Convective Instabilities in Concurrent Two Phase Flow:

Part I. Linear Stability

The linear stability of thermally stratified horizontal two-phase Couette flow is analyzed for the case of a constant vertical temperature gradient. Instabilities driven by buoyancy, surface tension gradients, or shear are allowed for. It is shown that the instability can take three possible forms: streamwise oriented roll vortices, long interfacial waves, and short Tollmien-Schlichting waves. It is shown that the stability limits for rolls are identical to those for plane, stagnant layers. A long wave expansion is presented and the stability limits for this mode are given algebraically. The nonexistence of a Squire's Theorem is demonstrated and some numerical experiments at moderate Reynolds numbers are described. Detailed comparisons with previous work are possible for only one fluid pair, but it is shown that reasonably accurate statements may be made to determine which mode may manifest itself in any given experimental situation.

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SCOPE

Under certain conditions, diffusive heat or mass transfer across a fluid-fluid interface can result in a convective instability. The secondary convection is driven by the temperature or concentration gradients. Because rates of transport are enhanced severalfold over that occurring by the diffusion mechanism alone, an understanding of the phenomena is of great practical interest. Study of this instability of diffusive transport has attracted considerable attention since it was first observed by Bénard (1900). It is now well known to be due to either surface tension variations or buoyantly unstable density variations.

Many theoretical analyses of instabilities driven by buoyancy and surface tension gradients exist for the case of stagnant plane fluid layers, (Berg et al., 1966; Sawistowski, 1971). The results of these studies have been substantiated by experiments, but it has been difficult to apply these results to cases of practical importance because of several restrictive assumptions. One of these is the assumption of initially stagnant phases. The objective in this work is to examine the effect of a rectilinear flow on some of the predictions made in stagnant systems. The work is motivated by the pervasiveness of shear in situations of practical interest such as liquid-liquid extraction, film evaporators, and wetted wall columns. All of these are situations where convective instabilities would be expected. In this paper, the first of a three part series, we employ the techniques of linear stability theory to (1) categorize the possible modes of instability in two-phase concurrent flow and (2) determine quantitative instability criteria which would allow one to predict which of these modes would occur.

CONCLUSIONS AND SIGNIFICANCE

The model system used here is the simplest incorporating the important physics of the problem. Thermally stratified two-phase Couette flow is considered. We take the gravity vector to be perpendicular to the boundaries generating the shear field and the thermal stratification to be linear. The treatment for mass transfer is identical except for the stabilizing effect of the Gibbs adsorption layer. Buoyancy, surface tension, and shear induced instability modes are allowed.

It is found that the instability can take one of three forms. In the first, designated as the longitudinal roll mode, the mean flow is shown to have no effect on the linear stability limits. Thus the critical parameters for onset of instability are identical to those predicted by an analysis for plane stagnant layers. The second mode, denoted as the long interfacial wave mode, is characterized by a low Reynolds number instability. A long wave *expansion is developed and explicit criterion given for* the onset of this mode. The third form of instability, that of short Tollmien-Schlichting waves, is discussed but no detailed computations are reported for this mode. This is a high Reynolds number phenomenon, as opposed to the other two modes. The remainder of the work centers on the development of criteria which would allow one to predict which mode of instability will occur. It is shown that a Squire's transformation exists, but no Squire's Theorem is possible due to the competing physical effects of surface tension, gravity, and shear. Thus, one cannot state a priori what orientation the instability will take. The paper concludes with some representative calculations for specific fluid pairs. For fluid pairs of high interfacial tension and high density ratio, the instability is expected to take the form of longitudinal roll vortices. Conversely, density ratios near unity will favor the appearance of long interfacial waves as the observed mode of instability.

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Convective motions occurring spontaneously during heat or mass transfer between two fluid phases have attracted a great deal of attention since the early experiments of Bénard (1900). It is now well understood that, in general, these motions are produced by the combined effects of density and interfacial tension gradients. Excellent reviews of the vast amount of work which has been done in this area include Berg et al. (1966), Sawistowski (1971), and Berg (1972). However, the present understanding of the effect of shear upon convective instabilities is far from adequate. For example, there are no analyses of the effect of shear on interfacial convection, while experimentally the problem has been considered only by Clark and King (1970), Maroudas and Sawistowski (1964), and Linde and Schwarz (1964). A related problem that has received attention is that of buoyancy-driven instability in a horizontal fluid layer between surfaces in a relative motion, or alternatively, in a fluid layer flowing between two stationary surfaces. The Couette problem is important in the study of atmospheric circulation and cloud formation. The linear stability of stratified Couette flow was treated by Gallagher and Mercer (1965), Deardorff (1965), and Ingersoll (1966), and the related uniqueness problem was solved by Joseph (1966). Brunt (1951), Chandra (1938), Benard and Avsec (1938), and Graham (1933) studied the problem experimentally. Gage and Reid (1968) and Phillips and Walker (1932) considered the Poiseuille problem. Despite the rich history of this problem, certain experimental observations regarding preferred instability orientation are not predicted, as we will discuss below.

Since physically important applications of convective motion, for example, liquid-liquid extraction, film evaporators and transport in wetted wall columns commonly involve the presence of shear, it is surprising that the area has generated so little interest. The present work is an integrated approach to the hitherto neglected study of convective flow in systems undergoing shear. This study is confined to the case of horizontal fluid layers, the simplest system retaining the important physical aspects of the problem and includes the inevitable interaction of the interfacial tension and buoyancy driven modes. Results of linear and energy stability theory are presented in Parts I and II, while Part III deals with some companion experiments.

The linear stability of horizontal, stratified fluid layers subject to surface driven convection is treated in a number of works considering the various physical possibilities, that is, single or double fluid layers, flat or deformable interface, and simultaneous buoyancy instability. The significant works in the area are Pearson (1957), Sternling and Scriven (1959), Nield (1964), Smith (1966), Zeren and Reynolds (1972), and Palmer and Berg (1972). These analyses have assumed a quiescent and diffusive (steady) base state. The experiments of Palmer and Berg (1971) are the best check of the theory: the agreement is fair considering the experimental difficulties. Our treatment here will be for instabilities driven by energy transport between the two phases. Recent work by Palmer and Berg (1972) and Brian (1971) has shown that in the case of mass transfer accumulation of mass in the Gibbs layer may have profound stabilizing effects on surface tension driven instabilities. In cases for which the instability arises primarily as the effect of buoyancy or shear, however, the two transport mechanisms are analogous. Many of our overall conclusions regarding the orientation of disturbances are in fact uneffected by Gibbs adsorption.

Here, we extend linear theory to study the effect of shear in a physically realistic model. We consider steady state temperature stratification in a horizontal gap filled with two immiscible fluids. The two bounding surfaces will be in steady relative motion inducing a two-dimensional rectilinear flow in the base case, and since waves are a characteristic instability, the interface will be allowed to deform.

The same stratification that induces surface tension instability will cause a buoyancy gradient which may be stabilizing or destabilizing. Both modes are included in the following analysis. The modes are coactive when the direction of transfer (in this case heat transfer) is such that both modes would be excited singly, given severe enough stratification. In this case instability will arise at a less severe stratification than if either effect were present alone. Conversely, the modes are counteractive when they would be activated by transfer in opposing directions, and consequently the stratification necessary to cause instability is greater than if the effects were considered singly.

LINEAR STABILITY ANALYSIS

As a base case we consider two immiscible fluids located between two rigid and isothermal boundary surfaces of infinite extent. A reference frame is chosen such that the interface at $z = 0$ is stationary: the upper surface (at temperature T_1) is located at $z = d_a$ and moves in the y direction at rate V_0 while the lower plate (at temperature T_2) at $z = -d_b$ moves in the opposite direction at rate $\mu_a/\mu_b\, d_b/d_a\, V_0$. After scaling with the characteristic quantities V_0, $d(= d_a + d_b)$, and $L_a d$, the base state is described by

$$V_a = (1 + r)z \tag{2.1a}$$

$$V_b = et(1 + r)z \tag{2.1b}$$

$$T_a = z + \left(\frac{sr}{1+r} + \frac{T_2}{T_1 - T_2} \cdot \frac{1 + rs}{1 + r}\right) \tag{2.1c}$$

$$T_b = sz + \left(\frac{sr}{1+r} + \frac{T_2}{T_1 - T_2} \cdot \frac{1 + rs}{1 + r}\right) \tag{2.1d}$$

where the upper plate is located at $z = (1 + r)^{-1}$ and the lower plate at $z = -r(1 + r)^{-1}$.

Employing the Boussinesq approximation, the linearized disturbance momentum equations are found in the normal way to be

$$\frac{\partial \vec{u}'}{\partial t} + V\frac{\partial \vec{u}'}{\partial y} + \hat{j}w'\frac{\partial V}{\partial z} = -\frac{1}{\rho}\nabla p' + \nu\nabla^2\vec{u}' - \beta g\hat{k}\theta' \tag{2.2}$$

where

$$\beta = -\left(\frac{1}{\rho}\frac{\partial \rho}{\partial T}\right) \tag{2.3}$$

Pressure and horizontal velocity components are removed by taking the scalar product of $\hat{k}$ and the double curl of (2.2a):

$$\frac{\partial}{\partial t}\nabla^2 w' + V\frac{\partial}{\partial y}\nabla^2 w' = \nu\nabla^2\nabla^2 w' + \beta g\nabla_1^2\theta' \tag{2.4}$$

The linearized disturbance energy equation is

$$\frac{\partial \theta'}{\partial t} + V\frac{\partial \theta'}{\partial y} + w'\frac{\partial T}{\partial z} = \kappa\nabla^2\theta' \tag{2.5}$$

We nondimensionalize the two disturbance equations using the scales

$$\{t, T, V, w', \theta'\} = \left\{\frac{d}{V_0}, L_a d, V_0, V_0, L_a d\right\} \quad (2.6)$$

to give in the upper phase

$$Re_a\left(\frac{\partial}{\partial t} + V_a\frac{\partial}{\partial y}\right)\nabla^2 w_a' = \nabla^4 w_a' + \frac{Ra_a}{Pe_a}\nabla_1{}^2\theta_a' \quad (2.7a)$$

$$\frac{\partial\theta_a'}{\partial t} + V_a\frac{\partial\theta_a'}{\partial y} + w_a' = \frac{1}{Pe_a}\nabla^2\theta_a' \quad (2.7b)$$

and in the lower phase

$$Re_b\left(\frac{\partial}{\partial t} + V_b\frac{\partial}{\partial y}\right)\nabla^2 w_b' = \nabla^4 w_b' + \frac{Ra_b}{sPe_b}\nabla_1{}^2\theta_b' \quad (2.7c)$$

$$\frac{\partial\theta_b'}{\partial t} + V_b\frac{\partial\theta_b'}{\partial y} + w_b's = \frac{1}{Pe_b}\nabla_1{}^2\theta_b' \quad (2.7d)$$

The periodic horizontal dependence of the disturbance is expressed using normal mode analysis

$$\begin{bmatrix}\theta'\\ w'\\ \eta'\end{bmatrix} = \begin{bmatrix}\theta(z)\\ w(z)\\ \eta\end{bmatrix} e^{i(\delta x+\beta y)}e^{\sigma t} \quad (2.8)$$

where β and δ are horizontal wave numbers. η is the surface deformation in the vertical direction, appearing in the boundary conditions below. σ is the complex time constant of the disturbance, its real part is positive if the disturbance grows, that is unstable base case, and negative if it decays, that is, stable base case. In the transformed equations δ appears implicitly in the total wave number $\alpha = (\delta^2 + \beta^2)^{1/2}$. These equations are

$$Re_a(\sigma + V_a i\beta)(D^2 - \alpha^2)w_a = (D^2 - \alpha^2)^2 w_a - \frac{Ra_a}{Pe_a}\alpha^2\theta_a \quad (2.9a)$$

$$Re_b(\sigma + V_b i\beta)(D^2 - \alpha^2)w_b = (D^2 - \alpha^2)^2 w_b - \frac{Ra_b}{sPe_b}\alpha^2\theta_b \quad (2.9b)$$

$$(\sigma + V_a i\beta)\theta_a + w_a = \frac{1}{Pe_a}(D^2 - \alpha^2)\theta_a \quad (2.9c)$$

$$(\sigma + V_b i\beta)\theta_b + w_b s = \frac{1}{Pe_b}(D^2 - \alpha^2)\theta_b \quad (2.9d)$$

The boundary conditions at the walls reflect the isothermal no-slip character

$$w_a = Dw_a = \theta_a = 0 \quad \text{at} \quad z = (1 + r)^{-1} \quad (2.10a)$$

$$w_b = Dw_b = \theta_b = 0 \quad \text{at} \quad z = -r(1 + r)^{-1} \quad (2.10b)$$

In the disturbed state, the interface has dimensional position $z = \eta'(x, y)$ in the vertical direction. At this position the disturbed flow has the following dimensional continuity conditions:

$$T_a + \theta_a' = T_b + \theta_b' \quad (2.11a)$$

$$k_a\frac{\partial}{\partial z}(\theta_a' + T_a) = k_b\frac{\partial}{\partial z}(\theta_b' + T_b) \quad (2.11b)$$

$$\nabla_1\gamma + \gamma\hat{k}\nabla_1{}^2\eta' = +\hat{k}(P_a' - P_b') + \hat{k} \cdot [\mu_b(\nabla\vec{u}_b + (\nabla\vec{u}_b')^T) - \mu_a(\nabla\vec{u}_a' + (\nabla\vec{u}_a')^T)] \quad (2.11c)$$

$$\vec{u}_a' + V_a\hat{j} = \vec{u}_b' + V_b\hat{j} \quad (2.11d)$$

$$\left(\frac{\partial}{\partial t} + V_b\frac{\partial}{\partial y}\right)\eta' = w' \quad (2.11e)$$

These represent continuity of temperature, heat flux, stress, and velocity. Equation (2.11e) is the kinematic condition relating change in interface position to velocity. These conditions are transferred to $z = 0$ using a Taylor series, and the basic state is subtracted out giving the following linearized disturbance equations:

$$\theta_a' + \eta' L_a = \theta_b' + \eta' L_b \quad (2.12a)$$

$$k_a\frac{\partial\theta_a'}{\partial z} = k_b\frac{\partial\theta_b'}{\partial z} \quad (2.12b)$$

$$\nabla_1{}^2\gamma = \mu_b\left(-\frac{\partial^2 w_b'}{\partial z^2} + \nabla_1{}^2 w_b'\right) - \mu_a\left(-\frac{\partial^2 w_a'}{\partial z^2} + \nabla_1{}^2 w_a'\right) \quad (2.12c)$$

$$(P_a' - P_b') + (\rho_b - \rho_a)g\eta' + 2\mu_b\frac{\partial w_b'}{\partial z} - 2\mu_a\frac{\partial w_a'}{\partial z} = \gamma\nabla_1{}^2\eta' \quad (2.12d)$$

$$w_a' = w_b' \quad (2.12e)$$

$$\eta'\frac{\partial V_a}{\partial z} + v_a' = \eta'\frac{\partial V_b}{\partial z} + v_b' \quad (2.12f)$$

$$u_a' = u_b' \quad (2.12g)$$

$$\frac{\partial\eta'}{\partial t} = w_b' \quad (2.12h)$$

To eliminate pressure from the normal stress condition, we can use an expression derived from taking the scalar product of (2.2a) and the horizontal gradient ∇_1, specifically,

$$\nabla_1{}^2 p' = \rho\left(-\nu\nabla^2\frac{\partial w'}{\partial z} + \frac{\partial}{\partial t}\frac{\partial w'}{\partial z} - \frac{\partial V}{\partial z}\frac{\partial w'}{\partial y} + V\frac{\partial}{\partial y}\frac{\partial w'}{\partial z}\right) \quad (2.13)$$

Next we make the horizontal Fourier decomposition of Equations (2.12) and (2.13) using (2.13) to eliminate pressure in (2.12d). The scalings of Equation (2.6) are used to give

$$\theta_a = \theta_b + \eta(s - 1) \quad (2.14a)$$

$$sD\theta_a = D\theta_b \quad (2.14b)$$

$$(D^2 w_a + \alpha^2 w_a) - \frac{1}{et}(D^2 w_b + \alpha^2 w_b) = -\alpha^2\frac{Ma}{Pe_a}[\theta_a + \eta] \quad (2.14c)$$

$$-(D^3 w_a - 3\alpha^2 D w_a) + \frac{1}{et}(D^3 w_b - 3\alpha^2 D w_b) + \sigma Re_a(Dw_a - Dw_b t^{-1})\, i\beta Re_a\left(-w_a DV_a + \frac{w_b}{t}DV_b\right) = \alpha^2 Re_a\eta(F + \alpha^2 S) \quad (2.14d)$$

$$w_a = w_b \quad (2.14e)$$

$$Dw_b = Dw_a + i\beta(DV_b - DV_a)\eta \quad (2.14f)$$

$$\sigma\eta = w_b \qquad (2.14g)$$

The disturbance can take one of two basic orientations which we will define as longitudinal and transverse, in conformity with the usual definition. In the former case, $\beta = 0$ and the wavefronts are parallel to the base state velocity vector. In the latter case, $\beta = \alpha$ and the wavefronts are perpendicular to that vector. It is especially important to note that the equations for longitudinally oriented disturbances are identical to those for plane, stagnant layers, since the terms in (2.9) and (2.14) involving the mean flow are zero for $\beta = 0$. This has been previously noted in similar studies and is due, of course, to the fact that a longitudinal roll vortex with disturbance streamwise velocity independent of streamwise coordinate has no Reynolds stress in the flow direction. Thus, a roll does not interact with the mean flow and stability limits for such rolls are independent of the features of this base flow. This is true for all rectilinear flows, not just the simple Couette flow treated in detail here.

SQUIRE'S TRANSFORMATION

Squire (1934) proved that, in parallel flows, transverse disturbances are excited at lower rates of shear than oblique disturbances. His proof involved transforming the linear stability equations for any oblique disturbance to an equivalent transverse disturbance with Reynolds number dependent upon the orientation. Oblique disturbances have the same stability limits as transverse disturbances at a lower Reynolds number. The theorem follows with the a priori knowledge that shear is destabilizing for that problem.

We attempt here to prove a theorem for the present problem in the spirit of Squire. This could be especially valuable since, as noted above, the longitudinal limit of Equations (2.9), (2.10), and (2.14) is the system of Zeren and Reynolds (1972). First, Equations (2.9), (2.10), and (2.14) will be transformed to remove β. Then the known effects of gravity, surface tension, and shear upon convective instabilities will be used to predict the relative stability of different orientations.

The three-dimensional problem is reduced to an equivalent two-dimensional problem under the following transformation:

$$\sigma = \beta \underline{\sigma}$$

$$\eta = \frac{\alpha}{\beta} \underline{\eta}$$

$$\theta_a = \frac{\alpha}{\beta} \underline{\theta}_a$$

$$Re = \frac{\alpha}{\beta} \underline{Re}; \; Pe = \frac{\alpha}{\beta} \underline{Pe} \qquad (3.1)$$

$$Ma = \underline{Ma} \text{ and } Ra = \underline{Ra}$$

$$S = \frac{\beta^2}{\alpha^2} \underline{S}$$

$$F = \frac{\beta^2}{\alpha^2} \underline{F}$$

where the underlined quantities are associated with the transformed equations (that is, β removed). It is clear that oblique disturbances have the same critical Ma or Ra as transverse waves at smaller Re and larger S and F.

On the basis of previous work it is possible to predict the effects of shear, surface tension, and gravity. The last two effects limit the system's freedom by providing a restoring force for small and large wavelengths, respectively; hence in the stagnant and diffusive Marangoni problem Scriven and Sternling (1964) clearly demonstrated the stabilizing effect of increasing surface tension while Smith (1966) dramatically showed that gravity stabilizes long waves. These effects carry over to the present problem, tending to stabilize the longitudinal mode relative to the transverse mode.

At this point it is appropriate to briefly review previous work on density driven instabilities in a shear environment, all of it dealing with a single fluid between horizontal, isothermal plates. Gage and Reid (1968) considered Poiseuille flow; while Ingersoll (1966), Gallagher and Mercer (1965), and Deardorf (1965) examined Couette flow. All of these theoretical works indicate longitudinal rolls to be the preferred mode. The experiments of Bernard and Avsec (1938), Chandra (1938), and Brunt (1951) (all on Couette flow) verify predictions except that transverse traveling waves are possible for $Re < 7$ at highly supercritical Rayleigh numbers, $Ra \sim 17{,}000$. On the basis of this work it can be inferred that, in contrast to the usual situation in parallel flow instability, shear stabilizes convective instabilities at moderate Re, causing the longitudinal mode to be preferred.

Hence, even though there is a Squire's transformation, no theorem is possible in this case due to the competing effects of shear, surface tension, and gravity. While the linear stability equations must still be solved for transverse disturbances to allow comparison with previous work, the transformation does suggest two possibilities. Namely, we would expect longitudinal rolls at large flow rates for systems where γ and ρ_b/ρ_a are large, and transverse interfacial waves in the opposite extreme.

LONGWAVE EXPANSION

The Equations (2.9), (2.10), and (2.14) have a transverse, interfacial wave solution that can be described using a long wave expansion technique similar to that of Yih (1967). We develop here a means of calculating a wave speed, growth rate, and critical Rayleigh number for this mode.

For ease of comparison with Yih in the appropriate limit we introduce stream functions having Fourier transforms φ and χ for the upper and lower phases, respectively. Hence

$$w_a = -i\beta\varphi \qquad (4.1a)$$

and

$$w_b = -i\beta\chi \qquad (4.1b)$$

Also we make the conversion $\sigma = -ic\beta$ so that the growth factor becomes $c_i\beta$ and the wave speed c_r where c_r and c_i are the real and imaginary parts of the complex numbers c. The linear disturbance equations in the transverse limit ($\alpha = \beta$) becomes

$$Re_a\beta[(1+r)z - c](D^2\varphi - \beta^2\varphi) + i(D^4\varphi - 2\beta^2D^2\varphi + \beta^4\varphi) + \beta\theta_a\frac{Ra_a}{Pe_a} = 0 \qquad (4.2a)$$

$$Re_b\beta[(1+r)et\,z - c](D^2\chi - \beta^2\chi) + i(D^4\chi - 2\beta^2D^2\chi + \beta^4\chi) + \beta\theta_b\frac{Ra_b}{Pe_b s} = 0 \qquad (4.2b)$$

$$i\beta[(1+r)z - c]\theta_a - i\beta\varphi = Pe_a^{-1}(D^2\theta_a - \beta^2\theta_a) \qquad (4.2c)$$

$$i\beta[(1+r)et\,z - c]\theta_b - i\beta s\varphi = Pe_b^{-1}(D^2\theta_b - \beta^2\theta_b) \qquad (4.2d)$$

At $z = 0$ the interface conditions become

$$sD\theta_a = D\theta_b \qquad (4.3a)$$

$$\theta_a = \theta_b + \eta(s-1) \quad (4.3b)$$

$$\eta = \varphi/c \quad (4.3c)$$

$$\varphi = \chi \quad (4.3d)$$

$$D\varphi = D\chi - \eta(1-et)(1+r) \quad (4.3e)$$

$$D^2\varphi + \beta^2\varphi - e^{-1}t^{-1}(D^2\chi + \beta^2\chi) = - Ma\, i\beta Pe_a{}^{-1}(\theta_a + \eta) \quad (4.3f)$$

$$D^3\varphi - 3\beta^2 D\varphi - e^{-1}t^{-1}(D^3\chi - 3\beta^2 D\chi) + Re_a i\beta c(D\varphi - t^{-1}D\chi) + \varphi i\beta Re_a(1+r)(1-e) + \beta\eta i Re_a(F + \alpha^2 S) = 0 \quad (4.3g)$$

The boundary conditions on the rigid surfaces remain as before.

In order to determine the critical parameters for the onset of long interfacial waves, one is faced with much tedious algebra, the details of which are omitted here for the sake of brevity. The technique used is identical in spirit to that of Yih (1967). The variables χ, φ, θ_a, θ_b, c, and η are expanded in an asymptotic power series in β. At each level in the expansion, a coupled set of equations is solved subject to the boundary conditions at that order. The kinematic condition is then used to determine successive approximations to the wave speed c_R and growth rate c_i.

The differential equations at lowest order are

$$D^4\varphi_0 = 0 \quad (4.4a)$$

$$D^4\chi_0 = 0 \quad (4.4b)$$

$$D^2\theta_{a,0} = 0 \quad (4.4c)$$

$$D^2\theta_{b,0} = 0 \quad (4.4d)$$

with the interfacial conditions ($z = 0$):

$$\eta_0 = \varphi_0/c_0 \quad (4.5a)$$

$$\varphi_0 = \chi_0 \quad (4.5b)$$

$$D\varphi_0 = D\chi_0 - \eta_0(1-et)(1+r) \quad (4.5c)$$

$$D^2\varphi_0 - e^{-1}t^{-1}D^2\chi_0 = 0 \quad (4.5d)$$

$$D^3\varphi_0 - e^{-1}t^{-1}D^3\chi_0 = 0 \quad (4.5e)$$

In addition we adopt the normalizing condition:

$$\varphi_0(0) = 1 \quad (4.6)$$

Then

$$\varphi_0 = 1 + A_1 z + A_2 z^2 + A_3 z^3 \quad (4.7a)$$

$$\chi_0 = 1 + B_1 z + B_2 z^2 + B_3 z^3 \quad (4.7b)$$

$$\theta_{a,0} = A_4 + A_5 z \quad (4.7c)$$

$$\theta_{b,0} = B_4 + B_5 z \quad (4.7d)$$

and it is a straightforward matter to determine the coefficients A_i and B_i. (See Table 1 for their expressions.) To zeroth order, c is real:

$$c_0 = \frac{2r^2et(1+r)(1-et)}{(et)^2r^2 + 2ret(2r^2+3r+2)+1} \quad (4.8)$$

The wave speed expressed in (4.8) is identical to Yih's equation (34) with appropriate modifications for reference frame and velocity scaling. To first order the governing equations are

$$Re_a[(1+r)z - c_0]D^2\varphi_0 + iD^4\varphi_1 + \theta_{a,0}\frac{Ra_a}{Pe_a} = 0 \quad (4.9a)$$

$$Re_b[(1+r)et\, z - c_0]D^2\chi_0 + iD^4\chi_1 + \theta_{b,0}\frac{Ra_b}{Pe_b s} = 0 \quad (4.9b)$$

with interfacial conditions ($z = 0$):

$$\varphi_1 = \chi_1 \quad (= 0 \text{ as a consequence } 4.6) \quad (4.10a)$$

$$\eta_1 = \frac{\varphi_1 - \eta_0 c_1}{c_0} = -\frac{c_1}{c_0{}^2} \quad (4.10b)$$

$$D\varphi_1 = D\chi_1 - \eta_1(1-et)(1+r) \quad (4.10c)$$

$$D^2\varphi_1 - e^{-1}t^{-1}D^2\chi_1 = - Ma\, iPe_a{}^{-1}(\theta_{a,0} + \eta_0) \quad (4.10d)$$

$$D^3\varphi_1 - e^{-1}t^{-1}D^3\chi_1 + Re_a ic_0(D\varphi_0 - D\chi_0 t^{-1}) + \varphi_0 iRe_a(1+r)(1-e) + \eta_0 iFRe_a = 0 \quad (4.10e)$$

The solutions of (4.9) are a homogeneous part (of polynomial form) and a particular part. With the interfacial and boundary surface conditions, the polynomial coefficients can be eliminated in a straightforward and tedious manner to give a relation for c_1:

$$c_1 P_{21} + \frac{iRa_a}{Re_a}P_{23} + iRe_a P_{24} = 0 \quad (4.11)$$

Note that c_1 is imaginary, and at marginal conditions ($c_1 = 0$)

$$Ra_a = -\frac{P_{24}}{P_{23}}Re_a{}^2 \quad (4.12)$$

P_{21}, P_{23}, and P_{24} are complicated, but real, functions of the numerous dimensionless groups (see Table 1).

It should be noted that for a given system, Ma, Ra_a, and Ra_b are fixed ratios of each other irrespective of temperature stratification, hence

$$Ra_b = \frac{esx}{E}Ra_a \quad (4.13a)$$

$$Ma = Q\, Ra_a \quad (4.13b)$$

and the criteria of (4.11) and (4.12) can be expressed in terms of Ra_b or Ma just as readily. Similarly,

$$Re_b = e\, Re_a \quad (4.14)$$

irrespective of shear.

Equations (4.11) and (4.12) give growth rates and wave speeds for the long wave solution which will be compared below to the general solution.

CALCULATIONS AND RESULTS

It was not feasible to do a parametric study in this problem due to the large number of dimensionless groups. Instead, solutions were calculated for particular fluid systems, in particular for benzene/water which Zeren and Reynolds (1970) used in their calculations.

While (4.11) and (4.12) provide the necessary results for long wavelength transverse waves, it remains to solve (9.9), 9.10), (9.14) for shorter wavelengths of the transverse mode. Since these equations are linear, it is possible to generate linearly independent solutions at a given (Ra, c) and superpose them to satisfy the boundary conditions if the chosen (Ra, c) pair is an eigenvalue. In the current work three linearly independent solutions were generated by integrating from the upper to the lower surface using a Runge-Kutta method. These solutions were constructed to satisfy the upper surface boundary conditions, the chosen (Ra, c) pair being an eigenvalue if it was possible to satisfy the boundary conditions on the lower surface. Eigenvalues pairs were located using bivariate interpolation of the complex determinant in the

TABLE 1. DIMENSIONLESS GROUPS OF 4.11 AND 4.12

$$A_1 = + \frac{+1 + et\,r^2(4r+3)}{2r^2 et}$$

$$A_2 = \frac{et\,r^3 + 1}{r^2(1+r)et}$$

$$A_3 = \frac{et\,r^2 - 1}{2\,et\,r^2}$$

$$A_4 = \frac{s-1}{c_0(sr+1)}$$

$$A_5 = \frac{(1+r)(s-1)}{c_0(sr+1)}$$

$$B_2 = etA_2$$

$$B_3 = etA_3$$

$$P_1 = 6(1+r)A_3$$

$$P_2 = 2(1+r)A_2 - 6c_0 A_3$$

$$P_3 = -2c_0 A_2$$

$$P_4 = 6(1+r)(et)^2 eA_3$$

$$P_5 = 2(1+r)(et)^2 eA_2 - 6c_0 e^2 t A_3$$

$$P_6 = -2c_0 e^2 t A_2$$

$$P_7 = \frac{1}{6} et \left[c_0 (A_1 - B_1 t^{-1}) + (1+r)(1-e) + \frac{F}{c_0} \right]$$

$$P_8 = \frac{et(A_4 + c_0^{-1})}{2\,Pr_a}$$

$$P_9 = -\frac{(1-et)(1+r)}{c_0^2}$$

$$P_{10} = -\frac{Qr^2P_8}{(1+r)^2} + \frac{esx}{E}\left[\frac{A_5 r^5}{120xPr_a(1+r)^5} - \frac{P_{17}r^4}{24(1+r)^4}\right]$$

$$P_{11} = \frac{P_7 r^3}{(1+r)^3} - \frac{P_4 r^6}{360(1+r)^6} + \frac{P_5 r^5}{120(1+r)^5} - \frac{P_6 r^4}{24(1+r)^4}$$

$$P_{12} = \frac{(1-et)(1+r)}{c_0^2}$$

$$P_{13} = \frac{2rP_8Q}{(1+r)} + \frac{esx}{E}\left[\frac{A_5 r^4}{(1+r)^4\,24xPr_a} + \frac{P_{17}r^3}{6(1+r)^3}\right]$$

$$P_{14} = -\frac{3P_7 r^2}{(1+r)^2} - \frac{P_5 r^4}{24(1+r)^4} + \frac{P_6 r^3}{6(1+r)^3} + \frac{P_4 r^5}{60(1+r)^5}$$

$$P_{15} = -\frac{A_5}{120Pr_a(1+r)^5} - \frac{A_4}{24Pr_a(1+r)^4}$$

$$P_{16} = -\frac{P_1}{360(1+r)^6} - \frac{P_2}{120(1+r)^5} - \frac{P_3}{24(1+r)^4}$$

$$P_{17} = \frac{A_4 + \dfrac{1-s}{c_0}}{xs\,Pr_a}$$

$$P_{18} = -\frac{A_5}{24Pr_a(1+r)^4} - \frac{A_4}{6Pr_a(1+r)^3}$$

$$P_{19} = -\frac{P_1}{60(1+r)^5} - \frac{P_2}{24(1+r)^4} - \frac{P_3}{6(1+r)^3}$$

$$P_{20} = \frac{2(1+r\,et)(1+r)^3}{r^2\,et}$$

$$P_{21} = \frac{3(1-et\,r^2)(1+r)^2}{2r^2\,et}$$

$$P_{22} = P_{12} + \left(\frac{P_{12}r}{1+r} + P_9\right)\left(P_{21} - \frac{P_{20}}{(1+r)}\right)\frac{1}{(1+r)^2}$$

$$P_{23} = P_{13} - P_{18} + \frac{r^2\,et}{(1+r)^2}\left(\frac{P_{18}}{1+r} - P_{15}\right)\left(\frac{rP_{20}}{(1+r)} + P_{21}\right) + \frac{1}{(1+r)^2}\left(P_{21} - \frac{P_{20}}{(1+r)}\right)\left(\frac{rP_{13}}{(1+r)} + P_{10}\right)$$

$$P_{24} = P_{14} - P_{19} + \frac{r^2\,et}{(1+r)^2}\left(\frac{P_{19}}{1+r} - P_{16}\right)\left(\frac{rP_{20}}{(1+r)} + P_{21}\right) + \frac{1}{(1+r)^2}\left(P_{21} - \frac{P_{20}}{(1+r)}\right)\left(\frac{rP_{14}}{(1+r)} + P_{11}\right)$$

superposition step.

This numerical routine was used on the water-benzene pair studied by Zeren and Reynolds. No small or moderate wavelength solutions were found at marginal stability for this particular pair although this result may not be conclusive. The routine did locate solutions at small wave numbers and these results are in excellent agreement with the long wave expansion (4.12). We hasten to qualify the above remarks by noting that, at sufficiently high Reynolds number, the flow will become unstable to short waves of the Tollmein-Schlichting type, regardless of the stratification. By Tollmein-Schlichting waves we refer to a class of well-studied parallel shear flow instabilities occurring in flows which are stable according to Rayleigh's inviscid criterion, but become unstable at sufficiently high Reynolds number; see Stuart (1963). Little theoretical work appears to have been done in two-phase flow for disturbances of this class, even for the case of homogeneous fluids. Such work would involve numerical solutions of the Orr-Sommerfeld equation at high *Re* for both phases, which is well known to entail severe numerical difficulties. The experimental work of Charles and Lilleleht (1965) indicates that instabilities of this class can be expected for individual phase Reynolds numbers of the order of 2000. In the work reported here, we have focused upon low to moderate Reynolds numbers, for which this mode can be safely neglected.

An overview of the stability diagram for the flow may be succinctly summarized as follows. Given a fluid pair and the depth ratio, we choose the Rayleigh number Ra_a as a stability parameter. This choice is somewhat arbitrary since Ra_a may be expressed in terms of the Marangoni number if desired. A convenient parameter describing the effect of the flow is the Reynolds number *Re*. Thus a description of the effect of shear on convective instabilities is given by a stability diagram in the (*Ra*, *Re*) plane. This is shown schematically in Figure 1. On such a diagram, the linear limit for onset of rolls appears as a horizontal line since, as we have seen, the base flow does not interact with roll vortices. At some high Reynolds number,

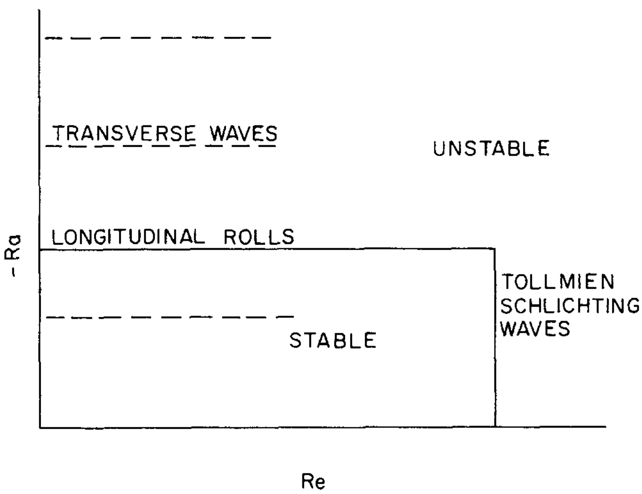

Fig. 1. Relationship of the various instability modes: Dashed lines represent possible locations of the transverse wave solution.

the flow becomes unstable to Tollmien-Schlichting waves; this limit appears as a cut-off Reynolds number above which the flow is certainly unstable. Finally, the presence of an interface allows a long interfacial wave disturbance whose locus in the (Ra, Re) plane is given by the complicated expression (4.12). The fact that we have chosen to depict this limit schematically as horizontal lines will be discussed below.

In Table 2 long wave results of (4.12) are compared with convective stability criteria of Zeren and Reynolds and Smith, all being expressed in terms of Ra_a. As expected the pure surface tension driven mode described by Smith required greater stratification than the combined density-surface tension mode. The traveling long wave is excited at an intermediate Ra_a, and presumably will not be observed—longitudinal rolls excited at the Ra_a of Zeren and Reynolds will dominate.

Long wave results for other fluid systems are given in Table 3. Although there are no results for comparison, in light of the close results in Table 2 it seems probable that long waves will dominate in some of the systems, especially when ρ_a/ρ_b is close to unity.

The results in Table 3 were found to be independent of Re_a with one exception. It is interesting that shear rate had a significant effect on the stability criteria of only one of the fluid systems studied. This is possible since F appears in the numerator of (4.12), canceling the velocity dependence if other numerator terms are sufficiently small. Thus for over a significant range of Reynolds numbers, we find that the most important stability parameter describing the onset of long interfacial waves is the modified Froude number F. The physical interpretation of this group's importance may be seen as follows. The Froude number appears in the normal stress balance and is a dimensional measure of the hydrostatic force caused by the presence of a wave on the interface. (Note that $F = 0$ if $\rho_a = \rho_b$.) Now the disturbance under discussion is a long interfacial wave, and for such long waves, surface tension cannot act as a restoring force. Thus the only restoring force on a long wave is gravity, manifested by the magnitude of F. It is thus not surprising that this is the major stability parameter for this mode of disturbance. The one fluid pair showing any shear dependence was that for which the density ratio is near 1.0 (n-octanol/water: $\rho_a/\rho_b \cong 0.83$).

Comparison with Nield's (1964) results for the combined Marangoni-Rayleigh problem is omitted since certain conflicting predictions arise when the one-fluid model is applied to a two-fluid system. For example, in heating from above for the usual case where $\partial\gamma/\partial T$, $\partial\rho/\partial T < 0$, the one fluid model predicts unconditional stability for the lower fluid whereas the upper fluid is unstable for sufficiently severe stratification, and suitably small thicknesses. Many such odd predictions derive from neglecting the effect of the adjacent phase upon interface temperature, for example, if fluid moves from both phases towards the same spot on the interface no surface tension driven instability occurs if the hotter fluid has larger ρC_p. The one-fluid model has found application in certain limiting cases, namely, convective instabilities in a liquid layer which adjoins an upper gas phase, Palmer and Berg (1971).

CONCLUSIONS

The existence of traveling wave disturbances was analytically demonstrated. Since the equations for longitudinal roll disturbances are identical to those for stagnant systems, a simple criteria now exists to determine whether rolls or waves will be excited. This is schematically illustrated in Figure 1: waves exist at conditions where the dotted curve described by equation (4.12) falls below the solid horizontal line. For the one fluid pair studied by Zeren and Reynolds (1970), longitudinal rolls are the preferred mode, but this is not a global result. Unfortunately, calculations similar to those of Zeren and Reynolds are not available for other fluid pairs. However the calculations of Smith (1966) may be used for comparison against results calculated by (4.12) when surface tension driven instabilities dominate. Similarly, for cases of high interfacial tension with coactive buoyancy and surface tension driven convection, Nield's (1964) results may be employed with some degree of confidence, provided the second phase is a gas.

In Part 2 of this work, the nonlinear formulation of this problem is solved to yield sufficient conditions for stability. In Part 3, we will discuss some related experimental observations.

TABLE 2. BENZENE/WATER—COMPARISONS WITH MARGINAL STABILITY CRITERIA OF OTHER WORKERS

Worker	d (cm)	Critical Ra_a	c_0
Long wave	0.4	-5.75×10^6	9.73×10^{-2}
Long wave	0.2	-3.04×10^5	9.73×10^{-2}
Zeren and Reynolds	0.2	-1.1×10^5	—
Smith	0.2	-4.38×10^5	—

Note that the $Ra_a < 0$ for heating from below. Long wave results for this fluid pair are shear independent.

TABLE 3. LONG WAVE RESULTS FOR DIFFERENT FLUID PAIRS. EXCEPT WHERE INDICATED, THERE IS STABILITY FOR $Ra > Ra_a$. $r = 1.0$ FOR ALL CASES

Fluids	d (cm)	Critical Ra_a	Re_a	c_0
Water/dibromoethane	0.2	1.39×10^5*	Independent	-4.32×10^{-1}
	0.4	-2.41×10^5	Independent	-4.32×10^{-1}
Water/carbon tetrachloride	0.2	-7.78×10^4	Independent	-2.50×10^{-2}
	0.4	-4.09×10^5	Independent	-2.50×10^{-2}
Air/n-octane	0.2	-337	Independent	8.63×10^{-2}
	0.4	-1.07×10^4	Independent	8.63×10^{-2}
Benzene-mercury	0.2	-2.17×10^7	Independent	1.40×10^{-1}
	0.4	-3.66×10^8	Independent	1.40×10^{-1}
n-octanol/water	0.2	-1.44×10^4	1	-1.36
	0.2	1.04×10^5	16	-1.36
	0.2	4.6×10^5	32	-1.36
	0.4	-3.87×10^5	1	-1.36
	0.4	-1.45×10^2	16	-1.36
	0.4	1.17×10^6	32	-1.36

* Stable for $Ra < Ra_a$.

NOTATION

c = $i\sigma/\beta$, dimensionless
d = $d_a + d_b$, m
d_a = depth of upper fluid, m
d_b = depth of lower fluid, m
D = $\partial/\partial z$, m^{-1}
e = ν_a/ν_b, dimensionless
E = β_a/β_b, dimensionless
F = $(\rho_b - \rho_a) gd/\rho_a V_0^2$, dimensionless
g = acceleration of gravity, m/s^2
$\hat{j}$ = unit vector parallel to base flow, dimensionless
k = thermal conductivity, W//m · K
$\hat{k}$ = vertical unit vector, dimensionless
L = base temperature gradient, k/m
Ma = $\partial\gamma/\partial T\ L_a d^2/\kappa_a \mu_a$, dimensionless
p = dynamic pressure, N/m^2
Pe_a = dV_0/κ_a, dimensionless
Pe_b = dV_0/κ_b, dimensionless
Q = $\partial\gamma/\partial T/d^2 g\beta_a\rho_a$, dimensionless
r = d_b/d_a, dimensionless
Ra_a = $\beta_a g L_a d^4/\kappa_a\mu_a$, dimensionless
Ra_b = $\beta_b g L_b d^4/\kappa_b\mu_b$, dimensionless
Re_a = $V_0 d/\nu_a$, dimensionless
Re_b = $V_0 d/\nu_b$, dimensionless
s = k_a/k_b, dimensionless
S = $\gamma/V_0^2\rho_a d$, dimensionless
t = ρ_a/ρ_b, dimensionless; or time, s or dimensionless
T = base temperature, K
T_1 = temperature of upper boundary, K
T_2 = temperature of lower boundary, K
u = perturbation velocity (x direction), m/s
$\vec{u}'$ = perturbation velocity vector, m/s
v = perturbation velocity (y direction), m/s
V = base velocity, m/s
V_0 = velocity of upper boundary, m/s
w = perturbation velocity (z direction), m/s
x = κ_a/κ_b, dimensionless; or horizontal (perpendicular to flow), m
y = horizontal (streamwise) coordinate, m
z = vertical coordinate, m

Greek Letters

α = total wavenumber, dimensionless
β = wavenumber in y direction (without subscript), or coefficient of expansion, dimensionless
γ = surface tension, N/m
∇_1 = horizontal component of gradient operator, m^{-1}
∇_1^2 = horizontal component of Laplacian operator, m^{-2}
δ = wavenumber in x direction, dimensionless
η = surface deformation, m
θ = perturbation temperature, K
κ = thermal diffusivity, m^2/s
μ = viscosity, N · s/m^2
ν = kinematic viscosity, m^2/s
ρ = density, kg/m^3
σ = disturbance growth constant, dimensionless
φ = upper fluid stream function, dimensionless
χ = lower fluid stream function, dimensionless

Superscripts

$(')$ = a perturbation quantity before normal mode transformation, dimensionless

Subscripts

a = a property of the upper fluid, dimensionless
b = a property of the lower fluid, dimensionless
0 = zeroth-order expansion term, dimensionless
1 = first-order expansion term, dimensionless

LITERATURE CITED

Bernard, H., and D. Avsec, "Travaux Recents sur les Tourbillons Cellulaires et les Tourbillons en Bandes Applications a L'Astrophysique et a la Metearologie," *J. Phys. Radium*, **9**, 486 (1938).

Berg, J., "Interfacial Phenomena in Fluid Phase Separation Processes," in *Recent Developments in Separation Science, II*. N. Li (ed.), Chemical Rubber Co., Cleveland (1972).

———., A. Acrivos, and M. Boudart, "Natural Convection in Pools of Evaporating Liquids," *J. Fluid Mech.*, **24**, 721 (1966).

Brian, P. L. T., "Effect of Gibbs Adsorption on Marangoni Instability," *AIChE J.*, **17**, 765 (1971).

Brunt, D., "Experimental Cloud Formation" in *Compendium of Meterology*, Am. Meteorological Soc., Boston (1951).

Chandra, K., "Instability of Fluids Heated From Below," *Proc. Royal Soc. London*, **164A**, 231 (1938).

Charles, M. E., and L. U. Lilleleht, "An Experimental Investigation of Stability and Interfacial Waves in Co-Current Flow of Two-Liquids," *J. Fluid Mech.*, **22**, 217 (1965).

Clark, M., and C. King, "Evaporation Rates of Volatile Liquids in a Laminar Flow System," *AIChE J.*, **16**, 64 (1970).

Deardorff, J., "Gravitational Instability between Horizontal Plates with Shear," *Phys. Fluids*, **8**, 1027 (1965).

Gage, K., and W. Reid, "The Stability of Thermally Stratified Plane Poiseuille Flow," *J. Fluid Mech.*, **33**, 21 (1968).

Gallagher, A., and A. Mercer, "On the Behavior of Small Disturbances in Plane Couette Flow with a Temperature Gradient," *Proc. Royal Soc. London*, **286A**, 117 (1965).

Graham, A., "Shear Patterns in an Unstable Layer of Air," *Phil. Trans. Roy Soc. London*, **232A**, 285 (1933).

Ingersoll, A., "Convective Instabilities in Plane Couette Flow," *Phys. Fluids*, **9**, 682 (1966).

Joseph, D., "Nonlinear Stability of the Boussinesq Equations by The Method of Energy," *Arch. Rat. Mech. Anal.*, **22**, 163 (1966).

Linde, H., and E. Schwarz, "Über groBräumige Rollzellen der freien Grenzflächenkonvecktien," *Mber dt. Akad. Wiss. Berl.*, **6**, 330 (1964).

Maroudas, N., and H. Sawistowski, "Simultaneous Transfer of Two Solutes across Liquid-Liquid Interfaces," *Chem. Eng. Sci.*, **19**, 919 (1964).

Nield, D., "Surface Tension and Buoyancy Effects in Cellular Convection," *J. Fluid Mech.*, **19**, 341 (1964).

Palmer, H., and J. Berg, "Convective Instabilities in Liquid Pools Heated From Below," *ibid.*, **47**, 779 (1971).

———., "Hydrodynamic Stability of Surfactant Solutions Heated from Below," *ibid.*, **51**, 385 (1972).

Pearson, J., "On Convection Cells Induced by Surface Tension," *ibid.*, **4**, 489 (1958).

Phillips, A., and G. Walker, "The Forms of Stratified Clouds," *Quart. J. R. Meteor. Soc.*, **58**, 23 (1932).

Sawistowski, H., "Interfacial Phenomena" in "Recent Advances in Liquid-Liquid Extraction," C. Hanson (ed.), Pergamon Press, Oxford (1971).

Scriven, L., and C. Sternling, "On Cellular Convection Driven by Surface-Tension Gradients," *J. Fluid Mech.*, **19**, 321 (1964).

Smith, K., "On Convective Instability Induced by Surface-Tension Gradients," *ibid.*, **24**, 401 (1966).

Squire, H., "On the Stability for Three-Dimensional Disturbances of Viscous Fluid Flow Between Parallel Walls," *Proc. Roy. Soc. London*, **A142**, 621 (1933).

Sternling, C., and L. Scriven, "Interfacial Turbulence: Hydrodynamic Instability and the Marangoni Effect," *AIChE J.*, **5**, 514 (1959).

Stuart, J. T., "Hydrodynamic Stability" in *Laminar Boundary Layers* (L. Rosenhead, ed.), Oxford University Press (1963).

Yih, C., "Instability due to Viscosity Stratification," *J. Fluid Mech.*, **27**, 337 (1967).

Zeren, R., and W. Reynolds, "Thermal Instabilities in Two-Fluid Horizontal Layers," *ibid.*, **53**, 305 (1972).

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