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Thermocapillary instability of irradiated transparent liquid films on absorbing solid substrates

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The thermocapillary instability of irradiated transparent liquid films on absorbing solid substrates is investigated by means of linear stability analysis. Under such circumstances, incident light passes through a film and is absorbed by the substrate, and the film is then heated by the heat influx across the interface with the substrate. The optical absorption in the substrate is affected by optical reflection. The energy reflectance varies periodically with the film thickness due to optical interference between light waves reflected from the gas-liquid and liquid-solid interfaces. The periodic variation of the reflectance strongly affects the film stability, which also varies periodically with the film thickness. Characteristic scales of the instability are also affected by the substrate thickness and incident light intensity. While qualitative aspects of the stability can be easily obtained from the analysis based on a simplified model that is derived under the thin-substrate assumption, the quantitative evaluation for the case of substrates of moderate to large thickness should be based on a more generalized model that allows for substrates of arbitrary thickness. © 2013 AIP Publishing LLC.

I. INTRODUCTION

Thermocapillary instability, also known as thermal Marangoni instability, of thin liquid films has been investigated extensively in various fields of research.1, 2 In particular, thermocapillary instability of a liquid film that is heated (or cooled) uniformly from below has been well studied, where the surface tension gradient contributes to the destabilization (or stabilization) of the film if the surface tension decreases monotonically with temperature.1–6

Irradiation is one method of heating liquids. Thermocapillary flow induced by irradiative heating has been applied to the control of convection patterns,7 control of flow patterns near contact lines,8–11 mixing in microdroplets,12, 13 laser surface texturing,14–16 and manipulation of micro-objects.17–26 Irradiative heating involves internal heat generation due to optical absorption in a medium. The absorption of light is generally described by the Bouguer-Lambert-Beer law,27 which states that the radiation intensity in the medium attenuates exponentially with the distance from the surface facing the light source. Irradiative heating of liquid films on solid substrates can be classified as direct or indirect heating. In direct heating, the film is directly heated by the absorption of light. In contrast, in indirect heating, incident light passes through a film and is absorbed by the substrate, and the film is then heated by the heat influx across the interface with the substrate. Oron28 introduced the Bouguer-Lambert-Beer law in a theoretical study of the dynamics of directly heated liquid films, in which the dynamics was described by an evolution equation that is based on the thin-film approximation, also known as the long-wave or lubrication approximation, as outlined in Ref. 1. The formulation was extended by Grigoriev,29 who took the heat transfer in the substrate into account. Grigoriev29 also referred to indirect heating and considered the case in which the substrate is optically thick, such that all radiation passing through a film can be assumed to be absorbed on the film-substrate interface.

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Later, Atena and Khenner\textsuperscript{30} took optical reflection into account in a study targeting absorbing films. Saeki \textit{et al.}\textsuperscript{31} recently proposed a more generalized formulation in which the heat transfer in the substrate, the thickness of which is arbitrary, and the optical reflection in which the film and substrate can be either transparent or absorbing are taken into account. Note that the derived model can properly deal with indirect heating of a transparent film, the thickness of which is comparable to the light wavelength, on an absorbing substrate. Under such circumstances, the effect of optical interference between light waves reflected from the gas-liquid and liquid-solid interfaces is not negligible,\textsuperscript{27} and the energy reflectance then varies periodically with the film thickness. Focusing on such film/substrate systems irradiated by monochromatic light with a laterally uniform intensity distribution, Saeki \textit{et al.}\textsuperscript{31} also performed numerical simulations based on the derived model and found that the stability and evolution of a film are strongly affected by the first derivative of the reflectance with respect to the film thickness. The results have been qualitatively confirmed by linear stability analysis based on a simplified model.\textsuperscript{31} However, the detailed stability properties have not yet been clarified.

In the present paper, we focus on uniformly irradiated transparent thin liquid films on absorbing solid substrates, as in the previous study,\textsuperscript{31} and perform linear stability analysis to investigate the stability of the films in more detail. The analysis is based on two models: a simplified model in which both the film and substrate are assumed to be sufficiently thin, so that the thin-film approximation is valid, and a more generalized model in which the film is assumed to be sufficiently thin but the substrate is not. We compare the results obtained from these two models and evaluate the stability both qualitatively and quantitatively.

The remainder of the present paper is organized as follows. In Sec. II, we pose a problem in which a transparent liquid film/absorbing solid substrate system is irradiated by monochromatic light and then derive two models that describe the evolution of both the profile of the gas-liquid interface (also called the free surface) and the temperature field. The linear stability analysis based on the models is presented in Sec. III. We present the summary and conclusions in Sec. IV. The Appendix describes the numerical methods used for analysis.

II. FORMULATION

A. Configuration and coordinate system

We consider a liquid film/solid substrate system in 2D space, as shown in Fig. 1. The ambient gas and liquid film are completely transparent, whereas the solid substrate is absorbing. The substrate, the upper and lower surfaces of which are flat and parallel to each other, is placed horizontally on an isothermal blackbody. Monochromatic light traveling in the vertical direction falls on the free surface. A Cartesian coordinate system is selected such that the origin is at a point on the liquid-solid interface, the $\tilde{x}$-axis is in the plane of the interface, and the $\tilde{z}$-axis is perpendicular to the plane of the interface. The free surface profile is described by the local film thickness $\tilde{h}(\tilde{x}, \tilde{t})$, where $\tilde{t}$ is the time, and the substrate has uniform thickness $\tilde{d}$.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure1.png}
\caption{Transparent liquid film/absorbing solid substrate system exposed to irradiation.}
\end{figure}
B. Basic equations

The liquid is assumed to be an incompressible Newtonian fluid, and the temperature variation is assumed to be so small that all physical properties, except the surface tension of the liquid, are constant. We also assume that the temperature variation due to viscous dissipation and the effects of gravity are negligible. Although the contributions of optical radiation to heat transfer generally include reflection, absorption, scattering, and emission, for simplicity, we consider only the first two factors. As will be mentioned later, we consider the case in which the film thickness is much smaller than the characteristic horizontal length. The free surface can then be assumed to be optically smooth. Therefore, the traveling direction of the light can be limited to the vertical direction.28

The continuity, Navier-Stokes, and energy equations for the liquid layer are expressed as

$$\nabla \cdot \mathbf{u} = 0, \quad \rho \left[ \partial_t \mathbf{u} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right] = -\nabla p + \mu \nabla^2 \mathbf{u}, \quad \rho c \left[ \partial_t \theta + (\mathbf{u} \cdot \nabla) \theta \right] = k \nabla^2 \theta,$$

where $\nabla = (\partial_x, \partial_z, \partial_t)$, $\mathbf{u} = (u(x, z, t), \bar{w}(x, z, t))$ is the velocity field in the liquid layer, $\bar{p}(x, z, t)$ is the pressure, $\bar{\theta}(x, z, t)$ is the liquid temperature, and $\rho$, $\mu$, $c$, and $k$ are the density, viscosity, specific heat at constant pressure, and thermal conductivity of the liquid, respectively. The heat equation for the solid layer is expressed as

$$\rho_s c_s \partial_t \theta_s = k_s \nabla^2 \theta_s + \bar{a}_s \bar{R}(\bar{h}) e^{i \theta_s z},$$

where $\bar{\theta}_s(x, z, t)$ is the solid temperature, $\bar{R}(\bar{h})$ is the energy reflectance, and $\rho_s$, $c_s$, $k_s$, and $\bar{a}_s$ are the density, specific heat at constant pressure, thermal conductivity, and absorption coefficient of the solid, respectively. The second term on the right-hand side of Eq. (2) is the internal heat generation per unit volume, which is based on the Bouguer-Lambert-Beer law.27 The absorption coefficient $\bar{a}_s$ is given by

$$\bar{a}_s = \frac{4\pi}{\lambda} k_2,$$

where $\lambda$ is the light wavelength in vacuum and $k_2$ is the extinction coefficient of the solid. The reflectance $\bar{R}(\bar{h})$ is expressed as

$$\bar{R}(\bar{h}) = \frac{C_1 + C_2 + C_3 \cos(\bar{h}) + C_4 \sin(\bar{h})}{1 + C_1 C_2 + C_3 \cos(\bar{h}) + C_4 \sin(\bar{h})},$$

with

$$\bar{h} = \frac{4\pi}{\lambda} n_1,$$

$$C_1 = g_1^2, \quad C_2 = g_2^2 + g_3^2, \quad C_3 = 2g_1g_2, \quad C_4 = 2g_1g_3,$$

$$g_1 = \frac{n_0 - n_1}{n_0 + n_1}, \quad g_2 = \frac{n_1^2 - n_2^2}{(n_1 + n_2)^2 + k_2^2}, \quad g_3 = \frac{2n_1k_2}{(n_1 + n_2)^2 + k_2^2},$$

where $n_0$, $n_1$, and $n_2$ are the refractive indices of the gas, liquid, and solid, respectively. Note that the refractive indices and extinction coefficient depend on the light wavelength $\lambda$. Equation (3) is obtained by applying a more generalized expression32 to the system shown in Fig. 1. The sine and cosine functions on the right-hand side of Eq. (3) are derived from optical interference between light waves reflected from the gas-liquid and liquid-solid interfaces. On the free surface $\bar{z} = \bar{h}(x, t)$, the kinematic condition, the normal and tangential stress balances, and Newton’s
The heat flux continuity conditions are imposed:

\[
\dot{w} = \partial_j \ddot{h} + \ddot{u} \partial_j \ddot{h},
\]

(4a)

\[-(\ddot{p} - \ddot{p}_\infty) + \mathbf{n} \cdot (\mathbf{r} \cdot \mathbf{n}) = -\gamma K,
\]

(4b)

\[\mathbf{t} \cdot (\mathbf{r} \cdot \mathbf{n}) = \mathbf{t} \cdot \nabla \gamma,
\]

(4c)

\[-k \nabla \bar{\theta} \cdot \mathbf{n} = \alpha (\bar{\theta} - \bar{\theta}_\infty),
\]

(4d)

where \(\mathbf{n}\) is the outward unit normal vector to the surface, \(\mathbf{t}\) is the unit tangential vector to the surface, \(K\) is the curvature of the surface, \(\mathbf{r}\) is the deviatoric stress tensor, each component of which is given by \(\tau_{ij} = \mu (\partial_j \ddot{u}_i + \partial_i \ddot{u}_j)\), \(\gamma\) is the surface tension of the liquid, \(\alpha\) is the convection heat transfer coefficient, and \(\ddot{p}_\infty\) and \(\bar{\theta}_\infty\) are the pressure and temperature of the ambient gas, respectively. The unit vectors, \(\mathbf{n}\) and \(\mathbf{t}\), and the curvature \(K\) are given by

\[
\mathbf{n} = \frac{(-\partial_j \ddot{h}, 1)}{(1 + (\partial_j \ddot{h})^2)^{1/2}}, \quad \mathbf{t} = \frac{(1, \partial_i \ddot{h})}{(1 + (\partial_i \ddot{h})^2)^{1/2}}, \quad K = \nabla \cdot \mathbf{n} = -\frac{\partial_i^2 \ddot{h}}{(1 + (\partial_i \ddot{h})^2)^{3/2}}.
\]

We assume that the surface tension is expressed as a linearly decreasing function of the free surface temperature \(\bar{\theta}_h(\bar{x}, \bar{t}) \equiv \bar{\theta}(\bar{x}, \bar{h}(\bar{x}, \bar{t}), \bar{t})
\]

\[\gamma(\bar{\theta}_h) = \gamma_\infty - \gamma'(\bar{\theta}_h - \bar{\theta}_\infty),
\]

where \(\gamma_\infty\) is the surface tension at the reference temperature \(\bar{\theta}_\infty\), and \(\gamma'\) is a positive constant. On the liquid-solid interface \(\bar{z} = 0\), the no-slip and no-penetration conditions and the temperature and heat flux continuity conditions are imposed:

\[
\ddot{u} = 0,
\]

(5a)

\[
\bar{\theta} = \bar{\theta}_s, \quad k \partial_z \bar{\theta} = k_s \partial_s \bar{\theta}_s.
\]

(5b)

On the bottom of the solid substrate \(\bar{z} = -\bar{d}\), a fixed temperature \(\bar{\theta}_b\) is assumed:

\[
\bar{\theta}_s = \bar{\theta}_b.
\]

(6)

**C. Thin-film approximation of the fluid dynamics equations**

We introduce the parameter

\[
\epsilon = \frac{h_0}{\bar{\ell}},
\]

where \(h_0\) and \(\bar{\ell}\) are the reference film thickness and horizontal length, respectively. In the liquid layer \(0 \leq \bar{z} \leq h(\bar{x}, \bar{t})\), the independent and dependent variables are normalized as follows:

\[
x = \epsilon \frac{\bar{x}}{h_0}, \quad \bar{z} = \frac{\bar{z}}{h_0}, \quad \bar{t} = \frac{\epsilon U}{h_0},
\]

\[
h(x, t) = \frac{\ddot{h}}{h_0}, \quad u(x, z, t) = \frac{\ddot{u}}{U}, \quad w(x, z, t) = \ddot{w},
\]

\[
p(x, z, t) = \frac{\epsilon h_0}{\mu U}(\ddot{p} - \ddot{p}_\infty), \quad \theta(x, z, t) = \frac{\ddot{\theta} - \ddot{\theta}_\infty}{\ddot{\theta}_\infty},
\]

where \(U\) is the reference velocity in the horizontal direction. Note that the normalized horizontal coordinate \(x\) and time \(t\) are also used in the solid layer. Equations (1), (4), and (5a) can be normalized.
as follows: in $0 < z < h(x, t)$,

\begin{equation}
\partial_t u + \partial_z w = 0, \tag{7a}
\end{equation}

\begin{equation}
\epsilon \text{Re} (\partial_t + u \partial_x + w \partial_z) u = -\partial_z p + (\epsilon^2 \partial^2_x + \partial^2_z) u, \tag{7b}
\end{equation}

\begin{equation}
\epsilon^3 \text{Re} (\partial_t + u \partial_x + w \partial_z) w = -\partial_z p + (\epsilon^4 \partial^2_x + \epsilon^2 \partial^2_z) w, \tag{7c}
\end{equation}

\begin{equation}
\epsilon \text{Pe} (\partial_t + u \partial_x + w \partial_z) \theta = (\epsilon^2 \partial^2_x + \partial^2_z) \theta, \tag{7d}
\end{equation}

on $z = h(x, t)$,

\begin{equation}
w = \partial_t h + u \partial_x h, \tag{8a}
\end{equation}

\begin{equation}
-p + 2\epsilon^2 \left( \frac{(\partial_t h)^2 - 1}{1 + \epsilon^2 (\partial_t h)^2} \right) \partial_z u - (\partial_t h) (\partial_z u + \epsilon^2 \partial_x w) \right) \right) = \frac{(S - \epsilon^2 M \theta_0) \partial_x h}{\left(1 + \epsilon^2 (\partial_t h)^2 \right)^{3/2}}, \tag{8b}
\end{equation}

\begin{equation}
-4\epsilon^2 (\partial_t h) \partial_z u + (1 - \epsilon^2 (\partial_t h)^2)(\partial_z u + \epsilon^2 \partial_x w) \right) \right) = -M \partial_x \theta_0, \tag{8c}
\end{equation}

\begin{equation}
\frac{\epsilon^2 (\partial_t h) \partial_z \theta - \partial_x \theta}{\left(1 + \epsilon^2 (\partial_t h)^2 \right)^{1/2}} = B \theta_0, \tag{8d}
\end{equation}

and on $z = 0$,

\begin{equation}
(u, w) = (0, 0), \tag{9}
\end{equation}

where $\theta_0(x, t) \equiv \theta(x, h(x, t), t)$, and $\text{Re}$, $\text{Pe}$, $M$, $B$, and $S$, which are defined in the following, are the Reynolds, Péclet, Marangoni, Biot, and inverse capillary numbers, respectively:

\begin{equation}
\text{Re} = \frac{\rho U h_0}{\mu}, \quad \text{Pe} = \frac{\rho c U h_0}{k}, \quad M = \frac{\epsilon \tilde{\theta}_\infty \gamma'}{\mu U}, \quad B = \frac{\alpha h_0}{k}, \quad S = \frac{\epsilon^3 \gamma_\infty}{\mu U}.
\end{equation}

Considering that the film thickness is much smaller than the horizontal length, i.e., $\epsilon \ll 1$, we expand $u$, $w$, $p$, and $\theta$ in powers of the small parameter $\epsilon$,

\begin{equation}
(u, w, p, \theta) = (u_0, w_0, p_0, \theta_0) + \epsilon (u_1, w_1, p_1, \theta_1) + \cdots.
\end{equation}

In addition, we assume that $\text{Re}$, $\text{Pe}$, $M$, $B$, and $S$ are $O(1)$. At leading order in $\epsilon$, Eqs. (7)–(9) are reduced to the following: in $0 < z < h(x, t)$,

\begin{equation}
\partial_t u_0 + \partial_z w_0 = 0, \tag{10a}
\end{equation}

\begin{equation}
\partial_t p_0 = \partial^2_x u_0, \tag{10b}
\end{equation}

\begin{equation}
\partial_z p_0 = 0, \tag{10c}
\end{equation}

\begin{equation}
\partial^2_x \theta_0 = 0, \tag{10d}
\end{equation}

\begin{equation}
\partial_t h_0 + u \partial_x h_0 + w \partial_z h_0 = 0, \tag{11a}
\end{equation}

\begin{equation}
\partial_t p_0 + \partial_x h_0 \partial_z p_0 + \partial_z h_0 \partial_x p_0 + \partial^2_x h_0 \partial_z w_0 + \partial^2_z h_0 \partial_x w_0 = 0, \tag{11b}
\end{equation}

\begin{equation}
\partial_t \theta_0 + u \partial_x \theta_0 + w \partial_z \theta_0 = 0, \tag{11c}
\end{equation}

\begin{equation}
\partial_t h_0 + u \partial_x h_0 + w \partial_z h_0 = 0, \tag{11d}
\end{equation}

\begin{equation}
\partial_t h_0 + u \partial_x h_0 + w \partial_z h_0 = 0, \tag{11e}
\end{equation}

\begin{equation}
\partial_t h_0 + u \partial_x h_0 + w \partial_z h_0 = 0. \tag{11f}
\end{equation}
on \( z = h(x, t) \),

\[
\begin{align*}
\omega_0 &= \partial_t h + u_0 \partial_x h, \quad (11a) \\
p_0 &= -S \partial_x^2 h, \quad (11b) \\
\partial_t u_0 &= -M \partial_x \theta_0, \quad (11c) \\
\partial_z \theta_0 &= -B \theta_0, \quad (11d)
\end{align*}
\]

and on \( z = 0 \),

\[
(u_0, w_0) = (0, 0), \quad (12)
\]

where \( \theta_{h0}(x, t) \equiv \theta_0(x, h(x, t), t) \). Hereinafter, the subscript 0 is dropped for simplicity. It follows from Eqs. (10)–(12) that

\[
p = -S \partial_x^2 h, \quad (13a)
\]

\[
u = \left( \frac{z^2}{2} - h z \right) \partial_x p - M z \partial_x \theta h, \quad (13b)
\]

\[
\partial_t h + \partial_x \int_0^h u \, dz = 0, \quad (13c)
\]

\[
\theta(x, z, t) = \frac{1 + B(h - z)}{1 + Bh} \theta(x, 0, t), \quad (13d)
\]

\[
\theta_b(x, t) \equiv \theta(x, h(x, t), t) = \frac{\theta(x, 0, t)}{1 + Bh}. \quad (13e)
\]

Substituting Eq. (13a) into Eq. (13b) and the result into Eq. (13c) yields

\[
\partial_t h = -S \frac{\partial_x (h^3 \partial_x^3 h)}{3} + M \frac{\partial_x (h^2 \partial_x \theta_0)}{2}. \quad (14)
\]

This equation is referred to as the thin-film equation.

### D. Normalization and approximation of the heat equation

#### 1. Thin-substrate case

Here we consider that the thickness of the substrate is of the same order as that of the liquid film. The vertical coordinate in the solid layer can then be normalized in the same manner as in the liquid layer, and correspondingly, the substrate thickness and absorption coefficient are normalized as

\[
d = \frac{\tilde{d}}{h_0}, \quad \alpha_s = h_0 \tilde{\alpha}_s.
\]

The solid temperature and the lower boundary temperature are normalized in a manner similar to the liquid temperature:

\[
\theta_s(x, z, t) = \frac{\tilde{\theta}_s - \tilde{\theta}_0}{\tilde{\theta}_\infty}, \quad \theta_b = \frac{\tilde{\theta}_b - \tilde{\theta}_0}{\tilde{\theta}_\infty}.
\]
Equations (2), (5b), and (6) can then be normalized as

\[ \epsilon^2 \partial_t \theta_s = \mathcal{K}(\epsilon^2 \partial_z^2 + \partial_z^2)\theta_s + \frac{\mathcal{K}}{\kappa_s} \alpha_s I (1 - R(h)) e^{\alpha_z}, \]  
\[ \text{Eq. (15a)} \]

\[ \theta_s(x, 0, t) = \theta(x, 0, t), \quad \partial_z \theta_s|_{z=0} = \frac{1}{\kappa_s} \partial_z \theta|_{z=0}, \]  
\[ \text{Eq. (15b)} \]

\[ \theta_t(0, -d, t) = \theta_b. \]  
\[ \text{Eq. (15c)} \]

Parameters \( \mathcal{K}, \kappa_s, \) and \( I(x, t) \) are defined as

\[ \mathcal{K} = \frac{\epsilon \kappa_s}{P \rho}, \quad \kappa_s = \frac{k_s}{\kappa}, \quad I(x, t) = \frac{h_0}{k \theta_{\infty} I}, \]

where \( \kappa_s = \kappa_s / \kappa \), in which \( \kappa = k \rho c \) and \( \kappa_s = k_s \rho c_s \) are the thermal diffusivities of the liquid and solid, respectively. Using \( h \) and \( \chi = h_0 \tilde{\chi} \) instead of \( \tilde{h} \) and \( \tilde{\chi} \), we can rewrite the reflectance as

\[ R(h) = \frac{C_1 + C_2 + C_3 \cos(\chi h) + C_4 \sin(\chi h)}{1 + C_1 C_2 + C_3 \cos(\chi h) + C_4 \sin(\chi h)}, \]  
\[ \text{Eq. (16)} \]

We expand \( \theta \) and \( \theta_s \) in the power series \( (\theta, \theta_s) = (\theta_0, \theta_{s0}) + \epsilon(\theta_1, \theta_{s1}) + \cdots \) and assume that \( \mathcal{K}, \kappa_s, \) and \( \alpha_s I \) are \( O(1) \). At leading order in \( \epsilon \), Eq. (15) is reduced to

\[ \partial_z^2 \theta_s = -\frac{1}{\kappa_s} \alpha_s I (1 - R(h)) e^{\alpha_z}, \]  
\[ \text{Eq. (17a)} \]

\[ \theta_{s0}(x, 0, t) = \theta_0(x, 0, t), \quad \partial_z \theta_{s0}|_{z=0} = \frac{1}{\kappa_s} \partial_z \theta_0|_{z=0}. \]  
\[ \text{Eq. (17b)} \]

\[ \theta_{s0}(0, -d, t) = \theta_b. \]  
\[ \text{Eq. (17c)} \]

Hereinafter, the subscript 0 is dropped for simplicity. Solving Eq. (17) for \( \theta_s \) and substituting Eq. (13d) into the result yields

\[ \theta_s(x, z, t) = \theta_b + \frac{1}{\kappa_s} \left[ \frac{-B(z + d)}{1 + Bh} \theta_s(x, 0, t) + I(1 - R(h)) \left( \frac{e^{-a_s d} - e^{a_s z}}{a_s} + z + d \right) \right]. \]  
\[ \text{Eq. (18a)} \]

The liquid-solid interface temperature is obtained as

\[ \theta(x, 0, t) = \theta_s(x, 0, t) = \frac{k_s (1 + Bh)}{k_s (1 + Bh) + Bd} \left[ \theta_b + \frac{1}{\kappa_s} I(1 - R(h)) \frac{e^{-a_s d} - 1 + a_s d}{a_s} \right]. \]  
\[ \text{Eq. (18b)} \]

2. General case

Here we consider a formulation that allows for a substrate of arbitrary thickness. The vertical coordinate in the solid layer, the substrate thickness, and the absorption coefficient are then normalized as

\[ Z = \frac{z}{h_0}, \quad D = \frac{d}{h_0}, \quad A_s = \frac{h_0}{\epsilon}, \]

where \( Z \equiv \epsilon z, D \equiv \epsilon d, \) and \( A_s = \alpha_s / \epsilon. \) Following the change of the normalized vertical coordinate from \( z \) in the thin-substrate case to \( Z \) in the present case, we replace the function \( \theta_s(x, z, t) \) with \( \theta_s(x, Z, t) \), where, although one of the independent variables is changed, the normalization itself is not
changed. Equations (2), (5b), and (6) can then be normalized as

\[ \partial_t \theta_s = \mathcal{K} (\partial_x^2 + \partial_Z^2) \theta_s + \frac{\mathcal{K}}{\epsilon k_s} A_I (1 - R(h)) e^{A_Z}, \quad (19a) \]

\[ \frac{\partial Z}{\epsilon k_s} \partial_z \theta_s \bigg|_{Z=0} = \frac{1}{\epsilon k_s} \partial_z \theta_s \bigg|_{Z=0}, \quad (19b) \]

\[ \theta_s (x, -D, t) = \theta_b. \quad (19c) \]

We expand \( \theta \) in the power series \( \theta = \theta_0 + \epsilon \theta_1 + \cdots \) and evaluate Eq. (19b) at leading order in \( \epsilon \) under the assumption that \( \epsilon k_s = O(1) \). Dropping the subscript 0 for simplicity, we obtain equations of the same form as Eq. (19b). Therefore, we regard Eq. (19) as the leading order system for the present case.

III. LINEAR STABILITY ANALYSIS

Addressing a uniform irradiation problem, in which the incident light intensity \( I \) is taken to be constant, we discuss the linear stability of laterally uniform steady states. We first derive the linear dispersion relation from a simplified model in which the substrate is assumed to be sufficiently thin. We then derive the linear dispersion relation from a more generalized model that allows for a substrate of arbitrary thickness. Then, showing the results for specific parameter values, we compare the two models and evaluate the stability both qualitatively and quantitatively.

A. Linear dispersion relations and their basic properties

1. Thin-substrate model

The thin-substrate model consists of the thin-film equation (14) with the surface temperature obtained under the thin-substrate assumption:

\[ \partial_t h = -\frac{S}{3} \partial_x (h^3 \partial_x^3 h) + \frac{M}{2} \partial_x (h^2 \partial_x \theta_h), \quad (20a) \]

\[ \theta_h (x, t) = \frac{k_s}{k_s (1 + B h) + B d} \left[ \theta_h + \frac{1}{k_s} I (1 - R(h)) e^{-a_d d} \frac{1 + a_d d}{a_x} \right] \equiv \Theta_h(h), \quad (20b) \]

where Eq. (20b) is obtained by substituting Eq. (18b) into Eq. (13e). When \( d = 0 \),

\[ \Theta_h(h) = \frac{\theta_h}{1 + B h}. \]

As \( d \to \infty \),

\[ \Theta_h(h) \to \frac{I}{B} (1 - R(h)). \]

We introduce a solution given by

\[ h(x, t) = \tilde{h} + \tilde{h} e^{i \omega t + i \beta x}, \quad (21) \]

where \( \tilde{h} = O(1) \), is the steady solution, \( \tilde{h} = O(\delta) \), in which \( 0 < \delta \ll 1 \), is the perturbation amplitude, and \( \omega \) and \( \beta \) are the growth rate and wavenumber of the perturbation, respectively. Substituting Eq. (21) into Eq. (20) and linearizing the result with respect to the perturbation yields the following linear dispersion relation:

\[ \omega = -\frac{S}{3} \tilde{h}^2 \beta^2 \left( \beta^2 + \frac{3M}{2S} \frac{1}{\tilde{h}} \frac{d\Theta_h}{dh} \bigg|_{h=\tilde{h}} \right), \quad (22a) \]
Typical relations between the growth rate $\omega$ and wavenumber $\beta$ are shown in Fig. 2. If $d\Theta_b/dh|_{h=h_b} > 0$, the steady solution is stable because $\omega < 0$ for $\beta > 0$. On the other hand, if $d\Theta_b/dh|_{h=h_b} < 0$, the solution is unstable because $\omega > 0$ in the following range:

$$0 < \beta < \beta_c = \sqrt{-\frac{3M}{2S} \frac{1}{h} \frac{d\Theta_b}{dh}|_{h=h_b}}.$$  \hspace{1cm} (23)

where $\beta_c$ is referred to as the cutoff wavenumber, and $\beta_{\text{max}} = \beta_c / \sqrt{2}$, which gives the maximum growth rate $\omega_{\text{max}}$, is referred to as the most unstable wavenumber. If $d\Theta_b/dh|_{h=h_b} = 0$, the solution is marginally stable ($\beta_c = 0$). When the bottom of the substrate is maintained at a higher (or lower) temperature than the ambient gas, i.e., $\theta_b > 0$ (or $\theta_b < 0$), the first term on the right-hand side of Eq. (22b) contributes to destabilization (or stabilization). When the bottom is maintained at the same temperature as the ambient gas, i.e., $\theta_b = 0$, Eq. (22b) is reduced to

$$d\Theta_b/\Theta_h|_{h=h_b} = -\frac{I}{a_s} \left[ k_s B (1 - R(h)) \right] \left[ k_s (1 + B h) + B d \right] + \frac{dR}{dh}|_{h=h_b}.$$ \hspace{1cm} (24)

The set of coefficients outside the brackets on the right-hand side of Eq. (24), which includes the negative sign, is always negative, and the first term inside the brackets is always positive. Therefore, it is found that

$$d\Theta_b/\Theta_h|_{h=h_b} \geq 0 \Rightarrow \frac{dR}{dh}|_{h=h_b} < 0, \quad d\Theta_b/\Theta_h|_{h=h_b} < 0 \Leftrightarrow \frac{dR}{dh}|_{h=h_b} \geq 0.$$ \hspace{1cm} (25)

2. Generalized model

The generalized model that allows for a substrate of arbitrary thickness consists of the thin-film equation (14), the surface temperature (13e), the heat equation (19a), and the boundary conditions...
(19b) and (19c):

\[
\partial_t h = - \frac{S}{3} \partial_t (h^3 \partial_z^3 h) + \frac{M}{2} \partial_t (h^2 \partial_z \theta), \tag{26a}
\]

\[
\theta_h(x, t) = \frac{\theta_h(x, 0, t)}{1 + Bh}, \tag{26b}
\]

\[
\partial_t \theta_s = K (\partial_x^2 + \partial_Z^2) \theta_s + \frac{K}{\epsilon k_s} A_s I (1 - R(h)) e^{A_s Z}, \tag{26c}
\]

\[
\partial_Z \theta_s \bigg|_{Z=0} = - \frac{B \theta_s(x, 0, t)}{\epsilon k_s (1 + Bh)}, \tag{26d}
\]

\[
\theta_s(x, -D, t) = \theta_b, \tag{26e}
\]

where Eq. (26b) is obtained by substituting the first equation in Eq. (19b) into Eq. (13e), and Eq. (26d) is obtained from Eq. (19b) by using Eq. (13d).

In addition to Eq. (21), we introduce solutions given by

\[
\theta_h(x, t) = \bar{\theta}_h + \hat{\theta}_h e^{\omega t + i\beta x}, \tag{27a}
\]

\[
\theta_s(x, Z, t) = \bar{\theta}_s(Z) + \hat{\theta}_s(Z) e^{\omega t + i\beta x}, \tag{27b}
\]

where \(\bar{\theta}_h\) and \(\bar{\theta}_s(Z)\), which are \(O(1)\), are the steady solutions, and \(\hat{\theta}_h\) and \(\hat{\theta}_s(Z)\), which are \(O(\delta)\), are the perturbation amplitudes. Substituting Eqs. (21) and (27a) into Eq. (26a) and linearizing the result with respect to the perturbations yields the following linear dispersion relation:

\[
\omega = - \frac{S}{3} h^3 \beta^4 - \frac{M}{2} h^2 \frac{\partial h}{h} \beta^2. \tag{28}
\]

Substituting Eqs. (21) and (27) into Eq. (26b), we obtain the following relations at \(O(1)\) and \(O(\delta)\):

\[
O(1) : \bar{\theta}_h = \frac{\bar{\theta}_h(0)}{1 + Bh}, \tag{29a}
\]

\[
O(\delta) : \theta_h = \frac{1}{1 + Bh} \left( \bar{\theta}_h(0) - \frac{B \bar{\theta}_h(0)}{1 + Bh} \hat{h} \right). \tag{29b}
\]

Substituting Eqs. (21) and (27b) into Eq. (26c), we obtain the following equations:

\[
O(1) : \frac{d^2 \bar{\theta}_s}{dZ^2} = - \frac{1}{\epsilon k_s} A_s I (1 - R(h)) e^{A_s Z}, \tag{30a}
\]

\[
O(\delta) : \frac{d^2 \hat{\theta}_s}{dZ^2} - \eta^2 \hat{\theta}_s = \frac{1}{\epsilon k_s} A_s I \frac{dR}{dh} \bigg|_{h=\hat{h}} \hat{h} e^{A_s Z}, \tag{30b}
\]

where

\[
\eta(\omega, \beta) = \sqrt{\frac{\omega}{K} + \beta^2}. \tag{31}
\]
In addition, substituting Eqs. (21) and (27b) into Eq. (26d), we obtain the following boundary conditions:

\[ O(1) : \left. \frac{d\hat{\theta}_s}{dZ} \right|_{Z=0} = -\frac{B\bar{\theta}_s(0)}{\epsilon k_s(1+B\hat{h})}, \]  

\[ (32a) \]

\[ O(\delta) : \left. \frac{d\hat{\theta}_s}{dZ} \right|_{Z=0} = -\frac{B}{\epsilon k_s(1+B\hat{h})}\left( \hat{\theta}_s(0) - \frac{B\bar{\theta}_s(0)}{1+B\hat{h}} \right). \]

\[ (32b) \]

On \( Z = -D \), the following conditions hold at \( O(1) \) and \( O(\delta) \):

\[ O(1) : \bar{\theta}_s(-D) = \theta_b, \]

\[ (33a) \]

\[ O(\delta) : \hat{\theta}_s(-D) = 0. \]

\[ (33b) \]

Solving Eq. (30a) under the boundary conditions (32a) and (33a) yields

\[ \bar{\theta}_s(Z) = \theta_b + \frac{1}{\epsilon k_s} I(1 - R(\hat{h}))(\frac{e^{-A_s D} - e^{A_s Z}}{A_s} + Z + D) - \frac{B\bar{\theta}_s(0)}{\epsilon k_s(1+B\hat{h})}(Z + D), \]

\[ (34a) \]

with

\[ \bar{\theta}_s(0) = \frac{\epsilon k_s(1+B\hat{h})}{\epsilon k_s(1+Bh) + BD}\left[ \theta_b + \frac{1}{\epsilon k_s} I(1 - R(\hat{h}))\frac{e^{-A_s D} - 1 + A_s D}{A_s} \right]. \]

\[ (34b) \]

Solving Eq. (30b) under the boundary conditions (32b) and (33b) yields

\[ \hat{\theta}_s(Z) = \frac{1}{\epsilon k_s A_s I} \left. \frac{dR}{dh} \right|_{h=\hat{h}} \]

\[ \times \frac{1}{A_s^2 - \eta^2} \left( e^{A_s Z} - \frac{A_s \sinh(\eta(Z + D))}{\eta \cosh(\eta D)} - e^{-A_s D} \frac{\cosh(\eta Z)}{\cosh(\eta D)} \right) \hat{h} \]

\[ \times -\frac{B}{\epsilon k_s(1+B\hat{h})} \frac{\sinh(\eta(Z + D))}{\eta \cosh(\eta D)} \left( \hat{\theta}_s(0) - \frac{B\bar{\theta}_s(0)}{1+B\hat{h}} \right) \]

\[ \hat{\theta}_s(0) = F(\eta(\omega, \beta))\hat{h}, \]

\[ (35a) \]

where

\[ F(\eta(\omega, \beta)) = \frac{F_N(\eta(\omega, \beta))}{F_D(\eta(\omega, \beta))}. \]

\[ (35b) \]

\[ F_N(\eta(\omega, \beta)) = A_s I \left. \frac{dR}{dh} \right|_{h=\hat{h}} \frac{1}{A_s^2 - \eta^2} \left( \eta - A_s \tanh(\eta D) - e^{-A_s D} \frac{\eta}{\cosh(\eta D)} \right) \]

\[ \times + \left( \frac{B}{1+B\hat{h}} \right)^2 \hat{\theta}_s(0) \tanh(\eta D), \]

\[ (35d) \]

\[ F_D(\eta(\omega, \beta)) = \epsilon k_s \eta + \frac{B}{1+B\hat{h}} \tanh(\eta D). \]

\[ (35e) \]
When \( D = 0 \),
\[
\tilde{\theta}_s(0) = \theta_b,
\]
\[
\hat{\theta}_b(0) = 0 \quad \left( F(\eta(\omega, \beta)) = 0 \right).
\]
As \( D \to \infty \),
\[
\tilde{\theta}_s(0) \to \frac{1 + B\bar{h}}{B} I(1 - R(\bar{h})),
\]
\[
F_N(\eta(\omega, \beta)) \to -A_i I \frac{dR}{dh} \bigg|_{h=\bar{h}} A_s + \eta + \left( \frac{B}{1 + B\bar{h}} \right)^2 \tilde{\theta}_s(0),
\]
\[
F_D(\eta(\omega, \beta)) \to \epsilon k_s \eta + \frac{B}{1 + B\bar{h}}.
\]
Substituting Eq. (35b) into Eq. (29b) and the result into Eq. (28) yields
\[
\omega = -\frac{S}{3} h^3 \beta^4 - \frac{M}{2} \frac{\bar{h}^2}{1 + B\bar{h}} F(\eta(\omega, \beta)) - \frac{B \tilde{\theta}_s(0)}{1 + B\bar{h}} \beta^2.
\]
This does not give the growth rate \( \omega \) as an explicit function of the wavenumber \( \beta \), and, therefore, specific relations between \( \omega \) and \( \beta \) should be calculated numerically, except that it can be analytically proven that \( \omega \to 0 \) and \( d\omega/d\beta \to 0 \) as \( \beta \to 0 \) (see the Appendix for details).

**B. Model comparison and stability evaluation**

Here we assume that the gas, liquid, and solid are air, polydimethylsiloxane, and amorphous carbon, respectively. We also assume the reference temperature \( \tilde{\theta}_\infty \) = 298 K and the light wavelength \( \lambda = 632.8 \) nm. The corresponding physical properties are listed in Table I. Nondimensional parameters obtained from the dimensional quantities are listed in Table II, in which \( \epsilon \) is set to an appropriate value, the reference film thickness \( h_0 \) is determined from \( \chi = 2\pi \), i.e., \( h_0 = \lambda/(2n_1) \),

**TABLE I. Physical properties at 298 K, where the optical constants correspond to those at \( \lambda = 632.8 \) nm. For details, see Ref. 31 and references therein.**

<table>
<thead>
<tr>
<th></th>
<th>( \text{Gas (air)} )</th>
<th>( \text{Liquid (polydimethylsiloxane)} )</th>
<th>( \text{Solid (amorphous carbon)} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \alpha )</td>
<td>5 \text{ W/(m}^2\cdot\text{K)}</td>
<td>( \mu/\rho )</td>
<td>( k_s )</td>
</tr>
<tr>
<td>( n_0 )</td>
<td>1.0</td>
<td>2.0 \times 10^{\text{-6}} \text{ m}^2/\text{s}</td>
<td>1.08</td>
</tr>
<tr>
<td>( \rho )</td>
<td>8.68 \times 10^2 \text{ kg/m}^3</td>
<td>0.108 \text{ W/(m} \cdot \text{K)}</td>
<td>1.08</td>
</tr>
<tr>
<td>( \kappa )</td>
<td>9.19 \times 10^{\text{-8}} \text{ m}^2/\text{s}</td>
<td>1.74 \times 10^{\text{-2}} \text{ N/m}</td>
<td>1.08</td>
</tr>
<tr>
<td>( \gamma_\infty )</td>
<td>6.7 \times 10^{\text{-5}} \text{ N/(m} \cdot \text{K)}</td>
<td>1.3902</td>
<td></td>
</tr>
<tr>
<td>( \gamma' )</td>
<td>\text{Refractive index}</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( k )</td>
<td>0.74</td>
<td>( n_1 )</td>
<td>( k_2 )</td>
</tr>
<tr>
<td>( \kappa_s )</td>
<td>9.19 \times 10^{\text{-6}} \text{ m}^2/\text{s}</td>
<td>2.43</td>
<td></td>
</tr>
<tr>
<td>( \epsilon )</td>
<td>1.0</td>
<td>0.74</td>
<td></td>
</tr>
</tbody>
</table>
TABLE II. Nondimensional parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film parameter (aspect ratio)</td>
<td>ϵ</td>
</tr>
<tr>
<td>Marangoni number</td>
<td>M</td>
</tr>
<tr>
<td>Inverse capillary number</td>
<td>S</td>
</tr>
<tr>
<td>Solid-to-liquid thermal conductivity ratio</td>
<td>k_s</td>
</tr>
<tr>
<td>Nondimensional thermal diffusivity of the solid</td>
<td>K</td>
</tr>
<tr>
<td>Biot number</td>
<td>B</td>
</tr>
<tr>
<td>Nondimensional absorption coefficients of the solid</td>
<td>A_s</td>
</tr>
<tr>
<td>Other optical parameters</td>
<td>χ, C₁, C₂, C₃, C₄</td>
</tr>
</tbody>
</table>

and the reference velocity \( U \) is determined from \( M = 1 \), i.e., \( U = \epsilon \tilde{\theta}_0 \gamma'/\mu \). In addition, we assume that the bottom of the substrate is maintained at the same temperature as the ambient gas, i.e., \( \theta_b = 0 \).

The film thickness dependence of the stability obtained from the thin-substrate model for \( I = 0.001 \) and \( d = 1 \) is shown in Fig. 3. The energy reflectance \( R \), which is given by Eq. (16), varies periodically with the film thickness \( h \) due to optical interference, and, consequently, the free surface temperature \( \Theta_h \), which is given by Eq. (20b), involves periodic variation with \( h \). As shown in Sec. III A 1, the stability is determined by \( d\Theta_h/dh |_{h=\bar{h}} \). Therefore, stable and unstable thickness regions appear alternately, as shown in Fig. 3. Similar stability characteristics are observed in Fig. 4, which shows the film thickness dependence of the stability obtained from the generalized model for \( I = 0.001 \) and \( D = 0.1 \) (i.e., \( d = 100 \)). Figure 5 shows the substrate thickness dependence of the dispersion relation for an unstable case. The results obtained from the two models agree.

FIG. 3. Film thickness dependence of the stability obtained from the thin-substrate model for \( I = 0.001 \) and \( d = 1 \). (a) Free surface temperature \( \Theta_h \), (b) energy reflectance \( R \), (c) cutoff and most unstable wavenumbers, \( \beta_c \) and \( \beta_{\text{max}} \), and (d) maximum growth rate \( \omega_{\text{max}} \) as a function of the film thickness \( h \), where the regions indicated by S and U, which are bounded by vertical dotted lines, correspond to stable and unstable regions, respectively. (e) Growth rate \( \omega \) as a function of the wavenumber \( \beta \) for various values of \( \bar{h} \).
well with each other in a small-substrate-thickness region, as implied by the assumption underlying
the thin-substrate model. However, the difference between these results increases with the substrate
thickness. In the case of the thin-substrate model, both $\beta_c$ and $\beta_{\text{max}}(=\beta_c/\sqrt{2})$ increase monotonically
with the substrate thickness. In contrast, in the case of the generalized model, while $\beta_c$ increases
monotonically with the substrate thickness, $\beta_{\text{max}}$, which is not always equal to $\beta_c/\sqrt{2}$, has a local
maximum at $D = 0.0334$ and a local minimum at $D = 0.2110$. In both cases, $\beta_c$, $\beta_{\text{max}}$, and $\omega_{\text{max}}$
converge to finite values as $d$ or $D \to \infty$. However, these limiting values obtained from
the thin-substrate model, $\beta_c \to 1.22 \times 10^4$, $\beta_{\text{max}} \to 8.64 \times 10^3$, and $\omega_{\text{max}} \to 4.45 \times 10^8$, are much larger than those obtained from the generalized model, $\beta_c \to 67.9$, $\beta_{\text{max}} \to 45.0$, and

---

**FIG. 4.** Film thickness dependence of the stability obtained from the generalized model for $I = 0.001$ and $D = 0.1$ (i.e., $d = 100$). (a) Energy reflectance $R$, (b) cutoff and most unstable wavenumbers, $\beta_c$ and $\beta_{\text{max}}$, and (c) maximum growth rate $\omega_{\text{max}}$ as a function of the film thickness $\bar{h}$, where the regions indicated by S and U, which are bounded by vertical dotted lines, correspond to stable and unstable regions, respectively. (d) Growth rate $\omega$ as a function of the wavenumber $\beta$ for various values of $\bar{h}$.

---

**FIG. 5.** Substrate thickness dependence of the dispersion relation for $\bar{h} = 0.65$ and $I = 0.001$. (a) Cutoff and most unstable wavenumbers, $\beta_c$ and $\beta_{\text{max}}$, and (b) maximum growth rate $\omega_{\text{max}}$ as a function of the substrate thickness, where $\beta_c$, $\beta_{\text{max}}$, and $\omega_{\text{max}}$ obtained from the generalized model converge to the values indicated by the dotted lines as $D \to \infty$. The horizontal scales, $D$ and $d$, coincide with each other by conversion to the dimensional quantity $\tilde{d}$. (c) Growth rate $\omega$ as a function of the wavenumber $\beta$, as obtained from the generalized model for various values of $D$. The dashed curve in (c) indicates the trajectory of $(\beta_{\text{max}}, \omega_{\text{max}})$. 
FIG. 6. Film thickness dependence of the stability obtained from the generalized model, where all parameter values are the same as in Fig. 4, but $R(h) \equiv 0$. (a) Cutoff and most unstable wavenumbers, $\beta_c$ and $\beta_{\text{max}}$, and (b) maximum growth rate $\omega_{\text{max}}$ as a function of the film thickness $\bar{h}$. (c) Growth rate $\omega$ as a function of the wavenumber $\beta$ for various values of $\bar{h}$.

$\omega_{\text{max}} \to 0.609$, respectively. The major difference between the two models is that lateral thermal diffusion, which suppresses the instability, is taken into account in the generalized model but not in the thin-substrate model. Such diffusion effects do not have much importance when the substrate thickness is sufficiently small (i.e., $d = O(1)$ or $D = O(\epsilon)$), but the importance increases with the thickness.

In order to clarify the effect of optical interference on the stability, we consider the case in which $R(h) \equiv 0$ for comparison. In such a case, optical interference is absent, and heat generation in the substrate is independent of $x$. The film thickness dependence of the stability obtained from the generalized model is shown in Fig. 6, in which the parameters are set to be the same as in Fig. 4, but the range of graphs is different. In this case, the solution is unstable for any value of $\bar{h}$, where $\beta_c$ and $\beta_{\text{max}}$ decrease monotonically with $\bar{h}$, while $\omega_{\text{max}}$ increases monotonically with $\bar{h}$. Comparison of Figs. 4 and 6 implies that $dR/dh|_{h=\bar{h}} > 0$ (or $dR/dh|_{h=\bar{h}} < 0$) has a destabilizing (or stabilizing) effect. In addition, the dispersion relation for $\bar{h}$ values that satisfy $dR/dh|_{h=\bar{h}} = 0$ in the case of $R(h) \neq 0$ is shown in Fig. 7, in which the curves for the case of $R(h) \neq 0$ are quantitatively, but not qualitatively, different from those for the case of $R(h) \equiv 0$. These results indicate that the stability is strongly affected by $dR/dh|_{h=\bar{h}}$ rather than $R(\bar{h})$ itself, which agrees qualitatively with the results obtained from the thin-substrate model (see Sec. III A 1).

FIG. 7. Growth rate $\omega$ as a function of the wavenumber $\beta$, as obtained from the generalized model for $I = 0.001$, $D = 0.1$, and various values of $\bar{h}$ that satisfies $dR/dh|_{h=\bar{h}} = 0$ in the case of $R(h) \neq 0$. The solid and dotted curves are for the cases of $R(h) \neq 0$ and $R(h) \equiv 0$, respectively. The inset shows the energy reflectance $R$ as a function of the film thickness $\bar{h}$, where $h_A$ and $h_C$ are the thicknesses corresponding to minima of the curve, and $h_B$ and $h_D$ are the thicknesses corresponding to maxima of the curve.
The dispersion relation also varies depending on the incident light intensity $I$. Figure 8 shows the incident light intensity dependence of the dispersion relation for an unstable case, where $\beta_c$, $\beta_{\text{max}}$, and $\omega_{\text{max}}$ increase monotonically with $I$.

IV. SUMMARY AND CONCLUSIONS

We have addressed the problem of the stability of uniformly irradiated transparent liquid films on absorbing solid substrates. Under such circumstances, the energy reflectance varies periodically with the film thickness due to optical interference, and the film stability then varies depending on the film thickness. The stability is strongly affected by the first derivative of the reflectance with respect to the film thickness rather than the reflectance itself.

We have performed numerical simulations based on the generalized model for $D = O(1)$ and have briefly mentioned that linear stability analysis based on the thin-substrate model can confirm the numerical results qualitatively. However, the dominant wavenumber obtained from the simplified analysis is much larger than that obtained from the simulations because lateral thermal diffusion, which suppresses the instability, is ignored in the simplified model. On the other hand, the results of the analysis based on the generalized model are naturally comparable to the numerical results. Therefore, we can conclude that the generalized model is necessary for quantitative evaluation of the stability in the case of substrates of moderate to large thickness, whereas the thin-substrate model is useful for clarifying the stability qualitatively and intuitively.

We have found that the instability and its characteristic scales, such as the most unstable and cutoff wavelengths, of uniformly irradiated transparent films can be controlled by the film and substrate thicknesses and the incident light intensity. In applications that require the stability of a film, e.g., radiation curing of coatings, the destabilization of the film can be prevented by selecting an appropriate film thickness. The destabilization can also be prevented by selecting an appropriate light source because the stable thickness range, in which the reflectance decreases with the film thickness, varies depending on the light wavelength. Another application is to microfluidics, where the instability can be utilized to drive microflow. In addition to the present results, we have obtained numerical simulation results for the same problem, which show that a small perturbation around an unstable uniform state evolves into a phase separation or periodic wavy pattern. These findings imply that uniform irradiation can produce patterned films on substrates that have no channels, no electrodes, and no chemical preparations. The nonlinear evolution will be discussed in more detail elsewhere.
APPENDIX: NUMERICAL METHODS FOR ANALYSIS BASED ON THE GENERALIZED MODEL

1. Growth rate for a given wavenumber

In order to calculate $\omega$ for $\beta > 0$, which is given as a parameter, from Eq. (36), we consider the following equation:

$$ W_1(\omega; \beta) = 0, $$  \hspace{1cm} (A1a)

where

$$ W_1(\omega; \beta) = \omega + \frac{S}{3} \bar{h}^3 \beta^4 + \frac{M}{2} \frac{\bar{h}^2}{1 + B \bar{h}} \left( F(\eta(\omega, \beta)) - \frac{B \bar{h}_s(0)}{1 + B \bar{h}} \right) \beta^2. $$  \hspace{1cm} (A1b)

We solve Eq. (A1a) for $\omega$ using Newton’s method:

$$ \omega^{(n+1)} = \omega^{(n)} - \left( \frac{\partial W_1}{\partial \omega} \bigg|_{\omega=\omega^{(n)}} \right)^{-1} W_1(\omega^{(n)}; \beta), $$  \hspace{1cm} (A2)

where the superscript is the iteration number.

2. Cutoff wavenumber

The following equality holds for $\beta = \beta_c > 0$:

$$ \omega = 0. $$  \hspace{1cm} (A3)

Therefore, the equation to be solved is obtained as

$$ W_2(\beta) = 0, $$  \hspace{1cm} (A4a)

where

$$ W_2(\beta) \equiv W_1(0; \beta) = \frac{S}{3} \bar{h}^3 \beta^4 + \frac{M}{2} \frac{\bar{h}^2}{1 + B \bar{h}} \left( F(\eta(0, \beta)) - \frac{B \bar{h}_s(0)}{1 + B \bar{h}} \right) \beta^2. $$  \hspace{1cm} (A4b)

We solve Eq. (A4a) for $\beta$ using Newton’s method:

$$ \beta^{(n+1)} = \beta^{(n)} - \left( \frac{dW_2}{d\beta} \bigg|_{\beta=\beta^{(n)}} \right)^{-1} W_2(\beta^{(n)}). $$  \hspace{1cm} (A5)

Note that in all cases, $\omega \to 0$ as $\beta \to 0$ because $W_2(\beta) \to 0$ as $\beta \to 0$.

3. Most unstable wavenumber

The following equality holds for $\beta = \beta_{\text{max}} > 0$:

$$ \frac{d\omega}{d\beta} = 0, $$  \hspace{1cm} (A6)

where $\omega = \omega(\beta)$. From Eq. (36), we obtain

$$ \frac{d\omega}{d\beta} = -\frac{W_3(\beta)}{1 + \frac{M}{4K} \frac{\bar{h}^2}{1 + B \bar{h}} \frac{\beta^2}{\eta} \frac{dF(\eta)}{d\eta}}, $$  \hspace{1cm} (A7a)

with

$$ W_3(\beta) = \frac{4}{3} S \bar{h}^3 \beta^3 + \frac{M}{2} \frac{\bar{h}^2}{1 + B \bar{h}} \left[ 2\beta \left( F(\eta) - \frac{B \bar{h}_s(0)}{1 + B \bar{h}} \right) + \frac{\beta^3}{\eta} \frac{dF(\eta)}{d\eta} \right], $$  \hspace{1cm} (A7b)

where $\eta = \eta(\omega, \beta)$. From Eqs. (A6) and (A7a), the equation to be solved is obtained as

$$ W_3(\beta) = 0. $$  \hspace{1cm} (A8)
We solve Eq. (A8) for $\beta$ using Newton’s method:

$$\beta^{(n+1)} = \beta^{(n)} - \left( \frac{dW_3}{d\beta} \right)_{\beta=\beta^{(n)}}^{-1} W_3(\beta^{(n)}),$$  (A9)

where $\omega(\beta^{(n)})$ in $W_3(\beta^{(n)})$ and $dW_3/d\beta|_{\beta=\beta^{(n)}}$ is updated at each iteration step by means of the method described in Subsection 1 of the Appendix. Note that in all cases, $d\omega/d\beta \to 0$ as $\beta \to 0$ because $W_3(\beta) \to 0$ as $\beta \to 0$.