Effect of gravity-induced shape change on the diffusion-limited evaporation of thin sessile and pendant droplets

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A comprehensive study of the effect of gravity-induced shape change on the diffusion-limited evaporation of thin sessile and pendant droplets on a horizontal substrate is performed. Specifically, theoretical predictions for the evolution, and hence the lifetime, of sessile and pendant droplets evaporating in four modes of evaporation, namely, the constant contact radius (CR), the constant contact angle (CA), the stick-slide (SS), and the stick-jump (SJ) modes, are obtained. In particular, it is shown that gravity-induced shape change can cause quantitative differences in the evolution of sessile and pendant droplets compared to that of a droplet in the absence of (or in the neglect of) the effect of gravity (a "zero-gravity droplet"). For example, whereas sessile and pendant droplets evaporating in the CR mode evolve in qualitatively the same manner as a zero-gravity droplet, the evolution of droplets evaporating in the CA mode is more complicated. Specifically, while a zero-gravity droplet evaporating in the CA mode evolves according to the well-known d^2 and 2/3 laws, an initially large sessile droplet evolves according to qualitatively different d and 1/2 laws, and an initially large pendant droplet evolves with the contact radius and the volume (but not, of course, the contact angle) behaving as if the droplet was evaporating in the CR mode. It is also found, perhaps somewhat unexpectedly, that the maximum height of a sessile droplet evaporating in the CA mode is a nonmonotonic function of time when the initial volume of the droplet is sufficiently large. Furthermore, it is found that for all four modes of evaporation a sessile droplet always evaporates faster, and hence has a shorter lifetime, than a zero-gravity droplet with the same initial volume, which in turn always evaporates faster, and hence has a shorter lifetime, than a pendant droplet with the same initial volume. It is also shown that for all four modes of evaporation the lifetime of a droplet is a monotonically increasing function of the initial volume of the droplet, that the lifetime of a droplet evaporating in the CA mode is always longer than that of the same droplet evaporating in the CR mode, and that the lifetimes of droplets evaporating in the SS and SJ modes both always lie between the lifetimes of the same droplet evaporating in the extreme modes.

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I. INTRODUCTION

The evaporation of sessile droplets has been the subject of extensive experimental, numerical, and theoretical investigations in recent years (see, for example, the review articles and books by Cazabat and Guéna [1], Routh [2], Kovalchuk *et al.* [3], Larson [4], Brutin [5], Lohse and Zhang [6], Brutin and Starov [7], Giorgiutti-Dauphiné and Pauchard [8], Zang *et al.* [9], Brutin and Sefiane [10], Gelderblom *et al.* [11], Erbil and McHale [12], Wilson and D'Ambrosio [13], and the many references therein) due to its relevance in a wide range of physical, biological, and industrial processes, such as in agricultural spraying (see, for example, Tredenick *et al.* [14]), chemical and biological assays (see, for example, Garcia-Cordero and Fan [15]), and inkjet printing (see, for example, Kuang *et al.* [16]). Particular attention has been paid to the evolution and lifetime of an evaporating droplet (see, for example, Picknett and Bexon [17], Birdi *et al.* [18], Hu

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and Larson [19], McHale *et al.* [20], Nguyen and Nguyen [21], Stauber *et al.* [22,23], and Wilson and Duffy [24]), and to the deposition onto the substrate from an evaporating particle-laden droplet (see, for example, Deegan *et al.* [25,26], and the additional review articles by Sefiane [27], Zhong *et al.* [28], Mampallil and Eral [29], Parsa *et al.* [30], and Yang *et al.* [31]).

The evolution, and hence the lifetime, of an evaporating droplet depends on the manner (i.e., the mode) in which it evaporates, which, in turn, depends on the physical properties of the system, including the surface structure and roughness, chemical heterogeneity, permeability, and porosity of the substrate. In their pioneering work, Picknett and Bexon [17] identified two so-called extreme modes of evaporation for a droplet, namely, the constant contact radius (CR) mode, in which the contact line remains pinned while the contact angle decreases, and the constant contact angle (CA) mode, in which the contact angle is constant while the contact radius decreases, both of which have also been reported by many subsequent authors (see, for example, Erbil *et al.* [32], Panwar et al. [33], Sobac and Brutin [34], Talbot et al. [35], Gleason and Putnam [36], Gao et al. [37], and Armstrong et al. [38,39]). In practice, droplets often evaporate in a so-called mixed mode, in which both the contact radius and the contact angle vary in time. Perhaps the most frequently reported mixed mode is the stick-slide (SS) mode (see, for example, Bourgès-Monnier and Shanahan [40], Erbil et al. [41], Semenov et al. [42], Nguyen et al. [21,43,44], Dash and Garimella [45], Stauber et al. [22,23], and Kadhim et al. [46]), in which the droplet initially evaporates in a CR (i.e., a "stick") phase until a critical receding angle is reached, after which it evaporates in a CA (i.e., a "slide") phase. Another commonly reported mixed mode is the stick-jump (SJ) mode (see, for example, Adachi et al. [47], Shanahan [48], Bodiguel et al. [49], Orejon et al. [50], Askounis et al. [51-53], and Dietrich *et al.* [54]), in which the droplet evaporates in a series of CR phases separated by short "jump" phases in which the contact line rapidly recedes and the contact angle rapidly increases. Of course, an infinite variety of other mixed modes can occur, including modes in which the contact radius and the contact angle change simultaneously, but the CR, CA, SS, and SJ modes capture a selection of the most frequently observed behaviors.

Many of the previous studies of evaporating droplets either ignore or neglect gravity. For example, the basic diffusionlimited model for the evaporation of a small droplet (see, for example, Murisic and Kondic [55] and Wilson and D'Ambrosio [13]) does not include any gravitational effects. However, there are a number of ways in which gravity can play a role, including the effect of buoyancy in the atmosphere (see, for example, Shahidzadeh-Bonn et al. [56], Kelly-Zion et al. [57], Carle et al. [58], Carrier et al. [59], Moore et al. [60], Dollet and Boulogne [61], and Kadhim *et al.* [46]), the effect of buoyancy within binary droplets (see, for example, Prahdan and Panigrahi [62], Edwards et al. [63], and Li et al. [64]), and the effect of gravity on the deposition onto the substrate from a particle-laden droplet (see, for example, Sommer [65], Sandu and Fleaca [66], Hampton et al. [67], Devlin et al. [68], and Moore and Wray [69]). There has also been some work on the effect of gravity-induced shape change

on the evolution, and hence the lifetime, of an evaporating droplet. For an (at least initially) pinned droplet on an inclined substrate (see, for example, Du and Deegan [70], Kim et al. [71], Timm et al. [72], Dhar et al. [73], and Cai et al. [74]), it has been observed experimentally that inclining the substrate can asymmetrically deform the droplet in a manner that promotes depinning of the upper portion of the contact line, which decreases the total evaporative flux and hence increases the lifetime of the droplet. On the other hand, for a sessile droplet on a horizontal substrate, Kadhim et al. [46] made experimental observations of sessile droplets of water with initial contact angles of approximately 49° and 120° evaporating in the SS mode. They found that a theoretical model based on the diffusion-limited evaporation of a spherical-cap droplet overpredicted the experimentally measured lifetimes of large droplets (with volumes of 30 µl) more significantly than those of small droplets (with volumes of less than 8 µl), suggesting that the effect of gravity-induced shape change is to decrease the lifetime of a sessile droplet. More recently, Tonini and Cossali [75,76] used the diffusion-limited model to calculate the total evaporative flux from a sessile droplet relative to that of the total evaporative flux from a droplet in the absence of gravity, i.e., a spherical-cap droplet, with the same volume and contact angle for increasing values of the Bond number. They found that the effect of increasing the value of the Bond number (i.e., increasing the relative strength of gravity) is to increase the evaporative flux for droplets with contact angles less than $\pi/2$ (i.e., in hydrophilic situations) but to first decrease and then increase it for droplets with contact angles greater than $\pi/2$ (i.e., in hydrophobic situations). Subsequently, Tonini and Cossali [77] extended this investigation to pendant droplets and found that for droplets with contact angles less than $\pi/2$, the minimum value of the total evaporative flux is obtained for pendant droplets, but for droplets with contact angles greater than $\pi/2$, the minimum value of the total evaporative flux is obtained for sessile droplets. In addition, for a fixed value of the Bond number, the total evaporative flux relative to that from a sphericalcap droplet decreases as the contact angle increases for sessile droplets, but increases with the contact angle for pendant droplets. They did not, however, use any of these results to calculate the effect of gravity-induced shape change on the evolution of the droplet. Indeed, no one has yet performed a comprehensive study of the effect of gravity-induced shape change on the evaporation of sessile and pendant droplets on a horizontal substrate. This is the aim of the present work. Specifically, in Sec. II we formulate a mathematical model describing the diffusion-limited evaporation of a thin droplet of arbitrary volume. In Sec. III we summarize the key results about the shape of sessile and pendant droplets. In Sec. IV we obtain theoretical predictions for the evolution, and hence the lifetime, of sessile and pendant droplets evaporating in the two extreme modes of evaporation. In Secs. V and VI we extend the investigation to two mixed modes of evaporation, specifically to the SS and SJ modes, respectively. Finally, in Sec. VII we summarize our findings and indicate possible directions for future work. In the Appendix we validate the present approach by comparing the asymptotic solution for the evaporative flux from a thin droplet with the numerical solution for a nonthin droplet.



FIG. 1. Sketch of an evaporating thin axisymmetric (a) sessile and (b) pendant droplet with contact radius $\hat{R}(\hat{t})$, contact angle $\hat{\theta}(\hat{t})$, and free-surface profile $\hat{z} = \hat{h}(\hat{r}, \hat{t})$ on a horizontal substrate in the presence of gravity \hat{g} . The arrows indicate the local evaporative flux $\hat{J}(\hat{r}, \hat{t})$.

II. PROBLEM FORMULATION

Consider a thin axisymmetric sessile or pendant droplet on a horizontal substrate undergoing quasistatic diffusion-limited evaporation in the presence of gravity $\hat{\mathbf{g}}$. We refer the description to cylindrical polar coordinates $(\hat{r}, \hat{\varphi}, \hat{z})$, with $O\hat{z}$ along the axis of the droplet, perpendicular to the substrate at $\hat{z} = 0$, pointing upward or downward for a sessile or a pendant droplet, respectively, as sketched in Fig. 1. The contact radius, contact angle, volume, and free-surface profile of the droplet are denoted by $\hat{R} = \hat{R}(\hat{t}), \hat{\theta} = \hat{\theta}(\hat{t}), \hat{V} = \hat{V}(\hat{t}), \text{ and } \hat{h} = \hat{h}(\hat{r}, \hat{t}),$ respectively, where \hat{t} denotes time. The initial values of $\hat{R}, \hat{\theta}$, and \hat{V} at $\hat{t} = 0$ are denoted by $\hat{R}_0, \hat{\theta}_0$, and \hat{V}_0 , respectively, and the lifetime of the droplet (i.e., the time it takes for the droplet to entirely evaporate) when it evaporates in the CR, CA, SS, and SJ modes is denoted by $\hat{t}_{CR}, \hat{t}_{CA}, \hat{t}_{SS}$, and \hat{t}_{SJ} , respectively.

A. The hydrostatic problem

We consider situations in which the droplet is thin and surface tension is sufficiently strong that the free surface of the droplet evolves quasistatically, but, unlike in most of the previous studies of evaporating droplets, in which the volume of the droplet is sufficiently large that gravity has an effect on its shape. More specifically, we consider situations in which both the initial contact angle of the droplet, $\hat{\theta}_0 \ll 1$, and the capillary number, $Ca = \hat{\mu}\hat{U}/(\hat{\theta}_0^3\hat{\sigma}) \ll 1$, are small, but the characteristic radial length scale of the droplet, denoted by \hat{L} , is the capillary length, $\hat{\ell} = (\hat{\sigma}/(\hat{\rho}\hat{g}))^{1/2}$, where $\hat{\sigma}$ and $\hat{\rho}$ are the constant surface tension and density of the fluid, and $\hat{g} = |\hat{g}|$ is the magnitude of acceleration due to gravity, i.e., in which the Bond number, Bo = $(\hat{L}/\hat{\ell})^2 = 1$, is unity. Although formally only valid in the asymptotic limit of small contact angle, in practice, theoretical predictions for thin evaporating droplets have been found to be in good agreement with experimental results for droplets with contact angles up to around $\pi/2$ (see, for example, Dunn et al. [78] and Wray et al. [79,80]).

The pressure in the droplet, denoted by $\hat{p} = \hat{p}(\hat{r}, \hat{z}, \hat{t})$, satisfies

$$\hat{\boldsymbol{\nabla}}\hat{\boldsymbol{p}} = \mp \hat{\rho}\hat{\boldsymbol{g}}\,\mathbf{e}_{z},\tag{1}$$

where \mathbf{e}_z denotes the unit vector in the *z* direction and the upper and lower signs correspond to a sessile and a pendant droplet, respectively, subject to the Young-Laplace equation at the free surface of the droplet, which for a thin droplet takes

the form

$$\hat{p} - \hat{p}_{a} = -\frac{\hat{\sigma}}{\hat{r}} \frac{\partial}{\partial \hat{r}} \left(\hat{r} \frac{\partial \hat{h}}{\partial \hat{r}} \right) \quad \text{at} \quad \hat{z} = \hat{h}.$$
 (2)

Solving Eq. (1) subject to Eq. (2) yields the solution for the pressure \hat{p} ,

$$\hat{p} = \hat{p}_{a} - \frac{\hat{\sigma}}{\hat{r}} \frac{\partial}{\partial \hat{r}} \left(\hat{r} \frac{\partial \hat{h}}{\partial \hat{r}} \right) \pm \hat{\rho} \hat{g} (\hat{h} - \hat{z}), \qquad (3)$$

and hence the governing equation for the free-surface profile \hat{h} ,

$$\frac{\partial}{\partial \hat{r}} \left[\frac{\hat{\sigma}}{\hat{r}} \frac{\partial}{\partial \hat{r}} \left(\hat{r} \frac{\partial \hat{h}}{\partial \hat{r}} \right) \mp \hat{\rho} \hat{g} \hat{h} \right] = 0.$$
 (4)

B. The evaporative problem

According to the basic diffusion-limited model for the evaporation of a thin droplet (see, for example, Murisic and Kondic [55] and Wilson and D'Ambrosio [13]), the quasistatic concentration of vapor in the atmosphere, denoted by $\hat{c} = \hat{c}(\hat{r}, \hat{z}, \hat{t})$, satisfies Laplace's equation,

$$\hat{\nabla}^2 \hat{c} = 0 \quad \text{in} \quad \hat{z} > 0, \tag{5}$$

subject to the usual boundary and far-field conditions

$$\hat{c} = \hat{c}_{\text{sat}}$$
 on $\hat{z} = 0$ for $0 \leq \hat{r} \leq \hat{R}$, (6)

$$\frac{\partial \hat{c}}{\partial \hat{z}} = 0 \quad \text{on} \quad \hat{z} = 0 \quad \text{for} \quad \hat{r} > \hat{R}, \tag{7}$$

$$\hat{c} \to \hat{c}_{\infty} \quad \text{as} \quad \hat{r}^2 + \hat{z}^2 \to \infty,$$
 (8)

where \hat{c}_{sat} is the constant saturation concentration and $\hat{c}_{\infty} = H\hat{c}_{sat}$ is the constant ambient concentration, where H ($0 \leq H \leq 1$) is the relative humidity of the atmosphere. Larson [4] gives a detailed discussion of the parameter regimes in which the diffusion-limited model is expected to apply and, in particular, discusses the competition between diffusive and convective mass transfer effects in the atmosphere. Note that the basic diffusion-limited model used here is not, as is sometimes erroneously claimed, isothermal. However, within this model the thermal problems for the temperature distributions in the droplet, the substrate, and the atmosphere are decoupled from the evaporative problem, and so the thermal properties of

the system have no effect on the evolution or lifetime of the droplet. The basic diffusion-limited model has been extended by various authors to include a variety of additional physical effects, including coupling the thermal and evaporative problems by accounting for the temperature dependence of the saturation concentration (see, for example, Dunn *et al.* [81], Sefiane *et al.* [82], Ait Saada *et al.* [83], and Schofield *et al.* [84]), but, for simplicity and clarity, we restrict our attention to the basic model in the present work.

The local evaporative mass flux from the free surface of the droplet, denoted by $\hat{J} = \hat{J}(\hat{r}, \hat{t})$, is given by

$$\hat{J} = -\hat{D}\frac{\partial\hat{c}}{\partial\hat{z}}$$
 on $\hat{z} = 0$ for $0 \leq \hat{r} \leq \hat{R}$, (9)

where \hat{D} is the constant diffusivity of vapor in the atmosphere. Note that, because the droplet is thin, both Eq. (6) and Eq. (9) are evaluated on $\hat{z} = 0$ rather than on $\hat{z} = \hat{h}$.

Integrating \hat{J} over the free surface of the droplet gives the total evaporative mass flux from the droplet, denoted by $\hat{F} = \hat{F}(\hat{t})$, namely,

$$\hat{F} = 2\pi \int_0^{\hat{R}} \hat{J}(\hat{r}, \hat{t}) \, \hat{r} \, \mathrm{d}\hat{r}, \tag{10}$$

and the droplet evolves according to the global massconservation condition,

$$\hat{\rho}\frac{\mathrm{d}\hat{V}}{\mathrm{d}\hat{t}} = -\hat{F}.$$
(11)

C. The nondimensional problem

We nondimensionalize and scale the variables appropriately for a thin droplet (see, for example, Wilson and Duffy [24]) according to

$$\hat{r} = \hat{\ell}r, \quad \hat{z} = \hat{\theta}_0 \hat{\ell}z, \quad \hat{R} = \hat{\ell}R, \quad \hat{\theta} = \hat{\theta}_0 \theta, \quad \hat{h} = \hat{\theta}_0 \hat{\ell}h,$$

$$\hat{V} = \hat{\theta}_0 \hat{\ell}^3 V, \quad \hat{p} - \hat{p}_a = \frac{\hat{\sigma}\hat{\theta}_0}{\hat{\ell}}p, \quad \hat{t} = \frac{\hat{\rho}\hat{\theta}_0 \hat{\ell}^2}{\hat{D}(\hat{c}_{\text{sat}} - \hat{c}_{\infty})}t,$$
(12)

for the droplet and

$$\hat{r} = \hat{\ell}r, \quad \hat{z} = \hat{\ell}\tilde{z}, \quad \hat{c} = \hat{c}_{\infty} + (\hat{c}_{\text{sat}} - \hat{c}_{\infty})c,$$

$$\hat{J} = \frac{\hat{D}(\hat{c}_{\text{sat}} - \hat{c}_{\infty})}{\hat{\ell}}J, \quad \hat{F} = \hat{D}(\hat{c}_{\text{sat}} - \hat{c}_{\infty})\hat{\ell}F,$$
(13)

for the atmosphere.

Using Eqs. (12) the governing equation for the nondimensional free-surface profile h given by Eq. (4) becomes

$$\frac{\partial}{\partial r} \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial h}{\partial r} \right) \mp h \right] = 0, \qquad (14)$$

subject to the contact-line conditions

$$h = 0, \quad \frac{\partial h}{\partial r} = -\theta \quad \text{at} \quad r = R,$$
 (15)

and the regularity condition

$$\frac{\partial h}{\partial r} = 0 \quad \text{at} \quad r = 0.$$
 (16)

The nondimensional volume of the droplet, V = V(t), is given by

$$V = 2\pi \int_0^R h(r, t) r \, \mathrm{d}r.$$
 (17)

Using Eqs. (13), the problem for the nondimensional concentration of vapor in the atmosphere, c = c(r, z, t), given by Eqs. (5)–(8) with Eqs. (9)–(11), becomes that of solving Laplace's equation $\nabla^2 c = 0$ in the half-space $\tilde{z} > 0$ subject to c = 1 on $\tilde{z} = 0$ for $0 \le r \le R$, $\partial c/\partial \tilde{z} = 0$ on $\tilde{z} = 0$ for r > R, and $c \to 0$ as $r^2 + \tilde{z}^2 \to \infty$. The exact solution of this problem is well known, and leads to the familiar expression for *J*, namely,

$$J = \frac{2}{\pi (R^2 - r^2)^{1/2}},$$
(18)

which is singular (but integrable) at the contact line of the droplet at r = R (see, for example, Wilson and Duffy [24]). Hence, the corresponding expression for *F* is

$$F = 4R. \tag{19}$$

Note that, the expressions for J and hence F given by Eqs. (18) and (19), but not the expressions for h and hence V given by Eqs. (14) and (17), coincide exactly with the corresponding expressions for a small droplet in which gravity plays no role (see, for example, Wilson and Duffy [24]), and that, in general, c and, hence, J and F depend on t parametrically via their dependence on R. Furthermore, also note that, because the droplet is thin, it is only its contact radius R, and not any other aspect of its shape h, that appears in the evaporative problem, and hence in the expression for F given in Eq. (19). However, as we show in Secs. IV–VI, gravity-induced shape change can cause quantitative differences in the evolution of sessile and pendant droplets compared to that of a zero-gravity droplet.

Substituting Eq. (19) into the nondimensional version of the global mass-conservation condition (11) yields the governing equation describing the evolution of the droplet,

$$\frac{\mathrm{d}V}{\mathrm{d}t} = -4R.\tag{20}$$

In general, Eq. (20) relates the rates of change of R and θ , but is not by itself sufficient to determine their evolutions. To do this it is necessary to provide additional information about R and/or θ by, for example, specifying the mode in which the droplet is evaporating. This is what we will do in Secs. IV–VI. However, before doing this we summarize the key results about the shape of sessile and pendant droplets in Sec. III.

III. THE SHAPE OF THE DROPLET

A. A sessile droplet

For a sessile droplet, solving Eq. (14) subject to Eqs. (15) and (16) yields the free-surface profile h,

$$h = \frac{\theta[I_0(R) - I_0(r)]}{I_1(R)},$$
(21)

where I_n denotes a modified Bessel function of the first kind of order *n*. In particular, the height at the middle of the droplet



FIG. 2. Plots of the scaled free-surface profile h/θ of a sessile droplet as a function of r for (a) $V/\theta = 10, 20, ..., 100$ and (b) $V/\theta = 100, 200, ..., 1000$. The arrows indicate the direction of increasing V/θ .

(which, as we will see, is also the maximum height of the droplet), denoted by $h_m = h_m(t) = h(0, t)$, is given by

$$h_{\rm m} = \frac{\theta[I_0(R) - 1]}{I_1(R)},\tag{22}$$

and, using Eq. (17), the volume of the droplet V is given by

$$V = \frac{\pi \theta R^2 I_2(R)}{I_1(R)}.$$
(23)

The initial values of R, θ , $h_{\rm m}$, and V are given by

$$R = R_0, \quad \theta = 1, \quad h_{\rm m} = h_{\rm m0} = \frac{I_0(R_0) - 1}{I_1(R_0)},$$
$$V = V_0 = \frac{\pi R_0^2 I_2(R_0)}{I_1(R_0)}.$$
(24)

In principle, any one of the three initial values R_0 , h_{m0} , and V_0 may be prescribed, with the other two determined from Eqs. (24), but in the present work we choose to prescribe the initial volume V_0 , which is typically the easiest to control in a physical experiment.

Figure 2 shows plots of the scaled free-surface profile of a sessile droplet h/θ given by Eq. (21) for a range of values of the scaled volume V/θ given by Eq. (23). In particular, Fig. 2 shows that, as expected, the droplet widens and flattens as its volume increases. Figure 3 shows plots of the scaled maximum height at the middle of the droplet $h_{\rm m}/\theta$ given by Eq. (22) and the contact radius R given implicitly by Eq. (23) as functions of the scaled volume V/θ . In particular, Fig. 3(a) shows, perhaps somewhat unexpectedly, that $h_{\rm m}/\theta$ is nonmonotonic in V/θ , increasing from zero at $V/\theta = 0$ to a maximum value $h_{\rm m}/\theta = h_{\rm m,max}/\theta \simeq 1.081$ at $V/\theta \simeq 73.175$, corresponding to $R \simeq 5.586$, and then decreasing to unity in the limit $V/\theta \to \infty$. However, this nonmonotonic behavior of $h_{\rm m}/\theta$ as a function of V/θ is in agreement with the numerical results of Padday [85] for a nonthin sessile droplet, and has also been found by Aussillous and Quéré [86] for liquid marbles (i.e., perfectly nonwetting droplets) and is mentioned by Finn (see Sec. 3.5 of [87]). On the other hand, Fig. 3(b) shows that R is monotonic in V/θ , increasing from zero at $V/\theta = 0$ to infinity in the limit $V/\theta \to \infty$.

In the limit of small contact radius, $R \rightarrow 0^+$, corresponding to the limit of small scaled volume, $V/\theta \rightarrow 0^+$, the droplet becomes narrow and shallow according to

$$\frac{h}{\theta} = \frac{R^2 - r^2}{2R} - \frac{(R^2 - r^2)^2}{32R} + O\left(\frac{(R^2 - r^2)^3}{R}\right) \to 0^+,$$
(25)

$$\frac{h_{\rm m}}{\theta} = \frac{R}{2} - \frac{R^3}{32} + O(R^5) \to 0^+, \tag{26}$$

$$\frac{V}{\theta} = \frac{\pi R^3}{4} - \frac{\pi R^5}{96} + O(R^7) \to 0^+.$$
 (27)

In particular, at leading order in this limit, Eqs. (25)–(27) reduce to the familiar "paraboloidal cap" solution for a small droplet, namely,

$$\frac{h}{\theta} = \frac{R^2 - r^2}{2R}, \quad \frac{h_{\rm m}}{\theta} = \frac{R}{2}, \quad \frac{V}{\theta} = \frac{\pi R^3}{4}.$$
 (28)

Note that this is, of course, also the solution for a droplet of *arbitrary* volume in the absence of (or in the neglect of) the effect of gravity, and so, in order to avoid any confusion which might be caused by subsequently discussing a small droplet of arbitrary volume, henceforth we refer to this solution as a "zero-gravity droplet."

On the other hand, in the limit of large contact radius, $R \to \infty$, corresponding to the limit of large scaled volume, $V/\theta \to \infty$, surface-tension effects become negligible away from the contact line, and the droplet becomes wide and flat away from the contact line according to

$$\frac{h}{\theta} \sim 1 - \sqrt{\frac{R}{r}} e^{-(R-r)}, \qquad (29)$$

$$\frac{h_{\rm m}}{\theta} = 1 + \frac{1}{2R} + O\left(\frac{1}{R^2}\right) \to 1^+,$$
 (30)

$$\frac{V}{\theta} = \pi R^2 - \frac{3\pi R}{2} + O(1) \to \infty.$$
 (31)

In particular, at leading order in this limit, Eqs. (29)–(31) reduce to the familiar "gravity pancake" or "puddle" solution



FIG. 3. Plots of (a) the scaled maximum height at the middle of a sessile droplet, h_m/θ , and (b) the contact radius *R* of the droplet as functions of the scaled volume V/θ . The dot and the dashed line in (a) correspond to the maximum value $h_m/\theta = h_{m,max}/\theta \simeq 1.081$ at $V/\theta \simeq 73.175$, and the limiting value $h_m/\theta = 1$ as $V/\theta \to \infty$, respectively.

for a large sessile droplet, namely,

$$\frac{h}{\theta} \equiv 1, \quad \frac{h_{\rm m}}{\theta} = 1, \quad \frac{V}{\theta} = \pi R^2.$$
 (32)

B. A pendant droplet

For a pendant droplet, solving Eq. (14) subject to Eqs. (15) and (16) yields

$$h = \frac{\theta[J_0(r) - J_0(R)]}{J_1(R)},$$
(33)

where J_n denotes a Bessel function of the first kind of order n, and hence

$$h_{\rm m} = \frac{\theta [1 - J_0(R)]}{J_1(R)} \tag{34}$$

and

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$$V = \frac{\pi \theta R^2 J_2(R)}{J_1(R)}.$$
(35)

Unlike for a sessile droplet, for a pendant droplet there are infinitely many branches of solutions for *h*; however, only the first branch corresponds to physically relevant solutions satisfying $h \ge 0$ for $0 \le r \le R$. The initial values of R, θ , $h_{\rm m}$, and *V* are given by

$$R = R_0, \quad \theta = 1, \quad h_{\rm m} = h_{\rm m0} = \frac{1 - J_0(R_0)}{J_1(R_0)},$$
$$V = V_0 = \frac{\pi R_0^2 J_2(R_0)}{J_1(R_0)}.$$
(36)

Figure 4 shows plots of the scaled free-surface profile of a pendant droplet h/θ given by Eq. (33) for a range of values of the scaled volume V/θ given by Eq. (35). In particular, Fig. 4 shows that, unlike for a sessile droplet, the droplet widens to a finite limiting width and deepens as its volume increases. Figure 5 shows plots of the scaled maximum height at the middle of the droplet, h_m/θ , given by Eq. (34) and the contact radius *R* given implicitly by Eq. (35) as functions of the scaled volume V/θ . In particular, Fig. 5 shows that, unlike for a sessile droplet, both h_m/θ and *R* are monotonic in V/θ ,

with $h_{\rm m}$ increasing from zero at $V/\theta = 0$ to infinity in the limit $V/\theta \rightarrow \infty$ and *R* increasing from zero at $V/\theta = 0$ to a maximum value of $R = R_{\rm max} \simeq 3.832$, corresponding to the first zero of $J_1(R)$, in the limit $V/\theta \rightarrow \infty$. Moreover, comparison of Eqs. (23), (28), and (35) shows that the contact radius of a sessile droplet is always greater than that of a zero-gravity droplet with the same scaled volume, which in turn is always greater than that of a pendant droplet with the same scaled volume.

In the limit of small contact radius, $R \rightarrow 0^+$, corresponding to the limit of small scaled volume, $V/\theta \rightarrow 0^+$, the droplet becomes narrow and shallow according to

$$\frac{h}{\theta} = \frac{R^2 - r^2}{2R} + \frac{(R^2 - r^2)^2}{32R} + O\left(\frac{(R^2 - r^2)^3}{R}\right) \to 0^+,$$
(37)

$$\frac{h_{\rm m}}{\theta} = \frac{R}{2} + \frac{R^3}{32} + O(R^5) \to 0^+, \tag{38}$$

$$\frac{V}{\theta} = \frac{\pi R^3}{4} + \frac{\pi R^5}{96} + O(R^7) \to 0^+,$$
(39)

which, as for a sessile droplet, at leading order reduce to the solution for a zero-gravity droplet given by Eq. (28), but differ from the corresponding expressions for a sessile droplet given by Eqs. (25)–(27) at higher order.

On the other hand, in the limit of maximum contact radius, $R \rightarrow R_{\text{max}}^-$, corresponding to the limit of large scaled volume, $V/\theta \rightarrow \infty$, the droplet approaches a finite width and becomes deep according to

$$\frac{h}{\theta} \sim \frac{J_0(r) - J_0(R_{\max})}{J_2(R_{\max})(R_{\max} - R)},$$
(40)

$$\frac{h_{\rm m}}{\theta} \sim \frac{1 - J_0(R_{\rm max})}{J_2(R_{\rm max})(R_{\rm max} - R)} \to \infty, \tag{41}$$

$$\frac{V}{\theta} \sim \frac{\pi R_{\max}^2}{R_{\max} - R} \to \infty.$$
 (42)

In particular, Eqs. (40)–(42) show that, unlike for a sessile droplet, for a pendant droplet the assumption that the droplet is thin eventually fails in the limit $V/\theta \rightarrow \infty$. Hence, unlike



FIG. 4. Plots of the scaled free-surface profile h/θ of a pendant droplet as a function of r for (a) $V/\theta = 10, 20, ..., 100$ and (b) $V/\theta = 100, 200, ..., 1000$. The arrows indicate the direction of increasing V/θ . Note that the vertical scales in (a) and (b) differ by an order of magnitude.

for a sessile droplet, for a pendant droplet the solution of the evaporative problem for a thin droplet described in Sec. II C will also eventually fail in the limit $V/\theta \rightarrow \infty$. In the Appendix we validate the present approach by comparing the asymptotic solution for the evaporative flux from a thin droplet with the numerical solution for a nonthin droplet, and show that the asymptotic solution provides a good approximation for a range of values of V/θ for both sessile and pendant droplets.

IV. EVAPORATION IN THE EXTREME MODES

A. Evaporation in the CR mode

For a droplet evaporating in the CR mode, i.e., with $R \equiv R_0$ and $\theta = \theta(t)$, the global mass-conservation equation (20) becomes

$$\frac{\partial V}{\partial \theta} \frac{\mathrm{d}\theta}{\mathrm{d}t} = -4R_0. \tag{43}$$

Solving Eq. (43) yields the general (implicit) solution for the evolution of the droplet, namely,

$$t = \frac{V(R_0, 1) - V(R_0, \theta)}{4R_0},$$
(44)

where, here and subsequently, we express the volume $V = V(t) = V(R, \theta)$ as a function of the contact radius *R* and the contact angle θ of the droplet. Setting $\theta = 0$ in Eq. (44) yields the general (explicit) expression for the lifetime of a droplet evaporating in the CR mode, namely,

$$t_{\rm CR} = \frac{V(R_0, 1)}{4R_0}.$$
 (45)

For a sessile droplet, substituting the expression for V from Eq. (23) into Eqs. (44) and (45) yields

$$R \equiv R_0, \quad \theta = 1 - \frac{4I_1(R_0)}{\pi R_0 I_2(R_0)} t, \tag{46}$$



FIG. 5. Plots of (a) the scaled maximum height at the middle of a pendant droplet, h_m/θ , and (b) the contact radius *R* of the droplet as functions of the scaled volume V/θ . The dashed line in (b) corresponds to the limiting value $R = R_{max} \simeq 3.832$ as $V/\theta \rightarrow \infty$.



FIG. 6. Evolutions of (a) R, (b) θ , (c) h_m , and (d) V for a sessile (solid line), a zero-gravity (dotted line), and a pendant (dashed line) droplet evaporating in the CR mode when $V_0 = 10$. The squares in (a) correspond to $t = t_{CR} = t_{CR,s}$, $t_{CR,0}$, and $t_{CR,p}$.

and

$$t_{\rm CR} = t_{\rm CR,s} = \frac{\pi R_0 I_2(R_0)}{4I_1(R_0)}.$$
(47)

For a pendant droplet, the corresponding expressions are

$$R \equiv R_0, \quad \theta = 1 - \frac{4J_1(R_0)}{\pi R_0 J_2(R_0)} t, \tag{48}$$

and

$$t_{\rm CR} = t_{\rm CR,p} = \frac{\pi R_0 J_2(R_0)}{4 J_1(R_0)}.$$
(49)

Both here and for the other modes considered subsequently, the corresponding expressions for a zero-gravity droplet are recovered at leading order in the limit $R_0 \rightarrow 0^+$ (see, for example, Wilson and Duffy [24] and Table II of Wilson and D'Ambrosio [13]).

Figure 6 (included primarily for comparison with the corresponding results for other modes) shows the evolutions of R, θ , $h_{\rm m}$, and V for a sessile, a zero-gravity, and a pendant droplet all with the same initial volume $V_0 = 10$ evaporating in the CR mode, and illustrates that θ , $h_{\rm m}$, and V are all linearly decreasing functions of t.

In the limit of an initially small droplet, $V_0 \sim \pi R_0^3/4 \rightarrow 0^+$, corresponding to the limit $R_0 \rightarrow 0^+$, the evolution of θ and the lifetime t_{CR} are given by

$$\theta = 1 - \frac{16}{\pi R_0^2} \left(1 \pm \frac{R_0^2}{24} \right) t + O(R_0^4)$$
(50)

and

$$t_{\rm CR} = t_{\rm CR,0} \left(1 \mp \frac{R_0^2}{24} \right) + O(R_0^6) \to 0^+,$$

where $t_{\rm CR,0} = \frac{\pi R_0^2}{16},$ (51)

where, again, the upper and the lower signs correspond to a sessile and a pendant droplet, respectively, i.e., $t_{CR,0} = O(V_0^{2/3})$. In particular, at leading order in this limit we recover the well-known expressions for the evolution of a zero-gravity droplet in the CR mode, namely,

$$\theta = 1 - \frac{t}{t_{\text{CR},0}}, \quad \frac{V}{V_0} = 1 - \frac{t}{t_{\text{CR},0}}.$$
 (52)

In the limit of an initially large sessile droplet, $V_0 \sim \pi R_0^2 \rightarrow \infty$, corresponding to the limit $R_0 \rightarrow \infty$, the evolution



FIG. 7. Plot of the lifetime of a droplet evaporating in the CR mode, t_{CR} , as a function of the initial volume V_0 for a sessile (solid line), a zero-gravity (dotted line), and a pendant (dashed line) droplet.

of θ and the lifetime $t_{CR,s}$ are given by

$$\theta \sim 1 - \frac{4}{\pi R_0} t \tag{53}$$

and

$$t_{\mathrm{CR},\mathrm{s}} \sim \frac{\pi R_0}{4} \to \infty,$$
 (54)

i.e., $t_{CR,s} = O(V_0^{1/2})$.

On the other hand, in the limit of an initially large pendant droplet, $V_0 \sim \pi R_{\text{max}}^2 / (R_{\text{max}} - R_0) \rightarrow \infty$, corresponding to the limit $R_0 \rightarrow R_{\text{max}}^-$, the evolution of θ and the lifetime $t_{\text{CR},p}$ are given by

$$\theta \sim 1 - \frac{4(R_{\max} - R_0)}{\pi R_{\max}}t \tag{55}$$

and

$$t_{\mathrm{CR},\mathrm{p}} \sim \frac{\pi R_{\mathrm{max}}}{4(R_{\mathrm{max}} - R_0)} \to \infty, \tag{56}$$

i.e., $t_{CR,p} = O(V_0)$.

Figure 7 shows a plot of t_{CR} as a function of V_0 , and illustrates that t_{CR} is a monotonically increasing function of V_0 that satisfies $t_{\rm CR} = O(V_0^{2/3}) \rightarrow 0^+$ as $V_0 \rightarrow 0^+$ and $t_{\rm CR,s} =$ $O(V_0^{1/2}), t_{CR,0} = O(V_0^{2/3}), \text{ and } t_{CR,p} = O(V_0) \to \infty \text{ as } V_0 \to$ ∞ for a sessile, a zero-gravity, and a pendant droplet, respectively. Figure 7 also illustrates that a sessile droplet always evaporates faster, and hence has a shorter lifetime, than a zerogravity droplet with the same initial volume, which in turn always evaporates faster, and hence has a shorter lifetime, than a pendant droplet with the same initial volume. This behavior can be understood by recalling that R_0 is a monotonically increasing function of V_0 , that the contact radius of a sessile droplet is always greater than that of a zero-gravity droplet with the same initial volume, which in turn is always greater than that of a pendant droplet with the same initial volume, and that the total evaporative flux from a droplet F given by Eq. (19) is proportional to R_0 . Note that the observation that F is always larger for a sessile droplet than for a pendant droplet with the same volume is consistent with the numerical results of Tonini and Cossali [77] for nonthin droplets with contact angles less than or equal to $\pi/2$ mentioned in Sec. I.

B. Evaporation in the CA mode

For a droplet evaporating in the CA mode, i.e., with R = R(t) and $\theta \equiv 1$, the global mass-conservation equation (20) becomes

$$\frac{\partial V}{\partial R}\frac{\mathrm{d}R}{\mathrm{d}t} = -4R.\tag{57}$$

Solving Eq. (57) using integration by parts yields the general (implicit) solution for the evolution of the droplet, namely,

$$t = \frac{V(R_0, 1)}{4R_0} - \frac{V(R, 1)}{4R} + \int_R^{R_0} \frac{V(\tilde{R}, 1)}{4\tilde{R}^2} \,\mathrm{d}\tilde{R}.$$
 (58)

Taking the limit $R \rightarrow 0^+$ in Eq. (58) yields the general (explicit) expression for the lifetime of a droplet evaporating in the CA mode, namely,

$$t_{\rm CA} = \frac{V(R_0, 1)}{4R_0} + \int_0^{R_0} \frac{V(\tilde{R}, 1)}{4\tilde{R}^2} \,\mathrm{d}\tilde{R}.$$
 (59)

For a sessile droplet, substituting the expression for V from Eq. (23) into Eqs. (58) and (59) and evaluating the integrals yields

$$t = \frac{\pi}{4} \left[\frac{R_0 I_2(R_0)}{I_1(R_0)} - \frac{R I_2(R)}{I_1(R)} - \ln\left(\frac{R_0 I_1(R)}{R I_1(R_0)}\right) \right], \quad \theta \equiv 1,$$
(60)

and

$$t_{\rm CA} = t_{\rm CA,s} = \frac{\pi}{4} \left[\frac{R_0 I_2(R_0)}{I_1(R_0)} - \ln\left(\frac{R_0}{2I_1(R_0)}\right) \right].$$
(61)

For a pendant droplet, the corresponding expressions are

$$t = \frac{\pi}{4} \left[\frac{R_0 J_2(R_0)}{J_1(R_0)} - \frac{R J_2(R)}{J_1(R)} + \ln\left(\frac{R_0 J_1(R)}{R J_1(R_0)}\right) \right], \quad \theta \equiv 1,$$
(62)

and

1

$$t_{\rm CA} = t_{\rm CA,p} = \frac{\pi}{4} \left[\frac{R_0 J_2(R_0)}{J_1(R_0)} + \ln\left(\frac{R_0}{2J_1(R_0)}\right) \right].$$
 (63)

Note the different sign of the logarithmic terms in Eqs. (62) and (63) compared to that of the corresponding terms in Eqs. (60) and (61) for the sessile case.

Figure 8 shows the evolutions of R, θ , $h_{\rm m}$, and V for a sessile, a zero-gravity, and a pendant droplet all with the same initial volume $V_0 = 10$ evaporating in the CA mode, and shows that, at least for this value of V_0 , R, $h_{\rm m}$, and Vare all nonlinearly decreasing functions of t. However, unlike for the CR mode, in which this behavior occurs for sessile, zero-gravity, and pendant droplets for all values of V_0 , for the CA mode, the nonmonotonic variation of $h_{\rm m}/\theta$ with V_0 described in Sec. III A and shown in Fig. 3(a) means that for a sessile droplet with $V_0 > 73.175$, corresponding to $R_0 >$ 5.586, $h_{\rm m}$ (but not R or V) is a nonmonotonic function of twhich increases to $h_{\rm m} = h_{\rm m,max}$ before eventually decreasing to $h_{\rm m} = 0$ at $t = t_{\rm CA,s}$. This behavior is illustrated in Fig. 9, which shows the evolution of $h_{\rm m}$ for a range of values of V_0 satisfying $V_0 > 73.175$.

In the limit of an initially small droplet, $R_0 \rightarrow 0^+$, the evolution of *R* and the lifetime t_{CA} are given by

$$t = \frac{3\pi}{32} \left[R_0^2 - R^2 \mp \frac{5(R_0^4 - R^4)}{144} \right] + O(R_0^6) \qquad (64)$$



FIG. 8. Evolutions of (a) R, (b) θ , (c) h_m , and (d) V for a sessile (solid line), a zero-gravity (dotted line), and a pendant (dashed line) droplet evaporating in the CA mode when $V_0 = 10$. The squares in (b) correspond to $t = t_{CA} = t_{CA,s}$, $t_{CA,0}$, and $t_{CA,p}$.

and

$$t_{CA} = t_{CA,0} \left(1 \mp \frac{5R_0^2}{144} \right) + O(R_0^6) \to 0^+,$$

where $t_{CA,0} = \frac{3\pi R_0^2}{32}.$ (65)



FIG. 9. Evolution of $h_{\rm m}$ for a sessile droplet evaporating in the CA mode when $V_0 = 200, 400, \ldots, 1000 (>73.175)$. The arrow indicates the direction of increasing V_0 .

In particular, at leading order in this limit we recover the wellknown expressions for the evolution of a zero-gravity droplet in the CA mode, namely,

$$\left(\frac{R}{R_0}\right)^2 = 1 - \frac{t}{t_{\text{CA},0}}$$
 and $\left(\frac{V}{V_0}\right)^{2/3} = 1 - \frac{t}{t_{\text{CA},0}}$, (66)

which, since the square of the contact radius (and hence the square of the contact diameter) and the volume raised to the 2/3 power are linear in *t*, are sometimes referred to as the " d^2 law" and the "2/3 law", respectively (see, for example, Wilson and D'Ambrosio [13]). Moreover, comparing the expressions for $t_{CR,0}$ and $t_{CA,0}$ recovers the well-known result that $t_{CA,0} = 3t_{CR,0}/2 \rightarrow 0^+$, with the factor of 3/2 reflecting the fact that for a zero-gravity droplet the average value of *R*, and hence the average value of *F*, in the CA mode is two-thirds of that in the CR mode.

In the limit of an initially large sessile droplet, $R_0 \rightarrow \infty$, the evolution of *R* and the lifetime $t_{CA,s}$ are given by

$$t \sim \frac{\pi}{4} \left[2R_0 - \frac{RI_2(R)}{I_1(R)} - \ln\left(\frac{\left(2\pi R_0^3\right)^{1/2} I_1(R)}{R}\right) \right]$$
(67)

and

$$t_{\mathrm{CA},\mathrm{s}} \sim \frac{\pi}{4} \left[2R_0 - \frac{1}{2} \ln \left(\frac{\pi R_0^3}{2} \right) \right] \to \infty.$$
 (68)

In particular, at leading order in this limit we obtain

$$\frac{R}{R_0} \sim 1 - \frac{2t}{\pi R_0}$$
 and $\left(\frac{V}{V_0}\right)^{1/2} \sim 1 - \frac{2t}{\pi R_0}$, (69)

which, since the contact radius (and hence the contact diameter) and the square root of the volume are linear in *t*, we term the "*d* law" and the "1/2 law," respectively, by analogy with the corresponding results for a zero-gravity droplet given by Eqs. (66). Moreover, comparing Eqs. (54) and (68) reveals that $t_{CA,s} \sim \pi R_0/2 \sim 2t_{CR,s} \rightarrow \infty$, with the factor of 2 reflecting the fact that the average value of *R*, and hence the average value of *F*, in the CA mode is half of that in the CR mode at leading order in this limit.

On the other hand, in the limit of an initially large pendant droplet, $R_0 \rightarrow R_{\text{max}}^-$, the evolution of *R* and the lifetime $t_{\text{CA},p}$ are given by

$$t \sim \frac{\pi}{4} \left[\frac{R_{\max}}{R_{\max} - R_0} - \frac{RJ_2(R)}{J_1(R)} + \ln\left(\frac{R_{\max}J_1(R)}{RJ_2(R_{\max})(R_{\max} - R_0)}\right) \right]$$
(70)

and

$$t_{\text{CA,p}} \sim \frac{\pi}{4} \left[\frac{R_{\text{max}}}{R_{\text{max}} - R_0} + \ln\left(\frac{R_{\text{max}}}{2J_2(R_{\text{max}})(R_{\text{max}} - R_0)}\right) \right]$$

$$\rightarrow \infty. \tag{71}$$

In particular, at leading order in this limit we obtain

$$\frac{R}{R_0} \sim 1$$
 and $\frac{V}{V_0} \sim 1 - \frac{4(R_{\max} - R_0)t}{\pi R_{\max}}$, (72)

i.e., *R* and *V* (but not, of course, θ) behave as if the droplet was evaporating in the CR mode. Moreover, comparing Eqs. (56) and (71) reveals that $t_{CA,p} \sim \pi R_{max}/(4(R_{max} - R_0)) \sim t_{CR,p} \rightarrow \infty$, with the equality reflecting the fact that the lifetimes of the extreme modes coincide at leading order in this limit.

The expressions for the evolution of the volume of a droplet evaporating in the CA mode given by Eqs. (66), (69), and (72) are all of the form

$$\left(\frac{V}{V_0}\right)^m = 1 - \frac{t}{t_{\rm CA}} \tag{73}$$

with different values of the exponent *m*, and rearranging Eq. (73) yields

$$m = \frac{\ln\left(1 - \frac{t}{t_{CA}}\right)}{\ln\left(\frac{V}{V_0}\right)}.$$
(74)

Figure 10 shows plots of the instantaneous value of *m* given by Eq. (74) evaluated at $t = t_{CA}/2$ (the corresponding plots for other choices of *t* are very similar) as a function of $\ln(V_0)$ for a sessile and a pendant droplet evaporating in the CA mode. In particular, Fig. 10 illustrates how for a sessile droplet the evolution of the droplet transitions from the 2/3 law [Eq. (66)] to the 1/2 law [Eq. (69)] as V_0 increases, and how for a pendant droplet it transitions from the 2/3 law [Eq. (66)] to that of a droplet evaporating in the CR mode [Eq. (72)] as V_0 increases. Specifically, Fig. 10 shows how *m* decreases from



FIG. 10. Plot of the instantaneous value of the exponent *m* given by Eq. (74) evaluated at $t = t_{CA}/2$ as a function of $\ln(V_0)$ for a sessile (solid line) and a pendant (dashed line) droplet evaporating in the CA mode. The dotted lines correspond to the limiting values m = 1/2and m = 1 as $V_0 \rightarrow \infty$.

m = 2/3 for a zero-gravity droplet toward the limiting value m = 1/2 for a large sessile droplet, and how it increases from m = 2/3 toward the limiting value m = 1 for a large pendant droplet.

Figure 11 shows a plot of t_{CA} as a function of V_0 , and illustrates that t_{CA} has qualitatively the same behavior as t_{CR} shown in Fig. 7 and described in Sec. IV A. Furthermore, comparing Eqs. (45) and (59) reveals that the lifetime of a droplet evaporating in the CA mode is always longer than that of the same droplet evaporating in the CR mode, i.e., $t_{CR} < t_{CA}$ for the same value of V_0 . This behavior, which is well known for a zero-gravity droplet, is readily understood by recalling that whereas $R \equiv R_0$ is constant in the CR mode, $R \leq R_0$ is a decreasing function of t in the CA mode, and that the total evaporative flux F given by Eq. (10) is proportional to R.

V. EVAPORATION IN THE SS MODE

As described in Sec. I, the SS mode consists of a CR phase, which ends when $\theta = \theta^*$, where θ^* ($0 \le \theta^* \le 1$) is the critical receding contact angle, followed by a CA phase, which ends when R = 0.



FIG. 11. Plot of the lifetime of a droplet evaporating in the CA mode, t_{CA} , as a function of the initial volume V_0 for a sessile (solid line), a zero-gravity (dotted line), and a pendant (dashed line) droplet.

During the CR phase, the evolution of the droplet is governed by Eq. (43) and hence is given by Eq. (44) for $0 \le t \le t^*$, where t^* ($0 \le t^* \le t_{SS}$) is the critical time at which the CR phase ends and the CA phase begins. Setting $\theta = \theta^*$ in Eq. (44) yields

$$t^* = \frac{V(R_0, 1) - V(R_0, \theta^*)}{4R_0}.$$
(75)

Thereafter, during the CA phase, the evolution of the droplet is governed by Eq. (57) and hence, using Eq. (75), is given implicitly by

$$t = \frac{V(R_0, 1)}{4R_0} - \frac{V(R, \theta^*)}{4R} + \int_R^{R_0} \frac{V(\tilde{R}, \theta^*)}{4\tilde{R}^2} \,\mathrm{d}\tilde{R}.$$
 (76)

Taking the limit $R \rightarrow 0^+$ in Eq. (76) yields the lifetime of a droplet evaporating in the SS mode, namely,

$$t_{\rm SS} = \frac{V(R_0, 1)}{4R_0} + \int_0^{R_0} \frac{V(\tilde{R}, \theta^*)}{4\tilde{R}^2} \,\mathrm{d}\tilde{R}.$$
 (77)

In the special cases $\theta^* = 0$ and $\theta^* = 1$ the SS mode reduces to the CR mode and the CA mode, respectively.

For a sessile droplet, the evolution of the droplet is given by Eq. (46) for $0 \le t \le t^*$ and

$$t = \frac{\pi}{4} \left[\frac{R_0 I_2(R_0)}{I_1(R_0)} - \frac{\theta^* R I_2(R)}{I_1(R)} - \theta^* \ln\left(\frac{R_0 I_1(R)}{R I_1(R_0)}\right) \right],$$

$$\theta \equiv \theta^*$$
(78)

for $t^* \leq t \leq t_{SS}$, where

$$t^* = t_{\rm s}^* = \frac{\pi (1 - \theta^*) R_0 I_2(R_0)}{4 I_1(R_0)}$$
(79)

and

$$t_{\rm SS} = t_{\rm SS,s} = \frac{\pi}{4} \left[\frac{R_0 I_2(R_0)}{I_1(R_0)} - \theta^* \ln\left(\frac{R_0}{2I_1(R_0)}\right) \right].$$
(80)

For a pendant droplet, the evolution of the droplet is given by Eq. (48) for $0 \le t \le t^*$ and

$$t = \frac{\pi}{4} \left[\frac{R_0 J_2(R_0)}{J_1(R_0)} - \frac{\theta^* R J_2(R)}{J_1(R)} + \theta^* \ln\left(\frac{R_0 J_1(R)}{R J_1(R_0)}\right) \right],\$$

$$\theta \equiv \theta^*$$
(81)

for $t^* \leq t \leq t_{SS}$, where

$$t^* = t_{\rm p}^* = \frac{\pi (1 - \theta^*) R_0 J_2(R_0)}{4 J_1(R_0)}$$
(82)

and

$$t_{\rm SS} = t_{\rm SS,p} = \frac{\pi}{4} \left[\frac{R_0 J_2(R_0)}{J_1(R_0)} + \theta^* \ln\left(\frac{R_0}{2J_1(R_0)}\right) \right].$$
 (83)

Again, note the different sign of the logarithmic terms in Eqs. (81) and (83) compared to that of the corresponding terms in Eqs. (78) and (80) for the sessile case.

Figures 12 and 13 show plots of the free-surface profile h as a function of r at various times when $V_0 = 10$, and the evolutions of R, θ , h_m , and V for a range of values of V_0 for a sessile and a pendant droplet, respectively, evaporating in the SS mode when $\theta^* = 1/2$. In particular, these figures show that the behavior in this mode inherits the appropriate behaviors of the extreme modes described in Sec. IV in the corresponding

phases. For example, Fig. 12(d) shows that h_m is a linearly decreasing function of t in the CR phase, but is a nonmonotonic function of t in the CA phase when $V_0 > 73.175$, corresponding to $R_0 > 5.586$.

In the limit of an initially small droplet, $R_0 \rightarrow 0^+$, the evolutions of *R* and θ are given by Eq. (50) for $0 \le t \le t^*$ and by

$$t = \frac{\pi}{32} \bigg[(2+\theta^*) R_0^2 - 3\theta^* R^2 \mp \frac{(4+\theta^*) R_0^4 - 5\theta^* R^4}{48} \bigg] + O(R_0^6)$$
(84)

for $t^* \leq t \leq t_{SS}$, where

$$t^* = t_0^* \left(1 \mp \frac{R_0^2}{24} \right) + O(R_0^6) \to 0^+,$$

where $t_0^* = \frac{\pi (1 - \theta^*) R_0^2}{16},$ (85)

and the lifetime t_{SS} is given by

$$t_{\rm SS} = t_{\rm SS,0} \left[1 \mp \frac{(4+\theta^*)R_0^2}{48(2+\theta^*)} \right] + O(R_0^6) \to 0^+,$$

where $t_{\rm SS,0} = \frac{\pi (2+\theta^*)R_0^2}{32}.$ (86)

In particular, comparing the expressions for $t_{CR,0}$, t_0^* , and $t_{SS,0}$ reveals that $t_0^* = (1 - \theta^*)t_{CR,0} \rightarrow 0^+$ and $t_{SS,0} = (2 + \theta^*)t_{CR,0}/2 \rightarrow 0^+$.

In the limit of an initially large sessile droplet, $R_0 \rightarrow \infty$, the evolutions of *R* and θ are given by Eq. (53) for $0 \le t \le t_s^*$ and by

$$t \sim \frac{\pi}{4} \left[(1 + \theta^*) R_0 - \frac{\theta^* R I_2(R)}{I_1(R)} - \theta^* \ln\left(\frac{\left(2\pi R_0^3\right)^{1/2} I_1(R)}{R}\right) \right]$$
(87)

for $t_{s}^{*} \leq t \leq t_{SS,s}$, where

$$t_{\rm s}^* \sim \frac{\pi (1-\theta^*)R_0}{4} \to \infty, \tag{88}$$

and the lifetime $t_{SS,s}$ is given by

$$t_{\text{SS,s}} \sim \frac{\pi}{4} \left[(1+\theta^*) R_0 - \frac{\theta^*}{2} \ln\left(\frac{\pi R_0^3}{2}\right) \right] \to \infty.$$
 (89)

In particular, comparing Eqs. (54), (88), and (89) reveals that $t_s^* \sim (1 - \theta^*) t_{CR,s} \to \infty$ and $t_{SS,s} \sim (1 + \theta^*) t_{CR,s} \to \infty$ at leading order in this limit.

On the other hand, in the limit of an initially large pendant droplet, $R_0 \rightarrow R_{\text{max}}^-$, the evolutions of *R* and θ are given by Eq. (55) for $0 \le t \le t_p^*$ and by

$$t \sim \frac{\pi}{4} \left[\frac{R_{\max}}{R_{\max} - R_0} - \frac{\theta^* R J_2(R)}{J_1(R)} + \theta^* \ln \left(\frac{R_{\max} J_1(R)}{R J_2(R_{\max})(R_{\max} - R_0)} \right) \right]$$
(90)



FIG. 12. Plot of (a) the free-surface profile h as a function of r at times $t = 0, t_s^*/2, t_s^*, 2(t_{SS,s} - t_s^*)/5, 3(t_{SS,s} - t_s^*)/4, 29(t_{SS,s} - t_s^*)/30$ when $V_0 = 10$, and the evolutions of (b) R, (c) θ , (d) h_m , and (e) V when $V_0 = 10, 20, \ldots, 100$ for a sessile droplet evaporating in the SS mode when $\theta^* = 1/2$. The dashed line in (a) corresponds to h at $t = t_s^*$, the circles in (b)–(e) correspond to $t = t_s^*$, and the squares in (c) correspond to $t = t_{SS,s}$. The arrow in (a) indicates the direction of increasing t.

for $t_{p}^{*} \leq t \leq t_{SS,p}$, where $t_{p}^{*} \sim \frac{\pi (1 - \theta^{*}) R_{\max}}{4(R_{\max} - R_{0})} \rightarrow \infty$, (91)

and the lifetime $t_{SS,p}$ is given by

$$t_{\rm SS,p} \sim \frac{\pi}{4} \left[\frac{R_{\rm max}}{R_{\rm max} - R_0} + \theta^* \ln\left(\frac{R_{\rm max}}{2J_2(R_{\rm max})(R_{\rm max} - R_0)}\right) \right]$$

$$\rightarrow \infty. \tag{92}$$

In particular, comparing Eqs. (56), (71), (91), and (92) reveals that $t_p^* \sim (1 - \theta^*) t_{\text{CR},p} \to \infty$ and $t_{\text{SS},p} \sim t_{\text{CR},p} \sim t_{\text{CA},p} \to \infty$ at leading order in this limit.

Figure 14 shows a plot of t_{SS} as a function of V_0 , and illustrates not only that t_{SS} has qualitatively the same behavior as t_{CR} and t_{CA} , but also that the lifetime of a droplet evaporating in the SS mode is a monotonically increasing function of θ^* , *i.e.* t_{SS} increases from $t_{SS} = t_{CR}$ when $\theta^* = 0$ to $t_{SS} = t_{CA}$



FIG. 13. Plot of (a) the free-surface profile h as a function of r at times t = 0, $t_p^*/2$, t_p^* , $2(t_{SS,p} - t_p^*)/5$, $3(t_{SS,p} - t_p^*)/4$, $29(t_{SS,p} - t_p^*)/30$ when $V_0 = 10$, and the evolutions of (b) R, (c) θ , (d) h_m , and (e) V when $V_0 = 10, 20, ..., 100$ for a pendant droplet evaporating in the SS mode when $\theta^* = 1/2$. The dashed line in (a) corresponds to h at $t = t_p^*$, the circles in (b)–(e) correspond to $t = t_p^*$, and the squares in (c) correspond to $t = t_{SS,p}$. The arrow in (a) indicates the direction of increasing t.

when $\theta^* = 1$, and satisfies $t_{CR} \leq t_{SS} \leq t_{CA}$ for the same value of V_0 .

VI. EVAPORATION IN THE SJ MODE

As described in Sec. I, the SJ mode consists of a (theoretically infinite) series of CR phases, separated by short (theoretically instantaneous) jump phases in which the contact line rapidly recedes and the contact angle rapidly increases from θ_{\min} to θ_{\max} , where θ_{\min} and θ_{\max} ($0 \le \theta_{\min} \le \theta_{\max} \le 1$) are the critical depinning and pinning contact angles, respectively. We denote the constant value of the contact radius during the *n*th CR phase by $R = R_n$ and the time at which the *n*th jump phase occurs by $t = t_n$ for n = 1, 2, 3, ..., and



FIG. 14. Plot of the lifetime of a droplet evaporating in the SS mode t_{SS} as a function of V_0 for a sessile and a pendant droplet when $\theta^* = 1/4$, 1/2, 3/4 (solid lines). The dotted and dashed lines correspond to $\theta^* = 0$ (i.e., the CR mode) and $\theta^* = 1$ (i.e., the CA mode), respectively. The corresponding curves for a zero-gravity droplet (which lie between those for a sessile and a pendant droplet of the same initial volume) are omitted for clarity.

so the *n*th CR phase begins at $t = t_{n-1}$ and ends at $t = t_n$, at which time the *n*th jump phase, in which *R* jumps instantaneously down from $R = R_n$ to $R = R_{n+1} (\leq R_n)$ and θ jumps instantaneously up from $\theta = \theta_{\min}$ to $\theta = \theta_{\max} (\geq \theta_{\min})$, occurs. The first CR phase begins with $R = R_1 = R_0$ and $\theta = 1$ at $t = t_0 = 0$.

During the *n*th (n = 2, 3, 4, ...) CR phase the evolution of the droplet is governed by Eq. (43) with R_n in place of R_0 and hence is given by

$$R \equiv R_n, \quad t = t_{n-1} + \frac{V(R_n, \theta_{\max}) - V(R_n, \theta)}{4R_n}$$
(93)

for $t_{n-1} < t < t_n$. Setting $\theta = \theta_{\min}$ in Eq. (93) yields

$$t_n = t_{n-1} + \frac{V(R_n, \theta_{\max}) - V(R_n, \theta_{\min})}{4R_n}.$$
 (94)

The behavior is the same during the first CR phase, except with θ_{max} replaced by 1. Mass is conserved during the *n*th (n = 1, 2, 3, ...) jump phase, and so

$$V(R_n, \theta_{\min}) = V(R_{n+1}, \theta_{\max})$$
(95)

at $t = t_n$. The lifetime of a droplet evaporating in the SJ mode is given by

$$t_{\rm SJ} = \lim_{n \to \infty} t_n. \tag{96}$$

In the special case $\theta_{\min} = 0$ and in the limit $\theta_{\min} \rightarrow \theta_{\max}^- \rightarrow 1^-$ the SJ mode reduces to the CR mode and the CA mode, respectively.

For a sessile droplet, the evolution of the droplet during the nth (n = 2, 3, 4, ...) CR phase is given by

$$R \equiv R_n, \quad \theta = \theta_{\max} - \frac{4I_1(R_n)}{\pi R_n I_2(R_n)} (t - t_{n-1,s})$$
(97)

for $t_{n-1,s} < t < t_{n,s}$, where

$$t_n = t_{n,s} = t_{n-1,s} + (\theta_{\max} - \theta_{\min}) \frac{\pi R_n I_2(R_n)}{4I_1(R_n)}, \quad (98)$$

i.e.,

$$t_{n,s} = \frac{\pi}{4} \left[(1 - \theta_{\min}) \frac{R_0 I_2(R_0)}{I_1(R_0)} + (\theta_{\max} - \theta_{\min}) \sum_{k=2}^n \frac{R_k I_2(R_k)}{I_1(R_k)} \right].$$
(99)

The condition that mass is conserved during the *n*th (n = 1, 2, 3, ...) jump phase [Eq. (95)] yields

$$\frac{\theta_{\min}R_n^2 I_2(R_n)}{I_1(R_n)} = \frac{\theta_{\max}R_{n+1}^2 I_2(R_{n+1})}{I_1(R_{n+1})},$$
(100)

and hence

$$\frac{R_{n+1}^2 I_2(R_{n+1})}{I_1(R_{n+1})} = \frac{\theta_{\min}}{\theta_{\max}} \frac{R_n^2 I_2(R_n)}{I_1(R_n)} = \left(\frac{\theta_{\min}}{\theta_{\max}}\right)^n \frac{R_0^2 I_2(R_0)}{I_1(R_0)}.$$
(101)

Taking the limit $n \to \infty$ in Eq. (99) yields

$$t_{\rm SJ} = t_{\rm SJ,s} = \frac{\pi}{4} \left[(1 - \theta_{\rm min}) \frac{R_0 I_2(R_0)}{I_1(R_0)} \right] + (\theta_{\rm max} - \theta_{\rm min}) \sum_{k=2}^{\infty} \frac{R_k I_2(R_k)}{I_1(R_k)} \right], \quad (102)$$

where R_k for k = 2, 3, 4, ... are given implicitly in terms of θ_{max} , θ_{min} , and R_0 by Eq. (101).

For a pendant droplet, the evolution of the droplet during the *n*th (n = 2, 3, 4, ...) CR phase is given by

$$R \equiv R_n, \quad \theta = \theta_{\max} - \frac{4J_1(R_n)}{\pi R_n J_2(R_n)} (t - t_{n-1,p})$$
(103)

for $t_{n-1,p} < t < t_{n,p}$, where

$$t_n = t_{n,p} = t_{n-1,p} + (\theta_{\max} - \theta_{\min}) \frac{\pi R_n J_2(R_n)}{4J_1(R_n)}, \quad (104)$$

i.e.,

$$t_{n,p} = \frac{\pi}{4} \left[(1 - \theta_{\min}) \frac{R_0 J_2(R_0)}{J_1(R_0)} + (\theta_{\max} - \theta_{\min}) \sum_{k=2}^n \frac{R_k J_2(R_k)}{J_1(R_k)} \right].$$
(105)

The condition that mass is conserved during the *n*th (n = 1, 2, 3, ...) jump phase [Eq. (95)] yields

$$\frac{\theta_{\min}R_n^2 J_2(R_n)}{J_1(R_n)} = \frac{\theta_{\max}R_{n+1}^2 J_2(R_{n+1})}{J_1(R_{n+1})},$$
 (106)

and hence

$$\frac{R_{n+1}^2 J_2(R_{n+1})}{J_1(R_{n+1})} = \frac{\theta_{\min}}{\theta_{\max}} \frac{R_n^2 J_2(R_n)}{J_1(R_n)} = \left(\frac{\theta_{\min}}{\theta_{\max}}\right)^n \frac{R_0^2 J_2(R_0)}{J_1(R_0)}.$$
(107)

Taking the limit $n \to \infty$ in Eq. (105) yields

$$t_{\rm SJ} = t_{\rm SJ,p} = \frac{\pi}{4} \left[(1 - \theta_{\rm min}) \frac{R_0 J_2(R_0)}{J_1(R_0)} + (\theta_{\rm max} - \theta_{\rm min}) \sum_{k=2}^{\infty} \frac{R_k J_2(R_k)}{J_1(R_k)} \right], \quad (108)$$



FIG. 15. Plot of (a) the free-surface profile *h* as a function of *r* at $t = t_{n,s}$ for n = 1, 2, 3, ... and the evolutions of (b) *R*, (c) θ , (d) h_m , and (e) *V* when $V_0 = 10$ for a sessile droplet evaporating in the SJ mode when $\theta_{\min} = 1/2$ and $\theta_{\max} = 1$. The solid and dash-dotted lines in (a) correspond to *h* at $t = t_{n-1,s}$ and at $t = t_{n,s}$, respectively, the dotted and dashed lines in (b)–(e) correspond to $\theta_{\min} = 0$ (i.e., the CR mode) and the limit $\theta_{\min} \rightarrow \theta_{\max}^- = 1^-$ (i.e., the CA mode), respectively, and the dots in (e) correspond to $t = t_{n,s}$. The arrow in (a) indicates the direction of increasing *t*.

where R_k for k = 2, 3, 4, ... are given implicitly in terms of θ_{max} , θ_{min} , and R_0 by Eq. (107).

Figures 15 and 16 show plots of the free-surface profile *h* as a function of *r* at $t = t_n$ for n = 1, 2, 3, ..., and the evolutions of *R*, θ , h_m , and *V* when $V_0 = 10$ for a sessile and a pendant droplet, respectively, evaporating in the SJ mode when $\theta_{\min} = 1/2$ and $\theta_{\max} = 1$. In particular, these figures illustrate that $R = R_n$ is constant and θ , h_m , and V are linearly decreasing functions of t during each CR phase, and that R jumps down discontinuously and θ and h_m jump up discontinuously, but V is continuous, during each jump phase. They also illustrate how an infinite number of CR and jump phases combine to give the finite lifetime t_{SJ} .



FIG. 16. Plot of (a) the free-surface profile *h* as a function of *r* at $t = t_{n,p}$ for n = 1, 2, 3, ... and the evolutions of (b) *R*, (c) θ , (d) h_m , and (e) *V* when $V_0 = 10$ for a pendant droplet evaporating in the SJ mode when $\theta_{\min} = 1/2$ and $\theta_{\max} = 1$. The solid and dash-dotted lines in (a) correspond to *h* at $t = t_{n-1,p}$ and at $t = t_{n,p}$, respectively, the dotted and dashed lines in (b)–(e) correspond to $\theta_{\min} = 0$ (i.e., the CR mode) and the limit $\theta_{\min} \rightarrow \theta_{\max}^- = 1^-$ (i.e., the CA mode), respectively, and the dots in (e) correspond to $t = t_{n,p}$. The arrow in (a) indicates the direction of increasing *t*.

In the limit of an initially small droplet, $R_0 \rightarrow 0^+$, the evolutions of *R* and θ during the *n*th CR phase are given by

for
$$t_{n-1,0} < t < t_{n,0}$$
, where

and the lifetime t_{SJ} is given by

$$t_{\rm SJ} \sim t_{\rm SJ,0} = \frac{\pi R_0^2}{16} \left[1 - \theta_{\rm max} + (\theta_{\rm max} - \theta_{\rm min}) \frac{\theta_{\rm max}^{2/3}}{\theta_{\rm max}^{2/3} - \theta_{\rm min}^{2/3}} \right] \to 0^+.$$
(111)

In the limit of an initially large sessile droplet, $R_0 \rightarrow \infty$, the evolutions of R and θ during the *n*th CR phase are given by

$$R \equiv R_n \sim \left(\frac{\theta_{\min}}{\theta_{\max}}\right)^{(n-1)/2} R_0, \quad \theta \sim \theta_{\max} - \frac{4}{\pi R_n} (t - t_{n-1,s})$$
(112)

for $t_{n-1,s} < t < t_{n,s}$, where

$$t_{n,s} \sim \frac{\pi R_0}{4} \left\{ 1 - \theta_{\max} + (\theta_{\max} - \theta_{\min}) \frac{\left[1 - \left(\frac{\theta_{\min}}{\theta_{\max}}\right)^{n/2}\right]}{\left[1 - \left(\frac{\theta_{\min}}{\theta_{\max}}\right)^{1/2}\right]} \right\},\tag{113}$$

and the lifetime $t_{SJ,s}$ is given by

$$t_{\rm SJ,s} \sim \frac{\pi R_0}{4} \left[1 - \theta_{\rm max} + (\theta_{\rm max} - \theta_{\rm min}) \frac{\theta_{\rm max}^{1/2}}{\theta_{\rm max}^{1/2} - \theta_{\rm min}^{1/2}} \right] \to \infty.$$
(114)

On the other hand, in the limit of an initially large pendant droplet, $R_0 \rightarrow R_{\text{max}}^-$, the evolutions of *R* and θ during the *n*th CR phase are given by

$$R \equiv R_n \sim R_{\max} - \left(\frac{\theta_{\max}}{\theta_{\min}}\right)^{n-1} (R_{\max} - R_0),$$

$$\theta \sim \theta_{\max} - \frac{4(R_{\max} - R_n)}{\pi R_{\max}} (t - t_{n-1,p})$$
(115)

for $t_{n-1,p} < t < t_{n,p}$, where

$$t_{n,p} \sim \frac{\pi R_{\max}}{4(R_{\max} - R_0)} \bigg[1 - \theta_{\max} \bigg(\frac{\theta_{\min}}{\theta_{\max}} \bigg)^n \bigg], \qquad (116)$$

and the lifetime $t_{SJ,p}$ is given by

$$t_{\rm SJ,p} \sim \frac{\pi R_{\rm max}}{4(R_{\rm max} - R_0)} \to \infty,$$
 (117)

which is independent of θ_{\min} and θ_{\max} . In particular, comparing Eqs. (56), (71), (92), and (117) reveals that $t_{SJ,p} \sim t_{CR,p} \sim t_{CA,p} \sim t_{SS,p} \rightarrow \infty$ at leading order in this limit.

Figure 17 shows a plot of t_{SJ} as a function of V_0 , and illustrates not only that t_{SJ} has qualitatively the same behavior as the lifetimes of the other three modes described previously, but also that the lifetime of a droplet evaporating in the SJ mode is a monotonically increasing function of θ_{min} for a fixed value of θ_{max} , and that when $\theta_{max} = 1$ it increases from $t_{SJ} =$ t_{CR} when $\theta_{min} = 0$ to $t_{SJ} = t_{CA}$ in the limit $\theta_{min} \rightarrow \theta_{max}^- = 1^-$, and satisfies $t_{CR} \leq t_{SJ} \leq t_{CA}$ for the same value of V_0 .

VII. CONCLUSIONS

In the present work we performed a comprehensive study of the effect of gravity-induced shape change on the diffusionlimited evaporation of thin sessile and pendant droplets on



FIG. 17. Plot of the lifetime of a droplet evaporating in the SJ mode, t_{SJ} , as a function of V_0 for a sessile and a pendant droplet when $\theta_{\min} = 1/4$, 1/2, 3/4 and $\theta_{\max} = 1$ (solid lines). The dotted and dashed lines correspond to $\theta_{\min} = 0$ (i.e., the CR mode) and the limit $\theta_{\min} \rightarrow \theta_{\max}^- = 1^-$ (i.e., the CA mode), respectively. The corresponding curves for a zero-gravity droplet (which lie between those for a sessile and a pendant droplet of the same initial volume) are omitted for clarity.

a horizontal substrate. Specifically, we obtained theoretical predictions for the evolution, and hence the lifetime, of sessile and pendant droplets evaporating in four modes of evaporation, namely, the CR, CA, SS, and SJ modes. In particular, we showed that gravity-induced shape change can cause quantitative differences in the evolution of sessile and pendant droplets compared to that of a zero-gravity droplet. For example, whereas sessile and pendant droplets evaporating in the CR mode evolve in qualitatively the same manner as a zerogravity droplet, i.e., their contact angles θ , and hence their volumes V, decrease linearly in t, the evolution of droplets evaporating in the CA mode is more complicated. Specifically, while a zero-gravity droplet evaporating in the CA mode evolves according to the well-known d^2 and 2/3 laws given by Eq. (66), an initially large sessile droplet evolves according to qualitatively different d and 1/2 laws given by Eq. (69), and an initially large pendant droplet evolves with the contact radius R and the volume V (but not, of course, the contact angle θ) behaving as if the droplet was evaporating in the CR mode according to Eq. (72). We also found, perhaps somewhat unexpectedly, and unlike for a droplet evaporating in the CR mode and for zero-gravity and pendant droplets evaporating in the CA mode, that the maximum height of a sessile droplet evaporating in the CA mode is a nonmonotonic function of t when $V_0 > 73.175$, corresponding to $R_0 > 5.586$. Furthermore, we found that, consistent with the results of Kadhim et al. [46] for sessile droplets evaporating in the SS mode mentioned in Sec. I, for all four modes of evaporation a sessile droplet always evaporates faster, and hence has a shorter lifetime, than a zero-gravity droplet with the same initial volume, which in turn always evaporates faster, and hence has a shorter lifetime, than a pendant droplet with the same initial volume. This behavior can be understood by recalling that the contact radius of a sessile droplet is always greater than that of a zero-gravity droplet with the same initial volume, which in turn is always greater than that of a pendant droplet with the same initial volume, and that the total evaporative flux

from a droplet F is proportional to R. We also showed that for all four modes of evaporation the lifetime of a droplet is a monotonically increasing function of the initial volume of the droplet that becomes short like $O(V_0^{2/3}) \rightarrow 0^+$ in the limit of an initially small droplet, $V_0 \rightarrow 0^+$, and becomes long like $O(V_0^{1/2})$, $O(V_0^{2/3})$, and $O(V_0) \to \infty$ for a sessile, a zero-gravity, and a pendant droplet, respectively, in the limit of an initially large droplet, $V_0 \rightarrow \infty$. In addition, we found that the lifetime of a droplet evaporating in the CA mode is always longer than that of the same droplet evaporating in the CR mode, and that the lifetime of a droplet evaporating in the SS mode with critical receding contact angle θ^* ($0 \le \theta^* \le 1$) and the lifetime of a droplet evaporating in the SJ mode with critical depinning and pinning contact angles θ_{\min} and θ_{\max} $(0 \leq \theta_{\min} \leq \theta_{\max} = 1)$ both always lie between the lifetimes of the same droplet evaporating in the extreme modes, i.e., $t_{\rm CR} \leq t_{\rm SS}, t_{\rm SJ} \leq t_{\rm CA}$ for the same value of V_0 .

It should, of course, be noted that in the present work we have focused on just one of the ways in which gravity can influence the evaporation of a sessile droplet, and in practice others ways can be equally or more important. In particular, larger droplets are likely to generate more buoyancy-driven convection in the atmosphere than smaller ones (see, for example, Larson [4]), which could quantitatively or even qualitatively change their behavior from that predicted by the present work.

In the Appendix, we validated the present approach by comparing the asymptotic solution for the evaporative flux from a thin droplet described in Sec. II C with the numerical solution for a nonthin droplet. However, for a nonthin droplet the situation is more complicated (for example, for a zero-gravity droplet, Picknett and Bexon [17] showed that $\hat{t}_{CA} < \hat{t}_{CR}$ for sufficiently large values of $\hat{\theta}_0$, and Stauber *et al.* [22,23] showed that \hat{t}_{SS} does not always lie between \hat{t}_{CR} and \hat{t}_{CA} for $\pi/2 < \hat{\theta}_0 < \pi$), and so a full description of the evolution of a nonthin droplet including the effect of gravity-induced shape change remains an open problem. It would also be of interest to use the present approach to investigate the effect of gravity on two closely related problems mentioned in Sec. I,

namely, the evaporation of a droplet on an inclined substrate and the deposition onto the substrate from an evaporating particle-laden droplet.

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The original concept was proposed by B.R.D., H.-M.D'A. performed the original calculations in close collaboration with S.K.W., A.W.W., and B.R.D. and created all of the figures. The manuscript was prepared by S.K.W. based on an initial draft by H.-M.D'A. H.-M.D'A., S.K.W., and A.W.W. revised, reviewed, and approved the final version of the manuscript prior to submission.

DATA AVAILABILITY

No data were created or analyzed in this study. All of the results reported in the present work can be reproduced from the equations detailed within the manuscript.

APPENDIX: THE EVAPORATIVE FLUX FROM A NONTHIN DROPLET

In this appendix, we validate the present approach by comparing the asymptotic solution for the evaporative flux from a thin droplet described in Sec. II C with the numerical solution for a nonthin droplet.

Nondimensionalizing the variables appropriately for a nonthin droplet according to

$$=\hat{\ell}r, \quad \hat{z}=\hat{\ell}\tilde{z}, \quad \hat{R}=\hat{\ell}R, \quad \hat{h}=\hat{\ell}\tilde{h}, \quad \hat{V}=\hat{\ell}^{3}\tilde{V},$$
$$\hat{p}-\hat{p}_{a}=\frac{\hat{\sigma}}{\hat{\ell}}\tilde{p}, \quad \hat{t}=\frac{\hat{\rho}\hat{\ell}^{2}}{\hat{D}(\hat{c}_{\text{sat}}-\hat{c}_{\infty})}\tilde{t}$$
(A1)



r

FIG. 18. Plots of the scaled local evaporative flux *JR* as a function of the scaled radial coordinate r/R for (a) a sessile droplet with $\theta = 2\pi/9$ and (b) a pendant droplet with $\theta = \pi/36$ showing the leading-order asymptotic solution for a thin droplet given by Eq. (18) (dashed lines) and the numerical solution for a nonthin droplet (solid lines) for $V/\theta = 1, 5, 10, 20, 40, 60, 80$, and 100. The arrows indicate the direction of increasing V/θ .

for the droplet and Eq. (13) again for the atmosphere, the governing equation for the nondimensional free-surface profile \tilde{h} of a nonthin droplet is

$$\frac{\partial}{\partial r} \left[\frac{1}{\left(1 + \left(\frac{\partial \tilde{h}}{\partial r}\right)^2\right)^{3/2}} \frac{\partial^2 \tilde{h}}{\partial r^2} + \frac{1}{r \left(1 + \left(\frac{\partial \tilde{h}}{\partial r}\right)^2\right)^{1/2}} \frac{\partial \tilde{h}}{\partial r} \mp \tilde{h} \right]$$
$$= 0, \qquad (A2)$$

and so the hydrostatic problem for a nonthin droplet is obtained by replacing Eq. (14) with Eqs. (A2) and (15) with the contact-line conditions

$$\tilde{h} = 0, \quad \frac{\partial \tilde{h}}{\partial r} = -\tan\theta \quad \text{at} \quad r = R.$$
 (A3)

The evaporative problem for a nonthin droplet is the same as that for a thin droplet described in Sec. II C, except that the condition c = 1 on z = 0 is replaced by c = 1 on the free surface of the droplet, the expression $J = -\partial c/\partial z$ on z = 0 for $0 \le r \le R$ is replaced by $J = -\partial c/\partial \tilde{\mathbf{n}}$ on the free surface of the droplet, where $\tilde{\mathbf{n}}$ is the unit outward normal to the free-surface profile, and *F* is given in terms of the integral of *J* over the free surface of the droplet, with the additional complication that the free-surface profile \tilde{h} becomes multivalued for sufficiently large pendant droplets (see, for example, Padday and Pitt [88] and Kumar *et al.* [89]).

The hydrostatic and evaporative problems were solved using *Mathematica* and COMSOL Multiphysics 5.5a, respectively. Specifically, the hydrostatic problem was solved in *Mathematica* using NDSolve. The free-surface profile of the droplet \tilde{h} was then imported into the geometry section of COMSOL as an interpolating function, and the evaporative problem was solved in COMSOL to determine the concentration of vapor in the atmosphere, *c*, and hence the local and total evaporative fluxes, *J* and *F*.

Figure 18 shows plots of the scaled local evaporative flux *JR* as a function of the scaled radial coordinate r/R for a sessile [Fig. 18(a)] and a pendant [Fig. 18(b)] droplet showing the leading-order asymptotic solution for a thin droplet given by Eq. (18) (dashed lines) and the numerical solution for a nonthin droplet (solid lines) for a range of values of V/θ . In particular, Fig. 18(a) shows that, since, as described in Sec. III A, a sessile droplet becomes wider (and hence thinner) as V/θ increases, the asymptotic solution for a thin droplet provides an increasingly more accurate approximation as V/θ increases. On the other hand, Fig. 18(b) shows that, since, as described in Sec. III B, a pendant droplet becomes deeper (and hence thicker) as V/θ increases, the asymptotic solution for a thin droplet in Sec. III B, a pendant droplet becomes deeper (and hence thicker) as V/θ increases, the asymptotic solution for a thin droplet provides an increasingly less accurate approximation as thin droplet provides an increasingly less accurate approximation as V/θ increases.



FIG. 19. Plot of the total evaporative flux *F* as a function of the scaled volume V/θ . The upper solid line and the circles show the leading-order asymptotic solution for a thin droplet given by Eq. (19) and the numerical solution for a nonthin droplet for a sessile droplet with $\theta = 2\pi/9$, while the lower solid line and the squares show the corresponding solutions for a pendant droplet with $\theta = \pi/36$. The dotted and dashed lines show the solutions for a nonthin droplet in the absence of the effect of gravity (i.e., for the corresponding spherical-cap droplet) for $\theta = 2\pi/9$ and $\theta = \pi/36$, respectively.

Figure 19 shows a plot of the total evaporative flux F as a function of the scaled volume V/θ . The upper solid line and the circles show the leading-order asymptotic solution for a thin droplet given by Eq. (19) and the numerical solution for a nonthin droplet for a sessile droplet with $\theta = 2\pi/9$, while the lower solid line and the squares show the corresponding solutions for a pendant droplet with $\theta = \pi/36$. In particular, Fig. 19 shows that, despite the differences between the asymptotic and numerical solutions for the local evaporative flux for a pendant droplet evident in Fig. 18(b), the asymptotic solution for the total evaporative flux for a thin droplet given by Eq. (19) provides a good approximation (specifically, accurate to less than 1% for the sessile droplet and to less than 3% for the pendant droplet for the values shown in the figure) to the key quantity driving the evolution of the droplet for a range of values of V/θ for both sessile and pendant droplets. However, as mentioned earlier, the free-surface profile \tilde{h} of a pendant droplet becomes multivalued for sufficiently large values of V/θ , and so, unlike for sessile droplets, the present approach will eventually fail for sufficiently large pendant droplets. Figure 19 also includes dotted and dashed lines showing the corresponding solutions for a nonthin droplet in the absence of the effect of gravity (i.e., for the corresponding spherical-cap droplet) (see, for example, Wilson and D'Ambrosio [13]), and illustrates that the common approach of ignoring the effect of gravity leads to increasingly inaccurate predictions for the total evaporative flux from both sessile and pendant droplets as V/θ increases.

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