

## Dynamics of Convective Instabilities in a Horizontal Liquid Layer

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A system of nonlinear equations describing the single-mode dynamics of both the Rayleigh-Bénard instability (RBI) and the Soret-driven instability (SDI) is derived. The system predicts saturation effects on the steady-state vertical temperature (concentration) gradient for the RBI (SDI), and transient relaxation oscillations above a given threshold. Good quantitative agreement is found with the experimental results. The possibility of observing giant pulses analogous to those observed in solid-state-laser transients is suggested.

Several experiments have been recently performed on the dynamics of the Rayleigh-Bénard instability (RBI)<sup>1-4</sup> and of the Soret-driven instability (SDI).<sup>5</sup> The aim of this Letter is to provide a theoretical description of both the RBI and SDI transients, starting from the general conservation equations and exploiting a technique similar to that used for the treatment of the laser instability.

The RBI arises in thin horizontal liquid layers heated from below when the temperature difference across the layer  $\Delta T_c$ .<sup>6</sup> The less well-known SDI may occur in two-component fluid layers, above a critical temperature difference, when the direction of the heat flux in the layer and the sign of the thermal diffusion ratio  $k_T$  are such that the molecules of the heavier component migrate upwards.<sup>7</sup>

The treatment presented here is limited to the region not too far above threshold where a single-mode convection pattern sets in. The starting point of the calculation is represented by the well-known equations expressing mass, momentum, and energy conservation in a two-component liquid mixture as they are written in Ref. 7. Those equations reduce to the standard equations for a single-component system by putting the concentration  $c$  of the heavier component identically equal to zero.

I consider first the RBI case in a single-component liquid. Following the single-mode assumption, the velocity  $v_z$  and the temperature  $T$  are expressed as

$$v_z = B(t)v(z) \cos(\vec{k} \cdot \vec{r}), \quad (1)$$

$$T = \bar{T} + A(z, t) + C(t)w(z) \cos(\vec{k} \cdot \vec{r}), \quad (2)$$

where  $\bar{T}$  is the average temperature of the layer,  $\vec{r}$  designates a horizontal vector in the configuration space, and  $\vec{k}$  is the corresponding wave number. Equations (1) and (2) differ from those in a classical linear-mode analysis because the time

dependence of velocity and temperature is not exponential, and  $A(z, t)$  which represents the average over the horizontal plane of the reduced temperature  $T - \bar{T}$  is not assumed *a priori* to be a linear function of  $z$ . In fact, when  $\Delta T$  exceeds the critical value  $\Delta T_c$ , a  $z$ -dependent convective heat flux is generated in the liquid layer. Consequently, the temperature gradient cannot stay constant over the cell height.

The dimensionless functions  $v(z)$  and  $w(z)$  are taken here as known functions.<sup>8</sup> They are indeed the solutions of the eigenvalue problem which defines the neutral stability curve.<sup>6</sup> The two functions take their maximum value at the midplane  $z = 0$ . I assume that  $v(0) = w(0) = 1$ , without loss of generality since the dependence on the temperature difference  $\Delta T$  can be included in the time-dependent amplitudes  $B$  and  $C$ . Consistently with the single-mode approach,  $A(z, t)$  can be written as

$$A(z, t) = (\Delta T/a)z - A(t)f(z), \quad (3)$$

where  $a$ , is the height of the liquid layer and the dimensionless function  $f(z)$  is determined by assigning  $v(z)$  and  $w(z)$ .

By inserting Eqs. (1)–(3) into the general conservation equations, by neglecting the nonlinear terms  $v_j \partial v_i / \partial x_j$  in the momentum conservation equation, by eliminating the pressure, and finally by averaging over the horizontal plane, the following set of dynamic equations for the three variables  $\Delta(t)$ ,  $C(t)$ , and  $B(t)$  is obtained:

$$\dot{\Delta} = (h_1/2a^2)BC - (h_2\chi/a^2)(\Delta - \Delta_0), \quad (4)$$

$$\dot{C} = -\Delta B - h_3\chi k^2 C, \quad (5)$$

$$\dot{B} = -h_4\nu k^2 [B + (h_3\chi k^2/\Delta_c)C], \quad (6)$$

where the new variable  $\Delta(t) = \Delta T/a - A(t)(df/dz)_{z=0}$  represents the temperature gradient, averaged over the horizontal plane, at  $z = 0$ . The constants  $\chi$  and  $\nu$  are, respectively, the thermal dif-

fusivity and the kinematic viscosity,  $\Delta_0 = \Delta T/a$ , and  $\Delta_c = \Delta T_c/a$ . The dimensionless constants  $h_j$ , with  $j = 1, \dots, 4$ , depend only on  $v(z)$ ,  $w(z)$ , and the product  $ka$ . By taking  $ka = 3.117$ , and by using the approximate expressions  $v(z) = 1 - 8(z/a)^2 + 16(z/a)^4$ ,  $w(z) = 1 - 4(z/a)^2$ , one finds  $h_1 = 21$ ,  $h_2 = 38.6$ ,  $h_3 = 1.8$ , and  $h_4 = 3.2$ .

The SDI case requires the introduction of a new variable which is chosen to be the mass fraction of the heavier component  $c$ . If the thermal diffusivity  $\chi$  is much larger than the mass diffusion coefficient  $D$ , as happens usually in liquid mixtures, and if fractional changes of  $c$  due to the instability are small, it is reasonable to assume, following Ref. 7, that the temperature distribution is given at any time by  $T = \bar{T} + (\Delta T/a)z$ . This assumption makes the SDI problem formally identical to the RBI problem, with the concentration  $c$  playing the role of the temperature. By repeating for the SDI the treatment outlined above for the RBI, the same set of Eqs. (4)–(6) is derived, where now  $C(t)$  is the time-dependent amplitude of the concentration mode,  $\Delta$  is the concentration gradient at the midplane of the layer, and  $\chi$  is replaced in all three equations by  $D$ . Taking into account the different boundary conditions, I put  $v(z) = 1 - 8(z/a)^2 + 16(z/a)^4$ ,  $w(z) = 1$ , and find  $h_1 = h_2 = 10$ ,  $h_3 = 1$ .<sup>7</sup> In the SDI case the minimum of the marginal-stability curve occurs for  $k_c = 0$ , and the actual value of  $k$  is dependent on the finite width of the cell. For small  $ka$ ,  $h_4 \approx 24/(ka)$ .

The steady-state solutions  $\Delta_s$ ,  $C_s$ , and  $B_s$  of Eqs. (4)–(6) are  $\Delta_s = \Delta_c$ ,  $C_s = -[2h_2/(h_1 h_3)]^{1/2}(\Delta_c/k)\epsilon^{1/2}$ , and  $B_s = (2h_2 h_3/h_1)^{1/2} k \chi \epsilon^{1/2}$ , where  $\epsilon = (\Delta_0 - \Delta_c)/\Delta_c$ . Whereas the expressions for  $B_s$  and  $C_s$  have already been given in the literature,<sup>6,7</sup> the theoretical result for  $\Delta_s$  is new. The fact that the temperature gradient (concentration gradient for the SDI) at the midplane of the cell takes, above threshold, a value independent of  $\Delta T$  and coincident with the critical temperature (concentration) gradient shows that convective instabilities present saturation effects very similar to those found in the laser, where the population inversion, above threshold, is locked to the threshold value. Direct measurements of the steady-state concentration gradient, averaged over the horizontal plane, have been performed by Giglio and Vendramini for the SKI in the mixture ethanol-toluene.<sup>9</sup> The results reported in Fig. 1 of their paper<sup>9</sup> show indeed that  $\Delta_s = \Delta_c$  above threshold.

Equations (4)–(6) fully describe the transient evolution of the instability toward the steady state

starting from an arbitrary initial condition at  $t = 0$ . In many relevant cases the description of the transient can be considerably simplified by using the so-called adiabatic approximation which is based upon the comparison of the different time scales involved in the problem and the consequent elimination of the faster variables. I discuss here for simplicity only the case  $\nu/\chi \gg 1$  (large Prandtl number) for the RBI and  $\nu/D \gg 1$  for the SDI, since these are the cases investigated in all the experiments mentioned above, except for Ref. 3. Under these assumptions, the “fast” variable in  $B$ , and the variable showing critical slowing down is  $C$ . In the region very close to threshold ( $\epsilon \ll 1$ ), also the dynamics of  $\Delta$  is fast in comparison with that of  $C$ , and therefore both  $B$  and  $\Delta$  can be adiabatically eliminated by putting  $\dot{B} = \dot{\Delta} = 0$ . The resulting dynamic equation for  $C$  shows the well-known cubic nonlinearity, and allows one to compute the time constant  $\tau$  for the decay of small deviations from the steady state. One finds  $\tau_0 = \tau_0 \epsilon^{-1}$ , where  $\tau_0 = (\chi k^2 h_3)^{-1}$ . In the RBI case  $\tau_0 = a^2/(17.71\chi)$ , in good agreement with previous theoretical<sup>6,10</sup> and experimental<sup>2</sup> results.

Slightly above threshold, when  $\tau$  cannot be considered much larger than the characteristic evolution time of the temperature (concentration) gradient which is  $(\chi h_2/a^2)^{-1}$ , it is not possible to eliminate adiabatically  $\Delta$ . The single condition  $\dot{B} = 0$  leads to the following pair of equations:

$$\dot{\Delta} = (h_1/a^2)I - (\chi h_2/a^2)(\Delta - \Delta_0), \quad (7)$$

$$\dot{I} = (2\chi k^2 h_3/\Delta_c)(\Delta - \Delta_c)I, \quad (8)$$

where  $I = (BC/2)$  represents the convective heat flux (convective mass flux for the SDI) at the midplane of the liquid layer. Note that, under the assumption  $\dot{B} = 0$ ,  $I$  is proportional to  $C^2$  and to  $B^2$ .

Some typical evolutions of  $\Delta$  and  $I$ , obtained by numerical computation for three distinct values of  $\epsilon$ , are shown in Figs. 1 and 2. The initial conditions are  $\Delta(0) = \Delta_0$ ,  $I(0) = I_i$ , where  $I_i$  is a small value simulating the noise which triggers the onset of the instability. The curves relative to  $\epsilon = 9$  are particularly striking because the gradient  $\Delta$  goes through a negative peak and the convective flux  $I$  has an overshoot 4 times larger than the steady-state value. Figure 3 reports the evolution of  $I$  when the system at the steady state above threshold is subjected to a sudden increase of  $\epsilon$ . The values of  $h_i$  appropriate to the RBI case have been used for all the computer runs. The most interesting and new feature of the tran-

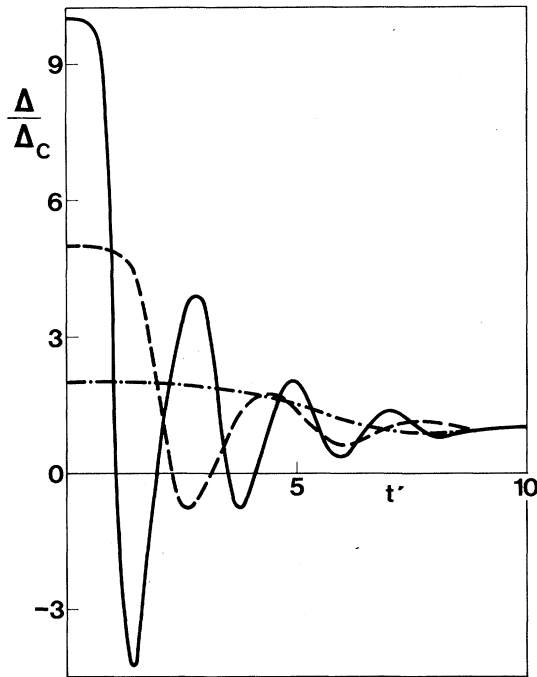


FIG. 1. Time evolution of the normalized temperature (concentration) gradient  $\Delta/\Delta_c$  starting from the initial condition  $\Delta(0) = \Delta_0$  for three distinct values of  $\epsilon = (\Delta_0 - \Delta_c)/\Delta_c$ ;  $\epsilon = 1$  (dot-dashed line),  $\epsilon = 4$  (dashed line), and  $\epsilon = 9$  (full line). The normalized time  $t'$  is defined as  $t' = (\chi h_2/a^2)t$ .

sients is the damped oscillatory behavior shown for not too small  $\epsilon$ . Linearization of Eqs. (7) and (8) around the steady-state values provides the threshold  $\epsilon_t$  for the appearance of oscillations,

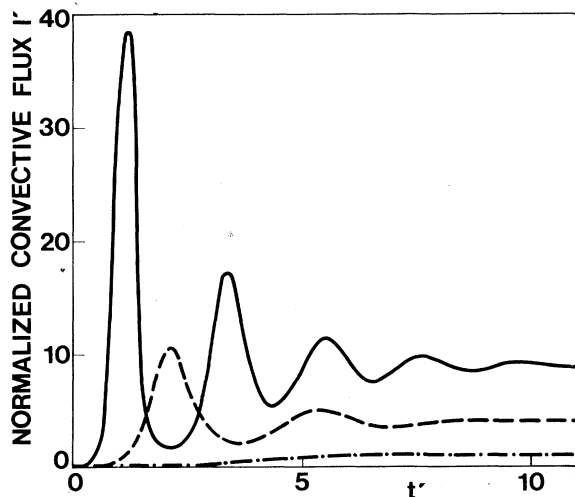


FIG. 2. Time evolution of the normalized convective flux  $I' = (h_1/2\chi\Delta_c h_2)I$ , starting from the initial condition  $I'(0) = 10^{-2}$  for the same three values of  $\epsilon$  used in Fig. 1.

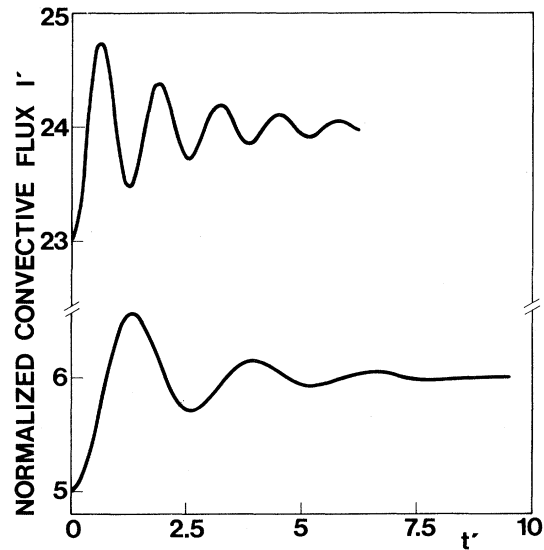


FIG. 3. Time evolution of the normalized convective flux  $I' = (h_1/2\chi\Delta_c h_2)I$  starting from the initial condition  $I'(0) = \epsilon - 1$ , with  $\epsilon = 6$  and  $24$ .

tions,  $\epsilon_t = h_2/(8a^2k^2h_3)$ , the expression of the period of the oscillations,  $\tau_{osc} = (2a^2k^2h_2h_3/\pi^2)^{-1/2}(a^2\chi)(\epsilon - \epsilon_t)^{-1/2}$ , and the damping time of the oscillations,  $\tau_D = (\chi h_2/a^2)^{-1}$ . Damped oscillatory transients have been experimentally observed both in the RBI<sup>1,4</sup> and the SDI.<sup>5</sup> All the qualitative features are correctly predicted by Eqs. (7) and (8) as one can judge by comparing Figs. 1, 2, and 3 with the figures presented in Refs. 1, 4, and 5. The empirical law  $\tau_{osc} = 200\epsilon^{-0.45 \pm 0.05}$  proposed in Ref. 1 for the range  $5 < \epsilon < 40$  is in good agreement with the expression for  $\tau_{osc}$  written above, taking into account that  $\epsilon_t = 0.27$  for the RBI case. Preliminary measurements of  $\tau_{osc}$  for the SDI in the range  $2 < \epsilon < 8$  also give the predicted power-law dependence.<sup>11</sup>

For large values of  $\epsilon$ , no adiabatic elimination can be performed, and the system may even show persistent oscillations for  $\epsilon > \epsilon_{tt}$ , where  $\epsilon_{tt} \approx \nu h_4/(\chi h_3)$  for large  $\nu/\chi$ . Persistent oscillations have indeed been observed in the RBI experiment of Ref. 1 with a threshold  $\epsilon_{tt} \approx 220$  for a liquid having  $\nu/\chi = 130$ , in good agreement with the theoretical value  $\epsilon_{tt} = 230$ . The irregular oscillations observed by Ahlers<sup>12</sup> in the stationary regime of the RBI have a threshold,  $\epsilon_{tt} \approx 2$ , which is also of the expected order of magnitude, taking into account that  $\nu/\chi \approx 1$  for that experiment.

It is known<sup>13</sup> that higher modes appear in the RBI only for  $\epsilon > 2$ , and that the amplitude of the fundamental mode is still predominant up to  $\epsilon \approx 10$ . It is, however, possible that some relevant

features of the dynamics of convective instabilities are fairly well described by a single-mode approach even for  $\epsilon > 10$ .

It should be noted that Eqs. (4)–(6) have the same structure as the equations derived in the so-called Lorenz model<sup>14</sup> of fluid instabilities. Discussions of the Lorenz model have been always concerned with the transition to turbulence, whereas the emphasis in this paper has been on the quantitative prediction of yet unexplained phenomena observed in the RBI and SDI transients in the region of regular roll convection. The analogy of the Lorenz-model equations with the Maxwell-Bloch laser equations has been considered by Haken.<sup>15</sup> A very interesting point open to investigation is the possibility of observing in convective instability experiments phenomena of the type already studied with the laser, such as the giant or superradiant pulses in the transient regime.

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## Charge-Transfer Excitation of Impurity Ions in Tokamaks

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Detailed studies of spectra from the ISX-A (Impurity Study Experiment) tokamak at the Oak Ridge National Laboratory have shown that certain oxygen-ion lines appear too anomalously intense to have been excited solely by electron collisions. These results are interpreted as being due to charge transfer and suggest the necessity of incorporating this mechanism into analyses of tokamak plasmas.

Although electron collisions usually dominate atomic processes in tokamak-produced plasmas, charge transfer from hydrogen atoms should theoretically constitute an important recombination process for certain impurity ions. The charge transfer takes place into excited states; and, in, some circumstances, excitation *via* this mechanism should dominate excitation by

electrons. Charge transfer has previously been observed through the sudden increase of radiation from the  $n = 3 \rightarrow n = 2$  transition of  $O^{7+}$  when 10–30-keV hydrogen beams are injected into a tokamak<sup>1</sup>; but detection of this process has not been reported for the more typical, noninjected discharges where the temperature of hydrogen atoms is less than 1 keV and their ambient cur-