

Analysis and Reduction of a Simplified Stochastic Chemical System via CSP

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Abstract—We explore the application of computational singular perturbation (CSP) to analyze the stochastic dynamics of an idealized, stiff, 3-component chemical system, characterized by a single small parameter ϵ . Attention is focused on the dynamics of the system under uncertain stiffness parameter, ϵ , and uncertain initial conditions. Polynomial chaos (PC) expansions are used to represent stochastic quantities, and the stochastic dynamics are described in terms of the Galerkin form of the PC reformulated system. The Galerkin form of the Jacobian of the PC reformulated system is also obtained, based on re-using the known form of the deterministic system Jacobian. A stochastic CSP framework is developed based on an analysis of the stochastic eigenvalue problem. Specifically, we seek an efficient, compact representation of the stochastic eigenvectors in terms of a linear combination of the eigenvectors of a nominal deterministic system. Two approaches are used to determine the stochastic coefficients in this non-orthonormal basis, based on non-intrusive spectral projections, or on a residual minimization technique. Computational tests indicate that both approaches result in suitable representations of the stochastic eigenvectors which are in close agreement with each other. The stochastic eigenvector representation is then exploited to conduct a CSP reduction of the chemical system. For systems involving small or moderate input uncertainty levels, computational experiments indicate that the resulting stochastic CSP formalism provides an effective means of dimension reduction, and accordingly leads to significant speedup in the integration of the stochastic dynamics.

I. INTRODUCTION

The equations of motion of reacting flows are frequently stiff, primarily due to the large spectrum of timescales that characterize individual reactions. It is by now well established that efficient simulation of such systems can tremendously benefit from methods that can address the stiffness of the governing equations and/or reduce the dimension of the system being modeled. In particular, CSP [1], [2], [3] has been found to be an effective tool in identifying disparate chemical timescales, and where appropriate formulating reduced models and/or substantially enhancing integration efficiency.

In this work, we explore the application of CSP to a simplified, stiff, stochastic chemical system. We focus specifically on the case of a purely reactive system with uncertain rate parameters and initial conditions. A spectral approach to uncertainty quantification is adopted, based on the use of polynomial chaos (PC) expansions. As outlined [4], these essentially amount to approximating random variables in terms of a linear combination of orthogonal basis functions. The coefficients in the expansion can then be determined based on suitable discrete

projections or a weighted residual formalism. We will specifically focus on the latter approach, which leads to a reformulated system of coupled ordinary differential equations (ODEs) that govern the evolution of the expansion coefficients.

Though CSP may be directly applied to the reformulated stochastic chemical system, an alternative methodology is explored in Section II based on an analysis of the Jacobian of the reformulated system. Specifically, we exploit the known structure of this Jacobian to construct a spectral representation of eigenvalues and eigenvectors in the original solution space. A compact representation of the stochastic eigenvectors is then constructed in terms of a linear combination of the eigenvectors of a nominal deterministic system. The coefficients in the corresponding expansions are determined based on non-intrusive spectral projections [4], or on a residual minimization technique [5].

The availability of spectral representations of stochastic eigenvalues and eigenvectors in the form outlined above is exploited to construct a stochastic CSP reduction methodology that is especially geared for exploring conditions under which a deterministic reduced model remains valid under uncertainty. Thus, this methodology specifically identifies situations when stochastic eigenvectors associated with a nominal deterministic direction can all be considered exhausted. Application of this formalism is briefly illustrated in Section III using a three-dimensional, stiff, chemical system [2].

II. APPROACH

Development of the methodology outlined above is illustrated for the simple case of a non-linear, stiff chemical system governing the evolution of three species [3]:

$$\begin{aligned} \dot{y}_1 &= -\frac{5y_1}{\epsilon_0} - \frac{y_1y_2}{\epsilon_0} + y_2y_3 + \frac{5y_2^2}{\epsilon_0} + \frac{y_3}{\epsilon_0} - y_1 \\ \dot{y}_2 &= \frac{10y_1}{\epsilon_0} - \frac{y_1y_2}{\epsilon_0} - y_2y_3 - \frac{10y_2^2}{\epsilon_0} + \frac{y_3}{\epsilon_0} + y_1 \\ \dot{y}_3 &= \frac{y_1y_2}{\epsilon_0} - y_2y_3 - \frac{y_3}{\epsilon_0} + y_1 \end{aligned} \quad (1)$$

where y_i , $i = 1, \dots, N$ denote the vector of concentrations ($N = 3$ presently), and $\epsilon \ll 1$ is a “stiffness” parameter. A stochastic variant of the above deterministic system is considered, in which the inverse of the stiffness parameter and the initial concentrations are modelled as independent uniformly distributed random variables,

each with a given mean and range. Specifically, they are expressed as $y_i(0) = p_i + \alpha_i \xi_i$, $i = 1, \dots, N$, and $\epsilon^{-1} = \epsilon_0^{-1} + \alpha_4 \xi_4$, where ξ_i , $i = 1, \dots, 4$ are independent random variables that are uniformly distributed on $[-1, 1]$.

The dynamics of the resulting stochastic chemical system are described in terms of a truncated PC expansion:

$$\dot{y}_i(t) = \sum_{k=0}^P y_i^k(t) \Psi_k(\boldsymbol{\xi}) \quad (2)$$

where the Ψ_k are orthogonal multidimensional Legendre polynomials, $\boldsymbol{\xi} = (\xi_1, \xi_2, \xi_3, \xi_4)^T$ is the vector of independent random variables, and $P + 1$ is the number of polynomials of order $\leq p$, and $y_i^k(t)$, $i = 1, \dots, N$, $k = 0, \dots, P$ are deterministic coefficients. To describe the evolution of the stochastic solution, it is sufficient to determine the evolution of the deterministic coefficients, y_i^k . A Galerkin procedure is used for this purpose, based on inserting (2) into (1), and projecting onto the PC basis. This leads into a reformulated system of the form:

$$\dot{y}_i^k = G_i^k, \quad i = 1, \dots, N; \quad k = 0, \dots, P \quad (3)$$

where G_i^k is a nonlinear source term that is a function of all the coefficients.

Straightforward differentiation of the source term in (3) results in a stochastic Jacobian of the form \mathcal{J}_{ij}^{kl} . Following [5], this matrix is stored in 2-index form $\mathcal{J}_{\{ik\}\{jl\}}$ using the convention $\{ik\} \equiv i + kN$. Thus, \mathcal{J} acts on $N(P + 1)$ dimensional vectors, which consist of $P + 1$ N -dimensional column vectors arranged consecutively.

While the CSP formalism may be directly applied using \mathcal{J} , an alternative approach is developed based on relating \mathcal{J} to the $N \times N$ Jacobian $J_{ij}(\boldsymbol{\xi})$ that corresponds to a specific realization of $\boldsymbol{\xi}$. Applying the appropriate Gâteaux derivatives to the stochastic source term, G_i^k , one can show that the Jacobian J_{ij} has the PC representation:

$$J_{ij}(\boldsymbol{\xi}) = \sum_{k=0}^P \mathcal{J}_{ij}^{k0} \Psi_k(\boldsymbol{\xi}) \quad (4)$$

which provides a direct connection between J and \mathcal{J} .

The PC expansion of the Jacobian (4) may be readily exploited to construct PC representations of the eigenvalues and eigenvectors of the stochastic chemical system. We have explored two approaches for this purpose. The first is based on non-intrusive spectral projection, based on deterministic sampling of a fully-tensored Gauss quadrature formula, whereas the second is based on an extension of the residual minimization formalism of Ghanem & Gosh [5]. Both approaches lead to suitable representations of the eigenvalues and eigenvectors of the stochastic system, respectively:

$$\lambda_i(\boldsymbol{\xi}) = \sum_0^P \lambda_i^k \Psi_k(\boldsymbol{\xi}) \quad (5)$$

$$\mathbf{u}_i(\boldsymbol{\xi}) = \sum_0^P \mathbf{u}_i^k \Psi_k(\boldsymbol{\xi}). \quad (6)$$

In the implementations below, we focus on a simplified setting in which the nominal system corresponding to

$\boldsymbol{\xi} = 0$ has linearly independent (normalized) eigenvectors, \mathbf{V}_i , $i = 1, \dots, 3$. We exploit this assumption by re-expressing the stochastic eigenvectors in terms of this generally non-orthogonal basis, namely according to:

$$\mathbf{u}_i(\boldsymbol{\xi}) = \sum_{k=1}^N g_i^k(\boldsymbol{\xi}) \mathbf{V}_k \quad (7)$$

where g_i^k is the coefficient of the i -th eigenvector, \mathbf{u}_i , “along” the k -th nominal eigenvector. Note that the $g_i^k(\boldsymbol{\xi})$ are (non-orthogonal) polynomials in $\boldsymbol{\xi}$ of order $\leq p$.

III. ILLUSTRATIONS

One of the advantages of the representations (5) and (7) is that they directly enable us to quantify the dependence of the eigenvalues and the orientation of normalized eigenvectors on the germ $\boldsymbol{\xi}$. Figure 1 shows PDFs of the

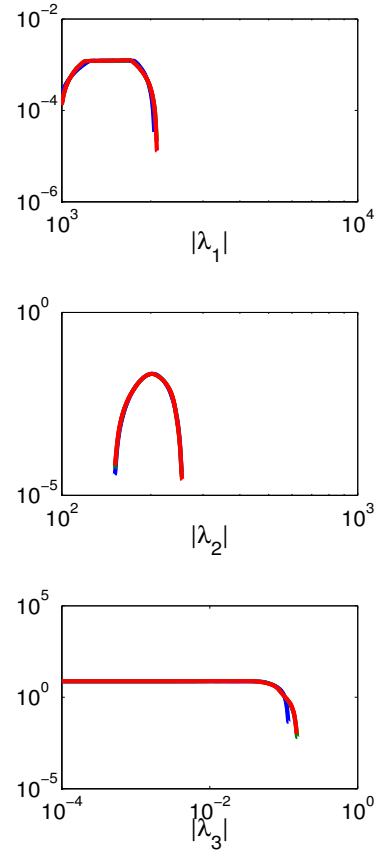


Fig. 1. PDFs of $|\lambda_i|$ at $t = 0$, $i = 1, \dots, 3$. Plotted are curves expansion orders $p = 1$ (blue), $p = 2$ (green), and $p = 3$ (red). Results are obtained with $p_i = 0.5$, $\alpha_i = 0.2$, $i = 1, \dots, 3$, $\epsilon_0 = 0.01$, $\alpha_4 = 10$.

eigenvalues for the stochastic system at $t = 0$. The results indicate good agreement between first, second, and third-order PC expansions, and that for the present example the eigenvalues remain well separated despite the large variability associated with the stochastic inputs.

To quantify the response of the normalized eigenvectors on ξ , we compute the spectra of the cosines between $\mathbf{u}_i(\xi)$ and the nominal eigenvector \mathbf{V}_i . Specifically, we set $\cos(\zeta_i) = \mathbf{u}_i(\xi) \cdot \mathbf{V}_i$, and plot in Fig. 2 the PC spectra of $\cos(\zeta_i)$ at time $t = 0$. The spectra clearly indicate that

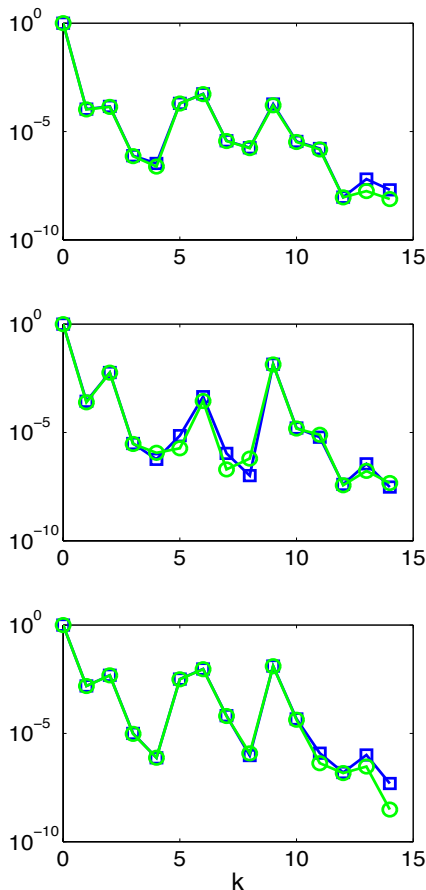


Fig. 2. PC spectra of $\cos(\zeta_i)$ at $t = 0$, for $i = 1, \dots, 3$, arranged from top. Plotted are curves obtained with residual minimization (green) NISP (blue). Same parameters as in Fig. 1.

the amplitude of the zeroth mode is near unity, and that this mode is several orders of magnitude larger than the remaining modes. Thus, for this system, uncertainty in initial conditions and stiffness parameter does not result in large variation in the orientation of the normalized eigenvectors at $t = 0$.

Examination of the dynamics of the present stochastic system (not shown) also indicates that the stochastic eigenvectors remain weakly sensitive to ξ at later times as well. Thus, for the present system, CSP reduction of the dynamics may be conducted using either the nominal eigenvectors or the stochastic eigenvectors as represented in (7). Both approaches have been attempted, based on the use of a simplified stochastic exhaustion criterion, and in both cases nearly identical results were obtained.

A sample of this exercise is shown in Fig. 3, which depicts the evolution of selected modes of the solution. The results in Fig. 3 are consistent with the analysis

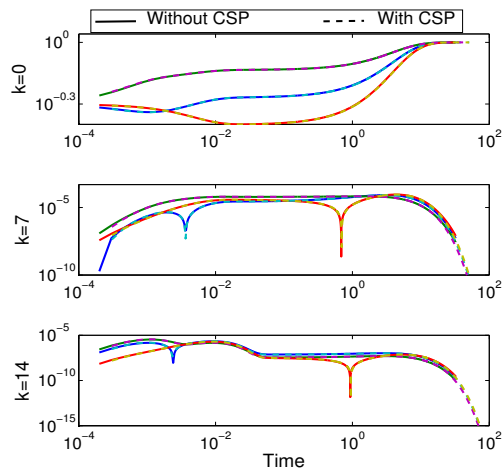


Fig. 3. Evolution of PC modes 0, 7, and 14. Plotted are curves for y_1 (blue), y_2 (green) and y_3 (red), obtained using fine-step integration (solid) and with CSP acceleration (dash). Same parameters as in Fig. 1.

in Ref. [3], the present system is attracted towards the deterministic equilibrium, $y_i = 1$, $i = 1, \dots, 3$, regardless of the initial condition and of the stiffness parameters. They also indicate a close agreement between direct and CSP-accelerated integration. For the present application, CSP integration of the stochastic chemical system with $p = 3$ leads to about a 50-fold speed up when nominal eigenvectors are used, and about a 35-fold speed up when the stochastic eigenvector representation is used.

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