Linear instability and the codimension-2 region in binary fluid convection between rigid impermeable boundaries

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The parameters of the linear instability to oscillatory convection (critical Rayleigh number, onset frequency, and others) are calculated for the experimentally common situation of rigid, impermeable boundaries, both near and away from the degenerate (codimension-2) bifurcation with stationary convection. This gives all linear coefficients of the standard and degenerate-amplitude equations. The small-Lewis-number limit is explicitly calculated. Wave-number and frequency jumps are confirmed in the vicinity of the codimension-2 point.

I. INTRODUCTION

Rayleigh-Benard convection in binary fluid mixtures has a number of new features compared with convection in pure fluids that have stimulated a large number of experiments recently.[1] For certain values of the fluid parameters the onset of convection, as the temperature difference between the horizontal plates containing the fluid is increased above a threshold, leads to an oscillatory state that may be manifested as traveling or standing waves. There is considerable interest in the study of these waves in the nonlinear state, in pattern formation in a dynamic state, and in the breakdown of simple wave states to more complicated, chaotic states. The fluid parameters may be tuned so that the frequency at onset goes to zero.[2] The behavior near this degenerate bifurcation point between stationary and oscillatory convection (convectionally called the codimension-2 point) is also of great interest.[3] Finally, novel states in the stationary convection region have been observed.[4,5]

The advances of precise experimental measurements and visualization of the flow field have stimulated much theoretical work. Surprisingly, until very recently the parameters describing the linear instability (e.g., critical Rayleigh number, onset frequency, and group velocity of the waves) were not precisely predicted from the fluid equations. Instead, various trial-function methods have been used.[6,7] Since these methods are not variational (i.e., do not give bounds) the calculations are not systematic, and one cannot have great confidence in their predictions. In addition, the careful measurements of the onset parameters experimentally[8] warrant a more accurate theoretical treatment.

A direct treatment of the linear instability is straightforward with modern numerical packages and computing power. We have implemented a numerical scheme to calculate all the parameters needed to construct the linearized complex-amplitude equation of oscillatory convection away from the codimension-2 point, and of stationary convection where this occurs. The linearized complex-amplitude equation for waves moving to positive x takes the form

$$\tau_0[\partial_t A + s\partial_x A] = \epsilon(1+ic_0)A + \xi_0^2(1+ic_1)\partial_x^2 A \ , \qquad (1.1)$$

with $A(x,t)$ the complex-amplitude modulating the basic wave $\exp[i(k_o x - \Omega_c t)]$, and $\epsilon = (R - R_o)/R_o$. The parameters τ_0, s, c_0, c_1, ξ_0^2, Ω_c, k_o, and R_o are to be calculated, as functions of Lewis number L, Prandtl number σ, and separation ratio ψ. Here Ω_c is the frequency of the wave at onset at the critical wave number k_o minimizing the onset Rayleigh number at R_o; τ_0 sets the basic time scale of the modulations and ξ_0 is the correlation length. Then $\tau_0^{-1}c_0$ gives the dependence of the frequency of small-amplitude waves on Rayleigh number (ϵ) and $\tau_0^{-1}\xi_0^2 c_1$ gives the dispersion. The group velocity of the waves is s. The calculation must be done as a function of many fluid parameters, and we cannot quote a complete set of results in any compact form. We expect to present detailed comparison with recent experiments for specific values of the parameters elsewhere. Recently, Zielinska and Brand[9] presented results for the onset Rayleigh number and frequency for oscillatory convection. We improve on the accuracy of their results, and calculate the additional linear parameters. In addition, we comment in some detail on an important conceptual question: namely, whether there are jumps in the wave number of the onset solution in the codimension-2 region. We disagree with the conclusion of Brand and Zielinska, and do indeed find jumps in agreement with a mode-truncation calculation of Linz and Lücke.[10] A codimension-2 point is approached smoothly only if the wave number of the rolls is fixed. If the wave number is allowed to adjust to the value giving the lowest threshold, then the wave number jumps, and the frequency jumps discontinuously to zero at some values of ψ. These latter results have been reported in a short communication.[11] Similar calculations have also been done simultaneously by Knobloch and Moore.[12]

Considerable numerical difficulty is introduced by the smallness of another fluid parameter, the Lewis number L (the ratio of the diffusivity of the impurity to the diffusivity of heat). In all experiments to date L has been very small (10^{-2}–10^{-3}). This introduces a rapid variation in the vertical spatial dependence of the solutions over a distance of order $(L/\Omega)^{1/2}$ with Ω the onset frequency. We consider the $L=0$ problem in the oscillatory regime. This provides a test of the small-L behavior, and also gives an easy approach to find quite accurate approximate results for the small-L problem.

In the codimension-2 region the simple-amplitude equation breaks down. Brand *et al.*[2] derived a new amplitude equation, now second-order in time, for this region for the physically unnatural free-slip, pervious boundary conditions. We perform the same calculation for the *linear* terms for the experimentally relevant boundary conditions. The degenerate-amplitude equation for the amplitude $A_k(t)$ at a *fixed* wave number k may be written

$$\partial_t^2 A_k + \alpha \partial_t A_k + \beta A_k = 0 \ , \tag{1.2}$$

where α and β are real numbers depending on R, k, L, σ, and ψ. The oscillatory instability occurs at $\alpha=0$, the stationary instability at $\beta=0$, so that the codimension-2 point $\psi^*(k,L,\sigma), R^*(k,L,\sigma)$ is the point $\alpha=\beta=0$. The calculation of α and β provide an additional check on the wave-number jump described above. Finally, we consider a small-L limit of the codimension-2 region. We explicitly factor out the leading-order L dependence from the problem, leaving a numerical calculation with no small parameters. We calculate the value of $\gamma=-\psi^*/L^2$ for $L\to 0$ as a function of k and σ. In addition, we write

$$\begin{aligned} \alpha &= \alpha_\epsilon \epsilon + \alpha_\psi \delta\psi \ , \\ \beta &= \beta_\epsilon \epsilon + \beta_\psi \delta\psi \ , \end{aligned} \tag{1.3}$$

with $\delta\psi=\psi-\psi^*$ and $\epsilon=(R-R^*)/R_p$ for each k [where $R_p \simeq 1707$ is the critical Rayleigh number for pure ($\psi=0$) stationary convection] and quote explicit values for the $L\to 0$ limit of $\alpha_\epsilon, \alpha_\psi, \beta_\epsilon, \beta_\psi$. These parameters define the behavior near the codimension-2 point for small L. In particular, we calculate an explicit expression for the wave-number jump, which is proportional to L, and the frequency jump proportional to $L^{3/2}$.

The plan of the paper is as follows. Section II sets out the equations and defines the parameters. Section III describes the derivation of the complex-amplitude equation (1.1). The results are described in Sec. IV. In Sec. V we present the derivation of the degenerate-amplitude equation (1.2) in the codimension-2 region, and describe the numerical implementation. We do not quote explicit results of these calculations, but have used them to check the results of Sec. IV and the small-L limit. The extraction of the small-L limit of the degenerate-amplitude equation is described in Sec. VI and explicit results are discussed in Sec. VII. Readers interested only in the result may confine their reading to Secs. II, IV, VII, and the conclusions in Sec. VIII.

II. EQUATIONS OF BINARY FLUID CONVECTION

We use the Boussinesq equations[6,7]

$$\partial_t \mathbf{u} + (\mathbf{u}\cdot\nabla)\mathbf{u} = -\nabla P + \sigma[\Theta(1+\psi)-\eta] + \sigma\nabla^2\mathbf{u} \ , \tag{2.1}$$

$$\partial_t \Theta + (\mathbf{u}\cdot\nabla)\Theta = Rw + \nabla^2\Theta \ , \tag{2.2}$$

$$\partial_t \eta + (\mathbf{u}\cdot\nabla)\eta = L\nabla^2\eta + \psi\nabla^2\Theta \ , \tag{2.3}$$

together with the incompressibility equation

$$\nabla\cdot\mathbf{u} = 0 \ . \tag{2.4}$$

The fields of the equation are the velocity $\mathbf{u}=(u,w)$; Θ, the deviation of the temperature from the linear conducting profile; and a field η defining c, the deviation of the concentration of the second component from the linear conducting profile driven by the Soret effect, through

$$\eta = c + \psi\Theta \ . \tag{2.5}$$

The boundary conditions at the upper and lower plates (rigid, perfect thermal conductors, impervious) are

$$\mathbf{u} = \Theta = \partial_z \eta = 0, \ z = \pm\tfrac{1}{2} \tag{2.6}$$

where the latter condition corresponds to no flux through the boundaries, including the Soret driven flux. As is conventional, we have neglected the very small Dufour effect in Eq. (2.2).

The parameters of the fluid are the Prandtl number σ (ratio of thermal to viscous diffusivities), Lewis number L (ratio of impurity to thermal diffusivities), separation ratio ψ (giving the ratio of the importance of thermal and concentration buoyancy effects), and Rayleigh number R giving in dimensionless form the driving temperature gradient. The equations have been made dimensionless in the usual way.[6,7] In particular, time is measured in units of the vertical thermal diffusion time and space is measured in terms of the vertical separation of the plates.

III. DERIVATION OF THE AMPLITUDE EQUATION

It is convenient to eliminate the pressure field from Eq. (2.1), and to write the horizontal velocity in terms of the vertical velocity using the continuity equation (2.4). Thus we write the linear solution as the vector

$$\mathbf{V} = [w(z), \Theta(z), \eta(z)]e^{ikx} \ , \tag{3.1}$$

with k the horizontal wave number. The equations become

$$\hat{O}\mathbf{V} = \hat{M}\partial_t \mathbf{V} \ , \tag{3.2}$$

with

$$\hat{O} = \begin{bmatrix} \nabla^4 & -(1+\psi)k^2 & k^2 \\ R & \nabla^2 & 0 \\ 0 & \psi\nabla^2 & L\nabla^2 \end{bmatrix} \tag{3.3}$$

and $\hat{M}$ the diagonal matrix with elements $(\sigma^{-1}\nabla^2,1,1)$, where $\nabla^2=\partial_z^2-k^2$. Although w, Θ, η, $\hat{O}$, and $\hat{M}$ depend on the wave number k, we will not explicitly show this dependence since we solve for each k independently. Since $\hat{O}$ is not self-adjoint, we will also need the adjoint equation

$$\hat{O}^\dagger \mathbf{V}^\dagger = \hat{M}\partial_t \mathbf{V}^\dagger \ , \tag{3.4}$$

with $\hat{O}^\dagger$ defined by requiring

$$(\mathbf{V}^\dagger, \hat{O}\mathbf{V}') = (\hat{O}^\dagger\mathbf{V}^\dagger, \mathbf{V}') \tag{3.5}$$

for any $\mathbf{V},\mathbf{V}'$. Here

$$(\mathbf{V}^\dagger, \mathbf{V}') = \langle w^{\dagger *}(z)w'(z) + \Theta^{\dagger *}(z)\Theta'(z) + \eta^{\dagger *}(z)\eta'(z)\rangle \ , \tag{3.6}$$

with $\langle\ \rangle$ denoting the average over the z coordinate. Then defining eigenvectors and eigenvalues for each k,

$$\hat{O}\mathbf{e}_i=\lambda_i\hat{M}\mathbf{e}_i\ ,\qquad \hat{O}^{\dagger}\mathbf{e}_i^{\dagger}=\lambda_i^{\dagger}\hat{M}\mathbf{e}_i^{\dagger}\ , \tag{3.7}$$

we have

$$(\mathbf{e}_j^{\dagger},\hat{M}\mathbf{e}_i)=0 \quad \text{for } \lambda_i\neq\lambda_j\ [=(\lambda_j^{\dagger})^*]\ . \tag{3.8}$$

The explicit form of $\hat{O}^{\dagger}$ is

$$\hat{O}^{\dagger}=\begin{bmatrix} \nabla^4 & R & 0 \\ -(1+\psi)k^2 & \nabla^2 & \psi\nabla^2 \\ k^2 & 0 & L\nabla^2 \end{bmatrix}\ , \tag{3.9}$$

with boundary conditions

$$w^{\dagger}=\partial_z w^{\dagger}=\partial_z\eta^{\dagger}=\Theta^{\dagger}+\psi\eta^{\dagger}=0 \quad \text{at } z=\pm\tfrac{1}{2}\ . \tag{3.10}$$

The critical Rayleigh number $R_o(k)$ and onset frequency $\Omega_c(k)$ for each k are given by the requirement that the eigenvalue with the greatest real part be pure imaginary (no growth or decay), i.e.,

$$\hat{O}(R_o)\mathbf{e}_0=-i\Omega_c\hat{M}\mathbf{e}_0\ . \tag{3.11}$$

Equation (3.11) clearly reduces to a single equation [e.g., for $w(z)$] that is eighth order in ∂_z (actually involving only powers of ∂_z^2) with constant coefficients. The solution for $w(z)$, which is even because of the boundary conditions, is given in terms of four unknown complex constants c_i and vertical complex wave numbers q_i,

$$w_0(z)=\sum_{i=1}^{4} c_i\cos(q_i z)\ , \tag{3.12}$$

with q_i given by the roots of the fourth-order polynomial in q^2, and expressions for $\Theta_0(z),\eta_0(z)$ given in terms of the same constants by Eqs. (2.2) and (2.3). The four boundary conditions at $z=+\frac{1}{2}$ [Eq. (2.6)] give four complex linear homogeneous equations for the unknowns. The Rayleigh number $R_o(k)$ and frequency $\Omega_c(k)$ are tuned to make the real and imaginary parts of the characteristic determinant zero.

This procedure was implemented in a straightforward way using the nonlinear root-finding routine from the Port library[13] in double precision on a 32-bit machine.

The amplitude equation is given by the expansion

$$\mathbf{V}(\mathbf{k})=A_{\mathbf{k}}(t)\mathbf{e}_0(\mathbf{k})\exp[i(kx-\Omega_c t)]+\delta\mathbf{V}\ , \tag{3.13}$$

with $\delta\mathbf{V}$ having no projection on the zero growth rate eigenvector, i.e., $(\mathbf{e}_0^{\dagger},\delta V)=0$. In Eq. (3.13) we have factored out the oscillatory time dependence at threshold for the critical wave number k_o minimizing the onset Rayleigh number $R_o(k)$ at R_o with frequency Ω_c. If $A(x,t)\exp(ik_o x)$ is the Fourier transform of A_k, then $A(x,t)$ satisfies in the linear regime the amplitude equation

$$\tau_0^{-1}[\partial_t+s\partial_x]A=(1+ic_0)\epsilon A+(1+ic_1)\xi_0^2\partial_x^2 A\ , \tag{3.14}$$

with τ_0,s,ξ_0,c_0,c_1 real numbers to be calculated and $\epsilon=(R-R_o)/R_o$.

Although the characteristic time τ_0 could be calculated by numerically differentiating the growth rate as a function of $(R-R_o)$, a more accurate approach is to use an integral expression involving $\mathbf{e}_0$ and $\mathbf{e}_0^{\dagger}$ at k_o. Substituting Eq. (3.13) into (3.2) and taking the inner product with $\mathbf{e}_0^{\dagger}$ gives

$$\tau_0(1+ic_0)=\frac{R_o\langle\Theta_0^{\dagger *}(z)w_0(z)\rangle}{\langle\sigma^{-1}w_0^{\dagger *}(z)\nabla^2 w_0(z)+\Theta_0^{\dagger *}(z)\Theta_0(z)+\eta_0^{\dagger *}(z)\eta_0(z)\rangle}\ . \tag{3.15}$$

The other parameters are calculated by numerically differentiating $\Omega_c(k)$ and $R_o(k)$ about k_o:

$$s=\partial\Omega_c/\partial k\ ,\qquad \xi_0^2=\frac{1}{2}\frac{\partial^2}{\partial k^2}\left[\frac{R_o(k)}{R_o}\right]\ ,\qquad c_1=\tfrac{1}{2}(\tau_0\xi_0^2)^{-1}\frac{\partial^2\Omega_c(k)}{\partial k^2}\ . \tag{3.16}$$

The numerical procedure is made more difficult by small parameters characteristic of typical physical systems. In particular, the Lewis number L is typically 10^{-3} to 10^{-2}. It is clear from Eq. (2.3) that for the solution at the onset of oscillatory convection there is a rapid variation in the z direction over a boundary layer of order $(L/\Omega)^{1/2}$ in size, and Ω is $O(1)$ away from the codimension-2 point. This corresponds to one of the roots q_i in Eq. (3.12) becoming large $|q|\sim(\Omega/L)^{1/2}$, perhaps leading to ill-conditioned linear equations to be solved. Simple reparametrization of the equations eliminates the exponential dependence of the entries in the determinants on this large number. Fortunately, we can check the behavior for small L by studying the approach to the results of an $L=0$ calculation. Note that for $L\to 0$ the boundary layer becomes of vanishing thickness. Thus we may replace Eq. (2.3) by

$$\eta=i\psi\nabla^2\Theta/\Omega\ , \tag{3.17}$$

eliminating the term in $\nabla^2\eta$, and we may ignore the boundary condition on η. There are now so small numbers in the system of equations, and the numerical procedure can be carried through as before with considerably greater ease. For oscillatory convection away from the codimension-2 point this $L=0$ limit is quite smooth: a finite onset frequency is obtained, for example. (For stationary convection this is not the case: there, it is easy to see that the solution depends on the ratio ψ/L for small L.) We have checked the smooth approach of the finite-L calculations for small L to the $L=0$ results down to $L=5\times10^{-4}$, considerably smaller than typical experimental values.

IV. RESULTS FOR THE NONDEGENERATE-AMPLITUDE EQUATION

A. General results

We calculate eight parameters (τ_0^{-1}, s, ξ_0, c_0, c_1, R_o, k_o, and Ω_c) as a function of the three fluid parameters L, σ, and ψ. There are clearly too many data to completely display, and we can only show examples. A comparison of results for specific values of the fluid parameters with experiment will be presented elsewhere. All experiments to date have been performed on systems with small Lewis number $L \sim 10^{-2}$–10^{-3}. The $L=0$ limit therefore provides a useful reference point. In addition, we show $L=0.05$ results to display the dependence on small L. We have chosen $\sigma=10$, a value that is found in alcohol-water mixtures. The results are displayed in Fig. 1. We have multiplied the frequency quantities by $\tau_p=(\sigma+0.5117)/19.65\sigma$, the quantity corresponding to τ_0 for pure stationary convection, to remove some of the Prandtl-number dependence. The differences between the $L=0$ and $L=0.05$ results are typically very small for most values of ψ and are linear in L. For $|\psi| \lesssim L$ the deviations become somewhat larger; for example, see Fig. 2. Note that the codimension-2 region is $-\psi \sim L^2$, and the deviations from $L=0$ again become linear in L in this region (see below). In Fig. 3 results for $L=0$ and various values of σ are shown.

B. Wave-number jumps

There has been some controversy in the literature[9,10] over the question of jumps in the properties, in particular the critical wave number, approaching the codimension-2 point. Linz and Lücke,[10] in an approximate mode-truncation calculation, found jumps in the wave number. Zielinska and Brand[9] disputed these results based on full calculations similar to ours. We have addressed this question in a short communication.[11] We indeed find jumps that, however, go to zero for $L \to 0$. This makes it rather a delicate question to resolve by direction calculation for small L, requiring a fine resolution in ψ (cf. Fig. 2) and highly accurate numerics. The small-L question is better answered by the analytic methods presented below, although we have confirmed those results by the direct calculations.

The conclusion must be stated quite carefully. For a fixed wave number k the onset frequency goes continuously to zero as a system parameter is varied, such as varying ψ to $\psi^*(k)$. In this situation a codimension-2 point is approached continuously. However, if the wave number of the onset solution is allowed to change to yield the lowest threshold Rayleigh number, then the frequency of the onset solution jumps discontinuously to zero at some value ψ_{pc}, together with a jump in the critical wave number.

These results are most easily illustrated for larger values of the Lewis number. We choose $L=0.8$, $\sigma=10$ purely for illustrative purposes. The results are shown in Fig. 4. Here we show the values of the onset Rayleigh numbers at the quadratic minima $R_s(k_s)$ and $R_o(k_o)$ for stationary and oscillatory convection, and the corresponding critical wave numbers k_s, k_o. For separation ratio corresponding to $\psi \simeq -0.135$ the critical Rayleigh numbers become equal: this is the point where the lowest threshold switches between the stationary and oscillatory instabilities. The wave numbers are quite different, and the frequency of the oscillatory mode is finite. The dependence of $R_s(k)$ and $R_o(k)$ are shown in Fig. 5. At $\psi=-0.112$ the minimum in the $R_o(k)$ curve disappears. Here the frequency of the onset solution at k_o tends continuously to zero, but the lowest threshold $R_o(k_o)$ is already considerably larger than stationary threshold $R_s(k_s)$.

For smaller L the jumps get much smaller and more care is needed in the numerics. Note in Fig. 2 that the wave number varies over a narrow range of ψ approaching the codimension-2 region, so that a fine grid of ψ values is needed to determine k_o, properly. This may explain the conclusion of Zielinska and Brand, who state that there is no wave-number jump. For the parameter values these authors use we find by the same methods as above that the wave-number jump when the minimum Rayleigh numbers coincide at ψ_{pc} is from $k_o=3.07$ to $k_s=3.15$ for $L=0.04$ and $\sigma=0.75$ and from $k_o=3.10(5)$ to $k_s=3.13$ for $L=0.02$ and $\sigma=17$. For small L the results of Sec. V give a better method to produce these results.

V. DERIVATION OF THE DEGENERATE-AMPLITUDE EQUATION NEAR THE CODIMENSION-2 POINT

A. Method

At the codimension-2 point $R^*(k), \psi^*(k)$ for fixed σ, L the eigenvalue equation (3.7) has two degenerate roots at $\lambda=0$. In this region we must expand the solution in terms of two amplitudes A,B giving the amplitudes of the two vectors spanning this null space.[2] As we will see, B is linearly related to $\partial_t A$, and the coupled equations for A and B can be rewritten as a second-order equation in time for A. Such an equation was derived for convection between free-slip, permeable boundaries by Brand *et al.*[2] We essentially repeat their method for rigid, impermeable boundaries. Again all quantities depend on the chosen horizontal wave number k, but we will usually not display this symbolically.

The null space is spanned by vectors $\mathbf{V}_1$ and $\mathbf{V}_2$ defined by

$$\hat{O}^*\mathbf{V}_1=0 , \tag{5.1}$$

$$\hat{O}^*\mathbf{V}_2=M\mathbf{V}_1 , \tag{5.2}$$

with $\hat{\mathbf{O}}^*$ the operator $\hat{O}$ with ψ and R tuned to their codimension-2 values $\psi^*(L,\sigma;k), R^*(L,\sigma;k)$. The vector $\mathbf{V}_2$ may be thought of as the limit of $(\mathbf{e}_1-\mathbf{e}_2)/(\lambda_1-\lambda_2)$, with $\mathbf{e}_1$ and $\mathbf{e}_2$ the eigenvectors with smallest eigenvalues λ_1, λ_2, as ψ, R approach ψ^*, R^* and $\lambda_1, \lambda_2 \to 0$.

We define the adjoint eigenvectors by

$$\hat{O}^{*\dagger}\mathbf{V}_2^{\dagger}=0 , \tag{5.3}$$

$$\hat{O}^{*\dagger}\mathbf{V}_1^\dagger = \hat{M}\mathbf{V}_2^\dagger , \tag{5.4}$$

with $\hat{O}^{*\dagger}$ the codimension-2 point adjoint operator. With this definition it is easy to see that $\mathbf{V}_2^\dagger$ is orthogonal to $\mathbf{V}_1$:

$$(\mathbf{V}_2^\dagger, \hat{M}\mathbf{V}_1) = 0 . \tag{5.5}$$

Since any amount of $\mathbf{V}_1$ may be added to $\mathbf{V}_2$ we may choose $\mathbf{V}_1^\dagger$ orthogonal to $\mathbf{V}_2$:

$$(\mathbf{V}_1^\dagger, \hat{M}\mathbf{V}_2) = 0 . \tag{5.6}$$

We now expand

$$\mathbf{V} = A(t)\mathbf{V}_1 + B(t)\mathbf{V}_2 + \delta\mathbf{V} , \tag{5.7}$$

with $\delta\mathbf{V}$ spanning the remainder of the space, i.e., $(\mathbf{V}_1^\dagger, \hat{M}\delta\mathbf{V})$ and $(\mathbf{V}_2^\dagger, \hat{M}\delta\mathbf{V})$ both zero, and $\hat{O} = \hat{O}^* + \delta\hat{O}$ with

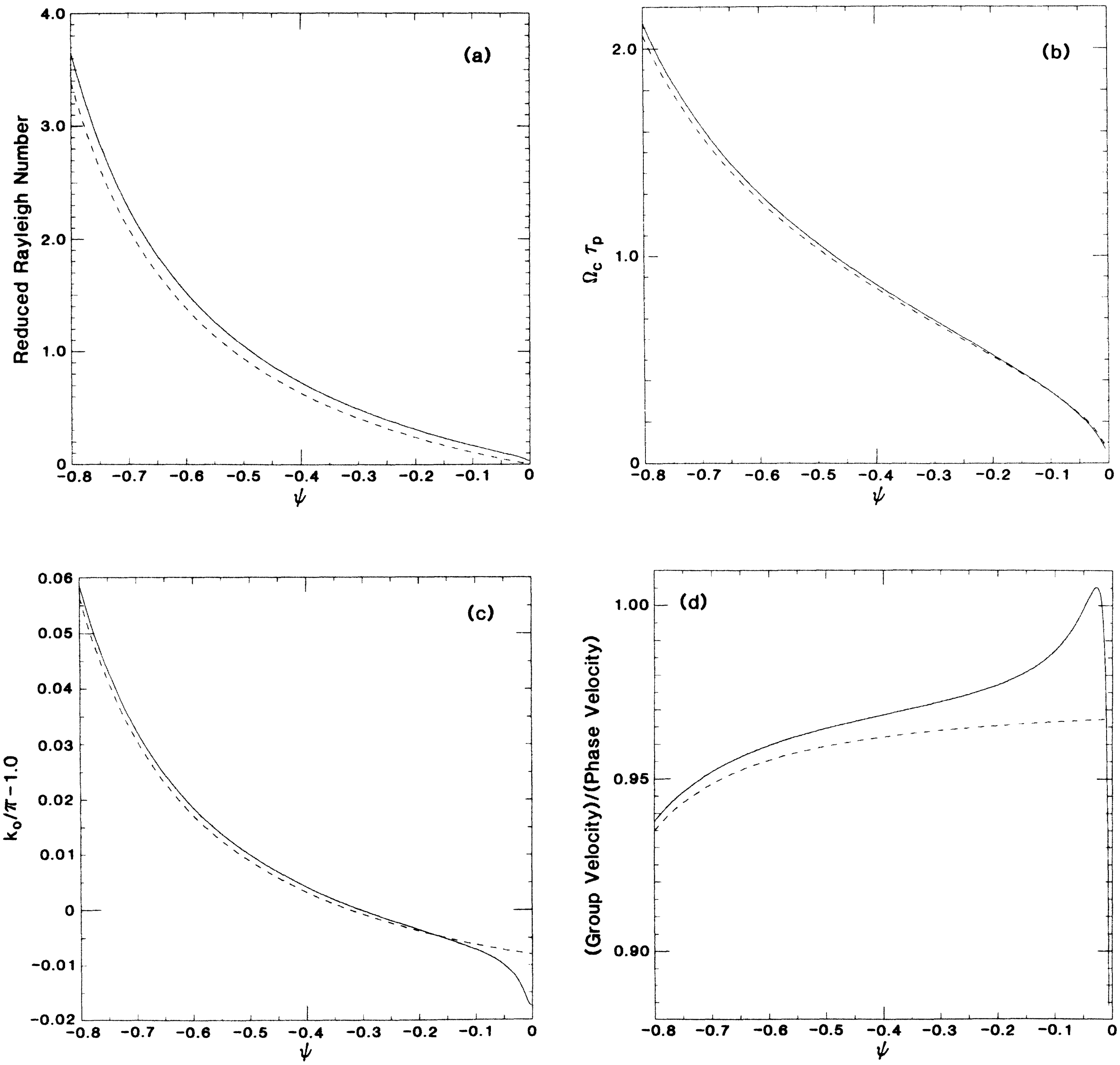

FIG. 1. Parameters of the linear onset and amplitude equation (1.1) for values of the separation-ratio ψ for Prandtl number $\sigma = 10$ and Lewis number $L = 0.05$ (solid line) and $L = 0$ (dashed line); (a) reduced critical Rayleigh number $(R_o - R_p)/R_p$ with $R_p = 1707.762$, the critical Rayleigh number for stationary convection in pure fluids; (b) oscillatory frequency at onset Ω_c scaled with $\tau_p = (\sigma + 0.5117)/19.65\sigma$; (c) critical wave number k_o; (d) ratio of group velocity s to phase velocity Ω_c/k_o; (e) τ_0^{-1} scaled with τ_p; (f) the square of the correlation length ξ_0^2; (g) c_0; and (h) $c_1 - c_0$.

$$\delta\hat{O}=\begin{bmatrix} 0 & -\delta\psi k^2 & 0 \\ \delta R & 0 & 0 \\ 0 & \delta\psi\nabla^2 & 0 \end{bmatrix} \tag{5.8}$$

and $\delta R = R - R^*$, $\delta\psi=\psi-\psi^*$.

Substituting Eq. (5.7) into Eq. (3.2), taking inner products with $\mathbf{V}_1^\dagger,\mathbf{V}_2^\dagger$, and keeping only first-order small quantities leads to the equations

$$\begin{aligned}(\mathbf{V}_1^\dagger,\hat{M}\mathbf{V}_1)\partial_t A &= (\mathbf{V}_1^\dagger,\delta\hat{O}\mathbf{V}_1)A + [(\mathbf{V}_1^\dagger,\hat{M}\mathbf{V}_1) \\ &\quad + (\mathbf{V}_1^\dagger,\delta\hat{O}\mathbf{V}_2)]B\ , \\ (\mathbf{V}_2^\dagger,\hat{M}\mathbf{V}_2)\partial_t B &= (\mathbf{V}_2^\dagger,\delta\hat{O}\mathbf{V}_1)A + (\mathbf{V}_2^\dagger,\delta\hat{O}\mathbf{V}_2)B\ .\end{aligned} \tag{5.9}$$

Eliminating B yields the second-order equation

$$\partial_t^2 A + \alpha\partial_t A + \beta A = 0\ , \tag{5.10}$$

with

$$\alpha = -\frac{(\mathbf{V}_1^\dagger,\delta\hat{O}\mathbf{V}_1)}{(\mathbf{V}_1^\dagger,\hat{M}\mathbf{V}_1)} - \frac{(\mathbf{V}_2^\dagger,\delta\hat{O}\mathbf{V}_2)}{(\mathbf{V}_2^\dagger,\hat{M}\mathbf{V}_2)}\ , \tag{5.11}$$

$$\beta = -\frac{(\mathbf{V}_2^\dagger,\delta\hat{O}\mathbf{V}_1)}{(\mathbf{V}_2^\dagger,\hat{M}\mathbf{V}_2)}\ . \tag{5.12}$$

Finally we write

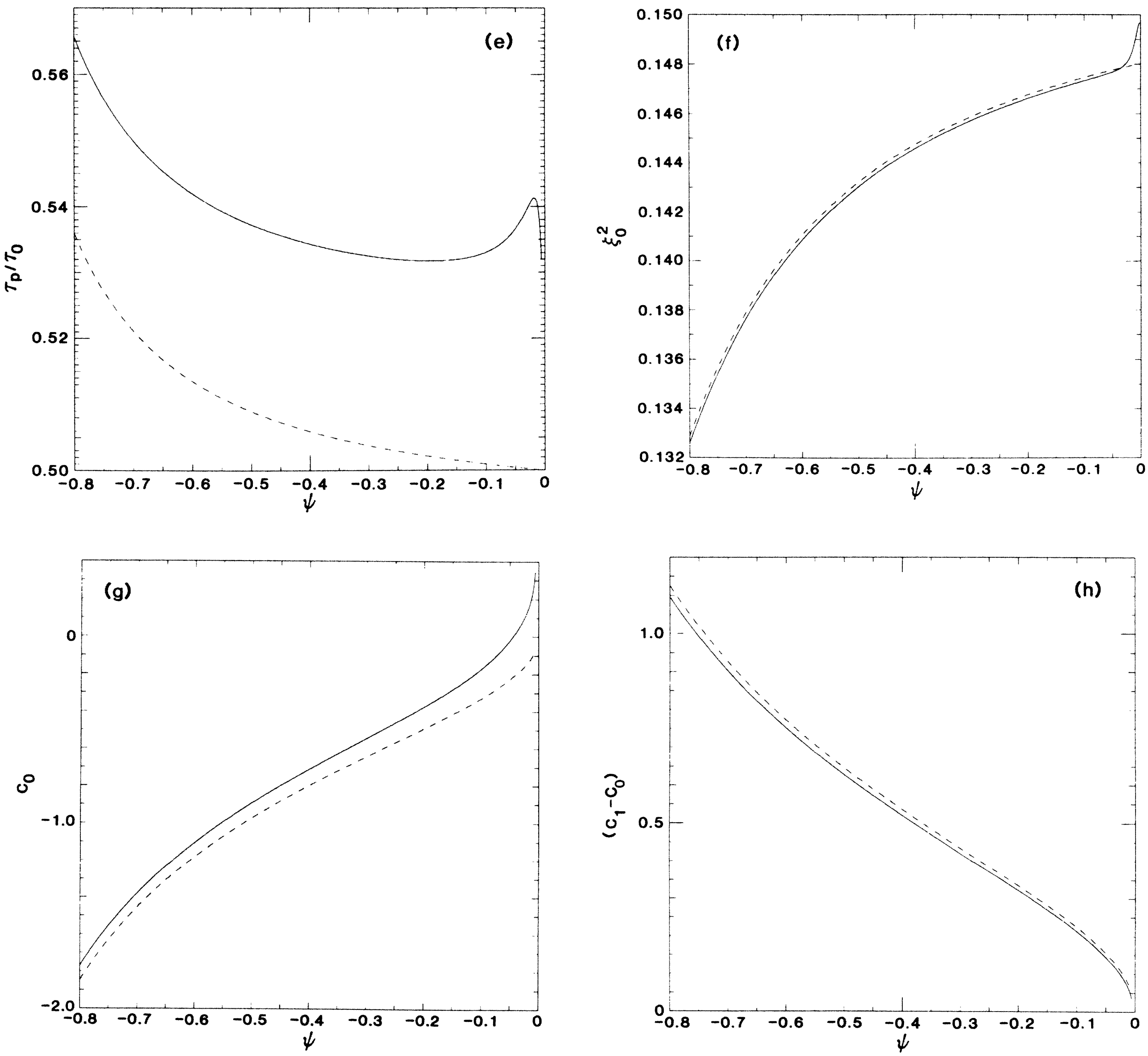

FIG. 1. (*Continued*).

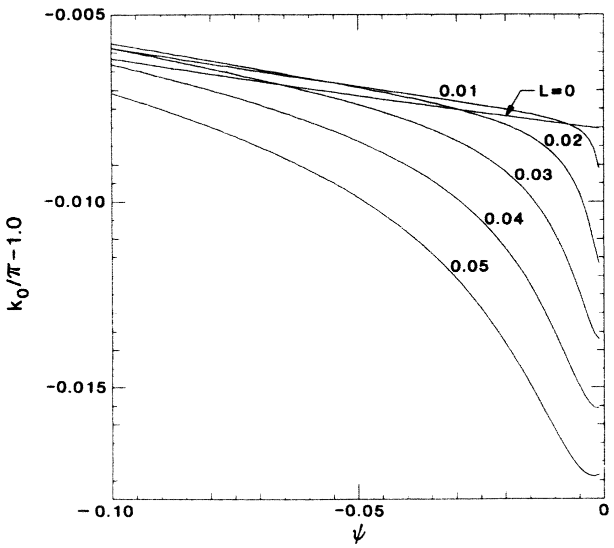

FIG. 2. Expanded region of Fig. 1(c) showing the dependence of the critical wave number on L for $|\psi|\sim L$.

$$\alpha=\alpha_\epsilon\epsilon+\alpha_\psi\delta\psi\ , \tag{5.13}$$
$$\beta=\beta_\epsilon\epsilon+\beta_\psi\delta\psi\ ,$$

where $\epsilon=(R-R^*)/R_p$ with R_p the critical Rayleigh number for pure stationary convection at its critical wave number (a convenient scaling) and calculate $\alpha_\psi,\alpha_\epsilon,\beta_\psi,\beta_\epsilon$ for each k and given L,σ.

B. Implementation

Away from the codimension-2 point Eq. (5.4) is singular. Requiring solvability of this equation together with Eq. (5.3) provides an accurate scheme for determining R^*,ψ^*. The actual procedure is as follows.

The solution $\mathbf{V}_1$ is given as before by the form (3.12) with q_i given by the roots of

$$Q[Q^3-Rk^2(1+\psi+\psi L^{-1})]=0\ , \tag{5.14}$$

with $Q=q^2+k^2$, which, of course, may be solved analytically. (Note that care is needed in manipulating the $Q=0$ solution as outlined in Ref. 9.) As before, the c_i are determined up to an overall normalization factor by the boundary condition providing $R=R_c(\psi)$.

The vector $\mathbf{V}_2$ takes the form

$$w_2=\sum_{i=1}^{4}\bar{c}_{1i}\cos q_iz+d_{1i}z\sin q_iz\ , \tag{5.15a}$$

$$\Theta_2=\sum_{i=1}^{4}\bar{c}_{2i}\cos q_iz+d_{2i}z\sin q_iz\ , \tag{5.15b}$$

$$\eta_2=\sum_{i=1}^{4}\bar{c}_{3i}\cos q_iz+d_{3i}z\sin q_iz\ . \tag{5.15c}$$

Matching the coefficients of $\cos q_iz$ and $z\sin q_iz$ in Eq. (5.4) gives only five independent equations for the six unknowns $\bar{c}_{ji},d_{ji}$ for each i, since an arbitrary multiple of the component in $\cos q_iz$ in $\mathbf{V}_1$ can be added to $\mathbf{V}_2$ at this stage. We now have four undetermined parameters. For the four linear, homogeneous boundary-condition equations to be satisfied we need an extra parameter to vary: This finally fixes ψ^* and $R^*=R_c(\psi^*)$, and all parameters $\bar{c}_{ji},d_{ji}$.

We will not explicitly display results here. We have used this independent method to check the approach to the codimension-2 region, including wave-number jumps, in Sec. IV, and the small-L results derived below.

VI. DERIVATION OF THE CODIMENSION-2 AMPLITUDE EQUATION FOR SMALL L

From the results of Sec. V, we expect the codimension-2 region to be $-\psi\sim L^2$ as $L\to 0$, as is true for the free-slip, permeable problem. The forward equations may then be approximated for small L by

$$\nabla^4w-k^2\Theta+k^2\eta=\sigma^{-1}\nabla^2\partial_tw\ , \tag{6.1a}$$

$$Rw+\nabla^2\Theta=\partial_t\Theta\ , \tag{6.1b}$$

$$\psi\nabla^2\Theta+L\nabla^2\eta=\partial_t\eta\ , \tag{6.1c}$$

with boundary conditions as before, and we have made the replacement $1+\psi\to 1$ in the first equation since $\psi\sim L^2$. We now want to solve these equations, explicitly extracting the small-L behavior.

(i) *Solve* $\hat{O}\mathbf{V}_1=0$. If we assume η is small, then the first two equations (6.1a) and (6.1b) for this result are solved by $w_1=w_p$, $\Theta_1=\Theta_p$ with $w_p(z),\Theta_p(z)$ the $O(1)$ solutions for *pure* (i.e., $\psi=0$), stationary convection and, providing we choose R to be $R_p(k)$, the critical Rayleigh number for pure, stationary convection at this wave number. Equation (6.1c) then gives

$$\nabla^2\eta_1=L(\gamma\nabla^2\Theta_p)\ , \tag{6.2}$$

with $\gamma=-\psi/L^2=O(1)$, and is to be solved with the boundary conditions $\partial_z\eta=0$ at $z=\pm\frac{1}{2}$. We write the solution as

$$\eta_1=L(\gamma\Theta_p+\eta_p)\ , \tag{6.3}$$

with $\nabla^2\eta_p=0$ and η_p chosen so that η satisfies the boundary conditions. Clearly, η_p is $O(1)$ and η_1 is $O(L)$, consistent with the original assumption. Thus

$$\mathbf{V}_1=[w_p+O(L),\Theta_p+O(L),L(\gamma\Theta_p+\eta_p)]\ . \tag{6.4}$$

(ii) *Solve* $\hat{O}\mathbf{V}_2=\mathbf{V}_1$. First we solve

$$\nabla^2\eta_2=(\gamma\Theta_p+\eta_p)\ , \tag{6.5}$$

together with the boundary conditions on η, having neglected the $O(L)$ term. We then have

$$\begin{bmatrix}\nabla^4 & -k^2\\ R & -k^2\end{bmatrix}\begin{bmatrix}w_2\\ \Theta_2\end{bmatrix}=\begin{bmatrix}\sigma^{-1}\nabla^2w_p-k^2\eta_2\\ \Theta_p\end{bmatrix}\ . \tag{6.6}$$

The parameter $\gamma=-\psi/L^2$ is determined through Eq. (6.5) and the solvability condition that the right-hand side of Eq. (6.6) be orthogonal to the null vector of the adjoint of the operator acting on (w_2,Θ_2), i.e., to $(w_p^\dagger,\Theta_p^\dagger)$. Equations (6.6) are then solved as before, so that $\mathbf{V}_2=O(1)$ and may be calculated.

The adjoint problem is defined by the equations

$$\nabla^4 w^\dagger + R\Theta^\dagger = \sigma^{-1}\nabla^2\partial_t w^\dagger \ , \tag{6.7a}$$

$$-k^2 w^\dagger + \nabla^2\theta^\dagger + \psi\nabla^2\eta^\dagger = \partial_t\Theta^\dagger \ , \tag{6.7b}$$

$$k^2 w^\dagger + L\nabla^2\eta^\dagger = \partial_t\eta^\dagger \ , \tag{6.7c}$$

with boundary conditions (3.10).

(iii) *Solve* $\hat{O}^\dagger \mathbf{V}_2^\dagger = 0$. Following the same arguments the solution is

$$\mathbf{V}_2^\dagger = (Lw_p^\dagger, L\Theta_p^\dagger, \eta_p^\dagger) \ , \tag{6.8}$$

with $\eta_p^\dagger$ solving

$$\nabla^2\eta_p^\dagger = -k^2 w_p^\dagger \tag{6.9}$$

and the boundary conditions, and $(w_p^\dagger, \Theta_p^\dagger)$ the solutions of the adjoint problem of pure, stationary convection.

(iv) *Solve* $\hat{O}^\dagger \mathbf{V}_1^\dagger = \mathbf{V}_2^\dagger$. The solution $V_1^\dagger$ is given by solving

$$\nabla^2\eta_1^\dagger = L^{-1}\eta_p^\dagger \tag{6.10}$$

and then

$$\begin{bmatrix} \nabla^4 & R \\ -k^2 & \nabla^2 \end{bmatrix} \begin{bmatrix} w_1^\dagger \\ \Theta_1^\dagger \end{bmatrix} = L \begin{bmatrix} \sigma^{-1}\nabla^2 w_p^\dagger \\ \Theta_p^\dagger + \gamma\eta_p^\dagger \end{bmatrix} \ . \tag{6.11}$$

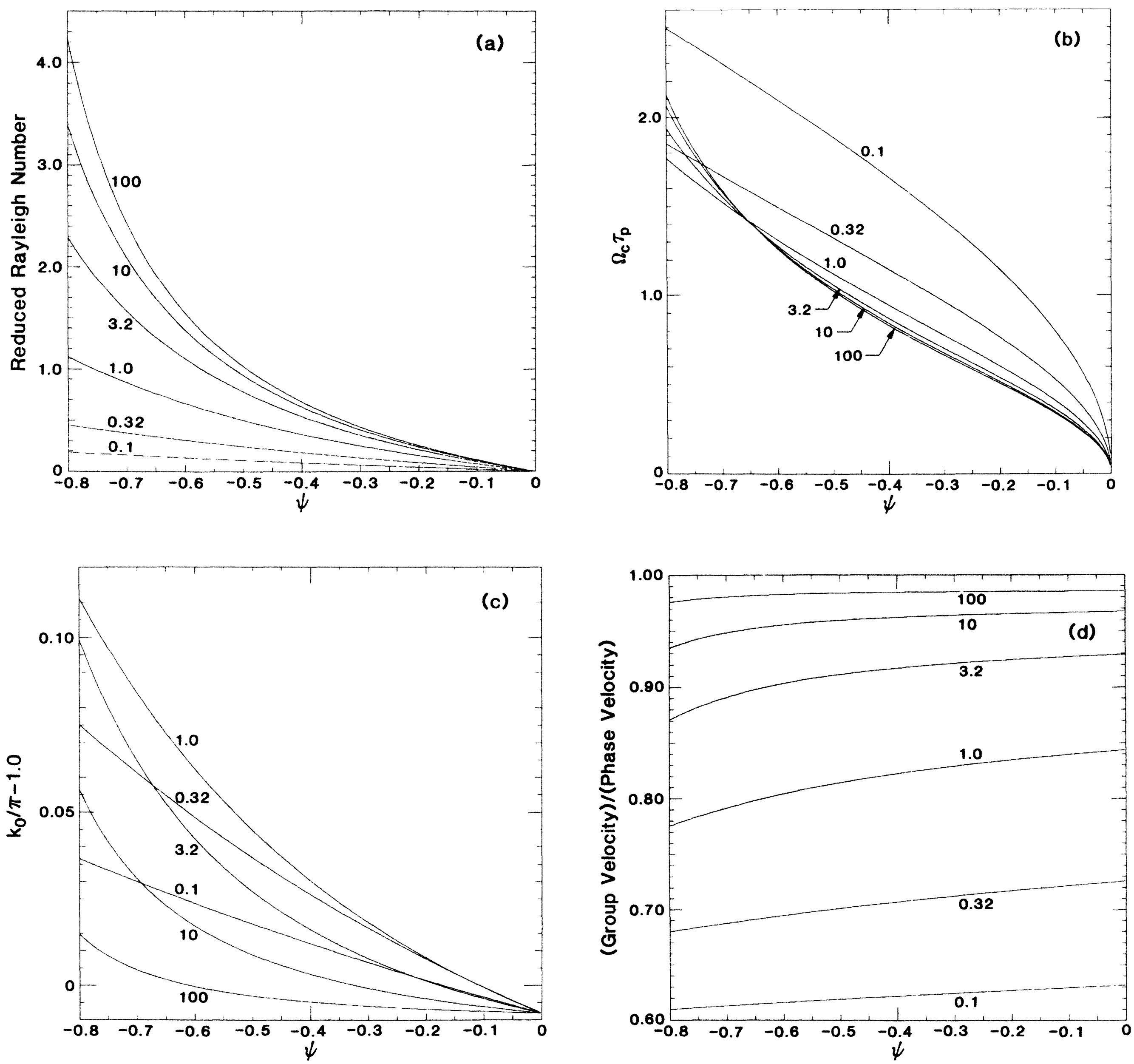

FIG. 3. Prandtl-number dependence of the linear parameters as in Fig. 1 for $L=0$ for Prandtl numbers from 0.1 to 100.

Again the (same) value of γ is given by the solvability condition and $\mathbf{V}_1^\dagger$ may be chosen as

$$\mathbf{V}_1^\dagger=[O(L),O(L),L^{-1}\bar{\eta}_1^\dagger] , \qquad (6.12)$$

with $\bar{\eta}_1^\dagger=L\eta_1^\dagger=O(1)$. Note that with this normalization $(\mathbf{V}_1^\dagger,\hat{M}\mathbf{V}_1)$ and $(\mathbf{V}_2^\dagger,\hat{M},\mathbf{V}_2)$ are $O(1)$ and the $O(L)$ terms in $\mathbf{V}_1,\mathbf{V}_2,\mathbf{V}_1^\dagger,\mathbf{V}_2^\dagger$ not explicitly quoted above are not needed. Also Eq. (5.6) is not satisfied, and must be enforced explicitly by the Gramm-Schmidt procedure.

Finally, we have, keeping each result to leading order in L,

$$\alpha_\epsilon=\frac{\langle L\eta_1^{\dagger *}\eta_2\rangle\langle L^{-1}\Theta_2^{\dagger *}w_1\rangle}{\langle L\eta_1^{\dagger *}L^{-1}\eta_1\rangle\langle\eta_2^{\dagger *}\eta_2\rangle}=O(1) , \qquad (6.13)$$

$$\alpha_\psi=L^{-1}\left[\frac{\langle L\eta_1^{\dagger *}\eta_2\rangle\langle\eta_2^{\dagger *}\nabla^2\Theta_1\rangle}{\langle L\eta_1^{\dagger *}L^{-1}\eta_1\rangle\langle\eta_2^{\dagger *}\eta_2\rangle}-\frac{\langle L\eta_1^{\dagger *}\nabla^2\Theta_1\rangle}{\langle L\eta_1^{\dagger *}L^{-1}\eta_1\rangle}\right] =O(L^{-1}) , \qquad (6.14)$$

$$\beta_\epsilon=-L\frac{\langle L^{-1}\Theta_2^{\dagger *}w_1\rangle}{\langle\eta_2^{\dagger *}\eta_2\rangle}=O(L) , \qquad (6.15)$$

$$\beta_\psi=-\frac{\langle\eta_2^{\dagger *}\nabla^2\Theta_1\rangle}{\langle\eta_2^{\dagger *}\eta_2\rangle}=O(1) . \qquad (6.16)$$

It can readily be checked from the derivation that the only Prandtl-number dependence of the α's and β's in this small-L limit comes through the proportionality of

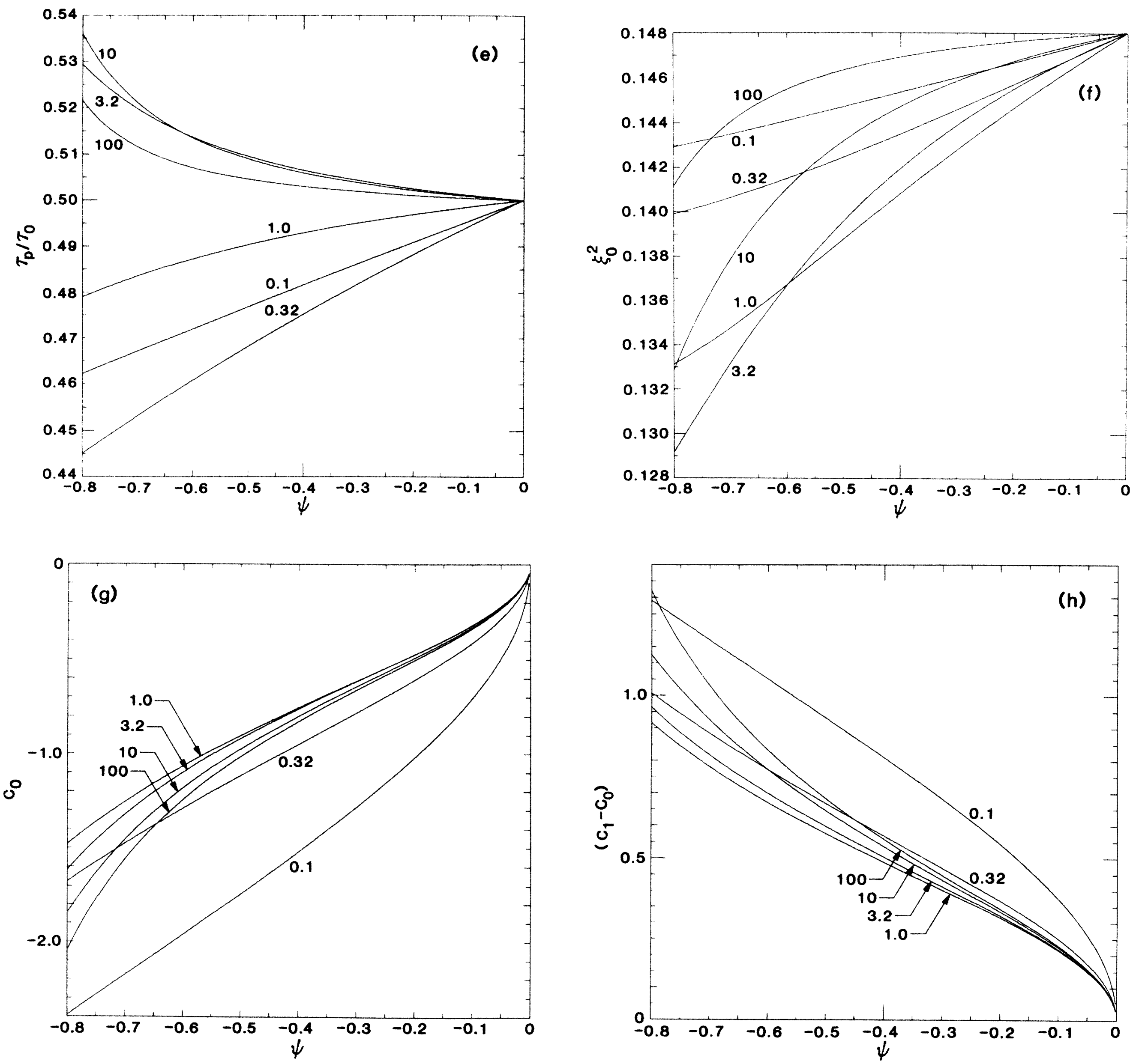

FIG. 3. (*Continued*).

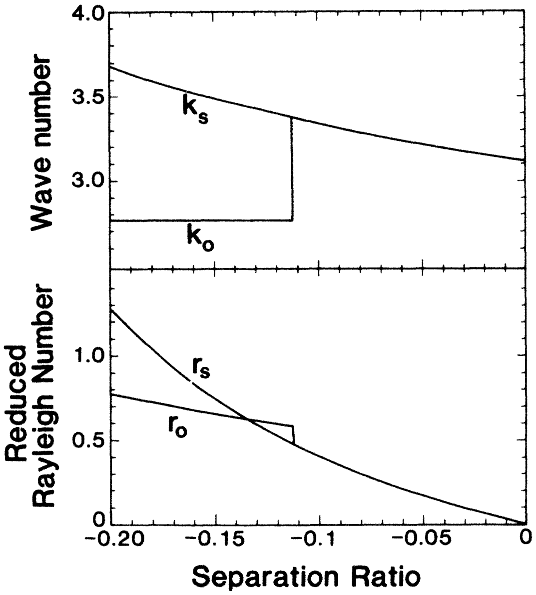

FIG. 4. Critical wave numbers k_s and k_o and reduced critical Rayleigh number $(R_s - R_p)/R_p$ and $(R_o - R_p)/R_p$ (with $R_p = 1707.76$) for stationary and oscillatory instabilities for $L = 0.8$, $\sigma = 10$.

η_1 and η_2 to $\gamma = -\psi^*/L^2$, and the Prandtl-number dependence of this quantity.

VII. RESULTS FOR CODIMENSION-2 AMPLITUDE EQUATION FOR SMALL L

The Prandtl-number dependence of the separation ration at the codimension-2 point given by $\gamma(k) = -\psi^*(k)/L^2$, and also its wave-number derivative, both evaluated at wave number k_p (the critical wave number for stationary convection in pure fluids), are shown in Fig. 6 and tabulated in Table I. The quantities $\gamma(k)\alpha_\epsilon(k)$, etc., are independent of Prandtl number, and take the values, again at wave number k_p,

$$\begin{aligned} \gamma\alpha_\epsilon &= -6.70 , \\ \gamma\alpha_\psi &= -0.623L^{-1} , \\ \gamma\beta_\epsilon &= -65.3L , \\ \gamma\beta_\psi &= -101.9 . \end{aligned} \tag{7.1}$$

We have checked these results using the method of Sec. V, calculating for values of L down to 10^{-4}. The approach of α_ψ to the small-L limit is rather slow, approximately $L\gamma\alpha_\psi = -0.623(1+10.6L)$. The wave-number derivatives at $k = k_p$ are

$$\begin{aligned} k_p(\gamma\alpha_\epsilon)'/\gamma\alpha_\epsilon &= 2.97 , \\ k_p(\gamma\alpha_\psi)'/\gamma\alpha_\psi &= 2.97 , \\ k_p(\gamma\beta_\epsilon)'/\gamma\beta_\epsilon &= 4.98 , \\ k_p(\gamma\beta_\psi)'/\gamma\beta_\psi &= 4.10 \end{aligned} \tag{7.2}$$

(where the prime denotes d/dk), again independent of Prandtl number.

From these results we may immediately calculate the slope $m = \epsilon/\delta\psi$ of the approach of the instability lines $\epsilon(\delta\psi)$ to the codimension-2 point for the oscillatory and

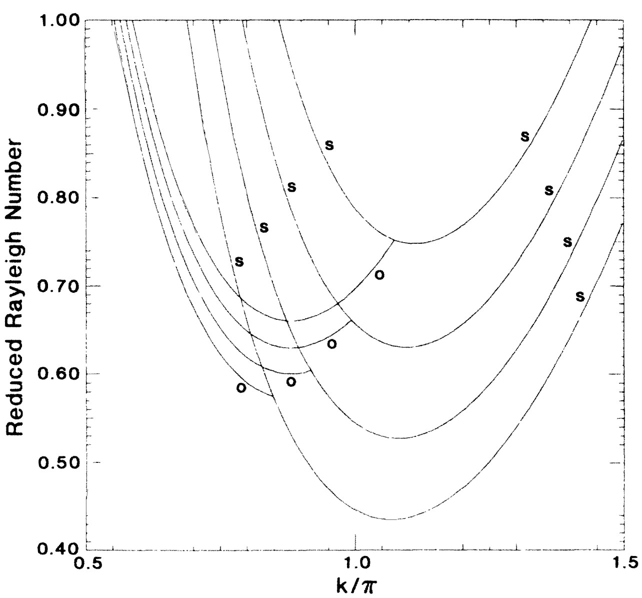

FIG. 5. Onset reduced Rayleigh numbers $[R_s(k) - R_p]/R_p$ and $[R_o(k) - R_p]/R_p$ for $L = 0.8$ as a function of wave number k for stationary (s) and oscillatory (o) instabilities. Values of the separation ratio are (top to bottom) -0.150, -0.135, -0.120, and -0.105.

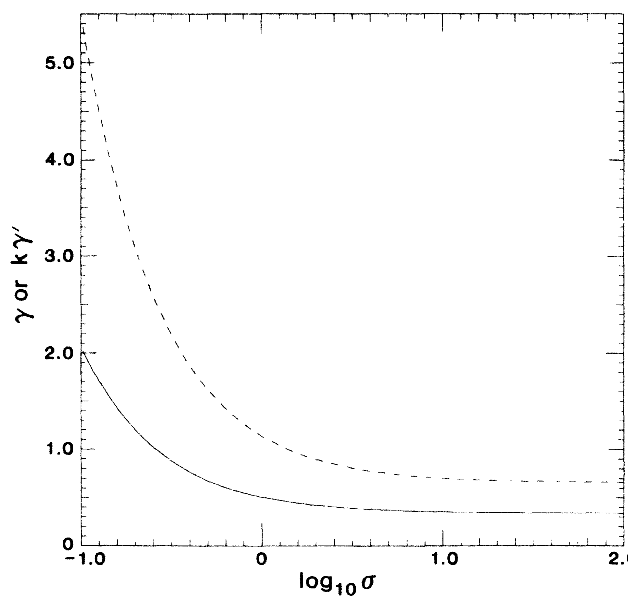

FIG. 6. $\gamma(k_p)$ (solid line) and $k\, d\gamma/dk\,|_{k=k_p}$ (dashed line) as a function of Prandtl number σ. Here $\gamma(k) = -\psi^*(k)/L^2$ with $\psi^*(k)$ the value of the separation ratio at the codimension-2 point for fixed wave number k, and $k_p = 0.992\pi$ is the critical wave number for pure stationary convection.

TABLE I. Values of $\gamma=-\psi^*(k)/L^2$ and $k\gamma'=k\,d\gamma/dk$ evaluated at $k=k_p$, the critical wave number for the onset of stationary convection in a pure fluid.

σ	γ	$k\gamma'$
0.1	2.064	5.497
0.2	1.201	3.077
0.3	0.913	2.271
0.4	0.769	1.868
0.5	0.683	1.626
0.6	0.625	1.465
0.8	0.553	1.263
0.9	0.529	1.196
1.0	0.510	1.142
2.0	0.424	0.900
3.0	0.395	0.820
4.0	0.381	0.779
5.0	0.372	0.755
6.0	0.366	0.739
7.0	0.362	0.727
9.0	0.357	0.712
10.0	0.355	0.707
∞	0.337	0.658

stationary instabilities. At $k=k_p$, for the stationary instability,

$$m_s(k_p)=-\frac{\beta_\psi(k_p)}{\beta_\epsilon(k_p)}=-1.561L^{-1}\ , \tag{7.3}$$

and for the oscillatory instability,

$$m_o(k_p)=-\frac{\alpha_\psi(k_p)}{\alpha_\epsilon(k_p)}=-0.093L^{-1}\ . \tag{7.4}$$

We may also calculate the wave-number jump for small L when the minimum onset Rayleigh numbers coincide. The stationary instability is given by

$$\epsilon_s(k)=\frac{R_s(k)-R^*(k)}{R_p}=-m_s(k)[\psi-\psi^*(k)]\ , \tag{7.5}$$

with $\psi^*(k)$ the value of ψ at the codimension-2 point for each value of k and $R^*(k)$ the degenerate onset Rayleigh number for this k,ψ^*. To find k_s, the critical wave number for the stationary instability, we minimize $R_s(k)$ with respect to k. Similarly, for the oscillatory instability we find k_o by minimizing $R_o(k)$ given by

$$\epsilon_o(k)=\frac{R_o(k)-R^*(k)}{R_p}=-m_o(k)[\psi-\psi^*(k)]\ . \tag{7.6}$$

For $L\to 0$, k_o and k_s coincide at the value k_p, the critical wave number for stationary convection. To find $\Delta k=k_s-k_o$ for small L we expand $R^*(k)$ about its quadratic minimum, and m_o,m_s and ψ^* linearly about this point. To lowest order

$$R^*(k)\simeq R_p(k)\simeq R_p[1+\xi_p^2(k-k_p)^2]\ , \tag{7.7}$$

with ξ_p again calculated for pure stationary convection. Substituting into the minimization and equating $R_s(k_s)$ and $R_o(k_o)$ we find to lowest order in powers of L:

$$\frac{\Delta k}{k_p}=\frac{k_s-k_o}{k_p}\simeq\frac{[m_o(k_p)-m_s(k_p)]}{2(k_p\xi_p)^2}[k_p\psi'(k_p)]\ . \tag{7.8}$$

Using $k_p=0.992\pi$, $\xi_p^2=0.148$, and the values of m_o,m_s above gives

$$\Delta k/k_p=0.511k_p\gamma'(k_p)L\ , \tag{7.9}$$

with $k_p\gamma'$ plotted in Fig. 6. The values for $\sigma=0.75$ and $L=0.04$, and $\sigma=17$ and $L=0.02$, are consistent with the direct calculations reported in Sec. IV. Results for other parameter values are easily found from Fig. 6.

We may also calculate the frequency Ω_c of the oscillatory solution at this point, using $\Omega_c^2=\beta$ and $\alpha=0$ at the oscillatory onset, so that

$$\Omega_c^2=\beta_\psi[\psi-\psi^*(k_o)]\left[1-\frac{\beta_\epsilon\alpha_\psi}{\beta_\psi\alpha_\epsilon}\right]\ . \tag{7.10}$$

Using the value of ψ calculated by the equality of $R_s(k_s)$ and $R_o(k_o)$ gives

$$\Omega_c^2=-\frac{\beta_\psi^2}{\beta_\epsilon}\frac{'(k_p\gamma')^2}{4(k_p\xi_p)^2}\left[1-\frac{\beta_\epsilon\alpha_\psi}{\beta_\psi\alpha_\epsilon}\right]^2\ , \tag{7.11}$$

using the values given above this yields

$$\Omega_c=4.90(k_p\gamma'/\gamma^{1/2})L^{3/2}\ . \tag{7.12}$$

This gives the "jump" in the oscillatory frequency for the threshold solution for any Prandtl number with $k_p\gamma'$ and γ evaluated from Fig. 6. For example, for $L=0.04$ and $\sigma=0.75$, we predict jump of 0.067 and for $L=0.02$ and $\sigma=17$, a frequency jump of 0.016.

VIII. CONCLUSIONS

We have derived the linear parameters for the amplitude equation describing the onset of oscillatory convection, and of the degenerate-amplitude equation near the codimension-2 point, for the experimentally common situation of rigid, impervious boundaries. We have particularly focused on the small-Lewis-number limit. Away from the codimension-2 region we suggest that the $L=0$ calculation (which does not suffer from the numerical difficulties of the small$-L$ calculation) provides quite accurate answers for typical experimental values. We have also considered the codimension-2 region for small L, explicitly controlling the leading L dependence of the terms in the degenerate-amplitude equation, so that again we have an easy numerical scheme that does not involve very small numbers. These results allow us to calculate accurate values for the separation ratio at the codimension-2 point at fixed wave number k for various Prandtl numbers, and other interesting quantities such as the wave-number and frequency jumps approaching the codimension-2 region if the onset wave number is allowed to change to follow the lowest threshold. In this regime all the Prandtl-number dependence is contained in $\psi^*(k)$, and results for any Prandtl number are easily evaluated from the plots in Fig. 6.

The codimension-2 region warrants some comments.

From our calculations we find, as might be expected, the quantities calculated from the codimension-2 amplitude equation (e.g., onset frequency Ω_c as a function of $\delta\psi=\psi-\psi^*$) are valid only for $|\delta\psi| \lesssim L^2$. Outside of this region, tiny for small L, Ω_c^2 is apparently approaching zero linearly [cf. Fig. 1(b)], but with a quite different slope than for $|\delta\psi| \lesssim L^2$. In fact, for $-\delta\psi \gtrsim L$ the behavior is close to the $L=0$ calculations, where again $\Omega_c^2(\psi)$ approaches zero linearly. The reason for this crossover is the expansion of the concentration boundary layer of width $\sim(L/\Omega)^{1/2}$ from a negligible value for large Ω to fill the cell for small Ω: as $\Omega \to 0$ the character of the solution dramatically changes. Thus the range of validity of the codimension-2 behavior in $\delta\psi$ is of order L^2, shrinking to zero with L. For L identically zero the onset frequency does indeed go smoothly to zero at $\psi=0$, with the characteristic square-root dependence [Fig. 3(b)] suggesting a "codimension-2" description. However, the point $\psi=0$ is highly singular for $L=0$, and we have not been able to construct a smooth expansion about this point. (For example, for $\psi=0^+$ the critical wave number of the *stationary* solution drops immediately to zero.) We also expect the nonlinearities to have a singular behavior for $L \to 0$, again with very low amplitude flows strongly affecting the nature of the boundary layers for small L. Thus (unless the degenerate $L=0$ behavior can be controlled) it appears to us that descriptions based on the degenerate bifurcation at $\psi \sim -L^2$ have a range of validity limited to the very small $O(L^2)$ region about this point.

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[1] Unfortunately there is no review summarizing the recent flurry of activity. Some of the most recent papers are by E. Moses, J. Fineberg, and V. Steinberg, Phys. Rev. A **35**, 2751 (1987); R. Heinrichs, G. Ahlers, and D. S. Cannell, *ibid.* **35**, 2761 (1987); P. Kolodner, A. Passner, C. M. Surko, and R. W. Walden, Phys. Rev. Lett. **56**, 2621 (1986). A review of work in helium mixtures is R. P. Behringer, Rev. Mod. Phys. **57**, 657 (1985).

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