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Regular hexagons and stripes on Marangoni and Rayleigh–Taylor unstable volatile thin films

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ABSTRACT

Thin volatile Newtonian liquid films with a free surface on a cooled horizontal substrate are studied theoretically and numerically. We show that if the fluid is initially in equilibrium with its own vapor in the gas phase, regular surface deformation patterns in the form of long-wave hexagons or stripes having a well defined lateral length scale are observed, depending on the instability mechanism. This is different to the case without evaporation where rupture or coarsening to larger and larger spatially disordered patterns is seen in the long time limit. We propose to use such a system to create a regular structuring of the film's surface. Heat production by latent heat and the influence of a temperature dependent surface tension (Marangoni effect) are included as well. Special emphasis is laid on the so-called anomalous Marangoni effect. In this case a parameter region where stripes should occur already at threshold is found by means of a systematic weakly non-linear analysis.

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1. Introduction

Regular self-organized patterns of fluid motion are known since the experiments of Michael Faraday in 1831 [1]. Faraday excited the fluid layer mechanically by harmonic vibrations and observed regular squares along its free surface. Seventy years later, Henri Bénard performed his famous convection experiments [2], where he obtained liquid motion organized in the form of regular hexagons. The fluid rises in the center of each hexagon and descends along its six sidewalls. This is the standard pattern obtained in liquids and therefore called ℓ-hexagon(s). Hexagons break the up–down symmetry of the patterns. Mirroring at the midplane, ℓ-hexagons turn into g-hexagons commonly found in gases. Here, the fluid descends in the center of the hexagons.

In his seminal paper in 1960, Enok Palm [3] was the first who used these symmetry arguments to show that ℓ-hexagons are the preferred patterns in convection, if the up–down symmetry is broken by a temperature dependent viscosity. In a contemporary notation, one would write down an amplitude equation in the form (see Section 3 of this contribution)

$$\dot{\Psi} = \varepsilon\,\Psi + A\,\Psi^2 - \Psi^3$$

where Ψ represents the pattern amplitude, ϵ is the distance to threshold and A stands for the symmetry breaking effects [4]. Besides temperature dependent viscosity, symmetry breaking may originate from a temperature dependent thermal conductivity or a nonlinear temperature dependence of the density of the liquid [5]. All these effects are summarized under the notion "Non-Boussinesqian" and give rise to a non-vanishing A. The change of sign of A marks the codimension-one hyperplane in parameter space where ℓ-hexagons $A > 0$ turn into g-hexagons ($A < 0$). As an example we mention the Bénard–Marangoni instability where A depends on the Prandtl number Pr and $A = 0$ is achieved for $\mathrm{Pr} \approx 0.23$, as shown in [6].

The above mentioned patterns have been observed in layers having the depth of some millimeters or more. The structures that emerge at instability possess a finite horizontal wave vector and are of the same size than the layer depth. In the present paper, we wish to study a thin liquid film showing a long wave instability. Then the lateral length scale of the unstable patterns is much larger than the depth. Motivated by the shallow water equations for inviscid fluids [7], a long-wave equation for viscous thin layers was derived systematically from the Navier–Stokes equations [8] by expansion with respect to a small parameter. In very thin viscous films, inertia can be neglected compared to friction and a closed equation for the location of the liquid's free surface can be derived (for reviews see [9] and the quite recent paper [10]).

The dynamics of a free surface of a viscous liquid is also of high interest from the viewpoint of self-organized pattern formation [4,11]. Surface deformation patterns of thin liquid films on a solid support were studied during the past decade in numerous experimental and theoretical contributions (see [9,10,12–16] and references therein). The typical scenario is the following: due to an instability mechanism (thermal, solutal, gravitational, etc.) the flat surface is unstable and the fluid begins to move. Thereby surface patterns are formed and show coarsening. Rather irregular cells

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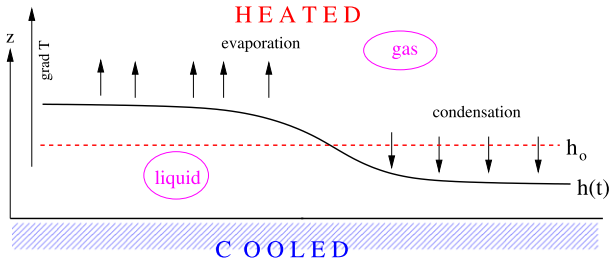

Fig. 1. The flat surface of a fluid on a horizontal plate cooled from below is stable at position h_0 where liquid and gas layers are in thermodynamic equilibrium. If the surface is at position $h > h_0$ the liquid would evaporate, if it would be below h_0 it would condense until h_0 is reached.

evolve first in the form of drops, holes or mazes. Although already large scaled with respect to the film's depth, they increase their size continuously in course of time and end on a much larger scale than the initial ones. Another possibility is that rupture occurs during the temporal evolution.

For technological applications the creation and control of ordered and regular micro structures come more and more into the focus of interest. Surface patterns with prescribed length scales and geometries can be produced in several ways, one simple possibility is to use a structured substrate [17–19]. Here, we propose another method: the self-organized pattern growth due to an instability mechanism of the initially flat film. There are several mechanisms that may destabilize a flat surface and that allow to control the growth of surface patterns. Flat ultra-thin films may become unstable by van der Waals forces between surface and substrate [13–15]. Thicker films can be destabilized by inhomogeneous tangential surface tensions (Marangoni effect), which in turn are often caused by lateral gradients of temperature and/or, in mixtures, of concentration [16,20,21].

In a previous publication [22] we showed that the inclusion of evaporation and condensation at the free surface may change pattern formation qualitatively. Rupture can be avoided for large enough evaporation rates and, even more important, due to the modified character of the instability, coarsening does no longer occur in the long time limit. Instead we found very regular cell structures in the form of hexagons, known from their morphology from small scale convection in thicker fluid layers [23–25].

If a temperature gradient vertically to the surface is applied, evaporation can stabilize or destabilize the flat surface, depending on the sign of the gradient. If the film is located on a heated support, evaporation acts destabilizing: thinner regions have their surface closer to the hotter substrate, evaporate stronger, getting even more thinner and finally rupture. Now assume that the fluid is heated from the gas side (or cooled from the support). If the partial pressure of the vapor in the gas layer is equal to the saturation pressure belonging to the surface temperature of the initial flat film, then a small depression of the surface into colder regions leads to local condensation, a small elevation into hotter regions causes evaporation, see Fig. 1. Thus, evaporation acts stabilizing.

Throughout the present paper, evaporation is assumed to be stabilizing. As a consequence, another destabilizing mechanism is necessary to obtain self-organized structures. The perhaps most simple method for destabilization is to put the film upside down, i.e. to position it under a flat horizontal plate. Then gravity acts against the stabilizing surface tension and inhomogeneous surface patterns can result [26]. This is called Rayleigh–Taylor instability (RTI) and was examined in [22].

Here we shall first concentrate on the Rayleigh–Taylor instability. We assume a vertical heat gradient applied from outside. If the fluid is heated from the gas side, this would in addition stabilize the flat film by the Marangoni effect which was not included in [22]. Neglecting evaporation, it was shown in [26] that RTI may occur if the temperature gradient is not too large so that gravity may overcome the Marangoni effect. Since we are interested in the emergence and saturation of regular surface patterns, we perform a weakly non-linear analysis close to threshold. We show that for RTI combined with stabilizing evaporation, latent heat production and surface tension, the typical 2D-patterns (hexagons, stripes, squares) bifurcate subcritically. Thus the validity of a third order amplitude equation is rather restricted. Finally we shall also consider the standard configuration where gravity is stabilizing, i.e. the light gas layer is located on top of the fluid. To obtain an unstable situation of the flat film for this case, a fluid showing the anomalous Marangoni effect is used. Here, surface tension increases with temperature, a situation encountered in certain aqueous alcohol solutions [27]. Strong enough heating from the gas side may cause a Marangoni instability of the flat and motionless layer. A weakly non-linear analysis shows now that the cubic coefficients of the amplitude equations have a stabilizing (negative) sign and a stability diagram in parameter space can be computed.

2. Thin film equation and evaporation

2.1. Thin film equation

Up to now, most of the theoretical work is based on an interface equation, often called thin film equation, describing the location $z = h(x, y, t)$ of the free surface of the liquid [9,28,29]. This equation can be systematically derived from the Navier–Stokes equation using the lubrication approximation. For the case of an externally applied temperature gradient vertically to the free surface, it reads:

$$\partial_t h = -\nabla\left[-\frac{h^2}{2}M\frac{dT_l(h)}{dh}\nabla h + \frac{1}{3}h^3C^{-1}\nabla^2\nabla h - \frac{1}{3}Gh^3\nabla h\right], \quad (1)$$

where $T_l(h)$ is the temperature at the free surface and ∇ and ∇^2 denote the horizontal gradient and Laplace operators. In (1) a scaling for lengths and time according to

$$(\tilde{x}, \tilde{y}, \tilde{z}, \tilde{h}) = (x, y, z, h)\cdot d, \qquad \tilde{t} = t\cdot\tau, \quad \tau = d^2/\kappa$$

is used (variables with a tilde bear dimensions). The thickness of the (flat) fluid layer in rest is hd, τ denotes the vertical diffusion time of heat, and κ is the thermal diffusivity of the liquid. Viscosity, density, and thermal conductivity of the fluid are denoted with ν, ρ, and λ_1. The width of the gas layer is d_g, λ_2 is its thermal conductivity and g the gravitational acceleration.

Scaling results into the dimensionless numbers M (Marangoni number), G (gravity number), C (Crispation number) defined as

$$M = \frac{\gamma_T \Delta T d}{\rho\nu\kappa}, \qquad G = \frac{gd^3}{\nu\kappa}, \qquad C = \frac{\kappa\nu\rho}{\gamma_0 d}. \quad (2)$$

Here, surface tension is assumed to be a linear function of surface temperature T_l.

$$\Gamma = \gamma_0 - \gamma_T(\tilde{T}_l - \tilde{T}_0), \quad \gamma_0, \gamma_T > 0. \quad (3)$$

Temperatures will be scaled to the externally applied temperature difference $\Delta T = \tilde{T}_0 - \tilde{T}_1$, $\tilde{T}_1$ being the temperature of the boundary at $z = d + d_g$, T_0 that of the solid support ($z = 0$). The mean control parameter is M which is directly linked to the externally applied temperature difference. Note that M is negative if the system is cooled from the liquid side, G is negative if gravity acts in direction to the gas side (RTI).

Previous work shows that in the case of a surface-driven thermal instability, the film thickness may reach very small values

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