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Three-dimensional dynamics of thin liquid films on vertical cylinders with Marangoni effect

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The effects of thermocapillary force on the three dimensional dynamics of thin liquid films flowing down a uniformly heated vertical cylinder are investigated using a thin film model. Linear and nonlinear analyses predict the existence of a non-axisymmetric mode due to the Marangoni effect. Symmetry-breaking of axisymmetric steady traveling waves is observed when the Marangoni number exceeds a critical value. Linear stability analysis demonstrates that the steady traveling wave can be unstable to azimuthal disturbances due to the Marangoni effect, leading to the formation of non-axisymmetric patterns. Published by AIP Publishing. [<http://dx.doi.org/10.1063/1.4974076>]

A thin liquid film flowing down a vertical cylinder driven by gravity experiences complex dynamics. The Plateau-Rayleigh mechanism modulated by the effects of gravity gives rise to the emergence of organized sliding beads and the breakup of liquid films.^{1,2} In industries, such a flow system is connected to coating systems, such as the coating of optical fibers and protection coating of tube walls.

The modeling work of a thin liquid film on a cylinder surface was pioneered by Frenkel.³ A thin film model equation was derived to investigate the linear dynamics of the interface based on the assumption that the film's thickness is much smaller than the cylinder radius.³ The nonlinear evolution of the thin film model³ has been examined by Kalliadasis and Chang,⁴ Chang and Demekhin.⁵ The formation of axisymmetric viscous beads in such a system has been discussed.^{4,5} Kliakhandler *et al.* extended the problem to thick film flows and reported three typical flow regimes.⁶ A Benney-type model⁶ was derived which, however, showed a poor agreement with the experimental data in the flow regime of large spaced droplets. An asymptotic model proposed by Craster and Matar⁸ showed an excellent agreement with experimental results of this flow regime. The Benney-type model⁶ has been extended to other complex problem, such as liquid films on vertical porous cylinders.⁷ The asymptotic model⁸ has been also extended to other complex flow systems.^{9,10} All these aforementioned models^{3–10} are valid to study the dynamics of liquid films at small flow rates. To model liquid films on vertical cylinders at moderate flow rates, an integral boundary layer model (IBL model)^{11–13} and a weighted-residual model (WR model)^{14,15} have been used, in which the fluid inertia is taken into account. It is noteworthy that the dynamics of liquid films was investigated under the assumption of axisymmetric flows in these previous studies.^{3–15} Additionally, non-axisymmetric nonlinear long wave dynamics is not amenable to analysis using the Benney-type model nor the IBL and WR models.

Practically, coating of fiber is operated in non-isothermal systems, for example, pulling out a cylinder from a hot liquid bath¹⁶ or cooling of hot glass fibers.¹⁷ Thermocapillary effect would be significant when the temperature difference between the cylinder surface and the surrounding environment is large. Hung *et al.*¹⁸ discussed the influence of cylinder radius on the weakly nonlinear stability of a condensate film and concluded that the film is more stable when the cylinder radius is large. Liu and Liu¹⁰ investigated the fully nonlinear long-wave dynamics of a uniformly heated film by extending the Benney-type model.⁸ Liu and Liu¹⁰ found that the Marangoni effect reinforced the Plateau-Rayleigh mechanism leading to the formation of small viscous beads. However, these studies were restricted to the two-dimensional axisymmetric flows.^{10,18} A careful survey of the available literatures on the dynamics of thin liquid films on vertical cylinders with Marangoni effects indicates that study on the non-axisymmetric dynamics is very limited. This paper aims to provide insights into the problem: will the non-axisymmetric mode emerge under the action of thermocapillary force?

We consider a thin Newtonian liquid film of constant density ρ and viscosity μ flowing down a uniformly heated vertical cylinder as shown in Figure 1. There is no slip and penetration on the surface of the system. The thickness of the film is h and the radius of the cylinder is a . In this paper, the cylindrical coordinate (r, θ, z) is chosen and the three dimensional velocity field $\mathbf{u} = ue_r + ve_\theta + we_z$ is considered. All the variables are non-dimensionalized by introducing the following scales: $(u, v, w) = (u', v', w')W$, $(r, z, h) = a(r', z', h')$, $(p - p_g) = \frac{\mu W}{a}p'$, $t = \frac{a}{W}t'$, and $T = (T_s - T_\infty)T'$. The velocity scale $W = \frac{\rho g a^2}{\mu}$. The primed variables are dimensionless. The dimensionless groups arise naturally,

$$\text{Re} = \frac{\rho W a}{\mu}, \quad \text{Ma} = \frac{\sigma_T(T_s - T_\infty)}{\mu W}, \quad C = \frac{\mu W}{\sigma_0}, \quad \text{Bi} = \frac{h_g a}{\lambda}, \quad (1)$$

which, respectively, are the Reynolds number, Marangoni number, capillary number, and Biot number. Here, h_g is

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FIG. 1. Geometry of the system.

the Newton's cooling coefficient and λ is the thermal conductivity. This paper considers a very small Reynolds number flow $Re \ll 1$ such that the fluid inertia and thermal convection can be neglected.¹⁰

In order to derive a nonlinear evolution equation describing the dynamics of the liquid film, we assume that the mean thickness of the film $\epsilon = h_0/a$ (h_0 the mean film thickness) is thin compared to the cylinder radius $\epsilon \ll 1$. In order to include the surface tension effect, we introduce $Ca = \epsilon^{-1}C$ as a rescaled capillary number. Furthermore, we introduce a new radial coordinate: $r = 1 + \epsilon y$, and the interfacial thickness is re-scaled as $h = \epsilon H$.²⁰ We then expand the variables in series of ϵ ,^{20,21}

$$\begin{aligned} u &= \epsilon^3 u_0 + \dots, & v &= \epsilon^2 v_0 + \dots, & w &= \epsilon^2 w_0 + \dots, \\ p &= p_0 + \dots, & T &= \epsilon T_0 + \dots, \end{aligned} \quad (2)$$

where the primes have been dropped.

Furthermore, we introduce a slow time $\tilde{t} = \epsilon^2 t$ to capture the interfacial dynamics. The evolution equation of H is obtained by solving the leading order problem of the Navier-Stokes equations and the energy equation,^{20,21}

$$\begin{aligned} \frac{\partial H}{\partial \tilde{t}} + \frac{\partial}{\partial z} \left(\frac{H^3}{3} \right) + \nabla \cdot \left[\frac{H^3}{3Ca} \nabla (H + \nabla^2 H) \right. \\ \left. + \frac{Ma}{2} \frac{Bi H^2 \nabla H}{(1 + BiH)^2} \right] = 0. \end{aligned} \quad (3)$$

Here, $\nabla = \mathbf{e}_\theta \frac{\partial}{\partial \theta} + \mathbf{e}_z \frac{\partial}{\partial z}$ and $\nabla^2 = \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial z^2}$. The tilde decoration on t is dropped. For $Ma = 0$ and an axisymmetric flow, the thin film model by Frenkel³ is recovered. The effects of capillary number on the axisymmetric dynamics have been discussed in many previous studies.^{3,10} This paper will concentrate the discussion on the Marangoni effect on the non-axisymmetric dynamics. The rescaled capillary number is fixed at $Ca = 1$.

The following discusses the linear stability of the thin film. The three dimensional stability of the basic state $\tilde{H} = 1$ is investigated using the normal mode analysis $H = 1 + \hat{H} \exp(\omega t + ikz + in\theta)$ which gives the dispersive relation,

$$\omega = -ik + \frac{1}{3Ca} (n^2 + k^2)(1 - n^2 - k^2) + \frac{MaBi}{2(1 + Bi)^2} (n^2 + k^2), \quad (4)$$

where $\omega = \omega_r + i\omega_i$ is the eigenvalue, whose real part is the effective growth rate and imaginary part ω_i is the frequency. k and n are the streamwise and azimuthal wave numbers, respectively.

The effective growth rate of the most unstable mode is given by

$$\omega_m = \frac{1}{12Ca} \left[1 + \frac{3MaBiCa}{2(1 + Bi)^2} \right]^2, \quad (5)$$

which occurs at $n^2 + k^2 = \frac{1}{2} [1 + \frac{3MaBiCa}{2(1 + Bi)^2}]$. Equation (5) indicates that the film becomes more stable when it is cooled; the film becomes more unstable when heated. In addition, Eq. (5) shows that the maximum growth rate $\omega_m \sim Ca^{-1}$, when the Marangoni effect is absent. It indicates that the surface tension is always destabilizing which agrees with the previous studies.³⁻⁵

Equation (5) also shows that for a heated film, the Marangoni effect always enhances the Rayleigh-Plateau instability which agrees with the study by Liu and Liu.¹⁰ However, Liu and Liu's model is only valid for an axisymmetric flow. When the flow is perturbed by a three dimensional disturbance, our results demonstrate that the flow is more unstable in the long wave range for $Ma > 0$ as illustrated in Figure 2(a). For a cooled film, the Marangoni effect stabilizes the flow as demonstrated in Figure 2(a). Moreover, when $\frac{MaBi}{2(1 + Bi)^2} < -1$, the flow is always linearly stable. It indicates that the instability in this flow system can be reinforced through heating up the cylinder and suppressed via cooling the cylinder. To explain the physical mechanism, let us assume that the film is depressed at a certain region due to external perturbations. When the film is heated, the surface tension reaches a minimum at the thinnest point while the surface tension in its neighboring zone is larger. Hence, liquids will flow out from this region due to the thermocapillary force, thereby enhancing the instability. However, when the film is cooled, liquids will flow into this region under the action of the thermocapillary force. Thereby, the instability is impeded. For critical stability, the azimuthal disturbance always stabilizes the flow since critical stability line rises rapidly as shown in Figure 2(b). This is due to the fact that the surface tension will impede the deformation of interface for $n \geq 1$.

FIG. 2. (a) The effective growth rate ω_r vs. the wave number k . “1” for $Ma = 0$, $n = 0$; “2” for $Ma = 2$, $n = 0$; “3” for $Ma = 2$, $n = 1$; “4” for $Ma = -0.5$, $n = 0$. (b) Marginal stability curve in k - Ma plane.

FIG. 3. Evolution of norm E_2 versus time. (a) $Ma = 1$, $Bi = 1$, $L_z = 15$. (b) $Ma = 2$, $Bi = 1$, $L_z = 15$. Lines are obtained by direct numerical simulation of Eq. (3). Triangle symbols are for the axisymmetric problem. “NAIC” stands for non-axisymmetric initial condition and “AIC” stands for axisymmetric initial condition.

The direct numerical simulation of Eq. (3) is carried out below. A numerical experiment in a periodic domain with $(z, \theta) \in [0, L_z) \times [0, 2\pi)$ is performed to demonstrate that either an axisymmetric unstable pattern or a non-axisymmetric unstable pattern can be observed in the system. L_z is the periodic length in the axial direction.

The evolution Equation (3) is solved numerically by the Crank-Nicolson method.¹⁹ The time step $\Delta t = 10^{-4} - 10^{-3}$ is chosen here. We use the Fourier method to discretize the problem in space,

$$H = \sum_{n=0}^{N_\theta} \sum_{m=0}^{N_z} \hat{H}_{mn} \exp\left(im \frac{2\pi}{L_z} z + in\theta\right) + c.c. \quad (6)$$

Typically, we use 64-128 Fourier modes in the axial direction and 16-32 Fourier modes in the azimuthal direction. When the base state is perturbed by an axisymmetric disturbance, the following equation is considered as an initial condition:

$$H(0, \theta, z) = 1 + 0.1 \sin\left(\frac{2\pi}{L_z} z\right). \quad (7)$$

When the non-axisymmetric disturbance is considered, we use the following profile as the initial condition:

$$H(0, \theta, z) = 1 + 0.1 \sin\left(\frac{2\pi}{L_z} z\right) \sin(\theta). \quad (8)$$

An energy norm $E_2 = \int_0^{L_z} \int_0^{2\pi} H^2 d\theta dz$ is defined to monitor the evolution behavior of the film as shown in Figure 3. The results are compared with the axisymmetric flow case

which neglects the azimuthal dynamics. When the Marangoni number is small, for example, $Ma = 1$, the film evolves in the same way as a two-dimensional axisymmetric film when subjected to an axisymmetric disturbance. After a long time, E_2 becomes constant, indicating that the film reaches a saturated state. This saturated state is a “steady” traveling-wave train propagating downstream. However, when the flow is perturbed by a three dimensional disturbance, E_2 oscillates with time and there is no “steady” state as demonstrated in Figure 3(a). Results in Figure 3(a) indicate that the film could either be two-dimensionally nor three-dimensionally unstable, depending on the initial condition. As the Marangoni number is increased to $Ma = 2$, for an axisymmetric initial condition, the interface evolves in the same way as an axisymmetric film in a short time and a saturated traveling wave is observed. However, an interesting phenomenon observed in Figure 3(b) is that this saturated traveling wave is not stable and starts to lose symmetry and evolve into a three-dimensional non-axisymmetric pattern spontaneously. Then, the film breaks up very quickly and non-axisymmetric droplets emerge. When a three dimensional initial condition is imposed, we observe that the film ruptures much more quickly as demonstrated in Figure 3(b). Our results are significantly different from that by Liu and Liu,¹⁰ in which only axisymmetric droplets would emerge due to the breakup of film. To exemplify the two different kinds of unstable modes in the system: the axisymmetric mode and the non-axisymmetric mode, we plot the interfacial shape of the film in Figure 4. Figure 4(b) clearly demonstrates that the Marangoni effect supports the three-dimensional non-axisymmetric unstable mode.

FIG. 4. Profiles of the liquid interface. $Ma = 2$, $Bi = 0.1$, $L_z = 50$, $\epsilon = 0.1$. (a) Initial condition: an axisymmetric disturbance. (b) Initial condition: non-axisymmetric disturbance.

We have observed that the film can transit from an axisymmetric state to a non-axisymmetric state as shown in Figure 3(b). Below we discuss the stability of axisymmetric traveling waves so as to provide physical insights into this phenomenon. First, we seek the solutions of the axisymmetric traveling waves which serve as the nonlinear base state. In a moving frame of velocity c , the two-dimensional axisymmetric traveling waves are “steady.” Applying the traveling wave transformation, $\zeta = z - ct$ and setting $H = H(\zeta)$, the two-dimensional axisymmetric traveling wave solution satisfies the following equation:

$$-cH\zeta + \left[\frac{H^3}{3} + \frac{H^3}{3Ca} \left(\frac{\partial H}{\partial \zeta} + \frac{\partial^3 H}{\partial \zeta^3} \right) + \frac{Ma}{2} \frac{BiH^2}{(1 + BiH)^2} \frac{\partial H}{\partial \zeta} \right]_{\zeta} = 0. \quad (9)$$

To fix the wave speed c , the mass conservative condition is imposed,

$$\frac{1}{L_z} \int_0^{L_z} H d\zeta = 1, \quad (10)$$

in which L_z is the length of computational domain in the axial direction which also corresponds to the droplet-droplet spacing for a single-droplet solution.^{8,9} The solution of H is approximated by Fourier series.^{8,9} Typically, 200 Fourier modes are enough to provide sufficient accuracy. The nonlinear Equation (9) is solved by a Newton iteration method²² which converges quickly with a good guess of the wave speed c and the profile of H . A continuation method is used to track the solution branch as parameter changes. Typical wave profiles are shown in Figure 5. It is shown here that a larger Marangoni number causes a steeper wave with a higher amplitude. Even though the linear wave speed $c = 1$ is not affected by the Marangoni effect as can be seen from the dispersive relation

FIG. 5. The steady traveling wave profile in the moving framework.

Eq. (4), the nonlinear wave speed is significantly influenced by the Marangoni effect as shown in Figure 6. Figure 6(a) shows that the nonlinear wave speed c decreases with increasing Ma , when droplet spacing L_z is short. For a large droplet spacing L_z , the nonlinear wave speed c is promoted by the Marangoni effect. In addition, the long-spaced droplet has a bigger size for a larger Ma as illustrated in Figures 5 and 6. Our results agree well with that by Liu and Liu,¹⁰ which also show that the sliding speed of long-spaced droplet is promoted by the Marangoni effect.

Furthermore, we investigate the stability of the steady axisymmetric traveling waves. In a moving frame ($\zeta = z - ct, \theta$), Eq. (3) is restated as

$$\frac{\partial H}{\partial t} - c \frac{\partial H}{\partial \zeta} + \frac{\partial}{\partial \zeta} \left(\frac{H^3}{3} \right) + \nabla \cdot \left[\frac{H^3}{3Ca} \nabla (H + \nabla^2 H) \right] + \frac{Ma}{2} \frac{BiH^2 \nabla H}{(1 + BiH)^2} = 0. \quad (11)$$

The stability of the axisymmetric traveling wave which serves as the basic state $\bar{H}(\zeta)$ is investigated by perturbing it with infinitesimal disturbances $H'(t, \zeta, \theta)$. Linearization of Eq. (11) gives

$$\begin{aligned} \frac{\partial H'}{\partial t} - c \frac{\partial H'}{\partial \zeta} + \frac{\partial}{\partial \zeta} (\bar{H}^2 H') + \frac{\partial}{\partial \zeta} \left[\frac{\bar{H}^3}{3Ca} \frac{\partial}{\partial \zeta} (H' + \nabla^2 H') \right] + \frac{\partial}{\partial \zeta} \left[\frac{\bar{H}^2 H'}{Ca} \frac{\partial}{\partial \zeta} (\bar{H} + \nabla^2 \bar{H}) \right] \\ + \frac{\partial}{\partial \zeta} \left[\frac{Ma}{2} \frac{Bi\bar{H}^2}{(1 + Bi\bar{H})^2} \frac{\partial H'}{\partial \zeta} + \frac{Ma}{2} \frac{2Bi\bar{H}H'}{(1 + Bi\bar{H})^2} \frac{\partial \bar{H}}{\partial \zeta} - \frac{Ma}{2} \frac{2Bi^2\bar{H}^2 H'}{(1 + Bi\bar{H})^3} \frac{\partial \bar{H}}{\partial \zeta} \right] \\ + \frac{\partial}{\partial \theta} \left[\frac{\bar{H}^3}{3Ca} \frac{\partial}{\partial \theta} (H' + \nabla^2 H') + \frac{Ma}{2} \frac{Bi\bar{H}^2}{(1 + Bi\bar{H})^2} \frac{\partial H'}{\partial \theta} \right] = 0. \end{aligned} \quad (12)$$

FIG. 6. (a) Nonlinear wave speed c vs. the wave length L_z for $Bi = 1$. (b) c vs. Ma for $L_z = 15$.

FIG. 7. (a) The effective growth rate s_r vs. the wave length L_z for $Ma = 2$, $Bi = 1$. (b) The effective growth rate s_r vs. the Marangoni number for $L_z = 7.5, 15, 30$. For $n \geq 2$, $s_r < 0$.

Now, we consider the Floquet analysis: $H' = \hat{H}(\zeta) \exp(i\phi k\zeta + in\theta + st)$ where the detuning parameter $\phi \in [0, 1/2]$ and $s = s_r + is_i$ is eigenvalue. An important class of disturbances is the disturbance with the same wave length as the nonlinear base state. For this case, the detuning parameter $\phi = 0$. In this paper, we only consider this particular case that the disturbance has the same wave length as the base state. We follow the previous study by Scheid *et al.*²³ and impose the periodic boundary conditions to solve the eigenvalue problem in Eq. (12).

Figure 7(a) shows that the axisymmetric traveling wave is unstable when the wave length is large for $Ma = 2$. Such a phenomenon indicates that bigger size droplets are unstable. However, when the axisymmetric film is perturbed by azimuthal disturbances, non-axisymmetric unstable mode can be supported by the Marangoni effect for short-spacing droplets as seen in Figure 7(a). In Figure 3(b), when the initial axisymmetric initial condition is imposed, two sub-waves of the same amplitude were observed in the “steady” traveling wave train. Stability analysis of a sub-wave reveals that it is unstable to the azimuthal disturbance which agrees with the direct numerical simulation of the symmetry-breaking phenomenon. Figure 7(b) indicates that in an isothermal film flow, $Ma = 0$, the long spaced droplets (big size) are unstable, while short spaced beads (small size) are stable. Our stability analysis agrees well with previous experimental and numerical studies that large droplets will break up into small droplets.^{8,10} If the Marangoni effect is present, the flow is more unstable when it is perturbed by the axisymmetric disturbances as seen in Figure 7(b). As the wave length becomes larger, for example, $L_z = 30$, the axisymmetric mode always dominates the non-axisymmetric mode as shown in Figure 7(b). However, as the axisymmetric big-sized beads are unstable which will break up into small-sized beads,^{8,10} it would be expected to see that non-axisymmetric unstable mode may emerge due to these unstable small-sized beads.

In this paper, we investigated the three dimensional dynamics of a thin liquid film falling down a vertical cylinder. A thin film model was derived to discuss the linear stability and nonlinear evolution of the film.

The three-dimensional stability analysis of the film demonstrated that the axisymmetric unstable mode always dominates the system when the Marangoni effect is absent. However, the non-axisymmetric unstable mode could be supported by the Marangoni effect. Direct numerical simulation of the thin film model demonstrated that the film

evolves in the same way as an axisymmetric system when the initial condition is seeded by axisymmetric disturbances at small Marangoni numbers. When a non-axisymmetric, three dimensional initial condition was imposed, the film may oscillate or break up quickly and formation of three dimensional structures was observed. Interestingly, nonlinear simulation revealed that symmetry-breaking phenomenon can occur in the system when the Marangoni number exceeds a critical value. The film can evolve into a non-axisymmetric pattern even if an axisymmetric initial condition is imposed. To explain the symmetry-breaking phenomenon, the linear stability analysis of steady traveling waves was carried out. Results demonstrated that the non-axisymmetric mode could emerge due to the azimuthal instability of these axisymmetric steady waves.

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