Exact sine series solution for oscillatory convection in a binary fluid

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The linear equations governing the onset of convection in a binary fluid subject to the Soret and Dufour effects are solved exactly for rigid boundary conditions by expanding the velocity and temperature as Fourier sine series. The velocity and temperature distributions are found to be those of the free-boundary problem, plus small admixtures of higher harmonics. Exact expressions for the Rayleigh number and oscillation frequency are obtained in terms of infinite series. For the special case of negligible Dufour effect our results agree with exact numerical calculations. Results for the general case are compared with previous approximate methods of solution.

I. INTRODUCTION

Binary fluids, i.e., mixtures of two nonreacting pure components allow observation of a much wider variety of convective phenomena than pure fluids. The most important of these is a transition from equilibrium to oscillatory convection in which the convective motion appears as standing or traveling waves. Convection in binary mixtures is also influenced by the Soret effect, in which a temperature gradient gives rise to a diffusive mass flux, and the converse phenomenon of a concentration gradient producing a heat flux, known as the Dufour effect.¹ Binary fluids are important experimentally because the fluid parameters can be varied by altering the composition of the mixture. Typical mixtures used include alcohol-water and liquid ³He-⁴He.

Although the onset of convection is described by linear equations, it is surprisingly difficult to find exact solutions for realistic rigid boundary conditions. Solutions have recently been obtained by brute force for the special case of zero Dufour coefficient by expanding the vertical velocity as a linear combination of exponentials.²⁻⁴ However, the algebra involved is heavy as it includes calculation of the real and imaginary parts of an 8×8 matrix. In practice this calculation is done numerically. Knobloch and Moore³ report that the matrix involved is ill-conditioned, necessitating use of a double-precision representation. In this paper we present a technique for solving the oscillatory convection equations with rigid boundary conditions exactly, avoiding the numerical problems inherent in the previous approach. An exact treatment of the Dufour effect is presented. In addition, our method shows that the velocity and temperature distributions are those of the free-boundary problem, plus small admixtures of higher harmonics.

The onset of oscillatory convection in a layer of binary fluid confined between two horizontal plates is governed by the equations⁵

$$(D^2-a^2)(D^2-a^2-i\omega)w = Ra^2(1+S)\theta + RSa^2\eta$$
, (1a)

$$(D^2 - a^2 - \sigma i\omega)\theta = -w + AL(D^2 - a^2)\eta , \qquad (1b)$$

$$\left[D^2 - a^2 - \frac{\sigma}{(1+A)L}i\omega\right]\eta = \frac{1}{(1+A)L}(D^2 - a^2)\theta . \quad (1c)$$

The upper and lower boundaries are maintained at temperatures T_0 , $T_0 + \Delta T$ and concentrations $c_0 + \Delta c$, c_0 respectively. Once ΔT has been chosen, Δc is fixed by the requirement that the vertical component of diffusive mass flux vanishes at the boundaries. In Eq. (1) w is the vertical component of velocity and θ and χ represent departures from equilibrium of temperature and concentration respectively, with $\eta = -\chi - \theta$. Physically $\nabla \eta$ is proportional to the diffusive mass flux. The separation ratio Smeasures the relative contribution of concentration and temperature to density variations, σ is the Prandtl number, L the Lewis number, and A the Dufour parameter. D denotes the operator d/dz. The eigenvalues are the thermal Rayleigh number R and the oscillation frequency ω . It is assumed that the layer is of infinite horizontal extent and convection sets in with horizontal wave number a. All quantities are dimensionless. In particular, lengths are scaled with the layer thickness so that the horizontal boundaries are at z=0,1 and frequencies are scaled with v/d^2 , where v is the kinematic viscosity.

Solutions are easily found for free-boundary conditions

$$w = D^2 w = \theta = \eta = 0 \quad \text{at } z = 0, 1 \tag{2}$$

for the eigenfunctions are then proportional to $\sin(n\pi z)$. However, such boundary conditions are artificial as they allow fluid to diffuse through the boundaries. Exact solutions using realistic rigid boundary conditions

$$w = Dw = \theta = D\eta = 0 \quad \text{at } z = 0,1 \tag{3}$$

have proved hard to find. Consequently initial efforts concentrated on finding approximate solutions for R and ω . The earliest approach was the Galerkin technique of Hurle and Jakeman.⁶ The temperature is expanded in terms of a finite number of orthogonal basis functions satisfying the boundary condition. Equations (1) are then solved for w and η in terms of this approximate temperature distribution. Expoliting orthogonality gives expres-

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sions for R and ω . This technique was subsequently rediscovered by other authors^{7,8} and recently extended to treat a rotating binary mixture.⁹ Indeed it is the same method as was originally applied to pure fluids by Chandrasekhar.¹⁰

An alternative Galerkin technique,² expected to give lower accuracy, consists of expanding all three variables as independent series of orthogonal basis functions and applying orthogonality conditions to all three governing equations.

A different approach introduced by Gutkowicz-Krusin, Collins, and Ross⁸ resulted in an expression for Rin terms of integrals over the layer of quantities quadratic in w, θ, η . The approximation $\theta = \sin(\pi z)$ was made for the temperature and w and η were solved for exactly in terms of hyperbolic and trigonometric functions.

We label this expression a pseudovariational principle; it is not a true variational principle if $\omega \neq 0$, but reduces to a variational principle in the $\omega \rightarrow 0$ limit. Although Gutkowicz-Krusin, Collins, and Ross⁸ calculated the eigenfunctions w and η corresponding to higher temperature modes $\theta = \sin(n\pi z)$, n > 1, there is no way they can be used to improve the accuracy of the lowest-order solution, using a series of form $\sum_n a_n \sin(n\pi z)$ —as their integral expression is not a true variational principle one is not at liberty to determine the a_i 's by setting $\partial R / \partial a_i = 0$. Thus the method should give good results for separation ratios near zero but may become less accurate for large negative S.

II. SINE SERIES SOLUTION

Recent experiments on liquid ³He-⁴He mixtures^{11,12} give critical Rayleigh numbers in good agreement with theoretical values obtained from the pseudovariational expression provided S is greater than -1.4. As S is decreased beyond -1.4 theory and experiment diverge rapidly, the theoretical Rayleigh number becoming several times larger than the experimental value. Therefore it is clearly desirable to solve Eqs. (1) with rigid boundary conditions exactly. This system is symmetric about the layer midpoint so solutions will have a definite parity. We will restrict attention to even parity solutions because they give the lowest R value.

The standard Rayleigh-Bénard problem is recovered by neglecting η and setting S=0, A=0, $\omega=0$,

$$(D^2 - a^2)^2 w = Ra^2\theta ,$$

$$(D^2 - a^2)\theta = -w .$$
(4)

Naively, one might expect to get a solution by expanding w, θ as Fourier series

$$w = \sum_{n \text{ odd}} a_n \sin(n \pi z), \quad \theta = \sum_{n \text{ odd}} b_n \sin(n \pi z) , \qquad (5)$$

satisfying the boundary conditions $w = \theta = 0$ at z = 0, 1. However, substituting in (4) gives $a_n = b_n = 0 \forall n$. The error in this approach is that it will turn out that $a_n \sim n^{-3}$ for large *n*. Thus the series for D^2w and higher derivatives diverge.

Jeffreys¹³ got around this difficulty by first eliminating

one of the variables from (4) giving

$$(D^2 - a^2)^3 \theta + Ra^2 \theta = 0 .$$
⁽⁶⁾

He then expanded $D^6\theta$ as a Fourier series

$$\frac{d^6\theta}{dz^6} = \sum_n c_n \sin(n\,\pi z) \,. \tag{7}$$

Integrating gives an expression for θ in the form Fourier series plus polynomial:

$$\theta = B_0 + \frac{B_2}{2!} (z - \frac{1}{2})^2 + \frac{B_4}{4!} (z - \frac{1}{2})^4 - \sum_n \frac{c_n}{(n\pi)^6} \sin(n\pi z) .$$
(8)

(Remember that a solution symmetric about $z = \frac{1}{2}$ will produce the lowest Rayleigh number.) The boundary conditions (3) translated into conditions on θ are

$$\theta = D^2 \theta = (D^3 - a^2 D) \theta = 0 \quad \text{at } z = 0, 1 . \tag{9}$$

The first two conditions fix the relative sizes of the constants B_0, B_2, B_4 ; (6) then determines the coefficients c_n and the third boundary condition gives an expression for R

$$\sum_{\substack{n=1\\n \text{ odd}}}^{\infty} \frac{n^2 \pi^2 (n^2 \pi^2 + a^2)}{(n^2 \pi^2 + a^2)^3 - Ra^2} = 0 .$$
 (10)

Allowed R values can be found from (10) by iteration.

Extension of Jeffreys's method to more complicated problems¹⁴ is straightforward provided all the boundary conditions except one involve only even derivatives. The reason is that even derivatives of the Fourier series in (8) vanish at the boundaries, allowing the B_i 's to be determined independently of the c_n 's.

However, in a binary fluid two boundary conditions involve odd derivatives. Nield¹⁵ was successful in applying Jeffreys's technique to stationary convection in a doublediffusive system but the algebra is very complex, the result being expressed in the form of a determinant. It is questionable whether this technique would work in the presence of the cross coupling induced by the Soret and Dufour effects; certainly the amount of algebra involved would be prohibitive. Instead we return to the original expansion (5) and avoid problems due to higher derivatives diverging by integrating over the layer. Two undetermined constants appear; one is removed by an appeal to homogeneity while the other is determined from the exact solution of (1c).

It will prove convenient to recast Eqs. (1) in the form

$$(D^2 - a^2)(D^2 - a^2 - i\omega)w = R_1\theta + R_2\eta$$
, (11a)

$$[(1-\epsilon)(D^2-a^2)-\sigma i\omega]\theta = -w + \gamma \eta , \qquad (11b)$$

$$(D^2 - a^2 - \sigma i \omega / \lambda) \eta = (D^2 - a^2) \theta / \lambda$$
, (11c)

where $R_1 = Ra^2(1+S)$, $R_2 = RSa^2$, $\lambda = (1+A)L$, $\epsilon = A/(1+A)$, and $\gamma = \epsilon \sigma i \omega$. In what follows it is understood that all sums are over odd *n*. Equations (11) are multiplied by $2 \sin(n\pi z)$ and integrated from 0 to 1. Integration by parts if used repeatedly until all the differential operators operate on $\sin(n\pi z)$ rather than w, θ , and η . Only then are the series expansions (5) inserted, to give

$$N^{2}F_{1}a_{n} + 4n\pi w_{0}^{\prime\prime} = R_{1}b_{n} + R_{2}I,$$

$$F_{2}b_{n} = a_{n} - \gamma I,$$

$$F_{3}I - 4n\pi\eta_{0} = N^{2}b_{n}/\lambda,$$
(12)

where we have introduced the notation

$$F_{1} = N^{2} + i\omega,$$

$$F_{2} = (1 - \epsilon)N^{2} + \sigma i\omega,$$

$$F_{3} = N^{2} + \sigma i\omega/\lambda,$$
(13)

and

$$I = 2 \int_{0}^{1} \sin(n\pi z) \eta \, dz ,$$

$$N^{2} = n^{2} \pi^{2} + a^{2} ,$$

$$w_{0}^{"} = D^{2} w|_{z=0} \quad (=D^{2} w|_{z=1} \text{ by symmetry}) ,$$

$$\eta_{0} = \eta(0) \quad [=\eta(1) \text{ by symmetry}] .$$
(14)

Elimination of I results in

$$a_{n} = \frac{4n\pi}{\Delta} \left[\eta_{0}(R_{2}F_{2} - \gamma R_{1}) - w_{0}^{"} \left[F_{2}F_{3} + \frac{\gamma N^{2}}{\lambda} \right] \right],$$

$$b_{n} = \frac{4n\pi}{\Delta} \left[\eta_{0}(R_{2} - \gamma N^{2}F_{1}) - F_{3}w_{0}^{"} \right],$$
 (15)

with

$$\Delta = N^2 F_1 F_2 F_3 - R F_4 + \gamma N^4 F_1 / \lambda ,$$

$$F_4 = a^2 (1+S) F_3 + N^2 S a^2 / \lambda .$$
(16)

The problem is homogeneous, so we fix a scale by choosing $w_0'' = -R_2$. It only remains to calculate η_0 . Introducing $c^2 = a^2 + \sigma i \omega / \lambda$, the even solution of (11c) is

$$\eta = \sum_{n=1}^{\infty} \frac{N^2}{\lambda F_3} b_n \sin(n \pi z) + B \cosh[c(z - \frac{1}{2})] .$$
 (17)

The constant B is found from the boundary condition $D\eta=0$ at z=0,1

$$B = \frac{1}{\lambda c \sinh(\frac{1}{2}c)} \sum_{n=1}^{\infty} \frac{n \pi N^2}{F_3} b_n$$
(18)

using (11) and (12). Hence

$$\eta_0 = B \cosh(\frac{1}{2}c) = \frac{1}{\lambda c \tanh(\frac{1}{2}c)} \sum_{n=1}^{\infty} \frac{n \pi N^2}{F_3} b_n .$$
 (19)

Substituting (15) in (19) yields an equation linear in η_0 , which when solved gives

$$\eta_0 = \frac{\Sigma_1}{L' - \Sigma_2} , \qquad (20)$$

where

$$\Sigma_{1} = \sum_{n=1}^{\infty} \frac{n^{2} \pi^{2}}{\Delta} N^{2} ,$$

$$\Sigma_{2} = \sum_{n=1}^{\infty} \frac{n^{2} \pi^{2} N^{2}}{\Delta F_{3}} \left[1 - \frac{\gamma N^{2} F_{1}}{R_{2}} \right] ,$$
(21)

and

$$L' = \frac{\lambda c \tanh(\frac{1}{2}c)}{4RSa^2} .$$
 (22)

The boundary condition Dw = 0 at z = 0, 1 applied to (11) with a_n given by (15) gives the required relation for R and ω :

$$\sum_{n \text{ odd}} \frac{n^2 \pi^2}{\Delta} \left[\left[F_2 F_3 + \frac{\gamma N^2}{\lambda} \right] + \eta_0 \left[F_2 - \gamma \frac{R_1}{R_2} \right] \right] = 0.$$
(23)

Writing Σ_3 for the left-hand side of (23) one can quickly solve for R and ω using two-dimensional Newton-Raphson iteration. With initial guesses R_0, ω_0 for R, ω : to first order

$$\Sigma_{3}(R_{0} + \Delta R, \omega_{0} + \Delta \omega) = \Sigma_{3}(R_{0}, \omega_{0}) + \Delta R \frac{\partial \Sigma_{3}(R_{0}, \omega_{0})}{\partial R} + \Delta \omega \frac{\partial \Sigma_{3}(R_{0}, \omega_{0})}{\partial \omega} .$$
(24)

The partial derivatives can easily be calculated from (23). Equating real and imaginary parts of (24) to zero fixes ΔR and $\Delta \omega$; now $R_0 + \Delta R$ and $\omega_0 + \Delta \omega$ are better approximations to R and ω . Iterating gives second-order convergence to the solution of (23). Thus we have obtained an exact solution for the Rayleigh number at the price of evaluating three infinite series. The slowest of these to converge is the first term in (23) which converges like n^{-2} . [Remember that $N^2 = n^2 \pi^2 + a^2$. Note also that (15) verifies the claim made earlier that $a_n \sim n^{-3}$ for large n.] The convergence can be radically improved, by using the standard technique of adding and subtracting a series whose sum is known.¹³ Using the identity

$$\sum_{n \text{ odd}} \frac{n^2 \pi^2}{(n^2 \pi^2 + a^2)(n^2 \pi^2 + b^2)} = \frac{1}{4(a^2 - b^2)} [a \tanh(\frac{1}{2}a) - b \tanh(\frac{1}{2}b)] \quad (25)$$

and selecting $b^2 = a^2 + i\omega$ we have

$$\sum_{n \text{ odd}} \frac{n^2 \pi^2}{N^2 F_1} = \frac{1}{4i\omega} \left[b \tanh(\frac{1}{2}b) - a \tanh(\frac{1}{2}a) \right]$$
$$\equiv K_1 \text{ say }. \tag{26}$$

Hence



and the convergence is now like n^{-8} rather than n^{-2} .



FIG. 1. (a) Critical Rayleigh number and (b) oscillation frequency as a function of separation ratio S for $\sigma = 0.75$, L = 0.04, A = 0. Solid lines denote the exact solution; dashed lines, the pseudovariational approximation.

TABLE I. Critical Rayleigh number as a function of A and S for $\sigma = 0.75$, L = 0.04.

S	A = 0	A = 0.005	A = 0.01	A = 0.015	
-0.25	2 162.6	2 163.2	2 163.7	2 164.1	
-0.5	2 629.6	2 630.2	2 630.9	2 631.5	
-1.0	4 472.6	4 473.8	4 475.0	4 476.3	
-1.5	10 900.6	10 903.8	10907.1	10910.3	

Similarly, the second term in Σ_2 can be made to converge like n^{-4} by adding and subtracting

$$K_{2} \equiv \frac{1}{1-\epsilon} \sum \frac{n^{2} \pi^{2}}{F_{3}^{2}}$$

= $\frac{1}{8c(1-\epsilon)} [\tanh(\frac{1}{2}c) + \frac{1}{2}c \operatorname{sech}^{2}(\frac{1}{2}c)].$ (28)

Further improvements in convergence stem from applying the formula

$$\sum_{n \text{ odd}} \frac{n^2 \pi^2}{(n^2 \pi^2 + e^2)(n^2 \pi^2 + b^2)(n^2 \pi^2 + c^2)} = -\frac{1}{4} \left[\frac{e \tanh(\frac{1}{2}e)}{(b^2 - e^2)(c^2 - e^2)} + \frac{b \tanh(\frac{1}{2}b)}{(e^2 - b^2)(c^2 - b^2)} + \frac{c \tanh(\frac{1}{2}c)}{(e^2 - c^2)(b^2 - c^2)} \right]$$
(29)

and an associated formula which comes from differentiating (29) with respect to e.

III. RESULTS

The critical Rayleigh number is found by minimizing R as a function of wave number a. We first consider the case where the Dufour effect is neglected. This special case was solved previously in Refs. 2-4 using numerical (although still exact) techniques. Results of the sine series calculation are plotted in Fig. 1 for negative values of S. The parameter values selected are typical of liquid ³He-⁴He mixtures. Also shown in the figure are the results of the pseudovariational method. These are in good agreement with the exact calculation, differing by about 10% at S = -1.6. Since the Dufour effect is small in normal liquid helium mixtures ($A \leq 0.01$) its inclusion has little effect on calculated values for R and ω as can be seen from the data presented in Table I.

Thus discrepancies observed between theory and experiment^{11,12} are probably due to differences between the experimental system and the theoretical model. For example, if the bifurcation to convection were backwards, noise of finite amplitude could trigger a transition to convection at a Rayleigh number smaller than that predicted by linear theory.

We have also performed a sine series calculation for the onset of stationary convection. For S values just below zero, $R_{\text{stationary}} < R_{\text{oscillatory}}$ and stationary convection should be observed. The converse applies at large negative S. For $\sigma = 0.75$, L = 0.04 the crossover from stationary to oscillatory convection occurs at $S = -0.000\,87$ with a jump in wave number from a = 3.1491 (stationary) to a = 3.0686 (oscillatory). The oscillatory branch continues to exist until $S = -0.000\,84$, at which point the frequency vanishes. These results are consistent with Knobloch and Moore³ and the revised calculations of Zielinska and Brand.²

The eigenfunctions can be constructed from (5). Reinstating the dependence on horizontal coordinate x and time t gives for a standing wave solution

$$w = \operatorname{Re}\left[\sum_{n} a_{n} \sin(n \pi z) \exp(i \omega t)\right] \cos(ax) .$$
 (30)

This can be rewritten

$$w = [|a_1|\cos(\omega t + \phi_1)\sin(\pi z) + |a_3|\cos(\omega t + \phi_3)\sin(3\pi z) + \cdots]\cos(ax) .$$
(31)

The phase angles ϕ_i are determined by $\tan \phi_i = \operatorname{Im} a_i /$



Re a_i . In general $|a_1| > |a_3| > |a_5| \cdots$. Therefore it is clear that when $\omega t + \phi_1$ is close to a multiple of 2π , the velocity will appear to be proportional to $\sin(3\pi z)$. This explains the three cell structure found in the calculations of Ref. 3.

Inclusion of the Dufour effect is illustrated in Fig. 2 using the parameter values $\sigma = \frac{2}{3}$, L = 0.64, A = 0.25 considered appropriate to binary gas mixtures in Ref. 8. The jump in wave number at the crossover from oscillatory (a=2.49) to stationary convection (a=3.50) is now much greater. Again, the pseudovariational approximation is in relatively good agreement with the exact results. Figure 3 demonstrates the effect of varying the Dufour parameter A, with the other parameters held fixed. For small A, the critical Rayleigh number increases approximately linearly with A. However at large A, the Rayleigh number can actually decrease as A increases.

IV. CONVECTION DRIVEN BY THE DUFOUR EFFECT

Linz¹⁶ considered a rather different system in which there is still a concentration difference Δc between top and bottom boundaries but the temperature difference ΔT is set to zero, in an attempt to enhance concentration effects relative to thermal effects. The disadvantage of this setup is that the condition of zero mass flux through the boundaries can no longer be satisfied, except for the trivial case $\Delta c = 0$. Instead the permeable boundary condition $\eta = 0$ at z = 0, 1 was imposed. In our notation, the governing equations for this system are

$$(D^{2}-a^{2})(D^{2}-a^{2}-i\omega)w = R_{s}a^{2}(1+1/S)\theta + R_{s}a^{2}\eta ,$$

$$(D^{2}-a^{2}-\sigma i\omega)\theta = \epsilon\lambda(D^{2}-a^{2})\eta , \qquad (32)$$

$$(D^{2}-a^{2}-\sigma i\omega/\lambda)\eta = -w/\lambda + (D^{2}-a^{2})\theta/\lambda ,$$



FIG. 2. (a) Critical Rayleigh number and (b) oscillation frequency as a function of separation ratio S for $\sigma = \frac{2}{3}$, L = 0.64, A = 0.25. Solid lines denote the exact solution; dashed lines, the pseudovariational approximation. The dotted line denotes the threshold for stationary convection.

FIG. 3. The change in critical Rayleigh number relative to its value at A=0. The curves are labeled with the value of S. The other parameters were held constant at $\sigma = \frac{2}{3}$, L=0.64. The two lowest curves terminate when $\omega=0$.



FIG. 4. (a) Reduced critical Rayleigh number \hat{R} and (b) oscillation frequency for Dufour driven convection with $\sigma = 1$, L = 0.5, and Q = 10. \hat{R} is obtained by dividing R by its value at S = 0: $\hat{R} = R/1708$ for the exact solution (labeled E); $\hat{R} = R/657.5$ for the approximate solution (labeled A).

where R_s is the solutal Rayleigh number, together with boundary conditions

$$w = Dw = \theta = \eta = 0$$
 at $z = 0, 1$. (33)

Using the techniques of Sec. II, it is straightforward to

derive the following expression determining R_s and ω :

$$\sum_{n \text{ odd}} \frac{n^2 \pi^2 (F'_2 F_3 - \epsilon N^4)}{N^2 F_1 (F'_2 F_3 - \epsilon N^4) - R_s a^2 [(1 + 1/S) \epsilon N^2 + F'_2 / \lambda]} = 0, \quad (34)$$

where $F'_2 = N^2 + \sigma i \omega$ and the other symbols are as defined previously. Linz made the further approximation of replacing the boundary condition Dw = 0 by the freeboundary condition $D^2w = 0$. Results from his expressions are compared with the exact solution (34) in Fig. 4. Good agreement for the Rayleigh number is achieved only if it is scaled by its value at S = 0, i.e., 657.5L for the approximate solution and 1708L for the exact solution. Note that following Ref. 16 we work at fixed $Q \equiv A/S^2$ rather than fixed A.

V. CONCLUSION

The sine series expansion provides a simple means of obtaining an exact solution for the onset of convection in a binary fluid subject to the Soret and Dufour effects. The form of the velocity and temperature eigenfunctions can easily be visualized. In liquids the Dufour effect is small and causes only a small increase in the critical Rayleigh number. The size of this increase is approximately proportional to the Dufour coefficient A. In gases, the Dufour effect again increases the critical Rayleigh number, but the magnitude of the increase is no longer a monotonic function of A, if A is sufficiently large. The extension of the sine series expansion to treat other systems, e.g., rotating binary mixture should be straightforward.

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