On application of lubrication approximations to nonunidirectional coating flows with clean and surfactant interfaces

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In this work, the characteristic properties of the lubrication approximation are studied and its *weak ellipticity* is established, in contradistinction to the commonly accepted parabolic character of the lubrication equations resulting from the underlying unidirectional flow assumption. The weak ellipticity property allows the lubrication analysis to capture flow topologies around stagnation points, contact lines, and flows over edges, all of which normally require elliptic operators to be accounted for. This is used to explain the empirically observed overperformance of the lubrication approximation from the perspective of characteristic analysis. While the analysis is developed in the context of the classical Landau–Levich problem of dip-coating, which is known to possess an interfacial stagnation point both in the clean and surfactant interface cases, the analysis is general since the Landau–Levich equation is common to many other lubrication problems. The analytical approach presented here when applied to the surfactant interface case, also allows one to establish a new physical result: a variation of the bulk surfactant concentration is the necessary condition for the film thickening phenomenon in the Landau–Levich problem to occur due to surfactant-induced Marangoni effects. © *2010 American Institute of Physics*. [doi:10.1063/1.3484276]

I. INTRODUCTION

The lubrication approximation for thin film flows has received pervasive use in fluid dynamics since the pioneering work of Reynolds,[1,2] which is based on the assumption that the flow is almost unidirectional. The original idea is analogous to that of Prandtl for boundary-layer type flows,[3] i.e., that the dynamics of the bulk fluid is subject to the approximation of parabolic characteristic type as will be discussed in detail in Sec. II C. However, the lubrication equations have been applied to the situations where the assumption of a unidirectional flow fails and thus the elliptic nature of the flow should not be neglected; examples include flows near contact lines,[4] over a cavity,[5] and over an edge[6] as depicted in Fig. 1. The obvious reasons for such "extensions" of the lubrication approach without rigorous justification are that the lubrication equations are much simpler than the Navier–Stokes or Stokes equations, and, as was found empirically, such equations work reasonably well even for the aforementioned "prohibited" flows.[4–6]

The *first goal* of this work is to give a justification from the point of view of classical method of characteristic analysis for the application of lubrication approximations to thin film flows with special points, which normally require a full elliptic (Stokes) operator to correctly resolve the structure of the flow. Such points may include eddy centers and stagnation points[7] and are referred to here as *elliptic* points. This notion should not be confused with the one in dynamical systems[8] used to describe fixed points $\boldsymbol{x}_0$ of a map $\dot{\boldsymbol{x}}\mapsto\boldsymbol{f}(\boldsymbol{x})$, $\boldsymbol{x}\in\mathbb{R}^N$ such that the eigenvalues of the linearization of the map around $\boldsymbol{x}_0$, i.e., $\boldsymbol{Df}(\boldsymbol{x}_0)$ have a unit modulus. The analog of an elliptic fixed point for vector fields is a center, i.e., when trajectories of the system $\dot{\boldsymbol{x}}=\boldsymbol{f}(\boldsymbol{x})$ appear as "ellipses" in a sufficiently small neighborhood of the fixed point $\boldsymbol{x}_0$. While one can apply the dynamical systems terminology if the flow field is considered as a vector field of some dynamical system (e.g., stagnation points in the bulk in Fig. 6 can be regarded as hyperbolic fixed points), here we deal with flows having free and rigid boundaries and therefore such terminology would not always be appropriate. In view of that and because of the nature of the analysis developed in this work, we classify fixed points of the flow based on the nature of a partial differential operator needed to resolve them as discussed above. For concreteness and because of their practical importance, we will focus on stagnation points, such as that illustrated in Fig. 2. However, due to the generality of the characteristics property of the lubrication approximation established below, the general origin of the unexpected performance of this approximation in a variety of situations[4–6] is clarified.

First, in Sec. II, we will perform the characteristics analysis of the clean interface case in the context of the classical Landau–Levich problem of dip-coating, i.e., when a solid substrate is coated with a thin film by being pulled out of a liquid bath as depicted in Fig. 3(a). As the name implies, the problem was first studied by Landau and Levich[9] by exploiting the lubrication approximation, which was later put on a firm foundation of matched asymptotic expansions.[10] The same approximation applies to the Bretherton problem in Fig. 3(b) of film deposition in a channel by a penetrating bubble. The Landau–Levich (-Bretherton) problem is particularly suitable for the purpose of our discussion since the elliptic stagnation point (cf. Fig. 3) can be captured by the lubrication approximation as will be demonstrated in Sec. II.

The behavior of this elliptic point—the stagnation point in Fig. 3(a)—also has nontrivial consequences for the physics of dip-coating in the presence of surfactants. Thus, the

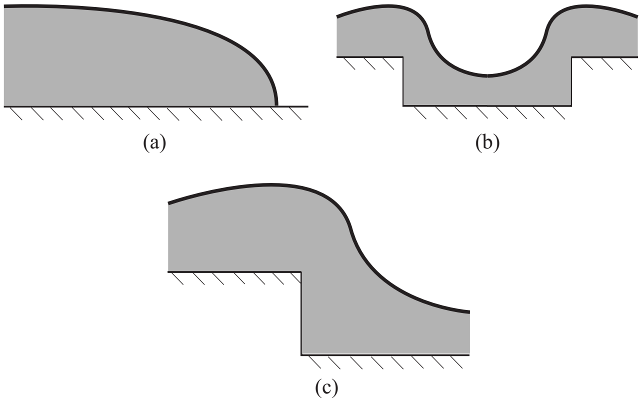

FIG. 1. Thin film flows containing elliptic regions, which are often analyzed with lubrication approximations (Refs. 4–6): (a) flow near the contact line, (b) flow over the cavity, and (c) flow over the edge.

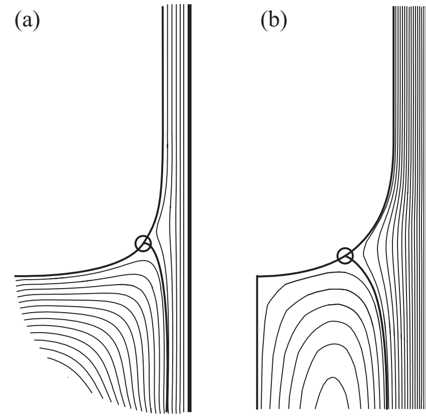

FIG. 3. Flow topologies in the classical film deposition: (a) Landau–Levich flow (the flow region extends to infinity) and (b) Bretherton flow (only half of a channel is shown). Encircled are elliptic points of interest to this study. Bretherton problem has a second stagnation point (at the axis of symmetry), which is not captured by the lubrication approximation, as discussed in Sec. III B.

second goal of this paper is to understand the characteristic properties of the lubrication approximation in the presence of surfactants and to clarify the behavior of the stagnation point in the Landau–Levich problem, which is done in Sec. III. One is thereby able to gain new insight into the surfactant-laden Landau–Levich problem and establish a necessary condition for the film thickening due to surfactant-induced Marangoni effects, which is still an unexplained phenomenon[11,12] as discussed in detail in Sec. III.

II. WEAK ELLIPTICITY: CLEAN INTERFACE CASE

In this section, we first provide a brief but self-contained derivation of the lubrication approximation in the context of the Landau–Levich problem (Sec. II A), which in Sec. II B is shown to be able to capture elliptic points. An explanation of this fact is given in Sec. II C based on the characteristic analysis, and implications of these results for other lubrication flows are discussed in Sec. II D.

A. A concise derivation of the lubrication approximation

We consider a two-dimensional steady dip-coating flow and choose the (x,y)-system of coordinates with the unit vectors $\mathbf{i}$ and $\mathbf{j}$, respectively. The interface is given by $y=-h(x)$ with $h,h_x,h_{xx}\geq 0$, as shown in Fig. 4, so that the outer normal to the interface is $\mathbf{n}=-(\mathbf{i}h_x+\mathbf{j})/\sqrt{1+h_x^2}$, the tangent vector $\mathbf{t}=-(\mathbf{i}-\mathbf{j}h_x)/\sqrt{1+h_x^2}$, and the interfacial curvature $\kappa=\nabla\cdot\mathbf{n}=-h_{xx}/(1+h_x^2)^{3/2}$. The flat bath interface at $y\to-\infty$ corresponds to $x=0$. We consider the entrainment of a film due to the vertical motion of the solid boundary with velocity $-U$. For convenience, the analysis will be done in nondimensional variables defined by $(x,y)\to l_c(x,y)$, $(u,v)\to U(u,v)$, $p\to\sqrt{\rho g\sigma}p$, where $l_c=\sqrt{\sigma/\rho g}$ is the capillary length and $\sqrt{\rho g\sigma}$ is the capillary pressure. With the above choice of nondimensional variables, the problem contains two nondimensional parameters: the capillary number $\mathsf{Ca}=\mu U/\sigma$, which is considered to be asymptotically small here, $\mathsf{Ca}\ll 1$, and an inertial parameter $La=\rho U^4/(\rho g)$ referred to as the Landau number,[11] the square root of which multiplies the inertial terms in the Navier–Stokes equations, cf. Krechetnikov and Homsy.[11] The classical Landau–Levich problem corresponds to the case when the meniscus is static on the scale l_c, which implies low values of $La\ll 1$ and thus the creeping flow regime. As a result, the bulk dynamics obeys the Stokes equations

$$u_x+v_y=0, \tag{1a}$$

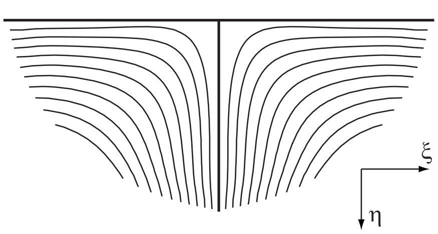

FIG. 2. Streamlines $\psi(x,y)$ of the flow field in the neighborhood of a stagnation point at the interface. If $u=\psi_\eta$ and $v=-\psi_\xi$ are the velocity components, then based on the symmetry of the flow field, i.e., that $u(-\xi)=-u(\xi)$ and $v(-\xi)=v(\xi)$, it is straightforward to show that the Taylor series of the stream-function ψ in the neighborhood of the interfacial stagnation point is $\psi\sim C_1\xi\eta+C_2\xi^3+\cdots$, where constant coefficients $C_1,C_2,\ldots$ are determined from the interfacial boundary conditions.

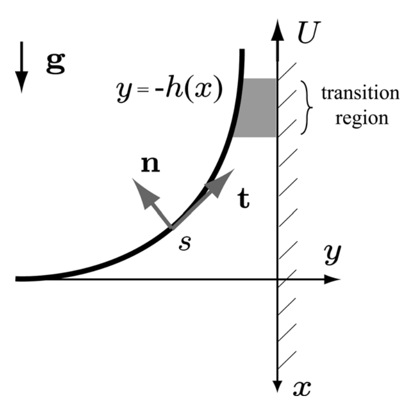

FIG. 4. Coordinate system: $h,h_x,h_{xx}\geq 0$. Thick lines are walls and thin lines are streamlines. The transition region is shaded. The arclength s is measured in the direction from the bath to the film.

$$0 = -p_x + 1 + \mathrm{Ca}\Delta u, \quad (1b)$$

$$0 = -p_y + \mathrm{Ca}\Delta v, \quad (1c)$$

augmented by (a) the normal $\mathbf{n}\cdot T\cdot\mathbf{n} = -\sigma\nabla_s\cdot\mathbf{n}$ and the tangential $\mathbf{t}\cdot T\cdot\mathbf{n} = \mathbf{t}\cdot\nabla_s\sigma$ dynamic boundary conditions at the interface $y=-h(x)$

$$p = \kappa + \frac{2\mathrm{Ca}}{1+h_x^2}[v_y + h_x^2 u_x + h_x(u_y + v_x)], \quad (2a)$$

$$\sigma_s = \frac{\mathrm{Ca}}{1+h_x^2}[2h_x(u_x - v_y) + (1-h_x^2)(u_y + v_x)], \quad (2b)$$

respectively, where s is the natural arclength parameterization, $\mathrm{d}s = -\sqrt{1+h_x^2}\mathrm{d}x$, and (b) the no-slip condition at the wall $y=0$, i.e., $u=-1$ and $v=0$. The kinematic condition at the interface, $uh_x + v = 0$, closes the problem formulation. In this section, we consider the clean interface case $\sigma_s = 0$ only.

With the idea to develop a lubrication approximation for the transition region in the low capillary number regime, $\mathrm{Ca} \ll 1$, which connects the thin film and the meniscus regions, one introduces a thin-layer assumption

$$y \to h_\infty y, \quad x \to \delta x, \quad u \to u, \quad v \to (h_\infty/\delta)v, \quad p \to p, \quad (3)$$

where $1 \gg \delta \gg h_\infty$, so that the dimensional film thickness is $\bar{h}_\infty = \alpha h_\infty l_c$ with the $O(1)$ numerical factor α to be determined later. The assumption of a thin-layer approximation, $\delta \gg h_\infty$, and the balance of capillary and viscous forces (or a formal application of the principle of least degeneracy of Ref. 13) in the x-momentum equation yield

$$\delta = h_\infty^2/\mathrm{Ca}, \quad (4)$$

which leads to the following system for the bulk dynamics:

$$u_x + v_y = 0, \quad (5a)$$

$$0 = -p_x + u_{yy}, \quad (5b)$$

$$0 = -p_y. \quad (5c)$$

Since we are working in two dimensions, it is convenient to introduce the stream-function defined by $u=\psi_y$ and $v=-\psi_x$, so that Eq. (5) can be immediately integrated to produce

$$\psi = p_x\frac{y^3}{6} + C_2\frac{y^2}{2} + C_1 y + C_0, \quad (6)$$

where thanks to the no-slip at the solid wall, $C_0=0$ and $C_1=-1$. In the clean interface case, the interfacial boundary conditions (2) at $y=-h(x)$ yield to the leading order

$$p - h_{xx} = O(\mathrm{Ca}^{2/3}), \quad (7a)$$

$$\psi_{yy} = O(\mathrm{Ca}^{2/3}), \quad (7b)$$

where the requirement that the capillary pressure should be present at the leading order gives $h_\infty \sim \delta^2$, i.e., together with Eq. (4) produces the classical result $\delta = \mathrm{Ca}^{1/3}$ and $h_\infty = \mathrm{Ca}^{2/3}$.

Taking into account the asymptotically simplified interfacial boundary conditions (7a) and (7b), we find $C_2 = p_x h$, and thus the expression for the stream-function (6) becomes

$$\psi = h_{xxx}\frac{y^2}{6}(y + 3h) - y. \quad (8)$$

The classical Landau–Levich equation[9] is deduced via equating the values of the stream-function at $h(x)$ and at $h=\alpha$ for $x=-\infty$, since in the steady case the value of the stream-function at the interface is constant, i.e., $\psi|_{y=-h(x)} = \psi|_{y=-\alpha, x=-\infty}$, which produces

$$h_{xxx}h^2 + 3\left(1 - \frac{\alpha}{h}\right) = 0. \quad (9)$$

The solution of this equation matches the thin film $(x\to-\infty : h\to\alpha)$ and the static meniscus; the latter is determined from the capillary-hydrostatic equilibrium

$$-h_{xx}/(1+h_x^2)^{3/2} = x + D_1. \quad (10)$$

Since $h(x)$ approaches the flat bath interface for $x\to 0$, the constant D_1 vanishes in the above expression. Integrating Eq. (10) once

$$-h_x/(1+h_x^2)^{1/2} = \frac{x^2}{2} + D_2, \quad (11)$$

and using the condition as $x\to 0$ again, i.e., $h_x\to+\infty$, we get $D_2=-1$. Therefore, the point of zero tangency, $h_x(x^*)=0$, is at $x^* = -\sqrt{2}$, and from Eq. (10), we determine the interfacial curvature at that point, $h_{xx} = \sqrt{2}$, which is the second condition for Eq. (9). Scaling $h\to\alpha H$ and $x\to\alpha\xi$, we get the canonical Landau–Levich boundary-value problem

$$H_{\xi\xi\xi}H^2 + 3\left(1 - \frac{1}{H}\right) = 0, \quad (12a)$$

$$\xi\to-\infty: \quad H\to 1, \quad H_\xi \to 0, \quad H_{\xi\xi}\to 0, \quad (12b)$$

integration of which gives $\xi\to+\infty : H_{\xi\xi}\to\sqrt{2}\alpha$ and the value of the factor $\alpha \simeq 0.945$. The deduced Landau–Levich law is also confirmed experimentally: see the discussion in Krechetnikov and Homsy.[12]

B. Capture of elliptic points

The expression for the stream-function (8) yields the interfacial velocity

$$u_{(s)} = \psi_y|_{y=-h} = -\tfrac{1}{2}h_{xxx}h^2 - 1. \quad (13)$$

With the use of Eq. (9), the interfacial velocity is expressed as

$$u_{(s)} = \frac{3}{2}\left(1 - \frac{\alpha}{h}\right) - 1, \quad s \in (-\infty, +\infty), \quad (14)$$

which indicates that $u_{(s)}$ changes sign along the interface at least once

$$s\to+\infty: \quad u_{(s)} = -1,$$

$$s \to -\infty: \quad u_{(s)} = 1/2.$$

As follows from Eq. (14), the interfacial velocity vanishes at $h=3\alpha$, i.e., the lubrication approximation is able to capture this stagnation point and the velocity field around it. Its location and the surrounding flow field are consistent (within asymptotic accuracy) with the full numerical solution of the Stokes equations (1), cf. Krechetnikov and Homsy[11]—this is, of course, of no surprise as the location of the stagnation point controls the mass flux into the film, cf. Fig. 3(a), and should the lubrication analysis fail to capture it correctly, the resulting Landau–Levich law[9] would be inconsistent with reality. Alternatively, representing the interfacial shape near the stagnation point x^* in the Taylor series form $h(x)=\Sigma_{i=0}^{\infty} c_i (x-x^*)^i / i!$, where $c_0=3\alpha$ and $c_3=-2/(9\alpha^2)$ result from the solution constructed in Sec. II A, it is straightforward to show through a change of coordinates $(x,y)\to(\xi,\eta)$ that the flow field of the oblique stagnation point (8) shown in Fig. 3(a) can be mapped to the Stokes flow field in the neighborhood of the "orthogonal" stagnation point at the flat interface discussed and shown in Fig. 2. More precisely, from the point of view of differential topology,[14] these two flows are diffeomorphic, i.e., there is a smooth bijective map between them with smooth inverse. The fact that the lubrication approximation captures a stagnation point, which is obviously of elliptic character, may appear to contradict the fact of the parabolic characteristic type of the bulk dynamics (5) that naturally follows from the originally assumed thin-layer approximation [the parabolic character of Eq. (5) will also be established in Sec. II C]. This paradox is resolved in Sec. II C with the help of characteristic analysis.

C. Characteristics analysis of the lubrication equations

In our characteristics analysis and classification of characteristic types of partial differential operators, we essentially follow Petrovsky[15] and Courant;[16] for the reader's convenience some technical details of the analysis are given in the Appendix. In order to determine the characteristic type of Eq. (5), let us first consider the case when the pressure p in Eq. (5) is a given function, which is standard in the boundary layer theory[3] since the pressure cannot be found from system (5) alone. If Ω denotes the characteristic surface in the (x,y)-plane, then the corresponding characteristic determinant for the system (5) with prescribed pressure $p(x,y)$ is obtained by direct application of the formula (A3) for the characteristic determinant from the Appendix

$$\begin{vmatrix} \Omega_x & \Omega_y \\ \Omega_y^2 & 0 \end{vmatrix} = \underset{\text{parabolic}}{\overset{\text{continuity}}{(\Omega_y)}} \times \underset{\text{parabolic}}{\overset{\text{diffusion}}{(\Omega_y^2)}} = \Omega_y^3 = 0, \tag{15}$$

where the terms in the first row of the determinant matrix originate from the continuity equation, while the second row is due to the reduced diffusion. The factors Ω_y and Ω_y^2 in the determinant (15), corresponding to the continuity and reduced diffusion, both contribute to the parabolic behavior.[17] Indeed, because all three roots of Eq. (15) are real with the degenerate characteristics $\Omega(x)=\text{const}$ normal to the wall, then system (5) is parabolic[15,16] and, therefore, disturbances propagate in both positive- and negative-y directions with an infinite speed. Also, even if the pressure $p(x,y)$ is considered as an unknown function, in which case the characteristic determinant for Eq. (5) is of the form

$$\begin{vmatrix} \Omega_x & \Omega_y & 0 \\ \Omega_y^2 & 0 & -\Omega_x \\ 0 & 0 & -\Omega_y \end{vmatrix} = \underset{\text{parabolic}}{\overset{\text{continuity}}{(\Omega_y)}} \times \underset{\text{parabolic}}{\overset{\text{diffusion}}{(\Omega_y^2)}} \times \underset{\text{parabolic}}{\overset{\text{pressure}}{(\Omega_y)}} = \Omega_y^4 = 0, \tag{16}$$

system (5) is clearly still of parabolic type.

This is opposed to the Stokes equations (1), in which case the characteristic determinant is

$$\begin{vmatrix} \Omega_x & \Omega_y & 0 \\ \mathrm{Ca}(\Omega_x^2+\Omega_y^2) & 0 & -\Omega_x \\ 0 & \mathrm{Ca}(\Omega_x^2+\Omega_y^2) & -\Omega_y \end{vmatrix} = \underset{\text{elliptic}}{\overset{\text{incompressibility}}{(\Omega_x^2+\Omega_y^2)}} \; \mathrm{Ca}^2 \underset{\text{elliptic}}{\overset{\text{diffusion}}{(\Omega_x^2+\Omega_y^2)^2}} = 0, \tag{17}$$

thus implying that there are no real characteristics, i.e., system (1) is of elliptic type. The property of elliptic operators, such as the Stokes operator in Eq. (1) defined in the (x,y)-plane, is that disturbances propagate in all directions with infinite speed. Let us now demonstrate that the lubrication approximation possesses the same property, although the mechanism for disturbance propagation is different.

To make the analysis transparent, first note that the unsteady version of the Landau–Levich equation (9) is obtained simply by inserting the stream-function solution (8) into the unsteady kinematic condition at $y=-h(t,x)$

$$h_t + u h_x + v = 0, \tag{18}$$

which produces

$$h_t - h_x - \partial_x[h_{xxx} h^3/3] = 0. \tag{19}$$

In order to understand the characteristic type of Eq. (19), in particular, the propagation of disturbances in this physical system, Eq. (19) needs to be linearized around the steady state $H(x)$ by introducing an unsteady perturbation $h(t,x)=H(x)+h'(t,x)$. The resulting linearized equation reads (omitting the primes)

$$h_t - h_x - \partial_x[h_{xxx}(H^3/3) + H_{xxx} H^2 h] = 0, \tag{20}$$

which has the characteristic surface $\tilde{\Omega}(t,x)$ defined by

$$\tilde{\Omega}_x^4 = 0, \tag{21}$$

thus implying that Eqs. (9) and (19) are parabolic and the corresponding characteristics are the surfaces $\tilde{\Omega}(t)$ parallel to the x-axis. Therefore, on the basis of the characteristic analysis, propagation of disturbances in the x direction is unlimited and occurs with an infinite speed in both positive- and negative-x directions.

Altogether, the above leads to the *weak ellipticity* property of the lubrication approximation, as illustrated in Fig. 5. Namely, the characteristics Ω of Eq. (5) allow for the propagation of disturbances from the point P in the y-direction with an infinite speed, while the characteristics $\tilde{\Omega}$ of Eqs. (9)

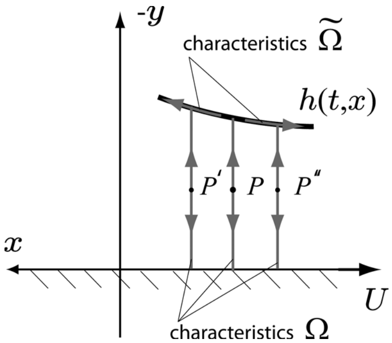

FIG. 5. On the weak ellipticity of the lubrication approximation: characteristics of Eqs. (9) and (19).

and (19) allow for the propagation of disturbances in the x-direction. Therefore, a disturbance from P can reach arbitrary points downstream P'' and upstream P' with an infinite speed analogously to a true (bulk) elliptic operator. It should be stressed, though, that the weak ellipticity property comes from the boundary conditions operator (19) in the problem with a parabolic bulk operator (5), and remains unchanged when passing to the steady case in Eq. (19).

D. Discussion

The above characteristics analysis does not change for coating flows over topography as the presence of the substrate curvature κ_{sub} modifies Eq. (19) to

$$h_t - \partial_x\left[\frac{h^3}{3}\partial_x(h_{xx} + \kappa_{\text{sub}})\right] = 0, \tag{22}$$

where the Galilean transformation $(t,x)\to(t,x-t)$ was also applied to bring the Landau–Levich equation to the standard form used for surface tension driven flows on curved substrates.[18–20] In addition to the Landau–Levich problem considered here, there are other situations when stagnation points arise in lubrication approximations: a couple of examples are given in Fig. 6. The first example—tip-streaming flow[21] in Fig. 6(a)—has a stagnation point in the bulk. It must be noted that the presence of a free boundary is not necessary for the lubrication approximation to capture a stagnation point, as illustrated by the problem of the flow near the nip of the co-rotating two-roll mill, cf. Fig. 6(b). In this case the net pressure gradient p_x across the nip is zero, i.e., $\int_{-\infty}^{+\infty} p_x \mathrm{d}x = 0$, but the pressure inside the nip is nonuniform and is found as a part of the solution as it adjusts itself to preserve the constraint of constant mass flux at each cross-section of the nip.

The analysis in Sec. II C also explains why the lubrication approximation works better than expected from equations of parabolic type in the situation such as a contact line or a flow over an edge shown in Fig. 1. In all these situations, the flow topology requires an elliptic operator to be accounted for—while the bulk operator in Eq. (5) is still parabolic, the overall weak elliptic character of the problem does allow the solution to exist and to capture elliptic points instead of blowing up. While the established weak ellipticity property is a necessary condition for the lubrication approximation to capture elliptic points, it does not allow the lubrication equations to capture elliptic behavior too far from the region of a thin film approximation, i.e., too far from the region where a thin-layer scaling applies. For example, in the context of the considered coating problems, the fact that the lubrication approximation is inapplicable far away from the wall follows from the fact that it cannot capture the second stagnation point present, for example, at the centerline in the Bretherton flow, cf. Fig. 3(b) or in dip-coating in confined geometries. In fact, in all known examples, where the lubrication approximation works, the elliptic points are localized in the otherwise global thin film approximation (stagnation points, contact lines, flows over the edge, etc.). However, while due to their weak ellipticity lubrication approximations are able to capture flows with elliptic behavior, they do not necessarily reflect the true behavior in a precise quantitative manner.[4] In the Landau–Levich problem though, the lubrication approximation for $\mathrm{Ca} \ll 1$ does conform with the true dynamics quantitatively, as was seen in Sec. II A.

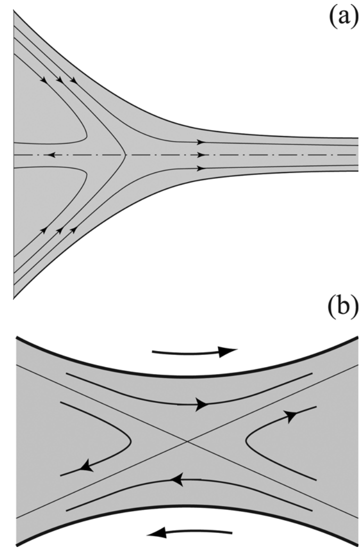

FIG. 6. Examples of flows with stagnation points studied with lubrication approximations: (a) tip-streaming phenomena (Ref. 21) and (b) flow near the nip of the corotating two-roll mill.

An interesting parallel can be drawn with boundary layer theory,[22,23] where the classical steady Prandtl boundary layer equations are parabolic, as we know from the characteristic analysis (15) [the neglected inertia terms in Eq. (5) are of lower order and thus do not contribute to the characteristic determinant]. Because of their parabolic character, the boundary layer equations cannot account for separation phenomenon, cf. Fig. 7, i.e., their solution blows up and thus cannot be continued beyond this point in a meaningful way[3,24] without modifying the assumption of a prescribed pressure. Indeed, the flow structure around a separation point, as in Fig. 7, clearly violates the built-in assumption of an almost unidirectional flow in the derivation of the boundary layer equations and requires an elliptic operator (i.e., nonreduced diffusion) to capture this structure. At the characteristics level, this is due to the fact that the boundary layer equations cannot allow for the propagation and thus the influence of disturbances upstream due to the parabolic char-

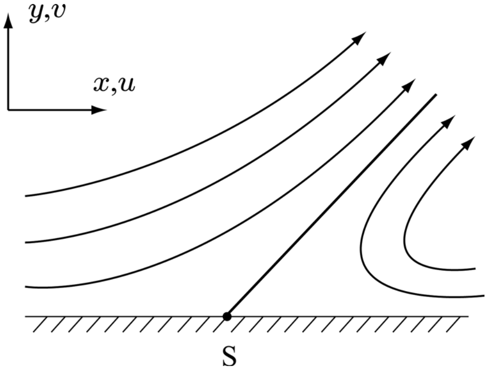

FIG. 7. Steady boundary layer streamlines near a separation point.

acter of the equations; however, experiments clearly demonstrate upstream influence: see discussion in Ref. 23. The contradiction is resolved by allowing the boundary layer and outer inviscid flow to interact through the boundary condition between them, which allows for adjustments of the pressure similar to the above free boundary problems and thus leads to weak ellipticity, i.e., disturbances propagate both upstream and downstream.

III. MARANGONI EFFECTS IN THE BEHAVIOR OF ELLIPTIC POINTS

With the above-introduced characteristic analysis of lubrication approximations, let us now understand how surfactant dynamics affects the characteristics properties of the lubrication approximation and the behavior of elliptic points.

A. Characteristic analysis of the surfactant interface case

To stay within the lubrication approximation, let us follow the work of Ratulowski and Chang[25] and consider the case of asymptotically small concentration of the surfactant γ,

$$\gamma \to \gamma_{\mathrm{m}} \mathrm{Ca}^{2/3} \tilde{\gamma}, \tag{23}$$

where γ_{m} is the saturation interfacial concentration. With Eq. (23), the tangential interfacial condition (7b) in the transition region is replaced with

$$\psi_{yy} = E \frac{\mathrm{d}\tilde{\gamma}}{\mathrm{d}s} + O(\mathrm{Ca}^{2/3}), \tag{24}$$

where the arclength s is scaled in the same way as x and $E = -\mathrm{d}\sigma/\mathrm{d}\gamma|_{\gamma=0}$ is the nondimensional *elasticity number*. Note that the elasticity number is positive, which can be seen from the derivative $\mathrm{d}\sigma/\mathrm{d}\gamma$ expressed in terms of the bulk concentration c, which is more suitable for the analysis of the interfacial material behavior,

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\gamma} = \frac{\mathrm{d}\sigma}{\mathrm{d}c}\frac{\mathrm{d}c}{\mathrm{d}\gamma} \quad \text{with} \quad \frac{\mathrm{d}\sigma}{\mathrm{d}c} < 0 \quad \text{and} \quad \frac{\mathrm{d}\gamma}{\mathrm{d}c} > 0. \tag{25}$$

Indeed, from the Szyszkowsky equation, $\sigma = \sigma - 2RT\gamma_{\mathrm{m}} \ln \times (1 + K_L c)$, and the Langmuir isotherm, $k_a c(1 - \gamma/\gamma_{\mathrm{m}}) - k_d \gamma = 0$, where R is the gas constant, T is the temperature, $K_L = k_a/(k_d \gamma_{\mathrm{m}})$ is the Langmuir constant, k_a and k_d are the adsorption and desorption coefficients, respectively, it follows that

$$\frac{\mathrm{d}\sigma}{\mathrm{d}c} = -\frac{2RT\gamma_{\mathrm{m}}}{1 + K_L c} = -2RT\gamma_{\mathrm{m}} K_L \left(1 + \frac{\gamma}{\gamma_{\mathrm{m}}}\right) < 0. \tag{26}$$

Also,

$$\frac{\mathrm{d}c}{\mathrm{d}\gamma} = \frac{k_d}{k_a}\left(1 + \frac{\gamma}{\gamma_{\mathrm{m}}}\right)^{-2} > 0, \tag{27}$$

which together with Eq. (26) produces

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\gamma} = -2RT\left(1 + \frac{\gamma}{\gamma_{\mathrm{m}}}\right)^{-1} < 0. \tag{28}$$

This behavior agrees (qualitatively) with the actual material properties of surfactants.[26]

As a result, the expression for the stream-function generalizes to

$$\psi = h_{xxx}\frac{y^2}{6}(y + 3h) + E\tilde{\gamma}_s \frac{y^2}{2} - y, \tag{29}$$

where we distinguish s- and x-derivatives for convenience, although in the transition region $\mathrm{d}s \simeq -\mathrm{d}x + O(\mathrm{Ca}^{2/3})$ to the leading order. It is interesting to note that, based on the meaning of the stream-function ψ in the steady case as a measure of the flow rate, the Marangoni term in Eq. (29) appears to provide an additional mass flux into the film if $E\tilde{\gamma}_s < 0$; however, one has to keep in mind that the dependence on Marangoni effects enters also through the modified dependence of h_{xxx}, which makes the flow rate ψ a nontrivial function of the Marangoni effects.

The classical Landau–Levich equation[9] is generalized for the surfactant presence case to

$$h_{xxx}h^2 + 3\left(1 - \frac{\alpha}{h}\right) + \frac{3}{2}E\tilde{\gamma}_s h = 0, \tag{30}$$

which is derived in the same fashion as Eq. (9). Its unsteady version is also derived as in Sec. II C and reads

$$h_t - h_x + E\partial_x(\tilde{\gamma}_x h^2/2) - \partial_x[h_{xxx}h^3/3] = 0. \tag{31}$$

At this point, it is necessary to invoke the analysis of the surfactant transport equation, which in the nondimensional form in the unsteady case can be written for the scaled interfacial concentration defined by Eq. (23) as

$$\frac{\partial\tilde{\gamma}}{\partial t} - \frac{\partial}{\partial x}\left[\tilde{\gamma}\left(1 - Eh\tilde{\gamma}_x + h_{xxx}\frac{h^2}{2}\right)\right] = \frac{1}{\mathrm{Pe}_s^*}\frac{\partial^2\tilde{\gamma}}{\partial x^2} + j^*(\tilde{\gamma},\tilde{c}) + O(\mathrm{Ca}^{2/3}), \tag{32}$$

where $\mathrm{Pe}_s^* = \mathrm{Ca}^{1/3}\mathrm{Pe}_s$ is the scaled Peclet number and $j^* = -\mathrm{Ca}^{1/3}[K_L c_{\mathrm{m}}\tilde{c} + \tilde{\gamma}]l_c k_d/U$ is the flux from the bulk to the interface. For the sake of brevity of the present characteristic analysis we keep the bulk concentration c constant—as we will show, this assumption does not affect the characteristic type of the system. In order to understand the characteristic type of the system of equations (31) and (32), in particular, the propagation of disturbances in this physical system, one needs to linearize around some steady state $[H(x), \Gamma(x)]$ by introducing the perturbation $h \to H(x)+h'(x,t)$ and $\tilde{\gamma} \to \tilde{\Gamma}(x)+\tilde{\gamma}'(x,t)$. The resulting linearized system is (dropping the tildes and primes)

$$h_t - h_x + E\partial_x\left[\gamma_x\frac{H^2}{2}+\Gamma_x Hh\right] - \partial_x\left[\frac{H^3}{3}h_{xxx}+H^2H_{xxx}h\right] = 0,$$

$$\gamma_t - \gamma_x + \partial_x\left[\gamma\left(EH\Gamma_x - \frac{H^2}{3}H_{xxx}\right)\right] + \partial_x\left[\Gamma\left(EH\gamma_x + E\Gamma_x h - \frac{H^2}{2}h_{xxx} - HH_{xxx}h\right)\right] = \frac{1}{\mathrm{Pe}_s^*}\tilde{\gamma}_{xx} + j^*(\gamma), \quad j^*(\gamma) = -\mathrm{Ca}^{1/3}\gamma l_c k_d/U,$$

the characteristic determinant of which is found by the transformation used Sec. II C, i.e., $(t,x)\to\tilde{\Omega}(t,x)$

$$\begin{pmatrix} -\frac{H^3}{3}\tilde{\Omega}_x^4 & E\frac{H^2}{2}\tilde{\Omega}_x^2 \\ -\Gamma\frac{H^2}{2}\tilde{\Omega}_x^4 & [\Gamma EH - \mathrm{Pe}_s^{*-1}]\tilde{\Omega}_x^2 \end{pmatrix}\begin{pmatrix} h^{(4)} \\ \gamma^{(2)} \end{pmatrix} = \text{l.o.t.},$$

where $h^{(4)}$ and $\gamma^{(2)}$ stand for the derivatives with respect to the characteristic coordinate $\tilde{\Omega}$, and the terms on the right-hand side contain lower order derivatives with respect to $\tilde{\Omega}$. When the determinant of the matrix on the left vanishes, this gives the characteristic condition

$$\tilde{\Omega}_x^6\frac{H^3}{3}\left(\frac{1}{\mathrm{Pe}_s^*} - \frac{1}{4}\Gamma EH\right) = 0. \quad (33)$$

Since $H>0$ and if the expression in brackets does not vanish, the characteristic surface $\tilde{\Omega}(t,x)$ is defined by $\tilde{\Omega}_x^6=0$, which implies that the system of equations (31) and (32) is parabolic and the corresponding characteristics are the surfaces $\tilde{\Omega}$ parallel to the x-axis. Therefore, following the analysis in Sec. II C, we conclude that the system of lubrication equations (31) and (32) possesses the weak ellipticity property.

While in the above argument, we kept the bulk concentration c constant, this does not affect the results as the bulk concentration obeys the convection-diffusion equation, which is parabolic and thus the whole coupled system with varying h, γ, and c remains parabolic. As we established here, Marangoni effects do not affect the weak ellipticity of the lubrication approximation, since the system of coupled Landau–Levich and surfactant transport equations is still of parabolic character. Therefore, one should be able to capture the stagnation point at the surfactant-laden interface within a lubrication approximation similar to the clean interface case; this intuition is also confirmed by the analysis below.

B. Behavior of the stagnation point

In the asymptotic analyses of surfactant effects in the Landau–Levich problem,[25,27,28] there are two scalings for the interfacial concentrations that admit self-consistent asymptotic lubrication approximations, namely, the case when the equilibrium concentration and its variations are of the order of $\mathrm{Ca}^{2/3}$ (so-called trace amounts of surfactants[25]) and the case when the concentration deviates from its equilibrium value γ_0 in the bath on the order of $\mathrm{Ca}^{2/3}$, cf. works of Stebe and Barthès-Biesel[27] and Park.[28] Below we consider each case separately.

1. Case 1: Trace amounts of surfactant

In this case, the surfactant at the interface and thus in the bulk (as a consequence of the Langmuir isotherm) is present in trace amounts, cf. Eq. (23). Due to the tangential boundary condition (24) modified to account for surface tension gradients, the precise expression for interfacial velocity $u_{(s)}$ derived from Eq. (29) becomes

$$u_{(s)} = \frac{3}{2}\left(1-\frac{\alpha}{h}\right) - 1 + \frac{1}{4}E\tilde{\gamma}_s h. \quad (34)$$

The fact that the interfacial velocity changes its sign is independent of the presence of Marangoni effects and thus clearly there must be at least one stagnation point at the interface contrary to the idea of the paper[29] that it can move into the bulk.

Thus the equation for zeros of $u_{(s)}$ is quadratic in h

$$E\tilde{\gamma}_s\frac{h^2}{4} - \frac{h}{2} + \frac{3}{2}\alpha = 0 \Rightarrow h_{1,2} = \frac{1\mp\sqrt{1-6\alpha E\tilde{\gamma}_s}}{E\tilde{\gamma}_s}. \quad (35)$$

Since the elasticity number E is positive (cf. discussion in Sec. III A), there are two options:

(1) $\tilde{\gamma}_s<0$: one stagnation point because the only physically meaningful solution is $h_1=(1-\sqrt{1-6\alpha E\tilde{\gamma}_s})/(E\tilde{\gamma}_s)$ since $h>0$. If $|E\tilde{\gamma}_s|\ll 1$, which corresponds to negligible Marangoni effects, then $h_1\simeq 3\alpha$.

(2) $\tilde{\gamma}_s>0$: two stagnation points as long as $E\tilde{\gamma}_s<1/(6\alpha)$. If $E\tilde{\gamma}_s\ll 1$, then $h_1\simeq 3\alpha$, which is the same as in the clean interface case, and $h_2\simeq 2/(E\tilde{\gamma}_s)>12\alpha$. However, the presence of this second stagnation point is not physically feasible in the Landau–Levich problem in view of the topological impossibility, cf. Fig. 8. Indeed, the velocity fields at the interface in the film and the bath have opposite directions in the interfacial arclength s coordinate as shown in Fig. 8. Therefore, the velocity field in the region between the streamlines A and B terminating at the two stagnation points, indicated in the same figure, is not feasible: the velocity along the vortex streamline C would have to change its sign, which is impossible.

2. Case 2: Small variation of surfactant concentration

In this case, i.e., when the dimensional surfactant concentration γ_0 in the bath away from the wall is such that $\gamma_0/\gamma_{\mathrm{m}}\sim O(1)$, the appropriate scaling should be with respect to its equilibrium value γ_0. Therefore, in the case of asymp-

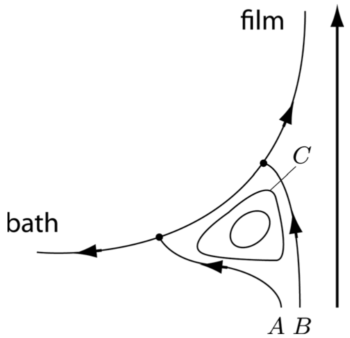

FIG. 8. Topological impossibility of the presence of two stagnation points in the Landau–Levich problem. The velocity fields at the interface in the film and the bath have the opposite directions, and thus the velocity field between the stagnation points, marked by dots in the figure, is not feasible unless there is a third stagnation point between these two, which is also impossible in view of Eq. (35).

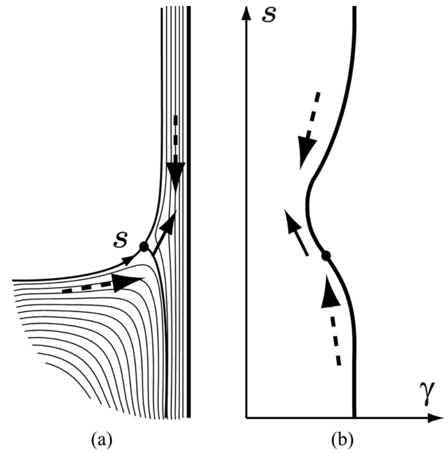

FIG. 9. Marangoni effects in the Landau–Levich problem: (a) Marangoni flows and (b) surfactant distribution. Dot designates the location of the stagnation point. Dashed and solid arrows are Marangoni stresses leading to film thinning and thickening, respectively. Note that the leftmost Marangoni stresses (dashed arrow) push the separating streamline, which emanates from the stagnation point, and thus reduce the mass flow into the film.

totically small deviations of the surfactant concentration γ from the equilibrium one γ_0, cf. the work of Stebe and Barthès-Biesel,[27] we find

$$\gamma \to \gamma_0(1 - \mathrm{Ca}^{2/3}\tilde{\gamma}), \tag{36}$$

which allows the balance of viscous and Marangoni stresses in Eq. (24). As a result, the reduced tangent dynamic boundary condition (24) has the same form as in the previous case if the elasticity number is defined as $E = \mathrm{d}\sigma/\mathrm{d}\gamma|_{\gamma=1}$, which is negative now. Indeed, Eq. (28) gives $\mathrm{d}\sigma/\mathrm{d}\gamma|_{\gamma=1} < 0$.

Since the only change compared to the previous case is the sign of the elasticity number, then the behavior of the stagnation point is reversed compared to the previous case:

(1) $\tilde{\gamma}_s < 0$: two stagnation points as long as $E\tilde{\gamma}_s < 1/(6\alpha)$. If $E\tilde{\gamma}_s \ll 1$, then $h_1 \simeq 3\alpha$, which is the same as in the clean interface case, and $h_2 \simeq 2/(E\tilde{\gamma}_s) > 12\alpha$. However, the presence of this second stagnation point is not physically feasible in the Landau–Levich problem in view of the topological impossibility, cf. Fig. 8, analogously to the case of trace amounts of surfactants.

(2) $\tilde{\gamma}_s > 0$: one stagnation point because the only physically meaningful solution is $h_1 = (1 - \sqrt{1 - 6\alpha E\tilde{\gamma}_s})/(E\tilde{\gamma}_s)$ in view of $h > 0$. If $|E\tilde{\gamma}_s| \ll 1$, then $h_1 \simeq 3\alpha$.

3. Discussion

While the value of the factor α is affected by Marangoni stresses, its value must be $O(1)$ for the lubrication assumption to remain valid; also, all known experimental observations, e.g., by Groenveld[30] and Krechetnikov and Homsy,[12] report $\alpha = O(1)$. Because $\alpha = O(1)$, the physically realizable stagnation points are always at a finite distance from the wall, which is of the order of the film thickness. Also, apparently, there are no physical conditions under which the stagnation point at the interface disappears. The latter observation is critical for the subsequent analysis in Sec. III C.

Notably, in none of the above cases does the location of the stagnation point go to infinity, $y \to -\infty$, i.e., the expectations of the works[29,31] cannot be justified.

C. Application: A necessary condition for film thickening in the Landau–Levich problem

As motivated by our newly gained understanding of the ability of the lubrication approximation to capture stagnation points and their behavior in the Landau–Levich problem, it is worth revisiting the long-standing question on surfactant effects in this classical problem, which was originally studied in the lubrication approximation.[25,28] While surfactant effects are usually deemed responsible for the experimentally observed film thickening phenomenon,[30] i.e., film turns out to be thicker compared to the one in the clean interface analysis of Landau and Levich,[9] there is no general firm justification of this assertion. Namely, in the regime when asymptotic analyses[25,28] are not applicable,[32] the experimental[12] and numerical[11] studies have demonstrated that Marangoni stresses cannot explain the experimentally observed film thickening. In particular, as illustrated in Fig. 9, the distribution of surfactant concentration relative to the stagnation point is such that there are two Marangoni flows (dashed arrows: see interpretation in the caption of Fig. 9), which contribute to film thinning, and one Marangoni flow (solid arrow), which thickens the film. As shown numerically[11] in the Stokes flow approximation, if the bulk concentration is maintained constant, which takes place for high concentrations of surfactant, then the Marangoni thinning stresses dominate and thus the net effect in the framework of the conventional macroscopic interfacial conditions should be film thinning. It is the goal here to establish a necessary condition for film thickening to occur due to Marangoni effects by analytical means. This will clarify the role of surfactants in the film thickening, the explanation of which is still an open problem.[11,12] In this work, we consider surfactant effects in the steady Landau–Levich problem in the case of soluble surfactants only, as the fact of the presence of a stagnation point at the interface makes it impossible for an insoluble surfactant to affect the dynamics (contrary to the analysis in Refs. 28 and 33).

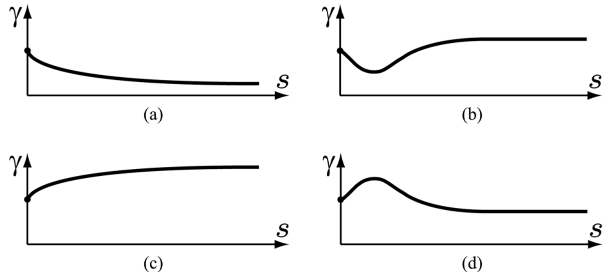

FIG. 10. All possible interfacial surfactant concentration distribution $\gamma(s)$ after the stagnation point (dot on the graph).

Let us go back to the original nondimensional concentration scaled with respect to the saturation value $\gamma_{\rm m}$, i.e., with no assumption on its smallness, and consider the steady case of its transport

$$\frac{d}{ds}(\gamma u_{(s)}) = \frac{1}{\text{Pe}_s}\frac{d^2\gamma}{ds^2} + j, \tag{37}$$

where Pe_s is the Peclet number and j is the flux from the bulk to the interface. Integrating this equation along the interface from arbitrary s to $s=\infty$,

$$-\gamma_\infty - \gamma(s)u_{(s)}(s) = -\frac{1}{\text{Pe}_s}\frac{d\gamma}{ds} + \int_s^\infty j(\tilde{s})d\tilde{s}, \tag{38}$$

and evaluating at the point where the interfacial velocity vanishes, furnishes the result

$$\frac{d\gamma}{ds} = \text{Pe}_s\left[\gamma_\infty + \int_s^\infty j(\tilde{s})d\tilde{s}\right], \tag{39}$$

where γ_∞ is the nondimensional interfacial concentration in the film away from the meniscus.

Obviously, based on the generic distribution of Marangoni stresses, cf. Figs. 9 and 10(b), one needs $d\gamma/ds<0$ at the stagnation point in order to get Marangoni stresses contributing to film thickening (solid arrow). The latter can be achieved only if the total flux—the second term on the right-hand side of Eq. (39)—is negative (i.e., cumulative surfactant net flux in the region after the stagnation point, $(s,+\infty)$, is from the interface into the bulk) and outweighs γ_∞ as in Fig. 10(b). This is, obviously, possible only if there can be a desorption of surfactant from the interface into the bulk, i.e., the bulk surfactant concentration cannot be constant. To prove that the generic concentration distribution in Fig. 10(b) is the only possible one, let us consider all other potential surfactant distributions shown in Fig. 10. Because interface stretching takes place only in the meniscus region, there is only one local extremum of $\gamma(s)$ in the meniscus region, i.e., it occurs before or after the stagnation point provided that the bulk concentration dynamics is governed only by diffusion, advection, and kinetic exchange with the interface. The situation in Fig. 10(c) cannot lead to film thickening because the Marangoni stresses in this case contribute to film thinning. Cases (a) and (d) are nonphysical since one cannot achieve higher interfacial concentrations in the region of interfacial stretching, i.e., in the meniscus and thus in the neighborhood of the stagnation point. Thus, the only option left is (b), which was also found numerically,[11] and which is generic in the sense that it does not depend on the details of the bulk concentration dynamics. Thus, logically, the necessary condition for film thickening within the framework of the standard hydrodynamic models is to allow for a variation of the surfactant concentration in the bulk. If the bulk concentration is lower in the thin film region than in the bath, in particular, $\gamma_\infty<1$, this weakens the film thinning Marangoni stresses and opens an opportunity for the film to thicken. Notably, the necessary condition for film thickening to occur due to Marangoni stresses established above does not rely upon the fact of the lubrication approximation and is valid in general, as follows from the analysis which led to Eq. (39).

IV. CONCLUSIONS

First, with the help of classical characteristics analysis, we have established the property of weak ellipticity of the lubrication approximation, which justifies the qualitative applicability of the latter to thin film flows with stagnation points. This explains how the performance of the lubrication approximation may exceed expectations in situations where the assumption of an almost unidirectional flow is patently invalid (e.g., flows over edges, cavities, and with contact lines).

Second, we also have developed the characteristics analysis in the surfactant interface case and analyzed the behavior of elliptic (stagnation) points within the lubrication approximation. In particular, based on the newly derived exact expressions for the interfacial velocity and surfactant flux in the surfactant-laden Landau–Levich problem, we proved rigorously that the stagnation point can neither go to infinity nor can it reside in the bulk. This new analysis also led to a necessary condition for the film thickening to occur due to Marangoni stresses.

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APPENDIX: ON COMPUTING CHARACTERISTIC DETERMINANTS

The discussion here follows Sec. III in Petrovsky[15] with the notation adopted to the present needs. The characteristic surface $\Omega(\boldsymbol{x})$ in the space of independent variables $\boldsymbol{x}$ is defined in the context of proving the Cauchy–Kovalevskaya theorem, namely, an initial value (Cauchy) problem has no unique solution, and thus is ill-posed, if the initial data are given at $\Omega(\boldsymbol{x})$. This allows a straightforward determination of the characteristic surface by making a transformation from the original independent variables $\boldsymbol{x}$ to $\Omega(\boldsymbol{x})$, in which resolution of the governing partial differential equation(s) for the highest order derivatives of the solution becomes impossible.[15] For example, in the case of the wave equation, $u_{tt}-c^2u_{xx}=0$, the transformation of independent variables $\boldsymbol{x}=(t,x)$ to the characteristic one $\Omega(t,x)$ yields $(\Omega_t^2$

$-c^2\Omega_x^2)u_{\Omega\Omega}+\text{l.o.t.}=0$, where only the highest order derivative terms are shown as they define the characteristic type of the equation. Thus the characteristics, defined by the undetermined highest order derivative $u_{\Omega\Omega}$, are given by the solutions of the vanishing characteristic determinant $\Omega_t^2-c^2\Omega_x^2=0$, i.e., $\Omega_t-c\Omega_x=0$ and $\Omega_t+c\Omega_x=0$, which are the familiar straight lines $x\pm ct=\text{const}$ responsible for the wave propagation in both positive and negative x directions with the speed c. This is reflected in the general form of the solution for the wave equation $u(t,x)=f(x+ct)+g(x-ct)$, where f and g are general functions.

In general, suppose we are given a system of nonlinear partial differential equations of dimension N,

$$\mathbf{\Phi}(\boldsymbol{x},\boldsymbol{u},\boldsymbol{u}_\sigma)=\mathbf{0}, \tag{A1}$$

where $\mathbf{\Phi}=(\Phi_1,\ldots,\Phi_N)$, $\boldsymbol{u}=(u^1,\ldots,u^N)$, and $\boldsymbol{x}=(x_1,\ldots,x_n)$ represent dependent and independent variables respectively, and the partial derivatives $\boldsymbol{u}_\sigma$ of the order of σ are given explicitly by

$$u_\sigma^j=\frac{\partial^{|\sigma|}u_j}{\partial x_1^{k_1}\ldots\partial x_n^{k_n}}, \quad |\sigma|\le n_j, \tag{A2}$$

with $\sigma=(k_1,\ldots,k_n)$ being the multi-index, $|\sigma|=k_1+\cdots+k_n$, and n_{ij} the highest order of the derivative of the variable u^j in the ith equation $\Phi_i=0$. Then the characteristic determinant for the system (A1) is[15]

$$\left|\sum_{\sigma=n_j}\frac{\partial\Phi_i}{\partial u_\sigma^j}\left(\frac{\partial\Omega}{\partial x_1}\right)^{k_1}\cdots\left(\frac{\partial\Omega}{\partial x_n}\right)^{k_n}\right|=0, \tag{A3}$$

where only the (i,j)-element of the determinant matrix is shown. Equation (A3) defines the characteristic surface $\Omega(\boldsymbol{x})$. Classification of characteristic types of Eq. (A1) at a particular point $\boldsymbol{x}$ is based on the type of solutions $\Omega(\boldsymbol{x})$ of Eq. (A3). Namely, if characteristic surfaces $\Omega(\boldsymbol{x})$ are all real and nondegenerate, then Eq. (A1) is hyperbolic; if characteristics are real and degenerate, then Eq. (A1) is parabolic; if there are no real characteristics, then Eq. (A1) is elliptic. As a test, the reader may want to apply Eq. (A3) to the wave equation in its original form $u_{tt}-c^2u_{xx}=0$ and in the form of the system $u_t=v$, $v_t=c^2u_{xx}$.

In the case of the system (5), we have $\boldsymbol{x}=(x,y)$, $\boldsymbol{u}=(u,v,p)$, and

$$\mathbf{\Phi}=\begin{pmatrix}u_x+u_y\\ p_x-u_{yy}\\ -p_y\end{pmatrix}. \tag{A4}$$

Thus, application of Eqs. (A3) and (5a)–(5c) produces the determinant (16) if the pressure p is an unknown function and Eq. (15) if the pressure p is a given function as is common in boundary layer theory.[3]

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