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Onset of convection in binary liquid mixtures : improved Galerkin approximations

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Résumé. — Deux modèles approchés (l'un valable lorsque le nombre de Lewis L est différent de zéro, l'autre lorsque $L = 0$) sont présentés en vue de décrire les conditions d'apparition de la convection dans un mélange binaire lorsque les bords horizontaux sont rigides et imperméables. Les seuils de stabilité ainsi que la fréquence de Hopf sont déterminés analytiquement. Les deux modèles sont comparés avec les approximations précédentes et avec la solution exacte obtenue numériquement.

Abstract. — Two approximate mode truncations (one for nonzero and one for zero Lewis number) are presented in order to describe the onset of convection of a binary mixture between realistic no slip impermeable horizontal boundaries. Stability thresholds as well as the Hopf frequency are determined analytically. The outcomes are compared with earlier approximations and the exact result obtained numerically.

1. Introduction.

In the last few years much effort has been spent by experimentalists [1-5] and theoreticians [6-13] to understand linear and nonlinear properties of convection in a horizontal layer of binary liquid mixtures driven by a vertical temperature gradient. One interesting aspect in this field is the question, how well the boundary conditions (b.c.) at the plates enclosing the layer on top and bottom have to be modelled to get qualitative or even quantitative agreement with the experiments. This question is already important for the onset of convection [6-8]. In contrast to Rayleigh-Bénard convection [14] for an one-component fluid, where the use of idealized free slip b.c. only changes the numerical value of the critical Rayleigh number and the critical wave number, things are different in binary mixtures. Here also the boundary conditions for the concentration field have to be taken into account. In early theoretical works [9, 10] idealized, permeable b.c. have been used which prohibit any convective concentration fluctuations at the plates. Only recently Galerkin approximations [6] have demonstrated that the impermeability b.c. is important for the onset of convection as well as weakly nonlinear convection. Later on numerical stability calculations [7, 8] assuming impermeability of the plates and realistic no slip boundary conditions have provided the *exact treatment* for a lateral

infinite or periodic layer. But for these b.c. the *exact* stability thresholds, the critical wave numbers, and the Hopf frequency are no longer accessible analytically. Since these quantities vary with the three fluid properties Prandtl number, Lewis number, and separation ratio, we feel that

(a) from an experimentalist's point of view there is need to provide simple approximative analytical expressions for the stability quantities which take realistic no slip, impermeable b.c. (NSI) into account and differ by a few percent only from the exact results,

(b) from the theoretical point of view it is instructive to compare the different kinds of b.c. used in the literature to give some more insights into their effects on the stability properties.

These two reasons motivate the investigation presented in this work

2. The system.

The experimental setup we consider consists of a binary fluid layer of height d and mean concentration C_m enclosed between two horizontal, parallel, perfectly heat conducting and impervious plates at $z = -d/2$ and $z = +d/2$. Perpendicular to the plates acts the gravitational field $\mathbf{g} = -g\mathbf{e}_z$. The upper plate is on temperature T_0 , the lower one on $T_0 + \Delta T$ with ΔT being the externally applied vertical temperature gradient.

If lengths are nondimensionalized by layer height d , times by d^2/κ (where κ the thermal diffusivity), temperatures $T(x, z, t)$ by $\kappa\nu/\alpha gd^3$ (where ν is the kinematic viscosity and α the thermal expansion coefficient) and concentrations $C(x, z, t)$ by $\kappa\nu/\beta gd^3$ (where β is the solutal expansion coefficient), the conductive profiles of temperature and concentration read [11]

$$T_{\text{cond}}(z) - T_0 = R \left(\frac{1}{2} - z \right) \quad \text{and} \quad C_{\text{cond}}(z) - C_m = -R\psi z. \quad (1)$$

Here we have introduced the Rayleigh number $R = \alpha gd^3 \Delta T / \kappa \nu$ being the non-dimensionalized temperature difference between the plates and the separation ratio ψ which is proportional to the thermodiffusion ratio k_T depending basically on the mean temperature and mean concentration in the mixture. To facilitate the handling of the impermeable b.c. we replace the concentration field C by

$$\zeta = C - \psi T. \quad (2)$$

Up to a prefactor equation (2) is the potential field of the mass diffusion current, $\mathbf{J}_c = -L \nabla \zeta$. The linearized equations for the vertical velocity field w , the deviation from the conductive temperature profile, $\theta = T - T_{\text{cond}}(z)$, and the ζ -field read in Oberbeck-Boussinesq approximation

$$(\partial_t - \sigma \nabla^2) \nabla^2 w = \sigma \partial_x^2 [(1 + \psi) \theta + \zeta] \quad (3a)$$

$$(\partial_t - \nabla^2) \theta = R w \quad (3b)$$

$$(\partial_t - L \nabla^2) \zeta = -\psi \nabla^2 \theta. \quad (3c)$$

Here $\sigma = \nu/\kappa$ denotes the Prandtl number and $L = D/\kappa$ the Lewis number where D is the concentration diffusivity. Since in (3) the Dufour effect has been ignored, this set of equations is only appropriate [15] to study the onset of convection in *liquid* mixtures like ^3He - ^4He [1] or ethanol/water [2-5].

3. The boundary conditions.

In the literature on convection in binary mixtures [6-13] three types of horizontal b.c. are discussed :

$$(i) \text{ free slip, permeable (FSP) . } w = \partial_z^2 w = \theta = \zeta = 0 \quad \text{at } z = \pm 1/2 ; \quad (4a)$$

$$(ii) \text{ free slip, impermeable (FSI) : } w = \partial_z^2 w = \theta = L \partial_z \zeta = 0 \quad \text{at } z = \pm 1/2 , \quad (4b)$$

$$(iii) \text{ no slip, impermeable (NSI) . } w = \partial_z w = \theta = L \partial_z \zeta = 0 \quad \text{at } z = \pm 1/2 . \quad (4c)$$

As an aside we note that the equations (3) together with the *impermeable* b.c (4b) or (4c) are invariant with respect to a constant shift of the ζ -field, $\zeta(\mathbf{x}, t) \rightarrow \zeta(\mathbf{x}, t) + a$. This invariance is broken for *permeable* b.c

Although NSI are the most realistic b.c. with respect to the experiments the idealized FSI and even FSP b.c. have led to some important insights into stability of the conductive state (FSP b.c., show qualitatively the right global stability behaviour [10] : stationary instability for $R > 0$ and $\psi > 0$, oscillatory instability for $R > 0$ and enough negative ψ , FSI b.c. treated within a Galerkin approximation [6] qualitatively predict the right behaviour at the codimension two (CT) point (where stationary and oscillatory instability meet), like wave number splitting and frequency jump, and the vanishing of the critical wave number for the onset of stationary convection). The « disadvantage » of impermeable b.c. is that the exact stability analysis of the conductive state can no longer be carried out analytically . one has to perform numerical analyses, i.e solving a complex eight order polynomial of the underlying eigenvalue problem [8] or using a shooting method [12, 16]. As an alternative we suggest here simple Galerkin approximations [6] for NSI b.c leading to stability thresholds in good agreement with the numerically determined exact ones.

4. Mode expansion of the fields.

The first step to construct a Galerkin approximation is to expand the spatial dependences of the basic fields in appropriate mode systems *which satisfy the boundary conditions*. For the x dependence of w , θ , and ζ a lateral Fourier expansion in terms of e^{iknx} (where k is the wave number and n is an integer) is suitable since the system is assumed to be laterally periodic or unbounded. To model the z dependence for *no slip, impermeable boundary conditions* (4c) one needs different orthonormal systems reflecting the different boundary conditions for w , θ , and ζ at $z = \pm 1/2$. An appropriate orthonormal system for the no slip b.c $w = \partial_z w = 0$ at $z = \pm 1/2$ are the Chandrasekhar functions [17]. To satisfy $\theta = 0$ at $z = \pm 1/2$ one can use $\{\sqrt{2} \cos \ell \pi z, \sqrt{2} \sin m \pi z ; \text{ with positive odd } \ell \text{ and even } m\}$. For the impermeability condition $L \partial_z \zeta = 0$ the system $\{1, \sqrt{2} \sin m \pi z, \sqrt{2} \cos \ell \pi z ; \text{ with positive even } \ell \text{ and odd } m\}$ seems to be adequate as long as $L > 0$.

5. The three mode approximation for $L > 0$.

We approximate the underlying hydrodynamic fields by using exclusively the basic modes of the above mentioned expansions :

$$w(x, z, t) = [\tilde{w}(t) e^{ikx} + c.c.] C_1(z) \quad (5a)$$

$$\theta(x, z, t) = [\tilde{\theta}(t) e^{ikx} + c.c.] \sqrt{2} \cos \pi z \quad (5b)$$

$$\zeta(x, z, t) = [\tilde{\zeta}(t) e^{ikx} + c.c.] . 1 \quad (5c)$$

Here

$$C_1(z) = \frac{\cosh(\lambda_1 z)}{\cosh(\lambda_1/2)} - \frac{\cos(\lambda_1 z)}{\cos(\lambda_1/2)} \quad (6)$$

denotes the first Chandrasekhar function with $\lambda_1 = 4.73$. Inserting these Galerkin truncations into equations (3a-c) and projecting these equations onto w , θ and ζ respectively leads to a set of ordinary differential equations for the time evolution of the amplitudes $\tilde{w}(t)$, $\tilde{\theta}(t)$ and $\tilde{\zeta}(t)$. Performing standard linear stability analysis of the conductive state, $\tilde{w} = \tilde{\theta} = \tilde{\zeta} = 0$, yields the following results:

(i) *stationary instability* the conductive state becomes linearly unstable towards a *monotonous* growth of small initial perturbations with wave number \hat{k} at the reduced stationary stability threshold

$$r_{\text{stat}}(\hat{k}) = \hat{q}^4(\hat{f}/\hat{g}) \frac{1}{1 + \psi[1 + (\alpha/\beta)(1/\hat{L})]} \quad (7)$$

(ii) *oscillatory instability* the conductive state becomes linearly unstable towards an oscillatory growth of small initial perturbations with frequency ω_H and wave number \hat{k} at the reduced oscillatory stability threshold

$$r_{\text{osc}}(\hat{k}) = \hat{q}^4(\hat{f}/\hat{g}) \frac{(1 + \beta \hat{L})(\bar{\sigma} + 1/\hat{f})(\hat{f} + \beta \hat{L}\bar{\sigma})}{(1 + \hat{f}\bar{\sigma})(1 + \psi) - \alpha\psi} \quad (8)$$

provided that the square of the Hopf frequency ω_H , given by

$$\tau^2 \omega_H^2(\hat{k}) = -\hat{q}^4 \left[\beta^2 \hat{L}^2 + \psi \frac{\alpha(1 + \beta \hat{L})(\hat{f}\bar{\sigma} + \beta \hat{L})}{(1 + \hat{f}\bar{\sigma})(1 + \psi) - \alpha\psi} \right], \quad (9)$$

is positive. Writing down equations (7)-(9) we have introduced the following reductions

$$\hat{k} = k/k_0; \quad r = R/R_0$$

where $k_0 = 3.097$ and $R_0 = 1\,728\,38$ are the critical wave number and critical Rayleigh number for $\psi = 0$. Moreover we have introduced

$$\hat{q}^2(\hat{k}) = [\hat{k}^2 + \pi^2/\hat{k}_0^2]/[1 + \pi^2/\hat{k}_0^2] \quad (10a)$$

$$\hat{g}(\hat{k}) = \hat{k}^2(1 - \gamma)/(\hat{k}^2 - \gamma) \quad (10b)$$

$$\hat{f}(\hat{k}) = \hat{g}(\hat{k})[\hat{k}^4 - 2\hat{k}^2\gamma + \lambda]/[(1 - 2\gamma + \lambda)\hat{q}^2\hat{k}^2] \quad (10c)$$

and $\hat{L}(\hat{k}) = (\hat{k}^2/\hat{q}^2)L/3$, $\bar{\sigma} = 1.944\sigma$, $\tau = 1/(k_0^2 + \pi^2) = 0.0514$. The constants $\gamma = -1\,282$, $\lambda = 5.437$, $\alpha = 0.758$, and $\beta = 1.479$ arise from projections of the field equations. Let us note that setting $\psi = 0$ reproduces the results of Niederländer *et al.* [14] for Rayleigh-Bénard convection in a pure fluid. There the approximate critical wave number k_0 and the critical Rayleigh number R_0 agree within 0.8% and 1.2%, respectively, with the exact results [17].

As in the earlier FSI approximation [6] our stability thresholds and the Hopf frequency are explicitly wave number dependent. To get the *critical* reduced Rayleigh numbers one has to minimize $r_{\text{stat}}(\hat{k})$ and $r_{\text{osc}}(\hat{k})$ with respect to \hat{k} .

Let us now discuss our approximative results for two typical liquid mixtures used in recent experiments, ethanol-water with $\sigma = 10$, $L = 0.01$ and ${}^3\text{He}-{}^4\text{He}$ with $\sigma = 0.6$, $L = 0.03$. We compare with the corresponding results of the FSI approximation [6] and the exact results obtained numerically in the same way as in reference [8]. In all figures we mark the exact values by full lines, the NSI approximation by dashed lines and the FSI approximation by dotted lines.

In figure 1 we present the critical stationary Rayleigh numbers r_{stat}^c for positive ψ and the corresponding critical wave numbers \hat{k}_{stat}^c . Figure 1a and b represent ethanol water mixtures, figure 1c and d the helium mixture. For both mixtures one finds that the FSI approximation underestimates the onset of stationary convection by a factor of 1/2, whereas our NSI three mode approximation deviates from the exact result less than 8%. The stationary critical wave number tends in all three cases to zero, but the crossing point ψ_0 with the ψ axis significantly differs. In all three cases one can derive an analytical expression

$$\psi_0 = \frac{L}{f - L}$$

where $f = 0.26$ for the exact solution [8], $f = 0.37$ in our NSI approximation and $f = 1.62$ in the FSI approximation [6]. In contrast to the FSI approximation our NSI approximation shows the right topological behaviour of \hat{k}_{stat}^c .

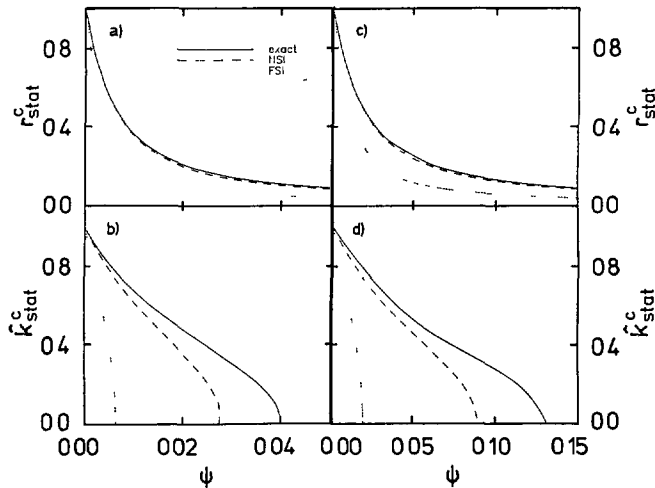


Fig. 1. — Critical stationary Rayleigh numbers r_{stat}^c and critical stationary wave numbers \hat{k}_{stat}^c as function of the separation ratio ψ for ethanol-water mixtures a) and b) with $\sigma = 10$, $L = 0.01$ and for ${}^3\text{He}-{}^4\text{He}$ mixtures c) and d) with $\sigma = 0.6$ and $L = 0.03$. Full lines exact numerical solution for NSI b c, dashed lines our NSI approximation, dotted lines FSI approximation

In figure 2 we present the critical oscillatory Rayleigh numbers r_{osc}^c , the corresponding critical wave numbers \hat{k}_{osc}^c and the Hopf frequencies $\omega_H = \omega_H(\hat{k}_{\text{osc}}^c)$ for the two liquids (a-c for ethanol-water, d-f for helium mixtures). In that case the oscillatory stability threshold for the FSI approximation deviates about 20% from the exact result, whereas our NSI approximation less than 3%. We find qualitative change due to the NS b c. for the critical wave number in

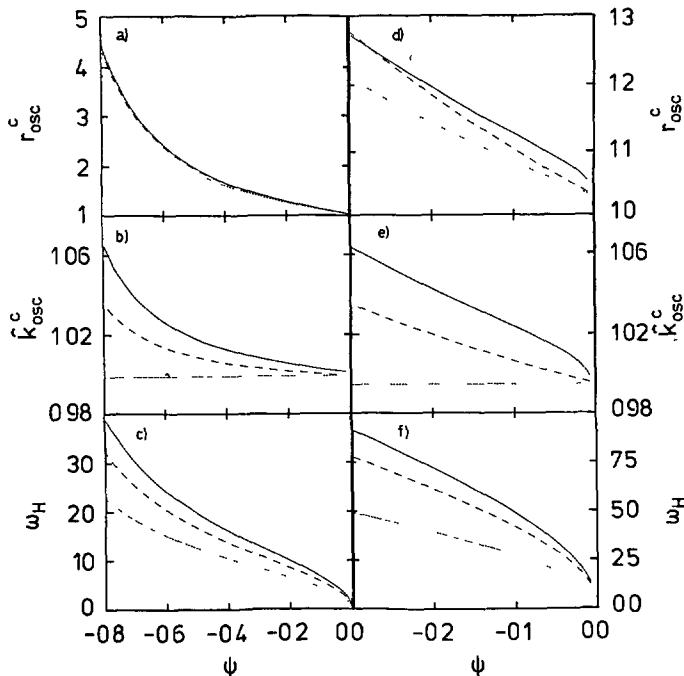


Fig 2 — Critical oscillatory Rayleigh numbers r_{osc}^c , critical oscillatory wave numbers \hat{k}_{osc}^c , and Hopf frequencies ω_H as function of the separation ratio ψ a)-c) for ethanol-water mixtures, d)-f) for helium mixtures σ, L and meaning of the line types as in figure 1

contrast to the FSI approximation where \hat{k}_{osc}^c is ψ independent there is a slight growth of \hat{k}_{osc}^c for NSI b.c as in the exact result Also here the accuracy is of the order of 2-3 % The Hopf frequency is worse approximated as one can see e.g. for ethanol-water mixtures in figure 2c ω_H differs for the FSI approximation by 38 %, but for the NSI approximation by 18 %.

Due to the weak ψ -dependence of the critical wave number \hat{k}_{osc}^c it is reasonable to approximate r_{osc}^c and ω_H by evaluating (8), (9) at the fixed wave number $\hat{k} = 1$ This simplifies equations (10) remarkably since $\hat{q}^2(\hat{k} = 1) = \hat{f}(\hat{k} = 1) = \hat{g}(\hat{k} = 1) = 1$, and $\hat{L}(\hat{k} = 1) = L/3$ One finds

$$r_{osc}(\hat{k} = 1) = \frac{(1 + \beta \hat{L})(1 + \bar{\sigma})(1 + \beta \hat{L}/\bar{\sigma})}{(1 + \bar{\sigma})(1 + \psi) - \alpha \psi}, \tag{11}$$

$$\tau^2 \omega_H^2(\hat{k} = 1) = - \left[\hat{L}^2 + \psi \frac{\alpha (1 + \beta \hat{L})(\bar{\sigma} + \beta \hat{L})}{(1 + \bar{\sigma})(1 + \psi) - \alpha \psi} \right] \tag{12}$$

These expressions for $\hat{k} = 1$ can not be distinguished from the corresponding dashed curves in figure 2 where $\hat{k} = \hat{k}_{osc}^c(\psi)$. Since α in the denominator of (11) and (12) arises from the projection procedure, one has to expect that the errors in our r_{osc} and ω_H increase whenever the absolute value of $[1 - \alpha/(1 + \bar{\sigma})] \psi$ is of the order 1. Thus the larger the Prandtl number and the smaller the absolute value of the separation ratio ψ , the better (11) and (12) work.

The error increases for σ - ψ -combinations where the nominator in (11), (12) becomes small. In comparison to $r_{osc}(\hat{k} = 1)$ for FSI b.c. equation (11) differs by the prefactor 1.479 in front of L and by a prefactor 1.944 in front of the Prandtl number σ . Furthermore the FSI-factor $8/\pi^2 \approx 0.811$ in the nominator is exchanged by $\alpha \approx 0.758$. In comparison to the calculation [6] of $r_{stat}(\hat{k} = 1)$ with FSI b.c. only the prefactor in front of \hat{L} in (7) is diminished by a factor of 0.632. Thus for fixed wave number $\hat{k} = 1$ and not to large negative ψ FSI and NSI approximation are formally connected *via* a simple scaling of the physical parameters L and σ . But the scaling of L differs for the two types of instabilities.

To summarize The assumption of free slip b.c. underestimates in general the stability thresholds, the critical wave numbers and the Hopf frequency, partly up to a factor 1/2. We emphasize that all critical Rayleigh numbers discussed here are reduced quantities scaled by their corresponding values at $\psi = 0$. Thus the unscaled Rayleigh numbers for FSI and NSI b.c. differ by a factor of about 5.

The simplicity of our mode truncation ignores the boundary layer behaviour of the marginal stable concentration field found in the numerical solution for $\psi \leq -0.2$. But from the accuracy of our approximation we infer that this detail seems to have only a minor influence on the stability thresholds.

6. Three mode approximation for $L = 0$.

The Lewis number of typical liquid binary mixtures is of the order 10^{-2} . This suggests to discuss as a further reasonable approximation the limit $L = 0$. Since the stationary instability is quite sensitive to $L = 0$ (one finds $r_{stat} = 0$ for $\psi > 0$, and $r_{stat} = 1$ for $\psi = 0$, and for $\psi < 0$ there is a stationary instability at $r_{stat} = 0$ when the layer is heated from above) this approximation is useful for the oscillatory instability only as already noticed in reference [7]. But when setting $L = 0$ one has to remember that the impermeability $L \partial_z \zeta = 0$ at the plates is guaranteed independently of the mode truncation of ζ . Thus with regard to equation (3c), $\partial_t \zeta = -\psi \nabla^2 \theta$, a spatial ζ -field expansion in the same manner as for the θ -field should be more appropriate than the one used in chapter 4. Thus for $L = 0$ also $\zeta = 0$ at the plates has to hold implying that there is no distinction between permeability b.c. and impermeability b.c.

Substituting (5c) by

$$\zeta(x, z, t) = [\tilde{\zeta}(t) e^{ikx} + c.c.] \sqrt{2} \cos \pi z \tag{13}$$

leads after performing standard stability analysis and setting again $\hat{k} = 1$ to

$$r_{osc} = \frac{1}{1 + a\psi} \tag{14}$$

For the squared Hopf frequency one obtains

$$\omega_H^2 = b \frac{\psi}{1 + c\psi} \tag{15}$$

where

$$a = c = 1 - \rho / (1 + \bar{\sigma}) \tag{16a}$$

$$b = -\bar{\sigma} \rho / [(1 + \bar{\sigma}) \tau^2] \tag{16b}$$

and $\bar{\sigma} = 1.944 \sigma$, $\rho = \pi / (2 \sqrt{2})$, and $1/\tau = 19.461$. In particular for $\sigma = 10$ we find $a = 0.946$ and $b = 399$. This is in good agreement with the exact numerical result [16] for

$L = 0$, $\sigma = 10$ and not too large negative ψ which can be fitted to $a = 0.937$, $b = 373$ and $c = 0.968$. Here the error of our approximative expression for r_{osc} is about $(10^{-2}|\psi|)\%$, the accuracy of ω_{H} is about 3.3%. Semiquantitative formulas [18] for r_{osc} and ω_{H}^2 , used by the experimentalists [4, 5], can also be brought into the form of (14) and (15). For $\sigma = 10$ they yield $a = c = 0.91$ and $b = 407$ being a little bit worse than our result. Setting $L = 0$ in our $L \neq 0$ approximation, equations (11) and (12), leads to $a = 0.94$ and $b = 273$. In particular the slope of ω_{H}^2 differs significantly (by about 26%) from the exact result, which reflects the fact that the mode truncation leading to (11) and (12) is not appropriate in the limit $L = 0$.

Let us finally note that the $L = 0$ result coincides within the pencil's width with the numerically determined exact results for $L = 0.01$ (water ethanol) shown in figures 2a and 2c. Obviously for small Lewis number (e.g. water ethanol mixture) one has to prefer the $L = 0$ formulas (14), (15) rather than the $L \neq 0$ approximation of Chapter 5. However if one is interested in the critical wavenumber dependence or the stationary instability one has to use (7)-(9).

Why a $L = 0$ approximation works very well also for small nonzero Lewis numbers seems to be caused by the basic mechanism producing the oscillatory instability: Stern [19] has argued that opposite conductive temperature and concentration gradients and a fast relaxation of temperature fluctuations in comparison to concentration fluctuations, i.e. $\kappa \gg D$ or $\bar{L} \ll 1$, can lead to an oscillatory rising and falling of small fluid volumes for appropriate r and ψ . This scenario seems to be quite insensitive to the limit $D \rightarrow 0$ or $L \rightarrow 0$.

7. Summary and discussion.

We have given a simple approximate analytical treatment of the stability properties of the conductive state in binary fluid mixtures in the Rayleigh-Bénard geometry with regard to the realistic no slip impermeable (NSI) boundary conditions at the plates. The critical stationary and oscillatory instabilities of our $L \neq 0$ Galerkin approximation agree well with the exact numerical ones as demonstrated for water-ethanol and ^3He - ^4He mixtures. The critical wave numbers and the Hopf frequency show the right qualitative behaviour and approximate much better the exact numerical results than an earlier free slip, impermeable (FSI) approximation. We have not discussed the immediate vicinity of the CT point since there the Oberbeck-Boussinesq approximation fails [20]. As an alternative to the NSI approximation for $L > 0$ we have also presented a NSI mode approximation for $L = 0$ which is a reasonable way to estimate the behaviour on the oscillatory branch for low Lewis number liquids.

To improve further the approximative stability properties one has to extend the mode expansion for the z -dependence of the hydrodynamic fields. To find out which mode seems to be the next important one we remember that the z -component of the velocity field w and the temperature field θ each fulfill two boundary conditions at each plate: $w = \partial_z w = 0$ and $\theta = \partial_z^2 \theta = 0$ at $z = \pm 1/2$ ($\partial_z^2 \theta = 0$ follows directly from (3b) by using $w = \theta = 0$ at $z = \pm 1/2$). However the ζ field fulfills *only one* boundary condition: $\partial_z \zeta = 0$. A second one can easily be calculated from (3c) and $\partial_z^2 \theta = 0$ at $z = \pm 1/2$. One finds that $\partial_z^2 \zeta$ has to be proportional to ζ at the plates. To fulfill this condition one should extend the ζ field truncation in (5c) by an additional term $\sim \cos 2\pi z$. Carrying out the calculation of the stability properties [21] (which we do not want to present here since the additional mode blows up considerably the analytical formulas) shows that there is indeed no appreciable improvement of the stability thresholds but the critical wave numbers and the Hopf frequency are better approximated. Roughly speaking, there the differences between exact numerical results and three mode approximation are halved in the extended version.

Based on our approximate truncation for the critical modes a Galerkin model similar to the one given in [6, 11] can be constructed. For details we refer to reference [21]

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