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Interfacial stability and self-similar rupture of evaporating liquid layers under vapor recoil

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We investigate interfacial stability of an evaporating viscous liquid layer above/below a horizontal heated substrate in the framework of a long-wave model that accounts for surface tension, positive/negative gravity, and evaporation effects of mass loss and vapor recoil. With the time-dependent linear stability analysis, it is found that the interface instability is enhanced by vapor recoil with time using an effective growth rate. The destabilizing mechanism of vapor thrust competes with the stabilizing surface tension, and the effects of the latter are not asymptotically negligible near rupture, reflected by a rescaled effective interfacial pressure. A two-dimensional nonlinear evolution is investigated for the quasi-equilibrium evaporating layers with different evaporative conditions for Rayleigh–Taylor unstable and sessile layers. For weak mass loss and strong vapor recoil, the well-defined capillary ridges emerge around a deepening narrow valley with increasing wavelength under a positive gravity, while, on the basis of initial condition, main and secondary droplets are either coalesced partially or separated by a sharp dry-out point under a negative gravity. The rupture location depends strongly on the characteristics of a given initial condition, except for the random perturbation. For both the cases, an increase in the modified evaporation number tends to reduce the rupture time t_r and droplet thickness remarkably. Similarity analysis along with numerical strategy is presented for the final stage of touch-down dynamics, determined by a physical balance between the vapor recoil and capillary force. The evaporation-driven rupture with a significant vapor recoil and negligible mass loss is shown to contain a countably infinite number of similarity solutions whose horizontal and vertical length scales behave as $(t_r - t)^{1/2}$ and $(t_r - t)^{1/3}$. The first similarity solution represents a stable single-point rupture. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4972179>]

I. INTRODUCTION

Dynamics and stability of evaporating liquid layers on substrates have been an important subject for both the scientific interests and engineering applications of cooling, depositing, and drying. During the last decades, numerous experimental and theoretical works have been devoted to various physical mechanisms including phase change, capillarity, thermocapillarity, gravity, and van der Waals interactions to predict and control the pattern formation of a free surface.^{1,2} The finite time rupture of evaporating layers on substrates can lead to the formation of droplets with contact lines, dry-out points, or drained regions, and thus dramatically influences the self-organized patterning in the (de)wetting process or the local heat flux in the cooling process.

At an evaporating interface, a mass balance reads $j = \rho(\mathbf{v} - \mathbf{v}_I) \cdot \mathbf{n} = \rho_g(\mathbf{v}_g - \mathbf{v}_I) \cdot \mathbf{n}$, where j , $\mathbf{v}$, and ρ are the evaporative mass flux, liquid velocity, and density, $\mathbf{n}$ stands for a unit vector normal to the interface, and the subscripts “g” and “I” refer to the gas phase and interface. As $\rho_g/\rho = O(10^{-3})$, it implies that the normal velocity evaluated on the gas side of the interface is much larger than that on the liquid side. Thus the departing vapor would provide a large acceleration which exerts

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a reactive pressure to the interface. A thinner part of a layer has a greater momentum flux, which acts as a destabilizing effect. The force is referred to as vapor recoil, interpreted first for the convection instability under reduced pressure by Hickman.³ The destabilizing influence could be tangibly recognizable for a rapidly evaporating interface as shown by Palmer⁴ through a quasi-steady linear stability analysis (LSA). Prosperetti and Plesset⁵ revisited the problem without the Marangoni effect and viscosity, who stated that the destabilizations of vapor recoil, inertia, and moving boundary were all negligible.

A nonlinear model of the rupture in an evaporating film was first proposed by Burelbach *et al.*⁶ by means of the long-wave theory and one-sided assumption. The effects of evaporation, thermo-capillarity, and negative disjoining pressure arising from long-range molecular forces due to van der Waals attractions were incorporated into a nonlinear evolution equation and studied numerically. Joo *et al.*⁷ extended the theory to an evaporating layer falling down a heated inclined plate. With a generalized intermolecular potential, Oron⁸ analyzed the three-dimensional (3D) evaporating films. Later, Shklyaev and Fried⁹ extended the model with a set of interface conditions under a general thermodynamic framework. However, in subsequent studies, with the growing number of applications in evaporating layers, surprisingly there was little attention paid to the influence of vapor recoil on their rupture events. A further explanation of the vapor-recoil instability on the interface rupture and its interaction with the surface tension, gravity, and evaporative mass loss is among the motivations of the present study.

Since the evaporation tends to drive the interfacial temperature towards an equilibrium value under the system pressure,¹⁰ Marangoni convection is usually inconsequential for evaporating layers in contact with own pure vapor. Consequently, a thermodynamic quasi-equilibrium condition (interface kinetics parameter^{4,6} $K = 0$) is a reasonable simplification in many applications,^{11,12} in which the evaporation rate is maximum while the greatest vapor recoil is achieved,⁶ a limiting case the current paper attempts to look at since vapor thrust can be a driving force of the local depression and rupture of the interface.^{4,6} Next, the heat flux through the gas phase is negligible in comparison with the latent heat, as considered by Palmer⁴ for the relatively intense evaporation. However, the effects of the conductive/convective heat flow between ambient gas and the free surface should be incorporated in non-equilibrium cases with a relatively low latent heat fluid ($K > 0$ for a lower evaporation rate).^{13–15} Furthermore, the extension to $(2 + 1)$ -dimensions, required for studying the 3D pattern formation, is technically trivial and will be discussed with more physical effects elsewhere, in which “2” and “1” indicate the horizontal spatial and temporal dimensionalities. For example, Fig. 1 illustrates a rupture pattern of a 3D layer. Therefore, in this paper, we put many of the complications, including nonequilibrium and Marangoni effects, vapor dynamics of heat and mass transfer, and so on, aside for the moment, and focus on one of the fundamental problems in the

FIG. 1. An unstable evaporating layer covering a heated horizontal substrate under vapor recoil. (a) Schematic of a sessile layer with $\tilde{\lambda} \gg h_0$, for a suspended layer subject to RTI, the direction of g should be reversed. (b) Illustration of a typical 3D rupture pattern of the Rayleigh–Taylor unstable layer, obtained by solving a canonical form of Eq. (12) and the 2D version of the random initial condition (16c) with $\mathcal{E} = 0.05$ and $\mathcal{D} = 1$ in a 151×151 mesh. The side length of the square domain is 2π and $t_r = 5.108$.

interfacial instability, namely, the two-dimensional (2D) nonlinear evolution and finite-time rupture of quasi-equilibrium evaporating layers subject to a constant surface tension and gravity.

Many models dealing with evaporating layers are limited to ultrathin films of 10–100 nm^{6,8,9} that gravity effects are negligible and van der Waals attractions are effective, while the experiments and applications often involve evaporating layers which are thick enough^{10,14,16,17} that the gravity plays a role and long-range molecular forces can be neglected. A simple destabilizing mechanism for the interface is the Rayleigh–Taylor instability (RTI). For a suspended liquid layer under a horizontal plate, the negative gravity acts against the stabilizing surface tension. Such a system has been studied extensively without evaporation, e.g., various patterns consisting of separated droplets and drained regions have been shown in Refs. 18 and 19. With interface initially in equilibrium with its vapor below, Bestehorn and Merkt¹¹ incorporated the phase change and showed that a layer hanging on a cooled substrate could be stabilized with localized mass loss/gain even without stabilizing thermocapillarity; however, the effect of vapor recoil was neglected. Here, we aim to investigate the influences of both evaporation effects of vapor recoil and mass loss on the Rayleigh–Taylor unstable layers and illustrate the differences from the relevant results.

A relevant question concerning a thin-film equation involves the similarity solution of the nonlinear partial differential equation (PDE) governing the finite-time rupture, as discussed by Oron *et al.*,¹ Eggers,²⁰ and Chou and Kwong.²¹ Since the self-similar solution distinguishes the limiting state of the nonlinear dynamical system, an understanding of the asymptotic behavior is useful in both selecting a cutoff criterion for the interface-singularity simulation and providing insights into the subtle dynamics, e.g., invariant nature, which could not be resolved by either stability analysis or numerical simulation. The rupture singularity of evaporating films under van der Waals forces was examined by Burelbach *et al.*,⁶ who suggested that the surface tension was negligible near rupture and the self-similar dynamics was governed by the competition of mass loss, vapor recoil, viscous dissipation, and van der Waals attractions; nevertheless, the specific similarity solution was not their major concern. From a balance of van der Waals and viscous forces with negligible surface tension, a separable solution was given analytically by Oron *et al.*¹ Under a balance of van der Waals and capillary forces, a discrete set of similarity solutions for the rupture of viscous films has been provided by Zhang and Lister.²² Witelski and Bernoff²³ developed an alternative technique to obtain the similarity solutions and showed that the “lowest-order” solution was stable for point rupture with LSA. For freely thin viscous sheets, Vaynblat *et al.*²⁴ showed that a balance between inertial, viscous, and van der Waals forces allowed a family of similarity solutions with a dimensional analysis, while the surface tension was negligible. However, most studies did not include evaporation effects. We consider the evaporation-driven rupture with significant vapor recoil in place of van der Waals forces to show, by time-dependent simulations of the relevant PDE, that the effects of the surface tension are not asymptotically inconsequential and explore the possibility of local similarity solutions. The challenges of the numerical strategy for solving the self-similar ordinary differential equation (ODE) arise from the nonlinearity and high-order derivatives.

II. PROBLEM FORMULATION

We consider a viscous, single-component, Newtonian liquid layer bounded above/below by its semi-infinite vapor and below/above by a rigid and conducting horizontal substrate in the case of positive/negative gravity. The layer evaporates into its pure vapor on the heated substrate of temperature, θ_w , higher than its saturation temperature, θ_s . The disjoining pressure arising from long-range molecular forces due to van der Waals attractions^{1,6,25} is negligible for the layer thickness of interest. The system is formulized with a 2D Cartesian coordinate (x, z) system with an initially unperturbed thickness, h_0 , as shown in Fig. 1. The gas-liquid interface is described by $z = h(x, t)$ with time t . The unit vector normal to the interface and directed towards the gas phase is expressed as $\mathbf{n} = (-h_x, 1)(1 + h_x^2)^{-1/2}$, and $\mathbf{t} = (1, h_x)(1 + h_x^2)^{-1/2}$ represents the unit vector tangent to the interface. The subscripts pertaining to independent variables denote the derivative throughout this paper. The governing equations for the liquid layer take the form

$$\rho[\mathbf{v}_t + (\mathbf{v} \cdot \nabla) \mathbf{v}] = -\nabla p + \mu \nabla^2 \mathbf{v} + \rho \mathbf{g}, \quad (1)$$

$$\nabla \cdot \mathbf{v} = 0, \quad \rho c_p [\theta_t + (\mathbf{v} \cdot \nabla) \theta] = k_{th} \nabla^2 \theta, \quad (2)$$

where $\nabla = (\partial_x, \partial_z)$ and ∇^2 are the gradient and Laplacian, $\mathbf{v} = (u, w)$ and θ denote the velocity and temperature, p is the pressure of liquid relative to the vapor, $\mathbf{g}$ represents the gravitational acceleration, and μ , c_p , and k_{th} are viscosity, heat capacity, and thermal conductivity.

At the liquid-solid interface, $z = 0$, we apply the Dirichlet conditions, $u = w = 0$ and $\theta = \theta_w$. We consider the quasi-equilibrium evaporation, in which the liquid molecules interchange quickly between the interface and its vicinity where the gas phase is (quasi-)saturated and the interface temperature is its saturated value θ_s so that the Marangoni effect is absent. In addition to the mass balance (see Sec. I) at the gas-liquid interface, $z = h(x, t)$, the energy and stress balance equations take the following simplified form under the one-sided approximation:⁶ $\rho_g \ll \rho$, $\mu_g \ll \mu$, and $k_{th,g} \ll k_{th}$, which decouples the vapor dynamics from that of the liquid

$$j\tilde{L} = -k_{th}\nabla\theta \cdot \mathbf{n}, \quad (3)$$

$$j^2\rho_g^{-1} + \mathbf{n} \cdot \mathbf{T} \cdot \mathbf{n} = 2\kappa\sigma_0, \quad \mathbf{n} \cdot \mathbf{T} \cdot \mathbf{t} = 0, \quad (4)$$

where $\tilde{L}$ denotes the latent heat of the liquid, $\mathbf{T}$ is the stress tensor, and the mean curvature $\kappa = -(\nabla \cdot \mathbf{n})/2$. The viscous heating, kinetic energy of vapor particles, and conductive/convective heat fluxes through the gas phase are omitted in Eq. (3). We also note that a more general boundary condition should include a temperature jump across the evaporating interface to account for the non-equilibrium effect.²⁶ However, the temperature jump is negligible when the phase change rate is not too rapid (here $0 < E \ll 1$ for slow evaporation, see Table I), as shown by Ward and Stanga²⁷ experimentally. Consequently, we assume that the temperature of liquid and vapor phases

TABLE I. Thermophysical properties of a typical vapor-liquid system in equilibrium and the corresponding experimental conditions and dimensionless parameters with $|g| = 9.8 \text{ m s}^{-2}$. p_g denotes vapor pressure.

Property and parameter	Water ^a	Mercury ^b	1,2-ethanediol ^b
p_g (Pa)	611.65	37.45	10
θ_s (K)	273.16	373.15	297.15 ^c
$\Delta\theta$ (K)	9	0.4	4
h_0 (mm)	0.12	0.10	0.11
ρ (kg m ⁻³)	999.79	13351	1113.2
ρ_g (kg m ⁻³)	4.8546×10^{-3}	2.4213×10^{-3} ^d	2.5122×10^{-4} ^d
μ (mPa s)	1.791	1.245	16.482 ^e
k_{th} (W m ⁻¹ K ⁻¹)	0.561	9.475	0.254
σ_0 (mN m ⁻¹)	75.646	470.11	48.079 ^e
$\tilde{L}$ (J g ⁻¹)	2500.9	294.68 ^f	1005.3 ^g
M_w (g mol ⁻¹)	18.015	200.59	62.068
$j^2\rho_g^{-1}$ (Pa)	0.06	6.83	0.34
E	1.1272×10^{-3}	1.0330×10^{-2}	6.1315×10^{-5}
S	9.4290×10^2	1.3498×10^5	7.2240
$ G $	1.7586	375.68	1.9834×10^{-2}
D	7.2834×10^{-6}	2.7202×10^{-7}	3.3851×10^{-7}
$\mathcal{E}$	0.34	0.01	1.12
$ \mathcal{D} $	0.10	1.04	0.56
$\text{Bo} = G /S$	1.9×10^{-3}	2.8×10^{-3}	2.7×10^{-3}

^aCalculated using the NIST REFPROP program and Standard Reference Database 23, Version 8.0.

^bReference 35.

^cExtrapolation with an Antoine equation beyond the region where experimental measurements exist.

^dApproximated using the ideal gas law with $R_g = 8.3145 \text{ J mol}^{-1} \text{ K}^{-1}$, most accurate for monatomic gas at high temperature and low pressure, e.g., Hg here.

^eObtained by linear extrapolation.

^fEstimated with $\tilde{L}$ at a normal boiling point due to its small variation over the wide range of temperatures [298.15, 629.73] K and pressures [0.278 64, 101 325] Pa according to Ref. 30.

^gEstimated with $\tilde{L}$ in the range of [307, 384] K according to Ref. 31.

is continuous at the interface under the quasi-equilibrium condition. Equations (4) include the normal- and tangent-stress boundary conditions. The former balances the jump in the normal stress with curvature stresses. We have used the continuity of the tangential velocity along the interface ($\mathbf{v} \cdot \mathbf{t} = \mathbf{v}_g \cdot \mathbf{t}$) and ignored the Marangoni term by assuming the surface tension σ_0 to be constant in the tangent stress condition. These interfacial boundary conditions are supplemented with the kinematic condition, $j = \rho(w - h_t - uh_x)(1 + h_x^2)^{-1/2}$. Then the mass flux is related to the local interface temperature by the modified Hertz–Knudsen law:⁴ $\tilde{K}j = \theta_l - \theta_s \equiv \Delta\theta_l$. If $\Delta\theta_l = 0$, the interface is in thermal equilibrium, while for a net mass transfer, a vapor pressure driving force must exist, representing as the non-equilibrium parameter, $\tilde{K} = \theta_s^{3/2}(a\rho_g\tilde{L})^{-1}\sqrt{2\pi R_g/M_w}$.^{1,6} Here, R_g and M_w denote the universal gas constant and molar mass, and a is the accommodation coefficient ($0 < a \leq 1$).

We then introduce the following scalings:

$$\left. \begin{aligned} (X, Z, H) &= h_0^{-1}(x, z, h), \quad T = h_0^{-2}\nu t, \quad (U, W) = h_0\nu^{-1}(u, w), \\ P &= h_0^2\rho\mu^{-2}p, \quad J = \tilde{L}h_0(k_{th}\Delta\theta)^{-1}j, \quad \Theta = \Delta\theta^{-1}(\theta - \theta_s), \end{aligned} \right\} \quad (5)$$

where $\nu = \mu/\rho$ is the kinematic viscosity and $\Delta\theta = \theta_w - \theta_s$ is used as the temperature scale. The non-dimensionalization yields the dimensionless groups,

$$E = \frac{k_{th}\Delta\theta}{\mu\tilde{L}}, \quad S = \frac{h_0\sigma_0\rho}{3\mu^2}, \quad G = \frac{gh_0^3\rho^2}{3\mu^2}, \quad D = \frac{3\rho_g}{2\rho}. \quad (6)$$

The evaporation number, E , describes the rate of mass loss, S and G are the surface-tension and gravity numbers, and D is the density ratio. We assume that the thickness and flow velocity of the layer change slowly enough to justify the lubrication approximation^{1,6} and consider a long-wave disturbance in the x -direction by introducing a small wavenumber, $k = 2\pi h_0/\tilde{\lambda}$, with the characteristic length $\tilde{\lambda} \gg h_0$. The derivation of an evolution equation mostly follows the approach in Ref. 25 with the lubrication variables, $\xi = kX$, $\zeta = Z$, and $\tau = kT$. We define the asymptotic transformation,

$$(D, E, G, S) = (\bar{D}k^3, \bar{E}k, \bar{G}k^{-1}, \bar{S}k^{-3}), \quad (7)$$

to preserve the effects of vapor recoil, mass loss, gravity, and surface tension at leading order, where the quantities with overbar are $O(1)$ as $k \rightarrow 0$. In particular, to incorporate the effects of gravity, we let $G = O(k^{-1})$ to make it comparable with the pressure term. The dependent variables then are expanded for small k , $(U, W, J, \Theta, P) = (U_0, kW_0, J_0, \Theta_0, k^{-1}P_0) + k(U_1, kW_1, J_1, \Theta_1, k^{-1}P_1) + \dots$, in which the quantities with subscript “0” are of $O(1)$. As a result, we arrive at the set of $O(1)$ dimensionless equations,

$$0 = -\partial_\xi P_0 + \partial_\zeta^2 U_0, \quad 0 = -\partial_\zeta P_0 - 3\bar{G}, \quad (8)$$

$$H_\tau + \partial_\xi \int_0^H U_0 d\zeta + \bar{E}J_0 = 0, \quad (9)$$

$$0 = \partial_\zeta^2 \Theta_0, \quad (10)$$

where $H(\xi, \tau)$ is an undetermined $O(1)$ function, and Eq. (9) is the conservation form of the kinematic condition, obtained by integrating the rescaled continuity equation. The boundary conditions at $\zeta = 0$ read $U_0 = W_0 = 0, \Theta_0 = 1$, and at $\zeta = H$,

$$\bar{E}J_0 = -H_\tau - U_0H_\xi + W_0, \quad (11a)$$

$$J_0 = -\partial_\zeta \Theta_0, \quad (11b)$$

$$\frac{3}{2}\bar{E}^2\bar{D}^{-1}J_0^2 - P_0 = 3\bar{S}H_{\xi\xi}, \quad \partial_\zeta U_0 = 0, \quad (11c)$$

$$KJ_0 = \Theta_0, \quad (11d)$$

where $K = \tilde{K}k_{th}/(h_0\tilde{L})$ is the scaled non-equilibrium parameter. The non-evaporative, quasi-, and non-equilibrium cases correspond to $K \rightarrow \infty$, $K = 0$, and $K > 0$, respectively.

Finally, with the procedure outlined in [Appendix A](#), the system [Eqs. (8)–(11)] is collapsed into the following (2 + 1)-dimensional evolution equation for the quasi-equilibrium case:

$$H_T + EH^{-1} + S\nabla_H \cdot [H^3 \nabla_H (\nabla_H^2 H)] - G\nabla_H \cdot (H^3 \nabla_H H) + E^2 D^{-1} \nabla_H^2 H = 0, \quad (12)$$

where $\nabla_H = (\partial_X, \partial_Y)$ and ∇_H^2 are the components of the gradient vector and Laplacian parallel to the substrate with the y -coordinate for a cross horizontal stream in 3D case. Within the long-wave regime, the Bond number $\text{Bo} \equiv |G|/S = \rho|g|h_0^2\sigma_0^{-1} = O(k^2) \ll 1$ as $k \rightarrow 0$, Eq. (12) is a valid limit to the governing system only in an excluded time interval since the lubrication assumption breaks down near $T = 0$ and the disappearance time T_e , as explained in Ref. 6 and [Appendix B](#). Given the appropriate initial and boundary conditions, we can investigate the interfacial instability by solving Eq. (12) as an initial-value problem. The second and fourth terms on the left hand side (LHS) account for mass losses and gravity. The stabilization of the capillary force acts through a fourth-order dissipation term responsible for the viscous flow. The last term is associated with the vapor recoil. Note that the $O(1)$ temperature profile in the evaporating film is linear (pseudo-steady), the same as in the basic state [see Eqs. (A1) and (B10)], because they are governed by the pseudo-steady conduction equations (10) and (B5); thus the temperature fields cannot satisfy an arbitrary initial condition. However, Eq. (12) still contains most of the richness of the original dynamics, which turn out to be effectively governed.

Rupture patterns observed in the experiments with evaporating layers are often quite complicated; however, the fundamental physical effects responsible for evaporating interface dynamics can be demonstrated and explained with the (1 + 1)-dimensional version of Eq. (12),

$$H_T + EH^{-1} + S(H^3 H_{XX})_X - G(H^3 H_X)_X + E^2 D^{-1} H_{XX} = 0. \quad (13)$$

To make the presentation of results more concise, we introduce the rescaling,

$$\hat{x} = \sqrt{|G|S^{-1}}X = \sqrt{\text{Bo}}X, \quad \hat{t} = G^2 S^{-1}T, \quad (14)$$

to absorb the parameters G and S and examine Eq. (13) in its canonical form

$$H_{\hat{t}} + \mathcal{E}H^{-1} \pm [(H^3 + \mathcal{D})H_{\hat{x}}]_{\hat{x}} + (H^3 H_{\hat{x}\hat{x}})_{\hat{x}} = 0, \quad (15)$$

where $\mathcal{E} = ESG^{-2}$ and $\mathcal{D} = E^2(-GD)^{-1}$ are the modified evaporation and the vapor thrust numbers, characterizing the mass loss and interfacial pressure caused by departing vapor; “+” corresponds to $G < 0$, “−” for $G > 0$, and “^” will be suppressed in the rescaled independent variables hereafter.

III. NUMERICAL METHOD AND VALIDATION

The strongly nonlinear PDE (15) is solved with the periodic boundary conditions on $[0, \lambda)$, in which $\lambda = \tilde{\lambda}/h_0 = 2\pi/k$. With the temporal derivative retained, the spatial derivatives are calculated by a pseudo-spectral method on a uniform mesh. The resulting system of semi-discrete ODEs is integrated in time, referred to as the “method of lines”.²⁸ An implicit time-step scheme is implemented for the numerical stability. The initial conditions (ICs) are small-amplitude sinusoidal or pseudo-random disturbances,

$$\text{IC1: } H(\hat{x}, 0) = 1 + \varepsilon_0 \cos(2\pi\hat{x}/\lambda), \quad (16a)$$

$$\text{IC2: } H(\hat{x}, 0) = 1 + \varepsilon_0 [\sin(2\pi\hat{x}/\lambda) + \sin(4\pi\hat{x}/\lambda)], \quad (16b)$$

$$\text{IC3: } H(\hat{x}, 0) = 1 + m + \varepsilon_1 \text{Rand}(\hat{x}), \quad (16c)$$

where $|\varepsilon_0|$ and $|\varepsilon_1| \ll 1$, we take $\varepsilon_0 = 0.01$ and $\varepsilon_1 = -0.05$; $\text{Rand}(\hat{x})$ is the pseudo-random function in $(-1, 1)$, and m matches the initial average-thickness with unity.

The numerical analysis shows that the rupture time, t_r , depends on the grid resolution and temporal accuracy. We refine the grid until the occurrence of a stable convergence, and a further increase in the resolution does not change the results significantly. The grid-independency is tested for t_r and minimum thickness $H_{\min}$, both of which are the major concerns in the dynamical system. An example is shown in Fig. 2. It is found that t_r differs from the previous one by an amount

FIG. 2. The effects of the spatial grid on t_r and $H_{\min}$ for a quasi-equilibrium evaporating layer subject to RTI with $\lambda = 2\sqrt{2}\pi$, $\mathcal{E} = 0.05$, $\mathcal{G} = 1$, $N = 40, 50, 100, 150, 200$, and 250 .

from 0.0013 to 0.0072 for $N \leq 150$, a large difference for the rupture instability. These differences then reduce to $O(10^{-4})$ for $N > 150$, where t_r is not sensitive to N as well. On the other hand, the values of $H_{\min}$ are $O(10^{-5})$ and the variations are negligible in effect, whereas the rate of change is minimized with N between 150 and 250. Thus $N = 200$ is chosen for $\lambda = 2\sqrt{2}\pi$, and the other resolutions are determined similarly.

Consider the disjoining pressure associated with van der Waals attractions that would become important and lead to an instantaneous rupture if the effect was retained and the local thickness entered the range of 10–100 nm.^{6,25} If the initial thickness, h_0 , of order 1 mm corresponds to $H = 1$, $H = O(10^{-4})$ corresponds to the thickness of order 100 nm, then the integration is terminated once $H_{\min}$ becomes less than a threshold of $O(10^{-4})$. Accordingly, this instant is defined as t_r . In this way, the “rupture” here is never affected by this disjoining pressure as a thin “adsorbed” layer remains, and the touchdown is not completely dry, similar to the phenomena in Ref. 17. In addition, we shall be content to neglect the inertial effects under lubrication approximation which become significant just before the complete rupture by the disjoining pressure.^{1,6} However, to follow the evolution properly up to the “true” rupture or extend the theory to falling films with a Reynolds number of $O(1)$, one should add a van der Waals term,^{6,25} $A(H^{-1}H_X)_X$, on the LHS of Eq. (13) with the dimensionless Hamaker constant $A > 0$ and/or introduce inertial effects.

The time step is restricted to be consistent with the spatial resolution, and the temporal accuracy is set quite high; thus the error should come from the spatial discretization and accumulative errors. With the grid-independency evaluation, the error arising from the spatial mesh is minimized. Particularly, when $H_{\min}$ varies rapidly, the time step reduces until the solution satisfies the local relative error. The convergence is successfully achieved with the tolerance error of $O(10^{-20})$ for t_r . Therefore, our numerical solutions have been determined precisely. In addition, the numerical value of t_r may not be accurate to the decimal place reported here. We show the actual value used due to the computational sensitivity and take only three significant decimal digits for comparison.

To validate the numerical method, we consider the simplest case of non-evaporative isothermal layers subject to pure RTI and compare with the results of Yiantsios and Higgins.¹⁸ Furthermore, a special case is checked against Burelbach’s results⁶ in Sec. V B. When $E = 0$ and $G < 0$, Eq. (13) has the canonical form

$$H_t + (H^3 H_x)_x + (H^3 H_{xxx})_x = 0, \quad (17)$$

which governs the RTI of a non-evaporative layer subject to the gravity and capillarity. A similar equation has been used in Refs. 18, 19, and 28. We examine the linear stability by perturbing the basic state with $H = 1 + H'(x, t)$. Substituting it into Eq. (17) and linearizing, one obtains $H'_t + H'_{xx} + H'_{xxx} = 0$. We employ the normal mode to express its solution in the exponential form, $H'(x, t) = H'_0 \exp(st + ikx)$ with $|H'_0| \ll 1$. Substitution of the normal mode into the stability equation yields $s = k^2(1 - k^2)$. The cutoff wavenumber $k_c = 1$ and maximum growth rate $s_m = 1/4$, corresponding to $\lambda_m = 2\sqrt{2}\pi$. The numerical solutions of Eq. (17) with IC (16a) are presented in Fig. 3. One can observe the superposition of higher-order harmonics as a result of the nonlinear

FIG. 3. Interface evolution of Rayleigh-Taylor unstable non-evaporative isothermal layers during $t_{\max} = 100$ with $\Delta t = 2$ presented for half-period: (a) $\lambda = \lambda_m$; (b) $\lambda = 2\lambda_m$; (c) $\lambda = 3\lambda_m$.

growth of the interface instability, taking the familiar form of long-wave pendent droplets. At the early stage, the disturbance grows at an exponential rate according with LSA and the interface patterns are dominated by the Fourier mode of maximum growth rate, as presented in Ref. 18. The slight differences in the amplitude are identifiable, since our results are obtained from the rescaled equation different from theirs by a factor of 1/3. The agreement validates the reliability of our numerical method.

IV. TIME-DEPENDENT LINEAR STABILITY ANALYSIS: VAPOR-RECOIL DYNAMICS

The leading-order basic-state solution to the non-dimensional governing system is given in Appendix B. The basic-state thickness, $\bar{H} = (1 - 2\mathcal{E}t)^{1/2}$, vanishes at $t_e = 1/(2\mathcal{E})$, where “ $\wedge$ ” is suppressed. We examine the time-dependent linear stability by perturbing Eq. (15) with $H = \bar{H} + H'$ and linearizing in H' ,

$$H'_t - \mathcal{E}\bar{H}^{-2}H' \pm (\bar{H}^3 + \mathcal{D})H'_{xx} + \bar{H}^3H'_{xxx} = 0. \quad (18)$$

One substitutes the normal mode $H' = A(t)e^{ikx}$ into Eq. (18) to obtain the characteristic equation for the growth rate, s ,

$$s = A_t A^{-1} = \mathcal{E}\bar{H}^{-2} \pm (\bar{H}^3 + \mathcal{D})k^2 - \bar{H}^3k^4, \quad (19)$$

where the time-dependent normal-mode amplitude, $A(t) \equiv H'_0 e^{st}$, is introduced. The general solution of this ODE governs the full dynamics of the disturbed basic state,

$$A(t) = A_0 \exp\left[\int_0^t (\cdot) dt\right] = \frac{A_0}{\bar{H}} \exp\left\{\frac{k^2(1 - \bar{H})}{\mathcal{E}}\left[\pm \frac{1}{2}\mathcal{D}(1 + \bar{H}) + \frac{1}{3}(\pm 1 - k^2) \sum_{n=0}^4 \bar{H}^n\right]\right\}, \quad (20)$$

where “+/-” corresponds to $G < 0/G > 0$, the upper limit of integral $t < t_e$, $(\cdot)$ denotes the right hand side of Eq. (19), and A_0 is the integration constant. The coefficient proportional to $\bar{H}^{-1}$, resulting from mass loss, has an algebraic contribution and no effect on the exponential stability. With the exponent in Eq. (20) divided by t to exclude the algebraic variation, an effective growth rate can be defined as follows:^{6,9}

$$s_{\text{eff}}(t) = \frac{2k^2}{1 + \bar{H}} \left[\pm \frac{1}{2}\mathcal{D}(1 + \bar{H}) + \frac{1}{3}(\pm 1 - k^2) \sum_{n=0}^4 \bar{H}^n\right], \quad (21)$$

where $\bar{H} = (1 - 2\mathcal{E}t)^{1/2}$ has been used. A positive value of s_{eff} corresponds to the linear instability. And the time-dependent cutoff wavenumber $k_c^2(t) = \pm [1 + 5\mathcal{D}(1 + \bar{H})/(2 \sum_{n=0}^4 \bar{H}^n)]$, given by $s_{\text{eff}}(t) = 0$. Note that $\mathcal{D} > 0/\mathcal{D} < 0$ for $G < 0/G > 0$. It follows from Eq. (21) that for $G < 0$ the instability is derived from the vapor recoil and Rayleigh-Taylor mechanism while the stabilization is due solely to the surface tension, and for $G > 0$ the instability arises from the vapor recoil only while stabilized by gravity additionally. There are two limiting cases: (1) $s_{\text{eff}} \rightarrow k^2[(\pm 1 - k^2) \pm \mathcal{D}]$,

FIG. 4. s_{eff} as a function of t and k for quasi-equilibrium evaporating layers for (a) $G < 0$ and (b) $G > 0$. The dispersion curves present the increase in $s_{\text{eff,m}}$ and broadening in the band of linearly unstable modes $0 < k^2 < k_c^2(t)$. The parameter $|\mathcal{D}| = 2$ for the illustration of the vapor recoil effect, which could be feasible for a relatively small ambient pressure (density).

$k_c^2 \rightarrow \pm(1 + \mathcal{D})$ as $t \rightarrow 0$ and (2) $s_{\text{eff}} \rightarrow k^2[2(\pm 1 - k^2)/5 \pm \mathcal{D}]$, $k_c^2 \rightarrow \pm(1 + 5\mathcal{D}/2)$ as $t \rightarrow t_c$. A maximum effective growth rate, $s_{\text{eff,m}}$, occurs at $k_m^2 = k_c^2/2 = \pm[1/2 + 5\mathcal{D}(1 + \bar{H})/(4 \sum_{n=0}^4 \bar{H}^n)]$. These reflect the fact that as time goes on an initially perturbed surface has its thin regions thinned faster under vapor thrust while the surface tension cuts off the shorter wavelength. As indicated by Fig. 4, both the unstable interval and $s_{\text{eff,m}}$ increase with time. It implies that the interface instability is enhanced by vapor recoil, $\mathcal{D}$, which competes with the stabilizing surface tension in both cases of $G < 0$ and $G > 0$. The destabilizing mechanism makes sense physically because the $O(1)$ basic-state temperature difference across the thinning layer is constant [actually unity for $K = 0$, see the temperature profile in Eq. (B10)]; the heat/mass fluxes and thus momentum flux associated with vapor thrust become greater as it thins out. Comparison of the magnitude of s_{eff} for $G < 0$ and $G > 0$ indicates that s_{eff} is significantly larger with $G < 0$ as RTI reinforces the vapor-recoil effect through the creation of depression while the hydrostatic pressure stabilizes the surface against the vapor thrust with $G > 0$. It is thus suggested that the rupture would be remarkably earlier with $G < 0$ provided the mass-loss effect is much smaller than that of vapor recoil (recalling s_{eff} defined by excluding the contribution of $\mathcal{E}$). This is consistent with the physical nature of evaporation as illustrated by the subsequent nonlinear simulations.

The most unstable perturbation is expected to grow faster than the basic-state vanishing that one can suppose a frozen basic state at $\bar{H}(0) = 1$ to compare with the quasi-steady LSA.^{4,6,9} Equation (19) becomes $s_s = \mathcal{E} \pm (1 + \mathcal{D})k^2 - k^4$ with subscript “s” for the quasi-steady theory, which gives a factitious instability due to the nature of thinning, i.e., $s_s = \mathcal{E} > 0$ even for the undisturbed interface ($k = 0$). The cutoff wavenumber is $k_{c,s}^2 = [\pm(1 + \mathcal{D}) + \sqrt{(1 + \mathcal{D})^2 + 4\mathcal{E}}]/2$. With $\mathcal{E} = 0$ to eliminate the algebraic contribution again, one finds $s_s = \pm(1 + \mathcal{D})k^2 - k^4$ and $k_{c,s}^2 = \pm(1 + \mathcal{D})$, corresponding to the asymptotically initial s_{eff} -curve from the time-dependent theory, i.e., $s_{\text{eff}}(t) \rightarrow s_s$ and $k_c(t) \rightarrow k_{c,s}$ as $t \rightarrow 0$.

V. TIME-DEPENDENT NONLINEAR SIMULATIONS: EVAPORATION-DRIVEN RUPTURE

Time-dependent LSA suggests that the rupture instability is enhanced due to the vapor recoil with the effective growth rate of the instability. However, our LSA is valid when the perturbation amplitude is much smaller than the layer thickness with weak mass loss. To follow the evolution approaching to rupture in strong nonlinear regimes and/or investigate the case in which the mass-loss effect is comparable to that of vapor recoil, the evolution equation should be solved numerically. The rest of this paper is devoted to the numerical treatment of Eq. (15) in two evaporation cases of strong vapor thrust [$\mathcal{D} = O(1)$]: case I for the weak mass loss of $\mathcal{E} = O(10^{-2})$ and case II for the moderate mass loss of $\mathcal{E} \approx 0.1 - 1$. Hereafter, we will refer to the two cases frequently. Before proceeding to our simulations, it is beneficial to take a couple of typical fluids and calculate the non-dimensional numbers using their thermodynamic properties to place them into aforementioned broad categories.

A. Typical working fluids

Several experiments have been conducted to examine the interfacial convection and heat-transfer characteristics in evaporating liquid layers or liquids in pools at the reduced pressure;^{3,10,27,29} however, no experimental investigations quantitatively identify the interfacial rupture rate and pattern of such thin-film systems due to difficulties in establishing, maintaining, and measuring a thin evaporating layer at subatmospheric pressures. Nevertheless, to substantiate the cases of interest, we quantify the dimensionless numbers characterizing the applicability of Eq. (12); thus the numerical analyses can be used to predict the regime and trend of experimental consequences. In particular, the first LHS-term of the normal-stress balance in Eq. (4) represents the vapor thrust, which can be considered as an externally imposed normal stress. We can estimate its magnitude with

$$j^2 \rho_g^{-1} \sim \frac{1}{\rho_g} \left(\frac{k_{th} \Delta \theta}{\tilde{L} h_0} \right)^2, \quad (22)$$

where Eq. (3) has been used. As a result of the increase in the local temperature gradient, the destabilizing influence of the vapor recoil is expected to be reinforced with touchdown.

We examine the typical vapor-liquid equilibrium systems with the material properties in Table I and calculate the dimensionless groups in Eq. (6) and the control parameters, $\mathcal{E}$ and $\mathcal{D}$. These fluids were selected for their wide variations in properties, relatively low vapor pressures and thus low D to make $\mathcal{D} = O(1)$ possible, and relatively high surface tension to obey $Bo \ll 1$. Mercury, a metallic liquid at standard conditions, is used in the mercury-vapor lamp and in the dropping mercury electrode or hanging mercury drop electrode for an electrochemical study. It seems to be a good candidate to present case I. As the data shown in Table I, it allows relatively small $\Delta \theta$ due to its small Prandtl number ($Pr \approx 0.02$) and thus small $\mathcal{E}$ with the order-of-magnitude larger in ρ . On the other hand, the major use of organic compound 1,2-ethanediol is a medium for the convective heat transfer in, for example, automobiles and liquid cooled computers. It has a relatively low vapor pressure at normal temperature; thus the importance of vapor recoil increases markedly as the pressure is reduced. The high viscosity and medium density produce an $\mathcal{E}$ of moderate magnitude. Therefore, 1,2-ethanediol appears to be a proper liquid to demonstrate the dynamics of case II. In addition, one could use water under the recommended conditions for another case of moderate evaporation where $\mathcal{E} \approx 0.1$ – 1 and $\mathcal{D} = O(0.1)$. The link to the real system serves as a motivation to test our numerical predictions experimentally.

In the experiments, the initial layer of the working fluid, lying on a temperature-controlled plate and subjected to superheat $\Delta \theta$, will evaporate in a pressure-controlled chamber free of non-condensable gas. Since the liquid is evaporating into its own vapor, $\Delta \theta$ could be controlled directly by changing the pressure of the vapor, which allows a larger $\Delta \theta$ range than a typical configuration, where a liquid layer evaporates into ambient air. As the pressure is gradually reduced with the liquid properties (ρ , μ , k_{th} , σ_0 , and $\tilde{L}$) insensitive to the pressure, the velocity of the vapor particles departing/impinging the interface increases and the density ratio, D , reduces that significantly increasing the experimental value of $\mathcal{D}$ thus reducing the stability threshold of the system. The dynamic thickness can be measured using a system consisting of a high speed camera and ultrasound ranging sensors until rupture.

B. Evaporating layer subject to negative gravity

The interface evolution is presented in Figs. 5 and 6 for $G < 0$ and $\lambda = \lambda_m$ or $2\lambda_m$, where $\lambda_m = 2\sqrt{2}\pi$ is the fastest-growing mode for a non-evaporative isothermal layer from the LSA. As shown in Figs. 5(a), 5(b), 5(e), 6(a), and 6(b) for case I with different ICs, one can observe that the layers thin nearly uniformly in a time of $O(t_r/3)$ (indicated by a dot-dashed line) as a result of the initially significant effects of the mass loss and stabilizing surface tension. The trough tends to flatten due to the Laplace pressure and a minor nonlinear evolution occurs during the subsequent adjustment period. The destabilizing effects of the vapor recoil and negative gravity then prevail over the stabilization of the surface tension, manifested by the rapid development of new

FIG. 5. Spatio-temporal evolution of quasi-equilibrium evaporating layers for $G < 0$ (RTI) and $\mathcal{G} = 1$ with symmetrically sinusoidal perturbation IC1. ((a) and (c)) $\lambda = 2\sqrt{2}\pi$ with $N = 200$ grid: (a) case I ($\mathcal{E} = 0.05$) at $t = 1, 2, 3, 4, 4.5, 4.9, 5.12, 5.23$, and t_r ; (c) case II ($\mathcal{E} = 0.5$), corresponding to $0 \leq t \leq 0.9$ with $\Delta t = 0.1$, and instants of 0.96 and t_r . ((b) and (d)–(f)) $\lambda = 4\sqrt{2}\pi$ with $N = 400$ grid: (b) case I ($\mathcal{E} = 0.03$) at $t = 1, 3, 5, 7, 8.9, 9.7, 10, 10.3$, and t_r ; (d) case II ($\mathcal{E} = 0.3$), corresponding to $0.1 \leq t \leq 1.5$ with $\Delta t = 0.2$, and instants of $1.6, 1.616$, and t_r ; (e) case I ($\mathcal{E} = 0.05$), corresponding to $0 \leq t \leq 8$ with $\Delta t = 1$, and instants of $8.5, 8.6, 8.7, 8.77, 8.79$, and t_r ; (f) case II ($\mathcal{E} = 0.5$), corresponding to $0 \leq t \leq 0.9$ with $\Delta t = 0.1$, and instants of 0.97 and t_r . Red arrows track the two $H_{\min}$ positions in subfigures ((a) and (b)) and $\uparrow$ indicates the adjustment period. Initial disturbances are dashed.

troughs/crest(s) to conserve mass under weak mass loss. Eventually, the interfaces rupture symmetrically at two points for IC1 and at a single point for IC2 and IC3 with a large contact angle (or small radius of curvature). The two-point rupture is reminiscent of the ring-like rupture (for axisymmetric problem) in Figs. 2 and 7 of Refs. 23 and 24, instead of the general single-point mode driven by van der Waals forces, see, e.g., Refs. 6 and 22.

Particularly, in Figs. 5(a) and 5(b), the growth rate of the main droplet (highest one) seems to follow the exponential rate of the linear theory until the neighborhood of t_r . Its growth rate reduces with decrease in k (even $\mathcal{E}$ minished from 0.05 to 0.03, recalling that $\mathcal{E}$ has only an algebraic contribution) which agrees with the LSA, that is, s_{eff} lowers as k decreases on $(0, k_m]$, shown in Fig. 4(a). For this reason, we use $\mathcal{E} = 0.03$ in Fig. 5(b) for the longer wavelength to compensate the decrease in the rate of nonlinear growth and allow the growth in the main droplet. The bottom widths of the main droplets are both about $(2\sqrt{2} - 1)\pi$, while their heights decrease with increasing λ in contrary to the non-evaporative situation (see Fig. 3).

When $\mathcal{E} = 0.05$ is held in Fig. 5(e) to isolate the effects of the wavelength, no growth in the crest of the main droplet is observed and it equilibrates at ~ 0.57 since $t = 8$. The larger t_r and smaller surface wave amplitude with a longer transition time of monotonically thinning demonstrate that a longer wavelength is indeed more stable. The secondary droplet in Fig. 5(b) consists of two

FIG. 6. Spatio-temporal evolution of quasi-equilibrium evaporating layers for $G < 0$ (RTI), $\mathcal{D} = 1$, and $\lambda = 2\sqrt{2}\pi$. ((a) and (c)) With an asymmetrically sinusoidal perturbation IC2 and $N = 200$ grid: (a) case I ($\varepsilon = 0.05$) at $t = 1, 2, 3, 3.5, 4, 4.2, 4.3$, and t_r ; (c) case II ($\varepsilon = 0.5$), corresponding to $0 \leq t \leq 0.9$ with $\Delta t = 0.1$, and instants of 0.91 and t_r . ((b) and (d)) With a random perturbation IC3 of 50 sample points and $N = 1000$ grid: (b) case I ($\varepsilon = 0.05$) at $t = 1, 2, 3, 4, 4.1, 4.2, 4.3, 4.4, 4.45$, and t_r ; (d) case II ($\varepsilon = 0.5$), corresponding to $0 \leq t \leq 0.9$ with $\Delta t = 0.1$, and instants of 0.96, 0.97, 0.98, and t_r . $\uparrow$ indicates the adjustment period. Initial disturbances are dashed.

partially coalesced ones originating from the intrinsic dynamics of nonlinear interactions between the main mode and higher-order harmonics. On the contrary, the main droplet consists of three partially coalesced ones, while the secondary droplet is unimodal in Fig. 5(e). This implies that a wider lobe is more susceptible to the interfacial instability to become multihumped due to a smaller Laplace pressure. Comparison of t_r in Figs. 5(b) and 5(e) indicates that a larger ε leads to an earlier rupture with thinner droplets. The subtle influences of ε will be evident when proceeding to case II.

Due to the larger thickness of the main droplet, the viscous force is lower there relative to the secondary one (intervening lower droplet). Thus more fluid from the pinch region is collected by the main droplet that results in its fast growth [see Figs. 5(a) and 5(b)] or sustains a local balance between the mass loss and flow of liquid there [see Fig. 5(e)]. The gradually decreasing amount of liquid remaining in the deepened regions is driven by increasing capillary-pressure gradients under favorable viscous-resistance gradients. Thus the local acceleration of liquid grows indefinitely, and the rupture of the interface corresponds to a finite-time singularity of the evolution

FIG. 7. $H_{\min}$ and $\hat{P}_1$ evolution of the evaporating layer for $G < 0$ (RTI), $\mathcal{D} = 1$, $\varepsilon = 0.05$, and $\lambda = 2\sqrt{2}\pi$ with a sinusoidal perturbation IC1. (a) $-dH_{\min}/dt$ versus $H_{\min}$: computation extended until $H_{\min}$ reaches $O(10^{-5})$ (denoted by bracket) using an adaptive grid scheme with $N_{\max} = 1000$, after a transient it has slope -1 . (b) $\hat{P}_1$ corresponding to the instants in Fig. 5(a) until $t = 5.28$ (dashed line) using $N = 200$ grid.

equation (15), in which the local acceleration and interface gradients diverge. The inspection of Figs. 5(a), 5(b), and 5(e) indicates that the evolution mode of Fig. 5(a) eventually dominates the case I evaporation with $G < 0$ and the symmetric initial condition. The analysis on case I demonstrates an evaporation-dominant pendent droplet pattern. It is found that the interaction between the quasi-equilibrium evaporation and RTI causes the long-wave pendent droplets divided by a sharp dry-out point instead of coarsening into large and sparse droplets in the absence of evaporation, see Fig. 6(c) in Ref. 11, Figs. 7(b), 9(a), and 9(b) in Ref. 18, or Fig. 2 in Ref. 19 for large drops separated by wide drained regions. And the droplet thickness is notably reduced due to the mass-loss effect when compared with that in the absence of evaporation, cf., Figs. 3(a) and 3(b), for example. With analogous conditions (RTI and no Marangoni effect), a similar conclusion on the long-wave characteristics was reached by Bestehorn and Merkt¹¹ with a 3D simulation. However, the dissimilarity is the rupture event of the large curvature in our case due to the incorporation of significant vapor recoil.

As depicted in Figs. 5(c), 5(d), and 5(f) for case II with IC1, t_r reduces roughly by one order of magnitude with a different pathway of evolution (no excited-mode grows drastically or quasi-steady profile surrounds the rapidly growing valley near t_r) from the relevant case I as $\mathcal{E}$ is increased by one order. There is no chance to develop a trapped secondary droplet between two rupture points in case II even with a strong vapor recoil, caused by a sufficient surface tension (S) and mass-loss rate (E) for a moderate value of $\mathcal{E}$ (recalling $\mathcal{E} = ESG^{-2}$). These points suggest that both t_r and evolution pattern are sensitive to $\mathcal{E}$. The layers thin nearly uniformly with gradually increased velocity due to the initial actions of mass loss and capillarity again. The local depression then deepens rapidly to form a valley and eventually destabilizes into a middle rupture with a relatively smaller contact angle. It is explained that the gradually enhancing effect of the vapor recoil prevails over the local surface tension as it thins out that accords with the LSA. The evolutions in Figs. 5(c) and 5(f) are very similar except for the drop thickness, which decreases with increasing λ , while the rupture becomes faster for reduced λ (or increased k). This is ascribed to (1) s_{eff} increases on $(0, k_m]$ [see Fig. 4(a)] and (2) less mass loss occurs in a shorter duration. The comparison between Figs. 5(d) and 5(f) reveals that an increase in moderate $\mathcal{E}$ has a minor effect on the rupture pattern but leads to a remarkably earlier rupture. This is in sharp contrast to case I of $\mathcal{E} = O(10^{-2})$ where an increase in $\mathcal{E}$ causes a significant change in the pattern formation but a secondary influence on t_r , cf., Figs. 5(b) and 5(e).

Figures 6(a) and 6(c) illustrate the evolution of the asymmetrically sinusoidal IC2 (16b) for case I and case II with $\lambda = 2\sqrt{2}\pi$. Aside from a nearly uniform thinning in the incipience, both cases proceed with an asymmetric pathway, ending up with two partially coalesced droplets of different sizes and one noncentral dry-out spot, which essentially corresponds to the initial $H_{\min}$. Both $t_r = 4.389$ and 0.917 are somewhat earlier than the respective counterpart with symmetric IC shown in Figs. 5(a) and 5(c), while both t_r for case I are much larger than that for case II because of the weaker mass loss. With the random IC3 (16c), Figs. 6(b) and 6(d) reveal new scenarios for cases I and II evaporation-driven rupture, respectively. The rupture spot translates randomly with a slight change in t_r among successive runs for different pseudo-random initial seeds. The rupture usually does not occur in the center of the domain, just as the observation in Ref. 32, and not correspond to the initial $H_{\min}$. This is in contrast to the cases with the (a)symmetric ICs, in which the rupture always occurs in the initial $H_{\min}$ or two symmetric locations with respect to it. Nevertheless, the computational costs increase exponentially for a random IC, since the irregular undulation in the initial data cannot be handled reliably for the same accuracy as sinusoidal disturbances without a substantial increase in grid points. For this reason, we use $N = 1000$ grid in the simulations with IC3.

The 2D evolution depends on the type of ICs; however, the dependence of its topological structure is weak, as found by Oron³³ as well, especially its behavior close to the rupture does not, such as with $\lambda = 2\sqrt{2}\pi$ recalling Figs. 5(a), 6(a), and 6(b) for case I, and Figs. 5(c), 6(c), and 6(d) for case II. Particularly, in Figs. 6(a) and 6(b), there are slight differences in t_r and heights of droplets with a phase shift between the two patterns. Thus the 2D solutions for sinusoidal disturbances capture the crucial spatial transformations with small computational cost. Therefore, a sinusoidal perturbation has been adopted for most 2D computations. However, it is necessary to employ a

random IC, which is more realistic, in a 3D simulation for the study of the pattern formation in view of the strong dependence of 3D dynamics on ICs.³³ For example, as shown in Fig. 1(b), we obtained the 3D rupture pattern as the counterpart of Fig. 6(b). Furthermore, these results imply that only 1-point rupture appears to be stable to a general disturbance, as verified in Sec. VI.

The rupture pattern in Fig. 5(a) represents two droplets of different sizes isolated by a sharp dry-out spot. The plot of $-dH_{\min}/dt$ versus $H_{\min}$ in Fig. 7(a) suggests $H_{\min} \sim (t_r - t)^{1/2}$ near rupture; thus the growth rate of the troughs varies as $(t_r - t)^{-1/2}$, which increases as $t \rightarrow t_r$ and becomes much larger than the disturbance growth rate predicted by the linear theory extrapolated to t_r . Now, the vapor recoil is considered as the main destabilizing effect to which the capillarity counteracts. Therefore we could specify the rescaled effective interfacial pressure, $\hat{P}_1(x, t) \equiv P_{10}|G|^{-1} = 3k(|\mathcal{D}|H^{-2}/2 - H_{xx})$, as the dimensionless criterion characterizing the relation of these two effects, which is obtained by setting $K = 0$ and $\zeta = H$ in Eq. (A2) and using Eqs. (7) and (14). As plotted in Fig. 7(b), the two pressure peaks of the power-law growth are responsible for the rapid evolution toward the pattern in Fig. 5(a). The pressure peaks are finite and differentiable anytime but approach two discontinuity points as $t \rightarrow t_r$, exactly corresponding to the rupture locations. The local curvature increases rapidly at the valley, suggesting an increasing Laplace pressure. Meanwhile, the local vapor recoil, varying as $(t_r - t)^{-1}$, augments more quickly. It turns out that the peak value of $\hat{P}_1$ varies as $(t_r - t)^{-0.7}$ near rupture, which reflects the significant stabilization of the surface tension. From a physical point of view, the vapor thrust is stronger at troughs, where the interfacial momentum transfer is greater and augments rapidly under the quasi-equilibrium condition. On the other hand, the capillary pressure increases quickly at a comparable degree to counteract the aforementioned destabilization at the valley. However, the vapor recoil is strong enough to lead to rupture eventually. Thus the liquid is squeezed into the droplets to reduce the free energy that resulting in the rapid growth in crests and troughs simultaneously.

Figure 8 illustrates the effects of the interaction between the mass loss and vapor thrust on the rupture instability when the gravity is negligible or significant ($G = 0$ or -1) compared with the surface tension ($S = 1/3$). The results are obtained by solving the unrescaled Eq. (13) with IC: $H(X, 0) = 1 + 0.1 \sin(kX)$ for $k = \sqrt{3}/2$. When the gravity is neglected [Fig. 8(a)], our equation reduces to a special case in Ref. 6, see Fig. 10 in their paper for the case without a body force of the van der Waals attraction. If we follow them with $N = 30$, we recover their results within a relative error of 2%, which further confirm our numerical method. However, our grid-independency test suggests that both t_r and $H_{\min}$ depend on the grid when $N = 30$, but not when $N = 120$; hence our results in Fig. 8(a) should be more accurate. Figure 8(b) shows the $H_{\min}$ evolution of a layer subject to RTI with different evaporative conditions. If both evaporative effects are negligible ($E, E^2D^{-1} \rightarrow 0$), the non-evaporative isothermal layer is recovered. If $E = 0.1, E^2D^{-1} \rightarrow 0$, it ruptures at $T_r = 2.947$ owing solely to the mass loss. If $E \rightarrow 0, E^2D^{-1} = 1$, breakup occurs faster at $T_r = 1.065$ resulting from the vapor thrust. With both effects included ($E = 0.1, E^2D^{-1} = 1$), it ruptures in the fastest manner, $T_r = 0.956$. Finally, the destabilization of the negative gravity is revealed by comparing the relevant curves and T_r for the same evaporation conditions between Figs. 8(a) and 8(b).

FIG. 8. $H_{\min}$ evolution of quasi-equilibrium evaporating layers with different degrees of mass loss and vapor thrust for $k = \sqrt{3}/2$ and $S = 1/3$ using $N = 120$ grid. Dashed line indicates the unperturbed thickness $H = 1$. (a) $G = 0$; (b) $G = -1$.

FIG. 9. Spatio-temporal evolution of quasi-equilibrium evaporating layers for $G > 0$ and $\mathcal{D} = -1$. ((a) and (c)) $\lambda = 2\sqrt{2}\pi$ with a symmetrically sinusoidal perturbation IC1 and $N = 200$ grid: (a) case I ($\mathcal{E} = 0.05$) at $t = 1, 2, 3, 4, 5, 6, 7, 7.5, 8, 8.15, 8.36$, and t_r ; (c) case II ($\mathcal{E} = 0.5$), corresponding to $0 \leq t \leq 0.9$ with $\Delta t = 0.1$, and instants of 0.95, 0.97, t_r . ((b) and (d)–(f)) $\lambda = 4\sqrt{2}\pi$: (b) case I ($\mathcal{E} = 0.03$) with IC1 and $N = 400$ grid, at $t = 1, 3, 5, 7, 9, 11, 13, 13.8, 14.01, 14.14$, and t_r ; ((d)–(f)) with different pseudo-random perturbations IC3 of 50 sample points and $N = 1000$ grid, (e) case II ($\mathcal{E} = 0.5$) at $0 \leq t \leq 0.9$ with $\Delta t = 0.1$, and instants of 0.93 and t_r ; ((d) and (f)) case I ($\mathcal{E} = 0.03$), at $t = 1, 3, 5, 7, 9, 11, 11.5, 12, 12.1, 12.13, 12.15$, and t_r in subfigure (d); and $t = 1, 3, 5, 7, 9, 11, 12, 12.5, 12.8, 12.9, 12.93$, and t_r in subfigure (f). Initial disturbances are dashed.

C. Evaporating layer subject to positive gravity

For the evaporating layers sitting on the substrate ($G > 0$ and $\mathcal{D} < 0$), Fig. 9 presents the representative solutions of Eq. (15) for $\lambda = \lambda_m$ and $2\lambda_m$ with the IC1 and IC3. The comparisons between Figs. 9(a), 9(b) and 5(a), 5(b) illustrate the difference between the stabilizing and destabilizing gravity. In Figs. 9(a) and 9(b), $t_r = 8.421$ and 14.169 are longer than their RTI counterparts by 59.2% and 35.4%, respectively. And only one droplet/dry-out point fits the interval without growth in the crest since the positive gravity tends to fill the trough, which should not be attributed to the symmetry of IC1 but a consistent pattern for $G > 0$, as attested by the evolution from two different IC3 shown in Figs. 9(d) and 9(f). The vapor thrust must overcome both stabilizations of the gravity and surface tension to rupture the interface that t_r will be more or less larger than its RTI counterpart depend on the evaporative conditions. The stabilizing effect of the gravity, however, is significant in the early stage and dominated by the local vapor recoil later on, which is suggested by the rapidly localized dimpling following a nearly uniform thinning (see also Sec. V D).

As shown in Fig. 9(c) for case II, $t_r = 0.976$ is larger than that of its RTI counterpart [see Fig. 5(c)] by merely 1.1% and reduces again by one order of magnitude from its weak mass-loss counterpart [see Fig. 9(a)]. The recognizable difference in the heights of the resulting droplets between Figs. 9(c) and 5(c) can be ascribed mainly to the opposite hydrostatic pressure. Its evolution and rupture pattern are also analogous to that shown in Figs. 5(c), 5(d), 5(f), 6(c), and 6(d) for case II with $G < 0$, indicating

that the moderate $\mathcal{E}$ could dominate over the gravity effect. It is worth noting that, in contrast to the scenarios in case II, for all the case I with $G < 0$ or $G > 0$, the local depression deepens quickly in the later stage to form a narrow and deep valley in a certain “similar” way, which suggests that the vapor thrust along with the surface tension ultimately controls the local dynamics of rupture regardless of the direction of the gravity (see Sec. VI for further discussion). We can say that the quasi-equilibrium rupture pattern is (1) sensitive to the direction of gravity with small $\mathcal{E}$ of $O(10^{-2})$ but not with a moderate $\mathcal{E}$ of $O(10^{-1})$, (2) rather susceptible to $\mathcal{E}$ for both $G > 0$ and $G < 0$, and (3) seems not very sensitive to λ with a fixed direction of gravity and similar evaporative conditions, as seen in Figs. 5(a), 5(e) and 5(c), 5(f) for $G < 0$ and Figs. 9(a), 9(b) and 9(c), 9(e) for $G > 0$.

For case I with $G > 0$, capillary ridges can emerge around the valley in general, see Figs. 9(a), 9(b), 9(d), and 9(f). They are driven by the increasing capillary-pressure gradients, which accelerate the local liquid outwards against the viscous resistance as $H_{\min}$ decreases rapidly. The capillary ridges become well-defined for a larger λ and/or a smaller $\mathcal{E}$. There are two reasons related to the latter as follows: (1) the fluid, squeezed quickly into the edges of the droplet, overcompensates the amount of evaporation there (low E) and (2) the replenishing flow causes a fast increase in the local normal viscous stresses under the viscous resistance, generating a local pressure capable of deforming the adjacent surface of a relatively weaker capillary pressure (small S). The well-developed ridges could obviously exceed the level of the center of the droplet, where a shallow valley is formed under the mass loss and vapor recoil, as illustrated in Fig. 9(d) that obtained with the random IC3. In addition, the predicted sharp rupture point can recall the numerical results of Tan *et al.*³⁴ without the surface tension. In the present cases, however, it is due to (1) the temperature difference across the layer is constant to precipitate the locally intense vapor recoil, and (2) the absence of the thermocapillarity arising from a non-equilibrium effect, which would allow viscous stresses to spread and uniformize underneath a depression. It should be mentioned that the vapor recoil is significant in the case of sufficient heat flux and/or low ambient pressure (see Sec. V A).

In the solutions of case II with random or asymmetric ICs shown in Figs. 6(c) and 6(d) for $G < 0$ and Fig. 9(e) for $G > 0$, the rupture patterns appear as two partially coalesced droplets, which are inherited from the multi-bump/dimple interface after the self-organization of the small-amplitude noise or asymmetric perturbation driven by the initial linear instability.

It should be emphasized that the interface evolution given for the specific random ICs in Figs. 6(b), 6(d), and 9(d)–9(f) are representative, based on our numerical experiments using different pseudo-random ICs (made repeatable with a pseudo-random generator). There was no qualitative change in the dynamics/stability of the interface for other random ICs. Specifically, we found finite quantitative variations in the form of the phase shift with slight differences in t_r and droplet thickness between patterns evolved from different IC3. A pair of examples in Figs. 9(d) and 9(f) manifests that their dynamics are insensitive to the random ICs.

Moreover, the numerical results for case I show that the interfaces are quasi-steady (not changed too much) in the “outer” region surrounding the rapidly growing valley in the later stage. Physically, this nontrivial property could be described by a local balance between the evaporation and liquid flow out of the pinch region near rupture,

$$\mathcal{E}H^{-1} = \mp[(H^3 + \mathcal{D})H_x]_x - (H^3H_{xxx})_x. \quad (23)$$

Although the vapor thrust effect has not been investigated systematically except for its interaction with the gravity and mass loss, we indicate that the evolution is also sensitive to $\mathcal{D}$ (or E^2D^{-1}), and the “cusp” of the rupture point will be rounded with a reduced $\mathcal{D}$ in addition to incorporating the Marangoni effect. Of particular significance here is the competition between the mechanisms of destabilizing vapor recoil and stabilizing capillarity, which determines the most rapidly amplified disturbance wavelength of the initial instability (discussed in Sec. IV) and the touch-down dynamics of the final stage that will be investigated further.

D. Locally vanishing effect of gravity: Time and length scales

The evaporation of the liquid layer starts with a finite thickness, where the gravity is important. However, as the film evaporates, the local thickness reduces and correspondingly the local effect

FIG. 10. The evolution of the ratio of vapor thrust to gravity terms (dashed line) and the ratio of surface tension to gravity terms (solid line) given by Eq. (24) using the numerical results of case I obtained in Figs. 5(e) and 9(d): (a) $G < 0$, (b) $G > 0$. For clarity, a logarithmic x -scale is used. Inset: the evolution of $H_{\min}$. The effects of the vapor recoil and surface tension become dominant when $H_{\min} \lesssim 0.4$. Also indicated in the insets is the time scale when the local effect of the gravity can be neglected, corresponding to the length scale of 0.4.

of the gravity weakens. Since the maximum evaporation flux and interface curvature are attained near $H_{\min}$, this would be the most likely position for the vapor recoil and capillary effects to dominate over gravity. To make this statement explicit, we consider the numerical results of case I to shed some light on the time and length scales when the gravity can be neglected. According to Eq. (15), the ratios of the vapor recoil and surface tension term to the gravity term, the stabilizing/destabilizing mechanism behind the dynamics for $G > 0/G < 0$, are

$$\frac{\text{vapor recoil term}}{\text{gravity term}} = \frac{|\mathcal{D}|}{H^3}, \quad \frac{\text{surface tension term}}{\text{gravity term}} = \left| \frac{H_{xxx}}{H_x} \right|. \quad (24)$$

These ratios are plotted in Fig. 10 for $H_{\min}$ along with an indication of the length and time scales obtained in the numerical results of Figs. 5(e) and 9(d) for $G < 0$ and $G > 0$. In both cases, it is found that the gravity term is initially important as compared with the vapor recoil and surface tension terms. However, as the film evaporates and interfacial instability evolves, its local thickness decreases and thus the relative importance of the gravity reduces significantly, while the local evaporation flux and interface curvature become progressively larger. It is thus that these two effects of the vapor recoil and surface tension are critical in the final stage of touch-down dynamics.

Specifically, the local effect of the gravity becomes negligible when $H_{\min} \lesssim 0.4$ in both cases. The corresponding time scale is $\sim 0.87t_r$ for $G < 0$, which is earlier than $\sim 0.96t_r$ for $G > 0$ because the droplets feed liquid into surrounding valleys under the stabilizing effect of the positive gravity. Since $H_{\min}$ will become small enough near rupture for the effects of the vapor recoil and surface tension to become dominant and comparable, it is reasonable to neglect the gravity but preserve the surface tension in a local analysis (see Sec. VI). This also justifies our discussion in Fig. 7(b). By changing the control parameters or experimental conditions, future simulations and experiments could reveal the dependence of the length and time scales on $\mathcal{E}$ and $\mathcal{D}$.

VI. FURTHER DISCUSSION ON RUPTURE OF EVAPORATING LAYERS WITH SIMILARITY ANALYSIS

A. From time-dependent simulation to similarity equation

As $t \rightarrow t_r$ and $H_{\min} \rightarrow 0$, the local interface behaviors of case I are reminiscent of the line, point, and ring (for axisymmetric problem) ruptures under the van der Waals forces^{6,23} in view of the dynamic length scales much smaller than the system dimensions. Burelbach *et al.*⁶ found a variation in the exponent q of the length scale, $H_{\min} \sim (t_r - t)^q$, e.g., $1/2 \leq q \leq 1$ corresponding to $1 \geq \mathcal{E} \geq 0$ from a quasi-equilibrium behavior of $-\ln H_{\min}$ versus $-\ln(t_r - t)$ with $\mathcal{D} = 1$. For a van der Waals rupture without evaporation, a characteristic horizontal length scale varying as $H_{\min}^2$ with $q = 1/5$ was found from a time-dependent simulation by Zhang and Lister,²² who showed that the

FIG. 11. Local “similarity” profiles close to the rupture from time-dependent simulations using an adaptive grid scheme with $N_{\max} = 1000$ for $|\mathcal{D}| = 1$. (a) Right-rupture point of the layer subject to RTI with $\varepsilon = 0.05$ and $\lambda = 2\sqrt{2}\pi$. (b) 1-point rupture for $G > 0$ or $G < 0$ with $\varepsilon = 0$ and $\lambda = 4\sqrt{2}\pi$.

effects of the surface tension were not asymptotically negligible, also supported by our discussion in Fig. 7(b). As suggested in Fig. 7(a) for a particular case I of Fig. 5(a), we find $H_{\min} \sim (t_r - t)^{1/2}$ near rupture. Accordingly, Fig. 11(a) plots $H(t_r - t)^{-1/2}$ as a function of x around the right-rupture point of Fig. 5(a), for a linear region indicated by “star” (★) in Fig. 7(a), to illustrate the local similarity behavior. The minimum point for each profile remains approximately in the same position that the local behavior may be described by the “similarity” form $H \propto (t_r - t)^{1/2} F(x)$, where $F(x)$ is a spatial similarity function. The asymmetry of the profiles in Fig. 11(a) results from the different growth rates of the adjacent droplets and numerical instability.

As $H_{\min} \rightarrow 0$, Eq. (15) is singular as H_t goes to infinity. If the surface tension has a minor effect near the rupture point,^{1,6} it can be reduced to an ODE: $H_t \approx -\varepsilon H^{-1}$ in view of the changing of H_{xx} and H_{xxxx} not too fast as compared to the indefinitely increasing H^{-1} , and the local value of H_x not too large and tending to vanish near $H_{\min}$. With $\varepsilon = 1$, one thus could infer $H_{\min} \sim (t_r - t)^{1/2}$ whether $|\mathcal{D}| = 1$ or 0. The simple scaling argument coincides with Ref. 6 even without van der Waals attractions. It is therefore of interest to pose the question about our physical problem: Could similarity solution still occur in the evaporation-driven rupture with a significant vapor recoil in place of the van der Waals forces?

With film thinning, the local gravity effect weakens compared to the surface tension and vapor thrust for the time integration near $H_{\min}$, it is reasonable to neglect the gravity in the local analysis (see Sec. V D), as an analogous argument in Sec. II.E of Ref. 1. Thus Eq. (15) degenerates into

$$H_t + \varepsilon H^{-1} \pm \mathcal{D} H_{xx} + (H^3 H_{xxx})_x = 0, \quad (25)$$

which governs the local touch-down dynamics of the interface. Recalling Sec. V C, Eq. (25) reflects that the evaporation-driven rupture of a viscous layer is controlled by a balance between the vapor thrust, surface tension, and viscous dissipation. It turns out that the requirement of they are equally important near rupture determines the scalings of the horizontal and vertical length scales, $\mathcal{H}$ and $\mathcal{V}$. It is assumed that $F(\eta) = H(x, t)/\mathcal{V}(t)$, where $\eta = (x - x_r)/\mathcal{H}(t)$ is a similarity variable and x_r denotes the rupture position. Then $\mathcal{H}$ and $\mathcal{V}$ can be determined with the appropriate auxiliary conditions when Eq. (25) is transformed into

$$(F^3 F''')' \pm \mathcal{D} \frac{\mathcal{H}^2}{\mathcal{V}^3} F'' - \frac{\mathcal{H}^3 \mathcal{H}'}{\mathcal{V}^3} \eta F' + \frac{\mathcal{H}^4 \mathcal{V}'}{\mathcal{V}^4} F + \varepsilon \frac{\mathcal{H}^4}{\mathcal{V}^5} F^{-1} = 0, \quad (26)$$

where “'” denotes the derivative of single-variable functions. If the similarity ansatz is correct, each fraction in Eq. (26) must be constant. The two unknown scale factors provide two degrees of freedom in specifying the constants. We assume $\mathcal{H}^3 \mathcal{H}' \mathcal{V}^{-3} = c_1$ and $\mathcal{H}^4 \mathcal{V}' \mathcal{V}^{-4} = c_2$ with constants c_1 and c_2 . Because the length scales shrink with time, we need $\mathcal{H}', \mathcal{V}' < 0$, and $c_1, c_2 < 0$. Otherwise, the values of c_1 and c_2 are unimportant in the sense of scaling. In the present context, only the limit of case I with $\varepsilon = 0$ and $|\mathcal{D}| = 1$ is of interest.

As the first choice, let $\mathcal{H}^2 = \mathcal{V}^3$ to make the coefficient of the vapor-recoil term simplest, then set $c_1 = -1/2 = (\mathcal{H}^2 \mathcal{V}')/2$, where the first relationship has been used. We could obtain $\mathcal{H} = (t_r - t)^{1/2}$ and $\mathcal{V} = (t_r - t)^{1/3}$ with the constraint of $\mathcal{H} \rightarrow 0$ as $t \rightarrow t_r$. It also follows that $c_2 = -1/3 < 0$

as required. The length scale $\mathcal{V}$ implies $H_{\min} \sim (t_r - t)^{1/3}$, which is different from the linear law in Ref. 6 for the same evaporation conditions. It should not be of surprise because the singularity of their equation originated from the fractional terms associated with mass loss and van der Waals forces. Figure 11(b) represents $H(t_r - t)^{-1/3}$ around the single rupture point from the time-dependent simulation of Eq. (25) for $\mathcal{E} = 0$ and $|\mathcal{D}| = 1$ with the similar numerical scheme in Ref. 22. The slight asymmetry of the profiles is caused by a numerical singularity near rupture. The convergence of minimum points confirms the deduced scaling of $\mathcal{V}$. Equation (26) thus becomes a fourth-order nonlinear ODE for the self-similar film thickness,

$$(F^3 F''')' + |\mathcal{D}| F'' + \frac{1}{2} \eta F' - \frac{1}{3} F = 0. \quad (27)$$

It requires four boundary conditions: $F'(0) = F'''(0) = 0$ are specified for regularity and $F(0)$ and $F''(0)$ can be determined by shooting for the quasi-steady conditions at infinity.

B. Far-field asymptotics and similarity solutions

The far-field asymptotics of the locally self-similar behavior must satisfy the quasi-steady condition: $(F - 3\eta F'/2) \rightarrow 0$. It forces the similarity solutions to have the structure $F \sim \mathcal{A} \eta^{2/3}$ as $|\eta| \rightarrow \infty$ for asymptotic matching to an insensitive outer solution, where the positive free constant $\mathcal{A}$ provides a global parameter for the comparison of different solutions of Eq. (27). With a perturbation analysis about the asymptotic form,²² we find that the similarity solutions locate in some isolated points in the $(F(0), F''(0))$ parameter space. It is also informative to exploit the alternative far-field behaviors of Eq. (27) in the $(F(0), F''(0))$ plane,

$$\begin{aligned} (F^3 F''')' &\sim \frac{1}{3} (F - \frac{3}{2} \eta F') \quad \text{as } |\eta| \rightarrow \infty, \\ (F^3 F''')' &\sim 0 \quad \text{as } |\eta| \rightarrow \infty. \end{aligned} \quad (28)$$

The first one yields a nontrivial asymptotics $F \sim \mathcal{B} \eta^{4/3}$ with the fixed constant $\mathcal{B} = (27/56)^{1/3}$, and the latter gives $F \sim \mathcal{C} \eta^2$ with the free constant $\mathcal{C}$.

We therefore expect to determine the isolated values of $(F(0), F''(0))$, which induce $\eta^{2/3}$ asymptotics and allow the behavior of Eq. (28) as well. The numerical problem is complicated by the capillary term, which gives rise to oscillatory solutions to Eq. (27) and possibly nonmonotonic far-field behavior with respect to the shooting parameters. First, we search for the $\eta^{2/3}$ solutions with a two-parameter shooting method in which Eq. (27) is integrated using a fourth-order Runge-Kutta scheme from the origin to a certain large $\eta_{\max}$ as the end of the integration interval. The $o(\eta^9)$ Maclaurin series are used to initiate the integration. The parameters are determined to satisfy the following boundary conditions:

$$F - \frac{3}{2} \eta F' = 0, \quad (29a)$$

$$F + \frac{9}{2} \eta^2 F'' = 0, \quad (29b)$$

FIG. 12. Shooting method searching for solutions with different far-field behaviors in the parameter space for $\mathcal{E} = 0$ and $|\mathcal{D}| = 1$. (a) $\eta^{2/3}$ solutions occur at the intersections. The values of $(F(0), F''(0))$ lying on the solid and dashed curves satisfy Eqs. (29a) and (29b) at $\eta_{\max} = 6$. (b) $\eta^{4/3}$ solutions occur at the curves that satisfy $F - 9\eta^2 F''/4 = 0$ at $\eta_{\max} = 40$. Terminating points correspond to parameter values for $\eta^{2/3}$ solutions in subfigure (a).

TABLE II. Parameters for the first seven $\eta^{2/3}$ similarity solutions with $\mathcal{E} = 0$ and $|\mathcal{D}| = 1$.

Solution No.	1	2	3	4	5	6	7
$F(0)$	0.564 709	0.385 308	0.307 045	0.268 729	0.242 702	0.224 105	0.210 448
$F''(0)$	0.305 821	0.119 509	0.108 368	0.089 287	0.082 457	0.074 947	0.070 944
$\mathcal{A}$	0.595 655	0.382 568	0.303 221	0.264 998	0.239 01	0.220 65	0.207 154

which are specified independently to require $F \propto \eta^{2/3}$ at $\eta_{\max}$ and involve two low-order derivatives. Then the scheme can be optimized by plotting the parameter curves for $\eta^{4/3}$ solutions with only one end condition $F - 9\eta^2 F''/4 = 0$ in an extended interval. The $\eta^{4/3}$ curves will reflect their relation to the $\eta^{2/3}$ solutions. Here, only those allowing $F'''(\eta_{\max}) < 0$ give $\eta^{4/3}$ solutions, and those with $F''' > 0$ are removed as they are transitions to the η^2 behavior.

Figure 12 illustrates the results of searching for $\eta^{2/3}$ and $\eta^{4/3}$ solutions with $\mathcal{E} = 0$ and $|\mathcal{D}| = 1$. In Fig. 12(a), the solid and dashed curves satisfy Eqs. (29a) and (29b) at $\eta_{\max} = 6$, respectively, where the intersections correspond to the $\eta^{2/3}$ solutions. As $\eta_{\max}$ increases, the numerical experiment reveals that the structures of the curves vary substantially; however, the intersections are fixed essentially. Moreover, the separation between intersections reduces monotonously as $(F(0), F''(0)) \rightarrow (0, 0)$, implying that Eq. (27) admits a countably infinite number of solutions. The values of $(F(0), F''(0))$ and $\mathcal{A}$ for the first seven $\eta^{2/3}$ similarity solutions are listed in Table II. Noted that both $F(0)$ and $F''(0)$ decrease monotonically with $\mathcal{A}$, instead of an oscillatory fashion for the curvature $F''(0)$ as shown in Ref. 22. In Fig. 12(b) for $\eta^{4/3}$ solutions with $\eta_{\max} = 40$, some curves pass through smoothly, while the others terminate in a spiral around a certain $\eta^{2/3}$ solution on the dynamical landscape. By plotting the $\eta^{4/3}$ spirals with increased $\eta_{\max}$, we have confirmed this observation. Specifically, the latter method is simpler and more robust than locating the roots of Eq. (29) for the $\eta^{2/3}$ solutions due to the less variation of parameter curves with one end condition only.

Figure 13 shows $F(\eta)$ and $F'''(\eta)$ for the first, third, fifth, and seventh similarity solutions of Eq. (27) along with the self-similar results from the simulation of Eq. (25) for $\mathcal{E} = 0$ and $|\mathcal{D}| = 1$, which agree well with the first one $F_1(\eta)$ with $F_1'''(\eta)$ of the least oscillation near the rupture point. Our simulations have shown that the local behavior close to the rupture does not depend on the choice of IC, which suggests that only the $F_1(\eta)$ solution represents the stable, self-similar, 1-point rupture, as observed in time-dependent simulations. The fundamental differences of $F(\eta)$ lie in $F(0)$ and $F'''(\eta)$: as $F(0)$ decreases, $F'''(\eta)$ undergoes an additional half-oscillation before approaching to the $\eta^{2/3}$ far field, which provides a well-defined method for counting of the solutions. There is no further similarity solution for larger $F(0)$ as found in Ref. 22. Thus, the ODE describing the evaporation-driven rupture of viscous films under a balance of the surface tension and significant vapor recoil has a countably infinite number of similarity solutions.

FIG. 13. Profiles of $F(\eta)$ and $F'''(\eta)$ for the first, third, fifth, and seventh similarity solutions. Red dots corresponding to the grid points show the rescaled solutions of the time-dependent simulation for $\mathcal{E} = 0$ and $|\mathcal{D}| = 1$ that closely follow the first solution $F_1(\eta)$.

VII. CONCLUSIONS

We have examined the dynamics and instabilities of the quasi-equilibrium evaporating layers subject to the constant surface tension and negative/positive gravity coupled with different degrees of mass-loss and strong vapor-recoil effects. The results of the simple model uncovered some interesting physical features and dynamics on the rupture by the vapor thrust in evaporating layers, which were affected appreciably by the direction of the gravity, evaporation effects, and initial condition. Special emphasis was laid on the touch-down dynamics. Time-dependent LSA led to the estimates of the effective growth rate, $s_{\text{eff}}(t)$, and suggested that the rupture instability was enhanced with time by vapor recoil. The cutoff and maximum growth wavenumbers were functions of the basic thickness. The numerical simulations of the evolution equation provided the spatio-temporal characterization of a variety of pattern formations including rupture time, capillary ridges, and partially coalesced or separated droplets with sharp dry-out points. For the evaporation-driven rupture with significant vapor recoil and negligible mass loss, a countably infinite number of similarity solutions were found by the far-field asymptotic analysis of a similarity ODE, their horizontal and vertical length scales behaved as $(t_r - t)^{1/2}$ and $(t_r - t)^{1/3}$, respectively.

Finite-time rupture always occurred independent of the direction of the gravity, the degree of mass loss, or the type of initial conditions, and thus must be an intrinsic instability of the evaporating interface under vapor recoil. For $G < 0$, the asymmetric and random perturbations evolved into two partially coalesced droplets in both cases I and II, separated from an adjacent period by a noncentral rupture point, with different height scales by one order of magnitude between case I and case II. With a symmetric sinusoidal IC, it ruptured at two symmetric locations on the substrate in case I, while at the midpoint in case II. For $G > 0$, the random IC could evolve into two partially coalesced droplets in case II only, and capillary ridges emerged around a deepening narrow valley along with a single sharp rupture point in case I. For both $G < 0$ and $G > 0$, t_r reduced roughly by one order of magnitude and the thicknesses of droplets decreased remarkably at rupture as the modified evaporation number, $\mathcal{E}$, was increased by one order from case I. The interface instability of the quasi-equilibrium evaporation was sensitive to (i) the direction of the gravity with $\mathcal{E}$ of $O(10^{-2})$ but not with $\mathcal{E}$ of $O(10^{-1})$ and (ii) $\mathcal{E}$ for both $G > 0$ and $G < 0$.

It was reasonable to neglect the *local* effect of the gravity on a local late-time analysis as long as $|\mathcal{D}|/H^3 \gg 1$ and $|H_{xxx}/H_x| \gg 1$ at $H_{\min}$. Nevertheless, there were no uniform length and time scales to neglect the *overall* effect of the gravity because the gravity played a stabilizing($G > 0$)/destabilizing($G < 0$) role in the dynamics up to the rupture on the horizontal scale λ and the time scale t_r . The stabilization of the capillarity was dominant in the early evolution and then overcome by vapor recoil; however, its stabilizing action was still significant in the final stage. The competition between the vapor recoil and surface tension governed the local dynamics close to the rupture. Only a single-point rupture was stable with respect to nonsymmetric disturbances. The self-similar results from the pertinent PDE accorded with the first similarity solution of $F_1(\eta)$ corresponding to the least oscillatory $F_1'''(\eta)$ near the rupture point, which represented the stable 1-point rupture. Finally, the thinness of the layer and the shortness of the late-time scale have caused considerable difficulties in obtaining the reliable results on the interface rupture, for which the relevant experimental data are still limited in the literature, especially for evaporating layers. To test the cases of interest, we suggested the typical working fluids together with the experimental conditions. The experimental study can be conducted to compare with these theoretical predictions.

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APPENDIX A: LEADING-ORDER SOLUTION OF RESCALED GOVERNING SYSTEM

Here we outline the $O(1)$ solution that leads to Eq. (12). First, we find the evaporative flux and temperature profile by integrating Eq. (10) with $\theta_0 = 1$ and Eqs. (11b) and (11d),

$$J_0(\xi, \tau) = (K + H)^{-1}, \quad \theta_0(\xi, \zeta, \tau) = 1 - (K + H)^{-1}\zeta. \quad (\text{A1})$$

Solving the ζ -component of Eq. (8) with (11c) and (A1), one obtains the pressure profile

$$P_0 = \frac{3}{2} \bar{E}^2 \bar{D}^{-1} J_0^2 - 3 \bar{S} H_{\xi\xi} + 3 \bar{G} (H - \zeta), \quad (\text{A2})$$

which consists of vapor recoil and Laplace pressure on the surface and hydrostatic pressure in the bulk. Substituting it into the ξ -component in Eq. (8) and integrating in ζ yields $U_0 = \Lambda(\frac{1}{2}\zeta^2 - H\zeta)$ with $\Lambda = -3\bar{E}^2\bar{D}^{-1}H_\xi(K+H)^{-1}J_0^2 - 3\bar{S}H_{\xi\xi\xi} + 3\bar{G}H_\xi$. Finally, one plugs J_0 and U_0 in Eq. (9) and makes the substitutions $(\bar{D}, \bar{E}, \bar{G}, \bar{S}) \rightarrow (D, E, G, S)$ and $(\xi, \zeta, \tau) \rightarrow (X, Z, T)$ to obtain Eq. (12) by setting $K = 0$ and assuming ∂_X and ∂_T are comparable.

APPENDIX B: BASIC STATE

The basic state is a thinning, x -independent, and motionless layer, whose dependent variables are denoted by overbar. The z -components of the non-dimensional Navier–Stokes and energy equations reduce to

$$\partial_Z \bar{P}_d = 0, \quad \text{Pr } \bar{\Theta}_T = \bar{\Theta}_{ZZ}, \quad (\text{B1})$$

where $\bar{P}_d = \bar{P} + \Phi$ is the dimensionless dynamic pressure and $\Phi = h_0^2 \rho \mu^{-2} \phi$ defines the dimensionless potential function with $\phi = \rho g z$. At $Z = 0$, the temperature boundary condition is $\bar{\Theta}(0, T) = 1$. At $Z = \bar{H}$, the non-dimensional kinematic condition, constitutive equation, and energy and normal-stress balance reduce to

$$E\bar{J} = -\bar{H}_T, \quad K\bar{J} = \bar{\Theta}_1, \quad \bar{J} = -\bar{\Theta}_Z, \quad \frac{3}{2}E^2\bar{D}^{-1}\bar{J}^2 - \bar{P} = 0. \quad (\text{B2})$$

The x -components of Navier–Stokes and energy equations, and shear-stress balance are not used in the basic-state problem.

To preserve the mass-loss effects in the kinematic condition at leading order, we rescale T as $T' = ET$. The dependent variables then are expanded as power series in small E ,

$$(\bar{J}, \bar{\Theta}, \bar{P}_d) = (\bar{J}_0, \bar{\Theta}_0, E^{-1}\bar{P}_{d0}) + E(\bar{J}_1, \bar{\Theta}_1, E^{-1}\bar{P}_{d1}) + \dots \quad (\text{B3})$$

Here, we assume that $\bar{J}, \bar{\Theta} = O(1)$, and $\bar{P}_d = O(E^{-1})$ as $E \rightarrow 0$, and $\bar{H}(T')$ is regarded as an undetermined $O(1)$ function. Further, assuming $D = \bar{D}E^3$ the vapor thrust is retained in the normal-stress condition to the leading order, where $\bar{D} = O(1)$. With the rescaled variable and the expansions (B3), the basic-state problem takes the leading-order form

$$\partial_Z \bar{P}_{d0} = 0, \quad (\text{B4})$$

$$\partial_Z^2 \bar{\Theta}_0 = 0. \quad (\text{B5})$$

The boundary conditions become $\bar{\Theta}_0(0, T') = 1$ at $Z = 0$, and

$$\bar{J}_0(T') = -\bar{H}_{T'}, \quad (\text{B6})$$

$$K\bar{J}_0(T') = \bar{\Theta}_0(\bar{H}, T'), \quad (\text{B7})$$

$$\bar{J}_0(T') = -\partial_Z \bar{\Theta}_0(\bar{H}, T'), \quad (\text{B8})$$

$$E^{-1}[\frac{3}{2}\bar{D}^{-1}\bar{J}_0^2(T') - \bar{P}_{d0}(\bar{H}, T')] = -\Phi(\bar{H}), \quad (\text{B9})$$

at $Z = \bar{H}$, supplemented with an initial condition $\bar{H}(T' = 0) = 1$.

Then the basic-state solution can be straightforwardly obtained. Integration of Eq. (B5) with (B8) gives $\bar{J}_0 = -f(T')$, where f is an unspecified function. Integrating Eq. (B5) again and using $\bar{\Theta}_0(0, T') = 1$ yield $\bar{\Theta}_0(Z, T') = f(T')Z + 1$. Substituting them in Eq. (B7) and solving for f , one obtains

$$\bar{J} \approx \bar{J}_0 = (K + \bar{H})^{-1}, \quad \bar{\Theta} \approx \bar{\Theta}_0 = 1 - (K + \bar{H})^{-1}Z. \quad (\text{B10})$$

We substitute $\bar{J}_0$ in Eq. (B6) and integrate to find $\bar{H} = -K + [(1 + K)^2 - 2ET]^{1/2}$ when using the dimensionless time T . Solving Eq. (B4), subject to condition (B9), one finds

$$\bar{P}_d \approx E^{-1}\bar{P}_{d0} = \frac{3}{2} \frac{E^2}{D(K+\bar{H})^2} + 3G\bar{H}. \quad (\text{B11})$$

When the gravity is negligible, our basic-state solution reduces to that of Burelbach *et al.*⁶ If we use the rescaled time $\hat{t}$ in Eq. (14) and the modified parameter $\mathcal{E}$, we have

$$\bar{H} = -K + [(1 + K)^2 - 2\mathcal{E}\hat{t}]^{1/2}. \quad (\text{B12})$$

By setting $\bar{H} = 0$, the evaporating time is found to be $\hat{t}_e = (1 + 2K)/(2\mathcal{E})$. According to Eq. (B12), for $K = 0$ the thickness, $\bar{H} = (1 - 2\mathcal{E}\hat{t})^{1/2}$, decreases monotonously with velocity $\bar{H}_t \rightarrow -\infty$ and $\bar{J}$, $\bar{P}_d \rightarrow \infty$ as $\hat{t} \rightarrow \hat{t}_e = 1/(2\mathcal{E})$. These imply underlying singularities in the leading-order solution near $\hat{t}_e$ where the higher-order terms in (B3) play a role comparable with that of leading-order ones.

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