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Asymptotic analysis of two reduction methods for systems of chemical reactions

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Abstract

This paper concerns two methods for reducing large systems of chemical kinetics equations, namely, the method of intrinsic low-dimensional manifolds (ILDMs) due to Maas and Pope [Combust. Flame 88 (1992) 239] and an iterative method due to Fraser [J. Chem. Phys. 88 (1988) 4732] and further developed by Roussel and Fraser [J. Chem. Phys. 93 (1990) 1072]. Both methods exploit the separation of fast and slow reaction time scales to find low-dimensional manifolds in the space of species concentrations where the long-term dynamics are played out. The asymptotic expansions of these manifolds ($\varepsilon \downarrow 0$, where ε measures the ratio of the reaction time scales) are compared with the asymptotic expansion of $\mathcal{M}_\varepsilon$, the slow manifold given by geometric singular perturbation theory. It is shown that the expansions of the ILDM and $\mathcal{M}_\varepsilon$ agree up to and including terms of $\mathcal{O}(\varepsilon)$; the former has an error at $\mathcal{O}(\varepsilon^2)$ that is proportional to the local curvature of $\mathcal{M}_0$. The error vanishes if and only if the curvature is zero everywhere. The iterative method generates, term by term, the asymptotic expansion of $\mathcal{M}_\varepsilon$. Starting from $\mathcal{M}_0$, the ith application of the algorithm yields the correct expansion coefficient at $\mathcal{O}(\varepsilon^i)$, while leaving the lower-order coefficients invariant. Thus, after ℓ applications, the expansion is accurate up to and including the terms of $\mathcal{O}(\varepsilon^\ell)$. The analytical results are illustrated on a planar system from enzyme kinetics (Michaelis–Menten–Henri) and a model planar system due to Davis and Skodje. © 2002 Published by Elsevier Science B.V.

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1. Introduction and summary of results

Many chemical reaction mechanisms in combustion [43,56,85], atmospheric science [72], enzyme kinetics [13], and biochemistry [28] involve large numbers of species, multiple chains of chemical reactions, and widely disparate

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time scales. A typical model of hydrocarbon combustion, for example, may well involve several hundred species, which participate in hundreds of reactions that proceed on time scales ranging from nanoseconds to minutes. The size and complexity of these mechanisms has stimulated the search for methods that reduce the number of species and chemical reactions but retain a desired degree of accuracy. Typically, these *reduction methods* select a small number of species, which are marked as *reaction progress variables*, and determine the concentrations of the remaining species as functions of the latter, either by table look-ups or by direct computation. The critical step in these methods is, of course, the definition of the reaction progress variables, which may be actual concentrations of selected species or combinations thereof.

Research into reduction methods has increased dramatically over the past decade, and several methods have been proposed in the literature and implemented in computer codes. We mention the quasi-steady-state approximation [71,81,90], the partial-equilibrium approximation [75], methods based on details of the chemistry [56,75], an iterative method [16,61], the method of intrinsic low-dimensional manifolds (ILDMs) [40,41], the computational singular perturbation method [19,20,29–32,36,44], a principal-component analysis [5,22,82], lumping techniques [35], repro-modeling [80], an inertial-manifold approach [89], a dynamic dimension-reduction method [9,10], a saddle-point method [7,8], a predictor-corrector method [7,8], an optimization method [57], and a global-eigenvalue method [74].

In this paper, we focus on two reduction methods, namely, the ILDM method due to Maas and Pope [40,41] and the iterative method due to Fraser and further developed by Roussel and Fraser [16,61]. Both methods have been developed for and extensively applied to problems with slow manifolds that attract nearby initial conditions. The long-time behavior of such systems is governed by the dynamics on the slow manifold, whose dimension is generally much less than that of the total composition space, resulting in a considerable reduction of complexity.

Given the importance of slow manifolds, a central question for any reduction method is: How accurately does it approximate a slow manifold? The present investigation answers this question for the ILDM method of Maas and Pope and the iterative method of Fraser and Roussel.

In the ILDM method, the Jacobian of the vector field is partitioned at each point of phase space into a fast and a slow component, and bases for the corresponding subspaces are generated by means of a Schur decomposition. The ILDM is defined as the locus of points where the vector field lies entirely in the slow subspace and is an approximation of the slow manifold. The efficacy of the ILDM method is evident, for example, by the reduction achieved in the prototypical example of a $CO–H_2–O_2–N_2$ combustion model [38,41]. Disregarding only the production of NO, the model comprises evolution equations for the enthalpy, pressure, and concentration of each of 13 species, making for a 15-dimensional phase space, and a total of 67 chemical reactions. With the proper choice of the reaction progress variable (CO_2), a reduction to a one-dimensional ILDM can be achieved that retains a certain accuracy after an initial transient [38,41]. Reduction to a two-dimensional ILDM gives a better approximation, albeit at the expense of keeping track of two reaction progress variables and the storage of a correspondingly larger look-up table. Refinements, applications, and evaluations of the ILDM method against direct numerical simulations can be found in Refs. [1–4,11,12,37,39,42,47,58,68–70,73,86–88].

The iterative method was inspired by the phase space geometry of an enzyme kinetics model involving a fast and a slow species, where the slow manifold is a curve in the phase plane. The method is derived formally from the *invariance equation*—an equation that is satisfied on any trajectory of the dynamical system and, in particular, on the slow manifold and extends naturally to multidimensional systems with (possibly) higher-dimensional slow manifolds. The procedure is explicit if the force field is linear in the fast variable, and implicit otherwise; hence, it generally requires the use of a nonlinear equation solver. The method has been developed further and applied, in particular, to several problems of enzyme kinetics and metabolism in Refs. [17,48,59,60,62–65].

A natural framework for the analysis of these and similar reduction methods is provided by geometric singular perturbation theory (GSPT) [15,24,27,66]. The presence of a fast and a slow time scale leads naturally to the

introduction of a small positive parameter ε measuring the ratio of the characteristic times. If, in the limit as $\varepsilon \downarrow 0$ (infinite separation of time scales), the system of kinetics equations has a slow manifold, $\mathcal{M}_0$, in phase space and this manifold is asymptotically stable, then GSPT identifies a (usually nonunique) slow manifold $\mathcal{M}_\varepsilon$ for ε sufficiently small positive and gives a complete geometric and analytical description of all solutions in the vicinity of the slow manifold, including how trajectories approach the manifold. By comparing the asymptotics of the slow manifold $\mathcal{M}_\varepsilon$ found by GSPT with the asymptotics of the low-dimensional manifolds generated by the ILDM method and the iterative method we can evaluate the accuracy of these reduction methods for small values of ε (finite but large separation of time scales). The evaluation leads to the following conclusions.

- ILDM method:
  - (i) The asymptotic expansion of the ILDM agrees with the asymptotic expansion of the slow manifold $\mathcal{M}_\varepsilon$ up to and including the $\mathcal{O}(\varepsilon)$ term, for all fast–slow systems. In general, however, the $\mathcal{O}(\varepsilon^2)$ terms differ.
  - (ii) The error at $\mathcal{O}(\varepsilon^2)$ is proportional to the local curvature of the slow manifold $\mathcal{M}_0$. It vanishes if and only if the curvature of $\mathcal{M}_0$ is zero everywhere. (The "if" part was observed previously in Ref. [41].)
- Iterative method:
  - (i) The iterative method, if started from $\mathcal{M}_0$, generates term by term the asymptotic expansion of the slow manifold $\mathcal{M}_\varepsilon$. In particular, ℓ applications of the iterative method generate an approximation to the slow manifold $\mathcal{M}_\varepsilon$ that is asymptotically correct up to and including the $\mathcal{O}(\varepsilon^\ell)$ term, albeit with extraneous terms at $\mathcal{O}(\varepsilon^{\ell+1})$.
  - (ii) The ℓth iteration leaves the terms at $\mathcal{O}(1)$ through $\mathcal{O}(\varepsilon^{\ell-1})$ invariant. (This observation is important because the lower-order terms have already been determined correctly in the preceding iterations.)

Remark. In Ref. [58], it is shown that the ILDM coincides with the slow manifold $\mathcal{M}_0$ in the limit of infinite separation of the fast and slow time scales ($\varepsilon = 0$).

Remark. In Ref. [7], it is shown that, for an elementary nonlinear example consisting of a planar linear system plus one quadratic term, the error in the ILDM increases with increasing curvature.

Remark. The slow manifold $\mathcal{M}_0$ can often be found analytically; otherwise, it can be obtained by one application of the iterative method to the coordinate axis, see Refs. [16,61].

The conclusions of this paper show that the ILDM method finds a highly accurate approximation of the slow manifold $\mathcal{M}_\varepsilon$, and this approximation may be sufficient in many applications. However, if an even higher order of accuracy is required, for example if the curvature correction is significant, then the conclusions listed above suggest that the desired further accuracy might be obtained by using an appropriate number of applications of the iterative method. In addition, there are important numerical issues, such as stability and computation time, that one needs to take into account when selecting a method for a given problem. Davis and Skodje [7] discuss a number of these issues in the context of a comparison between the ILDM method and three other methods. They find that higher accuracy can be obtained for some systems by applying either a predictor-corrector method or a modified Fraser scheme. Moreover, they often use the ILDM as the first approximation, or initialization, for these other schemes. Since issues associated with implementation are beyond the scope of the present paper, we refer to the literature cited above for a discussion of these important issues.

This paper is organized as follows. In Section 2, we review the general framework of fast–slow systems of ordinary differential equations (ODEs) and recall the asymptotic expansion of the slow manifold. In Section 3, we define the ILDM and indicate briefly how it is computed. We present the asymptotic expansion of the ILDM for planar fast–slow systems (one fast and one slow variable) in Section 4 and for general fast–slow systems (n fast and m

slow variables) in Section 5. The results are summarized in Corollary 5.1. In Section 6, we describe the iterative method of Fraser and Roussel. We discuss its asymptotics in Section 7. The results are summarized in Corollary 7.1. We illustrate the analytical results with two planar examples, namely the Michaelis–Menten–Henri (MMH) mechanism of enzyme kinetics (Section 8) and an example due to Davis and Skodje (Section 9). In Section 10 we remark on several generalizations and discuss some remaining issues.

2. Fast–slow systems of ODEs

We consider reaction mechanisms in homogeneous media, where the concentrations of the chemical species depend on time only. The concentrations evolve on two distinct and widely separated time scales. The slowly evolving concentrations are the entries of the vector y, the remaining concentrations the entries of the vector z; the former has m components, the latter n ($m, n \geq 1$). The separation of time scales is measured by ε, an arbitrarily small positive parameter. The limit $\varepsilon \downarrow 0$ corresponds to infinite separation. The reaction mechanism is thus modeled by a system of ODEs:

$$y' = \varepsilon f(y, z, \varepsilon), \tag{2.1}$$

$$z' = g(y, z, \varepsilon). \tag{2.2}$$

The unknowns y and z are functions of t with values in $\mathbf{R}^m$ and $\mathbf{R}^n$, respectively; $'$ denotes differentiation with respect to t; and f and g are smooth functions with values in $\mathbf{R}^m$ and $\mathbf{R}^n$, respectively. We assume that f and g, as well as all their derivatives, are $\mathcal{O}(1)$ as $\varepsilon \downarrow 0$.

Remark. The system of equations (2.1) and (2.2) is, of course, an idealization of the complex systems that occur in chemical kinetics. The model is adopted here because it is suitable for mathematical analysis. We claim, however, that it also captures the essential elements of any reaction mechanism whose long-term dynamics evolve on slow manifolds and offers a paradigm for the analysis of reduction methods. The validity of our conclusions extends therefore well beyond the idealized system of equations (2.1) and (2.2). We comment on the implications for more realistic systems in Section 10.

The independent variable t is called the *fast time* because it defines the time scale on which the fast variables evolve, and the system of equations (2.1) and (2.2) is labeled the *fast system*. While the fast time scale is appropriate for the study of the transient dynamics, the long-time dynamics are more naturally studied in terms of the *slow time* $\tau = \varepsilon t$. On the scale of τ, the system of equations (2.1) and (2.2) assumes the form

$$\dot{y} = f(y, z, \varepsilon), \tag{2.3}$$

$$\varepsilon \dot{z} = g(y, z, \varepsilon). \tag{2.4}$$

Here, $\dot{}$ denotes differentiation with respect to τ. We refer to the system of equations (2.3) and (2.4) as the *slow system*.

The fast system (2.1) and (2.2) and the slow system (2.3) and (2.4) are, of course, equivalent as long as $\varepsilon > 0$, but they approach different limits as $\varepsilon \downarrow 0$, that is, as the separation of the fast and slow time scales becomes infinite. The fast system reduces to

$$y' = 0, \tag{2.5}$$

$$z' = g(y, z, 0), \tag{2.6}$$

which is essentially a single equation for the fast variable z with y as a parameter. The slow system, on the other hand, reduces to

$$\dot{y} = f(y, z, 0), \tag{2.7}$$

$$0 = g(y, z, 0). \tag{2.8}$$

The first equation describes the motion of the slow variable y, and the second equation is an algebraic constraint that forces the motion to take place on the zero set of g.

Our focus is on systems for which the zero set of g is represented by the graph of a function. That is, we assume that there exists a single-valued function h_0, which is defined on a compact domain $K = [0, Y]^m$ in $\mathbf{R}^m$, such that

$$g(y, h_0(y), 0) = 0, \quad y \in K. \tag{2.9}$$

The zero set of g thus defines a *manifold*, $\mathcal{M}_0$, in phase space,

$$\mathcal{M}_0 = \{(y, z) \in \mathbf{R}^{m+n} : z = h_0(y), y \in K\}, \tag{2.10}$$

to which the motion of the reduced slow system is confined.

Our analysis requires a second assumption that holds for many, though not all, of the systems in which reductions have been sought, namely, that each point $(y, h_0(y))$ on $\mathcal{M}_0$ is an *asymptotically stable fixed point* of Eq. (2.6). The assumption guarantees that the eigenvalues of the matrix $D_z g(y, h_0(y), 0)$ all have negative real parts.

Remark. The two assumptions are justified in most enzyme kinetics and some combustion and atmospheric chemistry problems. In certain more complex reaction mechanisms, however, they may need justification. Toward this end, we observe that, in those cases where reduction methods are expected to be effective, h_0 can be found locally by the implicit function theorem (since the second assumption guarantees that the matrix $(D_z g)(y, h_0(y), 0)$ is invertible for each $y \in K$), and GSPT can be applied to each local portion. In the absence of singularities, these local functions can be pieced together to form a smooth global function over the entire domain under consideration.

Under the above conditions, standard asymptotic theory (see, for example, Refs. [6,15,24,34,45,49,79]) guarantees that, when ε is positive but arbitrarily small, there exists a *slow manifold* $\mathcal{M}_\varepsilon$ that is invariant under the dynamics of the system of equations (2.1) and (2.2), has the same dimension as $\mathcal{M}_0$, and lies near $\mathcal{M}_0$. All nearby solutions relax exponentially fast to $\mathcal{M}_\varepsilon$, and their long-term evolution is determined by an associated solution on the slow manifold itself. The manifold $\mathcal{M}_\varepsilon$ is usually not unique; typically there is a family of slow manifolds, all exponentially close ($\mathcal{O}(\mathrm{e}^{-c/\varepsilon})$ for some $c > 0$).

Theorem 2.1 (Fenichel, asymptotically stable slow manifolds). *For any sufficiently small ε, there is a function h_ε that is defined on K such that the graph*

$$\mathcal{M}_\varepsilon = \{(y, z) : z = h_\varepsilon(y), y \in K\} \tag{2.11}$$

is locally invariant under the dynamics of Eqs. (2.1) *and* (2.2). *The function h_ε admits an asymptotic expansion*

$$h_\varepsilon(y) = h_0(y) + \varepsilon h^{(1)}(y) + \varepsilon^2 h^{(2)}(y) + \cdots \quad \text{as } \varepsilon \downarrow 0, \tag{2.12}$$

where the coefficients $h^{(\ell)} : K \to \mathbf{R}^n$ are found successively from the equation

$$(D_z g)h^{(\ell)} = \sum_{i=0}^{\ell-1} (Dh^{(i)}) f^{(\ell-1-i)} - \sum_{j=2}^{\ell} \frac{1}{j!} (D_z^j g) \sum_{|i|=\ell} (h^{(i_1)}, \ldots, h^{(i_j)}) - \sum_{k=1}^{\ell-1} \frac{1}{k!} \sum_{j=1}^{\ell-k} \frac{1}{j!} (D_z^j (\partial_\varepsilon^k g)) \sum_{|i|=\ell-k} (h^{(i_1)}, \ldots, h^{(i_j)}) - \frac{1}{\ell!} (\partial_\varepsilon^\ell g) \tag{2.13}$$

for $\ell = 1, 2, \ldots$, with $h^{(0)} = h_0$. Here, the functions f and g and their derivatives are evaluated at $(y, z = h_0(y), 0)$, and it is understood that a sum is empty when the lower bound exceeds the upper bound. In particular, $h^{(1)}$ and $h^{(2)}$ are given by

$$(D_z g)h^{(1)} = (Dh_0) f - g_\varepsilon, \tag{2.14}$$

$$(D_z g)h^{(2)} = (Dh^{(1)}) f + (Dh_0)((D_z f)h^{(1)} + f_\varepsilon) - \tfrac{1}{2}(D_z^2 g)(h^{(1)}, h^{(1)}) - (D_z g_\varepsilon)h^{(1)} - \tfrac{1}{2} g_{\varepsilon\varepsilon}. \tag{2.15}$$

Furthermore, $h_\varepsilon \in C^r(K)$ for any finite r, and the dynamics of the system of equations (2.1) *and* (2.2) *on $\mathcal{M}_\varepsilon$ are given by the reduced equation*

$$\dot{y} = f(y, h_\varepsilon(y), \varepsilon). \tag{2.16}$$

Proof. The theorem is a direct restatement of [24, Theorem 2] for the special case in which $\mathcal{M}_0$ is asymptotically stable. It also follows directly from Nipp [49, Theorem] and is a special case of the Fenichel theory [15]. The asymptotics of the slow manifold $\mathcal{M}_\varepsilon$ are given explicitly, for example, in Refs. [45,50]. □

Remark. In many instances—for example, in the MMH reaction mechanism discussed in Section 8 and various combustion problems—the reduced slow system $\dot{y} = f(y, h_0(y), 0)$ has an asymptotically stable fixed point at $(y_0, h_0(y_0))$, say. In such cases, the reaction scheme has a global attracting equilibrium. Under the hypotheses made above, the system of equations (2.1) and (2.2) has a fixed point at $(y_{0,\varepsilon}, h_\varepsilon(y_{0,\varepsilon}))$, and the slow manifold $\mathcal{M}_\varepsilon$ is its weak stable manifold.

Remark. While we have used it here only for the case of attracting manifolds, the Fenichel theorem and Theorem 2 in Ref. [24] hold for the more general case of fast–slow systems of ODEs for which the manifold $\mathcal{M}_0$ is normally hyperbolic—that is, where there can be both fast stable (exponentially contracting) and fast unstable (exponentially expanding) dynamics in the directions transverse to $\mathcal{M}_0$. In the more general case, the matrix $(D_z g)(y, h_0(y), 0)$ has s eigenvalues with a negative real part and u eigenvalues with a positive real part, the fast variable z decomposes into a u-dimensional and an s-dimensional component with $u + s = n$, and the dynamics of all solutions near $\mathcal{M}_\varepsilon$ are governed by the Fenichel normal form [25]. The asymptotics of $\mathcal{M}_\varepsilon$ remains unchanged.

Remark. The papers of Tikhonov [79] and Levin and Levinson [33,34] present the original theory of persistence of asymptotically stable manifolds (see also Ref. [53]). The theory of persistence of normally hyperbolic manifolds can be found in the monographs of Fenichel [14,15] and Hirsch et al. [23]; see also Ref. [84]. Other relevant references are [45,49] for singularly perturbed systems of ODEs with asymptotically stable slow manifolds and [24,66] for singularly perturbed systems of ODEs with general normally hyperbolic slow manifolds. An introductory exposition of GSPT is given in Ref. [27].

Remark. The Fenichel theory [15] has been used in the important work of Stiefenhofer [78] to study the quasi-steady-state approximation in the context of fast–slow systems of ODEs modeling reaction kinetics. The main results

of Ref. [78] are: (i) the identification of the essential assumptions underlying the quasi-steady-state approximation of Schauer and Heinrich [67], (ii) a clear geometric interpretation of the quasi-steady-state approximation in terms of the leading order asymptotics of the slow manifold $\mathcal{M}_\varepsilon$, and (iii) a geometric interpretation of how the slow system enslaves the fast system.

Remark. A numerical procedure for finding asymptotically stable slow manifolds in fast–slow systems, which is stable and highly accurate for small values of ε, has been given by Nipp [50].

3. The ILDM method of Maas and Pope

The ILDM method starts from the slow system, Eqs. (2.3) and (2.4), takes the local vector field F and the associated Jacobian J, and reduces the latter at each point to a fast and a slow component. The vector field F and its Jacobian J are

$$F = \begin{pmatrix} f \\ \varepsilon^{-1} g \end{pmatrix}, \qquad J = \begin{pmatrix} D_y f & D_z f \\ \varepsilon^{-1} D_y g & \varepsilon^{-1} D_z g \end{pmatrix}, \tag{3.1}$$

where $D_y f$ is the $m \times m$ matrix of partial derivatives $\partial f_i / \partial y_j$, $D_z f$ the $m \times n$ matrix of partial derivatives $\partial f_i / \partial z_j$, $D_y g$ the $n \times m$ matrix of partial derivatives $\partial g_i / \partial y_j$, and $D_z g$ the $n \times n$ matrix of partial derivatives $\partial g_i / \partial z_j$.

By assumption, the real part of each eigenvalue of J is negative. The sum of the eigenvalues is equal to the trace of J, which is $\mathcal{O}(\varepsilon^{-1})$ as $\varepsilon \downarrow 0$, and their product is equal to the determinant of J, which is $\mathcal{O}(\varepsilon^{-n})$ as $\varepsilon \downarrow 0$. The eigenvalues of J fall therefore into two groups: one group of m eigenvalues with $\mathcal{O}(1)$ negative real parts and another group of n eigenvalues with $\mathcal{O}(\varepsilon^{-1})$ negative real parts. The eigenvectors associated with the first group span the *slow subspace*, those associated with the second group the *fast subspace*. The Maas and Pope algorithm defines the ILDM as the locus of all points (y, z) where the vector field F lies entirely in the slow subspace.

The algorithm uses a Schur decomposition [76, Section 6.3] of J,

$$J = QNQ' \tag{3.2}$$

with Q unitary ($QQ' = Q'Q = I_{m+n}$, $'$ denoting the transpose) and N upper triangular,

$$Q = (Q_s\, Q_f), \qquad N = \begin{pmatrix} N_s & N_{sf} \\ 0 & N_f \end{pmatrix}. \tag{3.3}$$

The dimensions of Q_s and Q_f are $(m+n) \times m$ and $(m+n) \times n$, respectively; N_s is an $m \times m$ upper triangular matrix, N_f an $n \times n$ upper triangular matrix, and N_{sf} an $m \times n$ full matrix. The eigenvalues of J appear on the diagonal of N in descending order of their real parts, from least negative at the $(1, 1)$ position to most negative at the $(m+n, m+n)$ position. This particular ordering is accomplished in Ref. [41] by means of a modification of Stewart's implementation of the Schur algorithm [77] and in Ref. [38] by means of a standard Schur decomposition followed by a sequence of Givens rotations [18, Section 5.1].

The first m Schur vectors—that is, the columns of Q_s—form an orthogonal basis for the slow subspace, while the remaining n Schur vectors—the columns of Q_f—form an orthogonal basis for the orthogonal complement of the slow subspace. The vector field F is entirely in the slow subspace if it is orthogonal to the orthogonal complement of the slow subspace, that is, if

$$Q_f' F = 0. \tag{3.4}$$

This equation defines the ILDM, the latter being an approximation of the slow manifold $\mathcal{M}_\varepsilon$. We analyze its asymptotics (as $\varepsilon \downarrow 0$) in the following sections.

Remark. The matrix Q'_f corresponds to Q_L^T, the number n to n_f, and the sum $m+n$ to n in Ref. [41].

In the numerical implementation of the ILDM method for general, closed, adiabatic, and isobaric reaction mechanisms, the system of equations is closed by supplementing the ILDM equation, Eq. (3.4), by a set of parameter equations. The parameter equations fix the enthalpy, pressure, and element composition. In addition, the reaction progress variables are treated as parameters. Each fixed set of parameters yields one point of the ILDM, and the entire ILDM is obtained by sweeping over the admissible set of parameter values. As noted in Ref. [41], the parameters can generally be chosen so the ILDM is at least defined piecewise, and, most important, the choice of the parameter equations does not influence the construction of the manifold.

In Eqs. (2.1) and (2.2), the enthalpy, pressure, and conserved quantities have been neglected. In this case, the parameter equations fix the values of the slow variables y, and the ILDM is obtained by sweeping over all points $y \in K$.

4. Asymptotics of the ILDM—planar case

We first restrict our attention to planar fast–slow systems, Eqs. (2.3) and (2.4) with $m = n = 1$, for which the computations are relatively straightforward and the asymptotic analysis more transparent. We address the general case in Section 5.

In the planar case, the vector field F and its Jacobian J are

$$F = \begin{pmatrix} f \\ \varepsilon^{-1} g \end{pmatrix}, \qquad J = \begin{pmatrix} f_y & f_z \\ \varepsilon^{-1} g_y & \varepsilon^{-1} g_z \end{pmatrix}. \tag{4.1}$$

The eigenvalues of J are

$$\lambda_{s,f} = \frac{1}{2}(\varepsilon^{-1} g_z + f_y) \pm \sqrt{\frac{1}{4}(\varepsilon^{-1} g_z + f_y)^2 - \varepsilon^{-1}(f_y g_z - f_z g_y)}, \tag{4.2}$$

where the upper (lower) sign is associated with λ_s (λ_f). Thus,

$$\lambda_s = f_y - \frac{f_z g_y}{g_z} + \mathcal{O}(\varepsilon), \qquad \lambda_f = \varepsilon^{-1} g_z + \mathcal{O}(1) \quad \text{as } \varepsilon \downarrow 0. \tag{4.3}$$

The derivatives of f and g, which are evaluated at (y, z, ε), are all $\mathcal{O}(1)$ as $\varepsilon \downarrow 0$. The (nonnormalized) slow eigenvector is

$$v_s = \begin{pmatrix} \lambda_s - \varepsilon^{-1} g_z \\ \varepsilon^{-1} g_y \end{pmatrix}, \tag{4.4}$$

and there is a corresponding fast eigenvector v_f. The vector v_s spans the slow subspace, v_f the fast subspace. The vectors v_s and v_f are not necessarily orthogonal. To determine the points (y, z) in the phase plane where the vector field F lies entirely in the slow subspace, we work with the orthogonal complement of the slow subspace, which is spanned by the row vector

$$v_s^\perp = (\varepsilon^{-1} g_y, \varepsilon^{-1} g_z - \lambda_s). \tag{4.5}$$

The locus of all points in the phase plane where the vector field F is in the slow subspace coincides with the set of all points (y, z) where F is orthogonal to $v_s^\perp$, that is, where

$$fg_y + g(\varepsilon^{-1}g_z - \lambda_s) = 0. \tag{4.6}$$

This equation defines the ILDM.

Remark. A Schur decomposition of the matrix J gives the vectors $q_f = v_f/|v_f|$ and $q_f^\perp = v_f^\perp/|v_f|$ directly. The algorithm must be modified to find the vectors $q_s = v_s/|v_s|$ and $q_s^\perp = v_s^\perp/|v_s|$, as described in Section 3.

Theorem 4.1 (Planar case). *The equation for the ILDM,* Eq. (4.6), *admits an asymptotic solution in the form of a power series expansion,*

$$z = \psi(y, \varepsilon) = \psi^{(0)}(y) + \varepsilon\psi^{(1)}(y) + \varepsilon^2\psi^{(2)}(y) + \cdots \quad \text{as } \varepsilon \downarrow 0. \tag{4.7}$$

The functions $\psi^{(0)}$, $\psi^{(1)}$, and $\psi^{(2)}$ are defined by the equations

$$\psi^{(0)} = h_0, \tag{4.8}$$

$$g_z\psi^{(1)} = fh_0' - g_\varepsilon, \tag{4.9}$$

$$g_z\psi^{(2)} = f\psi^{(1)'} - \frac{f^2}{g_z}h_0'' + (f_z\psi^{(1)} + f_\varepsilon)h_0' - \frac{1}{2}g_{zz}(\psi^{(1)})^2 - g_{z\varepsilon}\psi^{(1)} - \frac{1}{2}g_{\varepsilon\varepsilon}. \tag{4.10}$$

Here, $h_0 \equiv h_0(y)$ is defined by the equation $g(y, h_0(y), 0) = 0$, $'$ denotes differentiation with respect to y, and the functions f and g and their derivatives are evaluated at $(y, h_0(y), 0)$.

Proof. Assume that $z = \psi(y, \varepsilon)$, where ψ is given by the power series expansion (4.7). Then

$$f(y, \psi(y, \varepsilon), \varepsilon) = f + \varepsilon(f_z\psi^{(1)} + f_\varepsilon) + \varepsilon^2(f_z\psi^{(2)} + \tfrac{1}{2}f_{zz}(\psi^{(1)})^2 + f_{z\varepsilon}\psi^{(1)} + \tfrac{1}{2}f_{\varepsilon\varepsilon}) + \cdots, \tag{4.11}$$

where in the right member f and its derivatives are evaluated at $(y, \psi^{(0)}(y), 0)$. Similar expansions hold for g and the derivatives of f and g. The leading term in the expansion of λ_s follows immediately from Eq. (4.3):

$$\lambda_s = \lambda_s^{(0)} + \mathcal{O}(\varepsilon), \quad \lambda_s^{(0)} = f_y - \frac{f_z g_y}{g_z}, \tag{4.12}$$

where the derivatives of f and g are similarly evaluated at $(y, \psi^{(0)}(y), 0)$. We substitute the various expansions into Eq. (4.6) and equate the coefficients of like powers of ε.

$\mathcal{O}(\varepsilon^{-1})$. The ILDM equation, Eq. (4.6), gives

$$(gg_z)(y, z = \psi^{(0)}(y), 0) = 0, \tag{4.13}$$

which is satisfied if $\psi^{(0)} = h_0$. This result confirms Eq. (4.8).

$\mathcal{O}(1)$. From the equation for the ILDM, Eq. (4.6), we obtain

$$fg_y + (g_z\psi^{(1)} + g_\varepsilon)g_z = 0. \tag{4.14}$$

Here, we have used the identity $g \equiv g(y, h_0(y), 0) = 0$. The same identity implies that

$$g_y + g_z h_0' = 0, \tag{4.15}$$

so Eq. (4.14) reduces to

$$(g_z\psi^{(1)} + g_\varepsilon - fh_0')g_z = 0. \tag{4.16}$$

The assumption of attractive manifolds implies that $g_z < 0$, so Eq. (4.9) follows.

$\mathcal{O}(\varepsilon)$. From the equation for the ILDM, Eq. (4.6), we obtain

$$f(g_{yz}\psi^{(1)} + g_{y\varepsilon} + (g_{zz}\psi^{(1)} + g_{z\varepsilon} - \lambda_s^{(0)})h_0') - (f_z\psi^{(1)} + f_\varepsilon)g_z h_0' + (g_z\psi^{(2)} + \tfrac{1}{2}g_{zz}(\psi^{(1)})^2 + g_{z\varepsilon}\psi^{(1)} + \tfrac{1}{2}g_{\varepsilon\varepsilon})g_z = 0. \tag{4.17}$$

Here, we have used Eqs. (4.8) and (4.9) and the identity (4.15). The same identity also results in a simplification of the expression (4.12) for $\lambda_s^{(0)}$,

$$\lambda_s^{(0)} = f_y + f_z h_0'. \tag{4.18}$$

Furthermore, differentiating Eq. (4.16) with respect to y, we find

$$g_{yz}\psi^{(1)} + g_{y\varepsilon} + (g_{zz}\psi^{(1)} + g_{z\varepsilon})h_0' + g_z\psi^{(1)'} = fh_0'' + (f_y + f_z h_0')h_0'. \tag{4.19}$$

With Eqs. (4.18) and (4.19), Eq. (4.17) simplifies to

$$\left(g_z\psi^{(2)} + \tfrac{1}{2}g_{zz}(\psi^{(1)})^2 + g_{z\varepsilon}\psi^{(1)} + \tfrac{1}{2}g_{\varepsilon\varepsilon} - f\psi^{(1)'} + \left(\frac{f^2}{g_z}\right)h_0'' - (f_z\psi^{(1)} + f_\varepsilon)h_0'\right)g_z = 0. \tag{4.20}$$

Since $g_z < 0$, Eq. (4.10) follows. □

In the following section, we will generalize Theorem 4.1 to the multidimensional case (Theorem 5.1) and compare the asymptotics of the ILDM with the asymptotics of the slow manifold $\mathcal{M}_\varepsilon$ (Corollary 5.1).

5. Asymptotics of the ILDM—general case

The definition of the ILDM, Eq. (3.4), is based on a partition of the Jacobian, Eq. (3.2), into a fast and a slow component at each point of phase space and a Schur decomposition to generate bases for the corresponding fast and slow subspaces. Practical implementations of the Schur decomposition rely typically on the method of deflation [18, Chapter 7]; hence, the eigenvalues are generated in the order of descending *absolute values* of their real parts. This procedure yields a unitary matrix of the form $Q = (\,Q_f \quad Q_s\,)$. The columns of Q are then reordered, for example by a sequence of Givens rotations, as in Ref. [38].

Although this procedure is practical for numerical computations, it is not amenable to analysis. We start therefore from the standard Schur decomposition *before* reordering,

$$J = QTQ', \tag{5.1}$$

where

$$T = \begin{pmatrix} \Lambda_f & \Lambda \\ 0 & \Lambda_s \end{pmatrix} \tag{5.2}$$

with Λ_f an $n \times n$ upper triangular matrix, Λ_s an $m \times m$ upper triangular matrix, and Λ an $n \times m$ full matrix. The diagonal elements of Λ_f are the $\mathcal{O}(\varepsilon^{-1})$ eigenvalues of J, and the diagonal elements of Λ_s are the $\mathcal{O}(1)$ eigenvalues of J. The structure of the unitary matrix Q is

$$Q = \begin{pmatrix} Q_{11} & Q_{12} \\ Q_{21} & Q_{22} \end{pmatrix}, \tag{5.3}$$

where Q_{11} is an $m \times n$ matrix, Q_{12} an $m \times m$ matrix, Q_{21} an $n \times n$ matrix, and Q_{22} an $n \times m$ matrix. The columns of

$$\begin{pmatrix} Q_{11} \\ Q_{21} \end{pmatrix} \quad \text{and} \quad \begin{pmatrix} Q_{12} \\ Q_{22} \end{pmatrix}$$

form an orthogonal basis of the fast subspace and its orthogonal complement, respectively.

Since the fast and slow subspaces are not necessarily mutually orthogonal, the orthogonal complement of the fast subspace does not necessarily coincide with the slow subspace, and a further operation is needed to identify a basis for the slow subspace. This operation consists of solving the Sylvester equation

$$\Lambda_f X - X\Lambda_s = -\Lambda \tag{5.4}$$

for the $n \times m$ matrix X. With the definition

$$Y = \begin{pmatrix} I_n & X \\ 0 & I_m \end{pmatrix}, \tag{5.5}$$

we obtain a *block diagonalization* of J,

$$J = (QY)T_d(QY)^{-1}, \tag{5.6}$$

where

$$T_d = \begin{pmatrix} \Lambda_f & 0 \\ 0 & \Lambda_s \end{pmatrix}, \tag{5.7}$$

$$QY = \begin{pmatrix} Q_{11} & Q_{11}X + Q_{12} \\ Q_{21} & Q_{21}X + Q_{22} \end{pmatrix}, \qquad (QY)^{-1} = \begin{pmatrix} Q'_{11} - XQ'_{12} & Q'_{21} - XQ'_{22} \\ Q'_{12} & Q'_{22} \end{pmatrix}. \tag{5.8}$$

Thus, QY reduces the matrix J to its fast and slow components, and the condition that the vector field F given in Eq. (3.1) must lie entirely in the slow subspace is satisfied if

$$(Q'_{11} - XQ'_{12})f + \varepsilon^{-1}(Q'_{21} - XQ'_{22})g = 0. \tag{5.9}$$

The ILDM obtained from Eq. (5.9) is the same as the ILDM obtained from Eq. (3.4) and also the same as the ILDMs obtained in Refs. [38,41].

Theorem 5.1 (General case). *The equation for the ILDM*, Eq. (3.4), *admits an asymptotic solution in the form of a power series expansion,*

$$z = \psi(y, \varepsilon) = \psi^{(0)}(y) + \varepsilon\psi^{(1)}(y) + \varepsilon^2\psi^{(2)}(y) + \cdots \quad \text{as } \varepsilon \downarrow 0. \tag{5.10}$$

The $\mathbf{R}^n$*-valued functions* $\psi^{(0)}$, $\psi^{(1)}$, *and* $\psi^{(2)}$ *are defined by the equations*

$$\psi^{(0)} = h_0, \tag{5.11}$$

$$(D_z g)\psi^{(1)} = (Dh_0)f - g_\varepsilon, \tag{5.12}$$

$$\begin{aligned}(D_z g)\psi^{(2)} = {} & (D\psi^{(1)})f - (D_z g)^{-1}(D^2 h_0)(f, f) + (Dh_0)((D_z f)\psi^{(1)} + f_\varepsilon) - \tfrac{1}{2}(D_z^2 g)(\psi^{(1)}, \psi^{(1)}) \\ & - (D_z g_\varepsilon)\psi^{(1)} - \tfrac{1}{2}g_{\varepsilon\varepsilon}.\end{aligned} \tag{5.13}$$

Here, $h_0 \equiv h_0(y)$ is the $\mathbf{R}^n$-valued function defined by Eq. (2.9), *$g(y, h_0(y), 0) = 0$; $Dh_0 \equiv (Dh_0)(y)$ is a linear operator from $\mathbf{R}^m$ to $\mathbf{R}^n$, which is represented by the $n \times m$ matrix of partial derivatives $\partial h_{0,i}/\partial y_j$, and $D^2h_0 = D(Dh_0) \equiv (D^2h_0)(y)$ is a bilinear map from $\mathbf{R}^m \times \mathbf{R}^m$ to $\mathbf{R}^n$, $(D^2h_0)(u, v) = ((D^2h_0)u)v$ for all $u, v \in \mathbf{R}^m$. The functions f and g and their derivatives are evaluated at $(y, h_0(y), 0)$; $D_z f \equiv D_z f(y, h_0(y), 0)$ is a linear operator from $\mathbf{R}^n$ to $\mathbf{R}^m$, $D_z g \equiv D_z g(y, h_0(y), 0)$ a linear operator from $\mathbf{R}^n$ to $\mathbf{R}^n$, and $D_z^2 g = D_z(D_z g) \equiv D_z^2 g(y, h_0(y), 0)$ a bilinear map from $\mathbf{R}^n \times \mathbf{R}^n$ to $\mathbf{R}^n$.*

Proof. Assume that $z = \psi(y, \varepsilon)$ and that ψ is given by the expansion (5.10). For the asymptotic analysis of Eq. (5.9), we take

$$Q \equiv Q(\varepsilon) = \begin{pmatrix} 0 & I_m \\ Q_{21}^{(0)} & 0 \end{pmatrix} + \varepsilon \begin{pmatrix} Q_{11}^{(1)} & 0 \\ 0 & -Q_{21}^{(0)} Q_{11}^{(1)'} \end{pmatrix} + \cdots \tag{5.14}$$

with $Q_{21}^{(0)}$ a unitary $n \times n$ matrix and $Q_{11}^{(1)}$ an $m \times n$ matrix to be determined. Thus, Q is unitary to $\mathcal{O}(\varepsilon)$. Higher-order terms can be found in a consistent manner so $Q(\varepsilon)$ is unitary to any desired order. We take, furthermore,

$$\Lambda_f \equiv \Lambda_f(\varepsilon) = \varepsilon^{-1}\Lambda_f^{(-1)} + \Lambda_f^{(0)} + \cdots, \tag{5.15}$$

$$\Lambda \equiv \Lambda(\varepsilon) = \varepsilon^{-1}\Lambda^{(-1)} + \Lambda^{(0)} + \cdots, \tag{5.16}$$

$$\Lambda_s \equiv \Lambda_s(\varepsilon) = \Lambda_s^{(0)} + \cdots, \tag{5.17}$$

$$X \equiv X(\varepsilon) = X^{(0)} + \varepsilon X^{(1)} + \cdots. \tag{5.18}$$

The generalization of the expansion (4.11) to the present case is

$$\begin{aligned} f(y, \psi(y, \varepsilon), \varepsilon) = f &+ \varepsilon((D_z f)\psi^{(1)} + f_\varepsilon) \\ &+ \varepsilon^2((D_z f)\psi^{(2)} + \tfrac{1}{2}(D_z^2 f)(\psi^{(1)}, \psi^{(1)}) + (D_z f_\varepsilon)\psi^{(1)} + \tfrac{1}{2} f_{\varepsilon\varepsilon}) + \cdots. \end{aligned} \tag{5.19}$$

In the right member, f and its derivatives are evaluated at $(y, \psi^{(0)}(y), 0)$. Similar expansions hold for g and the derivatives of f and g.

To prove the theorem, we substitute the various expansions into Eq. (5.9) and equate the coefficients of like powers in ε in the usual manner.

$\mathcal{O}(\varepsilon^{-1})$. The ILDM equation, Eq. (5.9), gives

$$Q_{21}^{(0)'} g = 0. \tag{5.20}$$

Since $Q_{21}^{(0)}$ is unitary, Eq. (5.20) reduces to

$$g(y, \psi^{(0)}(y), 0) = 0. \tag{5.21}$$

This equation is satisfied if $\psi^{(0)} = h_0$, which confirms Eq. (5.11).

$\mathcal{O}(1)$. From the ILDM equation, Eq. (5.9), we obtain

$$-X^{(0)} f + Q_{21}^{(0)'}((D_z g)\psi^{(1)} + g_\varepsilon) = 0. \tag{5.22}$$

Here, we have already used the identity $g = 0$.

The matrix $X^{(0)}$ is determined from the $\mathcal{O}(\varepsilon^{-1})$ terms in the Sylvester equation, Eq. (5.4),

$$\Lambda_f^{(-1)} X^{(0)} = -\Lambda^{(-1)}. \tag{5.23}$$

The matrices $\Lambda_f^{(-1)}$ and $\Lambda^{(-1)}$, in turn, follow from the $\mathcal{O}(\varepsilon^{-1})$ terms in the Schur decomposition, Eq. (5.1),

$$Q_{21}^{(0)}\Lambda^{(-1)} = D_y g, \qquad Q_{21}^{(0)}\Lambda_f^{(-1)}Q_{21}^{(0)'} = D_z g. \tag{5.24}$$

The second equation is the Schur decomposition of $D_z g$, so $Q_{21}^{(0)}$ is determined by the ordering of the elements of $\Lambda_f^{(-1)}$. Both equations can be inverted,

$$\Lambda^{(-1)} = Q_{21}^{(0)'}(D_y g), \qquad \Lambda_f^{(-1)} = Q_{21}^{(0)'}(D_z g)Q_{21}^{(0)}. \tag{5.25}$$

Hence,

$$X^{(0)} = -Q_{21}^{(0)'}(D_z g)^{-1}(D_y g). \tag{5.26}$$

We can simplify this expression if we use the identity $g(y, h_0(y), 0) = 0$, which holds for all y. Upon differentiation, the identity gives a relation between $D_y g$ and $D_z g$,

$$D_y g + (D_z g)(Dh_0) = 0. \tag{5.27}$$

Note that this is a relation in the space of linear operators from $\mathbf{R}^m$ to $\mathbf{R}^n$. With this identity, Eq. (5.26) becomes

$$X^{(0)} = Q_{21}^{(0)'}(Dh_0), \tag{5.28}$$

and Eq. (5.22) reduces to

$$Q_{21}^{(0)'}[(D_z g)\psi^{(1)} + g_\varepsilon - (Dh_0)f] = 0. \tag{5.29}$$

Since $Q_{21}^{(0)}$ is unitary, Eq. (5.12) follows.

$\mathcal{O}(\varepsilon)$. From the ILDM equation, Eq. (5.9), we obtain

$$\begin{aligned}&(Q_{11}^{(1)'} - X^{(1)} + Q_{21}^{(0)'}(Dh_0)Q_{11}^{(1)}Q_{21}^{(0)'}(Dh_0))f - Q_{21}^{(0)'}(Dh_0)((D_z f)\psi^{(1)} + f_\varepsilon)\\ &\quad + Q_{21}^{(0)'}((D_z g)\psi^{(2)} + \tfrac{1}{2}(D_z^2 g)(\psi^{(1)}, \psi^{(1)}) + (D_z g_\varepsilon)\psi^{(1)} + \tfrac{1}{2}g_{\varepsilon\varepsilon}) = 0.\end{aligned} \tag{5.30}$$

Here, we have already made use of Eqs. (5.11) and (5.12) and substituted the expression (5.28) for $X^{(0)}$.

The matrix $X^{(1)}$ is determined from the $\mathcal{O}(1)$ terms in the Sylvester equation, Eq. (5.4),

$$\Lambda_f^{(-1)}X^{(1)} + \Lambda_f^{(0)}X^{(0)} - X^{(0)}\Lambda_s^{(0)} = -\Lambda^{(0)}. \tag{5.31}$$

The matrices $\Lambda_f^{(0)}$, $\Lambda_s^{(0)}$, and $\Lambda^{(0)}$ follow in turn from the $\mathcal{O}(1)$ terms in the Schur decomposition, Eq. (5.1),

$$Q_{11}^{(1)}\Lambda^{(-1)} + \Lambda_s^{(0)} = D_y f, \tag{5.32}$$

$$Q_{11}^{(1)}\Lambda_f^{(-1)}Q_{21}^{(0)'} = D_z f, \tag{5.33}$$

$$Q_{21}^{(0)}(\Lambda_f^{(-1)}Q_{11}^{(1)'} + \Lambda^{(0)}) = (D_z(D_y g))\psi^{(1)} + D_y g_\varepsilon, \tag{5.34}$$

$$Q_{21}^{(0)}(-\Lambda^{(-1)}Q_{11}^{(1)}Q_{21}^{(0)'} + \Lambda_f^{(0)}Q_{21}^{(0)'}) = (D_z^2 g)\psi^{(1)} + D_z g_\varepsilon. \tag{5.35}$$

We proceed as follows. First, we solve Eq. (5.33) for $Q_{11}^{(1)}$,

$$Q_{11}^{(1)} = (D_z f)Q_{21}^{(0)}(\Lambda_f^{(-1)})^{-1} = (D_z f)(D_z g)^{-1}Q_{21}^{(0)}. \tag{5.36}$$

Then, we obtain $\Lambda_s^{(0)}$ from Eq. (5.23),

$$\Lambda_s^{(0)} = D_y f - Q_{11}^{(1)} \Lambda^{(-1)} = D_y f + (D_z f)(Dh_0). \tag{5.37}$$

(We have used the relation (5.27) to rewrite the expression (5.25) for $\Lambda^{(-1)}$.) Next, we solve Eqs. (5.34) and (5.35) for $\Lambda^{(0)}$ and $\Lambda_f^{(0)}$,

$$\begin{aligned}\Lambda^{(0)} &= Q_{21}^{(0)'}((D_z(D_y g))\psi^{(1)} + D_y g_\varepsilon - Q_{21}^{(0)} \Lambda_f^{(-1)} Q_{11}^{(1)'}) \\ &= Q_{21}^{(0)'}((D_z(D_y g))\psi^{(1)} + D_y g_\varepsilon - (D_z g)((D_z f)(D_z g)^{-1})'),\end{aligned} \tag{5.38}$$

$$\begin{aligned}\Lambda_f^{(0)} &= Q_{21}^{(0)'}((D_z^2 g)\psi^{(1)} + D_z g_\varepsilon + Q_{21}^{(0)} \Lambda^{(-1)} Q_{11}^{(1)} Q_{21}^{(0)'}) Q_{21}^{(0)} \\ &= Q_{21}^{(0)'}((D_z^2 g)\psi^{(1)} + D_z g_\varepsilon - (D_z g)(Dh_0)(D_z f)(D_z g)^{-1}) Q_{21}^{(0)}.\end{aligned} \tag{5.39}$$

After these steps, we find $X^{(1)}$ from Eq. (5.31),

$$\begin{aligned}X^{(1)} &= (\Lambda_f^{(-1)})^{-1}(-\Lambda^{(0)} - \Lambda_f^{(0)} X^{(0)} + X^{(0)} \Lambda_s^{(0)}) \\ &= Q_{21}^{(0)'}(D_z g)^{-1}[-((D_z(D_y g))\psi^{(1)} + D_y g_\varepsilon) + (D_z g)((D_z f)(D_z g)^{-1})' \\ &\quad - ((D_z^2 g)\psi^{(1)} + D_z g_\varepsilon - (D_z g)(Dh_0)(D_z f)(D_z g)^{-1})(Dh_0) + (Dh_0)(D_y f + (D_z f)(Dh_0))].\end{aligned} \tag{5.40}$$

Substituting $Q_{11}^{(1)}$ from Eq. (5.36) and $X^{(1)}$ from Eq. (5.40) into Eq. (5.30), we obtain

$$\begin{aligned}&Q_{21}^{(0)'}[(D_z g)^{-1}((D_z(D_y g))(\psi^{(1)}, f) + (D_y g_\varepsilon) f + (D_z^2 g)(\psi^{(1)}, (Dh_0) f) + (D_z g_\varepsilon)((Dh_0) f) \\ &\quad - (Dh_0)(D_y f + (D_z f)(Dh_0)) f) - (Dh_0)((D_z f)\psi^{(1)} + f_\varepsilon) + (D_z g)\psi^{(2)} + \tfrac{1}{2}(D_z^2 g)(\psi^{(1)}, \psi^{(1)}) \\ &\quad + (D_z g_\varepsilon)\psi^{(1)} + \tfrac{1}{2} g_{\varepsilon\varepsilon}] = 0.\end{aligned} \tag{5.41}$$

The bilinear maps $D_z(D_y g)$ and $D_z^2 g$ satisfy the symmetry relations

$$(D_z(D_y g))(u, v) = (D_y(D_z g))(v, u), \quad u \in \mathbf{R}^n, \quad v \in \mathbf{R}^m, \tag{5.42}$$

$$(D_z^2 g)(u, v) = (D_z^2 g)(v, u), \quad u, v \in \mathbf{R}^n, \tag{5.43}$$

so Eq. (5.41) is equivalent with

$$\begin{aligned}&Q_{21}^{(0)'}[(D_z g)^{-1}((D_y(D_z g))(f, \psi^{(1)}) + (D_y g_\varepsilon) f + (D_z^2 g)((Dh_0) f, \psi^{(1)}) + (D_z g_\varepsilon)((Dh_0) f) \\ &\quad - (Dh_0)(D_y f + (D_z f)(Dh_0)) f) - (Dh_0)((D_z f)\psi^{(1)} + f_\varepsilon) + (D_z g)\psi^{(2)} + \tfrac{1}{2}(D_z^2 g)(\psi^{(1)}, \psi^{(1)}) \\ &\quad + (D_z g_\varepsilon)\psi^{(1)} + \tfrac{1}{2} g_{\varepsilon\varepsilon}] = 0.\end{aligned} \tag{5.44}$$

We simplify this expression by means of Eq. (5.12). Upon differentiation, this equation gives the identity

$$\begin{aligned}&(D_y(D_z g))\psi^{(1)} + D_y g_\varepsilon + (D_z^2 g)(Dh_0)\psi^{(1)} + (D_z g_\varepsilon)(Dh_0) + (D_z g)(D\psi^{(1)}) \\ &\quad = (D^2 h_0) f + (Dh_0)(D_y f + (D_z f)(Dh_0)).\end{aligned} \tag{5.45}$$

This is a relation in the space of linear operators from $\mathbf{R}^m$ to $\mathbf{R}^n$. When applied to the vector f, it gives the identity

$$\begin{aligned}&(D_y(D_z g))(f, \psi^{(1)}) + (D_y g_\varepsilon) f + (D_z^2 g)((Dh_0) f, \psi^{(1)}) + (D_z g_\varepsilon)((Dh_0) f) + (D_z g)((D\psi^{(1)}) f) \\ &\quad = (D^2 h_0)(f, f) + (Dh_0)(D_y f + (D_z f)(Dh_0)) f.\end{aligned} \tag{5.46}$$

With this result, Eq. (5.44) simplifies to

$$Q_{21}^{(0)'}[(D_z g)\psi^{(2)} + \tfrac{1}{2}(D_z^2 g)(\psi^{(1)}, \psi^{(1)}) + (D_z g_\varepsilon)\psi^{(1)} + \tfrac{1}{2} g_{\varepsilon\varepsilon} - (D\psi^{(1)})f + (D_z g)^{-1}(D^2 h_0)(f, f) - (Dh_0)((D_z f)\psi^{(1)} + f_\varepsilon)] = 0. \tag{5.47}$$

Since $Q_{21}^{(0)}$ is unitary, Eq. (5.13) follows. □

The following corollary summarizes the result of the asymptotic analysis.

Corollary 5.1. *The ILDM is an approximation to the slow manifold $\mathcal{M}_\varepsilon$ of the fast–slow system of* equations (2.1) and (2.2), *which is asymptotically accurate up to and including the $\mathcal{O}(\varepsilon)$ term as $\varepsilon \downarrow 0$. The approximation is asymptotically accurate up to and including the $\mathcal{O}(\varepsilon^2)$ term if and only if $D^2 h_0(y) = 0$ for all y. The asymptotic expansion of the ILDM is given by* Eq. (5.10). *A comparison of the coefficients in the expansion with the coefficients in the expansion of the slow manifold $\mathcal{M}_\varepsilon$,* Eq. (2.12), *shows that*

$$\psi^{(0)} = h_0, \tag{5.48}$$

$$\psi^{(1)} = h^{(1)}, \tag{5.49}$$

$$\psi^{(2)} = h^{(2)} - (D_z g)^{-2}(D^2 h_0)(f, f). \tag{5.50}$$

The difference $\psi^{(2)} - h^{(2)}$ involves the bilinear form $D^2 h_0$, which is proportional to the curvature of the zero set of g at $\varepsilon = 0$. It is present in any fast–slow system, unless the curvature vanishes everywhere. Because of it, the ILDM is in general not invariant under the dynamics of the system of equations (2.1) and (2.2).

6. The iterative method of Fraser and Roussel

The iterative method of Fraser and Roussel was developed originally for planar fast–slow systems that are linear in the fast variable,

$$\dot{y} = f_1(y, \varepsilon)z + f_2(y, \varepsilon), \tag{6.1}$$

$$\varepsilon\dot{z} = g_1(y, \varepsilon)z + g_2(y, \varepsilon). \tag{6.2}$$

Here, y and z are scalar-valued functions of time. These systems of equations are typical for enzyme kinetics [13] and other biochemical systems whose dynamics can be reduced to slow manifolds. In this case, the slow manifolds are curves in the phase plane.

On any trajectory $z = z(y, \varepsilon)$ in the phase plane, we have the identity $\dot{z} = z_y\dot{y}$, or, in terms of the functions f and g,

$$\varepsilon z_y(f_1 z + f_2) = g_1 z + g_2. \tag{6.3}$$

This identity is known as the *invariance equation*. In the present case, it can be solved for z in terms of y and z_y,

$$z = \frac{-g_2 + \varepsilon f_2 z_y}{g_1 - \varepsilon f_1 z_y}. \tag{6.4}$$

The equation holds, in particular, along trajectories on invariant manifolds. Fraser used Eq. (6.4) to propose the following functional iteration procedure to approximate the slow manifold.

Starting from an initial function $\varphi^{(0)}$, one computes a sequence of functions $\{\varphi^{\ell}\colon\ \ell = 1, 2, \dots\}$ using the definitions

$$\varphi^{(\ell)} = \frac{-g_2 + \varepsilon f_2 \varphi_y^{(\ell-1)}}{g_1 - \varepsilon f_1 \varphi_y^{(\ell-1)}}, \quad \ell = 1, 2, \dots. \tag{6.5}$$

Under appropriate conditions, the sequence $\{\varphi^{(\ell)}(y,\varepsilon)\colon\ \ell = 1, 2, \dots\}$ approaches $z(y,\varepsilon)$ (in a sense to be made precise) as ℓ goes to infinity, so the algorithm generates successive approximations to a slow manifold.

The iterative procedure generalizes to the fast–slow system of equations (2.1) and (2.2). The invariance equation is

$$\varepsilon(D_y z)(y,\varepsilon) f(y, z(y,\varepsilon), \varepsilon) = g(y, z(y,\varepsilon), \varepsilon) \tag{6.6}$$

for any trajectory $z = z(y,\varepsilon)$ in phase space. Starting from a function $\varphi^{(0)}$, one computes a sequence of functions $\{\varphi^{(\ell)}\colon\ \ell = 1, 2, \dots\}$ by solving the equation

$$\varepsilon(D_y \varphi^{(\ell-1)})(y,\varepsilon) f(y, \varphi^{(\ell)}(y,\varepsilon), \varepsilon) = g(y, \varphi^{(\ell)}(y,\varepsilon), \varepsilon). \tag{6.7}$$

The sequence $\{\varphi^{(\ell)}(y,\varepsilon)\colon\ \ell = 1, 2, \dots\}$ approaches $z(y,\varepsilon)$ (again, in a sense to be made precise) as ℓ goes to infinity.

Notice that Eq. (6.7) amounts to an *implicit definition* of $\varphi^{(\ell)}$, unless both f and g are linear in the fast variable z, as in the planar case discussed above, Eqs. (6.1) and (6.2). Hence, the numerical computation of $\varphi^{(\ell)}$ generally requires the solution of a nonlinear equation.

7. Asymptotics of the iterative method

Because the iterative method of Fraser and Roussel is closely related to the invariance equation, its asymptotic properties are most easily analyzed in terms of those of Eq. (6.6).

Lemma 7.1. *The invariance equation,* Eq. (6.6), *admits an asymptotic solution in the form of a power series expansion,*

$$z(y,\varepsilon) = z^{(0)}(y) + \varepsilon z^{(1)}(y) + \cdots \quad \text{as } \varepsilon \downarrow 0, \tag{7.1}$$

where

$$z^{(0)} = h_0, \tag{7.2}$$

and the functions $z^{(\ell)} : K \to \mathbf{R}^n$, $\ell = 1, 2, \dots$, are found successively from Eq. (7.17). *In particular, $z^{(1)}$ and $z^{(2)}$ are found from the equations*

$$(D_z g) z^{(1)} = (Dz^{(0)}) f - g_\varepsilon, \tag{7.3}$$

$$(D_z g) z^{(2)} = (Dz^{(1)}) f + (Dz^{(0)})((D_z f) z^{(1)} + f_\varepsilon) - \tfrac{1}{2}(D_z^2 g)(z^{(1)}, z^{(1)}) - (D_z g_\varepsilon) z^{(1)} - \tfrac{1}{2} g_{\varepsilon\varepsilon}, \tag{7.4}$$

where f and g and their derivatives are evaluated at $(y, z^{(0)}(y), 0)$.

Proof. We begin by expanding the function f,

$$f(y, z(y,\varepsilon), \varepsilon) = f^{(0)}(y) + \varepsilon f^{(1)}(y) + \varepsilon^2 f^{(2)}(y) + \cdots, \tag{7.5}$$

where

$$f^{(0)}(y) = f(y, z^{(0)}(y), 0), \tag{7.6}$$

and

$$f^{(\ell)}(y) = \sum_{j=1}^{\ell} \frac{1}{j!}(D_z^j) f(y, z^{(0)}(y), 0) \sum_{|i|=\ell} (z^{(i_1)}(y), \ldots, z^{(i_j)}(y)) + \sum_{k=1}^{\ell-1} \frac{1}{k!} \sum_{j=1}^{\ell-k} \frac{1}{j!} (D_z^j(\partial_\varepsilon^k f))(y, z^{(0)}(y), 0)$$
$$\times \sum_{|i|=\ell-k} (z^{(i_1)}(y), \ldots, z^{(i_j)}(y)) + \frac{1}{\ell!}(\partial_\varepsilon^\ell f)(y, z^{(0)}(y), 0), \quad \ell = 1, 2, \ldots. \tag{7.7}$$

The derivative $(D_z^j f)(y, z^{(0)}(y), 0)$ in the first term is a j-linear map from $(\mathbf{R}^n)^j$ to $\mathbf{R}^m$. The inner sums are taken over all multiindices $i = (i_1, \ldots, i_j)$ of positive integers $i_1, \ldots, i_j$ with length $|i| = \sum_{k=1}^{j} i_k = \ell$ and $\ell - k$, respectively. The first few coefficients are

$$f^{(1)}(y) = (D_z f) z^{(1)} + f_\varepsilon, \tag{7.8}$$

$$f^{(2)}(y) = (D_z f) z^{(2)} + \tfrac{1}{2}(D_z^2 f)(z^{(1)}, z^{(1)}) + (D_z f_\varepsilon) z^{(1)} + \tfrac{1}{2} f_{\varepsilon\varepsilon}, \tag{7.9}$$

$$f^{(3)}(y) = (D_z f) z^{(3)} + (D_z^2 f)(z^{(1)}, z^{(2)}) + \tfrac{1}{6}(D_z^3 f)(z^{(1)}, z^{(1)}, z^{(1)}) + (D_z f_\varepsilon) z^{(2)} + \tfrac{1}{2}(D_z^2 f_\varepsilon)(z^{(1)}, z^{(1)})$$
$$+ \tfrac{1}{2}(D_z f_{\varepsilon\varepsilon}) z^{(1)} + \tfrac{1}{6} f_{\varepsilon\varepsilon\varepsilon}, \tag{7.10}$$

where f and its derivatives are evaluated at $(y, z^{(0)}(y), 0)$, and the argument of each $z^{(i)}, i = 1, 2, \ldots$, is y. A similar expansion holds for $g(y, z(y, \varepsilon), \varepsilon)$,

$$g(y, z(y, \varepsilon), \varepsilon) = g^{(0)}(y) + \varepsilon g^{(1)}(y) + \varepsilon^2 g^{(2)}(y) + \cdots, \tag{7.11}$$

where

$$g^{(0)}(y) = g(y, z^{(0)}(y), 0), \tag{7.12}$$

and

$$g^{(\ell)}(y) = \sum_{j=1}^{\ell} \frac{1}{j!}(D_z^j g)(y, z^{(0)}(y), 0) \sum_{|i|=\ell} (z^{(i_1)}(y), \ldots, z^{(i_j)}(y)) + \sum_{k=1}^{\ell-1} \frac{1}{k!} \sum_{j=1}^{\ell-k} \frac{1}{j!} (D_z^j(\partial_\varepsilon^k g))(y, z^{(0)}(y), 0)$$
$$\times \sum_{|i|=\ell-k} (z^{(i_1)}(y), \ldots, z^{(i_j)}(y)) + \frac{1}{\ell!}(\partial_\varepsilon^\ell g)(y, z^{(0)}(y), 0), \quad \ell = 1, 2, \ldots. \tag{7.13}$$

Termwise differentiation of the asymptotic expansion (7.1) gives

$$(D_y z)(y, \varepsilon) = Dz^{(0)} + \varepsilon Dz^{(1)} + \cdots. \tag{7.14}$$

Equating the coefficients of like powers of ε in the left and right members of the invariance equation, Eq. (6.6), we obtain a sequence of functional identities,

$$g^{(0)} = 0, \tag{7.15}$$

$$g^{(\ell)} = \sum_{i=0}^{\ell-1} (Dz^{(i)}) f^{(\ell-1-i)}, \quad \ell = 1, 2, \ldots. \tag{7.16}$$

We satisfy the $\mathcal{O}(1)$ equation, Eq. (7.15), by taking $z^{(0)} = h_0$; see Eq. (7.2). Then $f^{(0)}(y) = f(y, h_0(y), 0)$, and $f^{(\ell)}(y)$ and $g^{(\ell)}(y)$ are given by Eqs. (7.7) and (7.13), respectively, with $z^{(0)}(y)$ replaced by $h_0(y)$.

Next, we turn to the $\mathcal{O}(\varepsilon^\ell)$ equation, Eq. (7.16). We observe that $z^{(\ell)}$ occurs in $g^{(\ell)}$ only in the first term with $j = 1$; the remaining terms involve $z^{(1)}$ through $z^{(\ell-1)}$ but not $z^{(\ell)}$. The right member of Eq. (7.16) similarly involves $z^{(1)}$ through $z^{(\ell-1)}$ but not $z^{(\ell)}$. Therefore, the identities (7.16) can be solved successively for $z^{(1)}$, $z^{(2)}$, and so on. Thus we find

$$\begin{aligned}(D_z g)z^{(\ell)} = &\sum_{i=0}^{\ell-1}(Dz^{(i)})f^{(\ell-1-i)} - \sum_{j=2}^{\ell}\frac{1}{j!}(D_z^j g)\sum_{|i|=\ell}(z^{(i_1)},\ldots,z^{(i_j)})\\ &- \sum_{k=1}^{\ell-1}\frac{1}{k!}\sum_{j=1}^{\ell-k}\frac{1}{j!}(D_z^j(\partial_\varepsilon^k g))\sum_{|i|=\ell-k}(z^{(i_1)},\ldots,z^{(i_j)}) - \frac{1}{\ell!}(\partial_\varepsilon^\ell g)\end{aligned} \tag{7.17}$$

for $\ell = 1, 2, \ldots$. Here, the functions f and g and their derivatives are evaluated at $(y, z = h_0(y), 0)$, and it is understood that a sum is empty when the lower bound exceeds the upper bound. The equations for $\ell = 1$ and 2 are given in the statement of the theorem. □

The following theorem shows that ℓ successive applications of the iterative algorithm of Fraser and Roussel, starting from $\varphi^{(0)} = h_0$, generate an approximation $\varphi^{(\ell)}$ to the slow manifold $\mathcal{M}_\varepsilon$ that is accurate up to and including the $\mathcal{O}(\varepsilon^\ell)$ term as $\varepsilon \downarrow 0$.

Theorem 7.1. *Let $\varphi^{(\ell)}$ and $z^{(\ell)}$ be defined recursively for $\ell = 1, 2, \ldots$ by Eqs.* (6.7) *and* (7.17)*, respectively. If $\varphi^{(0)} = z^{(0)} = h_0$, then*

$$\varphi^{(\ell)} \equiv \varphi^{(\ell)}(y, \varepsilon) = \sum_{i=0}^{\ell}\varepsilon^i z^{(i)}(y) + \mathcal{O}(\varepsilon^{\ell+1}), \quad \ell = 1, 2, \ldots. \tag{7.18}$$

Proof. The proof is by induction. Taking $\ell = 1$, we have

$$\varepsilon(D\varphi^{(0)})(y)f(y, \varphi^{(1)}(y, \varepsilon), \varepsilon) = g(y, \varphi^{(1)}(y, \varepsilon), \varepsilon).$$

Since $\varphi^{(0)} = z^{(0)}$, this equation is the same as

$$\varepsilon(Dz^{(0)})(y)f(y, \varphi^{(1)}(y, \varepsilon), \varepsilon) = g(y, \varphi^{(1)}(y, \varepsilon), \varepsilon).$$

We expand the terms in this equation in powers of ε and equate the coefficients of like powers of ε. To leading order, we find the equation

$$g(y, \varphi^{(1)}(y, 0), 0) = 0,$$

which is precisely the equation for $z^{(0)}$, so

$$\varphi^{(1)}(y, 0) = z^{(0)}(y).$$

To the next order, we find the equation

$$(D_z g)(y, z^{(0)}, 0)\varphi_\varepsilon^{(1)}(y, 0) + g_\varepsilon(y, z^{(0)}, 0) = (Dz^{(0)})(y)f(y, z^{(0)}(y), 0),$$

which is precisely Eq. (7.3), so

$$\varphi_\varepsilon^{(1)}(y, 0) = z^{(1)}(y).$$

Thus,

$$\varphi^{(1)}(y,\varepsilon) = z^{(0)}(y) + \varepsilon z^{(1)}(y) + \mathcal{O}(\varepsilon^2),$$

and the theorem is true for $\ell = 1$.

Suppose the theorem is true for $\ell - 1$, $\varphi^{(\ell-1)} = \sum_{i=0}^{\ell-1} \varepsilon^i z^{(i)} + \mathcal{O}(\varepsilon^\ell)$. The function $\varphi^{(\ell)}$ is defined by Eq. (6.7),

$$\varepsilon(D_y\varphi^{(\ell-1)})(y,\varepsilon) f(y,\varphi^{(\ell)}(y,\varepsilon),\varepsilon) = g(y,\varphi^{(\ell)}(y,\varepsilon),\varepsilon).$$

We expand each term in powers of ε and equate the coefficients of like powers,

$$\sum_{i=0}^{j-1} (D_y z^{(i)}) f^{(j-1-i)} = g^{(j)}, \quad j = 1, 2, \ldots, \ell,$$

where $f^{(\cdot)}$ and $g^{(\cdot)}$ are the functions defined after Eq. (7.5). For $j = 1, \ldots, \ell$, these are exactly the same functional identities as we derived above. Hence, order by order, the solution is

$$\partial_\varepsilon^i \varphi^{(\ell)}(y,0) = z^{(i)}(y), \quad i = 0, 1, \ldots, \ell.$$

This proves that the theorem is true for ℓ. □

Remark. In general, the coefficient of ε^2 in $\varphi^{(1)}$ will not be equal to $z^{(2)}$. This may be seen by direct examination of the $\mathcal{O}(\varepsilon^2)$ equation,

$$(Dz^{(0)})(y)((D_z f)z^{(1)} + f_\varepsilon) = \tfrac{1}{2}(D_z^2 g)(z^{(1)}, z^{(1)}) + (D_z g_\varepsilon)z^{(1)} + \tfrac{1}{2} g_{\varepsilon\varepsilon},$$

which differs from Eq. (7.4). The latter has two additional terms, namely, $(D_z g)z^{(2)}$ and $(Dz^{(1)})f$. Hence, the first iterate generally involves an error of $\mathcal{O}(\varepsilon^2)$. Similarly, the ℓth iterate does not give the same equation for the term at order $\mathcal{O}(\varepsilon^{\ell+1})$ as compared with that obtained from invariant manifold theory; hence, the error in the approximation at this stage is generally $\mathcal{O}(\varepsilon^{\ell+1})$.

A comparison of the results given in Theorem 7.1 with Theorem 2.1 leads to the following conclusions.

Corollary 7.1. *The iterative method of Fraser and Roussel gives successively higher-order asymptotic approximations to the slow manifold $\mathcal{M}_\varepsilon$. Starting from $\varphi^{(0)} = h_0$, ℓ applications of the iterative procedure give an approximation $\varphi^{(\ell)}$ that satisfies*

$$\varphi^{(\ell)} = \sum_{i=0}^{\ell} \varepsilon^i h^{(i)} + \mathcal{O}(\varepsilon^{\ell+1}), \quad \ell = 1, 2, \ldots, \tag{7.19}$$

where the functions $h^{(i)}$ are the coefficients in the asymptotic expansion of $\mathcal{M}_\varepsilon$ given in Theorem 2.1, Eq. (2.12).

Remark. The result of Corollary 7.1 was shown for a special class of planar fast-slow vector fields in Ref. [46]. Also, in Ref. [62], Roussel and Fraser noted the connection between their iterative approach and techniques from dynamical systems theory, such as deriving the existence of certain invariant manifolds via application of the contraction mapping principle to appropriate integral equations.

8. The MMH model

In this section, we illustrate the analytical results of the preceding sections on the MMH model. The MMH model is a prototype reaction mechanism for enzyme kinetics in biochemistry [21]. It is a planar fast–slow system, which is given in nondimensional form by the equations

$$\dot{y} = -y + (y + a - b)z, \tag{8.1}$$

$$\varepsilon\dot{z} = y - (y + a)z. \tag{8.2}$$

The variables are y, the concentration of the substrate, and z, the concentration of an intermediate substrate–enzyme complex. The parameters a and b satisfy the inequalities $a > b > 0$.

Remark. The MMH model, Eqs. (8.1) and (8.2), is derived from a more complicated system involving four species (enzyme, substrate, enzyme–substrate complex, and product) and two reactions (one reversible, one irreversible). The full system can be reduced to the planar system, because it has two conserved quantities; see Ref. [21].

The system of equations (8.1) and (8.2) has a family of slow manifolds $\mathcal{M}_\varepsilon$, whose asymptotics are given by Eq. (2.12),

$$h_\varepsilon(y) = h_0(y) + \varepsilon h^{(1)}(y) + \varepsilon^2 h^{(2)}(y) + \cdots, \quad y > 0, \tag{8.3}$$

where

$$h_0(y) = \frac{y}{y + a}, \tag{8.4}$$

$$h^{(1)}(y) = \frac{aby}{(y + a)^4}, \tag{8.5}$$

$$h^{(2)}(y) = \frac{aby(2ab - 3by - ay - a^2)}{(y + a)^7}. \tag{8.6}$$

We now show that the ILDM method yields an approximation of the slow manifold that is accurate up to and including the $\mathcal{O}(\varepsilon)$ term, but differs at $\mathcal{O}(\varepsilon^2)$ because of the curvature of h_0.

The Jacobian of the vector field associated with Eqs. (8.1) and (8.2) is

$$J = \begin{pmatrix} -(1 - z) & y + a - b \\ \varepsilon^{-1}(1 - z) & -\varepsilon^{-1}(y + a) \end{pmatrix}, \tag{8.7}$$

and its eigenvalues are

$$\lambda_{s,f}(y, z) = -\frac{1}{2}\left(\frac{y + a}{\varepsilon} + 1 - z\right) \pm \sqrt{\frac{1}{4}\left(\frac{y + a}{\varepsilon} + 1 - z\right)^2 - \frac{b(1 - z)}{\varepsilon}}. \tag{8.8}$$

The (nonnormalized) slow eigenvector is

$$v_s = \begin{pmatrix} \lambda_s + \varepsilon^{-1}(y + a) \\ \varepsilon^{-1}(1 - z) \end{pmatrix}, \tag{8.9}$$

and there is a corresponding fast eigenvector. The vector v_s spans the slow subspace. As noted before, the fast and slow subspace are not orthogonal, so we work with $v_s^\perp$ and define the ILDM as the set of points where the vector field is orthogonal to $v_s^\perp$,

$$(1-z)[-y+(y+a-b)z]-(\lambda_s+\varepsilon^{-1}(y+a))[y-(y+a)z]=0. \tag{8.10}$$

Asymptotically, the ILDM is given by

$$z=\psi(y,\varepsilon)=\psi^{(0)}(y)+\varepsilon\psi^{(1)}(y)+\varepsilon^2\psi^{(2)}(y)+\cdots, \tag{8.11}$$

where

$$\psi^{(0)}(y)=\frac{y}{y+a}, \tag{8.12}$$

$$\psi^{(1)}(y)=\frac{aby}{(y+a)^4}, \tag{8.13}$$

$$\psi^{(2)}(y)=\frac{aby(2ab-by-ay-a^2)}{(y+a)^7}. \tag{8.14}$$

A comparison of the coefficients in the expansions (8.3) and (8.11) shows agreement of the $\mathcal{O}(1)$ and $\mathcal{O}(\varepsilon)$ terms. On the other hand, the $\mathcal{O}(\varepsilon^2)$ terms differ; their difference is proportional to the curvature of h_0,

$$\psi^{(2)}-h^{(2)}=\frac{2ab^2y^2}{(y+a)^7}=-\frac{f^2}{g_z^2}h_0''. \tag{8.15}$$

Remark. For planar systems, Ref. [51] introduces a curve, called the A-inflector, which is equivalent to the ILDM. It is found as the set of points at which the vector field is parallel to the slow eigenvector, and it is interpreted as the set of points at which the trajectories near the slow manifold have inflection points in the (y,z) plane. This A-inflector is given for the MMH model in Ref. [52, Eq. (5.2)]; the explicit formula involves y as a function of z.

Remark. For the planar MMH problem, Roussel [59] also finds a curve, labeled the slow tangent manifold, that is equivalent to the ILDM. The formula for this curve [59, Eq. (3.2.19)], is derived by solving the ILDM equation, which is a quadratic in y and a cubic in z, explicitly for y as a function of z, as in Ref. [52].

The iterative method of Fraser and Roussel starts from the invariance equation,

$$z=\frac{y+\varepsilon z_y y}{y+a+\varepsilon z_y(y+a-b)} \tag{8.16}$$

or, equivalently,

$$z=\frac{y}{y+a}+\varepsilon\frac{byz_y}{(y+a)^2}-\varepsilon^2\frac{by(y+a-b)z_y^2}{(y+a)^3}+\varepsilon^3\frac{by(y+a-b)^2z_y^3}{(y+a)^4+\varepsilon(y+a)^3(y+a-b)z_y}. \tag{8.17}$$

Successive applications of the iterative algorithm lead to the approximations

$$\varphi^{(0)}=\frac{y}{y+a}, \tag{8.18}$$

$$\varphi^{(1)}=\frac{y}{y+a}+\varepsilon\frac{aby}{(y+a)^4}-\varepsilon^2\frac{a^2by(y+a-b)}{(y+a)^7}+\mathcal{O}(\varepsilon^3), \tag{8.19}$$

$$\varphi^{(2)} = \frac{y}{y+a} + \varepsilon \frac{aby}{(y+a)^4} + \varepsilon^2 \frac{aby(2ab - 3by - ay - a^2)}{(y+a)^7} + \mathcal{O}(\varepsilon^3). \tag{8.20}$$

A comparison with Theorem 2.1 (and Eqs. (8.4)–(8.6)) shows that $\varphi^{(\ell)}$ is asymptotically correct up to and including terms of $\mathcal{O}(\varepsilon^\ell)$ for $\ell = 1, 2$ (and beyond), as predicted by the analysis.

Remark. We refer the reader to Refs. [54,55,71] for further analyses of the MMH model in the context of singular perturbation theory. The last two references significantly extend the domain in parameter space where the model can be analyzed by using separation of time scales.

9. The Davis–Skodje model

The planar fast–slow system

$$\dot{y} = -y, \tag{9.1}$$

$$\varepsilon \dot{z} = -z + \frac{y}{1+y} - \frac{\varepsilon y}{(1+y)^2}, \tag{9.2}$$

was introduced by Davis and Skodje [7] as a model on which to compare various reduction methods. (The inverse, ε^{-1}, which is large, equals the large parameter γ of Ref. [7].)

For any ε, the curve

$$z = h_\varepsilon(y) = \frac{y}{1+y}, \quad y \geq 0, \tag{9.3}$$

is invariant under the dynamics of Eqs. (9.1) and (9.2). Therefore, the function h_ε represents the slow manifold exactly on $y \geq 0$ for all small $\varepsilon > 0$. (The nonlinearity in Eqs. (9.1) and (9.2) was, in fact, chosen so the slow manifold is given by the simple expression of Eq. (9.3).)

The Jacobian of the vector field is

$$J = \begin{pmatrix} -1 & 0 \\ \varepsilon^{-1}((1+y) + \varepsilon(y-1))(1+y)^{-3} & -\varepsilon^{-1} \end{pmatrix}, \tag{9.4}$$

and the eigenvalues are $\lambda_s = -1$ and $\lambda_f = -1/\varepsilon$ for all (y, z). The corresponding (nonnormalized) eigenvectors are

$$v_s = \begin{pmatrix} 1 \\ (1-\varepsilon)^{-1}(1+y+\varepsilon(y-1))(1+y)^{-3} \end{pmatrix}, \qquad v_f = \begin{pmatrix} 0 \\ 1 \end{pmatrix}. \tag{9.5}$$

The ILDM is given by the expression

$$z = \psi(y, \varepsilon) = \frac{y}{1+y} + \frac{2\varepsilon^2 y^2}{(1-\varepsilon)(1+y)^3}, \tag{9.6}$$

cf. [7, Eq. (3.8)]. Its asymptotic expansion is

$$z = \frac{y}{1+y} + \varepsilon^2 \frac{2y^2}{(1+y)^3} + \cdots. \tag{9.7}$$

The error in the expansion is $\mathcal{O}(\varepsilon^2)$ and proportional to the curvature, $h_0'' = -2/(1+y)^3$.

The invariance equation is

$$z = \frac{y}{1+y} - \varepsilon \frac{y}{(1+y)^2} + \varepsilon y z_y, \tag{9.8}$$

from which one readily verifies that the iterative method of Fraser and Roussel yields the approximation $\varphi^{(\ell)}(y) = y/(1+y)$ for $\ell = 1, 2, \ldots$.

Remark. If one restricts the variable y to a finite interval, then the system of equations (9.1) and (9.2) has a family of slow manifolds, all exponentially close ($\mathcal{O}(\mathrm{e}^{-c/\varepsilon})$ for some $c > 0$) to the exact slow manifold, Eq. (9.3).

10. Discussion

The fast–slow system of equations (2.1) and (2.2) captures the essential elements of any reaction mechanism whose long-time dynamics evolve on a slow manifold in the composition space. As stated in Section 2, however, this system is a mathematical idealization, and we need to consider how the results of the analysis carry over to more general reaction mechanisms. In this section, we consider issues related to the separation of time scales (Section 10.1), the inclusion of conserved quantities (Section 10.2), and the development and analysis of reduction mechanisms for reaction–diffusion equations (Section 10.3).

10.1. Separation of time scales

In this section, we discuss the partition of variables into a fast and a slow group and the assumption that the groups evolve on time scales that are and remain well separated at all times. This assumption underlies the definition of the small parameter ε in the model of Eqs. (2.1) and (2.2). We also discuss the possibility of partitioning the variables in more than two groups.

While many of the systems to which reduction methods have been applied satisfy this assumption, there are a significant number of reaction mechanisms where the fast and slow time scales are separated, but not well separated. This is the case, for example, when ε is no longer an asymptotically small parameter but a fixed (relatively small) number. In such cases, the spectral gap between the fast and slow eigenvalues of the Jacobian of the vector field is small, much smaller than the chasm between the fast $\mathcal{O}(\varepsilon^{-1})$ eigenvalues and the slow $\mathcal{O}(1)$ eigenvalues for asymptotically small ε, even at points near a low-dimensional manifold. Nevertheless, as long as there exists a nonzero spectral gap, the Jacobian can still be reduced to a fast and a slow component, and the ILDM method can be (and has been) implemented numerically. Part of our ongoing research is aimed at using spectral projection operators to analyze these applications of the ILDM method.

In addition, there are reaction mechanisms where the number of fast and slow species changes over time, as may happen, for example, when the temperature in the chemical reactor changes and the least-slow of the slow species transits to the group of fast species. To account for this type of occurrence, practical implementations of the ILDM method may use one ILDM until the crossover occurs and another after the crossover. The two procedures can be linked by doing a numerical integration of the full system of equations during the crossover. The analysis presented in this paper applies to each ILDM separately. However, it should be noted that the relationship between these two ILDMs (and, more generally, slow manifolds) depends on the bifurcation that occurs at crossover. As an alternative, one may consider repartitioning the species to avoid crossover altogether.

In some systems, a component of y may evolve on an even slower time scale than that given by $\tau = \varepsilon t$. For example, in Eq. (2.1) one may have $y_i' = \varepsilon^{1+\gamma} f_i(y, z, \varepsilon)$ with $\gamma > 0$ for some index i. Such cases are accounted

for by the present analysis; in fact, it suffices to absorb the factor ε^{γ} in f_i. On the other hand, the fact that there are slower time scales in the model may point to the existence of still lower-dimensional slow manifolds. By systematically eliminating fast variables, starting with the fastest and proceeding up the hierarchy, one can reduce the dimensionality of the slow manifold in a systematic way until no further reduction is possible. Such an approach has been used, for example, in Refs. [62,89]. The idea of a hierarchy of time scales was first explored in singular perturbation theory by Tikhonov [79].

10.2. Conserved quantities and the ILDM method

In this section, we briefly consider the ILDM method for fast–slow systems whose dynamics are described by Eqs. (2.1) and (2.2), where the unknowns satisfy one or more conservation laws. For models of chemical reactions, a conserved quantity is, typically, a linear combination of several unknowns that is constant in time. Conserved quantities give rise to zero eigenvalues of the Jacobian of the vector field, and the ILDM method groups these zero eigenvalues with the slow ones.

Degeneracies in the Jacobian affect the analysis of the ILDM method. Consider, for example, a system given by Eqs. (2.1) and (2.2) with $m = 2$ and $n = 2$, which has one conserved quantity. If the conserved quantity is a linear combination of the two slow variables, then the first and second row of the Jacobian are linearly dependent. In this case, there is effectively only one slow variable, and the analysis presented in this paper applies to the one-dimensional slow manifold $\mathcal{M}_0$. If, by contrast, the conserved quantity is a linear combination of the two fast variables, then not only is J degenerate, but also the columns of $D_z g(y, h_0(y), 0)$ are linearly dependent; hence, $\mathcal{M}_0$ is not asymptotically stable in the four-dimensional composition space. A reduction of the number of fast variables by one lifts this degeneracy, because $\mathcal{M}_0$ is asymptotically stable in the reduced three-dimensional composition space. Other possibilities are that a conserved quantity depends on a mix of fast and slow variables or that there are multiple conserved quantities. In each case, the analysis of the ILDM method presented here applies once the system has been reduced to a system for which the manifold $\mathcal{M}_0$ is asymptotically stable.

10.3. Reaction–diffusion equations

Reduction methods have also been developed for systems of *reaction–diffusion equations* [20,26,29,32,36,73,83, 89,90]. The elimination of the fast species affects not only the reaction kinetics of the slow species (as is the case for the kinetics reduction methods considered in this paper) but also their diffusivities. For systems where the fast and slow species have been separated, the "effective" diffusivities differ from the ordinary diffusivities by concentration-dependent terms that are higher order in ε. See [29, Section 7.6] for an example where the diffusivities are changed to leading order.

We illustrate this phenomenon on the MMH model with diffusion of the slow species,

$$\dot{y} = -y + (y + a - b)z + D\Delta y, \tag{10.1}$$

$$\varepsilon\dot{z} = y - (y + a)z. \tag{10.2}$$

The variables y and z depend not only on time but also on space; Δ is the Laplace operator. Ideas from inertial-manifold theory have been applied to this reaction–diffusion system [89]. The slow manifold is infinite dimensional, its asymptotics are given by

$$z = \frac{y}{y + a} + \varepsilon\left[\frac{aby}{(y + a)^4} - \frac{a}{(y + a)^3}D\Delta y\right] + \mathcal{O}(\varepsilon^2), \tag{10.3}$$

and the reduced reaction–diffusion equation up to and including terms of $\mathcal{O}(\varepsilon)$ is

$$\dot{y} = -y + (y + a - b)\left(\frac{y}{y+a} + \varepsilon\frac{aby}{(y+a)^4}\right) + D\left(1 - \varepsilon\frac{a(y+a-b)}{(y+a)^3}\right)\Delta y. \tag{10.4}$$

The regular diffusivity D is seen to be corrected by an ε-dependent term that involves the concentration of the slow species.

Singular perturbation theory provides an alternative method to find the infinite-dimensional slow manifold and, hence, the reduced reaction–diffusion equation for this and similar systems. In particular, one finds an asymptotic expansion for the slow manifold of the form

$$z = h_0(y) + \varepsilon h^{(1)}(y, \Delta y) + \varepsilon^2 h^{(2)}(y, \Delta y, \Delta^2 y) + \cdots. \tag{10.5}$$

For the MMH model, this procedure leads to the same expansion of the slow manifold, Eq. (10.3), and dynamical systems theory states that $\mathcal{M}_\varepsilon$ is the infinite-dimensional weak stable manifold of the spatially homogeneous state $(0, 0)$.

We remark that one can include diffusion of the fast variable z, but only if the diffusion coefficient is $\mathcal{O}(\varepsilon)$,

$$\dot{y} = f(y, z, \varepsilon) + D_1\Delta y, \tag{10.6}$$

$$\varepsilon\dot{z} = g(y, z, \varepsilon) + \varepsilon D_2\Delta z. \tag{10.7}$$

In this case, $z^{(0)}$ is independent of D_2, and one can follow the same asymptotic procedure as before; the diffusion coefficient D_2 enters into the equation of order $\mathcal{O}(\varepsilon)$. However, if the diffusion term in the fast equation is $\mathcal{O}(1)$, the asymptotic procedure for finding $z^{(0)}$ breaks down.

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