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COLLEGE OF ENGINEERING

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Subject: Analytical and Computational Analysis of the Dynamics of Combustion

cc: Dr. Yi Zhao, Professor of Aerospace Engineering
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INTRODUCTION

The project I worked on is part of a major research project that is currently being conducted by Professors Joseph Powers and Samuel Paolucci, together with several Ph.D. students of the Aerospace and Mechanical Engineering Department of the University of Notre Dame. The project is mainly sponsored by Los Alamos National Laboratories, Los Alamos, New Mexico.

The main goal of the project is to obtain a simplified representation of complicated reaction processes. The part of the project I was responsible for was the computation and graphical representation of **invariant low dimensional manifolds** for several systems of Ordinary Differential Equations (ODEs) associated with some simple chemical reactions.

My cooperative educational research experience started on June 23, 2003, and ended on August 22, 2003. I had a desk in the Aero/Mechanical Engineering Graduate Students' Office in the Fitzpatrick Hall of Engineering. I was suggested to work eight hours per day, and besides a stipend, I had all the benefits of being a student of the University of Notre Dame.

UNDERSTANDING THE BASICS OF COMBUSTION

During the first two weeks of my research I read several chapters chosen from many books, dealing with the thermodynamics of combustion, ordinary differential equations, and boundary value problems. Next, I went through published and unpublished papers regarding different ways of representing low-dimensional manifolds of ODEs. I considered theories of manifolds by Powers and Paolucci, Davis and Skodje, Mass and Pope, and Roussel and Fraser, all of them authors of several papers in the Journal of Chemical Physics among others.

The aim of this part of the project was understanding the basics of combustion. Several combustion processes in the real world involve a considerable number of reaction steps (e.g. Table 1). During these steps, several intermediate molecular species are formed. At any particular instant, the species that are observed are those whose atomic configurations have a lower energy state. This procedure will go on, until the reaction reaches equilibrium.

Reaction	Rate
$H+O_2 \rightarrow OH+O$	5.55×10^6
$OH+O \rightarrow H+O_2$	8.83×10^7
$O+H_2 \rightarrow OH+H$	1.54×10^7
$OH+H \rightarrow O+H_2$	1.29×10^7
$H+H_2O \rightarrow OH+H_2$	1.10×10^6
$OH+H_2 \rightarrow H+H_2O$	3.76×10^7
$O+H_2O \rightarrow OH+OH$	1.07×10^6
$OH+OH \rightarrow O+H_2O$	3.11×10^7

Table 1. Example of a multi-step reaction: first reduction of H2-O2 mechanism. Units for Rates are cm^3 molecule^-1 s^-1

Naturally all these steps are so fast, that usually engineers do not even consider that intermediate steps actually occur. However, a better understanding of combustion comes from the analysis of the dynamics of all these steps. The crucial point of this analysis is the formulation of the rate of change with time of all molecular species. From here it is easy to understand that we would have as many equations, as the number of species. All these equations will form a system of ODEs, and its analysis would reveal the "secrets" of the combustion process itself.

For instance, considering a combustion process that involves fuel (F), *a* moles of oxidizer (O) producing *b* moles of combustion product (P), we can write the reaction as follows:

$$F + aO \rightarrow bP \tag{1}$$

The rate at which the fuel is consumed can be expressed by the ODE (law of mass action):

$$\frac{d[X_F]}{dt} = -k_G(T)[X_F]^n [X_{Ox}]^m \tag{2}$$

Where $[X_i]$ refers to the molar concentration of the *i*th species of the reaction, $k_G(T)$ is the reaction rate coefficient and is a function of temperature (given by Arrhenius kinetics), and the exponents *n* and *m* are the reaction orders for a particular reaction component. The law of mass action applied to all the intermediate species will yield to a non-linear system of ODEs. A complete solution of this system, after appropriate simplifications, gives a description of the chemical kinetics of the system evolution over time.

ANALYSIS OF COMBUSTION WITH NUMERICAL AND COMPUTATIONAL METHODS

Most of the time I succeeded in obtaining the ODEs needed, but I had several difficulties in solving the system and drawing the solutions in the phase-plane for several cases I considered. In fact the system is usually **non-linear**; therefore I had to go through the process of local linearization using the Jacobian matrix. Once the system is linearized, one finds the associated eigenvalues and eigenvectors at different critical points.

Furthermore, since the problems we are considering are taken from the physical world, critical points with negative molar concentration can be neglected.

At this point we should pay attention to the nature of the eigenvalues and eigenvectors we obtained. In fact, the chemical reaction reaches equilibrium only at the critical points where both the associated eigenvalues are negative. Also, in order to understand how the reaction goes to equilibrium, we want to plot a trajectory that, starting from a particular initial condition will eventually reach the equilibrium point. Next, we have to consider the eigenvectors associated with local linearizations, since they give us the directions of the trajectories towards the steady-state value. For different initial conditions we have different trajectories. However, all these trajectories, even though starting from different initial conditions, seem to be attracted by another invariant trajectory, and all of them will eventually merge on it: the **invariant manifold**. The exact manifold for simple systems can be found with simple analytical methods. However, invariant manifold are not always attracting, but if they are locally attracting, we say they are locally stable. Among other trajectories, they only have the property that a point which begins on an invariant manifold will remain on an invariant manifold. For a better understanding of how trajectories and invariant manifolds look in the phase plane, please refer to Figure 1.

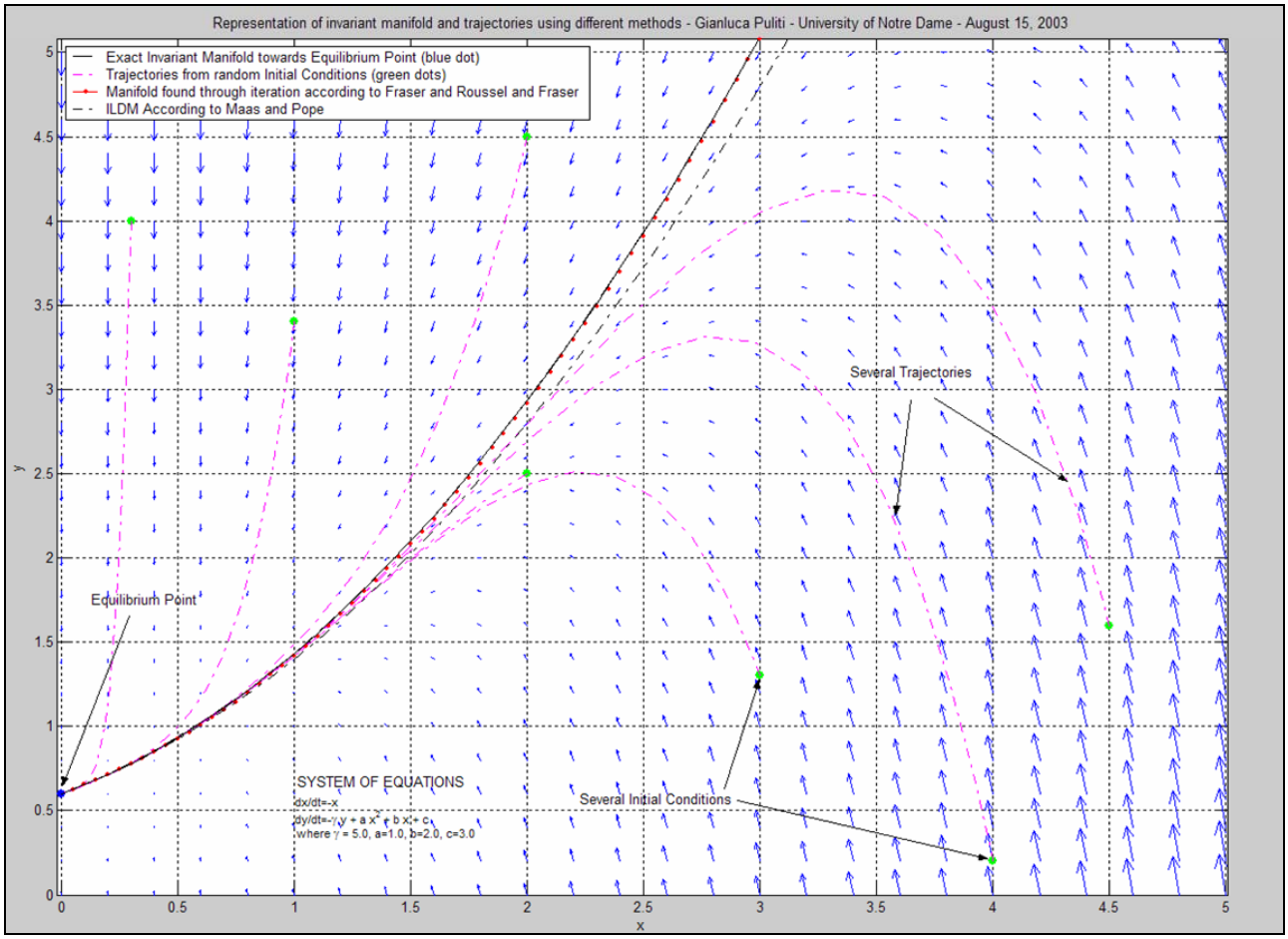


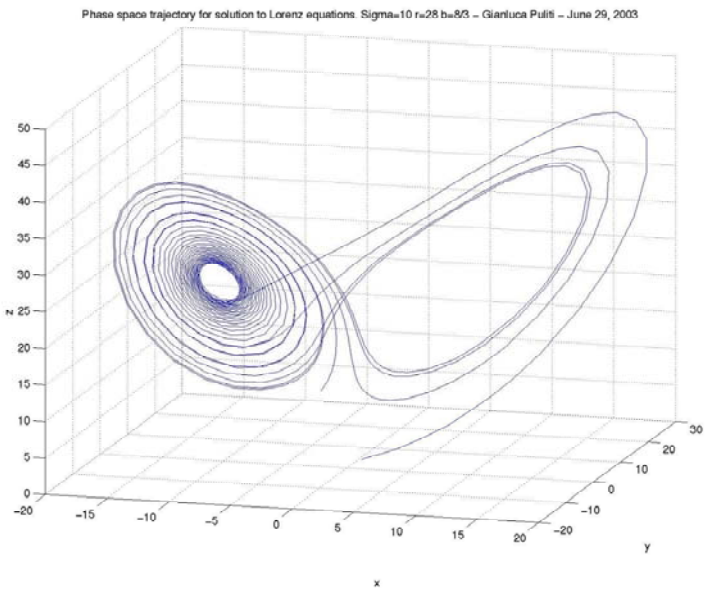
Figure 1. Manifolds for a simple system of ODEs using different methods

Another difficulty I had during the analysis was the huge computational time needed by the powerful SunOS Microsystems in solving some systems of ODEs. This is caused by the so-called "**stiffness**" of the system, associated with the range of time-scales. The reaction time-scales of the system are proportional to the reciprocal of the real components of the eigenvalues determined from the Jacobian matrix. If the fastest time-scale is several orders of magnitude bigger than the slowest one, this means that the system is stiff. In this case the computer would take weeks to compute the critical points. Also, as the number of species increases, the resulting manifold increases in dimension and become impossible to represent via the phase plot; therefore, the most important manifold and most easily understood is a **low-dimensional manifold**.

Thus far the manifolds we are more familiar with are the following:

- *Zero-dimensional manifold*: a point in the phase space (when $t \to \infty$)
- *One-Dimensional manifold*: a curve in the phase plane (slowest time-scale dominates the system)
- *Two-Dimensional manifold*: a surface (two longest time-scales dominate the responses)
- *"Two-Plus"- Dimensional manifold*: e.g. Lorenz strange attractor (Figure 2)
- *Three-Dimensional manifold*: in this case we can only plot sections of it.

Figure 2 (below): Lorenz strange attractor



However, we are not always able to find exact invariant low-dimensional manifold with simple analytical methods since the systems could be too complex. In these cases we can find an approximation: I studied in particular the ILDM (Intrinsic Low Dimensional Manifold) method by Maas and Pope, the iteration method by Roussel and Fraser, and the ASIM (Approximate Slow Invariant Manifold) method by Singh, Powers, and Paolucci. Some of these methods are used in Figure 1.

I repeated this analysis for several systems of ODEs, and in particular for those associated with the Zeldovich Mechanism (formation of nitric acid from atmospheric nitrogen, 4 reactions), and with the Decomposition of Ozone (14 reactions).

Hopefully, in the near future, the project will succeed in representing low dimensional invariant manifolds for combustion processes, involving hundreds of reactions.

CONCLUSION

My cooperative educational experience at the University of Notre Dame gave me the real feeling of scientific research, and allowed me to experience what graduate school has to offer. It surely improved my skills in computational methods, since I have been using Matlab and Maple on a daily basis. This experience taught me also how advanced mathematics and numerical methods can be used to understand fluid-thermodynamics problems. I proved to myself that mathematics and engineering are actually closer to each other than what we often think in undergraduate studies.

The research project is still going on at the University of Notre Dame, and I hope to keep collaborating with them in the future. I wish the best of luck to the University of Notre Dame for a great success on the project. Hopefully it will open new frontiers to new developments in the Aerospace and Mechanical Engineering fields.

Furthermore, I would like to thank Embry-Riddle for its excellence in education that gave me the possibility to work on this kind of research without any difficulty. This does not mean I never felt frustrated during my research experience, since frustration is part of scientific research. However, I always knew what to do next, and what the goal of the day was. This means that Embry-Riddle, besides giving me an excellent engineering education, gave me a great sense of responsibility and willingness.

Sincerely,

Gianluca Puliti