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Buoyancy–thermocapillary instability: the role of interfacial deformation in one- and two-component fluid layers heated from below or above

By J. L. CASTILLO AND M. G. VELARDE

Departamento Física Fundamental – U.N.E.D., Apdo Correos 50487, Madrid, Spain

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Energy stability theory has been used to study Bénard convection in one- and two-component horizontal fluid layers heated from below or above when there is a deformable upper surface. To a first approximation in the crispation number, we provide sufficient conditions for stability of the motionless state of the layer, and delineate regions of possible subcritical instability.

1. Introduction

Surface-tension forces play a major role in a low-gravity environment, and new impetus in analysing their influence on materials processing stems from space programmes. Scientifically and technologically significant results are expected from low-gravity melt processing in the near future. Metal-melting experiments in SKYLAB (Bourgeois & Brashears 1977) gave evidence of surface-tension-driven cellular convection, and it is generally well established that Marangoni–Bénard effects are responsible for triggering steady and time-dependent flows in crystal-growth melts with free surfaces (Schwabe 1981). On the other hand there is growing evidence of the role played by mere p.p.m. impurities in drastically affecting Bénard–Marangoni convection (Coriell *et al.* 1980; Hurle 1977). Recently, double-diffusive convection with free surfaces has been highlighted as one of the problems relevant to space processing of liquids under thermal constraints (Malméjac *et al.* 1981). Moreover, interface stability can be a measure of material purity (Fisher & Kurz 1980; Coulet, Billia & Capella 1981).

In a recent paper, Davis & Homsy (1980) have studied the role of interfacial deformation in the stability of a horizontal fluid layer heated from below and open to the ambient air They found that a deformable interface leads to a stabilization relative to the case of a planar interface. In the present paper we extend some of their results by considering a fluid layer heated from above (negative Rayleigh number R) and a fluid where the density depends on two variables such as temperature and impurity (or solute) concentration (Shir & Joseph 1968; Schechter, Velarde & Platten 1974). In our study, solutal and thermal effects can be described with the same analytical theory with the only exception that the solutal Marangoni number or elasticity number E does not generally follow the sign of the corresponding solutal Rayleigh number R_s. In contradistinction to the thermal Marangoni number M, the elasticity number can have either sign, positive or negative, for a given sign of the solutal Rayleigh number. Thus, the results reported here extend on the one hand the work of Davis & Homsy (1980) to binary mixtures, and on the other they extend the work of Shir & Joseph (1968) to include capillary phenomena. We discuss the

competition between the two Marangoni numbers, with emphasis on the case of vanishing gravity, as this should be the most relevant limiting case in experiments aboard spacecraft.

An interesting feature of binary mixtures is the occurrence of instability in liquid layers even when the vertical density distribution is *statically* stable (Turner 1973), no matter how small a gravitational field exists. This may be the case when the layer is heated from above and a solute less dense than a solvent accumulates at the bottom. An instability is possible owing to the large disparity between the thermal and mass diffusivities in the mixture. A parcel of fluid displaced from the bottom upwards rather quickly warms up, but scarcely changes its solute concentration. Thus it still tends to rise and convection sets in provided that the viscous forces are overcome (Turner 1973; Velarde 1977; Velarde & Normand 1980).

In the present paper we discuss the possibility of steady and time-dependent convection (overstability) and subcritical instability when a binary liquid mixture is subjected to thermal constraints and there is a free surface open to the ambient air.

2. Two-dimensional buoyancy–thermocapillary problem

We shall remain as close as possible to the formulation of the problem given by Davis & Homsy (1980). We shall use the following notation (figure 1): d is the mean distance between two infinite horizontal surfaces; the lower surface is a rigid heat-conducting plate at constant temperature, while the upper surface is free, and open to the ambient air on which the heat flux is prescribed. These surfaces bound an incompressible, Newtonian and Boussinesq (Pérez Cordón & Velarde 1975; Velarde & Pérez Cordón 1976) liquid of mean density ρ, viscosity μ and kinematic viscosity $\nu = \mu/\rho$. The acceleration due to gravity is g; α and γ are respectively the coefficients of thermal and solutal expansion; χ and D are respectively the thermal and mass diffusivity of the liquid mixture. The ambient air plays a passive role and it is assumed to have negligible density and viscosity. The free surface can be deformable and has no uniform temperature distribution.

We use a system of Cartesian coordinates whose origin lies in the rigid plate and whose dimensionless coordinates x and z are scaled on d. We thus consider two-dimensional fields. The velocity vector $\mathbf{v} = (u, w)$, the temperature θ, the mass fraction of one of the components (one, say, the solvent or the solute and the heavier in the binary mixture) Γ, the time t, the pressure p, and the surface tension σ are referred to scales χ/d, ΔT, ΔN_1 (with $N_i = \rho_i/\rho$, $i = 1, 2$, and $N_1 + N_2 = 1$), d^2/χ, $\mu\chi/d^2$, σ_0, where ΔT is the temperature excess or defect at the bottom compared with the top and σ_0 is the *mean* surface tension on the free surface. Under the above assumptions and conventions, the following dimensionless groups are introduced:

thermal Rayleigh number $$R = \frac{\alpha g d^3 \Delta T}{\chi \nu},$$

solutal Rayleigh number $$R_s = \frac{-\gamma g d^3 \Delta N_1}{D\nu},$$

Prandtl number $$P = \frac{\nu}{\chi},$$

(inverse) Lewis number $$L = \frac{D}{\chi},$$

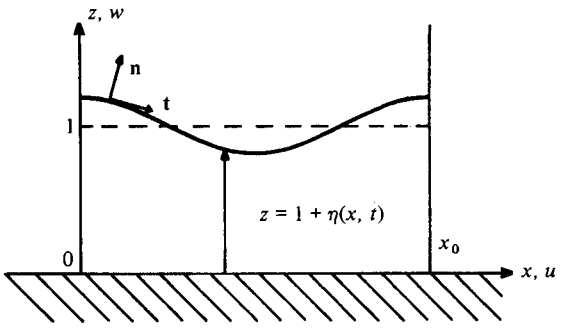

FIGURE 1. An exaggerated view of the interfacial deformation in a single cell. Boundary conditions are taken at $z = 0$ and $z = 1+\eta(x,t)$.

crispation or capillary number $C = \dfrac{\mu\chi}{\sigma_0 d}$,

Bond number $G = \dfrac{\rho g d^2}{\sigma_0}$,

thermal Marangoni number $M = -\left(\dfrac{\partial\sigma}{\partial T}\right)\dfrac{d\Delta T}{\mu\chi}$,

solutal Marangoni or elasticity number $E = -\left(\dfrac{\partial\sigma}{\partial N_1}\right)\dfrac{d\Delta N_1}{\mu D}$.

R, R_s, M and E are dimensionless measures of the thermal constraints operating in the fluid layer. These constraints induce positive or negative buoyancy and variations of surface tension, thus leading to surface tractions and eventual deformation of the open interface. The capillary number C measures the degree of deformability, and the limit of vanishing capillary number reduces the problem to a plane boundary, which is the case of an interface with mean surface tension very large. In this note we focus on the role of C to a first approximation.

We locate the liquid–air interface $S(t)$ by writing

$$S(t): z = 1+\eta(x,t). \tag{1}$$

For a sufficiently smooth general two-dimensional deformation of the interface, the unit outward normal vector $\mathbf{n}$ to S has the form

$$\mathbf{n} = \left(\frac{-\partial\eta}{\partial x}, 1\right)\Big/N, \tag{2}$$

and the unit tangent vector $\mathbf{t}$ to S

$$\mathbf{t} = \left(1, \frac{\partial\eta}{\partial x}\right)\Big/N, \tag{3}$$

where $N = \{1+(\partial\eta/\partial x)^2\}^{\frac{1}{2}}$. Thus the curvature $K(\eta)$ is

$$K(\eta) = \frac{1}{N^3}\frac{\partial^2\eta}{\partial x^2}. \tag{4}$$

The stress balance at the open interface in compact notation is

$$\tau_{ij}n_j = -\frac{G}{C}\{\eta+\tfrac{1}{2}A\eta^2\}n_i + \frac{K}{C}\{1-MC(\theta-\eta)-ECL(\Gamma-\eta)\}n_i$$
$$-t_i(\mathbf{t}.\nabla)\{M(\theta-\eta)+EL(\Gamma-\eta)\} \quad (i,j=1,2), \tag{5}$$

where $A = \alpha\Delta T - \gamma\Delta N_1$ is the group that must remain small and with small parts for the Boussinesquian approximation to be valid (Pérez Cordón & Velarde 1975): A is not independent of the dimensionless groups introduced earlier. Rather, we have $A = \{R + R_s L\}\, C/G$, which must be vanishingly small for given R and R_s. The stress tensor of the liquid is

$$\tau_{ij} = -p\delta_{ij} + \epsilon_{ij} \quad \text{with} \quad \epsilon_{ij} = \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i};$$

δ_{ij} is the Kronecker delta. The summation convention over repeated indices is assumed.

We assume that the liquid at the interface moves with the interface's velocity, i.e. the kinematic boundary condition at the open interface takes the form

$$\frac{\partial \eta}{\partial t} = N v_i n_i \quad \text{on} \quad z = 1 + \eta. \tag{6}$$

As indicated earlier, we assume that the heat flux is prescribed on the open interface, i.e. the interface is a rather poor heat-conducting boundary. We have

$$(\mathbf{n} \,.\, \nabla)\theta = \frac{1-N}{N}. \tag{7}$$

Thus the value of this flux is fixed at the value in the motionless steady state. For solute transport we also prescribe the value of the flux. An alternative condition would be to prescribe the value of the salt concentration at the interface, but for simplicity we shall not consider this case here. We take

$$(\mathbf{n} \,.\, \nabla)\Gamma = \frac{1-N}{N}. \tag{8}$$

On the rigid lower plate, the temperature and salt concentration are fixed:

$$v_i = \theta = \Gamma = 0 \quad \text{on} \quad z = 0. \tag{9}$$

Finally, the equations for the disturbances upon the motionless steady state of the bulk liquid enclosed in the region $0 \leqslant z \leqslant 1 + \eta(x, t)$, $-\infty < x < \infty$, with $t \geqslant 0$, are

$$P^{-1}\left(\frac{\partial v_i}{\partial t} + (\mathbf{v} \,.\, \nabla)\, v_i\right) = \frac{\partial \tau_{ij}}{\partial x_j} + R\theta k_i + R_s L \Gamma k_i, \tag{10a}$$

$$\frac{\partial \theta}{\partial t} + (\mathbf{v} \,.\, \nabla)\, \theta = \nabla^2\theta + w, \tag{10b}$$

$$\frac{\partial \Gamma}{\partial t} + (\mathbf{v} \,.\, \nabla)\, \Gamma = L\nabla^2\Gamma + w, \tag{10c}$$

$$\nabla \,.\, \mathbf{v} = 0, \tag{10d}$$

where $k_i = (0, 1)_i$.

3. Energy stability analysis

Energy theory (Davis 1969; Davis & Homsy 1981; Joseph 1976; Shir & Joseph 1968; Normand, Pomeau & Velarde 1977) provides sufficient conditions for stability of a given state of the fluid layer. It amounts to the construction of a suitable Lyapunov functional for arbitrary disturbances upon the initial state of the fluid

layer. We define the integral over the free surface of a quantity f as follows:

$$\int_{S(t)} f \equiv \int_0^{S_0(t)} f ds = \int_0^{x_0} f(z = 1+\eta)\, N dx, \tag{11}$$

where ds is an element of arclength along $S(t)$, and $S_0(t)$ is the arclength of the free surface in one period in x. Then the 'volume' integral (in two dimensions) of a quantity over one such period is

$$\langle f \rangle \equiv \int_0^{x_0} \int_0^{1+\eta(x,t)} f(x, z, t)\, dz\, dx. \tag{12}$$

Now we define the Lyapunov energy functional as follows:

$$\epsilon = P^{-1} \langle \tfrac{1}{2} v_i^2 \rangle + \lambda \langle \tfrac{1}{2} \theta^2 \rangle + \Lambda L \langle \tfrac{1}{2} \Gamma^2 \rangle + \frac{G}{C} \int_{S(t)} \frac{\eta^2 + \frac{1}{3} A \eta^3}{2N}. \tag{13}$$

Note that $|\eta| > 3/A$ for the Boussinesquian approximation to be valid. Then the fourth term on the right-hand side is positive, and the same property applies to ϵ for non-negative λ and Λ. The latter parameters are the linking parameters of the functional (Joseph 1965, 1976). Their choice is dictated by convenience to obtain the largest parameter region of stability. Our choice here departs from that of Davis & Homsy (1980), as we do not introduce the Rayleigh number in the Lyapunov energy functional. This procedure allows a straightforward discussion of positive and negative Rayleigh numbers. On the other hand the choice of the surface-integral term in (13) with the factor G permits an elimination of this parameter without, however, setting it to zero, thus overcoming the difficulties with zero-wavenumber modes at $G = 0$ (see e.g. Scriven & Sternling 1964; Smith 1966; Velarde 1977).

We search for solutions to

$$\delta \frac{d\epsilon}{dt} = 0,$$

where δ accounts for an arbitrary variation of the rate of evolution of ϵ with, however, the constraint that quantities have periodicity in x, there is conservation of volume elements ($\int_x \eta = 0$), incompressibility, and the boundary conditions are satisfied at the respective boundaries. Let β and $2p(x, z, t)$ be the relevant Lagrange multipliers. Then we pose the following variational condition:

$$\delta \left(\frac{d\epsilon}{dt} + \langle 2p \nabla . \mathbf{v} \rangle + \beta \int_x \eta \right) = 0. \tag{14}$$

From (14) we get the corresponding Euler–Lagrange equations together with their natural boundary conditions. As we are interested in the role played by the crispation number, i.e. by the deformation of the interface, we assume that all fields have a series expansion in C. In compact form we have

$$(v_i, \theta, \Gamma, \eta) = (v_i^{(0)}, \theta^{(0)}, \Gamma^{(0)}, \eta^{(0)}) + (v_i^{(1)}, \theta^{(1)}, \Gamma^{(1)}, \eta^{(1)})\, C + O(C^2) \tag{15}$$

together with a similar expansion for R, R_s, M or E. Then the Euler–Lagrange equations to the lowest-order approxmation are

$$2 \frac{\partial \tau_{ij}^{(0)}}{\partial x_j} + (R+\lambda)\, \theta^{(0)} k_i + (R_s + \Lambda)\, L \Gamma^{(0)} k_i = 0, \tag{16a}$$

$$(R+\lambda)\, w^{(0)} + 2\lambda \nabla^2 \theta^{(0)} = 0, \tag{16b}$$

$$(R_s + \Lambda)\, w^{(0)} + 2\Lambda L\, \nabla^2 \Gamma^{(0)} = 0, \tag{16c}$$

$$\nabla . \mathbf{v}^{(0)} = 0, \tag{16d}$$

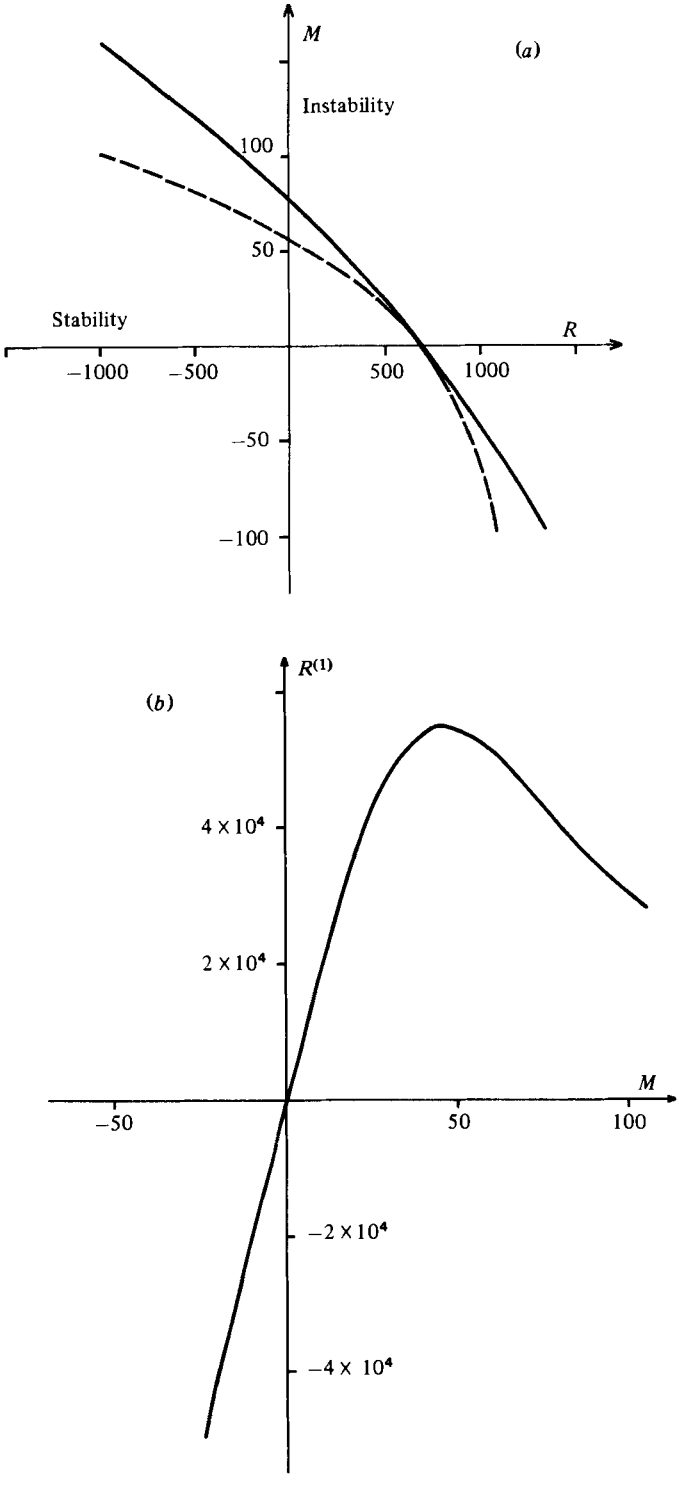

FIGURE 2. For caption see facing page.

together with the boundary conditions

$$v_i^{(0)} = \theta^{(0)} = \Gamma^{(0)} = 0 \quad \text{on} \quad z = 0. \tag{17}$$

On $z = 1$, we have the natural boundary conditions

$$w^{(0)} = 0, \tag{18a}$$

$$2\tau_{ij}^{(0)}\, n_j^{(0)}\, t_i^{(0)} + M\frac{\partial\theta^{(0)}}{\partial x} + EL\frac{\partial\Gamma^{(0)}}{\partial x} = 0, \tag{18b}$$

$$2\lambda\frac{\partial\theta^{(0)}}{\partial x} - M\frac{\partial u^{(0)}}{\partial x} = 0, \tag{18c}$$

$$2\Lambda L\frac{\partial\Gamma^{(0)}}{\partial x} - E\frac{\partial u^{(0)}}{\partial x} = 0. \tag{18d}$$

Note that when $C = 0$ we have $\eta = 0$ and there is no deformation of the interface. The solutions can be sought in the form $f(z)\exp(iax)$, where a is a Fourier wavenumber. We have

$$\theta^{(0)} = \left\{\sum_{i=1}^{6} \Omega_i \exp q_i z + \Omega_7\{\exp az - \exp -az\}\right\} \exp iax, \tag{19}$$

where q_i are the roots of the polynomial equation

$$(q_i^2 - a^2)^3 + \tfrac{1}{4}(\lambda + \Lambda)\, a^2 = 0. \tag{20}$$

Similar expressions are obtained for the remaining fields. Note that the Ω_7 term does not appear in single-component liquid layers.

4. The role of the crispation number: discussion of results

To a first-order approximation in the crispation number the solution of the problem posed in §§2 and 3 yields

$$R^1 = \frac{2(M+EL)}{a^2}\frac{\int_x \tau_{ij}^{(0)}\, n_j^{(0)}\, n_i^{(0)}\, w_z^{(0)}}{\langle\theta^{(0)} w^{(0)}\rangle}, \tag{21a}$$

or

$$R_S^{(1)} = \frac{2(ML^{-1}+E)}{a^2}\frac{\int_x \tau_{ij}^{(0)}\, n_j^{(0)}\, n_i^{(0)}\, w_z^{(0)}}{\langle\Gamma^{(0)} w^{(0)}\rangle}, \tag{21b}$$

FIGURE 2. (*a*) Marangoni versus Rayleigh number for a plane open interface (case $C = 0$). Solid and broken lines correspond respectively to linear and energy stability analyses. The region between the two lines corresponds to the possibility of subcritical instability. Note that the diagram corresponds either to the single-component Bénard problem heated from below or above, or to an isothermal horizontal layer subjected to a solutal gradient. In the latter case M and R should be replaced respectively by E and R_S. (*b*) Bénard convection with a deformable interface: first-order correction in the crispation number ($C \neq 0$) to the Rayleigh number versus Marangoni number. For a given value of M, the actual Rayleigh number provided by energy theory is given by the corresponding value in (*a*) added to $R^{(1)}C$. A similar result holds for (isothermal) solutal convection when R is replaced by R_S, M by E, and C by LC. Note that R is negative for $M > 56{\cdot}77$ and $R > 669$ for negative Marangoni numbers.

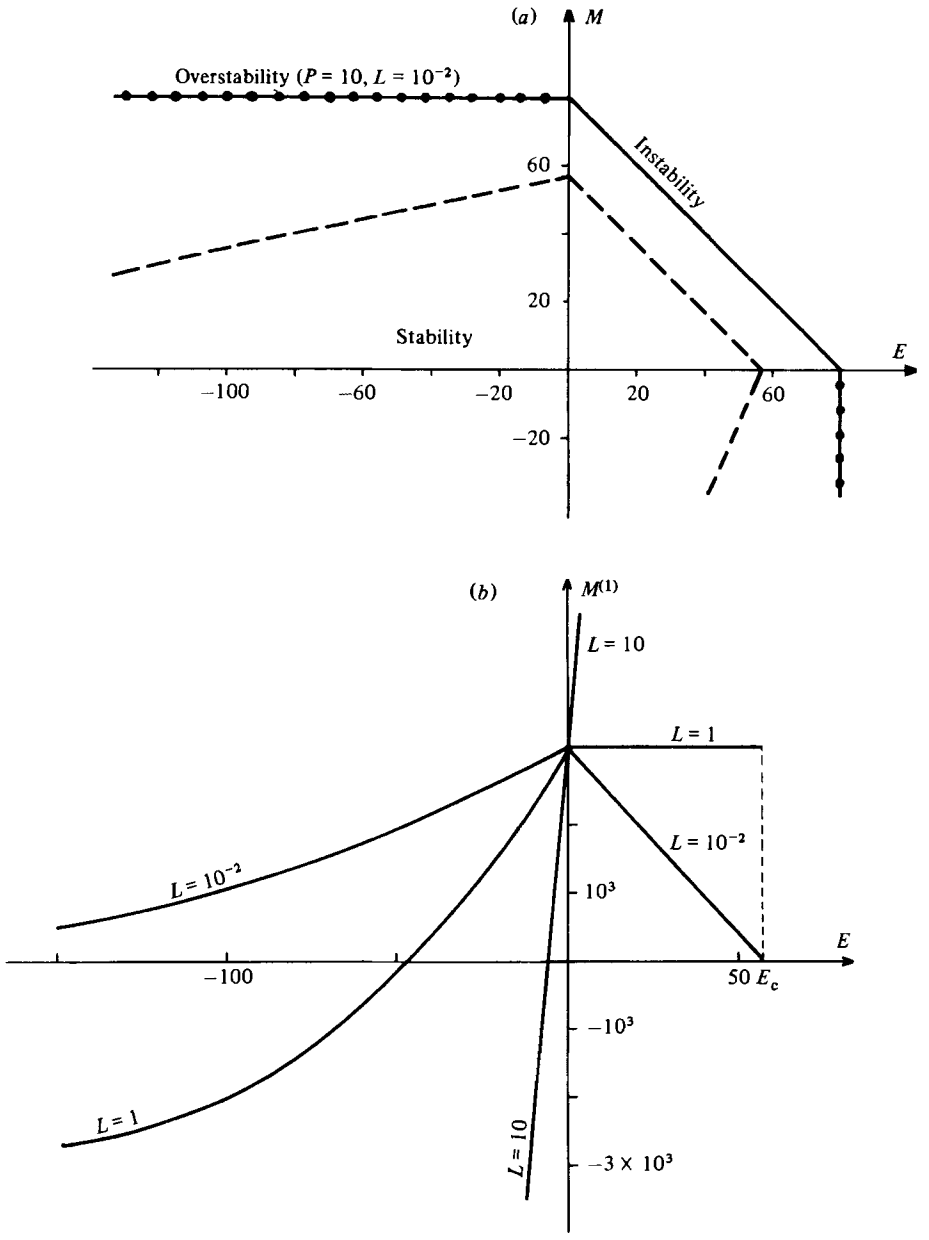

FIGURE 3. (*a*) Zero-gravity Bénard convection in a two-component fluid layer when the thermal and solutal (elasticity) Marangoni numbers compete or cooperate to induce interfacial instability. Solid and broken lines correspond respectively to energy and linear stability analyses. The dotted parts of the solid line correspond to overstability. The onset of overstability depends on Prandtl and Lewis numbers. For illustration, here we have chosen $P = 10$ and $L = 10^{-2}$. As in figure 2(*a*) the region between the two lines corresponds to the possibility of subcritical instability. (*b*) Zero-gravity Bénard convection in a binary mixture with a deformable interface: first-order corrections in the crispation number ($C \neq 0$) to the thermal Marangoni number versus solutal Marangoni (elasticity) number. For a given value of E, the actual thermal Marangoni number provided by energy theory is given by the corresponding value in (*a*) added to $M^{(1)}C$.

or

$$M^{(1)} = -\frac{2(M+EL)}{a^2}\frac{\int_x \tau_{ij}^{(0)} n_j^{(0)} n_i^{(0)} w_z^{(0)}}{\int_x \theta^{(0)} w_z^{(0)}}, \tag{21c}$$

or

$$E^{(1)} = -\frac{2(ML^{-1}+E)}{a^2}\frac{\int_x \tau_{ij}^{(0)} n_j^{(0)} n_i^{(0)} w_z^{(0)}}{\int_x \Gamma^{(0)} w_z^{(0)}}, \tag{21d}$$

where $w_z = \partial w/\partial z$, and the superscript (0) indicates the solution of the zeroth-order problem. Quantities like M and E without superscript refer to values at $C = 0$. The choice of any of the above given parameters mean that when we focus on $R^{(1)}$, say, we keep fixed all the other parameters in the problem.

For simplicity, we now illustrate the results found in a series of relevant cases. Figure 2(*a*) provides sufficient conditions for stability (energy theory) and sufficient conditions for instability (linear theory), thus delineating the region of possible subcritical instability. Presumably when the layer is heated from above ($R < 0$) the possibility of subcritical instability is enhanced owing to the lack of symmetry in the thermal boundary conditions, as the bottom plate is considered a good heat conductor, whereas the heat flux is fixed at the upper surface, which is also kept level. In thermal convection the standard case referes to the region of positive Marangoni and Rayleigh numbers, as the sign of the Marangoni number generally follows the sign of the Rayleigh number (for exceptional cases see e.g. Guyon & Pantaloni 1980). However, with solutal convection, regions of opposite signs of E and R_s are easily accessible (Sørensen 1979). Thus, the results depicted in figure 2 also correspond to solutal convection when R and M are replaced by R_s and E respectively.

The effect of interface deformation, to first order in the crispation number, is described in figure 2(*b*), which shows the solution (21*a*). The novelty of our results with respect to those reported by Davis & Homsy (1980) is twofold. (i) we have extended their predictions to negative Rayleigh or Marangoni numbers; when M is positive there is some stabilization of the fluid layer relative to the case of a planar interface, i.e. the region of absolute stability of the motionless steady state is enlarged (for $M > 56{\cdot}77$ we have negative Rayleigh numbers). The opposite behaviour appears for negative Marangoni numbers where R is positive and greater than 669, which is the critical Rayleigh number for the onset of convective instability in the absence of interfacial tractions ($M = 0$). At $M = 0$ both linear and energy theory (figure 2*a*) provide the same prediction, and there is no subcritical instability. (ii) We have improved the numerical estimates given by Davis & Homsy (1980) in the region $0 \leqslant M \leqslant 56{\cdot}77$ and $R > 0$. A cross-check of our computational scheme has been done by performing a self-consistent computation of λ (Lebon & Pérez García 1980):

$$\lambda = R^{(0)} - M^{(0)}\frac{\int_x \theta^{(0)} w_z^{(0)}}{\langle \theta^{(0)} w^{(0)} \rangle}. \tag{22}$$

Figure 3(*a*) refers to the zeroth-order approximation in the crispation number in the case of thermosolutal capillary instability in the absence of gravity. Energy stability corrections to first-order approximation in the crispation number are given in figure 3(*b*). These results are made more complete with figure 4, which shows the role played by the Rayleigh numbers. Figure 3(*a*) should be of relevance to

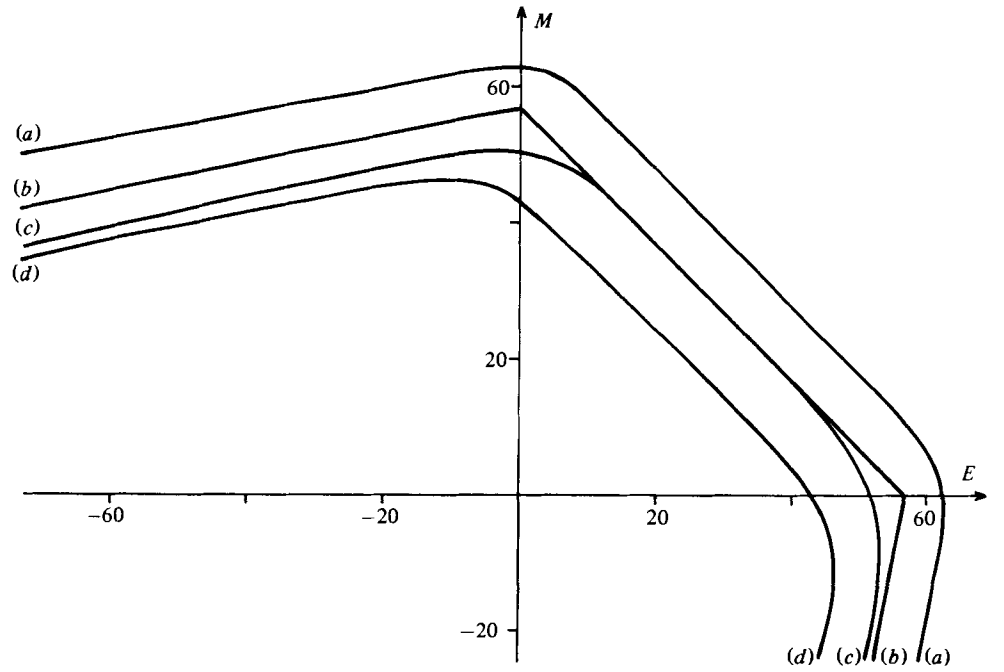

FIGURE 4. M and E energy stability lines for various values of the thermal and solutal Rayleigh numbers in the absence of interface deformation ($C = 0$): (*a*) $R = R_s = -100$; (*b*) $R = R_s = 0$; (*c*) $R = -R_s = 100$; (*d*) $R = R_s = 100$.

experiments aboard space vessels. Linear theory (Castillo & Velarde 1980) predicts regions of exchange of stabilities and overstability respectively. The latter depend on Prandtl and Lewis numbers (for illustration we have chosen $P = 10$ and $L = 10^{-2}$). The energy stability lines, however, do not depend on any of these parameters. It appears that, for large negative values of either Marangoni number, the region of expected subcritical instability is rather wide. There is symmetry with respect to the bisector of quadrant $M > 0$, $E > 0$. In this quadrant only there is a tight and perfect coupling between the two Marangoni numbers, as the critical wavenumber is the same all along the neutral stability curve $a_c = 2{\cdot}24$, in contradistinction to the case depicted in figure 2(*a*), where the critical wavenumber is not the same along the line (see also Nield 1964). In accordance with the energy theory the stability curve is a straight line, and along it $M+E = M_c(E = 0) = E_c(M = 0)$. Moreover $\Lambda+\lambda = 953{\cdot}52$, i.e. the sum of the two linking parameters remains constant in the quadrant $M > 0$, $E > 0$. On the other hand all, over the three relevant quadrants of the problem, $\lambda/\Lambda = |M/E|$.

Figure 3(*b*) provides the values of $M^{(1)}$, (21*c*), for given values of E and several values of the Lewis number. Note that a similar result appears when E is changed into M, $M^{(1)}$ into $E^{(1)}L$, and L into L^{-1}. Note also the relevance, albeit quantitatively minor, played by the Lewis number, i.e. by the role of the impurity incorporated through the ratio of the two diffusivities involved in the thermosolutal dynamics.

Figure 4 shows the influence of the two Rayleigh numbers for various illustrative values. It appears that when $R+R_s = 0$ there is no appreciable change in the stability line for the quadrant $M > 0$ and $E > 0$. When $R+R_s$ is positive the energy stability line is slightly lowered, whereas for negative values of $R+R_s$ there is a relative stabilization with respect to the case of vanishing gravity.

Lastly, as a byproduct of our energy theory we have also obtained the stability line in the absence of any interfacial traction (stress-free boundary conditions). Such a state can be achieved with the addition of surface-active agents along the open interface (Block 1956; Sørensen 1979). For this specific case our results reproduce the earlier finding reported by Shir & Joseph (1968).

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