Traveling Waves and Chaos in Convection in Binary Fluid Mixtures

R. W. Walden, Paul Kolodner, A. Passner, and C. M. Surko
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 2 May 1985)

Rayleigh-Bénard convection is studied in alcohol-water mixtures in which the diffusion of concentration opposes convection via the Soret effect. Near onset, the convective rolls are found to move continuously as traveling waves, in contrast to the stationary roll patterns observed in homogeneous fluids. Dependent upon the temperature difference across the fluid layer (i.e., Rayleigh number), these traveling-wave states are either periodic or chaotic. At larger Rayleigh numbers, time-independent flow is observed which is the same as that expected for the homogeneous fluid mixture.

PACS numbers: 47.25.–c

There is considerable interest in understanding the nature of convection in a horizontal fluid layer heated from below (i.e., Rayleigh-Bénard convection).[1–4] Convection provides a particularly useful model of nonequilibrium dissipative systems, since the fluid equations which describe the system are well known, and, from an experimental viewpoint, the boundary conditions on the system can be well controlled. Convection in a homogeneous fluid is described by two parameters: the Rayleigh number R, which is proportional to the temperature difference across the fluid layer, and the Prandtl number P, which is the ratio of the rate of dissipation of momentum to the rate of diffusion of heat. For such a fluid, the first flow pattern above the onset of convection is known to be a set of nearly parallel rolls which are stationary in space.

The description of convection in a binary fluid mixture requires two additional parameters, the Lewis number $L=D/\kappa$ (where D is the diffusion coefficient and κ is the thermal diffusivity of the mixture) and the separation parameter ψ.[5,6] The quantity ψ is the ratio of the coupling of concentration fluctuations with gravity to the coupling (via thermal expansion) of temperature fluctuations with gravity. Specifically, $\psi=c(1-c)(\alpha'/\alpha)S_T$, where S_T is the Soret coefficient which relates the concentration flux to an imposed temperature gradient, c is the weight concentration of one component, $\alpha'=\rho^{-1}(\partial\rho/\partial c)_{P,T}$, $\alpha=\rho^{-1}(\partial\rho/\partial T)_{P,c}$, ρ is the density, and T is the temperature.[5,6] If ψ is negative, the thermally induced concentration gradients oppose the onset of convection. For the experiments described here, $L \ll 1 < P$. In this case, when $\psi \lesssim -L^2$, the first convecting state is known to be time dependent.[5,7,8] To our knowledge, the flow pattern associated with this time-dependent state has not previously been studied experimentally. Recently, it has also been suggested that, for ψ sufficiently negative, the first convecting state might exhibit both spatially and temporally incoherent behavior.[9]

We have studied convection in mixtures of ethyl alcohol and water for negative values of ψ. We find that the flow is chaotic at the lowest values of R for which convection persists. At larger R, periodic states are observed. Associated with all of these time-dependent states are *qualitative* changes in the flow pattern, perhaps best described as a continuous motion of the rolls in the lateral direction. As Rayleigh number is increased further, the time dependence ceases, and the convection pattern and heat transport are those which would be expected for convection in the homogeneous fluid.

The experimental apparatus is similar to that described previously[4] and consists of a container of height $d=0.52$ cm with horizontal dimensions 2.3×4.9 cm^2. The bottom plate of the container is copper, the top plate is sapphire, and the cell walls are glass. The temperature field associated with the convective flow pattern is visualized from above by use of a shadowgraph technique.[4] The Nusselt number N (i.e., the ratio of the heat transport through the layer to that due to thermal conduction alone) is also measured.

We define the reduced Rayleigh number $r\equiv R/R_c$, where R_c is the Rayleigh number for the onset of convection calculated with use of viscosity, thermal diffusivity, and thermal expansion coefficient of the *homogeneous* fluid mixture.[10] Shown in Figs. 1(a) and 1(b) are measurements of N as a function of r for an 8%-by-weight mixture of ethyl alcohol in water at 10 and 20 °C. Since the heat current is held constant, r decreases and N increases when convection begins (dashed lines with ascending arrows). As shown by the solid curves in Fig. 1, the dependence of N on r is quite similar to that expected[11] for time-independent convection in a fluid with the properties of the homogeneous mixture.[10] A large hysteresis is observed between the onset and the cessation of the flow, as is expected from previous experiments[5,7] on similar mixture. For these mixtures $L\sim10^{-2}$. In this case convection begins at the onset of a time-dependent instability; the reduced Rayleigh number r_c at the onset of convection is

$$r_c=1-\psi(1+\psi+1/P)^{-1}, \tag{1}$$

where we have neglected terms of order $L \ll 1$.[5–7]

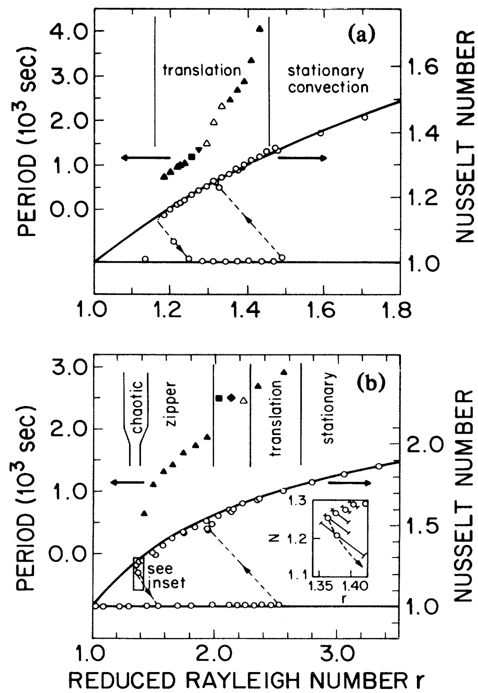

FIG. 1. The Nusselt number (i.e., the heat transport through the fluid relative to that due to conduction) as a function of $r = R/R_c$ for values of the separation parameter (a) $\psi = -0.35$ and (b) $\psi = -0.6$. The solid curves are those expected for steady convection in a simple fluid with the properties of the fluid mixture. The dashed lines with arrows indicate transitions between conducting and convecting states. Also shown is the period τ of the oscillations: (solid triangles) periodic states, (open triangles) chaotic states with a dominant spectral component at the period indicated, and (squares, lozenges, and inverted triangles) periodic states which exhibit subharmonics at 2, 3, and 5 times the period indicated. In the inset in (b), the amplitude of the chaotic motion, which is large only for these values of r, is denoted by the error bars. The spatial patterns of the flow are also indicated. The indicated boundaries are those obtained by changing r away from that corresponding to the initial convecting state [i.e., $r = 1.31$ in (a) and $r = 1.94$ in (b)].

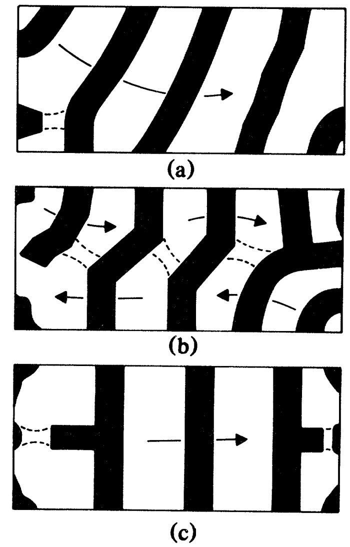

FIG. 2. Observed flow patterns shown schematically. Heavy lines indicate upflow boundaries which move in the direction indicated by the arrows: (a) translation state at $\psi = -0.35$, (b) "zipper state" at $\psi = -0.6$, and (c) translation state at $\psi = -0.6$.

Equation (1) assumes free-slip boundary conditions. Numerical calculations[5] indicate that, for the parameters of our experiments, the assumption of rigid, impervious boundaries increases the quantity $r_c - 1$ by about 5%. By measuring r_c, we determine ψ using Eq. (1); we find $\psi = -0.35$ for the data in Fig. 1(a), and $\psi = -0.6$ for the data in Fig. 1(b).[12]

The flow is observed to be time dependent when $r \lesssim 1.44$ for $\psi = -0.35$ [Fig. 1(a)] and when $r \lesssim 2.7$ for $\psi = -0.6$ [Fig. 1(b)]. The time-dependent flow pattern observed for $\psi = -0.35$ is illustrated in Fig. 2(a): Rolls are generated in one corner of the cell and move continuously to the opposite end of the cell. The particular direction of propagation in this translational state appears to depend on slight asymmetries in the initial conditions. We have, for example, been able to reverse the direction of propagation by transient heating of the bottom plate of the container. The period τ of the motion is indicated by the upper data set in Fig. 1(a). For comparsion, the vertical diffusion time, $\tau_v(\kappa) \equiv d^2/\kappa$, is 218 s for this mixture. Depending on the value of r, either periodic or chaotic states are observed. These same time-dependent states are observed when r is held constant as well as when the heat current through the fluid is held constant. As r is increased, the period of the motion increases. The flow is periodic above $r \simeq 1.34$, while the period τ continues to increase with r until the time dependence ceases abruptly above $r \simeq 1.44$. If r is then decreased, the onset of time dependence is hysteretic, beginning at $r \simeq 1.28$. When the time dependence does begin again, $N(r)$ and $\tau(r)$ are the same as those observed when r is approached from below. The period of the motion continues to decrease with decreasing r. The flow becomes chaotic very near the value of r at which convection ceases again via a first-order transition ($r \simeq 1.17$).

Data for $\psi = -0.6$ are shown in Fig. 1(b). Convection begins as r is raised above 2.53. The first convecting state, illustrated schematically in Fig. 2(b), is composed of two moving sets of rolls which originate in diagonally opposite corners of the cell. The subsequent motion of the two sets is confined to different halves of the cell. The motion of the two sets of rolls in the long direction of the cell is in opposite directions, and the roll ends near the center of the cell connect and disconnect as the motion proceeds. This "zipper" state is periodic at $r = 1.94$ with a period $\tau \sim 1900$ s. [For this case $\tau_v(\kappa)$ is 227 s.] As r is increased, the pattern undergoes a transformation to a purely translational state [illustrated in Fig. 2(c)], which is observed above $r = 2.3$. The time dependence ceases as r is increased beyond 2.7. The convection patterns and periods of motion obtained by decreasing r are similar (except for the hysteresis mentioned above) to those obtained by increasing r.

If r is decreased below $r = 1.9$ for $\psi = -0.6$, the zipper state is observed with τ continuing to decrease until the motion becomes chaotic at $r \simeq 1.4$. Associated with this chaotic state are flow patterns which are similar to that illustrated in Fig. 2(b), but which vary chaotically both in space and time. As r is decreased further, convection ceases at $r \simeq 1.35$ via a first-order transition. In Fig. 3 are shown the power spectra of the Nusselt number for three values of r in the vicinity of this chaotic region. The dominant frequency (which has increased to ~ 0.003 Hz near $\epsilon = 1.35$) and its harmonics and subharmonic broaden as r is decreased while an increasing, broad background is observed which peaks at zero frequency.

For the values of ψ studied here, previous theoretical work indicates that the flow should be time dependent at the onset of convection. For $L << 1 \lesssim P$, the period of this motion is predicted to be[5-7]

$$\tau_0 = (4/3\pi)\tau_v(\kappa)[(1+\psi+1/P)/(-\psi)]^{1/2}. \qquad (2)$$

Equation (2) assumes free-slip boundary conditions. Numerical calculations[5] indicate that the assumption of rigid, impervious boundaries decreases τ_0 by about 30%. Using Eq. (2), we find for $\psi = -0.35$ [Fig. 1(a)], $\tau_0 = 135$ s; and for $\psi = -0.6$ [Fig. 1(b)], $\tau_0 = 84$ s. In all cases, the period τ observed in our experiments is large compared with the predictions of Eq. (2); τ approaches τ_0 only at the smallest values of r for which convection is observed.

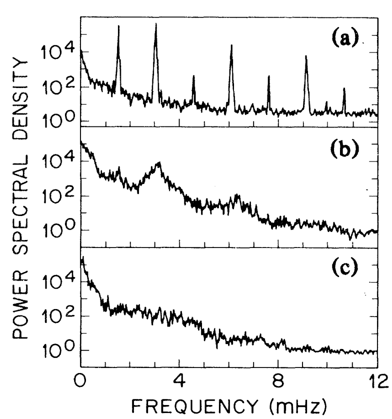

FIG. 3. Power spectra of the Nusselt number which illustrate the transition to chaos as r is decreased for $\psi = -0.6$: (a) $r = 1.424$, (b) $r = 1.392$, and (c) $r = 1.365$. The state shown in (a) is periodic; the width of the spectral lines and the level of the background noise in (a) are instrumental.

The theory for the oscillatory instability assumes small-amplitude flow. Thus, the relevance of the predicted oscillatory state to our experimental results is not clear. As in previous experiments,[5,7,8] we find that the oscillatory instability appears to trigger the onset of convection.[13] However, many of the properties of the resulting, finite-amplitude states are those which would be expected for time-independent convection. For example, excluding the chaotic region at lowest r, the time-averaged Nusselt number is that which one would expect for steady-state convection, and the observed fluctuations in N are less than 1%.

The observed time dependence is due to the translation of the rolls with velocity v. The period of the oscillation is then the time $\tau = \lambda/v$ for a roll pair to move one wavelength. The wavelength λ is approximately constant over the range of r studied; thus v has a strong dependence on r [i.e., for constant λ, $v(r) \propto \tau^{-1}(r)$].

A physical argument that a traveling-wave state might be preferred can be made in the following way. When convection begins, the rolls establish a lateral temperature wave of wavelength λ on a timescale $t \sim \tau_v(\kappa)$. For $\psi < 0$, the alcohol concentration readjusts on a slower time scale $t \sim d^2/D$ via the Soret effect to oppose convection with alcohol-concentration modulations and temperature modulations which are exactly opposite in phase. If there is then a relative displacement of the temperature and concentration waves, the convection can increase. But this relative displacement results in gradients of the alcohol concentration at the roll boundaries, and these gradients produce changes in buoyancy which tend to translate the roll pattern in the lateral direction. Whether such a concentration wave, not precisely opposite in phase with the temperature wave, is a self-consistent solution of the fluid equations and whether this is the origin of the traveling-wave states reported here awaits a realistic and nonlinear calculation. The cessation of time dependence at large r is probably due to the fact that the convective flow homogenizes the fluid.

The role of the aspect ratio of the cell and the cell walls in stabilizing or destabilizing the traveling-wave and chaotic states has not been investigated. The frequencies reported here are smaller by a factor of 2 or more than those observed[7] in larger–aspect-ratio containers of similar height, indicating that aspect ratio may play a role in the dynamics.[14] Unfortunately, the flow patterns were not studied in the previous experiments.[5,7]

The relationship of our experiments to the predictions[9] of spatially and temporally incoherent behavior near the onset of convection is unclear. As predicted, we do observe chaos for ψ large and negative. However, the theory assumes a slow modulation, both in space and time, of a flow pattern which is initially stationary in space but which oscillates with frequency $2\pi/\tau_0$. In constrast, our experiments indicate that the time dependence is due to a translation of the rolls.

We have observed traveling-wave states and states chaotic in both space and time in convection in binary fluid mixtures for which the separation parameter $\psi < 0$. The flow pattern near the onset of convection is chaotic. More generally, the traveling-wave states described here provide evidence of a new and interesting relationship between pattern selection and time dependence in convection in binary fluid mixtures.[15]

We would like to thank P. Hohenberg, G. Ahlers, M. C. Cross, and H. Greenside for helpful conversations and G. Dimino and N. Hartsough for their technical assistance.

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[9]H. R. Brand and V. Steinberg, Phys. Rev. A **29**, 2303 (1984). This work describes convection in a porous medium; an amplitude equation is derived which had previously been shown to exhibit spatially and temporally incoherent behavior. The authors suggest that similar behavior should occur in convection in bulk fluid mixtures.

[10]The thermodynamic properties are interpolated from the data in the *International Critical Tables for Physics, Chemistry, and Technology,* edited by E. W. Washburn (McGraw Hill, New York, 1928).

[11]G. E. Willis, J. W. Deardorff, and R. C. J. Somerville, J. Fluid. Mech. **54**, 351 (1972). To correct for finite aspect ratio, these predictions for infinite aspect ratio were multiplied by 0.73, which is the correction factor measured for pure water in the apparatus used in our experiments.

[12]This method of determination of ψ relies on the fact that the oscillatory instability triggers the onset of convection. We have directly confirmed that this is the case. (These results will be published elsewhere.) The values of ψ determined by this procedure agree reasonably well with the extrapolation of the only previous measurements for ethanol-water mixtures [L. J. Tichacek, W. S. Kmak, and H. G. Drickamer, J. Phys. Chem. **60**, 660 (1956)].

[13]For the values of ψ and L studied here, the fluid at rest is (linearly) stable to the onset of time-independent convection for all values of $r > 0$ (see Refs. 5–8).

[14]In Ref. 7, a circular container with a height d of 0.32 cm and a radius of 13 cm was used. The observed frequencies were more than a factor of 5 larger than those reported here. When the respective data are scaled by d^2, a discrepancy of a factor of 2 remains.

[15]There has been recent work on convection in binary ^3He-^4He fluid mixtures at low temperatures: I. Rehberg and G. Ahlers, Phys. Rev. Lett. **55**, 500 (1985) (this issue); and H. Gao and R. P. Behringer, private communication.