Regular Article

Implications of the Marangoni effect on the onset of Rayleigh–Benard convection in a two-layer system with a deformable interface

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Received 1 August 2014 / Received in final form 16 February 2015 Published online 8 April 2015

Abstract. The paper deals with the investigation of the implications of the Marangoni effect on the onset of Rayleigh-Benard convection in a two-layer system with deformable fluid interface. The study of the conductive state stability to the longwave perturbations shows that in the case of heating from above the thermocapillary effect leads to the increase of instability domain to the monotonic longwave perturbations. In the case of heating from below, the thermocapillarity makes stabilizing effect on the longwave perturbations and at some values of the parameters the configuration where more dense fluid is located above less dense one turns out to be stable. However, the analysis of the perturbations with finite wavelength in the presence of thermocapillary effect shows that in the case of heating from below the Rayleigh-Taylor instability is not suppressed. For any values of the parameters the perturbations with finite wavelength turn out to be more dangerous. In this situation the instability domain becomes wider with the increase of the Marangoni number modulus. In the case of heating from above, for any values of the Marangoni number, at the Rayleigh numbers small in the modulus, long-wave monotonic perturbations are most dangerous whereas at the Rayleigh numbers large in the modulus, the most dangerous mode is cellular monotonic instability.

1 Introduction

The onset of thermal buoyancy convection in two-layer systems of immiscible fluids has been explored in many theoretical and experimental works, see e.g. [1-4]. Most of these studies have been performed in the framework of the Boussinesq approximation, neglecting interface deformations. In [5] it is shown that accounting for interface deformations under the ordinary Boussinesq approximation can yield physically incorrect results. If the relative density difference is of the same order of magnitude as the density inhomogeneities caused by non-isothermality, then gravity is unable

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to retain the interface planar at finite Rayleigh numbers. In this case interface deformations may be large and therefore should be properly accounted for. A generalized Boussinesq approximation in the case where both the density inhomogeneities and the Boussinesq parameter are small was independently formulated by Busse [6] and Lyubimov [7].

In [6] the stability of the conductive state of a two-layer system of immiscible fluids with a deformable interface and perfectly conductive external boundaries was studied for fluids with close densities on the basis of the generalized Boussinesq approximation. The layer thicknesses and other parameters of fluids, except the densities, were assumed to be the same. Monotonic and oscillatory instability modes with finite wavelength were found. Later on, Lobov, Lyubimov and Lyubimova [7] found a solution to this problem for fluids with different properties and discovered a monotonic long-wave instability mode associated with the interface deformability (such mode does not exist in a particular case considered in [6]). They noted that long-wave perturbations are most dangerous in a wide range of parameters.

In the case of a single horizontal layer of fluid under prescribed heat fluxes at the external boundaries, the conductive state instability occurs at heating from below due to the development of monotonic long-wave perturbations [8]. The stability of the conductive state of a two-layer system of horizontal layers of immiscible fluids under prescribed heat fluxes at the external boundaries and non-deformable interface was studied in [3]. The calculations showed the existence of the long-wave monotonic instability when the ratio of the lower layer thickness to the total thickness of the two-layer system is close to zero or unity, i.e. the system was close to a single layer under given heat fluxes at the external boundaries. They also revealed the presence of long-wave monotonic instability at the intermediate values of layer thickness ratio close to 0.5.

Two long-wave instability modes (monotonic and oscillatory) were discovered in [9, 10] for the two-layer system of horizontal layers of immiscible fluids with a deformable interface subjected to the prescribed heat fluxes at the external boundaries.

In the systems with free surfaces or fluid interfaces, thermocapillary forces (for the transverse layer heating) may lead to the development of initial perturbations (Marangoni instability). When the layers are of small thicknesses, the Marangoni convection has a significant influence on the stability of conductive state.

In a series of papers [11,12], thermocapillary instability in model two-layer systems was studied, and the impact of various factors on this type of instability was analysed. It was found that in the system of fluids with equal properties and a planar interface the monotonic instability occurs only in the case of heating from the thick layer, and the oscillatory instability takes place under cooling from this layer [11]. In thermocapillary waves, the longitudinal motions dominate near the interface between the layers. The temperature field keeps these oscillations at the expense of motion phase differences in different layers. In [12] the conditions for monotonic and oscillatory instabilities in a two-layer system with a deformable interface at zero thermal Rayleigh number are determined. In the oscillatory convection regimes, two different mechanisms were distinguished: capillary, when the kinetic energy of flow is converted into the potential energy of the deformable interface (capillary waves), and thermocapillary, when during the motion the surface free energy varies due to a change in its temperature (thermocapillary waves). Capillary and thermocapillary waves differ significantly in the flow structure.

In [13] the Rayleigh – Benard – Marangoni convection in a two-layer system of fluids with a deformable interface and perfectly conductive external boundaries was investigated within conventional Boussinesq approximation. The results of theoretical and experimental studies for the benzene-water system are provided. Within the framework of the linear stability theory, it was found that in the case of heating from below the critical Marangoni number is smaller than in the case of heating from above. The experiments showed that the critical Rayleigh number in the system heated from below was in the range between the critical Rayleigh number predicted by the theory in the absence and in the presence of thermocapillary convection; in the system heated from above no instability was detected even for Marangoni numbers five times greater than the critical value calculated based on the linear theory. Discrepancies between the theoretical and experimental critical values of the Rayleigh number were attributed to the interface contamination.

In [14] theoretical and experimental studies of the Rayleigh-Benard-Marangoni instability in a two-layer system of fluids with a deformable interface were performed for different layer thickness and perfectly conductive external boundaries. The longwave oscillatory instability was observed for fluids with close densities. Long-wave oscillatory perturbations grow when the layer of greater thickness is heated.

The purpose of the present paper is to study the Marangoni effect on the onset of the Rayleigh-Benard convection in a two-layer system of immiscible fluids with a deformable interface under the prescribed heat flux at the external boundaries. The study is performed in the framework of a generalized Boussinesq approximation using the Busse-Lyubimov model [6,7].

2 Problem formulation and mathematical model

Consider a two-layer system of horizontal layers of immiscible fluids with a deformable interface. The layers are bounded by rigid plates $z = -h_1$, h_2 , at which a constant heat flux is preset: $\partial T_1/\partial z = const$ and $\partial T_2/\partial z = const$. The system of coordinates is taken in such a way that the z-axis is directed vertically upward, and the x-y plane coincides with the undisturbed interface. In virtue of the problem symmetry in the x-y plane (problem isotropy), the two-dimensional problem in the x-z plane is examined; the layers of equal thicknesses $h_1 = h_2 = h$ are considered.

We investigate the fluids of close densities, which allows to correctly take into account the deformations of the interface by solving the problem in terms of the Busse-Lyubimov model. In accordance with this model, the density of each fluid depends only on temperature; the density variations due to thermal expansion are assumed to be small and satisfying the linear law $\rho_j = \rho_{0j}(1 - \beta_j(T_j - T_0))$, where β_j are the thermal expansion coefficients, ρ_{0j} is the density of the *j*-th fluid at temperature T_0 , taken as a reference point and equal to the conductive state value of the interface temperature and

$$\delta = 2 \frac{\rho_{02} - \rho_{01}}{\rho_{02} + \rho_{01}}, \quad |\delta| \ll 1.$$
(1)

Within the Busse-Lyubimov model, the conventional Galileo number $Ga_* = gh^3/\nu_*^2$ is assumed to be an asymptotically large parameter, and its product with every small parameter – finite: Ra = $\beta_*(\Theta_1 - \Theta_2)Ga_*\nu_*/2\chi_*$ is the Rayleigh number, and Ga = $\delta Ga_*\nu_*/\chi_*$ is the modified Galileo number. Here, β_*, ν_*, χ_* are the average values of thermal expansion, kinematic viscosity and thermal diffusivity coefficients. Thus, in this model, the densities of two fluids are assumed to be constant and equal to the average value ρ_* everywhere, except the terms describing gravity force in stress balance condition at the interface. Positive modified Galileo numbers correspond to the potentially unstable stratification of fluids (the lighter fluid is at the bottom), and negative Ga the stable stratification. According to the Busse-Lyubimov model, the system of equations for thermal buoyancy convection is written as

$$\frac{1}{\Pr} \left(\frac{\partial \mathbf{v}_j}{\partial t} + (\mathbf{v}_j \nabla) \mathbf{v}_j \right) = -\nabla p_j + \nu_j \Delta \mathbf{v}_j + \operatorname{Ra}\beta_j T_j \boldsymbol{\gamma}$$
(2)

$$\frac{\partial T_j}{\partial t} + (\mathbf{v}_j \nabla) T_j = \chi_j \Delta T_j, \quad \text{div} \, \mathbf{v}_j = 0.$$
(3)

Here j = 1, 2, index 1 denotes the quantities pertaining to the lower fluid, and index 2 those pertaining to the upper fluid; γ is the unit vector directed vertically upward. The equations are written in dimensionless form. For the scales of time, length, velocity, temperature and pressure we use h^2/χ_* , h, χ_*/h , A_*h , $\rho\nu_*\chi_*/h^2$, respectively. As the scales for viscosity, thermal conductivity, thermal expansion, thermal dif-

As the scales for viscosity, thermal conductivity, thermal expansion, thermal diffusivity and conductive state temperature gradients, we take their arithmetic mean values $\nu_* = (\nu_{1*} + \nu_{2*})/2$, $\kappa_* = (\kappa_{1*} + \kappa_{2*})/2$, $\beta_* = (\beta_{1*} + \beta_{2*})/2$, $\chi_* = (\chi_{1*} + \chi_{2*})/2$, $A_* = (A_{1*} + A_{2*})/2$. With this choice of scales, the following relations are fulfilled:

$$\nu_1 + \nu_2 = 2, \quad \kappa_1 + \kappa_2 = 2, \quad \beta_1 + \beta_2 = 2, \quad \chi_1 + \chi_2 = 2, \quad A_1 + A_2 = 2.$$
 (4)

On rigid external boundaries, the no-slip conditions and prescribed heat fluxes are imposed:

$$z = \pm 1$$
: $\mathbf{v}_j = 0, \qquad \frac{\partial T_j}{\partial z} = const.$ (5)

The thermal conductivities of fluids are assumed to be constant, therefore the condition of constancy of the heat fluxes means the condition of constancy of the temperature gradients.

The kinematic condition and the conditions of continuity of velocity, temperature and heat flux at the interface take the form:

$$z = \zeta; \quad \frac{\partial \zeta}{\partial t} + (\mathbf{v}\nabla) \,\zeta = \mathbf{v}\gamma, \quad [\mathbf{v}] = 0, \quad [T] = 0, \quad [\kappa \nabla T] \,\mathbf{n} = 0, \tag{6}$$

where [f] is a jump of the quantity f across the interface, i.e. $[f] = (f_1 - f_2)_{z=\zeta}$, **n** is the vector of the normal to the interface directed from the first medium to the second one; $\zeta = z(x,t)$ is the equation for the interface ($\zeta = 0$ for the planar interface).

The continuity conditions for normal and tangential stresses at the interface, written under the assumption that the surface tension is linearly dependent on the temperature, have the form:

$$-[p] + [\sigma_{nn}] + \operatorname{Ga}\zeta = (\operatorname{Ca} + \operatorname{Ma} T) K, \quad [\sigma_{n\tau}] = \operatorname{Ma} \nabla T$$
(7)

where K is the interface curvature.

The boundary-value problem (2)–(7) contains the following dimensionless parameters: Prandtl number Pr, Rayleigh number Ra, capillarity parameter Ca, modified Galileo number Ga, Marangoni number Ma.

$$Pr = \nu_*/\chi_*, \quad Ra = g\beta_*A_*h^4/(\nu_*\chi_*), \quad Ca = \alpha_0 h/(\nu_*\chi_*\rho_0), Ga = (\rho_{02} - \rho_{01})gh^3/(\eta_*\chi_*), \quad Ma = (\partial \alpha/\partial T) \left(h^2 A_*/(\chi_*\eta_*)\right).$$
(8)

Here, $\eta_* = \rho_0 \nu_*$ is the average value of dynamic viscosity, α_0 is the value of surface tension at temperature taken as the reference point. Note that, for the majority of fluids, $\partial \alpha / \partial T$ is negative, and therefore the positive values of the Marangoni number correspond to heating from above, negative to heating from below.

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The problem (2)–(7) admits a stationary solution related to the quiescent fluids with a planar horizontal interface $\mathbf{v}_{0j} = 0$, $\zeta_0 = 0$. Temperature distribution in the layers corresponds to the heat-conductive regime:

$$T_{0j} = -z A_j, \quad p_{0j} = -\text{Ra}\beta_j A_j z^2/2, \quad A_1 = \kappa_2, \quad A_2 = \kappa_1.$$
 (9)

3 Long-wave instability of conductive state

Let us investigate the linear stability of the conductive state of the system to small normal perturbations of the form: $\exp(\lambda t + i\mathbf{kr})$.

To study stability to long-wave perturbations, all the fields and the increment λ are expanded into a power series of the wavenumber:

$$\lambda = k\lambda_1 + k^2\lambda_2 + \dots, \quad q_j = q_j^{(0)} + kq_j^{(1)} + \dots$$

$$u_j = ku_j^{(1)} + k^2u_j^{(2)} + \dots, \quad w_j = kw_j^{(1)} + k^2w_j^{(2)} + \dots$$

$$\theta_j = \theta_j^{(0)} + k\theta_j^{(1)} + \dots, \quad \zeta = \zeta^{(0)} + k\zeta^{(1)} + \dots$$
(10)

Here we introduced the following notations for the amplitudes of the perturbations: u_j – for the projection of the velocity in the direction of the wave vector, w_j – for the vertical velocity component, θ_j – for temperature, q_j – for the pressure.

Calculations showed that λ_1 vanishes identically, and for λ_2 a quadratic equation is obtained

$$\lambda_2^2 + B\lambda_2 + C = 0$$

whose coefficients are the cumbersome functions of the parameters (their explicit form is not given here).

The problem under consideration contains a large number of dimensionless parameters, so the calculations were performed for a specified pair of fluids – formic acid – transformer oil; the study of the stability of such a system in the absence of Marangoni convection was carried out in [3, 7, 9, 10].

3.1 Heating from below

Figure 1 presents the boundaries of conductive state instability to the long-wave monotonic and oscillatory perturbations in the parameter plane modified Galileo number – Rayleigh number in the case of heating from below.

In this and all subsequent figures the solid lines indicate the boundaries of instability to monotonic perturbations, and the dashed lines to oscillatory perturbations. When the thermocapillary effect is absent [9] (Ma = 0, the black lines in Fig. 1), two branches of the monotonic instability of hyperbolic type 1 and 2 occur. The branch 1 corresponds to the case when the heavier fluid is at the bottom (negative modifed Galileo numbers). In this case the perturbations have two-floor structure (Fig. 2c) and the interface is nearly non-deformed. To the right of the branch 2, the Rayleigh-Taylor instability develops which correspond to the critical perturbations having the single-floor structure (Fig. 2a).

Between the branches of monotonic instability there is an oscillatory instability boundary in the form of a straight line. The structure of critical perturbations associated with the threshold of oscillatory instability continuously changes with the modifed Galileo number from the two-floor structure at negative Ga large enough in the modulus to the through flow (single-floor perturbations which cover the total



Fig. 1. Boundaries of long-wave instability for the system heated from below; black lines – Ma = 0, grey lines – Ma = -50; solid lines – monotonic instability, dashed lines – oscillatory instability.

layer thickness) at Ga close to zero (Fig. 2b). For weak thermocapillary effect (grey lines in Fig. 1) the long-wave instability boundaries are qualitatively similar to those at Ma = 0. At negative Galileo numbers large in the modulus (Ga < -219.22), weak thermocapillary forces make stabilizing effect on the two-floor long-wave monotonic perturbations and at Ga > -219.22 destabilizing effect. The effect of weak thermocapillary forces on oscillatory long-wave perturbations is stabilizing and, as one can see from Fig. 1, this results in the extension of the stability domain. Moreover at some parameter values, the configuration, where the fluid of higher density is located above, may become stable. However, whether these long-wave perturbations are most dangerous and whether the suppression of the Rayleigh-Taylor instability is possible should be revealed in the analysis of the stability to the perturbations with finite wavelengh.

Figure 3 describes the evolution of the instability boundaries with the increase of thermocapilary effect. The instability boundaries are depicted in the parameter plane modified Galileo number – Rayleigh number. As one can see, at the negative Marangoni numbers smaller in the modulus than 187.1 (grey thick lines in Fig. 3a, Fig. 3d, Ma = -170) the location of the long-wave instability boundaries remains qualitatively the same as in the absence of the thermocapillary effect. At Ma \approx -187.1 the qualitative rearrangement takes place: recoupling of the branches of monotonic long-wave instability boundaries. As the result the system turns out to be unstable for any parameter values (Fig. 3c, Fig. 3d). In our opinion this rearrangement is related to the exchange of contributions of two governing mechanisms: at Ma > -187.1 the gravity effect dominates and at Ma < -187.1 the thermocapillary effect. Similar arrangement takes place for coupled magneto-sound waves (see, for example, [15]).

Despite the qualitative difference, the monotonic instability branches corresponding to different values of the Marangoni number have two common points. One point (Ga, Ra) = (127.90, 160.62) lies in the instability area and presents thus no interest. The second common point (Ga, Ra) = (-219.22, 252.32) corresponds to the boundary between the areas of stabilization and destabilization of conductive state with respect to monotonic long-wave perturbations discussed above.



Fig. 2. Velocity profiles and flow patterns of neutral monotonic perturbations for two instability modes at Ma = 0, (a) Ga = -0.1, (b) Ga = -100, (c) Ga = -370. Velocity profiles are plotted for the cross-sections passing through the centers of the vortices.

3.2 Heating from above

At nonzero Marangoni numbers, the instability of the system heated from above may take place when the lighter fluid is located above the heavier one, which is not observed at Ma = 0 (Fig. 4, grey and thin black lines). The instability has a monotonic type. As Ma increases, the boundary of the instability domain shifts to the left, i.e. instability domain is extended.

4 Instability with respect to perturbations with finite wave numbers

Investigation of the stability of the conductive state to perturbations with finite wave numbers (cellular perturbations) has been carried out numerically using a differential sweep method and a shooting method [16]. Calculation results were used to create full stability maps for different values of the Marangoni number. The stability



Fig. 3. Boundaries of long-wave instability in the system heated from below: thin black lines (a, b, c) – Ma = 0, dark grey lines (a, d) – Ma = -170, bold black lines (b, d) – Ma = -187.1, light grey lines (c, d) – Ma = -200.



Fig. 4. Boundaries of monotonic long-wave instability for the system heated from above; bold black line – Ma = 0, grey line – Ma = 50, thin black line – Ma = 200.

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Fig. 5. Stability maps: (a) Ma = -100, (b) Ma = -200. Black lines correspond to the boundaries of long-wave instability, and grey lines denote cellular instability. Solid lines indicate instability boundaries with respect to monotonic perturbations, and dashed lines show instability boundaries with respect to oscillatory perturbations.

maps presented below show the boundaries of instability to monotonic perturbations (solid lines) and oscillatory perturbations (dashed lines). Grey lines correspond to the boundaries of cellular instability and black ones to the boundaries of longwave instability.

4.1 Heating from below

Instability boundaries with respect to finite-wavenumber perturbations for the system heated from below are shown in Fig. 5 by the grey lines. The results are presented for the two values of the Marangoni number, Ma = -100 (a) and Ma = -200 (b). As one can see, any values of the parameters, the perturbations with a finite wave length are most dangerous. In the case when the denser fluid is above the less dense one, the Rayleigh-Taylor instability is observed, and convection develops monotonically in the form of rolls of finite length. However, it is worth noting that this result holds valid only for the examined system, where the layer thickness is large enough. For thin films in a well-known work by Burgess et al. [17], the long-wave modes are found to be more dangerous.

With the increase of thermocapillary effect the oscillatory cellular perturbations become less dangerous than monotonous ones. In the case of a non-deformable interface, at negative Marangoni numbers large in modulus, the instability threshold decreases with increasing Marangoni number modulus.

Thus, in the case of heating from below, finite-wavelength perturbations are most dangerous. Convection can develop both monotonically for nearly non-deformable interface and oscillatory when the deformations of the interface are essential.

4.2 Heating from above

For heating from above, the evolution of the instability boundaries with the increase of the Marangoni number is shown in Fig. 6. The grey lines correspond to the instability



Fig. 6. Stability map: (a) Ma = 100, (b) Ma = 200. Black lines correspond to the boundaries of long-wave monotonic instability, and grey lines denote cellular monotonic instability.

 Table 1. The wave numbers of most dangerous perturbations for different domains of instability boundaries.

Ga	k	Ma
-200	1.87	0
	$1.89 \\ 1.91$	$-100 \\ -200$
-300	1.73	0
	$1.85 \\ 1.93$	-100 -200

boundaries with respect to cellular perturbations, the instability domains are located above the curves. As one can see comparing Fig. 6a and Fig. 6b, the thermocapillary effect leads to the expansion of the parameter domain, where the most dangerous perturbations are those with a finite wavenumber. Oscillatory perturbations are absent and only monotonic perturbations may occur.

Table 1 presents the numerical data on the wave number of most dangerous perturbations for different domains of instability boundaries. As one can see, the wavelength of most dangerous perturbations is of the order of layer thickness; it slightly decreases with the thermocapillary effect growth.

5 Discussion

We have investigated the system of two horizontal layers of immiscible fluids bounded by two rigid plates. The layers are infinite in the horizontal directions. The fluid interface is deformable. The interaction of two mechanisms responsible for the excitation of convection: due to density changes with temperature (Rayleigh-Benard convection) and surface tension with temperature (Marangoni convection) are considered in the framework of the generalized Boussinesq approximation (Busse-Lyubimov model). The stability of the conductive state to long-wave perturbations was studied analytically using a series expansion with respect to the perturbation wave number. The

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stability maps in the parameter plane Rayleigh number – modified Galileo number at different Marangoni numbers are obtained. It is shown that, as in the absence of the Marangoni effect, the boundary of monotonic instability consists of two branches of a hyperbolic type. As the thermocapillary effect increases, the instability domain is expanded. With respect to oscillatory perturbations, the stabilization of conductive state with increasing Marangoni number is observed: the oscillatory instability boundary shifts to the positive Rayleigh numbers and at strong enough thermocapillary effect the oscillatory instability mode degenerates; the conductive state becomes unstable only with respect to monotonic perturbations.

The work was supported by Russian Scientific Foundation (grant No. 14-01-00090).

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