Linear stability of experimental Soret convection

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Linear stability results for convection in binary fluid mixtures are given for experimental boundary conditions and parameter values. Normal $^3$He-$^4$He and ethanol-water mixtures are considered with no-slip boundary conditions on the velocity and no outward mass flux. Two cases are considered in detail: fixed temperature at top and bottom, and fixed temperature at the top and fixed thermal flux at the bottom. The role played by the Biot number is emphasized. The results are presented in a manner most useful to experimentalists. The errors incurred in determining the separation ratio $S$ from experimental observations using scaled results with free boundaries are quantified. Neutral curves for both oscillatory and steady-state instabilities together with the corresponding critical wave numbers $k_c$ are determined as functions of $S$. For the latter, $k_c$ vanishes as $S$ increases. The critical values of $S$ for which this first occurs are determined analytically. Growth rates and frequencies for supercritical Rayleigh numbers are also given. The codimension-two point is found to be masked by steady-state instabilities that set in at smaller Rayleigh numbers, but the masking vanishes in the limit of small Lewis numbers and large Prandtl numbers. The lowest observable frequency along the Hopf curve is determined.

I. INTRODUCTION

Binary fluid mixtures provide a convenient and easily controllable continuous system exhibiting a variety of pattern formation phenomena when driven out of equilibrium by thermal forcing. In contrast to other pattern-forming systems, such as nematics, binary fluids offer several advantages. They are described by known equations with a relatively small number of controllable parameters. In addition, the physics of the convective instability is well understood. It is therefore natural that these systems have attracted much interest among both experimenters and theorists interested in pattern formation. The systems are characterized by a dimensionless separation ratio $S$, proportional to the Soret coefficient. When $S$ is positive the heavier component migrates towards the colder wall, while the opposite is the case when $S < 0$. Consequently, Soret diffusion promotes instability in a fluid heated from below when $S > 0$, and hinders it when $S < 0$. In the former case the instability that sets in is exponentially growing; in the latter case it can be oscillatory. Thus binary fluid mixtures can exhibit pattern formation arising from both steady-state and Hopf bifurcations.

In addition to the separation ratio, the system is specified by the Rayleigh number $R$ measuring the thermal driving, and by the Lewis and Prandtl numbers, denoted by $\tau$ and $\sigma$, describing, respectively, the solute and momentum diffusivities relative to the thermal diffusivity. In the experiments $R$ is determined externally from the measured temperature difference across the system, while the temperature and concentration dependence of the parameters $\tau$ and $\sigma$ is reasonably well known. The separation ratio $S$, however, is typically unknown although it can be varied widely not only by changing the solute concentration but also by changing the mean temperature of the system. Consequently binary fluid mixtures provide an ideal system in which a two-dimensional parameter space can be explored experimentally. This is one of the main reasons why so much interesting nonlinear behavior has been discovered in these systems.

In spite of the recent experimental and theoretical activity detailed linear stability results for the experimental boundary conditions and parameter values remain unavailable. Instead the procedure that has generally been adopted is to use results for the critical Rayleigh number for stress-free boundary conditions and fixed temperature and concentration at the top and bottom, and then to "correct" for the fact that the experiments demand no-slip boundary conditions with no outward mass flux. In practice this correction is accomplished by scaling the analytical critical Rayleigh number for the simple boundary conditions by 2.597. This number is the ratio, for fixed temperature at the top and bottom, of the critical Rayleigh numbers for the onset of convection in a pure fluid with no-slip and with stress-free boundary conditions. For oscillatory convection with fixed temperatures at the top and bottom and moderate values of $S$ this is known to yield results that are correct to 4–5%. However, we consider this procedure to be rather unreliable in general since (a) it assumes that the parameter dependence in the realistic case remains identical to that with the simple boundary conditions, and (b) it ignores the fact that for the cryogenic experiments imposed heat flux at the bottom may be a more appropriate
boundary condition. Naively this should change the scaling factor to 1.971. This is the ratio of the critical Rayleigh numbers for the onset of convection in a pure fluid with no-slip boundaries, fixed temperature at the top and fixed heat flux at the bottom, and convection with stress-free boundaries and fixed temperature on the boundaries.

If the separation ratio \( S \) were easily measurable the validity of the above extrapolation could be checked against the experiments. However, since this is often not possible the corrected Rayleigh numbers for oscillatory convection are actually used to deduce the value of \( S \) by fitting to the experimentally observed critical Rayleigh numbers. The possible error in the determination of \( S \) that this procedure introduces becomes particularly important for modeling the nonlinear evolution of the instability. The purpose of the present paper is therefore to provide a detailed description of the linear stability properties of the pure conduction state for the experimental boundary conditions and parameter values which could be used to provide a more reliable determination of the separation ratio \( S \). Even so we are forced to make the following simplification. We assume that the layer is unbounded in both horizontal directions, and therefore do not include the effects of sidewalls. Thus our results should be applicable to large-aspect-ratio systems only.

Study of convection in binary fluid mixtures has a long history. The first detailed studies were given in the context of thermohaline convection by Veronis\(^1\) and Baines and Gill\(^2\) for the simple boundary conditions, and by Nield\(^3\) for other types of boundary conditions. In these problems the concentration gradient is imposed externally by the boundary conditions rather than forming in response to the applied thermal gradient through the Soret effect. The two problems are, however, equivalent in the sense that there exists a linear transformation of one into the other.\(^4\) References 2 and 3 serve as a model for the present work, in that we extend the type of analysis and results presented in the former to the experimental boundary conditions, and use the technique described in the latter to derive various analytical results concerning the onset of stationary instability. Convection in binary fluid mixtures driven by the Soret effect was first studied for realistic boundary conditions by Hurle and Jakeman,\(^5\) as well as several times subsequently.\(^6\)–\(^8\) None of these papers can be used, however, to determine the separation ratio \( S \) from the experimental observations. In addition the techniques used by these authors (expansion in trigonometric functions in the vertical in Ref. 5, variational methods in Refs. 6 and 7) have to be used with considerable care for boundary-value problems of this type. This is because the linear eigenfunctions need not have straight or vertical boundaries (see Sec. III below). For this reason we adopt the normal-mode approach of Ref. 8.

In the paper we present largely numerical results for two sets of boundary conditions of experimental interest; both have no-slip boundaries for the velocity field, no mass flux through them and a fixed temperature at the top. They differ in the choice of the temperature boundary condition on the bottom boundary, which can be either fixed temperature or fixed heat flux. The latter boundary condition is not considered in any of the existing literature and yet is relevant to some of the recent experiments. Throughout we make every attempt to present the results in a manner that we hope will be found useful by the experimenters, and consider parameter values that bracket the experimental values. The method of analysis is described in Sec. II followed by the results for the two types of boundary conditions in Sec. III. These are compared not only to each other, but also to those with the simple boundary conditions in order to highlight the source of error that can result from the use of incorrect boundary conditions. A brief summary and conclusions follow in Sec. IV.

II. LINEAR THEORY

In this section we formulate the mathematical stability problem and describe the technique we use to solve it. Since we are considering a horizontally unbounded system, we restrict attention without loss of generality to two-dimensional disturbances. Then the linear stability properties of the conduction state (\( \Psi = \Theta = \Sigma = 0 \)) are described by the equations\(^9\):

\[
\frac{1}{\sigma} \partial_t \nabla^2 \Psi - \nabla^4 \Psi - R \partial_x \Theta - SR \partial_x \Sigma = 0 ,
\]

\[
\partial_t \Theta - \partial_x \Psi - \nabla^2 \Theta = 0, \tag{2.1b}
\]

\[
\frac{1}{\tau} \partial_t \Sigma - \frac{1}{\tau} \partial_x \Sigma - \nabla^2 \Psi + \nabla^2 \Theta = 0, \tag{2.1c}
\]

where \( \Psi \) is the stream function, and \( \Theta \) and \( \Sigma \) denote departures from the linear temperature and concentration profiles present in the absence of convection. With appropriate boundary conditions Eqs. (2.1) constitute an eigenvalue problem for the critical Rayleigh number \( R_c \). These are no-slip conditions on the velocity, and no outward mass flux on the concentration. The thermal boundary conditions vary from experiment to experiment. Two types of experiments are performed: those for which both the top and the bottom plates confining the fluid are in contact with heat baths, the lower surface being maintained at a higher temperature, and those in which the top plate is maintained at constant temperature, with the lower plate heated at constant electrical power. Before the stability properties of the resulting temperature profile can be discussed the experimental setup has to be translated into boundary conditions on the temperature perturbation. If the boundaries are excellent conductors and have in addition a higher thermal mass compared to the fluid, the temperature remains constant on the boundaries, and hence the temperature perturbation \( \Theta \) vanishes on \( z = 0, 1 \). This is true regardless of whether the bottom boundary is maintained at fixed temperature or is heated with constant current. These appear to be the appropriate boundary conditions for the room-temperature experiments on salt-water\(^10\) and ethanol-water\(^11\)–\(^18\) mixtures. Under these conditions convection in a pure fluid sets in at the critical Rayleigh number \( R_c = 1707.762 \). In the cryogenic experiments,\(^20\) however, the situation can be quite
different. Here, even if the boundaries are made of a
good conductor, their thermal capacity will nearly al-
ways be less than that of the fluid, and hence the
constant-temperature boundary condition applies at best
on the upper surface which is in contact with a
constant-temperature heat bath. Heating the lower sur-
face at constant power supplies a constant heat flux, and
this translates to a boundary condition that is better ap-
proximated by \( \partial \Theta / \partial z = 0 \) on \( z = 0 \). In all the ex-
periments the appropriate thermal boundary conditions are
determined by the value of the Biot number \( B \). If \( B \gg 1 \)
the boundary condition \( \Theta = 0 \) is appropriate, while if
\( B \ll 1 \) the condition \( \partial \Theta / \partial z = 0 \) becomes correct. In
general, the linearized boundary conditions take the form

\[
\frac{\partial \Theta}{\partial z} + B \Theta = 0 \, .
\] (2.2)

Since the values of \( B \) are not specified for any of the ex-
periments reported in the literature, we present below re-
results for both \( \Theta = 0 \) on \( z = 0,1 \), and for \( \Theta = 0 \) on \( z = 1 \)
and \( \partial \Theta / \partial z = 0 \) on \( z = 0 \), bracketing the range of pos-
sibilities. It should be mentioned that in the latter case the
critical Rayleigh number for a pure fluid is
\( R_c = 1295.778 \)\(^{,22} \). Values in this range have been re-
ported for pure \( ^4 \)He in Refs. 23. In addition, Sparrow et al.\(^{,22} \)
explore in detail the Biot number dependence of
\( R_c \) for a pure fluid in both cases of interest in this paper,
\( \Theta = 0 \) on \( z = 0 \), and \( \partial \Theta / \partial z = 0 \) on \( z = 0 \), providing a
means for extrapolating the results reported below to in-
termediate Biot numbers.

In the following we study Eqs. (2.1) with the following
sets of boundary conditions:

\[
\begin{bmatrix}
D^2 - k^2 - i \frac{\omega}{\sigma} & -R & -SR \\
0 & D^2 - k^2 - i \omega & 0 \\
\frac{k^2}{\tau} & -D^2(k^2) & D^2 - k^2 - i \frac{\omega}{\tau}
\end{bmatrix}
\]

and \( D \) denotes \( d/dz \). Equation (2.8) is an eighth order
boundary-value problem with constant coefficients. We
look for solutions proportional to \( \exp \lambda z \), and find that
such solutions exist provided

\[
\rho^4 - i \rho \omega^3 \left[ 1 + \frac{1}{\sigma} + \frac{1}{\tau} \right] - \rho^2 \omega^2 \left[ \frac{1}{\sigma} + \frac{1}{\tau} + \frac{1}{\sigma \tau} \right] + \rho \left[ \frac{\omega^3}{\sigma \tau} + R k^2 \left( 1 + S + \frac{S}{\tau} \right) \right] - i \frac{\omega^3}{\tau} R k^2 (1 + S) = 0 \, .
\] (2.9)

where \( \rho \equiv \lambda^2 - k^2 \). Equation (2.9) determines eight roots
\( \lambda_j \), so that the general solution is

\[
f(z) = \sum_{j=1}^{4} a_j^+ e^{\lambda_j z} + a_j^- e^{-\lambda_j z} \, .
\] (2.10)
(2.12) may be solved for \( \omega = \Omega + i\gamma \), i.e., the frequency \( \Omega \) and the growth rate \( \gamma \) of the instability. Equation (2.12) can also be used to determine the neutral curve, by imposing the condition \( \gamma = 0 \) and solving for \( \Omega \) and \( R \) instead. We denote these critical values by \( \Omega_c \) and \( R_c \). The quantity \( R_c \) is a function of \( k^2 \) and may be minimized with respect to \( k^2 \) to determine \( R_c(k_c) \), the Rayleigh number at which the instability first sets in, and the wave number \( k_c \) with which it does so. When \( \Omega = 0 \) the determinant becomes real and the same procedure can be used to find \( R_c(k_c) \) for the onset of steady convection, or to find the growth rate \( \gamma \) for a given value of \( R \) and \( k \). Finally, Eq. (2.12) can be solved for the coefficients \( a_{i2}^I \) at \( R = R_c \) and \( k = k_c \) and these used to construct the eigenfunctions. Eigenfunctions in the form of both traveling waves and standing waves have been computed and reveal several unexpected features that have a bearing on the experiments.

For the values of \( \sigma, \tau \), and \( S \) used in the experiments, the condition number\(^{25} \) of the matrix \( M \) is very large. This obliged us to resort to a 128-bit representation of the numbers to maintain accuracy of the results presented below.

**III. RESULTS**

(i) Case (C). We have investigated in detail the solutions to the problem (2.1) with the boundary conditions (C). These boundary conditions apply to many of the laboratory experiments. These include experiments on normal \(^3\)He-\(^4\)He mixtures,\(^8\) salt-water mixtures,\(^10\) and ethanol-water mixtures\(^11\) between plates maintained at constant temperatures\(^{26} \) as well as several room-temperature experiments on ethanol-water mixtures in which the lower plate is heated at constant electrical power.\(^{12-16} \) For the latter experiments the Biot number is presumably sufficiently large that the boundary conditions (C) are more appropriate than (D). For the cryogenic experiments we choose \( \tau = 0.03 \), \( \sigma = 0.6 \) as typical parameter values;\(^20\) the Dufour effect which may be important under some conditions is not included.\(^8\) The ethanol-water experiments cover the range \( \tau = 0.005 \), \( \sigma = 14.9 \) (Ref. 11); \( \tau = 0.0068 \), \( \sigma = 9.7 \) (Ref. 11); \( \tau = 0.015 \), \( \sigma = 18.4 \) (Ref. 18); and \( \tau = 0.02 \), \( \sigma = 17.0 \) (Ref. 15).

The linear-theory stream function \( \Psi \) is shown in Fig. 1 for each of the three cases: (a) steady state (SS), (b) left-traveling wave (LTW), and (c) standing wave (SW) and representative choices of parameter values. For SS the matrix \( M \) is real, and the solution takes, without loss of generality, the form

\[
\Psi^\mathrm{SS} = kf(z) \sin(kx), \tag{3.1}
\]

where \( f(z) \) is real. The cell boundary is vertical, and the streamlines are symmetric about the cell midplane, as shown in Fig. 1(a). In contrast, at the Hopf bifurcation we can get an arbitrary superposition of left- and right-traveling waves, of which a pure LTW is given by

\[
\Psi^\mathrm{LTW} = - \Re\{ikf(z)e^{ikx+iot}\} = k[f_R(z)\sin(kx+iot)+f_I(z)\cos(kx+iot)]
\]

\[
\equiv A(z)\sin[kx+iot+\theta(z)], \tag{3.2b}
\]

where \( \tan\theta = f_I/f_R \). Thus at any instant in time the cell boundary is given by

\[
kx+iot+\theta(z) = n\pi, \tag{3.3}
\]

and the boundary is not vertical, in contrast with the analytically solvable case (A). Figure 1(b) shows that for \( \tau = 0.03 \), \( \sigma = 0.6 \) the boundary is bowed symmetrically towards the right; thus the cell leads at the top and bottom, and lags in the middle. Remarkably, for \( \tau = 0.005 \), \( \sigma = 14.9 \) the boundary bows in the opposite direction, and the cell leads in the middle and lags at the top and bottom. Finally, a SW is the superposition of two oppositely traveling waves of equal amplitude, and is given by

\[
\Psi^\mathrm{SW} = \Re\{ikf(z)e^{iot}\} \sin(kx)
\]

\[
\equiv A(z) \sin[iot+\theta(z)] \sin(kx), \tag{3.4b}
\]

for a suitably chosen origin in \( x \). Such a solution has vertical cell boundaries [see Fig. 1(c)], but owing to the \( z \)-dependent phase \( \theta \), there is no instant in time at which the cell is completely at rest. This is again in contrast to case (A). This aspect of the solution is illustrated in Fig. 1(d) showing the root-mean-square velocity \( v_{\mathrm{RMS}} \) of a
FIG. 2. Neutral stability curves $R_c(k)$ for a $^3$He-$^4$He mixture with boundary conditions (C) for (a) the Hopf bifurcation, and (b) the steady-state bifurcation. The dashed lines indicate the values of $k_c$ which minimize $R_c(k)$ for each value of $S$. Note that $k_c$ vanishes when $S \geq 0.130$.

Cell as a function of the time $t$, and in Fig. 1(c) showing the streamlines near a minimum in $u_{\text{RMS}}$. Note that the cell appears divided, temporarily, into three cells, stacked vertically, with a countercell in the middle. Neither of these aspects of the linear theory appears to have been noted before.

In Fig. 2 we show the neutral stability curves $R_c(k)$ for $\tau=0.03$, $\sigma=0.6$, as a function of the horizontal wave number $k$. Several different values of $S$ are compared, and results for both oscillatory and steady convection are presented. The Hopf neutral curves do not extend to arbitrary wave numbers but terminate on a SS neutral curve where the oscillation frequency $\Omega_c$ vanishes. The neutral curves have minima at $k=k_c$ which yield the thresholds $R_c(k_c)$ for instability. Observe that for oscillatory convection such minima always occur at finite $k_c$ but that for steady convection $k_c$ may vanish, as explained further below. The neutral curves are useful in that they show how sharp the minima actually are. Figure 3 summarizes most of the results for oscillatory convection that will be of use to the experimenters. For various values of $\sigma$, $\tau$ the figures show, in the $(R,S)$ plane, the neutral stability curve for oscillations, $R_c(k_c)$, denoted by $\gamma=0$. Contours of constant growth rates $\gamma$, in units of the vertical diffusion time, are also shown. These lines terminate on the right along a line labeled $\Omega=0$, where the dominant eigenvalues become real. Contours of constant frequencies $\Omega$ are superposed. The $\gamma$ contours are computed by minimizing $R(k,\gamma,S,\sigma,\tau)$ at fixed $\gamma,S,\sigma,\tau$ with respect to $k$. If $R$ is thought of as a function of $\Omega(k,\gamma,S,\sigma,\tau)$ it is necessary to first eliminate $\Omega$ before proceeding with the minimization. This procedure yields the largest growth rate at each point in the $(R,S)$ plane. The $\Omega$ contours are obtained by splines connecting the corresponding frequencies in this landscape. Using a contour plot of this form observed growth rates and frequencies can be assigned unique $(R,S)$ values. Finally, the different values of $k_c$ along the $\gamma$ contours are indicated by tick marks. Note that in

FIG. 3. Critical Rayleigh numbers as a function of the separation ratio $S$ for the boundary conditions (C) and (a) $\tau=0.03$, $\sigma=0.6$; (b) $\tau=0.02$, $\sigma=10.0$; (c) $\tau=0.005$, $\sigma=14.9$. Shown are the curves of constant growth rate $\gamma$ and constant frequency $\Omega$. The tick marks on the constant $\gamma$ curves indicate the wave numbers $k_c$ in intervals of 0.05 increasing to the left. The horizontal dashed line indicates the result of scaling the $\gamma=0$ curve for boundary conditions (A) so as to coincide with that for the conditions (C) at the codimension-2 point. Note that these curves diverge significantly at larger negative $S$, particularly in (a). The vertical dashed line near the right axis gives the neutral stability curve $(\gamma=0)$ for the steady state.
contrast to the analytically solvable case (A) the critical wave number does depend on the parameters albeit rather weakly. Moreover, its value lies near $k_c = 3.116$ and not near 2.221, the critical wave number for case (A). In view of the use of the scaled results obtained with boundary conditions (A), we have also shown, in a dotted line, the critical Rayleigh number one obtains by multiplying this result by the factor necessary to bring the $\Omega_c(k_c) = 0$ end points of the corresponding neutral curves into coincidence. For $\tau = 0.0005$, $\sigma = 14.9$ this factor is 2.590. On the scale of this plot it is not possible to distinguish this procedure from scaling with the factor 2.597 to bring instead the steady-state critical Rayleigh numbers into coincidence at $S = 0$. One sees that for moderately large negative $S$ values the discrepancy can be quite large. For example, for $\tau = 0.03$, $\sigma = 0.6$, $S = -0.6$ [see Fig. 3(a)] the discrepancy is 16.0%, although for $\tau = 0.005$, $\sigma = 14.9$, $S = -0.6$ [see Fig. 3(c)] it is only 6.6%; moreover the discrepancy decreases for less-negative $S$. Similar neutral stability curves have been obtained in experiments on water-alcohol mixtures,\(^5\) normal $^3$He-$^4$He mixtures,\(^6\) as well as salt-water mixtures.\(^10\)

Because of the scale used in Fig. 3 the neutral curve for steady convection is very close to the $S = 0$ axis. Since the details of the behavior near $S = 0$ are important we show in Fig. 4 an enlargement of this region. Observe that the Hopf neutral curve crosses the steady-state neutral curve terminating at a point where the frequency $\Omega_c(k_c)$ of the neutrally stable oscillations vanishes. We call this point the codimension-2 (CT) point, and denote the corresponding values of $R$, $S$, and $k$ by the subscript CT. The CT point lies on the steady-state neutral curve with fixed $k = k_{CT}$, indicated by a broken line in Fig. 4(a). Since this value of $k$ does not minimize the Rayleigh number for the onset of steady convection the curve $R_{\text{SS}}(k_{CT})$ lies above the curve $R_{\text{SS}}(k_c)$. Thus if the system is allowed to select its wave number, the CT point will not be observable (cf. Ref. 24). Instead, as one traverses the Hopf curve in the direction of increasing $S$, the frequency $\Omega_c(k_c)$ decreases, with oscillations superseded by steady convection when $\Omega_c(k_c)$ is still finite. Thus, in spite of its importance\(^28\) for theories of the nonlinear evolution of the instabilities, the signature of the CT point, the vanishing of the frequency $\Omega_c(k_c)$, will not be observable in experiments, although the minimum frequency $\Omega_{\text{min}}$ along the Hopf neutral curve can get very small.\(^28\) Indeed, only in one small-aspect-ratio experiment, in which the wave number is to a large extent "quantized" by the sidewalls, has the CT point been apparently observed.\(^29\) Note that unlike the frequency which decreases monotonically along the Hopf

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**FIG. 5.** Location of the codimension-2 point for boundary conditions (C) and small values of $\tau$. Shown are (a) $R_{\text{CT}}$, (b) $(-S_{\text{CT}})^{1/2}$, and (c) $k_{\text{CT}}$ relative to the pure fluid values for three values of $\sigma$. Note the approximately linear scaling with $\tau$.

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**FIG. 6.** Shielding of the codimension-2 point for boundary conditions (C). Shown are (a) $(-\Delta R_{\text{CT}})^{1/2}$, (b) $\Delta k_{\text{CT}}$, (c) $\Omega_{\text{min}}$, and (d) $S_{\text{min}}$ as functions of $\tau$ for three values of $\sigma$. 

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**FIG. 4.** (a) An enlargement of the $R$-$S$ plane near the codimension-2 point in a $^3$He-$^4$He mixture with the boundary conditions (C). The curve $R_c(k_c)$ of Hopf bifurcations terminates at the point labeled CT where $\Omega$ vanishes and $k_c = 3.0754$. This point lies on the neutral curve $R_c(k)$ for steady-state bifurcations. However, $R_c(k_c)$ for steady-state bifurcations lies to the left, shielding the CT point. The tick marks indicate values of $k$; on this curve, with $k_c$ increasing in units of 0.0001 to the left. (b) shows the variation of $k_c$ as a function of $S$.
curve, the critical wave number \( k_c \) has a minimum near \( k_{CT} \), as shown in Fig. 4(b). In Figs. 5(a)–5(c) we present graphs of \( R_{CT} - R_{RB} \), \( -S_{CT} \), and \( k_{RB} - k_{CT} \) as functions of \( \tau \) for \( \sigma = 10, 0.1, \) and 0.01, where the subscript \( RB \) denotes the pure fluid values (\( \tau = 0, S = 0 \));
\[
R_{RB} = 1707.762, \ k_{RB} = 3.116.
\]
All these curves are approximately linear for small values of \( \tau \) and approach zero as \( \tau \) vanishes; in all cases the \( \tau \) dependence is strong for small \( \sigma \), becoming weaker with increasing \( \sigma \). If \( k \) is kept fixed at \( k_{RB} \), the curves are even closer to being linear, in agreement with the scaling \( R_{CT} - R_{RB} \propto \tau \) and \( S_{CT} \propto -\tau^2 \) suggested in Ref. 30. In Figs. 6(a) and 6(b) we show \(( -\Delta R_{CT} )^{1/2} \), where \( \Delta R_{CT} = R_{CT} - R_{CS}(S_{CT}) \), and \( \Delta k_{CT} = k_{CT} - k_{CS}(S_{CT}) \), with \( R_{CS} \) denoting the critical steady-state Rayleigh number and \( k_{CS} \) the corresponding minimizing wave number. The results show that, for a wide range of \( \sigma \), these differences vanish linearly as \( \tau \to 0 \). For small \( \sigma \), of order \( 10^{-1} \), \( \Delta R_{CT} \) and \( \Delta k_{CT} \) approach 30–40\%, though they decrease continuously with increasing \( \sigma \). In addition, in Fig. 6(c) we show \( \Omega_{min} \), the minimum frequency observable along the Hopf neutral curve, and in Fig. 6(d) the corresponding value \( S_{min} \) of the separation ratio \( S \). Both approach zero with \( \sigma \). It is therefore only in the limit \( \tau \to 0, \sigma \to \infty \) that the codimension-2 bifurcation provides the correct description of the initial instability of an unbounded layer. Only with the boundary conditions (A) do the differences \( \Delta R_{CT} \) and \( \Delta k_{CT} \) vanish identically, and the CT point exists even when the wave numbers are minimized.

In Fig. 7 we show \( R_{CT}(k_c) \) and \( k_c \), as functions of \( S \) for steady convection, for various values of \( \tau \). As evident from Eqs. (2.1) these curves are all independent of the Prandtl number. The neutral curves diverge to infinity already at small negative values of \( S \). For smaller values of \( S \) a second branch of each neutral curve exists, primarily for negative Rayleigh numbers. These branches are not shown. For the branches of interest both \( R_{CT}(k_c) \) and \( k_c \) decrease rapidly with increasing \( S \). The \( k_c \) curves all intersect at \( S = 0 \) where \( k_c = 3.116 \), the value for convection in a pure fluid with no-slip boundary conditions and fixed temperature at top and bottom. Notice, however, that for \( S > 0 \) the values of \( k_c \) continue to decrease, and eventually at critical values \( S_c(\tau) \) they vanish, as shown in Fig. 7(b). At this point the modes that first lose stability as \( R \) is increased have an infinite horizontal extent [cf. Fig. 2(b)], and this is the case for all \( S > S_c(\tau) \), provided that \( R_{CT}(0) > 0 \). Other stability calculations also show this feature.\(^5,8\) The reason for the zero critical wave number for sufficiently large \( S \) can be understood quite simply.\(^3,5,8\) For these values of \( S \) a small destabilizing temperature difference produces a large destabilizing solute distribution. Thus the system behaves as if it were a binary fluid destabilized by the solute distribution \( \Sigma \) which is determined by the zero mass flux boundary condition, or effectively \( \partial \Sigma / \partial z = 0 \) on \( z = 0 \). The resulting problem is therefore entirely analogous to ordinary Rayleigh-Bénard convection with fixed thermal flux boundary conditions, which is known to have \( k_c = 0.22 \). Note that because the destabilizing effects add, the critical Rayleigh number decreases with increasing \( S \). As suggested by the above explanation qualitatively similar behavior is found also with the boundary conditions (B) and (D).

In fact it is possible to be much more precise, and to find analytically the Rayleigh numbers and separation ratios for which \( k_c = 0 \).\(^3\) The technique consists of expanding the characteristic equation (2.12) to \( O(k^4) \). One finds that the \( k \)-independent terms vanish; from the remaining terms there results the relation
\[
\frac{R_S}{R} = 720 + \mu k^2 + O(k^4),
\]
where \( \mu \) is a function of \( R(1 + S) \). This relation holds for both the boundary conditions (C) and (D). Hence in the limit \( k \to 0 \), we find that
\[
\frac{R_S}{R} = 720.
\]

Using our numerically determined values of \( S_c(\tau) \) and the corresponding \( R_{CT}(0) \) we find that (3.6) is obeyed typically to seven significant figures, providing a further test of the numerical procedure. In producing Fig. 7(a) we have therefore used (3.6) to extend the numerically determined critical curves for \( S < S_c(\tau) \) to \( S > S_c(\tau) \), as indicated by the short-dashed lines in Fig. 7(a). In order to obtain the values of \( R \) and \( S \) at which \( k_c \) vanishes for the first \( \tau \), the coefficient \( \mu \) in (3.5) has to be calculated. As seen from Fig. 2(b), when \( \mu > 0 \) \( R_S / R \) is minimized by setting \( k = 0 \), but when \( \mu < 0 \) the minimum occurs for a nonzero value of \( k \). Hence the condition which yields the values of \( R \) and \( S \) for which \( k_c = 0 \) for the first \( \tau \) is given by \( \mu = 0 \). After a considerable amount of algebra, the details of which are not reproduced here, we find that \( k_c = 0 \) for
\[
R(1 + S) < 186.86,
\]

![FIG. 7. Steady-state bifurcations for boundary conditions (C). (a) shows the neutral curves \( R_{CT}(k_c) \) as a function of \( S \) for several values of \( \tau \), while (b) shows the corresponding critical wave numbers. Note that \( k_c \) vanishes at \( S_c(\tau) \). The neutral curves are extended for \( S > S_c(\tau) \) using Eq. (3.6) as indicated by the short-dashed lines. The long-dashed line shows the locus of critical Rayleigh numbers at which \( k \) first vanishes [Eq. (3.7)].](image)
or, equivalently,

\[ R < R_{\text{w}c}(\tau) \equiv 186.86 - 720\tau. \]  

(3.8)

The dividing line (3.7) is also drawn in Fig. 7(a). Note that the analysis predicts how \( R \) on the dividing line scales with both \( S \) and \( \tau \). In addition, combining (3.6) and (3.7) one finds that \( S_{\text{w}c}(\tau) \) scales according to

\[ S_{\text{w}c}(\tau) = \frac{\tau}{0.26 - \tau}. \]  

(3.9)

As shown in Fig. 8 this prediction is in complete agreement with the numerical results. In addition the analytical result (3.9) predicts that \( S_{\text{w}c}(\tau) \) reaches unity when \( \tau = 0.13 \) and infinity when \( \tau = 0.26 \). The concentration and thermal fields are then so tightly coupled that the no-mass-flux boundary conditions on the mass flow are no longer capable of forcing the zero critical wave number.

These results apply to the large-aspect-ratio experiments on pattern selection in water-ethanol mixtures.\(^{16}\) These experiments, carried out for \( \tau = 0.018 \) and \( 0.01 < S < 0.1 \), do reveal the long wavelength of the initial instability,\(^ {16}\) although this is superseded by finite wavelength patterns for only slightly supercritical Rayleigh numbers. This observation is suggestive of the results obtained from two-dimensional simulations of mantle convection with distributed heating and thermally insulating boundaries. Here \( k \) also vanishes but the finite amplitude solutions have finite wavelength of order a few times the layer depth.\(^{32}\)

(ii) Case (D). The boundary conditions (D) are likely to be relevant to cryogenic experiments in which the lower plate is heated at constant electrical power. We present below detailed results for both \(^4\)He-\(^3\)He mixtures and ethanol-water mixtures under circumstances for which the Biot number characterizing the thermal properties of the lower boundary is small. The discussion that follows parallels that given for case (C), and therefore provides a useful comparison between both extremes in the Biot number for the same values of the parameters \( \tau \) and \( \sigma \).

We begin by showing in Fig. 9 the eigenfunctions both for steady convection and for oscillatory convection in the form of LTW and SW for \( \tau = 0.03, \sigma = 0.6 \). In contrast to Fig. 1(a) the SS eigenfunction is no longer symmetric about the midplane. From Fig. 9(b) we see that the left-traveling wave shown leads with the part that is in contact with the conducting boundary, with the result that the cell leans forward. Similar behavior is found for \( \tau = 0.005, \sigma = 14.9 \), although the cell boundaries continue to curve in opposite directions. The standing pattern once again has vertical boundaries, but at no time is the cell completely at rest, as shown in Fig. 9(d). In Fig. 9(c) we show the SW at a time near the kinetic energy minimum. Notice that the cell is temporarily divided

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**FIG. 8.** Calculated values of \( S_{\text{w}c}(\tau) \) for boundary conditions (C) and (D). The crosses indicate numerical results; the solid lines are the theoretical curves (3.9) and (3.13), respectively.

**FIG. 9.** Eigenfunctions for \(^3\)He-\(^4\)He and the boundary conditions (D). (a) shows a steady-state solution for \( S = -0.012 \), (b) shows a left-traveling wave for \( S = -0.6 \), (c) shows a standing wave for \( S = -0.6 \) at a time near minimum kinetic energy, and (d) shows the root-mean-square velocity \( v_{\text{RMS}} \) for a standing wave. Note that \( v_{\text{RMS}} \) does not vanish during an oscillation period.

**FIG. 10.** Neutral stability curves \( R_c(k) \) for a \(^4\)He-\(^3\)He mixture with boundary conditions (D) for (a) the Hopf bifurcation, and (b) the steady-state bifurcation. The dashed lines indicate the values of \( k \), which minimize \( R_c(k) \) for each value of \( S \). Note that \( k \) vanishes when \( S \geq 0.471 \).
into two counterrotating subcells. In Fig. 10 we show the neutral curves \( R_c(k_c) \) for both the oscillatory and steady-state instabilities for a \(^3\text{He} - ^4\text{He}\) mixture \((\tau = 0.03, \sigma = 0.6)\) and several choices of the separation ratio. Again for large enough values of \( S \) \((S > 0.471)\) the minimizing wave number \( k_c \) for the steady-state bifurcation vanishes. Note, however, that for the steady-state neutral curves the minimum is very shallow for a wide range of \( S \) \((0.05 < S < 0.471)\). In Fig. 11 we show the neutral curve \( R_c(k_c) \) for the oscillatory instability, as well as the contours of constant growth rate \( \gamma \) and frequency \( \Omega \) constructed as in (i). The wave-number variation is also indicated, as is the result of scaling the theoretical neutral curve for boundary conditions (A) so as to coincide with the CT point. It is important to observe that with the present boundary conditions the scaled idealized theory indicated by the dashed lines in Figs. 11 is in substantially worse agreement with the exact results than in the case of the boundary conditions (C). For example, Fig. 11(b) shows a discrepancy between the approximate theory and the exact results that reaches 45% at \( S = -0.6 \). Note also that the parameter dependence of the critical wave number is much stronger than in case (C). In a pure fluid \((S = 0)\) \( k_c = 2.552 \) but as \( S \) decreases \( k_c \) increases quite rapidly, reaching 3.228 at \( S = -0.6 \). These results therefore show that \( k_c \) is more sensitive to the Biot number for small negative values of \( S \) than for larger negative values. We also note that the wave number of the fastest growing modes for supercritical Rayleigh numbers tends to be in excess of \( k_c = 3.0 \). Figure 12 shows an enlargement of the region near \( S = 0 \) showing both neutral curves. Once again the Hopf neutral curve does not terminate on the steady-state neutral curve, because the wave number \( k_{CT} \) minimizing \( R_{CT}(k) \) at the end of the Hopf curve \((\Omega_c = 0)\) does not coincide with the wave number that minimizes \( R_c(k) \) for the steady-state bifurcation at \( S = S_{CT} \). In Fig. 13 we show \( R_{CT} - R_{RB}, (S_{CT})^{1/2} \) and \( k_{CT} - k_{RB} \) as functions of \( \tau \) for \( \sigma = 10, 1, \)

![FIG. 11. Critical Rayleigh numbers as a function of the separation ratio \( S \) for the boundary conditions (D) and (a) \( \tau = 0.03, \sigma = 0.6 \); (b) \( \tau = 0.005, \sigma = 14.9 \). Shown are the curves of constant growth rate \( \gamma \) and constant frequency \( \Omega \). The tick marks on the former indicate the wave numbers \( k_c \) in intervals of 0.05 increasing to the left. The horizontal dashed lines indicate the result of scaling the \( \gamma = 0 \) curve for boundary conditions (A) so as to coincide with that for the conditions (D) at the codimension-2 point. The vertical dashed line by the right axis shows the neutral stability curve for the steady state.]

![FIG. 12. (a) An enlargement of the \( R - S \) plane in a \(^3\text{He} - ^4\text{He}\) mixture with boundary conditions (D), illustrating the shielding of the codimension-2 point. The tick marks indicate values of \( k_c \) on the Hopf curve, increasing in units of 0.0001 to the left. (b) As for Fig. 4 but with boundary condition (D).]

![FIG. 13. As for Fig. 5 but with boundary conditions (D).]
and 0.1, while Fig. 14 describes the $\tau$ dependence of $(-\Delta R_{CT})^{1/2}$ and $\Delta k_{CT}$, also for $\sigma = 10, 1,$ and 0.1. As in case (C) $(-\Delta R_{CT})^{1/2}$ and $\Delta k_{CT}$ approach zero linearly with $\tau$; in addition, for small values of $\sigma$ the difference $\Delta R_{CT}$ is not as large as in case (C). We conclude from these results that in order to observe the dynamics associated with the presence of a codimension-2 point it is necessary to employ small values of $\tau$ and large values of $\sigma$, as is the case with boundary conditions (C).

In Fig. 15 we present our results for the steady-state neutral curves and the associated critical wave numbers. The main difference between these results and the earlier ones is in the values of $R_c$ and $k_c$, which for small values of $S$ lie in the vicinity of 1295.778 and 2.552, respectively, the results for Rayleigh-Bénard convection with thermal flux imposed on the bottom boundary and the temperature on the top. As already indicated in Fig. 10(b) the critical wave number $k_c$ vanishes for sufficiently large values of $S$. As in the case of the boundary conditions (C) it is possible to obtain analytically the critical curves for $k_c = 0$. This time one obtains

$$\frac{R S}{\tau} = 720,$$

in the region given by

$$R(1 + S) < 67.634.$$  

These results in turn yield the predictions that $k_c = 0$ in the region

$$R < R_\infty(\tau), \quad S > S_\infty(\tau),$$

where

$$R_\infty(\tau) = 67.634 - 720\tau, \quad S_\infty(\tau) = \frac{\tau}{0.094 - \tau},$$

both of which are again in complete agreement with the numerical results (see Fig. 8). Observe that this time for $\tau > 0.047$ $k_c$ does not vanish for any $S < 1$. Hence with physically reasonable values of $S$ ($0.1 < S < 0.5$) it may not always be possible to reach $k_c = 0$ given the values of $\tau$ that are available.

**IV. DISCUSSION AND CONCLUSIONS**

In this paper we have undertaken a systematic study of the linear stability properties of binary fluid mixtures heated from below. We have focused on parameter values and boundary conditions of direct relevance to the recent experiments on normal $^3$He-$^3$He and ethanol-water mixtures. In this section we summarize and reiterate our main conclusions. The most important of these concerns the accuracy of estimating the separation ratio for a mixture by fitting the observed critical Rayleigh number to a suitably scaled theoretical result obtained for stress-free boundary conditions. To be more quantitative we use the results of Ref. 11 for an 8% ethanol-water mixture. When the top plate is maintained at 10°C and the lower at a higher temperature (not stated), the critical Rayleigh number is 2.215$R_0$, where $R_0 = 1707.765$. Thus $R_c = 3782$. Since for this mixture $\tau = 0.005$ and $\sigma = 14.9$ we obtain from Fig. 3(a) the value $S = -0.55$; the frequency of the neutrally stable oscillations is $\Omega_c = 21.75$ in units of the thermal diffusion time in the vertical, and the critical wave number is 3.167. In contrast, from the linear theory with free boundaries and fixed temperature and concentration at top and bottom, we obtain the result $R_c/R_0 = (1 + \sigma)/(1 + \sigma(1 + S))$, where $R_0 = 657.511$. Interpreting $R_0$ as the critical Rayleigh number for convection in a pure fluid so that $R_c/R_0 = 2.215$, and applying an additional 5% correction to $(R_c/R_0) - 1$, one obtains the result $S = -0.57$ given in Ref. 11. This constitutes a 3.6% error. To confirm this determination of $S$ the Biot number for the experimental boundaries should be estimated, and the effects of sidewalls on the neutral curve should be investigated as in Refs. 5 and 8. Note that with a fixed flux boundary condition at the bottom the scaling procedure yields substantially less accurate results [see Fig. 11(b)].

Throughout this paper we have emphasized the importance of the Biot number in specifying the correct temperature boundary conditions for each experimental
setup. To bracket the range of possibilities we have presented results both for fixed temperature at the bottom and for fixed heat flux at the bottom. In all cases we took the upper boundary to be at constant temperature since in the experiments it is kept in thermal contact with a constant-temperature heat bath. The fixed flux critical Rayleigh numbers and wave numbers are both found to be significantly smaller than those for the fixed temperature case. An extrapolation between these results as a function of the Biot number can be obtained using the results of Ref. 22.

The present study has revealed several other ancillary results of importance to the experiments. The detailed investigation of the parameter dependence of the codimension-2 point revealed that the nonlinear phenomena associated with such a point are most likely to be found in systems with a small Lewis number and a large Prandtl number, when this point is masked by steady-state instabilities with different wave numbers that set in for smaller Rayleigh numbers. In addition, our analytical discussion of the zero critical wave-number regime for steady convection gave a complete picture of the linear analysis in an experimentally interesting parameter regime.\(^{16}\)

After this work was completed we became aware of a related calculation\(^{34}\) for our boundary conditions (C). While there is broad agreement between the two calculations, there are also significant differences. In particular we find that the codimension-2 point is always masked by steady-state instabilities with different critical wave numbers at \(R < R_{CT}\). In Ref. 34 the claim is made that this is never the case for exact eigenfunctions. This could be because the authors apparently minimize \(R_c\) with respect to both \(k\) and \(\Omega\), even though these are not independent, or because of numerical inaccuracy. For example, after correcting for a transposition, the values of \(S_\infty(\tau)\) stated in Ref. 34 are 0.09 for \(\tau = 0.02\) and 0.2 for \(\tau = 0.04\), whereas the exact results are 0.083 and 0.182, respectively (see i). With errors as large as these the masking of the codimension-2 point will not be observable since accuracy to at least six significant figures is necessary (see Fig. 6).

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