On Computing Reactive Flows Using Slow Manifolds

by

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Outline

- Motivation.
- Comparison of ILDM and Slow Invariant Manifold (SIM) for spatially homogeneous premixed reactive systems.
- Theoretical development of the Elliptic Convection Diffusion Corrector (ECDC) method for the extension of the ILDM method to couple convection and diffusion with reaction.
- Comparison of the ECDC method with the Maas and Pope projection (MPP) method for simple model problem of Davis and Skodje, 1999.
- Comparison of the ECDC method with the MPP method for 1-D premixed laminar flame for ozone decomposition.
- Conclusions.

Motivation

- Severe stiffness in reactive fluid mechanical systems with detailed gas phase chemical kinetics renders fully resolved simulations of many systems to be impractical.
- ILDM method of Maas and Pope, 1992, offers a systematic robust method to equilibrate fast time scale phenomenon for spatially homogeneous premixed reactive systems with widely disparate reaction time scales.
- ILDM method can reduce computational time while retaining essential fidelity to full detailed kinetics.
- ILDM method effectively reduces large n species reactive systems to user-defined low order m-dimensional systems by replacing differential equations with algebraic constraints.
- The ILDM is only an approximation of the SIM, and contains a small intrinsic error for large stiffness.
- Using ILDM in systems with convection and diffusion can lead to large errors when convection and diffusion time scales are comparable to those of reactions.
- The ILDM method is extended to systems where reaction couples with convection and diffusion. An improvement over the MPP method is realized through a formulation of an elliptic system of equations via the ECDC method.

Chemical Kinetics Modeled as a Dynamical System

- ILDM developed for spatially homogeneous premixed reactor:

$$\frac{d\mathbf{y}}{dt} = \mathbf{f}(\mathbf{y}), \quad \mathbf{y}(0) = \mathbf{y}_\circ, \quad \mathbf{f}(\mathbf{0}) = 0, \quad \mathbf{y} \in \mathcal{R}^n,$$

$$\mathbf{y} = (T, P, Y_1, Y_2, ..., Y_n)^T.$$

- $\mathbf{f}(\mathbf{y})$ from Arrhenius kinetics.

Eigenvalues and Eigenvectors from Decomposition of Jacobian

$$\mathbf{J} = \mathbf{V}\Lambda\tilde{\mathbf{V}}, \qquad \mathbf{V}^{-1} = \tilde{\mathbf{V}},$$

$$\mathbf{V} = \left(\begin{array}{ccc:ccc} | & & | & | & & | \\ \mathbf{v}_1 & \cdots & \mathbf{v}_m & \mathbf{v}_{m+1} & \cdots & \mathbf{v}_n \\ | & & | & | & & | \end{array}\right) = \left(\begin{array}{c:c} \mathbf{V}_s & \mathbf{V}_f \end{array}\right),$$

$$\Lambda = \left(\begin{array}{ccc:ccc} \lambda_{(1)} & & 0 & & & \\ & \ddots & & & 0 & \\ 0 & & \lambda_{(m)} & & & \\ \hdashline & & & \lambda_{(m+1)} & & 0 \\ & 0 & & & \ddots & \\ & & & 0 & & \lambda_{(n)} \end{array}\right) = \left(\begin{array}{c:c} \Lambda_{(s)} & 0 \\ \hdashline 0 & \Lambda_{(f)} \end{array}\right),$$

$$\tilde{\mathbf{V}} = \left(\begin{array}{ccc} - & \tilde{\mathbf{v}}_1 & - \\ & \vdots & \\ - & \tilde{\mathbf{v}}_m & - \\ \hdashline - & \tilde{\mathbf{v}}_{m+1} & - \\ & \vdots & \\ - & \tilde{\mathbf{v}}_n & - \end{array}\right) = \left(\begin{array}{c} \tilde{\mathbf{V}}_s \\ \hdashline \tilde{\mathbf{V}}_f \end{array}\right).$$

The time scales associated with the dynamical system are the inverse of the eigenvalues:

$$\tau_i = \frac{1}{\lambda_{(i)}}.$$

Mathematical Model for ILDM

- With $\mathbf{z} = \mathbf{V}^{-1}\mathbf{y}$

$$\frac{1}{\lambda_{(i)}}\left(\frac{dz_i}{dt} + \tilde{\mathbf{v}}_i \sum_{j=1}^{n} \frac{d\mathbf{v}_j}{dt} z_j\right) = z_i + \frac{\tilde{\mathbf{v}}_i \mathbf{g}}{\lambda_{(i)}}, \text{ for } i = 1, \ldots, n,$$

  where $\mathbf{g} = \mathbf{f} - \mathbf{f}_\mathbf{y}\mathbf{y}$.

- $\frac{1}{\lambda_{(m+1)}}, \ldots, \frac{1}{\lambda_{(n)}}$ are the small parameters.
- Equilibrating the fast dynamics

$$\underbrace{z_i + \frac{\tilde{\mathbf{v}}_i \mathbf{g}}{\lambda_{(i)}} = 0}_{\text{ILDM}}, \text{ for } i = m+1, \ldots, n.$$

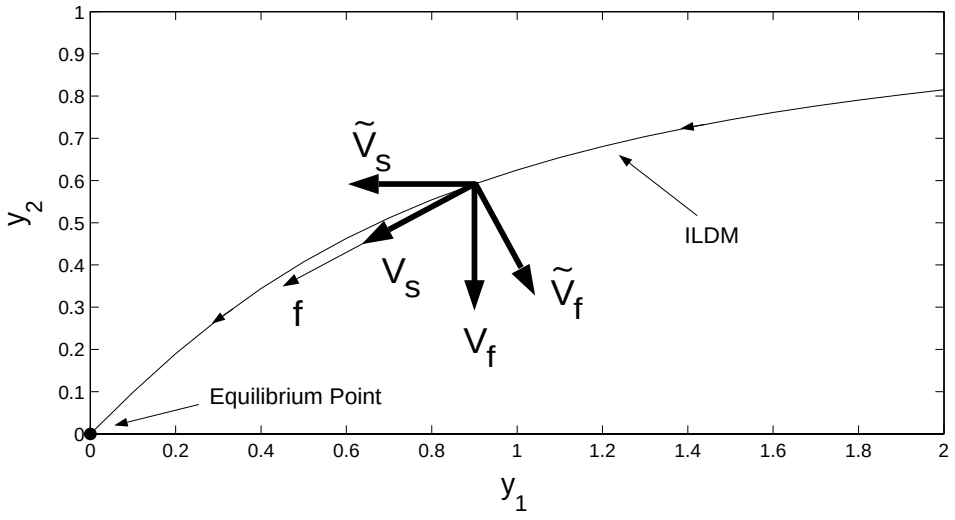
$$\Rightarrow \quad \underbrace{\tilde{\mathbf{V}}_f \mathbf{f} = 0}_{\text{ILDM}}.$$

- Slow dynamics approximated from differential algebraic equations on the ILDM

$$\begin{aligned} \tilde{\mathbf{V}}_s \frac{d\mathbf{y}}{dt} &= \tilde{\mathbf{V}}_s \mathbf{f}, \\ 0 &= \tilde{\mathbf{V}}_f \mathbf{f}. \end{aligned}$$

SIM vs. ILDM

- An invariant manifold is defined as a subspace $S \subset \mathcal{R}^n$ if for any solution $\mathbf{y}(t)$, $\mathbf{y}(0) \in S$, implies that for some $T > 0$, $\mathbf{y}(t) \in S$ for all $t \in [0, T]$.
- Slow Invariant Manifold (SIM) is also a trajectory in phase space and the vector $\mathbf{f}$ must be tangent to it.
- *ILDM is an approximation of the SIM and is not a phase space trajectory.*



- ILDM approximation gives rise to an intrinsic error which decreases as stiffness increases.

Demonstration that ILDM is not a trajectory in phase space.

- Normal vector to the ILDM is given by

$$\nabla(\underbrace{\tilde{\mathbf{V}}_f \mathbf{f}}_{\text{ILDM}}) = \tilde{\mathbf{V}}_f \mathbf{J} + (\nabla \tilde{\mathbf{V}}_f)\mathbf{f}$$

$$= \lambda_{(f)} \tilde{\mathbf{V}}_f + (\nabla \tilde{\mathbf{V}}_f)\mathbf{f},$$

  where in two dimensions $\lambda_{(f)} = \lambda_{(2)}$.

- If $\mathbf{f}$ is linear, $\nabla \tilde{\mathbf{V}}_f = \mathbf{0}$. Normal to the ILDM is parallel to $\tilde{\mathbf{V}}_f$ and orthogonal to $\mathbf{f}$ in two dimensions. ILDM is a trajectory.

- If $\mathbf{f}$ is non-linear, $\nabla \tilde{\mathbf{V}}_f \neq \mathbf{0}$. Normal to the ILDM is not parallel to $\tilde{\mathbf{V}}_f$ and not orthogonal to $\mathbf{f}$ in two dimensions. ILDM is not a trajectory.

- In the limit of large $\lambda_{(f)}$ the deviation of the ILDM from a phase space trajectory, and the SIM is small.

Comparison of the SIM with the ILDM

- Example from Davis and Skodje, 1999:

$$\frac{d\mathbf{y}}{dt} = \frac{d}{dt}\begin{pmatrix} y_1 \\ y_2 \end{pmatrix} = \begin{pmatrix} -y_1 \\ -\gamma y_2 + \frac{(\gamma-1)y_1+\gamma y_1^2}{(1+y_1)^2} \end{pmatrix} = \mathbf{f}(\mathbf{y}),$$

-

$$\mathbf{f_y} = \begin{pmatrix} -1 & 0 \\ \frac{\gamma-1+(\gamma+1)y_1}{(1+y_1)^3} & -\gamma \end{pmatrix}, \qquad \text{Jacobian}$$

$$\tilde{\mathbf{v}}_1 = \tilde{\mathbf{V}}_s = \begin{pmatrix} 1 & 0 \end{pmatrix}, \qquad \lambda_{(1)} = \lambda_{(s)} = -1, \qquad \text{slow}$$

$$\tilde{\mathbf{v}}_2 = \tilde{\mathbf{V}}_f = \begin{pmatrix} -\frac{\gamma-1+(\gamma+1)y_1}{(\gamma-1)(1+y_1)^3} & 1 \end{pmatrix}, \qquad \lambda_{(2)} = \lambda_{(f)} = -\gamma, \qquad \text{fast}$$

- The ILDM for this system is given by

$$\tilde{\mathbf{V}}_f\mathbf{f} = 0 \quad \Rightarrow \quad y_2 = \frac{y_1}{1+y_1} + \frac{2y_1^2}{\gamma(\gamma-1)(1+y_1)^3}.$$

Comparison of the SIM with the ILDM

- SIM assumed to be a polynomial

$$y_2 = \sum_{k=0}^{\infty} c_k y_1^k.$$

- Substituting the polynomial in the following equation

$$-\gamma y_2 + \frac{(\gamma - 1)y_1 + \gamma y_1^2}{(1 + y_1)^2} = -y_1 \frac{dy_2}{dy_1}.$$

- SIM is given by

$$y_2 = y_1(1 - y_1 + y_1^2 - y_1^3 + y_1^4 + \dots) = \frac{y_1}{1 + y_1}.$$

- ILDM

$$\tilde{\mathbf{V}}_f \mathbf{f} = 0 \quad \Rightarrow \quad y_2 = \frac{y_1}{1 + y_1} + \frac{2y_1^2}{\gamma(\gamma - 1)(1 + y_1)^3}.$$

- For large γ or stiffness, the ILDM approaches the SIM.
- For even a slightly more complicated systems we have to resort to a numerical computation of the SIM using the Roussel and Fraser method, 1992.

Comparison of the SIM with the ILDM

- Projection of the system on the slow and fast basis.

$$\text{Slow:} \qquad \tilde{\mathbf{V}}_s \frac{d\mathbf{y}}{dt} = \tilde{\mathbf{V}}_s \mathbf{f} \;\; \Rightarrow \;\; \frac{dy_1}{dt} = -y_1$$

$$\begin{aligned} \text{Fast:} \qquad \tilde{\mathbf{V}}_f \frac{d\mathbf{y}}{dt} &= \tilde{\mathbf{V}}_f \mathbf{f} \;\; \Rightarrow \\ \frac{1}{\gamma}\left(-\frac{\gamma - 1 + (\gamma+1)y_1}{(\gamma-1)(1+y_1)^3}\frac{dy_1}{dt} + \frac{dy_2}{dt}\right) &= -y_2 + \frac{y_1}{1+y_1} + \frac{2y_1^2}{\gamma(\gamma-1)(1+y_1)^3} \end{aligned}$$

Order of terms in the fast equation:

$$\mathcal{O}\left(\frac{1}{\gamma}\right) + \mathcal{O}\left(\frac{1}{\gamma^2}\right) + \ldots \;=\; \mathcal{O}(1) + \mathcal{O}\left(\frac{1}{\gamma}\right) + \ldots$$

- The ILDM approximation neglects all terms on the LHS while retaining all terms on RHS of the fast equation.
- Systematic matching of terms of all orders in a singular perturbation scheme correctly leads to the SIM

$$y_2 = \frac{y_1}{1+y_1}.$$

- The ILDM approximates the SIM well for large γ or stiffness and is a more practical method for complicated systems such as chemical kinetics.

Reaction Convection Diffusion Equations

- In one spatial dimension

$$\frac{\partial \mathbf{y}}{\partial t} = \underbrace{\mathbf{f}(\mathbf{y})}_{\text{reaction}} - \underbrace{\frac{\partial}{\partial x}\mathbf{h}(\mathbf{y})}_{\text{convection-diffusion}} .$$

- Substituting $\mathbf{z} = \mathbf{V}^{-1}\mathbf{y}$

$$\underbrace{\frac{1}{\lambda_{(i)}}\left(\frac{dz_i}{dt} + \tilde{\mathbf{v}}_i \sum_{j=1}^{n} \frac{d\mathbf{v}_j}{dt} z_j\right)}_{=0 \text{ for i=m+1,...,n}} = z_i + \frac{\tilde{\mathbf{v}}_i \mathbf{g}}{\lambda_{(i)}} - \frac{1}{\lambda_{(i)}}\left(\tilde{\mathbf{v}}_i \frac{\partial \mathbf{h}}{\partial x}\right) \text{ for } i = 1, \ldots, n.$$

- Equilibrating the fast dynamics, we get the elliptic equations

$$z_i + \frac{\tilde{\mathbf{v}}_i \mathbf{g}}{\lambda_{(i)}} - \frac{1}{\lambda_{(i)}}\left(\tilde{\mathbf{v}}_i \frac{\partial \mathbf{h}}{\partial x}\right) = 0, \text{ for } i = m+1, \ldots, n.$$

- For diffusion time scales which are of the order of the chemical time scales

$$z_i + \frac{\tilde{\mathbf{v}}_i \mathbf{g}}{\lambda_{(i)}} - \frac{1}{\lambda_{(i)}}\left(\tilde{\mathbf{v}}_i \frac{\partial \mathbf{h}}{\partial x}\right) = 0, \text{ for } i = m+1, \ldots, p.$$

- For fast chemical time scales we obtain the ILDM

$$z_i + \frac{\tilde{\mathbf{v}}_i \mathbf{g}}{\lambda_{(i)}} = 0, \text{ for } i = p+1, \ldots, n.$$

Elliptic Convection Diffusion Corrector (ECDC)

- Slow dynamics can be approximated by the ECDC method

$$\begin{aligned} \tilde{\mathbf{V}}_s \frac{\partial \mathbf{y}}{\partial t} &= \tilde{\mathbf{V}}_s \mathbf{f} - \tilde{\mathbf{V}}_s \frac{\partial \mathbf{h}}{\partial x}, \\ 0 &= \tilde{\mathbf{V}}_{fs} \mathbf{f} - \tilde{\mathbf{V}}_{fs} \frac{\partial \mathbf{h}}{\partial x}, \\ 0 &= \tilde{\mathbf{V}}_{ff} \mathbf{f}. \end{aligned}$$

where $\tilde{\mathbf{V}}_{fs} = \begin{pmatrix} - & \tilde{\mathbf{v}}_{m+1} & - \\ & \vdots & \\ - & \tilde{\mathbf{v}}_p & - \end{pmatrix}$ and $\tilde{\mathbf{V}}_{ff} = \begin{pmatrix} - & \tilde{\mathbf{v}}_{p+1} & - \\ & \vdots & \\ - & \tilde{\mathbf{v}}_n & - \end{pmatrix}$

- The diffusion term can be neglected only if the following term is small

$$\frac{1}{\lambda_{(i)}} \left(\tilde{\mathbf{v}}_i \frac{\partial \mathbf{h}}{\partial x} \right).$$

- Difficult to determine appropriate p, which is spatially dependent!
- It is difficult to *a priori* determine the the diffusion length scales and hence the diffusion time scales.
- A cautious approach, which we adopt, is to require $p = n$ and solve elliptic PDEs instead of the algebraic ILDM equations.

Davis Skodje Example Extended to Reaction Diffusion

$$\frac{\partial \mathbf{y}}{\partial t} = \frac{\partial}{\partial t}\begin{pmatrix} y_1 \\ y_2 \end{pmatrix} = \begin{pmatrix} -y_1 \\ -\gamma y_2 + \frac{(\gamma-1)y_1+\gamma y_1^2}{(1+y_1)^2} \end{pmatrix} - \frac{\partial}{\partial x}\begin{pmatrix} -\mathcal{D}\frac{\partial y_1}{\partial x} \\ -\mathcal{D}\frac{\partial y_2}{\partial x} \end{pmatrix} = \mathbf{f}(\mathbf{y}) - \frac{\partial}{\partial x}\mathbf{h}(\mathbf{y})$$

- Boundary conditions are chosen on the ILDM

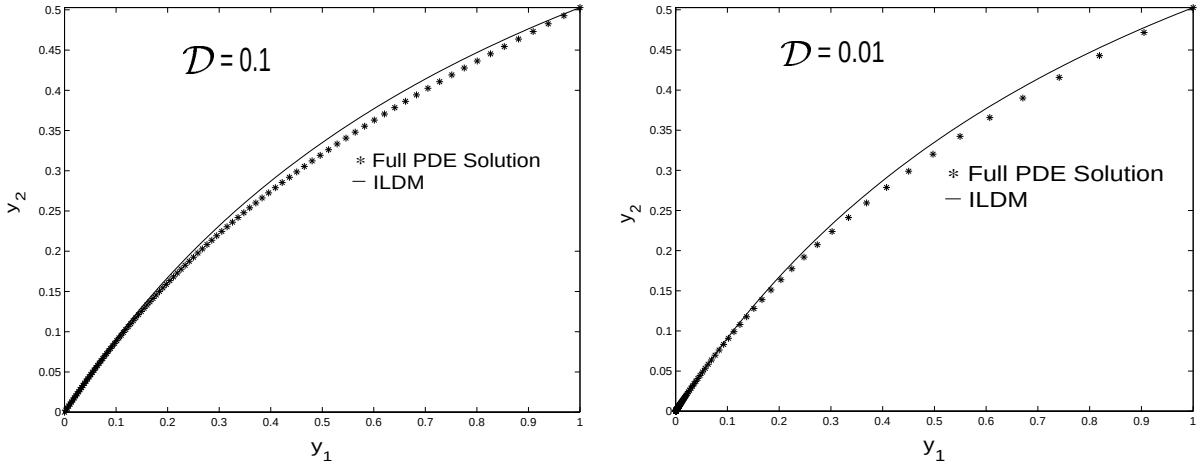
$$y_1(t,0) = 0, \;\; y_1(t,1) = 1,$$

$$y_2(t,0) = 0, \;\; y_2(t,1) = \tfrac{1}{2} + \tfrac{1}{4\gamma(\gamma-1)}.$$

- Initial conditions

$$y_1(0,x) = x, \quad y_2(0,x) = \left(\frac{1}{2} + \frac{1}{4\gamma(\gamma-1)}\right) x$$

Reaction Diffusion Example Results: Low Stiffness

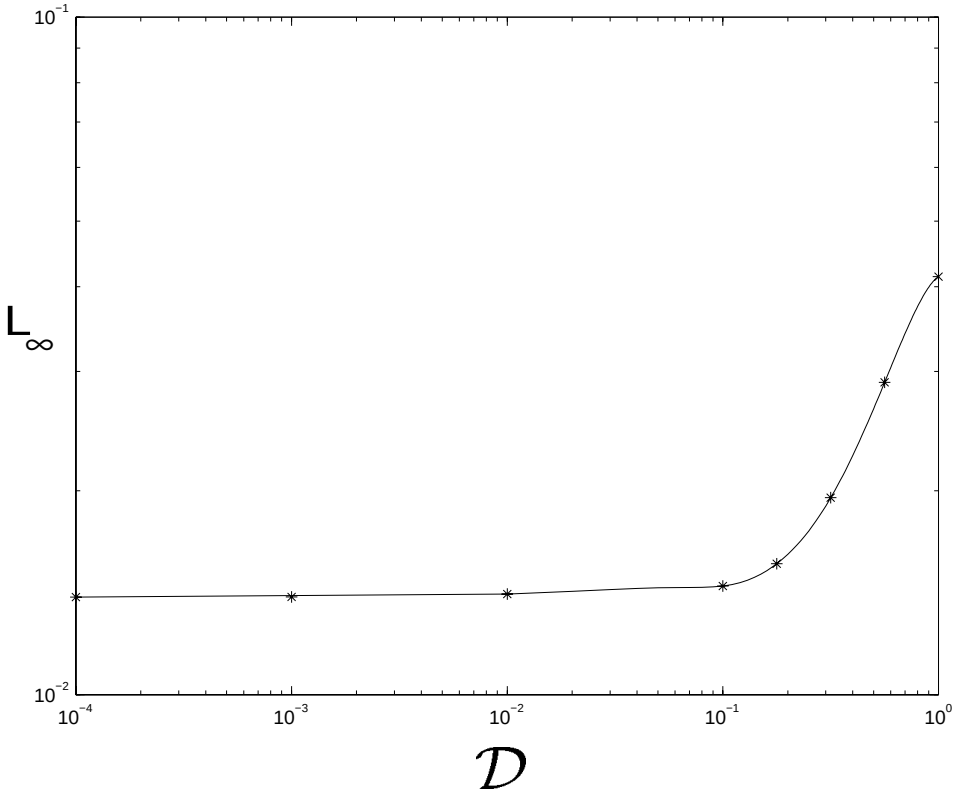


(a) (b)

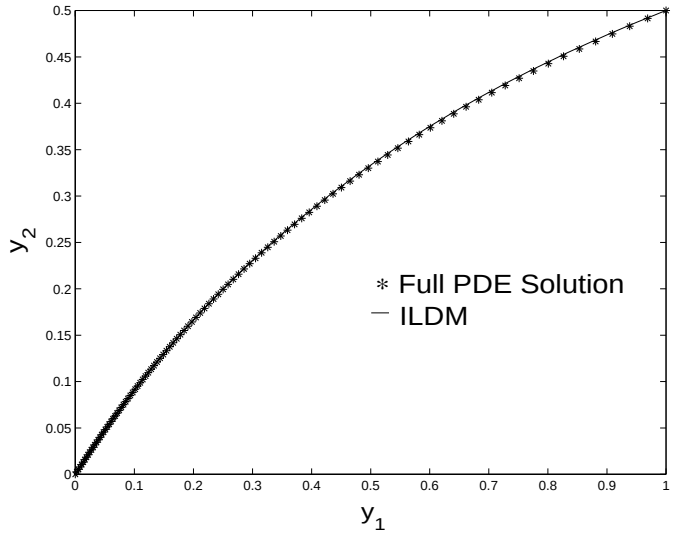
- Solution at $t = 5$, for $\gamma = 10$ with varying $\mathcal{D}$.
- PDE solution fully resolved; no ILDM or convection-diffusion correction.
- Forcing the solution onto the ILDM will induce large errors.

Reaction Diffusion Example Results: Low Stiffness

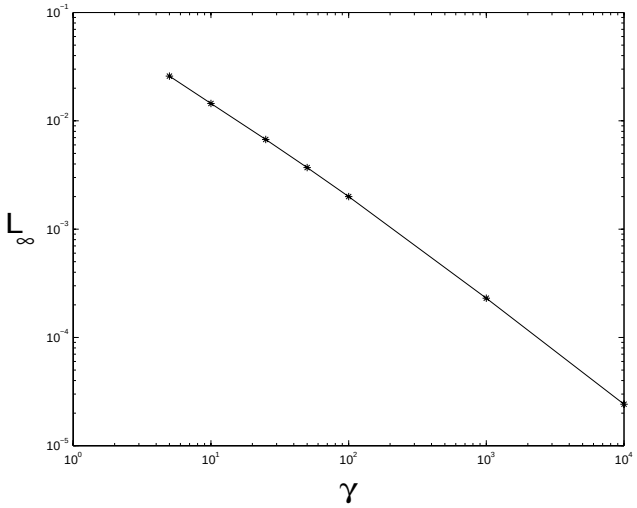
- Modification of $\mathcal{D}$ alone for fixed γ does not significantly change the error.



Reaction Diffusion Example Results: High Stiffness



- Solution at $t = 5$, for $\gamma = 100$ and $\mathcal{D} = 0.1$.
- Increasing γ moves solution closer to the ILDM.



Comparison of the ECDC method and the MPP method

- The slow dynamics for the simple system obtained using the ECDC method by taking $n = 2$, $m = 1$, $p = n$

$$\begin{aligned} \frac{\partial y_1}{\partial t} &= -y_1 + \mathcal{D}\frac{\partial^2 y_1}{\partial x^2} \\ y_2 - \frac{1}{\gamma}\frac{\partial^2 y_2}{\partial x^2} &= \frac{y_1}{1+y_1} + \frac{2y_1^2}{\gamma(\gamma-1)(1+y_1)^3} - \left(\frac{\gamma - 1 + (\gamma+1)y_1}{\gamma(\gamma-1)(1+y_1)^3}\right)\frac{\partial^2 y_1}{\partial x^2} \end{aligned}$$

- The Maas and Pope projection (MPP) method is given by projecting the convection diffusion term along the local slow subspace on the reaction ILDM, hence, ensuring that the slow dynamics occurs on the ILDM

$$\frac{\partial \mathbf{y}}{\partial t} = \mathbf{f}(\mathbf{y}) - \mathbf{V}_s\tilde{\mathbf{V}}_s\frac{\partial}{\partial x}\left(\mathbf{h}(\mathbf{y})\right).$$

- For the simple system the corresponding equations are given by

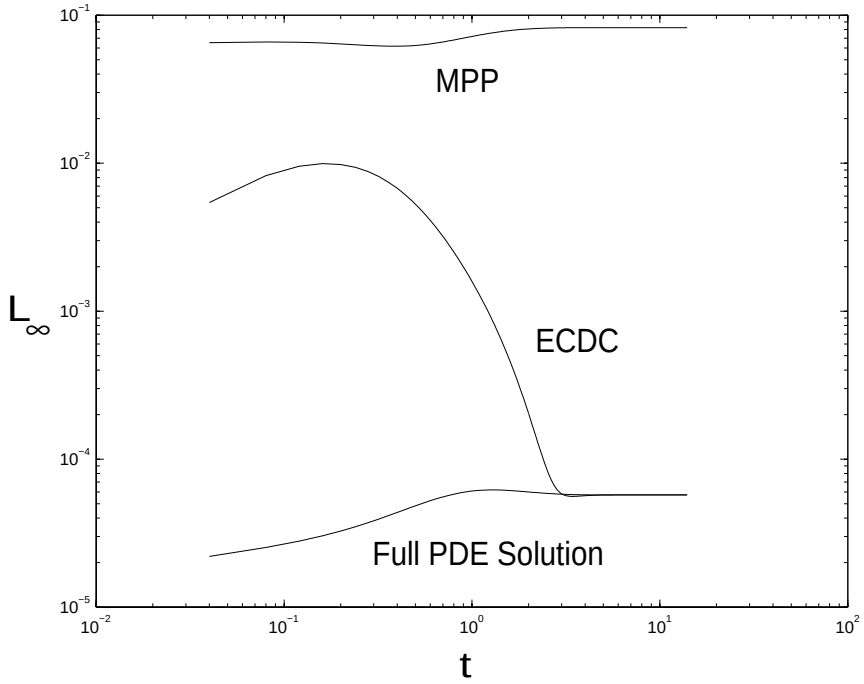
$$\begin{aligned} \frac{\partial y_1}{\partial t} &= -y_1 + \mathcal{D}\frac{\partial^2 y_1}{\partial x^2}, \\ \frac{\partial y_2}{\partial t} &= -\gamma y_2 + \frac{(\gamma-1)y_1 + \gamma y_1^2}{(1+y_1)^2} - \left(\frac{\gamma - 1 + (\gamma+1)y_1}{\gamma(\gamma-1)(1+y_1)^3}\right)\mathcal{D}\frac{\partial^2 y_1}{\partial x^2}. \end{aligned}$$

- The slow dynamics for the simple system obtained using the MPP method by taking $n = 2$ and $m = p = 1$.

$$\begin{aligned} \frac{\partial y_1}{\partial t} &= -y_1 + \mathcal{D}\frac{\partial^2 y_1}{\partial x^2}, \\ y_2 &= \frac{y_1}{1+y_1} + \frac{2y_1^2}{\gamma(\gamma-1)(1+y_1)^3}. \end{aligned}$$

Comparison of the ECDC method and the MPP method

- Solutions obtained by the MPP method, the ECDC method, and full equations, all calculated on a fixed grid with 100 points.
- All results compared to solution of full equations at high spatial resolution of 10000 grid points.
- $\gamma = 10$ and $\mathcal{D} = 0.1$.
- Forcing the solution onto the ILDM leads to large errors in the MPP method.
- Overall the error in the ECDC method is lower than the MPP method, and is similar to that incurred by the full equations near steady state.



1D Premixed Laminar Flame for Ozone Decomposition

- Governing equations of one-dimensional, isobaric, premixed laminar flame for ozone decomposition in Lagrangian coordinates for low Mach number flows, Margolis, 1978.

$$\begin{aligned}
\frac{\partial T}{\partial t} &= -\frac{1}{\rho c_p}\sum_{k=1}^{3}\dot{\omega}_k M_k h_k + \frac{1}{c_p}\frac{\partial}{\partial \psi}\left(\rho\lambda\frac{\partial T}{\partial \psi}\right), \\
\frac{\partial Y_k}{\partial t} &= \frac{1}{\rho}\dot{\omega}_k M_k + \frac{\partial}{\partial \psi}\left(\rho\mathcal{D}_k^2\frac{\partial Y_k}{\partial \psi}\right), \text{ for } k = 1, 2, 3,
\end{aligned}$$

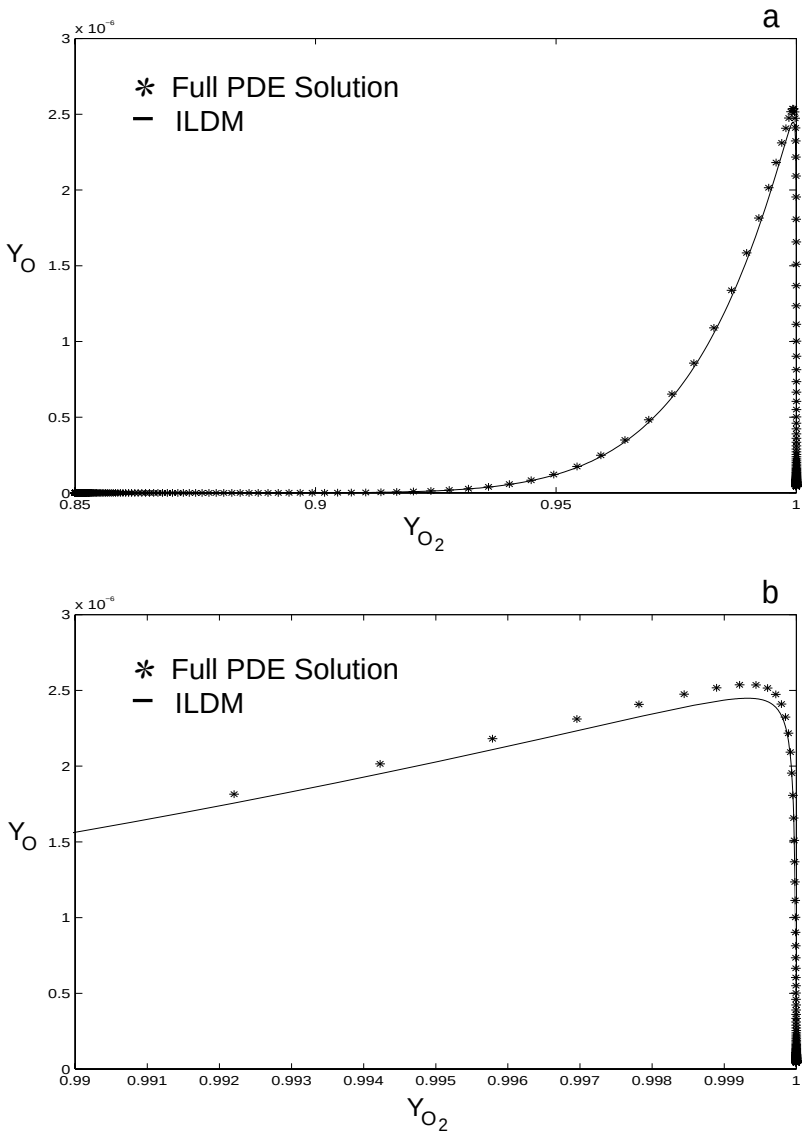
- Equation of state

$$p_0 = \rho\Re T\sum_{k=1}^{3}\frac{Y_k}{M_k},$$

- $Le = 1$.
- $c_p = 1.056 \times 10^6$ erg/(g-K).
- $\rho\lambda = 4.579 \times 10^{-2}$ g^2/(cm^2-s^3-K).
- $\mathcal{D}_1 = \mathcal{D}_2 = \mathcal{D}_3 = \mathcal{D}$.
- $\rho^2\mathcal{D} = 4.336 \times 10^{-7}$ g^2/(cm^4-s).
- $Y_1 = Y_O$, $Y_2 = Y_{O_2}$, $Y_3 = Y_{O_3}$.
- $p_0 = 8.32 \times 10^5$ dynes/cm^2.

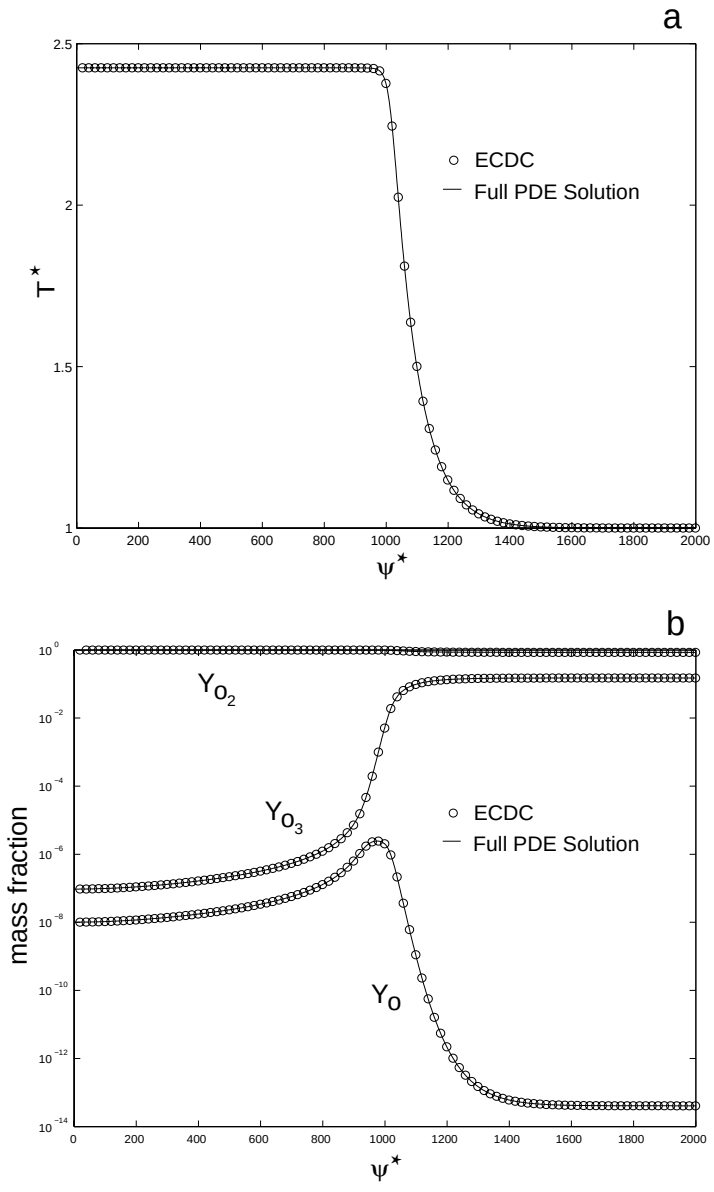
Comparison of the ILDM with the PDE solution for ozone decomposition flame

For unreacted mixture of $Y_{O_2} = 0.85$, $Y_{O_3} = 0.15$ at $T = 300K$



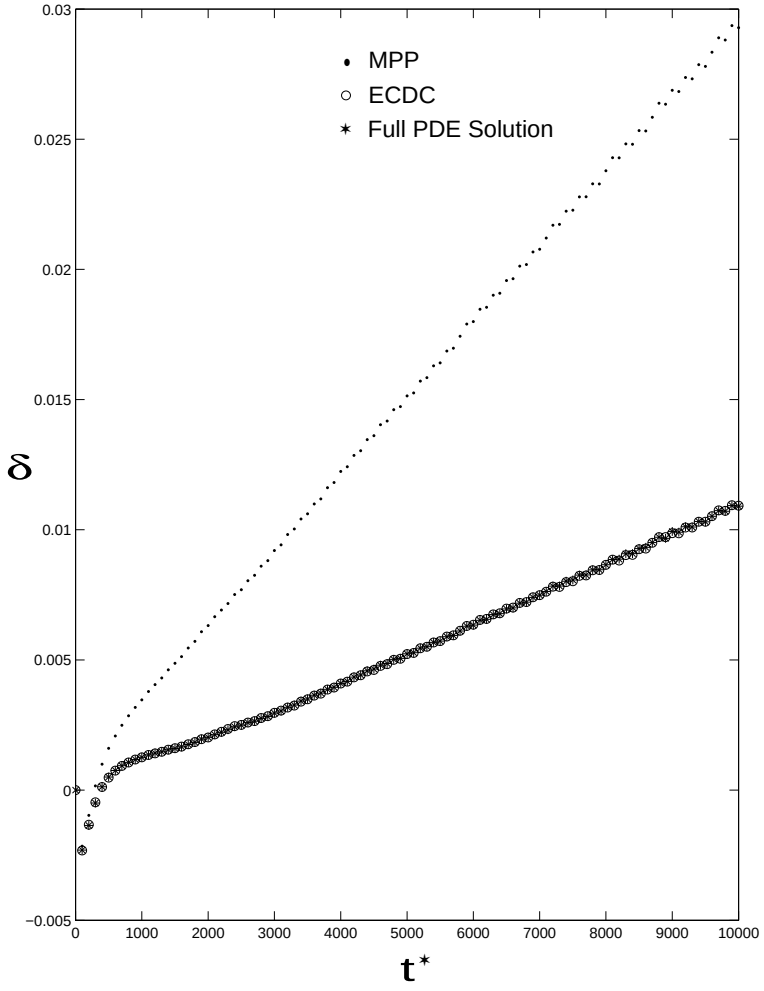
Profiles for 1D Premixed Laminar Flame for Ozone Decomposition

For unreacted mixture of $Y_{O_2} = 0.85$, $Y_{O_3} = 0.15$ at $T = 300K$



Comparison of phase errors incurred by the MPP method, the ECDC method and full integration.

- For unreacted mixture of $Y_{O_2} = 0.85$, $Y_{O_3} = 0.15$ at $T = 300K$.
- The phase error δ is measured as the Lagrangian distance between the location within the flame front where the mass fraction of O_3 is 0.075, for the solution obtained by the three methods, for 1000 grid points, and the full integration solution at 10000 grid points.



Conclusions

- ILDM approaches SIM in the limit of large stiffness for spatially homogeneous systems.
- Difficult to use SIM in practical combustion calculations while ILDM works well.
- No robust analysis currently exists to determine convection and diffusion time scales *a priori*.
- For systems in which convection and diffusion have time scales comparable to those of reaction, MPP method can lead to a large transient and steady state error.
- In the ECDC method and elliptic PDE is developed and solved to better couple reaction, convection and diffusion while systematically equilibrating fast time scales.
- At this point the fast and slow subspace decomposition is dependent only on reaction and should itself be modified to include fast and slow convection-diffusion time scales.
- The error incurred in approximating the slow dynamics by the ECDC method is smaller than that incurred by the MPP method.