# The stability of uniformly accelerated flows with application to convection driven by surface tension

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The method of energy is used to study the stability of uniformly accelerated flows, i.e. those flows characterized by an impulsive change in boundary temperature or velocity. Two stability criteria are considered: strong stability, in which there is exponential decay of the disturbance energy, and marginal stability, in which the disturbance energy is less than or equal to its initial value. For the important case in which the critical stability parameter (measured by the Marangoni, Rayleigh or Reynolds number) decreases with time, it is proved that an onset time exists. Furthermore, it is shown that the experimental onset time is bounded below by the marginal stability limit, which in turn is bounded below by the strong stability limit.

The method is then applied to the problem of an impulsively cooled liquid layer susceptible to instabilities driven by interfacial-tension gradients. The strong stability and marginal stability boundaries are calculated and bounds on the onset time are given. These results represent the first rigorous bounds for convective instability problems of this class. Comparison with the limited available experimental data shows the calculated results to be lower bounds on the experimental onset times, and hence the theory is in agreement with available experimental results.

#### 1. Introduction

The energy stability method has been recently applied to a number of timedependent flows, notably by von Kerczek & Davis (1972), Davis & von Kerczek (1973) and Homsy (1973, 1974). In the case of modulated base states, the important reformulation of the energy method by Davis & von Kerczek (1973) allowed the definition of two criteria for 'stability'. In Homsy (1974, hereafter referred to as II), these were referred to as *strong global stability* and *asymptotic stability*. The former definition is due to Joseph (1971) and implies an exponential decrease of a generalized 'energy' functional with time for  $t \in [0, \infty)$ . The latter is a weaker concept of stability and simply implies a net decrease of the energy functional over one cycle of modulation. This criterion implies asymptotic stability in the mean, but does not exclude a large disturbance which may grow

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(in a suitable norm) over a portion of a cycle and indeed, which may be extraordinarily persistent in time. Detailed calculations are available for the oscillatory Stokes layer, in Davis & von Kerczek (1973), and fluid layers subject to gravity or surface-temperature modulations, in II.

In Homsy (1973, hereafter referred to as I), the method was applied to impulsively heated fluid layers. Below we shall refer to this class of problems as uniformly accelerated flows', although we include impulsive heating and cooling and flows experiencing a sudden change in boundary velocity. The discussion in I dealt with the possible shapes of the stability boundaries in the Rayleighnumber/time plane, which result in either lower bounds on the 'onset time', defined in detail below, or a lowering of the global stability limit below its value for steady flows. Also in I, a full discussion was given of the inadequacies the results of both the quasi-static and the amplification linear theory for accelerating flows. Indeed, the results of Rosenblat & Herbert (1970) and those in II demonstrate that the quasi-static approximation is valid only when the base state and disturbances have widely different time scales, i.e. a disturbance Strouhal number is small. There is no equivalent separation of scales in the case of uniformly accelerated flows in which both the base state and disturbances evolve on a diffusive time scale. It is thus apparent that no linear instability theory exists for accelerated flows of this class, and it is in this context that the method of energy finds great utility.

In this paper we return to a study of this class of flows. We show in §2 that, in addition to the discussion in I, the reformulation of the energy method allows the introduction of a weaker definition of 'stability' which requires only that the disturbance energy be less than its initial value. This definition was motivated, as was the introduction of a similar weak concept of stability in II, by experimental evidence which supports its applicability in certain circumstances.

This work was motivated by the applicability of the method of energy to instabilities driven by surface tension; we treat this problem in  $\S3$  in some detail following the general discussion. The relevant studies dealing with surfacedriven instabilities of time-varying base states are Blair & Quinn (1969), Vidal (1967) and Vidal & Acrivos (1968). In addition to their careful experimental work on buoyancy-driven instabilities, Blair & Quinn (1969) briefly considered horizontal liquid layers subject to flows driven by surface tension. A 10% solution of ethyl ether in monochlorobenzene, initially in equilibrium with its atmosphere, was exposed to a step change in pressure. This resulted in the desorption of the ethyl ether. Both schlieren photography and quantitative measurements of mass transport rates were used to determine the onset time. We define this quantity as the time between the impulsive change and the manifestation of a deviation from the stagnant, penetrative concentration (temperature) field. The concept of an onset time is thus intimately linked to the observed experimental fact that flows accelerated from a quiescent state require a finite time before disturbances, assumed initially small, grow to an amplitude sufficient to become detectable. It will emerge below, however, that in certain cases disturbances of any initial norm decay exponentially in time with decay rates which are quantitatively predictable. Blair & Quinn have dramatically shown that the

onset time is unambiguously defined, as they found good agreement between onset times measured from deviations in transport rates and those determined using schlieren visualization of the instability. Unfortunately, owing to the unavailability of physical-property data for this system, we can make no direct comparison with the predictions developed below.

Vidal & Acrivos (1968) have attempted a linear stability analysis of a timedeveloping penetrative temperature profile. In their model, at zero time thermal energy is withdrawn at a constant rate from the free surface of an initially stagnant and isothermal fluid layer. The stability of the resulting time-varying base state with respect to instabilities driven by surface tension was investigated using the quasi-static approximation, which as we have noted above, is inappropriate for problems of this class. The resulting predictions of critical Marangoni numbers were compared with experimental data on evaporating pools of acetone and methyl alcohol (Vidal 1967) and propyl alcohol (Vidal & Acrivos 1968). In the experimental work, the temperature profile at onset (onset being determined by schlieren photography) was used to calculate a critical Marangoni number. While the agreement between experiment and the quasistatic results is fair, it may be due to judicious reading of the experimental temperature profile, since the onset times themselves are in wide disagreement.

The present work follows naturally from Davis & von Kerzcek and I. We seek to determine regions of stability in the Marangoni-number/time plane using the method of energy. The importance of the work is obvious owing to the absence of previous theoretical results on flow driven by surface tension. We note, too, that the model of a penetrative temperature profile considered here is often more representative of the true state of affairs than is the linear temperature profile treated by Pearson (1958) and refined by many others.

The main result of this paper is that, for the first time, the energy method provides lower bounds on the onset time. We note that the results given for impulsive Couette flow by Conrad & Criminale (1965) are not true bounds since they were computed for a restricted class of disturbances. They do, however, suggest that the results developed in §2 below might be applicable to a wider class of accelerating flows.

# 2. Formulation

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# The energy identities

Let  $\mathbf{\tilde{u}}(x, y, z, t)$ ,  $\overline{T}(x, y, z, t)$  be a dimensionless solution to the Boussinesq equations appropriate to an impulsive change in boundary velocity, temperature and/ or heat flux. We call this the base state. Following Joseph (1966), it can be shown that all dimensionless disturbances  $(\mathbf{u}, \Theta)$  satisfy the energy equality (with  $\phi = \Theta(\lambda R)^{\frac{1}{2}}$ )  $dF/dt = PL(t) = \emptyset$  (2.1)

$$dE/dt = RI_{\lambda}(t) - \mathscr{D}. \tag{2.1}$$

Here  $E = \frac{1}{2} \langle |\mathbf{u}|^2 / \sigma + \phi^2 \rangle$  is a positive-definite 'energy' functional and the brackets refer to integration over a domain which is (i) finite in at least one dimension, (ii) finite in one dimension with periodicity in the remaining dimensions or (iii) finite in all dimensions. The remaining quantities in (2.1) are  $I_{\lambda}(t)$ ,

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an 'energy production' integral,  $\mathcal{D}$ , a positive-definite generalized dissipation, and R, a physical parameter of the flow. The following three cases are among the most common.

(1) Impulsive flow. R is the Reynolds number based on the maximum velocity and maximum finite dimension of the domain.  $\mathscr{D}$  is the dissipation function  $\langle \nabla \mathbf{u} : \nabla \mathbf{u} \rangle$  and  $I_{\lambda}$  is the symmetric 'Reynolds stress'

$$I(t) = \langle \mathbf{u} . \mathbf{D} . \mathbf{u} \rangle,$$

where **D** is the rate-of-strain tensor of the base flow. In cases when the disturbances correspond to longitudinal rolls, specialized definitions of E and  $I_{\lambda}(t)$  are possible (Joseph & Hung 1971).

(2) Impulsive heating. R is the square root of the Rayleigh number and

$$\begin{split} I_{\lambda}(t) &= \lambda^{-\frac{1}{2}} \langle w\phi \rangle - \lambda^{\frac{1}{2}} \langle w\phi \,\partial \overline{T} / \partial z \rangle, \\ \mathscr{D} &= \langle \nabla \mathbf{u} \colon \nabla \mathbf{u} + |\nabla \phi|^2 \rangle + \oint L \phi^2. \end{split}$$

with  $\mathscr{D} = \langle \nabla \mathbf{u} : \nabla \mathbf{u} + | \nabla \phi$ 

Here  $w = \mathbf{k} \cdot \mathbf{u}$  is the vertical component of velocity.

(3) Impulsive evaporation with variable surface tension. This case is developed in §3 below. R is the square root of the Marangoni number, and we have

$$I_{\lambda}(t) = -\left(\lambda^{\frac{1}{2}} \left\langle w\phi \frac{\partial \overline{T}}{\partial z} \right\rangle + \lambda^{-\frac{1}{2}} \int_{z=1} \phi \frac{\partial w}{\partial z}\right),$$

with  $\mathscr{D}$  as in case 2.

We then have

We emphasize that (2.1) is identically satisfied for all solutions to the dynamic equations, regardless of their amplitude. We now develop our stability criteria.

#### Strong stability

We briefly recapitulate and strengthen the results in I. Consider the maximum problem  $e^{-1}(t) = \max(L/Q)$ (2.2)

$$\rho_{\lambda}^{-1}(t) = \max_{h} \left( I_{\lambda} / \mathcal{D} \right). \tag{2.2}$$

(For a discussion of the space h, see I, Davis (1969), and §4.)

$$dE/dt \leq \mathscr{D}\{-1 + R/\rho_{\lambda}(t)\}.$$
(2.3)

We shall be primarily concerned in this paper with cases for which  $\rho_{\lambda}(t)$  is a monotone decreasing function of time. We state the main result as the following theorem.

THEOREM 1. Let  $\rho_{\lambda}(t)$  be monotone decreasing with at most an integrable singularity at t = 0. Then for a given  $R(>\rho_{\lambda}(\infty))$  the flow is strongly stable for all times  $0 \le t \le t^*$ , where  $t^*$  satisfies

$$\rho_{\lambda}(t^*) = R. \tag{2.4}$$

Equation (2.4) is an implicit relation between the time interval  $(0, t)^*$  during which the energy decreases and the physical flow parameter R. The proof of the theorem follows from standard manipulations. For all times such that  $R < \rho_{\lambda}(t)$ , (2.3) may be bounded by  $dE/dt < \xi^2 E (-1 + B/c_{\lambda}(t))$  (2.5)

$$dE/dt \leq \xi^2 E\{-1 + R/\rho_\lambda(t)\},\tag{2.5}$$

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whence

$$E(t) \leqslant E(0) \exp\left\{-\xi^2\left(t-R\int_0^t \frac{dt}{
ho_\lambda}
ight)
ight\},$$

for any time  $t \leq t^*$ . Now since  $t \leq t^*$  and  $\rho_{\lambda}$  is monotone decreasing by hypothesis, we have

$$E(t) \leq E(0) \exp\{-\xi^2 t (1 - R/\rho_\lambda(t^*))\},$$
(2.6)

which proves the theorem. An important interpretation of theorem 1 is the following corollary.

COROLLARY 1. For a given flow parameter R, the onset time  $t_{on}$  is bounded from below, viz.

$$t_{\rm on} > t^*.$$
 (2.7)

*Proof.* By the mean-value theorem,

$$E(t^*) \leqslant E(0) \exp\{-\xi^2 t^* (1 - R/\rho_\lambda(\bar{t}))\},\tag{2.8}$$

where  $0 < \bar{t} < t^*$ . Thus any disturbance at  $t^*$  has suffered an attenuation which is bounded from above by  $\exp(-\hat{\xi}^2 t^*)$ , where  $\hat{\xi}^2 (= \xi^2(1 - R/\rho_\lambda(\bar{t})))$  is bounded away from zero. Thus the onset time (in the sense described in §1) must clearly be greater than  $t^*$ .

In I, it was shown that the 'optimal' stability boundary is given by the solution to the maximum problem

$$\tilde{\rho}(t) = \max_{\lambda > 0} \rho_{\lambda}(t) \tag{2.9}$$

and that this additional parametric time dependence of the optimal coupling constant does not invalidate the results. Thus the bound on the onset time is given as the solution of  $\tilde{\rho}(t^*) = R$ .

#### Marginal stability

In this subsection we propose a new criterion for 'stability' of accelerated flows which is weaker than that developed above and which may have practical utility in a restricted sense. A number of carefully executed experiments have shown that, following an impulsive change in boundary conditions, an *initially* quiescent and isothermal fluid layer remains so for times approaching the onset time. Thus omnipresent small disturbances due to thermal and mechanical fluctuations are damped for a finite time interval, after which they presumably grow exponentially until becoming manifest. Thus, a suitable criterion might be expressed as

$$E(t) \leqslant E(0) \tag{2.10}$$

if one has some a priori knowledge that E(0) is small. We refer to the criterion (2.10) as marginal stability. Note that this criterion differs from marginal conditions in linear stability theory, the latter being more appropriately called marginal instability. The development of the marginal stability criterion follows from the energy identity (2.1) and the work of Davis & von Kerczek.

Consider the following maximum problem:

$$\nu_{\lambda}(t) = \max_{h} \{ (RI_{\lambda}(t) - \mathcal{D})/E \}.$$
(2.11)



FIGURE 1. The function  $\nu_{\lambda}(t)$ . The lower curve is for the case  $R < \tilde{\rho}(\infty)$ . The upper curve gives the probable behaviour for  $R > \tilde{\rho}(\infty)$ .

The numbers  $\nu_{\lambda}$  are parametrically functions of time. Coupling (2.11) with (2.1) we arrive at the energy inequality

$$dE/dt \leqslant \nu_{\lambda}(t) E. \tag{2.12}$$

As was pointed out in II, (2.12) serves as a bound for energy *growth* as well as decay. For periodic base states,  $\nu_{\lambda}(t)$  is also periodic and it is possible to integrate (2.12) over one cycle to arrive at an alternative definition of stability, namely, asymptotic stability. The same ideas apply to the present class of problems.

Before developing the marginal stability criterion, we must establish a few preliminary results. The first of these, due to Davis & von Kerczek, is the fact that the base state is strongly globally stable for all values of the flow parameter R for which  $\nu_{\lambda}(t) < 0$ . This follows directly from (2.12). The main emphasis in this paper, however, is on flow parameters R and base states for which  $\nu_{\lambda}$  cannot be shown to be negative. Indeed, from the definition (2.11),  $\nu_{\lambda}(t)$  will be negative only for sufficiently small values of R, which yields the global stability limit for the flow. Conversely, it is clear that, for sufficiently large R,  $\nu_{\lambda}(t)$  can be made to become positive. We now address ourselves to a discussion of the behaviour of the function  $\nu_{\lambda}(t)$ . In the following, we assume that the flow may become unstable, i.e.  $R > \tilde{\rho}(\infty)$ .

THEOREM 2. Let the hypothesis of theorem 1 hold, and let  $t^0$  be a zero of  $\nu_{\lambda}(t)$ . Then (for any fixed  $\lambda$ ),  $t^0 \ge t^*$ .

THEOREM 3. Let the hypothesis of theorem 1 hold; then for any fixed  $\lambda$ ,  $\nu_{\lambda}(t) < 0$  for all  $0 < t < t^{0}$ .

These theorems are particularly useful in that they serve to establish that  $\nu_{\lambda}$  is initially negative and that, if it becomes positive (i.e. the flow is not globally stable), its first zero is bounded from below by the quantity  $t^*$ .

These results are depicted in figure 1, where we show the probable behaviour of  $\nu_{\lambda}(t)$ . The location of the curves depends upon the value of the flow parameter

*R*. For *R* below  $\tilde{\rho}(\infty)$ , the flow is strongly globally stable, i.e.  $\nu_{\lambda}(t)$  is always negative. For *R* sufficiently large,  $\nu_{\lambda}(t)$  will become positive for  $t \ge t^0 \ge t^*$ . We have not been able to prove that  $\nu_{\lambda}$  is monotonic as depicted, but it will be shown to be computationally true for the problem driven by surface tension treated in §3.

The proofs of theorems 2 and 3 rely on use of the definition of  $t^*$ , and its relation to the flow parameter R. Recall that  $t^*$  is defined implicitly through the relation (2.4), which we write as

$$\frac{1}{R} = \max_{h} \left\{ \frac{I_{\lambda}(t^*)}{\mathscr{D}} \right\} = \frac{1}{\rho_{\lambda}(t^*)}$$
(2.13)

for any given flow parameter R. Now any zero of  $\nu_{\lambda}(t)$  must evidently satisfy

$$0 = \max_{h} \left\{ \frac{RI_{\lambda}(t^{0}) - \mathcal{D}}{E} \right\} = \max_{h} \left\{ \frac{\mathcal{D}}{E} \left( \frac{RI_{\lambda}(t^{0})}{\mathcal{D}} - 1 \right) \right\}.$$
 (2.14)

But from (2.13) we have

$$\nu_{\lambda}(t) = \max\left\{\frac{\mathscr{D}}{E}\left(\frac{I_{\lambda}(t)}{\mathscr{D}}\rho_{\lambda}(t^{*}) - 1\right)\right\}$$
(2.15*a*)

and in particular,

$$0 = \max\left\{\frac{\mathscr{D}}{E}\left(\left[\frac{I_{\lambda}(t^{0})}{\mathscr{D}}\rho_{\lambda}(t^{*})\right] - 1\right)\right\}.$$
(2.15b)

The proof of theorem 2 follows from (2.15b). Recall that by hypothesis  $I_{\lambda}/\mathscr{D}$  is bounded from above by a quantity  $(\rho_{\lambda}^{-1})$  which increases monotonically with time. Thus any zero of  $\nu_{\lambda}$  must occur for  $t^{0} \ge t^{*}$ , since  $t^{*}$  is the earliest time for which (2.15b) can hold.

The proof of theorem 3 follows directly from (2.15a). Since  $\mathscr{D}/E$  is positive definite, the sign of  $\nu_{\lambda}$  is determined by the sign of the term square brackets in (2.15a). Now if  $t^0$  is the first zero of  $\nu_{\lambda}$ , then  $I_{\lambda}/\mathscr{D}$  is bounded above by its value at  $t^0$ , and hence  $\nu_{\lambda} < 0$  for all  $t < t^0$ .

We now state the main result of this section.

THEOREM 4. Let  $\nu_{\lambda}(t)$  be a monotone increasing function of t with  $\nu_{\lambda}(t)$  initially negative with at most an integrable singularity at t = 0. Then there exists a unique time  $t = t_{\lambda}^{\dagger} > 0$  such that  $E(t) \leq E(0)$  for  $0 < t < t_{\lambda}^{\dagger}$ .

Integration of (2.12) yields

$$E(t) \leq E(0) \exp\left\{\int_0^t \nu_{\lambda}(t) \, dt\right\}$$
(2.16)

and the proof follows directly. The quantity  $t^{\dagger}$  is evidently determined implicitly from the condition

$$\int_{0}^{t^{\dagger}} \nu_{\lambda}(t) \, dt = 0. \tag{2.17}$$

For any given R, we denote the solution to this problem as  $t^{\dagger}(\lambda, R)$ . The determination of that (constant) value of  $\lambda$  which maximizes  $t^{\dagger}(\lambda; R)$  yields the optimal bound on the onset time, viz.

$$t_{\text{on}} \ge \tilde{t}^{\dagger}(R) = \max_{\lambda > 0} t^{\dagger}(\lambda, R).$$



FIGURE 2. The stability plot in the R, t plane. ---, locus of the function  $\tilde{\rho}(t)$ ; ----,  $\tilde{t}^{\dagger}(R)$ .

The interpretation of theorems 1 and 4 is best displayed in the R, t plane; see figure 2. The dashed curve denotes the locus of the solutions to (2.9). By theorem 1, we have strong stability everywhere to the left of the curve. The intersection of the curve and the horizontal line  $R = \text{constant gives the lower bound } t^*$  for the onset time. Also shown is the locus of points  $\tilde{t}^{\dagger}(R)$ ; in the region between the curves we have  $E(t) \leq E(0)$ . Note that theorem 2, together with the hypothesis of theorem 4, ensures that the curves have this relation to one another.

To the right of the upper curve, one can bound the energy by integrating (2.13) beyond  $t^{\dagger}$ . Such results would be of dubious value, however, since the amplification factor corresponding to the manifestation of the instability is unknown. In closing we note that the hypothesis of (at most) integrable singularities in  $\rho_{\lambda}(t)$  and  $\nu_{\lambda}(t)$  may be removed by integration of the respective inequalities over a time interval bounded away from zero (Davis 1972).

# 3. Flows driven by surface tension

#### Formulation

We propose modelling evaporating liquids subject to instabilities driven by surface tension. If surface effects are to dominate buoyancy, we are restricted to thin liquid layers, which in most cases implies depths less than about 3 mm. A condition of constant heat flux from the upper surface is used to model the evaporation. Vidal (1967) and Vidal & Acrivos (1968) have shown this to be a realistic picture by demonstrating the agreement between the measured surface temperature and the diffusive solution to the appropriate conduction problem. For the small observed changes in surface temperature (less than 1 °C) constant heat flux is equivalent to constant mass flux. Our lower surface condition is that of an isothermal (constant concentration) plate. This condition allows comparison of strong stability results at large times with those of Davis (1969), who solved the corresponding steady-state problem. Other boundary conditions are obviously possible.

The base state we consider is a stagnant liquid layer of thickness d resting on a horizontal and isothermal plate. The upper surface of the liquid layer is free. At time zero, the layer is impulsively cooled by a constant outwardly directed heat flux Q (the product of the latent heat of vaporization and the mass flux of evaporation). Carslaw & Jaeger (1959, p. 113) give the solution for the dimensionless penetrative profile

$$\overline{T} = T_0 - z + \frac{8}{\pi^2} \sum_{m=0}^{\infty} \frac{(-1)^m}{(2m+1)^2} \exp\left(\frac{-(2m+1)^2 \pi^{2t}}{4}\right) \sin\frac{(2m+1)\pi z}{2}, \quad (3.1)$$

where  $T_0$  is the initial (dimensionless) temperature of the fluid layer. We have used the scalings

$$\{r, t, T\} = \{d, d^2/\kappa, Qd/k\}.$$
(3.2)

 $\kappa$  and k are the thermal diffusivity and conductivity of the liquid, respectively.

For the disturbances, let the scalings be

$$\{\mathbf{r}, t, \mathbf{u}, p, \Theta\} = \{d, d^2/\kappa, \kappa/d, \kappa\mu/d^2, Qd/k\}.$$
(3.3)

 $\rho$  and  $\nu = \mu/\rho$  are the liquid density and kinematic viscosity, respectively, and  $\mathbf{u} = (u, v, w)$ . The disturbances then satisfy the dimensionless nonlinear equations

$$\sigma^{-1}(\partial \mathbf{u}/\partial t + \mathbf{u} \cdot \nabla \mathbf{u}) = -\nabla p + \nabla^2 \mathbf{u}, \qquad (3.4a)$$

$$\partial \Theta / \partial t + \mathbf{u} \cdot \nabla \Theta + w \partial T / \partial z = \nabla^2 \Theta, \qquad (3.4b)$$

$$\nabla \mathbf{.} \, \mathbf{u} = 0, \tag{3.4c}$$

with boundary conditions

$$\frac{\partial u}{\partial z} = -Ma \frac{\partial \Theta}{\partial x}, \quad \frac{\partial v}{\partial z} = -Ma \frac{\partial \Theta}{\partial y}, \quad (3.5a, b)$$

$$w = 0, \qquad (3.5c)$$

$$\partial \Theta / \partial z = L \Theta$$
 (3.5d)

at 
$$z = 1$$
 and  $\mathbf{u} = 0$ ,  $\Theta = 0$  (3.5  $e, f$ )

at z = 0. We have defined the following familiar dimensionless groups: the Prandtl number  $\sigma = \nu/\kappa$ , the Marangoni number  $Ma = (-Qd^2/\mu k\kappa) \partial \gamma/\partial T$  and the Biot number L = Qd/k.  $\partial \gamma/\partial T$  is the (constant) variation of surface tension with temperature. Equations (3.5*a*, *b*) express the balance of surface-tension forces due to temperature variation, and viscous stress in the adjacent liquid.

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Equation (3.5c) states that the interface is not allowed to deform. This approximation is made here for simplicity in analysis, and it is justified for fluids of sufficiently high surface tension and density (relative to the adjacent upper phase); cf. Zeren & Reynolds (1972). In particular, in a linear stability analysis Smith (1966) explicitly demonstrated that deformation does not appreciably alter the theoretical results for most liquids. In (2.5d), the Biot number L at the free interface will henceforth be set equal to zero to simulate the conditions already discussed.

As in I and II, we now use (3.4) and (3.5) to derive the energy identity (2.1). Taking the scalar product of **u** with (3.4a), multiplying (3.4b) by  $\Theta$  and integrating over the layer, we have (see Davis (1969) for notation)

$$\frac{\sigma^{-1}}{2}\frac{\partial\langle |\mathbf{u}|^2\rangle}{\partial t} = -\langle \nabla \mathbf{u} : \nabla \mathbf{u} \rangle - Ma \int_{z=1} \Theta \frac{\partial w}{\partial z}, \qquad (3.6a)$$

$$\frac{1}{2}\partial \left\langle \Theta^2 \right\rangle / \partial t = -\left( \Theta w \,\partial T / \partial z \right\rangle - \left\langle \left| \nabla \Theta \right|^2 \right\rangle. \tag{3.6b}$$

We define a disturbance modulus  $E = \frac{1}{2} \langle |\mathbf{u}|^2 \sigma^{-1} + \lambda \Theta^2 \rangle$ , which for  $\lambda \ge 0$  is positive definite. E satisfies the energy equality

$$\frac{dE}{dt} = -\langle \nabla \mathbf{u} : \nabla \mathbf{u} \rangle - \lambda \langle |\nabla \Theta|^2 \rangle - Ma \int_{z=1} \Theta \frac{\partial w}{\partial z} - \lambda \left\langle \Theta w \frac{\partial T}{\partial z} \right\rangle.$$
(3.7)

It is enlightening to survey (3.7) in order to understand the dynamics of the instability. The first two terms, representing dissipation and conduction, are negative definite and so act to decrease the disturbance norm. The third term is the input of surface energy to the disturbance: it must be positive if there is to be an unstable disturbance. This is easily seen since Ma is typically positive (since  $\partial \gamma / \partial T < 0$ ) and for upwelling fluid at z = 1,  $\partial w / \partial z < 0$  and  $\Theta < 0$ . Similarly, for downwelling fluid at z = 1,  $\partial w / \partial z > 0$  and  $\Theta < 0$ . Using similar reasoning we can deduce that the last term in (2.9) is an energy input since for upwelling fluid  $\Theta, w < 0$ . We note that  $\partial T / \partial z < 0$  in either case.

Equation (3.7) may be cast into a symmetric form using the rescaling  $\phi = \Theta \lambda^{\frac{1}{2}}$ and  $\eta = \lambda/Ma$ . Thus,

$$dE/dt = -\mathscr{D} + Ma^{\frac{1}{2}}I_{\eta}(t), \qquad (3.8a)$$

(3.8b)

$$\mathscr{D} = \langle 
abla \mathbf{u} \colon 
abla \mathbf{u} + |
abla \phi|^2 
angle$$

where

$$I_{\eta}(t) = -\left(\eta^{\frac{1}{2}} \left\langle \phi w \frac{\partial T}{\partial z} \right\rangle + \eta^{-\frac{1}{2}} \int_{z=1} \phi \frac{\partial w}{\partial z} \right).$$
(3.8c)

We thus arrive at a form compatible with our general discussion in §2. Computationally however, it was found to be more convenient to work with the Euler-Lagrange equations in their asymmetric form, which results from the scaling  $\phi = \Theta \lambda^{\frac{1}{2}}$  with  $\lambda$  retained as the coupling parameter. The two forms are obviously equivalent, so that all of the results of §2 hold.



FIGURE 3. The stability curve for convection driven by surface tension. ——, strong stability limit;  $\odot$ , Vidal's experiments; A, 3 mm deep propyl alcohol; B, 1 mm deep methyl alcohol; C, 1 mm deep acetone.

# Strong stability

The strong stability limits are solutions to the mini-max problem

$$\tilde{\rho}(t) = \max_{\lambda > 0} \min_{h} \left( D/I_{\lambda}(t) \right),$$

$$h = \{ \mathbf{u}, \phi | \nabla \cdot \mathbf{u} = 0, \quad w = 0 \quad \text{at} \quad z = 1, \quad \mathbf{u} = \Theta = 0 \quad \text{at} \quad z = 0 \}.$$
(3.9)

It is convenient to determine the  $\rho_{\lambda}(t)$  as eigenvalues of the Euler-Lagrange equations corresponding to (3.9). These can be shown to reduce to

$$\nabla^4 w - \frac{\lambda^{\frac{1}{2}}}{2} \frac{\partial T}{\partial z} \nabla_1^2 \phi = 0, \qquad (3.10a)$$

$$\nabla^2 \phi - \frac{\lambda^{\frac{1}{2}} \partial T}{2 \partial z} w = 0, \qquad (3.10b)$$

with the natural boundary conditions

$$\rho_{\lambda}^{2} \partial w / \partial z + 2\lambda^{\frac{1}{2}} \partial \phi / \partial z = 0, \qquad (3.11a)$$

$$\rho_{\lambda}^{2} \nabla_{1}^{2} \phi - 2\lambda^{\frac{1}{2}} \partial^{2} w / \partial z^{2} = 0 \qquad (3.11b)$$

at z = 1 together with w = 0 at z = 1 and  $w = \partial w/\partial z = \phi = 0$  at z = 0. We note that (3.11) bear a close relation to the dynamic conditions (3.5*a*, *b*), but not the thermal condition (3.5*d*). Since (3.10) and (3.11) are cyclic in the directions normal to the z co-ordinate, we can Fourier decompose  $(w, \phi)$  into modes characterized by a single wavenumber  $\alpha$ . We then seek  $\tilde{\rho}(t) = \max_{\lambda} \min_{\alpha} \rho_{\lambda}(t)$ . The stability-region boundary is given by  $Ma(t) = \tilde{\rho}^2(t)$ . The solution for  $\rho_{\lambda}(t)$  was accomplished using an adaptation of a Galerkin technique due to Nield (1964). The development is traced in the

appendix. The resulting generalized algebraic eigenvalue problem was solved using a method due to Moler & Stewart (1973).

The results of this study are presented in figure 3. The remarkable result is that the energy theory yields bounds on the onset time. An experimental trajectory is horizontal on figure 3. Hence there is stability until that time at which the stability boundary is crossed. That time of crossing is a lower bound on the experimentally observed onset time. More important, the results also indicate that the analysis of §2 may apply to a wide class of accelerated flows.

At large times the strong stability limit of our work must agree with the steady-state results of Davis (1969). Our limit of Ma = 55.75 compares well with his result of 56.77 (in the limit of zero Rayleigh number). Our results are thus believed to be accurate to within 2 %.

#### Marginal stability

The Euler–Lagrange equations corresponding to (2.11) may be manipulated to yield

$$\nabla^4 w - \frac{\lambda^{\frac{1}{2}} dT}{2 dz} \nabla_1^2 \phi = \frac{\nu_\lambda(t)}{2\sigma} \nabla^2 w, \qquad (3.12a)$$

$$-\nu_{\lambda}(t)\phi + 2\nabla^2\phi - \lambda^{\frac{1}{2}}(dT/dz)w = 0 \qquad (3.12b)$$

and natural boundary conditions

$$2\lambda^{\frac{1}{2}} \partial \phi / \partial z + Ma \, \partial w / \partial z = 0, \qquad (3.13a)$$

$$Ma\nabla_1^2 \phi - 2\lambda^{\frac{1}{2}} \partial^2 w / \partial z^2 = 0 \tag{3.13b}$$

at z = 1, in addition to w = 0 at z = 1 and  $w = \partial w / \partial z = \phi = 0$  at z = 0.

We note that, as in II, the introduction of a weaker criterion for stability reintroduces the Prandtl number  $\sigma$  as a parameter of the problem.

The computation of the marginal stability boundary is involved, so we detail the steps here. As in the strong stability formulation  $(w, \phi)$  may be a Fourier decomposed by introducing a wavenumber  $\alpha$ . For any given  $\alpha$ , Ma,  $\lambda$  and  $\sigma$ , the function  $\nu_{\lambda}$   $(t; \alpha, Ma, \sigma)$  may be determined by the solution of a generalized eigenvalue problem; see appendix. Referring to (2.14), the problem is then to find the value of  $t^{\dagger}(\lambda; Ma, \alpha, \sigma)$  for which

$$\int_0^{t^{\dagger}} \nu_{\lambda}(s) \, ds = 0 \tag{3.14}$$

for the most dangerous Fourier mode, i.e. we seek min  $t^{\dagger}(\lambda; Ma, \alpha, \sigma)$ . We em-

phasize that this most dangerous Fourier mode may have no relation to the so-called 'wavenumber of maximum growth rate' of linear amplification theory, since the maximizing  $(w, \phi)$  need not be a solution to the dynamic equations.



FIGURE 4. The net amplification as a function of  $\alpha$  and t.

	$t^*$	$ ilde{t}^{\dagger}$	$t_{exp}$
$Ma = 421, \sigma = 1$	0.01	0.02	
$Ma = 618, \sigma = 30$ (propanol)	0.006	0.012	0.08
$Ma = 2023, \sigma = 6.6$ (methanol)	0.001	0.003	0.7

TABLE 1. A comparison of stability bounds and experimental onset times

The 'optimal' lower bound on the onset time is found by suitably varying  $\lambda$ , viz.

$$\tilde{t}^{\dagger} = \max_{\lambda} \min_{\alpha} t^{\dagger}(\lambda; Ma, \alpha, \sigma).$$

Since (3.14) involves the unknown  $t^{\dagger}$ , it was necessary to adopt the following iterative procedure. For fixed  $(Ma, \sigma)$ , the 'amplification'

$$\overline{\nu}_{\lambda} \equiv \int_0^t \nu_{\lambda}(s) \, ds$$

was determined as a function of  $\alpha$  and t. Three-point Gaussian quadrature over (0, t) was found to be sufficiently accurate. The main features of the results are depicted schematically in figure 4. The results for t near  $t^{\dagger}$  clearly indicate a most dangerous Fourier mode. The value of  $t^{\dagger}$  is that for which  $\bar{\nu}_{\lambda}$  first becomes zero for any  $\alpha$ . The process was then repeated for different  $\lambda$  to determine  $\tilde{t}^{\dagger}$ .

Because of the large number of computations necessary to determine the

marginal stability curve, we have computed  $\tilde{t}^{\dagger}$  for a limited number of cases only. These are given in table 1. The first point (Ma = 421,  $\sigma = 1$ ) was chosen as representative of the improvement for moderate Marangoni numbers. The other two cases are for parameter values corresponding to the available experimental results of Vidal. It is seen that  $\tilde{t}^{\dagger} \ge t^*$ , as must be the case, and furthermore, assuming these comparisons to be typical, one can obtain large improvements in the bounds with the marginal stability criterion in situations where its use is appropriate.

#### 4. Discussion and conclusions

The stability curve is presented in figure 3. This curve should asymptotically approach Davis' (1969) strong stability limit. This was verified in the current work. It is interesting to note that the frozen-time results of Vidal & Acrivos (1968) (not shown) nearly overlay the strong stability limits presented here.

A number of experimental points are shown in figure 3 and in table 1. These results are from Vidal (1967) and Vidal & Acrivos (1968) and are selected so that the buoyancy effect is minimal. The abscissae of these points are only approximate: in Vidal & Acrivos (1968) the onset time for 3 mm deep propyl alcohol is stated to be  $8 \pm 1$  s. The onset times for methyl alcohol and acetone (1 mm deep pools) are from Vidal (1967). They are not explicitly reported in that work, so we equated them to that time at which the surface temperature deviates from the predictions of the conductive solution.

The results of the experiments are seen to be in agreement with the theory in the sense that none of the experimentally determined onset times lies to the left of the strong stability limit. The comparisons made in table 1 also indicate that the marginal stability criterion provides bounds for the onset time in this context. If this improvement in the bound is typical, this weaker definition of 'stability' may indeed find some utility. Owing to the lack of good experimental data, those conclusions must necessarily remain tentative. It can be said, however, that the energy method can be successfully applied to uniformly accelerated flows to yield lower bounds on that elusive quantity the 'onset time'.

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# Appendix. Expansion technique for the impulsive cooling problem

In this appendix we present the solution method, which is an adaptation of Nield's (1964) Fourier sine-series method.

#### Strong stability

The Euler-Lagrange equations are first Fourier decomposed in the directions normal to the z axis. This introduces the wavenumber  $\alpha$ :

$$(D^2 - \alpha^2)^2 w + \frac{\lambda^{\frac{1}{2}}}{2} \frac{\partial T}{\partial z} \alpha^2 \phi = 0, \qquad (A \, 1 \, a)$$

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$$(D^2 - \alpha^2) \phi - \frac{\lambda^{\frac{1}{2}}}{2} \frac{\partial T}{\partial z} w = 0, \qquad (A \, 1 \, b)$$

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where  $D = \partial/\partial z$ . The boundary conditions become

$$\begin{array}{l} (\rho_{\lambda}^{2}/2\lambda^{\frac{1}{2}}) Dw + D\phi = 0\\ (\rho_{\lambda}^{2}/2\lambda^{\frac{1}{2}}) \alpha^{2}\phi + D^{2}w = 0\\ w = 0 \end{array} \right\} \quad \text{at} \quad z = 1,$$
 (A2a)

$$w = Dw = \phi = 0 \quad \text{at} \quad z = 0. \tag{A2b}$$

We want to expand both  $\phi$  and w in a sine series. However, if we differentiate term by term, the expansion will not approximate the odd derivatives of the function unless the preceding derivative vanishes at the end points z = 0, 1. See Jeffreys & Jeffreys (1946, §14.062). For the derivatives of interest here, if the expansions are to be valid approximations then the following conditions must be satisfied:

$$w(0) = w(1) = \phi(0) = \phi(1) = D^2 w(1) = D^2 w(0) = 0.$$
 (A 3)

The first three conditions are satisfied but the last three are not.

Consequently, to use a sine expansion it is necessary to introduce two auxilliary functions  $\phi^*$  and  $w^*$  satisfying the conditions (A 3):

$$\phi^* = \phi - \phi(1)z = \sum_{n=1}^{\infty} \phi_n \sin n\pi z, \qquad (A4a)$$

$$w^* = w + p_1(z) D^2 w(0) - p_2(z) D^2 w(1) = \sum_{n=1}^{\infty} w_n \sin n\pi z, \qquad (A4b)$$

where

$$p_1(z) = \frac{1}{6}(z^3 - 3z^2 + 2z), \quad p_2(z) = \frac{1}{6}(z^3 - z).$$
 (A4*c*, *d*)

Rearranging (A4) gives

$$\phi = \phi(1)z + \sum_{n=1}^{\infty} \phi_n \sin n\pi z, \qquad (A \, 5a)$$

$$w = -p_1(z) D^2 w(0) + p_2(z) D^2 w(1) = \sum_{n=1}^{\infty} w_n \sin n\pi z.$$
 (A 5b)

The Galerkin expansion now proceeds by substituting the above expansions into (A1) and the boundary conditions, and making the residuals orthogonal to each of the expansion functions  $\sin m\pi z$ . (Note that this is equivalent to Nield's approach of first expanding z,  $p_2(z)$  and  $p_1(z)$  in their sine series.)

Using the definition

$$\langle u, v \rangle = 2 \int_0^1 uv \, dz \tag{A 6}$$

we generate the following algebraic set for the  $w_n, \phi_n,$  etc.:

$$\begin{split} & [(m\pi)^2 + \alpha^2]^2 w_m + 2\alpha^2 [-\langle 1-z, \sin m\pi z \rangle D^2 w(0) - \langle z, \sin m\pi z \rangle D^2 w(1)] \\ & + \alpha^4 \langle p_2, \sin m\pi z \rangle D^2 w(1) - \alpha^4 \langle p_1, \sin m\pi z \rangle D^2 w(0) \\ & + \frac{\alpha^2 \lambda}{2} \bigg[ \sum_n \phi_n \left\langle \frac{\partial T}{\partial z} \sin n\pi z, \quad \sin m\pi z \right\rangle + \phi(1) \left\langle \frac{\partial T}{\partial z} z, \quad \sin m\pi z \right\rangle \bigg] = 0, \quad (A7a) \end{split}$$

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$$-\left[(m\pi)^{2} + \alpha^{2}\right]\phi_{m} - \alpha^{2}\langle z, \sin m\pi z \rangle \phi(1) - \frac{1}{2}\lambda^{\frac{1}{2}} \langle p_{2}(z) \partial T / \partial z, \sin m\pi z \rangle D^{2}w(1) + \frac{1}{2}\lambda^{\frac{1}{2}} \langle p_{1}(z) \partial T / \partial z, \sin m\pi z \rangle D^{2}w(0) - \frac{\lambda^{\frac{1}{2}}}{2} \sum_{n} w_{n} \left\langle \frac{\partial T}{\partial z} \sin n\pi z, \sin m\pi z \right\rangle = 0.$$
 (A 7b)

The three boundary conditions not identically satisfied yield

$$\sum_{n} (n\pi) w_n + D^2 w(1) Dp_2(0) - D^2 w(0) Dp_1(0) = 0, \qquad (A7c)$$

$$\sum_{n} (n\pi) (-1)^{n} \phi_{n} + \phi(1)$$

$$= -\frac{\rho_{\lambda}^{2}}{2\lambda^{\frac{1}{2}}} [\sum_{n} (-1)^{n} (n\pi) w_{n} + D^{2} w(1) Dp_{2}(1) - D^{2} w(0) Dp_{1}(1)], \quad (A7d)$$

$$D^{2} w(1) = -(\alpha^{2}/2)^{\frac{1}{2}} \alpha^{2} \phi(1) \qquad (A7d)$$

$$D^2 w(1) = -\left(\rho_{\lambda}^2/2\lambda^{\frac{1}{2}}\right) \alpha^2 \phi(1). \tag{A 7e}$$

With  $\lambda$  given, (A7) pose a generalized algebraic eigenvalue problem for  $\rho_{\lambda}$ . If the order of the Galerkin expansion is N, then the problem takes the form

 $\mathbf{A}\mathbf{x} = \rho_{\lambda} \mathbf{B}\mathbf{x},$ 

where **A** and **B** are square matrices of order 2N + 3, **B** is singular and has rank 2, and **x** is the column vector  $\mathbf{x} = [w, w_2, ..., w_N, \phi_1, \phi_2, ..., \phi_N, D^2w(1), D^2w(0), \phi(1)]^T$ . This problem is most conveniently solved by the method of Moler & Stewart (1973).

# Marginal stability

Equations (3.12a, b) and (3.13a, b) are Fourier decomposed to give, respectively,

$$(D^2 - \alpha^2)^2 w + \frac{\lambda^{\frac{1}{2}}}{2} \frac{\partial T}{\partial z} \alpha^2 \phi = \frac{\nu_{\lambda}(t)}{2\sigma} (D^2 - \alpha^2) w, \qquad (A \, 8 \, a)$$

$$2(D^2 - \alpha^2)\phi - \lambda^{\frac{1}{2}} \partial T / \partial z w = v(t)\phi, \qquad (A 8b)$$

$$2\lambda^{\frac{1}{2}}\partial\phi/\partial z + Ma\,\partial w/\partial z = 0, \qquad (A\,9a)$$

$$Ma\alpha^2\phi + 2\lambda^{\frac{1}{2}}\partial^2 w/\partial z^2 = 0.$$
 (A9b)

The expansions in (A5) are also used for this formulation. Substitution and orthogonalization result in the following relations. Equation (A8a) becomes

$$\begin{split} &[(m\pi)^2 + \alpha^2]^2 w_m - 2\alpha^2 (D^2 w(0) \langle 1 - z, \sin m\pi z \rangle + D^2 w(1) \langle z, \sin m\pi z \rangle) \\ &+ D^2 w(1) \alpha^4 \langle p_2, \sin m\pi z \rangle - D^2 w(0) \alpha^4 \langle p_1, \sin m\pi z \rangle \\ &+ \frac{\lambda^{\frac{1}{2}}}{2} \alpha^2 \sum_n \phi_n \left\langle \frac{\partial T}{\partial z} \sin m\pi z, \sin n\pi z \right\rangle + \frac{\lambda^{\frac{1}{2}}}{2} \alpha^2 \phi(1) \left\langle z \frac{\partial T}{\partial z}, \sin m\pi z \right\rangle \\ &= (\nu_\lambda(t)/2\sigma) \left[ -((m\pi)^2 + \alpha^2) w_m + D^2 w(1) \left( \langle z, \sin m\pi z \rangle - \alpha^2 \langle p_2, \sin m\pi z \rangle \right) \\ &+ D^2 w(0) \left( \langle (1 - z), \sin m\pi z \rangle + \alpha^2 \langle p_1, \sin m\pi z \rangle \right). \end{split}$$

Equation (A8b) results in

$$-2[(m\pi)^{2} + \alpha^{2}] \phi_{m} - 2\alpha^{2} \phi(1) \langle z, \sin m\pi z \rangle - \lambda^{\frac{1}{2}} \sum_{n} w_{n} \left\langle \frac{\partial T}{\partial z} \sin n\pi z, \sin m\pi z \right\rangle$$
$$-\lambda^{\frac{1}{2}} D^{2} w(1) \langle p_{2} \partial T / \partial z, \sin m\pi z \rangle + \lambda^{\frac{1}{2}} D^{2} w(0) \langle p_{1} \partial T / \partial z, \sin m\pi z \rangle$$
$$= \nu_{\lambda}(t) [\phi_{m} + \phi(1) \langle z, \sin m\pi z \rangle].$$

The conditions Dw(0) = 0, (A9a) and (A9b) becomes respectively

$$\begin{split} \sum_{n} (n\pi) w_n - \frac{1}{6} D^2 w(1) - \frac{1}{3} Dw(0) &= 0\\ \sum_{n} (n\pi) (-1)^n \phi_n + \phi(1) + \frac{Ma}{2\lambda^{\frac{1}{2}}} \sum_{n} (-1)^n (n\pi) w_n, \\ &+ \frac{Ma}{6\lambda^{\frac{1}{2}}} \frac{D^2 w(1)}{3} + \frac{Ma}{12\lambda^{\frac{1}{2}}} D^2 w(0) = 0,\\ 2D^2 w(1) + Ma\lambda^{-\frac{1}{2}} \alpha^2 \phi(1) &= 0. \end{split}$$

The eigenvalue  $\nu_{\lambda}(t)$  in these equations is found in the same way as  $\rho_{\lambda}(t)$  in the strong stability formulation.

The inner products in the equations in this appendix were all obtained analytically.

The strong stability results, as mentioned in §3 above, are believed to be accurate to within 2%. It was necessary to carry the Galerkin expansion to 30 terms to achieve this accuracy. The solution for the base state temperature field, equation (3.1), was carried to as many terms as were necessary to achieve a term-to-term variation of less than 0.1% in  $\partial T/\partial z$  throughout the layer.

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