

# Oscillatory Marangoni convection in a liquid–gas system heated from below

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**Abstract.** We investigate a longwave Marangoni convection in a two-layer system which consists of a liquid layer and a poorly conductive gas layer. The system is heated from below and confined between two rigid walls: the upper wall is ideally conductive, the lower one is thermally insulated. We aim at finding the analogue of the novel oscillatory mode that was detected analytically within the one-layer approach in [S. Shklyaev, M. Khenner, and A. A. Alabuzhev, “Oscillatory and monotonic modes of long-wave Marangoni convection in a thin film,” *Phys. Rev. E* **82**, 025302 (2010)]. To properly account for the influence of processes in gas on deformation of the interface we apply the two-layer approach. Considering only the heat transfer in gas phase we derive nonlinear amplitude equations that describe the coupled evolution of the layer thickness and temperature perturbation. Linear stability analysis of these equations yields the results similar to those obtained for a single layer whereas nonlinear equations reveal certain differences. The new oscillatory mode is found to be critical in a certain range of parameters, which allows us to provide the recommendations for a possible experiment.

## 1 Introduction

Marangoni convection is known to be studied so thoroughly that every discovery made in that field deserves a great attention. Such a discovery was a new oscillatory mode in a thin film heated from below, see [1], where the authors worked within the one-layer approach. To investigate the influence of processes in fluids on both sides of the interface on the new mode we employ the two-layer model and compare our results with the one-layer results from the above-mentioned paper.

Pearson in [2] considered a single liquid layer with nondeformable free surface and two types of solid substrates: ideally thermally conductive and poorly conductive, when temperature and heat flux are specified, respectively. In the latter case he found a monotonic long-wave instability for the layer heated from below with thermally insulated free surface (this corresponds to zero value of the Biot number).

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Later it was shown that for a small value of the Biot number the critical wavenumber is proportional to  $Bi^{1/4}$ , see, for example [3,4]. The deformational mode was discovered by Scriven&Sternling who took into account free surface deflection and found that in the absence of gravity the layer should be unstable at the infinitely small heating from below, see [5]. Smith in [6] explained the counter-intuitive result of [5]. He demonstrated that gravity suppresses the free surface deflection, which leads to nonzero instability threshold. Later the oscillatory mode was detected in [7] for a heated from above layer atop an ideally thermally conductive substrate. In the monograph [8] a careful numerical analysis of the convection in a liquid layer was provided for both types of thermal boundary conditions at the substrate. However, the authors concluded that the oscillatory mode exists only for heating from above. It was shown in [1,9] that a long-wave oscillatory mode is critical for heating from below; in [10] the new oscillatory mode existence was confirmed by numerical analysis of finite-wavenumber perturbations. The present study is the continuation of the last two works.

In most of the aforesaid works Marangoni convection is studied within the simplest, one-layer, approach which considers the interface as free surface and convection in the liquid layer only. Within such approach the influence of the gas phase is described in a phenomenological way by means of the Biot number, while the Biot number cannot be determined through the physical parameters of the system with a free surface. Within the realistic two-layer approach, processes in fluids on both sides of the interface are considered. Such an approach provides a better agreement with the experimental results (see, for example, [11]) and even reveals the phenomena which cannot be predicted by the one-layer model (“anticonvection”, several oscillatory modes, see [12]). The two-layer approach is unavoidable when the deformability of the interface is important [13,14].

In [1] the authors couple the two long-wave instabilities, Pearson’s and deformational ones, applying the one-layer model which described the Marangoni convection in a thin film heated from below atop a poorly heat conductive substrate. The following assumptions are made: (i) the capillary number is large, (ii) the Biot number is small whereas the product of the capillary number and the Biot number is finite, (iii) the Galileo number is  $O(1)$ . These assumptions allow one (i) to take into account the surface tension within the lubrication approximation, (ii) to prescribe a small heat flux from the free surface, (iii) to prevent the gravity-induced suppression of free surface deflection. Under these restrictions a new oscillatory mode was detected.

In our work we modify the above model to investigate a realistic two-layer system. Within the modified model assumptions (i) and (iii) remain the same, whereas (ii) is rewritten. A small heat flux from liquid across the interface leads to high temperature gradients in the gas phase. Consequently, if the surface is deformable, even small surface deformations can lead to large temperature deviations at the interface and change considerably the liquid-to-gas heat transfer rate. Therefore, we take into account the heat transfer in gas layer assuming its thermal conductivity to be small and its thickness not to be large.

We apply the multiscale expansion to derive the set of nonlinear amplitude equations that describes the coupled evolution of liquid layer thickness and temperature perturbation. Then we perform linear stability analysis of these equations concentrating on searching for an oscillatory mode.

## 2 Problem statement

Let us consider a two-layer liquid-gas system with a deformable interface, sandwiched between horizontal rigid walls. The system is heated from below; the thermal

conductivity of liquid  $\tilde{\kappa}$  is assumed to be large in comparison with that of a lower wall, so that the vertical component of the heat flux  $\tilde{\kappa}A$  is fixed at the substrate. The thermal conductivity of gas  $\tilde{\kappa}_g$  is considered small in comparison with those of liquid and upper rigid wall. (In what follows, we prescribe tildes to the dimensional properties of fluids; the subscript “ $g$ ” refers to gas.)

The liquid density  $\tilde{\rho}$  and the dynamic viscosity  $\tilde{\eta}$  are large in comparison with those for gas, thereby we take into account the heat transfer but not the hydrodynamics in gas phase. Liquid-gas interfacial tension depends linearly on temperature:

$$\tilde{\sigma} = \tilde{\sigma}_0 + \tilde{\alpha}\tilde{T};$$

hence, the Marangoni mechanism of convective instability in a liquid is considered predominant. The unperturbed liquid-layer thickness  $h_0$  is sufficiently small, so that the interface deflection should be taken into account.

The surface-tension-driven convection in this system is governed by the following system of dimensionless equations and boundary conditions:

$$\begin{aligned} \frac{1}{\text{Pr}} \left( \frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) &= -\nabla p + \nabla^2 \mathbf{v} - \text{Ga} \mathbf{k}, \\ \frac{\partial T}{\partial t} + \mathbf{v} \cdot \nabla T &= \nabla^2 T, \quad \frac{\partial T_g}{\partial t} = \chi_g \nabla^2 T_g, \\ \nabla \cdot \mathbf{v} &= 0, \end{aligned} \quad (1)$$

$$z = 0: \mathbf{v} = 0, \quad \frac{\partial T}{\partial z} = -1,$$

$$z = h(x, y, t): \frac{\partial h}{\partial t} = w - \mathbf{u} \cdot \nabla h,$$

$$\sigma_{n\tau} = -\text{Ma} \frac{\partial}{\partial \tau} (T|_{z=h}), \quad \sigma_{nn} = p - \text{Ca}K, \quad (2)$$

$$T = T_g, \quad \frac{\partial}{\partial n} (T - \kappa_g T_g) = 0,$$

$$z = H: T_g = 0,$$

where  $v = (\mathbf{u}, w)$  is the liquid velocity ( $\mathbf{u}$  is velocity in the horizontal  $x - y$  plane and  $w$  is the vertical component),  $p$  is the pressure,  $\sigma$  is the viscous stress tensor of the liquid,  $h$  is the local liquid-layer thickness,  $H$  is the overall thickness of two layers,  $\mathbf{k}$  is the upward unit vector;  $K$ ,  $n$  and  $\tau$  are the curvature, the normal and tangential unit vectors of the interface, respectively.  $T$  and  $T_g$  are the temperature in the liquid and the gas, respectively.

We use the following scalings:  $h_0$  for the length,  $h_0^2/\tilde{\chi}$  for the time,  $\tilde{\chi}/h_0$  for the velocity,  $\tilde{\eta}\tilde{\chi}/h_0^2$  for the pressure,  $Ah_0$  for the temperature ( $\tilde{\chi}$  is the thermal diffusivity of liquid). The problem is characterized by the following dimensionless parameters:

$$\text{Ca} = \frac{\tilde{\sigma}_0 h_0}{\tilde{\eta}\tilde{\chi}}, \quad \text{Ma} = \frac{\tilde{\alpha} h_0^2 A}{\tilde{\eta}\tilde{\chi}}, \quad \text{Ga} = \frac{g h_0^3}{\tilde{\nu}\tilde{\chi}}, \quad \text{Pr} = \frac{\tilde{\nu}}{\tilde{\chi}}, \quad \kappa_g = \frac{\tilde{\kappa}_g}{\tilde{\kappa}}, \quad \chi_g = \frac{\tilde{\chi}_g}{\tilde{\chi}},$$

which are the capillary, Marangoni, Galileo and Prandtl numbers, dimensionless heat conductivity and thermal diffusivity, respectively. Here  $\tilde{\nu}$  is the kinematic viscosity of the liquid.

Equations (1)–(2) have the obvious base solution, that corresponds to the conductive state:

$$h^{(0)} = 1, \quad T^{(0)} = -z + 1 + \frac{H-1}{\kappa_g}, \quad T_g^{(0)} = \frac{H-z}{\kappa_g}, \quad p^{(0)} = \text{Ga}(1-z). \quad (3)$$

Below we study the linear stability of the conductive state with respect to the long-wave perturbations and the evolution of the large-scale perturbations into the conductive state.

### 3 Lubrication approximation

To study the evolution of a large-scale flow, we rescale the coordinates, the time and the velocity according to the relations

$$X = \epsilon x, \quad Y = \epsilon y, \quad Z = z, \quad \tau = \epsilon^2 t, \quad \mathbf{U} = \epsilon \mathbf{u}, \quad W = \epsilon^2 w, \quad (4)$$

where  $\epsilon \ll 1$  can be thought of as the ratio of  $h_0$  to a typical horizontal lengthscale. We apply the following expansion

$$\begin{aligned} \mathbf{U} &= \mathbf{U}_0 + \epsilon^2 \mathbf{U}_1 + \dots, \quad W = W_0 + \epsilon^2 W_1 + \dots, \quad p = p_0 + \epsilon^2 p_1 + \dots, \\ T &= -z + \frac{H-1}{\kappa_g} + T_0 + \epsilon^2 T_1 + \dots, \\ T_g &= \frac{H-z}{\kappa_g} + T_{g0} + \epsilon^2 T_{g1} + \dots \end{aligned} \quad (5)$$

We do not drop out the equilibrium fields from  $T$  and  $p$ ; it is clear that the base conductive state corresponds to  $T_0 = 1, p_0 = p^{(0)}$ .

### 4 Amplitude equations

As our aim is to find the analogue of the new oscillatory mode in the two-layer liquid-gas system by coupling two monotonic modes, Pearson's and deformational, we apply the same scalings as in [1]: large values of the capillary number, a finite Galileo number, and a small gas thermal conductivity

$$\text{Ca} = \epsilon^{-2} C, \quad \text{Ga} = O(1), \quad \kappa_g = \epsilon^2 \kappa_{g2}. \quad (6)$$

The latter assumption allows us to prescribe a small heat flux from the liquid through the interface.

Substituting Eqs. (4)–(6) into problem (1)–(2) we obtain the following zeroth-order boundary-value problem

$$\begin{aligned} p_{0Z} &= -\text{Ga}, \quad \nabla p_0 = \mathbf{U}_{0ZZ}, \quad W_{0Z} = -\nabla \cdot \mathbf{U}_0, \\ T_{0ZZ} &= 0, \quad T_{g0ZZ} = 0, \end{aligned} \quad (7)$$

$$Z = 0: W_0 = \mathbf{U}_0 = T_{0Z} = 0,$$

$$\begin{aligned} Z = h: h_\tau &= W_0 - \mathbf{U}_0 \cdot \nabla h, \quad p_0 = -C \nabla^2 h, \quad \mathbf{U}_{0Z} = -M \nabla (T_0 - h), \\ T_0 - h &= T_{g0}, \quad T_{0Z} = T_{g0Z}, \end{aligned} \quad (8)$$

$$Z = H: T_{g0} = 0.$$

Hereafter  $\nabla \equiv (\partial_X, \partial_Y, 0)$  is a two-dimensional projection of the gradient operator onto the  $X - Y$  plane.

The solution of the problem is

$$\begin{aligned} p_0 &= P(X, Y, \tau) - \text{Ga}Z, \\ \mathbf{U}_0 &= \frac{Z}{2}(Z - 2h)\nabla P - Z\text{Ma}\nabla f, \\ W_0 &= \frac{Z^2}{2}\nabla \cdot \left[ \frac{1}{3}(3h - Z)\nabla P + \text{Ma}\nabla f \right], \\ T_0 &= \Theta(X, Y, \tau), T_{g0} = \frac{H - Z}{H - h}\Theta, \end{aligned} \quad (9)$$

where  $P = \text{Ga}h - C\nabla^2 h$ , and  $f = \Theta - h$  is the perturbation of the interface temperature. The evolution of the liquid layer thickness is governed by the well-known condition  $h_\tau = -\nabla \cdot \int_0^h U_0 dz$ , which provides the first amplitude equation

$$h_\tau = \nabla \cdot \left[ \frac{h^3}{3}\nabla P + \text{Ma}\frac{h^2}{2}\nabla f \right] = \nabla \cdot \mathbf{j}, \quad (10)$$

where vector  $-\mathbf{j}$  is the longitudinal flux of liquid averaged across the layer.

From the first order of the expansion we need only the equation that describes heat transfer in liquid

$$T_{1ZZ} = \Theta_\tau - \nabla^2 \Theta + \mathbf{U}_0 \cdot \nabla \Theta - W_0, \quad (11)$$

$$Z = 0 : T_{1Z} = 0, \quad Z = h : T_{1Z} = \nabla h \cdot \nabla \Theta - \frac{\kappa_{g2}}{H - h} f. \quad (12)$$

The solvability condition of this problem provides the second amplitude equation. The integration of Eq. (11) with the boundary conditions (12) results in

$$h\Theta_\tau = \nabla \cdot (h\nabla \Theta) - \frac{\kappa_{g2}}{H - h} f + \mathbf{j} \cdot \nabla f + \nabla \cdot \left( \frac{h^4}{8}\nabla P + \frac{h^3}{6}\text{Ma}\nabla f \right). \quad (13)$$

Equations (10) and (13) form a closed set of amplitude equations which describes the nonlinear dynamics of long-wave perturbations. These equations include the following effects: in the right-hand side of Eq. (10), suppression of the interface deflection by gravity and surface tension, and influence of the thermocapillary flow on the layer thickness; in the right-hand side of Eq. (13), heat conductivity in the longitudinal directions (the first term), heat losses from the interface into gas phase (the second term), and advective heat transfer by the flow (the third and fourth terms).

The base state, corresponding to motionless fluid and gas with a constant heat flux maintained through the layers, is given by  $h = 1$  and  $\Theta = 1$ .

#### 4.1 Comparison with the one-layer model

Longwave Marangoni convection in a thin film heated from below atop a poorly conducting substrate was investigated in [1] within the one-layer approach. The problem (1)–(2) was formulated in liquid phase only, where the thermal conditions at the free surface were prescribed by Newton's law of cooling. The authors used similar scalings to Eqs. (4)–(6) except for the assumptions of smallness of the heat flux from the free surface, which was replaced by the following scaling of the Biot number  $\text{Bi} = \beta\epsilon^2$ .

Our set of amplitude Eqs. (10) and (13) is very close to the amplitude equations derived within the one-layer approach. Evolutionary equations of liquid layer thickness in both models are absolutely the same. There are two differences in the evolutionary equation for temperature perturbation. The first one is that in the two-layer approach the term corresponding to the heat losses from the interface into gas phase is  $\kappa_{g2}\Theta/(H-h)$ , whereas the similar term from the one-layer model is  $\beta\Theta$ . The second difference is that the term from the one-layer model  $(\nabla h)^2/2$  corresponding to the heat losses from the free surface is absent in the two-layer model, see Eq. (13). Apparently all the differences between one-layer and two-layer evolutionary equations are nonlinear, therefore, the results of linear analysis are similar if we denote  $\kappa_{g2}/a \equiv \beta$  (where  $a = H - 1$  means dimensionless thickness of the gas layer). Note that it is not valid when the gas layer becomes thick enough or especially infinite.

## 5 Linear stability analysis

Considering small perturbations of temperature and interface deflection,  $\Theta = 1 + \theta$  and  $h = 1 + \zeta$ , and linearizing the equations we arrive at

$$\zeta_\tau = \nabla^2 \left[ \frac{1}{3} (\text{Ga}\zeta - C\nabla^2\zeta) + \frac{\text{Ma}}{2} (\theta - \zeta) \right], \quad (14)$$

$$\Theta_\tau = \nabla^2 \left[ \theta + \frac{1}{8} (\text{Ga}\zeta - C\nabla^2\zeta) + \frac{\text{Ma}}{6} (\theta - \zeta) \right] - \frac{\kappa_{g2}}{a} (\theta - \zeta), \quad (15)$$

from which we obtain a quadratic equation for the growth rate, representing the perturbation fields proportional to  $\exp(\lambda\tau + ikX)$ ,

$$\lambda^2 + \lambda \left[ k^2 \left( 1 + \frac{\text{G} - \text{Ma}}{3} \right) + \frac{\kappa_{g2}}{a} \right] + \frac{k^2}{3} \left( k^2 + \frac{\kappa_{g2}}{a} \right) \text{G} - \frac{Mk^4}{2} \left( 1 + \frac{\text{G}}{72} \right) = 0, \quad (16)$$

where  $\text{G} = \text{Ga} + Ck^2$ . This equation possesses both real (monotonic instability) and complex (oscillatory instability) solutions. The neutral stability curves for the monotonic ( $\text{Ma}_m$ ) and oscillatory ( $\text{Ma}_o$ ) modes are given by

$$\text{Ma}_m = \frac{48 \left( k^2 + \frac{\kappa_{g2}}{a} \right) \text{G}}{k^2 (72 + \text{G})}, \quad \text{Ma}_o = 3 + \frac{3}{k^2} \frac{\kappa_{g2}}{a} + \text{G}. \quad (17)$$

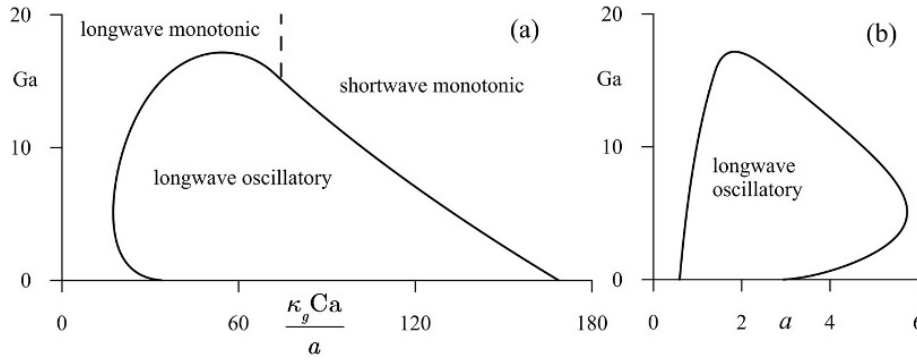
Neutral curve for the monotonic mode has a minimum at the finite values of  $k$  only if

$$\frac{\kappa_{g2}C}{a} < 72, \quad (18)$$

otherwise the critical value  $\text{Ma}_c^{sw} = 48$  is achieved in the limit  $k \rightarrow \infty$  (i.e., the shortwave mode is critical). The imaginary part of the growth rate (frequency) for neutral oscillatory perturbations is

$$\lambda_i = \frac{k^2}{12} \sqrt{(72 + \text{Ga} + k^2) (\text{Ma}_m - \text{Ma}_o)}. \quad (19)$$

It is clear that the oscillatory mode is present only at  $\text{Ma}_o(k) < \text{Ma}_m(k)$ .



**Fig. 1.** (a) The domain of oscillatory instability. The dashed vertical line marks the boundary of the longwave monotonic instability, Eq. (18). Panel (b) shows the domain where the oscillatory mode is critical on the  $Ga - a$  plane for  $Ca = 1000$  and  $\kappa_g = 0.1$ .

Keeping in mind the possible future experiments, below we present the expressions for the neutral stability curves in terms of the unscaled wave number  $K = \epsilon k$ , gas thermal conductivity  $\kappa_g$  and capillary number  $Ca$ . For the monotonic mode the stability threshold is

$$Ma_m = \frac{48 (K^2 + \frac{\kappa_g}{a}) (Ga + CaK^2)}{K^2 (72 + Ga + CaK^2)}. \quad (20)$$

For the oscillatory mode the stability threshold is

$$Ma_o = 3 + \frac{3}{K^2} \frac{\kappa_g}{a} + Ga + CaK^2. \quad (21)$$

The parameter range where the oscillatory mode is critical is shown in Fig. 1a. The boundaries between the domains of monotonic and oscillatory instabilities obtained within the one-layer and two-layer approaches coincide if we denote  $\kappa_g/a \equiv Bi$ . However, our two-layer model reveals severe restrictions on the thickness of gas layer: for silicon oil-air system under earthly conditions oscillatory mode is critical only if the gas layer is sufficiently thin (see Fig. 1b). Therefore, the two-layer approach allows us to provide a realistic estimates for a possible experiment. Considering the 1 mm air layer over the 0.1 mm film of the silicon fluid of the kinematic viscosity 100 cSt we obtain  $Ga = 10$  and  $Ca = 2000$ . The characteristic wavelength of the convective structure is 1 cm and the period of the oscillations is 50 s. The critical Marangoni number is attained at the temperature difference 4 K.

## 6 Conclusion

We have investigated the longwave Marangoni convection in a two-layer system with a deformable interface which consists of a liquid layer and a poorly conductive gas layer. We assume capillary number  $Ca \gg 1$ , ratio of gas and liquid thermal conductivities  $\kappa_g \ll 1$ , whereas Galileo number and ratio of gas and liquid thicknesses  $a$  to be finite. Within the two-layer approach we take into account heat transfer but not hydrodynamics in the gas phase. The set of amplitude equations, Eqs. (10) and (13), which describes a coupled evolution of the liquid film height and the averaged across the liquid layer part of the temperature is derived.

This set of equations is similar to the amplitude equations derived by the authors in [1] within the one-layer approach with a minor difference in nonlinear terms. The linear stability analysis of our amplitude equations confirms the existence of the new oscillatory mode, see Fig. 1a. The comparison of linear analysis results provided within the two-layer and one-layer approaches demonstrates their coincidence if we denote  $\kappa_g/a \equiv \text{Bi}$ . However, the important distinction of the two-layer results is that the effective empirical parameter – the Biot number – is replaced by exact liquid and gas characteristics. Thereby, within the two-layer approach we found a severe restriction on gas layer thickness. The new oscillatory mode is critical only if the gas layer is sufficiently thin in comparison with the liquid layer, see Fig. 1b. This allowed us to provide better estimates for possible future experiments.

Finally, it is worth noting that low heat conductivity of the gas in combination with small thickness of the gas layer leads to high temperature gradients in the gas phase. If the interface is deformable, even small surface deformations can lead to large temperature deviations at the interface. Therefore, the convective heat transport in the gas phase should be considered in order to get well-substantiated results in Marangoni convective pattern formation in systems with a deformable interface. However, this task is way beyond the present investigation.

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