## **Oscillatory Traveling-Wave Convection in a Finite Container**

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It has been proposed that the oscillatory traveling-wave states recently observed at the onset of convection in ethanol-water mixtures can be explained in terms of the linear properties of the waves, taking into account their reflection at lateral boundaries. We present experimental results which support this picture and explore a number of properties of this physical situation, including the existence of states which are modulated by a second frequency at onset.

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The recent discovery that convection in binary fluid mixtures takes the form of traveling waves has been the subject of both experimental<sup>1-4</sup> and theoretical<sup>5,6</sup> work. In this Letter, we demonstrate the validity of a model<sup>6</sup> of the nature of the oscillatory, traveling-wave states which are observed at onset in finite-sized containers, and we explore properties of these states which are qualitatively different from those observed in convection in pure fluids.

Convection in mixtures is described by four dimensionless parameters: the Rayleigh number R, which is proportional to the temperature difference across the layer; the Prandtl number P, which is the ratio of the viscosity to thermal diffusivity; the separation ratio  $\Psi$ ; and the Lewis number  $\mathcal{L}$ . The quantity  $\Psi$  is a measure of the destabilizing effect of concentration gradients produced by the Soret effect:

$$\Psi = -c(1-c)(\partial \rho/\partial c)_T (\partial \rho/\partial T)_c^{-1} S_T,$$

where c is concentration,  $\rho$  is density, T is temperature, and  $S_T$  is the Soret coefficient; and the Lewis number,  $\mathcal{L}$ , is the ratio of the diffusivity of concentration to that of heat.

For  $\Psi \lesssim -\mathcal{L}^2$ , convection is expected to begin as an oscillatory state with a well-defined frequency. Recent studies of the onset of convection in ethanol-water mixtures show that the conducting (zero-flow) state does become unstable to oscillatory convection as expected, but that, at fixed Rayleigh number, the oscillations grow until overturning convection is triggered.<sup>3</sup> Similar transients had been observed previously in thermohaline convection in a beautiful set of experiments by Caldwell,<sup>7</sup> but to our knowledge, the implications of these results had not been generally appreciated.

Experiments in ethanol-water mixtures<sup>3</sup> established that these oscillatory states can be described in terms of counterpropagating traveling waves which grow exponentially in space as they propagate. Cross suggested that these states can be understood in terms of the linear properties of the oscillatory traveling waves.<sup>6</sup> The spatial growth is determined by the temporal growth rate and the group velocity; and, in order to achieve a state in which the amplitude of oscillation is constant in time, the Rayleigh number must be adjusted so that the exponential growth in space compensates the loss on reflection at the end walls. In this Letter, we present experimental results which provide quantitative tests of Cross's model,<sup>8</sup> and we go on to demonstrate fundamental differences between the nature of these oscillatory flow patterns and those observed in pure fluids. Examples include the boundary conditions on the traveling waves at the end walls of the container and the observation of self-modulated states, which are composed of two waves with different frequencies and wave numbers, each satisfying a resonance condition in a container of finite extent.

The experimental apparatus, which is described elsewhere,<sup>3,9</sup> consists of a cell with plastic walls, a copper bottom plate, and a sapphire top plate. The cell is 0.47 cm in height and has one lateral dimension of 3.76 cm. An important feature of the experiments reported here is the ability to vary the second lateral dimension, L, of the container continuously from 5.5 to 8.5 cm by a movable end wall. The working fluids are mixtures of ethanol and water, with weight concentrations of ethanol ranging from 0.35% to 27%, operating at top-plate temperatures of from 10 to 30 °C. The Lewis number  $\mathcal{L}$  ranges from 0.005 to 0.009. The entire flow pattern is visualized from above with use of a shadowgraph technique. The image of the flow is recorded with a charge-coupleddevice camera and analyzed by use of digital imageprocessing techniques.

In previous studies of oscillatory convection in ethanol-water mixtures, it was established that the oscillations have a frequency and an onset Rayleigh number which are consistent with linear theory, and that the flow pattern consists of rolls oriented parallel to the shorter lateral dimension.<sup>3</sup> The observed convection amplitude as a function of space and time for  $\Psi = -0.55$  is shown in Fig. 1. The data are well described by a function of the form

$$A(x,t) = A_0 e^{\gamma t} [e^{x/t} \cos(k_0 x - \omega_0 t) - e^{-x/t} \cos(k_0 x + \omega_0 t)],$$

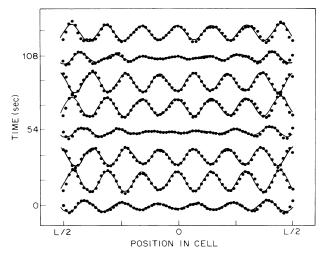


FIG. 1. The image intensity (closed circles) as a function of position along the long dimension of the cell at several times for  $\Psi = -0.55$ . The period of oscillations is 51 s, and the aspect ratio is 14.24. The solid line is a fit to the data with use of Eq. (1).

where  $\omega_0$  and  $k_0$  are the frequency and wave number of the waves, and  $\gamma$  and  $l^{-1}$  are the temporal and spatial growth rates of the slowly varying envelope. We have shown experimentally that  $\gamma$  is proportional to the reduced Rayleigh number,  $\epsilon = (T - R_{c0})/R_{c0}$ , where  $R_{c0}$  is the observed Rayleigh number at the onset of convection.<sup>3</sup> The data in Fig. 1 were taken by the adjustment of  $\epsilon$  so that  $\gamma = 0$  in order to achieve a (neutrally stable) steady state.

Cross proposed that convection patterns such as those illustrated in Fig. 1 can be explained in terms of the linear properties of traveling waves of oscillatory convection.<sup>6</sup> In particular, he pointed out that, if the reflectivity r of the end wall is less than unity, then, to achieve a steady state, the wave must grow in amplitude as it traverses the cell. For such a state, the exponential growth length l will be given by<sup>6</sup>

$$l = L [\ln(1/r)]^{-1}.$$
 (2)

Thus *l* is directly proportional to *L*. Shown in Fig. 2 are measured values of l/L for various values of aspect ratio,  $\Gamma \equiv L/d$ , where *d* is the height of the cell. Cross's prediction that l/L is independent of  $\Gamma$  is verified by the data.

Cross calculates values of the reflection coefficient r as a function of  $\Psi$  in a model which assumes rigid, impermeable end walls and free-slip, permeable, horizontal boundaries. Values of r inferred from the measured growth length l and Eq. (2) as compared with Cross's predictions (indicated in brackets) are 0.31 [0.12], 0.30 [0.20], and 0.29 [0.28] for values of  $\Psi$  of -0.06, -0.28, and -0.55, respectively. Thus, the predicted and measured values of r are in good agreement at large negative

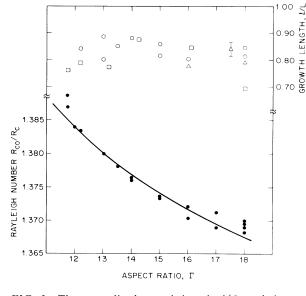


FIG. 2. The normalized growth length, l/L, and the onset Rayleigh number,  $R_{c0}/R_c$ , as functions of aspect ratio, for  $\Psi = -0.28$  (circles), -0.45 (triangles), and -0.55 (squares). The quantity  $R_{c0}$  is calculated with use of the observed temperature difference at onset and the thermal properties of the mixture, and  $R_c = 1708$ .

 $\Psi$  but disagree at smaller  $|\Psi|$ .

The model also predicts that the attenuation of the traveling waves upon reflection at the boundaries will lead to a shift,  $\Delta\epsilon$ , in the onset Rayleigh number. This is because the Rayleigh number of the quasisteady state must be sufficiently far above the onset of oscillatory convection in an infinite system to produce the spatial growth required to compensate the loss on reflection. We have previously observed that the temporal growth rate,  $\gamma$ , is a linear function of  $\epsilon$  and have measured  $\partial \gamma / \partial \epsilon$ .<sup>3</sup> If the onset of convection is exceeded by  $\Delta\epsilon$ , then the growth length *l* is given by the expression  $(\partial \gamma / \partial \epsilon) \Delta \epsilon = (\partial \omega / \partial k)_{k_0} l^{-1.6}$  Thus  $\Delta \epsilon = B / \Gamma$ , where

$$B = (\partial \omega / \partial k)_{k_0} (\partial \gamma / \partial \epsilon)^{-1} (ld/L)^{-1}.$$

Since l/L is independent of L in a steady state,  $\Delta \epsilon$  is inversely proportional to  $\Gamma$ . Note that this expression is qualitatively different from the effect of aspect ratio on the onset Rayleigh number in pure fluids, where the lowest-order correction is  $\Delta \epsilon \sim \Gamma^{-2}$ . For the flow patterns shown in Fig. 1, this second-order correction is 2 orders of magnitude smaller than that which we observe.<sup>6</sup>

Shown in Fig. 2 are data for  $R_{c0}/R_c$  as a function of  $\Gamma$  for  $\Psi = -0.28$ . The solid line is a fit by the form  $\Delta \epsilon = B/\Gamma$ . The value of *B* inferred from this fit is 0.44. For comparison, measurements of *l*,  $(\partial \omega/\partial k)_{k_0}$  (described below), and  $\partial \gamma/\partial \epsilon$  yield a value of B = 0.39.

Under most conditions, the onset of oscillatory convec-

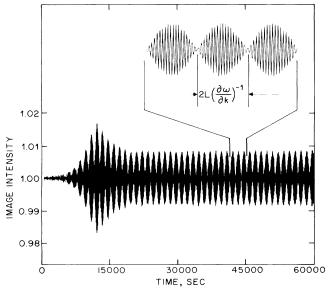


FIG. 3. The image intensity at one spatial point as a function of time for a self-modulated state with  $\Psi = -0.45$ . Initially, the Rayleigh number was adjusted for a positive growth rate; later, the growth rate was reduced to zero to achieve a steady state.

tion has been observed to begin with a smooth, exponentially growing transient with frequency  $\omega_0$  and growth rate  $\gamma$ . However, we have also observed a qualitatively different behavior, illustrated in Fig. 3, where the transient is modulated at a lower frequency. These states also have an overall growth rate which is proportional to  $\epsilon$  and can be made neutrally stable by suitable adjustment of  $\epsilon$ . The time interval between zeros in this modulated state is found to be approximately  $2L(\partial \omega/\partial k)_{k_0}^{-1}$ , which is the round-trip transit time of the oscillatory traveling waves. As shown in Fig. 4, this low-frequency modulation corresponds to an oscillation of the wave amplitude from one side of the cell to the other. Observation of the waves at times near 250 and 750 s reveals traveling waves propagating in one direction with a phase velocity equal to  $\omega_0/k_0$ . These fully modulated states are observed at values of the aspect ratio which are separated from each other approximately by integers.

The existence of such modulated states at particular values of  $\Gamma$  can be understood by our noting that, for constructive interference of the waves in a cavity of length L, the wave number must satisfy a resonance condition such that  $2(k_nL + \phi_r) = 2n\pi$ , where  $\phi_r$  is the phase shift on reflection. The system has a linear gain<sup>6</sup>

$$\gamma(\epsilon,k) = (\partial \gamma/\partial \epsilon) [\epsilon - \xi_0^2 (k - k_c)^2], \qquad (3)$$

where  $\xi_0$  is a coherence length and  $k_c$  is the critical wave number. In a steady, unmodulated state, such as that shown in Fig. 1, the mode with wave number  $k_n$  closest to  $k_c$  will have zero growth rate, while all others will decay. However, if the gain curve is centered between two

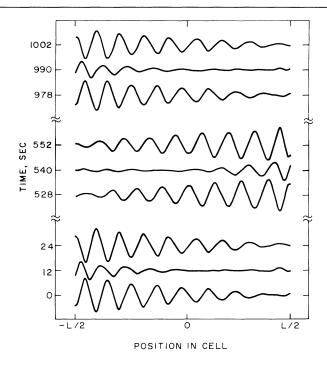


FIG. 4. The image intensity as a function of position along the length of the cell at several times during one-half of the cycle of modulation in a self-modulated state at  $\Psi = -0.55$ .

modes with mode numbers n-1 and n, then both modes will have comparable growth rates, and the convection pattern will exhibit their interference. For example, if we consider the two waves traveling in the positive xdirection in an infinite system, the amplitude A(x,t) will be

$$A(x,t) = \cos(\pi u/2L)\cos kv, \qquad (4)$$

where  $u = x - (\partial \omega / \partial k)_{\bar{k}}t$  and  $v = x - (\bar{\omega} / \bar{k})t$ , with  $\bar{k} = (k_n + k_{n-1})/2$  and  $\bar{\omega} = (\omega_{k_n} + \omega_{k_{n-1}})/2$ . The low-frequency modulation as a function of the variable u with spatial period 4L corresponds to that shown in Figs. 3 and 4. The boundary conditions at the end walls determine the relative phase of left- and right-going waves, and the resulting expression for the total amplitude is in excellent agreement with the data in Fig. 4.<sup>10</sup>

Measurement of the temporal modulation frequency gives a direct measure of the group velocity  $(\partial \omega/\partial k)_{k_0}$ . For the data shown in Fig. 4, we find  $(\partial \omega/\partial k)_{k_0} \approx (0.93 \pm 0.01)(\omega_0/k_0)$ . By measuring the relative growth rate of the two modes in the modulated state as a function of cell length, we are also able to measure the coefficient,  $\xi_0^2$ , in Eq. (3) and find  $\xi_0^2/d^2 = 0.14 \pm 0.01$ . For comparison, recent calculations assuming rigid, impermeable, horizontal boundaries predict  $(\partial \omega/\partial k)_{k_0} = 0.97(\omega_0/k_0)$ and  $\xi_0^2/d^2 = 0.145$  for this value of  $\Psi$ .<sup>10</sup>

The experiments reported here provide strong support for Cross's model of oscillatory, traveling-wave convection in a finite container. Specifically, we have verified that the growth length l is independent of L and that there is a shift in the onset Rayleigh number as a function of aspect ratio which is in quantitative agreement with that expected.

Cross's theory and the experiments described here reveal qualitative differences between the onset of convection in binary and in pure fluids. In convection in pure fluids near onset, there is a macroscopic healing length  $\tilde{\lambda} \sim d/\epsilon^{1/2}$  near an end wall because the fluid velocity must vanish at the position of the wall (which also corresponds to a roll boundary). In contrast, for the convecting states studied here, this length is found to be very short. For example, for the data in Fig. 1, one would expect  $\tilde{\lambda} \gtrsim 5d \sim L$ , whereas we find no evidence of a healing length  $\lambda \gtrsim 0.3d$  in any of our experiments. We note that, in spite of the short healing length which is observed, the system selects oscillatory flow patterns which are composed of rolls parallel to the short side of the rectangular container, just as in the case of convection in pure fluids. In the traveling-wave case, this is because waves traveling parallel to the long dimension of the container can have zero net growth rate at Rayleigh numbers at which waves traveling parallel to the short dimension are damped by reflection losses.

The modulated states show directly that the wave number in the system is quantized and have allowed measurement of the group velocity and the coherence length  $\xi_0$ . These self-modulated states introduce a second frequency into the flow at onset. In general, we find that this frequency is incommensurate with  $\omega_0$ , and one might expect such a situation to exhibit chaotic trajectories of fluid elements at the onset of convection.<sup>11,12</sup> We would like to acknowledge extensive interactions with M. C. Cross, helpful discussions with D. Bensimon, P. C. Hohenberg, S. L. McCall, and B. Shraiman, and the invaluable technical assistance of A. Passner and H. L. Williams.

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<sup>8</sup>In Ref. 6, Cross also presents nonlinear solutions to a set of amplitude equations. These solutions, which treat a different, larger-amplitude regime than that discussed here, appear to be related to recent experiments by Heinrichs, Ahlers, and Cannell and Moses, Fineberg, and Steinberg (Ref. 4).

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 $^{12}$ The chaotic advection of fluid elements in binary fluid mixture at a *second* bifurcation to a two-frequency state has recently been considered by E. Knobloch (unpublished). The results presented here show that, in a finite system, the *first* bifurcation can be to a two-frequency state.