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Finite amplitude cellular convection induced by surface tension

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A non-linear analysis of cellular convection driven by surface tension in a semi-infinite liquid layer heated from below has been made. The purpose is to determine whether or not one can predict the emergence of the hexagonal flow pattern from the interaction of a certain large class of important disturbances. The principal conclusion is that, compared with gravity driven convection, there is generally a much greater band of imposed temperature difference associated with hexagonal convective patterns. Partial results for the more realistic assumption of finite depth support this conclusion.

1. Introduction

In the past fifty years, beginning with Lord Rayleigh's analysis of the phenomenon, a great deal of effort has been expended treating cellular convection as a buoyancy driven flow. Despite generally impressive qualitative agreement between theory and various observations, this approach has not been satisfactory in describing Bénard's classic experiments which motivated the original theoretical investigations. Working with thin horizontal liquid layers heated from below, with the upper surface free to the atmosphere, Bénard observed that motion occurred only when a critical temperature gradient was exceeded, and that the stable secondary flow pattern was one of contiguous hexagonal cells.

The inappropriateness of Rayleigh's model to Bénard's experiments was not adequately explained until more recent experimental and analytical studies by Block (1956), Pearson (1958) and Nield (1964) showed that, rather than being a buoyancy driven flow, Bénard cells are primarily induced by the surface tension gradients resulting from temperature variations across the free surface. Nield accounted for both mechanisms in his analysis and found that at the onset of convection the driving force for the motion is approximately equal to the sum of the surface and buoyancy forces in the layer and, furthermore, as the depth of the layer decreases the surface tension mechanism becomes more dominant. For the thin (about 1 mm) layers used in Bénard's experiments the flow is due mainly to surface tension effects.

Although it does not describe Bénard's work, Rayleigh's theory is an appropriate model for similar situations where there is no free surface or, for most fluids, when the fluid layer is thicker than about 1 cm.

All the theoretical investigations of the surface mechanism cited above are linear stability analyses. The linear theory predicts the critical temperature gradient at which motion first occurs and the wave-number of the fastest growing disturbance. It does not specify the final size or shape of the cell or how the initial growing disturbances reach a finite steady-state amplitude. This information is essential if one is to understand the secondary steady flow field and can only be obtained by considering the non-linear theory.

The purpose of our investigation is to elucidate the non-linear behaviour of the surface tension mechanism. The problem of cell size is not considered here. We are concerned with the question of whether or not one can predict the emergence of the hexagonal flow pattern from the interaction of a certain large class of important disturbances. Since the calculations required for a non-linear analysis of this type can be inordinately long, we have used as our primary model the limiting case of infinite Prandtl number and semi-infinite liquid phase.

It may seem surprising that a semi-infinite model is used to describe a thin layer of liquid. A two-part argument supports this step. (i) In very thin layers convection is driven primarily by surface forces so, to a first approximation, buoyancy effects may be neglected. (ii) Consider then an instability due entirely to surface tension gradients. By continuity, surface motion requires motion of the adjacent bulk phase. Replacing a layer of finite thickness by a semi-infinite layer will only roughly model the bulk motion. However, qualitative agreement can still be expected because the surface motion which drives the flow is well modelled. Properly interpreted the semi-infinite model should retain the essential characteristics of the physical problem. This is confirmed by our partial results for the finite depth layer.

The analysis is carried out using a modified successive approximation technique based on the Stuart–Watson (1960) approach to non-linear stability problems. The particular formulation given here is explained in detail by Segel (1965*a*) for a model equation and has been applied to the problem of buoyancy driven convection by Davis & Segel (1965). In adapting these methods to the surface dominated problem the chief mathematical difficulty is connected with the fact that in the linearized problem the eigenvalue appears in the surface boundary condition.

2. Formulation

Consider a semi-infinite horizontal liquid layer which is unbounded in the horizontal (x, y) directions and extends to $z = -\infty$ in the vertical z direction. The undisturbed system whose stability we shall study is a quiescent fluid with a free surface at $z = 0$ and a constant temperature gradient, $-\beta$, in the z direction. The perturbation temperature is T, the perturbed surface is given by $z = \xi(x, y)$ and (u, v, w) are velocity components in the (x, y, z) directions.

Physical variables are scaled using d, d^2/κ, κ/d and βd as the length, time, velocity and temperature scale factors respectively. Here κ is thermal diffusivity and d is the characteristic length usually taken to be the depth of the layer. Because of the infinite depth in this model no specific length scale is chosen, d being left arbitrary for now.

The stability problem is formulated in the usual manner. The solution to the general equations governing the system is assumed to be the quiescent solution plus a perturbation. Substitution into the full momentum, energy and continuity equations gives the non-linear partial differential equations for the perturbation quantities,

$$\nabla^4 w = N_{Pr}^{-1}\left[\frac{\partial}{\partial t}\nabla^2 w + \nabla_1^2 N(w) - \frac{\partial^2}{\partial x\,\partial z}N(u) - \frac{\partial^2}{\partial y\,\partial z}N(v)\right], \tag{1}$$

$$\nabla^2 T + w = \frac{DT}{Dt}, \tag{2}$$

where $\nabla_1^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2$, $\nabla = (\partial/\partial x, \partial/\partial y, \partial/\partial z)$, $\bar{u} = (u, v, w)$,

$\nabla^2 = \nabla_1^2 + \partial^2/\partial z^2$, $N = \bar{u}\,.\,\nabla$, $D/Dt = \partial/\partial t + N$, N_{Pr} = Prandtl number.

The perturbation equations are presented in the form which best lends itself to an iteration scheme based on the linear theory. Equation (1) is obtained by combining the momentum and continuity equations to eliminate u and v from the linear terms and to eliminate pressure entirely. Other eliminations give the expressions needed to determine u and v, namely:

$$\nabla^2\left(\nabla_1^2 u + \frac{\partial^2}{\partial z\,\partial x}w\right) = N_{Pr}^{-1}\left[\frac{\partial}{\partial t}\left(\nabla_1^2 u + \frac{\partial^2}{\partial z\,\partial x}w\right) + \frac{\partial^2}{\partial y^2}N(u) - \frac{\partial^2}{\partial x\,\partial y}N(v)\right], \tag{3}$$

$$\nabla^2\left(\nabla_1^2 v + \frac{\partial^2}{\partial z\,\partial y}w\right) = N_{Pr}^{-1}\left[\frac{\partial}{\partial t}\left(\nabla_1^2 v + \frac{\partial^2}{\partial z\,\partial y}w\right) + \frac{\partial^2}{\partial x^2}N(v) - \frac{\partial^2}{\partial x\,\partial y}N(u)\right]. \tag{4}$$

In the analysis it turns out that the terms on the right side of (3) and (4) do not contribute to the solution for the disturbances considered in the iteration scheme and can be neglected. Therefore $(\nabla_1^2 u + w_{xz})$ and $(\nabla_1^2 v + w_{yz})$ are harmonic. Using the continuity equation and boundary conditions these functions can be shown to be zero at a rigid surface and to have zero normal derivatives at the free surface. Assuming appropriate boundedness of the solutions everywhere in the layer, it follows from the uniqueness of such harmonic functions that

$$\nabla_1^2 u = -w_{zx}, \tag{5}$$

$$\nabla_1^2 v = -w_{zy}. \tag{6}$$

It is clear from inspection of (1) that the calculations could be appreciably simplified if N_{Pr} was taken to be infinite. Preliminary calculations assuming a finite N_{Pr} and experience with other convection problems indicate that N_{Pr} typically appears in the combination $N_{Pr}(1+N_{Pr})^{-1}$. We therefore expect the infinite Prandtl number solution to be a good approximation for $N_{Pr} > 5$. Most liquids used in experiments are within this range; many have Prandtl numbers in the hundreds, or even thousands. For the present, therefore, we shall use the infinite Prandtl number equations (see Bray 1966):

$$\nabla^4 w = 0, \tag{7}$$

$$\nabla^2 T + w = DT/Dt, \tag{8}$$

along with (5) and (6) for u and v. Now the only non-linear terms are those appearing in the energy equation.

For a surface driven flow an essential feature of the formulation is the establishment of boundary conditions at the interface $z = \xi(x, y)$. Pearson first formulated and analysed the surface tension mechanism for the Bénard problem using a simple idealized model, a non-deforming free surface with a general heat transfer condition. Subsequent investigations have extended Pearson's model to include the effects of surface deformation, elasticity, and viscosity and the dynamics of the upper phase. Although there are situations in which such effects would greatly influence stability, these linear analyses indicate that for most experiments the upper phase, surface viscosity, and surface elasticity have little effect on the critical conditions at marginal stability. In a qualitative study of the non-linear problem it is justifiable to neglect them.

The consideration of surface deflexion, on the other hand, is important for two reasons. First, it has a destabilizing effect on the system and, secondly, it gives a criterion for determining the driving mechanism. A major conflict between Rayleigh's theory and experiments concerned the flow field. Bénard observed warm fluid rising below the depressions in the free surface, whereas Jeffreys (1951) proved that in a buoyancy driven flow the free surface over rising fluid is elevated. Davis (1964) and Sternling & Scriven (1964) independently explained this conflict by showing that for surface driven flows the free surface above a rising current is depressed as the experiments indicate.

For our model the infinite N_{Pr} assumption, which linearizes the momentum equation, also results in a simplification of the boundary conditions. It can be verified that, because the momentum equation is linear and the depth infinite, the surface deformation $\xi(x, y)$ is zero at all orders of perturbation. Therefore Pearson's formulation will be used to describe the free surface of the semi-infinite layer; only his notation is changed.

Since the surface heat transfer process is not easily describable, Pearson assumed a constant heat transfer coefficient N_{Nu} and considered the general condition

$$T_z + N_{Nu} T = 0 \quad \text{at} \quad z = 0.$$

He found that the stability of the system increases slowly with N_{Nu} and estimated that for Bénard's experiments $N_{Nu} \ll 1$. For our purposes it suffices to take $N_{Nu} = 0$. The kinematic condition for a non-deforming surface is $w = 0$.

A third boundary condition is found by making a horizontal force balance at the surface. Surface tension S is assumed to vary linearly with temperature: $S = S_0 - \sigma T$. Equating the net surface tension force due to temperature variations with the viscous shear force at the surface gives, after some manipulation with the continuity equation,

$$\frac{\partial^2 w}{\partial z^2} = N_{Ma} \nabla_1^2 T \quad \text{at} \quad z = 0,$$

$$N_{Ma} = \sigma \beta d^2 / \rho \nu \kappa.$$

The Marangoni number N_{Ma} contains the undisturbed temperature gradient β and therefore its magnitude will govern the stability of the system. Since σ is positive for most liquids, a positive N_{Ma} would correspond to a negative gradient, $-\beta$, in the undisturbed system. When, as here, the upper phase is ignored, instability of the motionless layer is expected for a sufficiently large positive Marangoni number (Smith 1966).

Surface deformation ξ is zero; consequently, the normal surface stress condition is automatically satisfied. The boundary conditions are completed by taking w and the derivatives of w and T to be bounded as $z \to -\infty$.

3. Solution

To solve the non-linear problem we shall utilize the successive approximation method mentioned above. This is most efficiently done if one writes the steady-state linearized equations in operator form as an eigenvalue problem with N_{Ma} as the eigenvalue. The reader is referred to Friedman (1956) for a vector formulation of a boundary-value problem, with an ordinary differential equation, having the eigenvalue in one of the boundary conditions. Generalizing to a system of partial differential equations, let the vector $\mathbf{U}$ be a 3-vector whose first two components are functions of the space variables and time and whose third component, a function of x, y and t, is evaluated at the surface:

$$\mathbf{U} = \begin{bmatrix} w(x,y,z,t) \\ T(x,y,z,t) \\ T(x,y,z=0,t) \end{bmatrix}. \tag{9}$$

We define an inner product of two such vectors

$$\mathbf{A} = (a_1, a_2, a_3) \quad \text{and} \quad \mathbf{B} = (b_1, b_2, b_3)$$

as
$$(\mathbf{A},\mathbf{B}) = \lim_{L\to\infty} \frac{1}{4L^2} \int_{-L}^{L} \int_{-L}^{L} dx\, dy \left[a_3 b_3 + \int_{-\infty}^{0} dz (a_1 b_1 + a_2 b_2) \right]. \tag{10}$$

For periodic cellular motion, the (x,y) integration may be taken over a single cell.

Our problem can now be considered as the determination of the solution to

$$\begin{bmatrix} \nabla^4 w \\ \nabla^2 T + w \\ w_{zz}(z=0) \end{bmatrix} = \begin{bmatrix} 0 \\ DT/Dt \\ 0 \end{bmatrix} + N_{Ma} \begin{bmatrix} 0 \\ 0 \\ \nabla_1^2 T\,(z=0) \end{bmatrix} \tag{11}$$

subject to the boundary conditions

$$\left. \begin{array}{ll} w = T_z = 0 & \text{at} \quad z = 0, \\ w, w_z, T_z & \text{bounded as} \quad z \to -\infty. \end{array} \right\} \tag{12}$$

We define the operators $\mathscr{L}$, $\mathscr{M}$ and $\mathscr{N}$ in the obvious way so that the vector equation (11) may be written

$$\mathscr{L}(\mathbf{U}) = \mathscr{N}(\mathbf{U}) + N_{Ma} \mathscr{M}(\mathbf{U}), \tag{13}$$

where $\mathscr{L}(\mathbf{U})$ contains all linear steady-state terms and $\mathscr{N}(\mathbf{U})$ contains all non-linear terms and time derivatives.

A linear analysis can be considered as the first step in any stability theory. By applying the principle of exchange of stabilities (see Vidal & Acrivos 1966) and assuming infinitesimal disturbances we can neglect $\mathscr{N}(\mathbf{U})$ in equation (13), thereby obtaining the linear problem for marginal stability ($\partial/\partial t = 0$):

$$\mathscr{L}(\mathbf{U}_1) - N_{Ma}^c \mathscr{M}(\mathbf{U}_1) = 0, \tag{14}$$

subject to the boundary conditions (12). The characteristic equation for this eigenvalue problem specifies the critical Marangoni number N^c_{Ma} and critical wave number α_c. According to linear theory, if $N_{Ma} > N^c_{Ma}$, the quiescent layer is unstable and the fastest growing disturbance has a wave number α_c.

Solutions to the non-linear problem, which we expect to be small for all time when $|N_{Ma} - N^c_{Ma}|$ is sufficiently small, can be obtained by means of an iteration scheme with the eigenvector $\mathbf{U}_1$ as the first approximation. Equation (13) is rearranged and written as

$$[\mathscr{L} - N^c_{Ma}\mathscr{M}](\mathbf{U}_{n+1}) = [\mathscr{N} + (N_{Ma} - N^c_{Ma})\mathscr{M}](\mathbf{U}_n) \quad (n = 1, 2, \ldots). \tag{15}$$

Boundary conditions (12) are imposed at each stage and equations (5) and (6) are used to determine u and v in $\mathscr{N}(\mathbf{U}_n)$.

The main point of interest is whether or not hexagonal cells are a stable secondary flow. Consequently, the initial disturbance is taken to be proportional to a function ϕ which contains two primary horizontal space modes of the same overall wave-number α; namely,

$$\phi = Z(t)\cos\alpha y + Y(t)\cos\tfrac{1}{2}\sqrt{3}\,\alpha x\cos\tfrac{1}{2}\,\alpha y.$$

It is known (see Chandrasekhar 1961) that hexagonal cells occur if the amplitude functions $Y(t)$ and $Z(t)$ have equilibrium solutions which satisfy the relationship $Y = \pm 2Z$. If this relationship can be shown ultimately to hold regardless of the initial values of Y and Z, then a step will have been taken in predicting the emergence of hexagonal cells.

Solutions to the linear problem (14) are assumed to be of the form

$$\mathbf{U}_1 = \begin{bmatrix} w_1(z) \\ T_1(z) \\ T_1(z=0) \end{bmatrix} \phi(x, y, t),$$

where

$$\nabla_1^2\phi + \alpha^2\phi = 0.$$

Separating variables reduces (14) to a pair of ordinary differential equations in z for w_1 and T_1:

$$(D^2 - \alpha^2)^2 w_1 = 0, \quad D \equiv d/dz,$$

$$(D^2 - \alpha^2) T_1 + w_1 = 0,$$

$$D^2 w_1 = -\alpha^2 N^c_{Ma} T_1, \quad \text{at} \quad z = 0.$$

The solution is, after imposing the boundary conditions (12),

$$w_1 = Cz\,e^{\alpha z}, \quad T_1 = -C(4\alpha^3)^{-1}(\alpha^2 z^2 - \alpha z + 1)\,e^{\alpha z}.$$

C is an arbitrary constant which we shall incorporate into the amplitudes Y and Z in $\mathbf{U}_1$.

The critical Marangoni number is given by $N^c_{Ma} = 8\alpha^2$. According to linear theory then, a disturbance with $\alpha_c = 0$, corresponding to infinite wavelength, is always unstable. That the critical wavelength is infinite is not surprising since from experimental observation one expects a critical wavelength comparable to the depth of the layer.

An interpretation of the critical Marangoni number for a semi-infinite layer which permits quantitative comparison with results for a layer having finite depth can be made by using the disturbance wavelength as the length scale d. Since overall cell wavelength and layer depth are expected to be of the same magnitude, similar results should be obtained for both models when scaled in this manner. In dimensional variables the wave-number and wavelength are a and λ respectively. For the linear analysis above, the dimensionless wave-number α was used, where $\alpha = ad$. With a length scale d given by

$$d = a^{-1} = \lambda/2\pi,$$

N^c_{Ma} based on the disturbance wavelength is

$$N^c_{Ma} = (\sigma\beta\lambda^2/\rho\nu\kappa) = 32\pi^2$$

for the semi-infinite layer. The calculations for a layer of finite depth (see below) give, at the critical wavelength λ_c,

$$N^c_{Ma} = (\sigma\beta\lambda_c^2/\rho\nu\kappa) = 80\pi^2.$$

Our infinite depth result of $32\pi^2$ is an underestimate, as would be expected for a model which ignores the stabilizing effect of the lower rigid boundary.

Corrections to the linear theory are found by solving the linear inhomogeneous equation (15) for $\mathbf{U}_{n+1}$ using the method of undetermined coefficients. It is well known that if solutions to this equation are to exist, the inhomogeneous term $[\mathcal{N} + (N_{Ma} - N^c_{Ma})\mathcal{M}](\mathbf{U}_n)$ must be orthogonal to the eigenfunctions $\mathbf{U}^*$ of the linear adjoint problem (see appendix for details). Until now no conditions have been imposed on the amplitude functions $Y(t)$ and $Z(t)$ but it turns out that the existence condition requires that they satisfy

$$Y' = \epsilon Y - \gamma YZ - RY^3 - PYZ^2 + \ldots, \tag{16a}$$

$$Z' = \epsilon Z - \tfrac{1}{4}\gamma Y^2 - R_1 Z^3 - \tfrac{1}{2}PY^2Z + \ldots, \tag{16b}$$

$$P = 4R - R_1, \quad ' = d/dt.$$

This pair of coupled ordinary differential equations describes the behaviour in in time of the orginal disturbance.

If ϵ and γ are small, the amplitude equations can be truncated at third order, the higher order terms being negligible. The qualitative behaviour of solutions to (16) is discussed in the appendix.

4. Conclusions

The constants ϵ, γ, P, R and R_1 are fixed by the existence condition. For the case of infinite depth and infinite Prandtl number they are

$$\left.\begin{array}{ll} \epsilon = \tfrac{1}{8}(N_{Ma} - N^c_{Ma}), & R = (0{\cdot}02970)\alpha^{-2}, \\ \gamma = (0{\cdot}1296), & R_1 = (0{\cdot}05745)\alpha^{-2}, \\ P = (0{\cdot}06135)\alpha^{-2} & N^c_{Ma} = 8\alpha^2. \end{array}\right\} \tag{17}$$

If $(N_{Ma} - N^c_{Ma})$ is small, inspection of (16) and (17) indicates that the conditions necessary for truncation are met.

Since $\alpha_c = 0$, it is at first unsettling that the factor α^{-2} appears in the third-order coefficients given in (17). However, we have seen that the appropriate length scale for the semi-infinite model is the disturbance wavelength. The quantity α^{-2} disappears when this length scale is used.

The form of the amplitude equations obtained here is exactly that obtained by Segel & Stuart (1962) in their analysis of buoyancy driven flows. More recently Segel (1965*b*) showed that once the coefficients in equation (16) are obtained, one can determine the final equilibrium states which result from the interaction of any finite number of modes associated with the same overall wave-number (see appendix).

	↑
Rolls	$196 < \Delta N_{Ma}$
Hexagons, rolls	$64 < \Delta N_{Ma} < 196$
Hexagons	$0 < \Delta N_{Ma} < 64$
Hexagons, no motion	$-0{\cdot}023 < \Delta N_{Ma} < 0$
No motion	$\Delta N_{Ma} < -0{\cdot}023$

$$\Delta N_{Ma} = (N_{Ma} - N^c_{Ma})/N^c_{Ma}$$

TABLE 1. Stable flows for various ranges of N_{Ma} (infinite Prandtl number, infinite depth)

Segel's results, restated for the surface driven flow, are given in table 1. The stable equilibrium solutions are shown for different ranges of the Marangoni number. Below the solid line at $|N_{Ma} - N^c_{Ma}| = 0$ the motionless state is stable according to linear theory. 'Rolls' refers to a two-dimensional pattern of vortices whose regularly spaced axes are parallel to each other and to the bounding planes. In ranges where two cell patterns are possible the steady flow field would be determined by the initial conditions.

From table 1 a principal formal conclusion of our analysis is that the hexagonal pattern is stable for a range of Marangoni number from just below critical to 64 times critical. Referring to (13) it is clear that the iteration scheme used to solve the non-linear problem requires sufficiently small non-linear terms and hence sufficiently small $(N_{Ma} - N^c_{Ma})$ for convergence. The supercritical ranges given in table 1 appear to be well beyond the limits of the analysis. Taking into account the probable non-convergence of our iteration scheme at large $(N_{Ma} - N^c_{Ma})$ the principal genuine conclusion is that hexagonal cells should be the convective pattern observed for a range of Marangoni number extending 'considerably' above critical. We also note that, just as in gravity driven convection when fluid property variation is considered, there is only an extremely narrow range of N_{Ma} in which sufficiently large disturbances grow although linear theory predicts stability.

In order to check the effect of the surface heat transfer coefficient N_{Nu} and Prandtl number N_{Pr} on the coefficients of the amplitude equations a one disturbance analysis, $Y(t) \equiv 0$, to third order with finite N_{Pr}, and a two disturbance analysis to second order with $N_{Nu} \neq 0$ and N_{Pr} finite were made on the semi-infinite model.

For
$$\phi = Z(t)\cos\alpha y, \quad N_{Pr}^{-1} \neq 0 \quad \text{and} \quad N_{Nu} = 0,$$
$$\epsilon = \tfrac{1}{4}(N_{Ma} - N_{Ma}^{c})(2 + N_{Pr}^{-1})^{-1}, \quad \gamma = 0,$$
$$R_1 = \tfrac{1}{3}(4^6\alpha^2)^{-1}(2 + N_{Pr}^{-1})^{-1}(218N_{Pr}^{-2} + 196N_{Pr}^{-1} + 1412).$$
For
$$\phi = Z(t)\cos\alpha y + Y(t)\cos\tfrac{1}{2}\sqrt{3}\,\alpha x\cos\tfrac{1}{2}\alpha y, \quad N_{Nu} \neq 0 \quad \text{and} \quad N_{Pr} \neq 0,$$
$$\epsilon = \tfrac{1}{4}(N_{Ma} - N_{Ma}^{c})(1 + [N_{Nu}\alpha^{-1} + 1][N_{Pr}^{-1} + 1])^{-1},$$
$$\gamma = (1/3^3)(7 - 2N_{Pr}^{-1})(N_{Nu}\alpha^{-1} + 1)(1 + [N_{Nu}\alpha^{-1} + 1][N_{Pr}^{-1} + 1])^{-1}.$$

It appears that if $N_{Pr} > 5$ and N_{Nu} is small compared to the wave-number then the simplifying assumptions $N_{Pr} = \infty$ and $N_{Nu} = 0$ will not appreciably affect the final results.

Originally our purpose was first to obtain a qualitative understanding of non-linear surface driven convection by a study of the semi-infinite layer and then to obtain quantitative results with a computer-aided analysis of a layer of finite thickness. It was clear from the outset that the required machine program would be very lengthy, so we deemed it all the more important to begin with an analytic approach whose results could serve as a check on the machine calculations.

We recently learned that R. Sani of the University of Illinois is carrying out an analysis of the finite depth layer at least as comprehensive as that which we contemplated. We therefore will be content with presenting qualitative conclusions. With this in mind we turn to the finite depth layer for which we have obtained results through second order. While hand computations for the full problem are forbidding, the second-order computations are not unduly long and were carried out to get some idea as to whether our earlier results would be greatly modified and as a further check on future computer programs.

For the finite layer the depth is used as the scale factor d. The boundary conditions at the rigid plate $z = -1$ are taken to be $w = w_z = T = 0$. It is assumed that $N_{Nu} = N_{Pr}^{-1} = 0$. As is the case for commonly used fluids, we assume that surface deflexion can be neglected (see below).

The analytic expressions for the coefficients ϵ and γ are such long and involved functions of α that it was necessary to evaluate them at the critical wave-number in order to get an idea of their magnitude. It should be mentioned here that in these calculations subtraction of nearly equal numbers seems unavoidable so a large number of significant figures must be carried in order to obtain any degree of accuracy.

The marginal stability curve is given by the expression
$$N_{Ma} = \frac{8\alpha^2(\alpha - \cosh\alpha\sinh\alpha)\cosh\alpha}{(\alpha^3\cosh\alpha - \sinh^3\alpha)},$$
which has a minimum value of $N_{Ma}^{c} = 80$ at approximately $\alpha = 2$. Letting $\alpha_c = 2$ we find $\epsilon = 0{\cdot}07565(N_{Ma} - N_{Ma}^{c})$, $\gamma = 0{\cdot}056108$. There is quantitative but no qualitative change compared to the results for the semi-infinite layer so we still expect hexagonal cells to be a stable flow for a considerable range of supercritical Marangoni numbers.

Other conclusions from our analysis are that fluid rises at the cell centres as observed in experiments and that there is an extremely small range of temperatures where the quiescent layer is unstable to sufficiently large disturbances although small disturbances decay (subcritical instability).

5. Comments: other papers

Surface driven flows are encountered in many practical situations involving heat and mass transfer for which the Bénard problem is a useful prototype (see Scriven & Sternling 1960). For this reason the relevant linear stability theory has received much attention recently. It was acknowledged above that Pearson's model was an idealization of a very complex situation. Recent investigators have considered more general and realistic surface conditions.

The most restrictive assumption of Pearson's model was felt to be that of a non-deforming free surface. Sternling & Scriven (1964) allowed the surface to deform, accounting for capillary waves but not gravity waves. The case of zero wave-number was found to be always unstable and, furthermore, there was no critical Marangoni number below which all disturbances decay. These disturbing results were clarified by Smith's (1966) more comprehensive study of surface curvature in which both capillary and gravity waves were considered. It was shown that gravity has a stabilizing effect at small α and there was a critical N_{Ma}. When the surface deformation is small, both studies indicate that Pearson's results are changed only at very small α and they accurately predict the critical conditions. However, if the surface deformation is appreciable, the nature of the solution is completely different from Pearson's.

Smith also accounted for the dynamics of the upper phase and found that it was possible to have instability with respect to heat transfer in either direction, a result which Block observed experimentally and which Pearson's model does not allow for.

Berg & Acrivos (1965) accounted for surface active agents and demonstrated their strong stabilizing effect on surface tension induced convection. Adding surfactants can increase the critical N_{Ma} several orders of magnitude.

It can be concluded that Pearson's analysis adequately predicts the critical conditions at the onset of instability for most experiments with thin layers (1 mm) of ordinary liquids. It is also clear that for certain liquids or for very small layer depth, a more careful description of the surface mechanism is necessary.

All non-linear analyses of the Bénard problem have, until now, used Rayleigh's model. It is informative to consider this work here because of the physical and mathematical similarities between the two problems.

The amplitude equations obtained for both problems are of the same form; only the value of the coefficients differ. For buoyancy driven convection the linear growth rate constant ϵ is proportional to $(N_{Ra} - N_{Ra}^c)$ where N_{Ra} is the Rayleigh number. The second-order coefficient γ controls the development of the flow. Hexagonal cells cannot be a stable flow pattern unless γ is non-zero. It has been shown (Palm 1960; Segel & Stuart 1962; Davis & Segel 1965) that second-order terms do not appear for a buoyancy driven flow unless fluid property variations with temperature (other than linear density variations) or surface deflexion are considered. For a layer of finite depth the coefficient γ is proportional to the usually slight property variation and surface deflexion effect. Segel estimated that in the most favourable situations, which require thin fluid layers and/or large property variations, hexagonal cells would be stable just a few percent above critical. Even when property variations and surface deflexion are accounted for, γ is zero for the semi-infinite model. Stable hexagonal cells are not possible for this limiting case.

No photographs of truly hexagonal patterns have yet been published for layers confined between rigid boundaries, a system in which the flow is buoyancy driven. However, such patterns should appear under appropriate experimental conditions. They have been observed by R. Krishnamurti of UCLA (unpublished) when the mean temperature varies slowly.

Two experimentalists have obtained very regular hexagonal cell patterns, Bénard (see Chandrasekhar 1961) and Koschmieder (1966). In both cases the upper surface was free. As we have already remarked, the flows Bénard observed were dominated by surface tension. In Koschmieder's experiments, although it was not easy to determine the location of elevations and depressions, the free surface appeared to be depressed above rising columns so here, too, surface tension dominated. Koschmieder (private communication) did not observe the theoretically predicted transition from hexagonal cells to rolls even though he increased the applied temperature difference as much as he could (two or three times critical). This can be taken as evidence that even our highly idealized model captured the essence of the phenomenon, the presence of a hexagonal pattern in surface driven convection for a range of temperature gradients large compared to the corresponding range for gravity driven convection.

More experimental checks on the theory are feasible and desirable.

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Appendix

Existence condition

The vector space of our problem is defined by (9) and (10). Using this notation we can state the existence condition for equation (15) in the following way. Since $(\mathscr{L} - N^c_{Ma}\mathscr{M})$ is a linear homogeneous operator, the inhomogeneous equation

$$(\mathscr{L} - N^c_{Ma}\mathscr{M})(\mathrm{U}_{n+1}) = \mathscr{F}(\mathrm{U}_n)$$

will have a solution only if $\mathscr{F}(\mathrm{U}_n)$ is orthogonal to the solution U^* of the adjoint problem; that is, $[\mathrm{U}^*, \mathscr{F}(\mathrm{U}_n)] = 0$. This requirement fixes the constants in the amplitude equation.

The adjoint operators and boundary conditions are defined by the requirement that for all $\mathbf{U}$ whose components satisfy (12) and all $\mathbf{U}^*$ whose components satisfy the adjoint boundary conditions we have

$$[(\mathscr{L}-N_{Ma}\mathscr{M})(\mathbf{U}),\mathbf{U}^*] = [\mathbf{U},(\mathscr{L}^*-N_{Ma}\mathscr{M}^*)(\mathbf{U}^*)].$$

They can be determined by using equation (10) and Green's theorem:

$$\iiint_V [(\psi\nabla^2\theta-\theta\nabla^2\psi)\,dV = \iint_S (\psi\,\partial\theta/\partial n-\theta\,\partial\psi/\partial n)\,dS,$$

where $\partial/\partial n$ denotes the derivative in the exterior direction normal to the boundary of V. If the adjoint eigenvector is taken as $\mathbf{U}^* = [w^*,T^*,w^*(z=0)]$ with the condition $\nabla_1^2\mathbf{U}^*+\alpha^2\mathbf{U}^* = 0$, the adjoint problem is

$$\begin{bmatrix} \nabla^4 w^*+T^* \\ \nabla^2 T^* \\ (T_z^*+N_{Nu}T^*)\,(z=0) \end{bmatrix} = N_{Ma}\begin{bmatrix} 0 \\ 0 \\ -\nabla_1^2 w_z^*\,(z=0) \end{bmatrix},$$

$$w^* = w_{zz}^* = 0 \quad \text{at} \quad z=0,$$

$$w^*, T^* \text{ and their derivatives bounded as } z\to-\infty.$$

For the semi-infinite model the adjoint solution is

$$T^* = e^{\alpha z}\phi(x,y),$$
$$w^* = -(8\alpha^3)^{-1}(\alpha z^2-z)\,e^{\alpha z}\phi(x,y).$$

The boundary conditions at the plate for the finite depth case are

$$w^* = w_z^* = T^* = 0 \quad \text{at} \quad z=-1.$$

Second-order solution

The second-order solution $\mathbf{U}_2$ is determined by solving

$$(\mathscr{L}-N_{Ma}^c\mathscr{M})(\mathbf{U}_2) = (\mathscr{N}+[N_{Ma}-N_{Ma}^c]\mathscr{M})(\mathbf{U}_1)$$

with the boundary conditions (12). Referring to $\mathbf{U}_1$ the inhomogeneous term can be written

$$\begin{bmatrix} 0 \\ T_1\phi_1+(w_{1z}T_1)(\phi_5+\tfrac{1}{2}\phi_2-\tfrac{1}{2}\phi_3-\phi_4)+(w_1T_{1z})(\phi_5+\phi_4+\phi_3+\phi_2) \\ (N_{Ma}-N_{Ma}^c)(4\alpha)^{-1}\phi_0 \end{bmatrix},$$

where

$$\phi_0 = Z(t)\cos\alpha y+Y(t)\cos m\alpha x\cos n\alpha y,$$
$$\phi_1 = Z'(t)\cos\alpha y+Y'(t)\cos m\alpha x\cos n\alpha y,$$
$$\phi_2 = \tfrac{1}{4}Y^2\cos\alpha y+YZ\cos m\alpha x\cos n\alpha y,$$
$$\phi_3 = \tfrac{1}{4}Y^2\cos 2m\alpha x+YZ\cos m\alpha x\cos 3n\alpha y,$$
$$\phi_4 = \tfrac{1}{2}Z^2\cos 2\alpha y+\tfrac{1}{4}Y^2\cos 2m\alpha x\cos\alpha y,$$
$$\phi_5 = \tfrac{1}{2}Z^2+\tfrac{1}{4}Y^2,$$
$$m = \tfrac{1}{2}\sqrt{3}, \quad n = \tfrac{1}{2}.$$

U_2 is easily found by the method of undetermined coefficients to be

$$w_2 = D_1 z\, e^{2m\alpha z}\phi_3 + D_2 z\, e^{2\alpha z}\phi_4,$$
$$T_2 = \sum_{r,p,s} D_{psr} z^r e^{s\alpha z}\phi_p,$$
$$p = 1,2,3,4,5 \quad r = 0,1,2,3, \quad s = 1,2,2m.$$

	↑ $\epsilon \sim (N_{Ra} - N_{Ra}^c)$
$\gamma^2(4R+R_1)Q^{-2}$	Roll
$\gamma^2 R_1 Q^{-2}$	Hexagon, roll
0	Hexagon
$-\gamma^2(4T)^{-1}$	No motion, hexagon
	No motion
$Q \equiv 2(2R - R_1)$	$T \equiv 8R - R_1$

TABLE 2. (after Segel and Davis). Stable flow for various ranges of the Rayleigh number.

Typical non-zero coefficients are

$$D_1 = 3(480 - m554)/4\alpha^2, \quad D_2 = 7/24\alpha^2,$$
$$D_{110} = 1/4\alpha^5, \quad D_{330} = (227 - 960m)/16\alpha^5.$$

The total solution to second order is then $\mathbf{U}_1 + \mathbf{U}_2$.

It can now be seen that the existence condition for $\mathbf{U}_3$ determines the amplitude equations to third order. Therefore it is only necessary to obtain solutions $\mathbf{U}_n$ through second order. The amplitude equations (16) are given by

$$[\mathbf{U}^*, (\mathcal{N} + [N_{Ma} - N_{Ma}^c]\mathcal{M})(\mathbf{U}_1 + \mathbf{U}_2)] = 0,$$

or, neglecting higher order terms,

$$\iiint_{\text{cell}} T^*(T_{1_t} + N_1 T_1 + N_1 T_2 + N_2 T_1)\, dx\, dy\, dz + (N_{Ma} - N_{Ma}^c)\int_{z=0} w_z^* \nabla_1^2 T_1\, dx\, dy = 0$$
$$N_1 = \mathbf{u}_1 . \nabla, \quad N_2 = \mathbf{u}_2 . \nabla.$$

Amplitude equations

The behaviour of solutions to the amplitude equations can be determined by making a linear stability analysis around each equilibrium solution. These solutions are found by obtaining the roots of equations (16) at steady state. The linear analysis classifies the equilibrium points as either stable nodes, corresponding to stable solutions of the original equations with respect to the interacting disturbances, or unstable nodes or saddle-points which correspond to unstable solutions. Since the equilibrium points are the only points where solution trajectories can cross, knowledge of the behaviour near these points is usually sufficient to determine global behaviour. For a detailed analysis of amplitude equations the reader is referred to Segel & Stuart (1962) and Segel (1965*a, b*). Segel's results for the interaction of N-disturbances of the same overall wave-number are summarized in table 2.

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