

**Chaotic advection by modulated traveling waves**

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The predicted transition from a traveling wave to a modulated traveling wave with increasing Rayleigh number was recently observed in an experiment on a water-ethanol mixture heated from below. Close to the codimension-two bifurcation the particle trajectories in such a modulated wave are predicted to be chaotic.

Recent experiments,<sup>1,2</sup> on water-ethanol mixtures heated from below have revealed a variety of novel behavior, some of which had been predicted theoretically.<sup>3,4</sup> Particular interest has centered around various nonlinear traveling wave forms, many of which are approximately two dimensional and spatially periodic. Of these, a regular translating pattern of rolls, hereafter called a traveling wave, may undergo a transition to a modulated wave in which the pattern is modulated in time with an independent frequency as it translates. Such a pattern, predicted theoretically,<sup>4</sup> has recently been observed experimentally<sup>2</sup> and in numerical simulations of thermosolutal convection.<sup>5</sup> The theory<sup>3,4</sup> assumes a fluid layer that is invariant under horizontal translations and reflections in vertical planes, and seeks spatially periodic solutions. These assumptions introduce a symmetry into the problem, which restricts the form of the amplitude equation describing the growth and equilibration of the instability. The key to much of the observed behavior is provided by the presence of a codimension-two bifurcation,<sup>6</sup> the Takens-Bogdanov bifurcation, for appropriate values  $R_c, S_c$  of the Rayleigh number  $R_T$  and the separation ratio  $S$ . Near this point the stream function  $\psi$  may be written in the form

$$\psi(x, z, t) = \text{Re}[\epsilon v e^{ikx} + O(\epsilon^3)] \sin(\pi z) , \quad (1)$$

where the complex amplitude  $v$  satisfies the equation

$$v'' = \mu v + \epsilon v v' + A |v|^2 v + \epsilon C(v\bar{v}' + \bar{v}v')v + \epsilon D |v|^2 v' + O(\epsilon^2) , \quad (2)$$

and the prime denotes differentiation with respect to a slow time  $t' = \epsilon p t$ ,  $p = k^2 + \pi^2$ . Here  $A, C, D$  are real constants that have to be computed from the basic equations,

$\mu, v$  are unfolding parameters proportional to linear combinations of  $R_T - R_c$ ,  $S - S_c$ , and  $0 < \epsilon \ll 1$ . With  $v = r \exp(i\phi)$  Eq. (2) turns into the Kepler problem

$$r'' + \frac{\partial V}{\partial r} = \epsilon(v + Mr^2)r' + O(\epsilon^2) , \quad (3a)$$

$$L' = \epsilon(v + Dr^2)L + O(\epsilon^2) , \quad (3b)$$

where  $V = \frac{1}{2}(L^2/r^2) - \frac{1}{2}\mu r^2 - \frac{1}{4}Ar^4$ ,  $L = r^2\phi'$ , and  $M = 2C + D$ . These equations admit four types of solutions, the trivial solution  $r = 0$ , here corresponding to pure conduction, standing waves (SW's) for which  $L = 0$ , traveling waves (TW's) for which  $r = r_0$  and  $\phi' = \omega_0$ , and modulated waves (MW's) for which  $r' \neq 0$  and  $L \neq 0$ . A complete classification of these solutions and their stability properties is given elsewhere.<sup>7</sup> For binary bulk mixtures the necessary coefficients  $A, C, D$ , and the unfolding parameters have been computed in the limit of large Prandtl number using idealized boundary conditions,<sup>8</sup> and the results used to predict the possibility of a bifurcation from TW's to MW's.<sup>4</sup> In this Rapid Communication we describe some characteristics of the modulated waves and show that the particle trajectories in such a flow may be chaotic.

A traveling wave, given by  $v + Dr_0^2 = 0$ ,  $\omega_0^2 = -\mu + (A/D)v$ , can lose stability at a secondary Hopf bifurcation to a modulated wave. In the case of interest,<sup>4</sup>  $A > 0$ ,  $M < 2D < 0$ , this bifurcation is supercritical<sup>7</sup> and occurs when

$$\mu = \left[ \frac{3M - 5D}{2M - 4D} \right] \frac{A}{D} v , \quad \mu < \frac{A}{D} v . \quad (4)$$

Near this bifurcation

$$\psi = \epsilon r_0 \cos(\omega_0 t' + kx) \sin(\pi z) + \frac{1}{2} \epsilon r_0 \eta \left[ \left( 1 - \frac{2}{\alpha} \right) \cos[(1 + \alpha)\omega_0 t' + kx + \theta] + \left( 1 + \frac{2}{\alpha} \right) \cos[(1 - \alpha)\omega_0 t' + kx - \theta] \right] \sin(\pi z) , \quad (5)$$

where  $\eta$  is the (small) amplitude of the oscillations about the TW,  $\omega = \alpha\omega_0$ , where  $\alpha = \sqrt{2D/C}$  is their frequency, and  $\theta$  is a phase. Thus  $\omega$  is small when  $|D|$  is small. Just before the bifurcation the spectrum of  $\psi$  at a fixed reference point  $x_0$  will contain only one prominent peak, at frequency  $\omega_0$  corresponding to a TW translating past  $x_0$  with

a phase velocity  $\omega_0/k$ . After the bifurcation, two sidebands with frequencies  $(1 \pm \alpha)\omega_0$  appear, one on each side of the original peak, and their power increases with distance beyond the bifurcation point. The power in the lower frequency sideband is larger by a factor  $[(\alpha + 2)/(\alpha - 2)]^2$ . In experiments in which the flow is

visualized by a shadowgraph technique, the same conclusion applies to the spectrum of the shadowgraph intensity. A transition with exactly these characteristics was recently observed by Heinrichs, Ahlers, and Cannell,<sup>2</sup> and may be identified with the theoretically predicted bifurcation (cf. Figs. 1 and 4 of Refs. 2 and 4, respectively). The experimental results can be used to estimate an effective value of the coefficient  $D$  which vanishes with idealized boundary conditions.<sup>3,4</sup> Taking the ratio of the observed

frequencies  $\omega_0$  and  $\alpha\omega_0$  to be  $\alpha=0.195$  and evaluating the coefficient  $C$  from Ref. 4 using  $k=\pi/\sqrt{2}$  and the value  $L=0.015$  for the Lewis number, we obtain  $D_{\text{eff}}=-0.047$ .<sup>9</sup>

The particle motion in the flow (5) is given by  $dx/dt=-\partial\psi/\partial z$ ,  $dz/dt=\partial\psi/\partial x$ . We define the scaled time  $\tau=\omega_0 t'$  and introduce the comoving variables  $\xi=\tau+kx$ ,  $\zeta=\pi z$ , yielding

$$\xi_\tau = 1 - R \cos \xi \cos \zeta - \frac{1}{2} \delta \left[ \left( 1 - \frac{2}{\alpha} \right) \cos(\xi + \alpha\tau + \theta) + \left( 1 + \frac{2}{\alpha} \right) \cos(\xi - \alpha\tau - \theta) \right] \cos \zeta + O(\epsilon^2, \epsilon\delta^2), \quad (6a)$$

$$\zeta_\tau = -R \sin \xi \sin \zeta - \frac{1}{2} \delta \left[ \left( 1 - \frac{2}{\alpha} \right) \sin(\xi + \alpha\tau + \theta) + \left( 1 + \frac{2}{\alpha} \right) \sin(\xi - \alpha\tau - \theta) \right] \sin \zeta + O(\epsilon^2, \epsilon\delta^2), \quad (6b)$$

where  $R=\pi k r_0/p\omega_0$  is the normalized TW amplitude and  $\delta=R\eta$ . These equations are derived from the Hamiltonian (the comoving stream function)

$$H_0 + \delta H_1 = -\zeta + R \cos \xi \sin \zeta + \frac{1}{2} \delta \left[ \left( 1 - \frac{2}{\alpha} \right) \cos(\xi + \alpha\tau + \theta) + \left( 1 + \frac{2}{\alpha} \right) \cos(\xi - \alpha\tau - \theta) \right] \sin \zeta. \quad (7)$$

When  $R > 1$  the Hamiltonian  $H_0$  (corresponding to a pure TW) contains heteroclinic orbits connecting pairs of saddle points that appear on  $\zeta=0, \pi$  as  $R$  increases through unity. The particles inside the heteroclinic orbit are carried with the wave. We call them "trapped." Those outside are not trapped and drift steadily backwards, relative to the wave. Both types of orbits are shown in Fig. 1.<sup>10</sup> The heteroclinic orbit  $\gamma$  is given by

$$\zeta_\tau = -\sqrt{R^2 \sin^2 \zeta - \zeta^2}, \quad R \cos \xi \sin \zeta = \zeta, \quad \tau > 0, \quad (8a)$$

with

$$\zeta(\tau) = \zeta(-\tau), \quad \xi(\tau) = -\xi(-\tau), \quad \tau < 0. \quad (8b)$$

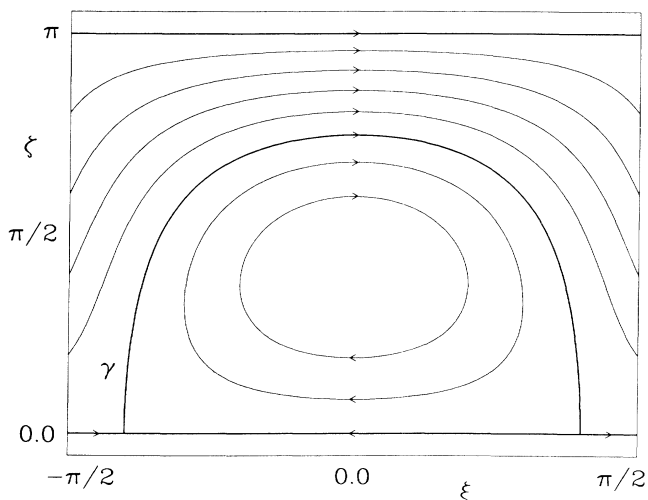


FIG. 1. The comoving stream lines in a pure traveling wave for  $R=3.266$ , showing the heteroclinic orbit  $\gamma$  separating regions of trapped and untrapped particles.

The situation described above occurs at the secondary Hopf bifurcation to MW where the amplitude

$$R = (\pi k/p) \sqrt{(1/A)[(2M-4D)/(M-D)]}.$$

Assuming that  $D$  is small and using the value of  $A$  calculated in Ref. 4 we find  $R=3.266$ . Beyond this bifurcation, the flow of  $H_0$  is perturbed by the time-periodic Hamiltonian  $H_1$ . As is well known, such a perturbation typically destroys the heteroclinic orbits resulting in

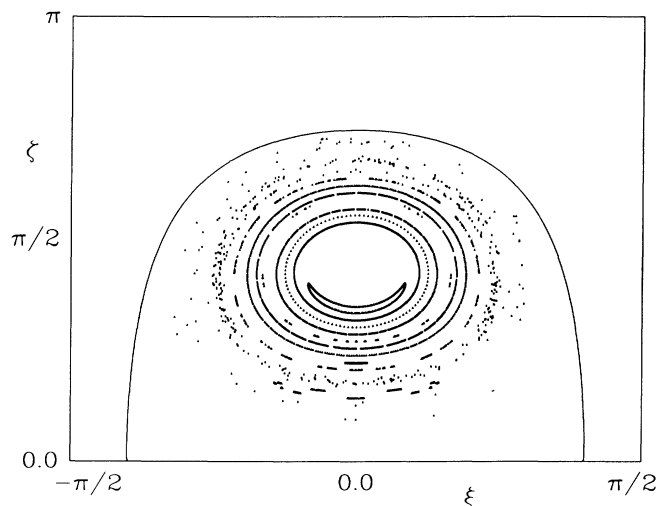


FIG. 2. Poincaré map for a modulated wave with  $R=3.266$ ,  $\alpha=0.195$ , and  $\delta=0.4$ . The position of the trajectory is plotted at times  $2\pi n/\alpha$ , where  $n$  is an integer, for several initial conditions along the line  $\xi=0$ . The solid line represents the heteroclinic orbit  $\gamma$  (Fig. 1) when  $\delta=0$ . Trajectories starting in the outermost chaotic region quickly escape the trapping region and are carried downstream by the flow.

chaotic motion. Below we calculate the Mel'nikov function<sup>11</sup>

$$M(\tau_0) \equiv \int_{-\infty}^{\infty} \{H_0(\xi, \zeta), H_1(\xi, \zeta, \tau + \tau_0)\} d\tau, \quad (9)$$

where  $\xi(\tau), \zeta(\tau)$  solve (8), and show that it has a countable number of isolated zeros. According to Mel'nikov's theorem<sup>11</sup> when this is the case the map

$$P: (\xi(\tau), \zeta(\tau)) \rightarrow \left[ \xi \left( \tau + \frac{2\pi}{\alpha} \right), \zeta \left( \tau + \frac{2\pi}{\alpha} \right) \right],$$

$$\{H_0, H_1\} = \sin\zeta \sin\xi \cos(\alpha\tau + \alpha\tau_0 + \theta) + \frac{2}{\alpha} \sin\zeta (R \cos\zeta - \cos\xi) \sin(\alpha\tau + \alpha\tau_0 + \theta). \quad (10)$$

Using the symmetry properties (8b) of  $\gamma$ , the Mel'nikov function becomes

$$M(\tau_0) = 2 \sin(\alpha\tau_0 + \theta) \int_0^{\infty} d\tau \sin\zeta \left[ -\sin\xi \sin(\alpha\tau) + \frac{2}{\alpha} (R \cos\zeta - \cos\xi) \cos(\alpha\tau) \right]. \quad (11)$$

With the help of (8a) this expression reduces to the simple form

$$M(\tau_0) = -\frac{6\alpha}{R} \sin(\alpha\tau_0 + \theta) \int_0^{\infty} \zeta \cos(\alpha\tau) d\tau. \quad (12)$$

The integral on the right side cannot be evaluated in closed form, but can be shown to be positive for the parameter values of interest. Thus, as a function of its argument,  $M(\tau_0)$  oscillates indefinitely and the conclusion follows. In Fig. 2 we show the map  $P$  obtained by numerical integration of the system (6) with  $\theta=0$  for  $R=3.266$ ,  $\alpha=0.195$ , and  $\delta=0.4$ , starting with several initial conditions along the line  $\xi=0$ . As  $\delta$  increases the invariant tori near the heteroclinic orbit  $\gamma$  are progressively broken. Ini-

generated by Eqs. (6), contains a Smale "horseshoe."<sup>11</sup> Thus, near the bifurcation to MW, there are a countable number of periodic orbits of arbitrarily high period and an uncountable number of nonperiodic orbits. Although all of these orbits are nonstable, we expect to see at least long chaotic transients. Moreover, it is possible that for larger values of the Rayleigh number the chaos becomes stable.

The Poisson bracket  $\{H_0, H_1\}$  simplifies considerably, yielding

tial conditions starting on such tori are eventually mapped outside the trapping region and are rapidly carried downstream (in the comoving frame). A detailed report on the properties of the resulting flow will be published elsewhere.<sup>12</sup>

In this Rapid Communication we have proved rigorously the existence of chaotic particle trajectories in a type of simple ordered flow that has recently been observed experimentally. We hope that this result will stimulate experimental studies in this direction.

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<sup>7</sup>G. Dangelmayr and E. Knobloch, Philos. Trans. R. Soc. London (to be published).  
<sup>8</sup>These coefficients have not been calculated with the boundary conditions appropriate for the experiments.  
<sup>9</sup>Although the experiments suggest that  $D > 0$ , the TW branch rapidly turns round and becomes supercritical, i.e., it behaves as if  $D < 0$ . This is the effect that is described by  $D_{\text{eff}}$ . When  $D > 0$  and small a negative fifth-order term must be added to the amplitude equation (1), but the conclusions of Ref. 7 remain unchanged.  
<sup>10</sup>With no-slip boundary conditions the saddle points cannot lie on the boundary, but the qualitative features associated with the bifurcation to MW are expected to persist.  
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