

Stability of a viscoelastic falling film with surfactant subjected to an interfacial shear

Hsien-Hung Wei

Department of Chemical Engineering, National Cheng Kung University, Tainan 701, Taiwan

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The long-wavelength stability is analyzed for a surfactant-laden, viscoelastic liquid flowing down an inclined plane when the liquid undergoes additional interfacial shear. The upper convected Maxwell model is employed for describing the elastic nature of the fluid. The system stability is characterized by the interface and the surfactant modes. The interface mode involves both elastic and Marangoni effects that modulate the stability with applied shears and gravity-driven flow. The surfactant mode is only determined by the shear-induced Marangoni effects. A phase diagram is established to identify the dominant mode and the overall features of instability. It reveals that the system is susceptible to instability, except in a stable window when the applied shear opposes the gravity-driven flow.

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I. INTRODUCTION

Liquid flowing down an inclined plane is a subject of long-standing interest in fluid mechanics and engineering processes. Benjamin [1] and Yih [2] first examined the stability of a Newtonian film for long-wavelength perturbations. A hydrodynamic instability arises from a combination of surface waves and inertial effects. Since destabilizing effects can be compromised by hydrostatic pressures, instability occurs when the Reynolds number is greater than a certain critical value. The development of such surface waves has received much attention, and extensive research studies on this subject can be found in an excellent review given by Chang [3].

Most of the stability analyses of falling film flows are based on Newtonian fluids which have been studied thoroughly. The corresponding analysis based on non-Newtonian fluids, especially on viscoelastic liquids, has been addressed by fewer studies due to the complex nature of the fluid rheology. Gupta [4] used a second-order fluid model to first examine the effects of viscoelasticity on the stability of falling film flow. He found that viscoelasticity has a destabilizing effect on long-wavelength disturbances since it decreases the critical Reynolds number. Shaqfeh *et al.* [5] examined the linear stability of an Oldroyd-B falling film in more detail. They solved the modified Orr-Sommerfeld equation and showed that although the system can be destabilized by viscoelasticity, the resulting growth rate is too small to be discernible in practice.

The stability analysis has also been extended to systems of two viscoelastic fluids flowing down an inclined plane [6,7]. In addition to the surface-wave mode (called the Yih mode) usually found in a single-layer problem, there is an interfacial mode that can modify the features of stability. The origin of instability of this mode is identified by a jump in the first normal stress difference across the interface [8]. This jump arises from an interfacial deflection; it works in the form of basic interfacial shear and can be reflected in the elasticity contrast between the two fluids. The general mechanism of elastic instability was clearly illustrated by Hinch *et al.* [9].

Studies so far on the stability of viscoelastic liquids are restricted to clean-interface systems. Since most of the appli-

cations involve surface-active agents or impurities on the interface, their influence on the stability could be critical to processes. Although there are a few studies addressing the effects of surfactants on the stability of Newtonian liquid flow, to our best knowledge, the roles of surfactants in affecting the stability features of viscoelastic liquid flow have not yet been explored. It is, first of all, essential to understand the effects of surfactants on the stability of Newtonian fluid flow systems. We describe some features thereof below.

The dominant effects introduced by surfactant are Marangoni forces that act on the interface and drive the fluid to flow toward lower-tension regions. For stationary systems, surfactant has a stabilizing effect [10]. When basic flows are present, however, the surfactant could be either stabilizing or destabilizing, depending on the nature of the basic flow. Whitaker and Jones [11] examined the long-wave stability of a falling film flow and found that a surfactant increases the critical Reynolds number, indicating stabilizing effects. Their finding is essentially a correction to the Yih mode. In related work, Ji and Setterwall [12] demonstrated the existence of an unstable mode due to Marangoni effects in the presence of a soluble surfactant. Pozrikidis [13] studied the stability of a surfactant-laden falling film flow in the limit of vanishing Reynolds number. He identified, in addition to the surface-wave mode that is stabilized by the surfactant, a surfactant mode that can make the growth rate decay more slowly than that of the Yih mode. In this viewpoint, surfactant can be said to have a destabilizing influence. Blyth and Pozrikidis [14] recently solved the Orr-Sommerfeld equation numerically and confirmed the above findings.

Frenkel and Halpern [15,16] analyzed the linear stability of a two-layer Couette flow with surfactant in the limit of zero Reynolds number. They demonstrated that surfactant could introduce destabilization to a system that is inherently stable in the absence of surfactant. Their results were also later confirmed by Blyth and Pozrikidis [14]. Such destabilizing effects due to surfactant are also found in cylindrical geometries in which the prevailing capillary instability could be enhanced by the surfactant [17,18].

The key to the flow-induced Marangoni destabilization lies in the interfacial shear of the basic flow. Perturbations to the basic interfacial shear can redistribute surfactant along

the interface. This creates a phase difference between the surfactant concentration and interface deflection; the resulting Marangoni forces can promote interface growth. Wei [19] considered the long-wave stability of a surfactant-laden falling film subjected to an additional interfacial shear. He identified that there is a surfactant mode that is solely excited by an imposed shear and irrelevant to gravity-driven basic flow. The surfactant mode is destabilizing (stabilizing) when the imposed shear acts in the direction in favor of (opposite to) gravity-driven flow. In the absence of imposed shear [11], this mode has a zero growth rate for long-wavelength disturbances; it is the mode that has a destabilizing influence in the finite-wavelength regime [13]. As for the Yih mode, the surfactant correction in response to the action of shear has effects opposite to those of the surfactant mode. The resulting system stability is determined by the competition between these modes, depending on the strength and direction of the applied shear.

The motivation of this study arises from the efforts to construct an appropriate model for understanding liquid lining flows in airways. The lining liquid is typically a bilayer structure that comprises a Newtonian ciliary layer overlaid by a mucous layer that has a viscoelastic nature [20]. Airflow travels back and forth during breathing and could exert shear forces on the air-liquid interface. Since the interface is often populated with surfactant, the dynamics are also influenced by Marangoni effects. As mentioned earlier, the effects of basic flows are twofold. On the one hand, they can induce elastic instability; on the other hand, they also can modulate the Marangoni influence on the stability. Since an imposed shear can act to either assist or oppose the flow caused by gravity, it is not clear how these effects interplay in response to various basic flow conditions. In this paper, we shall address this issue by examining the combined effects of interfacial shear and surfactant on the long-wave stability of a viscoelastic liquid down an inclined plane. The rheology of the liquid is assumed to follow the upper convected Maxwell (UCM) model. The UCM model can be derived from a molecular theory that idealizes the polymer molecules as non-interacting dumbbells [21]. It serves as a relatively simple constitutive model that attains the essential physics and enables us to assess its impact on the stability.

II. MATHEMATICAL FORMULATION

Consider an incompressible, UCM liquid flowing down an inclined plane with an inclined angle θ (see Fig. 1). The liquid has density ρ , viscosity μ , and relaxation time λ . An additional constant shear stress τ_s^* induced by an airflow is applied along the air-liquid interface and its direction can either assist or oppose the gravity-driven flow. The base state configuration consists of a liquid layer with a uniform thickness of h . The air-liquid interface is covered by an insoluble surfactant monolayer of a uniform concentration Γ_0^* . The surface tension is σ_0^* corresponding to Γ_0^* . We choose h as the characteristic length and scale the velocities with respect to the basic interfacial velocity $U_s^* = \rho g h^2 (\sin \theta) / (2\mu)$. The time is scaled by h/U_s^* . Both stress and pressure are scaled by $\mu U_s^*/h$. The surfactant concentration is normalized by Γ_0^* .

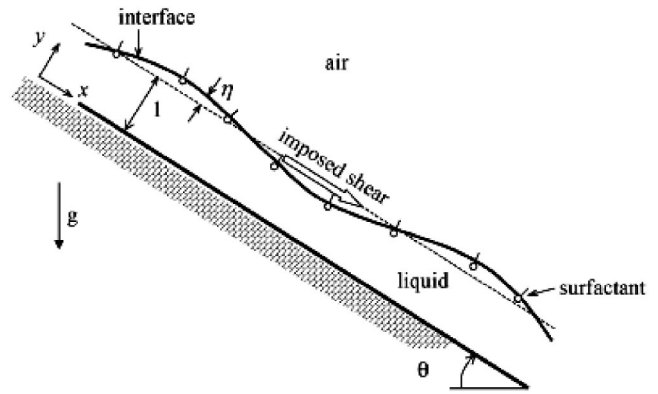


FIG. 1. Geometry of a viscoelastic liquid with surfactant flow down an inclined plane when the liquid undergoes an interfacial shear.

We define x to be the coordinate along the plane and y to be the coordinate perpendicular to the liquid layer with $y=0$ defining the plane, as shown in Fig. 1. The velocity vector of the flow is $\mathbf{V}=(u,v)$ where u and v denote the velocity components in the x and y directions, respectively. p is the pressure. In the dimensionless form, the continuity and momentum equations are

$$\nabla \cdot \mathbf{V} = 0, \quad (1)$$

$$\text{Re} \left(\frac{\partial \mathbf{V}}{\partial t} + \mathbf{V} \cdot \nabla \mathbf{V} \right) = -\nabla p + 2\mathbf{e}_g + \nabla \cdot \mathbf{S}, \quad (2)$$

where \mathbf{S} is the viscous stress tensor and $\mathbf{e}_g = \mathbf{e}_x - (\cot \theta)\mathbf{e}_y$ indicates the direction of the gravity force. The Reynolds number is defined by $\text{Re} = \rho U_s^* h / \mu$. The UCM constitutive equation is

$$\text{We} \mathbf{S} + \mathbf{S} = \nabla \mathbf{V} + (\nabla \mathbf{V})^T, \quad (3)$$

$$\mathbf{S} = \frac{\partial \mathbf{S}}{\partial t} + (\mathbf{V} \cdot \nabla) \mathbf{S} - \nabla \mathbf{V} \cdot \mathbf{S} - \mathbf{S} \cdot (\nabla \mathbf{V})^T, \quad (4)$$

where $\text{We} = \lambda U_s^*/h$ is the Weissenberg number, the ratio of elastic relaxation to flow time scales. The system is subject to the following boundary conditions. The velocity vanishes on the wall:

$$u = v = 0 \quad \text{on } y = 0. \quad (5)$$

At the interface $y=1+\eta$ with η being an interfacial displacement, the tangential stress and normal stress conditions are given by

$$\frac{1}{[1 + (\partial \eta / \partial x)^2]^{1/2}} \left\{ \left[1 - \left(\frac{\partial \eta}{\partial x} \right)^2 \right] S_{xy} - \left(\frac{\partial \eta}{\partial x} \right) (S_{xx} - S_{yy}) \right\} = \frac{1}{\text{Ca}} \frac{\partial \sigma}{\partial x} + \tau_s, \quad (6)$$

$$-p + \frac{1}{[1 + (\partial\eta/\partial x)^2]^{1/2}} \left[S_{yy} + \left(\frac{\partial\eta}{\partial x} \right)^2 S_{xx} - \frac{\partial\eta}{\partial x} S_{xy} \right] = \frac{\sigma}{\text{Ca}[1 + (\partial\eta/\partial x)^2]^{3/2}} \frac{\partial^2 \eta}{\partial x^2}, \quad (7)$$

where $\tau_s = \tau_s^* h / \mu U_s^*$ is the dimensionless imposed interfacial stress and $\text{Ca} = \mu U_s^* / \sigma_0^*$ is the capillary number. σ denotes the dimensionless surface tension scaled by σ_0^* , and its dependence on the surfactant concentration can be assumed to follow the linear equation of state

$$\sigma = 1 - E(\Gamma - 1), \quad (8)$$

where $E = -(\Gamma_0^* / \sigma_0^*) (\partial\sigma^* / \partial\Gamma^*)_{\Gamma_0^*}$ reflects the surfactant elasticity. The kinematic condition at the interface is

$$v = \frac{\partial\eta}{\partial t} + u \frac{\partial\eta}{\partial x}. \quad (9)$$

For an insoluble surfactant with negligible surface diffusion, the transport equation along the interface is given by Ref. [16]:

$$\frac{\partial}{\partial t} \left[\sqrt{1 + \left(\frac{\partial\eta}{\partial x} \right)^2} \Gamma \right] + \frac{\partial}{\partial x} \left[\sqrt{1 + \left(\frac{\partial\eta}{\partial x} \right)^2} \Gamma u \right] = 0. \quad (10)$$

The base states (denoted by overbars) are given by

$$\bar{u} = (2y - y^2) + \tau_s y, \quad \bar{p} = p_0 + 2(\cot\theta)(1 - y), \quad \bar{\Gamma} = 1, \\ \bar{S}_{xy} = \bar{u}', \quad \bar{S}_{xx} = 2 \text{We}(\bar{u}')^2, \quad (11)$$

where p_0 is a constant pressure of air and the primes indicate the derivatives with respect to y .

With the base states above, we now analyze the corresponding linear stability. We consider perturbations to be only two dimensional by appealing to Squire's theorem for the UCM fluid used here [22]. The perturbation quantity f (i.e., u , v , p , S , η , or Γ) is expressed in the form of the normal mode, $f = \hat{f} \exp[ik(x - ct)]$ where k is the wave number of the perturbation and c is the complex wave speed. The system is stable (unstable) if the imaginary part of c , $\text{Im}(c)$, $< 0 (> 0)$. After substituting perturbation quantities into Eqs. (1)–(9) and linearizing these equations, the normal-mode equations (dropping the overheads for perturbation quantities now) can be written as

$$iku + v' = 0, \quad (12)$$

$$\text{Re}[ik(\bar{u} - c)u + \bar{u}'v] = -ikp + S'_{xy} + ikS_{xx}, \quad (13)$$

$$ik \text{Re}(\bar{u} - c)v = -p' + ikS_{xy} + S'_{yy}, \quad (14)$$

$$S_{xx} + \text{We}[ik(\bar{u} - c)S_{xx} + \bar{S}'_{xx}v - 2(ik\bar{S}_{xx}u + \bar{S}'_{xy}u' + \bar{u}'S_{xy})] = 2iku, \quad (15)$$

$$S_{xy} + \text{We}[ik(\bar{u} - c)S_{xy} + \bar{S}'_{xy}v - ik\bar{S}_{xy}v - \bar{u}'S_{yy}] = u' + ikv, \quad (16)$$

$$S_{yy} + \text{We}[ik(\bar{u} - c)S_{yy} - 2ik\bar{S}_{xy}v] = 2v', \quad (17)$$

$$u = v = 0 \quad \text{at } y = 0 \quad (18)$$

$$S_{xy} + \bar{S}'_{xy}\eta - ik\bar{S}_{xx}\eta = -ikM\Gamma \quad \text{at } y = 1 \quad (19)$$

$$-p - \bar{p}'(1)\eta - S_{yy} - 2ik\eta\bar{S}_{xy} = -\frac{k^2}{\text{Ca}}\eta \quad \text{at } y = 1, \quad (20)$$

$$v = ik[\bar{u}(1) - c]\eta \quad \text{at } y = 1, \quad (21)$$

$$[\bar{u}(1) - c]\Gamma + \bar{u}'(1)\eta + u = 0 \quad \text{at } y = 1, \quad (22)$$

where $M = E/\text{Ca}$ is the Marangoni number. For each k , the system of equations (12)–(22) constitutes an eigenvalue problem that can be used to determine c .

III. LONG-WAVELENGTH STABILITY ANALYSIS

In the limit of long wavelengths ($k \rightarrow 0$), we follow the regular perturbation technique first adopted by Yih [2] to expand perturbation quantities as

$$(u, v, p, S, \eta, \Gamma, c) = (u_0, kv_0, p_0, S^0, \eta_0, \Gamma_0, c_0) \\ + k(u_1, kv_1, p_1, S^1, \eta_1, \Gamma_1, c_1) + \dots \quad (23)$$

Here we rescale v as $O(k)$ in view of the continuity equation (12). We substitute Eq. (23) into Eqs. (12)–(22) and collect the terms in each order of k . We further assume that Re , We , and M are all $O(1)$. At $O(1)$, we obtain

$$iu_0 + v'_0 = 0, \quad (24)$$

$$(S^0_{xy})' = 0, \quad (25a)$$

$$p'_0 = 0, \quad (25b)$$

$$S^0_{xy} + \text{We}[-\bar{u}'S^0_{yy}] = u'_0, \quad (26a)$$

$$S^0_{xx} + \text{We}[-2(\bar{S}_{xy}u' + \bar{u}'S^0_{xy})]u'_0 = 0, \quad (26b)$$

$$S^0_{yy} = 0, \quad (26c)$$

$$u_0(0) = v_0(0) = 0, \quad (27a)$$

$$S^0_{xy}(1) + \bar{S}'_{xy}(1)\eta_0 = 0, \quad (27b)$$

$$p_0(1) = -\bar{p}'(1)\eta_0, \quad (27c)$$

$$v_0(1) = [\bar{u}(1) - c_0]\eta_0, \quad (28a)$$

$$[\bar{u}(1) - c_0]\Gamma_0 + \bar{u}'(1)\eta_0 + u_0(1) = 0. \quad (28b)$$

The leading order solution is

$$u_0 = -\bar{U}'' \eta_0 y, \quad v_0 = \frac{i}{2} \bar{U}'' \eta_0 y^2,$$

$$S_{xy}^0 = -\bar{U}'' \eta_0, \quad S_{xx}^0 = 4 \text{We} \bar{u}' u_0', \quad p^0 = 2 \eta_0 \cot \theta, \quad (29)$$

$$\left(c_0 - \bar{U} + \frac{1}{2} \bar{U}'' \right) \eta_0 = 0, \quad (30a)$$

$$(c_0 - \bar{U}) \Gamma_0 = (\bar{U}' - \bar{U}'') \eta_0. \quad (30b)$$

Here we define $\bar{U} \equiv \bar{u}(1) = 1 + \tau_s$, $\bar{U}' \equiv \bar{u}'(1) = \tau_s$, and $\bar{U}'' \equiv \bar{u}''(1) = -2$ for simplicity. As revealed by Wei [19], Eqs. (30a) and (30b) suggest that there are two modes as follows. For $\eta_0 \neq 0$, these equations result in, respectively,

$$c_0 = 2 + \bar{U}', \quad (31a)$$

$$\Gamma_0 = (2 + \bar{U}') \eta_0. \quad (31b)$$

Since c_0 is determined by the leading order kinematic condition (30a), we call this mode an *interface* mode as usually found in free-falling systems [2,11]. The surfactant concentration thus acts in response to the dynamics of the interface via Eq. (30b) and is in phase (out of phase) with the interface when $2 + \bar{U}' > 0$ (< 0). In addition to the interface mode, there is a *surfactant* mode which can be triggered by the surfactant concentration perturbations ($\Gamma_0 \neq 0$) without necessarily having an interfacial deflection. For this mode, Eqs. (30a) and (30b) yield

$$c_0 = \bar{U}, \quad (32a)$$

$$\eta_0 = 0. \quad (32b)$$

Note that c_0 here is determined from the leading order surfactant transport equation (30b). As shown above, since c_0 is real, the $O(1)$ problem does not contribute to the stability of the system. The above results were also shown previously for a Newtonian fluid [19]. Neither viscoelastic nor surfactant effects influence the stability at this order because both We and M are $O(1)$ here. As we shall show next, their impacts on the stability will appear in the $O(k)$ problem.

For the $O(k)$ problem, we obtain the following equations:

$$i u_1 + v_1' = 0, \quad (33)$$

$$(S_{xy}^1)' + i S_{xx}^0 - i p_0 = \text{Re}[i(\bar{u} - c_0) u_0 + \bar{u}' v_0], \quad (34a)$$

$$i S_{xy}^0 + (S_{yy}^1)' - p_1' = 0, \quad (34b)$$

$$\begin{aligned} S_{xx}^1 + \text{We}[i(\bar{u} - c_0) S_{xx}^0 + v_0 \bar{S}'_{xx} - 2(i \bar{S}'_{xx} u_0 + \bar{S}_{xx} u_1' + \bar{u}' S_{xy}^1)] \\ = 2i u_0' \end{aligned} \quad (35a)$$

$$S_{xy}^1 + \text{We}[i(\bar{u} - c_0) S_{xy}^0 + v_0 \bar{S}'_{xy} - \bar{u}' S_{yy}^1] = u_1', \quad (35b)$$

$$S_{yy}^1 + \text{We}[i(\bar{u} - c_0) S_{yy}^0] = 2v_1', \quad (35c)$$

$$u_1(0) = v_1(0) = 0, \quad (36a)$$

$$S_{xy}^1(1) = 2i \text{We}(\bar{U}')^2 \eta_0 - \bar{U}'' \eta_1 - iM \Gamma_0, \quad (36b)$$

$$-p_1(1) - \bar{p}'(1) \eta_1 + S_{yy}^1(1) - 2i \eta_0 \bar{S}'_{xy}(1) = 0, \quad (36c)$$

$$v_1(1) = -i c_1 \eta_0 + i(\bar{U} - c_0) \eta_1, \quad (37a)$$

$$(\bar{U} - c_0) \Gamma_1 - c_1 \Gamma_0 + \bar{U}' \eta_1 + u_1 = 0. \quad (37b)$$

Making use of Eqs. (35b) and (35c), Eq. (34a) leads to

$$u_1'' = i p_0 - i \text{We}(\bar{u}' u_0' - \bar{u}'' u_0) + \text{Re}[i(\bar{u} - c_0) u_0 + \bar{u}' v_0]. \quad (38)$$

The velocity field that satisfies Eqs. (36a) and (36b) is

$$\begin{aligned} u_1 = i p_0 \left(\frac{y^2}{2} - y \right) - 4i \text{We} \eta_0 (\bar{U}' + 2) \left(\frac{y^2}{2} - y \right) \\ + [2 \eta_1 + 2i \text{We} \bar{U}' (\bar{U}' + 2) \eta_0 - iM \Gamma_0] y \\ + i \eta_0 \text{Re} \left[\frac{1}{3} (\bar{U}' + 2) \left(\frac{y^4}{4} - y \right) - c_0 \left(\frac{y^3}{3} - y \right) \right], \end{aligned} \quad (39a)$$

$$\begin{aligned} v_1 = p_0 \left(\frac{y^3}{6} - \frac{y^2}{2} \right) - \text{We}(\bar{U}' + 2) \eta_0 \left(\frac{y^3}{3} - y^2 \right) \\ - i \left(\eta_1 + i \text{We} \bar{U}' (\bar{U}' + 2) \eta_0 - i \frac{M}{2} \Gamma_0 \right) y^2 \\ + \eta_0 \text{Re} \left[\frac{1}{3} (\bar{U}' + 2) \left(\frac{y^5}{20} - \frac{y^2}{2} \right) - c_0 \left(\frac{y^4}{12} - \frac{y^2}{2} \right) \right]. \end{aligned} \quad (39b)$$

p_1 can be obtained using Eqs. (34b), (35c), and (36c). Since it is not used for determining the $O(k)$ wave speed c_1 , it does not affect the stability at this order. We thus do not pursue it further.

In view of the two modes derived in the $O(1)$ problem, we show the corresponding complex wave speed c_1 for each mode of the $O(k)$ problem below.

A. The interface mode

As in the $O(1)$ problem, the c_1 for the interface mode should be determined from the $O(k)$ kinematic condition (37a). Substituting Eqs. (39a) and (39b) into Eq. (37a), with the aid of Eq. (31), yields

$$c_1 = -\frac{2}{3} i \cot \theta + i \left[\frac{4}{15} \text{Re} + \text{We} \left(\bar{U}' + \frac{2}{3} \right) - \frac{1}{2} M \right] (\bar{U}' + 2). \quad (40)$$

In the absence of elasticity, the result agrees with that of Wei [19]; surfactant has a stabilizing (destabilizing) influence when $\bar{U}' + 2 > 0$ (< 0). The viscoelastic effects are reflected

by the term $i \text{We}(\bar{U}' + 2)(\bar{U}' + 2/3)$. In the absence of imposed shears and surfactant, the result is also consistent with that of Gupta [4]; viscoelastic effects are destabilizing.

Similar to the mechanism of purely elastic instability illustrated by Hunag and Khomami [7] for free surface flow, imposing an additional interfacial shear can either encourage or discourage the viscoelastic destabilization. A basic flow working through elasticity has two contributions to instability. First, it drives a perturbation flow through the elastic relaxation via $-i \text{We}(\bar{u}' u'_0 - \bar{u}'' u_0) = -2i \text{We}(\bar{U}' + 2) \eta_0$ of Eq. (38). For $\bar{U}' + 2 > 0$, this effect drives a perturbation flow forward (backward) for $\partial \eta / \partial x > 0$ (< 0). It in turn drives flows from troughs to peaks of the interface, promoting the growth of the interface. $\bar{U}' + 2 < 0$ acts in the opposite way; it is thus stabilizing. This elastic contribution induces a parabolic flow of $u_1 = -2i \text{We}(\bar{U}' + 2) \eta_0 (y^2/2 - y)$; the corresponding flow rate is thus $Q_1 = (2i/3) \text{We}(\bar{U}' + 2) \eta_0$ from which the dependence of stability on $\bar{U}' + 2$ explained above becomes evident.

The second elastic contribution arises from the elastic stress jump at the interface: $S_{xy}^1 = -(\bar{S}_{xy})' \eta_1 + i \bar{S}_{xx} \eta_0$ from Eq. (36b). There are three contributions from this condition as follows. For $S_{xy}^1(1)$, one can show that $S_{xy}^1(1) = u_1'(1) - 4i \text{We} \bar{U}' \eta_0$ from Eq. (35b). Hence, the term $4i \text{We} \bar{U}' \eta_0$ is destabilizing (stabilizing) for $\bar{U}' > 0$ (< 0). As for the term $-(\bar{S}_{xy})' \eta_1 = 2\eta_1$, it is only dispersive to the stability. The term $i \bar{S}_{xx} \eta_0 = 2i \text{We}(\bar{U}')^2 \eta_0$ can cause instability as demonstrated by Chen [8]. The instability due to this term can be analogous to the Rayleigh-Taylor instability, as illuminated by Graham [23]; that is, the elastic stress acts as an extra force pointing upward upon the interface, which promotes the interface growth. This term is always destabilizing regardless of the direction of the applied shear because of the value of $(\bar{U}')^2$. Hence, combining the effects of $S_{xy}^1(1)$ and $i \bar{S}_{xx} \eta_0$ induces a linear flow of $u_1 = 2i \text{We} \bar{U}' (\bar{U}' + 2) \eta_0 y$, and the flow rate is $Q_1 = i \text{We} \bar{U}' (\bar{U}' + 2) \eta_0$. Notice that for free surface flow [4,7], $\bar{U}' = 0$, there is no impact on the stability from this elastic contribution.

Consequently, the elastic effects combining both contributions shown above characterize c_1 as $i \text{We}(\bar{U}' + 2)(\bar{U}' + 2/3)$ which manifests elastic stabilization if $-2 < \bar{U}' < -2/3$ and destabilization otherwise. In contrast to the previous studies [4,7], applications of shear on the interface could have stabilizing effects due to elasticity.

The presence of surfactant also can modulate the stability via $-iM(\bar{U}' + 2)$. It is stabilizing if $\bar{U}' > -2$. Marangoni effects are generated in response to flow caused by interface deflections. Stability and instability can be explained by the phase difference between η_0 and Γ_0 [19]. Equation (31b) reveals that for $\bar{U}' > -2$, η_0 and Γ_0 are in phase, so Marangoni stresses push fluid toward the interface's troughs, relaxing corrugations of the interface. Similarly, the effects of $\bar{U}' < -2$ are the reverse.

For a vertical flow system with small Re, combining elastic and surfactant effects lead to instability when

$$\bar{U}' > \max(M/\text{We} - 2/3, -2) \quad \text{or} \quad \bar{U}' < \min(M/\text{We} - 2/3, -2).$$

For a sufficiently strong applied shear $|\bar{U}'| \gg 1$ [but still $\ll O(k^{-1})$ for ensuring the validity of the small- k expansion], the elastic effects dominate the instability, i.e., $c_1 \sim i \text{We}(\bar{U}')^2$. In this situation, strong imposed shears lead the destabilizing effects of the elastic-stress jump $i \bar{S}_{xx} \eta_0 = 2i \text{We}(\bar{U}')^2 \eta_0$ to become more dominant compared to other elastic contributions that are linear in \bar{U}' . Such an instability is independent of the direction of applied shear, as expected.

B. The surfactant mode

We now turn our attention to the surfactant mode. As in the $O(1)$ problem, we should apply the $O(k)$ surfactant transport equation (37b) for obtaining c_1 . Applying Eqs. (32a) and (32b) and (39a) and (39b) to Eq. (37b), we find

$$c_1 = \frac{i}{2} M \bar{U}'. \quad (41)$$

Unlike the interface mode, here the stability is solely determined by Marangoni effects. It depends on whether the imposed shear acts to assist or oppose gravity: the former ($\bar{U}' > 0$) destabilizes while the latter ($\bar{U}' < 0$) stabilizes. This result is also identical to the study on Newtonian flow [19], suggesting that the stability of this mode does not depend on the detailed information of the fluid rheology.

The key feature of this mode is that the instability arises from imbalance of surfactant mass, rather than that of fluid mass as in the interface mode. In addition, since this mode has $\eta_0 = 0$, interfacial deflections do not have sufficiently large amplitudes [of $O(k)$ at most] to furnish elastic instability. This is evidenced by the fact that $\eta_0 = 0$ makes all of the We terms vanish in Eqs. (39a) and (39b). Since both hydrostatic and inertial contributions also vanish as well when $\eta_0 = 0$, the resulting velocity field is simply a linear flow:

$$u_1 = (2\eta_1 - iM\Gamma_0)y, \quad (42a)$$

$$v_1 = -i \left(\eta_1 - \frac{i}{2} M \Gamma_0 \right) y^2. \quad (42b)$$

This is just a result of the stress-driven flow, viz.,

$$u_1'' = 0 \quad \text{with} \quad u_1'(1) = 2\eta_1 - iM\Gamma_0, \quad (43)$$

which is deduced from Eqs. (34a), (35a), and (36b). Again, the term $2\eta_1$ comes from a perturbation to the basic interfacial stress $-\bar{U}'' \eta_1$ and it is only dispersive to the stability. The instability is caused by the Marangoni stress $iM\Gamma_0$. The mechanism of the Marangoni-induced instability was explained previously [19]. It is provided here for completeness.

As revealed by Eqs. (37a), (32a), and (32b), the flat interface $\eta_0 = 0$ admits a zero normal velocity $v_1(1) = 0$. This demands $\eta_1 = i(M/2)\Gamma_0$; that is, the perturbation to the basic interfacial stress balances the Marangoni stress on the interface. As a consequence of the momentum conservation from Eq. (43), there is no perturbation flow everywhere, i.e., $u_1 = v_1 = 0$. Now inspecting Eq. (37b) for the surfactant transport, we find

$$-ikc_1\Gamma_0 + k\bar{U}'\eta_1 = 0. \quad (44)$$

Written back in the form of a differential equation, Eq. (44) is equivalent to

$$\frac{\partial\Gamma}{\partial t} + \bar{U}'\frac{\partial\eta}{\partial x} = 0, \quad (45)$$

wherein the time derivative is defined in the frame moving with the basic interfacial velocity \bar{U} in view of Eq. (32a). Hence, Eq. (45) just tells how surfactant transports along the interface when undergoing a shear flow. For $\bar{U}' > 0$, interface perturbations [with amplitudes of $O(k)$ here] lead the surfactant concentration to decrease (increase) for the interface portions of $\partial\eta/\partial x > 0$ (< 0), causing Γ to be ahead of η with a phase of $\pi/2$. To maintain the interface nearly flat without growth, the induced Marangoni stress has to be in balance with the perturbation shear stress due to the gravity-driven basic flow. Such a nearly stationary interface working with the shear flow keeps magnifying the amplitude of the surfactant concentration perturbation. This accelerates local accumulation or consumption of the surfactant mass on the interface; hence the instability.

In comparison with the interface mode Eq. (40), the influence of imposed shear on the Marangoni parts for both modes has the same form $i\bar{U}'M/2$ but with opposite signs. If Marangoni effects dominate the instability, e.g., very large M compared to both Re and We , imposing shears with $|\bar{U}'| \gg 1$ will lead to instability regardless of their direction of exertion. In that case, since the dispersion equation for c is in the form of $(c - \bar{U})^2 = O(k)$, the correction to c is $O(k^{1/2})$. The resulting wave-speed correction is $k^{1/2}c_1 = (i/2)M|\bar{U}'|k^{1/2}$ which is of $O(k^{1/2})$ larger than the case of moderate $|\bar{U}'|$. Such $O(k^{1/2})$ wave speed is consistent with previous findings in two-layer systems [15].

C. Overall stability behaviors as combining both the interface and surfactant modes

As demonstrated above, we discuss the respective stability features for the interface and the surfactant modes. In general, since development of any perturbation can be regarded as a linear combination of these two modes, the system stability hinges on their competition for given fluid properties and flow conditions. The overall behavior of the stability is discussed below.

Application of interfacial shear can affect both elastic and surfactant influences on the system stability. As shown in the interface mode Eq. (40), elastic effects can be either stabilizing or destabilizing, depending on the strength and direction of the applied shear. In addition to hydrostatic and inertial effects, the elastic influence on the stability of the interface mode is further mediated by Marangoni effects. As for the surfactant mode, it is only reflected by Marangoni effects that determine the stability through the direction of the applied shear. To illuminate the competition between these two modes in response to applied shear, we consider a system with $\theta = \pi/2$ and $Re = 0$ below for eliminating both hydrostatic and inertial effects.

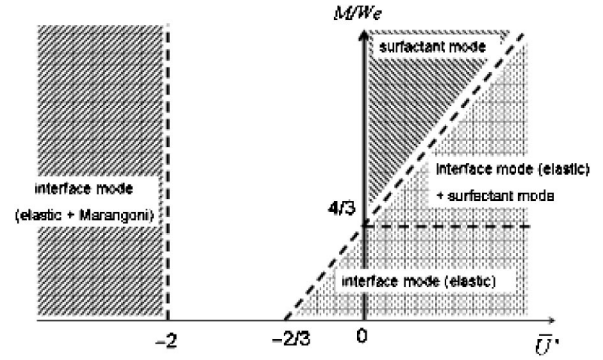


FIG. 2. Schematic phase diagram. Unstable (stable) regions are shaded (white). In unstable regions, the dominant mode and the associated effects of instability are also indicated. $Re = 0$, $\theta = \pi/2$.

For $\bar{U}' > 0$, the surfactant mode $c_1 = iM\bar{U}'/2$ is purely destabilizing. The interface mode $c_1 = i[We(\bar{U}' + 2/3) - M/2](\bar{U}' + 2)$ could be also destabilizing due to elastic effects if the applied shear can make elastic destabilization overcome the Marangoni stabilization, i.e., $\bar{U}' > M/(2We) - 2/3$. The interface mode dominates the instability if $M < 4We/3$. If $\bar{U}' < 0$, the surfactant mode now becomes stabilizing. The instability of the interface mode depends on M/We . When $M < 4We/3$, instability occurs if $\bar{U}' < -2$ or $M/(2We) - 2/3 < \bar{U}' < 0$. When $M > 4We/3$, instability takes place if $\bar{U}' < -2$. As such, $\bar{U}' < -2$ can cause instability for all M/We , which is just a consequence of the combination of both elastic and Marangoni destabilization of the interface mode. A phase diagram summarizing the instability features discussed above is illustrated in the upper half of the $\bar{U}' - M/We$ plane shown in Fig. 2. Figure 2 reveals that in most of the range of \bar{U}' , the system is unstable to long-wavelength perturbations, except in a stable window that is confined within the $-2 < \bar{U}' < 0$ region with the boundary $M/We = 2\bar{U}' + 4/3$. Such a stable window is mainly attributed to the Marangoni stabilization in the region of $-2 < \bar{U}' < 0$ modified by the elastic destabilization in the range of $-2/3 < \bar{U}' < 0$.

The above discussion is based on the absence of both inertial and hydrostatic effects. Clearly, these effects can modify the stability diagram (Fig. 3). They are only attributed to the interface mode, but not to the surfactant mode, as shown by Eqs. (40) and (41). Since the Re term of Eq. (40) is also proportional to $\bar{U}' + 2$, the stability boundary of $\bar{U}' = -2$ does not change. However, since the Re effect destabilizes the system for $\bar{U}' > 0$, it makes the line $M/We = 2\bar{U}' + 4/3$ shift toward the left, viz., $M/We = 2\bar{U}' + 4/3 + (8/15)(Re/We)$. This shrinks the stable window, depending on Re/We . As $Re = 5We$ or larger, the system becomes vulnerable to instability for all values of \bar{U}' ; a possible stabilization due to Marangoni effects for $-2 < \bar{U}' < 0$ requires M at least greater than $4We/3 + 8Re/15$. As for the effect of θ , it is evident that the smaller θ , the wider stable window due to hydrostatic stabilization.

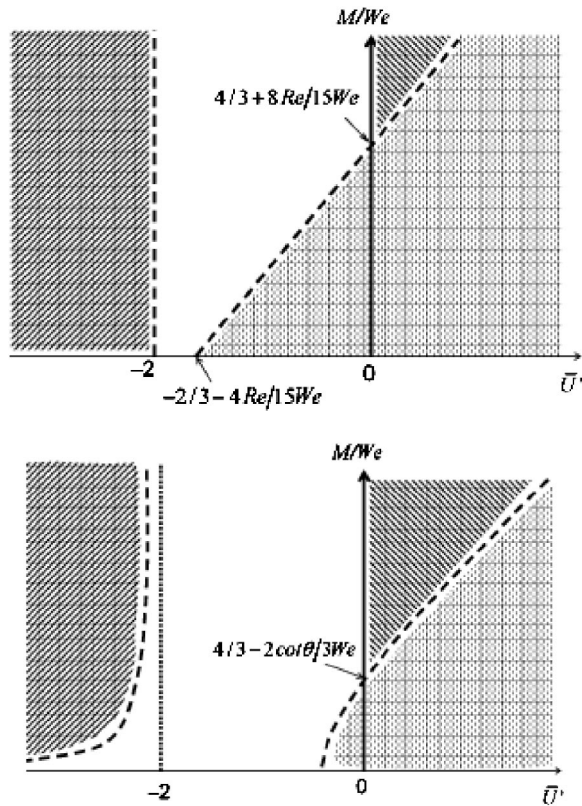


FIG. 3. Effects of Re and θ on the phase diagram. (a) The Re effect. $\theta = \pi/2$. (b) The θ effect. $Re = 0$.

Notice that in the regime of large $|\bar{U}'|$, instability could differ from that above mentioned. On the one hand, the interface mode has a large growth rate of $-ikc_1 \sim k^2 We(\bar{U}')^2$ due to elastic instability; on the other hand, the surfactant mode could have a growth rate of $M|\bar{U}'|k^{3/2}/2$. This suggests that the instability of the system could be dominated by the Marangoni destabilization unless $|\bar{U}'| \gtrsim k^{-1/2}M We^{-1}/2$ for which the elastic instability becomes at least comparable to or more important than the Marangoni one.

IV. CONCLUDING REMARKS

As shown above, we demonstrate how viscoelastic and Marangoni effects influence the stability of a falling film subjected to interfacial shear. In the absence of applied shear, elastic effects are destabilizing while Marangoni forces are stabilizing. Application of interfacial shear can make these effects stabilizing or destabilizing, depending on the strength or direction of the applied shear with respect to gravity-driven flow. The stability features are identified by the competition between the interface and surfactant modes. The interface mode is the mode usually found in free falling flow. Its stability is determined by both elastic and Marangoni forces that can act to oppose each other or reinforce their effects additively, depending on the modulation between shear and gravity-driven flows. The surfactant mode only involves Marangoni forces whose influence on stability merely relies on the action of the applied shear. For each mode, we identify the corresponding condition for occurrence of instability. Overall, the system is susceptible to instability under most condition of applied shear, as shown in the phase diagram.

Extension of the present analysis to explore nonlinear effects could be rather interesting. In principle, one can follow the standard procedures [24] to derive a coupled set of nonlinear evolution equations for the film thickness and the surfactant concentration. Although the previous weakly nonlinear stability analyses [25,26] in falling viscoelastic films without surfactant have suggested the possibility of nonlinear saturation of elastic instability, a similar study on the present system may reveal even rich dynamics. On the one hand, an instability can be excited by the exertion of interfacial shear; on the other hand, strong applied shear could suppress the instability [27]. The issue herein is to identify whether the instability can be restrained within the nonlinear regime or not. The ultimate fate of the system seems to arise from the competition between these effects, depending on the sizes of the perturbations and the ranges of the parameters.

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