Microgravity Sci. Technol. (2010) 22:415–423
DOI 10.1007/s12217-010-9214-9

ORIGINAL ARTICLE

Long-Wavelength Marangoni Convection in a Liquid Layer with Insoluble Surfactant: Linear Theory

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Received: 25 January 2010 / Accepted: 1 June 2010 / Published online: 19 June 2010

Abstract The subject of this paper is the long-wave Marangoni convection in a horizontal liquid layer with insoluble surfactant absorbed on the free surface. The surfactant is convected by interfacial velocity field and diffuses over the interface but not into the bulk of the fluid. The layer is subjected to a transverse temperature gradient. The buoyancy effects are negligible as compared to the Marangoni forces. We consider both cases of flat nondeformable and deformable surface. The linear stability analysis of this system is performed. It is shown that in both cases of the upper surface monotonic and oscillatory modes exist. Convection thresholds are determined and the critical Marangoni numbers for monotonic as well as for oscillatory mode are obtained. It is shown that the monotonic long-wave instability is more dangerous than oscillatory one only for small elasticity numbers, if the Lewis number is small.

Keywords Marangoni convection · Instabilities · Surfactants · Thin films

Introduction

There is a significant amount of research done on the Marangoni instability in a pure fluid layer (Davis 1987). This traditional surface-tension-driven convection can be important factor in the no-gravity environment.

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A lot of works are devoted to the Marangoni convection in binary-liquid mixtures where both the temperature and the solute concentration change the value of the surface tension. Unlike the case of pure liquid, an oscillatory Marangoni instability is possible due to the presence of two instability mechanisms related to both temperature and solute concentration gradients (Takashima 1980; Castillo and Velarde 1980). Surface deformation can lead to the emergence of a long-wave instability mode with minimum of neutral stability curve at zero value of the wave number $k = 0$ (Char and Chiang 1996; Bhattacharjee 1994; Oron and Nepomnyashchy 2004). Recently Podolny et al. (2005) developed the theory of long-wave Marangoni instability in a binary-liquid layer with a deformable interface in the limit of small Biot number B and in the presence of the Soret effect.

Another factor that can be important in the stability problems is the influence of surface-active agents on the thermocapillary convection. The surfactant concentrations being small, the convective motion leads to a heterogeneous surfactant distribution on the interface. Owing to the dependence of the surface tension on the surfactant concentration additional tangential stresses arise which have significant influence on convection. The investigation of the Marangoni convection in non-isothermic liquids with adsorbed surface-active agents, insoluble as well as soluble, remains still hardly explored. Palmer and Berg (1972) have considered the case of non-deformable free surface. The stability analysis including surface deformation has be done by Ryabitskii (1993). In the book by Simanovskii and Nepomnyashchy (1993), the problem has been considered for two-layer fluid systems. In all previous works, the temperature of the solid substrate was constant.

In the present paper, we consider the opposite case, where the heat flux is fixed on the solid substrate. On the free surface, the Biot number is assumed to be small (poorly conducting surface). Using the technique of Podolny et al. (2005), we investigate the linear stability problem of long-wave Marangoni convection in a liquid layer in the presence of an insoluble surfactant at a nondeformable interface and a deformable interface. The limit of small Biot number (poorly conducting interface) is studied.

Formulation of the Problem

Let us consider a horizontal liquid layer of thickness H, bounded by a rigid bottom plate located at $z = 0$ and an upper free surface with an ambient gas over it at $z = H$. It is assumed that the z axis is directed vertically upward. Because of the rotational symmetry of the problem, the consideration of two-dimensional disturbances is sufficient for the development of the linear stability theory. The upper surface can be flat and nondeformable or deformable. In both cases the heating is subjected to a transverse temperature gradient, $-a(a > 0)$ and on the bottom plate we have specified heat flux

$$z = 0: \qquad \partial_z T = -a. \tag{1}$$

At the upper free surface the normal heat transfer is governed by the Newton's law of cooling

$$\lambda \partial_{\mathbf{n}} T + qT = 0, \tag{2}$$

where λ is the thermal conductivity of the fluid, $\mathbf{n}$ is the normal unit vector to the surface and q is the rate of heat transfer by convection at the free surface. For the flat nondeformable surface $\partial_{\mathbf{n}} \to \partial_z$. It is assumed that the film is sufficiently thin, so the effect of buoyancy can be neglected. Surfactant with surface concentration $\Gamma(x, t)$ is localized on the interface of finite-thickness liquid layer. Its distribution is changed with time, and it is governed, as described in Stone (1990), by the following expression:

$$\partial_t \Gamma + \nabla_s \cdot (u_\tau \Gamma) + \kappa u_n \Gamma = D_0 \nabla_s^2 \Gamma. \tag{3}$$

Here u_n and u_τ are normal and tangential velocities on the surface, D_0 the surface diffusivity, ∇_s the surface gradient, and κ is the local surface curvature. For the flat nondeformable surface Eq. 3 reduces to

$$\partial_t \Gamma + \partial_x (u\Gamma) = D_0 \partial_{xx} \Gamma, \tag{4}$$

where u is the x-component of the velocity on the flat surface. In the absence of convection, the concentration of surfactant on the interface is constant, $\Gamma = \Gamma_0$, and later it is used as a scale for the surfactant concentration.

The surface tension σ is assumed to depend on both temperature T and surfactant concentration Γ and, therefore, both Marangoni effects are taken into account. We suppose that this dependence is linear:

$$\sigma = \sigma_0 - \sigma_1 T - \sigma_2 \Gamma, \tag{5}$$

where σ_0 is reference value of surface tension, $\sigma_1 = -\partial_T \sigma$, $\sigma_2 = -\partial_\Gamma \sigma$.

First, we present the set of linearized equations in the non-dimensional form. For this purpose we define scales for length, time, velocity, temperature and pressure as H, H^2/χ, χ/H, aH and $\rho\nu\chi/H^2$, respectively (ν, ρ and χ are kinematic viscosity, density and thermal diffusivity). The non-dimensional system of governing equations in the linear approximation is

$$P^{-1} \partial_t \mathbf{v} = -\nabla p + \nabla^2 \mathbf{v}; \quad \nabla \cdot \mathbf{v} = 0;$$
$$\partial_t \theta + w \left(\partial_z T_{eq} \right) = \nabla^2 \theta, \, w = (\mathbf{v} \cdot \mathbf{e}), \quad \mathbf{e} = (0, 1). \tag{6}$$

Here, the small disturbances of velocity, temperature and pressure are denoted by $\mathbf{v} = (u, w)$, θ and p, respectively. $\nabla = (\partial_x, \partial_z)$. T_{eq} is the non-dimensional temperature field corresponding to the motionless mechanical equilibrium state, see Podolny et al. (2005),

$$T_{eq} = -z + \frac{1 + B}{B}. \tag{7}$$

The non-dimensional parameters of the problem (the Prandtl number, P, and the Biot number, B) are defined as

$$P = \nu/\chi, \quad B = qH/\lambda. \tag{8}$$

Eliminating the pressure disturbances, p, the horizontal component of velocity, u, we obtain the following system of governing equations:

$$P^{-1} \left(w_{xxt} + w_{zzt} \right) = w_{xxxx} + 2w_{xxzz} + w_{zzzz},$$
$$\theta_t - w = \theta_{xx} + \theta_{zz}. \tag{9}$$

The linearized non-dimensional boundary conditions at the bottom rigid surface reflect the no-slip condition for the velocity and specified heat flux (Eq. 1), and they are given as

$$z = 0: \quad w = w_z = \theta_z = 0. \tag{10}$$

Conditions at the free upper surface, which depend on its type, are discussed below.

Nondeformable Free Surface

At the free nondeformable surface the boundary conditions are, respectively, the kinematic boundary condition, Newton's cooling law (Eq. 2), surfactant distribution equation (Eq. 4), and the stress balance condition, which can be written in the dimensional form as

$$\eta(\partial_z u + \partial_x w) = -\sigma_1 \partial_x T - \sigma_2 \partial_x \Gamma.$$

The linearized non-dimensional form of these conditions with the scales defined in "Formulation of the Problem"

$$z = 1: \quad w = 0, \quad \theta_z = -B\theta,$$

$$\gamma_t - w_z = L\gamma_{xx},$$

$$w_{xx} - w_{zz} = -M\theta_{xx} - N\gamma_{xx}. \tag{11}$$

Here we eliminated the horizontal velocity component, u, and introduced new non-dimensional parameters: the Lewis number, $L = D_0/\chi$, the Marangoni number, $M = \sigma_1 a H^2/\eta\chi$, and the elasticity number, $N = \sigma_2 H\Gamma_0/\eta\chi$. γ is perturbation function for the surfactant concentration.

Introducing normal perturbations in the form

$$(w, \theta, \gamma) = \exp(ikx + rt)(\hat{w}, \hat{\theta}, \hat{\gamma}), \tag{12}$$

where k and r are, respectively, the dimensionless wave number and growth rate of the disturbance, and substituting Eq. 12 into Eq. 9 and boundary conditions we obtain the following linearized problem (hats are omitted):

$$P^{-1} r\left(w_{zz} - k^2 w\right) = w_{zzzz} - 2k^2 w_{zz} + k^4 w, \tag{13}$$

$$\theta_{zz} - k^2\theta = r\theta - w, \tag{14}$$

$$z = 0: \quad w = w_z = \theta_z = 0,$$

$$z = 1: \quad w = 0, \quad \theta_z = -B\theta,$$

$$r\gamma - w_z = -k^2 L\gamma,$$

$$w_{zz} + k^2 w = -k^2(M\theta + N\gamma). \tag{15}$$

Eliminating γ from the last two conditions we obtain the following one

$$w_{zz} + k^2\left(w^2 + M\theta + \frac{N w_z}{r + k^2 L}\right) = 0. \tag{16}$$

We investigate the long-wavelength instability of the system with poorly conducting boundaries. According to this we introduce the scaling:

$$k = \epsilon K, \quad B = \epsilon^4\beta, \tag{17}$$

where ϵ is a small scaling parameter serving as a measure of supercriticality. The Marangoni number and the growth rate of the disturbances are expanded near the stability threshold as

$$M = M_0 + \epsilon^2 M_2 + \epsilon^4 M_4 + \ldots,$$

$$r = \epsilon^2\left(r_0 + \epsilon^2 r_2 + \epsilon^4 r_4 + \ldots\right). \tag{18}$$

The amplitudes of the perturbation functions are also expanded as

$$\theta = \theta_0 + \epsilon^2\theta_2 + \epsilon^4\theta_4 + \ldots,$$

$$w = \epsilon^2\left(w_0 + \epsilon^2 w_2 + \epsilon^4 w_4 + \ldots\right). \tag{19}$$

Substituting these expansions into the system of equations and boundary conditions, and collecting terms with the identical degrees of ϵ, we obtain equations describing the terms in the asymptotic expansion of the solution of the whole problem.

At the zeroth order of expansion we obtain

$$w_{0,zzzz} = 0, \tag{20}$$

$$\theta_{0,zz} = 0 \tag{21}$$

with conditions

$$z = 0 \quad w_0 = w_{0,z} = \theta_{0,z} = 0,$$

$$z = 1 \quad w_0 = \theta_{0,z} = 0,$$

$$w_{0,zz} + K^2 M_0\theta_0 + K^2\frac{N w_{0,z}}{r_0 + K^2 L} = 0. \tag{22}$$

At this stage we obtain the following solution

$$\theta_0 = \Theta_0, \quad w_0 = \frac{K^2 M_0\left(K^2 L + r_0\right)\Theta_0}{K^2(4L + N) + 4r_0} z^2(1 - z), \tag{23}$$

where Θ_0 is a constant, that we can choose equal to 1. Additionally, we integrate Eq. 14 over the interval $0 \le z \le 1$ and the resulting integral relation serves as solvability condition of the second order problem. In leading order of the approximation this condition looks like

$$K^2 < \theta_0 > + r_0 < \theta_0 > - < w_0 >= 0, \tag{24}$$

$< \cdots >= \int_0^1 \ldots dz$. Integrating and introducing the new rescaled growth rate, $\Lambda_0 = r_0 K^{-2}$, we obtain for it the following expression

$$\Lambda_0 = \frac{1}{96}\{M_0 - 48L - 12N - 48$$

$$\pm\sqrt{192(M_0 L - 48L - 12N) + (48L + 12N + 48 - M_0)^2}\}. \tag{25}$$

This expression shows the existence of two instability modes, the monotonic and the oscillatory, that will be considered separately below.

Monotonic Instability Mode

We first study the case of monotonic instability, hence we assume that at its threshold $\Lambda_0 = 0$. Equation 25 gives the critical value of Marangoni number

$$M_0 = 48 + \frac{12N}{L}. \tag{26}$$

This number is positive if $N > N_1 = -4L$, and negative for $N < N_1$. Recall that because the surfactant diminishes the surface tension, N is positive. Differentiating Eq. 25 with respect to M_0, we find that at the critical Marangoni number

$$\frac{d\Lambda_0}{dM_0} = \frac{L^2}{48L^2 + 12N(L-1)}. \tag{27}$$

Thus, we find that $d\Lambda_0/dM_0$ is positive if $N < N_2 = 4L^2/(1-L)$ and negative if $N > N_2$. That means that the growth rate is negative for M below the critical value M_0 and positive above, i.e., $M = M_0$ is indeed the threshold of the monotonic instability of the equilibrium state, only if $N < N_2$.

To determine the dependence of the growth rate on K, we need the next order of the approximation. At the second order the equations and boundary conditions read:

$$w_{2,zzzz} = \left(P^{-1}r_0 + 2K^2\right) w_{0,zz}, \tag{28}$$

$$\theta_{2,zz} = \left(r_0 + K^2\right)\theta_0 - w_0 \tag{29}$$

and

$$z = 0: \quad w_2 = w_{2,z} = \theta_{2,z} = 0,$$

$$z = 1: \quad w_2 = \theta_{2,z} = 0,$$

$$w_{2,zz} + K^2 M_0 \theta_2 + K^2 \frac{N w_{2,z}}{r_0 + K^2 L} = \frac{K^2 N r_2 w_{0,z}}{\left(K^2 L + r_0\right)^2} - K^2\left(w_0 + M_2\theta_0\right). \tag{30}$$

The solutions of the equation set read

$$\theta_2 = K^2\left(\frac{z^2}{2} - z^4 + 3\frac{z^5}{5}\right),$$

$$w_2 = \frac{12Nr_2\left(z^2 - z^3\right)}{L(4L+N)} + \frac{K^2 L M_2\left(z^2 - z^3\right)}{(4L+N)} + K^4\left\{\frac{4Nz^2 - 8(2L+N)z^3}{5(4L+N)} + 2z^4 - 6\frac{z^5}{5}\right\}. \tag{31}$$

The solvability condition in this order yields

$$K^2 < \theta_2 > + < r_0\theta_2 > + < r_2\theta_0 > - < w_2 > = -\beta\theta_0. \tag{32}$$

This condition yields expression for the growth rate r_2

$$r_2 = \frac{-60(4L+N)L\beta + 5L^2 M_2 K^2 - 16LK^4}{60(4L^2 - N + LN)}. \tag{33}$$

It follows from this expression that the neutral curve has its minimum for $K = O(1)$, i.e. in the longwave region $k = O(\epsilon)$. The shape of the neutral curve is determined by the expression

$$M_2 = \frac{60(4L+N)\beta + 16K^4}{5LK^2}. \tag{34}$$

Figure 1 depicts the typical behavior of M_2 (solid line) for $L = 0.1$, $\beta = 1$ and $N = 0.03$.

Oscillatory Instability Mode

The oscillatory instability boundary is determined by the relation $\Lambda_0 = \pm i\Omega_0$, where the oscillation frequency Ω_0 is real. Using the dispersion relation 25 we find that the oscillatory instability appears for $N > N_2$ at

$$M_{osc} = 48 + 12(4L + N) \tag{35}$$

with the frequency

$$\Omega_0 = \frac{1}{2}\sqrt{N - LN - 4L^2}. \tag{36}$$

The second-order correction for the Marangoni oscillatory number, $M_{2(osc)}$, is given below in the Appendix. Figure 1 reveals the typical form of the $M_{2,osc}$ (dashed line) as function of wave number k at $L = 0.1$, $P = 7$, $\beta = 1$ and $N = 40$.

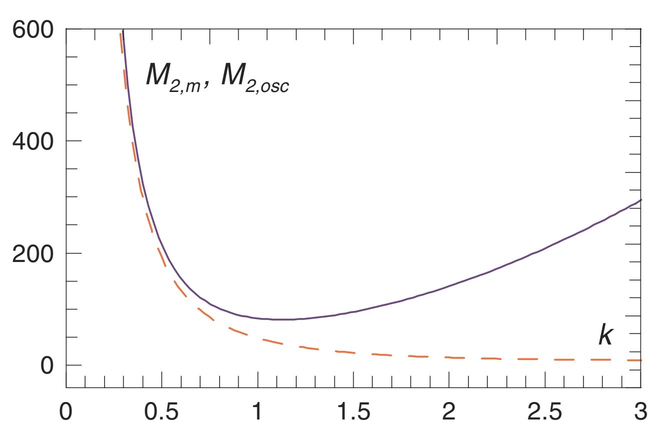

Fig. 1 Typical behavior of the second-order correction for the Marangoni monotonic number $M_{2,m}$ (*solid line*) and the oscillatory number $M_{2,osc}$ (*dashed line*) via the wave number k for the long-wave instability ($L = 0.1$, $\beta = 1$, $P = 7$, $N = 0.03$ and $N = 40$, respectively)

Deformable Free Surface

Here we suppose that the free liquid–gas interface is not flat, and it depends on time and on the horizontal position, i.e. $z = h(x, t)$. At the surface the boundary conditions are, respectively, the kinematic boundary condition, Newton cooling law, balance of both normal and tangential interfacial stresses given as

$$h_t + uh_x = w, \tag{37}$$

$$\lambda \nabla T \cdot \mathbf{n} + qT = 0, \tag{38}$$

$$-p + \frac{2\eta}{\left(1+h_x^2\right)}\left\{u_x\left(h_x^2-1\right) - h_x\left(u_z+w_x\right)\right\} = \frac{\sigma h_{xx}}{\left(1+h_x^2\right)^{3/2}}, \tag{39}$$

$$\frac{\eta}{\left(1+h_x^2\right)^{1/2}}\left\{2\left(w_z-u_x\right)h_x + \left(u_z+w_x\right)\left(1-h_x^2\right)\right\} = \sigma_s. \tag{40}$$

Here $\mathbf{n}$ is the unit outward vector normal to the surface, η is the dynamic viscosity, σ_s is the full derivative of the surface tension with respect to s, arc length along the interface. In addition to these conditions we need to consider Eq. 3 of the surfactant concentration over the deformable surface. The equations describing the dynamics and the heat transfer in the liquid are the same as in "Nondeformable Free Surface".

Now we rewrite the conditions in non-dimensional form and linearizing the normal perturbations of the variables, $(u, w, p, T, \gamma, \zeta) = (\hat{u}, \hat{w}, \hat{p}, \hat{\theta}, \hat{\gamma}, \hat{\zeta})\exp(ikx + rt)$, around the equilibrium state, which is the same as in "Nondeformable Free Surface". Here $\zeta = \zeta(x, t)$ is a deviation of the interface from the flat state $h = 1$.

Reducing the system by eliminating the horizontal component of velocity, the pressure and surfactant concentration disturbances we obtain for the amplitudes the following set of equations with the boundary conditions (hats are omitted):

$$P^{-1}r\left(w_{zz} - k^2 w\right) = w_{zzzz} - 2k^2 w_{zz} + k^4 w, \tag{41}$$

$$\theta_{zz} - k^2\theta = r\theta - w, \tag{42}$$

$$z = 0: \quad w = w_z = \theta_z = 0,$$
$$z = 1: \quad r\zeta = w, \quad \theta_z = -B(\theta - \zeta),$$
$$P^{-1}rw_z + 3k^2 w_z - w_{zzz} + k^2\left(G + \Sigma k^2\right)\zeta = 0,$$
$$w_{zz} + k^2 w = -k^2\left[M(\theta - \zeta) + N\frac{w_z}{(k^2L + r)}\right]. \tag{43}$$

The new dimensionless parameters in this section (the Galileo number, G, and the inverse capillary number, Σ) are defined as

$$G = gH^3/(\nu\chi), \quad \Sigma = \sigma H/(\eta\chi).$$

Here we again investigate the long-wave instability of the system with poorly conducting boundaries, i.e. $B << 1$. We employ the standard scaling for the wave number of disturbances and for the Biot number, both small in our consideration,

$$k = \epsilon K, \quad B = \epsilon^4 \beta \tag{44}$$

(scaling parameter ϵ is a measure of the smallness). Marangoni number, the growth rate of the disturbances, amplitudes of perturbations of velocity and temperature we expand as Eqs. 18 and 19. The deviation of the interface is expanded as

$$\zeta = \zeta_0 + \epsilon^2\zeta_2 + \epsilon^4\zeta_4 + \dots. \tag{45}$$

Like in the "Nondeformable Free Surface" at the zeroth order we obtain the following set of equations:

$$w_{0,zzzz} = 0, \tag{46}$$

$$\theta_{0,zz} = 0 \tag{47}$$

with conditions

$$z = 0: \quad w_0 = w_{0,z} = \theta_{0,z} = 0,$$
$$z = 1: \quad w_0 = r_0\zeta_0, \quad \theta_{0,z} = 0, \quad w_{0,zzz} = K^2 G\zeta_0,$$
$$w_{0,zz} + K^2\left[M_0(\theta_0 - \zeta_0) + \frac{Nw_{0,z}}{(K^2L + r_0)}\right]. \tag{48}$$

The solution is given by

$$\theta_0 = \Theta_0,$$
$$\zeta_0 = \frac{6K^2M_0(K^2L + r_0)\Theta_0}{K^4C_1 - 2K^2C_2r_0 - 12r_0^2},$$
$$w_0 = \frac{K^2G\zeta_0}{6}z^2(z-1) + r_0\zeta_0 z^2,$$
$$C_1 = 6LM_0 - G(4L + N),$$
$$C_2 = 2G + 6L - 3M_0 + 6N. \tag{49}$$

The solvability condition is the same as in "Nondeformable Free Surface" and yields in terms of $\Lambda_0 = r_0K^{-2}$

$$1 + \Lambda_0 = \frac{M_0\left(L + \Lambda_0\right)\left(2\Lambda_0 - \frac{G}{12}\right)}{C_1 - 2C_2\Lambda_0 - 12\Lambda_0^2}. \tag{50}$$

Equation 50 is now employed to investigate the monotonic and oscillatory types of instability.

Monotonic Instability Mode

The solution given by Eqs. 46–48 reduces to

$$\Theta_0 = -\frac{G\zeta_0}{72}, \quad w_0 = \frac{1}{6}GK^2\zeta_0 z^2(z-1).$$

Assuming in Eq. 50 $\Lambda_0 = 0$ we obtain the critical Marangoni number as

$$M_0 = \frac{12G(4L+N)}{L(72+G)}. \tag{51}$$

Below we assume that $G > 0$ (the free surface is on the top of the layer).

At the second order the equations and the boundary conditions read

$$w_{2,zzzz} = \left(2K^2 + P^{-1}r_0\right) w_{0,zz}, \tag{52}$$

$$\theta_{2,zz} = \left(K^2 + r_0\right)\theta_0 - w_0 \tag{53}$$

and

$$z = 0: \quad w_2 = w_{2,z} = \theta_{2,z} = 0,$$

$$z = 1: \quad r_2\zeta_0 + r_0\zeta_2 = w_2, \quad \theta_{2,z} = 0,$$

$$\left(P^{-1}r_0 + 3K^2\right) w_{0,z} - w_{2,zz} + GK^2\zeta_2 + \Sigma K^4\zeta_0 = 0,$$

$$w_{2,zz} + K^2M_0\left(\theta_2 - \zeta_2\right) + \frac{K^2Nw_2}{(K^2L + r_0)}$$
$$= -K^2\left(w_0 + M_2\left(\theta_0 - \zeta_0\right) - \frac{Nr_2w_{0,z}}{\left(K^2L + r_0\right)^2}\right). \tag{54}$$

The solution at second order is given by

$$\theta_2 = \frac{1}{24}GK^2\zeta_0\left(-\frac{z^2}{6} + \frac{z^4}{3} - \frac{z^5}{5}\right), \tag{55}$$

$$w_2 = \frac{\zeta_0}{360G(4L+N)}(K^2z^2(z-1)[-144G\{-5LM_2 + 4K^2(N+6L)\} + 4320\{6LM_2 - K^2(N+4L)\Sigma\} + G^2\{2K^2N[2+z(3z-2)] + L[5M_2 + 8K^2z(3z-2)]\}]\}. \tag{56}$$

Using the solvability condition at the fourth order we obtain

$$-\beta(\theta_0 - \zeta_0) = -< w_2 > +K^2 < \theta_2 >$$

and substituting solutions results in

$$M_2 = \frac{16K^4A_1 + A_2\beta}{5(G+72)^2K^2L}, \tag{57}$$

where $A_1 = G^2L + 36G(N+6L) + 270(N+4L)\Sigma$ and $A_2 = 60G(G+72)(N+4L)$. Thus, the second-order correction M_2 has form of $M_2 = a_1K^2 + a_2K^{-2}$, where a_1 and a_2 are constants. The necessary condition for the long-wavelength instability is $a_1 > 0$ and it takes when

$$N > N_3 = -L\left(4 + \frac{G(G+72)}{36G + 270\Sigma}\right). \tag{58}$$

Note that for all parameters $N_3 < N_1$. In the domain $N > N_1$, $M_0 > 0$, $a_1 > 0$ and $a_2 > 0$ $M = M_0 + \epsilon(a_1K^2 + a_2K^{-2} > 0$ for all K and has a minimum at

$$K^2_{min} = \frac{15^{1/2}\left[G(G+72)(4L+N)\beta\right]^{1/2}}{2\left[G^2L + 36G(N+6L) + 270(N+4L)\Sigma\right]^{1/2}}. \tag{59}$$

Finally we consider the dependence of the rescaled growth rate of disturbances Λ_0 on M. We find that at $M = M_0$ the value

$$\frac{d\Lambda_0}{dM_0} = \frac{B_1}{B_2},$$

$$B_1 = (G+72)^2L^2,$$

$$B_2 = 48L^2(G^2 + 27G + 216) - 12GN(G+72) + 12LN(G^2 + 36G + 864). \tag{60}$$

This value is positive if $N < N_4$, where

$$N_4 = \frac{4L^2(G^2 + 27G + 216)}{G(G+72) - L(G^2 + 36G + 864)}. \tag{61}$$

For real situations of small Lewis number, $L < 1$, N_4 is positive when

$$G > G_c = \left(\frac{6}{L-1}\right)(6 - 3L - \sqrt{36 - 12L - 15L^2}). \tag{62}$$

Oscillatory Instability Mode

Equation 50 gives us the critical Marangoni number as

$$M_0 = \frac{12(1+\Lambda_0)\left[12\Lambda_0(L+\Lambda_0+N) + G(4(L+\Lambda_0)+N)\right]}{(L+\Lambda_0)(G+72+48\Lambda_0)}. \tag{63}$$

Substituting $\Lambda_0 = i\Omega_0$ with real Ω_0 into this expression and looking for real values of M_0 we obtain two neutral

Marangoni numbers, $M_{osc}^{(\pm)}$, corresponding to the oscillatory case with two respective values for the squared neutral frequency

$$\Omega_{\pm}^2 = \frac{1}{288}\{-G^2 + 9G(N-3) - 72[N+3+2L(L+N)] \pm \sqrt{D}\}, \quad (64)$$

where

$$D = -144\left\{4(216+G(G+27))L^2 - G(G+72)N + (864+G(G+36))LN\right\} + \left\{G^2 - 9G(N-3) + 72(N+3+2L(L+N))\right\}^2. \quad (65)$$

Thus, two critical Marangoni numbers are

$$M_{osc}^{(\pm)} = \frac{1}{2(G+72+48L)}\{G^2 + 3G(41+32L+5N) + 72[5N+3+2L(L+N+2)] \mp \sqrt{D}\}. \quad (66)$$

Equations 66 and 64 describe a boundary of oscillatory instability, if M_0 is real and Ω_0^2 is real and positive. Numerical analysis shows that for all small Lewis numbers ($L < 1$), which are physically relevant, $\Omega_-^2 < 0$, i.e. this type of instability does not exist.

Figure 2 shows the typical dependence of the monotonic, M_m, and one of the oscillatory Marangoni number, M_{osc}^+, on the elasticity number, N. Here the Lewis number is small, $L = 0.01$, and the Galileo number equals to 100. As we see from the figure the more dangerous instability at small elasticity numbers is monotonic, but exists critical elasticity number when the oscillatory mode begins to be the most dangerous one. In this example the critical number $N_{cr} = 0.0003$. For all small Lewis numbers ($L < 1$), which are physically relevant, the situation is the same and only one oscillatory branch exists.

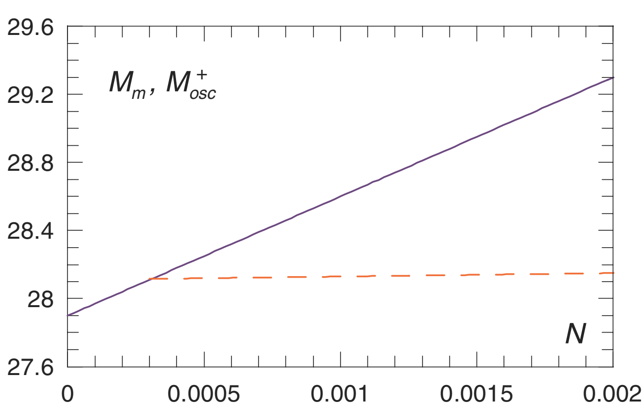

Fig. 2 Monotonic M_m (*solid line*) and oscillatory M_{osc}^+ (*dashed line*) Marangoni numbers via the elasticity number N for $L = 0.01$ and $G = 100$

If the elasticity number is fixed, then as we see from Eq. 51, the critical Marangoni number for monotonic instability grows with increasing the Galileo number and tends to $48 + 12N/L$ for $G \to \infty$ (situation of the flat free surface). A decrease of G corresponding to the increase of interface deformability results in lowering of the stability of the system for both types of the instability. It should be noted that in the limit $G \to \infty$, M_{osc}^+ also tends to the critical Marangoni number for the oscillatory instability found for the nondeformable case.

The stabilizing effect of the surfactant on the monotonic mode is due to the fact that for monotonic disturbances the tangential forces, caused by temperature and surfactant distribution heterogeneity, are of opposite directions. The oscillatory mode is characterized by a phase shift between the oscillations of temperature and surfactant disturbances, which weakens the stabilizing action of surfactant.

Case $k = O(B^{1/2})$

During the analysis of monotonic instability mode in the "Deformable Free Surface" we obtained, that the second-order correction M_2 for the Marangoni number in case of deformable surface has form of $M_2 = a_1 K^2 + a_2 K^{-2}$, where a_1 and a_2 are constants. The values a_1 and a_2 are always positive, because we consider only positive parameters G, N and L. The term $a_2 K^{-2}$ describes the intermediate asymptotics of the neutral curve. In previous sections we considered $k \sim B^{1/4}$, but not $k \ll B^{1/4}$. Thus, it is impossible to take the limit $K \to 0$, because k will then become much smaller than $B^{1/4}$. In order to carry out the full linear stability analysis one her we consider the case when

$$k = \tilde{\epsilon} q, \quad B = \tilde{\epsilon}^2 \tilde{\beta}. \quad (67)$$

Here $\tilde{\epsilon}$ is a new scaling parameter, measure of the smallness. We repeat expansions like in the previous sections.

The same operations as in "Deformable Free Surface" leads to the following relation for the monotonic instability boundary

$$M_{mon} = \frac{12G(4L+N)(q^2+\tilde{\beta})}{(G+72)q^2 L}. \quad (68)$$

In the limit $q \to \infty$ this expression reduces to the result of the case $k \sim B^{1/4}$.

Using solvability condition 14 we obtain for the leading term of the oscillatory Marangoni number

$$M_{osc} = \frac{12(F_1+F_2)(q^2(1+\Lambda_0)+\beta)}{q^2(L+\Lambda_0)(72+G+48\Lambda_0)}$$
$$F_1 = 12\Lambda_0(L+\Lambda_0+N),$$
$$F_2 = G(4(L+\Lambda_0)+N). \quad (69)$$

The oscillatory instability boundary is determining by substituting $\Lambda_0 = i\Omega_0$ in the previous expression and seeking for the real value of M_0. Finally, we obtain critical squared frequencies corresponding to the two branches of the neutral curve

$$\Omega_\pm^2 = -\frac{1}{288q^2}\{q^2(G^2-9G(N-3) + 72(N+3+2L(L+N))) - 9(-24+5G+16N)\beta \pm \sqrt{D_1}\}, \quad (70)$$

where

$$D_1 = -144q^2(-\beta(36(5G-24)L^2+GN(G+72) + 36LN(G-24)) + (4(216+G(G+27))L^2 - GN(G+72) + (864+G(G+36))LN)q^2) + (-9\beta(5G+16N-24) + (G^2-9G(N-3) + 72(3+N+2L(L+N)))q^2)^2. \quad (71)$$

Conclusions

In this paper we performed linear stability analysis of long-wave Marangoni convection in a liquid layer in the presence of an insoluble surfactant at a non-deformable and deformable interfaces. By means of multi-scale method, it was shown that monotonic and oscillatory instability modes exist in both cases. Convection onset thresholds for both modes were determined. The surfactant stabilizes the monotonic instability, which is dangerous only at small elasticity number. However, there exists a critical value of the elasticity number such that above that value an oscillatory instability starts.

Acknowledgements The authors gratefully acknowledge the financial support from the Israeli Ministry of Science (Grant No. 3-5799). The research was partially supported by the European Union via the FP7 Marie Curie scheme [Grant No. PITN-GA-2008-214919 (MULTIFLOW)].

Appendix: Derivation of $M_{2(osc)}$ of the Oscillatory Instability Mode for Nondeformable Free Surface

Introducing the rescaled growth rate $\Lambda_0 = r_0K^{-2}$ into Eq. 23 we rewrite θ_0 and w_0 (here we choose $\Theta_0 = 1$)

$$\theta_0 = 1, \quad w_0 = \frac{M_0K^2(L+\Lambda_0)}{4(L+\Lambda_0)+N}z^2(1-z). \quad (72)$$

Expression 25 is the solution of the following equation for M_0

$$M_0 = \frac{12(1+\Lambda_0)(4L+N+4\Lambda_0)}{(L+\Lambda_0)}. \quad (73)$$

Solving the set of equations for the second-order approximation with boundary conditions 28–30 we obtain

$$\theta_2 = K^2\left(\frac{z^2}{2} - z^4 + 3\frac{z^5}{5}\right)(1+\Lambda_0), \quad (74)$$

$$w_2 = \frac{12Nr_2(1-z)z^2(1+\Lambda_0)}{(L+\Lambda_0)(N+4L+4\Lambda_0)} + K^2\frac{M_2(L+\Lambda_0)(1-z)z^2}{(N+4L+4\Lambda_0)} + K^4\left(c_1z^2+c_2z^3+c_3z^4+c_4z^5\right). \quad (75)$$

Here

$$c_1 = \frac{(1+\Lambda_0)\left[12(2P-1)\Lambda_0(L+\Lambda_0)+N(P(4+6\Lambda_0)-\Lambda_0)\right]}{5P(N+4L+4\Lambda_0)},$$
$$c_2 = \frac{-(1+\Lambda_0)}{5P(N+4L+4\Lambda_0)}\{8(2L+N)P+4(6P-1)\Lambda_0{}^2 + [N-4L+2(8+12L+3N)P]\Lambda_0\},$$
$$c_3 = \frac{(2P+\Lambda_0)(1+\Lambda_0)}{P},$$
$$c_4 = -3\frac{(2P+\Lambda_0)(1+\Lambda_0)}{5P}.$$

Substituting $\Lambda_0 = i\Omega_0$ in θ_2 and w_2 and using solvability condition

$$K^2(1+i\Omega_0) < \theta_2 > +r_2 - < w_2 > +\beta = 0$$

for the second-order corrections to both the real part of the growth rate r_2 and the neutral Marangoni number $M_{2(osc)}$ we obtain

$$Re(r_2) = \frac{1}{480P}\{5K^2PM_2 + K^4[-3L(N+4+4L) + 2(N-8P)+2L(N+4L-4)P] - 240P\beta\},$$
$$M_{2(osc)} = \frac{1}{5K^2P}\{K^4[3L(4+4L+N)-2(N-8P) - 2L(-4+4L+N)P] + 240P\beta\}. \quad (76)$$

The condition for the long-wavelength instability is positivity of the term with K^4 and it takes when

$$N > N_5 = \frac{4(1+L)[3L + 2(2-L)P]}{2 + L(2P-3)}.$$

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