

Weakly Nonlinear Traveling-Wave Convection

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Experimental results are presented for traveling-wave states near the onset of convection in a fluid mixture in a rectangular container. When the Soret-effect-induced stabilization is sufficiently weak, the nonlinear oscillatory states at onset evolve in time from the linear states of the system and remain in a pattern of parallel rolls with a frequency which is always near the Hopf frequency (i.e., $\Delta f/f_0 \lesssim 0.15$). However, these states exhibit erratic spatial and temporal behavior which is apparently due to the nonlinear coupling of the counterpropagating traveling waves.

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Rayleigh-Benard convection in a fluid layer heated from below has been an extremely useful system in which to test our understanding of pattern selection and dynamics in nonequilibrium systems. An important variant of this physical situation is convection in the class of fluid mixtures where the first convecting state is oscillatory in time. In this case, the bifurcation is to a state whose wave number and frequency are both finite.¹ This allows us the possibility of studying traveling-wave phenomena in a model nonequilibrium system.²⁻⁴ Thus far, however, in experiments where it has been possible to visualize the often-complicated flow patterns, the only flows which have been studied are those in which the flow evolves to a state of large-amplitude convection^{2,5,6} whose character is quite different from that of the linearly unstable modes which are now well understood.^{3,7,8} Specifically, the oscillation frequency, which is related to the stabilizing, vertical concentration gradient, measures the departure of the concentration profile from that in the conducting state. In the linear state, the oscillations occur at the Hopf frequency, and the concentration gradient is the same as that in the conducting state. In previously studied nonlinear states, however, the frequency is lower by a factor of from 2 to 100, indicating that the concentration gradient has been strongly altered. Thus, it is not likely to be possible to describe such strongly nonlinear states by a perturbative analysis of the conducting state.

In this Letter, we show that, by suitably adjusting the parameter, ψ , which measures the stabilization of the fluid layer due to the Soret effect, it is possible to weaken the nonlinearity and still produce complex dynamical states near onset. These states exhibit spatial patterns which are closely related to those of the linear states, and their frequency at onset is within 15% of the Hopf frequency. We interpret this to mean that the concentration profile is not strongly modified. Thus, it appears possible to develop an accurate perturbative description of these states which could account for their interesting dynamical behavior. This would be the first time that such dynamical behavior could be described in detail

from first principles. Recent results from both an amplitude-equation model⁹ and from a numerical simulation¹⁰ exhibit many of the features reported in this Letter.

The apparatus, which has been described elsewhere,⁸ consists of a rectangular container of dimensions $1 \times 4.9 \times \Gamma$, in units of the depth, $d = 0.351$ cm. The major aspect ratio, Γ , can be varied from 11 to 20. The bottom plate is polished, gold-plated copper, and the top plate is sapphire. The flow is visualized from above with the shadowgraph method.

Convection in a pure fluid can be parametrized by 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 viscosity to the thermal diffusivity. The description of convection in fluid mixtures requires two additional parameters.^{1,11} The Lewis number, L , is the ratio of the diffusivity of concentration to that of heat. Finally, the separation ratio ψ is a measure of the stabilizing effect on the fluid layer produced by Soret-effect-driven concentration gradients; it is defined by $\psi = -c(1-c)(\partial\rho/\partial c)_T(\partial\rho/\partial T)_c^{-1}S_T$, where S_T is the Soret coefficient, ρ is the density, T is the temperature, and c is the concentration.

In this Letter, we study a mixture of 0.3 wt. % ethanol in water at mean temperatures from 10 to 30°C, for which ψ is negative (i.e., the vertical concentration gradient is stabilizing) and varies from -0.01 to -0.04 .¹² The Lewis number varies from 0.006 to 0.01, and P varies from 8.9 to 5.5.¹² For these values of ψ and L , convection begins with an oscillatory instability at a Rayleigh number $R_{c0} \approx R_c(1-\psi)$,^{8,11,13} where R_c is the onset of convection in the homogeneous fluid mixture. The oscillation period is $\tau_0 \approx A(-\psi)^{-1/2}$, where A depends on the boundary conditions and is of the order of the vertical thermal diffusion time.^{8,11,13}

Previous experiments^{3,8} and those reported here indicate that, for $-0.01 \gtrsim \psi \gtrsim -0.55$, convection begins with a subcritical bifurcation to a state of linear, counterpropagating traveling waves (CPTW),⁷ consisting of equal amplitudes of right- and left-going waves which

propagate parallel to the long sides of the container.³ The amplitude of this state can be made neutrally stable by setting the reduced Rayleigh number $\epsilon \equiv (R - R_{c0})/R_{c0}$ equal to zero after allowing the oscillations to grow to some amplitude. For $\Gamma = 15.51$ and $\psi = -0.021$, the observed oscillation period of these linear CPTW is $\tau_0 = 190$ s, which is close to the value of 189.6 s computed from the known parameters¹² of the system. If ϵ is then increased, the amplitude of the oscillations grows smoothly, and the CPTW state evolves to a state with the time dependence illustrated in Fig. 1(a). In the regions marked I, a CPTW state is observed with an oscillation period τ which increases smoothly from the Hopf period τ_0 as time proceeds [see Fig. 1(b)]. As the period lengthens and the amplitude increases, this state evolves to a *modulated state* (regions denoted by II in Fig. 1) in which the relative amplitudes of left- and right-going waves vary with time, leading to a modulation of both the amplitude and the period [e.g., as seen between times $t = 2.0$ and 4.3 h in Figs. 1(a) and 1(b)]. For this aspect ratio, the modulated state is itself unstable; the convection amplitude is observed to decrease abruptly by a factor of 5 or more (e.g., at $t = 4.5$ h in Fig. 1); then, after a transient, the cycle repeats, beginning with the linear growth of the CPTW state.

The character of these states depends sensitively on the aspect ratio Γ and is approximately periodic with integer period. At $\Gamma = 15.51$ [Figs. 1(a) and 1(b)], the separation of regions of modulation (regions II) from the

regions of linear growth and CPTW (regions I) is obvious. At other values of Γ (e.g., $15.8 \lesssim \Gamma \lesssim 16.2$), the state is closer to one of continual modulation [e.g., Figs. 1(c) and 1(d)]. This dependence on aspect ratio contrasts with the linear-mode beating observed previously,⁸ where persistent modulation was seen only at particular values of Γ which differ by an integer.

Shown in Fig. 2 is an example of the behavior of a modulated state in space and time, near the extremes of the modulation. Note that the amplitude of convection is very small in one side of the container when it is large in the other side, and that this state consists primarily of waves propagating in the direction of large amplitude. The strength of the modulation, which is quite large in Fig. 2, is sensitive to Γ and ϵ ; for other values of these parameters [e.g., for the data in Fig. 1(a)], we observe a smaller modulation of the amounts of left- and right-going waves. The period of the modulation varies from cycle to cycle and ranges from $\Gamma\tau_0$ to $3\Gamma\tau_0$. For appropriate values of Γ , we have previously observed modulation due to the beating of two *linear* modes; however, in this case, the modulation is accurately periodic with period $\Gamma\tau_0(\partial\omega/\partial k)^{-1} \approx 1.07\Gamma\tau_0$.⁸

The modulated states observed near $\Gamma = 16$ appear to be chaotic in time. However, at the small values of ϵ studied, we cannot completely rule out the possibility that some of the variations on time scales longer than

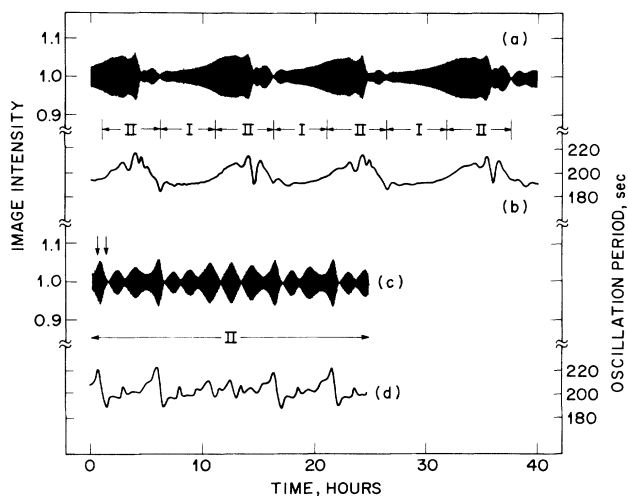


FIG. 1. The shadowgraph image intensity at a point in the container and the period of oscillation are shown as functions of time: (a) and (b) correspond to $\Gamma = 15.51$ and (c) and (d) to $\Gamma = 16.25$. The intensity is measured at a position of $\frac{1}{4}$ of the long dimension of the cell from one end wall (i.e., at position $L/4$ in Fig. 2). The reduced Rayleigh number is $\epsilon = 3 \times 10^{-4}$, and the calculated Hopf period is 189.6 s. As discussed in the text, numerals I and II refer to the spatial state of the convection.

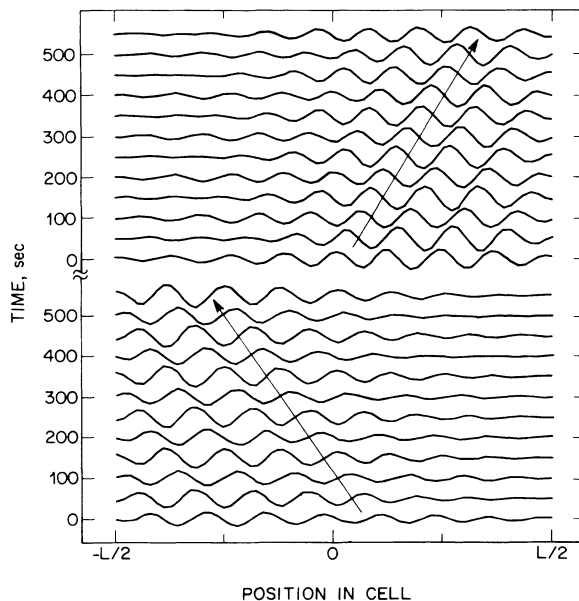


FIG. 2. The amplitude of convection in a modulated state, as measured by the shadowgraph intensity averaged along the direction parallel to the roll axes, is plotted as a function of position parallel to the long axis of the container. Two 700-s intervals near the extrema of the modulation are shown, corresponding to the intervals marked with arrows in Fig. 1(c). The aspect ratio Γ is 16.25, and $\epsilon = 3 \times 10^{-4}$.

$\Gamma\tau_0$ may be due to small fluctuations in Rayleigh number. When the aspect ratio is reduced to $\Gamma \approx 10$, the modulation is regular, and the states are quasiperiodic.

The rms amplitude of the oscillatory convection is shown as a function of ϵ in Fig. 3. The amplitude depends weakly on ϵ , increasing by a factor of 3 as ϵ is changed from 3×10^{-4} to 0.02. As ϵ is increased, τ_{\max} also increases, so that, at $\epsilon \approx 0.02$, $\tau_{\max} \approx 2.0\tau_0$. If ϵ is decreased below zero, the flow ceases. The solid curve in Fig. 3 has the form

$$\epsilon A + \alpha A^2 - \beta A^4 = 0, \quad (1)$$

chosen to represent the observed subcritical bifurcation. The constants were adjusted to best represent the data corresponding to two-dimensional patterns of straight rolls (filled circles in Fig. 3). We note that this curve predicts a small amount of hysteresis which is not observed in the experiment.

The spatial pattern of the flow also changes with ϵ . For $\epsilon \lesssim 6 \times 10^{-3}$, the rolls are straight and parallel to the short side of the container. For $\epsilon \gtrsim 3 \times 10^{-4}$, convection is suppressed near the long sides of the container. At larger ϵ , the pattern becomes more complicated. At $\epsilon \approx 1 \times 10^{-2}$, the amplitude of the flow is very small in all but a narrow, cigar-shaped region which is nearly but not exactly parallel to the long axis of the cell¹⁴ and whose location oscillates in time between the two ends of

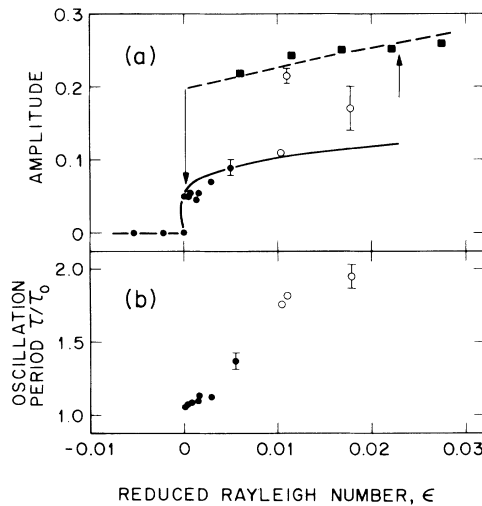


FIG. 3. (a) The maximum amplitude of convection, and (b) the maximum period of oscillation normalized to the Hopf period, τ_{\max}/τ_0 , are shown as functions of reduced Rayleigh number ϵ . Filled circles correspond to patterns of straight rolls parallel to the short side of the container. Open circles correspond to patterns in which the rolls are confined to oblique, cigar-shaped regions whose positions oscillate between the two ends of the cell. Squares indicate the amplitude of stationary convection.

the cell.

For $\psi = 0.021$, we observe a transition at $\epsilon = 0.022$ to stationary convection whose amplitude near $\epsilon = 0$ is approximately 4 times that of the oscillatory state at $\epsilon = 0$. As a result of the stabilizing effect of the concentration gradients for $\psi < 0$, we expect the amplitude in this state to be no greater than that in a pure fluid with the same physical properties. This indicates that the amplitude of the oscillatory state described here is quite small at onset, being at most $\frac{1}{4}$ that of convection in a pure fluid at a reduced Rayleigh number $\epsilon \equiv (R_{c0} - R_c)/R_c = 0.029$.

As ϵ is decreased from $\epsilon > 0.023$, the flow continues to be stationary until $\epsilon \approx 6 \times 10^{-4}$, where oscillatory convection is again triggered. The character of this oscillatory state is similar to that observed when this value of ϵ is approached from below.

At $\psi = 0.011$, the states near onset are qualitatively similar to those described for $\psi = -0.021$. In both cases, the range of ϵ in which oscillatory convection is observed is $\Delta\epsilon \approx |\psi|$, and the maximum amplitude of the observed oscillatory convection increases by about a factor of 3 when ϵ is increased from zero to $\Delta\epsilon \approx |\psi|$, at which point stationary convection is triggered. Similar results have been obtained at $\psi = -0.043$, where we have also observed a transition from modulated states to a time-independent confined state^{5,6} before the transition to stationary convection.

Cross has recently carried out numerical simulations of a one-dimensional, amplitude-equation model of oscillatory, traveling-wave convection in a finite container.^{7,9} This calculation assumes a supercritical bifurcation, which we do not observe, and so quantitative comparison with the experiment is not possible. However, the qualitative nature of the solutions is similar to that of the modulated states described here. Cross's solutions depend on the parameter $\bar{s} = s(\epsilon^{1/2}\gamma'\xi_0)^{-1}$, where s is the group velocity, $\gamma'\epsilon$ is the linear growth rate, and ξ_0 is a coherence length.⁷ Large \bar{s} implies that a wave packet propagates many coherence lengths in an exponential growth time and corresponds to small-amplitude states near threshold, such as those studied here. For such values of \bar{s} , Cross finds time-dependent solutions which can be thought of in the following way: The left- and right-going waves are fed back due to reflections from the end walls, and the sign of the nonlinear coupling between these two components leads to a competition between them, causing a temporal modulation of their relative amplitudes. The ratio, $\bar{\tau}$, of the period of the modulation to the linear, round-trip, pulse-propagation period is $1.7 \lesssim \bar{\tau} \lesssim 3.7$ for the theory, which is in reasonable agreement with the values $1 \lesssim \bar{\tau} \lesssim 3$ observed in the experiment.

Recently, Deane, Krobloch, and Toomre¹⁰ have carried out numerical simulations of thermosolutal convection, a problem which is closely related to that considered here. They observe unidirectional traveling

waves which exhibit aperiodic reversals of the propagation direction on a time scale similar to that observed in our experiments. More detailed comparisons will require similar calculations to be done for the Soret problem.

The fact that the nonlinear states observed here are more closely related to the linear states of the system than those studied previously is likely to be due, in part, to the fact that, at small ψ , the maximum-amplitude convection is expected to be smaller. In addition, at the values of ϵ where we observe states similar to the linear states of the system (i.e., $\epsilon \lesssim L^2$), the characteristic boundary layer for the concentration profile¹⁵ is comparable to the cell depth. Both of these effects can be expected to lead to a profile of the stabilizing concentration gradient which is similar to that of the conducting state (i.e., a simple linear profile). Thus, the observation that these states have a frequency near the Hopf frequency is plausible and may help to justify the validity of a perturbation analysis.

In this Letter, we have reported a study of traveling-wave convection near onset, where the nonlinear states are similar in frequency, wavelength, and pattern to the linear states of the system. These states are not steady, but vary aperiodically both in amplitude and in propagation direction.¹⁶ Since these nonlinear states retain much of the character of the linearly unstable waves and since their dynamics appear to be due to a weak, nonlinear coupling of left- and right-going traveling waves, it is likely that a rigorous theory of this rich dynamical behavior can be developed from the hydrodynamic equations. The qualitative agreement of the results presented here with recent theoretical work is encouraging in this regard.

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berg, E. Knobloch, and B. Shraiman, and we wish to thank A. Passner and H. L. Williams for their excellent technical assistance.

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¹⁶At the time of submission of this manuscript, we received a manuscript from J. Fineberg, E. Moses, and V. Steinberg, preceding Letter [*Phys. Rev. Lett.* **61**, 838 (1988)], which describes related observations on the same experimental system.