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Different convection dynamics in mixtures with the same separation ratio

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We report on convection near threshold in circular samples of radial aspect ratio $\Gamma = 11.5$ using ethanol-water mixtures with two concentrations x, but with the same separation ratio $\Psi \simeq -0.08$. For $x = 0.250$, convection begins with transients that lead to a time-independent cell-filling state, typically in a day. For $x = 0.011$, there is a range of temperature differences where repeated transients and aperiodically fluctuating localized regions of disordered convection persist for many days. In rare cases these lead to an apparently stable "wall" state of traveling waves surrounding pure conduction.

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Experiments on thermal convection in binary mixtures [1–5] have revealed a wealth of pattern-forming phenomena and stimulated theoretical efforts to study the behavior of closely related complex Ginzburg-Landau (CGL) equations [6–8]. It has been generally assumed that the phenomena which occur are fully described by three dimensionless parameters. These are the Lewis number $\mathscr{L}$ (the ratio of the mass diffusivity to the thermal diffusivity), the Prandtl number σ (the ratio of the kinematic viscosity to the thermal diffusivity), and the separation ratio Ψ [for positive (negative) Ψ concentration gradients induced by the imposed temperature gradient destabilize (stabilize) the conduction state]. Variations of $\mathscr{L}$ and σ over typical experimental ranges have little influence on the system. Thus the pattern-formation phenomena are expected to be determined essentially by Ψ. Mixtures with sufficiently negative Ψ are predicted to undergo a Hopf bifurcation to traveling-wave (TW) convection when the temperature difference ΔT exceeds the critical value ΔT_c (i.e., when $\epsilon \equiv \Delta T/\Delta T_c - 1 > 0$). We find that two mixtures in cells with circular cross sections and with the same $\Psi \simeq -0.08$, but with different alcohol weight fractions $x = 0.250$ and $x = 0.011$, behave very differently for the same small ϵ. In the former, transients lasting a day or so lead to a cell-filling time-independent state; the latter reveals non-periodically-repeated transients involving pure conduction and disordered localized regions of TWs lasting for many days. On rare occasions the repeated transients can lead to a novel, apparently stable, state of radially-localized waves traveling parallel to the wall with the cell interior remaining in the conduction state. Our results raise the question of whether phenomena observed in one-dimensional systems, such as stable localized pulses [1], periodic and aperiodic "blinking" states [2,4], and "dispersive chaos" [5] remain unaltered when x is changed at constant Ψ. Most (but not all [9]) of these phenomena have been described, at least qualitatively, by CGL equations [6–8,10,11] with coefficients which are expected to depend only on Ψ, σ, and $\mathscr{L}$. The difference in behavior at the same Ψ but different x which we report may require a reexamination of this theoretical approach.

The convection cell consisted of a sapphire top-plate and a silver bottom-plate separated by circular Delrin sidewalls. The temperature of the upper surface of the sapphire was controlled near 19.83 °C to ± 1mK by circulating water, and the bottom-plate temperature was controlled to ± 0.1mK. All experiments were carried out at fixed ΔT. The cell height d was uniform to $\pm 0.05\%$, and the cell diameter was 7.90 cm. For $x = 0.250$ (0.011) we had $d = 0.343$ (0.345) cm [$\Gamma \equiv$radius/height$= 11.52$ (11.45)]. For $x = 0.250$ (0.011) the mixtures had $\Psi = -0.082$ (-0.078), $\sigma = 23.7$ (7.2), and $\mathscr{L} = 0.007$ (0.008). The vertical diffusion time $\tau_v \equiv d^2/\kappa$ was 114.6 (85.5) s (κ is the thermal diffusivity). The Ψ values were determined [12–14] from the measured linear TW frequencies $\omega = 5.87$ (5.56) for $x = 0.250$ (0.011) (here $\omega = \tilde{\omega}\tau_v$ with $\tilde{\omega}$ in radians/s). They agree well with the values -0.080 (-0.077) derived from x and Ref. [15]. The convective threshold was crossed quasistatically making steps in ϵ of 0.0013 every 3 h. The onset ΔT_c was defined to be midway between the last point in the conduction regime and the first point for which convection occurred. For $x = 0.250$ (0.011), the critical temperature difference was $\Delta T_c = 2.244$ (3.183) °C. For $\Delta T = \Delta T_c$, we estimate [15] $\Delta\Psi/\Psi = 0.00$ and 0.25 for $x = 0.250$ and 0.011 respectively (here $\Delta\Psi$ is the difference in Ψ at the top and the bottom of the sample). The effect of departures from the Boussinesq approximation due to variations in Ψ has been considered only for linear properties near $\Psi = 0$ [16]. However, on the basis of a comparison with typical non-Boussinesq effects in pure fluids [17] one would not expect large departures from the Boussinesq approximation for these values of $\Delta\Psi/\Psi$. The patterns were visualized using the shadowgraph technique.

For both mixtures, convection immediately above onset initially consisted of a superposition of radially inward- and outward-traveling waves [Figs. 1(a) and 2(h)] [3,18,19]. The only observable difference was occasional azimuthal modulation of the roll pair nearest the wall [Fig. 2(b)] for $x = 0.011$. For both x, these waves eventually focused azimuthally [Figs. 1(b), 1(c), and 2(a), 2(e)] and then collapsed radially [Figs. 1(e) and 2(d)] to form a localized region of convection surrounded by quiescent fluid as reported before [3] for $x \simeq 0.25$. The localized region was often a long-lived localized pulse, similar in appearance to those studied in one-dimensional cells [1], but sometimes it was a region of disorganized TW convection.

For $x = 0.250$ and for $\epsilon < 0.001$, the localized region always decayed in amplitude, leaving a "hole" [Fig. 1(f)] of pure conduction. Convection then grew around the "hole"

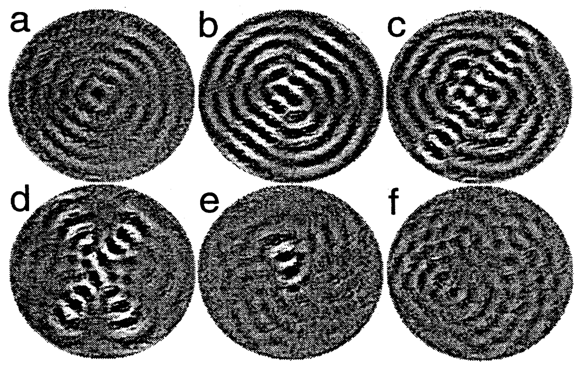

FIG. 1. Image sequence for $x=0.250$ and $\epsilon=7\times10^{-4}$, at times (a) $69\tau_v$ (b) $125\tau_v$ (c) $130\tau_v$ (d) $152\tau_v$ (e) $184\tau_v$ and (f) $236\tau_v$ after crossing the convective threshold.

and spread into it, leading to a steady state in which the entire container was filled with stationary convection rolls about a day later like the image shown in Fig. 3(a).

Whereas for $x=0.250$ convection always grew to a stable time-independent cell-filling state within a day or so, the long-time behavior in the $x=0.011$ mixture at similar values of ϵ was different. Convection also began with radially traveling linear transients and then focused to form a localized region of convection. However, this localized state died away, returning the cell to the conduction state. After some time convection began again in a qualitatively similar manner as before. In most experimental runs this behavior persisted apparently indefinitely (a week). Typical patterns seen at late times during this process are shown in Fig. 2. However, in one case an apparently stable state consisting of a narrow ring of TWs next to the cell wall and surrounding pure conduction in the interior was reached [18–20]. This "wall" state is shown in Fig. 3(b). It consisted of azimuthally-traveling waves along the cell periphery, with the roll axes perpendicular to the cell wall. Radially this state was approximately one roll wavelength wide. The state continued to exist over a range of ϵ, and was monitored for a time period of about three days. The TW frequency was $\omega=1.52$ at $\epsilon=0.0025$, and fell to 0.96 at $\epsilon=0.0179$. We observed the "wall" state only in a single experimental run, but there were other runs during which an *unstable* "wall" state covering part or most of the circumference of the cell had formed temporarily. In all of the runs but the one leading to the seemingly stable "wall" state, the erratically repeated transients persisted for days.

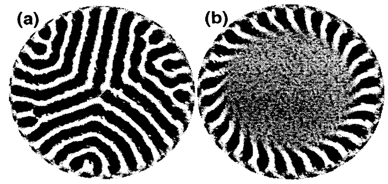

FIG. 3. Two apparently stable states for $\epsilon\simeq0.001$. (a) Cell-filling state for $x=0.011$. A state like this always formed for $x=0.250$ and $\epsilon>0$ and formed for $x=0.011$ for sufficiently large ϵ. It is representative of the uppermost branches in Figs. 5(a) and 5(b). (b) A "wall" state consisting of clockwise-traveling rolls which formed rarely for $x=0.011$.

Figure 4 shows the time dependence of the Nusselt number $\mathcal{N}(\epsilon,t)$ (the ratio of the total heat transport through the fluid to the heat transport due to conduction) for the two mixtures during three runs at similar values of ϵ. For

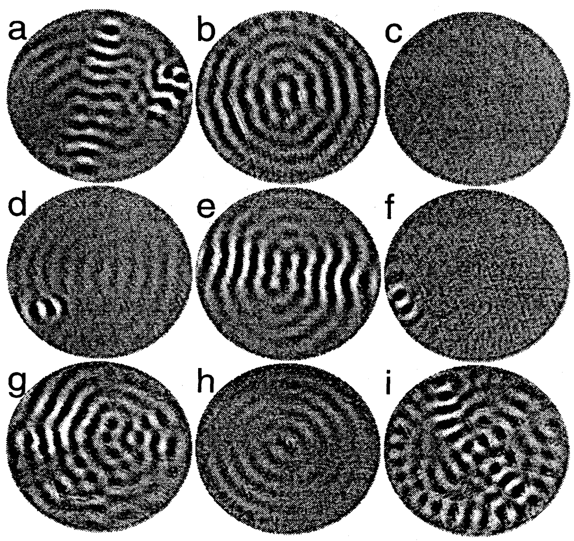

FIG. 2. Images during repeated transients at $\epsilon=6\times10^{-4}$ for $x=0.011$ from the same run as the Nusselt numbers in Fig. 4 (lower solid line) at times (a) $3620\tau_v$, (b) $3863\tau_v$, (c) $4062\tau_v$, (d) $4285\tau_v$, (e) $4358\tau_v$, (f) $4731\tau_v$, (g) $4954\tau_v$, (h) $5177\tau_v$, (i) $5400\tau_v$.

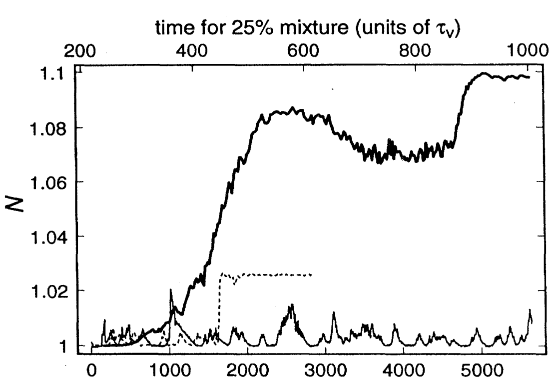

FIG. 4. Time series of the Nusselt number for two different mixtures. For $x=0.250$ (upper curve) at $\epsilon=7\times10^{-4}$, convection grew to a cell-filling state [Fig. 3(a)] in about $900\tau_v$ (upper time axis). The Nusselt-number record started after the image in Fig. 1(f) was taken. For $x=0.011$ at $\epsilon=6\times10^{-4}$, a seemingly stable "wall" state [Fig. 3(b)] formed during one run (dashed line, lower time axis). During all other runs at the same ϵ, irregularly repeated transients (lower solid curve) were seen over a period of $5600\tau_v$ (lower time axis). Images in Fig. 2 illustrate the patterns observed in this run.

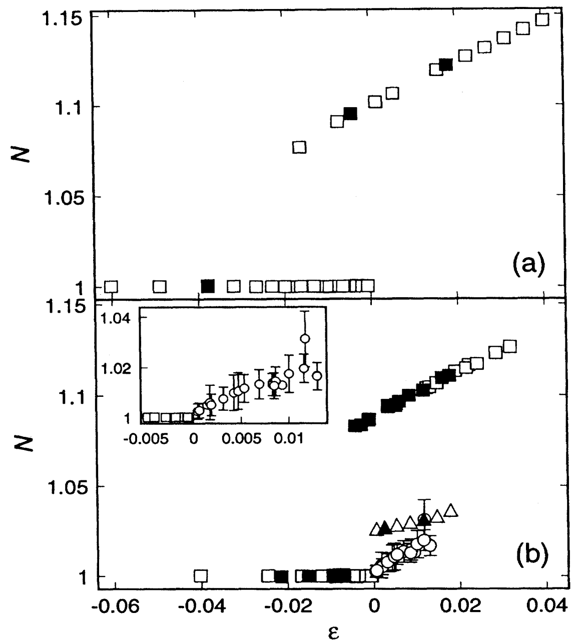

FIG. 5. Nusselt numbers vs ϵ. Open (solid) symbols correspond to increasing (decreasing) ϵ. (a) $x=0.250$ and $\Gamma=11.53$. (b) $x=0.011$, showing conduction and stationary convection branches (squares), an apparently continuous branch corresponding to erratically fluctuating convection (circles), and the "wall" state (triangles). The inset shows details of the repeating-transients branch.

$x=0.250$ at $\epsilon=7\times10^{-4}$ (upper curve), the data for $\mathcal{N}(t)$ began immediately after the image in Fig. 1(f) was taken. In that run, at $t\simeq900\tau_v$, the system reached a steady state in which the cell was filled with stationary convection rolls like those shown in Fig. 3(a). The dashed line in Fig. 4 shows $\mathcal{N}(t)$ for $x=0.011$ at $\epsilon=6\times10^{-4}$. This run yielded the "wall" state [Fig. 3(b)] at $t\simeq1600$. During another run at $x=0.011$ and shown in the figure (solid lower curve), repeated transients persisted over a period of about $5\frac{1}{2}$ days ($\approx5600\tau_v$) at the same value of ϵ, until the run was terminated.

Figure 5 compares $\mathcal{N}(\epsilon)$ for the two mixtures. As shown in Fig. 5(a), we observed only two branches for $x=0.250$, corresponding to the stationary rolls filling the cell and to pure conduction. Open (solid) symbols show data obtained while increasing (decreasing) ϵ. For $0<\epsilon<10^{-3}$ the upper branch was reached by decay of a pulse, leading to a "hole" and subsequent growth of convection as described above. For runs in which ϵ was increased beyond 10^{-3} during the initial linear transient, an azimuthally focused line of convection yielded either a long-lived (≥40 h $\simeq1300\tau_v$) pulse which eventually spread, or a localized but somewhat larger region of disordered convection which also spread to fill the cell.

Figure 5(b) shows $\mathcal{N}(\epsilon)$ for $x=0.011$. Again, open (solid) symbols show data for increasing (decreasing) ϵ. As in Fig. 5(a), data for the conducting and the cell-filling branches are shown by squares. The triangles correspond to the "wall" state but do not indicate its full stability range, which we have not determined. The open circles correspond to repeated transients for which $\mathcal{N}$ is erratic, with the symbols giving the average value and the vertical bars the standard deviation of $\mathcal{N}$.

Steinberg *et al.* reported [21] spatiotemporal chaos in a wide rectangular cell with an aspect ratio of 1:9:20. Their mixture had $x=0.25$ [22] and $\Psi=-0.1$. The chaotic state was observed under conditions of constant applied heat flux q, with $\epsilon=0.004$ to 0.014. For these parameters, the chaotic state persisted in time, and it was characterized by alternating localization of a line of convection near either of the long walls. Occasionally, the line of convection split into one or two pulses near the long wall. We observed a similar aperiodically fluctuating state for $x=0.250$ in a cylindrical container, but only at constant q [as described above, for $x=0.250$ and constant ϵ, a cell-filling state like the one shown in Fig. 3(a) always was reached after about one day]. At constant q the chaotic state persisted for days, and it was characterized by a repeated azimuthal focusing of convection rolls along alternating diameters of the container. The line of enhanced convection broke up into weak pulses, which died, while another line of convection, roughly perpendicular to the first, was growing stronger. Such persistent, aperiodically fluctuating states were observed for a value of q just above threshold, and also when q was reduced to a slightly subcritical value. While the chaotic dynamics were somewhat reminiscent of the repeated-transient regime seen at constant ϵ and for $x=0.011$, there are significant differences. For $x=0.250$, persistent aperiodic states were never observed at fixed ϵ. Nusselt-number fluctuations were about ten times smaller in the constant q experiments than in the low-concentration mixture at fixed ϵ. Finally, large-amplitude localized regions of disordered convection never died out at a fixed value of q, as they were observed to do in the $x=0.011$ mixture at constant ϵ.

We believe that the oscillations observed by Steinberg *et al.* and by ourselves at $x=0.25$ and fixed q are related to those predicted by Busse and are fundamentally different from the chaotic states seen for $x=0.011$ at fixed ϵ. Busse suggested [23] that a system near a transcritical or subcritical bifurcation can show periodic oscillations when q is kept fixed. Under these conditions, the system, in a sense, repeatedly falls off and is pushed back onto the convection branch. However, unlike the theoretical prediction, the oscillations we observe are aperiodic.

The erratic states we observe at constant ϵ and $x=0.011$ bear close resemblance to the "dispersive chaos" seen in an annular cell with $x=0.004$, [5] which was attributed to strong nonlinear dispersion. However, our failure to observe persistent chaotic dynamics in the $x=0.250$ mixture with a very similar separation ratio leads us to believe that some other effect is responsible for the behavior we observe. The most obvious and striking difference between the two mixtures is the separation ratio's dependence on the concentration. The derivative $(\partial\Psi/\partial x)_T$ for $x=0.250$ is positive and equal to 1.8, whereas for $x=0.011$ it is negative and equal to -6.5. Even though the total variation of Ψ across our cells was modest, the difference in sign of $(\partial\Psi/\partial x)_T$ seems to have a decisive effect on the behavior of the system. As far as we know, this "non-Boussinesq" effect has not been considered before. It may become important because, at least for one-dimensional systems, traveling waves induce a large-scale concentration current [24,25]. Although this current does not change the vertical average of the concentration, it affects the local stability properties by changing the vertical concentration *gradient* ahead of and behind a localized state. This mechanism contributes to the stability of one-dimensional pulses, because the vertical concentration gradient built up ahead of the pulse stabilizes the quiescent fluid and prevents the invasion of this region by convection rolls. We speculate that the difference in the separation ratio's dependence on concentration, coupled with concentration transport by localized traveling waves of convection, may lead to the differences in the dynamics of the two mixtures.

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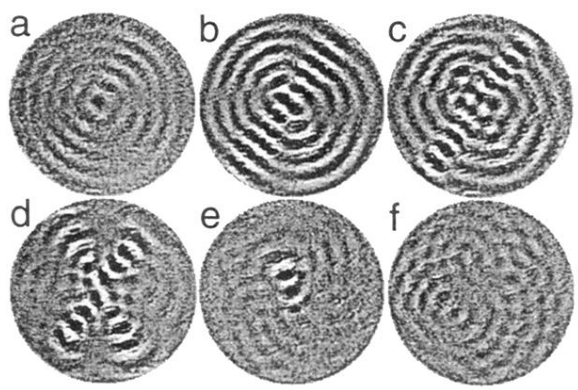

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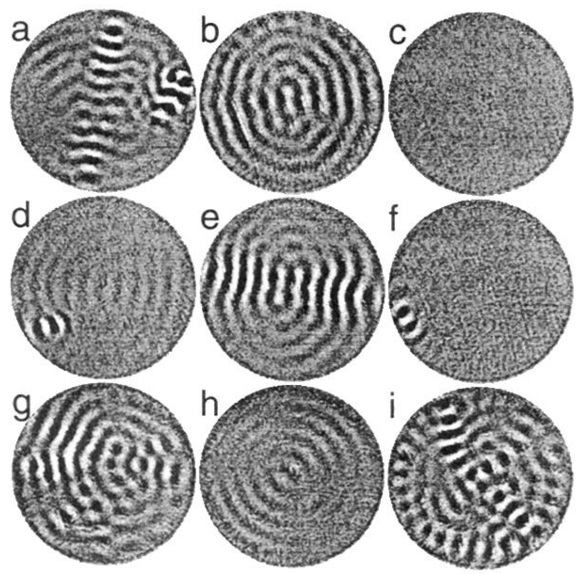

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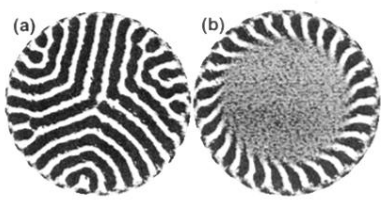

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