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OSCILLATORY CONVECTION INSTABILITIES IN SYSTEMS WITH AN INTERFACE

A. NEPOMNYASHCHY¹ and I. SIMANOVSKII²

¹Department of Mathematics, Technion–Israel Institute of Technology, Haifa 32000, Israel ²Ha-Jarden 46-6, Ramat-Gan, 52333, Israel

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Abstract—On the basis of the adjoint approach, different types of oscillatory mechanisms of instabilities in two-layer systems are investigated. It is shown that for Marangoni convection oscillatory instability may become the only possible mechanism of instability in the system. The evolution of non-linear regimes of oscillations is analysed. For different aspect ratios different kinds of bifurcations, including period doubling bifurcation and transition to the steady state through a homoclinic bifurcation, are observed. The combined action of Marangoni and Rayleigh mechanisms of instabilities can lead to oscillations even in the case where "pure" Marangoni and "pure" Rayleigh instabilities are stationary. It is shown that the presence of the surface active agents may lead to the specific types of oscillations: the frequency of oscillations is proportional to the wave number in the long-wave limit. Oscillatory Marangoni convection in systems with a deformable interface is investigated.

Key Words: convection, instabilities, oscillations, interface

1. INTRODUCTION

It is well known that when heating a one-layer system with a free surface from below one observes stationary convection cells generated by buoyancy (Rayleigh-Benard convection) and/or thermocapillary effects (Marangoni-Benard convection). However, it was known already in the 1950s (Sternling & Scriven 1959), that systems of fluids with an interface (especially two-layer systems) are subject to oscillatory instabilities, too. From a mathematical point of view, such an instability is a consequence of the non-selfadjointness of the linearized stability problem. The physical nature of oscillatory instabilities is, however, quite diverse.

In the present paper, we summarize some theoretical predictions concerning different kinds of oscillatory instabilities in order to facilitate their recognition.

2. FORMULATION OF THE PROBLEM

Let the space between two horizontal solid plates be filled by two immiscible viscous fluids. The plates are kept at different constant temperatures; the full temperature difference is θ . Two variants of heating—from below and from above—are considered. The coefficient of the surface tension is the linear function of temperature: $\sigma = \sigma_0 \pm \alpha T$. Except section 7, the interface is assumed to be flat (y = 0). All variables referring to the upper fluid, which occupies the region $0 < y < a_1$, are marked by subscript 1, and the variables referring to the lower fluid, $a_2 < y < 0$, are marked by subscript 2. The densities, coefficients of dynamic and kinematic viscosities, heat conductivities, temperature diffusivities and heat expansion coefficients are respectively equal to ρ_i , η_i , ν_i , κ_i , χ_i , β_i , i = 1, 2. The lateral boundaries of both layers x = 0 and x = l are rigid and well conducting.

We introduce the notation $\rho = \rho_1 \rho_2$, $\eta = \eta_1/\eta_2$, $v = v_1/v_2$, $\kappa = \kappa_1/\kappa_2$, $\chi = \chi_1/\chi_2$, $\beta = \beta_1/\beta_2$, $L = l/a_1$, $a = a_2/a_1$. As the units of length, time, the stream function, velocity and the temperature we choose, respectively, a_1 , a_1^2/v_1 , v_1 , v_1/a_1 and θ .

We write the complete non-linear equations of free convection for the stream function ψ_i , the vorticity ϕ_i and the temperature T_i in the following dimensionless form:

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$$\frac{\partial \phi_i}{\partial t} + \frac{\partial \psi_i}{\partial y} \frac{\partial \phi_i}{\partial x} - \frac{\partial \psi_i}{\partial x} \frac{\partial \phi_i}{\partial y} = d_i \Delta \phi_i + G b_i \frac{\partial T_i}{\partial x},$$

$$\Delta \psi_i = -\phi_i, G = \frac{g \beta_1 \theta a_1^3}{v_1^2}$$

$$\frac{\partial T_i}{\partial t} + \frac{\partial \psi_i}{\partial y} \frac{\partial T_i}{\partial x} - \frac{\partial \psi_i}{\partial x} \frac{\partial T_i}{\partial y} = \frac{c_i}{P} \Delta T_i, P = \frac{v_1}{\chi_1}$$

$$d_1 = b_1 = c_1 = 1, d_2 = 1/v, b_2 = 1/\beta, c_2 = 1/\chi;$$
[1]

G is the Grashof number and P is the Prandtl number (i = 1, 2).

The conditions satisfied at the interface are the vanishing of the normal components of velocities and the continuity conditions of the tangential components of velocities, the tangential stresses, the temperatures and the heat fluxes:

$$y = 0: \psi_1 = \psi_2 = 0, \frac{\partial \psi_1}{\partial y} = \frac{\partial \psi_2}{\partial y},$$

$$T_1 = T_2, \kappa \frac{\partial T_1}{\partial y} = \frac{\partial T_2}{\partial y},$$

$$\phi_2 = \eta \phi_1 + Mr \frac{\partial T_1}{\partial x},$$

$$Mr = \frac{\eta M}{P}, M = \frac{\alpha \theta a_1}{\eta_1 \chi_1}.$$
[2]

where Mr is the analogue of the Marangoni number M.

Here we do not explicitly write conditions on rigid boundaries y = 1, y = -a, x = 0 and x = L, which are obvious.

The boundary-value problem ([1] and [2]) is determined by eight physical (G, P, Mr, η , ν , κ , χ , β) and two geometrical (L, a) parameters. This problem has a solution corresponding to the mechanical equilibrium state which is characterized by the absence of any motion and constant vertical temperature gradients $A_1 = -s/(1 + \kappa a)$; $A_2 = -s\kappa/(1 + \kappa a)$ (s = 1 for heating from below and s = -1 for heating from above). The instability of this stage generates the thermocapillary convection. As a first step, one considers the growth of infinitesimally small disturbances of the stream function ψ_i and temperature T_i on the background of the equilibrium temperature gradients. The linearized convection equations take the form

$$(\lambda + i\omega)D\psi_i = -d_i D^2 \psi_i + ik \, Gb_i T_i,$$

$$-(\lambda + i\omega)T_i - ik\psi_i A_i = \frac{c_i}{P}DT_i,$$

$$D = \frac{d^2}{dy^2} - k^2,$$
[3]

k is the wave number, $\lambda + i\omega$ is the complex decay rate.

Then conditions at the solid boundaries and at the interface are:

$$y = 1: \psi_{1} = \psi_{1}' = T_{1} = 0,$$

$$y = -a: \psi_{2} = \psi_{2}' = T_{2} = 0,$$

$$y = 0: \psi_{1} = \psi_{2} = 0, \psi_{1}' = \psi_{2}',$$

$$T_{1} = T_{2}, \kappa T_{1}' = T_{2}',$$

$$\eta \psi_{1}'' - ik \operatorname{Mr} T_{1} = \psi_{2}''.$$
[4]

Depending on the ratio of the Marangoni number and the Grashof number, the thermocapillary or thermogravitational mechanism of instability plays the dominant role.

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3. THERMOGRAVITATIONAL CONVECTION

In the case of Rayleigh instability ($G \neq 0$, M = 0), the oscillations can arise in two-layer systems in the situation where the instability conditions are satisfied simultaneously in both layers. This kind of instability is connected with the interaction between convection motions in both layers and has nothing to do with the gravity-capillary waves, although it can generate some passive deformations of the interface.

Let us introduce the "local" Rayleigh numbers, characterizing the stability conditions in each layer.

$$\mathbf{R}_1 = \frac{\boldsymbol{g}\beta_1 A_1 a_1^4}{v_1 \chi_1}, \quad \mathbf{R}_2 = \frac{\boldsymbol{g}\beta_2 A_2 a_2^4}{v_2 \chi_2}.$$

If these Rayleigh numbers differ considerably, the intensive convective motion arises only in one fluid; in the second fluid the weak induced motion is present (Simanovskii 1979). If R_1 and R_2 are close, heat and hydrodynamic interactions on the interface play the dominating role. The work of Busse (1981) was the first where the situation with close values of the local Rayleigh numbers was considered. The author constructed two stationary neutral curves, corresponding to the arising of convection in each layer, the minima of which lie at considerably different values of the wave numbers. In the work of Gershuni & Zhukhovitsky (1982) it was shown that, in contrast to the one-layer case, the two-layer system may become unstable to the oscillatory disturbances (a physical example is the transformer oil–formic acid system). An example where the oscillatory neutral curve becomes the most "dangerous" is presented in figures 1 and 2 (Gilev *et al.* 1987).

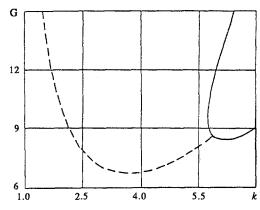


Figure 1. Neutral curves for the model system ($\eta = 0.123$, $\nu = 15.408$, $\kappa = 0.41$, $\chi = 0.714$, $\beta = 0.672$, P = 306.32, a = 0.54).

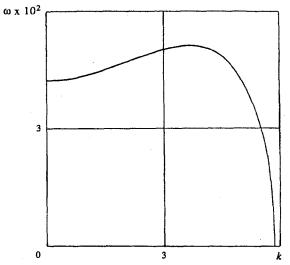


Figure 2. The dependence of the frequency of neutral oscillatory disturbances on wave number.

The linearized boundary-value problem ([3] and [4]) was solved by the Runge-Kutta method. The oscillation region occupies the interval $|k| < k_m \simeq 5.8$. The neutral curve minimum is realized at $k = k_c \simeq 3.8$ for oscillatory disturbances with frequency $\omega_c \simeq 0.051$.

Further, Rasenat *et al.* (1989) returned to investigation of the situation considered by Busse (1981) and obtained the oscillatory neutral curve. In the recent work of Colinet & Legros (1994), the existence of the oscillatory instability was confirmed: in the non-linear region the authors found the travelling wave, going to the left or to the right.

We shall not discuss in detail here the numerous attempts to consider the active influence of the interface deformation at the onset of convection (see for example, Benguria & Depassier 1987; Wahal & Bose 1988; Benguria & Depassier 1989 etc.). Let us note only that an oscillatory instability, essentially connected with deformations, was discovered in the case of small differences between fluid densities ($|\rho - 1| \ll 1$) by Renardy & Joseph (1985).

4. THERMOCAPILLARY CONVECTION (LINEAR THEORY)

There are several types of oscillatory instability in the case of the Marangoni convection (G = 0, $M \neq 0$). The first type, discovered by Sternling & Scriven (1959), does not need an interface deformation and generates longitudinal temperature waves. The original theory of Sternling & Scriven was developed in the case of two layers of infinite depths. As a matter of fact, it deals with the short-wavelength limit of the neutral curve, and cannot describe the most dangerous disturbances, which have wavelengths of the same order as the layer depth. Typically, the oscillatory instability is replaced by a stationary one in the case of finite depths of layers (Reichenbach & Linde 1981; Nepomnyashchy & Simanovskii 1983c). However, longitudinal Marangoni waves can be found in several fluid systems.

As an example, let us consider the transformer oil-formic acid system. The dependences of sMr and ω on k are shown in figure 3.

For $k < k^*$ the stationary mode of instability occurs for heating from below, and for $k < k^*$ the stationary mode of instability occurs for heating from above (line 1). In the long-wave region the oscillatory mode of instability appears (line 2). In the final point of the oscillatory neutral curve on the monotonic one, frequency ω vanishes. In the long-wave limit frequency is constant. The more exotic situation can be found for the model system: oscillatory instability becomes the only possible mechanism of instability in the system (Nepomnyashschy & Simanovskii 1983c), see figure 4.

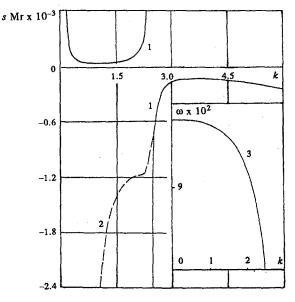


Figure 3. Transformer oil-formic acid system (a = 2). Monotonic (1) and oscillatory (2) neutral curves; frequency ω dependence on wave number k (3).

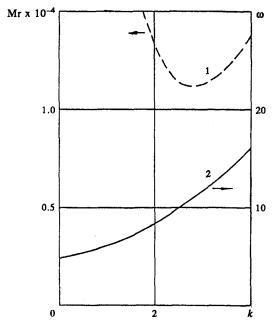


Figure 4. Oscillatory neutral curve (1) and frequency ω dependence on wave number k (2) for the system with parameters $\eta = v = 0.5$; $\kappa = \chi = P = a = 1$.

5. DEVELOPED REGIMES OF THERMOCAPILLARY CONVECTION

Let us now consider the non-linear convection regimes for the previous model system filling up the closed cavity (a = 1, L = 2.5) (Nepomnyashchy & Simanovskii 1983a). The non-linear boundary-value problem [1] and [2] was solved by the finite-difference method [a computational procedure was developed by Simanovskii (1979)]. At small values of Marangoni number the system maintains the equilibrium state: in the subcritical region ($Mr < Mr^* \simeq 15700$), initial four-vortex disturbance decreases in an oscillatory manner. With the increase in the Marangoni number the equilibrium state becomes unstable and regular convective oscillations develop in the system. For $Mr < Mr^*$ we shall characterize the intensity of motion by the variables

$$S_1(t) = \int_0^{L/2} dx \int_0^1 dy \,\psi_1(x, y, t),$$

$$S_2(t) = \int_{L/2}^L dx \int_0^1 dy \,\psi_1(x, y, t).$$

The motion is of a four-vortex structure symmetrical about the vertical axis x = L/2 and for it $S_1 = -S_2$. The dependence of $S_1(t)$ on different values of Marangoni number is shown in figure 5.

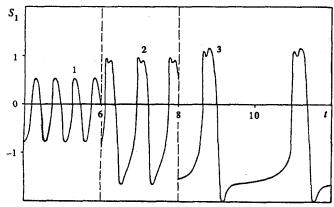


Figure 5. Thermocapillary oscillation forms for $Mr = 1.8 \times 10^4$ (line 1); 2.25×10^4 (2); 2.4×10^4 (3); L = 2.5.

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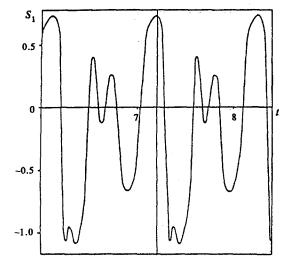


Figure 6. Thermocapillary oscillation forms for $Mr = 2.5 \times 10^4$; L = 2.

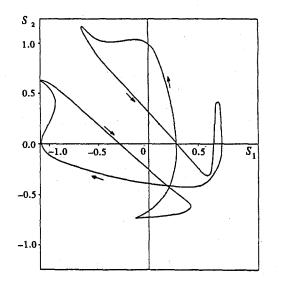


Figure 7. Phase trajectory for thermocapillary oscillations (Mr = 2.5×10^4 ; L = 2).

Close to the threshold the oscillations are nearly sinusoidal (line 1). With the increase in Mr the oscillations become essentially non-linear, their period grows (lines 2 and 3) and steady motion is further established [the oscillatory cycle is transformed into the separatrix of a saddle point (saddle-node)].

A more complicated sequence of transitions is observed for L = 2. As for the case L = 2.5 an oscillatory motion arises as a result of the instability of the equilibrium. With the increase in Mr, there is a doubling bifurcation of the period. The dependence of $S_1(t)$ takes on a more complicated nature (see figure 6). The typical phase trajectory is shown in figure 7. A further increase in Mr leads to the establishment of steady state through homoclinic bifurcation.

6. THE COMBINED ACTION OF THE MARANGONI AND RAYLEIGH MECHANISM OF INSTABILITY

The oscillatory instability is much more typical under the combined action of the thermogravitational and thermocapillary mechanisms of instability. Let us consider the water-silicon oil DC N 200 system (P = 6.28, $\eta = 0.915$, $\nu = 1.16$, $\kappa = 0.169$, $\chi = 0.472$, $\alpha = 7.16$, a = 1.6) (Gilev *et al.* 1987b). When Mr = 0 the threshold Grashof number for the excitation of convection in the upper

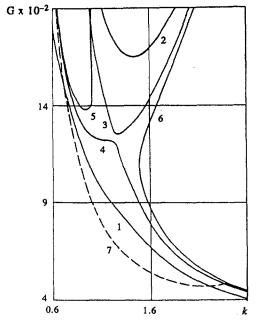


Figure 8. Neutral curves for a water-silicone oil No. 200 system (a = 1.6).

fluid $G_1 = 270$ (figure 8, line 1), while for excitation in the lower fluid $G_2 = 2860$ (line 3); both instability modes are monotonic. As Mr increases, the neutral curves approach each other (curves 4 and 5; Mr = 201). Subsequently, they hook and separate into disconnected parts, lying in the long-wave and short-wave regions (curves 6 and 7, respectively; Mr = 210).

At Mr > 125 an oscillatory interval appears on the neutral curve for disturbances in the upper fluid. It is preserved after hook up of the monotonic neutral curves, linking the long-wave and short-wave fragments (line 2 in figure 8, Mr = 210). At Mr > 400 the oscillatory instability becomes the more dangerous.

7. THE INFLUENCE OF SURFACE ACTIVE AGENTS ON THERMOCAPILLARY CONVECTION

Let the surface active agents (SAA) that lower the surface tension be concentrated at the interface. We assume that the concentration of SAA is low, so that its molecules form a "surface gas". The transport of SAA along the interface is described by the equation

$$\frac{\partial\Gamma}{\partial t} + \frac{\partial}{x} \left(v_x \Gamma \right) + \frac{\partial}{\partial y} \left(v_y \Gamma \right) = D_0 \left[\frac{\partial^2 \Gamma}{\partial x^2} + \frac{\partial^2 \Gamma}{\partial y^2} \right],$$
[5]

where v_x and v_y are the horizontal components of velocity at the interface and D_0 is the surface diffusion coefficient. At equilibrium the SAA concentration at the interface is constant: $\Gamma = \Gamma_0$. The linearized condition for tangential stresses at the interface takes the form

$$\psi_1'' - ik (\operatorname{Mr} T_1 + B\Gamma) = \psi_2'',$$

$$B = -\frac{\partial \omega}{\partial \Gamma} \frac{\Gamma_0 a_1}{\eta_2 v_1}.$$
[6]

The appearance of parameter B is stipulated by the presence of SAA.

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After having been made dimensionless and linearized [5] takes the form

$$(\lambda + i\omega - D_{s}k^{2})\Gamma = ik\psi_{1}'(0), D_{s} = D_{0}/v_{1}.$$
[7]

Eliminating Γ from [6] and [7] we obtain the condition on the interface:

$$y = 0: \eta \psi_1'' - ik \left(\operatorname{Mr} T_1 + \frac{ikB}{\lambda - D_s k^2 + i\omega} \psi_1' \right) = \psi_2''.$$
[8]

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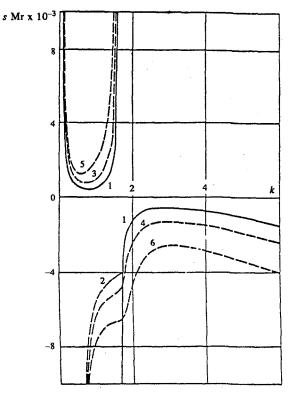


Figure 9. Neutral curves for values of parameter B = 0 (lines, 1, 2); 2 (3, 4); 8 (5, 6); a = 2.

In the case where B = 0 the boundary-value problems [3], [4] and [8] were solved by Nepomnyashchy & Simanovskii (1985). For the water-DC N 200 system (a = 2, $D_s = 10^{-3}$) the graph of function $s \operatorname{Mr}(k)$ for the monotonic mode has discontinuity (figure 9, line 1); in the long-wave region where heating from above the instability is oscillatory in character (figure 9, line 2).

With heating from below (sMr > 0), the presence of SAA leads to the splitting of the monotonic neutral curves to a monotonic one and an oscillatory one (Nepomnyashchy & Simanovskii 1989). The monotonic neutral curves lie in the region of higher values of Mr and are not shown in the graph. For heating from above (sMr > 0), with an increase in *B* the final point of the oscillatory neutral curve on the monotonic curve is shifted towards higher values of *k*. Attention should be drawn to the difference between the long-wave asymptotic form of frequency ω for oscillations caused by SAA (it can be shown analytically that ω is of the wave number order) and that for pure thermocapillary oscillations with B = 0 (ω is constant). Note, that we also found very similar oscillations generated by SAA for Rayleigh instability and for a specific non-Rayleigh type of instability ("anticonvection") when heating from above (see Simanovskii & Nepomnyashchy 1993).

Unlike the oscillations described in previous sections, which arise only in two-layer systems, the SAA-induced oscillations also exist in one-layer systems (Palmer & Berg 1972).

8. SYSTEMS WITH A DEFORMABLE INTERFACE

In the case of Marangoni convection the transverse oscillatory instability is also possible. The instability generates waves of the surface deformation. For one-layer systems, this type of instability was considered by Garcia-Ybarra & Velarde (1987) and investigated in detail in a series of papers (see, for example, Chu & Valarde 1989; Henneberg *et al.*, 1992 and references therein). An attempt to develop a non-linear theory can be found in the papers of Garazo & Velarde (1991) and Nepomnyashchy & Velarde (1994). The analysis of the two-layer problem was fulfilled by Nepomnyashchy & Simanovskii (1991).

On the interface we impose the conditions for normal and tangential stresses, the conditions of continuity of the velocity vector, temperature and the normal component of the heat flux, and the kinematic condition relating the deviation of the boundary h and the velocity of liquids on the

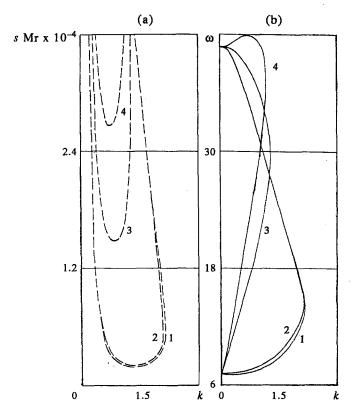


Figure 10. Dependence of the parameter sMr (a) and of the frequency ω (b) on the wave number k (Ga = 0, curve 1; 10⁴, 2; 10⁶, 3; 2 × 10⁶, 4).

interface. As a result of transfer to the plane y = 0 the conditions on the deformed interface take the form:

$$p_{1} - p_{2} + [Ga(\rho^{-1} - 1) + Wk^{2}]h = 2(v_{1y}' - \eta^{-1}v_{2y}')$$
$$\eta(v_{1x}' + ikv_{1y}) - (v_{2x}' + ikv_{2y}) - ik \operatorname{Mr} T_{1} - \left(\frac{s}{1 + \kappa a}h\right) = 0,$$
$$v_{1x} = v_{2x}, -(\lambda + i\omega)h = v_{1y} = v_{2y}$$
$$T_{1} - T_{2} = \left(\frac{s(1 - \kappa)}{1 + \kappa a}\right)h, \kappa T_{1}' - T_{2}' = 0,$$
$$W = \sigma_{0}a_{1}/\eta_{1}v_{1},$$

 $Ga = ga_1^3/v_1^2$ is the Galileo number.

Since the effect of deformation of the interface is assumed to be greatest in the case of similar liquid densities, we will consider a model system with $\rho = 0.999$. The other parameters of the system are as follows: $\chi = a = W = 1$, v = 0.5. The deformation of the interface in this system may lead to the onset of oscillatory instability but even in the absence of deformation the mechanical equilibrium is absolutely stable. The neutral curve is pocket-shaped [figure 10(a)], i.e. to any value of the wave number on a certain interval $k < k_1$ (Ga) there are two values of s Mr. The dependence of the oscillation frequency ω on the wave number k on both branches of the neutral curve is shown in figure 10(b). The threshold value of Mr tends to infinity as Ga $\rightarrow \infty$ which indicates that this mode of instability is essentially related to the deformation of the interface.

In conclusion, let us mention a new direction for investigation: convective instabilities in multilayer systems. The longitudinal oscillatory instability has been found, which is connected with the interaction of thermocapillary motions generated by both interfaces, and is much more typical than in the two-layer case (Georis *et al.* 1993; Simanovskii *et al.* 1993). Very recently, we have found a new type of long-wavelength transfer oscillatory instability which is produced by a hybridization

of two long-wavelength stationary modes. In the long-wave limit, frequency of these oscillations is proportional to the squared wave number.

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