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Marangoni convection induced by a nonlinear temperature-dependent surface tension

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Résumé. — On étudie l'instabilité de Marangoni dans une mince lame horizontale de fluide lorsque la tension de surface est une fonction non linéaire de la température. Un tel comportement est typique de solutions aqueuses d'alcools à longue chaîne. La zone des solutions stationnaires convectives est déterminée en fonction du nombre d'onde et d'un nouveau nombre sans dimension, le nombre de Marangoni du second ordre. On montre que les cellules prenant la forme de rouleaux et de rectangles sont instables alors que les hexagones sont stables. Les équations de champ sont exprimées sous forme d'équations d'Euler-Lagrange d'un principe variationnel qui constitue le point de départ de la procédure numérique, basée sur la méthode de Rayleigh-Ritz.

Abstract. — Marangoni instability in a thin horizontal fluid layer exhibiting a nonlinear dependence of the surface-tension with respect to the temperature is studied. This behaviour is typical of some aqueous long chain alcohol solutions. The band of allowed steady convective solutions is determined as a function of the wavenumber and a new dimensionless number, called the second order Marangoni number. We show that the cells which take the shape of rolls and rectangles are unstable while hexagonal planforms remain allowed. The field equations are expressed as Euler-Lagrange equations of a variational principle which serves as the starting point of the numerical procedure, based on the Rayleigh-Ritz method.

1. Introduction.

In most papers on Marangoni instability, the surface-tension is supposed to be a monotonically linearly decreasing function of temperature [1-4]. This behaviour is typical of a large class of fluids like water, silicone oil, water-benzene solutions, etc. In exceptional cases, one may find systems like some alloys, molten salts or liquid crystals with a surface-tension growing linearly with temperature [5-7]. There exists however a third class of fluid systems characterized by a surface-tension exhibiting a nonlinear dependence with respect to temperature and passing through a minimum (see Fig. 1). This behaviour is representative of aqueous long chain alcohol solutions and some binary metallic alloys [8-9].

The purpose of this paper is to study the effect of a nonlinear temperature dependence of the surface-tension on the convective motion observed in a thin horizontal fluid layer subject to heating (Marangoni problem). Buoyancy forces are neglected, which is a reasonable hypothesis in a microgravity environment \(10^{-6} \text{ g to } 10^{-3} \text{ g}\).

Our analysis departs from previous works in two respects. First, it is out of question to treat the problem within the linear normal mode technique. Second, the classical Marangoni number which measures the linear variation of the surface-tension with temperature is meaningless; it is replaced by a new dimensionless quantity expressing the curvature of the surface-tension-temperature curve.

The numerical treatment to be used is close to the procedure proposed in earlier papers [3, 4]. It consists of replacing the set of field equations by a variational principle. Approximate solutions are then obtained by appealing to Rayleigh-Ritz's technique which involves the construction of trial functions, selected as Tchebyshev's polynomials.

The mathematical model is presented in section 2, with special emphasis on the boundary conditions. In section 3, steady convective solutions are derived. As it appears that the number of mathematical steady solutions is infinite, it is necessary to determine which ones are physically admissible. This is achieved by examining the stability of the solutions with respect to infinitesimally small superimposed perturbations (section 4). Final comments are found in section 5.
2. The mathematical model

The system consists of a thin horizontal fluid layer of thickness $d$, extending laterally to infinity and subject to a temperature gradient. The lower face is in contact with a perfectly rigid heat conductor while the upper surface is free, adiabatically insulated, flat and undeformable. The surface-tension $\xi$ exerted at the upper boundary is supposed to be a quadratic function of the temperature, with a minimum value $\xi_m$ at $T = T_m$:

$$\xi(T) = \xi_m + \frac{b}{2}(T - T_m)^2,$$

(2.1)

$\xi_m$ and $b$ are given positive parameters. Law (2.1) is well representative of aqueous alcohol solutions, as observed in figure 1 which shows the $\xi(T)$ curves for a n-heptanol-water solution for various concentrations: $b$ is of the order of $10^{-6}$ N/m$^2$ while $\xi_m$ ranges from $3 \times 10^{-2}$ to $7 \times 10^{-2}$ N/m.

It is also assumed that the fluid is Boussinesquian with constant values of density $\rho$, heat diffusivity $\kappa$, kinematic viscosity $v$, and no viscous dissipation.

Let $\Delta T$ be the temperature drop between the lower and upper boundaries of the layer. In absence of gravity effects, the velocity $u(u, v, w)$ and temperature $\theta$ fields satisfy the following balance equations:

$$\nabla \cdot u = 0 \quad \text{(balance of mass)}, \quad (2.2)$$

$$Pr^{-1}(\partial_t + u \cdot \nabla)u = -\nabla p + \nabla^2 u \quad \text{(balance of momentum)}, \quad (2.3)$$

(\partial_t + u \cdot \nabla)\theta = \nabla^2 \theta + w \quad \text{(balance of energy)}, \quad (2.4)$$

where

$$\nabla \equiv (\partial_x, \partial_y, \partial_z), \quad \nabla^2 = \partial_x^2 + \partial_y^2 = \partial_x^2 + \partial_y^2 + \partial_z^2.$$

Cartesian coordinates are selected with horizontal axes located in the lower boundary and a vertical axis pointing upwards; equations (2.2) to (2.4) are written in non-dimensional form with the space coordinates, time, and temperature scaled by $d$, $d^2/\kappa$, $\Delta T$ respectively, $Pr$ is the usual Prandtl number given by

$$Pr = \frac{v}{\kappa}.$$

The relevant boundary conditions are:

at $z = 0$ (lower rigid, heat conducting):

$$w = \theta = 0, \quad (2.5)$$

at $z = 1$ (upper free, adiabatically insulated):

$$w = \partial_\theta = 0, \quad (2.6)$$

$$\partial^2_\theta = -M[(\nabla \cdot \nabla)^2 + \theta \nabla^2 \theta + f \nabla^2 \theta], \quad (2.7)$$

where $M$ and $f$ stand respectively for

$$M = \frac{(\partial^2 \xi/\partial T^2)(\Delta T)^2 d}{\rho v \kappa} > 0 \quad (M \neq 0), \quad (2.8)$$

$$f = (T_1 - T_m)/\Delta T. \quad (2.9)$$

For positive values of $\Delta T$, $f$ is positive, zero or negative according to whether the temperature $T_1$ at the upper surface is larger, equal or smaller than $T_m$, the temperature at which $\xi$ is a minimum. The quantity $M$ is strictly positive and is called the Marangoni number of second order: it is related to the inverse of the radius of curvature of the $\xi - T$ curve, it takes values between 100 and 1000 for aqueous alcohols when $\Delta T$ is of the order of a few degrees and $d$ about one centimeter.

It is worth comparing (2.7) with the analogous boundary condition formulated in the classical Marangoni problem and expressed by

$$\partial^2_\theta = Ma \nabla^2 \theta, \quad (2.10)$$

$Ma$ is the Marangoni number defined by

$$Ma = -\frac{(\partial^2 \xi/\partial T) \Delta T d}{\rho v \kappa}. \quad (2.11)$$

In contrast with (2.10), expression (2.7) is no longer linear; moreover, the central quantity ceases to be the Marangoni number but rather $M$, a strictly positive quantity whereas $Ma$ may be either positive or negative. When $f \gg 1$, it is a good approximation to drop all the nonlinear terms in (2.7), which reduces...
then to (2.10) upon writing

$$- Mf \equiv Ma. \quad (2.12)$$

On the other hand, for small values of \( f \), which means that \( T_1 \) is close to \( T_m \), the nonlinearity is dominant.


Our objective is to determine the steady solutions of the eigenvalue problem set up by equations (2.2) to (2.7). Since it is desired to focus on the boundary effects, we shall here neglect the nonlinear terms \( u \cdot \nabla u \) and \( u \cdot \nabla \theta \) appearing in (2.3) and (2.4): the validity of this approximation will be discussed at the end of section 5.

After eliminating the pressure, expressions (2.3) and (2.4) reduce to

$$\nabla^4 w_{ss} = 0 \quad (0 < z < 1), \quad (3.1)$$

$$\nabla^2 \theta_{ss} + w_{ss} = 0 \quad (0 < z < 1), \quad (3.2)$$

where the subscript \( ss \) refers to the steady solution. The corresponding boundary conditions are still given by (2.5)-(2.7) where all the quantities are affected by the subscript \( ss \).

In view of the numerical solutions, it is convenient to replace the relations (3.1), (3.2) and the associated boundary conditions by the variational equation

$$\delta I(w_{ss}, \theta_{ss}) = \frac{1}{2} \int_V \left[ (\nabla^2 w_{ss})^2 + (\nabla \theta_{ss})^2 \right] dV -$$

$$- \int_V w_{ss} \delta \theta_{ss} dV + M \int_{S_1} \left[ (\nabla \theta_{ss})^2 + \nabla^2 \theta_{ss} + f (\nabla \theta_{ss}) \right] \delta(\hat{z} \cdot w_{ss}) dS_1 = 0 \quad (3.3)$$

where \( \delta \) is the usual variation symbol, and \( V \) and \( S_1 \) represent the volume of the convective cell and the area of its upper boundary respectively. It is directly checked that the Euler-Lagrange equations corresponding to arbitrary variations \( \delta w_{ss} \) and \( \delta \theta_{ss} \) are relations (3.1) and (3.2); moreover the boundary conditions (2.6b) and (2.7) are also recovered as natural boundary conditions. It is worth noting that (3.3) is not a classical variational principle in the sense that some quantities like \( w_{ss} \) in the second volume integral and the quantities between brackets in the surface integral are not submitted to variation. Such a principle is generally classified as a quasi-variational principle in the literature [11, 12].

It must be realized that an « exact » variational principle cannot be formulated in relation with the present problem because the particular boundary condition (2.7) renders the problem not self-adjoint. By the way, many principles pertain to the class of quasi-variational principles [11, 12], a typical example is Hamilton’s principle in classical mechanics when friction is present. Despite its quasi-variational property, one is however allowed to use the classical Rayleigh-Ritz technique.

Assume that there exist steady solutions of the form

$$w_{ss} = W(z) \phi(x, y), \quad \theta_{ss} = \Theta(z) \phi(x, y), \quad (3.4)$$

with amplitudes \( W(z) \) and \( \Theta(z) \), and where \( \phi(x, y) \) represents the planform in the horizontal plane; \( \phi \) satisfies the relation

$$\nabla^2 \phi + k^2 \phi = 0, \quad (3.5)$$

and is normalized so that

$$\langle \phi^2 \rangle = 1, \quad (3.6)$$

\( k \) is the wavenumber in the horizontal plane and \( \langle \cdots \rangle \) denotes the average over the horizontal plane.

Setting \( \hat{z} = D \) and substituting (3.4) into (3.3) leads to the following variational equation

$$\delta I(W, \Theta) = \frac{1}{2} \int_0^1 \left[ \frac{1}{k^2} (D^2 W - k^2 W)^2 + (D\Theta)^2 + k^2 \Theta^2 \right] dz -$$

$$- \int_0^1 W \delta \Theta dz - M[\Theta(f + \Theta Q) \delta(DW)]_z = 1 = 0, \quad (3.7)$$

where \( Q \) stands for [10]

$$Q = \frac{1}{2} \langle \phi^3 \rangle. \quad (3.8)$$

It should be noticed that \( Q \) vanishes for cells taking the shape of rolls, squares and rectangles while for hexagons, one has

$$Q = \frac{1}{\sqrt{6}}. \quad (3.9)$$
The Euler-Lagrange equations corresponding to (3.7) are given by

\[
(D^2 - k^2)^2 W = 0 \quad (0 < z < 1), \quad (3.10)
\]

\[
(D^2 - k^2) \Theta + W = 0 \quad (0 < z < 1), \quad (3.11)
\]

while the natural boundary conditions are

\[
D\Theta = 0 \quad (z = 1), \quad (3.12)
\]

\[
D^2 W = k^2 M\Theta (f + \Theta Q) \quad (z = 1). \quad (3.13)
\]

It is easily checked that equations (3.10) to (3.13) are obtained by replacing (3.4) in (3.1) and (3.2) and the boundary conditions (2.6) and (2.7) and making use of (3.5), (3.6) and (3.8).

The set (3.10) to (3.13), together with the essential boundary conditions

\[
W = DW = \Theta = 0 \quad (at \ z = 0), \quad (3.14)
\]

\[
W = 0 \quad (at \ z = 1), \quad (3.15)
\]

is solved using the Rayleigh-Ritz method. It consists of expanding the unknown amplitudes \( W(z) \) and \( \Theta(z) \) in terms of polynomial functions

\[
W = \sum_{n=1}^{N} a_n f_n(z), \quad \Theta = \sum_{n=1}^{N} b_n g_n(z), \quad (3.16)
\]

where \( f_n(z) \) and \( g_n(z) \) are a priori selected functions verifying the essential boundary conditions; they are chosen as

\[
f_n(z) = z^{\rho}(1 - z) T_{n-1}^*(z), \quad (3.17)
\]

\[
g_n(z) = z \left( 1 - \frac{1}{z^2} \right) T_{n-1}^*(z), \quad (3.18)
\]

where \( T_{n}^*(z) \) are the modified Tchebyshev polynomials. The constant coefficients \( a_n \) and \( b_n \) in (3.16) are unknown quantities derived from the stationary conditions

\[
\frac{\partial J}{\partial a_i} = 0, \quad \frac{\partial J}{\partial b_i} = 0, \quad (i = 1, 2, ..., N). \quad (3.19)
\]

This numerical procedure predicts a steady solution for each value of \( Q \). This means that any particular cellular pattern, like rolls, squares, hexagons, ... is mathematically admissible. However, observations show a tendency toward a single well defined cellular structure. In order to determine the preferred form of convection, we shall examine the stability of the various steady solutions by superimposing infinitesimally small disturbances. In the next section, the stability of solutions consisting of rolls, rectangles and hexagons is investigated; other planforms, like pentagons, octagons, etc. are not considered because no experimental evidence of their existence has ever been displayed.

4. The preferred planform.

The disturbances are assumed to be given by

\[
\bar{u} = u(x, y, z) \exp(\sigma t), \quad (4.1)
\]

\[
\bar{\Theta} = \Theta(x, y, z) \exp(\sigma t), \quad (4.2)
\]

where \( u'(u', v', w') \) and \( \Theta' \) are their amplitudes, \( \sigma \) is a real parameter whose sign determines the stability of the steady solutions.

The disturbances \( \Theta' \) and \( u' \) obey the linearized equations

\[
\sigma \tilde{\Theta} + u_n \cdot \nabla \Theta + u' \cdot \nabla \Theta_{ns} = \omega' + \nabla^2 \Theta', \quad (4.3)
\]

\[
Pr^{-1}[\sigma \nabla^2 w' + \nabla^2 (u_n \cdot \nabla w' + u' \cdot \nabla w_{ns}) - \nabla^2 (u_n \cdot \nabla u' + u' \cdot \nabla u_{ns}) - \nabla^2 (u_n \cdot \nabla v' + u' \cdot \nabla v_{ns})] = \nabla^4 w', \quad (4.4)
\]

plus two similar equations for the \( u', v' \) components, which are of no use in the following. The boundary condition at the upper boundary involving the second-order Marangoni number is written

\[
\delta_{zz}^2 w' = -M \nabla \cdot [(\Theta_{ns} + f) \nabla \Theta' + \Theta' \nabla \Theta_{ns}]. \quad (4.5)
\]

To avoid costly and lengthy calculations, the Prandtl number is assumed to be infinite. This is certainly a good approximation for highly viscous oils. Moreover from calculation and experimental observations, it is expected that \( Pr = \infty \) is a reasonable hypothesis in the description of fluids with \( Pr > 5 [13] \).

The variational equation giving back the set (4.3) to (4.5) reads:

\[
\delta J(u', \Theta') = \frac{1}{2} \delta \left[ \int_{V} \left[ (\nabla^2 w')^2 + (\nabla \Theta')^2 + \sigma \Theta'^2 \right] dV - \int_{V'} w' \delta \Theta' dV + \int_{V'} (u_n \cdot \nabla \Theta' + u' \cdot \nabla \Theta_{ns}) \delta \Theta' dV + M \int_{S_1} \left[ f \nabla^2 \nabla \Theta' + \nabla^2 (\Theta_{ns} \Theta') \right] \delta (\nabla^2 w') dS_1. \quad (4.6)
\]

By analogy with the form (3.4) of the steady solutions, it is assumed that \( u' \) and \( \Theta' \) are separable and given by

\[
u' = U'(z, V(z), \Theta'(z) \phi'(x, y), \quad (4.7)
\]

\[
\Theta' = \Theta'(z, \phi'(x, y), \quad (4.8)
\]

with

\[
\nabla^2 \phi' + k^2 \phi' = 0,
\]

where \( k' \) is the wavenumber of the superimposed perturbation. The continuity equation \( \nabla \cdot u' = 0 \) requires that \( U' \) and \( V' \) be expressed by

\[
U' \phi' = \frac{1}{k^2} \delta_z W' \delta_x \phi', \quad (4.9)
\]
Substituting (4.7) to (4.10) into the variational principle (4.6), one obtains

\[ \delta J(W', \theta') = \frac{1}{2} \delta \left[ \frac{1}{k^2} (D^2 W' - k^2 W')^2 + (D\theta')^2 + (k^2 + \sigma) \theta'^2 \right] dz \]

\[ + \int_0^1 \left[ Q'\left( \theta' DW' + \frac{k^2}{k^2} \theta DW'' + 2W' D\theta' + 2 WD\theta' \right) - W' \right] \delta \theta' dz \]

\[ - M[(f \theta' + 2 \theta' \theta Q') \delta(DW')]_{z=1} = 0. \quad (4.11) \]

This equation holds for the arbitrary variations of the amplitudes \( \theta' \) and \( W' \), compatible with the essential boundary conditions

\[ W' = DW' = \theta' = 0 \quad \text{at} \quad z = 0, \quad (4.12) \]

\[ W' = 0 \quad \text{at} \quad z = 1, \quad (4.13) \]

under the condition that the following Euler-Lagrange equations and natural boundary conditions are satisfied:

\[ (D^2 - k^2 - \sigma) \theta' + W' - Q'\left[ \theta' DW' + \frac{k^2}{k^2} \theta DW'' + 2W' D\theta' + 2 WD\theta' \right] = 0 \quad (0 < z < 1), \quad (4.14) \]

\[ (D^2 - k^2)^2 W' = 0 \quad (0 < z < 1), \quad (4.15) \]

\[ D\theta' = 0, \quad D^2 W' = k^2 M(f \theta' + 2 \theta' \theta Q') \quad (z = 1). \quad (4.16) \]

The quantity \( Q' \) stands for

\[ Q' = \frac{1}{2} \left< \phi'^2 \phi \right>, \quad (4.17) \]

and describes the correlations between the superimposed and the reference steady planforms; in particular, \( Q' \) vanishes when the wavenumber \( k' \) of the superimposed pattern differs from the basic wavenumber \( k \). Various values of \( Q' \) are reported in table I.

As in section 3, the unknowns \( W' \) and \( \theta' \) are determined via the Rayleigh-Ritz procedure. This results in an eigenvalue problem for the parameters \( \sigma, f, Q', M \) and \( k' \). We fix two of them, namely \( f \) and \( Q' \), and calculate \( \sigma \) for various values of \( M \) and \( k' \). Recalling that \( \sigma > 0 \) means instability, we are able to divide the plane \( M - k' \) into two regions: one corresponding to stable solutions, the other to unstable ones. We first examine the stability of the steady solutions when the superimposed disturbance has a wavenumber \( k' \) equal to the wavenumber \( k \) of the reference state. Numerical results show that roll and rectangle patterns are characterized by a positive growth rate of the disturbance (see the last column of Table I): these configurations are clearly unstable. In contrast, hexagons may be stable: in the \( M - k' \) plane one can find

<table>
<thead>
<tr>
<th>Reference steady-state ((k))</th>
<th>Superimposed disturbance ((k' = k))</th>
<th>(Q')</th>
<th>(\sigma)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hexagon</td>
<td>Roll</td>
<td>0</td>
<td>&gt; 0 or &lt; 0</td>
</tr>
<tr>
<td>Hexagon</td>
<td>Rectangle</td>
<td>(1/\sqrt{6})</td>
<td>&gt; 0 or &lt; 0</td>
</tr>
<tr>
<td>Hexagon</td>
<td>Hexagon</td>
<td>(1/\sqrt{6})</td>
<td>&gt; 0 or &lt; 0</td>
</tr>
<tr>
<td>Roll</td>
<td>Roll</td>
<td>0</td>
<td>&gt; 0</td>
</tr>
<tr>
<td>Roll</td>
<td>Rectangle</td>
<td>(1/2\sqrt{2})</td>
<td>&gt; 0</td>
</tr>
<tr>
<td>Roll</td>
<td>Hexagon</td>
<td>(1/3\sqrt{2})</td>
<td>&gt; 0</td>
</tr>
<tr>
<td>Rectangle</td>
<td>Roll</td>
<td>0</td>
<td>&gt; 0</td>
</tr>
<tr>
<td>Rectangle</td>
<td>Rectangle</td>
<td>0</td>
<td>&gt; 0</td>
</tr>
<tr>
<td>Rectangle</td>
<td>Hexagon</td>
<td>(1/3)</td>
<td>&gt; 0</td>
</tr>
<tr>
<td>Any pattern ((k))</td>
<td>Any pattern with (k' \neq k)</td>
<td>0</td>
<td>&gt; 0 or &lt; 0</td>
</tr>
</tbody>
</table>
regions corresponding to negative values of the growth parameter $\sigma$. These regions are represented by dashed areas in figures 2 to 4. The analysis must be completed by examining the stability of hexagons with respect of disturbances with a different wavenumber ($k' \neq k$), which will provide an upper limit to the domain of stability in the form of a parallel to the $k'$ axis; this parallel is located at $M_{\lim}$ whose value is derived from

$$Ma^c = -fM_{\lim}, \quad (4.18)$$

$Ma^c$ is the critical Marangoni number obtained from the classical Marangoni problem: for an adiabatically insulated upper surface and in the absence of buoyancy effects, $Ma^c$ is equal to 79.6. The result (4.18) follows directly from the boundary condition (4.16b): recalling that $k' \neq k$ implies $Q' = 0$, it is seen that (4.16b) reduces to the classical Marangoni boundary condition under the condition to set $-fM$ equal to $Ma$.

Determination of the stability domains is performed for three different values of the parameter $f$, namely $-0.1$, $0$ and $0.1$. These values describe situations for which the temperature at the upper face is respectively smaller, equal or larger than the temperature at which the surface-tension reaches its minimum. The limiting value $M_{\lim}$ has been drawn only for negative values of $f$: when $f$ vanishes, $M_{\lim}$ is infinite whereas for positive values of $f$, $M_{\lim}$ should be negative but such negative values are excluded by the very definition (Eq. (2.8)) of $M$. The area of the domain of stability is strongly affected by the sign of $f$: for a given value of $f$, the area of the stability zone is the narrowest for negative $f$'s. The minima of the $M - k'$ curves define a critical value $M^c$ below which hexagonal steady solutions are unconditionally stable whatever the value of the wavenumber; the critical $M^c$ values are listed in table II for three values of $f$.

<table>
<thead>
<tr>
<th>$f$</th>
<th>$-0.1$</th>
<th>$0$</th>
<th>$0.1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$M^c$</td>
<td>405</td>
<td>550</td>
<td>800</td>
</tr>
</tbody>
</table>

Table II. — Critical values of the second-order Marangoni number.

It must be noticed that the above results remain unchanged when the fluid layer is heated from above, since the square of the temperature drop $\Delta T$ between the boundary faces appears in the definition of the governing second-order Marangoni number.

It is not significant to compare the present results with a classical analysis based on the usual Marangoni number as the latter is representative of fluid systems which are not included in our model.

5. Conclusions.

The role of a nonlinear dependence of the surface-tension with respect to temperature on the tension-driven instability in a thin fluid layer is examined. Steady solutions in the form of stable hexagonal cells are predicted.
The proposed model introduces several simplifications: gravity and non-Boussinesqian effects are ignored, the Prandtl number is taken to be infinite and the momentum and energy equations are linearized. The first approximation is reasonable in a microgravity environment, the Boussinesq approximation provides a good description for a wide class of fluids and mixtures. As discussed earlier, an infinite Prandtl number hypothesis is also readily acceptable for viscous fluids. The validity of the approximation that consists of linearizing the field equations of momentum and energy has been checked by calculating the ratio between $w_{ss}$ and the nonlinear term $v_{ss} \cdot \nabla \theta_{ss}$, for different values of $k$ and $M$ at $z = 0.5$ where the error is the most important. In Table III, we have reported the error percentage for $f = 0$:

<table>
<thead>
<tr>
<th>$M$</th>
<th>100</th>
<th>200</th>
<th>500</th>
<th>600</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>16 %</td>
<td>17 %</td>
<td>5 %</td>
<td>6 %</td>
</tr>
<tr>
<td>1.5</td>
<td>15 %</td>
<td>6 %</td>
<td>3 %</td>
<td>—</td>
</tr>
<tr>
<td>1</td>
<td>18 %</td>
<td>5 %</td>
<td>10 %</td>
<td>8 %</td>
</tr>
</tbody>
</table>

Similar orders of magnitude are obtained for non-zero $f$ values. Although the maximum error percentage is 18%, it falls around 5% in the vicinity of the curve separating the stable from the unstable convective cells, which is undoubtedly the region of interest. It is therefore reasonable to expect that our conclusions should not be drastically modified by performing a fully, but costly, nonlinear analysis.

In contrast, the nonlinear contribution cannot be neglected in the boundary condition (2.7). This appears clearly from table IV, where the ratio of the nonlinear terms $(v_{ss} \cdot \nabla \theta_{ss})^2 + \theta v_{ss}^2$ to the linear term $f v_{ss}^2 \theta$ at $z = 1$ is represented, for various values of the parameters.

<table>
<thead>
<tr>
<th>$M$</th>
<th>100</th>
<th>200</th>
<th>400</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>17</td>
<td>7</td>
<td>2.4</td>
</tr>
<tr>
<td>1.5</td>
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</tr>
<tr>
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<td>28</td>
<td>13</td>
<td>5.2</td>
</tr>
</tbody>
</table>

Of course, a decisive check of our model can only be provided by experimental observations. To this aim, in collaboration with the E.S.A. we plan to carry out some Marangoni experiments in microgravity environment using such mixtures, as aqueous alcohols, whose surface-tension is a parabolic function of the temperature.

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References