



Dependence of fluid flows in an evaporating sessile droplet on the characteristics of the substrate



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ABSTRACT

Temperature distributions and the corresponding vortex structures in an evaporating sessile droplet are obtained by performing detailed numerical calculations. A Marangoni convection induced by thermal conduction in the drop and the substrate is demonstrated to be able to result not only in a single vortex, but also in two or three vortices, depending on the ratio of substrate to fluid thermal conductivities, on the substrate thickness and the contact angle. The “phase diagrams” containing information on the number, orientation and spatial location of the vortices for quasistationary fluid flows are presented and analysed. The results obtained demonstrate that the fluid flow structure in evaporating droplets can be influenced in a controlled manner by selecting substrates with appropriate properties.

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1. Introduction

It is known that the evaporating flux density along the surface of a drying sessile droplet is inhomogeneous and diverges on approach to the pinned contact line [1,2]. The evaporation can induce temperature variations in the vicinity of the substrate–liquid interface and along the drop surface. A temperature variation along the liquid–vapor interface can in turn generate a thermocapillary flow inside the drop which has been intensively studied (see, for example, the review articles [3,4] and references therein). Sessile drop evaporation processes and structures of the fluid flow are of interest for important applications in ink-jet printing [5,6], spraying of pesticides [7], micro/nano fabrication [8,9], thin film coatings [10], biochemical assays [11], spray cooling [12], disease diagnosis [13,14], deposition of DNA/RNA microarrays [15–17], and manufacture of novel optical and electronic materials [18].

The substrate temperature distribution under the drop can be measured using thermo-chromic liquid crystals [19] and with IR thermography [20]. The substrate properties play an important role in the nanotechnology applications of the problem. One of the examples is self-assembly of superlattices of nanoparticles taking place during evaporation of colloidal solutions. It is known

that substrate characteristics can strongly influence both the deposition patterns and the self-assembly process [21–26].

Numerical calculations of Marangoni convection in an axially symmetrical evaporating droplet agree well with corresponding experimental data [27,28]. A sensitivity of Marangoni fluid flows to the droplet contact angle is known since Hu and Larson demonstrated that fluid circulation in the vortex can reverse its sign at a critical contact angle for a drop placed onto substrates with finite thicknesses [29]. It was originally observed and described by Ristenpart et al. that the circulation direction depends on the substrate to liquid ratio of the thermal conductivities [30]. Specifically, the authors found that the ratio determines the sign of the tangential component of the temperature gradient at the surface close to the contact line, and, therefore, it determines a direction of the circulation in that region. Assuming the key role of a small vicinity of the contact line in forming the circulation direction in a single vortex, the shape of the liquid–vapor interface in that small region was approximated as a plane, which is actually inappropriate since the three-phase contact line has finite radius of curvature. However, the conditions for the circulation sign change have been found within such framework. While the approach and predictions of Ref. [30] generally are qualitatively insightful and quite useful, the particular approximations made and the corresponding quantitative results obtained in [30] have not been justified by more accurate numerical calculations. For example, it follows from the analytical results of [30] that for $k_R < 1.45$ and $k_R > 2$ the circulation direction is insensitive to the contact angle. The results

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Table 1
The notations and the parameter values used for obtaining the evaporation rates, temperature distribution and hydrodynamics in the drop. The tabular data are taken from [37].

Drop parameters	Initial temperature	$T_0 = 293.15 \text{ K}$
	Contact line radius	$R = 10^{-3} \text{ m}$
	Contact angle	θ
Substrate parameters	Radius	$R_S = 1.25 \cdot 10^{-3} \text{ m}$
	Thickness	h_S
	Ratio of substrate thickness to contact line radius	$h_R = h_S/R$
	Thermal conductivity	k_S
	Substrate to liquid ratio of thermal conductivities	$k_R = k_S/k_L$
Fluid characteristics (1-hexanol)	Density	$\rho = 813.6 \text{ kg/m}^3$
	Molar mass	$\mu = 0.10217 \text{ kg/mole}$
	Thermal conductivity	$k_L = 0.15 \text{ W/(m}\cdot\text{K)}$
	Thermal diffusivity	$\kappa = k_L/(\rho c_p) = 7.84 \cdot 10^{-8} \text{ m}^2/\text{s}$
	Dynamic viscosity	$\eta = 4.578 \cdot 10^{-3} \text{ kg/(m}\cdot\text{s)}$
	Surface tension	$\sigma = 0.02581 \text{ kg/s}^2$
	Temperature derivative of surface tension	$-\partial\sigma/\partial T = 8.0 \cdot 10^{-5} \text{ kg/(s}^2\cdot\text{K)}$
	Latent heat of evaporation	$L = 6.03 \cdot 10^5 \text{ J/kg}$
1-hexanol Vapor characteristics	Diffusion constant	$D = 6.21 \cdot 10^{-6} \text{ m}^2/\text{s}$
	Saturated 1-hexanol vapor density	$u_s = 6.55 \cdot 10^{-3} \text{ kg/m}^3$

of more recent numerical calculations, as well as the results of the present study, do not confirm this statement.

An alternative approach suggested by Xu et al. [31], focuses on a heat transfer in the immediate vicinity of the symmetry axis piercing the apex. The change of sign of the tangential gradient of the temperature near the apex and, hence, the corresponding transition between the opposite circulation directions, taking place with a variation of the relative substrate–liquid thermal conductivity, has been identified by the authors under different conditions as compared to the results of [30]. Transition points at various contact angles obtained in [31] are much closer to the results of subsequent numerical calculations and are in agreement with the corresponding experimental data.

The above studies [29–31] assumed a monotonic temperature profile along the droplet surface, and, hence, a single-vortex fluid flow. However, such an assumption only partially explains the phenomenon. The thermal conduction processes throughout the droplet can generally result in a nonmonotonic spatial dependence of the surface temperature and in more complicated convection patterns inside a drop. In particular, either a single vortex or several vortices are formed in the droplet depending on the thermal conductivity of the substrate [32]. With varying the relative substrate–liquid thermal conductivity, transitions between regimes with different numbers of vortices and/or circulation directions take place. This has been described recently in more detail by Zhang et al. in [33], where the authors presented the “phase diagram” characterizing, for a fixed substrate thickness $h_S = 0.1 R$, the distribution of surface temperature in the k_R – θ plane, where the notations k_R , R and θ are explained in Table 1.

The three regions in the k_R – θ plane have been demonstrated in [33]. In region I the surface temperature monotonically increases from the center to the edge of the droplet. In region III the surface temperature decreases monotonically from the center to the edge of the droplet. Finally, in region II the temperature exhibits a non-monotonic spatial dependence along the droplet surface. Naturally, regions I and III correspond to single-vortex states with opposite circulations of the fluid flows. In the present work we focus on the substructure of the region II. Specifying the number of the temperature extrema along the droplet surface, we find the subregions, which correspond to two or three vortices inside the droplet. We also identify the dependence of the borders between the subregions on the substrate thickness. The results obtained demonstrate that the vortex state structure in evaporating droplets of capillary size can be prepared in a controlled manner by selecting substrates with appropriate thermal conductivity and thickness.

2. Basic equations and methods

The basic hydrodynamic equations inside the drop are the Navier–Stokes equations and the continuity equation for the incompressible fluid

$$\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} + \frac{1}{\rho} \text{grad } p = \nu \Delta \mathbf{v}, \quad (1)$$

$$\text{div } \mathbf{v} = 0. \quad (2)$$

Here $\Delta = \partial^2/\partial r^2 + \partial/r\partial r + \partial^2/\partial z^2$, $\nu = \eta/\rho$ is kinematic viscosity,

The calculation of thermal conduction inside the droplet and the substrate is carried out without taking into account the convective heat transfer, which is justified when the Péclet number $Pe = \bar{u}R/\kappa$ is much smaller than unity. The following equation is solved:

$$\frac{\partial T}{\partial t} = \kappa \Delta T, \quad (3)$$

where $\kappa = k/(\rho c_p)$ is thermal diffusivity. The boundary conditions take the form $\partial T/\partial r = 0$ for $r = 0$; $\partial T/\partial n = -Q_0(r)/k$ at the drop surface. Here $Q_0(r) = Lj_s(r)$ is the rate of heat loss per unit area of the upper free surface, \mathbf{n} is a normal vector to the drop surface, j_s is the local evaporation rate determined by

$$j_s(r) = J_0(\theta)(1 - r^2/R^2)^{-\lambda(\theta)}, \quad (4)$$

where $\lambda(\theta) = 1/2 - \theta/\pi$, fitting expression for $J_0(\theta)$ is taken from [35], other notations are explained in Table 1.

The relation (4) fits well with the analytical solution for a stationary spatial distribution of the vapor concentration for a drop with the shape of a spherical cap (see [2,35]). The problem is mathematically equivalent to that solved by Lebedev [36], who obtained the electrostatic potential of a charged conductor having the shape defined by two intersecting spheres. Nonstationary effects in vapor concentration and effects resulting from deviations of droplet shape from spherical cap are considered in detail in [34]. Nonstationary effects in vapor concentration may only effectively result in a change of the constant $J_0(\theta)$ in (4) and become small for $t \gg R^2/D$, where t is the duration of the evaporation process. Effects resulting from deviations of a droplet shape from spherical cap are very small provided that the Bond number $B_o = \rho ghR/(2\sigma \sin \theta)$ is much smaller than unity, which is true for the droplets considered. Evaporation rate measurements for such droplets agree well with the calculations based on the relation (4) [2,34,35].

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