

## HIERARCHICAL CONCEPTS FOR MODEL REDUCTION FOR REACTING FLOWS BASED ON LOW-DIMENSIONAL MANIFOLDS

U. Maas\*, A. Y. Klimenko\*\*

\*Karlsruhe Institute of Technology, 76131 Karlsruhe, Germany E-mail: Ulrich.Maas@kit.edu

\*\*The University of Queensland, QLD 4072, Australia, E-mail: a.klimenko@uq.edu.au

### ABSTRACT

The description of chemically reacting systems leads very often to reaction mechanisms with far above hundred chemical species (and, therefore, to more than a hundred partial differential equations), which possibly react within more than a thousand of elementary reactions. These kinetic processes cover time scales from nanoseconds to seconds. An analogous scaling problem arises for the length scales. Due to these scaling problems the detailed simulation of three-dimensional turbulent flows in practical systems is beyond the capacity of even today's super-computers. Using simplified sub-models is a way out of this problem. The question arising in mathematical modeling of reactive flows is then: How detailed, or down to which scale has each process to be resolved (chemical reaction, chemistry-turbulence-interaction, molecular transport processes) in order to allow a reliable description of the entire process. Both the chemical source term and the transport term have one important property, namely that they cause the existence of low-dimensional attractors in composition space. These manifolds can be parameterized by a small number of variables. In this work we discussed several model reduction aspects based on the concept of low-dimensional manifolds, namely the efficient identification of the low-dimensional manifolds, the efficient implementation to simplify the chemical kinetics, the hierarchical nature of the low-dimensional manifolds the use of the hierarchical nature to devise hierarchical modeling concepts for turbulent reacting flows.

### INTRODUCTION

Reacting flows are governed by a complex interplay of chemical reaction, flow and molecular transport. They can be described mathematically based on conservation equations for mass, momentum, energy and species masses. These conservation equations form a large system of stiff partial differential equations and, therefore, their solution is a great challenge [1; 2]. In the past many attempts have been made to simplify the description of chemically reacting flows while still capturing the essential features of the dynamics of the system. The developed methods focus both on a simplified description of the chemical kinetics and on a simplified description of the turbulent flow and the chemistry-turbulence interaction (see e.g. [3; 4; 5]). Different methods for dimension reduction have been proposed for the chemical kinetics. Many of them are based on a detailed analysis of the chemical source term (see e.g. [6; 7; 8; 9; 10; 11; 12; 13; 14]). Methods that account for the interaction of chemical reactions and physical processes are e.g. [15; 16; 17; 18; 19; 20]. An overview of several methods can be found in [21]. Furthermore many methods for the description of the chemistry-turbulence interaction make use of the existence of low-dimensional manifolds in composition space. Examples are the flamelet concept [22], conditional moment closure [23] and multiple mapping conditioning [24].

The dynamics of reacting flows is governed by the system of conservation equations for mass, momentum, energy, and species masses [25]. For the following analysis it is useful to separate the equations for the thermokinetic state variables from the equations for the flow field and to assume (for sake of simplicity) a low Mach number flow with constant thermodynamic pressure (a generalization to general flows is straight forward

and shall not be considered here). The governing equation system for the scalar field can be written as:

$$\frac{\partial \psi}{\partial t} = F - \vec{v} \text{grad} \psi - \frac{1}{\rho} \text{div} (D \text{grad} \psi), \quad (1)$$

where  $t$  denotes the time,  $\psi$  the  $(n = n_s + 2)$ -dimensional thermokinetic state vector (which is, e.g. given as  $\psi = (h, p, y_1, \dots, y_{n_s})^T$ , where  $n_s$  is the number of species,  $h$  is the specific enthalpy,  $p$  the pressure, and  $y_i$  the mass fraction of species  $i$ ), and  $\rho$  is the density,  $\vec{v}$  the velocity,  $D$  the  $(n$  by  $n)$ -dimensional matrix of transport coefficients (see, e.g., [26] for details), and  $F$  the  $(n)$ -dimensional vector of source terms. The source terms  $F$  as well as the transport matrix  $D$  are complicated nonlinear functions of the thermokinetic state vector  $\psi$  [27; 26; 28]. Because (1) does not invoke any modeling procedure, it is valid for laminar as well as for turbulent flows.

Both the chemical source term and the transport term have one important property, namely that they cause the existence of low-dimensional attractors in composition space. They result from fast chemical processes leading to species in steady states or reactions in partial equilibria. These manifolds can be parameterized by a small number  $m$  of variables, represented by the vector  $\theta$  of reduced coordinates ( $\theta = (\theta_1, \theta_2, \dots, \theta_m)^T$ ).

$$\mathcal{M} = \{\psi = \psi(\theta), \theta \in R^m, \psi \in R^n\}, \quad m \ll n \quad (2)$$

At each point  $\psi$  of a manifold  $\mathcal{M}^m$  perturbations off the manifold are relaxed according to a relaxation rate  $|\omega|$  that is larger than a given  $|\omega^m(\psi)|$ . Note that this relaxation rate  $\omega$  is for example for intrinsic low-dimensional manifolds given by the

value of the smallest (in magnitude) decoupled eigenvalue [8; 9]. Many model reduction techniques make use of the existence of such low-dimensional attractors (see e.g. [21]). Furthermore, methods like conditional moment closure [23] and multiple mapping conditioning [24] rely on the existence of such attractors in composition space. However, there exists also a hierarchy of the attractors, i. e. in the state space relaxation to equilibrium can be represented as a cascade through a nested hierarchy of smooth hypersurfaces [29]. While it is now well accepted that such a hierarchy exists, it is still necessary and useful to show, whether mathematical constructions of low-dimensional manifolds (i.e. model reduction concepts) can represent this behavior. It can be shown for several manifold based reduction concepts (see below) that attractors  $\mathcal{M}^m$  of dimension  $m < m + p$  are under certain conditions embedded in attractors  $\mathcal{M}^{m+p}$  of dimension  $m + p$ , i.e.  $\mathcal{M}^m \subset \mathcal{M}^{m+p}$ . This hierarchical nature can be used in many applications, e.g.

for an efficient construction of low-dimensional manifolds  
for a hierarchical improvement of the model accuracy in  
reacting flow calculations  
for devising hierarchical methods for turbulence/chemistry  
interaction closure.

These issues will be addressed below.

In the following we shall focus only on some examples of manifold concepts, namely those based on quasi steady state assumptions (QSSA) and partial equilibrium assumptions (PEA) [30], intrinsic low-dimensional manifolds (ILDM) [8] and global quasi-linearization (GQL) [11], which are based on an analysis of the chemical source term only, and the concept of reaction diffusion manifolds (REDIM) [19], which is based on a coupled analysis of reaction and molecular transport.

## THE HIERARCHICAL NATURE OF QSSA, PEA, ILDM AND GQL

For a pure homogeneous reaction system (1) reduces to

$$\frac{\partial \psi}{\partial t} = F(\psi). \quad (3)$$

In principle the assumption of species  $i$  being in steady state simply implies  $F_{i+2} \approx 0$  (note that the first two entries in  $\psi$  are the enthalpy and the pressure, and the first species evolution equation has index 3). For  $n_f = n - m$  species being in steady state the steady state conditions can be written as

$$\tilde{C}^m F(\psi) = 0, \quad (4)$$

with

$$\tilde{C} = \begin{pmatrix} 0 & 0 & \delta_{1k_1} & \delta_{2k_1} & \cdots & \delta_{n_s k_1} \\ 0 & 0 & \delta_{1k_2} & \delta_{2k_2} & \cdots & \delta_{n_s k_2} \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \delta_{1k_{n_f}} & \delta_{2k_{n_f}} & \cdots & \delta_{n_s k_{n_f}} \end{pmatrix}, \quad (5)$$

where  $\delta$  denotes the Kronecker- $\delta$ , and  $k_j$  is the index of the species in the  $j^{\text{th}}$  steady state assumption. This yields a definition of the  $(m = n - n_f)$ -dimensional QSSA manifold:

$$\mathcal{M}^m = \{\psi | \tilde{C}^m F(\psi) = 0\} \quad (6)$$

It is easy to see that if we have a set of  $(n - (m + p))$  species assumed to be in steady state, which is a subset of  $(n - m)$  species assumed to be in steady state

$$\{k_1, k_2, \dots, k_{n-(m+p)}\} \subset \{\hat{k}_1, \hat{k}_2, \dots, \hat{k}_{n-m}\}, \quad (7)$$

(where  $\hat{\cdot}$  is only used to indicate a possible different ordering of the species in the two subsets), then the matrix  $C^{m+p}$  is in the span of the matrix  $C^m$ , and it follows directly from (6) that

$$\mathcal{M}^m \subset \mathcal{M}^{m+p} \quad (8)$$

In partial equilibrium approximations (in the simplest formulation, see [21]) each partial equilibrium assumption defines a nonlinear equation via equating the rates of the forward and backward reaction. It is easy to show that if we define two low-dimensional manifolds via  $(n - (m + p))$  and  $(n - m)$  partial equilibrium conditions, respectively, where the  $(n - (m + p))$  vectors of the stoichiometric coefficients of the partial equilibrium reactions for  $\mathcal{M}^{m+p}$  are in the span of the  $(n - m)$  vectors of stoichiometric coefficients of the partial equilibrium reactions for  $\mathcal{M}^m$ , then again the  $m$ -dimensional manifold is a subset of the  $m + p$ -dimensional manifold.

This means that QSSA and PEA manifolds can be constructed in such a way, that a hierarchy of the manifolds exists, although it shall be noted that very often different sets of assumptions for different dimensions are used, i.e.

$$\{k_1, k_2, \dots, k_{n_f}\} \not\subset \{\hat{k}_1, \hat{k}_2, \dots, \hat{k}_{n_f}\}, \quad (9)$$

In this case a hierarchical nature of the manifolds is not guaranteed.

Let us now discuss the hierarchy in the context of ILDM and GQL. The mathematical model of these methods is described in detail in previous works (see e.g. [8; 31; 9; 32; 33; 34; 11; 35]). Here, only a short repetition to clarify the presentation shall be given. ILDM and QGL differ in principle how the fast and slow processes are identified. For ILDM [8] the  $(n \text{ by } n)$ -dimensional Jacobian matrix  $V^{ILDM} = F_\psi$  (with  $\{F_\psi\}_{ij} = \partial F_i / \partial \psi_j$ ) of the chemical source terms is used to identify the fast/slow decomposition of chemical processes. For GQL [11] the analysis is based on the so-called global quasi-linearization matrix. This matrix is obtained by picking  $n$  random states in the composition space and looking for a linear representation of  $F(\psi)$ .

$$V^{GQL} = \begin{pmatrix} | & & | \\ F(\psi_1) & \cdots & F(\psi_n) \\ | & & | \end{pmatrix} \begin{pmatrix} | & & | \\ \psi_1 & \cdots & \psi_n \\ | & & | \end{pmatrix}^{-1} \quad (10)$$

Issues like the choice of the reference points cannot be discussed here, and we simply refer to [11; 35]. The equation for the low-dimensional manifolds is then obtained by an invariant subspace decomposition of the matrices  $V^{ILDM}$  or  $V^{QGL}$

$$V = \begin{pmatrix} Z_s & Z_f \end{pmatrix} \cdot \begin{pmatrix} N_s & 0 \\ 0 & N_f \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix}. \quad (11)$$

The matrices  $Z$  and  $\tilde{Z}$  span up the right invariant subspace and

the left invariant subspace, respectively:

$$\tilde{Z} = Z^{-1} = (Z_s \ Z_f)^{-1} = \begin{pmatrix} \tilde{Z}_s \\ \tilde{Z}_f \end{pmatrix}. \quad (12)$$

Here  $Z_s$  is the  $(n$  by  $n_s)$ -dimensional invariant subspace belonging to the  $n_s$  eigenvalues  $\lambda(N_s)$  having the largest real parts and  $Z_f$  is the  $(n$  by  $n_f)$ -dimensional invariant subspace belonging to the  $n_f$  eigenvalues  $\lambda(N_f)$  having the smallest real parts, respectively, where  $n_s$  and  $n_f$  denote the number of eigenvalues according to slow and fast processes. This means that the eigenvalues of the matrix  $V$  are divided into two groups

$$i = 1, \dots, n_s \quad k = n_s + 1, \dots, n_s + n_f \quad n_s + n_f = n \quad (13)$$

$$|\operatorname{Re}(\lambda_i(V))| \leq a \ll b \leq |\operatorname{Re}(\lambda_k(V))| \quad \operatorname{Re}(\lambda_k(V)) < 0 \quad (14)$$

and fulfill a spectral gap condition (for more details see [36; 11]).

The general assumption that the fast processes have already relaxed defines an  $m = n_s = n - n_f$ -dimensional manifold in the state space (note that additional constraints for the conserved variables can be used to further reduce the dimension  $m$  of the manifold[8; 36]). This subspace is composed of points where the reaction rates in direction of the  $n_f$  fast processes vanish

$$\tilde{Z}_f(\psi)F(\psi) = 0, \quad (15)$$

This under-determined equation system can be solved using path following algorithms [36] to yield an  $(m = n - n_f)$ -dimensional manifold. In this case the manifolds are parameterized by a set of  $m$  reduced coordinates (e.g. mixture fraction, reaction progress variables)  $\theta = (\theta_1, \theta_2, \dots, \theta_m)^T$ :

$$\mathcal{M} = \{\psi = \psi(\theta), \theta \in R^m, \psi \in R^n\} \quad (16)$$

The investigation of the hierarchical nature of ILDM and GQL is now quite straight forward, due to the algebraic definition of the manifolds. Let us assume that we decompose the matrix  $V$  into invariant subspaces according to two different splitting conditions with  $n_s = m$  and  $n_s = m + p$ , respectively ( $p > 0$ ).

$$V = \begin{pmatrix} Z_s^m & Z_f^m \\ 0 & N_f^m \end{pmatrix} \cdot \begin{pmatrix} N_s^m & 0 \\ 0 & N_f^m \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s^m \\ \tilde{Z}_f^m \end{pmatrix}. \quad (17)$$

$$V = \begin{pmatrix} Z_s^{m+p} & Z_f^{m+p} \\ 0 & N_f^{m+p} \end{pmatrix} \cdot \begin{pmatrix} N_s^{m+p} & 0 \\ 0 & N_f^{m+p} \end{pmatrix} \cdot \begin{pmatrix} \tilde{Z}_s^{m+p} \\ \tilde{Z}_f^{m+p} \end{pmatrix}. \quad (18)$$

In this case it follows directly from the definition of the invariant subspaces that the  $(n - (m + p))$  by  $n$ -dimensional matrix  $\tilde{Z}_f^{m+p}$  is in the span of the  $(n - m)$  by  $n$ -dimensional matrix  $\tilde{Z}_f^m$ , and together with the manifold equation (16) we obtain

$$\{\psi | \tilde{Z}_f^m F(\psi) = 0\} \subset \{\psi | \tilde{Z}_f^{m+p} F(\psi) = 0\} \quad (19)$$

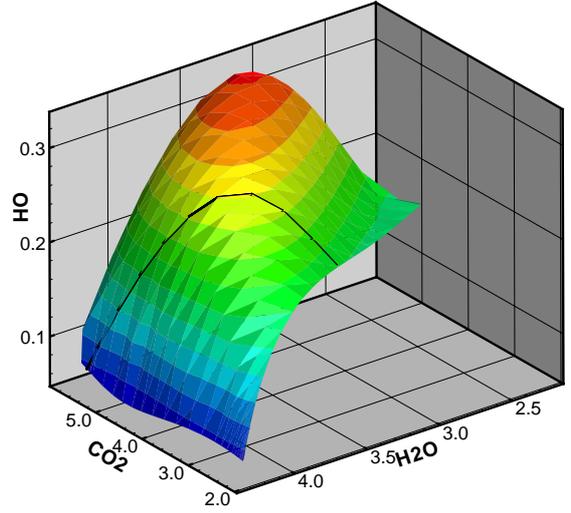


Figure 1. Example of a 1-dimensional ILDM (thick curve) embedded in a 2D-ILDM (mesh) plotted into the space of specific mole numbers (mass fractions divided by the molar mass in mol/kg) for the syngas/air system (composition and conditions are the same as in [8])

yielding

$$\mathcal{M}^m \subset \mathcal{M}^{m+p} \quad (20)$$

An example of a one-dimensional manifold embedded in a two-dimensional manifold for a syngas-air system is shown in Figure 1. Note that there exists also a hierarchy caused by the chemistry of higher hydrocarbon oxidation in the case of combustion processes. A discussion on this subject can be found in [33].

## THE HIERARCHICAL NATURE OF REDIM

In contrast to ILDM and GQL the REDIM concepts takes into account the molecular transport processes in the identification of the low-dimensional manifolds. Based on equation (1) the REDIM method uses estimates for the spatial gradients and solves an evolution equation for an  $m$ -dimensional manifold parameterized by the reduced coordinates  $\theta = (\theta_1, \theta_2, \dots, \theta_m)^T$  for a pseudo-time  $\tau \rightarrow \infty$

$$\rho \frac{\partial \psi}{\partial \tau} = (I - \psi_\theta \psi_\theta^+) \{ \rho F + (D \psi_\theta \chi)_\theta \chi \} \quad (21)$$

where  $F = F(\psi(\theta))$ ,  $D = D(\psi(\theta))$ ,  $\rho = \rho(\psi(\theta))$ ,  $\chi = \chi(\psi(\theta))$ ,  $\psi_\theta = \psi_\theta(\theta)$ ,  $\psi_\theta^+ = \psi_\theta^+(\theta)$ . In these equations  $I$  is the identity matrix,  $\psi_\theta$  is the matrix of partial derivatives of  $\psi$  with respect to  $\theta$  ( $\{\psi_\theta\}_{ij} = \partial \psi_i / \partial \theta_j$ , and  $\psi_\theta$  spans the tangent space to the manifold), and  $\psi_\theta^+$  is a pseudo-inverse (e.g. the Moore Penrose pseudo-inverse) of  $\psi_\theta$ , see [11]. Note that this is a partial differential equation system with the thermokinetic state vector  $\psi$  as dependent and the time  $t$  and the reduced coordinates  $\theta$  as independent variables.

The  $m$ -dimensional vector  $\chi$  of gradient estimates used in this equation relies on a gradient guess  $\zeta(\psi)$  for the thermokinetic

state vector  $\zeta(\psi) = \text{grad } \psi$ , which is assumed to be a function of the local thermodynamic state (see [19; 37; 38]) The gradient estimates  $\chi$  are calculated via

$$\chi = \psi_{\theta}^{+} \zeta(\psi) \quad (22)$$

The solution

$$\mathcal{M}^m = \{\psi = \psi(\theta), \theta \in R^m\} \quad (23)$$

for  $\tau \rightarrow \infty$  fulfills the invariance condition

$$(I - \psi_{\theta} \psi_{\theta}^{+}) \{\rho F + (D\psi_{\theta} \chi)_{\theta} \chi\} = 0 \quad (24)$$

For the following analysis it is useful to investigate the dependence of the REDIM evolution equation with respect to a change of the parametrization. Let a change of the parametrization of the manifold from  $\theta$  to  $\hat{\theta}$  be given by the  $(m \times m)$ -dimensional regular transformation matrix  $X = \hat{\theta}_{\theta}$ . It can be shown (see Appendix A) that the projection matrix is invariant with respect to a change of the parameterization (equation 35), and it can also be shown (see Appendix A) that the diffusion term  $\Delta = (D\psi_{\theta} \chi)_{\theta} \chi$  is invariant with respect to a change of the parameterization (equation 36). This means that the governing equation system does not depend on the choice of the parameterizing coordinates, a result which allows a simple analysis of the hierarchy of manifolds of different dimensions.

Let  $\mathcal{M}^m$  denote an  $m$ -dimensional Manifold with a tangent space defined by the matrix  $\psi_{\theta}$  of tangent vectors, and  $\mathcal{M}^{m+p}$  denote an  $m+p$ -dimensional Manifold with a tangent space defined by the matrix  $\psi_{\eta}$  of tangent vectors, where  $\theta = (\theta_1, \dots, \theta_m)^T$ , and  $\eta = (\eta_1, \dots, \eta_{m+p})^T$ . From the invariance of the equation system with respect to a change of the parameterization, it follows that we can represent the manifold  $\mathcal{M}^{m+p}$  by new coordinates  $\theta = (\theta_1, \dots, \theta_m, \xi_1, \dots, \xi_p)^T$  with

$$\psi_{\xi} \in \text{span}(\psi_{\eta}) \quad \wedge \quad \psi_{\xi} \perp \psi_{\theta}. \quad (25)$$

This has several consequences for the evolution equation (21) for the REDIM. The projection operators  $P^m$  and  $P^{m+p}$  are related via (see Appendix B).

$$(I - \psi_{\eta} \psi_{\eta}^{+}) = (I - \psi_{\theta} \psi_{\theta}^{+} - \psi_{\xi} \psi_{\xi}^{+}), \quad (26)$$

In Appendix C it is shown that if the vector of gradient estimates for  $\psi \in \mathcal{M}^m$  is tangent to the  $m$ -dimensional manifold ( $\zeta \parallel \psi_{\theta}$ ), then it follows that the  $m$ -dimensional manifold is invariant with respect to the evolution equation of the  $(m+p)$ -dimensional manifold, and therefore the  $m$ -dimensional manifold is embedded in the  $(m+p)$ -dimensional manifold.

Examples of one-dimensional manifolds embedded in a two-dimensional manifold for a syngas-air system are shown in Fig. 2 for a premixed reaction system (conditions were taken from [19]) and in Fig. 3 for a non-premixed system (conditions were taken from [20]).

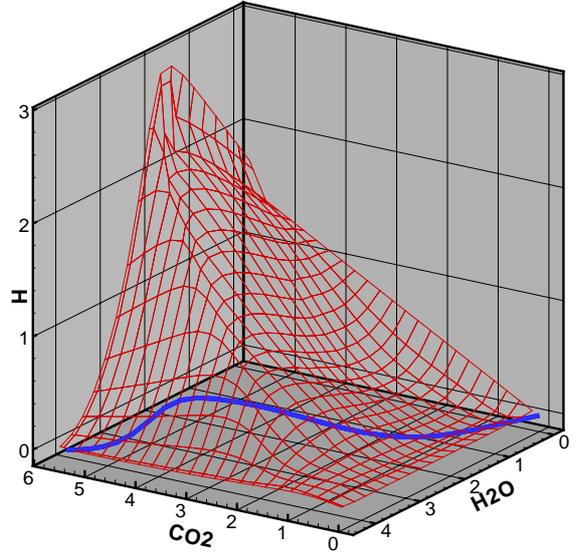


Figure 2. Example of a 1-dimensional REDIM (thick curve) embedded in a 2D-REDIM (mesh) plotted into the space of specific mole numbers (mass fractions divided by the molar mass in mol/kg) for the syngas/air system.

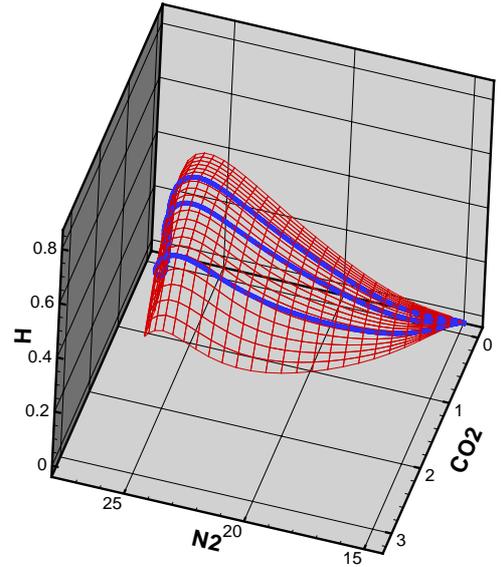


Figure 3. Example of 1-dimensional REDIMs (thick curves) for different gradient estimates embedded in a 2D-REDIM (mesh) plotted into the space of specific mole numbers (mass fractions divided by the molar mass in mol/kg) for the syngas/air system.

## CONSEQUENCES OF THE HIERARCHICAL NATURE OF LOW-DIMENSIONAL MANIFOLDS

Having shown that REDIMs exhibit a similar hierarchical nature as QSSA, PEA, ILDM and QGL, we can state that in all these concepts lower dimensional manifolds  $\mathcal{M}^m$  are embedded in higher dimensional manifolds  $\mathcal{M}^{m+p}$ . This has several consequences for the identification and the use of these low-dimensional manifolds.

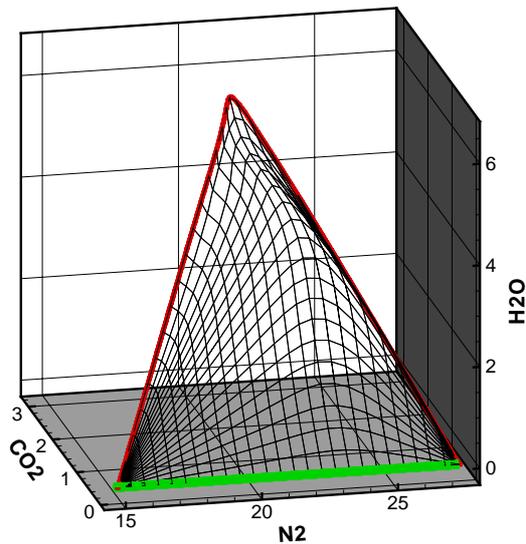


Figure 4. Example of two 1-dimensional REDIMs (thick curves) defining the boundary of a 2D-REDIM (mesh) plotted into the space of specific mole numbers (mass fractions divided by the molar mass in mol/kg) for the syngas/air system.

### Efficient hierarchical construction of higher dimensional manifolds

One aspect where the hierarchy of the low-dimensional manifolds can be used efficiently is the hierarchical generation of higher dimensional manifolds starting from lower-dimensional manifolds. In the case of ILDM this concept has already been applied successfully [39], and due to the similarity of GQL and REDIM in the identification of the manifolds [11], an extension to GQL is straight forward. Therefore we shall focus here on discussing a possible implementation in the REDIM concept. One possibility to calculate higher dimensional REDIMs starting from lower-dimensional ones is based on two ingredients. The first is to construct boundaries for  $m + 1$ -dimensional manifolds starting from  $m$ -dimensional ones. This can be illustrated by Fig. 4. It shows a two-dimensional REDIM for a non-premixed syngas system (same system as above, but Lewis number assumed to be 1). The green curve corresponds to a one-dimensional REDIM with a very high estimated gradient (corresponding to a dissipation rate much above the quenching limit). The red curve corresponds to a REDIM with gradient estimates taken from a counterflow flame with a small strain rate. An initial guess for the construction of the 2D REDIM has been obtained by a simple interpolation method (see [38]). Then the evolution equation for the REDIM has been solved to yield the 2D REDIM (mesh in the figure). This strategy can be further improved if lower-dimensional manifolds are not only used as boundaries, but also within the domain to improve convergence of the REDIM equation (cf. Fig 3).

### Efficient adaptive use of REDIMs and control of the model error

In typical computational fluid dynamics (CFD) applications the overall computational domain exists of many different domains, which are governed, e.g., by mixing and diffusion, convection and chemical reaction. The hierarchical nature of the

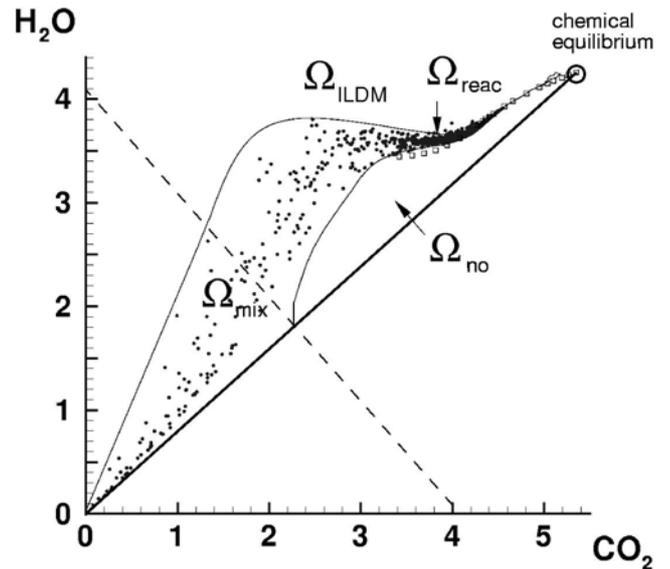


Figure 5. Effects of mixing and chemistry. Scatter plots of specific mole numbers of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  (mass fractions divided by the molar mass in mol/kg) for the syngas/air system in a partially stirred reactor, reprinted with permission from [41]

manifolds allows to use different degrees of approximation in different domains. In domains far from equilibrium (e.g. in the transient fuel conversion zone in combustion processes) high dimensional manifolds can be used, whereas close to equilibrium a representation by one or two-dimensional REDIMs is sufficient. Recently methods have become popular, which can estimate the modelling error [40]. Such methods could in future be used to estimate the error resulting from an approximation of the state space by low-dimensional manifolds of different dimensions.

### Hierarchical concepts for the chemistry/turbulence coupling

Due to the enormous computational effort, reduced mechanisms based on low-dimensional manifolds are frequently used for modeling of turbulent reacting flows [5; 4; 3]. In many cases reduced models are only used to describe the kinetics. On the other hand the coupling of the chemical kinetics with the turbulent micro-mixing is governed by the hierarchy of the kinetics, too. This can be seen from a scatter plot of a PDF-calculation of a partially stirred reactor with a syngas/air mixture [41]. Calculations were performed until a statistically stationary solution was obtained. The scatter plots in the state space using detailed chemistry are shown in Fig. 5. Obviously only a small domain is actually accessed, a behavior which is also well known from direct numerical simulations [42]. The domain  $\Omega_{mix}$  shows considerable scatter, and  $\Omega_{reac}$  shows considerably less scatter, which is caused by a one-dimensional attracting manifold (note that in this representation it cannot be identified whether in the domain  $\Omega_{mix}$  higher dimensional attractors exist. The lower boundaries of the accessed domain is the mixing line between the unburnt mixture (origin) and the completely burnt mixture (chemical equilibrium state). Due to fast chemical reaction the domain  $\Omega_{no}$  is never accessed, because even if mixing brought particles into this domain, fast chemistry would relax the states towards the accessed domain (note that the re-

sults of the calculation were plotted after the chemistry step in the PDF method [41]. From these observations we can state that the chemical kinetics does not only give rise to low-dimensional manifolds in composition space, but that it also influences the chemistry/turbulence coupling. In methods like the conditional moment closure (CMC) [43; 23] or multiple mapping conditioning (MMC) [24] exactly this effect is used in the statistical treatment of turbulent flows. Whereas in CMC the correlations are used explicitly to describe the thermokinetic state, in MMC the correlations introduced by the low-dimensional attractors in composition space are used only for conditioning mixing operation, which is not necessarily confined to a low-dimensional manifold associated with the attractor.

### The Manifold's Hierarchy in the context of MMC

Based on the observations made above, we suggest to use the hierarchical structure of the manifolds in the context of MMC. The basic idea is to use the results from a low-dimensional manifold  $\mathcal{M}^m$  to represent the evolution of the reference variables. Let  $\psi(\theta(\mathbf{x}, t))$  represent a solution of a transport problem obtained with a high spatial resolution and the use of low-dimensional manifolds parametrized by  $\theta = (\theta_1, \dots, \theta_m)^T$ . The function  $\theta^\circ = \theta(\mathbf{x}^\circ(t), t)$  denotes the values of  $\theta$  evaluated at Lagrangian trajectories — these values are deployed as MMC reference variables. The scalars are modelled by quantities  $\psi^*(t)$  that represent unrestricted kinetics (or kinetics confined to a manifold of a larger dimension  $\mathcal{M}^{m+p}$  in a hierarchical treatment of the problem) evaluated on a system of Pope particles; only relatively few particles are to be used in calculations (the reader is referred to sparse-Lagrangian methods [44]). Mixing operation is performed in MMC with localization in the combined space of the reference variables and physical coordinates. This enforces transport properties, which are accurately evaluated for  $\theta^\circ$ , on scalars  $\psi^*$  that have only sparse representations in physical space. Unrestricted treatment of chemical kinetics gives indication whether solution deviates from the  $m$ -dimensional manifold. Let us consider this point by introducing  $\psi^\circ = \psi(\theta^\circ(t))$ . These values do not coincide with  $\psi^*(t)$  but, if the chemical kinetics of the fast subspace is indeed fast,  $\psi^*$  can be represented by  $\psi^* = \psi(\theta^*(t))$ . Note that values  $\theta^*$  do not coincide with  $\theta^\circ$ , and can experience so called minor fluctuations with respect to  $\theta^\circ$ . The main difference between  $\psi^*$  and  $\psi^\circ$  is that  $\psi^*$  is not forced to be confined to the  $m$ -dimensional manifold and may deviate from the manifold when the absolute values of the eigenvalues of the fast subspace are not sufficiently large. The use of the manifold-constructed reference variables  $\theta^\circ$  plays an important role in the method, as these variables allow to perform economical evaluations of  $\psi^*$  on a very sparse system of particles. The suggested method combines advantages of the manifold-based reduction techniques and MMC. In principle, a hierarchical system of manifolds of increasing dimensions can be deployed to form a hierarchical system of MMC reference variables.

### ACKNOWLEDGMENT

The authors acknowledge funding by the Australian Research Council and the Deutsche Forschungsgemeinschaft.

### A INVARIANCE OF THE REDIM WITH RESPECT TO THE PARAMETERIZATION

Let a change of the parametrization of the manifold from  $\theta$  to  $\hat{\theta}$  be given by the  $(m \times m)$ -dimensional regular transformation matrix

$$X = \hat{\theta}_\theta \quad (27)$$

where  $\psi_\theta$  transforms according to

$$\psi_{\hat{\theta}} = \psi_\theta \theta_{\hat{\theta}} \quad (28)$$

and  $\chi(\theta)$  transforms according to

$$\hat{\chi}(\hat{\theta}) = \psi_{\hat{\theta}}^+ \zeta(\psi) = (\psi_\theta \theta_{\hat{\theta}})^+ \zeta(\psi) = \theta_{\hat{\theta}}^{-1} \psi_\theta^+ \zeta(\psi) = \theta_{\hat{\theta}}^{-1} \chi(\theta) \quad (29)$$

and

$$\hat{\theta}_\theta \theta_{\hat{\theta}} = I, \quad \theta_{\hat{\theta}} = \hat{\theta}_\theta^{-1} \quad (30)$$

The projection matrix is given as

$$P = (I - \psi_\theta \psi_\theta^+), \quad (31)$$

and in the new parameterization the projection matrix  $\hat{P}$  is given as

$$\hat{P} = (I - \psi_{\hat{\theta}} \psi_{\hat{\theta}}^+) \quad (32)$$

Using  $\psi_{\hat{\theta}} = \psi_\theta \theta_{\hat{\theta}}$  we obtain

$$\psi_{\hat{\theta}}^+ = (\psi_\theta \theta_{\hat{\theta}})^+ \quad (33)$$

which yields

$$\psi_{\hat{\theta}}^+ = \theta_{\hat{\theta}}^{-1} \psi_\theta^+ \quad (34)$$

yielding

$$\hat{P} = (I - \psi_{\hat{\theta}} \psi_{\hat{\theta}}^+) = (I - \psi_\theta \theta_{\hat{\theta}} \theta_{\hat{\theta}}^{-1} \psi_\theta^+) = (I - \psi_\theta \psi_\theta^+) = P \quad (35)$$

This means that the projection matrix is invariant with respect to a change of the parameterization. The diffusion term  $\Delta = (D\psi_\theta \chi)_\theta \chi$  with the changed parametrization is:

$$\begin{aligned} \hat{\Delta} &= (D\psi_{\hat{\theta}} \hat{\chi})_{\hat{\theta}} \hat{\chi} = (D\psi_\theta \theta_{\hat{\theta}} \hat{\theta}_\theta \chi)_{\hat{\theta}} \hat{\chi} \\ &= (D\psi_\theta \theta_{\hat{\theta}} \hat{\theta}_\theta \chi)_\theta \theta_{\hat{\theta}} \hat{\theta}_\theta \chi = (D\psi_\theta \chi)_\theta \chi = \Delta \end{aligned} \quad (36)$$

This means that the diffusion term is invariant with respect of a change of the parameterization, too.

## B EMBEDDING REACTION/DIFFUSION MANIFOLDS

At each  $\psi \in \mathcal{M}^m$  the tangent space of  $\mathcal{M}^{m+p}$  can be represented by an alternative set of basis vectors given by

$$\psi_\eta = \begin{pmatrix} | & | & & | \\ \psi_{\eta_1} & \psi_{\eta_2} & \cdots & \psi_{\eta_{m+p}} \\ | & | & & | \end{pmatrix} = \begin{pmatrix} | & | & | & | \\ \psi_{\theta_1} & \cdots & \psi_{\theta_m} & \psi_{\xi_1} & \cdots & \psi_{\xi_p} \\ | & | & | & | \end{pmatrix} \cdot Z \quad (37)$$

or in simplified notation

$$\psi_\eta = (\psi_\theta \quad \psi_\xi) \cdot Z, \quad (38)$$

where  $Z$  denotes an  $(m+p)$  by  $(m+p)$ -dimensional regular transformation matrix, and where  $\psi_\xi$  is constructed such that it is in the span of  $\psi_\eta$  and orthogonal to  $\psi_\theta$ :

$$\psi_\xi \in \text{span}(\psi_\eta) \quad \wedge \quad \psi_\xi \perp \psi_\theta. \quad (39)$$

Noting that the projection operator  $P$  is invariant with respect to a change of the parameterization (see 35), we obtain

$$(I - \psi_\eta \psi_\eta^+) = (I - (\psi_\theta \quad \psi_\xi)(\psi_\theta \quad \psi_\xi)^+), \quad (40)$$

and because  $\psi_\xi$  is orthogonal to  $\psi_\theta$  it follows that

$$\begin{aligned} (I - \psi_\eta \psi_\eta^+) &= \left( I - (\psi_\theta \quad \psi_\xi) \begin{pmatrix} \psi_\theta^+ \\ \psi_\xi^+ \end{pmatrix} \right) \\ &= \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right). \end{aligned} \quad (41)$$

From this several useful properties can be derived:

$$\begin{aligned} (I - \psi_\eta \psi_\eta^+) \psi_\xi &= \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) \psi_\xi \\ &= \left( I - \psi_\xi \psi_\xi^+ \right) \psi_\xi = 0, \end{aligned} \quad (42)$$

$$\begin{aligned} (I - \psi_\eta \psi_\eta^+) \psi_\theta &= \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) \psi_\theta \\ &= \left( I - \psi_\theta \psi_\theta^+ \right) \psi_\theta = 0, \end{aligned} \quad (43)$$

$$\begin{aligned} P^{n+p} P^n &= (I - \psi_\eta \psi_\eta^+) (I - \psi_\theta \psi_\theta^+) \\ &= \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) (I - \psi_\theta \psi_\theta^+) \\ &= (I - \psi_\theta \psi_\theta^+) (I - \psi_\theta \psi_\theta^+) - (\psi_\xi \psi_\xi^+) (I - \psi_\theta \psi_\theta^+) \\ &= (I - \psi_\theta \psi_\theta^+) - (\psi_\xi \psi_\xi^+) (I - \psi_\theta \psi_\theta^+) \\ &= \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) = P^{n+p} \end{aligned} \quad (44)$$

$$\begin{aligned} P^n P^{n+p} &= (I - \psi_\theta \psi_\theta^+) (I - \psi_\eta \psi_\eta^+) \\ &= (I - \psi_\theta \psi_\theta^+) \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) \\ &= (I - \psi_\theta \psi_\theta^+) (I - \psi_\theta \psi_\theta^+) - (I - \psi_\theta \psi_\theta^+) (\psi_\xi \psi_\xi^+) \\ &= (I - \psi_\theta \psi_\theta^+) - (I - \psi_\theta \psi_\theta^+) (\psi_\xi \psi_\xi^+) \\ &= \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) = P^{n+p} \end{aligned} \quad (45)$$

## C ANALYSIS OF THE HIERARCHY OF THE REDIM INVARIANCE CONDITION

Let  $\psi \in \mathcal{M}^m$  fulfill the invariance equation

$$0 = \psi_\theta^{\perp T} \{ \rho F + (D\psi_\theta \chi)_\theta \chi \} \quad (46)$$

which is in this case equivalent to

$$\{ \rho F + (D\psi_\theta \chi)_\theta \chi \} = \psi_\theta X, \quad (47)$$

where  $X$  denotes the coordinates in the local coordinate system of the  $\psi_\theta$ . The evolution of the  $\mathcal{M}^{m+p}$ -dimensional manifold is given by

$$\rho \frac{\partial \psi}{\partial t} = (I - \psi_\eta \psi_\eta^+) \{ \rho F + (D\psi_\eta \vartheta)_\eta \vartheta \} \quad (48)$$

with

$$\vartheta = \psi_\eta^+ \zeta(\psi) \quad (49)$$

Using the transformation equations for the projection term we obtain.

$$\rho \frac{\partial \psi}{\partial t} = \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) \{ \rho F + (D\psi_\eta \vartheta)_\eta \vartheta \} \quad (50)$$

In the local coordinate system the gradient estimate vector is given by

$$\hat{\vartheta} = \begin{pmatrix} \chi \\ \sigma \end{pmatrix} = \begin{pmatrix} \psi_\theta^+ \\ \psi_\xi^+ \end{pmatrix} \zeta \quad (51)$$

Because of the invariance of the diffusion term we have

$$\begin{aligned} (D\psi_\eta \vartheta)_\eta \vartheta &= \left\{ D\psi_{(\theta\xi)} \begin{pmatrix} \chi \\ \sigma \end{pmatrix} \right\}_{(\theta\xi)} \begin{pmatrix} \chi \\ \sigma \end{pmatrix} \\ &= (D\psi_\theta \chi)_\theta \chi + (D\psi_\theta \chi)_\xi \sigma + (D\psi_\xi \sigma)_\theta \chi + (D\psi_\xi \sigma)_\xi \sigma \end{aligned} \quad (52)$$

If  $\zeta \parallel \psi_\theta$  it follows with  $\psi_\xi \perp \psi_\theta$  that  $\sigma = 0$ , and we obtain

$$(D\psi_\eta \vartheta)_\eta \vartheta = (D\psi_\theta \chi)_\theta \chi \quad (53)$$

This results in an evolution equation for the subset  $\mathcal{M}^m$  according to

$$\rho \frac{\partial \psi}{\partial t} = \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) \{ \rho F + (D\psi_\theta \chi)_\theta \chi \} \quad (54)$$

and using  $\{ \rho F + (D\psi_\theta \chi)_\theta \chi \} = \psi_\theta X$

$$\begin{aligned} \rho \frac{\partial \psi}{\partial t} &= \left( I - \psi_\theta \psi_\theta^+ - \psi_\xi \psi_\xi^+ \right) \psi_\theta X \\ &= \left( \psi_\theta - \psi_\theta \psi_\theta^+ \psi_\theta - \psi_\xi \psi_\xi^+ \psi_\theta \right) X = 0 \end{aligned} \quad (55)$$

This means that an  $m$ -dimensional manifold is (for the given conditions) invariant with respect to the evolution equation of the  $(m+p)$ -dimensional manifold.

## REFERENCES

- [1] J. O. Hirschfelder and C. F. Curtiss. *Theory of Propagation of Flames. Part I: General Equations*, page 121. Williams and Wilkins, Baltimore, 1949.
- [2] C.K. Westbrook, Y. Mizobuchi, T.J. Poinso, P.J. Smith, and J. Warnatz. Computational combustion. *Proc. Comb. Inst.*, 30(1):125 – 157, 2005.
- [3] N. Peters. Multiscale combustion and turbulence. *Proceedings of the Combustion Institute*, 32(1):1 – 25, 2009.
- [4] Stephen B. Pope. Small scales, many species and the manifold challenges of turbulent combustion. *Proceedings of the Combustion Institute*, 34(1):1 – 31, 2013.
- [5] T. Echekki and E. Mastorakos. *Turbulent Combustion Modeling: Advances, New Trends and Perspectives*. Fluid mechanics and its applications. Springer Netherlands, 2011.
- [6] S. H. Lam and D. A. Goussis. Understanding complex chemical kinetics with computational singular perturbation. In *22<sup>nd</sup> Symposium (International) on Combustion*, page 931. The Combustion Institute, Pittsburgh, PA, 1988.
- [7] S.H. Lam and D.A. Goussis. The csp method for simplifying kinetics. *Intl. J. of Chem. Kinetics*, 26:461–486, 1994.
- [8] U. Maas and S.B. Pope. Simplifying chemical kinetics: Intrinsic low-dimensional manifolds in composition space. *Combustion and Flame*, 88:239–264, 1992.
- [9] U. Maas and S.B. Pope. Laminar flame calculations using simplified chemical kinetics based on intrinsic low-dimensional manifolds. In *25<sup>th</sup> Symposium (International) on Combustion*, pages 1349–1356. The Combustion Institute, Pittsburgh, PA, 1994.
- [10] Z. Ren, S.B. Pope, A. Vladimirov, and J.M. Guckenheimer. The invariant constrained equilibrium edge preimage curve method for the dimension reduction of chemical kinetics. *The Journal of Chemical Physics*, 124(11):114111, 2006.
- [11] V. Bykov, V. Gol'dshteyn, and U. Maas. Simple global reduction technique based on decomposition approach. *Combustion Theory and Modelling*, 12(2):389–405, 2008.
- [12] A.N. Gorban and I.V. Karlin. Method of invariant manifold for chemical kinetics. *Chemical Engineering Science*, 58(21):4751 – 4768, 2003.
- [13] M. Valorani, H.N. Najm, and D.A. Goussis. Csp analysis of a transient flame-vortex interaction: Time scales and manifolds. *Comb. Flame*, 134:35–53, 2003.
- [14] D. Lebiecz. Computing minimal entropy production trajectories: An approach to model reduction in chemical kinetics. *The Journal of Chemical Physics*, 120(15):6890–6897, 2004.
- [15] J.A Van Oijen and L.P.H. De Goeij. Modelling of premixed laminar flames using flamelet-generated manifolds. *Combustion, Science and Technology*, 161:113, 2000.
- [16] P.D. Nguyen, L. Vervisch, V. Subramanian, and P. Domingo. Multidimensional flamelet-generated manifolds for partially premixed combustion. *Combustion and Flame*, 157(1):43 – 61, 2010.
- [17] S.H. Lam. Reduced chemistry-diffusion coupling. *Comb. Sci. Tech.*, 179:767–786, 2007.
- [18] D.A. Goussis, M. Valorani, F. Creta, and H.N. Najm. Reactive and reactive-diffusive time scales in stiff reaction-diffusion systems. *Prog. Comp. Fluid Dyn.*, 5:316–326, 2005.
- [19] V. Bykov and U. Maas. The extension of the ildm concept to reaction-diffusion manifolds. *Combustion Theory and Modelling*, 11(6):839–862, 2007.
- [20] U. Maas and V. Bykov. The extension of the reaction/diffusion manifold concept to systems with detailed transport models. *Proceedings of the Combustion Institute*, 33(1):1253 – 1259, 2011.
- [21] Dimitris A. Goussis and Ulrich Maas. Model reduction for combustion chemistry. In Tarek Echekki and Epaminondas Mastorakos, editors, *Turbulent Combustion Modeling*, volume 95 of *Fluid Mechanics and Its Applications*, pages 193–220. Springer Netherlands, 2011.
- [22] N. Peters. Laminar diffusion flamelet models in non-premixed turbulent combustion. *Prog. Energy Combust. Sci.*, 10:319–339, 1984.
- [23] A.Y. Klimenko and R.W. Bilger. Conditional moment closure for turbulent combustion. *Progress in Energy and Combustion Science*, 25(6):595–687, 1999. cited By (since 1996)333.
- [24] A.Y. Klimenko and S.B. Pope. The modeling of turbulent reactive flows based on multiple mapping conditioning. *Physics of Fluids*, 15(7):1907–1925, 2003. cited By (since 1996)51.
- [25] J. O. Hirschfelder and C. F. Curtiss. *Molecular Theory of Gases and Liquids*. John Wiley & Sons, Inc., New York, 1964.
- [26] A. Ern and V. Giovangigli. *Multicomponent Transport Algorithms*. Lecture Notes in Physics. Springer, Berlin, Heidelberg, New York, 1994.
- [27] J.O. Hirschfelder, C.F. Curtiss, and R.B. Bird. *Molecular Theory of Gases and Liquids*. Wiley, New York, 1954.
- [28] R.B. Bird, W.E. Stewart, and E.N. Lightfoot. *Transport Phenomena*. Wiley Interscience, New York, 1960.
- [29] M.R. Roussel and S.J. Fraser. On the geometry of transient relaxation. *The Journal of Chemical Physics*, 94(11):7106–7113, 1991.
- [30] M. Bodenstein. Zur kinetik des chlorknallgases. *Z. Phys. Chem.*, 85:329, 1913.
- [31] U. Maas and S.B. Pope. Implementation of simplified chemical kinetics based on intrinsic low-dimensional manifolds. In *24<sup>th</sup> Symposium (International) on Combustion*, page 103. The Combustion Institute, Pittsburgh, PA, 1992.
- [32] T. Blasenbrey, D. Schmidt, and U. Maas. Automatically simplified chemical kinetics and molecular transport and its application in premixed and non-premixed laminar flame calculations. In *27<sup>th</sup> Symposium (International) on Combustion*. The Combustion Institute, Pittsburgh, PA, 1998.
- [33] T. Blasenbrey and U. Maas. ILDMs of higher hydrocarbons and the hierarchy of chemical kinetics. *Proc. Comb. Inst.*, 28:1623–1630, 2000.
- [34] J. Nafe and U. Maas. A general algorithm for improving ILDMs. *Combustion Theory and Modeling*, 6(4):697–709, 2002.
- [35] V. Bykov and U. Maas. Investigation of the hierarchical structure of kinetic models in ignition problems. *Zeitschrift für Physikalische Chemie*, 233(4-5):461–479, 2009.
- [36] U. Maas. Efficient calculation of intrinsic low-dimensional manifolds for the simplification of chemical kinetics. *Computing and Visualization in Science*, 1:69–

- 82, 1998.
- [37] V. Bykov and U. Maas. Problem adapted reduced models based on reaction-diffusion manifolds (redims). In *Proc. Comb. Inst.*, volume 32, pages 561–568. Elsevier, 2009.
  - [38] Gerd Steinhilber and Ulrich Maas. Reaction-diffusion manifolds for unconfined, lean premixed, piloted, turbulent methane/air systems. *Proceedings of the Combustion Institute*, 34(1):217 – 224, 2013.
  - [39] J. Nafe and U. Maas. Hierarchical generation of {ILDMS} of higher hydrocarbons. *Combustion and Flame*, 135(12):17 – 26, 2003.
  - [40] M. Braack and A. Ern. A posteriori control of modeling errors and discretization errors. *Multiscale Modeling and Simulation*, 1(2):221–238, 2003.
  - [41] R. Bender, T. Blasenbrey, and U. Maas. Coupling of detailed and ildm-reduced chemistry with turbulent mixing. *Proc. Comb. Inst.*, 28:101–106, 2000. Figure reprinted with permission from Elsevier.
  - [42] U. Maas and D. Thevenin. Correlation analysis of direct numerical simulation data of turbulent non-premixed flames. In *27<sup>th</sup> Symposium (International) on Combustion*. The Combustion Institute, Pittsburgh, PA, 1998.
  - [43] R. W. Bilger. Conditional moment closure for the turbulent reacting flow. *Phys. Fluids A*, 5:436, 1993.
  - [44] Yipeng Ge, M.J. Cleary, and A.Y. Klimenko. Sparse-lagrangian {FDF} simulations of sandia flame e with density coupling. *Proceedings of the Combustion Institute*, 33(1):1401 – 1409, 2011.