Chemical Turbulence:
Chaos in a Simple Reaction-Diffusion System

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Nonperiodic oscillation ("chaos"), formerly found in several 3-variable homogeneous abstract reaction systems, is also possible in 2-morphogen compartmental systems. In a 2-cellular (and hence 4-variable) symmetrical morphogenetic system of Rashevsky-Turing type, a nonperiodic return toward an (almost) undifferentiated state is observed under numerical simulation. The system hereby shows a novel, "bi-chaotic" (rather than bistable), type of behavior. The 2 chaotic regimes are of the screw type each. They are separated by a symmetrical saddle-limit cycle. This behavior is preserved under a "contraction" of the system to 3 variables. A second bichaotic mode (2 spiral-type chaotic regimes separated by a symmetrical steady state) is also possible. A preliminary result on a third of chaos is also presented. Thus, "turbulence" (or chaos) may be a general behavioral possibility of interaction-type morphogenetic systems.

Introduction

Nonperiodic oscillation [1] or chaos [2] was, in a simple differential system, first observed by Lorenz [1] in his model of turbulence. Ruelle and Takens [3] later claimed applicability of the abstract dynamical theory of horseshoe maps [4] to turbulence generation. Then an irregular "meandering" was observed in the dish, by Winfree [5], who studied the well-known Belousov-Zhabotinsky reaction in a modified version under nonstirred conditions [6]. Following Winfree's question whether this may be chaos, several 3-variable homogeneous abstract reaction systems producing nonperiodic oscillations were found [7, 8]. Hereby two prototypes of chaotic flow in state space, screw type chaos, and spiral type chaos, were distinguished [8]. A simpler analog to the Lorenz equation, being of single, rather than double, spiral type and containing just one (instead of two) nonlinearities of second order, has also been described [9]. Most recently, a class of 4-variable, symmetrically-built Hamiltonian systems (of Hénon-Heiles [10] type) has been shown to be chaos-producing by detailed analysis of the (locally) 3-dimensional state space in which a modified horseshoe map was found [11]. In these systems, the coupling between the 2 nonlinear sub-oscillators contained is of a highly nonlinear type.

Based on the idea that a chain of cross-inhibiting identical nonlinear oscillators should be capable of

producing a "boiling-like" type of chaos — in analogy to the cross-inhibitory recurrent formation of vapor bubbles in a saucepan of boiling spinach sauce —, the suspicion has been uttered earlier [8] that the well-known morphogenetic systems of Rashevsky-Turing type (which do consist of cross-inhibitorily coupled, potentially oscillating subsystems) may be capable of producing a chaotic type of morphogenesis.

In the following, some numerical evidence substantiating this suspicion is presented.

The Equation

Symmetry-breaking morphogenesis was apparently invented three times: Poincaré [12] observed it in the equations of a rotating star (coining the term "bifurcation" for the phenomenon); Rashevsky [13] proposed a simple reaction-diffusion-transport equation as a model to explain the sudden emergence of spatial "polarity" in a formerly symmetrical cell; and Turing [14] observed a "breakdown of symmetry" in a multi-cellular (in the simplest case, 2-cellular) "morphogenetic" system. Following the four sets of equations indicated by Turing (on pp. 43, 61, 64, and 65 of Ref. [14]), several more examples have been described in literature [15, 16]. The Gierer-Meinhardt equations [16] are of the same type, but were invented independently as concrete models of hydra morphogenesis. Turing-type equations are of interest, therefore, for mathematical, chemical, and biological systems.

In an earlier communication [17] it has been shown that Turing's simplest equation (described only ver-

bally on pages $42-43$ of his paper [14]) can be simplified further (by omitting a constant influx to the second variable and setting the diffusion-coefficient for the less diffusible first substance equal to zero as a matter of idealization). The single switching-type nonlinearity contained in Turing's equation (namely the term $-6\,b\cdot\{{}^1_0\text{ if }a\gtrless0\}$) could be replaced by a (chemically more convenient) Michaelis-Menten term: $-6\,b\cdot a/(a+K)$, where $K\to0$ [17]. Using these conventions, Turing's original equation [14] becomes

$$\dot a = k_1{}' a - k_2\,b\;\frac{a}{a+K}\;+k_5$$
$$\dot b = k_3\,a - k_4\,b + D\,(b'-b) \qquad (1)$$
$$\dot b{}' = k_3\,a' - k_4\,b' + D\,(b-b')$$
$$\dot a{}' = k_1{}'\,a' - k_2\,b'\;\frac{a'}{a'+K}\;+k_5\,,$$

where a, b, a', $b'=$ concentrations of the two morphogens A and B in compartment 1 (unprimed) and 2 (primed), respectively; $k_1{}'=k_1-k_3$, with $k_1>k_3$; $K=$ a phenomenological Michaelis-Menten constant; $D=$ diffusion (or rather, permeability) coefficient for B. Only two "cells" are considered. Well-stirredness, an appropriate concentration range, and fast relaxation of intermediate substances, are presupposed as usual. Turing's original constants (omitting two, as mentioned) were: $k_1{}'=5$, $k_2=k_3=6$, $k_4=7$, $k_5=1$, $D=4.5$, and $K\to0$ (as described above).

A First Simulation Result

Figure 1 gives the simulation result for one particular set of parameter values (which is not too critical). All six side-views of the flow of trajectories in 4-dimensional state space are displayed. The first side-view (Fig. 1 a) is the most instructive. It shows irregularly recurring "differentiation" between A and A': Whenever the trajectory returns toward the symmetrical unstable limit cycle from the side and below, it is going to be pushed away again (much like the flow in a water vessel containing a magnetic stirrer at the bottom). Figure 1 b gives a view of the "active" one of the 2 sub-oscillators, while Fig. 1 d represents the "passive" cell. The other side-views do not carry much additional information. Figure 2 gives the time course of the 4 variables.

It is evident that under a symmetrical change of initial conditions, the primed and the unprimed variables will simply change their roles. It is also clear that whenever the system has returned toward the neighborhood of the symmetry axis for a while (see Fig. 1 a, c), its state might easily be pushed over toward the other side (for example, in the presence of irregular exogeneous perturbations, or after the introduction of a slight parametric asymmetry).
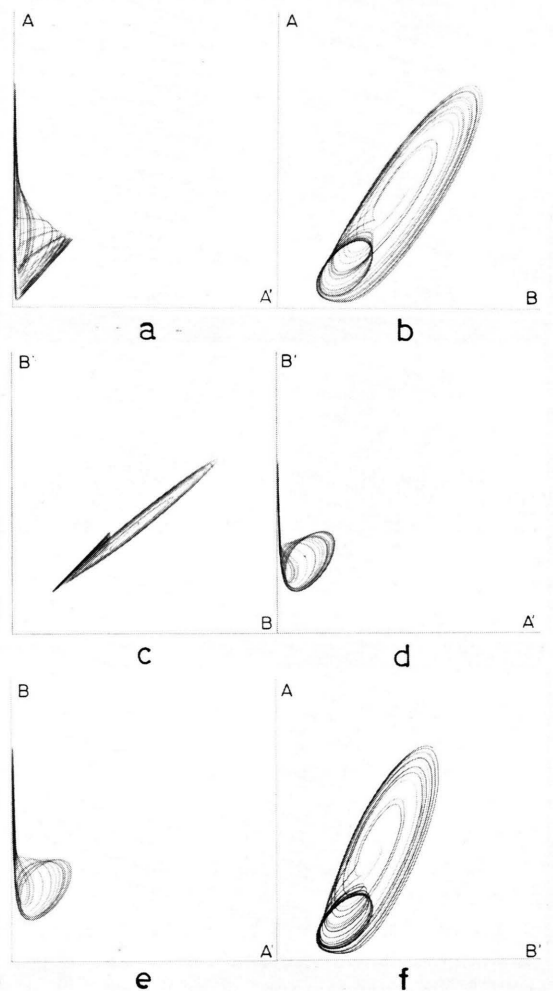

Fig. 1. Apparently nonperiodic flow in a Rashevsky-Turing equation [Equation (1)]. $\mathbf{a-f}$: six side-views of the 4-dimensional flow. Parametric values assumed: $k_1{}'=4.8$, $k_2=k_3=6$, $k_4=3$, $k_5=1$, $K=0.03$, $D=12$. Initial conditions: $A(0)=0.75$, $B(0)=B'(0)=0.6$, $A'(0)=0.005$ concentration units; $t_{\mathrm{end}}=64.4$ time units. Axes: $0\ldots1.5$ for all 4 variables. Numerical simulation performed on a HP 9820 A calculator with peripherals, using a standard Runge-Kutta-Merson integration routine (adapted by F. Göbber).
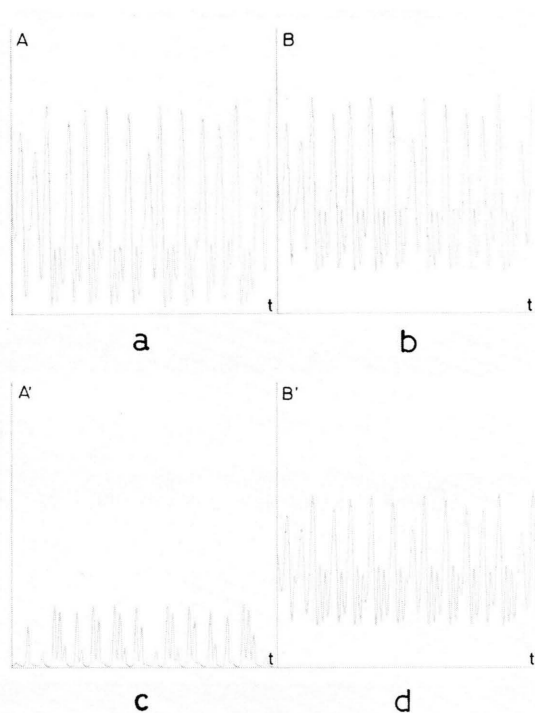
Fig. 2. Time-behavior of the 4 variables of Figure 1. $t(0) = 0$, $t_{end} = 64.4$ time units.

Discussion of Qualitative Behavior

Numerical evidence is never sufficient for the demonstration of truly nonperiodic behavior (for $t \to \infty$). As for chaotic systems governed by a 1-dimensional horseshoe map (called hair-pin map [8]), no matter whether they are discrete [2, 18] or continuous [7], it is well-known that they virtually always tend toward a periodic global attractor in the long run (cf. the review by May [19]). However, it is also well-known [19] that this means little from a practical point of view since (a) the period is usually very long, and (b) extremely small perturbations are sufficient to prevent the global attractor from being reached in most cases.

The situation is more favorable with 2- and higher-dimensional horseshoe maps. For systems governed by such maps it seems that truly nonperiodic behavior is possible for finite domains in their parameter spaces [20]. Thus, in order to make sure that the time course of Fig. 2 has a finite probability to be part of a truly nonperiodic oscillation, it will suffice to show that the flow of Fig. 1 actually admits a 2- or higher-dimensional cross-section of horseshoe type.

Unfortunately, this is very hard to do for 4-dimensional flows (cf. [11]). The 4-variable systems of Ref. [11] have, unlike Eq. (1), the asset of possessing an integral of motion, so that the dimensionality of their state space is reduced (the state space actually being a 3-manifold). While numerical simulation is an appropriate means to show the presence of a cross-section of horseshoe-type (barring the exclusion of subtleties like local "twists" in the horseshoe) in 3-variable systems [8], it has no similar significance in higher dimensions. A satisfactory understanding of Eq. (1) is, therefore, apparently only possible after putting $D = \infty$, thus reaching a "contracted" 3-variable system. As it turns out, this brute procedure is fully compatible with the preservation of the qualitative properties of the flow of Figure 1. After a slight readjustment of parameters (example: $k_1' = 5$, $k_3 = 3.9$, $k_4 = 1.5$, $K = 0.04$), in addition to $D \to \infty$, a flow much like that in Fig. 1, but with the projection of Fig. 1 c contracted to the first bisector and the projections of Fig. 1 e, f collapsed toward those of Fig. 1 d, b, is obtained. For this flow it is not hard to show that it belongs to the "screw-type" of chaos described earlier [8] as being governed by a horseshoe map. The behavior of this contracted system is interesting in its own right and separate communication [21].

Thus, up to isomorphy with its contracted analog [21], the system of Fig. 1 can be considered as a first example of a bichaotic dynamical system, the 2 chaotic regimes being separated by a saddle-limit cycle.

A Second Type of Chaos in the Same System

In Fig. 3, another simulation of Eq. (1), using a different set of parameter values, is shown. The side-view (Fig. 3 b) now shows the typical "shell-like" picture characteristic of "spiral-type" chaos [8]. Again two symmetrically placed flows are possible. However, there is no longer a symmetrical (saddle-)limit cycle, but only a symmetrical stable (saddle-)focus, just as in Turing's original system.

A better qualitative understanding of the flow can, again, be achieved following the above-described "contraction" procedure. Thus, up to isomorphy with the contracted analog [21], a second type of bichaotic flow has been found for Equation (1).
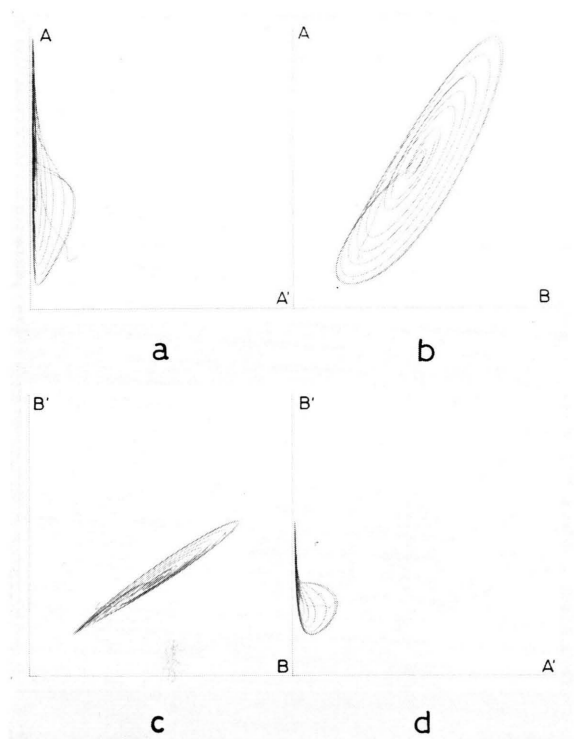
Fig. 3. A second type of apparently nonperiodic flow, obtained for Eq. (1) with a different parameter set. **a—d**: 4 side-views (corresponding to those of Figures 1 a—d). Numerical simulation as in Figure 1. Parameter values: $k_1'=8$, $k_2=6$, $k_3=12$, $k_4=4$, $k_5=1$, $K=0.5$, $D=9$. Initial conditions: $A(0)=0.61$, $B(0)=B'(0)=1.7$, $A'(0)=0.59$ concentration units; $t_{end}=26.2$ time units. Axes: $0\ldots3.5$ for A and A'; $0\ldots7$ for B and B'.

Fig. 4. Numerical evidence for a third type of chaos in Equation (1). **a—d**: 4 side-views (corresponding to those of Fig. 1 a, b, c, and b again). Numerical simulations as in Figure 1. Parameter values: $k_1'=5$, $k_2=k_3=6$, $k_4=3$, $k_5=1$, $K=0.03$, $D=11$. Initial conditions: $A(0)=0.75$, $B(0)=B'(0)=0.25$, $A'(0)=1/15$ concentration units; $t_{end}=25.7$ time units for **a** and **b**, and $t_{end}=130$ for **c** and **d**. Axes$= 0\ldots1.3$ for all variables.

Possible Existence of a Third Type of Chaotic Flow

In Fig. 4, numerical evidence is presented that the 2 screw-type chaotic flows of Fig. 1 can be made to merge "back-to-back", not only under exogenous perturbations (as mentioned), but also spontaneously. The result then is a single globally attracting chaotic regime.

Figure 4 has been included because it seems to reflect a behavioral peculiarity of Eq. (1) observed earlier in a different context [17] already: an intrinsic readiness to "overshoot". In the original, bistable mode, Eq. (1) was found to respond to a certain transient exogenous perturbation by a complete reversal of differentiation [17]. As it turned out later [22], this type of triggering of a bistable system with built-in overshoot, so that it acts as a **T** flip-flop, had been described earlier in an electronic context
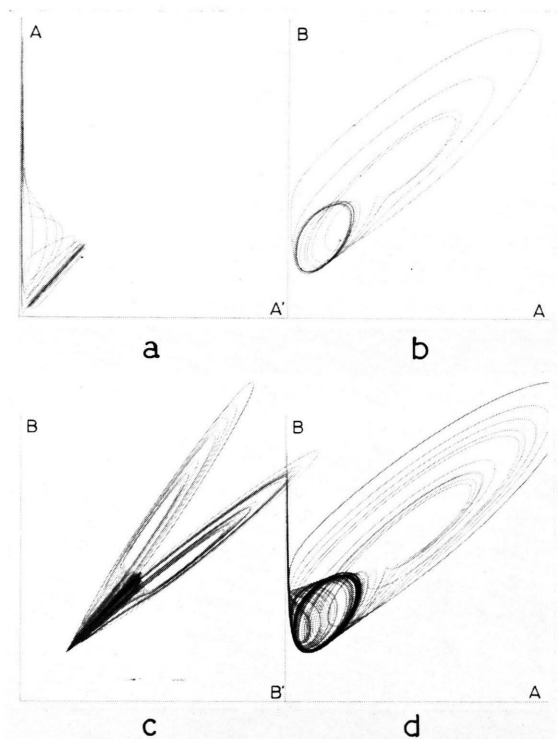
already by von Neumann [23]. Thus, it is conceivable that the additional "half" degree of freedom distinguishing the 4-variable system of Eq. (1) from its 3-dimensional "contracted" analog, is responsible for a sort of "endogenous perturbation" which allows the 4-variable system to behave spontaneously in a way for which the 3-variable system would need an exogenous perturbation.

Nonetheless it must be said that so far, the possibility that the behavior observed in Fig. 4 is due to "numerical perturbation" only, has not been excluded. The easiest way to do this will be to search for a much more robust type of "endogenous overshooting" than the one observed in Figure 4. Analytical counterarguments against such a possibility have not been found so far.

Thus, the interesting behavior of Fig. 4 (interesting, because not only the "amplitude", but also

the "polarity" of differentiation is affected by the nonperiodic oscillation) is nothing but a hypothesis at present.

Discussion

A well-known equation from chemical systems theory [Eq. (1)] turned out to possess a much richer catalog of behaviors than formerly anticipated. If it is true that the system's flow in 4-dimensional state space admits cross-sections of horseshoe type, as has been made plausible above, the system is capable of an infinite number of periodic solutions [4]. Equation (1) then could be ascribed a certain dynamical universality. While such a behavioral complexity is no longer unsuspected with respect to diffusion-coupled 3-variable chemical systems (see [7]), the fact that two variables appear sufficient now, is unexpected.

It is conceivable, therefore, that the "meandering" observed [5] in a reaction-diffusion system of a different (namely cross-activating) type might be explainable also in terms of a 2-variable model (like that of Ref. [24]). For example, it may be interesting to look into the consequences of increasing the slope of the "walls" of the core-region in a 2-variable excitable medium of this type (by decreasing simultaneously the minimum period of the involved monostable relaxation oscillator and the value of the diffusion coefficient). The formerly stable singularity inside the core (cf. [6]) should turn unstable hereby, with "repulsive" effects analogous to those observed in Fig. 1 becoming possible.

The numerical results described above apply to a compartmental system. A necessary next step will be their attempted reproduction in the partial differential equation which directly corresponds to Eq. (1) (cf. [14]). It is well possible that for chaos to occur in this partial differential equation, not all of the constraints present in the system of Fig. 1 (or 3) are required. Thus, an ordinary bi-oscillatory regime (with two limit cycles, rather than chaotic regimes, present) as it occurs in the 2-cellular system over a larger range of parameter values, might be sufficient already. The implications of the mere presence of a symmetrical saddle limit cycle will also be worth studying.

The above presented results endorse the idea that chaos may be a general behavioral potentiality of systems composed of more or less identical subsystems (like the brain, society, etc.). An irregular spiking observed in lasers has already been explained by an equation identical to the Lorenz equation [25]. Thus, chaos may, just like "differentiation", be a behavioral characteristic of "synergetic" systems [26] in general.

As a final remark, the fact that hydra morphogenesis has been explained by morphogen equations very similar to Eq. (1) may be recalled. Thus, there is a finite chance to observe chaos in a concrete biological system someday. It can be conjectured that this (so-called pathological [11]) dynamical behavior will, if occurring in a biological self-differentiating system, correspond to a truly "pathological" type of functioning.

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