

## Thermosolutal Marangoni instability of film falling down a flexible substrate

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A linear stability analysis is conducted to decipher how the thermal and surfactant Marangoni numbers affect primary instabilities when a two-dimensional incompressible viscous film flows down a uniformly heated flexible substrate under gravity. Since the viscous film is nonisothermal and a soluble surfactant is dissolved in it, we require additional heat- and mass-transfer equations along with the Navier-Stokes equations for describing this film flow. To explore linear stability, we formulate the Orr-Sommerfeld-type boundary value problem (OS BVP). The analytical long-wave series expansion and the numerical Chebyshev spectral collocation method are applied to solve the OS BVP. The long-wave results disclose that surface mode instability intensifies in the presence of the thermal Marangoni number but weakens as the surfactant Marangoni number rises, creating a surfactant-free isothermal film flowing over a flexible substrate when these two opposing impacts balance each other. Moreover, we identify a range of the inclination angle for which the surface mode becomes linearly stable, primarily because the stabilizing effects of the surfactant Marangoni stress and depthwise gravitational force dominate over the destabilizing effects of the thermal Marangoni stress and spring stiffness coefficient  $C_K$ . In the arbitrary wave-number regime, the numerical results reveal that surface mode, S-mode, and P-mode instabilities are amplified with rising values of the thermal Marangoni number but are weakened with rising values of the surfactant Marangoni number. Moreover, we observe a modal coalescence between the surface mode and the S-mode in the long-wave regime as the thermal Marangoni number increases. The base surface surfactant concentration parameter,  $\Gamma_E$ , exhibits both stabilizing and destabilizing influences on all the surface mode, S-mode, and P-mode instabilities. However, the surfactant solubility parameter,  $\xi_s$ , shows only a stabilizing influence on all instability modes. If the magnitudes of the spring stiffness coefficient  $C_K$ , the longitudinal tension parameter  $C_T$ , and the flexural rigidity parameter  $C_B$  are significantly lowered, unstable domains of the P-mode and the wall mode, which emerged separately in the finite wave-number regimes, coalesce at higher values of  $\Gamma_E$ . Additionally, we observe a competition between the surface mode and the wall mode. When the damping coefficient,  $C_D$ , is equal to 0.32, the wall mode controls the primary instability in the finite wavelength regime. As soon as  $C_D$  increases to 0.35, the surface mode governs the primary instability in the long-wave regime at low values of the surfactant Marangoni number. On the other hand, we see the existence of the unstable shear mode in the high Reynolds number regime when the inclination angle is small enough. The initiation of shear mode instability can be quickened by raising the thermal Marangoni number, while its initiation will be delayed if the surfactant Marangoni

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number goes up. Finally, we demonstrate that the primary instabilities induced by the surface mode and the wall mode may also appear in different ranges of  $C_D$  due to the destabilizing impact of the thermal Marangoni number even when the Reynolds number approaches zero.

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## I. INTRODUCTION

Over the last few decades, several studies have been conducted to probe how various compliant coatings reduce drag in fluid flows, whether through a flexible channel or over a flexible plate, following Kramer's seminal experimental work [1]. Moreover, there is evidence that the transition from laminar flow to turbulence in boundary-layer flow can be made slower by using a specific type of flexible coating on the flat plate, which causes the flow to be in a laminar state for a longer period of time. This physical phenomenon results in alterations to the mass and momentum fluxes, drag characteristics, acoustic properties, and heat- and mass-transfer rates near the flexible surface [2]. Apart from that, the compliant-wall flow model has immense applications in the biomedical field. For instance, exploring fluid flow interactions with the solid structures is crucial in figuring out how a pulse propagates and how the transmural pressure varies in arteries in the cardiovascular system [3,4]. Moreover, investigating surface instability for the soluble-surfactant-laden fluid flow over a compliant wall can be an appropriate flow model for understanding how pulmonary surfactant, found in the fluid lining the alveolar surface of the lungs, plays an important role in preventing alveolar collapse during the breathing cycle by reducing surface tension, as it is a highly surface-active material and the lung surface is elastic [5,6].

In the context of boundary-layer flows over a flexible plate, Benjamin [7] and Landhal [8] began theoretical studies on the linear stability analysis, and their results predicted that the Tollmien-Schlichting instability is destabilized by the internal damping parameter. To decipher the instability and transition of an incompressible Blasius boundary-layer flow over a flexible plate, Gyorgyfalvy [9] preferred a simple flexible skin model, which consists of a spring-backed membrane with internal damping. He demonstrated that the primary cause of the significant delay in the evolution of the Tollmien-Schlichting instability is a reduction in the amplification rate. Subsequently, a detailed linear stability analysis was performed by Carpenter and Garrad [10,11] for flows over Kramer-type compliant surfaces based on the Orr-Sommerfeld-type boundary value problem. They unravelled two distinct types of finite wavelength instabilities: the flow-induced surface instability and the Tollmien-Schlichting instability. Their numerical findings exhibited that the plate damping parameter stabilizes the flow-induced surface instability but destabilizes the Tollmien-Schlichting instability. Moreover, they showed that there occurs a modal coalescence between the flow-induced surface mode and the Tollmien-Schlichting mode, which was further ensured by Sen and Arora [12]. On the other hand, Carpenter and Gajjar [13] conducted a study on boundary-layer flow over a flexible plate to probe how isotropic and anisotropic compliant surfaces affect traveling wave flutter instability. Consequently, they introduced an asymptotic theory to examine disturbances in both two and three dimensions. To inspect the short-time disturbance energy growth phenomenon on boundary-layer flow and how it is influenced by the wall flexibility, Malik *et al.* [14] carried out a stability analysis using the normal velocity and normal vorticity formulations. They concluded that as the Reynolds number increases, the maximum transient amplification rate accelerates slowly.

On the other hand, Hains and Price [15] first instigated a study on the linear stability of Poiseuille flow bounded by flexible walls, assuming the wall to be a stretched membrane without mass and neglecting the effect of the elastic foundation. Thereafter, Tsvlodub [16] also conducted a linear stability analysis of a fluid flow through an elastic channel with flexible walls. To delve into the finite-amplitude nonlinear wave, Rotenberry [17] considered a flow between compliant walls. According to his discussion, the presence of a flexible wall makes the traveling wave's amplitude larger. Based on the results, he concluded that there is no significant qualitative difference in the

transition process from laminar flow to turbulence with or without a flexible wall. Ehrenstein and Rossi [18] later also performed a nonlinear stability analysis for a fluid flow in a channel with a single compliant wall. Using the method of multiple scales, they derived the complex Ginzburg-Landau equation. To investigate the Tollmien-Schlichting wave and its nonlinear evolution, Gajjar and Sibanda [19] further revisited their compliant-wall flow model. In the study, they mainly focused on finding out the impacts of various compliant-wall parameters. Davies and Carpenter [20,21], on the contrary, confined themselves to investigating the flow-induced surface instability. In the case of the compliant-wall flow model, the physical mechanism of instability was elucidated by Huang [22]. However, Huang restricted his study to the two-dimensional inviscid shear flow. Recently, Hoepffner *et al.* [23] accomplished a nonmodal stability analysis for the channel flow bounded by flexible walls. They noticed that the fluid-structure system becomes unstable owing to the flow-induced surface instability as the wall stiffness decreases.

In parallel, numerous studies have already been conducted on the stability of isothermal falling films on a rigid impermeable plane, following Kapitza's seminal experimental work [24]. In deciphering the process of how the transition of laminar flow to turbulence occurs, the investigation of such flow problems is very relevant [25–27]. Furthermore, engineering applications and natural processes frequently encounter these flow problems. In this context, Benjamin [28] and Yih [29] pioneered the study of surface mode instability in the low Reynolds number regime and predicted that the instability initiates when the Reynolds number exceeds a certain threshold that depends on the angle of inclination. Although there is long-wave surface mode instability in film flows, the finite wavelength shear mode instability can also be perceived in the high Reynolds number regime when the inclination angle is small enough [30–32]. In fact, they compete with each other to trigger the primary instability. Since thin film flows over a flexible substrate have extensive applications in the biomedical field, Matar and Kumar [33] originated a study in this direction to comprehend the fluid-structure interactions and their role in the surface mode instability. In particular, they explored the rupture phenomenon for a falling surfactant-covered liquid film over a flexible plate based on the evolution equations for the deflections of the air-liquid and wall-liquid interfaces. They reported that increasing the damping coefficient, the longitudinal tension, and the surfactant Marangoni stress stabilizes the primary instability. Their further investigation [34] reveals that the maximum temporal growth rate for surface mode instability increases as the plate damping coefficient and the plate longitudinal tension decrease. To elucidate nonlinear surface mode instability, Matar *et al.* [35] also derived three strongly coupled nonlinear evolution equations using the integral theory in terms of the film thickness, substrate deflection, and volumetric flow rate, respectively. Their numerical results demonstrated that the existence of spatiotemporal chaos in the weakly nonlinear regime is the consequence of the decreasing values of the plate damping coefficient or the plate longitudinal tension. Later, Sisoiev *et al.* [36] examined how the flexible wall affects steady-state traveling waves and their bifurcations in the nonlinear regime. As a result, they developed depth-averaged evolution equations using the boundary-layer theory together with the von Kármán-Polhausen approximation. Using lubrication approximation, Howell *et al.* [37] showed that the steady-state solution of the gravity-driven thin film flowing over a flexible substrate, which is modeled as an elastic beam, is strongly dependent on the length and stiffness of the beam. Since heat transfer to the flexible plate introduces additional Marangoni stress, Chao and Ding [38] first executed a stability analysis for a thin film flow over a uniformly heated flexible plate. Specifically, they analyzed the impact of thermocapillary forces on nonlinear traveling wave solutions. Their numerical findings ensured that the speed and maximum amplitude of the steady-state traveling waves amplify as the thermal Marangoni stress rises. Recently, a detailed study on surface-mode-, wall-mode-, and shear-mode-induced instabilities in the linear regime was accomplished by Alexander *et al.* [39] for a liquid film falling down an inclined flexible plate under the framework of the Orr-Sommerfeld-type boundary value problem. According to their results, the flow can be susceptible to surface mode instability at a vanishing Reynolds number if the spring stiffness coefficient is less than its critical value. Moreover, the intensity of surface mode instability increases as the spring stiffness coefficient decreases. The above compliant-wall flow model was further revisited by Samanta [40,41] to look into how the

insoluble surfactant and three-dimensional disturbances influence surface mode and wall mode instabilities. His numerical results confirmed that the insoluble surfactant stabilizes surface mode instability but destabilizes wall mode instability. After further searching with a soluble surfactant, Samanta [42] reported that surface mode and wall mode instabilities can be made stronger with increasing values of the base surface surfactant concentration parameter. Recently, Jia and Jian [43], Chattopadhyay *et al.* [44], and Desai *et al.* [45] deciphered how a compliant substrate affects surface mode instability for the odd-viscous liquid. Using the Orr-Sommerfeld-type boundary value problem (OS BVP) and the nonlinear surface evolution equation, they demonstrated that odd viscosity stabilizes surface mode instability.

Upon reviewing the existing literature, we notice that no studies have been devoted to exploring the combined thermosolutal Marangoni effect on the primary instabilities of thin film flows over a flexible substrate. Moreover, the solo surfactant Marangoni number exhibits a stabilizing influence on the hydrodynamic surface mode and a destabilizing influence on the wall mode, generated by the deformation of the flexible plate. On the other hand, the solo thermal Marangoni number shows a destabilizing impact on the hydrodynamic surface mode, the wall mode, and the thermocapillary S- and P-modes that emerged on account of the thermocapillary effect [46]. Since the thermocapillary modes are absent in the isothermal surfactant-laden film flow [42], we do not have any information on how a soluble surfactant affects the thermocapillary modes. Second, is there a modal coalescence between the wall mode and the thermocapillary P-mode, and how is this physical phenomenon influenced by a soluble surfactant? Third, how are the surface-mode- and wall-mode-induced inertialess instabilities altered in the presence of a soluble surfactant? To provide answers to these questions, we perform thorough research on the linear stability analysis of a soluble-surfactant-laden film flow over a uniformly heated flexible substrate. On the other hand, conducting an experimental study on the dilute surfactant solution with a weight concentration of 150 ppm, Hara *et al.* [47] reported that there was a 65% drag reduction rate. Therefore, such solutions have a significant practical relevance in industry owing to their potential for energy saving. The current study is organized as follows: In Sec. II, we discuss the governing equations and boundary conditions employed to describe the current flow problem. In Sec. III, we execute a detailed linear stability analysis based on the analytical long-wave analysis, while the numerical simulation for disturbances of arbitrary wave numbers is supplied in Sec. IV. In Sec. V, we demonstrate the numerical results for the shear mode in the high Reynolds number regime. Finally, Sec. VI presents the conclusions of the findings decoded from the current study.

## II. GOVERNING EQUATIONS

We analyze a two-dimensional, nonisothermal, incompressible viscous film mixed with a soluble surfactant that is flowing down an inclined flexible substrate under the action of a gravitational force. The flexible substrate is inclined at an angle  $\theta$  with the horizontal and is uniformly heated to a temperature  $T_w$ , which is higher than the ambient temperature  $T_a$  (see Fig. 1).

Since the viscous film is contaminated with a soluble surfactant, we choose  $c_b(x, y, t)$  as the bulk surfactant concentration and  $\Gamma(x, t)$  as the surface surfactant concentration. Moreover, both concentrations are below the critical concentration level, and thereby surfactants do not form micelles. Here,  $x$  and  $y$  are the spatial coordinates aligned in the streamwise and cross-stream directions, respectively, and  $t$  is the time coordinate. In fact, we have selected the Cartesian coordinate system to describe the current nonisothermal surfactant-contaminated flow arrangement, where the origin is placed on the equilibrium position of the flexible substrate. We assume that the dynamic viscosity  $\mu$  and the density  $\rho$  of the incompressible viscous film remain unchanged with temperature  $T(x, y, t)$ . Specifically, we assume that the temperature variations are small so that the density and viscosity variations with temperature are neglected in the current study (see, e.g., Refs. [48–50]). However, the surface tension  $\sigma(x, t)$  varies with both surface temperature  $T^i(x, t)$  and surface surfactant concentration  $\Gamma(x, t)$  according to the following linear relation [51,52]:

$$\sigma(T^i, \Gamma) = \sigma_a - E_T(T^i - T_a) - E_\Gamma(\Gamma - \Gamma_a), \quad (1)$$

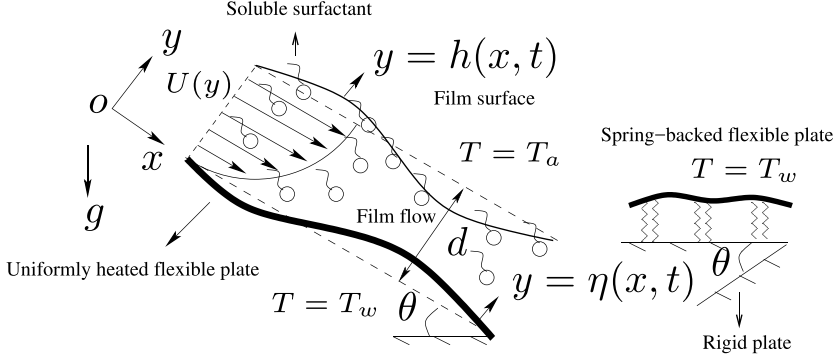


FIG. 1. A schematic diagram of a two-dimensional, gravity-driven, soluble-surfactant-impured, nonisothermal incompressible viscous film flowing over a uniformly heated flexible substrate that is assumed to be a spring-backed plate model. Here,  $U(y)$  represents the streamwise base velocity,  $T = T_w$  is a constant temperature at the flexible substrate, and  $T = T_a$  is the ambient temperature above the film surface.

where  $T = T^i$  is the temperature of the liquid film at the surface  $y = h(x, t)$ ,  $\sigma_a$  is the base surface tension, and  $\Gamma_a$  is the base surface surfactant concentration when  $T^i = T_a$ . Here, both  $E_T = -\frac{d\sigma}{dT^i}|_{T^i=T_a} > 0$  and  $E_\Gamma = -\frac{d\sigma}{d\Gamma}|_{\Gamma=\Gamma_a} > 0$ , as surface tension decreases with the increase in the values of surface temperature and surface surfactant concentration for most liquids. To justify our assumption of negligible variation in viscosity with small changes in temperature, we present the experimental findings reported by Khattab *et al.* [53] for the dilute solution of ethanol in water. Their experimental measurements show that for the aqueous solution of ethanol with viscosity  $\mu = 2.62 \times 10^{-3}$  Pa s and density  $\rho = 9.47 \times 10^2$  kg/m<sup>3</sup>, the rate of change of surface tension with temperature can be estimated approximately as  $E_T \approx 9 \times 10^{-5}$  N/mK. On the basis of the given experimental data, Ellaban *et al.* [54] calculated the temperature difference, which is of  $\mathcal{O}(10^{-3})$  K. To this temperature difference, the variation in viscosity is very small and can be omitted. Moreover, we neglect the effect of buoyancy compared to the Marangoni effects because we assume that the viscous film layer is very thin. A similar example can also be found in the studies of Podolny *et al.* [55,56]. However, there are some self-rewetting liquids, like dilute aqueous solutions of certain long-chain alcohols, that exhibit a nonmonotonic behavior of surface tension as temperature rises [57]. The gravity-driven, nonisothermal, surfactant-laden, incompressible viscous film flowing over a uniformly heated flexible substrate is governed by the following Navier-Stokes equations, the energy equation, and the bulk surfactant concentration equation:

$$\partial_x u + \partial_y v = 0, \quad (2)$$

$$\rho(\partial_t u + u\partial_x u + v\partial_y u) = -\partial_x p + \mu(\partial_{xx} u + \partial_{yy} u) + \rho g \sin \theta, \quad (3)$$

$$\rho(\partial_t v + u\partial_x v + v\partial_y v) = -\partial_y p + \mu(\partial_{xx} v + \partial_{yy} v) - \rho g \cos \theta, \quad (4)$$

$$\rho c_p(\partial_t T + u\partial_x T + v\partial_y T) = \kappa(\partial_{xx} T + \partial_{yy} T), \quad (5)$$

$$(\partial_t c_b + u\partial_x c_b + v\partial_y c_b) = D_b(\partial_{xx} c_b + \partial_{yy} c_b), \quad (6)$$

where  $u(x, y, t)$ ,  $v(x, y, t)$ ,  $p(x, y, t)$ ,  $T(x, y, t)$ , and  $g$  represent the  $x$  and  $y$  components of liquid velocity, liquid pressure, liquid temperature, and the gravitational acceleration, respectively. We are also assuming that the specific heat capacity  $c_p$  at constant pressure, the thermal conductivity  $\kappa$ , and the bulk surfactant diffusivity  $D_b$  are constant throughout the study. Since the fluid is incompressible and the temperature variations are small, the compressible effect and the heating due to viscous dissipation are neglected from the energy equation [48–50]. Next, we provide the boundary conditions

that the governing equations (2)–(6) implement to close the present flow configuration. At the film surface,  $y = h(x, t)$ , the gradients of surface tension caused by varying surface temperature and surface surfactant concentration induce both the thermal and surfactant Marangoni stresses. These additional stresses balance the hydrodynamic stress, yielding the following dynamic boundary conditions for the tangential stress and normal stress, respectively [51,52]:

$$[4\mu \partial_y v \partial_x h + \mu(\partial_y u + \partial_x v)\{1 - (\partial_x h)^2\}] + [E_T(\partial_x T + \partial_x h \partial_y T) + E_\Gamma \partial_x \Gamma] \sqrt{1 + (\partial_x h)^2} = 0, \quad (7)$$

$$p_a - p + \frac{2\mu}{[1 + (\partial_x h)^2]} [\partial_y v \{1 - (\partial_x h)^2\} - (\partial_y u + \partial_x v) \partial_x h] = \frac{\sigma \partial_{xx} h}{[1 + (\partial_x h)^2]^{3/2}}, \quad (8)$$

where  $p_a$  specifies the ambient pressure above the film surface. To describe the convection of heat transfer at the film surface,  $y = h(x, t)$ , we use Newton's law of cooling [27,58]:

$$\kappa \frac{(\partial_x T \partial_x h - \partial_y T)}{\sqrt{1 + (\partial_x h)^2}} = \lambda(T - T_a), \quad (9)$$

in which  $\lambda$  indicates the heat-transfer coefficient. Moreover, for describing the transport of surface surfactant concentration on the film surface,  $y = h(x, t)$ , we use the following transport equation [51,59,60]:

$$\begin{aligned} \partial_t \Gamma + u \partial_x \Gamma + \frac{\Gamma[\partial_x u + \partial_x h \partial_x v + \partial_x h(\partial_y u + \partial_x h \partial_y v)]}{[1 + (\partial_x h)^2]} \\ = \frac{D_s}{\sqrt{1 + (\partial_x h)^2}} \partial_x [\partial_x \Gamma / \sqrt{1 + (\partial_x h)^2}] + J_{ad}, \end{aligned} \quad (10)$$

in which  $D_s$  denotes the surface surfactant diffusivity and  $J_{ad}$  denotes the net flux of soluble surfactant that characterizes the mass exchange between the bulk and the film surface. The mass flux,  $J_{ad}$ , can also be articulated in terms of the adsorption and desorption rate constants  $k_a$  and  $k_d$  as follows [61,62]:

$$J_{ad} = k_a(1 - \Gamma/\Gamma_\infty)c_b - k_d \Gamma. \quad (11)$$

Here,  $\Gamma_\infty$  is the surface surfactant concentration at maximum packing. The net flux of bulk surfactant concentration at the film surface,  $y = h(x, t)$ , can also be described by the following Fickian diffusion [51]:

$$J_{ad} = \frac{D_b}{\sqrt{1 + (\partial_x h)^2}} (\partial_x h \partial_x c_b - \partial_y c_b). \quad (12)$$

Finally, we use the kinematic boundary condition that describes the time evolution of the film surface,  $y = h(x, t)$ :

$$\partial_t h + u \partial_x h = v. \quad (13)$$

On the other hand, we propose the following linear, spring-backed, isotropic flexible plate model [10,11,20,21,39,46] that governs the deformation,  $y = \eta(x, t)$ , of the uniformly heated flexible substrate:

$$M \partial_{tt} \eta + D_c \partial_t \eta + B \partial_{xxxx} \eta - T_l \partial_{xx} \eta + K \eta = (p_w - p) + 2\mu[\partial_y v - (\partial_y u + \partial_x v) \partial_x \eta]. \quad (14)$$

Here, we restrict our study to the scenario where the flexible substrate is constrained to move only in the normal direction from its equilibrium position. Here,  $M$ ,  $D_c$ ,  $B$ ,  $T_l$ ,  $K$ , and  $p_w$ , respectively, indicate the plate mass per unit area, damping coefficient, flexural rigidity, longitudinal tension per unit width, spring stiffness per unit width, and pressure below the flexible substrate, which are assumed to be constant in the current study. Since the flexible substrate is uniformly heated, it has no thermal expansion, and thus, we neglect the thermal stress [38]. Finally, the velocity components of the liquid, the temperature of the liquid, and the bulk surfactant concentration at

the uniformly heated flexible substrate,  $y = \eta(x, t)$ , must satisfy the following no-slip, continuity of velocity, constant temperature, and no mass flux boundary conditions [42,46]:

$$u = 0, \quad v = \partial_t \eta, \quad T = T_w, \quad (\partial_x \eta \partial_x c_b - \partial_y c_b) = 0. \quad (15)$$

### Analysis of base flow

To execute the linear stability analysis of the base flow, it is necessary to have its solution. Consequently, we consider a steady, unidirectional parallel flow driven by gravity, the so-called base flow, which has a constant film layer thickness,  $y = d$ , from the undeformed flexible plate location,  $y = \eta = 0$ . In this situation,  $\Gamma_a$  is the surface surfactant concentration and  $\sigma_a$  is the surface tension of the incompressible viscous film. These assumptions reduce the governing equations (2)–(15) to the following forms:

$$\mu \partial_{yy} u + \rho g \sin \theta = 0, \quad \partial_y p + \rho g \cos \theta = 0, \quad (16)$$

$$\partial_{yy} T = 0, \quad \partial_{yy} c_b = 0, \quad (17)$$

$$\partial_y u = 0, \quad \lambda(T - T_a) + \kappa \partial_y T = 0, \quad \text{at } y = d, \quad (18)$$

$$p = p_a, \quad J_{ad} = 0, \quad \partial_y c_b = 0, \quad \text{at } y = d, \quad (19)$$

$$u = 0, \quad p = p_w, \quad T = T_w, \quad \partial_y c_b = 0, \quad \text{at } y = 0. \quad (20)$$

Solving Eqs. (16)–(20), we obtain the following nondimensional analytical solution for the current base flow:

$$\begin{aligned} \bar{U}(y) &= (2y - y^2), \quad \bar{V}(y) = 0, \quad \bar{\Gamma} = \Gamma_E, \\ \bar{P}(y) &= P_a + 2 \cot \theta (1 - y), \quad \bar{T}(y) = 1 - \frac{\text{Bi } y}{(1 + \text{Bi})}, \\ P_w &= P_a + 2 \cot \theta, \quad \bar{c}_b(y) = \frac{\Gamma_E}{\xi_s(1 - \Gamma_E)} = c_E, \end{aligned} \quad (21)$$

where the surface velocity for the base flow,  $U_s = \rho g d^2 \sin \theta / (2\mu)$ , is selected as the characteristic velocity scale; the base film thickness,  $d$ , is selected as the characteristic length scale;  $\mu U_s / d$  is selected as the characteristic pressure scale;  $d / U_s$  is selected as the characteristic timescale;  $(T_w - T_a)$  is selected as the characteristic scale for the temperature difference  $(T - T_a)$ ;  $\Gamma_\infty / d$  is selected as the characteristic scale for the bulk surfactant concentration; and  $\Gamma_\infty$  is selected as the characteristic scale for the surface surfactant concentration. Here,  $P_a = p_a d / (\mu U_s)$  denotes the nondimensional ambient pressure;  $P_w = p_w d / (\mu U_s)$  denotes the nondimensional pressure below the flexible plate;  $\text{Bi} = \lambda d / \kappa$  denotes the Biot number, which compares the convective heat transfer at the film surface to the heat conduction within the viscous film;  $\Gamma_E = \Gamma_a / \Gamma_\infty$  denotes the nondimensional base surface surfactant concentration; and  $\xi_s = k_a / (k_d d)$  denotes the measure of surfactant solubility. If  $\xi_s$  remains low ( $\xi_s \ll 1$ ), it creates a highly soluble surfactant-laden nonisothermal film flow over a flexible substrate, while if  $\xi_s$  is very high ( $\xi_s \gg 1$ ), it creates an insoluble-surfactant-impured nonisothermal film flow over a flexible substrate. Clearly,  $\Gamma_E$  has an admissible range of values,  $0 \leq \Gamma_E \leq 1$ . It should be noted that the presence of heat transfer and soluble surfactant has no impact on the base flow velocity  $\bar{U}(y)$  in the streamwise direction. However, the base flow temperature  $\bar{T}(y)$ , which is maximum at the flexible substrate and minimum at the film surface, is dependent on the Biot number  $\text{Bi}$ . In fact,  $\bar{T}(y)$  decreases linearly in the cross-stream direction as  $y$  increases due to pure conduction, provided  $\text{Bi} \neq 0$ . However, the base flow temperature will be uniform when  $\text{Bi} = 0$ , because  $\bar{T}(y) = 1$  at  $\text{Bi} = 0$ ,  $\forall y \in [0, 1]$ . On the other hand, the base flow bulk surfactant concentration,  $\bar{c}_b(y)$ , remains constant and becomes infinity at  $\Gamma_E \rightarrow 1$ . Hence,  $\xi_s$  must be very large in the base flow to ensure that the bulk surfactant concentration remains

bounded. Moreover,  $\bar{c}_b(y)$  approaches zero as soon as  $\xi_s(1 - \Gamma_E) \rightarrow \infty$ , yielding a viscous film flow contaminated by an insoluble surfactant over a flexible substrate [40] if we remove the effect of heat transfer.

### III. LINEAR STABILITY ANALYSIS

#### A. Linearized perturbation equations

This section concerns the linear stability analysis for disturbances of infinitesimal amplitudes. To execute this, we apply an infinitesimal perturbation on the base flow. As a result, the flexible substrate also undergoes an infinitesimal deformation from its equilibrium position,  $\eta = 0$ . Accordingly, we can decompose each flow variable into a steady base part and an unsteady perturbation part:

$$\begin{aligned} u(x, y, t) &= \bar{U}(y) + u'(x, y, t), \\ v(x, y, t) &= v'(x, y, t), \\ p(x, y, t) &= \bar{P}(y) + p'(x, y, t), \\ T(x, y, t) &= \bar{T}(y) + T'(x, y, t), \\ c_b(x, y, t) &= c_E + c'_b(x, y, t), \\ \eta(x, t) &= \eta'(x, t), \\ h(x, t) &= 1 + h'(x, t), \\ \Gamma(x, t) &= \Gamma_E + \Gamma'(x, t). \end{aligned} \quad (22)$$

Here, the bar notation variables are the steady base part, and the prime notation variables are the unsteady perturbation part. After substituting ansatz (22) into the governing equations (2)–(15), we linearize them with respect to the base flow solution. This mathematical process yields the following set of nondimensional perturbation equations:

$$\partial_x u' + \partial_y v' = 0, \quad (23)$$

$$\text{Re}(\partial_t u' + \bar{U} \partial_x u' + v' D \bar{U}) + \partial_x p' - (\partial_{xx} u' + \partial_{yy} u') = 0, \quad (24)$$

$$\text{Re}(\partial_t v' + \bar{U} \partial_x v') + \partial_y p' - (\partial_{xx} v' + \partial_{yy} v') = 0, \quad (25)$$

$$\text{Pe}_T(\partial_t T' + \bar{U} \partial_x T' + v' D \bar{T}) - (\partial_{xx} T' + \partial_{yy} T') = 0, \quad (26)$$

$$\text{Pe}_b(\partial_t c'_b + \bar{U} \partial_x c'_b) - (\partial_{xx} c'_b + \partial_{yy} c'_b) = 0. \quad (27)$$

Here,  $\text{Re} = \rho U_s d / \mu$  denotes the Reynolds number, which compares inertia to the viscous stress;  $\text{Pe}_T = \rho c_p U_s d / \kappa = \text{Re} \text{Pr}$  denotes the thermal Péclet number, which compares the advective heat transfer to the diffusive heat transfer;  $\text{Pr} = \mu c_p / \kappa$  denotes the Prandtl number; and  $\text{Pe}_b = U_s d / D_b$  denotes the solutal Péclet number, which compares the advective mass transfer to the diffusive mass transfer. The above governing equations (23)–(27) are subjected to the following linearized nondimensional boundary conditions: (1) At the film surface,  $y = 1$ ,

$$\partial_y u' + \partial_x v' + D^2 \bar{U} h' + \text{Ma}_T(\partial_x T' + D \bar{T} \partial_x h') + \text{Ma}_\Gamma \partial_x \Gamma' = 0, \quad (28)$$

$$-p' + 2 \cot \theta h' + 2 \partial_y v' - (\text{Ca}^{-1} - \text{Ma}_T \bar{T}) \partial_{xx} h' = 0, \quad (29)$$

$$\partial_y T' + \text{Bi}(D \bar{T} h' + T') = 0, \quad (30)$$

$$\partial_t \Gamma' + \bar{U} \partial_x \Gamma' + \Gamma_E \partial_x u' - (1/\text{Pe}_\Gamma) \partial_{xx} \Gamma' - k_s [\xi_s (1 - \Gamma_E) c'_b - (1 + \xi_s c_E) \Gamma'] = 0, \quad (31)$$

$$(1/\text{Pe}_b) \partial_y c'_b + k_s [\xi_s (1 - \Gamma_E) c'_b - (1 + \xi_s c_E) \Gamma'] = 0, \quad (32)$$

$$\partial_t h' + \bar{U} \partial_x h' - v' = 0. \quad (33)$$

Here,  $\text{Ma}_T = E_T(T_w - T_a)/(\mu U_s)$  denotes the thermal Marangoni number, which compares the thermal Marangoni stress generated by the surface tension gradient due to a change in temperature to the viscous stress;  $\text{Ma}_\Gamma = E_\Gamma \Gamma_\infty/(\mu U_s)$  denotes the surfactant Marangoni number, which compares the surfactant Marangoni stress generated by the surface tension gradient due to a change in surface surfactant concentration to the viscous stress;  $\text{Ca} = \mu U_s/\sigma_a$  denotes the capillary number, which compares the viscous stress to the capillary stress generated by the base surface tension;  $\text{Pe}_\Gamma = U_s d/D_s$  denotes the surfactant Péclet number, which compares the advective mass transfer to the diffusive mass transfer at the film surface; and  $k_s = k_d d/U_s$  denotes the ratio of the advective timescale to the desorptive timescale. (2) At the flexible substrate,  $y = 0$ ,

$$C_M \partial_{tt} \eta' + C_D \partial_t \eta' + C_B \partial_{xxxx} \eta' - C_T \partial_{xx} \eta' + C_K \eta' + p' - 2 \cot \theta \eta' - 2(\partial_y v' - D\bar{U} \partial_x \eta') = 0, \quad (34)$$

$$u' + D\bar{U} \eta' = 0, \quad v' - \partial_t \eta' = 0, \quad (35)$$

$$T' + D\bar{T} \eta' = 0, \quad \partial_y c'_b = 0. \quad (36)$$

Here,  $C_M = MU_s/\mu$  denotes the measure of nondimensional plate inertia,  $C_D = D_c d/\mu$  denotes the measure of nondimensional damping coefficient,  $C_B = B/(\mu U_s d^2)$  denotes the measure of nondimensional plate flexural rigidity,  $C_T = T_l/(\mu U_s)$  denotes the measure of nondimensional longitudinal tension,  $C_K = K d^2/(\mu U_s)$  denotes the measure of nondimensional spring stiffness, and  $D^i = d^i/dy^i$  ( $i = 1, 2$ ) denote the first and second derivatives with respect to  $y$ . The order of magnitudes of various physical dimensional parameters and the reasonable range of values for the associated nondimensional flow parameters are provided in Sec. S1 of the Supplemental Material [63].

Next, the solution of the perturbation equations (23)–(36) is assumed to be in the form of a normal mode:

$$\begin{aligned} u'(x, y, t) &= \tilde{u}(y) \exp[ik(x - ct)] + \text{c.c.}, \\ v'(x, y, t) &= \tilde{v}(y) \exp[ik(x - ct)] + \text{c.c.}, \\ p'(x, y, t) &= \tilde{p}(y) \exp[ik(x - ct)] + \text{c.c.}, \\ T'(x, y, t) &= \tilde{T}(y) \exp[ik(x - ct)] + \text{c.c.}, \\ c'_b(x, y, t) &= \tilde{c}_b(y) \exp[ik(x - ct)] + \text{c.c.}, \\ h'(x, t) &= \tilde{h} \exp[ik(x - ct)] + \text{c.c.}, \\ \eta'(x, t) &= \tilde{\eta} \exp[ik(x - ct)] + \text{c.c.}, \\ \Gamma'(x, t) &= \tilde{\Gamma} \exp[ik(x - ct)] + \text{c.c.}, \end{aligned} \quad (37)$$

where c.c. is the complex conjugate, and the variables marked with a tilde notation represent the amplitudes of the perturbation variables. The wave speed and the wave number of the infinitesimal perturbation are denoted by  $c$  and  $k$ , respectively. As we will focus on exploring the temporal stability analysis, the wave speed  $c = c_r + ic_i$  is a complex quantity, while the wave number  $k$  is a real quantity. After substitution of the normal mode solution (37) in the perturbation

equations (23)–(36), we obtain the following boundary value problem:

$$ik\tilde{u} + D\tilde{v} = 0, \quad (38)$$

$$\text{Re}[ik(\bar{U} - c)\tilde{u} + \tilde{v}D\bar{U}] + ik\tilde{p} - (D^2\tilde{u} - k^2\tilde{u}) = 0, \quad (39)$$

$$\text{Re } ik(\bar{U} - c)\tilde{v} + D\tilde{p} - (D^2\tilde{v} - k^2\tilde{v}) = 0, \quad (40)$$

$$\text{Pe}_T[ik(\bar{U} - c)\tilde{T} + \tilde{v}D\bar{T}] - (D^2\tilde{T} - k^2\tilde{T}) = 0, \quad (41)$$

$$\text{Pe}_b ik(\bar{U} - c)\tilde{c}_b - (D^2\tilde{c}_b - k^2\tilde{c}_b) = 0, \quad (42)$$

$$D\tilde{u} + ik\tilde{v} + D^2\bar{U}\tilde{h} + \text{Ma}_T ik(\tilde{T} + D\bar{T}\tilde{h}) + \text{Ma}_\Gamma ik\tilde{r} = 0, \quad \text{at } y = 1, \quad (43)$$

$$-\tilde{p} + 2 \cot \theta \tilde{h} + 2D\tilde{v} + k^2(\text{Ca}^{-1} - \text{Ma}_T \bar{T})\tilde{h} = 0, \quad \text{at } y = 1, \quad (44)$$

$$D\tilde{T} + \text{Bi}(D\bar{T}\tilde{h} + \tilde{T}) = 0, \quad \text{at } y = 1, \quad (45)$$

$$(\bar{U} - c)ik\tilde{r} + ik\Gamma_E\tilde{u} + (1/\text{Pe}_\Gamma)k^2\tilde{r} - k_s[\xi_s(1 - \Gamma_E)\tilde{c}_b - (1 + \xi_s c_E)\tilde{r}] = 0, \quad \text{at } y = 1, \quad (46)$$

$$(1/\text{Pe}_b)D\tilde{c}_b + k_s[\xi_s(1 - \Gamma_E)\tilde{c}_b - (1 + \xi_s c_E)\tilde{r}] = 0, \quad \text{at } y = 1, \quad (47)$$

$$ik(\bar{U} - c)\tilde{h} - \tilde{v} = 0, \quad \text{at } y = 1, \quad (48)$$

$$\begin{aligned} & -C_M k^2 c^2 \tilde{\eta} - C_D ikc \tilde{\eta} + C_B k^4 \tilde{\eta} + C_T k^2 \tilde{\eta} + C_K \tilde{\eta} + \tilde{p} - 2 \cot \theta \tilde{\eta} \\ & - 2(D\tilde{v} - ikD\bar{U}\tilde{\eta}) = 0, \quad \text{at } y = 0, \end{aligned} \quad (49)$$

$$\tilde{u} + D\bar{U}\tilde{\eta} = 0, \quad \tilde{v} + ikc \tilde{\eta} = 0, \quad \text{at } y = 0, \quad (50)$$

$$\tilde{T} + D\bar{T}\tilde{\eta} = 0, \quad D\tilde{c}_b = 0, \quad \text{at } y = 0. \quad (51)$$

## B. Long-wave analysis

To explore primary instability governed by Eqs. (38)–(51), we employ the long-wave series expansion [29] in the limit  $k \sim (1/\lambda) \rightarrow 0$ , where  $\lambda$  is the wavelength of the infinitesimal disturbance:

$$\begin{aligned} \tilde{u}(y) &= \tilde{u}_0(y) + k\tilde{u}_1(y) + k^2\tilde{u}_2(y) + \dots, \\ \tilde{v}(y) &= k\tilde{v}_1(y) + k^2\tilde{v}_2(y) + k^3\tilde{v}_3(y) + \dots, \\ \tilde{p}(y) &= \tilde{p}_0(y) + k\tilde{p}_1(y) + k^2\tilde{p}_2(y) + \dots, \\ \tilde{T}(y) &= \tilde{T}_0(y) + k\tilde{T}_1(y) + k^2\tilde{T}_2(y) + \dots, \\ \tilde{c}_b(y) &= \tilde{c}_{b0}(y) + k\tilde{c}_{b1}(y) + k^2\tilde{c}_{b2}(y) + \dots, \\ \tilde{h} &= \tilde{h}_0 + k\tilde{h}_1 + k^2\tilde{h}_2 + \dots, \\ \tilde{\eta} &= \tilde{\eta}_0 + k\tilde{\eta}_1 + k^2\tilde{\eta}_2 + \dots, \\ \tilde{r} &= \tilde{r}_0 + k\tilde{r}_1 + k^2\tilde{r}_2 + \dots, \\ c &= c_0 + kc_1 + k^2c_2 + \dots. \end{aligned} \quad (52)$$

It is important to note that the series expansions presented in Eq. (52) are only valid in the limit  $k \rightarrow 0$ , since they may deviate from the true numerical solution at finite wave number. Using the Cauchy ratio test, if we calculate the radius of convergence,  $\mathcal{R}$ , we see that

$\mathcal{R} = \lim_{n \rightarrow \infty} \left| \frac{k^n}{k^{n+1}} \frac{Y_n}{Y_{n+1}} \right| = \left| \frac{1}{k} \frac{Y_n}{Y_{n+1}} \right|$ , where  $Y_{n+1}$  and  $Y_n$  represent the  $(n+1)$ th and  $n$ th terms for the amplitudes of flow variables in the long-wave series expansions. This shows that the radius of convergence depends on  $k$  as well as the flow parameters. Hence, the radius of convergence will increase gradually and become very large as  $k$  decreases and approaches zero. However, if  $k$  increases, the radius of convergence will decrease for a given set of flow parameters. Therefore, one can conclude that the long-wave series solution will deviate from the true numerical solution in the finite wave-number regime as  $k$  increases. This is why we will focus here solely on the long-wave mode.

### C. Zeroth-order [ $\mathcal{O}(k^0)$ ] long-wave equations

After substitution of Eq. (52) in Eqs. (38)–(51), we collect the zeroth-order [ $\mathcal{O}(k^0)$ ] long-wave equations:

$$D^2 \tilde{u}_0 = 0, \quad D \tilde{p}_0 = 0, \quad D^2 \tilde{T}_0 = 0, \quad D^2 \tilde{c}_{b0} = 0, \quad (53)$$

$$D \tilde{u}_0 + D^2 \tilde{U} \tilde{h}_0 = 0, \quad \tilde{p}_0 - 2 \cot \theta \tilde{h}_0 = 0, \quad \text{at } y = 1, \quad (54)$$

$$D \tilde{T}_0 + \text{Bi}(D \tilde{T} \tilde{h}_0 + \tilde{T}_0) = 0, \quad D \tilde{c}_{b0} = 0, \quad \text{at } y = 1, \quad (55)$$

$$C_K \tilde{\eta}_0 + \tilde{p}_0 - 2 \cot \theta \tilde{\eta}_0 = 0, \quad D \tilde{c}_{b0} = 0, \quad \text{at } y = 0, \quad (56)$$

$$\tilde{u}_0 + D \tilde{U} \tilde{\eta}_0 = 0, \quad \tilde{T}_0 + D \tilde{T} \tilde{\eta}_0 = 0, \quad \text{at } y = 0. \quad (57)$$

Here, it is assumed that the capillary number  $\text{Ca}$  is of order 1 [ $\text{Ca} \sim \mathcal{O}(1)$ ] in the long-wave calculation, which explains the absence of the capillary term in the zeroth-order normal stress boundary condition. However, there is no restriction on the choice of the capillary number when the numerical simulation is performed (see Sec. IV for details). After solving the zeroth-order equations (53)–(57), the following solution can be obtained:

$$\begin{aligned} \tilde{u}_0(y) &= (2\tilde{h}_0 y - 2\tilde{\eta}_0), \quad \tilde{p}_0(y) = 2 \cot \theta \tilde{h}_0, \\ \tilde{c}_{b0}(y) &= \frac{(1 + \xi_s c_E) \tilde{T}_0}{\xi_s (1 - \Gamma_E)}, \quad \tilde{\eta}_0 = -\frac{2 \cot \theta \tilde{h}_0}{C_K - 2 \cot \theta}, \\ \tilde{T}_0(y) &= \frac{\text{Bi}^2 (\tilde{h}_0 - \tilde{\eta}_0) y}{(1 + \text{Bi})^2} + \frac{\text{Bi} \tilde{\eta}_0}{(1 + \text{Bi})}, \end{aligned} \quad (58)$$

provided  $C_K \neq 2 \cot \theta$  and  $\xi_s (1 - \Gamma_E) \neq 0$ . Now, we denote the critical value of the spring stiffness coefficient,  $C_K$ , by  $C_K^* = 2 \cot \theta$ . In fact, when  $C_K = C_K^*$ , the zeroth-order amplitude,  $\tilde{\eta}_0$ , of the flexible plate deflection is indeterminable. On the other hand,  $\tilde{\eta}_0$  approaches zero at  $C_K \rightarrow \infty$  when  $\theta \neq 0$  and  $\theta \neq \pi/2$ . Indeed, this is the situation when the flexible substrate behaves as a rigid substrate. Moreover, the zeroth-order amplitude,  $\tilde{u}_0(y)$ , of the streamwise perturbation velocity is explicitly dependent on the coefficient of spring stiffness  $C_K$ , and therefore, it is affected by the distortion of the flexible substrate. In particular, the zeroth-order amplitude,  $\tilde{u}_0(y)$ , increases if  $\tilde{h}_0$  and  $\tilde{\eta}_0$  are out of phase but decreases if they are in phase. Unlike the amplitude of the streamwise perturbation velocity, the zeroth-order amplitude,  $\tilde{T}_0(y)$ , of the temperature perturbation is also explicitly dependent on the deformation of the flexible substrate. Interestingly, if we remove the effect of spring stiffness ( $C_K = 0$ ), we obtain  $\tilde{h}_0 = \tilde{\eta}_0$ , which converts the zeroth-order amplitude,  $\tilde{T}_0(y)$ , of the temperature perturbation uniform across the zeroth-order perturbation flow for the given parameter values. This is because  $\tilde{T}_0(y)$  is independent of the cross-stream coordinate  $y$ , as shown below:

$$\tilde{T}_0(y) = \frac{\text{Bi} \tilde{\eta}_0}{(1 + \text{Bi})}.$$

Additionally, in the limit  $\xi_s (1 - \Gamma_E) \rightarrow \infty$ ,  $\tilde{c}_{b0}(y) \rightarrow 0$ , i.e., the zeroth-order amplitude of the perturbation bulk surfactant concentration vanishes. This leads to the situation of an

insoluble-surfactant-impured isothermal film flowing over a flexible substrate if the effect of heat transfer is excluded [40].

#### D. First-order $[\mathcal{O}(k^1)]$ long-wave equations

Now, we collect the first-order  $[\mathcal{O}(k^1)]$  long-wave equations:

$$i\tilde{u}_0 + D\tilde{v}_1 = 0, \quad (59)$$

$$\text{Re}[i(\bar{U} - c_0)\tilde{u}_0 + D\bar{U}\tilde{v}_1] + i\tilde{p}_0 - D^2\tilde{u}_1 = 0, \quad (60)$$

$$D\tilde{p}_1 - D^2\tilde{v}_1 = 0, \quad (61)$$

$$\text{Pe}_T[i(\bar{U} - c_0)\tilde{T}_0 + D\bar{T}\tilde{v}_1] - D^2\tilde{T}_1 = 0, \quad (62)$$

$$\text{Pe}_b i(\bar{U} - c_0)\tilde{c}_{b0} - D^2\tilde{c}_{b1} = 0, \quad (63)$$

$$D\tilde{u}_1 + \tilde{h}_1 D^2\bar{U} + i\text{Ma}_T(\tilde{T}_0 + D\bar{T}\tilde{h}_0) + i\text{Ma}_\Gamma\tilde{F}_0 = 0, \quad \text{at } y = 1, \quad (64)$$

$$-\tilde{p}_1 + 2\cot\theta\tilde{h}_1 + 2D\tilde{v}_1 = 0, \quad \text{at } y = 1, \quad (65)$$

$$D\tilde{T}_1 + \text{Bi}(D\bar{T}\tilde{h}_1 + \tilde{T}_1) = 0, \quad \text{at } y = 1, \quad (66)$$

$$D\tilde{c}_{b1} + i\text{Pe}_b[(\bar{U} - c_0)\tilde{F}_0 + \Gamma_E\tilde{u}_0] = 0, \quad \text{at } y = 1, \quad (67)$$

$$i(\bar{U} - c_0)\tilde{h}_0 - \tilde{v}_1 = 0, \quad \text{at } y = 1, \quad (68)$$

$$\begin{aligned} & -iC_D c_0 \tilde{\eta}_0 + C_K \tilde{\eta}_1 + \tilde{p}_1 - 2\cot\theta\tilde{\eta}_1 \\ & - 2(D\tilde{v}_1 - D\bar{U}\tilde{\eta}_0) = 0, \quad \text{at } y = 0, \end{aligned} \quad (69)$$

$$\tilde{u}_1 + D\bar{U}\tilde{\eta}_1 = 0, \quad \tilde{v}_1 + ic_0\tilde{\eta}_0 = 0, \quad \text{at } y = 0, \quad (70)$$

$$\tilde{T}_1 + D\bar{T}\tilde{\eta}_1 = 0, \quad D\tilde{c}_{b1} = 0, \quad \text{at } y = 0. \quad (71)$$

The solution of the first-order long-wave equations (59)–(71) can be found in Sec. S2 of the Supplemental Material [63]. We can see that when  $C_K = C_K^* = 2\cot\theta$ , the first-order amplitude,  $\tilde{\eta}_1$ , of the flexible plate deflection cannot be determined from the plate deformation equation (69). From the first-order long-wave solution, we can also report that the surface mode generated owing to the perturbation on the film surface propagates with a constant phase speed of  $c_0 = 2$  in the limit  $k \rightarrow 0$ . Clearly, the presence of a uniformly heated flexible plate and soluble surfactant does not affect the phase speed of the first-order long-wave disturbance.

#### E. Second-order $[\mathcal{O}(k^2)]$ long-wave equations

Now, we collect the second-order  $[\mathcal{O}(k^2)]$  long-wave equations:

$$i\tilde{u}_1 + D\tilde{v}_2 = 0, \quad (72)$$

$$\text{Re}[i(\bar{U} - c_0)\tilde{u}_1 - ic_1\tilde{u}_0 + D\bar{U}\tilde{v}_2] + i\tilde{p}_1 - (D^2\tilde{u}_2 - \tilde{u}_0) = 0, \quad (73)$$

$$\text{Re } i(\bar{U} - c_0)\tilde{v}_1 + D\tilde{p}_2 - D^2\tilde{v}_2 = 0, \quad (74)$$

$$\text{Pe}_T[i(\bar{U} - c_0)\tilde{T}_1 - ic_1\tilde{T}_0 + D\bar{T}\tilde{v}_2] - (D^2\tilde{T}_2 - \tilde{T}_0) = 0, \quad (75)$$

$$\text{Pe}_b[i(\bar{U} - c_0)\tilde{c}_{b1} - ic_1\tilde{c}_{b0}] - (D^2\tilde{c}_{b2} - \tilde{c}_{b0}) = 0, \quad (76)$$

$$D\tilde{u}_2 + i\tilde{v}_1 + \tilde{h}_2 D^2\bar{U} + i\text{Ma}_T(\tilde{T}_1 + D\tilde{T}\tilde{h}_1) + i\text{Ma}_T\tilde{T}_1 = 0, \quad \text{at } y = 1, \quad (77)$$

$$-\tilde{p}_2 + 2\cot\theta\tilde{h}_2 + 2D\tilde{v}_2 + (\text{Ca}^{-1} - \text{Ma}_T\tilde{T})\tilde{h}_0 = 0, \quad \text{at } y = 1, \quad (78)$$

$$D\tilde{T}_2 + \text{Bi}(D\tilde{T}\tilde{h}_2 + \tilde{T}_2) = 0, \quad \text{at } y = 1, \quad (79)$$

$$D\tilde{c}_{b2} + i\text{Pe}_b[(\bar{U} - c_0)\tilde{T}_1 - c_1\tilde{T}_0 + \Gamma_E\tilde{u}_1] + (\text{Pe}_b/\text{Pe}_T)\tilde{T}_0 = 0, \quad \text{at } y = 1, \quad (80)$$

$$i(\bar{U} - c_0)\tilde{h}_1 - ic_1\tilde{h}_0 - \tilde{v}_2 = 0, \quad \text{at } y = 1, \quad (81)$$

$$\begin{aligned} & -C_M c_0^2 \tilde{\eta}_0 - iC_D(c_0\tilde{\eta}_1 + c_1\tilde{\eta}_0) + C_T\tilde{\eta}_0 + C_K\tilde{\eta}_2 + \tilde{p}_2 - 2\cot\theta\tilde{\eta}_2 \\ & - 2(D\tilde{v}_2 - iD\bar{U}\tilde{\eta}_1) = 0, \quad \text{at } y = 0, \end{aligned} \quad (82)$$

$$\tilde{u}_2 + D\bar{U}\tilde{\eta}_2 = 0, \quad \tilde{v}_2 + i(c_0\tilde{\eta}_1 + c_1\tilde{\eta}_0) = 0, \quad \text{at } y = 0, \quad (83)$$

$$\tilde{T}_2 + D\tilde{T}\tilde{\eta}_2 = 0, \quad D\tilde{c}_{b2} = 0, \quad \text{at } y = 0. \quad (84)$$

After solving the second-order equations (72)–(84), one can obtain the solution for  $\tilde{u}_2$ ,  $\tilde{v}_2$ ,  $\tilde{p}_2$ ,  $\tilde{T}_2$ , and  $\tilde{c}_{b2}$ . Next, we insert  $\tilde{v}_2$  in the kinematic boundary condition (81), which produces the following expression for  $c_1$  corresponding to the surface mode:

$$c_1 = i \left[ \frac{8}{15} \text{Re} - \frac{2}{3} \cot\theta + \frac{\text{Bi Ma}_T}{2(1 + \text{Bi})^2} + \frac{4 \cot^2\theta}{3 C_K} - \frac{3\text{Ma}_T \Gamma_E \xi_s (1 - \Gamma_E)^2}{4 + 3\xi_s (1 - \Gamma_E)^2} \right], \quad (85)$$

provided  $C_K \neq 0$ . Finally, we obtain the following critical Reynolds number,  $\text{Re}_c$ , for the surface mode using the neutral stability criterion ( $c_i \approx kc_1 = 0$  as  $k \rightarrow 0$ ):

$$\begin{aligned} \text{Re}_c = & \underbrace{\frac{5}{4} \cot\theta}_{\text{Effect of depthwise gravity}} - \underbrace{\frac{15}{16} \frac{\text{Bi Ma}_T}{(1 + \text{Bi})^2}}_{\text{Effect of temperature}} - \underbrace{\frac{5 \cot^2\theta}{2 C_K}}_{\text{Effect of flexible plate}} + \underbrace{\frac{45}{8} \left[ \frac{\text{Ma}_T \Gamma_E \xi_s (1 - \Gamma_E)^2}{4 + 3\xi_s (1 - \Gamma_E)^2} \right]}_{\text{Effect of soluble surfactant}}. \end{aligned} \quad (86)$$

In the formula for the critical Reynolds number  $\text{Re}_c$ , the first term reflects the stabilizing effect of depthwise gravity as  $\text{Re}_c$  increases if this term is increased, the second term reflects the destabilizing effect of temperature as  $\text{Re}_c$  decreases if this term is increased, the third term reflects the destabilizing effect of a flexible plate as  $\text{Re}_c$  decreases if this term is increased, and the fourth term reflects the stabilizing effect of soluble surfactant as  $\text{Re}_c$  increases if this term is increased. Clearly, in the absence of soluble surfactant and heat transfer, the critical Reynolds number for the surface mode matches with that of the isothermal film flowing over a flexible substrate [39]. Moreover, it captures the results of a falling film over a flexible substrate contaminated with a soluble surfactant [42] once we negate the effect of heat transfer from the current flow configuration. Furthermore, we obtain the critical Reynolds number for the nonisothermal film flowing over a uniformly heated flexible substrate if the fluid is free from soluble surfactant [46]. Additionally, in the limits of isothermal, insoluble-surfactant-laden film flow [ $\xi_s(1 - \Gamma_E) \rightarrow 0$ ,  $\text{Ma}_T \rightarrow 0$ ], the critical Reynolds number,  $\text{Re}_c$ , for the surface mode simplifies to the following form:

$$\begin{aligned} \text{Re}_c = & \underbrace{\frac{5}{4} \cot\theta}_{\text{Effect of depthwise gravity}} - \underbrace{\frac{5 \cot^2\theta}{2 C_K}}_{\text{Effect of flexible plate}} + \underbrace{\frac{15}{8} \text{Ma}_T \Gamma_E}_{\text{Effect of insoluble surfactant}}. \end{aligned} \quad (87)$$

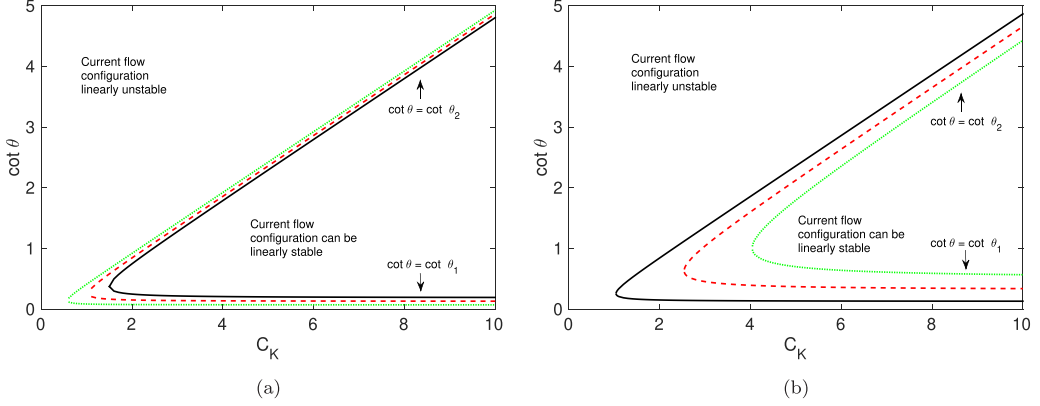


FIG. 2. (a) Variation of  $\cot \theta$  as a function of the spring stiffness coefficient  $C_K$  for different values of  $Ma_\Gamma$  when  $Ma_T = 1$ ,  $Bi = 1$ ,  $\xi_s = 100$ , and  $\Gamma_E = 0.9$ . Solid, dashed, and dotted lines represent the results for  $Ma_\Gamma = 0$ ,  $Ma_\Gamma = 0.1$ , and  $Ma_\Gamma = 0.2$ , respectively. (b) Variation of  $\cot \theta$  as a function of the spring stiffness coefficient  $C_K$  for different values of  $Ma_T$  when  $Ma_\Gamma = 0.1$ ,  $Bi = 1$ ,  $\xi_s = 100$ , and  $\Gamma_E = 0.9$ . Solid, dashed, and dotted lines represent the results for  $Ma_T = 1$ ,  $Ma_T = 2$ , and  $Ma_T = 3$ , respectively.

Indeed, expression (87) coincides with the finding reported in the study of insoluble-surfactant-laden film flowing over a compliant wall when there is no extra shear stress imposed at the film surface [40]. In addition, if we further set  $C_K \rightarrow \infty$  in expression (87),  $Re_c$  recovers the result of Blyth and Pozrikidis [64] and Wei [65] for an insoluble-surfactant-laden film flowing over a rigid inclined plane when a constant shear stress is not applied at the film surface. It should also be noted that, in the formula for  $Re_c$ , the second and third terms are negative, while the first and fourth terms are positive for  $0 < \theta < \pi/2$ . Hence, the critical Reynolds number for the onset of surface mode instability can be zero when these terms balance each other. If we set  $Re_c = 0$ , we obtain the following quadratic equation for  $\cot \theta$ :

$$\cot^2 \theta - \frac{C_K}{2} \cot \theta + \frac{3}{8} \left[ \frac{C_K Bi Ma_T}{(1 + Bi)^2} \right] - \frac{9}{4} \left[ \frac{C_K Ma_\Gamma \Gamma_E \xi_s (1 - \Gamma_E)^2}{4 + 3 \xi_s (1 - \Gamma_E)^2} \right] = 0. \quad (88)$$

Using Eq. (88), we can calculate a range of the inclination angle,  $\cot \theta_1 < \cot \theta < \cot \theta_2$ , where the current nonisothermal surfactant-contaminated film falling down a flexible substrate can be linearly stable, although the heat transfer and flexibility of the impermeable plate promote surface mode instability. This behavior arises from the stronger stabilizing effects of the surfactant Marangoni stress and depthwise gravitational force, which outweigh the destabilizing effects of the thermal Marangoni stress and spring stiffness of the flexible plate. In Fig. 2, we illustrate the associated results.

First, we produce outcomes when the surfactant Marangoni number,  $Ma_\Gamma$ , rises. Here, we observe that the unstable zone induced by the surface mode reduces, indicating a stabilizing influence of  $Ma_\Gamma$  [see Fig. 2(a)]. In contrast, if we increase the thermal Marangoni number  $Ma_T$ , the unstable zone created by the surface mode magnifies significantly, indicating a destabilizing influence of  $Ma_T$  [see Fig. 2(b)]. Additionally, by setting  $Re_c = 0$  in Eq. (86), we can determine the critical thermal Marangoni number for the onset of inertialess surface mode instability, which can be articulated as follows:

$$Ma_{Tc} = \frac{(1 + Bi)^2}{Bi} \left[ \frac{4}{3} \cot \theta - \frac{8 \cot^2 \theta}{3 C_K} + \frac{6 Ma_\Gamma \Gamma_E \xi_s (1 - \Gamma_E)^2}{4 + 3 \xi_s (1 - \Gamma_E)^2} \right]. \quad (89)$$

Therefore, if  $Ma_T > Ma_{Tc}$ , the inertialess instability induced by the surface mode will grow over time and make the current flow system unstable even though the Reynolds number vanishes; other-

wise, the flow system will be stable if  $\text{Ma}_T < \text{Ma}_{T_c}$  because the surface-mode-induced inertialess disturbance will decay over time. The physical mechanism of surface mode instability based on the speeds of the kinematic and dynamic hyperbolic waves is discussed in Sec. S3, while Sec. S4 of the Supplemental Material [63] provides the details of the inertialess instabilities generated by the surface mode and the wall mode.

#### IV. LINEAR STABILITY ANALYSIS IN LOW-TO-MODERATE REYNOLDS NUMBER ZONE

To execute the linear stability analysis in the arbitrary wave-number regime for infinitesimal disturbances, we will formulate the OS BVP. To this end, we introduce the perturbation stream function  $\psi'(x, y, t)$  from the perturbation continuity equation (23) with the aid of the relations  $u' = \partial_y \psi'$  and  $v' = -\partial_x \psi'$ . Assuming a normal mode solution of the perturbation equations, we can write the amplitude,  $\tilde{\psi}$ , of the perturbation stream function in the following form:

$$\tilde{u} = D\tilde{\psi}, \quad \tilde{v} = -ik\tilde{\psi}. \quad (90)$$

Here,  $\psi'(x, y, t) = \tilde{\psi} \exp[ik(x - ct)]$ . After insertion of Eq. (90) into Eqs. (38)–(51), we eliminate the pressure terms. This mathematical process yields the following OS BVP:

$$(D^2 - k^2)^2 \tilde{\psi} - \text{Re } ik[(\bar{U} - c)(D^2 - k^2)\tilde{\psi} - D^2 \bar{U} \tilde{\psi}] = 0, \quad (91)$$

$$(D^2 - k^2)\tilde{T} - \text{Pe}_T ik[(\bar{U} - c)\tilde{T} - D\tilde{T} \tilde{\psi}] = 0, \quad (92)$$

$$(D^2 - k^2)\tilde{c}_b - \text{Pe}_b ik(\bar{U} - c)\tilde{c}_b = 0, \quad (93)$$

$$(D^2 + k^2)\tilde{\psi} + D^2 \bar{U} \tilde{h} + ik\text{Ma}_T(\tilde{T} + D\tilde{T} \tilde{h}) + \text{Ma}_T ik \tilde{F} = 0, \quad \text{at } y = 1, \quad (94)$$

$$(D^2 - 3k^2)D\tilde{\psi} - \text{Re } ik(\bar{U} - c)D\tilde{\psi} - ik\tilde{h}[2 \cot \theta + k^2(\text{Ca}^{-1} - \text{Ma}_T \tilde{T})] = 0, \quad \text{at } y = 1, \quad (95)$$

$$D\tilde{T} + \text{Bi}(D\tilde{T} \tilde{h} + \tilde{T}) = 0, \quad \text{at } y = 1, \quad (96)$$

$$D\tilde{c}_b + ik\text{Pe}_b[(\bar{U} - c)\tilde{F} + \Gamma_E D\tilde{\psi}] + \frac{\text{Pe}_b}{\text{Pe}_T} k^2 \tilde{F} = 0, \quad \text{at } y = 1, \quad (97)$$

$$(\bar{U} - c)\tilde{h} + \tilde{\psi} = 0, \quad \text{at } y = 1, \quad (98)$$

$$-C_M ik^3 c^2 \tilde{\eta} + C_D k^2 c \tilde{\eta} + C_{\text{Bi}} k^5 \tilde{\eta} + C_T ik^3 \tilde{\eta} + C_K ik \tilde{\eta} - 2ik \cot \theta \tilde{\eta} + (D^2 - k^2)D\tilde{\psi} = 0, \quad \text{at } y = 0, \quad (99)$$

$$D\tilde{\psi} + D\bar{U} \tilde{\eta} = 0, \quad \tilde{\psi} - c \tilde{\eta} = 0, \quad \text{at } y = 0, \quad (100)$$

$$\tilde{T} + D\tilde{T} \tilde{\eta} = 0, \quad D\tilde{c}_b = 0, \quad \text{at } y = 0. \quad (101)$$

Next, our purpose is to solve this OS BVP [Eqs. (91)–(101)] numerically by using the Chebyshev spectral collocation method [66]. To achieve that, the amplitude of the perturbation stream function  $\tilde{\psi}(y)$  is expanded into a truncated series of the Chebyshev polynomials of the first kind  $\hat{P}_i(y)$  as written below:

$$\tilde{\psi}(y) = \sum_{i=0}^N \hat{\psi}_i \hat{P}_i(y), \quad \tilde{T}(y) = \sum_{i=0}^N \hat{T}_i \hat{P}_i(y), \quad \tilde{c}_b(y) = \sum_{i=0}^N \hat{c}_{bi} \hat{P}_i(y). \quad (102)$$

Here,  $N$  is the positive integer;  $\hat{\psi}_i$ ,  $\hat{T}_i$ , and  $\hat{c}_{bi}$  are unknown constants that have to be determined numerically. Since the OS BVP is defined in the domain  $[0, 1]$ , while the Chebyshev polynomials  $\hat{P}_i(y)$  are defined in the domain  $[-1, 1]$ , it is necessary to shift the domain of the OS BVP from

TABLE I. Comparison between the long-wave analytical and numerical values of the critical Reynolds number  $Re_c$  for the onset of surface mode instability when the thermal Marangoni number  $Ma_T$  varies. The other parameter values are  $C_M = C_D = 1$ ,  $C_B = C_T = 10^3$ ,  $C_K = 5$ ,  $Ca = 1$ ,  $Bi = 1$ ,  $Pr = 7$ ,  $Ma_\Gamma = 1$ ,  $Pe_b = Pe_\Gamma = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ .

| Critical Reynolds number | $Ma_T = 0$ | $Ma_T = 0.1$ | $Ma_T = 0.2$ | $Ma_T = 0.3$ | $Ma_T = 0.4$ | $Ma_T = 0.5$ |
|--------------------------|------------|--------------|--------------|--------------|--------------|--------------|
| Analytical ( $Re_c$ )    | 0.9345     | 0.9110       | 0.8876       | 0.8641       | 0.8407       | 0.8173       |
| Numerical ( $Re_c$ )     | 0.9347     | 0.9113       | 0.8878       | 0.8644       | 0.8410       | 0.8175       |

$[0, 1]$  to  $[-1, 1]$  using the transformation  $x = (2y - 1)$ . As a result, the various order derivatives are also converted as  $D \rightarrow 2D$ ,  $D^2 \rightarrow 4D^2$ , and so on. By inserting the series expansions (102) into the OS BVP, the Chebyshev polynomials are computed at the Gauss-Lobatto collocation points  $x_j = \cos(\pi j/N)$ , for  $j = 0, \dots, N$ . This mathematical process leads to the following polynomial eigenvalue problem:

$$[c^2 \mathcal{A}_2 + c \mathcal{A}_1 + \mathcal{A}_0] \mathcal{X} = 0. \quad (103)$$

Here,  $c$  is the eigenvalue;  $\mathcal{A}_2$ ,  $\mathcal{A}_1$ , and  $\mathcal{A}_0$  are the square matrices of dimension  $(3N + 5) \times (3N + 5)$ ;  $\mathcal{X} = [\hat{\psi}_0, \hat{\psi}_1, \dots, \hat{\psi}_N, \hat{h}, \hat{\eta}, \hat{T}_0, \hat{T}_1, \dots, \hat{T}_N, \hat{c}_{b0}, \hat{c}_{b1}, \dots, \hat{c}_{bN}]^T$  is a column matrix of dimension  $(3N + 5) \times 1$ . Finally, the eigenvalues are computed numerically using the matlab routine `polyeig`. First, we verify our numerical code by comparing the numerical and long-wave analytical results. To this end, we calculate the critical Reynolds number for the onset of surface mode instability when the thermal Marangoni number and the surfactant Marangoni number vary, while other parameters are kept fixed to their values at  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = C_T = 10^3$ ,  $C_K = 5$ ,  $Ca = 1$ ,  $Pr = 7$ ,  $Bi = 1$ ,  $Pe_b = Pe_\Gamma = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ . In Tables I and II, we have demonstrated a comparison of the critical Reynolds number for the onset of surface mode instability. Clearly, there is an excellent match between the long-wave analytical and numerical findings. This fact also confirms the accuracy of the current numerical code. From Table I, we can conclude that the onset of surface mode instability decreases as the thermal Marangoni number rises, while Table II exhibits that the onset of surface mode instability increases as the surfactant Marangoni number rises. Therefore, surface mode instability is destabilized by the thermal Marangoni stress, while it is stabilized by the surfactant Marangoni stress. In Fig. 3(a), we further graphically illustrate these two opposing events.

Physically, if the thermal Marangoni stress, or equivalently  $Ma_T$ , is increased, a local film flow is generated from the trough to the crest of the deformed film surface by the surface tension gradient caused by temperature differences, which is high at the trough and low at the crest. This thermal Marangoni stress pushes the fluid toward the crest and elevates the surface amplitude, resulting in a destabilizing influence on surface mode instability. In contrast, when the surfactant Marangoni stress, or equivalently  $Ma_\Gamma$ , is increased, a local film flow is generated from the crest to the trough of the deformed film surface by the surface tension gradient caused by the differences in surfactant concentration, which is high at the crest and low at the trough because the surfactant concentration

TABLE II. Comparison between the long-wave analytical and numerical values of the critical Reynolds number  $Re_c$  for the onset of surface mode instability when the surfactant Marangoni number  $Ma_\Gamma$  varies. The other parameter values are  $C_M = C_D = 1$ ,  $C_B = C_T = 10^3$ ,  $C_K = 5$ ,  $Ca = 1$ ,  $Bi = 1$ ,  $Pr = 7$ ,  $Ma_T = 0.5$ ,  $Pe_b = Pe_\Gamma = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ .

| Critical Reynolds number | $Ma_\Gamma = 1$ | $Ma_\Gamma = 2$ | $Ma_\Gamma = 3$ | $Ma_\Gamma = 4$ | $Ma_\Gamma = 5$ | $Ma_\Gamma = 6$ |
|--------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Analytical ( $Re_c$ )    | 0.8173          | 1.0017          | 1.1862          | 1.3707          | 1.5551          | 1.7396          |
| Numerical ( $Re_c$ )     | 0.8175          | 1.0022          | 1.1869          | 1.3716          | 1.5563          | 1.7409          |

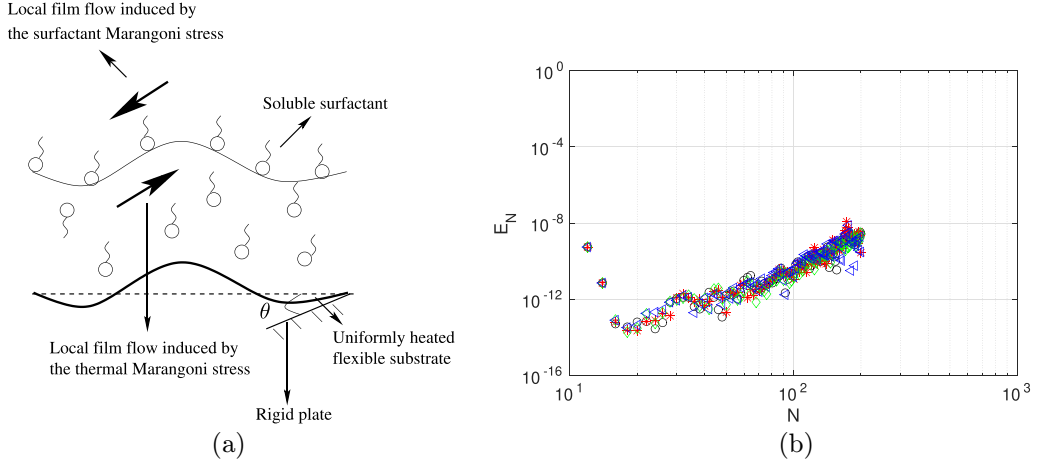


FIG. 3. (a) A sketch of a physical mechanism of surface mode instability induced by the thermal and surfactant Marangoni stresses. (b) The variation of the relative error  $E_N$  is shown as a function of the number  $N$  of the Chebyshev polynomials using a logarithmic plot when  $N$  varies within the range  $[10, 200]$ . Here, the black circle symbols are the results for  $Ma_L = 1$  and  $Ma_T = 0.2$ , the red star symbols are the results for  $Ma_L = 1$  and  $Ma_T = 0.4$ , the green diamond symbols are the results for  $Ma_L = 2$  and  $Ma_T = 0.5$ , and the blue triangle symbols are the results for  $Ma_L = 4$  and  $Ma_T = 0.5$ , respectively. The other parameter values are  $Re = 10$ ,  $k = 0.1$ ,  $C_M = C_D = 1$ ,  $C_B = C_T = 10^3$ ,  $C_K = 5$ ,  $Ca = 1$ ,  $Bi = 1$ ,  $Pr = 7$ ,  $Pe_b = Pe_L = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ .

and the perturbation film surface are in the same phase. This surfactant Marangoni stress pushes the fluid toward the trough and suppresses the surface amplitude, yielding a stabilizing influence on surface mode instability. To examine the convergence criterion for the spectrum as the number  $N$  of the Chebyshev polynomials varies, we define the relative error  $E_N$  as follows [67,68]:

$$E_N = \frac{\|C_{N+1} - C_N\|_2}{\|C_N\|_2}, \quad (104)$$

in which  $\|\cdot\|_2$  represents the  $L_2$  norm and  $C_N$  is a column vector of the ten least stable eigenvalues collected from the spectrum of the eigenvalue problem (103). The relative error  $E_N$  is computed with  $N$  numerically for different values of the thermal and surfactant Marangoni numbers when the other parameters are fixed:  $Re = 10$ ,  $k = 0.1$ ,  $C_M = C_D = 1$ ,  $C_B = C_T = 10^3$ ,  $C_K = 5$ ,  $Ca = 1$ ,  $Bi = 1$ ,  $Pr = 7$ ,  $Pe_b = Pe_L = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ . The result is displayed using a logarithmic plot when  $N$  varies within the range  $[10, 200]$ . In Fig. 3(b), we observe that the relative error  $E_N$  varies within the range  $[10^{-13}, 10^{-8}]$  when the number  $N$  of the Chebyshev polynomials increases. It appears that  $N = 20$  will be sufficient to obtain accurate numerical results for the surface mode and wall mode instabilities.

#### A. Effect of the modified thermal and surfactant Marangoni numbers on the hydrodynamic surface mode and the thermocapillary S- and P-modes

Since the current study includes a large set of nondimensional numbers, it is not feasible to analyze their individual effects separately in a single study. Moreover, the nondimensional numbers  $Ma_T$ ,  $Ma_L$ , and  $Bi$  depend not only on the thickness of the basic parallel flow but also on the physical properties of the nonisothermal surfactant-laden viscous film when the inclination angle is given. Additionally, as discussed by Goussis and Kelly [49], it is essential to rescale the nondimensional numbers for identifying the thermocapillary S-mode and P-mode. To achieve this, we introduce a set of relevant nondimensional numbers that are analogous to those employed in the studies of Goussis

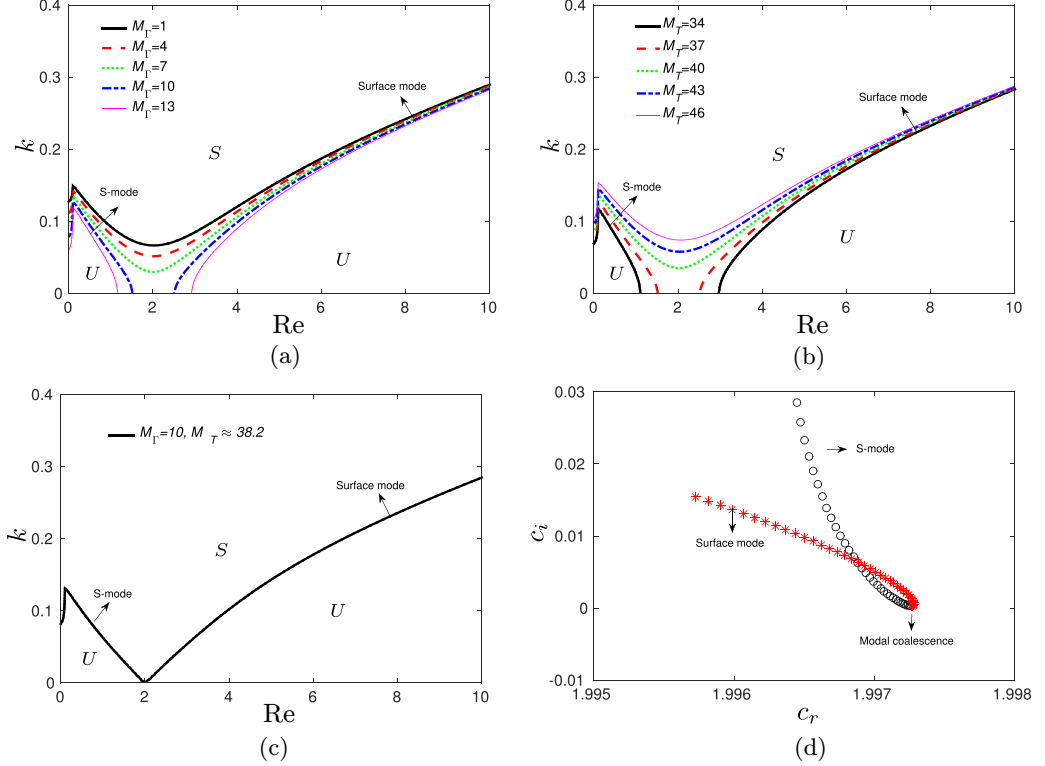


FIG. 4. (a) Variation of the neutral stability curves corresponding to the surface mode and the S-mode in the  $(\text{Re}, k)$  plane for different values of the modified surfactant Marangoni number  $M_\Gamma$  when  $M_T = 37$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $M_\Gamma = 1$ ,  $M_\Gamma = 4$ ,  $M_\Gamma = 7$ ,  $M_\Gamma = 10$ , and  $M_\Gamma = 13$ , respectively. (b) Variation of the neutral stability curves corresponding to the surface mode and the S-mode in the  $(\text{Re}, k)$  plane for different values of the modified thermal Marangoni number  $M_T$  when  $M_\Gamma = 10$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $M_T = 34$ ,  $M_T = 37$ ,  $M_T = 40$ ,  $M_T = 43$ , and  $M_T = 46$ , respectively. (c) Variation of the neutral stability curves for the surface mode and the S-mode in the  $(\text{Re}, k)$  plane when  $M_\Gamma = 10$  and  $M_T \approx 38.2$ . (d) Trajectories of the eigenvalues for the S-mode and the surface mode in the  $(c_r, c_i)$  plane when  $k = 0.033$ ,  $M_\Gamma = 10$ , and  $M_T = 40$ . Here,  $\text{Re} \in [0.5, 2.02]$  for the S-mode specified by circle symbols and  $\text{Re} \in [2.02, 4]$  for the surface mode specified by star symbols. The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 10^3$ ,  $C_T = 10^3$ ,  $C_K = 10^3$ ,  $\text{Ka} = 500$ ,  $\text{Pr} = 7$ ,  $B = 1$ ,  $\text{Pe}_b = \text{Pe}_\Gamma = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 15^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

and Kelly [49] and Trevelyan and Kalliadasis [69]:  $M_T = \text{Ma}_T(2\text{Re})^{2/3} = \frac{2E_T(T_w - T_a)}{\rho[g \sin \theta v^4]^{1/3}}$ ,  $M_\Gamma = \text{Ma}_\Gamma(2\text{Re})^{2/3} = \frac{2E_\Gamma \Gamma_\infty}{\rho[g \sin \theta v^4]^{1/3}}$ ,  $B = \text{Bi}/(2\text{Re})^{1/3} = \frac{\lambda v^{2/3}}{\kappa (g \sin \theta)^{1/3}}$ , and  $\text{Ka} = (2\text{Re})^{2/3}/\text{Ca} = \frac{2\sigma_a}{\rho[g \sin \theta v^4]^{1/3}}$ , where  $M_T$ ,  $M_\Gamma$ ,  $B$ , and  $\text{Ka}$  represent the modified thermal Marangoni number, the modified surfactant Marangoni number, the modified Biot number, and the Kapitza number, respectively.

First, we have explored how the modified surfactant Marangoni number and the modified thermal Marangoni number affect both the surface mode and the thermocapillary S-mode. As a result, we follow the work by Samanta [46] and choose the following parameter values for the numerical simulation:  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 10^3$ ,  $C_T = 10^3$ ,  $C_K = 10^3$ ,  $\text{Ka} = 500$ ,  $\text{Pr} = 7$ ,  $B = 1$ ,  $\text{Pe}_b = \text{Pe}_\Gamma = 10^3$ ,  $\xi_s = 10^2$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 15^\circ$ . In Fig. 4(a), we illustrate the variation of the neutral stability curves for the surface mode and the thermocapillary S-mode as the modified surfactant Marangoni number  $M_\Gamma$  rises.

We can see in the result that there is only one neutral stability curve that separates the unstable and stable zones in the  $(\text{Re}, k)$  plane when  $M_\Gamma = 1$  and  $M_T = 37$ . However, as soon

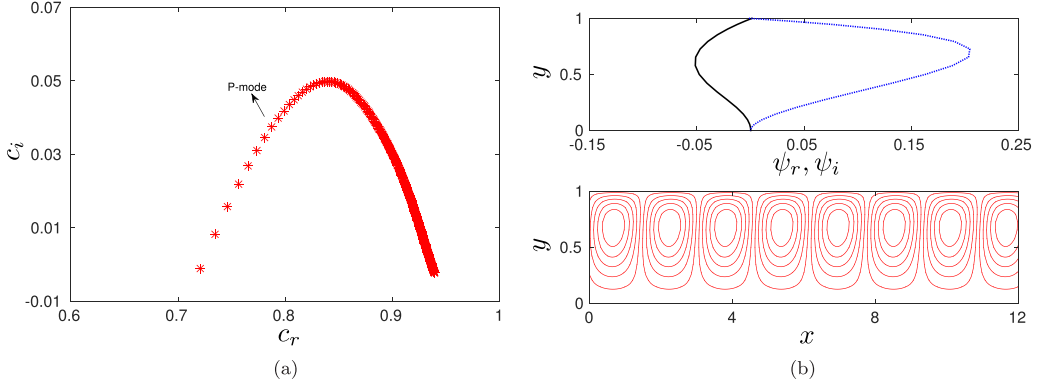


FIG. 5. (a) Trajectory of the eigenvalue for the P-mode in the  $(c_r, c_i)$  plane when  $k = 2$  and  $\text{Re} \in [1, 40]$ . (b) (top) Variation of the eigenfunction for the P-mode in the cross-stream direction  $y$  when  $\text{Re} = 15$  and  $k = 2$ . Solid and dotted lines represent the real and imaginary parts of  $\psi$ . (bottom) Contour plot of the stream function for the P-mode in the  $(x, y)$  plane when  $\text{Re} = 15$  and  $k = 2$ . The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 10^3$ ,  $C_T = 10^3$ ,  $C_K = 10^3$ ,  $\text{Ka} = 500$ ,  $M_T = 55$ ,  $\text{Pr} = 7$ ,  $B = 1$ ,  $M_\Gamma = 4$ ,  $\text{Pe}_b = \text{Pe}_T = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 15^\circ$ .

as  $M_\Gamma$  increases, the unstable zone attenuates and, ultimately, divides into two separate unstable zones bounded by two neutral stability curves, indicating a stabilizing impact of  $M_\Gamma$ . Specifically, the curve that emerged in the low Reynolds number regime is associated with the S-mode, while the curve that emerged in the low-to-moderate Reynolds number regime is associated with the surface mode. Moreover, the unstable zone generated by the surface mode appears away from that generated by the S-mode. Hence, the thermocapillary S-mode instability occurs before the hydrodynamic surface mode instability in the  $(\text{Re}, k)$  plane. In contrast, if we raise the modified thermal Marangoni number  $M_T$ , we observe a completely opposite scenario [see Fig. 4(b)]. In particular, we notice two unstable zones that are separated by two distinct neutral stability curves for the S-mode and the surface mode in the  $(\text{Re}, k)$  plane when  $M_T = 34$  and  $M_\Gamma = 10$ . However, if we further increase the value of  $M_T$ , unstable zones magnify and, suddenly, merge together and form a single unstable zone bounded by a single neutral stability curve, implying a destabilizing influence of  $M_T$  on both S-mode and surface mode instabilities. In fact, the merging process between the S-mode and the surface mode neutral stability curves takes place approximately at  $M_T \approx 38.2$  and  $M_\Gamma = 10$  [see Fig. 4(c)]. To ensure modal coalescence between the surface mode and the S-mode, we also demonstrate the trajectories of eigenvalues for the S-mode and the surface mode in the  $(c_r, c_i)$  plane [see Fig. 4(d)]. Clearly, there occurs a modal coalescence between the thermocapillary S-mode and the hydrodynamic surface mode in the long-wave regime. This outcome also strengthens the findings shown in Figs. 4(a) and 4(b), respectively. It is worth mentioning here that the hydrodynamic surface mode and the thermocapillary S-mode are recognized based on their phase speeds in the numerical simulation. They also arise in different ranges of the Reynolds number.

Second, we will unravel the thermocapillary P-mode. To this end, we compute the spectrum numerically from the eigenvalue problem (103) for the P-mode when  $M_T = 55$  and  $M_\Gamma = 4$ . Then, we demonstrate the trajectory of the eigenvalue for the P-mode in the  $(c_r, c_i)$  plane when the Reynolds number changes [see Fig. 5(a)].

The movement of the P-mode shows that it is initially placed in the stable lower half plane ( $c_i < 0$ ) at  $\text{Re} = 1$ . As the Reynolds number  $\text{Re}$  increases, the imaginary part  $c_i$  of  $c$  grows and eventually crosses the real  $c_r$  axis, entering the unstable upper half plane ( $c_i > 0$ ), where it reaches a maximum positive value. Then, it finally returns to the stable lower half plane ( $c_i < 0$ ) at higher values of  $\text{Re}$ . On the basis of this result, we can conclude that there exists a range of  $c_r$  where  $c_i$  is

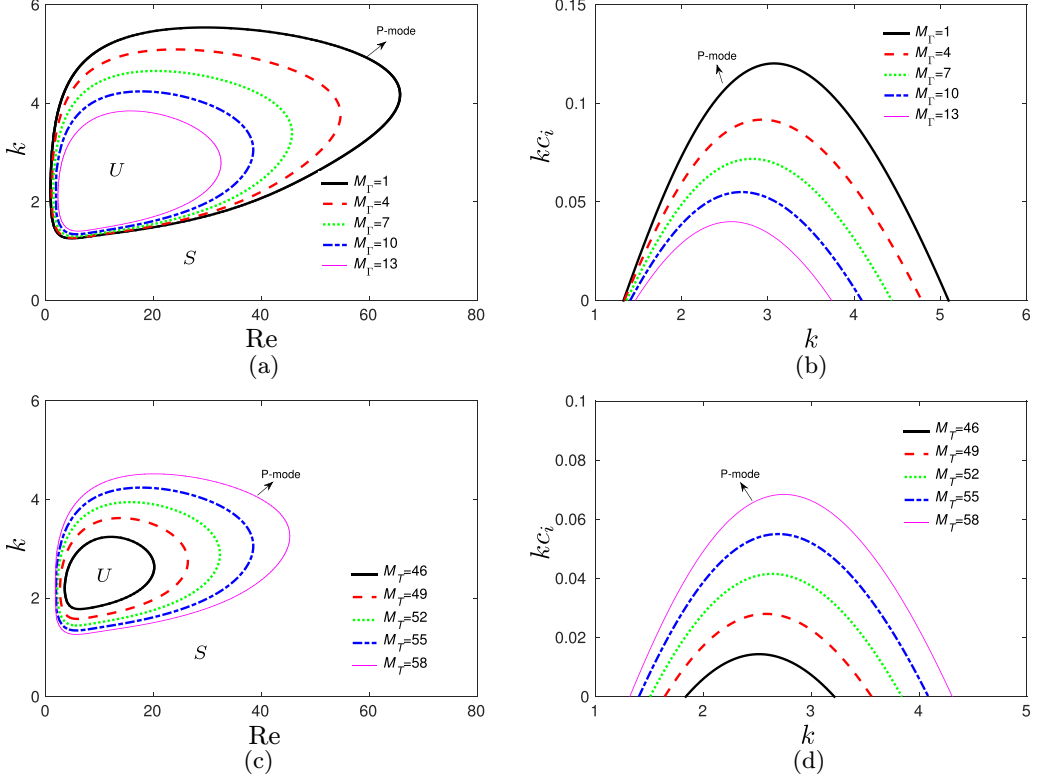


FIG. 6. (a) Variation of the neutral stability curve corresponding to the P-mode in the  $(\text{Re}, k)$  plane for different values of the modified surfactant Marangoni number  $M_F$  when  $M_T = 55$ . (b) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the P-mode for different values of  $M_F$  when  $M_T = 55$  and  $\text{Re} = 10$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $M_F = 1$ ,  $M_F = 4$ ,  $M_F = 7$ ,  $M_F = 10$ , and  $M_F = 13$ , respectively. (c) Variation of the neutral stability curve corresponding to the P-mode in the  $(\text{Re}, k)$  plane for different values of the modified thermal Marangoni number  $M_T$  when  $M_F = 10$ . (d) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the P-mode for different values of  $M_T$  when  $M_F = 10$  and  $\text{Re} = 10$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $M_T = 46$ ,  $M_T = 49$ ,  $M_T = 52$ ,  $M_T = 55$ , and  $M_T = 58$ , respectively. The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 10^3$ ,  $C_T = 10^3$ ,  $C_K = 10^3$ ,  $\text{Ka} = 500$ ,  $\text{Pr} = 7$ ,  $B = 1$ ,  $\text{Pe}_b = \text{Pe}_r = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 15^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

positive, implying the existence of an unstable P-mode. Analyzing the eigenfunction corresponding to the perturbation stream function for the unstable P-mode reveals a rapid change in the center zone [see Fig. 5(b), (top)]. As a result, the contours of the perturbation stream function induce a set of close vortices in the center zone [see Fig. 5(b), (bottom)]. Next, in Fig. 6(a), we depict how the neutral stability curve for the thermocapillary P-mode changes as the modified surfactant Marangoni number  $M_F$  rises.

In this case, the unstable zone for the thermocapillary P-mode appears in the finite wave-number regime in the  $(\text{Re}, k)$  plane, and it forms a closed-island-type shape when  $M_T = 55$ . Moreover, this unstable zone decays continuously as long as the modified surfactant Marangoni number  $M_F$  increases and, ultimately, vanishes at higher values of  $M_F$ , indicating a stabilizing influence of  $M_F$  on thermocapillary P-mode instability. To ensure this result, we also compute the temporal growth rate for the P-mode and plot it in Fig. 6(b) when  $M_F$  varies. It should be noted that the temporal growth rate curve does not pass through  $k = 0$ . Instead, it appears in the finite wave-number

regime, confirming that the thermocapillary P-mode is a finite wavelength mode. Moreover, the temporal growth rate and the associated unstable range of wave number for the P-mode gradually become weaker as  $M_F$  rises, supporting the stabilizing impact of  $M_F$  on thermocapillary P-mode instability. On the contrary, in Fig. 6(c), we observe that the unstable zone for thermocapillary P-mode instability grows continuously as long as the modified thermal Marangoni number  $M_T$  increases, indicating a destabilizing influence of  $M_T$  on thermocapillary P-mode instability. To endorse this finding, we also compute the temporal growth rate for the thermocapillary P-mode and display it in Fig. 6(d) when  $M_T$  varies. Clearly, the temporal growth rate and the associated unstable range of wave number for the P-mode become stronger gradually as  $M_T$  rises, which is in favor of the destabilizing impact of  $M_T$  on thermocapillary P-mode instability.

### B. Effect of the base surface surfactant concentration and surfactant solubility parameter on the hydrodynamic surface mode and the thermocapillary S- and P-modes

To explore the individual effect of the base surface surfactant concentration parameter  $\Gamma_E$  on the hydrodynamic surface mode and the thermocapillary S-mode and P-mode, we compute the neutral stability curves for these modes in the  $(\text{Re}, k)$  plane when  $\Gamma_E$  changes. In Fig. 7(a), we observe a single neutral stability curve at  $\Gamma_E = 0.1$ .

As the value of  $\Gamma_E$  increases, the unstable zone bounded by this single neutral stability curve decays and, suddenly, splits into two unstable zones bounded by two separate neutral stability curves: one corresponds to the S-mode, and the other corresponds to the surface mode. In fact, this splitting process into the S-mode and the surface mode neutral stability curves occurs approximately at  $\Gamma_E \approx 0.385$  [see Fig. 7(b)]. Based on the above result, we can conclude that increasing the base surface surfactant concentration parameter  $\Gamma_E$  stabilizes both instabilities created by the hydrodynamic surface mode and the thermocapillary S-mode. However, when  $\Gamma_E$  reaches a very large value, i.e., when  $\Gamma_E \geq 0.9$ , the physical scenario is completely opposite. Specifically, there are both the S-mode neutral stability curve and the surface mode neutral stability curve in the  $(\text{Re}, k)$  plane at  $\Gamma_E = 0.9$  [see Fig. 7(c)]. If  $\Gamma_E$  is increased, these neutral stability curves approach each other and merge, forming a single neutral stability curve, which further moves up with rising values of  $\Gamma_E$ . In this case, the merging process between the S-mode neutral stability curve and the surface mode neutral stability curve happens approximately at  $\Gamma_E \approx 0.909$  [see Fig. 7(d)]. Since the unstable zone magnifies, we report that increasing the base surface surfactant concentration parameter  $\Gamma_E$  destabilizes both hydrodynamic surface mode and thermocapillary S-mode instabilities when  $\Gamma_E$  takes a large value in its reasonable range of  $0 \leq \Gamma_E \leq 1$ . Hence,  $\Gamma_E$  has a dual role in hydrodynamic surface mode and thermocapillary S-mode instabilities. Interestingly, the splitting and merging processes of the neutral stability curves do not happen in a similar fashion. For instance, when the S-mode and surface mode neutral stability curves approach during the merging process, the slopes of the curves almost vanish, while there are finite slopes during the splitting process of the curve.

On the other hand, in Fig. 8(a), we display the neutral stability curve for the thermocapillary P-mode in the  $(\text{Re}, k)$  plane when  $\Gamma_E$  alters.

As shown in Fig. 6, the unstable zone for the P-mode emerges in the finite wave-number regime, and it is a closed-island-type shape. Moreover, this unstable zone reduces as  $\Gamma_E$  rises, indicating a stabilizing impact of  $\Gamma_E$  on thermocapillary P-mode instability. The result in Fig. 8(b) also confirms this finding, where the temporal growth rate is displayed for the P-mode. Obviously, the maximum temporal growth rate and the unstable range of wave number for the P-mode diminish monotonically with increasing values of  $\Gamma_E$ . However, the reduction process for the unstable region of the P-mode and the associated temporal growth rate will discontinue when  $\Gamma_E \approx 1$ . Instead, we perceive that the unstable region of the P-mode amplifies with rising values of  $\Gamma_E$  when  $\Gamma_E \geq 0.95$ , indicating a destabilizing impact of  $\Gamma_E$  on thermocapillary P-mode instability [see Fig. 8(c)]. To corroborate this unusual behavior of the thermocapillary P-mode at large values of  $\Gamma_E$ , we also compute the temporal growth rate for the P-mode and depict it in Fig. 8(d) when  $\Gamma_E$  changes. Obviously, the temporal

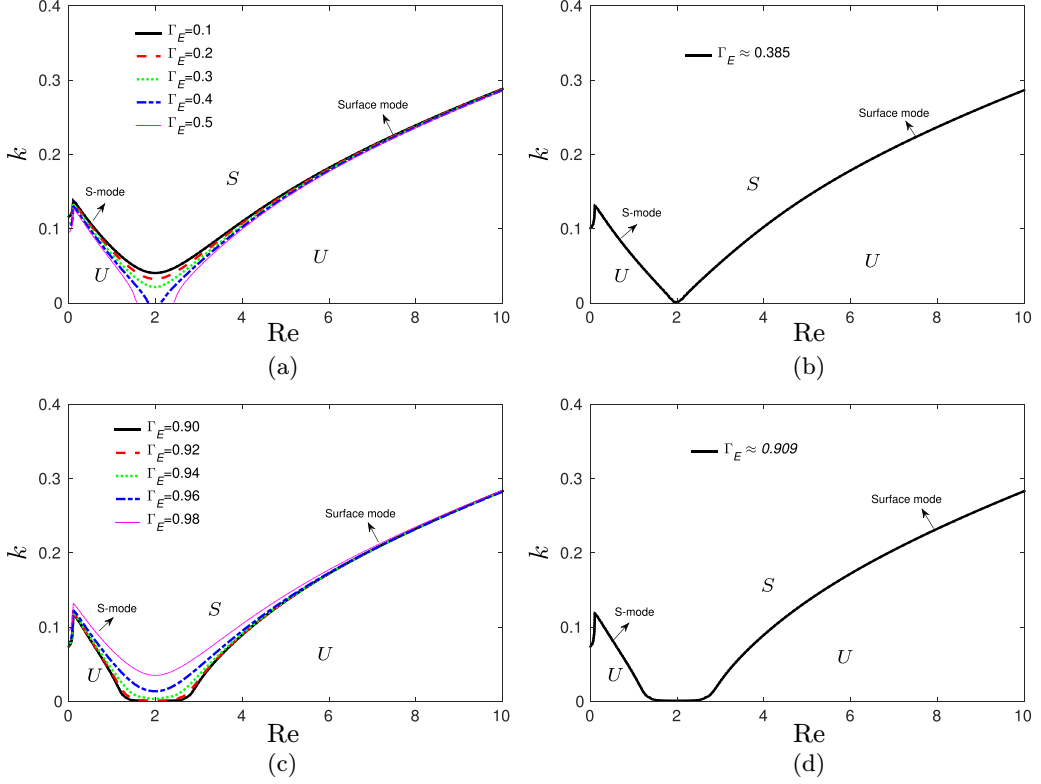


FIG. 7. (a) Variation of the neutral stability curves corresponding to the surface mode and the S-mode in the  $(\text{Re}, k)$  plane for different values of the base surface surfactant concentration parameter  $\Gamma_E$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\Gamma_E = 0.1$ ,  $\Gamma_E = 0.2$ ,  $\Gamma_E = 0.3$ ,  $\Gamma_E = 0.4$ , and  $\Gamma_E = 0.5$ , respectively. (b) Variation of the neutral stability curves for the surface mode and the S-mode in the  $(\text{Re}, k)$  plane when  $\Gamma_E \approx 0.385$ . (c) Variation of the neutral stability curves corresponding to the surface mode and the S-mode in the  $(\text{Re}, k)$  plane for different values of  $\Gamma_E$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\Gamma_E = 0.90$ ,  $\Gamma_E = 0.92$ ,  $\Gamma_E = 0.94$ ,  $\Gamma_E = 0.96$ , and  $\Gamma_E = 0.98$ , respectively. (d) Variation of the neutral stability curves for the surface mode and the S-mode in the  $(\text{Re}, k)$  plane when  $\Gamma_E \approx 0.909$ . The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 10^3$ ,  $C_T = 10^3$ ,  $C_K = 10^3$ ,  $\text{Ka} = 500$ ,  $\text{Pr} = 7$ ,  $B = 1$ ,  $M_T = 33$ ,  $\text{Pe}_b = \text{Pe}_T = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $M_T = 1$ , and  $\theta = 15^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

growth rate and the associated unstable range of wave number grow monotonically for the P-mode as  $\Gamma_E$  increases ( $\Gamma_E \geq 0.95$ ). This result fully supports the finding shown in Fig. 8(c). Therefore, we can conclude that the base surface surfactant concentration parameter  $\Gamma_E$  has a stabilizing influence on primary instabilities induced by the hydrodynamic surface mode and the thermocapillary S-mode and P-mode when it keeps low values. However,  $\Gamma_E$  destabilizes all surface mode, S-mode, and P-mode instabilities when it keeps very large values ( $\Gamma_E \approx 1$ ) within its reasonable range of  $0 \leq \Gamma_E \leq 1$ . The dual role of the base surface surfactant concentration parameter  $\Gamma_E$  in the primary instabilities can be explained in the spirit of Georgantaki *et al.* [70]. If a surfactant is added, a surfactant concentration gradient is formed along the deformed film surface due to in-phase distribution of surfactant concentration. This distribution creates a surface elasticity and induces a surfactant Marangoni stress at the film surface. As the base surface surfactant concentration parameter  $\Gamma_E$  increases, the surface elasticity, or equivalently, the surfactant Marangoni stress, rises. This fact causes an additional energy loss to the perturbation compared to the clean film

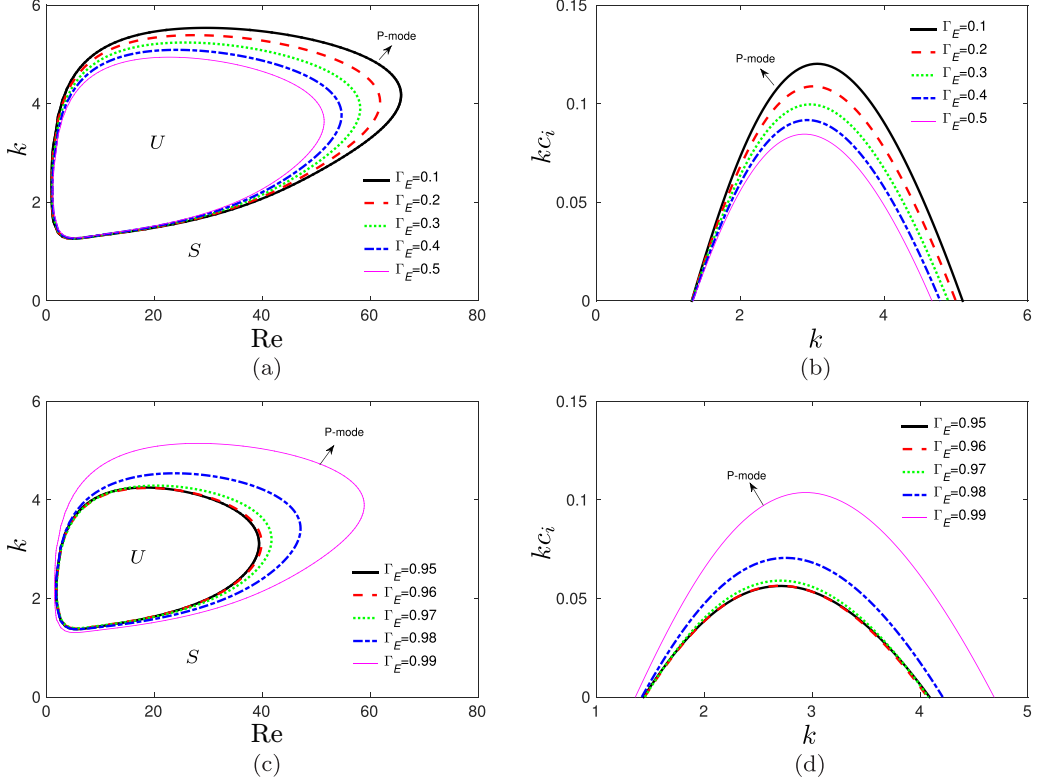


FIG. 8. (a) Variation of the neutral stability curve corresponding to the P-mode in the  $(Re, k)$  plane for different values of the base surface surfactant concentration parameter  $\Gamma_E$ . (b) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the P-mode for different values of  $\Gamma_E$  when  $Re = 10$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\Gamma_E = 0.1$ ,  $\Gamma_E = 0.2$ ,  $\Gamma_E = 0.3$ ,  $\Gamma_E = 0.4$ , and  $\Gamma_E = 0.5$ , respectively. (c) Variation of the neutral stability curve corresponding to the P-mode in the  $(Re, k)$  plane for different values of  $\Gamma_E$ . (d) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the P-mode for different values of  $\Gamma_E$  when  $Re = 10$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\Gamma_E = 0.95$ ,  $\Gamma_E = 0.96$ ,  $\Gamma_E = 0.97$ ,  $\Gamma_E = 0.98$ , and  $\Gamma_E = 0.99$ , respectively. The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 10^3$ ,  $C_T = 10^3$ ,  $C_K = 10^3$ ,  $Ka = 500$ ,  $Pr = 7$ ,  $B = 1$ ,  $M_T = 55$ ,  $Pe_b = Pe_\Gamma = 10^3$ ,  $\xi_s = 100$ ,  $k_s = 1$ ,  $M_\Gamma = 1$ , and  $\theta = 15^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

flow case and stabilizes primary instabilities. Since the current study deals with a soluble surfactant, the bulk surfactant concentration gradient is also formed in the vicinity of the film surface due to local equilibrium, and it intensifies with the rising values of  $\Gamma_E$ . As discussed by Karapetsas and Bontozoglou [71], increasing the bulk surfactant concentration gradient not only strengthens the mass exchange between bulk and surface but also mitigates the surface surfactant concentration gradient. This physical fact weakens the stabilizing influence of the surfactant Marangoni stress, which is the consequence of the destabilizing influence of  $\Gamma_E$  when it keeps large values close to  $\Gamma_E \approx 1$ .

To decipher how the surfactant solubility parameter  $\xi_s$  affects primary instabilities induced by the surface mode, S-mode, and P-mode, we present the neutral stability curves for these modes in Figs. 9(a) and 9(b), respectively, when the surfactant solubility parameter  $\xi_s$  varies.

In the numerical computation, we obtain only one neutral stability curve for the hydrodynamic surface mode and the thermocapillary S-mode at  $\xi_s = 1$  [see Fig. 9(a)]. As the parameter  $\xi_s$  rises, the neutral stability curve moves down gradually, which is followed by the successive reduction of

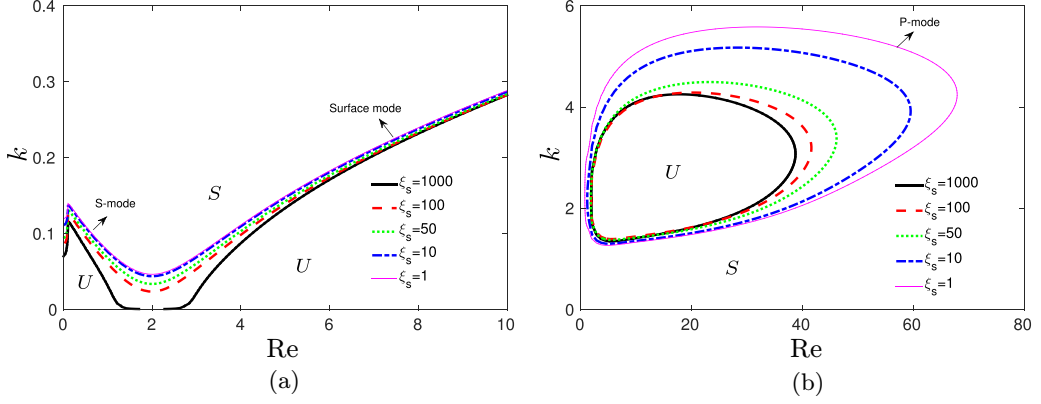


FIG. 9. (a) Variation of the neutral stability curves corresponding to the surface mode and the S-mode in the  $(Re, k)$  plane for different values of the surfactant solubility parameter  $\xi_s$  when  $M_T = 33$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\xi_s = 1000$ ,  $\xi_s = 100$ ,  $\xi_s = 50$ ,  $\xi_s = 10$ , and  $\xi_s = 1$ , respectively. (b) Variation of the neutral stability curve corresponding to the P-mode in the  $(Re, k)$  plane for different values of  $\xi_s$  when  $M_T = 55$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\xi_s = 1000$ ,  $\xi_s = 100$ ,  $\xi_s = 50$ ,  $\xi_s = 10$ , and  $\xi_s = 1$ , respectively. The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 10^3$ ,  $C_T = 10^3$ ,  $C_K = 10^3$ ,  $Ka = 500$ ,  $Pr = 7$ ,  $B = 1$ ,  $Pe_b = Pe_\Gamma = 10^3$ ,  $\Gamma_E = 0.97$ ,  $k_s = 1$ ,  $M_\Gamma = 1$ , and  $\theta = 15^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

the unstable zone, indicating a stabilizing impact of  $\xi_s$ . Moreover, when  $\xi_s$  keeps a high value, the single neutral stability curve is suddenly divided into two neutral stability curves for the thermocapillary S-mode and the hydrodynamic surface mode, respectively. Specifically, the neutral stability curve that appeared in the low Reynolds number zone corresponds to the thermocapillary S-mode, while the other neutral stability curve that appeared in the low-to-moderate Reynolds number zone corresponds to the hydrodynamic surface mode. On the other hand, in Fig. 9(b), we always find a single neutral stability curve for the thermocapillary P-mode, which induces a closed-island-type-shaped unstable region. Additionally, the unstable region for the P-mode progressively diminishes as the value of  $\xi_s$  increases, implying that  $\xi_s$  has a stabilizing effect on thermocapillary P-mode instability. Physically, as the surfactant solubility parameter  $\xi_s$  rises, the current flow problem gradually converts into the insoluble-surfactant-impured film flowing over a flexible substrate, which is more stable than the soluble-surfactant-laden film flow. This is why increasing  $\xi_s$  has a stabilizing impact on primary instability.

### C. Modal coalescence between the P-mode and wall mode

To examine the modal coalescence between the thermocapillary P-mode and the wall mode, we choose the following parameter values:  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 30$ ,  $Ka = 500$ ,  $Pr = 7$ ,  $B = 1$ ,  $Pe_b = Pe_\Gamma = 10^3$ ,  $\Gamma_E = 0.1$ ,  $k_s = 1$ ,  $\xi_s = 100$ , and  $\theta = 15^\circ$ . In Fig. 10(a), we present the variation of the neutral stability curves in the  $(Re, k)$  plane for several values of the modified surfactant Marangoni number  $M_\Gamma$  when  $M_T = 50$ .

When  $M_\Gamma > 1$ , we notice two neutral stability curves: one corresponds to the P-mode, while the other corresponds to the wall mode. In fact, both the neutral stability curves emerge in the finite wave-number regime but in different ranges of the Reynolds number. Specifically, when  $M_\Gamma = 1$ , the P-mode and the wall mode neutral stability curves merge together. However, as soon as  $M_\Gamma$  increases, the previously combined neutral stability curve separates into two neutral stability curves for the P-mode and the wall mode, respectively. The separation process between the neutral stability curves takes place approximately at  $M_\Gamma \approx 1.17$  and  $M_T = 50$ , as shown in Fig. 10(c). Moreover, unstable zones bounded by the neutral stability curves for the P-mode and the wall mode

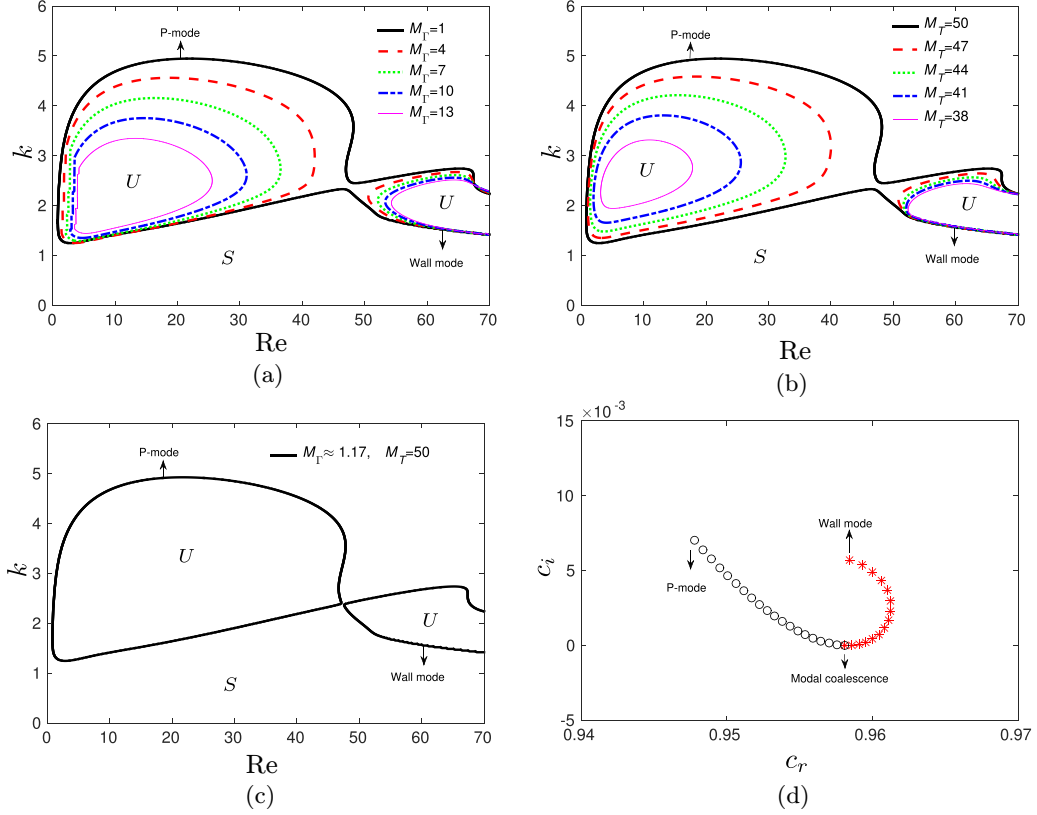


FIG. 10. (a) Variation of the neutral stability curves corresponding to the P-mode and the wall mode in the  $(Re, k)$  plane for different values of  $M_F$  when  $M_T = 50$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $M_F = 1$ ,  $M_F = 4$ ,  $M_F = 7$ ,  $M_F = 10$ , and  $M_F = 13$ , respectively. (b) Variation of the neutral stability curves corresponding to the P-mode and the wall mode in the  $(Re, k)$  plane for different values of  $M_T$  when  $M_F = 1$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $M_T = 50$ ,  $M_T = 47$ ,  $M_T = 44$ ,  $M_T = 41$ , and  $M_T = 38$ , respectively. (c) Variation of the neutral stability curves for the P-mode and the wall mode in the  $(Re, k)$  plane when  $M_F \approx 1.17$  and  $M_T = 50$ . (d) Trajectories of the eigenvalues for the P-mode and the wall mode in the  $(c_r, c_i)$  plane when  $k = 2.455$ ,  $M_F = 1$ , and  $M_T = 50$ . Here,  $Re \in [30, 48.38]$  for the P-mode specified by circle symbols and  $Re \in [48.38, 60]$  for the wall mode specified by star symbols. The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 30$ ,  $Ka = 500$ ,  $Pr = 7$ ,  $B = 1$ ,  $Pe_b = Pe_f = 10^3$ ,  $\Gamma_E = 0.1$ ,  $k_s = 1$ ,  $\xi_s = 100$ , and  $\theta = 15^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

shrink with increasing values of  $M_F$ , indicating a stabilizing effect of  $M_F$  on both P-mode and wall mode instabilities. Additionally, we observe that P-mode instability occurs before wall mode instability because the critical Reynolds number for the onset of wall mode instability lies far away from that of P-mode instability. In contrast, if we investigate the effect of the modified thermal Marangoni number  $M_T$ , we see the opposite behavior. That is, there exist distinct neutral stability curves for the P-mode and the wall mode in the  $(Re, k)$  plane when  $M_T = 38$  and  $M_F = 1$ . But if  $M_T$  is raised, the neutral stability curves for the P-mode and the wall mode approach each other and ultimately coalesce for higher values of  $M_T$ , forming a single neutral stability curve [see Fig. 10(b)]. Moreover, unstable zones for P-mode and wall mode instabilities magnify as  $M_T$  rises, indicating a destabilizing effect of  $M_T$  on primary instabilities caused by the P-mode and the wall mode. The trajectories of eigenvalues in Fig. 10(d) further endorse the modal coalescence between the P-mode and the wall mode.

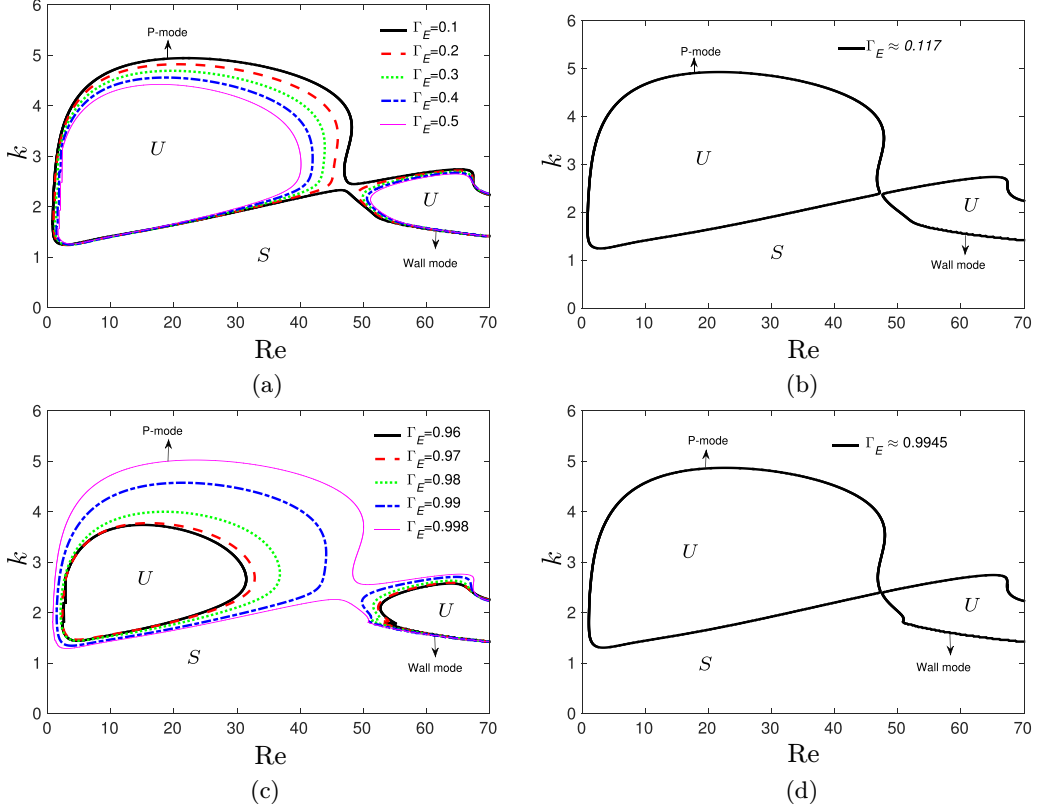


FIG. 11. (a) Variation of the neutral stability curves corresponding to the P-mode and the wall mode in the  $(\text{Re}, k)$  plane for different values of  $\Gamma_E$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\Gamma_E = 0.1$ ,  $\Gamma_E = 0.2$ ,  $\Gamma_E = 0.3$ ,  $\Gamma_E = 0.4$ , and  $\Gamma_E = 0.5$ , respectively. (b) Variation of the neutral stability curves for the P-mode and the wall mode in the  $(\text{Re}, k)$  plane when  $\Gamma_E \approx 0.117$ . (c) Variation of the neutral stability curves corresponding to the P-mode and the wall mode in the  $(\text{Re}, k)$  plane for different values of  $\Gamma_E$ . Solid, dashed, dotted, dash-dotted, and thin lines stand for  $\Gamma_E = 0.96$ ,  $\Gamma_E = 0.97$ ,  $\Gamma_E = 0.98$ ,  $\Gamma_E = 0.99$ , and  $\Gamma_E = 0.998$ , respectively. (d) Variation of the neutral stability curves for the P-mode and the wall mode in the  $(\text{Re}, k)$  plane when  $\Gamma_E \approx 0.9945$ . The other parameter values are  $C_M = 1$ ,  $C_D = 1$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 30$ ,  $\text{Ka} = 500$ ,  $\text{Pr} = 7$ ,  $B = 1$ ,  $M_T = 50$ ,  $\text{Pe}_b = \text{Pe}_r = 10^3$ ,  $k_s = 1$ ,  $\xi_s = 100$ ,  $M_\Gamma = 1$ , and  $\theta = 15^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

To investigate how the base surface surfactant concentration parameter  $\Gamma_E$  affects the modal coalescence between the P-mode and the wall mode, we alter the parameter  $\Gamma_E$  in the numerical simulation while the other parameters remain constant. In Fig. 11(a), the result reveals that the combined neutral stability curve for the P-mode and the wall mode, which appeared at  $\Gamma_E = 0.1$ , splits into two distinct neutral stability curves for the P-mode and the wall mode when the parameter  $\Gamma_E$  is increased.

Indeed, the splitting process between the P-mode and the wall mode neutral stability curves happens approximately at  $\Gamma_E \approx 0.117$  [see Fig. 11(b)]. Moreover, increasing  $\Gamma_E$  reduces unstable domains for P-mode and wall mode instabilities in the  $(\text{Re}, k)$  plane. However, this reduction process no longer continues at the higher values of  $\Gamma_E$ . In fact, we notice an opposite scenario in Fig. 11(c). That is, unstable domains for P-mode and wall mode instabilities grow for higher values of  $\Gamma_E (\geq 0.96)$ . Interestingly, we also discover that the separated unstable domains for the P-mode and the wall mode further merge together and create a single combined unstable domain. In

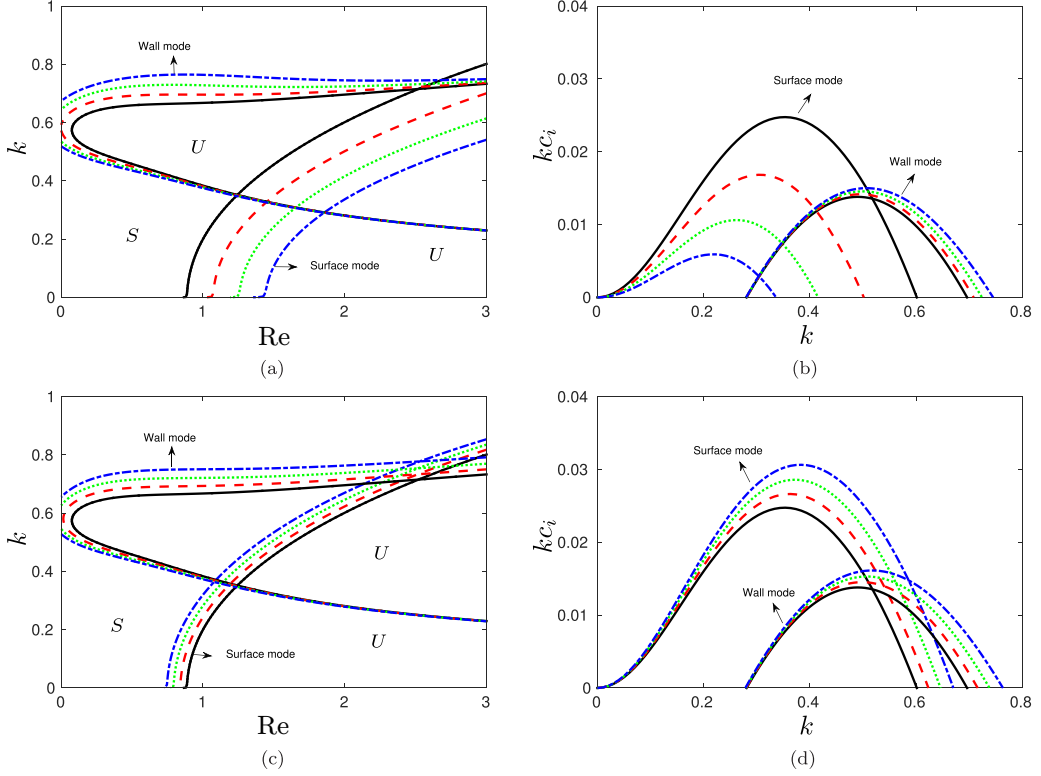


FIG. 12. (a) Variation of the neutral stability curves corresponding to the surface mode and the wall mode in the  $(\text{Re}, k)$  plane for different values of the surfactant Marangoni number  $\text{Ma}_\Gamma$  when  $\text{Ma}_T = 0.2$ . (b) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the surface mode and the wall mode for different values of  $\text{Ma}_\Gamma$  when  $\text{Ma}_T = 0.2$  and  $\text{Re} = 2$ . Solid, dashed, dotted, and dash-dotted lines stand for  $\text{Ma}_\Gamma = 1$ ,  $\text{Ma}_\Gamma = 2$ ,  $\text{Ma}_\Gamma = 3$ , and  $\text{Ma}_\Gamma = 4$ , respectively. (c) Variation of the neutral stability curves corresponding to the surface mode and the wall mode in the  $(\text{Re}, k)$  plane for different values of the thermal Marangoni number  $\text{Ma}_T$  when  $\text{Ma}_\Gamma = 1$ . (d) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the surface mode and the wall mode for different values of  $\text{Ma}_T$  when  $\text{Ma}_\Gamma = 1$  and  $\text{Re} = 2$ . Solid, dashed, dotted, and dash-dotted lines stand for  $\text{Ma}_T = 0.2$ ,  $\text{Ma}_T = 0.4$ ,  $\text{Ma}_T = 0.6$ , and  $\text{Ma}_T = 0.8$ , respectively. The other parameter values are  $C_M = 1$ ,  $C_D = 0.32$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 5$ ,  $\text{Ca} = 1$ ,  $\text{Pr} = 7$ ,  $\text{Bi} = 1$ ,  $\text{Pe}_b = \text{Pe}_\Gamma = 10^3$ ,  $\xi_s = 10$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

this case, the merging process between the P-mode and the wall mode neutral stability curves occurs approximately at  $\Gamma_E \approx 0.9945$ , as depicted in Fig. 11(d). Additionally, we notice that the impact of  $\Gamma_E$  is stronger when it keeps value in the vicinity of  $\Gamma_E \approx 1$ .

#### D. Competition between the surface mode and the wall mode

In this subsection, we discuss the competition between the surface mode and the wall mode when the surfactant Marangoni number  $\text{Ma}_\Gamma$  and the thermal Marangoni number  $\text{Ma}_T$  vary. To this end, we choose the following parameter values:  $C_M = 1$ ,  $C_D = 0.32$ ,  $C_B = 1$ ,  $C_K = 5$ ,  $C_T = 1$ ,  $\text{Ca} = 1$ ,  $\text{Pr} = 7$ ,  $\text{Bi} = 1$ ,  $\text{Pe}_b = \text{Pe}_\Gamma = 10^3$ ,  $\xi_s = 10$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ . In Fig. 12(a), we illustrate the neutral stability curves for the surface mode and the wall mode when the surfactant Marangoni number  $\text{Ma}_\Gamma$  alters but  $\text{Ma}_T = 0.2$ .

It is important to note that surface mode instability emerges at  $k = 0$  when the Reynolds number exceeds the critical value for its initiation at  $\text{Ma}_F = 1$ . However, the critical Reynolds number for the onset of wall mode instability emerges in the finite wave-number regime when  $\text{Ma}_F = 1$ . As long as  $\text{Ma}_F$  rises, the critical Reynolds number for surface mode instability appearing on the Re axis shifts toward the right, which is followed by the successive reduction of the unstable domain of the surface mode in the  $(\text{Re}, k)$  plane. Hence, the surfactant Marangoni number  $\text{Ma}_F$  stabilizes surface mode instability. On the contrary, the critical Reynolds number for wall mode instability appearing in the finite wave-number regime shifts toward the left, which is followed by the successive amplification of the unstable domain of the wall mode in the  $(\text{Re}, k)$  plane with increasing values of  $\text{Ma}_F$ . Hence,  $\text{Ma}_F$  destabilizes wall mode instability. Moreover, we observe that the neutral stability curve for wall mode instability touches the  $k$  axis at higher values of  $\text{Ma}_F$ , indicating the existence of finite wavelength inertialess instability generated by the wall mode (for details, see the Supplemental Material [63]). Additionally, we see that the long-wave surface mode instability occurs after the finite wavelength wall mode instability when  $C_D = 0.32$ , as the critical Reynolds number for surface mode instability is greater than that of wall mode instability. To ensure the above findings at  $C_D = 0.32$ , we further demonstrate the temporal growth rate for the surface mode and the wall mode in Fig. 12(b) when  $\text{Ma}_F$  changes but  $\text{Ma}_T = 0.2$  and  $\text{Re} = 2$ . It is clear that the temporal growth rate emerging in the small-to-finite wave-number regime attenuates for surface mode instability, but the temporal growth rate emerging in the finite wave-number regime intensifies for wall mode instability with rising values of  $\text{Ma}_F$ , confirming the outcomes shown in Fig. 12(a). Interestingly, the maximum temporal growth rate of surface mode instability is larger than that of wall mode instability when  $\text{Ma}_F = 1$  and  $\text{Re} = 2$ . However, as  $\text{Ma}_F$  increases, the situation reverses because the temporal growth rate for the wall mode intensifies. That is, the maximum temporal growth rate of wall mode instability overtakes the maximum temporal growth rate of surface mode instability. Since the surface mode is a long-wave mode, the temporal growth rate for the surface mode passes through  $k = 0$ . However, the temporal growth rate for the wall mode does not pass through  $k = 0$ , as it is a finite wavelength mode.

On the other hand, analyzing the effect of the thermal Marangoni number  $\text{Ma}_T$  reveals that surface mode instability behaves oppositely, while wall mode instability behaves the same as the result of  $\text{Ma}_F$ . Specifically, unstable domains bounded by the neutral stability curves and temporal growth rates for both the surface mode and the wall mode amplify as  $\text{Ma}_T$  rises when  $\text{Ma}_F = 1$  [see Fig. 12(c)]. Hence,  $\text{Ma}_T$  destabilizes both surface mode and wall mode instabilities. Although  $\text{Ma}_T$  makes the long-wave surface mode instability stronger, wall mode instability always initiates first in the finite wave-number regime because its critical Reynolds number is lower than that of surface mode instability. Albeit wall mode instability occurs first, its temporal growth rate is weaker than that of surface mode instability with rising values of  $\text{Ma}_T$  when  $C_D = 0.32$  [see Fig. 12(d)].

If the value of the damping coefficient  $C_D$  is slightly increased to  $C_D = 0.35$  in the numerical computation, we find that the critical Reynolds number for the onset of the long-wave surface mode instability arises on the Re axis earlier than that of the finite wavelength wall mode instability when  $\text{Ma}_F = 1$  [see Fig. 13(a)]. This fact indicates that the long-wave surface mode instability occurs before the finite wavelength wall mode instability.

However, as  $\text{Ma}_F$  increases, the strength of surface mode instability decreases, while wall mode instability grows stronger. As a result, the critical Reynolds number for the surface mode is higher than that for the wall mode. Hence, the finite wavelength wall mode instability outweighs the long-wave surface mode instability with the increase in the value of  $\text{Ma}_F$ . To support this outcome, we have also plotted the temporal growth rates in Fig. 13(b) for both the surface mode and the wall mode. Clearly, the maximum temporal growth rate for surface mode instability is larger than that of wall mode instability when  $\text{Ma}_F = 1$ . But this physical phenomenon no longer holds when  $\text{Ma}_F$  is at a high value. Indeed, the maximum temporal growth rate for surface mode instability is smaller than that of wall mode instability when  $\text{Ma}_F = 4$  [see Fig. 13(b)].

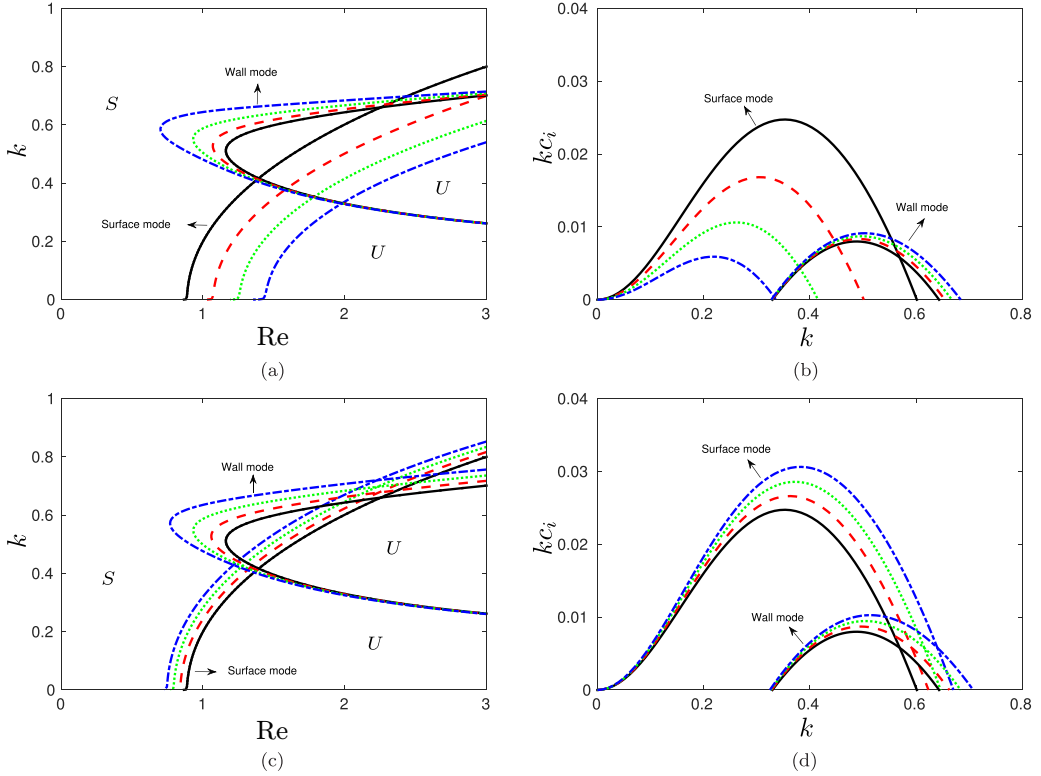


FIG. 13. (a) Variation of the neutral stability curves corresponding to the surface mode and the wall mode in the  $(Re, k)$  plane for different values of the surfactant Marangoni number  $Ma_F$  when  $Ma_T = 0.2$ . (b) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the surface mode and the wall mode for different values of  $Ma_F$  when  $Ma_T = 0.2$  and  $Re = 2$ . Solid, dashed, dotted, and dash-dotted lines stand for  $Ma_F = 1$ ,  $Ma_F = 2$ ,  $Ma_F = 3$ , and  $Ma_F = 4$ , respectively. (c) Variation of the neutral stability curves corresponding to the surface mode and the wall mode in the  $(Re, k)$  plane for different values of the thermal Marangoni number  $Ma_T$  when  $Ma_F = 1$ . (d) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  corresponding to the surface mode and the wall mode for different values of  $Ma_T$  when  $Ma_F = 1$  and  $Re = 2$ . Solid, dashed, dotted, and dash-dotted lines stand for  $Ma_T = 0.2$ ,  $Ma_T = 0.4$ ,  $Ma_T = 0.6$ , and  $Ma_T = 0.8$ , respectively. The other parameter values are  $C_M = 1$ ,  $C_D = 0.35$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 5$ ,  $Ca = 1$ ,  $Pr = 7$ ,  $Bi = 1$ ,  $Pe_b = Pe_F = 10^3$ ,  $\xi_s = 10$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 45^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

Next, we examine the impact of the thermal Marangoni number  $Ma_T$  when  $C_D = 0.35$ . In this case, we notice that unstable domains for both surface mode and wall mode instabilities enhance successively with rising values of  $Ma_T$ , resulting in a destabilizing influence of  $Ma_T$  on both surface mode and wall mode instabilities [see Fig. 13(c)]. But the effect of  $Ma_T$  on wall mode instability is more significant than that of surface mode instability. As a result, even though the surface mode dominates primary instability in the long-wave regime initially at  $Ma_T = 0.2$ , the wall mode takes over primary instability in the finite wavelength regime with the increase in the value of  $Ma_T$ . However, the temporal growth rate for the long-wave surface mode instability is sufficiently greater than that of the finite wavelength wall mode instability when  $Ma_T$  alters [see Fig. 13(d)].

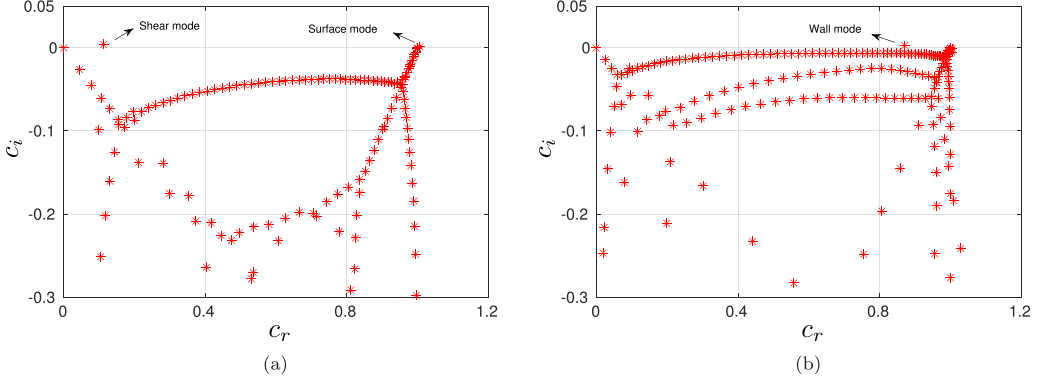


FIG. 14. Spectrum of the eigenvalue problem (103) when  $\text{Re} = 10^5$ . (a)  $k = 0.5$  and (b)  $k = 3$ . The other parameter values are  $C_M = 1$ ,  $C_D = 10$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 10^5$ ,  $\text{Ca} = 1$ ,  $\text{Ma}_T = 1$ ,  $\text{Pr} = 7$ ,  $\text{Bi} = 1$ ,  $\text{Ma}_F = 1$ ,  $\text{Pe}_b = \text{Pe}_F = 10^3$ ,  $\xi_s = 10^3$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 4^\circ$ .

## V. SHEAR MODE INSTABILITY ANALYSIS IN THE HIGH REYNOLDS NUMBER ZONE

In this section, we focus on the investigation of shear mode instability for a soluble-surfactant-laden nonisothermal film falling down a uniformly heated flexible substrate. Specifically, shear mode instability for a surfactant-free isothermal falling film over a nonflexible substrate emerges in the finite wave-number regime when the Reynolds number is sufficiently large and the inclination angle is small enough [30,31]. However, the study of Alexander *et al.* [39] was the first to report the existence of shear mode instability in the case of an isothermal falling film over a flexible substrate. That is why we adhere to the research work of Alexander *et al.* [39] and select the parameter values as suggested in their numerical simulations, such as  $C_M = 1$ ,  $C_K = 10^5$ ,  $C_D = 10$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $\text{Ca} = 1$ ,  $\text{Ma}_T = 1$ ,  $\text{Pr} = 7$ ,  $\text{Bi} = 1$ ,  $\text{Ma}_F = 1$ ,  $\text{Pe}_b = \text{Pe}_F = 10^3$ ,  $\xi_s = 10^3$ ,  $\Gamma_E = 0.1$ ,  $k_s = 1$ , and  $\theta = 4^\circ$ . In Fig. 14(a), we display the spectrum of the eigenvalue problem (103) when  $\text{Re} = 10^5$  and  $k = 0.5$ .

It should be noted that two dominant unstable modes arise in the  $(c_r, c_i)$  plane when  $k = 0.5$ . One mode corresponds to the shear mode, and the other mode corresponds to the surface mode. In fact, they are recognized by their phase speeds, as the surface mode has a higher phase speed than that of the shear mode. Clearly, the shear mode is placed at  $(c_r = 0.1131, c_i = 0.003\,907\,7)$ , while the surface mode is placed at  $(c_r = 1.0056, c_i = 0.001\,502\,1)$ . It appears that the phase speed of the shear mode is lower than that of the surface mode, but its temporal growth rate ( $\propto c_i$ ) is greater than that of the surface mode. Hence, at  $k = 0.5$ , the shear mode, although slower than the surface mode, is the most unstable mode and dominates primary instability. To figure out the characteristics of both the unstable shear mode and the unstable surface mode in the high Reynolds number regime, we compute their eigenfunctions and the contours of the associated perturbation stream functions. In Fig. 15(a), we illustrate the eigenfunction corresponding to the perturbation stream function for the shear mode.

It should be noted that the numerical outcome shows a substantial change in the real part of the eigenfunction in the vicinity of the center of the film layer. This fact results in the induction of large open vortices in the contours of the stream function near the center of the film layer, as depicted in Fig. 15(b). On the other hand, the result for the surface mode in Fig. 15(c) shows a significant change in the real part of the eigenfunction close to the film surface. This physical fact generates open vortices in the contours of the stream function for the surface mode near the film surface rather than the center of the film layer [see Fig. 15(d)]. Moreover, the induced open vortices for the surface mode are not as big as observed for the shear mode.

If we increase the wave number to  $k = 3$  while keeping the other parameters fixed and compute the spectrum, we observe a new unstable mode, the so-called wall mode, that arises in the

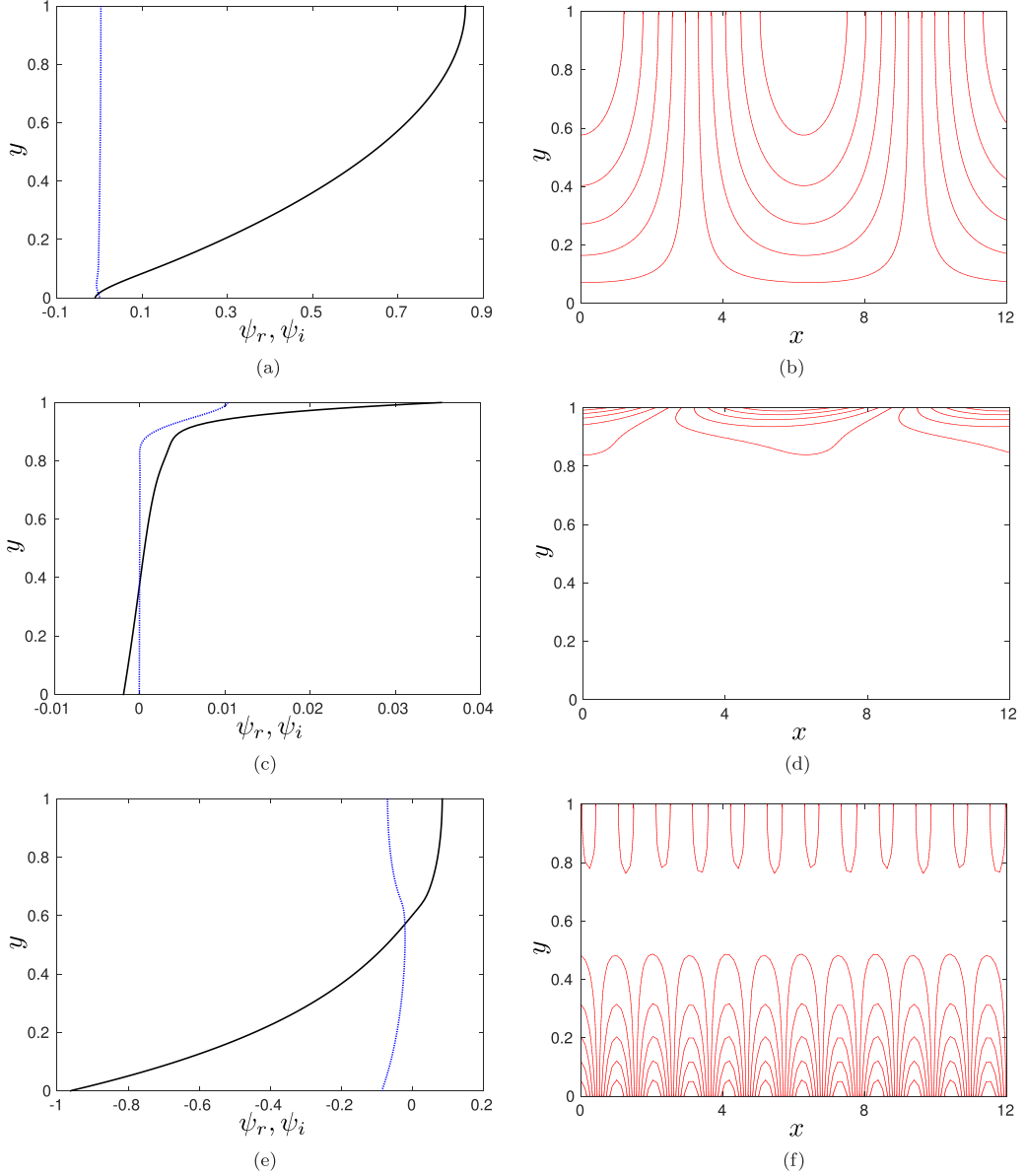


FIG. 15. (a) Eigenfunction for the shear mode varies in the  $y$  direction when  $k = 0.5$ . (b) Contour plot of the stream function for the shear mode in the  $(x, y)$  plane when  $k = 0.5$ . (c) Eigenfunction for the surface mode varies in the  $y$  direction when  $k = 0.5$ . (d) Contour plot of the stream function for the surface mode in the  $(x, y)$  plane when  $k = 0.5$ . (e) Eigenfunction for the wall mode varies in the  $y$  direction when  $k = 3$ . (f) Contour plot of the stream function for the wall mode in the  $(x, y)$  plane when  $k = 3$ . Solid and dotted lines, respectively, represent the real and imaginary parts of  $\psi$ . The other parameter values are  $\text{Re} = 10^5$ ,  $C_M = 1$ ,  $C_D = 10$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 10^5$ ,  $\text{Ca} = 1$ ,  $\text{Ma}_T = 1$ ,  $\text{Pr} = 7$ ,  $\text{Bi} = 1$ ,  $\text{Ma}_\Gamma = 1$ ,  $\text{Pe}_b = \text{Pe}_\Gamma = 10^3$ ,  $\xi_s = 10^3$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 4^\circ$ .

$(c_r, c_i)$  plane [see Fig. 14(b)]. In particular, the unstable wall mode is placed at  $(c_r = 0.8707, c_i = 0.0027699)$ . Since the wall mode is the only unstable mode at  $k = 3$ , it dominates primary instability in this case. To understand the characteristics of the unstable wall mode in the high

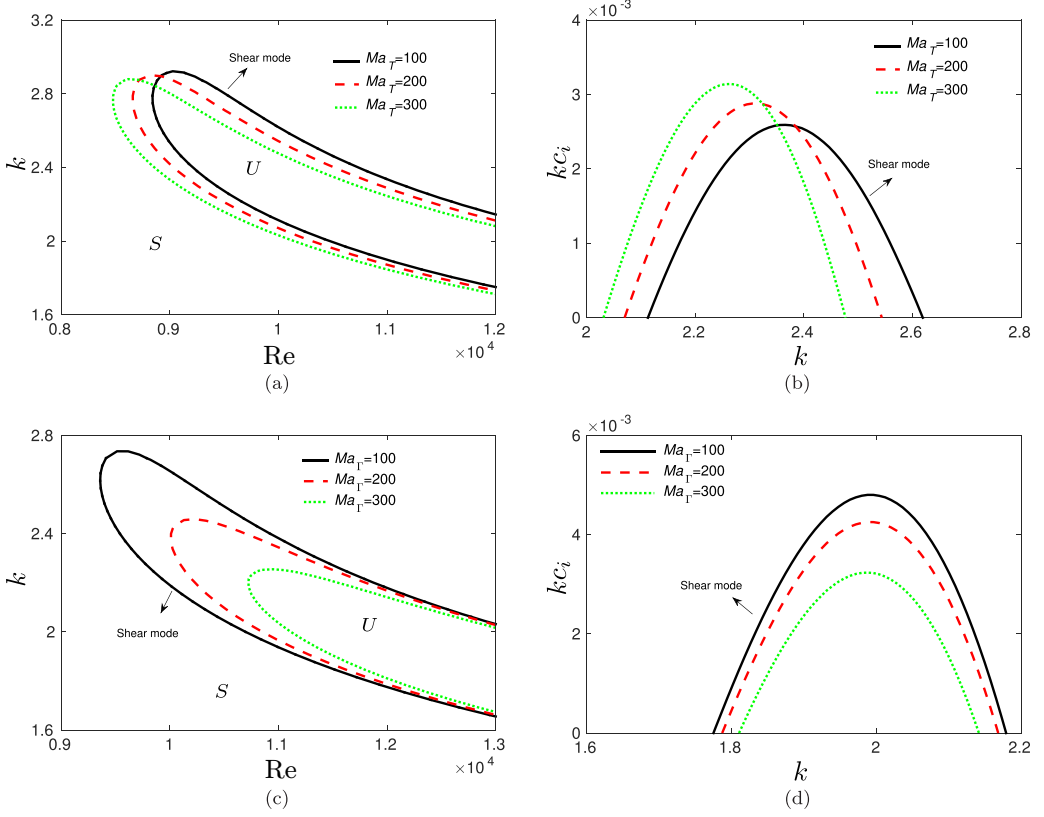


FIG. 16. (a) Variation of the neutral stability curves corresponding to the shear mode in the  $(\text{Re}, k)$  plane for different values of  $\text{Ma}_T$  when  $\text{Ma}_\Gamma = 1$ . (b) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  for the shear mode when  $\text{Re} = 10000$ . Solid, dashed, and dotted lines stand for  $\text{Ma}_T = 100$ ,  $\text{Ma}_T = 200$ , and  $\text{Ma}_T = 300$ , respectively. (c) Variation of the neutral stability curves corresponding to the shear mode in the  $(\text{Re}, k)$  plane for different values of  $\text{Ma}_T$  when  $\text{Ma}_\Gamma = 1$ . (d) Variation of the temporal growth rate  $kc_i$  with wave number  $k$  for the shear mode when  $\text{Re} = 12000$ . Solid, dashed, and dotted lines stand for  $\text{Ma}_T = 100$ ,  $\text{Ma}_T = 200$ , and  $\text{Ma}_T = 300$ , respectively. The other parameter values are  $C_M = 1$ ,  $C_D = 10$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 10^5$ ,  $\text{Ca} = 0.001$ ,  $\text{Pr} = 7$ ,  $\text{Bi} = 1$ ,  $\text{Pe}_b = \text{Pe}_\Gamma = 10^3$ ,  $\xi_s = 10^3$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 1' = (1/60)^\circ$ . Here,  $U$  and  $S$  represent the unstable and stable regions.

Reynolds number regime, we also calculate the eigenfunction and the contours of the associated perturbation streamfunction for the wall mode. In this case, a significant change in the real part of the eigenfunction corresponding to the perturbation stream function for the wall mode takes place in the vicinity of the flexible substrate [see Fig. 15(e)]. As a result, the contours of the stream function for the wall mode ensue a cluster of open vortices that occupies the region near the flexible substrate [see Fig. 15(f)]. Moreover, when we compare the result of the wall mode with that of the shear mode depicted in Fig. 15(b), we find that the sizes of vortices reduce for the wall mode, but surprisingly, their patterns reverse.

To examine how the thermal Marangoni number  $\text{Ma}_T$  and the surfactant Marangoni number  $\text{Ma}_\Gamma$  affect shear mode instability, we compute the neutral stability curves and the temporal growth rate for the shear mode when  $C_M = 1$ ,  $C_D = 10$ ,  $C_B = 1$ ,  $C_T = 1$ ,  $C_K = 10^5$ ,  $\text{Ca} = 0.001$ ,  $\text{Pr} = 7$ ,  $\text{Bi} = 1$ ,  $\text{Pe}_b = \text{Pe}_\Gamma = 10^3$ ,  $\xi_s = 10^3$ ,  $k_s = 1$ ,  $\Gamma_E = 0.1$ , and  $\theta = 1' = (1/60)^\circ$ . In Figs. 16(a) and 16(b), we display the influence of  $\text{Ma}_T$  when  $\text{Ma}_\Gamma = 1$ . Here, we observe that the unstable region bounded by the neutral stability curve for the shear mode becomes slightly bigger as  $\text{Ma}_T$  rises, and

this fact is followed by the successive decrease in the critical Reynolds number for the onset of shear mode instability [see Fig. 16(a)]. Therefore, increasing  $Ma_T$  destabilizes shear mode instability. Since the critical Reynolds number decreases, we can quicken the appearance of shear mode instability by increasing the thermal Marangoni number. To strengthen this finding, we further compute the temporal growth rate for the shear mode and depict it in Fig. 16(b) with rising values of  $Ma_T$ . It is clear that the maximum temporal growth rate and the unstable range of wave number for the shear mode grow as  $Ma_T$  increases, endorsing the outcome shown in Fig. 16(a). Moreover, the temporal growth rate curve for the shear mode does not pass through  $k = 0$ . Instead, it arises in the finite wave-number regime, indicating that the shear mode is a finite wavelength mode. On the contrary, in Fig. 16(c), the neutral stability curve for the shear mode exhibits an opposite scenario in the  $(Re, k)$  plane when the surfactant Marangoni number  $Ma_F$  alters. Specifically, the unstable region decays significantly, and the critical Reynolds number for the onset of shear mode instability accelerates with the increase in the value of  $Ma_F$ . Therefore, increasing  $Ma_F$  stabilizes shear mode instability. Since the critical Reynolds number increases, we can delay the appearance of shear mode instability by increasing the surfactant Marangoni number. Plotting the temporal growth rate for the shear mode when  $Ma_F$  increases further confirms the stabilizing influence of the surfactant Marangoni number. The resulting temporal growth rate for the shear mode can be found in Fig. 16(d). Indeed, the maximum temporal growth rate and the unstable range of wave number for the shear mode reduce successively with rising values of  $Ma_F$ .

## VI. DISCUSSION AND CONCLUSIONS

We conduct a detailed linear stability analysis of a two-dimensional, incompressible, gravity-driven, nonisothermal viscous film flowing down a flexible substrate for disturbances of arbitrary wave numbers. Here, the viscous film is contaminated by a soluble surfactant, and the flexible substrate is uniformly heated. As a result, we have ignored thermal expansion of the flexible plate. Because of the nonisothermal soluble-surfactant-impured viscous film, we incorporate both heat- and mass-transfer equations along with the Navier-Stokes equations for describing the current flow configuration. We derive the OS BVP to decipher primary instabilities induced by unstable modes, including the hydrodynamic surface mode, the thermocapillary S- and P-modes, the wall mode, and the shear mode. Both the analytical long-wave asymptotic expansion and the numerical Chebyshev spectral collocation techniques are employed to solve the OS BVP. In the long-wave regime, we observe that surface mode instability exists, and this instability is intensified in the presence of the thermal Marangoni number but weakened by an increase in the surfactant Marangoni number. As a result, when these two opposing effects cancel each other out, the critical Reynolds number of the surface mode becomes equal to that reported by Alexander *et al.* [39], resulting in a surfactant-free isothermal film flowing over a flexible substrate. Since the stabilizing influences of the surfactant Marangoni stress and depthwise gravitational force outweigh the destabilizing influences of the thermal Marangoni stress and spring stiffness coefficient  $C_K$  in a certain range of the inclination angle, the surface mode becomes linearly stable in that range. Considering the physical mechanism of instability, we can specifically attribute the rationale for surface mode instability to the increase in speed of the dynamic hyperbolic wave. This increase is caused by the thermal Marangoni stress, which results from the surface tension gradient created by temperature differences. However, the surfactant Marangoni stress, which is induced by the surface tension gradient ensuing from changes in surface surfactant concentration, can weaken surface mode instability.

In the arbitrary wave-number regime, the numerical results indicate that there are four dominant unstable temporal modes when the Reynolds number keeps a low-to-moderate value. These temporal modes include the surface mode, wall mode, S-mode, and P-mode. Specifically, the hydrodynamic surface mode arises in the low-to-finite wave-number regime, the wall mode arises in the finite wave-number regime, the thermocapillary S-mode arises in the low wave-number regime, and the thermocapillary P-mode arises in the finite wave-number regime. Moreover, it

is necessary to rescale the dimensionless parameters for the appearance of the thermocapillary S-mode and P-mode. We observe that as the modified surfactant Marangoni number  $M_F$  goes up, unstable domains for the thermocapillary S-mode, P-mode, and the surface mode shrink. Hence, increasing  $M_F$  stabilizes surface mode, S-mode, and P-mode instabilities. In contrast, rising values of the modified thermal Marangoni number  $M_T$  expand unstable domains for the thermocapillary S-mode, P-mode, and the surface mode. Hence, increasing  $M_T$  destabilizes surface mode, S-mode, and P-mode instabilities. Moreover, there occurs a modal coalescence between the hydrodynamic surface mode and the thermocapillary S-mode in the long-wave regime as the modified thermal Marangoni number increases. As a result, the neutral stability curves for the surface mode and the S-mode merge and form a single neutral stability curve in the  $(\text{Re}, k)$  plane. However, if the modified surfactant Marangoni number is increased, the previously combined neutral stability curve splits again into two separate neutral stability curves for the surface mode and the S-mode, respectively.

Analyzing the impact of the base surface surfactant concentration parameter,  $\Gamma_E$ , reveals two opposite events for the hydrodynamic surface mode and the thermocapillary S- and P-modes. In particular, primary instabilities caused by the surface mode, S-mode, and P-mode are suppressed as  $\Gamma_E$  increases when it chooses low values within its reasonable range of  $0 \leq \Gamma_E \leq 1$ . If  $\Gamma_E$  chooses high values in the vicinity of  $\Gamma_E \approx 1$ , increasing  $\Gamma_E$  amplifies all surface mode, S-mode, and P-mode instabilities. On the other hand, the surfactant solubility parameter  $\xi_s$  exhibits only a stabilizing behavior for all primary instabilities generated by the hydrodynamic surface mode and the thermocapillary S- and P-modes.

To explore the modal coalescence between the P-mode and the wall mode in the finite wave-number regime, we significantly reduce the magnitudes of the spring stiffness coefficient  $C_K$ , the longitudinal tension parameter  $C_T$ , and the flexible rigidity parameter  $C_B$ . Initially, both P-mode- and wall-mode-induced instabilities appear in the finite wave-number regime but in different ranges of the Reynolds number. Interestingly, the P-mode and wall mode neutral stability curves merge together, and the combined unstable region grows with rising values of the modified thermal Marangoni number. If the modified surfactant Marangoni number is increased, the previously combined unstable region for the P-mode and the wall mode separates and decays with rising  $M_F$ . Hence,  $M_T$  destabilizes, but  $M_F$  stabilizes both P-mode and wall mode instabilities. Moreover, the finite wavelength P-mode instability occurs first, as its onset lies ahead of that of the finite wavelength wall mode instability. On the other hand, the base surface surfactant concentration parameter,  $\Gamma_E$ , exhibits a dual role in P-mode and wall mode instabilities. At the beginning, when  $\Gamma_E$  increases, unstable domains for the P-mode and the wall mode deplete. But when  $\Gamma_E \approx 1$ , the numerical result shows a completely reverse behavior. That is, unstable domains for the P-mode and the wall mode widen as  $\Gamma_E$  rises.

To probe how the competition between the surface mode and the wall mode is affected by the surfactant and thermal Marangoni numbers, we display their neutral stability curves in the  $(\text{Re}, k)$  plane. The numerical findings exhibit that the finite wavelength wall mode instability manifests before the long-wave surface mode instability when  $C_D$  equals 0.32. As the surfactant Marangoni number increases, wall mode instability strengthens, while surface mode instability weakens, always indicating the first appearance of wall mode instability in the finite wave-number regime. Albeit wall mode instability happens before surface mode instability, the maximum temporal growth rate for the surface mode is greater than that of the wall mode for low values of  $\text{Ma}_F$ . As the value of  $\text{Ma}_F$  rises, the maximum temporal growth rate for the wall mode surpasses that of the surface mode. Increasing the thermal Marangoni number, on the other hand, destabilizes both surface mode and wall mode instabilities by magnifying their respective unstable domains. As a result, wall mode instability always begins before surface mode instability with rising values of  $\text{Ma}_T$ . If  $C_D$  slightly increases to 0.35, the long-wave surface mode instability begins before the finite wavelength wall mode instability for low values of  $\text{Ma}_T$ . Since the effect of  $\text{Ma}_T$  on wall mode instability is more significant than on surface mode instability, wall mode instability initiates again before surface mode instability as  $\text{Ma}_T$  increases.

In the high Reynolds number regime, we perceive an additional mode, called the shear mode, when the angle of inclination is sufficiently low. Specifically, the shear mode has different characteristics than the surface mode. For instance, shear mode instability has a slower phase speed than surface mode instability, and it begins in the finite wave-number regime. Moreover, the stream function contours associated with the shear mode create large open vortices that occupy the center zone of the film layer rather than the film surface, as observed for the surface mode. In addition, shear mode instability intensifies as the thermal Marangoni number increases, while it becomes weaker with increasing values of the surfactant Marangoni number. Hence, we can quickly initiate shear mode instability by increasing the thermal Marangoni number. In contrast, we can delay its generation by increasing the surfactant Marangoni number.

Finally, we show that the inertialess instabilities caused by both the surface mode and the wall mode emerge in different ranges of wave number. Specifically, the surface-mode-induced inertialess instability initiates in the long-wave regime when  $C_D$  is equal to 1, while the wall-mode-induced inertialess instability initiates in the finite wave-number regime when  $C_D$  is equal to 0.32. Moreover, we observe that the thermal Marangoni number destabilizes both surface mode and wall mode instabilities. On the contrary, the surfactant Marangoni number stabilizes surface mode instability but destabilizes wall mode instability. Additionally, the influence of the thermal Marangoni number on the inertialess primary instability appears to be stronger than that of the surfactant Marangoni number.

#### DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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