))^3} \right. \\ & \left. + Sh \frac{(16\gamma^2 - 3)\sinh(\gamma) + 6\gamma\cosh(\gamma) + 3\gamma\sinh(3\gamma)}{6\gamma(Sh\cosh(\gamma) + \gamma\sinh(\gamma))^3} \right) \Bigg\} \Bigg]. \end{aligned} \tag{34}$$

Again, if the absorption does not influence the flow, this formula reduces to the result given in Lin and Wang [3].

An obvious advantage of the long-wave approximation compared to numerical solutions of the Orr–Sommerfeld equations is that the results are given in symbolic form and hence allow for a qualitative understanding of the mechanisms. As an example we analyze the

dependence of the growth rate (32) on the Sherwood number *Sh*: If the mass transfer is very fast ($Sh \to \infty$) the stabilizing effect of the absorption vanishes since the surface concentration will be always equal to the concentration in the air and hence the surface tension will be constant. On the other hand, if there is no mass transfer at all ($Sh \to 0$), Eq. (32) reduces to the same result since there is now no chemical reaction and hence no concentration gradient in the film. It should be noted that the capillary number S, which was calculated with the surface tension corresponding to the air concentration, should actually be replaced in this case by $S + \alpha^2 M$ in Eq. (32), since the concentration at the surface, as well as in the whole film, is now zero. This small difference is of course consistent with the truncation order of (32) and is corrected if higher orders are considered.

From the two limits above it is clear that there is a finite value of *Sh* for which the stabilizing effect is maximal. This suggests enhancing the stability of the flow by controlling the Sherwood number, which might be done, at least to some extent, by modifying the air flow above the film. The stabilizing effect becomes maximal for $Sh = \gamma \tanh \gamma$. As an example, Fig. 2 shows the dependence of the spatial growth rate for the values $Sh = 0$, $Sh = 5$, and $Sh = 0.762$ which is the "optimal" Sherwood number for $\gamma = 1$.

While the above example shows the usefulness of low-order long wave approximations for the stability analysis of film flow problems with complex transport processes, there is no decisive way to determine a priori whether the asymptotic result is in fact reliable for a given small but finite wavenumber. The computer algebra methods offer a heuristical way to estimate the range of validity of the approximation, simply by comparing the low-order results with higher order results. Figure 3 compares the predictions of the dependence of the critical wavenumber (i.e., the smallest positive value α_c such that $k_i = 0$) on the Reynolds number by the first-, third-, and fifth-order approximation. For $R > 3$ the first and third order deviate considerably from each other which indicates that the first-order approximation is

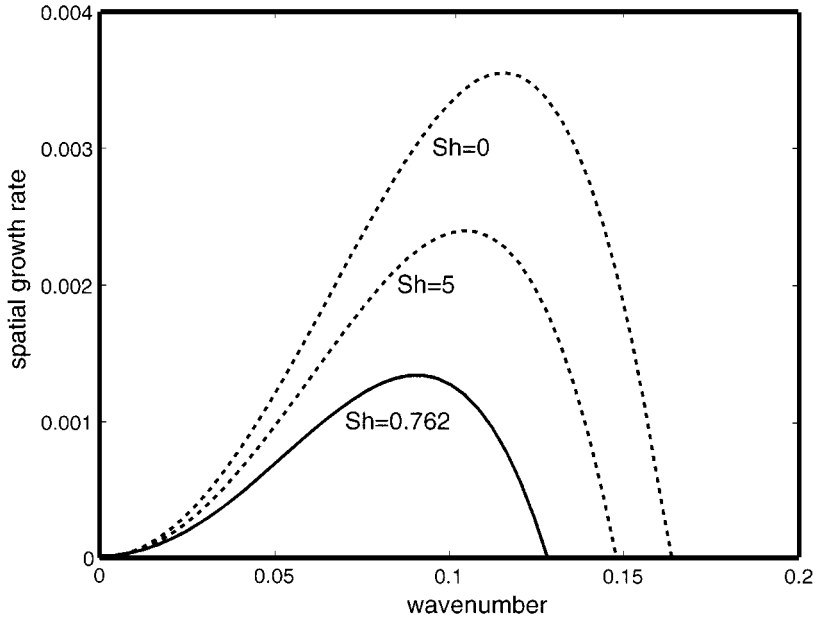

FIG. 2. Stabilization of film flow by controlling the absorption rate via the Sherwood number. Other parameters are given by $R = 2$, $S = 60$, $\beta = \pi/2$, $M = 3$, $Sc = 5$, $\gamma = 1$.

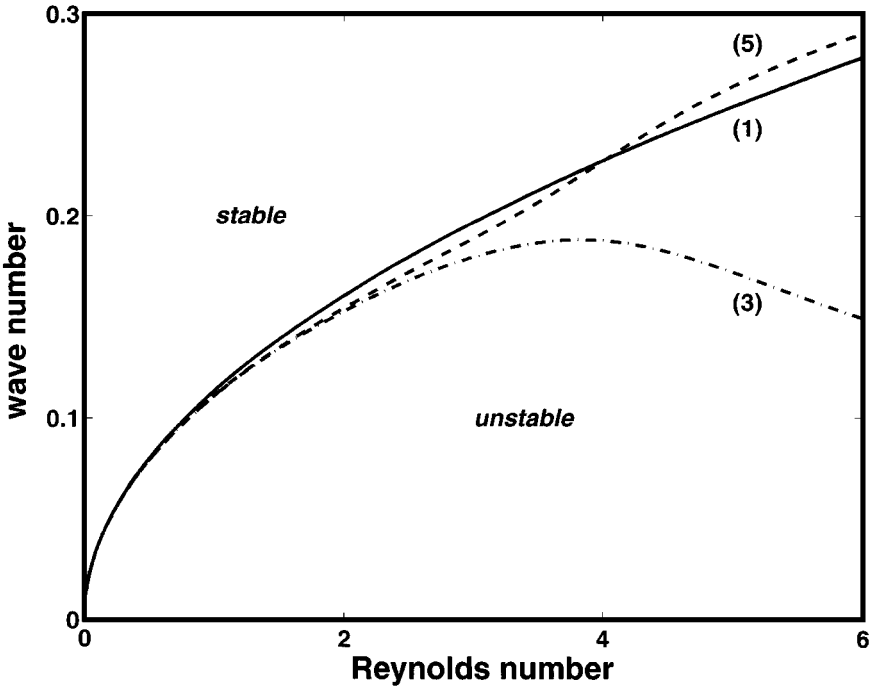

FIG. 3. Neutral stability curves as predicted by different orders of the long-wave approximation ($S = 60$, $\beta = \pi/2$, no absorption).

no longer valid. Comparison with the fifth-order approximation shows that the range of validity of the third-order approximation is not significantly larger.

However, the weak improvement in the range of stability for higher-order approximations is mainly due to the fact that the polynomial approximations (31) of k_i and c are not very convenient because of the—clearly unphysical—tendency of high-order polynomials to oscillate for increasing α. This unphysical behaviour can be overcome by using a rational (Padé-) approximation instead:

$$-k_i = \alpha \frac{p_0 + p_2\alpha^2 + \cdots}{1 + q_2\alpha^2 + \cdots}, \qquad c = \frac{r_0 + r_2\alpha^2 + \cdots}{1 + s_2\alpha^2 + \cdots}. \tag{35}$$

The easiest way to find appropriate Padé-approximants of k_i and c is to calculate the Padé-approximations of the polynomial approximants which we have already determined. Although the range of validity of these polynomial approximations is very small, they do match the first few derivatives at $\alpha = 0$ of the exact growth rate and phase velocity, which is all we need to calculate Padé-approximants of the exact solutions.

In addition, the rational approximations can be constructed in such a way that they reflect the expectation that very short waves should decay rapidly because of surface tension, i.e., $k_i \to \infty$ as $\alpha \to \infty$. The (3,2)-Padé-approximation for k_i is the only one which can be derived from our fifth-order polynomial approximation which has the expected behaviour for $\alpha \to \infty$ as well as the correct symmetry properties. In Fig. 4, we present predictions of the first-, third-, and fifth-order polynomial approximations and a (3,2)-Padé-approximation. These predictions are compared with experimental data taken from Krantz and Owens [15] for the growth rate and the phase velocity of a disturbance of a film flow on a vertical wall.

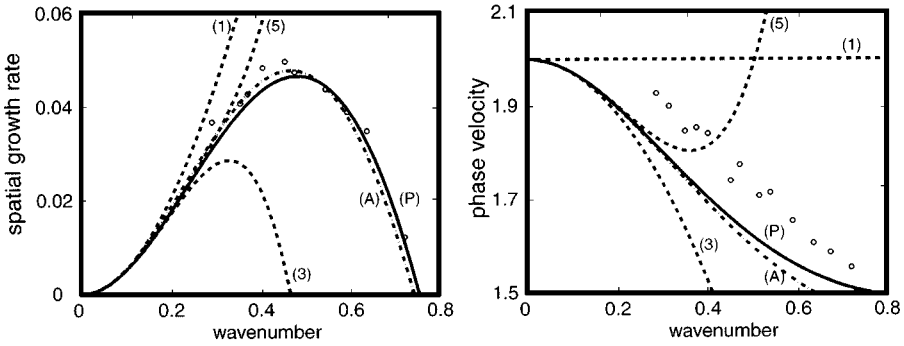

FIG. 4. Spatial growth rate and phase velocity as predicted by different polynomial approximation orders ((1), (3) and (5)), the Padé-approximation (P), and the numerical solution of the simplified Orr–Sommerfeld equation (A). Experimental data taken from Krantz and Owens. Parameters are $R = 1.94$, $S = 0.815$, and $\beta = \pi/2$.

A numerical solution of the approximate Orr–Sommerfeld equation as proposed by Anshus and Goren [17] is also included.

As shown in Fig. 4, the agreement of the predictions by the fifth-order Padé-approximation with experimental data is as good as that of the Anshus/Goren method, while the polynomial approximations are only valid up to a wavenumber of 0.25. In particular, they predict neither the critical wavenumber nor the wavenumber of maximum growth rate correctly. As indicated by Krantz and Owens [15], the disagreement between the predictions of the Anshus/Goren method for the phase velocity and the experimental data is likely due to nonlinear effects.

Figure 5 shows a similar comparison with data obtained by Liu *et al.* [16] for an very small inclinination angle β and $R = 23$. Even for this comparatively high Reynolds number the Padé-approximation still agrees at least qualitatively with the numerical solution and the data, although the polynomial approximations already fail for wavenumbers as small as 0.02.

5. PERMANENT WAVES

The main objective of this section is to illustrate how higher-order theories obtained by computer algebra techniques provide a useful heuristic criterion to assess the range

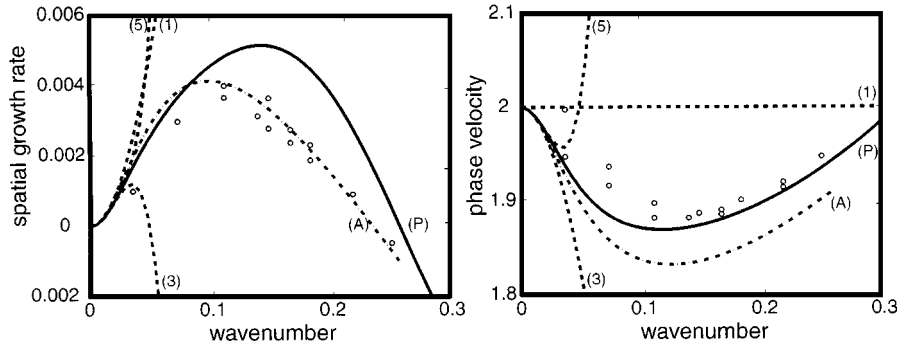

FIG. 5. Spatial growth rate and phase velocity as predicted by different polynomial approximation orders ((1), (3) and (5)), the Padé-approximation (P), and the numerical solution of the simplified Orr–Sommerfeld equation (A). Experimental data of Liu, Paul, and Gollub. Parameters are $R = 23$, $S = 62$, and $\beta = 0.08$.

of validity of standard weakly nonlinear theories. As an example for a weakly nonlinear analysis, we use Landau theory in order to describe the equilibration of a linearly unstable monochromatic disturbance due to the nonlinear interaction with its stable higher harmonics. In order to account for these interactions up to third order of the amplidude of the disturbance, we follow Nakaya [9] and express $h(\tau, \xi)$ as a Fourier series

$$\begin{aligned} h = {} & \varepsilon(e^{i\varphi} + e^{-i\varphi}) + \varepsilon^2\left(h_{2,2}\, e^{2i\varphi} + h_{0,0} + h_{-2,2}\, e^{-2i\varphi}\right) \\ & + \varepsilon^3\left(h_{3,3}\, e^{3i\varphi} + h_{1,3}\, e^{i\varphi} + h_{-1,3}\, e^{-i\varphi} h_{-3,3}\, e^{-3i\varphi}\right). \end{aligned} \tag{36}$$

Here the coefficients $h_{-k,l}$ are the complex conjugates of $h_{k,l}$. The dependence of the amplitude ϵ and the phase φ on (τ, ξ) is approximated by the expansions

$$\begin{aligned} \varepsilon_\xi &= 0, \qquad \varepsilon_\tau = s_1\varepsilon + s_3\varepsilon^3 + \cdots \\ \varphi_\xi &= 1, \qquad \varphi_\tau = \omega_1 + \omega_3\varepsilon^2 + \cdots. \end{aligned} \tag{37}$$

Again the substitution of the expansions (36) and (37) into the evolution equation is readily implemented in a computer algebra system. Equating like powers in ε yields a hierarchy of linear equations for the coefficients $h_{l,k}$, s_k, and ω_k. The symbolic solutions of these linear equations are easily found if $h_{3,1}$ is set to zero, following the arguments given in Nakaya [9].

Since $\varepsilon_\tau = 0$ for a permanent wave, its amplitude is found from (37) to be

$$\epsilon = \sqrt{-\frac{s_1}{s_3}}. \tag{38}$$

Permanent waves only exist if this amplitude is real. If we base the analysis on the first-order evolution equation for the example problem, this condition is given by the symbolic expression

$$\begin{aligned} 0 < \varepsilon^2 = {} & -\frac{\alpha^2}{450}\left(-4R + 5\cot\beta + 5S\alpha^2 + \frac{15}{2}\frac{M\,Sh\,\gamma^2}{(Sh\cosh\gamma + \gamma\sinh\gamma)^2}\right) \\ & \times\left(-4R + 5\cot\beta + 35S\alpha^2 + \frac{15}{2}\frac{M\,Sh\,\gamma^2}{(Sh\cosh\gamma + \gamma\sinh\gamma)^2}\right). \end{aligned} \tag{39}$$

The positive root of the first factor of (39) is the critical wavenumber already found from linear stability analysis, which is an upper bound for the existence of permanent waves. The second factor of expression (39) also suggests a lower bound for the existence of permanent waves. It is intuitively clear that the equilibrating effect of the nonlinear interaction will vanish as the wavenumber decreases, since the first harmonics of an unstable monochromatic wave become eventually unstable, too. However, the relevance of the lower bound predicted by (39) is questionable since the amplitude for the first harmonic $\varepsilon^2|h_{2,2}|$ becomes larger than ε close to that wavenumber, which violates the assumptions of Landau theory.

Using a higher-order evolution equation for the weakly nonlinear analysis corroborates that the lower bound given by (39) is meaningless, as is illustrated in Fig. 6. It compares the dependence of the amplitudes for the first three harmonics (ε, $\varepsilon^2|h_{2,2}|$, $\varepsilon^3|h_{3,3}|$) on the wavenumber as predicted by the first- and third-order evolution equation. In the case shown, the results agree reasonably well down to wavenumber 0.07, but exhibit extreme qualitative differences as the wavenumber decreases further. Instead of the treacherously regular

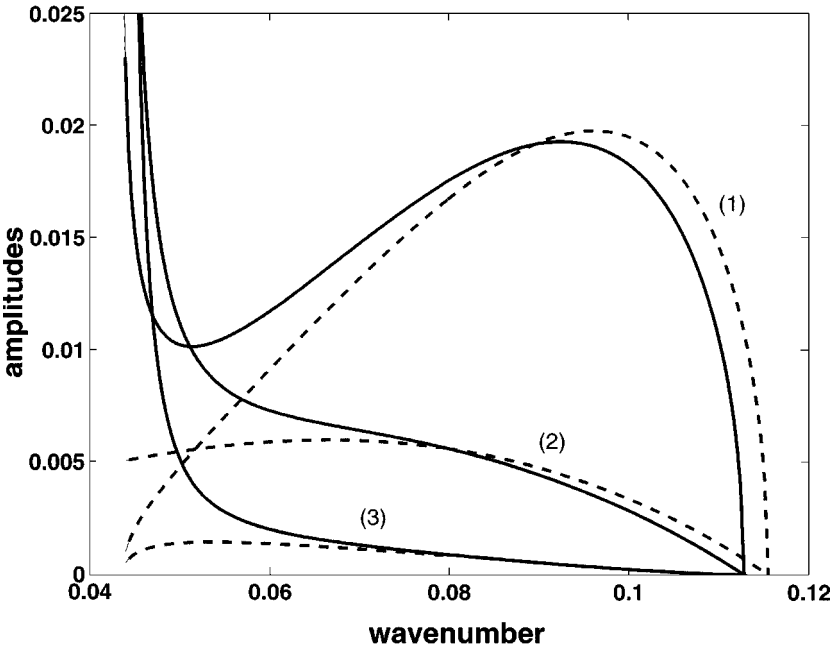

FIG. 6. Illustration of the invalidity of Landau theory far away from the bifurcation point: The contributions of the first three modes as predicted by Landau-theory using the first- (dashed line) and third-order (solid line) evolution equation differ even qualitatively for small wavenumbers ($R = 1$, $S = 60$, $\beta = \pi/2$, no absorption).

behaviour of the first-order results, the third-order amplitudes finally reach a singularity. This is due to the fact that the third-order evolution equation captures more details of the complex nonlinear dynamics of the original system and this nonlinear interaction causes increasing contributions of higher harmonics to the solution of the evolution equation as the wavenumber decreases, such that the representation (36) finally fails to approximate it. In this context, it is also interesting to look at numerical solutions of the "fully nonlinear" evolution equation (30) which are obtained by a pseudospectral approach similar to the method used by Joo *et al.* [12]. Figure 7 shows the first three Fourier coefficients of the equilibrium solutions that evolved from a initial sinusoidal disturbance of a given wavenumber. These numerical solutions agree well with the first-order Landau theory down to wavenumber 0.065, where they undergo an aprupt change. As the wavenumber decreases, the contributions of higher modes become more and more important, and it takes longer and longer to reach the equilibrium. At wavenumber 0.054, the numerically obtained solution appeared not to equilibrate anymore but to become time-periodic. For higher Reynolds numbers, the numerically calculated equilibrium solutions resemble the results of the third-order Landau-theory rather than the first-order results. In the vicinity of the pole the numerical solutions exhibit the "catastrophic" behaviour in time, also observed by Joo *et al.* [12].

These numerical observations obviously corroborate that a comparison between third-order and first-order results can be used as a heuristic criterion of the range of validity of the weakly nonlinear theory beyond the bifurcation point. (The intermediate chararacter of the "fully nonlinear" evolution equation should be pointed out: while taking only first-order terms into account, it captures the full nonlinear mode interaction which is pruned by first- and third-order Landau theory.) The situation illustrated above for Landau theory agrees also qualitatively with results obtained by a higher-order version of Gjevik's method [6]

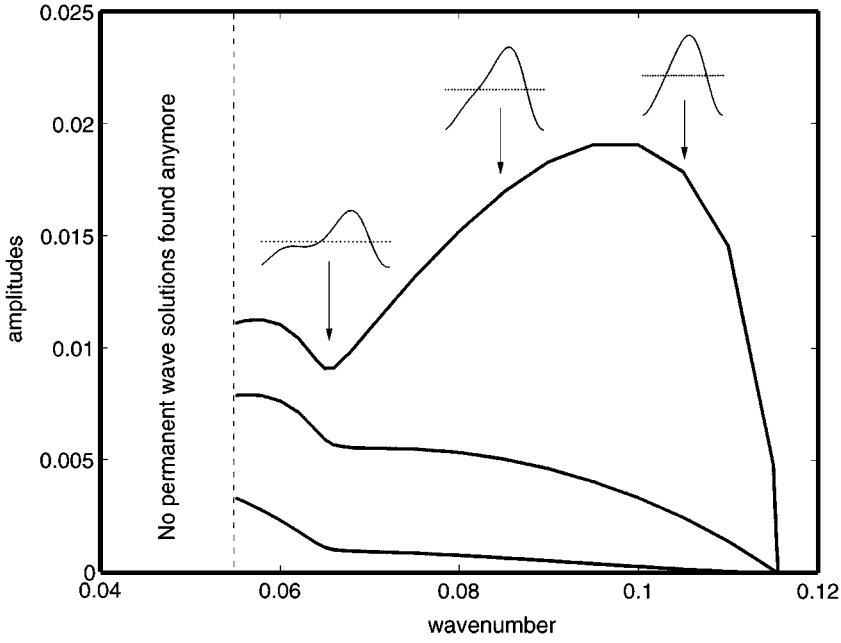

FIG. 7. Contributions of the first three modes versus the wavenumber of the initial disturbance as predicted by numerical solution of the "fully nonlinear" first-order evolution equation. Also shown are some characteristic waveforms ($R = 1$, $S = 60$, $\beta = \pi/2$ no absorption).

and by a weakly nonlinear analysis based on higher-order equations of the Kuramoto–Sivashinski hierarchy (see, e.g., Shlang and Sivashinsky [18]), which again can be derived automatically from the higher-order evolution equations using computer algebra.

6. CONCLUSION

We illustrated how a large class of thin film flow problems coupled with heat and mass transfer processes can be investigated within a common framework using computer algebra methods. Even in situations involving complex transport processes interacting with the flow, the appropriate long-wave evolution equations can be derived automatically, and their dynamic behaviour is readily studied by standardized implementations of the methods of linear and nonlinear stability analysis.

On one hand, an analysis based on low-order equations has the advantage to result in relatively simple formulas describing the stability behaviour, which provide a good orientation for problems depending on many parameters and may suggest strategies for optimization of industrial processes involving thin film flows. On the other hand, considering higher-order evolution equations yields heuristic criteria estimating the ranges of validity for both the linear and the nonlinear results obtained at first order. Using Padé-approximations, linear stability analysis based on the high-order long-wave approximation agrees well with experimental results and numerical solutions of the Orr–Sommerfeld equations even for relatively short waves and moderately high Reynolds numbers.

While presented here for the stream function formulation of the Navier–Stokes equation for simplicity, the automatic derivation of evolution equations is readily extended to three

dimensions. Another straightforward extension which is particularly useful for applications to coating processes is to consider multilayer films, which yields a system of evolution equations for the thicknesses of the different layers.

The available computer hardware, in particular memory, clearly limits the applicability of the symbolic methods presented in this paper. The storage space needed by our MAPLE-implementation of the derivation of the evolution equation increased roughly by a factor 4 from one order to the next for a film without transport processes. For the fifth order, a maximum of 40 Megabytes was necessary. The computing time for the fifth order was about 20 minutes on a IBM Risc 6000 workstation.

A more fundamental limitation of the computer algebra methods is given by the fact that the zeroth order of the equation hierarchy must allow a closed form solution. Thus they cannot be applied to film flows of fluids with complex rheological behaviour or transport processes involving higher-order chemical reactions. Despite these restrictions, there is a large number of applications involving thin film flow coupled with transport processes, for which the computer algebra methods outlined in this paper are a very useful engineering tool.

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