Control of Evaporatively Driven Instabilities of Thin Liquid Films

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In the process of drying, thin volatile liquid films often undergo a long-wavelength instability leading to nonuniformities or formation of dry spots, with the strength of the instability increasing with the volatility and temperature of the liquid. Perhaps counterintuitively, this evaporative instability can be actively suppressed by an appropriate heating procedure. We use linear stability analysis of the lubrication approximation to show that spatially nonuniform time-dependent radiative heating can indeed have a stabilizing effect. Evaporation is shown to introduce several fundamentally new aspects into the control problem for heated liquid films, compared to the relatively well studied case of thermal convection. The control problem becomes especially interesting and non-trivial for mixtures and solutions with negative Marangoni numbers due to a peculiar cancellation effect rendering the system insensitive to temperature control at a certain wavelength. It is shown that taking the time-dependence of the mean thickness of the film into account is necessary to circumvent this insensitivity.

I. INTRODUCTION

Evaporating liquid layers are found in many areas of science and technology, so their practical significance is rather high. Examples include evaporative cooling and numerous coating applications. In the latter a substrate is coated with a layer of solution which, upon the evaporation of solvent, leaves a layer of solute on the surface of the substrate. The main difference between various coating techniques is in the way the initial liquid coating is produced. For instance, dip-coating technique is used for optical fiber coating [1], and (anti)reflective optical coatings of lenses and mirrors. Similar techniques are used to produce hydrophobic [2] and hydrophilic coatings. Spray coating is used to produce sol-gel coatings of TV screens [3] and, more routinely, for painting. Spin coating [4] which was originally developed for microelectronics applications has also found numerous applications in the optical industry.

One of the critical issues related to the quality of produced coatings is stability of the liquid layer during evaporation. For instance in spin-coating applications, striations (radially oriented lines of thickness variation) arise due to the thermal and solutal effects induced by evaporation [5]. These effects are also responsible for the rupturing instability [6] which results in the formation of a pattern of dry spots [7], a phenomenon often referred to as reticulation. The ultimate fate of the linearly unstable liquid film, with or without evaporation, is eventually determined by the properties of the substrate: if it is nonwetting, the film will generally rupture [8], otherwise it may remain continuous and nonuniform.

In most circumstances dewetting is a highly undesirable effect, so understanding its mechanisms and learning to control it is very important. The process of dewetting is intrinsically nonlinear, so it cannot be adequately described by the linear theory. This means that either numerical [8,9] or approximate analytical [10] solutions have to be obtained. However, neither approach is suitable for control purposes. The more practical approach is to prevent the instability from forming *at the linear stage*, rather than suppress the rupturing itself. This approach has an additional advantage that, if the liquid layer is uniform at all times during the evaporation, the produced coating will be uniform as well.

Several different approaches have been suggested to enforce control of evaporating liquid films. All of them are properly classified as *passive*. The examples include the use of surfactants [11] and internal (volumetric) heating of the liquid layer [12,13]. Albeit the passive approaches are relatively simple, their applicability is very restricted (e.g., surfactants may contaminate the coating). *Active*, or *feedback*, control schemes are generally significantly more flexible and efficient. Although none currently exist, several approaches designed to suppress Marangoni-Bénard convection in thin nonvolatile films can be generalized for the case with evaporation. The underlying idea of the latter approaches is to exploit the same physical mechanism that leads to instability, in this case the thermocapillary effect: a spatially distributed thermal perturbation is applied to either the bottom [14] or the top [15] interface, which opposes the spontaneously produced disturbances, an approach sometimes referred to as *noise-cancellation*.

When evaporation is present, thermocapillarity represents only one of several destabilizing mechanisms: vapor recoil (normal pressure on the liquid-gas interface due to non-equilibrium evaporation), differential evaporation (dependence of the evaporation rate on the thickness of the film) and, sometimes, solutocapillarity all contribute to the development of the interfacial instability. There might also be additional destabilizing mechanisms not related to evaporation, e.g., van der Waals interactions, gravity, and curvature effects. Since all mechanisms in the former group are evaporatively driven, it should come as no surprise that all of them can be altered (and hence controlled) by varying the evaporation rate, which is most easily accomplished by applying thermal perturbations. The evaporative instability, however, is much more complicated than the thermally driven instability leading to Marangoni-Bénard convection: the disturbances produced by several different mechanisms cannot be simultaneously canceled by applying perturbations to a single parameter of the system, such as the interfacial temperature of the liquid layer. An additional challenge posed by the presence of evaporation is time-dependence of the mean thickness of the film.

In this paper we use linear stability analysis to design a control scheme which uses distributed thermal perturbations to suppress the instability by exploiting a *combination* of different physical effects. The thermal perturbations will be applied by varying the intensity of electromagnetic radiation absorbed by the liquid film and the substrate. The intensity profile will be calculated and adjusted in real time based on the measurements of either the local thickness of the liquid layer or the local interfacial temperature, which is related to the local thickness. The distributed nature of the feedback requires sensing and thermal actuation capabilities with the spatial resolution comparable to the scale of characteristic disturbances, which can be rather small (e.g., submillimeter) for thin films. Such capabilities can be implemented rather easily using commercially available optical components such as CCD arrays in video cameras and MEMS mirrors in video projectors.

The outline of the paper is as follows. We derive the nonlinear lubrication equations describing the dynamics in the limiting case of thin substrate and perform the linear stability analysis in section II. We compare different ways of perturbing the system by radiative heating from the control standpoint in section III and derive the equations describing the effect of perturbations on the dynamics in section IV. The modifications of these equations required by relaxing the restriction on the thickness of substrate are discussed in section V. Section VI contains the analysis of the feedback control problem and discusses the new features introduced by the presence of competing destabilization mechanisms. The spatial structure of the distributed feedback is discussed in section VII and section VIII presents summary and conclusions.

II. EVOLUTION EQUATIONS

The ability to apply thermal perturbations is essential for suppression of the evaporative instability. Evaporative cooling provides a natural way to lower the temperature of the liquid, Alternatively, the liquid can be heated by electromagnetic radiation absorbed by either the liquid itself or the substrate. If the film is thin, it will not absorb radiation effectively, so it will be difficult to change its temperature significantly by modulating the intensity of radiation. A more flexible approach is to use the substrate as the absorbing medium. This approach requires that heat conduction in the substrate is modeled properly. In particular, the substrate should have finite conductivity and finite thickness, h_s. Therefore, we have to consider a three-layer system depicted in Fig. 1. The liquid layer has a free upper boundary where it interfaces with a gas layer partially saturated with the vapor. We will orient our coordinate system such that the z-axis is orthogonal to the substrate's upper boundary, with $z = 0$ at the bottom of the liquid layer. The initial mean height of the liquid layer is h_l, and its instantaneous height is $h(x, t)$. (For simplicity of notation we ignore the variation in the y-direction – y-dependence can be trivially restored – so the problem becomes 2-dimensional.)

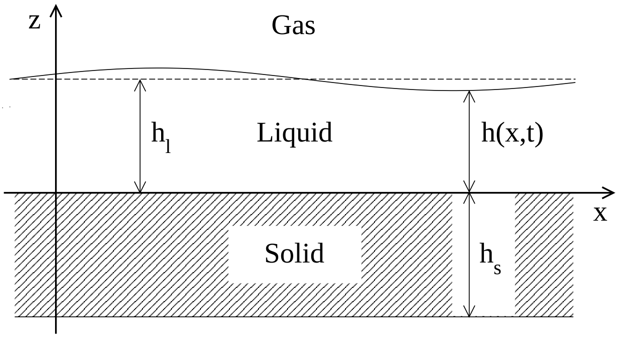

FIG. 1. Evaporating liquid film on a solid substrate.

The combined three-layer system is described by the Navier-Stokes equation

$$\rho_l(\partial_t \mathbf{v} + (\mathbf{v} \cdot \nabla)\mathbf{v}) = -\nabla p + \mu \nabla^2 \mathbf{v} + \rho_l \mathbf{g} \tag{1}$$

augmented by the incompressibility condition $\nabla \cdot \mathbf{v} = 0$, the energy equation

$$\rho_l c_l(\partial_t \theta_l + (\mathbf{v} \cdot \nabla)\theta_l) = \kappa_l \nabla^2 \theta_l, \tag{2}$$

for the liquid layer, and the heat equation for the solid layer

$$\rho_s c_s \partial_t \theta_s = \kappa_s \nabla^2 \theta_s, \tag{3}$$

where ρ_l, κ_l, c_l, θ_l and ρ_s, κ_s, c_s, θ_s are the densities, thermal conductivities, heat capacities, and temperatures of the liquid and solid layer, respectively, μ, p, and $\mathbf{v}$ are the viscosity, pressure, and velocity of the liquid, and $\mathbf{g}$ – the gravitational acceleration in the vertical (negative z) direction. For simplicity we assume that, if the liquid is a mixture or solution with two components, none of its physical properties (viscosity, density, surface tension and so on) depend on the relative concentration of the components. This assumption means that the solutal Marangoni effect is absent and we don't need to include an additional evolution equation for the relative concentration [8,9].

The gas layer is not modeled explicitly. Instead its effect is represented by the appropriate boundary conditions at the liquid-gas interface $z = h$, which describe the exchange of mass, momentum, and heat between the layers. We will use the simplified boundary conditions [16] for the energy, stress, and mass balance

$$jL = -\kappa_l \partial_z \theta_l, \tag{4}$$

$$T_{nn} = -k_i\sigma + \frac{j^2}{\rho_g}, \quad T_{nt} = (\mathbf{t}\cdot\nabla)\sigma, \tag{5}$$

$$\partial_t h = -\partial_x \int_0^h v_x dz - \frac{j}{\rho_l}, \tag{6}$$

which are valid when the density, viscosity, and thermal conductivity of the liquid are significantly greater than those of the vapor. L denotes the latent heat of vaporization per unit mass, ρ_g – vapor density, σ – surface tension, and k_i is the curvature of the interface which can be expressed through the local thickness h of the liquid layer

$$k_i = \partial_x^2 h / \sqrt{1 + (\partial_x h)^2}. \tag{7}$$

An additional boundary condition is imposed by a constitutive relationship [16] between the interfacial temperature θ_i and mass flux j

$$\tilde{K} j = \theta_i - \theta_a, \tag{8}$$

where θ_a is the absolute saturation temperature, and $\tilde{K}$ describes the volatility of the liquid. The limit of vanishing volatility, $\tilde{K} \to \infty$, corresponds to the pure Marangoni-Bénard convection.

At the liquid-solid interface $z = 0$ we impose the no-slip boundary condition for the velocity and conditions of continuity for the temperature and heat flux

$$\mathbf{v} = 0, \tag{9}$$

$$\theta_s = \theta_l, \tag{10}$$

$$\kappa_s \partial_z \theta_s = \kappa_l \partial_z \theta_l. \tag{11}$$

Finally, the temperature at the bottom of the solid layer $z = -h_s$ is assumed constant

$$\theta_s = \theta_b. \tag{12}$$

The horizontal velocity profile needed to close the evolution equation (6) for the height h can be determined by solving the Navier-Stokes equation (1) subject to boundary conditions (5) and (9). The solution in the lubrication approximation is obtained in a standard way [17]:

$$\mu v_x = z\partial_x\sigma + \left(\frac{1}{2}z^2 - hz\right)\partial_x \bar{p}, \tag{13}$$

where $\bar{p} = \rho_l g h - k_i\sigma + j^2/\rho_g$ is the modified pressure. We choose to ignore the disjoining pressure terms [7,8] representing the effect of van der Walls forces, because they are only significant for extremely thin (sub-micron scale) films. These terms can be easily included in the following analysis. Combining (6) and (13) one obtains a nonlinear evolution equation

$$\begin{aligned}\partial_t h = &-\frac{j}{\rho_l} - \frac{1}{2\mu}\partial_x(h^2\partial_x\sigma) + \frac{\rho_l g}{3\mu}\partial_x(h^3\partial_x h)\\ &- \frac{\sigma}{3\mu}\partial_x(h^3\partial_x^3 h) + \frac{2}{3\mu\rho_g}\partial_x(h^3 j\partial_x j).\end{aligned} \tag{14}$$

Both the mass flux j and the coefficient of surface tension σ depend on the interfacial temperature θ_i, which can be found by solving (2) and (3) subject to the boundary conditions given above. In the lubrication approximation the left-hand sides of both equations can be neglected, so that in the dimensionless coordinates $X = x/h_l$, $Z = z/h_l$, $T = (\kappa_l/\rho_l c_l h_l^2)t$, and $\Theta = (\theta - \theta_a)/\Delta\theta$ with $\Delta\theta = \theta_b - \theta_a$, both equations reduce to

$$\partial_X^2\Theta + \partial_Z^2\Theta = 0. \tag{15}$$

(Since the mean thickness of the liquid film varies in time due to evaporation, we use its *initial* value h_l as the length scale.) The general solution of (15) is

$$\Theta = \int [c(k)\cosh kZ + s(k)\sinh kZ]\, e^{ikX} dk \tag{16}$$

and simplifies considerably in the limit of thin layers (or long wavelength disturbance), $k \ll H$, $k \ll H_s$, where $H = h/h_l$ and $H_s = h_s/h_l$ are the dimensionless thicknesses of the liquid and solid layer, respectively. Expanding (16) in powers of kZ and retaining the two leading terms yields

$$\Theta = \mathcal{C}(X) + \mathcal{S}(X)Z. \tag{17}$$

Introducing the ratio of thermal resistances of the liquid and solid layer $F = h_s\kappa_l/h_l\kappa_s$, the Biot number $B = h_l L/\kappa_l\tilde{K}$ characterizing heat transfer at the liquid-gas interface, and the dimensionless mass flux $J = (h_l L/\kappa_l\Delta\theta)j$, the boundary conditions (4), (8), (10), (11), and (12) can also be written in dimensionless form:

$$-\partial_Z\Theta_l(H) = J, \tag{18}$$

$$B\Theta_l(H) = J, \tag{19}$$

$$\Theta_l(0) = \Theta_s(0), \tag{20}$$

$$\partial_Z\Theta_s(0) = FH_s^{-1}\partial_Z\Theta_l(0), \tag{21}$$

$$\Theta_s(-H_s) = 1. \tag{22}$$

The particular solution (17) satisfying these boundary conditions gives the temperature of the liquid-gas interface:

$$\Theta_i = \frac{1}{1 + BF + BH}, \tag{23}$$

such that the height uniquely determines the interfacial temperature and vice versa.

Assuming linear dependence of the surface tension coefficient on temperature, upon substitution of (19) the dimensionless version of (14) can be written as

$$\partial_T H = -EB\Theta_i + \frac{M}{2}\partial_X(H^2\partial_X\Theta_i) + \frac{G}{3}\partial_X(H^3\partial_X H) - \frac{1}{3C}\partial_X(H^3\partial_X^3 H) + \frac{2E^2B^2}{3PD}\partial_X(H^3\Theta_i\partial_X\Theta_i), \quad (24)$$

where we have defined the evaporation number $E = c_l\Delta\theta/L$, Marangoni number $M = -\partial_\theta\sigma\Delta\theta\rho_l c_l h_l/\mu\kappa_l$, gravity number $G = g\rho_l^2 c_l h_l^3/\mu\kappa_l$, capillary number $C = \mu\kappa_l/\sigma\rho_l c_l h_l$, Prandtl number $P = \mu c_l/\kappa_l$, and the density ratio for the liquid and its vapor $D = \rho_g/\rho_l$. This equation shows that the interfacial temperature plays a very important role in the dynamics of the liquid layer, entering three of the five terms on the right-hand side. This justifies our control strategy: we can influence the evolution of the film's thickness by making appropriate changes to the interfacial temperature.

Substituting (23) into (24) and linearizing with respect to small deviations about the (time-dependent) mean height of the film $H(X,T) = H_0(T) + H_1(X,T)$ yields two equations, one for the mean H_0

$$\partial_T H_0 = -\frac{E}{K + H_0} \quad (25)$$

and one for the deviation H_1

$$\partial_T H_1 = \frac{E}{(K+H_0)^2}H_1 - \frac{M}{2B}\frac{H_0^2}{(K+H_0)^2}\partial_X^2 H_1 - \frac{2E^2B}{3PD}\frac{H_0^3}{(K+H_0)^3}\partial_X^2 H_1 + \frac{GH_0^3}{3}\partial_X^2 H_1 - \frac{H_0^3}{3C}\partial_X^4 H_1, \quad (26)$$

where we have denoted $K = B^{-1} + F$. The first equation describes the gradual reduction in the mean thickness of the liquid layer with time. It can be integrated [17]

$$H_0(T) = -K + \sqrt{(K+1)^2 - 2ET} \quad (27)$$

and shows that the amount of time it takes for the layer to completely evaporate is

$$T_0 = \frac{2K+1}{2E}. \quad (28)$$

It is conventionally called the disappearance time.

Equation (26) describes the stability of the uniform solution (27). The first term in (26) represents differential evaporation and is always destabilizing. The physical mechanism responsible for differential evaporation is simple. The top of the liquid layer has a lower temperature than the bottom due to evaporative cooling. In the regions where the interface is closer to the substrate the liquid is warmer and hence evaporates more rapidly than in the regions where the interface is further away. This can also be thought of as a result of the evaporation speed-up: according to (27) the intensity of evaporation increases with decreasing thickness of the layer, which is a weakly destabilizing effect. This effect has never been properly considered in the literature, although it becomes dominant at very long wavelengths. The second term describes the effect of thermocapillarity which is destabilizing whenever $\partial_\theta\sigma < 0$ (i.e., for pure liquids, $M > 0$) and stabilizing otherwise (for some mixtures and solutions, $M < 0$): when surface tension decreases with temperature its variation produces tangential stresses which draw liquid from warmer to cooler regions, amplifying the disturbance. The third term represents the effect of vapor recoil [16] and is also destabilizing. Destabilization is due to larger evaporation rates in the warmer regions, leading to a larger number of vapor molecules leaving the surface with a larger momentum (hence quadratic dependence on the evaporation rate), which produces increased normal stress on the interface. The fourth and fifth terms represent the effects of gravity and surface tension, respectively, and are both stabilizing in the geometry considered here. Gravity can also play a destabilizing role, if the liquid layer coats the bottom of the solid layer, rather than the top [18]. In cylindrical geometry, which is appropriate, e.g., for the case of optical fiber coating, the fourth term describes the effect of curvature instead of gravity, with $G = -(h_l/r_0)^2 C^{-1}$, where r_0 is the radius of the fiber. This geometrical effect is always destabilizing.

As we have already mentioned, the first term dominates the evolution for the longest wavelength disturbances, so in the presence of evaporation the uniform solution (27) is always linearly unstable. In the case of pure liquids, the asymptotic state is the same, regardless of the initial conditions or existing spatial nonuniformities in the substrate and so on – all liquid eventually evaporates. In the case of solutions, however, the instability results in a nonuniform layer of solute left on the surface of the substrate once the solvent has evaporated completely. This problem becomes even more severe, if either additional (laterally uniform) heating or a more volatile solvent is used to speed up the drying process: the first three terms in (26) increase with $\Delta\theta$, while the third term also increases with B, i.e., with increasing volatility.

III. RADIATIVE HEATING

Consider now what happens when the system is heated internally to enhance evaporation, e.g., through the absorption of electromagnetic radiation, such as microwaves [19], infrared, or visible light. Depending on the absorption properties of the liquid and the substrate, either both layers or just one of them could be heated by the radiation. Extending the results of Oron [13] who has considered the effect of *spatially uniform* irradiation of a liquid film on a substrate of *infinite* conductivity, we will consider different limiting cases which cover the spectrum of possibilities for substrates of *finite* conductivity and thickness. In addition, we will allow for *spatial* and *temporal* variation of the intensity of radiation, a prerequisite for active control. This variation will be assumed slow compared to the characteristic time scale, such that (15) is valid. Furthermore, in the next few sections we will assume that the substrate is thin, so we can use the approximation (17) instead of the exact solution (16) for both layers. We will lift this restriction and discuss the effect of substrate thickness in section V.

Since, for a given frequency, the fraction of the absorbed radiation does not depend on its intensity, the intensity (power) of a monochromatic radiation decays exponentially with the distance from the interface, a fact expressed by the Bouguer's law [20]

$$q(z) = q(z_0)e^{(z-z_0)/d}, \tag{29}$$

where z_0 is the position of the interface, d is the (frequency dependent) penetration depth, and for simplicity we assume that there is no reflection at the interface. Given the mathematical structure of the lubrication approximation equations we expect that the sole effect of introducing an external heat source will be to change the functional dependence (23) between the thickness of the liquid layer and the temperature at the liquid-gas interface.

A. Volumetric absorption

First, assume that the liquid is optically thin, $d \gg h_l$, and the substrate either optically transparent or reflecting. In this case the absorption and hence heating is essentially uniform across the depth of the liquid layer. Correspondingly, while the temperature distribution inside the substrate still obeys (15), inside the liquid this equation has to be modified by including a source term with uniform density $A^{-1}Q$:

$$\partial_X^2 \Theta_l + \partial_Z^2 \Theta_l = -A^{-1}Q, \tag{30}$$

where $Q = (h_l/\kappa_l \Delta\theta)q$ is the dimensionless power of the incident radiation per unit area and A is the dimensionless absorption length defined as $A = d/h_l$ for a transparent non-reflecting substrate and $A = d/2h_l$ for a reflecting substrate (the radiation traverses the liquid layer twice in the latter case). The solution of (30) in the thin layer limit is

$$\Theta_l = \mathcal{C} + \mathcal{S}Z - (2A)^{-1}QZ^2, \tag{31}$$

so using the standard boundary conditions (18)-(22) we obtain the temperature of the liquid-gas interface in the form

$$\Theta_i = \frac{1 + (2A)^{-1}Q(H^2 + 2FH)}{1 + BF + BH}. \tag{32}$$

This relation shows that the interfacial temperature is a function of the *local* thickness H and *local* intensity Q. The dependence on Q, however, is very weak due to the smallness of the coefficient $(2A)^{-1}$, making this a poor arrangement for the purpose of active control.

Interestingly, the volumetric heating by laterally uniform radiation $Q(X,T) = Q_0$ can stabilize the evaporating liquid film when the heating intensity is sufficiently large. Indeed, substituting (32) into (24) and linearizing, we obtain an equation similar to (26)

$$\begin{aligned}\partial_T H_1 = {} & \frac{E\alpha(H_0)}{(K+H_0)^2}H_1 - \frac{M}{2B}\frac{\alpha(H_0)H_0^2}{(K+H_0)^2}\partial_X^2 H_1 \\ & - \frac{2E^2B}{3PD}\frac{\alpha(H_0)\gamma(H_0)H_0^3}{(K+H_0)^3}\partial_X^2 H_1 \\ & + \frac{GH_0^3}{3}\partial_X^2 H_1 - \frac{H_0^3}{3C}\partial_X^4 H_1,\end{aligned} \tag{33}$$

where

$$\begin{aligned}\alpha(H_0) &= 1 - (2A)^{-1}Q_0\beta(H_0), \\ \beta(H_0) &= 2K(H_0 + F) + H_0^2, \\ \gamma(H_0) &= 1 + (2A)^{-1}Q_0(H_0^2 + 2FH_0).\end{aligned} \tag{34}$$

Since $\gamma(H_0)$ is positive, the effects of differential evaporation, thermocapillarity (for $M > 0$), and vapor recoil all change sign and become stabilizing for negative values of $\alpha(H_0)$, i.e., when

$$Q_0 > \frac{2A}{\beta(H_0)}. \tag{35}$$

The same conclusion has been reached by Oron [13], who ignored differential evaporation and assumed that the substrate was perfectly conducting. It served as the basis for suggesting the use of volumetric heating for passive control.

In reality the situation is more complicated. Even though the radiation absorbed in the liquid layer provides a stabilizing effect, some fraction of it will necessarily be absorbed by the substrate. As we will see in the next section this has a destabilizing effect. Since A has to be large for the absorption in the liquid layer to be relatively uniform across its depth, Q_0 will also have to be large, so substantial power will be dissipated in the substrate, negating the stabilizing effect of the radiation absorbed by the liquid layer. In other words, passive control is likely to work only in a highly idealized situation with the substrate which is either almost perfectly reflecting or perfectly transparent (or perfectly conducting, as was assumed by Oron) and even then it will be quite inefficient energetically. (Intense irradiation may also damage the substrate, limiting the practically achievable values of Q_0.)

B. Absorption at the bottom interface

The problems faced by passive control can be overcome by employing active control, which uses non-uniform heating with intensity changing in response to the deviation of the interface from the flat profile. The active control is most effective when the radiation couples strongly to the interfacial temperature. Since many liquids are transparent in a wide range of frequencies, it is often easiest to change their temperature indirectly by heating the substrate. This approach has not been previously considered in the framework of radiative heating, although the basic idea is not new: heat flux control using heaters embedded in the substrate was suggested in the context of Rayleigh-Bénard convection by Howle [21]. Radiative heating is a vastly superior approach which requires no preparation of the substrate, yet achieves spatial resolution several orders of magnitude higher.

To simplify the discussion, let us assume that the liquid is completely transparent and the substrate is optically thick, $d \ll h_s$, such that essentially all radiation is absorbed at the liquid-solid interface (optically thin substrates produce essentially identical results, but are less efficient energetically and hence will not be considered here). The temperature distribution in both layers in this limit is described by the original equation (15), but the boundary condition (21) has to be replaced with

$$\partial_Z \Theta_s(0) = F H_s^{-1} \left[\partial_Z \Theta_l(0) + Q\right] \tag{36}$$

to reflect the extra heat flux. Substitution of the modified boundary conditions into (17) yields the interfacial temperature

$$\Theta_i = \frac{1 + QF}{1 + BF + BH}. \tag{37}$$

This result is very instructive from the control viewpoint, because it directly relates the effect of perturbations on the dynamics to the material properties. For instance, one can easily see that, if the substrate is a very good conductor, $F \ll 1$, we have $\partial_Q \Theta_i = O(F)$. In other words, changes in the intensity of the radiation have a very small effect on the interfacial temperature, a situation one would like to avoid, if the spatial variation in the intensity is to be used to alter the dynamics. In particular, in the case of a perfectly conducting substrate, for which $F = 0$, the interfacial temperature becomes independent of Q, i.e., we completely loose our ability to control the dynamics. This unfavorable, from control perspective, effect provides additional motivation to study the dynamics and stability of liquid films using a more complicated model which explicitly includes conduction in the substrate.

This difficulty could have been predicted, as the present arrangement effectively attempts to change the temperature of one interface of the liquid layer heating the other one. Such an arrangement becomes less efficient as the relative heat resistance of the liquid layer (compared to the substrate) increases and a larger fraction of the extra heat flux is directed towards the bottom of the substrate.

C. Absorption at the top interface

To avoid this difficulty, it is preferable to direct the heat flux right at the location, whose temperature we are attempting to change, i.e., at the liquid-gas interface. This can be achieved by choosing the frequency of the radiation at which the liquid is optically thick, $d \ll h_l$, such that the dominant fraction of the radiation is absorbed at the liquid-gas interface. This case is conceptually similar to the case of an optically thick substrate considered above, so we proceed in the same way by replacing the boundary condition (18) with

$$-\partial_Z \Theta_l(H) = J - Q, \tag{38}$$

while (21) is left unchanged. Again substituting the modified boundary conditions into (17) we obtain

$$\Theta_i = \frac{1 + Q(F + H)}{1 + BF + BH}. \tag{39}$$

The dependence of the interfacial temperature Θ_i on Q in this case indeed turns out to be very similar to (37) obtained for absorption at the liquid-solid interface, so these two ways of applying thermal perturbations are essentially equivalent from the control standpoint when F is not small. For $F \ll 1$ the latter arrangement clearly has an advantage as the thermal response of the system $\partial_Q \Theta_i = O(H)$ does not vanish even for a perfectly conducting substrate.

We should contrast our approach with the one developed by Bau [15], who suggested *temperature* rather than *heat flux* control at the top boundary to suppress the short-wavelength instability leading to Marangoni-Bénard convection in a liquid layer on a substrate of infinite conductivity. No realistic mechanism to control the temperature directly has been suggested, however, so Bau's approach has a rather limited practical significance.

Summing up, we find that in order to effectively control the dynamics via radiative heating, it is preferable to use radiation with the frequency for which either the liquid or the solid layer is optically thick (highly absorbing), and the solid substrate of finite (and preferably not very high) conductivity.

IV. EVOLUTION IN THE PRESENCE OF DISTRIBUTED FEEDBACK

In the following we will assume that the radiation is absorbed at the liquid-solid interface, mainly because this will likely be the case most frequently encountered in practice. This is also the case that is easiest to implement in experiments by, e.g, painting the substrate. The other two cases considered above can be treated in essentially the same way. Substituting (37) into (24) we obtain the following nonlinear evolution equation for the height H, where the field Q plays a role of the distributed control parameter:

$$\partial_T H = -E\frac{1+QF}{K+H} + \frac{M}{2B}\partial_X\left(H^2\partial_X\frac{1+QF}{K+H}\right) + \frac{2E^2}{3PD}\partial_X\left(\frac{H^3(1+QF)}{K+H}\partial_X\frac{1+QF}{K+H}\right) + \frac{G}{3}\partial_X(H^3\partial_X H) - \frac{1}{3C}\partial_X(H^3\partial_X^3 H). \quad (40)$$

Linearizing this equation with respect to small deviations in the height $H(X,T) = H_0(T)+H_1(X,T)$ and radiation intensity $Q(X,T) = Q_0 + Q_1(X,T)$ in a manner similar to section II we obtain

$$\begin{aligned}\partial_T H_1 &= \frac{E(1+Q_0F)}{(K+H_0)^2}H_1 - \frac{M}{2B}\frac{(1+Q_0F)H_0^2}{(K+H_0)^2}\partial_X^2 H_1 \\ &- \frac{2E^2B}{3PD}\frac{(1+Q_0F)^2H_0^3}{(K+H_0)^3}\partial_X^2 H_1 \\ &+ \frac{GH_0^3}{3}\partial_X^2 H_1 - \frac{H_0^3}{3C}\partial_X^4 H_1 \\ &- \frac{EF}{K+H_0}Q_1 + \frac{MF}{2B}\frac{H_0^2}{K+H_0}\partial_X^2 Q_1 \\ &+ \frac{2E^2BF}{3PD}\frac{(1+Q_0F)H_0^3}{(K+H_0)^2}\partial_X^2 Q_1, \end{aligned} \quad (41)$$

where the mean height decreases according to

$$H_0(T) = -K + \sqrt{(K+1)^2 - 2E(1+Q_0F)T}. \quad (42)$$

Since the evaporation intensity increases by a factor of $1+Q_0F$, the amount of time it takes for the layer to completely evaporate decreases by the same factor:

$$T_0 = \frac{2K+1}{2E(1+Q_0F)}. \quad (43)$$

We should comment on the physical significance of the last three terms in (41) (the first five have been discussed in section II). The first term represents the variation in the evaporation rate produced by the variation of the interfacial temperature. The second term represents the tangential stress produced by the thermocapillary effect, while the third term represents the normal stress due to vapor recoil. The first effect is most effective in controlling the dynamics because it can directly remove the elevated regions by vaporizing the excess of the liquid, rather than by redistributing the liquid inside the film.

As a consequence of translational invariance of the problem the evolution equation (41) is diagonalized in the Fourier space. Using the Fourier transforms $\mathcal{H}(k,T) = \mathcal{F}[H_1(X,T)]$ and $\mathcal{Q}(k,T) = \mathcal{F}[Q_1(X,T)]$ of the variations in the height of the liquid layer and radiation intensity we can transform (41) into a system of uncoupled linear ODEs with time-dependent coefficients

$$\partial_T\mathcal{H}(k,T) = a(k,T)\mathcal{H}(k,T) + b(k,T)\mathcal{Q}(k,T). \quad (44)$$

In other words, the problem of determining the stability of the uniform solution of a PDE (40) is reduced to the problem of determining the stability of a family of *uncoupled* ODEs, which dramatically simplifies the analysis. Both the spectrum of growth rates of the unperturbed system

$$a(k,T) = a_1(T) + a_2(T)k^2 + a_3(T)k^4 \quad (45)$$

and the linear response of the system to perturbations which describes the effect of irradiation on the growth rates of the disturbance at different wavenumbers

$$b(k,T) = b_1(T) + b_2(T)k^2 \quad (46)$$

are polynomials in even powers of k with coefficients

$$\begin{aligned} a_1(T) &= \frac{E(1+Q_0F)}{(K+H_0(T))^2}, \\ a_2(T) &= -\frac{GH_0^3(T)}{3} + \frac{M}{2B}\frac{(1+Q_0F)H_0^2(T)}{(K+H_0(T))^2} \\ &+ \frac{2E^2B}{3PD}\frac{(1+Q_0F)^2H_0^3(T)}{(K+H_0(T))^3}, \\ a_3(T) &= -\frac{H_0^3(T)}{3C}, \\ b_1(T) &= -\frac{EF}{K+H_0(T)}, \\ b_2(T) &= -\frac{MF}{2B}\frac{H_0^2(T)}{K+H_0(T)} \\ &- \frac{2E^2BF}{3PD}\frac{(1+Q_0F)H_0^3(T)}{(K+H_0(T))^2}. \end{aligned} \quad (47)$$

Examination of these coefficients confirms our previous argument that laterally uniform heating tends to further destabilize the system. Indeed, the $0th$ order coefficient a_1 is always positive and increases with increasing heating intensity Q_0, making the system unstable toward long wavelength disturbances. The quadratic coefficient a_2 also increases with increasing Q_0 (at least for pure liquids, for which $M > 0$) extending the range of unstable wavenumbers. In fact, for thin enough films (i.e., when $|G| \ll 1$) the effect of gravity can be ignored, so that a_2 can be assumed strictly positive for $M > 0$.

The description presented above includes the classical Marangoni-Bénard convection as a special case in the limit of vanishing evaporation ($B \to 0$, $K \to \infty$). Suppression of the long wavelength instability leading to Marangoni-Bénard convection has been considered by Or, Kelly, Cortelezzi, and Speyer [14], who ignored conduction in the substrate and instead assumed a predetermined temperature at the liquid-solid interface in their analysis. Evaporation introduces some nontrivial physical effects which are absent in the case of thermal convection. We consider them next in the limits of slow and fast evaporation.

A. Slow evaporation

Let us consider what happens when the evaporation rate is small but not negligible. By this we mean that the disturbances grow on a time scale much smaller than the disappearance time T_0, rather than that the disappearance time itself is large. Since the first term in (41) changes on the same time scale as the mean H_0, this implies that either $M \gg EB$ (thermocapillarity dominates) or $PD(K+1) \ll EB(1+Q_0F)$ (vapor recoil dominates). In both cases we can ignore the change in the height $H_0(T)$ with time, so that (44) reduces to a system of ODEs

$$\partial_T \mathcal{H}(k,T) = a(k)\mathcal{H}(k,T) + b(k)\mathcal{Q}(k,T), \tag{48}$$

with time-independent coefficients. The most general linear relationship between the deviation $H_1(X,T)$ from the flat profile and the variation in heating intensity $Q_1(X,T)$ which leaves the system (44), and therefore (48), uncoupled is

$$\mathcal{Q}(k,T) = r(k,T)\mathcal{H}(k,T), \tag{49}$$

where the (as yet undetermined) coefficient of proportionality r is called the feedback gain. In the limit of slow evaporation both the growth rates and the response function are time-invariant, so without loss of generality the gain can be made time-invariant as well, and each of the equations (48) can be immediately integrated to yield at time T

$$\mathcal{H}(k,T) = \mathcal{H}(k,0)\exp[(a(k) + b(k)r(k))T]. \tag{50}$$

As we will see later, the control problem is crucially affected by the structure of the response function $b(k)$. Substituting $H_0(T) = 1$ into (47) one concludes that b_1 is always negative, while the sign of b_2 depends on the sign and relative magnitude of M. For pure liquids M is positive, so b_2 is negative and $b(k)$ is negative-definite. However, for mixtures and solutions it is not unusual to have the surface tension *increase* with temperature, $\partial_\theta \sigma > 0$, so that $M < 0$. If the thermocapillary effect opposes, and is stronger than, the effect of vapor recoil, we can have $b_2 > 0$. If b_2 is positive, $b(k)$ is no longer negative-definite and will pass through zero at some wavenumber

$$k_b = \sqrt{|b_1 b_2^{-1}|}. \tag{51}$$

At k_b the effects of differential evaporation, thermocapillarity, and vapor recoil produced by the imposed variation in the temperature cancel each other. This phenomenon is unique to the evaporatively driven convection and has no analog in the case of thermally driven Marangoni-Bénard convection.

The spectrum of growth rates will also pass through zero at the marginally stable wavenumber

$$k_a = \sqrt{-\frac{a_2}{2a_3} - \frac{1}{2a_3}\sqrt{a_2^2 - 4a_1a_3}}, \tag{52}$$

which defines the upper limit of the band of unstable Fourier modes, $|k| < k_a$. It is easy to check that k_b is always greater than k_a, so that the Fourier mode with wavenumber k_b is always stable. It is interesting to note, however, that $k_b \approx k_a$ in the limit when the gravity and surface tension are negligible, such as when the liquid film is intensively heated by radiation.

B. Fast evaporation

When the disturbances grow on the time scale comparable to the disappearance time T_0, the change in the mean height $H_0(T)$ cannot be ignored. However, (44) can be integrated, if we again assume linear dependence (49) between H_1 and Q_1. In particular, for time-invariant feedback gain we obtain an expression similar to (50) for the amplitude of the Fourier mode with wavenumber k at the time required for a uniform film to completely evaporate:

$$\mathcal{H}(k,T_0) = \mathcal{H}(k,0)\exp[a(k) + b(k)r(k)]. \tag{53}$$

We have to make a note regarding the validity of this solution. Since the thickness of the liquid layer is generally nonuniform, in certain regions the film will disappear sooner than the time T_0 is reached. This means that the equation (41) and its solution (53) are uniformly valid only until the time $T_0' < T_0$ at which the interface first touches the substrate. Because the thickness of the film cannot become negative, (41) has to be augmented with

$$\partial_T H_1(X,T) = 0 \tag{54}$$

for X such that $H_1(X,T) \le -H_0(T)$. However, since the limit $T \to T_0$ is nonsingular, (53) provides a good approximation to the height profile at the time T_0' as long as $T_0 - T_0' \ll T_0$, i.e., in the limit of small deviation from flat profile.

Although the solution (53) is similar in form to (50), $a(k)$ and $b(k)$ are now the integrated, rather than instantaneous, growth rates and response function. They also have the functional form given by (45) and (46), but with different coefficients:

$$\begin{aligned}
a_1 &= \ln(1 + K^{-1}),\\
a_2 &= -\frac{G(5K+4)}{60E(1+Q_0F)} + \frac{M}{4EB}(1 - 2K + 2K^2 a_1)\\
&\quad + \frac{EB(1+Q_0F)}{3PD(K+1)}(1 - 3K - 6K^2 + 6K^2(K+1)a_1),\\
a_3 &= -\frac{5K+4}{60EC(1+Q_0F)},\\
b_1 &= -\frac{F}{1+Q_0F},\\
b_2 &= -\frac{MF}{6EB(1+Q_0F)}\\
&\quad - \frac{EBF}{9PD}(2 - 3K + 6K^2 - 6K^3 a_1).
\end{aligned} \tag{55}$$

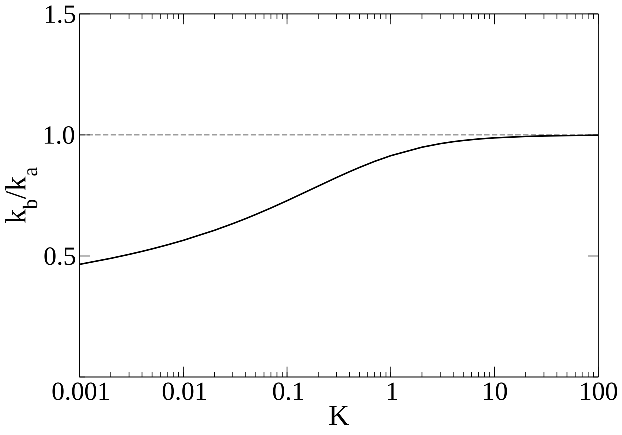

FIG. 2. The ratio of the wavenumbers at which the integrated response function $b(k)$ and spectrum of growth rates $a(k)$ pass through zero as a function of parameter K in the case of dominant thermocapillarity.

In this case a_1 is again strictly positive, indicating absolute instability at small k. The logarithmic dependence of a_1 on K means that differential evaporation is a very weakly destabilizing effect. Even though its physical significance might be rather small compared to thermocapillarity and vapor recoil in typical conditions, we include it because of its dominance at small wavenumbers. Furthermore, a_3 and b_1 are always negative, while the sign of a_2 and b_2 depends on the sign of the Marangoni number, M (all K-dependent expressions in parentheses are strictly positive). The case of fast evaporation is less trivial (and hence more interesting) from the control perspective than the case of slow evaporation, however, because now k_b can lie inside the unstable band $|k| < k_a$ (k_a and k_b are still given by (52) and (51), respectively). For instance, if vapor recoil, gravity, and surface tension are negligible compared to the thermocapillary effect, which is the case for sufficiently thin films, the ratio

$$\frac{k_b^2}{k_a^2} = \frac{3}{2}\frac{1 - 2K + 2K^2 \ln(1 + K^{-1})}{\ln(1 + K^{-1})} \tag{56}$$

is smaller than unity for any K (see Fig. 2), i.e., $k_b < k_a$.

If time-dependent feedback is chosen, the evolution equations cannot be explicitly integrated. This does not pose any fundamental problems for the analysis, although the control problem does become somewhat more complicated. In particular, because the instantaneous coefficients in (47) depend on time through the mean thickness of the liquid layer $H_0(T)$, the band of unstable modes $|k| < k_a(T)$ will change in time. Although $b_1(T)$ is negative at all times, the sign of $b_2(T)$ in general depends on time. As a consequence, the sign of the linear response $b(k, T)$ can change for certain wavenumbers as the liquid layer becomes thinner due to evaporation. However, contrary to the case of time-invariant feedback gain, if $b(k, T)$ does change sign at some wavenumber $k_b(T)$, the corresponding Fourier mode will always be instantaneously stable, $k_b(T) > k_a(T)$, regardless of the values of physical parameters.

V. THICK SUBSTRATE

In a typical coating application the thickness of the substrate will be much larger than the thickness of the liquid layer, but what is more important, H_s could be comparable to, or even larger than, the wavelength of a characteristic unstable disturbance, $\lambda \approx 2\pi/k_a$, where k_a is determined from (52) and (55). If the thickness of the substrate is not small compared to λ, the approximation (17) does not adequately describe the temperature profile in the substrate due to non-negligible heat flux in the horizontal direction. It has to be replaced with the exact solution (16) which, when written in the form

$$\Theta_s = \int [\delta(k) + s_s(k) \sinh k\,(Z + H_s)]\, e^{ikX} dk, \tag{57}$$

automatically satisfies the boundary condition (22). The liquid layer is still considered thin, so it is appropriate to use (17) which can be written in a form similar to (57):

$$\Theta_l = \int [c_l(k) + s_l(k)Z]\, e^{ikX} dk. \tag{58}$$

The boundary conditions (20) and (21) can also be replaced with their Fourier space counterparts in a straightforward way:

$$c_l(k) = \delta(k) + s_s(k) \sinh kH_s, \tag{59}$$

$$s_s(k)\, k \cosh kH_s = FH_s^{-1}[s_l(k) + \mathcal{Q}(k) + Q_0\delta(k)]. \tag{60}$$

In addition to the above boundary conditions, the solution has to satisfy conditions (18) and (19) which can be combined into a single equation

$$-\partial_Z \Theta_l(H) = B\Theta_l(H). \tag{61}$$

This boundary condition does not have a simple representation in the Fourier space, because the height H is itself a function of X. However, if we assume small deviations from a uniform height, at the leading order (61) can be written as

$$-s_l(k) = B[c_l(k) + s_l(k)H_0] + O(\mathcal{H}(k)). \tag{62}$$

Solving (59), (60), and (62) one obtains the distribution of the temperature at the liquid-gas interface

$$\Theta_i = \frac{1 + Q_0F}{1 + BH_0 + BF} + \int \frac{F\mathcal{Q}(k)}{kH_s \coth kH_s(1 + BH_0) + BF} e^{ikX} dk. \tag{63}$$

This expression generalizes (37) by including the effects of spatially varying intensity of radiation, but ignoring, at the leading order, the effects of spatially varying thickness of the liquid layer. Looking back at the structure of the evolution equation (41) obtained in the limit of thin substrate, we see that it has two groups of terms. The first five terms that make up $a(k,T)$ in (44) only account for the variation in H and ignore the variation in Q, while the last three, making up $b(k,T)$, ignore the variation in H and only account for the variation in Q. This means that the evolution equation for a liquid layer on a thick substrate is identical in its structure to (44), but the response function is now defined by

$$b(k,T) = \frac{(B^{-1} + F + H_0(T))(b_1(T) + b_2(T)k^2)}{kH_s \coth kH_s(B^{-1} + H_0(T)) + F} \tag{64}$$

rather than (46), with the coefficients $b_1(T)$ and $b_2(T)$ still determined by (47). Clearly (64) reduces to (46) when the thickness of the substrate is much less than the wavelength of the disturbance, $kH_s \ll 1$. Summing up, we see that the magnitude of the response to spatial variation in the heating intensity decreases with increasing thickness of the substrate. However, if $b(k,T)$ does change sign, it will do so exactly at the wavenumber (51) calculated in the assumption of a thin substrate.

The result (64) is easy to interpret: high wavenumbers correspond to very dense patterns of “hot” and “cold” spots imposed on the system. When the thickness of the solid layer is comparable to the size of such spots, the heat can “short-circuit” through the substrate, washing out the resulting variation in the temperature, compared to the thin limit case where essentially all heat transfer is between the top and the bottom of the solid layer. Smaller variation in the temperature, in turn, leads to a weaker response of the system.

Since the structure of $b(k,T)$ is essentially independent of the thickness h_s, in the remainder of the paper we will use the simpler expression (46) which is valid in the limit of thin substrate to simplify the algebra. Generalization to the case of arbitrary thickness is straightforward.

VI. ACTIVE CONTROL OF INTERFACIAL INSTABILITIES

As we saw above, it was rather easy to predict the effect of laterally uniform heating, described by the component Q_0, on the dynamics. However, the effect of introducing spatial variation $Q_1(X,T)$ is much less obvious. In fact, the structure of the linear response $b(k)$ turns out to be quite non-trivial, producing a number of unexpected results. Most significantly, nonuniform heating can produce the thermocapillary effect opposing the effect of vapor recoil and differential evaporation. The balance of these three effects will determine whether, and how, the interfacial instability can be controlled.

The idea of active control is to choose and apply a feedback (here $Q_1(X,T)$) that would stabilize the flat profile by converting the exponentially growing modes (50) or (53) into exponentially decaying ones. This requires finding some feedback gain $r(k)$ satisfying

$$a(k) + b(k)r(k) < 0, \quad \forall k. \tag{65}$$

Let us study how the solutions of this inequality depend on the structure of $a(k)$ and $b(k)$ in more detail. The analysis of the control problem with time-varying feedback gain is conceptually similar, so we will concentrate mostly on the time-invariant feedback in the next few sections.

A. Pure liquids

We have determined previously that for pure liquids both b_1 and b_2 are negative, so $b(k)$ is negative-definite. In this case we can easily see that a naive approach of heating the cooler (elevated) regions of the films and cooling the warmer (depressed) regions of the film, as suggested by Or, Kelly, Cortelezzi, and Speyer [14], successfully suppresses the interfacial instability even in the presence of evaporation. Indeed, let us apply a feedback

$$Q_1(X,T) = rH_1(X,T), \tag{66}$$

locally (in real space) proportional to the deviation of the interface from its mean position* (we will refer to it as “locally proportional” from now on). This feedback law corresponds to a wavenumber-independent gain $r(k) = r$ in Fourier space. If the constant r is chosen large enough to satisfy (65), we ensure that each Fourier mode is stable, so the deviation from the flat profile decreases exponentially in time.

Since $b(k)$ is negative-definite, (65) is equivalent to

$$r > f(k) \equiv -\frac{a(k)}{b(k)}, \quad \forall k. \tag{67}$$

The maximal value of $f(k)$ immediately gives the minimal value of the feedback gain that achieves stabilization:

$$r > r_{min}, \tag{68}$$

where $r_{min} = f(k_d)$ and k_d^2 is the greater of 0 and

$$-\frac{b_1}{b_2} + \frac{1}{a_3 b_2}\sqrt{a_3^2 b_1^2 - a_2 a_3 b_1 b_2 + a_1 a_3 b_2^2}. \tag{69}$$

It is important to note that r is not bounded from above. This choice of the feedback law is not unique (and certainly not optimal from many perspectives), but being extremely simple has its advantages for experimental implementation. We will consider other possibilities below in the context of mixtures.

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*In fact, Or *et al.* have suggested the use of weakly nonlinear control, with different gains corresponding to the first, second, and third powers of the interfacial deflection. Our linear analysis ignores the higher order gains.

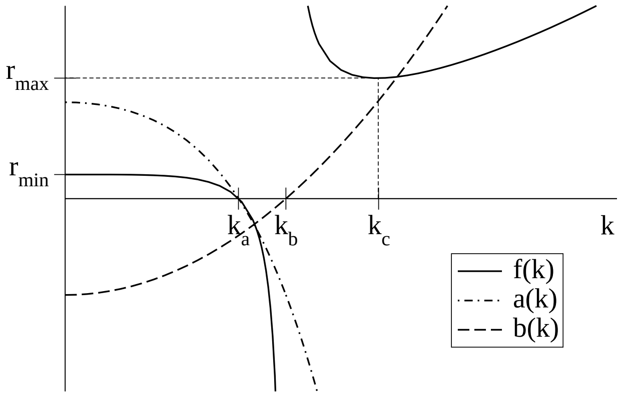

FIG. 3. Spectrum of growth rates $a(k)$, linear response $b(k)$, and their ratio $f(k)$ for $k_b > k_a$.

B. Mixtures

Mixtures and solutions can have a negative Marangoni number, so b_2 could be positive as well as negative, while b_1 is always negative. If b_2 is negative, the situation is identical to the case of pure liquids. If b_2 is positive, the situation is much more interesting. In the latter case (65) reduces to the following system of inequalities:

$$\begin{aligned} r(k) &> f(k), \quad |k| < k_b, \\ r(k) &< f(k), \quad |k| > k_b. \end{aligned} \tag{70}$$

As long as $k_b > k_a$ (this case is illustrated in Fig. 3) one can show that the system can still be stabilized by choosing locally proportional feedback (66): inequality (65) has a range of wavenumber-independent solutions

$$r_{min} < r < r_{max}, \tag{71}$$

where $r_{min} = f(k_d)$, $r_{max} = f(k_c)$, and

$$k_c^2 = -\frac{b_1}{b_2} - \frac{1}{a_3 b_2}\sqrt{a_3^2 b_1^2 - a_2 a_3 b_1 b_2 + a_1 a_3 b_2^2}. \tag{72}$$

This scenario describes, for example, the case of slowly evaporating thin films. As the heating intensity increases, $k_b \to k_a$. As a consequence $k_d \to k_c$, so the range (71) shrinks to a point. In this situation locally proportional feedback has a serious downside: control becomes extremely sensitive to intrinsic noise, modeling and measurement errors, i.e., it is not *robust* in the language of control theory. The reason for this is that wavenumber-independent feedback which is stabilizing for unstable modes becomes destabilizing for the stable ones due to the change in the sign of the response function, resulting in the amplification of small disturbances with wavenumbers close to the marginal wavenumber k_a.

The case $k_b < k_a$, corresponding to time-invariant feedback control of quickly evaporating thin films, is much more complicated and deserves a closer look. The wavenumber at which $b(k)$ vanishes is rather special: at $k = \pm k_b$ (53) reduces to

$$\mathcal{H}(k, T_0) = \mathcal{H}(k, 0)\exp[a(k)], \tag{73}$$

i.e., the feedback has no effect on the dynamics. Close to $\pm k_b$ this effect is very small, so perturbations of very large magnitude, $O(1/|k \pm k_b|)$ to be exact, have to be applied to produce $O(1)$ changes in the dynamics. In the language of control theory the system becomes *weakly controllable* in the vicinity of, and *uncontrollable* at, $\pm k_b$. This poses little difficulty when k_b lies outside the band of unstable wavenumbers, $k_b > k_a$, but causes significant problems when k_b happens to fall inside this band. Indeed, in order to suppress the instability we only need the ability to influence the *unstable*[†] modes.

As illustrated in Fig. 4, if the uncontrollable mode lies inside the unstable band, (65) cannot be satisfied by a feedback gain independent of the wavenumber, so locally proportional feedback fails. In principle, all unstable modes (except $k = \pm k_b$, of course) can still be converted into stable ones at the expense of allowing the feedback gain to depend on the wavenumber k. The price one has to pay for this is that the feedback law becomes delocalized in the real space:

$$Q_1(X, T) = \int R(X - X')H_1(X', T)dX', \tag{74}$$

where according to the convolution theorem, the kernel $R(X)$ is given by the inverse Fourier transform of $r(k)$:

$$R(X) = \frac{1}{2\pi}\int r(k)e^{ikX}dk. \tag{75}$$

Since $f(k)$ diverges at $\pm k_b$, the time-invariant feedback gain $r(k)$ would have to diverge as well. The perturbation $\mathcal{Q}(k, T)$, however, is bounded (for instance, the total radiation intensity cannot be negative, so $|Q_1(X, T)| < Q_0$). Therefore, in a laterally unbounded system disturbances with wavenumbers close to k_b will grow uncontrollably and eventually the film will rupture, producing dry spots with spatial periodicity $2\pi/k_b$. A laterally bounded system, of course, might not support any modes with wavenumbers close to k_b, if its size is sufficiently small.

In fact, the existence of the uncontrollable mode is not a property of the system, but rather a consequence of our choice of time-invariant feedback. The rupturing instability described above can be easily prevented, if time-varying, rather than time-invariant, feedback gain is used. In reality, because the height of the liquid layer will be changing in time due to the evaporation, the coefficients (47) will be changing too, so generically $k_b(T)$

[†] Strictly speaking, influence over a certain range of stable modes might also be necessary to deal with the *control spillover* effect [22], if a *nonlinear* control is desired. It might also be needed in order to design linear control with certain desired properties as we will see in the next section.

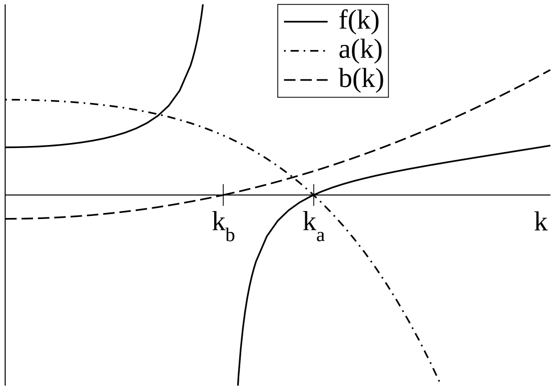

FIG. 4. Spectrum of growth rates $a(k)$, linear response $b(k)$, and their ratio $f(k)$ for $k_b < k_a$.

will be time-dependent. In order to make the flat profile stable using time-varying feedback, one simply has to choose $r(k,T)$ satisfying

$$\int_T^{T_0} b(k,\tau)r(k,\tau)d\tau < -\int_T^{T_0} a(k,\tau)d\tau \qquad (76)$$

for every $0 \le T < T_0$ instead of (65). The existence of bounded solutions to (76) is guaranteed, since $k_b(T) > k_a(T)$, as we have determined in section IV. It is easiest to satisfy this inequality with a feedback gain that simply changes sign wherever $b(k,T)$ changes sign to ensure that the action of the feedback is stabilizing for every k. In fact, (76) should have bounded solutions even if $k_b(T) < k_a(T)$, as long as $k_b(T)$ is nonconstant.

VII. SPATIAL LOCALIZATION

As the analysis of the previous section suggests, locally proportional feedback (which has a singularly localized kernel, $R(X) = r\delta(X)$) is not always robust and can fail in rather harmless situations. In this section we consider an alternative approach of using nonlocal feedback (74) for control of interfacial instabilities. We have already seen that (65) does not uniquely determine the feedback law, it is only a necessary condition. The selection has to be made using some additional criteria. It is useful to define the feedback which is optimal in some sense. For instance, one could require that the least amount of energy be expended on stabilizing the system, or that the terminal deviation from the uniform state be minimal. Alternatively, one could look for a feedback law that is most localized (this requirement, however, still does not specify the feedback uniquely).

The nonlocal nature of the feedback means that the perturbation applied at some spatial location X depends on the state of the system at some other location X'. It is important to understand the degree of spatial (de)localization, i.e., how fast the kernel $R(X-X')$ decays with the distance $|X-X'|$, and how this localization is related to the structure of the response function $b(k)$ and the choice of the criterion used to select some particular feedback law out of all possibilities satisfying (65) (or (76)), because it determines the complexity of our controller. For instance, it determines how much information has to be processed in order to calculate the perturbation at a single spatial location.

A. Time-invariant feedback

To get an idea of what kinds of nonlocal kernels could arise, let us begin by restricting our attention to the time-invariant problem which arises in the limit of slow evaporation. For simplicity, let us ignore the effect of surface tension, which is always stabilizing, and find the feedback gain that produces the same convergence rate a_0 for every k, such that

$$a_1 + a_2k^2 + (b_1 + b_2k^2)r(k) = -a_0. \qquad (77)$$

Clearly, this requires

$$r(k) = -\frac{a_2}{b_2} - \frac{a_0 + a_1 - a_2b_1b_2^{-1}}{b_1 + b_2k^2}. \qquad (78)$$

If $b(k)$ is negative-definite ($b_2 < 0$), the Fourier transform (75) yields an exponentially decaying kernel

$$R(X) = -\frac{a_2}{b_2}\delta(X) + \frac{a_0 + a_1 - a_2b_1b_2^{-1}}{2\sqrt{b_1b_2}}\exp(-k_b|X|). \qquad (79)$$

Because of such fast (exponential) decay, in practice the kernel can be truncated at a distance of order few characteristic lengths, k_b^{-1}, with no loss in performance.

Alternatively, if $b(k)$ changes sign at $\pm k_b$ ($b_2 > 0$), the gain (78) becomes singular at these points. One can nevertheless formally evaluate the Fourier transform using contour integration in the complex k-plane to obtain

$$R(X) = -\frac{a_2}{b_2}\delta(X) + \frac{a_0 + a_1 - a_2b_1b_2^{-1}}{4\sqrt{-b_1b_2}}\sin(k_b|X|). \qquad (80)$$

Clearly, in this case the kernel is completely delocalized; it does not decrease with the distance $|X|$. This complete delocalization is a direct consequence of the singularity, indicating the loss of controllability. Indeed, if a disturbance with wavenumber k_b is present, the integral (74) will diverge, indicating that feedback of infinite magnitude is required to achieve the desired rate of decay.

More generally, one would expect that, if k_b lies in the unstable band, $k_b < k_a$, the kernel will necessarily be completely delocalized regardless of the particular choice of the feedback law. On the other hand, if the Fourier mode k_b is stable, the feedback gain does not have to diverge. In fact, one could always choose a wavenumber-independent feedback gain, yielding a kernel localized at a single point.

In fact, by shifting the integration contour in the complex k-plane it can be shown [23] that, if $r(k)$ is analytic in a strip $|\Im(k)| < \beta$ and bounded on the real axis, $\Im(k) = 0$, the kernel can be represented in the form

$$R(X) = R_0\delta(X) + R_1(X), \tag{81}$$

where $R_0 = \lim_{|k|\to\infty} r(k)$ and $R_1(X)$ decays exponentially at a rate faster than any $\alpha < \beta$:

$$R_1(X)e^{\alpha|X|} \to 0, \quad |X| \to \infty. \tag{82}$$

For instance, if $b_2 < 0$, the poles of $r(k)$ lie on the imaginary axis and $\beta = |\Im(\pm ik_b)| = k_b$, so (79) is seen to be just a special case of the more general relationship (81). Similarly, for $b_2 > 0$, the poles lie on the real axis, so $\beta = |\Im(k_b)| = 0$, and we obtain a delocalized kernel (80), which does not decay as $|X| \to \infty$. (The fact that $\beta = 0$ does not mean, however, that the kernel should be *completely* delocalized: we will see later that, if $r(k)$ has no *poles* on the real axis, $R(X)$ will decay as a power of X.)

Additional restrictions on the domain of analyticity are imposed, if $r(k)$ is required to satisfy certain optimality conditions. For instance, practical considerations might require that the time-averaged and/or terminal deviation of the liquid film from the flat profile is minimized. An optimal feedback achieving this goal and subject to the physical constraint of finite power available for control can be obtained by minimizing the following cost functional[‡]:

$$V[r] = \int_0^{T_0}\int_{-\infty}^{\infty}[\psi H_1^2(X,T) + Q_1^2(X,T)]dXdT + \int_{-\infty}^{\infty}\phi H_1^2(X,T_0)dX, \tag{83}$$

where $\psi \geq 0$ and $\phi \geq 0$ are costs associated with time-averaged and terminal deviation from the flat profile, respectively, per unit cost of the applied control signal.

Conventional control theoretic approach calls for making the problem finite-dimensional at this stage by employing the formalism of Galerkin projection onto a finite number of Fourier modes. This involves fixing the values of all parameters and solving the resulting optimization problem numerically. This approach has recently been used by Or, Cortelezzi, and Speyer [24] in the context of Rayleigh-Bénard convection. The most serious drawback of this approach is the lack of generality common to all numerical methods. By forcing special choices of parameters it obscures the connections between the geometrical structure of the problem, the optimality criteria, and the structure of the feedback.

Considerable progress can be made without resorting to numerics, if we allow our problem to remain infinite-dimensional. We can readily rewrite the functional (83) in the Fourier space as

$$V[r] = \int_0^{T_0}\int_{-\infty}^{\infty}[\psi|\mathcal{H}(k,T)|^2 + |\mathcal{Q}(k,T)|^2]dkdT + \int_{-\infty}^{\infty}\phi|\mathcal{H}(k,T_0)|^2dk. \tag{84}$$

In the case of slow evaporation it is appropriate to take $T_0 = \infty$ and $\phi = 0$. Minimization of the functional (84) subject to the constraints (48) and (49) with time-invariant feedback results in an infinite system of scalar Riccati equations

$$-b^2(k)p^2(k) + 2a(k)p(k) + \psi = 0, \tag{85}$$

whose positive-definite solutions give the feedback gain $r(k) = -b(k)p(k)$ for each wavenumber k:

$$r(k) = -\frac{a(k) + \sqrt{a^2(k) + \psi b^2(k)}}{b(k)}. \tag{86}$$

When the analytic continuation of $r(k)$ is considered the branch of the square root which is positive on the real axis has to be taken. Despite $b(k)$ appearing in the denominator, $r(k)$ does not necessarily have poles at the zeroes of $b(k)$. Close to the zeroes of $b(k)$ (86) can be expanded as

$$r(k) = -\frac{a(k) + \sqrt{a^2(k)}}{b(k)} + \frac{\psi}{2}\frac{\sqrt{a^2(k)}}{a^2(k)}b(k) + \cdots, \tag{87}$$

so $r(k)$ has no poles, if the zeroes of $b(k)$ lie on the real axis ($b_2 > 0$) *and* the Fourier mode k_b is stable ($k_b > k_a$). On the other hand, $r(k)$ generally has four pairs of branch points determined by

$$a^2(k) + \psi b^2(k) = 0, \tag{88}$$

some of which can coalesce forming branch cuts, or discontinuities, that cross the real axis. In general β is determined by the imaginary part of either a pole or a branch point closest to the real axis.

When the cost of control is finite, $0 < \psi < \infty$, none of the branch points lie on the real axis, because generically $k_a \neq k_b$. If b_2 is negative, there are two poles lying on the imaginary axis, so $0 < \beta \leq k_b$ and $R(X)$ decays exponentially. If b_2 is positive, but $k_b > k_a$, the poles disappear, so $R(X)$ will again decay exponentially fast. As either $k_b \to k_a$ or $\psi \to 0$, several of the branch points approach the real axis, so that $\beta \to 0$. For $k_b < k_a$, two poles are created on the real axis resulting in a completely delocalized kernel.

[‡] This minimization problem corresponds to the Linear-Quadratic Regulator (LQR) problem [25] of control theory.

In the limit of "expensive" control, $\psi = 0$, a couple of branch points coalesce at k_a and another at $-k_a$, producing two discontinuities. As a result, on the real axis we have

$$r(k) = \begin{cases} -2a(k)/b(k), & |k| < k_a, \\ 0, & |k| > k_a. \end{cases} \tag{89}$$

so the feedback gain vanishes for all stable modes. If the zeroes of $b(k)$ lie on the real axis, and $k_b < k_a$, the gain (89) has poles at $\pm k_b$, so the feedback gain diverges and we again obtain a completely delocalized kernel. If, on the other hand, the zeroes of $b(k)$ are real but lie outside the band of unstable modes, $k_b > k_a$, the gain has no poles, only the discontinuities crossing the real axis at $\pm k_a$. In either case $\beta = 0$.

Generally, if the feedback gain is given by a sufficiently smooth even function for unstable modes, e.g., $r(k) \in C^1(-k_a, k_a)$, and vanishes for all stable modes, one can use integration by parts to show that

$$\begin{aligned} R(X) &= \frac{1}{\pi}\int_0^{k_a} r(k)\cos(kX)dk \\ &= \frac{r(k_a)}{\pi}\frac{\sin(k_a X)}{X} + \frac{r'(k_a)}{\pi}\frac{\cos(k_a X)}{X^2} + O(X^{-3}), \end{aligned} \tag{90}$$

so that the kernel will be localized, with a power law, rather than exponential, rate of decay

$$R(X)|X|^\alpha \to 0, \quad |X| \to \infty, \tag{91}$$

where $\alpha < 2$ (we cannot have $\alpha \geq 2$ because of the inequality (70), which requires that either $r(k_a) \neq 0$ or $r'(k_a) \neq 0$). In particular, for the optimal feedback gain (89) one obtains

$$R(X) = -\frac{2}{\pi}\frac{a'(k_a)}{b(k_a)}\frac{\cos(k_a X)}{X^2} + O(X^{-3}), \tag{92}$$

provided, of course, $k_b > k_a$.

In principle, as long as $k_b > k_a$, one can always obtain a kernel $R(X)$ exhibiting power-law decay with an arbitrarily large exponent α, even if the gain $r(k)$ has compact support, $r(k) = 0$ for $|k| > k_s > k_a$. This requires a tradeoff: the feedback should be applied not only to the unstable modes, but also to a range of stable modes, $k_a < |k| < k_s$, and it should be sufficiently smooth in the Fourier space, i.e., $r(k) \in C^{n-1}$ with n equal to the integer part of α.

As we mentioned above, the kernel $R(X - X')$ of the spatial convolution operator determines how much information from distant regions X' of the system we need to have (and use) in order to suppress the instability at any given location X. If the control is "cheap", the kernel is very localized, i.e., we only need local information. In other words, we can afford *not* using information from regions that are far away, but instead use "brute force" approach to control the dynamics with strong perturbations. In the opposite case of "expensive" control, small intelligently chosen perturbations are used to guide the system in the desired direction. This intelligent approach, however, requires a much more extensive knowledge of the state of the system. As a consequence, the respective kernel has to be more delocalized, extending to remote locations that are further away from the point where the feedback is applied.

B. Time-varying feedback

We have learned above that time-invariant feedback can experience serious difficulties when disturbances grow on the time scale comparable to the disappearance time T_0. These difficulties are easily resolved, if time-varying feedback gain is used instead. In fact, optimal (in the sense defined above) feedback will always be time-dependent as long as the spectrum of growth rates and the response function vary in time. The solution to the time-varying problem (84) is given by a generalization of the Riccati equations (85):

$$\begin{aligned} -\dot{p}(k,T) &= -b^2(k,T)p^2(k,T) + 2a(k,T)p(k,T) + \psi, \\ p(k,T_0) &= \phi. \end{aligned} \tag{93}$$

The algebraic equations are thus replaced by nonlinear ordinary differential equations. If we leave T_0 finite and set $\psi = 0$, which is a natural choice for the case of fast evaporation, i.e., require that the terminal deviation from the flat profile be minimal, these equations can be simplified considerably by using the ansatz $p(k,T) = 1/s(k,T)$:

$$\begin{aligned} \dot{s}(k,T) &= 2a(k,T)s(k,T) - b^2(k,T), \\ s(k,T_0) &= \phi^{-1}. \end{aligned} \tag{94}$$

The solution of (94) defines the feedback gain $r(k,T) = -b(k,T)/s(k,T)$ which is both time- and wavenumber-dependent:

$$r(k,T) = -\frac{b(k,T)u^2(k,T)}{\phi^{-1} + \int_T^{T_0} b^2(k,\tau)u^2(k,\tau)d\tau}. \tag{95}$$

The evolution operator

$$u(k,T) = \exp\left[\int_T^{T_0} a(k,\tau)d\tau\right] \tag{96}$$

can be explicitly evaluated and has the following structure:

$$u(k,T) = \exp[u_1(T) + u_2(T)k^2 + u_3(T)k^4], \tag{97}$$

where the time-dependent coefficients are given by

$$\begin{aligned} u_1(T) &= \ln(1 + K^{-1}H_0(T)), \\ u_2(T) &= -\frac{G(5K + 4H_0(T))H_0^4(T)}{60E(1 + Q_0 F)} \end{aligned}$$

$$
\begin{aligned}
&+\frac{M}{4BE}(H_0^2(T)-2KH_0(T)+2K^2u_1(T))\\
&+\frac{EB(1+Q_0F)}{3PD(K+H_0(T))}\left[H_0^3(T)-3KH_0^2(T)\right.\\
&\quad\left.-6K^2H_0(T)+6K^2(K+H_0(T))u_1(T)\right],\\
u_3(T)&=-\frac{(5K+4H_0(T))H_0^4(T)}{60EC(1+Q_0F)}. \qquad (98)
\end{aligned}
$$

As expected, $u_i(0)=a_i$ for $i=1,2,3$, where a_i are the integrated coefficients of $a(k,T)$ defined in (55).

The integral in (95) cannot be evaluated explicitly. In certain special cases (e.g., in the limit of slow evaporation) one can obtain a good approximation using the saddle-point method. Alternatively one can employ numerical integration. However, even without making this last step a number of important conclusions can be drawn. First of all, the optimal feedback (95) is wavenumber-dependent, so its kernel will necessarily be nonlocal. Furthermore, its kernel will always decay exponentially fast in real space, since $r(k,T)$ is analytic on the real k-axis. This is to be expected: we have determined that $k_b(T)>k_a(T)$ for all T. Finally, the optimal feedback gain will change sign wherever $b(k,T)$ changes sign, also in agreement with our expectations.

A stabilizing time-varying feedback can be easily obtained analytically, if optimality is not required. Indeed, in order to satisfy (76) it is sufficient to choose a feedback gain which makes the growth rate of the perturbed system negative at each instant in time

$$a(k,T)+b(k,T)r(k,T)<0. \qquad (99)$$

This brings us back to the time-invariant problem considered above: most of the results obtained in the preceding sections can be naturally generalized by merely replacing growth rates $a(k)$ and the response function $b(k)$ with their time-dependent counterparts. In particular, since either $b(k,T)$ has no zeroes or its zeroes correspond to instantaneously stable modes, one could always choose a wavenumber-independent feedback $r(T)$ producing the kernel $R(X,T)=r(T)\delta(X)$ which is localized at a point. If $b(k,T)$ is not negative-definite, locally proportional feedback has an adverse effect on the stability of the modes with wavenumbers $|k|>k_b(T)$. To get rid of this effect it makes sense to choose the gain in the form $r(k,T)=-b(k,T)p(k,T)$ with some $p(k,t)>0$, such that the gain changes sign wherever the linear response changes sign, so the net result is always stabilizing. Then any $p(k,t)$ that satisfies the following two conditions

$$p(k,T)>\frac{a(k,T)}{b^2(k,T)}, \quad |k|<k_a(T), \qquad (100)$$

$$p(k,T)k^2\rightarrow \text{const}<\infty, \quad |k|\rightarrow\infty, \qquad (101)$$

will achieve stabilization. Such feedback will be both wavenumber- and time-dependent, so its kernel will be non-local with exponential or power law decay, depending on the smoothness properties of $p(k,T)$.

VIII. SUMMARY

A few remarks concerning the problem of controlling the evaporatively driven instability are now in order. First of all, we have determined that this type of instability could be effectively suppressed by applying distributed feedback through thermal perturbations. Such perturbations can be easily produced by irradiating the liquid film and/or the substrate with a spatially modulated source of visible, infrared, or microwave radiation. The frequency of radiation should be tuned to produce the desired absorption characteristics. The sensing can also be implemented rather easily. Laser interferometry or shadowgraphy can be used to measure the local thickness of the liquid film, if it is transparent. Otherwise, thermal imaging could be employed to calculate the thickness from the surface temperature measurements (the feedback can also be easily reformulated in terms of the deviation of the temperature from its mean value).

The evaporation brings a new dimension into the control problem, compared to the relatively well studied case of thermally driven Marangoni-Bénard convection. One of the most important results of the above analysis is that in certain cases the simple noise-cancellation approach resulting in locally proportional time-invariant feedback does not work well or does not work at all. In those cases we are forced to use either a more complicated nonlocal time-invariant feedback, or switch to a time-varying feedback, local or nonlocal, depending on the physical parameters of the liquid. To be precise, the degree of spatial localization of the feedback law is defined by the control goals, subject to the limiting conditions imposed by the physical parameters.

Proper modeling of the system with feedback requires using the substrate of finite conductivity and finite thickness, rather than imposing a constant temperature or constant flux boundary conditions at the liquid-solid interface. Explicit modeling of the gas layer, on the other hand, appears to be unnecessary. Even with these complications, control of the long-wavelength evaporative instability represents a unique problem from mathematical perspective. It is sufficiently rich to produce a variety of qualitatively different regimes, but at the same time simple enough to allow explicit analytical solutions in most cases.

Finally, although quite a few assumptions and simplifications were made in deriving the evolution equations and the feedback laws, most of them were rather technical and could be relaxed at the expense of making the algebra a bit more lengthy. None of the important conclusions should change though. One possible exception is neglecting the effect of the composition of n-component liquids on the physical parameters (density, viscosity, and so on). The proper treatment of such liquids would include the evolution equations for the concentration of each of the components, yielding a system of n PDEs in the lubrication approximation. These PDEs will still diagonalize in the Fourier space due to the translational invariance of the system, but instead of a 1-dimensional control problem we had to solve for each wavenumber, we would end up with an n-dimensional problem. Solution of matrix Riccati equations is a much less trivial task, even in the time-invariant case. More important, the existence of a solution is predicated on the controllability of all degrees of freedom: for instance, it is far from obvious that thermal perturbations will be effective in controlling the concentration profiles of multiple components. This problem is the subject of further investigation.

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