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Non-linear Marangoni Convection in a Layer of Finite Depth.

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Abstract. – Non-linear thermal convection driven by surface tension in a thin layer of fluid heated from below is studied. The present analysis, based on the amplitude method, amplifies previous results obtained by other authors. The thin fluid layer is modelized by means of a finite-depth layer instead of a semi-infinite one, as proposed by Scanlon and Segel. The main differences with Scanlon and Segel analysis are emphasized.

The present note concerns instabilities in fluid layers induced by variations of the surface tension with temperature. This effect is dominant in very thin layer and (or) in a microgravity environment where buoyancy effects are negligible. Very few works on non-linear Marangoni convection have been done in the past. A first non-linear approach of the problem was proposed a few years ago by Scanlon and Segel [1] and Scanlon [2]. However, their work left some questions unanswered, as these authors modelled a thin fluid layer by a layer of semi-infinite depth. Another non-linear analysis was proposed by Kraska and Sani [3] but their results were contested by Rosenblat, Davis and Homsy [4]. Starting from a different point of view, these authors studied the influence of side walls on Bénard-Marangoni convection; however, they examined only the occurrence of rolls and excluded *a priori* the possibility for hexagonal cells to appear. Another non-linear approach was proposed by Cloot and Lebon [5] who used the Malkus and Veronis technique [6]. In contrast, the present analysis will be based on the amplitude method developed by Stuart [7] and Segel and Stuart [8]. Our objective is to answer the following four questions.

i) How far are the Scanlon and Segel amplitude equations modified by introducing a finite-depth layer?

ii) Are the convective cells keeping the same form as predicted by Scanlon and Segel?

iii) Is the direction of circulation in the cells found by Scanlon and Segel correct?

iv) Is the subcritical domain of instability widely modified by considering a finite-depth layer?

Consider a fluid layer of infinite horizontal (x, y) extent bounded below $(z = 0)$ by a rigid plane and above $(z = d)$ by a free flat surface with a temperature-dependent surface tension σ whose equation of state is $\sigma = \sigma_0 - \gamma(T - T_0)$, where σ_0 is the surface tension at temperature T_0, γ the constant rate of change of surface tension with temperature (γ is positive for most current liquids). The layer is submitted to a vertical temperature gradient, in the reference state the fluid is at rest and heat propagates only by conduction.

The fluid is Newtonian, incompressible and the Boussinesq approximation is taken for granted. It is convenient to express the variables in dimensionless form: distances are scaled by the thickness d of the layer, velocity $\boldsymbol{v}$ (u, v, w), time t, temperature T and surface tension are scaled by κd^{-1}, κd^{-2}, $\Delta T = T_0 - T_d$ and σ_0, respectively, T_0 is the temperature at the lower boundary and T_d at the upper one, κ is the heat diffusivity. The usual Prandtl and Marangoni numbers are also introduced:

$$Pr = \nu/\kappa\,, \qquad Ma = \gamma\,\Delta T\,d/\mu\kappa\,;$$

ν and μ stand for the kinematic and dynamic viscosities, respectively. Within Boussinesq's approximation and large values of the Prandtl number, the governing dimensionless equations for the perturbations of the quiescent conductive state are [1,2]

$$\nabla^2 T + w = D_t T\,, \tag{1}$$

$$\nabla^4 w = 0\,, \tag{2}$$

$$\nabla_1^2 u = -\,w_{,xz}\,, \qquad \nabla_1^2 v = -\,w_{,yz}\,, \tag{3}$$

where $\nabla = (\partial_x, \partial_y, \partial_z)$, $\nabla_1^2 = \partial_{xx}^2 + \partial_{yy}^2$, $D_t = \partial_t + v_i \partial_{x_i}$, a comma followed by a subscript denotes derivation with respect to the corresponding space derivative. Assume that the lower boundary ($z = 0$) is rigid and perfectly heat conducting, while the upper surface ($z = 1$) is free, subject to a temperature-dependent surface tension and adiabatically isolated. The relevant boundary conditions for w and T are then given by

$$\text{i) at } z = 0\text{:} \quad w = w_{,z} = T = 0\,, \tag{4}$$

$$\text{ii) at } z = 1\text{:} \quad w = w_{,zz} - Ma\,\nabla_1^2 T = T_{,z} = 0\,. \tag{5}$$

Following Scanlon and Segel [1], this non-linear problem is solved by means of an iterative process. For simplicity, introduce the following differential operators:

$$\boldsymbol{L} = \begin{bmatrix} \nabla^4 & 0 & 0 \\ 1 & \nabla^2 & 0 \\ \partial_{zz}^2\,|_{z=1} & 0 & 0 \end{bmatrix}, \qquad \boldsymbol{N} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & D_t & 0 \\ 0 & 0 & 0 \end{bmatrix}, \qquad \boldsymbol{M} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \nabla_1^2 \end{bmatrix}.$$

In terms of these operators, the set of equations reads in symbolic form

$$\boldsymbol{L}(\boldsymbol{u}) = \boldsymbol{N}(\boldsymbol{u}) + Ma\boldsymbol{M}(\boldsymbol{u})\,, \tag{6}$$

where $\boldsymbol{u}$ is the perturbation vector with components

$$\boldsymbol{u} = [w(x, y, z, t),\, T(x, y, z, t),\, T|_{z=1}(x, y, t)]\,. \tag{7}$$

For further purpose, it is also necessary to define the scalar product of two vectors by

$$\langle \boldsymbol{a}, \boldsymbol{b} \rangle = \lim_{L \to \infty} \frac{1}{4L^2} \int_{-L}^{+L} \int_{-L}^{+L} \mathrm{d}x\,\mathrm{d}y \left[a_3 b_3 + \int_0^1 \mathrm{d}z (a_1 b_1 + a_2 b_2) \right], \tag{8}$$

where L represents the horizontal extent. To ensure the convergence of the iterative process, expression (6) will be rewritten in the following form where the right-hand side contains only small terms:

$$[\boldsymbol{L} - Ma^{c}\boldsymbol{M}](\boldsymbol{u}^{(N)}) = [\boldsymbol{N} + (Ma - Ma^{c})\boldsymbol{M}](\boldsymbol{u}^{(N-1)}), \tag{9}$$

Ma^{c} is the critical Marangoni number obtained from the linear theory. Defining the distance to the threshold by $\varepsilon = (Ma - Ma^{c})/Ma^{c}$, it is expected that convergence will be ensured for sufficiently small values of ε.

The first step in the resolution of the set (9) consists in calculating the linear solution, which corresponds to $N = 1$. The solution is supposed to be written as

$$\boldsymbol{u} = [W_1(z), T_1(z), T_1(z = 1)]\,\Phi(x, y, t), \tag{10}$$

where the form function Φ is solution of the Helmholtz equation

$$\nabla_1^2 \Phi + a^2 \Phi = 0, \tag{11}$$

with a the dimensionless wave number. The expressions of $W_1(z)$ and $T_1(z)$ were derived by Pearson [9]:

$$W_1(z) = C\{[1 + (a \operatorname{ctgh} a - 1)z]\sinh az - az\cosh az\}, \tag{12}$$

$$T_1(z) = C\left\{\sinh az\left[\frac{1}{4a^2} + \frac{\operatorname{ctgh} a}{4a} + \frac{\operatorname{tgh} a}{4a^3} + \frac{a\operatorname{ctgh} a - 1}{4a^2}z + \frac{z^2}{4}\right] - \right.$$
$$\left. - \cosh az\left[\frac{3}{4a}z + \frac{a\operatorname{ctgh} a - 1}{4a}z^2\right]\right\}. \tag{13}$$

C is an arbitrary constant to be taken equal to one in second-order developments. Solutions (12), (13) were calculated by assuming exchange of stability; this property was proved to be correct by Vidal and Acrivos [10]. The stability marginal curve is given by the relation

$$Ma = \frac{8a^2(\cosh a \sinh a - a)\cosh a}{\sinh^3 a - a^3 \cosh a}. \tag{14}$$

The critical Marangoni number and the critical wave number are obtained by minimizing (14) with respect to a; it is deduced that

$$Ma^{c} = 79.607, \qquad a^{c} = 1.993. \tag{15}$$

The linear theory does not predict the shape of the convective cells which experimentally usually take the form of hexagons or rolls. To determine the geometry of the pattern, one needs the explicit expression of the form function supposed to contain two horizontal space modes with two unknown amplitudes depending only on time, namely

$$\Phi(x, y, t) = Z(t)\cos ay + Y(t)\cos(\sqrt{3}/2)\,ax\cos(1/2)\,ay. \tag{16}$$

The convective pattern possesses a hexagonal symmetry when the amplitudes verify the relation: $Y = \pm 2Z$, while for $Y = 0$ and $Z = \text{const}$, it corresponds to a roll symmetry.

The second-order solution of (9) is obtained by setting $N = 2$ and integrating expression (9). The integrability of this system is insured by the Fredholm condition stating that

$$\langle \boldsymbol{u}^*, [\boldsymbol{N} + (Ma - Ma^{c})\boldsymbol{M}](\boldsymbol{u}^{(1)})\rangle = 0, \tag{17}$$

where $\boldsymbol{u}^*$ is the solution of the linear adjoint problem. This condition leads to two amplitude equations containing first- and second-order terms in Y and Z. The second-order solution and a new application of the Fredholm alternative yield the two amplitude equations for $Y(t)$ and $Z(t)$ up to third order. After rather lengthy calculations, it was found that

$$\dot{Y} = L\varepsilon Y - \gamma YZ - RY^3 - PYZ^2, \tag{18}$$

$$\dot{Z} = L\varepsilon Z - (\gamma/4)Y^2 - R_1 Z^3 - (P/2)Y^2 Z, \tag{19}$$

with coefficient values

$$L = 5.9972, \quad \gamma = -0.20079, \quad R_1 = 0.0789, \quad P = 0.110475, \quad R = (P + R_1)/4 = 0.04735.$$

The analysis of the two differential equations (18), (19) is well known [8] and we recall here just the essential points. The main problem consists in finding the fixed point—nine in the present problem—and to analyse their linear stability. Each fixed point has a physical meaning and represents either a conductive state, roll cells, hexagonal cells or hybrid cells. According to the value taken by the parameter ε, these points are locally stable or unstable.

The results of the present analysis are summarized in table I:

TABLE I. – *Stable configurations according to the ε-values.*

$\varepsilon = (Ma - Ma^c)/Ma^c$	stable configurations
$\varepsilon < \varepsilon_c$	conductive satate
$\varepsilon_c < \varepsilon < 0$	conductive state, hexagons (subcritical range)
$0 < \varepsilon < \varepsilon_1$	hexagons
$\varepsilon_1 < \varepsilon < \varepsilon_2$	hexagons, rolls
$\varepsilon > \varepsilon_2$	rolls

The values of the constants ε_c, ε_1 and ε_2 are, respectively,

$$\varepsilon_c = -0.0056, \quad \varepsilon_1 = 0.53, \quad \varepsilon_2 = 1.8. \tag{20}$$

For ε less than ε_c, the layer remains at rest and heat propagates by conduction, while for $\varepsilon_c < \varepsilon < 0$, a subcritical region is displayed where hexagons coexist with the static state. Increasing ε between 0 and ε_1 shows the presence of hexagonal cells; in the interval $\varepsilon_1 < \varepsilon < \varepsilon_2$ both hexagons and rolls are stable, the observed configuration depending on the initial value of the amplitudes. For still larger values of ε ($> \varepsilon_2$), only roll patterns are stable. These conclusions are qualitatively similar to those drawn by Scanlon and Segel [1] and Scanlon [2] who modelled the fluid layer by an unrealistic semi-infinite layer. It appears that by modifying the depth of the layer, one does not change the hierarchy of the transitions between hexagons and rolls as exhibited by table I. However, it should be mentioned that by repeating Scanlon and Segel's calculations [1] for an infinite layer, we have found different values for the constants ε_c, ε_1, ε_2, namely

$$\varepsilon_c = -0.0216, \quad \varepsilon_1 = 7.8, \quad \varepsilon_2 = 25, \tag{21}$$

instead of Scanlon and Segel's original results

$$\varepsilon_c = -0.023\,, \qquad \varepsilon_1 = 64\,, \qquad \varepsilon_2 = 196\,. \tag{22}$$

Clearly, there is a large discrepancy between the values of these parameters and the corresponding values found in the case of a finite-depth layer. Moreover, by starting from Scanlon and Segel's model, we have found that in hexagonal cells the fluid moves downwards at the centre of the hexagon, in contradiction with experiments. For a finite-depth layer, stable hexagons are those with upward flow at the centre of the cells. It should also be noticed that the present analysis predicts a smaller subcritical region than for the infinite-depth layer (0.56% *vs.* 2.16%). This is not surprising since the bottom conductive plate has a stabilizing effect. In that respect, it is found that our results agree with those of Cloot and Lebon [5] who obtained a subcritical range of 0.3% for $Pr = 7$.

The above calculations rest on an iterative procedure requiring that ε remains finite but smaller than one; this means that the result predicting rolls for ε greater than 1.8 must be regarded with caution. In contrast, our analysis shows without any doubt that hexagons appear when the conductive state loses its stability. Far from this first bifurcation, our results are only qualitative but appear to have received experimental confirmations. It was indeed shown by Cerisier *et al.* [11] that in rectangular boxes of small thickness, hexagonal cells are the only stable configuration for ε-values lower than 0.45. Coexistence of rolls and hexagons are observed by Cerisier at relatively large thickness (> 2 cm) when buoyant effects become significant. Other experimental investigations by Koschmieder and Prahl [12] confirm also our results, as it was found that hexagonal-type cells are the preferred stable patterns at not too high values of ε. Koschmieder and Prahl also displayed the presence of a subcritical region.

Recent theoretical works by Bestehorn and Pérez-García [13] and Pérez-García *et al.* [14], based on a generalized Ginzburg-Landau equation, suggest that the occurrence of two different stable patterns, like rolls and hexagons, is typical of fluids whose transport coefficients are highly temperature dependent. This condition is met in the present work, since the surface tension is taken as temperature dependent. It should however be added that our results are only in qualitative agreement with Bestehorn and Pérez-García analyses as they found possible coexistence of rolls and hexagons for ε_1 larger than 0.04, while our calculations predict that ε_1 should exceed 0.53. This difference is not surprising as Bestehorn and Pérez-García worked in a rather general context and did not particularize their study to the special case of Marangoni instability.

To summarize and answering the questions raised at the beginning of this note, it can be stated:

1) with the exception of ε_c, the values ε_1 and ε_2 predicted by the present analysis (20) are appreciably modified compared with the ε_1 and ε_2 values (22) obtained for an infinite depth; it is found that ε_1 and ε_2 in (21) are smaller by a factor of the order of 15;

2) the convective cells keep the same form as predicted by Scanlon and Segel with the same hierarchy exhibited by table I;

3) the direction of the motion inside the cells is now found in agreement with experiments;

4) the subcritical domain is greatly reduced (more or less four times) in the case of a finite-depth layer.

Experimental observations do not only exhibit hexagons but also more complicated patterns like heptagons and pentagons. The occurrence of such kind of cells is certainly due to the lateral boundaries not included in our analysis. Another unsolved problem is the wave number selection beyond the critical threshold. A more reliable analysis needs to take into account a finite band of wave number as done by Newell and Whitehead [15] for the Bénard problem with free-free boundaries. Clearly the problems concerning finite-dimension containers and non-linear wave number selection remain open.

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