Chaotic Behavior in Simple Reaction Systems

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Deterministic nonperiodic flow (of "chaotic" or "strange" or "tumbling" type, respectively) was first observed, in a 3-component differential system, by E. N. Lorenz in 1963. A 3-component abstract reaction system showing the same qualitative behavior is indicated. It consists of (1) an ordinary 2-variable chemical oscillator and (2) an ordinary single-variable chemical hysteresis system. According to the same dual principle, many more analogous systems can be devised, no matter whether chemical, biochemical, biophysical, ecological, sociological, economic, or electronic in nature. Their dynamics are determined by the presence of a "folded" Poincaré map. Under numerical simulation, the proposed chemical system provides an almost ideal illustration to the underlying dynamical prototype, the "3-dimensional blender". Thus, continuous Euklidean dynamics (and with it chemical kinetics) proves to be of equal interest in studying chaos as discrete dynamical systems already have.

Introduction

"Chaotic", or "tumbling" behavior as a qualitative behavioral mode of dynamical systems is known since a long time. Poincaré already observed that not only large ensembles of coupled systems (as in statistical mechanics) may produce the phenomenon, but that 2 strongly coupled nonlinear oscillators may already be sufficient[1]. Later on, numerous treatises on "ergodic" (and "mixing", and "axiom A", and "Anosov", respectively) flows have appeared in continuous dynamics[2–5], rendering the mathematical existence of "strange attractors", as the underlying limit sets have been called[4], a well-established fact. However, the historical origin (2 coupled oscillators, which means 4 state variables) may have been the reason that in search for mathematically simple examples, mostly a non-Euklidean metric has been assumed (since a 2-dimensional torus is the natural surface for treating a pair of oscillators). Hereby the fact that a 2-torus can be re-embedded in Euklidean 3-space was somehow not exploited.

An impulse toward reconsideration of Euklidean systems was provided by E. N. Lorenz's paper[6] on a 3-variable "non-periodic" differential system, derived from a more complicated model of turbulence. However, the mode of action of this system, described by a deceptively simple set of equations, was apparently too complicated in order to lead to the formulation of a simple 3-dimensional prototype directly.

Nonetheless, Lorenz[6, 7] made an important quantitative observation concerning the amplitudes of successive oscillations in his system: when he viewed those amplitudes as being generated by a discrete system, the transition function determining the latter's behavior revealed an interesting "cap-shaped" form (as a two-to-one mapping). Concerning the class of chaos-generating discrete systems opened up by this finding, a number of papers have appeared recently[8] or are in the process of appearing[9–12], whereby a potential ecological application of these equations is emphasized.

Since any continuous oscillator gives rise to a discrete dynamical system governed by a so-called Poincaré map[13] (which describes nothing else than the transition law from one amplitude to the next[13], though usually being considered only in the neighborhood of a limit cycle), it is straightforward to suggest a reversal of Lorenz's procedure: to look for further 3-variable dynamical systems possessing a cap-shaped difference equation as a Poincaré map. (Some of the systems to be detected may then prove to be of a similar practical importance as the derived difference equations already have.)

The particular 3-dimensional flow to be described below was not found in this way, however. In an attempt at "translating" Lorenz's original differential system into the non-negative domain in order

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to arrive at a possible reaction kinetic analog, the mode of action of the anticipated analog (depicted in Fig. 8g) proved so intricate[14] that a simpler mechanism had to be looked for (possibly within the class of chemical universal circuits considered earlier[15]). Only after such a system had been found did its properties suggest the above-named identity (Lorenz map = Poincaré map).

A Principle for 3-dimensional Chaos Generation

In 1930, Khaikin[16] described an electronic device which he called a "universal circuit" since it could produce both nearly linear and typical relaxation oscillations on the turn of a single parameter (with a sharp transition point). As described in Andronov et al. 's well-known textbook[17], the system's trajectorial flow consists of an autonomous oscillation in 2 variables, being molded upon an either ʃ- or S-shaped slow manifold[18] formed by the dynamics of a third variable.

As depicted in Fig. 1, a slight modification is sufficient to turn the device into a chaos-generating machine: by simply introducing a different orientation of flowing on the other stable branch of the slow manifold (with the consequence of a "reinjection" of part of the flow after its having passed through a twisted roundabout loop).

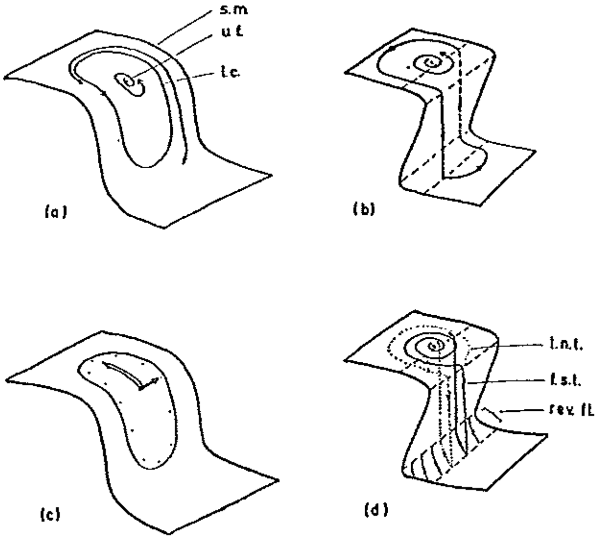

Fig. 1. Main trajectorial flow of a universal circuit. a) Nearly linear mode. b) Relaxation mode. c) Analogous "Soft Watch" (after Salvador Dali's synonymous painting, 1933). d) Chaos-producing mode (see text). s.m. = slow manifold, u.f. = unstable focus, l.c. = limit cycle, the intermediate part of slow manifold in (b) and (d) is unstable, f.s.t. = "first switched trajectory", l.n.t. = "last nonswitched trajectory", rev.fl. = reversed direction of flow "downstairs".

Since this is a very minor modification, the circuit appears to be even more "universal" than originally thought. In addition to chaos-type oscillations (to be considered here), the system also can produce coil-type[19] oscillations (when the width of the hysteresis loop is reduced) and, when used as a morphogenetic system (under diffusion-type coupling), "veined" patterns, as evidenced by a recent model on leaf-morphogenesis[20]. The limits of the circuit's "universality" thus are still undetermined.

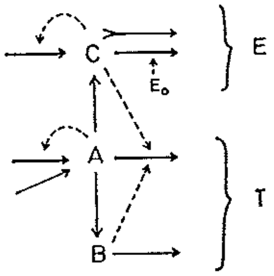

Fig. 2. Combination of an Edelstein switch[22, 23] with a Turing oscillator[24, 23] in a reaction system producing chaos. E = switching subsystem, T = oscillating subsystem; constant pools (sources and sinks) have been omitted from the scheme as usual.

The following reaction scheme (Fig. 2) constitutes one possible way to realize the principle by chemical means. It combines a 2-variable chemical oscillator (variables a, b) with a single-variable chemical hysteresis system (c), as prescribed by the recipe.

The system obeys, under the usual assumptions of wellstirredness and isothermy as well as an appropriate concentration range, the following set of rate equations:

$$\begin{aligned} \dot a &= k_1 + k_2 a - (k_3 b + k_4 c)\, a/(a+K)\,, \\ \dot b &= k_5 a - k_6 b\,, \\ \mu \dot c &= k_7 a + k_8 c - k_9 c^2 - k_{10}\, c/(c+K')\,, \end{aligned} \tag{1}$$

where a denotes the concentration of substance A, etc., $\dot{} = \mathrm{d}/\mathrm{d}t$, $k_{10} = k_{10}'\, e_0$, $e_0 =$ constant, and K, K' are Michaelis constants. The equations thus are non-explicit, assuming validity of a steady-state approximation of fast-reacting intermediate products[21].

A simulation result is shown in Figure 3. It may be noted that due to the asymmetry of the slow manifold (cf. Fig. 3b), only one of its two thresholds is effective at the assumed, relatively low value

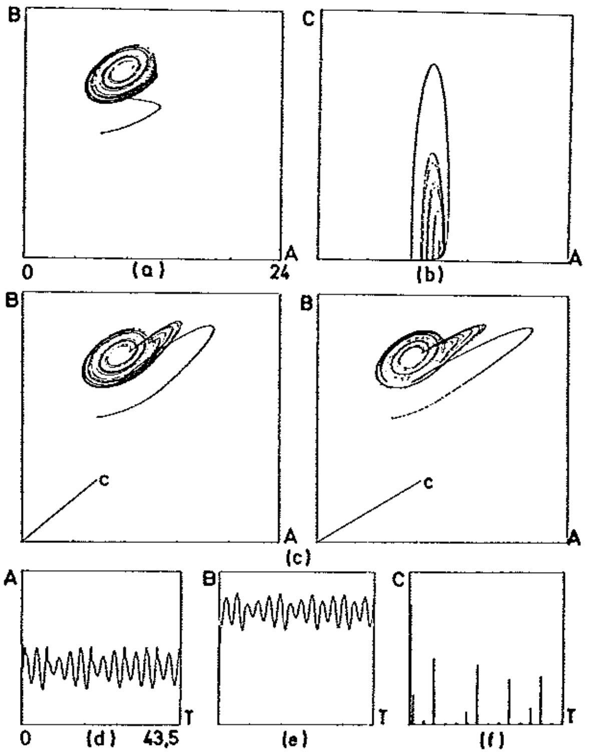

Fig. 3. Numerical simulation of Eq. (1), using Gear's integration method for the numerical solution of stiff differential equations[25]. a) a/b-projection. b) a/c-projection. c) Stereoplot. (Parallel projection; the right-hand picture is for the left eye; c is pointing out of the paper.) d) Time behavior of a. e) Time behavior of b. f) Time behavior of c. Parameters assumed: $k_1 = 37.8$, $k_2 = 1.4$, $k_3 = 2.8$, $k_4 = 2.8$, $k_5 = 2$, $k = 1$, $k_7 = 8$, $k_8 = 1.84$, $k_9 = 0.0616$, $k_{10} = 100$, $K = 0.05$, $K' = 0.02$, $\mu = 1/25$; $a_0 = 7$, $b_0 = 12$, $c_0 = 0.2$, $t_0 = 0$, $t_{\text{end}} = 43.51$.

of μ. (Therefore, k_9 could as well have been set equal to zero, rendering the Edelstein switch a non-resettable, or single-threshold, chemical switch[23, 26].) The "reinjection principle" as postulated above is nonetheless perfectly valid, as evidenced by the "down-view" (Fig. 3 a) as well as the stereo-plot (Figure 3 c). Both the time behavior of the 3 variables and the apparent relatively homogeneous covering of a whole region of state space by trajectories suggest absence of a limit cycle of low period. The qualitative properties of the flow cannot be deduced from simulation results alone, however.

Existence of a Chaos-generating Poincaré Map

In Fig. 1a, a one-dimensional Poincaré map[13] can be constructed along a radius emerging from the unstable focus and staying within the stable manifold, supposed that μ is tending to zero. The same holds true for Figure 1 b. In either case, the Poincaré map has the form indicated in Figure 4. It is identical with the Poincaré map of a simple 2-dimensional limit cycle oscillator.

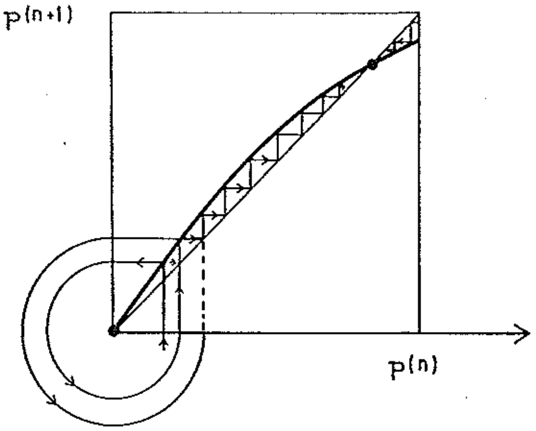

Fig. 4. Poincaré map of a universal circuit in the nearly linear and the relaxation mode, respectively (see Figs. 1a and b), supposed that $\mu \to 0$. $\to$ = Poincaré radius.

The map is depicted as a function over the radius. Any trajectory re-enters the radius (abscissa) at the corresponding function value (ordinate), such that identity circles are needed for the transfer. These identity circles are conveniently replaced by the identity map (first bisector), as indicated. Both a monotonously repelling and a monotonously attracting fixed point are found in this way, the former (at the origin) corresponding to the unstable focus, the latter (on the right hand side) to the stable limit cycle. For more details, see[13].

When the same map is constructed now for the system of Fig. 1 d (with the radius pointing in a direction either parallel to or away from the cliff), the more interesting picture of Fig. 5 results.

The map now possesses a "cap-shaped" region. All trajectories coming from the left are attracted by, and trapped in, the quadratic box which is bounded by the "first reinjected" and the "last non-reinjected" trajectory, respectively. The formation of this box is decisive because, whenever within such a box 2 upward moves (increases of amplitude) followed by a decrease below the initial point are possible, the conditions of the Li-Yorke theorem[11] are fulfilled, which means that the presence of chaos has been proved. Obviously, this condition is easy

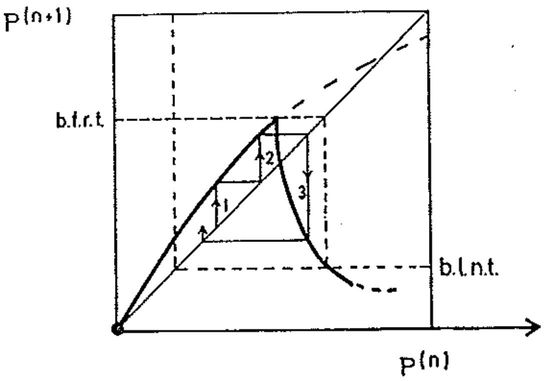

Fig. 5. Poincaré map of a universal circuit in the chaotic mode (see Fig. 1d), supposed that $\mu \to 0$. b.f.r.t. = borderline determined by first reinjected trajectory; b.l.n.t. = borderline determined by last non-reinjected trajectory; 1, 2, 3 = steps proving chaos (see text).

to meet by adjusting the parameters of the system. The simulation results of Fig. 3 provide a case in point.

Thus, the very technique which has been introduced recently for proving chaos in discrete systems [9–11] could be carried over to the continuous domain, simply by identifying the former next-state map with a Poincaré map.

As to the detailed mathematical implications of the Lorenz-Li-Yorke map (existence of an uncountable set of measure zero of repelling periodic attractors; all solutions in between are non-periodic; structural stability of flow), see [9–11].

Extension to the Non-idealized Case

The same trick which has been used above (substitution of a Poincaré map for a next-amplitude map) can still be applied when the idealizing assumption $\mu \to 0$ is dropped, such that the cross-section over which the Poincaré map is defined no longer is one-dimensional, but 2-dimensional. The resulting, still "folded", Poincaré map then is similar to a so-called Barker's transformation (as cited in [2]) or a so-called horseshoe map ([27], cited after [12]), respectively. A discrete system based on a "modified horseshoe map" has been studied only recently [12].

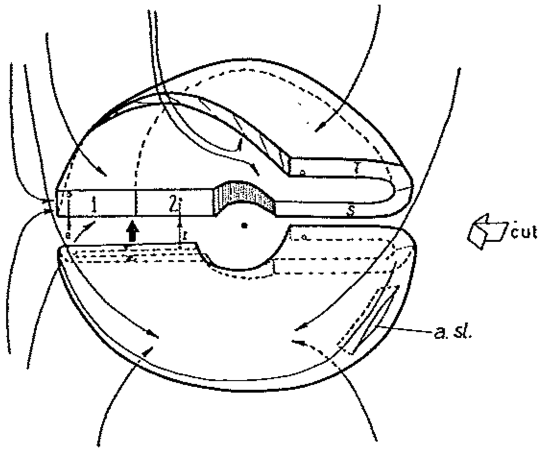

Fig. 6. The "three-dimensional blender". (Cf. Fig. 3a.) → = trajectories entering the structure from the outside; 1, 2 = half cross-sections (demonstrating the 'mixing transformation' that occurs), e = entry point of some arbitrarily chosen trajectory, r = reentry point of the same trajectory after one cycle, ↑ = 'horseshoe map', a.sl. = allowed slit (see text).

What actually happens in 3-space is shown in Figure 6. The "folded pancake" does not display the trajectories themselves, but only an "envelope" (made up of surfaces without contact, cf. [17]) which is entered by trajectories (as depicted), but never left. The picture is directly derived from Figs. 1 d (turned upside down) and 3, respectively, displaying the principal properties only. The rectangular cross-section on the left-hand side is seen to be mapped diffeomorphically onto a subset of itself, as required from a 2-dimensional Poincaré map. The "horseshoe" which is formed upon reinjection is also clearly visible.

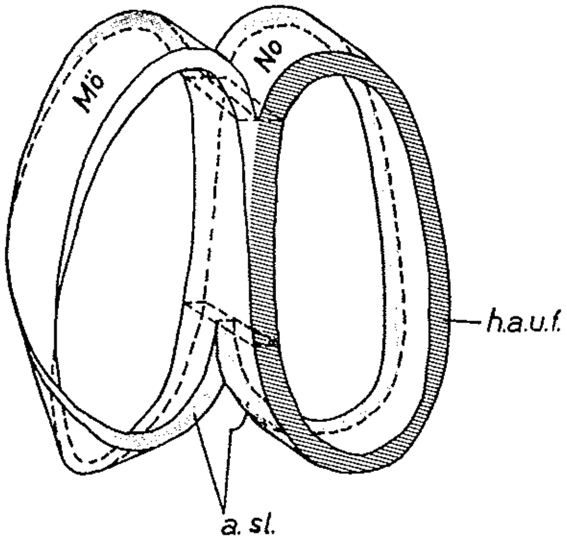

Fig. 7. A structure equivalent to that shown in Figure 6. Mö = Möbius loop, No = normal loop; h.a.u.f. = hole around the unstable focus in Fig. 6; a.sl. = boundaries of the allowed slit in Figure 6.

Due to the simplicity of the picture, it may be conjectured that it represents a sort of prototype for the generation of a "mixing" transformation [2] of horseshoe shape in 3 dimensions, realizing Smale's [3] suspension principle.

Figure 7 finally displays an, in a certain sense, equivalent structure. It is topologically equivalent to the cake, once a slit has been allowed in its right-hand back in such a way that no trajectories are damaged. Its essential part is the central rod which is carved in two mutually orthogonal directions on its top and its bottom, respectively.

As to the details already known about the 2-dimensional map (point patterns formed by the periodic repellors; existence of an "uncertainty principle" with respect to the predictable future time course of a trajectory in terms of the map's two coordinates; structural stability), see [12, 4].

Discussion

A continuous chemical system has been described which realizes a prototypically simple chaos-generating machine. Mathematically, the system is new only insofar as it provides a simpler example to some well-established facts (thereby perhaps acting as a conceptual catalyst). The observed "lateral reinjection" of a whole bundle of trajectories appears, as a principle of flowing in state space, possible only beyond the second dimension – just as "recurrence" of a single trajectory is a new principle in the transition from 1 to 2 dimensions (allowing for the phenomenon of oscillation). Thus, chaos can be classified as a dynamical property emergent with the third dimension. In this respect it is a sort of "superoscillation". Whether similar qualitative jumps are provided by the next-higher dimensions is an open question.

Chemically, the described system is just one out of a huge variety of possible combinations of an oscillator, on the one hand, and a switching system, on the other (cf. [26]). Therefore, further, simpler-to-realize examples should be easy to find. Some candidate systems have been listed in Figure 8. The fact that coil-type oscillations have already been observed in the well-known Belousov-Zhabotinsky reaction [28] (an oscillating system known to contain a hysteresis type subsystem [29]) renders the probability of finding a chaotic mode in this concrete chemical system great enough to warrant a systematic experimental investigation. The phenomenon of a "meandering" core, observed in a nonstirred excitable medium of the same type [30], speaks in the same direction (Winfree, personal communication). Incidentally, the behavior of diffusion-coupled chaotic systems poses a challenging dynamical problem in its own right.

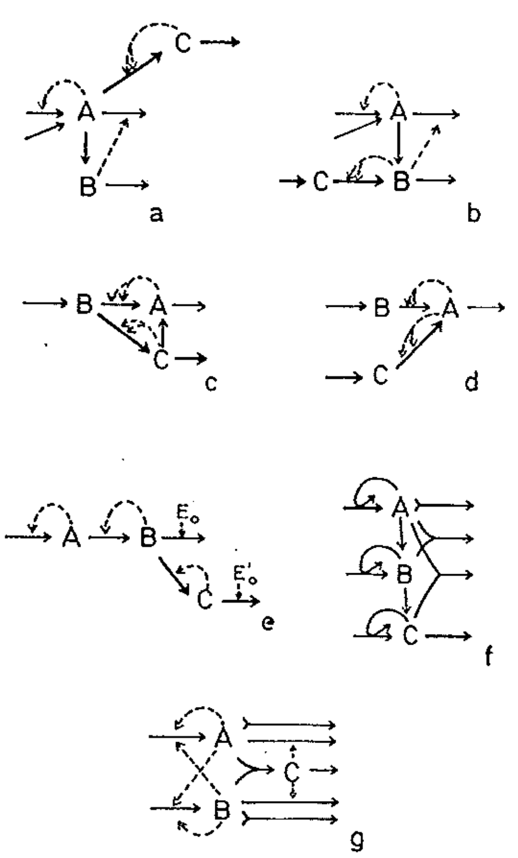

Fig. 8. Some further abstract reaction systems likely to produce chaos. — Thicker arrows = switching subsystems, thinner arrows = oscillatory subsystems.

Beyond facilitating artificial design, the described recipe (of combining an oscillation with a threshold in state space) is already realized in many natural systems, for example in certain metabolic and membrane-bound biochemical systems; in certain hormonal, neuronal and behavioral physiological systems; and in certain ecological, sociological and economic networks. Search for further distinct chaos-generating mechanisms (beyond the difference equation system of ecology already considered [8–11]) is therefore desirable also from an applicational point of view, in order for preventive and counter measures to be found for those cases in which the actual onset of chaotic behavior would be harmful.

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