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Bénard convection in a binary mixture with a nonlinear density-temperature relation

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Bénard convection of a two-component liquid in a porous medium is considered. The mixture displays Soret effects and shows a nonlinear density-temperature relation, i.e., non-Boussinesq properties. A two-parameter perturbation analysis is used to determine the effects on the stability of the basic state and the finite-amplitude convection. Linear theory demonstrates the nonlinear density-temperature relation to have a destabilizing effect; the critical Rayleigh numbers for the onset of oscillatory and steady-state convection are decreased. For the case of two-dimensional convection, traveling waves and steady-state solutions are considered. They are determined up to fifth order of the amplitude parameter. In the vicinity of the codimension-two point, a stable branch of traveling wave solutions exists near the onset. If the Rayleigh number is increased the wave motion vanishes and a transition to steady-state convection occurs. Due to the symmetry of the two-dimensional solutions this bifurcation is not affected by the non-Boussinesq properties of the mixture. However, for the case of three-dimensional convection the nonlinear density-temperature relation leads to an unfolding of the bifurcation of steady-state hexagonal solutions. As a consequence the branch of the oscillatory solution terminates at an isolated point in the parameter plane.

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I. INTRODUCTION

In a two-component liquid layer heated from below and cooled from above the imposed temperature gradient can generate a concentration gradient as well. This molecular separation phenomenon is known as the Soret effect [1,2]. In binary mixtures with negative Soret effects, characterized by negative separation ratios $\psi<0$, the more dense component migrates towards the lower warm boundary [3]. Hence buoyancy forces are reduced and the layer is stabilized by the induced concentration stratification. In such a system the essential results of linear stability analyses [4–12] can be described in terms of the separation ratio ψ. For $\psi<\psi_{CT}$ and $\psi_{CT}<0$ the basic state of heat conduction becomes initially unstable to oscillatory perturbations as the temperature difference across the height of the layer exceeds a critical value. For $\psi>\psi_{CT}$ a steady-state instability occurs. The case $\psi=\psi_{CT}$ denotes the codimension-two point (CTP) where both instabilities coincide at the same critical value of the control parameter.

In previous years the nonlinear convection in binary mixtures, subject to negative Soret effects, has stimulated an enormous amount of effort both theoretically and experimentally; for details see, e.g., [13–25] and the recent surveys of the literature by Schöpf [26], Schöpf and Zimmermann [27], and Zimmermann, Müller, and Davis [28]. Focusing on the vicinity of the CTP, i.e., $\psi_{CT}-\psi\ll 1$, the main experimental observations and theoretical findings can be summarized as follows: For slightly supercritical temperature differences across the layer traveling waves (TWs), consisting of a laterally propagating roll pattern, are the preferred mode at the onset of convection. Increasing the forcing temperature difference the amplitude of the wave increases while its phase velocity decreases. At an even higher value of the temperature difference the propagation of the wave ceases, and a transition to steady-state two-dimensional convection occurs. In some cases the TWs may lose stability to modulated traveling waves before the transition takes place.

In the present paper we investigate the influence of a nonlinear density-temperature relation on the Bénard convection in a binary liquid layer. We consider a mixture in a porous medium [29–31]. In this case similar results as in bulk fluid mixtures are expected while the analysis is simplified. The work is motivated by experiments on Bénard convection performed by Zimmermann and Müller [32]. These experiments were conducted in a liquid mixture of 15 wt. % ethyl alcohol in water at various mean temperatures. Zimmermann and Müller [32] observed that stable TWs are suppressed when the mean temperature is close enough to the solidification temperature of the mixture. Instead, a steady-state three-dimensional convection pattern is identified. They conjectured that this observation is due to the presence of significant nonlinearities in the density profile of the basic state at low mean temperatures. Studies of convection in single-component fluids demonstrate that these non-Boussinesq properties lead to a preference of three-dimensional hexagonal patterns at onset of convection; see [33–38].

The paper is organized as follows. In Sec. II the governing equations and the method of solution are presented. The results of a linear stability analysis are

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given in Sec. III. Section IV shows results of the finite-amplitude analysis. In the case of two-dimensional convection the TW solution and the steady-state solution are calculated up to fifth order in the amplitude. A bifurcation diagram is shown representing the situation in the vicinity of the CTP. We discuss the stability of these solutions using standard results of bifurcation theory [39–41]. Furthermore in Sec. IV we calculate three-dimensional oscillatory and steady-state solutions with hexagonal symmetry. For these solutions a bifurcation diagram valid near the CTP is derived. The main results of our study are summarized in Sec. V. To obtain a readable text details of rather lengthy calculations are presented separately in the Appendix.

II. FORMULATION OF THE PROBLEM

A. Governing equations

The basic equations describing thermal convection in a horizontal layer of a binary liquid mixture are the conservation equations of mass, solute and energy. In addition we use Darcy's law to model the conservation of momentum in a porous medium. The full set of equations is

$$\nabla\cdot\mathbf{v}=0\ , \tag{2.1a}$$

$$\partial_t C+(\mathbf{v}\cdot\nabla)C=-\nabla\cdot\mathbf{j}\ , \tag{2.1b}$$

$$\partial_t T+(\mathbf{v}\cdot\nabla)T=\kappa\nabla^2 T\ , \tag{2.1c}$$

$$\mathbf{v}=-\frac{K}{\mu}(\nabla p+\rho_f g\mathbf{e}_z),\quad \mathbf{e}_z=(0,0,1)\ . \tag{2.1d}$$

Here $\mathbf{v}=(u,v,w)$ is the filtration velocity, C the concentration of the solute, T the temperature, and p the pressure. Material properties are the thermal conductivity κ of the layer, the dynamic viscosity μ of the mixture, and the permeability K of the porous medium. In Eq. (2.1d), g is the acceleration of gravity acting vertically downward. Furthermore, in this equation the density ρ_f of the mixture is given by the nonlinear relation

$$\rho_f=\rho^*[1-\alpha(T-T^*)-\beta(T-T^*)^2-\alpha'(C-C^*)]\ , \tag{2.2}$$

where α and α' are the coefficients of thermal and solutal expansion and β denotes the nonlinear coefficient of thermal expansion. We define the reference density ρ^* at the mean temperature T^* and the mean concentration C^* within the mixture. In Eq. (2.1b) the solute flux j, allowing for Soret diffusion, takes the form

$$\mathbf{j}=-D_0[\nabla C-S_0C^*(1-C^*)\nabla T]\ . \tag{2.3}$$

Here D_0 is the coefficient of mass diffusion and S_0 is the Soret diffusion coefficient. All the coefficients are assumed to be constant.

We consider a layer of height h and of infinite extend in the horizontal x-y plane subject to fixed temperature and concentration at the top ($z=h$) and bottom ($z=0$). In this case the boundary conditions read as

$$w=0,\quad T=T_0,\quad C=C_0\quad \text{at } z=0\ , \tag{2.4a}$$

$$w=0,\quad T=T_1,\quad C=C_1\quad \text{at } z=h\ . \tag{2.4b}$$

Using these idealized conditions the boundaries are assumed to be permeable for the solute flux. Hence the present problem qualitatively corresponds to the thermohaline problem [42–48] in which the concentration gradient is not induced by Soret effects but externally imposed.

The system possesses a basic state of heat conduction characterized by $\mathbf{v}'=0$ and $\mathbf{j}'=0$, a hydrostatic pressure distribution and linear temperature and concentration profiles. We find

$$T'(z)=T^*-(T_0-T_1)\left[\frac{z}{h}-\frac{1}{2}\right]\ , \tag{2.5a}$$

$$C'(z)=C^*-S_0C^*(1-C^*)(T_0-T_1)\left[\frac{z}{h}-\frac{1}{2}\right]\ . \tag{2.5b}$$

Moreover, these equations define the mean values T^*,C^* and the values of C_0 and C_1 used in the boundary conditions (2.4).

In the following we are interested in dimensionless deviations from the basic state. Using the scales

$$(x,y,z)\propto h,\quad t\propto\frac{h^2}{\kappa},\quad \mathbf{v}\propto\frac{\kappa}{h},\quad p\propto\frac{\kappa\mu}{K}\ , \tag{2.6}$$

$$T-T^*\propto T_0-T_1,\quad C-C^*\propto -S_0C^*(1-C^*)(T_0-T_1)\ ,$$

eliminating the pressure and introducing the field $S=T+C$ eventually leads to a system of nonlinear perturbation equations. We obtain

$$\nabla^2 w=R\,\nabla_H^2[\{1+\psi-2\gamma(z-\tfrac{1}{2})\}T+\gamma T^2-\psi S]\ , \tag{2.7a}$$

$$\partial_t T+(\mathbf{v}\cdot\nabla)T=w+\nabla^2 T\ , \tag{2.7b}$$

$$\partial_t S+(\mathbf{v}\cdot\nabla)S=L\nabla^2 S+\nabla^2 T\ , \tag{2.7c}$$

and the homogenous boundary conditions

$$w=0,\quad T=0,\quad S=0\quad \text{at } z=0,1\ . \tag{2.8}$$

In this representation the horizontal components of the velocity vector can be expressed in terms of the vertical component:

$$\nabla_H^2 u=-\partial_x\partial_z w,\quad \nabla_H^2 v=-\partial_y\partial_z w\ , \tag{2.9}$$

where ∇_H^2 denotes the horizontal Laplacian. The system (2.7) is characterized by four dimensionless groups, the Rayleigh number R, the separation ratio ψ, the non-Boussinesq number γ, and the Lewis number L:

$$R=\frac{\alpha g(T_0-T_1)Kh}{\kappa(\mu/\rho^*)},\quad \psi=S_0C^*(1-C^*)\frac{\alpha'}{\alpha}\ ,$$
$$\gamma=\frac{\beta}{\alpha}(T_0-T_1),\quad L=\frac{D_0}{\kappa}\ . \tag{2.10}$$

We consider R as the forcing parameter of convection. ψ is an additional control parameter characterizing Soret effects. The parameter γ quantifies the nonlinearity of the density profile. The Lewis number L defines the dimensionless time scale of molecular diffusion. For typical water/ethanol mixtures at low mean temperatures L is of the order 10^{-3} [11].

B. Method of solution

We use a perturbation analysis [49,50] to solve Eqs. (2.7) subject to the boundary conditions (2.8). Regarding the amplitude of convection ε and the non-Boussinesq number γ as small parameters, the variables of state may be expressed in form of a double series:

$$(w,T,S)=\sum_{n=1,m=0}\varepsilon^n\gamma^m(w_{nm},T_{nm},S_{nm}) . \qquad (2.11)$$

The forcing parameter R and the frequency ω of oscillatory solutions are likewise expanded:

$$R=\sum_{n=1,m=0}\varepsilon^{n-1}\gamma^m R_{n-1,m} , \qquad (2.12a)$$

$$\omega=\sum_{n=1,m=0}\varepsilon^{n-1}\gamma^m \omega_{n-1,m} . \qquad (2.12b)$$

In writing Eq. (2.12b) we are looking for time-periodic solutions, characterized by $\partial_t(w,T,S)=i\omega q(w,T,S)$. Here $i=\sqrt{-1}$ and q is a real multiplier which will be defined later on. Inserting expansions (2.11) and (2.12) into the nonlinear equations (2.7) and (2.8) and collecting terms of equal power in the small parameters ε and γ results in a sequence of linear inhomogenous equations. This sequence can be successively solved starting with the eigenvalue problem of the order $\varepsilon^1\gamma^0$. The coefficients of expansion of higher order, $R_{n-1,m}$ and $\omega_{n-1,m}$ with $n>1$ and $m>0$, respectively, are determined from solvability conditions for inhomogenous systems of differential equations. Then, for a fixed parameter γ, Eq. (2.12a) defines the amplitude ε as a function of the Rayleigh number R. Moreover, from Eq. (2.12b) by demanding $\omega^2\geq 0$ we determine the parameter range in which oscillatory solutions exist. To evaluate the linear stability problem ($n=1$) the coefficients are calculated up to the order γ^2. In the case of two- and three-dimensional finite-amplitude convection ($n>1$) we calculate the coefficients up to the order ε^5 and ε^3, respectively. Hence the interrelation between the parameters used in the perturbation expansion are assumed to be $\gamma\propto\varepsilon^2$ and $\gamma\propto\varepsilon$, respectively.

III. LINEAR STABILITY ANALYSIS

To the order $\varepsilon^1\gamma^0$ we obtain the eigenvalue problem

$$\mathbf{L}\{\mathbf{X}_{10}\}\equiv\begin{bmatrix} D^2-k^2 & R_{00}k^2(1+\psi) & -R_{00}k^2\psi \\ 1 & D^2-k^2-i\omega_{00}q & 0 \\ 0 & D^2-k^2 & L(D^2-k^2)-i\omega_{00}q \end{bmatrix}\begin{bmatrix} w_{10} \\ T_{10} \\ S_{10}\end{bmatrix}=\mathbf{0} , \qquad (3.1)$$

and the boundary conditions

$$\mathbf{X}_{10}=\mathbf{0} \quad \text{at } z=0,1 . \qquad (3.2)$$

Here ∂_z is denoted by D. Furthermore we have introduced the horizontal wave number k according to the lateral periodicity of the first order solutions, i.e., $\nabla_H^2\mathbf{X}_{1m}=-k^2\mathbf{X}_{1m}$. Since the boundary conditions (3.2) suggest a solution of the form $\mathbf{X}_{10}(z)\propto\sin\pi z$, Eq. (3.1) can be reduced to a set of homogenous algebraic equations. The evaluation of the solvability condition of this set yields the bifurcation points $R_{00}^{(\text{st})}$ and $R_{00}^{(\text{os})}$ of steady-state ($\omega=0$) and oscillatory ($\omega\neq 0$) solutions, respectively. According to results of Brand and Steinberg [29] and Knobloch [31] we find

$$R_{00}^{(\text{st})}=\frac{q^2}{k^2}\{1+\psi(1+L^{-1})\}^{-1} \qquad (3.3)$$

and

$$R_{00}^{(\text{os})}=\frac{q^2}{k^2}\frac{1+L}{1+\psi} , \qquad (3.4a)$$

$$\omega_{00}^2=-\left[L^2+\psi\frac{1+L}{1+\psi}\right] , \qquad (3.4b)$$

where $q=\pi^2+k^2$. The solution $\mathbf{X}_{10}(z)$ of Eq. (3.1) together with the solution $\mathbf{X}_{10}^*(z)$ of the adjoint problem are given in the Appendix [Eqs. (A1) and (A2)].

To the order $\varepsilon^1\gamma^1$ we have to solve the inhomogenous equations

$$\mathbf{L}\{\mathbf{X}_{11}\}=\begin{bmatrix} -R_{01}k^2[(1+\psi)T_{10}-\psi S_{10}]+2R_{00}k^2(z-\frac{1}{2})T_{10} \\ i\omega_{01}qT_{10} \\ i\omega_{01}qS_{10}\end{bmatrix} \qquad (3.5)$$

with respect to the boundary conditions

$$\mathbf{X}_{11}=\mathbf{0} \quad \text{at } z=0,1 \; . \tag{3.6}$$

The solvability of Eq. (3.5) requires

$$\langle \mathbf{L}\{\mathbf{X}_{11}\},\mathbf{X}_{10}^{*}\rangle=0 \; , \tag{3.7}$$

where the angular brackets denote the weighted scalar product. By this condition for the right-hand side of Eq. (3.5) resonant terms are eliminated. The evaluation of relation (3.7) gives

$$R_{01}=0 \; , \tag{3.8a}$$

$$\omega_{01}=0 \; . \tag{3.8b}$$

A partial solution $\mathbf{X}_{11}(z)$ of Eq. (3.5) can be easily obtained by expanding the remaining inhomogenity in terms of a rapidly converging Fourier's series [see Appendix, Eq. (A3)].

To the order $\varepsilon^1\gamma^2$ the set of equations and the corresponding boundary conditions are

$$\mathbf{L}\{\mathbf{X}_{12}\}=\begin{bmatrix} -R_{02}k^2\{(1+\psi)T_{10}-\psi S_{10}\}+2R_{00}k^2(z-\tfrac{1}{2})T_{11} \\ i\omega_{02}qT_{10} \\ i\omega_{02}qS_{10} \end{bmatrix} , \tag{3.9}$$

$$\mathbf{X}_{12}=\mathbf{0} \quad \text{at } z=0,1 \; . \tag{3.10}$$

The evaluation of the solvability condition of this problem, i.e.,

$$\langle \mathbf{L}\{\mathbf{X}_{12}\},\mathbf{X}_{10}^{*}\rangle=0 \; , \tag{3.11}$$

leads to the following coefficients of expansion (2.12a) and (2.12b):

$$R_{02}^{(\mathrm{st})}=-\frac{R_{00}^{(\mathrm{st})}}{[1+\psi(1+L^{-1})]^2}[f_1(q)+f_2(q)] \tag{3.12}$$

and

$$R_{02}^{(\mathrm{os})}=-\frac{R_{00}^{(\mathrm{os})}}{[1+\psi]^2}\left[f_1(q)\left\{1+\frac{3\pi^2L^3(q+3\pi^2)\left[1-\dfrac{q}{3\pi^2}L\right]}{L^2(q+3\pi^2)^2+\omega_{00}^2q^2(1+L)^2}\right\}+f_2(q)\left\{1+\frac{15\pi^2L^3(q+15\pi^2)\left[1-\dfrac{q}{15\pi^2}L\right]}{L^2(q+15\pi^2)^2+\omega_{00}^2q^2(1+L)^2}\right\}\right] , \tag{3.13a}$$

$$2\omega_{00}\omega_{02}=-\frac{L^2+\omega_{00}^2}{[1+\psi]^2}\left[f_1(q)\left\{1+\frac{L^3(q+3\pi^2)^2\left[1+\dfrac{q(1+L)}{3\pi^2}\right]}{L^2(q+3\pi^2)^2+\omega_{00}^2q^2(1+L)^2}\right\}+f_2(q)\left\{1+\frac{L^3(q+15\pi^2)^2\left[1+\dfrac{q(1+L)}{15\pi^2}\right]}{L^2(q+15\pi^2)^2+\omega_{00}^2q^2(1+L)^2}\right\}\right] . \tag{3.13b}$$

Here an implicit representation of the results is used where

$$f_1(q)=\frac{(32q)^2}{9^2\times 3\pi^6(2q+3\pi^2)} , \quad f_2(q)=\frac{(64q)^2}{225^2\times 15\pi^6(2q+15\pi^2)} \; . \tag{3.14}$$

Minimization of the expressions for the Rayleigh numbers with regard to the wave number k leads to the critical conditions for the onset of steady-state and oscillatory convection. After some lengthy algebra we obtain

$$R_c^{(\mathrm{st})}=\frac{4\pi^2}{1+\psi(1+L^{-1})}\left[1-\gamma^2\frac{f_1+f_2}{\{1+\psi(1+L^{-1})\}^2}\right] , \tag{3.15}$$

$$k_c^{(\mathrm{st})^2}=\pi^2\left[1+\gamma^2\frac{\frac{10}{7}f_1+\frac{34}{19}f_2}{\{1+\psi(1+L^{-1})\}^2}\right] , \tag{3.16}$$

and

$$R_c^{(\mathrm{os})}=4\pi^2\frac{1+L}{1+\psi}\left[1-\frac{\gamma^2}{(1+\psi)^2}\left\{f_1\left[1+\frac{15L^3(1-\frac{2}{3}L)}{25L^2+4\omega_{00}^2(1+L)^2}\right]+f_2\left[1+\frac{255L^3(1-\frac{2}{15}L)}{289L^2+4\omega_{00}^2(1+L)^2}\right]\right\}\right] , \tag{3.17}$$

$$\omega_c^2=-L^2-\psi\frac{1+L}{1+\psi}\left[1-\frac{\gamma^2}{(1+\psi)^2}\left\{f_1\left[1+\frac{35L^3(1+\frac{2}{7}L)}{25L^2+4\omega_{00}^2(1+L)^2}\right]+f_2\left[1+\frac{323L^3(1+\frac{2}{19}L)}{289L^2+4\omega_{00}^2(1+L)^2}\right]\right\}\right], \tag{3.18}$$

$$k_c^{(\mathrm{os})^2}=\pi^2\left[1+\frac{\gamma^2}{(1+\psi)^2}\left\{\tfrac{10}{7}f_1\left[1+\tfrac{9}{5}L^3\frac{150L^2(1-\frac{71}{54}L)-4\omega_{00}^2(1+L)^2(1+\frac{29}{9}L)}{[25L^2+4\omega_{00}^2(1+L)^2]^2}\right]\right.\right.$$
$$\left.\left.+\tfrac{34}{19}f_2\left[1+\tfrac{225}{17}L^3\frac{17^2\times 18L^2(1-\frac{863}{4050}L)-4\omega_{00}^2(1+L)^2(1+\frac{293}{225}L)}{[289L^2+4\omega_{00}^2(1+L)^2]^2}\right]\right\}\right], \tag{3.19}$$

where

$$f_1=\frac{4\times 32^2}{9^2\times 7\times 3\pi^4},\quad f_2=\frac{4\times 64^2}{225^2\times 19\times 15\pi^4}. \tag{3.20}$$

Figure 1 shows the stability diagram of the present problem. The critical Rayleigh numbers $R_c^{(\mathrm{st})}$ and $R_c^{(\mathrm{os})}$ given by the Eqs. (3.15) and (3.17), respectively, are plotted versus the separation ratio ψ. Two different cases are illustrated: The dashed curves correspond to the well-known case $\gamma=0$ [29,31]. The new results for a binary mixture with a nonlinear density-temperature relation, characterized by $\gamma\neq 0$, are given by solid curves. We find the non-Boussinesq properties to have a destabilizing effect; for a fixed value of ψ the critical Rayleigh numbers are decreased. This result holds independent of the sign of the parameter γ since the critical values depend on γ^2. In single-component fluids ($\psi=0$) the destabilizing effect of a nonlinear density profile was already identified by Busse [35,36].

Following Refs. [9–16,26–28] we define the CTP at $\psi=\psi_{\mathrm{CT}}$ by setting $R_c^{(\mathrm{os})}=R_c^{(\mathrm{st})}$; cf. Eqs. (3.15) and (3.17). We find that the CTP shifts to lower values of the separation ratio when the nonlinear density profile is present; see Fig. 1. We obtain

$$\psi_{\mathrm{CT}}=\frac{-L^2}{1+L+L^2}[1+\gamma^2(1+L+L^2)\{\tfrac{7}{5}f_1+\tfrac{19}{17}f_2\}]. \tag{3.21}$$

Hence, for $\gamma\neq 0$ the parameter range in which an oscillatory instability occurs is obviously reduced. Moreover, we define the value $\psi=\tilde{\psi}$ by calculating Eq. (3.18) in the limit $\omega_c^2=0$. We find

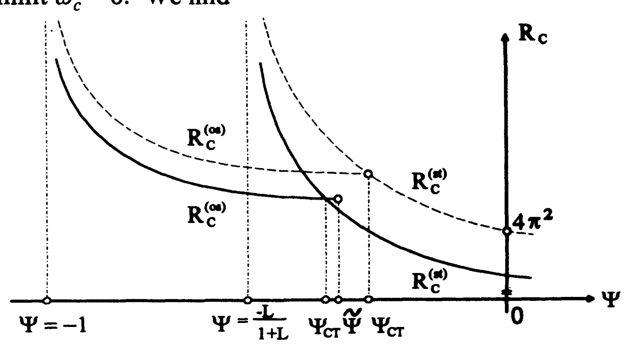

FIG. 1. Stability diagram for a binary mixture subject to Soret effects, critical Rayleigh numbers $R_c^{(\mathrm{st})}$ and $R_c^{(\mathrm{os})}$ for the onset of steady-state and oscillatory convection as functions of the separation ratio ψ, and for the non-Boussinesq numbers $\gamma=0$ (dashed lines) and $\gamma\neq 0$ (solid lines). ψ_{CT} denotes the codimension-two point.

$$\tilde{\psi}=\frac{-L^2}{1+L+L^2}\left[1+\gamma^2\frac{1+L+L^2}{1+L}\times\{f_1(1+\tfrac{7}{5}L)+f_2(1+\tfrac{19}{17}L)\}\right]. \tag{3.22}$$

Since $\tilde{\psi}>\psi_{\mathrm{CT}}$, where $\tilde{\psi}-\psi_{\mathrm{CT}}\propto\gamma^2L^2$, the oscillatory stability curve $R_c^{(\mathrm{os})}(\psi)$ does not end at the CTP as for the case $\gamma=0$, but exists in a small range for $\psi>\psi_{\mathrm{CT}}$. In this range we have $R_c^{(\mathrm{st})}<R_c^{(\mathrm{os})}$; see Fig. 1. Thus, at the CTP the onset of oscillatory convection is characterized by a nonzero critical frequency when a nonlinear density profile is present. Similar results are predicted by linear theory if the boundaries are assumed to be impermeable for solute flux; see Refs. [9,10,15,16].

Figure 2 shows qualitatively the critical wave numbers $k_c^{(\mathrm{st})}$ and $k_c^{(\mathrm{os})}$ according to Eqs. (3.16) and (3.19), respectively, as functions of ψ for the cases $\gamma=0$ (dashed lines) and $\gamma\neq 0$ (solid lines). When the nonlinear density profile is absent we find constant wave numbers and $k_c^{(\mathrm{st})}=k_c^{(\mathrm{os})}$ [29,31]. In contrast, if $\gamma\neq 0$ the critical wave numbers of the steady-state and oscillatory instability differ as they depend on the separation ratio ψ. Especially at the CTP we find $k_c^{(\mathrm{st})^2}-k_c^{(\mathrm{os})^2}\propto\gamma^2L$. A similar wave number splitting at the CTP was found by Refs. [9–12,15,16] by assuming impermeable boundary conditions. Furthermore, for $\gamma\neq 0$ the wave numbers are increased. This fact can be attributed to the destabilizing effect of the nonlinear density profile. Since the reduction of the Rayleigh number corresponds to a reduction of the active layer height the horizontal length scale is likewise reduced. As one

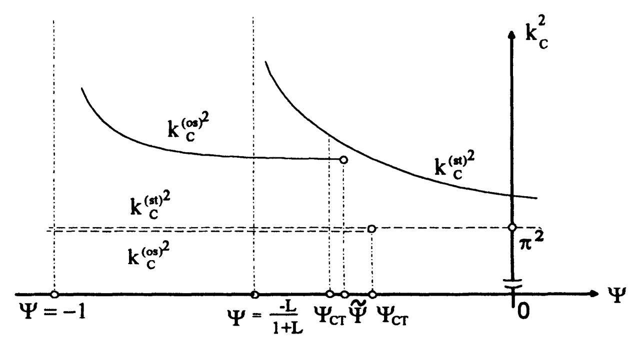

FIG. 2. Plot of the critical wave number $k_c^{(\mathrm{st})}$ and $k_c^{(\mathrm{os})}$ at onset of steady-state and oscillatory convection as functions of the separation ratio ψ and for the non-Boussinesq numbers $\gamma=0$ (dashed lines) and $\gamma\neq 0$ (solid lines). ψ_{CT} denotes the codimension-two point.

can see from Eqs. (3.16) and (3.19) there are singularities at values $\psi=-L(1+L)^{-1}$ and $\psi=-1$, respectively, in the representation of the wave numbers. Hence the convergence of the perturbation expansion is ensured only in the parameter ranges $\gamma^2<\{1+\psi(1+L^{-1})\}^2$ and $(1+\psi)^2$, respectively.

IV. FINITE-AMPLITUDE CONVECTION

A. Convective pattern

In the regime of finite-amplitude convection we focus on two- and three-dimensional steady-state (SS) and TW solutions. Standing waves are not considered, since these solutions are not expected to be a stable form of convection: In the case $\gamma=0$ Knobloch [31] has shown that two-dimensional standing waves are unstable with respect to larger-amplitude TWs. Following the idea of Knobloch, Karcher [51] has demonstrated that this result holds also for a small nonlinearity in the density profile as well as for the three-dimensional case.

The horizontal and temporal periodicity of the linear modes (see Sec. III) is achieved by introducing a pattern function of the form

$$\Phi(x,y,t)=A_H\sum_N \exp\{-i[k(\boldsymbol{\varphi}_N\cdot\mathbf{r})-\omega q t]\}\ , \qquad (4.1)$$

so that the complete solutions $\mathbf{x}_{1m}(x,y,z,t)$ in the order $\varepsilon^1\gamma^m$ can be written as a product, i.e., $\mathbf{x}_{1m}(x,y,z,t)=\mathbf{X}_{1m}(z)\Phi(x,y,t)$. In Eq. (4.1) $\mathbf{r}=(x,y)$ and $\boldsymbol{\varphi}_N$ is a horizontal unit wave vector. We restrict the analysis to so-called regular patterns [35,36]. These are characterized by a single horizontal amplitude A_H. The particular value of A_H can be determined from the normalization condition $\langle\Phi,\Phi\rangle=1$.

In the notation of Eq. (4.1) a two-dimensional roll pattern is given by $N=1$ and $A_H=\pm 2^{1/2}$. A three-dimensional hexagonal pattern is characterized by $N=3$, $A_H=\pm(\frac{2}{3})^{1/2}$, and $\varphi_{12}=\varphi_{13}=\varphi_{23}=-\frac{1}{2}$, where $\varphi_{NL}=\boldsymbol{\varphi}_N\cdot\boldsymbol{\varphi}_L$. In the time-periodic case ($\omega\neq 0$) the resulting pattern is a superposition of three TWs of the same phase velocity, while their axes are each shifted by an angle of 120°. This yields a nontraveling structure which may be interpreted as oscillating triangles (OTs); see Roberts, Swift, and Wagner [52].

The differential equations and the boundary conditions to the order $\varepsilon^n\gamma^m$, with $n>1$, can be written in the general form

$$\mathbf{M}\{\mathbf{x}_{nm}\}\equiv\begin{bmatrix}\nabla^2 & R_{00}(1+\psi)\nabla_H^2 & R_{00}\Psi\nabla_H^2\\ 1 & \nabla^2-\partial_t & 0\\ 0 & \nabla^2 & L\nabla^2-\partial_t\end{bmatrix}\begin{bmatrix}w_{nm}\\ T_{nm}\\ S_{nm}\end{bmatrix}=\mathbf{S}_{nm}\ ,$$
$$\mathbf{x}_{nm}=\mathbf{0}\quad\text{at } z=0,1\ . \qquad (4.2)$$

Here $\mathbf{S}_{nm}$ represents the corresponding right-hand side vector containing the relevant coefficients of the perturbation expansion (2.12a) and (2.12b), i.e., $R_{n-1,m}$ and $\omega_{n-1,m}$. Moreover, $\mathbf{S}_{nm}$ contains terms which are due to the nonlinearities in the basic equations (2.7). These terms can be expressed in form of products of n first order solutions. Due to the homogenous boundary conditions the calculation of $R_{n-1,m}$ and $\omega_{n-1,m}$ is straightforward; we apply the solvability condition

$$\langle\mathbf{S}_{nm},\mathbf{x}^*_{10,K}\rangle=0 \qquad (4.3)$$

within the respective order. Here $\mathbf{x}^*_{10,K}$ denotes an arbitrary solution of the adjoint eigenvalue problem characterized by the wave vector $\boldsymbol{\varphi}_K$. According to Eq. (4.3), the coefficients $R_{n-1,m}$ and $\omega_{n-1,m}$ are fixed by eliminating resonant terms generated by nonlinear interactions of first order solutions. Moreover, together with the coefficients $R_{n-1,m}$ we discuss the stability of the various patterns. This is done with the aid of standard stability theorems of bifurcation theory; see Refs. [39–41] for details. For a supercritical bifurcation to occur it is necessary and sufficient that the first nonvanishing coefficient $R_{n-1,m}$ is positive. In this case the bifurcating solution is stable with respect to infinitesimally small disturbances which are themselves of the same spatiotemporal structure. If the first nonvanishing coefficient is negative, the bifurcation is subcritical and the corresponding solution is unstable.

In this section we focus again on the presentation of the main results and refer to the Appendix for details of the calculations. In the following the cases $N=1$ and 3 will be treated separately.

B. Two-dimensional finite-amplitude convection

We consider first the equations of the order $\varepsilon^2\gamma^m$. Since by nonlinear interactions of two roll solutions ($n=2$), characterized by the same wave vector, resonant terms cannot be generated all coefficients of the perturbation expansion vanish, i.e.,

$$R_{1m}=0\ , \qquad (4.4a)$$

$$\omega_{1m}=0,\quad m=0,1,2,\ldots\ . \qquad (4.4b)$$

To the orders $\varepsilon^3\gamma^0$ and $\varepsilon^3\gamma^1$ the relevant equations are

$$\mathbf{M}\{\mathbf{x}_{30}\}=\begin{bmatrix}R_{20}\nabla_H^2\{(1+\psi)T_{10}-\psi S_{10}\}\\ (\mathbf{v}_{10}\cdot\nabla)T_{20}+i\omega_{20}qT_{10}\\ (\mathbf{v}_{10}\cdot\nabla)S_{20}+i\omega_{20}qS_{10}\end{bmatrix},$$
$$\mathbf{x}_{30}=\mathbf{0}\quad\text{at } z=0,1 \qquad (4.5)$$

and

$$\mathbf{M}\{\mathbf{x}_{31}\}=\begin{bmatrix}R_{21}\nabla_H^2\{(1+\psi)T_{10}-\psi S_{10}\}-2(z-\frac{1}{2})\nabla_H^2(R_{00}T_{30}+R_{20}T_{10})+2R_{00}\nabla_H^2(T_{10}T_{20})\\ (\mathbf{v}_{11}\cdot\nabla)T_{20}+i\omega_{21}qT_{10}+i\omega_{20}qT_{11}\\ (\mathbf{v}_{11}\cdot\nabla)S_{20}+i\omega_{21}qS_{10}+i\omega_{20}qS_{11}\end{bmatrix},$$
$$\mathbf{x}_{31}=0\quad\text{at } z=0,1\ , \qquad (4.6)$$

respectively. In writing Eqs. (4.5) and (4.6) we have already used results of the lower-order analysis to simplify the vectors $\mathbf{S}_{30}$ and $\mathbf{S}_{31}$ (see the Appendix). The evaluation of the solvability condition of the problems (4.5) and (4.6) yields the following coefficients for the SS solution:

$$R_{20}=R_{00}^{(\mathrm{st})}\frac{q}{4}\frac{1+\psi(1+L^{-1}+L^{-2}+L^{-3})}{1+\psi(1+L^{-1})}\ , \qquad (4.7a)$$

$$R_{21}=0\ . \qquad (4.7b)$$

For the TW solution we obtain

$$R_{20}=0\ , \qquad (4.8a)$$

$$R_{21}=0\ , \qquad (4.8b)$$

$$2\omega_{00}\omega_{20}=-\frac{q}{4}(1+\omega_{00}^2)\ , \qquad (4.9a)$$

$$\omega_{21}=0\ . \qquad (4.9b)$$

There is no dependence of the perturbation expansion (2.12) on the non-Boussinesq number γ to this order. The results are therefore similar to findings of Knobloch [31] for $\gamma=0$. The SS solution shows a so-called [31,46–48] tricritical point ($R_{20}=0$) at $\psi=\psi_{\mathrm{TC}}$, where $\psi_{\mathrm{TC}}=-(1+L^{-1}+L^{-2}+L^{-3})^{-1}$. For $\psi>\psi_{\mathrm{TC}}$ the bifurcation to steady-state convection is supercritical ($R_{20}>0$) and for $\psi<\psi_{\mathrm{TC}}$ it is subcritical ($R_{20}<0$). Since $\psi_{\mathrm{CT}}<\psi_{\mathrm{TC}}$ [cf. Eq. (3.21)], at the CTP the bifurcation is subcritical and the corresponding coefficient reads

$$R_{20}^{(\mathrm{CT})}=-R_{00}^{(\mathrm{st})}\frac{q}{4}L^{-1}\ . \qquad (4.10)$$

Hence, when the first instability of the basic state is oscillatory, the SS solution is unstable at finite amplitudes. Note that the bifurcation to TW convection is degenerate ($R_{20}=0$) and the frequency of the TW decreases with increasing amplitude ($\omega_{20}<0$). To describe the bifurcation of the solutions in more detail we perform a higher-order perturbation analysis which strictly holds near the CTP.

It is easy to show that to the order $\varepsilon^4\gamma^0$ no resonant terms are generated and the coefficients vanish. Thus we obtain

$$R_{30}=0\ , \qquad (4.11a)$$

$$\omega_{30}=0\ . \qquad (4.11b)$$

To the order $\varepsilon^5\gamma^0$ the relevant equations read as

$$\mathbf{M}\{\mathbf{x}_{50}\}=\begin{pmatrix} R_{40}\nabla_H^2\{(1+\psi)T_{10}-\psi S_{10}\}+R_{20}\nabla_H^2\{(1+\psi)T_{30}-\psi S_{30}\} \\ (\mathbf{v}_{10}\cdot\nabla)T_{40}+(\mathbf{v}_{30}\cdot\nabla)T_{20}+i\omega_{40}qT_{10}+i\omega_{20}qT_{30} \\ (\mathbf{v}_{10}\cdot\nabla)S_{40}+(\mathbf{v}_{30}\cdot\nabla)S_{20}+i\omega_{40}qS_{10}+i\omega_{20}qS_{30}\end{pmatrix}\ , \qquad (4.12)$$

$$\mathbf{x}_{50}=0 \quad \text{at } z=0,1\ .$$

The evaluation of the solvability condition of Eq. (4.12) turns out to be a cumbersome task. The full equations determining the coefficients R_{40} are given in the Appendix [see Eqs. (B10) and (B11)]. However, in the vicinity of the CTP and for small Lewis numbers L the expressions can be considerably simplified. By inserting $\psi=\psi_{\mathrm{CT}}-\Delta\psi$ into Eqs. (B10) and (B11) and calculating the expressions in the limit $\Delta\psi\ll 1$, the following results were derived: For the SS solution we find

$$R_{40}^{(\mathrm{CT})}=\tfrac{6}{5}\pi^6L^{-3}[1+O(L)+O(\Delta\psi)]\ , \qquad (4.13)$$

where we have used $k^2=\pi^2$. For the TW solution the corresponding coefficients are

$$R_{40}^{(\mathrm{CT})}=\tfrac{5}{24}\pi^6L^{-2}[1+O(L)+O(\Delta\psi)]\ , \qquad (4.14)$$

$$2\omega_{00}\omega_{40}^{(\mathrm{CT})}=-\tfrac{1}{16}\pi^4\frac{(1+\omega_{00}^2)^2}{\omega_{00}^2}[1+O(\Delta\psi)]\ . \qquad (4.15)$$

Note that for the SS solution we have $R_{40}^{(\mathrm{CT})}>0$ [Eq. (4.13)] but $R_{20}^{(\mathrm{CT})}<0$ [Eq. (4.10)]. Hence the subcritically bifurcating branch of the steady-state solution develops a limit point, characterized by $R=R^{(\mathrm{LP})}$ and $R^{(\mathrm{LP})}<R_c^{(\mathrm{st})}$, where it changes direction. Bifurcation theory assures [39–41] that at the limit point the SS solution is stabilized with respect to infinitesimally small disturbances. Moreover, at the CTP the bifurcation to TW convection is supercritical since $R_{40}^{(\mathrm{CT})}>0$; see Eq. (4.14). Thus TWs are stable near the onset. Furthermore, we find that the corresponding frequency of the wave is not affected to the order $\varepsilon^5\gamma^0$. According to Eqs. (4.9b) and (4.15) the relation $2\omega_{00}\omega_{40}^{(\mathrm{CT})}=-(\omega_{20}^{(\mathrm{CT})})^2$ holds, and both coefficients contributing to the expansion (2.12b) compensate.

We insert all the calculated coefficients into the perturbation expansion of the Rayleigh number [Eq. (2.12a)] and solve for the amplitude ε. The result is plotted in Fig. 3. The diagram shows the bifurcations to steady-state and traveling wave convection in the vicinity of the CTP, where we have $R_c^{(\mathrm{st})}-R_c^{(\mathrm{os})}\propto\Delta\psi$ and $\Delta\psi\ll 1$. Hatched curves indicate unstable branches of the solutions. Increasing the Rayleigh number quasisteadily (cf. the arrow symbols in Fig. 3) the basic state loses stability at $R=R_c^{(\mathrm{os})}$ with respect to TWs. At a supercritical value of the Rayleigh number, i.e., $R=R_c^{(\mathrm{TW})}$ and $R_c^{(\mathrm{TW})}-R_c^{(\mathrm{os})}\propto(\Delta\psi)^2$, the TW branch terminates at the lower unstable branch of the SS solution. At this particular Rayleigh number the wave motion disappears ($\omega^2\rightarrow 0$) and a transition to the upper branch of the SS solution occurs leading to stable steady-state convection at large amplitudes. On the other hand, starting from a supercritical point on the upper SS branch and decreasing R in small steps, at the limit point $R=R^{(\mathrm{LP})}$ a transi-

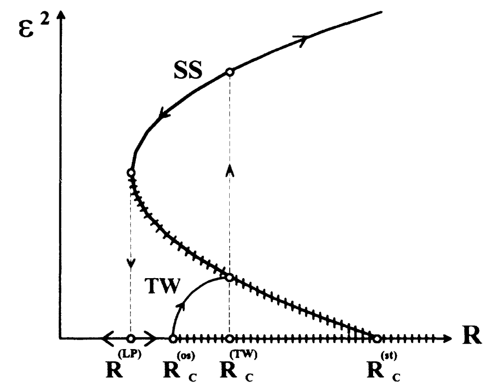

FIG. 3. Qualitative sketch of the bifurcations from the basic state to two-dimensional convection in the vicinity of the codimension-two point showing the amplitudes ε^2 of traveling waves (TW) and steady-state rolls (SS) as functions of the Rayleigh number R. Hatched curves indicate unstable branches of the solutions. Arrows indicate sequence of states which are taken when increasing or decreasing R quasisteadily.

tion to the basic state occurs and convection dies out. We find $R_c^{(\mathrm{st})}-R^{(\mathrm{LP})}\propto L$; thus the TW branch is completely overshadowed by the subcritical region of the SS solution provided $\Delta\psi<L$ holds. Similar bifurcation diagrams were derived by Deane, Knobloch, and Toomre [46,47] and Knobloch and Moore [48], who studied thermohaline convection using numerical methods.

As stated before, the bifurcation to two-dimensional convection is not affected by the non-Boussinesq number to the first order. The evaluation of the effects at higher order requires the calculation of the coefficients R_{22} and ω_{22}. However, this seems beyond the scope of an analytical procedure and numerical methods have to be employed. Thus the result of the present investigation does not allow to confirm or reject the conjecture of Zimmermann and Müller [32], namely, that the observed suppression of TWs in binary mixtures at low mean temperatures is due to a strong nonlinear relation between density and temperature.

C. Three-dimensional finite-amplitude convection

We outline next the bifurcations of steady-state and oscillatory solutions reflecting hexagonal symmetry. The evaluation of the solvability condition to the order $\varepsilon^2\gamma^0$ yields vanishing coefficients since the nonlinearity in the density profile is absent. We obtain

$$R_{10}=0\ , \tag{4.16a}$$

$$\omega_{10}=0\ . \tag{4.16b}$$

According to Eq. (4.1) the spatiotemporal dependence of the solutions $\mathbf{x}_{20}$ can be represented by a product of two sums with the wave vectors $\boldsymbol{\varphi}_N$ and $\boldsymbol{\varphi}_L$, respectively.

To the order $\varepsilon^2\gamma^1$ we have to consider

$$\mathbf{M}\{\mathbf{x}_{21}\}=\begin{Bmatrix} R_{11}\nabla_H^2\{(1+\psi)T_{10}-\psi S_{10}\}-R_{00}\nabla_H^2\{2(z-\tfrac{1}{2})T_{20}-T_{10}^2\} \\ (\mathbf{v}_{10}\cdot\nabla)T_{11}+(\mathbf{v}_{11}\cdot\nabla)T_{10}+i\omega_{11}qT_{10} \\ (\mathbf{v}_{10}\cdot\nabla)S_{11}+(\mathbf{v}_{11}\cdot\nabla)S_{10}+i\omega_{11}qS_{10}\end{Bmatrix}\ ,$$
$$\mathbf{x}_{21}=\mathbf{0}\quad\text{at } z=0,1\ . \tag{4.17}$$

The solvability condition of Eq. (4.17) indicates a nontrivial coefficient R_{11} for steady-state hexagons due to the symmetry breaking effect on non-Boussinesq properties; see Busse [35]. This is the case whenever the wave vectors form an equilateral triangle, i.e., $\boldsymbol{\varphi}_N\pm\boldsymbol{\varphi}_L\pm\boldsymbol{\varphi}_K=0$ holds. We find

$$R_{11}=-R_{00}^{(\mathrm{st})}\frac{40}{21\pi}A_H\frac{1+\psi(1+L^{-1}+\frac{4}{25}L^{-2})}{\{1+\psi(1+L^{-1})\}^2}\ . \tag{4.18}$$

Especially near the CTP, where $\Delta\Psi\ll 1$, the corresponding coefficient writes as

$$R_{11}^{(\mathrm{CT})}=-\tfrac{32\pi}{5}A_H(1+L+L^2)^2+O(\Delta\psi)\ . \tag{4.19}$$

According to expansion (2.12a) and Eq. (4.19) the bifurcation depends on both the sign of the amplitude A_H and the sign of the non-Boussinesq number γ. Here we only discuss the case $\gamma>0$, which is typical for binary liquid mixtures at low mean temperatures. For so-called l-hexagons [35], characterized by an upstream in the center, $A_H>0$ and thus $R_{11}^{(\mathrm{CT})}<0$. In this case the bifurcation is subcritical. On the other hand, for so-called g-hexagons [35], with downstream in the center, the bifurcation is supercritical since $A_H<0$ and $R_{11}^{(\mathrm{CT})}>0$. However, for the oscillatory pattern the distinguishing feature of up- and downstream in the center is lost. Thus the nonlinear density profile does not affect the bifurcation in this case. Consequently for the OT solution we find

$$R_{11}=0\ , \tag{4.20a}$$

$$\omega_{11}=0\ . \tag{4.20b}$$

Finally by applying the solvability condition to the equations of the order $\varepsilon^3\gamma^0$ [Eq. (4.5)], the following results are obtained. For both SS solutions, i.e., l-hexagons and g-hexagons, we find

$$R_{20}=\frac{\frac{1}{6}\pi^2R_{00}^{(\mathrm{st})}}{1+\psi(1+L^{-1})}\left[\tfrac{1079}{259}\{1+\psi(1+L^{-1}+L^{-2})\}+\tfrac{139}{35}\psi L^{-3}\left\{1+\frac{7\times 36}{37\times 139}L\frac{1+\psi(1+L^{-1}+L^{-2})}{1+\psi(1+L^{-1})}\right\}\right]. \quad (4.21)$$

In the vicinity of the CTP the result (4.21) simplifies to

$$R_{20}^{(\mathrm{CT})}=-\tfrac{2}{3}\tfrac{139}{35}\pi^4L^{-1}(1+L+L^2)+O(\Delta\psi)\,. \quad (4.22)$$

Therefore the bifurcation is subcritical at finite amplitudes. For the OT solution in the vicinity of the CTP we obtain

$$R_{20}^{(\mathrm{CT})}=\tfrac{2}{3}\tfrac{1061}{1225}\pi^4L^{-1}(1+L+L^2)+O(\Delta\psi)\,, \quad (4.23)$$

$$2\omega_{00}\omega_{20}^{(\mathrm{CT})}=-\tfrac{1}{6}\tfrac{11\,257}{2450}\pi^2L^{-1}+O(\Delta\psi)\,. \quad (4.24)$$

Note that the branch of the OT solution bifurcates supercritically ($R_{20}^{(\mathrm{CT})}>0$) and the frequency decreases with increasing amplitudes ($\omega_{20}<0$).

By combining the results in the same way as in Sec. IV B, the bifurcation diagram of the solutions in the vicinity of the CTP can be given. The results are shown in Fig. 4. Figure 4(a) illustrates the case $\gamma=0$ while Fig. 4(b) shows the bifurcations in the presence of a nonlinear density profile, i.e., for $\gamma>0$. For reasons of clarity both graphs are limited to the first quadrant of the R-ε plane. We conclude (see [39–41]) that the subcritical portion of the SS branches is unstable. In Fig. 4 the corresponding curves are given by hatched lines. To evaluate the stability of the OT solution we follow the ideas of Roberts, Swift, and Wagner [52]. They have shown that all other oscillatory solutions bifurcating on a hexagonal lattice are unstable if TWs are stable. In the present problem the instability of the OT solution is inferred from the fact that TWs develop the larger amplitude at the onset of oscillatory convection. Therefore in Fig. 4 the OT branch is marked by hatched lines.

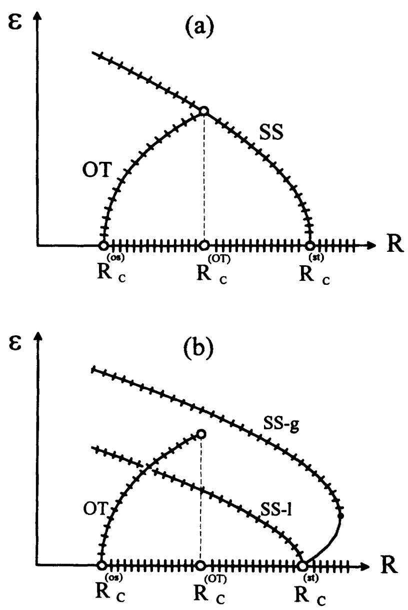

FIG. 4. Qualitative sketch of the bifurcations from the basic state to three-dimensional convection in the vicinity of the codimension-two point showing the amplitudes ε of the solutions as functions of the Rayleigh number R. Hatched curves indicate unstable branches of the solutions. (a) Oscillating triangles (OT) and steady-state hexagons (SS) for the non-Boussinesq number $\gamma=0$. (b) OT, steady-state l-hexagons (SS-l) and g-hexagons (SS-g) for a non-Boussinesq number $\gamma>0$.

For $\gamma=0$, it is easily shown that increasing the Rayleigh number towards the value $R=R_c^{(\mathrm{OT})}$, where $R_c^{(\mathrm{OT})}-R_c^{(\mathrm{os})}\propto\Delta\psi$, leads to a termination of the OT branch ($\omega^2\to 0$) on the unstable SS branch representing both l-hexagons and g-hexagons [see Fig. 4(a)]. In this case the bifurcation is similar to that described in Sec. IV B. However, in the presence of a nonlinear density profile ($\gamma>0$), according to Eq. (4.14) the single SS branch is unfolded into a branch of l-hexagons and a branch of g-hexagons; see Fig. 4(b). On the other hand, according to Eqs. (4.20a) and (4.20b) the bifurcation to OT convection is not changed for $\gamma\neq 0$. As a consequence, the branch of the OT solution terminates at $R=R_c^{(\mathrm{OT})}$ at an isolated point in the parameter plane if a nonlinear density profile is present.

In the experiments of Zimmermann and Müller [32] at low mean temperatures within the mixture a steady-state three-dimensional convection pattern was observed. They explained their observation by the effect of a strong nonlinear density profile of the basic state. This conjecture cannot be strictly confirmed by the present analysis. Since the perturbation expansion is truncated at third order in ε, at finite amplitudes only unstable hexagons are predicted. A higher-order analysis is necessary to demonstrate the existence of stable hexagons at larger amplitudes.

V. SUMMARY

In this paper we have studied Bénard convection of a binary liquid in a porous medium. The mixture displays negative Soret effects and shows a nonlinear dependence of density on temperature, i.e., non-Boussinesq properties. The main parameters controlling both the stability of the basic state and the bifurcation to finite-amplitude convection are the Rayleigh number R, the separation ratio ψ, and the non-Boussinesq number γ.

Linear theory is examined for the case of permeable boundary conditions. The stability of the basic state is characterized by a codimension-two point at $\psi=\psi_{\mathrm{CT}}$ and $\psi_{\mathrm{CT}}<0$. When $\psi<\psi_{\mathrm{CT}}$ the basic state is replaced by oscillatory convection as R exceeds a critical value at $R=R_c^{(\mathrm{os})}$. At an even higher, second critical value of the Rayleigh number, $R=R_c^{(\mathrm{st})}$ with $R_c^{(\mathrm{st})}>R_c^{(\mathrm{os})}$, steady-state convection is predicted. If a nonlinear density-temperature relation is present, i.e, $\gamma\neq 0$, the critical Rayleigh numbers and the value of ψ_{CT} are decreased. We find that the parameter range in which a bifurcation to oscillatory convection may occur is decreased. Moreover, we find the nonlinearity in the density profile to have similar effects as impermeable boundary conditions [9–12,15,16]; at the CTP a wave number splitting and a nonzero frequency of oscillations is predicted. When $\psi=0$ or $\gamma=0$ the present results agree with those of Busse [35,36] and Brand and Steinberg [29], respectively.

The bifurcations to finite-amplitude convection are examined in the vicinity of the CTP, i.e., $\psi_{CT}-\psi \ll 1$. For the case of two-dimensional convection the nonlinear density profile does not affect the bifurcation. Thus the present results correspond qualitatively to those presented by Knobloch [31], Deane, Knobloch, and Toomre [46,47], and Knobloch and Moore [48]. A stable branch of traveling wave solutions exists near the onset. At a slightly supercritical Rayleigh number, $R=R_c^{(TW)}$ with $R_c^{(TW)}>R_c^{(os)}$, this branch terminates at the subcritical unstable portion of the steady-state branch. There a jump transition to the upper stable part of the steady-state branch of large-amplitude convection is expected to occur.

The bifurcation to three-dimensional steady-state convection is affected by the nonlinear density-temperature relation of the mixture. If $\gamma=0$ there is only a single branch of hexagonal solutions. If a nonlinear density profile is present, i.e., $\gamma\neq 0$, this branch is unfolded into a branch of l-hexagons and a branch of g-hexagons. In single-component liquids ($\psi=0$) this unfolding phenomenon was already predicted by Busse [35]. The bifurcation to oscillatory hexagonal convection is not influenced by the parameter γ. Thus, if $\gamma\neq 0$ we find that the oscillatory branch does not terminate at the subcritical steady-state branch as for the case $\gamma=0$, but ends at an isolated point in the parameter plane. Using stability considerations of Roberts, Swift, and Wagner [52] we conclude that the oscillatory solution is unstable.

In experiments on Bénard convection in binary mixtures Zimmermann and Müller [32] observed that the traveling wave solution was suppressed when the mean temperature within the mixture was close to the solidification temperature of the liquid. Instead they observed a steady-state hexagonal-type pattern. They attributed these experimental findings to the presence of a significant nonlinearity in the density profile of the basic state. The present analysis cannot conclusively confirm their conjecture since the experimental range of amplitudes is not fully covered by our perturbation analysis. However, if the nonlinear density-temperature relation is taken into account both findings, the shift of the CTP and the unfolding of the steady-state hexagons support the conjecture of Zimmermann and Müller [32].

APPENDIX A: SOLUTIONS OF THE LINEAR STABILITY ANALYSIS

To the order $\varepsilon^1\gamma^0$ the steady-state ($\omega_{00}=0$) and oscillatory ($\omega_{00}\neq 0$) solutions are

$$\mathbf{X}_{10}=\begin{bmatrix} q(1+i\omega_{00}) \\ 1 \\ -(L+i\omega_{00})^{-1}\end{bmatrix}\sin\pi z\ . \tag{A1}$$

The solutions $\mathbf{X}_{10}^*$ of the adjoint steady-state $[\omega_{00}=0,\ r=(1+\psi(1+L^{-1})^{-1})]$ and oscillatory $[\omega_{00}\neq 0,\ r=(1+L)(1+\psi)^{-1}]$ problem are

$$\mathbf{X}_{10}^*=\begin{bmatrix} q^{-1} \\ 1 \\ -r\psi(L-i\omega_{00})^{-1}\end{bmatrix}\sin\pi z\ . \tag{A2}$$

To the order $\varepsilon^1\gamma^1$ the steady-state and oscillatory solutions write as

$$\mathbf{X}_{11}=\begin{bmatrix} q(1+i\omega_{00})+3\pi^2 \\ 1 \\ -\dfrac{q+3\pi^2}{q(L+i\omega_{00})+3\pi^2}\end{bmatrix}\frac{32q^2r}{3^3\pi^4(2q+3\pi^2)}\frac{L(q+3\pi^2)+i\omega_{00}q}{L(q+3\pi^2)+i\omega_{00}q(1+L)}\sin 2\pi z$$
$$+\begin{bmatrix} q(1+i\omega_{00})+15\pi^2 \\ 1 \\ -\dfrac{q+15\pi^2}{q(L+i\omega_{00})+15\pi^2}\end{bmatrix}\frac{64q^2r}{15^3\pi^4(2q+15\pi^2)}\frac{L(q+15\pi^2)+i\omega_{00}q}{L(q+15\pi^2)+i\omega_{00}q(1+L)}\sin 4\pi z+\text{h.o.t}\ , \tag{A3}$$

where we have expanded terms proportional to $(z-\frac{1}{2})\sin\pi z$ into a Fourier's series and h.o.t. represents higher-order terms of the expansion.

APPENDIX B: SOLUTIONS OF THE FINITE-AMPLITUDE ANALYSIS

To the order $\varepsilon^2\gamma^0$ the solution can be written in the form

$$\mathbf{x}_{20}=\sin 2\pi z\left[\begin{bmatrix}A_{11}\\A_{12}\\A_{13}\end{bmatrix}+\sum_{N\neq L}\exp[-ik(\boldsymbol{\varphi}_N-\boldsymbol{\varphi}_L)\cdot\mathbf{r}]\begin{bmatrix}A_{21}\\A_{22}\\A_{23}\end{bmatrix}+\sum_{N\neq L}\exp[-i\{k(\boldsymbol{\varphi}_N+\boldsymbol{\varphi}_L)\cdot\mathbf{r}-2\omega qt\}]\begin{bmatrix}A_{31}\\A_{32}\\A_{33}\end{bmatrix}\right]\ , \tag{B1}$$

where the components A_{ij} are defined as follows:

$$A_{11}=0\ , \tag{B2a}$$

$$A_{12}=\frac{c_1}{\Delta_1}\ , \tag{B2b}$$

$$A_{13}=-L^{-1}\left[A_{12}+\frac{c_1}{\Delta_1}(L+i\omega_{00})^{-1}\right]\ , \tag{B2c}$$

$$A_{21}=-\Delta_2 A_{22}+c_2\ , \tag{B2d}$$

$$A_{22}=c_2\frac{\Delta_2-\Delta_{H2}R_{00}\psi(L\Delta_2(L+i\omega_{00}))^{-1}}{\Delta_2^2+\Delta_{H2}R_{00}\{1+\psi(1+L^{-1})\}}\ , \tag{B2e}$$

$$A_{23}=-L^{-1}\left[A_{22}+\frac{c_2}{\Delta_2}(L+i\omega_{00})^{-1}\right]\ , \tag{B2f}$$

$$A_{31}=-[\Delta_3-2i\omega_{00}q]A_{32}+c_3\ , \tag{B2g}$$

$$A_{32}=c_3\frac{\Delta_3-\Delta_{H3}(L\Delta_3-2i\omega_{00}q)^{-1}R_{00}\psi(L+i\omega_{00})^{-1}}{\Delta_3(\Delta_3-2i\omega_{00}q)+\Delta_{H3}R_{00}\{1+\psi(1+\Delta_3[L\Delta_3-2i\omega_{00}q]^{-1})\}}\ , \tag{B2h}$$

$$A_{33}=-(L\Delta_3-2i\omega_{00}q)^{-1}\left[A_{32}+\frac{c_3}{\Delta_3}(L+i\omega_{00})^{-1}\right]\ , \tag{B2i}$$

and

$$c_1=\tfrac{1}{2}N\pi q A_H^2(1-i\omega_{00})\ , \tag{B3a}$$

$$c_2=\tfrac{1}{4}\pi q A_H^2(1-i\omega_{00})(1+\varphi_{NL})\ , \tag{B3b}$$

$$c_3=\tfrac{1}{4}\pi q A_H^2(1+i\omega_{00})(1-\varphi_{NL})\ . \tag{B3c}$$

Here the Δ_i and Δ_{Hi} correspond to the Laplacian and horizontal Laplacian, respectively, of the different modes of the solution $\mathbf{x}_{20}$, i.e.,

$$\Delta_1=-4\pi^2\ , \tag{B4a}$$

$$\Delta_2=-[4\pi^2+2k^2(1-\varphi_{NL})]\ , \tag{B4b}$$

$$\Delta_3=-[4\pi^2+2k^2(1+\varphi_{NL})]\ , \tag{B4c}$$

$$\Delta_{H2}=-2k^2(1-\varphi_{NL})\ , \tag{B4d}$$

$$\Delta_{H3}=-2k^2(1+\varphi_{NL})\ . \tag{B4e}$$

In this general formulation rolls are given by $N=1$, $N=L$, and $A_H^2=2$ and hexagons by $N=3$, $N\neq L$, and $A_H^2=\frac{2}{3}$. To obtain the steady-state and oscillatory solutions we use $\omega_{00}=0$, $R_{00}=R_{00}^{(\mathrm{st})}$ and $\omega_{00}\neq 0$, $R_{00}=R_{00}^{(\mathrm{os})}$, respectively.

To the order $\varepsilon^3\gamma^0$ the roll solution reads as

$$\mathbf{x}_{30}=A_H^3\exp[-i\{k(\boldsymbol{\varphi}_1\cdot\mathbf{r})-\omega q t\}]\left[\begin{pmatrix}B_{11}\\0\\B_{12}\end{pmatrix}\sin\pi z+\begin{pmatrix}B_{31}\\B_{32}\\B_{33}\end{pmatrix}\sin 3\pi z\right]\ . \tag{B5}$$

For the steady-state solution the components B_{ij} are

$$B_{11}=\frac{q^2}{8}\ , \tag{B6a}$$

$$B_{13}=\frac{q}{8}L^{-3}(1+L)\ , \tag{B6b}$$

$$B_{31}=(q+8\pi^2)B_{32}-\frac{q^2}{8}\ , \tag{B6c}$$

$$B_{32}=\frac{q^2}{128\pi^2}\frac{q+8\pi^2}{q+4\pi^2}\left[1+\frac{q^2}{(q+8\pi^2)^2}\frac{L^{-3}\psi(1+L)}{1+\psi(1+L^{-1})}\right]\ , \tag{B6d}$$

$$B_{33}=-L^{-1}B_{32}-\frac{q^2}{8(q+8\pi^2)}L^{-3}(1+L)\ . \tag{B6e}$$

For TWs we find the following components:

$$B_{11}=\frac{q^2}{8}(1+i\omega_{00})+i\omega_{20}q\ , \tag{B7a}$$

$$B_{13}=\frac{q}{8}(L^2+\omega_{00}^2)^{-1}\frac{1+i\omega_{00}}{L+i\omega_{00}}(1+L)+i\omega_{20}(L+i\omega_{00})^{-2}\ , \tag{B7b}$$

$$B_{31}=-\frac{q^2}{8}(1+i\omega_{00})+\{q(1+i\omega_{00})+8\pi^2\}B_{32}\ , \tag{B7c}$$

$$B_{32}=\frac{q^2}{128\pi^2}\frac{q+8\pi^2}{q+4\pi^2}(1+i\omega_{00})\frac{\{L^2(q+8\pi^2)^2+\omega_{00}^2q^2-q^2(1+L)(L-qi\omega_{00}[q+8\pi^2]^{-1})\}}{L^2(q+8\pi^2)^2+\omega_{00}^2q^2(1+L)+i\omega_{00}q(q+8\pi^2)L^2}\ , \tag{B7d}$$

$$B_{33}=-\frac{q^2}{4}\frac{(1+i\omega_{00})(1+L)(L^2+\omega_{00}^2)^{-1}}{\{q(L+i\omega_{00})+8\pi^2L\}}-\frac{q+8\pi^2}{q(L+i\omega_{00})+8\pi^2L}B_{32}\ . \tag{B7e}$$

To the order $\varepsilon^4\gamma^0$ the steady-state solution can be written as

$$\mathbf{x}_{40}=\frac{1}{4\pi}A_H^4\begin{bmatrix}0\\(q+4\pi^2)B_{32}-\frac{1}{8}q^2\\-L^{-2}(1+L)\left[(q+4\pi^2)B_{32}-\frac{q^2}{8}\left[1-L^{-2}\frac{q+4\pi^2}{q+8\pi^2}\right]\right]\end{bmatrix}\sin 2\pi z+\text{n.r.t.}\ , \tag{B8}$$

and for TWs we find

$$\mathbf{x}_{40}=\frac{1}{2\pi}A_H^4\begin{bmatrix}0\\q(1-i\omega_{00})B_{32}-(\overline{B_{11}}-\overline{B_{31}})\\L^{-1}\left[q(1-i\omega_{00})(B_{32}+B_{13}-B_{33})+(\overline{B_{11}}-\overline{B_{31}})\left[1+\frac{L-i\omega_{00}}{L^2+\omega_{00}^2}\right]\right]\end{bmatrix}\sin 2\pi z+\text{n.r.t.} \tag{B9}$$

Here the components according to Eqs. (B6a)–(B6e) and (B7a)–(B7e), respectively, are used and the bar denotes the complex conjugate. n.r.t. denotes nonresonant terms.

To the order $\varepsilon^5\gamma^0$ the full dependence of the coefficients R_{40} on the parameters are as follows: For the SS solution we find

$$R_{40}=\frac{q^2}{16}A_H^4R_{00}^{(\text{st})}(1+L)\left[\frac{1}{4}\psi L^{-3}\frac{1+\psi(1+L^{-1}+L^{-2}+L^{-3})}{\{1+\psi(1+L^{-1})\}^2}\left\{1-\frac{q^2(3q+16\pi^2)}{16\pi^2(q+4\pi^2)(q+8\pi^2)}\right\}\right.$$
$$\left.-\frac{1}{2}\frac{q+4\pi^2}{q+8\pi^2}\frac{\psi L^{-5}}{1+\psi(1+L^{-1})}+\left\{1+\frac{\psi L^{-3}}{1+\psi(1+L^{-1})}\right\}\left\{1-\frac{(3q+16\pi^2)(q+8\pi^2)}{64\pi^4(q+4\pi^2)}\right\}\right]\ . \tag{B10}$$

For the TW using $k^2=\pi^2$ we find

$$R_{40}=\frac{5}{16}\pi^4A_H^4R_{00}^{(\text{os})}\frac{(1+\omega_{00}^2)(1+L)}{25L^2+\omega_{00}^2}\left[1+\frac{25L^2+\omega_{00}^2}{24}\frac{25(L^2+L^2\omega_{00}^2+\omega_{00}^4)+\omega_{00}^2(1-24L)^2}{\{25L^2+\omega_{00}^2(1+L)\}^2+25L^4\omega_{00}^2}\right]\ . \tag{B11}$$

Note that TWs bifurcate always supercritically since $R_{40}>0$.

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