MODE LOCKING, THE BELOUSOV–ZHABOTINSKY REACTION, AND ONE-DIMENSIONAL MAPPINGS

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We demonstrate that complex sequences of periodic states such as those observed in the Belousov–Zhabotinsky reaction can be generated by simple one-dimensional maps. Motivated by the experimental data, we construct a map which reproduces most of the complicated devil's staircase observed by Maselko and Swinney as well as chaos and other experimentally observed periodic sequences. An interesting property of the devil's staircase observed here is that it remains complete through a wide range of parameters, in contrast to the devil's staircases observed in critical circle maps. We also comment on a new class of mode-locking sequences.

One of the most fascinating phenomena in nature is the spontaneous oscillation seen in the Belousov–Zhabotinsky (BZ) reaction. In a typical experiment reactants are pumped into a stirred tank at a constant rate. In spite of the fact that none of the external conditions vary in time, the concentrations of the reactants oscillate with temporal patterns ranging from simple periodic motion to aperiodic chaotic behavior. An interesting aspect recently observed by Maselko and Swinney [1] is that by changing the catalyst from the normally used cerous ion to the manganese ion, there is a regime in which variations of the flow rate cause the pattern of the oscillation to go through a complicated sequence of different periodic states. In particular, the amplitude of the peaks of the oscillations are quantized into high and low values, as depicted in fig. 1d. An overall property characterizing this pattern is what we will call the firing rate, defined as the ratio of the number of small peaks to the total number of peaks per oscillation. Maselko and Swinney demonstrate that a plot of the firing rate versus the flow rate consists of a sequence of flat steps, corresponding to different types of periodic behavior, joined together by jumps of infinite slope, corresponding to bifurcations. If the jumps in this curve form a Cantor set it is called a devil's staircase, and if it has zero measure the devil's staircase is said to be complete.

Behavior of this type has also been observed in many other systems. In a periodically driven analog neuron model, for example, Harmon [2] found that as the driving frequency was varied a plot of the firing rate versus the frequency formed a complete devil's staircase. Building on this work, Sato and others [3] simplified Harmon's model to a piece-wise linear one-dimensional map and showed that it generated the same kind of devil's staircase. Tomita and Tsuda [4] argued that a similar map might apply to the BZ reaction. Further study of the map led Tsuda to predict self-similarity in the bifurcation structure of the BZ reaction in some parameter regimes as the flow rate is varied [5].

A close examination of the Tomita and Tsuda model reveals, however, that the class of maps they studied is only capable of producing a small number of possible periodic states. Combinations of consecutive large peaks and small peaks are not possible, for example, three large peaks followed two small peaks. Furthermore, the map they devised does not look like maps previously constructed from experimental data. As described below, we study a map whose shape is motivated by experiments performed in the chaotic regime of the BZ reaction, and show that it is capable of generating behavior qualitatively like that of the Maselko–Swinney experiments.

In certain parameter regimes the chaotic behavior

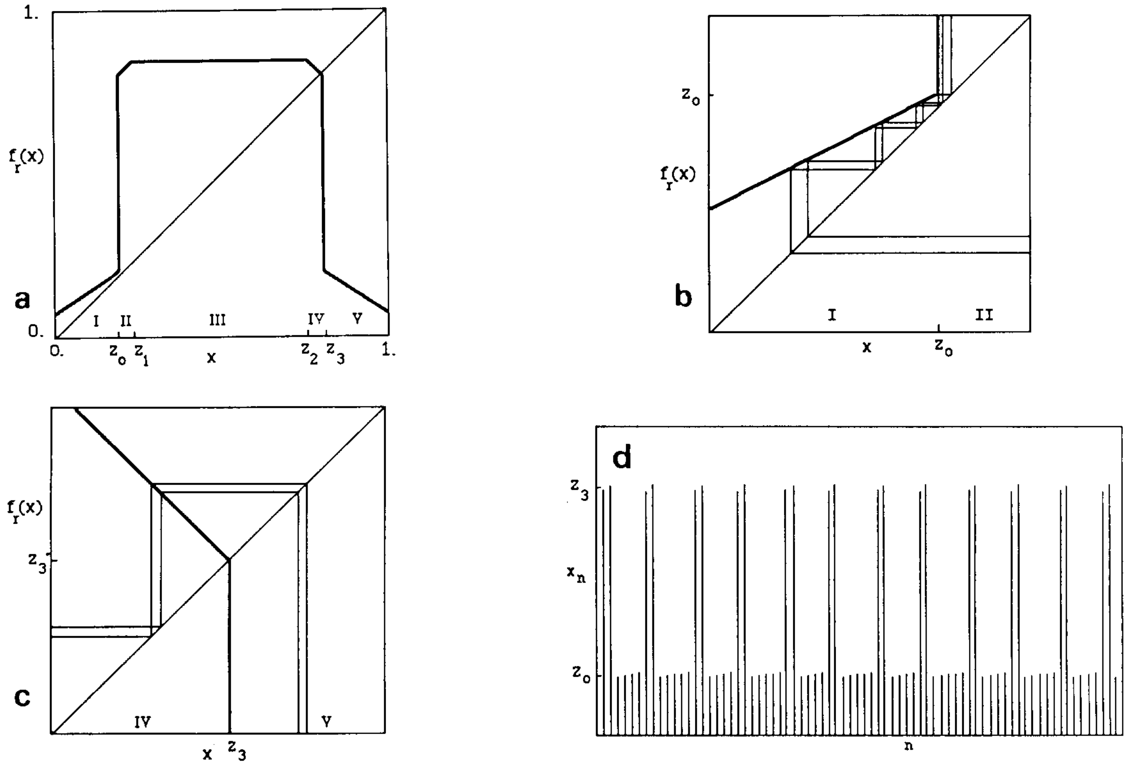

Fig. 1(a) The piecewise linear hump map f_i is defined in regions I, II, III, IV, and V as follows: $f_I = m_1(x - z_0) + z_0 + c_1$, $f_{II} = m_2(x - z_0) + z_3 + c_2$, $f_{III} = m_3(z_1 - z_0) + z_3 + c_2$, $f_{IV} = m_4(z_3 - x) + z_3 + c_2$, $f_V = m_5(z_3 - x) + z_0 + c_3$. (b, c) Magnification of regions II and IV near the identity line to show the action of the orbit. (d) An example of a time series for the orbit $L^2S^5L^2S^4$.

of the BZ reaction can be described quite well in terms of a simple one-dimensional map. This map consists of a single hump that is tapered off on one side, roughly similar to $f(x) = x\,e^{-x}$. Motivated by this, and in an attempt to explain the devil's staircase phenomena we have constructed the piecewise linear map as shown in fig. 1a. By construction the iterates of this map are quantized into large values, denoted L, and small values, denoted S. A given periodic state can be characterized by its sequence of S and L values. To generate a sequence of periodic orbits, we pick a function $f(x)$ and introduce a bifurcation parameter r which scales the height of the function in the form:

$$f_r(x) = rf(x). \tag{1}$$

It is convenient to describe a symbolic sequence Ω which represents a particular periodic orbit in the following form:

$$\Omega = \prod_{i=1}^{k} L^{m_i} S^{n_i},$$

where L (S) denotes large (small) amplitude peaks and m (n) denotes the number of consecutive large (small) peaks which occur in k groups, the period being

$$P = \sum_{i=1}^{k} (m_i + n_i).$$

With a choice of parameters which determine the shape of the map, we generate a progression of periodic states as we increment r as shown in fig. 2. As a measure of the mapping's success in reproducing the behavior of the Maselko–Swinney experiments, we compare in table 1 a subset of the periodic states shown in fig. 2 with the complete sequence of pat-

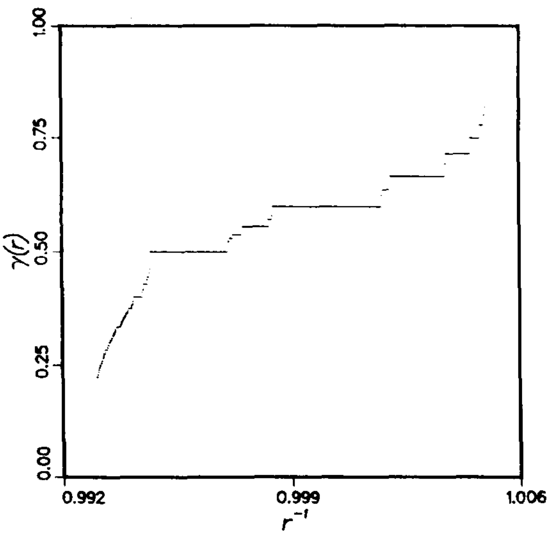

Fig. 2. Plot of "firing rate" versus r generated from the map in fig. 1b. For this sequence, $m_1 = m_5 = 0.4, m_2 = m_3 = m_4 = 1.5, c_1 = 0.001, c_2 = -0.01, c_3 = -0.003, z_0 = 0.20, z_1 = 0.23, z_2 = 0.77$, and $z_3 = 0.80$.

terns generated from the chemical experiment. Clearly, we can obtain much higher resolution with a numerical experiment and thus can identify intervening periodicities which span smaller intervals in r. By slightly varying these parameters, we have reproduced most of the experimental periods which are missing in this table as well. To get from any given periodic state to another there are many different paths through parameter space, with many different sequences along each path. Some of these sequences of patterns generate devil's staircases which are more complicated than the one shown in fig. 2. Thus, there is a considerable amount of arbitrariness in the particular sequences that we have shown here, since by varying parameters it is clear that we can sweep out an infinite sequence of different periodic orbits of arbitrarily large periods.

Several of the transitions in Maselko and Swinney's experimental results involve simple period adding (L^4S^1 from L^3S^1). Other examples of period adding are more complex (L^2S^6 to $L^2S^6L^2S^5$ to L^2S^5) be-

Table 1
Comparison of two sequences of periodic orbits (Ω) generated from the chemical experiment (A) and the mapping (B) with the onset (r) and the firing rate (γ). The parameter settings for the mapping are the same as for fig. 2.

<table>
<tr><th rowspan="2">r_{BZ}</th><th colspan="2">$\Omega(\gamma)$</th><th rowspan="2">r_{map}</th></tr>
<tr><th>A</th><th>B</th></tr>
<tr><td>0.1360</td><td>$S^1L^4(1/5)$</td><td rowspan="2">period > 200</td><td rowspan="2">r > 1.00641</td></tr>
<tr><td>0.1294</td><td>$S^1L^3(1/4)$</td></tr>
<tr><td>0.1254</td><td colspan="2">$S^2L^4(2/6)$</td><td>1.00641</td></tr>
<tr><td>0.1230</td><td>$S^1L^2(1/3)$</td><td></td><td></td></tr>
<tr><td>0.1148</td><td>$S^2L^3(2/5)$</td><td>$S^2L^2S^2L^4(4/10)$</td><td>1.00587</td></tr>
<tr><td>0.1114</td><td>$S^2L^2S^1L^2(3/7)$</td><td>$(S^2L^2)^2S^2L^4(6/14)$</td><td>1.0057</td></tr>
<tr><td>0.1100</td><td>$(S^2L^2S^1L^2)^2(S^3L^3)(9/20)$</td><td></td><td></td></tr>
<tr><td>0.1094</td><td>$(S^2L^2S^1L^2)^2(S^3L^3)^2(12/26)$
$(S^2L^2S^1L^2)(S^3L^3)(6/13)$</td><td>$(S^2L^2)^5S^2L^4(12/26)$</td><td>1.00536</td></tr>
<tr><td>0.1092</td><td>$S^3L^3(3/6)$</td><td rowspan="2">$S^2L^2(2/4)$</td><td rowspan="2">1.00534</td></tr>
<tr><td>0.1040</td><td>$S^1L^1(1/2)$</td></tr>
<tr><td>0.0998</td><td colspan="2">$S^2L^2S^3L^2(5/9)$</td><td>1.00252</td></tr>
<tr><td>0.0980</td><td colspan="2">$S^3L^2(3/5)$</td><td>1.00155</td></tr>
<tr><td>0.0974</td><td colspan="2">$S^3L^2S^4L^2(7/11)$</td><td>0.99818</td></tr>
<tr><td>0.0968</td><td colspan="2">$S^4L^2(4/6)$</td><td>0.99796</td></tr>
<tr><td>0.0948</td><td colspan="2">$S^4L^2S^5L^2(9/13)$</td><td>0.99629</td></tr>
<tr><td>0.0940</td><td colspan="2">$S^5L^2(5/7)$</td><td>0.99625</td></tr>
<tr><td>0.0928</td><td colspan="2">$S^5L^2S^6L^2(11/15)$</td><td>0.99553</td></tr>
<tr><td>0.0924</td><td colspan="2">$S^6L^2(6/8)$</td><td>0.99532</td></tr>
<tr><td>0.0896</td><td colspan="2">$S^6L^2S^7L^2(13/17)$</td><td>0.99523</td></tr>
<tr><td>0.0876</td><td colspan="2">$S^7L^2(7/9)$</td><td>0.99522</td></tr>
<tr><td>0.0872</td><td colspan="2">$S^nL^2(n > 7)(n/(n+2))$</td><td>0.99510</td></tr>
<tr><td>0.0816</td><td>$S^nL^1(n > 7)(n/(n+1))$</td><td>S</td><td>0.99502</td></tr>
</table>

cause the route from one periodic state to another involves an intermediate state which is a mixture of both states. In another set of data from the BZ reaction [6], the following period adding sequence is reported: L^2S^1 to $L^2S^1(L^1S^1)^n$ to L^1S^1, where $n = 1, ..., 7$. We have generated many examples of the above transitions with our mapping. Examination of the $f^n(x)$ versus x mappings, where n was the period of the orbit, revealed that in analogy to smooth, continuous mappings, these transitions proceeded via the route of a "saddle node" bifurcation. Such processes in smooth mappings usually give rise to type 1 intermittency [7]; in our case the "saddle point" can be thought of as being infinitely unstable. The stability of a fixed point is given by the modulus of the slope of the mapping at that fixed point. At the discontinuity our mapping can be approximated by a smooth function with infinite slope Maselko and Swinney also report that an interval in bifurcation space is found where either the L^1S^1 state or the L^2S^2 state may exist, but that a finite perturbation can result in transition from one state to the other. Study of a similar situation produced by our mapping indicates that there are coexisting basins of attraction over a finite interval of bifurcation space.

As we consider smaller and smaller intervals in the bifurcation path, we find states of larger and larger periodicities, sometimes consisting of 100 peaks or more. Usually these large period states are composed of combinations of bordering states. Also, chaotic states are found within the sequence in other parameter regimes both in our simulations and in the experiment [6]. (For the mapping, this occurs whenever the average slope of the orbit is greater than one.) In these cases the staircase is not complete, since the chaotic regions smooth out the behavior of the firing rate. We can constrain our parameters in order to prevent this if we like, but generically we expect that there will be some parameter regions in which choas is mixed in.

The mode locking phenomena and devil's staircase here bear some similarities to that observed in circle maps near the transition to chaos [8]. The suggestive nature of this connection led Maselko and Swinney to call what we have labeled the firing rate the "winding number", in analogy to the number of times an orbit winds around a torus one way as compared to the other way. It is not clear a priori, though, that the devil's staircase seen in critical circle maps has anything to do with that observed here. In fact, there is a very marked difference in the phenomenology, since the complete devil's staircase observed in circle maps is only complete along a critical surface, which is of measure zero in the space of parameters. Picking a curve through parameter space at random will never yield a complete devil's staircase, unless a great deal of luck or premeditation is involved. The devil's staircase seen here, in contrast, is complete over a wide range of parameters. We should note that there are classes of highly nonlinear circle maps, not associated with the transition to chaos, that generate generic complete devil's staircases [9]. Although the connection is not clear, we think it is much more likely that this behavior is related to what we describe here and to the BZ reaction. In particular, we can alter our map somewhat to make it into a circle map. This map continues to have a well defined firing rate, defined as before, but at the same time it is also possible to define a winding number. However, the experimental data from the BZ reaction does not show a relationship between the winding number and the firing rate [10].

We have constructed variations of our mapping which do not have a fixed point and have generated sequences of periodic orbits which produce a new class of mode-locking sequences, which we term "multiple" staircases. These staircases are complete but not monotonically increasing over the bifurcation range. Fig. 3a shows a staircase where a string of firing rates overlaps another; each string of firing rates is characterized by an integer in the numerator. Fig. 3b shows a multiple staircase where there is no overlap between strings, but instead the staircase will monotonically increase for a while, fall to a lower firing rate, and begin to increase again, etc. We note that in both these sequences as well as those from the original mapping, simple ratios produce the longest plateaus in the staircases, which appears to be generic for mode-locking phenomena [11]. Also, the firing rate increases with r converse to the behavior of the mapping represented in figs. 1a and 2. This behavior is analogous to that of the circle map which also lacks an unstable fixed point on the periodic orbit. For the mapping represented in fig. 3b, the Farey relation is always satisfied within an interval of monotonically increasing firing rates [12]. It would be interesting to see if this second type of mode-locking behavior will be experimentally observed.

In summary, if maps of this type do indeed reproduce the dynamics which underly mode locking phe-

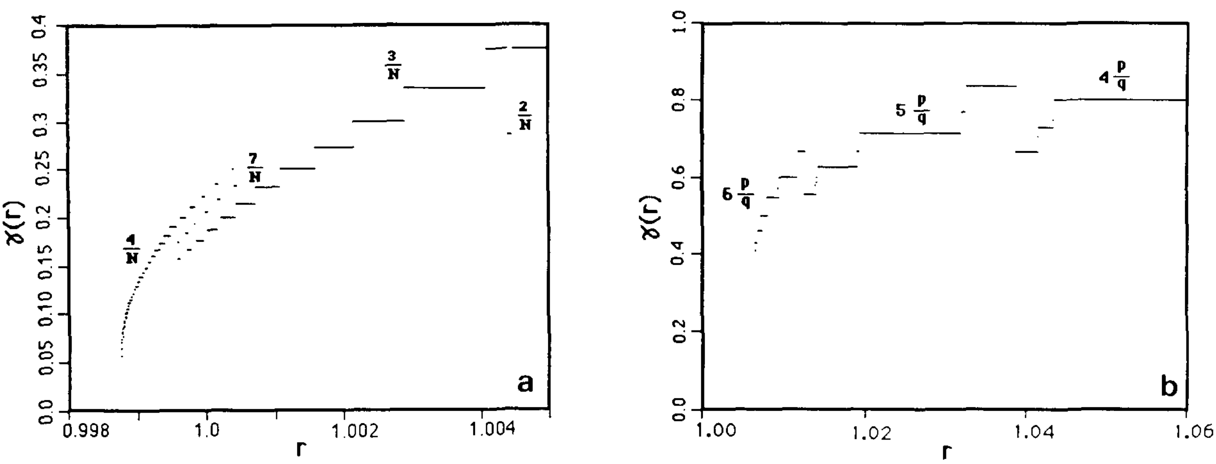

Fig. 3. Two examples of staircases generated from 2 variations of the map in fig. 1b which do not have a fixed point. For (a) the parameter values are: $m_1 = m_5 = 0.7, m_2 = m_3 = -m_4 = 0.9, c_1 = 0.001, c_2 = 0.001, c_3 = -0.003, z_0 = 0.20, z_1 = 0.23, z_2 = 0.77$, and $z_3 = 0.80$. For (b) the parameter values are: $m_1 = -m_5 = 0.42, m_2 = m_3 = -m_4 = 1.5, c_1 = 0.001, c_2 = 0.01, c_3 = -0.173, z_0 = 0.20, z_1 = 0.23, z_2 = 0.77$, and $z_3 = 0.80$. In (b) the complete sequence of periodic orbits is: 18/44, 6/14, 6/13, 6/12, 6/11, 12/21 6/10, 6/9, 5/9, 15/26, 10/17, 15/25, 5/8, 10/15, 5/7, 10/13, 5/6, 4/6, 12/17, 8/11, 12/16, and 4/5.

nomena, then we would expect that some of the properties that we observe here should also occur in experiments. In particular, if parameters other than the flow of rate of x are varied, then it is likely that more complicated behavior will be observed. The possibilities include:

(i) For two given patterns of oscillation, the sequence of patterns linking the two should in general depend on the exact path taken through the parameter space.

(ii) There are likely to be regions where the devil's staircase is smoothed out due to chaos.

(iii) Alternative patterns of mode-locking are likely to occur.

Simple variations of our map can also produce chaos, period-doubling, the U-sequence [13], and the alternation of periodic and chaotic behavior, all of which are observed in other parameter regimes. We emphasize, though, that we by no means consider that the work reported here constitutes a proper physical explanation of the phenomena seen in the BZ reaction. We only intend to give one possible geometry that is capable of qualitatively reproducing the experimental results. Presumably the correct answer can be gotten directly from a system of equations describing the BZ system. By working with equations directly motivated by the chemistry, we are attempting to derive a map similar to that studied here from first principles.

It is our feeling that the mode-locking phenomena described here are the natural result of the interaction between an oscillator and an excitable system with several steady states. As suggested by Koppel and Ermentrout [14], the BZ reaction can also be viewed as an excitable system. In the future we hope to make the connection between our family of mappings and continuous systems of this type.

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