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Gerold Baier and Sven Sahle

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# Hyperchaos and chaotic hierarchy in low-dimensional chemical systems

Gerold Baier and Sven Sahle
*Institute for Chemical Plant Physiology, University of Tübingen, D-72076 Tübingen, Germany*

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Chemical reaction chains with feedback of one of the products on the source of the chain are considered. A strategy is presented in terms of ordinary differential equations which creates one, two, and three positive Lyapunov exponents as the finite dimension of the system is increased. In particular, a nonlinear inhibition loop in a chemical reaction sequence controls the type of chaos. The bifurcation scenarios are studied and chaos and hyperchaos are found for broad regions of bifurcation parameter. Some implications for the occurrence of higher chaos in real systems are discussed.

## INTRODUCTION

The notion of hyperchaos was introduced by Rössler.[1] He considered the possibility of more than one positive Lyapunov exponent (LCE), i.e., more than one direction of mean exponential divergence for attractors in dynamical systems. He then argued that a system with $N$ state variables may possess at most $(N-2)$ positive LCEs and introduced the idea that a hierarchy of chaotic behavior can be realized in principle in systems with many variables, e.g., chemical reaction networks.[2] Hyperchaos was observed numerically in the Mackey–Glass equation by Farmer[3] and in a limit cycle system with delayed feedback by Schell and Ross.[4] Both systems use continuous time delays and are infinite-dimensional dynamical systems. Farmer showed that an increasing number of positive LCEs can occur in such systems when the delay time is increased. Here, we show how this can be realized in autonomous homogeneous reaction systems with few variables.

Hyperchaos and the chaotic hierarchy can easily be modeled in discrete maps (i.e., explicit models of cross-sections of attractors) in two different ways: One can either couple chaos-producing map units of the logistic type (or any unimodal map with chaotic behavior under iteration); or one can instead linearly delay the nonlinear variable of a unimodal map with chaotic behavior. This second way is a means to generate higher-dimensional chaos in diffeomorphisms with only one nonlinear term.[5] We demonstrate that a similarly simple mechanism works for an abstract ordinary differential equation as well. We translate this result into a chemical context. We study bifurcation scenarios and attractors for chemical chaos and hyperchaos in the system with nonlinear feedback and discuss implications of the proposed mechanism for catalytic reaction networks.

### A prototype equation

We introduce the $N$-dimensional system of ordinary differential equations

$$\begin{aligned} \dot{x}_1 &= -x_2 + a(1-x_N^2), \\ \dot{x}_i &= x_{i-1} - x_{i+1}, \qquad (1) \\ \dot{x}_N &= x_{N-1} + b x_N(1-x_N^2) \end{aligned}$$

with $x,a,b \in \mathbf{R}$, $a,b \geqslant 0$, and $i=2,\ldots,N-1$, $N \in \mathbf{N}$. For parameter $a=0$ the system is an $N$-dimensional chain of harmonic oscillators with a nonlinear dissipation function governed by dissipation parameter $b$. The dissipation function causes instability of the fixed point at the origin and creates an attracting limit cycle. The quadratic function governed by parameter $a$ is the chaos-producing nonlinearity. Here, we keep parameter $b=0.2$ constant and study the bifurcation behavior of the limit cycle in Eq. (1) as parameter $a$ is varied.

With $N=3$, a period-doubling sequence from limit cycle to chaos and chaos is found as $a$ is increased from 0 to $a \approx 0.829$ (compare the qualitatively similar bifurcation diagram Fig. 6 in Ref. 6). For instance, spiral chaos is obtained at $a=0.8$ and screw chaos at $a=0.825$.

With $N=4$ and $N=5$, the bifurcation diagram becomes more complex and, in contrast to the three-dimensional system, a chaotic attractor with Lyapunov dimension larger than 3 can occur. For example, at $a=0.68$ the spectrum of Lyapunov characteristic exponents (LCEs in bits/time unit) for $N=4$ is (0.158, 0, −0.126, −0.766). This spectrum fulfills the condition $\lambda_1+\lambda_3>0$ and leads to a Lyapunov dimension $3<D_L<4$ according to the Kaplan–Yorke formula.[7]

With $N=6$, hyperchaos occurs in Eq. (1). Figure 1 shows a bifurcation diagram of the cross sections of attractors obtained as $a$ is *decreased* from 0.7 to 0.4. In this case the sequence hyperchaos→chaos→locking→ quasiperiodicity→ limit cycle is observed. This is similar to the sequence in the three-dimensional diffeomorphism of the delayed Hénon map (see Fig. 15 in Ref. 6) except that in the present case the period-four cycle loses stability via torus-bifurcation as $a$ is increased. However, the dynamics of the six-variable ODE is more complex. It possesses coexisting attractors in the region of quasiperiodicity and of the period-four cycle of Fig. 1 as can be demostrated in bifurcation diagrams obtained for *increasing* parameter $a$ adiabatically in the same range as Fig. 1. For $0.6<a<0.7$, however, hyperchaos is the only finite attractor around the origin. For $a=0.65$, the four largest LCEs are (0.077, 0.027, 0.000, −0.038). As can be seen from the LCEs the Kaplan–Yorke transition occurred for one of the negative exponents yielding a Lyapunov dimension of $4<D_L<5$ for the attractor at $a=0.65$. This type of behavior cannot be described by a 3D diffeomorphism. Note that the difference between the cases $N=3$ and $N=6$ in Eq. (1) is the fact that the nonlinear feedback from $x_N$ to $x_1$ is



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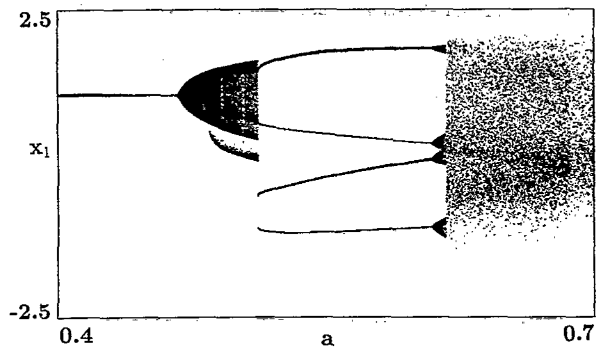


FIG. 1. Bifurcation diagram of Eq. (1) with $N=6$. Cross-section points of variable $x_1$ are plotted whenever a maximum in variable $x_3$ was reached. Plot started at $a=0.7$ and parameter $a$ was decreased adiabatically.

simply delayed by three more linear variables in the 6D case. Neither a new nonlinearity nor any new expanding term have been added. Thus the same mechanism of delayed feedback can lead to chaos and hyperchaos, depending only on the size of the delay chain.

The number of positive Lyapunov exponents for attractors in Eq. (1) can be increased further. With $N=10$, it is easy to find chaos with three positive LCEs. As in the preceding cases, the attractors are periodic for small $a$ and chaos appears as $a$ is increased. For $a=0.54$ we calculated the five largest LCEs to be (0.030, 0.017, 0.005, 0, −0.007). Thus, in principle, the differential equation works to some extent similar to the delayed Hénon map: increasing delay of the chaos-producing nonlinearity generates more independent directions of stretching and folding in phase space. In the case of the ODE, however, the creation of one more positive LCE cannot be achieved by a single additional delay variable as in the map.

Equation (1) possesses a rich variaty of dynamics and is prototypic due to its simplicity of construction. For the present purpose it is sufficient to understand that the mechanism of delayed nonlinear feedback can yield an increasingly complex dynamics as the number of delay variables is increased. This can now be formulated in terms of a chemical reaction network.

## A chemical equation

To design an equation which describes a sequence of chemical reactions we need to introduce a chaos-generating nonlinearity. We first study the iteration scheme:

$$x^{k+1}=Ax^k/(B+(x^k)^C) \tag{2}$$

with $x,A,B\in\mathbf{R}$, and $C\in\mathbf{N}$.

This is a map with a single maximum in $0<x<\infty$. Variable $x^k$ is mapped to $x^{k+1}$ linearly with slope $A/B$ for $x^k\to 0$, and hyperbolic decay towards zero for large $x^k$ when $C>1$. In order to find the universal properties of a transition to chaos it is requested that the fixed point at the origin is unstable (i.e., the derivative at this point must be larger than 1), and that the fixed point in the positive quadrant undergoes a subharmonic bifurcation (i.e., derivative equal to $-1$) as a parameter is varied. Both requirements can be fulfilled if $C>2$. In general, the larger exponent $C$ is, the larger parameter $B$ may be chosen. For instance, when $C=10$ and $B=1$, Eq. (2) shows a period-doubling cascade to chaos as parameter $A$ is increased, followed by a broad range of chaos with comparatively small periodic windows. Thus the rational function in Eq. (2) is a suitable chaos-producing nonlinearity.

This one-dimensional map is noninvertible and must be embedded in a two-dimensional diffeomorphism to be a valid model for a cross section of a flow. Similarly as for the Hénon map, a $D$-dimensional diffeomorphism can be constructed which implements the idea of delayed feedback:

$$x_1^{k+1}=Ax_{D-1}^k/(B+(x_{D-1}^k)^C)+(-1)^{D-1}\cdot bx_D^k,$$
$$x_i^{k+1}=x_{i-1}^k \tag{3}$$

with $x,A,B,b\in\mathbf{R}$, $C,D,k\in\mathbf{N}$, and $i=2,\dots,D$.

The map Eq. (3) produces the whole discrete chaotic hierarchy.[4] As an example, with $D=3$ a folded towel attractor [hyperchaos with LCE spectrum $(+,+,-)$] is found for $A=1.5$, $B=1$, $C=10$, where $B$ and $C$ were chosen as in the Mackey–Glass delay differential equation which uses the same function to generate chaos.[3] Because Eq. (3) is a diffeomorphism it is in principle possible to find equivalent dynamical behavior generically in flows.

From the map Eq. (2) and the results for the abstract system Eq. (1) we construct the following set of differential equations:

$$\frac{dx_1}{dt}=\frac{Ax_N}{1+x_N^C}-k_1x_1,$$
$$\frac{dx_2}{dt}=k_1x_1-k_2x_2x_3,$$
$$\frac{dx_i}{dt}=k_2x_{i-1}x_i-k_2x_ix_{i+1}, \tag{4}$$
$$\frac{dx_N}{dt}=k_2x_{N-1}x_N-k_1x_N$$

with $i=3,\dots,N-1$.

This set of equations describes a sequence of reactions from species $x_1$ through $x_N$. The inlet of compound $x_1$ is nonlinearly regulated by variable $x_N$. The nonlinear function is chosen as in the map Eq. (2). This is the chaos-generating nonlinearity. There is a linear first-order reaction leading from $x_1$ to compound $x_2$. Variables $x_2$ through $x_N$ form the delay chain of length $(N-1)$. The delay chain was formulated as a sequence of autocatalytic reactions and contains elements of the Lotka–Volterra equation for predator-prey interaction. The Lotka–Volterra system is a nonlinear transformation of the harmonic oscillator into the positive quadrant of phase space. The delay chain of the chemical system in Eq. (4) is thus implemented in accordance with the linear delay in Eq. (1). The beginning and the end of the chain are assumed to be first-order reactions and the whole system is dissipative. For the parameters chosen the delay chain without feedback possesses an attracting focus as stable solution. In the simulations discussed here $k_1=0.1$, $k_2=0.2$, $C=10$, and $A$ is used as a bifurcation parameter.

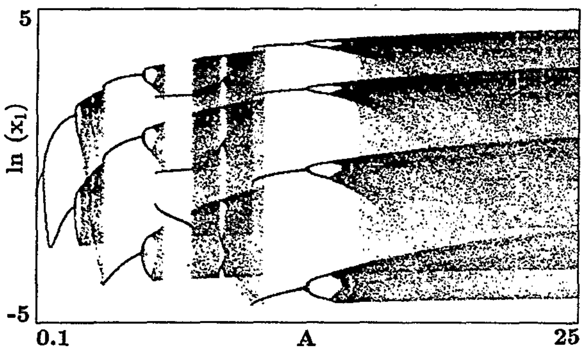


FIG. 2. Bifurcation diagram of Eq. (4) with $N=4$. Cross-section points are taken at maxima of variable $x_3$. Plot started at $A=0.1$ and parameter $A$ was increased adiabatically.

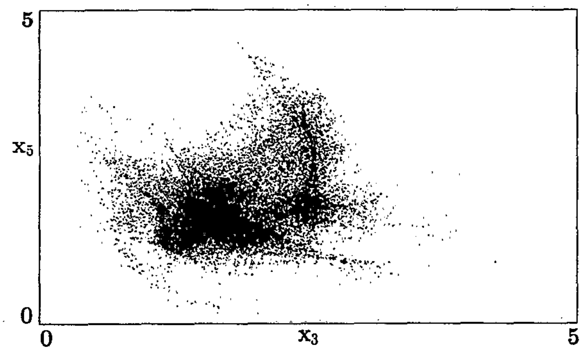


FIG. 4. Poincaré cross-section of hyperchaotic attractor in Eq. (4) with $N=9$ at $A=1.0$. Cross-section taken at maxima of variable $x_8$.

For $N=3$ the system shows limit cycle oscillations over a wide range of parameter $A$. Figure 2 shows the bifurcation diagram for the reaction system with $N=4$. It can be seen that Eq. (4) is successful in terms of chaos. Wide ranges of parameter $A$ display chaotic behavior separated by windows with periodic solutions. Period-doubling cascades to chaos can be seen to originate in limit cycles of period two, three, and four. The respective chaotic regions consequently show two, three, and four overlapping bands in the density distribution of cross-section points in Fig. 2. The corresponding cross sections in three dimensions possess two, three, and four branches of folded lines. For the period-three cycle originating from a crisis and the period-four cycle we find numerical evidence for bistability of the periodic cycle with chaos (left end of either window in parameter space in Fig. 2). Chaotic behavior yielded one positive Lyapunov exponent in all cases tried. For example, we calculated the four LCEs to be (0.010, 0, $-0.064$, $-0.234$), when $N=4$ and $A=2.5$. For $N=7$ the qualitative picture in the bifurcation diagram is similar. Periodic windows born through crisis of a chaotic attractor disappear after a period-doubling sequence to chaos as parameter $A$ is increased. Also, several regions of bistability were detected, and in all cases tried chaotic attractors with only one positive exponent were found.

As dimension $N$ is increased further chaos with two and three positive LCEs can be observed. Figure 3 shows the bifurcation diagram of Eq. (4) with $N=9$. The chosen parameter range (from $A=0.1$ to $A=25$) is widely dominated by chaotic behavior. The system starts with a limit cycle for small values of parameter $A$ and only few periodic windows in the chaotic domain are left as $A$ is increased. Different types of chaos cannot be distinguished from the bifurcation diagram, however. Calculating the spectrum of Lyapunov exponents we found hyperchaos (two positive exponents) on the left side of the chaotic region in Fig. 3. For example, we calculated the four largest LCEs to be (0.015, 0.004, 0, $-0.008$), when $N=9$ and $A=1.0$. Figure 4 shows a Poincaré cross section of the hyperchaotic flow with some foldings discernible in the sheetlike structure. The figure is too distorted, however, to visually distinguish the two independent directions of stretching and folding. Figure 5 shows a grey-coded time series of all variables governed by the dynamics of this hyperchaotic attractor. The members of the delay chain arrange themselves in pairs due to the oscillatory components, otherwise the dynamics is highly irregular. The complexity in this plot arises from the competition of (i) the desire to simply pass the oscillatory information created by variable $x_1$ down the delay line from variable $x_2$ to $x_N$; (ii) the strong nonlinear interference of the actual value of $x_N$ on the production of $x_1$; and (iii) the possibility to propagate information of oscillatory states backwards through the autocatalytic loops.

For $N=12$ the system possesses attractors with three positive LCEs, i.e., three directions of stretching and folding.

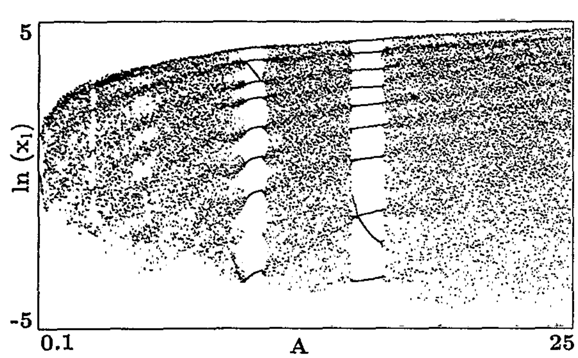


FIG. 3. Bifurcation diagram of Eq. (4) with $N=9$. Cross-section points are taken at maxima of variable $x_6$. Plot started at $A=0.1$ and parameter $A$ was increased adiabatically.

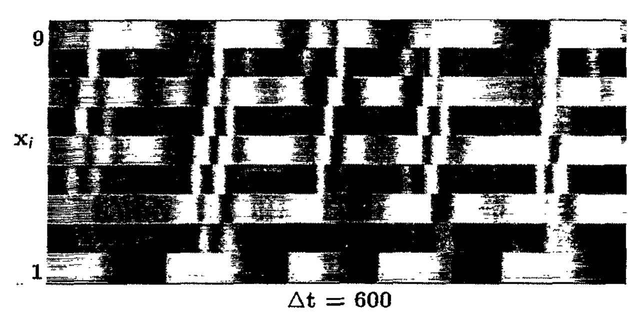


FIG. 5. Time evolution of nine variables on hyperchaotic attractor in Eq. (4) with $N=9$ at $A=1.0$. Grey coding of logarithms of variable values from black ($-7$) to white (3).

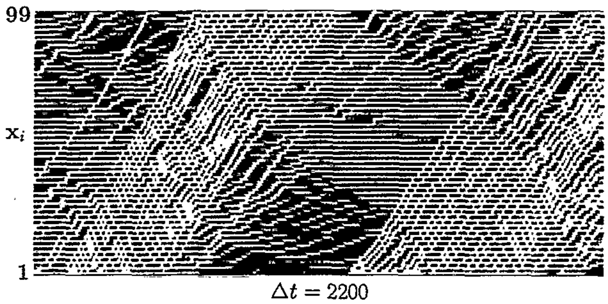


FIG. 6. Time evolution of all variables on attractor in Eq. (4) with $N=99$ and $A=1.0$. Grey coding of logarithms of variable values from black ($-7$) to white (3).

As an example, for $A=1$ the five largest LCEs were calculated to be (0.014, 0.008, 0.003, 0, $-0.002$). Calculating the LCE spectra of our system for further increase of $N$ as $A$ is held constant we make similar observations as Farmer who reports increase of number of positive LCEs and decay of the value of the largest LCE as the delay time is increased in the Mackey–Glass equation.[3] For all $N$ investigated the reported behavior was not altered qualitatively when parameters $k_1$ and $k_2$ deviated slightly from the given values; when slightly nonequal values were assigned to the autocatalytic steps; and when small dissipative terms (first-order reactions) were added to each chain member. Finally we present a simulation of a higher-dimensional case of Eq. (4). Figure 6 is a plot of the time evolution of all variables in Eq. (4) with $N=99$. Transients were allowed to die out and the system had settled to an attractor at the beginning of the displayed time series. One can see that a large spike (coded white) in a variable with low number is passed along the delay chain from member to member until it reaches $x_N$. Spikes in variable $x_1$ occur irregularly. In particular, there are bundles with many spikes separated by calm periods with low average value of $x_1$ (middle of Fig. 6). Propagation of spikes is not parallel, however. Rather, propagating spikes are slowed down at several points along the chain. It turns out that at these points they are interfering either with other spikes in neighboring variables or with oscillations that move "backwards," i.e., start at $x_N$. Even though we have not yet explicitly analysed all higher-dimensional cases of Eq. (4) it appears that the trends found at low values of dimension $N$ continues as $N$ is increased further.

To test the applicability of our results under less rigid assumptions we numerically investigated a random net based on Eq. (4). For this purpose we kept variables $x_1$, $x_2$, and $x_N$ as in Eq. (4) and choose an arbitrary number of net variables, e.g., 50. Then, an arbitrary number of autocatalytic reactions were arranged at random among net members and rate constants were assigned randomly within a given range. It was made sure that the net was connected to variables $x_2$ and $x_N$. Numerical details will be reported elsewhere, however, for the present context the following observations can be mentioned: Nonequilibrium self-organized behavior is found frequently. Among the attractors observed are limit cycles, $n$-tori, chaos and a large variety of attractors which require higher-dimensional embedding. Also occurring is intermittent behavior where laminar phases alternate aperiodically with irregular bursts. We also observed attractors where the trajectory switched between different chaotic subregimes in phase space. Due to the complexity of phase space we also frequently recorded long-lived chaotic transients for initial conditions near basin boundaries or far from the final attractor. The most important feature was that the probability of finding a high-dimensional attractor (a criterion being $D_L>3$ for the Lyapunov dimension $D_L$, for example) decreased dramatically when, under otherwise identical conditions, the loop with nonlinear feeback from variable $x_N$ to $x_1$ was omitted.

## DISCUSSION

Examples of hyperchaos in chemical ODEs have been mostly the result of searches in parameter space of nonlinear systems. Killory *et al.* found hyperchaos numerically in a system of equations describing mass-action kinetics that was derived from Rössler's original model.[8] Badola *et al.* saw hyperchaotic behavior in weakly coupled chaotic subsystems for a finite range of parameter.[9] Apart from weakly coupled chaotic subsystems a model of bacteria-phage interaction suggested that higher-order chaos and the chaotic hierarchy could be generated by coupling of oscillatory components to a common source.[10] Now we successfully exploited the idea that a single chaos-producing mechanism can be used to generate steps of the chaotic hierarchy as well. An oscillating signal is fed back with a finite number of delays. The delay variables may even be trivially behaving. Best results were obtained when using a set of delay-variables with a focus, either unstable as in Eq. (1) or stable as in Eq. (4). The feedback function was either a simple square term or a single-humped rational function of a delayed variable. Both functions have been shown to produce the chaotic hierarchy in maps. This suggests that, in principle, it is possible to generate all kinds of (hyper)chaos from a single nonlinear feedback function. The proposed reaction scheme is to our knowledge the first explicit candidate for a generic finite-dimensional chaotic hierarchy in reaction kinetics.

Schell and Ross explain the occurrence of hyperchaos with the fact that for increasing time delay real parts of eigenvalues of an unstable steady state in their system successively become positive and they assume that at least some of these new locally unstable directions result in a globally positive LCEs.[4] In Eqs. (1) and (4) new unstable directions around an unstable steady state are created as $N$ is increased, but no analytic knowledge of the global flow is available to make predictions about the qualitative behavior of the LCE spectrum.

It will now be necessary to compare hyperchaos with one source of divergence [as in the present examples Eq. (1) and Eq. (4)] to hyperchaos with two sources of divergence (as in the Rössler equation[1]) in terms of chaos control. Preliminary results by Pyragas suggest that in the chaos of Eq. (1) it is sufficient to add only one control term to one of the system's variables whereas in the Rössler equation two variables have to be controlled in order to be able to stabilize unstable periodic orbits of the system.[11] If this can be explained theoretically chaos control could provide a means to distinguish different mechanisms of hyperchaos generation.

Both the spectrum of Lyapunov exponents and the estimated metric entropy seem to behave similar to the Mackey–Glass equation as the delay is increased.[3] Similarly, Schell and Ross observed the transition from chaos to hyperchaos as the delay time was increased.[4] They argued that equivalent results can be expected in oscillatory biochemical reactions with imposed time delays. In a sense, Eq. (4) could be viewed as a finite-dimensional version of an infinite-dimensional delay differential equation. Our approach indicates that indeed a high-dimensional dynamics can be realized in a *finite* biochemical or physiological control chain with nonlinear feedback inhibition. The fact that all delay steps are autocatalytic in our artificial example Eq. (4) might appear as a severe limitation. However, this formulation has been chosen to construct the system in accordance with Eq. (1). As argued by Rössler[12] autocatalysis is a special case of the generalized autocatalytic set which is much easier to implement and can be expected to appear much more frequently in a biochemical context. For example, the reaction sequence $A+X \rightarrow 2Y$, $Y \rightarrow X$, which appears in the mechanism of the Belousov–Zhabotinsky reaction, is a member of the same set. These more realistic models have yet to be investigated.

Experimentally autonomous hyperchaos was demonstrated in a semiconductor experiment by Stoop *et al.*[13] The first observation of two positive LCEs in a chemical experiment was in a time series from the catalytic oxidation of CO on the (110) surface of a platinum single crystal.[14] In both experiments, however, spatial degrees of freedom have to be considered in modeling and the connection between spatial structure and temporal hyperchaos is open at present.

For dimension $N$ going to infinity in Eq. (1) we expect high-dimensional attractors with many LCEs close to or equal to zero. In this case many directions of weak divergence can no longer be distinguished from a high-dimensional attracting hypertorus. Of special interest is the case where the chain of $x_i$ can be interpreted as approximation to a spatial direction, e.g., the transport system in Eq. (1). Then the transition to $N=\infty$ would correspond to a one-dimensional partial differential equation with highly complex space-time behavior. This result seems to converge to Landau's picture of turbulence as an interaction of many oscillatory modes.[15]

We have demonstrated the occurrence of chaos and hyperchaos in low-dimensional chains of the chemical system Eq. (4). Furthermore we showed an example that the trend of increasing the number of positive Lyapunov exponents continues as the number of chain members increases. In random networks composed of reactions as considered in Eq. (4) self-organized high-dimensional dynamics appeared with high probability. In contrast, high-dimensional attractors were found with very low probability when the feedback regulation of the input to the network was missing. We conclude that it is not sufficient to simply couple many variables nonlinearly at random, and not even sufficient to introduce many autocatalytic steps to generate robust high-dimensional attractors. It seems important to add a proper chaos-supporting strategy. Nonlinear delayed feedback is such a strategy in chemical systems. It is likely that the present results can be formulated in terms of other systems as well, for example in the dynamics of biological neural nets.

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