A course in . . .

NONLINEAR SYSTEMS

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DURING THE PAST three years we have provided our graduate students with a third, optional, course on the mathematics of nonlinear systems. The course follows two required courses that formalize the structures of linear, or vector, spaces and nonlinear metric spaces leading to the solution of partial differential equations. These two courses have been described by Lauffenburger, *et al* [1].

The nonlinear math course provides an opportunity for the students to examine the complex solution spaces that chemical engineers encounter in modeling many chemical processes, especially those involving reaction and diffusion, autocatalytic reactions, phase equilibrium in the critical region, and multistaged operations. Some of the simplest exothermic reactions in CSTRs with heat transfer exhibit branches in their solution diagrams that contain limit and bifurcation points, both steady-state and periodic, and trace out isolas as parameters are varied. For such systems, solution diagrams are calculated to show the importance of characterizing the singular points and expressing their normal forms and universal unfoldings so as to determine the number of steady state solutions in their vicinity. Examples are selected to demonstrate steady-state foci that bifurcate to time-periodic limit cycles which, in turn, undergo secondary bifurcations that lead to chaotic behavior, and even intermittent interchanges between periodic and chaotic modes of operation. Experimental observations of these phenomena are reviewed to drive home the importance of developing models that have corresponding solutions. In many cases, the models have complex solution diagrams that *don't* correspond to the experimental measurements and which emphasize the importance of locating the solution that most closely matches the data.

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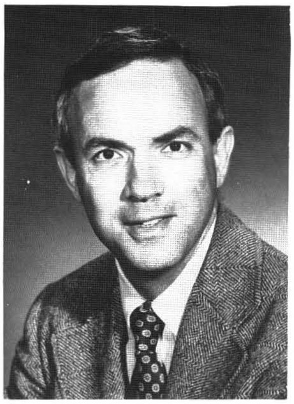

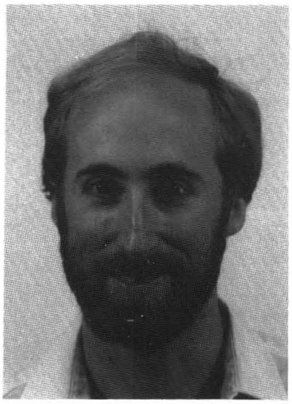

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Lyle Ungar joined the faculty at Penn in 1984 as assistant professor, having received his BS degree at Stanford and his PhD at MIT. His research interests include application of perturbation methods, bifurcation theory, and finite element analysis to transport problems in continuum physics, crystal growth and rapid solidification materials processing. He is also applying artifical intelligence programming techniques to process control problems. **(R)**

Nonlinear phenomena, such as the formation of spatial and temporal patterns and chaotic behavior, arise naturally in many systems with fluid flow or chemical reaction. Combustion, natural and forced convection, biological systems with competing species, and catalytic reactions can all require nonlinear analysis. Nonlinearities can also be generated in the design of complex processes, such as those integrated to achieve a high thermodynamic efficiency, and can introduce oscillatory and chaotic regimes that can present pitfalls and obstacles to easy operation and control. The design of these processes usually begins with the analysis of simple structures using approximate models. Gradually, as the synthesis tree is pruned, more complex models can be justified to represent the real phenomena more accurately. However, the more complex models usually have a richer solution space and the number, type (steady, periodic, or chaotic) and stability of the solutions varies with the specifications and the parameters of the model. In design calculations, as in all mathematical modeling, the student must learn to beware of algorithms that converge to solutions that are not physically correct. The seriousness of this problem in the design stage, when experimental data are not available, is emphasized. Of course, when the solutions are observed experimentally, it is important to recognize the possible existence of multiple solutions and to design control systems to achieve the desired performance. This permits focusing on designs that have few regimes of operation; that is, less complex solution diagrams.

EVOLUTION OF THE COURSE

The initial version of the course concentrated on the general aspects of bifurcation and singularity theories with examples of many applications that arise in chemical processing. Emphasis was placed on the use of analytical perturbation methods to analyze nonlinear systems. A variety of techniques were covered for describing steady and oscillatory bifurcations and how they change as parameters are altered [18]. No computations were carried out, so complete solution diagrams were obtained only for very simple problems. Continuum problems such as natural convection were studied at the end of the course, but only the onset of instability could be covered in the homework exercises. This approach closely parallels that of Iooss and Joseph [3] in their text, *Elementary Stability and Bifurcation Theory*, which was the principal reference in that initial offering. That text has several limitations, the most severe being the lack of physical examples and the lack of coverage of chaotic phenomena. A mixture of papers from the math, physics and engineering literatures was therefore used as supplements.

In the most recent version of the course (Spring, 1987), emphasis was shifted more toward the process models and the methods of computing the singular points and the branches that connect them in solution diagrams. This was facilitated by improvements in available texts and software. Kubicek and Marek's [2] text, *Computational Methods in Bifurcation Theory and Dissipative Structures*, provides a unified approach to the analysis of solution diagrams. Several process models are introduced in Chapter 1. Then, as

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the singular and bifurcation points are defined, examples are illustrated in the solution diagrams for these process models. General methods are presented to compute the singular and bifurcation points and the branches that connect them. These are very helpful, but unfortunately many of the definitions are stated briefly and the figures and tables in which the results are presented are explained insufficiently. Many questions arise which can only be answered by computational experiments. To accomplish this, we initially introduced our own program for the continuation of steady-state solutions [8] and placed some emphasis on the logic that enables it to traverse turning-points effectively. Midway through the semester we obtained a copy of the AUTO program [29], and this added immeasurably to the course. AUTO enabled us to perform computational experiments with ease and to answer many questions, especially those concerning branches of periodic solutions. These will be considered in the next section, in which the syllabus for the course is presented and the role of AUTO is described.

COURSE CONTENTS AND PHILOSOPHY

A central aspect of the course as it has evolved is the integration of analytical and numerical techniques and their application to physical problems. Analytical techniques and theorems provide a general framework for understanding how and when stability can change and new solution branches can arise. Numerical calculations provide complete solution diagrams for specific physical problems. Interpreting these solution diagrams in the contexts of both singularity theory and the physical problem from which they arise gives students a better understanding of nonlinear phenomena.

The core of the course concentrates on the methods of analyzing what happens at and near different steady and time-periodic bifurcations (see Table 1 for a list of topics covered). To calculate bifurcations of nontrivial solutions, one must generally turn to the computer. Hence, major effort was devoted to numerical techniques. All numerical techniques essentially grow out of analytical perturbation techniques. When using Newton's method, the linearized equations are present and so one can, for example, monitor the determinant to find steady bifurcations and check for changes in stability without calculating the eigenvalues. The implicit function theorem guarantees that stability can only change when the determinant vanishes; arc-length continuation around limit points [5] naturally grows out of the classification of limit points and steady bifurcation points, and Poincare maps provide the basis for calculating time-periodic solutions. The latter can be difficult to compute, especially when the branches of limit cycles are unstable. Hence, the Newton-Fox procedure for locating a point on the limit cycles is described, following the approach of Aluko and Chang [27]. Stability analysis of the Monodromy matrix, according to the Floquet Theory, and the methods of continuation to locate secondary bifurcations lead naturally to studies of the transitions to chaos.

We have tried to integrate smoothly the three different aspects of the course: analytical methods, numerical methods, and physical insight. The general theory is illustrated throughout with examples, and relevant computational techniques such as homotopy methods are covered as they are used.

For example, we have used the Belousov-Zhabotinskii reaction system as an example of steady and time-dependent bifurcations and of chaotic behavior. This relatively simple set of reactions has been observed to produce a bewildering variety of spatial and temporal patterns, and has been widely studied as a simple prototype for many reaction-diffusion systems. These have been summarized nicely in a review article by Epstein [41] that illustrates the regimes of periodic behavior with beautiful color photographs of the oscillations in a stirred beaker and in a Petri dish with diffusion effects. In our coverage, the initial kinetic model of Field and Noyes [33] was presented and mass balances were derived for the three principal intermediates, $HBrO_2$, Br^- and Ce^{4+}:

$$\frac{dy_1}{dt} = 77.27\ (y_2 - y_1y_2 + y_1 - ky_1^2)$$

$$\frac{dy_2}{dt} = (-\ y_2 - y_1y_2 + y_3)/77.27$$

$$\frac{dy_3}{dt} = 0.161\ (y_1 - y_3)$$

whose dimensionless concentrations are y_1, y_2, and y_3, respectively. For this mechanism, which assumes the autocatalytic formation of $HBrO_2$, the rate constant k is a key parameter. Steady-state continuation calculations show that as k is decreased, the L_2-norm of **y** increases, as illustrated in Figure 1. At k = 0.02394, a Hopf bifurcation point is encountered. The steady-state branch becomes unstable and a new branch is born. Early in the course, the students computed Figure 1 using our continuation program and located the Hopf bifurcation point by computing the eigenvalues of the Jacobian along the steady-state

TABLE 1
Course Topics

BACKGROUND
- **Implicit function theorem**
- **Stability theory**

STEADY BIFURCATIONS
- **Single and multiple limit points**
- **Continuation methods**
- **Liapunov-Schmidt reduction**
- **Effect of a second parameter**
  - **Perturbed and "broken" bifurcations**
- **Singularity theory**
  - **Normal forms, unfoldings**
  - **Representation of energy for conservative sytems**

TIME DEPENDENT BIFURCATIONS
- **Floquet theory**
- **Hopf bifurcations**
- **Integration of stiff ODEs**
- **Secondary bifurcations**

CHAOTIC BEHAVIOR
- **Transitions to chaos**
  - **Period doubling**
  - **Incomensurate frequencies**
  - **Intermittency**
- **Characterization of chaos**
  - **Power spectra**
  - **Poincare sections and maps**
  - **Liapunov exponents**

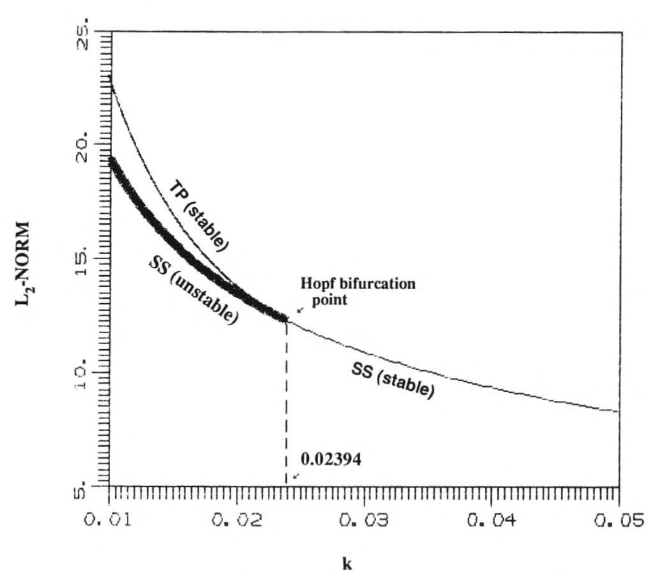

FIGURE 1. Steady and time-periodic branches for the Belousov reaction system (TP—time periodic, SS—steady-state).

branch. Then, with the LSODE program, they plotted the limit cycles in the time-domain (see Figure 2), and showed the decrease in the frequency of oscillation as k decreases. Finally, the AUTO program performed these calculations with much less preparation and traced the periodic branch in more detail, showing the variation of the frequency with k. This analysis led naturally to several papers that show how chaotic behavior arises in CSTRs and introduces alterations in the model to track these strange attractors [38, 39, 40]. As expected, computational experiments by the students with the AUTO program were unable to track the strange attractors, but success was achieved with LSODE. These results are displayed in the phase-plane of Figure 3, which closely resembles the results illustrated by Epstein and others.

The AUTO program was a great aid in enabling students to calculate solution structures and to get a feel for how nonlinear systems behave. AUTO is a collection of FORTRAN routines whose primary purpose is to compute the branches of stable or unstable periodic solutions of systems of ODEs that are functions of a free (bifurcation) parameter. AUTO also determines the branches of steady-state solutions, locates limit and real bifurcation points along solution branches, and can switch branches at these points. It can also locate limit points and curves of Hopf bifurcation points using two-parameter, continuation methods. A tape containing the AUTO routines was installed on our VAX computer, under the VMS operating system, in less than two hours. Of special note is that AUTO is currently limited to small ODE systems (up to twelve state variables). Computations of steady branches normally proceed very rapidly, whereas time-periodic branches can be slow to compute, especially when they are unstable.

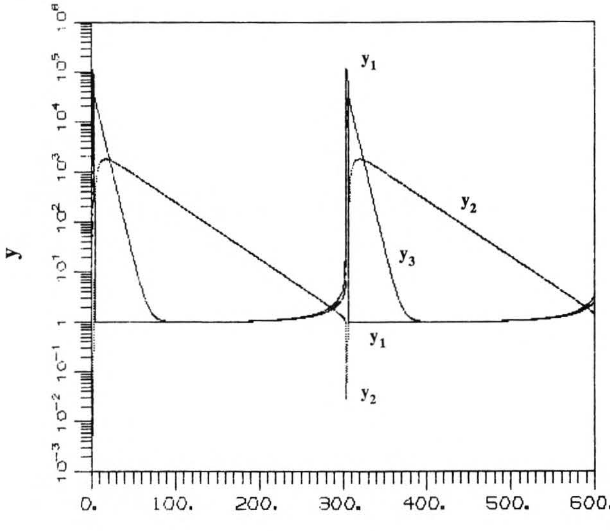

FIGURE 2. Dynamic simulation of Belousov reaction system with $k=8.375 \times 10^{-6}$.

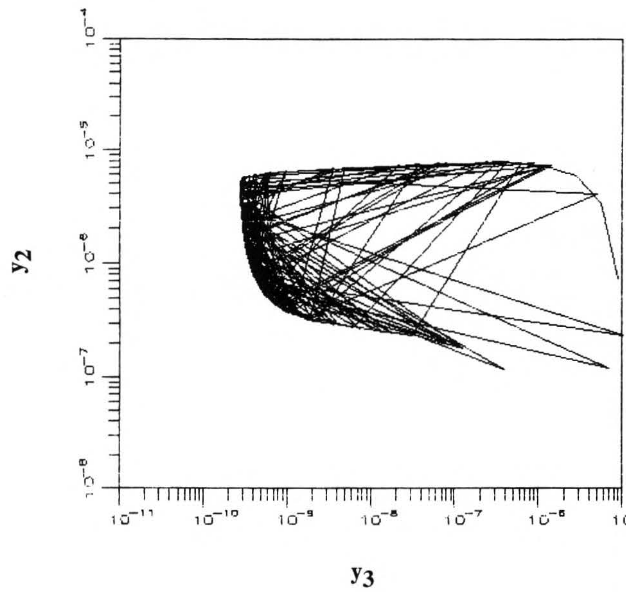

FIGURE 3. Phase-plane for Belousov reaction system. Turner model with $\tau=0.2962$.

At the end of the course, the students studied articles on either analytical or numerical techniques or on specific systems that exhibit interesting nonlinear behavior and made presentations to the class. A list of project areas and references is given in Table 2. In the initial version of the course emphasis was placed on classic papers describing the effect of container shape on the onset of natural convection, pattern formation due to competing biological species (predator-prey systems with similarities to the Belousov-Zhabotinskii system) and different transitions to turbulence in forced convection. Several mechanical engineering students enrolled in the course studied bifurcation-based descriptions of buckling. In 1987, the papers focused on chemical processes that exhibit complex solution diagrams, usually with transitions to chaos. Several of these papers present the latest results of studies of systems that naturally exhibit chaotic behavior or become chaotic under the influence of forced oscillations.

CONCLUSIONS

Nonlinear phenomena are ubiquitous. They have received little attention largely because the required mathematics is less well-developed and harder to comprehend than for linear systems. Bifurcation and singularity theory provide a framework for classifying and understanding nonlinear phenomena. They follow very easily from a linear operator approach and provide a dramatic demonstration of the constructive use of Fredholm's alternative. This has enabled us to successfully touch on bifurcation theory in the last days of the prior required portion of our graduate math sequence. When bifurcation techniques are implemented in a computer package such as AUTO, they also provide a means of mapping out solution structures. Students can obtain an intuitive understanding of nonlinear phenomena by examining the solutions to physical problems. With the right software, they can also generate these solutions themselves and prepare an array of two- and three-dimensional drawings that permit more thorough analysis and visualization than is possible with the few drawings that typically accompany technical articles and books.

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TABLE 2
Sample of Articles Reviewed

AUTHOR	TITLE (Reference No.)
EARLIER ARTICLES	
Hall, Walton	Benard Convection in a Finite Box: Secondary and Imperfect bifurcations (42)
Keener	Oscillatory Coexistence in the Chemostat: A Codimension Two Unfolding (43)
Segel	Mathematical Models in Molecular and Cellular Biology (44)
Eckmann	Roads to Turbulence in Dissipative Dynamical Systems (55)
Thompson, Hunt	A General Theory of Elastic Stability (45)
Triantafyllidis, Tvergaard	On the Development of Shear Bands in Pure Bending (46)
FOCUS ON CHAOTIC BEHAVIOR—1987	
Kahlert, Rossler, Varma	Chaos in a Continuous Stirred Tank Reactor with Two Consecutive First-order Reactions: One Exo-, One Endothermic (50)
Mankin, Hudson	Oscillatory and Chaotic Behavior of a Forced Exothermic Chemical Reaction (51)
Mankin, Hudson	The Dynamics of Coupled Nonisothermal Continuous Stirred Tank Reactors (52)
Kim, Hlavacek	On the Detailed Dynamics of Coupled Continuous Stirred Tank Reactors (32)
Nandapurkar, Hlavacek, Van Rompay	Chaotic Behavior of a Diffusion-Reaction System (53)
Chang, Chen	Bifurcation Characteristics of Nonlinear Systems Under Conventional PID Control (54)

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