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TITLE MASS TRANSPORT IN PROPAGATING PATTERNS OF CONVECTION

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Mass Transport in Propagating Patterns of Convection

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I. Introduction

Recent studies of propagating waves in an oscillatory convection of binary mixtures¹⁻⁷ arise questions about transport properties of this flow. Optical visualization of a field of refraction index due to a shadowgraph technique gives information on the temperature and concentration fields. However, experimental observation of rolls propagating along the cell as travelling waves (TW) does not necessarily imply that mass is transferred hydrodynamically by the convective motion along the cell. One of the possibilities discussed, e.g., is that TW observed is only a phase propagation. The traditional examples of such situations come from the domain of linear, superposition-oriented physics. Acoustic waves transfer momentum and energy, but do not cause the mass to make excursions from their equilibrium point that are larger than the oscillation amplitude. In the case of nonlinear physics we were aware that small amplitude surface waves cause only small oscillatory motion round the equilibrium point, while larger amplitudes can cause the mass to start moving in the direction of the TW.

Our first suggestion^{2,3} about possible transfer of mass by TW was based on the fact that the fields of velocity, concentration and temperature are all coupled through the Navier Stokes equation. So our initial guess about the form of the flow was aimed to reconcile the possibility of mass transport due to TW with the necessary mass conservation and the restrictions that follow from the finite geometry of the cell^{2,3}. On the basis of

the two-dimensional parabolic shape of the propagating roll pattern in the vicinity of the lateral walls, we tended toward a picture of mean drift flow, rather than global circulation with the back-flow returning along the cell sides. We now know that the models suggested for the global mass flow were naive and incorrect. However, they have shed light on how convection takes care of itself, and of some different basic possibilities for the form that a flow in general can take.

Since the oscillatory convection in binary mixtures at the onset has been found to consist of different types of flow patterns, we were also intrigued by the possibility that the different states and regimes would display different forms of mass transport. For example, it is hard to imagine mass moving at the finite velocity of the linear counter-propagating waves when the amplitude of the velocity goes to zero, as is the case for small ϵ ($\epsilon = \frac{\Delta T}{\Delta T_c} - 1$).

Can we then use the differences in mass transport for understanding the underlying mechanisms that determine the different oscillatory states? Are a global mass flow and back-flow, compensating for mass conservation, described by the amplitude equation approach? To resolve these questions we undertook a study of the transport properties of the oscillatory states trying to identify the motion of mass and the mechanism that ensured mass conservation in a finite cell⁸.

By using a technique of molecularly mixed photochromic component we ensure that these impurities do not modify the velocity field. This condition requires for passive markers transport. It is a direct way to follow the fluid particle trajectories under the action of the fluid velocity field.

Problem of mixing and stirring⁹ is closely related to the question raised in this paper. In order to understand how mixing occurs it is necessary to know fluid particle trajectories. It is well known that particle trajectories coincide with streamlines only in a stationary flow. In time dependent flow this coincidence does not occur, and the streamlines define the velocity field of different particles at a certain moment, while the trajec

tories define the particle velocities at successive moments. In stationary flows and flows with non-zero mean velocity like TW where flow is stationary in a moving frame, the velocity field as well as the particle trajectories are always well ordered. On the other hand, in time-dependent flows the fluid particle motion may be extremely complicated. For example, while the motion of three point vortices in an unbounded domain is integrable, particle motion in this flow can be chaotic. We consider in this paper in detail the simplest case of two-dimensional flow when both fluid dynamics and particle motion are integrable. However, even in this simple case the Lagrangian presentation is very useful to describe the global structure of the particle trajectories and to elucidate the problem of mixing and transport in a flow of propagating patterns. Our experiments revealed that depending on the ratio of velocity amplitude inside the roll to propagation velocity part of the fluid is trapped and propagates with TW while the rest in accordance with a mass conservation propagates in average in the opposite direction. This global circulation of mass (or mass transport) along the regular trajectories without mixing occurs in a cell infinite in a horizontal direction. In a finite geometry cell this simple global mass transport is complicated by reflection at lateral boundaries and by corresponding back flow.

This simple scenario can be observed only in a case when existing TW propagates in one direction. However, in both nonlinear TW states - spatially modulated TW (confined TW) and spatially and temporally modulated TW (blinking TW) - left and right propagating waves together with waves reflecting from the lateral boundaries exist simultaneously ¹⁰⁻¹². It means that in a moving frame for one of TW one gets time-dependent behaviour from the rest of TW. In case of confined TW an amplitude ratio of reflecting and incident TW is, probably, so small that resolution of our visualization method is not enough to detect chaotic excursions from regular behaviour on time scale of the experiment. On the other hand, we do observe irregular behaviour in a transport in the case of blinking TW where temporal modulation of TW is significant. We did not

analyse this irregularity due to lack of a model which can transform the chaotic trajectories from a moving to a laboratory frame in order to compare them with the experimental results. Besides the transformation this phenomenon, probably, should be similar to other recently studied examples of chaotic mixing of fluids in two-dimensional time-periodic flows¹³⁻¹⁴. It would be definitely interesting to study the structures produced by this time-dependent flow. It provides useful insight into the mechanism of mixing and transport of fluid flows. And secondly, it provides a visual analogue for chaos in area-preserving maps in terms of directly observable physical phenomena.

II. Review of experimental and theoretical results

On the experimental side very few studies have been conducted on topology of fluid particle trajectories and transport in convective flows. This is especially true for propagative cellular flows, like TW patterns, since these occur near threshold only in few known systems. Work that is close to ours in its experimental approach, is that of Croquette et. al.¹⁵. In this work convection of methanol in a cylindrical cell with aspect ratio $\Gamma = 20$ was studied by the use of a photochromic technique. A strip of ultra-violet radiation was applied to the cell, causing coloration of the dye. The subsequent motion of the strip of dye was monitored visually, together with the shadowgraph signal. In this geometry the umbilical has a tendency to move off center. It was shown that the distorted pattern is accompanied by a large scale flow that does not exist for the pattern with the umbilical in the center. This motion is representative of one type of large scale motion. This is a secondary flow, and for this type of flow to occur there must be curvature of the basic main flow roll pattern.

Another work on convection has been carried out by Solomon and Gollub¹⁶. This work studied mass transport of an impurity in stationary convection in an undistorted pattern that is due to interplay between advection of particles along the streamlines within convection rolls and their diffusion between rolls. They injected an impurity, and observed its propagation along the convective pattern. The motion within the rolls is de

terminated by the convection amplitude, and is a fast process. The motion between rolls occurs through diffusion, and depends on the boundary layer that separates two adjacent rolls and on the diffusion constant of the impurity. This flow of the impurity can be characterized by an effective diffusion constant, that is larger than the original one by a factor proportional to the square root of the Peclet number $p = \frac{vl}{D}$, with v the velocity, l the length scale, and D the diffusion coefficient of the impurity.

Second paper by the same authors¹³ is devoted to study mixing and transport in a time-dependent convective flow. In the time-periodic regime, the transport is dominated by advection of tracer particles across roll boundaries. It was shown that in this regime transport is dramatically enhanced due to chaotic advection caused by lateral oscillations of the roll boundaries. The latter conclusion is true for many types of time-periodic flows. Experimental¹⁴ and theoretical studies^{9,17} of two-dimensional incompressible time-periodic flows show that dramatical enhancement of mixing and transport occurs due to crossing of stable and unstable manifolds of hyperbolic points. As the result of it a fluid element becomes stretched and folded so that points originally close become widely separated and others are drawn close together. This process causes mixing of the fluid similar to the chaotic behaviour in dynamical systems.

Mass transport has been studied by Tam and Swinney¹⁸ in turbulent Couette-Taylor flow, again by the injection of an impurity and monitoring its transport in the flow pattern. They also find that the transport mechanism can be characterized as a diffusion process, and measure an effective diffusion constant that is orders of magnitude larger than D .

The importance of the Lagrangian representation, i.e. studying the equations of motion as opposed to the Eulerian description, where the equations for the fields are studied, has been emphasized in the work of Aref¹⁹. The trajectories of particles can be very complex, even if the equations for the field are simple. This can lead to chaotic motion of the particles themselves. Rich behaviour can occur for 3D flows, as was shown

by Feingold et. al.²⁰ who used the analogue that exists between Hamiltonian systems and flow streamlines (where mass is the conserved quantity) to characterize the different flows possible. In cases of extreme complexity the motion of a “single” particle that can be monitored is very valuable, since any finite volume will disintegrate within a short time. These abstract particles are called passive scalars, and are used as a tool for following the flow lines.

An example of the problems involved in using the Eulerian description can be found in the work of Linz et. al.²¹. These authors ask if mass is transferred by looking at the behaviour of the fields of convection in a few mode truncation of the equations for convection in a binary mixture. Besides wrong conclusions that mass transportation due to global circulation in TW is negligible and undetectable experimentally unlike transport of concentration, which is the result of a trivial mistake, the authors of Ref.21 used the spatial averaging based on Eulerian description. This averaging procedure, perfectly legitimate in a stationary flow, occurs to be incorrect for time-dependent flow or flow with non-zero mean velocity. Here Lagrangian vs. Eulerian description becomes crucial. In a stationary convection with rigid boundaries horizontal component of velocity is symmetric in a vertical plane. It leads due to mass conservation to symmetric counter-flows in any vertical plane in the cell. However, it is not the case for TW convection contrary to the result of Ref.21. From Lagrangian presentation one can conclude that two types of particle trajectories take place: open and closed. Part of a fluid volume trapped inside separatrix between two types of trajectories depends on the ratio of velocity amplitude and TW propagation velocity. This parameter, naturally, does not appear in Eulerian description at all. Then a velocity distribution in the vertical plane is asymmetric and depends on this parameter. It is obvious that even a total mass flux through any vertical plane is zero, counter-flow is nonsymmetric in the vertical plane. Thus, only Lagrangian approach gives a possibility to describe correctly a global mass circulation due to TW.

The interesting point also is that, large scale flow can exist even though no mean

flow exists. In a finite container this means that the convection "takes care" of itself, and we do not have to introduce the back-flow by hand, as it is implicit already in the Lagrangian description. In this case the large scale flow is circulatory, taking mass (in an infinite system) from $-\infty$ to $+\infty$, but also returning an equal amount from $+\infty$ to $-\infty$.

The most relevant work is one that dealt with a one dimensional equation of motion. Knobloch and Weiss²² have shown that a TW in 1D causes a particle to drift from its original position. This model showed a continuous growth in drift velocity of the particle as a function of the TW amplitude, saturating when it reaches the TW velocity.

The same authors used²³ the Lagrangian description to show that for modulated TW chaotic trajectories of particles can occur. Although they deal with a more complicated situation than ours, we have used the Hamiltonian description which they introduce.

III. Experimental Methods

The technique we chose for monitoring particle motion is that of introducing a strip of dye in the flow, measuring the attenuation of a parallel beam of light as it is passed through the sample. We did this by admixing a small amount of photochromic spiropyran ($4 \cdot 10^{-4}$ by weight) that is colorless until irradiated by UV light. It then attains a vivid dark color that remains until thermal processes cause the color to fade. This cycle can be repeated many times with only minimal optical degradation. Our choice of spiropyran was guided by the need to minimize irradiation time while maximizing the fading time. A characteristic passage time for a roll to propagate along the TW pattern (half the cell length) is 225 sec. Thus the fading time should be a few times larger. The irradiation time should be kept down to a few seconds, which we shall show is the characteristic time scale for spreading of the dye within the roll by the rotational convective motion. The spiropyran has been supplied by Dr. R.C. Bertelson, and is 5-bromo-8-methoxy-6-nitro-1',3',3'-trimethylspiro(2H-1-benzopyran-2,2'-indoline), sold

as "Chromadye 2" by Chroma Chemicals of Dayton, OH. The absorption spectrum at this spiropyran is shown in fig. 1, and is characterized by a broad peak around 572 nm. Thus by using a green filter we were sensitive to the "shadow" of the dye.

This spiropyran has best sensitivity to UV when admixed with aromatic hydrocarbons, while mixing with alcohols decreases its fading rate. With this in mind we chose a benzene - methanol mixture. The best data for the physical properties of a benzene - methanol mixture at 95% by weight benzene and at 30°C found in the literature are $\lambda = 1.48 \frac{mW}{cmK}$, $\nu = 6.7 \cdot 10^{-3} \frac{cm^2}{sec}$, $D = 2.3 \cdot 10^{-5} \frac{cm^2}{sec}$, $\kappa = 0.92 \cdot 10^{-3} \frac{cm^2}{sec}$, $\rho = 0.863 \frac{g}{cm^3}$, $\beta_1 = -\frac{1}{\rho} \frac{\partial \rho}{\partial T} = 1.22 \cdot 10^{-3} K^{-1}$, $\beta_2 = \frac{1}{\rho} \frac{\partial \rho}{\partial C} = 0.103$, $k_T = -0.16$ and $\Delta T_c = 2.86 K$ for $d = 0.141 cm$ ($\Delta t_{exp} = 2.85 K$).

The preparation of the sample consisted of mixing typically $4 \cdot 10^{-4}$ by weight of the spiropyran with 50 mg of benzene. This was filtered through filter paper, and then weighed again before methanol was added in the right amount. This ensured that all losses due to evaporation during the filtering process could be accounted for and did not reduce the final concentration of benzene in the mixture. In the filtering process any large particles of undissolved photochromic material were removed. Although we could not check this, it is reasonable that these losses more than compensated the loss of benzene due to evaporation, so that the concentration of photochromic material in the fluid did not increase in the process.

Our experimental setup is shown in fig. 2, clarifying some of the experimental difficulties. The UV light source we used is a commercial mercury lamp with a fluid filled light guide cord, manufactured by the Dr. Houle Co. of Martinsried, Germany. This light source can output 800 mW, with multiple spectra both in the UVA and UVB. The optical components we used were of quartz or sapphire where possible, so that losses through the optical setup are negligible. The main source of losses is in the 1mm pin hole used, and in absorption in the ~ 50 mm of cooling water that lie in the optical path. We estimate the amount of UV light to finally reach the cell to be on the order

of 10 mW. Even such an amount could potentially change the convection field drastically. Therefore we checked that the absorption of the UV radiation in the mixture was not enough over the radiation times we used to affect the convection at all. We did this by looking at the amplitude and form of the TW before irradiation, and comparing to its form immediately after.

The setup for the UV radiation (shown in fig. 2) is such that the UV light source is fixed, and the mirror and cylindrical lens are movable. The cylindrical lens can also be rotated to produce a strip in any direction we choose in the cell. During irradiation the mirror protects the camera from the intense radiation and reflections. Immediately following the end of the irradiation time the mirror and lens are removed, to clear the view for the camera. The ability to follow irradiation by immediate monitoring sets a limit on the speed of the processes we could study. For irradiation times of 3 seconds, for example, the additional 1-2 seconds needed till the first measurement of the camera can cause some information to be lost. Irradiation times were between 2 and 10 seconds, with 5-7 seconds being typical.

The need to monitor both the shadowgraph and the attenuation caused by the dye are contradictory. The shadowgraph signal is strong away from the cell, and is largest at the focal plane of the lenslets produced by the convection rolls. In our case this distance was on the order of 60cm. To view the motion of the dye, we must eliminate the effect of the shadowgraph, so we had to focus directly onto the bottom of the cell. In that way the shadowgraph signal does not interfere, and the parallel light beam can be used for both measurements. In order to obtain a large image of the 18mm cell, we had to bring the camera as close as we could to the cell itself. Thus the compromise was between the size of the image when we followed the dye (spatial resolution) and the resolution in intensity when we monitored the shadowgraph (grey scale resolution). Although this problem could in theory be solved by use of an appropriate zoom lens, we did not find an adequate solution in practice.

To obtain the maximum from the image we had we used averaging in time of the signal. Our camera is an old model made by Hamamatsu, very reliable in response and linearity, but limited to transferring 10 lines per second (compared to the possibility of 25 per second plus the possibility of spatial averaging when using a video frame grabber). We used assembly language routines to minimize computer time while averaging lines. Typical averaging numbers were between 10 and 50, and typical time intervals between measurements were between 3 and 12 seconds.

At a mean temperature of 30°C we can change the sign of ψ by varying a concentration. This resolves a controversy that existed about the Soret coefficient for this mixture²⁴. The controversy stems from the fact that the region of oscillatory convection is very small, so that techniques for determining the sign of ψ by looking for oscillatory convection can be misleading. At benzene concentrations of 10% and 50% by weight square stationary patterns typical of positive ψ were observed. At 95% by weight benzene an inverse bifurcation leads to finite amplitude TW via CPW, and ψ is negative. The TW branch exists in a narrow range of $\sim 20\text{mK}$ (not including hysteretic regime) for a cell of height 1.47 mm and aspect ratio 1:4:12.25. Our estimates based on data from the literature give $\psi = -0.045$. We can compare this with the value $\psi = -0.044$ deduced from the experimental value for the neutral frequency, $\omega_0 = 4.4$ using the dependence²⁵

$$\omega_0 = \frac{\kappa}{d^2} \left(\frac{3\pi^2}{2} a \right) \sqrt{\frac{-\psi}{1 + \psi + P^{-1}}}$$

with $a=1.43$. We have observed in this mixture confined TW states^{2,4-7} in which convection exists only in part of the cell. For small enough step size from conduction, the first state was a temporally and spatially modulated one (a "blinking state")²⁶. The confined state always appeared high on the TW branch. The spatial pattern of the blinking state consists of alternation between left and right TW, both being modulated in space, with periods of coexistence in mid-cycle. The fact that the TW state that fills the cell was not observed may be due to the narrow range of the TW branch, or to the

small thermal conductivity of the mixture when compared to the cell boundaries²⁷.

Thus, the experimental data we present on mass transport in the nonlinear TW state is that of the confined TW and the blinking state patterns.

What we actually measure is not concentration but attenuation, and we must take this into account. The transmitted intensity is given by

$$I(x) = I_0 \int e^{-\frac{C(z)}{x_0}} dz$$

where $I(x)$ is the measured intensity at point x (x is the direction along the cell), I_0 the initial intensity, $C(z)$ the concentration of excited photochromic dye at point z along the height of the cell (y is the direction across the cell), and x_0 the attenuation length that characterizes the absorption of light by the dye (we assume that the green filter allows only a small bandwidth to pass). The integral should be taken along the full optical path, i.e. through the cell depth and back. If we want to use this relation in a quantitative way we must make some assumptions on the form of $C(z)$. This is usually done by assuming a uniform distribution of the dye along the cell^{15,16,18}. As we shall see presently this is not so in our case. However, we note that the actual attenuation is small enough so that plotting the attenuation or its logarithm is essentially the same.

IV. Experimental Results

Conduction

In fig. 3 we show the result of irradiating a single line across the cell, at a distance from the cell wall which is typical of the experiments on the convective state. We see that the initial shape is close to a gaussian one. This is due to the optical setup, and is convenient for our analysis. The FWHM of the gaussian is about 1.2mm, slightly less than the size of a convection roll, 1.5mm. The evolution in time is that of a decay in amplitude and widening of the gaussian. We can model this by $C(x, t) = A(t) \cdot \text{Gaussian}(x, t)$, where $A(t) = e^{-\frac{t}{\tau}}$:

$$C(x, t) = \frac{C_0}{2\sqrt{\pi Dt}} e^{-\frac{t}{\tau}} e^{-\frac{(x-x_1)^2}{4Dt}}$$

with t the time, τ the characteristic decay time for the thermal fading of the photochromic, and x_1 the center of the gaussian. We have 2 parameters to fit every line of fig. 3, $A(t)$ and $\sigma^2(= 2Dt)$ of the gaussian. Fig. 4 shows a representative measurement. The dots are data, and the curve is a fit using the above function. We see that the model gives a good fit. This quality of the fit is characteristic of all these measurements.

In figs. 5 and 6 we show the results of fitting for D and τ . Fig. 5 shows the decay prefactor as a function of time. The dots are experimental points, and the curve is a best fit to an exponential behaviour. We see a relatively long transient, after which the data fits an exponential decay with time constant $\tau = 490 \pm 70$ seconds. The fact that our initial profile was actually gaussian eliminates a common cause for transients. The transient we see may be due to processes that we cannot view, i.e. in the unseen direction, the depth of the cell. The time scale of the transient is about 200 seconds. This is approximately the time scale for diffusion of the dye on a length on the order of the cell depth. We conjecture that this transient is largely due to a diffusion process in which the dye diffuses downwards, into the depth of the cell, until the profile in the z direction homogenizes. During this period the decay of concentration is less than expected by the simple model, since the diffusion in the z direction comes instead of diffusion in the x direction. The reason for an inhomogeneity in the z direction is that our irradiation enters the sample from the top. As it penetrates the fluid, it excites the photochromic material, causing opaqueness. This opaqueness causes the deeper layers to be less effectively excited, with the final outcome that the concentration of dye decreases with the depth. We shall present evidence for the depth dependence of the dye concentration below. Although methods of treatment for such a self induced opaqueness exists, we shall not make an attempt at characterizing the z dependence of the concentration. Where possible (i.e. no quantitative data is taken from the amplitudes), we make use of the signal from the intensity itself, rather than make claims about the concentration.

Fig. 6 shows the fit for D , plotting the variance σ^2 of the gaussian versus $2t$. Again, the dots are experimental points, while the curve is a linear fit, yielding $D = (1.0 \pm 0.2) \cdot 10^{-5} \frac{cm^2}{sec}$. The transient is not as noticeable here, but is still apparent. Here the effect is to reduce the diffusion in the x direction until the profile in the z direction is homogenized. The reduced sensitivity to the transient may be due to the linear behaviour. At large time scales the width of the gaussian along with small amplitude cause scatter round the linear fit.

Stationary convection

The state following the oscillatory branch is one of stationary convection, similar to that in pure fluids. In this section we present results on mass transport in this regime. For figs. 7-9 $\epsilon = 0.02$ still quite close to onset. The results presented are mainly qualitative, aimed at comparison with the following section on mass transport in the oscillatory regime. We can, however, obtain some interesting insight as to what can be observed with the photochromic technique.

Fig. 7 shows the result of irradiating a strip across the cell at the typical $\frac{1}{3}$ of the cell length. The irradiation time is 5 seconds, and we see that the initial gaussian shape is distorted already by the time of our first measurement. The gaussian spans two rolls, with a tendency to the one at the right. Within 2-3 measurement times (time between measurements is 5 seconds) the dye has homogenized over the size of the roll. This indicates a velocity $v \sim 200 \frac{\mu m}{sec}$, assuming half a revolution of a particle on the boundary of the roll for homogenizing the dye over the roll in the x direction. The two rolls that are initially irradiated settle to a constant amplitude of dye, that decays at the rate measured above. We can just barely discern an enhancement of the two adjacent rolls on either side, coming purely from an effective diffusion process of the type measured in ref.16. Although we can see the effect here, obviously the decay rate of the photochromic spiropyran we chose does not permit us to make any measurement of the effective diffusion coefficient.

To follow the process of homogenization within the rolls by the convection we do two things. First, go down to 3 seconds exposure and measurement intervals. Second, we irradiate a strip at the side of the cell, where the boundary of the wall has a damping effect on the amplitude of the convection. We see in fig. 8 for such a test that the amplitudes of the roll next to the wall takes about 2-2.25 times longer to homogenize than does the adjacent roll (whose time is similar to that in fig. 7). The estimate for the velocity of the roll near the boundary is $v = 90 \frac{\mu m}{sec}$.

Finally, we show in fig. 9 the result of irradiating a strip along the cell, i.e. along the x direction. The signal, although homogenous at the onset, is seen to be highly modulated, at the characteristic wavelength of the convection (2 rolls). We understand this as stemming from an initial inhomogeneity in the z direction. After a quarter of a cycle the high concentration from the top will go in each roll to one side, while the bottom will rotate to the opposite side. The rolls are paired so that this effect is reinforced, each pair enhancing or decreasing together the intensity of the boundary between them. We can see that a quarter of a cycle occurs faster than our first measurement, giving again an estimate for $v = 200 - 250 \frac{\mu m}{sec}$, with an uncertainty due to time loss between irradiation and the first measurement, as explained above. This result is the one we rely on in our analysis of the transients in the decay of the gaussian in the section above.

Another observation, not shown here, was that although secondary motion in the y direction may be expected (due to 3D effects in the flow), these motions were not apparent within our resolution. An initial strip along the cell (x direction) showed no sign of spreading in the y direction faster than would be expected from a diffusion process.

Oscillatory Convection

We show here the effect of the existence of TW on mass transport. We do this for the confined TW state, then briefly for the blinking state and CPW.

Confined states: Figs. 10 and 11 show the result of irradiating a strip across the cell, at $\frac{1}{4}$ the length. In fig. 10 two rolls are affected, while in fig. 11 the initial gaussian en

closed only one roll. Mass is definitely transferred in the TW direction, at the TW velocity, in a periodic structure with the size of the convection rolls. The existence of a flow in the opposite direction is evident. This is the backflow needed to maintain mass conservation. The motion of the part that moves with the TW is seen to be rather straightforward along the 1D line depicted. The roll detaches from the patch of dye, and propagates smoothly and continuously till the edge of the cell. The task of extracting the velocity of the TW from these figures is complicated by the existence of changes in wavenumber (see fig. 13a below). Since the frequency is constant over the cell, the velocity varies over the TW pattern. We can take the signal from the shadowgraph to obtain the frequency, multiplying by the critical wavelength to obtain an estimate. This is in practice a lower bound on the velocity.

To obtain further information on the backflow we irradiate an initial strip located just next to the short wall of the cell.

This is shown in fig. 12. The backflow appears in a discontinuous fashion, with strips appearing at distances corresponding to multiples of the roll size. This discreteness can only be explained as a local concentration of dye at fixed points on the roll structure along the cell. The motion can best be described as follows: a peak of intensity appears at multiple distances of the roll size, enhances and starts to drift back in the direction of and at the velocity of the TW itself. Thus we see that the back flow is restricted by the frame of the traveling rolls. The small number of convection rolls limit the accuracy in determining the backflow front velocity in fig 12. Our estimate is $V_b = 42 \pm 10 \frac{\mu m}{sec}$. We were not able to estimate the variations in V_b over the cell.

In fig. 13 we show the full 2D behavior, using a series of images of the dye to follow the behaviour of initial strips of dye, placed in positions analogous to those of figs. 10-12. Exposure time is 7 seconds, and time between measurements is 7 seconds. Fig. 13a displays the shadowgraph image of the convective pattern. We see three dimensional effects, a large deviation from the straight rolls pattern. The rolls are much shorter,

near the edge of the cell, occupying as little as one half of the width of the cell. While a boundary layer is expected to exist, the measured size of the layer exceeds any theoretical estimate.

In fig. 13 b) and c) we show the spreading sequence for the two different initial positions of the dye. In the TW direction the form of the dye front is parabolic at the beginning, then changes to the form of the roll structure shown in Fig. 13a, filling about half the cell width. The backflow also has a parabolic profile along the width of the cell, and seems to maintain it.

In fig. 14 we show the analogous situation to that of fig. 9, irradiating along the cell. In this case we see that the nonuniformity caused by an inhomogeneous profile in the z direction is also found here. However, the mixing mechanism that existed in the stationary regime does not work here, and the modulation of the profile is maintained in time. We shall see below that this can be linked to the behaviour of the boundaries of the rolls.

Temporally and spatially modulated TW (blinking state): In fig. 15 we show the mass transport properties of the time dependent "blinking state"²⁶, in which left and right TW periodically alternate. This state is found at very low ϵ , for very small jumps from conduction, as a precursor to the confined states. The two opposite TW are modulated on a slow time scale, but with a phase difference between them. At first one TW dominates, then the other grows and the first diminishes, till the second dominates, and so on. The spatial profile is similar to that of the linear CPW, where the waves grow as they propagate, but with nonlinear saturation apparent. The time spectrum of the signal at a single spot in the cell reveals a transition from a quasiperiodic, ordered spectrum at low ϵ , to a many peaked disordered one.

The blinking state has been independently discovered in numerical simulations of 2 coupled Ginzburg-Landau equations, as have the confined states it precedes¹⁰. The selection mechanism that chooses between the states depends on the ratio of the TW

propagation velocity s to the local growth rate τ^{-1} on characteristic local scale where s is the group velocity and $\xi = \xi_0 \epsilon^{-1/2}$ and $\tau = \tau_0 \epsilon^{-1}$ are the spatial and temporal characteristic scales. This selection is related to the transition from convected to absolute instability conditions, which occurs at a defined value of this ratio.

Fig 15a shows a right TW, and we see that at the opposite side no mass motion is apparent. We note that the division of left and right TW occurs at the center of the cell, emphasizing that the nonlinear profile has a rapid variation in midcell. Fig. 15b shows a time in which both TW have comparable amplitude, and both are seen to transfer mass (this coexistence is for a brief period of time). We have also measured brief occurrences at which both TW were at a minimum, and no mass was transferred in either direction. Finally, in fig. 15c the left TW dominates, and in the right side of the cell no mass is transferred. It is impressive that the large scale motion can be initiated and stopped within the short time scales that separate periods of domination (order of $0.1 \tau_h$ horizontal diffusion time)

We would like to point out here that spatio temporal plots of dye intensity profiles in the blinking state exhibit less regular behaviour than the corresponding plots (Fig.10-12) for confined TW. A possible reason for this disorder is a temporal modulation of TW. As we already mentioned in the Introduction the time-periodic perturbations cause the chaotic advection across the separatrix between different topology trajectories.

It is not clear what kind of behaviour should be observed then in a laboratory frame in which the data are presented. It is also possible that much faster mixing (homogenization) observed on Fig.15 than on Figs. 10-12 is the result of the chaotic advection. This point needs further experimental and theoretical studies.

Linear CPW: The stable nonlinear TW states are reached via a transient of linear CPW oscillation. Upon close investigation of this regime we found that in the linear CPW state no mass transfer is evident within our resolution. Fig. 16 shows the result of such an experiment. We first raise the power input q from 0.995 times the critical value q_c to

$q = 1.0015q_c$ at $t = 0$. A linear CPW transient is discernible after about 40 min. Fig. 16a shows the shadowgraph signal beginning at $t = 52$ min, with two counterpropagating waves filling the cell. Their velocity is $V_p = 84 \frac{\mu m}{sec}$. We then irradiated two strips of UV light at each side of the cell for a period of 6 sec, and monitored how they spread. No mass motion is apparent until the nonlinear TW state is reached, and this is shown in Fig. 16b. The first lines (beginning at $t = 65$ min) show no signs of mass propagation. Towards the top of Fig. 16b, at $t \approx 66.5$ min, the right strip of dye undergoes a breakdown, and mass propagation begins. The left strip remains untouched, slowly fading, as a nonlinear TW state is reached, with convection only in the right hand side of the cell. This is the stage where the linear CPW develops into nonlinear TW. Thus the transition from linear CPW to nonlinear TW is tied to a transition from no mass propagation to large scale flow.

We have checked that the amplitude of the CPW before the transition is not markedly different from that of the TW after the transition, (while a jump in the TW frequency does occur). This indicates to our mind either a critical transition, or that the two states are governed by different mechanisms.

V. Theoretical Model

To model this behaviour of the convective flow in confined TW state we have used a simple form of TW for the amplitude of convection, that assumes the existence of only one mode, and is a reasonable approximation for the rigid boundaries of the experimental cell:

$$V_x = V_0 \frac{k}{\pi} \cos(\frac{k}{d}x - \omega t) \sin^2 \frac{\pi}{d} z \quad (1)$$

$$V_z = 2V_0 \sin(\frac{k}{d}x - \omega t) \sin \frac{\pi}{d} z \cos \frac{\pi}{d} z \quad (2)$$

with $V_0 = 1$, $k = \pi$, $d = 1$, x the horizontal and z the vertical directions. The TW frequency ω is a parameter we vary.

There are two significant assumptions made for this model. we ignore the diffusion

effects and consider that only one TW existing in the confined state. First assumption is based on the fact that the Peclet number p is typically much larger than 1 (of order 100), so that diffusive processes are really negligible in comparison with the flow effects. Second assumption is less obvious since as known¹⁰⁻¹² a spatial modulation of TW is the result of interaction of left and right-propagating waves and waves reflected from lateral boundaries. Probably, in the case considered the amplitude ratio of left and right-propagating waves in a right part of the cell is small enough that we did not observe effects due to modulation by left-propagating wave. These equations were integrated in time using a standard Runge-Kutta algorithm (Fig. 17a).

The flow is seen to separate into two distinct regions. One part moves with the TW while the second flows on the boundaries of the roll, with an average motion in the direction opposite to the TW propagation. This second part is the backflow. We can now associate the discrete peaks appearing in the backflow in Figs. 10-12 with the arrival of the backflow front at a boundary between rolls, where optical absorption is large due to our viewing the cell from the top. This is exemplified in Fig. 17b which depicts the equivalent of our measuring technique, integrating the dyed particle density along z .

If we vary our parameter ω , we see that the flow changes drastically. Fig 18a-b show the effect of increasing the TW velocity. The volume that move with the TW decreases, and the backflow velocity falls below the TW velocity. Further decrease of the frequency, such that the TW moves at a velocity faster than V_0 , is shown in figs. 19a-b. At this value the motion with the TW ceases, and the motion is almost symmetric in both directions. This is because the particles do not move a significant amount during one TW cycle, and the changes in direction of their velocities are the dominant effect in determining the motion.

We can analyze this behaviour by going to the comoving frame, where a Hamiltonian can be written¹⁹. Rescaling $\xi = \frac{k}{d}x - \omega t$, $\zeta = \frac{\pi}{d}z$ and $\tau = \omega t$ we obtain

$$H = \frac{V_0 k}{\omega d} \sin \xi \sin^2 \zeta + \zeta \quad (3)$$

We see that the combination of parameters $R = \frac{V_0 k}{\omega d} = \frac{V_0}{V_{TW}}$ is more appropriate for describing the system than just the frequency ω . For $R > 1$ this Hamiltonian has a separatrix that divides (ξ, ζ) space into closed and open orbits. Thus it is the open orbits that comprise the backflow, while the closed ones define a new “reduced” comoving roll. We can estimate the experimental value of $R = \frac{V_0 k}{\omega d}$ by measuring the backflow and TW velocities. The data of Fig. 2b at a TW velocity of $\approx 40 \frac{\mu m}{sec}$ can be described by $R \approx 2$, giving $V_0 \approx 80 \frac{\mu m}{sec}$. An interesting feature of this Hamiltonian is the existence of a transition in the topology of the trajectories. As we decrease R the separatrix orbit shrinks, till at $R = 1$ it disappears. This is the point we associate with the transition from fig. 18 to fig. 19, when mass propagation with the TW velocity ceases to exist. Fig. 20 shows this transition in the streamlines of H for different values of the parameter R .

We can find the origin of this transition in the fixed point behaviour of H . If we look at the $[0, 1] \times [0, 1]$ square these are the points $\xi = \frac{\pi}{2}, \zeta = \frac{1}{2} \arcsin(-\frac{1}{R})$ (for the $[-1, 0] \times [0, 1]$ square the relations are similar but reflected round $\zeta = \frac{\pi}{2}$). The value of H at the two fixed points is

$$H_{fp} = \frac{R}{2} \left(1 \mp \sqrt{1 - \frac{1}{R^2}} \right) + \frac{1}{2} \arcsin(-\frac{1}{R}) \quad (4)$$

For $R > 1$ these fixed points exist, while for $R < 1$ they are undefined. Looking along the line $\xi = \frac{\pi}{2}$ these points are extremes of H . One of these fixed points (the maximum of H) describes the center of the comoving roll, while the other sits on the separatrix (actually a homoclinic orbit). In the limit $R \rightarrow \infty$ the fixed point at the center of the roll remains, becoming the standard fixed point of stationary convection. Indeed $R = \infty$ is the $\omega \rightarrow 0$ limit of no TW. The other fixed point is then pushed towards the boundary $\zeta \rightarrow \pi$, which in the comoving frame is always moving at velocity $V_T = 1$. In the limit $R \rightarrow 1$ the two points coalesce into an inflection point, and disappear for $R < 1$.

VI. Discussion

The main objective of this work has been to identify the processes of mass transfer by TW. We have found that for nonlinear TW a “reduced” comoving roll carries mass at the TW velocity. The rest of the fluid, in the area that surrounds the reduced rolls, carries mass back, taking care of mass conservation. Both of these are reproduced from a simple model for the TW. This result reinforces the ideas of the amplitude equation approach, that information on the velocity can be obtained from information on the temperature (and concentration).

A step beyond this is the differentiation between linear and nonlinear regimes based on mass transport. We have found a transition to mass transport that coincides with the onset of the nonlinear state. Mass transport can obviously perturb the concentration field, leading to the breaking of the linear profile. In the linear regime any given fluid volume is executing oscillatory motion round its equilibrium point, and the concentration profile to be a linear. Once mass begins to move with the TW the concentration stratification may be disturbed by a varying degree. This change would lead away from the linear regime, into the various nonlinear TW states. Such an effect would dominate over any boundary layer process¹¹ (that also tends to distort the linear concentration profile), since this depend on changes in V_0 , which are small for the confined and blinking states (the exact change depends on the final state and on ϵ).

It is interesting that the blinking state, although only weakly nonlinear, is robust under this distortion of the linear concentration profile. It would be interesting to simulate numerically (perhaps for the exact solutions) the behaviour of the concentration field under the various types of flow.

The difference between the regimes is seen also when we look at possible models for the system. For the translational symmetry of the TW we can go to the comoving frame, removing the explicit time dependence from the equations of motion. This leads to a time independent Hamiltonian. In the case of coexisting left and right TW such a time independent H cannot be written. The time dependence of H may lead to interest

ing new phenomena, related to chaotic mixing.

A few questions are interesting and as yet remain unanswered. Although the model we use is a crude one, it gives a prediction for the R dependence seen by performing a scan along the TW branch. Measuring the main and backflow velocities, for example, would enable checking the R dependence of the relative size of the two oppositely moving volumes.

Another open question concerns the mixing region where main and back flows join at the edges of the convection pattern. Obviously fluid transported to the edge of the cell with the TW must find its way into the backflow, be transported to the other edge of the convection pattern, and there be reinjected into the main flow to maintain the cycle. The way in which this is done could reveal much about the role of the boundaries. The work done by Cross¹⁰ has shown the importance of the boundaries and the reflection there, and it would be interesting to assess the effect of a mixing layer on these. The model considered does not incorporate this mixing. We did not model also 3D effects in a mass transport and did not find an explanation for 3D structure and parabolic shape of the fronts observed in the experiment.

Thus the distinct feature of nonlinear TW states is the existence of the mass transport with propagation velocity of TW. From our experiments²⁰ we know depending that on a stepsize in a temperature difference from the convection threshold different TW states can be reached at different values of the amplitude. It means that transition from linear to a nonlinear state occurs at different values of the parameter R which determines the transition in a topology of the particle trajectories. Therefore we can conclude that the onset of the mass transport accompanies the transition from the linear to the nonlinear oscillatory state, but is not the primary reason for the instability. The absence of mass transport in the linear TW is perhaps related to the fact that the CPW are basically a form of standing waves.

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Figure Captions

- Fig. 1:** Absorption spectrum of the photochromic compound used, in a mixture of 95% wt. benzene and 5% wt. methanol.
- Fig. 2:** Schematic diagram of the experimental setup.
- Fig. 3:** Logarithm of the attenuation profiles along the cell show the diffusion and decay of the dye in the conductive state. Time progresses upwards, and the interval between profiles is 12 sec. Initial irradiation lasted 10 sec.
- Fig. 4:** Single profile of the dye distribution along the cell. Points are data, and the curve is a best fit to the model. Taken at $t = 300$ seconds.
- Fig. 5:** Decay prefactor versus time for the profiles obtained from fig. 3. Points are values from the individual fits, and the curve is a best fit to an exponential behaviour.
- Fig. 6:** Variance of the gaussian versus $2 \cdot t$ for the profiles obtained from fig. 2. Points are values from the individual fits, and the curve is a best fit to a linear behaviour.
- Fig. 7:** Attenuation profiles along the cell show the spreading of the dye from a strip near midcell in the stationary convection state. $\epsilon \approx 0.02$. Time progresses upwards, and the interval between profiles is 5 sec.
- Fig. 8:** Attenuation profiles along the cell show the spreading of the dye from a strip near the edge of the cell in the same state as fig. 7. Time progresses upwards, and the interval between profiles is 3 sec.
- Fig. 9:** Attenuation profiles across the cell show the modulation of an initially homogenous strip of dye along the cell in the same state as fig. 7. Time progresses upwards, and the interval between profiles is 3 sec.
- Fig. 10:** Attenuation profiles along the cell in a confined state in the hysteretic region of the TW branch with $\frac{q}{q_c} = 0.998$, after initial UV irradiation near mid cell. Time progresses upwards, and the interval between profiles is 7 sec.

- Fig. 11:** Attenuation profiles along the cell in a confined state in a different cell ($d = 1.41$) in the confined TW state with $\epsilon = 0.007$, after initial UV irradiation near mid-cell. Time progresses upwards, and the interval between profiles is 4 sec.
- Fig. 12:** As in fig. 10, but with initial irradiation near edge of cell.
- Fig. 13:** a) Shadowgraph image of the confined state at $\frac{q}{q_c} = 1.008$.
b) Consecutive images of the dye show its propagation in a confined state with $\frac{q}{q_c} = 1.008$, for an initial UV irradiation near the center of the cell.
c) As in b), but for initial irradiation near the edge of the cell.
Solid frame indicates the borders of the experimental cell.
- Fig. 14:** Attenuation profiles across the cell show the modulation of an initially homogenous strip of dye along the cell in the same state as fig. 10. Time progresses upwards, and the interval between profiles is 7 sec.
- Fig. 15:** Attenuation profiles for a blinking state. $\epsilon = 0.0027$. a) Right TW.
b) convection in both parts of cell. c) Left TW. Time progresses upwards, and the interval between profiles is 5 sec.
- Fig. 16:** a) Shadowgraph intensity profiles along the cell during the CPW transient. Time progresses upwards, and the interval between profiles is 15 sec.
b) Attenuation profiles along the cell during CPW and their transition to TW after initial UV irradiation of strips in both sides of the cell. Time progresses upwards, and the interval between profiles is 4 sec.
- Fig. 17:** a) Computer simulation of the TW equations of motion. Shown are the positions of 2048 particles in the convection cell at constant time intervals, after being evenly distributed at $t = 0$ in the $[0, 1] \times [0, 1]$ square. The grid moves at the TW velocity, and $R = 3.2$
b) Particle density (integrated along z) as a function of x from the numerical simulation of a). The x interval is divided in 128 bins. Time progresses

upwards, and the interval between profiles is 0.5 units of time.

Fig. 18: a) As in fig. 17a, for $R = 1.1$

b) As in fig. 17b, for $R = 1.1$

Fig. 19: a) As in fig. 17a, for $R = 0.5$

b) As in fig. 17b, for $R = 0.5$

Fig. 20: Contours of equal H on the (ξ, ζ) plane bounded by $0, \pi$ in both directions.
Different value of R are signified above each figure.

Fig. 1

Fig. 2

F.g. 4

F. 5. 5

F. 3 6

F. 5. 7

Fig. 2

Fig. 1

Fig. 10

Fig. 11

Fig. 12

(b)

(c)

Fig. 11

1. 11.

(a)

(b)