Transient Localized States in 2d Binary Liquid Convection

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We report an experimental study of the onset of convection in ethanol-water mixtures confined in a circular cell of radial aspect ratio 11.6. The initial bifurcation was to radial traveling waves; however, the linear state typically gave way to a nonlinear one in which convection alternately focused along one cell diameter and then another roughly perpendicular to the first. After a number of cycles, this state frequently collapsed to a localized pulse of traveling-wave convection very similar to those observed in 1d geometries. The pulses we observed either decayed back to pure conduction or grew to fill or nearly fill the cell.

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Convection in binary-liquid mixtures has proved to be a rich model system for the study of pattern formation and evolution,[1–10] particularly for the case of a negative separation ratio, ψ. For sufficiently negative ψ the initial (backward) bifurcation is to a state of traveling-wave (TW) convection.[1] Extensive study of this system in one-dimensional (1d) geometries has revealed a remarkable wealth of phenomena, including highly localized pulses of TW convection,[2–5] localized convecting regions of arbitrary, history-dependent, spatial extent,[6] "blinking" states in which convection appears alternately on one side and then another of a rectangular cell,[7] and states of very complicated spatio-temporal chaos emerging directly from the conduction state.[8] It is now known[11–13] that the complex quintic Ginzburg-Landau equation (CGL), which describes the onset of traveling wave convection in binary mixtures, has spatially localized solutions in 1 dimension. In the limit of large imaginary coefficients the CGL equation reduces to the non-linear Schrödinger equation (NLS), and there seems to exist a mathematical connection between the solitonic solutions of the NLS equation and the pulses of the CGL equation.[12,14] In two dimensions (2d) it is known that the NLS equation possesses no solutions which are localized in 2d, thus raising the question of whether localized solutions of the 2d CGL equation and the corresponding 2d pulses in binary fluid convection exist and are stable. To date, however, very little experimental work[9] has been carried out on binary liquid convection in 2d, perhaps because of the obvious complexity of the states. Here we report experimental results which show that 2d pulses form spontaneously in this system, but so far we have always found them to be unstable.

We report here the results of an extensive survey of the initial bifurcation to convection for ethanol-water mixtures confined in a circular convection cell of radius to height ratio 11.6. We made these observations for mixtures having separation ratios of $-0.09, -0.10, -0.14$, and -0.16, both under conditions of constant heat current and constant temperature difference, and usually observed similar sequences of transient phenomena for either condition. The initial state we observed was a superposition of radially inward and outward traveling circular convection rolls. This linear state always evolved to a nonlinear one in which radial TW convection focused strongly along a cell diameter, leaving the remainder of the cell with greatly reduced amplitude. This azimuthally focused state typically gave way to another similar state with convection focused along another diameter roughly perpendicular to the first. This process often continued for some time, with azimuthal focusing occurring alternately along one diameter and then another, but eventually *radial* focusing along a diameter of high amplitude resulted in the formation of a localized state of TW convection. These states frequently appeared to be very similar to the pulses observed in 1d rectangular and annular geometries, both in spatial extent and TW frequency. They had a long lifetime, but unlike the 1d case, they were not stable. They either died out altogether, which resulted in a repetition of the entire transient sequence, or grew to a disordered, although still localized, state which gradually expanded so as to fill or nearly fill the entire cell.

The convection cell consisted of a single crystal sapphire top plate and a polished silver bottom plate separated by an annular Delrin spacer sealed to both plates by O-rings. The central circular region containing the convecting fluid had a radius of 3.95 cm and a height of 0.34 cm, uniform to $\pm 0.15\%$. The upper surface of the sapphire was held at fixed temperature to $\pm 1mK$ by means of circulating temperature-controlled water, while the lower surface of the bottom plate was in close contact with a resistive heater of nearly the same size as the bottom plate. We were able to control either the heat flux through the cell, $\dot{Q}$, or the temperature difference across it, ΔT, to better than 0.1%. We visualized the convection pattern by means of the shadowgraph method. Video images were digitized and divided pixel by pixel by a background image taken in the pure conduction state. In addition to revealing the pattern of convection present, sequences of such images taken at a frequency about 10 times the linear TW frequency were used to measure the TW frequency. Two different mixtures (25.0 wt% and 20.8 wt% ethanol) were studied at several mean temperatures in order to obtain different separation ratios.[15] For these solutions the vertical thermal diffusion time t_v ranged from 102 to 110 seconds. The initial bifurcation from conduction was studied by increasing either $\dot{Q}$ or ΔT in steps of about 0.1% of the critical value, at intervals of about $200 t_v$. We take as ΔT_c and $\dot{Q}_c$ the values midway between those for which convection was first observed and the preceding values. Our resolution in $\epsilon \equiv \Delta T/\Delta T_c - 1$ thus is about 0.05%.

The initial linear state observed upon crossing threshold consisted of nearly azimuthally symmetric waves as shown in Fig. 1a. They traveled in the radial direction, both inward and outward, with a dimensionless angular frequency ($\Omega \equiv \omega t_v$) which we measured (calculated[16]) to be 7.45(7.89) for $\psi = -0.14$ and 8.28(8.56) for $\psi = -0.16$. The amplitude of this state grew exponentially in time. The weak linear state always gave way to a nonlinear state in which convection focused along a cell diameter, as shown in Fig. 1b. Typically this state was followed by a similar one in which convection focused along a diameter roughly perpendicular to the first (Figs.1c-1e). This sequence was usually repeated many times. We have not determined the average period of focusing, but it appeared to be aperiodic. During each focusing process the TW frequency gradually decreased to about half the linear value. Decomposition of time series of the convective amplitude for the linear state along a cell diameter into "left" and "right" TW's showed that the amplitude of waves traveling inward toward the cell center was less than that of the outward traveling waves. This is very similar to what has been observed in narrow rectangular cells, where left and right TW's are reflected weakly at the ends of the cell and grow in amplitude as they travel through the cell.[10] The nonlinear focused state eventually contracted radially to a localized state of TW convection. These localized states were often pulses like that shown in Fig. 1f (localized states resembling those shown in Figs. 2 and 3 were also observed on occasion). For comparison, a stable pulse observed[2] in a one-dimensional rectangular cell of length 25.3d, width 4.87d and depth d = 0.308 cm for $\psi = -0.09$ is shown as Fig. 1g, while Fig. 1h shows the pulse from Fig. 1f expanded and reoriented. The similarity is quite striking. It indicates that the pulse observed in the rectangular geometry of width 4.87d already had essentially the shape of the pulses observed in 2d and was stabilized by the proximity of the sidewalls which may have suppressed transverse perturbations.

Like the pulses observed in 1d geometries, the TW frequency in the pulses we observed was about half the linear frequency, but unlike the 1d case these pulses appeared to be intrinsically unstable in 2 dimensions even though they formed spontaneously. They usually either died slowly in amplitude, leading back to the conduction state, or grew, over a period of $70t_v - 200t_v$, to a roughly circular state of disorganized TW convection having a diameter about equal to the original pulse length. This state then expanded over a period of about $300t_v$, to fill or nearly fill the cell. For $\psi \leq -0.10$ the final state consisted of stationary convection rolls which filled the cell completely, while for $\psi = -0.09$ the final state consisted of traveling-wave convection which nearly filled the cell.

The process whereby a pulse of TW convection grew to a localized but disordered state was always the same and is shown in Fig. 2. The pulse initially spread in the transverse direction, i.e. parallel to the rolls (Figs. 2a & 2b). The trailing edge, where the waves entered the pulse, then became unstable with respect to a transverse perturbation (Fig. 2b & 2c), which lead to a defect which moved in the TW direction (Fig. 2d). The defect was then expelled, and the pulse appeared reoriented but essentially unchanged (Fig. 2e). This process often occurred several times, causing the pulse to effectively rotate, but eventually the defect was always retained, and the pulse assumed a more nearly circular shape, as shown in Fig. 2f. Defects never appeared in a pulse while it was decaying to the conduction state, and such pulses appeared to be stationary, moving less than $d/2$ in $140t_v$.

Since the pulses we observed were not stable at onset, we attempted to stabilize them by suddenly reducing the heat flux or temperature difference as they were forming. We found it was possible to produce long-lived pulses in this fashion, but they were not ultimately stable. Under conditions of constant temperature difference for $\psi = -0.09$ the longest pulse lifetime observed was about $200t_v$ at $\epsilon = 0.0006$. For $\psi = -0.10$ the maximum lifetime was about $600t_v$ (18 h) at $\epsilon = -0.0031$. For both values of ψ, pulses ultimately expanded spatially (see Fig. 2) or decreased in amplitude and vanished depending on whether ϵ was greater or less than that resulting in the maximum lifetime, respectively. This suggests that such pulses are stable at only one unique value of ϵ, for any given ψ, if at all.

In the course of attempting to stabilize pulses as described above, we often generated a different localized state, an example of which is shown in Fig. 3. The waves traveled along the perimeter of this "triangular" state, in a manner similar to that described for the propagation of defects through the pulse. Unlike the pulse, which would reorient itself at certain times, this localized state was characterized by a continuous and rapid rotation. We have observed it for $\psi = -0.14$ and -0.16 at negative ϵ. Under conditions of constant heat flux ($0.977\dot{Q}_c$), the state shown in Fig. 3 lasted about $1700t_v$ (2 days), before growing into a more disordered state. A very similar state was obtained with $\psi = -0.14$ at $\epsilon = -0.0291$ by following an entirely different procedure. In this case we allowed the cell to fill with stationary rolls at threshold while controlling ϵ. We then reduced ϵ in steps. Upon reaching $\epsilon = -0.0279$ a transition to traveling waves was observed, and the cell began to empty of convection. We continued to reduce ϵ in small steps (0.0031) waiting about $800t_v$ (1 day) between steps. Following each step the portion of the cell supporting convection decreased, but only very slowly; the heat flux did not reach steady state during the time we waited. When the convecting region had shrunk in size to about that shown in Fig. 2f, we began controlling heat flux rather than temperature difference. The convection pattern then evolved to closely resemble the "triangular" state shown in Fig. 3. This localized state traveled slowly around the periphery of the cell, and lasted for about $2700t_v$, at which time the run was interrupted by an equipment failure. This behavior suggests that this state may be stable, although possibly only under conditions of constant heat flux. Such conditions automatically decrease the temperature difference if the state begins to expand spatially or increase in amplitude and increase it if the state shrinks or decreases in amplitude, which may stabilize the pulse.

Although the transient sequence described above was usually what we observed, at least one other possibility exists. Figure 4 shows the continuation of the run at $\psi = -0.09$, $\epsilon = 0.000$ presented in Fig. 1. Figure 4a was taken $370t_v$ (11 h) after Fig. 1f, and shows convection just beginning for the second time following the decay of the pulse shown in Fig. 1f. After the focused state shown in Fig. 4b, convection collapsed to another pulse, Fig. 4c, which again died away. This time, however, as the pulse was decaying, a pair of spiral states appeared. They grew in amplitude, Fig. 4d, and evolved directly into an extended disorganized state of convection, Fig. 4e, which continued to expand, Fig. 4f, to fill most of the cell. Although striking in appearance, such spiral states were relatively rare. The decay of a pulse at slightly positive ϵ was usually followed by a repetition of the transient sequence and formation of a localized pulse which grew into a disorganized localized state of convection and then expanded to fill or nearly fill the cell.

In summary, we have observed in cylindrical geometry some of the convection states studied in narrow 1d cells, such as linear counter propagating waves and localized pulses. We also found novel phenomena, such as azimuthally focused transients and localized states like the one shown in Fig. 3. Although the localized pulses we have studied in 2d were not stable, they had very long ϵ-dependent lifetimes. We have not been able to create pulses of arbitrary length such as those which have been observed in 1d systems.[6] Previous experiments in wide rectangular cells[9] produced transient states that look similar to those displayed in Figs. 2a & 2f. In addition, the initial transient involved focusing in the direction transverse to the of the wave motion, very reminiscent of the azimuthal focusing we have described. At present, numerical studies of the CGL equation seem only qualitatively related to the experiment. While the simulations done by Bestehorn and Haken[17] do capture some of the phenomena we have observed, e.g. growth and decay of pulses and the existence of spirals, it is not clear to us how to relate the coefficients in the equation being simulated to the parameters of the physical system.

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FIGURE CAPTIONS

Fig. 1. Shadowgraph images showing the initial transient leading to convection in a water/ethanol mixture having a separation ratio $\psi = -0.09$. Images (a)-(f) were obtained using a circular cell of radial aspect ratio 11.6, with δT at which convection first began. Images (b)-(f) were taken at intervals of $100t_v$ (3 h), while image (a) is from a different run and shown with an enhanced contrast. In (g) and (h), a stable pulse (g) observed in a rectangular cell of width 4.87, and length 25.3 times its depth at $\psi = -0.09$ (see reference 2) is compared to the 2d pulse (h) from image (f), expanded and reoriented. The arrows in images (g) and (h) indicate the direction of roll propagation.

Fig. 2. Shadowgraph images showing the typical evolution of a localized pulse of convection which is expanding at $\epsilon = 0.0010$ in a mixture with $\psi = -0.10$. Image (a) shows the pulse shortly after its formation. Image (b), taken about $30t_v$ later, shows that the pulse has spread somewhat and is becoming unstable on its trailing edge, where the waves enter. Images (c)&(d) show the resulting defect traveling along the pulse, leaving it essentially unchanged, but reoriented (image (e)). Images (b) through (e) were taken at intervals of about $3t_v$. Image (f), which is from another run with $\psi = -0.10$ and $\epsilon = -0.0031$, shows a typical disordered but still localized state which results (usually after many cycles of reorientation) when the pulse retains several of these defects.

Fig. 3. Shadowgraph image of a localized state obtained at $\psi = -0.14$ by reducing the heat current to 0.977 of its critical value during the initial transient. After about $1700t_v$ (over 2 days) this state began to expand. Arrows indicate the direction of traveling wave motion.

Fig. 4. Shadowgraph images showing the subsequent (rather atypical) evolution observed in the run shown in Fig. 1 (b)-(f). Image (a) was taken $370t_v$ after image (f) of Fig. 1, and is shown here with enhanced contrast. Subsequent images were taken at intervals of $70t_v$.