Transition from Traveling-Wave to Stationary Convection in Fluid Mixtures

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The transition from traveling-wave to stationary convection in a binary-fluid mixture is studied as a function of Rayleigh number. The experiments are conducted in an annular container which provides periodic boundary conditions in the direction of the wave motion. In agreement with the predictions of recent theories, the transition is shown to be continuous with no measurable hysteresis. The functional dependence of the wave phase velocity on Rayleigh number is in agreement with the predictions over the entire traveling-wave branch.

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Convection in binary-fluid mixtures provides a model system with which to study a wide range of spatiotemporal behavior. In experiments with ethanol-water mixtures, as the temperature difference across the fluid layer is increased, the conducting (motionless) state undergoes a Hopf bifurcation to a state of traveling-wave (TW) convection. For the parameter range of interest, this Hopf bifurcation is subcritical, and the linear convecting state grows into a large-amplitude nonlinear TW state before saturating. A wide variety of nonlinear phenomena have now been studied in this system, including the competition of counterpropagating traveling waves, stable fronts separating conduction and convection, and the dynamics of spatiotemporal defects.[1-3] Recently, nonlinear theories have been developed[4,5] and numerical calculations performed[6] to describe this large-amplitude TW state. This Letter reports experiments which directly test the predictions of these theories. The agreement between theory and experiment indicates that we now understand the nature of the uniform TW state in this system.

In a binary-fluid mixture the separation ratio ψ, which is proportional to the Soret coefficient, parametrizes the concentration-driven fluid density changes.[1] For $\psi < 0$, the lighter component diffuses toward the colder region, thereby stabilizing the fluid layer against convection, and the Rayleigh number at onset is larger than that for a pure fluid. In this Letter all Rayleigh numbers r will be normalized by the onset Rayleigh number R_c of convection in a pure fluid with the same thermal properties as the mixture; thus $r \equiv R/R_c > 1$.

At the onset of convection, the convective amplitude is observed to grow via a long transient to a state of slow-moving traveling waves;[7] at $\psi = -0.25$ the frequency is a factor of 30 lower than the Hopf frequency (ω_0). If the Rayleigh number is then reduced, this state of nonlinear traveling waves remains stable down to a saddle-node bifurcation back to conduction at r_s. Numerical calculations indicate that the linear concentration profile present in the conduction state is destroyed by convective mixing in the TW state.[8] Thus, the vertical concentration gradient remains only in thin, horizontal boundary layers at the top and bottom of the fluid layer, and the fluid is well mixed in the interior of the rolls. Concentration from these boundary layers is fed into the upflow and downflow boundaries between the rolls, forming a lateral concentration wave. It is the phase shift between this lateral concentration field and the lateral temperature field which leads to traveling convection rolls.[9]

Previous experiments have shown that, as r increases, the TW phase speed decreases and eventually stops, at a Rayleigh number which we denote by r^*, and a state of stationary overturning convection (SOC) results.[7,10] For sufficiently large convective amplitude, the concentration gradient due to the Soret effect is eliminated, and the resulting homogeneous fluid mixture exhibits the usual stationary convection as in a pure fluid.[9] In these experiments, which were conducted in a rectangular cell, the transition from TW to SOC was observed to be hysteretic,[7,10] and the TW phase speed was reported[10] to decrease linearly to zero as a function of $r^* - r$.

Recent theoretical work[5] and numerical calculations[6,8] have considered the nature of this TW state. The theory is a perturbation expansion in $|\psi|$ and $|r-1|$ around the solution for convection in a pure fluid, assuming infinite Prandtl number, while the numerical work is a finite-difference calculation. Both assume the Oberbeck-Boussinesq approximation of the fluid equations and two-dimensional flow perpendicular to the roll axes. They impose rigid, impermeable boundary conditions at the upper and lower plates of the container and periodic horizontal boundary conditions. In contrast to the experimental results described above, both the theory and the numerical calculations predict that the transition from TW to SOC should be continuous with no hysteresis at r^*, and the phase speed should decrease asymptotically as $(r^* - r)^{1/2}$.

To resolve these discrepancies, we have conducted experiments using an annular cell with a rectangular cross section which provides periodic horizontal boundary conditions in the direction of the TW propagation to match the assumptions of the theory and numerical calculations. The apparatus is similar to that described in Refs. 2 and 3. The convection cell has a rhodium-plated, mirror-polished copper bottom plate and a sapphire top plate. The top-plate temperature is set to 25.00 °C and regulated to ± 0.7 mK, and the bottom-plate temperature varies from 29.5 to 31.0 °C with a similar regulation. The vertical walls are machined plastic (ULTEM polyetherimide) of height $d=0.309$ cm. The cell is a long, narrow annulus with dimensions, in units of d, 1.288 in width ($\equiv\Gamma_r$) by 67.09 in mean circumference. The working fluid is a mixture of 8.00% by weight ethanol in water. At the onset of convection, the average temperature is 27.53 °C, and the fluid parameters are[11] $\psi=-0.257$, $P=9.16$, and $L=0.008$. The vertical thermal diffusion time $\tau_v\equiv d^2/\kappa$ is 74.2 sec, where κ is the thermal diffusivity.

The nonlinear convective state is composed of radially aligned rolls which propagate azimuthally when traveling. The flow is visualized from above with the shadowgraph method. In the shadowgraph image, the downflow roll boundaries are bright; their positions are followed in time with a circular photodiode array, and the image is digitized and stored by computer. The traveling-wave speed is calculated as a function of time from the average position of the entire pattern of the peaks in shadowgraph intensity.

Experimentally, the conduction state becomes unstable to linear traveling waves at $\Delta T_{c0}=5.062\pm0.003$ °C, corresponding to $r_{c0}=1.80\pm0.01$, where the error limit on r_{c0} is due to uncertainty in the fluid parameters. In a laterally infinite system, theory predicts onset at $r_{c0}=1.342$ at the critical wave number $k=3.137$.[12] In pure fluids, the onset of convection is suppressed in a cell with small lateral dimension perpendicular to the roll axis.[13] If we assume the same suppression factor $\Lambda\equiv 1+(1/2\Gamma_r^{1.75})$, the expected theoretical onset value would be $r_{c0}=1.773$, which is 1.6% smaller than observed.[14] The measured linear period $2\pi/\omega_0$ is 33.8 sec which corresponds to a phase velocity of $4.40d/\tau_v$ for the critical wave number, while the phase velocity expected[12] in a laterally infinite system for a linear wave at the Hopf frequency is smaller,[15] i.e., $3.63d/\tau_v$. To our knowledge, there are, at present, no predictions for the effect of Γ_r on phase velocity.

The linear TW state does not saturate but triggers the slow nonlinear TW state which grows to fill the cell with an integral number of roll pairs. The measured phase speed of the uniform TW convection as a function of Rayleigh number is shown in Fig. 1. The data correspond to a state with wave number $k=3.278$, traveling in a direction which we arbitrarily designate as positive. After a decrease (increase) in r, the speed increases (de-

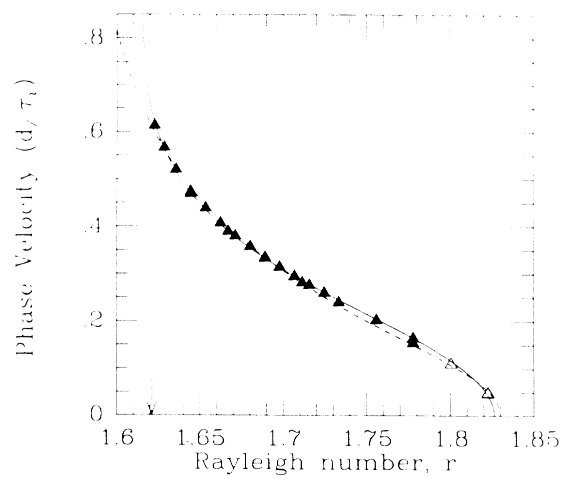

FIG. 1. The traveling-wave phase velocity as a function of Rayleigh number $r=R/R_c$ for $k=3.278$. The solid (open) symbols indicate decreasing (increasing) r. The arrow indicates the experimental value of r_s. The solid line is a least-squares fit of the prediction from the theory of Ref. 5, and the dashed line from the numerical calculations of Ref. 6.

creases) quasiexponentially, and we wait at least 5 e-folding times (corresponding to 12 to 36 hours) for the speed to come to equilibrium. The speed is then measured for typically twenty TW periods. The error in the measured TW speed is smaller than the symbol size. There are two data points at $r=1.645$ (1.689, 1.778, 1.832) which are taken during different ramps in Rayleigh number and occurred 9 (7, 60, 6) days apart. Thus the speeds are repeatable; however, the discrepancy at $r=1.778$ indicates that there may be a small, long-term drift.

The suppression of the linear TW instability by the narrow aspect ratio Γ_r appears to be consistent with the factor Λ computed for the onset of convection in pure fluids. However, it is not obvious that r^* and r_s, which characterize the nonlinear state, should be affected in the same way. Experimentally, we find $r^*=1.826$ and $r_s=1.621$, while the theory (numerical calculation) for a laterally infinite system predicts $r^*=1.226$ (1.650) and $r_s=1.152$ (1.214). To compare the actual magnitude of the TW phase speeds to the predictions, we examine the center of the TW branch at $r=(r_s+r^*)/2$. Here, the nonlinear TW phase speed is measured to be $0.263d/\tau_d$ for $k=3.278$, while the theory (numerical calculation) predicts 0.367 (0.249) at an assumed wave number of $k=3.117$ (π). Differences between the theory and numerical calculation are probably due to the fact that the former is a perturbation expansion. A likely source of discrepancy between the predictions and experiment for r^*, r_s, and the phase speed is the effect of the finite transverse aspect ratio. Thus, calculations including these effects would be of interest.[16]

In Fig. 1, we compare the functional dependence of the phase velocity on the Rayleigh number between theory and experiment. The two curves shown are least-squares fits of the theoretical and numerical predictions from Refs. 5 and 6 for $\psi=-0.25$ and $L=0.01$, and wave numbers $k=3.117$ and $k=\pi$, respectively. The free parameters in the fits are r^* and the scale in each direction. The fit with the perturbation theory[5] is excellent over the entire TW branch, from the fastest nonlinear TW state near the saddle node at r_s to the slowest at r^*, with an average rms error of $0.0040 d/\tau_v$, i.e., about the size of the experimental uncertainty near the center of the TW branch. The fit with the finite-difference, numerical calculation[6] gives an rms error about twice as large; the discrepancy is due mostly to the difference in shape between this calculation and both the theory and experiment near r_s. In the experiments, TW convection was lost to conduction at $r_s=1.621$, compared to 1.616 for the fit by the theory and 1.581 for the fit by the numerical calculation.

Near r^*, the theory and numerical calculations agree with each other and can be well fitted to the experimental data. Figure 2 shows an expanded view of phase-velocity data near the transition to stationary convection for a slightly different uniform state. These TW's also have wave number $k=3.278$, but travel in the negative direction. The predictions from the perturbation theory and numerical calculations are fitted to the data by allowing r^* and the vertical scale to vary but using the same value of r_s as the fits shown in Fig. 1. Both fits are excellent near the transition to stationary convection and give $r^*=1.8235$ and 1.8236, respectively. The discrepancy in Fig. 1 of the fits near r^* arises from the balance of the least-squares error minimization near r_s.

The data for increasing and decreasing Rayleigh numbers shows no hysteresis in the value of r^*. In particular, separately fitting the perturbation theory to just the increasing- or decreasing-Rayleigh-number data in Fig. 2 gives values for r^* which differ by less than 0.02%. In contrast, a 15% hysteresis in r^* was reported in experiments in a rectangular cell.[7] Thus, the prediction[5] that the transition should be continuous in an annular geometry is supported by our experimental results.

From bifurcation analysis, the phase velocity should decrease asymptotically as $(r^*-r)^{1/2}$ near r^*,[4] and both the theory and numerical calculations predict this dependence. The data in Fig. 2 and data taken for another state with $k=3.184$ follow a power law, $a(r^*-r)^b$, over the range in r shown. A least-squares fit varying a, b, and r^* gives an average rms error 20% smaller than the fits in Fig. 2, with $b=0.58\pm0.01$ (0.57 ± 0.01) for data with $k=3.278$ (3.184). Although the exponent b is close to 0.5, the discrepancy with the predictions remains even when the least-squares fits are limited to points closer to r^*.

In Fig. 3, all the data of Figs. 1 and 2 along with additional data at larger Rayleigh numbers and data at an additional wave number are shown on a logarithmic scale. We can resolve TW speeds greater than $\sim 3\times10^{-4} d/\tau_v$ (corresponding to the horizontal dashed line in Fig. 3); speeds below that were regarded as stationary convection. For $r>r^*$, we sometimes observe a very slow state which usually travels in the positive direction with speeds $(0.001\text{–}0.005) d/\tau_v$. In other runs, we observe stationary convection at the same values of r.

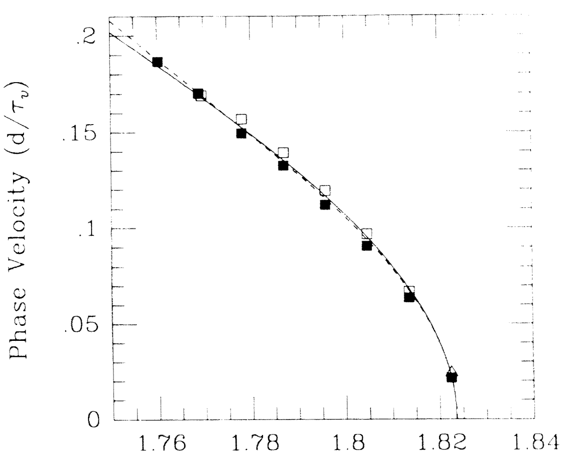

FIG. 2. TW phase velocity vs Rayleigh number on an expanded scale near r^* for $k=3.278$. The solid (open) symbols indicate decreasing (increasing) r. Squares (triangle) denote negative (positive) direction traveling waves. Lines are least-squares fits as in Fig. 1, using the values of r_s determined in Fig. 1.

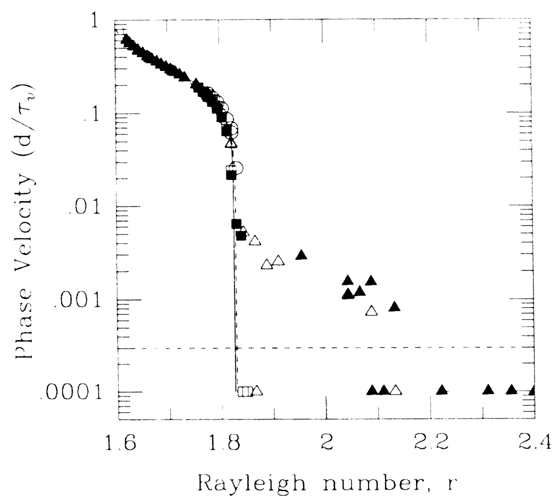

FIG. 3. Phase velocity vs Rayleigh number on a semilogarithmic plot. The horizontal dashed line indicates the limits of our resolution; the points below are regarded as stationary convection. Solid (open) symbols indicate decreasing (increasing) r. Triangles denote positive-direction TW's with $k=3.278$; squares, negative with $k=3.278$; circles, negative with $k=3.184$. The lines are the fits in Fig. 1.

For $r \sim 2.1$, this state can be intermittent, and the rolls sometimes stop and start aperiodically. When approaching r^* from below, rolls traveling in the negative direction were sometimes observed to stop and reverse direction over very long times at fixed r just below r^*, perhaps indicating that a low-level current in our apparatus may be responsible. However, we once observed the slow state to start in the negative direction from stationary convection upon lowering r. Further studies of these slow states will be described elsewhere.[17] The excellent fit with the theory and numerical calculations for the data below r^* indicates that some other physics may be important in the stationary-convection regime.

In this Letter, we have presented a quantitative study of the speed of nonlinear traveling-wave convection in a binary-fluid mixture as a function of Rayleigh number. There is agreement of the dependence of TW phase velocity on Rayleigh number with a recent theoretical model and a numerical calculation, from the saddle-node bifurcation to conduction at low r to the transition from TW to stationary convection at higher r, spanning a factor of 30 in phase velocity. The prediction that this transition is continuous with no hysteresis is verified to a level $\Delta r^*/r^* < 0.02\%$. Near r^*, the dependence of the TW phase velocity on Rayleigh number is found to be $(r^* - r)^b$, with $b \approx 0.57$, while the predicted value is $b = 0.5$. These results indicate that we understand nonlinear TW convection in mixtures in the case where the TW amplitude is uniform in the direction of propagation. Agreement between the predictions and experiment at the TW$\rightarrow$SOC transition is particularly significant, since we are considering not a primary but a secondary bifurcation. The model of TW convection tested here is likely to be an important starting point in understanding the dynamics of the fronts, pulses, and defects observed[1-3] in this nonequilibrium system.

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