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Effect of surfactants on the long-wave stability of oscillatory film flow

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Effects of insoluble surfactants on the stability of film flow driven by an oscillatory plate are investigated in the limit of long-wavelength perturbations. Two particular Floquet modes are identified and the corresponding growth rates are obtained by solving a quadratic equation. Results show that the oscillatory film flow can be stabilized by surface surfactant in the sense of raising the critical Froude number and narrowing the bandwidths of the unstable frequencies.

1. Introduction

The stability of film flow adjacent to walls is of considerable importance in a variety of problems, such as coating, crystal growth and materials processing. However, unsteadiness usually occurs and may not be neglected. Some relevant topics, e.g. stability of a fluid layer driven by an oscillating plane, and stability of steady film flow with surfactant, have been investigated and are briefly reviewed below.

Linear stability of a fluid layer driven by an oscillating plane was first studied theoretically by Yih (1968), who proposed an analysis for two-dimensional, long-wavelength disturbances based on Floquet theory. He found that the stability of the flow depends on the Froude number and the frequency of oscillation, and the long-wavelength instability can only exist for certain separated bandwidths of the frequency. Or (1997) extended the analysis to investigate the same problem with arbitrary wavenumbers and found that finite-wavelength instability occurs once the imposed frequency exceeds a certain threshold. Further, Or & Kelly (1998) studied the effects of both wall oscillation and themocapillarity on the instability of a fluid layer.

The stability of steady film flows in the presence of surfactants has been investigated extensively. Insoluble surfactants may have either a stabilizing or a destabilizing effect. In surfactant-laden flow down an inclined plane, Whitaker & Jones (1966) and Lin (1970) found that the critical Reynolds number associated with the Yih mode for long-wave instability increases with surfactant elasticity, indicating a stabilizing influence of surfactants. In the limit of Stokes flow, Pozrikidis (2003) identified another Marangoni mode due to the presence of surfactant, which is always damped even though inertial effects are considered (Blyth & Pozrikidis 2004*a*). When an interfacial shear is imposed, a falling film or a two-fluid flow system may be destabilized by surfactants (Frenkel & Halpern 2002; Halpern & Frenkel 2003; Blyth & Pozrikidis 2004*b*; Wei 2005*a*). A unified view of the mechanisms of Marangoni effects was proposed by Wei (2005*b*).

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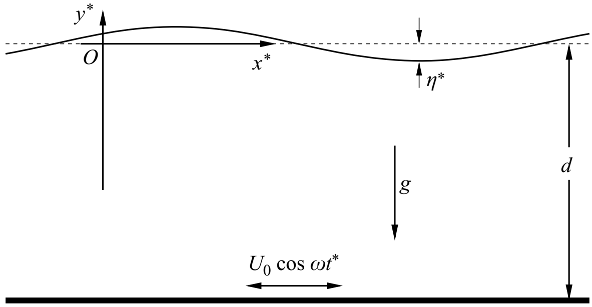

FIGURE 1. Schematic of the flow configuration.

Little work has been performed on the stability of unsteady film flows with insoluble surfactants to our knowledge. The main aim of the present paper is to study the effect of surfactants on the long-wave stability of oscillatory film flow. Although we recognize the limitation of the long-wave stability analysis (Or 1997) we nevertheless feel that the analysis will be of help in revealing the complicated stability characteristics.

2. Flow configuration and the stability problem

Consider a horizontal layer of incompressible Newtonian fluid with density ρ and viscosity μ on an infinite flat plate, as shown in figure 1. We use an asterisk to denote dimensional variables. The plate located at $y^* = -d$ oscillates in the x^*-direction with velocity $U_0 \cos \omega t^*$, where ω is the modulation frequency and U_0 is the amplitude. The upper free surface of the film is described by $y^* = \eta^*(x^*, t^*)$, and is covered by a monolayer of insoluble surfactant. Let u^* and v^* be the velocity components in the horizontal and vertical directions, respectively. As in Halpern & Frenkel (2003), the surfactant concentration $\Gamma^*(x^*, t^*)$ obeys the transport equation

$$\frac{\partial (H\Gamma^*)}{\partial t^*} + \frac{\partial}{\partial x^*}(H\Gamma^* u^*) = D_s \frac{\partial}{\partial x^*}\left(\frac{1}{H}\frac{\partial \Gamma^*}{\partial x^*}\right), \tag{2.1}$$

where $H = \sqrt{1 + \eta_{x^*}^{*2}}$ and D_s is the surfactant diffusivity, which is usually negligible and is discarded below. For the linear stability problem considered, the relation between the surface tension γ^* and the surfactant concentration Γ^* can be approximated as $\gamma^* = \gamma_0 - E(\Gamma^* - \Gamma_0)$, where E is the surface elasticity, and Γ_0 is the basic value of the surfactant concentration, corresponding to a uniform surface tension γ_0.

The motion of the fluid is governed by the Navier–Stokes equation and the continuity equation, together with the no-slip and no-penetration boundary conditions at the wall. At the free surface, the kinematic condition $\partial \eta^*/\partial t^* = v^* - u^* \partial \eta^*/\partial x^*$ and the dynamic condition requires a balance among the hydrodynamic traction, the surface tension and the Marangoni traction (e.g. Halpern & Frenkel 2003; Pozrikidis 2003; Blyth & Pozrikidis 2004*a*). We choose U_0 as the characteristic scale of velocity, the mean thickness of the film d as the scale of length, ω^{-1} as the scale of time, ρU_0^2 as the scale of pressure. The surfactant concentration and surface tension are normalized by Γ_0 and γ_0, respectively. Then, the dimensionless form of the basic velocity profile is given by

$$U(y, t) = \mathrm{Re}\left[\frac{\cosh By}{\cosh B} \mathrm{e}^{\mathrm{i}t}\right], \tag{2.2}$$

where $B=(1+\mathrm{i})\beta$ and $\beta=\sqrt{\rho\omega d^2/2\mu}$ is the ratio of the mean thickness of the film to the thickness of the Stokes layer induced by wall oscillation.

Since only two-dimensional perturbations are considered, we introduce a disturbance streamfunction $\psi'(x, y, t)$, related to the velocity perturbations (u', v') by $u'=\psi'_y$, $v'=-\psi'_x$. The surfactant concentration Γ is perturbed as $\Gamma(x,t)=1+\Gamma'(x,t)$. Since the basic state is independent of x, the disturbances ψ', Γ' and the position of the perturbed surface η can be assumed to be of the form

$$[\psi'(x, y, t), \Gamma'(x, t), \eta(x, t)] = \varepsilon[\phi(y, t), \xi(t), h(t)]\mathrm{e}^{\mathrm{i}kx} + \text{complex conjugate}, \tag{2.3}$$

where $|\varepsilon| \ll 1$, k is real and denotes the streamwise wavenumber. Substituting (2.3) into the governing equations and linearizing, we obtain the time-dependent Orr–Sommerfeld equation

$$\left(2\beta^2\frac{\partial}{\partial t} + \mathrm{i}kRU\right)\left(\frac{\partial^2}{\partial y^2} - k^2\right)\phi - \mathrm{i}kRU_{yy}\phi = \left(\frac{\partial^2}{\partial y^2} - k^2\right)^2\phi, \tag{2.4}$$

where $R=\rho U_0 d/\mu$ is the Reynolds number. The boundary conditions at the wall $y=-1$ satisfy

$$\phi = \frac{\partial \phi}{\partial y} = 0. \tag{2.5}$$

The linearized conditions for the normal and tangential stresses at $y = 0$ are, respectively,

$$2\beta^2\frac{\partial^2\phi}{\partial t\partial y} - \left(\frac{\partial^2}{\partial y^2} - 3k^2 - \mathrm{i}kRU\right)\frac{\partial\phi}{\partial y} + \mathrm{i}k\left(F^{-2}R + \frac{k^2}{Ca}\right)h = 0, \tag{2.6}$$

$$U_{yy}h + \frac{\partial^2\phi}{\partial y^2} + k^2\phi + \frac{Ma}{Ca}\mathrm{i}k\xi = 0, \tag{2.7}$$

where the Froude number F, the Marangoni number Ma and the capillary number Ca are defined as $F^{-2} = gd/U_0^2$, $Ma = E\Gamma_0/\gamma_0$ and $Ca = \mu U_0/\gamma_0$. The linearized kinematic boundary condition and transport equation for surfactant are expressed by

$$2\beta^2\frac{\mathrm{d}h}{\mathrm{d}t} + \mathrm{i}kRUh + \mathrm{i}kR\phi = 0, \tag{2.8}$$

$$2\beta^2\frac{\mathrm{d}\xi}{\mathrm{d}t} + \mathrm{i}kRU\xi + \mathrm{i}kR\frac{\partial\phi}{\partial y} = 0, \tag{2.9}$$

where ϕ, $\partial\phi/\partial y$ and U are evaluated at $y=0$.

The Floquet system (2.4) to (2.9) governs the linear stability problem. For finite-wavelength instabilities, the differential system should be solved numerically, while long-wavelength solutions can be analytically obtained by an expansion in k and will be discussed in the following.

3. Long-wavelength stability analysis

Considering the limit of long waves, i.e. $k \ll 1$, the disturbances are assumed as

$$\phi(y, t) = \mathrm{e}^{\mu t}[\phi_0(y, t) + k\phi_1(y, t) + k^2\phi_2(y, t) + \cdots], \tag{3.1a}$$

$$h(t) = \mathrm{e}^{\mu t}[h_0(t) + kh_1(t) + k^2h_2(t) + \cdots], \tag{3.1b}$$

$$\xi(t) = \mathrm{e}^{\mu t}[\xi_0(t) + k\xi_1(t) + k^2\xi_2(t) + \cdots], \tag{3.1c}$$

$$\mu = \mu_0 + k\mu_1 + k^2\mu_2 + \cdots, \tag{3.1d}$$

in which μ is the complex growth rate of the disturbance, ϕ_j, h_j and ξ_j ($j = 0, 1, 2, \ldots$) are 2π-periodic in time. Substituting these expansions into (2.4) to (2.9), we obtain a sequence of problems at each order of k. The purpose of this procedure is to find the first non-zero μ_j, corresponding to exponential growth or decay of the disturbance.

At the leading order, $O(1)$, the kinematic condition and the transport equation are

$$\frac{\mathrm{d}h_0}{\mathrm{d}t} + \mu_0 h_0 = 0, \qquad \frac{\mathrm{d}\xi_0}{\mathrm{d}t} + \mu_0 \xi_0 = 0. \tag{3.2}$$

The constraint that h_0 and ξ_0 are periodic in t leads to

$$\mu_0 = 0, \quad h_0 = \text{const}, \quad \xi_0 = \text{const}. \tag{3.3}$$

Another possibility, $\mu_0 \neq 0$ and $h_0 = \xi_0 = 0$, corresponds to damped Floquet modes as demonstrated in Yih (1968), and is not of interest here. Then the leading-order system for ϕ_0 becomes

$$2\beta^2 \frac{\partial^3 \phi_0}{\partial t \partial y^2} = \frac{\partial^4 \phi_0}{\partial y^4}, \tag{3.4}$$

$$\phi_0(-1, t) = \frac{\partial \phi_0}{\partial y}(-1, t) = 0, \tag{3.5a, b}$$

$$\frac{\partial^3 \phi_0}{\partial y^3}(0, t) = 2\beta^2 \frac{\partial^2 \phi}{\partial t \partial y}(0, t), \quad U_{yy}(0, t) h_0 + \frac{\partial^2 \phi_0}{\partial y^2}(0, t) = 0. \tag{3.5c, d}$$

This system admits the periodic solution

$$\phi_0 = h_0 \mathrm{Re}\left[\frac{1 - \cosh B(y+1)}{\cosh^2 B} \mathrm{e}^{\mathrm{i}t}\right]. \tag{3.6}$$

The associated flow field is identical with that of Yih (1968). It is shown that the surface surfactant does not affect the velocity distribution at this order.

For the first-order approximation, $O(k)$, (2.8) and (2.9) give

$$2\beta^2 \left(\frac{\mathrm{d}h_1}{\mathrm{d}t} + \mu_1 h_0\right) + \mathrm{i}RU(0, t) h_0 + \mathrm{i}R\phi_0(0, t) = 0, \tag{3.7}$$

$$2\beta^2 \left(\frac{\mathrm{d}\xi_1}{\mathrm{d}t} + \mu_1 \xi_0\right) + \mathrm{i}RU(0, t)\xi_0 + \mathrm{i}R\frac{\partial \phi_0}{\partial y}(0, t) = 0. \tag{3.8}$$

Since ϕ_0 and U_0 are time periodic with a zero average, to obtain periodic solutions $h_1(t)$ and $\xi_1(t)$, we must have

$$\mu_1 = 0, \tag{3.9}$$

$$h_1 = -\frac{Rh_0}{B^2} \mathrm{Im}\left[\frac{1}{\cosh^2 B} \mathrm{e}^{\mathrm{i}t}\right], \tag{3.10}$$

$$\xi_1 = -\frac{R\xi_0}{B^2} \mathrm{Im}\left[\frac{1}{\cosh B} \mathrm{e}^{\mathrm{i}t}\right] + \frac{Rh_0}{B^2} \mathrm{Im}\left[\frac{B \sinh B}{\cosh^2 B} \mathrm{e}^{\mathrm{i}t}\right]. \tag{3.11}$$

The differential system for ϕ_1 contains inhomogeneous terms which are products of functions having a time dependence given by $\mathrm{e}^{\pm \mathrm{i}t}$. This leads to $\phi_1(y, t) = \phi_1^{(S)}(y) + \hat{\phi}_1(y)\mathrm{e}^{2\mathrm{i}t} + \check{\phi}_1(y)\mathrm{e}^{-2\mathrm{i}t}$ with the superscript, (S), denoting the steady part. As will be shown later, it is sufficient to solve for $\phi_1^{(S)}(y)$ to obtain μ_2 and hence to determine the stability of the flow. The corresponding time-independent system is

$$\frac{\mathrm{d}^4\phi_1^{(S)}}{\mathrm{d}y^4} = \mathrm{i}Rh_0\mathrm{Re}\left[\frac{2\cosh By\cos B(y+1) - \cosh By}{2B^{-2}\cos^2 B\cosh B}\right], \tag{3.12}$$

$$\phi_1^{(S)}(-1) = \frac{\mathrm{d}\phi_1^{(S)}}{\mathrm{d}y}(-1) = 0, \tag{3.13a,b}$$

$$\frac{\mathrm{d}^3\phi_1^{(S)}}{\mathrm{d}y^3}(0) - \mathrm{i}F^{-2}Rh_0 - \mathrm{i}Rh_0\mathrm{Re}\left[\frac{B\sin B}{2\cos^2 B\cosh B}\right] = 0, \tag{3.13c}$$

$$\frac{\mathrm{d}^2\phi_1^{(S)}}{\mathrm{d}y^2}(0) + \mathrm{i}Rh_0\mathrm{Re}\left[\frac{1}{2\cos^2 B\cosh B}\right] + \frac{Ma}{Ca}\mathrm{i}\xi_0 = 0. \tag{3.13d}$$

The solution to this system is represented by

$$\phi_1^{(S)}(y) = \phi_{1c}(y) - \mathrm{i}Rh_0\mathrm{Re}\left[\frac{2\cosh By + \cosh By\cos B(y+1)}{4B^2\cos^2 B\cosh B}\right], \tag{3.14a}$$

in which ϕ_{1c} is the complementary solution

$$\phi_{1c}(y) = A_0 + A_1 y + A_2 y^2 + A_3 y^3. \tag{3.14b}$$

Here these four coefficients can be determined by the boundary conditions and are expressed as

$$A_0 = -\tfrac{1}{3}\mathrm{i}RF^{-2}h_0 - \frac{Ma}{2Ca}\mathrm{i}\xi_0 + \mathrm{i}Rh_0\mathrm{Re}\left[\frac{3\cosh B - 3B\sinh B}{4B^2\cos^2 B\cosh B}\right],$$
$$A_1 = -\tfrac{1}{2}\mathrm{i}RF^{-2}h_0 - \frac{Ma}{Ca}\mathrm{i}\xi_0 - \mathrm{i}Rh_0\mathrm{Re}\left[\frac{3\sinh B}{4B\cos^2 B\cosh B}\right],$$
$$A_2 = -\frac{Ma}{2Ca}\mathrm{i}\xi_0, \qquad A_3 = \tfrac{1}{6}\mathrm{i}F^{-2}Rh_0.$$

Further, considering the $O(k^2)$ problem, the kinematic condition and the surfactant transport equation can be written as

$$2\beta^2\left(\frac{\mathrm{d}h_2}{\mathrm{d}t} + \mu_2 h_0\right) + \mathrm{i}RU(0,t)h_1 + \mathrm{i}R\phi_1(0,t) = 0, \tag{3.15}$$

$$2\beta^2\left(\frac{\mathrm{d}\xi_2}{\mathrm{d}t} + \mu_2\xi_0\right) + \mathrm{i}RU(0,t)\xi_1 + \mathrm{i}R\frac{\partial\phi_1}{\partial y}(0,t) = 0. \tag{3.16}$$

Since $h_2(t)$ and $\xi_2(t)$ are periodic in t, it follows that

$$2\beta^2 R^{-1}\mu_2 h_0 = -\mathrm{i}\left[U(0,t)h_1 + \phi_1(0,t)\right]^{(S)}, \tag{3.17}$$

$$2\beta^2 R^{-1}\mu_2\xi_0 = -\mathrm{i}\left[U(0,t)\xi_1 + \frac{\partial\bar{\phi}_1}{\partial y}(0,t)\right]^{(S)}. \tag{3.18}$$

From (2.2), (3.10), (3.11) and (3.14), we obtain

$$\left[U(0,t)h_1 + \phi_1(0,t)\right]^{(S)} = -\tfrac{1}{3}\mathrm{i}F^{-2}Rh_0 - \frac{Ma}{2Ca}\mathrm{i}\xi_0 + \mathrm{i}Rh_0 I_1, \tag{3.19}$$

$$\left[U(0,t)\xi_1 + \frac{\partial\bar{\phi}_1}{\partial y}(0,t)\right]^{(S)} = -\tfrac{1}{2}\mathrm{i}F^{-2}Rh_0 - \frac{Ma}{Ca}\mathrm{i}\xi_0 + \mathrm{i}Rh_0 I_2, \tag{3.20}$$

in which

$$I_1 = \mathrm{Re}\left[\frac{3\cosh B - 3B\sinh B}{4B^2\cos^2 B\cosh B}\right], \quad I_2 = \mathrm{Re}\left[\frac{3\sin B - 3\sinh B}{4B\cos^2 B\cosh B}\right]. \tag{3.21}$$

Note that I_1 and I_2 only depend on β. Upon substituting (3.19) and (3.20) into (3.17) and (3.18), we obtain two equations related to μ_2, which can be written in the matrix form

$$\begin{pmatrix} I_1 - \frac{1}{3}F^{-2} & -\frac{1}{2}M \\ I_2 - \frac{1}{2}F^{-2} & -M \end{pmatrix} \begin{pmatrix} h_0 \\ \xi_0 \end{pmatrix} = \frac{2\beta^2}{R^2}\mu_2 \begin{pmatrix} h_0 \\ \xi_0 \end{pmatrix}, \tag{3.22}$$

where $M = Ma/RCa$. Obviously, (3.22) is an eigenvalue problem, with $2\beta^2\mu_2/R^2$ being the eigenvalue and $[h_0, \xi_0]^T$ the eigenvector. For clarity, let $\theta = 2\beta^2\mu_2/R^2$, then θ can be obtained by solving the quadratic equation

$$\theta^2 + b\theta + c = 0, \tag{3.23a}$$

where the coefficients b and c are constants and can be obtained as

$$b = \tfrac{1}{3}F^{-2} + M - I_1, \qquad c = \tfrac{1}{12}M(F^{-2} - 12I_1 + 6I_2). \tag{3.23b}$$

When surfactant is absent, i.e. $M = 0$, only (3.17) is needed, which gives

$$\theta = I_1 - \tfrac{1}{3}F^{-2}. \tag{3.24}$$

Note that $3I_1$ is just a reformulation of the parameter L defined in Yih (1968), which has also been numerically calculated by Or (1997). Equation (3.24) is identical with the results obtained by Yih (1968) and the instability criterion for a clean surface, i.e. $3I_1 > F^{-2}$, is reproduced. For a general case, there exist two roots, corresponding to two Floquet modes, which are associated with the deformation of the interface and the presence of surfactant. Since b and c are real, (3.23*a*) has two real or conjugate complex roots, which can be expressed using the well-known formula. It is easy to find that the neutral condition corresponds to the two possibilities $c = 0, b \geqslant 0$ and $b = 0, c \geqslant 0$, which combine to yield, according to (3.23*b*),

$$F^{-2} = 3\max\left(4I_1 - 2I_2, I_1 - M\right). \tag{3.25}$$

Obviously, no solution exists if the right-hand side of (3.25) is negative. The flow parameters to determine the onset of the long-wavelength instability have been reduced to F, M and β.

4. Results and discussion

First we consider a special case in the absence of gravity, i.e. $F^{-2} = 0$, which may be useful in microgravity environments. This leads to the critical condition of the instability only depending on M and β. According to (3.25), the neutral conditions become $\max(4I_1 - 2I_2, I_1 - M) = 0$. In figure 2(*a*), the neutral stability curves are plotted in the (β, M^{-1})-plane. For a clean surface at $M = 0$, the flow is unstable only for certain intervals of β satisfying $I_1 > 0$. When surfactant is introduced, these unstable intervals of β, corresponding to the unstable frequencies of oscillation, are narrowed and their width decreases as M increases, indicating a stabilizing effect of surfactant. Further increasing M will result in the neutral curves becoming vertical straight lines determined by $2I_1 - I_2 = 0$, i.e. the criterion for the onset of instability being independent of M and hence the surfactant elasticity. Note that, based on previous work (e.g. Or 1997), the regions marked S for stable in figure 2(*a*) may not be entirely stable if finite-wave disturbances are considered.

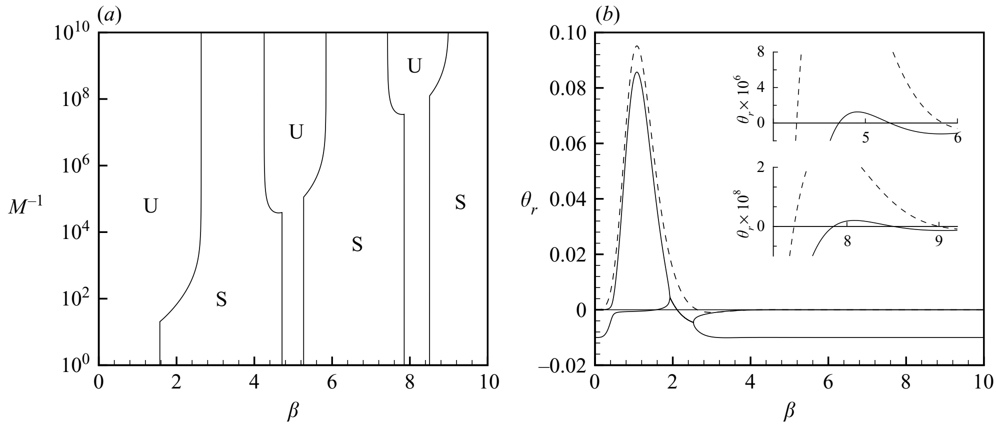

FIGURE 2. (a) Stability limits in the (β, M^{-1})-plane in the absence of gravity. Stable and unstable regions are denoted by S and U, respectively. (b) Variation of the real part of θ, θ_r, as a function of β for $M^{-1} = 100$ (solid line) and $M = 0$ (dashed line). Insets show the first mode.

To deal with the behaviour of the growth rates of the two Floquet modes, figure 2(b) exhibits the real part of θ, θ_r, as a function of β for a typical value $M^{-1} = 100$. The clean-surface results, described simply by $\theta = I_1$, are also presented for comparison. Clearly, the film flow is stabilized by surfactant since the growth rate of a contaminated surface is lower than that of a clean surface for the parameters considered here. It is also shown that both modes could be unstable. In most ranges of β, the Floquet exponents are real, corresponding to standing wave modes, while for $1.92 < \beta < 2.53$ approximately they are complex, and the corresponding curves merge together. These complex eigenvalues are associated with travelling wave disturbances, one of which propagates to the right along the interface with a phase velocity of $O(k)$ and the other one to the left. This is different from the clean-surface results, in which, according to (3.24), only standing wave solutions exist. The maximum of the eigenvalues occurs at $\beta = 1.08$ approximately, indicating that the flow is most unstable when the Stokes layer thickness is close to the depth of the film, in accordance with Yih (1968) and Or (1997). For high frequencies, typically $\beta > 4$, hence $|I_1|,\ |I_2| \ll 1$, and the eigenvalues can be approximated as $\theta_1 \approx I_1 - \frac{1}{2}I_2$ and $\theta_2 \approx -M + \frac{1}{2}I_2$. The first mode, as shown in the insets in figure 2(b), results in the unstable regions bounded by the vertical curves in figure 2(a), while the second mode is always stable. Note that the growth rate of the unstable mode is exponentially small and the flow is only mildly unstable.

When the effect of gravity is involved, according to (3.25), it is convenient to plot the neutral stability curves in the (β, F)-plane parameterized by the value of M. As shown in Or (1997), the neutral curve for a clean surface is composed of several isolated open-ended loops. When surfactant is introduced, each loop is modified to form a family of neutral curves. Figure 3 shows the neutral curves for three families. The region underneath each curve corresponds to stable modes, i.e. long-wavelength instability disappearing in these regions. We consider the neutral curves in the first family in figure 3(a). The neutral curves for $M > 7.64 \times 10^{-2}$ have the same shape, which is described by $F^{-2} = 6(2I_1 - I_2)$ and denoted by a thick line, and the corresponding neutral modes are associated with standing wave perturbations. For a slight decrease of M (e.g. $M = 7 \times 10^{-2}$), a dip occurs at the bottom of the thick curve around $\beta = 1.1$; the corresponding neutral curve joins the thick curve with discontinuous slope. Then, as M decreases gradually, the neutral curves branch off the thick curve

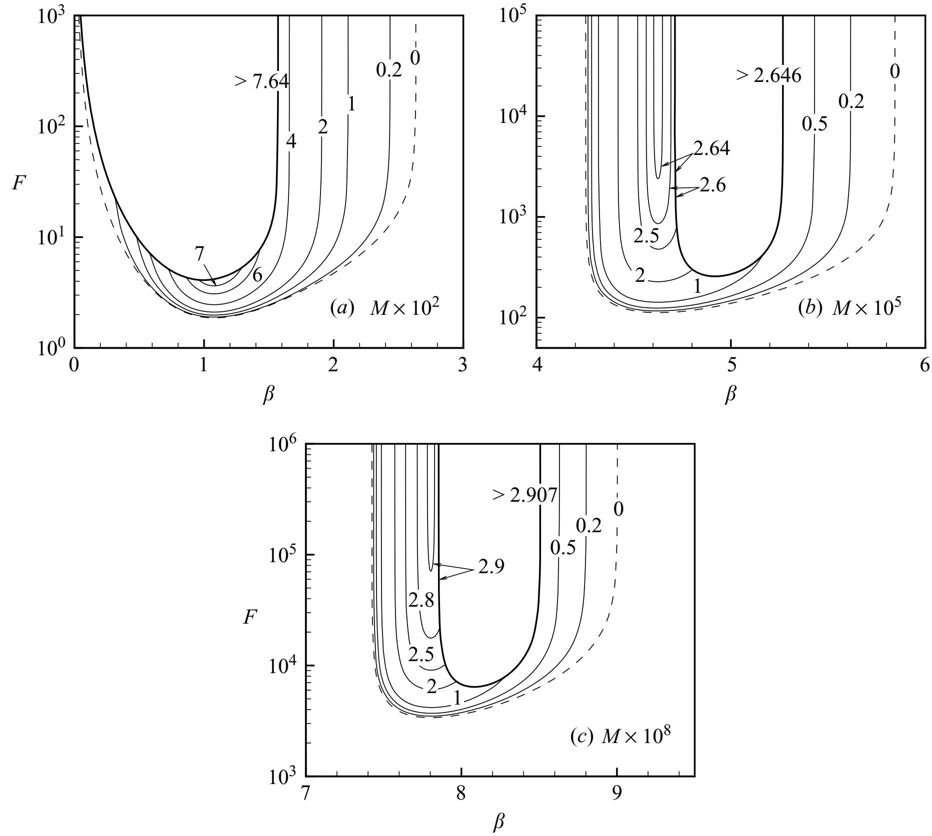

FIGURE 3. The (*a*) first, (*b*) second and (*c*) third family of the neutral stability curves for the long-wavelength instability in the (β, F)-plane for various values of M as labelled on the curves. The region above (below) each curve is unstable (stable) to long-wavelength disturbances.

and extend to a wide range of β. The right-hand portions of these curves correspond to travelling wave perturbations. Eventually, the neutral curve tends to the clean-surface one as $M \to 0$ (dashed curve). Further, as shown in figure 3(*b*), there exist different features of the neutral curves in the second family. When $M < 0.8857 \times 10^{-5}$, the neutral curves do not intersect the thick loop corresponding to $M > 2.646 \times 10^{-5}$. When $M > 0.8857 \times 10^{-5}$, the neutral curves branch off the thick loop at a point which moves to the left along the loop as M increases until $M = 2.570 \times 10^{-5}$, at which the curve is divided into two loops. The left loop shrinks as M increases further and disappears for $M > 2.646 \times 10^{-5}$, while the right one (i.e. the thick line loop) remains unchanged. Similar behaviours are demonstrated for the neutral curves in the third family in figure 3(*c*), except for higher values of F and smaller values of M. Since the neutral curves lie in the interior of the regions bounded by the clean-surface curve, the critical Froude number is raised, and long-wavelength instability for particular frequencies is fully eliminated due to the presence of surfactant.

Previous studies for steady film flows mainly interpreted the mechanism of instability based on Marangoni forces (Frenkel & Halpern 2002; Blyth & Pozrikidis 2004*b*) or disturbance vorticity (Hinch 1984; Kelly *et al.* 1989; Charru & Hinch 2000; Wei 2005*b*). As indicated by Wei (2005*b*), only considering the effects of Marangoni forces may result in an inconsistent interpretation, while the viewpoint of vorticity is only appropriate when inertial effects are small, which is not the case in the present

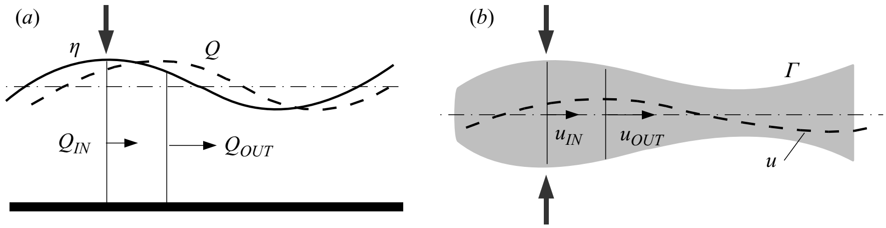

FIGURE 4. Sketch for the instability mechanism: (*a*) positive Q–η phase difference suppresses the interfacial deflection. The large arrow represents the downward motion of the crest. (*b*) Positive u–Γ phase difference smooths the surfactant distribution. The surfactant concentration Γ is denoted by the width of the grey area and the large arrows represent the decrease of the surfactant concentration.

study. The instability of oscillatory film flow can be straightforwardly explained by considering the effects of the disturbance flow field. First, the disturbance velocity causes a spatially distributed mass flow rate, related to the disturbance streamfunction,

$$Q(x,t) = \int_{-1}^{\eta} u \, \mathrm{d}y = \int_{-1}^{0} U(y,t)\, \mathrm{d}y + \psi'(x,0,t) + U(0,t)\eta + O(\varepsilon^2), \tag{4.1}$$

which may induce the growth of the interface. Second, the surface velocity

$$u(x,\eta,t) = U(0,t) + u'(x,0,t) + O(\varepsilon^2) \tag{4.2}$$

rearranges the distribution and causes local convergence of surfactant. The stability of the flow system depends on the phase difference of the mass flow rate relative to the interface as well as that between the interfacial velocity and the surfactant concentration. As shown in figure 4(*a*), for a control volume enclosed by the vertical lines and the interface, the net flow rate is negative since the flow entering the volume, Q_{IN}, is less than that leaving it, Q_{OUT}, resulting in the decay of the interfacial deformation. Thus, we can interpret that positive (negative) Q–η phase difference, defined in the interval $[-\pi, \pi]$, is associated with a stable (unstable) flow. A similar result can be obtained from the relation between the u–Γ phase difference and the stability, as shown in figure 4(*b*). Since we are concerned with the mean growth of the flow, only the steady part of the disturbance flow, which determines the phase difference, is relevant to the stability. The flow rate perturbation as well as the interfacial velocity perturbation are induced by gravity, the Marangoni traction, the flow due to inertia and the advective effects of the basic flow. It is clearly indicated from (3.19) and (3.20) that the flow rate due to gravity leads η by a phase $\pi/2$, indicating that gravity always plays a stabilizing role. Inertial effects together with the advective effects of the basic flow are destabilizing (stabilizing) when I_1 is positive (negative), corresponding to the phase difference $-\pi/2$ ($\pi/2$). The effects of Marangoni flow are determined by the argument of $\mathrm{i}\xi_0/h_0$, which is always positive at the critical conditions based on our extensive calculations, indicating a stabilizing effect of surfactant.

Finally, we emphasize that the instability in the present problem is due to the interfacial deformation and the presence of surfactant, instead of the classical shear modes associated with the basic velocity profile. For sufficiently high frequencies, however, the shear modes may dominate the stability, and the stability characteristics should tend to those of plane Stokes layers. The relevant critical Reynolds number based on the thickness of flat Stokes layers has been studied by Blennerhassett & Bassom (2002) and Gao & Lu (2006).

5. Summary

The stability of film flow driven by an oscillatory plate and covered by an insoluble surfactant has been studied analytically in the limit of long-wavelength perturbations. An eigenvalue problem is derived through the asymptotic expansion of the differential system governing the stability of the flow. Two Floquet modes are reported and both could be unstable. In addition to standing wave modes, travelling wave modes are also detected in certain regions of the parameters. The inertial instability is suppressed by surfactant in the absence of gravity, while the unstable regions shrink due to the stabilizing effects of surfactant in the presence of gravity. We also find that the criterion of instability does not change when the surfactant elasticity (i.e. M) exceeds a threshold. The relevant instability mechanism is interpreted based on the disturbance flow rate.

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