Transient Rayleigh-Bénard-Marangoni Convection due to Evaporation : a Linear Non-normal Stability Analysis Electronic Annex.

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Appendix A. Basic State

We obtain here the scaling laws given in section 3 which provide the evolution of a purely conductive basic state. It is recalled that the basic temperature field is initialy uniform i.e. $\theta_0(z, t = 0) = 0$ and satisfies a heat equation

$$\frac{\partial \theta_0}{\partial t} = \frac{\partial^2 \theta_0}{\partial z^2} \tag{A 1}$$

with the boundary conditions

$$\partial_z \theta_0 = 0 \quad \text{at z=0}, \qquad \partial_z \theta_0 + Bi\, \theta_0 + Bi = 0 \quad \text{at z=1}. \tag{A 2}$$

Due to the evaporation, a cooled layer of characteristic thickness $\delta(t)$ develops from the upper surface. Equation (A 1) provides the standard estimate

$$\delta_0(t) \sim \min(\sqrt{t}, 1). \tag{A 3}$$

In the following, we determine the scaling laws for the two extreme cases $Bi \ll 1$ and $1 \ll Bi$.

A) Case $Bi \ll 1$

Since $\theta_0(z, t)$ is initially zero, it remains small during a period of time $t \lesssim \tau_1$ (the time τ_1 is determined below and shown to be much larger than 1). Diffusion in the liquid and evaporation terms thus dominate in the free-surface boundary condition (A 2). Such a balance can be expressed in terms of order of magnitude as follows

$$\frac{\Delta\theta_0(t)}{\delta_0(t)} \sim Bi \tag{A 4}$$

One thus obtains using equation (A 3)

$$\Delta\theta_0 \sim Bi\sqrt{t}, \quad \delta_0(t) \sim \sqrt{t} \quad \text{for} \quad 0 \lesssim \sqrt{t} \lesssim 1, \tag{A 5}$$

During this time interval, the cooling layer has not reached the bottom at $z=0$ so that temperature field θ_0 reads in terms of orders of magnitude

$$|\theta_0| \sim \Delta\theta_0 \sim Bi\sqrt{t} \quad \text{for} \quad 0 \lesssim \sqrt{t} \lesssim 1. \tag{A 6}$$

Thereafter the thickness remains constant $\delta_0(t) \sim 1$ and the following condition holds

$$\Delta\theta_0(t) \sim Bi, \quad \delta_0(t) \sim 1 \quad \text{for} \quad 1 \lesssim t \lesssim \tau_1. \tag{A 7}$$

During this latter time interval, the heat equation (A 1) can be used with scaling (A 7) to get the temperature field θ_0 in terms of orders of magnitude

$$|\theta_0| \sim Bi\ t, \quad \text{for} \quad 1 \lesssim t \lesssim \tau_1. \tag{A 8}$$

These equations are valid if evaporation dominates heat transfer in the gas phase. This requires that $|\theta_0| \ll 1$ and determines the value $\tau_1 \sim Bi^{-1}$. For $\tau_1 \lesssim t$, the temperature field θ_0 relaxes towards the steady uniform temperature equal to $\theta_0(z,t) = -1$. Since the temperature gradient is equal to zero in that regime, all the energy due to evaporation is transfered by convection in the gas phase.

B) Case $1 \ll Bi$

For small times, the condition $|\theta_0| \ll 1$ holds and an analysis similar to the one performed for the case $Bi \ll 1$ is valid leading to

$$\Delta\theta_0 \sim Bi\sqrt{t}, \quad \delta_0(t) \sim \sqrt{t} \quad \text{for} \quad 0 \lesssim t \lesssim \tau_2, \quad \text{where} \quad \tau_2 \sim Bi^{-2}. \tag{A 9}$$

The value of τ_2 is obtained by determining the time when $\Delta\theta_0 \sim 1$. At that time, the heat flux in the gas phase becomes of the same order of the evaporation. During this time interval, the cooling layer has not reached the bottom at $z=0$ so that temperature field θ_0 reads in terms of orders of magnitude

$$|\theta_0| \sim \Delta\theta_0 \sim Bi\sqrt{t} \quad \text{for} \quad 0 \lesssim t \lesssim Bi^{-2}. \tag{A 10}$$

For $Bi^{-2} \lesssim t$, a new regime begins where the surface temperature remains constant $|\theta_0(z=1,t)| \sim 1$ while the cooled layer thickness keeps increasing

$$\Delta\theta_0 \sim 1, \quad \delta_0 \sim \sqrt{t} \quad \text{for time} \quad Bi^{-2} \lesssim t \lesssim 1. \tag{A 11}$$

This regime ends when $\delta_0 \sim 1$ at time $t \sim 1$. Thereafter the temperature decreases in the whole layer thickness to reach the steady state regime $\theta_0(z,t) = -1$.
The case $Bi \sim 1$ is the limiting case of the two previous ones. The equation (A 5) is valid until $t \sim 1$, when the cooled layer thickness and the surface temperature both reach their extremum. Thereafter the temperature decreases in the whole layer thickness, to reach the steady state.

Appendix B. Obtaining the Adjoint Equations

Let us denote by $q_j(z,t), j = 1, \ldots, 4$, the components of the vector field

$$(\hat{u}(z,t), \hat{w}(z,t), \hat{\theta}(z,t), \hat{p}(z,t)).$$

To find the maximum amplification at a given time t_1, we maximize the perturbation norm $E(\boldsymbol{q}(t_1))$

$$E(\boldsymbol{q}(t_1)) \equiv \sum_{j=1}^{3} C_j \int q_j(z,t_1) q_j^+(z,t_1) \mathrm{d}z \tag{B 1}$$

at time t_1 with respect to the set of all possible initial perturbations $\boldsymbol{q}(0)$ such that $E(\boldsymbol{q}(0)) = 1$. It is recalled that the integration is performed over the entire layer depth and the superscript $+$ denotes complex conjugation. Coefficients C_j are weight coefficients chosen to put emphasis on temperature or velocity acoording to the case considered. To analyse the initial perturbation in velocity, the kinetic energy norm E_V is used and one takes $C_1 = C_2 = 1$ and $C_3 = 0$. To analyse the initial perturbation in temperature, the temperature norm E_T is used and $C_1 = C_2 = 0$ and $C_3 = 1$.

The variation $\delta E(\boldsymbol{q}(t_1))$ with respect to a variation $\delta\boldsymbol{q}(0)$ of the initial perturbation is to be evaluated. This computation cannot be performed in a straightforward manner since the energy $E(\boldsymbol{q}(t_1))$ can be explicitly written in terms of $\boldsymbol{q}(t_1)$ but only implicitely in terms of $\boldsymbol{q}(0)$. It is known *via* several constraints: normalization of $\boldsymbol{q}(0)$ and time integration over the interval $[0, t_1]$ of equations (3.8)–(3.10). These dynamic equations relating $\boldsymbol{q}(0)$ to $\boldsymbol{q}(t_1)$, are formally written here as $F_j(\boldsymbol{q}) = 0, \;\; j = 1 \dots 4$. This optimization with constraints necessitates the introduction of Lagrangian multipliers, the so-called adjoint fields $\tilde{\boldsymbol{q}}(t) \equiv (\tilde{u}(z,t), \tilde{w}(z,t), \tilde{\theta}(z,t), \tilde{p}(z,t))$.

More specifically, a Lagrangian function L is defined, which depends on direct $\boldsymbol{q}(t)$ and adjoint $\tilde{\boldsymbol{q}}(t)$ variables over the interval $[0, t_1]$, and a normalization scalar s_0:

$$L(\boldsymbol{q}, \tilde{\boldsymbol{q}}, s_0, t_1) = E(\boldsymbol{q}(t_1)) - s_0(E(\boldsymbol{q}(0)) - 1) - \sum_{j=1}^{4} \int_0^{t_1} dt\,(\langle F_j(\boldsymbol{q}(t)), \tilde{q}_j(t)\rangle + c.c.) \quad \text{(B 2)}$$

where *c.c.* means complex conjugate and $\langle \cdot, \cdot \rangle$ stands for the scalar product

$$\langle a_1, a_2 \rangle \equiv \int \hat{a}_1(z)\hat{a}_2^+(z)\mathrm{d}z. \quad \text{(B 3)}$$

When $\boldsymbol{q}(t)$ satisfies the constraints (direct problem plus normalization at $t = 0$), all terms but the first one on the r.h.s. of equation (B 2) are zero and, by consequence, $L = E$ and $\delta L = \delta E$. At this stage, the adjoint variables and the quantity s_0 are left unspecified. Formally the variation δL reads as

$$\delta L = \sum_{j=1}^{3} C_j \left(\int q_j^+(z,t_1)\delta q_j(z,t_1)\mathrm{d}z - s_0 \int q_j^+(z,0)\delta q_j(z,0)\mathrm{d}z \right) \quad \text{(B 4)}$$
$$- \sum_{j=1}^{4} \int_0^{t_1} dt[\langle \delta F_j(\boldsymbol{q}(t)), \tilde{q}_j(t)\rangle + \langle F_j(\boldsymbol{q}(t)), \delta\tilde{q}_j(t)\rangle] + c.c..$$

The expression $\langle F_j(\boldsymbol{q}(t)), \delta\tilde{q}_j(t)\rangle$ in equation (B 4) is zero if the governing equations $F_j(\boldsymbol{q}) = 0$ are satisfied during the time interval $[0, t_1]$. The main idea then amounts to rewriting quantity $\langle \delta F_j(\boldsymbol{q}(t)), \tilde{q}_j(t)\rangle$ in terms of $\delta q_k(t)$. This is done by integrating by parts in space or time. After some tedious algebra, the following identity

$$\sum_{j=1}^{4} \int_0^{t_1} dt\langle \delta F_j(\boldsymbol{q}(t)), \tilde{q}_j(t)\rangle = \sum_{j=1}^{4} [\int_0^{t_1} dt\langle \tilde{F}_j(\tilde{q}(t), q), \delta q(t)\rangle] + \quad \text{(B 5)}$$
$$+ \frac{1}{Pr} \sum_{j=1}^{2} [\int \tilde{q}_j^+(z,t_1)\delta q_j(z,t_1)\mathrm{d}z - \int \tilde{q}_j^+(z,0)\delta q_j(z,0)\mathrm{d}z]$$
$$+ \int \tilde{q}_3^+(z,t_1)\delta q_3(z,t_1)\mathrm{d}z - \int \tilde{q}_3^+(z,0)\delta q_3(z,0)\mathrm{d}z + B(\delta\boldsymbol{q}, \tilde{\boldsymbol{q}})$$

can be established, where $\tilde{F}_j$ is an expression containing spatial or time derivatives of $\tilde{\boldsymbol{q}}$.

Note that the second, third and fourth r.h.s terms originate from integrations by parts of time derivatives in equations (3.8)–(3.10) and terms $B(\delta\boldsymbol{q},\tilde{\boldsymbol{q}})$ are generated from the boundary terms resulting from integrations by parts of spatial derivatives. These latter terms hence involve only quantities $\delta\boldsymbol{q}$ and $\tilde{\boldsymbol{q}}$ evaluated at the boundaries $z=0$ and $z=1$.

At this stage, the freedom of the Lagrangian multipliers can be used to impose some added constraints on the adjoints fields $\tilde{\boldsymbol{q}}$, namely: (i) equations $\tilde{F}_j(\tilde{\boldsymbol{q}}(t))=0, j=1\dots 4$, which are similar to $F_j(\boldsymbol{q}(t))$ for $\boldsymbol{q}$ and define the evolution equations (3.15)–(3.17); and (ii) boundary conditions $B(\delta\boldsymbol{q},\tilde{\boldsymbol{q}})=0$, which are the counterpart of boundary conditions (3.10)–(3.12) on $\boldsymbol{q}$ and define the boundary conditions (3.18)–(3.19) for $\tilde{\boldsymbol{q}}$. This new system can be simulated as the direct problem. It is easily seen that the adjoint system (3.15)–(3.19) must be integrated backwards in time. When it is satisfied, the variation δL reads

$$
\begin{aligned}
\delta L = \sum_{j=1}^{2}\Bigg(&\int (C_j q_j^{+}(z,t_1) - \frac{1}{Pr}\tilde{q}_j^{+}(z,t_1))\delta q_j(z,t_1)\mathrm{d}z \\
&- \int (s_0 C_j q_j^{+}(z,0) - \frac{1}{Pr}\tilde{q}_j^{+}(z,0))\delta q_j(z,0)\mathrm{d}z\Bigg) \\
&+ \int (C_3 q_3^{+}(z,t_1) - \tilde{q}_3^{+}(z,t_1))\delta q_3(z,t_1)\mathrm{d}z \\
&- \int (s_0 C_3 q_3^{+}(z,0) - \tilde{q}_3^{+}(z,0))\delta q_j(z,0)\mathrm{d}z + c.c.
\end{aligned}
\tag{B 6}
$$

Two relations can be still imposed, a first one at time $t=t_1$ which relates $\tilde{q}_j(z,t_1)$ and $q_j(z,t_1)$ and a second one at time $t=0$ which relates $\tilde{q}_j(z,0)$ and $q_j(z,0)$. These two constraints are satisfied so that $\delta L=0$ and are defined precisely below according to the norm and Prandtl number. The condition $\delta L=0$ means that an optimal perturbation is attained. However this process should be self-consistent : one uses the iteration procedure (3.20). When the iterative process has converged, an initial optimal perturbation for time t_1 is found.

A) **Finite Prandtl and zero initial temperature perturbations**

If one considers only initial perturbations in velocity field so that variation of temperature field $\delta q_3(z,0)$ is zero, it is consistent to use the norm E_V, i.e., $C_1=C_2=1$ and $C_3=0$. Equation (B 6) then naturally leads to the relation

$$
\tilde{q}_j(z,t_1) = Pr q_j(z,t_1), \quad j=1,2\ ; \qquad \tilde{q}_3(z,t_1)=0 \tag{B 7}
$$

at time $t=t_1$ and the relation

$$
q_j(z,0) = \frac{1}{Pr\, s_0}\tilde{q}_j(z,0), \quad j=1,2\ ; \qquad q_3(z,0)=0 \tag{B 8}
$$

at time $t=0$, where s_0 is chosen such that the normalization condition $E(\boldsymbol{q}(0))=1$ is satisfied.

B) **Finite Prandtl and zero initial velocity perturbations**

When considering only initial perturbations in temperature field so that $\delta q_1(z,0) = \delta q_2(z,0) = 0$, it is consistent to use the norm E_T, i.e., $C_1=C_2=0$ and $C_3=1$. Equation (B 6) then leads to the relation

$$
\tilde{q}_j(z,t_1)=0 \quad j=1,2\ ; \quad \tilde{q}_3(z,t_1)=q_3(z,t_1) \tag{B 9}
$$

at time $t = t_1$ and

$$q_j(z,0) = 0, \quad j = 1,2\ ; \qquad q_3(z,0) = \frac{1}{s_0}\tilde{q}_3(z,0) \tag{B 10}$$

at time $t = 0$ so that the normalization is satisfied.

C) **Infinite Prandtl number**

For the infinite Prandtl number, the norm E_T is chosen since the velocity is slaved to the temperature field in that instance, hence $C_1 = C_2 = 0$ and $C_3 = 1$. The equations are then identical to the previous case except that only the equation for temperature appears, i.e.,

$$\tilde{q}_3(z,t_1) = q_3(z,t_1), \tag{B 11}$$

and at time $t = 0$

$$q_3(z,0) = \frac{1}{s_0}\tilde{q}_3(z,0) \tag{B 12}$$

so that the normalization is satisfied.

Appendix C. Numerical Method.

For the numerical solution, the direct and adjoint equations are reformulated as fourth-order problem in a streamfunction-like approach. The incompressibility constraint is thereby satisfied automatically, and the pressure and horizontal velocity are eliminated from the equations. For finite Prandtl number, the direct equations take the form

$$\frac{1}{Pr}\frac{\partial}{\partial t}\hat{\eta} = \left[\frac{\partial^2}{\partial z^2} - k^2\right]\hat{\eta} - Rak^2\hat{\theta}, \tag{C 1}$$

$$\hat{\eta} = \left[\frac{\partial^2}{\partial z^2} - k^2\right]\hat{w}, \tag{C 2}$$

$$\frac{\partial}{\partial t}\hat{\theta} + \hat{w}\frac{\partial \theta_0}{\partial z} = \left[\frac{\partial^2}{\partial z^2} - k^2\right]\hat{\theta}. \tag{C 3}$$

The boundary conditions are

$$\hat{w} = \frac{\partial \hat{w}}{\partial z} = \frac{\partial \hat{\theta}}{\partial z} = 0 \quad \text{at } z = 0 \tag{C 4}$$

and

$$\hat{w} = 0, \quad \hat{\eta} + k^2 Ma\hat{\theta} = 0, \quad \frac{\partial \hat{\theta}}{\partial z} + Bi\hat{\theta} = 0 \quad \text{at } z = 1. \tag{C 5}$$

The adjoint fields satisfy the system

$$\frac{1}{Pr}\frac{\partial}{\partial \tau}\tilde{\eta} = \left[\frac{\partial^2}{\partial z^2} - k^2\right]\tilde{\eta} - k^2\tilde{\theta}\frac{\partial \theta_0}{\partial z}, \tag{C 6}$$

$$\tilde{\eta} = \left[\frac{\partial^2}{\partial z^2} - k^2\right]\tilde{w}, \tag{C 7}$$

$$\frac{\partial}{\partial \tau}\hat{\theta} = \left[\frac{\partial^2}{\partial z^2} - k^2\right]\hat{\theta} + Ra\tilde{w} \tag{C 8}$$

with the boundary conditions

$$\tilde{w} = \frac{\partial \tilde{w}}{\partial z} = \frac{\partial \tilde{\theta}}{\partial z} = 0 \quad \text{at } z = 0 \tag{C 9}$$

and

$$\tilde{w} = \tilde{\eta} = 0, \quad \frac{\partial \tilde{\theta}}{\partial z} + Bi\tilde{\theta} + Ma\frac{\partial \tilde{w}}{\partial z} = 0 \quad \text{at } z = 1. \tag{C 10}$$

These equations are discretized in time with a backward Euler method for the diffusive terms. The product term with the basic temperature profile is treated with the explicit Euler method. For the direct problem the solution at the new time level $n+1$ is obtained by solving the following equations in sequence:

$$\left[\frac{\partial^2}{\partial z^2} - k^2 - \frac{1}{\Delta t}\right]\hat{\theta}^{n+1} = -\frac{\hat{\theta}^n}{\Delta t} + \hat{w}^n\frac{\partial \theta_0^n}{\partial z}, \tag{C 11}$$

$$\left[\frac{\partial^2}{\partial z^2} - k^2 - \frac{1}{Pr\Delta t}\right]\hat{\eta}^{n+1} = -\frac{\hat{\eta}^n}{Pr\Delta t} + Rak^2\hat{\theta}^{n+1}, \tag{C 12}$$

$$\left[\frac{\partial^2}{\partial z^2} - k^2\right]\hat{w}^{n+1} = \hat{\eta}^{n+1}. \tag{C 13}$$

The boundary equations for $\hat{\eta}$ are given in terms of $\hat{w}$. To satisfy them, the solution of the second and third equation is represented by the linear combination

$$\hat{\eta}^{n+1} = \hat{\eta}_P + \lambda\hat{\eta}_1 + \mu\hat{\eta}_2, \tag{C 14}$$

$$\hat{w}^{n+1} = \hat{w}_P + \lambda\hat{w}_1 + \mu\hat{w}_2, \tag{C 15}$$

where the solution with subscript P is a particular solution of the $\hat{\eta}$-equation (C 12) with $\hat{\eta}_P = 0$ on the boundaries $z = 0$ and $z = 1$. The functions with subscripts 1 and 2 satisfy the homogeneous $\hat{\eta}$-equation with zero right hand side and two linearly independent boundary conditions, which we choose as

$$\hat{\eta}_1(z = 1) = \hat{\eta}_1(z = 0) = 1, \tag{C 16}$$

$$\hat{\eta}_2(z = 1) = -\hat{\eta}_2(z = 0) = 1. \tag{C 17}$$

The boundary conditions $\partial\hat{w}/\partial z = 0$ at $z = 0$ and $\hat{\eta} + k^2 Ma\hat{\theta} = 0$ at $z = 1$ determine the coefficients λ and μ in the linear combination. We note that the functions $\hat{w}_P$, $\hat{w}_1$ and $\hat{w}_2$ satisfy zero boundary conditions at $z = 0$ and $z = 1$.

The adjoint solution at the new time level $n+1$ is likewise found by solving the equations

$$\left[\frac{\partial^2}{\partial z^2} - k^2 - \frac{1}{Pr\Delta\tau}\right]\tilde{\eta}^{n+1} = -\frac{\tilde{\eta}^n}{Pr\Delta\tau} - k^2\tilde{\theta}^n\frac{\partial \theta_0^n}{\partial z}, \tag{C 18}$$

$$\left[\frac{\partial^2}{\partial z^2} - k^2\right]\tilde{w}^{n+1} = \tilde{\eta}^{n+1}, \tag{C 19}$$

$$\left[\frac{\partial^2}{\partial z^2} - k^2 - \frac{1}{\Delta\tau}\right]\tilde{\theta}^{n+1} = -\frac{\tilde{\theta}^n}{\Delta\tau} - Ra\tilde{w}^{n+1} \tag{C 20}$$

in the given sequence. The solution for $\tilde{w}^{n+1}$ and $\tilde{\eta}^{n+1}$ must again be represented as a linear combination with auxiliary functions satisfying the homogeneous $\tilde{\eta}$-equation in order to satisfy the boundary conditions.

Discretization in space is realized with an expansion in Chebyshev polynomials (see Canuto *et al.* (1988)). Product terms with the perturbation and the basic state are calculated in physical space by a fast cosine transform. The Helmholtz equations for the variables $\hat{\eta}$, $\hat{w}$, $\hat{\theta}$ and the adjoint variables $\tilde{\eta}$, $\tilde{w}$, $\tilde{\theta}$ reduce to essentially tridiagonal linear systems. The boundary conditions are treated with the tau method, which produces two filled rows in the matrix representation. The limit of infinite Prandtl number requires no changes in the solution procedure.

The basic temperature profile is computed with the same numerical method as the perturbations, i.e. using the backward-Euler method and a Chebyshev polynomial expansion with the same time step and number of polynomials. The entire field $\theta_0(z,t)$ is stored in an array in order to speed up the backward integration of the adjoint equations.
The code was tested for infinite Prandtl number with a stationary basic temperature profile. It was verified that exponential growth of the optimal perturbations appeared at the proper threshold values of $Ma \approx 79.6$ for pure Marangoni convection with $Bi = 0$ (Pearson 1958) and for $Ra \approx 1100$ for pure Rayleigh convection with fixed temperature on the free surface (Chandrasekhar 1961) For this verification, the boundary condition at the bottom was changed to constant temperature.

Appendix D. Analysis of Stability for the Marangoni case

D.1. *The approach for the Marangoni case ($Ra = 0$).*

This section presents an approach valid for infinite Prandtl number, which evaluates the evolution in terms of orders of magnitude. It is based on two hypotheses which make the analysis tractable. First the initial perturbation of wavenumber k in the x direction is only a temperature perturbation i.e. $\hat{u}(z,t=0) = \hat{w}(z,t=0) = 0$ and the temperature perturbation $\hat{\theta}(z,t=0)$ is uniform along the z-direction. Second, the flow is supposed unstable i.e. convection sets in, if there exists a time and a region in the flow in which the advection term in equation (3.10) becomes greater or equal to the two diffusion terms which tend to damp the initial perturbation.
Practically, one first determines the orders of magnitude for temperature perturbations when the system is in a stable regime or near the critical curve, i.e., when the term corresponding to advected heat transfer $\hat{w}\frac{\partial\theta_0}{\partial z}$ in equation (3.10) can be neglected, according to the second hypothesis. Thereafter one computes the order of magnitude of the term $k^2\hat{\theta}$ i.e., the diffusion in the x-direction, and of the term $\frac{\partial^2\hat{\theta}}{\partial z^2}$, i.e., the diffusion in the z-direction, corresponding to the stable regime. This is done in subsection D.2. On the other hand, the order of magnitude for velocity $\hat{w}$ is found in subsection D.3, as well as the corresponding advection term, $\hat{w}\frac{\partial\theta_0}{\partial z}$.
Since the velocity is computed using the temperature perturbation field estimated for the stable configuration, this approach is consistent only if the advection term remains much smaller than one of the diffusion terms. The sets of parameters Ma, Bi, Pr that give consistent results for each time and mode are considered in the "stable" domain. Otherwise, if there exists a time and a mode of wavenumber k such that the advection term is larger in order of magnitude than the two diffusion terms, the corresponding set of parameters is associated with a situation where convection sets in (subsection D.4). The scaling laws for critical parameters are then derived by solving the resulting set of inequalities (subsection D.5).

D.2. *Scaling Analysis for the Temperature Perturbation Field*

As mentioned in the previous paragraph, we determine the orders of magnitude for temperature perturbations by neglecting the advected heat transfer $\hat{w}\frac{\partial\theta_0}{\partial z}$ in equation (3.10). One thus obtains

$$\frac{\partial\hat{\theta}}{\partial t} - \left[\frac{\partial^2}{\partial z^2} - k^2\right]\hat{\theta} = 0 \tag{D 1}$$

The temperature perturbation field also satisfies the boundary conditions

$$\frac{\partial\hat{\theta}}{\partial z} + Bi\,\hat{\theta} = 0, \quad \text{at } z = 1, \tag{D 2}$$

$$\frac{\partial \hat{\theta}}{\partial z} = 0 \quad \text{at } z = 0. \tag{D 3}$$

The cooling due to evaporation imposes that a thermal boundary layer for the temperature perturbation $\hat{\theta}(z,t)$ develops. One can easily check that the solution

$$\hat{\theta}(z,t) = a_0(1 + \theta_0(z,t)) \exp(-k^2 t) \tag{D 4}$$

satisfies the above system and the condition of uniformity at $t = 0$. Note that a_0 is simply the initial amplitude of the temperature perturbation which is taken to be equal to one in the sequel. From the above equation, it is readily seen that the thickness of the thermal boundary layer for the perturbation field $\hat{\theta}(t)$ is equal to $\delta_0(t)$ and that the scale of variation in the z-direction of the perturbed field $\hat{\theta}(z,t)$ denoted by $\Delta\hat{\theta}$ satisfies

$$\Delta\hat{\theta}(t) \sim \Delta\theta_0(t) \exp(-k^2 t) \tag{D 5}$$

The scale $\hat{\theta}_s$ of the perturbed field $\hat{\theta}(z = 1, t)$ on the surface satisfies according to equation (D 2)

$$\hat{\theta}_s \sim \frac{1}{Bi} \frac{\Delta\hat{\theta}(t)}{\delta_0(t)} \tag{D 6}$$

Using the results of Annex A, it is straightforward to find the following estimates:
For $Bi \ll 1$:

$$\Delta\hat{\theta}(t) \sim Bi\sqrt{t} \exp(-k^2 t), \quad \hat{\theta}_s \sim \exp(-k^2 t), \quad \hat{\theta} \sim \exp(-k^2 t) \quad \text{for} \quad 0 \lesssim \sqrt{t} \lesssim 1, \tag{D 7}$$

$$\Delta\hat{\theta}(t) \sim Bi \exp(-k^2 t), \quad \hat{\theta}_s \sim \exp(-k^2 t), \quad \hat{\theta} \sim \exp(-k^2 t) \quad \text{for} \quad 1 \lesssim t \lesssim Bi^{-1}. \tag{D 8}$$

For $1 \ll Bi$:

$$\Delta\hat{\theta}(t) \sim Bi\sqrt{t} \exp(-k^2 t), \quad \hat{\theta}_s \sim \exp(-k^2 t), \quad \hat{\theta} \sim \exp(-k^2 t) \quad \text{for} \quad 0 \lesssim t \lesssim Bi^{-2}, \tag{D 9}$$

$$\Delta\hat{\theta}(t) \sim \exp(-k^2 t), \quad \hat{\theta}_s \sim \frac{\exp(-k^2 t)}{Bi\sqrt{t}}, \quad \hat{\theta} \sim \exp(-k^2 t), \text{ for} \quad Bi^{-2} \lesssim t \lesssim 1. \tag{D 10}$$

D.3. *Scalings for the Velocity Field in the Bénard-Marangoni problem ($Ra = 0$).*

The equation of the vorticity field can be easily deduced from equations (3.8) and (3.9). Denoting by $\hat{\omega}$ the y-component of vorticity, we obtain the diffusion equation

$$\frac{1}{Pr} \partial_t \hat{\omega} = \partial_z^2 \hat{\omega} - k^2 \hat{\omega} \tag{D 11}$$

For infinite Prandtl number, this equations simplifies

$$\partial_z^2 \hat{\omega} - k^2 \hat{\omega} = 0 \tag{D 12}$$

The vorticity is slaved to the temperature evolution *via* the boundary condition at the free surface given by equation (3.11) :

$$\hat{\omega} + ikMa\hat{\theta} = 0 \quad \text{at} \quad z = 1. \tag{D 13}$$

Equation (D 12) plus the forcing (D 13) defines an hydrodynamic boundary layer δ_H. It is easily seen that the proper scaling reads

$$\delta_H \sim \min(1/k, 1) \tag{D 14}$$

The hydrodynamic boundary layer either reaches the bottom, i.e., $\delta_H \sim 1$, or the diffusion term along x becomes of the same order of the diffusion term along z and $\delta_H \sim 1/k$.

From equation (3.11), a relation between $\hat{\theta}_s$ and the order of magnitude of velocity $\hat{u}$ can be found :

$$\hat{u} \sim k\delta_H \; Ma \; \hat{\theta}_s(t). \tag{D 15}$$

The order of magnitude of the vertical component $\hat{w}$ of velocity is provided *via* mass conservation

$$\hat{w} \sim k\delta_H \hat{u} \equiv (k\delta_H)^2 \; Ma \; \hat{\theta}_s(t). \tag{D 16}$$

D.4. *Condition for the onset of convection for the Marangoni flow ($Ra = 0$)*

To describe the time evolution of perturbations, one must distinguish two regions along the z direction i.e. inside and outside the thermal boundary layer. Outside the layer ($\delta_0 \lesssim 1-z \lesssim 1$), the advection term in equation (3.10) is zero since the basic temperature field $\theta_0(z,t)$ vanishes : hence diffusion dominates and perturbations are always damped. Instability thus only arises inside the thermal layer ($0 \lesssim 1-z \lesssim \delta_0$).
To determine the onset of instability, one first compares the order of magnitude of the advection and the diffusion along the x-direction in the thermal layer

$$\frac{\hat{w}_{th}\Delta\theta_0}{\delta_0} \; , \; k^2\hat{\theta} \tag{D 17}$$

where $\hat{w}_{th}$ denotes the order of magnitude of the vertical velocity in the thermal boundary layer, and second the order of magnitude of the advection term and of the diffusion term in the z-direction

$$\frac{\hat{w}_{th}\Delta\theta_0}{\delta_0} \; , \; \frac{\Delta\hat{\theta}}{\delta_0^2} \tag{D 18}$$

The existence of a convection onset thus implies that there exists a time and a mode of wavenumber k for which the two conditions

$$\hat{w}_{th}(\frac{\Delta\theta_0}{\delta_0}) \gtrsim k^2\hat{\theta} \quad \text{and} \quad \hat{w}_{th}(\frac{\Delta\theta_0}{\delta_0}) \gtrsim \frac{\Delta\hat{\theta}}{\delta_0^2} \tag{D 19}$$

hold. We need quantity $\hat{w}_{th}$ since it explicitely appears in the above inequalities. Two possibilities should be considered at each time : $\delta_0(t) \lesssim \delta_H$ or $\delta_H \lesssim \delta_0(t)$. In the first case, the thermal layer is included in the hydrodynamic layer and one may use the scaling $\hat{w}_{th} \sim \frac{\delta_0}{\delta_H}\hat{w}$. In the second case $\hat{w}_{th} \sim \hat{w}$. Using equation (D 16), this implies that the scaling $\hat{w}_{th}$ is such that

$$\hat{w}_{th} \sim \min\left(1, \frac{\delta_0}{\delta_H}\right)\hat{w} = \min\left(1, \frac{\delta_0}{\delta_H}\right)(k\delta_H)^2 \; Ma \; \hat{\theta}_s. \tag{D 20}$$

It is now possible to rewrite inequalities (D 19) as

$$\min\left(1, \frac{\delta_0}{\delta_H}\right)\delta_H^2 \; Ma \; \hat{\theta}_s(\frac{\Delta\theta_0}{\delta_0}) \gtrsim \hat{\theta} \quad \text{and} \quad \min\left(1, \frac{\delta_0}{\delta_H}\right)(k\delta_H)^2 \; Ma \; \hat{\theta}_s\Delta\theta_0 \gtrsim \frac{\Delta\hat{\theta}}{\delta_0} \tag{D 21}$$

D.5. *Derivation of scaling laws*

One must now introduce the various expressions previously obtained for $\Delta\theta_0$, δ_0, $\hat{\theta}$, $\Delta\hat{\theta}$, $\hat{\theta}_s$, δ_H inside instability conditions D 21. The expressions for δ_0 and $\Delta\theta_0$ are obtained in section (A), the expressions for $\hat{\theta}$, $\Delta\hat{\theta}$ and $\hat{\theta}_s$ in section (D.2), the expression for δ_H in section (D.3). To ease the discussion, three separate cases are studied :
A) $Bi \ll 1$, B) $1 \ll Bi$ and $t \lesssim Bi^{-2}$, C) $1 \ll Bi$ and $Bi^{-2} \lesssim t$.

A) Case $Bi \ll 1$

First let us recall from Annex A, that the following relations hold :

$$\delta_0(t) \sim \min(\sqrt{t}, 1) \quad \text{and} \quad \frac{\Delta\theta_0(t)}{\delta_0(t)} \sim Bi. \tag{D 22}$$

From section D.2, one easily verifies that the temperature perturbation field is such that

$$\Delta\hat{\theta} \sim Bi\delta_0\hat{\theta}_s \quad \text{and} \quad \hat{\theta} \sim \hat{\theta}_s \tag{D 23}$$

Using equations (D 22) and (D 23), condition (D 21) can be transformed into

$$\min\left(1, \frac{\delta_0}{\delta_H}\right) \; Ma \; Bi \; \delta_H^2 \; \gtrsim 1 \quad \text{and} \quad \min\left(1, \frac{\delta_0}{\delta_H}\right) \; \delta_0 \; Ma \; k^2 \; \delta_H^2 \; \gtrsim 1 \tag{D 24}$$

Note that only the period $t \lesssim 1/Bi$ should be considered here since, for $1/Bi \lesssim t$, the basic state has relaxed to a uniform temperature.
To ease the discussion, two separate cases must be considered for the wavenumber k, namely $k \lesssim 1$ and $1 \lesssim k$.

• $k \lesssim 1$

In that instance, $\delta_H \sim \min(1/k, 1) = 1$ (see equation (D 14)) leading to the equality $\min(1, \frac{\delta_0}{\delta_H}) \sim \delta_0$. Condition (D 24) reads

$$\delta_0 \; Ma \; Bi \; \gtrsim 1 \quad \text{and} \quad \delta_0^2 \; Ma \; k^2 \; \gtrsim 1 \tag{D 25}$$

The smallest Marangoni number i.e. the critical Marangoni number which satisfies such inequalities, is obtained for $\delta_0(t) \sim 1$ i.e. for $1 \lesssim t$. Condition (D 25) becomes

$$Ma \; \gtrsim \frac{1}{Bi} \quad \text{and} \quad k^2 \; \gtrsim \frac{1}{Ma} \tag{D 26}$$

From the above onditions, one easily gets the critical value

$$Ma_c \sim 1/Bi, \quad \text{with} \quad \sqrt{Bi} \lesssim k_c \lesssim 1 \quad \text{and} \quad 1 \lesssim t_c \lesssim 1/Bi. \tag{D 27}$$

• $1 \lesssim k$.

In that instance, $\delta_H \sim \min(1/k, 1) = 1/k$. Since the two functions $\min(1, k\delta_0)$ and $\min(1, k\delta_0)\delta_0$ are both increasing functions of δ_0 when $\delta_0 \lesssim 1/k$, the critical Marangoni must be obtained when $1/k \lesssim \delta_0$ for which $\min(1, k\delta_0) = 1$. Conditions (D 24) become

$$Ma \; Bi \; \gtrsim k^2 \quad \text{and} \quad Ma \; \delta_0(t) \; \gtrsim 1 \tag{D 28}$$

A straigthforward discussion directly leads to the conditions

$$Ma_c \sim 1/Bi \quad \text{and} \quad k_c \sim 1 \quad \text{and} \quad 1 \lesssim t_c \lesssim 1/Bi. \tag{D 29}$$

which is a limiting case of condition (D 27). As a consequence, the critical conditions in the case $Bi \ll 1$ corresponds to condition (D 27).

B) Case $1 \ll Bi$ *and* $t \lesssim Bi^{-2}$.

From results obtained on the basic flow, it is easily seen that relations (D 22), (D 23) and thus (D 24) still hold. Moreover note that the largest value of $\delta_0(t)$ is obtained at the largest time $t \sim Bi^{-2}$: $\delta_0(Bi^{-2}) \sim Bi^{-1}$. One must consider the three cases $k \lesssim 1$, $1 \lesssim k \lesssim Bi$ and $Bi \lesssim k$.

• $k \lesssim 1$

In that case, $\delta_H \sim \min(1/k, 1) = 1$ and condition (D 25) is again satisfied. The critical

Marangoni number, is obtained for the largest possible value of $\delta_0(t)$ i.e. $\delta_0(Bi^{-2}) = Bi^{-1}$. Condition (D 25) now reads

$$Ma \gtrsim 1 \quad \text{and} \quad Ma\, k^2 \gtrsim Bi^2 \tag{D 30}$$

A straigthforward discussion directly leads to the critical conditions for instability

$$Ma_c \sim Bi^2 \quad \text{with} \quad k_c \sim 1 \quad \text{and} \quad t_c \sim Bi^{-2} \tag{D 31}$$

• $1 \lesssim k \lesssim Bi$
In that instance, $\delta_H \sim \min(1/k, 1) = 1/k$. Conditions (D 24) become

$$\min(1, k\delta_0(t))\; k^{-2}\; Ma\; Bi \gtrsim 1 \quad \text{and} \quad \min(1, k\delta_0(t))\; \delta_0(t)\; Ma \gtrsim 1 \tag{D 32}$$

The critical Marangoni number, is obtained for the largest possible value of $\delta_0(t)$ obtained at the largest time i.e. $t = Bi^{-2}$ for which $\delta_0(Bi^{-2}) = Bi^{-1}$. As a consequence

$$\min(1, k/Bi)\; k^{-2}\; Ma\; Bi \gtrsim 1 \quad \text{and} \quad \min(1, k/Bi)\; Bi^{-1}\; Ma \gtrsim 1 \tag{D 33}$$

For this wavenumber interval, $\min(1, k/Bi) = k/Bi$ and equation (D 33) becomes

$$k^{-1}\; Ma \gtrsim 1 \quad \text{and} \quad k\; Bi^{-2}\; Ma \gtrsim 1 \tag{D 34}$$

This leads to the critical conditions for instability

$$Ma_c \sim Bi \quad \text{with} \quad k_c \sim Bi \quad \text{and} \quad t_c \sim Bi^{-2} \tag{D 35}$$

• $Bi \lesssim k$
In that case, $\delta_H \sim \min(1/k, 1) = 1/k$ and conditions (D 32) are verified. Again the critical Marangoni number, is obtained for the largest time i.e. $t = Bi^{-2}$ corresponding to the largest possible value of $\delta_0(t)$. Moreover, for this wavenumber interval, $\min(1, k\delta_0(t)) = 1$ and equation (D 32) becomes

$$k^{-2}\; Ma\; Bi \gtrsim 1 \quad \text{and} \quad Bi^{-1}\; Ma \gtrsim 1 \tag{D 36}$$

This leads to the same critical conditions (D 35) for instability:

$$Ma_c \sim Bi \quad \text{with} \quad k_c \sim Bi \quad \text{and} \quad t_c \sim Bi^{-2} \tag{D 37}$$

C) Case $1 \ll Bi$ *and* $Bi^{-2} \lesssim t \lesssim 1$

First let us recall from results on the basic flow that the following relation holds:

$$\delta_0(t) \sim \sqrt{t} \quad \text{and} \quad \Delta\theta_0(t) \sim 1 \tag{D 38}$$

From section D.2, one easily verifies that the temperature perturbation field is such that

$$\Delta\hat{\theta} \sim Bi\delta_0\hat{\theta}_s \;\text{ and }\; \hat{\theta} \sim \Delta\hat{\theta} \tag{D 39}$$

Using equations (D 38), (D 39), conditions (D 21) can be transformed into

$$\min\left(1, \frac{\delta_0}{\delta_H}\right)\; \delta_0^{-2}\; \delta_H^2\; Ma\; Bi^{-1} \gtrsim 1 \quad \text{and} \quad \min\left(1, \frac{\delta_0}{\delta_H}\right)\; \delta_H^2\; k^2\; Ma\; Bi^{-1} \gtrsim 1 \tag{D 40}$$

At this stage, two possibilites should be considered : $k \lesssim 1$ and $1 \lesssim k$.
• $k \lesssim 1$
In that case, $\delta_H \sim \min(1/k, 1) = 1$ and $\min(1, \frac{\delta_0}{\delta_H}) \sim \delta_0$. Conditions (D 40) become

$$\delta_0^{-1}\; Ma\; Bi^{-1} \gtrsim 1 \quad \text{and} \quad \delta_0\; k^2\; Ma\; Bi^{-1} \gtrsim 1. \tag{D 41}$$

By multiplying both conditions, one gets:

$$Ma \gtrsim Bi\, k^{-1}. \tag{D 42}$$

This implies that $k_c \sim 1$ and $Ma_c \sim Bi$. Introducing the latter two equalities back into equation (D 41) provides $\delta_0 = 1$ i.e. $t_c \sim 1$. Finally the critical conditions can be written as

$$Ma_c \sim Bi \quad \text{with} \quad k_c \sim 1 \quad \text{and} \quad t_c \sim 1. \tag{D 43}$$

• $1 \lesssim k$

In that instance, $\delta_H \sim \min(1/k, 1) = 1/k$. If one introduces the new variable $\xi \equiv k\delta_0$, conditions (D 40) read

$$Ma \gtrsim BiF(\xi) \quad \text{and} \quad Ma \gtrsim BiG(\xi) \tag{D 44}$$

in which

$$F(\xi) \equiv \xi^2 \min(1, \xi)^{-1} \quad \text{with} \quad G(\xi) \equiv \min(1, \xi)^{-1}. \tag{D 45}$$

A straightforward analysis of these two functions shows that the critical Marangoni is reached for $\xi = 1$ hence $Ma_c \sim Bi$. Moreover, since $1/Bi \lesssim \delta_0 \lesssim 1$ in this time interval, a large bandwith of modes k are equivalent leading to the following critical conditions for instability:

$$Ma_c \sim Bi \quad \text{with} \quad 1 \lesssim k_c \lesssim Bi \quad \text{and} \quad t_c \sim k_c^{-2} \tag{D 46}$$

Finally, by taking the lowest Marangoni numbers of the conditions (D 31)-(D 35)-(D 43)-(D 46), one deduces the true critical conditions for $1 \ll Bi$, namely

$$Ma_c \sim Bi \quad \text{with} \quad 1 \lesssim k_c \lesssim Bi \quad \text{and} \quad t_c \sim k_c^{-2}. \tag{D 47}$$

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