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ORIGINAL ARTICLE

Large-Scale Liquid Motion in Free Thermocapillary Films

Hendrik C. Kuhlmann

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Abstract Thermocapillary flows in thin films are considered which are differentially heated from lateral side walls. The competition between two different types of motion is addressed. One type is the so-called return flow in which a return flow opposes the thermocapillary free-surface flow to preserve mass conservation. The other type of motion is a large-scale flow which arises as a plug flow in which the velocity is independent of the coordinate perpendicular to the film. We provide physical arguments, based on the minimization of the surface energy, for the preference of the large scale flow over the return flow as the film thickness decreases. The large scale motion arises as a cellular flow with alternating vorticity perpendicular to the film surface. The direction of rotation of these vortices is not determined when the film is adiabatic and of constant thickness. If, however, the film thickness varies perpendicular to the applied temperature gradient the flow direction is dictated by the minimization of the surface energy. Our predictions are consistent with independent experiments and numerical simulations.

Keywords Thin film · Marangoni effect · Thermocapillary convection

Background

Investigations of thin thermocapillary liquid films with two free surfaces (Watanabe and Ueno 2009; Ueno and Torii 2010; Yamamoto et al. 2013) have revealed a large-scale cellular fluid motion (Fig. 1b). The liquid films have been heated non-uniformly from a bounding solid frame which was either circular or rectangular. Similar as in soap films the velocity field of the large-scale fluid motion was found to be independent of the coordinate z normal to the plane defined by the thin film and it arises as two vortices occupying the whole liquid film laterally (x and y directions).

To date, the only attempt of an explanation for the large-scale motion is due to Yamamoto et al. (2013) who aimed at understanding a demonstration experiment carried out onboard of the ISS by astronaut Pettit (2003) under conditions of weightlessness. Yamamoto et al. (2013) numerically simulated the flow generated by suddenly placing a heat source at one point of a circular solid frame holding the liquid film. The heat source induces strong local temperature variations along the two liquid–gas interfaces of the film which, via the thermocapillary effect, drive a pair of wall-attached counter-rotating vortices. Far away from the wall they find a large-scale flow. The authors argue that the return flow of the two stacked wall-attached vortices which is directed towards the solid wall in the symmetry plane of the problem cannot transport the same amount of liquid as the free-surface flow directed away from the hot wall if the film has either a concave or a convex shape. Thus a net flow away from (towards) the stacked vortices and towards (away from) the center of the film should arise for a concave (convex) shape of the liquid–gas interfaces of the film. While this is a description of the numerically computed flow field, it is not a physical reason for the existence and the direction of a large-scale motion. The argument is essentially *two-dimensional* and purely mechanical and it relies on the static free-surface deformation in wall-normal direction, i.e. in the direction parallel to the surface-temperature gradient. However, for an incompressible *two-dimensional* flow an imbalance between the volume flux in the surface

H. C. Kuhlmann (✉)
Vienna University of Technology, Institute of Fluid Mechanics and Heat Transfer, Resselgasse 3, 1040 Vienna, Austria
e-mail: h.kuhlmann@tuwien.ac.at

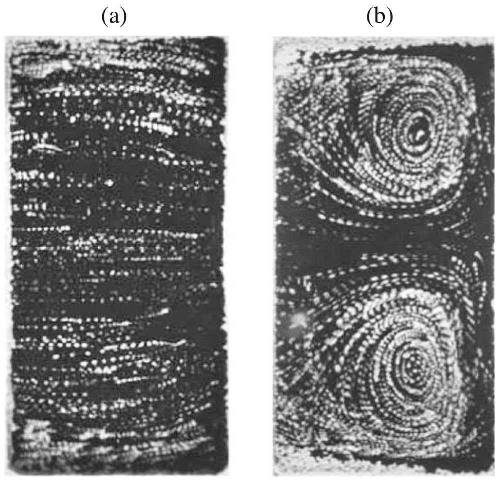

Fig. 1 Streakline patterns for the return flow **a** and for the large-scale flow **b** in a rectangular frame of 2×4 mm as viewed perpendicular to the thin film. The thickness of the frames in (**a**) and (**b**) is 0.6 mm and 0.2 mm, respectively. The right side is heated while left side is cooled. Reproduced from Figs. 5a and 6a of Ueno and Torii (2010) courtesy I. Ueno

flow and the one in the return flow is impossible. Another shortcoming of the explanation is the absence the thermal budget.

Here, we propose an alternative explanation for the existence and the direction of the large-scale motion. The explanation applies to the work of Yamamoto et al. (2013) as well as to the work of Watanabe et al. (2009) and Ueno et al. (2010) who investigated the flow in a film bounded by a rectangular frame heated from one side of the frame and cooled from the opposite side. Our explanation is based on the surface energy, the convective heat transport, and a static variation of the film thickness perpendicular to the temperature gradient. It does not rely on flow-induced interface deformations.

Model System

We consider a liquid film bounded by a rectangular frame of width L heated differentially such that $T(x = -L/2) = T_0 - \Delta T/2$ (cold wall) and $T(x = L/2) = T_0 + \Delta T/2$ (hot wall) under zero-gravity conditions (Fig. 2), similar as in Ueno et al. (2010). We assume that the other two facing sides of the frame at $y = \pm\Lambda L/2$ are adiabatic, where $\Lambda = O(1)$ is the cross-stream aspect ratio. The gas is also assumed to be adiabatic. Let us first consider a film of constant thickness h equal to the thickness of the frame. The problem is essentially two-dimensional in x and z, apart from end effects caused by the adiabatic side walls due to the no-slip conditions. If viscous effects in the gas are neglected the thermocapillary stresses are balanced by viscous stresses in the liquid which, due to symmetry, drive a symmetric return flow in the liquid with a parabolic velocity profile (Figs. 1a and 2a) which does not depend on the transverse coordinate y, except near the adiabatic side walls. Such flows have been observed by Ueno et al. (2010) and computed by Limsukhawat et al. (2013).

The system investigated by Yamamoto et al. (2013) is somewhat different, because the flow is transient and the geometry in conjunction with a point heat source does not allow for a strict symmetry. But the idealization of the problem considered by Yamamoto et al. (2013) would be a point heat source at the apex of a wedge-shaped frame which is free-slip and adiabatic. In this idealization the return flow would be axisymmetric with respect the vertical axis through the point at which the heat source is located, giving rise to an axisymmetric return flow.

For small Marangoni numbers, measuring the strength of the thermocapillary driving force, the basic flow with the same symmetries as the boundary conditions will be stable. Upon parameter variation deviations from the symmetric flow typically arise via symmetry-breaking instabilities. The classical example is the onset of hydrothermal waves (Smith and Davis 1983).

Origin and Direction of the Large-Scale Motion

Thermocapillary flow is caused by the tendency of the system to minimize the total surface energy $E = \int_A \sigma(T)\mathrm{d}A$, where $\sigma(T)$ is the temperature-dependent surface tension and A the total area of the interface. In linear approximation $\sigma = \sigma_0 - \gamma(T - T_0)$, thus $E = \sigma_0 A - \gamma \int_A (T - T_0)\mathrm{d}A$. For the normal thermocapillary effect ($\gamma > 0$) E is minimized by maximizing the mean temperature of the free surface under the constraints given by the Navier–Stokes and energy equations together with the boundary conditions. The surface forces $\nabla\sigma$ driving the system to a dynamic equilibrium must be balanced by viscous forces which is expressed by the tangential stress balance on the interface

$$\mathbf{t} \cdot \mathsf{S} \cdot \mathbf{n} = \mathbf{t} \cdot \mathsf{S}_{\mathrm{gas}} \cdot \mathbf{n} + \mathbf{t} \cdot \nabla\sigma, \tag{1}$$

where $\mathsf{S} = \mu\left(\partial_i u_j + \partial_j u_i\right)$ is the stress tensor in the liquid with dynamic viscosity μ, $\mathsf{S}_{\mathrm{gas}}$ the one in the gas and $\mathbf{n}$ and $\mathbf{t}$ the unit vectors normal and tangential to the free surface, respectively.

Assume, we have a two-dimensional return flow with zero mean $\int \mathbf{u}\mathrm{d}z = 0$. Usually, the dynamic viscosity of the liquid is much larger than that of the gas, $\mu \gg \mu_{\mathrm{gas}}$, such that for sufficiently thick liquid layers $\|\mathsf{S}\| \gg \|\mathsf{S}_{\mathrm{gas}}\|$. Neglecting $\mathsf{S}_{\mathrm{gas}}$ the tangential stress condition (1) leads to the velocity estimate for the return flow $U \sim \gamma\Delta T h/(\mu L)$. Therefore, the horizontal convective heat transport is governed by $U\partial_x T \sim \gamma\Delta T^2 h/(\mu L^2)$, while the diffusive heat transport across the film scales like $\kappa\Delta T/h^2$, where κ is the

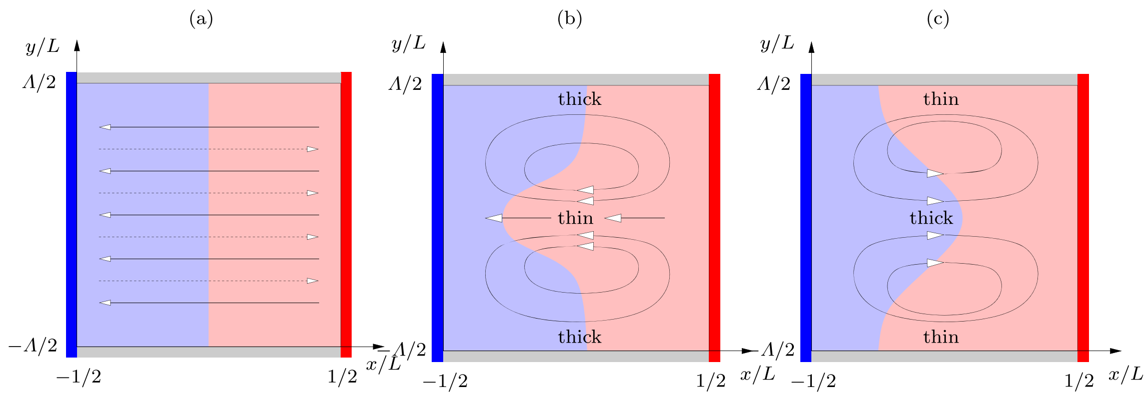

Fig. 2 Sketch of characteristic streamlines and the temperature distribution in thin thermocapillary films. **a** Return flow in a film of constant thickness. **b** Large-scale flow in an underfilled film which is thinner in the center than at the periphery. **c** Large-scale flow in an overfilled film which is thicker in the center than at its periphery. In cases (**b**) and (**c**) the mean temperture of the film is higher than in case (**a**)

thermal diffusivity of the liquid. The ratio of the horizontal-to-vertical heat-transport terms is $\sim$ Ma$(h/L)^3$ with the Marangoni number Ma $= \gamma \Delta T L/(\kappa\mu)$. For a thick film with $h = O(L)$ and high Marangoni number the horizontal convective transport dominates the vertical one. As a result the temperature at the surface of the liquid will be higher than the one in the bulk. This effect reduces the surface energy and it is the typical situation in, e.g., thermocapillary liquid bridges when $h/L = O(1)$. If, however, the film thickness becomes very small ($h/L \to 0$) the horizontal convective heat transport diminishes, because the velocity diminishes ($U \sim h/L \to 0$), and the temperature distribution will be dominated by thermal conduction everywhere in the liquid. Therefore, the return flow cannot lead to a reduction of the surface energy for very thin films.

In contrast, a large-scale flow can transport heat much more efficiently in thin films, because of the mean flow $\int \mathbf{u} dz \neq 0$. Moreover, the viscous stresses in the liquid are much smaller than for the return flow and the thermocapillary stress in (1) will mainly be balanced by the viscous stress S_{gas} in the gas phase. For this mode of convection the film thickness h does not enter the stress balance anymore, and the characteristic velocity of the large-scale motion will not vanish in the limit $h \to 0$ if thermocapillary stresses exist which can drive the large-scale motion. For example, for a Marangoni number as small as Ma $= 2$ Watanabe et al. (Watanabe and Ueno 2009) found a significant large-scale motion in thin films, much larger than the surface velocity in the return flow in thicker films at the same Marangoni number.

The large-scale motion observed the experiments in form of a pair of large vortices breaks the translational symmetry in y (Watanabe and Ueno 2009; Ueno and Torii 2010) (Fig. 2). On the other hand, the temperature gradient is essentially unidirectional during the growth phase when the large-scale motion is yet a small disturbance to the nearly quiescent flow with a nearly conducting temperature field (Fig. 2a). Therefore, the convective transport of temperature in positive and negative x directions will be balanced and the large-scale motion cannot reduce the surface energy (cannot increase the mean surface temperature) in a film of constant thickness.

To see how the large-scale flow can reduce the total surface energy we consider a film whose thickness varies in y direction, perpendicular to the applied temperature gradient. Such static interface deformation can arise due to over- or underfilling of the film whose contact lines are pinned at the edges of the frame. For adiabatic boundary conditions this surface deformation would not alter the conductive temperature profile. However, due to mass conservation, the absolute value of the velocity of the large-scale flow must be higher in the thin-film region than in the thick-film region, provided the horizontal extent in which the flow is directed in positive and negative directions is approximately the same. This is the case in the experiments of Ueno et al. (2010) (Fig. 1b). In this situation, the large-scale flow is more efficient in transporting heat in the thin-film region than in the thick-film region and leads to an imbalance of the horizontal energy transport. Two directions of circulation are possible.

1. **Large-scale flow is parallel to the surface-tension gradient where the film is thin:** In this case the convective transport of hot fluid in negative x direction due to the large-scale motion is strong in the thin-film region because of the relatively high flow velocity. This effect leads to a strong increase of the surface temperature and thus to a decrease of the total surface energy. In the other region where the film is thick and the flow is directed opposite to the surface-tension gradient the convective transport of cold fluid in positive x direction is weaker, because the velocity is smaller there.

Therefore, the transport of cold fluid in the thick-film region cannot compensate the decrease of the surface energy caused by the flow in the thin-film region. The large-scale motion will thus lead to a reduction of the total surface energy by increasing the average surface temperature. The flow directions for the two cases of concave (underfilling) and convex (overfilling) interfaces are illustrated schematically in Figs. 2b and c, respectively.

2. **Large-scale flow is opposite to the surface-tension gradient where the film is thin:** In this case the transport of cold fluid will be strong where the film is thin, because the absolute velocity of the large-scale motion is high. Moreover, the transport of hot fluid in the region where the film is thick will be weak due to the small flow velocity. In this case the average surface temperature will be reduced by the large scale motion which represents an increase of the surface energy. Therefore, a large scale motion *antiparallel* to the surface-tension gradient where the film is thin would be energetically less favorable than a large-scale motion *parallel* to the surface-tension gradient where the film is thin, and even energetically less favorable than the absence of a large-scale motion. Therefore, such a motion cannot arise.

Summary and Conclusion

A large-scale motion in which the velocity does not vary perpendicular to the film can be driven by small thermocapillary forces which are mainly balanced by the weak viscous stresses in the gas phase. Under the symmetries described the convective transport of heat by the large scale motion is balanced if the film thickness is constant. If, however, the film thickness is non-uniform in a direction perpendicular to the applied temperature gradient, the velocity depends on the film thickness and the convective heat transport is not balanced anymore. As a result the surface energy will change. The convective heat transport leads to a reduction of the surface energy if the large-scale flow is parallel to the surface-tension gradient in the region where the film is thinnest, because this flow orientation leads to a reduction of the total surface energy. Since the reduction of the surface energy by the large-scale flow is larger than for the return flow for $h \rightarrow 0$, the large-scale flow is favored over the return flow in sufficiently thin films.

The interpretation given is in qualitative agreement with the experiments of Watanabe et al. (2009) and Ueno et al. (2010). In the numerical simulations of Yamamoto et al. (2013) who neglected the stress in the gas phase localized thermocapillary stresses in the liquid may have caused the large-scale flow. The interpretation is also consistent with the results of Limsukhawat et al. (2013) who did not find any large-scale flow in their three-dimensional simulations of free liquid films with constant thickness.

In contrast to the argument of Yamamoto et al. (2013) which is based on pure mechanical quantities, i.e. the flow field and the surface deformation *parallel* to the temperature gradient, our argument is based on the total surface energy, the convective transport of heat, and the surface deformation *perpendicular* to the temperature gradient. It would be interesting to verify the present predictions by, e.g., measurements of the mean temperature of the film as well as the film-thickness distribution for different steady flow states as functions of filling factor (concave or convex interface shape).

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