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Tangential Stretching Rate Analysis of Ignition in a Non Premixed System

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Outline of the talk

We analyze ignition phenomena by resorting to the stretching rate concept introduced in the study of dynamical systems.

We construct a Tangential Stretching Rate (TSR) parameter by combining:

stretching rate concept

and

local tangent space decomposition in eigenmodes

TSR identifies unambiguously the **most energetic scale** at a given space location and time instant

TSR is a **state function** (TSR depends only on local mixture composition, temperature, and pressure)

TSR can be readily computed during the post processing of computed reactive flow fields, both for
1) Spatially homogeneous (auto-ignition in batch reactors) and
2) In-homogenous systems (premixed and non premixed systems)

We verified the properties of TSR with reference to hydro-carbon oxidation kinetics:

1) Ignition in batch reactors
2) Unsteady flamelet model
   a. Ignition
   b. Quenching
   c. Re-ignition

We will discuss how to extend the definition of the TSR to PDEs models including transport as well as kinetics

Stretching Rate Analysis

$$\frac{d\vec{z}}{dt} = \vec{g}(\vec{z}) \qquad ICs : \vec{z}(0) = \vec{z}_0$$

State vector z: species concentration vector
Vector field: g (z) = S r (z) with the species reaction rate vector
S : stoichiometric coefficients matrix
r : net reaction rates vector
z0 : initial concentrations vector

Consider two nearby trajectories: $\vec{\mathbf{z}} = \vec{\mathbf{z}}_0 + \varepsilon$

Define a scaled vector distance between the two as: $\vec{\mathbf{v}} := \lim_{\varepsilon \to 0} \left(\frac{\vec{\mathbf{z}}_2 - \vec{\mathbf{z}}_1}{\varepsilon} \right)$

Vector Dynamics $\frac{d\vec{\mathbf{v}}}{dt} = \mathbf{Jac_g}(\mathbf{z})\vec{\mathbf{v}}(t) \qquad \vec{\mathbf{v}}(0) = \vec{1} \qquad \mathbf{Jac_g} := \frac{\partial \mathbf{g}(\mathbf{z})}{\partial \mathbf{z}}$

Vector Norm Dynamics $\frac{d\left\|\vec{\mathbf{v}}\right\|^2}{dt} = 2\frac{\vec{\mathbf{v}}^T \mathbf{Jac_g} \vec{\mathbf{v}}}{\left\|\vec{\mathbf{v}}\right\|^2}\left\|\vec{\mathbf{v}}\right\|^2 \qquad \left\|\vec{\mathbf{v}}\right\|(0) = 1$

Stretching Rate along any $\vec{\mathbf{u}}$ $\omega_{\vec{\mathbf{u}}} := \vec{\mathbf{u}}^T \mathbf{Jac_g} \vec{\mathbf{u}} \qquad \vec{\mathbf{u}} := \frac{\vec{\mathbf{v}}(t)}{\left\|\vec{\mathbf{v}}\right\|}$

Stretching Rate and CSP decomposition (ODEs)

Stretching rate along vector field direction

TSR can be recast after CSP expansion of J = A Λ B and g = Σ_i $\mathbf{a}_i$ f^i

$$\omega_\tau = \vec{\tau}^T \cdot J_g \cdot \vec{\tau} = \frac{1}{g^2}\left(\vec{g}^T \cdot A\Lambda B \cdot \vec{g}\right) = \frac{1}{g^2}\sum_{i=1}^{N}\left(\vec{g}^T \cdot \vec{a}_i\right)\lambda_i f^i = \sum_{i=1}^{N} W_i \lambda_i$$

$$f^i := \vec{b}^i \cdot \vec{g}$$

$$\omega_\tau := \sum_{i=1}^{N} \bar{W}_i \,\mathrm{Sgn}(\mathrm{Re}(\lambda_i))\left|\lambda_i\right|, \qquad \bar{W}_i = \frac{W_i}{\sum_{j=1}^{N}\left|W_j\right|}$$

Def.#1:Accounting for angle/phase between $\vec{g}$ and $\vec{a}_i$

$$W_i := \frac{f^i}{g}\frac{\vec{g}^T \cdot \vec{a}_i}{g}$$

Def.#2:Not accounting for angle/phase between $\vec{g}$ and $\vec{a}_i$

$$W_i := \left(\frac{f^i}{g}\right)^2$$

TSR is a weighted sum of the eigenvalues

Participation indices related to tangential stretching rate (ODEs)

CSP PI index between reaction & mode

$$f^i = \mathbf{b}^i \cdot \mathbf{g} = \sum_{k=1,Nr} (\mathbf{b}^i \cdot \mathbf{S}_k) r^k$$

$$P_k^i = \frac{\left|(\mathbf{b}^i \cdot \mathbf{S}_k) r^k\right|}{\sum_{k'=1,Nr} \left|(\mathbf{b}^i \cdot \mathbf{S}_{k'}) r^{k'}\right|}$$

TSR PI index between mode & TSR

$$\omega_\tau = \sum_{i=1,N} \tilde{W}_i \left|\lambda_i\right|$$

$$P_i^{\omega_\tau} = \frac{\left|\tilde{W}_i \left|\lambda_i\right|_i\right|}{\sum_{j=1,N} \left|\tilde{W}_j \left|\lambda_j\right|\right|}$$

PI index between reaction & TSR

$$P_k^{\omega_\tau} = P_i^{\omega_\tau} P_k^i$$

Modes with a large $P_i^{\omega_\tau}$ are the most contributing to the ω_τ scale (energy containing)

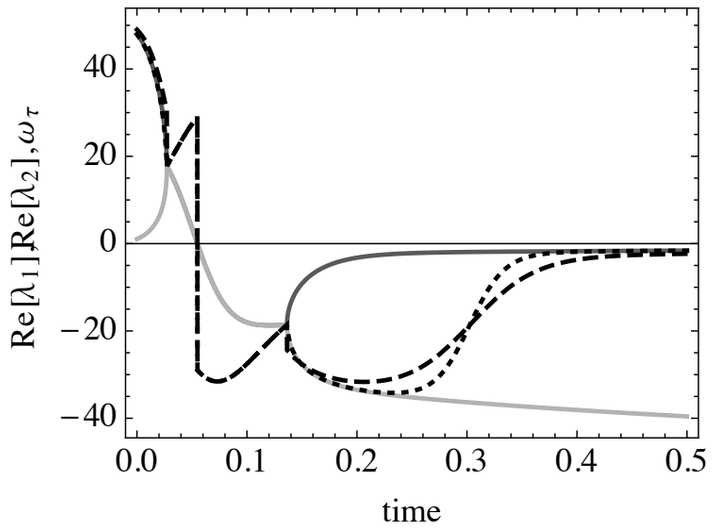
Reactions with a large P_k^i are the most contributing to the i-th mode

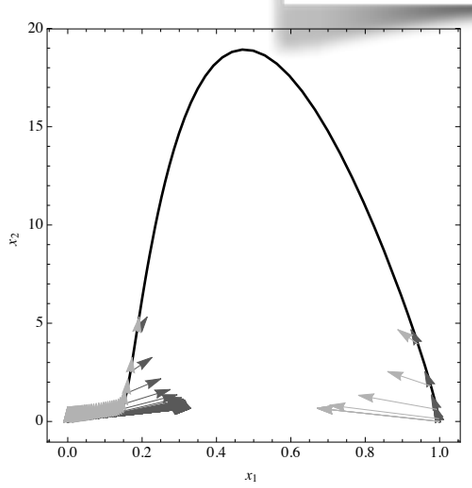
Reactions with a large $P_k^{\omega_\tau} = P_i^{\omega_\tau} P_k^i$ are the most contributing to the ω_τ scale

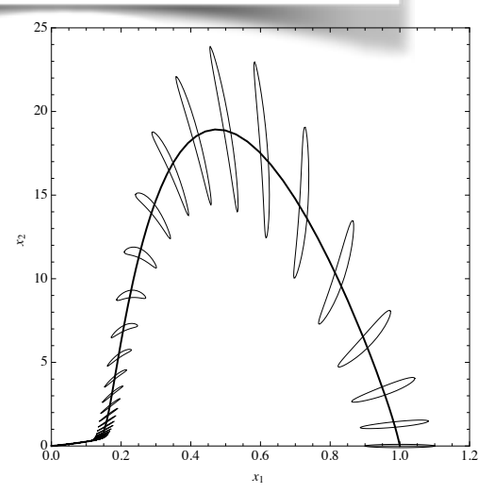
Williams model

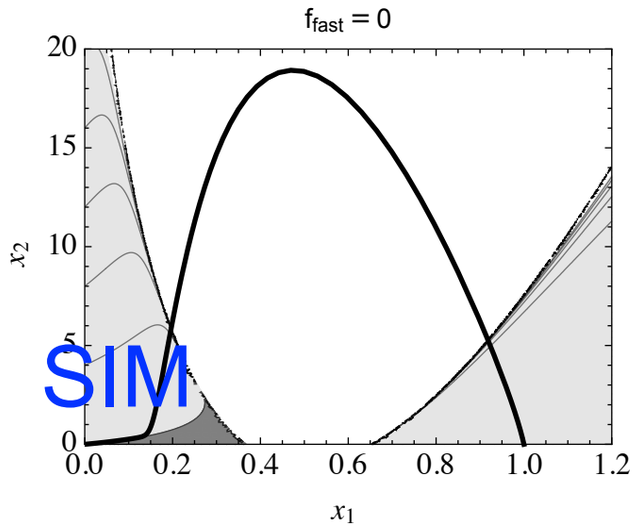
Branched-chain reactions (polynomial) system

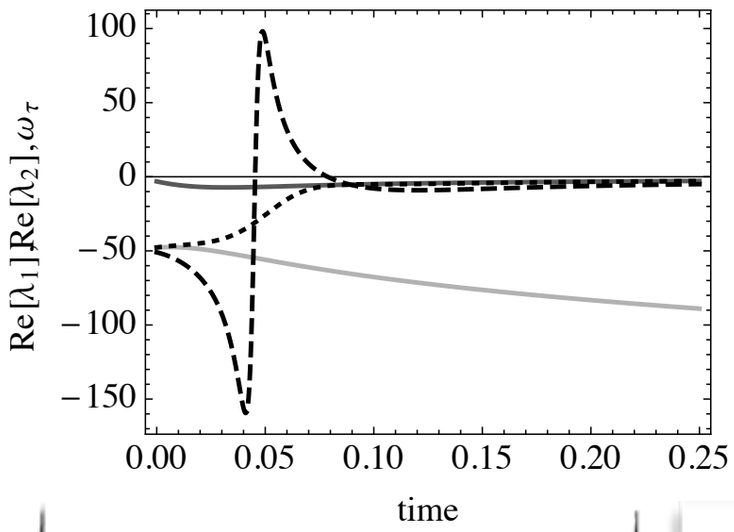
$$R \to C \quad \text{(initiation)}$$
$$R + C \to \alpha\, C + P \quad \text{(propagation)}$$
$$C \to P \quad \text{(termination)}$$

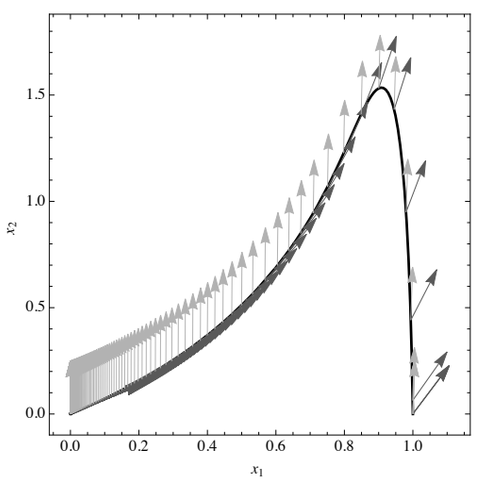


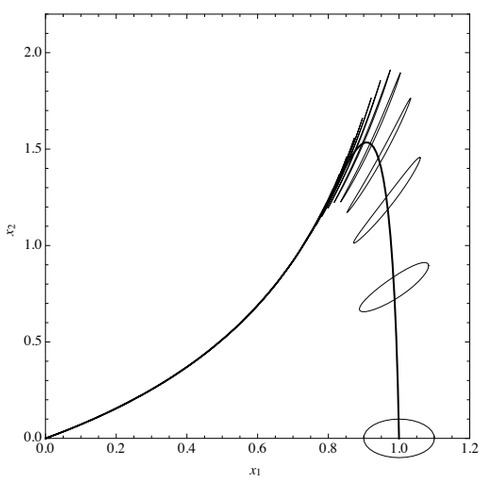


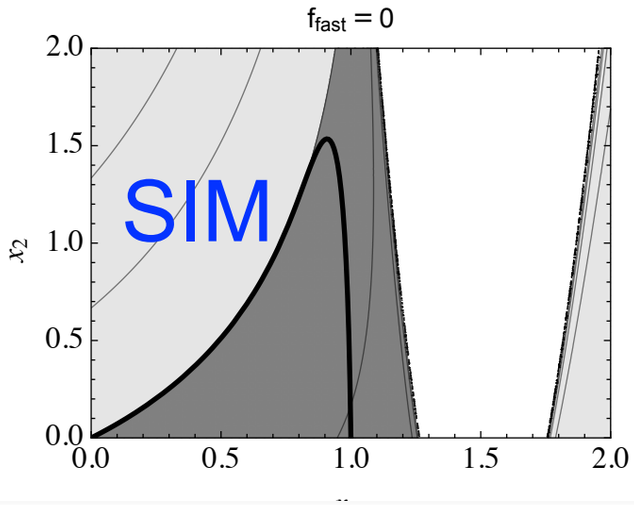










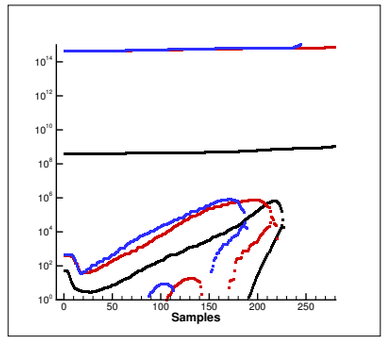


$$\frac{dx_1}{d\tau} = -x_1 - x_1 x_2$$
$$\epsilon \frac{dx_2}{d\tau} = x_1 + (\alpha - 1) x_1 x_2 - \gamma x_2$$
$$\gamma \frac{dx_3}{d\tau} = \gamma x_2 + x_1 x_2,$$
$$x_1(0) = 1, \quad x_2(0) = 0, \quad x_3(0) = 0$$

- Ignition might initiate with a pair of real positive eigenvalues
- Transition from positive to negative sign can occur while crossing a region of complex eigenvalues
- Crossing a region of complex eigenvalues can occur with a change of sign (positive to negative)
- Non-normality in subcritical regime results in overshoots of the TSR index associated with the strong curvature of the trajectory in the phase space

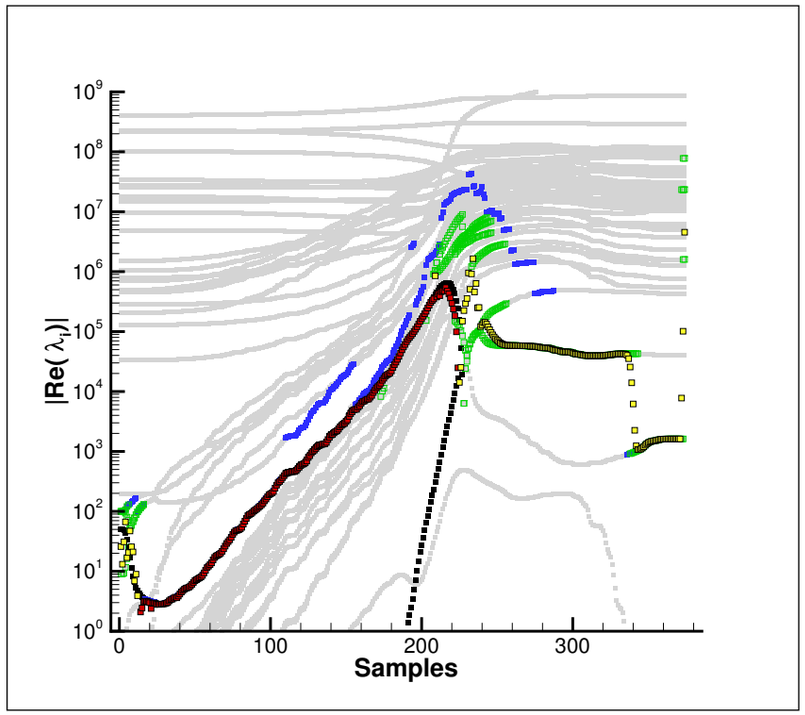
TSR Analysis of Autoignition

Legend
black: pos eigvals
blue: first non-exhausted mode
red/yellow: pos/neg TSR
green: active subspace

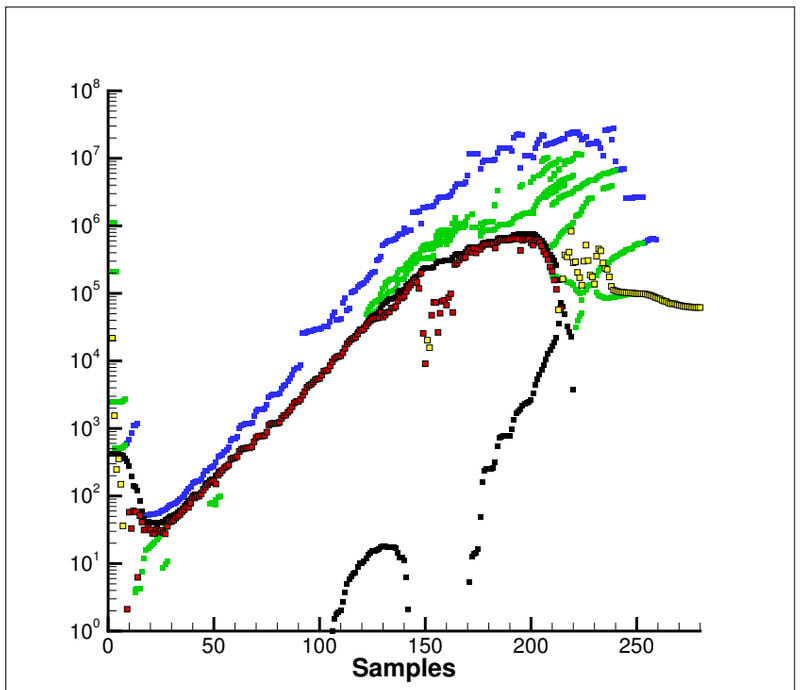


Batch Reactor Iso-choric Autoignition

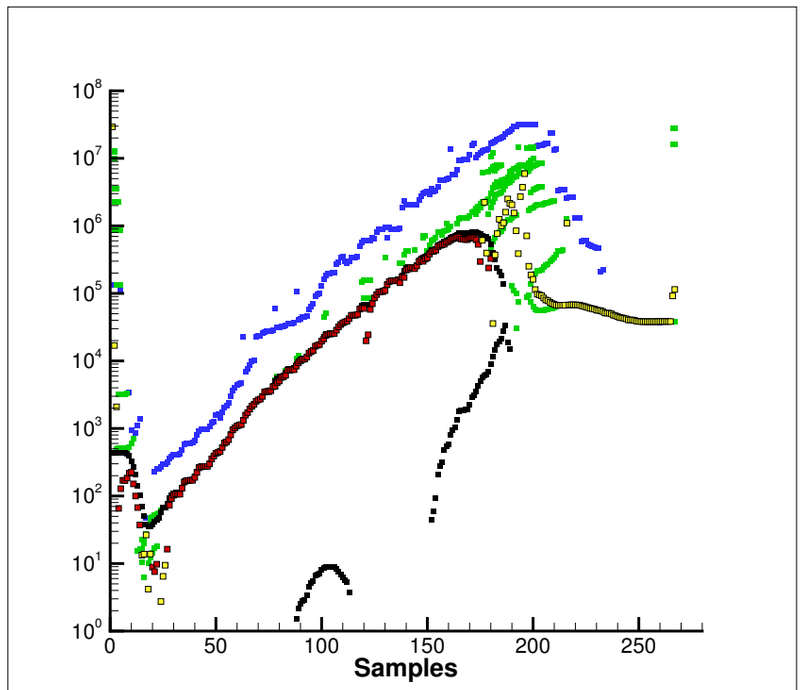
T_0=1000K p_0=1 atm, stoichiometric, non-diluted air



Methane (GRI 3.0)



Propane



n-Heptane

- Same qualitative behavior for all fuels: a pair of positive eigenvalues merge as in Williams model

- Chain branching: TSR coincides with the **fast (largest) positive eigenvalue**
- Thermal explosion: across the merging, **TSR is contributed by some dissipative modes**
- Recombination phase: TSR tracks the driving dissipative mode

- Max value of TSR is same order [tau ~1/(2 x 105) s ~ 50 x 10-6 s = 50 microsec]
- Explosive time scales are much slower than the fastest time scales because all other faster time scales become exhausted.

Stretching Rate and CSP decomposition

Extension to PDEs

$$\frac{d\vec{z}}{dt} = \vec{L}(\vec{z}) + \vec{g}(\vec{z}) \qquad ICs + BCs$$

$$\omega_\tau = \vec{\tau}_g \cdot J_g \cdot \vec{\tau}_{L+g} = \frac{1}{|\vec{g}\,||\vec{L}+\vec{g}\,|}\left(\vec{g}\cdot A\Lambda B\cdot(\vec{L}+\vec{g})\right)$$

$$= \frac{1}{|\vec{g}\,||\vec{L}+\vec{g}\,|}\sum_{i=1}^{N}\left(\vec{g}\cdot\vec{a}_i\right)\lambda_i h^i = \sum_{i=1}^{N} W_i\lambda_i$$

$$h^i := \vec{b}^{\,i}\cdot(\vec{L}+\vec{g}) \qquad W_i := \frac{h^i(\vec{g}^{\,T}\cdot\vec{a}_i)}{|\vec{g}\,||\vec{L}+\vec{g}\,|}$$

Weights depend on transport as well as kinetics

$$\omega_\tau := \sum_{i=1}^{N}\bar{W}_i\,\mathrm{Sgn}(\mathrm{Re}(\lambda_i))|\lambda_i|, \qquad \bar{W}_i = \frac{W_i}{\sum_{j=1}^{N}|W_j|}$$

Participation indices related to tangential stretching rate (PDEs)

CSP PI index between reaction & mode

$$h^i = \mathbf{b}^i \cdot (\mathbf{L} + \mathbf{g}) = \sum_{j=1,N} (\mathbf{b}^i \cdot \mathbf{e}_j) L^j + \sum_{k=1,Nr} (\mathbf{b}^i \cdot \mathbf{S}_k) r^k$$

$$P_k^i = \frac{\left|(\mathbf{b}^i \cdot \mathbf{S}_k) r^k\right|}{\sum_{j'=1,N} \left|(\mathbf{b}^i \cdot \mathbf{e}_{j'}) L^{j'}\right| + \sum_{k'=1,Nr} \left|(\mathbf{b}^i \cdot \mathbf{S}_{k'}) r^{k'}\right|}$$

$$P_k^i = \frac{\left|(\mathbf{b}^i \cdot \mathbf{e}_j) L^j\right|}{\sum_{j'=1,N} \left|(\mathbf{b}^i \cdot \mathbf{e}_{j'}) L^{j'}\right| + \sum_{k'=1,Nr} \left|(\mathbf{b}^i \cdot \mathbf{S}_{k'}) r^{k'}\right|}$$

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PI index between reaction & TSR

$$P_k^{\omega_\tau} = P_i^{\omega_\tau} P_k^i$$

Modes with a large $P_i^{\omega_\tau}$ are the most contributing to the ω_τ scale (energy containing)

Reactions with a large P_k^i are the most contributing to the i-th mode

Reactions with a large $P_k^{\omega_\tau} = P_i^{\omega_\tau} P_k^i$ are the most contributing to the ω_τ scale

Flamelet model

$$\frac{\partial Y_\alpha}{\partial t} = \frac{1}{2}\chi\frac{\partial^2 Y_\alpha}{\partial \xi^2} + \frac{\dot{\omega}_\alpha}{\rho}, \quad \alpha = 1, N_s$$

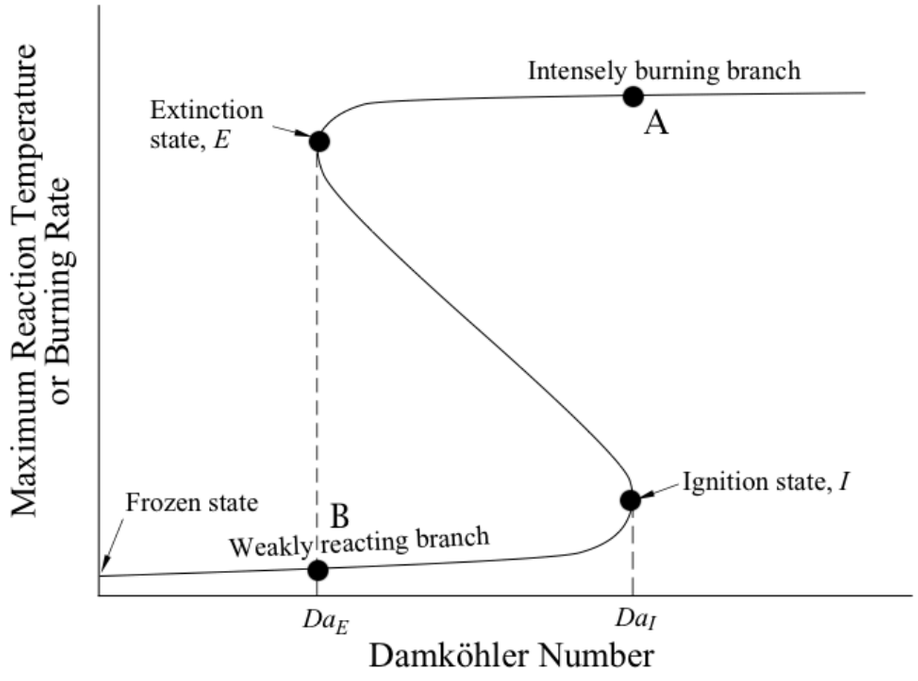
$$\frac{\partial T}{\partial t} = \frac{1}{2}\chi\left[\frac{\partial^2 T}{\partial \xi^2} + \frac{1}{c_p}\frac{\partial c_p}{\partial \xi}\frac{\partial T}{\partial \xi}\right] + \frac{\dot{\omega}_T}{c_p \rho}$$

where ξ is the mixture fraction

$\dot{\omega}_\alpha$, $\dot{\omega}_T$ are the species and temperature source terms

N_s is the number of species in the mixture

$\chi = 2D(\partial\xi / \partial x_i)^2$ is the scalar dissipation rate

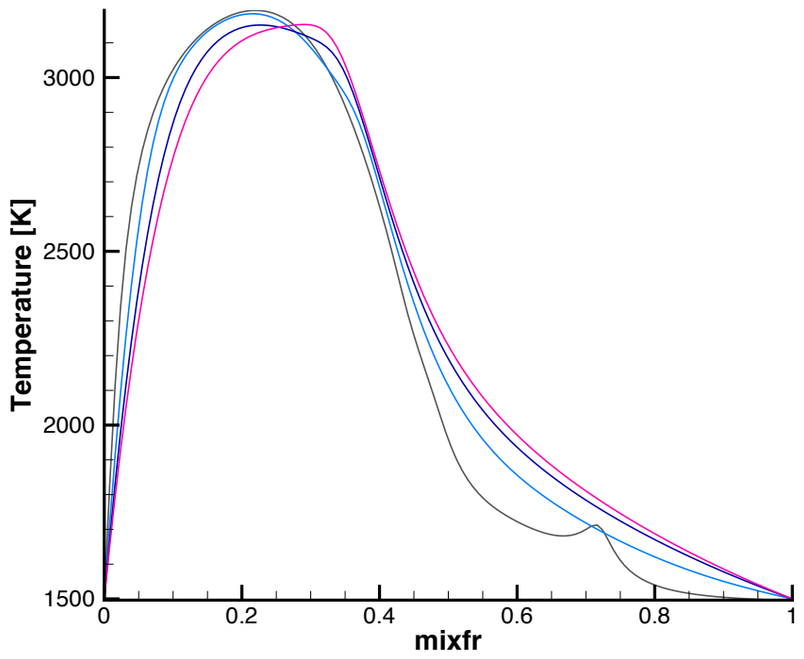


S-shaped temperature behavior as a function of the Damkohler number, Da:

$$Da = \frac{1}{\tau_c \chi} = \frac{1}{\tau_c D(\partial\xi/\partial x_i)^2}$$

High Scalar Dissipation yields:
- longer ignition delay time
- faster diffusion wave
- lower max temperature at steady state

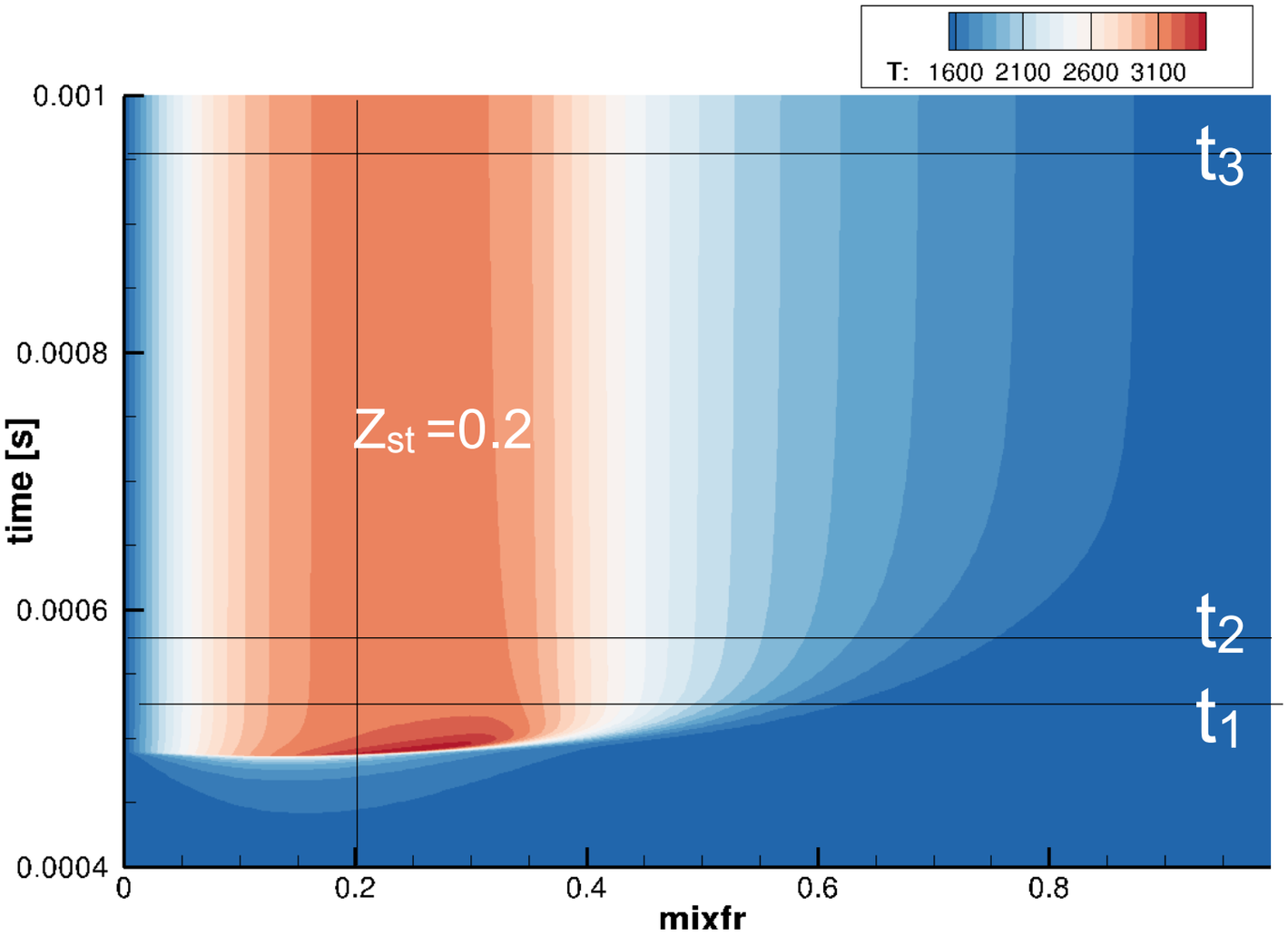
Ignition occurs only below a Limiting Scalar Dissipation ($N_{ignition}$)
Quenching occurs only above a Limiting Scalar Dissipation ($N_{quenching}$)

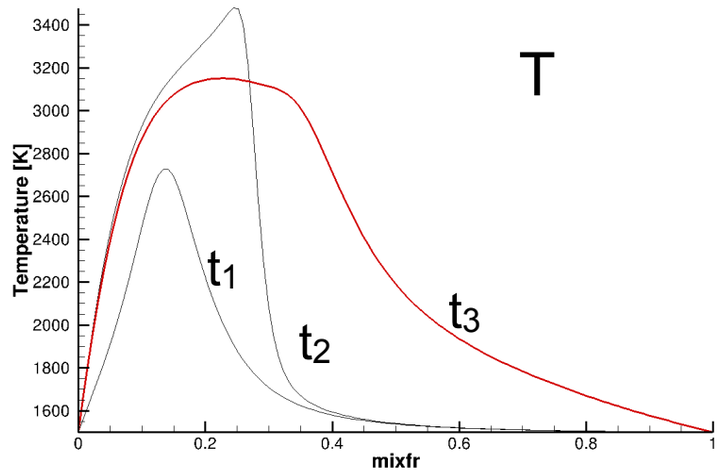


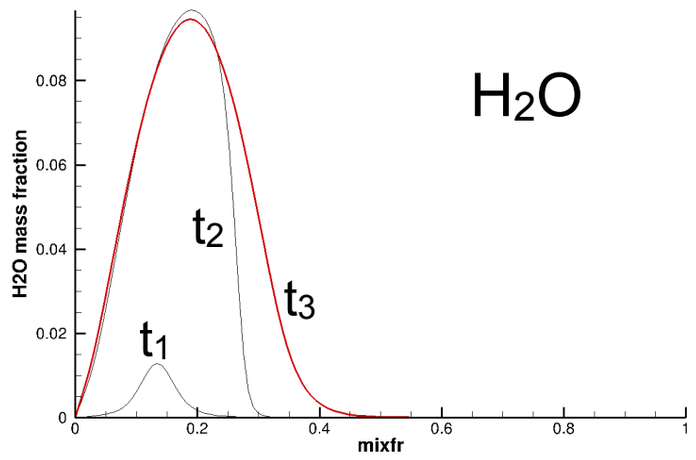
Unsteady Flamelet Dynamics N=250s^{-1}

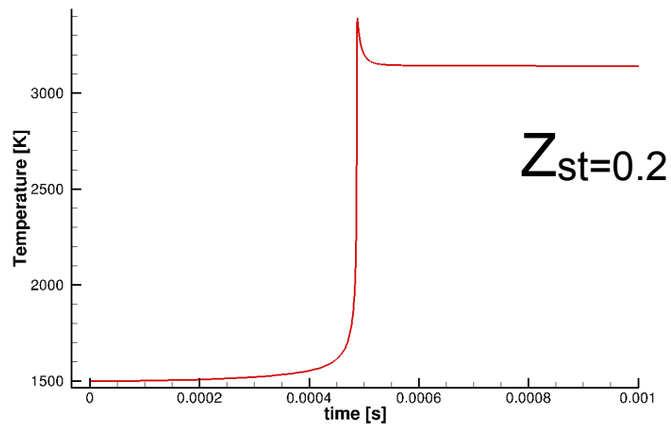
- ICs T(Z) = 1500 K $Y_{CH4}(Z)=Z$ $Y_{O2}(Z=0)=1-Z$

- Chemical mechanism: gri3.0 (53/356)
- Space resolution: 128 cells
- Numerical integration: BDF implicit technique (DVODE)

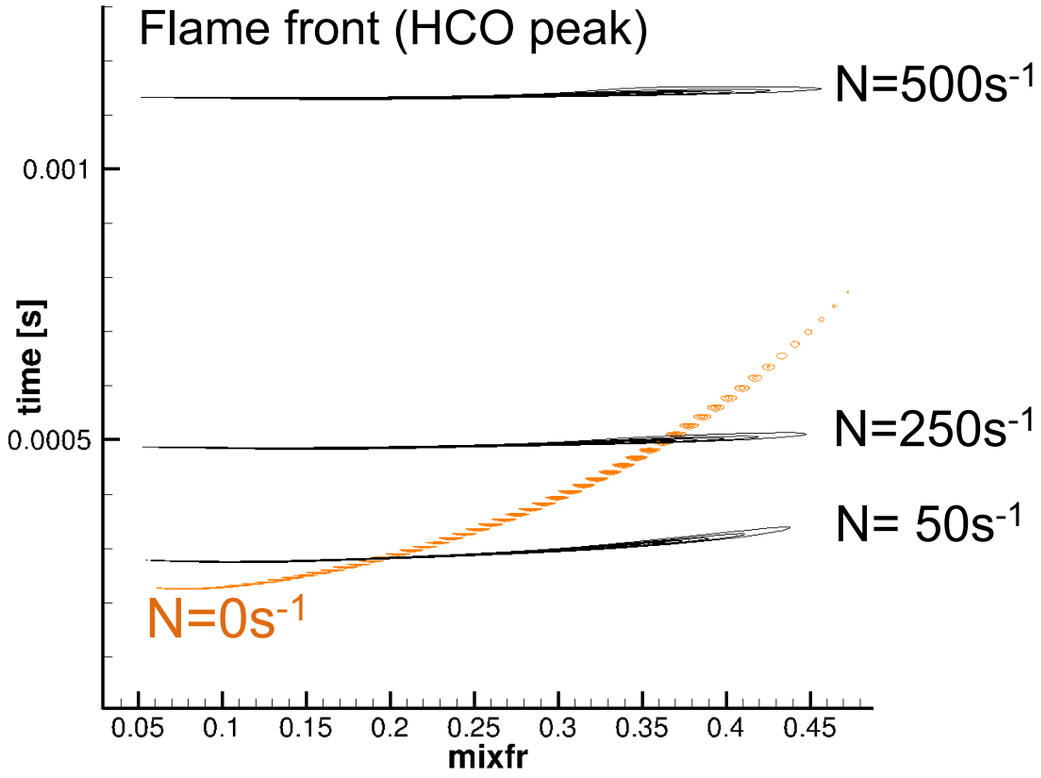








Role of Scalar Dissipation in Flamelet Ignition



High Scalar Dissipation yields:

- longer ignition delay time
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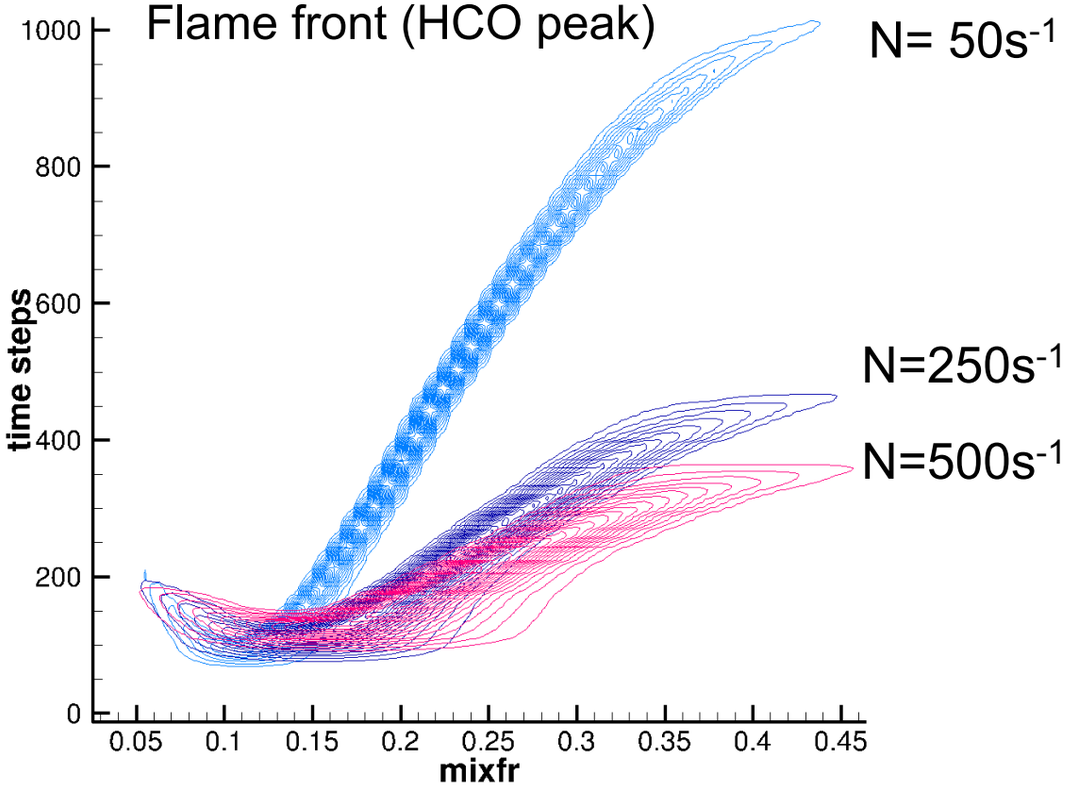
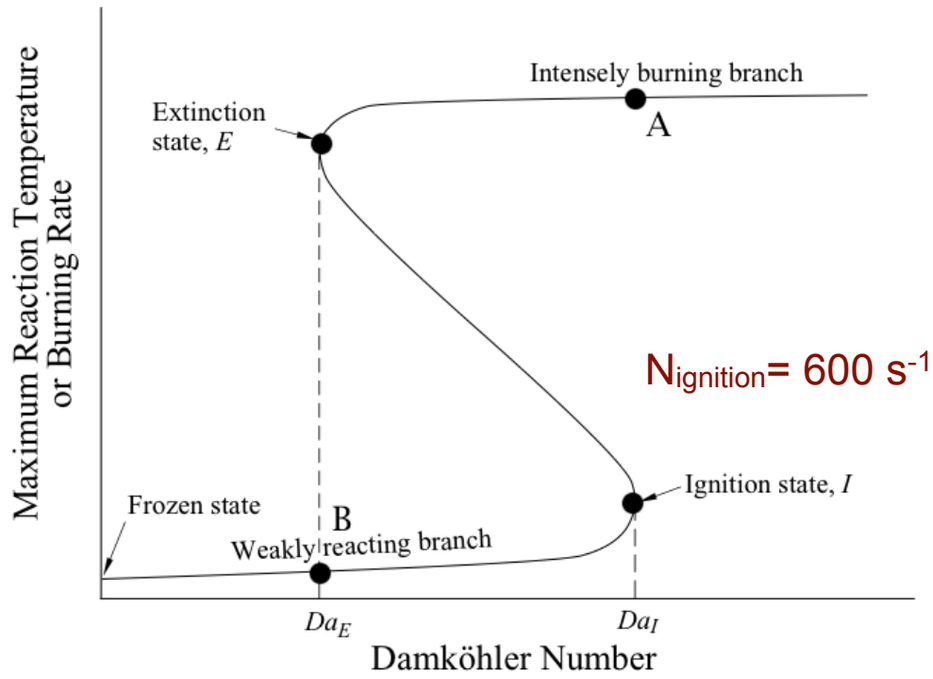
Ignition occurs only below a Limiting Scalar Dissipation ($N_{ignition}$)

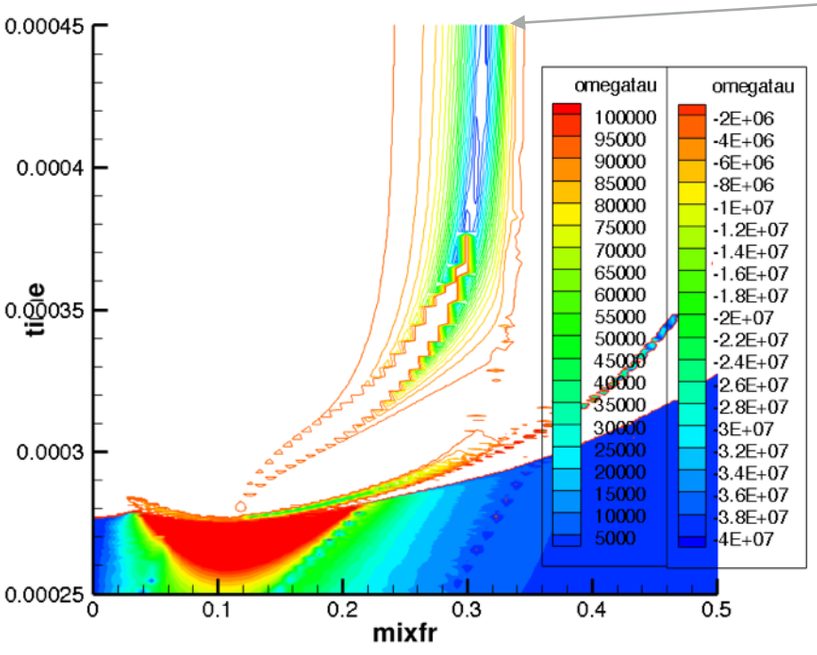


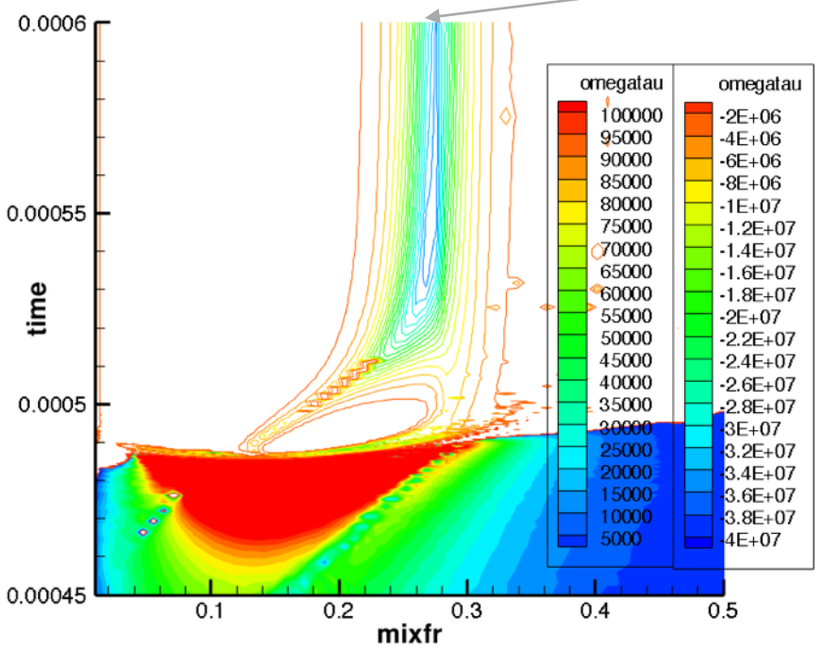


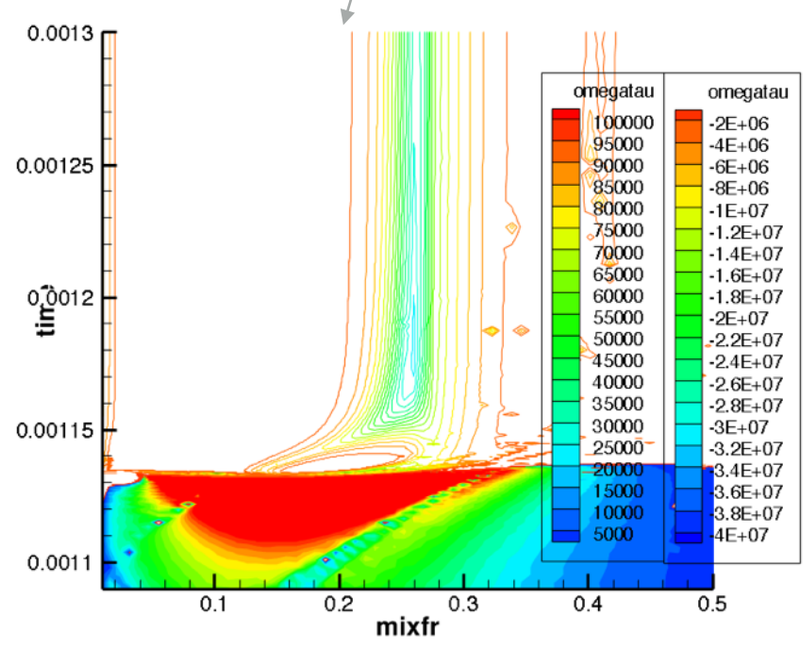
TSR Analysis of Ignition

TSR v CEMA

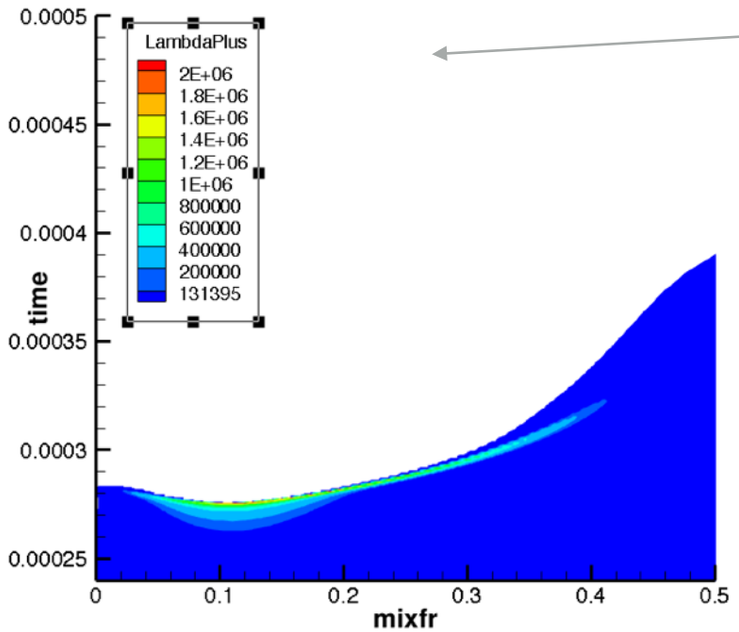
Negative TSR



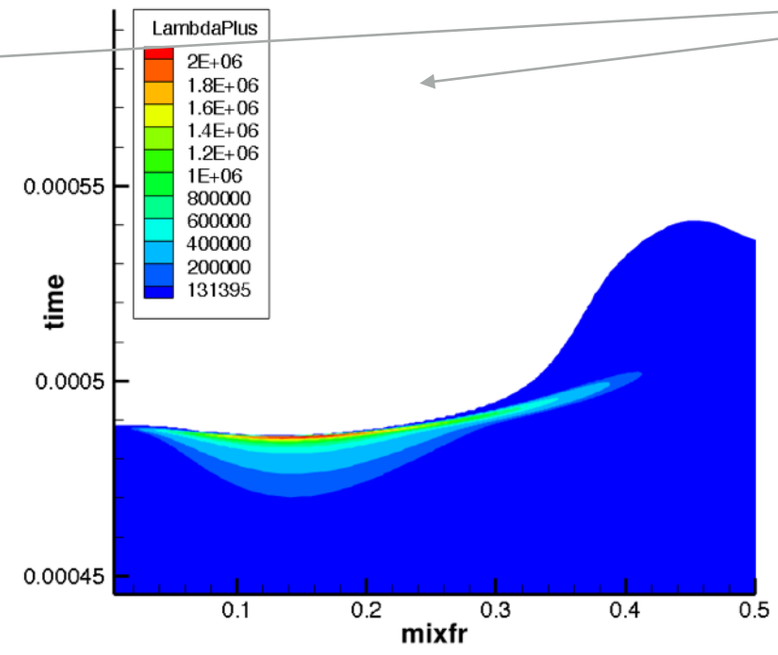




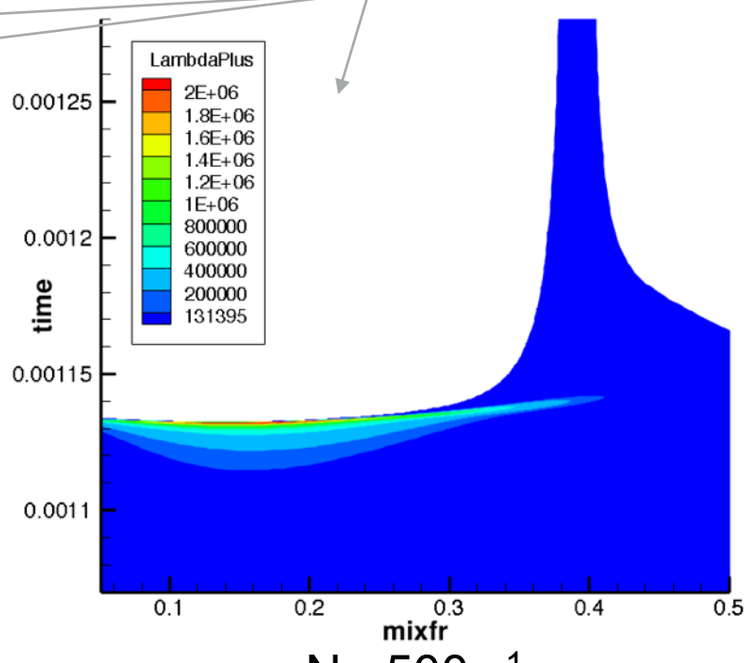
LambdaPlus not defined



N= 50s^{-1}

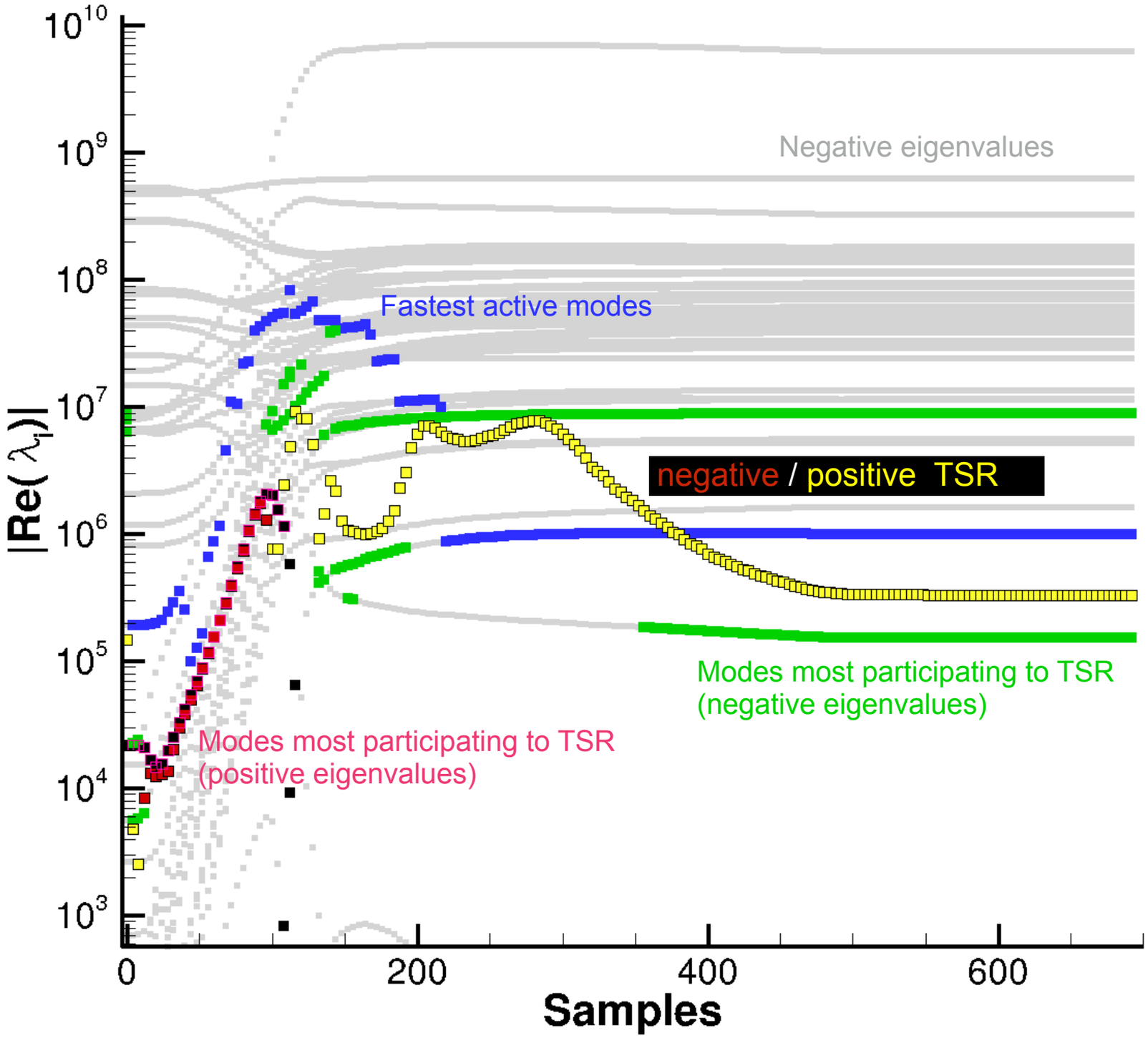


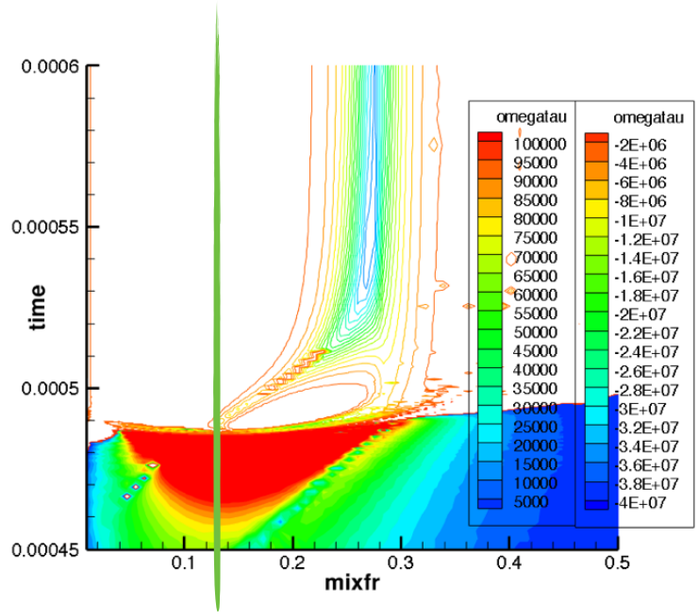
N= 250s^{-1}



N= 500s^{-1}

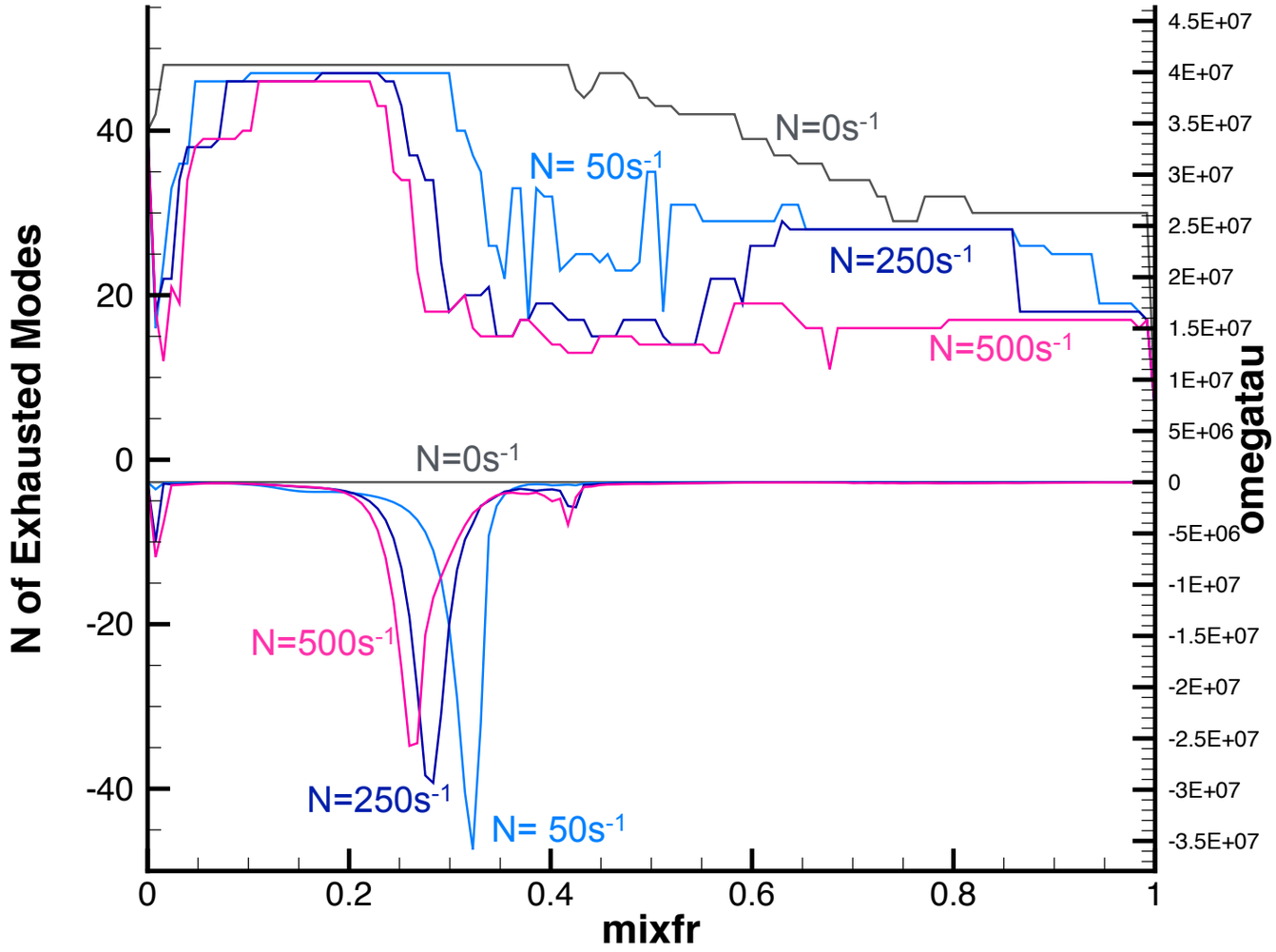
CSP analysis and TSR of the ignition

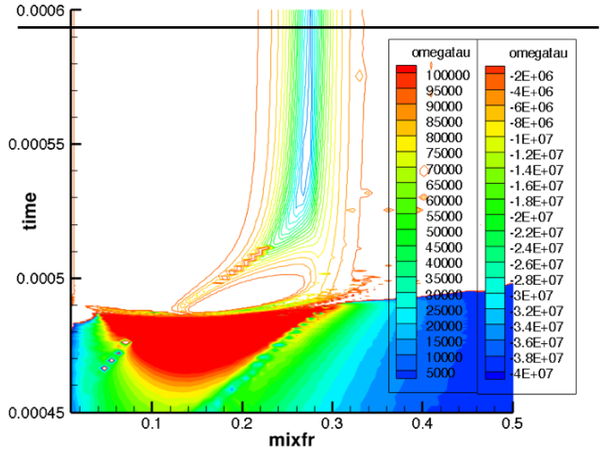


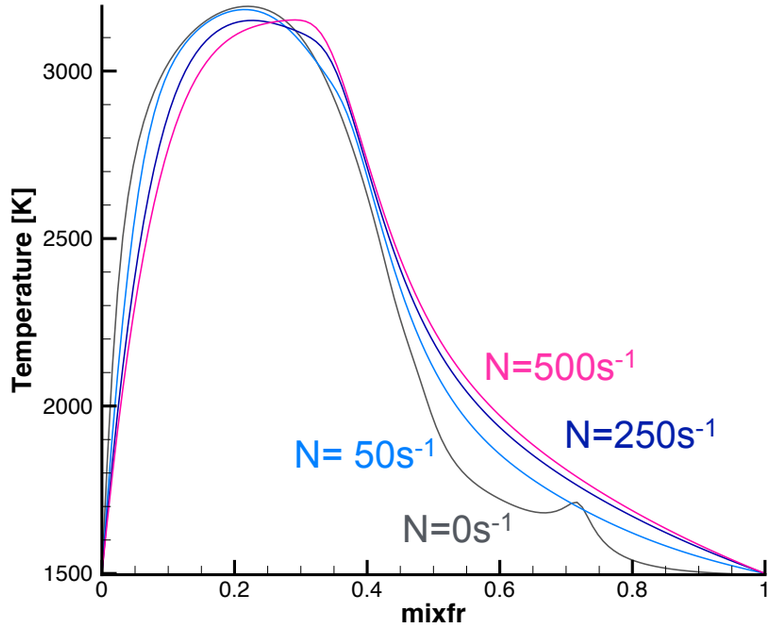


- N=250s-1
- $Z=Z_{mr}$ = 0.13

TSR at Steady-State







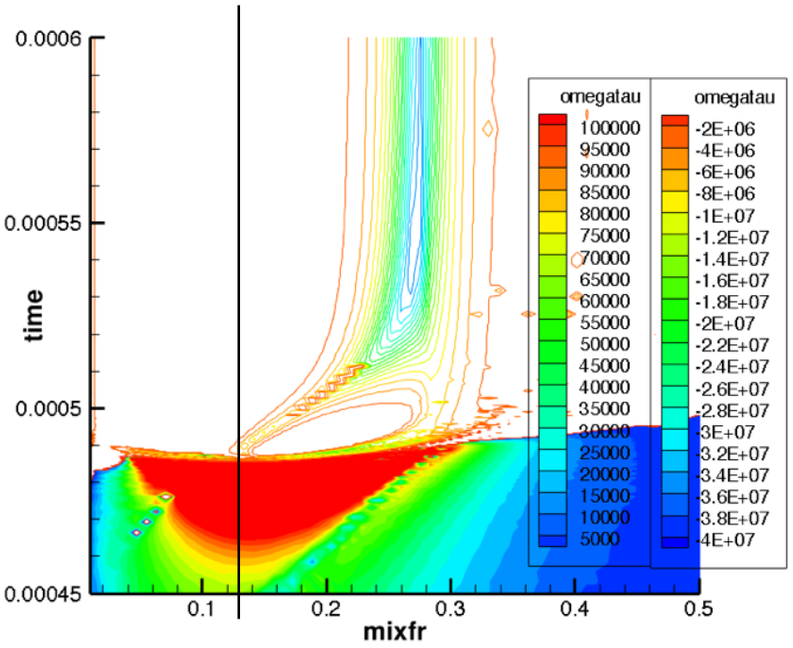
Diffusion at stationary conditions drives kinetics off equilibrium

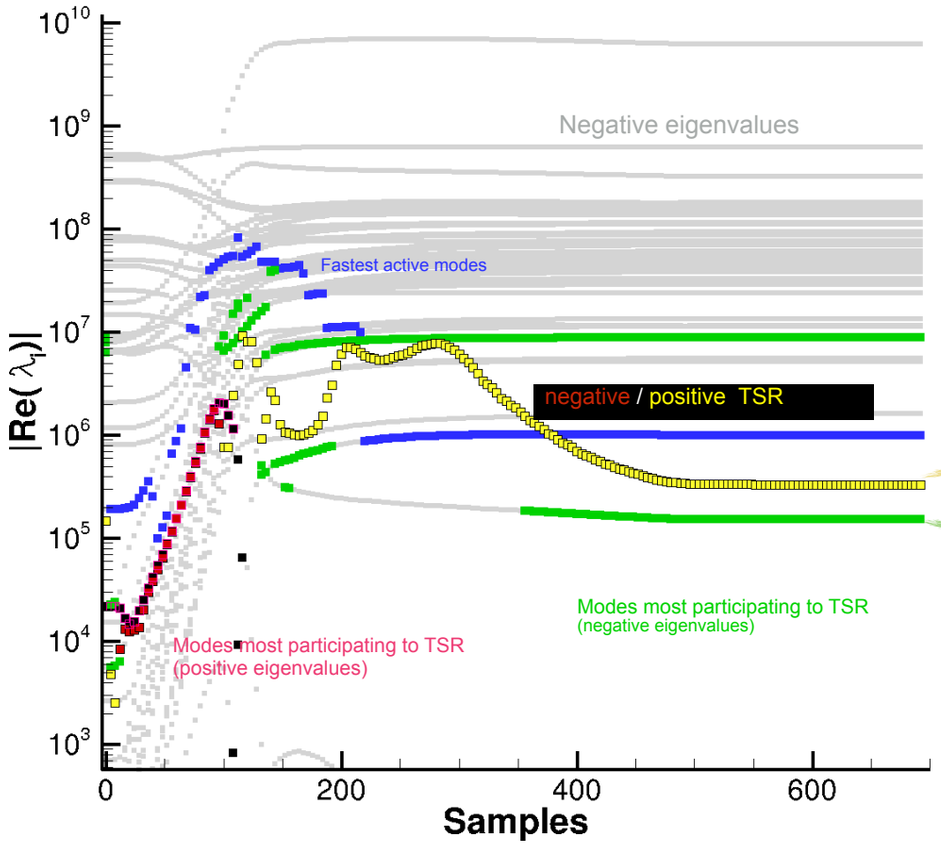
$\vec{b}^{\,i} \cdot (\vec{L} + \vec{g}) = 0$ steady state

$\vec{b}^{\,i} \cdot \vec{g} = -\vec{b}^{\,i} \cdot \vec{L}$ chemical non equilibrium $\quad \omega_\tau \sim \sum_{act} (\vec{g} \cdot \vec{a}_{act})(\vec{b}^{\,act} \cdot \vec{g}) \,|\, \lambda_{act} \,| \;= \sum_{act} (\vec{g} \cdot \vec{a}_{act})(-\vec{b}^{\,act} \cdot \vec{L}) \,|\, \lambda_{act} \,|$

$\vec{b}^{\,i} \cdot \vec{g} = 0$ chemical equilibrium $\quad \omega_\tau \sim (\vec{g} \cdot \vec{a}_{slowest})(\vec{b}^{\,slowest} \cdot \vec{g}) \,|\, \lambda_{slowest} \,|$

CSP analysis and TSR of ignition





MODE No 42

R_fwd#	3 O+H2<=>H+OH	7.84E-02
R_bwd#	3 O+H2<=>H+OH	7.82E-02
R_fwd#	38 H+O2<=>O+OH	2.47E-01
R_bwd#	38 H+O2<=>O+OH	2.44E-01
R_fwd#	84 OH+H2<=>H+H2O	1.03E-01
R_bwd#	84 OH+H2<=>H+H2O	1.03E-01
R_fwd#	86 2OH<=>O+H2O	5.65E-02
R_bwd#	86 2OH<=>O+H2O	5.66E-02

MODE No 47

R_fwd#	3 O+H2<=>H+OH	5.43E-02
R_bwd#	3 O+H2<=>H+OH	5.41E-02
R_fwd#	35 H+O2+H2O<=>HO2+H2O	9.03E-02
R_bwd#	35 H+O2+H2O<=>HO2+H2O	5.21E-02
R_fwd#	**85 2OH(+M)<=>H2O2(+M)**	**5.26E-02**
R_fwd#	86 2OH<=>O+H2O	7.93E-02
R_bwd#	86 2OH<=>O+H2O	7.94E-02
R_fwd#	88 OH+HO2<=>O2+H2O	7.36E-02
R_bwd#	88 OH+HO2<=>O2+H2O	6.81E-02

Mode 42 for 47%

$\lambda_{42} \simeq -10^{7}$

$W_{42} \simeq 1.75 \times 10^{-2}$

TSR = -332415.6

2 Modes mostly participating to TSR

Mode 47 for 44%

$\lambda_{47} \simeq -1.5 \times 10^{5}$

$W_{47} \simeq 0.98$

Role of Scalar Dissipation in Flamelet Reignition/Quenching

Initial condition
Steady-state solution:

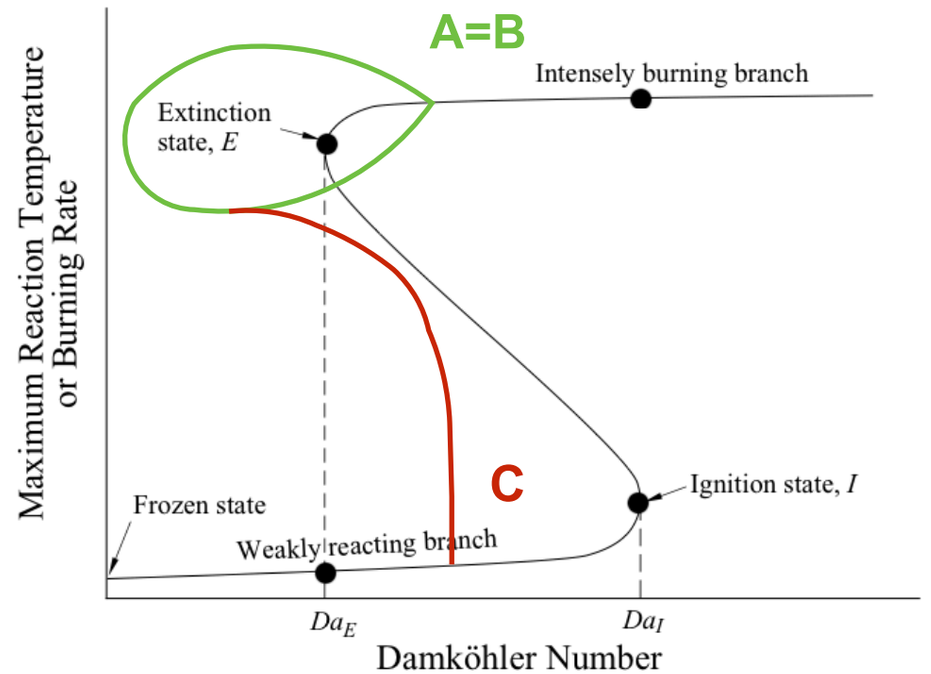
T(Ox) = T(Fu)=300 K
N=10000s^{-1}

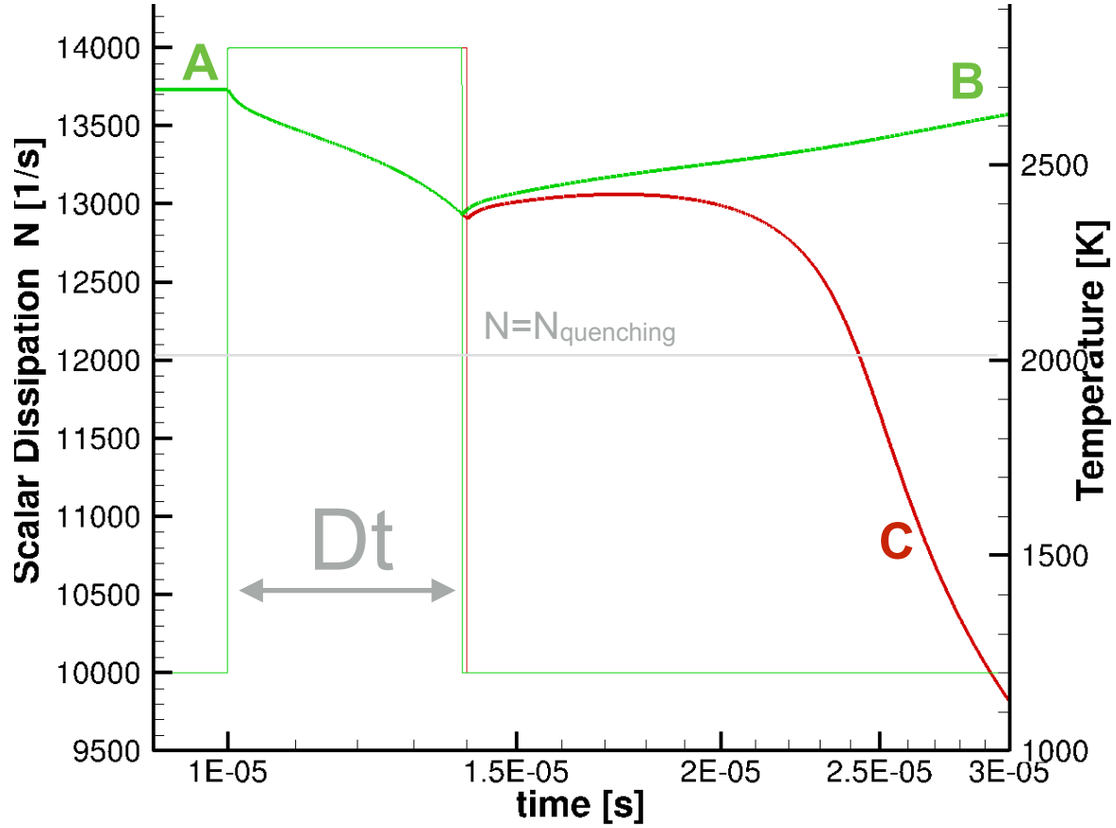
40% increase in scalar dissipation rate for a limited time Dt

Nmax =14000 > 12000 = Nquenching

Case 1 reignition
Dt1 = 3.9 10-6 s

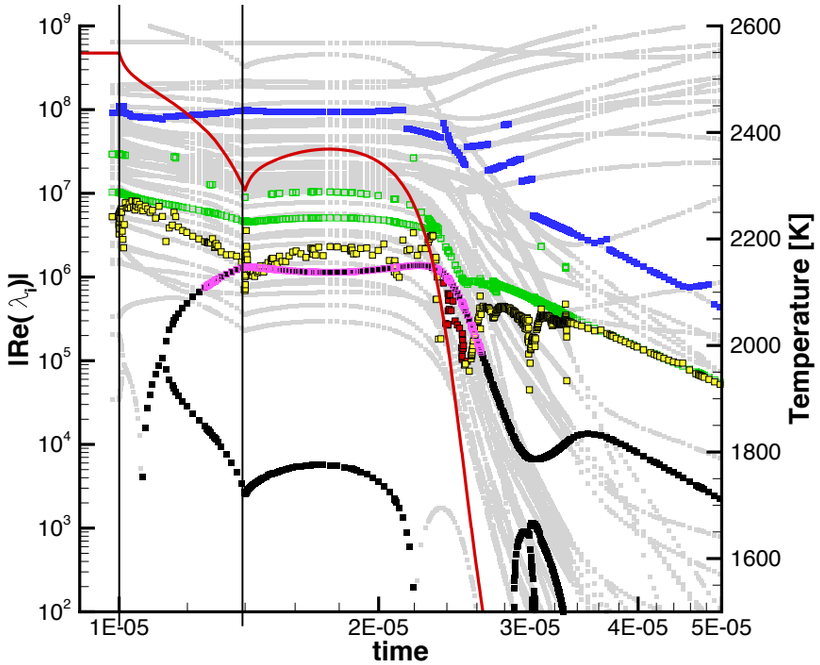
Case 2 quenching
Dt1 = 4.0 10-6 s

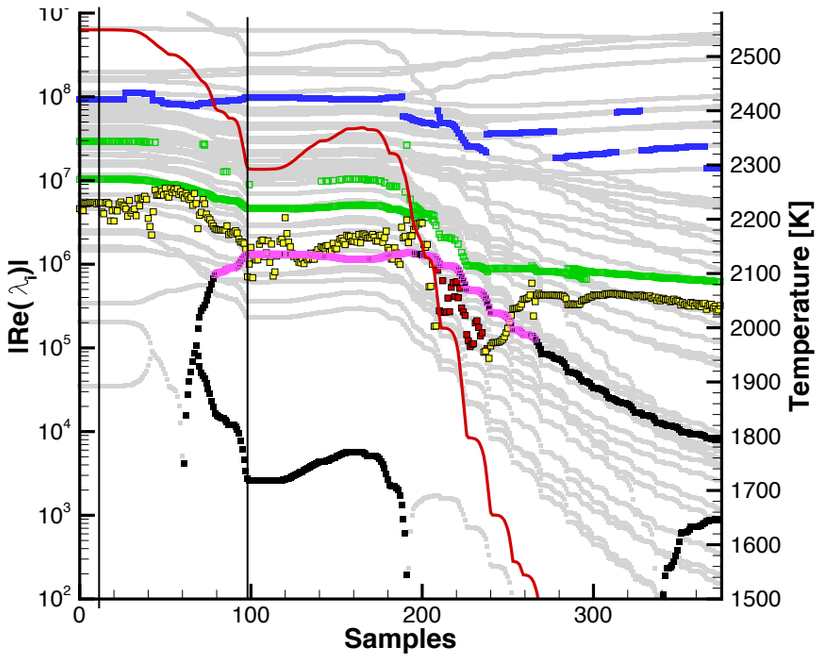


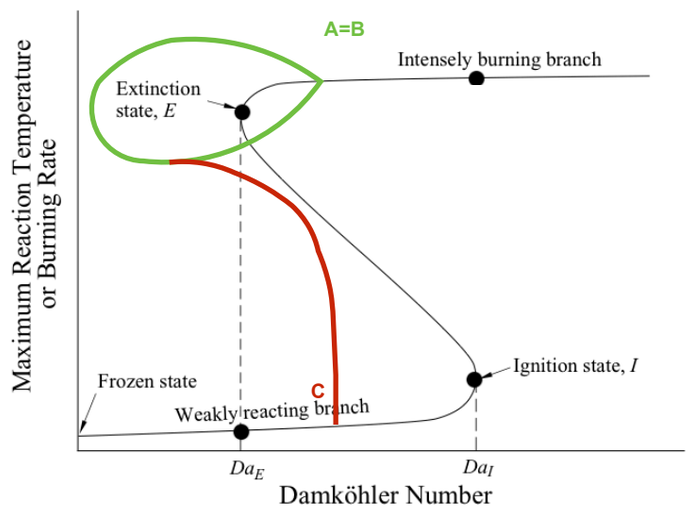


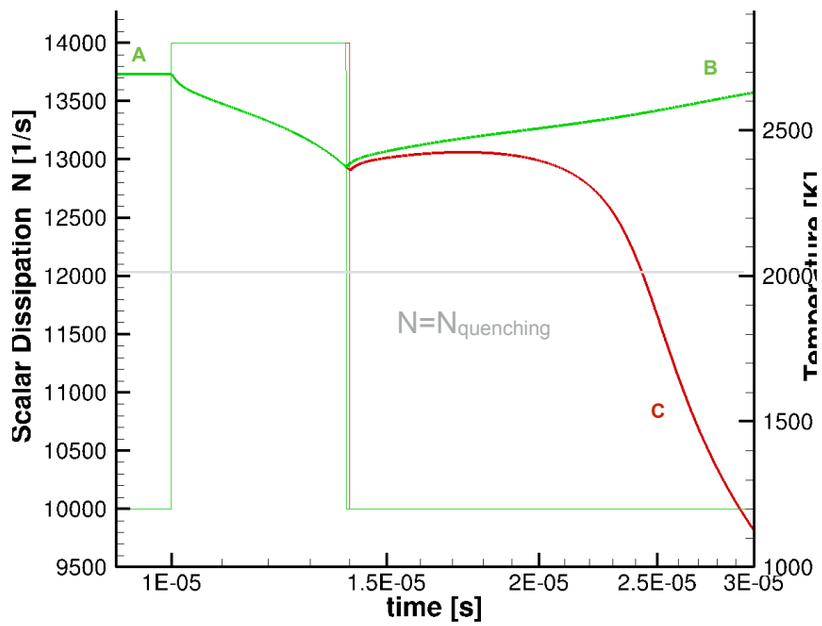
TSR Analysis of Quenching and Reignition

Quenching



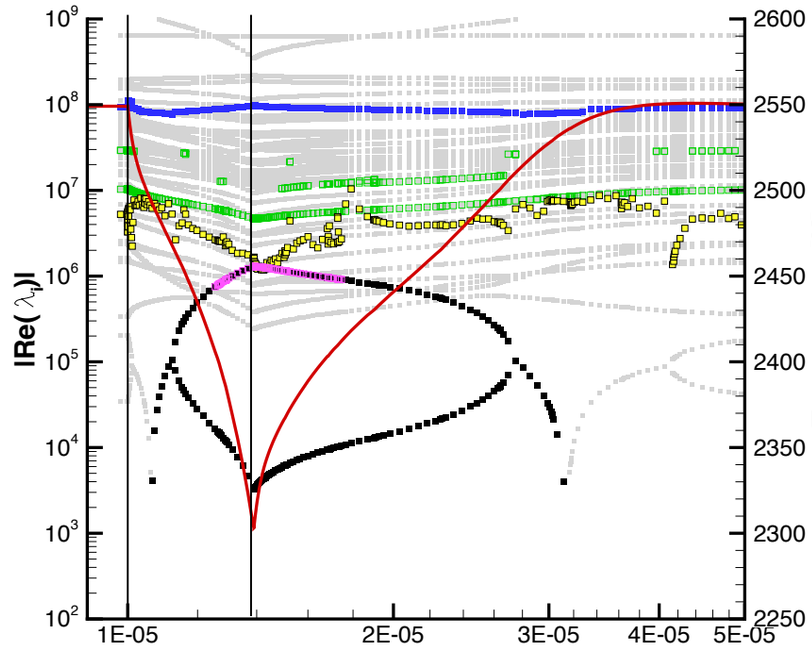


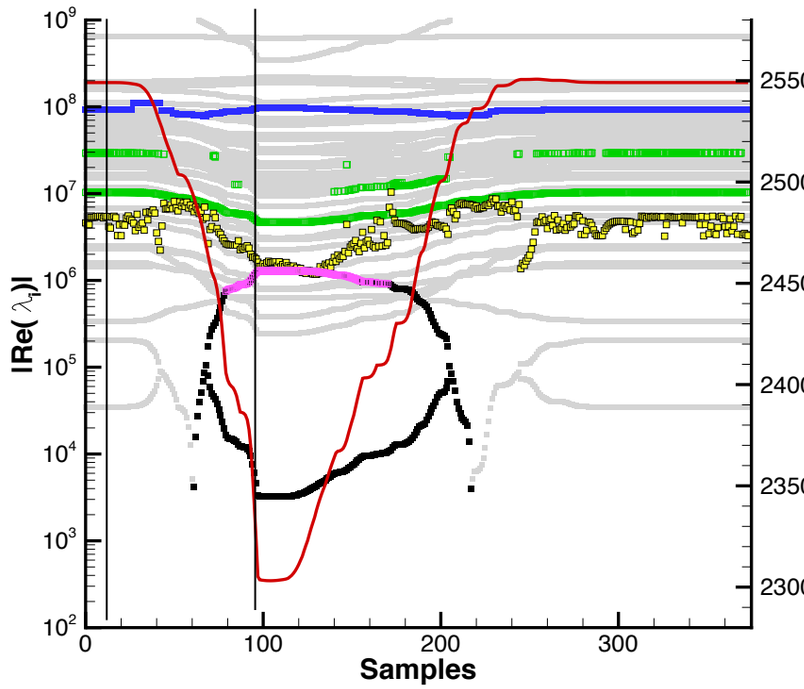




Z= 0.165

Reignition





TSR Analysis
Quenching and Reignition

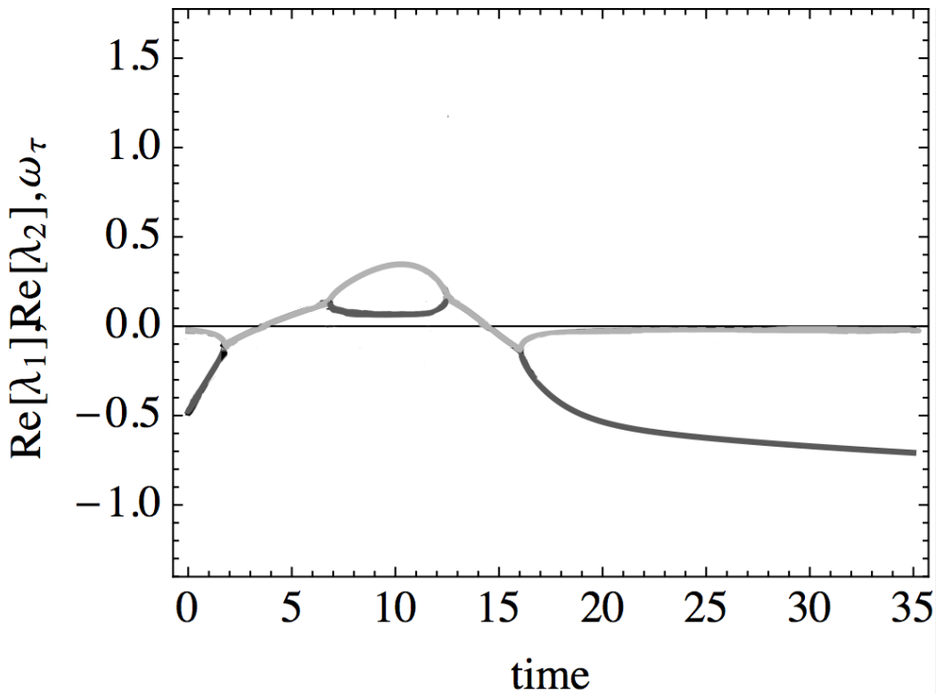
Z= 0.165

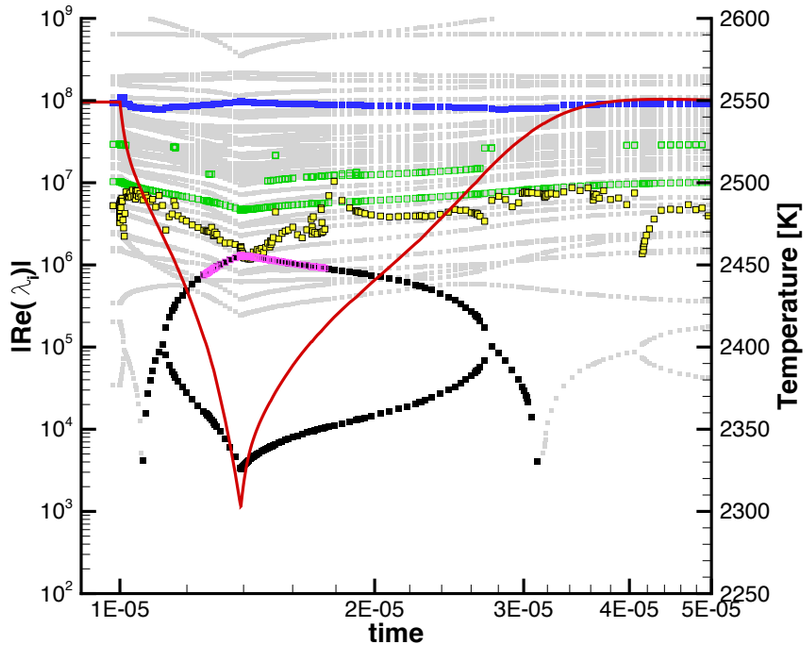
Semenov Model

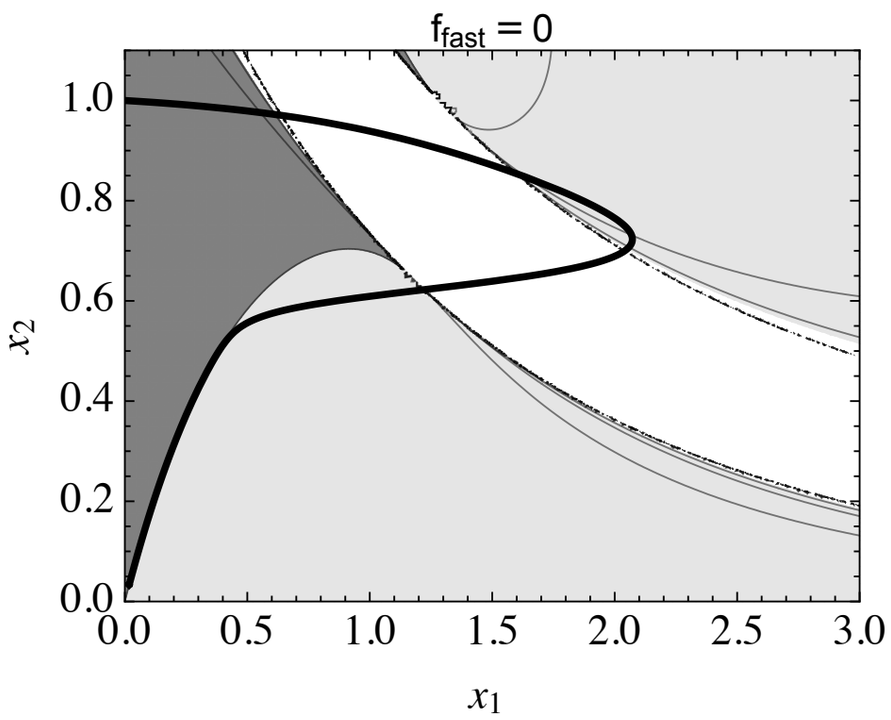
Non-isothermal (exponential) system

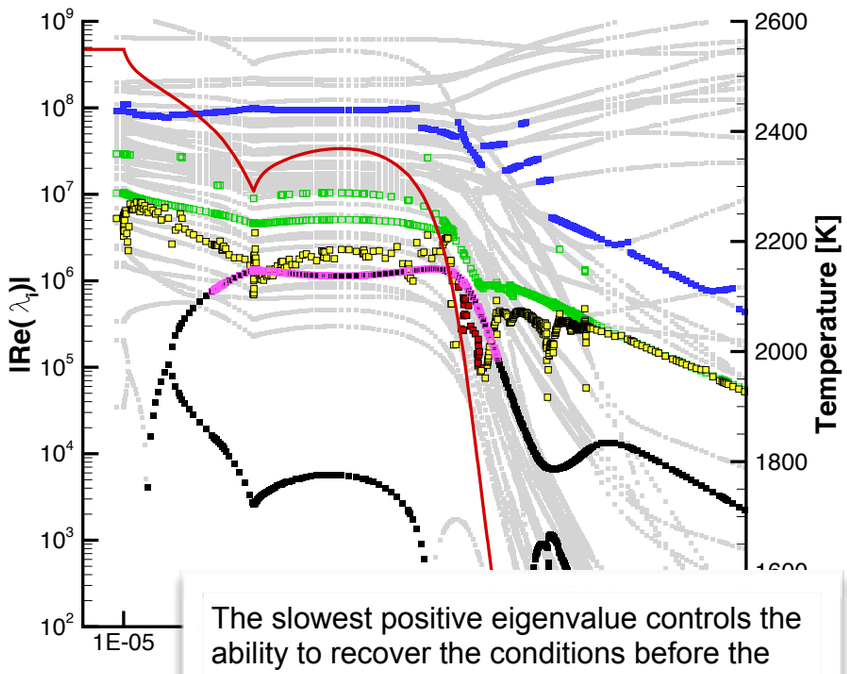
$$W(T,C_f) = K(T)C_f^1$$
$$V\rho C_v T' = +VQ_F W(T,C_f) - q_{wall}(T)$$
$$VC_{f'} = -VW(T,C_f) - \dot{m}_{fuel}(C_f)$$

- Ignition might initiate with a **pair of real negative** eigenvalues
- Transition from positive to negative sign occur with crossing a region of complex eigenvalues
- Crossing a region of complex eigenvalues can occur with a persistent sign (**positive)**









The slowest positive eigenvalue controls the ability to recover the conditions before the perturbation:

➡ Reignition occurs when the fast/slow positive eigenvalue merging occurs

➡ Otherwise, quenching occurs

Conclusions

TSR definition has been extended to PDEs

TSR analysis of non premixed systems (unsteady flamelet model) has been carried out for:
1)Ignition with different scalar dissipation rates
2)Quenching/Reignition bifurcative behavior

TSR allows to identify:
1)region in mixture fraction space of highest propensity to ignition
2)region in mixture fraction space of weak/none propensity to ignition
- Kinetics proceeds only because of diffusion

3)time scales associated with ignition
4)reactions most contributing to ignition

TSR analysis showed the role of diffusion in driving the kinetics off equilibrium at steady conditions

Quenching/Reignition bifurcative behavior exhibits similarities with the Semenov model of thermal explosion

Credits

The CSP Tool Box code (written in Mathematica) used in the analysis of the **model problems** can be obtained by sending a request to mauro.valorani@uniroma1.it

The library (CSP Tool Kit, **CSPTk**) used in the analysis of the **batch reactor model** can be obtained by sending a request to mauro.valorani@uniroma1.it

The library (TChem) used in the analysis of the **batch reactor model** can be obtained by sending a request to cosmin.safta@sandia.gov

The library (RFlamelet) used in the analysis of the **unsteady flamelet model** has been developed by P.P.Ciottoli, P.Lapenna, F.Creta

This work is under joint development among

NTUA: D.A.Goussis for the CSP library

CRF/SANDIA: H.N.Najm, C.Safta for the TChem library

KAUST: F.Bisetti, H.Im, M.Sarathi, et al., for the CSPTk library

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Thanks for your attention