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Dynamical system analysis of ignition phenomena using the Tangential Stretching Rate concept

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ABSTRACT

We analyze ignition phenomena by resorting to the stretching rate concept formerly introduced in the study of dynamical systems. We construct a Tangential Stretching Rate (TSR) parameter by combining the concepts of stretching rate with the decomposition of the local tangent space in eigen-modes. The main feature of the TSR is its ability to identify unambiguously the most energetic scale at a given space location and time instant. The TSR depends only on the local composition of the mixture, its temperature and pressure. As such, it can be readily computed during the post processing of computed reactive flow fields, both for spatially homogeneous and in-homogenous systems.

Because of the additive nature of the TSR, we defined a normalized participation index measuring the relative contribution of each mode to the TSR. This participation index to the TSR can be combined with the mode amplitude participation Index of a reaction to a mode – as defined in the Computational Singular Perturbation (CSP) method – to obtain a direct link between a reaction and TSR. The reactions having both a large participation index to the TSR and a large CSP mode amplitude participation index are those contributing the most to both the explosive and relaxation regimes of a reactive system. This information can be used for both diagnostics and for the simplification of kinetic mechanisms.

We verified the properties of the TSR with reference to three nonlinear planar models (one for isothermal branched-chain reactions, one for a non-isothermal, one-step system, and for non-isothermal branched-chain reactions), to one planar linear model (to discuss issues associated with non-normality), and to test problems involving hydro-carbon oxidation kinetics.

We demonstrated that the reciprocal of the TSR parameter is the proper characteristic chemical time scale in problems involving multi-step chemical kinetic mechanisms, because (i) it is the most relevant time scale during both the explosive and relaxation regimes and (ii) it is intrinsic to the kinetics, that is, it can be identified without the need of any ad hoc assumption.

1. Introduction

Ignition of hydrocarbon (and hydrogen) fuels is controlled by branched-chain reactions and its complexity increases with the length/size of the fuel molecule.

The Computational Singular Perturbation (CSP) Method has been used to analyze two-stage ignition of n-heptane by Kazakov et al. [1] and Goussis et al. [2,3]. They observed the occurrence in spatially homogeneous systems of two branches of positive eigenvalues during both chain-branching and thermal ignition. Chen et al. [4] resorts to CSP to study high temperature ignition and combustion enhancement by dimethyl ether addition to methane–air mixtures. Lu et al. [7–10] proposed an explosion index for Chemical Explosive Modes (CEM) to analyze DNS datasets of turbulent flames. Lu et al. [11] also discussed the role of eigenvalues with a positive real part in the context of limit phenomena in combustion. Najm et al., [12], found explosive modes in edge flame data sets. Gupta et al. adopted CSP to classify ignition regimes in HCCI combustion [5], and n-heptane auto-ignition characteristics in DNS datasets [6]. All these ignition studies stress the role of eigenvalues with a positive real part in ignition and other limit phenomena for all fuels tested (hydrogen and hydrocarbons).

The present work analyzes the kinetic behavior related to ignition events by adopting the concept of local stretching rates introduced in [13]. The local stretching rates find their theoretical justification in the theory of normal hyperbolicity applied along system orbits/trajectories.

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Upon close analysis, we found that the basic definition of the local stretching rate given in [13] fails to provide meaningful information when the trajectory proceeds through a region of complex eigenvectors. To overcome this shortcoming, we propose in this paper a modification of the local Tangential Stretching Rate (TSR) definition, which involves combining the eigen-decomposition of the Jacobian matrix of the vector field with the basic definition of the local TSR given in [13].

The main result of this work is the recognition that the TSR can be computed as a weighted average of the modulus of the eigenvalues of the problem, with weights that depend on (i) the (square of) mode amplitudes, and (ii) the degree of co-linearity between each eigenvector and the vector field.

In addition, because of its additive nature, we introduce a normalized index measuring the relative contribution of each mode to the TSR amplitude. This index can be further combined with the standard participation index of a reaction to a mode amplitude – as defined in CSP – to obtain a direct link between reactions and the TSR. This information can be used for both diagnostics and for the simplification of kinetic mechanisms.

We explore ignition with a planar (2D) model of a spatially homogeneous system according with an isothermal branched-chain explosion model proposed by Williams [20]. We analyze Williams' model on the basis of the CSP and TSR theories. Similar to what is found in detailed kinetic systems, Williams' model develops one pair of eigenvalues, both real and complex, with a positive real part during the explosive regime of ignition.

Our findings show that the TSR is able to track the most energetic scale(s) at all times, and that its own time evolution is smooth.

We carry out the TSR analysis of a planar (2D) model of thermal explosion model – firstly proposed by Semenov in [33,34] – to study the non-isothermal explosion process, which might occur in a reservoir where combustion generates heat released to the ambient through non-adiabatic walls. The Semenov system features an exponential non linearity.

We finally consider a planar (2D) model of a spatially homogeneous system for a non-isothermal branched-chain explosion model proposed by Kapila [22]. The main motivation to analyze this system is to assess the role of the underlying nonlinearities characterizing this model problem. In fact, Williams' model features a polynomial (quadratic) non linearity, whereas Kapila's model combines the exponential dependence of the temperature with the quadratic non linearity of the branching-chain reaction.

The critical comparison of the Williams, Semenov, and Kapila models allowed us to learn about the influence of the nature of the nonlinearities on the qualitative evolution of ignition.

The qualitative results obtained in the analysis of the kinetics of three fuels (methane [18], propane and n-heptane [19]) are very similar to those observed in the Williams model, this suggesting that the dominant behavior is controlled by the polynomial nonlinearities typical of branched-chain reactions. This finding is consistent with the results obtained by Lu [7–10] and Goussis [2,3] in their (independent) analyses.

We will show on the basis of the TSR analysis that during ignition, events are controlled not only by the two modes with positive real parts but also by a few modes having negative real parts. This finding extends what has been already assessed in the literature (e.g., [1–3,7–10]) about the role of modes with positive real part.

While carrying out the TSR analysis of the hydrocarbon system, we observed the TSR to occasionally exhibit significant overshoots with respect to the underlying eigenvalues. We were eventually able to attribute this behavior to the development of nearly co-linear eigenvalues, which occurs when the Jacobian matrix becomes non-normal. To explore in depth this issue, we designed a planar linear system with tunable non-normality, and we analyzed its dynamics with the TSR index.

We found that non-normality is a necessary condition to generate the overshoots, but not sufficient: overshoots of the TSR index manifest only along trajectories exhibiting a strong curvature in non-normal systems.

Finally, the combination of the participation index of reactions to modes and the participation index of modes to the TSR allows us to identify the most important reactions controlling the ignition in the three fuels considered. Among other things, this provides information on the reactions and species that are common in the ignition of the three fuels.

The paper is organized as follows. The theoretical derivation that leads to the definition of the TSR is presented in Section 2. The derivation of the equation for the vector dynamics is offered in Section 2.1. The derivation of the equation for the vector norm dynamics is obtained in Section 2.2. In Section 3, the Williams model is presented and discussed in light of both the CSP and TSR analyses, where we also introduce a two-dimensional definition of the TSR. The definition of the TSR is extended to the N-dimensional case in Section 4. The Participation Index of a mode to the TSR is introduced in Section 5. The TSR analysis of the Semenov model is in Appendix 7. The non-isothermal system by Kapila is defined in Section 8, and analyzed in Section 9. The outcome of the TSR analysis applied to the auto-ignition of three hydrocarbon fuels is illustrated in Section 10. Finally, issues connected with non-normality of the Jacobian are discussed in Section 11.

In Appendix A, we provide the detail of the implementation of the TSR when complex eigenvalues are present.

2. Theory

Consider a chemical kinetic system whose dynamics is described by the a Cauchy problem of the form:

$$\frac{d\boldsymbol{x}}{dt} = \boldsymbol{g}(\boldsymbol{x}), \quad \boldsymbol{x}(0) = \boldsymbol{x}_0, \quad \boldsymbol{x} \in \mathbb{R}^N. \tag{1}$$

The state vector $\boldsymbol{x}$ can be identified with the species concentration vector, the vector field $\boldsymbol{g}(\boldsymbol{x}) = S\mathbf{r}(\boldsymbol{x})$ with the species reaction rate vector, S with the stoichiometric coefficients matrix, $\mathbf{r}(\boldsymbol{x})$ with the net reaction rates vector, and $\boldsymbol{x}_0$ with the initial concentrations vector.

The definition of the state vector $\boldsymbol{x}$ can be extended to include thermo-dynamic variables (temperature, pressure, internal energy, entropy, etc.), this requiring a suitable generalization of the coefficients matrix, to accommodate laws of energy conservation, entropy production, and so forth.

Now, consider two nearby initial conditions, $\boldsymbol{x}_{0,1}$ and $\boldsymbol{x}_{0,2}$, for the point dynamics of Eq. (1), such that:

$$\boldsymbol{x}_{0,2} - \boldsymbol{x}_{0,1} = \boldsymbol{\epsilon} \tag{2}$$

with $\boldsymbol{\epsilon}$ a small (vector) perturbation. Eq. (1) will generate two trajectories $\boldsymbol{x}_1(t)$ and $\boldsymbol{x}_2(t)$. Let us now define the vector $\boldsymbol{v}(t)$ as follows:

$$\boldsymbol{v}(t) := \lim_{|\boldsymbol{\epsilon}|\to 0} \frac{(\boldsymbol{x}_2 - \boldsymbol{x}_1)}{|\boldsymbol{\epsilon}|}. \tag{3}$$

By construction, the vector $\boldsymbol{v}(t)$ belongs to the tangent bundle at $\mathbf{x}_{0,1}$. The vector $\boldsymbol{v}(t)$ is a scaled measure, at time t, of the difference between the two trajectories emanating from the two initial conditions.

2.1. Derivation of the vector dynamics equation

The characterization of the dynamical features defined by Eq. (1) can be carried out by viewing the dynamical system as a generalized "advection machine" of all the possible geometric entities