

Exact solution of the linear-stability problem for the onset of convection in binary fluid mixtures

Barbara J. A. Zielinska* and Helmut R. Brand†

Department of Physics, The Weizmann Institute of Science, 76 100 Rehovot, Israel

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We solve the linearized problem for the onset of convection in binary fluid mixtures in a layer of infinite lateral extent for rigid-rigid, no-flux boundary conditions exactly. We present detailed data for the critical Rayleigh number and for the neutral frequency as a function of the separation ratio for the two experimentally most relevant systems: for ethanol-water and ^3He - ^4He mixtures. For both the oscillatory and the stationary onset we compare our results with those obtained for simple trial functions. The influence of the effect of rigid-rigid, no-flux boundary conditions on the location of the codimension-2 point is examined.

I. INTRODUCTION

Stimulated by earlier theoretical work on the onset of convection in binary fluid mixtures,¹⁻⁹ several groups started to investigate this problem experimentally.¹⁰⁻¹⁶ Some of the experimental results, especially those on the behavior close to the intersection between the stationary and the oscillatory onset in a porous medium¹⁰ were in agreement with the theoretical predictions.^{5,8} Other observations such as finite-amplitude propagating waves at onset^{13,14} have not been predicted theoretically. Naturally the question arises how these unexpected phenomena can be explained theoretically. Scanning the theoretical approaches one finds out easily that all nonlinear calculations reported so far were based on the use of unphysical free slip, impervious boundary conditions. In addition, it appears that even the linearized stability problem on the oscillatory branch has not been solved exactly for the case of physically realistic rigid-rigid, no-flux boundary conditions.

In the present paper we solve exactly the linearized stability problem for a layer of laterally infinite extent for the otherwise physically realistic rigid-rigid, no-flux boundary conditions, both on the stationary and on the oscillatory branch. We present detailed results of our numerical calculations for the two practically most important and experimentally studied cases: ethanol-water mixtures at room temperatures and ^3He - ^4He mixtures at low temperatures. We give detailed plots for the critical Rayleigh number and the neutral frequency at onset as a function of the separation ratio and we discuss the behavior of the critical wave vector. We investigate the shift in the location of the codimension-2 (CT) point for rigid-rigid, no-flux boundary conditions as compared to the free-free, impervious ones. We compare all our exact results with those obtained from various types of test functions using the Galerkin method.¹⁷ The paper is organized as follows. In the following section we describe the problem and we sketch the procedure used to solve it; in Sec. III we present the results and in Sec. IV we give the conclusions including the discussion of the limitations of the linear-

ized stability analysis when applied to experimental results.

II. THE LINEARIZED BOUNDARY VALUE PROBLEM

In the present work we focus on a laterally infinite layer of a binary mixture of miscible fluids and the physically realistic rigid-rigid (stick), no-flux boundary conditions. The solution for the rigid-rigid, no-flux boundary conditions we will refer to as *exact*, in contrast to the solution for the physically unrealistic free-free (slip), impervious boundary conditions. The stability problem with rigid-rigid boundary conditions for simple fluids was solved many years ago (cf., e.g., Chandrasekhar¹⁸). One obtains $R_c = 1707.8$ for the critical Rayleigh number and $k_c = 3.117$ for the critical wave number. For binary mixtures the exact problem for the stationary bifurcation was solved by Gutkowicz-Krusin *et al.*³ Various authors have also used expansions in trigonometric functions, the method of Galerkin, or variational principles to obtain approximate solutions both on the stationary and oscillatory branch. While the Galerkin method has been proven to yield results converging to the exact solution for self-adjoint operators (i.e., for the stationary bifurcation), no such proof exists for non-self-adjoint operators (i.e., for the oscillatory bifurcation). Therefore it was not clear how the results obtained by this method for the onset of the oscillatory convection compared to the exact solution. In this paper we present the exact solution for the oscillatory branch and compare it with results obtained from the Galerkin method using two types of simple trial functions. We find that the trial functions $(z^2 - \frac{1}{4})^2$ for the vertical velocity field and for the mass flux, and $z^2 - \frac{1}{4}$ for the temperature (z being the direction perpendicular to the fluid layer) are a good approximation to the exact solution and can be used as a reliable estimate for both the critical Rayleigh number and the neutral frequency.

We consider a laterally infinite layer of a binary fluid mixture between two plates in an external temperature gradient. The equations linearized around the heat conduction state (in dimensionless units) describing this system are, in generalized Boussinesq approximation,^{1-3,5}

$$\begin{aligned}\Delta(\partial_t/P - \Delta)w &= \Delta_2[(1 + \Psi)\theta + \Psi\eta], \\ (\partial_t - \Delta)\theta &= R w, \\ (\partial_t - \mathcal{L}\Delta)\eta &= -\Delta\theta,\end{aligned}\quad (1)$$

where the scaled variables w , θ , and $\eta = c - \theta$ are the z component of the velocity field, the temperature, and the mass flux, respectively, c being the concentration of one component of the mixture. $R = \beta_1 g l^3 \Delta T / \nu \kappa$ is the Rayleigh number corresponding to the temperature difference ΔT , $\Psi = -k_T \beta_2 / T \beta_1$ is the separation ratio, g is the gravitational constant, l is the height of the fluid layer, $\beta_1 = -\rho^{-1}(\partial\rho/\partial T)_{P,c}$ is the thermal expansion coefficient, $\beta_2 = -\rho^{-1}(\partial\rho/\partial c)_{P,T}$, ν is the kinematic viscosity, κ is the thermodiffusivity, and k_T is the thermodiffusion ratio. $P = \nu/\kappa$ is the Prandtl number and $\mathcal{L} = D/\kappa$ is the Lewis number, D being the diffusion coefficient. $\Delta \equiv \partial_x^2 + \partial_y^2 + \partial_z^2$, $\Delta_2 \equiv \partial_x^2 + \partial_y^2$, and we assumed that the positive z axis points in the direction of the temperature gradient. The time is scaled with l^2/κ , velocity with κ/l , temperature with $\nu\kappa/\beta_1 g l^3$, and concentration with $-\nu\kappa k_T / T \beta_1 g l^3$, and we shall choose units of length such that $l = 1$.

We impose the following physically important rigid-rigid, no-flux boundary conditions:

$$w = \partial_z w = \theta = \partial_z \eta = 0 \quad \text{at } z = \pm \frac{1}{2}. \quad (2)$$

The even solution of Eqs. (1) will in general have a form

$$\begin{pmatrix} w(t, \mathbf{x}, z) \\ \theta(t, \mathbf{x}, z) \\ \eta(t, \mathbf{x}, z) \end{pmatrix} = \sum_{\mathbf{k}} e^{\sigma t + i\mathbf{k} \cdot \mathbf{x}} \sum_{i=1}^4 \begin{pmatrix} w_i^0 \\ \theta_i^0 \\ \eta_i^0 \end{pmatrix} A_i \cosh(q_i z) + \text{c.c.} \quad (3)$$

where \mathbf{x} is the location in the plane perpendicular to the temperature gradient and q_i are the roots of the polynomial:

$$\begin{aligned}\tilde{\Delta}(\sigma/P - \tilde{\Delta})(\sigma - \mathcal{L}\tilde{\Delta})(\sigma - \tilde{\Delta}) \\ + k^2 R [(1 + \Psi)(\sigma - \mathcal{L}\tilde{\Delta}) - \Psi\tilde{\Delta}] = 0,\end{aligned}\quad (4)$$

with $\tilde{\Delta} \equiv k^2 - q^2$. It is clear from inspection of Eq. (4) that for stationary convection the critical values for R and k depend only on $\Psi(1 + 1/\mathcal{L})$ and not on Ψ and \mathcal{L} separately. It is easily checked that the even solutions give the lowest value for the Rayleigh number R , so consequently we can discard the odd solutions in Eq. (3), since the two sets separate. Inserting solutions (3) into the boundary conditions (2) one finds a set of four algebraic equations for the amplitudes A_i ($i = 1, \dots, 4$). The coefficients of these amplitudes form a matrix \mathcal{M} and the solvability condition requires that $\det \mathcal{M} = 0$. This condition together with Eq. (4) forms a set of two implicit equations for R , k , σ , and q_i . For given values of Ψ , \mathcal{L} , and P the value of R must be minimized with respect to σ and k . We shall consider two cases: $\sigma = 0$ and $\sigma = i\omega$. The first case corresponds to the stationary bifurcation and was solved previously. The second case corresponds to the oscillatory bifurcation. To determine the solution in this case we minimize with respect to ω for fixed k and repeat this procedure for a grid of k values in order to reach the minimum value for R .

In order to avoid singularities in the limit $\omega \rightarrow 0$ and $\Psi \rightarrow \Psi_{CT}$ it turns out to be convenient to choose $\eta_i^0 = 1$ ($i = 1, \dots, 4$) in Eq. (3). In this case, the elements m_{ij} of \mathcal{M} have the following form:

$$\begin{aligned}m_{1i} &= q_i \sinh(q_i/2), \\ m_{2i} &= \frac{i\omega + \mathcal{L}\tilde{\Delta}_i}{\tilde{\Delta}_i} \cosh(q_i/2), \\ m_{3i} &= \frac{(i\omega + \tilde{\Delta}_i)(i\omega + \mathcal{L}\tilde{\Delta}_i)}{R\tilde{\Delta}_i} \cosh(q_i/2), \\ m_{4i} &= \frac{(i\omega + \tilde{\Delta}_i)(i\omega + \mathcal{L}\tilde{\Delta}_i)q_i}{2R\tilde{\Delta}_i} \sinh(q_i/2),\end{aligned}\quad (5)$$

where $\tilde{\Delta}_i \equiv k^2 - q_i^2$ and $i = 1, \dots, 4$. For $\omega \neq 0$, Eq. (4) has four different solutions for q_i^2 . It has been previously shown that in the case of a stationary bifurcation one of the roots (say, q_4^2) is equal to k^2 , which yields $\tilde{\Delta}_4 \equiv 0$. Therefore, special attention is required in taking the limit $\omega \rightarrow 0$ in m_{24} in Eq. (5). The correction to q_4^2 due to small ω can be calculated from Eq. (4) yielding

$$q_4^2 = k^2 - i\omega \frac{1 + \Psi}{\mathcal{L} + \mathcal{L}\Psi + \Psi} + O(\omega^2). \quad (6)$$

Using Eq. (6), one finds that

$$m_{24} \rightarrow -\frac{\Psi}{1 + \Psi} \cosh(k/2) \quad \text{as } \omega \rightarrow 0, \quad (7)$$

in agreement with previous results for the stationary bifurcation.

III. RESULTS AND DISCUSSION

Equation (4), together with the condition $\det \mathcal{M} = 0$, was solved numerically for R , ω , and k as a function of Ψ for two choices of the Lewis and Prandtl numbers. Namely, for $\mathcal{L} = 0.02$, $P = 17$ (corresponding to ethanol-water mixtures at room temperature) and for $\mathcal{L} = 0.04$, $P = 0.75$ (a typical value for ^3He - ^4He mixtures). The values of R for the oscillatory bifurcation are shown in Figs. 1(a) and 1(b). In these figures the lowest curve represents the exact solution. The two other curves were obtained by performing the stability analysis using the Galerkin method with two sets of trial functions $(z^2 - \frac{1}{4})^2$ and $\cos^2(\pi z)$ for the vertical velocity w and for the mass flux, and $z^2 - \frac{1}{4}$ and $\cos(\pi z)$ for the temperature, respectively. Clearly the trial functions $(z^2 - \frac{1}{4})^2$ and $z^2 - \frac{1}{4}$ yields a better approximation. It deviates from the exact solution by at most 3%. We note that the calculations on the oscillatory branch done previously (see Refs. 1 and 3) used trigonometric functions as trial functions. The exact values of R for both, ethanol-water and ^3He - ^4He mixtures, for the stationary bifurcation are shown in Fig. 1(c). For both choices of \mathcal{L} and P the values of ω following from the trial functions are nearly identical to the exact solution for small ω , and have slightly lower values (no more than 3%) for large ω [see Figs. 2(a) and 2(b)]. For both choices of \mathcal{L} and P the values of the wave vector k for the oscillatory bifurcation are close to π and vary little as a function of Ψ . For room-temperature mixtures

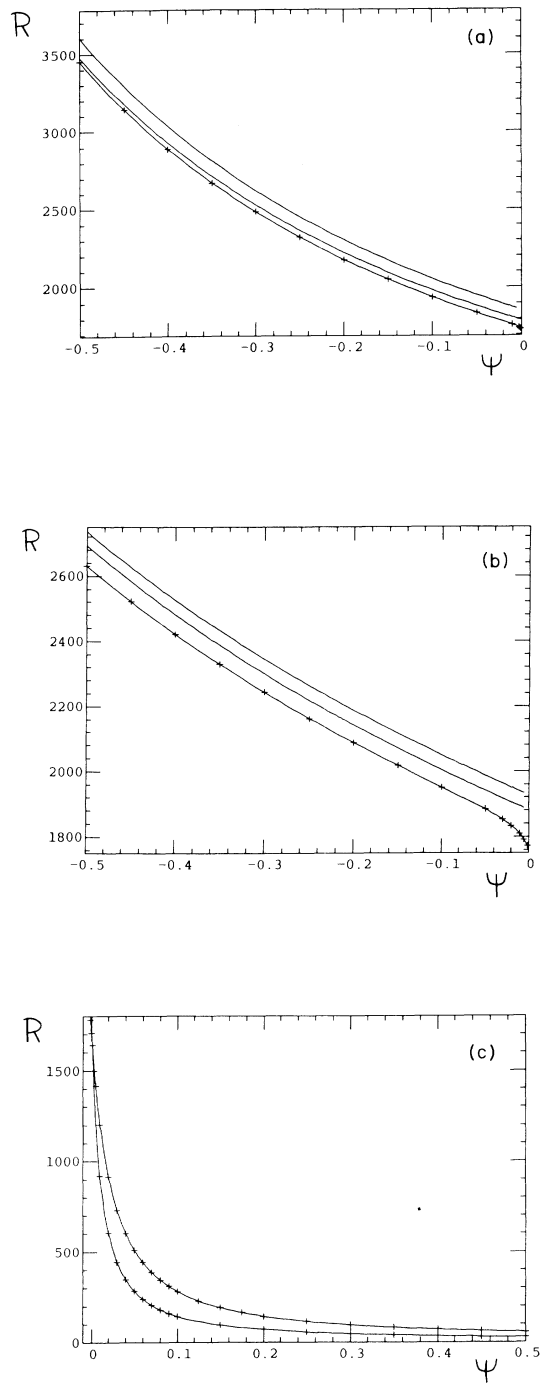


FIG. 1. Rayleigh number R as a function of separation ratio Ψ . (a) The oscillatory branch for ethanol-water mixtures. The lowest curve corresponds to the exact solution. The curve in the center is obtained using the trial function $(z^2 - \frac{1}{4})^2$ for the vertical velocity w , and the highest curve using the trial function $\cos^2(\pi z)$ for w . (b) The oscillatory branch for ${}^3\text{He}$ - ${}^4\text{He}$ mixtures. Here the curves from the lowest to the highest have the same meaning as in (a). (c) Exact results for the stationary branch. The upper curve corresponds to ${}^3\text{He}$ - ${}^4\text{He}$ mixtures, the lower one to ethanol-water mixtures.

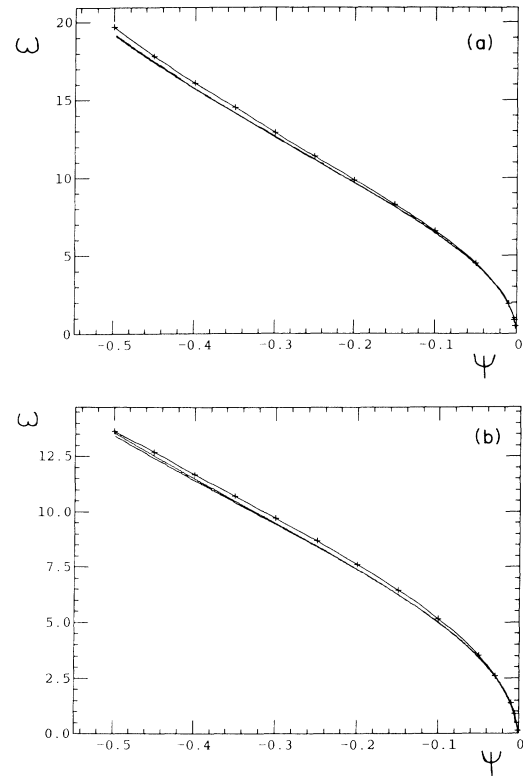


FIG. 2. Neutral frequency ω as a function of the separation ratio Ψ for (a) ethanol-water mixtures, (b) ${}^3\text{He}$ - ${}^4\text{He}$ mixtures. In both cases the highest curve corresponds to the exact solution. The two lower curves are obtained using the test functions quoted in the text. The latter two curves nearly coincide.

($\mathcal{L}=0.02$ and $P=17$) k varies from 3.12, for Ψ close to zero, to 3.13, for $\Psi=-0.5$. The trial function gives the range 3.12–3.14 in the case of $(z^2 - \frac{1}{4})^2$ and 3.11–3.13 if $\cos^2(\pi z)$ is used. For the helium mixtures ($\mathcal{L}=0.04$ and $P=0.75$), k varies from 3.15 to 3.22 for the same range of Ψ , while the trial functions yield values from 3.10 to 3.26 for $(z^2 - \frac{1}{4})^2$ and from 3.08 to 3.24 for $\cos^2(\pi z)$. These small variations in k are unlikely to be of practical importance in a finite container because the vertical boundaries have dominating influence on imposing a value for the wave vector. For practical purposes it can be therefore assumed that on the oscillatory branch $k \simeq \pi$. On the stationary branch k decreases sharply with increasing Ψ and reaches zero at $\Psi = \Psi_s$. The value of Ψ_s depends on the Lewis number. We found that for $\mathcal{L}=0.02$ (room-temperature mixtures), $\Psi_s=0.2$, and for $\mathcal{L}=0.04$ (helium mixtures), $\Psi_s=0.09$. The plot of k as a function of Ψ for both mixtures is shown in Fig. 3.

From the previous discussion we conclude that the trial function $(z^2 - \frac{1}{4})^2$ for w and η , and $z^2 - \frac{1}{4}$ for θ , are a good approximation to the exact solution for the oscillatory bifurcation. In particular, it can be used for quick and reliable estimates of R and ω for various binary mixtures.

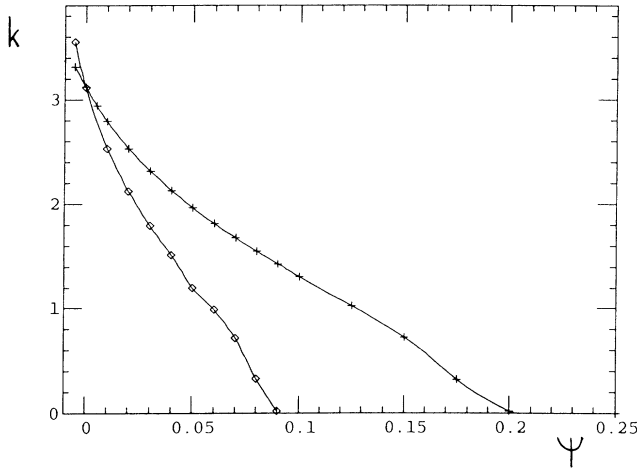


FIG. 3. The critical wave vector on the stationary branch. The diamonds correspond to ethanol-water mixtures and the crosses to ${}^3\text{He}$ - ${}^4\text{He}$ mixtures.

The relevant expressions can be easily obtained from Eqs. (1):

$$R(k) = \frac{\beta\tilde{\gamma} - \alpha\tilde{\delta}}{\alpha\delta_R - \beta\gamma_R}, \quad (8)$$

$$\omega^2(k) = \frac{\tilde{\delta} + R\delta_R}{\beta} = \frac{\tilde{\gamma} + R\gamma_R}{\alpha},$$

where

$$\begin{aligned} \alpha &= 0.1P^{-1}b_1, \\ \beta &= 12.6(\mathcal{L}/P)b_1^2 + (1/P)b_1b_2 + 0.1b_3, \\ \tilde{\gamma} &= 126(\mathcal{L}/P)b_1^2b_2 + 12.6\mathcal{L}b_1b_3 + b_2b_3, \\ \gamma_R &= -\frac{3}{3920}k^2(1+\Psi), \\ \tilde{\delta} &= 126\mathcal{L}b_1b_2b_3, \\ \delta_R &= -\frac{27}{280}b_1k^2(1+\Psi)\mathcal{L} - \frac{3}{140}k^2\Psi(k^2/28 + \frac{1}{3}), \end{aligned}$$

and

$$\begin{aligned} b_1 &= (k^2/126 + 2/21), \quad b_2 = (1 + k^2/10), \\ b_3 &= (4 + 4k^2/21 + k^4/126). \end{aligned}$$

The condition of minimum R determines the value of the wave vector k in Eq. (8).

It has been found for both choices of \mathcal{L} and P that $\omega^2 \sim \Psi$ for small ω , similar to the case of free-free, impervious boundary conditions. A careful extrapolation of this result to $\omega=0$ yields the value of Ψ at the codimension-2 point. In both cases we found that $\Psi_{\text{CT}} < 0$, but it is shifted towards $\Psi=0$ as compared to the free-free case. For $\mathcal{L}=0.02$ and $P=17$ the exact solution yields $\Psi = -1.5 \times 10^{-4}$ ($R_{\text{CT}}=1726.9$ and $k_{\text{CT}}=3.12$), while $\Psi_{\text{CT}} = -4.15 \times 10^{-4}$ for free-free boundary conditions. For $\mathcal{L}=0.04$ and $P=0.75$ we found $\Psi_{\text{CT}} = -9.0 \times 10^{-4}$ ($R_{\text{CT}}=1771.2$ and $k_{\text{CT}}=3.15$), as compared with $\Psi_{\text{CT}} = -3.40 \times 10^{-3}$ for free-free boun-

dary conditions. Even though the shift in Ψ_{CT} is substantial in absolute terms it is much smaller than the present experimental resolution in Ψ . Therefore it is not expected to be detected experimentally in the near future.

IV. CONCLUSIONS

In the present paper we have calculated exactly for a laterally infinite layer of a binary fluid mixture the critical temperature difference and the critical frequency as a function of the separation ratio for various practically important values of the Prandtl and Lewis number. We find that on the oscillatory branch the critical transverse wave vector at onset is nearly independent of the separation ratio and that the system tends to form circular rolls. This is in strong contrast to the results for the stationary branch for which one expects above a critical value for the separation ratio ($\Psi_s=0.20$ for ${}^3\text{He}$ - ${}^4\text{He}$ mixtures and $\Psi_s=0.09$ for ethanol-water mixtures) one big roll to appear and to fill the whole container. This prediction seems to be in accord with very recent experimental observations¹⁵ which indeed show one big roll at onset. We have also investigated to what extent the exact critical Rayleigh number and critical frequency can be approximated by simple test functions and we find that, taking for the velocity field the profile $(z^2 - \frac{1}{4})^2$, we can easily obtain an accuracy of better than 3%. In addition, we have evaluated the shift in the location of the codimension-2 point and we have found this shift to be too small to be detected experimentally given the present experimental resolution in the determination of the separation ratio. It is important to note, however, that the codimension-2 value always appears at slightly negative values of Ψ .

We would like to add a word of caution here, however. The values for the critical Rayleigh number and the frequency can only be expected to be practically relevant if the transition to the first convective pattern is continuous¹⁰ (second-order-type nonequilibrium phase transition) or very weakly first order¹⁴ (small hysteresis). The present analysis is not applicable if the transition to the convective structure observed is strongly first order¹² and associated with a large hysteresis as it is the case, e.g., for ethanol-water mixtures for $\Psi = -0.5$.

Note added in proof. After submitting this paper for publication, we received a copy of a report by S. J. Linz and M. Lücke (unpublished) reporting a jump in wave number and in frequency at the codimension-2 point using an eight-mode truncation of the basic equations.

From our analysis it follows that there is no jump in the wave vector nor frequency at the codimension-2 point in the exact solution. On the contrary, taking as an example normal fluid ${}^3\text{He}$ - ${}^4\text{He}$ mixtures with $\mathcal{L}=0.04$ and $P=0.75$ we obtain for the test functions proportional to $\sin^2(\pi Z)$ and $(Z^2 - \frac{1}{4})^2$ for the vertical velocity field the following result. At the codimension-2 point we get a jump in wave vector of 1.42% for $\sin^2(\pi Z)$ and 0.31% for $(Z^2 - \frac{1}{4})^2$, respectively. For the frequency we obtain 0.045 in the former and 0.03 in the latter case. As has been discussed above the test functions $(Z^2 - \frac{1}{4})^2$ yield a better approximation to the exact solution and we find,

correspondingly, that the jumps in the wave vector and frequency are smaller.

To demonstrate that there is no jump in wave number and in frequency in the exact solution at the codimension-2 point we have repeated the *exact* calculations reported in the main part of the paper with higher accuracy. We find no jump, neither in the wave vector nor in the frequency. For the case of ${}^3\text{He}$ - ${}^4\text{He}$ mixtures we find at the codimension-2 point

$$\psi_{\text{CT}} = -8.97 \times 10^{-4} \pm 0.02 ,$$

$$R_{\text{CT}} = 1771.06 \pm 0.01 ,$$

and

$$k_{\text{CT}} = 3.150 \pm 0.001 .$$

Therefore we conclude that the exact solutions both for the free-free, impervious case and the rigid-rigid, no-flux boundary conditions give a codimension-2 point of the same type. On the other hand it seems clear from the above results that any finite truncation of the basic equations will give a jump in k and ω .

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*Address after 1 October 1986: Département de Physique, Université de Nice, Parc Valrose, 06034 Nice Cedex, France.

†Address after 1 October 1986: Fachbereich Physik, Universität Essen, D-4300 Essen 1, West Germany.

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