The oscillatory longwave Marangoni convection in a thin film heated from below

Anna Samoilova¹ · Alexander Nepomnyashchy²

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Abstract

A novel type of Marangoni convection was predicted theoretically a decade ago. The thin liquid film atop a substrate of low thermal conductivity was considered. In the case of heating from below, the Marangoni convection emerges not only in a conventional stationary regime, but also as oscillatory flows. Specifically, the oscillatory Marangoni convection emerges if (1) the heat flux from the free surface is small, and (2) the large-scale deformation of the free surface is allowed. During the past decade, this novel Marangoni convection was detected and investigated in several other theoretical works. The review discusses the recent achievements in studying the oscillatory Marangoni convection in a thin film heated from below. The guiding data for observation of the oscillatory regime are also provided.

Article Highlights

Several recent theoretical works reveal the oscillatory Marangoni instability in thin liquid film atop a heated substrate. The oscillatory regime of convection can be observed in ultrathin films (~ 0.01 mm for water) or under microgravity conditions. The surface tension should allow interface deformations, and the heat transfer from the liquid to the ambient gas should be small.

Keywords Marangoni effect · Long-wave instability · Thin film · Deformable interface · Wave patterns · Feedback control · Insoluble surfactant

Mathematics Subject Classification 76A20 · 76E30 · 76E06 · 76E17 · 76D55

Nomenclature

λ	thermal conductivity	α	surface tension (α_0 is the one at the reference temperature)
χ	thermal diffusivity	α_T	temperature coefficient of the surface tension
ρ	density	α_Γ	concentration coefficient of the surface tension
ν	kinematic viscosity		
g	gravitational acceleration		
Q	vertical component of the heat flux at the substrate	H	mean layer thickness
		L	typical horizontal length scale
q	heat transfer coefficient from the free surface	$\varepsilon = H/L$	lubrication parameter
		z	vertical Cartesian coordinate

✉ Anna Samoilova, annsomeoil@gmail.com | ¹Department of Theoretical Physics, Perm State University, Perm, Russia 614990. ²Department of Mathematics, Technion–Israel Institute of Technology, 32000 Haifa, Israel.

x, y	horizontal coordinates (rescaled ones are X, Y)	$Le = D_0/\chi$	Lewis number		
t	time (rescaled one is τ)	$N = \alpha_T \Gamma_0 H/\rho\nu\chi$	elasticity number		
$\mathbf{n}$	normal unit vector	$\lambda_g = \tilde{\lambda}_g/\lambda$	dimensionless gas heat conductivity (rescaled one is Λ)		
$\mathbf{u}$	horizontal velocity vector (rescaled one is $\mathbf{U}$)	$\chi_g = \tilde{\chi}_g/\chi$	dimensionless gas thermal diffusivity		
w	vertical velocity component (rescaled one is W)	Abbreviations TR traveling rolls SR standing rolls TRa traveling rectangles AR alternating rolls SSq standing squares			
T	difference between the temperatures of the liquid and the gas				
p	pressure				
h	surface location				
Θ	deviation of the temperature T from that in the conductive state				
$f = \Theta - h$	temperature on the free surface	1 Introduction Interfacial convection in a thin liquid film atop a heated substrate is a classical non-equilibrium phenomenon that has been studied for over a century [1]. Recently, it has been placed in the spotlight again due to development of microfluidics [2, 3]. In many technological processes in microelectronics and bioengineering, a thin liquid film placed on a solid heated substrate is present in which undesirable convective instability can occur. On the contrary, mixing caused by convection in a thin film can be beneficial for heat and mass transfer, and convective structures on the surface of a thin film can be used to create spatially periodic microstructures. There are two main mechanisms of convective instability in a heated liquid layer with a free surface: the buoyancy and the dependence of the surface tension on the temperature (thermocapillary effect). For sufficiently thin layers or under microgravity conditions, the thermocapillary (Marangoni) effect predominates over the buoyancy effect. Two basic types of Marangoni instability have been revealed. Pearson discovered an instability mode that does not need surface deformations [4]. In the case of boundaries of finite heat conductivity, that is a short-wave instability with a finite wavelength of the critical perturbations. But in the case of poorly heat conducting boundaries, the critical wave number is asymptotically small as $k_c \sim \text{Bi}^{1/4}$, where Bi is a small Biot number (dimensionless heat flux from the free surface). A specific feature of a longwave instability is the existence of a slowly evolving (active) variable that governs the dynamics [5]. In the case of poorly conducting boundaries, the active variable is the temperature [6, 7]. At moderate Galileo number, another type of longwave Marangoni instability (with $k_c = 0$ has been found [8, 9], where the surface deformation is the active variable [10, 11]. In both cases, the instability leads to the monotonic growth of large-scale perturbations on the liquid surface.			
Σ	growth rate (rescaled one is σ)				
K	wavenumber (rescaled one is k)				
K_X, K_Y	rescaled wave numbers in X and Y directions				
ω	frequency of the neutral perturbations				
ψ	perturbation of the stream function				
θ	perturbation of temperature T				
ξ	perturbation of the surface location h				
δ	small parameter of supercriticality				
A_j	amplitude functions of perturbations				
ϕ	angle between waves				
$K_{0,1}$	Landau coefficients				
K_f	control gain (rescaled one is κ)				
Subscripts					
g	related to the gas phase				
c	related to the critical perturbations				
1, 2, 3 ...	related to components of a series expansion				
τ	tangent to the surface				
Superscripts					
mon	related to the monotonic instability				
osc	related to the oscillatory instability				
$\sim$	denotes dimensional quantities. The same quantities without ($\sim$) are nondimensional				
Dimensionless groups					
$\text{Bi} = qH/\lambda$	Biot number (rescaled one is β)				
$\text{Pr} = \nu/\chi$	Prandtl number				
$\text{Ga} = gH^3/\nu\chi$	Galileo number				
$\text{Ca} = \alpha_0 H/\rho\nu\chi$	capillary number (rescaled one is C)				
$\text{Ma} = \alpha_T QH^2/\lambda\rho\nu\chi$	Marangoni number				

However, the Marangoni instability is not always monotonic. The oscillatory surface-tension-driven convection caused by the dependence of the surface tension on the solute concentration (solutocapillary effect) was first predicted [12] and observed [13] in the case of the mass transfer through the interface between two fluids. In the case of the heat transfer, oscillations were described starting with early observations of Linde and collaborators (see, e.g., [14].) However, the nature of those oscillations was elusive.

The theoretical analysis revealed several situations where oscillatory Marangoni instability can appear.

First, if the temperature gradient is directed upward, the oscillatory instability can appear due to a special kind of mixing of two otherwise decaying modes, capillary-gravity waves and dilatational interfacial waves [15, 16]. A similar kind of oscillatory instability can develop by heating from above due to mixing of internal waves and dilatational waves [17]. For more details, see [18].

Typically, the appearance of oscillations needs the presence of an additional stabilizing physical factor ("negative feedback") that transforms a monotonic instability into an "overstability", i.e., an oscillatory instability. That factor can be a surface-active agent on the interface [19, 20] or a stabilizing solute concentration gradient in a binary mixture [21–23]. Note that in [24] authors found that in the case of a poorly conducting interface, there are two important long-wave regimes with different asymptotics, namely, $k_c \sim \text{Bi}^{1/4}$ and $k_c \sim \text{Bi}^{1/2}$ caused by the presence of two instability mechanisms, namely thermocapillarity and solutocapillarity. A detailed description of oscillatory instabilities of this kind can be found in [5]. In the recent paper [25], the oscillatory Marangoni convection was detected in a thin film of a viscoelastic liquid. The authors suggested that viscoelasticity of the fluid is responsible for the emergence of oscillatory instability. Let us mention also the oscillatory Marangoni instability in a system of two liquid layers, which is developed due to the hydrodynamic and thermal interaction between the layers (for review, see [26]).

However, neither of instability mechanisms listed above can produce oscillations in a single layer of a pure Newtonian liquid heated from below.

A decade ago, a novel mode of thermohydrodynamic instability in liquid layer was discovered in the paper of Shklyaev and co-authors [27]. Keeping in mind the idea from [24] of coupling two instability mechanisms to obtain Marangoni oscillations, Shklyaev and co-authors detected a novel oscillatory mode of Marangoni instability in thin film heated from below [8, 27]. They considered thin film atop poorly conducting substrate under certain physical conditions. Namely, authors assumed (1) low heat flux from the thin film surface, (2) sufficiently thin film or

microgravity and finite surface tension to allow large-scale free surface deflections. Separately, (1) and (2) assumptions correspond to the Pearson's and the deformational modes, respectively, of stationary Marangoni convection in thin film heated from below. Eventually, coupling of two monotonic longwave modes can produce a novel oscillatory mode. The dynamics of the system is governed by two active variables, temperature and interface deformation.

2 Motivation and contents

The novel oscillatory instability discovered in [27] is important from both fundamental and technological points of view. Unfortunately, that excellent example of scientists' intuition and courage did not receive the attention that it deserved. Neither numerical nor experimental attempts to detect the novel mode have not yet been presented to a wider audience. The goal of this review is to summarize the theoretical results obtained by now and suggest guiding data for observation of that novel instability outside a piece of paper.

In the present review paper, we collect the novel results dispersed over different publications and present the up-to-date picture of new achievements in the field of oscillatory pattern formation.

In Sect. 3 we formulate the physical problem and present its mathematical description. Also, we describe briefly the results of the pioneering works [27, 28].

Extension of the initial one-layer model to the two-layer model (solution of adjoint problem in ambient gas layer) is presented in Sect. 4. In Sect. 5 the extension of the initial result to the finite-wavenumbers is provided by modeling beyond the longwave approximation. Influence of surface-active agents at the free surface on the novel oscillatory mode is presented in Sect. 6. In Sect. 7 the non-linear development of instabilities is investigated by mean of the weakly nonlinear analysis. Applying the feedback control to govern the long-wave Marangoni instability is presented in Sect. 8. Finally, in Sect. 9, we discuss possibility of observation of oscillatory mode in thin film heated from below.

3 Basic concepts

A horizontal liquid layer confined between a gas phase and solid substrate is heated from below, see Fig. 1a. A detailed consideration of the processes inside solid and gas phases is avoided in the following way. The substrate is assumed to be poorly conductive, i.e., the thermal conductivity of the liquid λ is large in comparison with that of

Fig. 1 **a** A schematic of thin liquid film atop a solid substrate. The dashed line represents the locations of the unperturbed interface. The deformable interface is located at $z = h(x, y, t)$. **b** A schematic of thermocapillary mechanism

the substrate, so that the vertical component of the heat flux Q is fixed (see [29]).

The viscous stresses on the liquid–gas interface created by the flow in the gas phase are neglected due to the low viscosity of the gas. The heat transfer on the interface is determined by the Newton's law of cooling,

$$\lambda \frac{\partial T}{\partial n} = -qT. \quad (1)$$

Here and below, T is the difference between the temperatures of the liquid and the gas; $\partial T / \partial n$ is the normal derivative of T , and q is the heat transfer coefficient. The system is subjected to transverse external fields: gravity $\mathbf{g}$ and temperature gradient $-(Q/\lambda)\hat{z}$. It is known that this system has a conductive state of motionless liquid. This state can be broken due to instability resulting in global convective motion. The mean liquid layer thickness H is assumed sufficiently small, so that the influence of bulk effects, such as buoyancy, is negligible and the free surface deformation is important. In this case, the main mechanism of convective instability is the thermocapillary (Marangoni) mechanism, see Fig. 1b. The linear dependence of surface tension on the temperature is assumed: $\alpha = \alpha_0 - \alpha_T T$, where α_0 is the surface tension of the liquid at the reference temperature and $-\alpha_T = d\alpha/dT$ is the temperature coefficient of surface tension.

The thermal diffusivity, density, and the kinematic viscosity of liquid are χ , ρ and ν , respectively. The Cartesian x - and y -axes are placed in the substrate plane and the z -axis is opposite to the gravity acceleration.

The problem is characterized by the following non-dimensional parameters. The properties of the liquid are characterized by the Prandtl number, $Pr = \nu/\chi$, which is the ratio of kinematic viscosity to thermal diffusivity. The

non-dimensional parameter $Ga = gH^3/\nu\chi$ named Galileo number represents a ratio between gravity and dissipative factors. The heat transfer from the liquid to the ambient gas phase is described by the Biot number, $Bi = qH/\lambda$. The non-dimensional parameter characterizing the surface tension is the capillary number, $Ca = \alpha_0 H/\rho\nu\chi$, and the strength of the thermocapillary stresses is determined by the Marangoni number, $Ma = \alpha_T QH^2/\lambda\rho\nu\chi$.

As mentioned in Sect. 1, the conduction state can be subject to longwave instabilities: the ratio ε of the mean layer thickness H to a typical horizontal length scale L , which is referred to as the "lubrication parameter", is small, and it can be used as the basis of perturbation expansions. The appropriate rescaling of coordinates and velocity components is as follows:

$$X = \varepsilon x, \quad Y = \varepsilon y, \quad Z = z, \quad \tau = \varepsilon^2 t, \quad \mathbf{U} = \frac{\mathbf{u}}{\varepsilon}, \quad W = \frac{w}{\varepsilon^2}$$

Here $\mathbf{u}$ is the horizontal velocity vector and w is the vertical velocity component.

Let us present the full mathematical description of the problem using the rescaled variables [26]. The rescaled non-dimensional Navier–Stokes, energy and continuity are:

$$\frac{\varepsilon^2}{Pr} \left(\frac{\partial \mathbf{U}}{\partial \tau} + \mathbf{U} \cdot \nabla \mathbf{U} + W \frac{\partial \mathbf{U}}{\partial Z} \right) = -\nabla p + \frac{\partial^2 \mathbf{U}}{\partial Z^2} + \varepsilon^2 \nabla^2 \mathbf{U}, \quad (2)$$

$$\frac{\varepsilon^4}{Pr} \left(\frac{\partial W}{\partial \tau} + \mathbf{U} \cdot \nabla W + W \frac{\partial W}{\partial Z} \right) = -\frac{\partial p}{\partial Z} + \varepsilon^4 \nabla^2 W + \varepsilon^2 \frac{\partial^2 W}{\partial Z^2} - Ga, \quad (3)$$

$$\varepsilon^2 \frac{\partial T}{\partial \tau} + \mathbf{V} \cdot \nabla T = \frac{\partial^2 T}{\partial Z^2} + \varepsilon^2 \nabla^2 T, \quad (4)$$

$$\nabla \cdot \mathbf{U} + \frac{\partial W}{\partial Z} = 0. \quad (5)$$

The no-penetration, no-slip boundary condition at the solid boundary, together with the prescribed vertical heat flux, give

$$Z = 0 : \mathbf{U} = W = 0, \quad \frac{\partial T}{\partial Z} = -1. \quad (6)$$

The boundary conditions on the interface are:

$$\begin{aligned} Z = h(X, Y, \tau) : \quad & \frac{\partial h}{\partial \tau} = W - \mathbf{U} \cdot \nabla h, \\ & \varepsilon^2 \nabla h \cdot \nabla T - \frac{\partial T}{\partial Z} = \text{BiT} [1 + \varepsilon^2 (\nabla h)^2]^{1/2}, \\ & \left(\varepsilon^2 \nabla W + \frac{\partial \mathbf{U}}{\partial Z} \right) [1 - \varepsilon^2 (\nabla h)^2] + 2\varepsilon^2 \nabla h \left(\frac{\partial W}{\partial Z} - \nabla \cdot \mathbf{U} \right) \\ & = -\text{Ma} \left(\nabla T + \frac{\partial T}{\partial Z} \nabla h \right) [1 + \varepsilon^2 (\nabla h)^2]^{1/2}, \\ & 2\varepsilon^2 \left[\varepsilon^2 \nabla \cdot \mathbf{U} (\nabla h)^2 + \frac{\partial W}{\partial Z} - \nabla h \cdot \left(\varepsilon^2 \nabla W + \frac{\partial \mathbf{U}}{\partial Z} \right) \right] \\ & - \frac{2}{3} \varepsilon^2 \left(\frac{\partial W}{\partial Z} + \nabla \cdot \mathbf{U} \right) [1 + \varepsilon^2 (\nabla h)^2] \\ & = p [1 + \varepsilon^2 (\nabla h)^2] + \frac{\varepsilon^2 \text{Ca} \nabla^2 h}{[1 + \varepsilon^2 (\nabla h)^2]^{1/2}}. \end{aligned} \quad (7)$$

Hereafter, $\nabla = (\partial/\partial X, \partial/\partial Y, 0)$. The nonlinear terms in the expression for the curvature, which are $O(\varepsilon^4)$, are omitted. The system of equations and boundary conditions (2–7) is supplemented by some initial conditions. Integration of the continuity Eq. (5) over Z from 0 to $h(X, Y, \tau)$, using integration by parts, the boundary condition (6) and kinematic condition from (7), gives

$$\frac{\partial h}{\partial \tau} + \nabla \cdot \int_0^h \mathbf{U} dZ = 0. \quad (8)$$

The key point of the following analysis is the scaling of dimensionless parameters Ga , Ca and Bi . In realistic systems, these parameters vary significantly depending on values of thin film thickness H , physical properties of the liquid and the rate of the heat transfer from the interface.

For 1-mm water layer at room temperature and atmospheric pressure, Galileo number $\text{Ga} \sim 10^5$. It is reasonable that under normal gravitational acceleration $g = 9.8 \text{ m/s}^2$ Galileo number usually is assumed large even for thin films [30]. However, as $\text{Ga} \sim H^3$, for ultrathin layers Galileo number should be assumed finite (for example, for 0.1 mm water layer $\text{Ga} = 70$). The capillary number $\text{Ca} \sim 5 \times 10^5$ for 1 mm water layer, so it is reasonably assumed large. However, this assumption is usually adopted for the

conductive substrate [11, 31], while for the nearly insulated substrate considered here the capillary number is often taken finite [24, 32].

In the case of low heat transfer from the interface to the ambient gas, $\text{Bi} \ll 1$, and the deformation of the surface is negligible due to the action of gravity, the linear stability theory of Pearson predicts the Marangoni instability in the interval of wavenumbers $\varepsilon \sim \text{Bi}^{1/4}$. Application of the longwave asymptotic approach with the scaling $\text{Bi} = O(\varepsilon^4)$ and $\text{Ga} \gg 1$ leads to the Knobloch equation [7] with the temperature being the only active variable. For $\text{Bi} = O(1)$ and $\text{Ga} = O(1)$, the longwave approach leads to an equation containing the deformation of the film as the only active variable [11].

Shklyaev and his collaborators [27, 28] discovered the non-trivial distinguished limit, $\text{Ca} = \varepsilon^{-2} C$, $\text{Bi} = \varepsilon^2 \beta$, $\text{Ga} = O(1)$, where the longwave dynamics is characterized by two active variables, deviation of the temperature from that in the conductive state, $\Theta(X, Y, \tau)$, and the surface deformation, $h(X, Y, \tau)$. They have derived two coupled nonlinear equations. The first equation is the consequence of the mass conservation condition (8):

$$\frac{\partial h}{\partial \tau} = \nabla \cdot \mathbf{j}, \quad \mathbf{j} = \frac{h^3}{3} \nabla P + \text{Ma} \frac{h^2}{2} \nabla f, \quad (9)$$

where $-\mathbf{j}$ is the local flow rate, $P = \text{Ga}h - C\nabla^2 h$ is the pressure and $f = \Theta - h$ is the temperature on the free surface. The second equation,

$$\begin{aligned} h \frac{\partial \Theta}{\partial \tau} = & \nabla \cdot (h \nabla \Theta) - \frac{1}{2} (\nabla h)^2 - \beta f \\ & + \mathbf{j} \cdot \nabla f + \nabla \cdot \left(\frac{h^4}{8} \nabla P + \frac{h^3}{6} \text{Ma} \nabla f \right), \end{aligned} \quad (10)$$

describes the heat transfer in the film. The nonlinear dynamics of longwave perturbations can be fully described by Eqs. (9 and 10) [27].

Note that the right-hand sides of Eqs. (9 and 10) contain only the gradients of the amplitude function Θ (because the Boussinesq approximation is used in (2, 3, 4, 5)), while the amplitude function h itself appears in the equations.

Within amplitude Eqs. (9 and 10), one can investigate the behavior of small disturbances to the conductive state with motionless fluid and flat interface, that corresponds to $h = 1, \Theta = 1$.

Linearizing Eqs. (9 and 10), one obtains the following dispersion relation for the normal perturbations proportional to $\exp(\sigma \tau + ik_X X + ik_Y Y)$

$$\sigma^2 + \sigma \left[\beta + k^2 \left(1 + \frac{G - \text{Ma}}{3} \right) \right] + \frac{Gk^2}{3} (\beta + k^2) - \frac{\text{Ma}k^4(72 + G)}{144} = 0, \quad (11)$$

where $G \equiv Ga + Ck^2$, $k^2 = k_x^2 + k_y^2$. The wavenumber k is connected with the wavenumber K of the original physical problem as

$$K = \varepsilon k. \quad (12)$$

At the stability border for the *monotonic* mode, $\sigma = 0$, one obtains the following neutral curve

$$Ma^{\text{mon}} = \frac{48(\beta + k^2)(Ga + Ck^2)}{k^2(72 + Ga + Ck^2)}. \quad (13)$$

The critical wave number corresponding to the minimum of this neutral curve is

$$(k_c^{\text{mon}})^2 = \frac{\beta GaC + \sqrt{72\beta GaC(Ga + 72 - \beta C)}}{C(72 - \beta C)}. \quad (14)$$

At the stability border for the *oscillatory* mode, $\text{Re}(\sigma) = 0$, $\text{Im}(\sigma) = \omega$ is the frequency of the neutral perturbations. The neutral curve, critical wave number, and frequency of the neutral perturbations are determined by the following expressions

$$Ma^{\text{osc}} = 3 + 3\frac{\beta}{k^2} + Ga + Ck^2, \quad (15)$$

$$(k_c^{\text{osc}})^2 = \sqrt{\frac{3\beta}{C}}, \quad (16)$$

$$\omega = \frac{k^2}{12} \sqrt{(72 + Ga + Ck^2)(Ma^{\text{mon}} - Ma^{\text{osc}})}. \quad (17)$$

Figure 2a illustrates neutral stability curves for the monotonic and oscillatory modes. It is clear from Eq. (17) that the oscillatory mode is present only at $Ma^{\text{mon}}(k) > Ma^{\text{osc}}(k)$ (see Fig. 2a). Even then the oscillatory mode is critical only if the minimum of $Ma^{\text{osc}}(k)$ is located below the minimum of $Ma^{\text{mon}}(k)$. The parameter range where the novel oscillatory mode is critical is shown in Fig. 3a (solid line).

The neutral curve (13) reproduces two conventional asymptotics in the short-wave limit $k \rightarrow \infty$ (i.e., $K \gg \varepsilon \sim \text{Bi}^{1/2}$). The first one is attained for the finite C , and its value $Ma_c^{\text{mon}} = 48$. This value corresponds to the onset of monotonic instability in the layer with a non-deformable surface (Ca is infinitely large) [4]. In the case $C = 0$ the short-wave limit of (13) represents the result $Ma = 48Ga/(Ga + 72)$ for the onset of monotonic mode in a layer with a deformable surface (Ca is finite) [32]. The same critical Marangoni value is attained in the case $\beta = 0$ (conventional scaling $\text{Bi} \sim \varepsilon^4$), in the minimum of the neutral curve at $k_c = 0$.

Fig. 2 Neutral stability curves for the monotonic (solid lines) and the oscillatory (dashed lines) longwave modes in case of **a** a pure liquid and **b** liquid layer covered by insoluble surfactant, $N=0.001$, $Le=0.003$. Lines 1 and 2 correspond to $\beta C=10$ and 100, respectively. Here $Ga=10$. The domain of stability is located below every line

Fig. 3 **a** The domain of oscillatory instability for one-layer model. Solid and dashed lines correspond to results obtained within the longwave approximation (LO) and for arbitrary wavenumbers (SO), respectively. Here $\text{Pr}=1$, $Ca=1000$. **b** The domain of longwave oscillatory mode for two-layer model for $Ca=2000$, $\lambda_g=0.2$

Importantly, the neutral curves Eqs. (13) and (15) reveal the meaning of the scalings for capillary, Biot and Galileo numbers. Namely, the term $Ga + Ck^2$ indicates the comparability of the gravity and the surface tension. Moreover, terms $\beta + k^2$ and $1 + \beta/k^2$ show that the unusual scaling for the Biot number prescribes that the heat flux from the free surface is of the same order as the heat diffusivity.

Remarkably, one can set any finite value of parameter C without loss of generality in (13, 14, 15, 16, 17), as it corresponds to the definition of the small parameter $\varepsilon = \sqrt{C/Ca}$. Thus, the novel oscillatory mode depends on the product $\beta C = \text{Bi}Ca$, not on each of Bi and Ca separately. Therefore, large capillary number and small Biot number does not ensure occurrence of the oscillatory mode. To succeed, one should balance the surface tension and the heat losses from the free surface as $\text{Bi} \sim 1/Ca \sim K^2$.

4 Two-layer model

A potentially weak point of the initial model developed in [27, 28] is the empirical law for heat transfer from the liquid to the gas phase. The aforesaid works are based on the one-layer approach which treats the interface between gas and liquid phases as a free surface. Within this approach hydrodynamics and heat transfer are considered only in the liquid phase, whereas the gas phase is accounted in a phenomenological way by the Newton's law of cooling. This law includes a dimensionless parameter—the Biot number that plays crucial role in the long-wave instability. However, the Biot number (specifically, heat transfer coefficient q , see Eq. (1)) cannot be calculated within the one-layer approach. Besides, in the case of small Bi (low heat conductivity of the interface), large temperature deviations can occur even for small interface deformations. Thus, it is important to consider heat transport in the gas phase when investigating the longwave deformational instability [30, 33]. As we aimed to provide the guidance for the observation, it is essential to discuss the results obtained in the realistic two-layer approach. Because processes inside both phases are considered within such approach, it provides results that are closer to the experimental data [31].

In [34] the authors investigate longwave instability in a thin liquid film using two-layer approximation. The initial fluid dynamics model is modified to include the gas layer atop the thin film. It is assumed that the gas layer has finite thickness $\tilde{H}_g$ (see Fig. 1a) and small thermal conductivity $\tilde{\lambda}_g$ to assure small heat flux through the interface. The liquid density and the dynamic viscosity are assumed large in comparison with those of gas, thereby one can take into account the heat transfer but not the hydrodynamics in the gas phase.

In addition to dimensionless parameters mentioned above: the capillary, Marangoni, Galileo, and Prandtl numbers, the two-layer system is characterized by dimensionless heat conductivity $\lambda_g = \tilde{\lambda}_g/\lambda$ and thermal diffusivity $\chi_g = \tilde{\chi}_g/\chi$.

The initial-boundary value problem of thermocapillary convection should be changed for the two-layer system. Dimensionless equations are supplemented with the Fourier equation for gas temperature T_g

$$\frac{\partial T_g}{\partial t} = \chi_g \nabla^2 T_g. \quad (18)$$

Here we assume that the thermal diffusivity of the gas is large compared to that of liquid, therefore the heat advection by the gas flow can be neglected.

On the interface, phenomenological Newton's law for cooling is replaced by continuity conditions for

temperatures and normal heat fluxes on both sides of the interface:

$$z = h(x, y, t) : T = T_g, \quad \frac{\partial}{\partial n} (T - \lambda_g T_g) = 0 \quad (19)$$

The upper rigid wall above the gas phase now is considered within reach (see Fig. 1a). The thermal conductivity of gas is considered small in comparison with that of the upper rigid wall, and thus the temperature of a gas is fixed at this boundary

$$z = H_g : T_g = 0 \quad (20)$$

Aiming to find the analogue of the new longwave oscillatory mode from Sect. 3, one applies the same scalings $Ca = \varepsilon^{-2}C$, $Ga = O(1)$ but replaces the phenomenological condition of small Biot number by the physical condition of the relatively small thermal conductivity of the gas:

$$\lambda_g = \varepsilon^2 \Lambda. \quad (21)$$

Under the latter assumption, the disturbance of the heat flux from the liquid through the interface is of the same order as the heat flux along the film.

Following the same procedure as in Sect. 3, one attains the same solution of the leading order and consequently the same first amplitude Eq. (9).

Essential distinctions arise in the first order of the expansion in small ε . The boundary condition on the interface is now

$$Z = h : \frac{\partial T_1}{\partial Z} = \nabla \Theta \cdot \nabla h - \frac{\Lambda}{H_g - h} f \quad (22)$$

That results in the modified second amplitude equation

$$h \frac{\partial \Theta}{\partial \tau} = \nabla \cdot (h \nabla \Theta) - \frac{\Lambda}{H_g - h} f + \mathbf{j} \cdot \nabla f + \nabla \cdot \left(\frac{h^4}{8} \nabla P + \frac{h^3}{6} \text{Ma} \nabla f \right) \quad (23)$$

Comparing the evolutionary Eqs. (10) and (23), one can notice that they differ in nonlinear terms of heat losses from the interface. Consequently, the results of linear stability analysis for the one-layer problem should be suitable for the two-layer problem as well. Indeed, the linearized version of Eq. (23) reduces to the linearized version of the one-layer Eq. (10) after denotation $\Lambda/(H_g - 1) \rightarrow \beta$. Thus, the exact properties of liquid and gas are used instead of the empirical parameter of heat transfer from the interface. However, this two-layer approach imposes the restriction on the thickness of the gas phase. As the novel oscillatory mode cannot be observed at infinitely small β (see Fig. 3a), dimensionless gas layer thickness H_g in the denominator must be finite. Recalling that liquid film thickness H is used as the length scale, thus the novel oscillatory mode

is critical when thicknesses of the gas and the liquid layers are of the same order of magnitude. This is illustrated in Fig. 3b, where the domain of criticality of oscillatory instability is presented on $Ga-(H_g-1)$ plane. Here the realistic system of 0.1 mm silicon fluid of the kinematic viscosity 100 cSt contacting the air layer is considered, which corresponds to dimensionless parameters $Ca = 2000$ and $\lambda_g = 0.2$. It is seen that the thickness of air layer is limited to about 20-fold silicon film thickness.

5 Beyond the lubrication approximation

As explained in the previous sections, the nature of the oscillatory instability is the interaction of slowly evolving, longwave disturbances of temperature and interface deformation, which can be described in the longwave ("lubrication") approximation. However, the oscillatory instability can be obtained numerically without any a priori assumptions on the values of characteristic parameters. The linear eigenvalue problem for small normal perturbations $\sim \exp(iKx + \Sigma t)$, is written as

$$\frac{\Sigma}{Pr}(\psi'' - K^2\psi) = \psi^{IV} - 2K^2\psi'' + K^4\psi, \quad (24)$$

$$\Sigma\theta = \theta'' - K^2\theta - iK\psi, \quad (25)$$

$$z = 0 : \psi = \psi' = 0, \theta' = 0; \quad (26)$$

$$\begin{aligned} z = 1 : \Sigma\xi &= -iK\psi, \\ \psi'''' - \left(\frac{\Sigma}{Pr} + 3K^2\right)\psi' &= iK(Ga + CaK^2)\xi, \\ \psi' + K^2\psi &= iKMa(\xi - \theta), \theta' = Bi(\xi - \theta), \end{aligned} \quad (27)$$

where $\psi(z)$, $\theta(z)$ and ξ correspond to disturbances of stream function, temperature and surface shape, correspondingly.

Solutions of problem (24, 25, 26, 27) at the stability border ($\text{Re}(\Sigma) = 0$) were obtained analytically for the monotonic mode in [35] and for the oscillatory mode in [36]. Expressions that determine the neutral stability curves turn into Eqs. (13) and (15) in the limit $K \ll 1$.

The oscillatory mode thus has been revealed beyond the longwave approximation. Moreover, the parameter range where the oscillatory mode is critical, is extended substantially (see Fig. 3a) compared with one found within the longwave approximation.

6 Influence of insoluble surfactant

In a real system, it is challenging to obtain pure thin film, as the free surface can be contaminated with the *surface active agents* (surfactants). For example, for a water layer, a surfactant is adsorbed fast by the water surface and can form a motionless monolayer on the surface, even when its concentration is tiny [37]. In this section, we discuss the influence of surfactant on the long-wave Marangoni oscillations [38].

Let us consider an insoluble surfactant adsorbed on the interface between the liquid and the ambient gas over it. Thus, the surface tension depends both on temperature and surfactant concentration Γ . The linear thermocapillary and solutocapillary effect are assumed: $\alpha = \alpha_0 - \alpha_T T - \alpha_\Gamma \Gamma$, where $-\alpha_\Gamma = \partial\alpha/\partial\Gamma$.

In addition to the conserved liquid volume and the nearly conserved average temperature, there is one more conserved quantity, the total amount of the surfactant. The longwave disturbances of the surfactant concentration evolve slowly, and hence, they provide one more active variable.

The evolution equation for the surfactant concentration can be written in the non-dimensional form [39] as

$$\frac{\partial\Gamma}{\partial t} - \frac{\partial h}{\partial t}(\mathbf{e}_z \cdot \nabla_\tau)\Gamma + \nabla_\tau \cdot (\mathbf{v}_\tau \Gamma) + (\mathbf{v} \cdot \mathbf{n})\Gamma(\nabla_\tau \cdot \mathbf{n}) = \text{Le} \nabla_\tau^2 \Gamma \quad (28)$$

where $\text{Le} = D_0/\chi$ is the Lewis number, D_0 is the surface diffusion coefficient, $\nabla_\tau = \nabla - \mathbf{n}(\mathbf{n} \cdot \nabla)$. The presence of surfactant also should be taken into account in the balance of tangential stresses on the interface according to

$$\sigma_{n\tau} = -Ma \frac{\partial}{\partial \tau}(T|_{z=h}) - N \frac{\partial}{\partial \tau}(\Gamma|_{z=h}) \quad (29)$$

here $N = \alpha_\Gamma \Gamma_0 H / \rho \nu \chi$ is the elasticity number, Γ_0 is the mean surface concentration.

Applying lubrication theory to the governing equations, one arrives at the following amplitude equations

$$\frac{\partial h}{\partial \tau} = \nabla \cdot \mathbf{j}_1, \quad \mathbf{j}_1 = \frac{h^3}{3} \nabla P + Ma \frac{h^2}{2} \nabla f + N \frac{h^2}{2} \nabla \Gamma, \quad (30)$$

$$\begin{aligned} h \frac{\partial \Theta}{\partial \tau} &= \nabla \cdot (h \nabla \Theta) - \frac{1}{2}(\nabla h)^2 - \beta f + \mathbf{j}_1 \cdot \nabla f \\ &+ \nabla \cdot \left(\frac{h^4}{8} \nabla P + \frac{h^3}{6} Ma \nabla f + \frac{h^3}{6} N \nabla \Gamma \right) \end{aligned} \quad (31)$$

$$\frac{\partial \Gamma}{\partial \tau} = \nabla \cdot \mathbf{j}_2, \quad \mathbf{j}_2 = h \Gamma \left(\frac{h}{2} \nabla P + Ma \nabla f + N \nabla \Gamma \right) + \text{Le} \nabla \Gamma \quad (32)$$

here besides the small Biot number, large capillary number and finite Galileo number, the Lewis number and elasticity number are assumed to be finite.

Obviously, Eqs. (30) and (31) coincide with the Eqs. (9) and (23) if the surfactant is absent.

Linearizing system (30, 31, 32) around the base solution $h = \Theta = \Gamma = 1$ with respect to normal disturbances proportional to $\exp(\sigma\tau + ik_X X + ik_Y Y)$, one obtains cubic equation for the growth rate

$$\begin{aligned} & \sigma^3 + \sigma^2 \left[\beta + k^2 \left(1 + \text{Le} + N + \frac{G - \text{Ma}}{3} \right) \right] \\ & + \sigma \left[\frac{Gk^4}{3} \left(\frac{\beta}{k^2} + 1 + \text{Le} + \frac{1}{3}N \right) + k^2\beta(N + \text{Le}) \right. \\ & \left. - \frac{\text{Ma}k^4(72 + 48\text{Le} + G)}{144} - \frac{k^4(N + 24\text{Le})}{24} \right] \\ & + \frac{Gk^4}{12}(\beta + k^2)(N + 4\text{Le}) - \frac{\text{Ma}k^6\text{Le}(72 + G)}{144} = 0. \end{aligned} \quad (33)$$

This equation possesses both real (monotonic instability) and complex (oscillatory instability) solutions. Neutral curves for monotonic and oscillatory mode are obtained in [38].

The influence of insoluble surfactant on the longwave Marangoni convection is shown in Fig. 2b. It is seen, that even a rather small quantity of insoluble surfactant on the free surface ($N = 0.001$) creates a new type of oscillatory instability, which makes the oscillatory instability mode the most dangerous one for the full range of wave numbers, i.e., for all wave numbers the marginal curves for oscillatory mode are below the monotonic ones.

7 Nonlinear dynamics

An important issue about observation of any instability is investigation of nonlinear development of disturbances beyond the instability threshold. The first scenario is the subcritical excitation, which usually leads to the film rupture [31, 40–43]. Alternatively, the nonlinear interaction of the disturbances can result in pattern formation, which is important for practice [5, 44]. Below the nonlinear development of novel oscillatory Marangoni instability is investigated by means of weakly nonlinear analysis.

7.1 Pure liquid

Consider nonlinear dynamics of small perturbations close to the threshold of the oscillatory instability

$$\text{Ma} - \text{Ma}_c = \delta^2 \text{Ma}_2, \quad (34)$$

where δ is a small parameter characterizing “supercriticality”; $\text{Ma}_c = \text{Ma}^{\text{osc}}$ was obtained from the linear analysis (see Eq. (15)). To describe evolution of wave solutions of Eqs. (9, 10) near the neutral curve, one needs to introduce two timescales, the “fast” and the “slow” times. The timescale $\tau_0 = \tau$ is characteristic for oscillations and the timescale $\tau_2 = \delta^2 \tau$ is characteristic for the growth of the oscillation amplitude near the threshold, so that

$$\frac{\partial}{\partial \tau} = \frac{\partial}{\partial \tau_0} + \delta^2 \frac{\partial}{\partial \tau_2} + \dots \quad (35)$$

The interface deflections and temperature disturbances should also be presented as series in powers of the supercriticality parameter δ

$$h = 1 + \delta h_1 + \delta^2 h_2 + \dots, \quad \Theta = 1 + \delta \Theta_1 + \delta^2 \Theta_2 + \dots \quad (36)$$

At the first order, one obtains the linear stability problem. In order to investigate the nonlinear interaction of waves, consider a particular solution corresponding to a system of traveling waves with the wave vectors $\mathbf{k}_j$

$$h_1 = \sum_{j=1}^N A_j(\tau_2) \exp(i\mathbf{k}_j \cdot \mathbf{R} + i\omega\tau_0) + c.c., \quad (37)$$

$$\Theta_1 = c_0 \sum_{j=1}^N A_j(\tau_2) \exp(i\mathbf{k}_j \cdot \mathbf{R} + i\omega\tau_0) + c.c., \quad (38)$$

where $c_0 = 1 - G/3\text{Ma}_c - 2i\omega/\text{Ma}_c k^2$, $|\mathbf{k}_j| = k$, $\mathbf{R} = (X, Y)$.

At higher orders in small δ , one arrives at the inhomogeneous partial differential equations with the same linear operator as in linear stability problem. Inhomogeneities in these PDEs contain some secular terms, that have to be eliminated according to the following solvability condition

$$\left(i\omega + \frac{\text{Ma}_c k^2}{6} + k^2 + \beta \right) F_1 = \frac{\text{Ma}_c k^2}{2} F_2, \quad (39)$$

where F_1 and F_2 are secular parts of inhomogeneities that arise from Eqs. (9 and 10), respectively. It yields a set of ordinary differential equations, which governs the slow dynamics of amplitudes A_j .

For example, in the case $N = 2$, i.e., a pair of traveling waves with the wave numbers $\mathbf{k}_1 = (k, 0)$ and $\mathbf{k}_2 = (k \cos \phi, k \sin \phi)$, one obtains the set of complex Landau equations that govern evolution of the wave amplitudes A_1 and A_2

$$\frac{dA_1}{d\tau_2} = \left(\gamma - K_0 |A_1|^2 - K_1(\phi) |A_2|^2 \right) A_1, \quad (40)$$

Fig. 4 Selection of oscillatory patterns in a thin film heated from below **(a)** on a square lattice and **(b)** on a hexagonal lattice. Domains of stability for alternating rolls and traveling rectangles are marked by “AR” and “TRa”, respectively. The domains of subcriticality are shaded. The dashed line outlines the area where the oscillatory mode is critical. [28, 45]

$$\frac{dA_2}{d\tau_2} = \left(\gamma - K_0 |A_2|^2 - K_1(\phi) |A_1|^2 \right) A_2, \quad (41)$$

where $\gamma = d\sigma/dMa$, K_0 and K_1 are self-interaction and cross-interaction Landau coefficients. Those coefficients depend on the dimensionless parameters of the system: Galileo number, Biot number and capillary number. Their real parts determine the bifurcation type (super/subcritical) and stability of ordered structures (patterns) on the interface.

Pattern formation in a wide range of problem's parameters was investigated in [27, 45, 46]. The cases of a pair of counterpropagating waves, and patterns on a rhombic lattice, a square lattice, a hexagonal lattice (nonlinear interaction of three waves), a rhombic superlattice (nonlinear interaction of four waves) in the Fourier space were considered. It was shown that, unlike previous results, the supercritical bifurcation is possible, meaning that the stable oscillatory convection can be observed. Among numerous formations, several patterns are stable, i.e., can be observed in experiment; see Fig. 4. Namely, on the square lattice (nonlinear interaction of two cross-propagating traveling waves), the traveling rolls are stable almost in the whole parameter domain, where the oscillatory mode is critical, excluding a small area, where alternating rolls are stable. On the hexagonal lattice (nonlinear interaction of three traveling waves propagating at angle $\phi = 2\pi/3$ between wavenumbers) traveling rolls and traveling rectangles can be observed.

7.2 Influence of insoluble surfactant at the interface

The same procedure of weakly nonlinear analysis can be carried out within the amplitude Eqs. (30–32), that govern the local film thickness, the perturbation of the

free-surface temperature and the surfactant concentration [38].

Two types of solutions were considered: a single traveling wave and a pair of traveling waves propagating in opposite directions. The analysis shows that the bifurcation is typically supercritical near the critical wave number for sufficiently small values of the elasticity parameter. It was shown that the elasticity parameter determines the pattern selection between a possible traveling rolls regime and standing rolls regime. However, at greater values of the elasticity number; for example, at $N = 0.5$, the wave solutions can bifurcate subcritically.

7.3 Influence of nonlinear Marangoni effect

It is known, that some of binary liquids demonstrate quadratic dependence of the surface tension on the temperature [47]. While in the case of a linear dependence of the surface tension on the temperature the film always ruptures, Oron and Rosenau found stable steady states [48]. The authors in [49] consider the influence of the nonlinearity of the surface tension's temperature dependence on the nonlinear development of the oscillatory Marangoni convection in a thin liquid film. They provide the same weakly nonlinear analysis as described in Sect. 6.1, but the Marangoni number is taken into account as the function of temperature of the free surface of the liquid

$$Ma(\Theta) = Ma_0 + M_1 f + M_2 f^2 + \dots, \quad (42)$$

where Ma_0 is the Marangoni number in the conductive state (Eq. (13) for the monotonic mode and (15) for the oscillatory mode), $M_1 = dMa/df$, $M_2 = (1/2)d^2Ma/df^2$.

The authors considered spatially-periodic patterns corresponding to two lattices in the Fourier space: square lattice and hexagonal lattice. Among the oscillatory patterns, single traveling waves as well as traveling rectangles were considered. The influence of nonlinearity of the surface tension function on these regular waves was demonstrated.

8 Active control

Finally, one can think about control of thin film to attain the conditions for observation of the novel mode. Here we discuss the application of the feedback control as efficient instrument of impact. The main advantage of the feedback (or active) control over more popular passive control methods (vibrations, modulations) is that the impact vanishes as soon as the system comes to the desired state.

In [50] the authors compare two control schemes that were applied to the longwave instabilities before.

Following [51] the authors apply the control based on the fluid temperature deviation from conductive value. This control strategy was compared with the one that uses the free surface deflection from planarity as the measure of instability as in [52]. It was demonstrated that the most effective control strategy is the one that relies on the deviation of free surface temperature from its conductive value. In that case the local change of the flux applied on the solid substrate is

$$\left. \frac{\partial T}{\partial z} \right|_{z=0} = -1 - K_f(f)f, \quad f = T|_{z=h} - \bar{T}|_{z=1} \quad (43)$$

where K_f is the non-dimensional scalar control gain, which is rescaled within lubrication approximation as

$$K_f = \varepsilon^2 \chi. \quad (44)$$

Feedback control is aimed to weaken the surface-tension-induced flow. Varying the heat flux at the substrate, one influences the heat transfer from the liquid–gas interface. Consequently, under the feedback control nonlinear Eqs. (9, 10) is simply modified as $\beta \rightarrow \beta - \chi(f)$.

Applying the feedback control one can influence both on the instability threshold and on the nonlinear development of the instability. The linear control law affects the linear stability, thus one can stabilize/destabilize stationary state. Nonlinear control law affects nonlinear dynamics, allowing to eliminate subcritical instability and to change stability of patterns. Below we assume that the additional term for feedback control is a quadratic polynomial of the free surface temperature perturbation

$$\chi(f)f = \chi_l f + \chi_q f^2, \quad (45)$$

where χ_l and χ_q are constants. Thus, in amplitude Eq. (10) the additional terms $(\chi_l f + \chi_q f^2)$ arise.

8.1 Linear feedback control

Neutral curves under feedback control read as [50]

$$\text{Ma}^{\text{mon}} = \frac{48(\beta - \chi_l + k^2)(\text{Ga} + Ck^2)}{k^2(72 + \text{Ga} + Ck^2)}, \quad (46)$$

$$\text{Ma}^{\text{osc}} = 3 + \text{Ga} + Ck^2 + \frac{3(\beta - \chi_l)}{k^2},$$

Obviously, the linear control changes the stability threshold by variation of the effective Biot number β . Consequently, negative values of the linear control gain χ_l increase Ma^{mon} and Ma^{osc} , i.e., stabilize both monotonic and oscillatory instabilities. Under linear feedback control with the positive χ_l oscillatory instability occurs for smaller Ma (i.e. smaller thermal gradient) then for the uncontrolled

convection. Moreover, when $\chi_l > \beta$, additional heat at the substrate prevails against the heat that is carried off from the free surface, so no stable state occurs.

8.2 Nonlinear feedback control

Nonlinear control gain affects nonlinear interaction of perturbations by changing values of coefficients in a set of Landau equations that governs slow evolution of the amplitudes [45, 53]. Thus, under the quadratic control gain one can eliminate subcritical instability.

In the case of interaction of two counterpropagating waves, for the negative quadratic control gain in the interval $-0.2 < \chi_q < -0.07$ there is no subcritical instability neither for single traveling waves nor for a pair of counterpropagating traveling waves in the whole region of parameters where the oscillatory mode is critical (see Fig. 5a). Besides, stable standing rolls can be observed in addition to conventional traveling rolls.

In the case of two cross-propagating waves (corresponded to square lattice) quadratic feedback control can produce such exotic stable patterns as standing squares, see Fig. 5b.

9 Guidance for observations

Finalizing this work, we provide the guidance data for experimental or numerical observations of the oscillatory Marangoni instability in a thin liquid film heated from below.

First, the layer should be very thin, as the oscillatory mode is critical within the domain, that is limited in the Galileo number by the value $\text{Ga}^* < 17.16$ to prevent gravity-induced suppression of the interface deformation.

Fig. 5 Selection of oscillatory patterns under quadratic feedback control with $\chi_q = -0.2$ **(a)** for a pair of counterpropagating waves and **(b)** on a square lattice. Domains of stability for traveling rolls, standing rolls and standing squares are marked by “TR”, “SR” and “SSq” respectively. The domain of subcriticality is shaded. The dashed line outlines the area, where the oscillatory mode is critical. [45, 53]

The substrate should be made from insulated material, not metals. For example, 0.05 mm water layer, which corresponded to $Ga \approx 10$, atop a Plexiglas wall seems to be appropriate system. Indeed, for such water layer $Ga \approx 5 \times 10^4$ and $Bi = 0.001$ (assuming heat transfer coefficient $q = 10 \text{ W/m}^2\text{K}$), that gives $CaBi = 50$, i.e., one gets into domain, where the oscillatory mode is critical. Critical temperature difference in that layer is 0.5 K, when novel oscillatory mode can be found.

Water–air interface, due to the high surface tension, is highly subjected to the uncontrolled contamination by a surfactant. Even small amount of the surface-active agent decreases surface tension, allowing interface deformation and justifying oscillatory instability. However, surfactant absorbed on the interface can form a motionless monolayer, which prevents any interface flow.

The analysis of nonlinear development of instability shows that for the water–air system described above subcritical instability occurs, leading to the film rupture. That unfortunate situation can be eliminated by applying the feedback control.

As the problem does not depend on the Prandtl number, one can choose more viscous fluid, which also allows increasing the thickness of the layer. For example, one can choose the silicone fluid with the kinematic viscosity 100 cSt. The one-layer model predicts the emergence of the oscillatory mode above temperature difference 5 K for 0.1 mm film. However, a correct accounting for the ambient gas shows that the oscillatory mode can be observed if the gas layer is only thinner than 19 thicknesses of the liquid layer. The upper wall above the gas layer should be ideally conductive in this case.

10 Summary

The theoretical results reviewed in this article concern the exploration of the possibility to observe oscillatory Marangoni instability in thin liquid film atop a heated substrate. This oscillatory instability emerges under specific physical parameters, allowing Pearson's and deformational long-wave Marangoni instabilities occur simultaneously. However, estimations show that the instability predicted theoretically can be observed even in a simplest system—thin layer of water ($\sim 0.05 \text{ mm}$). Together with the fact that thin film itself is not the easiest object for experimental or computational study, the limited conditions for observations made this discovery to be perceived as a fully “academic” work. This idea is disproved in this review by several examples of studies that provide additional information about oscillatory Marangoni instability.

We simultaneously relax base assumptions of initial model to demonstrate that the novel mode can be

observed in realistic system. Proper accounting of the heat transfer from thin film to the ambient gas instead of the empirical Newton's law of cooling at the interface results in that the thickness of the ambient gas is important as much as thickness of liquid layer itself. Reconsideration of instability problem beyond the lubrication approximation yields an extension of conditions of emergence of oscillatory mode, as well as extension of the range of wavelength of critical perturbations (emergence of short-wave oscillations). It should be noted however that the Marangoni oscillations in thin film heated from below can be caused by a different instability mechanism. For instance, even a small amount of surface-active agent at the interface of thin film can produce a specific oscillatory mode.

Finally, it should be mentioned that when heating is from below, a subcritical bifurcation takes place due to a monotonic instability and the film usually ruptures (i.e., there is no stable state with a finite deformation of a free surface). However, a novel oscillatory instability can lead to development of stable structures [28, 45], which is certainly very important from a practical standpoint.

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Declarations

Conflict of interest The authors have no conflicts of interest to declare that are relevant to the content of this article.

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References

- Bénard H (1900) Les tourbillons cellulaires dans une nappe liquide. *Rev Gén Sci Pures Appl* 11:1261
- Lappa M (2004) Fluids materials and microgravity: numerical techniques and insights to physics. Elsevier Science, Oxford
- Manz PN, O'Connor JS, Simone G (2020) Microfluidics and Lab-on-a-Chip. RSC Publishing
- Pearson JRA (1958) On convection cells induced by surface tension. *J Fluid Mech* 4:489
- Shklyaev S, Nepomnyashchy A (2017) Longwave instabilities and patterns in fluids. Birkhaeuser, New York
- Sivashinsky GI (1982) Large cells in nonlinear marangoni convection. *Physica D* 4:227
- Knobloch E (1990) Pattern selection in long-wavelength convection. *Physica D* 41:450
- Sternling CV, Scriven LE (1964) On cellular convection driven by surface tension gradients: effects of mean surface tension and surface viscosity. *J Fluid Mech* 19:321
- Smith KA (1966) On convection instability induced by surface tension gradient. *J Fluid Mech* 24:401
- Davis SH (1983) Rupture of thin liquid films, in waves on fluid interfaces. In: Meyer RE, Academic, New York
- Oron A, Davis SH, Bankoff SG (1997) Long-scale evolution of thin liquid films. *Rev Mod Phys* 69:931
- Sternling CV, Scriven LE (1959) Interfacial turbulence: hydrodynamic instability and the Marangoni effect. *AIChE J* 5:514
- Sawistowski H (1981) Interfacial phenomena. In: Hanson C (ed) Recent advances in liquid-liquid extraction. Pergamon, Oxford, p 293
- Linde H, Loeschke K (1966) Rollzellen und oszillation beim waermeuebergang zwischen gas und flussigkeit. *Chem Ing Tech* 39:65
- Levchenko EB, Chernyakov AL (1981) Instability of surface waves in a nonuniformly heated liquid. *Sov Phys JETP* 54:102
- Takashima M (1981) Surface tension driven instability in a liquid layer with a deformable free surface. *J Phys Soc Jpn* 50:2745
- Rednikov AE, Colinet P, Velarde MG, Legros JC (2000) Rayleigh-Marangoni instability in a horizontal liquid layer heated from above: coupling and mode mixing of internal and surface dilatational waves. *J Fluid Mech* 405:57
- Nepomnyashchy AA, Velarde MG, Colinet P (2002) Interfacial phenomena and convection. Chapman and Hall/CRC, Boca Raton
- Nepomnyashchy AA, Simanovskii IB (1986) On the onset of the thermocapillary convection in the presence of surfactants. *Fluid Dyn* 21:469
- Mikishev AB, Nepomnyashchy AA (2010) Long-wavelength Marangoni convection in a liquid layer with insoluble surfactant: linear theory. *Microgravity Sci Technol* 22:415
- Takashima M (1980) Surface tension: driven instability in a horizontal layer of binary liquid mixture in the presence of solet effect II overstability. *J Phys Soc Jpn* 49:802
- Castillo JL, Velarde MG (1982) Buoyancy-thermocapillary instability: the role of interfacial deformation in one- and two-component fluid layers heated from below or above. *J Fluid Mech* 125:463–474
- Oron A, Nepomnyashchy AA (2004) Long-wavelength thermocapillary instability with Soret effect. *Phys Rev E* 69:016313
- Podolny A, Oron A, Nepomnyashchy AA (2005) Long-wave Marangoni instability in a binary-liquid layer with deformable interface in the presence of solet effect: linear theory. *Phys Fluids* 17:104104
- Patne R, Agnon Y, Oron A (2020) Marangoni instability in the linear Jeffreys fluid with a deformable surface. *Phys Rev Fluids* 5:084005
- Nepomnyashchy A, Simanovskii I, Legros JC (2012) Interfacial convection in multilayer systems, 2nd edn. Springer, New York
- Shklyaev S, Khenner M, Alabuzhev AA (2010) Oscillatory and monotonic modes of long-wave Marangoni convection in a thin film. *Phys Rev E* 82:025302
- Shklyaev S, Alabuzhev AA, Khenner M (2012) Long-wave Marangoni convection in a thin film heated from below. *Phys Rev E* 85:016328
- Gershuni GZ, Zhukhovitskii EM (1976) Convective stability of incompressible fluids. Keter, Jerusalem
- Golovin AA, Nepomnyashchy AA, Pismen LM (1995) Pattern formation in large-scale Marangoni convection with deformable interface. *Physica D* 81:117
- VanHook SJ, Schatz MF, Swift JB, McCormick WD, Swinney HL (1997) Long-wavelength surface-tension-driven benard convection: experiment and theory. *J Fluid Mech* 345:45
- Garcia-Ybarra PL, Castillo JL, Velarde MG (1987) Bénard–Marangoni convection with a deformable interface and poorly conducting boundaries. *Phys Fluids* 30:2655
- Golovin AA, Nepomnyashchy AA, Pismen LM (1997) Nonlinear evolution and secondary instabilities of Marangoni convection in a liquid-gas system with deformable interface. *J Fluid Mech* 341:317
- Samoilova AE, Shklyaev S (2015) Oscillatory Marangoni convection in a liquid–gas system heated from below. *Eur Phys J Spec Topics* 224:241
- Samoilova AE, Lobov NI (2014) On the oscillatory Marangoni instability in a thin layer heated from below. *Phys Fluids* 26:064101
- Sarma R, Mondal PK (2018) Marangoni instability in a thin film heated from below: effect of nonmonotonic dependence of surface tension on temperature. *Phys Rev E* 97:043105
- Mizev AI, Trofimenko AI (2014) Effect of an insoluble surfactant film on the stability of the concentration-driven Marangoni flow. *Fluid Dyn* 49:26
- Mikishev AB, Nepomnyashchy AA (2019) Weakly nonlinear analysis of long-wave Marangoni convection in a liquid layer covered by insoluble surfactant. *Phys Rev Fluids* 4:094002
- Mikishev A, Nepomnyashchy A (2015) On dynamic excitation of Marangoni instability in a liquid layer with insoluble surfactant on the deformable surface. *Eur Phys J Spec Top* 224:229
- Schatz MF, VanHook SJ, McCormick WD, Swift JB, Swinney HL (1995) Onset of surface-tension-driven benard convection. *Phys Rev Lett* 75:1938
- Krishnamoorthy S, Ramaswamy SB, Joo SW (1995) Spontaneous rupture of thin liquid films due to thermocapillarity: a full-scale direct numerical simulation. *Phys Fluids* 7:2291
- Oron A (2000) Nonlinear dynamics of three-dimensional long-wave Marangoni instability in thin liquid films. *Phys Fluids* 12:1633
- Yeo LY, Craster RV, Matar OK (2003) Marangoni instability of a thin liquid film resting on a locally heated horizontal wall. *Phys Rev E* 67:056315
- Craster RV, Matar OK (2009) Dynamics and stability of thin liquid films. *Rev Mod Phys* 81:1131
- Samoilova AE, Nepomnyashchy A (2020) Nonlinear feedback control of Marangoni wave patterns in a thin film heated from below. *Physica D* 412:132627

46. Samoilova AE, Nepomnyashchy A (2021) Marangoni patterns on a rhombic lattice in a thin film heated from below. *Phys Fluids* 33:014101
47. Legros JC (1986) Problems related to non-linear variations of surface tension. *Acta Astronaut* 13:697
48. Oron A, Rosenau P (1994) On a nonlinear thermocapillary effect in thin liquid layers. *J Fluid Mech* 273:362
49. Mikishev AB, Nepomnyashchy AA (2020) Influence of nonlinear thermocapillary effect on Marangoni patterns in thin film. *Phys Rev Fluids* 5:054001
50. Samoilova AE, Nepomnyashchy A (2019) Feedback control of Marangoni convection in a thin film heated from below. *J Fluid Mech* 876:573
51. Tang J, Bau HH (1994) Stabilization of the no-motion state in the Rayleigh-Bénard problem. *Proc R Soc A* 447:587
52. Or AC, Kelly RE, Cortelezzi L, Speyer JL (1999) Control of long-wavelength Marangoni-Bénard convection. *J Fluid Mech* 387:321
53. Samoilova AE, Nepomnyashchy A (2021) Controlling of long-wave oscillatory Marangoni patterns on a rhombic lattice. *Math Model Nat Phenom* 16:1

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