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Fronts and trigger wave patterns in an array of oscillating vortices

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Abstract – We present experiments on reaction fronts and self-sustaining trigger wave patterns in an advection-reaction-diffusion system with chaotic mixing. The flow is a two-dimensional array of oscillating vortices, and the reaction is the excitable Belousov-Zhabotinsky chemical reaction. Reaction fronts are found to mode-lock for a wide range of frequencies, and the mode-locking results in "faceted" fronts that line up along the directions of the underlying vortex array. Self-sustaining trigger wave patterns are also found composed of large scale spiral and spiral-pair patterns, similar to patterns in the reaction-diffusion (no flow) limit but with a significantly larger typical length scale and with clear anisotropy that reflects the vortex array.

Reaction-diffusion (RD) systems are often characterized by front propagation and by the formation of trigger wave patterns – self-sustaining patterns of outward propagating rings or of rotating spirals. Reaction-diffusion models [1] are used to explain a wide range of phenomena, including propagating flame fronts [2], waves of electrical activity in the heart [3], patterns formed by predator-prey population systems [4], and morphogenesis [5,6]. By definition, an RD system is one in which there is no fluid motion; however, many reacting systems are affected significantly by enhanced mixing due to fluid flows. Flows play a significant role in, for example, microfluidic reactors, solidification in a flowing liquid, forest fires in the presence of a wind, and the spreading of a disease in a moving population. The general advection-reaction-diffusion (ARD) problem has only recently begun to receive attention [7–9]. In this article, we present ARD experiments on reaction fronts and trigger waves in a laminar fluid flow with chaotic advection [10,11]. We use the well-known Belousov-Zhabotinsky (BZ) chemical reaction [12–15] in a flow that is composed of a two-dimensional (2D) array of counter-rotating vortices that oscillate laterally in a circular motion.

Reaction fronts in an RD system propagate with a speed given by the Fisher-Kolmogorov-Petrovsky-Piskunov (FKPP) prediction [16,17]: $v=2\sqrt{D/\tau}$, where D is the molecular diffusion coefficient and τ is the reaction time scale. The presence of a flow with chaotic mixing results in enhanced mixing that often is diffusive with enhanced diffusivity D^* [18]. However, it is not sufficient simply to replace D with D^* in the FKPP prediction to determine front speeds for ARD systems. Recent theories [19,20] and experiments [21,22] showed that vortices in a flow have a significant effect on one-dimensional front propagation. In particular, a moving vortex tends to pin and drag a front [23]; in the reference frame of the vortex, the front is frozen in the face of an imposed wind.

If a chain of vortices oscillates periodically, mode-locking often occurs. Mode-locking is often found in oscillating systems that are forced periodically by an external perturbation. If the amplitude of the perturbation is large enough, the natural oscillation of the system may change and "lock" to the external perturbation, with the two oscillating with frequencies that are rationally related, *e.g.*, 1 : 1 or 1 : 2 or 3 : 2, etc. For front propagation in a cellular flow, mode-locking behavior has been found [20–22] in which the front propagates an integer number N of wavelengths λ (vortex pairs) in an integer number M of drive periods T, resulting in a front velocity:

$$v=N\lambda/MT=\frac{N}{M}\lambda f, \tag{1}$$

where f is the frequency of oscillation. The previous studies were essentially one-dimensional (1D), conducted for a chain of vortices. The experiments presented here extend the previous studies to a geometry that allows for fully 2D propagation.

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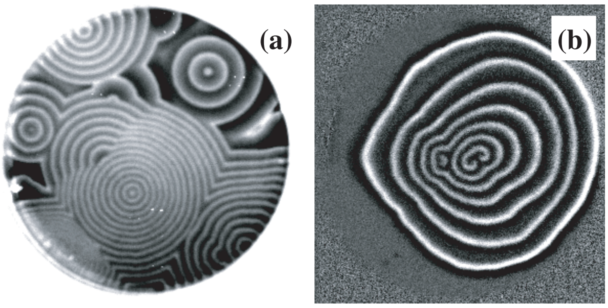

Fig. 1: Self-sustaining trigger wave patterns in reaction-diffusion (no flow) limit. (a) Outward-propagating target patterns. (b) Spiral-pair generator.

Self-sustaining trigger wave patterns are common in RD systems in which fronts can propagate in two dimensions. Figure 1 shows two different mechanisms by which these patterns can form. Target patterns (concentric, outwardly-propagating rings) form if there are slight imperfections or impurities that continually retrigger fronts at one or more locations in the system (fig. 1a). Alternately, if a front is broken, the ends curl inward to form a spiral that continually rotates. If both ends of a broken front curl inward, the result is a pair of spirals that acts as a generator for a self-sustaining pattern of outward propagating rings (fig. 1b). Alternately, individual rotating single spiral patterns are also possible.

The flow studied in these experiments is a square array of counter-rotating vortices, generated using magneto-hydrodynamic forcing (fig. 2). An electrical current (85 mA) passes horizontally through a 3 mm thick layer of an electrolytic solution. This current interacts with an alternating magnetic field produced by a 40×40 array of 1/4 inch (0.64 cm) diameter Nd-Fe-B magnets located below the fluid layer. Lorentz magnetic forces push the fluid horizontally in one direction above magnets with the north side up and push the fluid in the opposite direction above magnets with the south side facing up. The result is a 25.4 cm $\times$ 25.4 cm square array of 1600 vortices in the fluid layer.

For small electrical current, the vortex array described above is time-independent, and there is no chaotic mixing between adjacent vortices. Periodic time dependence and chaotic fluid mixing are achieved by oscillating the fluid layer horizontally across the vortex array in a circular motion. This oscillatory time dependence is imposed with the use of two pairs of displacement plungers on a pivoting lever that oscillate in the side reservoirs, displacing fluid slowly back and forth across the vortex array. For all cases in this paper, the amplitude of the oscillation is 0.12 as a fraction of the vortex width. The phase difference between the two perpendicular plunger sets is $\pi/2$ radians, ensuring circular oscillations of the fluid over the vortex array. Previous studies of a 1D analog of this flow (a chain of oscillating vortices) have demonstrated chaotic mixing between vortices and (for most frequencies) enhanced diffusive mixing [18,24]. Similar chaotic transport and enhanced diffusion apply to the 2D vortex array when oscillating.

The fluid layer is composed of the chemicals for the ferroin-catalyzed, excitable BZ reaction [15][1]. Before each run, a line of 1.0 M NaBr is injected around the perimeter to shield the region of interest (ROI) from fronts that trigger spontaneously in the reservoirs. A front is then intentionally triggered within the ROI by momentarily dipping the end of a silver wire into the fluid.

In the absence of any lateral oscillations of the fluid (*i.e.*, the plunger sets are inactive) a front triggered near the left side of the ROI progresses as shown in fig. 3a. The front advances by a combination of advection around individual vortices and reactive burning across separatrices between one vortex and its neighbors. Since the excitable BZ reaction produces pulse-like fronts with a refractory process, the front continually re-triggers within individual vortices, leaving significant activity in the vortices behind the advancing front. The result is a pattern that reflects the underlying vortex structure on small scales and resembles a roughly circular front on large scales.

The behavior is quite different in the presence of lateral oscillations and chaotic mixing (fig. 3b–d). First, the front is an isolated, pulse-like front with no significant activity in its wake. This is due to chaotic advection which enhances mixing of the inhibitor in the wake of the advancing front, preventing re-triggering of the reaction within individual vortices. Second, signatures of chaotic advection are evident in the advancing front. The front no longer relies on reactive burning to cross between vortices – it is instead advected across separatrices between vortices, following the lobe structure typical of chaotic advection in an oscillating vortex flow (see fig. 2 from ref. [18]).

Third, the underlying anisotropy of the vortex array can have a significant effect on the shape of the front, especially if the front is mode-locked to the external forcing. To investigate mode-locking in these experiments, spacetime plots are made from time sequences along a horizontal strip in the images. Figures 4a–c show the motion of the fronts in figs. 3b–d, respectively. The spacetime plots in figs. 4a and c are taken at the mid-height of the images, whereas that for fig. 4b is taken near the bottom. Front velocities can be determined from the inverse of the slope of these spacetime plots. When mode-locked, the slope is clearly defined, such as in fig. 4a and the second half of fig. 4b; if the front is not mode-locked, the slope is less well defined, as in fig. 4c and the first half of fig. 4b.

Mode-locking can be verified rigorously by comparing front velocities to those from eq. (1). As seen in fig. 4d,

[1]The recipe used: separately mix 48.9 g sodium bromate and 202 ml of 1 M sulfuric acid in 472 ml water, 11.25 g Malonic acid in 112.5 ml water, and 11.25 g sodium bromide in 112.5 ml water. Mix the three solutions together under a vent hood until clear, mix in 8 ml ferroin indicator and then pour into apparatus.

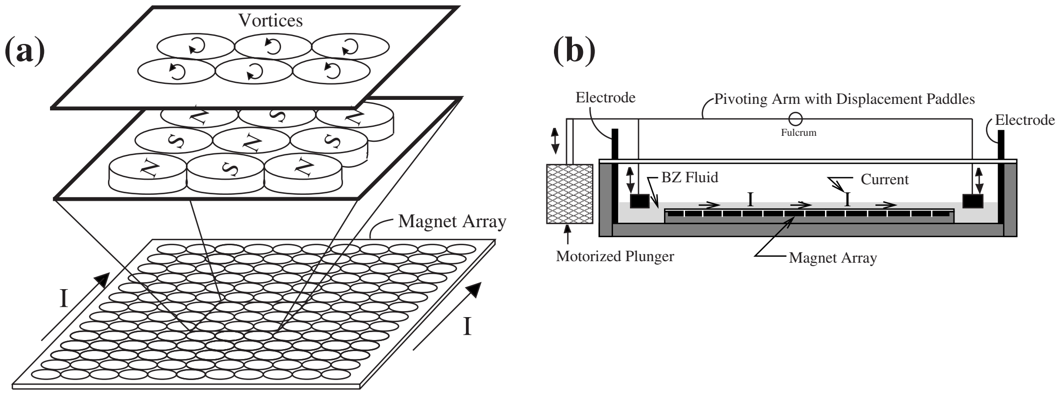

Fig. 2: (a) Exploded view of magnetohydrodynamic forcing. A 40×40 array of magnets with alternating polarity sits below a fluid layer through which a current passes, producing an array of vortices. (b) Side view of apparatus. The fluid layer is 3 mm thick. A pair of displacement plungers slowly oscillates the fluid left-right across the vortex array; another set (not shown) oscillates the fluid horizontally in-out of the plane of the page.

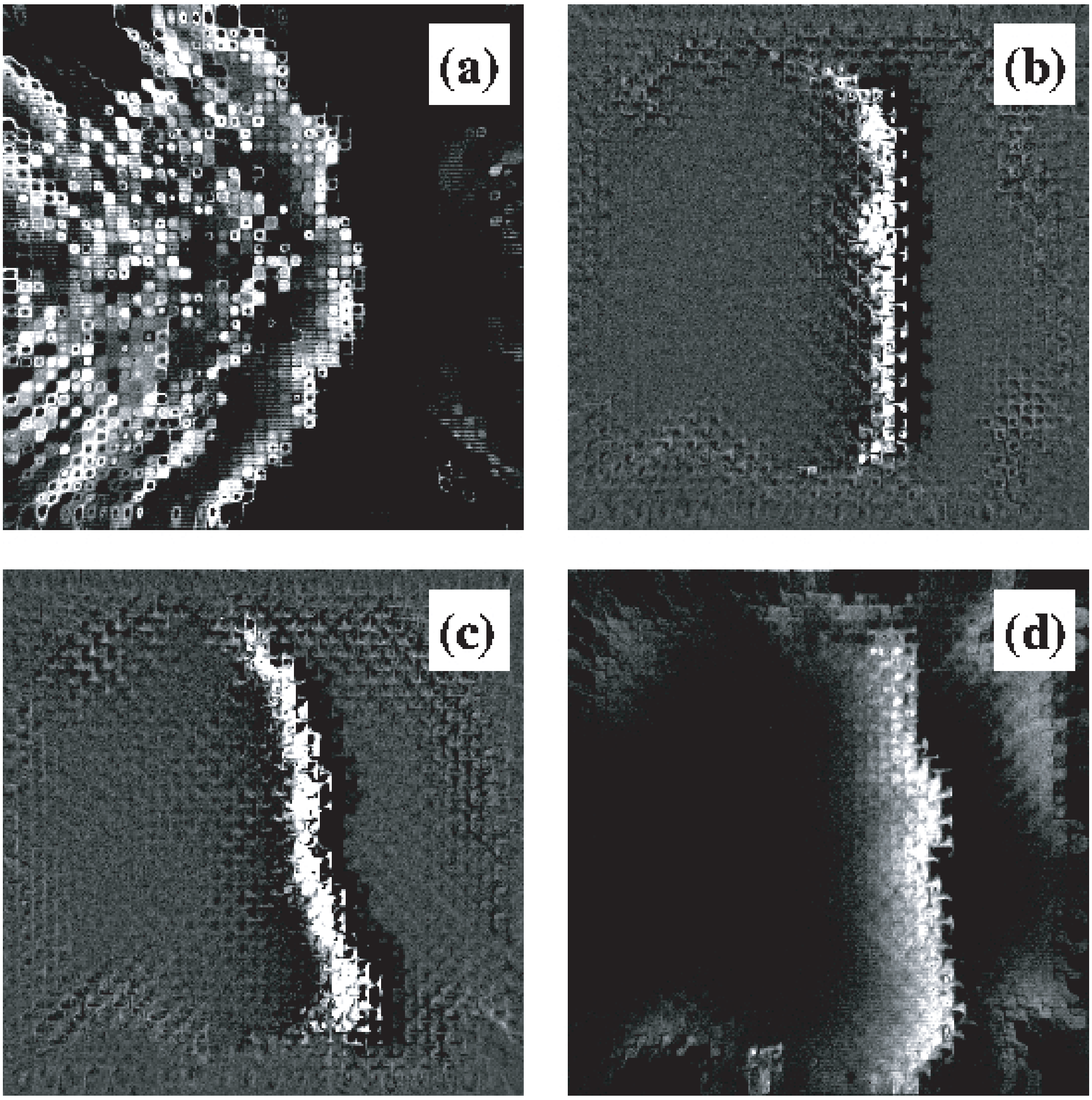

Fig. 3: Fronts propagating in vortex array. (a) No oscillations or chaotic mixing. (b) Lateral oscillations, $f = 0.050$ Hz. The entire front is mode-locked in this case. A movie of this sequence is available online (`fig3b_movie.avi`, 4.6 MB). (c) Oscillations with $f = 0.070$ Hz. The bottom part of the front is mode-locked, but the rest is not. (d) Oscillations with $f = 0.080$ Hz. Mode-locking is not evident in this case.

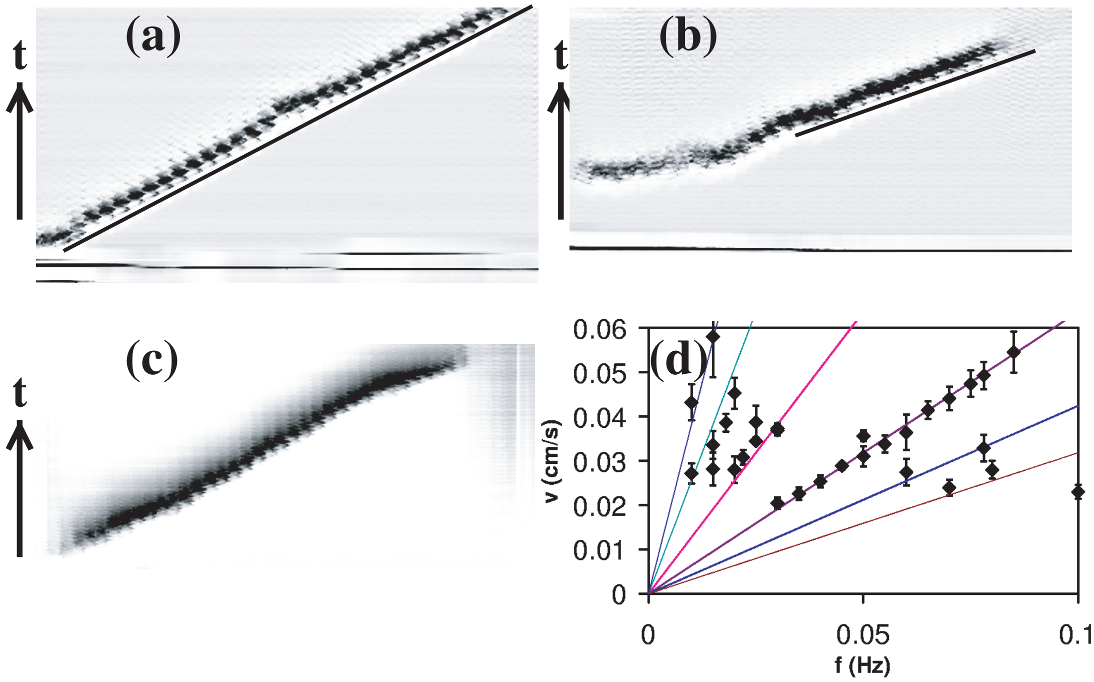

Fig. 4: (a) Spacetime plot corresponding to front motion along a line at the midheight in fig. 3b; frequency $f = 0.050$ Hz, mode-locked. (b) Spacetime plot along line near bottom of fig. 3c; $f = 0.070$ Hz; mode-locking starts about halfway through the run. (c) Spacetime plot along line at midheight of fig. 3d; $f = 0.080$ Hz; mode-locking not evident. (d) Front velocities; experimental values (filled diamonds) are compared with theoretical mode-locking predictions from eq. (1). The diagonal solid lines show theoretical predictions for mode-locking with $(N, M) = (3, 1)$, (2,1) (1,1), (1,2), (1,3) and (1,4), respectively, from left to right.

for a large range of frequencies, the front velocities agree quite well with those for $(N, M) = (1, 2)$ mode-locking, *i.e.*, where the front propagates one wavelength every two drive periods. It is reasonable that the (1,2) mode-locking regime dominates since this is the locking ratio for which the front propagates one vortex every period of the forcing. Mode-locking in the (1,1) regime is also apparent in fig. 4d, although the range of frequencies is significantly less than that for the (1,2) regime. There are also hints of mode-locking with other locking ratios, but the evidence is not as clear as for (1,2) and (1,1) locking. For more discussion of mode-locking in front propagation, see refs. [21,22] and [23].

When mode-locked, the front tends to be flat and "faceted". After a brief transient, a mode-locked front advances in rhythm with the external forcing, with a periodic pattern of lobes, as seen in fig. 3b. The front initially grows as a coarse-grained arc, but it quickly starts to align with the underlying vortex array. Trailing parts of the front catch up to the leading edge, due to sideways propagation of the front – chaotic advection applies in both the x- and y-directions. The result is an almost perfectly straight, "faceted" front whose macroscopic orientation reflects the anisotropy of the underlying (small-scale) vortex array, similar to the manner in which faceted crystals reflect the microscopic anisotropy of packing of individual molecules in the crystals. This faceted behavior is self-correcting; if any part of the front momentarily lags, the neighboring parts of the front propagate laterally to fill in the gap.

If a front is not mode-locked or is only partially locked, the faceted behavior is not as striking, although the large-scale structure is still influenced by the small-scale anisotropy of the vortex array. An example of a partially locked front is shown in figs. 3c and 4b. The frequency of oscillation (0.070 Hz) is larger than that for figs. 3b and 4a (0.050 Hz), and most of the front cannot keep up with the larger velocity needed to remain locked. Although still anisotropic, there is less of a self-correcting mechanism to correct for deviations from a faceted front. About halfway through this particular run, the bottom portion of the front manages to mode-lock to the external forcing and surges ahead of the rest of the front due to the higher frequency. A front at a higher oscillation frequency is shown in figs. 3d and 4c; either this front is unlocked or is locked (temporarily) in a regime with a higher-order (N, M) combination.

Self-sustaining patterns of trigger waves are found in this ARD system with chaotic mixing, as seen in fig. 5. These patterns are generated in a slightly different manner than those of fig. 3. After a front is triggered, a few drops of 0.25 M NaBr are added directly in front of part of the advancing reaction, breaking the reaction pulse and allowing it to curl back on itself. If a small portion

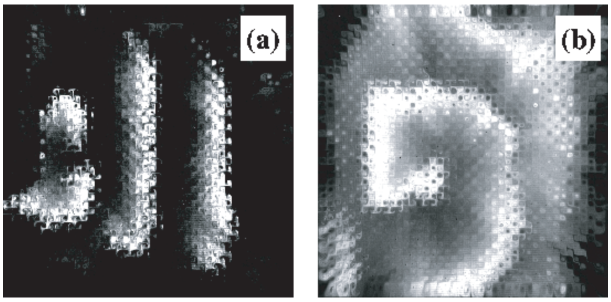

Fig. 5: Self-sustaining trigger wave patterns; $f = 0.050\,\text{Hz}$, in the regime with (1,2) mode-locking. (a) Spiral-pair generator. A movie of this sequence is available on-line (fig5a_movie.avi, 2.0 MB). (b) Single spiral.

of the middle of the pulse is broken (fig. 5a), a spiral-pair generator is formed (near the left of fig. 5a). Similar to the spiral-pair generator in the RD case (fig. 1b), the generator in fig. 5a produces a series of fronts that propagate away from the generator. It is also possible to produce a single spiral, as shown in fig. 5b. Square-shaped, outwardly propagating target patterns have also been found in this ARD system, similar to those in fig. 1a, but with a much larger average wavelength.

Since propagating fronts are mode-locked at the frequency for figs. 5a and b, the self-sustaining trigger wave patterns for the ARD case – which are composed of a series of propagating pulses – show strong anisotropic behavior, lining up with the underlying vortex array. The increased length scale of these patterns is due to the significant enhancements in front velocity due to fluid advection. Assuming roughly the same refractory time for recovery of the reactants as for the RD case, the average wavelength should increase proportionally to the propagation speed. Note also that even a 40×40 array of vortices is limited in size with respect to these patterns. Presumably, the fronts in fig. 5a would be part of a pattern of outward-propagating squared pulses.

The pattern formation process seen here is inherently two dimensional. Earlier experiments [21,22] with the excitable BZ reaction in an annular chain of oscillating vortices revealed mode-locking behavior, but large-scale pattern formation was absent in that effectively 1D geometry. The self-sustaining patterns seen here also require chaotic mixing in this system. In the absence of lateral oscillations responsible for chaotic mixing between vortices, there are no large-scale patterns at all, as seen in fig. 3a.

Summarizing, for reactions in a 2D vortex array, we find front propagation and trigger wave patterns similar in some respects to those in a reaction-diffusion system with no flows. However, the underlying vortex structure imposes significant anisotropy on the fronts and on the pattern formation process, particularly when the fronts are mode-locked to external perturbations. We are currently conducting experiments to extend theses results to spatially random vortex flows and to flows with more complicated time dependence, including weakly turbulent flows. The goal is to determine how dependent the large-scale pattern formation is on the specific flow used and how general the phenomena is. Ultimately, this work has the potential for long-term applications to microfluidic reactors, many of which involve chaotic mixing in small-scale periodic lattices. The effect of small-scale cellular structures on large-scale patterns may also be relevant to understanding the effects of discrete cells and nerves on waves of electrical activity in the heart and brain. Finally, ARD studies such as this may shed light on pattern formation in ecosystems in fluid flows, *e.g.*, phytoplankton and algae blooms in the Atlantic Ocean and Gulf of Mexico.

* * *

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