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On dynamic excitation of Marangoni instability in a liquid layer with insoluble surfactant on the deformable surface

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Abstract. This paper is a continuation of our previous work presented at the IMA-6, see [1]. We continue to analyze the parametric excitation of Marangoni instability by a periodic flux modulation in a liquid layer with insoluble surfactant. Contrary to the previous investigation here the upper free surface of the layer is deformable. The linear stability analysis for the disturbances with arbitrary wave numbers is performed. Three response modes of the system to an external periodic stimulation were found – synchronous, subharmonic, and quasi-periodic ones. Results for different Galileo and inverse capillary parameters are presented. It is shown that contrary to the situation with nondeformable interface, at small values of Galileo and inverse capillary parameters a new subharmonic instability region appears in the range of long waves.

1 Introduction

It is known that the influence of external periodic forcing on a physical system can change essentially the behavior of this system. Most often the frequencies of the response on this forcing are related to the driving frequency by an integer multiplier, and these responses are synchronous. However, under certain conditions a response with a frequency less than the driving one can also appear. These responses are known as subharmonics. Later on, we will consider subharmonic responses with frequency proportional to the half of the driving frequency. The response can also be quasi-periodic.

Thus, depending on the frequency and amplitude of the driving force the physical system exhibits a rich variety of different types of behavior. The convective system is only one example of these systems, see [2–4]. It is not so important what kind of

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driving mechanism is applied. It can be gravity modulation, as well as temperature or gradient of temperature modulation [5]. It is important that the considered physical system has its natural frequency, as an ordinary planar pendulum in a well-known Kapitza pendulum problem [6].

The adsorption of insoluble surfactants at the free liquid surface reduces the surface tension. The inhomogenity of surface temperature also may change the surface tension. Both these factors under certain conditions generate oscillations of the liquid in the layer heated from below with absorbed insoluble surfactant on the upper surface. These oscillations have their natural frequency and under external periodic forcing lead the system to its parametric excitation. Our previous investigation of the onset of Marangoni convection in a liquid layer with insoluble surfactant under heat flux modulation [7] and [1], considered the liquid with flat free surface. In the present paper we consider the deformable free surface of the liquid. Section 2 contains formulation of the problem. In the next section numerical method is described. The results of modeling are presented in Sect. 4. The last Section is devoted to concluding remarks.

2 Formulation of the problem

2.1 Basic equations and boundary conditions

Let us consider a horizontally infinite layer of an incompressible liquid, bounded by a rigid lower plane which is located at $z = 0$, and a free deformable upper boundary at $z = h(x, t)$ (the z axis is directed vertically upward). The rotational symmetry of the problem allows us to consider two-dimensional disturbances in the case of linear stability problem. The liquid layer is heated from below with periodically changing in time temperature gradient varying around the mean value $-a$ (for heating from below the value a is positive), while on the upper surface the standard Newton's cooling occurs,

$$z = 0: \qquad T_z = -a + d\cos(2\Omega t), \tag{1}$$

$$z = h(x, t): \qquad \lambda T_z + qT = 0. \tag{2}$$

Here T is the difference between the liquid temperature and the temperature of the ambient gas, d is the amplitude of the heat flux modulation, 2Ω is the external modulation frequency, λ is the thermal conductivity of the liquid, and q is the heat transfer coefficient at the free surface. The subscript denotes the partial derivative with respect to the corresponding variable.

The following set of equations governs the process:

$$u_x + w_z = 0, \tag{3}$$

$$u_t + uu_x + wu_z = -\rho^{-1}p_x + \nu(u_{xx} + u_{zz}), \tag{4}$$

$$w_t + uw_x + ww_z == -\rho^{-1}p_z + \nu(w_{xx} + w_{zz}) - g, \tag{5}$$

$$T_t + uT_x + wT_z = \chi(T_{xx} + T_{zz}). \tag{6}$$

Here u, w are the $x-$ and $z-$ components of velocity field, respectively, ρ is the density of the liquid, p is the pressure, ν and χ are the kinematic viscosity and thermal diffusivity, respectively, t is the time.

On the upper free surface of the liquid an insoluble surfactant is adsorbed. The dynamical distribution of the surfactant concentration, $\Gamma(x,t)$, is described by the following equation [8]:

$$z = h(x,t): \qquad \Gamma_t - h_t(\mathbf{e}_z \cdot \nabla_s)\Gamma + \nabla_s(\mathbf{v}_\tau \Gamma) + (\nabla_s \cdot \mathbf{n})(\mathbf{v}\cdot\mathbf{n})\Gamma = D{\nabla_s}^2\Gamma. \tag{7}$$

Here $\mathbf{e}_z = (0,1)$ is the unity vector directed upward, $\mathbf{n} = (-h_x, 1)(1+h_x^2)^{-1/2}$ is the unity vector normal to the interface, $\nabla_s = \nabla - \mathbf{n}(\mathbf{n}\cdot\nabla)$, $\mathbf{v}_\tau = \mathbf{v} - (\mathbf{v}\cdot\mathbf{n})$, $\nabla = (\partial_x, \partial_z)$, and D is the interfacial diffusion coefficient.

In addition to the above-mentioned boundary condition we have the balance conditions for the normal and tangential stresses on the upper surface, the augmented kinematic condition on the upper surface, the non-slip and non-penetration conditions on the bottom of the layer:

$$z = 0: \qquad u = w = 0, \tag{8}$$

$$z = h(x,t): \quad -p + 2\mu\mathbf{n}\cdot\mathcal{D}\cdot\mathbf{n} + 2\kappa\sigma = 0, \tag{9}$$

$$2\mu\mathbf{n}\cdot\mathcal{D}\cdot\mathbf{t} = \nabla\sigma\cdot\mathbf{t}, \tag{10}$$

$$h_t + uh_x = w. \tag{11}$$

Here $\mathcal{D}$ is the deviatoric stress tensor, κ is the mean interfacial curvature, σ is the surface tension, $\mathbf{t} = (1, h_x)(1+h_x^2)^{-1/2}$ is the unit tangent vector, $\mu = \rho\nu$.

The surface tension is assumed to be a linear function of the temperature and surfactant concentration:

$$\sigma = \sigma_0 - \sigma_1 T - \sigma_2\Gamma,$$

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

2.2 Nondimensionalization and set of equations for disturbances

To formulate the problem in a nondimensional form , the following scales are chosen: length is scaled by H_0 (the mean thickness of the layer), time by H_0^2/χ, velocity χ/H_0, pressure by $\rho\nu\chi/H_0^2$, temperature by aH_0, and the surfactant concentration by Γ_0 (the value for the quiescent state).

Following our previous paper [1] we decompose the base temperature into two components, $T_b = \bar{T}_b + \tilde{T}_b$ ($\bar{T}_b$ is the time-independent average temperature, $\tilde{T}_b$ is the fluctuation around the average temperature). In nondimensional form,

$$\bar{T}_b = -z + \frac{1+B}{B}, \tag{12}$$

$$\tilde{T}_b = \tilde{T}_b^+(z)e^{\alpha^2 t} + \tilde{T}_b^-(z)e^{-\alpha^2 t}, \tag{13}$$

$$\tilde{T}_b^+(z) = a_1 e^{\alpha z} + a_2 e^{-\alpha z}, \qquad \tilde{T}_b^-(z) = (\tilde{T}_b^+)^*, \tag{14}$$

$$a_1 = \frac{(B-\alpha)\delta}{4\alpha e^\alpha[B\cosh(\alpha) + \alpha\sinh(\alpha)]}, \qquad a_2 = \frac{-(B+\alpha)\delta e^\alpha}{4\alpha[B\cosh(\alpha) + \alpha\sinh(\alpha)]}. \tag{15}$$

("*" denotes a complex conjugate expression). Here we have the following parameters, $B = qH/\lambda$ is the Biot number, $2\omega = 2\Omega H^2/\chi$ is the nondimensional frequency of heat flux modulation, $\delta = d/a$ is the nondimensional amplitude of heat flux modulation,

$\alpha = \sqrt{2i\omega}$. The base state values of the surfactant concentration and liquid surface function are $\Gamma_b = 1, h_b = 1$.

The dimensionless form of the linearized equations and boundary conditions for disturbances of the vertical velocity, w, temperature, $\theta(x, z, t) = T(x, z, t) - T_b(z, t)$, surface concentration, $\gamma(x, t) = \Gamma(x, t) - 1$, and free surface function $\zeta(x, t) = h(x, t) - 1$, is (here the pressure, p, and the horizontal velocity component, u, are eliminated from the set):

$$P^{-1}(w_{zzt} + w_{xxt}) = w_{zzzz} + 2w_{xxzz} + w_{xxxx}, \tag{16}$$

$$\theta_t + w(T_b)_z = \theta_{xx} + \theta_{zz}, \tag{17}$$

$$z = 0: \qquad w = w_z = \theta_z = 0, \tag{18}$$

$$z = 1: \qquad w = \zeta_t, \qquad \theta_z + \zeta(T_b)_{zz} + B[\theta + \zeta(T_b)_z] = 0, \tag{19}$$

$$\gamma_t - w_z = L\gamma_{xx}, \tag{20}$$

$$w_{xx} - w_{zz} = -M[\theta_{xx} + \zeta_{xx}(T_b)_z] - N\gamma_{xx}, \tag{21}$$

$$-P^{-1}w_{zt} + w_{zzz} + 3w_{zxx} + G\zeta_{xx} - \Sigma\zeta_{xxxx} = 0. \tag{22}$$

Here we have the following dimensionless parameters: $M = \sigma_1 a H^2/\mu\chi$ is the Marangoni number, $N = \sigma_2 H\Gamma_0/\mu\chi$ is the elasticity number, $L = D/\chi$ is the Lewis number, $G = gH^3/\nu\chi$ is the Galileo number, $\Sigma = \sigma H/\mu\chi$ is the inverse capillary number.

The variables of the problem are decomposed into normal perturbations

$$(w, \theta, \gamma, \zeta) = (\hat{w}(z, t), \hat{\theta}(z, t), \hat{\gamma}(t), \hat{\zeta}(t)) \exp(ikx + rt), \tag{23}$$

where k and r are, respectively, the dimensionless wave number and growth rate of disturbances. The amplitudes $\hat{w}(z, t)$, $\hat{\theta}(z, t)$, $\hat{\gamma}(t)$, and $\hat{\zeta}(t)$ are periodic in time with the period π/ω (later on, the hats are omitted).

Finally, we obtain the following system for investigation:

$$P^{-1}[w_{zzt} + rw_{zz} - k^2 w_t - rk^2 w] = w_{zzzz} - 2k^2 w_{zz} + k^4 w, \tag{24}$$

$$\theta_t + r\theta + w(T_b)_z = \theta_{zz} - k^2\theta, \tag{25}$$

$$z = 0: \quad w = w_z = \theta_z = 0, \tag{26}$$

$$z = 1: \quad w = r\zeta + \zeta_t, \qquad \theta_z + \zeta(T_b)_{zz} + B[\theta + \zeta(T_b)_z] = 0, \tag{27}$$

$$\gamma_t + r\gamma - w_z = -k^2 L\gamma, \tag{28}$$

$$k^2 w + w_{zz} = -Mk^2[\theta + \zeta(T_b)_z] - k^2 N\gamma, \tag{29}$$

$$P^{-1}(w_{zt} + rw_z) - w_{zzz} + 3k^2 w_z + k^2(G + k^2\Sigma)\zeta = 0. \tag{30}$$

This system of equations with boundary conditions defines a spectral problem for the eigenvalue r. The condition $\Re(r) = 0$ determines the critical value of Marangoni number for the parametric excitation of the Marangoni instability.

3 Numerical method

In the systems with time-periodic forcing we can find three types of instability: synchronous, subharmonic, and quasi-periodic. To find the first two modes of instability we assume $r = 0$ and decompose the unknown variables into the sets of harmonic functions as the Fourier expansions

$$w(z,t) = \sum_m w_m(z)e^{im\omega t}, \qquad \theta(z,t) = \sum_m \theta_m(z)e^{im\omega t},$$
$$\gamma(t) = \sum_m \gamma_m e^{im\omega t}, \qquad \zeta(t) = \sum_m \zeta_m e^{im\omega t}, \tag{31}$$

where $m = \pm 1, \pm 3, \ldots$ correspond to the subharmonic instability mode and $m = 0, \pm 2, \pm 4, \ldots$ to the synchronous one. The subharmonic mode has the period which is twice the period of the heat flux forcing, the synchronous one has the same period as the external heat flux. The substitution of (31) into (24)–(30) yields two independent infinite systems for amplitudes w_m, θ_m, γ_m and ζ_m for synchronous and subharmonic disturbances.

Boundary condition (28) leads to the following relationship between amplitudes of surfactant concentration and amplitudes of vertical velocity component:

$$\gamma_m = w_{m,z}/(im\omega + k^2 L). \tag{32}$$

Equation (24) gives an infinite system for amplitudes $w_m(z)$

$$\frac{i\omega m}{P}[w_{m,zz} - k^2 w_m] = w_{m,zzzz} - 2k^2 w_{m,zz} + k^4 w_m. \tag{33}$$

The general solution of these equations contains the coefficients, which can be expressed by ζ_m. Substituting the solution with respect to w_m into (25) we obtain the infinite system of equations for θ_m, its formal solution also contains ζ_m. One obtains two infinite homogeneous systems of linear algebraic equations for coefficients ζ_m. One system is for the synchronous disturbances, another one is for the subharmonic ones. Each system has a nontrivial solution if the characteristic determinant of the system equals zero. The latter condition allows to find the eigenvalues of the problem, the critical values M. The calculations were performed using from 32 up to 512 harmonics for each mode which assured a good accuracy of the analysis. For more detailed information on the method see [4].

To find the quasi-periodic instability mode, we apply the Floquet theory. For that kind of instability, only the real part r_r of the growth rate $r = r_r + ir_i$ vanishes on the stability boundary, $r_r = 0$. The imaginary part, r_i, is defined modulo 2ω. Because the imaginary eigenvalues of system (24)–(30) appear in pairs, $r = ir_r$ and $r^* = -ir_i$, without loss of generality we can assume $0 \le r_i \le \omega$. The case $r_i = 0$ corresponds to the synchronous instability and $r_i = \omega$ to the subharmonic ones.

Before discussing the results of our calculation let us estimate the non-dimensional parameters of the problem. We suppose that typical values of the dimensional physical parameters have the following orders (in SI): kinematic viscosity $\sim 10^{-6}$, density of liquid $\sim 10^3$, thermal diffusivity $\sim 10^{-7}$, surface tension $\sim 10^{-2} - 10^{-1}$, heat transfer coefficient $\sim 10^2$–10^3, surface diffusion coefficient $\sim 10^{-9}$. The problem has a lot of parameters, so we fix some of them. The Lewis number is fixed as $L = 0.01$. It is known, see [9], that low-molecular-weight surfactants in emulsions and foams form monolayers with low elasticity number $N < 1$ and we will fix this number as $N = 0.1$.

4 Results and discussion

We begin the computation with high values of Galileo and inverse capillary parameters, $G = 10^5$, $\Sigma = 10^5$; the modulation frequency, as well as amplitude of the modulation are fixed: $\omega = 0.25$, $\delta = 0.6$. These parameters (G and Σ) are so high, that the liquid surface is flat and neutral curves look like in the case of nondeformable layer.

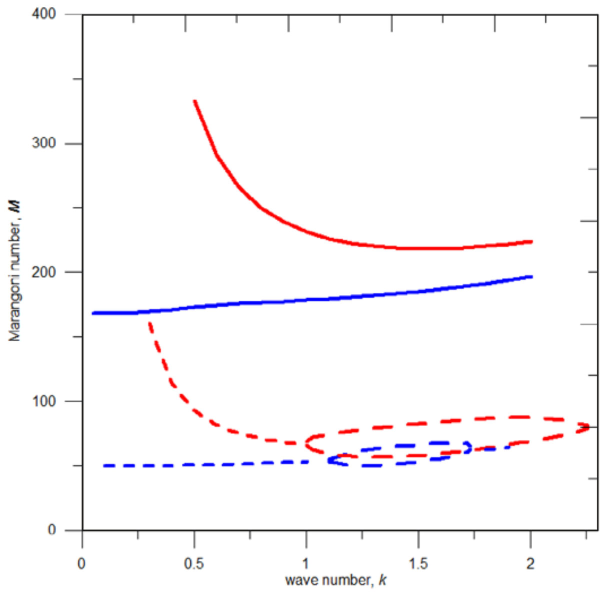

Fig. 1. Neutral curves in the (k, M) – plane for different Biot numbers. Blue color lines are for $B = 0$, red color lines – $B = 0.2$. Solid line is for synchronous mode, dashed one is for subharmonic mode, and short-dotted line for quasi-periodic mode. Parameters of the system $\omega = 0.25$, $\delta = 0.6$, $\Sigma = 10^5$, $G = 10^5$, $P = 7$, $L = 0.01$, $N = 0.1$.

Figure 1 shows typical neutral curves at different values of the Biot number. The blue lines represent the case with poorly conducting upper layer, $B = 0$. The solid line is for synchronous mode, the dashed line is for subharmonic one, and the dotted line is for quasi-periodic instability mode. The computation shows that when the wave number approaches zero the Marangoni number value for the synchronous threshold of instability tends to 168, that exactly coincides with the value obtained for the monotonic mode, $M_m = 48 + 12N/L$, of long-wave instabilities in the absence of modulation [11]. At the same time the quasi-periodic threshold tends to 49.7, which corresponds to a critical Marangoni number for oscillatory instability mode of non-modulated quiescent state, $M_{osc} = 48 + 12(4L + N)$.

Hence, the system at high values of Galileo and inverse capillary parameters behaves similarly to the situation with flat upper surface, see [1]. This system in the absence of heat modulation has two modes of instability–monotonic and oscillatory. When the modulation appears the monotonic neutral curve is transformed to a synchronous one, and the oscillatory into a quasi-periodic one. At small values of the modulation amplitude on the oscillatory neutral curve around the value of k, where the frequency of the neutral oscillatory mode is equal to ω (to half of the frequency of the external modulation), a subharmonic "bubble" appears, similarly to the problem of convective instability of gravity-modulated doubly cross-diffusive fluid layer [10]. This bubble grows as the amplitude modulation parameter δ grows and that causes existence of two minima of the stability curve–one is that related to the quasi-periodic mode, another one to the subharmonic mode.

As Fig. 1 shows the subharmonic bubble grows with the Biot number. The red lines represent the case with fixed value of the Biot parameter, $B = 0.2$. The lines are shifted up when the Biot parameter grows which means that the stability regions increase. This situation was described in detail in our previous work.

Here we consider in detail the situation with small values of Galileo and inverse capillary numbers, $G = 100$ and $\Sigma = 100$, the case when the deformability of the upper

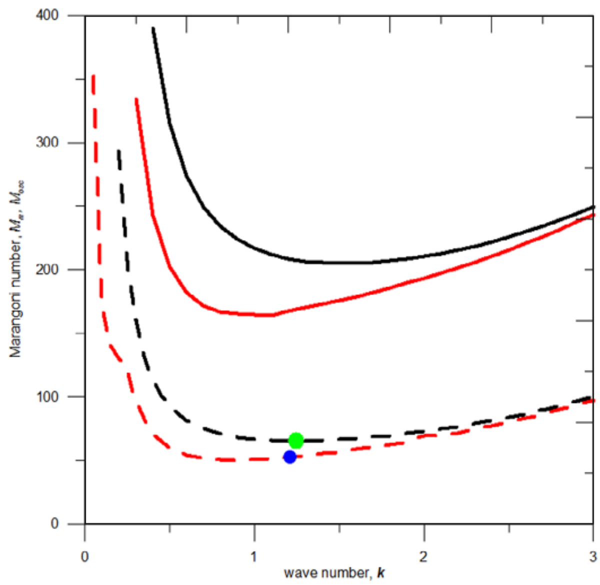

Fig. 2. Neutral curves in the (k, M) – plane for different parameters G and Σ (case without modulation). Black color lines are for $G = 10^5$ and $\Sigma = 10^5$, red color lines are for $G = 100$ and $\Sigma = 100$. Solid line is for synchronous mode, dashed one is for subharmonic mode. Parameters of the system: $B = 0.2, \Sigma = 100, G = 100, P = 7, L = 0.01, N = 0.1$. Points show where subharmonic mode appears.

surface is significant. First, we consider the liquid layer with insoluble surfactant without modulation of the mean temperature flux. In this case our system of Eqs. (16)–(22) coincides with system (41)–(43) of [11] and can be solved analytically using the Mathematica package. The results are shown in Fig. 2, which represents the neutral curves–black color lines are for the case of high-valued parameters, $G = 10^5$ and $\Sigma = 10^5$, red color lines are for the case of small values of parameters, $G = 100$ and $\Sigma = 100$. Solid lines are for the monotonic instability mode, dashed lines are for the oscillatory mode. Other parameters are as follows: $B = 0.2, P = 7, L = 0.01$, and $N = 0.1$. Here the unusual kink on the curve for the oscillatory mode (red color dashed line) is caused by the fast change of the eigenfrequency in that region (as shown in Fig. 3).

To check here the fact, that when the system is excited by external heat flux modulation the subharmonic bubble appears at the wave number where driving frequency equals the double eigenfrequency of the system, we plot the dependence of the eigenfrequency of the system on the wave number of disturbances. Figure 3 shows this dependence for both cases, the red solid line is for the case with $G = 10^5$, $\Sigma = 10^5$ and the black dashed line is for the case with $G = 100$, $\Sigma = 100$.

From Fig. 3 we can see that for disturbances with wave numbers $k \geq 0.5$ the eigenvalues of the system for both cases are very close one to another. For example, for the eigenfrequency of the system, $\omega_0 = 0.25$ the oscillatory instability mode appears at wave number $k = 1.21$ in the case with $G = 100$ and $\Sigma = 100$, and at $k = 1.25$ for the case with $G = 10^5$, $\Sigma = 10^5$. Numerical solution of the full system (24)–(30) shows that even at small value of δ parameter (ratio of amplitude of the external modulation to the mean temperature flux), $\delta = 0.001$, the subharmonic bubble appears, starting as "a germ point" at small δ parameter. The Marangoni numbers, respectively, are $M = 52.7$ for $k = 1.21$ and $M = 65.5$ for $k = 1.25$. As it might be seen in Fig. 2, the first point, blue color, lies on the red oscillatory line and another point, green color,

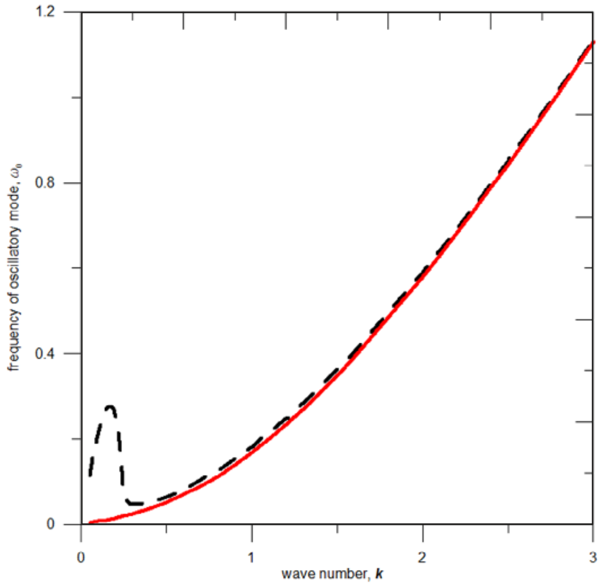

Fig. 3. Dependence of eigenfrequency of the system on wave number of disturbances (situation without modulation). Red solid line is for $G = 10^5$ and $\Sigma = 10^5$, black line is for $G = 100$ and $\Sigma = 100$. Other parameters of the system $B = 0.2, P = 7, L = 0.01, N = 0.1$.

lies on the black oscillatory line. Exactly this situation was observed in our previous investigation with flat surface [1].

However, as Fig. 3 shows we can obtain additional subharmonic regions in the region of long wavelengths at small values of modulation driving frequency. For example, for the driving frequency $\omega = 0.2$ in addition to the known "germ point" of subharmonical response of the system at $k = 1.06$ and $M = 51.1$ we have two additional points in the long-wavelength region, at $k = 0.09$ with $M = 180.4$ and at $k = 0.22$ with $M = 128.7$, respectively. These values of the Marangoni number are obtained analytically solving the system of equations without modulation.

The numerical calculation of the system with modulation where driving frequency $\omega = 0.2$ and $\delta = 0.01$ gives us these two additional subharmonic regions, red color bubbles on Fig. 4. When the δ parameter increases, the subharmonic regions also grows. On the same Fig. 4 the blue color region represents subharmonical instability region at $\delta = 0.1$, situation when two bubbles merge into one.

From Fig. 3 we can conclude that at modulation frequency greater than $\omega > 0.27$ the system will have only one subharmonic region as in the case of big values of G and Σ (non-deformable surface). However, the calculation done for the system at $\omega = 0.3$, as well as at $\omega = 0.4$ and $\omega = 0.5$ reveals the existence of subharmonic regions at long wavelengths. The difference between these cases is in the fact that this subharmonic region appears at values of the δ parameter greater than some critical value, δ_c (non-equal to zero). Figure 5 shows the situation when the driving frequency $\omega = 0.4$ (black color region) and $\delta = 0.1$. For the comparison the situation with $\omega = 0.2$ at the same value of δ is depicted by blue color. The calculation shows that the subharmonical region for $\omega = 0.4$ in the region of long-wavelengths can not be obtained numerically with the number of harmonics in Fourier series equal to 128, as well as to 256. In the case when driving frequency equals 0.5, $\delta_c = 0.14$.

If we extrapolate these three situations at $\omega = 0.3, 0.4$ and 0.5 to the non-modulated problem, localizing the "germ points" of the appearing subharmonic

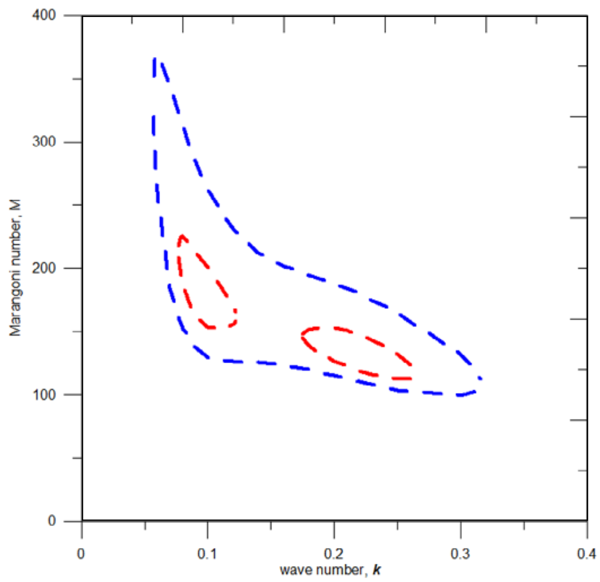

Fig. 4. Subharmonic regions in the long wave range. Red color bubbles are the subharmonic regions at $\delta = 0.01$ and blue color region corresponds to the case with $\delta = 0.1$. Other parameters of the system $\omega = 0.2, B = 0.2, \Sigma = 100, G = 100, P = 7, L = 0.01, N = 0.1$.

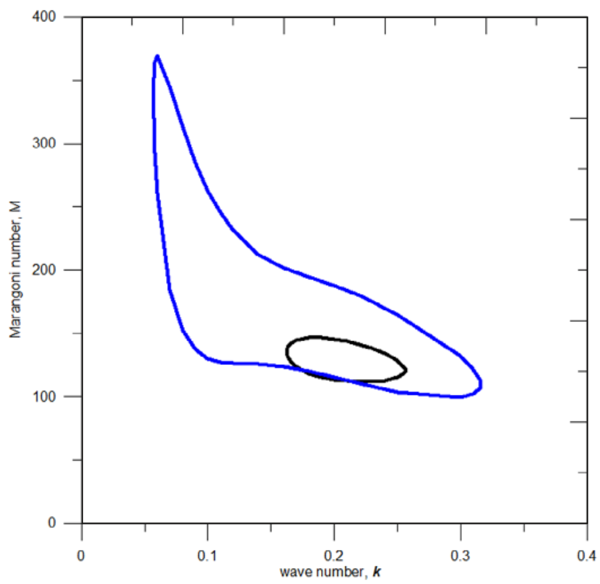

Fig. 5. Subharmonic regions in the range of long waves. Blue color region is for $\omega = 0.2$ and black color region is for $\omega = 0.4$. Both cases at $\delta = 0.1$. Other parameters of the system $B = 0.2, \Sigma = 100, G = 100, P = 7, L = 0.01, N = 0.1$.

regions, and plot these points on the graph of the eigenfrequency of the system, Fig. 3, we obtain Fig. 6. This Figure shows that all these three points lie on the same line which is the continuation of one of branches of the eigenfrequency graph. The red point corresponds to the driving frequency $\omega = 0.3$, the brown point – to the $\omega = 0.4$ and the purple point – to the $\omega = 0.5$.

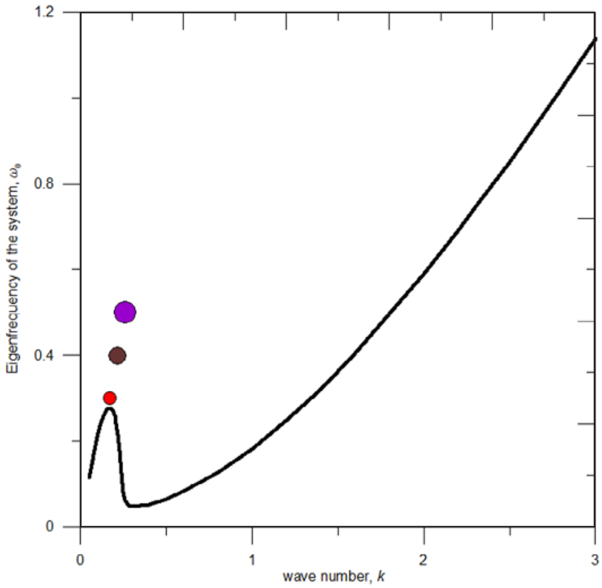

Fig. 6. Dependence of the eigenfrequency of the system on the wave number with extrapolated points. Here the red color point is for $\omega = 0.3$, the brown point is for $\omega = 0.4$ and the purple point is for $\omega = 0.5$. Other parameters of the system $B = 0.2$, $\Sigma = 100$, $G = 100$, $P = 7$, $L = 0.01$, $N = 0.1$.

The subharmonic regions for greater values of wave number, $k > 0.5$, behave as described in our previous paper [1].

Here on the Figures the quasi-periodic modes are omitted, because they coincide with oscillatory mode in the non-modulated case.

5 Conclusions

In this paper we have considered the parametric excitation of Marangoni convection by periodic heat flux modulation in a liquid layer with the insoluble surfactant absorbed on a deformable free surface. It is shown that at high-valued Galileo and inverse capillary numbers the behavior of this system is similar to the system with the insoluble surfactant on non-deformable surface.

We can summarize the behavior of the system as follows. In the case without external modulation the map of instability looks as Fig. 2. It contains three zones on the (k, M) – map. The first zone below the the dashed line is a stable zone. Any disturbances here disappear and dimension of the unstable manifold in this region equals 0. The second zone is above the upper solid line. This is zone of monotonic instability. Here the dimension of unstable manifold is equal to 1. Between the lower dashed line and the upper solid line we have the zone of oscillatory instability. This instability region is caused by competition of two mechanism, thermocapillary and solutocapillary, that changes the surface tension gradient. This region has the dimension of unstable manifold equals to 2. So, the third zone represents the parameters of the most unstable region. When we have external driving forcing on the oscillation line, it is transformed to the so called "quasi-periodic" line, and the subharmonic bubble appears where the dimension of the unstable manifold equals 1. This region, see for example Fig. 1, grows when the driving frequency increases as well as the ratio of the amplitude of the external forcing to the mean value of the temperature gradient increases. The appeared subharmonic bubble has a trend to go upward when the driving frequency increases and this means the system becomes more stable.

When the values of Galileo and inverse capillary numbers are small and the role of deformability in the system is significant, then in the range of long-wavelengths additional subharmonic region appears, that grows when the driving frequency increases.

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