

Shear-flow and thermocapillary interfacial instabilities in a two-layer viscous flow

Hsien-Hung Wei

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

(Received 19 August 2005; accepted 18 May 2006; published online 30 June 2006)

Combined effects of shear-flow and thermocapillary instabilities in a two-layer Couette flow are asymptotically examined in the thin-layer limit. The basic features of the system instability are revealed by first analyzing the two-dimensional stability problem. A scaling analysis is devised to identify dominant mechanisms in various parameter regimes. With an appropriate scaling, the leading order linear stability is reduced to a one-dimensional evolution equation containing a nonlocal contribution from viscosity stratification. Viscosity stratification destabilizes (stabilizes) the system with a more (less) viscous film, but the effect can be compromised by thermocapillary stabilization (destabilization) as the film is cooled (heated). Thermocapillary effects dominate over viscosity stratification effects for short-wave perturbations albeit the latter could be stronger than the former for long waves. The competition between these two effects gives rise to the critical Reynolds number for the onset of stability/instability. A nontrivial interplay is found within a window in the weak interfacial-tension regime. It demonstrates a possibility of the existence of two neutral states in the wavenumber space. The three-dimensional problem is also examined. For the first time, a two-dimensional film evolution equation with the inclusion of a nonlocal term is systematically derived for the corresponding stability. It can be shown analytically that three-dimensional perturbations can be more unstable than two-dimensional ones due to thermocapillarity in line with the nonexistence of Squires' theorem. The three-dimensional problem has the critical Reynolds number larger than the two-dimensional problem, but an instability in the latter does not necessarily suggest an instability in the former. An extension of each problem to the weakly nonlinear regime is also discussed in the context of the Kuramoto-Sivashinsky equation. © 2006 American Institute of Physics. [DOI: [10.1063/1.2213279](https://doi.org/10.1063/1.2213279)]

I. INTRODUCTION

As two immiscible fluids undergo shearing motions, interfacial instability could occur due to the fluids' viscosity difference. Yih¹ first identified this instability by showing that plane Couette flow of two superposed fluids is unstable to long waves when the thinner layer is more viscous. Later, Hooper² studied the long-wave stability of two-fluid Couette flow in which one fluid is overlaid by another semi-infinite layer. She also showed an instability if the lower fluid is more viscous, which was termed the "thin-layer" effect. Renardy³ analyzed the stability of two-layer Couette flow for arbitrary wavelength disturbances. For the two fluids having similar mechanical properties, the growth rate can be derived in a closed form. In the thin-layer limit, the role of viscosity stratification in determining the stability was further highlighted in accordance with the long-wave limit of Hooper.² Notice that this "thin-layer effect" is not restricted to two-layer plane flows; it also appears in multilayer systems⁴ and in core-annular flows.⁵

The above-mentioned instabilities arising from viscosity stratification mainly occur when wavelengths of disturbances are sufficiently long compared to the layer thickness. Viscosity stratification can also cause an instability for sufficiently short waves.^{6,7} However, this short-wave instability can be compromised by stabilizing effects of interfacial tension. In any case, the instability associated with viscosity stratifica-

tion must be accommodated with interfacial deflections; it is thus referred to as an "interfacial mode." Besides, there is another type of instability arising from a "wall mode." It corresponds to the classical instability of shear flow, and typically occurs at high shear rates when wavelengths are comparable to or shorter than the layer thickness.⁸ The identification of types of instabilities in two-layer flow can be found in the study of Charru and Hinch.⁹

Most of the studies on two-fluid stability are under the isothermal condition, and the stability features are fairly understood. In practice, however, fluids are often heated or cooled in processes. This could induce thermal effects and modify the features of stability. The dominant effects due to temperature variations are thermocapillarity and buoyancy. If a liquid layer is sufficiently thin, thermocapillarity generally dominates over buoyancy.¹⁰ Thermocapillarity occurs when the temperature along the fluid-fluid interface is not uniform. It induces a tension-gradient force, driving the fluid toward the higher-tension (lower-temperature) regions. Early studies^{10,11} on the thermocapillary instability of a stationary liquid layer suggested that thermocapillarity destabilizes systems with heated walls. Goussis and Kelly¹² examined the instability of a liquid film flowing down an inclined heated wall. They pointed out that there are two mechanisms responsible for thermocapillary instability. One arises from the interaction between the basic temperature and the disturbed

velocity field. The other is due to the modulation of the basic temperature at the deflected interface.

Gumerman and Homay (hereafter referred to as GH) performed a series of studies on the stability of a two-fluid Couette-Bénard problem. Their setting appears more general since they include all ingredients affecting the stability: buoyancy, interfacial-tension gradient and shear flow. We briefly review their work below. Part I¹³ of GH's work examined linear stability. One among their major findings is the nonexistence of Squire's theorem due to the competition between the three effects mentioned above. In fact, Squire's theorem does not always hold in the presence of gravity.¹⁴ Using a long-wavelength analysis,¹ GH illustrated the instability mainly driven by buoyancy and its modulation with the shear flow; thermocapillarity seems to merely serve a correction to their analysis. Part II¹⁵ applied an energy method to determine global stability. Under certain conditions, the global stability results are close to those of existing linear analyses only for low Reynolds numbers. In Part III,¹⁶ they experimentally examined effects of shear on instabilities that arise from interfacial tension gradient, buoyancy, and their combination. Their observations are generally in qualitative agreement with their theoretical predictions.

While GH's studies include all relevant effects, their efforts are mainly focused on the buoyancy-dominated instability. In this regime, their results are relatively insensitive to the details of the basic flow; the effect of the basic flow on the stability (via the classical one-fluid shear instability) appears merely important when the Reynolds number is sufficiently high.¹³ At another extreme, however, thermocapillarity and shear effects could dominate the instability at low/moderate Reynolds numbers. In this case, the shear flow can affect the stability through viscosity stratification, rather than through the classical shear instability occurring in the high Reynolds-number regime. Therefore, the interplay between thermocapillarity and the shear flow might lead the instability characteristics to differ from those found by GH. In addition, since GH did not explore all ranges of the parameters (see Ref. 13), it is also not clear how various effects play roles in determining stabilities in different parameter regimes. If one can understand instabilities in certain limiting cases, insights gained from which might provide some useful information about the complete instability pictures including all effects.

The main theme of this paper is to study combined effects of thermocapillarity and shear flow on the stability of two-layer Couette flow. We assume that the fluids' densities are matched, so gravity/buoyancy effects are excluded. To ensure negligible natural convection compared to thermocapillarity, we also assume that for each fluid layer the ratio of natural convection velocity scale $\beta_T l_c^2 g \Delta T^* / \mu$ to thermocapillary velocity scale $E^* l_c \Delta T^* / (\mu L)$ is sufficiently small; that is $\beta_T l_c^2 (L/l_c) g \Delta T^* / E^* \ll 1$. Here β_T is the thermal expansion coefficient, l_c is the fluid thickness, L is the longitudinal length scale, g is the gravitational acceleration, ΔT^* is the temperature difference between the two plates, μ is the viscosity, and E^* is a measure of the susceptibility of the interfacial-tension change to temperature variation. From the

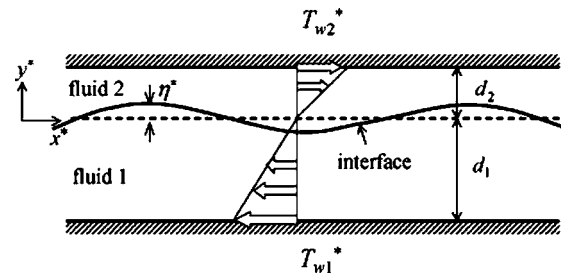


FIG. 1. Two-layer Couette flow in a channel.

above criterion, it is clear that for a given ΔT^* a sufficiently thin layer (large L/l_c) will make thermocapillarity outweigh buoyancy. We also restrict our attention to the case where one fluid thickness is much thinner than the other. This enables us to apply lubrication theory to asymptotically examine the features of stability. As we will demonstrate, the stability is dictated by the dynamics of the film and can be characterized by an evolution equation for the interface. It not only provides complementary views to previous studies,^{3,13} but also offers a lucid way to illuminate interplays among various effects. The paper is organized as follows. We begin with the base state, governing equations and boundary conditions in Sec. II. For simplicity, we first consider the stability problem to be two dimensional. In Sec. III, we devise a scaling analysis to identify dominant instability mechanisms. The leading order linear stability is formulated in Sec. IV. In Sec. V, we examine the three-dimensional stability and compare it with the two-dimensional counterpart. Extension to the weakly nonlinear regime is discussed in Sec. VI. We conclude the paper in Sec. VII.

II. BASE STATE, GOVERNING EQUATIONS, AND BOUNDARY CONDITIONS

Consider two immiscible liquid layers flowing in a channel as undergoing a shear action exerted by two parallel plates. The bottom layer is occupied by the fluid 1 of viscosity μ_1 and thickness d_1 . It is overlaid by the second fluid of viscosity μ_2 and thickness d_2 . Densities of both fluids are matched and denoted by ρ . It is more convenient to analyze the system in a frame moving with the steady-state interfacial velocity. With respect to the interface, the bottom plate moves with a speed of U_w , and the top does with $\mu_1 U_w / \mu_2$ in the opposite direction. The characteristic length and velocity are chosen as d_1 and U_w , respectively. The pressure scale is $\mu_1 U_w / d_1$ and time is scaled by d_1 / U_w . The flow system is defined in Cartesian coordinates aligned with the plates: x is the streamwise direction, and y is the direction normal to the interface defined at $y=0$, see Fig. 1. Let u and v be the velocity components in the x and y directions, respectively, and p be the pressure. Defining the viscosity ratio $m = \mu_2 / \mu_1$ and the thickness ratio $d = d_2 / d_1$, we write the basic flows in the dimensionless form

$$\bar{U} = y, \quad -1 \leq y \leq 0, \quad (1)$$

$$\bar{U} = y/m, \quad 0 \leq y \leq d.$$

The temperatures of the top and bottom plates are maintained at T_{w2}^* and T_{w1}^* , respectively. Define the dimensionless temperature $T = (T^* - T_{w1}^*) / (T_{w2}^* - T_{w1}^*)$ and the thermal-conductivity ratio $\lambda = k_2/k_1$. The basic temperature profile for each fluid can be expressed as

$$\bar{T} = \frac{\lambda}{\lambda + d}(y + 1), \quad \text{for } -1 \leq y \leq 0, \quad (2)$$

$$\bar{T} = \frac{1}{\lambda + d}(y - d) + 1, \quad \text{for } 0 \leq y \leq d.$$

For the stability problem, we first consider it to be two dimensional for simplicity. Extension to the three-dimensional problem will be analyzed later in Sec. V. Since a dynamic flow system consists of contributions from both base state and perturbations, we start with general formulation prior to performing linearization. For the two-dimensional problem, the nondimensional equations of each layer are governed by the continuity and the Navier-Stokes equations:

$$u_x + v_y = 0, \quad (3)$$

$$\text{Re}_1(u_t + uu_x + vv_y) = -p_x + m_i \nabla^2 u, \quad (4a)$$

$$\text{Re}_1(v_t + uv_x + vv_y) = -p_y + m_i \nabla^2 v. \quad (4b)$$

$\text{Re}_1 = \rho U_w d_1 / \mu_1$ is the Reynolds number based on the bottom layer. Here $m_i = 1$, for $i=1$, or $= m$ for $i=2$. The system is subject to the following boundary conditions. On the walls:

$$u_1 = -1, \quad v_1 = 0, \quad \text{at } y = -1, \quad (5)$$

$$u_2 = d/m, \quad v_2 = 0, \quad \text{at } y = d. \quad (6)$$

Define the jump notation $[] := (\)_1 - (\)_2$. Continuous velocities at the deformed interface $y = \eta(x, t)$ demand

$$[u] = [v] = 0. \quad (7)$$

The tangential stress and normal stress conditions are applied at $y = \eta(x, t)$:

$$\frac{1}{(1 + \eta_x^2)} [(u_y + v_x)(1 - \eta_x^2) + 2(v_y - u_x)\eta_x] = \frac{1}{\text{Ca}} \sigma_x, \quad (8)$$

$$\begin{aligned} & -[p - 2v_y - (-p + 2u_x)\eta_x^2 + 2(v_y + u_x)\eta_x] \\ & = \frac{\sigma(T)}{\text{Ca}} \eta_{xx}(1 + \eta_x^2)^{-3/2}, \end{aligned} \quad (9)$$

where $\text{Ca} = \mu_1 U_w / \sigma_0^*$ is the capillary number with σ_0^* being the interfacial tension at the reference temperature T_0^* . We assume the dependence of the interfacial tension on temperature to be linear:

$$\sigma = 1 - E(T - T_0), \quad (10)$$

where $E = -(T_{w2}^* - T_{w1}^*)(\partial \sigma^* / \partial T^*)_{T_0^*} / \sigma_0^*$ is a dimensionless parameter that reflects the change of the interfacial tension in response to the temperature variation. Here $E > 0 (< 0)$ corresponds to heating (cooling) the top layer, viz., $T_{w1}^* < T_{w2}^* (T_{w1}^* > T_{w2}^*)$. Finally, we require the kinematic condition at the interface:

$$v = \eta_t + u \eta_x. \quad (11)$$

For the heat transfer problem, the governing equation for each layer is

$$T_t + u T_x + v T_y = \frac{1}{\text{Pe}_i} \nabla^2 T, \quad (12)$$

where $\text{Pe}_i = U_w d_1 / \alpha_i$ ($i=1, 2$) is the Peclet number. $\alpha_i = k_i / \rho c_p$ is the thermal diffusivity with the specific heat capacity c_p . The boundary conditions are given by

$$T_1 = 0, \quad \text{at } y = -1, \quad T_2 = 1, \quad \text{at } y = d. \quad (13)$$

$$T_1 = T_2, \quad \text{at } y = \eta(x, t), \quad (14)$$

$$T_{1y} - \eta_x T_{1x} = \lambda(T_{2y} - \eta_x T_{2x}), \quad \text{at } y = \eta(x, t). \quad (15)$$

III. SCALING ANALYSIS

With the base state above, we now begin to analyze the corresponding stability. To asymptotically examine the stability in the thin-layer limit, we shall first carry out a scaling analysis to identify dominant mechanisms of stability. It also provides approximation for the sizes of perturbation quantities. We then expand governing equations and boundary conditions with perturbation quantities to derive the relevant equations governing the system's leading order stability.

Suppose that the top layer thickness is much thinner compared to the bottom layer, i.e., $d = \varepsilon \ll 1$. We introduce a stretched variable $Y = y/\varepsilon$ to the problem so that one can separate the transverse length scale of the thin layer from that of the thick layer. For $m = O(1)$, the basic flows can be rewritten as $\bar{U}_1 = y$ and $\bar{U}_2 = \varepsilon Y/m$ for the bottom and top layers, respectively. Also, assuming $\lambda = O(1)$, the leading order basic temperature profiles are expressed as

$$\bar{T}_1 = 1 + y - \frac{\varepsilon}{\lambda}(1 + y) + O(\varepsilon^2) \quad (-1 \leq y \leq 0), \quad (16)$$

$$\bar{T}_2 = 1 + \frac{\varepsilon}{\lambda}(Y - 1) + O(\varepsilon^2) \quad (0 \leq Y \leq 1).$$

Now consider the system perturbed by temperature disturbances, say, T'_1 and T'_2 for the thick and the thin layers, respectively. Suppose that T'_1 for the thick layer is of size $\delta_T (\ll 1)$. Then a continuous heat flux (15) across the interface demands $T'_2 \sim \varepsilon \delta_T / \lambda$ for the film. In what follows, for $\lambda = O(1)$ the interface feels most of the temperature variations from the thick layer, and so does the subsequent thermocapillary force.

To estimate the sizes of perturbation flow quantities, we consider the system perturbed by an interfacial disturbance

of size $\delta(\ll \varepsilon)$. Let (u', v', p') and (U', V', P') denote the perturbation flow quantities for the thin and the thick layers, respectively. In the thin-layer limit, the system stability is usually dictated by the dynamics of the film; we thus first inspect the relevant scales. The size of u' depends on flow mechanisms; the film flow can be sheared either by the adjacent thick fluid through viscosity stratification or by thermocapillary forces. It can be further driven by capillary pressures furnished by interfacial tension forces. Below we first consider viscosity stratification and thermocapillary effects, and then discuss the influence of the interfacial tension later.

We begin with estimating the scale of u' due to viscosity stratification. To do so, one has to find its scale relationship with U' . With the aid of the boundary condition (8), we find $u' \sim (\varepsilon/m)U'$. Expanding $[u]=0$ in (7) and writing it in the linearized form: $U' + \bar{U}_{1y}|_{y=0} \delta\xi = u' + \bar{U}_{2y}|_{y=0} \delta\xi$ [where $\delta\xi$ is an interfacial perturbation and ξ is an $O(1)$ function], we find that a perturbation to the interface causes a jump in the disturbance of the horizontal velocity across the unperturbed interface due to viscosity stratification. Since $u' \sim (\varepsilon/m)U'$, this perturbation will be perceived most by the thick layer and induces a velocity of scale $U' \sim (1-m^{-1})\delta$ to the thick layer. Both V' and P' have the same scales as U' due to a lack of separation in length scales in the thick layer. This velocity then generates a tangential stress on the interface, shearing the thin layer through (8) and in turn giving rise to $u' \sim \varepsilon(1-m^{-1})\delta/m$.

The temperature jump at the interface due to the thermal-conductivity difference in (14) provides a scale relationship between the interfacial and temperature perturbations: $\delta_T \sim (1-\lambda^{-1})\delta$. Hence, the thermocapillary force in (8) has a scale of $E\delta_T/Ca \sim (1-\lambda^{-1})E\delta/Ca$ and induces a velocity $u' \sim \varepsilon(1-\lambda^{-1})E\delta/(mCa)$ to the thin layer. Compared to the viscosity-stratification velocity scale $u' \sim \varepsilon(1-m^{-1})\delta/m$, the relative importance of thermocapillary to viscosity-stratification effects can be reflected by $(1-m^{-1})^{-1}(1-\lambda^{-1})E/Ca$. If the contrasts of mechanical and thermal properties between the two fluids are sufficiently manifest, say, both $(1-m^{-1})$ and $(1-\lambda^{-1})$ are $O(1)$, the prior scaling ratio can be approximated as E/Ca .

The above discussion is focused on the perturbation flows induced by viscosity stratification and thermocapillarity. We now include the influence of the interfacial tension and discuss the relevant scaling. In the thin-layer limit, for a sufficiently strong tension the capillary force usually dominates in the normal stress (9), and furnishes a pressure force $p' \sim \delta/Ca$ for the film. This pressure drives the film flow with $u' \sim \varepsilon^2\delta/(mCa)$ according to (4a). The ratio of this capillary velocity scale to the viscosity-stratification-induced velocity of $O(\varepsilon\delta/m)$ is thus ε/Ca , which reflects their relative influence on the stability.

As discussed above, we have established the relative velocity scales E/Ca and ε/Ca for thermocapillarity and capillarity, respectively, compared to viscosity stratification. With these scaling ratios, dominant effects can be identified when all the three effects are present. In view of the fact that both scaling ratios depend on Ca , we consider three different cases: (i) $Ca \gg \varepsilon$; (ii) $Ca \sim \varepsilon$; and (iii) $Ca \ll \varepsilon$. For case (i),

capillary effects are weak. The instability is determined by thermocapillarity (when $E \gg Ca$), viscosity stratification (when $E \ll Ca$), or their combination (when $E \sim Ca$). For case (ii) the tension is moderately strong, so its influence on the stability is comparable to viscosity stratification. Thermocapillary effects are not comparable to the other two until $E \sim \varepsilon$. As for case (iii), capillary effects dominate over viscosity stratification effects. The stability is mediated by thermocapillary effects if $E \gg Ca$.

In fact, the *Ansatz* can be generalized under the condition $Ca \sim E \sim \varepsilon$ in which all the three effects are comparable since other scenarios can be treated by taking appropriate limits, as discussed above. Using this scaling, we find $u' \sim \varepsilon\delta$, $v' \sim \varepsilon^2\delta$, and $p' \sim \varepsilon^{-1}\delta$ for the film. In order to capture nontrivial dynamics for the interface, we require a long time scale $t \sim \varepsilon^{-2}$ in the kinematic condition (11).

A few remarks on our analysis are made below. Since our approach is based on lubrication theory in the film, it requires $\varepsilon^3 \text{Re}_1/m \ll 1$ for neglecting the film's inertia. To further ensure the validity of the theory, disturbance wavelengths must be longer than the film thickness, viz. $k^{-1} \gg \varepsilon$. Hence the results that follow are not limited to wavelengths longer than the film thickness, but applicable to any wavelengths as long as the above constraint for k is satisfied. Finally, we assume the absence of gravity effects because of matched densities, so Rayleigh-Taylor instability will not occur in our system. Nevertheless, as we will show, gravity appears to have similar effects on the system's stability as thermocapillarity even though instability mechanisms of these two are seemingly different.

In the next section, we outline the formulation for the leading order stability based upon the above scaling, and then discuss the results derived therefrom.

IV. ANALYSIS OF THE LEADING ORDER LINEAR STABILITY

Define $M = E/Ca$ be the Marangoni number. For $Ca \sim \varepsilon$ and $E \sim \varepsilon$, we let $Ca = \varepsilon Ca_0$, with $Ca_0 = O(1)$ and $M = O(1)$. Dynamic quantities relevant to the stability can be expanded as follows:

$$\begin{aligned} u_1 &= \bar{U}_1 + \delta U + O(\varepsilon\delta), & v_1 &= \delta V + O(\varepsilon\delta), \\ p_1 &= \delta P + O(\varepsilon\delta), \\ u_2 &= \bar{U}_2 + \varepsilon \delta u + O(\varepsilon^2\delta), & v_2 &= \varepsilon^2 \delta v + O(\varepsilon^3\delta), \\ p_2 &= \varepsilon^{-1} \delta p + O(\delta), \\ \eta &= \delta\xi + O(\varepsilon\delta), & T_1 &= \bar{T}_1 + \delta\theta_1 + O(\varepsilon\delta), \\ T_2 &= \bar{T}_2 + \varepsilon \delta\theta_2 + O(\varepsilon^2\delta). \end{aligned} \tag{17}$$

After substituting the above into (3)–(15) and expanding them with respect to the base state, the leading order governing equations of the perturbation film flow become

$$u_x + v_y = 0, \tag{18a}$$

$$0 = -p_x + mu_{YY}, \quad (18b)$$

$$0 = -p_Y. \quad (18c)$$

The film flow field that satisfies the no-slip conditions at the wall $Y=1$ is

$$u = \frac{1}{2m} p_x (Y^2 - 1) + A(Y - 1), \quad (19a)$$

$$v = -\frac{1}{2m} p_{xx} \left(\frac{Y^3}{3} - Y + \frac{2}{3} \right) - A_x \left(\frac{Y^2}{2} - Y + \frac{1}{2} \right), \quad (19b)$$

where A is an as-yet-undetermined coefficient. The leading order tangential and normal stress conditions at the unperturbed interface are

$$mu_{Y'}(Y=0) - (U_{y'} + V_{x'})_{y=0} = M[\xi + \theta_1(y=0)]_{x'}, \quad (20)$$

$$p = \frac{\xi_{xx}}{Ca_0}. \quad (21)$$

Note that in (20), the temperature perturbation involves the variation of the basic temperature $\bar{T}_{1y}|_{y=0}\xi$ due to deflections of the interface. The leading order kinematic condition is

$$v = \xi_{\tau}, \quad (22)$$

where we have invoked the long time scale $\tau = \varepsilon^2 t$.

To determine the film flow solution (19) further requires the thick-layer flow solution due to their coupling through (20). For the thick layer the leading order governing equations are

$$U_x + V_y = 0, \quad (23a)$$

$$Re_1(yU_x + V) = -P_x + \nabla^2 U, \quad (23b)$$

$$Re_1 y V_x = -P_y + \nabla^2 V. \quad (23c)$$

Note that the time-derivative terms are negligible because the time scale is $O(\varepsilon^{-2})$ here. The boundary conditions for (23) include the no-slip conditions at the bottom wall (5), and the continuity of velocities at the interface (7). The latter, to leading order, becomes

$$U(y=0) = \left(\frac{1}{m} - 1 \right) \xi, \quad (24a)$$

$$V(y=0) = 0. \quad (24b)$$

To solve Eqs. (23), we introduce the streamfunction Ψ such that $U = \Psi_y$ and $V = -\Psi_x$ for satisfying the continuity equation (23a). Further, taking the Fourier transform, $\hat{\Psi} = \int_{-\infty}^{\infty} \Psi e^{-ikx} dx$, we reduce (23b) and (23c) to

$$ik Re_1 y (D^2 - k^2) \hat{\Psi} = (D^2 - k^2)^2 \hat{\Psi}, \quad (25)$$

where $D \equiv d/dy$. The solution is given by

$$\hat{\Psi} = k^{-1} \hat{a} \int_{-1}^y \sinh[k(y-y')] [g_2 f_1(y') - g_1 f_2(y')] dy', \quad (26)$$

with

$$\hat{a} = \frac{\left(\frac{1}{m} - 1 \right) \hat{\xi}}{\int_{-1}^0 \cosh(ky) [g_2 f_1(y) - g_1 f_2(y)] dy},$$

$$f_1(y) = \text{Ai}[e^{i\pi/6} (k Re_1)^{1/3} (y - ik Re_1^{-1})],$$

$$f_2(y) = \text{Ai}[e^{i5\pi/6} (k Re_1)^{1/3} (y - ik Re_1^{-1})],$$

$$g_1 = \int_{-1}^0 \sinh(ky) f_1(y) dy, \quad g_2 = \int_{-1}^0 \sinh(ky) f_2(y) dy.$$

Here Ai is Airy's function. With the solution above, we first rewrite $(U_x + V_y)_{y=0} = \Psi_{yy}(y=0)$ for (20) in the Fourier space as

$$\hat{\Psi}_{yy}(y=0) = \hat{a} \int_{-1}^0 \sinh(ky) [f_2(y) f_1(0) - f_1(y) f_2(0)] dy; \quad (27)$$

$(U_y + V_x)_{y=0}$ can then be expressed in terms of ξ by taking the inverse Fourier transform for (27).

Finally, we need the perturbed temperature field in the thick layer for describing thermocapillary forces in (20). To leading order, (14) yields

$$\theta_1(y=0) = (\lambda^{-1} - 1) \xi. \quad (28)$$

Combining $\bar{T}_{1y}|_{y=0}\xi$, we reduce the perturbed thermocapillary force in (20) to $M\lambda^{-1}\xi$. As such, how the temperature varies along the interface can be simply reflected by an interfacial deflection. The effects of the induced thermocapillary force on the interface motion can be understood below. Consider a sinusoidal perturbation to the interface. For a heated film layer ($M > 0$), the interface crests (the thinnest portion of the film) are hotter than the troughs (the thickest portion of the film). This induces a tension-gradient force and shears the fluid toward the troughs, making the interfacial deflection amplify as a consequence of mass conservation. Similarly, $M < 0$ does the opposite, and hence has a stabilizing influence on the system.

Furthermore, the strength of the thermocapillary force depends on the thermal-conductivity contrast between the two fluids. This can be explained below. If the thin layer is less conductive ($\lambda < 1$), its basic temperature profile is steeper for a given temperature difference across the channel. It follows that a small perturbation to the interface will give rise to a large temperature variation along the deflected interface, thereby inducing a greater thermocapillary force.

As such, the film velocities (19) can be solved in terms of ξ ; there is no need in solving the detailed solution of θ_1 .

Substituting the film solution into (22) yields the following evolution equation that governs the leading order linear stability:

$$\begin{aligned} \xi_\tau + \frac{M}{2m\lambda} \xi_{xx} + \frac{1}{3m \text{Ca}_0} \xi_{xxxx} + \frac{1}{2\pi m} \\ \times \left(\frac{1}{m} - 1 \right) \int_{-\infty}^{\infty} G(k) \int_{-\infty}^{\infty} \xi(x', \tau) e^{ik(x-x')} dx' dk = 0, \end{aligned} \quad (29)$$

with

$$G = \frac{ik \int_{-1}^0 \sinh(ky) [f_2(y)f_1(0) - f_1(y)f_2(0)] dy}{2 \int_{-1}^0 \cosh(ky) [g_2 f_1(y) - g_1 f_2(y)] dy}.$$

As shown by (29), physics associated with each term now emerge. The second-derivative term arises from thermocapillarity. A similar term can also arise from hydrostatic effects if the fluids' densities are mismatched.¹⁷ The fourth-derivative term is attributed to capillarity. The nonlocal term reflects viscosity stratification effects through the coupling to the dynamics of the thick layer. Although Eq. (29) is derived based on $\text{Ca} \sim \varepsilon$ and $M = E/\text{Ca} \sim O(1)$, it is still applicable in various limits, as discussed in the scaling analysis. That is, $\text{Ca}_0 \rightarrow 0$ and $\text{Ca}_0 \rightarrow \infty$ should be regarded as $\text{Ca} \ll \varepsilon$ (strong tension limit) and $\text{Ca} \gg \varepsilon$ (weak tension limit), respectively. Both small and large M limits can also be interpreted in a similar manner.

To analyze the linear stability, we use a normal mode: $\xi = \hat{\xi} \exp(ikx + s\tau)$ where k is the wave number and s is the complex growth rate. The system is unstable when the real part of s , s_r , is positive. The normal-mode form of (29) then becomes

$$s = \frac{M}{2m\lambda} k^2 - \frac{1}{3m \text{Ca}_0} k^4 + \frac{1}{m} \left(1 - \frac{1}{m} \right) G(k, \text{Re}_1). \quad (30)$$

Note that all the terms in (30) have a common factor $1/m$. Since s is measured with respect to the basic flow velocity scale of the thick layer, i.e., the *dimensional* growth rate scale is $\varepsilon^2 U_w / d_1$, the growth rate $m \cdot s$ should be understood on the basis of the thin-layer flow scale.

To gain more insight into the instability features, we first inspect (30) in the long-wave limit. Making use of the $k \rightarrow 0$ asymptote of $G: G = 2ik + (\text{Re}_1/60)k^2 + O(k^3)$ (see Appendix), we find

$$s = \frac{2i}{m} \left(1 - \frac{1}{m} \right) k + \left[\frac{M}{2m\lambda} + \frac{\text{Re}_1}{60m} \left(1 - \frac{1}{m} \right) \right] k^2 + O(k^3). \quad (31)$$

As a result, the $O(k)$ effect is only dispersive to the long-wave stability. The long-wave stability is determined at $O(k^2)$ by both thermocapillarity and viscosity stratification. The effect of the interfacial tension is $O(k^4)$ unless the tension is strong enough [viz., $\text{Ca}_0 \leq O(k^2)$] to compete with the other two. Thermocapillary effects are destabilizing (stabiliz-

ing) for $M > 0 (< 0)$. The viscosity-stratification term agrees with Yih¹ in the thin-layer limit. It reveals that $m > 1$ (i.e., a more viscous thin layer) is destabilizing to long waves, whereas $m < 1$ is stabilizing. Therefore, a competition between thermocapillarity and viscosity stratification can take place either when $M > 0$ and $m < 1$ or when $M < 0$ and $m > 1$. In these situations, there exists a critical Reynolds number, Re_1^* , at the marginal stability state by setting the $O(k^2)$ part of (31) to be zero:

$$\text{Re}_1^* = \frac{30m}{\lambda} \left| \frac{M}{(m-1)} \right|. \quad (32)$$

For $M > 0$ and $m < 1$, (32) provides the minimum Re_1 required for stability while for $M < 0$ and $m > 1$, it is the maximum Re_1 below which stability can be sustained. Since the velocity scale in (32) appears in both Re_1^* and M , the *dimensional* velocity U^* at the criticality given by (32) is

$$U^* = \left(\frac{30m}{\rho d\lambda} \left| \left(\frac{\partial \sigma^*}{\partial T^*} \right)_{T_0^*} \frac{(T_{w1}^* - T_{w2}^*)}{(m-1)} \right| \right)^{1/2}. \quad (33)$$

For given fluid properties and heat conditions, this is the critical velocity for the onset of stability/instability. Physically, for long-wave perturbations, dominant driving forces to perturbation flows are the thermocapillary force and the thick-layer shear stress in view of (20). Following this line, we can also obtain (33) using a simple scaling argument below. The thick-layer shear stress through viscosity stratification needs to accompany the inertia in order to make the instability at work (since an inertialess flow will not have instability due to the reversibility of Stokes flow); thus it has a scale of $O[\rho U^2(1-m^{-1})d/L]$, where L is the perturbation wavelength. Define $\Delta T \equiv T_{w1}^* - T_{w2}^*$. The thermocapillary force is $O[(\partial \sigma^* / \partial T^*) \Delta T^* / (\lambda L)]$. The critical velocity occurs when these two forces counterbalance. This at least requires balancing their scales: $\rho U^2(1-m^{-1})d/L \sim (\partial \sigma^* / \partial T^*) \Delta T^* / (\lambda L)$, which arrives at the same result given by (33). Also, note that U^* here does not depend on the interfacial tension σ_0^* as a consequence of negligible interfacial-tension effects on the long-wave stability.

In the short-wave limit, with the aid of the $k \rightarrow \infty$ asymptote of $G: G = ik^2 - ik + \frac{\text{Re}_1}{4} + O(k^{-1})$, we have

$$\begin{aligned} s = -\frac{1}{3m \text{Ca}_0} k^4 + \left[\frac{i}{m} \left(1 - \frac{1}{m} \right) + \frac{M}{2m\lambda} \right] k^2 - \frac{i}{m} \left(1 - \frac{1}{m} \right) k \\ + \frac{\text{Re}_1}{4m} \left(1 - \frac{1}{m} \right) + O(k^{-1}). \end{aligned} \quad (34)$$

Equation (34) holds in the sense that $1 \ll k \ll \varepsilon^{-1}$, for ensuring the validity of lubrication in the film. More precisely, to retain the strongest effect $O(k^4)$ in (34) due to the interfacial tension, it entails the unsteady term $\text{Re}_1 u_t (\sim \text{Re}_1 \varepsilon^2 k^4)$ to be much smaller than the viscous term $u_{yy} (\sim \varepsilon^{-2})$ in (4a), viz., $k \ll \text{Re}_1^{-1/4} \varepsilon^{-1}$ for preserving lubrication in the film. In addition, $k \ll \varepsilon^{-1/2}$ is also required for neglecting the unsteady term in the motion of the thick layer, i.e., $U_t \sim \varepsilon^2 k^4 \ll V \sim O(1)$ in (23b). Thereby, for $\text{Re}_1 \sim O(1)$ (more precisely, smaller than ε^{-2}), the restriction $k \ll \varepsilon^{-1/2}$ suffices to most of the situations of interest.

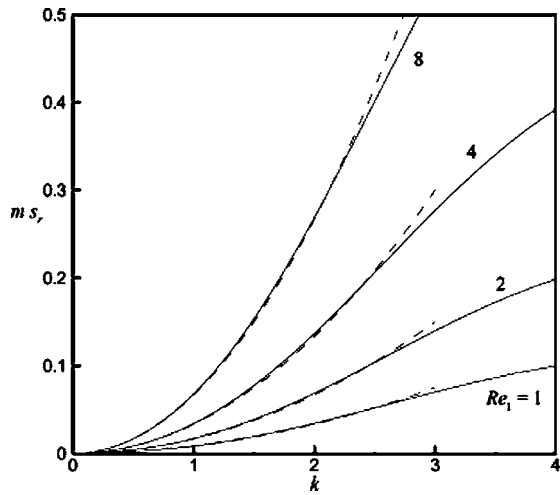


FIG. 2. The growth rates versus the wavenumber k . Here $m=2$; $M=0$; $Re_1=1, 2, 4,$ and 8 . The dashed lines are the corresponding long-wave asymptotes.

As indicated by (34), viscosity stratification determines the short-wave stability at $O(k^0)$, making the growth rate approach a constant. Clearly, such an $O(k^0)$ short-wave growth rate due to viscosity stratification will be either rapidly damped by the interfacial tension [of $O(k^4)$], or overwhelmed by thermocapillarity [of $O(k^2)$].

Intuitively, short-wave disturbances do not perceive the presence of the walls; the induced effects arising from viscosity stratification should be only localized in the neighborhood of the interface, just as in the unbounded problem.⁶ However, the present analysis is different from the unbounded problem since in the latter the short-wave instability occurs at $O(k^{-2})$. In fact, the short-wave analysis of the bounded problem does not necessarily deduce to that of the unbounded problem.^{8,18} The discrepancy lies in the fact that the only length scale in the unbounded problem is the transverse scale of the viscous momentum, whereas the bounded problem further involves a scale of the plate separation. Besides, our thin-layer *Ansatz* further imposes a restriction on $k(\ll \varepsilon^{-1})$ for assuring lubrication in the film, making the transverse length scale of the film separated from that of the thick layer. Below, we apply a scaling argument to illustrate how these length scales can lead to different orders of magnitudes for the short-wave growth rates caused merely by viscosity stratification.

With respect to the plate separation, there are three length scales in the problem: wavelength $L(=2\pi/k)$, viscous length scale l , and the thickness of the thin layer ε . For the thick-layer flow, the disturbed velocity has a scale $U' \sim \delta$ [more precisely, $\sim(1-m^{-1})\delta$] through the jump of the perturbed interfacial velocities due to viscosity stratification. Since the instability requires participation of the inertia of the thick layer, it entails the inertial term $Re_1 \bar{U}'_x \sim Re_1 l k \delta$ to balance the viscous term $\nabla^2 U' \sim U'_{xx} \sim k^2 \delta$ in the x -momentum equation. This yields the viscous length scale $l \sim k Re_1^{-1}$. The resulting shear stress at $y=0$ thus can be estimated by $U'_y \sim k^{-1} Re_1 \delta$ (because $y=0$ is near the wall $y=\varepsilon$, the normal velocity therein $V' \sim v' \sim \varepsilon^2 Re_1 \delta$ nearly

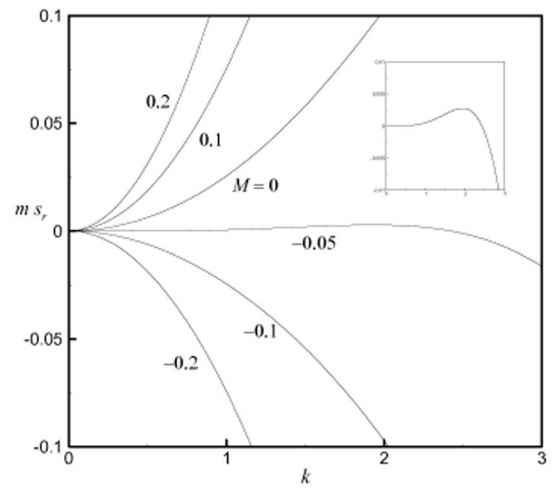


FIG. 3. Effects of thermocapillarity on the growth rate. $Re_1=3$; $m=2$; $\lambda=1$. The inset is for $M=-0.05$.

vanishes, making $V'_x \ll U'_y$ in the shear stress, as we can justify *a posteriori*). This stress drives the thin layer within which the transverse scale in the film remains $O(\varepsilon)$ and is much shorter than the viscous length scale in the thick layer, viz., $\varepsilon \ll l$. Balancing the shear stresses of the two fluids: $m\varepsilon^{-1}u'_y \sim U'_y$, we arrive at $u' \sim \varepsilon k^{-1} Re_1 \delta$, and hence $v' \sim \varepsilon^2 Re_1 \delta$ due to the continuity. Using $v' \sim \delta/t$ from the kinematic condition, the growth rate therefore has a scale of $t^{-1} \sim \varepsilon^2 Re_1$ or $\tau^{-1} \sim Re_1$, which agrees with the $O(k^0)$ term in (34).

We also demonstrate that if the above scaling scheme is not devised by balancing the inertia, then the resulting growth rate scale will be $O(k^2)$. The viscous length l is obtained by balancing $U'_{yy} \sim l^{-2} \delta$ with $U'_{xx} \sim k^2 \delta$ in the viscous term $\nabla^2 U'$. This leads to $l \sim k^{-1} (\gg \varepsilon)$. Following similar arguments outlined earlier, we have $U'_y \sim k \delta$, $u' \sim \varepsilon k \delta$, and $v' \sim \varepsilon^2 k^2 \delta$. Thereby, the growth rate has a scale $t^{-1} \sim \varepsilon^2 k^2$ or $\tau^{-1} \sim k^2$. This is again consistent with the $O(k^2)$ term of (34). Note that the growth rate of this case is purely imaginary—an instability will not occur simply because of the Stokes-flow reversibility for the lack of inertia.

With the above features gleaned in the limits of both long and short waves, we now analyze the stability for arbitrary wavelength disturbances. We first consider an isothermal system with a sufficiently weak tension (i.e., $Ca \gg \varepsilon$), so the instability manifests solely due to viscosity stratification. Figure 2 shows the growth rate curves for a more viscous thin layer ($m=2$). Here, the growth rate is represented by ms_r in measure with respect to the basic velocity scale of the film. As revealed by Fig. 2, for a fixed Re_1 the system is unstable and the growth rate increases as the wavelength becomes shorter. Increasing Re_1 exacerbates the instability. The results are also compared with those of long waves, and show an excellent agreement for $k \leq 2$. The agreement is also fairly good up to $k \sim 3$. For $m < 1$ (not shown), it is clear that the system is stable because $(1-m^{-1}) < 0$ in (30). It thus follows that increasing Re_1 expedites the stabilization in contrast to $m > 1$. The above stability features are consistent with those found by Renardy³ for the fluids with nearly matched

viscosities. The present work also complements her study in a sense that here we provide an alternative view for understanding the thin-layer effects.

Figure 3 shows the effects of thermocapillarity on the system stability with $Re_1=3, \lambda=1$ and $m=2$. Since $M>0$ (the heated thin layer) is destabilizing, it makes the growth rate faster than that of the isothermal case ($M=0$). $M<0$ (the cooled thin layer) is stabilizing and hence suppresses the growth. The critical state between instability and stability occurs at $M\sim-0.05$ below which the system is completely stabilized by thermocapillarity. This critical M also agrees with the long-wave estimate given by (34). Also, for $M=-0.05$ (see the inset of Fig. 3), the growth rate for small k is positive and increases with k . It reaches a maximum at $k\approx 2$, and then decays to a negative value at larger k with a neutral state at $k\approx 2.5$. This can be explained by the fact that the viscosity-stratification destabilization is slightly stronger than the thermocapillary stabilization for small and moderate waves, but the former is suppressed by the latter for short waves, in view of (34).

Figure 3 suggests that a slight change in M gives rise to a large change of the growth rate. In practice, the system stability is rather sensitive to thermocapillary effects. This can be seen by inspecting both long and short-wave results. For long waves as in (31), the growth rate due to viscosity stratification is proportional to $k^2 Re_1/60$ as opposed to $k^2 M/\lambda$ of thermocapillarity; thus thermocapillarity is generally more prevailing than viscosity stratification unless Re_1 or λ is sufficiently large. As for short waves as in (34), thermocapillarity clearly dominates over viscosity stratification. Therefore, for a given arbitrary wavelength thermocapillarity generally outweighs viscosity stratification effects even if the temperature difference across the plates is small. For example, consider a system with $-(\partial\sigma^*/\partial T^*)\sim 0.1$ dyne/cm $^\circ\text{C}$ and $\mu_1=0.1$ poise. Suppose that $M\sim 0.1$ suffices to cause an effect. For $U^*\sim 1$ cm/s, the required temperature difference for thermocapillarity at work is merely 0.1 $^\circ\text{C}$!

In fact, when the interfacial tension is negligible, the combined effects of viscosity stratification and thermocapillarity on the stability for $m>1$ can be used to infer those for $m<1$, or vice versa. It can be shown as follows. Letting $s'(M, m)=ms(M, m)$, we find

$$s'[-M, m/(2m-1)] = -s'(M, m). \quad (35)$$

Since $m' = m/(2m-1) < 1$ for $m > 1$, a stability for $m > 1$ and $M < 0$ suggests an instability for $m < 1$ and $M > 0$ via the property (35). That is, if there is a critical $M^* (< 0)$ below which a flow with $m > 1$ is stable, then there must exist $M^* \rightarrow -M^* (> 0)$ beyond which a flow with $m \rightarrow m/(2m-1) < 1$ is unstable.

The discussion so far excludes capillary effects, or is concerning a scenario that capillary forces are too weak (i.e., $Ca \gg \varepsilon$) to influence the stability compared to viscosity stratification or thermocapillarity. Since capillarity is always stabilizing, it breaks the symmetry of (35). Also, since the capillary growth rate decays at the rate of $O(k^4)$, the stabilizing effect is strong for moderate or short waves. In general, capillarity is often robust to completely stabilize the

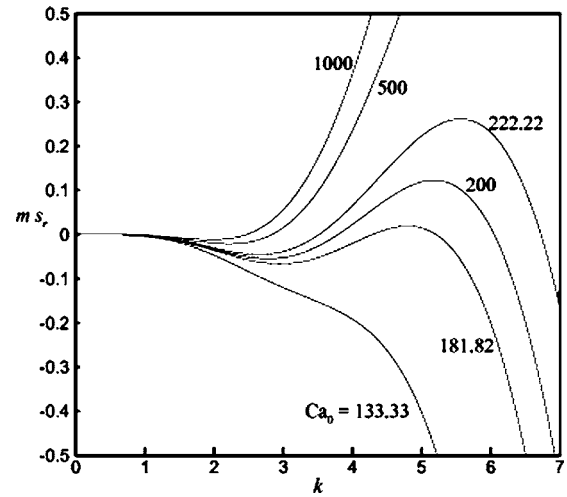


FIG. 4. An example of the existence of two critical states in the k -space in the weak tension regime. Here $Re_1=6; m=0.5; \lambda=1; M=0.05$. Here M is chosen such that viscosity-stratification stabilization is nearly offset by thermocapillary destabilization according to (32).

system. If capillarity is sufficiently weak (but not too weak), however, it could have some nontrivial effects on the stability for $m < 1$. Such an incident occurs when M is equal or close to the critical M^* estimated by (32). This is illustrated by Fig. 4 for $Re_1=6, m=0.5, \lambda=1$, and $M=M^*=0.2$. The result reveals that there are two critical wavenumbers k_c 's in a range of large Ca_0 . In the long-wave regime, the viscosity-stratification stabilization is offset by the thermocapillary destabilization at $M=M^*$ according to (32). In this regime, the capillary stabilization is of a higher order [of $O(k^4)$] in view of (31), and hence has no noticeable influence on the stability, as evidenced in Fig. 4. For moderate waves, the capillary stabilization becomes stronger; it works together with the $m < 1$ stabilizing effect, making the growth rate negative, and decreased with k . Meanwhile, the thermocapillary destabilization starts to become important, thus the growth rate increases with k and reaches the first neutral state at some k . The growth rate keeps increasing with k until a sufficiently large k for which the capillary stabilization becomes robust. The growth rate then decreases with k again, leading to the second neutral state.

The above result arises from the competition among all the three effects in a range of k , and only occurs in an appropriate window of Ca . If Ca is too small, then the system is completely stabilized by capillarity for all k . If Ca is too large, on the other hand, then only viscosity stratification and thermocapillarity compete to determine the stability. For parameters given in Fig. 4, the window occurs within $180 < Ca_0 < 1000$. For $\varepsilon=0.1$, the corresponding Ca ranges from $O(\varepsilon^{-1})$ to $O(\varepsilon^{-2})$, indicating that the required tension for such an occurrence is small.

V. THREE-DIMENSIONAL LINEAR STABILITY AND SQUIRE'S THEOREM

In the preceding sections, we consider the system subjected to two-dimensional perturbations and characterize the linear stability. If perturbations are three dimensional, two

issues will be raised. First, the condition at the criticality between stability and instability might change. Second, it is not clear if three-dimensional perturbations can grow more rapidly than two-dimensional ones, which is, in particular, critical to determining the fate of the interface.

The relationship between the behavior of three-dimensional perturbations and that of two-dimensional perturbations has been established by Squire.¹⁹ The theorem states that if there is an instability in the three-dimensional problem at a certain Reynolds number, then the equivalent two-dimensional problem given by Squire's transformation have an instability at a lower Reynolds number. In addition, the growth rate of the three-dimensional problem is also smaller than the corresponding transformed two-dimensional problem, so considering only two-dimensional perturbations will be sufficient. Note that at a given Reynolds number a three-dimensional perturbation is *not* necessarily more stable than a two-dimensional one.

Although Squire's theorem holds for a large class of flows, it could be invalid under the *nonisothermal* condition. GH¹³ showed previously for thermally stratified two-fluid flow that Squire's theorem does not hold even though Squire's transformation exists. Kallidasis *et al.*²⁰ studied thermocapillary instability on falling film flow and demonstrated the nonexistence of Squire's transformation. Therefore, the two issues mentioned in the beginning of the section will become more apparent. In this section, we will examine the three-dimensional stability problem and the validity of Squire's theorem. We again focus on the thin-film limit, so it can allow us to examine the problem analytically.

Let w' and W' denote the perturbation velocities in the cross-stream (z) direction for the thin and thick layers, respectively. Following the scaling procedures outlined in Sec. III, it can be shown that these velocities have the same scale as the corresponding x components. Thus we let $w' = \varepsilon \delta w$ and $W' = \delta W$. The scales of the other perturbation quantities remain the same as those in Sec. III. We now write the leading order linear-stability equations and boundary conditions. They are then formulated in terms of the normal-mode form \hat{q} by setting $q = \hat{q} \exp(ikx + inz + s\tau)$ for a given perturbation quantity q , where n is the wavenumber of perturbations in the z direction. Also recall that the long time scale $\tau = \varepsilon^2 t$ is used.

At leading order in ε , the film equations are

$$ik\hat{u} + in\hat{w} + \hat{v}_Y = 0, \quad (36a)$$

$$m\hat{u}_{YY} = ik\hat{p}, \quad (36b)$$

$$m\hat{w}_{YY} = in\hat{p}. \quad (36c)$$

Here \hat{p} is again independent of y because of $p_Y = 0$ from the y -momentum at leading order. As for the thick layer, using $D \equiv d/dy$, we have

$$ik\hat{U} + in\hat{W} + D\hat{V} = 0, \quad (37a)$$

$$\text{Re}_1(ik\bar{U}\hat{U} + \hat{V}D\bar{U}) = ik\hat{P} + [D^2 - (k^2 + n^2)]\hat{U}, \quad (37b)$$

$$\text{Re}_1 D\bar{U}\hat{V} = D\hat{P} + [D^2 - (k^2 + n^2)]\hat{V}, \quad (37c)$$

$$\text{Re}_1 ik\bar{W}\hat{W} = in\hat{P} + [D^2 - (k^2 + n^2)]\hat{W}. \quad (37d)$$

The boundary conditions are as follows.

At the walls:

$$\hat{u} = \hat{v} = \hat{w} = 0, \quad \text{at } Y = 1, \quad (38a)$$

$$\hat{U} = \hat{V} = \hat{W} = 0 \quad \text{at } y = -1. \quad (38b)$$

At the unperturbed interface ($y = Y = 0$):

$$m D\hat{u} = (D\hat{U} + ik\hat{V}) + ik\lambda^{-1}M\hat{\xi}, \quad (39a)$$

$$m D\hat{w} = (D\hat{W} + in\hat{V}) + in\lambda^{-1}M\hat{\xi}, \quad (39b)$$

$$\hat{p} = -(k^2 + n^2)\hat{\xi}/\text{Ca}_0, \quad (40)$$

$$\hat{U} = (m^{-1} - 1)\hat{\xi}, \quad \hat{V} = \hat{W} = 0, \quad (41)$$

$$\hat{v} = s\hat{\xi}. \quad (42)$$

Again, there is no need to solve the perturbation temperature field since we derive (39a) and (39b) using (28) and the base-state temperature, similar to (20).

Equations (36)–(42) constitute an eigenvalue problem that determines the three-dimensional linear stability in the thin-film limit. Note that the dynamics of the thick layer couple to the film's through (39a) and (39b). Since s appears only in (42), it can be uniquely determined. It can be shown analytically that s of the three-dimensional problem takes the form

$$s = \frac{M}{2m\lambda}(k^2 + n^2) - \frac{1}{3m \text{Ca}_0}(k^2 + n^2)^2 + \frac{1}{m} \left(1 - \frac{1}{m}\right) \Gamma(\text{Re}_1, k, n), \quad (43)$$

with

$$\Gamma = k(k^2 + n^2)^{-1/2} G[\text{Re}_1 k(k^2 + n^2)^{-1/2}, (k^2 + n^2)^{1/2}]. \quad (44)$$

Define $\hat{\nabla}^2 = \partial^2/\partial x^2 + \partial^2/\partial z^2$. Equation (43) is equivalent to the following two-dimensional evolution equation:

$$\begin{aligned} \xi_\tau + \frac{M}{2m\lambda} \hat{\nabla}^2 \xi + \frac{1}{3m \text{Ca}_0} \hat{\nabla}^2 \hat{\nabla}^2 \xi + \frac{1}{(2\pi)^2 m} \left(\frac{1}{m} - 1\right) \\ \times \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \Gamma(\text{Re}_1, k, n) \\ \times \left(\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \xi(x', z', \tau) e^{ik(x-x') + in(z-z')} dx' dz' \right) dk dn \\ = 0. \end{aligned} \quad (45)$$

Compared to (30) of the two-dimensional problem, we find that the three-dimensional problem has a stronger thermocapillary effect [because of $(k^2 + n^2) > k^2$ in the M term] while the viscosity stratification effect is weaker [because for

a fixed k , $\Gamma(n=0)=G$ and the real part of Γ is a positive function decreasing toward 0 as n increases]. Obviously, the capillary stabilization is also stronger in the three-dimensional problem. Therefore, three-dimensional perturbations will evolve (either grow or decay) more rapidly than two-dimensional ones; two-dimensional perturbations are *not* necessarily more dangerous. Similar to (32), the critical Reynolds number can be obtained by using the long-wave expansion in small k and n :

$$\text{Re}_1^* = \frac{30m}{\lambda} \left| \frac{M}{(m-1)} \right| \left[1 + \left(\frac{n}{k} \right)^2 \right]. \quad (46)$$

As a result, Re_1^* for the onset of stability/instability in the three-dimensional problem will be larger than that of the two-dimensional problem. For $M > 0$, thermocapillarity is destabilizing, but can be stabilized by viscosity stratification with $m < 1$. Thus (46) yields Re_1^* for the onset of stability and suggests that if the two-dimensional problem is unstable at some Re_1 , the three-dimensional system will also be unstable at the same Re_1 . For $M < 0$, on the other hand, thermocapillarity is stabilizing while viscosity stratification is destabilizing for $m > 1$. Re_1 given by (46) is the maximum Reynolds number allowed for the stability. Therefore, an instability in the two-dimensional problem at a given Re_1 *does not* necessarily imply an instability in the three-dimensional problem at the same Re_1 .

The results shown above suggest that the usual interpretation concerning the criticality using Squire's theorem is not applicable to our system. In fact, we find that Squire's transformation generally does not exist. But there are two exceptions: (i) $m=1$, and (ii) $m \neq 1$, $M=0$ and $\text{Ca}_0 \rightarrow \infty$. In case (i), the thick-layer dynamics are irrelevant to the leading order stability, so is Re_1 ; the stability is determined solely by the film flow. In this case, Squire's transformation can be simply carried out by $\tilde{k}=(k^2+n^2)^{1/2}$ and $\tilde{k}\tilde{u}=k\hat{u}+n\hat{w}$ while other quantities remain untransformed. Here, overtilde quantities/parameters are used to describe the transformed two-dimensional system. It is clear from (43) that for $m=1$ and the absence of interfacial tensions, three-dimensional perturbations can grow more rapidly than two-dimensional ones due solely to thermocapillary instability. Therefore, considering only two-dimensional perturbations will be insufficient; Squire's theorem does not hold even though there is a Squire transformation. Physically, the instability in this case is independent of basic flows, just like the case of a stationary base state. Thus interfacial perturbations will develop unbiasedly in both x and z directions. Also, since the instability in each direction can occur independently for arbitrary k or n ($\ll \varepsilon^{-1}$), their superposition will exacerbate the instability.

As for case (ii), the instability arises solely from viscosity stratification, and can be reduced to an equivalent two-dimensional problem under the following transformation:

$$\begin{aligned} \tilde{k}(\tilde{u}, \tilde{U}) &= k(\hat{u}, \hat{U}) + n(\hat{w}, \hat{W}), \\ (\tilde{v}, \tilde{\rho}, \tilde{V}, \tilde{P}, \tilde{\xi}) &= (\hat{v}, \hat{\rho}, \hat{V}, \hat{P}, \hat{\xi}), \end{aligned} \quad (47)$$

$$\tilde{k} = (k^2 + n^2)^{1/2}, \quad \tilde{s} = \tilde{k}s/k, \quad \tilde{\text{Re}}_1 = \text{Re}_1 k/\tilde{k}, \quad \tilde{m} = m,$$

$$\tilde{\lambda} = \lambda.$$

Since we have solved the two-dimensional problem analytically in Sec. IV, we can utilize its solution and the above transformation to solve the three-dimensional problem analytically. This is how we derive Γ in (43). From (30) for the two-dimensional problem, we know that instability occurs when $m > 1$ at any nonzero Reynolds number, so the critical Reynolds number for stability will be zero. Also, since $\text{Re}_1 = \tilde{\text{Re}}_1 \tilde{k}/k$, the critical Reynolds number in the three-dimensional problem is zero, too. Moreover, because $\tilde{k} \geq k$, $\text{real}(\tilde{s}) \geq \text{real}(s)$. In what follows, two-dimensional perturbations are the most dangerous. Thereby, Squire's theorem does hold in this case.

As such, Squire's theorem does not exist in the presence of thermocapillarity, as also evidenced by comparing the growth rates between (30) for the two-dimensional problem and (43) for the three-dimensional problem. This conclusion is consistent with those found in previous studies on thermocapillary instability.^{20,21} A similar situation can also be found due to adverse density stratification.¹³ In analogy to the effect of density stratification discussed by Joseph and Renardy,¹⁴ for the fully three-dimensional stability problem, there is no basic flow in the cross-stream direction, so thermocapillary instability could occur at any Reynolds number in the three-dimensional problem, while its two-dimensional counterpart still admits stability in a range of the Reynolds number. This is the reason why the usual interpretation of Squire's theorem does not hold when thermocapillary effects are present.

VI. EXTENSION TO WEAKLY NONLINEAR STABILITY REGIME

In the foregoing sections, we have analyzed the leading order linear stability that is only appropriate for sufficiently small perturbations. As the system undergoes instability, the interface grows with time and its amplitude could be so large that nonlinear effects become important. An issue here is often concerning if the instability can persist or be arrested, so that one can foresee if the film is ruptured or still keep its integrity. Here we restrict our attention on the weakly nonlinear regime in which the interface amplitude is assumed finite but still remains small compared to the undisturbed film thickness.

In view of the nonexistence of Squire's theorem shown in Sec. V, the three-dimensional problem could be more unstable than the two-dimensional problem. Thus, an appropriate nonlinear evolution equation of the interface should be on the two-dimensional basis like (45). Nevertheless, it might also be essential to understand differences of the interface

dynamics between these two problems. To gain some insights, first of all, we consider the weakly nonlinear extension to (29) for the two-dimensional problem. The corresponding extension to (45) for the three-dimensional problem will be discussed later.

A. Weakly nonlinear evolution equation for the two-dimensional stability problem

To extend (29) into the weakly nonlinear regime, we shall find the size of perturbations within which the equation holds. Our approach to finding appropriate scales in this regime is similar to that of core-annular flow studied by Papageorgiou *et al.*²² It is briefly provided below. For $Ca \sim \varepsilon$ and $M \sim O(1)$, using (17) and the long time scale $\tau = \varepsilon^2 t$, we expand the kinematic condition (11): $\varepsilon^2 \delta v = \varepsilon^2 \delta \xi_\tau + \delta^2 \bar{U}_{2y}|_{y=0} \xi \xi_x$. Balancing the terms, we find $\delta \sim \varepsilon^2$ appropriate for the problem. We can also show that the nonlinear effects contributing to the leading order come only from $\xi \xi_x$ shown above; all the other nonlinear terms will be of higher orders. For instance, the nonlinear terms arising from the evaluation of $\varepsilon^2 \delta v$ at the deflected interface $Y=1 + \delta \xi$ are $O(\varepsilon^2 \delta^2)$, at most. That from the inertia of the thick layer contributes the $O(\delta^2)$ correction to δU , so the associated term in $\varepsilon^2 \delta v$ is $O(\varepsilon^2 \delta^2)$. The nonlinearity from the interface curvature is $O(\delta^3)$ and yields $O(\varepsilon^{-1} \delta^3)$ to the capillary pressure. The resulting effect contributing to $\varepsilon^2 \delta v$ is merely $O(\varepsilon^2 \delta^3)$ and even of higher order than those considered above.

As a result, we derive the following weakly nonlinear evolution equation:

$$\begin{aligned} \xi_\tau + \frac{1}{m} \xi \xi_x + \frac{M}{2m\lambda} \xi_{xx} + \frac{1}{3mCa_0} \xi_{xxx} + \frac{1}{2\pi m} \\ \times \left(\frac{1}{m} - 1 \right) \int_{-\infty}^{\infty} G(k) \int_{-\infty}^{\infty} \xi(x', \tau) e^{ik(x-x')} dx' dk = 0. \end{aligned} \quad (48)$$

Letting $\zeta = Lx$, $\nu = 2\lambda(3|M|Ca_0)^{-1}L^{-2}$, $\phi = 2L\lambda|M|^{-1}\xi$, $\omega = 2\lambda(1-m^{-1})L^2/|M|$, and $T = |M|(2m\lambda)^{-1}L^{-2}\tau$, we reduce (48) to the following form:

$$\begin{aligned} \phi_T + \phi \phi_\zeta + \text{sign}(M) \phi_{\zeta\zeta} + \nu \phi_{\zeta\zeta\zeta} \\ - \frac{\omega}{2\pi} \int_{-\infty}^{\infty} G(k) \int_{-\infty}^{\infty} \phi(\zeta', T) e^{ik(\zeta-\zeta')} d\zeta' dk = 0. \end{aligned} \quad (49)$$

This equation is a Kuramoto-Sivashinsky (KS) type and generally admits nonlinear saturation of instability. Similar equations can also be found in core-annular film flows.^{22,23}

For disturbances with very short wavelengths, it is not possible for effects of the nonlocal term (via G) to override the capillary stabilization (via $\phi_{\zeta\zeta\zeta}$) since the real part of the Airy kernel G approaches a constant as $k \rightarrow \infty$ [see (34)] while the capillarity goes like k^4 . Therefore, short-wave disturbances will be rapidly damped by the capillarity; effects of the nonlocal term manifest only for long and moderate wavelengths. Furthermore, the quantitative behavior of G for moderate k can be adequately described by the small- k approximation (see Fig. 2, the agreement between the exact and

the long-wave results is good up to $k \sim 3$). Similar adequacy of the small- k approximation for the effect of the nonlocal term can also be found in core-annular film flows.²⁴

There is a possibility that one has to consider the full nonlocal term. This occurs only when the interfacial tension is too weak, so one needs even shorter waves to make an instability arrested within the weakly nonlinear regime. However, since the capillary stabilization is too weak, the amplitude of the interface could have been growing so large that it could be already beyond the weakly nonlinear regime. In this case, the derived KS equation is no longer appropriate to describe the nonlinear behavior for the interface.

In light of the above discussion, from both practical and theoretical points of view, it is sufficient to account only long and moderate waves in G . We therefore approximate G as

$$G = 2ik + \frac{\text{Re}_1}{60} k^2 + iG_3 k^3 + G_4 k^4 + O(k^5). \quad (50)$$

Note that we retain the terms up to $O(k^4)$ whose effect could mitigate the capillary stabilization $\nu \phi_{\zeta\zeta\zeta}$. Here, both G_3 and G_4 are real and given by Appendix . Using (50), we can adequately describe the interface dynamics without appealing to the detailed evaluation of the complex Airy kernel. Substituting (50) into (49) and using Galileo transformation $\zeta \rightarrow \zeta + 2\omega T$, we simplify Eq. (49) to

$$\phi_T + \phi \phi_\zeta + \alpha \phi_{\zeta\zeta} + \beta \phi_{\zeta\zeta\zeta} + \gamma \phi_{\zeta\zeta\zeta\zeta} = 0, \quad (51)$$

where $\alpha = M/|M| - \omega \text{Re}_1/60$, $\beta = -\omega G_3$, and $\gamma = \nu - \omega G_4$. We are more interested in the $\alpha > 0$ case since it introduces destabilizing effects. Here γ is generally positive, so linearly unstable waves can be stabilized through nonlinear modulation. Letting $T \rightarrow T/\alpha$ and $\phi \rightarrow \alpha \phi$, we reduce (51) in the following canonical form:

$$\phi_T + \phi \phi_\zeta + \phi_{\zeta\zeta} + (\beta/\alpha) \phi_{\zeta\zeta\zeta} + (\gamma/\alpha) \phi_{\zeta\zeta\zeta\zeta} = 0. \quad (52)$$

Equation (52) with $\beta=0$ is the prototype KS equation and can be found in a variety of contexts.^{23,25-29} The existence and boundedness of solutions of the KS equation have also been proved.³⁰⁻³⁴ In addition, the solutions reveal rich spatiotemporal dynamics such as traveling waves, time periodic motion, and chaos, depending on the parameters.³⁵⁻³⁹ The term $\phi_{\zeta\zeta\zeta}$ introduces dispersion to interface waves and hence can change their shapes or velocities.^{39,40} In fact, most of the film dynamics can be characterized by a more generalized form: the Shkadov equation,^{42,43} from which the KS equation can be derived in an appropriate limit.⁴⁴

Since the dynamics of (52) have been well studied in the past, we do not intend to solve the equation here. Rather, we discuss its qualitative behaviors related to our problem. For a periodic domain of 2π , linear instability and the subsequent nonlinear modulation occur if $\gamma/\alpha < 1$; otherwise a trivial solution $\phi=0$ will be obtained since all waves are damped by the interfacial tension. We can estimate the relevant scales below by balancing the terms in (52). A balance of the second and fourth derivative terms provides an estimate of the

length scale $(\gamma/\alpha)^{1/2}$ necessary for instability. For an unstable wave with that length scale, its amplitude can be estimated as $(\alpha/\gamma)^{1/2}$ by balancing the nonlinear term. Similarly, we can obtain a time scale as γ/α from balancing the unsteady term. With the above scales, the scale of the third derivative term relative to the remaining terms is $(|\beta|/\alpha) \times (\alpha/\gamma)^{1/2}$. From a dynamic point of view, large α/γ generally leads the spatiotemporal behavior to be less regular³⁹ while large $|\beta|/\alpha$ tends to regularize the waves.⁴¹ Therefore, $(|\beta|/\alpha)(\alpha/\gamma)^{1/2}$ can be served as the dispersion-dissipation ratio for indicating the dynamic tendency toward organized wave structures.

Using the scales shown above, we now take an example to illustrate the effects at work. Here, we choose $\gamma/\alpha=0.01$ to find the conditions for realizing nonlinear saturation of instability when destabilizing effects are much stronger than stabilizing effects in the linear regime. Suppose that the interface starts with a monochromatic wave of a small amplitude, say, $0.1 \sin(\zeta)$. An instability will set off for wavelengths longer than $(\gamma/\alpha)^{1/2} \sim 0.1$ (or the Fourier mode <10) according to the linear theory. The most unstable wave occurs at the Fourier mode $\sim(\alpha/2\gamma)^{1/2} \sim 7$, and grows rapidly in the early stages of the evolution. As the interface amplitude becomes large, the wave steepening due to the KS nonlinearity will make the wavelength shorter, exciting higher Fourier modes. As the amplitude grows up to an order of $(\alpha/\gamma)^{1/2} \sim 10$, the instability will be gradually attenuated due to the short-wave stabilization and finally arrested within the finite-amplitude regime. The dispersion-dissipation ratio is $(|\beta|/\alpha)(\alpha/\gamma)^{1/2} \sim 10|\beta|/\alpha$, so organized wave structures (e.g., traveling/pulse-like waves) are likely observed in this case unless $|\beta|$ is sufficiently small (e.g., nearly matched viscosities or a sufficiently slow flow).

Notice that for finite-amplitude interfacial waves, the true physical scale of the amplitude with respect to the plate separation should be read as $\alpha(\alpha/\gamma)^{1/2}M\lambda^{-1}\varepsilon^2$. For $\alpha \sim 1$, $M \sim 1$, and $\lambda \sim 1$, the amplitude scale is $10\varepsilon^2$. Therefore, one requires $\varepsilon \ll 0.1$ to ensure the validity of the weakly nonlinear analysis. That is, in order to keep the film from rupture, the film thickness must be much smaller than 10% of the plate separation. It is also clear that a stronger tension (larger γ) or a faster flow (smaller α) will render stabilization, making the interface evolution confined within a smaller amplitude and hence allowing a thicker film for preventing rupture.

B. Weakly nonlinear evolution equation for the three-dimensional stability problem

The preceding discussion is based on the *one-dimensional* interface evolution equation governing the stability of the two-dimensional problem. For the three-dimensional problem that could be more unstable than the two-dimensional one, the corresponding evolution equation of the interface should take a two-dimensional form. In a similar fashion to derive (48), we derive the two-dimensional evolution equation below for the three-dimensional problem:

$$\begin{aligned} & \xi_\tau + \xi\xi_x + \frac{M}{2m\lambda} \hat{\nabla}^2 \xi + \frac{1}{3m \text{Ca}_0} \hat{\nabla}^2 \hat{\nabla}^2 \xi + \frac{1}{(2\pi)^2 m} \left(\frac{1}{m} - 1 \right) \\ & \times \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \Gamma(\text{Re}_1, k, n) \\ & \times \left[\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \xi(x', z', \tau) e^{ik(x-x') + in(z-z')} dx' dz' \right] dk dn \\ & = 0. \end{aligned} \quad (53)$$

To the best of our knowledge, it is the first time that a two-dimensional evolution equation with the inclusion of a *nonlocal* term can be systematically derived. Making use of (44) and (50), we again approximate the kernel function Γ to make the problem more tractable:

$$\Gamma = 2ik + \frac{\text{Re}_1}{60} k^2 + iG_3 k \tilde{k}^2 + G_4 k \tilde{k}^3 + O(k \tilde{k}^4). \quad (54)$$

Here, Re_1 in G_3 and G_4 given by the Appendix is now replaced by $\text{Re}_1 k/\tilde{k}$. Similar to (52), we can show, with an appropriate transformation, that (53) can be reduced to the following two-dimensional KS equation (2-D KS):

$$\xi_\tau + \xi\xi_x + (\hat{\nabla}^2 \xi + \hat{\alpha}\xi_{xx}) + \hat{\beta} \hat{\nabla}^2 \xi_x + (\hat{\gamma}_1 \hat{\nabla}^4 \xi + \hat{\gamma}_2 \hat{\nabla}^2 \xi_{xz}) = 0. \quad (55)$$

Note that spatial derivatives are *not* symmetric between x and z because the basic flow acts in the x direction. The original equation (with $\hat{\gamma}_2=0$) has been postulated for modeling various nonlinear systems.⁴⁵ But here we find an additional dissipative term from $\hat{\gamma}_2 \neq 0$. Equation (55) is also similar to those derived in related problems.^{46,47} Although 2-D waves (with $s \sim (k+n)^2$) are more unstable than 1-D waves ($\sim k^2$) for long-wavelength disturbances, the nonlinear wave steepening makes the short-wave stabilization more effective in the former [$\sim (k+n)^4$] than the latter ($\sim k^4$). Therefore, we speculate that compared to 1-D waves, 2-D waves might be easier to be arrested in the weakly nonlinear regime, although they can grow faster than 1-D waves in the linear regime.

The main difference between the 1-D and 2-D KS equations lies in effects due to dispersion (from odd derivative terms).⁴⁸ When dispersion is weak, the dynamics of the 2-D case are qualitatively similar to those of the 1-D case.⁴⁵ If the effect is strong, the extra 2-D dispersion term can be important even at large tension (large $\hat{\gamma}_1$), leading to a change of wave patterns or complicated wave interactions in the 2-D interface's dynamics. The detailed dynamic behaviors of the 2-D KS equation can be found elsewhere,⁴⁸⁻⁵¹ and hence we do not pursue them here.

VII. CONCLUDING REMARKS

We have asymptotically examined the stability of two-layer Couette flow in the thin-layer limit. Effects of viscosity stratification, thermocapillarity, and interfacial tension are included in the analysis. Most of our efforts are devoted to the two-dimensional stability problem. We devise a scaling analysis to identify dominant mechanisms of instability in

various ranges of parameters. Using an appropriate scaling, we analytically derive an evolution equation of the interface governing the leading order linear stability of the system. The effects of viscosity stratification can be represented by a nonlocal contribution in terms of Airy functions.

Viscosity stratification linearly destabilizes (stabilizes) when the film is more (less) viscous. Thermocapillarity linearly stabilizes (destabilizes) as the thin layer is cooled (heated). The viscosity stratification effects could be stronger than the thermocapillary effects for long waves, but the former can be outweighed by the latter for short waves. The interplay between these two effects determines the Reynolds number at the criticality between stability and instability.

Interfacial tension is always stabilizing, and is often robust to make the system completely stable. For a less viscous film, however, we find a possibility of the existence of two neutral states in the wavenumber space when the flow is within a certain window in the weak-tension regime.

We also examine the three-dimensional stability problem. The corresponding stability is reduced to a two-dimensional film evolution equation consisting of a nonlocal term. It can be shown analytically that a three-dimensional perturbation can grow more rapidly than a two-dimensional one in the presence of thermocapillarity. More importantly, we prove the nonexistence of Squire's theorem. We also find that the critical Reynolds number in the three-dimensional problem is larger than that in the two-dimensional problem. However, an instability in the two-dimensional problem does not necessarily suggest an instability in the three-dimensional problem. For each problem, the corresponding extension to the weakly nonlinear regime is also discussed. With an appropriate scaling, the weakly nonlinear interfacial dynamics can be characterized by the Kuramoto-Sivashinsky equations. Consequently, the instability can be arrested in the finite-amplitude regime; that is, the film can still keep intact without rupture.

APPENDIX: LONG-WAVE EXPANSION OF THE AIRY KERNEL G

Here we provide the long-wave approximation up to $O(k^4)$ for the Airy kernel G in (29). We first expand the numerator and denominator of G :

$$G = \frac{ik a_1 + ib_1 k + c_1 k^2 + id_1 k^3 + O(k^4)}{2 a_2 + ib_2 k + c_2 k^2 + id_2 k^3 + O(k^4)}, \quad (\text{A1})$$

with the coefficients given below:

$$a_1 = \frac{\text{Re}_1^{1/3}}{6\pi}, \quad b_1 = -\frac{\text{Re}_1^{4/3}}{144\pi}, \quad c_1 = \frac{\text{Re}_1^{1/3}}{30\pi} - \frac{\text{Re}_1^{7/3}}{9072\pi},$$

$$d_1 = -\frac{\text{Re}_1^{4/3}}{720\pi} + \frac{\text{Re}_1^{10/3}}{1\,088\,640\pi},$$

$$a_2 = \frac{\text{Re}_1^{1/3}}{24\pi}, \quad b_2 = -\frac{\text{Re}_1^{4/3}}{720\pi},$$

$$c_2 = \frac{\text{Re}_1^{1/3}}{180\pi} - \frac{67\text{Re}_1^{7/3}}{3\,628\,800\pi}, \quad d_2 = -\frac{\text{Re}_1^{4/3}}{5040\pi} + \frac{\text{Re}_1^{10/3}}{748\,440\pi}.$$

An expansion of (A1) in a series of k yields

$$G = 2ik + \frac{\text{Re}_1}{60}k^2 + iG_3k^3 + G_4k^4 + O(k^5). \quad (\text{A2})$$

Here G_3 and G_4 can be expressed in terms of the coefficients given above:

$$G_3 = [c_1 - a_1c_2/a_2 - (a_1b_2 - b_1a_2)b_2/a_2^2]/(2a_2),$$

$$G_4 = -[d_1 - a_1d_2/a_2 - c_2(a_1b_2 - b_1a_2)/a_2^2 + (-c_1a_2^2 + a_1c_2a_2 - b_2b_1a_2 + a_1b_2^2)b_2/a_2]/(2a_2).$$

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