Toroidal Oscillation in a 3-Variable Abstract Reaction System

Otto E. Rössler

Institut für Physikalische und Theoretische Chemie der Universität Tübingen
und Institut für Theoretische Physik der Universität Stuttgart, W. Germany

(Z. Naturforsch. **32 a**, 299–301 [1977]; received February 22, 1977)

An example of a chemical reaction system producing a type of oscillation that is close to a quasiperiodic oscillation is presented. Such system may help clarify the relationship between quasiperiodicity and chaos.

Introduction

Abstract reaction kinetics is dynamically universal [1, 2]. It not only allows for 3-variable chaos, as shown recently [3, 4], but also for 3-variable toroidal oscillations [5]. These oscillations, of which "quasi-periodic oscillations" form a structurally unstable special case [6], are characterized by trajectories that form spirals in close neighborhood to a torus in state space. The time-behavior of the variables hereby looks like a periodically amplitude-modulated oscillation plus a DC-component.

An Example

A first chemical example system is depicted in Figure 1. The dashed arrows correspond to catalytic

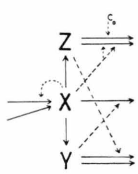

Fig. 1. Reaction scheme of a toroidal chemical oscillator. Constant pools (sources and sinks) have been omitted from the scheme as usual.

couplings. The corresponding rate equations are, under the usual assumptions of well-stirredness, isothermy, and an appropriate concentration range:

$$\begin{aligned} \dot{x} &= a\,x - y\,\frac{x}{x+K_1} + c \\ \dot{y} &= x - b\,y - z\,\frac{y}{y+K_2} \qquad (1) \\ \dot{z} &= d\,x - e\,(x^2+f)\,\frac{z}{z+K_3}\,, \end{aligned}$$

where $a = a' - 1 - d$ and $f = f'\,c_0/e$.

Requests for reprints to Doz. Dr. O. E. Rössler, Institut für Physikalische und Theoretische Chemie der Universität Tübingen, Auf der Morgenstelle 8, *D-7400 Tübingen*, Germany

The equation is written in the usual non-explicit convention, that is, fast-reacting intermediates have been eliminated via a Michaelis-Menten type steady state approximation (cf. [7]).

Simulation Results

In Fig. 2, a stereoscopic view of a segment of trajectorial flow in state space is presented. The trajectories form a coil [8] on the surface of a torus. The time-behavior of the 3 variables is shown in Figure 3. The system has been allowed some time to relax toward the asymptotic regime, after starting from nearby, but non-selected, initial conditions.

The flow of Fig. 2 looks very much like a quasi-periodic flow, that is, the trajectories do not close over the time span of the simulation. Nonetheless, none of the criteria for equations with truly quasi-periodic behavior (existence of a first integral of

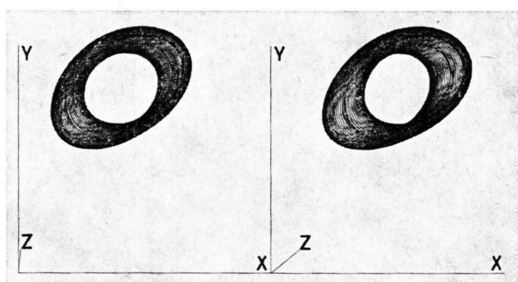

Fig. 2. Toroidal oscillation in the reaction scheme of Fig. 1 and Equation (1). Stereoscopic plot. (Parallel projections; the left-hand picture is for the right eye and vice versa: Try to fix a pencil in front of your eyes in such a way that, of the 4 pictures behind it, the 2 innermost pictures merge although being blurred. Then just wait until they become sharp.) Numerical simulation on a HP 9820A desk calculator with peripherals, using a standard Runge-Kutta-Merson integration routine (adapted by F. Göbber). Parameters assumed: $a=0.3026$, $b=0.3$, $c=3$, $d=0.4$, $e=0.1$, $f=3.4$, $K_1=K_2=K_3=K=0.001$. Initial conditions: $x(0)=z(0)=1$, $y(0)=4.1$. The plot begins at $t_1=1872$ and ends at $t_2=2345$. Axes: $0\ldots5$ for x and y, $0\ldots1$ for z.

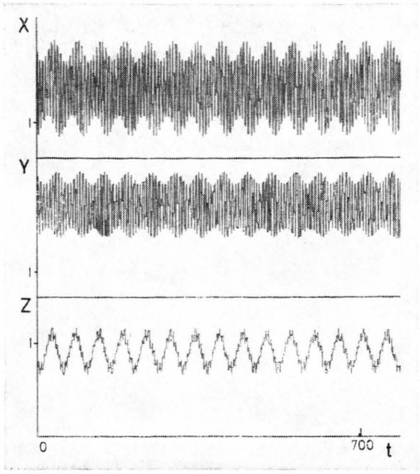

Fig. 3. Time behavior of the 3 variables of Figure 2. Numerical simulation as in Figure 2. $t_0 = t_2$ of Figure 2.

motion, vanishing divergence, or certain symmetry properties, respectively [6]) applies to Eq. (1) if $K \neq 0$. However, for $K \rightarrow 0$ and $a \rightarrow b$, Eq. (1) does possess truly quasi-periodic behavior [9] in the limit. In that case, a whole 1-parameter family of invariant tori exists [9].

It is to be expected that even simpler reaction schemes than the one of Fig. 1 will be able to produce the same type of behavior. The reason is, simply, that all the (formidable) nonlinearities of Michaelis-Menten type involved in Eq. (1) are not essential for the main result.

Discussion

Toroidal oscillations are of interest presently in nonlinear physics [10–12] and economics [13]. In both fields (especially in hydrodynamics and economy) very large recurrence times are the rule. In turbulence theory the problem whether toroidal oscillation is a necessary – or at least frequent – step toward "chaos" is still an open question (see [12]).

Chaos is possible in 3-variable reaction systems [3, 4], as well as in 3-variable models in hydrodynamics [14] and lasers [15, 16]. (Near-)quasiperiodic behavior, however, of 3-variable Euclidean systems has not yet been described in reaction kinetics. So far it was anticipated that chaos is easier to obtain than quasiperiodicity. The above example of a basically simple 3-variable "quasiperiodic" system of non-distributed type shows that such systems are not necessarily more complicated than the chaos-producing 3-variable systems. The essential component is a single nonlinear term of second order. Equations of the very type of Eq. (1) can also yield chaos [9]. In three dimensions, therefore, one way to obtain chaos is via an invariant torus. The underlying Poincaré map then is a "contracting ring map". Thus, the two basic types of chaos (horseshoe map chaos and sandwich map chaos; see [17]) can be complemented by a third type: ring map chaos [4, 9]. However, it seems that toroidal oscillations are not a necessary (although, perhaps, a sufficient) condition for chaos.

In concrete chemical and biochemical systems (for example, in the glycolysis system [18]) the occurrence probability for chaos is still greater than that for toroidal oscillation. While almost any double-loop reaction system (and even single-loop system) is likely to be capable of chaotic oscillations, especially of it contains a so-called double-periodicity, see [19], toroidal oscillation seems, at the time being, to require more constituents (namely, a double-loop structure and a combination of catalytic influences of both first and second order on the same variable), if it is to occur in a network of strongly coupled chemical reactions. Nonetheless it is possible that toroidal oscillations will be observed in one cuvette some day. In two cuvettes (that is, with 2 independent oscillators), quasiperiodicity is, of course, unavoidable.

To conclude, another type of "exotic" oscillation has been found in 3-variable reaction kinetics, toroidal oscillation.

I thank Professor H. Haken for discussions.

1 O. E. Rössler, A Synthetic Approach to Exotic Kinetics, with Examples. Lecture Notes in Biomathematics **4,** 546 [1974].

2 O. E. Rössler, Chemical Automata in Homogenous and Reaction-Diffusion Kinetics. Lecture Notes in Biomathematics **4,** 419 [1974].

3 O. E. Rössler, Chaotic Behavior in Simple Reaction Systems. Z. Naturforsch. **31 a,** 259 [1976].

4 O. E. Rössler, Chaos in Abstract Kinetics: Two Prototypes. Bull. Math. Biol. **35** [1977].

5 O. E. Rössler, Quasiperiodic Oscillation in Abstract Reaction System (Abstract). Biophys. J. **17,** 281a [1977].

6 J. Moser, Perturbation Theory of Quasiperiodic Solutions of Differential Equations. In: Bifurcation Theory and Nonlinear Eigenvalue Problems (J. B. Keller and S. Antman, eds.), p. 283. Benjamin, New York 1969.

[7] F. G. Heineken, H. M. Tsuchiya, and R. Aris, On the Mathematical Status of the Pseudo-steady State Hypothesis in Biochemical Kinetics. Math. Biosci. **1,** 95 [1967].
[8] O. Gurel, Topological Dynamics in Neurobiology. Int. J. Neuroscience **6,** 165 [1973].
[9] O. E. Rössler, Quasiperiodic Oscillations and Chaos in 2 Simple Differential Equations (in preparation).
[10] D. Ruelle and F. Takens, On the Nature of Turbulence. Comm. Math. Phys. **20,** 167 [1971].
[11] O. E. Lanford, Bifurcation of Periodic Solutions into Invariant Tori: the Work of Ruelle and Takens. Lecture Notes in Mathematics **322,** 159 [1973].
[12] M. McCracken and J. E. Marsden, The Hopf Bifurcation. Springer-Verlag, New York 1976.
[13] H. R. Grümm, Bifurcations and the Appearance of Attracting Tori. In: Analysis and Computation of Equilibria and Regions of Stability (H. R. Grümm, ed.), p. 83. International Institute for Applied Systems Analysis Conference Proceedings, Schloß Laxenburg, A-2361 Austria 1975.
[14] E. N. Lorenz, Deterministic Nonperiodic Flow. J. Atoms. Sci. **20,** 130 [1963].
[15] H. Haken, Analogy Between Higher Instabilities in Fluids and Lasers. Phys. Lett. **53 A,** 77 [1975].
[16] R. Graham, Onset of Self-pulsing in Lasers and the Lorenz Model. Phys. Lett. **58 A,** 440 [1976].
[17] O. E. Rössler, Different Types of Chaos in Two Simple Differential Equations. Z. Naturforsch. **31 a,** 1664 [1976].
[18] J. Higgins, R. Frenkel, E. Hulme, A. Lucas, and G. Ranzagas, The Control Theoretic Approach to the Analysis of Glycolytic Oscillators. In: Biological and Biochemical Oscillators (B. Chance, E. K. Pye, A. K. Ghosh, and B. Hess, eds.), p. 127. Academic Press, New York 1973.
[19] J. Boissonade, Aspects théoriques de la "double oscillation" dans les systèmes dissipatifs chimiques. J. Chimie Physique **73,** 540 [1976].